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**Quantum phase transitions with polar molecules:
Towards the realization of ferro–electric liquids**

Quantenphasenübergänge mit polaren Molekülen:
Realisierung von ferroelektrischen Flüssigkeiten

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Statutory Declaration:

I declare that I have written this thesis independently and have not used any other sources or aids than those referenced in this work.

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Abstract

Recently cold polar molecules trapped in optical lattices have been used to engineer strongly correlated quantum phases. The high tunability of the dipolar interaction and the internal rotational structure of polar molecules is suitable to realize field induced spin systems that can be efficiently tuned to simulate important model systems of condensed matter physics. In this diploma thesis we demonstrate the realization of spin- $1/2$ Heisenberg models and propose a method for the simulation of the t - J model with strong exchange interactions. In the first part, the spin- $1/2$ XXZ model in an optical two-dimensional square lattice is derived microscopically using external static electric and microwave fields. The influence of the slowly decaying dipolar $1/r^3$ interaction between polar molecules on the ground state phase diagram at $T = 0$ is studied by employing a spin wave analysis. In the second part of this work, an attempt to simulate the t - J model with polar molecules is undertaken and there might be several perspectives of an exact realization. As an outlook, a short discussion of using the results of this work to realize ferro-electric liquids is given.

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Contents

List of Figures	VII
List of Tables	IX
List of Program Codes	XI
List of Symbols	XIII
1 Introduction	1
2 Basic Concepts of Polar Molecules	3
2.1 Quantum Rigid Rotor	3
2.1.1 Linear Rigid Rotor of a Diatomic Molecule	3
2.1.2 Selection Rules for Rotational Transitions	4
2.2 Two Level Systems	5
2.3 Polar Molecules in External Electric Fields	13
2.3.1 Dipole Approximation	13
2.3.2 Polar Molecules in Static Electric Fields	16
2.3.3 Polar Molecules in Microwave Fields	20
2.3.4 Rotating Wave Approximation	21
2.4 Interactions between Polar Molecules	26
2.4.1 Dipole–Dipole Interaction	26
2.4.2 Van–der–Waals Interaction	29
3 Realization of Heisenberg Models	31
3.1 Heisenberg Models of Quantum Magnets	31
3.2 Realizing the Heisenberg Model with Polar Molecules	32
3.3 Spin Waves Excitations	39
3.3.1 Fourier Transform of the $1/R^3$ –Interaction	39
3.3.2 Ferromagnetic Spin Wave Excitations	46
3.3.3 Antiferromagnetic Spin Waves	53
3.3.4 In–plane magnetically ordered XXZ Model	65
3.3.5 In–Plane Ferromagnetic Spin Waves	66
3.3.6 In–Plane Antiferromagnetic Spin Waves	68
3.4 Mean Field Solution of the XXZ Model	71
3.5 Phase Diagram of the XXZ Model	76
4 t–J Model	81
4.1 t – J Model for strongly correlated electrons	81
4.2 Realizing the t – J Model with Polar Molecules	82

5 Summary & Conclusion	89
Appendix	91
A Spherical Tensor Operators	91
A.1 Clebsch–Gordan Coefficients	91
A.2 Wigner–Eckart Theorem	92
B Time Independent Perturbation Theory	95
C Coordinate Transformations	97
D Operators in Product Spaces	101
E General Solution of a Two–Level System	109
F Fourier Transformation	117
F.1 One–Dimensional Discrete Fourier Transformation	117
F.2 d –Dimensional Discrete Fourier Transformation	118
F.3 Continuous Fourier Transformation	118
F.4 Poisson Summation Formula	120
F.5 Physical Application of Fourier Transformations	121
F.5.1 $d = 3$ dimensional Fourier Series (discrete in \mathbf{k} –space)	121
F.5.2 $d = 3$ dimensional discrete Fourier transformation	121
F.5.3 $d = 3$ dimensional Fourier Series (discrete in real space)	124
F.5.4 Translational Invariant Systems	124
G Pauli Representation of the Lie Group $SU(2)$	127
G.1 General Properties of the Special Unitary Group $SU(n)$	127
G.2 Properties of $SU(2)$	128
G.3 Physical Application of $SU(2)$	129
H Documentation of Mathematica Programs	131
H.1 Numerical-Diagonalization.nb	131
H.2 Energy-Stark-Shift.nb	134
H.3 Fourier-Transform-Interaction.nb	136
H.4 Dipole-Moments-Coupling-Constants.nb	137
H.5 t-J-Model-Roots.nb	141
Bibliography	145
Acknowledgments	149

List of Figures

2.1	Rotational spectrum of rigid rotor	4
2.2	Diatomic Molecule	5
2.3	Two level system with ground and excited state	6
2.4	Oscillations between the ground state and the excited state of the two level system	10
2.5	Two level system with and without perturbation leading to an energy shift	12
2.6	Stark shifted states of polar molecules	18
2.7	Plots of Stark shifted perturbed energy levels	19
2.8	Numerically calculated Stark shift of the three lowest energy levels	21
2.9	Coupling of the dc-Stark shifted states by the external microwave field	26
2.10	Dipole-dipole interaction of two polar molecules	29
3.1	Setup for the realization of the spin- $1/2$ XXZ model	33
3.2	Plots of induced dipole moments and coupling constants	38
3.3	Possible realizations of antiferromagnetic spin models	39
3.4	First three shells in direct and reciprocal space	42
3.5	Fourier transform of the interaction potential	44
3.6	Dispersion relation of spin waves about in-axis ferro ground state	52
3.7	Ferro- and antiferromagnetically ordered ground states	53
3.8	Illustration of the two sublattices	56
3.9	Geometric construction of the g lattice factor – Brillouin zone of sublattices	62
3.10	Dispersion relation of spin waves about in-axis antiferro ground state	64
3.11	Dispersion relation of spin waves about in-plane ferro ground state	68
3.12	In-plane antiferromagnet dispersion relations	69
3.13	The two dispersion relation for the in-plane antiferromagnet	71
3.14	Neél ordered ground state divided in two sublattices	73
3.15	Non-analytical points in mean field ground state energy	77
3.16	Comparison of short-range interaction to long-range interaction phase diagram	78
4.1	Setup to realize the t - J model	82
4.2	Fixing the parameters for magnetic field and on-site repulsion	86
4.3	Fixing the parameter to obtain an isotropic coupling	87
E.1	Rotation of the basis vectors caused by the unitary transformation Q	114
F.1	Construction of the n^{th} Brillouin zone	122
F.2	Nearest neighbor shells in the direct lattice	123
H.1	Numerically determined eigenstates and eigenvalues	132
H.2	Coupling of Stark-shifted states	138

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List of Tables

2.1	Comparison between the two level system with and without perturbation	13
2.2	Physical explanation of the rotating wave approximation	24
2.3	Rotating Wave Hamiltonian compared the solution of the two level system	25
3.1	Numerical contributions to $\epsilon(\mathbf{0})$ for $\eta = 1$ and $\eta = \pi$	43
3.2	Comparison of numerical solution to approximated analytical solution	45
3.3	Overview over ground state energies	74
3.4	Overview over dispersion relations	80
A.1	List of the first three (unnormalized) spherical harmonics	92

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List of Program Codes

Numerical-Diagonalization-Running.nb	131
H.1 Numerical-Diagonalization.nb	133
H.2 Energy-Stark-Shift.nb	134
H.3 Fourier-Transform-Interaction.nb	136
H.4 Dipole-Moments-Coupling-Constants.nb	137
H.5 t-J-Model-Roots.nb	141

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List of Symbols

$\partial_x, \frac{\partial}{\partial x}$	partial derivative
∇	multi dimensional gradient or “nabla” operator
$ \rangle$	ket state vector
$\langle $	bra state vector
c^*	complex conjugate of a complex number
$ \cdot $	absolute value of a complex number or a vector
$\ \cdot\ $	Hilbert space induced norm of a state
A^t, \mathbf{v}^t	transpose of a matrix or vector
\mathcal{A}	operator in the Hilbert space of state vectors
\mathcal{A}^\dagger	hermitian conjugate of an operator
$[\mathcal{A}, \mathcal{B}]$	commutator of two operators
$\{\mathcal{A}, \mathcal{B}\}$	anticommutator of two operators
$\langle \alpha \mathcal{A} \beta \rangle$	matrix elements of operators
$\mathcal{A} \otimes \mathcal{B}$	tensor product
$A : B$	inner product of two matrices
$\langle \alpha \mathcal{T}_q^{(k)} \beta \rangle$	reduced matrix element
$\mathbb{1}$	identity matrix or identity operator
$ \uparrow\rangle, \downarrow\rangle$	two level system states (spin-1/2 states)
a	lattice spacing of two dimensional cubic lattice
\mathbf{A}	vector potential
β	expansion parameter of Stark shift perturbation
B	rotational constant of rigid rotor
\tilde{c}	coefficient in rotating wave approximation
$C_q^{(k)}(\theta, \phi)$	unnormalized spherical harmonics

d	permanent dipole moment of polar molecules
\mathbf{d}	electric dipole operator, electric dipole moment
d_x, d_y, d_z	components of the electric dipole operator in Cartesian coordinates
d_{-1}, d_0, d_1	spherical components of the electric dipole operator
$d^{\uparrow\downarrow} = \langle \uparrow \mathbf{d} \downarrow \rangle$	dipole transition elements
\mathcal{D}	reduced dipole–dipole interaction operator
δ_{ij}	Kronecker delta symbol
$\delta(\mathbf{r} - \mathbf{r}')$	delta distribution
Δ	detuning from resonant frequency
$ e\rangle$	excited state (of a two level system)
$\mathbf{e}_0, \mathbf{e}_{\pm 1}$	spherical basis vectors
\mathbf{e}_r	spherical unit vector
E_0	symmetrized ground state energy
E_{JM}	energy of perturbed Stark shift states
$E_{\uparrow}, E_{\downarrow}$	upper (lower) energy level of two level systems
E_+, E_-	eigenenergies of two level systems
\mathbf{E}_{ac}	external microwave field
\mathbf{E}_{dc}	static (external) electric field
$\epsilon(\mathbf{k})$	discrete Fourier transform of interaction range
\mathcal{F}, \tilde{f}	Fourier transform
Γ	generalized factorial function (Γ function)
$ g\rangle$	ground state (of a two level system)
g^F	ferromagnetic lattice factor
$g = g^{\oplus} - g^{\ominus}$	antiferromagnetic lattice factor
\mathbf{G}	reciprocal lattice vector
\hbar	reduced Planck's constant
h	magnetic field
$\mathcal{H}, \mathcal{H}, \mathcal{H}$	Hamilton operator
\mathcal{H}_{rot}	rigid rotor Hamiltonian
\mathcal{H}_{dc}	static electrical field Hamiltonian
\mathcal{H}_{ac}	microwave field Hamiltonian

i	imaginary unit
I	inertia tensor
I	moment of inertia
$\Im(c)$	imaginary part of a complex number
\mathcal{J}	angular momentum operator (divided by \hbar)
J	general magnetic coupling constant
J_z	“in axis” coupling constant
J_{\perp}	“in plane” coupling constant
$ JM\rangle$	eigenbasis of angular momentum operator
$ \phi_{JM}\rangle$	(perturbed) Stark shift states
k	rank of tensor operator
\mathbf{k}	wave vector (of plane waves)
\mathbf{K}	dimensionless wave vector
η	splitting factor between k -space summation and real space summation
μ	reduced mass of two body system
m	mass of particles
M	magnetic quantum number (of angular momentum eigenbasis)
N	total number of lattice points in the lattice
\bar{N}	number of lattice points in one direction
n_{\oplus}	index running over the same sublattice
n_{\ominus}	index running over different sublattices
φ	“mixing” angle of two level systems
ϕ	azimuth angle in spherical coordinates
Φ	electrical potential
$\mathcal{P}_{\uparrow}, \mathcal{P}_{\downarrow}$	spin 1/2 projectors
P_{α}	probability of finding a quantum system in state $ \alpha\rangle$
ρ	vector connecting nearest neighbor lattice points
$\rho(\mathbf{r})$	charge distribution
$\Re(c)$	real part of a complex number
\mathcal{R}	rotation matrix, rotating frame transformation
\mathbf{R}	separation vector, dimensionless lattice vector

q	electric quadrupole moment
q	polarization of microwave field
Q	transformation matrix, electric monopole
\mathbf{R}_i	position vector of i th molecule in lattice
$\sigma_x, \sigma_y, \sigma_z$	Pauli matrices
σ_+, σ_-	raising and lowering Pauli matrices
$\mathbf{S}, S^+, S^-, S^z$	spin $1/2$ -operators
ϑ	angle of coupling constants
θ	polar angle in spherical coordinates
$\mathcal{T}_q^{(k)}$	irreducible spherical tensor operator
\mathcal{T}	time ordering operator
\mathcal{U}	time evolution operator
\mathcal{V}^{dd}	dipole–dipole interaction operator
V_ρ	electrostatic energy
V_E	volume of primitive cell
$\chi(\mathbf{r}, t)$	gauge function
$Y_m^\ell(\theta, \phi)$	spherical harmonics
$ \psi(t)\rangle$	arbitrary state vector
ω	(resonant) transition frequency
ω_L	frequency of microwave field
$\omega_{\text{AF}}(\mathbf{k})$	antiferromagnet dispersion relation
$\omega_{\text{F}}(\mathbf{k})$	ferromagnet dispersion relation
Ω	generalized “Rabi” frequency
$\Omega_q^{\uparrow\downarrow}$	Rabi frequency coupling states $ \uparrow\rangle$ and $ \downarrow\rangle$

1

Introduction

Since the discovery of high temperature superconductors by Karl Müller and Johannes Bednorz [1] in 1986 intensive research has been undertaken to understand the physical mechanism causing the superconducting phase to appear. Unlike conventional superconductors the high temperature superconductors do not exhibit an isotope effect that explains the coupling mechanism of electrons via the exchange of virtual phonons, as has been worked out by Bardeen Cooper and Schrieffer in their highly successful BCS theory [2]. It is assumed that the mechanism is of purely electronic origin, for example antiferromagnetic exchange correlations. For this reason spin models are intensively studied [3] which offer many interesting phenomena despite their simple mathematical structure. Many known spin models possess topological phases that do not fit into the Landau symmetry breaking scheme described by standard theory of phase transitions. Topological order is not described by a local order parameter but a new set of quantum numbers like ground state degeneracy and their topology. Critical spin liquids arise from frustration for example the Heisenberg antiferromagnet on a triangular lattice (or other non bipartite lattices like the Kagomé lattice). Ultracold atomic or molecular gases offers interesting possibilities to experimentally realize these strongly correlated quantum phases. In this work cold polar molecules in optical lattices are used to engineer effective spin models which can be easily manipulated by external fields. The strong electric dipole–dipole interactions tunable with external fields and the rotational states that can be used to simulate internal degrees of freedom allow for simulations of spin models in the strong coupling regime [4]. In particular the realization of the XXZ Model and the t - J model describing strongly correlated electrons are investigated by trapping polar molecules in an two dimensional optical lattice.

Unlike classical phase transitions, quantum phase transitions are not driven by thermal fluctuations but they are driven by quantum fluctuations between different ground states at the absolute zero. Different quantum phases can be accessed by varying a physical parameter of the system in question. Interesting quantum phase transitions are the order–disorder transition where two phases are separated by a quantum critical point (indicates a 2nd order phase transition). The quantum critical fluctuations extend over the entire spatial and temporal domain and therefore the quantum critical region can be reached at finite temperature. Prominent example is ⁴He un-

der high pressure and low temperature that leads to a coexistence of supersolid and superfluid phase. Supersolids have long range diagonal and off diagonal ordering combining the properties of a superfluid (zero viscosity flow) and a spatial crystalline ordering.

Here we show the possibility to investigate quantum phase transitions with the help of polar molecules in optical lattices. First an short introduction to describe molecules by a simple model that approximates the behavior of molecules in their electronical and ro-vibrational ground state is given followed by an more extensive discussion about a generic two level system. Next we need to understand how to manipulate polar molecules by applying external electrical fields that couples to the internal charge distribution and concluding Chapter 2 by deriving the dipole-dipole interaction operator for two polar molecules. In Chapter 3 the realization of the Heisenberg model and various relatives thereof, like the XXZ model, are discussed extensively. After given a short overview of the different Heisenberg-like models a detailed derivation of the XXZ Hamiltonian is given in terms of polar molecules in the rotating wave approximation. Applying a spin wave analysis the phase diagram of the XXZ model on a two dimensional optical square lattice with slowly decaying $1/r^3$ interaction is investigated and compared to the phase diagram of the nearest neighbor XXZ model. In the second part the realization of the t - J model is determined by using a different setup of coupled dc-Stark shifted rotational states. Following the calculations in Chapter 3 the t - J model Hamiltonian is derived and the explicit parameters for the realization are calculated as stated in Chapter 4. Finally we conclude this work with a short summary and an outlook addressing the questions what can and needs to be done to actually realize ferro-electric liquids with the help of polar molecules in optical lattices.

2

Basic Concepts of Polar Molecules

2.1 Quantum Rigid Rotor

Polar molecules possess a much more complex spectrum than atomic systems. There exist different electronic-, vibrational-, and rotational excitations of the molecules which can also be coupled to each other. If we only concentrate on rotational excitations the Hamiltonian of free polar molecules corresponds to a rigid rotor

$$\mathcal{H}_{\text{rot}} = B\mathcal{J}^2 \quad (2.1)$$

where B is the rotational constant of the electronic–vibrational ground state (and can in general be regarded to be proportional to the inverse of the inertia tensor $\propto I^{-1}$). \mathcal{J} describes the total internal angular momentum (divided by \hbar) with spherical harmonics $Y_M^J(\theta, \phi)$ as eigenstates to \mathcal{J}^2 and \mathcal{J}_z denoted by $|J, M\rangle$. The spectrum of a rigid rotor is shown in Figure 2.1. The time independent Schrödinger equation for the rigid rotor in spherical coordinates reads

$$\mathcal{H}_{\text{rot}}|J, M\rangle = B\mathcal{J}^2|J, M\rangle = BJ(J+1)|J, M\rangle \quad (2.2)$$

A very instructive introduction to angular momentum operators and their eigenspectrum in terms of spherical harmonics can be found in [5].

2.1.1 Linear Rigid Rotor of a Diatomic Molecule

Considering a linear molecule, it is simple to give an explicit expressions for the rotational constant B . The rotational energy of a linear diatomic molecule is

$$E_{\text{rot}} = \frac{\hbar^2 J(J+1)}{2I} \quad (2.3)$$

where I is the moment of inertia and R the interatomic separation of two atoms with mass m_a and mass m_b , see Figure 2.2. The interesting rotational axis is perpendicular to the connection

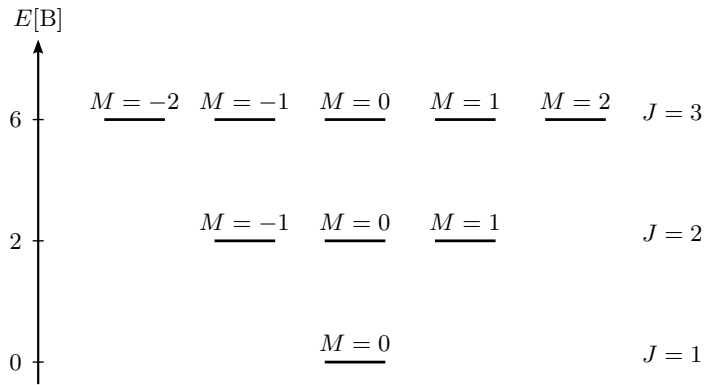


Figure 2.1 Rotational spectrum of a rigid rotor for $J = 0, 1, 2$.

line between the two atoms leading to

$$I = \mu R^2 \qquad \mu = \frac{m_a m_b}{m_a + m_b} \qquad (2.4)$$

since the two-particle problem can be reduced to an one particle problem with reduced mass μ rotating about the rotational axis of the diatomic molecule. By comparing equation (2.2) with (2.3) the rotational constant B of the diatomic molecule can be written as

$$B = \frac{\hbar^2}{2\mu R^2} \qquad (2.5)$$

2.1.2 Selection Rules for Rotational Transitions

Excitations of a rigid rotor can occur through interactions with an external electrical field. The molecule absorbs (or emits) photons with the resonant frequency, i.e. the energy difference between the ground state and the excited state. Typically, for pure rotational transition of molecules this frequency lies in the microwave to far-infrared regime of the electromagnetic wave spectrum ($\approx 1 \dots 100 \text{GHz} \hat{=} 300 \dots 0.3 \text{mm} \hat{=} 4 \cdot 10^{-6} \dots 4 \cdot 10^{-4} \text{eV}$). In order to couple the electromagnetic wave to the molecule, a permanent dipole moment must be present. This can be seen with the help of a multipole expansion and the use of the Wigner-Eckart theorem for spherical tensor operators (see Appendix A). The multipole expansion of the charge distribution of a molecule can be expressed as

$$\rho(\theta, \phi) = \sum_{J=0}^{\infty} \sum_{M=-J}^J A_M^J Y_M^J(\theta, \phi) \qquad (2.6)$$

The expansion coefficient A_M^J depends only on the expanded function. With the help of the Wigner-Eckart theorem (A.13) we find the selection rule for the different multipole coefficients

- Monopole A_0^0 :

$$\langle J' M' | A_0^0 | J M \rangle \neq 0 \quad \text{for } M' = M \text{ and } J' = J \qquad (2.7)$$

- Dipole A_{-1}^1, A_0^1, A_1^1 :

$$\langle J' M' | A_q^1 | J M \rangle \neq 0 \quad \text{for } M - M' = -1, 0, 1 \text{ and } J' = J + 1 \qquad (2.8)$$

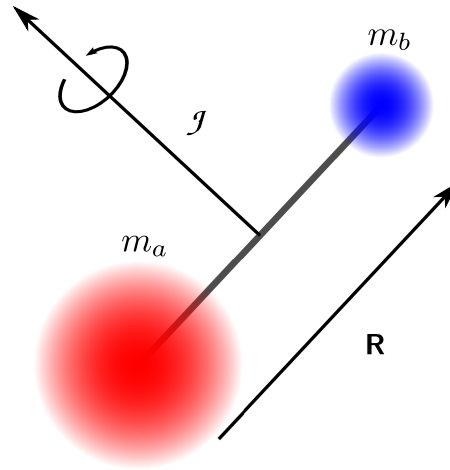


Figure 2.2 Diatomic molecule with masses m_a and m_b separated by $|\mathbf{R}|$.

- Quadrupole $A_{-2}^2, A_{-1}^2, A_0^2, A_1^2, A_2^2$:

$$\langle J' M' | A_q^2 | J M \rangle \quad \text{for } M - M' = q \text{ and } J' = J + 2, J + 1 \quad (2.9)$$

where $J' > J$ is assumed. The monopole term is proportional to the unit operator $\mathbb{1}$ and cannot couple different rotational states. Therefore only the dipole vector operator $\mathcal{d} \propto C_q^{(1)}(\theta, \phi)$ (and all higher tensor operators) are able to couple excited rotational states to the ground state, where $C_q^{(k)}(\theta, \phi)$ describes unnormalized spherical harmonics defined in equation (A.11). In the following only the dipole approximation will be employed because the electric dipole term is dominating the transition between ground state and excited states (and electric dipole transitions are much more likely to happen) than those of higher multipole terms.

2.2 Two Level Systems

Starting with a general discussion of a quantum mechanical two level system, we will derive general properties and formulae which can be applied to precise problems later on. In Section 2.3 at first an external static electric fields will be applied to polar molecules generating a Stark-shifted two level system. In a second step an external microwave field is additionally switched on to couple two Stark-shifted rotational states of the molecule, e.g. $|\phi_{00}^{\text{num}}\rangle \leftrightarrow |\phi_{10}^{\text{num}}\rangle$.

The most common description of a two level system is the Pauli representation of the $su(2)$ -algebra, i.e. mapping the two states onto a spin- $1/2$ system

$$|e\rangle = |\uparrow\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad |g\rangle = |\downarrow\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad (2.10)$$

where $|g\rangle$ describes the ground state and $|e\rangle$ the excited state of the two level system. The basis state of the spin- $1/2$ system $\{|\uparrow\rangle, |\downarrow\rangle\}$ allow a particular matrix representation of the $su(2)$ -algebra extensively explained in Appendix G.3 and in various group theoretical (or quantum mechanics) textbooks [6, 7]. A two level system can be described by a simple Hamiltonian

$$\mathcal{H}^{(0)} = E_{\uparrow} |\uparrow\rangle \langle\uparrow| + E_{\downarrow} |\downarrow\rangle \langle\downarrow| \quad (2.11)$$

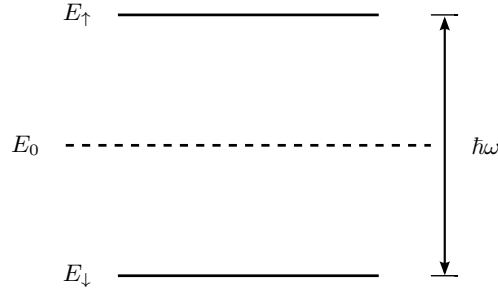


Figure 2.3 Two level system with ground state energy $E_{\downarrow} = E_0 - \frac{\hbar\omega}{2}$ and excited energy $E_{\uparrow} = E_0 + \frac{\hbar\omega}{2}$.

where E_{\uparrow} and E_{\downarrow} are the eigenenergies of $\mathcal{H}^{(0)}$. Writing this Hamiltonian with the help of the spin- $1/2$ raising and lowering operators represented by Pauli matrices σ defined in (G.7)

$$\sigma_+ = \frac{1}{2}(\sigma_x + i\sigma_y) = |\uparrow\rangle\langle\downarrow| \quad \sigma_- = \frac{1}{2}(\sigma_x - i\sigma_y) = |\downarrow\rangle\langle\uparrow| \quad \sigma_z = |\uparrow\rangle\langle\uparrow| - |\downarrow\rangle\langle\downarrow| \quad (2.12)$$

gives rise to

$$\mathcal{H}^{(0)} = \frac{1}{2} \underbrace{(E_{\uparrow} - E_{\downarrow})}_{\hbar\omega} \sigma_z + \frac{1}{2} \underbrace{(E_{\uparrow} + E_{\downarrow})}_{E_0} \mathbb{1} \quad (2.13)$$

with E_0 describing the centered energy and $E_{\uparrow} - E_{\downarrow}$ the transition energy. A general state of the two level system $|\psi\rangle$ can be written as superposition

$$|\psi\rangle = |\uparrow\rangle\langle\uparrow|\psi\rangle + |\downarrow\rangle\langle\downarrow|\psi\rangle = \psi_{\uparrow}|\uparrow\rangle + \psi_{\downarrow}|\downarrow\rangle = \begin{pmatrix} \psi_{\uparrow} \\ \psi_{\downarrow} \end{pmatrix} \quad (2.14)$$

The time evolution according to the full time dependent Schrödinger equation is given by the unitary time evolution operator

$$\mathcal{U}(t, t_0) = \mathcal{T} \left[e^{-\frac{i}{\hbar} \int_{t_0}^t dt' \mathcal{H}(t')} \right] \quad (2.15)$$

where \mathcal{T} is the time ordering operator. For Hamilton operators that are not explicitly time dependent, i.e. $\frac{\partial \mathcal{H}}{\partial t} = 0$, the unitary time evolution operator (2.15) can be simplified to

$$\mathcal{U}(t, t_0) = e^{-\frac{i}{\hbar}(t-t_0)\mathcal{H}} \quad (2.16)$$

Acting with $\mathcal{U}(t, t_0)$ on an arbitrary two level state $|\psi\rangle$ yields

$$\begin{aligned} |\psi(t)\rangle &= \mathcal{U}(t, t_0) |\psi(t_0)\rangle \\ &= e^{-\frac{i}{\hbar}(t-t_0)\mathcal{H}^{(0)}} |\psi(t_0)\rangle \\ &= \underbrace{e^{-\frac{i}{\hbar}(t-t_0)E_{\uparrow}} \psi_{\uparrow}(t_0)}_{\psi_{\uparrow}(t)} |\uparrow\rangle + \underbrace{e^{-\frac{i}{\hbar}(t-t_0)E_{\downarrow}} \psi_{\downarrow}(t_0)}_{\psi_{\downarrow}(t)} |\downarrow\rangle \end{aligned} \quad (2.17)$$

since $|\uparrow\rangle$ and $|\downarrow\rangle$ are eigenstates of $\mathcal{H}^{(0)}$ or solutions to the time independent Schrödinger equation respectively. Preparing a two level system in the ground state $|g\rangle = |\downarrow\rangle$ at $t_0 = 0$ it can be shown that the system cannot leave its initially prepared state since the norm

$$\| |\psi(0)\rangle \| = \| \psi_{\downarrow}(0) |\downarrow\rangle \| = |\psi_{\downarrow}(0)| = 1 \quad (2.18)$$

is preserved under unitary transformations

$$\begin{aligned} \| |\psi(t)\rangle \|^2 &= \| \mathcal{U}(t, 0) |\psi(0)\rangle \|^2 \\ &= |\langle \mathcal{U}(t, 0) \psi(0) | \mathcal{U}(t, 0) \psi(0) \rangle|^2 \\ &= |\langle \psi(0) | \mathcal{U}(t, 0)^\dagger \mathcal{U}(t, 0) | \psi(0) \rangle|^2 \\ &= |\psi_{\downarrow}(0)|^2 \end{aligned} \quad (2.19)$$

so that the system remains in the ground state for all times t . This is easily seen because the probability of finding the system in a state $|\alpha\rangle$ is

$$P_{\alpha}(t) = |\langle \alpha | \psi(t) \rangle|^2 \quad (2.20)$$

and hence with the preparation of the system in the ground state $|\psi(0)\rangle = |\downarrow\rangle$ the probability of the system being in $|\downarrow\rangle$ is

$$\begin{aligned} P_{\downarrow}(t) &= |\langle \downarrow | \psi(t) \rangle|^2 = |\langle \downarrow | \mathcal{U}(t, 0) | \psi(0) \rangle|^2 \\ &= |\langle \downarrow | \mathcal{U}(t, 0) | \downarrow \rangle|^2 = \left| \langle \downarrow | e^{-\frac{i}{\hbar} t \mathcal{H}^{(0)}} | \downarrow \rangle \right|^2 \\ &= \underbrace{|\langle \downarrow | \downarrow \rangle|^2}_{=1} \underbrace{\left| e^{-\frac{i}{\hbar} E_{\downarrow} t} \right|^2}_{=1} = 1 \end{aligned} \quad (2.21)$$

and for the probability of the system being in $|\uparrow\rangle$ we find

$$\begin{aligned} P_{\uparrow}(t) &= |\langle \uparrow | \psi(t) \rangle|^2 \\ &= |\langle \uparrow | \mathcal{U}(t, 0) | \downarrow \rangle|^2 = \underbrace{|\langle \uparrow | \downarrow \rangle|^2}_{=0} \left| e^{-\frac{i}{\hbar} E_{\downarrow} t} \right|^2 \end{aligned} \quad (2.22)$$

as would be expected since the probability distribution must be normalized

$$\sum_{i=1}^{\dim \mathcal{H}^{(0)}} P_i(t) = 1 \quad \Leftrightarrow \quad P_{\downarrow}(t) + P_{\uparrow}(t) = 1 \quad (2.23)$$

In order to induce a “spin flip” i.e. to bring the system from its ground state to the excited state, an external perturbation is needed.

$$\mathcal{H} = \mathcal{H}^{(0)} + \mathcal{H}' = \begin{pmatrix} E_{\uparrow} & 0 \\ 0 & E_{\downarrow} \end{pmatrix} + \begin{pmatrix} H'_{11} & H'_{12} \\ (H'_{12})^* & H'_{22} \end{pmatrix} \quad (2.24)$$

The Hamiltonian \mathcal{H}' describing the external perturbation can be divided in two diagonal elements and two off-diagonal elements. The diagonal elements are shifting the eigenenergies of $\mathcal{H}^{(0)}$ to $E_{\downarrow} + H'_{11}$ and $E_{\uparrow} + H'_{22}$ and therefore can be absorbed in the unperturbed Hamiltonian (2.11). Thus, the diagonal elements of \mathcal{H}' can be set to zero. Due to the fact that the Hamiltonian is a hermitian operator, the off-diagonal elements are complex conjugate to each other and can be written in the polar representation of complex numbers leading to the matrix representation

$$\mathcal{H} = \begin{pmatrix} E_{\uparrow} & |H'_{12}| e^{-i \arg H'_{12}} \\ |H'_{12}| e^{-i \arg H'_{12}} & E_{\downarrow} \end{pmatrix} \quad (2.25)$$

The operator \mathcal{H} can be diagonalized with the help of the formulae in Appendix E. Equation (E.12) states that the eigenenergies of a 2×2 -matrix are

$$\begin{aligned} E_{\pm} &= \frac{E_{\uparrow} + E_{\downarrow}}{2} \pm \sqrt{\left(\frac{E_{\uparrow} - E_{\downarrow}}{2}\right)^2 + |H'_{12}|^2} \\ &= E_0 \pm \sqrt{\left(\frac{\hbar\omega}{2}\right)^2 + |H'_{12}|^2} \end{aligned} \quad (2.26)$$

Setting $\arg H'_{12} = \gamma$ the corresponding eigenstates are

$$\begin{aligned} |+\rangle &= \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |\uparrow\rangle + \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |\downarrow\rangle \\ |-\rangle &= -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |\uparrow\rangle + \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |\downarrow\rangle \end{aligned} \quad (2.27)$$

where the “mixing” angle φ is given as

$$\varphi = \arctan \left(\frac{2|H'_{12}|}{E_{\uparrow} - E_{\downarrow}} \right) = \arctan \left(2 \frac{|H'_{12}|}{\hbar\omega} \right) \quad (2.28)$$

The diagonalization of \mathcal{H} can be seen as a two dimensional rotation in the space spanned by $\{|\uparrow\rangle, |\downarrow\rangle\}$ with rotation angle $\varphi/2$. The time evolution of the two level system is given according to equation (2.16) by

$$|\psi(t)\rangle_{\pm} = \mathcal{U}_{\pm}(t, 0) |\psi(0)\rangle_{\pm} \quad (2.29)$$

The subscript \pm indicates the basis in which the components of the state vector and the matrix elements of the time evolution operator are represented. In order to calculate the probabilities of finding a prepared system in one of the two distinct states after some time, the time evolution must be expressed in the $\{|\uparrow\rangle, |\downarrow\rangle\}$ basis. Following equation (E.43) the time evolution can be written as

$$|\psi(t)\rangle_{\uparrow\downarrow} = Q \mathcal{U}_{\pm}(t, 0) Q^{\dagger} |\psi(0)\rangle_{\uparrow\downarrow} = \mathcal{U}_{\uparrow\downarrow} |\psi(0)\rangle_{\uparrow\downarrow} \quad (2.30)$$

where Q is the transformation matrix that diagonalizes \mathcal{H} . This can be seen by writing out the transformation of \mathcal{H} explicitly

$$\begin{aligned} \mathcal{U}_{\uparrow\downarrow}(t, 0) &= e^{-\frac{i}{\hbar} \mathcal{H}_{\uparrow\downarrow} t} = e^{-\frac{i}{\hbar} (Q \mathcal{H}_{\pm} Q^{\dagger}) t} \\ &= \sum_{k=0}^{\infty} \left(-\frac{i}{\hbar} \right)^k \frac{(Q \mathcal{H}_{\pm} Q^{\dagger})^k t^k}{k!} \end{aligned}$$

$$\begin{aligned}
&= 1 + \left(-\frac{i}{\hbar}\right) Q \mathcal{H}_{\pm} Q^{\dagger} t + \left(-\frac{i}{\hbar}\right) \frac{Q \mathcal{H}_{\pm} Q^{\dagger} Q \mathcal{H}_{\pm} Q^{\dagger}}{2} t^2 + \dots \\
&\quad \dots + \left(-\frac{i}{\hbar}\right)^l \frac{Q \mathcal{H}_{\pm} Q^{\dagger} \dots \mathcal{H}_{\pm} \mathbb{1} \mathcal{H}_{\pm} \dots Q \mathcal{H}_{\pm} Q^{\dagger}}{l!} t^l + \dots \\
&= Q \left[\sum_{k=0}^{\infty} \left(-\frac{i}{\hbar}\right)^k \frac{\mathcal{H}_{\pm}^k}{k!} t^k \right] Q^{\dagger} = Q e^{-\frac{i}{\hbar} \mathcal{H}_{\pm} t} Q^{\dagger} \\
&= Q \mathcal{U}_{\pm}(t, 0) Q^{\dagger}
\end{aligned} \tag{2.31}$$

Thus the time evolution operator $\mathcal{U}_{\uparrow\downarrow}(t, 0)$ in the $\{|\uparrow\rangle, |\downarrow\rangle\}$ representation reads

$$\begin{aligned}
\mathcal{U}_{\uparrow\downarrow} &= \begin{pmatrix} \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} & -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \\ \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} & \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \end{pmatrix} \begin{pmatrix} e^{-\frac{i}{\hbar} E_+ t} & 0 \\ 0 & e^{-\frac{i}{\hbar} E_- t} \end{pmatrix} \begin{pmatrix} \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} & \sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \\ -\sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} & \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \end{pmatrix} \\
&= \begin{pmatrix} \cos^2 \frac{\varphi}{2} e^{-\frac{i}{\hbar} E_+ t} + \sin^2 \frac{\varphi}{2} e^{-\frac{i}{\hbar} E_- t} & \cos \frac{\varphi}{2} \sin \frac{\varphi}{2} e^{-i\gamma} \left[e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right] \\ \cos \frac{\varphi}{2} \sin \frac{\varphi}{2} e^{i\gamma} \left[e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right] & \sin^2 \frac{\varphi}{2} e^{-\frac{i}{\hbar} E_+ t} + \cos^2 \frac{\varphi}{2} e^{-\frac{i}{\hbar} E_- t} \end{pmatrix}
\end{aligned} \tag{2.32}$$

With the help of trigonometric identities

$$\cos \frac{\varphi}{2} \sin \frac{\varphi}{2} = \frac{1}{2} \sin \varphi \quad \cos^2 \frac{\varphi}{2} = \frac{1}{2} (1 + \cos \varphi) \quad \sin^2 \frac{\varphi}{2} = \frac{1}{2} (1 - \cos \varphi) \tag{2.33}$$

$\mathcal{U}_{\uparrow\downarrow}$ can eventually be written as

$$\begin{aligned}
\mathcal{U}_{\uparrow\downarrow} &= \frac{1}{2} \left[\left(e^{-\frac{i}{\hbar} E_+ t} + e^{-\frac{i}{\hbar} E_- t} \right) + \cos \varphi \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\uparrow\rangle \langle\uparrow| \\
&\quad + \frac{1}{2} \left[\sin \varphi e^{i\gamma} \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\uparrow\rangle \langle\downarrow| \\
&\quad + \frac{1}{2} \left[\sin \varphi e^{-i\gamma} \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\downarrow\rangle \langle\uparrow| \\
&\quad + \frac{1}{2} \left[\left(e^{-\frac{i}{\hbar} E_+ t} + e^{-\frac{i}{\hbar} E_- t} \right) - \cos \varphi \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\downarrow\rangle \langle\downarrow|
\end{aligned} \tag{2.34}$$

In the following the subscript $\uparrow\downarrow$ will be suppressed. Now, for a two level system prepared in its ground state $|\psi(0)\rangle = |g\rangle = |\downarrow\rangle$, one will observe oscillation between the ground and excited state with frequency Ω that corresponds to the difference between the eigenenergies $E_+ - E_-$

$$\begin{aligned}
|\psi^{\downarrow}(t)\rangle &= \mathcal{U}(t, 0) |\psi(0)\rangle = \mathcal{U} |\downarrow\rangle \\
&= \frac{1}{2} \left[\sin \varphi e^{i\gamma} \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\uparrow\rangle \\
&\quad + \frac{1}{2} \left[\left(e^{-\frac{i}{\hbar} E_+ t} + e^{-\frac{i}{\hbar} E_- t} \right) - \cos \varphi \left(e^{-\frac{i}{\hbar} E_+ t} - e^{-\frac{i}{\hbar} E_- t} \right) \right] |\downarrow\rangle
\end{aligned} \tag{2.35}$$

As can be seen in equation (2.35) the two states are intermingled and cannot be separated anymore. The probability of finding such a prepared system in the excited state is the absolute

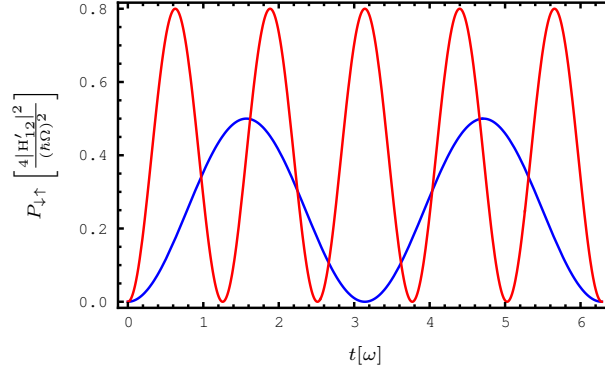


Figure 2.4 Oscillations between the ground state and the excited state of the two level system. The blue curve shows $P_{\downarrow\uparrow}$ with $|H'_{12}|/\hbar\omega = 1/2$ and the red curve $|H'_{12}|/\hbar\omega = 1$.

square of the projection onto $|e\rangle = |\uparrow\rangle$. In this case, we expect to have a time dependency of the probability

$$\begin{aligned}
 P_{\downarrow\uparrow} &= |\langle\uparrow|\psi^\downarrow(t)\rangle|^2 \\
 &= \frac{1}{4} \sin^2 \varphi \left| e^{-\frac{i}{\hbar}E_+t} - e^{-\frac{i}{\hbar}E_-t} \right|^2 \\
 &= \frac{1}{4} \sin^2 \varphi \left(e^{\frac{i}{\hbar}E_+t} - e^{\frac{i}{\hbar}E_-t} \right) \left(e^{-\frac{i}{\hbar}E_+t} - e^{-\frac{i}{\hbar}E_-t} \right) \\
 &= \frac{1}{4} \sin^2 \varphi \left(1 - e^{-\frac{i}{\hbar}(E_+ - E_-)t} - e^{\frac{i}{\hbar}(E_+ - E_-)t} + 1 \right) \\
 &= \frac{1}{4} \sin^2 \varphi \left[2 - 2 \cos \left(\frac{E_+ - E_-}{\hbar} t \right) \right] \\
 &= \sin^2 \varphi \cdot \sin^2 \left(\frac{E_+ - E_-}{2\hbar} t \right)
 \end{aligned} \tag{2.36}$$

Inserting φ from equation (2.28) and defining

$$\begin{aligned}
 \hbar\Omega &= E_+ - E_- = 2\sqrt{\left(\frac{E_\uparrow - E_\downarrow}{2}\right)^2 + |H'_{12}|^2} \\
 \hookrightarrow \hbar\Omega &= \sqrt{(\hbar\omega)^2 + 4|H'_{12}|^2}
 \end{aligned} \tag{2.37}$$

the system will oscillate between excited and ground state as can be seen in Figure 2.4

$$P_{\downarrow\uparrow} = \frac{|H'_{12}|^2}{\left(\frac{E_\uparrow - E_\downarrow}{2}\right)^2 + |H'_{12}|^2} \sin^2 \left(\frac{\Omega}{2} t \right) = \frac{4|H'_{12}|^2}{(\hbar\Omega)^2} \sin^2 \left(\frac{\Omega}{2} t \right) \tag{2.38}$$

Analogously the same result can be derived for the reversed setup, starting from the excited

state, thus $P_{\downarrow\uparrow} = P_{\uparrow\downarrow}$. The unnormalized probability of staying in the same state can be calculated from the projection onto the initially prepared state

$$\begin{aligned}
P_{\downarrow\downarrow} = P_{\uparrow\uparrow} &= |\langle\downarrow|\psi^\downarrow(t)\rangle|^2 \\
&= \frac{1}{4} \left| \left(e^{-\frac{i}{\hbar}E_+t} + e^{-\frac{i}{\hbar}E_-t} \right) - \cos\varphi \left(e^{-\frac{i}{\hbar}E_+t} - e^{-\frac{i}{\hbar}E_-t} \right) \right|^2 \\
&= \frac{1}{2} \left[1 + \cos\left(\frac{E_+ - E_-}{\hbar}t\right) + \cos^2\varphi \left(1 - \cos\left(\frac{E_+ - E_-}{\hbar}t\right) \right) \right] \\
&= \cos^2\left(\frac{\Omega}{2}t\right) + \cos^2\varphi \cdot \sin^2\left(\frac{\Omega}{2}t\right) \\
&= \cos^2\left(\frac{\Omega}{2}t\right) + \frac{\left(\frac{E_+ - E_-}{2}\right)^2}{\left(\frac{E_+ - E_-}{2}\right)^2 + |H'_{12}|^2} \sin^2\left(\frac{\Omega}{2}t\right) \\
&= \cos^2\left(\frac{\Omega}{2}t\right) + \frac{\omega^2}{\Omega^2} \sin^2\left(\frac{\Omega}{2}t\right) \tag{2.39}
\end{aligned}$$

In general the time evolution can be represented in the rotating basis $\{|+\rangle, |-\rangle\}$ or in the original two level system basis $\{|\uparrow\rangle, |\downarrow\rangle\}$ where the two states are mixed

$$\begin{aligned}
|\psi(t)\rangle &= \psi_+(t)|+\rangle + \psi_-(t)|-\rangle \\
&= e^{-\frac{i}{\hbar}E_+t} \psi_+(0)|+\rangle + e^{-\frac{i}{\hbar}E_-t} \psi_-(0)|-\rangle \\
&= \left(\cos\frac{\varphi}{2} e^{i\frac{\gamma}{2}} \psi_\uparrow(0) + \sin\frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \psi_\downarrow(0) \right) e^{-\frac{i}{\hbar}E_+t} |+\rangle \\
&\quad + \left(-\sin\frac{\varphi}{2} e^{i\frac{\gamma}{2}} \psi_\uparrow(0) + \cos\frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \psi_\downarrow(0) \right) e^{-\frac{i}{\hbar}E_-t} |-\rangle \tag{2.40}
\end{aligned}$$

where $\psi_{\uparrow\downarrow}(0)$ are the components of the initial two level state vector at $t_0 = 0$ and φ is defined in equation (2.28). In the $\{|\uparrow\rangle, |\downarrow\rangle\}$ basis $|\psi(t)\rangle$ can be written as

$$\begin{aligned}
|\psi(t)\rangle &= e^{-\frac{i}{\hbar}E_0t} \left\{ \left[\left(\cos^2\left(\frac{\Omega}{2}t\right) - i\frac{\omega}{\Omega} \sin^2\left(\frac{\Omega}{2}t\right) \right) \psi_\uparrow(0) - 2i\frac{|H'_{12}|}{\hbar\Omega} \sin^2\left(\frac{\Omega}{2}t\right) e^{-i\gamma} \psi_\downarrow(0) \right] |\uparrow\rangle \right. \\
&\quad \left. + \left[-2i\frac{|H'_{12}|}{\hbar\Omega} \sin^2\left(\frac{\Omega}{2}t\right) e^{i\gamma} \psi_\downarrow(0) + \left(\cos^2\left(\frac{\Omega}{2}t\right) + i\frac{\omega}{\Omega} \sin^2\left(\frac{\Omega}{2}t\right) \right) \psi_\downarrow(0) \right] |\downarrow\rangle \right\} \tag{2.41}
\end{aligned}$$

with $E_0 = \frac{1}{2}(E_\uparrow + E_\downarrow) = \frac{1}{2}(E_+ + E_-)$, the transition frequency ω of the two level states and the oscillation frequency $\Omega = \sqrt{\omega^2 + 4/\hbar^2 |H'_{12}|^2}$ between the two states. The effect of the non-diagonal element of the operator \mathcal{H}' on the two level system is on the one hand the oscillation between the two states for a system prepared only in one state (e.g. the ground state), and on the other hand the additional symmetric energy shift that scales with the magnitude of $|H'_{12}|$. This effect is visualized in Figure 2.5, but without the effect that the diagonal elements of \mathcal{H}' would shift E_0 to $E_0 + \frac{1}{2}(H'_{11} + H'_{22})$ and the transition energy $\hbar\omega$ to $\hbar\omega + H'_{11} - H'_{22}$. In Table 2.1 the unperturbed and the perturbed two level systems will be compared in a slightly different language namely

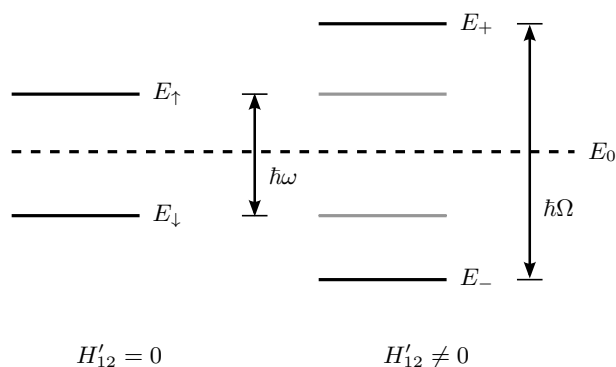


Figure 2.5 For $H'_{12} = 0$ there are two states with transition energy $\hbar\omega$. Switching on the perturbation (off-diagonal elements of \mathcal{H}') $H'_{12} \neq 0$ leads to an symmetric energy shift (with respect to E_0) and a new transition energy $\hbar\Omega$.

using the Pauli representation of spin- $1/2$ systems. Note that in spin language the diagonalization of \mathcal{H} can be seen as a rotation of the coordinate system onto the arbitrary direction of σ , such that the quantization axis in the new coordinates are along the z -axis.

$$* \quad \mathbf{h} \cdot \boldsymbol{\sigma} = \begin{pmatrix} h_z & h_x - ih_y \\ h_x + ih_y & -h_z \end{pmatrix} \quad \text{with} \quad \mathbf{h} = \begin{pmatrix} \Re(H'_{12}) \\ -\Im(H'_{12}) \\ \frac{\hbar\omega}{2} \end{pmatrix}$$

Hamiltonian	$\mathcal{H} = \mathcal{H}^{(0)} + \mathcal{H}'$	$\mathcal{H}^{(0)}$
Eigenenergies	$E_+ = E_0 + \frac{\hbar\Omega}{2}$ $E_- = E_0 - \frac{\hbar\Omega}{2}$	$E_\uparrow = E_0 + \frac{\hbar\omega}{2}$ $E_\downarrow = E_0 - \frac{\hbar\omega}{2}$
Representation $\{ \downarrow\rangle, \uparrow\rangle\}$	$\mathcal{H} = \mathbf{h} \cdot \boldsymbol{\sigma}_{\uparrow\downarrow} + E_0 \mathbb{1}_{\uparrow\downarrow}$	$\mathcal{H}^{(0)} = \frac{\hbar\omega}{2} \sigma_z \uparrow\downarrow + E_0 \mathbb{1}_{\uparrow\downarrow}$
Special case $E_0 = 0, \gamma = 0$	$\mathcal{H} = \frac{\hbar\omega}{2} \sigma_z \uparrow\downarrow + H'_{12} \sigma_x \uparrow\downarrow$	$\mathcal{H}^{(0)} = \frac{\hbar\omega}{2} \sigma_z \uparrow\downarrow$
Representation $\{ +\rangle, -\rangle\}$	$\mathcal{H} = \frac{\hbar\Omega}{2} \sigma_z \pm + E_0 \mathbb{1}_\pm$	—
Transition energy	$\hbar\Omega = \sqrt{(\hbar\omega)^2 + 4 H'_{12} ^2}$	$\hbar\omega$

Table 2.1 Comparison between the two level system with $|H'_{12}| \neq 0$ and without perturbation $|H'_{12}| = 0$.

2.3 Polar Molecules in External Electric Fields

2.3.1 Dipole Approximation

The internal charge distribution of the molecule leads to an permanent dipole moment which is describe by the dipole vector operator \mathbf{d} . In the dipole–approximation this is the leading term that is sensitive to an external electrical field. This can be derived from the classical electrostatic energy of an external electrical field (represented by Φ^{ext}) acting on a charge distribution $\rho(\mathbf{r}')$

$$V_\rho = \int_V d^3\mathbf{r}' \rho(\mathbf{r}') \Phi^{\text{ext}}(\mathbf{r}') \quad (2.42)$$

The potential $\Phi(\mathbf{r})$ can be expanded in a series expansion around $\mathbf{r} = \mathbf{0}$

$$\begin{aligned} \Phi^{\text{ext}}(\mathbf{r}) &= \Phi(\mathbf{0}) + \mathbf{r} \cdot (\nabla\Phi)\Big|_0 + \frac{1}{2}(\mathbf{r} \otimes \mathbf{r}) : (\nabla \otimes (\nabla\Phi))\Big|_0 \\ &= \Phi(\mathbf{0}) - \mathbf{r} \cdot \mathbf{E}(\mathbf{0}) - \frac{1}{2}(\mathbf{r} \otimes \mathbf{r}) : (\nabla \otimes \mathbf{E})\Big|_0 \end{aligned} \quad (2.43)$$

This expansion up to second order is only accurate if the source of the potential at $\mathbf{r} = \mathbf{0}$ is far away from the charge distribution it is acting on. In this case the last term of the expansion (2.43) can be rewritten in the following form using the fact that there are no sources. Hence $\nabla \cdot \mathbf{E} = 0$ and by applying the relation $\nabla \cdot \mathbf{E} = \mathbf{1} : (\nabla \otimes \mathbf{E})$ we find

$$\begin{aligned} -\frac{1}{2}(\mathbf{r} \otimes \mathbf{r}) : (\nabla \otimes \mathbf{E})\Big|_0 &= -\frac{1}{6} \left[3(\mathbf{r} \otimes \mathbf{r}) : (\nabla \otimes \mathbf{E}) - r^2 \nabla \cdot \mathbf{E} \right] \Big|_0 \\ &= -\frac{1}{6} (3(\mathbf{r} \otimes \mathbf{r}) - r^2 \mathbf{1}) : (\nabla \otimes \mathbf{E})\Big|_0 \end{aligned} \quad (2.44)$$

Reinserting the expansion (2.43) in the integral (2.42) gives rise to

$$\begin{aligned}
V_\rho &= \int_V d^3\mathbf{r}' \rho(\mathbf{r}') \left[\Phi(\mathbf{0}) - \mathbf{r}' \cdot \mathbf{E}(\mathbf{0}) - \frac{1}{6} \left((3(\mathbf{r}' \otimes \mathbf{r}') - r'^2 \mathbf{1}) : (\nabla \otimes \mathbf{E}) \right) \Big|_0 \right] \\
&= \underbrace{\int_V d^3\mathbf{r}' \rho(\mathbf{r}') \Phi(\mathbf{0})}_Q - \underbrace{\int_V d^3\mathbf{r}' \rho(\mathbf{r}') \mathbf{r}' \cdot \mathbf{E}(\mathbf{0})}_\mathbf{d} - \underbrace{\frac{1}{6} \int_V d^3\mathbf{r}' \rho(\mathbf{r}') (3(\mathbf{r}' \otimes \mathbf{r}') - r'^2 \mathbf{1}) : (\nabla \otimes \mathbf{E}) \Big|_0}_q \\
&= Q\Phi^{\text{ext}}(\mathbf{0}) - \mathbf{d} \cdot \mathbf{E}^{\text{ext}}(\mathbf{0}) - \frac{1}{6} q : (\nabla \otimes \mathbf{E}^{\text{ext}}) \Big|_0
\end{aligned} \tag{2.45}$$

The Q -term is the monopole part, the \mathbf{d} -term the dipole part and the q -term is the quadrupole part of the total energy V_ρ of the charge distribution. The monopole reacts directly to the external electrical potential Φ^{ext} , whereas the dipole is sensitive to the external electric field \mathbf{E}^{ext} and the higher multipole terms are sensitive to spatial derivations of the external electric field. In the following the dipole approximation is assumed since the spatial changes of the external electric field are very small compared to the spatial extension of a typical molecule, e.g. compare the wave length of microwaves $\approx 10^{-3}\text{m}$ to the dimension of a molecule $\approx 10^{-9}\text{m}$. Polar molecules are electrical neutral and therefore the integral over the charge distribution of the molecule will vanish, leaving only the dipole term of equation (2.45)

$$V_\rho \approx -\mathbf{d} \cdot \mathbf{E}^{\text{ext}} \tag{2.46}$$

Note that only the classical position is promoted to an operator – the position operator r – and hence the dipole moment is also described by an operator whereas the external electric field is still treated classically. Applying an external static electrical field \mathbf{E}_{dc} to a polar molecule leads to the following Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{rot}} + \mathcal{H}_{\text{dc}} = B\mathcal{J}^2 - \mathbf{d} \cdot \mathbf{E}_{\text{dc}} \tag{2.47}$$

In general this Hamiltonian cannot be solved directly, so for weak electrical fields $E_{\text{dc}} \ll B/d$ one can apply a time independent perturbation calculation. Before doing that we first want to express the dipole operator \mathbf{d} in spherical coordinates because then it is straightforward to express \mathbf{d} in the eigenbasis of \mathcal{J}^2 . The spherical components of the dipole operator in a spherical basis $\{\mathbf{e}_{-1}, \mathbf{e}_0, \mathbf{e}_1\}$ are proportional to (unnormalized) spherical harmonics $C_q^{(k)}(\theta, \phi)$ with rank $k = 0$. Thus we can write

$$d_q = dC_q^{(1)}(\theta, \phi) \tag{2.48}$$

The components can be directly calculated via the projection

$$\mathbf{e}_r \cdot \mathbf{d} = \sum_{q=-1}^1 \mathbf{e}_r \cdot (\mathbf{e}_q \otimes \mathbf{e}_q) \mathbf{d} = \sum_{q=-1}^1 \underbrace{(\mathbf{e}_r \cdot \mathbf{e}_q)}_{C_q^{(1)}(\theta, \phi)} \underbrace{(\mathbf{e}_q \cdot \mathbf{d})}_{d_q} = \sum_{q=-1}^1 C_q^{(1)}(\theta, \phi) d_q \tag{2.49}$$

Calculating the unnormalized spherical harmonics $C_q^{(k)}$ for $k = 1$ and $q = -1, 0, 1$ using the position vector of a unit sphere

$$\mathbf{e}_r = \begin{pmatrix} \cos \phi \sin \theta \\ \sin \phi \sin \theta \\ \cos \theta \end{pmatrix} \quad (2.50)$$

and the spherical basis as defined in Appendix C, equation (C.12)

$$C_0^{(1)} = \mathbf{e}_0 \cdot \mathbf{e}_r = \mathbf{e}_z \cdot \mathbf{e}_r = \cos \theta \quad (2.51)$$

$$\begin{aligned} C_{\pm 1}^{(1)} &= \mathbf{e}_{\pm 1} \cdot \mathbf{e}_r = \mp \frac{1}{\sqrt{2}} (\mathbf{e}_x \pm i\mathbf{e}_y) \cdot \mathbf{e}_r \\ &= \mp \frac{1}{\sqrt{2}} (\cos \phi \sin \theta \pm i \sin \phi \sin \theta) \\ &= \mp \frac{1}{\sqrt{2}} e^{\pm i\phi} \sin \theta \end{aligned} \quad (2.52)$$

The unnormalized spherical harmonics $C_q^{(k)}$ can be compared to the spherical harmonics $Y_M^J(\theta, \phi)$, see Table A.1. Writing out the terms of equation (2.49)

$$\begin{aligned} \mathbf{e}_r \cdot \mathbf{d} &= C_{-1}^{(1)}(\theta, \phi) d_{-1} + C_0^{(1)}(\theta, \phi) d_0 C_1^{(1)}(\theta, \phi) d_1 \\ &= \frac{1}{\sqrt{2}} e^{-i\phi} \sin \theta d_{-1} + \cos \theta d_0 - \frac{1}{\sqrt{2}} e^{i\phi} \sin \theta d_1 \\ &= \cos \phi \sin \theta d_x + \sin \phi \sin \theta d_y + \cos \theta d_z \end{aligned} \quad (2.53)$$

and comparing the coefficients give rise to the representation of the dipole operator in spherical coordinates

$$\begin{aligned} \cos \phi \sin \theta : \quad d_x &= \frac{1}{\sqrt{2}} d_{-1} - \frac{1}{\sqrt{2}} d_1 = -\frac{1}{\sqrt{2}} (d_1 - d_{-1}) \\ \sin \phi \sin \theta : \quad d_y &= -\frac{i}{\sqrt{2}} d_{-1} - \frac{i}{\sqrt{2}} d_1 = -\frac{i}{\sqrt{2}} (d_1 + d_{-1}) \\ \cos \theta : \quad d_z &= d_0 \end{aligned} \quad (2.54)$$

A different definition of the components of the electric dipole operator is given in analogy to raising and lowering operators of the angular momentum representation of the $SU(2)$, namely S_+ , S_z and S_- . The projection onto \mathbf{e}_r reads

$$\mathbf{e}_r \cdot \mathbf{d} = \sum_{q=-1}^1 (-1)^q C_q^{(1)} d_{-q} = -C_{-1}^{(1)}(\theta, \phi) d_+ + C_0^{(1)}(\theta, \phi) d_0 - C_1^{(1)}(\theta, \phi) d_- \quad (2.55)$$

Comparing the coefficients on the left hand side and on the right hand side in analogy to equation (2.54) leads to a slightly different representation

$$\begin{aligned}
 d_x &= -\frac{1}{\sqrt{2}}(d_+ - d_-) & d_+ &= -\frac{1}{\sqrt{2}}(d_x + id_y) = -d_{-1} \\
 d_y &= \frac{i}{\sqrt{2}}(d_+ + d_-) & d_0 &= d_z \\
 d_z &= d_0 & d_- &= \frac{1}{\sqrt{2}}(d_x - id_y) = -d_1
 \end{aligned} \tag{2.56}$$

Note that the components of the dipole operator \mathbf{d} defined in equation (C.17) and (2.56) are antihermitian conjugate to each other, i.e.

$$\begin{aligned}
 d_{-1}^\dagger &= \frac{1}{\sqrt{2}}(d_x + id_y)^\dagger = \frac{1}{\sqrt{2}}(d_x - id_y) = -d_1 \\
 d_1^\dagger &= -\frac{1}{\sqrt{2}}(d_x - id_y)^\dagger = -\frac{1}{\sqrt{2}}(d_x + id_y) = -d_{-1} \\
 d_+^\dagger &= -\frac{1}{\sqrt{2}}(d_x + id_y)^\dagger = -\frac{1}{\sqrt{2}}(d_x - id_y) = -d_- \\
 d_-^\dagger &= \frac{1}{\sqrt{2}}(d_x - id_y)^\dagger = \frac{1}{\sqrt{2}}(d_x + id_y) = -d_+
 \end{aligned} \tag{2.57}$$

whereas the angular momentum raising and lowering operators are hermitian conjugate to each other i.e. $S_\pm = S_x \pm iS_y$. Equation (2.57) can be simplified to

$$d_q^\dagger = (-1)^q d_{-q} \tag{2.58}$$

2.3.2 Polar Molecules in Static Electric Fields

Now we want to calculate the effect of an external static electrical field \mathbf{E}_{dc} in \mathbf{e}_0 -direction, using the time independent perturbation theory as described in Appendix B and setting $\mathcal{H}_{\text{tot}} \equiv \mathcal{H}^0$ and $\mathcal{H}_{\text{dc}} \equiv \mathcal{H}^1$

$$\mathcal{H} = \mathcal{H}^0 + \mathcal{H}^1 = BJ^2 - d_0 E_{\text{dc}} \quad \mathcal{H}^0 |JM\rangle = BJ(J+1) |JM\rangle \tag{2.59}$$

Zero order perturbations are the solution of the unperturbed Hamiltonian \mathcal{H}^0

$$E_{JM}^{(0)} = BJ(J+1) \quad |\phi_{JM}^{(0)}\rangle = |JM\rangle \tag{2.60}$$

First order perturbation in $\beta \equiv dE_{\text{dc}}/B$ are calculated using formula (B.4) and (B.5)

$$E_{JM}^{(1)} = \langle JM | \mathcal{H}^1 | JM \rangle = -E_{\text{dc}} \langle JM | d_0 | JM \rangle = 0 \tag{2.61}$$

$$\begin{aligned}
 |\phi_{JM}^{(1)}\rangle &= -E_{\text{dc}} \frac{\langle J-1, M | d_0 | JM \rangle}{E_{JM}^{(0)} - E_{J-1, M}^{(0)}} |J-1, M\rangle - E_{\text{dc}} \frac{\langle J+1, M | d_0 | JM \rangle}{E_{JM}^{(0)} - E_{J+1, M}^{(0)}} |J+1, M\rangle \\
 &+ \cdots \underbrace{(J \pm 2, J \pm 3, \dots)}_{=0}, \text{ do not couple to } J, M)
 \end{aligned}$$

$$\begin{aligned}
&= -\frac{E_{\text{dc}}d}{B[J(J+1) - (J-1)J]} \sqrt{\frac{(J-M)(J+M)}{4J^2-1}} |J-1, M\rangle \\
&\quad - \frac{E_{\text{dc}}d}{B[J(J+1) - (J+1)(J+2)]} \sqrt{\frac{(J-M+1)(J+M+1)}{4J(J+2)+3}} |J+1, M\rangle \\
&= -\frac{E_{\text{dc}}d}{2BJ} \sqrt{\frac{J^2-M^2}{4J^2-1}} |J-1, M\rangle - \frac{E_{\text{dc}}d}{2B(-J-1)} \sqrt{\frac{(J+1)^2-M^2}{4(J+1)^2-1}} |J+1, M\rangle \\
&= -\frac{\beta}{2J} \sqrt{\frac{J^2-M^2}{4J^2-1}} |J-1, M\rangle + \frac{\beta}{2(J+1)} \sqrt{\frac{(J+1)^2-M^2}{4(J+1)^2-1}} |J+1, M\rangle \quad (2.62)
\end{aligned}$$

Second order perturbation leads to the perturbed states, using equation (B.6) and (B.7)

$$\begin{aligned}
E_{JM}^{(2)} &= \langle JM | \mathcal{H}^1 | \phi_{JM}^{(1)} \rangle \\
&= -E_{\text{dc}} \left(-\frac{E_{\text{dc}}}{2BJ} \langle J-1, M | d_0 | JM \rangle \right) \langle JM | d_0 | J-1, M \rangle \\
&\quad - E_{\text{dc}} \left(\frac{E_{\text{dc}}}{2B(J+1)} \langle J+1, M | d_0 | JM \rangle \right) \langle JM | d_0 | J+1, M \rangle \\
&\quad + \dots \underbrace{\left(J \pm 2, J \pm 3, \dots \right)}_{=0}, \text{ do not couple to } J, M \\
&= E_{\text{dc}}^2 \frac{|\langle J-1, M | d_0 | JM \rangle|^2}{2BJ} - E_{\text{dc}}^2 \frac{|\langle J+1, M | d_0 | JM \rangle|^2}{2B(J+1)} \\
&= \frac{E_{\text{dc}}^2 d^2}{2BJ} \frac{J^2-M^2}{4J^2-1} - \frac{E_{\text{dc}}^2 d^2}{2B(J+1)} \frac{(J+1)-M^2}{4(J+1)^2-1} \\
&= \frac{B\beta^2}{2} \frac{\left(1 - \frac{3M^2}{J(J+1)}\right)}{(2J-1)(2J+3)} \quad (2.63)
\end{aligned}$$

Now the energy and the states of the perturbed Hamiltonian are calculated to the lowest order of $\beta \equiv dE_{\text{dc}}/B$

$$\begin{aligned}
E_{JM} &= E_{JM}^{(0)} + \beta E_{JM}^{(1)} + \beta^2 E_{JM}^{(2)} \\
&= BJ(J+1) + B \frac{\beta^2}{2} \frac{\left(1 - \frac{3M^2}{J(J+1)}\right)}{(2J-1)(2J+3)} \quad (2.64)
\end{aligned}$$

$$\begin{aligned}
|\phi_{JM}\rangle &= |\phi_{JM}^{(0)}\rangle + \beta |\phi_{JM}^{(1)}\rangle \\
&= |J, M\rangle - \frac{\beta}{2J} \sqrt{\frac{J^2-M^2}{4J^2-1}} |J-1, M\rangle + \frac{\beta}{2(J+1)} \sqrt{\frac{(J+1)^2-M^2}{4(J+1)^2-1}} |J+1, M\rangle \quad (2.65)
\end{aligned}$$

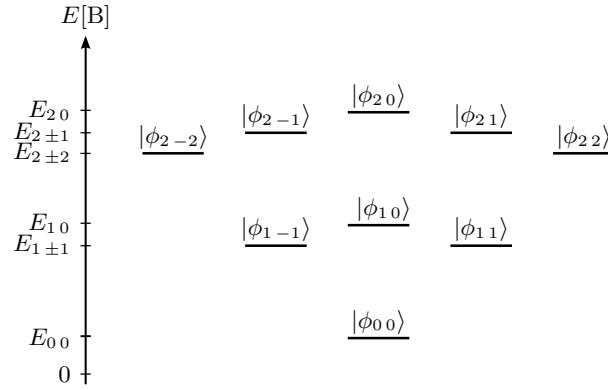


Figure 2.6 Stark shifted states of polar molecules for $J = 0, 1, 2$ and $M = -J, \dots, J$. For further details see [4]

The Stark shifted states – for a static electrical field in z -direction applied to the polar molecules – are now used as basis states for the rotational Hamiltonian $\mathcal{H} = B\mathcal{J}^2 - d_0 E_{\text{dc}}$. Since the static field is applied in z -direction, states with different quantum number M will not be mixed

$$E_{00} \equiv E_{\downarrow} = \frac{\beta^2}{2} \left(-\frac{1}{3} \right) B = -\frac{\beta^2}{6} B \quad (2.66)$$

$$E_{10} \equiv E_{\uparrow} = 2B + B \frac{\beta^2}{2} \frac{1}{5} = 2B + \frac{\beta^2}{10} B$$

$$|\phi_{00}\rangle \equiv |\downarrow\rangle = |0, 0\rangle + \frac{\beta}{2} \frac{1}{\sqrt{3}} |1, 0\rangle \quad (2.67)$$

$$|\phi_{10}\rangle \equiv |\uparrow\rangle = |1, 0\rangle - \frac{\beta}{2} \frac{1}{\sqrt{3}} |0, 0\rangle + \frac{\beta}{2} \frac{1}{\sqrt{15}} |2, 0\rangle$$

These equation can be used to calculate the $\Delta M = 0, \Delta J = 1$ transition with $\Delta E = E_{10} - E_{00} = \hbar\omega$, for example. The $M \neq 0$ degeneracy is not lifted by the stark shift since the electric field vector changes sign under a change of chirality of the coordinate system or a simple spatial inversion. This is true because \mathbf{E} is a genuine vector field whereas the magnetic vector field \mathbf{B} is a axial vector field gaining an additional sign flip when inverted. Put it another way, the time-reversal invariance of the external static electric field perturbs the rotational invariance but at least a two-fold degeneracy must persist because the time reversal operator is antiunitary. Thus the energy E_{11} and E_{1-1} are identical as well as the corresponding states $|\phi_{11}\rangle$ and $|\phi_{1-1}\rangle$

$$E_{1\pm1} = 2B - \frac{\beta^2}{2} B \quad (2.68)$$

$$|\phi_{1\pm1}\rangle = |1, \pm1\rangle + \frac{\beta}{4} \frac{1}{\sqrt{5}} |2, \pm1\rangle \quad (2.69)$$

With the help of the Wigner–Eckart Theorem (A.13) it is very simple to calculate the matrix elements of the dipole operator with respect to the \mathcal{J}^2 eigenbasis. The dipole operator \mathbf{d} in the

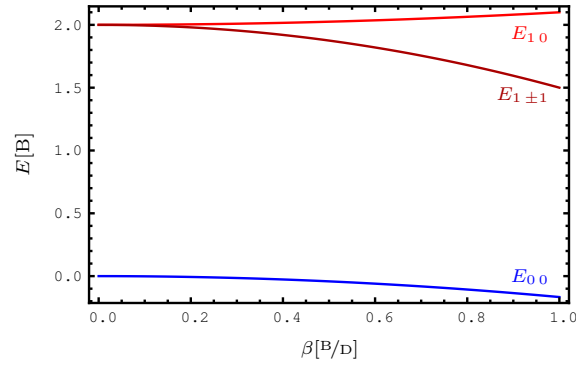


Figure 2.7 Stark shifted energy levels of polar molecules for $J = 0, 1$ and $M = -J, \dots, J$ dependent on the perturbation parameter β .

spherical basis $\{\mathbf{e}_0 \equiv \mathbf{e}_z, \mathbf{e}_{\pm 1} \equiv \mp(\mathbf{e}_x \pm i\mathbf{e}_y)\}$ couples only states with $\Delta J = 1$ and $\Delta M = q$ (see Subsection 2.1.2 “Selection Rules”)

$$\langle JM | \mathbf{d} | JM \rangle = \mathbf{0} \quad (2.70)$$

In the following we choose

$$\langle J \pm 1, M + q | \mathbf{d} | JM \rangle = \begin{cases} \dots & q = -1 \\ \dots & q = 0 \\ \dots & q = 1 \end{cases} \quad (2.71)$$

where \dots indicates the expectation value of the corresponding component as the ordering of the basis vectors. Then we find

$$\langle 1, -1 | \mathbf{d} | 0, 0 \rangle = \begin{pmatrix} \frac{d}{\sqrt{3}} \\ 0 \\ 0 \end{pmatrix} \quad \langle 1, 0 | \mathbf{d} | 0, 0 \rangle = \begin{pmatrix} 0 \\ \frac{d}{\sqrt{3}} \\ 0 \end{pmatrix} \quad \langle 1, 1 | \mathbf{d} | 0, 0 \rangle = \begin{pmatrix} 0 \\ 0 \\ \frac{d}{\sqrt{3}} \end{pmatrix} \quad (2.72)$$

$$\langle 2, -1 | \mathbf{d} | 1, 0 \rangle = \begin{pmatrix} \frac{d}{\sqrt{5}} \\ 0 \\ 0 \end{pmatrix} \quad \langle 2, 0 | \mathbf{d} | 1, 0 \rangle = \begin{pmatrix} 0 \\ \frac{2}{\sqrt{15}}d \\ 0 \end{pmatrix} \quad \langle 2, 1 | \mathbf{d} | 1, 0 \rangle = \begin{pmatrix} 0 \\ 0 \\ \frac{d}{\sqrt{5}} \end{pmatrix} \quad (2.73)$$

For our $\Delta J = 1$ and $\Delta M = 0$ transition we only need to insert two matrix elements in order to calculate the matrix elements of the Hamiltonian in the perturbed two-level system basis with the states $\{|\uparrow\rangle, |\downarrow\rangle\}$

$$\begin{aligned} \mathbf{d}^{\uparrow\uparrow} &\equiv \langle \uparrow | \mathbf{d} | \uparrow \rangle = -2 \frac{\beta}{2} \frac{1}{\sqrt{3}} \underbrace{\langle 1, 0 | \mathbf{d} | 0, 0 \rangle}_{\frac{d}{\sqrt{3}} \mathbf{e}_0} + 2 \frac{\beta}{2} \frac{1}{\sqrt{15}} \underbrace{\langle 1, 0 | \mathbf{d} | 2, 0 \rangle}_{\frac{2d}{\sqrt{15}} \mathbf{e}_0} \\ &= \left(-\frac{\beta}{\sqrt{3}} \frac{d}{\sqrt{3}} + \frac{2\beta}{\sqrt{15}} \frac{d}{\sqrt{15}} \right) \mathbf{e}_0 \end{aligned}$$

$$= \left(-\frac{d\beta}{3} + \frac{2d\beta}{15} \right) \mathbf{e}_0 \quad \hookrightarrow \quad d_0^{\uparrow\uparrow} = -\frac{d\beta}{5} \quad (2.74)$$

$$\begin{aligned} \mathbf{d}^{\uparrow\downarrow} &\equiv \langle \uparrow | \mathbf{d} | \downarrow \rangle = \underbrace{\langle 1, 0 | \mathbf{d} | 0, 0 \rangle}_{\frac{d}{\sqrt{3}} \mathbf{e}_0} - \frac{\beta^2}{4} \frac{1}{3} \underbrace{\langle 1, 0 | \mathbf{d} | 0, 0 \rangle}_{\frac{d}{\sqrt{3}} \mathbf{e}_0} \\ &= \left(\frac{d}{\sqrt{3}} - \frac{d\beta^2}{12\sqrt{3}} \right) \mathbf{e}_0 \quad \hookrightarrow \quad d_0^{\uparrow\downarrow} = \frac{d}{\sqrt{3}} \left(1 - \frac{\beta^2}{12} \right) \end{aligned} \quad (2.75)$$

$$\begin{aligned} \mathbf{d}^{\downarrow\downarrow} &\equiv \langle \downarrow | \mathbf{d} | \downarrow \rangle = 2 \frac{\beta}{2} \frac{1}{\sqrt{3}} \underbrace{\langle 1, 0 | \mathbf{d} | 0, 0 \rangle}_{\frac{d}{\sqrt{3}} \mathbf{e}_0} \\ &= \frac{d\beta}{3} \mathbf{e}_0 \quad \hookrightarrow \quad d_0^{\downarrow\downarrow} = \frac{d\beta}{3} \end{aligned} \quad (2.76)$$

A more generalized discussion on the perturbation of the rigid rotor spectrum of polar molecules can be found in [4].

Numerical Solution

Perturbation theory is only valid for small values of the perturbation parameter, i.e. $\beta \ll 1$. A solution for arbitrary large β can only be obtained numerically. Therefore a numerically diagonalization of the Hamiltonian in equation (2.47) is performed with the help of MATHEMATICA. For further details on the MATHEMATICA code see Appendix H.1 and H.2. Since there is no intermix between states with different M quantum number, it is useful to diagonalize each set of states for a fixed M . This leads to a breaking of the full symmetry, so that one must be cautious not to include matrix elements that are vanishing for symmetry reasons e.g. $\langle \phi_{00}^{\text{num}} | \hat{d}_1 | \phi_{10}^{\text{num}} \rangle$. The dimension of the matrix representation of the Hamiltonian is $\dim \mathcal{H} = 50$, i.e. the highest value for J is 50 and the error is of the order $\beta/J^2 = 10/50^2 \approx 10^{-3}$. The numerically determined eigenenergies E_{JM}^{num} are shown in Figure 2.8.

2.3.3 Polar Molecules in Microwave Fields

The coupling of a static electric field to the molecule leads only to a energy shift in the spectrum of the molecule, the so called dc-Stark shift. In order to couple different rotational states an external microwave field must be applied (see Subsection 2.1.2 “Selection Rules”). There the Hamiltonian (2.47) needs to be extended with an additional term accounting for an external microwave field $\mathbf{E}_{\text{ac}}(t)$

$$\mathcal{H}(t) = \mathcal{H}_{\text{rot}} + \mathcal{H}_{\text{dc}} + \mathcal{H}_{\text{ac}}(t) = B\mathcal{J}^2 - d_0 \mathbf{E}_{\text{dc}} - \mathbf{d} \cdot \mathbf{E}_{\text{ac}}(t) \quad (2.77)$$

The additional term couples the classical external field $\mathbf{E}_{\text{ac}}(t)$ in the dipole approximation. The equivalence of the dipole approximation to the minimal coupling $\mathbf{p} \rightarrow \mathbf{p} + e/c\mathbf{A}$ can be shown via an appropriate local gauge transformation

$$\mathbf{A} \rightarrow \mathbf{A} + \nabla\chi(\mathbf{r}, t) \quad \Phi \rightarrow \Phi - \frac{\partial\chi(\mathbf{r}, t)}{\partial t} \quad (2.78)$$

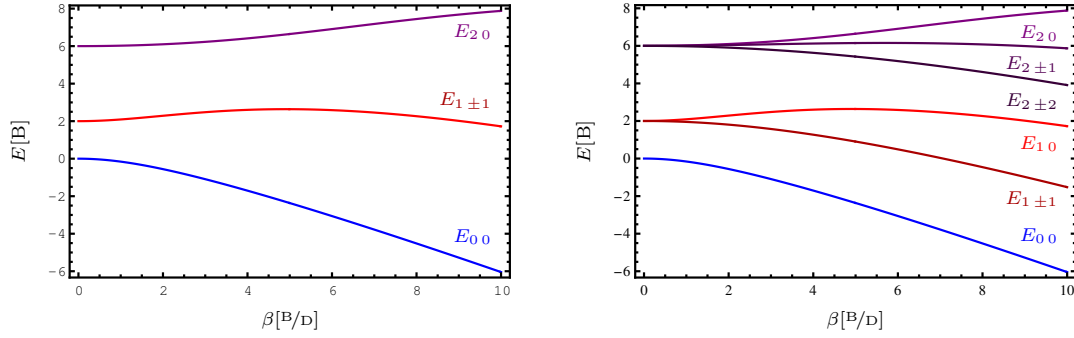


Figure 2.8 Numerically calculated Stark shift of the three lowest energy levels and the lifting of the M degeneracy.

The local gauge transformation $\chi(\mathbf{r}, t) = -\mathbf{r} \cdot \mathbf{A}(\mathbf{0}, t)$ acting on the wave function $\psi(\mathbf{r}, t)$ gives rise to the electric dipole interaction term \mathcal{H}_{ac} . Details about the connection between gauge transformations, gauge invariance of physical quantities and interaction can be found in various quantum optics books [8, 9, 10]. The applied microwave field is described as a electromagnetic wave

$$\mathbf{E}_{\text{ac}}(\mathbf{r}, t) = \sum_{q=-1}^1 (\mathbf{e}_q \cdot \mathbf{E}_{\text{ac}}) \mathbf{e}_q = E_{\text{ac}}(\mathbf{r}) e^{-i\omega_L t} \mathbf{e}_q + (-1)^q E_{\text{ac}}(\mathbf{r}) e^{i\omega_L t} \mathbf{e}_{-q} \quad (2.79)$$

where we consider a single microwave field with polarization q and frequency ω_L . For $q = 0$ the microwave is linear polarized whereas $q \neq 0$ leads to circular polarization. The Hamiltonian \mathcal{H}_{ac} takes the form

$$\mathcal{H}_{\text{ac}}(t) = -\mathbf{d} \cdot \mathbf{E}_{\text{ac}} = -d_q E_{\text{ac}} e^{-i\omega_L t} - (-1)^q E_{\text{ac}} e^{i\omega_L t} d_{-q} \quad (2.80)$$

The Hamiltonian (2.77) will be applied to a two level system consisting of two dc-Stark shifted rotational states of the molecule, e.g. $|\phi_{00}^{\text{num}}\rangle \leftrightarrow |\phi_{10}^{\text{num}}\rangle$. The corresponding time dependent Schrödinger equation can only be solved analytical with the help of the rotating wave approximation (see Subsection 2.3.4 “Rotating Wave Approximation”). The resulting matrix representation of the Hamiltonian (2.77) in the rotating wave approximation can be diagonalized easily and is used as standard description of polar molecules subject to external electrical fields in the following Section 2.4 where interactions between two polar molecules will be considered.

2.3.4 Rotating Wave Approximation

Finally we want to determine the time evolution of polar molecules subject to both external fields \mathbf{E}_{dc} and \mathbf{E}_{ac} . The solution of the time independent part of the Hamiltonian

$$\mathcal{H}^{(0)} = \mathcal{H}_{\text{rot}} + \mathcal{H}_{\text{dc}} \quad (2.81)$$

is already determined in Section 2.3.2 analytically for small static electric fields and numerically for nearly arbitrary large field strengths. Therefore we can assume that the time independent part is diagonal in the basis $\{|\phi_{JM}\rangle\}$ or $\{|\phi_{JM}^{\text{num}}\rangle\}$. Expanding the Hamiltonian $\mathcal{H}^{(0)}$ in this basis,

the solution to the time dependent full Hamiltonian containing the interaction with the external microwave field can be found by solving the corresponding Schrödinger equation in this basis

$$i\hbar \frac{\partial |\psi(t)\rangle}{\partial t} = \mathcal{H}(t) |\psi(t)\rangle = (\mathcal{H}^{(0)} - \mathbf{d} \cdot \mathbf{E}_{ac}(t)) |\psi(t)\rangle \quad (2.82)$$

with the full Hamiltonian from equation (2.77). Expanding $|\psi(t)\rangle$ in the basis $\{|\phi_{JM}\rangle\}$ and considering only the coupling of two Stark-shifted states leads to

$$|\psi(t)\rangle = c_{\uparrow} e^{-\frac{i}{\hbar} E_{\uparrow} t} |\uparrow\rangle + c_{\downarrow} e^{-\frac{i}{\hbar} E_{\downarrow} t} |\downarrow\rangle \quad (2.83)$$

Inserting $|\psi(t)\rangle$ in the Schrödinger equation (2.82) yields

$$\begin{aligned} i\hbar \left(\dot{c}_{\uparrow} e^{-\frac{i}{\hbar} E_{\uparrow} t} |\uparrow\rangle + \dot{c}_{\downarrow} e^{-\frac{i}{\hbar} E_{\downarrow} t} |\downarrow\rangle \right) &= (d_q E_{ac} e^{-i\omega_L t} - (-1)^q E_{ac} e^{i\omega_L t} d_{-q}) c_{\uparrow} e^{-\frac{i}{\hbar} E_{\uparrow} t} |\uparrow\rangle \\ &+ (d_q E_{ac} e^{-i\omega_L t} - (-1)^q E_{ac} e^{i\omega_L t} d_{-q}) c_{\downarrow} e^{-\frac{i}{\hbar} E_{\downarrow} t} |\downarrow\rangle \end{aligned} \quad (2.84)$$

Projecting out the coefficients $c_{\uparrow}(t)$

$$\begin{aligned} i\hbar \dot{c}_{\uparrow} &= - \left(c_{\uparrow} \langle \uparrow | d_q | \uparrow \rangle e^{-\frac{i}{\hbar} E_{\uparrow} t} + c_{\downarrow} \langle \uparrow | d_q | \downarrow \rangle e^{-\frac{i}{\hbar} E_{\downarrow} t} \right) E_{ac} e^{-i\omega_L t} e^{\frac{i}{\hbar} E_{\uparrow} t} \\ &- (-1)^q \left(c_{\uparrow} \langle \uparrow | d_{-q} | \uparrow \rangle e^{-\frac{i}{\hbar} E_{\uparrow} t} + c_{\downarrow} \langle \uparrow | d_{-q} | \downarrow \rangle e^{-\frac{i}{\hbar} E_{\downarrow} t} \right) E_{ac} e^{i\omega_L t} e^{\frac{i}{\hbar} E_{\uparrow} t} \\ &= - \left(c_{\uparrow} \langle \uparrow | d_q | \uparrow \rangle + c_{\downarrow} \langle \uparrow | d_q | \downarrow \rangle e^{\frac{i}{\hbar} (E_{\uparrow} - E_{\downarrow}) t} \right) E_{ac} e^{-i\omega_L t} \\ &- (-1)^q \left(c_{\uparrow} \langle \uparrow | d_{-q} | \uparrow \rangle + c_{\downarrow} \langle \uparrow | d_{-q} | \downarrow \rangle e^{\frac{i}{\hbar} (E_{\uparrow} - E_{\downarrow}) t} \right) E_{ac} e^{-i\omega_L t} \\ \Leftrightarrow \dot{c}_{\uparrow} &= \frac{i}{\hbar} (c_{\uparrow} d_q^{\uparrow\uparrow} + c_{\downarrow} d_q^{\uparrow\downarrow} e^{i\omega t}) E_{ac} e^{-i\omega_L t} + (-1)^q \frac{i}{\hbar} (c_{\uparrow} d_{-q}^{\uparrow\uparrow} + c_{\downarrow} d_{-q}^{\uparrow\downarrow} e^{i\omega t}) E_{ac} e^{i\omega_L t} \end{aligned} \quad (2.85)$$

and $c_{\downarrow}(t)$ analogously, leads to

$$\Leftrightarrow \dot{c}_{\downarrow} = \frac{i}{\hbar} (c_{\uparrow} d_q^{\downarrow\uparrow} e^{-i\omega t} + c_{\downarrow} d_q^{\downarrow\downarrow}) E_{ac} e^{-i\omega_L t} + (-1)^q \frac{i}{\hbar} (c_{\uparrow} d_{-q}^{\downarrow\uparrow} e^{-i\omega t} + c_{\downarrow} d_{-q}^{\downarrow\downarrow}) E_{ac} e^{i\omega_L t} \quad (2.86)$$

Equation (2.85) and (2.86) can be rewritten in a more convenient form

$$\dot{c}_{\uparrow} = \frac{i}{2} \left(\Omega_q^{\uparrow\uparrow} e^{-i\omega_L t} + \Omega_{-q}^{\uparrow\uparrow} e^{i\omega_L t} \right) c_{\uparrow} + \frac{i}{2} \left(\Omega_q^{\uparrow\downarrow} e^{-i\Delta t} + \Omega_{-q}^{\uparrow\downarrow} e^{i(\omega_L + \omega)t} \right) c_{\downarrow} \quad (2.87)$$

$$\dot{c}_{\downarrow} = \frac{i}{2} \left(\Omega_q^{\downarrow\uparrow} e^{-i(\omega_L + \omega)t} + \Omega_{-q}^{\downarrow\uparrow} e^{i\Delta t} \right) c_{\uparrow} + \frac{i}{2} \left(\Omega_q^{\downarrow\downarrow} e^{-i\omega_L t} + \Omega_{-q}^{\downarrow\downarrow} e^{i\omega_L t} \right) c_{\downarrow} \quad (2.88)$$

where $\Delta = \omega_L - \omega$ denotes the detuning of the external microwave field from the transition frequency and Ω_q denotes the Rabi frequency which is defined in terms of the dipole moments as

$$\Omega_q = \frac{2d_q E_{ac}}{\hbar} \quad \Omega_{-q} = (-1)^q \frac{2d_{-q} E_{ac}}{\hbar} \quad (2.89)$$

The additional factor $(-1)^q$ is needed to satisfy the Condon–Shortley phase convention because it is not included in the definition of the (unnormalized) spherical harmonics which is used for spherical tensor operators. In order to apply the rotating wave approximation it is useful to transform into the “rotating frame” of the microwave field via a rotating frame transformation that can be described by

$$\begin{aligned} \mathcal{R} &= \mathcal{R}^{\text{Microwave}} \left(\mathcal{R}^{\text{Molecule}} \right)^{-1} = e^{-i\omega_L \frac{\sigma_z}{2} t} e^{i\omega \frac{\sigma_z}{2} t} = e^{-i(\omega_L - \omega) \frac{\sigma_z}{2} t} \\ &\stackrel{(G.14)}{=} \cos\left(\frac{\Delta}{2}t\right) \mathbb{1} - i \sin\left(\frac{\Delta}{2}t\right) \sigma_z = \begin{pmatrix} e^{-i\frac{\Delta}{2}t} & 0 \\ 0 & e^{i\frac{\Delta}{2}t} \end{pmatrix} \end{aligned} \quad (2.90)$$

Transforming the components of $|\psi(t)\rangle$ into the rotating frame is done with the inverse transformation \mathcal{R}^{-1} yielding

$$c_\uparrow = e^{-i\frac{\Delta}{2}t} \tilde{c}_\uparrow \quad c_\downarrow = e^{i\frac{\Delta}{2}t} \tilde{c}_\downarrow \quad (2.91)$$

where “ \sim ” denotes the coefficients in the rotating frame. Inserting the transformed coefficients into (2.87) gives rise to

$$\begin{aligned} \dot{\tilde{c}}_\uparrow e^{-i\frac{\Delta}{2}t} - i\frac{\Delta}{2}\tilde{c}_\uparrow e^{-i\frac{\Delta}{2}t} &= \frac{i}{2} \left(\Omega_q^{\uparrow\uparrow} e^{-i\omega_L t} + \Omega_{-q}^{\uparrow\uparrow} e^{i\omega_L t} \right) e^{-i\frac{\Delta}{2}t} \tilde{c}_\uparrow \\ &\quad + \frac{i}{2} \left(\Omega_q^{\uparrow\downarrow} e^{-i\Delta t} + \Omega_{-q}^{\uparrow\downarrow} e^{i(\omega_L + \omega)t} \right) e^{i\frac{\Delta}{2}t} \tilde{c}_\downarrow \\ \Leftrightarrow \dot{\tilde{c}}_\uparrow &= \frac{i}{2} \left(\Delta + \Omega_q^{\uparrow\uparrow} e^{-i\omega_L t} + \Omega_{-q}^{\uparrow\uparrow} e^{i\omega_L t} \right) \tilde{c}_\uparrow + \frac{i}{2} \left(\Omega_q^{\uparrow\downarrow} + \Omega_{-q}^{\uparrow\downarrow} e^{i\Delta t} e^{i(\omega_L + \omega)t} \right) \tilde{c}_\downarrow \end{aligned} \quad (2.92)$$

and analogously equation (2.88) reads

$$\dot{\tilde{c}}_\downarrow = \frac{i}{2} \left(\Omega_q^{\downarrow\uparrow} e^{-i\Delta t} e^{-i(\omega_L + \omega)t} + \Omega_{-q}^{\downarrow\uparrow} \right) \tilde{c}_\uparrow + \frac{i}{2} \left(-\Delta + \Omega_q^{\downarrow\downarrow} e^{-i\omega_L t} + \Omega_{-q}^{\downarrow\downarrow} e^{i\omega_L t} \right) \tilde{c}_\downarrow \quad (2.93)$$

A further simplification can be done by eliminating $\Omega_{-q}^{\downarrow\uparrow}$, $\Omega_{-q}^{\uparrow\downarrow}$, $\Omega_{-q}^{\downarrow\downarrow}$ and $\Omega_{-q}^{\uparrow\uparrow}$. Since the matrix elements of the dipole operator are real, it follows that

$$\left(\Omega_{-q}^{\uparrow\uparrow} \right)^* \propto (-1)^q \langle \uparrow | \mathbf{d}_{-q} | \uparrow \rangle^* = (-1)^q \langle \uparrow | \mathbf{d}_{-q}^\dagger | \uparrow \rangle \stackrel{(2.58)}{=} (-1)^{2q} \langle \uparrow | \mathbf{d}_q | \uparrow \rangle \propto \Omega_q^{\uparrow\uparrow} \quad (2.94)$$

$$\left(\Omega_{-q}^{\downarrow\uparrow} \right)^* \propto (-1)^q \langle \downarrow | \mathbf{d}_{-q} | \uparrow \rangle^* = (-1)^q \langle \uparrow | \mathbf{d}_{-q}^\dagger | \downarrow \rangle \stackrel{(2.58)}{=} (-1)^{2q} \langle \uparrow | \mathbf{d}_q | \downarrow \rangle \propto \Omega_q^{\uparrow\downarrow} \quad (2.95)$$

and analogously one finds $\Omega_{-q}^{\downarrow\downarrow} = \Omega_q^{\downarrow\downarrow}$ and $\Omega_{-q}^{\uparrow\downarrow} = \Omega_q^{\uparrow\downarrow}$. Thus equation (2.92) and (2.93) can be simplified to

$$\dot{\tilde{c}}_\uparrow = \frac{i}{2} \left(\Delta + \Omega_q^{\uparrow\uparrow} e^{-i\omega_L t} + \Omega_q^{\uparrow\uparrow} e^{i\omega_L t} \right) \tilde{c}_\uparrow + \frac{i}{2} \left(\Omega_q^{\uparrow\downarrow} + \Omega_q^{\uparrow\downarrow} e^{i\Delta t} e^{i(\omega_L + \omega)t} \right) \tilde{c}_\downarrow \quad (2.96)$$

$$\dot{\tilde{c}}_\downarrow = \frac{i}{2} \left(\Omega_q^{\downarrow\uparrow} e^{-i\Delta t} e^{-i(\omega_L + \omega)t} + \Omega_q^{\downarrow\uparrow} \right) \tilde{c}_\uparrow + \frac{i}{2} \left(-\Delta + \Omega_q^{\downarrow\downarrow} e^{-i\omega_L t} + \Omega_q^{\downarrow\downarrow} e^{i\omega_L t} \right) \tilde{c}_\downarrow \quad (2.97)$$

Emission of a photon	Emission & activation of the molecule	Absorption & deactivation of the molecule	Absorption of a photon
$e^{i\omega_L t}$	$e^{i(\omega_L + \omega)t}$	$e^{-i(\omega_L + \omega)t}$	$e^{-i\omega_L t}$
$e^{i(\Delta + \omega)t}$	$e^{i(\Delta + 2\omega)t}$	$e^{-i(\Delta + 2\omega)t}$	$e^{-i(\Delta + \omega)t}$

Table 2.2 Expanding the time dependent terms of the wave function $|\psi(t)\rangle$ in a Fourier series would lead to higher terms in the frequency ω_L . The rotating wave approximation leads to a vanishing of these terms, which is required for energy conservation. In the second row it is shown that some of the processes are actually two-photon processes that are very unlikely to happen. Further details on the RWA can be found in [8, 11].

Now we are able to apply the rotating wave approximation. In this approximation we will neglect all fast rotating terms, i.e. terms with exponential $i\omega_L t$, $i(\omega_L + \omega)t$ and their complex conjugates. These terms are rapidly oscillating and their contribution averages to zero on reasonable time scales. Since we also employed the dipole approximation, that is the external microwave field must be near resonance with the transition, those terms with exponential $i(\Delta + n\omega)t$ with n being a integer, must vanish in order to keep the dipole approximation valid. Therefore, the rotating wave approximation accounts for a time average of the expectation values and enforces energy conservation. A more physical insight to this approximation is listed in Table 2.2. Thus after applying the rotating wave approximation (in the following abbreviated with “RWA”) our differential equation system can be written in the form of a Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \tilde{c}_\uparrow \\ \tilde{c}_\downarrow \end{pmatrix} = -\frac{\hbar}{2} \begin{pmatrix} \Delta & \Omega_q^{\uparrow\downarrow} \\ \Omega_q^{\uparrow\downarrow} & -\Delta \end{pmatrix} \begin{pmatrix} \tilde{c}_\uparrow \\ \tilde{c}_\downarrow \end{pmatrix} \quad (2.98)$$

In spin- $1/2$ language the RWA-Hamilton can be represented in the following form

$$\mathcal{H}^{\text{RWA}} = -\frac{\hbar\Delta}{2}\sigma_z - \frac{\hbar\Omega_q^{\uparrow\downarrow}}{2}\sigma_x + E_\downarrow\mathbb{1} \quad (2.99)$$

The rotating wave approximation has eliminated the time dependency and therefore the Schrödinger equation (2.98) is easy to solve. In fact the solution has already been derived in 2.2. Comparing the two Hamiltonians \mathcal{H}^{RWA} and \mathcal{H} from equation (2.25) ignoring the global minus sign

$$\mathcal{H} = \begin{pmatrix} E_\uparrow & |H'_{12}| e^{-i\arg H'_{12}} \\ |H'_{12}| e^{-i\arg H'_{12}} & E_\downarrow \end{pmatrix} \iff \mathcal{H}^{\text{RWA}} = \begin{pmatrix} \frac{\hbar\Delta}{2} & \frac{\hbar\Omega_q^{\uparrow\downarrow}}{2} \\ \frac{\hbar\Omega_q^{\uparrow\downarrow}}{2} & -\frac{\hbar\Delta}{2} \end{pmatrix} \quad (2.100)$$

leads to the replacements shown in Table 2.3. With the help of these replacements the solution is given in the form of

$$\tilde{c}_\uparrow = \left[\cos\left(\frac{\Omega}{2}\right) + i\frac{\Delta}{\Omega} \sin\left(\frac{\Omega}{2}\right) \right] \tilde{c}_\uparrow(0) + i\frac{\Omega_q^{\uparrow\downarrow}}{\Omega} \sin\left(\frac{\Omega}{2}\right) \tilde{c}_\downarrow(0) \quad (2.101)$$

$$\tilde{c}_\downarrow = i\frac{\Omega_q^{\uparrow\downarrow}}{\Omega} \sin\left(\frac{\Omega}{2}\right) \tilde{c}_\uparrow(0) + \left[\cos\left(\frac{\Omega}{2}\right) - i\frac{\Delta}{\Omega} \sin\left(\frac{\Omega}{2}\right) \right] \tilde{c}_\downarrow(0) \quad (2.102)$$

Operator	\mathcal{H}	\mathcal{H}^{RWA}
non-diagonalized	$E_0 = \frac{E_{\uparrow} - E_{\downarrow}}{2}$ $\hbar\omega = E_{\uparrow} + E_{\downarrow}$	$E_0 = \frac{1}{2} \left(\frac{\hbar\Delta}{2} - \frac{\hbar\Delta}{2} \right) = 0$ $\hbar\omega = \frac{\hbar\Delta}{2} - \left(-\frac{\hbar\Delta}{2} \right) = \hbar\Delta$
diagonalized	$E_{\pm} = E_0 \pm \frac{\hbar\Omega}{2}$ $\hbar\Omega = \sqrt{(\hbar\omega)^2 + 4 H'_{12} ^2}$	$E_{\pm} = \pm \frac{\hbar\Omega}{2}$ $\hbar\Omega = \sqrt{(\hbar\Delta)^2 + 4 \left(\frac{\hbar\Omega_q^{\uparrow\downarrow}}{2} \right)^2}$

Table 2.3 Eigenenergies of the diagonalized \mathcal{H}^{RWA} and generalized “Rabi” frequency are found by comparison with the solution from (2.25) and by setting $E_{\uparrow\downarrow} = \pm\hbar\Delta/2$, $|H'_{12}| = \hbar\Omega_q^{\uparrow\downarrow}/2$. Due to the fact that the dipole matrix elements are real γ is equal to zero.

The frequency Ω is called the generalized “Rabi” frequency and given according to Table 2.3 as

$$\Omega = \sqrt{\Delta^2 + \left(\Omega_q^{\uparrow\downarrow}\right)^2} = \sqrt{(\omega_L - \omega)^2 + 4/\hbar^2 (E_{\text{ac}} \langle \uparrow | \mathbf{d}_q | \downarrow \rangle)^2} \quad (2.103)$$

where the expressions for the detuning Δ and the Rabi frequency of the transition $\Omega_q^{\uparrow\downarrow}$ are inserted. The solution in the eigenbasis of

$$\mathcal{H}^{\text{RWA}} = \frac{\hbar}{2} \begin{pmatrix} -\Omega & 0 \\ 0 & -\Omega \end{pmatrix} + E_{\downarrow} \mathbf{1} \quad (2.104)$$

is then given in the form of

$$|\uparrow\rangle = \cos \left[\frac{1}{2} \arctan \left(\frac{\Omega}{\Delta} \right) \right] |+\rangle + \sin \left[\frac{1}{2} \arctan \left(\frac{\Omega}{\Delta} \right) \right] |-\rangle \quad (2.105)$$

$$|\downarrow\rangle = -\sin \left[\frac{1}{2} \arctan \left(\frac{\Omega}{\Delta} \right) \right] |+\rangle + \cos \left[\frac{1}{2} \arctan \left(\frac{\Omega}{\Delta} \right) \right] |-\rangle \quad (2.106)$$

As has been pointed out in Section 2.2 the perturbation of the two level system leads to an energy shift. In this case the shift is induced by the external microwave field \mathbf{E}_{ac} leading to an additional Stark shift, the so called ac-Stark shift. With the help of the expressions in Table 2.3 the induced ac-Stark shift can be calculated by subtracting the overall ground state energy of the system E_{\downarrow}

$$\begin{aligned}
E_{\pm} - E_{\downarrow} &= -\frac{\hbar\Delta}{2} \pm \frac{\hbar\Omega}{2} = -\frac{\hbar\Delta}{2} \pm \frac{\hbar\Delta}{2} \sqrt{1 + \left(\frac{\Omega_q^{\uparrow\downarrow}}{\Delta} \right)^2} \\
&\approx -\frac{\hbar\Delta}{2} \pm \frac{\hbar\Delta}{2} \left[1 + \frac{1}{2} \left(\frac{\Omega_q^{\uparrow\downarrow}}{\Delta} \right)^2 \right] = -\frac{\hbar\Delta}{2} \pm \frac{\hbar\Delta}{2} + \frac{\hbar(\Omega_q^{\uparrow\downarrow})^2}{4\Delta}
\end{aligned} \quad (2.107)$$

In the dipole approximation (external microwave field near resonance $\Delta \ll \Omega_q^{\uparrow\downarrow}$) the square root can be approximated with the series expansion up to linear order, i.e. $\sqrt{1+x} \approx 1+x/2$. Inserting

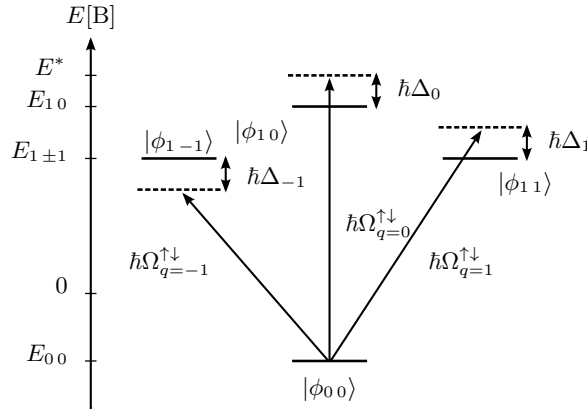


Figure 2.9 Coupling of the dc-Stark shifted states by the external microwave field with polarization q . Linear polarized microwaves with $q = 0$ couple states with $\Delta M = 0$ whereas circular polarized microwaves can couple only states with $\Delta M \neq 0$ (see Subsection 2.1.2 “Selection Rules”). The energy E_{JM} and the states $|\phi_{JM}\rangle$ can be analytically determined via perturbation theory for small static fields or numerically for arbitrary large field strength (see Subsection 2.3.2 “Polar Molecules in Static Electric Fields”).

$\Omega_q^{\uparrow\downarrow} = 2/\hbar E_{ac} d_q^{\uparrow\downarrow}$ we finally arrive at

$$E_+ - E_{\downarrow} = \frac{(E_{ac} d_q^{\uparrow\downarrow})^2}{\hbar\Delta} \quad (2.108)$$

$$E_- - E_{\downarrow} = -\hbar\Delta + \frac{(E_{ac} d_q^{\uparrow\downarrow})^2}{\hbar\Delta} \quad (2.109)$$

The ac-Stark shift depends linear on the detuning Δ and quadratic on the field strength of the external microwave field E_{ac} and the dipole moment of the polar molecule $d_q^{\uparrow\downarrow}$, respectively.

2.4 Interactions between Polar Molecules

2.4.1 Dipole–Dipole Interaction

The interaction between two polar molecules can be derived from the classical model of the electrostatic potential

$$\Phi(\mathbf{r}) = \int_V d^3 r' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \quad (2.110)$$

For well separated molecules, we can expand $\frac{1}{|\mathbf{r} - \mathbf{r}'|}$ in \mathbf{r}' because the molecules are in their mutual far field, fulfilling $r \gg r'$

$$\begin{aligned} \frac{1}{|\mathbf{r} - \mathbf{r}'|} &= \sum_{k=0}^{\infty} \frac{1}{k!} (-\mathbf{r}' \cdot \nabla) \frac{1}{r} \\ &= \frac{1}{r} - \frac{\mathbf{r} \cdot \mathbf{r}'}{r^3} + \frac{\mathbf{r} \otimes \mathbf{r}}{2r^5} : (\mathbf{3}\mathbf{r}' \otimes \mathbf{r}' - r'^2 \mathbf{1}) + \dots \end{aligned} \quad (2.111)$$

Inserting this series expansion into the electrostatic potential gives rise to the multipole expansion

$$\begin{aligned}\Phi(\mathbf{r}) &= \frac{1}{r} \underbrace{\int_V d^3 r' \rho(\mathbf{r}')}_Q + \frac{r}{r^3} \cdot \underbrace{\int_V d^3 r' \mathbf{r}' \rho(\mathbf{r}')}_\mathbf{d} + \frac{1}{2} \frac{\mathbf{r} \otimes \mathbf{r}}{r^5} : \underbrace{\int_V d^3 r' (3\mathbf{r}' \otimes \mathbf{r}' - r'^2 \mathbb{1}) \rho(\mathbf{r}')}_q + \dots \\ &= \frac{1}{r} Q + \frac{\mathbf{r}}{r^3} \cdot \mathbf{d} + \frac{1}{2} \frac{\mathbf{r} \otimes \mathbf{r}}{r^5} : q + \dots\end{aligned}\quad (2.112)$$

where the definitions of the monopole Q , the dipole d and the quadrupole q have been inserted. Considering two molecules 1 and 2 separated by the distance R , their potential energy due to their charge distribution is given by equation (2.45) as follows

$$V_{12}(\mathbf{R}) = Q_1 \phi_2(\mathbf{R}) + \mathbf{d}_1 \cdot \nabla \Phi_2(\mathbf{R}) + \frac{1}{2} q_1 : \nabla(\nabla \Phi_2(\mathbf{R})) + \dots \quad (2.113)$$

Since the two molecules are identical, there is no difference if we would have acted with the electrostatic potential of molecule 1 on the charge distribution of molecule 2. In fact all terms in (2.113) are symmetrical under a transposition of the subscripts 1 and 2. For a neutral polar molecule, i.e. $Q = 0$ the leading term is the dipole term and because of the small spatial extension of the molecules higher multipoles can be neglected. This is the well known dipole approximation we have employed earlier. Thus for two polar molecules with permanent dipole moment \mathbf{d} the interaction is reduced to

$$\begin{aligned}V^{\text{dd}}(\mathbf{R}) &= \mathbf{d}_1 \cdot \nabla \Phi_2(\mathbf{R}) = \mathbf{d}_1 \cdot \nabla \left(\frac{\mathbf{r} \cdot \mathbf{d}_2}{r^3} \right) \\ &= \mathbf{d}_1 \cdot \left(\nabla(\mathbf{R} \cdot \mathbf{d}_2) \frac{1}{R^3} + \nabla \left(\frac{1}{R^3} \right) \mathbf{R} \cdot \mathbf{d}_2 \right) \\ &= \mathbf{d}_1 \cdot \left[\frac{1}{R^3} \left(\underbrace{(\nabla \otimes \mathbf{R})}_{=\mathbb{1}} \mathbf{d}_2 + \underbrace{(\nabla \otimes \mathbf{d}_2)}_{=0} \mathbf{R} \right) - \frac{3\mathbf{R}}{R^5} (\mathbf{R} \cdot \mathbf{d}_2) \right] \\ &= \frac{\mathbf{d}_1 \cdot \mathbf{d}_2}{R^3} - \frac{3}{R^5} (\mathbf{d}_1 \cdot \mathbf{R}) (\mathbf{R} \cdot \mathbf{d}_2) \\ \hookrightarrow V^{\text{dd}}(\mathbf{R}) &= \frac{\mathbf{d}_1 \cdot \mathbf{d}_2 - 3(\mathbf{d}_1 \cdot \mathbf{e}_R)(\mathbf{e}_R \cdot \mathbf{d}_2)}{R^3}\end{aligned}\quad (2.114)$$

The scalar product of two dipole operators can be calculated following equation (D.11)

$$\mathbf{d}_1 \cdot \mathbf{d}_2 = (\mathbf{d} \otimes \mathbb{1})(\mathbb{1} \otimes \mathbf{d}) = d_x \otimes d_x + d_y \otimes d_y + d_z \otimes d_z \quad (2.115)$$

and can be written in terms of the spherical basis as

$$\begin{aligned}\mathbf{d}_1 \cdot \mathbf{d}_2 &= \frac{1}{2} (d_1 - d_{-1}) \otimes (d_1 - d_{-1}) - \frac{1}{2} (d_1 + d_{-1}) \otimes (d_1 + d_{-1}) + d_0 \otimes d_0 \\ &= d_0 \otimes d_0 - d_1 \otimes d_{-1} - d_{-1} \otimes d_1\end{aligned}\quad (2.116)$$

The calculation of the second term $(\mathbf{d}_1 \cdot \mathbf{e}_R)(\mathbf{e}_R \cdot \mathbf{d}_2)$ proceed in exactly the same way but the calculation is a little bit lengthy and rather tedious. A better way is to represent \mathcal{V}^{dd} directly with the help of the unnormalized spherical harmonics using equation (2.49) and a generalization thereof for higher tensor operators. Since \mathcal{V}^{dd} is a operator acting on the direct product space of two particles (or two polar molecules), it is actually a tensor operator of rank two and its expansion in spherical coordinates is given by

$$\mathcal{V}^{\text{dd}} = -\frac{1}{R^3} \sum_{q=-2}^2 C_q^{(2)}(\theta, \phi) [\mathbf{d} \otimes \mathbf{d}]_q^2 \quad (2.117)$$

Taking unnormalized spherical harmonics $C_q^{(2)}$ from Table A.1 and inserting them into

$$\begin{aligned} \mathcal{V}^{\text{dd}} = & -\frac{1}{R^3} \left[C_{-2}^{(2)}(\mathbf{d}_{-1} \otimes \mathbf{d}_{-1}) - C_{-1}^{(2)}(\mathbf{d}_{-1} \otimes \mathbf{d}_0 + \mathbf{d}_0 \otimes \mathbf{d}_{-1}) \right. \\ & + C_0^{(2)}(\mathbf{d}_1 \otimes \mathbf{d}_{-1} + \mathbf{d}_{-1} \otimes \mathbf{d}_1 + 2\mathbf{d}_0 \otimes \mathbf{d}_0) \\ & \left. - C_1^{(2)}(\mathbf{d}_1 \otimes \mathbf{d}_0 + \mathbf{d}_0 \otimes \mathbf{d}_1) + C_2^{(2)}(\mathbf{d}_1 \otimes \mathbf{d}_1) \right] \end{aligned} \quad (2.118)$$

gives rise to

$$\begin{aligned} \mathcal{V}^{\text{dd}} = & -\frac{1}{R^3} \left\{ (3 \cos^2 \theta - 1) \mathbf{d}_0 \otimes \mathbf{d}_0 + \frac{3}{2} \sin^2 \theta (e^{2i\phi} \mathbf{d}_1 \otimes \mathbf{d}_1 + \mathbf{d}_{-1} \otimes \mathbf{d}_{-1}) \right. \\ & + 3 \cos \theta \sin \theta \left[e^{i\phi} (\mathbf{d}_1 \otimes \mathbf{d}_0 + \mathbf{d}_0 \otimes \mathbf{d}_1) - e^{-i\phi} (\mathbf{d}_{-1} \otimes \mathbf{d}_0 + \mathbf{d}_0 \otimes \mathbf{d}_{-1}) \right] \\ & \left. + \frac{1}{2} (3 \cos^2 \theta - 1) (\mathbf{d}_1 \otimes \mathbf{d}_{-1} + \mathbf{d}_{-1} \otimes \mathbf{d}_1) \right\} \end{aligned} \quad (2.119)$$

where θ describes the polar angle of \mathbf{R} with respect to the fixed Cartesian coordinate system and ϕ the azimuthal angle, respectively. In Chapter 3 we will encounter a setup of polar molecules in a two dimensional optical lattice, subject to an external static electric field in z -direction. Therefore we can assume that $\theta = \pi/2$, taking the xy -plane as our two dimensional plane where the polar molecules – aligned along the z -axis – reside

$$\mathcal{V}^{\text{dd}} = \frac{2\mathbf{d}_0 \otimes \mathbf{d}_0 - (3 e^{2i\phi} \mathbf{d}_1 \otimes \mathbf{d}_1 - \mathbf{d}_1 \otimes \mathbf{d}_{-1} - \mathbf{d}_{-1} \otimes \mathbf{d}_1 + 3 e^{-2i\phi} \mathbf{d}_{-1} \otimes \mathbf{d}_{-1})}{2r^3} \quad (2.120)$$

Here ϕ denotes the in-plane angle measured from the x -axis. Due to the external microwave field, the polar molecules will rotate about a rotation axis in the xy -plane and therefore the dipole moment can be considered to be aligned along the x -direction. Finally our two particle interaction operator for this setup can be cast into

$$\mathcal{V}^{\text{dd}} = \frac{\mathbf{d}_0 \otimes \mathbf{d}_0 - \frac{3}{2}(\mathbf{d}_1 \otimes \mathbf{d}_1 + \mathbf{d}_{-1} \otimes \mathbf{d}_{-1}) + \frac{1}{2}(\mathbf{d}_1 \otimes \mathbf{d}_{-1} + \mathbf{d}_{-1} \otimes \mathbf{d}_1)}{r^3} \quad (2.121)$$

The optical square lattice described in Chapter 3 will be filled with $\sqrt{N} \times \sqrt{N}$ polar molecules, where N denotes the total number of molecules in the system. In general each molecule in the optical lattice can interact with all other molecules. Therefore the two particle operator \mathcal{V}^{dd} will be generalized to

$$\mathcal{V}_{ij}^{\text{dd}} = \frac{\mathbf{d}_{i0} \mathbf{d}_{j0} - \frac{3}{2}(\mathbf{d}_{i1} \mathbf{d}_{j1} + \mathbf{d}_{i-1} \mathbf{d}_{j-1}) + \frac{1}{2}(\mathbf{d}_{i1} \mathbf{d}_{j-1} + \mathbf{d}_{i-1} \mathbf{d}_{j1})}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \quad (2.122)$$

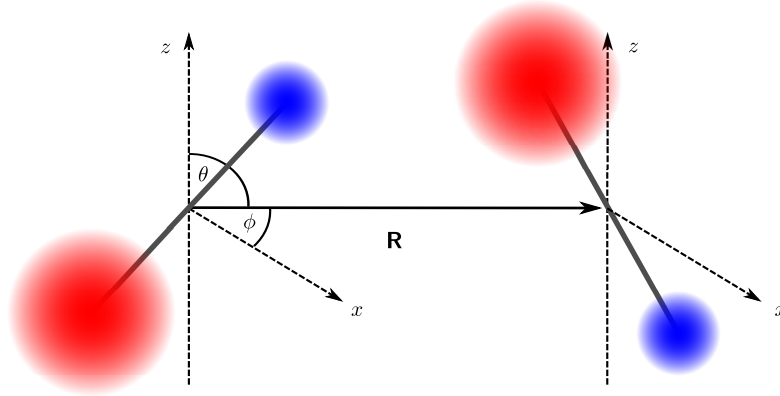


Figure 2.10 Dipole–dipole interaction of two polar molecules. The projections $\mathbf{e}_R \cdot \mathbf{d}_i$ define the polar angle θ_i and the azimuthal angle ϕ_i with respect to the space fixed frame centered at the molecules.

where a denotes the lattice spacing and \mathbf{R}_i is a dimensionless vector pointing on the i th lattice point.

2.4.2 Van–der–Waals Interaction

Without the external fields \mathbf{E}_{dc} and \mathbf{E}_{ac} the only interaction between two molecules in their rotational ground state is the van–der–Waals interaction which is a combination of the Coulomb interaction and quantum fluctuations. Suppose in one of the two molecules arises a dipole moment due to the fluctuation in the charge distribution. This gives rise to an electric field acting on the other molecule, according to equation (2.112) taking only the dipole term

$$\mathbf{E}_2 = -\nabla\Phi_1^{\text{induced}} = \frac{1}{R^3} \left(3\mathbf{e}_R(\mathbf{e}_R \cdot \mathbf{d}_1^{\text{induced}}) - \mathbf{d}_1^{\text{induced}} \right) \quad (2.123)$$

This electric field induces a dipole moment in the molecule 2

$$\mathbf{d}_2^{\text{induced}} = \alpha_2 \mathbf{E}_2 \quad (2.124)$$

where α denotes the polarizability tensor describing the linear response of the charge distribution to an external electric field. This dipole in turn acts on molecule 1 leading to a polarization induced dipole moment, so that we can write for the interaction

$$V^{\text{vdW}} = -\mathbf{d}_1^{\text{induced}} \cdot \mathbf{E}_1 = -\mathbf{d}_2^{\text{induced}} \cdot \mathbf{E}_2 \quad (2.125)$$

Since the two molecules are identical the interaction must be identical under a transposition of the subscripts 1 and 2. Physically speaking, we cannot distinguish which of the fluctuation in the charge distribution of the two molecules is starting the whole process. Finally for identical molecules we end up with the fluctuating dipole–dipole interaction

$$V^{\text{vdW}} = -\mathbf{d}_2^{\text{induced}} \cdot \mathbf{E}_2 \stackrel{(2.124)}{=} -\alpha_2 \mathbf{E}_2 \cdot \mathbf{E}_2 \stackrel{(2.123)}{=} -\frac{9(\mathbf{e}_R \cdot \alpha_1 \mathbf{E}_1)(\mathbf{e}_R \cdot \alpha_2 \mathbf{E}_2) + \dots}{R^6} \approx -\frac{C_6}{R^6} \quad (2.126)$$

The two equations for molecule 1 and molecule 2 can be inserted iteratively with the leading term being proportional to $-1/R^6$ and for isotropic polarizability to the product of $\alpha_1 \alpha_2$. The actual coefficient can be derived quantum mechanically with the help of second order perturbation

theory. The results of such a calculation will be more accurate because using the charge density expressed in terms of electronic wave functions provides a much better charge distribution. Additionally the perturbation of these wave functions up to second order respects the quantum nature of the fluctuations.

3

Realization of Heisenberg Models

Starting this chapter with a very short introduction of the Heisenberg model and its relative the XXZ model, we will then look in particular on the spin- $1/2$ XXZ model and its realization with cold polar molecules in optical lattices. In Section 3.2 the spin- $1/2$ XXZ model is derived microscopically with the help of the rotating wave approximation for polar molecules subject to external electric fields. In a next step the dispersion relation of harmonic excitations above the mean field ground state – the so called spin waves – are determined. The mean field ground state – the simplest possible solution of this model – will be presented in Section 3.4 by applying the mean field theory to determine the ground state in dependence of the tuning of the external fields. Finally the mean field ground state and the spin wave analysis enables us to identify the phase diagram of the spin- $1/2$ XXZ model.

3.1 Heisenberg Models of Quantum Magnets

The Heisenberg model is maybe the simplest model of magnetism but accounts for a wide range of physical phenomena. For example it describes coupled electronic spins in metals and a possible and mostly dominant coupling mechanism is the exchange interaction, i.e. an interplay of the Pauli exclusion principle and the Coulomb repulsion. These purely electronic interactions are much stronger than the magnetic interaction due to a magnetic dipole moment induced by the electron spins. Considering only the coupling of nearest neighbor spins the Heisenberg model is defined as

$$\mathcal{H} = - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mathbf{h}_i \cdot \mathbf{S}_i \quad (3.1)$$

where \mathbf{h} is an external magnetic field. If the exchange integral J_{ij} is positive the Heisenberg model describes a ferromagnet, where all spins are aligned parallel. This can be achieved in a first approximation by considering only the Coulomb interaction where the exchange integral J_{ij} is always positive. An antiferromagnet is Néel ordered that is an alternating sequence of spins pointing up and down, such that all neighbors are antiparallel aligned. In practice the J_{ij} are often introduced as phenomenological parameters. Since we are not interested in the real

physical mechanism of generating the coupling of spins we will set $J_{ij} = -J$. In the following section we will derive the coupling constants microscopically from the dipole–dipole interactions of cold polar molecules in optical lattices.

There are several models connected with the Heisenberg model. First of all the Heisenberg model can be treated classically, i.e. $S = \infty$ with a continuous spectrum of states. Quantum mechanically there exists Heisenberg model for arbitrary spins describing $2S + 1$ discrete states. Especially the spin- $1/2$ model with two degrees of freedom are of interest in several fields of physics. The Heisenberg model can be generalized to the spin- $1/2$ XXZ model

$$\mathcal{H}^{\text{XXZ}} = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left[J_{\perp} \left(S_{ix}S_{jx} + S_{iy}S_{jy} \right) + J_z S_{iz}S_{jz} \right] - \sum_{i=1}^N \mathbf{h}_i \cdot \mathbf{S}_i \quad (3.2)$$

with two different coupling strengths, the in-plane coupling J_{\perp} and the in-axis coupling J_z along the quantization axis, i.e. typically the z -axis. For strong in-plane interaction $J_{\perp} \gg J_z$ we can neglect the $S_{iz}S_{jz}$ term. This model is called the XY model which favors a strong exchange anisotropy. In contrast for strong in-axis interaction $J_z \gg J_{\perp}$ we will arrive at the Ising model. Classically, all these models can be classified as \mathbf{n} -vector models with $n = 1$ being the Ising model, $n = 2$ being the XY model and finally $n = 3$ the Heisenberg model. Quantum mechanically we cannot describe the spin by a classical vector since we have to deal with representatives of the $SU(2)$. In the following we will consider only the spin- $1/2$ Heisenberg models possessing the $SU(2)$ symmetry at the Heisenberg point $J_z = J_{\perp}$. Further information about models to describe (quantum) magnetism can be found in [12, 13]

3.2 Realizing the Heisenberg Model with Polar Molecules

In order to realize the spin- $1/2$ XXZ model with cold polar molecules we take the following approach. First a static electric field \mathbf{E}_{dc} is applied, creating a dipole moment and additionally inducing a dc-Stark shift of the rotational spectrum. Next an additional external microwave field with polarization q , field strength E_{ac} and frequency ω_{L} is irradiated to couple $|\phi_{00}\rangle$ and $|\phi_{10}\rangle$ of the dc-Stark shifted rotational spectrum. We will restrict ourselves only on near resonance dipole transitions with very small detuning Δ . For the $|\phi_{00}\rangle \leftrightarrow |\phi_{10}\rangle$ coupling, the microwave field needs to be linear polarized, i.e. $q = 0$. The respective Hamiltonian is given in the rotating wave approximation according to equation (2.99) as

$$\mathcal{H}^{\text{RWA}} = -\frac{\hbar\Delta}{2}\sigma_z - \frac{\hbar\Omega_0^{\uparrow\downarrow}}{2}\sigma_x + E_{00}\mathbb{1} = \begin{pmatrix} -\frac{\hbar\Delta}{2} + E_{00} & -\frac{\hbar\Omega_0^{\uparrow\downarrow}}{2} \\ -\frac{\hbar\Omega_0^{\uparrow\downarrow}}{2} & \frac{\hbar\Delta}{2} + E_{00} \end{pmatrix} \quad (3.3)$$

where we take $|\downarrow\rangle \equiv |\phi_{00}\rangle$ as ground state and $|\uparrow\rangle \equiv |\phi_{10}\rangle$ as excited state. The Rabi frequency of this transition can be written as

$$\Omega_0^{\uparrow\downarrow} = \frac{2E_{\text{ac}} \langle \phi_{10} | \mathbf{d}_0 | \phi_{00} \rangle}{\hbar} \quad (3.4)$$

In the language of the spin- $1/2$ XXZ model the Rabi frequency accounts for a transverse magnetic field whereas the detuning Δ can be seen as a static magnetic field, inducing magnetization. To describe N molecules in a two dimensional $\sqrt{N} \times \sqrt{N}$ -square lattice with lattice spacing a the

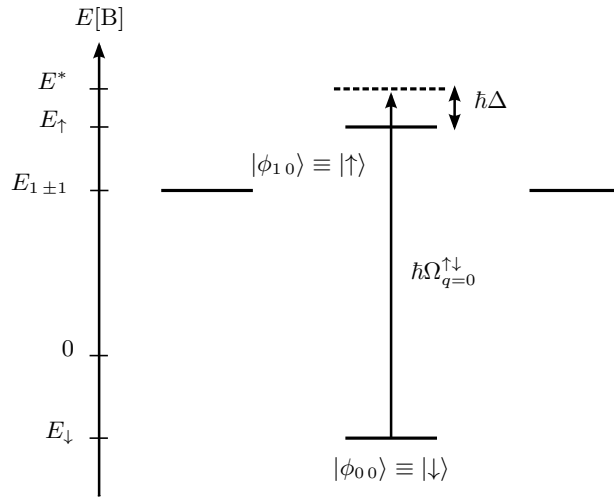


Figure 3.1 The spin- $1/2$ XXZ model is realized by weakly coupling $|\phi_{00}\rangle \longleftrightarrow |\phi_{10}\rangle$ with a linear polarized microwave field and a near resonant frequency detuned by Δ . In this way a two level system with ground state $|\downarrow\rangle$ and excited state $|\uparrow\rangle$ is engineered.

Hamiltonian (3.3) can be generalized to a two particle operator $\mathcal{H}^{(2)}$. Incorporating the dipole-dipole interaction between polar molecules the operator describing two molecules can be cast into

$$\mathcal{H}^{(2)} = \mathcal{H}_1^{\text{RWA}} + \mathcal{H}_2^{\text{RWA}} + \mathcal{V}_{ij}^{\text{dd}} \quad (3.5)$$

where the dipole-dipole interaction (2.122) between two molecules

$$\mathcal{V}_{ij}^{\text{dd}} = \frac{d_{i0}d_{j0} - \frac{3}{2}(d_{i1}d_{j1} + d_{i-1}d_{j-1}) + \frac{1}{2}(d_{i1}d_{j-1} + d_{i-1}d_{j1})}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \quad (3.6)$$

is reduced to

$$\mathcal{V}_{ij}^{\text{dd}} = \frac{d_{i0}d_{j0}}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \quad (3.7)$$

for $\Delta M = 0$ transitions. This is true because of the fact, that for symmetry reasons all matrix elements of $d_{\pm 1}$ are vanishing, leaving only the interaction terms in z -direction. The Hamiltonian for the two dimensional optical $\sqrt{N} \times \sqrt{N}$ -square lattice is the sum over all interactions between molecules at each site, omitting the diverging self interaction

$$\mathcal{H} = \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \mathcal{H}_{ij}^{(2)} = \frac{1}{2} \left\{ \sum_{i=1}^N \mathcal{H}_i + \sum_{j=1}^N \mathcal{H}_j + \sum_{\substack{i,j=1 \\ i \neq j}}^N \mathcal{V}_{ij}^{\text{dd}} \right\} \quad (3.8)$$

Since the dipole-dipole interaction operator $\mathcal{V}_{ij}^{\text{dd}}$ is a two particle operator we need to determine the matrix representation of $\mathcal{V}_{ij}^{\text{dd}}$ in the rotating wave approximation for two particle states, e.g. the direct product $|\uparrow\rangle \otimes |\uparrow\rangle$, $|\uparrow\rangle \otimes |\downarrow\rangle$, $|\downarrow\rangle \otimes |\uparrow\rangle$ and $|\downarrow\rangle \otimes |\downarrow\rangle$. The derivation of the dipolar interaction operator used projections onto the space fixed basis (see the definitions of θ and ϕ in Figure 2.10), so that we cannot use the rotating frame transformation from equation (2.90). We

need to define this rotation with respect to the space fixed frame instead

$$\left\{ \begin{array}{l} |\tilde{\uparrow}\rangle \\ |\tilde{\downarrow}\rangle \end{array} \right\} = \mathcal{R}^{\text{Microwave}} \underbrace{\left(\mathcal{R}^{\text{Space-Fixed-Frame}} \right)^{-1}}_{\mathbb{1}} \left\{ \begin{array}{l} |\uparrow\rangle \\ |\downarrow\rangle \end{array} \right\} = \begin{pmatrix} e^{-i\omega_L t} & 0 \\ 0 & e^{i\omega_L t} \end{pmatrix} \left\{ \begin{array}{l} |\uparrow\rangle \\ |\downarrow\rangle \end{array} \right\} \quad (3.9)$$

in the notation of Appendix C. The transformation for the single molecule basis states looks like

$$\begin{aligned} |\tilde{\uparrow}\rangle &= e^{-i\omega_L t} |\uparrow\rangle \\ |\tilde{\downarrow}\rangle &= e^{i\omega_L t} |\downarrow\rangle \end{aligned} \quad (3.10)$$

and for two molecule states we find

$$\begin{aligned} |\tilde{\uparrow}\tilde{\uparrow}\rangle &= |\tilde{\uparrow}\rangle \otimes |\tilde{\uparrow}\rangle = e^{-i\omega_L t} |\uparrow\rangle \otimes e^{-i\omega_L t} |\uparrow\rangle = e^{-2i\omega_L t} |\uparrow\rangle \otimes |\uparrow\rangle = e^{-2i\omega_L t} |\uparrow\uparrow\rangle \\ |\tilde{\uparrow}\tilde{\downarrow}\rangle &= e^{-i\omega_L t} |\uparrow\rangle \otimes e^{i\omega_L t} |\downarrow\rangle = |\uparrow\downarrow\rangle \\ |\tilde{\downarrow}\tilde{\uparrow}\rangle &= e^{i\omega_L t} |\downarrow\rangle \otimes e^{-i\omega_L t} |\uparrow\rangle = |\downarrow\uparrow\rangle \\ |\tilde{\downarrow}\tilde{\downarrow}\rangle &= e^{i\omega_L t} |\downarrow\rangle \otimes e^{i\omega_L t} |\downarrow\rangle = e^{2i\omega_L t} |\downarrow\downarrow\rangle \end{aligned} \quad (3.11)$$

Calculating the matrix elements and applying the rotating wave approximation

$$\begin{aligned} \langle \tilde{\uparrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\uparrow}\tilde{\uparrow} \rangle &= \langle \uparrow\uparrow | \mathcal{V}^{\text{dd}} | \uparrow\uparrow \rangle = \frac{1}{R^3} \langle \uparrow | d_0 | \uparrow \rangle \langle \uparrow | d_0 | \uparrow \rangle = \frac{(d_0^{\uparrow\uparrow})^2}{R^3} = \tilde{V}_{11}^{\text{dd}} \\ \langle \tilde{\uparrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\uparrow}\tilde{\downarrow} \rangle &= e^{2i\omega_L t} \langle \uparrow\uparrow | \mathcal{V}^{\text{dd}} | \uparrow\downarrow \rangle = \tilde{V}_{12}^{\text{dd}} \approx 0 \\ \langle \tilde{\uparrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\uparrow} \rangle &= e^{2i\omega_L t} \langle \uparrow\uparrow | \mathcal{V}^{\text{dd}} | \downarrow\uparrow \rangle = \tilde{V}_{13}^{\text{dd}} \approx 0 \\ \langle \tilde{\uparrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\downarrow} \rangle &= e^{2i\omega_L t} e^{2i\omega_L t} \langle \uparrow\uparrow | \mathcal{V}^{\text{dd}} | \downarrow\downarrow \rangle = \tilde{V}_{14}^{\text{dd}} \approx 0 \\ \langle \tilde{\uparrow}\tilde{\downarrow} | \mathcal{V}^{\text{dd}} | \tilde{\uparrow}\tilde{\uparrow} \rangle &= \langle \uparrow\downarrow | \mathcal{V}^{\text{dd}} | \uparrow\uparrow \rangle = \frac{1}{R^3} \langle \uparrow | d_0 | \uparrow \rangle \langle \downarrow | d_0 | \uparrow \rangle = \frac{d_0^{\uparrow\uparrow} d_0^{\downarrow\uparrow}}{R^3} = \tilde{V}_{22}^{\text{dd}} \\ \langle \tilde{\uparrow}\tilde{\downarrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\uparrow} \rangle &= \langle \uparrow\downarrow | \mathcal{V}^{\text{dd}} | \downarrow\uparrow \rangle = \frac{1}{R^3} \langle \uparrow | d_0 | \downarrow \rangle \langle \downarrow | d_0 | \uparrow \rangle = \frac{d_0^{\uparrow\downarrow} d_0^{\downarrow\uparrow}}{R^3} = \tilde{V}_{23}^{\text{dd}} \\ \langle \tilde{\uparrow}\tilde{\downarrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\downarrow} \rangle &= e^{2i\omega_L t} \langle \uparrow\downarrow | \mathcal{V}^{\text{dd}} | \downarrow\downarrow \rangle = \tilde{V}_{24}^{\text{dd}} \approx 0 \\ \langle \tilde{\downarrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\uparrow}\tilde{\uparrow} \rangle &= \langle \downarrow\uparrow | \mathcal{V}^{\text{dd}} | \uparrow\uparrow \rangle = \frac{1}{R^3} \langle \downarrow | d_0 | \uparrow \rangle \langle \uparrow | d_0 | \uparrow \rangle = \frac{d_0^{\downarrow\uparrow} d_0^{\uparrow\uparrow}}{R^3} = \tilde{V}_{33}^{\text{dd}} \\ \langle \tilde{\downarrow}\tilde{\uparrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\downarrow} \rangle &= e^{2i\omega_L t} \langle \downarrow\uparrow | \mathcal{V}^{\text{dd}} | \downarrow\downarrow \rangle = \tilde{V}_{34}^{\text{dd}} \approx 0 \\ \langle \tilde{\downarrow}\tilde{\downarrow} | \mathcal{V}^{\text{dd}} | \tilde{\downarrow}\tilde{\downarrow} \rangle &= \langle \downarrow\downarrow | \mathcal{V}^{\text{dd}} | \downarrow\downarrow \rangle = \frac{1}{R^3} \langle \downarrow | d_0 | \downarrow \rangle \langle \downarrow | d_0 | \downarrow \rangle = \frac{(d_0^{\downarrow\downarrow})^2}{R^3} = \tilde{V}_{44}^{\text{dd}} \end{aligned} \quad (3.12)$$

leads to the following matrix representation of \mathcal{V}^{dd}

$$\mathcal{V}^{\text{dd}} = \frac{1}{R^3} \begin{pmatrix} (d_0^{\uparrow\uparrow})^2 & 0 & 0 & 0 \\ 0 & d_0^{\uparrow\uparrow} d_0^{\downarrow\downarrow} & (d_0^{\uparrow\downarrow})^2 & 0 \\ 0 & (d_0^{\uparrow\downarrow})^2 & d_0^{\uparrow\uparrow} d_0^{\downarrow\downarrow} & 0 \\ 0 & 0 & 0 & (d_0^{\downarrow\downarrow})^2 \end{pmatrix} \quad (3.13)$$

which can be generalized for the two dimensional square lattice. The total Hamiltonian \mathcal{H} can be expressed in different operator sets that can be represented in the two level basis

$$\{|\tilde{\uparrow}\rangle, |\tilde{\downarrow}\rangle\} \equiv \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

$$\mathbb{1} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad \sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3.14)$$

$$\sigma_+ = \frac{1}{2}(\sigma_x + i\sigma_y) = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \sigma_- = \frac{1}{2}(\sigma_x - i\sigma_y) = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad (3.15)$$

$$\mathcal{P}_\uparrow = \frac{\mathbb{1} + \sigma_z}{2} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad \mathcal{P}_\downarrow = \frac{\mathbb{1} - \sigma_z}{2} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \quad (3.16)$$

The corresponding two particle operators can be easily generated with the help of the tensor product of matrices as can be seen in Appendix D equation (D.26) to (D.41). Describing the total Hamiltonian in this set of operators

$$\mathcal{H}_i + \mathcal{H}_j = -\hbar\Delta(\mathcal{P}_{i\uparrow} + \mathcal{P}_{j\uparrow} - \mathbb{1}) - \frac{\hbar\Omega_0^{\uparrow\downarrow}}{2}(\sigma_{i+} + \sigma_{i-} + \sigma_{j+} + \sigma_{j-}) + 2E_{00}\mathbb{1} \quad (3.17)$$

$$\mathcal{V}_{ij}^{\text{dd}} = \frac{1}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \left\{ (d_0^{\uparrow\uparrow})^2 \mathcal{P}_{i\uparrow} \mathcal{P}_{j\uparrow} + d_0^{\uparrow\uparrow} d_0^{\downarrow\downarrow} (\mathcal{P}_{i\uparrow} \mathcal{P}_{j\downarrow} + \mathcal{P}_{i\downarrow} \mathcal{P}_{j\uparrow}) \right. \\ \left. + (d_0^{\uparrow\downarrow})^2 (\sigma_{i+} \sigma_{j-} + \sigma_{i-} \sigma_{j+}) + (d_0^{\downarrow\downarrow})^2 \mathcal{P}_{i\downarrow} \mathcal{P}_{j\downarrow} \right\} \quad (3.18)$$

Summing up the free sums yields

$$\mathcal{H} = \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \mathcal{H}_{ij}^{(2)} = \sum_{i=1}^N \mathcal{H}_i + \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \mathcal{V}_{ij}^{\text{dd}} \\ = NE_{00} + \frac{\hbar\Delta}{2} - \hbar\Delta \sum_{i=1}^N \mathcal{P}_{i\uparrow} - \frac{\hbar\Omega_0^{\uparrow\downarrow}}{2} \sum_{i=1}^N (\sigma_{i+} + \sigma_{i-})$$

$$\begin{aligned}
& + \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \frac{\left(d_0^{\uparrow\uparrow}\right)^2}{a^3} \frac{\mathcal{P}_{i\uparrow}\mathcal{P}_{j\downarrow}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{d_0^{\uparrow\uparrow}d_0^{\downarrow\downarrow}}{a^3} \frac{\mathcal{P}_{i\uparrow}\mathcal{P}_{j\downarrow} + \mathcal{P}_{i\downarrow}\mathcal{P}_{j\uparrow}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\
& \quad \left. + \frac{\left(d_0^{\uparrow\downarrow}\right)^2}{a^3} \frac{\sigma_{i+}\sigma_{j-} + \sigma_{i-}\sigma_{j+}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{\left(d_0^{\downarrow\downarrow}\right)^2}{a^3} \frac{\mathcal{P}_{i\downarrow}\mathcal{P}_{j\uparrow}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \quad (3.19)
\end{aligned}$$

The set of operators $\{\sigma_+, \sigma_-, \mathcal{P}_\uparrow, \mathcal{P}_\downarrow\}$ can be converted to $\{\sigma_x, \sigma_y, \sigma_z, \mathbb{1}\}$ by

$$\sigma_+ = \frac{1}{2}(\sigma_x + i\sigma_y) \quad \mathcal{P}_\uparrow = \frac{1}{2}(\mathbb{1} + \sigma_z) \quad \mathbb{1} = \mathcal{P}_\uparrow + \mathcal{P}_\downarrow \quad \sigma_y = -i(\sigma_+ - \sigma_-) \quad (3.20)$$

$$\sigma_- = \frac{1}{2}(\sigma_x - i\sigma_y) \quad \mathcal{P}_\downarrow = \frac{1}{2}(\mathbb{1} + \sigma_z) \quad \sigma_x = \sigma_+ + \sigma_- \quad \sigma_z = \mathcal{P}_\uparrow - \mathcal{P}_\downarrow \quad (3.21)$$

By inserting this conversion into equation(3.17) and (3.18) the total Hamiltonian reads

$$\begin{aligned}
\mathcal{H} &= -\frac{\hbar\Omega_0^{\uparrow\downarrow}}{4} \underbrace{\left(\sum_{i=1}^N \sigma_{ix} + \sum_{j=1}^N \sigma_{jx} \right)}_{= 2 \sum_{i=1}^N \sigma_{ix}} - \frac{\hbar\Delta}{4} \underbrace{\left(\sum_{i=1}^N \sigma_{iz} + \sum_{j=1}^N \sigma_{jz} \right)}_{= 2 \sum_{i=1}^N \sigma_{iz}} + E_{00} \underbrace{\sum_{i=1}^N \mathbb{1}_i}_{= N} \\
&+ \frac{\left(d_0^{\uparrow\uparrow} + d_0^{\downarrow\downarrow}\right)^2}{8a^3} \underbrace{\sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\mathbb{1}_i \mathbb{1}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3}}_{N \sum_{\substack{\mathbf{R}_N \\ \mathbf{R}_n \neq 0}} \frac{\mathbb{1}_{\mathbf{R}_n}}{|\mathbf{R}_n|^3}} + \frac{\left(d_0^{\uparrow\uparrow}\right)^2 - \left(d_0^{\downarrow\downarrow}\right)^2}{8a^3} \underbrace{\sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\sigma_{iz} + \sigma_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3}}_{2 \sum_{i=1}^N \sigma_{iz} \sum_{\substack{\mathbf{R}_N \\ \mathbf{R}_n \neq 0}} \frac{1}{|\mathbf{R}_n|^3}} \\
&+ \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \frac{\left(d_0^{\uparrow\downarrow}\right)^2}{4a^3} \frac{\sigma_{ix}\sigma_{jx} + \sigma_{iy}\sigma_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{\left(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow}\right)^2}{8a^3} \frac{\sigma_{iz}\sigma_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \quad (3.22)
\end{aligned}$$

Using the fact that the two dimensional square lattice is translational invariant, one particle operator terms depending on one position can be rewritten in terms of the relative position vector $\mathbf{R}_n = \mathbf{R}_i - \mathbf{R}_j$ ignoring the reference to an explicit point in the lattice. Thus the sum over the second subscript can be carried out. In the following the summation over complete shells containing all n^{th} nearest neighbors will be denoted by

$$\sum_{\substack{\mathbf{R}_N \\ \mathbf{R}_n \neq 0}} \longrightarrow \sum_{n=1}^N \quad (3.23)$$

so that the notation will not be cluttered up needlessly. Inserting $\sigma = 2/\hbar\mathbf{S}$ into (3.22) the total

Hamiltonian can be cast into

$$\begin{aligned}
\mathcal{H} = & NE_{00} + \frac{N(d_0^{\uparrow\uparrow} + d_0^{\downarrow\downarrow})^2}{8a^3} \sum_{i=1}^N \frac{1}{|\mathbf{R}_i|^3} \\
& - \sum_{i=1}^N \left\{ \Omega_0^{\uparrow\downarrow} \mathcal{S}_{ix} + \left[\Delta - \frac{(d_0^{\uparrow\uparrow})^2 - (d_0^{\downarrow\downarrow})^2}{2\hbar a^3} \sum_{n=1}^N \frac{1}{|\mathbf{R}_n|^3} \right] \mathcal{S}_{iz} \right\} \\
& + \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \frac{(d_0^{\downarrow\downarrow})^2}{\hbar^2 a^3} \frac{\mathcal{S}_{ix} \mathcal{S}_{jx} + \mathcal{S}_{iy} \mathcal{S}_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow})^2}{2\hbar^2 a^3} \frac{\mathcal{S}_{iz} \mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \quad (3.24)
\end{aligned}$$

and compared to the spin-1/2 XXZ model Hamiltonian with an external magnetic field \mathbf{h}

$$\mathcal{H}^{\text{XXZ}} = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(J_{\perp} \frac{\mathcal{S}_{ix} \mathcal{S}_{jx} + \mathcal{S}_{iy} \mathcal{S}_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J_z \frac{\mathcal{S}_{iz} \mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) - \sum_{i=1}^N \mathbf{h}_i \cdot \mathbf{S}_i \quad (3.25)$$

This gives rise to the following relations connecting the dipole moments with the exchange coupling constants

$$J_{\perp} = J \sin \vartheta = \left(\frac{2}{\hbar} \right)^2 \frac{(d_0^{\uparrow\downarrow})^2}{4a^3} = \frac{(d_0^{\uparrow\downarrow})^2}{\hbar^2 a^3} \quad (3.26)$$

$$J_z = J \cos \vartheta = \left(\frac{2}{\hbar} \right)^2 \frac{(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow})^2}{8a^3} = \frac{(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow})^2}{2\hbar^2 a^3} \quad (3.27)$$

and the relation for the external magnetic field

$$\mathbf{h}_i = \Omega_0^{\uparrow\downarrow} \mathbf{e}_x + \left(\Delta - \frac{(d_0^{\uparrow\uparrow})^2 - (d_0^{\downarrow\downarrow})^2}{2\hbar a^3} \sum_{n=1}^N \frac{1}{|\mathbf{R}_n|^3} \right) \mathbf{e}_z \quad (3.28)$$

The magnetic field can be made homogeneous throughout the whole lattice because

$$\sum_{n=1}^N \frac{1}{|\mathbf{R}_n|^3} = g^{\text{F}} \quad (3.29)$$

will be explicitly calculated in Subsection 3.3.3, see for instance equation (3.99). The definition and detailed explanation of the ferromagnetic lattice constant g^{F} can be found in Section 3.3. In the case of a homogeneous \mathbf{h} -field, we can choose a global detuning Δ in such a way that h_z is vanishing. Since we will employ a weak coupling of the two states, $\Omega_0^{\uparrow\downarrow} \approx 0$ which will eventually be turned off, so that the transverse magnetic field is also vanishing, leading to $\mathbf{h} = \mathbf{0}$.

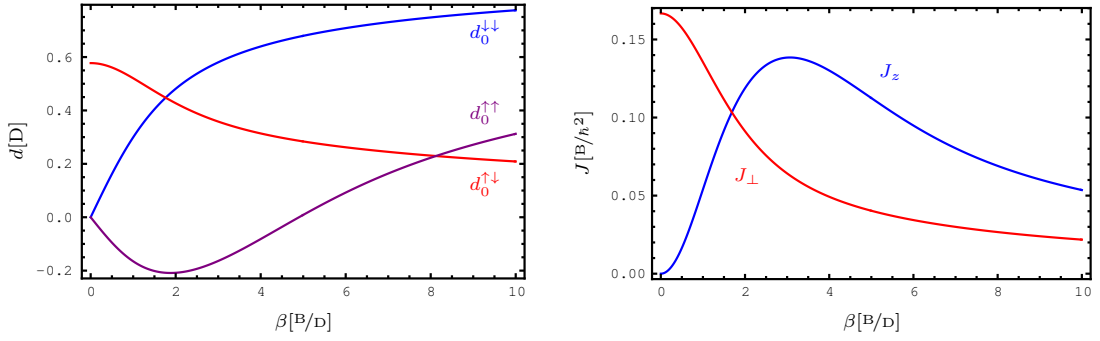


Figure 3.2 Left Panel: Induced dipole moments controlling the interaction strength. The $d_0^{\uparrow\uparrow}$ and $d_0^{\downarrow\downarrow}$ describes static interaction whereas the spin flip dipole moment $d_0^{\uparrow\downarrow}$ describes an exchange of photon between two molecules. Right Panel: In-axis and in-plane coupling constants J_z and J_{\perp} in dependence of the parameter β that can be tuned by the static external electric field E_{dc} (dc-Stark shift).

The global energy shift written in terms of g^F

$$NE_{00} + \frac{N(d_0^{\uparrow\uparrow} + d_0^{\downarrow\downarrow})^2}{8a^3} \sum_{i=1}^N \frac{\mathbb{1}_i}{|\mathbf{R}_i|^3} = N \left[E_{00} + g^F \frac{(d_0^{\uparrow\uparrow} + d_0^{\downarrow\downarrow})^2}{8a^3} \right] \quad (3.30)$$

can be scaled away as usual, since it gives no physical contribution. Finally we arrive at the spin- $1/2$ XXZ model

$$\mathcal{H}^{\text{XXZ}} = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\underbrace{\frac{(d_0^{\uparrow\downarrow})^2}{\hbar^2 a^3}}_{J_{\perp}} \frac{S_{ix}S_{jx} + S_{iy}S_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \underbrace{\frac{(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow})^2}{2\hbar^2 a^3}}_{J_z} \frac{S_{iz}S_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (3.31)$$

The first sum arises due to the dipole-dipole interaction between two polar molecules and its first term describes an exchange of photons between two polar molecules which is linked to a simultaneous spin flip of both spins. The second term of the dipole-dipole interaction part describes the static interaction in z -direction due to the field induced dipole moments of the ground state and the excited state. The spin- $1/2$ Heisenberg model can be realized for $\vartheta = \pi/4$ leading to $J_{\perp} = J_z$. For the $|\phi_{00}\rangle \leftrightarrow |\phi_{10}\rangle$ coupling, we can calculate the dipole moments $d_0^{\uparrow\uparrow}$, $d_0^{\downarrow\downarrow}$ and $d_0^{\uparrow\downarrow}$ by using the numerically determined eigensystem of $\mathcal{J} + \beta/da\mathcal{d}$. With the help of the MATHEMATICA program H.4 the dipole moments and the coupling constants are determined and plotted for the given setup. The possible values for the coupling constants J_{\perp} and J_z in dependence on $\beta = dE_{dc}/B$ are shown in Figure 3.2. We see that for a vanishing static field, i.e. $\beta = 0$ the field induced dipole moments of the static interaction are also vanishing. The angle of the coupling constants ϑ can be calculated by

$$\vartheta(\beta) = \arctan \left(\frac{J_{\perp}(\beta)}{J_z(\beta)} \right) \quad (3.32)$$

For vanishing in-axis interaction J_z we can realize the antiferromagnetic XY model. The values of ϑ ranges from $\pi/2$ to the minimal value $\approx 19.7^\circ$ for $\beta \approx 5.24$, as shown in Figure 3.3. For

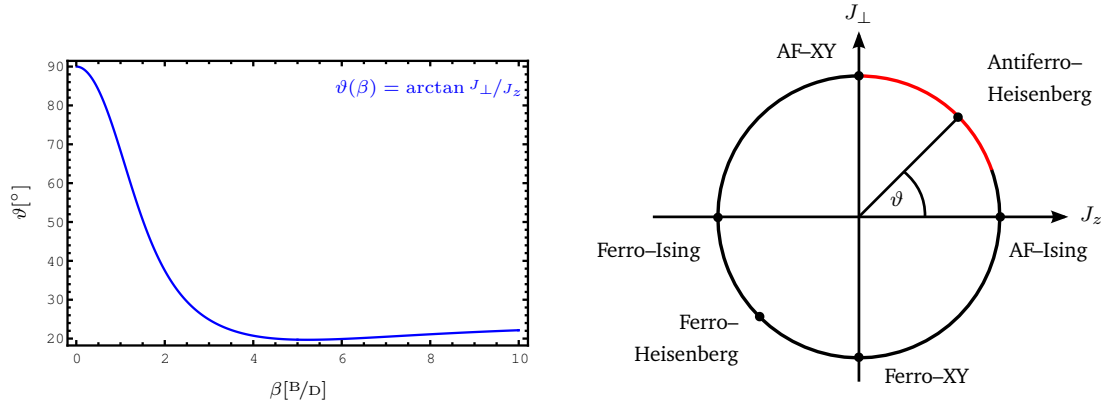


Figure 3.3 In dependence on the static electric field E_{dc} different models can be realized. The accessible values of $\vartheta(\beta)$ are indicated in the left picture by the red circular segment.

$\beta \approx 1.6876$ we can reach the Heisenberg point and realize the antiferromagnetic Heisenberg model for appropriate polar molecules in a two dimensional square lattice.

3.3 Spin Waves Excitations

Having derived the spin- $1/2$ XXZ model for polar molecules in an optical two dimensional square lattice, we now look at excitation above the mean field ground state which will be discussed in detail in the following Section 3.4. Here we will concentrate on non-interacting, i.e. linear harmonic excitations, the so called spin waves, that can be described by bosonic operators obeying the common commutator relations. A nice introduction to the theory of spin waves in general can be found in [14]. Following [15] we first use the Holstein-Primakoff transformation and linearize the resulting Hamiltonian which is then diagonalized by successive transformation in Fourier space from which we determine the dispersion relation of the spin waves. This procedure is carried out for the ferromagnetic as well as for the antiferromagnetic case. We introduce some precursory calculations in order to handle the Fourier transformation of the dipolar interaction which represents a crucial part in the understanding of the spin wave dispersion relation.

3.3.1 Fourier Transform of the $1/R^3$ -Interaction

The Fourier transform of the interaction potential can be written in compact notation as

$$\epsilon(\mathbf{k}) = \sum_{\mathbf{R}_n \neq 0} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} \quad |\mathbf{R}_n| = |\mathbf{R}_i - \mathbf{R}_j| \quad (3.33)$$

The sum is running over all n^{th} nearest neighbors as is described in (F.38) in detail. For small values of $\mathbf{k} \approx \mathbf{0}$ the discrete Fourier transformation of $1/R^3$ can be approximated by the continuous Fourier transform

$$\epsilon(\mathbf{k}) = \sum_{\mathbf{R}_n \neq 0} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} \approx \int_{\phi=0}^{2\pi} d\phi \int_{r=r_c}^{\infty} dr r \frac{e^{i\mathbf{k} \cdot \mathbf{r}}}{r^3} = -|\mathbf{k}| + C \quad (3.34)$$

where r_c describes an appropriate short distance cut-off and C is an arbitrary integration constant. For a more rigorous and precise treatment one needs to manipulate the above sum running over all lattice points. With the help of the so called Ewald-summation [16] one can reach a fast convergence. Rewriting the sum using the relation

$$\int_0^\infty d\lambda \lambda^\nu e^{-\lambda|\mathbf{R}_n|^2} = \int_0^\infty \frac{d\lambda'}{|\mathbf{R}_n|^2} \left(\frac{\lambda'}{|\mathbf{R}_n|^2} \right)^\nu e^{-\lambda'} = \underbrace{\int_0^\infty d\lambda' e^{-\lambda'} \lambda'^\nu}_{\Gamma(\nu+1)} \frac{1}{|\mathbf{R}_n|^{2+2\nu}} = \frac{\Gamma(\nu+1)}{|\mathbf{R}_n|^{2(\nu+1)}} \quad (3.35)$$

and applying the substitution $\lambda' = \lambda |\mathbf{R}_n|^2$ with the help of the so called Γ -function

$$\Gamma(\nu+1) = \int_0^\infty d\lambda \lambda^\nu e^{-\lambda} \quad (3.36)$$

In the special case of $|\mathbf{R}_n|^{-3}$, equation (3.35) can be cast into

$$\frac{1}{|\mathbf{R}_n|^3} = \frac{2}{\sqrt{\pi}} \int_0^\infty d\lambda \lambda^{\frac{1}{2}} e^{-\lambda|\mathbf{R}_n|^2} \quad (3.37)$$

by setting $2(\nu+1) = 3 \Leftrightarrow \nu = \frac{1}{2}$ and using

$$\Gamma\left(\frac{3}{2}\right) = \Gamma\left(1 + \frac{1}{2}\right) = \frac{1}{2}\Gamma\left(\frac{1}{2}\right) = \frac{\sqrt{\pi}}{2} \quad (3.38)$$

to solve equation (3.35) for $|\mathbf{R}_n|^{-3}$. Now we are able to rewrite the sum (3.34) as

$$\epsilon(\mathbf{k}) = \frac{2}{\sqrt{\pi}} \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} \int_0^\infty d\lambda \lambda^{\frac{1}{2}} e^{-\lambda|\mathbf{R}_n|^2} e^{i\mathbf{k}\cdot\mathbf{R}_n} \quad (3.39)$$

Splitting the sum at η into two parts

$$\epsilon(\mathbf{k}) = \frac{2}{\sqrt{\pi}} \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} \left(\int_0^\eta + \int_\eta^\infty \right) \left(d\lambda \lambda^{\frac{1}{2}} e^{-\lambda|\mathbf{R}_n|^2} \right) e^{i\mathbf{k}\cdot\mathbf{R}_n} \quad (3.40)$$

and applying the substitution $\lambda' = \frac{\eta}{\lambda}$ to the first integral gives rise to

$$\begin{aligned} \int_0^\eta d\lambda \lambda^{\frac{1}{2}} e^{-\lambda|\mathbf{R}_n|^2} &= - \int_\infty^1 d\lambda' \frac{\eta}{\lambda'^2} \left(\frac{\eta}{\lambda'} \right)^{\frac{1}{2}} e^{-\frac{\eta}{\lambda'}|\mathbf{R}_n|^2} \\ &= \int_1^\infty d\lambda' \left(\frac{\eta^3}{\lambda'^5} \right)^{\frac{1}{2}} e^{-\frac{\eta}{\lambda'}|\mathbf{R}_n|^2} \\ &= \eta^{\frac{3}{2}} \int_1^\infty d\lambda \lambda^{-\frac{5}{2}} e^{-\frac{\eta}{\lambda}|\mathbf{R}_n|^2} \end{aligned} \quad (3.41)$$

Applying a similar substitution $\lambda' = \frac{\lambda}{\eta}$ to the second integral yields

$$\int_{\eta}^{\infty} d\lambda \lambda^{\frac{1}{2}} e^{-\lambda|\mathbf{R}_n|^2} = \eta \int_1^{\infty} d\lambda' \eta^{\frac{1}{2}} \lambda'^{\frac{1}{2}} e^{-\eta\lambda'|\mathbf{R}_n|^2} = \eta^{\frac{3}{2}} \int_1^{\infty} d\lambda \lambda^{\frac{1}{2}} e^{-\eta\lambda|\mathbf{R}_n|^2} \quad (3.42)$$

Inserting both equations (3.41) and (3.42) in $\epsilon(\mathbf{k})$

$$\epsilon(\mathbf{k}) = \frac{2}{\sqrt{\pi}} \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} \eta^{\frac{3}{2}} \left\{ \int_1^{\infty} d\lambda \lambda^{-\frac{5}{2}} e^{-\frac{\eta}{\lambda}|\mathbf{R}_n|^2} + \int_1^{\infty} d\lambda \lambda^{\frac{1}{2}} e^{-\eta\lambda|\mathbf{R}_n|^2} \right\} e^{i\mathbf{k}\cdot\mathbf{R}_n} \quad (3.43)$$

Using the Poisson summation formula from Appendix F.4 for the first integral

$$\sum_{\mathbf{R}_n}^{\mathbf{R}_N} e^{-\frac{\eta}{\lambda}|\mathbf{R}_n|^2} e^{i\mathbf{k}\cdot\mathbf{R}_n} = \frac{\pi}{a^2 \left(\frac{\eta}{\lambda}\right)} \sum_{\mathbf{q}_i} e^{-\frac{|\mathbf{q}_i+\mathbf{k}|^2}{4\left(\frac{\eta}{\lambda}\right)}} = \frac{\pi\lambda}{a^2\eta} \sum_{\mathbf{q}_i} e^{-\frac{\lambda}{4\eta}|\mathbf{q}_i+\mathbf{k}|^2} \quad (3.44)$$

and considering that we must subtract the $\mathbf{R}_{n=0}$ term because the Poisson summation formula takes the sum over the whole lattice including the origin

$$\sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} = \sum_{\mathbf{R}_n}^{\mathbf{R}_N} - \sum_{\mathbf{R}_n=0}^{\mathbf{R}_N} \quad (3.45)$$

we finally find

$$\begin{aligned} \epsilon(\mathbf{k}) &= \frac{2\sqrt{\pi}}{a^2} \eta^{\frac{1}{2}} \sum_{\mathbf{q}_i} \int_1^{\infty} d\lambda \lambda^{-\frac{3}{2}} e^{-\frac{\lambda}{4\eta}|\mathbf{q}_i+\mathbf{k}|^2} - \underbrace{\frac{2}{\sqrt{\pi}} \eta^{\frac{3}{2}} \int_1^{\infty} d\lambda \lambda^{-\frac{5}{2}} e^{-\frac{\eta}{\lambda}|\mathbf{0}|^2}}_{\mathbf{R}_{n=0} \text{ term}} \\ &\quad + \frac{2}{\sqrt{\pi}} \eta^{\frac{3}{2}} \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} \int_1^{\infty} d\lambda \lambda^{\frac{1}{2}} e^{-\eta\lambda|\mathbf{R}_n|^2} e^{i\mathbf{k}\cdot\mathbf{R}_n} \end{aligned} \quad (3.46)$$

Integrating equation (3.46) over λ yields

$$\begin{aligned} \epsilon(\mathbf{k}) &= \sum_{\mathbf{q}_i} \left\{ \frac{4\sqrt{\pi}}{a^2} \sqrt{\eta} e^{-\frac{|\mathbf{q}_i+\mathbf{k}|^2}{4\eta}} - \frac{2\pi}{a^2} |\mathbf{q}_i + \mathbf{k}| \text{Errc} \left(\frac{|\mathbf{q}_i + \mathbf{k}|}{2\sqrt{\eta}} \right) \right\} - \frac{4\eta^{\frac{3}{2}}}{3\sqrt{\pi}} \\ &\quad + \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} \left\{ \left(2\sqrt{\frac{\eta}{\pi}} \frac{e^{-\eta|\mathbf{R}_n|^2}}{|\mathbf{R}_n|^2} + \frac{\text{Errc}(\sqrt{\eta}|\mathbf{R}_n|)}{|\mathbf{R}_n|^3} \right) e^{i\mathbf{k}\cdot\mathbf{R}_n} \right\} \end{aligned} \quad (3.47)$$

with the complementary error function defined as

$$\text{Errc}(x) = 1 - \frac{2}{\sqrt{\pi}} \int_0^x dt e^{-t^2} \quad (3.48)$$

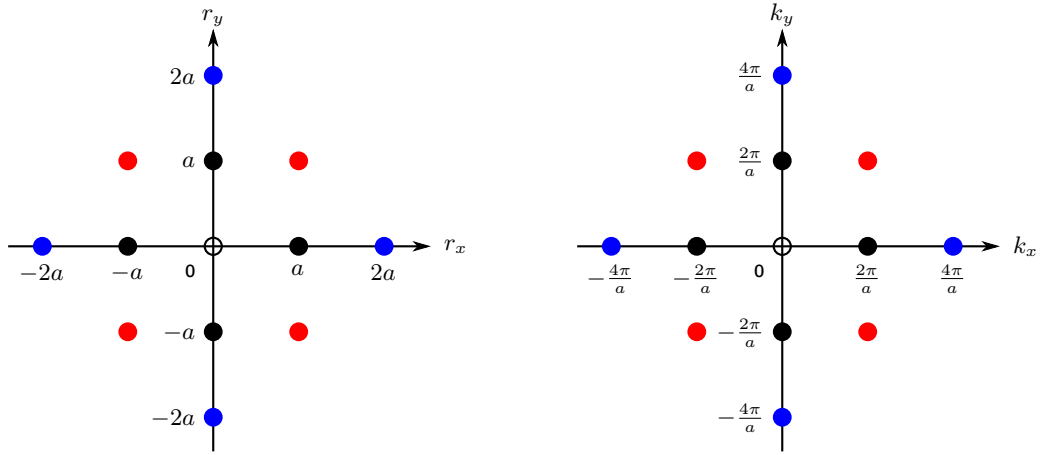


Figure 3.4 The first three shells to be summed over in direct space (see left panel) are $R_1 = (1, 0)^t$, $(0, 1)^t$, $(-1, 0)^t$ and $(0, -1)^t$. In \mathbf{k} -space (see right panel) the first shell consists of $(2\pi, 0)^t$, $(0, 2\pi)^t$, $(-2\pi, 0)^t$ and $(0, -2\pi)^t$. The second shell denoted with red dots consist of four terms with $|R_2| = \sqrt{2}$ and $|\mathbf{q}_2| = 2\sqrt{2}\pi$ and the third shell colored in blue with $|R_3| = 2$ and $|\mathbf{q}_3| = 4\pi$. For further details see also Figure F.2 in Appendix F.5.2.

Additionally we introduce the function $A_\eta(|\mathbf{R}_n|)$ as the amplitude of the plane waves in the real space summation

$$A_\eta(|\mathbf{R}_n|) = \left(2\sqrt{\frac{\eta}{\pi}} \frac{e^{-\eta|\mathbf{R}_n|^2}}{|\mathbf{R}_n|^2} + \frac{\text{Ercr}(\sqrt{\eta}|\mathbf{R}_n|)}{|\mathbf{R}_n|^3} \right) \quad (3.49)$$

and the summands in \mathbf{k} -space

$$C_\eta(|\mathbf{q}_i + \mathbf{k}|) = \frac{4\sqrt{\eta\pi}}{a^2} e^{-\frac{|\mathbf{q}_i + \mathbf{k}|^2}{4\eta}} - \frac{2\pi}{a^2} |\mathbf{q}_i + \mathbf{k}| \text{Ercr} \left(\frac{|\mathbf{q}_i + \mathbf{k}|}{2\sqrt{\eta}} \right) \quad (3.50)$$

leading to the simplification

$$\epsilon_\eta(\mathbf{k}) = \sum_{\mathbf{q}_i} C_\eta(|\mathbf{q}_i + \mathbf{k}|) + \sum_{\mathbf{R}_n \neq 0}^{\mathbf{R}_N} A_\eta(|\mathbf{R}_n|) e^{i\mathbf{k} \cdot \mathbf{R}_n} - \frac{4\eta^{\frac{3}{2}}}{3\sqrt{\pi}} \quad (3.51)$$

In order to compare it to the continuous Fourier transform in equation (3.34) for $\mathbf{k} \approx \mathbf{0}$ we expand $\epsilon_\eta(\mathbf{k})$ in \mathbf{k} around zero. It suffices to look at the series expansion of $C_\eta(|\mathbf{q}_i + \mathbf{k}|)$ and $e^{i\mathbf{k} \cdot \mathbf{R}_n}$ since $A_\eta(|\mathbf{R}_n|)$ does not depend on \mathbf{k}

$$C_\eta(|\mathbf{q}_i + \mathbf{k}|) = \frac{4\sqrt{\eta\pi}}{a^2} - \frac{2\pi}{a^2} |\mathbf{q}_i + \mathbf{k}| + O(|\mathbf{q}_i + \mathbf{k}|^2) \quad (3.52)$$

$$e^{i\mathbf{k} \cdot \mathbf{R}_n} = 1 + i\mathbf{k} \cdot \mathbf{R}_n - \frac{1}{2} |\mathbf{k}|^2 |\mathbf{R}_n|^2 + O(|\mathbf{k}|^3)$$

Since we will sum over complete shells in real space each term with an odd number of \mathbf{R}_n involved, will cancel out. These are also the imaginary terms, so that the series expansion of

Shell	$\eta = 1$		$\eta = \pi$	
	$ \mathbf{q}_i $	$ \mathbf{R}_n $	$ \mathbf{q}_i $	$ \mathbf{R}_n $
0 th	7.08982	–	12.5664	–
1 st	$6.51665 \cdot 10^{-5}$	2.28963	0.247377	0.394467
2 nd	$1.79156 \cdot 10^{-9}$	0.369766	$6.15741 \cdot 10^{-3}$	$8.0252 \cdot 10^{-3}$
3 rd	$2.47869 \cdot 10^{-19}$	0.0230059	$6.27331 \cdot 10^{-6}$	$7.24227 \cdot 10^{-6}$

Table 3.1 Numerical contributions to $\epsilon(\mathbf{0})$ are shown for $\eta = 1$ and $\eta = \pi$. As has been expected from the analytical comparison of the exponential cut off functions, the numerical values are nearly split symmetrical between the real space summands and the \mathbf{k} -space summands for $\eta = \pi$.

$\epsilon_\eta(\mathbf{k})$ remains real up to all orders as expected. Taking the limit $\mathbf{k} \rightarrow 0$, we see that $\epsilon(\mathbf{k})_\eta$ approaches zero linearly, leading to the same result as in (3.34).

What can we expect from the analytic expression of $\epsilon_\eta(\mathbf{k})$ in terms of the two sums over real and reciprocal space? Mathematically we used the fact that the second integrand is cut off with $e^{-\eta\lambda|\mathbf{R}_n|^2}$ which lead to a fast converging summation on the interval $[1, \infty)$ and the first integrand initially leads to a very slow convergence because $e^{-\eta\lambda|\mathbf{R}_n|^2}$ does not cut off higher summands. But with the help of the Poisson summation formula we can identify the first sum with the sum of the Fourier transform and since Gaussian function invert their half width, we end up with an fast converging integrand in the Fourier space. Physically one can realize that the summation in real space over long wavelengths will lead to a fast convergence whereas for short wavelength the sum in the reciprocal space is fast converging. The parameter η is now determining the scale where to split the summation in long and in short wavelength. Analytically we can compare the magnitude of the two cut off factors

$$e^{-\frac{\lambda}{4\eta}|\mathbf{q}_i+\mathbf{k}|^2} \text{ (sum over reciprocal lattice)} \quad \longleftrightarrow \quad e^{-\eta\lambda|\mathbf{R}_n|^2} \text{ (sum over real space)} \quad (3.53)$$

for the first shell in direct and reciprocal space as depicted in Figure 3.4. Thus for $\mathbf{k} = \mathbf{0}$, $|\mathbf{q}_1| = 2\pi$ and $|\mathbf{R}_1| = 1$ we find

$$e^{-\frac{\lambda}{4\eta}|2\pi|^2} = e^{-\lambda\frac{\pi^2}{\eta}} = e^{-\lambda\pi} \quad (3.54)$$

and

$$e^{-\eta\lambda|1|^2} = e^{-\lambda\eta} = e^{-\lambda\pi} \quad (3.55)$$

are equal for $\eta = \pi$. Therefore we will expect that the contributions to the sum in direct and reciprocal space will be of the same order of magnitude. For the second shell ($|\mathbf{q}_2| = 2\sqrt{2}\pi$ and $|\mathbf{R}_2| = \sqrt{2}$) we find for $\eta = \pi$ that the two exponential functions are identical as well

$$\frac{e^{-\frac{\lambda}{4\pi}8\pi^2}}{e^{-2\pi\lambda}} = 1 \quad (3.56)$$

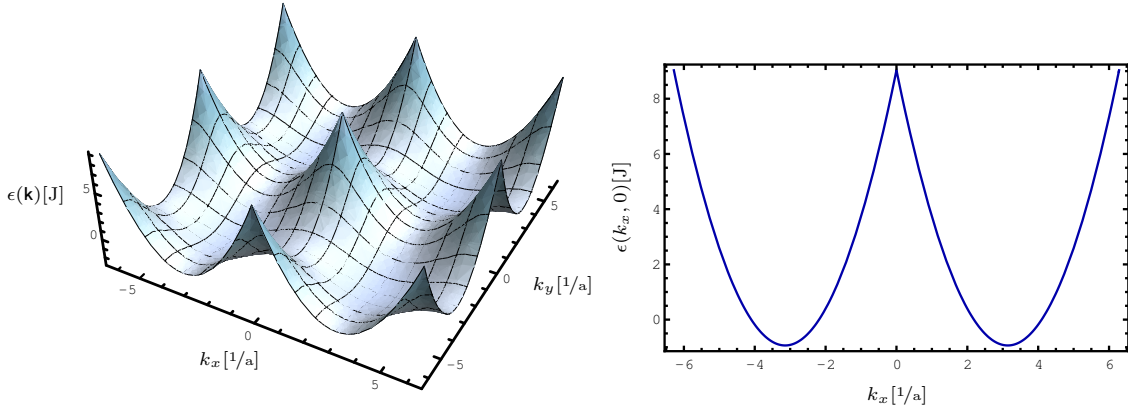


Figure 3.5 Left Panel: Fourier transform of the $1/R^3$ -interaction $\epsilon(\mathbf{k})$ in the first Brillouin zone
Right Panel: Profile of the two dimensional Fourier transform along the x -axis, showing $\epsilon(kx, 0)$.

Compared to taking $\eta = 1$ where the two exponential functions differ only by a factor of

$$\frac{e^{-2\pi^2\lambda}}{e^{-2\lambda}} = e^{\pi^2} \quad (3.57)$$

this factor is by $e^{-\pi^2} \approx 2 \cdot 10^{-4}$ smaller than the one with $\eta = 1$. In Table 3.1 the numerical values of the first three shells – consisting of for summands in a two dimensional lattice – for two different η are shown. Thus for $\eta = \pi$ it suffices to take only terms up to the third shell, i.e. $|\mathbf{q}_i| = 0, 2\pi, 2\sqrt{2}\pi, 4\pi$ and $|\mathbf{R}_n| = 1, \sqrt{2}, 2$. Note that we take the 0th shell in \mathbf{k} -space and therefore we have to take into account the correction of subtracting the \mathbf{R}_0 term

$$\begin{aligned} \epsilon_{\eta=\pi}(\mathbf{k}) &= \frac{2\pi}{a} \sum_{\mathbf{q}_i} \left\{ 2 e^{-\frac{|\mathbf{q}_i+\mathbf{k}|^2}{4\pi}} - |\mathbf{q}_i + \mathbf{k}| \text{Errc} \left(\frac{|\mathbf{q}_i + \mathbf{k}|}{2\sqrt{\pi}} \right) \right\} \\ &\quad + \sum_{\mathbf{R}_n \neq 0} \left\{ \left(2 \frac{e^{-\pi|\mathbf{R}_n|^2}}{|\mathbf{R}_n|^2} + \frac{\text{Errc}(\sqrt{\pi}|\mathbf{R}_n|)}{|\mathbf{R}_n|^3} \right) e^{i\mathbf{k} \cdot \mathbf{R}_n} \right\} - \frac{4\pi}{3} \\ &= \sum_{\mathbf{q}_i=0}^{\mathbf{q}_3} C_\pi(|\mathbf{q}_i + \mathbf{k}|) \sum_{\mathbf{R}_n=1}^{\mathbf{R}_3} A_\pi(|\mathbf{R}_n|) - \frac{4\pi}{3} \end{aligned} \quad (3.58)$$

$$\begin{aligned} \Rightarrow \epsilon_{\eta=\pi}(\mathbf{k}) &= C_\pi(|\mathbf{0} + \mathbf{k}|) \\ &\quad + C_\pi \left(\left| \begin{pmatrix} 2\pi \\ 0 \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} 0 \\ 2\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} -2\pi \\ 0 \end{pmatrix} + \mathbf{k} \right| \right) \\ &\quad + C_\pi \left(\left| \begin{pmatrix} 0 \\ -2\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} 2\pi \\ 2\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} -2\pi \\ 2\pi \end{pmatrix} + \mathbf{k} \right| \right) \\ &\quad + C_\pi \left(\left| \begin{pmatrix} -2\pi \\ -2\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} 2\pi \\ -2\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_\pi \left(\left| \begin{pmatrix} 4\pi \\ 0 \end{pmatrix} + \mathbf{k} \right| \right) \end{aligned}$$

Position	#n	$\epsilon_{\pi}^{\text{num}}(\mathbf{k})$	$\epsilon_{\pi}(\mathbf{k})$	$ \epsilon_{\pi}^{\text{num}} - \epsilon_{\pi} $
$\mathbf{k} = \mathbf{0}$	100	8.977335157		$5.63 \cdot 10^{-2}$
	1000	9.027967657	9.033620744	$5.65 \cdot 10^{-3}$
	10000	9.033056026		$5.65 \cdot 10^{-4}$
$\mathbf{k} \approx (\pi, 0)^{\text{t}}$	100	-0.9353221508		$1.81 \cdot 10^{-4}$
	1000	-0.9354607418	-0.9355033787	$4.26 \cdot 10^{-5}$
	10000	-0.9354621405		$4.12 \cdot 10^{-5}$
$\mathbf{k} \approx (\pi, \pi)^{\text{t}}$	100	-2.645886184		$5.30 \cdot 10^{-4}$
	1000	-2.645886532	-2.646416184	$5.30 \cdot 10^{-4}$
	10000	-2.645886532		$5.30 \cdot 10^{-4}$

Table 3.2 Comparison of the numerical solution $\epsilon_{\pi}^{\text{num}}(\mathbf{k})$ obtained with the help of the MATHEMATICA program H.3 and the three shell summation approximation of $\epsilon_{\pi}(\mathbf{k})$. For $\mathbf{k} = \mathbf{0}$ we actually need 10 000 summands in order to reach the accuracy of the three shell summation approximation, whereas on the edges of the first Brillouin zone $\mathbf{k} = (\pi, 0)^{\text{t}}$ and $\mathbf{k} = (\pi, \pi)^{\text{t}}$ only 100 summands suffices to reach the accuracy of approximately 10^{-4} .

$$\begin{aligned}
& + C_{\pi} \left(\left| \begin{pmatrix} 0 \\ 4\pi \end{pmatrix} + \mathbf{k} \right| \right) + C_{\pi} \left(\left| \begin{pmatrix} -4\pi \\ 0 \end{pmatrix} + \mathbf{k} \right| \right) + C_{\pi} \left(\left| \begin{pmatrix} 0 \\ -4\pi \end{pmatrix} + \mathbf{k} \right| \right) \\
& + 2A(1) (\cos k_x + \cos k_y) + 4A \left(\sqrt{2} \right) \cos k_x \cos k_y \\
& + 2A(2) \left[\cos(2k_x) + \cos(2k_y) \right] - \frac{4\pi}{3}
\end{aligned} \tag{3.59}$$

The graph of $\epsilon_{\eta=\pi}(\mathbf{k})$ is shown in Figure 3.5 for the first Brillouin zone. A numerical solution is generated with the help of the MATHEMATICA program H.3 by solving

$$\epsilon(\mathbf{k}) = \sum_{n \neq 0}^N \frac{e^{-i\mathbf{k} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} \approx \sum_{R_x=-100}^{100} \sum_{R_y=-100}^{100} \frac{\cos(k_x R_x + k_y R_y)}{\sqrt{(R_x)^2 + (R_y)^2}^3} \tag{3.60}$$

Due to the summation over complete shells the exponential function can be rewritten as a pure cos-function because in the series expansion of $e^{-i\mathbf{k} \cdot \mathbf{R}_n}$ all terms with a product of odd numbers of \mathbf{R}_n will vanish

$$e^{-i\mathbf{k} \cdot \mathbf{R}_n} = \sum_{l=0}^{\infty} \frac{(-i\mathbf{k} \cdot \mathbf{R}_n)^l}{l!} = \underbrace{\sum_{l'=0}^{\infty} \frac{(-i\mathbf{k} \cdot \mathbf{R}_n)^{2l'+1}}{(2l'+1)!}}_{=0} + \underbrace{\sum_{l=0}^{\infty} \frac{(-i\mathbf{k} \cdot \mathbf{R}_n)^{2l}}{(2l)!}}_{=\cos(\mathbf{k} \cdot \mathbf{R}_n)} \tag{3.61}$$

In Table 3.2 the solution of this program is compared to the analytical solution in the third shell approximation given in equation (3.59). As one can see the error in the three shell approximation

is of the order of 10^{-4} which is very impressive considering the fact that only 12 summands are taken into account in each sum over direct and reciprocal space, respectively, whereas at the edges of the first Brillouin zone at least 100 summands suffices and for the origin in \mathbf{k} -space we even need at least 10 000.

3.3.2 Ferromagnetic Spin Wave Excitations

Starting with the following spin- $1/2$ XXZ model

$$\mathcal{H} = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(J_{\perp} \frac{\mathcal{S}_{ix}\mathcal{S}_{jx} + \mathcal{S}_{iy}\mathcal{S}_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J_z' \frac{\mathcal{S}_{iz}\mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (3.62)$$

with

$$J_{\perp} = \frac{2 \left(d_0^{\uparrow\downarrow} \right)^2}{\hbar^2 a^3} \quad J_z = \frac{\left(d_0^{\uparrow\uparrow} - d_0^{\downarrow\downarrow} \right)^2}{\hbar^2 a^3} \quad (3.63)$$

the ferromagnetic case is described by $J_{\perp} < 0$ and $J_z < 0$ or the larger one of the two coupling constants being negative, i.e. $J_z < 0$ and $|J_z| > J_{\perp} > 0$ or $J_{\perp} < 0$ and $|J_{\perp}| > J_z > 0$. These cases corresponds to ϑ in the range of $[3\pi/4, 7\pi/4]$ and therefore in the mean field approximation one finds parallel aligned spins at each lattice site pointing in the same direction. As we will see later on (see Section 3.5 “Phase Diagram of the XXZ-Model”) this is only true for nearest neighbor interactions, whereas in the case of slowly decaying interactions or long-range interactions, the ferromagnetic domain is extended. Introducing the $SU(2)$ raising and lowering operators

$$\begin{aligned} \mathcal{S}_+ &= \mathcal{S}_x + i\mathcal{S}_y & \mathcal{S}_x &= \frac{1}{2}(\mathcal{S}_+ + \mathcal{S}_-) \\ \mathcal{S}_- &= \mathcal{S}_x - i\mathcal{S}_y & \mathcal{S}_y &= -\frac{i}{2}(\mathcal{S}_+ - \mathcal{S}_-) \end{aligned} \quad (3.64)$$

$$\hookrightarrow \mathcal{S}_{ix}\mathcal{S}_{jx} + \mathcal{S}_{iy}\mathcal{S}_{jy} = \frac{1}{2}(\mathcal{S}_{i+}\mathcal{S}_{j-} + \mathcal{S}_{i-}\mathcal{S}_{j+}) \quad (3.65)$$

(3.62) can be rewritten to

$$\mathcal{H}^F = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{J_{\perp}}{2} \frac{\mathcal{S}_{i+}\mathcal{S}_{j-} + \mathcal{S}_{i-}\mathcal{S}_{j+}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J_z \frac{\mathcal{S}_{iz}\mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (3.66)$$

First we will only consider the case of a strong in-axis coupling, i.e. $J_z < 0$ and $|J_z| > |J_{\perp}|$. Following [15] we now perform a Holstein-Primakoff transformation

$$\mathcal{S}_{i+} = \hbar\sqrt{2S}\varphi(n_i)a_i \quad \mathcal{S}_{i-} = \hbar\sqrt{2S}a_i^{\dagger}\varphi(n_i) \quad \mathcal{S}_{iz} = \hbar(S - n_i) \quad (3.67)$$

where φ is a function of the $n_i = a_i^{\dagger}a_i$ operator which can be expanded for different S . For $S = 1/2$ a rigorous solution is

$$\varphi(n_i) = 1 - n_i \quad (3.68)$$

Since we are only interested in non-interaction spin waves we will make the approximation $\varphi(\mathbf{n}_i) \approx 1$. The ground state of the ferromagnet is described by an alignment of all spins in z -direction:

$$\{|\uparrow\rangle_{i_1}, |\uparrow\rangle_{i_2}, \dots, |\uparrow\rangle_{i_N}\} \equiv |\uparrow\uparrow\uparrow \dots\rangle \quad \text{or} \quad \{|\downarrow\rangle_{i_1}, |\downarrow\rangle_{i_2}, \dots, |\downarrow\rangle_{i_N}\} \equiv |\downarrow\downarrow\downarrow \dots\rangle \quad (3.69)$$

The operator n_i can be considered as a “spin deviation” operator from the ferromagnetic ordered ground state and the total spin deviation can be defined as

$$n_i = S - \frac{S_{iz}}{\hbar} \quad \mathcal{N} = \sum_{i=1}^N n_i = NS - \frac{1}{\hbar} \sum_{i=1}^N S_{iz} \quad (3.70)$$

Using the commutation relations of spin operators

$$\begin{aligned} [S_{i\alpha}, S_{j\beta}] &= i\hbar\epsilon_{\alpha\beta\gamma}\delta_{ij}S_{j\gamma} & \alpha\beta\gamma &= x, y, z \\ \hookrightarrow [S_{iz}, S_{j\pm}] &= \pm\hbar\delta_{ij}S_{j\pm} & [S_{i+}, S_{j-}] &= 2\hbar\delta_{ij}S_{jz} \end{aligned} \quad (3.71)$$

one can derive the bosonic commutation relations of a_i and a_i^\dagger

$$[a_i, a_j^\dagger] = \delta_{ij} \quad [a_i, a_j] = 0 = [a_i^\dagger, a_j^\dagger] \quad (3.72)$$

Thus a_i and a_i^\dagger can be considered as annihilation and creation operators of spin deviations, i.e. harmonic excitations “above” the ferromagnetic ground state, e.g. $S_-|\uparrow\rangle = |\downarrow\rangle$. These low-lying collective excitation can be thought of as bosonic quasi-particles. However this concept is only well defined for spin waves which are spatial far apart and ideally non-interacting. Later on we will work in the spin wave approximation given in (3.78) where we can safely assume the magnon concept in analogy to phonons. Applying the Holstein–Primakoff transformation to the Hamiltonian \mathcal{H}^F (3.66) and setting $J' = \hbar^2 J$

$$\begin{aligned} \mathcal{H}^F &= \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \frac{J_\perp}{2} \frac{\hbar^2 2S}{|\mathbf{R}_i - \mathbf{R}_j|^3} \left(\varphi(\mathbf{n}_i) a_i a_j^\dagger \varphi(\mathbf{n}_j) + a_i^\dagger \varphi(\mathbf{n}_i) \varphi(\mathbf{n}_j) a_j \right) \right. \\ &\quad \left. + J_z \frac{\hbar^2}{|\mathbf{R}_i - \mathbf{R}_j|^3} (S - n_i)(S - n_j) \right\} \\ &= SJ'_\perp \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\varphi(\mathbf{n}_i) a_i a_j^\dagger \varphi(\mathbf{n}_j) + a_i^\dagger \varphi(\mathbf{n}_i) \varphi(\mathbf{n}_j) a_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) + J'_z \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{S^2 - S(n_j + n_i) + n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \end{aligned} \quad (3.73)$$

Due to the translational invariance of the square lattice, the second term can be simplified by summing over the free subscript respectively

$$g_i^F = \sum_{j=1}^N \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3} = \sum_{\mathbf{R}_n \neq 0}^{R_N} \frac{1}{|\mathbf{R}_n|^3} = g^F \quad (3.74)$$

$$g_j^F = \sum_{i=1}^N \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3} = \sum_{\mathbf{R}_n \neq 0}^{R_N} \frac{1}{|\mathbf{R}_n|^3} = g^F \quad (3.75)$$

Using the above relation the second term of (3.73) can be cast into

$$J'_z \left(S^2 \underbrace{\sum_{i=1}^N g_i^F}_{\stackrel{(3.75)}{=} N g^F} - S \underbrace{\left(\sum_{i=1}^N g_i^F n_i + \sum_{i=1}^N g_j^F n_j \right)}_{\stackrel{(3.75)}{=} 2g^F \sum_{i=1}^N n_i} + \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (3.76)$$

we end up with

$$\begin{aligned} \mathcal{H}^F = S J'_\perp \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\varphi(\mathbf{n}_i) \mathbf{a}_i \mathbf{a}_j^\dagger \varphi(\mathbf{n}_j) + \mathbf{a}_i^\dagger \varphi(\mathbf{n}_i) \varphi(\mathbf{n}_j) \mathbf{a}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \\ + J'_z \left(S^2 N g^F - 2S g^F \sum_{i=1}^N n_i + \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \end{aligned} \quad (3.77)$$

Since we do not want to have interactions between the spin waves we will apply the following approximation

$$\varphi(\mathbf{n}_i) \approx 1 \quad n_i n_j \approx 0 \quad (3.78)$$

to equation (3.77) yielding

$$\mathcal{H}^{\text{FSW}} = S J'_\perp \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\mathbf{a}_i \mathbf{a}_j^\dagger + \mathbf{a}_i^\dagger \mathbf{a}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J'_z \left(S^2 N g^F - 2S g^F \sum_{i=1}^N n_i \right) \quad (3.79)$$

By applying a unitary canonical transformation (a so called Bogoliubov transformation)

$$\mathbf{a}_i = \frac{1}{\sqrt{2}} (\mathcal{Q}_i + i\mathcal{P}_i) \quad \mathbf{a}_i^\dagger = \frac{1}{\sqrt{2}} (\mathcal{Q}_i - i\mathcal{P}_i) \quad (3.80)$$

\mathbf{a}_i and \mathbf{a}_i^\dagger can be written in terms of canonical conjugate operators \mathcal{Q}_i and \mathcal{P}_i satisfying

$$[\mathcal{Q}_i, \mathcal{P}_j] = i\delta_{ij} \quad \mathcal{Q}_i^\dagger = \mathcal{Q}_i \quad \text{and} \quad \mathcal{P}_i^\dagger = \mathcal{P}_i \quad (3.81)$$

Thus

$$\begin{aligned} \mathbf{a}_i \mathbf{a}_j^\dagger + \mathbf{a}_i^\dagger \mathbf{a}_j &= \frac{1}{2} (\mathcal{Q}_i + i\mathcal{P}_i) (\mathcal{Q}_j - i\mathcal{P}_j) + \frac{1}{2} (\mathcal{Q}_i - i\mathcal{P}_i) (\mathcal{Q}_j + i\mathcal{P}_j) \\ &= \frac{1}{2} [\mathcal{Q}_i \mathcal{Q}_j + \mathcal{P}_i \mathcal{P}_j - i(\mathcal{Q}_i \mathcal{P}_j - \mathcal{P}_i \mathcal{Q}_j) + \mathcal{Q}_i \mathcal{Q}_j + \mathcal{P}_i \mathcal{P}_j + i(\mathcal{Q}_i \mathcal{P}_j - \mathcal{P}_i \mathcal{Q}_j)] \\ &= \mathcal{Q}_i \mathcal{Q}_j + \mathcal{P}_i \mathcal{P}_j \end{aligned} \quad (3.82)$$

and analogous

$$\begin{aligned} n_i &= a_i^\dagger a_i = \frac{1}{2} (Q_i - iP_i)(Q_i + iP_i) \\ &= \frac{1}{2} [Q_i^2 + P_i^2 + i \underbrace{(Q_i P_i - P_i Q_i)}_{[Q_i, P_i] \stackrel{(3.81)}{=} i\mathbb{1}}] = \frac{1}{2} [Q_i^2 + P_i^2 - \mathbb{1}] \end{aligned} \quad (3.83)$$

Inserting equation (3.82) and (3.83) in the spin wave Hamiltonian (3.79) gives rise to

$$\begin{aligned} \mathcal{H}^{\text{FSW}} &= S J'_\perp \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{Q_i Q_j + P_i P_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J'_z \left(S^2 N g^F - S g^F \sum_{i=1}^N (Q_i^2 + P_i^2) + S g^F \underbrace{\sum_{i=1}^N \mathbb{1}}_{=N} \right) \\ &= N g^F J'_z S(S+1) + S J'_\perp \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{Q_i Q_j + P_i P_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} - J'_z S g^F \sum_{i=1}^N (Q_i^2 + P_i^2) \end{aligned} \quad (3.84)$$

A Fourier transformation will diagonalize the Hamiltonian \mathcal{H}^{FSW} quadratic in the canonical operators Q_i and P_i . The commutation relations for canonical conjugate operators must be preserved under the Fourier transformation. This can be achieved by defining the Fourier transformation in the following way

$$Q_i = \frac{1}{N} \sum_{\mathbf{k}} Q_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i} \quad P_i = \frac{1}{N} \sum_{\mathbf{k}} P_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_i} \quad (3.85)$$

Expressing the commutator in terms of the Fourier transforms

$$\begin{aligned} [Q_i, P_j] &= \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} [Q_{\mathbf{k}}, P_{\mathbf{k}'}] e^{i\mathbf{k} \cdot \mathbf{R}_i} e^{-i\mathbf{k}' \cdot \mathbf{R}_j} = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} \delta_{\mathbf{k}\mathbf{k}'} e^{i\mathbf{k} \cdot \mathbf{R}_i} e^{-i\mathbf{k}' \cdot \mathbf{R}_j} \\ &= \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)} = \delta(\mathbf{R}_i - \mathbf{R}_j) = \delta_{ij} \end{aligned} \quad (3.86)$$

we find the expected commutation relations of canonical conjugated operators. A more detailed discussion of the different variants of Fourier transformations and their physical applications can be found in Appendix F. For Fourier transforms of hermitian operators there exists also a symmetry in Fourier space

$$\begin{aligned} Q_i &= Q_i^\dagger = \frac{1}{N} \sum_{\mathbf{k}} Q_{\mathbf{k}}^\dagger e^{-i\mathbf{k} \cdot \mathbf{R}_i} \stackrel{!}{=} \frac{1}{N} \sum_{\mathbf{k}} Q_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i} \\ &\Rightarrow Q_{\mathbf{k}}^\dagger = Q_{-\mathbf{k}} = Q_{\mathbf{k}} \end{aligned} \quad (3.87)$$

Applying the Fourier transformation to the first sum of \mathcal{H}^{FSW} yields

$$\sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{Q_i Q_j + P_i P_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} = \sum_{\mathbf{R}_i, \mathbf{R}_j} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \left(\frac{1}{N} Q_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i} \frac{1}{N} Q_{\mathbf{k}'} e^{i\mathbf{k}' \cdot \mathbf{R}_j} \frac{1}{N} V(\mathbf{q}) e^{i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \right)$$

$$\begin{aligned}
& + \frac{1}{N^2} \mathcal{P}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}'} V(\mathbf{q}) e^{-i\mathbf{k} \cdot \mathbf{R}_i} e^{-i\mathbf{k}' \cdot \mathbf{R}_j} e^{i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \Big) \\
& = \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \left(\mathcal{Q}_{\mathbf{k}} \mathcal{Q}_{\mathbf{k}'} V(\mathbf{q}) \underbrace{\sum_{\mathbf{R}_i} \frac{1}{N} e^{-i(-\mathbf{k}-\mathbf{q}) \cdot \mathbf{R}_i}}_{\delta(-\mathbf{k}-\mathbf{q})} \underbrace{\sum_{\mathbf{R}_j} \frac{1}{N} e^{-i(-\mathbf{k}'+\mathbf{q}) \cdot \mathbf{R}_j}}_{\delta(-\mathbf{k}'+\mathbf{q})} \right. \\
& \quad \left. + \mathcal{P}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}'} V(\mathbf{q}) \delta(\mathbf{k}-\mathbf{q}) \delta(\mathbf{k}'+\mathbf{q}) \right) \\
& = \sum_{\mathbf{k}, \mathbf{k}'} \left(\mathcal{Q}_{\mathbf{k}} \mathcal{Q}_{\mathbf{k}'} V(\mathbf{k}') \delta(-\mathbf{k}-\mathbf{k}') + \mathcal{P}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}'} V(-\mathbf{k}') \delta(\mathbf{k}+\mathbf{k}') \right) \\
& = \sum_{\mathbf{k}} \left(\mathcal{Q}_{\mathbf{k}} \mathcal{Q}_{-\mathbf{k}} V(-\mathbf{k}) + \mathcal{P}_{\mathbf{k}} \mathcal{P}_{-\mathbf{k}} V(\mathbf{k}) \right) \tag{3.88}
\end{aligned}$$

Due to the time reversal symmetry of the dipole-dipole interaction, the Fourier transform $V(\mathbf{k})$ is symmetric and we can write $V(\mathbf{k}) = V(-\mathbf{k}) = \epsilon(\mathbf{k})$ where $\epsilon(\mathbf{k})$ is defined and calculated in Subsection 3.3.1. Equation (3.88) can be simplified with the help of (3.87) to

$$\sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\mathcal{Q}_i \mathcal{Q}_j + \mathcal{P}_i \mathcal{P}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} = \sum_{\mathbf{k}} (\mathcal{Q}_{\mathbf{k}}^2 + \mathcal{P}_{\mathbf{k}}^2) \epsilon(\mathbf{k}) \tag{3.89}$$

The second sum of \mathcal{H}^{FSW} can be Fourier transformed to

$$\begin{aligned}
\sum_{i=1}^N \mathcal{Q}_i^2 & = \sum_{i=1}^N \mathcal{Q}_i^\dagger \mathcal{Q}_i = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} \sum_{\mathbf{R}_i} \mathcal{Q}_{\mathbf{k}} \mathcal{Q}_{\mathbf{k}'} e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}_i} \\
& = \sum_{\mathbf{k}, \mathbf{k}'} \mathcal{Q}_{\mathbf{k}} \mathcal{Q}_{\mathbf{k}'} \delta(\mathbf{k}-\mathbf{k}') = \sum_{\mathbf{k}} \mathcal{Q}_{\mathbf{k}}^2 \\
\hookrightarrow \sum_{i=1}^N (\mathcal{Q}_i^2 + \mathcal{P}_i^2) & = \sum_{\mathbf{k}} (\mathcal{Q}_{\mathbf{k}}^2 + \mathcal{P}_{\mathbf{k}}^2) \tag{3.90}
\end{aligned}$$

Putting everything together, the completely Fourier transformed spin wave Hamiltonian is given as

$$\begin{aligned}
\widetilde{\mathcal{H}}^{\text{FSW}} & = N g^{\text{F}} J'_z S(S+1) + S J'_\perp \sum_{\mathbf{k}} \epsilon(\mathbf{k}) (\mathcal{Q}_{\mathbf{k}}^2 + \mathcal{P}_{\mathbf{k}}^2) - J'_z S g^{\text{F}} \sum_{\mathbf{k}} (\mathcal{Q}_{\mathbf{k}}^2 + \mathcal{P}_{\mathbf{k}}^2) \\
& = N g^{\text{F}} J'_z S(S+1) - g^{\text{F}} J'_z S \sum_{\mathbf{k}} \left[\left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k})}{g^{\text{F}}} \right) \mathcal{Q}_{\mathbf{k}}^2 + \left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k})}{g^{\text{F}}} \right) \mathcal{P}_{\mathbf{k}}^2 \right] \tag{3.91}
\end{aligned}$$

This Hamiltonian can be compared to the harmonic oscillator

$$\frac{1}{2m} \mathcal{P}^2 + \frac{m\omega^2}{2} \mathcal{Q}^2 = \hbar\omega \left(n + \frac{1}{2} \right) \tag{3.92}$$

expressed in creation and annihilation operators

$$Q = \sqrt{\frac{\hbar}{2m\omega}} (\mathcal{A}^\dagger + \mathcal{A}) \quad \mathcal{P} = i\sqrt{\frac{\hbar m\omega}{2}} (\mathcal{A}^\dagger - \mathcal{A}) \quad (3.93)$$

with the canonical commutation relation $[Q, \mathcal{P}] = i\hbar\mathbb{1}$ and the occupation number operator $\mathcal{N} = \mathcal{A}^\dagger \mathcal{A}$. In order to compare the harmonic oscillator to $\widetilde{\mathcal{H}}^{\text{FSW}}$ we need to rescale the operators in equation (3.92) with $1/\sqrt{\hbar}$

$$\frac{1}{m} \mathcal{P}^2 + m\omega^2 Q^2 = \omega(2n + 1) \quad (3.94)$$

and we finally obtain

$$\frac{1}{m} = \left(1 - \frac{J'_\perp \epsilon(\mathbf{k})}{J'_z g^{\text{F}}}\right) \quad m\omega^2 = \left(1 - \frac{J'_\perp \epsilon(\mathbf{k})}{J'_z g^{\text{F}}}\right) \quad (3.95)$$

so that

$$\frac{1}{m} m\omega^2 = \omega^2 = \left(1 - \frac{J'_\perp \epsilon(\mathbf{k})}{J'_z g^{\text{F}}}\right)^2 \quad \hookrightarrow \quad \omega = \left(1 - \frac{J'_\perp \epsilon(\mathbf{k})}{J'_z g^{\text{F}}}\right) \quad (3.96)$$

We can split the spin wave Hamiltonian in a ground state energy part $E_{0\text{F}}^z$ and a harmonic excitation with dispersion relation $\omega_{\text{F}}^z(\mathbf{k})$. The superscript z indicates that we derived the linear spin waves starting from the ferromagnetic ordered state in z-direction.

$$\widetilde{\mathcal{H}}^{\text{FSW}} = E_{0\text{F}}^z + \sum_{\mathbf{k}} \omega_{\text{F}}^z(\mathbf{k}) n_{\mathbf{k}} \quad (3.97)$$

with

$$\begin{aligned} \omega_{\text{F}}^z(\mathbf{k}) &= 2S (J'_\perp \epsilon(\mathbf{k}) - J'_z g^{\text{F}}) \\ &= 2J'S (\sin \vartheta \epsilon(\mathbf{k}) - \cos \vartheta \epsilon(\mathbf{0})) \end{aligned} \quad (3.98)$$

where we have expressed J'_\perp and J'_z in terms of the coupling constant angle ϑ . The ferromagnetic lattice factor g^{F} can be expressed with the help of the Fourier transform of the interaction potential (3.34)

$$\epsilon(\mathbf{k}) = \sum_{\mathbf{R}_n \neq \mathbf{0}}^{R_N} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} \quad \hookrightarrow \quad g^{\text{F}} = \sum_{\mathbf{R}_n \neq \mathbf{0}}^{R_N} \frac{e^{i\mathbf{0} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} = \epsilon(\mathbf{0}) \approx 9.0336 \quad (3.99)$$

Furthermore the ground state energy can be simplified to

$$E_{0\text{F}}^z = N g^{\text{F}} J'_z S(S+1) + \frac{1}{2} \sum_{\mathbf{k}} \omega_{\text{F}}^z(\mathbf{k}) \quad (3.100)$$

$$= N g^{\text{F}} J'_z S(S+1) + S \sum_{\mathbf{k}} \left(J'_\perp \epsilon(\mathbf{k}) - J'_z g^{\text{F}} \right) \quad (3.101)$$

$$= N g^{\text{F}} J'_z S(S+1) + \mathcal{F}^{-1}[\epsilon(\mathbf{k})](\mathbf{0}) - N g^{\text{F}} J'_z S = N g^{\text{F}} J'_z S^2 \quad (3.102)$$

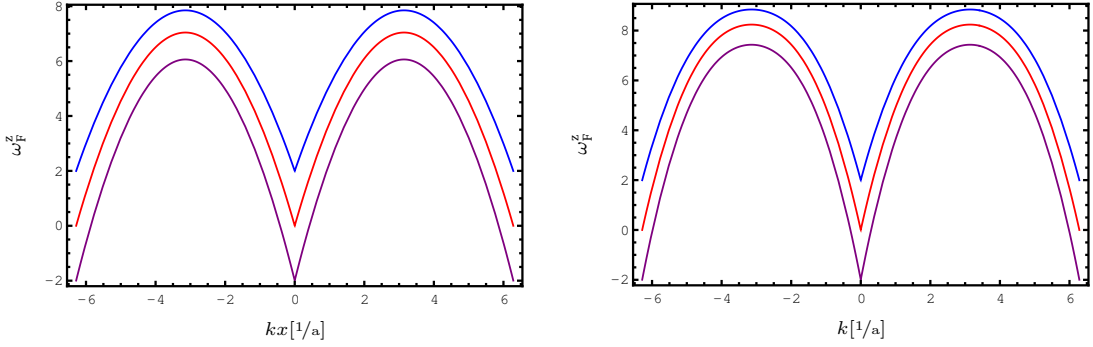


Figure 3.6 Dispersion relation ω_F^z for different $\vartheta = 1.2\pi, 1.25\pi, 1.3\pi$ in k_x -direction (left picture) and $(k_x, k_x)^t$ -direction (right picture). The dispersion relation is becoming negative for $\vartheta > 5\pi/4$ or $|J'_\perp| > |J'_z|$ and $J'_\perp < 0$ indicating the instability of the in-axis ground state.

The first sum corresponds to the inverse Fourier transformation evaluated at the real space point $\mathbf{R} = \mathbf{0}$. In order to define the Fourier transformation of the $1/R^3$ properly, the summand of $\mathbf{R} = \mathbf{0}$ was omitted by setting it to zero in the first place and therefore the first sum is vanishing. In the first Brillouin zone there exists precisely the same number of \mathbf{k} vectors as there are unit cells in the direct lattice and hence for a primitive lattice the number of \mathbf{k} vectors in the first Brillouin zone equals the number of particles in the real lattice. Additionally the summands in the second sum are not depending on \mathbf{k} and so it can be replaced by the sum over all N molecules in real space. For this reason the second sum yields just N , leading to the simplified ground state energy stated in (3.102). Since $J'_z < 0$, the ground state energy remains negative in the entire ferromagnetic domain with spins aligned in z -direction, i.e. in the range of $\vartheta \in [\approx 3\pi/5, 5\pi/4]$. In Section 3.5 we will look at the energy for different ground states and investigate the phase transition between these ground states in detail.

The dispersion relation is positive in the range $\approx 3\pi/5$ to $5\pi/4$ of the angle ϑ , whereas a negative dispersion relation indicates an instability. In fact for $\vartheta > 5\pi/4$, the ferromagnetic ground state $|\mathbf{z}\text{-F}\rangle$ aligned in z -direction is destroyed by the strong in-plane coupling $|J'_\perp| > |J'_z|$. In this case we need to start with an in-plane ferromagnetically ordered ground state, see Subsection 3.3.5.

Considering only nearest neighbor interaction one has to replace g^F with the coordination number of the lattice z – e.g. for a two dimensional square lattice $z = 4$. In this case the Fourier transformation can be reduced to a one shell summation

$$\epsilon(\mathbf{k})^{\text{NN}} = \sum_{\rho} \frac{e^{i\mathbf{k}\cdot\rho}}{|\rho|^3} = 2(\cos k_x + \cos k_y) \quad (3.103)$$

where ρ points to the nearest neighbor of a lattice point. The spin wave Hamiltonian in this case reads

$$\widetilde{\mathcal{H}}^{\text{FSW-NN}} = NzJ'_zS^2 + 2S \sum_{\mathbf{k}} (2J'_\perp (\cos k_x + \cos k_y) - J'_z z n_{\mathbf{k}}) \quad (3.104)$$

Intuitively it is clear, that the ground state energy of the slowly decaying $1/R^3$ -interaction is larger than the ground state energy of the nearest neighbor interaction, because the $1/R^3$ -interaction leads to a higher number of interacting spins as in the nearest neighbor case. The

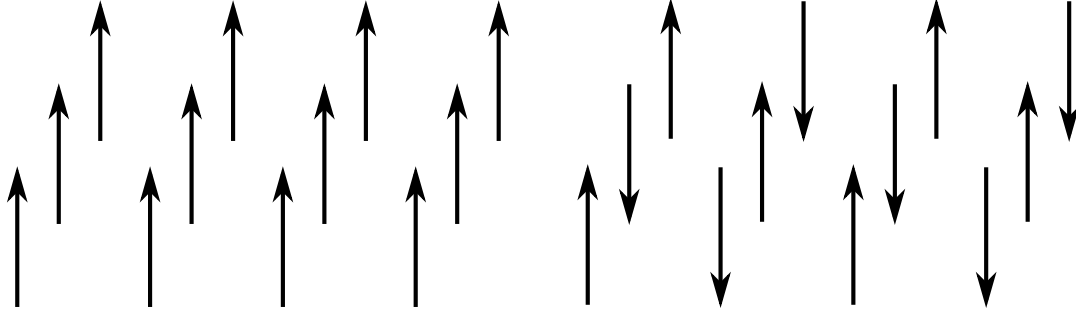


Figure 3.7 Left Panel: Ferromagnetically ordered ground state.
Right Panel: Antiferromagnetically ordered ground state

ferromagnetic lattice factor g^F can be considered as the number of spins that are interacting with a spin on an individual lattice site, whereas for the nearest neighbor interaction this factor is just the coordination number $z < g^F$.

3.3.3 Antiferromagnetic Spin Waves

The antiferromagnetic spin- $1/2$ XXZ model is described by the Hamiltonian (3.66) but with positive coupling constants $J_z > 0$ and $J_\perp > 0$ or at least the larger one of the two coupling constants being positive, i.e. $J_z > |J_\perp| > 0$ and $J_\perp > |J_z| > 0$. As we will see later on, this is only true for nearest neighbor interactions. For the slowly decaying $1/R^3$ -interaction we noted in the beginning of Subsection 3.3.2, that the ferromagnetic domain is extended and therefore the antiferromagnetic domain must be reduced to a smaller range.

$$\mathcal{H}^{\text{AF}} = \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{J_\perp}{2} \frac{S_{i+} S_{j-} + S_{i-} S_{j+}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J_z \frac{S_{i_z} S_{j_z}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (3.105)$$

In case of an in-axis antiferromagnet, we assume that the ground state is Néel ordered in the z -direction, so that the two dimensional square lattice can be divided in two sublattices, one with all spins pointing in $+z$ -direction which we will call the \bullet -sublattice and one with all spins pointing in $-z$ -direction which will be called the \circ -sublattice. The Néel ordered ground state, as illustrated in Figure 3.7 of the antiferromagnet is given as

$$\{|\uparrow\rangle_{i_1}, |\downarrow\rangle_{i_2}, \dots, |\uparrow\rangle_{i_N}\} \equiv |\uparrow\downarrow\uparrow \dots\rangle \quad \text{or} \quad \{|\downarrow\rangle_{i_1}, |\uparrow\rangle_{i_2}, \dots, |\downarrow\rangle_{i_N}\} \equiv |\downarrow\uparrow\downarrow \dots\rangle \quad (3.106)$$

These two ground states break the symmetry of the antiferromagnetic spin- $1/2$ XXZ model so we will expect gaped dispersion relation. The \bullet -sublattice possesses the ground state

$$\{|\uparrow\rangle_{i_{1+}}, |\uparrow\rangle_{i_{2+}}, \dots, |\uparrow\rangle_{i_{N+}}\} \quad (3.107)$$

whereas the ground state of the \circ -sublattice is given as

$$\{|\downarrow\rangle_{i_{1-}}, |\downarrow\rangle_{i_{2-}}, \dots, |\downarrow\rangle_{i_{N-}}\} \quad (3.108)$$

The subscript “+” denotes indices running over the ●-sublattice and “-” denotes indices of the ○-sublattice, respectively. Therefore the sum in (3.105) can be replaced by

$$\begin{aligned}
\sum_{\substack{i,j=1 \\ i \neq j}}^N &= \sum_{i_+=1}^{N/2} \left(\sum_{j=1}^N \right) + \sum_{i_-=1}^{N/2} \left(\sum_{j=1}^N \right) \\
&= \sum_{i_+=1}^{N/2} \left(\sum_{j_+=1}^{N/2} + \sum_{j_-=1}^{N/2} \right) + \sum_{i_-=1}^{N/2} \left(\sum_{j_+=1}^{N/2} + \sum_{j_-=1}^{N/2} \right) \\
&= \sum_{\substack{i_+,j_+=1 \\ i_+ \neq j_+}}^{N/2} + \sum_{\substack{i_+,j_-=1}}^{N/2} + \sum_{\substack{i_-,j_+=1}}^{N/2} + \sum_{\substack{i_-,j_-=1 \\ i_- \neq j_-}}^{N/2} \tag{3.109}
\end{aligned}$$

Now the Holstein–Primakoff transformation for the spin operators S^+ , S^- and S^z must be applied to both sublattices

$$\begin{aligned}
S_{i_+}^+ &= \hbar\sqrt{2S}\varphi(n_{i_+})a_{i_+} & S_{i_-}^+ &= \hbar\sqrt{2S}b_{i_-}^\dagger\varphi(n_{i_-}) \\
S_{i_+}^- &= \hbar\sqrt{2S}a_{i_+}^\dagger\varphi(n_{i_+}) & S_{i_-}^- &= \hbar\sqrt{2S}\varphi(n_{i_-})b_{i_-} \\
S_{i_+}^z &= \hbar(S - n_{i_+}) & S_{i_-}^z &= \hbar(n_{i_-} - S)
\end{aligned} \tag{3.110}$$

For the ●-sublattice with ground state $|\uparrow\uparrow\uparrow\dots\rangle$ and excited state $S_{i_+}^- |\uparrow\rangle = |\downarrow\rangle$ we define the local “spin deviation”

$$n_{i_+} = S - \frac{S_{i_+}^z}{\hbar} = a_{i_+}^\dagger a_{i_+} \tag{3.111}$$

$a_{i_+}^\dagger$ and a_{i_+} are creation and annihilation operators on the ●-sublattice with

$$[a_{i_+}, a_{j_+}^\dagger] = \delta_{i_+j_+} \tag{3.112}$$

$$[a_{i_+}^\dagger, a_{j_+}^\dagger] = 0 = [a_{i_+}, a_{j_+}] \tag{3.113}$$

The meaning of the operators on the ○-sublattice with ground state $|\downarrow\downarrow\downarrow\dots\rangle$ and excited state $S_{i_-}^+ |\downarrow\rangle = |\uparrow\rangle$ are constructed in the same way. The local “spin deviation” reads

$$n_{i_-} = S + \frac{S_{i_-}^z}{\hbar} = b_{i_-}^\dagger b_{i_-} \tag{3.114}$$

$b_{i_-}^\dagger$ and b_{i_-} are creation and annihilation operators on the ○-sublattice with

$$[b_{i_-}, b_{j_-}^\dagger] = \delta_{i_-j_-} \tag{3.115}$$

$$[b_{i_-}^\dagger, b_{j_-}^\dagger] = 0 = [b_{i_-}, b_{j_-}] \tag{3.116}$$

Inserting the Holstein–Primakoff transformation (3.110) into the Hamiltonian (3.105) and splitting the total lattice into two sublattices (3.109) yields

$$\begin{aligned}
\mathcal{H}^{\text{AF}} = & \sum_{\substack{i_+, j_+=1 \\ i_+ \neq j_+}}^{N/2} \left\{ \frac{J_{\perp}}{2} \frac{\hbar^2 2S}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \left(\varphi(\mathbf{n}_{i_+}) \mathbf{a}_{i_+} \mathbf{a}_{j_+}^{\dagger} \varphi(\mathbf{n}_{j_+}) + \mathbf{a}_{i_+}^{\dagger} \varphi(\mathbf{n}_{i_+}) \varphi(\mathbf{n}_{j_+}) \mathbf{a}_{j_+} \right) \right. \\
& \left. + J_z \frac{\hbar^2}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \left[(S - n_{i_+})(S - n_{j_+}) \right] \right\} \\
& + \sum_{\substack{i_+, j_-=1 \\ i_+ \neq j_-}}^{N/2} \left\{ \frac{J_{\perp}}{2} \frac{\hbar^2 2S}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}^3|} \left(\varphi(\mathbf{n}_{i_+}) \mathbf{a}_{i_+} \varphi(\mathbf{n}_{j_-}) \mathbf{b}_{j_-} + \mathbf{a}_{i_+}^{\dagger} \varphi(\mathbf{n}_{i_+}) \mathbf{b}_{j_-}^{\dagger} \varphi(\mathbf{n}_{j_-}) \right) \right. \\
& \left. + J_z \frac{\hbar^2}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \left[(S - n_{i_+})(n_{j_-} - S) \right] \right\} \\
& + \sum_{\substack{i_-, j_+=1 \\ i_- \neq j_+}}^{N/2} \left\{ \frac{J_{\perp}}{2} \frac{\hbar^2 2S}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}^3|} \left(\mathbf{b}_{i_-}^{\dagger} \varphi(\mathbf{n}_{i_-}) \mathbf{a}_{j_+}^{\dagger} \varphi(\mathbf{n}_{j_+}) + \varphi(\mathbf{n}_{i_-}) \mathbf{b}_{j_-} \varphi(\mathbf{n}_{j_+}) \mathbf{a}_{j_+} \right) \right. \\
& \left. + J_z \frac{\hbar^2}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \left[(n_{i_-} - S)(S - n_{j_+}) \right] \right\} \\
& + \sum_{\substack{i_-, j_-=1 \\ i_- \neq j_-}}^{N/2} \left\{ \frac{J_{\perp}}{2} \frac{\hbar^2 2S}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}^3|} \left(\mathbf{b}_{i_-}^{\dagger} \varphi(\mathbf{n}_{i_-}) \varphi(\mathbf{n}_{j_-}) \mathbf{b}_{j_-} + \varphi(\mathbf{n}_{i_-}) \mathbf{b}_{i_-} \mathbf{b}_{j_-}^{\dagger} \varphi(\mathbf{n}_{j_-}) \right) \right. \\
& \left. + J_z \frac{\hbar^2}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \left[(n_{i_-} - S)(n_{j_-} - S) \right] \right\} \tag{3.117}
\end{aligned}$$

The J_z term can be further simplified due to the translational invariance of the \bullet - and the \circ -sublattice. We can define antiferromagnetic lattice factors in the same way as we did in the ferromagnetic case. The division in \bullet - and \circ -sublattices is shown in Figure 3.8. The sums

$$g_{i_+}^+ = \sum_{j_+} \frac{1}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} = g^{(++)} \quad g_{j_+}^+ = \sum_{i_+} \frac{1}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} = g^{(++)} \tag{3.118}$$

are both running over the \bullet -sublattice because the summation index i and j can be swapped. The four sums

$$g_{i_+}^- = \sum_{j_-} \frac{1}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} = g^{(-+)} \quad g_{i_-}^+ = \sum_{j_+} \frac{1}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} = g^{(+-)} \tag{3.119}$$

$$g_{j_+}^- = \sum_{i_-} \frac{1}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} = g^{(-+)} \quad g_{j_-}^+ = \sum_{i_+} \frac{1}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} = g^{(+-)} \tag{3.120}$$

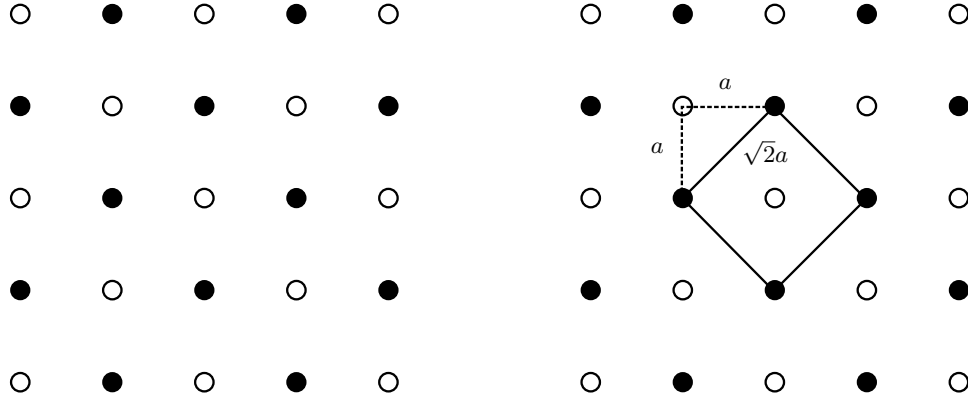


Figure 3.8 The Néel ordered ground state can be thought of as being composed of two ferromagnetically ordered sublattices ● and ○ with lattice spacing $\sqrt{2}a$.

start all on the ●-sublattice and run over the ○-sublattice and vice versa. So the four sums are belonging to the same class and we can write $g^{(-+)} = g^{(+ -)} = g^\ominus$ for the antiferromagnetic lattice factor connecting different sublattices. The last two combinations

$$g_{i_-}^- = \sum_{j_-} \frac{1}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} = g^{(--)} \quad g_{j_-}^+ = \sum_{i_-} \frac{1}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} = g^{(--) \quad (3.121)$$

are identical to the factor $g^{(++)}$ since the ○-sublattice is identical to the ●-sublattice. Therefore we can write for the sums over identical sublattices $g^{(++)} = g^{(--) = g^\oplus$. To compactify the notation we can write for the sum over all lattice points on identical sublattices $\sum_{\mathbf{n}_\oplus}$ with the vector \mathbf{n}_\oplus connecting identical sublattice points and for the sum starting from one sublattice running over the other sublattice $\sum_{\mathbf{n}_\ominus}$ with the vector \mathbf{n}_\ominus connecting the starting point with the other sublattice. Furthermore we define the complete antiferromagnetic lattice factor

$$g = g^\oplus - g^\ominus = \sum_{\mathbf{n}_\oplus} \frac{1}{|\mathbf{R}_{\mathbf{n}_\oplus}|^3} - \sum_{\mathbf{n}_\ominus} \frac{1}{|\mathbf{R}_{\mathbf{n}_\ominus}|^3} \quad (3.122)$$

All terms depending on $J'_z = \hbar^2 J_z$ can be simplified by summing over the respective free index

$$\begin{aligned} \mathcal{H}^{\text{AF}} = S J'_\perp (\dots) + S J'_z \left(S \frac{N}{2} g^\oplus - g^\oplus \sum_{j_+=1}^{N/2} n_{j_+} - g^\oplus \sum_{i_+=1}^{N/2} n_{i_+} + \sum_{\substack{i_+, j_+=1 \\ i_+ \neq j_+}}^{N/2} \frac{n_{i_+} n_{j_+}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \right. \\ \left. - S \frac{N}{2} g^\ominus + g^\ominus \sum_{j_-=1}^{N/2} n_{j_-} + g^\ominus \sum_{i_-=1}^{N/2} n_{i_-} - \sum_{i_+, j_-=1}^{N/2} \frac{n_{i_+} n_{j_-}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} \right. \\ \left. - S \frac{N}{2} g^\ominus + g^\ominus \sum_{j_+=1}^{N/2} n_{j_+} + g^\ominus \sum_{i_-=1}^{N/2} n_{i_-} - \sum_{i_-, j_+=1}^{N/2} \frac{n_{i_-} n_{j_+}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} \right) \end{aligned}$$

$$\begin{aligned}
& + S \frac{N}{2} g^\oplus - g^\ominus \sum_{j=1}^{N/2} n_{j-} - g^\oplus \sum_{i=1}^{N/2} n_{i-} + \sum_{\substack{i-,j-=1 \\ i- \neq j-}}^{N/2} \frac{n_{i-} n_{j-}}{|\mathbf{R}_{i-} - \mathbf{R}_{j-}|^3} \Bigg) \\
= & S J'_\perp (\dots) + S J'_z \left(\sum_{\substack{i+,j+=1 \\ i+ \neq j+}}^{N/2} \frac{n_{i+} n_{j+}}{|\mathbf{R}_{i+} - \mathbf{R}_{j+}|^3} - 2 \sum_{i+,j-=1}^{N/2} \frac{n_{i+} n_{j-}}{|\mathbf{R}_{i+} - \mathbf{R}_{j-}|^3} + \sum_{\substack{i-,j-=1 \\ i- \neq j-}}^{N/2} \frac{n_{i-} n_{j-}}{|\mathbf{R}_{i-} - \mathbf{R}_{j-}|^3} \right) \\
& + S J'_z \left[N(g^\oplus - g^\ominus) - 2 \underbrace{(g^\oplus - g^\ominus)}_{=g} \sum_{i+=1}^{N/2} n_{i+} - 2(g^\oplus - g^\ominus) \sum_{i-=1}^{N/2} n_{i-} \right] \tag{3.123}
\end{aligned}$$

Applying the spin-wave approximation $\varphi(n_{i_\pm}) \approx 1$ and $n_{i_\pm} n_{j_\pm} \approx 0$ to (3.123) gives rise to the following antiferromagnetic spin wave Hamiltonian

$$\begin{aligned}
\mathcal{H}^{\text{AFSW}} = & 4S J_\perp \left(\sum_{\substack{i+,j+=1 \\ i+ \neq j+}}^{N/2} \frac{a_{i+} a_{j+}^\dagger + a_{i+}^\dagger a_{j+}}{|\mathbf{R}_{i+} - \mathbf{R}_{j+}|^3} + \sum_{i+,j-=1}^{N/2} \frac{a_{i+} b_{j-} + a_{i+}^\dagger b_{j-}^\dagger}{|\mathbf{R}_{i+} - \mathbf{R}_{j-}|^3} \right. \\
& \left. + \sum_{i-,j+=1}^{N/2} \frac{b_{i-}^\dagger a_{j+}^\dagger + b_{i-} a_{j+}}{|\mathbf{R}_{i-} - \mathbf{R}_{j+}|^3} + \sum_{\substack{i-,j-=1 \\ i- \neq j-}}^{N/2} \frac{b_{i-}^\dagger b_{j-} + b_{i-} b_{j-}^\dagger}{|\mathbf{R}_{i-} - \mathbf{R}_{j-}|^3} \right) \\
& + 4J_z \left[S^2 N g - 2Sg \left(\sum_{i+=1}^{N/2} n_{i+} + \sum_{i-=1}^{N/2} n_{i-} \right) \right] \tag{3.124}
\end{aligned}$$

As a next step we apply the unitary transformation

$$\begin{aligned}
a_{i+} &= \frac{1}{\sqrt{2}} (Q_{i+} + iP_{i+}) & b_{i-} &= \frac{1}{\sqrt{2}} (\mathcal{R}_{i+} + iS_{i+}) \\
a_{i+}^\dagger &= \frac{1}{\sqrt{2}} (Q_{i+} - iP_{i+}) & b_{i-}^\dagger &= \frac{1}{\sqrt{2}} (\mathcal{R}_{i+} - iS_{i+})
\end{aligned} \tag{3.125}$$

with the canonical conjugate, hermitian operators Q_{i+} , P_{i+} and \mathcal{R}_{i-} , S_{i-} to $\mathcal{H}^{\text{AFSW}}$

$$\begin{aligned}
a_{i+} a_{j+}^\dagger + a_{i+}^\dagger a_{j+} &= \frac{1}{2} (Q_{i+} + iP_{i+}) (Q_{j+} - iP_{j+}) + \frac{1}{2} (Q_{i+} - iP_{i+}) (Q_{j+} + iP_{j+}) \\
&= \frac{1}{2} [Q_{i+} Q_{j+} + P_{i+} P_{j+} - i(Q_{i+} P_{j+} - P_{i+} Q_{j+}) \\
&\quad + Q_{i+} Q_{j+} + P_{i+} P_{j+} + i(Q_{i+} P_{j+} - P_{i+} Q_{j+})] \\
&= Q_{i+} Q_{j+} + P_{i+} P_{j+} \tag{3.126}
\end{aligned}$$

In the same way, we find for the remaining three terms

$$\begin{aligned}
a_{i_+} b_{j_-} + a_{i_+}^\dagger b_{j_-}^\dagger &= Q_{i_+} \mathcal{R}_{j_-} - \mathcal{P}_{i_+} S_{j_-} \\
b_{i_-}^\dagger a_{j_+}^\dagger + b_{i_-} a_{j_+} &= \mathcal{R}_{i_-} Q_{j_+} - S_{i_-} \mathcal{P}_{j_+} \\
b_{i_-}^\dagger b_{j_-} + b_{i_-} b_{j_-}^\dagger &= \mathcal{R}_{i_-} \mathcal{R}_{j_-} + S_{i_-} S_{j_-}
\end{aligned} \tag{3.127}$$

For the n_{i_+} and n_{i_-} operators we can write

$$\begin{aligned}
n_{i_+} &= a_{i_+}^\dagger a_{i_+} = \frac{1}{2} (Q_{i_+} - i\mathcal{P}_{i_+}) (Q_{i_+} + i\mathcal{P}_{i_+}) \\
&= \frac{1}{2} [Q_{i_+}^2 + \mathcal{P}_{i_+}^2 + i \underbrace{(Q_{i_+} \mathcal{P}_{i_+} - \mathcal{P}_{i_+} Q_{i_+})}_{[\mathcal{Q}_{i_+}, \mathcal{P}_{i_+}] = i\mathbb{1}}] = \frac{1}{2} (Q_{i_+}^2 + \mathcal{P}_{i_+}^2 - \mathbb{1})
\end{aligned} \tag{3.128}$$

$$\begin{aligned}
n_{i_-} &= b_{i_-}^\dagger b_{i_-} = \frac{1}{2} (\mathcal{R}_{i_-} - iS_{i_-}) (\mathcal{R}_{i_-} + iS_{i_-}) \\
&= \frac{1}{2} [\mathcal{R}_{i_-}^2 + S_{i_-}^2 + i \underbrace{(\mathcal{R}_{i_-} S_{i_-} - S_{i_-} \mathcal{R}_{i_-})}_{[\mathcal{R}_{i_-}, S_{i_-}] = i\mathbb{1}}] = \frac{1}{2} (\mathcal{R}_{i_-}^2 + S_{i_-}^2 - \mathbb{1})
\end{aligned} \tag{3.129}$$

Inserting equation (3.126), (3.127), (3.128) and (3.129) into the Hamiltonian

$$\begin{aligned}
\mathcal{H}^{\text{AFSW}} &= SJ'_\perp \left(\sum_{\substack{i_+, j_+ = 1 \\ i_+ \neq j_+}}^{N/2} \frac{Q_{i_+} Q_{j_+} + \mathcal{P}_{i_+} \mathcal{P}_{j_+}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} + \sum_{\substack{i_+, j_- = 1 \\ i_+ \neq j_-}}^{N/2} \frac{Q_{i_+} \mathcal{R}_{j_-} - \mathcal{P}_{i_+} S_{j_-}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} \right. \\
&\quad \left. + \sum_{\substack{i_-, j_+ = 1 \\ i_- \neq j_+}}^{N/2} \frac{\mathcal{R}_{i_-} Q_{j_+} - S_{i_-} \mathcal{P}_{j_+}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} + \sum_{\substack{i_-, j_- = 1 \\ i_- \neq j_-}}^{N/2} \frac{\mathcal{R}_{i_-} \mathcal{R}_{j_-} + S_{i_-} S_{j_-}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} \right) \\
&\quad + SJ'_z \left\{ SNg + g \left(\underbrace{\sum_{i_+ = 1}^{N/2} \mathbb{1} + \sum_{i_- = 1}^{N/2} \mathbb{1}}_{= N} \right) - g \left[\sum_{i_+ = 1}^{N/2} (Q_{i_+}^2 + \mathcal{P}_{i_+}^2) + \sum_{i_- = 1}^{N/2} (\mathcal{R}_{i_-}^2 + S_{i_-}^2) \right] \right\} \\
&= NgJ'_z S(S+1) \\
&\quad + SJ'_\perp \left(\sum_{\substack{i_+, j_+ = 1 \\ i_+ \neq j_+}}^{N/2} \frac{Q_{i_+} Q_{j_+} + \mathcal{P}_{i_+} \mathcal{P}_{j_+}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} + 2 \sum_{\substack{i_+, j_- = 1 \\ i_+ \neq j_-}}^{N/2} \frac{Q_{i_+} \mathcal{R}_{j_-} - \mathcal{P}_{i_+} S_{j_-}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} \right. \\
&\quad \left. + \sum_{\substack{i_-, j_- = 1 \\ i_- \neq j_-}}^{N/2} \frac{\mathcal{R}_{i_-} \mathcal{R}_{j_-} + S_{i_-} S_{j_-}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} \right) + SJ'_z \left[\sum_{i_+ = 1}^{N/2} (Q_{i_+}^2 + \mathcal{P}_{i_+}^2) + \sum_{i_- = 1}^{N/2} (\mathcal{R}_{i_-}^2 + S_{i_-}^2) \right]
\end{aligned} \tag{3.130}$$

Now in the third step we Fourier transform equation (3.130). The Fourier transformation of the canonical conjugate operators on their respective sublattice reads

$$\begin{aligned} Q_{i+} &= \frac{2}{N} \sum_{\mathbf{k}} Q_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_{i+}} & P_{i+} &= \frac{2}{N} \sum_{\mathbf{k}} P_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{R}_{i+}} \\ \mathcal{R}_{i-} &= \frac{2}{N} \sum_{\mathbf{k}} \mathcal{R}_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_{i-}} & S_{i-} &= \frac{2}{N} \sum_{\mathbf{k}} S_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{R}_{i-}} \end{aligned} \quad (3.131)$$

In the antiferromagnetic case the Fourier transform of the interaction potential must be divided in two classes V^{\oplus} and V^{\ominus}

$$V^{\oplus}(\mathbf{R}_{i\pm} - \mathbf{R}_{j\pm}) = V^{\oplus}(\mathbf{R}_{n\oplus}) \stackrel{(F.7)}{=} \left(\frac{2}{N}\right)^2 \sum_{\mathbf{k}} V^{\oplus}(\mathbf{k}) e^{i\mathbf{k}\cdot(\mathbf{R}_{i\pm} - \mathbf{R}_{j\pm})} = \frac{4}{N^2} \sum_{\mathbf{k}} V^{\oplus}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{R}_{n\oplus}} \quad (3.132)$$

$$V^{\ominus}(\mathbf{R}_{i\pm} - \mathbf{R}_{j\mp}) = V^{\ominus}(\mathbf{R}_{n\ominus}) \stackrel{(F.7)}{=} \left(\frac{2}{N}\right)^2 \sum_{\mathbf{k}} V^{\ominus}(\mathbf{k}) e^{i\mathbf{k}\cdot(\mathbf{R}_{i\pm} - \mathbf{R}_{j\mp})} = \frac{4}{N^2} \sum_{\mathbf{k}} V^{\ominus}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{R}_{n\ominus}} \quad (3.133)$$

where V^{\oplus} describes the interaction of molecules on the same sublattice and V^{\ominus} the interaction of molecules on different sublattices. Following the Fourier transformation of the ferromagnetic Hamiltonian, the solution for the sums over identical sublattices are already calculated in (3.88)

$$\sum_{\substack{i+,j+=1 \\ i+ \neq j+}}^{N/2} \frac{Q_{i+} Q_{j+} + P_{i+} P_{j+}}{|\mathbf{R}_{i+} - \mathbf{R}_{j+}|^3} = \sum_{\mathbf{k}} (Q_{\mathbf{k}}^2 + P_{\mathbf{k}}^2) \epsilon^{\oplus}(\mathbf{k}) \quad (3.134)$$

$$\sum_{\substack{i-,j-=1 \\ i- \neq j-}}^{N/2} \frac{\mathcal{R}_{i-} \mathcal{R}_{j-} + S_{i-} S_{j-}}{|\mathbf{R}_{i-} - \mathbf{R}_{j-}|^3} = \sum_{\mathbf{k}} (\mathcal{R}_{\mathbf{k}}^2 + S_{\mathbf{k}}^2) \epsilon^{\oplus}(\mathbf{k}) \quad (3.135)$$

For the term involving operators on different sublattices we insert (3.131) and (3.133)

$$\begin{aligned} \sum_{i+,j-=1}^{N/2} \frac{Q_{i+} \mathcal{R}_{j-} - P_{i+} S_{j-}}{|\mathbf{R}_{i+} - \mathbf{R}_{j-}|^3} &= \sum_{\mathbf{R}_i, \mathbf{R}_j} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \left(\frac{4}{N}\right)^2 \left(Q_{\mathbf{k}} \mathcal{R}_{\mathbf{k}'} V^{\ominus}(\mathbf{q}) e^{i\mathbf{k}\cdot\mathbf{R}_{i+}} e^{i\mathbf{k}'\cdot\mathbf{R}_{j-}} e^{i\mathbf{q}\cdot(\mathbf{R}_{i+} - \mathbf{R}_{j-})} \right. \\ &\quad \left. - P_{\mathbf{k}} S_{\mathbf{k}'} e^{-i\mathbf{k}\cdot\mathbf{R}_{i+}} e^{-i\mathbf{k}'\cdot\mathbf{R}_{j-}} e^{i\mathbf{q}\cdot(\mathbf{R}_{i+} - \mathbf{R}_{j-})} \right) \\ &= \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} \left(Q_{\mathbf{k}} \mathcal{R}_{\mathbf{k}'} V^{\ominus}(\mathbf{q}) \delta(-\mathbf{k} - \mathbf{q}) \delta(-\mathbf{k}' + \mathbf{q}) - P_{\mathbf{k}} S_{\mathbf{k}'} \delta(\mathbf{k} - \mathbf{q}) \delta(\mathbf{k}' + \mathbf{q}) \right) \\ &= \sum_{\mathbf{k}} \left(Q_{\mathbf{k}} \underbrace{\mathcal{R}_{-\mathbf{k}}}_{=\mathcal{R}_{\mathbf{k}}} V^{\ominus}(-\mathbf{k}) - P_{\mathbf{k}} \underbrace{S_{-\mathbf{k}}}_{=S_{\mathbf{k}}} V^{\ominus}(\mathbf{k}) \right) \\ &= \sum_{\mathbf{k}} \left(Q_{\mathbf{k}} \mathcal{R}_{\mathbf{k}} - P_{\mathbf{k}} S_{\mathbf{k}} \right) \epsilon^{\ominus}(\mathbf{k}) \end{aligned} \quad (3.136)$$

The second part of the spin wave Hamiltonian contains only hermitian operators squared, i.e. Q^2 . The Fourier transformation is isometric in the Hilbert space and so the square of an hermitian operator remains invariant, see for example equation (3.90)

$$\sum_{i_+=1}^{N/2} (Q_{i_+}^2 + P_{i_+}^2) = \sum_{\mathbf{k}} (Q_{\mathbf{k}}^2 + P_{\mathbf{k}}^2) \quad (3.137)$$

$$\sum_{i_-=1}^{N/2} (\mathcal{R}_{i_-}^2 + S_{i_-}^2) = \sum_{\mathbf{k}} (\mathcal{R}_{\mathbf{k}}^2 + S_{\mathbf{k}}^2) \quad (3.138)$$

Now we can write the Fourier transform of the spin wave Hamiltonian $\mathcal{H}^{\text{AFSW}}$ (3.130) as

$$\begin{aligned} \widetilde{\mathcal{H}}^{\text{AFSW}} &= NgJ'_z S(S+1) + J'_\perp S \sum_{\mathbf{k}} \left[(Q_{\mathbf{k}}^2 + P_{\mathbf{k}}^2 + \mathcal{R}_{\mathbf{k}}^2 + S_{\mathbf{k}}^2) \epsilon^\oplus(\mathbf{k}) + 2(Q_{\mathbf{k}}\mathcal{R}_{\mathbf{k}} - P_{\mathbf{k}}S_{\mathbf{k}}) \epsilon^\ominus(\mathbf{k}) \right] \\ &\quad - gJ'_z S \sum_{\mathbf{k}} (Q_{\mathbf{k}}^2 + P_{\mathbf{k}}^2 + \mathcal{R}_{\mathbf{k}}^2 + S_{\mathbf{k}}^2) \end{aligned} \quad (3.139)$$

In contrast to the ferromagnetic calculation the Fourier transformation does not diagonalize the Hamiltonian because of the mixed term $Q_{\mathbf{k}}\mathcal{R}_{\mathbf{k}} - P_{\mathbf{k}}S_{\mathbf{k}}$. In order to diagonalize

$$\begin{pmatrix} Q_{\mathbf{k}} \\ P_{\mathbf{k}} \\ \mathcal{R}_{\mathbf{k}} \\ S_{\mathbf{k}} \end{pmatrix} \cdot \begin{pmatrix} J'_\perp S \epsilon^\oplus(\mathbf{k}) - J'_z Sg & 0 & J'_\perp S \epsilon^\ominus(\mathbf{k}) & 0 \\ 0 & J'_\perp S \epsilon^\oplus(\mathbf{k}) - J'_z Sg & 0 & -J'_\perp S \epsilon^\ominus(\mathbf{k}) \\ J'_\perp S \epsilon^\ominus(\mathbf{k}) & 0 & J'_\perp S \epsilon^\oplus(\mathbf{k}) - J'_z Sg & 0 \\ 0 & -J'_\perp S \epsilon^\ominus(\mathbf{k}) & 0 & 0 \end{pmatrix} \begin{pmatrix} Q_{\mathbf{k}} \\ P_{\mathbf{k}} \\ \mathcal{R}_{\mathbf{k}} \\ S_{\mathbf{k}} \end{pmatrix} \quad (3.140)$$

we need a second unitary canonical transformation

$$\begin{aligned} Q_{\mathbf{k}} &= \frac{1}{\sqrt{2}} (q_{1\mathbf{k}} + q_{2\mathbf{k}}) & P_{\mathbf{k}} &= \frac{1}{\sqrt{2}} (p_{1\mathbf{k}} + p_{2\mathbf{k}}) \\ \mathcal{R}_{\mathbf{k}} &= \frac{1}{\sqrt{2}} (q_{1\mathbf{k}} - q_{2\mathbf{k}}) & S_{\mathbf{k}} &= \frac{1}{\sqrt{2}} (p_{1\mathbf{k}} - p_{2\mathbf{k}}) \end{aligned} \quad (3.141)$$

with the canonical commutator relations $[q_{i\mathbf{k}}, p_{i\mathbf{k}}] = i\delta_{ij}$. Applying (3.141) to the Hamiltonian $\widetilde{\mathcal{H}}^{\text{AFSW}}$ we have to insert

$$\begin{aligned} Q_{\mathbf{k}}^2 + P_{\mathbf{k}}^2 + \mathcal{R}_{\mathbf{k}}^2 + S_{\mathbf{k}}^2 &= \frac{1}{2} \left[(q_{1\mathbf{k}} + q_{2\mathbf{k}})^2 + (q_{1\mathbf{k}} - q_{2\mathbf{k}})^2 + (p_{1\mathbf{k}} + p_{2\mathbf{k}})^2 + (p_{1\mathbf{k}} - p_{2\mathbf{k}})^2 \right] \\ &= q_{1\mathbf{k}}^2 + q_{2\mathbf{k}}^2 + p_{1\mathbf{k}}^2 + p_{2\mathbf{k}}^2 \end{aligned} \quad (3.142)$$

$$\begin{aligned} Q_{\mathbf{k}}\mathcal{R}_{\mathbf{k}} - P_{\mathbf{k}}S_{\mathbf{k}} &= \frac{1}{2} \left[(q_{1\mathbf{k}} + q_{2\mathbf{k}})(q_{1\mathbf{k}} - q_{2\mathbf{k}}) - (p_{1\mathbf{k}} + p_{2\mathbf{k}})(p_{1\mathbf{k}} - p_{2\mathbf{k}}) \right] \\ &= \frac{1}{2} (q_{1\mathbf{k}}^2 - q_{2\mathbf{k}}^2 - p_{1\mathbf{k}}^2 + p_{2\mathbf{k}}^2) \end{aligned} \quad (3.143)$$

to get a completely diagonalized operator reading

$$\begin{aligned}
\widetilde{\mathcal{H}}^{\text{AFSW}} &= NgJ'_z S(S+1) + J'_\perp S \sum_{\mathbf{k}} \left[\epsilon^\oplus(\mathbf{k}) (q_{1\mathbf{k}}^2 + q_{2\mathbf{k}}^2 + p_{1\mathbf{k}}^2 + p_{2\mathbf{k}}^2) \right. \\
&\quad \left. + \epsilon^\ominus(\mathbf{k}) (q_{1\mathbf{k}}^2 - q_{2\mathbf{k}}^2 - p_{1\mathbf{k}}^2 + p_{2\mathbf{k}}^2) \right] - gJ'_z S \sum_{\mathbf{k}} (q_{1\mathbf{k}}^2 + q_{2\mathbf{k}}^2 + p_{1\mathbf{k}}^2 + p_{2\mathbf{k}}^2) \\
&= NgJ'_z S(S+1) - S \sum_{\mathbf{k}} \left\{ \left[gJ'_z - J'_\perp \underbrace{(\epsilon^\oplus(\mathbf{k}) + \epsilon^\ominus(\mathbf{k}))}_{=\epsilon(\mathbf{k})} \right] q_{1\mathbf{k}}^2 \right. \\
&\quad + \left[gJ'_z - J'_\perp \underbrace{(\epsilon^\oplus(\mathbf{k}) - \epsilon^\ominus(\mathbf{k}))}_{=\epsilon'(\mathbf{k})} \right] p_{1\mathbf{k}}^2 + \left[gJ'_z - J'_\perp \underbrace{(\epsilon^\oplus(\mathbf{k}) - \epsilon^\ominus(\mathbf{k}))}_{=\epsilon'(\mathbf{k})} \right] q_{2\mathbf{k}}^2 \\
&\quad \left. + \left[gJ'_z - J'_\perp \underbrace{(\epsilon^\oplus(\mathbf{k}) + \epsilon^\ominus(\mathbf{k}))}_{=\epsilon(\mathbf{k})} \right] p_{2\mathbf{k}}^2 \right\} \tag{3.144}
\end{aligned}$$

Comparing with the harmonic oscillator (3.94)

$$\frac{1}{m_i} p_{i\mathbf{k}}^2 + m_i \omega_i^2 q_{i\mathbf{k}}^2 = \omega_i (2n_{i\mathbf{k}} + 1) \tag{3.145}$$

we obtain

$$\begin{aligned}
\frac{1}{m_1} &= gJ'_z - J'_\perp \epsilon'(\mathbf{k}) = m_2 \omega_2^2 \\
m_1 \omega_1^2 &= gJ'_z - J'_\perp \epsilon(\mathbf{k}) = \frac{1}{m_2} \\
\hookrightarrow \frac{1}{m_i} m_i \omega_i^2 &= \omega_i^2 = (gJ'_z - J'_\perp \epsilon'(\mathbf{k})) (gJ'_z - J'_\perp \epsilon(\mathbf{k}))
\end{aligned} \tag{3.146}$$

Rewriting the spin wave Hamiltonian into a “classical” ground state energy part, ignoring the quantum fluctuation caused by the spin waves and two harmonic excitation gives rise to

$$\widetilde{\mathcal{H}}^{\text{AFSW}} = E_{\text{AF-class}}^z + \sum_{\mathbf{k}} \omega_{\text{AF}}^z(\mathbf{k}) \left[\left(n_{1\mathbf{k}} + \frac{1}{2} \right) + \left(n_{2\mathbf{k}} + \frac{1}{2} \right) \right] \tag{3.147}$$

with

$$E_{\text{AF-class}}^z = NgJ'_z S(S+1) \tag{3.148}$$

$$\omega_{\text{AF}}^z(\mathbf{k}) = 2S \sqrt{(gJ'_z - J'_\perp \epsilon'(\mathbf{k})) (gJ'_z - J'_\perp \epsilon(\mathbf{k}))} \tag{3.149}$$

In the in-axis antiferromagnet case we have found two excitation, yet they both possess the same dispersion relation ω_{AF}^z . In order to fully understand this result we should resolve how to express g and $\epsilon'(\mathbf{k})$ in terms of the Fourier transform of the interaction potential $\epsilon(\mathbf{k})$. Calculating the

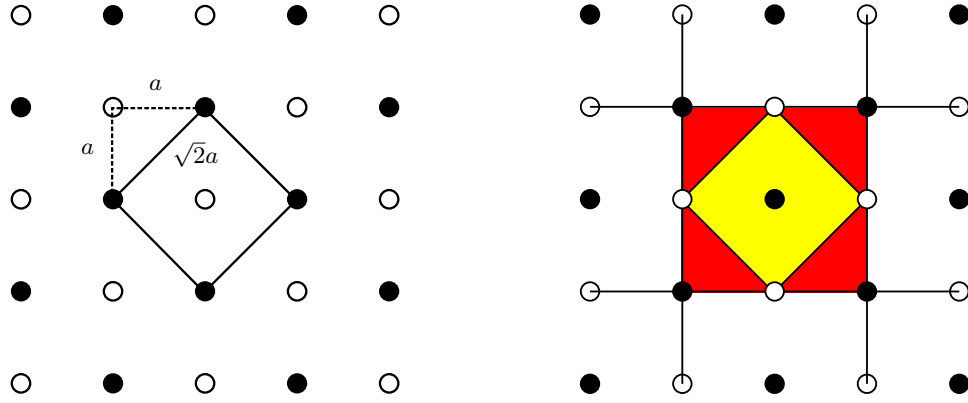


Figure 3.9 Left Panel: The two sublattices are also square lattices with a lattice constant $a\sqrt{2}$. Therefore we can rescale the sum over one of the sublattices by the factor $\sqrt{2}$ and retrieving the summation over the complete lattice with the rescaled lattice constant.

Right Panel: The red colored area denotes the first Brillouin zone of the total lattice, whereas the yellow colored area indicates the first Brillouin zone of one of the sublattices. Note that the vertices at $(\pi, 0)$ and $(0, \pi)$ are identical in both Brillouin zones as well as paths starting from the origin running in $(kx, 0)$ or $(0, ky)$ -direction.

antiferromagnetic lattice factor directly with the help of a geometric observation as shown in Figure 3.9 we find

$$g^{\oplus} = \sum_{\mathbf{n}_{\oplus}} \frac{1}{|\mathbf{R}_{\mathbf{n}_{\oplus}}|^3} = \frac{1}{\sqrt{2}^3} \sum_{\mathbf{R}_n} \frac{1}{|\mathbf{R}_n|^3} \stackrel{(3.99)}{=} \frac{1}{\sqrt{2}^3} \epsilon(\mathbf{0}) = 2^{-3/2} \epsilon(\mathbf{0}) \quad (3.150)$$

and respectively

$$g^{\ominus} = \sum_{\mathbf{R}_n} \frac{1}{|\mathbf{R}_n|^3} - \sum_{\mathbf{n}_{\oplus}} \frac{1}{|\mathbf{R}_{\mathbf{n}_{\oplus}}|^3} = \epsilon(\mathbf{0}) - 2^{-3/2} \epsilon(\mathbf{0}) = (1 - 2^{-3/2}) \epsilon(\mathbf{0}) \quad (3.151)$$

Combining equation (3.150) and (3.151) gives rise to

$$g = g^{\oplus} - g^{\ominus} = 2^{-3/2} \epsilon(\mathbf{0}) - (1 - 2^{-3/2}) \epsilon(\mathbf{0}) = \left(\frac{1}{\sqrt{2}} - 1 \right) \epsilon(\mathbf{0}) \quad (3.152)$$

On the other hand g can be determined with the help of a shift in \mathbf{k} -space

$$\begin{aligned} g = g^{\oplus} - g^{\ominus} &= \sum_{\mathbf{n}_{\oplus}} \frac{1}{|\mathbf{R}_{\mathbf{n}_{\oplus}}|^3} - \sum_{\mathbf{n}_{\ominus}} \frac{1}{|\mathbf{R}_{\mathbf{n}_{\ominus}}|^3} \\ &= \sum_{\mathbf{n}_{\oplus}} \frac{e^{i(\frac{\pi}{2}) \cdot \mathbf{R}_{\mathbf{n}_{\oplus}}}}{|\mathbf{R}_{\mathbf{n}_{\oplus}}|^3} + \sum_{\mathbf{n}_{\ominus}} \frac{e^{i(\frac{\pi}{2}) \cdot \mathbf{R}_{\mathbf{n}_{\ominus}}}}{|\mathbf{R}_{\mathbf{n}_{\ominus}}|^3} \\ &= \sum_{\mathbf{R}_n} \frac{e^{i(\frac{\pi}{2}) \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} = \epsilon(\pi, \pi) \approx -2.6464 \end{aligned} \quad (3.153)$$

where we have used the fact that $\pi(R_x + R_y)_\oplus = (\pi \cdot \text{even number})$ and $\pi(R_x + R_y)_\ominus = (\pi \cdot \text{odd number})$ are generating the sign in the respective sum and that the sum over the \bullet -sublattice and the \circ -sublattice is identical to the sum over the complete lattice. This derivation can be verified by checking the two factors involved in determining g , numerically

$$\left(\frac{1}{\sqrt{2}} - 1\right) \epsilon(\mathbf{0}) \stackrel{!}{=} \epsilon(\pi, \pi) \quad \hookrightarrow \quad \frac{\epsilon(\pi, \pi)}{\epsilon(\mathbf{0})} = \left(\frac{1}{\sqrt{2}} - 1\right) \quad (3.154)$$

$$\frac{\epsilon(\pi, \pi)}{\epsilon(\mathbf{0})} \approx -0.29295 \quad \left(\frac{1}{\sqrt{2}} - 1\right) \approx -0.29289 \quad (3.155)$$

In the same way we can calculate $\epsilon(\mathbf{k}) = \epsilon^\oplus(\mathbf{k}) + \epsilon^\ominus(\mathbf{k})$ and $\epsilon'(\mathbf{k}) = \epsilon^\oplus(\mathbf{k}) - \epsilon^\ominus(\mathbf{k})$

$$\epsilon^\oplus(\mathbf{k}) + \epsilon^\ominus(\mathbf{k}) = \sum_{\mathbf{n}_\oplus} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}_\oplus}}}{|\mathbf{R}_{\mathbf{n}_\oplus}|^3} + \sum_{\mathbf{n}_\ominus} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}_\ominus}}}{|\mathbf{R}_{\mathbf{n}_\ominus}|^3} = \sum_{\mathbf{R}_n} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} = \epsilon(\mathbf{k}) \quad (3.156)$$

$$\begin{aligned} \epsilon^\oplus(\mathbf{k}) - \epsilon^\ominus(\mathbf{k}) &= \sum_{\mathbf{n}_\oplus} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}_\oplus}}}{|\mathbf{R}_{\mathbf{n}_\oplus}|^3} - \sum_{\mathbf{n}_\ominus} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}_\ominus}}}{|\mathbf{R}_{\mathbf{n}_\ominus}|^3} \\ &= \sum_{\mathbf{n}_\oplus} \underbrace{e^{i\left(\frac{\pi}{\pi}\right) \cdot \mathbf{R}_{\mathbf{n}_\oplus}}}_{=1} \frac{e^{i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}_\oplus}}}{|\mathbf{R}_{\mathbf{n}_\oplus}|^3} + \sum_{\mathbf{n}_\ominus} \underbrace{e^{i\left(\frac{\pi}{\pi}\right) \cdot \mathbf{R}_{\mathbf{n}_\ominus}}}_{=-1} \frac{e^{i\left(\frac{\pi}{\pi}\right) \cdot \mathbf{R}_{\mathbf{n}_\ominus}}}{|\mathbf{R}_{\mathbf{n}_\ominus}|^3} \\ &= \sum_{\mathbf{R}_n} \frac{e^{i\left(\mathbf{k} + \left(\frac{\pi}{\pi}\right)\right) \cdot \mathbf{R}_n}}{|\mathbf{R}_n|^3} = \epsilon\left(\mathbf{k} + \begin{pmatrix} \pi \\ \pi \end{pmatrix}\right) \end{aligned} \quad (3.157)$$

Hence we can rewrite the dispersion relation and the “classical” ground state energy in (3.149) and (3.148), setting $\mathbf{K} = (\pi, \pi)^t$

$$E_{\text{AF-class}}^z = N\epsilon(\pi, \pi)J'_z S(S+1) \quad (3.158)$$

$$\begin{aligned} \omega_{\text{AF}}^z(\mathbf{k}) &= 2S\sqrt{(J'_z\epsilon(\mathbf{K}) - J'_\perp\epsilon(\mathbf{k} + \mathbf{K})) (J'_z\epsilon(\mathbf{K}) - J'_\perp\epsilon(\mathbf{k}))} \\ &= 2J'_z S\sqrt{(\sin\vartheta\epsilon(\mathbf{k} + \mathbf{K}) - \cos\vartheta\epsilon(\mathbf{K})) (\sin\vartheta\epsilon(\mathbf{k}) - \cos\vartheta\epsilon(\mathbf{K}))} \end{aligned} \quad (3.159)$$

Note that $g = \epsilon(\pi, \pi) < 0$ and so the “classical” ground state energy $E_{\text{AF-class}}^z$ is also negative. The complete quantum mechanical ground state energy is corrected by taking the quantum fluctuations of the spin wave ground state with $n_{1\mathbf{k}} = 0$ and $n_{2\mathbf{k}} = 0$ into account

$$\begin{aligned} E_{\text{0AF}}^z &= N\epsilon(\pi, \pi)J'_z S(S+1) + \sum_{\mathbf{k}} \omega_{\text{AF}}^z(\mathbf{k}) \\ &= N\epsilon(\mathbf{K})J'_z S(S+1) + 2\epsilon(\mathbf{K})J'_z S \sum_{\mathbf{k}} \sqrt{\left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k} + \mathbf{K})}{\epsilon(\mathbf{K})}\right) \left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{K})}\right)} \\ &= N\epsilon(\mathbf{K})J'_z S \left[(S+1) + \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k} + \mathbf{K})}{\epsilon(\mathbf{K})}\right) \left(1 - \frac{J'_\perp}{J'_z} \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{K})}\right)} \right] \end{aligned} \quad (3.160)$$

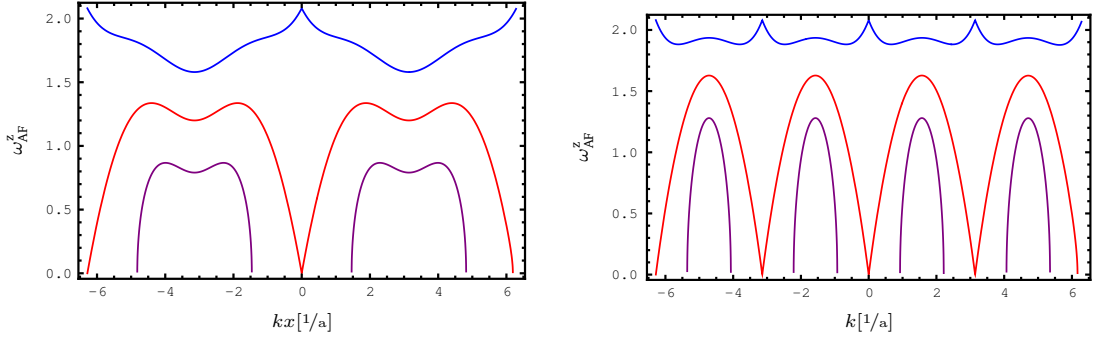


Figure 3.10 Left Panel: Dispersion relation ω_{AF}^z for different $\vartheta = 0.2\pi, 0.25\pi, 0.3\pi$ in k_x -direction. The dispersion relation is becoming imaginary for $\vartheta > \pi/4$ or $J'_\perp > J'_z$ indicating the instability of the in-axis ground state. Right Panel: The Dispersion relation ω_{AF}^z in $(k_x, k_x)^t$ -direction is actually $\pi/2$ periodic because the sublattices are rescaled square lattice with the factor $\sqrt{2}$. The edge of the first Brillouin zone of these sublattices is crossed in $(k_x, k_x)^t$ -direction at $(\pi/2, \pi/2)$, whereas in k_x -direction both first Brillouin zones match, e.g. see Figure 3.9.

where we have used the relation between the sum over all \mathbf{k} -vectors of the first Brillouin zone and the integral in \mathbf{k} -space as stated in equation (F.43). In Section 3.5 these quantum fluctuation are used to determine the type of phase transition between different ground states.

The dispersion relation is real for all \mathbf{k} in the first Brillouin zone for ϑ in the range $[\approx -\pi/10, \pi/4]$. For $\vartheta > \pi/4$ the in-plane coupling will destroy the Néel ordered ground state aligned in z -direction. Therefore in Subsection 3.3.6 we will determine the dispersion relation for harmonic excitations above the in-plane Néel ordered ground state. Due to the splitting of the total lattice in two sublattices there exists two identical dispersion relations and since the sublattices possess a larger lattice spacing the first Brillouin zone of these lattices leads to a different periodic behavior. In Figure 3.10 the dispersion relation in $(k_x, k_x)^t$ -direction is only $\pi/2$ periodic because in this direction the edge of the first Brillouin zone – as indicated in Figure 3.9 – is reached at $(\pi/2, \pi/2)$.

Considering only nearest neighbor interaction, the antiferromagnetic spin wave Hamiltonian $\widetilde{\mathcal{H}}^{\text{AFSW}}$ is simplified drastically. The sums over identical sublattices

$$\sum_{\substack{i_+, j_+ = 1 \\ i_+ \neq j_+}}^{N/2} \longrightarrow 0 \qquad \sum_{\substack{i_-, j_- = 1 \\ i_- \neq j_-}}^{N/2} \longrightarrow 0 \qquad (3.161)$$

can be set to zero since there are no nearest neighbor sites involved. This leads to vanishing $\epsilon^\oplus(\mathbf{k})$ and g^\oplus and to the replacements

$$g = -g^\ominus = -z \qquad \epsilon(\mathbf{k}) = \epsilon^\ominus(\mathbf{k}) = \epsilon(\mathbf{k})^{\text{NN}} \qquad \epsilon'(\mathbf{k}) = -\epsilon^\ominus(\mathbf{k}) = -\epsilon(\mathbf{k})^{\text{NN}} \qquad (3.162)$$

The nearest neighbor interaction is given according to (3.103) as

$$\epsilon(\mathbf{k})^{\text{NN}} = \sum_{\rho} \frac{e^{i\mathbf{k}\cdot\rho}}{|\rho|^3} = 2(\cos k_x + \cos k_y) \qquad (3.163)$$

Putting everything together the nearest neighbor antiferromagnetic Hamiltonian $\widetilde{\mathcal{H}}^{\text{AFSW-NN}}$ reads

$$\widetilde{\mathcal{H}}^{\text{AFSW-NN}} = -NzJ'_z S(S+1) + 2S \sum_{\mathbf{k}} \sqrt{(zJ'_z)^2 - (2J'_\perp (\cos k_x + \cos k_y))^2} \left(n_{\mathbf{k}} + \frac{1}{2} \right) \quad (3.164)$$

Here we see that the coordination number z is larger than the absolute value of the antiferromagnetic lattice constant, i.e. $z > |g|$ indicating a “screening” of the interaction between a spin on a particular lattice site and its neighboring spins, due to the fact that for long-range interactions also parallel aligned spins on the same sublattice can interact with each other.

3.3.4 In-plane magnetically ordered XXZ Model

As we have seen in Subsection 3.3.2 and 3.3.3 the magnetically ordered ground state is not stable for strong in-plane couplings $|J_\perp| > |J_z|$. The linear harmonic excitations above the magnetically ordered ground states aligned along the z -direction cannot be assumed to yield reasonable results. In fact for the strong in-plane ferromagnetic ordering the dispersion relation $\omega_{\text{F}}^z(\mathbf{k})$ is becoming negative and for the Néel ordered ground state we even find an imaginary dispersion relation $\omega_{\text{AF}}^z(\mathbf{k})$. So for strong in-plane coupling $|J_\perp| > |J_z|$ we must assume a new magnetically ordered ground state, i.e. aligned along the x -direction. Mathematically this can be achieved by a spatial rotation of the spin- $1/2$ XXZ model Hamiltonian about the y -axis

$$\mathcal{R}_\alpha = \begin{pmatrix} \cos \alpha & 0 & -\sin \alpha \\ 0 & 1 & 0 \\ \sin \alpha & 0 & \cos \alpha \end{pmatrix} \quad (3.165)$$

In order to create a magnetically ordered ground state in x -direction we have to rotate with the angle of $\pi/2$ about the y -axis

$$|x\text{-AF/F}\rangle = \mathcal{R}_{\pi/2}^{-1} |z\text{-AF/F}\rangle \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ -1 & 0 & 0 \end{pmatrix} |z\text{-AF/F}\rangle \quad (3.166)$$

Since the Holstein-Primakoff transformation acts on the operators in the Hamiltonian we need to transform the spin- $1/2$ XXZ-Hamiltonian (3.31)

$$\begin{aligned} \mathcal{H}^Z &= \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(J_\perp \frac{S_{ix}S_{jx} + S_{iy}S_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J_z \frac{S_{iz}S_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \\ &= J \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \sin \vartheta \left(\frac{\mathbf{S}_i \cdot \mathbf{S}_j - S_{iz}S_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) + \cos \vartheta \frac{S_{iz}S_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \\ &= J \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \sin \vartheta \frac{\mathbf{S}_i \cdot \mathbf{S}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} + (\cos \vartheta - \sin \vartheta) \frac{S_{iz}S_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \end{aligned} \quad (3.167)$$

In the following we will denote the rotated operators with an x superscript.

$$\mathcal{H}^x = \mathcal{R}_{x/2} H^Z \mathcal{R}_{x/2}^{-1} \quad \hookrightarrow \quad \mathbf{S} = \mathcal{R}_{x/2} \mathbf{S}^x \quad \text{and} \quad S_z = (-1) S_x^x \quad (3.168)$$

The scalar product of two vector operators is invariant under rotations, i.e. $\mathbf{S}_i^x \cdot \mathbf{S}_j^x = \mathbf{S}_i \cdot \mathbf{S}_j$. Inserting (3.168) into (3.167) we obtain

$$\begin{aligned} \mathcal{H}^x &= J \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \sin \vartheta \frac{\mathbf{S}_i^x \cdot \mathbf{S}_j^x}{|\mathbf{R}_i - \mathbf{R}_j|^3} + (\cos \vartheta - \sin \vartheta) \frac{(-1) S_{ix}^x (-1) S_{jx}^x}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \\ &= J \sum_{\substack{i,j=1 \\ i \neq j}}^N \left\{ \sin \vartheta \frac{S_{iy}^x S_{jy}^x + S_{iz}^x S_{jz}^x}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \cos \vartheta \frac{S_{ix}^x S_{jx}^x}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right\} \end{aligned} \quad (3.169)$$

As we can see the role of the operators S_x and S_z is interchanged, i.e. $S_z \longrightarrow S_x^x$. From now on, we will suppress the superscript x because it is clear that we will use this Hamiltonian to describe magnetically ordered spin- $1/2$ systems with aligned spins in x -direction.

3.3.5 In-Plane Ferromagnetic Spin Waves

Starting from the spin- $1/2$ XXZ Hamiltonian in x -direction (3.169) we need to express S_x and S_y in terms of the raising and lowering operators S_+ and S_-

$$\begin{aligned} S_{i+} &= S_{ix} + i S_{iy} & S_{ix} &= \frac{1}{2} (S_{i+} + S_{i-}) \\ S_{i-} &= S_{ix} - i S_{iy} & S_{iy} &= -\frac{i}{2} (S_{i+} - S_{i-}) \end{aligned} \quad (3.170)$$

The ferromagnetic Hamiltonian is then cast into

$$\begin{aligned} \mathcal{H}^{x-F} &= \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(-\frac{J_{\perp}}{4} \frac{S_{i+} S_{j+} - S_{i+} S_{j-} - S_{i-} S_{j+} + S_{i-} S_{j-}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\ &\quad \left. + \frac{J_z}{4} \frac{S_{i+} S_{j+} + S_{i+} S_{j-} + S_{i-} S_{j+} + S_{i-} S_{j-}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \end{aligned} \quad (3.171)$$

Following the calculation laid out in Subsection 3.3.2 and [15], we apply the Holstein-Primakoff transformation (3.67)

$$S_{i+} = \hbar \sqrt{2S} \varphi(n_i) a_i \quad S_{i-} = \hbar \sqrt{2S} a_i^{\dagger} \varphi(n_i) \quad S_{iz} = \hbar (S - n_i) \quad (3.172)$$

resulting in

$$\mathcal{H}^{x-F} = \frac{J_{\perp} S}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{-\varphi(n_i) a_i \varphi(n_j) a_j + \varphi(n_i) a_i a_j^{\dagger} \varphi(n_j) + a_i^{\dagger} \varphi(n_i) \varphi(n_j) a_j - a_i^{\dagger} \varphi(n_i) a_j^{\dagger} \varphi(n_j)}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right)$$

$$\begin{aligned}
& + \frac{J'_z S}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\varphi(\mathbf{n}_i) \mathbf{a}_i \varphi(\mathbf{n}_j) \mathbf{a}_j + \varphi(\mathbf{n}_i) \mathbf{a}_i \mathbf{a}_j^\dagger \varphi(\mathbf{n}_j) + \mathbf{a}_i^\dagger \varphi(\mathbf{n}_i) \varphi(\mathbf{n}_j) \mathbf{a}_j + \mathbf{a}_i^\dagger \varphi(\mathbf{n}_i) \mathbf{a}_j^\dagger \varphi(\mathbf{n}_j)}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \\
& + J'_\perp \left(S^2 N g^F - 2 S g^F \sum_{i=1}^N n_i + \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \tag{3.173}
\end{aligned}$$

Applying the spin wave approximation $\varphi(\mathbf{n}_i) \approx 1$ and $n_i n_j \approx 0$ we find

$$\begin{aligned}
\mathcal{H}^{\text{x-FSW}} & = J'_\perp S \left(S N g^F - 2 g^F \sum_{i=1}^N n_i \right) \\
& + \frac{J'_\perp S}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{-\mathbf{a}_i \mathbf{a}_j + \mathbf{a}_i \mathbf{a}_j^\dagger + \mathbf{a}_i^\dagger \mathbf{a}_j - \mathbf{a}_i^\dagger \mathbf{a}_j^\dagger}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \\
& + \frac{J'_z S}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\mathbf{a}_i \mathbf{a}_j + \mathbf{a}_i \mathbf{a}_j^\dagger + \mathbf{a}_i^\dagger \mathbf{a}_j + \mathbf{a}_i^\dagger \mathbf{a}_j^\dagger}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \tag{3.174}
\end{aligned}$$

As a next step we apply the unitary transformation (3.80)

$$\mathbf{a}_i = \frac{1}{\sqrt{2}} (\mathbf{Q}_i + i \mathbf{P}_i) \quad \mathbf{a}_i^\dagger = \frac{1}{\sqrt{2}} (\mathbf{Q}_i - i \mathbf{P}_i) \tag{3.175}$$

leading to

$$\begin{aligned}
\mathcal{H}^{\text{x-FSW}} & = N g^F J'_\perp S (S + 1) - J'_\perp S g^F \sum_{i=1}^N (\mathbf{Q}_i^2 + \mathbf{P}_i^2) \\
& + J'_\perp S \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\mathbf{P}_i \mathbf{P}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} + J'_z S \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\mathbf{Q}_i \mathbf{Q}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \tag{3.176}
\end{aligned}$$

The Fourier transform of this Hamiltonian using (3.85) is given by

$$\begin{aligned}
\widetilde{\mathcal{H}}^{\text{x-FSW}} & = N g^F J'_\perp S (S + 1) - J'_\perp S g^F \sum_{i=1}^N (\mathbf{Q}_i^2 + \mathbf{P}_i^2) \\
& + J'_\perp S \sum_{\mathbf{k}} \mathbf{P}_\mathbf{k}^2 \epsilon(\mathbf{k}) + J'_z S \sum_{\mathbf{k}} \mathbf{Q}_\mathbf{k}^2 \epsilon(\mathbf{k}) \\
& = N g^F J'_\perp S (S + 1) + \sum_{\mathbf{k}} \left[S (J'_z \epsilon(\mathbf{k}) - J'_\perp g^F) \mathbf{Q}_\mathbf{k}^2 + J'_\perp S (\epsilon(\mathbf{k}) - g^F) \mathbf{P}_\mathbf{k}^2 \right] \tag{3.177}
\end{aligned}$$

We can transform the diagonalized Fourier transform of $\mathcal{H}^{\text{x-FSW}}$ onto a harmonic oscillator with

$$\mathbf{Q} = \sqrt{\frac{\hbar}{2m\omega}} (\mathcal{A}^\dagger + \mathcal{A}) \quad \mathbf{P} = i \sqrt{\frac{\hbar m \omega}{2}} (\mathcal{A}^\dagger - \mathcal{A}) \tag{3.178}$$

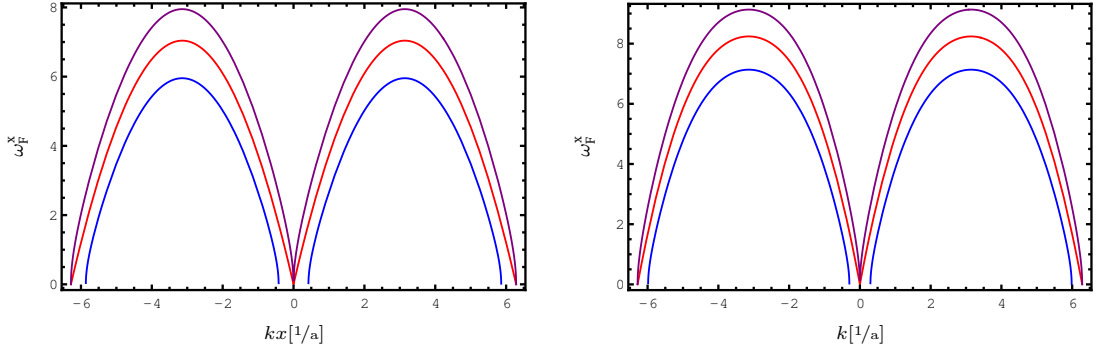


Figure 3.11 In-plane dispersion relation $\omega_{\text{F}}^{\text{x}}$ for different $\vartheta = 1.2\pi, 1.25\pi, 1.3\pi$ in k_x -direction (left picture) and $(k_x, k_x)^{\text{t}}$ -direction (right picture). For $\vartheta < 5\pi/4$ the in-plane ground state is unstable and the dispersion relation $\omega_{\text{F}}^{\text{x}}$ is becoming imaginary.

and read of the ground state energy $E_{0\text{F}}^{\text{x}}$ and the dispersion relation $\omega_{\text{F}}^{\text{x}}(\mathbf{k})$

$$\begin{aligned}\omega_{\text{F}}^{\text{x}}(\mathbf{k}) &= 2S\sqrt{(J'_z\epsilon(\mathbf{k}) - J'_\perp\epsilon(\mathbf{0}))J'_\perp(\epsilon(\mathbf{k}) - \epsilon(\mathbf{0}))} \\ &= 2J'S\sqrt{\sin\vartheta(\epsilon(\mathbf{0}) - \epsilon(\mathbf{k}))(\sin\vartheta\epsilon(\mathbf{0}) - \cos\vartheta\epsilon(\mathbf{k}))}\end{aligned}\quad (3.179)$$

$$\begin{aligned}E_{0\text{F}}^{\text{x}} &= Ng^{\text{F}}J'_\perp S(S+1) + \frac{1}{2}\sum_{\mathbf{k}}\omega_{\text{F}}^{\text{x}}(\mathbf{k}) \\ &= N\epsilon(\mathbf{0})J'_\perp S(S+1) + \epsilon(\mathbf{0})J'_\perp S\sum_{\mathbf{k}}\sqrt{\left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{0})}\right)\left(1 - \frac{J'_z\epsilon(\mathbf{k})}{J'_\perp\epsilon(\mathbf{0})}\right)} \\ &= N\epsilon(\mathbf{0})J'_\perp S\left[S+1 + \frac{a^2}{4\pi^2}\int d^2\mathbf{k}\sqrt{\left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{0})}\right)\left(1 - \frac{J'_z\epsilon(\mathbf{k})}{J'_\perp\epsilon(\mathbf{0})}\right)}\right]\end{aligned}\quad (3.180)$$

The dispersion relation is real only in the range of ϑ given by $[5\pi/4, 19\pi/10]$, where also the ground state energy remains negative since $J'_\perp < 0$.

3.3.6 In-Plane Antiferromagnetic Spin Waves

Last but not least we will discuss rather shortly the solution for the spin wave Hamiltonian starting from a Néel ordered ground state in x -direction. Since the steps of the calculation are identical to those in the previous Subsections, especially Subsection 3.3.3, we will sum up the results quickly. After splitting up the Hamiltonian in different sublattices (3.109) we apply a Holstein–Primakoff transformation (3.110) and the linearized spin wave approximation, yielding

$$\begin{aligned}\mathcal{H}^{\text{x-AFSW}} &= J'_\perp S\left[SNg - 2Sg\left(\sum_{i_+=1}^{\frac{N}{2}}n_{i_+} + \sum_{i_-=1}^{\frac{N}{2}}n_{i_-}\right)\right] \\ &\quad + \frac{J'_\perp S}{2}\sum_{\substack{i_+, j_+=1 \\ i_+ \neq j_+}}^N\left(\frac{-a_{i_+}a_{j_+} + a_{i_+}a_{j_+}^\dagger + a_{i_+}^\dagger a_{j_+} - a_{i_+}^\dagger a_{j_+}^\dagger}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3}\right)\end{aligned}$$

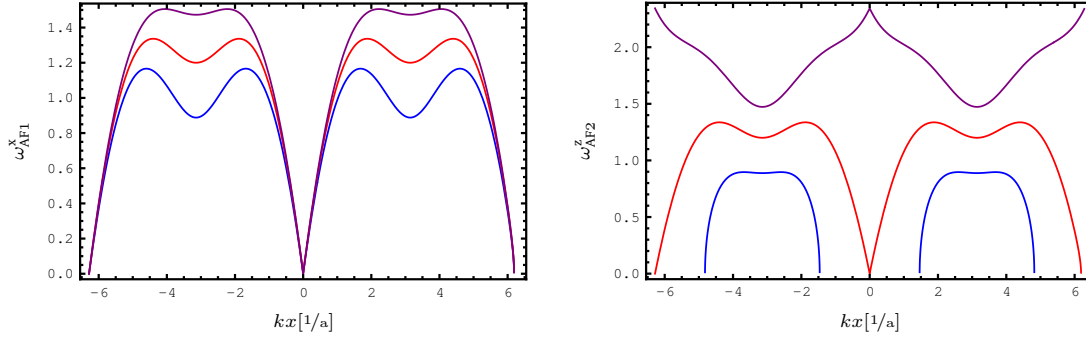


Figure 3.12 Left Panel: Gapless in-plane dispersion relation $\omega_{\text{AF1}}^x(\mathbf{k})$ in k_x -direction. Right Panel: Second gapped in-plane dispersion relation $\omega_{\text{AF2}}^x(\mathbf{k})$ in k_x -direction.

$$\begin{aligned}
& + \frac{J'_z S}{2} \sum_{\substack{i_+, j_+=1 \\ i_+ \neq j_+}}^N \left(\frac{a_{i_+} a_{j_+} + a_{i_+} a_{j_+}^\dagger + a_{i_+}^\dagger a_{j_+} + a_{i_+}^\dagger a_{j_+}^\dagger}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_+}|^3} \right) \\
& + \frac{J'_\perp S}{2} \sum_{\substack{i_+, j_-=1 \\ i_+ \neq j_-}}^N \left(\frac{-a_{i_+} b_{j_-}^\dagger + a_{i_+} b_{j_-} + a_{i_+}^\dagger b_{j_-}^\dagger - a_{i_+}^\dagger b_{j_-}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} \right) \\
& + \frac{J'_z S}{2} \sum_{\substack{i_+, j_-=1 \\ i_+ \neq j_-}}^N \left(\frac{a_{i_+} b_{j_-}^\dagger + a_{i_+} b_{j_-} + a_{i_+}^\dagger b_{j_-}^\dagger + a_{i_+}^\dagger b_{j_-}}{|\mathbf{R}_{i_+} - \mathbf{R}_{j_-}|^3} \right) \\
& + \frac{J'_\perp S}{2} \sum_{\substack{i_-, j_+=1 \\ i_- \neq j_+}}^N \left(\frac{-b_{i_-}^\dagger a_{j_+} + b_{i_-}^\dagger a_{j_+}^\dagger + b_{i_-} a_{j_+} - b_{i_-} a_{j_+}^\dagger}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} \right) \\
& + \frac{J'_z S}{2} \sum_{\substack{i_-, j_+=1 \\ i_- \neq j_+}}^N \left(\frac{b_{i_-}^\dagger a_{j_+} + b_{i_-}^\dagger a_{j_+}^\dagger + b_{i_-} a_{j_+} + b_{i_-} a_{j_+}^\dagger}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_+}|^3} \right) \\
& + \frac{J'_\perp S}{2} \sum_{\substack{i_-, j_-=1 \\ i_- \neq j_-}}^N \left(\frac{-b_{i_-}^\dagger b_{j_-}^\dagger + b_{i_-}^\dagger b_{j_-} + b_{i_-} b_{j_-}^\dagger - b_{i_-} b_{j_-}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} \right) \\
& + \frac{J'_z S}{2} \sum_{\substack{i_-, j_-=1 \\ i_- \neq j_-}}^N \left(\frac{b_{i_-}^\dagger b_{j_-}^\dagger + b_{i_-} b_{j_-}^\dagger + b_{i_-} b_{j_-}^\dagger + b_{i_-} b_{j_-}}{|\mathbf{R}_{i_-} - \mathbf{R}_{j_-}|^3} \right) \tag{3.181}
\end{aligned}$$

Applying a unitary transformation (3.125) and Fourier transforming the spin wave Hamiltonian $\mathcal{H}^{\text{x-AFSW}}$ we need to diagonalize the non-quadratic parts with the help of a second unitary transformation (3.141)

$$\widetilde{\mathcal{H}}^{\text{x-AFSW}} = NgJ'_\perp S(S+1)S \sum_{\mathbf{k}} \left[(J'_z \epsilon(\mathbf{k}) - gJ'_\perp) q_{1\mathbf{k}}^2 + (J'_z \epsilon'(\mathbf{k}) - gJ'_\perp) q_{2\mathbf{k}}^2 \right]$$

$$+ J'_\perp (\epsilon'(\mathbf{k}) - g) p_{1\mathbf{k}}^2 + J'_\perp (\epsilon(\mathbf{k}) - g) p_{2\mathbf{k}}^2 \Big] \quad (3.182)$$

Comparing with the harmonic oscillator (3.94) gives rise to the dispersion relation and the ground state energy

$$E_{0\text{AF}}^x = N\epsilon(\mathbf{K})J'_\perp S \left[(S+1) + \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k} + \mathbf{K})}{\epsilon(\mathbf{K})}\right) \left(1 - \frac{J'_z \epsilon(\mathbf{k})}{J'_\perp \epsilon(\mathbf{K})}\right)} \right] \quad (3.183)$$

$$\omega_{\text{AF1}}^x(\mathbf{k}) = 2S \sqrt{J'_\perp (\epsilon(\mathbf{K}) - \epsilon(\mathbf{k} + \mathbf{K})) (J'_\perp \epsilon(\mathbf{K}) - J'_z \epsilon(\mathbf{k}))} \quad (3.184)$$

$$\omega_{\text{AF2}}^x(\mathbf{k}) = 2S \sqrt{(J'_\perp \epsilon(\mathbf{K}) - J'_z \epsilon(\mathbf{k} + \mathbf{K})) J'_\perp (\epsilon(\mathbf{K}) - \epsilon(\mathbf{k}))} \quad (3.185)$$

Interestingly in the case of strong in-plane coupling $J_\perp > J'_z$ the two dispersion relation are not identical, but they are describing the same physics since they are only shifted by an reciprocal lattice vector $\mathbf{G} = (\pi, \pi)^t$ of the first Brillouin zone of one of the sublattices. Due to the 2π periodicity of $\epsilon(\mathbf{k})$ we find the following relation between the two dispersion relations $\omega_{\text{AF1}}^x(\mathbf{k})$ and $\omega_{\text{AF2}}^x(\mathbf{k})$

$$\omega_{\text{AF1}}^x \left(\mathbf{k} + \begin{pmatrix} \pi \\ \pi \end{pmatrix} \right) = \omega_{\text{AF2}}^x(\mathbf{k}) \quad \omega_{\text{AF2}}^x \left(\mathbf{k} + \begin{pmatrix} \pi \\ \pi \end{pmatrix} \right) = \omega_{\text{AF1}}^x(\mathbf{k}) \quad (3.186)$$

Therefore we can use one of the two dispersion relations, i.e. $\omega_{\text{AF1}}^x(\mathbf{k})$ and generate the other via shifting with \mathbf{G} . Since the linearized spin wave excitations are low lying collective excitations we deal in the quasi particle picture with bosonic particles called magnons that possess creation a^\dagger and annihilation operators a . By drawing an analogy to phonons we see that the second mode that is arising due to the two sublattices, corresponds to the optical mode of phonons, whereas the first mode can be considered as acoustical mode. The introduction of the two sublattices \bullet and \circ doubles the lattice constant of each sublattice and therefore the Brillouin zone of these sublattices is reduced as shown in Figure 3.9. Since we have an underlying symmetry of the two dimensional square lattice, we can shift the extended scheme back into the first Brillouin zone with the help of an reciprocal lattice vector \mathbf{G} (see Appendix F.5.2).

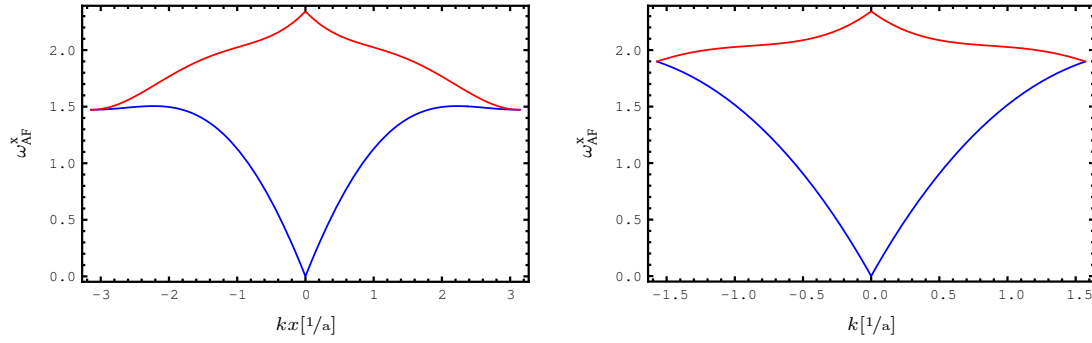


Figure 3.13 There are two dispersion relation for the in-plane antiferromagnet $\omega_{\text{AF1}}^x(\mathbf{k})$ and $\omega_{\text{AF2}}^x(\mathbf{k})$ in k_x -direction (left picture). The second dispersion relation possesses a finite energy gap for $\mathbf{k} = \mathbf{0}$. In the right picture the dispersion relations in $(k_x, k_x)^t$ -direction are shown. Note that due to the rescaled sublattice constant the first Brillouin zone in this direction is halved.

3.4 Mean Field Solution of the XXZ Model

The spin- $1/2$ XXZ model cannot be solved analytically, since we have to deal with correlated two particle operators. One possibility to simplify the complicated two particle operators is to apply the mean field approximation. In this approximation we consider only a single spin in the background field generated by all other spins. Thus, we reduce the interaction problem of two spins to non-interacting single spins in an magnetic field. This magnetic field must be calculated self-consistently, i.e. to reproduce the right interaction between the spins in the system. Since we replace all interaction between single spins by a mean field, we do not have any fluctuations or correlations between those spins and we can regard the mean field approximation as the zeroth order of the Hamiltonian in the fluctuation. It is clear, that the mean field approximation will only be good for systems with small fluctuations. This is true for long range interactions and for high dimensionality of the system. The Ginzburg-Landau criterion states, that the upper critical dimension is $d_c = 4$. Only above d_c mean field is still valid when approaching a critical point and will not break down due to the critical fluctuation with diverging correlation length.

To apply the mean field approximation to \mathcal{H}^{XXZ} one needs to construct effective one particle operators. Following the idea of expanding the fluctuation around the average or mean value, we can introduce the new operators

$$\delta\mathbf{S}_i = \mathbf{S}_i - \langle \mathbf{S}_i \rangle \quad \hookrightarrow \quad \mathbf{S}_i = \langle \mathbf{S}_i \rangle + \delta\mathbf{S}_i \quad (3.187)$$

where $\delta\mathbf{S}_i$ defines the deviation from the expectation value of the spin operator. Rewriting the interacting two spin operator $\mathbf{S}_i \cdot \mathbf{S}_j$ with the help of the deviation gives rise to

$$\begin{aligned} \mathbf{S}_i \cdot \mathbf{S}_j &= (\langle \mathbf{S}_i \rangle + \delta\mathbf{S}_i) \cdot (\langle \mathbf{S}_j \rangle + \delta\mathbf{S}_j) \\ &= \langle \mathbf{S}_i \rangle \cdot \delta\mathbf{S}_j + \delta\mathbf{S}_i \cdot \langle \mathbf{S}_j \rangle + \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle + \delta\mathbf{S}_i \cdot \delta\mathbf{S}_j \\ &= \langle \mathbf{S}_i \rangle \cdot (\mathbf{S}_j - \langle \mathbf{S}_j \rangle) + \mathbf{S}_i - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle + \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle + \delta\mathbf{S}_i \cdot \delta\mathbf{S}_j \\ &= \mathbf{S}_i \cdot \langle \mathbf{S}_j \rangle + \langle \mathbf{S}_i \rangle \cdot \mathbf{S}_j - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle + (\mathbf{S}_i - \langle \mathbf{S}_i \rangle) \cdot (\mathbf{S}_j - \langle \mathbf{S}_j \rangle) \end{aligned} \quad (3.188)$$

The first two terms have the correct structure of the product of a single spin with an averaged background field. The third term can be considered as a mere energy shift, with no physical contribution and finally the last term describes spin–spin correlation. In the mean field approximation we will replace the correlation term by its average and therefore neglect any correlations between the spins

$$\langle \delta \mathbf{S}_i \cdot \delta \mathbf{S}_j \rangle = \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle \quad (3.189)$$

This term is also an additional energy shift and can be safely neglected. Applying the mean field approximation, we end up with the following Hamiltonian

$$\begin{aligned} \mathcal{H}^{\text{Mean}} &= \sum_{i=1}^N \mathcal{H}_i = 2 \sum_{i=1}^N \left[J_{\perp} \left(\underbrace{h_{ix}}_{\langle S_{ix} \rangle} S_{ix} + \underbrace{h_{iy}}_{\langle S_{iy} \rangle} S_{iy} \right) + J_z \underbrace{h_{iz}}_{\langle S_{iz} \rangle} S_{iz} \right. \\ &\quad \left. - J_{\perp} (h_{ix} \langle S_{iy} \rangle + h_{iy} \langle S_{ix} \rangle) - J_z h_{iz} \langle S_{iz} \rangle \right] \\ &\approx \sum_{i=1}^N J_{\perp} \left[(h_{ix} S_{ix} + h_{iy} S_{iy}) + J_z h_{iz} S_{iz} \right] \end{aligned} \quad (3.190)$$

The self-consistency condition determines the magnetic background field \mathbf{h}_i acting on the spin occupying the i th lattice site. In the ferromagnetic case we assume the mean field ground state consists of all spins aligned parallel in z -direction. Thus the magnetic field is given by

$$\mathbf{h}_i^{\text{F}} \equiv \mathbf{h}^{\text{F}} = \sum_{n \neq 0} \frac{\langle \mathbf{S}_n \rangle}{|\mathbf{R}_n|^3} = \sum_{n \neq 0} \frac{\langle \uparrow \uparrow \uparrow \dots | \mathbf{S}_n | \uparrow \uparrow \uparrow \dots \rangle}{|\mathbf{R}_n|^3} = \langle \mathbf{S} \rangle \sum_{n \neq 0} \frac{1}{|\mathbf{R}_n|^3} = \langle \mathbf{S} \rangle g^{\text{F}} \quad (3.191)$$

where $n = i - j$ and $|\mathbf{R}_n| = |\mathbf{R}_i - \mathbf{R}_j|$ due to the translational invariance of the lattice. Since the components of the homogeneous magnetic field are all positive, the coupling constants J_z and J_{\perp} must be negative to minimize the mean field energy as we would have been expected for the ferromagnetic case. If we would have taken the other possible ferromagnetic ground state $|\downarrow \downarrow \downarrow \dots\rangle$, the components of \mathbf{h}^{F} would have turned out to be negative in agreement with the expectation value of the single spin operators and the ferromagnetic coupling constants. The mean field ground state energy can be calculated

$$\begin{aligned} \langle \uparrow \uparrow \uparrow \dots | \mathcal{H}^{\text{F}} | \uparrow \uparrow \uparrow \dots \rangle &= \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{J_{\perp}}{2} \frac{\langle \uparrow \uparrow \uparrow \dots | S_{i+} S_{j-} + S_{i-} S_{j+} | \uparrow \uparrow \uparrow \dots \rangle}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\ &\quad \left. + J_z \frac{\langle \uparrow \uparrow \uparrow \dots | S_{iz} S_{jz} | \uparrow \uparrow \uparrow \dots \rangle}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \\ &= J_z \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{\langle S_z \rangle \langle S_z \rangle}{|\mathbf{R}_i - \mathbf{R}_j|^3} = J_z \langle S_z \rangle^2 \sum_{i=1}^N \sum_{\substack{j=1 \\ j \neq i}}^N \frac{1}{|\mathbf{R}_i - \mathbf{R}_j|^3} \\ &= J_z \left(\frac{\hbar}{2} \right)^2 N g^{\text{F}} = N g^{\text{F}} J_z S^2 = E_{0\text{F}}^z \end{aligned} \quad (3.192)$$

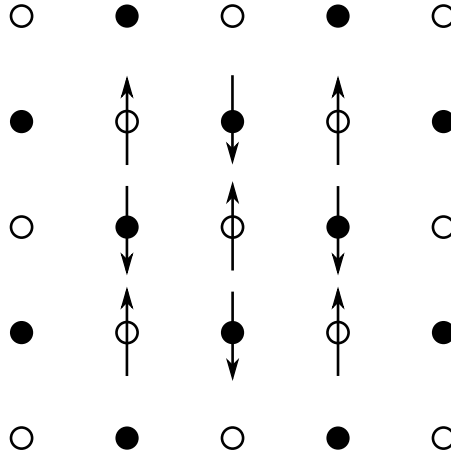


Figure 3.14 The Néel ordered ground state is divided in two sublattices • and ○. For the ground state $|\uparrow\downarrow\uparrow\dots\rangle$ the spins on the ○-sublattice are pointing up.

and is in agreement with the result of the spin wave analysis with $J'_z = \hbar^2 J_z$ and $S^2 = 1/4$. In fact the spin wave analysis delivered the functional dependency on ϑ of the mean field ground state energy for free which we now have verified. The mean field energy of the other possible ground state $|\downarrow\downarrow\downarrow\dots\rangle$ is identical since $\langle S_z \rangle^2$ is invariant under sign changes.

In the antiferromagnetic case, we start with an Néel ordered ground state $|\uparrow\downarrow\uparrow\dots\rangle$. If we take the spin on site i to be pointing up, all nearest neighbor spins must point down and the next nearest neighbors in the second shell are again pointing up. This can be carried out for all higher shells and we find that for the sum of $i+j$ = (an even number) the spins are pointing up and for $i+j$ = (an odd number) the spins are pointing down. In order to have a self-consistent solution, the magnetic background field \mathbf{h}_i^{AF} acting on site i must be negative since the antiferromagnetic coupling constants are positive

$$\begin{aligned} \mathbf{h}_i^{\text{AF}} &= (-1)^i \sum_{n \neq 0}^N \frac{\langle \mathbf{S}_n \rangle}{|\mathbf{R}_n|^3} = (-1)^i \sum_{n \neq 0}^N \frac{(-1)^n}{|\mathbf{R}_n|^3} \begin{pmatrix} |S_x| \\ |S_y| \\ |S_z| \end{pmatrix} \\ &= (-1)^i g \begin{pmatrix} |S_x| \\ |S_y| \\ |S_z| \end{pmatrix} = (-1)^{i+1} |g| \begin{pmatrix} |S_x| \\ |S_y| \\ |S_z| \end{pmatrix} \end{aligned} \quad (3.193)$$

Calculating the mean field ground state energy

$$\begin{aligned} \langle \uparrow\downarrow\uparrow\dots | \mathcal{H}^{\text{AF}} | \uparrow\downarrow\uparrow\dots \rangle &= \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{J_{\perp}}{2} \frac{\langle \uparrow\downarrow\uparrow\dots | S_{i+} S_{j-} + S_{i-} S_{j+} | \uparrow\downarrow\uparrow\dots \rangle}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\ &\quad \left. + J_z \frac{\langle \uparrow\downarrow\uparrow\dots | S_{i_z} S_{j_z} | \uparrow\downarrow\uparrow\dots \rangle}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \end{aligned}$$

Magnetical Ordering	Mean Field	Spin Wave
ferro in-axis	$-Ng^F J'_z S^2$	$-Ng^F J'_z S^2$
ferro in-plane	$-Ng^F J'_\perp S^2$	$-Ng^F J'_\perp S \left[(S+1) + \frac{a^2}{4\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k})}{g^F}\right) \left(1 - \frac{J'_z \epsilon(\mathbf{k})}{J'_\perp g^F}\right)} \right]$
antiferro in-axis	$-N g J'_z S^2$	$-N g J'_z S \left[(S+1) + \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{J'_z \epsilon(\mathbf{k}+\mathbf{K})}{g}\right) \left(1 - \frac{J'_z \epsilon(\mathbf{k})}{g}\right)} \right]$
antiferro in-plane	$-N g J'_\perp S^2$	$-N g J'_\perp S \left[(S+1) + \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k}+\mathbf{K})}{g}\right) \left(1 - \frac{J'_z \epsilon(\mathbf{k})}{J'_\perp g}\right)} \right]$

Table 3.3 Overview over the mean field and the spin wave ground state energy for the $1/R^3$ -interaction. Only for the in-axis ferromagnet both energies are identical. The mean field energy for the nearest neighbor interaction can be found via replacing g^F or $|g|$ with the coordination number z .

$$\begin{aligned}
&= J_z \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{(-1)^i |\langle \mathcal{S}_z \rangle| (-1)^j |\langle \mathcal{S}_z \rangle|}{|\mathbf{R}_i - \mathbf{R}_j|^3} = J_z |\langle \mathcal{S}_z \rangle|^2 \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{(-1)^{i+j}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \\
&= J_z \langle \mathcal{S}_z \rangle^2 \sum_{i=1}^N \sum_{n \neq 0}^N \frac{(-1)^n}{|\mathbf{R}_n|^3} = NgJ'_z S^2 = E_{\text{AF-mean}}^z \quad (3.194)
\end{aligned}$$

which is also in agreement with the “classical” ground state energy ignoring the quantum fluctuations obtained in the spin wave analysis in Section 3.3. For the other possible antiferromagnetic mean field ground state $|\downarrow\uparrow\downarrow\dots\rangle$ we only have to shift the index i and j to $i+1$ and $j+1$. Since $(-1)^2 = 1$ the ground state energy is identical and the two different ground states are degenerate.

In order to calculate the mean field energy of the in-plane magnetically ordered ground states we can use the Hamiltonian define in Subsection 3.3.4. The Hamiltonian $\mathcal{H}^{\text{x-F}}$ and $\mathcal{H}^{\text{x-AF}}$ are constructed by interchanging \mathcal{S}_x and \mathcal{S}_z . Following the calculations presented in equation (3.194) and (3.192) we see that we only have to interchange $\cos\vartheta$ by $\sin\vartheta$ or J_z by J_\perp to get $E_{\text{F-mean}}^{\text{x}}$ and $E_{\text{AF-mean}}^{\text{x}}$

$$E_{\text{F-mean}}^{\text{x}} = Ng^F J'_\perp S^2 \quad E_{\text{AF-mean}}^{\text{x}} = NgJ'_\perp S^2 \quad (3.195)$$

The mean field ground state energies are compared to the spin wave ground state energies in Table 3.3. There is an additional contribution in the spin wave ground state energy originating from the quantum fluctuation, except for the in-axis ferromagnetic case where both energies are identical. These corrections are numerically determined with the help of MATHEMATICA in Section 3.5.

In general we can show that for a two dimensional square lattice the mean field energy can be

written as

$$\langle \text{MF} | \mathcal{H} | \text{MF} \rangle = \left[J_{\perp} \left(\langle S_x \rangle^2 + \langle S_y \rangle^2 \right) + J_z \langle S_z \rangle^2 \right] \sum_{\substack{i,j=1 \\ i \neq j}}^N \frac{g(i-j)}{|\mathbf{R}_i - \mathbf{R}_j|^3} \quad (3.196)$$

with the continuous function $g \in [-1, 1]$ describing the relative orientation of two spins at i and j and the constraint

$$\sum_{n \neq 0}^N |g(n)|^2 \leq N \quad (3.197)$$

since the sum over the g cannot exceed the number of particles in the lattice. Expressing the sum with the help of the Poisson formula (F.23) from Appendix F.4 setting $q = 0$, we find

$$\sum_{\substack{\mathbf{R}_N \\ \mathbf{R}_n \neq 0}} \frac{g(n)}{|\mathbf{R}_n|^3} = \frac{1}{a^2} \sum_{\mathbf{k}} \tilde{g}(\mathbf{k}) \epsilon(\mathbf{k}) = \frac{1}{a^2} \left(\frac{\sqrt{Na}}{2\pi} \right)^2 \int d^2 \mathbf{k} \tilde{g}(\mathbf{k}) \epsilon(\mathbf{k}) \quad (3.198)$$

and inserting this integral in the mean field energy gives rise to

$$\langle \text{MF} | \mathcal{H} | \text{MF} \rangle = N \left[J_{\perp} \left(\langle S_x \rangle^2 + \langle S_y \rangle^2 \right) + J_z \langle S_z \rangle^2 \right] \int \frac{d^2 \mathbf{k}}{(2\pi)^2} \tilde{g}(\mathbf{k}) \epsilon(\mathbf{k}) \quad (3.199)$$

The constraint (3.197) can be written in terms of the Fourier transform $\tilde{g}(\mathbf{k})$ as

$$N \geq \sum_{n \neq 0} |g(n)|^2 = \sum_{\mathbf{k}} |\tilde{g}(\mathbf{k})|^2 \leq N \quad (3.200)$$

In order to find the extrema of the mean field energy, it suffices to search for the extrema of the integrand $\tilde{g}(\mathbf{k}) \epsilon(\mathbf{k})$. The integrand will be extremal if both factors are extremal. The extremal value of $\tilde{g}(\mathbf{k})$ which is in agreement with the constraint is

$$|g(n)|^2 = 1 \quad \Leftrightarrow \quad \sum_{n \neq 0}^N |g(n)|^2 = N \quad (3.201)$$

$$|\tilde{g}(\mathbf{k})|^2 = 1 \quad \Leftrightarrow \quad \sum_{\mathbf{k}} |\tilde{g}(\mathbf{k})|^2 = N \quad (3.202)$$

Thus the mean field ground state can only be a product state of spins aligned parallel or antiparallel, i.e. ferromagnetically or antiferromagnetically ordered. The extrema of $\epsilon(\mathbf{k})$ are at

$$\begin{aligned} \mathbf{k}_F = 0 & \quad \Leftrightarrow \quad \epsilon(\mathbf{0}) \text{ is local maxima with } \epsilon(\mathbf{0}) > 0 \\ \mathbf{k}_{AF} = 0 & \quad \Leftrightarrow \quad \epsilon(\pi, \pi) \text{ is local minima with } \epsilon(\pi, \pi) < 0 \end{aligned} \quad (3.203)$$

and we see that the ferromagnetically ordered ground states are given by $\epsilon(\mathbf{0})$, and since the integral is maximized the coupling constants must be negative in order to minimize the mean field ground state energy. For positive coupling constants we need to minimize the integral which is done by $\epsilon(\pi, \pi)$. As one can see in Figure 3.5 there are no more extrema of $\epsilon(\mathbf{k})$ and thus no more possible mean field ground states. Looking at the four dispersion relations of the harmonic

excitations about those four ground states we find a general form of the dispersion relation

$$\omega_{\mathbf{q}}^z = 2J'S \sqrt{(\sin \vartheta \epsilon(\mathbf{k} + \mathbf{q}) - \cos \vartheta \epsilon(\mathbf{q})) (\sin \vartheta \epsilon(\mathbf{k}) - \cos \vartheta \epsilon(\mathbf{q}))} \quad (3.204)$$

$$\omega_{\mathbf{q}}^x = 2J'S \sqrt{\sin \vartheta (\epsilon(\mathbf{q}) - \epsilon(\mathbf{k} + \mathbf{q})) (\sin \vartheta \epsilon(\mathbf{q}) - \cos \vartheta \epsilon(\mathbf{k}))} \quad (3.205)$$

In order to arrive at the ferromagnetic dispersion relation we need to “expand” about $\mathbf{q} = \mathbf{0}$ and for the antiferromagnetic dispersion relation about $\mathbf{q} = (\pi, \pi)^t$. In the same way we can define a general form of the spin wave ground state energy given by

$$E_{0\mathbf{q}}^z = N\epsilon(\mathbf{q})J'_z S \left[(S+1) + \left(-\frac{1}{2}\right)^{1-\frac{|\mathbf{q}|}{\sqrt{2}\pi}} \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{J'_z \epsilon(\mathbf{k} + \mathbf{q})}{\epsilon(\mathbf{q})}\right) \left(1 - \frac{J'_z \epsilon(\mathbf{k})}{\epsilon(\mathbf{q})}\right)} \right] \quad (3.206)$$

$$E_{0\mathbf{q}}^x = N\epsilon(\mathbf{q})J'_\perp S \left[(S+1) + \left(\frac{1}{2}\right)^{1-\frac{|\mathbf{q}|}{\sqrt{2}\pi}} \frac{a^2}{2\pi^2} \int d^2\mathbf{k} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k} + \mathbf{q})}{\epsilon(\mathbf{q})}\right) \left(1 - \frac{J'_\perp \epsilon(\mathbf{k})}{\epsilon(\mathbf{q})}\right)} \right] \quad (3.207)$$

3.5 Phase Diagram of the XXZ Model

In the previous two subsection we have determined the mean field ground state and the linear spin wave excitations about these ground states. Now we can use these results to identify the different quantum phases and their type of phase transition, i.e. first order or discontinuous and second order or continuous respectively. The phase transition between the mean field (anti)ferromagnetic ground state aligned in z -direction and the mean field (anti)ferromagnetic ground state aligned in x -direction can be found by looking at the mean field ground state energy while varying the parameter ϑ

$$E_{\text{AF-MF}}^z(\vartheta) = -JN |g| \cos \vartheta \quad E_{\text{AF-MF}}^x(\vartheta) = -JN |g| \sin \vartheta \quad (3.208)$$

$$E_{\text{F-MF}}^z(\vartheta) = JNg^F \cos \vartheta \quad E_{\text{F-MF}}^x(\vartheta) = JNg^F \sin \vartheta \quad (3.209)$$

Since the ground state energy must be minimal the phase transition occurs at values of ϑ where two energies of different ground states are identical. This allows us to find possible phase transitions as intersection of ground state energies. Beginning with the...

- in-axis antiferromagnet to in-plane antiferromagnet transition

$$\begin{aligned} E_{\text{AF-MF}}^z(\vartheta_{P1}) &\stackrel{!}{=} E_{\text{AF-MF}}^x(\vartheta_{P1}) && \hookrightarrow JNg \cos \vartheta_{P1} = JNg \sin \vartheta_{P1} \\ \tan \vartheta_{P1} &= 1 && \hookrightarrow \vartheta_{P1} = \pi/2 \end{aligned} \quad (3.210)$$

- in-plane antiferromagnet to in-axis ferromagnet transition

$$\begin{aligned} E_{\text{AF-MF}}^x(\vartheta_{P2}) &\stackrel{!}{=} E_{\text{F-MF}}^z(\vartheta_{P2}) && \hookrightarrow JNg \sin \vartheta_{P2} = JNg^F \cos \vartheta_{P2} \\ \tan \vartheta_{P2} &= \frac{g^F}{g} = \frac{\epsilon(\mathbf{0})}{\epsilon(\mathbf{K})} && \hookrightarrow \vartheta_{P2} \approx 106.27^\circ \end{aligned} \quad (3.211)$$

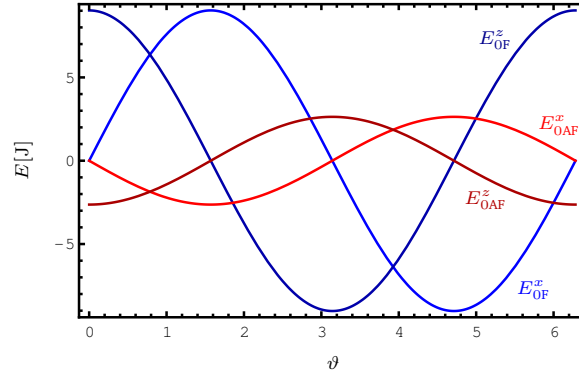


Figure 3.15 The mean field energy of all four possible ground states are shown in dependence of the parameter ϑ . There are four intersection points showing the critical angles ϑ_c where quantum phase transitions occur.

- in-axis ferromagnet to in-plane ferromagnet

$$\begin{aligned}
 E_{\text{F-MF}}^z(\vartheta_{\text{P3}}) &\stackrel{!}{=} E_{\text{F-MF}}^x(\vartheta_{\text{P3}}) && \hookrightarrow JNg^{\text{F}} \cos \vartheta_{\text{P3}} = JNg^{\text{F}} \sin \vartheta_{\text{P3}} \\
 \tan \vartheta_{\text{P3}} &= 1 && \hookrightarrow \vartheta_{\text{P3}} = \frac{5\pi}{4}
 \end{aligned} \tag{3.212}$$

- in-plane ferromagnet to in-axis antiferromagnet

$$\begin{aligned}
 E_{\text{F-MF}}^x(\vartheta_{\text{P4}}) &\stackrel{!}{=} E_{\text{AF-MF}}^z(\vartheta_{\text{P4}}) && \hookrightarrow JNg^{\text{F}} \sin \vartheta_{\text{P4}} = JNg \cos \vartheta_{\text{P3}} \\
 \tan \vartheta_{\text{P4}} &= \frac{g}{g^{\text{F}}} = \frac{\epsilon(\mathbf{K})}{\epsilon(\mathbf{0})} && \hookrightarrow \vartheta_{\text{P4}} \approx 343.73^\circ \hat{=} -16.27^\circ
 \end{aligned} \tag{3.213}$$

Compared to the phase transitions of the nearest neighbor interaction which occur symmetrically at $\pi/4$, $3\pi/4$, $5\pi/4$ and $7\pi/4$ the phase transition points between ferromagnetical and antiferromagnetical order are shifted towards the antiferromagnetic phases. Intuitively, it is clear that a long-range interaction favors the ferromagnetic ground state because in the antiferromagnetic case the spins on identical sublattices are also interacting with each other. Since spins on such sublattices are aligned parallel the antiferromagnetic ordering obtains an energy penalty that must be compensated by the respective positive coupling constant.

With the help of the spin wave dispersion relation, we can probe the long range ordering of the system and find indications about the type of the phase transition. If the dispersion relations at the phase transition are identical over the entire \mathbf{k} -space domain, we can expect a continuous or second order phase transition since there will be no energy gap between the two excitations. This is the case for the phase transitions between identical magnetical ordering, e.g. for $\vartheta_{\text{P1}} = \pi/4$ and $\vartheta_{\text{P3}} = 5\pi/4$, although the excitations about the in-axis ground state shows an finite energy gap in both cases. Since the transition takes place at the Heisenberg point, the $SU(2)$ symmetry allows a continuous transition that could be destroyed by explicitly breaking the $SU(2)$ symmetry of the Hamiltonian, e.g. by turning on an additional transverse magnetic field. Spontaneously breaking the continuous $SU(2)$ symmetry leads to gapless excitations the so called Goldstone modes,

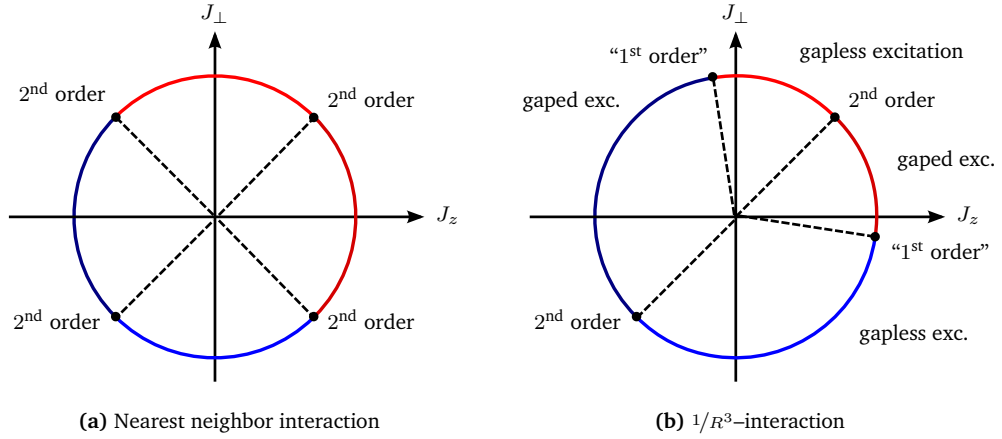


Figure 3.16 Due to the symmetric mapping between different coupling constant regimes the phase transitions of the nearest neighbor interaction is of 2nd order given by the $SU(2)$ symmetry of the Heisenberg point. The $1/R^3$ -interaction favors ferromagnetic ordered ground states that leads to an extension of the ferromagnetic domain. Independent of the interaction range the in-axis spin waves possess an excitation gap in the long wave length limit. Note that the transitions between the ferromagnetic and antiferromagnetic phases are only proposed to be of 1st order and that at those points there might be another unidentified phase.

i.e. fluctuations among the degenerate ground states. The magnetically ordered ground states posses a lower symmetry as the Hamiltonian, so we will expect gapless excitations. The deviation from the z -axis leads to a non-negligible energy cost, since the long wave length excitations are lowering the magnetization. Thus, we find gaped excitations in case of the in-axis ferromagnetic and antiferromagnetic spin waves for the $1/R^3$ -interaction as well as the nearest neighbor interaction. The in-plane excitations are gapless since the rotational symmetry in the xy -plane is not broken and spin flips can be performed continuously. Therefore the phase transitions at ϑ_{P2} and ϑ_{P4} could be of first order with non-identical spin wave excitations that lead to a jump in the long wave length energy.

Unfortunately, this simple mean field picture is not telling the whole truth about the real type of the phase transitions, because the effects of quantum fluctuations are not taken into account. A further complication arises from the fact, that at ϑ_{P2} and ϑ_{P4} the dispersion relation is becoming negative or imaginary, respectively, i.e. the spin waves are breaking down exactly at the alleged phase transitions. By looking at the quantum fluctuation, one finds the following corrections to the mean field ground state energies (3.208) and (3.209) at $\vartheta_{P4} = \arctan \epsilon(\mathbf{K})/\epsilon(\mathbf{0})$

$$E_{\text{OF}}^x(\vartheta_{P4}) = N\epsilon(\mathbf{0})J'_\perp S \left[(S+1) + \underbrace{\frac{a^2}{4\pi^2} \int d^2\mathbf{K} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{0})}\right) \left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{K})}\right)}}_{\approx 0.85895 a^2} \right] \quad (3.214)$$

$$E_{\text{OAF}}^z(\vartheta_{P4}) = N\epsilon(\mathbf{K})J'_\perp S \left[(S+1) + \underbrace{\frac{a^2}{2\pi^2} \int d^2\mathbf{K} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k} + \mathbf{K})}{\epsilon(\mathbf{0})}\right) \left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{0})}\right)}}_{\approx 1.93833 a^2} \right] \quad (3.215)$$

where $J_z/J_\perp = \cot \vartheta$ and $\cot(\arctan x) = x^{-1}$ has been used. Since the spin waves about the in-plane ferromagnetic ground states are collapsing, we cannot enter the antiferromagnetic domain with $\vartheta > \vartheta_{P4}$, although the ground state energy $E_{0F}^x(\vartheta_{P4})$ is smaller than $E_{0AF}^x(\vartheta_{P4})$. This gives rise to the existence of an intermediate phase that is bounded by a first order phase transition to the ferromagnetic domain and can be continuously transformed to the antiferromagnetic in-axis phase, indicating a second order phase transition. In the same way the peculiarities of the phase transition at $\vartheta_{P2} = \arctan \epsilon^{(0)}/\epsilon(\mathbf{K})$ can be investigated, using the spin wave ground state energies (3.206) and (3.207)

$$E_{0F}^{textz}(\vartheta_{P2}) = N\epsilon(\mathbf{0})J'_z S \quad (3.216)$$

$$E_{0AF}^x(\vartheta_{P2}) = N\epsilon(\mathbf{K})J'_\perp S \left[(S+1) + \underbrace{\frac{a^2}{2\pi^2} \int d^2\mathbf{K} \sqrt{\left(1 - \frac{\epsilon(\mathbf{k} + \mathbf{K})}{\epsilon(\mathbf{K})}\right) \left(1 - \frac{\epsilon(\mathbf{k})}{\epsilon(\mathbf{0})}\right)}}_{\approx 1.91089 a^2} \right] \quad (3.217)$$

Additionally, we see that at the points $\vartheta_{P1} = \pi/4$ and $\vartheta_{P3} = 5\pi/4$ the spin wave ground state energies are identical, because $J_z = J_\perp$ and the spin wave dispersion relation is also identical.

Magnetical Ordering	Dispersion Relation $\omega(\mathbf{k})$
ferro in-axis	$2J'S(\sin \vartheta\epsilon(\mathbf{k}) - \cos \vartheta\epsilon(\mathbf{0}))$
ferro in-plane	$2J'S\sqrt{\sin \vartheta(\epsilon(\mathbf{0}) - \epsilon(\mathbf{k}))(\sin \vartheta\epsilon(\mathbf{0}) - \cos \vartheta\epsilon(\mathbf{k}))}$
antiferro in-axis	$2J'S\sqrt{(\sin \vartheta\epsilon(\mathbf{k} + \mathbf{K}) - \cos \vartheta\epsilon(\mathbf{K}))(\sin \vartheta\epsilon(\mathbf{k}) - \cos \vartheta\epsilon(\mathbf{K}))}$
antiferro in-plane	$2J'S\sqrt{\sin \vartheta(\epsilon(\mathbf{K}) - \epsilon(\mathbf{k} + \mathbf{K}))(\sin \vartheta\epsilon(\mathbf{K}) - \cos \vartheta\epsilon(\mathbf{k}))}$ $2J'S\sqrt{(\sin \vartheta\epsilon(\mathbf{K}) - \cos \vartheta\epsilon(\mathbf{k} + \mathbf{K}))\sin \vartheta(\epsilon(\mathbf{K}) - \epsilon(\mathbf{k}))}$
Magnetical Ordering	Nearest Neighbor Dispersion Relation $\omega(\mathbf{k})$
ferro in-axis	$2S(2J'_\perp(\cos k_x + \cos k_y) - J'_z z)$
ferro in-plane	$2S\sqrt{J'_\perp(z - 2(\cos k_x + \cos k_y))(J'_\perp z - 2J'_z(\cos k_x + \cos k_y))}$
antiferro in-axis	$2S\sqrt{(J'_z z)^2 - (2J'_\perp(\cos k_x + \cos k_y))^2}$
antiferro in-plane	$2S\sqrt{(J'_\perp z + 2J'_z(\cos k_x + \cos k_y))J'_\perp(z - 2(\cos k_x + \cos k_y))}$ $2S\sqrt{(J'_\perp z - 2J'_z(\cos k_x + \cos k_y))J'_\perp(z + 2(\cos k_x + \cos k_y))}$

Table 3.4 Overview over the linear spin wave dispersion relations for the four different mean field ground states. As comparison the nearest neighbor dispersion relations are given in the lower part.

4

t - J Model

4.1 t - J Model for strongly correlated electrons

The t - J model is derived from the Hubbard model

$$\mathcal{H} = -t \sum_{\langle i,j \rangle \sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + c_{i\sigma} c_{j\sigma}^\dagger \right) + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad (4.1)$$

where t describes the hopping matrix element and U the Coulomb on-site repulsion of the electrons. In the strong coupling regime $t/U \ll 1$, we find for the limit of a Mott-Hubbard insulator, i.e. if the particle numbers are conserved and there is only single occupancy of a lattice site, that the t - J model can be written as

$$\mathcal{H} = -t \sum_{\langle i,j \rangle \sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + c_{i\sigma} c_{j\sigma}^\dagger \right) + J \sum_{i,j=1} \left(\mathbf{s}_i \cdot \mathbf{s}_j - \frac{1}{4} n_i n_j \right) \quad (4.2)$$

by the mapping

$$\sigma \longrightarrow c_{i\alpha}^\dagger \sigma_{\alpha\beta} c_{i\beta} \quad (4.3)$$

where $c_{i\alpha}^\dagger$ and $c_{i\alpha}$ are projected fermionic creation and annihilation operators with non-fermionic commutation relations [17]. Carrying out the mapping for the components of a spin-1/2 system gives rise to

$$\begin{aligned} \sigma_{ix} &\longrightarrow \begin{pmatrix} c_{i\uparrow}^\dagger \\ c_{i\downarrow}^\dagger \end{pmatrix} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} c_{i\uparrow} \\ c_{i\downarrow} \end{pmatrix} = c_{i\uparrow}^\dagger c_{i\downarrow} + c_{i\downarrow}^\dagger c_{i\uparrow} \\ \sigma_{iz} &\longrightarrow \begin{pmatrix} c_{i\uparrow}^\dagger \\ c_{i\downarrow}^\dagger \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} c_{i\uparrow} \\ c_{i\downarrow} \end{pmatrix} = c_{i\uparrow}^\dagger c_{i\uparrow} - c_{i\downarrow}^\dagger c_{i\downarrow} = n_{i\uparrow} - n_{i\downarrow} \end{aligned} \quad (4.4)$$

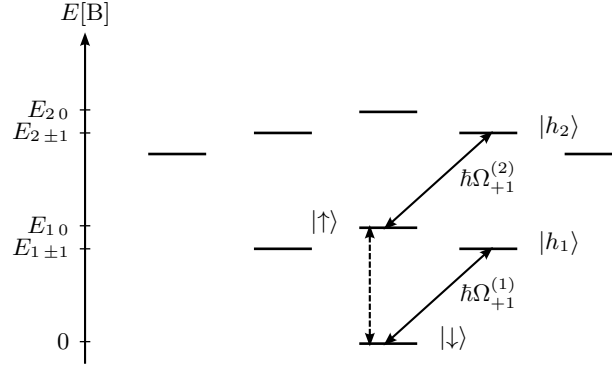


Figure 4.1 An attempt to realize the t - J model is given by the strong coupling of $|\phi_{00}\rangle \longleftrightarrow |\phi_{11}\rangle$ and $|\phi_{10}\rangle \longleftrightarrow |\phi_{21}\rangle$ in order to obtain three independent tuning parameters

4.2 Realizing the t - J Model with Polar Molecules

If we do not fill up all lattice points with molecules, that is we dope the lattice with holes, we can allow the molecules to hop from one site to the nearest neighbor site. For strongly correlated systems (e.g. non-Fermi liquids) this can be described by the t - J model. In order to realize the t - J model we need three tuning parameters to satisfy the three conditions

$$h_z = 0 \quad J'_\perp - J'_z = 0 \quad 4J_\perp + J_z = 0 \quad (4.5)$$

Therefore we propose the following setup shown in Figure 4.1. We first couple the states $|\phi_{10}\rangle \longleftrightarrow |\phi_{21}\rangle$ and $|\phi_{00}\rangle \longleftrightarrow |\phi_{11}\rangle$ strongly by irradiating two independent circular polarized microwave fields with polarization $q = 1$ and frequency ω_{L1} and ω_{L2} , respectively. In the rotating wave approximation we can write the linear combination of the coupled states in the following way

$$|+\rangle_2 = \cos \frac{\varphi_2}{2} |\phi_{21}\rangle + \sin \frac{\varphi_2}{2} |\phi_{10}\rangle \quad |-\rangle_2 = -\sin \frac{\varphi_2}{2} |\phi_{21}\rangle + \cos \frac{\varphi_2}{2} |\phi_{10}\rangle \quad (4.6)$$

$$|+\rangle_1 = \cos \frac{\varphi_1}{2} |\phi_{11}\rangle + \sin \frac{\varphi_1}{2} |\phi_{00}\rangle \quad |-\rangle_1 = -\sin \frac{\varphi_1}{2} |\phi_{11}\rangle + \cos \frac{\varphi_1}{2} |\phi_{00}\rangle \quad (4.7)$$

with

$$\begin{aligned} \varphi_1 &= \frac{1}{2} \arctan \left(\frac{\Omega_1^{(1)}}{\Delta_1} \right) & \varphi_2 &= \frac{1}{2} \arctan \left(\frac{\Omega_1^{(2)}}{\Delta_2} \right) \\ \Omega_1^{(1)} &= \frac{2 \langle \phi_{11} | \mathbf{d}_1 | \phi_{10} \rangle E_{ac}}{\hbar} & \Delta_1 &= \omega_{L1} - \omega_1 \\ \Omega_1^{(2)} &= \frac{2 \langle \phi_{21} | \mathbf{d}_1 | \phi_{10} \rangle E_{ac}}{\hbar} & \Delta_2 &= \omega_{L2} - \omega_2 \end{aligned} \quad (4.8)$$

Now we weakly couple each of the ground states $|-\rangle_2 \equiv |e\rangle$ and $|-\rangle_1 \equiv |g\rangle$ of the strongly coupled states

$$\begin{aligned} |e\rangle &= \alpha_2 |\uparrow\rangle + \beta_2 e^{i\omega_{L2}t} |h_2\rangle \\ |g\rangle &= \alpha_1 |\downarrow\rangle + \beta_1 e^{i\omega_{L1}t} |h_1\rangle \end{aligned} \quad (4.9)$$

by a third linear polarized microwave field with ω_L

$$|\tilde{g}\rangle = e^{i\omega_L t} |g\rangle \quad |\tilde{e}\rangle = e^{-i\omega_L t} |e\rangle \quad (4.10)$$

The coefficients α_i and β_i are given by

$$\alpha_i = \cos\left(\frac{1}{2} \arctan\left(\frac{\Omega_1^{(i)}}{\Delta_i}\right)\right) \quad \beta_i = -\sin\left(\frac{1}{2} \arctan\left(\frac{\Omega_1^{(i)}}{\Delta_i}\right)\right) \quad (4.11)$$

Calculating the matrix elements of the dipole-dipole interaction operator (2.122)

$$\mathcal{V}_{ij}^{\text{dd}} = \frac{\mathbf{d}_0 \otimes \mathbf{d}_0 - \frac{3}{2}(\mathbf{d}_1 \otimes \mathbf{d}_1 + \mathbf{d}_{-1} \otimes \mathbf{d}_{-1}) + \frac{1}{2}(\mathbf{d}_1 \otimes \mathbf{d}_{-1} + \mathbf{d}_{-1} \otimes \mathbf{d}_1)}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \quad (4.12)$$

we have to construct the two particle states

$$\begin{aligned} |\tilde{e}\tilde{e}\rangle &= e^{-2i\omega_L t} |ee\rangle & |\tilde{e}\tilde{g}\rangle &= |eg\rangle \\ |\tilde{g}\tilde{g}\rangle &= e^{2i\omega_L t} |gg\rangle & |\tilde{g}\tilde{e}\rangle &= |ge\rangle \end{aligned} \quad (4.13)$$

where

$$\begin{aligned} |ee\rangle &= \alpha_2^2 |\uparrow\uparrow\rangle + \alpha_2\beta_2 e^{i\omega_{L2}t} (|\uparrow h_2\rangle + |h_2 \uparrow\rangle) + \beta_2^2 e^{2i\omega_{L2}t} |h_2 h_2\rangle \\ |eg\rangle &= \alpha_1\alpha_2 |\uparrow\downarrow\rangle + \alpha_2\beta_1 e^{i\omega_{L1}t} |\uparrow h_1\rangle + \alpha_1\beta_2 e^{i\omega_{L2}t} |h_2 \downarrow\rangle + \beta_1\beta_2 e^{i(\omega_{L1}+\omega_{L2})t} |h_2 h_1\rangle \\ |ge\rangle &= \alpha_1\alpha_2 |\downarrow\uparrow\rangle + \alpha_1\beta_2 e^{i\omega_{L2}t} |\downarrow h_2\rangle + \alpha_2\beta_1 e^{i\omega_{L1}t} |h_1 \uparrow\rangle + \beta_1\beta_2 e^{i(\omega_{L1}+\omega_{L2})t} |h_1 h_2\rangle \\ |gg\rangle &= \alpha_1^2 |\downarrow\downarrow\rangle + \alpha_1\beta_1 e^{i\omega_{L1}t} (|\downarrow h_1\rangle + |h_1 \downarrow\rangle) + \beta_1^2 e^{2i\omega_{L1}t} |h_1 h_1\rangle \end{aligned} \quad (4.14)$$

Due to the hermitian symmetry of the interaction operator $\mathcal{V}^{\text{dd}\dagger} = \mathcal{V}^{\text{dd}}$ and the dipole moments, we only have to calculate ten matrix elements. By applying the rotating wave approximation for the weak coupling, i.e. $e^{i\omega_L t} \approx 0$ and $e^{2i\omega_L t} \approx 0$ only five matrix elements are left, see equation (3.12)

$$\begin{aligned} \langle \tilde{e}\tilde{e} | \mathcal{V}^{\text{dd}} | \tilde{e}\tilde{e} \rangle &= \langle ee | \mathcal{V}^{\text{dd}} | ee \rangle \approx \alpha_2^4 \langle \uparrow\uparrow | \mathcal{V}^{\text{dd}} | \uparrow\uparrow \rangle + \alpha_2^2\beta_2^2 \left(\langle \uparrow h_2 | \mathcal{V}^{\text{dd}} | \uparrow h_2 \rangle \right. \\ &\quad \left. + 2 \langle \uparrow h_2 | \mathcal{V}^{\text{dd}} | h_2 \uparrow \rangle + \langle h_2 \uparrow | \mathcal{V}^{\text{dd}} | h_2 \uparrow \rangle \right) \\ &\quad + \beta_2^4 \langle h_2 h_2 | \mathcal{V}^{\text{dd}} | h_2 h_2 \rangle \end{aligned} \quad (4.15)$$

$$\begin{aligned} \langle \tilde{e}\tilde{g} | \mathcal{V}^{\text{dd}} | \tilde{e}\tilde{g} \rangle &= \langle eg | \mathcal{V}^{\text{dd}} | eg \rangle \approx \alpha_1^2\alpha_2^2 \langle \uparrow\downarrow | \mathcal{V}^{\text{dd}} | \uparrow\downarrow \rangle + \alpha_2^2\beta_1^2 \langle \uparrow h_1 | \mathcal{V}^{\text{dd}} | \uparrow h_1 \rangle \\ &\quad + \alpha_1^2\beta_2^2 \langle h_2 \downarrow | \mathcal{V}^{\text{dd}} | h_2 \downarrow \rangle + \beta_1^2\beta_1^2 \langle h_1 h_2 | \mathcal{V}^{\text{dd}} | h_1 h_2 \rangle \end{aligned} \quad (4.16)$$

$$\begin{aligned} \langle \tilde{g}\tilde{e} | \mathcal{V}^{\text{dd}} | \tilde{g}\tilde{e} \rangle &= \langle ge | \mathcal{V}^{\text{dd}} | ge \rangle \approx \alpha_1^2\alpha_2^2 \langle \uparrow\downarrow | \mathcal{V}^{\text{dd}} | \downarrow\uparrow \rangle + \alpha_2^2\beta_1^2 \langle \uparrow h_1 | \mathcal{V}^{\text{dd}} | h_1 \uparrow \rangle \\ &\quad + \alpha_1^2\beta_2^2 \langle h_2 \downarrow | \mathcal{V}^{\text{dd}} | \downarrow h_2 \rangle + \beta_1^2\beta_1^2 \langle h_2 h_1 | \mathcal{V}^{\text{dd}} | h_1 h_2 \rangle \end{aligned} \quad (4.17)$$

$$\begin{aligned} \langle \tilde{g}\tilde{e} | \mathcal{V}^{\text{dd}} | \tilde{g}\tilde{e} \rangle &= \langle ge | \mathcal{V}^{\text{dd}} | ge \rangle \approx \alpha_1^2 \alpha_2^2 \langle \downarrow \uparrow | \mathcal{V}^{\text{dd}} | \downarrow \uparrow \rangle + \alpha_1^2 \beta_2^2 \langle \downarrow h_2 | \mathcal{V}^{\text{dd}} | \downarrow h_2 \rangle \\ &\quad + \alpha_2^2 \beta_1^2 \langle h_1 \uparrow | \mathcal{V}^{\text{dd}} | h_1 \uparrow \rangle + \beta_1^2 \beta_2^2 \langle h_1 h_2 | \mathcal{V}^{\text{dd}} | h_1 h_2 \rangle \end{aligned} \quad (4.18)$$

$$\begin{aligned} \langle \tilde{e}\tilde{e} | \mathcal{V}^{\text{dd}} | \tilde{e}\tilde{e} \rangle &= \langle ee | \mathcal{V}^{\text{dd}} | ee \rangle \approx \alpha_1^4 \langle \downarrow \downarrow | \mathcal{V}^{\text{dd}} | \downarrow \downarrow \rangle + \alpha_1^2 \beta_1^2 (\langle \downarrow h_1 | \mathcal{V}^{\text{dd}} | \downarrow h_1 \rangle \\ &\quad + 2 \langle \downarrow h_1 | \mathcal{V}^{\text{dd}} | h_1 \downarrow \rangle + \langle h_1 \downarrow | \mathcal{V}^{\text{dd}} | h_1 \downarrow \rangle) \\ &\quad + \beta_1^4 \langle h_1 h_1 | \mathcal{V}^{\text{dd}} | h_1 h_1 \rangle \end{aligned} \quad (4.19)$$

whereas all other matrix elements are vanishing in the rotating wave approximation. Now the matrix elements of the dipole operator \mathbf{d} must be calculated. Therefore we define the following operator

$$\mathcal{D} = a^3 |\mathbf{R}_i - \mathbf{R}_j|^3 \mathcal{V}^{\text{dd}} \quad (4.20)$$

and evaluate the five matrix elements using the symmetries stated in equation (2.57), especially $\tilde{d}_{-1} = -\tilde{d}_1^\dagger$ to eliminate \tilde{d}_{-1} and that \tilde{d}_0 only couple states with $\Delta M = 0$ while \tilde{d}_1 only couples states with $\Delta M = 1$

$$\begin{aligned} \tilde{D}^{\text{ee}} &= \langle ee | \mathcal{D} | ee \rangle = \alpha_2^4 (d_0^{\uparrow\uparrow})^2 + \alpha_2^2 \beta_2^2 \left[2d_0^{\uparrow\uparrow} d_0^{22} - (d_1^{2\uparrow})^2 \right] + \beta_2^4 (d_0^{22})^2 \\ \tilde{D}^{\text{eg}} &= \langle eg | \mathcal{D} | eg \rangle = \alpha_1^2 \alpha_2^2 d_0^{\uparrow\uparrow} d_0^{\downarrow\downarrow} + \alpha_2^2 \beta_1^2 d_0^{\uparrow\uparrow} d_0^{11} + \alpha_1^2 \beta_2^2 d_0^{\downarrow\downarrow} d_0^{22} + \beta_1^2 \beta_2^2 d_0^{11} d_0^{22} \\ \tilde{D}^{\text{gg}} &= \langle gg | \mathcal{D} | gg \rangle = \alpha_1^4 (d_0^{\downarrow\downarrow})^2 + \alpha_1^2 \beta_1^2 \left[2d_0^{\downarrow\downarrow} d_0^{11} - (d_1^{1\downarrow})^2 \right] + \beta_1^4 (d_0^{11})^2 \\ \tilde{D}^{\text{egge}} &= \langle eg | \mathcal{D} | ge \rangle = \alpha_1^2 \alpha_2^2 (d_0^{\uparrow\downarrow})^2 - \frac{1}{2} \left[\alpha_1^2 \beta_2^2 (d_1^{2\downarrow})^2 + \alpha_2^2 \beta_1^2 (d_1^{1\uparrow})^2 \right] + \beta_1^2 \beta_2^2 (d_0^{12})^2 \end{aligned} \quad (4.21)$$

where

$$\begin{aligned} d_0^{\uparrow\uparrow} &= \langle \uparrow | \tilde{d}_0 | \uparrow \rangle & d_0^{22} &= \langle h_2 | \tilde{d}_0 | h_2 \rangle \\ d_0^{\uparrow\downarrow} &= \langle \uparrow | \tilde{d}_0 | \downarrow \rangle & d_0^{12} &= \langle h_1 | \tilde{d}_0 | h_2 \rangle \\ d_0^{\downarrow\downarrow} &= \langle \downarrow | \tilde{d}_0 | \downarrow \rangle & d_0^{11} &= \langle h_1 | \tilde{d}_0 | h_1 \rangle \end{aligned} \quad (4.22)$$

and

$$d_1^{2\uparrow} = \langle h_2 | \tilde{d}_1 | \uparrow \rangle \quad d_1^{1\downarrow} = \langle h_1 | \tilde{d}_1 | \downarrow \rangle \quad (4.23)$$

Due to the above mentioned symmetries the matrix element of \tilde{D}^{ge} and \tilde{D}^{eg} are identical, so that only four independent matrix elements remain. With the help of equation (3.17) and (3.18) the general form of the Hamiltonian in spin language is given as

$$\begin{aligned} \tilde{\mathcal{H}}_{ij} &= \tilde{\mathcal{H}}_i + \tilde{\mathcal{H}}_j + \mathcal{V}_{ij}^{\text{dd}} \\ &= 2E_{00} \mathbb{1} - \frac{\hbar\Delta}{2} (\sigma_{iz} + \sigma_{jz}) - \frac{\hbar\Omega_0^{\uparrow\downarrow}}{2} (\sigma_{ix} + \sigma_{jx}) \end{aligned}$$

$$\begin{aligned}
& + \frac{1}{4} \left(\tilde{D}^{ee} + 2\tilde{D}^{eg} + \tilde{D}^{gg} \right) \frac{\mathbb{1}_i \mathbb{1}_j}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{1}{2} \tilde{D}^{eg} \frac{\sigma_{ix} \sigma_{jx} + \sigma_{iy} \sigma_{jy}}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} \\
& + \frac{1}{4} \left(\tilde{D}^{ee} - \tilde{D}^{gg} \right) \frac{\sigma_{iz} + \sigma_{jz}}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{1}{4} \left(\tilde{D}^{ee} - 2\tilde{D}^{eg} + \tilde{D}^{gg} \right) \frac{\sigma_{iz} \sigma_{jz}}{a^3 |\mathbf{R}_i - \mathbf{R}_j|^3}
\end{aligned} \tag{4.24}$$

The total interaction Hamiltonian on a two dimensional square lattice can then be cast into

$$\begin{aligned}
\mathcal{H}_{\text{int}} &= \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \tilde{\mathcal{H}}_{ij} = \sum_{i=1}^N \tilde{\mathcal{H}}_i + \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^N \mathcal{V}_{ij}^{\text{dd}} \\
&= NE_{00} - \frac{\hbar}{2} \sum_{i=1}^N \left(\Delta \sigma_{iz} + \Omega_0^{\uparrow\downarrow} \sigma_{ix} \right) + \frac{\tilde{D}^{ee} - \tilde{D}^{gg}}{2a^3} \sum_{i=1}^N \sigma_{iz} \sum_{n \neq 0}^N \frac{1}{|\mathbf{R}_n|^3} \\
&\quad \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\tilde{D}^{eg} \text{ge}}{\hbar^2 a^3} \frac{\mathcal{S}_{ix} \mathcal{S}_{jx} + \mathcal{S}_{iy} \mathcal{S}_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{\tilde{D}^{ee} - 2\tilde{D}^{eg} + \tilde{D}^{gg}}{2\hbar^2 a^3} \frac{\mathcal{S}_{iz} \mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\
&\quad \left. + \frac{\tilde{D}^{ee} + 2\tilde{D}^{eg} + \tilde{D}^{gg}}{8a^3} \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right)
\end{aligned} \tag{4.25}$$

Up to now, there are four parameters α_1 , β_1 , α_2 and β_2 but since the states must be normalized two of the four parameters are not independent. Therefore we can choose

$$\begin{aligned}
\alpha_1 &= \cos \left(\frac{1}{2} \arctan \left(\frac{\Omega_1^{(1)}}{\Delta_1} \right) \right) & \beta_1 &= -\sqrt{1 - \alpha_1^2} \\
\alpha_2 &= \cos \left(\frac{1}{2} \arctan \left(\frac{\Omega_1^{(2)}}{\Delta_2} \right) \right) & \beta_2 &= -\sqrt{1 - \alpha_2^2}
\end{aligned} \tag{4.26}$$

In order to evaluate the range of the two independent parameters α_1 and α_2 , we can look at the two limiting cases $\Delta_i \rightarrow 0$ and $\Omega_1^{(i)} \rightarrow 0$

$$\lim_{\Delta \rightarrow 0} \arctan \left(\frac{\Omega_1^{(i)}}{\Delta_i} \right) = \frac{\pi}{2} \qquad \lim_{\Omega_1^{(i)} \rightarrow 0} \arctan \left(\frac{\Omega_1^{(i)}}{\Delta_i} \right) = 0 \tag{4.27}$$

and find for the range of the parameters

$$\alpha_i \in \left[\frac{1}{\sqrt{2}}, 1 \right] \qquad \beta_i \in \left[0, \frac{1}{\sqrt{2}} \right] \tag{4.28}$$

If we insert the two limiting cases in the definition of our linear combination of the coupled states, equation (4.9), we see that in the case of vanishing detuning both states of the linear combination, e.g. $|\uparrow\rangle$ and $|h_2\rangle$, are equally contributing, whereas in the case of vanishing coupling only the states $|\uparrow\rangle$ and $|\downarrow\rangle$ are left (which is exactly the setup for the realization of the XXZ model). Using the projected fermionic mapping (4.4) the total interaction Hamiltonian \mathcal{H}_{int} can

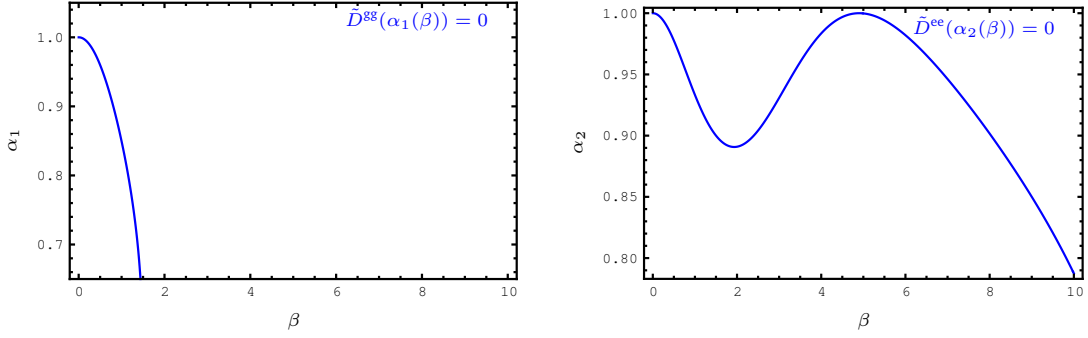


Figure 4.2 The conditions $h_z = 0$ and $4J_{\perp} + J_z = 0$ are fulfilled for the values of β given by the above curves. In the left figure the curve for $\tilde{D}^{\text{gg}}(\alpha_1(\beta)) = 0$ fixes $\alpha_1(\beta)$ and the curve in the right figure shows $\tilde{D}^{\text{ee}}(\alpha_2(\beta)) = 0$ which fixes $\alpha_2(\beta)$.

be recast

$$\begin{aligned}
\mathcal{H}_{\text{int}} = & NE_{00} - \frac{\hbar}{2} \sum_{i=1}^N \left[\Omega_0^{\uparrow\downarrow} (c_{i\uparrow}^\dagger c_{i\downarrow} + c_{i\downarrow}^\dagger c_{i\uparrow}) + \Delta (n_{i\uparrow} - n_{i\downarrow}) \right] + g^{\text{F}} \frac{\tilde{D}^{\text{ee}} - \tilde{D}^{\text{gg}}}{4a^3} \sum_{i=1}^N (n_{i\uparrow} - n_{i\downarrow}) \\
& + \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\tilde{D}^{\text{egge}}}{\hbar^2 a^3} \frac{\mathcal{S}_{ix} \mathcal{S}_{jx} + \mathcal{S}_{iy} \mathcal{S}_{jy}}{|\mathbf{R}_i - \mathbf{R}_j|^3} + \frac{\tilde{D}^{\text{ee}} - 2\tilde{D}^{\text{eg}} + \tilde{D}^{\text{gg}}}{2\hbar^2 a^3} \frac{\mathcal{S}_{iz} \mathcal{S}_{jz}}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right. \\
& \left. + \frac{\tilde{D}^{\text{ee}} + 2\tilde{D}^{\text{eg}} + \tilde{D}^{\text{gg}}}{8a^3} \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (4.29)
\end{aligned}$$

Ignoring the physically irrelevant energy shift and setting the detuning of the weak coupling Δ to zero, the effective interaction Hamiltonian can be written in terms of the three coupling constants J_{\perp} , J_z and J_{\parallel}

$$J_{\perp} = \frac{\tilde{D}^{\text{egge}}}{\hbar^2 a^3} \quad J_z = \frac{\tilde{D}^{\text{ee}} - 2\tilde{D}^{\text{eg}} + \tilde{D}^{\text{gg}}}{2\hbar^2 a^3} \quad J_{\parallel} = \frac{\tilde{D}^{\text{ee}} + 2\tilde{D}^{\text{eg}} + \tilde{D}^{\text{gg}}}{8a^3} \quad (4.30)$$

and compared to the exchange interaction part of the t - J model

$$J \sum_{\substack{i,j=1 \\ i \neq j}}^N \left(\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} - \frac{1}{4} \frac{n_i n_j}{|\mathbf{R}_i - \mathbf{R}_j|^3} \right) \quad (4.31)$$

The following conditions must be fulfilled, so that equation (4.29) and (4.31) match

$$h_z = 0 \quad \Leftrightarrow \quad \tilde{D}^{\text{ee}} - \tilde{D}^{\text{gg}} = 0 \quad (4.32a)$$

$$J_{\perp} - J_z = 0 \quad \Leftrightarrow \quad 2\tilde{D}^{\text{egge}} - \tilde{D}^{\text{ee}} + 2\tilde{D}^{\text{eg}} - \tilde{D}^{\text{gg}} = 0 \quad (4.32b)$$

$$4J_{\parallel} + J_z = 0 \quad \Leftrightarrow \quad \tilde{D}^{\text{ee}} + \tilde{D}^{\text{gg}} = 0 \quad (4.32c)$$

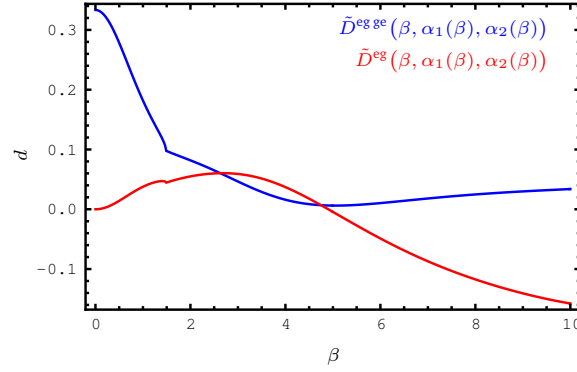


Figure 4.3 Plot of the last condition $J_{\perp} - J_z = 0$ that cannot be fulfilled for the given range of the parameter α since we cannot go to higher values of $\beta = 1.37$.

Condition (4.32a) and (4.32c) can only be fulfilled for $\tilde{D}^{ee} = 0$ and $\tilde{D}^{gg} = 0$. Reinserting these expressions in (4.32b) yields

$$\tilde{D}^{egge} + \tilde{D}^{eg} = 0 \quad (4.33)$$

Since we have three independently tunable parameter, α_1 which corresponds to the adjusted field strength E_{ac1} and frequency ω_{L1} of the applied microwave field coupling $|\phi_{00}\rangle \rightarrow |\phi_{11}\rangle$ and α_2 corresponding to the E_{ac2} and ω_{L2} , coupling $|\phi_{10}\rangle \rightarrow |\phi_{21}\rangle$. The third parameter corresponds to the field strength of the applied static field E_{dc} . We see that the two composite dipole moments \tilde{D}^{ee} and \tilde{D}^{gg} are only dependent on the applied microwave field of the respective coupling. Therefore we can determine $\alpha_1(\beta)$ from $\tilde{D}^{gg}(\alpha_1(\beta)) = 0$ and $\alpha_2(\beta)$ from $\tilde{D}^{ee}(\alpha_2(\beta)) = 0$ and insert the values into equation (4.33). Finally the parameter β will be fixed by

$$\tilde{D}^{egge}(\beta, \alpha_1(\beta), \alpha_2(\beta)) + \tilde{D}^{eg}(\beta, \alpha_1(\beta), \alpha_2(\beta)) = 0 \quad (4.34)$$

In Figure 4.2 the composite dipole moments are shown. As one can see in Figure 4.3 it is not possible with this setup to fulfill all conditions because inside the range $[1/\sqrt{2}, 1]$ of the parameter $\alpha_1(\beta)$ where $\tilde{D}^{gg}(\alpha_1(\beta)) = 0$ is zero, there is no combination of β that fulfills the condition (4.34). The minimum of $\tilde{D}^{egge} + \tilde{D}^{eg}$ describes the smallest deviation of J_z from J_{\perp} or vice versa and is approximately given by 0.0738 at $\beta = 1.37$ where $\alpha_1 = 1/\sqrt{2}$ or $\Delta_1 = 0$. Unfortunately this minimum leads to a ratio of $J_{\perp}/J_z \approx 2.58$ and therefore cannot be considered as a small anisotropy. Since this attempt to realize the t - J model fails, one could try to couple an additional state to the ground state $|\phi_{00}\rangle$, for example $|\phi_{1-1}\rangle$.

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5

Summary & Conclusion

In this work the possibility of realizing quantum simulators by using cold polar molecules trapped in optical lattices has been studied. It has been shown how to simulate spin models via the strong electric dipole–dipole interaction that can be tuned by external static fields to engineer desired spin interactions. Using the rotational excitation spectrum of polar molecules, at least three spin models, the antiferromagnetic XY model, the XXZ model and the Heisenberg model can be realized. The implications of the slowly decaying $1/r^3$ -interaction on the phase diagram of the general XXZ model has been investigated and the physical distinctions to a short-ranged nearest neighbor interaction have been worked out by employing a spin wave analysis. The magnetically ordered ground states of the XXZ model in dependence on the external static field are determined and their respective mean field ground states are calculated. In the second part the possibility to realize the t - J model of strongly correlated electrons is studied and an attempt for a concrete realization have been made. However, for an exact realization the setup has to be refined further, which is beyond the scope of this work, but could be done with little effort in a subsequent study.

Another interesting topic is the creation of ferroelectrics by coupling all four low-lying rotational states, that can yield interesting new physics. In this case one needs to couple all four states, the ground state of the dc-Stark shifted rotational spectrum and the first three excited states. This setup could then be used to create a liquid with a spontaneous electrical dipole moment, as has already be done with ferroelectric liquid crystals. The novelty of this setup would be to incorporate the advantages of using cold polar molecules as has been explained in the previous sections, to control and simulate such a ferroelectric liquid. As a starting point for a further study, the calculations presented in this work could be used.

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Spherical Tensor Operators

A.1 Clebsch–Gordan Coefficients

The Clebsch–Gordan coefficients are defined as a basis change from the product basis of two angular momenta to a common coupled momentum basis. Let $|j_1 m_1\rangle$ and $|j_2 m_2\rangle$ be the angular momentum of the first and the second particle, respectively. Then

$$|j_1 m_1\rangle \otimes |j_2 m_2\rangle \equiv |j_1 m_1, j_2 m_2\rangle \quad (\text{A.1})$$

defines the product basis of the two particle Hilbert space. Another possible basis of the two particle Hilbert space describes the total angular momentum \mathbf{J} of two coupled angular momenta with the corresponding state $|J M\rangle$. The total angular momentum is given by the sum of the two coupled angular momenta

$$\mathbf{J} = \mathbf{j}_1 + \mathbf{j}_2 \quad \mathcal{J}_z = j_{1z} + j_{2z} \quad (\text{A.2})$$

Since only \mathcal{J}^2 and \mathcal{J}_z are commuting and therefore share a common eigenbasis with eigenstates $|J M\rangle$, the total angular momentum must be squared

$$\mathcal{J}^2 = j_1^2 + j_2^2 + 2\mathbf{j}_1 \cdot \mathbf{j}_2 \quad (\text{A.3})$$

and therefore we see why two coupled angular momenta do not add but must be treated more complicated. The product state in equation (A.1) is no eigenstate of the total angular momentum operator. The change of basis from the product states to the total angular momentum eigenstates can be represented by an unitary transformation, since the product basis $\{|j_1 m_1, j_2 m_2\rangle\}$ is complete

$$|j_1 j_2, J M\rangle = \sum_{m_1=-j_1}^{j_1} \sum_{m_2=-j_2}^{j_2} |j_1 m_1, j_2 m_2\rangle \langle j_1 m_1, j_2 m_2 | j_1 j_2, J M\rangle \quad (\text{A.4})$$

The expansion coefficients of the coupled angular momentum state are called Clebsch–Gordan coefficients and can be chosen to be real. This is the Condon–Shortley phase convention that

rank	“polarization”	Unnormalized Spherical Harmonics $C_q^{(k)}$	Spherical Harmonics $Y_q^{(k)}$
$k = 0$	$q = 0$	1	$\sqrt{\frac{1}{4\pi}}$
$k = 1$	$q = 0$	$\cos \vartheta$	$\sqrt{\frac{3}{4\pi}} \cos \vartheta$
	$q = \pm 1$	$\mp \frac{1}{\sqrt{2}} e^{\pm i\varphi} \sin \vartheta$	$\mp \sqrt{\frac{3}{8\pi}} \sin \vartheta e^{\pm i\varphi}$
$k = 2$	$q = 0$	$\frac{1}{2} (3 \cos^2 \vartheta - 1)$	$\sqrt{\frac{5}{16\pi}} (3 \cos^2 \vartheta - 1)$
	$q = \pm 1$	$\mp \sqrt{\frac{3}{2}} \cos \vartheta \sin \vartheta e^{\pm i\varphi}$	$\mp \sqrt{\frac{15}{8\pi}} \cos \vartheta \sin \vartheta e^{\pm i\varphi}$
	$q = \pm 2$	$\frac{1}{2} \sqrt{\frac{3}{2}} \sin^2 \vartheta e^{\pm 2i\varphi}$	$\sqrt{\frac{15}{32\pi}} \sin^2 \vartheta e^{\pm 2i\varphi}$

Table A.1 List of the $k = 0, 1, 2$ unnormalized spherical harmonics $C_q^{(k)}$ and normalized spherical harmonics $Y_q^{(k)}$ as defined in equation (A.11).

includes a factor $(-1)^m$ in the associated Legendre functions (A.10) or in the definition of the spherical harmonics (A.12). The new states couple j_1 and j_2 to the total angular momentum state $J = |j_1 - j_2|, \dots, j_1 + j_2$ with $M = m_1 + m_2 (\hat{=} -J, \dots, J)$.

Analytic representations of the Clebsch–Gordan coefficients exist, but they are somewhat lengthy, so we will not state it here but they can be found in [18]. Solutions with $m < 0$ are calculated using the relation

$$\langle j_1 m_1, j_2 m_2 | j_1 j_2, J M \rangle = (-1)^{J-j_1-j_2} \langle j_1 - m_1, j_2 - m_2 | j_1 j_2, J M \rangle \quad (\text{A.5})$$

A.2 Wigner–Eckart Theorem

Now we have seen how to construct arbitrary representations of the angular momentum operator by coupling the appropriate angular momentum operators. In general every tensor operator can be build up with the help of the tensor product

$$\mathcal{T}_{i_1, i_2, \dots, i_n} = \bigotimes_{j=1}^{n=\dim \mathcal{V}} \mathcal{T}_{i_j} \quad (\text{A.6})$$

that possess the same transformation rules as vectors under rotation

$$\mathcal{T}'_{i_1, i_2, \dots, i_n} = \sum_{j_1, j_2, \dots, j_n=1}^3 \mathcal{R}_{i_1 j_1}^{(1)} \mathcal{R}_{i_2 j_2}^{(1)} \dots \mathcal{R}_{i_n j_n}^{(1)} \mathcal{T}_{j_1, j_2, \dots, j_n} \quad (\text{A.7})$$

But in order to construct a tensor operator behaving like an angular momentum operator we need to do the same trick as in deriving the Clebsch–Gordan coefficients, that is to use the

transformation behavior of coupled angular momentum. Employing irreducible spherical tensor operators of rank k that can be related to (unnormalized) spherical harmonics in the following way

$$\mathcal{T}_q^{(k)} \propto C_q^{(k)}(\theta, \phi) \quad (\text{A.8})$$

ensures that their behavior under rotations is identical to the corresponding angular momentum of “length” k .

$$\mathcal{T}_{q'}^{(k)} = \sum_{q=-k}^k \mathcal{R}_{q'q}^{(k)} \mathcal{T}_q^{(k)} \quad (\text{A.9})$$

The unnormalized spherical harmonics are defined in terms of the associated Legendre functions $P_m^\ell(x)$ that solves the generalized Legendre’s differential equation. There exists various recursive formulae to calculate the Legendre polynomials to specific orders ℓ , e.g. defined with the help of Rodrigues’ formula

$$P_m^\ell(x) = \frac{(-1)^m}{2^\ell \ell!} (1-x^2)^{m/2} \frac{d^{\ell+m}}{dx^{\ell+m}} (x^2-1)^\ell \quad (\text{A.10})$$

The unnormalized spherical harmonics can be written as

$$C_m^\ell(\theta, \phi) = \sqrt{\frac{(\ell-m)!}{(\ell+m)!}} P_m^\ell(\cos \theta) e^{im\phi} \quad (\text{A.11})$$

and finally normalized by integrating $C_m^\ell(\theta, \phi)$ over the whole three dimensional space which gives rise to

$$Y_m^\ell(\theta, \phi) = \sqrt{\frac{(2\ell+1)}{4\pi}} C_m^\ell(\cos \theta) \quad (\text{A.12})$$

The Wigner–Eckart theorem is useful to calculate the matrix elements of spherical tensor operators in the angular momentum basis $\{|j m\rangle\}$. It states that the matrix elements of spherical tensor operators of rank k are equal to the coupling of k to the angular momentum, while the components q of the spherical tensor operator affect m

$$\langle J' M' | \mathcal{T}_q^{(k)} | J M \rangle = \langle J M, k q | J k, J' M' \rangle \frac{\langle J' || \mathcal{T}^{(k)} || J \rangle}{\sqrt{2J'+1}} \quad (\text{A.13})$$

$\langle J' || \mathcal{T}^{(k)} || J \rangle$ is called the reduced matrix element and (maybe surprisingly) is independent of M . This allows a very quick and simple calculation of the matrix elements. Because of the Clebsch–Gordan coefficients the matrix elements are zero unless q and k satisfies

$$q = M - M' \quad |J - J'| \leq k \leq J + J' \quad (\text{A.14})$$

For irreducible spherical tensor operators there exists a nice formula to calculate the reduced matrix element

$$\langle J' || \mathcal{T}^{(k)} || J \rangle = (-1)^{J'} \sqrt{(2J'+1)(2J+1)} \underbrace{\begin{pmatrix} J' & k & J \\ 0 & 0 & 0 \end{pmatrix}}_{\text{Wigner-3j-Symbol}} \quad (\text{A.15})$$

The Wigner-3j-Symbol can be related to the Clebsch-Gordan coefficients with the help of Racah's formula

$$\begin{pmatrix} j_1 & j_2 & J \\ m_1 & m_2 & -M \end{pmatrix} \equiv \frac{(-1)^{j_1-j_2+M}}{\sqrt{2J+1}} \langle j_1 m_1, j_2 m_2 | j_1 j_2, J M \rangle \quad (\text{A.16})$$

and conversely the Clebsch-Gordan coefficients can be calculated from equation (A.16)

B

Time Independent Perturbation Theory

Suppose one encounters a system which can be split in a known and solved Hamiltonian \mathcal{H}^0 and a small deviation \mathcal{H}' which perturbs this known solution. Then it is easy to find solutions to the complete system using the time independent perturbation theory

$$\mathcal{H} = \mathcal{H}^{(0)} + \mathcal{H}' \quad \mathcal{H}^{(0)} |E_i^{(0)}\rangle = E_i^{(0)} |E_i^{(0)}\rangle \quad (\text{B.1})$$

\mathcal{H} solves the unperturbed system with eigenstates $|E_i^{(0)}\rangle$ and eigenenergies $E_i^{(0)}$. The perturbation is considered to be small, so one can expand the perturbation in a series of unperturbed terms, with a perturbation factor $\beta \ll 1$.

$$E = E^{(0)} + \beta E^{(1)} + \beta^2 E^{(2)} + \dots \quad (\text{B.2})$$

$$|E\rangle = |E^{(0)}\rangle + \beta |E^{(1)}\rangle + \beta^2 |E^{(2)}\rangle + \dots \quad (\text{B.3})$$

The 1st order corrections to the unperturbed solutions are

- $p = 1$:

$$E_i^{(1)} = \langle E_i^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle \quad (\text{B.4})$$

$$|E_i^{(1)}\rangle = \sum_{j \neq i} \frac{\langle E_j^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{E_i^{(0)} - E_j^{(0)}} |E_j^{(0)}\rangle \quad (\text{B.5})$$

The 2nd order corrections to the unperturbed solutions are

- $p = 2$:

$$E_i^{(2)} = \langle E_i^{(0)} | \mathcal{H}' | E_i^{(1)} \rangle$$

$$\begin{aligned}
& \stackrel{\text{(B.5)}}{=} \sum_{j \neq i} \frac{\langle E_i^{(0)} | \mathcal{H}' | E_j^{(0)} \rangle \langle E_j^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{E_i^{(0)} - E_j^{(0)}} \\
& = \sum_{j \neq i} \frac{|\langle E_j^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle|^2}{E_i^{(0)} - E_j^{(0)}} \tag{B.6}
\end{aligned}$$

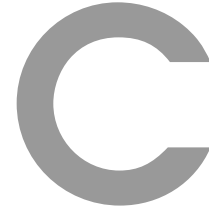
$$\begin{aligned}
|E_i^{(2)}\rangle & = \sum_{j \neq i} \frac{\langle E_j^{(0)} | \mathcal{H}' | E_i^{(1)} \rangle}{E_i^{(0)} - E_j^{(0)}} |E_j^{(0)}\rangle - E_i^{(1)} \sum_{j \neq i} \frac{\langle E_j^{(0)} | E_i^{(1)} \rangle}{E_i^{(0)} - E_j^{(0)}} |E_j^{(0)}\rangle \\
& \stackrel{\text{(B.5)}}{=} \sum_{j \neq i} \sum_{k \neq i} \frac{\langle E_j^{(0)} | \mathcal{H}' | E_k^{(0)} \rangle \langle E_k^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{(E_i^{(0)} - E_j^{(0)}) (E_i^{(0)} - E_k^{(0)})} |E_j^{(0)}\rangle \\
& \quad - E_i^{(1)} \sum_{j \neq i} \sum_{k \neq i} \frac{\langle E_j^{(0)} | E_k^{(0)} \rangle \langle E_k^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{(E_i^{(0)} - E_j^{(0)}) (E_i^{(0)} - E_k^{(0)})} |E_j^{(0)}\rangle \\
& \stackrel{\text{(B.4)}}{=} \sum_{j \neq i} \sum_{k \neq i} \left\{ \frac{\langle E_j^{(0)} | \mathcal{H}' | E_k^{(0)} \rangle \langle E_k^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{(E_i^{(0)} - E_j^{(0)}) (E_i^{(0)} - E_k^{(0)})} \right. \\
& \quad \left. - \langle E_i^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle \frac{\langle E_j^{(0)} | E_k^{(0)} \rangle \langle E_k^{(0)} | \mathcal{H}' | E_i^{(0)} \rangle}{(E_i^{(0)} - E_j^{(0)}) (E_i^{(0)} - E_k^{(0)})} \right\} \tag{B.7}
\end{aligned}$$

In general the p^{th} order corrections to unperturbed solutions

$$E_i^{(p)} = \langle E_i^{(0)} | \mathcal{H}' | E_i^{(p-1)} \rangle \tag{B.8}$$

$$|E_i^{(p)}\rangle = \sum_{j \neq i} \frac{\langle E_j^{(0)} | \mathcal{H}' | E_i^{(p-1)} \rangle}{E_i^{(0)} - E_j^{(0)}} |E_j^{(0)}\rangle - \sum_{q=1}^p E_i^{(q)} \sum_{j \neq i} \frac{\langle E_j^{(0)} | E_i^{(p-q)} \rangle}{E_i^{(0)} - E_j^{(0)}} |E_j^{(0)}\rangle \tag{B.9}$$

can be calculated recursively from order $(p-1)$ to order p . Notice that for non-degenerate eigenstates and orthonormal eigenbasis of $\mathcal{H}^{(0)}$, $\langle E_j^{(0)} | E_i^{(0)} \rangle = 0$ for all $j \neq i$. In this case the sum over q runs only from 1 to $(p-1)$.



Transformation of Vector-Components in Different Base

An orthonormal basis $\{\mathbf{e}_i\}_{i=1,\dots,\dim \mathcal{E}}$ can be expressed via a matrix, with the absolute value of the determinant equal to one, that contains all basis vectors in its columns. If there exists a different basis $\{\mathbf{f}_i\}_{i=1,\dots,\dim \mathcal{F}}$ (for the same vector space $\dim \mathcal{E} \equiv \dim \mathcal{F}$) then the linear mapping between the two bases can be expressed as follows

$$F = \mathcal{T}E \quad (\text{C.1})$$

$$F = \begin{Bmatrix} \mathbf{f}_1 \\ \mathbf{f}_2 \\ \mathbf{f}_3 \end{Bmatrix} \equiv (\mathbf{f}_1|\mathbf{f}_2|\mathbf{f}_3) = \mathcal{T}(\mathbf{e}_1|\mathbf{e}_2|\mathbf{e}_3) = \mathcal{T} \begin{Bmatrix} \mathbf{e}_1 \\ \mathbf{e}_2 \\ \mathbf{e}_3 \end{Bmatrix} = \mathcal{T}E \quad (\text{C.2})$$

where the mapping \mathcal{T} is understood as a simple matrix multiplication on the matrix $(\mathbf{e}_1|\mathbf{e}_2|\mathbf{e}_3)$.

$$\mathbf{f}_j = \sum_j^{\dim \mathcal{E}} T_{ji} \mathbf{e}_j \quad (\text{C.3})$$

Here T_{ij} are the components of the matrix representing the linear mapping between the two bases. For equal dimensionality the matrix T is quadratic and there exists an inverse T^{-1} which maps the basis $\{\mathbf{f}_i\}_{i=1,\dots,\dim \mathcal{F}}$ on the basis $\{\mathbf{e}_i\}_{i=1,\dots,\dim \mathcal{E}}$.

$$\mathbf{e}_j = \sum_j^{\dim \mathcal{F}} (T^{-1})_{ji} \mathbf{f}_j \quad (\text{C.4})$$

Using (C.1), the matrix T and its inverse T^{-1} can be express via the matrices E and F

$$T = FE^{-1} = FE^\dagger \quad T^{-1} = EF^{-1} = EF^\dagger \quad (\text{C.5})$$

since the bases are orthonormal one has unitary matrices E and F and hence T will also be unitary $T^{-1} = T^\dagger$, where $T^\dagger \equiv (T^*)^t = (T^t)^*$ denotes the hermitian conjugate.

Vectors are invariant under basis transformations but their components with respect to the basis vectors (projections onto the basis vectors) are transformed as well.

$$\begin{aligned} \mathbf{v} &= E \mathbf{v}_E \stackrel{!}{=} F \mathbf{v}_F \\ \stackrel{\text{(C.1)}}{\hookrightarrow} E \mathbf{v}_E &= T E \mathbf{v}_F \end{aligned} \quad (\text{C.6})$$

If the components with respect to the basis $\{\mathbf{e}_i\}_{i=1, \dots, \dim \mathcal{E}}$ are known, the components with respect to the basis $\{\mathbf{f}_i\}_{i=1, \dots, \dim \mathcal{F}}$ are then

$$\mathbf{v}_E = E^{-1} T E \mathbf{v}_F \quad (\text{C.7})$$

Furthermore, if one chooses canonical (cartesian) coordinates with basis vectors

$$\mathbf{e}_1 = \begin{pmatrix} 1 \\ 0 \\ 0 \\ \vdots \end{pmatrix} \quad \mathbf{e}_2 = \begin{pmatrix} 0 \\ 1 \\ 0 \\ \vdots \end{pmatrix} \quad \dots \quad \mathbf{e}_j = \begin{pmatrix} \vdots \\ j \\ \vdots \end{pmatrix} \quad \dots \quad \mathbf{e}_{\dim \mathcal{E}} = \begin{pmatrix} \vdots \\ 0 \\ 0 \\ 1 \end{pmatrix} \quad (\text{C.8})$$

such that $E = \mathbb{1}$. The components of the arbitrary vector \mathbf{v} are transformed with the inverse transformation $T^{-1} = T^\dagger$. A change of basis form $\{\mathbf{e}_i\}_{i=1, \dots, \dim \mathcal{E}}$ to $\{\mathbf{f}_i\}_{i=1, \dots, \dim \mathcal{F}}$ via T transforms the components of a vector with T^{-1} .

$$F = T E = T \quad \mathbf{v}_F = T^{-1} \mathbf{v}_E \quad (\text{C.9})$$

The transformation rules for linear mappings between vectors of the vector space can be deduced from these transformation rules. Let \mathcal{A} be a mapping \mathbf{v} on \mathbf{w} with the corresponding matrix A

$$\mathcal{A} : \mathbf{v}_E \longrightarrow \mathbf{w}_E \quad (\text{C.10})$$

then

$$\begin{aligned} A_E \mathbf{v}_E &= \mathbf{w}_E \\ A_E T \mathbf{v}_F &= T \mathbf{w}_F \\ \underbrace{T^{-1} A_E T}_{A_F} \mathbf{v}_F &= \mathbf{w}_F \\ \hookrightarrow A_F &= T^{-1} A_E T \end{aligned} \quad (\text{C.11})$$

Usually we adopt a more sloppy notation where we suppress the subscripts referring to the basis. Pictorially one can depict this fact in a commutative diagram*.

$$\begin{array}{ccc}
 \mathbf{v} & \xrightarrow{\mathcal{A}} & \mathbf{w} \\
 \mathcal{T}^{-1} \downarrow & & \uparrow \mathcal{T} \\
 \tilde{\mathbf{v}} & \xrightarrow{\tilde{\mathcal{A}}} & \tilde{\mathbf{w}} \\
 \mathcal{A} & & \tilde{\mathcal{A}}
 \end{array}
 \qquad
 \begin{array}{ccc}
 \mathbf{v} & \xrightarrow{\mathcal{A}} & \mathbf{w} \\
 \mathcal{T}^{-1} \downarrow & & \downarrow \mathcal{T}^{-1} \\
 \tilde{\mathbf{v}} & \xrightarrow{\tilde{\mathcal{A}}} & \tilde{\mathbf{w}} \\
 \mathcal{A} & & \tilde{\mathcal{A}}
 \end{array}$$

$$\mathcal{A} = \mathcal{T}^{-1} \tilde{\mathcal{A}} \mathcal{T} \qquad \mathcal{T}^{-1} \mathcal{A} = \tilde{\mathcal{A}} \mathcal{T}^{-1}$$

Example: Spherical Basis for Vector Operators

Suppose we want to transform vectors (or vector operators) from cartesian coordinates to spherical coordinates, which are convenient for spherical tensor operators that operates on the angular momentum eigenbasis. The spherical coordinates are defined as

$$\mathbf{e}_{-1} = \frac{1}{\sqrt{2}}(\mathbf{e}_x - i\mathbf{e}_y) \qquad \mathbf{e}_0 = \mathbf{e}_z \qquad \mathbf{e}_1 = -\frac{1}{\sqrt{2}}(\mathbf{e}_x + i\mathbf{e}_y) \qquad (\text{C.12})$$

and conversely

$$\mathbf{e}_x = \frac{1}{\sqrt{2}}(\mathbf{e}_{-1} - \mathbf{e}_1) \qquad \mathbf{e}_z = \mathbf{e}_0 \qquad \mathbf{e}_y = \frac{i}{\sqrt{2}}(\mathbf{e}_1 + \mathbf{e}_{-1}) \qquad (\text{C.13})$$

This can be written as a matrix multiplication in the sense of equation (C.3).

$$\begin{pmatrix} \mathbf{e}_{-1} \\ \mathbf{e}_0 \\ \mathbf{e}_1 \end{pmatrix} = \frac{1}{\sqrt{2}} \underbrace{\begin{pmatrix} 1 & 0 & -1 \\ -i & 0 & -i \\ 0 & \sqrt{2} & 0 \end{pmatrix}}_T \begin{pmatrix} \mathbf{e}_x \\ \mathbf{e}_y \\ \mathbf{e}_z \end{pmatrix} \qquad (\text{C.14})$$

and conversely

$$\begin{pmatrix} \mathbf{e}_x \\ \mathbf{e}_y \\ \mathbf{e}_z \end{pmatrix} = \frac{1}{\sqrt{2}} \underbrace{\begin{pmatrix} 1 & i & 0 \\ 0 & 0 & \sqrt{2} \\ -1 & i & 0 \end{pmatrix}}_{T^{-1} = T^\dagger} \begin{pmatrix} \mathbf{e}_{-1} \\ \mathbf{e}_0 \\ \mathbf{e}_1 \end{pmatrix} \qquad (\text{C.15})$$

Now if we transform let's say, the electric dipole operator \mathbf{d} , then the components reads in spherical coordinates

$$\begin{pmatrix} d_{-1} \\ d_0 \\ d_1 \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i & 0 \\ 0 & 0 & \sqrt{2} \\ -1 & i & 0 \end{pmatrix} \begin{pmatrix} d_x \\ d_y \\ d_z \end{pmatrix} \qquad (\text{C.16})$$

* It is called commutative diagram because the mapping \mathcal{A} commutes with the transformation \mathcal{T}

or equivalently

$$d_{-1} = \frac{1}{\sqrt{2}}(d_x + id_y) \quad d_0 = d_z \quad d_1 = -\frac{1}{\sqrt{2}}(d_x - id_y) \quad (\text{C.17})$$

and

$$d_x = -\frac{1}{\sqrt{2}}(d_1 - d_{-1}) \quad d_z = d_0 d_y = -\frac{i}{\sqrt{2}}(d_1 + d_{-1}) \quad (\text{C.18})$$



Operators in Product Spaces

Operators acting in two different vector spaces \mathcal{V}^1 and \mathcal{V}^2 can act on a combined vector space $\mathcal{V} = \mathcal{V}^1 \otimes \mathcal{V}^2$ that is build up from the two vector spaces via the so called tensor product

$$\mathcal{A} = \mathcal{A} \otimes \mathbb{1}_{\mathcal{V}^2} \quad (\text{D.1})$$

$$\mathcal{B} = \mathbb{1}_{\mathcal{V}^1} \otimes \mathcal{B} \quad (\text{D.2})$$

Since \mathcal{A} is acting only on \mathcal{V}^1 , there must be an identity operator acting on \mathcal{V}^2 . In the same way the two bases of the vector spaces are build up as tensor products of the single vector space basis.

$$|v^1\rangle \otimes |v^2\rangle = |v^1 v^2\rangle \quad (\text{D.3})$$

$$\langle v^1| \otimes \langle v^2| = \langle v^1 v^2| \quad (\text{D.4})$$

This leads to the action of \mathcal{A} and \mathcal{B} on the product space vectors

$$\begin{aligned} \langle v^1 v^2| \mathcal{A} |w^1 w^2\rangle &= (\langle v^1| \otimes \langle v^2|) (\mathcal{A} \otimes \mathbb{1}_{\mathcal{V}^2}) (|w^1\rangle \otimes |w^2\rangle) \\ &= \langle v^1| \mathcal{A} |w^1\rangle \langle v^2| \mathbb{1}_{\mathcal{V}^2} |w^2\rangle \\ &= \langle v^1| \mathcal{A} |w^1\rangle \langle v^2| w^2\rangle \end{aligned} \quad (\text{D.5})$$

and in the same way it can be defined for operator \mathcal{B}

$$\begin{aligned} \langle v^1 v^2| \mathcal{B} |w^1 w^2\rangle &= (\langle v^1| \otimes \langle v^2|) (\mathbb{1}_{\mathcal{V}^1} \otimes \mathcal{B}) (|w^1\rangle \otimes |w^2\rangle) \\ &= \langle v^1| \mathbb{1}_{\mathcal{V}^1} |w^1\rangle \langle v^2| \mathcal{B} |w^2\rangle \\ &= \langle v^1| w^1\rangle \langle v^2| \mathcal{B} |w^2\rangle \end{aligned} \quad (\text{D.6})$$

The composition of two operators can then be written as

$$\mathcal{A}^1 \mathcal{B}^2 = (\mathcal{A} \otimes \mathbf{1})(\mathbf{1} \otimes \mathcal{B}) = \mathcal{A} \otimes \mathcal{B} \quad (\text{D.7})$$

where the superscript denotes the action of the operator with respect to its vector space on which the action of the operator is defined. If there is a tensor product sign present, then one can deduce from the ordering of the factors on which space the operator is acting. The last equality in (D.7) holds because

$$\begin{aligned} \langle v^1 v^2 | \mathcal{A}^1 \mathcal{B}^2 | w^1 w^2 \rangle &\stackrel{\dagger}{=} \langle v^1 | \mathcal{A} | w^1 \rangle \langle v^2 | \mathcal{B} | w^2 \rangle \\ &\stackrel{(\text{D.7})}{=} (\langle v^1 | \otimes \langle v^2 |) (\mathcal{A} \otimes \mathbf{1})(\mathbf{1} \otimes \mathcal{B}) (|w^1 \rangle \otimes |w^2 \rangle) \\ &= (\langle v^1 | \otimes \langle v^2 |) (\mathcal{A} \otimes \mathcal{B}) (|w^1 \rangle \otimes |w^2 \rangle) \end{aligned} \quad (\text{D.8})$$

Following equation (D.7) one can easily define the action of a scalar product of a vector operator

$$\mathbf{d}^1 \cdot \mathbf{e}^2 = \sum_i d_i^1 e_i^2 = \sum_i (d_i^1 \otimes \mathbf{1})(\mathbf{1} \otimes e_i) \stackrel{(\text{D.7})}{=} \sum_i (d_i^1 \otimes e_i) \quad (\text{D.9})$$

For a three-dimensional vector operator in cartesian coordinates the scalar product (D.9) can be written in the following form

$$\mathbf{d}^1 \cdot \mathbf{e}^2 = (\mathbf{d} \otimes \mathbf{1})(\mathbf{1} \otimes \mathbf{e}) = d_x \otimes e_x + d_y \otimes e_y + d_z \otimes e_z \quad (\text{D.10})$$

This is easily generalized to scalar products of two vector operators

$$\mathbf{d}^1 \cdot \mathbf{d}^2 = (\mathbf{d} \otimes \mathbf{1}) \cdot (\mathbf{1} \otimes \mathbf{d}) = \sum_i d_i^1 d_i^2 = \sum_i (d_i^1 \otimes \mathbf{1})(\mathbf{1} \otimes d_i) \stackrel{(\text{D.7})}{=} \sum_i (d_i^1 \otimes d_i) \quad (\text{D.11})$$

Representations of operators in product spaces are determined by the coordinate representation of the tensor product of operators. Assume operator \mathcal{A} acts on \mathcal{V} and operator \mathcal{B} acts on \mathcal{W} , respectively. The basis vectors of \mathcal{V} is denoted by $\{\mathbf{v}_i\}_{i=1, \dots, \dim \mathcal{V}}$ and the basis of \mathcal{W} is denoted by $\{\mathbf{w}_j\}_{j=1, \dots, \dim \mathcal{W}}$ then the basis of the product space $\mathcal{V} \otimes \mathcal{W}$ is denoted by

$$\mathbf{v}_i \otimes \mathbf{w}_j = (v_{ik} w_{jl})_{k=1, \dots, \dim \mathcal{V}; l=1, \dots, \dim \mathcal{W}} \quad (\text{D.12})$$

Then the linear map from one vector $\mathbf{x} \in \mathcal{V} \otimes \mathcal{W}$ to another vector $\mathbf{y} \in \mathcal{V} \otimes \mathcal{W}$ can be written in coordinates as

$$\begin{aligned} \mathbf{y} &= (\mathcal{A} \otimes \mathcal{B})\mathbf{x} = \sum_{i j} (\mathcal{A} \otimes \mathcal{B})x_{ij}(\mathbf{v}_i \otimes \mathbf{w}_j) \\ &= \sum_{i j} (\mathcal{A}\mathbf{v}_i) \otimes (\mathcal{B}\mathbf{w}_j)x_{ij} \\ &= \sum_{i j r s} (\mathbf{v}_r A_{ri} \otimes \mathbf{w}_s B_{sj})x_{ij} \\ &= \sum_{i j r s} (A_{ri} B_{sj} x_{ij}) \mathbf{v}_r \otimes \mathbf{w}_s \end{aligned}$$

$$\hookrightarrow y_{rs} = A_{ri}B_{sj}x_{ij} = C_{risj}x_{ij} \quad \text{with} \quad C = \mathcal{A} \otimes \mathcal{B} \quad (\text{D.13})$$

If one chooses canonical basis vectors (i.e. in cartesian coordinates)

$$\mathbf{e}_1 \otimes \mathbf{e}_2 = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \quad \mathbf{e}_1 \otimes \mathbf{e}_1 = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \quad \mathbf{e}_2 \otimes \mathbf{e}_1 = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} \quad \mathbf{e}_2 \otimes \mathbf{e}_2 = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix} \quad (\text{D.14})$$

this leads to the following “multiplication” rule for the tensor product of two operators in this canonical basis

$$\begin{pmatrix} y_{11} \\ y_{12} \\ y_{21} \\ y_{22} \end{pmatrix} = \begin{pmatrix} A_{11}B_{11} & A_{11}B_{12} & A_{12}B_{11} & A_{12}B_{12} \\ A_{11}B_{21} & A_{11}B_{22} & A_{12}B_{21} & A_{12}B_{22} \\ A_{21}B_{11} & A_{21}B_{12} & A_{22}B_{11} & A_{22}B_{12} \\ A_{21}B_{21} & A_{21}B_{22} & A_{22}B_{21} & A_{22}B_{22} \end{pmatrix} \begin{pmatrix} x_{11} \\ x_{12} \\ x_{21} \\ x_{22} \end{pmatrix}$$

$$\hookrightarrow \mathbf{y} = \underbrace{\begin{pmatrix} A_{11}B & A_{12}B \\ A_{21}B & A_{22}B \end{pmatrix}}_{A \otimes B} \mathbf{x} \quad (\text{D.15})$$

Example: Two–Level Spin System

The usage of the symbol $|\uparrow\rangle$ indicates the ground state of the two–level system and the symbol $|\downarrow\rangle$ the excited state, respectively. Then the product space basis is made up of the following basis states

$$\begin{aligned} |\uparrow\uparrow\rangle &= |\uparrow\rangle \otimes |\uparrow\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} & |\uparrow\downarrow\rangle &= |\uparrow\rangle \otimes |\downarrow\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \\ |\downarrow\uparrow\rangle &= |\downarrow\rangle \otimes |\uparrow\rangle = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} & |\downarrow\downarrow\rangle &= |\downarrow\rangle \otimes |\downarrow\rangle = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix} \end{aligned} \quad (\text{D.16})$$

The components of a state in this two-level system basis are found in an expansion of the state in terms of the basis states

$$|\Psi\rangle = \begin{pmatrix} \Psi_{\uparrow\uparrow} \\ \Psi_{\uparrow\downarrow} \\ \Psi_{\downarrow\uparrow} \\ \Psi_{\downarrow\downarrow} \end{pmatrix} = |\uparrow\uparrow\rangle \langle\uparrow\uparrow|\Psi\rangle + |\uparrow\downarrow\rangle \langle\uparrow\downarrow|\Psi\rangle + |\downarrow\uparrow\rangle \langle\downarrow\uparrow|\Psi\rangle + |\downarrow\downarrow\rangle \langle\downarrow\downarrow|\Psi\rangle \quad (\text{D.17})$$

It is customary to define “spin operators” build up from Pauli matrices

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (\text{D.18})$$

$$\sigma_+ = \frac{1}{2}(\sigma_x + i\sigma_y) = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \sigma_- = \frac{1}{2}(\sigma_x - i\sigma_y) = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad (\text{D.19})$$

In the two-level system product space, the raising operator σ_+ , the lowering operator σ_- and the σ_z -operator are constructed via the tensor product as shown before

$$\sigma_+^1 = \sigma_+ \otimes \mathbf{1} = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} \quad \sigma_+^2 = \mathbf{1} \otimes \sigma_+ = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.20})$$

$$\sigma_-^1 = \sigma_- \otimes \mathbf{1} = \begin{pmatrix} 0 & 0 \\ 1 & 0 \\ 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \sigma_-^2 = \mathbf{1} \otimes \sigma_- = \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix} \quad (\text{D.21})$$

The projectors on the two basis states of the two-level system are build via the σ_z -operator

$$\mathcal{P}_\uparrow = \frac{\mathbf{1} + \sigma_z}{2} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad \mathcal{P}_\downarrow = \frac{\mathbf{1} - \sigma_z}{2} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \quad (\text{D.22})$$

$$\mathcal{P}_\uparrow^1 = \frac{\mathbf{1} + \sigma_z}{2} \otimes \mathbf{1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \mathcal{P}_\uparrow^2 = \mathbf{1} \otimes \frac{\mathbf{1} + \sigma_z}{2} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.23})$$

$$\mathcal{P}_{\downarrow}^1 = \frac{\mathbb{1} - \sigma_z}{2} \otimes \mathbb{1} = \begin{pmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{0} & 1 & 0 \\ & 0 & 1 \end{pmatrix} \quad \mathcal{P}_{\downarrow}^2 = \mathbb{1} \otimes \frac{\mathbb{1} - \sigma_z}{2} = \begin{pmatrix} 0 & 0 & \mathbf{0} \\ 0 & 1 & \\ \mathbf{0} & 0 & 0 \\ & 0 & 1 \end{pmatrix} \quad (\text{D.24})$$

An arbitrary operator in this two-level basis can then be written using the spin operators and the projectors stated in (D.20)–(D.24)

$$\mathcal{A} = \mathbb{1}\mathcal{A}\mathbb{1} = \sum_{ij} |i\rangle \langle i| \mathcal{A} |j\rangle \langle j| = \sum_{ij} A_{ij} |i\rangle \langle j| \quad (\text{D.25})$$

$$\begin{aligned} \hookrightarrow A_{11} |\uparrow\uparrow\rangle \langle \uparrow\uparrow| &= A_{11} \mathcal{P}_{\uparrow}^1 \mathcal{P}_{\uparrow}^2 = A_{11} \left(\frac{\mathbb{1} + \sigma_z}{2} \otimes \mathbb{1} \right) \left(\mathbb{1} \otimes \frac{\mathbb{1} + \sigma_z}{2} \right) \\ &= A_{11} \left(\frac{\mathbb{1} + \sigma_z}{2} \right) \otimes \left(\frac{\mathbb{1} + \sigma_z}{2} \right) = A_{11} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \\ &= A_{11} \begin{pmatrix} 1 & 0 & & \\ 0 & 0 & & \\ & & \mathbf{0} & \\ & & & \mathbf{0} \end{pmatrix} \end{aligned} \quad (\text{D.26})$$

$$A_{12} |\uparrow\uparrow\rangle \langle \uparrow\downarrow| = A_{12} \mathcal{P}_{\uparrow}^1 \sigma_+^2 = A_{12} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} = A_{12} \begin{pmatrix} 0 & 1 & & \\ 0 & 0 & & \\ & & \mathbf{0} & \\ & & & \mathbf{0} \end{pmatrix} \quad (\text{D.27})$$

$$A_{13} |\uparrow\uparrow\rangle \langle \downarrow\uparrow| = A_{13} \sigma_+^1 \mathcal{P}_{\uparrow}^2 = A_{13} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} = A_{13} \begin{pmatrix} & 1 & 0 & \\ \mathbf{0} & & 0 & 0 \\ & & \mathbf{0} & \\ & & & \mathbf{0} \end{pmatrix} \quad (\text{D.28})$$

$$A_{14} |\uparrow\uparrow\rangle \langle \downarrow\downarrow| = A_{14} \sigma_+^1 \sigma_+^2 = A_{14} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} = A_{14} \begin{pmatrix} & 0 & 1 & \\ \mathbf{0} & & 0 & 0 \\ & & \mathbf{0} & \\ & & & \mathbf{0} \end{pmatrix} \quad (\text{D.29})$$

$$A_{21} |\uparrow\downarrow\rangle \langle \uparrow\uparrow| = A_{21} \mathcal{P}_{\uparrow}^1 \sigma_-^2 = A_{21} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} = A_{21} \begin{pmatrix} 0 & 0 & & \\ 1 & 0 & & \\ & & \mathbf{0} & \\ & & & \mathbf{0} \end{pmatrix} \quad (\text{D.30})$$

$$A_{22} |\uparrow\downarrow\rangle \langle\uparrow\downarrow| = A_{22} \mathcal{P}_\uparrow^1 \mathcal{P}_\downarrow^2 = A_{22} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} = A_{22} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.31})$$

$$A_{23} |\uparrow\downarrow\rangle \langle\downarrow\uparrow| = A_{23} \sigma_+^1 \sigma_-^2 = A_{23} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} = A_{23} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.32})$$

$$A_{24} |\uparrow\downarrow\rangle \langle\downarrow\downarrow| = A_{24} \sigma_+^1 \mathcal{P}_\uparrow^2 = A_{24} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} = A_{24} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.33})$$

$$A_{31} |\downarrow\uparrow\rangle \langle\uparrow\uparrow| = A_{31} \sigma_-^1 \mathcal{P}_\uparrow^2 = A_{31} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} = A_{31} \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.34})$$

$$A_{32} |\downarrow\uparrow\rangle \langle\uparrow\downarrow| = A_{32} \sigma_-^1 \sigma_+^2 = A_{32} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} = A_{32} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.35})$$

$$A_{33} |\downarrow\uparrow\rangle \langle\downarrow\uparrow| = A_{33} \mathcal{P}_\downarrow^1 \mathcal{P}_\uparrow^2 = A_{33} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} = A_{33} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.36})$$

$$A_{34} |\downarrow\uparrow\rangle \langle\downarrow\downarrow| = A_{34} \mathcal{P}_\downarrow^1 \sigma_+^2 = A_{34} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} = A_{34} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} \quad (\text{D.37})$$

$$A_{41} |\downarrow\downarrow\rangle \langle\uparrow\uparrow| = A_{41} \sigma_-^1 \sigma_-^2 = A_{41} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} = A_{41} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \quad (\text{D.38})$$

$$A_{42} |\downarrow\downarrow\rangle \langle\uparrow\downarrow| = A_{42} \sigma_-^1 \mathcal{P}_\downarrow^2 = A_{42} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} = A_{42} \begin{pmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{pmatrix} \quad (\text{D.39})$$

$$A_{43} |\downarrow\downarrow\rangle \langle\downarrow\uparrow| = A_{43} \mathcal{P}_\downarrow^1 \sigma_-^2 = A_{43} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} = A_{43} \begin{pmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{pmatrix} \quad (\text{D.40})$$

$$A_{44} |\downarrow\downarrow\rangle \langle\downarrow\downarrow| = A_{44} \mathcal{P}_\downarrow^1 \mathcal{P}_\downarrow^2 = A_{44} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} = A_{44} \begin{pmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 1 \end{pmatrix} \quad (\text{D.41})$$

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E

General Solution of a Two–Level System

Two level systems can easily be mapped onto a spin system with two degrees of freedom, namely spin–up $|\uparrow\rangle$ and spin–down $|\downarrow\rangle$. If we consider a hermitian operator $\mathcal{H} = \mathcal{H}^\dagger$ there exists a unitary transformation that diagonalizes the representation of \mathcal{H} . It is useful to separate the diagonal elements from the non–diagonal element and write

$$\mathcal{H} = \mathcal{H} + \mathcal{H}' = \begin{pmatrix} H_{11} & 0 \\ 0 & H_{22} \end{pmatrix} + \begin{pmatrix} 0 & H'_{12} \\ (H'_{12})^* & 0 \end{pmatrix} \quad (\text{E.1})$$

Calculating the eigenvalues with the help of the eigenvalue equation

$$\begin{aligned} \mathcal{H}\mathbf{v} &= E\mathbf{v} \\ (\mathcal{H} - E\mathbf{1})\mathbf{v} &= 0 \quad \forall \mathbf{v} \in \mathbb{C}^2 : \mathbf{v} \neq \mathbf{0} \\ \Leftrightarrow \det(\mathcal{H} - E\mathbf{1}) &= 0 \end{aligned} \quad (\text{E.2})$$

leads to

$$\det(\mathcal{H} - E\mathbf{1}) = E^2 - \underbrace{(H_{11} + H_{22})}_{\text{tr } \mathcal{H}} E + \underbrace{(H_{11}H_{22} - H'_{12}(H'_{12})^*)}_{\det \mathcal{H}} = 0 \quad (\text{E.3})$$

Solving equation (E.3) for E yields

$$E_{\pm} = \frac{H_{11} + H_{22}}{2} \pm \sqrt{\left(\frac{H_{11} - H_{22}}{2}\right)^2 + |H'_{12}|^2} \quad (\text{E.4})$$

In order to find the eigenvectors and the unitary transformation we need to split \mathcal{H} in a different way

$$\begin{aligned}\mathcal{H} &= \begin{pmatrix} \frac{1}{2}(H_{11} + H_{22}) & 0 \\ 0 & \frac{1}{2}(H_{11} + H_{22}) \end{pmatrix} + \begin{pmatrix} \frac{1}{2}(H_{11} - H_{22}) & H'_{12} \\ (H'_{12})^* & -\frac{1}{2}(H_{11} - H_{22}) \end{pmatrix} \\ &= \frac{1}{2}(\text{tr } \mathcal{H})\mathbf{1} + \frac{1}{2}(H_{11} - H_{22})\mathcal{H} \end{aligned} \quad (\text{E.5})$$

whereas $\mathcal{H} = \mathcal{H}^\dagger$ is defined as

$$\mathcal{H} = \begin{pmatrix} 1 & \frac{2H'_{12}}{H_{11} - H_{22}} \\ \frac{2(H'_{12})^*}{H_{11} - H_{22}} & -1 \end{pmatrix} \quad (\text{E.6})$$

The matrix \mathcal{H} commutes with the matrix H

$$\begin{aligned}[H, \mathcal{H}] &= H\mathcal{H} - \mathcal{H}H \\ &= \frac{1}{2}(\text{tr } \mathcal{H})\mathbf{1}\mathcal{H} + \frac{1}{2}(H_{11} - H_{22})\mathcal{H}^2 - \frac{1}{2}(\text{tr } \mathcal{H})\mathcal{H}\mathbf{1} - \frac{1}{2}(H_{11} - H_{22})\mathcal{H}^2 \\ &= 0 \end{aligned} \quad (\text{E.7})$$

and therefore there exists a common eigenbasis denoted by $\{|+\rangle, |-\rangle\}$. Now we have two eigenvalue equations

$$\mathcal{H}|\pm\rangle = E_\pm|\pm\rangle \quad \mathcal{H}|\pm\rangle = \epsilon_\pm|\pm\rangle \quad (\text{E.8})$$

$$\Leftrightarrow \det(\mathcal{H} - E\mathbf{1}) = 0 \quad \det(\mathcal{H} - \epsilon\mathbf{1}) = 0 \quad (\text{E.9})$$

with the two eigenvalues satisfying the following relation

$$\begin{aligned}\mathcal{H}|\pm\rangle &= \frac{1}{2}(\text{tr } \mathcal{H})|\pm\rangle + \frac{1}{2}(H_{11} - H_{22})\epsilon_\pm|\pm\rangle = E_\pm|\pm\rangle \\ \Leftrightarrow E_\pm &= \frac{1}{2}(H_{11} + H_{22}) + \frac{1}{2}(H_{11} - H_{22})\epsilon_\pm \end{aligned} \quad (\text{E.10})$$

We can determine the eigenvalues ϵ_\pm of \mathcal{H} in a straightforward calculation

$$\begin{aligned}\det(\mathcal{H} - \epsilon\mathbf{1}) &= (-1 - \epsilon)(1 - \epsilon) - \frac{4|H'_{12}|^2}{(H_{11} - H_{22})^2} \\ &= -1 + \epsilon - \epsilon + \epsilon^2 - \frac{4|H'_{12}|^2}{(H_{11} - H_{22})^2} \\ &= 0 \\ \Leftrightarrow \epsilon_\pm &= \pm \frac{1}{2} \sqrt{4 \left(1 + \frac{4|H'_{12}|^2}{(H_{11} - H_{22})^2} \right)} = \pm \sqrt{1 + \left(\frac{2|H'_{12}|^2}{(H_{11} - H_{22})^2} \right)} \end{aligned} \quad (\text{E.11})$$

and see that these solutions for ϵ_{\pm} satisfies equation (E.10) together with equation (E.4)

$$\begin{aligned}
E_{\pm} &= \frac{1}{2}(H_{11} + H_{22}) + \frac{1}{2}(H_{11} - H_{22})\epsilon_{\pm} \\
&= \frac{(H_{11} + H_{22})}{2} \pm \frac{(H_{11} - H_{22})}{2} \sqrt{1 + \left(\frac{2}{H_{11} - H_{22}}\right)^2 |H'_{12}|^2} \\
&= \frac{H_{11} + H_{22}}{2} \pm \sqrt{\left(\frac{H_{11} - H_{22}}{2}\right)^2 + |H'_{12}|^2}
\end{aligned} \tag{E.12}$$

After rewriting $H'_{12} = |H'_{12}| e^{-i\gamma}$ it is possible to define a “mixing” angle φ , such that

$$\tan \varphi = \frac{2 |H'_{12}|}{H_{11} - H_{22}} \tag{E.13}$$

since a unitary transformation can be considered as a complex rotation. Now equation (E.6) can be written as

$$\mathcal{H} = \begin{pmatrix} 1 & \tan \varphi e^{-i\gamma} \\ \tan \varphi e^{i\gamma} & -1 \end{pmatrix} \tag{E.14}$$

and the eigenvalue equation (E.9) reads

$$\begin{aligned}
-(1 + \epsilon)(1 - \epsilon) - \tan^2 \varphi &= 0 \\
\hookrightarrow \epsilon_{\pm} &= \pm \sqrt{1 + \tan^2 \varphi} = \pm \sec \varphi
\end{aligned} \tag{E.15}$$

which can be easily verified with $\sec(\arctan x) = \sqrt{1 + x^2}$ and compared to equation (E.12). Now we are able to calculate the eigenvectors that diagonalize \mathcal{H} . Expanding the eigenbasis in $\{|+\rangle, |-\rangle\}$ with respect to the two level system basis $\{|\uparrow\rangle, |\downarrow\rangle\}$ one obtains the components of the eigenvectors

$$|+\rangle = |\uparrow\rangle \underbrace{\langle\uparrow|+ \rangle}_{v_1} + |\downarrow\rangle \underbrace{\langle\downarrow|+ \rangle}_{v_2} = v_1 |\uparrow\rangle + v_2 |\downarrow\rangle \tag{E.16}$$

$$|-\rangle = |\uparrow\rangle \underbrace{\langle\uparrow|-\rangle}_{w_1} + |\downarrow\rangle \underbrace{\langle\downarrow|-\rangle}_{w_2} = w_1 |\uparrow\rangle + w_2 |\downarrow\rangle \tag{E.17}$$

Then equation (E.8) for the eigenvalue $\epsilon_+ = \sec \varphi$ reads

$$\begin{pmatrix} 1 & \tan \varphi e^{-i\gamma} \\ \tan \varphi e^{i\gamma} & -1 \end{pmatrix} \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} = \sec \varphi \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} \tag{E.18}$$

Since $\det \mathcal{H} = 0$ we can express $v_1(v_2)$ by solving the first equation of the linear equation system

$$\begin{aligned}
v_1 + \tan \varphi e^{-i\gamma} v_2 &= \sec \varphi v_1 \\
(1 - \sec \varphi)v_1 + \tan \varphi e^{-i\gamma} v_2 &= 0 \quad | \cdot \cos \varphi
\end{aligned}$$

$$\begin{aligned}
(\cos \varphi - 1)v_1 + \sin \varphi e^{-i\gamma} v_2 &= 0 \\
\hookrightarrow v_1 &= \frac{\sin \varphi}{1 - \cos \varphi} e^{-i\gamma} v_2
\end{aligned} \tag{E.19}$$

Using trigonometric identities $1 - \cos \varphi = 2 \sin^2 \varphi/2$ and $\sin \varphi = 2 \sin \varphi/2 \cos \varphi/2$ we obtain

$$v_1 = \frac{2 \sin \frac{\varphi}{2} \cos \frac{\varphi}{2}}{2 \sin^2 \frac{\varphi}{2}} e^{-i\gamma} v_2 = \cot \frac{\varphi}{2} e^{-i\gamma} v_2 \tag{E.20}$$

Symmetrizing this expression by multiplying with $\sin \varphi/2 e^{i\gamma/2}$ yields

$$\sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} v_1 = \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} v_2 \tag{E.21}$$

The representation matrix of the unitary transformation must consist of normalized column vectors, so we have to impose the normalization condition

$$\mathbf{v}^2 = 1 \quad \hookrightarrow \mathbf{v}^* \cdot \mathbf{v} = 1 \quad \hookrightarrow v_1^* v_1 + v_2^* v_2 = |v_1|^2 + |v_2|^2 = 1 \tag{E.22}$$

that leads to a reasonable solution

$$v_1 = \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \quad \text{and} \quad v_2 = \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \tag{E.23}$$

which fulfills the normalization condition (E.22) and the eigenvalue equation (E.18)

$$v_1 = \cot \frac{\varphi}{2} e^{-i\gamma} v_2 = \cot \frac{\varphi}{2} e^{-i\gamma} \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} = \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \tag{E.24}$$

$$|v_1|^2 + |v_2|^2 = \left| \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \right|^2 + \left| \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \right|^2 = \cos^2 \left(\frac{\varphi}{2} \right) + \sin^2 \left(\frac{\varphi}{2} \right) = 1 \tag{E.25}$$

The eigenvector to the eigenvalue $\epsilon_+ = \sec \varphi$ is

$$|+\rangle = \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |\uparrow\rangle + \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |\downarrow\rangle \quad \varphi = \arctan \left(\frac{2|H'_{12}|}{H_{11} - H_{22}} \right) \tag{E.26}$$

where φ is defined by inverting equation (E.13).

Now calculating the second eigenvector to the eigenvalue $\epsilon_- = -\sec \varphi$ is carried out analogously

$$\begin{pmatrix} 1 & \tan \varphi e^{-i\gamma} \\ \tan \varphi e^{i\gamma} & -1 \end{pmatrix} \begin{pmatrix} w_1 \\ w_2 \end{pmatrix} = -\sec \varphi \begin{pmatrix} w_1 \\ w_2 \end{pmatrix} \tag{E.27}$$

Taking the first equation of (E.27)

$$w_1 + \tan \varphi e^{-i\gamma} w_2 = -\sec \varphi w_1$$

$$(1 + \sec \varphi)w_1 + \tan \varphi e^{-i\gamma} w_2 = 0 \quad | \cdot \cos \varphi$$

$$(\cos \varphi + 1)w_1 + \sin \varphi e^{-i\gamma} w_2 = 0$$

$$\hookrightarrow w_1 = -\frac{\sin \varphi}{1 + \cos \varphi} e^{-i\gamma} w_2 \tag{E.28}$$

Using trigonometric identities $1 + \cos \varphi = 2 \cos^2 \varphi/2$ and $\sin \varphi = 2 \sin \varphi/2 \cos \varphi/2$ we obtain

$$w_1 = -\frac{2 \sin \frac{\varphi}{2} \cos \frac{\varphi}{2}}{2 \cos^2 \frac{\varphi}{2}} e^{-i\gamma} w_2 = -\tan \frac{\varphi}{2} e^{-i\gamma} w_2 \quad (\text{E.29})$$

Symmetrizing this expression by multiplying with $\cos \varphi/2 e^{i\gamma/2}$ yields

$$\cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} w_1 = -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} w_2 \quad (\text{E.30})$$

Imposing the normalization condition (E.22) gives rise to the solution

$$w_1 = -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \quad \text{and} \quad w_2 = \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \quad (\text{E.31})$$

which can be expressed in terms of the $\{|\uparrow\rangle, |\downarrow\rangle\}$ basis

$$|-\rangle = -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |\uparrow\rangle + \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |\downarrow\rangle \quad (\text{E.32})$$

For a unitary transformation the column vectors of the representation matrix must be mutual orthogonal (this is expected for the eigenvectors of an hermitian operator as well)

$$\begin{aligned} \langle + | - \rangle &= \mathbf{v}^* \cdot \mathbf{w} = -\cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} + \sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \\ &= -\cos \frac{\varphi}{2} \sin \frac{\varphi}{2} + \sin \frac{\varphi}{2} \cos \frac{\varphi}{2} = 0 \end{aligned} \quad (\text{E.33})$$

The eigenvector representation of $|+\rangle$ and $|-\rangle$ can be inverted in order to represent the initial vectors $|\uparrow\rangle$ and $|\downarrow\rangle$ in the new eigenbasis $\{|+\rangle, |-\rangle\}$

$$|\uparrow\rangle = \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |+\rangle - \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} |-\rangle \quad (\text{E.34})$$

$$|\downarrow\rangle = \sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |+\rangle + \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} |-\rangle \quad (\text{E.35})$$

Summary: Unitary Transformation of Operators and Basis States

The unitary transformation of the basis $\{|\uparrow\rangle, |\downarrow\rangle\}$ to the eigenbasis of an operator $\{|+\rangle, |-\rangle\}$ can be expressed in the notation of Appendix C, equation (C.2).

$$\begin{aligned} \begin{Bmatrix} |+\rangle \\ |-\rangle \end{Bmatrix} &= Q \begin{Bmatrix} |\uparrow\rangle \\ |\downarrow\rangle \end{Bmatrix} = \begin{Bmatrix} |\uparrow\rangle \langle \uparrow | + \rangle + |\downarrow\rangle \langle \downarrow | + \rangle \\ |\uparrow\rangle \langle \uparrow | - \rangle + |\downarrow\rangle \langle \downarrow | - \rangle \end{Bmatrix} = \begin{Bmatrix} |\uparrow\rangle \overbrace{\langle \uparrow | + \rangle}^{v_1} + |\downarrow\rangle \overbrace{\langle \downarrow | + \rangle}^{v_2} \\ \underbrace{|\uparrow\rangle \langle \uparrow | - \rangle}_{w_1} + \underbrace{|\downarrow\rangle \langle \downarrow | - \rangle}_{w_2} \end{Bmatrix} \\ \hookrightarrow Q &= \begin{pmatrix} v_1 & w_1 \\ v_2 & w_2 \end{pmatrix} = \begin{pmatrix} \langle \uparrow | + \rangle & \langle \uparrow | - \rangle \\ \langle \downarrow | + \rangle & \langle \downarrow | - \rangle \end{pmatrix} \end{aligned} \quad (\text{E.36})$$

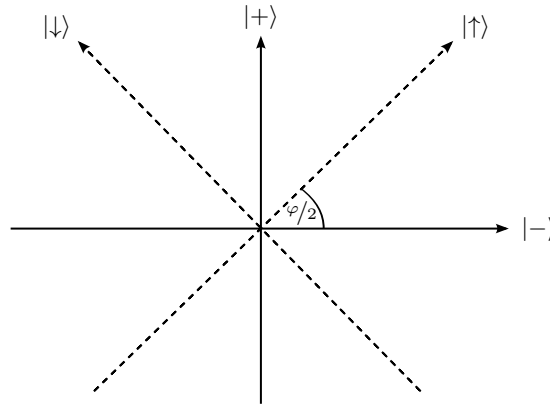


Figure E.1 Rotation of the basis vectors caused by the unitary transformation Q

In our case the transformation matrix Q looks like

$$\begin{Bmatrix} |+ \rangle \\ |- \rangle \end{Bmatrix} = Q \begin{Bmatrix} |\uparrow \rangle \\ |\downarrow \rangle \end{Bmatrix} = \begin{pmatrix} \cos \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} & -\sin \frac{\varphi}{2} e^{-i\frac{\gamma}{2}} \\ \sin \frac{\varphi}{2} e^{i\frac{\gamma}{2}} & \cos \frac{\varphi}{2} e^{i\frac{\gamma}{2}} \end{pmatrix} \begin{Bmatrix} |\uparrow \rangle \\ |\downarrow \rangle \end{Bmatrix} \quad (\text{E.37})$$

which can be considered as a rotation with the angle $\varphi/2$ combined with an phase or $U(1)$ -rotation with the angle $\gamma/2$ as shown in Figure E.1. The components of a state vector transforms with the inverse transformation matrix $Q^{-1} \equiv Q^\dagger$ which is the adjoint of the transformation matrix for unitary transformations. For a state vector $|\psi\rangle$ represented in the basis $\{|\uparrow\rangle, |\downarrow\rangle\}$ the transformed components read

$$\begin{aligned} |\psi\rangle &= \psi_+ |+ \rangle + \psi_- |- \rangle \\ &\stackrel{(\text{E.37})}{=} \psi_+ (Q_{11} |\uparrow \rangle + Q_{21} |\downarrow \rangle) + \psi_- (Q_{12} |\uparrow \rangle + Q_{22} |\downarrow \rangle) \\ &= \psi_\uparrow |\uparrow \rangle + \psi_\downarrow |\downarrow \rangle \end{aligned} \quad (\text{E.38})$$

As one can see the components of the state vector are transformed with the inverse transformation matrix

$$\hookrightarrow \psi_\uparrow = Q_{11}\psi_+ + Q_{12}\psi_- \quad \psi_\downarrow = Q_{21}\psi_+ + Q_{22}\psi_- \quad (\text{E.39})$$

$$\psi_+ = Q_{11}^*\psi_\uparrow + Q_{21}^*\psi_\downarrow \quad \psi_- = Q_{12}^*\psi_\uparrow + Q_{22}^*\psi_\downarrow \quad (\text{E.40})$$

Written in matrix-vector notation equation (E.39) and (E.40) yields

$$|\psi\rangle_{\uparrow\downarrow} = \begin{pmatrix} \psi_\uparrow \\ \psi_\downarrow \end{pmatrix} = Q |\psi\rangle_{\pm} = \begin{pmatrix} Q_{11} & Q_{12} \\ Q_{21} & Q_{22} \end{pmatrix} \begin{pmatrix} \psi_+ \\ \psi_- \end{pmatrix} \quad (\text{E.41})$$

$$|\psi\rangle_{\pm} = \begin{pmatrix} \psi_+ \\ \psi_- \end{pmatrix} = Q^\dagger |\psi\rangle_{\uparrow\downarrow} = \begin{pmatrix} Q_{11}^* & Q_{21}^* \\ Q_{12}^* & Q_{22}^* \end{pmatrix} \begin{pmatrix} \psi_\uparrow \\ \psi_\downarrow \end{pmatrix} \quad (\text{E.42})$$

Equivalently one can directly adopt the formula from equation (C.11) in Appendix C to find the unitary transformations of operators

$$A_{\pm} = Q^{-1} A_{\uparrow\downarrow} Q = Q^{\dagger} A_{\uparrow\downarrow} Q \quad (\text{E.43})$$

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F

Fourier Transformation

F.1 One-Dimensional Discrete Fourier Transformation

The discrete Fourier transform in one dimension is defined as the following summation over (one-dimensional) lattice points x_i , while the summation in the Fourier transformed lattice runs over $y_j = \frac{2\pi}{N}j$ with $j = 1, \dots, N$

$$f_i = f(x_i) = \frac{1}{N} \sum_{j=1}^N \tilde{f}(y_j) e^{iy_j x_i} = \frac{1}{N} \sum_{j=1}^N \tilde{f}_j e^{iy_j x_i} \quad (\text{F.1})$$

$$\tilde{f}_j = \tilde{f}(y_j) = \sum_{i=1}^N f(x_i) e^{-iy_j x_i} = \sum_{i=1}^N f_i e^{-iy_j x_i} \quad (\text{F.2})$$

Equation (F.2) is the inverse Fourier transformation which can be easily seen by inserting and using the Fourier representation of the delta distribution

$$\frac{1}{N} \sum_{j=1}^N 1 \cdot e^{iy_j x_i} = \delta(x_i) \quad \sum_{i=1}^N \frac{1}{N} e^{-iy_j x_i} = \tilde{\delta}(y_j) \quad (\text{F.3})$$

$$\sum_{i=1}^N \delta(x_i) e^{-iy_j x_i} = 1 \quad \frac{1}{N} \sum_{j=1}^N \tilde{\delta}(y_j) e^{iy_j x_i} = \frac{1}{N}^* \quad (\text{F.4})$$

into equation (F.1)

$$\begin{aligned} f(x_i) &= \frac{1}{N} \sum_{j=1}^N \tilde{f}(y_j) e^{iy_j x_i} = \frac{1}{N} \sum_{j=1}^N \sum_{k=1}^N f(x_k) e^{-iy_j x_k} e^{iy_j x_i} \\ &= \sum_{k=1}^N f(x_k) \frac{1}{N} \sum_{j=1}^N e^{iy_j (x_i - x_k)} = \sum_{k=1}^N f(x_k) \delta(x_i - x_k) = f(x_i) \end{aligned} \quad (\text{F.5})$$

* $\delta(x_i) = 1/N$ and $\tilde{\delta}(y_j) = 1$

and analogous for the Fourier transform

$$\begin{aligned}\tilde{f}(y_j) &= \sum_{i=1}^N f(x_i) e^{-iy_j x_i} = \frac{1}{N} \sum_{i=1}^N \sum_{k=1}^N \tilde{f}(y_k) e^{iy_k x_i} e^{-iy_j x_i} \\ &= \sum_{k=1}^N \tilde{f}(y_k) \frac{1}{N} \sum_{i=1}^N e^{-i(y_j - y_k)x_i} = \sum_{k=1}^N \tilde{f}(y_k) \delta(y_j - y_k) = \tilde{f}(y_j)\end{aligned}\quad (\text{F.6})$$

F.2 d -Dimensional Discrete Fourier Transformation

Now the generalization from the one-dimensional Fourier transformation to d dimensions is quite simple. Instead of a one-dimensional lattice, there is d -dimensional lattice with d -dimensional lattice vectors $\mathbf{r} = (r_1, \dots, r_d)^t$, as well as a d -dimensional reciprocal lattice with reciprocal lattice vectors $\mathbf{k} = 2\pi(\frac{k_1}{N_1}, \dots, \frac{k_d}{N_d})^t$

$$\begin{aligned}f(\mathbf{r}) &= \frac{1}{N_1} \sum_{k_1=1}^{N_1} \tilde{f}(\mathbf{k}) e^{i\frac{2\pi}{N_1} k_1 r_1} \times \dots \times \frac{1}{N_d} \sum_{k_d=1}^{N_d} \tilde{f}(\mathbf{k}) e^{i\frac{2\pi}{N_d} k_d r_d} \\ &= \prod_{i=1}^d \left[\frac{1}{N_i} \sum_{k_i=1}^{N_i} \tilde{f}(\mathbf{k}) e^{i\frac{2\pi}{N_i} k_i r_i} \right] = \frac{1}{N} \sum_{\mathbf{k}} \tilde{f}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}}\end{aligned}\quad (\text{F.7})$$

The formal notation denotes the sum over \mathbf{k} as the d -fold sum over each vector component and $N = \prod_i^d N_i$. The inverse Fourier transformation can be cast into

$$\tilde{f}(\mathbf{k}) = \sum_{\mathbf{r}} f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}}\quad (\text{F.8})$$

All properties of the one-dimensional Fourier transform are also valid in d dimensions, especially the representation of the delta distribution

$$\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} = \prod_{i=1}^d N_i \delta(\mathbf{r}) \qquad \sum_{\mathbf{r}} e^{-i\mathbf{k}\cdot\mathbf{r}} = \prod_{i=1}^d N_i \tilde{\delta}(\mathbf{k})\quad (\text{F.9})$$

or in the special case of a hypercubic lattice $N_i = \bar{N}$

$$\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} = \bar{N}^d \delta(\mathbf{r}) \qquad \sum_{\mathbf{r}} e^{-i\mathbf{k}\cdot\mathbf{r}} = \bar{N}^d \tilde{\delta}(\mathbf{k})\quad (\text{F.10})$$

F.3 Continuous Fourier Transformation

The discrete Fourier transformation can also be generalized to a continuous Fourier transformation via replacing the discrete sum with an Riemann sum that can be cast into an integral

$$f(x) = \int_{-\infty}^{\infty} \frac{dy}{2\pi} \tilde{f}(y) e^{iyx}\quad (\text{F.11})$$

$$\tilde{f}(y) = \int_{-\infty}^{\infty} dx f(x) e^{-iyx} \quad (\text{F.12})$$

In d dimensions the continuous Fourier transformation (and its inverse) reads

$$f(\mathbf{r}) = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \tilde{f}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} \quad (\text{F.13})$$

$$\tilde{f}(\mathbf{k}) = \int_{-\infty}^{\infty} d^d\mathbf{r} f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} \quad (\text{F.14})$$

Note that the factor $\frac{1}{2\pi}$ can be shifted to the Fourier space or the real space measure or even split into a symmetric form (e.g. $d\mathbf{r}/\sqrt{2\pi}$ and $d\mathbf{k}/\sqrt{2\pi}$). The important thing is that the product of f and \tilde{f} is normalized to $\frac{1}{2\pi}$. The convention that is used here will be as follows: For functions the factor $\frac{1}{2\pi}$ is always under the Fourier space measure whereas for operators it will be split symmetrically between both measures

$$a(\mathbf{r}) = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{\sqrt{2\pi}} \right)^d \tilde{a}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} \quad \left(\text{compare } a(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} \tilde{a}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} \right) \quad (\text{F.15})$$

$$\tilde{a}(\mathbf{k}) = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{r}}{\sqrt{2\pi}} \right)^d a(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} \quad \left(\text{compare } \tilde{a}(\mathbf{k}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{r}} a(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} \right) \quad (\text{F.16})$$

In the continuous Fourier transformation there exists also a representation of the delta distribution

$$\delta(\mathbf{r} - \mathbf{r}') = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \quad \tilde{\delta}(\mathbf{k} - \mathbf{k}') = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{r}}{2\pi} \right)^d e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \quad (\text{F.17})$$

Equation (F.17) can be easily derived using the definition of the delta distribution

$$\begin{aligned} \tilde{\delta}(\mathbf{k}) &= \int_{-\infty}^{\infty} d^d\mathbf{r} \delta(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} = e^0 = 1 \\ \delta(\mathbf{r}) &= \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \tilde{\delta}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} = \frac{e^0}{(2\pi)^d} = \frac{1}{(2\pi)^d} \end{aligned} \quad (\text{F.18})$$

and reinserting equation (F.18) into the definition of the Fourier Transformation yields

$$\delta(\mathbf{r}) = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \underbrace{\tilde{\delta}(\mathbf{k})}_{=1} e^{i\mathbf{k}\cdot\mathbf{r}} = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d e^{i\mathbf{k}\cdot\mathbf{r}}$$

$$\begin{aligned}\tilde{\delta}(\mathbf{k}) &= \int_{-\infty}^{\infty} d^d \mathbf{r} \underbrace{\delta(\mathbf{r})}_{= \frac{1}{(2\pi)^d}} e^{-i\mathbf{k}\cdot\mathbf{r}} = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{r}}{2\pi}\right)^d e^{-i\mathbf{k}\cdot\mathbf{r}}\end{aligned}\quad (\text{F.19})$$

F.4 Poisson Summation Formula

For periodic function f with period T one can find a periodic extension of f and its (continuous) Fourier transform \tilde{f} (see Section F.3). In this case the general Poisson summation formula

$$\sum_{n \in \mathbb{Z}} f(t + nT) e^{-i(2\pi\nu)nT} = \frac{1}{T} \sum_{k \in \mathbb{Z}} \tilde{f}\left(2\pi\nu + \frac{2\pi k}{T}\right) e^{i2\pi\nu t + i2\pi k \frac{t}{T}} \quad (\text{F.20})$$

holds with additional parameters t and ν . This formula can be easily generalized to d dimensions via introducing the following d dimensional vectors

$$\mathbf{R}_n = T\mathbf{n} = T \begin{pmatrix} n_1 \\ \vdots \\ n_d \end{pmatrix} \quad \mathbf{k} = \frac{2\pi}{T} \begin{pmatrix} k_1 \\ \vdots \\ k_d \end{pmatrix} \quad \mathbf{q} = 2\pi\nu \in \mathbb{R}^d \quad \mathbf{t} \in \mathbb{R}^d \quad (\text{F.21})$$

Notice that the integers in \mathbf{k} actually mean $\mathbf{k} = \frac{2\pi}{T}(k_1, \dots, k_d)^t$ with $k_i \in \mathbb{Z}$, which is an useful simplification for physical applications. For a d dimensional periodic function f with period T in all directions one obtains

$$\begin{aligned}\sum_{\mathbf{n} \in \mathbb{Z}^d} f(\mathbf{t} + \mathbf{n}T) e^{-i2\pi\nu \cdot \mathbf{n}T} &= \prod_{i=1}^d \left[\left(\frac{1}{T}\right) \sum_{k_i \in \mathbb{Z}} \tilde{f}(2\pi\nu + \mathbf{k}) e^{i2\pi\nu \cdot \mathbf{t}} e^{i\frac{2\pi}{T} k_i t_i} \right] \\ &= \left(\frac{1}{T}\right)^d \sum_{\mathbf{k} \in \mathbb{Z}^d} \tilde{f}(2\pi\nu + \mathbf{k}) e^{i2\pi\nu \cdot \mathbf{t} + i\mathbf{k} \cdot \mathbf{t}} \\ \hookrightarrow \sum_{\mathbf{R}_n \in \mathbb{Z}^d} f(\mathbf{t} + \mathbf{R}_n) e^{-i\mathbf{q} \cdot \mathbf{R}_n} &= \frac{1}{T^d} \sum_{\mathbf{k} \in \mathbb{Z}^d} \tilde{f}(\mathbf{q} + \mathbf{k}) e^{i(\mathbf{q} + \mathbf{k}) \cdot \mathbf{t}}\end{aligned}\quad (\text{F.22})$$

For $\mathbf{t} = \mathbf{0}$ and $T = a$ with a the lattice spacing of a d -dimensional hypercube, \mathbf{k} can be considered as the lattice vector of the reciprocal hypercubic lattice. Then an useful special case of equation (F.22) gives rise to

$$\sum_{\mathbf{R}_n \in \mathbb{Z}^d} f(\mathbf{R}_n) e^{-i\mathbf{q} \cdot \mathbf{R}_n} = \frac{1}{a^d} \sum_{\mathbf{k} \in \mathbb{Z}^d} \tilde{f}(\mathbf{q} + \mathbf{k}) \quad (\text{F.23})$$

This special formula can be interpreted as an identity of a discrete Fourier transformation relating real and Fourier space to one another and hence discrete Fourier transform (or Fourier series) to the continuous Fourier transform of a function.

F.5 Physical Application of Fourier Transformations

F.5.1 $d = 3$ dimensional Fourier Series (discrete in \mathbf{k} -space)

The physical picture of the d -dimensional Fourier series is a transformation between complementary physical quantities like time t and energy E or position \mathbf{r} and momentum k (in units $\hbar = 1$). In the following, using physical notation we will suppress the “ \sim ” on top of the function symbols and distinguish function and its Fourier transform just by looking at the argument indicating in which space the function resides. For quantum mechanical operators we can define the following series expansion (in plane waves $e^{i\mathbf{k}\cdot\mathbf{r}}$)

$$a_{\mathbf{k}}^{\dagger} = \frac{1}{\sqrt{V}} \int_V d\mathbf{r} e^{i\mathbf{k}\cdot\mathbf{r}} a(\mathbf{r})^{\dagger} \quad a_{\mathbf{k}} = \frac{1}{\sqrt{V}} \int_V d\mathbf{r} e^{-i\mathbf{k}\cdot\mathbf{r}} a(\mathbf{r}) \quad (\text{F.24})$$

$a_{\mathbf{k}}^{\dagger}$ and $a_{\mathbf{k}}$ are the coefficient of the different plane waves, building up the total wave packet that corresponds to the creation operator $a(\mathbf{r})^{\dagger}$ (or annihilation operator) that creates a particle at position \mathbf{r}

$$a(\mathbf{r})^{\dagger} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}}^{\dagger} \quad a(\mathbf{r}) = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}} \quad (\text{F.25})$$

whereas for general function $f(\mathbf{r})$ the Fourier series expansion is defined asymmetrically

$$f(\mathbf{r}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} f_{\mathbf{k}} \quad f_{\mathbf{k}} = \frac{1}{V} \int_V d^3\mathbf{r} e^{-i\mathbf{k}\cdot\mathbf{r}} f(\mathbf{r}) \quad (\text{F.26})$$

Since (F.26) is an integration over the entire real space V , the factor $\frac{1}{V}$ gives the right normalization.

F.5.2 $d = 3$ dimensional discrete Fourier transformation

A second application is the transformation of a periodic function $f(\mathbf{r} + \mathbf{R}) = f(\mathbf{r})$ defined only on discrete lattice points in real space with lattice vector \mathbf{R} . We assume for simplicity a cubic lattice with lattice constant a . A Fourier transformation to the \mathbf{k} -space with the reciprocal lattice defined by the basis vectors

$$\mathbf{b}_i = \frac{2\pi}{V_E} \frac{1}{2} \sum_{j,k=1}^3 \epsilon_{ijk} \mathbf{a}_j \times \mathbf{a}_k \quad \text{with} \quad V_E = \sum_{i,j,k=1}^3 \epsilon_{ijk} \mathbf{a}_i \cdot \mathbf{a}_j \cdot \mathbf{a}_k = \det(\mathbf{a}_1 | \mathbf{a}_2 | \mathbf{a}_3) \quad (\text{F.27})$$

gives the Fourier transform which is defined with the help of the reciprocal lattice vector \mathbf{G} . V_E denotes the volume of the primitive cell (spanned by the basis vectors \mathbf{a}_i) and is equal to a^3 in the cubic case. The volume of the complete system is the volume of the unit cell times the total number of lattice points, since the primitive unit cell just contains one lattice point.

$$V = NV_E = V_E \prod_{i=1}^3 \bar{N} = \bar{N}^3 V_E = \bar{N}^3 a^3 \quad (\text{F.28})$$

The lattice spacing of the (also cubic) reciprocal lattice is then $\frac{2\pi}{a}$. There exists the following identity between R and G

$$e^{i\mathbf{G}\cdot\mathbf{R}} = 1 \quad (\text{F.29})$$

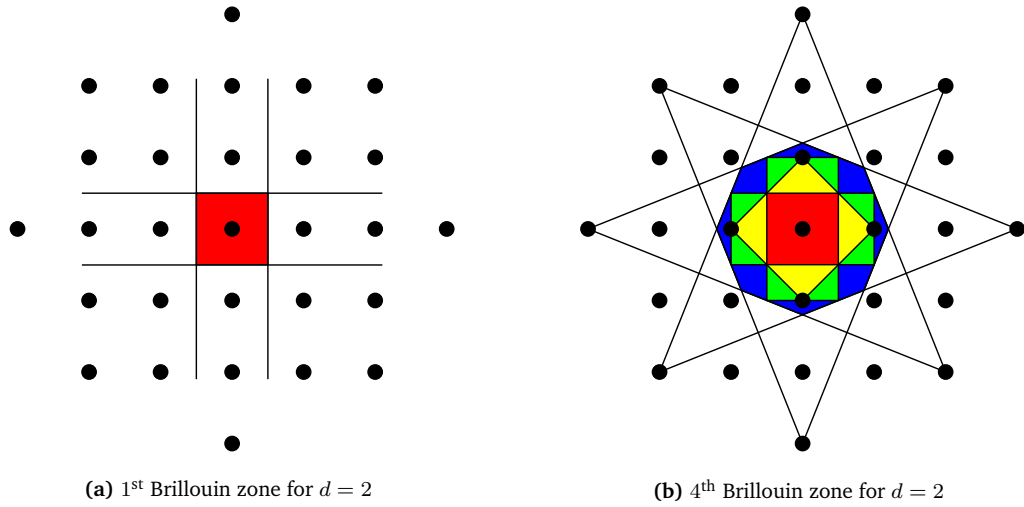


Figure F.1 The n^{th} Brillouin zone consist of all points in \mathbf{k} -space that crosses $n - 1$ Bragg lines

because the product of the lattice vector \mathbf{R} with the reciprocal lattice vector \mathbf{G} is

$$\mathbf{G} \cdot \mathbf{R} = \sum_{i=1}^3 G_i R_i \mathbf{a}_i \cdot \mathbf{b}_i = \pi \sum_{i,j,k=1}^3 G_i R_i \frac{\epsilon_{ijk} \mathbf{a}_i \cdot (\mathbf{a}_j \times \mathbf{a}_k)}{V_E} = 2\pi \underbrace{\sum_{i=1}^3 G_i R_i}_{\in \mathbb{Z}} \quad (\text{F.30})$$

The discrete Fourier transform of a periodic function $f(\mathbf{r} + \mathbf{R}) = f(\mathbf{r})$ is then given by

$$f(\mathbf{r}) = \sum_{\mathbf{G} \in \text{RL}} f_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}} \quad (\text{F.31})$$

Using the plane wave basis to expand the operators, only the wave vectors of the so called first Brillouin zone are needed. The first Brillouin zone is defined as all wave vectors \mathbf{k} of the reciprocal lattice that are closer to $\mathbf{G} = \mathbf{0}$ than to all other reciprocal vectors $\mathbf{G} \neq \mathbf{0}$. In Figure F.1a the 1st Brillouin zone is shown in red, whereas the solid line indicates the Bragg lines (intersecting the line segment connecting the (nearest) neighbors at a right angle). Now operators and functions can be expanded with respect to the discrete sum over all wave vectors in the first Brillouin zone

$$f(\mathbf{r} = a\mathbf{R}) = \frac{1}{\underbrace{(Na)^3}_{=V}} \sum_{\mathbf{K} \in \text{1.BZ}} e^{i\frac{\mathbf{K}}{a} \cdot a\mathbf{R}} f\left(\frac{\mathbf{K}}{a}\right) \quad (\text{F.32})$$

$$f\left(\mathbf{k} = \frac{\mathbf{K}}{a}\right) = \sum_{\mathbf{R}} e^{-i\frac{\mathbf{K}}{a} \cdot a\mathbf{R}} f(a\mathbf{R}) \quad (\text{F.33})$$

Here V denotes the real space volume of the cubic lattice (with lengths of the sides Na). The vector $\mathbf{R} = (R_1, R_2, R_3)^t$ and the vector $\mathbf{K} = \frac{2\pi}{N}(K_1, K_2, K_3)^t$ given by the discrete Fourier transformation defined in equation (F.7) are multiplied by a length scale, which is the lattice spacing, the physical distance between two lattice points. In order to leave the (general) scalar

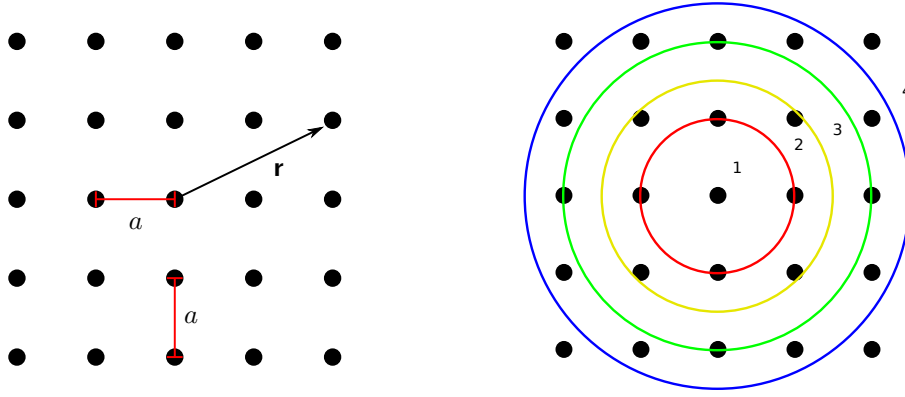


Figure F.2 Left Panel: Real space lattice with lattice spacing a .
 Right Panel: The four next nearest neighbors R_1, R_2, R_3, R_4

product

$$\mathbf{K} \cdot \mathbf{R} = 2\pi \sum_{i=1}^3 \frac{K_i}{N_i} R_i \quad (\text{F.34})$$

in the exponent of the e -function in the definition of the discrete Fourier transformation invariant, the \mathbf{k} -space must scale with the dimension (length) $^{-1}$

$$\mathbf{K} \cdot \mathbf{R} = \mathbf{k} \cdot \mathbf{r} \quad \text{with} \quad \mathbf{k} = \frac{\mathbf{K}}{a} = \frac{2\pi}{Na} (K_1, K_2, K_3)^t \quad \text{and} \quad \mathbf{r} = a\mathbf{R} = a(R_1, R_2, R_3)^t \quad (\text{F.35})$$

where \mathbf{r} denotes the position vector in the real space lattice and \mathbf{k} the vector pointing on points of the reciprocal lattice in the first Brillouin zone. Thus, the sum $\sum_{\mathbf{K}}$ is running over $k_i = -\bar{N}/2, \dots, \bar{N}/2$. Now functions and operators can be Fourier transformed in the following way

$$a(\mathbf{R}) = \frac{1}{N^{3/2}} \sum_{\mathbf{K}} e^{i\mathbf{K} \cdot \mathbf{R}} a(\mathbf{K}) \quad a(\mathbf{K}) = \frac{1}{N^{3/2}} \sum_{\mathbf{R}} e^{-i\mathbf{K} \cdot \mathbf{R}} a(\mathbf{R}) \quad (\text{F.36})$$

$$f(\mathbf{R}) = \frac{1}{N^3} \sum_{\mathbf{K}} e^{i\mathbf{K} \cdot \mathbf{R}} f(\mathbf{K}) \quad f(\mathbf{K}) = \sum_{\mathbf{R}} e^{-i\mathbf{K} \cdot \mathbf{R}} f(\mathbf{R}) \quad (\text{F.37})$$

Sometimes it is useful to rewrite these equation in terms of new summation variables, just counting the number of neighbors from the starting point. Then the real space sum over \mathbf{R} can be replaced by one single sum

$$\sum_{\mathbf{R}} \longrightarrow \sum_{R_n} \equiv \text{sum over all } n^{\text{th}} \text{ nearest neighbor} \quad R_i = -\frac{\bar{N}}{2}, \dots, \frac{\bar{N}}{2} \longrightarrow n = 1, \dots, \bar{N} \quad (\text{F.38})$$

where $|\mathbf{R}_1| = 1$ denotes the nearest neighbor, $|\mathbf{R}_2| = \sqrt{2}$ denotes the next nearest neighbor and so on as depicted in Figure F.2. In the following we will not distinguish between \mathbf{K} and \mathbf{k} and by writing

$$\sum_{\mathbf{k} \in \text{1.BZ}} \dots \equiv \sum_{k_1 = -\bar{N}/2}^{\bar{N}/2} \sum_{k_2 = -\bar{N}/2}^{\bar{N}/2} \sum_{k_3 = -\bar{N}/2}^{\bar{N}/2} \dots \quad (\text{F.39})$$

we assume the sum over the first Brillouin zone. Finally we have

$$a(\mathbf{R}_n) = \frac{1}{N^{3/2}} \sum_{\mathbf{k} \in 1.\text{BZ}} e^{i\mathbf{k} \cdot \mathbf{R}_n} a(\mathbf{k}) \quad a(\mathbf{k}) = \frac{1}{N^{3/2}} \sum_{\mathbf{R}_n} e^{-i\mathbf{k} \cdot \mathbf{R}_n} a(\mathbf{R}_n) \quad (\text{F.40})$$

$$f(\mathbf{R}_n) = \frac{1}{N^3} \sum_{\mathbf{k} \in 1.\text{BZ}} e^{i\mathbf{k} \cdot \mathbf{R}_n} f(\mathbf{k}) \quad f(\mathbf{k}) = \sum_{\mathbf{R}_n} e^{-i\mathbf{k} \cdot \mathbf{R}_n} f(\mathbf{R}_n) \quad (\text{F.41})$$

F.5.3 $d = 3$ dimensional Fourier Series (discrete in real space)

If one considers the continuum limit of the problem in equation (F.41) then we have to replace the sum over the first Brillouin zone with an integral over the whole \mathbf{k} -space volume \mathcal{V} .

$$\sum_{\mathbf{k} \in 1.\text{BZ}} \dots \longrightarrow \frac{1}{\mathcal{V}} \int_{\mathcal{V}} d^3\mathbf{k} \dots = \left(\frac{\bar{N}a}{2\pi} \right)^3 \int_{\mathcal{V}} d^3\mathbf{k} \dots \quad (\text{F.42})$$

or in general for a d -dimensional lattice

$$\sum_{\mathbf{k} \in 1.\text{BZ}} \dots \longrightarrow \left[\frac{\prod_{i=1}^d (N_i a_i)}{(2\pi)^d} \right] \int_{\mathcal{V}} d^d\mathbf{k} \dots \quad (\text{F.43})$$

or in the special case of a d -dimensional hypercubic lattice, respectively

$$\sum_{\mathbf{k} \in 1.\text{BZ}} \dots \longrightarrow \left(\frac{\bar{N}a}{2\pi} \right)^d \int_{\mathcal{V}} d^d\mathbf{k} \dots \quad (\text{F.44})$$

The volume of the real space V can be converted to the \mathbf{k} -space volume \mathcal{V} as follows

$$V = \frac{(2\pi)^d}{\mathcal{V}} \quad \mathcal{V} = \frac{(2\pi)^d}{V} \quad (\text{F.45})$$

F.5.4 Translational Invariant Systems

Consider a isotropic periodic hypercubic lattice such that, it is translational invariant. All physical observables must obey the invariance under the translational symmetry operation. Thus, all physical quantities are function of differences of lattice vectors only, since any point of the hypercubic lattice can be chosen as a reference point. Especially two point functions depending on two different position \mathbf{r} and \mathbf{r}' can only depend on their difference $\mathbf{r} - \mathbf{r}'$

$$f(\mathbf{r}, \mathbf{r}') = f(\mathbf{r} - \mathbf{r}') \quad (\text{F.46})$$

The Fourier transform of such a function will then have special symmetry properties

$$\begin{aligned} f(\mathbf{r}, \mathbf{r}') &= \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \left(\frac{d\mathbf{k}'}{2\pi} \right)^d \mathcal{F}(\mathbf{k}, \mathbf{k}') e^{i\mathbf{k} \cdot \mathbf{r}} e^{i\mathbf{k}' \cdot \mathbf{r}'} \\ &= \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \left(\frac{d\mathbf{k}'}{2\pi} \right)^d \mathcal{F}(\mathbf{k}, \mathbf{k}') e^{i(\mathbf{r}-\mathbf{r}') \cdot \mathbf{k}} e^{i(\mathbf{k}+\mathbf{k}') \cdot \mathbf{r}'} \end{aligned} \quad (\text{F.47})$$

Since the function cannot explicitly depend on \mathbf{r}' , the factor $e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}'}$ must be a constant, which leads to the following condition for the reciprocal vectors \mathbf{k} , \mathbf{k}' and the Fourier transform

$$\mathbf{k} = -\mathbf{k}' \quad \Rightarrow \quad \mathcal{F}(\mathbf{k}, \mathbf{k}') \propto \delta(\mathbf{k} - \mathbf{k}') \quad (\text{F.48})$$

By comparison with the Fourier transform of $f(\mathbf{r} - \mathbf{r}')$

$$f(\mathbf{r} - \mathbf{r}') = \int_{-\infty}^{\infty} \left(\frac{d\mathbf{k}}{2\pi} \right)^d \tilde{f}(\mathbf{k}) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \quad (\text{F.49})$$

one finds the following relation between the two Fourier transforms

$$\mathcal{F}(\mathbf{k}, \mathbf{k}') = (2\pi)^d \delta(\mathbf{k} - \mathbf{k}') \tilde{f}(\mathbf{k}) \quad (\text{F.50})$$

Therefore the Fourier transform of a translational invariant function can always be described by a single wave vector \mathbf{k} .

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Pauli Representation of the Lie Group $SU(2)$

G.1 General Properties of the Special Unitary Group $SU(n)$

The special unitary group $SU(n) = \{A \in (n \times n)\text{-matrices} \mid A^\dagger A = 1, \det A = 1\}$ is a subgroup of the unitary group, which consists of all unitary $n \times n$ -matrices. It is a real matrix group of dimension $n^2 - 1$ because there exists the constraint that $\det A = 1$, reducing the number of independent matrix elements by one. The corresponding Lie algebra $su(n)$ is constructed with regard to the tangential space to the identity element $\mathbb{1}$ of the group. With the help of the exponential map each Lie algebra element can be mapped onto a Lie group element

$$f: su(n) \longrightarrow SU(n) \tag{G.1}$$

$$su(n) \ni g_a \longmapsto e^{ig_a} = g \in SU(n)$$

The elements of the Lie algebra $su(n)$ are real, traceless and antihermitian matrices. The Lie group elements can be generated using infinitesimal generators T_a which can be represented as traceless hermitian matrices

$$\text{tr } T_a = 0 \qquad T_a^\dagger = T_a \tag{G.2}$$

There exists a fundamental representation where the generators can be represented as follows

$$T_a T_b = \frac{1}{2n} \delta_{ab} \mathbb{1} + \frac{1}{2} \sum_{c=1}^{n^2-1} (if_{abc} + d_{abc}) T_c \tag{G.3}$$

The structure constants f_{abc} are totally antisymmetric and describe the commuting part of the generators, whereas d_{abc} is totally symmetric and describe the anticommutating part

$$[T_a, T_b] = i \sum_{c=1}^{n^2-1} f_{abc} T_c \tag{G.4}$$

$$\{T_a, T_b\} = \frac{1}{n} \delta_{ab} \mathbb{1} + \sum_{c=1}^{n^2-1} d_{abc} T_c \quad (\text{G.5})$$

Additionally one can define the so called adjoint representation where the generators are represented by matrices with the structure constants f_{abc} as elements

$$(T_a)_{jk} = -i f_{ajk} \quad (\text{G.6})$$

G.2 Properties of $SU(2)$

The Lie group $SU(2)$ consists of all unitary (2×2) -matrices with unit determinant. In the fundamental representation the $n^2 - 1 = 3$ infinitesimal generators of the $SU(2)$ can be represented as Pauli matrices

$$\frac{1}{2} \sigma_1 = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \frac{1}{2} \sigma_2 = \frac{1}{2} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \frac{1}{2} \sigma_3 = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (\text{G.7})$$

From the matrix product relation

$$\left(\frac{1}{2} \sigma_a \right) \left(\frac{1}{2} \sigma_b \right) = \frac{1}{4} \delta_{ab} \mathbb{1} + \frac{1}{2} \sum_{c=1}^3 i \varepsilon_{abc} \left(\frac{1}{2} \sigma_c \right) \quad \leftrightarrow \quad \sigma_a \sigma_b = \delta_{ab} \mathbb{1} + \sum_{c=1}^3 i \varepsilon_{abc} \sigma_c \quad (\text{G.8})$$

one can immediately read off $f_{abc} = \varepsilon_{abc}$ and $d_{abc} = 0$ which leads to

$$[\sigma_a, \sigma_b] = 2i \varepsilon_{abc} \sigma_c \quad (\text{G.9})$$

$$\{\sigma_a, \sigma_b\} = 2 \delta_{ab} \mathbb{1} \quad (\text{G.10})$$

The group elements of the Lie groups can be described by $n^2 - 1 = 3$ continuous parameters which can be written as a three dimensional vector $\mathbf{n} = \varphi \mathbf{n}^0$ and the infinitesimal generators in terms of Pauli matrices as $\frac{1}{2} \boldsymbol{\sigma}$

$$\begin{aligned} e^{i \sum_a \alpha_a T_a} &= e^{i \frac{1}{2} \mathbf{n} \cdot \boldsymbol{\sigma}} = e^{i \frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma}} \\ &= \sum_{k=0}^{\infty} \frac{(i \frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma})^k}{k!} \\ &= \sum_{k=0}^{\infty} \frac{(i \frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k}}{2k!} + \sum_{k=0}^{\infty} \frac{(i \frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k+1}}{(2k+1)!} \\ &= \sum_{k=0}^{\infty} \frac{(i^2)^k (\frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k}}{2k!} + \sum_{k=0}^{\infty} \frac{i (i^2)^k (\frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k+1}}{(2k+1)!} \\ &= \sum_{k=0}^{\infty} \frac{(-1)^k (\frac{\varphi}{2})^{2k} (\mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k}}{(2k)!} + i \sum_{k=0}^{\infty} \frac{(-1)^k (\frac{\varphi}{2})^{2k+1} (\mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k+1}}{(2k+1)!} \end{aligned} \quad (\text{G.11})$$

After splitting the sum into an odd and an even part and using the following relations

$$\begin{aligned} (\mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k} &= [(\mathbf{n}^0 \cdot \boldsymbol{\sigma})(\mathbf{n}^0 \cdot \boldsymbol{\sigma})]^k = \left[(n_1^0 \sigma_1)^2 + (n_2^0 \sigma_2)^2 + (n_3^0 \sigma_3)^2 + n_1^0 n_2^0 (\sigma_1 \sigma_2 + \sigma_2 \sigma_1) \right. \\ &\quad \left. + n_1^0 n_3^0 (\sigma_1 \sigma_3 + \sigma_3 \sigma_1) + n_2^0 n_3^0 (\sigma_2 \sigma_3 + \sigma_3 \sigma_2) \right]^k \\ &= \left([(n_1^0)^2 + (n_2^0)^2 + (n_3^0)^2] \mathbf{1} \right)^k = \mathbf{1} \end{aligned} \quad (\text{G.12})$$

$$(\mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k+1} = (\mathbf{n}^0 \cdot \boldsymbol{\sigma})^{2k} (\mathbf{n}^0 \cdot \boldsymbol{\sigma}) = \mathbf{1} (\mathbf{n}^0 \cdot \boldsymbol{\sigma}) = (\mathbf{n}^0 \cdot \boldsymbol{\sigma}) \quad (\text{G.13})$$

equation (G.11) can be simplified

$$\begin{aligned} \hookrightarrow e^{i \frac{\varphi}{2} \mathbf{n}^0 \cdot \boldsymbol{\sigma}} &= \sum_{k=0}^{\infty} \frac{(-1)^k \left(\frac{\varphi}{2}\right)^{2k}}{(2k)!} \mathbf{1} + i (\mathbf{n}^0 \cdot \boldsymbol{\sigma}) \sum_{k=0}^{\infty} \frac{(-1)^k \left(\frac{\varphi}{2}\right)^{2k+1}}{(2k+1)!} \\ &= \cos\left(\frac{\varphi}{2}\right) \mathbf{1} + i (\mathbf{n}^0 \cdot \boldsymbol{\sigma}) \sin\left(\frac{\varphi}{2}\right) \end{aligned} \quad (\text{G.14})$$

The absolute value of the vector $|\mathbf{n}| = \varphi$ can be identified with the rotation angle and the direction unit vector \mathbf{n}^0 as the rotation axis in three dimensional space. The $SU(2)$ describes a rotation with an rotational angle of 4π to complete the circle. The reason for this lies in the fact that the Lie algebra $su(2)$ is isomorphic to the Lie algebra $so(3)$ (which means that the generators of $SU(2)$ are infinitesimal rotations in three dimensional space) but the Lie group $SU(2)$ is the double covering group of the Lie group $SO(3)$. This leads to a homomorphic two-to-one mapping from $SU(2)$ to $SO(3)$ and to the fact that a rotation about 2π gives the original state multiplied by (-1) (setting $\varphi = 2\pi$ in equation (G.14) gives -1). With the help of the tensor product representations of higher spin systems can be constructed, e.g.

$$2 \otimes 2 = 3 \oplus 1 \quad (\text{G.15})$$

where the numbers represent the $(2S - 1)$ -dimensional representation of the $SU(2)$. The 3-dimensional representation is the adjoint representation whereas the 1 denotes the trivial representation of $SU(2)$.

G.3 Physical Application of $SU(2)$

The fundamental representation using the Pauli matrices can be used to describe spin- $1/2$ particles like electrons or two state systems with only two different quantum states or degrees of freedom (e.g. the lowest two states of a multi-state system that can be excited). Defining the following two state basis

$$\{|\uparrow\rangle, |\downarrow\rangle\} \equiv \left\{ \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix} \right\} \quad (\text{G.16})$$

the spin- $1/2$ operators are defined as $\mathbf{S} = \frac{\hbar}{2} \boldsymbol{\sigma}$

$$\mathcal{S}_x = \frac{\hbar}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \mathcal{S}_y = \frac{\hbar}{2} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \mathcal{S}_z = \frac{\hbar}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (\text{G.17})$$

Additionally, we can define creation and annihilation operators. If we label the ground state as $|\downarrow\rangle$ and the excited state as $|\uparrow\rangle$ then \mathcal{S}_+ is a creation operator that “creates” the excited state $|\uparrow\rangle$ (and destroys the ground state $|\downarrow\rangle$) whereas \mathcal{S}_- “destroys” the excited state (and creates the ground state)

$$\mathcal{S}_+ = \mathcal{S}_x + i\mathcal{S}_y = \hbar \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \mathcal{S}_- = \mathcal{S}_x - i\mathcal{S}_y = \hbar \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad (\text{G.18})$$

These operators can be written directly in the basis (G.16) as

$$\mathcal{S}_+ = \hbar |\uparrow\rangle \langle \downarrow| \quad \mathcal{S}_- = \hbar |\downarrow\rangle \langle \uparrow| \quad \mathcal{S}_z = \frac{\hbar}{2} (|\uparrow\rangle \langle \uparrow| - |\downarrow\rangle \langle \downarrow|) \quad (\text{G.19})$$

The operators $\mathcal{S}_+ = \mathcal{S}_-^\dagger$ and $\mathcal{S}_- = \mathcal{S}_+^\dagger$ obey the following commutator relations

$$[\mathcal{S}_z, \mathcal{S}_\pm] = \pm \hbar \mathcal{S}_\pm \quad [\mathcal{S}_+, \mathcal{S}_-] = 2\hbar \mathcal{S}_z \quad (\text{G.20})$$

which can be derived from equation (G.9).



Documentation of Mathematica Programs

H.1 Numerical-Diagonalization.nb

The eigenbasis of the Hamiltonian

$$\mathcal{H} = Bg^2 - d_0 E_{dc} \qquad \frac{\mathcal{H}}{B} = g^2 - \frac{dE_{dc}}{B} \frac{d_0}{d} \qquad (\text{H.1})$$

is determined by numerically diagonalize \mathcal{H} with MATHEMATICA and the help of the build-in function **Eigensystem**. The matrix representation in the spherical tensor basis of \mathcal{H} is generated with

```

RME[J_, j_, k_] := (-1)^(J) Sqrt[(2 J + 1) (2 j + 1)]
10           ThreeJSymbol[{J, 0},{k, 0},{j, 0}]/Sqrt[2 J + 1]
WET[J_, M_, k_, q_, j_, m_] := ClebschGordan[{j, m}, {k, q}, {J, M}]
12           RME[J, j, k]

```

where the self-defined functions WET defines the Wigner-Eckart theorem for spherical tensor operators $\mathcal{T}_q^{(k)}$ which consists of the Clebsch-Gordan-Coefficients and the reduced matrix element, the self-defined function RME. The Wigner-Eckart theorem is now used to calculate the matrix representation of the z -component of the dipole operator d_0 via

```

d[J_, M_, j_, m_, q_] := WET[J + M, M, 1, q, j + m, m]
14 d0[J_, M_, j_, m_] := d[J, M, j, m, 0]
d0m0 = Table[d0[J, 0, j, 0], {J, 0, n}, {j, 0, n}];
16 d0m1 = Table[d0[J, 1, j, 1], {J, 0, n}, {j, 0, n}];
d0m2 = Table[d0[J, 2, j, 2], {J, 0, n}, {j, 0, n}];

```

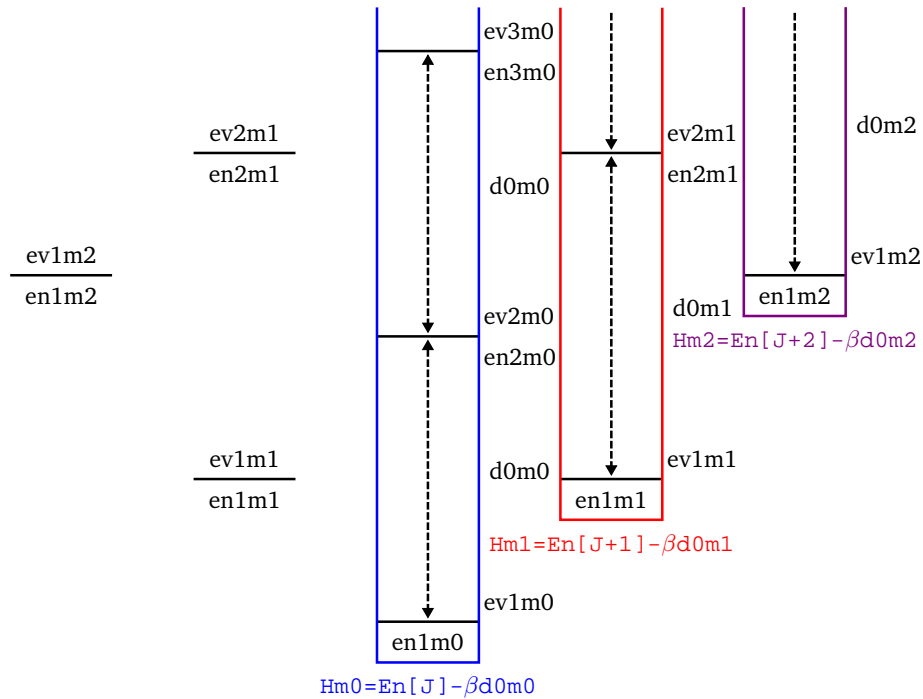


Figure H.1 Numerically determined eigenstates and eigenvalues of \mathcal{H} for $|M| = 0, 1, 2$

where the function $d[J, M, j, m, q]$ defines the dipole operator in spherical coordinates, $d0[J, M, j, m]$ the z -component of \mathbf{d} and $d0mi$ for $i = 0, 1, 2$ are the matrix representation of the dipole operators coupling $|J = i, |M| = i\rangle$. We are using the modulus of M since the matrix representations for the operators for $-M$ are identical and therefore the degeneracy between the $M \neq 0$ states is not lifted by applying a static electric field E_{dc} in z -direction. In the spherical tensor basis the \mathcal{J}^2 operator is diagonal and possesses the eigenvalues $J(J + 1)$. Thus the rotor term in the Hamiltonian \mathcal{H} can be described by the rotor-energy function

$$18 \quad \text{En}[J, M] := (J + M) (J + M + 1)$$

where we start for $|M| = 1, 2$ at $J = 1, 2$ since $|M| \neq 0$ states are only defined for $J \neq 0$ states. Now we are ready to calculate the matrix representation of \mathcal{H} for each value of M in the spherical tensor basis with the help of

$$20 \quad \begin{aligned} \text{Hm0}[\mathbf{Beta}] &:= \text{Table}[\text{En}[J, 0] \text{KroneckerDelta}[J, j] - \mathbf{Beta} \\ &\quad d0[J, 0, j, 0], \{J, 0, n\}, \{j, 0, n\}] \\ \text{Hm1}[\mathbf{Beta}] &:= \text{Table}[\text{En}[J, 1] \text{KroneckerDelta}[J, j] - \mathbf{Beta} \\ &\quad d0[J, 1, j, 1], \{J, 0, n\}, \{j, 0, n\}] \\ \text{Hm2}[\mathbf{Beta}] &:= \text{Table}[\text{En}[J, 2] \text{KroneckerDelta}[J, j] - \mathbf{Beta} \\ &\quad d0[J, 2, j, 2], \{J, 0, n\}, \{j, 0, n\}] \end{aligned}$$

For each β running from 0 to 10 in steps of 0.01 (see Mathematica Code H.1, lines 2 – 7) the eigensystem of the Hamiltonian matrix $H_{m0}[\beta]$ is calculated and then stored in the table $eigm0[\beta]$. The table is indexed by an index running from 1 to $imax+1 = 1001$. This procedure is carried out for each $|M| = 0, 1, 2$.

```
eigm0=Table[Eigensystem[Hm0[\beta]],{\beta, start, end, di}];
26 Export["/home/steffen/working/mathematica/data/Eigensystem0.m",eigm0];
eigm1=Table[Eigensystem[Hm1[\beta]],{\beta, start, end, di}];
28 Export["/home/steffen/working/mathematica/data/Eigensystem1.m",eigm1];
eigm2=Table[Eigensystem[Hm2[\beta]],{\beta, start, end, di}];
30 Export["/home/steffen/working/mathematica/data/Eigensystem2.m",eigm2];
```

The eigensystems of each set of states for $|M| = 0, 1, 2$ (as seen in Figure H.1) are exported in an external file named “Eigensystem0.m”, “Eigensystem1.m” and “Eigensystem2.m”, respectively. Because of the separated diagonalization for different $|M|$ the symmetry properties of the eigenstates for each set of states are lost and hence one should be cautious calculating matrix elements for operators that now may have no meaning anymore, like $\langle \phi_{00}^{num} | d_1 | \phi_{10}^{num} \rangle$.

Mathematica Code H.1 Numerical-Diagonalization.nb

```
(* Parameters (n=dimension of Hamiltonian, di=discretization length)*)
2 n=50;
imax=1000;
4 start=0.;
end=10.;
6 di=(end-start)/imax
0.01
8 (* Define Reduced Matrix Element :*)
RME[J_,j_,k_]:=(-1)^(J) Sqrt[(2J+1)(2j+1)]ThreeJSymbol[{J,0},{k,0},{j,0}]/
Sqrt[2J+1]
10 (* Define Wigner – Eckart Theorem :*)
WET[J_,M_,k_,q_,j_,m_]:=ClebschGordan[{j,m},{k,q},{J,M}]RME[J,j,k]
12 (* Define Dipole Operator (in units of d)*)
d[J_,M_,j_,m_,q_]:=WET[J+M,M,1,q,j+m,m]
14 d0[J_,M_,j_,m_]:=d[J,M,j,m,0]
d0m0=Table[d0[J,0,j,0],{J,0,n},{j,0,n}];
16 d0m1=Table[d0[J,1,j,1],{J,0,n},{j,0,n}];
d0m2=Table[d0[J,2,j,2],{J,0,n},{j,0,n}];
18 (* Define Hamiltonian H=BJ^2-d.E in |J,M> Basis (in units of B with \beta=dE
/B)*)
```

```

En[J_,M_]:= (J+M) (J+M+1)
20 Hm0[\[Beta]_]:= Table [En[J,0] KroneckerDelta[J,j]-\[Beta] d0[J,0,j,0],{J,0,n},{
      j,0,n}]
Hm1[\[Beta]_]:= Table [En[J,1] KroneckerDelta[J,j]-\[Beta] d0[J,1,j,1],{J,0,n},{
      j,0,n}]
22 Hm2[\[Beta]_]:= Table [En[J,2] KroneckerDelta[J,j]-\[Beta] d0[J,2,j,2],{J,0,n},{
      j,0,n}]
(* Eigensystem of the Hamiltonian *)
24 eigm0=Table [Eigensystem [Hm0[\[Beta]]],{\[Beta],start,end,di}];
Export ["/home/steffen/working/mathematica/data/Eigensystem0.m",eigm0];
26 eigm1=Table [Eigensystem [Hm1[\[Beta]]],{\[Beta],start,end,di}];
Export ["/home/steffen/working/mathematica/data/Eigensystem1.m",eigm1];
28 eigm2=Table [Eigensystem [Hm2[\[Beta]]],{\[Beta],start,end,di}];
Export ["/home/steffen/working/mathematica/data/Eigensystem2.m",eigm2];

```

H.2 Energy-Stark-Shift.nb

After importing the eigensystems from the files “Eigensystem0.m”, “Eigensystem1.m” and “Eigensystem2.m” with the same parameters as the tables containing the eigensystems of the Hamiltonian were exported (H.1) (compare Mathematica Code H.1 with Mathematica Code H.2). The built-in functions **Ordering** and **Part** are employed in order to sort each set of eigenstates and eigenenergies in ascending order and to extract the first three eigenstates and eigenenergies. The function `en1m0`, `en2m0`,... and `ev1m0`, `ev2m0`,... are plotted for each $\beta = 0, \dots, 10$ in discrete steps `di`.

Mathematica Code H.2 Energy–Stark–Shift.nb

```

(* Parameters (n=dimension of Hamiltonian , di=discretization length) *)
2 n=50;
imax=1000;
4 start=0.;
end=10.;
6 di=(end-start)/imax
0.01
8 (* Import Eigensystem of H=BJ^2-d.E (for m=0,1,2) *)
eigm0=Import ["/home/steffen/working/mathematica/data/Eigensystem0.m"];
10 eigm1=Import ["/home/steffen/working/mathematica/data/Eigensystem1.m"];
eigm2=Import ["/home/steffen/working/mathematica/data/Eigensystem2.m"];

```



```

12 (*Energies & Eigenvectors of the ground state and the two first excited
    states (with the index irunning form 0 to \[Beta]/di)*)
orderm0[i_]:=Ordering[Part[Part[eigm0,i],1],3]
14 en1m0[i_]:=Part[Part[Part[eigm0,i],1],First[orderm0[i]]]
en2m0[i_]:=Part[Part[Part[eigm0,i],1],Part[orderm0[i],2]]
16 en3m0[i_]:=Part[Part[Part[eigm0,i],1],Part[orderm0[i],3]]
ev1m0[i_]:=Part[Part[Part[eigm0,i],2],First[orderm0[i]]]
18 ev2m0[i_]:=Part[Part[Part[eigm0,i],2],Part[orderm0[i],2]]
ev3m0[i_]:=Part[Part[Part[eigm0,i],2],Part[orderm0[i],3]]
20 orderm1[i_]:=Ordering[Part[Part[eigm1,i],1],3]
en1m1[i_]:=Part[Part[Part[eigm1,i],1],First[orderm1[i]]]
22 en2m1[i_]:=Part[Part[Part[eigm1,i],1],Part[orderm1[i],2]]
en3m1[i_]:=Part[Part[Part[eigm1,i],1],Part[orderm1[i],3]]
24 ev1m1[i_]:=Part[Part[Part[eigm1,i],2],First[orderm1[i]]]
ev2m1[i_]:=Part[Part[Part[eigm1,i],2],Part[orderm1[i],2]]
26 ev3m1[i_]:=Part[Part[Part[eigm1,i],2],Part[orderm1[i],3]]
orderm2[i_]:=Ordering[Part[Part[eigm2,i],1],3]
28 en1m2[i_]:=Part[Part[Part[eigm2,i],1],First[orderm2[i]]]
en2m2[i_]:=Part[Part[Part[eigm2,i],1],Part[orderm2[i],2]]
30 en3m2[i_]:=Part[Part[Part[eigm2,i],1],Part[orderm2[i],3]]
ev1m2[i_]:=Part[Part[Part[eigm2,i],2],First[orderm2[i]]]
32 ev2m2[i_]:=Part[Part[Part[eigm2,i],2],Part[orderm2[i],2]]
ev3m2[i_]:=Part[Part[Part[eigm2,i],2],Part[orderm2[i],3]]
34 (* Calculation
    Plots of energy levels for m=0 and J=0,1,2*)
36 ListLinePlot[{Table[{(i-1)*di+start,en1m0[i]},{i,1,imax+1}],Table[{(i-1)*di+
    start,en2m0[i]},{i,1,imax+1}],Table[{(i-1)*di+start,en3m0[i]},{i,1,imax
    +1}]},PlotRange->Full,PlotStyle->{Directive[Blue,Thickness[.005]],
    Directive[Red,Thickness[.005]],Directive[Purple,Thickness[.005]]},
    FrameStyle->Thickness[.005],FrameLabel->{"x","y"},PlotLabel->"Caption",
    Frame->True,Axes->False]
    (* Plots of energy levels for |m|=0, 1, 2 and J=0,1,2*)
38 ListLinePlot[{Table[{(i-1)*di+start,en1m0[i]},{i,1,imax+1}],Table[{(i-1)*di+
    start,en2m0[i]},{i,1,imax+1}],Table[{(i-1)*di+start,en1m1[i]},{i,1,imax
    +1}],Table[{(i-1)*di+start,en3m0[i]},{i,1,imax+1}],Table[{(i-1)*di+start,
    en2m1[i]},{i,1,imax+1}],Table[{(i-1)*di+start,en1m2[i]},{i,1,imax+1}]},

```

```

PlotRange->Full, PlotStyle->{Directive[Blue, Thickness[.005]], Directive[Red,
, Thickness[.005]], Directive[Darker[Red], Thickness[.005]], Directive[Purple,
, Thickness[.005]], Directive[Darker[Purple], Thickness[.005]], Directive[
Darker[Darker[Purple]], Thickness[.005]]}, FrameStyle-> Thickness[.005],
FrameLabel->{"x", "y"}, PlotLabel->"Caption", Frame->True, Axes->False

```

H.3 Fourier-Transform-Interaction.nb

Calculation of $\epsilon(\mathbf{k})$ directly in real space via the sum given as

$$\epsilon(\mathbf{k}) \approx \sum_{R_x=-100}^{100} \sum_{R_y=-100}^{100} \frac{e^{-i(k_x R_x + k_y R_y)}}{\sqrt{(R_x)^2 + (R_y)^2}^3} \approx \sum_{R_x=-100}^{100} \sum_{R_y=-100}^{100} \frac{\cos(k_x R_x + k_y R_y)}{\sqrt{(R_x)^2 + (R_y)^2}^3} \quad (\text{H.2})$$

The cancellation of all imaginary terms of the exponential function is explicitly verified numerically by comparison of the two sums over `f1[i_,j_,kx_,ky_]` and `f2[i_,j_,kx_,ky_]` in Mathematica Code H.3. The `If`-condition in line 2 and 3 of Mathematica Code H.3 ensures that the origin is omitted. The results of this program are checked against the analytical expression (3.58) as shown in Table 3.2.

Mathematica Code H.3 Fourier-Transform-Interaction.nb

```

(* Defining the summand function, leaving out the origin *)
2 f1[i_,j_,kx_,ky_]:=If[i==0&&j==0,Exp[I(kx i+ky j)]/(i^2+j^2)^(3/2),0]
f2[i_,j_,kx_,ky_]:=If[i==0&&j==0,Cos[kx i+ky j]/(i^2+j^2)^(3/2),0]
4 F1[kx_,ky_]:=Sum[f1[i,j,kx,ky],{i,-100,100},{j,-100,100}];
F2[kx_,ky_]:=Sum[f2[i,j,kx,ky],{i,-100,100},{j,-100,100}];
6 (* Create sums for all kx, in [-3\[Pi],3\[Pi]] and export as CSV table *)
disp1=Table[{kx,F1[kx,0]},{kx,-3\[Pi],3\[Pi],.1}]
8 Export["/user/home/people/steffen/mathematica/data/disp1.dat",disp1,"CSV"]
disp2=Table[{kx,F2[kx,0]},{kx,-3\[Pi],3\[Pi],.1}]
10 Export["/user/home/people/steffen/mathematica/data/disp2.dat",disp2,"CSV"]
(* Create sums for all kx and ky in [-3\[Pi],3\[Pi]] and export as CSV table *)
12 disp13D=Table[{kx,ky,F1[kx,ky]},{kx,-3\[Pi],3\[Pi],.1},{ky,-3\[Pi],3\[Pi],.1}]
Export["/user/home/people/steffen/mathematica/data/disp13D.dat",disp13D,"CSV"]
]
14 disp23D=Table[{kx,ky,F2[kx,ky]},{kx,-3\[Pi],3\[Pi],.1},{ky,-3\[Pi],3\[Pi],.1}]
]

```

```

Export["/user/home/people/steffen/mathematica/data/disp23D.dat", disp23D, "CSV"
]
16 (*2D and 3D plots of the Fourier transform*)
ListPlot[disp1, PlotRange->Full, AxesLabel->{"x", "y"}, PlotLabel->"Caption"]
18 ListPlot[disp2, PlotRange->Full, AxesLabel->{"x", "y"}, PlotLabel->"Caption"]
ListPlot3D[disp13D, PlotRange->Full, ColorFunction->"Aquamarine", AxesLabel->{"x",
"y", "z"}, PlotLabel->"Caption"]
20 ListPlot3D[disp23D, PlotRange->Full, ColorFunction->"Aquamarine", AxesLabel->{"x",
"y", "z"}, PlotLabel->"Caption"]

```

H.4 Dipole-Moments-Coupling-Constants.nb

The calculation of the dipole moments coupling different states of the Stark-shifted spectrum is carried out by the program Mathematica Code H.4. The eigensystem of the operator (H.1) is imported and determined as is stated in Mathematica Code H.2 lines 2 – 33. The dipole operators d_{-1} and d_1 are generated by the functions $dminus1[J_, M_, j_, m_] := d[J, M, j, m, -1]$ and $dplus1[J_, M_, j_, m_] := d[J, M, j, m, 1]$. If one prints out the corresponding matrices, one finds that these operators are antihermitian conjugate to each other and that they possess only diagonal entries and entries on the second off-diagonal. Therefore only scalar products involving the physically coupled eigenvectors are physically correct, for example

$$\begin{array}{ll}
 d0m0 & |\phi_{J0}\rangle \longleftrightarrow |\phi_{J+10}\rangle \\
 d0m1 & |\phi_{J1}\rangle \longleftrightarrow |\phi_{J+11}\rangle \\
 d0m2 & |\phi_{J2}\rangle \longleftrightarrow |\phi_{J+12}\rangle \\
 dminus1m0m1 & |\phi_{J0}\rangle \longleftrightarrow |\phi_{J+11}\rangle \\
 dplus1m0m1 & |\phi_{J+11}\rangle \longleftrightarrow |\phi_{J0}\rangle \\
 dminus1m1m2 & |\phi_{J1}\rangle \longleftrightarrow |\phi_{J+12}\rangle \\
 dplus1m1m2 & |\phi_{J+11}\rangle \longleftrightarrow |\phi_{J2}\rangle
 \end{array}$$

as is also shown in Figure H.2. After generation of the matrices, we calculate the dipole moments and coupling constants (see lines 67 – 77 of Mathematica Code H.4) and finally make the plots that appear in Figure 3.2.

Mathematica Code H.4 Dipole-Moments-Coupling-Constants.nb

```

50 (* Define Reduced Matrix Element : *)
RME[J_, j_, k_] := (-1)^(J) Sqrt[(2J+1)(2j+1)] ThreeJSymbol[{J, 0}, {k, 0}, {j, 0}]/
Sqrt[2J+1]

```

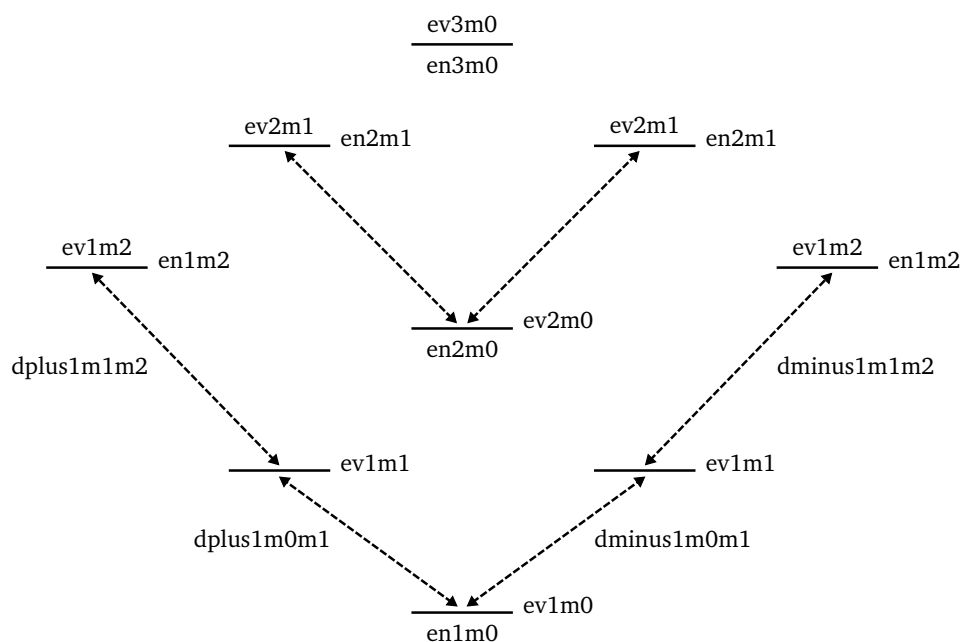


Figure H.2 Coupling of Stark-shifted states and their corresponding dipole moments. The matrices of $d_{\text{minus1}[J,M,j,m]}$ and $d_{\text{plus1}[J,M,j,m]}$ are used to determine the dipole moments of the coupled states indicated by the dashed lines.

```

52 (* Define Wigner - Eckart Theorem :*)
WET[J_,M_,k_,q_,j_,m_] := ClebschGordan[{j,m},{k,q},{J,M}]RME[J,j,k]
54 (* Define Dipole Operator (in units of d) for m=0,1,2*)
d[J_,M_,j_,m_,q_] := WET[J+M,M,1,q,j+m,m]
56 dminus1[J_,M_,j_,m_] := d[J,M,j,m,-1]
d0[J_,M_,j_,m_] := d[J,M,j,m,0]
58 dplus1[J_,M_,j_,m_] := d[J,M,j,m,1]
d0m0 = Table[d0[J,0,j,0],{J,0,n},{j,0,n}];
60 d0m1 = Table[d0[J,1,j,1],{J,0,n},{j,0,n}];
d0m2 = Table[d0[J,2,j,2],{J,0,n},{j,0,n}];
62 dminus1m0m1 = Table[dminus1[J,0,j,1],{J,0,n},{j,0,n}];
dplus1m0m1 = Table[dplus1[J,1,j,0],{J,0,n},{j,0,n}];
64 dminus1m1m2 = Table[dminus1[J,1,j,2],{J,0,n},{j,0,n}];
dplus1m1m2 = Table[dplus1[J,2,j,1],{J,0,n},{j,0,n}];
66 (* Calculate dipole operator matrix elements (for couplings of |0,0> -> |1,0>
and |1,0> -> |2,0> or \[CapitalDelta]m=0*)
d0dd0[i_] := ev1m0[i].d0m0.ev1m0[i]
68 d0pd0[i_] := ev1m0[i].d0m0.ev2m0[i]

```

```

d0pp0[i_]:=ev2m0[i].d0m0.ev2m0[i]
70 d0pd1[i_]:=ev2m0[i].d0m0.ev3m0[i]
d0pp1[i_]:=ev3m0[i].d0m0.ev3m0[i]
72 (* Define coupling constant Subscript[J, z] (in units of d^2/a^3) (for
    couplings of |0,0> -> |1,0> and |1,0> -> |2,0> or m=0) *)
Jz1[i_]:= .25(d0pp0[i]-d0dd0[i])^2
74 Jz2[i_]:= .25(d0pp1[i]-d0pp0[i])^2
(* Define coupling constant Subscript[J, \[UpTee]] (in units of d^2/a^3) (for
    couplings of |0,0> -> |1,0> and |1,0> -> |2,0> or m=0) *)
76 Jp1[i_]:= .5(d0pd0[i])^2
Jp2[i_]:= .5(d0pd1[i])^2
78 (* Angle of "Model Circle" (for couplings of |0,0> -> |1,0> and |1,0> -> |2,0>
    or m=0) *)
\[Theta]1[i_]:=ArcTan[Jp1[i]/Jz1[i]]/\[Pi]*180
80 \[Theta]2[i_]:=ArcTan[Jp2[i]/Jz2[i]]/\[Pi]*180
(* Plots of dipole operator matrix elements Subscript[d, 0]^\[UpArrow]\[
    UpArrow] Subscript[d, 0]^\[DownArrow]\[DownArrow] Subscript[d, 0]^\[
    UpArrow]\[DownArrow] ( \[CapitalDelta]M=0 transitions) for J=0,1,2 *)
82 ListLinePlot[{Table[{(i-1)*di+start, d0dd0[i]}, {i, 1, imax+1}], Table[{(i-1)*di+
    start, d0pd0[i]}, {i, 1, imax+1}], Table[{(i-1)*di+start, d0pp0[i]}, {i, 1, imax
    +1}], PlotRange->Full, PlotStyle->{Directive[Blue, Thickness[.005]],
    Directive[Red, Thickness[.005]], Directive[Purple, Thickness[.005]]},
    FrameStyle-> Thickness[.005], FrameLabel->{"x", "y"}, PlotLabel->"Caption",
    Frame->True, Axes->False]
ListLinePlot[{Table[{(i-1)*di+start, d0dd0[i]}, {i, 1, imax+1}], Table[{(i-1)*di+
    start, Abs[d0pd0[i]}, {i, 1, imax+1}], Table[{(i-1)*di+start, d0pp0[i]}, {i, 1,
    imax+1}], PlotRange->Full, PlotStyle->{Directive[Blue, Thickness[.005]],
    Directive[Red, Thickness[.005]], Directive[Purple, Thickness[.005]]},
    FrameStyle-> Thickness[.005], FrameLabel->{"x", "y"}, PlotLabel->"Caption",
    Frame->True, Axes->False]
84 ListLinePlot[{Table[{(i-1)*di+start, d0pd1[i]}, {i, 1, imax+1}], Table[{(i-1)*di+
    start, d0pp1[i]}, {i, 1, imax+1}], PlotRange->Full, PlotStyle->{Directive[Blue
    , Thickness[.005]], Directive[Red, Thickness[.005]]}, FrameStyle-> Thickness
    [.005], FrameLabel->{"x", "y"}, PlotLabel->"Caption", Frame->True, Axes->False
    ]
ListLinePlot[{Table[{(i-1)*di+start, Abs[d0pd1[i]}, {i, 1, imax+1}], Table[{(i-1)

```

```

*di+start , d0pp1[i]},{i,1,imax+1}},PlotRange->Full , PlotStyle->{Directive [
Blue , Thickness [.005]] , Directive [Red , Thickness [.005]]} , FrameStyle->
Thickness [.005] , FrameLabel->{"x" , "y"} , PlotLabel->"Caption" , Frame->True ,
Axes->False]
86 Plots of coupling constants  $\text{Subscript}[J, z]$  and  $\text{Subscript}[J, \text{\[UpTee]}$  for
|0,0> -> |1,0>
ListLinePlot[{Table[{{(i-1)*di+start , Jz1[i]},{i,1,imax+1}},Table[{{(i-1)*di+
start , Jp1[i]},{i,1,imax+1}}],PlotRange->Full , PlotStyle->{Directive [Blue ,
Thickness [.005]] , Directive [Red , Thickness [.005]]} , FrameStyle-> Thickness
[.005] , FrameLabel->{"x" , "y"} , PlotLabel->"Caption" , Frame->True , Axes->False
]
88 Plots of coupling constants  $\text{Subscript}[J, z]$  and  $\text{Subscript}[J, \text{\[UpTee]}$  for
|1,0> -> |2,0>
ListLinePlot[{Table[{{(i-1)*di+start , Jz2[i]},{i,1,imax+1}},Table[{{(i-1)*di+
start , Jp2[i]},{i,1,imax+1}}],PlotRange->Full , PlotStyle->{Directive [Blue ,
Thickness [.005]] , Directive [Red , Thickness [.005]]} , FrameStyle-> Thickness
[.005] , FrameLabel->{"x" , "y"} , PlotLabel->"Caption" , Frame->True , Axes->False
]
90 Plots of angle  $\text{\[Theta]}$  for couplings of |0,0> -> |1,0>
ListLinePlot[Table[{{(i-1)*di+start , \[Theta]1[i]},{i,1,imax+1}},PlotRange->
Full , PlotStyle->Directive [Blue , Thickness [.005]] , FrameStyle-> Thickness
[.005] , FrameLabel->{"x" , "y"} , PlotLabel->"Caption" , Frame->True , Axes->False
]
92 Plots of angle  $\text{\[Theta]}$  for couplings of |1,0> -> |2,0>
ListLinePlot[Table[{{(i-1)*di+start , \[Theta]2[i]},{i,1,imax+1}},PlotRange->
Full , PlotStyle->Directive [Blue , Thickness [.005]] , FrameStyle-> Thickness
[.005] , FrameLabel->{"x" , "y"} , PlotLabel->"Caption" , Frame->True , Axes->False
]
94 Comparison between numerical dipole matrix elements and perturbative
solutions (for |0,0> -> |1,0>)
ListLinePlot[{Table[{{(i-1)*di+start , d0dd0[i]},{i,1,10+1}},
96 Table[{{(i-1)*di+start , 1./3 i},{i,1,11}},Table[{{(i-1)*di+start , d0pd0[i]},{i
,1,10+1}},
Table[{{(i-1)*di+start , 1./Sqrt[3]i},{i,1,11}},
98 Table[{{(i-1)*di+start , d0pp0[i]},{i,1,10+1}},
Table[{{(i-1)*di+start , -1./5 i},{i,1,11}}],PlotRange->Full , PlotStyle->{

```

```

Directive[Blue, Thickness[.005]], Directive[Darker[Blue], Thickness[.005]],
Directive[Red, Thickness[.005]], Directive[Darker[Red], Thickness[.005]],
Directive[Purple, Thickness[.005]], Directive[Darker[Purple], Thickness
[.005]], FrameStyle-> Thickness[.005], FrameLabel->{"x", "y"}, PlotLabel->"
Caption", Frame->True, Axes->False]

```

H.5 t-J-Model-Roots.nb

In order to determine the values of the parameter α_1 , α_2 and β , we have to solve the three conditions $\tilde{D}^{ee} = 0$, $\tilde{D}^{gg} = 0$ and $\tilde{D}^{egge} = -\tilde{D}^{eg}$ which is done in Mathematica Code H.5. First the four composite dipole moments are defined (lines 78–81) and the roots of the equations $\tilde{D}^{ee} = 0$ and $\tilde{D}^{gg} = 0$ are calculated with the help of the build-in function `FindRoot` that determines the root of a function. After fixing the two parameters α_1 and α_2 , the parameter β is given by the intersection point of the two curves plotted in line 104 and 105.

Mathematica Code H.5 t-J-Model-Roots.nb

```

(* Define matrix elements of Subscript[V, dd]: Subscript[D, ee], Subscript[D,
  gg], Subscript[D, eg], Subscript[D, eg, ge] *)
78 Dee[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := \[Alpha]2^4 (d0pp[i]^2 + \[Alpha]2^2
  (1 - \[Alpha]2^2)(2d0pp[i]d022[i] - (dplus12p[i])^2) + (1 - \[Alpha]2^2)^2 (d022[
  i])^2)
Deg[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := \[Alpha]1^2 \[Alpha]2^2 (d0pp[i]d0dd[i
  ]) + \[Alpha]1^2 (1 - \[Alpha]2^2)(d0dd[i]d022[i]) + \[Alpha]2^2 (1 - \[Alpha
  ]1^2)(d0pp[i]d011[i]) + (1 - \[Alpha]2^2)(1 - \[Alpha]1^2)(d011[i]d022[i])
80 Dgg[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := \[Alpha]1^4 (d0dd[i])^2 + \[Alpha]1^2
  (1 - \[Alpha]1^2)(2d0dd[i]d011[i] - (dplus11d[i])^2) + (1 - \[Alpha]1^2)^2 (d011[
  i])^2
Degge[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := \[Alpha]1^2 \[Alpha]2^2 (d0pd[i])
  ^2 + (1 - \[Alpha]2^2)(1 - \[Alpha]1^2) (d012[i])^2
82 (* Define coupling constant Subscript[J, l], Subscript[J, z], Subscript[J, \[
  UpTee]] (in units of d^2/a^3) and Subscript[h, z] *)
hz[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := .5(Dee[i, \[Phi], \[Alpha]1, \[Alpha]2] -
  Dgg[i, \[Phi], \[Alpha]1, \[Alpha]2])
84 Jp[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := 2.Degge[i, \[Phi], \[Alpha]1, \[Alpha]2]
Jz[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := Dee[i, \[Phi], \[Alpha]1, \[Alpha]2] - 2Deg[
  i, \[Phi], \[Alpha]1, \[Alpha]2] + Dgg[i, \[Phi], \[Alpha]1, \[Alpha]2]

```

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86 J1[i_, \[Phi]_, \[Alpha]1_, \[Alpha]2_] := Dee[i, \[Phi], \[Alpha]1, \[Alpha]2] + 2Deg[
    i, \[Phi], \[Alpha]1, \[Alpha]2] + Dgg[i, \[Phi], \[Alpha]1, \[Alpha]2]
(* Condition Subscript[D, ee]=0 -> Subscript[\[Alpha], 2](\[Beta]) *)
88 root\[Alpha]21 = Table[\[Alpha]2/.FindRoot[Dee[i, 0, 0, \[Alpha]2], {\[Alpha]2, 1./
    Sqrt[2]}], {i, 1, imax+1}];
root\[Alpha]22 = Table[\[Alpha]2/.FindRoot[Dee[i, 0, 0, \[Alpha]2], {\[Alpha]
    ]2, .85}], {i, 1, imax+1}];
90 root\[Alpha]23 = Table[\[Alpha]2/.FindRoot[Dee[i, 0, 0, \[Alpha]2], {\[Alpha]
    ]2, 1.}], {i, 1, imax+1}];
ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]21, i]}, {i, 1, imax+1}],
    PlotRange -> {1.05, .65}, PlotStyle -> Directive[Blue, Thickness[.005]],
    FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption",
    Frame -> True, Axes -> False]
92 ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]22, i]}, {i, 1, imax+1}],
    PlotRange -> Full, PlotStyle -> Directive[Blue, Thickness[.005]], FrameStyle ->
    Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption", Frame -> True,
    Axes -> False]
ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]23, i]}, {i, 1, imax+1}],
    PlotRange -> Full, PlotStyle -> Directive[Blue, Thickness[.005]], FrameStyle ->
    Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption", Frame -> True,
    Axes -> False]
94 ListLinePlot[Table[{Tan[2ArcCos[Part[root\[Alpha]22, i]]], (i-1)*di+start}, {i
    ], 1, imax+1}], PlotRange -> {{0, .5}, {0, 6}}, PlotStyle -> Directive[Blue, Thickness
    [.005]], FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "
    Caption", Frame -> True, Axes -> False]
(* Condition Subscript[D, gg]=0 -> Subscript[\[Alpha], 1](\[Beta]) *)
96 root\[Alpha]11 = Table[\[Alpha]1/.FindRoot[Dgg[i, 0, \[Alpha]1, 0], {\[Alpha]1, 1./
    Sqrt[2]}], {i, 1, imax+1}];
root\[Alpha]12 = Table[\[Alpha]1/.FindRoot[Dgg[i, 0, \[Alpha]1, 0], {\[Alpha]
    ]1, .85}], {i, 1, imax+1}];
98 root\[Alpha]13 = Table[\[Alpha]1/.FindRoot[Dgg[i, 0, \[Alpha]1, 0], {\[Alpha]
    ]1, 1.}], {i, 1, imax+1}];
ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]11, i]}, {i, 1, imax+1}],
    PlotRange -> Full, PlotStyle -> Directive[Blue, Thickness[.005]], FrameStyle ->
    Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption", Frame -> True,
    Axes -> False]

```



```

100 ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]12, i]}, {i, 1, imax+1}],
  PlotRange -> {1.05, .65}, PlotStyle -> Directive[Blue, Thickness[.005]],
  FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption",
  Frame -> True, Axes -> False]
ListLinePlot[Table[{(i-1)*di+start, Part[root\[Alpha]13, i]}, {i, 1, imax+1}],
  PlotRange -> {1.05, .65}, PlotStyle -> Directive[Blue, Thickness[.005]],
  FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption",
  Frame -> True, Axes -> False]
102 ListLinePlot[Table[{Tan[2ArcCos[Part[root\[Alpha]12, i]]], (i-1)*di+start}, {i,
  1, imax+1}], PlotRange -> {{0, 4}, {0, 6}}, PlotStyle -> Directive[Blue, Thickness
  [.005]], FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "
  Caption", Frame -> True, Axes -> False]
(* Condition Subscript[D, eg, ge] = -Subscript[D, eg] -> \[Beta] *)
104 ListLinePlot[Table[{(i-1)*di+start, Degge[i, 0, Part[root\[Alpha]12, i], Part[root
  \[Alpha]22, i]] + Deg[i, 0, Part[root\[Alpha]12, i], Part[root\[Alpha]22, i]}], {i,
  1, imax+1}], PlotRange -> Full, PlotStyle -> Directive[Red, Thickness[.005]],
  FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "Caption",
  Frame -> True, Axes -> False]
ListLinePlot[{Table[{(i-1)*di+start, Degge[i, 0, Part[root\[Alpha]12, i], Part[
  root\[Alpha]23, i]}], {i, 1, imax+1}], Table[{(i-1)*di+start, -Deg[i, 0, Part[
  root\[Alpha]12, i], Part[root\[Alpha]23, i]}], {i, 1, imax+1}], PlotRange -> Full
, PlotStyle -> {Directive[Blue, Thickness[.005]], Directive[Red, Thickness
  [.005]]}, FrameStyle -> Thickness[.005], FrameLabel -> {"x", "y"}, PlotLabel -> "
  Caption", Frame -> True, Axes -> False]

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Bibliography

- [1] J. G. Bednorz and K. A. Müller. Possible high t_c superconductivity in the $Ba_x-La_{5-x}-Cu_{5(3-y)}$ system. *Zeitschrift für Physik B Condensed Matter*, 64(2):189–193, Jun 1986.
- [2] J. Bardeen, L. N. Cooper, and J. R. Schrieffer. Theory of superconductivity. *Phys. Rev.*, 108(5):1175–1204, Dec 1957.
- [3] X. G. Wen, Frank Wilczek, and A. Zee. Chiral spin states and superconductivity. *Phys. Rev. B*, 39(16):11413–11423, Jun 1989.
- [4] A. Micheli, G. Pupillo, H. P. Büchler, and P. Zoller. Cold polar molecules in two-dimensional traps: Tailoring interactions with external fields for novel quantum phases. *Physical Review A (Atomic, Molecular, and Optical Physics)*, 76(4):043604, 2007.
- [5] Klaus Schulten. *Notes on Quantum Mechanics*. 2000.
- [6] Howard Georgi. *Lie Algebras in Particle Physics*. World Scientific, second edition, 1999.
- [7] J.F. Cornwell. *Group Theory in Physics*. Academic Press, 1997.
- [8] Wolfgang P. Schleich. *Quantum Optics in Phase Space*. WILEY-VCH, 2001.
- [9] Claude Cohen-Tannoudji, Jacques Dupont-Roc, and Gilbert Grynberg. *Photons And Atoms – Introduction to Quantum Electrodynamics*. John Wiley & Sons, 1997.
- [10] Pierre Meystre and Murray Sargent III. *Elements of Quantum Optics*. Springer, 2007.
- [11] Marlan O. Scully and M. Suhail Zubairy. *Quantum Optics*. Cambridge University Press, 1997.
- [12] Ralph Skomski. *Simple Models of Magnetism*. Oxford Graduate Texts. Oxford University Press, 2008.
- [13] U. Schollwöck, J. Richter, D.J.J. Farnell, and R.F. Bishop. *Quantum Magnetism*. Lecture Notes in Physics. Springer, 2008.
- [14] J. van Kranendonk and J. H. van Vleck. Spin waves. *Rev. Mod. Phys.*, 30(1):1–23, Jan 1958.
- [15] Ryogo Kubo. The spin-wave theory of antiferromagnetics. *Physical Review*, 87(4):568–580, 1952.
- [16] Lynn Bonsall and A. A. Maradudin. Some static and dynamical properties of a two-dimensional wigner crystal. *Phys. Rev. B*, 15(4):1959–1973, Feb 1977.
- [17] Jozef Spalek. $t-j$ model then and now: A personal perspective from the pioneering times. *Acta Physica Polonica A*, 111:409, 2007.

- [18] Jürgen Fuchs and Christopf Schweigert. *Symmetries, Lie Algebras & Representations*. Cambridge Monographs on Mathematical Physics. Cambridge University Press, 1997.
- [19] H. P. Büchler, E. Demler, M. Lukin, A. Micheli, N. Prokof'ev, G. Pupillo, and P. Zoller. Strongly correlated 2d quantum phases with cold polar molecules: Controlling the shape of the interaction potential. *Physical Review Letters*, 98(6):060404, 2007.
- [20] Immanuel Bloch, Jean Dalibard, and Wilhelm Zwerger. Many-body physics with ultracold gases. *Rev. Mod. Phys.*, 80(3):885–964, Jul 2008.
- [21] H. P. Büchler, A. Micheli, and P. Zoller. Three-body interactions with cold polar molecules. *Nat Phys*, 3(10):726–731, Oct 2007.
- [22] A. Micheli, G. K. Brennen, and P. Zoller. A toolbox for lattice-spin models with polar molecules. *Nat Phys*, 2(5):341–347, May 2006.
- [23] Maciej Lewenstein. Atomic and molecular physics: Polar molecules in topological order. *Nat Phys*, 2(5):309–310, May 2006.
- [24] K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Pe'er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye. A high phase-space-density gas of polar molecules. *Science*, 322(5899):231–235, 2008.
- [25] Ryogo Kubo. The spin-wave theory as a variational method and its application to antiferromagnetism. *Rev. Mod. Phys.*, 25(1):344–351, Jan 1953.
- [26] Andreas Lüscher and Andreas M. Läuchli. Exact diagonalization study of the antiferromagnetic spin- $1/2$ heisenberg model on the square lattice in a magnetic field. *Phys. Rev. B*, 79(19):195102, May 2009.
- [27] Masanori Kohno and Minoru Takahashi. Magnetization process of the spin- $1/2$ xxz models on square and cubic lattices. *Phys. Rev. B*, 56(6):3212–3217, Aug 1997.
- [28] Alessandro Cuccoli, Valerio Tognetti, and Ruggero Vaia. Two-dimensional xxz model on a square lattice: A monte carlo simulation. *Phys. Rev. B*, 52(14):10221–10231, Oct 1995.
- [29] H.-Q. Lin, J. S. Flynn, and D. D. Betts. Exact diagonalization and quantum monte carlo study of the spin- $1/2$ xxz model on the square lattice. *Phys. Rev. B*, 64(21):214411, Nov 2001.
- [30] Alexander Reischl, Erwin Müller-Hartmann, and Götz S. Uhrig. Systematic mapping of the hubbard model to the generalized $t - j$ model. *Phys. Rev. B*, 70(24):245124, Dec 2004.
- [31] F. Hébert, G. G. Batrouni, R. T. Scalettar, G. Schmid, M. Troyer, and A. Dorneich. Quantum phase transitions in the two-dimensional hardcore boson model. *Phys. Rev. B*, 65(1):014513, Dec 2001.
- [32] Guido Schmid, Synge Todo, Matthias Troyer, and Ansgar Dorneich. Finite-temperature phase diagram of hard-core bosons in two dimensions. *Phys. Rev. Lett.*, 88(16):167208, Apr 2002.
- [33] R. G. Melko, A. Paramekanti, A. A. Burkov, A. Vishwanath, D. N. Sheng, and L. Balents. Supersolid order from disorder: Hard-core bosons on the triangular lattice. *Phys. Rev. Lett.*, 95(12):127207, Sep 2005.

-
- [34] B. Capogrosso-Sansone, C. Trefzger, M. Lewenstein, P. Zoller, and G. Pupillo. Quantum phases of cold polar molecules in 2d optical lattices. *arXiv:0906.2009v1*, Jun 2009.
- [35] Francesco Iachello. *Lie Algebras & Applications*. Lecture Notes in Physics. Springer, 2006.
- [36] Wendell Hill and Chi Lee. *Light-Matter Interaction*. WILEY-VCH, 2007.
- [37] Shlomo Sternberg. *Group Theory and Physics*. Cambridge University Press, 1995.
- [38] D.F. Walls and G.J. Milburn. *Quantum Optics*. Springer, second edition, 1995.
- [39] Howard Carmichael. *An Open System Approach to Quantum Optics*. Springer, 1993.
- [40] Jean-Louis Basdevant and Jean Dalibard. *Quantum Mechanics*. Springer, 2002.
- [41] Michel Le Bellac. *Quantum Physics*. Cambridge University Press, 2006.
- [42] J.C. Garrison and R.Y. Chiao. *Quantum Optics*. Oxford Graduate Texts. Oxford University Press, 2008.
- [43] Hartmut Haug and Stephan Koch. *Quantum Theory of the Optical Electronic Properties of Semiconductors*. World Scientific, 2004.
- [44] Henrik Bruus and Karsten Flensberg. *Many Body Quantum Field Theory*. Oxford Graduate Texts. Oxford University Press, 2004.
- [45] Xiao-Gang Wen. *Quantum Field Theory of Many Body Systems*. Oxford Graduate Texts. Oxford University Press, 2004.
- [46] Alexander Altland and Ben Simons. *Condensed Matter Field Theory*. Cambridge University Press, 2006.
- [47] Leonard Sander. *Advanced Condensed Matter Physics*. Cambridge University Press, 2009.
- [48] P.M. Chaikin and T.C. Lubensky. *Principles of Condensed Matter Physics*. Cambridge University Press, 1995.
- [49] Subir Sachdev. *Quantum Phase Transitions*. Cambridge University Press, 2001.

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