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Ultrafast Two-Magnon Excitation in Two-Dimensional Antiferromagnetic Insulators

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Master of Science, Nanotechnology

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NORWEGIAN UNIVERSITY OF SCIENCE AND
TECHNOLOGY

MASTER'S THESIS

**Ultrafast Two-Magnon Excitation in
Two-Dimensional Antiferromagnetic
Insulators**

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*A thesis submitted in fulfillment of the requirements
for the degree of Master of Science*

at the

Center for Quantum Spintronics
Department of Physics

June 22, 2020

Declaration of Authorship

I, Trond Hjerpekjøn HAUG, declare that this thesis titled, "Ultrafast Two-Magnon Excitation in Two-Dimensional Antiferromagnetic Insulators " and the work presented in it are my own. I confirm that:

- This work was done wholly while in candidature for a research degree at this University.
- Where I have consulted the published work of others, this is always clearly attributed.
- Where I have quoted from the work of others, the source is always given. With the exception of such quotations, this thesis is entirely my own work.
- I have acknowledged all main sources of help.
- Where the thesis is based on work done by myself jointly with others, I have made clear exactly what was done by others and what I have contributed myself.

Signed: Trond Hjerpekjøn Haug

Date: June 22nd 2020

"Eigi fellr tré við fyrsta hogg."

Old Norse proverb

NORWEGIAN UNIVERSITY OF SCIENCE AND TECHNOLOGY

Abstract

Faculty of Natural Sciences
Department of Physics

Master of Science

Ultrafast Two-Magnon Excitation in Two-Dimensional Antiferromagnetic Insulators

by Trond Hjerpekjøn HAUG

Spin excitations in two-dimensional antiferromagnetic materials take the form of spin waves that are more commonly referred to as magnons. Magnons are bosonic quasi-particles that can be excited by femtosecond laser-induced inhomogeneous perturbation of the exchange interaction between spins, allowing for ultrafast generation and control of these particles that have potential applications in high-speed information processing devices. I present the quantum theory for describing the dynamics triggered by the optical excitation and show how the magnons produce oscillations of the antiferromagnetic Néel vector that cannot be described using a semiclassical approach. A connection is drawn between operators that act on magnon pairs and the group $SU(1,1)$, which enables the representation of the quantum state of the antiferromagnet as a direct product of Perelomov coherent states. The effect of using a spatially nonuniform optical field intensity for magnon excitation is then studied using the techniques that proved effective for describing excitation by uniform fields. I discuss the generalization of the standard Bogoliubov transformation and apply it to the Hamiltonian that results from nonuniform optical fields before I present how the sudden approximation could potentially be used to numerically obtain the quantum state of the antiferromagnet after excitation by light with any spatially varying intensity distribution.

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Chapter 1

Introduction

Magnetic phenomena have been known to humans for at least 2500 years [1] and have grown increasingly important in technological applications as we have learned to exploit and control them. The first big breakthrough in the understanding of magnetism came in the early 1800s following Hans Christian Ørsted's discovery of its link to moving electrical charges in 1820 [2]. A series of experiments quickly followed at the hands of André-Marie Ampere [3], Jean-Baptiste Biot and Félix Savart [4] and Michael Faraday [5], among others, which laid the foundations for the theory of electromagnetism. The theory was unified and summarized by James Clark Maxwell [6] in 1873, although his famous equations and the description of the origin of electromagnetic waves must be ascribed to British scientists who came after him [7].

The second big leap came with the discovery of atoms and description of their structure using quantum mechanics in the 20th century. The contributions of people like Erwin Schrödinger [8, 9, 10], Werner Heisenberg [11], Wolfgang Pauli [12] and Paul Dirac [13] to the field of magnetism can hardly be understated given – as we shall see – its inherent quantum mechanical nature. This text concerns itself primarily with antiferromagnets, a group of materials that possess magnetic ordering below a characteristic temperature called the Néel temperature, without having a macroscopic magnetization. The magnetic ordering was first described by Louis Néel [14] who received the Nobel Prize in Physics in 1970 for his work on antiferromagnetism. Néel discovered that the behavior of these antiferromagnetic materials could be explained if the magnetic moment of an atom in the material was completely cancelled by the magnetic moment on a nearby atom. An example is shown in Fig. 1.1, where the magnetic moment of each lattice site is represented by an arrow. The magnetic moment is related to a property known as *spin* through the gyromagnetic ratio, meaning that the magnetic moment of an atom is proportional to its spin. The terms are therefore often

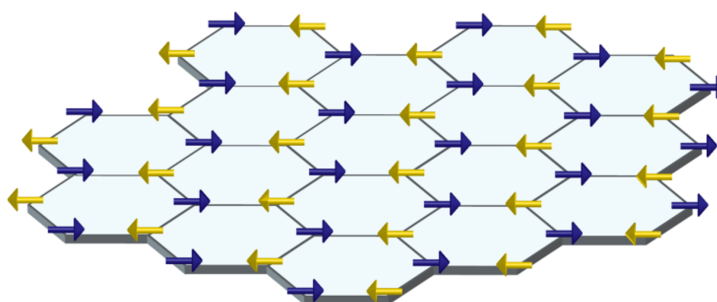


FIGURE 1.1: A honeycomb lattice of atoms with magnetic moments (spins) aligned in opposite directions. This material has an ordered magnetic structure, but the macroscopic magnetization, which is the sum of all the spins, is zero. Credit: E. Edwards.

used interchangeably, but since spin is the more fundamental quantity I will use this term.

By contrast, a ferromagnet is a material where all the spins are aligned parallel to one another. That is, turn all the yellow arrows in Fig. 1.1 by 180 degrees and you have a ferromagnet. The magnetization of the ferromagnet is simply the sum of all the magnetic moments divided by the volume (or in the two-dimensional case, the area) of the material. The magnetization can easily be measured and will tell us for instance the average orientation of the spins and their magnitude. We can also change the orientation of the spins by applying an external magnetic field, which will force the spins in the ferromagnet to align parallel to the field.

The lack of macroscopic magnetization in antiferromagnets was originally seen as a disadvantage when it came to possible technological applications for antiferromagnetic materials – in fact Néel himself famously said in his Nobel lecture that antiferromagnetic materials do not appear to have any interesting applications [15]. How can we exploit the magnetic order if there is no magnetization? In the recent decades we have learned to detect and interact with the magnetic moments of the atoms [16] rather than the macroscopic magnetization in the material, which has turned this disadvantage into an advantage in some areas. The absence of magnetic stray fields from antiferromagnets make them much more suited than ferromagnets for use in devices where we want to pack magnetic elements very close together, such as in memory storage and data processing. Another advantage of antiferromagnets is that the spins can be manipulated on ultrafast (sub-picosecond) timescales [17], which would make information processing and storage devices based on antiferromagnetic spin much faster than today's state-of-the-art computers that use electrical current (THz vs. GHz today).

The emerging fields that intend to exploit spin rather than electrical charge to carry information are called *spintronics* and *magnonics*. Spintronics seeks to use the spin of electrons to send electronic spin currents through circuits, whereas magnonics seeks to send magnon currents through magnetic insulators. A magnon is a quantum of excitation in a magnet that arises when the spins are moved out of their equilibrium orientation relative to one another. The situation is illustrated in Fig. 1.2. We see that the deflection from the equilibrium position traces out a sinusoidal wave, and magnons are therefore known as spin waves.

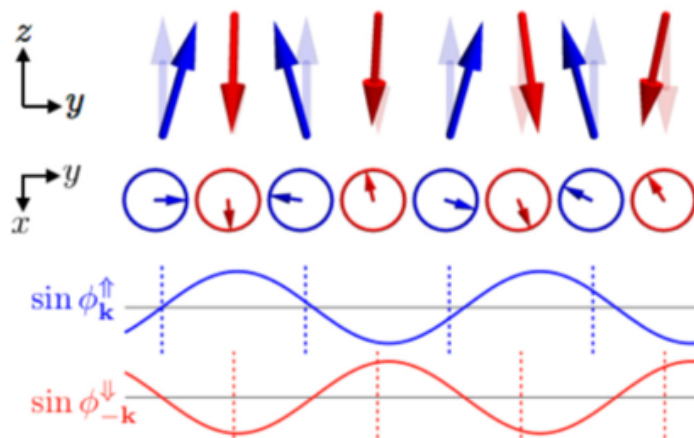


FIGURE 1.2: When spins are distorted out of their equilibrium orientations they precess around the equilibrium axis (shaded arrows). The deflection looks like a wave in the spin lattice, hence the term spin wave. Illustration originally from [18].

In this thesis we will encounter these pure spin waves and much more besides as we look at both static magnets and, later, antiferromagnetic dynamics. Although we restrict the discussion to some of the simplest antiferromagnets possible, we will need an arsenal of theories, approximations and techniques in condensed matter physics to get a grip on the problem. I have structured the thesis so as to serve as a gentle but thorough introduction to antiferromagnetic spin waves for graduate level students like myself who are relatively new to this branch of physics. It is my belief that the only priors needed to understand this text are a one-semester course in introductory quantum mechanics, a solid foundation in the core mathematical disciplines of multivariable calculus and linear algebra, and at least some familiarity with solid state physics, in particular reciprocal lattices and the Brillouin zone. As a consequence, readers with a strong background in condensed matter physics will probably find some phrases, in particular in the first couple of chapters, rather crude or perhaps even inaccurate. The writing style may be more colloquial than what you encounter in the average thesis as well. This is a feature, not a bug. While I have made every effort to make sure everything in this thesis is conveyed with as much precision as possible, I refuse to wrap the discussion in a veil of advanced terminology that only serves to alienate newcomers to the field. I also think that the discussion in Chapters 5 and 6 is technical enough as it is even for people who have some experience with magnonics.

As the reader has likely already noticed, I am switching frequently between "I" and "we". This is a small pedagogical experiment on my part. The vast majority of the text is written using the first person plural form because I want the readers to feel that they are taking part in the discussion and it is important that they understand what is going on. Sometimes, however, the use of the first person singular feels more natural because I want to express my own viewpoints and opinions, to which the readers might disagree. At some of the lengthier calculations I have also chosen to refer to myself with the singular pronoun to explain how I arrived at the result if it does not follow immediately from the written equations. On that note, I have tried to strike a balance between including as many intermediate calculations that are necessary to easily follow the derivations ("show your work"), and keeping the equations from filling up entire pages. The readers will therefore find that the derivations are more detailed than what you get from most scientific papers, but not as detailed as the typical calculus exam.

The thesis is divided into chapters of variable lengths, roughly corresponding to how much time it has taken me to study the material covered in them. Chapter 2 is short and introduces some much-needed concepts in quantum mechanics that are not always covered in low-level courses. Chapter 3 is even shorter and only scratches the surface of the semiclassical description of magnetic dynamics. I included it partly to illustrate the differences between the semiclassical and quantum theories, and partly because I reference the semiclassical theory in Chapter 5. The main segment of the thesis, which concerns quantum spin wave theory, is found in Chapters 4-6. The material covered in Chapters 4 and 5 is more or less properly developed by others, but is much more thoroughly explained than in any other single source I could find while studying for my degree. Chapter 6 relies to a less extent on published literature and is mostly a product of my own ideas, with the work of others duly cited when used, of course. I hope my efforts and ideas will prove useful in some way to the project currently undertaken by my supervisor and his collaborators, or at least help the graduate students succeeding me on the project build a better understanding of the challenges in a shorter period of time than I have managed.

Chapter 2

Background

2.1 Atomic origins of magnetism

In this section I outline the most fundamental aspects of how magnetism arises in insulators. The purpose is to give a motivation for why we model magnetic insulators as arrays of spin vectors. I treat the orbital angular momentum more thoroughly because we will need some of the derivations in later chapters, and because it is the easiest to get a grasp on since it has a classical analog. The origin of spin angular momentum and how the total angular momentum of an atom is calculated are discussed only briefly to avoid getting bogged down with relativistic effects, which would make the discussion unnecessarily long and conceptually difficult. Wave mechanics is avoided as well, which means restrictions on the orbital quantum numbers imposed by the spherical harmonics are not covered – both in the interest of brevity and because this should be familiar to students who have studied introductory quantum mechanics.

2.1.1 Orbital angular momentum

The magnetic moment m caused by a circulating current I around an area A is defined to be

$$m = IA. \quad (2.1)$$

If we expand this expression using $I = \frac{qN}{\tau}$ and $A = \pi r^2$, where q is the charge on the particles in the current, N is the number of charged particles passing through a cross section of the current loop in time τ and r is the radius of the current loop, then the magnitude of the magnetic moment is

$$m = \frac{qN}{\tau} \pi r^2. \quad (2.2)$$

The non-relativistic (linear) momentum of each particle is $p = m_0 v = m_0 \frac{2\pi r}{\tau}$, where m_0 is the rest mass of the particle. We can rewrite (2.2) as

$$\begin{aligned} m &= \frac{2m_0 qN}{2m_0 \tau} \pi r^2 \\ &= \frac{qN}{2m_0} r m_0 \frac{2\pi r}{\tau} \\ &= \frac{qN}{2m_0} r p \\ &= \frac{qN}{2m_0} L, \end{aligned} \quad (2.3)$$

where L is the angular momentum of one particle in the current. Clearly, charged particles with angular momentum are sources of magnetic moment. In an atom these

charged particles are electrons, and to find their angular momenta we need to find eigenvalues of the angular momentum operator $\hat{\mathbf{L}} = \hat{\mathbf{r}} \times \hat{\mathbf{p}}$. The individual components (x, y, z) of $\hat{\mathbf{L}}$ are

$$\hat{L}_x = \hat{y}\hat{p}_z - \hat{z}\hat{p}_x, \quad \hat{L}_y = \hat{z}\hat{p}_x - \hat{x}\hat{p}_z, \quad \hat{L}_z = \hat{x}\hat{p}_y - \hat{y}\hat{p}_x. \quad (2.4)$$

The commutator $[\hat{L}_x, \hat{L}_y]$ is

$$\begin{aligned} [\hat{L}_x, \hat{L}_y] &= [\hat{y}\hat{p}_z - \hat{z}\hat{p}_x, \hat{z}\hat{p}_x - \hat{x}\hat{p}_z] \\ &= [\hat{y}\hat{p}_z, \hat{z}\hat{p}_x] - [\hat{y}\hat{p}_z, \hat{x}\hat{p}_z] - [\hat{z}\hat{p}_x, \hat{z}\hat{p}_x] + [\hat{z}\hat{p}_x, \hat{x}\hat{p}_z]. \end{aligned} \quad (2.5)$$

Using the substitutions $\hat{q} \rightarrow q$ and $\hat{p}_q \rightarrow -i\hbar \frac{\partial}{\partial q}$ for $q \in \{x, y, z\}$, we get

$$\begin{aligned} [\hat{y}\hat{p}_z, \hat{z}\hat{p}_x] &= -\hbar^2 y \left(\frac{\partial}{\partial z} z \frac{\partial}{\partial x} \right) + \hbar^2 z y \frac{\partial^2}{\partial x \partial z} \\ &= -\hbar^2 y \left(\frac{\partial}{\partial x} + z \frac{\partial^2}{\partial z \partial x} \right) + \hbar^2 z y \frac{\partial^2}{\partial x \partial z} \\ &= -i\hbar y \left(-i\hbar \frac{\partial}{\partial x} \right) \\ &= -i\hbar \hat{y}\hat{p}_x. \end{aligned} \quad (2.6)$$

Similarly, we have

$$[\hat{y}\hat{p}_z, \hat{x}\hat{p}_z] = 0, \quad [\hat{z}\hat{p}_x, \hat{z}\hat{p}_x] = 0, \quad [\hat{z}\hat{p}_x, \hat{x}\hat{p}_z] = i\hbar \hat{x}\hat{p}_y. \quad (2.7)$$

Putting all this together yields

$$[\hat{L}_x, \hat{L}_y] = i\hbar (\hat{x}\hat{p}_y - \hat{y}\hat{p}_x) = i\hbar \hat{L}_z. \quad (2.8)$$

The commutators $[\hat{L}_y, \hat{L}_z]$ and $[\hat{L}_z, \hat{L}_x]$ follow by cyclic permutation of the coordinates:

$$\begin{aligned} [\hat{L}_y, \hat{L}_z] &= i\hbar \hat{L}_x, \\ [\hat{L}_z, \hat{L}_x] &= i\hbar \hat{L}_y. \end{aligned} \quad (2.9)$$

These are the fundamental commutation relations from which the rest of the theory of angular momentum follows. Crucially, the total angular momentum squared, $\hat{L}^2 \equiv \hat{L}_x^2 + \hat{L}_y^2 + \hat{L}_z^2$, commutes with each of the components of $\hat{\mathbf{L}}$. This means that there exist eigenstates of \hat{L}_z (or \hat{L}_y or \hat{L}_x) that are also eigenstates of \hat{L}^2 , i. e. we can know the magnitude of the total angular momentum and the magnitude of one of its components at the same time.

The eigenvalues are easiest to find if we introduce two new operators:

$$\begin{aligned} \hat{L}_+ &= \hat{L}_x + i\hat{L}_y, \\ \hat{L}_- &= \hat{L}_x - i\hat{L}_y. \end{aligned} \quad (2.10)$$

This way we obtain the commutation relations

$$[\hat{L}_z, \hat{L}_\pm] = [\hat{L}_z, \hat{L}_x] \pm i[\hat{L}_z, \hat{L}_y] = \pm\hbar (\hat{L}_x \pm i\hat{L}_y) = \pm\hbar \hat{L}_\pm, \quad (2.11)$$

$$[\hat{L}^2, \hat{L}_\pm] = 0. \quad (2.12)$$

Let $|\alpha\rangle$ be an eigenstate of \hat{L}^2 and \hat{L}_z so that

$$\hat{L}^2|\alpha\rangle = \alpha|\alpha\rangle, \hat{L}_z|\alpha\rangle = \beta|\alpha\rangle. \quad (2.13)$$

Then the commutation relations (2.11) and (2.12) ensure that

$$\hat{L}^2\hat{L}_\pm|\alpha\rangle = \hat{L}_\pm\hat{L}^2|\alpha\rangle = \alpha\hat{L}_\pm|\alpha\rangle \quad (2.14)$$

$$\hat{L}_z\hat{L}_\pm|\alpha\rangle = \pm\hbar\hat{L}_\pm|\alpha\rangle + \hat{L}_\pm\hat{L}_z|\alpha\rangle = (\pm\hbar + \beta)\hat{L}_\pm|\alpha\rangle. \quad (2.15)$$

Clearly, $\hat{L}_\pm|\alpha\rangle$ is also an eigenstate of both \hat{L}^2 and \hat{L}_z with eigenvalues α and $\beta \pm \hbar$, respectively. But α is the value of the total angular momentum squared and $\beta \pm \hbar$ is the z-component, which cannot be greater than the total. Evidently, there is an eigenstate $|\alpha_{max}\rangle$ for which $\hat{L}_+|\alpha_{max}\rangle = 0$. So for each eigenvalue α there exists a maximum eigenvalue β_{max} of \hat{L}_z . The value of α in terms of β_{max} is found by noting that

$$\hat{L}_\pm\hat{L}_\mp = (\hat{L}_x \pm i\hat{L}_y)(\hat{L}_x \mp i\hat{L}_y) = \hat{L}^2 - \hat{L}_z^2 \pm \hbar\hat{L}_z \implies \hat{L}^2 = \hat{L}_\pm\hat{L}_\mp + \hat{L}_z^2 \mp \hbar\hat{L}_z, \quad (2.16)$$

so that

$$\hat{L}^2|\alpha_{max}\rangle = (\hat{L}_-\hat{L}_+ + \hat{L}_z^2 + \hbar\hat{L}_z)|\alpha_{max}\rangle = (\beta_{max}^2 + \hbar\beta_{max})|\alpha_{max}\rangle. \quad (2.17)$$

Similarly, for each α there must exist a minimum eigenvalue β_{min} of \hat{L}_z . Applying the operator in (2.16) to $|\alpha_{min}\rangle$, we get

$$\hat{L}^2|\alpha_{min}\rangle = (\hat{L}_+\hat{L}_- + \hat{L}_z^2 - \hbar\hat{L}_z)|\alpha_{min}\rangle = (\beta_{min}^2 - \hbar\beta_{min})|\alpha_{min}\rangle. \quad (2.18)$$

From (2.17) and (2.18) we see that

$$\beta_{max}^2 + \hbar\beta_{max} = \beta_{min}^2 - \hbar\beta_{min}, \quad (2.19)$$

and by definition $\beta_{max} \geq \beta_{min}$. The only solution is

$$\beta_{min} = -\beta_{max}. \quad (2.20)$$

Moreover, since the eigenvalue of \hat{L}_z increases in n integer steps of units \hbar from $-\beta_{max}$ to β_{max} , it follows that

$$\beta_{max} = \hbar l, \quad l \in \{0, \frac{1}{2}, 1, \frac{3}{2}, \dots\} \quad (2.21)$$

and

$$\beta = \hbar m_l, \quad m_l \in \{-l, -l+1, \dots, l-1, l\} \quad (2.22)$$

The eigenvalue equations for \hat{L}^2 and \hat{L}_z in terms of the quantum numbers l and m_l are therefore

$$\hat{L}^2|l m_l\rangle = \hbar^2 l(l+1)|l m_l\rangle, \quad l \in \{0, \frac{1}{2}, 1, \frac{3}{2}, \dots\} \quad (2.23)$$

and

$$\hat{L}_z|l m_l\rangle = \hbar m_l|l m_l\rangle, \quad m_l \in \{-l, -l+1, \dots, l-1, l\}. \quad (2.24)$$

To summarize, we have found that a source of magnetic moment in atoms is electrons orbiting the nucleus. The angular momenta of the electrons are quantized. If we project the angular momentum of an electron with quantum number l onto the z-axis, we will find that the projection is $\hbar m_l$, where m_l is allowed to take any integer value between l and $-l$.

2.1.2 Spin angular momentum

There is a second contribution to the magnetic moments of atoms that arises from relativistic effects [19] and is believed to have no direct classical analog (although some disagreement exists [20]). It is called the electron's spin angular momentum. Fortunately, the theory of spin angular momentum (and indeed any general angular momentum) is identical to the theory of orbital angular momentum if we define the spin angular momentum operator $\hat{S} = \hat{S}_x\hat{x} + \hat{S}_y\hat{y} + \hat{S}_z\hat{z}$ to be an operator that satisfies the commutation relations in (2.8) and (2.9):

$$[\hat{S}_x, \hat{S}_y] = i\hbar\hat{S}_z, \quad [\hat{S}_y, \hat{S}_z] = i\hbar\hat{S}_x, \quad [\hat{S}_z, \hat{S}_x] = i\hbar\hat{S}_y. \quad (2.25)$$

This way the eigenvalue equations are simply

$$\hat{S}^2|s m_s\rangle = \hbar^2s(s+1)|s m_s\rangle, \quad s \in \{0, \frac{1}{2}, 1, \frac{3}{2}, \dots\}, \quad (2.26)$$

$$\hat{S}_z|s m_s\rangle = \hbar m_s|s m_s\rangle, \quad m_s \in \{-s, -s+1, \dots, s-1, s\}. \quad (2.27)$$

There is an important distinction between the quantum numbers l and s , however: the value for s is fixed at $\frac{1}{2}$ for electrons, whereas the value of l can vary. The fact that $s = \frac{1}{2}$ restricts m_s to the values $\pm\frac{1}{2}$. It is common to say that an electron with $m_s = +\frac{1}{2}$ is "spin up", while an electron with $m_s = -\frac{1}{2}$ is "spin down", and we mark them as \uparrow and \downarrow , respectively.

2.1.3 Total angular momentum

To find the atom's total angular momentum J we need to sum up all the contributions from the electrons' orbital and spin angular momenta. There are two strategies we can employ here:

1. We can add all the orbital contributions into a total orbital angular momentum, then add all the spin contributions into a total spin angular momentum, and finally add the total orbital and spin angular contributions:

$$L = \sum_i L_i, \quad S = \sum_i S_i, \quad J = L + S. \quad (2.28)$$

2. We can add each individual electron's orbital and spin angular momenta into a total angular momentum for the electron, and then sum the total angular momenta of the electrons in the atom:

$$J_i = L_i + S_i, \quad J = \sum_i J_i. \quad (2.29)$$

If the electrons' spin and orbital angular momenta were totally independent of each other, then it would make no difference which strategy we choose. However, the interaction between an electron's orbital angular momentum and its spin angular momentum causes an effect known as spin-orbit coupling. The coupling is weak in lighter atoms but increases rapidly as the nucleus becomes heavier [21]. The way the total angular momentum is calculated will depend on the degree of spin-orbit coupling in the atom. To our purposes the way the total angular momentum is calculated is of no importance. Therefore, to keep the notation consistent with convention, we shall from now on ignore the orbital angular momentum and only work with the spin

contribution to the atomic angular momentum. This does not reduce the generality of our results, since the physics would remain exactly the same had we not made this simplification. Thus for a known total spin quantum number s (the sum of the contributions from each electron's spin), we can express the state of the system in the basis of the spin projection along the z direction as $|m_s\rangle$.

2.2 Second quantization

As the number of atoms in a system grows, it becomes increasingly difficult to keep track of the state of every particle, which in turn determines the total state of the system. If we want to deal with macroscopic collectives of atoms, we need a new approach. Classically this was accomplished by introducing distribution functions so that the probability of finding a system in a given state could be calculated. We can hope to do something similar using quantum mechanics, but since we are operating with state vectors for the system as a whole, and all identical particles are fundamentally indistinguishable from one another, we must have that the observable state of the system is not affected if we exchange two identical particles. That is, the state can at most pick up a phase factor $e^{i\phi}$ in an exchange. In the position representation we have

$$|\Psi\rangle = \Psi(\zeta_1, \zeta_2, \dots, \zeta_i, \dots, \zeta_j, \dots, \zeta_N) = e^{i\phi} \Psi(\zeta_1, \zeta_2, \dots, \zeta_j, \dots, \zeta_i, \dots, \zeta_N) \quad (2.30)$$

where ζ_n is the coordinates and spin of the n -th particle in a system of N particles. If we exchange the particles back again, we get

$$\Psi(\zeta_1, \zeta_2, \dots, \zeta_j, \dots, \zeta_i, \dots, \zeta_N) = e^{i\phi} \Psi(\zeta_1, \zeta_2, \dots, \zeta_i, \dots, \zeta_j, \dots, \zeta_N) \quad (2.31)$$

and therefore $e^{2i\phi} = 1 \implies e^{i\phi} = \pm 1$, which means

$$\Psi(\zeta_1, \zeta_2, \dots, \zeta_i, \dots, \zeta_j, \dots, \zeta_N) = \begin{cases} +\Psi(\zeta_1, \zeta_2, \dots, \zeta_j, \dots, \zeta_i, \dots, \zeta_N) \\ -\Psi(\zeta_1, \zeta_2, \dots, \zeta_j, \dots, \zeta_i, \dots, \zeta_N) \end{cases}.$$

This requirement that Ψ be either symmetric or antisymmetric gives rise to two different forms of statistics: Bose-Einstein statistics for the symmetric wave functions and Fermi-Dirac statistics for antisymmetric wave functions. Relativistic quantum mechanics reveals that integer-spin particles obey Bose-Einstein statistics and are called bosons, whereas half-integer-spin particles obey Fermi-Dirac statistics and are called fermions [22].

When we expand Ψ in products of eigenfunctions of the one-particle Hamiltonian (which form a complete set), we therefore have the restriction that Ψ be symmetrized for bosons and anti-symmetrized for fermions. Mathematically, we have for bosons that

$$\Psi_N^B = \sqrt{\frac{n_1! n_2! \cdots n_N!}{N!}} \sum \psi_1(\zeta_1) \psi_2(\zeta_2) \cdots \psi_N(\zeta_N), \quad (2.32)$$

where the sum runs over all distinguishable permutations of the indices $\{1, 2, \dots, N\}$ and n_i counts the number of times the one-particle wave function ψ_i appears in Ψ_N . The expression for fermions is a bit more involved since the exchange of one particle with another changes the sign of Ψ_N^F . The wave function is best expressed with a

Slater determinant:

$$\Psi_N^F = \sqrt{\frac{1}{N!}} \begin{vmatrix} \psi_1(\zeta_1) & \psi_1(\zeta_2) & \psi_1(\zeta_3) & \cdots & \psi_1(\zeta_N) \\ \psi_2(\zeta_1) & \psi_2(\zeta_2) & \psi_2(\zeta_3) & \cdots & \psi_2(\zeta_N) \\ \psi_3(\zeta_1) & \psi_3(\zeta_2) & \psi_3(\zeta_3) & \cdots & \psi_3(\zeta_N) \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \psi_N(\zeta_1) & \psi_N(\zeta_2) & \psi_N(\zeta_3) & \cdots & \psi_N(\zeta_N) \end{vmatrix}. \quad (2.33)$$

In this basis an N -particle state Ψ_N is completely defined by the set of numbers $\{n_i\}$. A more convenient notation is therefore

$$\Psi_N = |n_1, n_2, \dots, n_N\rangle. \quad (2.34)$$

The Hilbert space (the space in which quantum mechanical states live) spanned by a finite or *denumerably* infinite number of $|n_i\rangle$ states is called a Fock space. The state $|n_1, n_2, \dots, n_N\rangle$ is called a Fock state and the operators in Fock space that act on Fock states are called second-quantized operators. These operators change the occupation numbers n_i in the Fock states and are therefore called creation and annihilation operators. I will make extensive use of these operators throughout this thesis, starting in Chapter 4.

2.3 The three pictures of quantum mechanics

Most introductory quantum mechanics courses teach a formulation of quantum mechanics known as the Schrödinger picture in which the operators are time-independent and the quantum states evolve with time. Taking the geometric view of states and operators, the Schrödinger picture represents a view where the state vectors move about in a static coordinate system. Most of the discussion in this thesis takes this view. However, there is nothing that prevents us from making a change of basis so that the coordinate system is moving as well. This might seem like an unnecessary complication at first, but by making this transformation it often becomes easier to deal with interactions. Since we will deal with interactions between spins and light in Chapters 5 and 6, I present here a short summary of some important aspects that will be useful to us from the three pictures in quantum mechanics: the Schrödinger, interaction (Dirac) and Heisenberg pictures.

2.3.1 The Schrödinger picture

As I already mentioned, the Schrödinger picture takes the view of static operators and evolving states. We will denote a state in the Schrödinger picture as $|\Psi_S(t)\rangle$. The Schrödinger state's evolution is described by a unitary operator $\hat{U}(t, t_0)$ which acts on the state at t_0 and rotates it so that at time t the state is $|\Psi(t)\rangle$. A unitary operator is an operator whose adjoint is equal to its inverse: $\hat{U}^\dagger = \hat{U}^{-1}$. Mathematically we have

$$|\Psi_S(t)\rangle = \hat{U}(t, t_0)|\Psi_S(t_0)\rangle. \quad (2.35)$$

We can put this into the Schrödinger equation $i\hbar \frac{d}{dt} |\Psi_S(t)\rangle = \hat{\mathcal{H}} |\Psi_S(t)\rangle$ and get an equation for the evolution operator \hat{U} :

$$i\hbar \frac{d}{dt} \hat{U}(t, t_0) = \hat{\mathcal{H}} \hat{U}(t, t_0). \quad (2.36)$$

The solution is

$$\hat{U}(t, t_0) = \exp \left[-\frac{i}{\hbar} \int_{t_0}^t \hat{\mathcal{H}}(t') dt' \right]. \quad (2.37)$$

I have assumed that the Hamiltonians at different times commute (this assumption is rarely wrong and certainly holds for the discussion in this text). Unless there is some explicit time-dependence in the Hamiltonian $\hat{\mathcal{H}}$ as a result of for instance a temporally varying potential, the Hamiltonian is a function of static operators only and can therefore be taken out of the integral. In that case we get

$$\hat{U}_S(t, t_0) = \exp \left[-i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right], \quad (2.38)$$

where I added the subscript 0 to emphasize that the Hamiltonian is time-independent.

2.3.2 The interaction picture

In the case that there *is* some time-dependence in the Hamiltonian, we can often write the Hamiltonian as a sum of a time-independent part and a time-varying part: $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \delta\hat{\mathcal{H}}(t)$. We could have chosen to split the Hamiltonian in any way we like, but this way of writing it will be most convenient. We now define a state in the interaction picture as

$$|\Psi_I(t)\rangle = \exp \left[i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] |\Psi_S(t)\rangle. \quad (2.39)$$

Using (2.35) and (2.37) we see that $|\Psi_I(t)\rangle$ still carries some time-dependence:

$$|\Psi_I(t)\rangle = \exp \left[-\frac{i}{\hbar} \int_{t_0}^t \delta\hat{\mathcal{H}}(t') dt' \right] |\Psi_S(t_0)\rangle, \quad (2.40)$$

and in order to preserve the expectation values of operators (observables such as momentum, spin, energy and so on cannot depend on which picture we are using to represent the system!), they must transform in a similar manner:

$$\hat{X}_I(t, t_0) = \exp \left[i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] \hat{X}_S \exp \left[-i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] = \hat{U}_S^\dagger \hat{X}_S \hat{U}_S, \quad (2.41)$$

where \hat{X} is any operator. As an example, the Hamilton operator in the interaction picture becomes

$$\begin{aligned} \hat{\mathcal{H}}_I &= \exp \left[i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] (\hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1(t)) \exp \left[-i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] \\ &= \hat{\mathcal{H}}_0 + \exp \left[i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] \hat{\mathcal{H}}_1(t) \exp \left[-i \frac{\hat{\mathcal{H}}_0(t - t_0)}{\hbar} \right] \\ &= \hat{\mathcal{H}}_0 + \hat{U}_S^\dagger \hat{\mathcal{H}}_1 \hat{U}_S. \end{aligned} \quad (2.42)$$

To conclude our discussion of the interaction picture, we derive the transformed Schrödinger equation which gives the time-evolution of states. We write $|\Psi_S(t)\rangle =$

$\hat{U}_S|\Psi_I(t)\rangle$, which once inserted into the Schrödinger equation yields

$$\begin{aligned}\hat{\mathcal{H}}\hat{U}_S|\Psi_I(t)\rangle &= i\hbar\frac{d}{dt}\hat{U}_S|\Psi_I(t)\rangle \\ &= i\hbar\left(\frac{d\hat{U}_S}{dt}|\Psi_I(t)\rangle + \hat{U}_S\frac{d}{dt}\hat{\mathcal{H}}\right) \\ &= \hat{\mathcal{H}}_0\hat{U}_S|\Psi_I(t)\rangle + i\hbar\hat{U}_S\frac{d}{dt}|\Psi_I(t)\rangle.\end{aligned}\tag{2.43}$$

Operating with \hat{U}_S^\dagger from the left and using $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \delta\hat{\mathcal{H}}$, we get

$$i\hbar\frac{d}{dt}|\Psi_I(t)\rangle = \left(\hat{U}_S^\dagger\hat{\mathcal{H}}_0\hat{U}_S + \hat{U}_S^\dagger\delta\hat{\mathcal{H}}\hat{U}_S - \hat{U}_S^\dagger\hat{\mathcal{H}}_0\hat{U}_S\right)|\Psi_I(t)\rangle = \hat{U}_S^\dagger\delta\hat{\mathcal{H}}\hat{U}_S|\Psi_I(t)\rangle.\tag{2.44}$$

This equation can be written even more compactly by using (2.41):

$$i\hbar\frac{\partial}{\partial t}|\Psi_I(t)\rangle = \delta\hat{\mathcal{H}}^I(t)|\Psi_I(t)\rangle.\tag{2.45}$$

2.3.3 The Heisenberg picture

In the interaction picture we transfer some of the time-dependence of the states into the operators. We can of course transfer *all* the time-dependence to the operators; this is called the Heisenberg picture. There are no surprises in the expressions for the states and operators – they follow from our discussion about the interaction picture:

$$|\Psi_H\rangle = |\Psi_S(t_0)\rangle\tag{2.46}$$

$$\hat{X}_H(t) = \exp\left[-\frac{i}{\hbar}\int_{t_0}^t\hat{\mathcal{H}}(t')dt'\right]\hat{X}_S\exp\left[\frac{i}{\hbar}\int_{t_0}^t\hat{\mathcal{H}}(t')dt'\right].\tag{2.47}$$

We will not be using the Heisenberg picture in this text, so we will end the discussion here. The pictures are treated more thoroughly in many books on quantum mechanics, for instance in [23].

2.4 Squeezed states

One of the most fundamental results of quantum mechanics is that there is some inherent uncertainty in quantities whose operators do not commute. In fact, this is the reason why we cannot have the angular momentum (spin) of an atom perfectly aligned with the z -axis: the spin operators in the x -, y - and z -directions do not commute, so there is no such thing as a state where the angular momentum has a precise direction. For two Hermitian operators \hat{A} and \hat{B} , the uncertainty relation is [23]

$$\sigma_A\sigma_B \geq \left|\frac{1}{2i}\langle[\hat{A}, \hat{B}]\rangle\right|,\tag{2.48}$$

where σ_X is the standard deviation in a measurement of the eigenvalue of the operator \hat{X} . It is important to note that the relation is an *inequality*, so there is no guarantee that we will hit the equality limit. Also, there is no lower limit on the uncertainty in one quantity alone: we can make the measurement of A as precise as we want at the expense of large uncertainty in B . The uncertainty in a physical quantity that follows

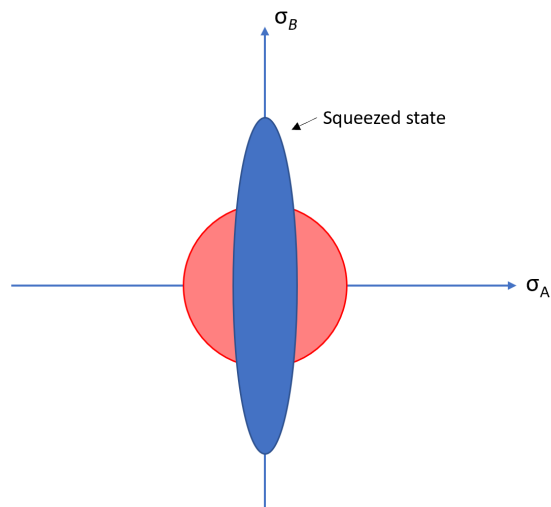


FIGURE 2.1: The shaded regions represent forbidden areas because of the uncertainty relation (2.48). The squeezed state (blue) represents a state in which the uncertainty in A is reduced compared to the vacuum state (red).

from its quantum nature is often called quantum noise. Put simply, squeezed states are states that minimize the quantum noise in one observable at the expense of the other. If we picture the uncertainties as axes of an ellipse, then reducing one of the uncertainties is like squeezing the ellipse from two sides, increasing its elongation (or eccentricity). The process is illustrated in Fig. 2.1. Quantum noise is obviously a quantum effect, and hence if we want to find a quantum description that most resembles a classical description of a system, then squeezing the quantum noise out of the system is a way to do so.

Squeezed states are mostly used in the field of quantum optics, so the concept of squeezed states in quantum magnetism can be a bit slippery. We will encounter squeezed states in Chapter 5 as we look to find states that allow us to link the quantum description of antiferromagnetic dynamics with the classical description in Chapter 3.

Chapter 3

Semiclassical Dynamics in Antiferromagnets

The goal of the semiclassical approach is to move from a description of individual spins on a lattice to a description of a continuous field in the antiferromagnet. In a ferromagnet this would amount to finding the magnetization $\mathbf{M}(\mathbf{r}, t)$, whose dynamics is described by the Landau-Lifshitz-Gilbert equation:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} + \frac{\alpha}{M_s} \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}, \quad (3.1)$$

where γ is the gyromagnetic ratio, \mathbf{H}_{eff} is the effective magnetic field, α is a damping parameter and M_s is the saturation magnetization. We would like to find a similar dynamic equation for an antiferromagnet, but there is a problem: classically the magnetization of an antiferromagnet is zero. To get around this we have to introduce two fields for the antiferromagnet. One of the techniques for doing this is called Haldane's mapping [24], where we map each spin \mathbf{S}_i onto two orthogonal vectors called the Néel unitary vector $\tilde{\mathbf{n}}_i$ and the canting vector $\tilde{\mathbf{m}}_i$:

$$\mathbf{S}_i = (-1)^i S \sqrt{1 - |\tilde{\mathbf{m}}_i|^2} \tilde{\mathbf{n}}_i + \tilde{\mathbf{m}}_i. \quad (3.2)$$

From this definition of $\tilde{\mathbf{n}}$ and $\tilde{\mathbf{m}}$ we can say something about the frequencies of oscillation in the spins \mathbf{S}_i . In a classical picture, we think of spin vectors as precessing around the equilibrium axis with a frequency ω . Taking the equilibrium axis to be the direction of the Néel unitary vector, this would mean that the projection of the spin vector onto the canting vector can be written

$$\tilde{\mathbf{m}}_i \sim \tilde{\mathbf{m}}_i^0 \sin(\omega t). \quad (3.3)$$

The projection of \mathbf{S}_i onto the Néel unitary field is

$$(-1)^i S \sqrt{1 - |\tilde{\mathbf{m}}_i|^2} \approx (-1)^i S \left(1 - \frac{1}{2} |\tilde{\mathbf{m}}_i|^2 \right). \quad (3.4)$$

Inserting the value of the canting field projection yields

$$\begin{aligned} \mathbf{S}_i \cdot \mathbf{n}_i &\approx (-1)^i S \left(1 - \frac{1}{2} |\tilde{\mathbf{m}}_i|^2 \right) \\ &\sim (-1)^i S \left(1 - \frac{1}{2} |\tilde{\mathbf{m}}_i^0|^2 \sin^2(\omega t) \right) \\ &= (-1)^i S \left(1 - \frac{1}{4} |\tilde{\mathbf{m}}_i^0|^2 [1 - \cos(2\omega t)] \right). \end{aligned} \quad (3.5)$$

We see that, to the leading order, the oscillations in the direction along the Néel unitary vector are twice as fast as the transverse oscillations, and that this is a nonlinear effect. When we treat the dynamics using quantum mechanics in Chapter 5, we will obtain the same result, but this time the phenomenon will be present even in a purely linear theory.

Rather awkwardly, there is another approach to parameterize spins to continuous fields in addition to Haldane's mapping which is somewhat standard. This is called the Hamiltonian approach [25] and introduces two similar-looking quantities:

$$\mathbf{m}_i = \frac{\mathbf{S}_A^i + \mathbf{S}_B^i}{2S}, \quad (3.6)$$

$$\mathbf{l}_i = \frac{\mathbf{S}_A^i - \mathbf{S}_B^i}{2S}, \quad (3.7)$$

where \mathbf{m}_i is the vector of magnetization on site i , which now contains *two* spins: one spin up (denoted with subscript A) and one spin down (denoted with subscript B). The pairing of spins causes the number of sites in the antiferromagnet to drop by half compared to Haldane's mapping. The vector \mathbf{l}_i is known as the antiferromagnetic vector or the Néel vector. This definition of the Néel vector is *not* equivalent to the definition in Haldane's mapping. We are done with Haldane's definition now: from now on the Néel vector is defined as in (3.7). We define the unitary Néel vector in the obvious way:

$$\mathbf{n}_i = \frac{\mathbf{l}_i}{|\mathbf{l}_i|}, \quad (3.8)$$

again stressing that $\mathbf{n}_i \neq \frac{1}{2}(\tilde{\mathbf{n}}_{2i-1} + \tilde{\mathbf{n}}_{2i})$ and $\mathbf{m}_i \neq \frac{1}{2}(\tilde{\mathbf{m}}_{2i-1} + \tilde{\mathbf{m}}_{2i})$. We now take the continuous limit

$$\mathbf{n}_i \rightarrow \mathbf{n}(\mathbf{r}, t), \quad \mathbf{m}_i \rightarrow \mathbf{m}(\mathbf{r}, t), \quad (3.9)$$

so that we obtain magnetization and Néel *fields*, which are expected to yield a good description in the case where difference between vectors on neighboring sites is small (i.e. the energy in the system is low). I will suppress the notation showing the spatial and temporal dependence of the fields in the rest of the chapter and simply use \mathbf{n} and \mathbf{m} . The fields are subject to the constraint

$$\mathbf{n} \cdot \mathbf{m} = 0. \quad (3.10)$$

Now we are ready to derive equations for the dynamics of \mathbf{n} and \mathbf{m} . These can be obtained as Euler-Lagrange equations from the principle of least action through the use of the Lagrangian density. In terms of the fields \mathbf{n} and \mathbf{m} it can be shown [26] that the Lagrangian density is

$$\mathcal{L} = \mathcal{K} - \mathcal{U} = \frac{1}{\gamma} \mathbf{m} \cdot (\dot{\mathbf{n}} \times \mathbf{n}) - \mathcal{U}, \quad (3.11)$$

where γ is the gyromagnetic ratio, \mathcal{K} is the kinetic energy density, \mathcal{U} is the potential energy density and $\dot{\mathbf{n}} = \frac{\partial \mathbf{n}}{\partial t}$. The Lagrangian L is therefore

$$L = \int_{-\infty}^{\infty} \mathcal{L} d^3r. \quad (3.12)$$

We seek functions \mathbf{n} and \mathbf{m} that are stationaries of the Lagrangian:

$$\frac{\delta L}{\delta \mathbf{n}} = 0, \quad \frac{\delta L}{\delta \mathbf{m}} = 0, \quad (3.13)$$

where $\delta \mathbf{n}$ and $\delta \mathbf{m}$ are small variations in \mathbf{n} and \mathbf{m} , respectively. The Néel and magnetization fields that satisfy (3.13) must also satisfy the Euler-Lagrange equation, which serves the same purpose in Lagrangian mechanics as Newton's laws of motion in Newtonian mechanics. Using (3.11) we get that the Euler-Lagrange equation reads

$$\frac{\partial \mathcal{L}}{\partial \mathbf{n}} - \frac{\partial}{\partial t} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{n}}} = 0 \implies \frac{\partial \mathcal{K}}{\partial \mathbf{n}} - \frac{\partial}{\partial t} \frac{\partial \mathcal{K}}{\partial \dot{\mathbf{n}}} - \frac{\delta U}{\delta \mathbf{n}} = 0 \quad (3.14)$$

for the Néel field \mathbf{n} and

$$\frac{\partial \mathcal{L}}{\partial \mathbf{m}} - \frac{\partial}{\partial t} \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{m}}} = 0 \implies \frac{\partial \mathcal{K}}{\partial \mathbf{m}} - \frac{\partial}{\partial t} \frac{\partial \mathcal{K}}{\partial \dot{\mathbf{m}}} - \frac{\delta U}{\delta \mathbf{m}} = 0 \quad (3.15)$$

for the magnetization field \mathbf{m} , where $U = \int \mathcal{U} d^3r$. Inserting the expression for the kinetic energy yields

$$\dot{\mathbf{n}} \times \mathbf{n} = \gamma \frac{\delta U}{\delta \mathbf{m}}, \quad (3.16)$$

$$2\mathbf{m} \times \dot{\mathbf{n}} + \dot{\mathbf{m}} \times \mathbf{n} = \gamma \frac{\delta U}{\delta \mathbf{n}}. \quad (3.17)$$

Explicit expressions for $\dot{\mathbf{n}}$ and $\dot{\mathbf{m}}$ can be obtained by noting that $\dot{\mathbf{n}} \perp \mathbf{n}$ because $|\mathbf{n}|^2 = 1$, so by crossing the equations with \mathbf{n} from the left we obtain

$$\dot{\mathbf{n}} = \gamma \mathbf{n} \times \frac{\delta U}{\delta \mathbf{m}}, \quad (3.18)$$

$$\dot{\mathbf{m}} = \gamma \left(\mathbf{n} \times \frac{\delta U}{\delta \mathbf{n}} + \mathbf{m} \times \frac{\delta U}{\delta \mathbf{m}} \right). \quad (3.19)$$

These coupled equations are valid independently of the sophistication of the model we use for the antiferromagnetic free energy functional U . They are the equivalent equations to the Landau-Lifshitz equation for ferromagnets. A typical, simple model of the free energy would include the magnetostatic energy, magnetocrystalline anisotropy with easy axis along the z-axis and an external magnetic field \mathbf{H} :

$$U = \int \mathcal{U} d^3r = \int \left(\frac{a}{2} |\mathbf{m}|^2 + \frac{A}{2} |\nabla \mathbf{n}|^2 - \frac{\kappa}{2} (\mathbf{n} \cdot \hat{\mathbf{z}})^2 + \mathfrak{L} \mathbf{m} \cdot (\nabla \mathbf{n}) - \mathbf{H} \cdot \mathbf{m} \right) d^3r, \quad (3.20)$$

where a is the homogeneous exchange constant, A is the exchange stiffness, κ is the anisotropy energy, $\nabla \mathbf{n} \equiv \sum_{i=x,y,z} \frac{\partial}{\partial i} \mathbf{n}$ and \mathfrak{L} parameterizes a parity-breaking term that arises because we divided the antiferromagnetic lattice into pairs [26]. This allows us to calculate the functional derivatives of U :

$$\frac{\delta U}{\delta \mathbf{m}} = a\mathbf{m} + (\mathfrak{L} \nabla \mathbf{n} - \mathbf{H}), \quad (3.21)$$

$$\frac{\delta U}{\delta \mathbf{n}} = -\kappa (\mathbf{n} \cdot \hat{\mathbf{z}}) \hat{\mathbf{z}} + A \nabla^2 \mathbf{n}. \quad (3.22)$$

Inserting (3.21) into (3.18) and crossing it with $-\mathbf{n}$ from the left we obtain (after rearranging)

$$\mathbf{m} = \frac{1}{a} \left(\frac{1}{\gamma} \dot{\mathbf{n}} \times \mathbf{n} + \mathbf{n} \times [\mathbf{n} \times (\mathfrak{L} \nabla \mathbf{n} - \mathbf{H})] \right), \quad (3.23)$$

from which we see that \mathbf{m} is a slave variable and we can therefore find a dynamic equation for \mathbf{n} that does not depend on \mathbf{m} . Moreover, it implies that the dynamic equation for \mathbf{n} is not affected by whether the restrictions $|\mathbf{n}|^2 = 1$ and $\mathbf{n} \cdot \mathbf{m} = 0$ are enforced through Lagrange multipliers in the derivation, which is why they were dropped. We could at this point take the expression for $\mathbf{m}(\mathbf{n}, \dot{\mathbf{n}}, \nabla \mathbf{n}, \mathbf{H})$ and plug it into (3.14) to obtain a differential equation of the form

$$\ddot{\mathbf{n}} = F(\mathbf{n}, \dot{\mathbf{n}}, \nabla \mathbf{n}, \mathbf{H}). \quad (3.24)$$

Although this is of the same form as Newton's equation of motion, F will not be a particularly pretty function and the equation will definitely not be analytically solvable. However, we have obtained an equation that serves the same purpose as the Landau-Lifshitz-Gilbert equation (3.1), which is all we really need: we can use (3.24) to find the evolution of the Néel field, which describes the motion of the spins in the antiferromagnet.

Chapter 4

The Antiferromagnetic Ground State

We now turn our attention to the problem of finding the ground state energy of an antiferromagnet at zero temperature using quantum mechanics. One of the most general methods, applicable to systems in one, two and three dimensions and with any value of s , is known as spin wave theory and revolves around the idea of replacing the spin operators with creation and annihilation operators for bosons. This will diagonalize the Hamiltonian and allow us to compare the quantum ground state to the classical ground state, which we call the Néel state.

4.1 Spin wave theory

In the discussion of orbital angular momentum we introduced operators \hat{L}_\pm that act on eigenstates of L_z and raise (lower) the state to another eigenstate of L_z with eigenvalue \hbar above (below) the previous state, as seen from (2.15). To keep the notation tidy, we will from now on be working in units of \hbar so that \hbar has value 1. Since the theory of spin is nothing but a carbon copy of the theory of orbital angular momentum, we have

$$\hat{S}_z \hat{S}_\pm |m_s\rangle = (m_s \pm 1) \hat{S}_\pm |m_s\rangle, \quad (4.1)$$

which implies

$$\hat{S}_\pm |m_s\rangle = c_\pm |m_s \pm 1\rangle. \quad (4.2)$$

The constants c_\pm remain to be determined. For c_+ we have

$$\langle m_s | \hat{S}_+^\dagger \hat{S}_+ |m_s\rangle = (\hat{S}_+ |m_s\rangle)^\dagger \hat{S}_+ |m_s\rangle = c_+^* c_+ = |c_+|^2. \quad (4.3)$$

But $\hat{S}_+^\dagger = \hat{S}_-$ and $\hat{S}_- \hat{S}_+ = \hat{S}^2 - \hat{S}_z^2 - \hat{S}_z$. So

$$\begin{aligned} \langle m_s | \hat{S}_+^\dagger \hat{S}_+ |m_s\rangle &= \langle m_s | \hat{S}^2 - \hat{S}_z^2 - \hat{S}_z |m_s\rangle \\ &= (s(s+1) - m_s^2 - m_s) \langle m_s | m_s\rangle \\ &= (s - m_s)(s + m_s + 1) \end{aligned} \quad (4.4)$$

and hence $c_+ = \sqrt{(s - m_s)(s + m_s + 1)}$ times a phase factor which we choose to be unity. This approach applied to c_- as well yields

$$\begin{aligned} \hat{S}_+ |m_s\rangle &= \sqrt{(s - m_s)(s + m_s + 1)} |m_s + 1\rangle, \\ \hat{S}_- |m_s\rangle &= \sqrt{(s + m_s)(s - m_s + 1)} |m_s - 1\rangle. \end{aligned} \quad (4.5)$$

Of course, the states $|s+1\rangle$ and $|-s-1\rangle$ are unphysical, so $\hat{S}_+|s\rangle = 0$ and $\hat{S}_-|-s\rangle = 0$.

The most common way to introduce the boson operators is called the Holstein-Primakoff transformation [27]. The idea is to set the state of maximum spin projected onto the z -axis $|s\rangle$ as the vacuum state and then apply the creation operator

$$\hat{a}^\dagger|s\rangle = c|s-1\rangle \quad (4.6)$$

to make an excited state. This creation operator can be applied multiple times until the state $|-s\rangle$ is reached. The number of bosons is interpreted as the number of deviations from the vacuum state. The adjoint \hat{a} is the annihilation operator, which has the opposite effect:

$$\hat{a}|m_s\rangle = c^*|m_s+1\rangle \quad (4.7)$$

The constant c is defined such that when we apply the number operator $\hat{n} \equiv \hat{a}^\dagger\hat{a}$, we get the number of bosons back:

$$\hat{a}^\dagger\hat{a}|m_s\rangle = (s-m_s)|m_s\rangle, \quad (4.8)$$

which obviously means $c = \sqrt{s-m_s}$ times a phase factor which we choose to be unity. Thus

$$\hat{S}_z|m_s\rangle = m_s|m_s\rangle = (s-s+m_s)|m_s\rangle = (s-\hat{a}^\dagger\hat{a})|m_s\rangle. \quad (4.9)$$

The expressions for \hat{S}_+ and \hat{S}_- are more easily verified than derived, so I will just state them here [28]:

$$\hat{S}_+ = \sqrt{2s}\sqrt{1-\frac{\hat{n}}{2s}}\hat{a}, \quad (4.10)$$

$$\hat{S}_- = \sqrt{2s}\hat{a}^\dagger\sqrt{1-\frac{\hat{n}}{2s}}. \quad (4.11)$$

It is straightforward to check that these expressions yield (4.5) if applied to a state $|m_s\rangle$. The Holstein-Primakoff transformation's main disadvantage is that working with square roots of operators is notoriously difficult. This can be overcome by using the Taylor expansion of the root around 0 – which is known as a Maclaurin expansion – and keeping only the first one or two terms, which works best when s is large and $\langle\hat{n}\rangle$ is small. The price to pay is that the theory is no longer exact, but we gain insights into the physical picture of the spin system that are otherwise not easily obtained.

So far we have only considered the case where the vacuum state is $|s\rangle$ for each lattice site. But for antiferromagnets this is not the case. The simplest antiferromagnetic spin systems consist of a bipartite spin lattice. Classically this means that atoms with nonzero spin on sublattice A are surrounded by neighboring atoms on sublattice B whose spins are *antiparallel*, because this minimizes the isotropic Heisenberg exchange interaction $\mathcal{H} = JS_A \cdot S_B$ between the atoms on A and their neighbors on B when the exchange integral J is positive. If the atoms on A have vacuum state $|s\rangle$, then the atoms on B have the vacuum state $|-s\rangle$ and creating bosons *increases* the value of S_z . The boson creation (annihilation) operator on sublattice B is \hat{b}^\dagger (\hat{b}) and the number operator now becomes

$$\hat{n}_b|m_s\rangle = \hat{b}^\dagger\hat{b}|m_s\rangle = (s+m_s)|m_s\rangle. \quad (4.12)$$

This yields

$$\hat{S}_z|m_s\rangle = m_s|m_s\rangle = (-s + s + m_s)|m_s\rangle = (-s + \hat{b}^\dagger\hat{b})|m_s\rangle, \quad (4.13)$$

$$\hat{S}_+ = \sqrt{2s}\hat{b}^\dagger\sqrt{1 - \frac{\hat{n}_b}{2s}}, \quad (4.14)$$

$$\hat{S}_- = \sqrt{2s}\sqrt{1 - \frac{\hat{n}_b}{2s}}\hat{b}. \quad (4.15)$$

Now expand in a Maclaurin series, keeping only the first term:

$$\hat{S}_+^A \approx \sqrt{2s}\hat{a}, \quad \hat{S}_-^A \approx \sqrt{2s}\hat{a}^\dagger, \quad \hat{S}_+^B \approx \sqrt{2s}\hat{b}^\dagger, \quad \hat{S}_-^B \approx \sqrt{2s}\hat{b}. \quad (4.16)$$

We rewrite the Hamiltonian as a sum over every spin's interaction with its neighbors

$$\mathcal{H} = \frac{J}{2} \sum_j \sum_\delta \hat{S}_j \cdot \hat{S}_{j+\delta}, \quad (4.17)$$

where δ is a vector connecting a spin on site j with its nearest neighbors. Using the definition of \hat{S}_+ and \hat{S}_- as in (2.10), we get

$$\mathcal{H} = \frac{J}{2} \sum_j \sum_\delta \left[\hat{S}_{j,z}\hat{S}_{j+\delta,z} + \frac{1}{2} (\hat{S}_{j,+}\hat{S}_{j+\delta,-} + \hat{S}_{j,-}\hat{S}_{j+\delta,+}) \right]. \quad (4.18)$$

Naturally, if the spin on site j is in sublattice A, then the neighbors are on sublattice B, and vice versa. Hence the Hamiltonian can be expressed as a sum over sublattice A only:

$$\mathcal{H} = -\frac{z_N s^2 J N}{2} + \frac{sJ}{2} 2 \sum_j \sum_\delta \left[\hat{a}_j^\dagger \hat{a}_j + \hat{b}_{j+\delta}^\dagger \hat{b}_{j+\delta} + \hat{a}_j^\dagger \hat{b}_{j+\delta}^\dagger + \hat{a}_j \hat{b}_{j+\delta} \right], \quad (4.19)$$

where j is now a lattice site on A, z_N is the number of nearest neighbors, N is the total number of spins and the factor 2 in front of the summation comes from the need to count sublattice B as well. I discarded all the non-linear terms since we are looking for a linear theory (we already discarded all non-linear terms in the Maclaurin expansion, so we can only keep the linear terms in the Hamiltonian if we want to be consistent).

Straight away we notice that the first term in the Hamiltonian is the classical ground state energy of the Néel state. However, the Néel state is not an eigenstate of the Hamiltonian because the third term in the sum creates a boson on all sites if applied to this state. Thus the Neel state is not the true quantum mechanical ground state, and the ground state energy must be different from $E_0^{Classic} = -\frac{z_N s^2 J N}{2}$. Worse, it is not immediately obvious what the quantum mechanical ground state energy is because the Hamiltonian is very far from diagonal in the basis $|m_{s,1} m_{s,2} \cdots m_{s,N}\rangle$: in addition to the boson-creating third term, there is an annihilating fourth term that also creates off-diagonal elements in the matrix.

In Chapter 1 I mentioned how spin excitations in antiferromagnets manifest themselves as a small deflection of each spin from the antiparallel position, and that these deflections trace out a wave in the spin lattice (see Fig. 1.2). If the quantum mechanical ground state is *close* to the Néel state, it seems reasonable that it will contain a small number of spin waves, so a natural next step is to make a Fourier transform of the Hamiltonian. I am getting tired of "hatting" all the operators, so from now on the

hat will be omitted – it should be clear from the context which symbols are operators and which are not. The creation and annihilation operators can be expressed in terms of their complex Fourier expansions as

$$a_j^\dagger = \sqrt{\frac{1}{N_{unit}}} \sum_k \exp [ik \cdot \mathbf{R}_j] a_k^\dagger, \quad (4.20)$$

$$a_j = \sqrt{\frac{1}{N_{unit}}} \sum_k \exp [-ik \cdot \mathbf{R}_j] a_k, \quad (4.21)$$

$$b_j^\dagger = \sqrt{\frac{1}{N_{unit}}} \sum_k \exp [-ik \cdot \mathbf{R}_j] b_{-k}^\dagger, \quad (4.22)$$

$$b_j = \sqrt{\frac{1}{N_{unit}}} \sum_k \exp [ik \cdot \mathbf{R}_j] b_{-k}, \quad (4.23)$$

where N_{unit} is the number of unit cells consisting of one spin up and one spin down site ($N_{unit} = \frac{N}{2}$). Inserting these expansions into (4.19) yields

$$\mathcal{H} = -\frac{z_N S^2 J N}{2} + z_N J s \sum_k \left(a_k^\dagger a_k + b_{-k}^\dagger b_{-k} + \gamma_k a_k b_{-k} + \gamma_k b_{-k}^\dagger a_k^\dagger \right), \quad (4.24)$$

where $\gamma_k = \frac{1}{z_N} \sum_\delta \exp [ik \cdot \delta]$.

So far it looks like we have not really accomplished much because the Hamiltonian still contains off-diagonal $a_k b_{-k}$ and $b_{-k}^\dagger a_k^\dagger$ terms. We continue by applying the Bogoliubov transformation, which assumes that the eigenvectors of \mathcal{H} are linear combinations of spin waves on sublattice A with wave vector k and spin waves on sublattice B with the opposite wave vector. The new operators are

$$\alpha_k = u_k a_k - v_k b_{-k}^\dagger, \quad (4.25)$$

$$\beta_{-k} = u_k b_{-k} - v_k a_k^\dagger. \quad (4.26)$$

These operators must satisfy the Bose commutation relations

$$[\alpha_k, \alpha_k^\dagger] = 1, \quad [\beta_k, \beta_k^\dagger] = 1. \quad (4.27)$$

All other pairs of operators in the set $\{\alpha_k, \alpha_k^\dagger, \beta_{-k}, \beta_{-k}^\dagger\}$ commute. The Bose commutators require that

$$u_k^2 - v_k^2 = 1, \quad (4.28)$$

so both u_k and v_k can be taken to be real without loss of generality. Then

$$a_k = u_k \alpha_k + v_k \beta_{-k}^\dagger, \quad (4.29)$$

$$b_{-k} = u_k \beta_{-k} + v_k \alpha_k^\dagger, \quad (4.30)$$

which produce the Hamiltonian

$$\begin{aligned} \mathcal{H} = & -\frac{z_N s^2 J N}{2} \\ & + z_N J s \sum_k (u_k^2 + v_k^2 + 2\gamma_k u_k v_k) (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1) \\ & + z_N J s \sum_k [\gamma_k (u_k^2 + v_k^2) + 2u_k v_k] (\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger) \\ & - z_N J s \sum_k 1. \end{aligned} \quad (4.31)$$

We see that the off-diagonal terms vanish if

$$\gamma_k (u_k^2 + v_k^2) + 2u_k v_k = 0. \quad (4.32)$$

Rearranging this equation, we obtain

$$\frac{\gamma_k}{2} \left(\frac{u_k}{v_k} \right)^2 + \frac{u_k}{v_k} + \frac{\gamma_k}{2} = 0, \quad (4.33)$$

from which it follows that

$$\frac{u_k}{v_k} = \frac{1}{\gamma_k} \left(-1 \pm \sqrt{1 - \gamma_k^2} \right). \quad (4.34)$$

From (4.28) we have

$$v_k^2 \left[\left(\frac{u_k}{v_k} \right)^2 - 1 \right] = 1, \quad (4.35)$$

which after we insert (4.34) and do the algebra yields

$$u_k^2 + v_k^2 = \pm \frac{1}{\sqrt{1 - \gamma_k^2}}. \quad (4.36)$$

I have already taken u_k and v_k to be real, so the solution with the minus sign must be discarded. We can now use (4.32) and (4.36) to rewrite the Hamiltonian in (4.31) as

$$\mathcal{H} = -\frac{z_N s^2 J N}{2} - z_N J s \sum_k 1 + z_N J s \sum_k \sqrt{1 - \gamma_k^2} (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1). \quad (4.37)$$

Noting that the number of k -vectors in the magnetic Brillouin zone is $N_{unit} = \frac{N}{2}$, we have the Hamiltonian in compact form:

$$\mathcal{H} = -\frac{z_N s(s+1)JN}{2} + \sum_k \omega_k (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1), \quad (4.38)$$

where $\omega_k = z_N J s \sqrt{1 - \gamma_k^2}$ is the energy of an excitation. A contour map of ω_k for a square lattice is shown in in Fig. 4.1.

We have now accomplished what we set out to do: the Hamiltonian is diagonal in the basis $|n_{\alpha_k}\rangle |n_{\beta_{-k}}\rangle$. Evidently, even if we make sure that there are absolutely no α_k - or β_{-k} -magnons present (i.e. the antiferromagnet is in its ground state), the magnet still behaves like it is host to one magnon with energy ω_k for every k , as seen in the

last term of (4.38). This is a purely quantum mechanical phenomenon, and these spin fluctuations in the ground state are therefore called quantum fluctuations. They are present even when the temperature is at absolute zero.

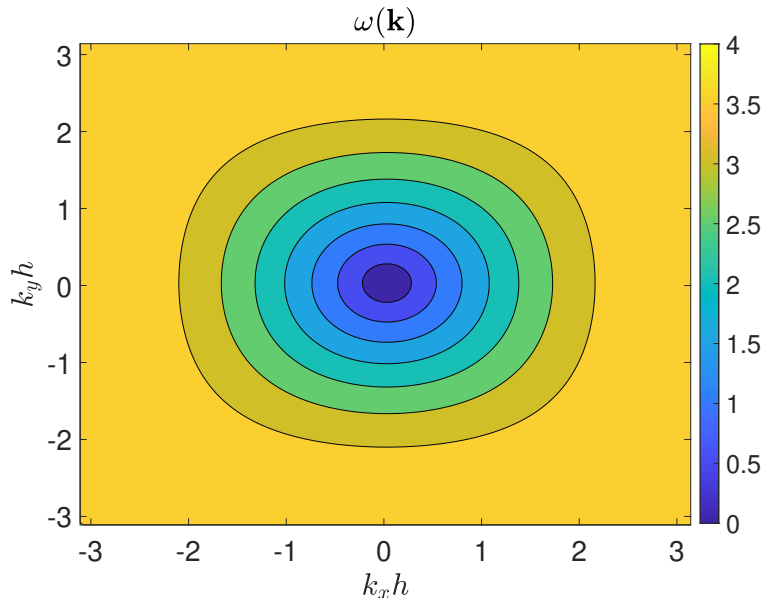


FIGURE 4.1: Contour plot of the dispersion $\omega(\mathbf{k})$ over the Brillouin zone for a square lattice in units of Js. The Brillouin zone and the axis labels are discussed in Section 4.2.

The presence of spin fluctuations raises a rather troubling question: how much fluctuation can the antiferromagnet bear without destroying the antiferromagnetic ordering of the spins? We know that thermal fluctuations cause an antiferromagnet to lose its ordering when the temperature reaches the Néel temperature. It is not unthinkable that the ground state fluctuations would effectively lower the Néel temperature to absolute zero, i.e. ordering of spins is not possible at any temperature. We can check if the ordering is preserved by looking at the average value of the z-component of S in one of the sublattices. Picking sublattice A for the calculation, we have

$$\frac{2}{N} \sum_{j \in A} \langle S_j^z \rangle = \frac{2}{N} \sum_{\mathbf{k}} (s - \langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \rangle), \quad (4.39)$$

where I used (4.9) and the Fourier expansions of the a_j, a_j^\dagger operators. As expected, the absence of $a_{\mathbf{k}}$ -magnons yields the classical result of maximum alignment with the z-axis. We may therefore write

$$\frac{2}{N} \sum_{j \in A} \langle S_j^z \rangle = s - \frac{2}{N} \sum_{\mathbf{k}} \langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \rangle = s - \Delta \bar{S}_A^z, \quad (4.40)$$

where $\Delta \bar{S}_A^z$ is the quantum correction to the classical result. For us to have a hope that the magnetic ordering is preserved, we need $\Delta \bar{S}_A^z$ to be smaller than s . To evaluate $\Delta \bar{S}_A^z$ we need to express $\langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \rangle$ in terms $\alpha_{\mathbf{k}}$ and $\beta_{\mathbf{k}}$ using (4.25):

$$\langle a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \rangle = u_{\mathbf{k}}^2 \langle \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} \rangle + v_{\mathbf{k}}^2 \langle \beta_{-\mathbf{k}}^\dagger \beta_{-\mathbf{k}} \rangle - u_{\mathbf{k}} v_{\mathbf{k}} \langle \alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}}^\dagger \rangle - u_{\mathbf{k}} v_{\mathbf{k}} \langle \alpha_{\mathbf{k}} \beta_{-\mathbf{k}} \rangle + v_{\mathbf{k}}^2. \quad (4.41)$$

But $\langle \alpha_k^\dagger \beta_{-k}^\dagger \rangle = \langle \alpha_k \beta_{-k} \rangle = 0$, so

$$\Delta \bar{S}_A^z = \frac{2}{N} \sum_k u_k^2 \langle \alpha_k^\dagger \alpha_k \rangle + v_k^2 \langle \beta_{-k}^\dagger \beta_{-k} \rangle + v_k^2. \quad (4.42)$$

We assume there are equally many α_k - and β_{-k} -magnons in the magnet (the number of these magnons is zero in the ground state, of course) and use (4.34), (4.35) and (4.36) to obtain

$$\Delta \bar{S}_A^z = -\frac{1}{2} + \frac{2}{N} \sum_k \frac{1}{\sqrt{1 - \gamma_k^2}} \left(\langle \alpha_k^\dagger \alpha_k \rangle + \frac{1}{2} \right). \quad (4.43)$$

Our theory predicts stable antiferromagnets if the last term in (4.43) converges to a value smaller than $s + \frac{1}{2}$ when converted to an integral over the Brillouin zone. The integral thus depends on the geometry of the magnet. In 1D there is only one possible lattice (the linear lattice), and if we do the integral we will find that it diverges for any $\langle \alpha_k^\dagger \alpha_k \rangle$ [28]. The conclusion must be that antiferromagnetic ordering in 1D is simply impossible with our crude theory.

The two-dimensional situation is somewhat more complicated. The integral obtained from (4.43) is not in general analytically solvable, but it can be shown [28] that ordering *is* possible in the presence of quantum fluctuations. However, any excited magnons will destroy the ordering. To put it another way: antiferromagnetic ordering is possible at zero temperature, but any nonzero temperature will immediately destroy it. This might seem devastating to the project of describing dynamics in two-dimensional antiferromagnets, which relies on the excitation of magnons. Yet we know experimentally that two-dimensional magnetic materials do exist above zero temperature [29, 30]. There must be something missing from our model that would provide an explanation for these observations.

As was briefly mentioned in Chapter 2, there is some degree of coupling between the spin and orbital angular momenta of the electrons in materials. When the spin of an electron is reoriented, the atomic orbital is also reoriented slightly because of this spin-orbit coupling. In most materials, though, there are some configurations of the atomic orbitals that give the maximum amount of energetically favourable overlap, and displacing the atomic orbitals from this configuration will be costly. The extra energy needed to rotate the spins out of their equilibrium orientation is called magnetocrystalline anisotropy. Often there is one crystallographic axis that is preferable to any other axis for the spins to orient themselves along, and we call this axis the easy axis. It is very natural to choose this axis as the quantization axis (the z-axis). We account for an easy axis anisotropy by adding two anisotropy terms to the Heisenberg Hamiltonian. It now reads

$$\mathcal{H} = \frac{1}{2} \sum_j \sum_\delta \left[J \mathbf{S}_j \cdot \mathbf{S}_{j+\delta} + K \left(S_j^z \right)^2 + K \left(S_{j+\delta}^z \right)^2 \right], \quad (4.44)$$

where $K < 0$ is the easy axis anisotropy constant. Swapping the spin operators with Bose operators and discarding all terms that contain more than two operators, we get

$$\mathcal{H}_0 = \sum_j \sum_\delta \left[(2K - J) s^2 + (J - 2K) s a_j^\dagger a_j + (J - 2K) s b_{j+\delta}^\dagger b_{j+\delta} + J s a_j^\dagger b_{j+\delta}^\dagger + J s a_j b_{j+\delta} \right]. \quad (4.45)$$

As before, we continue by replacing the Bose operators with their Fourier series expansions:

$$\begin{aligned} \mathcal{H}_0 = & -\frac{z_N s^2 (J - 2K) N}{2} \\ & + z_N J s \sum_k \left[\left(1 - \frac{2K}{J}\right) (a_k^\dagger a_k + b_{-k}^\dagger b_{-k}) + \gamma_k (a_k^\dagger b_{-k}^\dagger + a_k b_{-k}) \right] \end{aligned} \quad (4.46)$$

We will set $x = \frac{2K}{J}$ for neater notation and apply the Bogoliubov transformation:

$$\begin{aligned} \mathcal{H}_0 = & -\frac{z_N s^2 (J - 2K) N}{2} \\ & + z_N J s \sum_k ((1-x)u_k^2 + (1-x)v_k^2 + 2\gamma_k u_k v_k) (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1) \\ & + z_N J s \sum_k [\gamma_k (u_k^2 + v_k^2) + 2(1-x)u_k v_k] (\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger) \\ & - z_N J s \sum_k (1-x). \end{aligned} \quad (4.47)$$

The off-diagonal terms vanish if

$$\gamma_k (u_k^2 + v_k^2) + 2(1-x)u_k v_k = 0 \quad (4.48)$$

or, dividing the equation by $2v_k^2$,

$$\frac{\gamma_k}{2} \left(\frac{u_k}{v_k}\right)^2 + (1-x)\frac{u_k}{v_k} + \frac{\gamma_k}{2} = 0. \quad (4.49)$$

Use of the quadratic formula yields

$$\frac{u_k}{v_k} = \frac{1}{\tilde{\gamma}_k} \left(1 \pm \sqrt{1 - \tilde{\gamma}_k^2}\right), \quad (4.50)$$

where $\tilde{\gamma}_k = \frac{\gamma_k}{1-x}$. This means

$$u_k^2 + v_k^2 = \frac{1}{\sqrt{1 - \tilde{\gamma}_k^2}}, \quad (4.51)$$

which leaves us with the following Hamiltonian:

$$\mathcal{H} = -\frac{z_N s (s+1) \tilde{J}}{2} + \sum_k \omega_k (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1), \quad (4.52)$$

where $\tilde{J} = J - 2K$ and $\omega_k = z_N \tilde{J} s \sqrt{1 - \tilde{\gamma}_k^2}$ is the energy of an excitation. Interestingly, the addition of an easy axis magnetocrystalline anisotropy can be accounted for simply by using

$$J \rightarrow \tilde{J}, \quad \gamma_k \rightarrow \tilde{\gamma}_k. \quad (4.53)$$

The introduction of anisotropy also takes care of any potential problems with dividing by $\sqrt{1 - \tilde{\gamma}_k^2}$, since $\tilde{\gamma}_k < 1$ for all k . Because of the triviality of adding anisotropy to the calculations, I will not be taking this into consideration for the remainder of Chapters

4 and 5. If the reader feels uneasy about potential divergence problems of the form (4.43), use the replacements (4.53).

4.2 Comparison to the Néel state

To illustrate the effect of quantum fluctuations we will calculate the ground state energy of a square lattice antiferromagnet and compare it to the Néel state energy. The lattice and the magnetic unit cell is shown in Fig. 4.2. In terms of magnetic unit cells the square lattice looks like Fig. 4.3, where we define a coordinate system so that the nearest neighbor vectors are (with hats indicating unit vectors now!)

$$\delta_1 = \frac{1}{2}h\hat{x} + \frac{1}{2}h\hat{y} \quad (4.54)$$

$$\delta_2 = -\frac{1}{2}h\hat{x} - \frac{1}{2}h\hat{y} \quad (4.55)$$

$$\delta_3 = \frac{1}{2}h\hat{x} - \frac{1}{2}h\hat{y} \quad (4.56)$$

$$\delta_4 = -\frac{1}{2}h\hat{x} + \frac{1}{2}h\hat{y} \quad (4.57)$$

where $h = \frac{\sqrt{2}}{2}a$ and $\frac{a}{2}$ is the lattice constant. Then

$$\begin{aligned} \gamma_{\mathbf{k}} &= \frac{1}{4} \exp \left[i\frac{h}{2} (k_x + k_y) \right] + \exp \left[-i\frac{h}{2} (k_x + k_y) \right] \\ &\quad + \frac{1}{4} \exp \left[i\frac{h}{2} (k_x - k_y) \right] + \exp \left[-i\frac{h}{2} (k_x - k_y) \right] \\ &= \frac{1}{2} \left(\cos \left[\frac{h}{2} (k_x + k_y) \right] + \cos \left[\frac{h}{2} (k_x - k_y) \right] \right). \end{aligned} \quad (4.58)$$

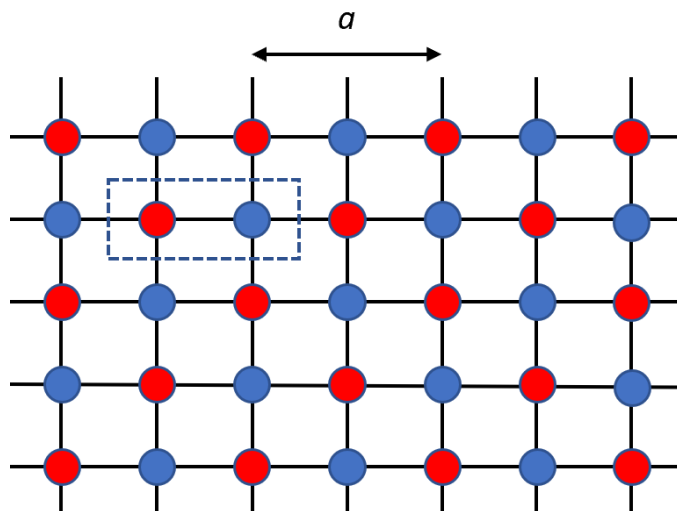


FIGURE 4.2: A part of a simple, square antiferromagnet with red circles indicating sublattice A sites and blue circles indicating sublattice B sites. The magnetic unit cell is shown with dotted lines.

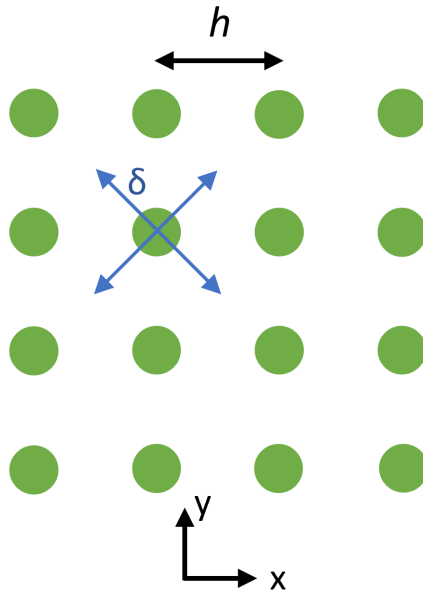


FIGURE 4.3: The lattice of unit cells is square with lattice constant $h = \frac{\sqrt{2}}{2}a$ and the nearest neighbor vectors δ make an angle $\frac{\pi}{4}$ with both the x -axis and the y -axis.

The number operators $\alpha_k^\dagger \alpha_k$ and $\beta_{-k}^\dagger \beta_{-k}$ will yield 0 in the ground state, so the ground state energy per lattice site is

$$\frac{E_0}{N} = -\frac{4s(s+1)J}{2} + \frac{4Js}{N} \sum_{k \in \text{BZ}} \sqrt{1 - \frac{1}{4} \left(\cos \left[\frac{h}{2} (k_x + k_y) \right] + \cos \left[\frac{h}{2} (k_x - k_y) \right] \right)^2}, \quad (4.59)$$

where the sum runs over all k -vectors in the Brillouin zone, which is described by $k_x, k_y \in [0, \frac{2\pi}{h})$. Our strategy for evaluating the sum over all k -states in the Brillouin zone will be to convert the sum into an integral and evaluate the integral numerically. There are $\frac{N}{2}$ states in the Brillouin zone and the total area of the Brillouin zone is $(\frac{2\pi}{h})^2 = \frac{4\pi^2}{h^2}$, so the area of one state is $\frac{8\pi^2}{Nh^2}$. Thus

$$\sum_{k \in \text{BZ}} \sqrt{1 - \gamma_k^2} \rightarrow \frac{Nh^2}{8\pi^2} \iint_{\text{BZ}} \sqrt{1 - \frac{1}{4} \left(\cos \left[\frac{h}{2} (k_x + k_y) \right] + \cos \left[\frac{h}{2} (k_x - k_y) \right] \right)^2} dA. \quad (4.60)$$

We can make the substitutions $u_x = \frac{h}{2}k_x$, $u_y = \frac{h}{2}k_y$ so that the integral becomes

$$\sum_{k \in \text{BZ}} \sqrt{1 - \gamma_k^2} \approx \frac{N}{2\pi^2} \int_0^\pi \int_0^\pi \sqrt{1 - \frac{1}{4} (\cos [u_x + u_y] + \cos [u_x - u_y])^2} du_x du_y. \quad (4.61)$$

Using WolframAlpha or another numerical integration tool we find that the approximate numerical value of (4.61) is 0.4210. Thus the energy difference between the ground state and the Néel state is

$$\Delta E_{\text{GS}} = \frac{E_0}{N} - \frac{E_0^{\text{Classic}}}{N} = \left(-\frac{4Js(s+1)}{2} + \frac{4Js}{N} 0.4210 \right) - \left(-\frac{4s^2J}{2} \right) = -0.3159Js. \quad (4.62)$$

The result is in perfect agreement with that originally obtained by Anderson [31].

Chapter 5

Ultrafast 2-Magnon Excitation in Antiferromagnets

In the previous chapter we saw how our efforts to diagonalize the Hamiltonian led to the introduction of operators (α_k and β_{-k}) that combine excitations and annihilation of magnons on the two sublattices of bipartite antiferromagnets. Taking this as a starting point, we will now look at what happens to the Hamiltonian when we induce small changes in the exchange interaction $J \rightarrow J + \Delta J$ by shining light on the antiferromagnet. Theoretical calculations and experiments [18, 32] have shown that light enables us to control the spin dynamics in antiferromagnets. The Hamiltonian that includes the light-induced modification will motivate us to introduce new operators that work on magnon pairs. Our goal will ultimately be to find a set of coherent states – which, rather awkwardly, are not coherent in the traditional sense but more like squeezed states – and look at the dynamics of the quantum mechanical Néel vector, which we will see reveals some interesting features of the quantum description of the antiferromagnet. From the coherent states we will be able to extract a parameter that captures the dynamics on the femtosecond timescale, and whose time-evolution can be treated classically to give a set of new equations to complement the Landau-Lifshitz equations, yielding a complete description of antiferromagnetic dynamics for a simple Heisenberg Hamiltonian.

5.1 Hamiltonian with light-spin interaction

The exchange interaction J that is the source of ferro- and antiferromagnetism can be derived from the Hubbard model of interacting spin particles on a lattice. The Hubbard model can be thought of as an extension of the tight-binding model into the second quantization regime, with the inclusion of particle-particle interactions. A full derivation of the exchange interaction perturbation ΔJ from the model is nearly a master's thesis in its own right [33]. I will therefore only present a short description of the model and qualitatively show how its mapping to the Heisenberg model reveals the nature of the exchange interaction.

5.1.1 A peak at the Hubbard model

We will consider a regular lattice of atoms on which electrons can reside. Each atom has one orbital available for electrons to enter or leave (all the lower-lying orbitals are already filled with bound electrons). Since electrons are fermions obeying the Pauli exclusion principle, each atom can accommodate up to two electrons. An atom can therefore host no electrons, one spin up electron, one spin down electron, or two electrons with opposite spins. For atoms with two electrons there is an associated

energy from the Coulombic repulsion of the negative charges. Thus the Hubbard Hamiltonian will contain the potential

$$V_{int} = U \sum_j n_{j,\uparrow} n_{j,\downarrow} \quad (5.1)$$

where U is the onsite Coulombic repulsive energy and $n_{j,\sigma}$ is the number of electrons with spin $\sigma \in \{\uparrow, \downarrow\}$ at site j . We assume that there is no Coulombic interaction between electrons on different atoms.

The electrons are free to move around on the lattice, so we need a kinetic term in our Hamiltonian as well. When an electron "hops" from one atom to the next, we can model this by destroying an electron on one atom and creating an electron on a neighboring atom. The operator $c_{j,\sigma}^\dagger c_{j+\delta,\sigma}$ accomplishes this, with $c_{j,\sigma}^\dagger$ now being the *fermionic* creation operator of an electron with spin σ on site j . The adjoint operator $c_{j,\sigma}$ is of course the fermionic destruction operator. We also will not allow an individual electron to change its spin.

The ease with which an electron can move between atoms will depend on the overlap between the atomic orbitals. Since an atomic orbital is centered on its atom and decays exponentially, we assume that only orbitals on nearest-neighbor atoms overlap. The energy associated with the "hopping" of an electron from an atom to another is a constant denoted by t_0 . The delocalization of electrons lowers the free energy of the material, so the kinetic term will be negative. Adding the kinetic and the Coulombic terms yields the Hubbard Hamiltonian:

$$\mathcal{H}_U = -t_0 \sum_{i,\delta,\sigma} c_{i,\sigma}^\dagger c_{i+\delta,\sigma} + U \sum_j n_{j,\uparrow} n_{j,\downarrow}. \quad (5.2)$$

Assume now that we have two atoms and two electrons of opposite spin. There are four ways to arrange the electrons, and we express them as kets:

$$\begin{aligned} &|\uparrow, \downarrow\rangle \\ &|\downarrow, \uparrow\rangle \\ &|\uparrow\downarrow, 0\rangle \\ &|0, \uparrow\downarrow\rangle. \end{aligned}$$

Using this basis, the Hubbard Hamiltonian can be written in matrix form as

$$\mathcal{H}_U = \begin{bmatrix} 0 & 0 & -t_0 & -t_0 \\ 0 & 0 & -t_0 & -t_0 \\ -t_0 & -t_0 & U & 0 \\ -t_0 & -t_0 & 0 & U \end{bmatrix}. \quad (5.3)$$

We can use a computer program to diagonalize the matrix for us (again, WolframAlpha is a convenient tool). We find $\mathcal{H}_U = PDP^{-1}$ with

$$D = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & U & 0 & 0 \\ 0 & 0 & \frac{U}{2} - \frac{U}{2} \sqrt{1 + \frac{16t_0^2}{U^2}} & 0 \\ 0 & 0 & 0 & \frac{U}{2} + \frac{U}{2} \sqrt{1 + \frac{16t_0^2}{U^2}} \end{bmatrix}. \quad (5.4)$$

The unitary matrix P is of no importance to us, since we are only interested in the eigenvalues of (5.3). We can expand the roots in (5.4) as a Maclaurin series $\sqrt{1+x^2} \approx$

$1 + \frac{x^2}{2}$ provided $U \gg t_0$ so that the eigenvalues are approximately

$$-\frac{4t_0^2}{U}, \quad 0, \quad U, \quad U + \frac{4t_0^2}{U}.$$

Thus the difference between the ground state and the first excited state is $\frac{4t_0^2}{U}$.

We will now see what we get if we use the Heisenberg Hamiltonian to find the energy difference between the ground state and the first excited state. For two interacting spins we get from (4.18):

$$\mathcal{H} = J \left[S_1^z S_2^z + \frac{1}{2} (S_1^+ S_2^- + S_1^- S_2^+) \right]. \quad (5.5)$$

The eigenstates should be familiar to anyone who has studied introductory quantum mechanics. They are

$$\text{triplet : } \begin{cases} |\uparrow, \uparrow\rangle \\ \frac{1}{\sqrt{2}} (|\uparrow, \downarrow\rangle + |\downarrow, \uparrow\rangle) \\ |\downarrow, \downarrow\rangle \end{cases} \quad \text{singlet : } \frac{1}{\sqrt{2}} (|\uparrow, \downarrow\rangle - |\downarrow, \uparrow\rangle).$$

The eigenvalues are $\frac{J}{4}$ for the triplet states and $-\frac{3J}{4}$ for the singlet state. So the difference is J and thus, upon comparison with the results from the Hubbard model, we get $J = \frac{4t_0^2}{U}$.

5.1.2 Light-induced perturbation to the exchange interaction

When light is interacting with the lattice, we model this by including in the hopping term a phase factor $\exp[ie\mathbf{A} \cdot \boldsymbol{\delta}]$ so that

$$t_0 \rightarrow t_0 \exp[ie\mathbf{A} \cdot \boldsymbol{\delta}], \quad (5.6)$$

where e is the elementary charge and $\mathbf{A} = \frac{\partial \mathbf{E}}{\partial t}$ is the vector potential of the electric field. For monochromatic laser light the field is simply $\mathbf{E}(t) = \mathbf{E}_0 \sin \omega t$ with \mathbf{E}_0 the electric field amplitude, so the vector potential is $\mathbf{A}(t) = -\frac{1}{\omega} \mathbf{E}_0 \cos \omega t$. Substituting (5.6) into the Hubbard Hamiltonian (5.2), the task is then to find the energy spectrum similarly to what we did for the unperturbed two-particle case above, but for a large number of particles.

In the derivation by Mentink, Balzer and Eckstein [34], the Hubbard Hamiltonian is brought into Fourier space and Floquet theory is used to find the energy spectrum. In the limit where the electric field amplitude is small compared to the electromagnetic energy, by which I mean $e\mathbf{E}_0 \cdot \boldsymbol{\delta} \ll \hbar\omega$, and the onsite Coulombic interaction is much stronger than the hopping integral, $U \gg t_0$, it is possible to simplify the Hubbard Hamiltonian to the point where standard perturbation theory on the hopping integral yields [34]

$$\Delta J = \frac{t_0^2}{2U} \frac{(e\mathbf{E}_0 \cdot \boldsymbol{\delta})^2}{U^2 - \hbar^2 \omega^2}. \quad (5.7)$$

We see that the sign of the perturbation changes when the energy of the light goes from being smaller than U to greater than U . Also worth noting is that this model breaks down for photon energies close to the band gap $\hbar\omega \approx U$.

5.1.3 The perturbed Hamiltonian

When we insert the exchange perturbation ΔJ into the Heisenberg Hamiltonian, we get

$$\mathcal{H} = \frac{J}{2} \sum_{j,\delta} \hat{\mathbf{S}}_j \cdot \hat{\mathbf{S}}_{j+\delta} + \frac{1}{2} f(t) \sum_{j,\delta} \Delta J(\delta) \hat{\mathbf{S}}_j \cdot \hat{\mathbf{S}}_{j+\delta} = \mathcal{H}_0 + \delta \mathcal{H}, \quad (5.8)$$

where the time profile of the light pulse is captured by the function f whose maximum amplitude is normalized to 1. The unperturbed Hamiltonian \mathcal{H}_0 we already diagonalized using the operators α_k and β_{-k} . We should therefore try to write $\delta \mathcal{H}$ in terms of these operators as well. Luckily, the perturbation looks very much like the unperturbed Hamiltonian, so we can apply the Holstein-Primakoff transformation to get

$$\begin{aligned} \delta \mathcal{H} = & -f(t) \frac{s^2 N}{2} \sum_{\delta} \Delta J(\delta) \\ & + f(t) \frac{s}{2} 2 \sum_j \sum_{\delta} \Delta J(\delta) \left[a_j^\dagger a_j + b_{j+\delta}^\dagger b_{j+\delta} + a_j^\dagger b_{j+\delta}^\dagger + a_j b_{j+\delta} \right]. \end{aligned} \quad (5.9)$$

We continue as before by expressing the Bose operators in the form of their complex Fourier series expansions given in (4.20)-(4.23). This yields

$$\begin{aligned} \delta \mathcal{H} = & -f(t) \frac{s^2 N}{2} \sum_{\delta} \Delta J(\delta) \\ & + f(t) \frac{s}{2} 2 \sum_k \sum_{\delta} \Delta J(\delta) \left[a_k^\dagger a_k + b_{-k}^\dagger b_{-k} + \exp[i\mathbf{k} \cdot \delta] \left(a_k^\dagger b_{-k}^\dagger + a_k b_{-k} \right) \right]. \end{aligned} \quad (5.10)$$

The next step is to apply the Bogoliubov transformation:

$$\begin{aligned} \delta \mathcal{H} = & -f(t) \frac{s^2 N}{2} \sum_{\delta} \Delta J(\delta) \\ & + f(t) s \sum_k \sum_{\delta} \Delta J(\delta) (u_k^2 + v_k^2 + 2 \exp[i\mathbf{k} \cdot \delta] u_k v_k) \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) \\ & + f(t) s \sum_k \sum_{\delta} \Delta J(\delta) [\exp[i\mathbf{k} \cdot \delta] (u_k^2 + v_k^2) + 2u_k v_k] \left(\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger \right) \\ & - f(t) s \sum_k \sum_{\delta} \Delta J(\delta). \end{aligned} \quad (5.11)$$

Now let

$$\Lambda = \sum_{\delta} \Delta J(\delta) \quad (5.12)$$

and

$$\tilde{\zeta}_k = \frac{1}{\Lambda} \sum_{\delta} \Delta J(\delta) \exp[i\mathbf{k} \cdot \delta]. \quad (5.13)$$

Using (4.36) and (4.32) linking u_k and v_k to γ_k , we get

$$\begin{aligned} \delta \mathcal{H} = & -\frac{f(t)\Lambda s(s+1)N}{2} \\ & + f(t)\Lambda s \sum_k \frac{1 - \tilde{\zeta}_k \gamma_k}{\sqrt{1 - \gamma_k^2}} \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) \\ & + f(t)\Lambda s \sum_k \frac{\tilde{\zeta}_k - \gamma_k}{\sqrt{1 - \gamma_k^2}} \left(\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger \right) \end{aligned} \quad (5.14)$$

We see that the operators α_k and β_{-k} do *not* diagonalize the perturbation to the Heisenberg Hamiltonian unless $\tilde{\zeta}_k = \gamma_k$. The picture becomes a little clearer if we collect constants so that $\Omega = z_N J s$ and $\delta\omega_R = \Lambda s$ and define

$$\delta\omega_k = \delta\omega_R \frac{1 - \tilde{\zeta}_k \gamma_k}{\sqrt{1 - \gamma_k^2}}, \quad (5.15)$$

$$V_k = \delta\omega_R \frac{\tilde{\zeta}_k - \gamma_k}{\sqrt{1 - \gamma_k^2}}. \quad (5.16)$$

The entire Hamiltonian can then be written

$$\begin{aligned} \mathcal{H} = & -\frac{(\Omega + f(t)\delta\omega_R) N (s+1)}{2} \\ & + \sum_k (\omega_k + f(t)\delta\omega_k) \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) \\ & + \sum_k f(t)V_k \left(\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger \right). \end{aligned} \quad (5.17)$$

In short, the perturbation shifts the reference energy (first term), shifts the frequency of magnons that are already present in the antiferromagnet (first sum), and excites and annihilates pairs of magnons (second sum). The frequency shift disappears after the light is turned off.

Before we conclude this section we will briefly discuss the light polarization dependence of the parameters $\delta\omega_k$ and V_k . First of all we notice that the z -component of the electric field has no effect on neither Λ nor $\tilde{\zeta}_k$, and hence any electric field perpendicular to the xy -plane will not induce any excitations of magnons. Moreover, the value of $\delta\omega_R$ is independent of the polarization in the xy -plane for both square and hexagonal lattices. The only anisotropy is found in the term $\tilde{\zeta}_k$, which is a result of the non-circular shape of the magnetic Brillouin zone. More pronounced polarization dependence could likely be derived by including anisotropic interactions such as the spin-orbit interaction and magnetoelastic strain the model, but is beyond the scope of this discussion.

5.2 Magnon pair operators

We see from the third line in the Hamiltonian (5.17) that the magnon excitations always come in pairs of α_k and β_{-k} magnons. The eigenstates of the number operators $\alpha_k^\dagger \alpha_k$ and $\beta_{-k}^\dagger \beta_{-k}$ are $|n_{\uparrow,k}\rangle$ and $|n_{\downarrow,-k}\rangle$, respectively. The reason I have included the spin states \uparrow and \downarrow in the notation is that for magnons close to the edge of the Brillouin zone, the oscillations are predominantly localized to one sublattice only. To see this

we can add (4.28) and (4.36) to get

$$2u_k^2 = 1 + \frac{1}{\sqrt{1 - \gamma_k^2}}. \quad (5.18)$$

This implies that $u_k \approx \pm 1$ if γ_k^2 is close to zero. The value of γ_k^2 for a square lattice is plotted as a contour map in Fig. 5.1 for $k_x, k_y \in [-\frac{\pi}{h}, \frac{\pi}{h}]$.

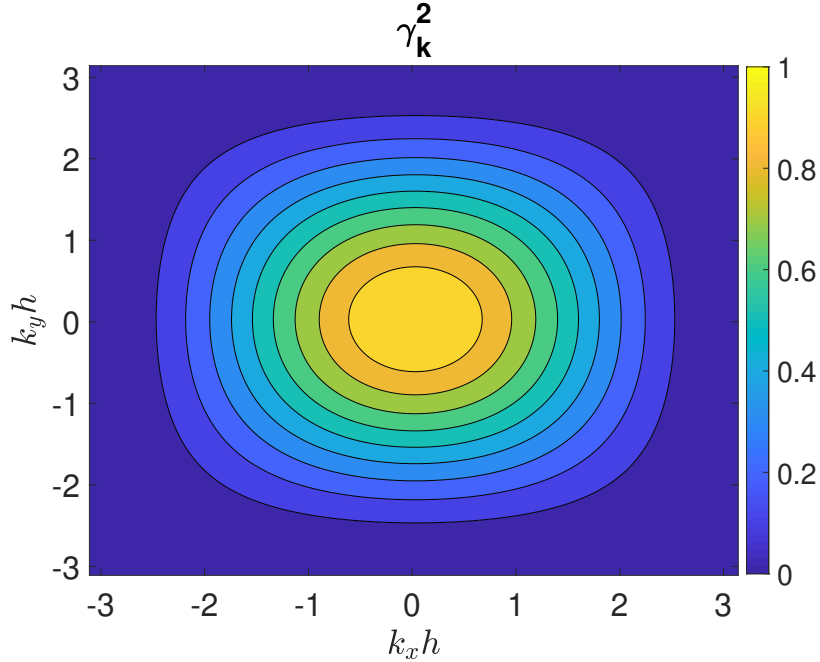


FIGURE 5.1: Plot of γ_k^2 for a square lattice.

We notice that γ_k^2 is very small for k near the Brillouin zone boundary. In that case u_k^2 is close to unity, v_k^2 must be close to zero and $\alpha_k^\dagger \approx \pm a_k^\dagger$. The magnonic density of states is larger near the edge of the Brillouin zone than near the center [18], so the majority of α_k and β_{-k} magnons will be located primarily on sublattices A (spin \uparrow) and B (spin \downarrow), respectively.

The fact that the magnons always come in pairs motivates us to introduce the two-magnon (2M) operators

$$K_k^+ = \alpha_k^\dagger \beta_{-k}^\dagger, \quad K_k^- = \alpha_k \beta_{-k}, \quad K_k^z = \frac{1}{2} (\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1). \quad (5.19)$$

Their commutation relations follow from the Bose commutation relations of α_k and β_{-k} :

$$\begin{aligned} [K_k^-, K_k^+] &= \alpha_k \beta_{-k} \alpha_k^\dagger \beta_{-k}^\dagger - \alpha_k^\dagger \beta_{-k}^\dagger \alpha_k \beta_{-k} \\ &= (1 + \alpha_k^\dagger \alpha_k) (1 + \beta_{-k}^\dagger \beta_{-k}) - (\alpha_k^\dagger \alpha_k) (\beta_{-k}^\dagger \beta_{-k}) \\ &= 1 + \alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} = 2K_k^z, \end{aligned} \quad (5.20)$$

$$\begin{aligned}
[K_k^z, K_k^+] &= \frac{1}{2} \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) \left(\alpha_k^\dagger \beta_{-k}^\dagger \right) - \frac{1}{2} \left(\alpha_k^\dagger \beta_{-k}^\dagger \right) \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) \\
&= \frac{1}{2} \left(\alpha_k^\dagger \alpha_k \alpha_k^\dagger \beta_{-k}^\dagger + \alpha_k^\dagger \beta_{-k}^\dagger \beta_{-k} \beta_{-k}^\dagger - \alpha_k^\dagger \alpha_k^\dagger \alpha_k \beta_{-k}^\dagger - \alpha_k^\dagger \beta_{-k}^\dagger \beta_{-k}^\dagger \beta_{-k} \right) \\
&= \frac{1}{2} \left(\alpha_k^\dagger \left[\alpha_k, \alpha_k^\dagger \right] \beta_{-k}^\dagger + \alpha_k^\dagger \beta_{-k}^\dagger \left[\beta_{-k}, \beta_{-k}^\dagger \right] \right) = K_k^+,
\end{aligned} \tag{5.21}$$

and similarly $[K_k^z, K_k^-] = -K_k^-$.

5.3 Groups, Lie algebra and symmetry

It is interesting to compare the 2M operators to the standard spin operators S_+, S_-, S_z that we started with:

$$\begin{aligned}
[K_k^z, K_k^\pm] &= \pm K_k^\pm, & [K_k^-, K_k^+] &= 2K_k^z \\
[S_z, S_\pm] &= \pm S_\pm, & [S_-, S_+] &= -2S_z.
\end{aligned}$$

To get a sense of the behavior of the operators and how they are related, we will employ the concept of groups. Group theory is branch of mathematics that studies transformations and their symmetry. To keep things from getting out of hand – and because this is strictly speaking a subject of graduate level mathematics – I will keep the discussion short and informal. A comprehensive coverage of the application of group theory to quantum mechanics can be found in [35].

A group G is a set of transformations $\{A, B, \dots\}$ with a composition rule (a rule for how to combine two transformations in the set) satisfying four axioms:

1. Closure: If $A, B \in G$, then $A \bullet B \in G$, where $A \bullet B$ is the composition of A and B .
2. Identity: There exists an element $I \in G$ so that $I \bullet A = A$.
3. Inverse: For any $A \in G$ there exists an inverse A^{-1} so that $A^{-1} \bullet A = I$.
4. Associativity: For $A, B, C \in G$, we have $(A \bullet B) \bullet C = A \bullet (B \bullet C)$.

As a very simple example, consider the set "rotations in a plane". The composition rule will be "addition of angles", so if A rotates by $\frac{\pi}{4}$ and B rotates by $\frac{\pi}{2}$, the composition $A \bullet B$ rotates by $\frac{3\pi}{4}$. The composition is also a rotation in the plane, and therefore the closure axiom is satisfied. The identity is rotation by 0 and the inverse will be rotation in the opposite direction. It is easy to check that the rest of the axioms are satisfied by this group.

Many groups can be represented as invertible matrices. A group of $n \times n$ invertible matrices with complex entries (or any closed subgroup of this group) is called a matrix Lie group. With any Lie group there is an associated Lie algebra that completely characterizes the behavior of the group near the identity element (e.g. small rotations in the example of 2D rotations). To our purposes it suffices to say that the Lie algebra is a vector space of matrices satisfying a specific condition. The Lie algebra is spanned by a set of matrices known as the generators of the group. A generator g_A is a vector of matrices such that the matrix A in the group can be written

$$A = \exp [i g_A \cdot v], \tag{5.22}$$

where v is a vector that parameterizes the transformation. The exponential function in this case is defined by its Maclaurin series expansion. Again returning to our rotation example, $v = v$ would be the rotation angle (a scalar, in this case) and $g_A = g_A$ would be a matrix so that

$$A = \exp [ig_A v] = \begin{bmatrix} \cos(v) & -\sin(v) \\ \sin(v) & \cos(v) \end{bmatrix}, \quad (5.23)$$

because A now rotates the vector $[x_0, y_0]^T$ by an angle v . The Lie algebra of the group "rotations in a plane" will tell us what the generators g_A that satisfy (5.23) are. Conversely, if we have matrices that satisfy the Lie algebra of the group "rotation in the plane", we know what these matrices in fact can be thought of as generating rotations in the plane.

5.3.1 The group SU(2) and its representations

We will take a closer look at a group known as the special unitary group of degree 2, or SU(2). The "special" means that in the matrix representation of SU(2), the matrices have determinant +1. The "unitary" means just that: the matrices are unitary, i. e. $U^\dagger U = I$. The degree 2 means that the matrices are 2×2 .

Any 2×2 unitary matrix with determinant 1 (i. e. any U in SU(2)) can be written

$$U = \begin{bmatrix} z & w \\ -w^* & z^* \end{bmatrix}; \quad |z|^2 + |w|^2 = 1, \quad (5.24)$$

or, with $z = v_4 + iv_3$ and $w = v_2 + iv_1$ for $v_1, v_2, v_3, v_4 \in \mathbb{R}$:

$$U = \begin{bmatrix} v_4 + iv_3 & v_2 + iv_1 \\ -v_2 + iv_1 & v_4 - iv_3 \end{bmatrix} = v_4 \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + v_3 \begin{bmatrix} i & 0 \\ 0 & -i \end{bmatrix} + v_2 \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix} + v_1 \begin{bmatrix} 0 & i \\ i & 0 \end{bmatrix}. \quad (5.25)$$

The matrices in the latter three terms we recognize as i times the Pauli matrices, so that

$$U = v_4 I + iv_3 \sigma_z + iv_2 \sigma_y + iv_1 \sigma_x = v_4 I + iv \cdot \sigma. \quad (5.26)$$

From (5.24) we have the condition

$$v_4^2 + |v|^2 = 1, \quad (5.27)$$

so setting $v_4 = \cos \frac{\theta}{2}$ and $|v| = \sin \frac{\theta}{2}$ makes sure this is satisfied and we get

$$U_n = \left(\cos \frac{\theta}{2} \right) I + i \left(\sin \frac{\theta}{2} \right) \hat{n} \cdot \sigma = \exp \left[i\theta \hat{n} \cdot \frac{\sigma}{2} \right] \quad (5.28)$$

where $\hat{n} = \frac{v}{|v|}$ and I used $(\hat{n} \cdot \sigma)^2 = I$. We have $\theta \in (0, 2\pi]$ but I will not justify it here (see [36]). Apparently the generators of SU(2) are the matrices $\frac{1}{2}\sigma_j$ for $j \in \{x, y, z\}$. Their commutators are

$$\left[\frac{\sigma_x}{2}, \frac{\sigma_y}{2} \right] = i \frac{\sigma_z}{2}, \quad \left[\frac{\sigma_y}{2}, \frac{\sigma_z}{2} \right] = i \frac{\sigma_x}{2}, \quad \left[\frac{\sigma_z}{2}, \frac{\sigma_x}{2} \right] = i \frac{\sigma_y}{2}, \quad (5.29)$$

exactly the commutators for the spin operators S_x, S_y and S_z from (2.25). However, it is not immediately obvious what the significance of this result is. After all:

1. The spin operators S_x, S_y and S_z are 2×2 matrices in the basis $|s m_s\rangle$ if $s = \frac{1}{2}$, but in general there are $2s + 1$ possible values of m_s , and so the spin operators must be $(2s + 1) \times (2s + 1)$ matrices. Are they still generators of SU(2)?

2. We need to know what the transformations in $SU(2)$ do. Why do we care that S_x, S_y and S_z are generators of this group?
3. The operators we are interested in are S_+, S_- and S_z , not S_x, S_y and S_z . What can we say about the raising and lowering operators from what we learn about the spin operators?

It turns out that spin operators that are $(2s + 1) \times (2s + 1)$ are also generators of $SU(2)$ because there exists a linear mapping that maps the generators in 2 dimensions to a set of generators in $2s + 1$ dimensions while preserving the commutation relations in (5.29). In other words, the spin operators $S_x, S_y, S_z \in \mathbb{C}^{(2s+1) \times (2s+1)}$ are *images* of the generators of $SU(2)$ under a linear map from 2 dimensions to $2s + 1$ dimensions. The mapping is called a representation of the Lie algebra of $SU(2)$. The representation with $s = \frac{1}{2}$ is called the defining representation because then every matrix $U \in SU(2)$ is represented by itself.

To get a sense of what the transformations in $SU(2)$ do, we will look at the set of transformations that rotate a vector in 3D around an arbitrary axis by an arbitrary angle. Let's say we want to rotate a vector \mathbf{r} around the direction $\hat{\mathbf{n}}$ by an angle ψ . The transformation $\mathcal{R}(\psi)$ can be written using Rodrigues' rotation formula:

$$\mathcal{R}(\psi)\mathbf{r} = \cos(\psi)\mathbf{r} + [1 - \cos(\psi)](\hat{\mathbf{n}} \cdot \mathbf{r})\hat{\mathbf{n}} + [\sin(\psi)]\hat{\mathbf{n}} \times \mathbf{r}. \quad (5.30)$$

The Maclaurin expansion for small angles $\delta\psi$ to first order is

$$\mathcal{R}(\delta\psi)\mathbf{r} \approx \mathbf{r} + \delta\psi\hat{\mathbf{n}} \times \mathbf{r} = \begin{bmatrix} r_x \\ r_y \\ r_z \end{bmatrix} + \delta\psi \begin{bmatrix} 0 & -n_z & n_y \\ n_z & 0 & -n_x \\ -n_y & n_x & 0 \end{bmatrix} \begin{bmatrix} r_x \\ r_y \\ r_z \end{bmatrix}. \quad (5.31)$$

This means we can write the rotation $\mathcal{R}(\delta\psi)$ as

$$\mathcal{R}(\delta\psi) = I + i\delta\psi \left(n_x \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \end{bmatrix} + n_y \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix} + n_z \begin{bmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \right). \quad (5.32)$$

We will denote the matrices as J_x, J_y and J_z (from left to right above) and $\mathbf{J} = [J_x, J_y, J_z]$. To construct a rotation by an angle ψ , we can make a series of p very small rotations: $\mathcal{R}(\psi) = \left[\mathcal{R}\left(\frac{\psi}{p}\right) \right]^p$. Then setting $p = \frac{1}{\delta\psi}$ yields

$$\mathcal{R}(\psi) = \lim_{\frac{1}{\delta\psi} \rightarrow \infty} [\mathcal{R}(\psi \cdot \delta\psi)]^{\frac{1}{\delta\psi}} = \lim_{\frac{1}{\delta\psi} \rightarrow \infty} [I + \delta\psi \cdot i\psi\hat{\mathbf{n}} \cdot \mathbf{J}]^{\frac{1}{\delta\psi}} = \exp[i\psi\hat{\mathbf{n}} \cdot \mathbf{J}], \quad (5.33)$$

where I used the limit definition of the exponential function $e^x = \lim_{p \rightarrow \infty} \left(1 + \frac{x}{p}\right)^p$ in the last step.

The expression for a rotation in 3D around a direction $\hat{\mathbf{n}}$ by an angle ψ looks very much like a general element from $SU(2)$ in (5.28). The 3D rotation group is known as $SO(3)$ because it consists of orthogonal 3×3 matrices with determinant $+1$, and the group is closely linked to $SU(2)$ [35]. The central idea here is that we can consider the group $SU(2)$ to be rotations as well, although obviously not in real space. In fact, transformations in $SU(2)$ do not act on vectors but on spinors, which have no real space representation at all. Nevertheless, it is sometimes convenient to think of the spin operators as generators of rotations constrained to the unit sphere in spin space.

Before we move on to consider the 2M operators, we note that the operators S_+ , S_- and S_z are a basis for the *complexified* Lie algebra of SU(2). In particular, by allowing the entries in v from (5.22) to be complex we can construct a transformation using this basis. This representation makes it harder to see that SU(2) is a rotation group because it is impossible to write the transformation in a form that looks like (5.28) and (5.33), but it is still a valid representation.

5.3.2 The group SU(1,1) and the 2M operators

The 2M operators are a basis for the (complexified) Lie algebra of the group SU(1,1). A general matrix V in SU(1,1) can be written

$$V = \begin{bmatrix} z & w \\ w^* & z^* \end{bmatrix}; \quad |z|^2 - |w|^2 = 1. \quad (5.34)$$

This way of writing an element of SU(1,1) shows us clearly the connection with the Bogoliubov transformation we made in Chapter 4. For the creation and annihilation operators a_k and b_{-k}^\dagger , we have

$$\begin{bmatrix} \alpha_k \\ \beta_{-k}^\dagger \end{bmatrix} = \begin{bmatrix} u_k & -v_k \\ -v_k & u_k \end{bmatrix} \begin{bmatrix} a_k \\ b_{-k}^\dagger \end{bmatrix}; \quad u_k^2 - v_k^2 = 1, \quad (5.35)$$

or simply

$$\begin{bmatrix} \alpha_k \\ \beta_{-k}^\dagger \end{bmatrix} = V \begin{bmatrix} a_k \\ b_{-k}^\dagger \end{bmatrix}. \quad (5.36)$$

Evidently, the Bogoliubov transformation is an SU(1,1) transformation. No wonder the 2M operators are generators of SU(1,1) – they were constructed from the Bogoliubov-transformed operators.

We saw in our description of SU(2) that we can think of an element of the group SO(3) as a rotation on the unit sphere of Euclidian space. We then looked at the similarity of SO(3) with SU(2) and said that the SU(2) group has in essence the same effect. This "detour" through an orthogonal group can be used to study the action of the group SU(1,1) as well. The group in question is SO(2,1), and it is related to SU(1,1) in the same way that SO(3) is related to SU(2).

The unit sphere S^2 in Euclidian space is

$$S^2 = \{\mathbf{r} = r_x \hat{\mathbf{x}} + r_y \hat{\mathbf{y}} + r_z \hat{\mathbf{z}} \mid r_x^2 + r_y^2 + r_z^2 = 1\}. \quad (5.37)$$

Any element in SO(3) preserves the relation $r_x^2 + r_y^2 + r_z^2 = 1$ when acting on a vector \mathbf{r} . Similarly, it can be shown that any element in SO(2,1) preserves the relation $u_x^2 + u_y^2 - u_z^2 = -1$ when acting on a vector \mathbf{u} . The set of points

$$H^2 = \{\mathbf{r} = u_x \hat{\mathbf{x}} + u_y \hat{\mathbf{y}} + u_z \hat{\mathbf{z}} \mid u_x^2 + u_y^2 - u_z^2 = -1\}. \quad (5.38)$$

form a hyperboloid of two sheets. The upper sheet ($u_z > 0$) of H^2 is sometimes called a pseudosphere because it can be mapped to a Poincaré disk in the (u_x, u_y) -plane in the same way that a sphere is mapped to a plane – a mapping called a stereographic projection. The Poincaré disk is a model of two-dimensional hyperbolic geometry, the description of which is beyond the scope of this discussion. However, this analogy justifies the statement that we can think of H^2 as a hyperbolic unit sphere, and the 2M operators as generators of rotations on this sphere.

The true power in connecting the 2M operators to the SU(1,1) group lies in the fact that it enables us to use the theory that has been developed by mathematicians to study our physical system. In a sense, we acquire a new set of tools that we can apply to structures that might seem very unfamiliar. An excellent example of how we can utilize mathematical results from studies of SU(1,1) in order to obtain a description of our physical system will be given when we look for coherent states at the end of this chapter.

5.4 Néel vector dynamics

The Néel vector \mathbf{l} was first introduced in Chapter 3 in an effort to find macroscopic fields that would enable a classical description of the dynamics of an antiferromagnet. Having defined the Néel vector twice already, I will give yet another definition of the Néel vector, although the similarity with the previous definition in (3.7) should be obvious. In this section I will be referring to the classical Néel vector as the difference between the sublattice magnetizations:

$$\mathbf{L}_{clas} = \mathbf{M}_A - \mathbf{M}_B. \quad (5.39)$$

Here the sublattice magnetizations $\mathbf{M}_A, \mathbf{M}_B$ are

$$\mathbf{M}_A = \gamma \sum_{i \in A} \mathbf{S}_i = \gamma \frac{N}{2} \mathbf{S}_A, \quad (5.40)$$

$$\mathbf{M}_B = \gamma \sum_{j \in B} \mathbf{S}_j = \gamma \frac{N}{2} \mathbf{S}_B, \quad (5.41)$$

where $\mathbf{S}_A, \mathbf{S}_B$ are the average spin per unit volume in sublattices A and B, respectively, and γ is the gyromagnetic ratio. Inserting this expression into the definition of \mathbf{L}_{clas} and setting $\gamma = 1$ yields

$$\mathbf{L}_{clas} = \frac{N}{2} (\mathbf{S}_A - \mathbf{S}_B). \quad (5.42)$$

The use of the volumetric average in the definition of the classical Néel vector is known as the mean field approximation and breaks down when moving to a quantum mechanical description because of quantum correlations. As an example, consider the (normalized) state for two spin- $\frac{1}{2}$ particles $|\phi\rangle = c|\uparrow, \downarrow\rangle + d|\downarrow, \uparrow\rangle$. The total spin is

$$\langle \mathbf{S}_{tot} \rangle = \langle \mathbf{S}_1 + \mathbf{S}_2 \rangle = \langle S_1^x + S_2^x \rangle \hat{x} + \langle S_1^y + S_2^y \rangle \hat{y} + \langle S_1^z + S_2^z \rangle \hat{z}. \quad (5.43)$$

Using $\langle \phi | S_j^x | \phi \rangle = \langle \phi | S_j^y | \phi \rangle = 0$ because $S_j^x = \frac{1}{2} (S_j^+ + S_j^-)$ and $S_j^y = \frac{-i}{2} (S_j^+ - S_j^-)$ with $j \in \{1, 2\}$, we get

$$\langle \mathbf{S}_{tot} \rangle = \langle S_1^z + S_2^z \rangle \hat{z} = \left(\frac{|c|^2}{2} + \frac{-|d|^2}{2} + \frac{-|c|^2}{2} + \frac{|d|^2}{2} \right) \hat{z} = \mathbf{0}. \quad (5.44)$$

The fact that the total spin is zero would force the classical spin vectors \mathbf{S}_A and \mathbf{S}_B to be oppositely aligned with equal magnitude, which again would mean that the length \mathbf{L}_{clas} is fixed. However, if we redefine the Néel vector and total magnetization in terms of quantum mechanical average:

$$\mathbf{L} = \sum_A \langle \mathbf{S}_i \rangle - \sum_B \langle \mathbf{S}_j \rangle, \quad (5.45)$$

$$\mathbf{M} = \sum_A \langle \mathbf{S}_i \rangle + \sum_B \langle \mathbf{S}_j \rangle, \quad (5.46)$$

we see that the length of the Néel vector can vary for two spin- $\frac{1}{2}$ particles even though $\langle \mathbf{M} \rangle = 0$:

$$|\mathbf{L}| = |\langle \mathbf{S}_1 - \mathbf{S}_2 \rangle| = \left| \frac{|c|^2}{2} + \frac{-|d|^2}{2} - \frac{-|c|^2}{2} - \frac{|d|^2}{2} \right| = ||c|^2 - |d|^2|. \quad (5.47)$$

The deviation from the classical result can be explained by looking at spin correlations $\langle \mathbf{S}_1 \mathbf{S}_2 \rangle$:

$$\langle S_1^z S_2^z \rangle = -\frac{|c|^2 + |d|^2}{4} = -\frac{1}{4}, \quad \langle S_1^x S_2^x \rangle = \langle S_1^y S_2^y \rangle = \frac{cd^* + c^*d}{4}. \quad (5.48)$$

Depending on the weights c and d , the system experiences changes in the spin correlations with an accompanying variation in the length of the Néel vector, which could take any value from 0 to 1. Time-dependent weights would therefore cause the length of the Néel vector to oscillate, as illustrated in Fig 5.2 for $|\phi\rangle = \cos t |\uparrow, \downarrow\rangle + \sin t |\downarrow, \uparrow\rangle$.

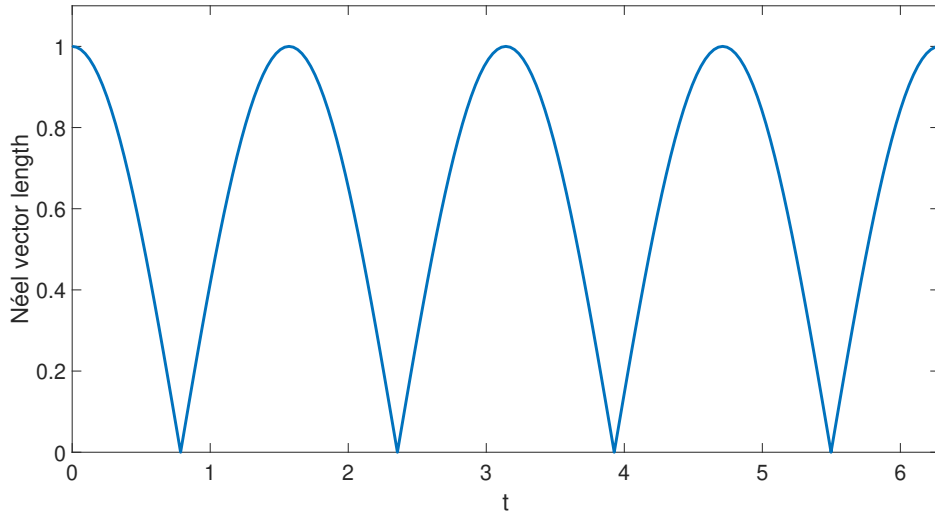


FIGURE 5.2: Oscillations of $|\mathbf{L}|$ in the state $|\phi\rangle = \cos t |\uparrow, \downarrow\rangle + \sin t |\downarrow, \uparrow\rangle$.

For a general bipartite antiferromagnet with arbitrary spin we can employ spin wave theory from Chapter 4 to express the longitudinal component of the Néel vector in terms of spin correlations. We start from (5.45) and invoke (4.9) and (4.13) to get

$$\begin{aligned} L^z &= \sum_A S_i^z - \sum_B S_j^z \\ &= \sum_{i=0}^{\frac{N}{2}} (s - a_i^\dagger a_i) - \sum_{j=0}^{\frac{N}{2}} (-s + b_j^\dagger b_j) \\ &= Ns - \sum_{i=0}^{\frac{N}{2}} a_i^\dagger a_i - \sum_{j=0}^{\frac{N}{2}} b_j^\dagger b_j \\ &= Ns - \sum_k (a_k^\dagger a_k + b_{-k}^\dagger b_{-k}). \end{aligned} \quad (5.49)$$

Here L^z is an operator so that $L \cdot \hat{z} = \langle L^z \rangle$. In the derivation of the ground state Heisenberg Hamiltonian (4.24) we found that

$$\sum_j \sum_\delta S_j^z S_{j+\delta}^z \approx -\frac{z_N s^2 N}{2} + z_N s \sum_k \left(a_k^\dagger a_k + b_{-k}^\dagger b_{-k} \right), \quad (5.50)$$

where we ignored the second-order term in the magnon number operators. The longitudinal component of the Néel vector operator can therefore be rewritten as

$$L^z = \frac{Ns}{2} - \frac{1}{z_N s} \sum_j \sum_\delta S_j^z S_{j+\delta}^z. \quad (5.51)$$

By applying the Bogoliubov transformation to (5.50) we get

$$\begin{aligned} \sum_j \sum_\delta S_j^z S_{j+\delta}^z &= -\frac{z_N s^2 N}{2} \\ &+ z_N s \sum_k \left\{ (u_k^2 + v_k^2) \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) - (u_k^2 - v_k^2) \right\} \\ &+ \sum_k 2u_k v_k \left(\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger \right). \end{aligned} \quad (5.52)$$

We again use (4.32) and (4.36) that link u_k and v_k to γ_k and insert the definition of the 2M operators from (5.19):

$$\begin{aligned} \sum_j \sum_\delta S_j^z S_{j+\delta}^z &= -\frac{z_N s^2 N}{2} \\ &+ z_N s \sum_k \left(\frac{2K_k^z}{\sqrt{1-\gamma_k^2}} - 1 - \frac{\gamma_k}{\sqrt{1-\gamma_k^2}} (K_k^- + K_k^+) \right), \end{aligned} \quad (5.53)$$

and finally we insert the expression for the spin correlations into (5.51), which then reads

$$L^z = Ns - \sum_k \left(\frac{2K_k^z}{\sqrt{1-\gamma_k^2}} - 1 \right) + \sum_k \frac{\gamma_k}{\sqrt{1-\gamma_k^2}} (K_k^- + K_k^+). \quad (5.54)$$

The fact that L^z contains the 2M raising and lowering operators K_k^\pm implies that the longitudinal component of the Néel vector is not conserved because these operators do not commute with the time-independent Hamiltonian. On the other hand, we can repeat (5.49) for the magnetization to get

$$\begin{aligned} M^z &= \sum_k \left(a_k^\dagger a_k - b_{-k}^\dagger b_{-k} \right) \\ &= \sum_k \left(\alpha_k^\dagger \alpha_k - \beta_{-k}^\dagger \beta_{-k} \right). \end{aligned} \quad (5.55)$$

The longitudinal magnetization is therefore constant as long as the magnons always come in pairs, which is precisely what we found for light-induced perturbations to the exchange interaction. Assuming no other excitation mechanism has created an uneven number of α_k and β_k magnons, we get that the longitudinal magnetization is zero. The state of the antiferromagnet, which in general lives in the full Hilbert space

spanned by $|n_{\alpha_k}\rangle|n_{\beta_{-k}}\rangle$, now lives on the subspace spanned by $|n_{\alpha_k}\rangle|n_{\beta_{-k}}\rangle = |n_k\rangle|n_k\rangle$, where it is implied that the β -magnon has opposite wave vector to the α -magnon. The states are eigenstates of K_k^z with eigenvalue $n_k + \frac{1}{2}$ and K_k^\pm create and annihilate the 2M states:

$$\begin{aligned} K_k^+ |n_k\rangle|n_k\rangle &= (n_k + 1) |n_k + 1\rangle|n_k + 1\rangle \\ K_k^- |n_k\rangle|n_k\rangle &= n_k |n_k - 1\rangle|n_k - 1\rangle. \end{aligned} \quad (5.56)$$

5.4.1 Transition to the interaction picture

So far we have been working in the Schrödinger picture with time-independent states. To simplify the analysis, we make a change of basis so that the operators and the state vectors are brought to the interaction picture. We split the Schrödinger Hamiltonian in (5.17) so that $\mathcal{H} = \mathcal{H}_0 + \delta\mathcal{H}$ with

$$\mathcal{H}_0 = -\frac{(\Omega + f(t)\delta\omega_R)N(s+1)}{2} + \sum_k \omega_k \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right), \quad (5.57)$$

$$\delta\mathcal{H} = f(t) \sum_k \left[\delta\omega_k \left(\alpha_k^\dagger \alpha_k + \beta_{-k}^\dagger \beta_{-k} + 1 \right) + V_k \left(\alpha_k \beta_{-k} + \alpha_k^\dagger \beta_{-k}^\dagger \right) \right]. \quad (5.58)$$

For small perturbations we have $\delta\omega_R \ll \Omega$, so \mathcal{H}_0 can be treated as time-independent. A general state in the interaction picture can be written

$$|\Psi_I(t)\rangle = U(t, t_0) |\Psi_I(t_0)\rangle = U(t, t_0) \exp[i\mathcal{H}_0 t] |\psi_S(t_0)\rangle, \quad (5.59)$$

where $U(t, t_0)$ is the unitary evolution operator in the interaction picture and $|\psi_S\rangle$ is a time-independent state in the Schrödinger picture. The raising operator in the the interaction picture becomes

$$\begin{aligned} K_k^+(t) &= \exp[i\mathcal{H}_0 t] K_k^+ \exp[-i\mathcal{H}_0 t] \\ &= \left(1 + i\mathcal{H}_0 t - \frac{1}{2} \mathcal{H}_0^2 t^2 + \dots \right) K_k^+ \exp[-i\mathcal{H}_0 t] \\ &= K_k^+ \left(1 + i[\mathcal{H}_0 + 2\omega_k] t - \frac{1}{2} [\mathcal{H}_0 + 2\omega_k]^2 t^2 + \dots \right) \exp[-i\mathcal{H}_0 t] \\ &= K_k^+ \exp[i(\mathcal{H}_0 + 2\omega_k) t] \exp[-i\mathcal{H}_0 t] = K_k^+ \exp[2i\omega_k t], \end{aligned} \quad (5.60)$$

where I used the commutation relation $[\mathcal{H}_0, K_k^\pm] = 2\omega_k K_k^\pm$. Similarly we obtain

$$K_k^-(t) = K_k^- \exp[-2i\omega_k t], \quad K_k^z(t) = K_k^z. \quad (5.61)$$

With the 2M operators in the interaction picture we obtain the interaction Hamiltonian

$$\mathcal{H}_0^I = -\frac{(\Omega + f(t)\delta\omega_R)N(s+1)}{2} + \sum_k 2\omega_k K_k^z(t) = \mathcal{H}_0, \quad (5.62)$$

$$\delta\mathcal{H}^I = f(t) \sum_k \left[V_k (K_k^-(t) + K_k^+(t)) + 2\delta\omega_k K_k^z(t) \right]. \quad (5.63)$$

We are now interested in finding an explicit expression for the unitary evolution operator $U(t, t_0)$ so that we can calculate the time-dependence of the expectation values of the 2M operators. Plugging $|\Psi_I\rangle = U(t, t_0) |\Psi(t_0)\rangle$ into the transformed

Schrödinger equation (2.45) yields the equation for the time evolution of $U(t, t_0)$:

$$i\partial_t U(t, t_0) = \delta \mathcal{H}^I U(t, t_0). \quad (5.64)$$

The differential equation is separable with solution

$$U(t, t_0) = \exp \left[-i \int_{t_0}^t \delta \mathcal{H}^I(t') dt' \right]. \quad (5.65)$$

We set $t_0 < 0$ as the starting point of the the evolution of the state. For exciting laser pulses τ much shorter than the oscillation period of the magnons we can model the exciting laser pulse as $f(t) = \tau\delta(t)$, where $\delta(t)$ is the Dirac delta function. Inserting this into (5.65) we get

$$U(t, 0) = \prod_k \exp \left[-i\tau \{ V_k K_k^+(0) + V_k K_k^-(0) + 2\delta\omega_k K_k^z(0) \} \right]. \quad (5.66)$$

The expectation value of 2M operator can therefore be written

$$\begin{aligned} \langle K_k^{\pm,z}(t) \rangle &= \langle \Psi_I(0) | U_k^\dagger K_k^{\pm,z} U_k | \Psi_I(0) \rangle \\ &= \langle \exp [i\tau \mathcal{H}_k] K_k^{\pm,z} \exp [-i\tau \mathcal{H}_k] \rangle, \end{aligned} \quad (5.67)$$

where $U = \prod_k U_k = \prod_k \exp [-i\tau\delta \mathcal{H}_k]$. To find explicit expressions for $\langle K_k^{\pm,z}(t) \rangle$ we use the identity

$$e^A B e^{-A} = B + [A, B] + \frac{1}{2!} [A, [A, B]] + \frac{1}{3!} [A, [A, [A, B]]] + \dots \quad (5.68)$$

Keeping only the first three terms leaves us with

$$\begin{aligned} \langle K_k^+(t) \rangle &= \langle K_k^+ \rangle (1 + i\tau 2\delta\omega_k - (i\tau)^2 V_k^2 + 2(i\tau)^2 \delta\omega_k^2) \exp [2i\omega_k t] \\ &\quad + \langle K_k^- \rangle (i\tau)^2 V_k^2 \exp [2i\omega_k t] \\ &\quad + \langle K_k^z \rangle 2i\tau V_k (1 + i\tau\delta\omega_k) \exp [2i\omega_k t]. \end{aligned} \quad (5.69)$$

But $\langle K_k^+ \rangle = \langle K_k^- \rangle = 0$, so

$$\langle K_k^+(t) \rangle = \langle K_k^z \rangle 2i\tau V_k (1 + i\tau\delta\omega_k) \exp [2i\omega_k t]. \quad (5.70)$$

To first order in τ we have $1 + i\tau\delta\omega_k = \exp [i\tau\delta\omega_k]$. Making this substitution yields

$$\langle K_k^+(t) \rangle = \langle K_k^z \rangle 2i\tau V_k \exp [i(2\omega_k t + \tau\delta\omega_k)]. \quad (5.71)$$

Similar calculations yield the following expressions for the two other 2M operators:

$$\langle K_k^-(t) \rangle = -\langle K_k^z \rangle 2i\tau V_k \exp [-i(2\omega_k t + \tau\delta\omega_k)], \quad (5.72)$$

$$\langle K_k^z(t) \rangle = \langle K_k^z \rangle (1 - 2(i\tau)^2 V_k^2) \approx \langle K_k^z \rangle. \quad (5.73)$$

We can now express the time-dependent longitudinal component of the Néel vector from (5.54) as

$$\begin{aligned} \langle L^z(t) \rangle &= Ns - \sum_k \left(\frac{2\langle K_k^z(t) \rangle}{\sqrt{1-\gamma_k^2}} - 1 \right) + \sum_k \frac{\gamma_k}{\sqrt{1-\gamma_k^2}} (\langle K_k^-(t) \rangle + \langle K_k^+(t) \rangle) \\ &= Ns - \Delta L + \delta L(t), \end{aligned} \quad (5.74)$$

where I collected terms so that

$$\Delta L = \sum_k \left(\frac{2\langle K_k^z \rangle}{\sqrt{1-\gamma_k^2}} - 1 \right). \quad (5.75)$$

and

$$\delta L(t) = 4\tau \sum_k \frac{\gamma_k V_k}{\sqrt{1-\gamma_k^2}} \langle K_k^z \rangle \sin(2\omega_k t + \tau\delta\omega_k). \quad (5.76)$$

From (5.75) it should be clear that magnons in the antiferromagnet reduce the length of the longitudinal component of the Néel vector, and hence reduce the length of the Néel vector itself. The fact that there are magnons present even in the ground state implies that the length of the quantum Néel vector is always smaller than that of its classical counterpart. We also see from (5.76) that the light pulse induces oscillations in the length of the Néel vector with a frequency twice as large as the magnon frequency ω_k . Thinking back to our example with just two spin- $\frac{1}{2}$ particles, we were able to explain oscillations in the Néel vector as a transition between the two states $|\uparrow, \downarrow\rangle$ and $|\downarrow, \uparrow\rangle$ in the superposition state $|\phi\rangle = c(t)|\uparrow, \downarrow\rangle + d(t)|\downarrow, \uparrow\rangle$. It is the same story for a general antiferromagnet: on top of the reduction in $|L|$ caused by ground state magnons, oscillations are triggered by varying contributions from the 2M states to the total state of the system $|\Psi(t)\rangle$. The amplitude of the oscillations is proportional to the energy of the excited magnons, which in turn is proportional to the change in the exchange interaction:

$$\delta L(t)_{\max} \propto V_k \propto \delta\omega_R \propto \Delta J. \quad (5.77)$$

The period of the oscillations is inversely proportional to the strength of the interaction:

$$T_k = \frac{\pi}{\omega_k} \sim \frac{1}{J}. \quad (5.78)$$

The oscillations in the Néel vector are illustrated in Fig. 5.3. We conclude this section by quickly noting that in the limit of large s and large N the first term (5.74) dominates the two latter terms, and so we recover the classical result for the Néel vector, as required.

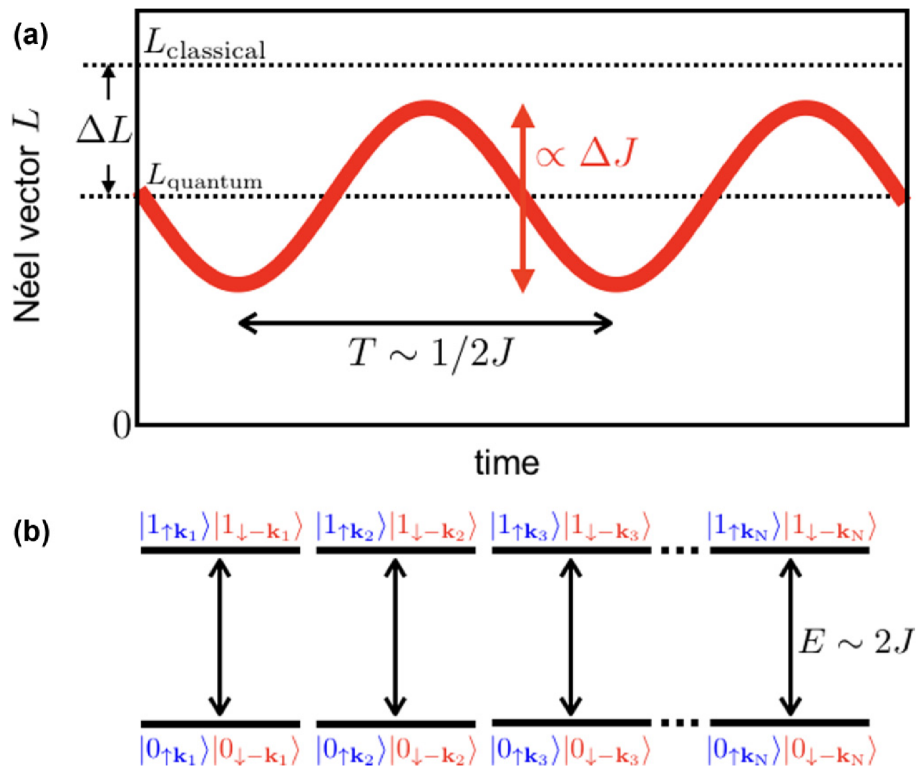


FIGURE 5.3: Illustration of the dynamics of the Néel vector length L . (a) The quantum Néel vector length is reduced compared to the classical Néel vector and oscillates as a result of light-induced excitations. (b) The oscillations in L can be viewed as stemming from varying contributions from the 2M states to the total state of the antiferromagnet. In this case transitions from the Néel state $|0_k\rangle|0_k\rangle$ to the state where one 2M magnon is present are shown. The coloring of the states corresponds to the sublattice in which the magnons are predominantly localized (see Fig. 4.2). Illustration originally from [18].

5.5 Spin correlations

In the previous section we found the expression (5.53) for the longitudinal spin correlator along the z -axis. While useful for calculating Néel vector dynamics, this expression tells us nothing about the correlations between two individual spins in the magnet. Another problem with the correlator is the approximation that we made in (5.50) where we ignored the term containing four Bose operators. Without making this approximation, the spin correlator in (5.53) picks up an extra term

$$\sum_j \sum_\delta a_j^\dagger a_j b_{j+\delta}^\dagger b_{j+\delta} = z_N \sum_{k_1} \sum_{k_2} \sum_{k_3} \sum_{k_4} \sum_j a_{k_1}^\dagger a_{k_2} b_{-k_3}^\dagger b_{-k_4} \exp [i(k_1 + k_4 - k_3 - k_2) \cdot r_j], \quad (5.79)$$

where the Fourier series expansions (4.20)-(4.23) were used. As before we can exploit destructive wave interference so that

$$\sum_j \exp [i(k_1 + k_4 - k_3 - k_2) \cdot r_j] = \frac{N}{2} \delta_{k_4, k_2 + k_3 - k_1}, \quad (5.80)$$

but we are still left with three wave vectors k_1, k_2 and k_3 that will yield six Bogoliubov magnon operators ($\alpha_k, \beta_{-k}, k = k_1, k_2, k_3$) after the transformation. The upshot is that

the right hand side of (5.79) will be a sum over every combination of three reciprocal lattice vectors of 16 terms with four magnon operators in each term. When we transition to the interaction picture we will have to find the commutator of all these terms with the unperturbed Hamiltonian, which in itself is a massive problem. Even worse, the time-evolution operator (5.66) contains three times as many magnon operators as the unperturbed Hamiltonian, tripling the number of commutators that have to be calculated. The procedure is straightforward but incredibly time-consuming.

The reason I bring this up is that this growing complexity as a result of more and more wave vectors entering the problem is a recurring theme in my project. Unfortunately, the number of terms in (5.79) is not even the worst horror story in this regard. As an example, consider the correlator

$$\langle S_j^z S_i^z \rangle = -s^2 + s \left(\langle a_j^\dagger a_j \rangle + \langle b_i^\dagger b_i \rangle \right) - \langle a_j^\dagger a_j b_i^\dagger b_i \rangle, \quad (5.81)$$

where r_j is a site on sublattice A and r_i is a site on sublattice B. The last term is similar to the term in (5.79) but lacking the sum over all lattice sites, meaning we can not exploit the destructive interference expressed by (5.80) to remove the fourth sum over the Brillouin zone. Hence the operator $a_j^\dagger a_j b_i^\dagger b_i$ in Fourier space will be a sum over every combination of four reciprocal lattice vectors. It is definitely doable to keep working in Fourier space and with the Bogoliubov transformation, but at some point the number of terms grows so large that this approach is no longer practical. For this reason I have spent a considerable amount of time looking for techniques that let us reduce the number of terms relative to the Bogoliubov transformation. An interesting candidate is the generalization of the Bogoliubov transformation, which is described in section 6.2. However, this generalization is not useful for dealing with the last term of (5.81) either. I am not aware of any method where the calculation of $\langle a_j^\dagger a_j b_i^\dagger b_i \rangle$ is not a tremendously tedious endeavor.

The expectation value of the operator $a_j^\dagger a_j$ is possible to calculate using the Bogoliubov transformation without too much difficulty, so we will go through the steps here. The expectation value of the number operator on any other site is calculated similarly, of course. In order to keep track of the different wave vectors more easily I will label wave vectors with λ, μ and ν , rather than k with a subscript. We will need the following commutators for the 2M number operator K_λ^z :

$$\left[2K_\lambda^z, \alpha_\nu^\dagger \alpha_\mu \right] = \alpha_\lambda^\dagger \alpha_\mu \delta_{\lambda, \nu} - \alpha_\nu^\dagger \alpha_\lambda \delta_{\lambda, \mu}. \quad (5.82)$$

$$\left[2K_\lambda^z, \beta_{-\nu}^\dagger \beta_{-\mu} \right] = \beta_{-\lambda}^\dagger \beta_{-\mu} \delta_{\lambda, \nu} - \beta_{-\nu}^\dagger \beta_{-\lambda} \delta_{\lambda, \mu}. \quad (5.83)$$

$$\left[2K_\lambda^z, \alpha_\nu^\dagger \beta_{-\mu}^\dagger \right] = \alpha_\lambda^\dagger \beta_{-\mu}^\dagger \delta_{\lambda, \nu} + \alpha_\nu^\dagger \beta_{-\lambda}^\dagger \delta_{\lambda, \mu}. \quad (5.84)$$

$$\left[2K_\lambda^z, \alpha_\nu \beta_{-\mu} \right] = -\alpha_\lambda \beta_{-\mu} \delta_{\lambda, \nu} - \alpha_\nu \beta_{-\lambda} \delta_{\lambda, \mu}. \quad (5.85)$$

For K_λ^+ we have:

$$\left[K_\lambda^+, \alpha_\nu^\dagger \alpha_\mu \right] = -\alpha_\nu^\dagger \beta_{-\lambda}^\dagger \delta_{\lambda, \mu}. \quad (5.86)$$

$$\left[K_\lambda^+, \beta_{-\nu}^\dagger \beta_{-\mu} \right] = -\alpha_\lambda^\dagger \beta_{-\nu}^\dagger \delta_{\lambda, \mu}. \quad (5.87)$$

$$\left[K_\lambda^+, \alpha_\nu^\dagger \beta_{-\mu}^\dagger \right] = 0. \quad (5.88)$$

$$\left[K_\lambda^+, \alpha_\nu \beta_{-\mu} \right] = -\beta_{-\lambda}^\dagger \beta_{-\mu} \delta_{\lambda, \nu} - \alpha_\lambda^\dagger \alpha_\nu \delta_{\lambda, \mu} - \delta_{\lambda, \nu} \delta_{\lambda, \mu}. \quad (5.89)$$

For K_λ^- we have:

$$\left[K_\lambda^-, \alpha_v^\dagger \alpha_\mu \right] = \alpha_\mu \beta_{-\lambda} \delta_{v,\lambda}. \quad (5.90)$$

$$\left[K_\lambda^-, \beta_{-v}^\dagger \beta_{-\mu} \right] = \alpha_\lambda \beta_{-\mu} \delta_{v,\lambda}. \quad (5.91)$$

$$\left[K_\lambda^-, \alpha_v^\dagger \beta_{-\mu}^\dagger \right] = \alpha_v^\dagger \alpha_\lambda \delta_{\mu,\lambda} + \beta_{-\mu}^\dagger \beta_{-\lambda} \delta_{v,\lambda} + \delta_{\lambda,v} \delta_{\lambda,\mu}. \quad (5.92)$$

$$\left[K_\lambda^-, \alpha_v \beta_{-\mu} \right] = 0. \quad (5.93)$$

The first step is to express the operator $a_j^\dagger a_j$ in terms of the Bogolubov magnons. Until now we have first made the Fourier transform and then used (4.29), but because (5.80) cannot be used we should combine (4.21) and (4.29) so that the Bogoliubov coefficients for site r_j are

$$u_{j,\lambda} = \sqrt{\frac{2}{N}} u_\lambda \exp[-i\lambda \cdot r_j], \quad v_{j,\lambda} = \sqrt{\frac{2}{N}} v_\lambda \exp[i\lambda \cdot r_j], \quad (5.94)$$

and

$$a_j = \sum_\lambda \left(u_{j,\lambda} \alpha_\lambda + v_{j,\lambda}^* \beta_{-\lambda} \right) \quad (5.95)$$

is the Bogoliubov transformation. Thus, the number operator $a_j^\dagger a_j$ in the Schrödinger picture is

$$a_j^\dagger a_j = \sum_v \sum_\mu u_{j,v}^* u_{j,\mu} \alpha_v^\dagger \alpha_\mu + u_{j,v}^* v_{j,\mu} \alpha_v^\dagger \beta_{-\mu}^\dagger + u_{j,\mu} v_{j,v} \alpha_\mu \beta_{-v} + v_v v_\mu^* \left(\beta_{-\mu}^\dagger \beta_{-v} + \delta_{v,\mu} \right). \quad (5.96)$$

The next step is to bring the operator over to the interaction picture. With our experience calculating the 2M interaction picture operators in the previous section, we immediately see that

$$a_j^\dagger a_j(t) = \exp \left[it \sum_\lambda 2\omega_\lambda K_\lambda^z \right] a_j^\dagger a_j \exp \left[-it \sum_\lambda 2\omega_\lambda K_\lambda^z \right]. \quad (5.97)$$

Making use of (5.68) and the commutators (5.82)-(5.85), we find

$$\begin{aligned} a_j^\dagger a_j(t) &= \sum_v \sum_\mu \left(e^{i(\omega_v - \omega_\mu)t} u_{j,v}^* u_{j,\mu} \alpha_v^\dagger \alpha_\mu + e^{i(\omega_v + \omega_\mu)t} u_{j,v}^* v_{j,\mu}^* \alpha_v^\dagger \beta_{-\mu}^\dagger \right) \\ &\quad + \sum_v \sum_\mu \left(e^{-i(\omega_v + \omega_\mu)t} u_{j,\mu} v_{j,v} \alpha_\mu \beta_{-v} + e^{-i(\omega_v - \omega_\mu)t} v_{j,v} v_{j,\mu}^* \left\{ \beta_{-\mu}^\dagger \beta_{-v} + \delta_{v,\mu} \right\} \right). \end{aligned} \quad (5.98)$$

To arrive at this expression I did not truncate the sum on the right hand side of (5.68), so it is exact. However, for the subsequent calculations we have to make the same approximations as before: we model the light pulse with the Dirac delta function so that $f(t) = \tau \delta(t)$ and keep terms only to second order in τ . That is,

$$\begin{aligned} U^\dagger a_j^\dagger a_j(t) U &\approx a_j^\dagger a_j(t) \\ &\quad + \left[i\tau \delta \mathcal{H}^I(t=0, f(t)=1), a_j^\dagger a_j(t) \right] \\ &\quad + \frac{1}{2} \left[i\tau \delta \mathcal{H}^I(t=0, f(t)=1), \left[i\tau \delta \mathcal{H}^I(t=0, f(t)=1), a_j^\dagger a_j(t) \right] \right]. \end{aligned} \quad (5.99)$$

Using (5.82)-(5.93) multiple times leaves us with

$$\begin{aligned}
\langle U^\dagger a_j^\dagger a_j(t) U \rangle &\approx \sum_\lambda \left\{ u_{j,\lambda}^* u_{j,\lambda} \langle \alpha_\lambda^\dagger \alpha_\lambda \rangle + v_{j,\lambda}^* v_{j,\lambda} \left(\langle \beta_{-\lambda}^\dagger \beta_{-\lambda} \rangle + 1 \right) \right\} \\
&\quad + i\tau \sum_\lambda 2V_\lambda \left(u_{j,\lambda}^* v_{j,\lambda}^* e^{2i\omega_\lambda t} - u_{j,\lambda} v_{j,\lambda} e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle \\
&\quad - 2(i\tau)^2 \sum_\lambda V_\lambda^2 \left(u_{j,\lambda}^* u_{j,\lambda} + v_{j,\lambda}^* v_{j,\lambda} \right) \langle K_\lambda^z \rangle \\
&\quad + 2(i\tau)^2 \sum_\lambda 2V_\lambda \delta\omega_\lambda \left(u_{j,\lambda}^* v_{j,\lambda}^* e^{2i\omega_\lambda t} + u_{j,\lambda} v_{j,\lambda} e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle.
\end{aligned} \tag{5.100}$$

Looking back at (5.94), we see that

$$u_{j,\lambda}^* u_{j,\lambda} = \frac{2}{N} u_\lambda^2, \quad v_{j,\lambda}^* v_{j,\lambda} = \frac{2}{N} v_\lambda^2, \quad u_{j,\lambda}^* v_{j,\lambda}^* = u_{j,\lambda} v_{j,\lambda} = \frac{2}{N} u_\lambda v_\lambda, \tag{5.101}$$

and so the expectation value $\langle a_j^\dagger a_j \rangle$ is the same for every site on sublattice A after the excitation. Thus,

$$\begin{aligned}
\frac{N}{2} \langle U^\dagger a_j^\dagger a_j(t) U \rangle &\approx \sum_\lambda \left\{ u_\lambda^2 \langle \alpha_\lambda^\dagger \alpha_\lambda \rangle + v_\lambda^2 \left(\langle \beta_{-\lambda}^\dagger \beta_{-\lambda} \rangle + 1 \right) \right\} \\
&\quad + i\tau \sum_\lambda 2V_\lambda u_\lambda v_\lambda \left(e^{2i\omega_\lambda t} - e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle \\
&\quad - 2(i\tau)^2 \sum_\lambda V_\lambda^2 \left(u_\lambda^2 + v_\lambda^2 \right) \langle K_\lambda^z \rangle \\
&\quad + 2(i\tau)^2 \sum_\lambda 2V_\lambda \delta\omega_\lambda u_\lambda v_\lambda \left(e^{2i\omega_\lambda t} + e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle.
\end{aligned} \tag{5.102}$$

Repeating the steps for the operator $b_i^\dagger b_i$ for a site r_i on sublattice B yields

$$\begin{aligned}
\frac{N}{2} \langle U^\dagger b_i^\dagger b_i(t) U \rangle &\approx \sum_\lambda \left\{ u_\lambda^2 \langle \beta_{-\lambda}^\dagger \beta_{-\lambda} \rangle + v_\lambda^2 \left(\langle \alpha_\lambda^\dagger \alpha_\lambda \rangle + 1 \right) \right\} \\
&\quad + i\tau \sum_\lambda 2V_\lambda u_\lambda v_\lambda \left(e^{2i\omega_\lambda t} - e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle \\
&\quad - 2(i\tau)^2 \sum_\lambda V_\lambda^2 \left(u_\lambda^2 + v_\lambda^2 \right) \langle K_\lambda^z \rangle \\
&\quad + 2(i\tau)^2 \sum_\lambda 2V_\lambda \delta\omega_\lambda u_\lambda v_\lambda \left(e^{2i\omega_\lambda t} + e^{-2i\omega_\lambda t} \right) \langle K_\lambda^z \rangle.
\end{aligned} \tag{5.103}$$

Using that the expectation values for the number operators in spin sites are all equal, we get

$$\langle S_j^z S_i^z \rangle = \frac{2}{Nz_N} \sum_j \sum_\delta \langle S_j^z S_{j+\delta}^z \rangle = 1 - \frac{2s}{N} \langle L^z(t) \rangle, \tag{5.104}$$

where I used (5.51) and $\langle L^z(t) \rangle$ is given by (5.74). The fact that the spin correlator in (5.104) is independent of the spin sites is unsurprising because each spin is treated equivalently by the perturbation, and the Bogoliubov transformation does not account for spins on the edges of the magnet (i.e. we do not consider edge effects).

5.6 Coherent states and macroscopic dynamics

In the final section of this chapter we will be looking for ways to incorporate the results from our quantum treatment of antiferromagnets into a semi-classical theory. We shall start by looking at a system where the connection between the classical and quantum states is easier to draw: the simple harmonic oscillator.

5.6.1 Harmonic oscillator coherent states

Classically the position of a particle in a harmonic oscillator potential is given by Newton's second law

$$\frac{d^2q}{dt^2} + \omega^2q = 0, \quad (5.105)$$

where $\omega = \sqrt{\frac{k}{m}}$, m is the particle's mass and $U = \frac{1}{2}kq^2$ is the potential for some constant k and displacement q from the center. The solution is simple harmonic motion of the form

$$q(t) = A \cos(\omega t + \phi), \quad (5.106)$$

where the amplitude A and phase ϕ is determined by boundary conditions. In quantum mechanics the particle's position is determined by its wavefunction $\Psi(q, t) = \psi(q) \exp[i\omega t]$, which satisfies the Schrödinger equation

$$\left(-\frac{\hbar^2}{2m} \frac{\partial^2}{\partial q^2} + \frac{1}{2}kq^2 \right) \Psi = i\hbar \frac{\partial}{\partial t} \Psi. \quad (5.107)$$

Once the wave function is obtained from (5.107), the probability density of finding the particle in position q at time t is $|\Psi(q, t)|^2$. That is, according to the statistical interpretation of quantum mechanics, the probability of finding the particle in some interval Δq is $\int_{\Delta q} |\Psi(q, t)|^2 dq$. Of course, we can never hope to find a wave function that yields a deterministic value for the position and momentum simultaneously because the operators for position and momentum do not commute, but we can certainly try to find states where the uncertainties are at a minimum (i.e. (2.48) is satisfied with equality). We also might hope to find states where the width of $|\psi(q)|^2$ is constant in time:

$$|\Psi(q, t)|^2 = |\Psi(q - f(t), 0)|^2, \quad (5.108)$$

and where the center of the distribution follows classical simple harmonic motion:

$$\frac{d^2f}{dt^2} + \omega^2f = 0 \quad (5.109)$$

These states would be the quantum states that come closest to reproducing the classical motion of the particle.

The remaining discussion of harmonic oscillator coherent states is inspired by the discussion presented in [37]. We will start with finding the $\psi(q)$ that satisfies (5.107) with minimal uncertainty. The possible solutions can be looked up in [23]. After normalization they are

$$\psi(q)_n = \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{4}} \frac{1}{\sqrt{2^n n!}} H_n \left(\sqrt{\frac{m\omega}{\hbar}} q \right) \exp \left[-\frac{m\omega}{2\hbar} q^2 \right] \quad (5.110)$$

or any linear combination of them, where H_n is the Hermite polynomial of degree n and n is a non-negative integer. Next we need to find a state that hits the uncertainty limit

$$\Delta_q \Delta_p = \frac{1}{2i} | [q, p] | = \frac{\hbar}{2}, \quad (5.111)$$

where p is the momentum of the particle and $\Delta_x = \sqrt{\langle x^2 \rangle - \langle x \rangle^2}$ is the uncertainty in $x \in \{q, p\}$. This is the case for the ground state:

$$\psi_0(q) = \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{4}} \exp \left[-\frac{m\omega}{2\hbar} q^2 \right], \quad (5.112)$$

because by symmetry we have

$$\langle q \rangle = \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{2}} \int_{-\infty}^{\infty} q \exp \left[-\frac{m\omega}{\hbar} q^2 \right] dq = 0 \quad (5.113)$$

$$\langle p \rangle = -i\hbar \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{2}} \int_{-\infty}^{\infty} \exp \left[-\frac{m\omega}{2\hbar} q^2 \right] \frac{d}{dq} \exp \left[-\frac{m\omega}{2\hbar} q^2 \right] dq = 0, \quad (5.114)$$

while the two non-vanishing integrals are

$$\langle q^2 \rangle = \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{2}} \int_{-\infty}^{\infty} q^2 \exp \left[-\frac{m\omega}{\hbar} q^2 \right] dq = \frac{\hbar}{2m\omega} \quad (5.115)$$

and

$$\langle p^2 \rangle = -\hbar^2 \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{2}} \int_{-\infty}^{\infty} \exp \left[-\frac{m\omega}{\hbar} q^2 \right] \frac{d^2}{dq^2} \exp \left[-\frac{m\omega}{\hbar} q^2 \right] dq = \frac{m\omega\hbar}{2}. \quad (5.116)$$

So we find that the uncertainty product in the ground state is

$$\Delta_q \Delta_p = \sqrt{\langle q^2 \rangle - \langle q \rangle^2} \sqrt{\langle p^2 \rangle - \langle p \rangle^2} = \frac{\hbar}{2}, \quad (5.117)$$

exactly what we wanted.

Equipped with a minimal uncertainty state, we are now able to write up the general form of the coherent wave function. Using the dimensionless variable

$$\varphi = \sqrt{\frac{m\omega}{\hbar}} q, \quad (5.118)$$

we have

$$\Psi \propto \exp \left[-\frac{1}{2} (\varphi - f(t))^2 \right] \exp [i\Phi(\varphi, f; t)], \quad (5.119)$$

where Φ is some real functional. The wave function must satisfy the Schrödinger equation (5.107), which in terms of the dimensionless variable φ reads

$$i\partial_t \Psi = \frac{\omega}{2} \left(-\frac{\partial^2}{\partial \varphi^2} + \varphi^2 \right) \Psi. \quad (5.120)$$

Inserting (5.119) into (5.120) yields

$$2i\dot{f}(\varphi - f) - 2\dot{\Phi} = \omega [2f\varphi + 2i\Phi'(\varphi - f) + \Phi'^2 - f^2 - i\Phi'' + 1], \quad (5.121)$$

where

$$\dot{f} = \frac{df}{dt}, \quad \dot{\Phi} = \frac{\partial\Phi}{\partial t}, \quad \Phi' = \frac{\partial\Phi}{\partial\varphi}.$$

We are free to choose the phase Φ as we please, so setting

$$\Phi(\varphi, f; t) = \frac{\varphi\dot{f}(t)}{\omega} - \chi(t) - \frac{i\omega t}{2} \quad (5.122)$$

makes (5.121) reduce to

$$\dot{f}^2 + 2\varphi(\ddot{f} + \omega^2 f) - f^2\omega^2 = 2\dot{\chi}\omega. \quad (5.123)$$

All that remains is to find χ so that any f that satisfies (5.109) also satisfies (5.123). We can get rid of \dot{f} if we set

$$\chi(t) = \frac{1}{2} \left(\frac{\dot{f}(t)f(t)}{\omega} + C \right) \implies \dot{\chi} = \frac{1}{2} \left(\frac{\dot{f}(t)^2}{\omega} + \frac{\ddot{f}(t)f(t)}{\omega} \right) \quad (5.124)$$

with C some irrelevant constant phase factor. This yields

$$2\varphi(\ddot{f} + \omega^2 f) - \omega^2 f^2 = \ddot{f}f, \quad (5.125)$$

which we see is indeed satisfied by any function that satisfies (5.109). The coherent state wave function for an harmonic oscillator is therefore

$$\begin{aligned} \Psi(q, t) = & \left(\frac{m\omega}{\pi\hbar} \right)^{\frac{1}{4}} \exp \left[-\frac{1}{2} \left(\sqrt{\frac{m\omega}{\hbar}} q - f(t) \right)^2 \right] \\ & \times \exp \left[i \frac{\sqrt{\frac{m\omega}{\hbar}} q \dot{f}(t)}{\omega} - i \frac{\dot{f}(t)f(t)}{2\omega} - i \frac{\omega t}{2} \right], \end{aligned} \quad (5.126)$$

where I excluded an arbitrary constant phase factor that is determined by the initial state.

Our analysis so far has revealed three distinct characteristics of coherent states for the harmonic oscillator:

1. They are minimum uncertainty states.
2. They evolve without spreading.
3. The peak of the probability density moves harmonically.

All of these are traits we associate with the classical harmonic oscillator. Before we can start to look for coherent magnon states, however, we need to introduce a different formalism that allows us to work with coherent states without resorting to wave mechanics. We start by writing up the usual ladder operators for the harmonic oscillator:

$$a^\dagger = \frac{1}{\sqrt{2\hbar m\omega}} (-ip + m\omega q), \quad a = \frac{1}{\sqrt{2\hbar m\omega}} (ip + m\omega q). \quad (5.127)$$

We can express the coordinate and momentum operators in terms of the raising and lowering operators:

$$q = \sqrt{\frac{\hbar}{2m\omega}} (a^\dagger + a), \quad p = i\sqrt{\frac{\hbar}{2m\omega}} (a^\dagger - a), \quad (5.128)$$

so that the harmonic oscillator Hamiltonian becomes

$$\mathcal{H}_{har} = \hbar\omega \left(a^\dagger a + \frac{1}{2} \right) \quad (5.129)$$

whose n -th normalized eigenstate is

$$|n\rangle = \frac{1}{\sqrt{n!}} (a^\dagger)^n |0\rangle. \quad (5.130)$$

We know from the wave mechanics analysis that the coherent states are formed from the ground state. Evidently, there must be some unitary operator $D(z)$ that produces the coherent state $|z\rangle$ from $|0\rangle$. The operator must be unitary because we require

$$\langle z|z\rangle = \langle 0|D^\dagger D|0\rangle = \langle 0|0\rangle = 1. \quad (5.131)$$

We can expand $|z\rangle$ in the energy eigenstates:

$$|z\rangle = \sum_{n=0}^{\infty} c_n |n\rangle = \sum_{n=0}^{\infty} \frac{c_n}{\sqrt{n!}} (a^\dagger)^n |0\rangle. \quad (5.132)$$

Interestingly, if we choose $c_n(z) = \frac{c_0(z)z^n}{\sqrt{n!}}$ then

$$|z\rangle = c_0(z) \sum_{n=0}^{\infty} \frac{(za^\dagger)^n}{n!} |0\rangle = c_0(z) e^{za^\dagger} |0\rangle. \quad (5.133)$$

Clearly, $D(z)|0\rangle = c_0(z)e^{za^\dagger}|0\rangle$ if the choice of c_n is valid. We must choose $c_0(z)$ so that

$$\Psi(q, 0) = \langle q|z\rangle = c_0 \sum_{n=0}^{\infty} \frac{z^n}{n!} \langle q| (a^\dagger)^n |0\rangle, \quad (5.134)$$

where $\Psi(q, 0)$ is given by (5.126) at $t = 0$. In view of (5.130), the sum in (5.134) contains terms of the form $\frac{1}{\sqrt{n!}} \langle q|n\rangle$. But $\langle q|n\rangle$ are the wave functions given by (5.110), so

$$\langle q|z\rangle = \left(\frac{m\omega}{\pi\hbar}\right)^{\frac{1}{4}} c_0 \left[\sum_{n=0}^{\infty} \left(\frac{z}{\sqrt{2}}\right)^n \frac{1}{n!} H_n \left(\sqrt{\frac{m\omega}{\hbar}} q\right) \right] \exp \left[-\frac{m\omega}{2\hbar} q^2\right]. \quad (5.135)$$

The sum in the bracket is the generating function of the Hermite polynomials:

$$\sum_{n=0}^{\infty} \left(\frac{z}{\sqrt{2}}\right)^n \frac{1}{n!} H_n \left(\sqrt{\frac{m\omega}{\hbar}} q\right) = \exp \left[-\frac{z^2}{2} + \sqrt{\frac{2m\omega}{\hbar}} zq\right], \quad (5.136)$$

so we get

$$\langle q|z\rangle = \left(\frac{m\omega}{\pi\hbar}\right)^{\frac{1}{4}} c_0 \exp \left[-\frac{z^2}{2} + \sqrt{\frac{2m\omega}{\hbar}} zq\right] \exp \left[-\frac{m\omega}{2\hbar} q^2\right]. \quad (5.137)$$

Comparing this expression with the one for $\Psi(q, 0)$ from (5.123), we have

$$-\frac{z^2}{2} + \sqrt{\frac{2m\omega}{\hbar}}zq + \log_e c_0(z) = \sqrt{\frac{m\omega}{\hbar}}qf(0) - \frac{1}{2}f(0)^2 + \frac{i}{\omega}\sqrt{\frac{m\omega}{\hbar}}q\dot{f}(0) - i\frac{\dot{f}(0)f(0)}{2\omega} \quad (5.138)$$

Collecting like powers of q on both sides, we see that

$$\sqrt{\frac{m\omega}{\hbar}}\sqrt{2}z = \sqrt{\frac{m\omega}{\hbar}}\left[f(0) + \frac{i}{\omega}\dot{f}(0)\right] \implies z = \frac{1}{\sqrt{2}}\left[f(0) + i\frac{\dot{f}(0)}{\omega}\right] \quad (5.139)$$

and hence

$$\begin{aligned} \log_e c_0 &= \frac{1}{2}\left(z^2 - f(0)^2 - i\frac{\dot{f}(0)f(0)}{\omega}\right) \\ &= \frac{1}{4}\left[-f(0)^2 - \left(\frac{\dot{f}(0)}{\omega}\right)^2\right] \\ &= -\frac{|z|^2}{2} \end{aligned} \quad (5.140)$$

After all that work we are finally ready to write up the general coherent state for the harmonic oscillator:

$$|z\rangle = e^{-\frac{|z|^2}{2}}e^{za^\dagger}|0\rangle = D(z)|0\rangle, \quad (5.141)$$

where z is a complex number whose real and complex parts are the starting position and starting velocity, respectively, of the corresponding classical oscillator. But wait: was not D supposed to be unitary? To fix this problem, we can expand (5.141) as

$$|z\rangle = e^{-\frac{|z|^2}{2}}e^{za^\dagger}e^{-z^*a}|0\rangle = D(z)|0\rangle \quad (5.142)$$

and use $[a, a^\dagger] = 1$ and the Baker–Campbell–Hausdorff formula

$$e^A e^B e^{-\frac{1}{2}[A, B]} = e^{A+B} \quad (5.143)$$

to write

$$D(z) = \exp\left[za^\dagger - z^*a\right]. \quad (5.144)$$

This is the fundamental result of our discussion of harmonic oscillator coherent states. D is called a displacement operator because it displaces a state by z in phase space. So a coherent state is constructed by taking the ground state and displacing it in phase space (i.e. giving the particle an initial position $q_0 = f(0)$ and initial velocity $v_0 = \dot{f}(0)$). Before we move on to the 2M coherent states, we note that the harmonic

oscillator coherent state evolves in time as

$$\begin{aligned}
\exp\left[-\frac{\mathcal{H}_{har} t}{\hbar}\right] |z\rangle &= e^{-\frac{|z|^2}{2}} e^{-\frac{i}{2}\omega t} e^{-i\omega t a^\dagger a} \sum_{n=0}^{\infty} \frac{z^n}{\sqrt{n!}} |n\rangle \\
&= e^{-\frac{|z|^2}{2}} e^{-\frac{i}{2}\omega t} \sum_{n=0}^{\infty} \frac{z^n}{\sqrt{n!}} e^{-i\omega t a^\dagger a} |n\rangle \\
&= e^{-\frac{|z|^2}{2}} e^{-\frac{i}{2}\omega t} \sum_{n=0}^{\infty} \frac{z^n}{\sqrt{n!}} e^{-i\omega t n} |n\rangle \\
&= e^{-\frac{|z|^2}{2}} e^{-\frac{i}{2}\omega t} \sum_{n=0}^{\infty} \frac{(ze^{-i\omega t})^n}{\sqrt{n!}} |n\rangle \\
&= e^{-\frac{i}{2}\omega t} |ze^{i\omega t}\rangle.
\end{aligned} \tag{5.145}$$

This means $z(t) = z(0)e^{-i\omega t} = \frac{1}{\sqrt{2}} \left(f(t) + i\frac{\dot{f}(t)}{\omega} \right)$, as expected, where $f(t)$ solves the classical equation of motion for the harmonic oscillator (5.109).

5.6.2 2M coherent states and macroscopic dynamics

Having found the coherent states for the harmonic oscillator, it is natural to look at what properties, if any, can be generalized to other quantum systems, and how we can construct such coherent states. We will start by looking at the latter problem. Our starting point will be the displacement operator $D(z)$ in (5.144), which we saw constructed the coherent states from the ground state of the harmonic oscillator. This operator is called the Weyl-Heisenberg translation operator because it is an element of a group called the Weyl-Heisenberg group. The Lie algebra of this group is spanned by operators q, p, I that satisfy

$$[q, p] = i\hbar I, \quad [q, I] = 0, \quad [p, I] = 0. \tag{5.146}$$

The more commonly used complexified Lie algebra is spanned by the raising and lowering operators a, a^\dagger together with the identity operator:

$$[a, a^\dagger] = I, \quad [a, I] = 0, \quad [a^\dagger, I] = 0. \tag{5.147}$$

The extension of coherent states from the Weyl-Heisenberg group to an arbitrary Lie group is due to Perelomov [38]. These states are therefore called Perelomov coherent states. The definition is rather long and involved, but the upshot is that for any system of operators X, Y, Z that form a closed algebra, that is

$$[X, Y] = c_z Z, \quad [Y, Z] = c_x X, \quad [Z, X] = c_y Y, \tag{5.148}$$

where $c_x, c_y, c_z \in \mathbb{C}$, the generalized Perelomov coherent states are constructed from a state $|\psi\rangle$ in Hilbert space by a unitary, irreducible representation T of a Lie group G whose Lie algebra is given by (5.148). So for any group element $g \in G$, the operator

$$T(g) = \exp [ig \cdot \mathbf{v}] \tag{5.149}$$

with $g = X\hat{e}_1 + Y\hat{e}_2 + Z\hat{e}_3$ and $\mathbf{v} = v_1\hat{e}_1 + v_2\hat{e}_2 + v_3\hat{e}_3$, produces a Perelomov coherent state

$$|\mu\rangle = T(g)|\psi\rangle, \tag{5.150}$$

from any state $|\psi\rangle$ in Hilbert space. Hence, for each state $|\psi\rangle$ there is an associated set of coherent states $\{T(g)|\psi\rangle\}$. As was pointed out in Section 5.3.2, the 2M operators are a basis for the complexified Lie algebra of SU(1,1). Our task, therefore, is to construct a set of coherent states for this group. In order to achieve this, we need to

1. Construct a basis for the Lie algebra of SU(1,1)
2. Choose a suitable state $|\psi\rangle$ from which to construct a set of coherent states
3. Parameterize the coherent states

The idea is to use the obtained parameter to study the ultrafast dynamics semiclassically.

1: Basis for the algebra. We will start with a basis for the regular Lie algebra of SU(1,1) and then move to the complexified version once we have constructed the coherent states. The basis we will use is $\{K^x, K^y, K^z\}$. The commutation relations between these operators are

$$[K^x, K^y] = -iK^z, \quad [K^y, K^z] = iK^x, \quad [K^z, K^x] = iK^y. \quad (5.151)$$

I will use the shorthand notation $\mathbf{K} = K^x\hat{x} + K^y\hat{y} + K^z\hat{z}$. The operator that produces the 2M coherent states can now be written

$$T(\mathbf{r}) = \exp[i\mathbf{K} \cdot \mathbf{v}(\mathbf{r})], \quad \mathbf{v}(\mathbf{r}) \in \mathbb{R}^3. \quad (5.152)$$

The significance of the vector \mathbf{r} will become apparent in a few moments. For now it is just a vector specifying an element of SU(1,1).

2: Choice of starting state. Next, we need to choose an appropriate starting state. It is difficult to say *a priori* which state will be easiest to work with, but we can take inspiration from the harmonic oscillator states and choose the 2M ground state. We will therefore define the 2M coherent states as the set of states

$$T(\mathbf{r})|0\rangle \quad (5.153)$$

where $K^z|0\rangle = \frac{1}{2}|0\rangle$.

3: Parameterization. With everything properly defined, we can begin the process of parameterizing the coherent states. We could of course use the vector \mathbf{v} from (5.152), but we can do better than that. According to the discussion in Section 5.3.2, SU(1,1) transformations are rotations on the upper half of the hyperboloid H^2 defined in (5.38) and restated below:

$$H^2 = \{\mathbf{r} = r_x\hat{x} + r_y\hat{y} + r_z\hat{z} \mid r_x^2 + r_y^2 - r_z^2 = -1\}.$$

That means we can associate each coherent state $T(\mathbf{r})|0\rangle$ with the point \mathbf{r} on the upper sheet of H^2 . Parameterizing the coherent states then becomes equivalent to parameterizing the vector \mathbf{r} . We will use the parameterization

$$\mathbf{r}(\theta, \phi) = (\sinh \theta \cos \phi) \hat{x} + (\sinh \theta \sin \phi) \hat{y} + (\cosh \theta) \hat{z}, \quad (5.154)$$

where $\theta, \phi \in \mathbb{R}$ are the new parameters. The unrotated vector $\mathbf{r}(0,0) = \mathbf{r}_0 = \hat{z}$ corresponds to the 2M starting state $|0\rangle$.

I claim that with the chosen parameterization, every \mathbf{r} can be written

$$\mathbf{r}(\theta, \phi) = \exp[-i\theta\mathbf{K} \cdot \mathbf{w}] \mathbf{r}_0, \quad \mathbf{w} = \cos\left[\phi - \frac{\pi}{2}\right] \hat{x} + \sin\left[\phi - \frac{\pi}{2}\right] \hat{y}. \quad (5.155)$$

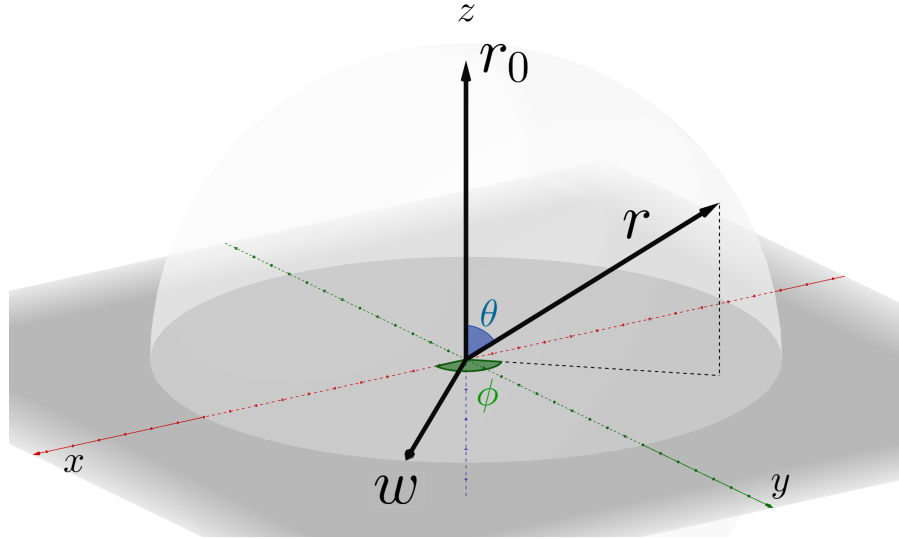


FIGURE 5.4: If K were a basis for the Lie algebra of $SO(3)$, the rotation $\exp[-i\theta K \cdot w]$ would produce $r = \sin \theta \cos \phi \hat{x} + \sin \theta \sin \phi \hat{y} + \cos \theta \hat{z}$ from r_0 .

That is, r can be written as a rotation of r_0 around the vector w by $-\theta$. If the operators K were a basis for the Lie algebra of $SO(3)$, this rotation would be on a sphere as depicted in Fig. 5.4. However, with K a basis for the $SU(1,1)$ Lie algebra, this rotation is lifted to the pseudosphere $\{r \in H^2 \mid r_z > 0\}$. When we insert the expression for w , we obtain

$$\begin{aligned}
 T(r) &= \exp \left[-i\theta \left(\cos \left[\phi - \frac{\pi}{2} \right] K^x + \sin \left[\phi - \frac{\pi}{2} \right] \hat{K}^y \right) \right] \\
 &= \exp \left[-i\theta (\sin \phi K^x - \cos \phi K^y) \right] \\
 &= \exp \left[\frac{\theta}{2} (-2i \sin \phi K^x + \cos \phi K^x - \cos \phi K^x + 2i \cos \phi K^y + \sin \phi K^y - \sin \phi K^y) \right] \\
 &= \exp \left[\frac{\theta}{2} \left(e^{-i\phi} K^x + i e^{-i\phi} K^y - \left(e^{i\phi} K^x - i e^{i\phi} K^y \right) \right) \right] \\
 &= \exp \left[\frac{\theta}{2} \left(e^{-i\phi} (K^x + i K^y) - e^{i\phi} (K^x - i K^y) \right) \right].
 \end{aligned} \tag{5.156}$$

But the operators $K^\pm = K^x \pm i K^y$ are the 2M creation and annihilation operators because their commutation relations with K^z are (5.19). So the operator

$$T(\epsilon) = \exp [\epsilon K^+ - \epsilon^* K^-] \tag{5.157}$$

is the constructor of a 2M coherent state

$$|\epsilon\rangle = \exp [\epsilon K^+ - \epsilon^* K^-] |0\rangle \tag{5.158}$$

parameterized by the complex number $\epsilon = \frac{\theta}{2} e^{-i\phi}$, where θ, ϕ parameterize a rotation on the upper half of the hyperboloid H^2 .

Looking back at the expression of the time-evolution operator (5.66), we see that

the evolution of a 2M mode $|n_k^\alpha\rangle|n_{-k}^\beta\rangle \equiv |n_k\rangle$ looks somewhat like a Perelomov coherent state:

$$U_k(t,0)|n_k\rangle = \exp[-i\tau\{V_k K_k^+(t) + V_k K_k^-(t) + 2\delta\omega_k K_k^z(t)\}] |n_k\rangle. \quad (5.159)$$

If we assume that each mode is unoccupied at $t = 0$, i.e. the antiferromagnet starts out in the ground state, then to first order in τ we have

$$\begin{aligned} U_k|0_k\rangle &\approx |0_k\rangle + (-i\tau\{V_k K_k^+(t) + V_k K_k^-(t) + 2\delta\omega_k K_k^z(t)\}) |0_k\rangle \\ &= |0_k\rangle - i\tau V_k K_k^+(t) |0_k\rangle - i\tau V_k K_k^-(t) |0_k\rangle - i\tau \delta\omega_k |0_k\rangle \\ &= (1 - i\tau \delta\omega_k) |0_k\rangle - i\tau V_k K_k^+(t) |0_k\rangle - i\tau V_k K_k^-(t) |0_k\rangle \\ &\approx e^{-i\tau \delta\omega_k} |0_k\rangle - i\tau V_k K_k^+(t) |0_k\rangle - i\tau V_k K_k^-(t) |0_k\rangle \end{aligned} \quad (5.160)$$

The first term on the right is just the unoccupied mode times an irrelevant phase factor, so we can absorb the phase into the ket:

$$e^{-i\tau \delta\omega_k} |0_k\rangle \rightarrow |0_k\rangle. \quad (5.161)$$

This leads to

$$\begin{aligned} U_k|0_k\rangle &\approx |0_k\rangle - i\tau V_k K_k^+(t) e^{i\tau \delta\omega_k} |0_k\rangle - i\tau V_k K_k^-(t) e^{i\tau \delta\omega_k} |0_k\rangle \\ &\approx \exp[-i\tau V_k K_k^+(t) e^{i\tau \delta\omega_k} - i\tau V_k K_k^-(t) e^{i\tau \delta\omega_k}] |0_k\rangle \\ &= \exp[-i\tau V_k K_k^+ e^{2i\omega_k t} e^{i\tau \delta\omega_k} - i\tau V_k K_k^- e^{-2i\omega_k t} e^{i\tau \delta\omega_k}] |0_k\rangle \\ &= T(\epsilon_k) |0_k\rangle, \end{aligned} \quad (5.162)$$

with $\epsilon_k = -i\tau V_k e^{2i\omega_k t + i\tau \delta\omega_k}$. We may write a 2M coherent state in terms of the Fock states as [39]

$$U_k|0_k\rangle \equiv |\mu_k\rangle = \sqrt{1 - |\mu_k|^2} \sum_{n=0}^{\infty} \mu_k^n |n\rangle, \quad (5.163)$$

where $\mu_k = \tanh\left(\frac{\theta}{2}\right) e^{-i\phi}$ with

$$\theta = -2\tau V_k, \quad \phi = 2\omega_k t + \tau \delta\omega_k + \frac{\pi}{2}. \quad (5.164)$$

It is interesting to see which properties of the harmonic oscillator coherent states have survived the generalization to Perelomov coherent states. For instance, are the Perelomov coherent states minimal uncertainty states? A paper by Wódkiewicz and Eberly [40] gives us the variances in the Hermitian operators K_k^x, K_k^y in a coherent state:

$$\Delta_{K_k^x}^2 = \frac{1}{4} \left(1 + \sinh^2(\theta) \cos^2(\phi)\right), \quad (5.165)$$

$$\Delta_{K_k^y}^2 = \frac{1}{4} \left(1 + \sinh^2(\theta) \sin^2(\phi)\right). \quad (5.166)$$

The expectation value of their commutator is

$$|\langle [K_k^x, K_k^y] \rangle| = i |\langle K_k^z \rangle| = \frac{i}{2} \cosh(\theta). \quad (5.167)$$

Inserted into the uncertainty relation (2.48), we obtain (after squaring both sides):

$$\left(1 + \sinh^2(\theta) \cos^2(\phi)\right) \left(1 + \sinh^2(\theta) \sin^2(\phi)\right) \geq \cosh^2(\theta). \quad (5.168)$$

We notice that the lower uncertainty limit is now a function of how far the coherent state $|\mu_k\rangle = T(\theta, \phi)|0_k\rangle$ has been displaced the ground state. The 2M ground state ($\theta = 0$) is the state with the lowest uncertainty, and any other coherent state ($\theta \neq 0$) will have greater uncertainty in K_k^x and K_k^y . This is true even if ϕ is an integer multiple of 2π , in which case (5.168) holds with equality.

The generalized coherent states have therefore lost the property of being minimal uncertainty states. In fact, the generalizations of coherent states (of which Perelomov's is one) from the harmonic oscillator to general systems are more concerned with preserving the *mathematical* properties of harmonic oscillator coherent states than the *physical* properties [39, 41]. In some cases the generalized coherent states are the quantum states that physically resemble classical states, but from what I can tell this is more a serendipitous perk possessed by a limited number of generalized coherent states than a requirement for the label "coherent" in mathematical physics. So what is the point of introducing the Perelomov coherent states if we cannot retain the relation to the classical description that we get with ordinary coherent states? The fact that the Perelomov coherent states are not very close to classical states does not mean they are not the *closest*, as stated by Bossini *et al.*[18] but not justified. So if we are looking for a classical description, the Perelomov coherent states might be the best we can do. However, and I suspect more importantly, the time-evolution operator naturally produce Perelomov coherent states from the ground state, which is very convenient. Since a coherent state is completely specified by the parameter μ_k , we have a parameter that can be used to represent the macroscopic spin dynamics in antiferromagnets. Also, for the 2M operators the displacement operator (5.157) is on the same form as the two-mode squeezing operator from quantum optics [42]:

$$S(\epsilon_k) = T(-\epsilon_k) = \exp[\epsilon_k^* K_k^- - \epsilon_k K_k^+]. \quad (5.169)$$

In this sense the coherent states can be viewed as a form of squeezed states. Usually squeezed states are time-independent states with reduced quantum noise compared to the some other time-independent state. The Perelomov coherent states used here are not time-independent, so some caution is advised when thinking about squeezing in this context.

We see from Fig. 5.3 that the oscillations triggered by the excitation of magnons actually periodically compensate for the reduction in the Néel vector length caused by ground state magnons. In the case where the coherent states $|\mu_k\rangle$ have the same frequency and phase (i.e. classical coherence), the oscillations in the states shown in Fig. 5.3 will add constructively and cause the Néel vector's oscillation to become detectable on a macroscopic level. The degree to which the coherent states are classically coherent is determined by the k -dependence of ω_k and $\delta\omega_k$; slowly varying ω_k and $\delta\omega_k$ with k will produce the strong classical coherence needed for macroscopically detectable Néel vector oscillations.

We will now treat μ_k as a classical parameter and look for the classical equation of motion that μ_k must satisfy. Of course, the classical dynamics of SU(1,1) coherent states has been studied before by mathematical physicists, for example in [43]. The equation of motion is

$$\frac{\partial \mu_k}{\partial t} = \{\mu_k, \mathcal{H}_{\text{class}}\}, \quad (5.170)$$

where

$$\mathcal{H}_{\text{class}} = \prod_k \langle \mu_k | (\mathcal{H}_0 + \delta \mathcal{H}) | \mu_k \rangle \quad (5.171)$$

and the Poisson bracket $\{ \}$ is shorthand for

$$\{ \mu_k, \mathcal{H}_{\text{class}} \} = \frac{(1 - |\mu_k|^2)^2}{i} \left(\frac{\partial \mu_k}{\partial \mu_k} \frac{\partial \mathcal{H}_{\text{class}}}{\partial \mu_k^*} - \frac{\partial \mu_k}{\partial \mu_k^*} \frac{\partial \mathcal{H}_{\text{class}}}{\partial \mu_k} \right). \quad (5.172)$$

Inserting the Schrödinger Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \delta \mathcal{H}$ from (5.17) into (5.171) yields

$$\mathcal{H}_{\text{class}} = \sum_k [(\omega_k + f(t)\delta\omega_k) \langle K_k^z \rangle_{\mu_k} + f(t)V_k (\langle K_k^+ \rangle_{\mu_k} + \langle K_k^- \rangle_{\mu_k})]. \quad (5.173)$$

In terms of μ_k the expectation values of the time-independent 2M operators are [42]

$$\langle K_k^+ \rangle_{\mu_k} = \frac{\mu_k^*}{1 - |\mu_k|^2}; \quad \langle K_k^- \rangle_{\mu_k} = \frac{\mu_k}{1 - |\mu_k|^2}; \quad \langle K_k^z \rangle_{\mu_k} = \frac{1}{2} \frac{1 + |\mu_k|^2}{1 - |\mu_k|^2}. \quad (5.174)$$

Using (5.171)-(5.174) we get that (5.170) reads

$$i \frac{\partial \mu_k}{\partial t} = (1 - |\mu_k|^2)^2 \frac{\partial \mathcal{H}_{\text{class}}}{\partial \mu_k^*} = 2((\omega_k + f(t)\delta\omega_k) \mu_k + f(t)V_k (1 + \mu_k^2)). \quad (5.175)$$

This is the quasi-classical equation of motion for the parameter that represents the spin dynamics on the femtosecond scale in antiferromagnets. In the classical description of Néel vector dynamics, any change in the Néel vector induces a change in the antiferromagnet's magnetization because purely longitudinal variations are not allowed. Not so in the quasi-classical description we have derived. To first order in τ we can express the longitudinal dynamics of the Néel vector using (5.54) and (5.174) as

$$\langle L^z \rangle = Ns - \sum_k \frac{1}{\sqrt{1 - \gamma_k^2}} \frac{1 + |\mu_k|^2}{1 - |\mu_k|^2} + \sum_k \frac{\gamma_k}{\sqrt{1 - \gamma_k^2}} \frac{2\text{Re}\{\mu_k\}}{1 - |\mu_k|^2}, \quad (5.176)$$

and together (5.175) and (5.176) complement the traditional Landau-Lifshitz equations for antiferromagnet dynamics. Whereas the Landau-Lifshitz equations describe precessional dynamics on the timescale of 100 ps [18], the parameters μ_k capture the quantum dynamics that happens on considerably smaller timescales: for an exchange interaction $J \sim 100$ meV we get a period of oscillations on the order of 10 fs. The average number of magnon pairs \bar{n}_k with wavevector $\pm k$ in the antiferromagnet is given by

$$\begin{aligned} \bar{n}_k &= \frac{1}{2} \left(\overline{\alpha_k^\dagger \alpha_k} + \overline{\beta_{-k}^\dagger \beta_{-k}} \right) \\ &= \langle K_k^z \rangle_{\mu_k} - \frac{1}{2} \\ &= \frac{1}{2} \frac{1 + |\mu_k|^2}{1 - |\mu_k|^2} - \frac{1}{2} \\ &= \frac{|\mu_k|^2}{1 - |\mu_k|^2}. \end{aligned} \quad (5.177)$$

The probability of observing n_k magnons in a mode is

$$\begin{aligned} |\langle n_k | \mu_k \rangle|^2 &= (1 - |\mu_k|^2) |\mu_k|^{2n_k} \\ &= (1 - |\mu_k|^2)^{n_k+1} \frac{|\mu_k|^{2n_k}}{(1 - |\mu_k|^2)^{n_k}} \\ &= \frac{\bar{n}_k^{n_k}}{(1 + \bar{n}_k)^{n_k+1}}. \end{aligned} \tag{5.178}$$

Chapter 6

Excitation from Nonuniform Fields

Until now we have assumed that the electric field that causes the change in the exchange interaction is uniform across the antiferromagnet. At any particular instance in time, the electric field from the laser pulse used to excite magnons was assumed to be

$$E(\mathbf{r}) = E_0. \quad (6.1)$$

We will now look at the consequences of allowing the electric field to vary spatially. We will use the electric field

$$E(\mathbf{r}) = E_0 \sqrt{g(\mathbf{r})}, \quad (6.2)$$

where the function $g(\mathbf{r}) \geq 0$ captures the spatial profile of the electric field strength. This allows us to write the light-induced perturbation to the exchange interaction as

$$\Delta J = \Delta J(\delta) g(\mathbf{r}). \quad (6.3)$$

The perturbation Hamiltonian is a discrete function of space, so we have to decide in advance how to sample g at various positions \mathbf{r} . We will choose to sample g at the middle of each bond. This means that for a spin located at \mathbf{r}_j , $\Delta J = \Delta J(\delta) g(\mathbf{r}_j + \frac{1}{2}\delta)$ and the perturbation Hamiltonian becomes

$$\delta \mathcal{H}(\mathbf{r}_j, t) = \frac{1}{2} f(t) \sum_j \sum_{\delta} \Delta J(\delta) g(\mathbf{r}_j + \frac{1}{2}\delta) \mathbf{S}_j \cdot \mathbf{S}_{j+\delta}. \quad (6.4)$$

This choice of sampling preserves the symmetry of the Hamiltonian so that we may express it as a sum over one sublattice only:

$$\delta \mathcal{H}(\mathbf{r}_j, t) = f(t) \sum_{j \in A} \sum_{\delta} \Delta J(\delta) g\left(\mathbf{r}_j + \frac{1}{2}\delta\right) \mathbf{S}_j \cdot \mathbf{S}_{j+\delta} \quad (6.5)$$

We may also continue using the Holstein-Primakoff transformation to express $\delta \mathcal{H}$ in terms of Bose operators. In particular, the Hamiltonian (6.4) after the transformation reads

$$\delta \mathcal{H} = f(t) \sum_j \sum_{\delta} \Delta J(\delta) \left(-s^2 + sa_j^\dagger a_j + sb_{j+\delta}^\dagger b_{j+\delta} + sa_j^\dagger b_{j+\delta}^\dagger + sa_j b_{j+\delta} \right) g\left(\mathbf{r}_j + \frac{1}{2}\delta\right) \quad (6.6)$$

However, the Bogoliubov transformation we applied in Chapter 5 will no longer help us reduce the complexity of the perturbation Hamiltonian. In Section 6.1 we continue to use the Bogoliubov transformation to (6.6) and look at the effect of $g(\mathbf{r})$ on the Néel vector dynamics. In Section 6.2 we will employ a more general transformation and look for a way to obtain the quantum states numerically after the perturbation is switched off, which involves use of the sudden approximation.

6.1 Fourier analysis

The success of the Bogoliubov transformation in diagonalizing a Hamiltonian of the form (4.24) is owed to the fact that every Bose operator a_k , b_{-k} and their adjoints always appear together with a Bose operator with the same or opposite wave vector. The Fourier transform takes advantage of the translational invariance of the Hamiltonian so that "interference" on the form (5.80) kills all other operators. Thus simple linear combinations of the Bose operators on the form of (4.25) and (4.26) provide an elegant solution to the diagonalization problem. We are bound to run into trouble once this translational invariance breaks down if we continue to use the same transformation. Still, there is one very good reason for sticking with it: the unperturbed Hamiltonian is diagonal after the transformation is applied. So as long as we are not forced to do too many calculations with the perturbation, continuing with the Bogoliubov transformation is reasonable. If we try to apply a different transformation to reduce the complexity of the perturbation Hamiltonian, we gain complexity in the unperturbed Hamiltonian. In this section we will continue using Fourier analysis – despite the loss of translational invariance – because \mathcal{H}_0 is easy to work with in Fourier space. Like before we express each operator and the function g in terms of Fourier series:

$$a_j = \sqrt{\frac{2}{N}} \sum_k \exp [ik \cdot r_j] a_k \quad (6.7)$$

$$b_{j+\delta} = \sqrt{\frac{2}{N}} \sum_k \exp [ik \cdot r_j] \exp [ik \cdot \delta] b_k \quad (6.8)$$

$$g \left(r_j + \frac{1}{2} \delta \right) = \sum_k \exp [ik \cdot r_j] \exp \left[i \frac{1}{2} k \cdot \delta \right] g_k. \quad (6.9)$$

The choice to factor out $\sqrt{N_{unit}^{-1}}$ from the Bose operators a_k and b_k but not from g_k is in anticipation of the resulting expression for $\delta \mathcal{H}$, which will take a simpler and more familiar form using this definition. Since we are now going to work with products of three different Fourier expansions, I will again switch notation so that wave vectors are represented by λ , μ and ν . The first double sum in the Hamiltonian (6.6) is

$$-s^2 \sum_j \sum_\delta \Delta J(\delta) g \left(r_j + \frac{1}{2} \delta \right) = -s^2 G, \quad (6.10)$$

The second double sum is

$$\begin{aligned} \sum_j \sum_\delta \Delta J(\delta) a_j^\dagger a_j g(r_j) &= \frac{2}{N} \sum_j \sum_\delta \Delta J(\delta) \left[\sum_\lambda e^{-i\lambda \cdot r_j} a_\lambda^\dagger \right] \left[\sum_\mu e^{i\mu \cdot r_j} a_\mu \right] \left[\sum_\nu e^{i\nu \cdot r_j} e^{\frac{i}{2} \nu \cdot \delta} g_\nu \right] \\ &= \frac{2}{N} \sum_j \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu \sum_\nu a_\lambda^\dagger a_\mu g_\nu e^{i(\nu - (\lambda - \mu)) \cdot r_j} e^{\frac{i}{2} \nu \cdot \delta} \\ &= \frac{2}{N} \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu \sum_\nu a_\lambda^\dagger a_\mu g_\nu e^{\frac{i}{2} \nu \cdot \delta} \sum_j e^{i(\nu - (\lambda - \mu)) \cdot r_j} \\ &= \frac{2}{N} \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu \sum_\nu a_\lambda^\dagger a_\mu g_\nu e^{\frac{i}{2} \nu \cdot \delta} \frac{N}{2} \delta_{\nu, \lambda - \mu} \\ &= \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu a_\lambda^\dagger a_\mu g_{\lambda - \mu} e^{\frac{i}{2} (\lambda - \mu) \cdot \delta}. \end{aligned} \quad (6.11)$$

We see that unless the only nonzero Fourier coefficient of $g(\mathbf{r}_j + \frac{1}{2}\delta)$ is g_0 , we get contributions from operators a_λ and a_μ^\dagger that act on states with different wave vectors. The upshot is that we have transformed the operators $a_j^\dagger a_j$, which produce diagonal terms in the Hamiltonian matrix, into a sum of operators $a_\lambda^\dagger a_\mu$ that produce off-diagonal terms in that matrix as well. By repeating the steps in (6.11) we find the last three double sums in (6.6) to be

$$\sum_j \sum_\delta \Delta J(\delta) b_{j+\delta}^\dagger b_{j+\delta} g(\mathbf{r}_j) = \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu b_\lambda^\dagger b_\mu g_{\lambda-\mu} e^{\frac{i}{2}(\mu-\lambda)\cdot\delta}, \quad (6.12)$$

$$\sum_j \sum_\delta \Delta J(\delta) a_j^\dagger b_{j+\delta}^\dagger g(\mathbf{r}_j) = \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu a_\lambda^\dagger b_{-\mu}^\dagger g_{\lambda-\mu} e^{\frac{i}{2}(\lambda+\mu)\cdot\delta}, \quad (6.13)$$

$$\sum_j \sum_\delta \Delta J(\delta) a_j b_{j+\delta} g(\mathbf{r}_j) = \sum_\delta \Delta J(\delta) \sum_\lambda \sum_\mu a_\lambda b_{-\mu} g_{\mu-\lambda} e^{-\frac{i}{2}(\lambda+\mu)\cdot\delta}. \quad (6.14)$$

Putting it all together yields the following expression for $\delta \mathcal{H}$ in Fourier space:

$$\begin{aligned} \delta \mathcal{H} = & -s^2 G f(t) \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu a_\lambda^\dagger a_\mu g_{\lambda-\mu} \xi_{\frac{\lambda-\mu}{2}} + b_\lambda^\dagger b_\mu g_{\lambda-\mu} \xi_{\frac{\mu-\lambda}{2}} \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu a_\lambda^\dagger b_{-\mu}^\dagger g_{\lambda-\mu} \xi_{\frac{\lambda+\mu}{2}} + a_\lambda b_{-\mu} g_{\mu-\lambda} \xi_{\frac{-\lambda-\mu}{2}} \end{aligned} \quad (6.15)$$

where Λ and ξ_k are as defined in (5.12) and (5.13). It is easy to check that (6.15) reduces to (5.10) if the electric field is uniform, because then

$$g(\mathbf{r}_j + \frac{1}{2}\delta) = 1 \implies g_k = \delta_{k,0} \quad (6.16)$$

and all terms with $\mu \neq \lambda$ vanish.

We continue by applying the Bogoliubov transformation given by (4.29) and (4.30). I will abandon the notation with superscripts for ξ and g and instead use parentheses for clarity. I believe this will not cause too much confusion, and it should be clear from the context whether g is in real or Fourier space. After the transformation the resulting expression for $\delta \mathcal{H}$ is

$$\begin{aligned} \delta \mathcal{H} = & -s^2 f(t) G \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu g(\Delta) \xi\left(\frac{\Delta}{2}\right) \left[u_\lambda u_\mu \alpha_\lambda^\dagger \alpha_\mu + u_\lambda v_\mu \alpha_\lambda^\dagger \beta_{-\mu}^\dagger + u_\mu v_\lambda \alpha_\mu \beta_{-\lambda} + v_\lambda v_\mu \beta_{-\lambda} \beta_{-\mu}^\dagger \right] \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu g(-\Delta) \xi\left(\frac{\Delta}{2}\right) \left[u_\lambda u_\mu \beta_{-\lambda}^\dagger \beta_{-\mu} + u_\lambda v_\mu \beta_{-\lambda}^\dagger \alpha_\mu^\dagger + u_\mu v_\lambda \beta_{-\mu} \alpha_\lambda + v_\lambda v_\mu \alpha_\lambda \alpha_\mu^\dagger \right] \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu g(\Sigma) \xi\left(\frac{\Sigma}{2}\right) \left[u_\lambda u_\mu \alpha_\lambda^\dagger \beta_{-\mu}^\dagger + u_\lambda v_\mu \alpha_\lambda^\dagger \alpha_\mu + u_\mu v_\lambda \beta_{-\lambda} \beta_{-\mu}^\dagger + v_\lambda v_\mu \alpha_\mu \beta_{-\lambda} \right] \\ & + f(t) s \Lambda \sum_\lambda \sum_\mu g(-\Sigma) \xi\left(\frac{\Sigma}{2}\right) \left[u_\lambda u_\mu \alpha_\lambda \beta_{-\mu} + u_\lambda v_\mu \alpha_\lambda \alpha_\mu^\dagger + u_\mu v_\lambda \beta_{-\lambda}^\dagger \beta_{-\mu} + v_\lambda v_\mu \alpha_\mu^\dagger \beta_{-\lambda}^\dagger \right] \end{aligned} \quad (6.17)$$

where I have defined

$$\Delta = \lambda - \mu, \quad \Sigma = \lambda + \mu, \quad (6.18)$$

for cleaner notation. We collect equal terms in the magnon operators and interchange summation indices where appropriate to obtain

$$\begin{aligned}
\delta \mathcal{H} = & -s^2 f(t)G + f(t)s\Lambda g(\mathbf{0}) \sum_{\lambda} (2v_{\lambda}^2 + 2u_{\lambda}v_{\lambda}\xi_{\lambda}) \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu} g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda}u_{\mu} + v_{\lambda}v_{\mu}] + \xi \left(\frac{\Sigma}{2} \right) [u_{\lambda}v_{\mu} + v_{\lambda}u_{\mu}] \right) \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \beta_{-\mu}^{\dagger} \beta_{-\lambda} g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda}u_{\mu} + v_{\lambda}v_{\mu}] + \xi \left(\frac{\Sigma}{2} \right) [u_{\lambda}v_{\mu} + v_{\lambda}u_{\mu}] \right) \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger} g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda}v_{\mu} + v_{\lambda}u_{\mu}] + \xi \left(\frac{\Sigma}{2} \right) [u_{\lambda}u_{\mu} + v_{\lambda}v_{\mu}] \right) \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \alpha_{\mu} \beta_{-\lambda} g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda}v_{\mu} + v_{\lambda}u_{\mu}] + \xi \left(\frac{\Sigma}{2} \right) [u_{\lambda}u_{\mu} + v_{\lambda}v_{\mu}] \right), \tag{6.19}
\end{aligned}$$

where I used the commutators

$$[\alpha_{\lambda}, \alpha_{\mu}^{\dagger}] = \delta_{\lambda, \mu}, \quad [\beta_{\lambda}, \beta_{\mu}^{\dagger}] = \delta_{\lambda, \mu}. \tag{6.20}$$

We see that $\delta \mathcal{H}$ for nonuniform fields contains operators $\alpha_{\lambda}^{\dagger} \alpha_{\mu}$ and $\beta_{-\mu}^{\dagger} \beta_{-\lambda}$ that shift the momentum of magnons as well as operators $\alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}$ and $\alpha_{\mu} \beta_{-\lambda}$ that create and destroy 2M states, respectively, that can carry a nonzero momentum. Using (6.18) to express μ in terms of λ and Δ , we get

$$\begin{aligned}
\delta \mathcal{H} = & -s^2 f(t)G + f(t)s\Lambda g(\mathbf{0}) \sum_{\lambda} (2v_{\lambda}^2 + 2u_{\lambda}v_{\lambda}\xi_{\lambda}) \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda}^{\dagger} \alpha_{\lambda-\Delta} g(\Delta) \xi \left(\frac{\Delta}{2} \right) [u_{\lambda}u_{\lambda-\Delta} + v_{\lambda}v_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda}^{\dagger} \alpha_{\lambda-\Delta} g(\Delta) \xi \left(\lambda - \frac{\Delta}{2} \right) [u_{\lambda}v_{\lambda-\Delta} + v_{\lambda}u_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \beta_{-\lambda+\Delta}^{\dagger} \beta_{-\lambda} g(\Delta) \xi \left(\frac{\Delta}{2} \right) [u_{\lambda}u_{\lambda-\Delta} + v_{\lambda}v_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \beta_{-\lambda+\Delta}^{\dagger} \beta_{-\lambda} g(\Delta) \xi \left(\lambda - \frac{\Delta}{2} \right) [u_{\lambda}v_{\lambda-\Delta} + v_{\lambda}u_{\lambda-\Delta}] \tag{6.21} \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda}^{\dagger} \beta_{-\lambda+\Delta}^{\dagger} g(\Delta) \xi \left(\frac{\Delta}{2} \right) [u_{\lambda}v_{\lambda-\Delta} + v_{\lambda}u_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda}^{\dagger} \beta_{-\lambda+\Delta}^{\dagger} g(\Delta) \xi \left(\lambda - \frac{\Delta}{2} \right) [u_{\lambda}u_{\lambda-\Delta} + v_{\lambda}v_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda-\Delta} \beta_{-\lambda} g(\Delta) \xi \left(\frac{\Delta}{2} \right) [u_{\lambda}v_{\lambda-\Delta} + v_{\lambda}u_{\lambda-\Delta}] \\
& + f(t)s\Lambda \sum_{\lambda} \sum_{\Delta} \alpha_{\lambda-\Delta} \beta_{-\lambda} g(\Delta) \xi \left(\lambda - \frac{\Delta}{2} \right) [u_{\lambda}u_{\lambda-\Delta} + v_{\lambda}v_{\lambda-\Delta}]
\end{aligned}$$

We cannot apply (4.32) and (4.36) that link u_{λ} and v_{λ} to the structure factor γ_{λ} directly except when $\Delta = \mathbf{0}$. I propose to use first-order expansions

$$u_{\lambda-\Delta} \approx u_{\lambda} - \nabla_{\lambda} u_{\lambda} \cdot \Delta, \quad v_{\lambda-\Delta} \approx v_{\lambda} - \nabla_{\lambda} v_{\lambda} \cdot \Delta, \tag{6.22}$$

which will carry an error of the order $|\Delta|^2$. The structure factor ξ can also be expanded in a first-order Taylor series. Keeping only terms that are linear in Δ yields

$$\begin{aligned} \xi\left(\frac{\Delta}{2}\right) [u_\lambda u_{\lambda-\Delta} + v_\lambda v_{\lambda-\Delta}] &= \xi(\mathbf{0}) [u_\lambda^2 + v_\lambda^2] + \frac{1}{2} \nabla_\lambda \xi(\mathbf{0}) \cdot \Delta [u_\lambda^2 + v_\lambda^2] \\ &\quad - \xi(\mathbf{0}) [u_\lambda \nabla_\lambda u_\lambda + v_\lambda \nabla_\lambda v_\lambda] \cdot \Delta. \end{aligned} \quad (6.23)$$

By using

$$\nabla_\lambda u_\lambda = \frac{\partial u}{\partial \gamma_\lambda} \nabla_\lambda \gamma_\lambda \quad (6.24)$$

and combining (4.28) and (4.36) to obtain

$$u_\lambda = \pm \sqrt{\frac{1}{2} + \frac{1}{2\sqrt{1-\gamma_\lambda^2}}}, \quad (6.25)$$

$$v_\lambda = \pm \sqrt{\frac{1}{2\sqrt{1-\gamma_\lambda^2}} - \frac{1}{2}}, \quad (6.26)$$

we get

$$\nabla_\lambda u_\lambda = \frac{\gamma_\lambda}{4u_\lambda} (1-\gamma_\lambda^2)^{-\frac{3}{2}} \nabla_\lambda \gamma_\lambda, \quad (6.27)$$

$$\nabla_\lambda v_\lambda = \frac{\gamma_\lambda}{4v_\lambda} (1-\gamma_\lambda^2)^{-\frac{3}{2}} \nabla_\lambda \gamma_\lambda. \quad (6.28)$$

In view of (4.36), the expression in (6.23) reduces to

$$\xi\left(\frac{\Delta}{2}\right) [u_\lambda u_{\lambda-\Delta} + v_\lambda v_{\lambda-\Delta}] = \frac{1}{\sqrt{1-\gamma_\lambda^2}} \left(1 + \frac{\Delta}{2} \left[\nabla_\lambda \xi(\mathbf{0}) - \frac{\gamma_\lambda \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right), \quad (6.29)$$

where I used $\xi(\mathbf{0}) = 1$. Similarly we see that

$$\xi\left(\lambda - \frac{\Delta}{2}\right) [u_\lambda u_{\lambda-\Delta} + v_\lambda v_{\lambda-\Delta}] = \frac{\xi(\lambda)}{\sqrt{1-\gamma_\lambda^2}} \left(1 - \frac{\Delta}{2} \left[\frac{\nabla_\lambda \xi(\lambda)}{\xi(\lambda)} + \frac{\gamma_\lambda \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right). \quad (6.30)$$

Next we will be looking at the cross terms of the form

$$\begin{aligned} \xi\left(\frac{\Delta}{2}\right) [u_\lambda v_{\lambda-\Delta} + v_\lambda u_{\lambda-\Delta}] &= \xi(\mathbf{0}) [2u_\lambda v_\lambda] + \frac{1}{2} \nabla_\lambda \xi(\mathbf{0}) \cdot \Delta [2u_\lambda v_\lambda] \\ &\quad - \xi(\mathbf{0}) [u_\lambda \nabla_\lambda v_\lambda + v_\lambda \nabla_\lambda u_\lambda] \cdot \Delta. \end{aligned} \quad (6.31)$$

We have

$$u_\lambda \nabla_\lambda v_\lambda + v_\lambda \nabla_\lambda u_\lambda = \left(\frac{u_\lambda}{v_\lambda} + \frac{v_\lambda}{u_\lambda} \right) \frac{\gamma_\lambda}{4} (1-\gamma_\lambda^2)^{-\frac{3}{2}} \nabla_\lambda \gamma_\lambda, \quad (6.32)$$

which, when we use (4.32) so that

$$\frac{u_\lambda}{v_\lambda} + \frac{v_\lambda}{u_\lambda} = \frac{u_\lambda^2 + v_\lambda^2}{u_\lambda v_\lambda} = -\frac{2}{\gamma_\lambda}, \quad (6.33)$$

transforms (6.31) into

$$\bar{\xi} \left(\frac{\Delta}{2} \right) [u_\lambda v_{\lambda-\Delta} + v_\lambda u_{\lambda-\Delta}] = -\frac{1}{\sqrt{1-\gamma_\lambda^2}} \left(\gamma_\lambda + \frac{\Delta}{2} \left[\gamma_\lambda \nabla_\lambda \bar{\xi}(\mathbf{0}) - \frac{\nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right). \quad (6.34)$$

Finally we have

$$\bar{\xi} \left(\lambda - \frac{\Delta}{2} \right) [u_\lambda v_{\lambda-\Delta} + v_\lambda u_{\lambda-\Delta}] = -\frac{\bar{\xi}(\lambda)}{\sqrt{1-\gamma_\lambda^2}} \left(\gamma_\lambda - \frac{\Delta}{2} \left[\frac{\gamma_\lambda \nabla_\lambda \bar{\xi}(\lambda)}{\bar{\xi}(\lambda)} + \frac{\nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right). \quad (6.35)$$

The structure factor $\bar{\xi}_\lambda$ is an even function for square antiferromagnets, so its gradient at the origin must be zero. Substituting (6.29), (6.30), (6.34) and (6.35) into the Hamiltonian (6.21) and collecting terms with the same operators, we obtain

$$\begin{aligned} \delta \mathcal{H} = & -f(t)s(sG + \Lambda g(\mathbf{0})) + f(t)s\Lambda g(\mathbf{0}) \sum_\lambda \delta\omega_\lambda \\ & + f(t)s\Lambda \sum_\lambda \sum_\Delta \alpha_\lambda^\dagger \alpha_{\lambda-\Delta} \frac{g(\Delta)}{\sqrt{1-\gamma_\lambda^2}} \left(1 - \gamma_\lambda \bar{\xi}_\lambda + \frac{\Delta}{2} \left[\gamma_\lambda \nabla_\lambda \bar{\xi}_\lambda + \frac{(\bar{\xi}_\lambda - \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right) \\ & + f(t)s\Lambda \sum_\lambda \sum_\Delta \beta_{-\lambda+\Delta}^\dagger \beta_{-\lambda} \frac{g(\Delta)}{\sqrt{1-\gamma_\lambda^2}} \left(1 - \gamma_\lambda \bar{\xi}_\lambda + \frac{\Delta}{2} \left[\gamma_\lambda \nabla_\lambda \bar{\xi}_\lambda + \frac{(\bar{\xi}_\lambda - \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right) \\ & + f(t)s\Lambda \sum_\lambda \sum_\Delta \alpha_\lambda^\dagger \beta_{-\lambda+\Delta}^\dagger \frac{g(\Delta)}{\sqrt{1-\gamma_\lambda^2}} \left(\bar{\xi}_\lambda - \gamma_\lambda - \frac{\Delta}{2} \left[\nabla_\lambda \bar{\xi}_\lambda - \frac{(1-\bar{\xi}_\lambda \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right) \\ & + f(t)s\Lambda \sum_\lambda \sum_\Delta \alpha_{\lambda-\Delta} \beta_{-\lambda} \frac{g(\Delta)}{\sqrt{1-\gamma_\lambda^2}} \left(\bar{\xi}_\lambda - \gamma_\lambda - \frac{\Delta}{2} \left[\nabla_\lambda \bar{\xi}_\lambda - \frac{(1-\bar{\xi}_\lambda \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1-\gamma_\lambda^2} \right] \right). \end{aligned} \quad (6.36)$$

In the limit of uniform fields, i.e. $g(\Delta) = \delta_{\Delta,0}$, we recover the expression in (5.14), as required. As always with Fourier transforms, symmetric light profiles centered in the middle of the magnet will yield exclusively real Fourier components $g(\Delta)$, while translation of the light distribution in the real domain causes the Fourier components to pick up a complex part as well. The hermicity of (6.36) is preserved by this translation on account of the properties of the Fourier transform. Still, it seems reasonable to assume that most practical laser pulses will be a symmetric distribution applied to the center of the magnet, so unless otherwise stated I will assume $g(\Delta)$ is real. Another advantage of working with symmetric, centered light profiles is that the Fourier coefficients $g(\Delta)$ in this case are even in Δ . We can exploit this property in (6.19) by swapping the two wave vectors λ and μ in the third and fifth lines. Subsequent use of (6.18) leaves us with (6.36) but with the substitutions

$$\beta_{-\lambda+\Delta}^\dagger \beta_{-\lambda} \rightarrow \beta_{-\lambda}^\dagger \beta_{-\lambda+\Delta}, \quad \alpha_{\lambda-\Delta} \beta_{-\lambda} \rightarrow \alpha_\lambda \beta_{-\lambda+\Delta}. \quad (6.37)$$

The reason I prefer to use these substitutions is simply because they bring more order to the Hamiltonian (6.36) by making sure Δ always occurs on the second operator from the left.

6.1.1 Spin correlations and Néel vector oscillation

The equations we obtained in Chapter 5 for the spin correlations (5.53) and the longitudinal component of the Néel vector (5.54) are valid independently of the form of the perturbation, so we will reuse those two equations here. Our strategy will be to "simply" repeat the steps we made to obtain (5.74). The perturbation Hamiltonian in this case will be more complicated and will require many more calculations to be made because of the off-diagonal terms in the wave vector. As a result I have had to move the calculations to Appendix A. I will only consider the case where $g(\Delta)$ is even as it simplifies the bookkeeping in the calculations tremendously. To see why, we will rewrite (6.19) as

$$\begin{aligned} \delta \mathcal{H}(t) = & -sf(t) (sG + \Lambda g(\mathbf{0})) + f(t)s\Lambda \sum_{\lambda} \delta\omega_{\lambda}^{\lambda} \\ & + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} + \beta_{-\lambda}^{\dagger} \beta_{-\mu} \right) \delta\omega_{\lambda}^{\mu} \\ & + f(t)s\Lambda \sum_{\lambda} \sum_{\mu} \left(\alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger} + \alpha_{\lambda} \beta_{-\mu} \right) V_{\lambda}^{\mu}, \end{aligned} \quad (6.38)$$

where I have defined

$$\delta\omega_{\lambda}^{\mu} = g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda} u_{\mu} + v_{\lambda} v_{\mu}] + \zeta \left(\frac{\Sigma}{2} \right) [u_{\lambda} v_{\mu} + v_{\lambda} u_{\mu}] \right) \quad (6.39)$$

and

$$V_{\lambda}^{\mu} = g(\Delta) \left(\xi \left(\frac{\Delta}{2} \right) [u_{\lambda} v_{\mu} + v_{\lambda} u_{\mu}] + \zeta \left(\frac{\Sigma}{2} \right) [u_{\lambda} u_{\mu} + v_{\lambda} v_{\mu}] \right) \quad (6.40)$$

and exploited the fact that $g(\Delta)$ is even so that I could apply the substitutions (6.37). Since the expressions in (6.39) and (6.40) are invariant with the interchange of wave vectors $\lambda \leftrightarrow \mu$, we do not have to worry about keeping track of where in the expressions two wave vectors enter. This might seem trivial when only two wave vectors are considered, but as seen from the calculations in Appendix A and in the discussions below, it will come in handy when we will be dealing with sums of four different wave vectors simultaneously that each can be any vector in the magnetic Brillouin zone.

As before, we will be working in the interaction picture. The time-evolution operator is given by (5.65) and we will again model the time pulse as $f(t) = \tau\delta(t)$. This yields

$$U = \exp \left[-i\tau \sum_{\lambda} \sum_{\mu} \left\{ \delta\omega_{\lambda}^{\mu} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} + \beta_{-\lambda}^{\dagger} \beta_{-\mu} + \delta_{\lambda,\mu} \right) + V_{\lambda}^{\mu} \left(\alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger} + \alpha_{\lambda} \beta_{-\mu} \right) \right\} \right], \quad (6.41)$$

where I have factored out the irrelevant phase $e^{i\tau s(sG + \Lambda g(\mathbf{0}))}$. We are interested in the expectation values of the 2M operators but, unfortunately, each 2M operator will commute with many off-diagonal terms in (6.41) and therefore it is necessary to keep working with U in its full glory. The expectation values we seek are

$$\langle K_v^{\pm,z}(t) \rangle = \langle \Psi_I(0) | U^{\dagger} K_v^{\pm,z}(t) U | \Psi_I(0) \rangle, \quad (6.42)$$

where $K_v^{\pm z}(t)$ are given by (5.60) and (5.61). By using (5.68) and keeping only the first three terms, we have

$$U^\dagger K_v^{\pm z}(t) U = K_v^{\pm z}(t) + [i\tau\delta\mathcal{H}(0), K_v^{\pm z}(t)] + \frac{1}{2} [i\tau\delta\mathcal{H}(0), [-i\delta\mathcal{H}(0), K_v^{\pm z}(t)]] . \quad (6.43)$$

The calculations of the commutators can be found in Appendix A. After taking the expectation values of (A.1)-(A.20) and collecting like terms we find

$$\langle K_v^+(t) \rangle = e^{2i\omega_v t} \left(2i\tau V_v^v \langle K_v^z \rangle + 2(i\tau)^2 \sum_\lambda \delta\omega_v^\lambda V_v^\lambda \langle K_\lambda^z \rangle \right) . \quad (6.44)$$

Similarly using (A.21)-(A.40), we obtain

$$\langle K_v^-(t) \rangle = -e^{-2i\omega_v t} \left(2i\tau V_v^v \langle K_v^z \rangle - 2(i\tau)^2 \sum_\lambda \delta\omega_v^\lambda V_v^\lambda \langle K_\lambda^z \rangle \right) . \quad (6.45)$$

Finally we can use all 40 equations in Appendix A to get

$$\langle K_v^z(t) \rangle = \langle K_v^z \rangle - (i\tau)^2 \sum_\lambda \left\{ \left(V_v^\lambda \right)^2 [\langle K_\lambda^z \rangle + \langle K_v^z \rangle] + \left(\delta\omega_v^\lambda \right)^2 [\langle K_\lambda^z \rangle - \langle K_v^z \rangle] \right\} . \quad (6.46)$$

We see that, compared to the uniform light profile results of (5.71)-(5.73), the expectation values for the 2M operators are now influenced by the average value of number of magnons in other modes ($\lambda \neq \nu$). Remember: the expectation value of K_λ^z is nonzero for every λ in every state. It should be said that the contribution is likely to be very small apart from the states with ν close to λ , since for realistic light profiles $g(\mathbf{r})$ the only significant Fourier coefficients $g(\Delta)$ are going to be very close to $\Delta = \lambda - \nu = \mathbf{0}$. As an example, consider the light profile in Fig. 6.1a for a square magnet consisting of $N = 200$ spins in each dimension. The profile is an amplitude-normalized, centered Gaussian

$$g(x, y) = \exp \left[-\frac{(x - \frac{N}{2})^2}{2\sigma_x^2} - \frac{(y - \frac{N}{2})^2}{2\sigma_y^2} \right] . \quad (6.47)$$

with standard deviations $\sigma_x = \sigma_y = 10$ spins. Its major Fourier components (also normalized) are plotted in Fig. 6.1b. (Other normalizations, e.g. volume-normalization, of the Gaussian will just scale the colorbars and has no effect on the *relative* amplitudes of the Fourier components). The off-diagonal contributions enter as a second-order effect and both $\delta\omega_v^\lambda$ and V_v^λ are proportional to $g(\Delta)$, as can be seen from

$$\delta\omega_v^\lambda \approx \frac{g(\Delta)}{\sqrt{1 - \gamma_\lambda^2}} \left(1 - \gamma_\lambda \xi_\lambda + \frac{\Delta}{2} \left[\gamma_\lambda \nabla_\lambda \xi_\lambda + \frac{(\xi_\lambda - \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1 - \gamma_\lambda^2} \right] \right) , \quad (6.48)$$

$$V_v^\lambda \approx \frac{g(\Delta)}{\sqrt{1 - \gamma_\lambda^2}} \left(\xi_\lambda - \gamma_\lambda - \frac{\Delta}{2} \left[\nabla_\lambda \xi_\lambda - \frac{(1 - \xi_\lambda \gamma_\lambda) \nabla_\lambda \gamma_\lambda}{1 - \gamma_\lambda^2} \right] \right) , \quad (6.49)$$

where I used the linear expansions in (6.22).

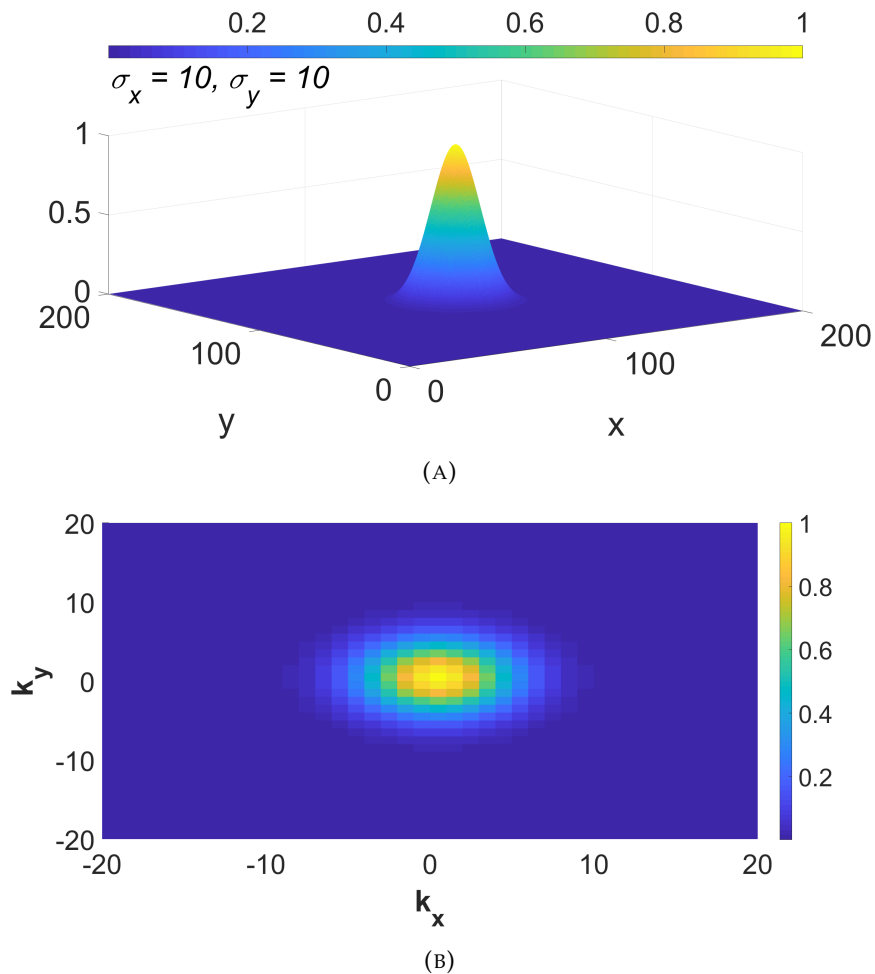


FIGURE 6.1: A) Gaussian light profile for a 200-by-200 square antiferromagnet and B) its fast Fourier transform (absolute values), both with amplitudes normalized. The Fourier transform is purely real since the light profile is symmetric about the center of the magnet, so no information is lost by taking the absolute value.

6.2 The Tyablikov-Bogoliubov transformation

In this section I will attempt to find the energy spectrum and eigenstates of the antiferromagnet Hamiltonian

$$\begin{aligned}
 \mathcal{H} = & -s^2 \sum_j \sum_{\delta} \left[\tilde{J} + \Delta J(\delta) g \left(\mathbf{r}_j + \frac{1}{2} \delta \right) \right] \\
 & + s \sum_j \sum_{\delta} \left\{ \left[\tilde{J} + \Delta J(\delta) g \left(\mathbf{r}_j + \frac{1}{2} \delta \right) \right] a_j^{\dagger} a_j + \left[J + \Delta J(\delta) g \left(\mathbf{r}_j + \frac{1}{2} \delta \right) \right] a_j^{\dagger} b_{j+\delta}^{\dagger} \right\} \\
 & + s \sum_l \sum_{\delta} \left\{ \left[\tilde{J} + \Delta J(\delta) g \left(\mathbf{r}_l + \frac{1}{2} \delta \right) \right] b_l^{\dagger} b_l + \left[J + \Delta J(\delta) g \left(\mathbf{r}_l + \frac{1}{2} \delta \right) \right] a_{l+\delta} b_l \right\},
 \end{aligned} \tag{6.50}$$

where the index j runs over sublattice A, l runs over sublattice B and $\tilde{J} = J - 2K$ as before. I will mostly neglect the constant term, as it only sets the reference energy and does not come into consideration as we seek the states that diagonalize the Hamiltonian. For cleaner notation I will from now on write the strength of the exchange

perturbation as

$$\Delta J(\delta)g\left(\mathbf{r}_j + \frac{1}{2}\delta\right) = \Delta J_{j,\delta} \quad (6.51)$$

and similarly with the interchange $j \leftrightarrow l$. Rather than using the standard Bogoliubov transformation, I will employ a generalized version that is due to Tyablikov and Bogoliubov [44]. The idea is similar to the standard Bogoliubov transformation (4.25) and (4.26) in that we seek new Bose operators that are linear combinations of the operators a_j and $b_{j+\delta}$. However, I will not express the operators as a Fourier series, since we saw in Section 6.1 that the perturbation Hamiltonian $\delta\mathcal{H}$ in Fourier space contains many more off-diagonal elements if the light profile $g(\mathbf{r})$ contains a considerable amount of non-zero Fourier coefficients g_k . The transformation is

$$a_j = \sum_k \left(u_{j,k} \alpha_k + v_{j,k}^* \beta_k^\dagger \right) \quad (6.52)$$

$$b_l = \sum_k \left(y_{l,k} \beta_k + z_{l,k}^* \alpha_k^\dagger \right), \quad (6.53)$$

where α_k, β_k are new Bose operators that satisfy

$$i\partial_t \alpha_k = \omega_k^\alpha \alpha_k, \quad i\partial_t \alpha_k^\dagger = -\omega_k^\alpha \alpha_k^\dagger, \quad (6.54)$$

$$i\partial_t \beta_k = \omega_k^\beta \beta_k, \quad i\partial_t \beta_k^\dagger = -\omega_k^\beta \beta_k^\dagger, \quad (6.55)$$

and $\omega_k^\alpha + \omega_k^\beta$ is an eigenvalue of (6.50) when we neglect the constant term. It is important to stress that k is in general *not* a wave vector; it is an index used to designate a specific eigenstate.

The equations of motion for the untransformed Bose operators a_j and $b_{j+\delta}$ and their adjoints are

$$i\partial_t a_j = [a_j, \mathcal{H}], \quad i\partial_t a_j^\dagger = [a_j^\dagger, \mathcal{H}], \quad i\partial_t b_l = [b_l, \mathcal{H}], \quad i\partial_t b_l^\dagger = [b_l^\dagger, \mathcal{H}]. \quad (6.56)$$

We proceed by calculating the explicit expressions using (6.50) and inserting (6.52)-(6.55) where appropriate. The result is

$$\begin{aligned} \sum_k \left(u_{j,k} \omega_k^\alpha \alpha_k - v_{j,k}^* \omega_k^\beta \beta_k^\dagger \right) &= +s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) u_{j,k} \alpha_k \\ &+ s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) v_{j,k}^* \beta_k^\dagger \\ &+ s \sum_k \sum_\delta \left(J + \Delta J_{j,\delta} \right) y_{j+\delta,k}^* \beta_k^\dagger \\ &+ s \sum_k \sum_\delta \left(J + \Delta J_{j,\delta} \right) z_{j+\delta,k} \alpha_k \end{aligned} \quad (6.57)$$

$$\begin{aligned} \sum_k \left(v_{j,k} \omega_k^\beta \beta_k - u_{j,k}^* \omega_k^\alpha \alpha_k^\dagger \right) &= -s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) u_{j,k}^* \alpha_k^\dagger \\ &- s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) v_{j,k} \beta_k \\ &- s \sum_k \sum_\delta \left(J + \Delta J_{j,\delta} \right) y_{j+\delta,k} \beta_k \\ &- s \sum_k \sum_\delta \left(J + \Delta J_{j,\delta} \right) z_{j+\delta,k}^* \alpha_k^\dagger \end{aligned} \quad (6.58)$$

$$\begin{aligned}
\sum_k \left(y_{l,k} \omega_k^\beta \beta_k - z_{l,k}^* \omega_k^\alpha \alpha_k^\dagger \right) &= +s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) y_{l,k} \beta_k \\
&+ s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) z_{l,k}^* \alpha_k^\dagger \\
&+ s \sum_k \sum_\delta \left(J + \Delta J_{l,\delta} \right) u_{l+\delta,k}^* \alpha_k^\dagger \\
&+ s \sum_k \sum_\delta \left(J + \Delta J_{l,\delta} \right) v_{l+\delta,k} \beta_k
\end{aligned} \tag{6.59}$$

$$\begin{aligned}
\sum_k \left(z_{l,k} \omega_k^\alpha \alpha_k - y_{l,k}^* \omega_k^\beta \beta_k^\dagger \right) &= -s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) y_{l,k}^* \beta_k^\dagger \\
&- s \sum_k \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) z_{l,k} \alpha_k \\
&- s \sum_k \sum_\delta \left(J + \Delta J_{l,\delta} \right) u_{l+\delta,k} \alpha_k \\
&- s \sum_k \sum_\delta \left(J + \Delta J_{l,\delta} \right) v_{l+\delta,k}^* \beta_k^\dagger
\end{aligned} \tag{6.60}$$

By recognizing that $\alpha_k, \beta_k, \alpha_k^\dagger$ and β_k^\dagger are linearly independent, we can exploit the fact that their coefficients in Eqs. (6.57) - (6.60) must match individually (for each k). This yields the following linear equations for the coefficients $u_{j,k}, v_{j,k}, y_{j+\delta,k}$ and $z_{j+\delta,k}$:

$$u_{j,k} \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) u_{j,k} + s \sum_\delta \left(J + \Delta J_{j,\delta} \right) z_{j+\delta,k}, \tag{6.61}$$

$$-z_{l,k}^* \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) z_{l,k}^* + s \sum_\delta \left(J + \Delta J_{l,\delta} \right) u_{l+\delta,k}^*, \tag{6.62}$$

$$y_{l,k} \omega_k^\beta = s \sum_\delta \left(\tilde{J} + \Delta J_{l,\delta} \right) y_{l,k} + s \sum_\delta \left(J + \Delta J_{l,\delta} \right) v_{l+\delta,k}, \tag{6.63}$$

$$-v_{j,k}^* \omega_k^\beta = s \sum_\delta \left(\tilde{J} + \Delta J_{j,\delta} \right) v_{j,k}^* + s \sum_\delta \left(J + \Delta J_{j,\delta} \right) y_{j+\delta,k}^*. \tag{6.64}$$

We see that the system of equations consists of two subsystems: Eqs. (6.61) and (6.62) form an independent system of $2N_{unit}$ linear equations and (6.63), and (6.64) form an *identical* system of equations. This underlines the equivalence between α -magnons and β -magnons. If we add a term to our Hamiltonian in (6.50) that breaks the reflectional symmetry about the xy -plane, for instance an external magnetic field, we would expect the above equations to no longer decouple into two equivalent subsystems. Given that our Hamiltonian splits nicely in two with the generalized α - and β -magnons, we shall simply need to solve for one and immediately have the solution for the other. Eqs. (6.61)-(6.64) can be solved numerically to obtain the energy spectrum and eigenstates of the antiferromagnet for any light profile $g(\mathbf{r})$. They are totally equivalent to the eigenvalue equation involving the Hamiltonian in (6.50).

6.2.1 Proof of diagonalization

We shall first prove that the operators α_k, β_k diagonalize (6.50). Let

$$S = \tilde{J} + \Delta J_{j,\delta}, \quad R = J + \Delta J_{j,\delta}. \tag{6.65}$$

When we insert the definitions (6.52) and (6.53) into (6.50), we get (up to a constant)

$$\begin{aligned}
\mathcal{H} = & \\
& + s \sum_{\lambda} \sum_{\mu} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} \sum_j \sum_{\delta} [S u_{j,\lambda}^* u_{j,\mu} + R u_{j,\lambda}^* z_{j+\delta,\mu}] + \beta_{\lambda}^{\dagger} \beta_{\mu} \sum_l \sum_{\delta} [S y_{l,\lambda}^* y_{l,\mu} + R v_{l+\delta,\lambda}^* y_{l,\mu}] \right) \\
& + s \sum_{\lambda} \sum_{\mu} \left(\alpha_{\lambda}^{\dagger} \beta_{\mu}^{\dagger} \sum_j \sum_{\delta} [S u_{j,\lambda}^* v_{j,\mu}^* + R u_{j,\lambda}^* y_{j+\delta,\mu}^*] + \beta_{\lambda}^{\dagger} \alpha_{\mu}^{\dagger} \sum_l \sum_{\delta} [S z_{l,\mu}^* y_{l,\lambda}^* + R v_{l+\delta,\lambda}^* z_{l,\mu}^*] \right) \\
& + s \sum_{\lambda} \sum_{\mu} \left(\beta_{\lambda} \alpha_{\mu} \sum_j \sum_{\delta} [S v_{j,\lambda} u_{j,\mu} + R v_{j,\lambda} z_{j+\delta,\mu}] + \alpha_{\lambda} \beta_{\mu} \sum_l \sum_{\delta} [S z_{l,\lambda} y_{l,\mu} + R u_{l+\delta,\lambda} y_{l,\mu}] \right) \\
& + s \sum_{\lambda} \sum_{\mu} \left(\beta_{\lambda} \beta_{\mu}^{\dagger} \sum_j \sum_{\delta} [S v_{j,\lambda} v_{j,\mu}^* + R v_{j,\lambda} y_{j+\delta,\mu}^*] + \alpha_{\lambda} \alpha_{\mu}^{\dagger} \sum_l \sum_{\delta} [S z_{l,\lambda} z_{l,\mu}^* + R u_{l+\delta,\lambda} z_{l,\mu}^*] \right)
\end{aligned} \tag{6.66}$$

Using (6.61)-(6.64) and the fact that ω_k , S and R are all real, we get

$$s \sum_j \sum_{\delta} [S u_{j,\lambda}^* u_{j,\mu} + R u_{j,\lambda}^* z_{j+\delta,\mu}] = \sum_j u_{j,\lambda}^* u_{j,\mu} \omega_{\mu}^{\alpha}, \tag{6.67}$$

$$s \sum_l \sum_{\delta} [S y_{l,\lambda}^* y_{l,\mu} + R v_{l+\delta,\lambda}^* y_{l,\mu}] = \sum_l y_{l,\lambda}^* y_{l,\mu} \omega_{\lambda}^{\beta}, \tag{6.68}$$

$$s \sum_j \sum_{\delta} [S z_{l,\mu}^* y_{l,\lambda}^* + R v_{l+\delta,\lambda}^* z_{l,\mu}^*] = \sum_j z_{l,\mu}^* y_{l,\lambda}^* \omega_{\lambda}^{\beta}, \tag{6.69}$$

$$s \sum_j \sum_{\delta} [S v_{j,\lambda} u_{j,\mu} + R v_{j,\lambda} z_{j+\delta,\mu}] = \sum_j v_{j,\lambda} u_{j,\mu} \omega_{\mu}^{\alpha}, \tag{6.70}$$

$$s \sum_j \sum_{\delta} [S z_{l,\lambda} y_{l,\mu} + R u_{l+\delta,\lambda} y_{l,\mu}] = - \sum_l y_{j,\mu} z_{l,\lambda} \omega_{\lambda}^{\alpha}, \tag{6.71}$$

$$s \sum_j \sum_{\delta} [S v_{j,\lambda} v_{j,\mu}^* + R v_{j,\lambda} y_{j+\delta,\mu}^*] = - \sum_j v_{j,\lambda} v_{j,\mu}^* \omega_{\mu}^{\beta}, \tag{6.72}$$

$$s \sum_l \sum_{\delta} [S z_{l,\lambda} z_{l,\mu}^* + R u_{l+\delta,\lambda} z_{l,\mu}^*] = - \sum_l z_{l,\mu}^* z_{l,\lambda} \omega_{\lambda}^{\alpha}, \tag{6.73}$$

$$s \sum_j \sum_{\delta} [S u_{j,\lambda}^* v_{j,\mu}^* + R u_{j,\lambda}^* y_{j+\delta,\mu}^*] = - \sum_j v_{j,\mu}^* u_{j,\lambda}^* \omega_{\mu}^{\beta}. \tag{6.74}$$

This yields the following Hamiltonian:

$$\begin{aligned}
\mathcal{H} = & + \sum_{\lambda} \sum_{\mu} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} \sum_j u_{j,\lambda}^* u_{j,\mu} \omega_{\mu}^{\alpha} + \beta_{\lambda}^{\dagger} \beta_{\mu} \sum_l y_{l,\lambda}^* y_{l,\mu} \omega_{\lambda}^{\beta} \right) \\
& + \sum_{\lambda} \sum_{\mu} \left(-\alpha_{\lambda}^{\dagger} \beta_{\mu}^{\dagger} \sum_j v_{j,\mu}^* u_{j,\lambda}^* \omega_{\mu}^{\beta} + \beta_{\lambda}^{\dagger} \alpha_{\mu}^{\dagger} \sum_l z_{l,\mu}^* y_{l,\lambda}^* \omega_{\lambda}^{\beta} \right) \\
& + \sum_{\lambda} \sum_{\mu} \left(\beta_{\lambda} \alpha_{\mu} \sum_j v_{j,\lambda} u_{j,\mu} \omega_{\mu}^{\alpha} - \alpha_{\lambda} \beta_{\mu} \sum_l y_{l,\mu} z_{l,\lambda} \omega_{\lambda}^{\alpha} \right) \\
& - \sum_{\lambda} \sum_{\mu} \left(\beta_{\lambda} \beta_{\mu}^{\dagger} \sum_j v_{j,\lambda} v_{j,\mu}^* \omega_{\mu}^{\beta} + \alpha_{\lambda} \alpha_{\mu}^{\dagger} \sum_l z_{l,\mu}^* z_{l,\lambda} \omega_{\lambda}^{\alpha} \right).
\end{aligned} \tag{6.75}$$

We can interchange the summation indices $\lambda \leftrightarrow \mu$ in the last line of (6.75) to get

$$\begin{aligned} \mathcal{H} = & + \sum_{\lambda} \sum_{\mu} \omega_{\mu}^{\alpha} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} \sum_j \left[u_{j,\lambda}^* u_{j,\mu} - z_{j+\delta,\lambda}^* z_{j+\delta,\mu} \right] - \sum_j z_{j+\delta,\lambda}^* z_{j+\delta,\mu} \delta_{\lambda,\mu} \right) \\ & + \sum_{\lambda} \sum_{\mu} \omega_{\lambda}^{\beta} \left(\beta_{\lambda}^{\dagger} \beta_{\mu} \sum_l \left[y_{j,\lambda}^* y_{l,\mu} - v_{l+\delta,\lambda}^* v_{l+\delta,\mu} \right] - \sum_l v_{l+\delta,\lambda}^* v_{l+\delta,\mu} \delta_{\lambda,\mu} \right) \\ & + \sum_{\lambda} \sum_{\mu} \alpha_{\lambda}^{\dagger} \beta_{\mu}^{\dagger} \omega_{\mu}^{\beta} \sum_l \left[z_{l,\lambda}^* y_{l,\mu}^* - v_{l+\delta,\mu}^* u_{l+\delta,\lambda}^* \right] \\ & + \sum_{\lambda} \sum_{\mu} \alpha_{\lambda} \beta_{\mu} \omega_{\lambda}^{\alpha} \sum_j \left[v_{j,\mu} u_{j,\lambda} - y_{j+\delta,\mu} z_{j+\delta,\lambda} \right], \end{aligned} \quad (6.76)$$

where I used the commutators for the new Bose operators. Since the system of equations (6.63) + (6.64) is identical to (6.61) + (6.62), we must have $\omega_k^{\alpha} = \omega_k^{\beta} = \omega_k$. Using (6.61)-(6.64), we obtain

$$\begin{aligned} \omega_{\lambda} \sum_j (u_{j,\lambda} v_{j,\mu} - y_{j+\delta,\mu} z_{j+\delta,\lambda}) &= s \sum_j \sum_{\delta} (S u_{j,\lambda} v_{j,\mu} + R v_{j,\mu} z_{j+\delta,\lambda}) \\ &+ s \sum_l \sum_{\delta} (S y_{l,\mu} z_{l,\lambda} + R y_{l,\mu} u_{l+\delta,\lambda}), \end{aligned} \quad (6.77)$$

$$\begin{aligned} -\omega_{\mu} \sum_j (u_{j,\lambda} v_{j,\mu} - y_{j+\delta,\mu} z_{j+\delta,\lambda}) &= s \sum_j \sum_{\delta} (S u_{j,\lambda} v_{j,\mu} + R u_{j,\lambda} y_{j+\delta,\mu}) \\ &+ s \sum_l \sum_{\delta} (S y_{l,\mu} z_{l,\lambda} + R z_{l,\lambda} v_{l+\delta,\mu}). \end{aligned} \quad (6.78)$$

But

$$\sum_j \sum_{\delta} R v_{j,\mu} z_{j+\delta,\lambda} = \sum_l \sum_{\delta} R v_{l+\delta,\mu} z_{l,\lambda}, \quad \sum_l \sum_{\delta} R y_{l,\mu} u_{l+\delta,\lambda} = \sum_j \sum_{\delta} R y_{j+\delta,\mu} u_{j,\lambda}, \quad (6.79)$$

so when we subtract (6.78) from (6.77) we obtain

$$(\omega_{\lambda} + \omega_{\mu}) \sum_j (u_{j,\lambda} v_{j,\mu} - y_{j+\delta,\mu} z_{j+\delta,\lambda}) = 0 \implies \sum_j (u_{j,\lambda} v_{j,\mu} - y_{j+\delta,\mu} z_{j+\delta,\lambda}) = 0. \quad (6.80)$$

Hence, the off-diagonal terms in (6.76) vanish:

$$\begin{aligned} \mathcal{H} = & + \sum_{\lambda} \sum_{\mu} \omega_{\mu}^{\alpha} \left(\alpha_{\lambda}^{\dagger} \alpha_{\mu} \sum_j \left[u_{j,\lambda}^* u_{j,\mu} - z_{j+\delta,\lambda}^* z_{j+\delta,\mu} \right] - \sum_j z_{j+\delta,\lambda}^* z_{j+\delta,\mu} \delta_{\lambda,\mu} \right) \\ & + \sum_{\lambda} \sum_{\mu} \omega_{\lambda}^{\beta} \left(\beta_{\lambda}^{\dagger} \beta_{\mu} \sum_l \left[y_{j,\lambda}^* y_{l,\mu} - v_{l+\delta,\lambda}^* v_{l+\delta,\mu} \right] - \sum_l v_{l+\delta,\lambda}^* v_{l+\delta,\mu} \delta_{\lambda,\mu} \right). \end{aligned} \quad (6.81)$$

All that remains is to show that $\sum_j (u_{j,\lambda}^* u_{j,\mu} - z_{j+\delta,\lambda}^* z_{j+\delta,\mu}) \propto \delta_{\lambda,\mu}$. From the inverse relations

$$\alpha_k = \sum_j \left(u_{j,k}^* a_j - z_{j+\delta,k}^* b_{j+\delta}^{\dagger} \right), \quad (6.82)$$

$$\beta_k = \sum_l \left(y_{j,k}^* b_l - v_{l+\delta,k}^* a_{l+\delta}^{\dagger} \right) \quad (6.83)$$

we find that

$$\sum_j \left(u_{j,\lambda}^* u_{j,\mu} - z_{j+\delta,\lambda}^* z_{j+\delta,\mu} \right) = \left[\alpha_{\lambda}, \alpha_{\mu}^{\dagger} \right] = \delta_{\lambda,\mu}, \quad (6.84)$$

$$\sum_l \left[y_{j,\lambda}^* y_{l,\mu} - v_{l+\delta,\lambda}^* v_{l+\delta,\mu} \right] = \left[\beta_\lambda, \beta_\mu^\dagger \right] = \delta_{\lambda,\mu}. \quad (6.85)$$

Therefore, the diagonal Hamiltonian is

$$\mathcal{H} = - \sum_k Q \left(\omega_k^\alpha + \omega_k^\beta \right) + \sum_k \left(\omega_k^\alpha \alpha_k^\dagger \alpha_k + \omega_k^\beta \beta_k^\dagger \beta_k \right), \quad (6.86)$$

where

$$Q = \sum_j |v_{j,k}|^2. \quad (6.87)$$

6.2.2 Tyablikov-Bogoliubov transformation with uniform electric field

As noted before, when dealing with arbitrary light profiles $g(\mathbf{r})$, Eqs. (6.61) and (6.62) will yield a system of $2N_{unit}$ equations for the coefficients $u_{j,k}$ and $z_{j,k}$ that have to be solved simultaneously. However, if the light profile is uniform, then Eqs. (6.61) and (6.62) become

$$u_{j,k} \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) u_{j,k} + s \sum_\delta \left(J + \Delta J(\delta) \right) z_{j+\delta,k}, \quad (6.88)$$

$$-z_{l,k}^* \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) z_{l,k}^* + s \sum_\delta \left(J + \Delta J(\delta) \right) u_{l+\delta,k}^*, \quad (6.89)$$

and we see that $u_{j,k}$ and $z_{j,k}$ must satisfy the same equation for every j, l . This effectively reduces the number of equations from $2N_{unit}$ to 2:

$$u_k \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) u_k + s \sum_\delta \left(J + \Delta J(\delta) \right) z_k, \quad (6.90)$$

$$-z_k^* \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) z_k^* + s \sum_\delta \left(J + \Delta J(\delta) \right) u_k^*. \quad (6.91)$$

Specifically, we will be looking for plane wave solutions:

$$u_{j,k} = \frac{u_k}{\sqrt{N_{unit}}} \exp [i \mathbf{k} \cdot \mathbf{r}_j], \quad z_{l,k} = \frac{z_k}{\sqrt{N_{unit}}} \exp [i \mathbf{k} \cdot \mathbf{r}_l], \quad (6.92)$$

where \mathbf{k} is a reciprocal lattice vector in the magnetic Brillouin zone. This yields the following equations for the (real) wave amplitudes u_k and z_k :

$$u_k \omega_k^\alpha = s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) u_k + s \sum_\delta \left(J + \Delta J(\delta) \right) \exp [i \mathbf{k} \cdot \delta] z_k \quad (6.93)$$

$$z_k \omega_k^\alpha = -s \sum_\delta \left(\tilde{J} + \Delta J(\delta) \right) z_k - s \sum_\delta \left(J + \Delta J(\delta) \right) \exp [i \mathbf{k} \cdot \delta] u_k. \quad (6.94)$$

The solution is subject to the normalization constraint (6.84), which becomes

$$|u_k|^2 - |z_k|^2 = 1, \quad (6.95)$$

just as in the normal Bogoliubov transformation. The solution can be easily obtained analytically. We find

$$\omega_k^\alpha = s \sqrt{\left(z_N \tilde{J} + \Lambda \right)^2 - \left(z_N J \gamma_k + \Lambda \xi_k \right)^2}, \quad (6.96)$$

$$u_k^2 = \frac{1}{2} \left(1 + \frac{z_N \tilde{J}s + \Lambda s}{\omega_k^\alpha} \right), \quad (6.97)$$

$$z_k^2 = \frac{1}{2} \left(\frac{z_N \tilde{J}s + \Lambda s}{\omega_k^\alpha} - 1 \right). \quad (6.98)$$

Of course, the energy of the β -magnons is also given by (6.96), and the equations for y_k^2 and v_k^2 are identical to (6.97) and (6.98), respectively.

The numerical solutions for $u_{j,k}$ in the ten lowest modes given by the system the system (6.88) and (6.89) for a 200-by-200 square antiferromagnet of the type shown in Fig. 6.2a are plotted in Fig. 6.3. Plotting $u_{j,k}$ is a challenge because sublattice A on which $u_{j,k}$ is defined is not square. To get around the problem, I have used the average of $u_{j,k}$ on the nearest neighbors to plot $u_{j,k}$ on sites of sublattice B, illustrated in Fig. 6.2b. It should be stressed that this averaging is only done to make the plots in Fig. 6.3 look smoother. I have included a small amount of anisotropy ($K = -10^{-4}J$) to ensure that the ground state is well behaved. The light is polarized so that $\Delta J(\delta)$ is independent of the direction δ and the perturbation is set to 5 % of the the unperturbed exchange J . The solutions were found using MATLAB's built-in function for finding eigenvalues and eigenvectors. Unlike our analytical solution for an infinitely large magnet, the solution obtained numerically for finite magnets yields real $u_{j,k}$ and $z_{l,k}$. The energies ω are also lower than the analytical solution, which can attributed to edge effects in the numerical solution; spins on the edges are coupled to fewer neighboring spins, which lowers the energy of excitation. As the size of the magnet increases, so does the spectrum $\omega(k)$. I am unable to verify convergence of the spectra toward the analytical result because the computational power available to me only allows magnets up to on the order of 10 000 spins to be solved, even if only the lowest eigenstates are sought. However, for a 2-by-2 magnet the analytical and numerical solutions are identical because each spin in that case has equally many nearest neighbors (two). The amplitudes $u_{j,k}$ and $z_{l,k}$ decrease with the size of the magnet because of the normalization condition (6.84).

Increasing the anisotropy in the magnet has the effect of flattening the distribution $u_{j,k}$ while increasing the amplitudes in opposing corners the magnet, as illustrated in

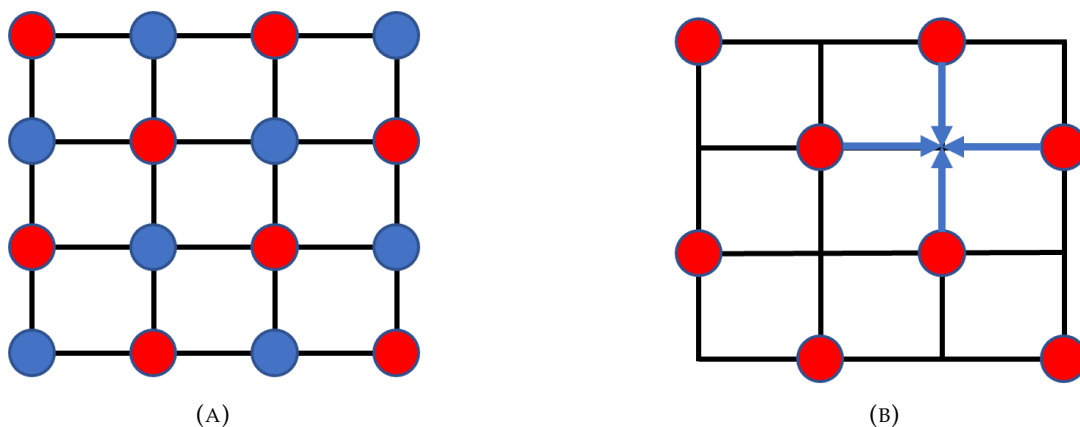


FIGURE 6.2: Illustration of A) a bipartite square antiferromagnet and B) how I obtain average values of $u_{j,k}$ on sites where it is not defined.

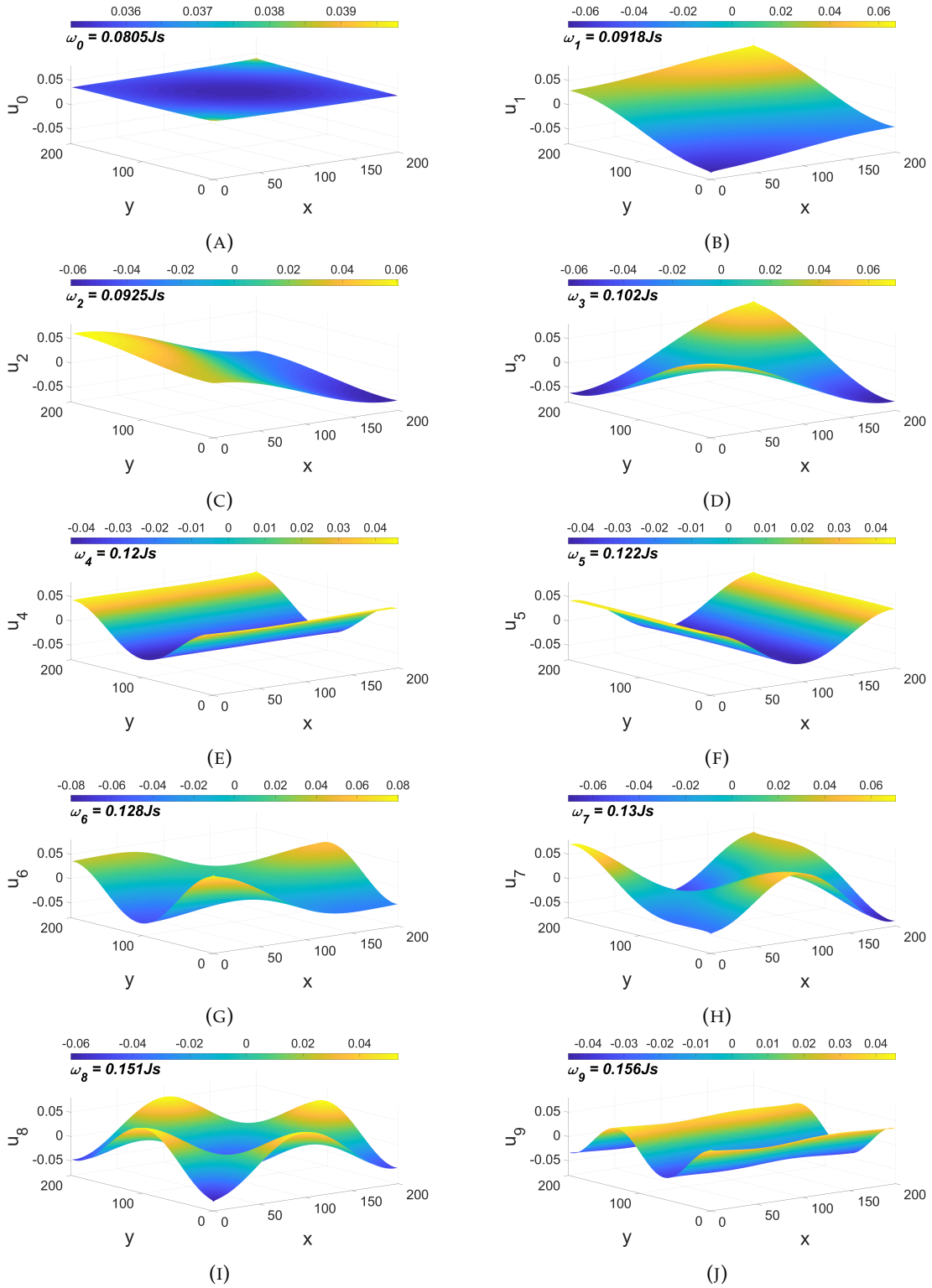


FIGURE 6.3: Surface plots of $u_{j,k}$ for the ten lowest values energies ω_k^α with $K = 10^{-4}J$, $\Delta J(\delta) = 0.05J \hat{x} \cdot \delta$ for a 200-by-200 square antiferromagnet.

Fig. 6.4. To explain this effect we will look at the average of S_j^z in a 2M state. We have

$$\begin{aligned}
 \langle S_j^z \rangle &= \sum_k \langle n_k | s | n_k \rangle - \sum_k \langle n_k | a_j^\dagger a_j | n_k \rangle \\
 &= s \sum_k 1 - \sum_k |u_{j,k}|^2 n_k - \sum_k |v_{j,k}|^2 (n_k + 1),
 \end{aligned} \tag{6.99}$$

where I used (6.52) and (6.20) and n_k is the number of magnons in the 2M state specified by the number k . We see that small values of $u_{j,k}$ and $v_{j,k}$ are associated with the spin on site \mathbf{r}_j being closely aligned with the z-axis. This is favored by the strong anisotropy. The exchange interaction favors all neighboring spins to be as close to parallel as possible, which is achieved by forming a smooth wave as in Fig. 6.4a. The profile in Fig. 6.4b is therefore a "compromise" between the the anisotropy and the exchange given that $u_{j,k}$ and $v_{j,k}$ cannot be zero everywhere: most of the deflection from the z-axis is put on the spins with the fewest nearest neighbors in the corners where the cost to the exchange is smallest, and then the spins rapidly align with the z-axis as you move inward towards the center of the magnet. I should stress that $v_{j,k}$ has a similar profile to $u_{j,k}$ but with smaller amplitude, and therefore does not affect the reasoning above. The anisotropy's effect on the profile of $u_{j,k}$ is not present in the analytical solution because it does not take into account that there are edges or corners.

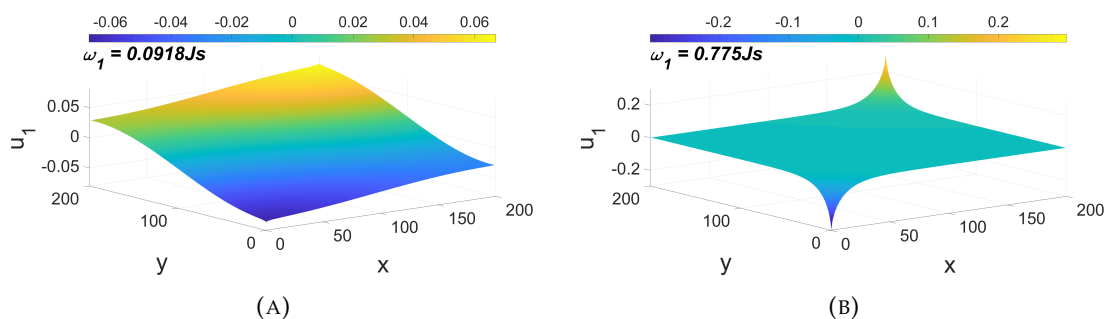


FIGURE 6.4: The first excited state of magnet with A) $K = 10^{-4}J$ and B) $0.01J$. The light is polarized so that ΔJ is $0.05J$ in the x-direction.

6.3 The sudden approximation for nonuniform electric fields

In Section 6.1 we studied the effect of the introduction of a non-uniform field to the Hamiltonian of our antiferromagnet in Fourier space. In this section we will try to find an estimate for the effect of nonuniform excitation fields using the sudden approximation technique. The idea is to model the laser pulse in time as a Heaviside function:

$$f(t) = \Theta(t) - \Theta(t - \tau), \quad (6.100)$$

where τ is the duration of the laser pulse (as before). We will then diagonalize the Hamiltonian (6.50) using the transformation of the previous section so that we can find its eigenstates. For a magnet that starts out in the ground state $|0\rangle$ of the unperturbed Hamiltonian, we will be able to calculate the transition probabilities to each of the eigenstates of (6.50) by calculating their inner products with $|0\rangle$. Specifically, we have

$$|\Psi(\tau)\rangle = \sum_j \langle \phi_j | 0 \rangle |\phi_j\rangle e^{-i\omega_{\phi_j}\tau} \quad (6.101)$$

where $|\phi_j\rangle$ is an eigenstate of (6.50) with eigenvalue ω_{ϕ_j} . After the perturbation is switched off instantly at $t = \tau$, we find that the (unnormalized) state of the magnet is

$$|\Psi(t > \tau)\rangle = \sum_h \langle \psi_h | \Psi(\tau) \rangle |\psi_h\rangle e^{-i\omega_{\psi_h}(t-\tau)}, \quad (6.102)$$

where $|\psi_h\rangle$ is an eigenstate of the unperturbed Hamiltonian. Inserting (6.101) into (6.102), we get

$$|\Psi(t > \tau)\rangle = \sum_h \sum_j \langle \psi_h | \phi_j \rangle \langle \phi_j | 0 \rangle |\psi_h\rangle \exp \left[-i \left\{ \omega_{\psi_h} t + \left(\omega_{\phi_j} - \omega_{\psi_h} \right) \tau \right\} \right]. \quad (6.103)$$

Obviously, for this method to work we need to be able to calculate inner products between states with relative ease. Because every state can be reached from the ground state by applying the appropriate magnon raising operators, this amounts to calculating matrix elements of the form

$$\psi \langle 0 | A | 0 \rangle_\phi, \quad (6.104)$$

where A is some product of annihilation and creation operators, $|0\rangle_\psi = |0\rangle$ is the ground state of the unperturbed Hamiltonian and $|0\rangle_\phi$ is the ground state of the perturbed Hamiltonian.

It turns out that the evaluation of matrix elements of the form (6.104) is not trivial, and so I have had to spend much more time exploring the machinery for evaluating them than I had originally planned. I will therefore only present a strategy for how the matrix elements can be evaluated and discuss briefly what the challenges are and what it will take to overcome them.

Before we start looking for ways to evaluate the matrix elements, we should make life a bit easier for ourselves by writing a Tyablikov-Bogoliubov transformation in a more compact form. For Bose operators $a_j, a_j^\dagger, b_l, b_l^\dagger$, we have

$$\begin{bmatrix} \mathbf{a} \\ \mathbf{b} \\ \mathbf{a}^\dagger \\ \mathbf{b}^\dagger \end{bmatrix} = \begin{bmatrix} U & 0 & 0 & V^* \\ 0 & Y & Z^* & 0 \\ 0 & V & U^* & 0 \\ Z & 0 & 0 & Y^* \end{bmatrix} \begin{bmatrix} \boldsymbol{\alpha} \\ \boldsymbol{\beta} \\ \boldsymbol{\alpha}^\dagger \\ \boldsymbol{\beta}^\dagger \end{bmatrix}, \quad (6.105)$$

where the vectors contain a sequence of the operators so that, for instance, $\mathbf{a} = a_1, a_2, \dots, a_N$, and the matrices U, Y, V, Z contain the Tyablikov-Bogoliubov coefficients. We may write the transformation even more compactly by merging the two sequences \mathbf{a} and \mathbf{b} into the sequence \mathbf{x} , so that

$$\begin{bmatrix} \mathbf{x} \\ \mathbf{x}^\dagger \end{bmatrix} = \begin{bmatrix} O & P^* \\ P & O^* \end{bmatrix} \begin{bmatrix} \boldsymbol{\chi} \\ \boldsymbol{\chi}^\dagger \end{bmatrix} = B \begin{bmatrix} \boldsymbol{\chi} \\ \boldsymbol{\chi}^\dagger \end{bmatrix}, \quad (6.106)$$

with

$$O = \begin{bmatrix} U & 0 \\ 0 & Y \end{bmatrix}, \quad P = \begin{bmatrix} 0 & V \\ Z & 0 \end{bmatrix}. \quad (6.107)$$

The $2N \times 2N$ matrix B is obviously the transformation on the operators. However, we are going to need the transformation on the states as well. There exists a theorem due to Thouless [45] which gives a way to express any state the vacuum state $|0\rangle_\psi$ when the states are Slater determinants. The theorem can be recast for bosons, and the version we will use is given by [46]:

$$|0\rangle_\psi = {}_\phi \langle 0 | 0 \rangle_\psi \exp \left[\frac{1}{2} \sum_k \sum_l M_{kl} \phi_k^\dagger \phi_l^\dagger \right] |0\rangle_\phi, \quad (6.108)$$

where, for two different Tyablikov-Bogoliubov transformations

$$\begin{bmatrix} \mathbf{x} \\ \mathbf{x}^\dagger \end{bmatrix} = \begin{bmatrix} O & P^* \\ P & O^* \end{bmatrix} \begin{bmatrix} \boldsymbol{\phi} \\ \boldsymbol{\phi}^\dagger \end{bmatrix} = \begin{bmatrix} W & X^* \\ X & W^* \end{bmatrix} \begin{bmatrix} \boldsymbol{\psi} \\ \boldsymbol{\psi}^\dagger \end{bmatrix}, \quad (6.109)$$

the matrix $M = Z^T$ is given by

$$M = \left(\left[O^T X - P^T W \right] \left[O^+ W - P^+ X \right]^{-1} \right)^*. \quad (6.110)$$

The overlap integral between the two vacuum states is

$$\phi \langle 0|0 \rangle_\psi = \exp \left[-\frac{1}{2} \text{Tr} \log_e \left(O^+ W - P^+ X \right) \right]. \quad (6.111)$$

With the expression (6.108) relating the vacuum states of two different Bose operators, we can in principle evaluate inner products of the form $\langle \phi_j | 0 \rangle$. Any eigenstate $|\phi_j\rangle$ of the full Hamiltonian (6.50) can be reached by application of a product of raising operators ϕ^\dagger to the vacuum state $|0\rangle_\phi$. Hence,

$$\prod_j \langle n_j^\phi | 0 \rangle = \phi \langle 0|0 \rangle_\psi \phi \langle 0| \prod_j \phi_j \exp \left[\frac{1}{2} \sum_k \sum_l M_{kl} \phi_k^\dagger \phi_l^\dagger \right] |0\rangle_\phi, \quad (6.112)$$

where n_j^ϕ is the number of ϕ -magnons in mode j . First of all we notice that odd-numbered magnon states have zero overlap with the ground state, so the laser pulse cannot excite odd numbers of magnons. Second, if we look closer at the matrix Z we will find that the overlap is nonzero only for states $\prod_j |n_j\rangle$ that contain an equal amount of magnons from the α -branch and β -branch. To see this, we can write

$$W = \begin{bmatrix} U' & 0 \\ 0 & Y' \end{bmatrix}, \quad X = \begin{bmatrix} 0 & V' \\ Z' & 0 \end{bmatrix}. \quad (6.113)$$

The matrix products in (6.110) are then easily found to be

$$O^T X - P^T W = \begin{bmatrix} 0 & U^T V' - Z^T Y' \\ Y^T Z' - V^T U' & 0 \end{bmatrix}, \quad (6.114)$$

$$O^+ W - P^+ X = \begin{bmatrix} U^+ U' - Z^+ Z' & 0 \\ 0 & Y^+ Y' - V^+ V' \end{bmatrix}. \quad (6.115)$$

Assuming $U^+ U' - Z^+ Z'$ and $Y^+ Y' - V^+ V'$ are invertible, we have [47]

$$\left(O^+ W - P^+ X \right)^{-1} = \begin{bmatrix} (U^+ U' - Z^+ Z')^{-1} & 0 \\ 0 & (Y^+ Y' - V^+ V')^{-1} \end{bmatrix}. \quad (6.116)$$

Thus,

$$M = \begin{bmatrix} 0 & (U^T V' - Z^T Y') (Y^+ Y' - V^+ V')^{-1} \\ (Y^T Z' - V^T U') (U^+ U' - Z^+ Z')^{-1} & 0 \end{bmatrix}^*. \quad (6.117)$$

The first $\frac{N}{2}$ elements of $\boldsymbol{\phi}$ belong to the α -branch and the remaining $\frac{N}{2}$ elements belong to the β -branch. So if $k \leq \frac{N}{2}$ so that ϕ_k is an α -magnon, we see that the nonzero

elements on the k -th row of M are all in columns with $l > \frac{N}{2}$, and ϕ_l in this case is a β -magnon.

Suppose we have a state with two magnons (one from each branch). Then,

$$\begin{aligned} \phi \langle 0 | \phi_i \phi_j \exp \left[\frac{1}{2} \sum_k \sum_l M_{kl} \phi_k^\dagger \phi_l^\dagger \right] | 0 \rangle_\phi &= \frac{1}{2} \sum_k \sum_l M_{kl} (\delta_{k,i} \delta_{l,j} + \delta_{k,j} \delta_{l,i}) \\ &= M_{j,i}, \end{aligned} \quad (6.118)$$

where the first equality follows from the Bose commutators and the last equality follows from the fact that M is symmetric. So the overlap with the ground state $|0\rangle_\psi$ is simply given by the matrix element $M_{j,i}$ times the overlap between the vacuum states given by (6.111). For states with four magnons (two from each branch) or more, we are required to do more complicated calculations. In general, a state with n magnon pairs will require calculation of

$$\phi \langle 0 | \prod_{j=1}^{j_{2n}} \phi_j \sum_{k_1=0}^N \cdots \sum_{k_n=0}^N \frac{1}{n!} \left(\frac{1}{2} \right)^n M_{k_1, k_2} M_{k_3, k_4} \cdots M_{k_{2n-1}, k_{2n}} \phi_{k_1}^\dagger \phi_{k_2}^\dagger \cdots \phi_{k_{2n}}^\dagger | 0 \rangle_\phi, \quad (6.119)$$

where the factor $\frac{1}{n!} \left(\frac{1}{2} \right)^n$ is from the exponential. There exists a theorem, known as Wick's theorem, that can be used to calculate such matrix elements by reordering the sequence of the operators ϕ, ϕ^\dagger to normal order, which means that all creation operators are to the left of all annihilation operators. I do not have the time to start evaluating the inner products of the form (6.119), but I will make some general remarks.

Straight away we notice that the factor $\frac{1}{n!} \left(\frac{1}{2} \right)^n$ will become very small when n grows large. However, according to Wick's theorem,

$$\phi \langle 0 | \phi_{j_1} \cdots \phi_{j_{2n}} \phi_{k_1}^\dagger \cdots \phi_{k_{2n}}^\dagger | 0 \rangle_\phi = \sum_{perm.} \delta_{k_1, j_j} \delta_{k_2, j_2} \cdots \delta_{k_{2n}, j_{2n}}, \quad (6.120)$$

where the sum runs over all permutations of the indices k and j (assuming each operator ϕ_j enters only once). From what I can tell this amounts to $2n!$ permutations, so states with many magnons need not have vanishingly small overlap with $|0\rangle_\phi$. We therefore have to calculate the overlap between $|0\rangle_\phi$ and every other state in Hilbert space, which on its face seems like a very large problem. We can write this problem compactly as

$$\phi \langle 1_{j_1} \cdots 1_{j_{2n}} 0_{j_{2n+1}} \cdots 0_{j_N} | 0 \rangle_\phi = \frac{1}{n! 2^n} \sum_{perm.} M_{j_1, j_2} \cdots M_{j_{2n-1}, j_{2n}}, \quad (6.121)$$

where the sum runs over all permutation of the indices j , which specify which magnons are excited in the state whose overlap with $|0\rangle_\phi$ we seek. For instance, if we have a state $|1_a 1_b 1_c 1_d\rangle$ with two magnon pairs (four magnons in total), the overlap is

$$\begin{aligned} \langle 1_a 1_b 1_c 1_d | 0 \rangle_\phi &= \frac{1}{8} (M_{ab} M_{cd} + M_{ab} M_{dc} + M_{ac} M_{bd} + M_{ac} M_{db} + M_{ad} M_{bc} + M_{ad} M_{cb} \\ &\quad + M_{ba} M_{cd} + M_{ba} M_{dc} + M_{bc} M_{ad} + M_{bc} M_{da} + M_{bd} M_{ac} + M_{bd} M_{ca} \\ &\quad + M_{ca} M_{bd} + M_{ca} M_{db} + M_{cb} M_{ad} + M_{cb} M_{da} + M_{cd} M_{ab} + M_{cd} M_{ba} \\ &\quad + M_{da} M_{bc} + M_{da} M_{cb} + M_{db} M_{ac} + M_{db} M_{ca} + M_{dc} M_{ab} + M_{dc} M_{ba}). \end{aligned} \quad (6.122)$$

Although each term is just a product of matrix elements and is calculated in a fraction of a millisecond on any computer, the fact that there are so many ($2n!$) of them makes this approach rather useless. The only thing I can think of that would save this approach is if we could prove that the highly occupied states have negligible overlap in some other way, but I have not found one.

The calculation of matrix elements of the form $\langle \psi_h | \phi_j \rangle = \langle \phi_j | \psi_h \rangle^*$ follows immediately from (6.108):

$$\langle \phi_j | \psi_h \rangle = {}_\phi \langle 0 | 0 \rangle_\psi {}_\phi \langle 0 | \prod_{j=j_1}^{j_{2n}} \prod_{h=h_1}^{h_{2m}} \phi_j \psi_h^\dagger \exp \left[\frac{1}{2} \sum_k \sum_l M_{kl} \phi_k^\dagger \phi_l^\dagger \right] | 0 \rangle_\phi, \quad (6.123)$$

where m is the number of ψ -magnon pairs. This is essentially the same problem as the previous overlap, but the magnons ψ_h have to be expressed in terms of the magnons ϕ_j by making the transformation

$$\begin{bmatrix} \psi \\ \psi^\dagger \end{bmatrix} = \begin{bmatrix} W & X^* \\ X & W^* \end{bmatrix}^{-1} \begin{bmatrix} O & P^* \\ P & O^* \end{bmatrix} \begin{bmatrix} \phi \\ \phi^\dagger \end{bmatrix}. \quad (6.124)$$

However, the problem is even more complex because of the number of terms in the transformation (6.124). Again, I think that unless it can be shown that the highly occupied states have very negligible overlap with states near the ground state, these calculations become too large for any reasonably sized magnet.

Chapter 7

Summary, Outlook & Some Reflections

In this thesis I have studied quantum antiferromagnets using spin wave theory. I began with the basics, building on little more than a fundamental understanding of quantum mechanics usually acquired in a one-semester undergraduate course. From there I started building up the theory of bosonic spin waves piece by piece. In Chapter 4 I outlined the approach for studying static antiferromagnets, i.e. how the excitation and annihilation of spin waves can be combined so that the time-independent Heisenberg Hamiltonian becomes diagonal, from which the eigenstates and energy eigenvalues are easily obtained. This was the first encounter with the Bogoliubov transformation, which plays a central role in the subsequent chapters. I also briefly discussed the stability of antiferromagnetic order, showed how magnetic anisotropy increases this stability and how the effect of easy-axis anisotropy can be included in the calculations with minimal difficulty. This triviality was used as justification for ignoring anisotropy and instability in the following discussion. The purpose of Chapter 4 was to introduce the techniques that are used in conventional spin wave theory, which are also used for the more interesting case of antiferromagnetic spin dynamics.

Chapter 5 was devoted to the study of these dynamics launched by interaction between the magnet and electromagnetic waves. I showed how the electric field triggers dynamics as a result of a perturbation of the exchange interactions, greatly elaborating on the discussion and theory section of a recently published article on the subject [18]. A series of important concepts were developed in that chapter, including a link between the physics of spin and the mathematical concept of groups. While the spin correlations and Néel vector dynamics were described well without the results of the group theory discussion, it turned out that knowledge of the $SU(1,1)$ group would allow us to study the state of the magnet after the excitation because the states were Perelomov coherent states of this group. Despite some confusion about what is meant by "coherent" in this regard, I showed how these states were fully described by a single parameter and I outlined how this parameter connects the action of the perturbations on the quantum state to the rotation of a vector on the upper sheet of a hyperboloid. As well as providing an elegant description of the magnetic quantum states, the coherent state parameter can be treated semiclassically to capture the quantum femtosecond dynamics that complement classical dynamics, which are described by the Landau-Lifshitz equations (Chapter 3) and characterized by considerably longer time scales.

In Chapter 6 I attempted to look at the implications of using spatially nonuniform excitation fields, which from what I can tell has not been done before. As a start I continued with the Fourier and Bogoliubov transformations that had been so effective in dealing with uniform excitation fields, following the same steps as in Chapter

5 but with a factor in the perturbation part of the Hamiltonian that captured the spatial profile of the exciting electric field. While this approach was tangible, the loss of translational invariance made the Fourier transform less suited to reduce the complexity in the calculations. The correction to the Néel vector dynamics caused by the nonuniformity of the exciting electric field was calculated and found to be of second order in the perturbation strength and duration. In an effort to reduce the complexity of the calculations, I studied the generalization of the Bogoliubov transformation described by Tyablikov [44], which can diagonalize any quadratic form such as the Hamiltonians in this thesis. I proved this for the Heisenberg Hamiltonian with a constant exchange perturbation with respect to time, and I did so in a way that illustrates the separation and independence of the two branches of the magnon spectrum.

After having written computer code that could diagonalize the Heisenberg Hamiltonian with or without the exchange perturbation, I started developing the machinery for finding the quantum state of the magnet following a sudden pulse of excitation in the final part of Chapter 6. I showed how the problem reduces to an evaluation of inner products and I outlined how those inner products can be evaluated using only the results from the numerical diagonalization. The discussion was cut short by my running out of time before the thesis submission deadline, so I ended with some thoughts on what it would take to make this approach viable. I left the discussion on a rather pessimistic note, though I am not certain that obstacles I see are as difficult to overcome as they appear as of this writing.

Going forward, I think it will be worthwhile to consider whether the evaluation of inner products can be made simple enough to make the sudden approximation a realistic approach. Its strength lies in its conceptual simplicity and ability to handle a wide range of perturbations for relatively large magnets; most current laptops would have little trouble handling magnets of size roughly 100-by-100 spins. It could turn out that the number of approximations needed to make this approach work is simply too great. If so, then at least the knowledge of *why* it does not work will guide the next attempt at studying the problem.

Having worked on this project for quite some time, it has become increasingly clear to me that there are shortcomings to all approaches, and that only by attacking the problem from multiple angles can a proper understanding be built. The main thing I take away from my thesis – apart from how to use all the techniques presented in the previous chapters – is that I now have a much better understanding of the strengths and weaknesses of some of the tools used in the study of quantum magnets. This is important because it enables me to appreciate what makes a problem difficult, and only by understanding the problem can I really hope to find a solution. Most of this thesis was written before I had my current level of understanding, and often have I felt that I was taking a shot in the dark, especially when the literature could no longer guide me. But every missed attempt at a solution has grown my understanding of the problem, like a lumberjack who learns where to strike a tree to bring it down. So the next time I swing my axe, I will have a better idea of where I should strike. And as we all know: a tree does not fall with the first swing. "Eigi fellr tré við fyrsta högg".

Appendix A

Spin correlations and Néel vector dynamics calculations

The following commutators were evaluated to arrive at the results in Section 6.1.1.

A.1 Commutators involving the 2M raising operator

A.1.1 First order

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, K_v^+ \right] = \sum_{\lambda} \delta\omega_{\lambda}^v \alpha_{\lambda}^{\dagger} \beta_{-\nu}^{\dagger}. \quad (\text{A.1})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, K_v^+ \right] = \sum_{\lambda} \delta\omega_{\lambda}^v \alpha_{\nu}^{\dagger} \beta_{-\lambda}^{\dagger}. \quad (\text{A.2})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, K_v^+ \right] = 0. \quad (\text{A.3})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, K_v^+ \right] = \sum_{\lambda} V_{\nu}^{\lambda} \left(\alpha_{\nu}^{\dagger} \alpha_{\lambda} + \beta_{-\nu}^{\dagger} \beta_{-\lambda} + \delta_{\lambda, \nu} \right). \quad (\text{A.4})$$

A.1.2 Second order

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.5})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.6})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} - \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}. \quad (\text{A.7})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = 0. \quad (\text{A.8})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\mu}^{\dagger} \beta_{-\lambda}^{\dagger}. \quad (\text{A.9})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\mu}^{\dagger} \beta_{-\lambda}^{\dagger}. \quad (\text{A.10})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = 0. \quad (\text{A.11})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} V_{\mu}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu} - \sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} V_{\mu}^{\lambda} \beta_{-\mu}^{\dagger} \beta_{-\lambda}. \quad (\text{A.12})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = 0. \quad (\text{A.13})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = 0. \quad (\text{A.14})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = - \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.15})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.16})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \left(V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} + V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \beta_{-\mu}^{\dagger} \beta_{-\lambda} + V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \delta_{\mu,\lambda} \right). \quad (\text{A.17})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta\omega_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \beta_{-\nu}^{\dagger} \right] = \sum_{\lambda} \sum_{\mu} \left(V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} + V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \beta_{-\mu}^{\dagger} \beta_{-\lambda} + V_{\lambda}^{\mu} \delta\omega_{\mu}^{\lambda} \delta_{\mu,\lambda} \right) \quad (\text{A.18})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda} \beta_{-\mu}. \quad (\text{A.19})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda} \beta_{-\mu}. \quad (\text{A.20})$$

A.2 Commutators involving the 2M lowering operator

A.2.1 First order

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, K_{\nu}^{-} \right] = - \sum_{\lambda} \delta\omega_{\lambda}^{\nu} \alpha_{\lambda} \beta_{-\nu}. \quad (\text{A.21})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta\omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, K_{\nu}^{-} \right] = - \sum_{\lambda} \delta\omega_{\lambda}^{\nu} \alpha_{\nu} \beta_{-\lambda}. \quad (\text{A.22})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, K_{\nu}^{-} \right] = - \sum_{\lambda} V_{\nu}^{\lambda} \left(\alpha_{\lambda}^{\dagger} \alpha_{\nu} + \beta_{-\lambda}^{\dagger} \beta_{-\nu} + \delta_{\lambda,\nu} \right). \quad (\text{A.23})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, K_k^{-} \right] = 0. \quad (\text{A.24})$$

A.2.2 Second order

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \delta \omega_{\mu}^{\lambda} \alpha_{\mu} \beta_{-\mu}. \quad (\text{A.25})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} \delta \omega_{\mu}^{\lambda} \delta \omega_{\lambda}^{\mu} \alpha_{\mu} \beta_{-\mu}. \quad (\text{A.26})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = \sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} V_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} - \sum_{\lambda} \sum_{\mu} \delta \omega_{\mu}^{\lambda} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}. \quad (\text{A.27})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \alpha_{\mu}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = 0. \quad (\text{A.28})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \delta \omega_{\mu}^{\lambda} \alpha_{\lambda} \beta_{-\mu}. \quad (\text{A.29})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} \delta \omega_{\mu}^{\lambda} \delta \omega_{\lambda}^{\mu} \alpha_{\mu} \beta_{-\mu}. \quad (\text{A.30})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = 0. \quad (\text{A.31})$$

$$\left[\sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = \sum_{\lambda} \sum_{\mu} \delta \omega_{\lambda}^{\mu} V_{\mu}^{\lambda} \beta_{-\lambda}^{\dagger} \beta_{-\mu} - \sum_{\lambda} \sum_{\mu} \delta \omega_{\mu}^{\lambda} V_{\lambda}^{\mu} \beta_{-\lambda}^{\dagger} \beta_{-\mu}. \quad (\text{A.32})$$

$$\begin{aligned} & \left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = \\ & - \sum_{\lambda} \sum_{\mu} \left(V_{\lambda}^{\lambda} \delta \omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} + V_{\lambda}^{\mu} \delta \omega_{\mu}^{\lambda} \beta_{-\mu}^{\dagger} \beta_{-\mu} + V_{\mu}^{\lambda} \delta \omega_{\lambda}^{\mu} \delta_{\mu, \lambda} \right). \end{aligned} \quad (\text{A.33})$$

$$\begin{aligned} & \left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = \\ & - \sum_{\lambda} \sum_{\mu} \left(V_{\lambda}^{\mu} \delta \omega_{\mu}^{\lambda} \alpha_{\lambda}^{\dagger} \alpha_{\mu} + V_{\mu}^{\lambda} \delta \omega_{\mu}^{\lambda} \beta_{-\mu}^{\dagger} \beta_{-\mu} + V_{\mu}^{\lambda} \delta \omega_{\lambda}^{\mu} \delta_{\mu, \lambda} \right). \end{aligned} \quad (\text{A.34})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} V_{\nu}^{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} \right] = - \sum_{\lambda} \sum_{\mu} V_{\mu}^{\lambda} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.35})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}, \sum_{\nu} V_{\nu}^{\nu} \beta_{-\nu}^{\dagger} \beta_{-\nu} \right] = - \sum_{\lambda} \sum_{\mu} V_{\mu}^{\lambda} V_{\lambda}^{\mu} \alpha_{\lambda}^{\dagger} \beta_{-\mu}^{\dagger}. \quad (\text{A.36})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = 0. \quad (\text{A.37})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta \omega_{\nu}^{\nu} \alpha_{\nu} \beta_{-\nu} \right] = 0. \quad (\text{A.38})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta \omega_k^{\nu} \alpha_{\nu}^{\dagger} \alpha_k \right] = \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \delta \omega_k^{\lambda} \alpha_k \beta_{-\mu}. \quad (\text{A.39})$$

$$\left[\sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \alpha_{\lambda} \beta_{-\mu}, \sum_{\nu} \delta \omega_k^{\nu} \beta_{-\nu}^{\dagger} \beta_{-k} \right] = \sum_{\lambda} \sum_{\mu} V_{\lambda}^{\mu} \delta \omega_k^{\mu} \alpha_{\lambda} \beta_{-k}. \quad (\text{A.40})$$

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