The Equation of Motion Method for Spin Systems with Multipolar Hamiltonians

Peter Balla

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1 Introduction

In the theory of magnetic insulators several models of magnetic ordering have been developed. The most well known ones are the (isotropic) Heisenberg ferro- and antiferromagnets. The usual ways to calculate the dispersion relations (and the form of approximate excitations) of these models are linearized bosonization techniques (e.g. Schwinger or Holstein-Primakoff bosons). But if we allow complex interactions, or even the seemingly innocent on-site anisotropies, or any term containing higher order polynomials of the spin components, these techniques fail. But these terms –if allowed by symmetry, and for spin lengths larger than one half– are present in the real world materials. To handle these complex models correctly the so-called multiboson theory has been developed, and used successfully to describe these materials. But this method –although very powerful– is very complicated, and to *understand* what is really happening to the spins is very hard to extract from them. Therefore we seek an alternative way of solving these complex models, perhaps one, that –besides being correct– gives a way of imagining the motion of the spins pictorially.

In this work we develop a method based on the quasiclassical approximation of the quantum mechanical Heisenberg equations of motion of complex spin systems. The idea came from the Bloch equations of electron spin resonance (ESR), and the already existing quasiclassical approximation of simple Heisenberg models. What we do in this thesis is the following: we write down the equations of motion for the quantum mechanical variables, and look at them as the classical equations of motion of the expectation values of the quantum mechanical variables. We linearize these equations based on physical arguments, and compute the physically interesting quantities (usually energies and susceptibilities). This picture, which is a powerful way of looking at the motion of the spin in an ESR problem gives us the opportunity to imagine the much more complex excitations in these complex materials. Another advantage is, that we have an alternative way of solving these problems.

The structure of this thesis is the following: we always start with simpler models, and step to the harder ones gradually. Our main goal is to write down the equations for complicated (multipolar) lattice problems, and solve them in a general form. So we start with the simplest on-site problem, the classical Bloch equations. After that we solve the two-spin problem of antiferromagnetic resonance. Next we introduce the so-called multipoles to the one-spin problem, which are just the building blocks of multipolar lattice problems. After that we put the spins on a lattice, and give the solutions of the quasiclassical Heisenberg magnets. And at last we write down, linearize and solve the equations of motion of the very general multipolar Hamiltonian.

We try to illustrate our method with problems that are known to the literature, in order to check the validity of our calculations. The only –but very painful– deficiency is in this respect, that we did not illustrate the general solution of the multipolar lattice model with a worked out example.

2 The Equation of Motion Method

2.1 Single Spin in a Magnetic Field: ESR and Bloch Equations

2.1.1 Introduction

We devote this subsection to the quasiclassical approximation of the Heisenberg equation of motion (EOM) of a *single spin* placed in a magnetic field, its excitations (eigenenergies and eigenoscillations) and its dynamical susceptibility. Actually this is the subject of the well known mastery of electron spin resonance (ESR, or EPR for electron paramagnetic resonance for chemists), or nuclear magnetic resonance (NMR, or MRI for magnetic resonance imaging for physicians). Although these results achieved by the equation of motion (especially for small spins) could be calculated much more precisely on the back of an envelope with simple quantum mechanics, or the use of the Schwinger bosonization technique (c.f. Chapter 3.9. of [26]) and with the use of Kubo's formula, the advantage of our approach is that it gives a simple physical picture of what is happening to the physical system. Another advantage of the quasiclassical description is that its generalization to much more complicated models conserves some of this pictorial view (and bosonization techniques become much more complicated for complicated Hamiltonians, and simple minded quantum mechanics either does not work or requires a supercomputer).

Our model system in this subsection will be a single spin of any length, placed in a static field, and excited by a harmonic, perturbatively weak field. We calculate the excitation energies, eigenoscillations and some typical forced oscillations. This is a warm up subsection of a very well known field, and it serves as an introduction to our notational system and calculational tools. As far as I know, this notational system is new (although it surely is not a big thing), and is very handy to generalize the concept of the quasiclassical approach of the quantum mechanical equation of motion to the more complicated problems of Hamiltonians containing multipolar operators (e.g. anisotropy, quadrupolar interactions, etc.). Another advantage is of this notational system, that the approximation used to solve the EOM are described in a controlled manner, and the method is easily implemented in an algebraic manipulation software (I used *Mathematica*). And –of course– it gives the same results as the textbook techniques.

Of course our exposition of the subject of ESR must be very sporadic, the intrigued reader should consult the very broad literature, of which we mention a few textbooks. The Holy Bible of magnetic resonance is the monograph by Slichter [27] (actually this book was much detailed for my purposes). A very brief introduction of ESR is contained in Chapter 3.2. of [28], and a whole (Chapter 13.) chapter of the classical textbook by Kittel [14] is devoted to the subject of magnetic resonance. What I found really useful was a set of lecture notes by Arovas [2], especially its Chapter 3. on linear response theory (the description of ESR follows that of [14], but is much more detailed). These notes can be found on the webpage [1]: http://physics.ucsd.edu/students/courses/winter2010/physics211b/lectures.html. The ones interested in the subject of angular momentum in quantum mechanics should consult with Chapter 3. of [26], or the books [3] and [4].

A comment about units: we rarely use real physical units (except in the rare cases

when we compare our results to existing ones or measurements), so \hbar is usually set to 1 (energy is measured in angular frequency units, and the words energy/frequency are used interchangeably), and the Bohr magneton, gyromagnetic ratios, etc. are absorbed into the definition of the generalized magnetic fields.

The structure of this subsection is the following: we start by stating the problem, derive the EOM, calculate the eigenfrequencies and -oscillations, the dynamical susceptibility matrix, and at the end we illustrate the method on a concrete example and a comparison with the literature. A discussion closes this subsection.

2.1.2 Hamiltonian, Introduction to the Equation of Motion

In order to develop a concise and consistent notational system we rederive the Bloch equations based on quantum mechanics here. We set $\hbar = 1$, and consider a spin of length S in a magnetic field $\underline{B} = (B^x, B^y, B^z)^T$ with Hamiltonian:

$$H^{h} = -g\mu_{B}\underline{B}^{T} \cdot \underline{S} = \underline{h}^{T} \cdot \underline{S} = \sum_{\alpha} h^{\alpha}S^{\alpha}, \qquad (1)$$

where S^{α} , $\alpha = x, y, z$ are the dimensionless spin operators, and <u>h</u> is the magnetic field containing all the necessary prefactors. Note that we do not use the Einstein summation convention, nor will we pull indices up or down. The spin operators are defined by their commutators, i.e. by the structure constants of the Lie-algebra $\mathfrak{su}(2)^1$:

$$\left[S^{\alpha}, S^{\beta}\right] = i \sum_{\gamma} \varepsilon_{\alpha\beta}{}^{\gamma} S^{\gamma}, \tag{2}$$

here we have used γ as an upper index, which is clearly unimportant yet, but this convention will be very helpful later, and $\varepsilon_{\alpha\beta}^{\gamma}$ is the three dimensional Levi-Civita symbol. The time evolution of a single spin component is governed by the Heisenberg equation of motion:

$$\dot{S}^{\zeta} = i \left[H^h, S^{\zeta} \right] = i \sum_{\alpha} h^{\alpha} \left[S^{\alpha}, S^{\zeta} \right] = -\sum_{\alpha, \gamma} \varepsilon_{\alpha \zeta}{}^{\gamma} h^{\alpha} S^{\gamma}.$$
(3)

Although this is a very simple system of equations, it has some very important structure. The cross product of two three-dimensional vectors \underline{a} , \underline{b} reads as:

$$\underline{c} = \underline{a} \times \underline{b},$$

or with components:

$$c^{\gamma} = \sum_{\alpha,\beta} \varepsilon_{\alpha\beta}^{\quad \gamma} a^{\alpha} b^{\beta}.$$

It is obvious, that this cross product can always be viewed as an antisymmetric linear operator $(\bigstar a)^2$ acting on <u>b</u>:

$$\underline{c} = \underline{a} \times \underline{b} = \underline{(\bigstar a)} \cdot \underline{b},$$

¹We will devote a small subsection to the properties of this algebra later.

²About the notation: we are speaking about Hodge-duality in 3D-space, i.e. $(\bigstar a) = \bigstar a$, and on the right-hand-side \bigstar is the usual Hodge operator. A little bit later we will generalize this concept further.

or componentwise:

$$c^{\gamma} = \sum_{\alpha,\beta} \varepsilon_{\alpha\beta}{}^{\gamma} a^{\alpha} b^{\beta} = \sum_{\beta} \underbrace{\left(\sum_{\alpha} \varepsilon_{\alpha\beta}{}^{\gamma} a^{\alpha}\right)}_{-(\bigstar a)_{\gamma\beta}} b^{\beta} = -\sum_{\beta} a_{\gamma\beta} b^{\beta},$$

from which the components of $(\bigstar a)$ can be read off, namely:

$$(\bigstar a)_{\beta\gamma} = \sum_{\alpha} \varepsilon_{\alpha\beta}{}^{\gamma} a^{\alpha}.$$

In matrix form the associated operator reads as:

$$\begin{pmatrix} c^x \\ c^y \\ c^z \end{pmatrix} = \begin{pmatrix} 0 & a^z & -a^y \\ -a^z & 0 & a^x \\ a^y & -a^x & 0 \end{pmatrix} \cdot \begin{pmatrix} b^x \\ b^y \\ b^z \end{pmatrix}.$$

With this notation at hand we can rewrite Eq. (3) as:

$$\dot{S}^{\zeta} = -\sum_{\gamma} \left(\sum_{\alpha} \varepsilon_{\alpha\zeta}{}^{\gamma} h^{\alpha} \right) S^{\gamma} = -\sum_{\gamma} \left((\bigstar h)_{\zeta\gamma} \right) S^{\gamma}, \tag{4}$$

$$\underline{\dot{S}} = -\underline{(\bigstar h)} \cdot \underline{S} = -\underline{h} \times \underline{S} = \underline{S} \times \underline{h}.$$
(5)

Hereafter we will take this set of equations as the quasiclassical approximation of the quantum mechanical problem, and try to solve it somehow. The solutions of these classical ordinary differential equations will be interpreted as the quantum mechanical expectation values of the corresponding observables, and we will dub this procedure as the equation of motion method, briefly the EOM.

As a concrete example we will show how to use the EOM to interpret the results of electron spin resonance (ESR) experiments. In the experiment the magnetic moment is placed in a large, constant field (which aligns it in the field direction) and a perturbatively weak oscillating field is applied perpendicularly to the DC-field, and the resonant absorption is measured³. In order to describe this situation we divide the external field into a large, static (i.e. time-independent) (\underline{h}^0) part, and a small oscillating $\underline{\delta h}(t) = \underline{\delta h}^{\omega} e^{-i\omega t}$ part, so the field reads as:

$$\underline{h}(t) = \underline{h}^{0} + \underline{\delta h}(t) = \underline{h}^{0} + \underline{\delta h}^{\omega} e^{-i\omega t},$$

where $\underline{\delta h}^{\omega}$ is a vector of complex amplitudes describing the polarization of the oscillating field (the physical field vector is $\Re \left(\underline{\delta h}^{\omega} e^{-i\omega t} \right)$). If only the static field was present, the spin would align in its direction, let us denote this –static– ground state of the spin by \underline{S}^{0} .

 $^{^{3}}$ In real experiments the sample is placed in a microwave resonator, and the static field is varied until a resonance is detected.

As a response to the excitation by the oscillating field a small time dependent variation of the spin results: $\underline{\delta S}(t) = \underline{\delta S}^{\omega} e^{-i\omega t}$, so the total spin becomes:

$$\underline{S}(t) = \underline{S}^0 + \underline{\delta}\underline{S}(t) = \underline{S}^0 + \underline{\delta}\underline{S}^{\omega}e^{-i\omega t},$$

where $\underline{\delta S}^{\omega}$ is a vector of complex amplitudes again, describing the polarization of the response (the physical spin vector is $\Re(\underline{\delta S}^{\omega}e^{-i\omega t})$). Hereafter we omit the time arguments, and remember, that the vectors with 0 upper indices denote static (ground state) quantities. With this notation Eq. (5) becomes:

$$\left(\underline{S}^{0} + \underline{\delta}\underline{S}\right) = \left(\underline{S}^{0} + \underline{\delta}\underline{S}\right) \times \left(\underline{h}^{0} + \underline{\delta}\underline{h}\right).$$
(6)

Since in the ground state (without the oscillating field) there are no fluctuations⁴ we can write:

$$\underline{\underline{S}}^{0} = \underline{\underline{S}}^{0} \times \underline{\underline{h}}^{0} = 0, \tag{7}$$

and substituting this into Eq. (6) we get:

$$\underline{\dot{\delta S}} = \underline{S}^0 \times \underline{\delta h} + \underline{\delta S} \times \underline{h}^0 + \underline{\delta S} \times \underline{\delta h}.$$
(8)

These equations are exact so far, and contain no dissipation terms. The last term in Eq. (8) is a product of two small terms: the perturbatively weak oscillating field and the small spin response to it, so this double- δ term can safely be neglected. Since in real experiments the spin always interacts with its surroundings, we have to somehow account for the dissipation of its energy to the environment. We will do this by using some phenomenological time-constants, the so-called relaxation rates. Let us suppose that there are no external oscillating fields present, and we tilt the spin by a little, then it is natural to expect that it will relax to its ground state with a velocity proportional to its deviation, i.e.: $\delta \dot{S}^{\alpha} = -\delta S^{\alpha}/T^{\alpha}$, where the T^{α} -s are the phenomenological relaxation time constants. We can put these constants in a matrix $\underline{T} = \text{diag}\{T^x, T^y, T^z\}$, or equivalently we can use the inverse lifetimes: $\underline{\Gamma} = \underline{T}^{-1} = \text{diag}\{1/T^x, 1/T^y, 1/T^z\} = \text{diag}\{\gamma^x, \gamma^y, \gamma^z\}$. These constants depend on the environment of the moments, and its interactions with it, so their calculation from first principles is a really hard task, we do not even try to do it. We use them as phenomenological constants instead, and set their values to fit the measurements. Putting all these together we arrive at (the somewhat modified form of) the celebrated *Bloch equations*:

$$\underline{\delta S} = \underline{S}^0 \times \underline{\delta h} + \underline{\delta S} \times \underline{h}^0 - \underline{\underline{\Gamma}} \cdot \underline{\delta S},\tag{9}$$

$$\underline{\dot{\delta S}} = -\left\{ \underline{(\bigstar h^0)} + \underline{\underline{\Gamma}} \right\} \cdot \underline{\delta S} + \underline{(\bigstar S^0)} \cdot \underline{\delta h}, \tag{10}$$

which we present in two equivalent forms. During the derivation of the last two equations we used the antisymmetry of the cross product, or equivalently, the *total* antisymmetry of the Levi-Civita tensor, or equivalently the antisymmetry of the associated dual matrix. We turn to solve the equation (10) for the eigenenergies (eigenfrequencies) and for the dynamical susceptibility within our formalism.

⁴Remember, this is a classical model.

2.1.3 Eigenenergies and Free Oscillations

First we substitute the harmonic time dependence of $\underline{\delta S}$ and $\underline{\delta h}$ (i.e. $\underline{\delta S} = -i\omega \underline{\delta S}^{\omega}$) in (10) to get an equation for the amplitudes:

$$-i\omega\underline{\delta S}^{\omega} = -\left\{\underline{(\bigstar h^0)} + \underline{\underline{\Gamma}}\right\} \cdot \underline{\delta S}^{\omega} + \underline{(\bigstar S^0)} \cdot \underline{\delta h}^{\omega}.$$
(11)

To get the eigenenergies and eigenmodes we set the dissipation an the small oscillating fields to zero: $\underline{\underline{\Gamma}} = 0$ and $\underline{\delta h}^{\omega} = 0$, multiply by $(-i)\underline{\underline{E}}$, with $\underline{\underline{E}}$ being the 3 × 3 identity matrix, and sort terms to the left hand side:

$$\left\{-i\underline{\left(\bigstar h^{0}\right)}-\omega\underline{\underline{E}}\right\}\cdot\underline{\delta S}^{\omega}=0.$$
(12)

So the eigenfrequencies and eigenmodes are clearly the eigenvalues and eigenvectors of the dynamical matrix $\underline{\Omega} = -i(\bigstar h^0)$. Please note, that the matrix we are searching the eigenvalues of is a 3×3 antisymmetric one times the imaginary unit, so it has eigenvalues of the form $\omega = \pm \omega_0$ and $\omega = 0$. This is reassuring, since the spin component pointing in the direction of \underline{h}^0 commutes with the Hamiltonian, and is therefore a constant of motion (the other two components can be chosen to play the role of the classical analogues of the usual ladder operators). Classically this means that the spin precesses about $-\underline{h}^0$ with angular frequency ω_0 , following the right hand rule (the minus sign comes from absorbing a negative constant in front of the physical field in \underline{h}^0). Next we calculate the susceptibilities.

2.1.4 Susceptibilities and Forced Oscillations

When the small perturbation $\underline{\delta h}^{\omega}$ is applied in Eq. (11) and the linear response is $\underline{\delta S}^{\omega}$, the susceptibility is by definition:

$$\underline{\delta S}^{\omega} = \underline{\chi}(\omega) \cdot \underline{\delta h}^{\omega}.$$
(13)

In order to solve for the susceptibility we sort the terms of $\underline{\delta S}^{\omega}$ on the left side in Eq. (11):

$$\left\{-i\omega\underline{\underline{E}} + \underline{(\bigstar h^0)} + \underline{\underline{\Gamma}}\right\} \cdot \underline{\delta S}^{\omega} = \underline{(\bigstar S^0)} \cdot \underline{\delta h}^{\omega}.$$
(14)

Solving for the oscillating spin components $\underline{\delta S}^{\omega}$ yields:

$$\underline{\delta S}^{\omega} = \underbrace{\left\{-i\omega\underline{\underline{E}} + \underline{(\bigstar h^{0})}_{\underline{\underline{E}}} + \underline{\underline{\Gamma}}\right\}^{-1} \cdot \underline{(\bigstar S^{0})}_{\underline{\underline{\chi}}(\omega)}} \cdot \underline{\delta h}^{\omega}, \tag{15}$$

so the dynamical susceptibility reads as:

$$\underline{\underline{\chi}}(\omega) = \left\{ -i\omega \underline{\underline{E}} + \underline{(\bigstar h^0)} + \underline{\underline{\Gamma}} \right\}^{-1} \cdot \underline{(\bigstar S^0)}.$$
(16)

This is the full frequency-dependent (dynamical) complex linear response matrix. In order to calculate the spin response in physical space all we have to do is choose some $\underline{\delta h}^{\omega}$ at wish (magnitude, excitation frequency and polarization) and calculate $\Re(\underline{\chi}(\omega) \cdot \underline{\delta h}^{\omega})$. Please note that without the dissipation term the susceptibility would diverge at the eigenenergies, as it should. The damping softens these divergences to Lorentzians/ derivative Lorentzians.

2.1.5 Bloch Equations, Comparison with Textbook Results

In order to show that our results reproduce the textbook ones, let us calculate the eigenenergies, eigen- and excited modes and susceptibility of the classical problem of a spin of length S, in a static magnetic field, with a perpendicular perturbing field. Let us suppose that the static field points in the negative \mathbf{z} direction: $\underline{h}^0 = (0, 0, -h^0)^T$, i.e. the physical field $\underline{B}^0 = -\underline{h}^0/(g\mu_B)$ points in the positive \mathbf{z} direction, and so the ground state of the spin points upwards $\underline{S}^0 = (0, 0, S)$. For the eigenmodes and energies we need the eigensystem of the dynamical matrix (Eq. (12)):

$$\underline{\underline{\Omega}} = -i \underbrace{(\bigstar h^0)}_{===} = i \begin{pmatrix} 0 & h^0 & 0 \\ -h^0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$
(17)

for which the eigenenergies are $\omega_1^0 = -h^0$, $\omega_2^0 = +h^0$, $\omega_3^0 = 0$, with eigenvectors: $\delta \underline{S}_1^0 = (-i, 1, 0)^T$, $\delta \underline{S}_2^0 = (i, 1, 0)^T$, $\delta \underline{S}_3^0 = (0, 0, 1)^{T5}$. As was already mentioned the eigenfrequencies come in \pm pairs, and there is no oscillation in the direction of the static field, i.e. $S^z = S$. To have a physical picture of the eigenmodes we have to plot $\Re(\underline{\delta S}_i^0 e^{(-i\omega_i^0 t)})$, for i = 1, 2 (with some initial tilting, i.e. initial condition). What we get is two identical circular precessions about the axis \mathbf{z} , showing that the two eigenmodes are physically identical. In Fig. (1) we show this free oscillation/precession in the xy-plane, with $h^0 = 1$ and of initial tilting of the spin 0.1 in the x-direction (left panel). The arrow shows the spin at t = 0.4. The right panel shows the illustration of the precession of one spin in a ferromagnet: Fig. 1.b. in [5]. In order to calculate the susceptibilities and forced oscillations (Eq. (16)) we set the relaxation times to $T^x = T^y = T_2$ and $T^z = T_1$, where T_1 , T_2 are called the longitudinal and transverse relaxation times, respectively. Since $\underline{S}^0 = (0, 0, S)$ the dual matrix takes the form:

$$\underline{(\bigstar S^0)} = \begin{pmatrix} 0 & S & 0 \\ -S & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$
 (18)

Since the real and imaginary susceptibilities are really complicated expressions, we only illustrate them with the real part of $\chi_{xx}(\omega)$:

$$\Re(\chi_{xx}(\omega)) = \frac{h^0 S T_2^2 \left((h^0)^2 T_2^2 - T_2^2 \omega^2 + 1\right)}{(h^0)^4 T_2^4 + (h^0)^2 \left(2T_2^2 - 2T_2^4 \omega^2\right) + \left(T_2^2 \omega^2 + 1\right)^2},\tag{19}$$

⁵As is clearly seen this problem is diagonal in the circular basis consisting of $S^{\pm} = S^x \pm iS^y$ and S^z , i.e. in the classical analogue of the ladder operators



Figure 1: Left panel: Path of the free oscillation/precession of the spin in the xy-plane, with static field $h^0 = 1$ parallel to the z-axis and of initial tilting of the spin 0.1 in the x-direction. The arrow shows the spin at t = 0.4. (The nice arrows on the curve were produced with the aid of the Mathematica package CurvesGraphics6.nb written by Gianluca Gorni [7], and it can be found on the website: http://sole.dimi.uniud. it/~gianluca.gorni/Mma/Mma.html.). Right panel: Almost identical picture from [5], Fig. 1.b. Please ignore the scripts on the right panel, they are there just beacause it is an illustration of propagating precession.

which is exactly the same as in Chapter 3. of [2] (besides Arovas's gyromagnetic factor is set $\gamma = 1$ in our case). The complex susceptibility is of the form (reassuringly satisfying Onsager reciprocity):

$$\underline{\underline{\chi}}(\omega) = \begin{pmatrix} \chi_{xx} & \chi_{xy} & 0\\ -\chi_{xy} & \chi_{xx} & 0\\ 0 & 0 & 0 \end{pmatrix},$$
(20)

with components:

$$\chi_{xx} = \frac{h^0 S T_2^2}{(h^0)^2 T_2^2 - (T_2 \omega + i)^2},\tag{21}$$

$$\chi_{xy} = \frac{ST_2(iT_2\omega - 1)}{-(h^0)^2 T_2^2 + (T_2\omega + i)^2}.$$
(22)

In order to illustrate the forced oscillations, we excite the spin with the linearly polarized field $(\delta h^x, \delta h^y) = (0.05, 0.1)$, and set $T_2 = \infty$ and $\omega = 6$, far away from the resonance, c.f. Fig. (2). This picture shows the time evolution of the components in the *xy*-plane as calculated from $\Re(\underline{\chi}(\omega) \cdot \underline{\delta h})$. These calculations were done with the (uncommented and very dirty (it is for personal use)) Mathematica notebook **bloch_eqs_dipi.nb**. Next we discuss our general results, and give an outlook to more general problems.



Figure 2: Forced oscillations of the spin as the response to the linearly polarized field $(\delta h^x, \delta h^y) = (0.05, 0.1)$, with no dissipation and $\omega = 6$, far away from the resonance. Time evolution of the spin components in the *xy*-plane.

2.1.6 Discussion

Here we discuss some properties of the results achieved so far. Please note, that the exact equation (5) conserves the spin length (since the "velocity" is perpendicular to the spin), just like its quantum mechanical counterpart. To show this simply dot-multiply Eq. (5) with \underline{S} , the right hand side is trivially zero, and the left hand side is simply half of the time derivative of the spin length. Or more formally multiply the componentwise equation by S^{ζ} and sum up to ζ , and use the *total* antisymmetry of the unit tensor. Our linearized equations do not conserve the spin length, but this is not a real problem, as long as the tilting of the spin is small.

As was already mentioned the eigenfrequencies are of the form $\omega_{1,2}^0 = \pm \omega_0$ and $\omega_3 = 0$. The last one corresponds to the fixedness of the component pointing in the direction of the field (say z), and the former two are associated to the same physical oscillation, as has already been seen. Although it seems as a simple consequence of the well known algebraic property of the eigenvalues of antisymmetric matrices it has deep roots in the structure of the algebra $\mathfrak{su}(2)$. The ground state corresponds to S^z , which is the only element of the so-called Cartan subalgebra of $\mathfrak{su}(2)$, and physically plays the role of the order parameter. And the other two components can be chosen to be the adjoint pairs of the ladder operators: $S^{\pm} = S^x \pm iS^y$, this is the so-called Cartan-Weyl basis. This concept –mutatis mutandis– survives to the general case $\mathfrak{su}(\mathfrak{n})$. There is another way of looking at this: quantum mechanically this $\mathfrak{su}(2)$ -model is a two component system, with a Hilbert space of dimension 2, so there is only one transition from the ground state to the only excited state, with the given eigenfrequency.

Our next observation is the following: this simple technique works for spins of any length S as long as the only operators present in the Hamiltonian are the spin components. Even an innocently looking tiny little anisotropy (e.g. the operator $(S^z)^2$, possible if S > 1/2) could cause real trouble, since it introduces higher order polynomials of spin components in the commutation relations (EOM-s), which will have dynamics themselves, and so on, as long as we exhaust the possibilities of the Hilbert space (there are at most $((2S + 1)^2 - 1)$ linearly independent, traceless, selfadjoint operators in the space). The reason why the spin components alone cannot cause a problem (i.e. they evolve between themselves), that they form a closed subalgebra in the selfadjoint operator-space for any spin length. (More on this, see later.)

Let us mention a few ingredients which we used in the derivations. For the eigenenergies/eigenoscillations we needed to solve the eigenproblem of a not too big dynamical matrix, a matrix which was defined through the structure constants of the algebra, which are extremely simple in this case. In any other algebra the structure constants in any basis are unknown (although they are tabulated for a few algebras and bases in dedicated books, e.g. [24]), so we have to calculate them ourselves, which is not an entirely trivial task to do. The interested reader should consult the Appendix about this topic. Another fact we thoroughly used was the total antisymmetry of the structure constants. Unfortunately this will not generalize to more complicated algebras trivially. For the calculations of the susceptibilities we needed to calculate the inverse of a matrix valued function. Another important ingredient was the ground state, which in this case was trivial, the spin pointed to the direction of the field. But in more complicated (i.e. interacting or lattice systems) finding the proper equilibrium state is a formidable task in itself.

As a summary: we derived the EOM for a single spin in a magnetic field (introducing a convenient notation that is a good subject to generalizations), calculated the eigenenergies and eigenoscillations, gave the frequency dependent complex susceptibility matrix and illustrated our results with a concrete example, which we compared to the literature. In what follows we turn our attention to use our method to rederive some classical results of antiferromagnetic resonance.

2.2 Two Spins: Antiferromagnetic Resonance

Here we rederive some results of the classic paper of Keffer and Kittel [12] on the theory of antiferromagnetic resonance, to show that our method is capable of attacking more complicated problems. An excerpt of this theory is presented in Chapter 13. [14]. Here we use notations that bear more resemblance to the ones in [12], we retain the gyromagnetic factor γ and use physical fields <u>H</u>, and denote magnetizations by <u>M</u>.

Consider an antiferromagnetic substance with two sublattices 1, 2 with sublattice magnetizations \underline{M}_1 , \underline{M}_2 respectively. Let us denote the field acting on the sublattice *i* by :

$$\underline{H}_{i} = \underbrace{\underline{H}^{0} + \underline{\delta}\underline{H}}_{\underline{H}^{ext}} + \underline{H}_{i}^{A}, \tag{23}$$

where \underline{H}^0 and $\underline{\delta H}$ are the static and small dynamic parts of the external field \underline{H}^{ext} , assumed to be common for the two sublattices, and \underline{H}_i^A is the anisotropy field felt by the spins because their surroundings, and we set $\underline{H}_1^A = -\underline{H}_2^A = \underline{H}^A$, parallel to the z-direction.⁶ The theoretical purpose of this anisotropy is clear: it stabilizes the equilibrium direction of the sublattice magnetizations in the z-direction. Since the the problem

⁶This choice of the anisotropy field is very unphysical, the real form of it would be something like

without the anisotropy field would be rotationally invariant (in the absence of external fields), hence the ground state undefined. If the length of the moments is M, then the ground state is $\underline{M}_1 = (0, 0, M)^T$ and $\underline{M}_2 = (0, 0, -M)^T$. The antiferromagnetic interaction of the two sublattices is handled by an effective (Weiss) field, with the parameter $\lambda > 0$: \underline{M}_1 "feels" the exchange field $\underline{H}^E = -\lambda \underline{M}_2$ and vice versa. With these the EOM-s of the magnetizations become:

$$\underline{\dot{M}}_1 = \gamma \underline{M}_1 \times (\underline{H}^0 + \underline{\delta H} + \underline{H}_1^A - \lambda \underline{M}_2), \qquad (24)$$

$$\underline{\dot{M}}_2 = \gamma \underline{M}_2 \times (\underline{H}^0 + \underline{\delta H} + \underline{H}_2^A - \lambda \underline{M}_1).$$
⁽²⁵⁾

We divide the magnetizations to static and small oscillating parts as usual: $\underline{M}_i = \underline{M}_i^0 + \underline{\delta M}_i$, substitute these expressions to the EOM-s (25), use the equilibrium conditions and neglect the double- δ terms to arrive at the set of equations for the oscillations of the sublattice magnetizations:

$$\frac{1}{\gamma} \underline{\delta \dot{M}}_1 = \left(-\underline{H}^0 - \underline{H}_1^A + \lambda \underline{M}_2^0\right) \times \underline{\delta M}_1 - \lambda \underline{M}_1^0 \times \underline{\delta M}_2 + \underline{M}_1^0 \times \underline{\delta H},\tag{26}$$

$$\frac{1}{\gamma}\underline{\delta\dot{M}}_2 = (-\underline{H}^0 - \underline{H}_2^A + \lambda \underline{M}_1^0) \times \underline{\delta M}_2 - \lambda \underline{M}_2^0 \times \underline{\delta M}_1 + \underline{M}_2^0 \times \underline{\delta H}.$$
(27)

Substituting the anisotropy field values and introducing the new notation \underline{H}_{i}^{0} for the static fields on each sublattice these equations become:

$$\frac{1}{\gamma} \underline{\delta \dot{M}}_{1} = \underbrace{(-\underline{H}^{0} - \underline{H}^{A} + \lambda \underline{M}_{2}^{0})}_{-\underline{H}_{1}^{0}} \times \underline{\delta M}_{1} - \lambda \underline{M}_{1}^{0} \times \underline{\delta M}_{2} + \underline{M}_{1}^{0} \times \underline{\delta H}, \tag{28}$$

$$\frac{1}{\gamma} \underline{\delta \dot{M}}_2 = \underbrace{(-\underline{H}^0 + \underline{H}^A + \lambda \underline{M}_1^0)}_{-\underline{H}_2^0} \times \underline{\delta M}_2 - \lambda \underline{M}_2^0 \times \underline{\delta M}_1 + \underline{M}_2^0 \times \underline{\delta H}.$$
(29)

With the star notation these equations become the following in block-matrix form:

$$\frac{1}{\gamma}\frac{d}{dt}\begin{pmatrix}\underline{\delta M_1}\\\underline{\delta M_2}\end{pmatrix} = -\begin{pmatrix}\underline{(\bigstar H_1^0)}\\\lambda\underline{(\bigstar M_2^0)}\end{pmatrix} & \underline{\lambda}\underline{(\bigstar M_1^0)}\\\underline{(\bigstar H_2^0)}\end{pmatrix} \cdot \begin{pmatrix}\underline{\delta M_1}\\\underline{\delta M_2}\end{pmatrix} + \begin{pmatrix}\underline{(\bigstar M_1^0)}\\0\\\underline{(\bigstar M_2^0)}\end{pmatrix} \cdot \begin{pmatrix}\underline{\delta H}\\\underline{\delta H}\end{pmatrix}.(30)$$

With a slight abuse of notation let us denote the 6×6 unit matrix by $\underline{\underline{E}}$, and of course we can introduce a 6×6 diagonal damping matrix $\underline{\underline{\Gamma}}$ again, with these at hand we have:

$$\frac{1}{\gamma} \underline{\underline{E}} \cdot \frac{d}{dt} \left(\frac{\delta M_1}{\delta M_2} \right) = - \left(\underbrace{\underbrace{(\bigstar H_1^0)}}_{\lambda \underline{(\bigstar M_2^0)}} \quad \underbrace{(\bigstar H_1^0)}_{\underline{(\bigstar H_2^0)}} \right) \cdot \left(\frac{\delta M_1}{\delta M_2} \right) - \underline{\underline{\Gamma}} \cdot \left(\frac{\delta M_1}{\delta M_2} \right) + \tag{31}$$

$$+ \left(\underbrace{\underbrace{(\bigstar M_1^0)}}{0} \quad \underbrace{0}_{\underline{(\bigstar M_2^0)}} \right) \cdot \left(\underbrace{\underline{\delta H}}{\underline{\delta H}} \right). \tag{32}$$

 $(M_i^z)^2$, but as already mentioned, a realistic anisotropy like this would ruin the use of the simple $\mathfrak{su}(2)$ -method presented here (and used by [12]).

In order to solve for the eigenoscillations and -energies we set $\underline{\Gamma}$ and $\underline{\delta H}$ to zero, and substitute the harmonic time dependence, and multiply by -i:

$$-\omega \frac{1}{\gamma} \underline{\underline{E}} \cdot \left(\underline{\delta M_1^{\omega}}_{\underline{2}} \right) = +i \left(\underbrace{\underbrace{(\bigstar H_1^0)}}_{\lambda \underline{(\bigstar M_2^0)}} \quad \underbrace{(\bigstar H_1^0)}_{\underline{(\bigstar H_2^0)}} \right) \cdot \left(\underline{\delta M_1^{\omega}}_{\underline{\delta M_2^{\omega}}} \right). \tag{33}$$

Which shows, that the eigenvalues and -vectors of the dynamical matrix $\underline{\Omega}$:

$$\underline{\underline{\Omega}} = -i\gamma \left(\underbrace{\frac{(\bigstar H_1^0)}{\lambda \underline{(\bigstar M_2^0)}}}_{(\bigstar \underline{M_2^0})} \quad \underbrace{\frac{\lambda (\bigstar M_1^0)}{(\bigstar H_2^0)}}_{(\bigstar \underline{M_2^0})} \right)$$
(34)

are the eigenfrequencies and eigenoscillation of the system, respectively.

Next we calculate the susceptibility. We define the 6×6 "susceptibility" matrix $\underline{\underline{\chi}}(\omega)$ as usual:

$$\left(\frac{\delta M_1^{\omega}}{\delta M_2^{\omega}}\right): = \underline{\chi}(\omega) \cdot \left(\frac{\delta H^{\omega}}{\delta H^{\omega}}\right), \tag{35}$$

with 3×3 components $\underline{\chi}_{ij}(\omega)$:

$$\underline{\underline{\chi}}(\omega) = \begin{pmatrix} \underline{\underline{\chi}}_{11}(\omega) & \underline{\underline{\chi}}_{12}(\omega) \\ \underline{\underline{\chi}}_{21}(\omega) & \underline{\underline{\chi}}_{22}(\omega) \end{pmatrix}.$$
(36)

Since the physical (net) magnetization is $\underline{\delta M} = \underline{\delta M}_1 + \underline{\delta M}_2$, the physical susceptibility must be defined as the 3×3 matrix:

$$\underline{\delta M}^{\omega} = \underline{\underline{\chi}}^{\text{phys}}(\omega) \cdot \underline{\delta H}^{\omega}.$$
(37)

To calculate the physical susceptibility we substitute the form (36) into the definition (35), and from this form, the calculation of the net magnetization yields:

$$\underline{\delta M}^{\omega} = \left(\underline{\chi}_{11}(\omega) + \underline{\chi}_{12}(\omega) + \underline{\chi}_{21}(\omega) + \underline{\chi}_{22}(\omega)\right) \cdot \underline{\delta H}^{\omega},\tag{38}$$

$$\underline{\chi}^{\text{phys}}(\omega) = \underline{\chi}_{=11}(\omega) + \underline{\chi}_{=12}(\omega) + \underline{\chi}_{=21}(\omega) + \underline{\chi}_{=22}(\omega).$$
(39)

As usual to calculate the "susceptibility" matrix $\underline{\underline{\chi}}(\omega)$, we use the EOM (32) (with the harmonic Ansatz). This yields:

$$\underline{\underline{\chi}}(\omega) = \left\{ -i\frac{\omega}{\gamma}\underline{\underline{E}} + -\left(\underbrace{\underbrace{(\bigstar H_1^0)}}_{\lambda\underline{\underline{(\bigstar M_2^0)}}} \quad \underbrace{\underline{\lambda}(\bigstar M_1^0)}_{\underline{\underline{(\bigstar M_2^0)}}} \right) + \underline{\underline{\Gamma}} \right\}^{-1} \cdot \left(\underbrace{\underbrace{(\bigstar M_1^0)}}_{0} \quad \underbrace{0}_{\underline{(\bigstar M_2^0)}} \right), \quad (40)$$

from which the physical susceptibility can easily be calculated. In what follows we turn to the concrete example used by [12], and compare our results with that.

For this purpose we choose the static field in the z-direction: $\underline{H}^0 = (0, 0, H^0)^T$, the magnitude of the exchange field is $H^E = \lambda M$. If we solve the secular equation of

the dynamical matrix (34) we get the six eigenfrequencies. As before two frequencies corresponding to oscillations in the z-direction are identically zero. In what follows we ignore motions in the z-direction, and we concentrate on the x and y components of the motions (and of course in the susceptibilities any component containing the index z vanishes). The other four eigenfrequencies come in pairs again, namely (we follow the notations of [12]):

$$\omega_1 = -\omega_2 = \gamma \left[H^0 + \sqrt{H^A (2H^E + H^A)} \right], \tag{41}$$

$$\omega_3 = -\omega_4 = \gamma \left[H^0 - \sqrt{H^A (2H^E + H^A)} \right].$$
(42)

The eigenvectors have a complicated form, so we do not give them here. All our eigenfrequencies and eigenoscillations agree with that of [12]. Their characteristic is the following: when viewed along the z-axis the two spins precess in the same direction with the same frequency circularly, but on circles of unequal size, always having opposite directions (of course the x, y components). In the left panel of Fig. 3 we show this motion, with the parameters: $\gamma = 1$, $H^0 = 0.01$, $H^A = 0.1$, and $H^E = 0.3$. The initial tilting of the spin precessing on the larger circle is 0.1. With these parameters $\omega_1 \approx 0.254$, the arrows show the spin positions at t = 2. The right panel is Fig. 3.b. in [5], and it shows similar behaviour (actually it shows the motion of a pair of nearest neighbor spins in an antiferromagnetic lattice). Another note: there are two types of motion here, the orientation of the precession is fixed by the the direction of the effective field, but either the first or the second spin can precess on the larger circle. Let us turn to the susceptibility calculation. For better agreement with [12] we set the dissipation term to zero: $\underline{\Gamma} = 0$. We calculate (40) and substitute in (39) to get the physical susceptibility (of course without the z-components):

$$\underline{\chi}_{\underline{\underline{\chi}}}^{\mathrm{phys}}(\omega) = \begin{pmatrix} \chi_{xx}(\omega) & \chi_{xy}(\omega) \\ -\chi_{xy}(\omega) & \chi_{xx}(\omega) \end{pmatrix}.$$
(43)

The components read as:

$$\chi_{xx}(\omega) = \frac{-2\gamma^2 H^A M(\omega^2 + \omega_1 \omega_3)}{(\omega^2 - \omega_1^2)(\omega^2 - \omega_3^2)},$$
(44)

$$\chi_{xy}(\omega) = \frac{4i\gamma^3 H^0 H^A M \omega}{(\omega^2 - \omega_1^2)(\omega^2 - \omega_3^2)}.$$
(45)

It is clear from the form of the physical susceptibility matrix (43) that it satisfies Onsager reciprocity. The diagonal components are real, and the offdiagonal ones are pure imaginary, this is a consequence of setting the dissipation to zero. In Fig. 4. we present $\chi_{xx}(\omega)$ for the parameter values $H^0 = 0.1$, $\gamma = 1$, M = 1, $H^A = 0.1$, and $H^E = 0.3$, it is (derivatively) peaked at the eigenfrequencies $\omega_1 = 0.164$, $\omega_3 = 0.365$, as it should. The divergences at these frequencies are the consequence of setting $\underline{\Gamma} = 0$. We conclude our discussion of the antiferromagnetic resonance.



Figure 3: Left panel: Path of the free oscillation/precession of the sublattice magnetizations in the xy-plane, with static field $H^0 = 0.01$, $H^A = 0.1$, and $H^E = 0.3$ parallel to the z-axis, and of initial tilting of the spin on the first sublattice 0.1. The arrows show the sublattice magnetizations at t = 2. Right panel: Almost identical picture from [5], Fig. 3.b. Please ignore the scripts on the right panel, they are there just beacause it is an illustration of a propagating AFM precession.

In this subsection we derived the eigenfrequencies and eigenoscillations and the full complex dynamical susceptibility matrix of an antiferromagnetic material (at zero wavevector $\mathbf{q} = 0$), and rederived some of the classical results of [12]. The anisotropy fields used by the mentioned article and us were very unphysical, but at this level of the EOM, the only ones that can be handled. In what follows we demonstrate that with the proper modifications, the EOM is capable of handling much more realistic anisotropy fields, i.e. fields that contain products of the spin component operators (e.g. easy plane single ion-anisotropy, $\Lambda(S^z)^2$, with positive Λ).

2.3 Single Spin: the General Hamiltonian

2.3.1 Introduction

In this subsection we derive the EOM for a single spin (S > 1/2), that contains higher (than first) order polynomials of spin components. For this purpose first we discuss some properties of the corresponding Hilbert space and its observables, define the multipolar operators, discuss how to handle them in a general framework, based on a few properties of the Lie-algebras $\mathfrak{su}(\mathfrak{n})$. We give a mild introduction of the very few properties of these algebras we will need in the calculations. A very short review of the literature follows, where we mention the standard bosonization technique usually used to handle spin models (we will not use bosonization in this work). After that we derive the EOM, and use it in a concrete example. Some technical details of the calculations are relegated to the Appendix. First we start with the structure of the Hilbert space of a spin S "particle".



Figure 4: $\chi_x x$ -component (without dissipation it is pure real) of the dynamic susceptibility of an antiferromagnet, the two derivative peaks are centered at the resonance frequencies $\omega_{1,3}$.

The Hilbert space structure and the observables on the Hilbert space for a spin S = 1 are described in the book chapter [23], and in the PhD thesis [30]. For a spin of length S = 3/2 the observables (classified for a concrete problem of a multipolar spin model) are given in Chapter 2. [25]. The book [11] is a good introduction to group and representation theory in physics, its Chapters 8. and 9. cover the topics of the groups SU(N) and their algebras, and their representation theory. The book [10] is a very practical mixture of a definition and theorem summary and a cookbook for Lie algebras, with a lot of useful information for practical calculations. Just like [24], where up to n = 4 the structure constants, useful bases, Casimir operators and representation theoretical tools for the algebras $\mathfrak{su}(\mathfrak{n})$ are summarized and tabulated. We used the tables in the latter to check our calculations about the structure of the algebras (c.f. Appendix). The book [8] is a mathematically precise, though very readable one about Lie groups, -algebras and their representations. We will not use representation theory in this work. In what follows we start with the description of the multipoles.

2.3.2 Multipoles

The Hilbert space of a spin of length S is of dimension n = (2S + 1), and is a module of the irreducible representation of the spin algebra $\mathfrak{su}(2)$, with generators S^{α} , $\alpha = x, y, z$, i.e. the spin components. The spin components (in this irrep) are traceless selfadjoint matrices of dimension $n \times n$, and as a Lie algebra they form a 3-dimensional real vector space, endowed with their usual commutator as an extra structure. These two structures (commutators and linear combinations) were the only structures on this Hilbert space we have used so far. But the space of quantum mechanical observables is much richer: any selfadjoint traceless matrix of dimension $n \times n$ could be chosen.⁷ A simple counting of real

 $^{^{7}}$ Tracelessness is taken just for convenience, we could always introduce the scalar matrix as -a trivial-observable.

parameters shows that these observables form a $(n^2 - 1)$ dimensional real vector space. Clearly these matrices can be constructed from the spin components, by multiplying them together. This way one can define the *multipolar operators*, or just multipoles. We give a few examples.

For S = 1/2, we have n = 2, so $(n^2 - 1) = 3$, and the only nontrivial observables are the spin components themselves, namely the *dipoles*. For S = 1, n = 3, $(n^2 - 1) =$ 8 = 3 + 5, so besides the dipoles we have five quadrupoles, of the form $Q^{\alpha\beta} = \overline{S^{\alpha}S^{\beta}} =$ $S^{\alpha}S^{\beta} + S^{\beta}S^{\alpha}$, where we have introduced a notation for the symmetrization of operator products. Symmetrization is needed to ensure hermiticity⁸. So in a spin-1 system there are observables of the form $Q^{zz} = (S^z)^2 - \text{Tr}((S^z)^2)$ (which can play the role of single ion anisotropy in an effective Hamiltonian), or of the form a typical quadrupole Q^{xy} = $S^{x}S^{y}+S^{y}S^{x}$. In an S=3/2 system n=4, $(n^{2}-1)=15=3+5+7$. Here we have besides the dipoles and quadrupoles also 7 octupoles, third order symmetrized polynomials of the spin components, e.g. $\overline{S^x S^z S^z} = 2O^{zxz} + O^{xzz} + O^{zzx}$, or simply $(S^z)^3 - \text{Tr}((S^z)^3)$, where the octupolar operators are defined as $O^{\alpha\beta\gamma} = S^{\alpha}S^{\beta}S^{\gamma}$. For longer spins more and more operators would appear. The enumeration and classification (e.g. under the symmetries of the site-symmetry group of an embedding lattice) of these operators is beyond the scope of this thesis. An example is shown in [25]. Several useful bases are known to the literature for these multipolar operators: the so-called irreducible/spherical tensors presented in [23] and [30], the tesseral harmonics and the Stevens operators. Classification and useful properties, relations, tables and definitions of the multipoles can be found in dedicated books, e.g. [31]. We give some useful multipolar bases in the Appendix. We turn to show that the presence of multipoles in a Hamiltonian has severe consequences.

The effect of these multipoles if present in a Hamiltonian is dramatic. Consider for example a spin of length S = 3/2 embedded in an environment whose effect is modeled by an effective anisotropy field in this toy-Hamiltonian:

$$H^{AI} = \Lambda(S^z)^2, \ \Lambda > 0, \tag{46}$$

classically this is easy to interpret. If we think about the magnetic moment as an (axial)vector this term simply forces it to lie in the easy xy-plane. Quantum mechanically this lifts the degeneracy of the S^z eigenstates (S^z is a good quantum number), with the states $S^z = \pm 1/2$ being the low-lying, and $S^z = \pm 3/2$ high-lying ones, respectively. We show the consequences of the presence of the anisotropy by writing down the EOM-s of the spin components:

$$\dot{S}^x = i \left[H^{AI}, S^x \right] = -\Lambda \overline{S^z S^y} = -\Lambda Q^{yz}, \tag{47}$$

$$\dot{S}^{y} = i \left[H^{AI}, S^{y} \right] = +\Lambda \overline{S^{z} S^{x}} = +\Lambda Q^{xz}, \tag{48}$$

$$\dot{S}^z = i \left[H^{AI}, S^z \right] = 0. \tag{49}$$

These equations are not closed, since they involve quadrupoles, namely Q^{yz} and Q^{xz} (the spin z-component is conserved because of the the z-rotational invariance of H^{AI}). To get

⁸We could use antisymmetrization and a multiplication by i to ensure hermiticity too, but this procedure would simply result in dipoles again.

a closed set of equations we need the time evolution of the quadrupolar operators:

$$\dot{Q}^{yz} = i \left[H^{AI}, Q^{yz} \right] = +\Lambda \{ 2O^{zxz} + O^{xzz} + O^{zzx} \} = +\Lambda O^1, \tag{50}$$

$$\dot{Q}^{xz} = i \left[H^{AI}, Q^{xz} \right] = -\Lambda \{ 2O^{zyz} + O^{yzz} + O^{zzy} \} = -\Lambda O^2, \tag{51}$$

(52)

the above combinations are symmetric in their component-indices, hence selfadjoint. Let the octupoles evolve, and because our spins are of length $S = \frac{3}{2}$ our equations finally close (we do not write down this last set of equations). The moral is the following: if there any multipolar operators in a Hamiltonian, it will generate EOM-s for the other multipolar operators. The sole exceptions are the spin components themselves, since they form a closed subalgebra of the observables, i.e. they evolve among themselves. This is the reason why we need to generalize the EOM-s if there are multipoles present. As a sidenote: we do not need this generalization as long as there are only dipoles present in the Hamiltonian, c.f. the first subsection. Very general models fall into this class: (anisotropic) ferro- or antiferromagnetic Heisenberg models, even containing Dzyaloshinskii-Moriya interaction. Next we describe the very few ingredients of Lie theory we use in this work.

2.3.3 The Lie-algebra $\mathfrak{su}(\mathfrak{n})$

In order to handle multipolar models we have to handle all the multipoles on equal footing. For this purpose we introduce a handy notation: given a spin of length S, its n = (2S + 1) dimensional Hilbert space can support $(n^2 - 1)$ independent, selfadjoint, traceless matrices (i.e. the observables), as already mentioned, let us denote them by A^{α} , $\alpha = 1, 2, \ldots (n^2 - 1)$. With the real linear structure on them together with the usual commutators they define the Lie-algebra $\mathfrak{su}(\mathfrak{n})$. The commutators are defined by the ordinary matrix multiplication:

$$\left[A^{\alpha}, A^{\beta}\right] = A^{\alpha}A^{\beta} - A^{\beta}A^{\alpha} = i\sum_{\gamma} f_{\alpha\beta}{}^{\gamma}A^{\gamma},$$
(53)

and $f_{\alpha\beta}^{\ \gamma}$ are the *structure constants* of the algebra. Here $f_{\alpha\beta}^{\ \gamma}$ is clearly antisymmetric in the indices $\alpha\beta$, but generally there holds no antisymmetry between the index γ and the others, putting γ in upper index reminds us of this fact. By writing out a prefactor *i* explicitly we ensure that the structure constants are real. With this definition the commutator is bilinear, antisymmetric, and instead of associativity it has a property called Jacobi-identity, which we will not use here. We recall some properties of the algebras.

As was already mentioned, and heavily used, the algebra $\mathfrak{su}(\mathfrak{n})$ always contains $\mathfrak{su}(2)$ as a subalgebra (a linear subspace closed under commutation), which was explicitly seen by constructing a basis for the algebra as the multipoles. A well known standard basis in $\mathfrak{su}(2)$ consists of the *Pauli matrices*, and in $\mathfrak{su}(3)$ they have their analogues, the *Gell-Mann matrices* (for their definition c.f. Chapter 4. of [24]). They have quite obvious extensions to higher order algebras $\mathfrak{su}(\mathfrak{n})$ (for $\mathfrak{su}(4)$ they are tabulated in Chapter 5. of [24]). The Pauli matrices are just twice the spin matrices $\sigma^{\alpha} = 2S^{\alpha}$, $\alpha = x, y, z$ in their two dimensional (S = 1/2) defining representation. Here we briefly recall some properties and definitions of the algebra $\mathfrak{su}(2)$ and summarize those of them which have analogues in higher order algebras, and the ones we will use later.

The defining relations of the $\mathfrak{su}(2)$ Lie-algebra are:

$$\begin{bmatrix} S^x, S^y \end{bmatrix} = iS^z, \qquad \begin{bmatrix} S^z, S^x \end{bmatrix} = iS^y, \qquad \begin{bmatrix} S^y, S^z \end{bmatrix} = iS^x, \tag{54}$$

$$S^{\pm} = S^{x} \pm iS^{y}, \quad [S^{z}, S^{\pm}] = \pm S^{\pm}, \quad [S^{+}, S^{-}] = 2S^{z}, \tag{55}$$

$$S^{x} = \frac{1}{2}(S^{+} + S^{-}), \quad S^{y} = \frac{1}{2i}(S^{+} - S^{-}).$$
(56)

Here we have introduced the so-called *ladder operators* S^{\pm} , which come as an adjoint pair. Together with S^z they form a non-selfadjoint basis for the algebra. They are useful in constructing representations, but as we have already seen they also correspond to the same frequency oscillation (excitation) over the ground state (a state in which the component S^z plays the role of the *order parameter*). If we choose $\hat{\mathbf{z}}$ as the quantization axis, than the defining representation of the spin-components become the matrices:

$$S^{x} = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \ S^{y} = \frac{1}{2} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \ S^{z} = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \ S^{+} = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \ S^{-} = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}.$$
(57)

This structure of the operator survives to the case of higher n, in a more complicated form.

Let us define the rank r of the algebra $\mathfrak{su}(\mathfrak{n})$ as the number of mutually commuting selfadjoint elements in the algebra: H^i , for which $[H^i, H^j] = 0, i, j = 1, \dots r$. As a fact r = (n-1). In the $\mathfrak{su}(2)$ r = 1, and H^1 can be chosen as S^2 . The subalgebra spanned by these mutually commuting operators is the Cartan subalgebra. The other n(n-1) operators can be chosen to play the role of generalized ladder operators E^{α} , and they actually come in n(n-1)/2 pairs $E^{\pm \alpha}$. Unfortunately the have much more complicated commutation relations in the n > 2 case than in the n = 2 case. They are also very useful tools for computing representations. A particularly useful choice of these generalized ladder operators and the Cartan subalgebra is called the Cartan-Weyl basis. We will not give all their defining properties here, the interested reader can found them in any of the books on Lie-algebras and their representations mentioned before. But to have a feeling of their complicated commutation relations we give a table of them for n = 4 in the Appendix. We have calculated this table with the aid of the Mathematica notebook su4_CartanWeyl.nb, and with a few technical tricks. We briefly describe these tricks in the Appendix. We used several other bases (namely multipolar ones), for which the structure factors were also calculated. We tried to check all these calculations, by comparing lots of commutators randomly chosen to the ones in the tables in Chapter 5. of [24]. We used these structure constants in the calculation of the properties of a multipolar onsite model (see later).

As for the $\mathfrak{su}(2)$ model the important ingredients of this construction for us are the following. Since the Cartan subalgebra contains mutually commuting operators, they can have simultaneously measurable ground state expectation values, i.e. they can play the role of *order parameters*. The generalized ladder operator pairs play the roles of the excitations. As a little counting shows, in an *n*-level system there are n(n-1)/2 transition

frequencies, and they correspond to the n(n-1)/2 operator pairs $E^{\pm \alpha}$. And as will be seen, in our linearized EOM method for the classical analogues of the elements of the Cartan subalgebra there correspond modes with zero frequency, as was already seen for the simple spin model (for S = 3/2, n = 4, r = 3, and the number of ladder operator pairs is 6). In what follows we give some references about the bosonization techniques that are usually used to handle spin systems.

2.3.4 A Very Brief Review of Bosonization Methods in Spin Systems

As the usual method of solving spin systems is some kind of bosonization method, here we give some references on them. Since our hope is that the EOM gives the same results as the techniques mentioned above, we feel an urge to mention them. We do not use bosonization here, so we will only give the simplest example (i.e. Schwinger bosons), to give a reader a feeling.

As we seen a spin oscillates in a magnetic field, and since there is a very deep relation between bosons and oscillators, one has a feeling that there must be some way spin operators can be mapped to bosonic ones. The feeling is right, one way to do this is by the use of the Schwinger bosons (c.f. [26]). As we have already seen, spin commutations are operator valued, hence very complicated in practical calculations, but the bosonic commutators are simple and beautiful, hence people usually use bosonic representations in real calculations of the properties of spin systems. Here follow the definitions of the Schwinger bosonization.

To represent the spin operators let us introduce two independent bosons: a, b, these are the so-called Schwinger bosons. They satisfy the usual bosonic algebra: $[a, a^+] = [b, b^+] = 1$ (where a^+ denotes the adjoint of a, i.e. the corresponding creation operator), and they are independent in the sense, that $[a, b^+] = 0$ (and so on). They annihilate a bosonic vacuum ket $|00\rangle = |0\rangle$, i.e. $a|0\rangle = b|0\rangle = 0$. The associated number operators are $n_a = a^+a$ and $n_b = b^+b$. Some useful relations are summarized in the following:

$$[a, a^+] = [b, b^+] = 1,$$
 (58)

$$[a,a] = [b,b] = [b,a^+] = [a,b^+] = [a,b] = [a^+,b^+] = 0,$$
(59)

$$a|0\rangle = b|0\rangle = 0, \ n_a = a^+a, \ n_b = b^+b, \ n = n_a + n_b,$$
 (60)

$$[n_a, a] = -a, \ [n_b, b] = -b, \ [n_a, a^+] = a^+, \ [n_b, b^+] = b^+.$$
(61)

These bosons are capable of representing general S spins, but we will soon restrict ourselves to the S = 1/2 case. The spin operators can be represented as boson-bilinears. For this purpose let us introduce the boson-valued spinors (a^+, b^+) and $(a, b)^T$. The bosonic representation of the spin operators consists of sandwiching the matrix forms of spin operators in, e.g.:

$$S^{z} = (a^{+}, b^{+}) \cdot D_{\frac{1}{2}} [S^{z}] \cdot {\binom{a}{b}} = (a^{+}, b^{+}) \frac{1}{2} {\binom{1}{0}} \frac{0}{-1} {\binom{a}{b}} = \frac{1}{2} (a^{+}a - b^{+}b) = \frac{1}{2} (n_{a} - n_{b}). (62)$$
$$S^{x} = \frac{1}{2} (a^{+}b + b^{+}a), \ S^{y} = \frac{1}{2i} (a^{+}b - b^{+}a), \ S^{+} = a^{+}b, \ S^{-} = b^{+}a. (63)$$

These bosons are well suited to describe paramagnetic materials, since they handle up and down spins on an equal footing. We close our introduction of an enumeration of the literature where the intrigued reader can find the details of this very powerful technique.

When handling ordered materials (of Heisenberg-like –say ferromagnetic– Hamiltonians, containing no multipoles) one usually uses some modified version of the Schwinger bosons, where one of the operators is condensed. This represents the fully polarized ground state, and the other boson is used as a dynamical variable that handles the oscillations about the ground state. One of this representation is known as the Holstein-Primakoff representation, used for ferromagnetic and antiferromagnetic systems in e.g. Chapter 6. of [6], or in Chapter 15. in [28]. One of the very first attempts to use bosons in the description of ferromagnets is the classic article [9]. Another useful representation is the so-called Dyson-Maleev representation.

If there are multipolar operators in the Hamiltonian (either as anisotropy fields, or multipolar interactions) the standard techniques mentioned above fail. The first attempt to generalize the bosonization technique to multipolar (i.e. $\mathfrak{su}(\mathfrak{n})$) models was these two articles [20] and [21] by Papanicolaou, where the so-called *multiboson* or *flavor wave* theory was introduced. After that very general introduction of the method a series of papers by Onufrieva followed: [17], [18], [19] and [32], where she used the method to describe multipolar (usually ferromagnets with anisotropy) Hamiltonians. In the paper [22] the authors used the flavor-wave method to describe the (very unusual) excitations of an antiferromagnetic insulator, with strong on-site anisotropy, that is either multiferroic. As far as I know, this is the first occasion when the multiboson technique was –actually very– successful in describe multipolar materials. The two authors of the dissertations [25] and [30] used the method to describe multipolar materials and Hamiltonians. Their exposition of the subject is very pedagogical, I would suggest [25] as a first introduction to flavor wave theory. We close this very brief subsection, and turn to the derivation of the EOM method for on-site multipolar Hamiltonians.

2.3.5 The Equation of Motion Method for the On-site Multipolar Hamiltonian

In order to pave the way to the construction of the EOM for multipolar lattice problems, here we develop our method for the multipolar on-site problem. The general (multipolar) on-site Hamiltonian reads as:

$$H^{h} = \underline{h}^{T} \cdot \underline{A} = \sum_{\alpha} h^{\alpha} A^{\alpha}, \tag{64}$$

with generalized magnetic field components h^{α} , and operators taking values in the Liealgebra $\mathfrak{su}(\mathfrak{n})$, and $\alpha = 1, 2, \ldots (n^2 - 1)$, with n = 2S + 1 the dimension of the Hilbert space. Please note that besides the usual magnetic field, the components of the general field play other roles. For example, if $A^4 = ((S^z)^2 - \operatorname{Tr}((S^z)^2))$ than $h^4 = \Lambda$ plays the role of the ons-site anisotropy energy. The generalized spin operators are defined by their commutators, i.e. by the structure constants of the Lie-algebra $\mathfrak{su}(\mathfrak{n})$:

$$\left[A^{\alpha}, A^{\beta}\right] = i \sum_{\gamma} f_{\alpha\beta}{}^{\gamma} A^{\gamma}.$$
(65)

The time evolution of a single component is governed by the Heisenberg equation of motion:

$$\dot{A}^{\zeta} = i \left[H^h, A^{\zeta} \right] = i \sum_{\alpha} h^{\alpha} \left[A^{\alpha}, A^{\zeta} \right] = -\sum_{\alpha, \gamma} f_{\alpha \zeta}^{\ \gamma} h^{\alpha} A^{\gamma}.$$
(66)

In order to calculate the eigenfrequencies and eigenoscillations, just like in the simple spin case we divide the fields and the observables into static and small oscillating parts, and remark that in the static case there is no time evolution:

$$A^{\alpha}(t) = A^{0,\alpha} + \delta A^{\alpha}(t) = A^{0,\alpha} + \delta A^{\omega,\alpha} e^{-i\omega t}, \qquad (67)$$

$$h^{\alpha}(t) = h^{0,\alpha} + \delta h^{\alpha}(t) = h^{0,\alpha} + \delta h^{\omega,\alpha} e^{-i\omega t}, \tag{68}$$

$$\dot{A}^{0,\zeta} = -\sum_{\alpha,\gamma} f_{\alpha\zeta}{}^{\gamma} h^{0,\alpha} A^{0,\gamma} = 0.$$
(69)

Putting these forms into Eq. (66) yields:

$$\dot{\delta A}^{\zeta} = -\sum_{\alpha,\gamma} f_{\alpha\zeta}^{\gamma} \left(h^{0,\alpha} + \delta h^{\alpha} \right) \left(A^{0,\gamma} + \delta A^{\gamma} \right) \approx \tag{70}$$

$$\approx -\sum_{\alpha,\gamma} f_{\alpha\zeta}^{\ \gamma} \left(h^{0,\alpha} \delta A^{\gamma} + A^{0,\gamma} \delta h^{\alpha} \right) =$$
(71)

$$= -\left(\sum_{\gamma} \left(\sum_{\alpha} f_{\alpha\zeta}{}^{\gamma} h^{0,\alpha}\right) \delta A^{\gamma} + \sum_{\alpha,\gamma} f_{\alpha\zeta}{}^{\gamma} A^{0,\gamma} \delta h^{\alpha}\right).$$
(72)

In the last term we interchange the dummy indices $\alpha \leftrightarrow \gamma$:

$$\dot{\delta A}^{\zeta} = -\left(\sum_{\gamma} \underbrace{\left(\sum_{\alpha} f_{\alpha\zeta}{}^{\gamma} h^{0,\alpha}\right)}_{(\textcircled{O}h^{0})_{\zeta\gamma}} \delta A^{\gamma} + \sum_{\gamma} \underbrace{\left(\sum_{\alpha} f_{\gamma\zeta}{}^{\alpha} A^{0,\alpha}\right)}_{(\textcircled{O}A^{0})_{\zeta\gamma}} \delta h^{\gamma}\right),\tag{73}$$

or in matrix form:

$$\underline{\dot{\delta A}} = -\underline{(\textcircled{\bullet}h^0)} \cdot \underline{\delta A} - \underline{(\textcircled{\bullet}A^0)} \cdot \underline{\delta h},\tag{74}$$

where we have introduced the notations:

$$(\mathbf{\mathfrak{O}}A)_{\zeta\gamma} = \sum_{\alpha} f_{\alpha\zeta}{}^{\gamma}A^{\alpha},\tag{75}$$

$$(\textcircled{A})_{\zeta\gamma} = \sum_{\alpha} f_{\gamma\zeta} \,^{\alpha} A^{\alpha}, \tag{76}$$

please note the different orientations and colors of the stars⁹, and the indices. The two matrices do not have that simple relation as in the simple spin case, because there is

⁹Especially because I sweated blood to manage to do them in LaTeX.

no obvious antisymmetry between lower and upper indices¹⁰. By introducing the phenomenological matrix of inverse lifetimes $\underline{\Gamma}$ like before the EOM becomes:

$$\underline{\dot{\delta A}} = -\left\{ \underbrace{\underline{(\mathbf{\mathfrak{O}}h^0)}}_{\underline{}} + \underline{\underline{\Gamma}} \right\} \cdot \underline{\delta A} - \underbrace{\underline{(\mathbf{\mathfrak{O}}A^0)}}_{\underline{}} \cdot \underline{\delta h}, \tag{77}$$

and substituting the harmonic time dependence the equation yields:

$$-i\omega\underline{\delta A}^{\omega} = -\left\{\underline{(\textcircled{}h^{0})}_{} + \underline{\underline{\Gamma}}\right\} \cdot \underline{\delta A}^{\omega} - \underline{(\textcircled{}A^{0})}_{} \cdot \underline{\delta h}^{\omega}.$$
(78)

In order to get the eigenmodes and eigenfrequencies we set $\underline{\underline{\Gamma}} = 0$ and $\underline{\delta h}^{\omega} = 0$ in Eq. (77), and multiply the equation by $(-i)\underline{\underline{E}}$, with $\underline{\underline{E}}$ being the $(n^2 - 1) \times (n^2 - 1)$ identity matrix:

$$\left\{ (-i)\underline{\left(\textcircled{}h^{0}\right)} - \omega \underline{\underline{E}} \right\} \cdot \underline{\delta A}^{\omega} = \underline{0},$$
(79)

so the eigenfrequencies and eigenmodes are the eigenvalues and eigenvectors of the dynamical matrix $\underline{\Omega} = (-i)(\underline{\textcircled{O}}h^0)$, respectively. When the small perturbation $\underline{\delta}h^{\omega}$ is applied and the response is $\underline{\delta}A^{\omega}$, the susceptibility is by definition:

$$\underline{\delta A}^{\omega} = \underline{\underline{\chi}}(\omega) \cdot \underline{\delta h}^{\omega}.$$
(80)

Sorting the terms of $\underline{\delta A}^{\omega}$ on the left side in Eq. (78) we have:

$$\left\{-i\omega\underline{\underline{E}} + \left(\underline{(\textcircled{}h^{0})}{\underline{\underline{E}}} + \underline{\underline{\Gamma}}\right)\right\} \cdot \underline{\delta A}^{\omega} = -\underline{(\textcircled{}A^{0})}{\underline{\underline{\delta}}} \cdot \underline{\delta h}^{\omega},\tag{81}$$

and solving for $\underline{\delta A}^{\omega}$ yields:

$$\underline{\delta A}^{\omega} = \underbrace{-\left\{-i\omega\underline{\underline{E}} + \left(\underbrace{(\textcircled{} h^{0})}_{\underline{\underline{E}}} + \underline{\underline{\Gamma}}\right)\right\}^{-1} \cdot \underbrace{(\textcircled{} A^{0})}_{\underline{\underline{X}}(\omega)} \cdot \underline{\delta h}^{\omega}, \tag{82}$$

so the susceptibility is:

$$\underline{\underline{\chi}}(\omega) = -\left\{-i\omega\underline{\underline{E}} + \left(\underline{(\textcircled{\bullet}h^0)} + \underline{\underline{\Gamma}}\right)\right\}^{-1} \cdot \underline{(\textcircled{\bullet}A^0)}.$$
(83)

It is time to discuss the results achieved so far.

 $^{^{10}}$ Actually there *is* a basis on a Lie-algebra, in which the structure constants are totally antisymmetric, by the use of the Cartan metric we can pull down the upper index to form a totally antisymmetric quantity, but we will not use this procedure here.

2.3.6 Discussion of General Results

We managed to improve our EOM technique to handle on-site problems where multipolar fields are present. We solved the equations to yield the eigenenergies and eigenoscillations of the system. So we have generalized the Bloch equations to include e.g. anisotropy fields. Though this result is not a big deal, but as far as I know, it is new. The eigenenergies could have been calculated by simply solving a little $n \times n$ eigensystem, which we referred to as the quantum mechanical solution. The power of our method is, that with the eigenvectors at hand we can give a pictorial view of the motions of the eigen- and forced oscillations of the multipoles, saving something from the soothing feeling of thinking about a spin as a small arrow precessing about the magnetic field, though the price we pay for this is, that our small arrow precesses in a (n^2-1) dimensional space. Another important observation is, that if multipolar fields are present, new dynamical variables – i.e. the multipolesare needed to describe the system. So if we have e.g. anisotropy fields, the spin, as a classical vector shrinks, in order to give magnitude to the multipolar components. This situation is dubbed in the article [22] as the stretching of the spin. In complete analogy with the spin calculations, the eigenenergies have a certain structure. In the linearized calculation we have r = (n-1) zero modes, these correspond to the steadiness of the r components, pointing in the direction of the external field (i.e. the order parameters), and algebraically these are related to the Cartan subalgebra of our algebra. Since these components mutually commute, they can have simultaneous ground state expectation values. The other (n(n-1)) eigenvalues come in \pm pairs, and each pair represents the same physical oscillation, which even manifests itself in the fact that in a $n \times n$ quantum mechanical system we can have (n(n-1))/2 transition frequencies. Algebraically they are related to the generalized ladder operators of the algebra.

We have calculated the $(n^2 - 1) \times (n^2 - 1)$ (zero temperature, as all calculations in this thesis) complex dynamical susceptibility matrix of the onsite problem that gives the cross-correlation effects, e.g. the multipolar responses to multipolar fields. This could be achieved in the quantum mechanical calculation by the use of Kubo's formula. This result of calculation of multipolar susceptibilities without the use of the mentioned formula is – up to our knowledge- new, although Kubo's formula is much more powerful, since it gives the temperature dependence of these susceptibilities. We have not yet been achieved to generalize our concepts to finite temperature, and at this point it seems to be a very hard task to do (not to mention, that at low temperatures we surely must bosonically quantize our oscillations in order that they have the correct –Bose– statistics). We must mention, that our calculation of the susceptibility needed the ground state as an essential ingredient (which is not a big surprise, since we analyzed small oscillations *about* the ground state). In all practical applications we used the quantum mechanically calculated ground state to calculate the expectation values of the multipoles. This may seem as cheating (as it surely is), but our point is that the onsite problem is only a brick in the road to handle multipolar lattice problems, where the ground state must be calculated in very different (and usually very intricate) ways. Another ingredient was for all the calculations the structure of the algebra. For the trick we used to calculate the structure constants, c.f. the Appendix. Note that the susceptibilities are peaked at the eigenfrequencies, as they

should.

Although this question -single site multipolar oscillations- seems very academical, this is not. We used the quantum version of these calculations to describe the high temperature behaviour of a multiferroic substance [29], but that calculation was simple, pure quantum mechanics (and high temperature susceptibility calculations with Kubo's Formula), unrelated to the EOM and we want to publish it elsewhere. So we do not give here the details. But that calculation is very useful here, since we can check our method's results with that. We turn to this concrete example.

2.3.7 Example: Spin in an Anisotropy Field

Here we briefly describe the results of a calculation of the dynamics of a spin (S = 3/2), when it feels an easy plane on-site anisotropy, and is placed in a magnetic field. The Hamiltonian reads as:

$$H = \Lambda(S^z)^2 - g\mu_B \underline{B}^T \cdot \underline{S},\tag{84}$$

with anisotropy constant $\Lambda = 1.4 \text{ meV} > 0$, gyromagnetic factor g = 2, and the external field *B* is measured in Teslas. We used the tesseral/multipolar basis in this case described in the Appendix, and the structure factors were calculated the way it is in the Appendix. We calculated the eigenvalues of the multipolar dynamical matrix, and they resulted as the transition frequencies in the quantum mechanical calculation. In Fig. 5. give these eigenvalues as a function of the external field, when it is in the easy plane. Actually this picture is taken from the quantum mechanical calculation. In Fig. 6. we show how



Figure 5: Transition frequencies of a spin (S = 3/2) in an easy-plane type anisotropic environment, as a function of the in-plane magnetic field.

the static multipole components evolve under the effect of the external in-plane field (their numbering is in the order dipoles, quadrupoles, octupoles, and the ordering of the multipoles is just like the ordering of the ones in the Appendix). It is clearly seen that besides the spin components other multipolar components have finite ground state expectation values as a consequence of anisotropy. Another thing is that simple magnetic



Figure 6: Evolution of static multipole components (Tesserals in Appendix) under the effect of the external in-plane field. (Their numbering is dipoles, quadrupoles, octupoles.)

field has an effect on the multipoles, too. We have calculated the full susceptibility matrix, but we do not give it here. These calculations are in the notebooks onsite_su4.nb (EOM) and onsite_QM.nb (quantum). As a conclusion: we derived the EOM for the most general onsite model, showed how the full zero temperature susceptibility matrix should be calculated, used the method in a concrete example, and checked with basic quantum mechanical calculations that the method works. We turn our attention to lattice models.

2.4 Lattice Spin Models: the General Hamiltonian

2.4.1 Introduction

In this section we derive the EOM for general, multipolar lattice models, and show a way how to linearize them in order to get tractable equations. We use our technique and notational system on well known models, to check their correctness. These reference models are a simple Heisenberg ferromagnet and a collinear antiferromagnet, with simple commensurate ordering. As a sidenote we mention –but not present here in full detail– an application of our method to a model where the ordering is incommensurate, but an article exists about it. With this last example we illustrate that the EOM is capable of attacking more complicated problems, and gives the same result as the Holstein-Primakoff method. At the end we transform our model to Fourier space, and give the linearized version of it in Fourier space. Unfortunately we give no application of the EOM to the full multipolar lattice model.

2.4.2 Definition of the Model

Our most general model Hamiltonian is of the form:

$$H = H^J + H^h = \frac{1}{2} \sum_{ij} \sum_{\alpha\beta} J_{ij}^{\alpha\beta} A_i^{\alpha} A_j^{\beta} + \sum_i \sum_{\beta} h_i^{\beta} A_i^{\beta},$$
(85)

or in vectorial notation:

$$H = H^J + H^h = \frac{1}{2} \sum_{ij} \underline{A}_i^T \underline{J}_{ij} \underline{A}_j + \sum_i \underline{h}_i^T \cdot \underline{A}_i.$$
(86)

Where the A-s are $\mathfrak{su}(\mathfrak{n})$ -valued fields over the lattice (with lattice points \mathbf{R}_i , \mathbf{R}_j), and their commutators read as:

$$\left[A_i^{\alpha}, A_j^{\beta}\right] = i\delta_{ij} \sum_{\gamma} f_{\alpha\beta}{}^{\gamma} A_j^{\gamma}.$$
(87)

Here i, j are site indices, with no restriction on them besides $i \neq j$, hence the $\frac{1}{2}$ against overcounting the bonds. $\alpha, \beta, \gamma = 1, 2 \dots (n^2 - 1)$ indexes the elements of the algebra, the $J_{ij}^{\alpha\beta}$ are generalized exchange constants (a real $(n^2-1) \times (n^2-1)$ matrix) for every pair of sites, h_i^{β} are generalized (at this point possibly inhomogeneous) fields (a vector with $(n^2 - 1)$ entries), δ_{ij} is the Kronecker symbol and $f_{\alpha\beta}^{\ \gamma}$ are the structure constants of the algebra. It is clear from this form of the model, that it is very general, it can have spin-spin interactions, but also multipolar interactions. It must not be isotropic, nor is it restricted to nearest neighbor interactions. Even an antisymmetric part of the coupling is allowed, and the external field vector can contain a very wide range of on-site fields, e.g. anisotropies and external magnetic or multipolar fields. At this point we do not suppose any other symmetries of the exchange matrix besides translation invariance, i.e. $J_{ij}^{\alpha\beta}$ depends only on $\Delta \mathbf{R} = \mathbf{R}_i - \mathbf{R}_j$: $J_{ij}^{\alpha\beta} = J^{\alpha\beta}(\Delta \mathbf{R})$. It is worth noting, that we can say something about the symmetric and antisymmetric

parts of the exchange matrices:

$$J_{ij}^{\alpha\beta}(S) = \frac{1}{2} \left\{ J_{ij}^{\alpha\beta} + J_{ij}^{\beta\alpha} \right\},\tag{88}$$

$$J_{ij}^{\alpha\beta}(A) = \frac{1}{2} \left\{ J_{ij}^{\alpha\beta} - J_{ij}^{\beta\alpha} \right\}.$$
(89)

By definition these matrices are symmetric (antisymmetric) in the $\alpha\beta$ indices, respectively. The symmetries with respect the change of the site indices ij can be guessed from imagining these generalized spins as rigid bodies attached to the lattice sites, and interchanging them mechanically (imagining the symmetric part as the ordinary exchange, and the antisymmetric part as the Dzyaloshinskii-Moriya interaction may be helpful). Summarizing these properties leads to:

$$J_{ij}^{\alpha\beta}(S) = J_{ij}^{\beta\alpha}(S) = J_{ji}^{\alpha\beta}(S) = J_{ji}^{\beta\alpha}(S), \tag{90}$$

$$J_{ij}^{\alpha\beta}(A) = -J_{ij}^{\beta\alpha}(A) = -J_{ji}^{\alpha\beta}(A) = J_{ji}^{\beta\alpha}(A),$$
(91)

$$J_{ij}^{\alpha\beta} = J_{ji}^{\beta\alpha}.$$
 (92)

These relations may be helpful later¹¹. We turn to the EOM-s.

¹¹The "derivation" of these properties is very handwaving, they surely work for simple (say Bravais) lattices and simple Hamiltonians, but I would not put much trust in them for the very general case. We try to avoid their usage if possible.

2.4.3 The Equation of Motion for the General Hamiltonian

In order to calculate the full time evolution we start with the field part of the Hamiltonian H^h in (85) with the aid of the commutation relations (87):

$$(\dot{A}^{\zeta}{}_{k})_{h} = i \left[H^{h}, A^{\zeta}_{k} \right] = i \sum_{\alpha, i} h^{\alpha}_{i} \left[A^{\alpha}_{i}, A^{\zeta}_{k} \right] = -\sum_{\alpha, \gamma} f_{\alpha\zeta}^{\gamma} h^{\alpha}_{k} A^{\gamma}_{k}, \tag{93}$$

just like in the one site case (66). Or with our star notation:

$$(\underline{\dot{A}}_k)_h = -\underline{(\mathbf{O}h_k)} \cdot \underline{A}_k.$$
(94)

We turn to the exchange part H^J in (85), use the commutation relations (87) and the very general operator identity [AB, C] = A[B, C] + [A, C]B, and calculate the time evolution:

$$(\dot{A}^{\zeta}{}_{k})_{J} = i \left[H^{J}, A^{\zeta}_{k} \right] = i \frac{1}{2} \sum_{ij} \sum_{\alpha\beta} J^{\alpha\beta}_{ij} \left[A^{\alpha}_{i} A^{\beta}_{j}, A^{\zeta}_{k} \right] =$$
(95)

$$= i\frac{1}{2}\sum_{ij}\sum_{\alpha\beta}J_{ij}^{\alpha\beta}\left(A_i^{\alpha}\left[A_j^{\beta}, A_k^{\zeta}\right] + \left[A_i^{\alpha}, A_k^{\zeta}\right]A_j^{\beta}\right) =$$
(96)

$$= -\frac{1}{2} \sum_{ij} \sum_{\alpha\beta\gamma} J_{ij}^{\alpha\beta} \left(\delta_{jk} f_{\beta\zeta}^{\ \gamma} A_i^{\alpha} A_j^{\gamma} + \delta_{ik} f_{\alpha\zeta}^{\ \gamma} A_i^{\gamma} A_j^{\beta} \right) =$$
(97)

$$= -\frac{1}{2} \left(\sum_{ij} \sum_{\alpha\beta\gamma} J_{ij}^{\alpha\beta} \delta_{jk} f_{\beta\zeta}^{\ \gamma} A_i^{\alpha} A_j^{\gamma} + \sum_{ij} \sum_{\alpha\beta\gamma} J_{ij}^{\alpha\beta} \delta_{ik} f_{\alpha\zeta}^{\ \gamma} A_i^{\gamma} A_j^{\beta} \right) =$$
(98)

$$= -\frac{1}{2} \left(\sum_{i \neq k} \sum_{\alpha \beta \gamma} J_{ik}^{\alpha \beta} f_{\beta \zeta}^{\gamma} A_i^{\alpha} A_k^{\gamma} + \sum_{j \neq k} \sum_{\alpha \beta \gamma} J_{kj}^{\alpha \beta} f_{\alpha \zeta}^{\gamma} A_k^{\gamma} A_j^{\beta} \right).$$
(99)

Since we began with $i \neq j$ in (85), in the first sum we have $i \neq k$, and in the second one we have $j \neq k$. As a consequence of the commutation relations (87) we are allowed to interchange the two operators in the second sum, so we have:

$$(\dot{A}^{\zeta}{}_{k})_{J} = -\frac{1}{2} \left(\sum_{i \neq k} \sum_{\alpha \beta \gamma} J_{ik}^{\alpha \beta} f_{\beta \zeta}^{\gamma} A_{i}^{\alpha} A_{k}^{\gamma} + \sum_{j \neq k} \sum_{\alpha \beta \gamma} J_{kj}^{\alpha \beta} f_{\alpha \zeta}^{\gamma} A_{j}^{\beta} A_{k}^{\gamma} \right).$$
(100)

In the second sum we change the dummy index $j \to i$, and interchange the dummy indices $\alpha \leftrightarrow \beta$:

$$(\dot{A}^{\zeta}{}_{k})_{J} = -\frac{1}{2} \left(\sum_{i \neq k} \sum_{\alpha \beta \gamma} J_{ik}^{\alpha \beta} f_{\beta \zeta}^{\gamma} A_{i}^{\alpha} A_{k}^{\gamma} + \sum_{i \neq k} \sum_{\alpha \beta \gamma} J_{ki}^{\beta \alpha} f_{\beta \zeta}^{\gamma} A_{i}^{\alpha} A_{k}^{\gamma} \right) =$$
(101)

$$= -\frac{1}{2} \sum_{i \neq k} \sum_{\alpha \beta \gamma} \left(f_{\beta \zeta}^{\gamma} \left(J_{ik}^{\alpha \beta} + J_{ki}^{\beta \alpha} \right) A_i^{\alpha} A_k^{\gamma} \right) =$$
(102)

$$= -\sum_{\beta\gamma} \left(f_{\beta\zeta}^{\gamma} \underbrace{\left(\frac{1}{2} \sum_{i \neq k} \sum_{\alpha} \left(J_{ik}^{\alpha\beta} + J_{ki}^{\beta\alpha} \right) A_{i}^{\alpha} \right)}_{h_{k}^{\text{eff},\beta}(\{A_{i}^{\alpha}\})} A_{i}^{\alpha} \right)} A_{k}^{\gamma} \right).$$
(103)

This is an equation that is easy to interpret. We define the *effective field* at site k as:

$$h_k^{\text{eff},\beta}\left(\{A_i^\alpha\}\right) = \frac{1}{2} \sum_{i \neq k} \sum_{\alpha} \left(J_{ik}^{\alpha\beta} + J_{ki}^{\beta\alpha}\right) A_i^\alpha,\tag{104}$$

$$\underline{h}_{k}^{\mathrm{eff}}\left(\{\underline{A}_{i}\}\right) = \frac{1}{2} \sum_{i \neq k} \left(\underline{J}_{ik}^{T} + \underline{J}_{ki}\right) \cdot \underline{A}_{i}, \qquad (105)$$

which is just the molecular field the generalized spin at site k feels from its surroundings. For later purposes it is worth to define the matrix:

$$\widetilde{J}_{ki}^{\beta\alpha} = \frac{1}{2} \left(J_{ik}^{\alpha\beta} + J_{ki}^{\beta\alpha} \right), \tag{106}$$

$$\widetilde{\underline{J}}_{ki} = \frac{1}{2} \left(\underline{J}_{ik}^T + \underline{J}_{ki} \right),$$
(107)

this yields:

$$\underline{h}_{k}^{\text{eff}}\left(\{\underline{A}_{i}\}\right) = \sum_{i \neq k} \underbrace{\widetilde{\underline{J}}}_{ki} \cdot \underline{A}_{i}.$$
(108)

With this notation (dropping the argument of the effective field) the EOM becomes:

$$(\dot{A}^{\zeta}{}_{k})_{J} = -\sum_{\gamma} \left(\sum_{\beta} f_{\beta\zeta} \,^{\gamma} h_{k}^{\text{eff},\beta} \right) A_{k}^{\gamma}, \tag{109}$$

or in vectorial form:

$$(\underline{\dot{A}}_k)_J = -\underbrace{(\textcircled{O}h_k^{\text{eff}})}_{k} \cdot \underline{A}_k, \tag{110}$$

$$(\underline{\dot{A}}_{k})_{J} = -\underbrace{\left(\bigotimes\left(\sum_{i \neq k} \underline{\widetilde{\underline{J}}}_{ki} \cdot \underline{A}_{i}\right) \right)}_{\underline{\underline{J}}} \cdot \underline{\underline{A}}_{k}.$$
(111)

With these results at the full time evolution can be written as:

$$\dot{A^{\zeta}}_{k} = (\dot{A^{\zeta}}_{k})_{J} + (\dot{A^{\zeta}}_{k})_{h} = -\sum_{\gamma} \left(\sum_{\beta} f_{\beta\zeta}^{\gamma} \left(h_{k}^{\text{eff},\beta} + h_{k}^{\beta} \right) \right) A_{k}^{\gamma}, \tag{112}$$

or in vectorial form:

$$\underline{\dot{A}}_{k} = -\underline{\left(\mathbf{\mathfrak{O}}(h_{k}^{\text{eff}} + h_{k})\right)} \cdot \underline{A}_{k}.$$
(113)

Although our notation hides the fact, this is a disgusting, coupled nonlinear set of equations, since in the effective field contains a linear combination of the dynamical variables, though these equations surely are exact. In order to solve them we must modify our linearization technique.

2.4.4 Linearization of the Lattice Model

We start to describe the linearization procedure for the easier part of the model: the EOM under the external field. This method will almost be the exact copy of the linearization of the one-ion problem. We start with the equations (93) or (94)

$$(\dot{A}^{\zeta}{}_{k})_{h} = -\sum_{\alpha,\gamma} f_{\alpha\zeta} \,^{\gamma} h_{k}^{\alpha} A_{k}^{\gamma}, \tag{114}$$

$$(\underline{\dot{A}}_k)_h = -\underline{(\mathbf{O}h_k)} \cdot \underline{A}_k.$$
 (115)

As usual we divide our fields to large, static ground state expectation values, and small time dependent fluctuating parts. We concentrate on one Fourier mode of the latter (this will be sufficient for our purposes)¹²:

$$h_k^{\alpha}(t) = \langle h_k^{\alpha} \rangle_0 + \delta h_k^{\alpha}(t) = \langle h_k^{\alpha} \rangle_0 + \delta h_{\mathbf{q},\omega}^{\alpha} e^{-i(\omega t - \mathbf{q}\mathbf{R}_k)}, \tag{116}$$

$$A_k^{\alpha}(t) = \langle A_k^{\alpha} \rangle_0 + \delta A_k^{\alpha}(t) = \langle A_k^{\alpha} \rangle_0 + \delta A_{\mathbf{q},\omega}^{\alpha} e^{-i(\omega t - \mathbf{q}\mathbf{R}_k)}, \qquad (117)$$

please note, that we let the ground state expectation values vary spatially. The spatiotemporal variations of the oscillations clearly form a well defined wave propagating in the $+\mathbf{q}$ direction. Since our first goal is to calculate the dispersion relations $\omega(\mathbf{q})$, i.e. the wave vector specifies the frequency (up to a branch index, of course). So the above notation is redundant, the index ω is unnecessary, so we will omit it. Let us suppose that the ordering forms a well defined pattern, i.e. the ground state's Fourier transform only consists of a *finite* –usually very small– set of amplitudes with ordering vectors \mathbf{Q}_l . With these at hand we have (here all the constants are correct):

$$h_k^{\alpha}(t) = \langle h_k^{\alpha} \rangle_0 + \delta h_k^{\alpha}(t) = \frac{1}{\sqrt{N}} \sum_{\mathbf{Q}_l} \left\langle h_{\mathbf{Q}_l}^{\alpha} \right\rangle_0 e^{+i(\mathbf{Q}_l \mathbf{R}_k)} + \frac{1}{\sqrt{N}} \sum_{\mathbf{q} \in \text{B.Z.}} \delta h_{\mathbf{q}}^{\alpha} e^{-i(\omega t - \mathbf{q} \mathbf{R}_k)}, (118)$$

$$A_k^{\alpha}(t) = \langle A_k^{\alpha} \rangle_0 + \delta A_k^{\alpha}(t) = \frac{1}{\sqrt{N}} \sum_{\mathbf{Q}_l} \left\langle A_{\mathbf{Q}_l}^{\alpha} \right\rangle_0 e^{+i(\mathbf{Q}_l \mathbf{R}_k)} + \frac{1}{\sqrt{N}} \sum_{\mathbf{q} \in \mathrm{B.Z.}} \delta A_{\mathbf{q}}^{\alpha} e^{-i(\omega t - \mathbf{q} \mathbf{R}_k)}, (119)$$

 $^{^{12}\}mathrm{For}$ more about Fourier transforms c.f. next subsection.

where N is the number of lattice points, and the wave-vector summation is over the ordering vectors, or runs over the Brillouin zone. In what follows we omit the sums, and only retain them at the end of the calculations. We copy the method of the on-site problem to calculate the linearized EOM: we substitute the Ansätze (119) in (115), us the non-evolving nature of the ground state (i.e. if in a product two ground state expectation values meet we drop them, this is exact), and the linearization consists of neglecting the double- δ terms. All these yield to (we introduced a dissipation term again, and use vectorial notation):

$$(\underline{\dot{\delta A}}_k)_h = -\left\{ \underbrace{(\underline{\bullet} \langle h_k \rangle_0)}_{\underline{\bullet}} + \underline{\underline{\Gamma}} \right\} \cdot \underline{\delta A}_k - \underbrace{(\underline{\bullet} \langle A_k \rangle_0)}_{\underline{\bullet}} \cdot \underline{\delta h}_k.$$
(120)

We turn to the part of the EOM which originates in the exchange interaction. As always we drop the terms where two expectation values multiply each other (being exactly zero), and neglect the double- δ terms. We start with (111):

$$(\underline{\dot{A}}_{k})_{J} = -\left(\bigoplus \left(\sum_{i \neq k} \underbrace{\widetilde{\underline{J}}_{ki}}_{i \neq k} \cdot \underline{A}_{i} \right) \right) \cdot \underline{A}_{k}, \quad (121)$$

$$(\underline{\delta \dot{A}}_{k})_{J} \approx -\underbrace{\left(\bigodot\left(\sum_{i \neq k} \underline{\widetilde{J}}_{ki} \cdot \underline{\delta A}_{i}\right)\right)}_{\underline{I}} \cdot \langle \underline{A}_{k} \rangle_{0} - \underbrace{\left(\bigodot\left(\sum_{i \neq k} \underline{\widetilde{J}}_{ki} \cdot \langle \underline{A}_{i} \rangle_{0}\right)\right)}_{\underline{I}} \cdot \underline{\delta A}_{k}.$$
(122)

The second sum is easy to interpret: it is just the effect of the ground state expectation value of the effective field:

$$-\underbrace{\left(\mathbf{O}\left(\sum_{i\neq k}\widetilde{\underline{\underline{J}}}_{ki}\cdot\langle\underline{A}_{i}\rangle_{0}\right)\right)}_{\underline{\underline{J}}}\cdot\underline{\underline{\delta}A}_{k} = -\sum_{i\neq k}\underbrace{\left(\mathbf{O}\left(\widetilde{\underline{\underline{J}}}_{ki}\cdot\langle\underline{A}_{i}\rangle_{0}\right)\right)}_{\underline{\underline{J}}}\cdot\underline{\underline{\delta}A}_{k} =$$
(123)

$$= -\underbrace{\left(\bigotimes \langle \underline{h}_k^{\text{eff}} \rangle_0 \right)}_{\underline{}} \cdot \underline{\delta A}_i. \tag{124}$$

In the first sum of Eq. (122) we use the usual trick, to interchange the argument of star and the last variable:

$$-\underbrace{\left(\bigotimes\left(\sum_{i\neq k} \underline{\widetilde{\underline{J}}}_{ki} \cdot \underline{\delta}\underline{A}_i\right) \right)}_{\underline{i\neq k}} \cdot \langle \underline{\underline{A}}_k \rangle_0 = -\sum_{i\neq k} \left(\underline{\underline{\widetilde{\underline{J}}}}_{ki} \cdot \underline{\underline{(\bigotimes\langle\underline{A}_k\rangle_0)}} \right) \cdot \underline{\delta}\underline{A}_i.$$
(125)

Rewriting the linearized evolution under the exchange (122) with (124) and (125) yields:

$$(\underline{\delta \dot{A}}_{k})_{J} = -\sum_{i \neq k} \underbrace{\left(\underbrace{\widetilde{\underline{J}}_{ki}}_{\underline{\underline{J}}_{ki}} \cdot \langle \underline{A}_{i} \rangle_{0} \right) \right)}_{i \neq k} \cdot \underline{\delta A}_{k} - \sum_{i \neq k} \left(\underbrace{\widetilde{\underline{J}}_{ki}}_{\underline{\underline{J}}_{ki}} \cdot \underline{\underbrace{(\underline{\delta} \langle \underline{A}_{k} \rangle_{0})}_{0} \right) \cdot \underline{\delta A}_{i}.$$
(126)

This equation together with evolution under the external field Eq. (120) (without the dissipation term) yields to the *full linearized equation of motion of the general Hamiltonian*

in real space:

$$= -\underline{(\textcircled{O}\langle h_k \rangle_0)} \cdot \underline{\delta A}_k - \sum_{i \neq k} \underline{(\textcircled{O}\left(\underline{\tilde{J}}_{ki} \cdot \langle \underline{A}_i \rangle_0\right))} \cdot \underline{\delta A}_k - \sum_{i \neq k} \underline{(\textcircled{O}\left(\underline{\tilde{J}}_{ki} \cdot \langle \underline{A}_i \rangle_0\right))} \cdot \underline{\delta A}_k - \sum_{i \neq k} \underline{(\textcircled{O}\langle \underline{A}_k \rangle_0)} \cdot \underline{\delta A}_i - (128) - \underline{(\textcircled{O}\langle A_k \rangle_0)} \cdot \underline{\delta h}_k.$$
(129)

Please note that we have not used the harmonic Ansätze (119) yet. Before we try to solve this model in general (with the harmonic Ansätze) we illustrate the technique that will be used by the simple example of isotropic Heisenberg magnets.

2.4.5 Lattice Spin Models: The Heisenberg Ferromagnet and Antiferromagnet

Before turning to solving the general linearized lattice model let us very briefly discuss the simple isotropic ferro- and antiferromagnetic Heisenberg models. This is a warm up subsection, where we calculate some well known dispersion relations. Of course this subject has a vast literature. The usual technique used to solve the Heisenberg model is quantum mechanical bosonization. A version of this technique (the so-called linearized Holstein-Primakoff transformation) is used in Chapter 6. of the excellent textbook [6], and in chapter 15. of [28], where the analogues of our quasiclassical spin wave calculations are present, too. In Chapter 12. [14] a simplified version of the classical spin wave calculation is given. In [5] a very pictorial description is given on spin waves in ferroand antiferromagnetic materials. Chapter 4. of [13] uses the bosonization technique to derive the spin wave dispersion, but gives the quasiclassical description of a ferromagnet as a (solved) exercise (Chapter 4. Exercise 6.). As far as I know one the first articles on the bosonization technique of ferromagnetic spin waves is [9], where the first published presentation of the derivation of the quasiclassical equations based on a quantum mechanical arguments is present. We turn to our exposition of solving for the dispersion relation of Heisenberg magnets.

Here the Hamiltonian is of the form:

$$H = \frac{1}{2} \sum_{ij} J_{ij} \underline{S}_i^T \cdot \underline{S}_j, \qquad (130)$$

where depending on the sign of the exchange constant(s) the model is either ferro- (negative) or antiferromagnetic (positive). Here we suppose simple, collinear orderings (although there exist counterexamples, i.e. spiral or conical orders, and our method is capable of handling them, but we want to keep this section as simple as possible) with a single ordering vector \mathbf{Q} , i.e.

$$\langle \underline{S}_i \rangle_0 = \underline{S}_0 e^{i \mathbf{Q} \mathbf{R}_i},\tag{131}$$

where $\underline{S}_0 = (0, 0, S)^T$, and in the ferromagnetic case $\mathbf{Q} = 0$ (equitranslational order), and in the antiferromagnetic case on some lattice points the spin points up, on the others down (so \mathbf{Q} is chosen to be in some special point in the Brillouin zone). Since we are using the simple spin algebra the linearized EOM-s (129) simplify considerably (also we do not have an external field):

$$(\underline{\delta S}_i) = \sum_j J_{ij} \left(-\langle \underline{S}_j \rangle_0 \times \underline{\delta S}_i + \langle \underline{S}_i \rangle_0 \times \underline{\delta S}_j \right).$$
(132)

Note that we retained the more familiar cross-product notation, instead of the starnotation. Substituting the ground states, and multiplying by $e^{-i\mathbf{QR}_i}$ yields:

$$(\underline{\delta S}_i)e^{-i\mathbf{QR}_i} = \sum_j \left(-J_{ij}e^{i\mathbf{Q}(\mathbf{R}_j - \mathbf{R}_i)}\underline{S}_0 \times \underline{\delta S}_i + J_{ij}\underline{S}_0 \times \underline{\delta S}_j \right).$$
(133)

If we suppose harmonic spacetime dependence of the fluctuating parts (we suppose that only one Fourier mode is excited):

$$\underline{\delta S}_i = \underline{\delta S}_{\mathbf{q}} e^{-i\omega t} e^{i\mathbf{q}\mathbf{R}_i},\tag{134}$$

and substitute this into Eq. (133) we arrive at the equation:

$$-i\omega\underline{\delta S}_{\mathbf{q}}e^{i(\mathbf{q}-\mathbf{Q})\mathbf{R}_{i}} = \underline{S}_{0} \times \underline{\delta S}_{\mathbf{q}} \sum_{j} \left(-J_{ij}e^{i(\mathbf{Q}(\mathbf{R}_{j}-\mathbf{R}_{i})+\mathbf{q}\mathbf{R}_{i}))} + J_{ij}e^{i\mathbf{q}\mathbf{R}_{j}} \right).$$
(135)

Let us rename our variables and define the Fourier transform of the exchange constants:

$$\mathbf{R}_j - \mathbf{R}_i = \Delta \mathbf{R},\tag{136}$$

$$\mathbf{R}_j = \mathbf{R}_i + \Delta \mathbf{R},\tag{137}$$

$$\sum_{\Delta \mathbf{R}} J(\Delta \mathbf{R}) e^{i\mathbf{q}\Delta \mathbf{R}} = J(\mathbf{q}).$$
(138)

With these definitions the EOM becomes:

$$-i\omega\underline{\delta S}_{\mathbf{q}}e^{i(\mathbf{q}-\mathbf{Q})\mathbf{R}_{i}} = \underline{S}_{0} \times \underline{\delta S}_{\mathbf{q}}e^{i\mathbf{q}\mathbf{R}_{i}} \sum_{\Delta \mathbf{R}} \left(-J(\Delta \mathbf{R})e^{i\mathbf{Q}\Delta \mathbf{R}} + J(\Delta \mathbf{R})e^{i\mathbf{q}\Delta \mathbf{R}}\right), \quad (139)$$

$$-i\omega\underline{\delta S}_{\mathbf{q}}e^{i(\mathbf{q}-\mathbf{Q})\mathbf{R}_{i}} = \underline{S}_{0} \times \underline{\delta S}_{\mathbf{q}}e^{i\mathbf{q}\mathbf{R}_{i}}\left(-J(\mathbf{Q})+J(\mathbf{q})\right).$$
(140)

Using the facts

$$\underline{\delta S}_{i} = \underline{\delta S}_{\mathbf{q}} e^{-i\omega t} e^{i\mathbf{q}\mathbf{R}_{i}} = \underline{\delta S}_{\mathbf{q}+\mathbf{Q}} e^{-i\omega t} e^{i(\mathbf{q}+\mathbf{Q})\mathbf{R}_{i}},\tag{141}$$

$$\underline{\delta S}_{\mathbf{q}} e^{i(\mathbf{q}-\mathbf{Q})\mathbf{R}_i} = \underline{\delta S}_{\mathbf{q}+\mathbf{Q}} e^{i\mathbf{q}\mathbf{R}_i}, \qquad (142)$$

we can rewrite the EOM:

$$-i\omega\underline{\delta S}_{\mathbf{q}+\mathbf{Q}} = \underline{S}_0 \times \underline{\delta S}_{\mathbf{q}} \left(-J(\mathbf{Q}) + J(\mathbf{q}) \right), \tag{143}$$

$$-i\omega\underline{\delta S}_{\mathbf{q}+\mathbf{Q}} = (J(\mathbf{q}) - J(\mathbf{Q}))(\underline{\bigstar S}_0) \cdot \underline{\delta S}_{\mathbf{q}}.$$
(144)

Several interesting things have happened. The first thing is that the time evolution of the components $\underline{\delta S}_{\mathbf{q}+\mathbf{Q}}$ and $\underline{\delta S}_{\mathbf{q}}$ is related. This is a consequence of the character of the

ordering. With nonzero ordering vector the resulting ground state pattern enlarges the unit cell (this is usually called the magnetic unit cell), hence shrinks the Brillouin zone, so formerly inequivalent wave vectors become equivalent. Our equation is true even in the larger, chemical Brillouin zone. In the chemical zone, for simple orderings supposed here, we can surely state that the vectors $\delta S_{\mathbf{q}+2\mathbf{Q}} = \delta S_{\mathbf{q}}$, since 2**Q** is supposed to be reciprocal lattice vector in the original Brillouin zone (we supposed a collinear, simple two sublattice antiferromagnet, or the even more simple ferromagnet). We solve the equations (144) for a ferromagnet and for a simple antiferromagnet.

In the ferromagnet the magnetic and chemical Brillouin zones are the same. So in Eq. (144) $\underline{\delta S}_{\mathbf{q}+\mathbf{Q}} = \underline{\delta S}_{\mathbf{q}}$, since $\mathbf{Q} = 0$. The eigenvalues of the dynamical matrix

$$\underline{\underline{\Omega}}(\mathbf{q}) = -i \left(J(0) - J(\mathbf{q}) \right) \underline{(\bigstar S_0)}.$$
(145)

are $\omega_{1,2} = \pm S (J(0) - J(\mathbf{q}))$, $\omega_0 = 0$, as usual. There is only one propagating circular precession about the effective field vector, and in the direction of the effective field, there is no motion, in perfect analogy with the one-spin problem. Fig. 7. shows the dispersion for a 1D ferromagnet, with only nearest neighbor interactions. It starts quadratically, and at the zone center the frequency vanishes. This last property is the consequence of the well known *Goldstone theorem*.



Figure 7: Dispersion relation for a 1D ferromagnet, with nearest neighbor interactions only, and without any external field, throughout the Brillouin zone. The plot is normalized to have saturation value of the frequency 1, and the lattice spacing is set to 1.

In the antiferromagnet the magnetic and chemical Brillouin zones are not the same. So in Eq. (144) only we know is $\delta S_{\mathbf{q}+2\mathbf{Q}} = \delta S_{\mathbf{q}}$, since $\mathbf{Q} \neq 0$, but we suppose a simple two-sublattice ordering. In Eq. (144) we substitute $\mathbf{q} \rightarrow \mathbf{q} + \mathbf{Q}$ to get:

$$-i\omega\underline{\delta S}_{\mathbf{q}} = -i\omega\underline{\delta S}_{\mathbf{q}+2\mathbf{Q}} = (J(\mathbf{q}+\mathbf{Q}) - J(\mathbf{Q}))\underline{(\bigstar S_0)} \cdot \underline{\delta S}_{\mathbf{q}+\mathbf{Q}}.$$
 (146)

We substitute this expression for $\underline{\delta S}_{\mathbf{q}}$ back in Eq. (144), and we arrive at:

$$-\omega^2 \underline{\delta S}_{\mathbf{q}} = (J(\mathbf{q}) - J(\mathbf{Q})) \left(J(\mathbf{q} + \mathbf{Q}) - J(\mathbf{Q}) \right) \underbrace{\left(\bigstar S_0 \right)^2}_{\underline{\mathbf{m}}} \cdot \underline{\delta S}_{\mathbf{q}}.$$
 (147)

And with this we arrive at the expression

$$\omega_{1,2} = \pm S \sqrt{\left(J(\mathbf{q}) - J(\mathbf{Q})\right) \left(J(\mathbf{q} + \mathbf{Q}) - J(\mathbf{Q})\right)},$$

and a zero mode again. From the expression of the frequency it is obvious that Goldstone's theorem is fulfilled, and we have two degenerate branches of excitations, jut like in the two-spin problem: the spins on the two sublattices precess in the same direction, on unequal size circles, and the precession propagates. The degeneracy means that we have two options for choosing on which sublattice the larger circles should be. Fig. 8. shows the dispersion relation for a 1D antiferromagnet, with nearest neighbor interactions only. It is clearly seen, that the dispersion starts linearly. We turn to the discussion of our results on simple Heisenberg magnets. Here we rederived the well known dispersion relations of



Figure 8: Dispersion relation for a 1D antiferromagnet, with nearest neighbor interactions only, and without any external field, throughout the magnetic Brillouin zone. The plot is normalized to have saturation value of the frequency 1, and the lattice spacing is set to 1.

classical Heisenberg models, with simple, collinear, commensurate orderings. Our result agree with the literature, e.g. c.f. Chapter 15. of $[28]^{13}$, or Chapter 6. of [6].

Although we only used our method for simple collinear, commensurate orderings, the model is capable of handling much more complicated problems. In the articles [15] and [16] the authors described two models, from which the second one is much more interesting. In this $J_1 - J_2$ model they postulated a Heisenberg spin chain with nearest and next nearest isotropic exchange in a magnetic field. The exchange constants are chosen to result in a spiral (without field) and conical order (with field), with an incommensurate ordering vector. The authors calculated the dispersions with a trick of using local coordinates, in which the z-axis on each site always points in the effective field direction. With

¹³There is a mismatch between this book's results and of ours. In the antiferromagnetic dispersion a factor of 2 appears in the book. This may be a consequence of the definition of the interaction Hamiltonian. Another mismatch is the sign of the ordering vectors, this may be a consequence of a definition, or I have made a mistake. In simple cases both results are the same.

this technique they essentially reduced the lattice problem to a wave-vector dependent, anisotropic one-site problem. The interesting point is that with field the dispersion are not even in q-space: i.e. $\omega(q) \neq \omega(-q)$. We would like to mention that our EOM method (with the proper modifications) is capable of reproducing these results. Since we would like to publish these results elsewhere we do not go into the details of the modification of the method (which is based on choosing proper local coordinates just like in the original article), just show the results in Fig. 9. Our results are in exact match with that of [16] These calculations were done with the notebook miyahara_repr.nb.



Figure 9: Dispersion relation for a magnet with incommensurate, conical order, with isotropic nearest and next nearest neighbor interactions in a magnetic field, throughout the chemical Brillouin zone. Left panel: our results with the quasiclassical approach. Right panel: results of [16] where they used linearized spin wave theory.

As a conclusion: with the EOM method we are able to calculate the dispersion relations of even complicated spin models, without the use of bosonization techniques. Next we turn to to solve the general linearized model (129).

2.4.6 Solving the Linearized Model

We substitute the harmonic dependencies (119) into the linearized EOM (129), but we omit the $1/\sqrt{N}$ prefactors, and suppose that we only have one ordering vector, and name it as **Q**, also we set the external fields equal to 0 (they can easily be reintroduced in the end). So the EOM becomes:

$$-i\omega(\mathbf{q})\underline{\delta A}_{\mathbf{q}}e^{i\mathbf{q}\mathbf{R}_{k}} = -\sum_{i\neq k} \underbrace{\left(\mathbf{\mathfrak{O}}\left(\underline{\widetilde{J}}_{ki}\cdot\left\langle\underline{A}_{\mathbf{Q}}\right\rangle_{0}\right)\right)}_{\underline{\widetilde{J}}_{ki}} \cdot \underline{\delta A}_{\mathbf{q}}e^{i(\mathbf{Q}\mathbf{R}_{i}+\mathbf{q}\mathbf{R}_{k})} - (148)$$

$$-\sum_{i\neq k} \left(\underbrace{\widetilde{\underline{J}}}_{ki} \cdot \underbrace{\left(\underbrace{\mathfrak{B}} \langle \underline{A}_{\mathbf{Q}} \rangle_{0} \right)}_{\mathbf{Q}} \right) \cdot \underline{\delta A}_{\mathbf{q}} e^{i(\mathbf{Q}\mathbf{R}_{k} + \mathbf{q}\mathbf{R}_{i})}.$$
(149)

As we have done to the spin-lattice models we multiply the equation by $e^{-i\mathbf{QR}_k}$: and define $\Delta \mathbf{R} = (\mathbf{R}_i - \mathbf{R}_k)$ and replace $\mathbf{R}_i = (\Delta \mathbf{R} + \mathbf{R}_k)$, and define the Fourier transform

of the exchange constant as:

$$\widetilde{\underline{J}}_{\mathbf{Q}} = \sum_{\Delta \mathbf{R}} \widetilde{\underline{\underline{J}}}_{\underline{\mathbf{I}}} (\Delta \mathbf{R}) e^{+i(\mathbf{Q}\Delta \mathbf{R})}$$
(150)

again. After some algebra we arrive at the expression (with a replacement of $\underline{\delta A}_{\mathbf{q}} \rightarrow \underline{\delta A}_{\mathbf{q}+\mathbf{Q}}$ after absorbing an appropriate exponential factor):

$$-i\omega(\mathbf{q})\underline{\delta A}_{\mathbf{q}+\mathbf{Q}} = -\left\{\underline{\left(\mathbf{\mathfrak{O}}\left(\underline{\widetilde{J}}_{\mathbf{Q}}\cdot\langle\underline{A}_{\mathbf{Q}}\rangle_{0}\right)\right)} + \left(\underline{\widetilde{J}}_{\mathbf{q}}\cdot\underline{\left(\mathbf{\mathfrak{O}}\langle\underline{A}_{\mathbf{Q}}\rangle_{0}\right)}\right)\right\}\cdot\underline{\delta A}_{\mathbf{q}}.$$
 (151)

This is the exact analogue of Eq. (144) for multipolar systems. And again, if we suppose that the ordering is simple enough (i.e. $\mathbf{Q} = 0$ for ferro-ordering or $\mathbf{Q} \neq 0$, but is in a special point, so $\underline{\delta A}_{\mathbf{q}+2\mathbf{Q}} = \underline{\delta A}_{\mathbf{q}}$)¹⁴ we can solve this equation. We start with the ferromagnet, just like before for the simple spin systems. For the ferro-ordering $\mathbf{Q} = 0$, and substituting this into (151) yields:

$$-i\omega(\mathbf{q})\underline{\delta A}_{\mathbf{q}} = -\left\{ \underbrace{\left(\mathbf{O}\left(\underbrace{\widetilde{\underline{J}}}_{\mathbf{q}=\mathbf{0}} \cdot \langle \underline{A}_{\mathbf{q}=\mathbf{0}} \rangle_{0} \right) \right)}_{\mathbf{q}} + \left(\underbrace{\widetilde{\underline{J}}}_{\mathbf{q}} \cdot \underbrace{\left(\mathbf{O}\left\langle \underline{A}_{\mathbf{q}=\mathbf{0}} \rangle_{0} \right)}_{\mathbf{q}} \right) \right\} \cdot \underline{\delta A}_{\mathbf{q}}.$$
(152)

From which the dispersion relations for a ferromultipolar material are the eigenvalues of the dynamical matrix:

$$\underline{\underline{\Omega}}(\mathbf{q}) = +i\left\{\underline{\left(\underline{\mathbf{O}}\left(\underline{\widetilde{J}}_{\mathbf{q}=\mathbf{0}} \cdot \langle \underline{A}_{\mathbf{q}=\mathbf{0}} \rangle_{0}\right)\right)} + \left(\underline{\widetilde{J}}_{\mathbf{q}} \cdot \underline{\left(\underline{\mathbf{O}}\langle \underline{A}_{\mathbf{q}=\mathbf{0}} \rangle_{0}\right)}\right)\right\}.$$
(153)

For the antiferro-ordered system we repeat the trick we used for the antiferromagnetic Heisenberg model: we substitute $\mathbf{q} \to \mathbf{q} + \mathbf{Q}$ in Eq. (151), use the facts $\underline{\delta A}_{\mathbf{q}+2\mathbf{Q}} = \underline{\delta A}_{\mathbf{q}}$, and $\omega(\mathbf{q}) = \omega(\mathbf{q} + \mathbf{Q})$:

$$-i\omega(\mathbf{q})\underline{\delta A}_{\mathbf{q}+2\mathbf{Q}} = -\left\{\underline{\left(\mathbf{\mathfrak{Q}}\left(\underline{\widetilde{J}}_{\mathbf{Q}}\cdot\left\langle\underline{A}_{\mathbf{Q}}\right\rangle_{0}\right)\right)} + \left(\underline{\widetilde{J}}_{\mathbf{q}+\mathbf{Q}}\cdot\underline{\left(\mathbf{\mathfrak{Q}}\left\langle\underline{A}_{\mathbf{Q}}\right\rangle_{0}\right)}\right)\right\}\cdot\underline{\delta A}_{\mathbf{q}+\mathbf{Q}}.$$
 (154)

We substitute the last equation in (151), and conclude that the square roots of the eigenvalues of the dynamical matrix:

$$\underline{\underline{\Omega}}(\mathbf{q})^2 = -\left\{ \underbrace{\left(\underbrace{\mathbf{O}}\left(\underline{\underline{\tilde{J}}}_{\mathbf{Q}} \cdot \langle \underline{A}_{\mathbf{Q}} \rangle_0 \right) \right)}_{\mathbf{Q}} + \left(\underline{\underline{\tilde{J}}}_{\mathbf{q}} \cdot \underbrace{\left(\underbrace{\mathbf{O}}\langle \underline{A}_{\mathbf{Q}} \rangle_0 \right)}_{\mathbf{Q}} \right) \right\} \cdot$$
(155)

$$\cdot \left\{ \underbrace{\left(\mathbf{\mathfrak{O}}\left(\underbrace{\widetilde{J}}_{\mathbf{Q}} \cdot \langle \underline{A}_{\mathbf{Q}} \rangle_{0} \right) \right)}_{\mathbf{\mathbf{I}}} + \left(\underbrace{\widetilde{J}}_{\mathbf{q}+\mathbf{Q}} \cdot \underbrace{\left(\mathbf{\mathfrak{O}}\langle \underline{A}_{\mathbf{Q}} \rangle_{0} \right)}_{\mathbf{\mathbf{I}}} \right) \right\},$$
(156)

are the dispersion relations. If an external field is present the modifications are trivial: we simply have to add $\langle \underline{h}_{\mathbf{Q}} \rangle_0$ to the effective field $\langle \underline{h}_{\mathbf{Q}}^{\text{eff}} \rangle_0 = \underline{J}_{\mathbf{Q}} \cdot \langle \underline{A}_{\mathbf{Q}} \rangle_0$ under the \mathfrak{Q} We close this subsection with a conclusion.

¹⁴If this conditions do not hold, but there is some integer multiple of the ordering vector which is a reciprocal lattice vector in the chemical reciprocal space, than the modification of our arguments in the solution of this equation is almost trivial.

What we achieved here is the solution for the eigenfrequencies for the general multipolar lattice Hamiltonian by the linearized EOM method. Although only for simple orderings. Unfortunately we have not used this formalism to any real multipolar lattice problem, so we cannot give an example here. As always in a homogeneous lattice problem, it should be worth to transform our equations to Fourier space. In the following we derive the Fourier transform of the lattice problem, and give its general solution. At first fix our conventions of the transformations.

2.4.7 The Model in Fourier Space

The direct and inverse transforms read as:

$$A^{\alpha}_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_{i} e^{-i\mathbf{q}\mathbf{R}_{i}} A^{\alpha}_{i}, \qquad (157)$$

$$A_i^{\alpha} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q} \in \mathbf{B.Z.}} e^{i\mathbf{q}\mathbf{R}_i} A_{\mathbf{q}}^{\alpha}, \tag{158}$$

where N is the number of lattice points, the real space summation runs over the lattice, and the wave-vector summation in the Brillouin zone. The commutation relations in Fourier space take the form of (using (87) with the expansion (158)):

$$\begin{bmatrix} A_{\mathbf{q}}^{\alpha}, A_{\mathbf{q}'}^{\beta} \end{bmatrix} = \frac{1}{N} \sum_{ij} e^{-i\mathbf{q}\mathbf{R}_{i}} e^{-i\mathbf{q}'\mathbf{R}_{j}} \begin{bmatrix} A_{i}^{\alpha}, A_{j}^{\beta} \end{bmatrix} = \frac{i}{N} \sum_{ij} e^{-i\mathbf{q}\mathbf{R}_{i}} e^{-i\mathbf{q}'\mathbf{R}_{j}} \delta_{ij} \sum_{\gamma} f_{\alpha\beta}^{\gamma} A_{j}^{\gamma} = (159)$$
$$= \frac{i}{N} \sum_{i,\gamma} e^{-i(\mathbf{q}+\mathbf{q}')\mathbf{R}_{i}} f_{\alpha\beta}^{\gamma} A_{i}^{\gamma} = \frac{i}{\sqrt{N}} \sum_{\gamma} f_{\alpha\beta}^{\gamma} \underbrace{\frac{1}{\sqrt{N}} \sum_{i} e^{-i(\mathbf{q}+\mathbf{q}')\mathbf{R}_{i}} A_{i}^{\gamma}}_{\mathbf{q}+\mathbf{q}'} (160)}_{A_{\mathbf{q}}^{\gamma}, A_{\mathbf{q}'}^{\gamma}} = i \frac{1}{\sqrt{N}} \sum_{\gamma} f_{\alpha\beta}^{\gamma} A_{\mathbf{q}+\mathbf{q}'}^{\gamma} (161)$$

Let us turn to the Fourier transform of the Hamiltonian (85). We start with the easier part H^h :

$$H^{h} = \sum_{i,\beta} h_{i}^{\beta} A_{i}^{\beta} = \frac{1}{N} \sum_{\mathbf{q}\mathbf{q}'} \sum_{i,\beta} e^{i(\mathbf{q}+\mathbf{q}')\mathbf{R}_{i}} h_{\mathbf{q}}^{\beta} A_{\mathbf{q}'}^{\beta} = \sum_{\beta} \sum_{\mathbf{q}\mathbf{q}'} h_{\mathbf{q}}^{\beta} A_{\mathbf{q}'}^{\beta} \frac{1}{N} \underbrace{\sum_{i} e^{i(\mathbf{q}+\mathbf{q}')\mathbf{R}_{i}}}_{N\delta_{\mathbf{q}+\mathbf{q}',\mathbf{0}}}, \quad (162)$$
$$H^{h} = \sum_{\mathbf{q},\beta} h_{\mathbf{q}}^{\beta} A_{-\mathbf{q}}^{\beta}, \quad (163)$$

where in the last part of the first line we used the lattice sum. Next we calculate the exchange part H^J of the Hamiltonian (85) in Fourier space:

$$H^{J} = \frac{1}{2} \sum_{ij,\alpha\beta} J^{\alpha\beta}_{ij} A^{\alpha}_{i} A^{\beta}_{j} = \frac{1}{2} \frac{1}{N} \sum_{\mathbf{q}\mathbf{q}'} \sum_{ij,\alpha\beta} J^{\alpha\beta}_{ij} e^{i(\mathbf{q}\mathbf{R}_{i}+\mathbf{q}'\mathbf{R}_{j})} A^{\alpha}_{\mathbf{q}} A^{\beta}_{\mathbf{q}'} =$$
(164)

$$=\frac{1}{2}\frac{1}{N}\sum_{\alpha\beta}\sum_{\mathbf{q}\mathbf{q}'}\sum_{\underline{\Delta\mathbf{R}}}J^{\alpha\beta}(\Delta\mathbf{R})e^{-i\mathbf{q}\Delta\mathbf{R}}A^{\alpha}_{\mathbf{q}}A^{\beta}_{\mathbf{q}'}\sum_{\underline{\mathbf{R}}_{i}}e^{i(\mathbf{q}+\mathbf{q}')\mathbf{R}_{i}},\qquad(165)$$

$$H^{J} = \frac{1}{2} \sum_{\mathbf{q},\alpha\beta} J^{\alpha\beta}_{\mathbf{q}} A^{\alpha}_{\mathbf{q}} A^{\beta}_{-\mathbf{q}}, \qquad (166)$$

where we have used the lattice sum, and defined the Fourier transform of the exchange matrix:

$$J_{\mathbf{q}}^{\alpha\beta} = \sum_{\Delta \mathbf{R}} J^{\alpha\beta}(\Delta \mathbf{R}) e^{-i\mathbf{q}\Delta \mathbf{R}},\tag{167}$$

with $\Delta \mathbf{R} = \mathbf{R}_i - \mathbf{R}_j$. So the full Hamiltonian in Fourier space looks like:

$$H = H^{J} + H^{h} = \frac{1}{2} \sum_{\mathbf{q},\alpha\beta} J^{\alpha\beta}_{\mathbf{q}} A^{\alpha}_{\mathbf{q}} A^{\beta}_{-\mathbf{q}} + \sum_{\mathbf{q},\beta} h^{\beta}_{\mathbf{q}} A^{\beta}_{-\mathbf{q}}.$$
 (168)

Here we recall some properties of the Fourier transforms which will be helpful later. These properties can easily be shown based on the definitions (158) and (167). If our quantum fields are self-adjoint, or our classical fields (i.e. the ground state expectation values of the quantum fields) are real, then we have:

$$A^{\alpha}_{\mathbf{q}} = A^{\alpha\dagger}_{-\mathbf{q}},\tag{169}$$

in the quantum case, and

$$A^{\alpha}_{\mathbf{q}} = A^{\alpha*}_{-\mathbf{q}},\tag{170}$$

in the classical case. If the configuration is real even in real space, then it is real and even in Fourier space, i.e. we can omit the conjugation mark. As for the exchange matrix: if it is real (as always) and even (odd) in real space, then it is real and even (odd) in Fourier space. Next we turn to the EOM-s in Fourier space.

Again we start with the easier part H^h of Eq. (163). Using the commutation relations Eq. (161) and the EOM as applied to the Fourier component $A_{\mathbf{q}'}^{\zeta}$ we have:

$$(\dot{A}_{\mathbf{q}'}^{\zeta})_{h} = i \left[H^{h}, A_{\mathbf{q}'}^{\zeta} \right] = i \sum_{\mathbf{q},\beta} h_{\mathbf{q}}^{\beta} \left[A_{-\mathbf{q}}^{\beta}, A_{\mathbf{q}'}^{\zeta} \right] = -\frac{1}{\sqrt{N}} \sum_{\mathbf{q},\gamma\beta} f_{\beta\zeta}^{\gamma} h_{\mathbf{q}}^{\beta} A_{\mathbf{q}'-\mathbf{q}}^{\gamma}.$$
(171)

This equation has a structure. With the aid of the star-notation Eq. (76) we can simplify it, just like in the single site case (since there are no interactions in this part of the Hamiltonian this is essentially a one-site problem). The β -sum is just the definition of the \bigcirc of the field vector, and the γ -sum is just a simple matrix product of the associated matrix and the "spin" vector. So this equation becomes in vectorial notation:

$$\left(\underline{\dot{A}}_{\mathbf{q}'}\right)_{h} = -\frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \underbrace{\left(\mathbf{\textcircled{O}}_{h\mathbf{q}}\right)}_{\mathbf{q}} \cdot \underline{A}_{\mathbf{q}'-\mathbf{q}}^{\gamma}.$$
(172)

Keeping the full wavevector dependence of the external field may seem to be purely academical, since we cannot really employ a field with high spatial variation, so it is tempting to drop all the terms besides $\underline{h}_{\mathbf{q}=\mathbf{0}}$. And this is absolutely true for the static external field $\underline{h}_{\mathbf{q}}^{0} = \underline{h}_{\mathbf{q}=\mathbf{0}}^{0} := \underline{h}^{0}$. But for calculating the spatial dependence of the susceptibility we have to retain the -small- spatially oscillating part of the field $\underline{\delta}\underline{h}_{\mathbf{q}}^{\omega}$ (in the calculations). But for the spatially homogeneous external field it is true, that:

$$(\underline{A}_{\mathbf{q}'})_{h^0} = -\frac{1}{\sqrt{N}} \underbrace{(\mathbf{O}h_{\mathbf{q}=\mathbf{0}}^0)}_{\mathbf{Q}} \cdot \underline{A}_{\mathbf{q}'}^{\gamma}.$$
(173)

Another caveat: since the generalized fields contain all the effective internal (e.g. anisotropy) fields, it can happen that they are neither zero, nor homogeneous, even in the absence of any external fields!

Next we turn our attention to the exchange part H^J of Eq. (163). Using the commutation relations Eq. (161) again and the EOM as applied to the Fourier component $A_{\mathbf{q}'}^{\zeta}$ we have (with the aid of the very general formula [AB, C] = A[B, C] + [A, C]B of operator products):

$$(\dot{A}_{\mathbf{q}'}^{\zeta})_{J} = i \left[H^{J}, A_{\mathbf{q}'}^{\zeta} \right] = i \frac{1}{2} \sum_{\mathbf{q}, \alpha\beta} J_{\mathbf{q}}^{\alpha\beta} \left[A_{\mathbf{q}}^{\alpha} A_{-\mathbf{q}}^{\beta}, A_{\mathbf{q}'}^{\zeta} \right] =$$
(174)

$$= i\frac{1}{2}\sum_{\mathbf{q},\alpha\beta} J_{\mathbf{q}}^{\alpha\beta} \left(A_{\mathbf{q}}^{\alpha} \left[A_{-\mathbf{q}}^{\beta}, A_{\mathbf{q}'}^{\zeta} \right] + \left[A_{\mathbf{q}}^{\alpha}, A_{\mathbf{q}'}^{\zeta} \right] A_{-\mathbf{q}}^{\beta} \right) =$$
(175)

$$= -\frac{1}{\sqrt{N}} \frac{1}{2} \sum_{\mathbf{q},\alpha\beta} J_{\mathbf{q}}^{\alpha\beta} \sum_{\gamma} \left(f_{\beta\zeta} \,^{\gamma} A_{\mathbf{q}}^{\alpha} A_{-\mathbf{q}+\mathbf{q}'}^{\gamma} + f_{\alpha\zeta} \,^{\gamma} A_{\mathbf{q}+\mathbf{q}'}^{\gamma} A_{-\mathbf{q}}^{\beta} \right). \tag{176}$$

Putting the two equations Eq. (171) and Eq. (176) together to get full time evolution we arrive at:

$$(\dot{A}_{\mathbf{q}'}^{\zeta}) = (\dot{A}_{\mathbf{q}'}^{\zeta})_{J} + (\dot{A}_{\mathbf{q}'}^{\zeta})_{h} = (177)$$

$$= -\frac{1}{\sqrt{N}} \left(\frac{1}{2} \sum_{\mathbf{q},\gamma\alpha\beta} J_{\mathbf{q}}^{\alpha\beta} \left(f_{\beta\zeta}^{\gamma} A_{\mathbf{q}}^{\alpha} A_{-\mathbf{q}+\mathbf{q}'}^{\gamma} + f_{\alpha\zeta}^{\gamma} A_{\mathbf{q}+\mathbf{q}'}^{\gamma} A_{-\mathbf{q}}^{\beta} \right) + \sum_{\mathbf{q},\gamma\beta} f_{\beta\zeta}^{\gamma} h_{\mathbf{q}}^{\beta} A_{-\mathbf{q}+\mathbf{q}'}^{\gamma} \right) (178)$$

this is an exact set of fully nonlinear, ugly, coupled equations, which we are clearly unable to solve. The problem is the nonlinearity of the time evolution of the exchange part. So what we have to do is somehow linearize it based on some physical assumptions. In what follows we try to do this. Concentrating on the exchange part (Eq. (176)) we seek a method to linearize it. So as before let us divide the fields to ground state and small fluctuating parts:

$$A^{\alpha}_{\mathbf{q}} = \left\langle A^{\alpha}_{\mathbf{q}} \right\rangle_0 + \delta A^{\alpha}_{\mathbf{q}}(t), \tag{179}$$

where we suppose that the first part is the ground state expectation value, hence large and time independent, and the second part is small and fluctuating in time. Let us substitute the Ansatz (179) in Eq. (176). The left hand side is easy: only the time evolution of $(\delta A_{\mathbf{q}}^{\zeta})_J$ remains, since the ground state is static (this -mutatis mutandis- remains true for the field evolution part). Of course the right hand side is the tricky one. One we can say for sure is, that if in the operator products an expectation value multiplies another expectation value, and we carefully sum them up, this sum must result zero, since it is just the non-evolving ground state part of the full time derivative. When in a product an expectation value multiplies a fluctuating δ -field we must keep it. And as before, we neglect the small double- δ terms. Similarly do we to the evolution under the field. So what is left for us is to enumerate all the terms where an expectation value meets a δ -term. To summarize the results we have so far let us define the division of the field as:

$$h_{\mathbf{q}}^{\alpha} = \left\langle h_{\mathbf{q}}^{\alpha} \right\rangle_{0} + \delta h_{\mathbf{q}}^{\alpha}(t), \tag{180}$$

or putting together (179) and (180) in vectorial form:

$$\underline{A}_{\mathbf{q}} = \left\langle \underline{A}_{\mathbf{q}} \right\rangle_0 + \underline{\delta A}_{\mathbf{q}}(t), \tag{181}$$

$$\underline{h}_{\mathbf{q}} = \left\langle \underline{h}_{\mathbf{q}} \right\rangle_0 + \underline{\delta h}_{\mathbf{q}}(t). \tag{182}$$

To write down the linearized EOM-s in Fourier space, as always we start with fielddependent part (and do the same dummy-index-exchange trick as in the single site problem):

$$(\delta \dot{A}_{\mathbf{q}'}^{\zeta})_{h} = -\frac{1}{\sqrt{N}} \sum_{\mathbf{q},\gamma\beta} \left(f_{\gamma\zeta}^{\ \beta} \left\langle A_{-\mathbf{q}+\mathbf{q}'}^{\beta} \right\rangle_{0} \delta h_{\mathbf{q}}^{\gamma} + f_{\beta\zeta}^{\ \gamma} \left\langle h_{\mathbf{q}}^{\beta} \right\rangle_{0} \delta A_{-\mathbf{q}+\mathbf{q}'}^{\gamma} \right),$$
(183)

or in vectorial form:

$$\left(\underline{\delta\dot{A}}_{\mathbf{q}'}\right)_{h} = -\frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \left(\underbrace{\left(\underline{\mathfrak{B}} \left\langle A_{-\mathbf{q}+\mathbf{q}'} \right\rangle_{0}\right)}_{\mathbf{q}} \cdot \underline{\delta h}_{\mathbf{q}} + \underbrace{\left(\underline{\mathfrak{B}} \left\langle h_{\mathbf{q}} \right\rangle_{0}\right)}_{\mathbf{q}} \cdot \underline{\delta A}_{-\mathbf{q}+\mathbf{q}'} \right).$$
(184)

Let us mention, that it is *really* reassuring after so many calculations, that these results are consistent with the real space result (74). To show this, multiply Eq. (184) with $\frac{1}{\sqrt{N}}e^{i\mathbf{q'R_i}}$, and sum up for $\mathbf{q'}$. Then on the left hand side simply $(\underline{\delta A_i})_h$ remains, and on the right hand side change the summation variable to $\mathbf{q'} = \mathbf{q} + \Delta \mathbf{q}$ and perform the sums. The result is (74), for the site \mathbf{R}_i .

Let us turn to the hard part, i.e. the linearization of the exchange part of the EOM. for this purpose let us assume, that the ordering is simple, i.e. only for one Fourier component (vector) with the *ordering vector* \mathbf{Q} is nonvanishing (of course with its nonvanishing

partner $-\mathbf{Q}$).¹⁵ So let us suppose, that only the following (real in real space!) ground state expectation values are nonvanishing:

$$\left\langle \underline{A}_{\mathbf{Q}} \right\rangle_{0} \neq \underline{0},$$
 (185)

$$\left\langle \underline{A}_{-\mathbf{Q}} \right\rangle_{0} = \left\langle \underline{A}_{\mathbf{Q}}^{*} \right\rangle_{0} \neq \underline{0},\tag{186}$$

$$\left\langle \underline{A}_{\mathbf{q}} \right\rangle_{0} = \underline{0}, \text{ if } \mathbf{q} \neq \pm \mathbf{Q}.$$
 (187)

Let us concentrate on the last line of Eq. (176), and denote the summands temporarily by:

$$I. = J_{\mathbf{q}}^{\alpha\beta} f_{\beta\zeta} \,^{\gamma} A_{\mathbf{q}}^{\alpha} A_{-\mathbf{q}+\mathbf{q}'}^{\gamma}, \tag{188}$$

$$II. = J_{\mathbf{q}}^{\alpha\beta} f_{\alpha\zeta}^{\ \gamma} A_{\mathbf{q}+\mathbf{q}'}^{\gamma} A_{-\mathbf{q}}^{\beta}.$$
(189)

Clearly for $(\delta \dot{A}_{\mathbf{q}'})_J$ there are only four terms for *I*. and four terms for *II*. that give nonvanishing contributions at linear level, namely:

$$\mathbf{q} = +\mathbf{Q}, \ -\mathbf{q} + \mathbf{q}' = \mathbf{q}' - \mathbf{Q}, \tag{190}$$

$$\mathbf{q} = -\mathbf{Q}, \ -\mathbf{q} + \mathbf{q}' = \mathbf{q}' + \mathbf{Q}, \tag{191}$$

$$-\mathbf{q} + \mathbf{q}' = +\mathbf{Q}, \ \mathbf{q} = \mathbf{q}' - \mathbf{Q}, \tag{192}$$

- $-\mathbf{q} + \mathbf{q}' = -\mathbf{Q}, \ \mathbf{q} = \mathbf{q}' + \mathbf{Q}, \tag{193}$
 - (194)

for I., and

$$\mathbf{q} = +\mathbf{Q}, \ \mathbf{q} + \mathbf{q}' = \mathbf{q}' + \mathbf{Q}, \tag{195}$$

$$\mathbf{q} = -\mathbf{Q}, \ \mathbf{q} + \mathbf{q}' = \mathbf{q}' - \mathbf{Q}, \tag{196}$$

$$\mathbf{q} + \mathbf{q}' = +\mathbf{Q}, \ \mathbf{q} = -\mathbf{q}' + \mathbf{Q}, \tag{197}$$

$$\mathbf{q} + \mathbf{q}' = -\mathbf{Q}, \ \mathbf{q} = -\mathbf{q}' - \mathbf{Q}, \tag{198}$$

(199)

for II. We put all these expressions into the summands (189), and remember that we have already gotten rid of the summation over wavevector \mathbf{q} . For the linearized time evolution of I, we have (without the structure constants explicitly written, and a new notation introduced):

$$[\delta I.] = \left\langle A^{\alpha}_{\mathbf{Q}} \right\rangle_0 \left(J^{\alpha\beta}_{\mathbf{Q}} \delta A^{\gamma}_{\mathbf{q}'-\mathbf{Q}} + J^{\alpha\beta}_{-\mathbf{Q}} \delta A^{\gamma}_{\mathbf{q}'+\mathbf{Q}} \right) +$$
(200)

$$+\left\langle A^{\gamma}_{\mathbf{Q}}\right\rangle_{0} (J^{\alpha\beta}_{\mathbf{q}'-\mathbf{Q}}\delta A^{\alpha}_{\mathbf{q}'-\mathbf{Q}} + J^{\alpha\beta}_{\mathbf{q}'+\mathbf{Q}}\delta A^{\alpha}_{\mathbf{q}'+\mathbf{Q}}).$$
(201)

For the linearized time evolution of II. we have:

$$[\delta II.] = \left\langle A^{\beta}_{\mathbf{Q}} \right\rangle_{0} \left(J^{\alpha\beta}_{\mathbf{Q}} \delta A^{\gamma}_{\mathbf{q}'+\mathbf{Q}} + J^{\alpha\beta}_{-\mathbf{Q}} \delta A^{\gamma}_{\mathbf{q}'-\mathbf{Q}} \right) +$$
(202)

$$+\left\langle A_{\mathbf{Q}}^{\gamma}\right\rangle_{0} (J_{-\mathbf{q}'+\mathbf{Q}}^{\alpha\beta}\delta A_{\mathbf{q}'-\mathbf{Q}}^{\beta}+J_{-\mathbf{q}'-\mathbf{Q}}^{\alpha\beta}\delta A_{\mathbf{q}'+\mathbf{Q}}^{\beta}).$$
(203)

¹⁵It is not a real restriction. If there are several ordering vectors \mathbf{Q}_i present, then we can repeat all the calculations putting all the ordering vectors in the formulae, and summing them up for *i*.

And collecting all these terms together for the EOM of $(\delta \dot{A}_{\mathbf{q}'}^{\zeta})_J$ we arrive at the expression:

$$(\delta \overset{\cdot}{A_{\mathbf{q}'}})_J = -\frac{1}{\sqrt{N}} \frac{1}{2} \sum_{\gamma \alpha \beta} \left\{ f_{\beta \zeta} \,^{\gamma} \left[\delta I. \right] + + f_{\alpha \zeta} \,^{\gamma} \left[\delta II. \right] \right\}.$$
 (204)

So these are the linearized equations of motion for a general multipolar Hamiltonian in Fourier space. They are not very simple, but they surely are linear. As we have not used them in any real world calculation, we cannot tell how easily tractable they are in practical calculations, but we believe that they can reproduce the results of the usual linearized multiboson theories, as every part of their ingredients did. Our work is now to use this theory in real world Hamiltonians. But –hopefully– this will be part of another thesis. We close our work with a conclusion, and enumerate problems we would like to solve in the future with the EOM method.

3 Conclusions

We want to write down some concluding remarks, and point to the possibilities of further work on the topics covered in this thesis. As a conclusion: we wrote down the EOM-s for a large class of multipolar spin models, developed a technique to linearize them in a systematic way, and gave the solutions. We were able to calculate the eigenenergies, and the eigenoscillations of our systems, and calculated the susceptibilities. Our examples included the following problems: single spin in a magnetic field, antiferromagnetic resonance, single spin in a multipolar field, Heisenberg models of ferro- and antiferromagnetism, and we gave the general form of the eigenenergies of the multipolar lattice problem. Our results are compatible with the ones in the literature. But there is much more work to do. Here we mention a few ways to extend our calculations.

The most important is to use the general multipolar model on a real lattice system, and demonstrate that it gives the same results as the linear multiboson theory, which is known to be able to describe the measurements very well. We have to compute the general form of the susceptibility for these models, too (this is just a few steps away). There are a great bunch of real materials for which these calculations could be relevant, so we have to compare our results with real and new measurements. This will finally show if this method is really capable of giving the same result as the powerful multiboson theories. What I would also like to do is to understand the characteristics of the excitations more deeply, and along with this, to understand the selection rules experienced by experimentalists on multipolar materials (i.e. classify the excitations). There are some failures of the linearized multiboson theories, they usually give excellent results, but sometimes they miss the microstructure found in real experiments. I hope that if I could retain some of the nonlinearities -in a kind of perturbative manner- than I would be able to describe the microstructures of the excitations (unfortunately this may very well turn out to be a false hope). And the last one: all our calculations were done on zero temperature, and I would like to extend them to finite temperatures.

4 Appendix

In order to make real calculations in the algebras $\mathfrak{su}(\mathfrak{n})$ we need several bases and have to compute the structure constants in these bases. Here we briefly mention some conventional bases, and describe the trick used to compute the structure constants We define some bases on the Lie algebra $\mathfrak{su}(\mathfrak{n})$, based on spin operators (n = 2S + 1).

4.1 Spherical Tensor Operators

As mentioned in the appropriate subsection there is a very useful basis on the Liealgebras $\mathfrak{su}(\mathfrak{n})$, especially when they are derived from spin models (if the spin length S > 1/2, than n > 2). We use definitions based on [23] and [30]. The Racah definition is the following: T_q^k is rank-k spherical tensor operator, where $k = 0, 1, \ldots, N - 1$ and $q = -k, -k + 1, \ldots k - 1, k$. They can be successively defined by starting with T_k^k (let $T_0^0 = 1$, but it is *not* an element of $\mathfrak{su}(\mathfrak{n})$), and applying the following commutation rules with the step operator S^- :

$$T_k^k = \beta(k) \cdot (S^+)^k, \tag{205}$$

$$\left[S^z, T^k_q\right] = qT^k_q,\tag{206}$$

$$\left[S^{\pm}, T_q^k\right] = \alpha_{\pm}(k, q) \cdot T_{q\pm 1}^k, \qquad (207)$$

$$\alpha_{\pm}(k,q) = \sqrt{k(k+1) - q(q\pm 1)},$$
(208)

where $\beta(k)$ is a real normalization constant to be chosen to taste. These operators are the $\mathfrak{su}(\mathfrak{n})$ generalization of the step operators (on their own, not only in the context of spin), with T_0^k analogous to S^z , and $T_{\pm q}^k$ to S^{\pm} . This basis clearly is not a selfadjoint one, but it can be the base to define a selfadjoint basis, i.e. the multipoles. Next we define the multipoles systematically.

4.2 Tesseral Harmonics, Multipoles, Stevens Operators

Based on the spherical tensor operators we can define the so-called tesseral harmonics (multipoles, Stevens operators), they form a selfadjoint basis of $\mathfrak{su}(\mathfrak{n})$:

$$\mathbb{T}_{k,q} = \begin{cases} \frac{1}{\sqrt{2}} \left[T_{-q}^k + (-1)^q T_q^k \right] & \text{if } q > 0 \\ T_0^k & \text{if } q = 0 \\ \frac{i}{\sqrt{2}} \left[T_q^k - (-1)^q T_{-q}^k \right] & \text{if } q < 0. \end{cases}$$
(209)

We enumerate the tesserals up to k = 3, sufficient for the S = 3/2, i.e. $\mathfrak{su}(4)$ calculations:

k	=	0
\mathbb{T}_{00}	=	$T_0^0 = 1.$
k	=	1
\mathbb{T}_{10}	=	$T_{0}^{1},$
\mathbb{T}_{11}	=	$\frac{1}{\sqrt{2}}(T_{-1}^1 - T_1^1),$
\mathbb{T}_{1-1}	=	$\frac{i}{\sqrt{2}}(T_{-1}^1 + T_1^1).$
k	=	2
\mathbb{T}_{20}	=	$T_{0}^{2},$
\mathbb{T}_{22}	=	$\frac{1}{\sqrt{2}}(T_{-2}^2 + T_2^2),$
\mathbb{T}_{2-2}	=	$\frac{i}{\sqrt{2}}(T_{-2}^2 - T_2^2),$
\mathbb{T}_{21}	=	$\frac{1}{\sqrt{2}}(T_{-1}^2 - T_1^2),$
\mathbb{T}_{2-1}	=	$\frac{i}{\sqrt{2}}(T_{-1}^2 + T_1^2).$
k	=	3
\mathbb{T}_{30}	=	$T_{0}^{3},$
\mathbb{T}_{33}	=	$\frac{1}{\sqrt{2}}(T_{-3}^3 - T_3^3),$
\mathbb{T}_{3-3}	=	$\frac{i}{\sqrt{2}}(T_{-3}^3 + T_3^3),$
\mathbb{T}_{32}	=	$\frac{1}{\sqrt{2}}(T_{-2}^3 + T_2^3),$
\mathbb{T}_{3-2}	=	$\frac{i}{\sqrt{2}}(T_{-2}^3 - T_2^3),$
\mathbb{T}_{31}	=	$\frac{1}{\sqrt{2}}(T_{-1}^3 - T_1^3),$
\mathbb{T}_{3-1}	=	$\frac{i}{\sqrt{2}}(T_{-1}^3 + T_1^3).$

4.3 Frobenius Inner Products, Commutators

We turn to a concrete representation of the operators in $\mathfrak{su}(4)$, and introduce some notions and tricks to calculate the structure constants in these bases.

Let us recall that the Frobenius (or Hilbert-Schmidt) inner product of two matrices is:

$$\langle\langle A, B \rangle\rangle = \operatorname{Tr}(A^{\dagger}B).$$
 (210)

It is clearly seen that it is an inner product on the linear space of the matrices. The tesserals and spherical operators form two orthogonal sets with respect to the Frobenius inner product:

$$\left\langle \left\langle T_{q}^{k}, T_{q'}^{k'} \right\rangle \right\rangle = N^{2}(k)\delta_{k,k'}\delta_{q,q'}, \qquad (211)$$

$$\left\langle \left\langle \mathbb{T}_{kq}, \mathbb{T}_{k'q'} \right\rangle \right\rangle = N^2(k)\delta_{k,k'}\delta_{q,q'},$$
(212)

where $N^2(k)$ is the squared norm of a rank-k tensor (the norm is denoted as $||A|| = \sqrt{\langle \langle A, A \rangle \rangle}$). The norms for the operators defined before read as:

$$N^{2}(k) = \left\langle \left\langle T_{0}^{k}, T_{0}^{k} \right\rangle \right\rangle = \left\langle \left\langle \mathbb{T}_{k0}, \mathbb{T}_{k0} \right\rangle \right\rangle = \operatorname{Tr}(T_{0}^{k\dagger}T_{0}^{k}) = \operatorname{Tr}((T_{0}^{k})^{2}),$$
(213)

$$N^{2}(1) = 10\beta^{2}(1), \ N^{2}(1) = 24\beta^{2}(2), \ N^{2}(1) = 36\beta^{2}(3).$$
 (214)

The important message is, that the physically relevant bases (the ones mentioned above, the Cartan-Weyl basis, or the generalized Gell-Mann matrices (c.f. [24]) fall into this class) are Frobenius-orthogonal, which will be crucial for the trick described below.

We want to calculate the expansion of the commutators of operators on a given basis, since these give us (-*i* times) the structure constants $f_{\alpha\beta}{}^{\gamma}$ of the algebra. Since $\mathfrak{su}(4)$ is 15 dimensional for each basis, we have to compute o(100) commutators (for each bases). We describe our method here.

Let us suppose that we want to compute the expansion of an operator A (probably given as a result of a commutator) on the given, not necessarily selfadjoint, Frobeniusorthogonal basis A_i of the algebra $\mathfrak{su}(\mathfrak{n})$:

$$A = \sum_{i} a_i A_i, \tag{215}$$

so our goal is here to calculate the expansion coefficients a_i , for a given operator A, and a given basis A_i , in a language a computer algebra software understands. Frobenius orthogonality of the basis means:

$$\langle\langle A_j, A_i \rangle\rangle = n_{j,i}\delta_{j,i},$$
(216)

where $n_{ii} = ||A_i||^2$. If we Frobenius multiply Eq. (215) by A_j from the left, and use the orthogonality of the basis we get:

$$\langle\langle A_j, A \rangle\rangle = \sum_i a_i \langle\langle A_j, A_i \rangle\rangle = \sum_i a_i n_{j,i} \delta_{j,i} = a_j n_{jj} = a_j ||A_j||^2.$$
(217)

And solving for the coefficients yields:

$$a_j = \frac{\langle \langle A_j, A \rangle \rangle}{||A_j||^2}.$$
(218)

Which is the result. Our calculations for the structure constants of $\mathfrak{su}(4)$ for the spherical tensors and tesserals are in the notebook comm_tesseral.nb, and for the generalized

Gell-Mann (i.e. Cartan-Weyl, c.f. Chapter 5. of [24]) matrices in the notebook comm_-Cartan_Weyl.nb. We checked the procedure by picking lots of commutators from the book mentioned, and from our calculations. The results match. To have a feeling of the much more complicated nature of these algebras, than that of $\mathfrak{su}(2)$ wee present the commutators for the Cartan-Weyl basis of $\mathfrak{su}(4)$ in a table on the next page. In this table the three matrices $H_{1,2,3}$ form the mutually commuting subalgebra of Cartan (c.f. the upper left corner). and $E_{\pm i}$, $i = 1, \ldots 6$ are the generalized ladder operators. Note that the if a \pm pair commutes the result is in the Cartan subalgebra, if a member H_i commutes with E_j , the result is always proportional to the latter operator. But if two nonpaired ladder commutes, the result is another *single* single member of the ladders. These nice algebraic properties make the Cartan-Weyl basis so comfortable, and play a very important part of the representation theory of Lie algebras.

E_6	0	$-\frac{2E_6}{3}$	$2E_6$	$H_2 - \frac{2H_3}{3}$	$-E_{-3}$	$-E_{-2}$	0	0	0	0	E_4	E_5	0	0	0
E_5	$-\frac{E_5}{2}$	$\frac{E_5}{3}$	$2\check{E}_5$	$-E_3$	$H_1 - \frac{H_2}{2} - \frac{2H_3}{3}$	$-\bar{E}_{-1}$	E_6	0	0	E_4	0	0	0	0	×
E_4	$\frac{E_4}{2}$	$\frac{E_4}{3}$	$2\check{E}_4$	$-E_2$	$-E_1$	$-H_1 - \frac{H_2}{2} - \frac{2H_3}{3}$	0	E_6	E_5	0	0	0	0	×	×
E_3	$-\frac{E_3}{2}$	E_3	0	0	E_{-6}	0	$H_1 - \frac{3H_2}{2}$	$-E_{-1}$	0	E_2	0	0	×	×	×
E_2	$\frac{E_2}{2}$	E_2	0	0	0	E_{-6}	$-E_1$	$-H_1 - \frac{3H_2}{2}$	E_3	0	0	×	×	×	×
E_1	E_1	0	0	0	0	E_{-5}	0	E_{-3}	$-2H_1$	0	×	×	×	×	×
E_{-1}	$-E_{-1}$	0	0	0	E_{-4}	0	E_{-2}	0	0	×	×	×	×	×	×
E_{-2}	$-\frac{E_{-2}}{2}$	$-E_{-2}$	0	E_{-4}	0	0	0	0	×	×	×	×	×	×	×
E_{-3}	$\frac{E_{-3}}{2}$	$-E_{-3}$	0	E_{-5}	0	0	0	×	×	×	×	×	×	×	×
E_{-4}	$-\frac{E_{-4}}{2}$	$-\frac{E_{-4}}{3}$	$-2\vec{E_{-4}}$	0	0	0	×	×	×	×	×	×	×	×	×
E_{-5}	$\frac{E_{-5}}{2}$	$-\frac{E_{-5}}{3}$	$-2\vec{E_{-5}}$	0	0	×	×	×	×	×	×	×	×	×	×
E_{-6}	0	$\frac{2E_{-6}}{3}$	$-2\tilde{E}_{-6}$	0	×	×	×	×	×	×	×	×	×	×	×
H_3	0	0	0	×	×	×	×	×	×	×	×	×	×	×	×
H_2	0	0	×	×	×	×	×	×	×	×	×	×	×	×	×
H_1	0	×	×	×	×	×	×	×	×	×	×	×	×	×	×
[A,B]	H_1	H_2	H_3	E_{-6}	E_{-5}	E_{-4}	E_{-3}	E_{-2}	E_{-1}	E_1	E_2	E_3	E_4	E_5	E_6

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