



PONTIFICIA UNIVERSIDAD CATÓLICA DE CHILE
FACULTAD DE FÍSICA
INSTITUTO DE FÍSICA

Characterization of Ferromagnetic and Antiferromagnetic Systems Using Nitrogen-Vacancy Center Relaxometry

by

Ricardo Javier Cabezón Heitzer

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Supervisor : Dr. Roberto Rodríguez (PUC, Chile)
Committee : Dr. Jerónimo Maze (PUC, Chile)
: Dr. Claudio González (PUC, Chile)

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Abstract

Relaxometry with nitrogen-vacancy (NV) centers in diamond is an innovative and non-invasive technique that has been increasingly used for characterizing magnetic materials. Because of their exceptional quantum coherence and capability to be manipulated optically, it is possible to measure their relaxation time and, from this quantity, probe physical characteristics of the environment. This technique is the so-called relaxometry. During the last few years, NV centers have been used for measuring magnetic properties in ferromagnetic materials, and a few times for antiferromagnetic systems with easy-axis anisotropy.

In this work, we develop a complete theoretical description of NV center relaxometry for several types of magnetic materials (ferromagnetic and antiferromagnetic systems), we indicate what physical parameters can be measured in each situation, when this technique will experimentally work, and how it depends on the sample anisotropy. We found that relaxation rates using ferromagnetic systems peak at certain values of the external magnetic field, where both NV center and sample resonate simultaneously, something that does not happen when using antiferromagnets because of their high resonance frequency. In this way, NV center relaxometry allows us to extract information about the spin waves of the system. Finally, we discuss prospects to non-intrusively probe magnetic properties and phase transitions in ferrimagnetic systems, in triangular-lattice antiferromagnets, such as Cs_2CuCl_4 and in 2D materials.

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Chapter 1

Introduction

1.1 Motivation

During the last decades, the development of electronic devices has been focused in finding out alternative and efficient ways of transport information [1]. Spintronics is the branch of condensed matter physics that deals with possible ways to generate, transport and detect polarized currents of spin angular momentum (spin currents).

Up to date, ferromagnetic systems have been widely used for spin currents generation, due to their capability of naturally presenting spin waves [2]. Ferromagnetic materials produce a spontaneous magnetization, due to the microscopic fact that spins tend to align parallelly to each other [2] (see Figure 1.1a). An exemplary material that is commonly treated as a ferromagnetic system is $\text{Y}_3\text{Fe}_5\text{O}_{12}$, also known as “YIG”, which stands out for its low spin waves damping [3].

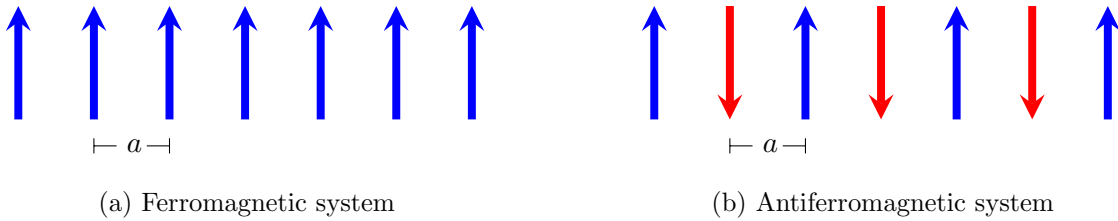


Figure 1.1: Comparison between ferromagnetic and antiferromagnetic basic structures. Blue and red arrows represent spin directions.

On the other hand, recent research [4] have proved the big potential that antiferromagnetic materials have in spintronics. In these systems, spins tend to align *anti-parallelly*, cancelling all net magnetization [5] (see Figure 1.1b). For this reason, such materials are robust against external magnetic perturbations.

Another advantage of these materials is the ultra-fast dynamics given by their resonance frequency, which is in the order of THz (while ferromagnetic frequencies are in the order of GHz) [5]. An intermediate situation between both cases are the *ferrimagnetic* systems: just like antiferromagnets, they also have spins pointing in opposite directions, but one direction dominates over the other, producing a net magnetization [6].

Due to the above, the development of new storage and processing technologies seeks to incorporate the use of antiferromagnetic materials, with an increasingly protagonist role [7, 8, 9]. However, when studying a new magnetic material, it is necessary to determine if it is ferromagnetic, ferrimagnetic or antiferromagnetic. In addition, it would be useful to determine magnetic properties of the sample, such as its (an)isotropy, magnetization, spin waves dissipation, etc. There exist appropriate techniques for every type of measurement [10, 11, 12, 13], but none of them is transverse; they tend to vary according to which parameter to measure and the type of material. Also, most of these techniques can only take global measurements of the sample, but not point-to-point.

A reliable method to deal these problems is using nitrogen-vacancy centers (NV^- centers, or NV for short), which are point defects in a diamond lattice [14], as illustrated in Figure 1.2, and whose optical properties make it a desirable sensor to probe environment fluctuations (electric, magnetic or thermal) [15]. In the next section we will explain more deeply how this can be done, but the main idea is to locate the NV center on the sample and measure the *relaxation time* a NV center takes to decay from one spin state to another, in a given external magnetic field. Such decay has an exponential form, and can be measured as photoluminescence in a photon detector.

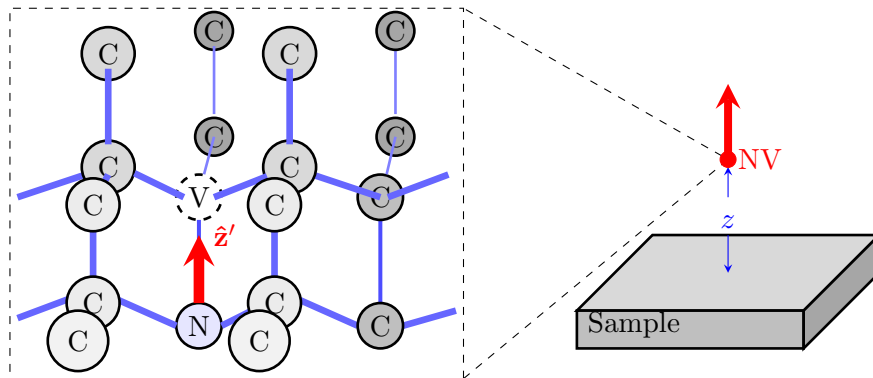


Figure 1.2: (a) Diamond lattice with a NV center and the coordinate system used attached to it. Here, the $\hat{z}'$ axis coincides with the anisotropy direction, which joins the nitrogen and the vacancy. From now on, NV centers will be represented as an arrow indicating its $\hat{z}'$ direction.

Measuring the photoluminescence decay as a function of time directly gives us how the spin occupation probability evolves in time. When a magnetic sample is located near to the NV center, this exponential decay becomes steeper (as illustrated in Figure 1.3), and one can obtain information of the sample from this faster decay. Particularly, the NV center has the big advantage that it can be initialized in one of its spin states (namely $|0\rangle$), almost with full probability, as will be explained in the next section.

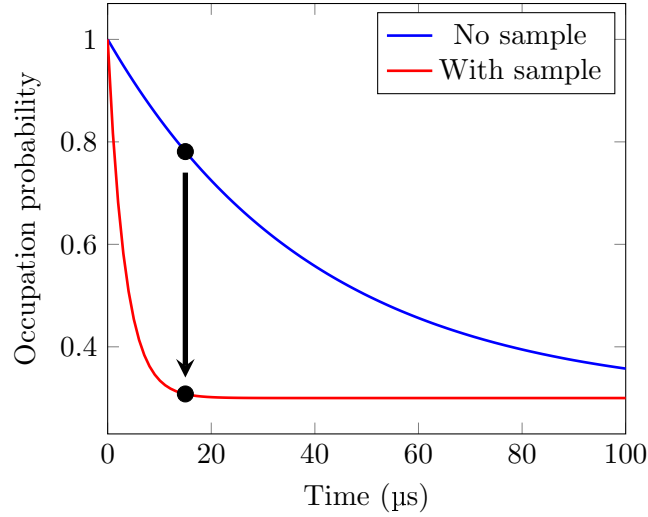


Figure 1.3: Occupation probability of the $|0\rangle$ spin state versus time, with and without a sample near to the NV center. In this case, these measurements were made using a paramagnetic GdCl_3 salt [16].

This molecule has been already used for measuring magnetic properties of YIG [17], and has demonstrated to be a good alternative to the conventional characterization techniques, such as ferromagnetic resonance. It also has been used a few times for similar measurements in antiferromagnetic systems [18], but only for a specific type of material, with uniaxial anisotropy (*easy-axis* anisotropy). For instance, there is still no experimental or theoretical characterization of antiferromagnetic materials with biaxial anisotropy (*easy-plane* anisotropy), even though their striking magnetic properties, like presenting two different resonances even at zero magnetic field [19].

The main goal of this work is to describe theoretically the interaction between a nitrogen-vacancy center and a magnetic material, developing with this an experimental method applicable to several types of samples. Specifically, we want to compare how NV center relaxation rates react against both ferromagnetic and antiferromagnetic materials, and determine the practical conditions the last materials must satisfy for these relaxation rates to be detected.

1.2 Description of this thesis

In this thesis, we describe theoretically the interaction between a magnetic sample and NV center relaxation times, as a method to measure physical quantities for magnetic materials. First, Chapter 2 introduces NV center dynamics. A three-level model is presented to describe how NV spin probabilities evolve in time, and how one can obtain the relaxation rates from this model. Also, we introduce NV center's electronic hamiltonian and explain mathematically how a time-dependent perturbation, generated by spin fluctuation in the sample, can be used to calculate the relaxation rates.

Chapter 3 describes the magnetic noise produced by the sample and how it is connected with the NV center via fluctuation-dissipation theorem. We model the sample as a 2D disk of magnetic dipoles with spin, displayed continuously. Using classical electrodynamics, we relate the spin fluctuations in the sample with the magnetic field detected by the NV center, and therefore obtain a general formula for the desired relaxation rates. This expression is given by the spin-spin correlation functions, so describing how spins statistically correlate in the sample plays a crucial role in this work.

Chapter 4 introduces the physical parameters of a ferromagnetic sample, necessary to calculate the relaxation rates. We describe here spin waves, their quanta (magnons) and how magnon energies are altered when anisotropies are present. Chapter 5 does the same but for antiferromagnetic materials. Both Chapters 4 and 5 are almost fully dedicated to introducing the required concepts of magnetic materials, without talking so much about NV centers.

Finally, Chapter 6 shows explicit calculations for the required spin-spin correlations, considering the materials introduced in previous chapters. Using these correlation functions, we obtain simulations for the relaxation rates, both for ferromagnetic and antiferromagnetic samples.

1.3 NV centers

The nitrogen-vacancy center is a point-defect in a diamond lattice, consisting of a nitrogen atom replacing a carbon, and therefore creating a vacancy next to it (as shown in Figure 1.2). Such molecule can be typically found in two charge states: NV^0 or NV^- , with different optical properties and angular momentum between them [14]. For our proposal, we use the NV^- , which we are going to write simply as NV. This charge configuration has a spin quantum number 1, so its energy ground state $|g\rangle$ is presented as a triplet $|g; -\rangle$, $|g; 0\rangle$ and $|g; +\rangle$, which from now on will be written as $|-\rangle$, $|0\rangle$ and $|+\rangle$, unless otherwise

indicated. Experimentally, it is known that between the states $|0\rangle$ and $|\pm\rangle$ exists a zero-field splitting, which in angular frequency units has the value $D_{\text{NV}} = 2\pi \times 2.87 \text{ GHz}$ [20], as shown in Figure 1.4. When an external magnetic field $\mathbf{B}_{\text{NV}}$ is applied, the degenerate spin states $|\pm\rangle$ split their energies via Zeeman effect. When $\mathbf{B}_{\text{NV}} = B_{\text{NV}}\hat{\mathbf{z}}'$, this energy difference is $2\hbar\gamma_{\text{NV}}B_{\text{NV}}$, where $\hbar$ is the reduced Planck's constant and $\gamma_{\text{NV}} = 2\mu_B/\hbar$ is the molecule gyromagnetic factor, being μ_B the Bohr magneton.

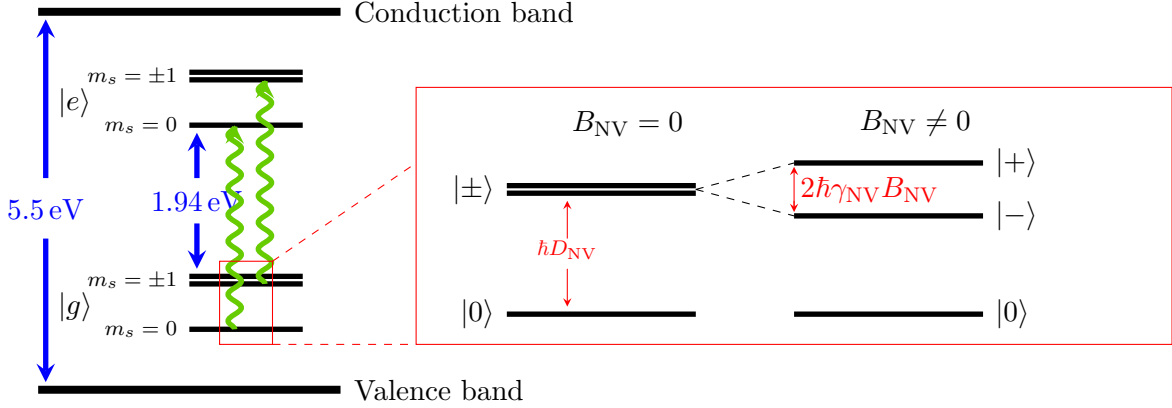


Figure 1.4: Energy levels of the NV center [20]. For the ground-state $|g\rangle$, there exists a zero-field splitting energy $\hbar D_{\text{NV}} = 2\pi\hbar \times 2.87 \text{ GHz}$ between the spin states $|0\rangle$ and $|-\rangle$, while the spin states $|+\rangle$ and $|-\rangle$ are degenerate. The orange box indicates how this degeneracy is lifted when an external magnetic field (in the z' direction) is applied.

Between the ground state $|g\rangle$ and the first excited state $|e\rangle$, there exists a *dark state* $|s\rangle$ of intermediate energy, whose transitions $|e\rangle \rightarrow |s\rangle$ and $|s\rangle \rightarrow |g\rangle$ do not irradiate visible light (see Figure 1.5). This state plays a crucial role for the optical manipulation of NV centers, due to its strong spin-selectivity and meta-stability. NV centers in the spin states $|e; \pm\rangle$ tend to pass through this meta-stable state instead of directly returning to $|g; \pm\rangle$. However, $|s\rangle$ is a singlet with spin state $m_s = 0$, so $|s\rangle \rightarrow |g; 0\rangle$ is the allowed transition. As a result, NV center's occupation is completely polarized in its $|g; 0\rangle$ state, almost with full probability, after exciting it into its $|e\rangle$ triplet.

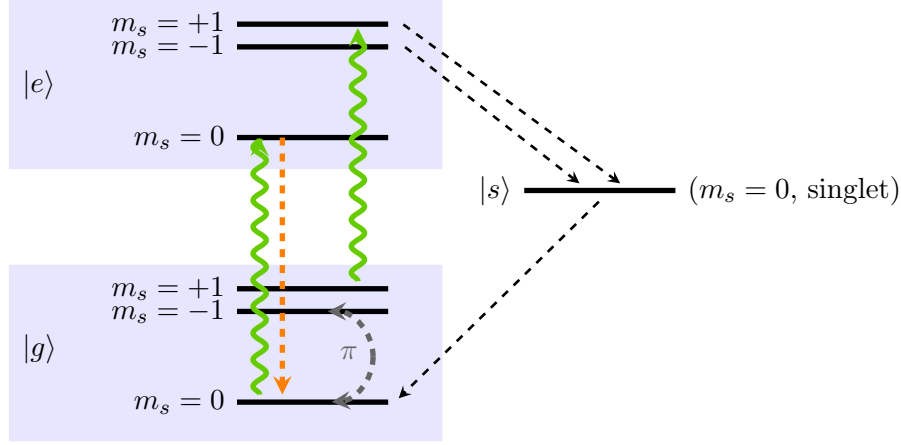


Figure 1.5: Allowed transitions between the states $|g\rangle$, $|e\rangle$ and the *dark state* $|s\rangle$ (meta stable). A NV center in the $|e; \pm\rangle$ state won't decay immediately to $|g; \pm\rangle$, but it passes through the meta stable $|s\rangle$, and then it decays to $|g; 0\rangle$, without emitting visible light. Dashed arrows between the $|g; 0\rangle$ and $|g; \pm\rangle$ states illustrate how a microwave (MW) of half-period (π -pulse) and resonant frequency ω_{MW} exchanges the occupation probabilities of $|g; 0\rangle$ and $|g; \pm\rangle$, depending on which spin state resonates.

The energy difference between the ground state $|g\rangle$ and the first excited state $|e\rangle$ is $\Delta E_{eg} = 1.94 \text{ eV}$, which is equivalent to a radiation wavelength¹ of $\lambda_{eg} \approx 640 \text{ nm}$. So, the necessary energy to excite a NV center (and therefore, to set it in the $|g; 0\rangle$ spin state) can be obtained from a typical green laser pulse² (532 nm) [20]. In this way, when this kind of pulse is received at instant $t = 0$, the initial condition of probability $P_0(t = 0) = 1$ is guaranteed. After some time t , the system will lose coherence due to the energetic exchange with its environment, as illustrated in Figure 1.6.

Moreover, one can measure this probability $P_0(t)$ at *any time* t by applying again a green laser pulse. As explained in Figure 1.4, NV centers excited from $|g; \pm\rangle$ spin states won't irradiate visible light when turning back to $|g\rangle$. In fact, the radiation given by the green laser is absorbed and re-emitted as non-visible radiation, resulting in less reply photons to count. This doesn't happen for NV centers in $|g; 0\rangle$ state. As a result, there is a direct relation between the photoluminescence probed by a photon detector and the occupation probability of the $|0\rangle$ state. Indeed, the *normalized* photoluminescence, with respect to this counting at $t = 0$, coincides with $P_0(t)$. One can also measure $P_{\pm}(t)$ (the occupation probability of the respective $|g; \pm\rangle$ spin states) at a given time t by applying a π -pulse microwave (MW) before reading out.

¹Here we use the relation $\Delta E_{eg} = h\nu_{eg} = hc/\lambda_{eg}$, being h the Planck's constant and c the speed of light.

²Actually, the green laser pulse gives the NV center an energy greater than ΔE_{eg} , as can be seen from their respective wavelengths ($532 \text{ nm} < \lambda_{eg}$). Fast, phonon-mediated relaxation takes the NV center into the electronic excited state $|e\rangle$, without emitting visible light.

As illustrated in Figure 1.5, this π pulse exchanges $P_0 \leftrightarrow P_+$ (if the microwave frequency resonates with the $|0\rangle \leftrightarrow |+\rangle$ transition) or $P_0 \leftrightarrow P_-$ (if the microwave frequency resonates with $|0\rangle \leftrightarrow |-\rangle$). In this way, one can measure all three probabilities $P_0(t)$, $P_+(t)$ and $P_-(t)$ and analyze their exponential decay to obtain information about NV center's environment. This experimental technique is the so-called *NV center relaxometry*.

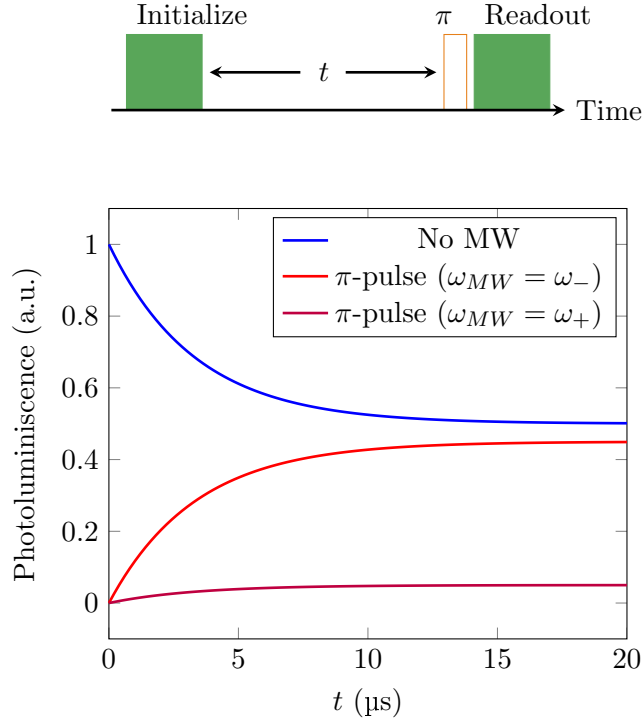


Figure 1.6: Photoluminescence versus time t . Top panel: optical and microwave (MW) sequence for NV relaxometry measurements. An initial green laser pulse (addressed as “Initialize”) is given to the NV center. After some variable time t , other green laser pulse is used for probing (“Readout”) the spin occupation probability ($P_0(t)$ if no microwave; $P_{\pm}(t)$ if the π -microwave angular frequency ω_{MW} coincides with the respective resonance frequencies $\omega_{\pm}$) as photoluminescence. Bottom panel: occupation probabilities P_0 , P_+ and P_- as functions of time t . In this case, external factors provoke the probability P_- to increase faster than P_+ , but the constraint $P_0(t) + P_+(t) + P_-(t) = 1$ is satisfied for each t .

Summarizing, the NV center has three interesting properties that makes it a great candidate for optical manipulation:

- Its occupation probability can be coherently initialized as $P_0(t=0) = 1$, just by giving it the necessary excitation energy.
- The later excitation energy can be obtained from a typical, nonresonant green laser radiation

(532 nm).

- The returning transition $|e; 0\rangle \rightarrow |g; 0\rangle$ irradiates visible light while $|e; \pm\rangle \rightarrow |s\rangle \rightarrow |g; 0\rangle$ don't, so one can also use green laser pulses for reading $|g; 0\rangle$ state's occupation.

1.4 NV center relaxometry

The central quantities in this work are the NV center relaxation rates $\Gamma_{\pm}$, associated with the transitions $|0\rangle \leftrightarrow |\pm\rangle$. In a three-level poblational model (see Figure 1.7), those $\Gamma_{\pm}$ are the transition rates that relate the occupation probabilities of each level.

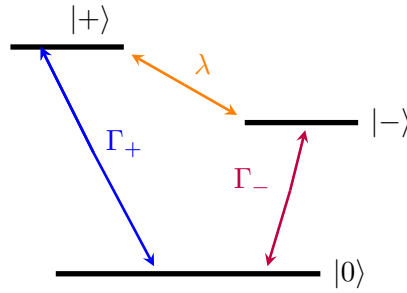


Figure 1.7: Energy-level diagrams with their respective transitions and relaxation rates. Transitions between $|+\rangle$ and $|-\rangle$ states are negligible up to first order. Also, we consider Γ_+ as the same factor both for absorption and decay (the same for Γ_-).

Based on the three-level diagram, we write

$$\begin{aligned}\frac{dP_0}{dt} &= -\Gamma_+ P_0 - \Gamma_- P_0 + \Gamma_+ P_+ + \Gamma_- P_- \\ \frac{dP_+}{dt} &= \Gamma_+ P_0 - (\Gamma_+ + \lambda) P_+ + \lambda P_- \\ \frac{dP_-}{dt} &= \Gamma_- P_0 + \lambda P_+ - (\Gamma_- + \lambda) P_-, \end{aligned}$$

In matrix form,

$$\frac{d}{dt} \begin{bmatrix} P_0 \\ P_+ \\ P_- \end{bmatrix} = \mathbf{\Gamma} \mathbf{P} = \begin{bmatrix} -(\Gamma_+ + \Gamma_-) & \Gamma_+ & \Gamma_- \\ \Gamma_+ & -\Gamma_+ - \lambda & \lambda \\ \Gamma_- & \lambda & -\Gamma_- - \lambda \end{bmatrix} \begin{bmatrix} P_0 \\ P_+ \\ P_- \end{bmatrix}. \quad (1.1)$$

The probabilities $P_0(t)$, $P_+(t)$ and $P_-(t)$ can be measured directly in the experiment, as photoluminescence. The relaxation rates $\Gamma_{\pm}$ define how these probabilities evolve, and during the last years, there has been a huge research topic to find how these rates depend on the physical properties of the sample, so one can measure the sample just analyzing NV photoluminescences. This experimental technique is

the so-called NV center relaxometry. Its application to magnetic samples was first reported in 2015 by Van der Sar et al. [21] using a permalloy sample, and six years later it was used to measure the effective magnetization M_{eff} of a YIG sample [17], giving excellent agreement with the ferromagnetic resonance technique. In both situations, they had to fit the relaxation rates $\Gamma_{\pm}(B_{NV}, M_{eff})$ to experimental data of photoluminescence. Chapters 2 and 3 are fully dedicated to show a derivation for this explicit formula, and in Chapter 4 and in Chapter 5 we adjust it for the ferromagnetic and antiferromagnetic cases, respectively.

Chapter 2

NV center dynamics

The nitrogen-vacancy center is a point-defect in a diamond lattice, consisting of a nitrogen atom replacing a carbon, and therefore creating a vacancy next to it (as shown in Figure 1.2). As a spin-1 molecule, its ground state $|g\rangle$ is disposed as a triplet of spin states $|0\rangle$ and $|\pm\rangle$, with a zero-field-splitting energy $\hbar D_{\text{NV}}$ between $|0\rangle$ and $|\pm\rangle$, as introduced in Section 1.3. In this chapter, we explain more deeply the NV dynamics and how it reacts against an external magnetic perturbation (caused, for example, by spin fluctuations in a near magnetic system). Considering these perturbations, we calculate the needed relaxation rates $\Gamma_{\pm}$ that appear in relaxometry techniques. Finally, we explain some technical details and limitations given by the NV center one must consider when doing the experiment.

2.1 Three-level model

In Section 1.4 was introduced the system of differential equations that describes the time evolution of the probabilities $P_0(t)$, $P_+(t)$ and $P_-(t)$ for the spin states $|0\rangle$, $|+\rangle$ and $|-\rangle$, respectively, given by (1.1) in the form $d\mathbf{P}/dt = \mathbf{\Gamma}\mathbf{P}$, with $\mathbf{P} = \begin{bmatrix} P_0 & P_+ & P_- \end{bmatrix}^\top$ and

$$\mathbf{\Gamma} = \begin{bmatrix} -(\Gamma_+ + \Gamma_-) & \Gamma_+ & \Gamma_- \\ \Gamma_+ & -\Gamma_+ & 0 \\ \Gamma_- & 0 & -\Gamma_- \end{bmatrix}. \quad (2.1)$$

Up to first order, the relaxation rate λ between the $|+\rangle$ and $|-\rangle$ spin states is unaffected by the magnetic noise [21], so $\lambda \ll \Gamma_{\pm}$ and we can set $\lambda \rightarrow 0$. The resulting differential equations bring a useful relation for $\Gamma_{\pm}$:

$$\left. \frac{dP_{\pm}}{dt} \right|_{t=0} = \Gamma_{\pm} P(t=0). \quad (2.2)$$

Diagonalizing $\mathbf{\Gamma}$, we find

$$\mathbf{\Gamma} = \mathbf{\Lambda} \mathbf{\Gamma}' \mathbf{\Lambda}^\top$$

where

$$\mathbf{\Lambda} = \begin{bmatrix} 1 & \mathcal{A}_+ & \mathcal{A}_- \\ 1 & \mathcal{B}_+ & \mathcal{B}_- \\ 1 & 1 & 1 \end{bmatrix},$$

being these coefficients given by

$$\begin{aligned} \mathcal{A}_{\pm} &= -\frac{\Gamma_-^2 + \Gamma_+^2 - (\Gamma_+ + \Gamma_-)\lambda \pm (\Gamma_+ + \Gamma_-)\sigma}{\Gamma_-^2 - \Gamma_+\lambda \pm \Gamma_-\sigma}, \\ \mathcal{B}_{\pm} &= -\frac{\mp\Gamma_+^2 \pm \Gamma_-\lambda - \Gamma_+\sigma}{\pm\Gamma_-^2 \mp \Gamma_+\lambda + \Gamma_-\sigma}, \end{aligned}$$

with

$$\sigma = \sqrt{\Gamma_-^2 - \Gamma_+\Gamma_- + \Gamma_+^2 - \Gamma_-\lambda - \Gamma_+\lambda + \lambda^2},$$

and

$$\mathbf{\Gamma}' = \begin{bmatrix} 0 & 0 & 0 \\ 0 & -\Gamma_+ - \Gamma_- - \lambda - \sigma & 0 \\ 0 & 0 & -\Gamma_+ - \Gamma_- - \lambda + \sigma \end{bmatrix}$$

is the diagonal matrix of eigenvalues. The general solution of the equation system (1.1) can be written in terms of these elements:

$$\mathbf{P} = c_1 \begin{bmatrix} 1 \\ 1 \\ 1 \end{bmatrix} + c_2 \begin{bmatrix} \mathcal{A}_+ \\ \mathcal{B}_+ \\ 1 \end{bmatrix} \exp(-\Gamma_+ - \Gamma_- - \lambda - \sigma)t + c_3 \begin{bmatrix} \mathcal{A}_- \\ \mathcal{B}_- \\ 1 \end{bmatrix} \exp(-\Gamma_+ - \Gamma_- - \lambda + \sigma)t$$

We know that NV centers can initialize in its $|0\rangle$ spin state by giving them a green laser pulse (532 nm), so we can realistically state the initial condition $\mathbf{P}(t=0) = \begin{bmatrix} 1 & 0 & 0 \end{bmatrix}^\top$. In this case, the coefficients c_1 , c_2 and c_3 are given by

$$c_1 + \mathcal{A}_+c_2 + \mathcal{A}_-c_3 = 1$$

$$c_1 + \mathcal{B}_+c_2 + \mathcal{B}_-c_3 = 0$$

$$c_1 + c_2 + c_3 = 0$$

For instance, the first component is

$$\begin{aligned} P_0(t) &= -\frac{\mathcal{B}_- - \mathcal{B}_+}{\mathcal{A}_- - \mathcal{A}_+ - \mathcal{B}_- + \mathcal{A}_+\mathcal{B}_- + \mathcal{B}_+ - \mathcal{A}_-\mathcal{B}_+} \\ &\quad - \frac{1 - \mathcal{B}_-}{\mathcal{A}_- - \mathcal{A}_+ - \mathcal{B}_- + \mathcal{A}_+\mathcal{B}_- + \mathcal{B}_+ - \mathcal{A}_-\mathcal{B}_+} \mathcal{A}_+ \exp(-\Gamma_+ - \Gamma_- - \lambda - \sigma)t \\ &\quad + \frac{1 - \mathcal{B}_+}{\mathcal{A}_- - \mathcal{A}_+ - \mathcal{B}_- + \mathcal{A}_+\mathcal{B}_- + \mathcal{B}_+ - \mathcal{A}_-\mathcal{B}_+} \mathcal{A}_- \exp(-\Gamma_+ - \Gamma_- - \lambda + \sigma)t. \end{aligned}$$

In the special case where $\Gamma_+ = \Gamma_- = \Gamma$ (e.g, $B_{\text{NV}} = 0$, without external magnetic field), the transformation matrix $\mathbf{\Lambda}$ reduces to

$$\mathbf{\Lambda} = \begin{bmatrix} 1 & 0 & -2 \\ 1 & -1 & 1 \\ 1 & 1 & 1 \end{bmatrix}.$$

and the solution for $\mathbf{P}$ is enormously simplified:

$$P_0(t) = \frac{1}{3} + \frac{2}{3} \exp(-3\Gamma t).$$

Note that this probability evolution considers the spontaneous emission and radiation absorption of the NV center. These are the mechanisms that allows the molecule to move from one state to another throughout the time t in this model. This time t is what one can actually measure through the experiment, and in that sense, it is a *macroscopic* time.

2.2 NV center electronic hamiltonian

Negatively charged nitrogen-vacancy (NV^-) centers can be seen as a pair of electrons, interacting between them as two magnetic dipoles. This gives rise to an interaction potential [22] of the form (see Appendix A)

$$\hat{H}_{int} = \frac{1}{\hbar} \hat{\mathbf{S}}^\dagger \mathbf{D} \hat{\mathbf{S}} \quad (2.3)$$

where $\hat{\mathbf{S}} = \hat{S}_x \hat{\mathbf{x}}' + \hat{S}_y \hat{\mathbf{y}}' + \hat{S}_z \hat{\mathbf{z}}'$ is the spin angular momentum vector (considering spin 1) and $\mathbf{D}$ matrix representation of the zero-field splitting tensor. This matrix is symmetric and diagonalizable, so one can always choose a coordinate system for the spatial coordinates such that

$$\hat{H}_{int} = \frac{1}{\hbar} \left(D_x \hat{S}_x^2 + D_y \hat{S}_y^2 + D_z \hat{S}_z^2 \right). \quad (2.4)$$

This matrix representation uses the three principal axes as canonical basis, being one of them the symmetry axis $\hat{\mathbf{z}}'$. The other two elements are the $\hat{\mathbf{x}}'$ and $\hat{\mathbf{y}}'$ directions.

Putting this interaction together with the Zeeman effect term, the total electronic NV hamiltonian is built:

$$\hat{H} = \gamma_{\text{NV}} \mathbf{B}_{\text{NV}} \cdot \hat{\mathbf{S}} + \frac{1}{\hbar} \left(D_x \hat{S}_x^2 + D_y \hat{S}_y^2 + D_z \hat{S}_z^2 \right) \quad (2.5)$$

where $\gamma_{\text{NV}} = 2\pi \times 2.86 \text{ MHz/G}$ is the NV center gyromagnetic factor and $\mathbf{B}_{\text{NV}}$ is the magnetic field detected by the molecule. Note that other energy contributions, like electron-nuclei interaction or nuclear Zeeman effect, were neglected. From now on, we will consider only electronic energies. It is possible to

reduce even more this expression by defining $D_{\text{NV}} = 3D_z/2$ and $E_{\text{NV}} = (D_x - D_y)/2$, obtaining

$$\hat{H} = \gamma_{\text{NV}} \mathbf{B}_{\text{NV}} \cdot \hat{\mathbf{S}} + \frac{D_{\text{NV}}}{\hbar} \left(\hat{S}_z^2 + \frac{2}{3} \right) + \frac{E_{\text{NV}}}{\hbar} (\hat{S}_x^2 - \hat{S}_y^2) \quad (2.6)$$

Here, $D_{\text{NV}} = 2\pi \times 2.87$ GHz is the same *zero-field splitting* factor mentioned in Chapter 1, while the E_{NV} anisotropy factor introduces a splitting between the $|+\rangle$ and $|-\rangle$ energies. This anisotropy in the $x'y'$ plane appears due to strain in the diamond lattice. However, compared to D_{NV} , this factor E_{NV} is relatively small; it is just of a few MHz, and rarely exceeds $2\pi \times 5$ MHz [23]. For this reason, we can neglect this term, too, resulting finally

$$\hat{H} = \gamma_{\text{NV}} \mathbf{B}_{\text{NV}} \cdot \hat{\mathbf{S}} + \frac{D_{\text{NV}}}{\hbar} \hat{S}_z^2. \quad (2.7)$$

where the constant term $2D/3$ was omitted. If we choose the spin state representation

$$|+\rangle = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix} \quad |0\rangle = \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix} \quad |-\rangle = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix} \quad (2.8)$$

the spin operators are written in matrix form as

$$\hat{S}_x = \frac{\hbar}{\sqrt{2}} \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix} \quad \hat{S}_y = \frac{\hbar}{\sqrt{2}} \begin{bmatrix} 0 & -i & 0 \\ i & 0 & -i \\ 0 & i & 0 \end{bmatrix} \quad \hat{S}_z = \hbar \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix}. \quad (2.9)$$

Using this representation, one can build a 3×3 square matrix for $\hat{H}$ in equation (2.7). The eigenvalues E_+ , E_0 and E_- of this matrix represent the eigenenergies, and the NV resonance frequencies are defined as $\omega_{\pm} = (E_{\pm} - E_0)/\hbar$. In this way, a NV center, initially in its spin state $|0\rangle$, can take two possible transitions: $|0\rangle \leftrightarrow |+\rangle$ or $|0\rangle \leftrightarrow |-\rangle$, whose respective resonance frequencies $\omega_{\pm}$ depend on the external magnetic field $\mathbf{B}_{\text{NV}}$, as illustrated in Figure 2.1. In the simplest case, when the external magnetic field is aligned with the direction $\hat{\mathbf{z}}'$ of the NV center ($\mathbf{B}_{\text{NV}} = B_{\text{NV}}\hat{\mathbf{z}}'$), we have $\omega_{\pm} = D_{\text{NV}} \pm \gamma_{\text{NV}} B_{\text{NV}}$. On the other hand, if $\mathbf{B}_{\text{NV}}$ is not parallel to $\hat{\mathbf{z}}'$, it is necessary to find $\omega_{\pm} = \omega_{\pm}(\mathbf{B}_{\text{NV}})$ by solving the eigenvalues of the hamiltonian operator (2.7).

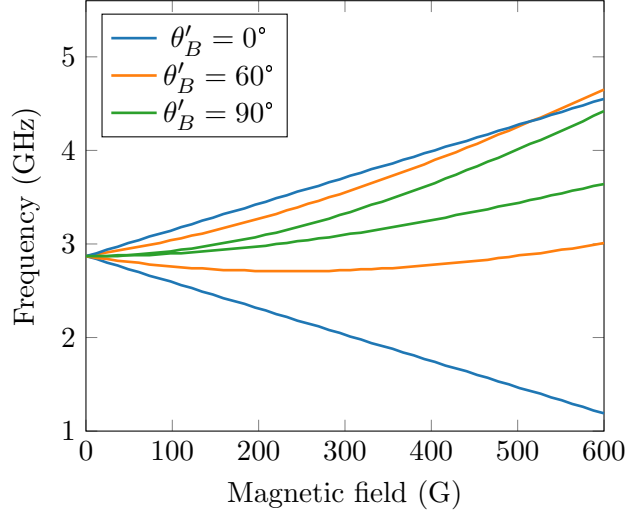


Figure 2.1: NV resonance frequencies $\omega_{\pm}$ versus external magnetic field $\mathbf{B}_{\text{NV}}$, using several angles θ'_B between $\mathbf{B}_{\text{NV}}$ and $\hat{\mathbf{z}}'$ [23]. For each angle, $\omega_+ \geq \omega_-$, and both frequencies converge to $D_{\text{NV}}/(2\pi) = 2.87 \text{ GHz}$ as $B_{\text{NV}} \rightarrow 0$.

2.3 Perturbed relaxation rates

One can find the relaxation rate coefficients $\Gamma_{\pm}$ by using time-dependent quantum perturbation theory [24]. Considering magnetic field perturbations, the unperturbed hamiltonian in (2.7) turns out to be the stationary part of the total hamiltonian

$$\hat{H}(t) = \hat{H}^{(0)} + \hat{H}'(t),$$

where

$$\hat{H}'(t) = \hbar \gamma_{\text{NV}} \sum_{j=1}^3 B'_j(t) \hat{S}_j \quad (2.10)$$

is the time-dependent perturbation for the Zeeman term $\gamma_{\text{NV}} \mathbf{B}_{\text{NV}} \cdot \hat{\mathbf{S}}$. This fluctuation contains all time-dependence of the magnetic field.

The ground state $|g\rangle$ can be expanded [25] as

$$\begin{aligned} |g\rangle &= c_0(t) \exp\left(-\frac{iE_0 t}{\hbar}\right) |0\rangle + c_+(t) \exp\left(-\frac{iE_+ t}{\hbar}\right) |+\rangle + c_-(t) \exp\left(-\frac{iE_- t}{\hbar}\right) |-\rangle \\ \Rightarrow |g\rangle &= \sum_{m=-1}^1 c_m(t) \exp\left(-\frac{iE_m t}{\hbar}\right) |m\rangle. \end{aligned}$$

Time-dependent perturbation theory is useful for finding the coefficients $c_0(t)$, $c_+(t)$ and $c_-(t)$. These coefficients allow us to calculate the transition probability $P_{\ell n}(t)$ of a NV center going from the $|\ell\rangle$ spin

state to the $|n\rangle$, within the time interval $[0, t]$. This probability can be formulated as follows: it's the probability of being in the state $|n\rangle$ at instant t , *given that it was initially in the state $|\ell\rangle$ at instant 0*:

$$P_{\ell n}(t) = |c_n(t)|^2 \Big|_{c_\ell(0)=1}, \quad (2.11)$$

whose derivative,

$$\frac{dP_{\ell n}}{dt} = c_n^* \frac{dc_n}{dt} + c_n \frac{dc_n^*}{dt} \quad (2.12)$$

can be used to obtain the relaxation rates $\Gamma_{\ell n}$, where the case $\ell = 0$ and $n = +$ gives us Γ_+ , while $n = -$ returns Γ_- .

Note that these transitions, unlike those shown in Section 2.1, are directly caused by the perturbation $\hat{H}'(t)$, and have nothing to do with spontaneous emission or radiation absorption. When $B_j(t) = 0$, this transition completely vanishes. In this way, these corrections caused by the perturbation are *too fast* when compared to the *macroscopic* transitions from Section 2.1, which is to say, within the interval $[0, t]$, only transitions due to perturbations occur, and the radiative transitions will only appear if we take $t \rightarrow \infty$. So, as the measurement time t lies around the initial time $t = 0$ in equation (2.2),

$$\Gamma_{\ell n} = \left\langle \frac{dP_{\ell n}}{dt} \Big|_{t=0} \right\rangle = \left\langle \left(c_n^* \frac{dc_n}{dt} + c_n \frac{dc_n^*}{dt} \right)_{c_\ell(t)=1} \right\rangle \quad (2.13)$$

where we used the definition (2.11) in the last identity and $\langle \dots \rangle$ is the canonical average. This average comes out due to the fact we are macroscopically measuring this correction to the relaxation rate caused by microscopic fluctuations $\{B_j(t)\}_{j=1}^3$.

According to Schrödinger's equation,

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} |g\rangle &= \hat{H} |g\rangle \\ \Rightarrow i\hbar \frac{\partial}{\partial t} \sum_{m=-1}^1 c_m(t) \exp\left(-\frac{iE_m t}{\hbar}\right) |m\rangle &= \hat{H} \sum_{m=-1}^1 c_m(t) \exp\left(-\frac{iE_m t}{\hbar}\right) |m\rangle \\ \Rightarrow \sum_{m=-1}^1 \left\{ i\hbar \frac{dc_m}{dt} \exp\left(-\frac{iE_m t}{\hbar}\right) |m\rangle + E_m c_m(t) \exp\left(-\frac{iE_m t}{\hbar}\right) |m\rangle \right\} \\ &= \sum_{m=-1}^1 c_m(t) \exp\left(-\frac{iE_m t}{\hbar}\right) [\hat{H}^{(0)} + \hat{H}'(t)] |m\rangle. \end{aligned}$$

Taking the inner product with $|n\rangle$, using the fact that eigenstates are orthogonal and defining $\omega_{mn} = (E_m - E_n)/\hbar$, we get

$$\frac{dc_n}{dt} = -\frac{i}{\hbar} \sum_{m=-1}^1 \langle n | \hat{H}'(t) | m \rangle c_m(t) \exp(-i\omega_{mn}t). \quad (2.14)$$

Up to first order (approximating $c_n(t) \approx c_n(0)$ in equation (2.14)),

$$c_n \approx c_n(0) - \frac{i}{\hbar} \sum_{m=-1}^1 c_m(0) \int_0^t \langle n | \hat{H}'(t') | m \rangle \exp(-i\omega_{mn}t') dt'. \quad (2.15)$$

So, inserting equations (2.14) and (2.15) into (2.13), for $\ell \neq n$, we obtain

$$\begin{aligned} \frac{dP_{\ell n}}{dt} &= \frac{1}{\hbar^2} \int_0^t \langle \ell | \hat{H}'(t') | n \rangle \langle n | \hat{H}'(t) | \ell \rangle \exp i\omega_{\ell n}(t' - t) dt' \\ &\quad + \frac{1}{\hbar^2} \int_0^t \langle n | \hat{H}'(t') | \ell \rangle \langle \ell | \hat{H}'(t) | n \rangle \exp i\omega_{\ell n}(t - t') dt'. \end{aligned} \quad (2.16)$$

Taking canonical average $\langle \dots \rangle$ and inserting this result into (2.13),

$$\begin{aligned} \Gamma_{\ell n} &= \frac{1}{\hbar^2} \int_{-t}^0 \left\langle \langle \ell | \hat{H}'(t + \tau) | n \rangle \langle n | \hat{H}'(t) | \ell \rangle \right\rangle \exp i\omega_{\ell n}\tau d\tau \\ &\quad - \frac{1}{\hbar^2} \int_t^0 \left\langle \langle n | \hat{H}'(t - \tau) | \ell \rangle \langle \ell | \hat{H}'(t) | n \rangle \right\rangle \exp i\omega_{\ell n}\tau d\tau. \end{aligned}$$

where the substitution $\tau = t' - t$ was performed in the first integral, while $\tau = t - t'$ was used in the second one. The quantity $G_{\ell m}(t, \tau) = \left\langle \langle \ell | \hat{H}'(t + \tau) | n \rangle \langle n | \hat{H}'(t) | \ell \rangle \right\rangle$ is a correlation function, and it must satisfy the properties $G_{\ell m}(t, \tau) = G_{\ell m}(0, \tau) = G_{\ell m}(\tau)$ and $G_{\ell m}(\tau) = G_{\ell m}(-\tau)$. As usual for time-dependent quantum perturbations theory, we consider $t \gg 1/\omega_{\ell n}$. So,

$$\Gamma_{\ell n} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \left\langle \langle \ell | \hat{H}'(\tau) | n \rangle \langle n | \hat{H}'(0) | \ell \rangle \right\rangle \exp i\omega_{\ell n}\tau d\tau.$$

The next step is to replace the perturbation $\hat{H}'(t)$ using equation (2.10):

$$\begin{aligned} \Gamma_{\ell n} &= \int_{-\infty}^{\infty} \left\langle \langle \ell | \gamma_{\text{NV}} \sum_{j=1}^3 \hat{S}_j B'_j(\tau) | n \rangle \langle n | \gamma_{\text{NV}} \sum_{k=1}^3 \hat{S}_k B'_k(0) | \ell \rangle \right\rangle \exp i\omega_{\ell n}\tau d\tau \\ \Rightarrow \Gamma_{\ell n} &= \gamma_{\text{NV}}^2 \sum_{j=1}^3 \sum_{k=1}^3 \underbrace{\langle \ell | \hat{S}_j | n \rangle \langle n | \hat{S}_k | \ell \rangle \int_{-\infty}^{\infty} \langle \hat{B}'_j(\tau) \hat{B}'_k(0) \rangle \exp i\omega_{\ell n}\tau d\tau}_{A_{jk}} \end{aligned}$$

Clearly $A_{jk} = A_{kj}^*$, so this sum can be rewritten¹ as

$$\Gamma_{\ell n} = \gamma_{\text{NV}}^2 \sum_{j=1}^3 \sum_{k \geq j}^3 2 \text{Re}(A_{jk})$$

From the explicit spin matrices (2.9) one may notice that, for $j \neq z$ and $k \neq z$, the product $\langle \ell | \hat{S}_j | n \rangle \langle n | \hat{S}_k | \ell \rangle$ will always be an imaginary number without real part, unless $j = k$. On the other hand, if $j = z$ or $k = z$, the product $\langle \ell | \hat{S}_j | n \rangle \langle n | \hat{S}_k | \ell \rangle$ will result immediately in 0 given that $\hat{S}_z$ is represented by a diagonal matrix. Therefore, this double sum can be reduced to a single one:

$$\Gamma_{\ell n} = \gamma_{\text{NV}}^2 \sum_{j \neq z} |\langle \ell | \hat{S}_j | n \rangle|^2 \int_{-\infty}^{\infty} \langle \hat{B}'_j(\tau) \hat{B}'_j(0) \rangle \exp i\omega_{\ell n}\tau d\tau.$$

¹Here, we have implicitly promoted the field fluctuations to operators: $B_j(t) \rightarrow \hat{B}_j(t)$ for $j = x, y, z$. How these are related to other operators will be explained in the next chapter.

Whether $j = x$ or $j = y$, we have $|\langle \ell | S_j | n \rangle|^2 = 1/2$,

$$\Rightarrow \Gamma_{\ell n} = \frac{\gamma_{\text{NV}}^2}{2} \sum_{j \neq z} \int_{-\infty}^{\infty} \left\langle \hat{B}'_j(\tau) \hat{B}'_j(0) \right\rangle \exp(i\omega_{\ell n} \tau) d\tau.$$

As mentioned before, the coefficients $\Gamma_{\pm}$ are the central quantity of the relaxometry technique, and finding them out gives us a complete description of the transitions between the spin states. If $\hbar\omega_{\pm}$ is the energy difference between the $|\pm\rangle$ y $|0\rangle$ eigenstates, we have

$$\Gamma_{\pm} = \Gamma(\omega_{\pm}) = \frac{\gamma_{\text{NV}}^2}{2} \sum_{j \neq z} \int_{-\infty}^{\infty} \left\langle \hat{B}'_j(\tau) \hat{B}'_j(0) \right\rangle \exp(i\omega_{\pm} \tau) d\tau. \quad (2.17)$$

We observe that the relaxation rates $\Gamma_{\pm}$ are given by the temporal Fourier transform of the transverse magnetic correlation functions, evaluated in the respective frequencies $\omega_{\pm}$.

2.4 NV center sensibility

One striking application of NV centers is its capability of measuring external magnetic fields at a position $\mathbf{r}$. The experimental setup has a very basic idea: a NV center is located at $\mathbf{r}$ and the external magnetic field $\mathbf{B}_{\text{NV}}$ we want to measure is turned on. At $t = 0$, the NV center receives a green laser pulse (532 nm) in such a way that it is initialized in its spin $|0\rangle$ state, emitting visible light due to the $|e\rangle \rightarrow |g\rangle$ transition (as explained earlier in Chapter 1). This light intensity is measured as photoluminescence PL in a photon detector.

As discussed before, the NV resonance frequencies $\omega_{\pm}$ depend on $\mathbf{B}_{\text{NV}}$, so one can also apply a microwave of angular frequency ω and sweep it around the zero field splitting $D_{\text{NV}} = 2\pi \times 2.87 \text{ GHz}$. When $\omega = \omega_-$, the molecule absorbs the electromagnetic radiation and uses it to perform a transition $|0\rangle \rightarrow |-\rangle$, resulting in a lesser photoluminescence PL . The same occurs when $\omega = \omega_+$. As a result, the curves of photoluminescence PL versus frequency ω have two down peaks, as shown in Figure 2.2. Such peaks have lorentzian forms, with a contrast C , typically below 0.3. The splitting between these two minima gives us the field magnitude B_{NV} . In fact, when $\mathbf{B}_{\text{NV}}$ is aligned with the anisotropy axis $\hat{\mathbf{z}}'$, we have $\omega_+ - \omega_- = 2\gamma_{\text{NV}} B_{\text{NV}}$. This technique is known as *magnetometry*.

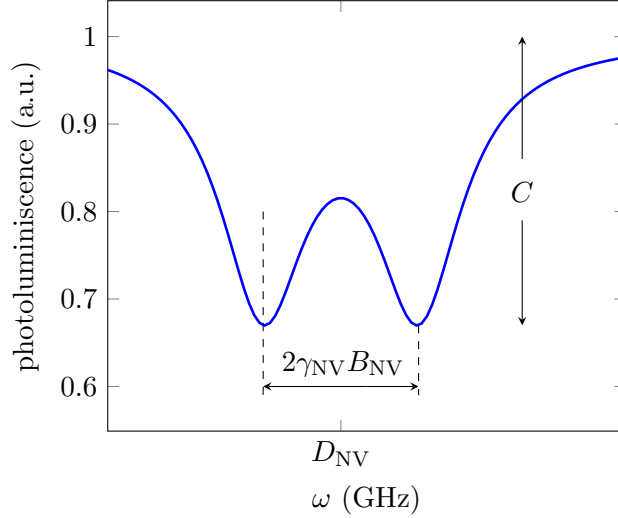


Figure 2.2: Photoluminescence versus frequency, for an external magnetic field B_{NV} . If $\mathbf{B}_{\text{NV}} = B_{\text{NV}}\hat{\mathbf{z}}'$, the splitting between the two resonance frequencies is $2\gamma_{\text{NV}}B_{\text{NV}}$.

This technique has a maximum theoretical precision, which can be obtained by considering the contrast C [20]. One can show that the minimum measurable difference between ω_+ and ω_- is

$$\Delta\omega \approx \frac{a}{2C\sqrt{I_0\Delta t}} \quad (2.18)$$

where a is the half-width at half-maximum of the Lorentzian bands, C is the optical contrast, I_0 is the photon counting rate and Δt is the measurement time. For instance, if $a \approx 9.5 \text{ MHz}$, $C \approx 0.3$, $I_0 \approx 5 \times 10^4 \text{ s}^{-1}$ and $\Delta t \approx 1 \text{ s}$, we get $\Delta\omega \approx 2\pi \times 73 \text{ kHz}$. So, in this situation, the minimum measurable variation of magnetic field is $\Delta B_{\text{NV}} \sim 1 \text{ } \mu\text{T}$. This sensitivity can be improved measuring several NV centers simultaneously (*ensemble magnetometry*), from structures like nanodiamonds, and one can even detect magnetic fields of the order of nT [15].

In summary, Nitrogen-Vacancy centers are able to detect magnetic fields just by measuring their resonance frequencies $\omega_{\pm} = \omega_{\pm}(\mathbf{B}_{\text{NV}})$. This field-dependence not only allows us to probe the magnitude B_{NV} , but also it is possible to describe how the relaxation rates $\Gamma_{\pm} = \Gamma(\omega_{\pm}) = \Gamma_{\pm}(\mathbf{B}_{\text{NV}})$ depend on the external magnetic field. So, the intuitive experiment proposed here is to create a graph of $\Gamma_{\pm}$ for several values of B_{NV} , and fitting that curve with the result (2.17) would determine the physical properties of the sample, hidden in the field correlators $\langle B'_j(t) B'_j(0) \rangle$. What we have to do now is to expand this correlation function in terms of the time-dependent spin fluctuations of the sample.

Chapter 3

Magnetic noise

The relaxation rates $\Gamma = \Gamma(\omega)$ of the NV center were expressed in Chapter 2 as functions of the magnetic noise detected by the molecule. However, we hardly talked about where these magnetic perturbations are generated. In this chapter, we consider a magnetic sample which generates this field variation through spin fluctuations, and we will relate it with the spin-spin correlation functions of the sample, using statistical mechanics tools.

3.1 Stray field generated by a magnetic sample

The main idea of the experimental setup consists of positioning a NV center at a distance z on the sample (see Figure 3.1). We model the sample as a continuous two-dimensional (2D) disk of magnetic dipoles. At each position $\mathbf{r}''$, there is a dipole and therefore a magnetization $\mathbf{M}(\mathbf{r}'')$. According to classical electrodynamics, this magnetization generates a magnetic field $\mathbf{B}(\mathbf{r})$ (also known as *stray field*)¹ at the position $\mathbf{r} = z\mathbf{z}$ and Maxwell's equations offers us a way to relate $\mathbf{B}(\mathbf{r})$ and $\mathbf{M}(\mathbf{r}'')$. Moreover, the disk is a continuous medium of dipoles with spin. In fact, $\mathbf{M}(\mathbf{r}'')$ is directly caused by the spin angular momentum $\mathbf{S}(\mathbf{r}'')$, seen now as a continuous function of $\mathbf{r}''$. This is the so-called *macrospin approximation*, which brings immediately the following expression to relate $\mathbf{S}(\mathbf{r}'')$ and $\mathbf{M}(\mathbf{r}'')$:

$$\mathbf{M}(\mathbf{r}'') = \frac{g\mu_B}{\hbar} n(\mathbf{r}'') \mathbf{S}(\mathbf{r}'') ,$$

where g is the Landè g-factor (typically, $g \approx 2$), μ_B is the Bohr magneton and $n(\mathbf{r}'')$ is the volumetric density of dipoles. The last quantity can be expressed in terms of a *surface* density σ by using $n(\mathbf{r}'') =$

¹Do not confuse $\mathbf{B}(\mathbf{r})$ (the stray field generated by the sample) with $\mathbf{B}_{\text{NV}}(\mathbf{r})$ (the *total* magnetic field detected by the NV center). The last one considers both the stray field $\mathbf{B}(\mathbf{r})$ and the external magnetic field $\mathbf{H}_0$. In this work, we assume that $|\mathbf{H}_0| \gg |\mathbf{B}(\mathbf{r})|$, so $\mathbf{B}_{\text{NV}}(\mathbf{r}) \approx \mathbf{H}_0$.

$\sigma\delta(\mathbf{r}'')$. Therefore, the magnetization is related to a *spin surface density* $\mathbf{s}(\mathbf{r}'') = \sigma\mathbf{S}(\mathbf{r}'')$

$$\mathbf{M}(\mathbf{r}'') = \gamma\mathbf{s}(\mathbf{r}'')\delta(\mathbf{r}'') \quad (3.1)$$

where we have defined the sample gyromagnetic ratio $\gamma = g\mu_B/\hbar$. In this way, a spin fluctuation $\mathbf{s}(\mathbf{r}'') \rightarrow \mathbf{s}(\mathbf{r}'', t)$ at position $\mathbf{r}''$ and instant t in the sample is detected by the NV center as a time-dependent magnetic field $\mathbf{B}_{\text{NV}}(\mathbf{r}) \rightarrow \mathbf{B}_{\text{NV}}(\mathbf{r}) + \mathbf{B}(\mathbf{r}, t)$, which is exactly what we have discussed in Chapter 2 using quantum perturbation theory.

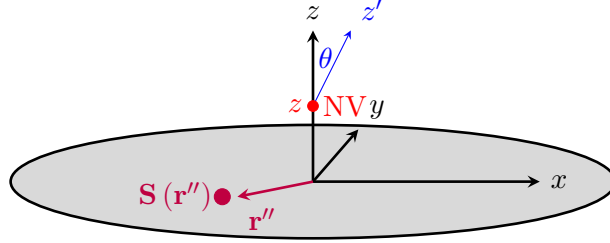


Figure 3.1: Disk modelling the thin film sample, along with the used coordinate system xyz . Note that this system is independent of the $x'y'z'$ axes used to describe the NV center.

We can relate both quantities (spin angular momentum and magnetic field) by using Maxwell's equations. To begin with, the magnetic vector potential $\mathbf{A}$ and the magnetization $\mathbf{M}$ follow the relation (in CGS units)

$$\mathbf{A}(\mathbf{r}) = \int \mathbf{M}(\mathbf{r}'') \times \frac{\mathbf{r} - \mathbf{r}''}{|\mathbf{r} - \mathbf{r}''|^3} d^3\mathbf{r}''.$$

Applying curl on both sides of this relation (knowing $\mathbf{B} = \nabla \times \mathbf{A}$), we get

$$\begin{aligned} \mathbf{B}(\mathbf{r}) &= \int \nabla \times \left(\mathbf{M}(\mathbf{r}'') \times \frac{\mathbf{r} - \mathbf{r}''}{|\mathbf{r} - \mathbf{r}''|^3} \right) d^3\mathbf{r}'' \\ \Rightarrow \mathbf{B}(\mathbf{r}) &= \int \left\{ \mathbf{M} \left(\nabla \cdot \frac{\mathbf{r} - \mathbf{r}''}{|\mathbf{r} - \mathbf{r}''|^3} \right) - (\mathbf{M} \cdot \nabla) \frac{\mathbf{r} - \mathbf{r}''}{|\mathbf{r} - \mathbf{r}''|^3} \right\} d^3\mathbf{r}'' \\ \Rightarrow \mathbf{B}(\mathbf{r}) &= \int \left\{ 4\pi\delta^3(\mathbf{r} - \mathbf{r}'') \mathbf{M}(\mathbf{r}'') - (\mathbf{M} \cdot \nabla) \nabla'' \frac{1}{|\mathbf{r} - \mathbf{r}''|} \right\} d^2\mathbf{r}'', \end{aligned} \quad (3.2)$$

and using the macrospin approximation (3.1) in (3.2), for $\mathbf{r} \neq \mathbf{r}''$,

$$\mathbf{B}(\mathbf{r}) = \gamma \int \left\{ -(\mathbf{s} \cdot \nabla) \nabla'' \frac{1}{|\mathbf{r} - \mathbf{r}''|} \right\} d^2\mathbf{r}''.$$

In a more compact form, this integral can be written as

$$\mathbf{B}(\mathbf{r}) = \gamma \int \mathbf{D}(\mathbf{r} - \mathbf{r}'') \cdot \mathbf{s}(\mathbf{r}'') d^2\mathbf{r}'', \quad (3.3)$$

being $\mathbf{D}(\mathbf{r} - \mathbf{r}') = -\nabla\nabla'|\mathbf{r} - \mathbf{r}'|^{-1}$ the dipole tensor. In the Cartesian system of coordinates xyz , it can be explicitly expressed as

$$D_{\alpha\beta}(\mathbf{r} - \mathbf{r}'') = -\partial_\alpha \partial_\beta'' \frac{1}{|\mathbf{r} - \mathbf{r}''|}, \quad (3.4)$$

which is to say,

$$D_{\alpha\beta}(\mathbf{r} - \mathbf{r}'') = \frac{3(x_\alpha - x''_\alpha)(x_\beta - x''_\beta)}{[(x - x'')^2 + (y - y'')^2 + (z - z'')^2]^{5/2}} \quad \text{if } \alpha \neq \beta$$

$$D_{\alpha\beta}(\mathbf{r} - \mathbf{r}'') = \frac{3(x_\alpha - x''_\alpha)(x_\beta - x''_\beta)}{[(x - x'')^2 + (y - y'')^2 + (z - z'')^2]^{5/2}} - \frac{1}{[(x - x'')^2 + (y - y'')^2 + (z - z'')^2]^{3/2}} \quad \text{if } \alpha = \beta.$$

The elements $D_{\alpha\beta}$ can be easily manipulated in the Fourier space, where differential operators just transform to algebraic products ($\mathcal{F}\{\partial_\alpha f\} = -ik_\alpha \mathcal{F}\{f\}$ if $\alpha \neq z$). Considering that we will need the two-dimensional Fourier transform $\mathcal{F}_{\mathbf{r}''}$, the needed identities here are

$$\begin{aligned} \mathcal{F}_{\mathbf{r}''} \left\{ \frac{1}{|\mathbf{r} - \mathbf{r}''|} \right\}(\mathbf{k}, z) &= 2\pi \frac{\exp(-k|z - z''|)}{k} \\ \mathcal{F}_{\mathbf{r}''} \left\{ \frac{\partial}{\partial z} \frac{1}{|\mathbf{r} - \mathbf{r}''|} \right\}(\mathbf{k}, z) &= -2\pi \exp(-k|z - z''|) \\ \mathcal{F}_{\mathbf{r}''} \left\{ \frac{\partial^2}{\partial z^2} \frac{1}{|\mathbf{r} - \mathbf{r}''|} \right\}(\mathbf{k}, z) &= 2\pi k \exp(-k|z - z''|) \end{aligned}$$

so

$$\begin{aligned} (\mathcal{F}D_{zz})(\mathbf{k}, z) &= 2\pi k \exp(-kz), \\ (\mathcal{F}D_{\alpha z})(\mathbf{k}, z) &= -2\pi i k_\alpha \exp(-kz) \quad \text{if } \alpha \neq z, \\ (\mathcal{F}D_{\alpha\beta})(\mathbf{k}, z) &= -2\pi k_\alpha k_\beta \frac{\exp(-kz)}{k} \quad \text{if } \alpha, \beta \neq z, \end{aligned}$$

where we have written the 2D Fourier transform just as $\mathcal{F}$ for short. Therefore,

$$\mathcal{F}\{\mathbf{D}\}(\mathbf{k}, z) = 2\pi k \exp(-kz) \begin{bmatrix} -\frac{k_x^2}{k^2} & -\frac{k_x k_y}{k^2} & -i \frac{k_x}{k} \\ -\frac{k_x k_y}{k^2} & -\frac{k_y^2}{k^2} & -i \frac{k_y}{k} \\ -i \frac{k_x}{k} & -i \frac{k_y}{k} & 1 \end{bmatrix}.$$

Or, in polar coordinates (defining $\cos \phi_k = k_x/k$ y $\sin \phi_k = k_y/k$),

$$\mathcal{F}\{\mathbf{D}\}(\mathbf{k}, z) = 2\pi k \exp(-kz) \begin{bmatrix} -\cos^2 \phi_k & -\frac{\sin 2\phi_k}{2} & -i \cos \phi_k \\ -\frac{\sin 2\phi_k}{2} & -\sin^2 \phi_k & -i \sin \phi_k \\ -i \cos \phi_k & -i \sin \phi_k & 1 \end{bmatrix}. \quad (3.5)$$

What we have in equation (3.3) is a convolution, whose Fourier transform results in a simple algebraic product:

$$\mathcal{F}\{\mathbf{B}\} = \gamma \mathcal{F}\{\mathbf{D}\} \mathcal{F}\{\mathbf{s}\}.$$

Finally, taking the inverse Fourier transform,

$$\mathbf{B}(\mathbf{r}) = \frac{\gamma}{(2\pi)^2} \int \mathcal{F}\{\mathbf{D}\}(\mathbf{k}, z) \cdot \mathcal{F}\{\mathbf{s}\}(\mathbf{k}) \cdot \exp(i\mathbf{k} \cdot \boldsymbol{\rho}) d^2\mathbf{k}, \quad (3.6)$$

where the components of $\mathcal{F}\{\mathbf{D}\}$ are given by equation (3.5). Strictly speaking, this integral is performed on the sample first Brillouin zone (1BZ). In the continuous limit, this 1BZ is the entire plane.

Result (3.6) shows how the spin density $\mathbf{s}(\mathbf{r}'')$ generates a stray magnetic field $\mathbf{B}(\mathbf{r})$ detected by the NV center at position $\mathbf{r}$. In particular, a spin fluctuation $\mathbf{s}(\mathbf{r}'', t)$ generates a time-dependent field fluctuation $\mathbf{B}(\mathbf{r}, t)$, whose components in the NV coordinate system $x'y'z'$ (remember that the z' and the z axes might form an angle θ) are exactly the ingredients that appear in equation (2.17). From now on, for simplicity, we write $\mathcal{F}\{\mathbf{D}\}(\mathbf{k}, z) \rightarrow \mathbf{D}(\mathbf{k}, z)$ and $\mathcal{F}\{\mathbf{s}\}(\mathbf{k}, t) \rightarrow \mathbf{s}(\mathbf{k}, t)$ and promote this quantity to quantum operator: $\mathbf{s}(\mathbf{r}, t) \rightarrow \hat{\mathbf{s}}(\mathbf{r}, t)$, which satisfies angular momentum algebra. Therefore, according to (3.6), field fluctuations $\left\{\hat{B}_j(\mathbf{r}, t)\right\}_{j=x,y,z}$ are also operators, whose components in the NV coordinate system $x'y'z'$ are

$$\hat{\mathbf{B}}'(t) = \frac{\gamma}{(2\pi)^2} \int \mathbf{R}_\theta \mathbf{D}(\mathbf{k}, z) \cdot \hat{\mathbf{s}}(\mathbf{k}, t) \cdot \exp(i\mathbf{k} \cdot \boldsymbol{\rho}) d^2\mathbf{k}, \quad (3.7)$$

where $\mathbf{R}_\theta$ is the rotation matrix needed for doing the rotation from the xyz coordinates to the $z'y'z'$. Remember that the magnetic field correlation $\langle \hat{B}'_j(t) \hat{B}'_j(0) \rangle$ requires the magnetic field components expressed in the NV coordinate system. Explicitly,

$$\mathbf{R}_\theta = \begin{bmatrix} \cos \theta & 0 & -\sin \theta \\ 0 & 1 & 0 \\ \sin \theta & 0 & \cos \theta \end{bmatrix}.$$

With this, one can calculate the field-field correlation $\langle \hat{B}'_j(t) \hat{B}'_j(0) \rangle$:

$$\langle \hat{B}'_j(t) \hat{B}'_j(0) \rangle = \frac{\gamma^2}{(2\pi)^4} \int \int R_{j\ell} D_{\ell n}(\mathbf{k}) R_{jp} D_{pm}(\mathbf{k}') \langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle \exp[i(\mathbf{k} + \mathbf{k}') \cdot \boldsymbol{\rho}] d^2\mathbf{k}' d^2\mathbf{k}. \quad (3.8)$$

where summation convention was used. It is possible to show that only the terms with $m = n$ contribute to the sum (see Appendix B), where (from now on, summation convention is no longer used)

$$\langle \hat{s}_n(\mathbf{k}, t) \hat{s}_n(\mathbf{k}', 0) \rangle = (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') \mathcal{F}_{\mathbf{R}=\mathbf{r}-\mathbf{r}''} \left\{ \langle \hat{s}_n(\mathbf{r}, t) \hat{s}_n(\mathbf{r}'', 0) \rangle \right\}(\mathbf{k}, t)$$

so, inserting this into equation (3.8), we obtain

$$\langle \hat{B}'_j(t) \hat{B}'_j(0) \rangle = \frac{\gamma^2}{(2\pi)^2} \sum_{n=x,y,z} \int N_{jn}(\mathbf{k}) \mathcal{F}_{\mathbf{R}=\mathbf{r}-\mathbf{r}'} \left\{ C_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) \right\}(\mathbf{k}, t) d^2\mathbf{k}, \quad (3.9)$$

where $\mathcal{F}_{\mathbf{R}}$ is the 2D Fourier transform operator with respect to the coordinate $\mathbf{R}$, $N_{jn}(\mathbf{k}, z) = |R_{j\ell} D_{\ell n}(\mathbf{k}, z)|^2$ and $C_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \langle \hat{s}_\alpha(\mathbf{r}, t) \hat{s}_\beta(\mathbf{r}', 0) \rangle$, for the coordinates $\alpha, \beta = x, y, z$. The explicit form of this matrix

$\mathbf{N}$ is

$$\mathbf{N} = (2\pi k)^2 \exp(-2kz) \times \begin{bmatrix} \cos^4 \phi_k \cos^2 \theta + \cos^2 \phi_k \sin^2 \theta & \frac{\sin^2 2\phi_k \cos^2 \theta}{4} + \sin^2 \phi_k \sin^2 \theta & \cos^2 \phi_k \cos^2 \theta + \sin^2 \theta \\ \frac{\sin^2 2\phi_k}{4} & \sin^4 \phi_k & \sin^2 \phi_k \\ \cos^4 \phi_k \sin^2 \theta + \cos^2 \phi_k \cos^2 \theta & \sin^2 \phi_k \cos^2 \theta + \frac{\sin^2 2\phi_k \sin^2 \theta}{4} & \cos^2 \theta + \cos^2 \phi_k \sin^2 \theta \end{bmatrix}. \quad (3.10)$$

This is the desired expansion that relates the field correlation functions $\langle \hat{B}'_j(t) \hat{B}'_j(0) \rangle$ with the spin-spin correlations $C_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \langle \hat{s}_\alpha(\mathbf{r}, t) \hat{s}_\beta(\mathbf{r}', 0) \rangle$. Particularly, C_{xx} and C_{yy} are known as the *transverse spin-spin correlations*, while C_{zz} is the *longitudinal correlation*.

Finally, inserting the result (3.9) into the relaxation rates (2.17) found in Chapter 2, we obtain a first formula that allows us to calculate $\Gamma(\omega)$, for a resonance frequency ω . From now on, unless otherwise, we will refer to the (2+1)D Fourier transform of the spin-spin correlations $\mathcal{F}_{(\mathbf{R}, t)} \{C_{\alpha\beta}(\mathbf{R}, t)\}(\mathbf{k}, \omega)$ just as $C_{\alpha\beta}(\mathbf{k}, \omega)$:

$$\Gamma(\omega) = \frac{(\gamma\gamma_{\text{NV}})^2}{2(2\pi)^2} \int \{N_{xx}(\mathbf{k}) C_{xx}(\mathbf{k}, \omega) + N_{xy}(\mathbf{k}) C_{yy}(\mathbf{k}, \omega) + N_{xz}(\mathbf{k}) C_{zz}(\mathbf{k}, \omega) + N_{yx}(\mathbf{k}) C_{xx}(\mathbf{k}, \omega) + N_{yy}(\mathbf{k}) C_{yy}(\mathbf{k}, \omega) + N_{yz}(\mathbf{k}) C_{zz}(\mathbf{k}, \omega)\} d^2\mathbf{k}.$$

Replacing the explicit elements of the matrix $\mathbf{N}(\mathbf{k}, z)$ found in equation (3.10),

$$\begin{aligned} \Gamma(\omega) = \frac{(\gamma\gamma_{\text{NV}})^2}{2} \int d^2\mathbf{k} k^2 \exp(-2kz) \{ & \left(\cos^4 \phi_k \cos^2 \theta + \cos^2 \phi_k \sin^2 \theta + \frac{\sin^2 2\phi_k}{4} \right) C_{xx}(\mathbf{k}, \omega) \\ & + \left(\frac{\sin^2 2\phi_k \cos^2 \theta}{4} + \sin^2 \phi_k \sin^2 \theta + \sin^4 \phi_k \right) C_{yy}(\mathbf{k}, \omega) \\ & + (\cos^2 \phi_k \cos^2 \theta + \sin^2 \theta + \sin^2 \phi_k) C_{zz}(\mathbf{k}, \omega) \} \end{aligned} \quad (3.11)$$

This is the most general form of the relaxation rates $\Gamma(\omega)$ in terms of the spin-spin correlation functions $C_{\alpha\beta}(\mathbf{k}, \omega)$ in the Fourier space. Symmetries are the key to reduce this expression. For instance, a $U(1)$ -symmetric system guarantees that the transverse spin-spin correlations are equal: $C_{xx} = C_{yy}$, which is the case we treat during the following chapters. Also, we assume that $C_{\alpha\beta}$ depend only on the wavevector length $k = |\mathbf{k}|$, which is always valid for isotropic systems and some anisotropic systems we will discuss later.

With these assumptions in mind, we can write $d^2\mathbf{k} = k dk d\phi_k$ and realize the integral over ϕ_k . Note

that

$$\begin{aligned} \int_0^{2\pi} d\phi_k \left(\cos^4 \phi_k \cos^2 \theta + \cos^2 \phi_k \sin^2 \theta + \frac{\sin^2 2\phi_k}{4} \right) &= \frac{\pi}{8} [11 - 3 \cos(2\theta)] \\ \int_0^{2\pi} d\phi_k \left(\frac{\sin^2 2\phi_k \cos^2 \theta}{4} + \sin^2 \phi_k \sin^2 \theta + \sin^4 \phi_k \right) &= \frac{\pi}{8} [9 - \cos(2\theta)] \\ \int_0^{2\pi} d\phi_k (\cos^2 \phi_k \cos^2 \theta + \sin^2 \theta + \sin^2 \phi_k) &= \frac{\pi}{2} [5 - \cos(2\theta)], \end{aligned}$$

so the sum of the first two integrals is exactly the last one. Therefore, the reduced expression for the relaxation rate is [26]

$$\boxed{\Gamma(\omega) = f(\theta) \int_0^\infty dk k^3 e^{-2kz} [C_{xx}(k, \omega) + C_{zz}(k, \omega)]} \quad (3.12)$$

where $f(\theta) = \pi(\gamma\gamma_{\text{NV}})^2(5 - \cos 2\theta)/4$. Originally the integral in equation (3.11) is defined on the first Brillouin zone (1BZ), but the filter function $k^3 \exp(-2kz)$ in the integrand suppresses all the contributions for large $k \gg 1/z$, including those near the edges of the 1BZ (as $a \sim 1 \text{ nm}$ and $z \sim 10 \text{ nm}$, we have $1/z \gg \pi/a$). Therefore, we are not making a significant error by extending the integration limits. Here, $C_{\alpha\beta}(\mathbf{k}, \omega)$ is the Fourier transform of the spin-spin correlation function $C_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \langle \hat{s}_\alpha(\mathbf{r}, t) \hat{s}_\beta(\mathbf{r}', 0) \rangle$, for the coordinates $\alpha, \beta = x, y, z$. So far, all this procedure is general and does not depend on the physical characteristics of the sample. All the information of the sample is implicitly content in the correlators $C_{\alpha\beta}$.

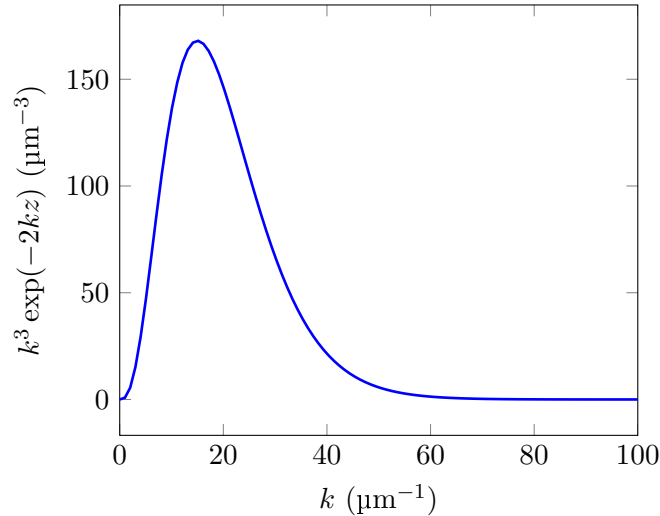


Figure 3.2: Filter function $k^3 \exp(-2kz)$ versus wave number k in the 1BZ, for $z = 100 \text{ nm} = 0.1 \mu\text{m}$.

Note that the correlations $C_{\alpha\beta}$ are defined using the canonical average $\langle \dots \rangle$ out of equilibrium (considering also the time-dependent spin fluctuations), so equilibrium statistical mechanics tools, such as

the use of the partition function, are no longer valid in this context. The strategy here is to use the fluctuation-dissipation theorem [27], which establishes that, at temperature $T > 0$,

$$C_{\alpha\beta}(\mathbf{k}, \omega) = \frac{2\hbar}{1 - \exp\left(-\frac{\hbar\omega}{k_B T}\right)} \chi''_{\alpha\beta}(\mathbf{k}, \omega), \quad (3.13)$$

where k_B is the Boltzmann constant and $\chi''_{\alpha\beta}$ is the imaginary part of the linear response function $\chi_{\alpha\beta}$, given in the real space by the Kubo formula [28]

$$\chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \frac{i}{\hbar} \theta(t) \langle [\hat{s}_\alpha(\mathbf{r}, t), \hat{s}_\beta(\mathbf{r}', 0)] \rangle_{eq}, \quad (3.14)$$

being $\langle \dots \rangle_{eq}$ the canonical average in thermal equilibrium (without time-dependent fluctuations). In this way, the calculation for correlators $C_{\alpha\beta}$ can be reduced to finding the linear response function $\chi_{\alpha\beta}$ through the spin-spin commutator. These spin density operators $\hat{\mathbf{s}}(\mathbf{r}, t)$ contain the the physical information of the magnetic sample.

In summary, we developed in this chapter a model to relate the time-dependent field fluctuations, detected by the NV center, with spin fluctuations produced in the material. These spin fluctuations can be produced, for example, by an external driving field that perturbrates at a certain instant the spin system. We modelled the sample as a 2D disk of spin magnetic dipoles displayed continuously (macrospin approximation), and using classical electrodynamics, we related the detected stray field with the spin density function $\mathbf{s}(\mathbf{r}'', t)$. As a result, the desired relaxation rates depend on the spin-spin correlation functions, so finding these correlators is crucial in order to calculate $\Gamma(\omega_{\pm})$. These functions can be calculated using the fluctuation-dissipation theorem, and to do that, it is necessary to describe the physical parameters of the sample.

Chapter 4

Ferromagnetic systems

In this chapter, we introduce a first kind of materials to consider in the calculation of the relaxation rate from equation (3.12). Ferromagnetic systems present long-range spin ordering, which can be explained using the Heisenberg spin-spin hamiltonian [2]. In this way, spins interact between them, and oscillations can propagate along the material through the so-called *spin waves*. In this chapter, we introduce spin waves in ferromagnetic insulators, present their quanta (magnons) and find their dispersion relation. As we are considering only insulator systems, we neglect presence of free electric charges in the material.

4.1 Ferromagnetic hamiltonian

According to Heisenberg's model, two atoms, molecules or dipoles with spins (operators) $\hat{\mathbf{S}}(\mathbf{r}_1)$ and $\hat{\mathbf{S}}(\mathbf{r}_2)$ can interact between them by electrostatic forces, whose energy, due to the anti-symmetry of fermionic wave functions, can be expressed as a simple dot product $-2J_{12}\hat{\mathbf{S}}(\mathbf{r}_1) \cdot \hat{\mathbf{S}}(\mathbf{r}_2)/\hbar^2$. Here, J_{12} is the *exchange parameter* or *coupling constant*, and its sign determines if the interaction is ferro- ($J_{12} > 0$) or antiferromagnetic ($J_{12} < 0$). In this chapter, we focus on ferromagnetic interactions, where all the possible coupling constants are positive or zero. With this in mind, one can build the sample hamiltonian:

$$\hat{H}_s = -\gamma \sum_{\mathbf{r}_i} \mathbf{H}_0 \cdot \hat{\mathbf{S}}(\mathbf{r}_i) - \sum_{\mathbf{r}_i} \sum_{\mathbf{r}_j \neq \mathbf{r}_i} \frac{J_{ij}}{\hbar^2} \hat{\mathbf{S}}(\mathbf{r}_i) \cdot \hat{\mathbf{S}}(\mathbf{r}_j), \quad (4.1)$$

where $\mathbf{H}_0$ is the external magnetic field, which for simplicity we will assume to be parallel to the sample perpendicular axis $\hat{\mathbf{z}}$. The first summation corresponds to the net Zeeman effect on the sample, while the double summation comes from the spin-spin interaction. In this toy model, we won't consider anisotropy yet.

One can characterize spin waves in many ways [2]. Here, we treat the quantum approach consisting

of diagonalizing the hamiltonian (4.1) in the Fourier space. First, we introduce the raising and lowering spin operators

$$\begin{aligned}\hat{S}_j^- &= \hat{S}_{j,x} - i\hat{S}_{j,y}, \\ \hat{S}_j^+ &= \hat{S}_{j,x} + i\hat{S}_{j,y},\end{aligned}$$

where we used the subindex j enumerates the dipole located at $\mathbf{r}_j$. In this way, we used the simplified notation $\hat{S}^-(\mathbf{r}_j) \rightarrow \hat{S}_j^-$ and $\hat{S}^+(\mathbf{r}_j) \rightarrow \hat{S}_j^+$. These operators transform the individual state $|s_{jz}\rangle$ of the j particle as

$$\begin{aligned}\hat{S}_j^- |s_{jz}\rangle &= \hbar\sqrt{s(s+1) - s_{jz}(s_{jz}+1)} |s_{jz}-1\rangle \\ \hat{S}_j^+ |s_{jz}\rangle &= \hbar\sqrt{s(s+1) - s_{jz}(s_{jz}-1)} |s_{jz}+1\rangle\end{aligned}$$

where s is the spin quantum number of each dipole (for example, $s = 1/2$). In order to know which creation-annihilation we should define here, it is convenient to establish the “spin deviation operator”

$$\hat{n}_j = s - \hat{S}_{jz}/\hbar$$

with associated quantum number

$$n_j = s - s_{jz}.$$

In terms of $\hat{n}_j$, the action of $\hat{S}_j^+$ and $\hat{S}_j^-$ on $|n_j\rangle$ is

$$\hat{S}_j^+ |n_j\rangle = \hbar\sqrt{2s}\sqrt{n_j}\sqrt{1 - \frac{n_j-1}{2s}} |n_j-1\rangle \quad (4.2)$$

$$\hat{S}_j^- |n_j\rangle = \hbar\sqrt{2s}\sqrt{n_j+1}\sqrt{1 - \frac{n_j}{2s}} |n_j+1\rangle \quad (4.3)$$

We know that the raising and lowering operators $\hat{a}_j^\dagger$ y $\hat{a}_j$ must satisfy

$$\begin{aligned}[\hat{a}_i, \hat{a}_j^\dagger] &= \delta_{ij} \\ [\hat{a}_i, \hat{a}_j] &= [\hat{a}_i^\dagger, \hat{a}_j^\dagger] = 0 \\ \hat{a}_j^\dagger |n_j\rangle &= \sqrt{n_j+1} |n_j+1\rangle \\ \hat{a}_j |n_j\rangle &= \sqrt{n_j} |n_j-1\rangle\end{aligned}$$

So, equations (4.2) and (4.3) suggest to define the operators $\hat{a}_j^\dagger$ and $\hat{a}_j$ such that

$$\begin{aligned}\hat{S}_j^+ &= \hbar\sqrt{2s} \left(1 - \frac{\hat{a}_j^\dagger \hat{a}_j}{2s}\right)^{1/2} \hat{a}_j \\ \hat{S}_j^- &= \hbar\sqrt{2s} \hat{a}_j^\dagger \left(1 - \frac{\hat{a}_j^\dagger \hat{a}_j}{2s}\right)^{1/2} \\ \hat{S}_{jz} &= \hbar \left(s - \hat{a}_j^\dagger \hat{a}_j\right)\end{aligned} \quad (4.4)$$

These are the *Holstein-Primakoff transformations* [29]. Explicitly, these square roots are expanded in power series as

$$\begin{aligned}\hat{S}_j^+ &= \hbar\sqrt{2s} \left(1 - \frac{\hat{a}_j^\dagger \hat{a}_j}{4s} + \dots \right) \hat{a}_j \approx \hbar\sqrt{2s} \hat{a}_j \\ \hat{S}_j^- &= \hbar\sqrt{2s} \hat{a}_j^\dagger \left(1 - \frac{\hat{a}_j^\dagger \hat{a}_j}{4s} + \dots \right) \approx \hbar\sqrt{2s} \hat{a}_j^\dagger \\ \hat{S}_{jz} &= \hbar \left(s - \hat{a}_j^\dagger \hat{a}_j \right)\end{aligned}$$

Here, the approximations were used at room temperature ($T \approx 300$ K), where one can assume that all spin states have similar occupation probability.

With the latest formalism, we can write the sample hamiltonian (4.1) as

$$\begin{aligned}\hat{H}_s &= -\hbar\gamma H_0 \sum_j \left(s - \hat{a}_j^\dagger \hat{a}_j \right) - \frac{1}{2} \sum_j \sum_{i \neq j} \frac{J_{ij}}{\hbar^2} \left(\hat{S}_j \hat{S}_i^\dagger + \hat{S}_j^\dagger \hat{S}_i + 2\hat{S}_{jz} \hat{S}_{iz} \right) \\ \Rightarrow \hat{H}_s &= -\hbar\gamma H_0 \sum_j \left(s - \hat{a}_j^\dagger \hat{a}_j \right) - \frac{1}{2} \sum_j \sum_{i \neq j} J_{ij} \left[2s \hat{a}_j^\dagger \hat{a}_i + 2s \hat{a}_j \hat{a}_i^\dagger + 2 \left(s - \hat{a}_j^\dagger \hat{a}_j \right) \left(s - \hat{a}_i^\dagger \hat{a}_i \right) \right] \\ \Rightarrow \hat{H}_s &= -\hbar\gamma H_0 \sum_j \left(s - \hat{a}_j^\dagger \hat{a}_j \right) - s \sum_j \sum_{i \neq j} J_{ij} \left(\hat{a}_j^\dagger \hat{a}_i + \hat{a}_j \hat{a}_i^\dagger - \hat{a}_i^\dagger \hat{a}_i - \hat{a}_j^\dagger \hat{a}_j + s \right).\end{aligned}$$

Now we return to the original notation $\hat{a}_j^\dagger \rightarrow \hat{a}^\dagger(\mathbf{r}_j)$ and $\hat{a}_j \rightarrow \hat{a}(\mathbf{r}_j)$, being $\mathbf{r}_j$ the position of the dipole j . This *field notation* is useful for considering only interaction between nearest neighbors, which is to say:

$$J_{ij} = \begin{cases} J > 0 & \text{if } |\mathbf{r}_i - \mathbf{r}_j| = a, \\ 0 & \text{any other case.} \end{cases}$$

With this in mind, the hamiltonian is

$$\begin{aligned}\hat{H}_s &= -\hbar\gamma H_0 \sum_{\mathbf{r}} \left[s - \hat{a}^\dagger(\mathbf{r}) \hat{a}(\mathbf{r}) \right] - s \sum_{\mathbf{r}} \sum_{\boldsymbol{\delta}} J[\hat{a}^\dagger(\mathbf{r}) \hat{a}(\mathbf{r} + \boldsymbol{\delta}) + \hat{a}(\mathbf{r}) \hat{a}^\dagger(\mathbf{r} + \boldsymbol{\delta}) \\ &\quad - \hat{a}^\dagger(\mathbf{r} + \boldsymbol{\delta}) \hat{a}(\mathbf{r} + \boldsymbol{\delta}) - \hat{a}^\dagger(\mathbf{r}) \hat{a}(\mathbf{r}) + s] \quad (4.5)\end{aligned}$$

The idea is to diagonalize this hamiltonian. To do that, we use the periodicity of the sample lattice to expand the operators $\hat{a}_j$ y $\hat{a}_j^\dagger$ in the Fourier space (as if they were classical quantities)

$$\begin{aligned}\hat{a}_j^\dagger &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k} \in \text{1BZ}} \exp(-i\mathbf{k} \cdot \mathbf{r}_j) \hat{a}_{\mathbf{k}}^\dagger \\ \hat{a}_j &= \frac{1}{\sqrt{N}} \sum_{\mathbf{k} \in \text{1BZ}} \exp(i\mathbf{k} \cdot \mathbf{r}_j) \hat{a}_{\mathbf{k}},\end{aligned}$$

where the coefficients $\hat{a}_{\mathbf{k}}$ are given by

$$\hat{a}_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_j \hat{a}_j \exp(-i\mathbf{k} \cdot \mathbf{r}_j).$$

The commutation relations of these new $a_{\mathbf{k}}$ are

$$\begin{aligned} [\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}}^\dagger] &= \frac{1}{N} \sum_j \hat{a}_j \exp(-i\mathbf{k} \cdot \mathbf{r}_j) \sum_p \hat{a}_p^\dagger \exp(i\mathbf{k} \cdot \mathbf{r}_p) - \frac{1}{N} \sum_p \hat{a}_p^\dagger \exp(i\mathbf{k} \cdot \mathbf{r}_p) \sum_j \hat{a}_j \exp(-i\mathbf{k} \cdot \mathbf{r}_j) \\ \Rightarrow [\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}}^\dagger] &= \frac{1}{N} \sum_j \sum_p \left[\hat{a}_j \hat{a}_p^\dagger \exp i\mathbf{k} \cdot (\mathbf{r}_p - \mathbf{r}_j) - \hat{a}_p^\dagger \hat{a}_j \exp i\mathbf{k} \cdot (\mathbf{r}_p - \mathbf{r}_j) \right] \\ \Rightarrow [\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}}^\dagger] &= 1, \end{aligned}$$

so we have a bosonic commutation relation:

$$[\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}. \quad (4.6)$$

To diagonalize the hamiltonian (4.5), it is necessary to write each summation on $\mathbf{r}$ as summations on $\mathbf{k}$:

$$\hat{H}_s = -\hbar\gamma H_0 N s - JNZs^2 - \sum_{\mathbf{k} \in 1\text{BZ}} \left\{ -\hbar\gamma H_0 + Js \left[\sum_{\boldsymbol{\delta}} \exp i\mathbf{k} \cdot \boldsymbol{\delta} - 2z + \sum_{\boldsymbol{\delta}} \exp(-i\mathbf{k} \cdot \boldsymbol{\delta}) \right] \right\} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}},$$

where $Z = \sum_{\boldsymbol{\delta}} 1$ is the number of nearest neighbors of each dipole. If we define the lattice structure factor

$$\gamma_{\mathbf{k}} = \frac{1}{Z} \sum_{\boldsymbol{\delta}} \exp i\mathbf{k} \cdot \boldsymbol{\delta}$$

and the dispersion relation

$$\boxed{\hbar\omega_{\mathbf{k}} = 2JZs(1 - \gamma_{\mathbf{k}}) + \hbar\gamma H_0} \quad (4.7)$$

we obtain finally

$$\hat{H}_s = \sum_{\mathbf{k} \in 1\text{BZ}} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}} \hbar\omega_{\mathbf{k}}$$

where the residual constant terms were omitted, and we have assumed central symmetry ($\gamma_{\mathbf{k}} = \gamma_{-\mathbf{k}}$). This is the desired diagonal form of the hamiltonian (4.1), in terms of *number operators* $\hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}}$. This form is interpreted as follows: for each normal mode $\mathbf{k}$ of spin oscillations, we have $\hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}}$ bosonic-type particles, namely *magnons*, and each of them contributes to the system an energy $\hbar\omega_{\mathbf{k}}$, given by the dispersion relation (4.7). The operator $\hat{a}_{\mathbf{k}}^\dagger$ creates a magnon associated to the normal mode state $|\mathbf{k}\rangle_1$ (interpreted as a state with momentum $\hbar\mathbf{k}$)¹

$$\hat{a}_{\mathbf{k}}^\dagger |0\rangle = |\mathbf{k}\rangle$$

¹The subindex 1 in the state $|\mathbf{k}\rangle_1$ refers to an *individual state*, which characterizes the state of one particle at a time. Don't confuse with $|\mathbf{k}\rangle$ (without subindex), which actually should be written as $|0 \ 0 \ \dots \ 1 \ 0 \ 0 \ \dots\rangle$, where that 1 locates at the slot of the $\mathbf{k}$ mode state and indicates that the individual state $|\mathbf{k}\rangle_1$ is being occupied by exactly 1 particle. By the way, the vacuum state $|0\rangle$ (without subindex) refers to $|0 \ 0 \ 0 \ \dots\rangle$, being all its inner numbers 0.

while the operator $\hat{a}_{\mathbf{k}}$ annihilates it:

$$\hat{a}_{\mathbf{k}} |\mathbf{k}\rangle = |0\rangle$$

As bosons, magnon creation and annihilation operators satisfy the commutation relation (4.6).

As a practical example for calculating the dispersion relation, consider a 3D cubical lattice ($Z = 6$) with lattice constant a ,

$$\hbar\omega_{\mathbf{k}} = 2JZs \left[1 - \frac{1}{6} \cdot (2 \cos k_x a + 2 \cos k_y a + 2 \cos k_z a) \right] + \hbar\gamma H_0.$$

For small wave vectors ($ka \ll 1$), we have a parabolic dispersion relation

$$\hbar\omega_{\mathbf{k}} \approx 2Jsa^2 k^2 + \hbar\gamma H_0 \quad (4.8)$$

which can be written as a non-relativistic energy with external potential $\hbar\gamma H_0$:

$$\hbar\omega_{\mathbf{k}} \approx \frac{\hbar^2 k^2}{2m} + \hbar\gamma H_0$$

where $m = \hbar^2 / (4Jsa^2)$ is the magnon effective mass. This individual energy presents a gap $\hbar\gamma H_0$, which increases as the magnetic field H_0 does (see Figure 4.1). When $H_0 \sim 100$ Oe, this gap is of the order of GHz.

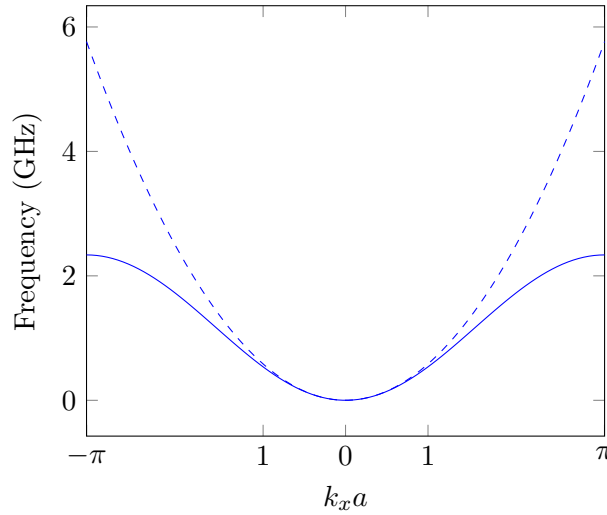


Figure 4.1: Magnon dispersion relation $\omega_{\mathbf{k}}/(2\pi)$ versus ka in one dimension ($k = k_x$), according to equations (4.7) (solid line, exact relation) and (4.8) (dashed line, quadratic approximation). Here, we used $J = 2.42$ meV and $H_0 = 100$ Oe.

4.2 Anisotropy in ferromagnetic systems

In the previous section, we only considered the simplest magnetic system, whose energy contributions were the Zeeman effect and the electrostatic coupling between spins. Here, the Zeeman effect tells us that the macroscopic magnetization vector $\mathbf{M}$ tend to align parallelly to the external magnetic field $\mathbf{H}_0$, which is expressed in the energy per unit volume (in CGS units) as a dot product, which has to be minimum at equilibrium

$$u_{Zeeman} = -\mathbf{M} \cdot \mathbf{H}_0. \quad (4.9)$$

However, we completely ignored the fact that magnetic dipoles can also interact *magnetically* between them, giving raise to a new term in the total energy density. This energy was already deduced for the case of NV centers; the zero-field-splitting term in the hamiltonian (2.7) has its origins just in dipole-dipole magnetic interaction (see Appendix A), so one can anticipate that this energy has to be proportional to M_z^2 :

$$u_D = 2\pi M_z^2. \quad (4.10)$$

In magnetic materials, this term is often known as the *demagnetizing* energy.

Besides the demagnetizing term, one can also find intrinsic uniaxial anisotropy terms that are pretty common in thin films and can be induced by the substrate on which they are grown. These terms tend to create an easy-axis of anisotropy [30], and have the form

$$u_A = -\frac{K^\perp}{M_S^2} M_z^2, \quad (4.11)$$

being $K^\perp > 0$ and M_S the saturation magnetization. Putting together the contributions (4.9), (4.10) and (4.11), one gets the total energy per unit volume, in the macrospin approximation:

$$u = -\mathbf{M} \cdot \mathbf{H}_0 + \frac{1}{2M_S} \left(4\pi M_S - \frac{2K^\perp}{M_S} \right) M_z^2$$

Here, we define the *effective magnetization* $4\pi M_{eff} = 4\pi M_S - 2K^\perp/M_S$. Also, keeping the module of $\mathbf{M}$ as $|\mathbf{M}| = M_S$, one can identify the angle θ_M between the vectors $\mathbf{M}$ and the sample normal, and the angle θ_H of $\mathbf{H}_0$. In this way, the previous equation is written as

$$\frac{u(\theta_M)}{M_S} = -H_0 \cos(\theta_M - \theta_H) + 2\pi M_{eff} \cos^2(\theta_M) \quad (4.12)$$

One can predict the equilibrium value of the magnetization angle θ_M minimizing this expression, which is to say, by solving the equation

$$\begin{aligned} \frac{du}{d\theta_M} \Big|_{\theta_M=\theta_M^*} &= 0 \\ \Rightarrow H_0 \sin(\theta_M^* - \theta_H) - 4\pi M_{eff} \cos(\theta_M^*) \sin(\theta_M^*) &= 0. \end{aligned}$$

In fact, this equation can be easily solved when $\theta_H = 0$ (magnetic field $\mathbf{H}_0$ parallel to $\hat{\mathbf{z}}$):

$$\theta_M^* = \arccos \frac{H_0}{4\pi M_{eff}}.$$

In this way, the effective magnetization M_{eff} defines if this quantity is truly a minimum or not. For instance, if $M_{eff} > 0$, this value of θ_M^* minimizes equation (4.12), and for $H_0 = 0$ we obtain $\theta_M^* = \pi/2$, which is to say, at zero field the magnetization is parallel to the sample plane. However, if $M_{eff} < 0$, this θ_M^* is not the global minimum of the function. One can check that $u(\theta_M = 0) < u(\theta_M = \theta_M^*)$, so the actual situation that minimizes the energy is when the magnetization $\mathbf{M}$ points perpendicular to the sample plane.

The main effect of the magnetization is the modification of the magnetic field that is detected by each dipole *inside* the sample. This magnitude is not H_0 anymore, but it is now an *internal field* B_M given by the system of equations [31]

$$B_M \cos(\theta_M) = H_0 \cos(\theta_H) - 4\pi M_{eff} \cos(\theta_M) \quad (4.13)$$

$$B_M \sin(\theta_M) = H_0 \sin(\theta_H), \quad (4.14)$$

where θ_M is the angle between $\mathbf{M}$ and the normal axis z (see Figure 4.2).

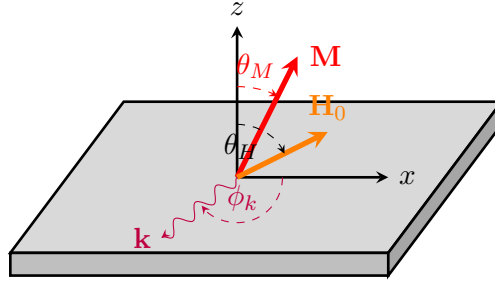


Figure 4.2: Magnetization vector $\mathbf{M}$ and external magnetic field $\mathbf{H}_0$ in the Cartesian xyz coordinate system. The wavy line on the xy represents an arbitrary magnon with momentum $\hbar\mathbf{k}$ in the thin film.

Equations (4.13) and (4.14) correspond to a system of equations with unknown variables θ_M and B_M . It is possible to reduce them into only one trigonometric relation $H_0 \cot(\theta_M) \sin(\theta_H) + 4\pi M_{eff} \cos(\theta_H) = H_0 \cos(\theta_H)$ by dividing (4.13) with (4.14). Taking as variable $x = \cos(\theta_M)$ and rewriting $\sin(\theta_M) = \sqrt{1 - x^2}$, one obtains a polynomial equation of fourth grade for x . Numerical solutions of this equation are calculated for each H_0 , and the associated values for $\theta_M = \theta_M(H_0, \theta_H)$ and $B_M = B_M(H_0, \theta_H)$ are illustrated in Figure 4.3. We observe that, if $M_{eff} > 0$, the magnetization vector $\mathbf{M}$ tends to point parallelly ($\theta_0 = \pi/2$ rad) to the film plane, but if $M_{eff} < 0$, the vector $\mathbf{M}$ points perpendicular to the thin film at zero magnetic field, as expected.

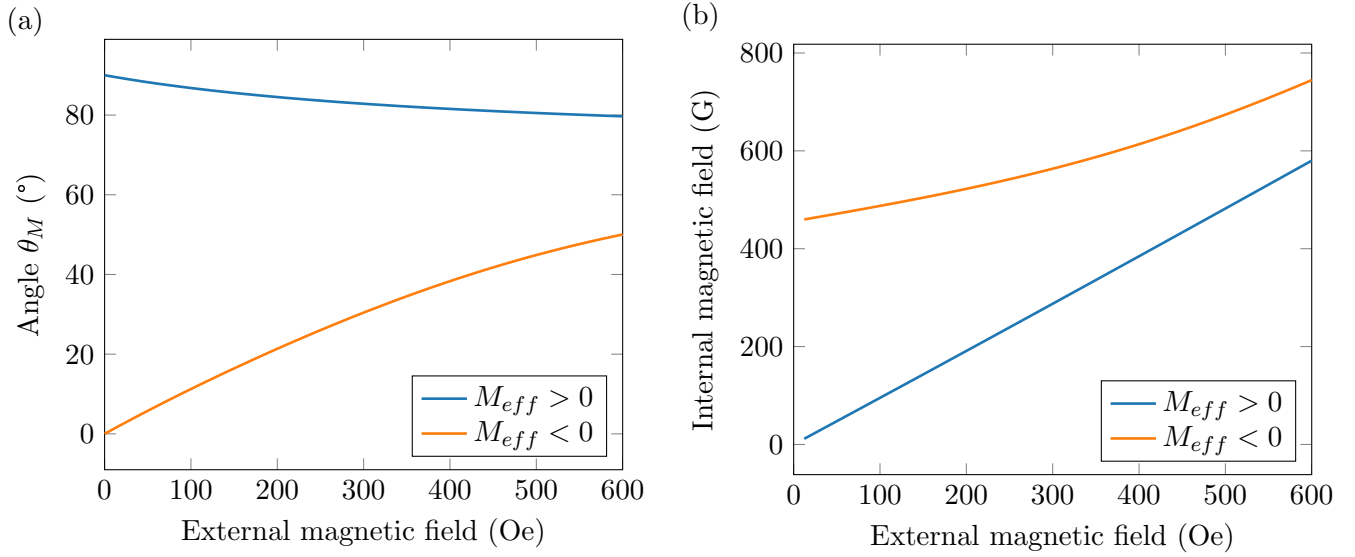


Figure 4.3: Graphic solution of the system of equations (4.13)–(4.14), taking the field angle as $\theta_H = 2\pi/5$ and the effective magnetization such that $4\pi|M_{eff}| = 456$ G. (a) Magnetization angle θ_M versus external magnetic field H_0 , both for positive effective magnetization and for negative. (b) Internal field B_M versus external magnetic field H_0 , using here the found solution for $\theta_M = \theta_M(H_0, \theta_H)$.

When anisotropy and demagnetizing effects are added to the ferromagnetic hamiltonian in equation (4.1), one must realize some extra steps in order to diagonalize this new hamiltonian and to find the modified magnon energies $\hbar\omega_{\mathbf{k}}$ [31]. In this case, the dispersion relation turns out to be

$$\hbar\omega_{\mathbf{k}} = \sqrt{(2Jsa^2k^2 + \hbar\gamma B_M)(2Jsa^2k^2 + \hbar\gamma B_M + 4\pi\hbar\gamma M_{eff}F(k, \phi_k))}, \quad (4.15)$$

where ϕ_k is the angle between the wave vector $\mathbf{k}$ and the x axis (as shown in Figure 4.3). Similarly to what we have done in equation (4.8), a 3D cubical lattice ($Z = 6$) was assumed and $ka \ll 1$. Note that, when $M_{eff} = 0$, this expression reduces to the simplified form (4.8), as expected. In the demagnetizing term $4\pi\gamma M_{eff}F(k, \phi_k)$, the anisotropy correction factor $F(k, \phi_k)$ is given by

$$F(k, \phi_k) = P(k) + \sin^2(\theta_0) \left[1 - P(k) (1 + \cos^2 \phi_k) + 4\pi\hbar\gamma M_{eff} \frac{P(k)(1 - P(k)) \sin^2 \phi_k}{2Jsa^2k^2 + \hbar\gamma B_M} \right]$$

with

$$P(k) = 1 - \frac{1 - \exp(-kt_{\text{film}})}{kt_{\text{film}}}.$$

being t_{film} the film thickness. This expression is useful for describing magnons in typical anisotropic materials like $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (Yttrium-Iron-Garnet, or YIG for short) [17], which is commonly treated as a ferromagnetic system and stands out for its low dissipation of spin waves.

To sum up, ferromagnetic materials are good candidates to take place of the dipole system model developed in Chapter 3. The goal here was to indicate how spins interact between them in this kind of systems, and we did it using the hamiltonian (4.1) which involucrates Heisenberg's spin-spin interaction and Zeeman effect. A collective deviation of spins is propagated in ferromagnetic systems as spin waves, whose quanta (magnons) are bosonic-type quasiparticles which obey the commutation relation $[\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$. Spin densities and magnon creation-annihilation operators are related by the HP transformations. All this information about magnon operators is useful to calculate the spin-spin correlation function introduced in Chapter 3, using the fluctuation-dissipation theorem –which requires to compute a spin-spin commutator. Also, we discussed about the magnon energies $\hbar\omega_{\mathbf{k}}$, whose gap is of the order of GHz (in frequency units), and how they are altered when anisotropies are present. We expect the desired relaxation rates to depend on these magnon energies, and therefore one may experimentally obtain parameters like the effective magnetization M_{eff} , the saturation magnetization M_S or the spin-spin exchange constant J just by fitting the theoretical relaxation rates in a graph of $\Gamma_{\pm}$ versus external magnetic field H_0 .

Chapter 5

Antiferromagnetic systems

In this chapter, we introduce a second kind of materials to consider in the calculation of the relaxation rate from equation (3.12). As mentioned in Chapter 1, antiferromagnetic systems have no net magnetization, but can present spin fluctuations in the form of spin waves. Here, we describe those spin waves, their quantization (magnons) and how they differ according to the anisotropy the material has.

5.1 Antiferromagnetic magnons

Ferromagnetic material scanning has already been developed by other authors [17, 32], so this work we focuses especially in antiferromagnetic insulators (AFI). The most common way to model a sample like that is to consider it as a two-sublattice system [19]: a L_1 sublattice with its spins tending to point parallelly to $\hat{\mathbf{z}}$, and the other L_2 with its spins parallel to $-\hat{\mathbf{z}}$. In general, a hamiltonian describing such system is of the form

$$\hat{H}_s = \sum_{\mathbf{k} \in 1\text{BZ}} \left(\hbar\omega_{\alpha\mathbf{k}} \hat{\alpha}_{\mathbf{k}}^\dagger \hat{\alpha}_{\mathbf{k}} + \hbar\omega_{\beta\mathbf{k}} \hat{\beta}_{\mathbf{k}}^\dagger \hat{\beta}_{\mathbf{k}} \right), \quad (5.1)$$

where 1BZ is the first Brillouin zone. This hamiltonian can be interpreted in the usual way: spin waves are carried by bosonic-type quasiparticles, namely magnons, with two modes α and β . The first mode contributes $\hat{\alpha}_{\mathbf{k}}^\dagger \hat{\alpha}_{\mathbf{k}}$ magnons, each one contributing an energy $\hbar\omega_{\alpha\mathbf{k}}$ to the system. Similarly, the other mode β adds up a gas of $\hat{\beta}_{\mathbf{k}}^\dagger \hat{\beta}_{\mathbf{k}}$ magnons, each one with energy $\hbar\omega_{\beta\mathbf{k}}$.

The dispersion relations $\omega_{\alpha\mathbf{k}}$ and $\omega_{\beta\mathbf{k}}$ in antiferromagnetic systems have typically values of the order of THz in frequency units, and have a minimum value (a gap) even at zero magnetic field. In fact, for small wave vector k , those energies can be reduced to a relativistic form

$$\hbar\omega_{\alpha(\beta)\mathbf{k}} = \sqrt{\hbar^2 c_{\alpha(\beta)}^2 k^2 + m_{\alpha(\beta)}^2 c_{\alpha(\beta)}^4} + U. \quad (5.2)$$

where $c_{\alpha(\beta)}$ is the spin wave phase speed of the α (β) mode (both of the order of km/s), m_α and m_β are the magnon effective masses, which depend on the physical parameters of the sample, and U is an external gap not considered in the mass term. Typically, antiferromagnets have mass terms $m_{\alpha(\beta)}c_{\alpha(\beta)}^2/\hbar$ of the order of THz.

5.2 Easy-axis anisotropy antiferromagnet

One possible case of antiferromagnetic system is when it presents easy-axis anisotropy. Materials like MnF_2 [33], FeF_2 [5] and RbMnF_3 [34] present anisotropy along the z axis (see Figure 5.1), in such a way they present an *easy axis* of magnetization. The hamiltonian that allows us to describe such system is

$$\hat{H}_s = -\gamma \sum_{\mathbf{r}_i} \mathbf{H}_0 \cdot \hat{\mathbf{S}}(\mathbf{r}_i) - \sum_{\mathbf{r}_i} \sum_{\mathbf{r}_j \neq \mathbf{r}_i} \frac{J_{ij}}{\hbar^2} \hat{\mathbf{S}}(\mathbf{r}_i) \cdot \hat{\mathbf{S}}(\mathbf{r}_j) - \frac{D_z}{\hbar^2} \sum_{\mathbf{r}_i} \hat{S}_z(\mathbf{r}_i)^2, \quad (5.3)$$

where $D_z > 0$ is the anisotropy coefficient and J_{ij} is the exchange parameter that couples the dipoles i and j . Typically, in antiferromagnetic systems, the negative interaction ($J_{ij} < 0$, which we'll call J_2) dominates over the positive one $J_1 > 0$. For example, for MnF_2 , the nearest-neighbors coupling constants are $J_1 = 0.028$ meV and $J_2 = -0.152$ meV [35], and the anisotropy parameter is $D_z = 0.019$ meV [33].

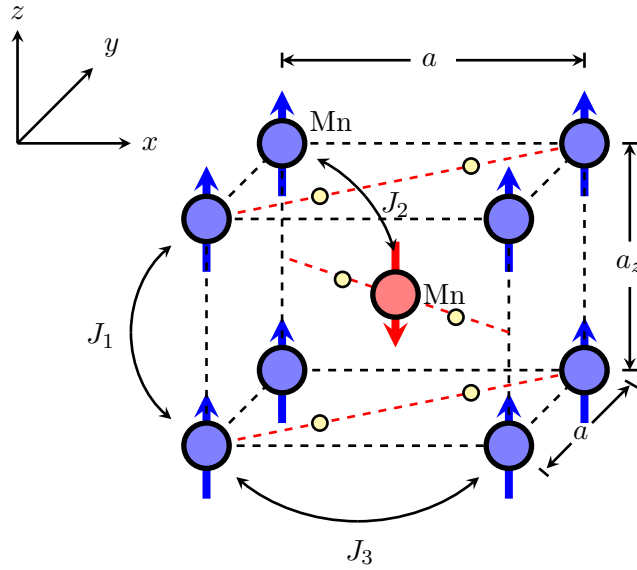


Figure 5.1: Crystal structure of MnF_2 [5]. Big blue circles represent Mn atoms, while small yellow circles represent fluorine. J_1 , J_2 and J_3 are the exchange parameters, with $J_1 > 0$ and $J_2, J_3 < 0$.

When an external magnetic field $\mathbf{H}_0 = H_0 \hat{\mathbf{z}}$ is applied on the sample, it will transport spin waves carried by magnons. Mathematically, this can be determined through the Holstein-Primakoff (HP) transformation

[29] for $\hat{S}^+ = \hat{S}_x + i\hat{S}_y$ and $\hat{S}^- = \hat{S}_x - i\hat{S}_y$:

$$\begin{aligned}\hat{S}_1^- (\mathbf{r}_i) &\approx \hbar\sqrt{2s}\hat{a}_i^\dagger, \\ \hat{S}_1^+ (\mathbf{r}_i) &\approx \hbar\sqrt{2s}\hat{a}_i, \\ \hat{S}_z (\mathbf{r}_i) &= \hbar \left(s - \hat{a}_i^\dagger \hat{a}_i \right)\end{aligned}\tag{5.4}$$

for $\mathbf{r}_i \in L_1$, and

$$\begin{aligned}\hat{S}_2^- (\mathbf{r}_j) &\approx \hbar\sqrt{2s}\hat{b}_j, \\ \hat{S}_2^+ (\mathbf{r}_j) &\approx \hbar\sqrt{2s}\hat{b}_j^\dagger, \\ \hat{S}_z (\mathbf{r}_j) &= \hbar \left(\hat{b}_j^\dagger \hat{b}_j - s \right)\end{aligned}\tag{5.5}$$

if $\mathbf{r}_j \in L_2$. The index i enumerates the magnetic dipole of each sublattice, and here was used to write $\hat{a}_i = \hat{a}(\mathbf{r}_i)$ and $\hat{b}_i = \hat{b}(\mathbf{r}_i)$. In this transformation, s is the spin quantum number of the particles ($s = 5/2$ for MnF_2 [35]). Next, expanding $\hat{a}_i$ and $\hat{b}_i$ in the Fourier space with variable $\mathbf{k}$, and using Bogoliubov transformations

$$\begin{aligned}\hat{a}_{\mathbf{k}} &= u_{\mathbf{k}}\hat{\alpha}_{\mathbf{k}} - v_{\mathbf{k}}\hat{\beta}_{-\mathbf{k}}^\dagger, \\ \hat{b}_{-\mathbf{k}}^\dagger &= -v_{\mathbf{k}}\hat{\alpha}_{\mathbf{k}} + u_{\mathbf{k}}\hat{\beta}_{-\mathbf{k}}^\dagger,\end{aligned}$$

we obtain the diagonal form (5.1) of the hamiltonian. The necessary coefficients $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ for this are

$$u_{\mathbf{k}} = \sqrt{\frac{\tilde{\omega} + \omega_{\mathbf{k}}}{2\omega_{\mathbf{k}}}} \quad v_{\mathbf{k}} = \sqrt{\frac{\tilde{\omega} - \omega_{\mathbf{k}}}{2\omega_{\mathbf{k}}}}\tag{5.6}$$

with $\omega_{\mathbf{k}} = \omega_{\alpha\mathbf{k}}(H_0 = 0) = \omega_{\beta\mathbf{k}}(H_0 = 0)$ and we have defined $\hbar\tilde{\omega} = 2s(Z|J_2| + D_z)$.

In this case, the magnon frequencies are

$$\omega_{\alpha(\beta)\mathbf{k}} = \sqrt{\tilde{\omega}^2 - \tilde{\Omega}_{\mathbf{k}}^2} \pm \gamma H_0.\tag{5.7}$$

where the positive sign is used for the α magnonic mode, while the negative is used for β . Also, γ is the gyromagnetic ratio of the sample, $\hbar\tilde{\Omega}_{\mathbf{k}} = 2sZ|J_2|\gamma_{\mathbf{k}}$ and

$$\gamma_{\mathbf{k}} = \frac{1}{Z} \sum_{\boldsymbol{\delta}} \exp(i\mathbf{k} \cdot \boldsymbol{\delta})\tag{5.8}$$

is a crystal structure factor, whose sum over $\boldsymbol{\delta}$ is effectuated over the Z first neighbors of each point inside the lattice. For tetragonal structures like MnF_2 or FeF_2 [5], one has

$$\gamma_{\mathbf{k}} = \cos\left(\frac{k_x a}{2}\right) \cos\left(\frac{k_y a}{2}\right) \cos\left(\frac{k_z a}{2}\right)\tag{5.9}$$

The dispersion relation for this situation is illustrated in Figure 5.2, where is clearly shown the difference between the two modes.

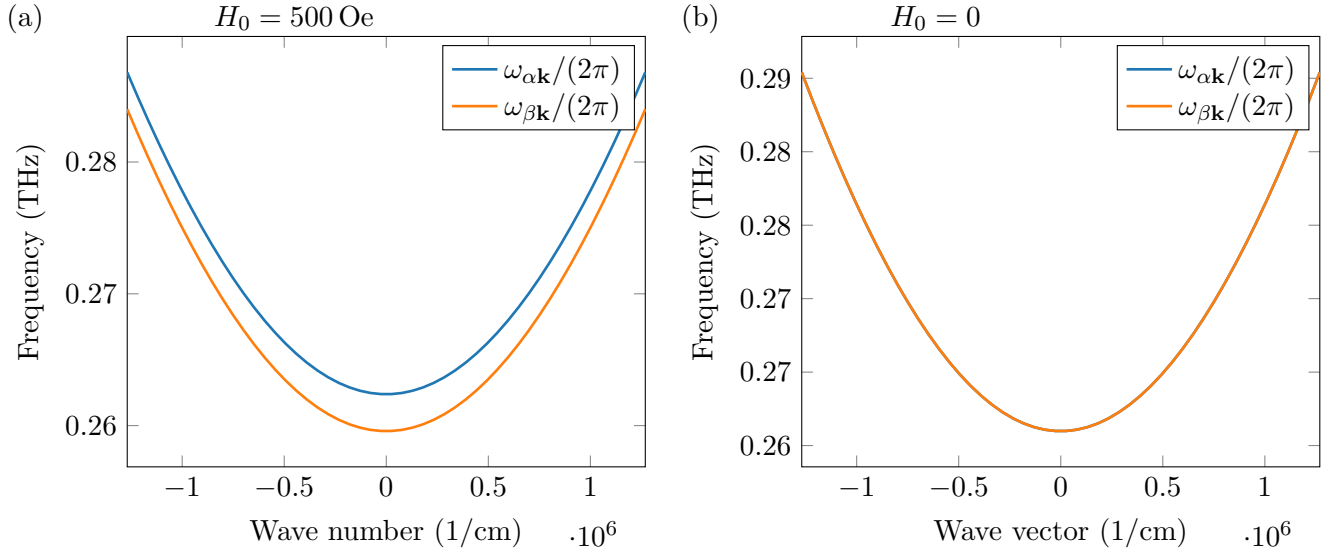


Figure 5.2: Magnon dispersion relation (in linear frequency units) versus wave vector in one dimension ($k = k_x$), for easy-axis antiferromagnet MnF_2 , using equations (5.7). (a) With magnetic field $H_0 = 500$ Oe. (b) When no external magnetic field is applied, both magnon modes have the same energy.

In this case, and for cubic structures in general, this structure factor for small $k \ll \pi/a$ reduces to

$$\gamma_{\mathbf{k}} \approx 1 - \frac{(ka)^2}{8} \quad (5.10)$$

with $k = |\mathbf{k}| = \sqrt{k_x^2 + k_y^2}$. Inserting this into equation (5.7), expanding $\hbar\tilde{\omega} = 2s(Z|J_2| + D_z)$ and $\hbar\tilde{\Omega}_{\mathbf{k}} = 2sZ|J_2|\gamma_{\mathbf{k}}$, we have

$$\begin{aligned} \hbar\omega_{\alpha(\beta)\mathbf{k}} &\approx 2s\sqrt{D_z^2 + 2ZJ_2D_z + Z^2J_2^2a^2k^2/4 \pm \hbar\gamma H_0} \\ \Rightarrow \hbar\omega_{\alpha(\beta)\mathbf{k}} &\approx \sqrt{4s^2D_z^2 + 8s^2Z|J_2|D_z + s^2Z^2J_2^2a^2k^2 \pm \hbar\gamma H_0}. \end{aligned} \quad (5.11)$$

Comparing with the relativistic-like energy (5.2), we read a spin wave speed

$$c = \frac{sZ|J_2|a}{\hbar} \quad (5.12)$$

and a magnon effective mass

$$m = \frac{\hbar^2}{s^2Z^2J_2^2a^2} \sqrt{4s^2D_z^2 + 8s^2Z|J_2|D_z}, \quad (5.13)$$

for both modes α and β . For even smaller k , this takes the form of a non-relativistic particle:

$$\hbar\omega_{\alpha(\beta)\mathbf{k}} \approx \frac{\hbar^2k^2}{2m} + mc^2 \pm \hbar\gamma H_0. \quad (5.14)$$

Note that the magnon frequency gap is mainly given by the mass term $mc^2/\hbar$, which goes to 0 as $D_z \rightarrow 0$. From Figure (5.2b) can be seen that the magnon frequency gap, given by the mass term $mc^2/\hbar$, is of the

order of THz, something pretty common for easy-axis AFI such as MnF_2 and FeF_2 . Few exceptions, like RbMnF_3 ($s = 5/2$ [36]), present a gap of the order of ~ 20 GHz because of to their low anisotropy [34]. Such gap, however, is still being greater than the NV resonance frequencies $\omega_{\pm}$.

5.3 Easy-plane antiferromagnet

Some other materials like NiO [37], CoCl_2 [38] and FeBO_3 [39] are antiferromagnetic systems with a *hard* anisotropy axis besides from the easy-axis mentioned in the previous section. Such configuration is the so-called *easy-plane* anisotropy. The crystal structure of the typical easy-plane antiferromagnet, NiO , is shown in Figure 5.3, where the z axis is, as before, the easy-axis, while the x axis is the hard axis. These anisotropies can be expressed in the hamiltonian as two anisotropy terms with constants $D_x > 0$ and $D_z > 0$, respectively:

$$\hat{H} = -\gamma \sum_{\mathbf{r}_i} \mathbf{H}_0 \cdot \hat{\mathbf{S}}(\mathbf{r}_i) - \sum_{\mathbf{r}_i} \sum_{\mathbf{r}_j \neq \mathbf{r}_i} \frac{J_{ij}}{\hbar^2} \hat{\mathbf{S}}(\mathbf{r}_i) \cdot \hat{\mathbf{S}}(\mathbf{r}_j) + \frac{D_x}{\hbar^2} \sum_{\mathbf{r}_i} \hat{S}_x(\mathbf{r}_i)^2 - \frac{D_z}{\hbar^2} \sum_{\mathbf{r}_i} \hat{S}_z(\mathbf{r}_i)^2. \quad (5.15)$$

There exist samples, like $\alpha\text{-Fe}_2\text{O}_3$, that are easy-plane antiferromagnets, but behave as easy-axis antiferromagnets for temperatures below the so-called *Morin transition* point. For $\alpha\text{-Fe}_2\text{O}_3$, this critical temperature is $T_M = 263$ K [18].

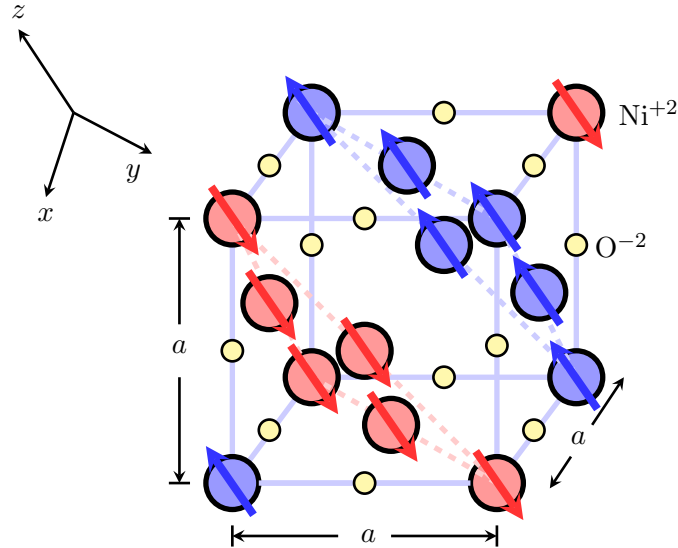


Figure 5.3: Lattice structure of NiO in AF phase. Arrows represent orientations of spins. The small yellow circles represent O^{2-} ions and the large circles are Ni^{2+} ions.

In a similar fashion to MnF_2 , NiO 's hamiltonian can be expressed in the diagonal form (5.1). The procedure is analogous, being the only difference that the Bogoliubov transformations $\hat{a}_{\mathbf{k}} = u_{\alpha\mathbf{k}}\hat{\alpha}_{\mathbf{k}} -$

$v_{\beta\mathbf{k}}\hat{\beta}_{-\mathbf{k}}^\dagger$ and $\hat{b}_{-\mathbf{k}}^\dagger = -v_{\alpha\mathbf{k}}\hat{\alpha}_{\mathbf{k}} + u_{\beta\mathbf{k}}\hat{\beta}_{-\mathbf{k}}^\dagger$ need now four coefficients [40]:

$$\begin{aligned} u_{\alpha(\beta),\mathbf{k}} &= \sqrt{\frac{\tilde{\omega} + \omega_{\alpha(\beta),\mathbf{k}}(0)}{2\omega_{\alpha(\beta),\mathbf{k}}(0)}}, \\ v_{\alpha(\beta),\mathbf{k}} &= \sqrt{\frac{\tilde{\omega} - \omega_{\alpha(\beta),\mathbf{k}}(0)}{2\omega_{\alpha(\beta),\mathbf{k}}(0)}}, \end{aligned} \quad (5.16)$$

with $\hbar\tilde{\omega} = 2s(ZJ_2 + D_z + D_x/2)$ in this case.

An interesting result of this procedure is that the dispersion relations for magnonic modes α and β are different even with no external magnetic field (see Figure 5.4). These are given by

$$\omega_{\alpha(\beta)\mathbf{k}}^2 = \tilde{\omega}^2 + \gamma^2 H_0^2 - \tilde{\omega}_x^2 - \tilde{\Omega}_{\mathbf{k}}^2 \pm 2\sqrt{\gamma^2 H_0^2 (\tilde{\omega}^2 - \tilde{\Omega}_{\mathbf{k}}^2) + \tilde{\Omega}_{\mathbf{k}}^2 \tilde{\omega}_x^2}, \quad (5.17)$$

with $\hbar\tilde{\Omega}_{\mathbf{k}} = 2sZ|J_2|\gamma_{\mathbf{k}}$ and $\hbar\tilde{\omega}_x = sD_x$. As in equation (5.7), the α mode uses the positive sign, while the β takes the negative one. It's a common practice to take a spherical first Brillouin zone, so that the structure factor $\gamma_{\mathbf{k}}$ is

$$\gamma_{\mathbf{k}} = \cos\left(\frac{k a}{2}\right), \quad (5.18)$$

and for small k , it reduces to the same form as equation (5.10).

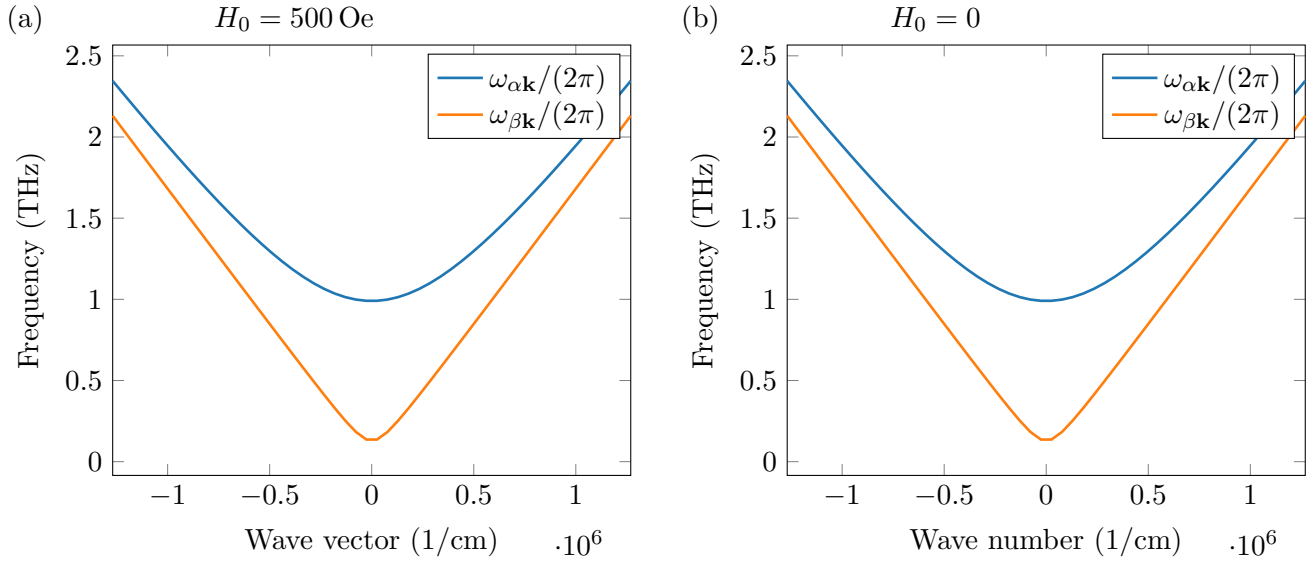


Figure 5.4: Magnon dispersion relation (in linear frequency units) versus wave vector in one dimension ($k = k_x$), using equations (5.17), for easy-plane antiferromagnet NiO. (a) With magnetic field $H_0 = 500$ Oe. (b) With $H_0 = 0$.

Dispersion relations for easy-plane anisotropic systems can be approximated in the same relativistic-way form of equation (5.2). However, unlike the easy-axis case, the magnetic term $\hbar\gamma H_0$ is not out of the

square root, but it's inside of it. This means the effective masses $m_{\alpha(\beta)}$ and the spin wave phase speeds $c_{\alpha(\beta)}$ will depend on the magnetic field H_0 , and therefore, they'll be different according to which mode α or β we are evaluating. After some approximations in (5.17), we find

$$c_{\alpha(\beta)} = \frac{sZ|J_2|a}{\hbar} \sqrt{1 \pm (H_0^2 - \frac{\hbar^2 \gamma^2}{4s^2} D_x^2/4) \frac{\hbar^2 \gamma^2}{4s^2 E_{\alpha(\beta)}^2}}$$

$$m_{\alpha(\beta)} c_{\alpha(\beta)}^2 = 2s \sqrt{D_z^2 + ZD_x|J_2| + 2Z|J_2|D_z + D_x D_z \pm 2E_{\alpha(\beta)}^2}$$

with $E_{\alpha(\beta)}^2 = \sqrt{\frac{\hbar^2 \gamma^2 H_0^2}{4s^2} (D_x^2/4 + D_z^2 + Z|J_2|D_x + 2Z|J_2|D_z + D_x D_z) + Z^2 J_2^2 D_x^2/4}$. This reproduces the relativistic form

$$\hbar \omega_{\alpha(\beta)\mathbf{k}} = \sqrt{\hbar^2 c_{\alpha(\beta)}^2 k^2 + m_{\alpha(\beta)}^2 c_{\alpha(\beta)}^4}, \quad (5.19)$$

with $U_{\alpha(\beta)} = 0$ in equation (5.2). For even smaller k , we obtain

$$\hbar \omega_{\alpha(\beta)\mathbf{k}} \approx \frac{\hbar^2 k^2}{2m_{\alpha(\beta)}} + m_{\alpha(\beta)} c_{\alpha(\beta)}^2. \quad (5.20)$$

5.4 Phase transitions in antiferromagnets

In general, antiferromagnetic materials may lose their *antiferromagnetic behavior* passing through some temperatures or critical points. A well-known situation is the Neel temperature T_N , above which the antiferromagnetic system behaves as paramagnetic (similar to the Curie temperature in ferromagnetic materials). In this work, we only consider materials at lower temperatures than their respective Neel temperatures.

Other common phase transition is spin-flop. One can find the points where this transition occurs, by considering the energy per unit volume, based on the hamiltonian (5.3) in the macrospin approximation

$$u(\mathbf{M}_1, \mathbf{M}_2) = -H_0(M_{1z} + M_{2z}) + \frac{H_E}{M} \mathbf{M}_1 \cdot \mathbf{M}_2 + \frac{H_{Ax}}{2M} (M_{1x}^2 + M_{2x}^2) - \frac{H_{Az}}{2M} (M_{1z}^2 + M_{2z}^2) \quad (5.21)$$

where

$$H_E = \frac{2sZ|J_2|}{\hbar\gamma}, \quad H_{Ax} = \frac{2sD_x}{\hbar\gamma}, \quad H_{Az} = \frac{2sD_z}{\hbar\gamma} \quad (5.22)$$

are convenient parameters (effective fields) that are commonly defined in this context [5], while $\mathbf{M}_1 = \hbar\gamma n \langle \mathbf{S}(\mathbf{r}_i \in L_1) \rangle$, $\mathbf{M}_2 = \hbar\gamma n \langle \mathbf{S}(\mathbf{r}_j \in L_2) \rangle$, n is the volume dipole density and $M = M_1(T=0) = M_2(T=0)$. Note that, if the external magnetic field is applied along the easy-axis of magnetization ($\mathbf{H}_0 = H_0 \hat{\mathbf{z}}$), we have $M_{1x} = M_{2x} = 0$ and the easy-plane case reduces to easy-axis. Therefore, it is enough to analyze only the easy-axis system energy. In terms of the angles θ_1 and θ_2 that vectors $\mathbf{M}_1$ and $\mathbf{M}_2$ form with the $\hat{\mathbf{z}}$ axis, we have

$$\frac{u(\theta_1, \theta_2)}{M} = -H_0(\cos \theta_1 + \cos \theta_2) + H_E \cos(\theta_1 - \theta_2) - \frac{H_{Az}}{2} (\cos^2 \theta_1 + \cos^2 \theta_2). \quad (5.23)$$

One can find the equilibrium condition by minimizing this volumetric energy $u(\theta_1, \theta_2)$, which results in the equations $\partial u / \partial \theta_1 = \partial u / \partial \theta_2 = 0$ with two possible solutions, depending on the value of the field H_0 :

$$\begin{cases} \theta_1 = 0, \theta_2 = \pi & \text{if } H_0 < H_{SF}, \\ \theta_1 = \theta_2 = \arccos[H_0 / (2H_E - H_{Az})] & \text{if } H_0 \geq H_{SF}, \end{cases} \quad (5.24)$$

where $H_{SF} := \sqrt{2H_E H_{Az} - H_{Az}^2} \approx \sqrt{2H_E H_{Az}}$. The first case ($\theta_1 = 0, \theta_2 = \pi$) is the typical antiferromagnetic (AF) phase we have been discussing so far. The other situation with $\theta_1 = \theta_2$ is known as *spin-flop* (SF), and this configuration defines a phase for antiferromagnets that can only be reached when H_0 exceeds the critical magnetic field $H_{SF} \propto \sqrt{|J_2|D_z}$. This critical field can be extremely high. For instance, considering MnF_2 , we have $H_E = 526 \text{ kOe}$ and $H_{Az} = 8.2 \text{ kOe}$, so $H_{SF} \approx (2H_E H_A)^{1/2} = 93 \text{ kOe}$ [5].

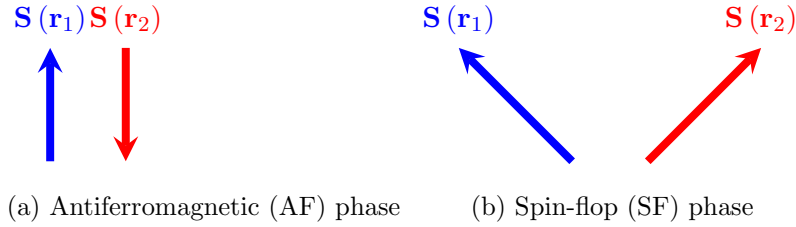


Figure 5.5: Spin orientations in different phases.

Other interesting transition, already mentioned before for $\alpha\text{-Fe}_2\text{O}_3$, is the Morin transition, which consists of a sudden rotation of the easy-axis $\hat{\mathbf{z}}$, creating an easy-plane anisotropy. For example, in $\alpha\text{-Fe}_2\text{O}_3$, for a temperature T less than $T_M = 263 \text{ K}$, the system has only uniaxial anisotropy, with easy-axis $\hat{\mathbf{z}}$. But for $T > T_M$, this easy axis turns out to be parallel to the xy plane, which is to say, the basis of directions for representing spins operators transforms as $\hat{\mathbf{x}}_1 \rightarrow \hat{\mathbf{z}}_2$ and $\hat{\mathbf{z}}_1 \rightarrow \hat{\mathbf{x}}_2$ (see Figure 5.6). Note that this coordinate transformation means a change in the way we express the magnetic field:

$$\mathbf{H}_0 = \begin{cases} H_0 \hat{\mathbf{z}}_1 & \text{if } T \leq T_M, \\ H_0 \hat{\mathbf{x}}_2 & \text{if } T > T_M, \end{cases}$$

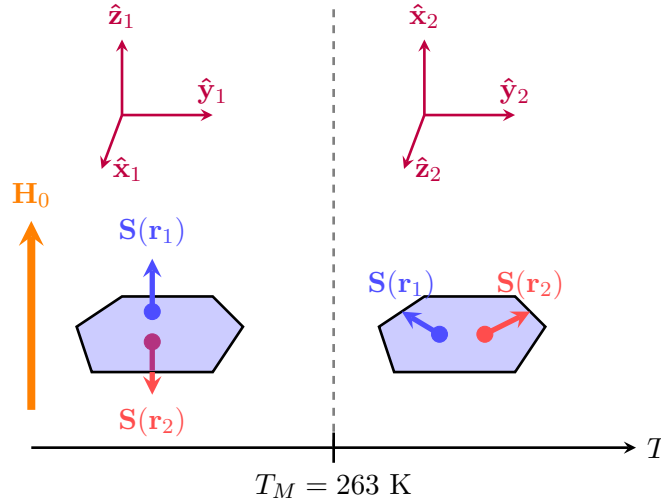


Figure 5.6: Morin transition in hematite ($\alpha\text{-Fe}_2\text{O}_3$), indicating the spin orientations relative to the external magnetic field $\mathbf{H}_0$.

Here, $\hat{\mathbf{z}}_1$ and $\hat{\mathbf{x}}_2$ are essentially the same vector (as the magnetic field direction remains constant), but they differ in which will be selected as element of the basis. With this in mind, the new anisotropy direction ($\hat{\mathbf{z}}_2$) and the magnetic field are now perpendicular. In this *canted phase*, the Zeeman effect term in the hamiltonian (5.3) must be expanded as $\gamma \mathbf{H}_0 \cdot \hat{\mathbf{S}}(\mathbf{r}) = \gamma H_0 \hat{S}_x(\mathbf{r})$, where x here is our *old* anisotropy direction. Doing the same procedure as the previous sections to diagonalize the hamiltonian $\hat{H}_s$, we find the dispersion relations in the canted phase:

$$\begin{aligned} \omega_{\alpha(\beta)\mathbf{k}}^2 = & \gamma (H_0 \cos \theta_c - H_E \cos(2\theta_c) + H_A (2 \sin^2 \theta_c - \cos^2 \theta_c) / 2 \mp \gamma_{\mathbf{k}} H_E (1 + \cos 2\theta_c) / 2)^2 \\ & - \gamma (\gamma_{\mathbf{k}} H_E \sin^2 \theta_c \mp H_A \cos^2 \theta_c / 2)^2, \end{aligned} \quad (5.25)$$

where H_E and H_{Az} are defined as before in (5.22), and $\theta_c = \arccos[H_0 / (2H_E + H_{Az})]$ (do not confuse with the spin-flop angle introduced in (5.24), which has a minus sign).

To sum up, antiferromagnetic insulators (AFI) materials are also interesting candidates to represent the dipole system model introduced in Chapter 3. As for the ferromagnetic case, the goal here was to indicate how spins interact between, and we did for both easy-axis and easy-plane anisotropy cases. Both situations involucrate Heisenberg's spin-spin interaction, Zeeman effect, and anisotropy terms. Antiferromagnetic systems present spin waves and magnons, too, but they have some practical differences compared to ferromagnetic materials. To begin, their frequency gap is of the order of THz, while the ferromagnetic gap is just of a few GHz. This is also greater than the NV center resonance frequencies $\omega_{\pm} \sim \text{GHz}$. Second, magnons have two modes of energy (α and β), which will eventually result in a more complex

expansion of the spin density operators $\hat{\mathbf{S}}(\mathbf{r}, t)$ in terms of the magnon creation-annihilation operators $\hat{\alpha}_{\mathbf{k}}^\dagger$ ($\hat{\alpha}_{\mathbf{k}}$) and $\hat{\beta}_{\mathbf{k}}^\dagger$ ($\hat{\beta}_{\mathbf{k}}$). Anyway, bosonic commutation relations $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$ and $[\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$ are still valid, and can be used to calculate the spin-spin correlation functions using the fluctuation-dissipation theorem mentioned in Chapter 3, and therefore, the desired relaxation rates $\Gamma(\omega_\pm)$. Experimentally, one can measure these rates versus the external magnetic field H_0 , and by fitting the theoretical expression for $\Gamma_\pm(\mathbf{H}_0)$ it is possible to extract the intersublattice spin-spin coupling constant $J_2 < 0$ and the anisotropy parameters D_z and D_x . We also discussed phase transitions that are typically present in AFI, like spin-flop transition and the Morin transition of $\alpha\text{-Fe}_2\text{O}_3$. We expect the NV center to detect these transitions, reflected in the relaxation rates.

Chapter 6

Relaxometry simulations

In order to calculate the NV center relaxation rates $\Gamma(\omega_{\pm})$, we utilize equation (3.12). For each material, it was necessary to calculate the spin-spin correlators C_{xx} and C_{zz} . Thanks to the fluctuation-dissipation theorem (3.13), the problem of calculating the correlation functions is reduced to find the commutators shown in equation (3.14), in this case,

$$\chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \frac{i}{\hbar} \theta(t) \langle [\hat{s}_{\alpha}(\mathbf{r}, t), \hat{s}_{\beta}(\mathbf{r}', 0)] \rangle_{eq}.$$

In this chapter, we calculate the spin-spin correlation functions, for each type of ferromagnetic and anti-ferromagnetic (AF) samples described in the last two chapters, using the fluctuation-dissipation theorem. With these correlators $C_{\alpha\beta}(\mathbf{k}, \omega)$, we present simulations for the desired relaxation rates $\Gamma(\omega_{\pm})$ as functions of the external magnetic field $\mathbf{H}_0$. In general, the field dependence of $\Gamma(\omega_{\pm})$ is contained in the NV resonance frequencies $\omega_{\pm} = \omega_{\pm}(\mathbf{B}_{\text{NV}})$, as explained in Chapter 2, being $\mathbf{B}_{\text{NV}}$ the magnetic field detected by the NV center. If the sample produces a low magnetic field compared to $\mathbf{H}_0$, we may simply take $\mathbf{B}_{\text{NV}} \approx \mathbf{H}_0$.

6.1 Spin-spin correlations in ferromagnetic systems

6.1.1 Transverse spin-spin correlation for ferromagnetic systems

First, we calculate the transverse spin-spin correlation functions $C_{xx} = C_{yy}$, which by fluctuation-dissipation theorem (3.13) is given by

$$C_{xx}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi''_{xx}(\mathbf{k}, \omega), \quad (6.1)$$

where the linear response function in real space is

$$\begin{aligned}\chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_x(\mathbf{r}, t), \hat{s}_x(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_x(\mathbf{r}, t), \hat{S}_x(\mathbf{r}', 0)] \rangle_{eq}.\end{aligned}$$

Here, σ is the dipole surface density $\hat{S}_x = (\hat{S}^+ + \hat{S}^-)/2$ and $\hat{S}_y = (\hat{S}^+ - \hat{S}^-)/(2i)$, so we may write the linear response function as

$$\begin{aligned}\chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{4\hbar} \theta(t) \langle [\hat{S}^+(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \\ &\quad + [\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^-(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \rangle_{eq}\end{aligned}$$

As these commutators are evaluated at different instants, it is not trivial to get rid off them. The strategy here is to use the simple time evolution that obey annihilation operators $\hat{a}_{\mathbf{k}}(t) = \hat{a}_{\mathbf{k}}(0) \exp(-i\omega_{\mathbf{k}}t - \eta t)$, with $\eta > 0$ a damping parameter. Therefore, it is convenient to use the Holstein-Primakoff transformations defined in equation (4.4), obtaining

$$\begin{aligned}\chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\hbar\sigma^2}{2} \theta(t) \langle [\hat{a}(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \\ &\quad + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \rangle_{eq}\end{aligned}$$

Now, expanding every raising and lowering operator in the Fourier space, we have

$$\begin{aligned}\chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\hbar\sigma^2}{2N} \theta(t) \sum_{\mathbf{k}' \in 1\text{BZ}} \sum_{\mathbf{k}'' \in 1\text{BZ}} \langle [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ &\quad + [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \\ &\quad + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ &\quad + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \rangle_{eq}\end{aligned}$$

and taking the Fourier transform for the relative position vector $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ (or equivalently¹, for $\mathbf{r}$ evaluating $\mathbf{r}' = \mathbf{0}$), in the convenient limit $\int d^2\mathbf{R} \rightarrow \frac{1}{\sigma} \sum_{\mathbf{R}}$, we obtain

$$\chi_{xx}(\mathbf{k}, t) = \frac{i\hbar\sigma}{2} \theta(t) \sum_{\mathbf{k}' \in 1\text{BZ}} \left\langle [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] \right\rangle_{eq} \quad (6.2)$$

Now it is possible to use the time evolution of the operators $\hat{a}_{\mathbf{k}}$ and $\hat{a}_{\mathbf{k}}^\dagger$ in Heisenberg picture:

$$\begin{aligned}\frac{d\hat{a}_{\mathbf{k}}}{dt} &= \frac{i}{\hbar} [\hat{H}_s, \hat{a}_{\mathbf{k}}] - \eta \hat{a}_{\mathbf{k}} = \frac{i}{\hbar} \left[\sum_{\mathbf{k}' \in 1\text{BZ}} \hat{a}_{\mathbf{k}'}^\dagger \hat{a}_{\mathbf{k}'} \hbar\omega_{\mathbf{k}'}, \hat{a}_{\mathbf{k}} \right] - \eta \hat{a}_{\mathbf{k}} \\ \Rightarrow \hat{a}_{\mathbf{k}}(t) &= \hat{a}_{\mathbf{k}}(0) \exp(-i\omega_{\mathbf{k}}t - \eta t).\end{aligned}$$

¹In the long run, the result will be the same as using $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, since the system has translational symmetry.

Using this result into equation (6.2), one clearly observes that the first and the last commutators vanish, while the rest reduce to the bosonic relation $[\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$, so

$$\Rightarrow \mathcal{F}\{\chi_{yy}\}(\mathbf{k}, t) = \frac{i\hbar\sigma}{2}\theta(t) [\exp(-i\omega_{\mathbf{k}}t) - \exp(i\omega_{\mathbf{k}}t)] \exp(-\eta t).$$

Next, taking temporal Fourier transform,

$$\begin{aligned} \chi'_{xx}(\mathbf{k}, \omega) + i\chi''_{xx}(\mathbf{k}, \omega) &= \frac{i\hbar\sigma}{2} \int_0^\infty \exp(i\omega t) [\exp(-i\omega_{\mathbf{k}}t) - \exp(i\omega_{\mathbf{k}}t)] \exp(-\eta t) dt \\ \Rightarrow \chi'_{xx}(\mathbf{k}, \omega) + i\chi''_{xx}(\mathbf{k}, \omega) &= -\frac{\hbar\sigma}{2} \frac{1}{(\omega - \omega_{\mathbf{k}}) + i\eta} + \frac{\hbar\sigma}{2} \frac{1}{(\omega + \omega_{\mathbf{k}}) + i\eta} \end{aligned}$$

Therefore, the imaginary part of the linear response function is

$$\chi''_{xx}(\mathbf{k}, \omega) = \frac{\hbar\sigma}{2} \frac{\eta}{(\omega_{\mathbf{k}} - \omega)^2 + \eta^2} - \frac{\hbar\sigma}{2} \frac{\eta}{(\omega_{\mathbf{k}} + \omega)^2 + \eta^2}.$$

which corresponds to a sum of lorentzians; one of them peaks at $\omega = \omega_{\mathbf{k}}$ (see Figure 6.1), while the other does at $\omega = -\omega_{\mathbf{k}}$. However, as both the dispersion relation $\omega_{\mathbf{k}}$ and the frequency ω are positive, the last lorentzian is negligible. With this result, it is possible to use the fluctuation-dissipation theorem (3.13) and obtain the transverse spin-spin correlation function

$$C_{xx}(\mathbf{k}, \omega) = \frac{\hbar\sigma k_B T}{\omega} \frac{\eta}{(\omega_{\mathbf{k}} - \omega)^2 + \eta^2} \quad (6.3)$$

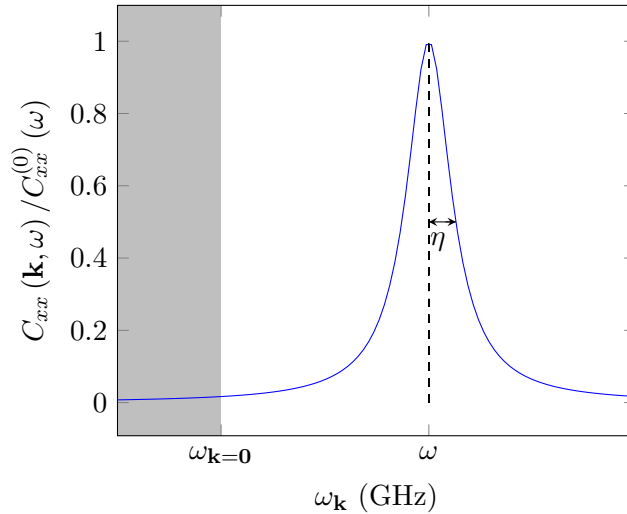


Figure 6.1: Normalized transverse spin-spin correlation function $C_{xx}(\mathbf{k}, \omega) / C_{xx}^{(0)}(\omega)$ versus frequency $\omega_{\mathbf{k}}$, being $C_{xx}^{(0)}(\omega) = \hbar\sigma k_B T / (\omega\eta)$. Here, $\eta \sim 2\pi \times 0.5$ GHz and $\omega \sim D_{\text{NV}}$. The shaded region corresponds to the forbidden values for $\omega_{\mathbf{k}}$ due to the gap in the dispersion relation.

This correlation has the following interpretation: normal modes of spin wave whose frequency $\omega_{\mathbf{k}}$ coincides with the NV center resonance ω are those who contribute the most to the spin-spin correlation.

In second quantization terms, as magnons are the quanta for the interaction between spins, creation-annihilation processes with energy $\hbar\omega$ are the most relevant for the relaxation rate. In this way, the NV center triggers the magnon creation-annihilation with energy $\hbar\omega$, and this is a single-magnon process.

However, if $\hbar\omega < \hbar\omega_{\mathbf{k}=0}$, this single-magnon process cannot occur, and in this case the transverse correlator decays to zero. This can be understood from Figure 6.1: if the peak at ω lies within the shaded region (which is to say, $\omega < \omega_{\mathbf{k}=0}$), then $\omega_{\mathbf{k}}$ will never coincide with ω , and the one-magnon processes are suppressed. In that case, two-magnon processes, contained in the longitudinal correlation C_{zz} , become relevant. This also can be explained graphically using the band diagram shown in Figure 6.2.

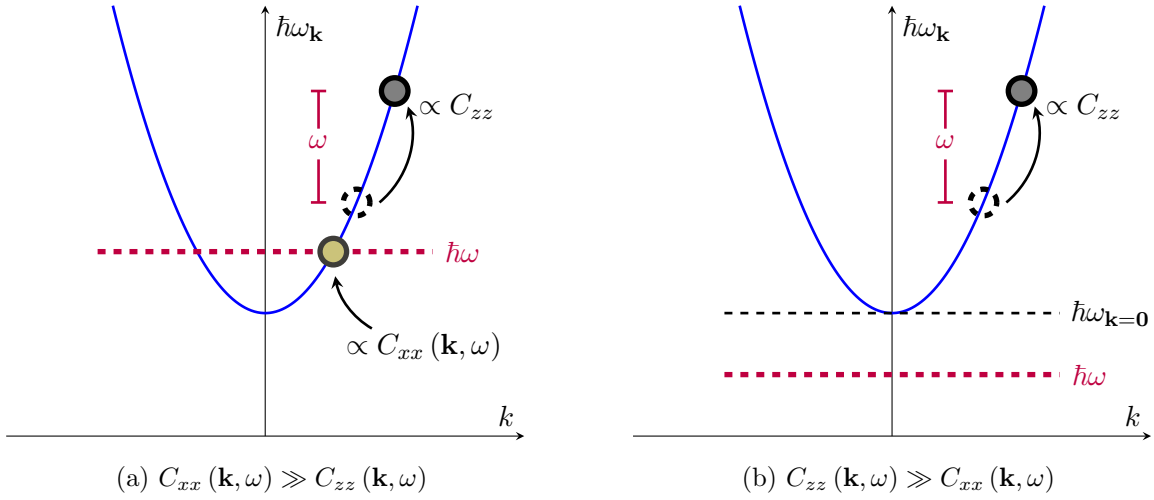


Figure 6.2: Magnon processes, depending on the values of ω . (a) If $\omega > \omega_{\mathbf{k}}$, then one-magnon events (creation and annihilation of a magnon with energy $\hbar\omega$) dominates, which mathematically means $C_{xx} \gg C_{zz}$. (b) If $\omega < \omega_{\mathbf{k}}$, two-magnon scattering dominates (magnons gaining or losing energy $\hbar\omega$), which implies that $C_{zz} \gg C_{xx}$.

6.1.2 Longitudinal spin-spin correlation for ferromagnetic systems

Finding the longitudinal spin-spin correlation function C_{zz} is far more difficult, because its associated linear response function

$$\begin{aligned}\chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_z(\mathbf{r}, t), \hat{s}_z(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_z(\mathbf{r}, t), \hat{S}_z(\mathbf{r}', 0)] \rangle_{eq}.\end{aligned}$$

is a two-magnon correlator, as one can check by using the HP transformations (4.4), in this case, $\hat{S}_z(\mathbf{r}, t) = \hbar (s - \hat{a}^\dagger(t) \hat{a}(t))$:

$$\chi_{zz}(\mathbf{r}, \mathbf{r}'; t) = i\hbar\sigma^2\theta(t) \left\langle \left[\hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}(\mathbf{r}', 0) \right] \right\rangle_{eq}.$$

It is possible to calculate this commutator by splitting it up into four terms using the product rule:

$$\begin{aligned} \chi_{zz}(\mathbf{r}, \mathbf{r}'; t) = i\hbar\sigma^2\theta(t) & \left\langle \left[\hat{a}^\dagger(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \right] \hat{a}(\mathbf{r}, t) \hat{a}(\mathbf{r}', 0) \right. \\ & + \hat{a}^\dagger(\mathbf{r}, t) \left[\hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \right] \hat{a}(\mathbf{r}', 0) \\ & + \hat{a}^\dagger(\mathbf{r}', 0) \left[\hat{a}^\dagger(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0) \right] \hat{a}(\mathbf{r}, t) \\ & \left. + \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}^\dagger(\mathbf{r}, t) \left[\hat{a}(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0) \right] \right\rangle_{eq}. \end{aligned}$$

As before, it is convenient to expand the raising and lowering operators in the Fourier space. It is clear that the first and the last term vanish, while

$$\begin{aligned} \left[\hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \right] &= \frac{1}{N} \sum_{\mathbf{k}' \in \text{1BZ}} \exp(-i\omega_{\mathbf{k}'}t - \eta t) \exp i\mathbf{k}' \cdot (\mathbf{r} - \mathbf{r}') \\ \left[\hat{a}^\dagger(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0) \right] &= -\frac{1}{N} \sum_{\mathbf{k}' \in \text{1BZ}} \exp(i\omega_{\mathbf{k}'}t - \eta t) \exp i\mathbf{k}' \cdot (\mathbf{r}' - \mathbf{r}) \end{aligned}$$

Also, we have to expand the canonical averages

$$\begin{aligned} \left\langle \hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}', 0) \right\rangle_{eq} &= \frac{1}{N} \sum_{\mathbf{k}'' \in \text{1BZ}} n_B(\hbar\omega_{\mathbf{k}''} - \mu) \exp(i\omega_{\mathbf{k}''}t - \eta t) \exp i\mathbf{k}'' \cdot (\mathbf{r}' - \mathbf{r}) \\ \left\langle \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}(\mathbf{r}, t) \right\rangle_{eq} &= \frac{1}{N} \sum_{\mathbf{k}'' \in \text{1BZ}} n_B(\hbar\omega_{\mathbf{k}''} - \mu) \exp(-i\omega_{\mathbf{k}''}t - \eta t) \exp i\mathbf{k}'' \cdot (\mathbf{r} - \mathbf{r}') \end{aligned}$$

where we used $\left\langle \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}''} \right\rangle_{eq} = n_B(\hbar\omega_{\mathbf{k}''} - \mu) \delta_{\mathbf{k}'\mathbf{k}''}$, being $n_B(\varepsilon) = \left[\exp\left(\frac{\varepsilon}{k_B T}\right) - 1 \right]^{-1}$ the Bose-Einstein distribution and μ the chemical potential.

Therefore, taking Fourier transform with respect to $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ and with respect to time t ,

$$\chi'_{zz}(\mathbf{k}, \omega) + i\chi''_{zz}(\mathbf{k}, \omega) = \frac{i\hbar\sigma}{N} \sum_{\mathbf{k}' \in \text{1BZ}} \left\{ \frac{n_B(\hbar\omega_{\mathbf{k}'-\mathbf{k}} - \mu)}{\tilde{\eta} - i(\omega - \omega_{\mathbf{k}'} + \omega_{\mathbf{k}'-\mathbf{k}})} - \frac{n_B(\hbar\omega_{\mathbf{k}'+\mathbf{k}} - \mu)}{\tilde{\eta} - i(\omega + \omega_{\mathbf{k}'} - \omega_{\mathbf{k}'+\mathbf{k}})} \right\}$$

with $\tilde{\eta} = 2\eta$. In the summation associated to the first term, it is convenient to replace $\mathbf{k}' - \mathbf{k} \rightarrow \mathbf{k}'$, which can be thanks to the periodicity of the reciprocal lattice. Finally, taking the continuous limit $\frac{1}{N} \sum_{\mathbf{k} \in \text{1BZ}} \rightarrow \frac{a^2}{(2\pi)^2} \int_{\text{1BZ}} d^2\mathbf{k}'$, and estimating $a^2\sigma \rightarrow 1$, we obtain the imaginary part of the linear response function,

$$\chi''_{zz}(\mathbf{k}, \omega) = \frac{\hbar}{(2\pi)^2} \int_{\text{1BZ}} d^2\mathbf{k}' \left\{ \frac{n_B(\hbar\omega_{\mathbf{k}'} - \mu) \tilde{\eta}}{\tilde{\eta}^2 + (\omega - \omega_{\mathbf{k}'+\mathbf{k}} + \omega_{\mathbf{k}'})^2} - \frac{n_B(\hbar\omega_{\mathbf{k}'+\mathbf{k}} - \mu) \tilde{\eta}}{\tilde{\eta}^2 + (\omega + \omega_{\mathbf{k}'} - \omega_{\mathbf{k}'+\mathbf{k}})^2} \right\}$$

which can be used in fluctuation-dissipation theorem (3.13) for obtaining the spin-spin correlation function C_{zz} . In the limit $\tilde{\eta} \rightarrow 0$, these lorentzians transform to Dirac deltas, thanks to the identity

$$\lim_{\tilde{\eta} \rightarrow 0^+} \frac{1}{\pi} \frac{\tilde{\eta}}{\tilde{\eta}^2 + (\omega - \omega_{\mathbf{k}+\mathbf{k}'} + \omega_{\mathbf{k}'})^2} = \delta(\omega - \omega_{\mathbf{k}+\mathbf{k}'} + \omega_{\mathbf{k}'}).$$

So,

$$C_{zz}(\mathbf{k}, \omega) = \frac{\hbar k_B T}{2\pi\omega} \int_{\text{1BZ}} d^2\mathbf{k}' \delta(\omega - \omega_{\mathbf{k}+\mathbf{k}'} + \omega_{\mathbf{k}'}) [n_B(\hbar\omega_{\mathbf{k}'} - \mu) - n_B(\hbar\omega_{\mathbf{k}'+\mathbf{k}} - \mu)] \quad (6.4)$$

It is possible to calculate this integral in an exact way using the non-relativistic approximation (4.8) for the dispersion relation $\omega_{\mathbf{k}}$. As this approximation is in the same order of magnitude as the original frequency (as illustrated in Figure 4.1), this estimation will be reliable enough. Writing $\mathbf{k} = k\hat{\mathbf{y}}''$ and $\mathbf{k} + \mathbf{k}' = k'_x\hat{\mathbf{x}}'' + (k'_y + k)\hat{\mathbf{y}}''$, for $\mu = 0$, we obtain

$$C_{zz}(\mathbf{k}, \omega) \approx \frac{m(k_B T)^2}{2\omega} \left\{ \frac{1}{\sqrt{(\hbar\omega - \hbar^2 k^2/(2m))^2 + 2U\hbar^2 k^2/m}} - \frac{1}{\sqrt{(\hbar\omega - \hbar^2 k^2/(2m))^2 + 2(U + \hbar\omega)\hbar^2 k^2/m}} \right\}, \quad (6.5)$$

where $U = \hbar\gamma H_0$ and m is the magnon effective mass. Plots of this correlation function, for $\omega = \omega_+$ and $\omega = \omega_-$, are shown in Figure 6.3, where they also are compared with the transverse correlation C_{xx} according to equation (6.3). We observe that, as expected, $C_{xx} \gg C_{zz}$, indicating the dominance of the one-magnon processes due to the fact that the gap U is of the order of GHz, comparable with ω .

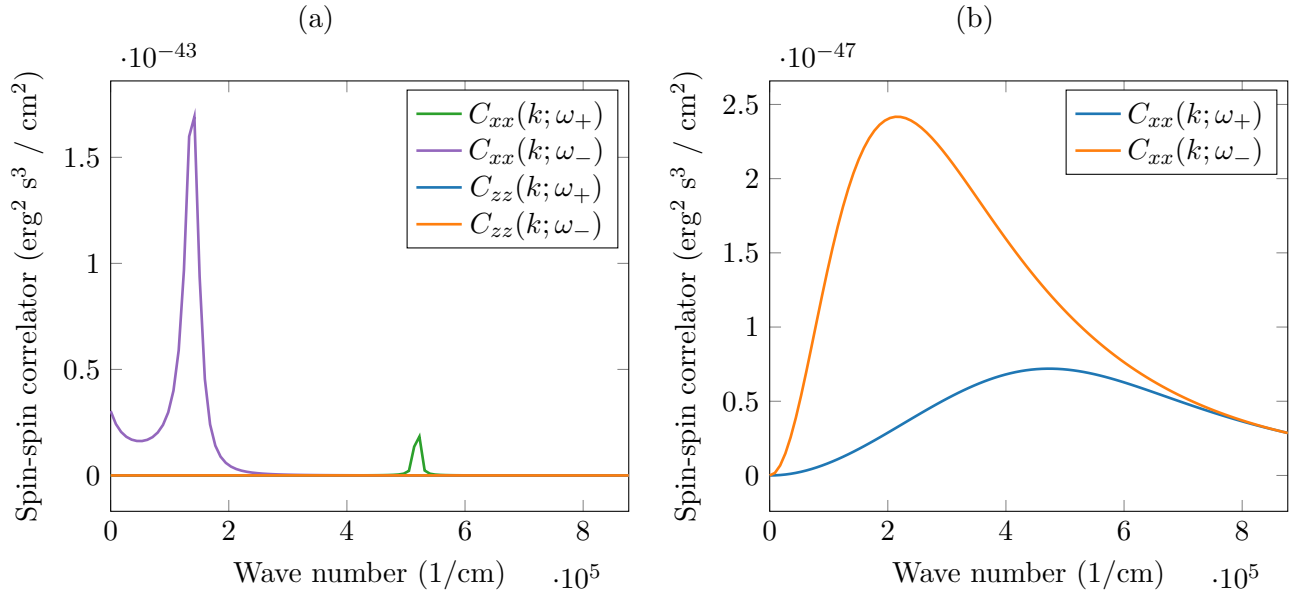


Figure 6.3: Transverse and longitudinal spin-spin correlation functions for a ferromagnetic sample, using equations (6.3) and (6.5). (a) Considering both transverse and longitudinal correlations. (b) Considering only longitudinal correlation.

6.2 Relaxation rates using ferromagnetic samples

In this section, we test the correlation functions found in last sections to compute the relaxation rates $\Gamma(\omega_{\pm})$. Here, we take as sample $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (Yttrium-Iron-Garnet, or YIG for short) thin films, which is a material commonly treated as a ferromagnet and stands out for its low dissipation of spin waves (meaning a low η in equation (6.3)). For our simulations, we consider as reference the experimental results obtained by Lee-Wong and co-authors [17] for this anisotropic material.

As the dispersion relation (4.15) depends on the angle ϕ_k , it is necessary to use the complete form (3.11) for $\Gamma(\omega)$, but neglecting the terms with C_{zz} for reasons already discussed in the previous sections. So, writing the dipole density as

$$\sigma = \frac{M_S t_{\text{film}}}{g\mu_B s} = \frac{M_S t_{\text{film}}}{\hbar\gamma s},$$

being t_{film} the film thickness, one obtains the relaxation rates for this situation:

$$\Gamma_{\pm} = \int_0^{\infty} dk \int_0^{2\pi} d\phi_k f(\phi_k, \theta) k^3 \exp(-2kz) \frac{M_S t_{\text{film}}}{\gamma} \frac{k_B T}{\omega_{\pm}} \frac{\eta}{\eta^2 + (\omega_{\mathbf{k}} - \omega_{\pm})^2}. \quad (6.6)$$

with

$$\begin{aligned} f(\phi_k, \theta) = (\gamma\gamma_{\text{NV}})^2 & \left(\sin^4 \phi_k + \frac{\sin^2 2\phi_k \cos^2 \theta}{4} + \sin^2 \phi_k \sin^2 \theta \right. \\ & \left. + \cos^4 \phi_k \cos^2 \theta + \cos^2 \phi_k \sin^2 \theta + \frac{\sin^2 2\phi_k}{4} \right) \end{aligned} \quad (6.7)$$

the geometric function containing all the explicit angular dependence of the integration.

Considering this, the shape of the resulting curves is modulated by parameters like η , J and M_{eff} , while its amplitude (its maximum value) is controlled by the saturation magnetization M_S .

First, we realize these simulations for a YIG thin film of $t_{\text{film}} = 8$ nm thick and effective magnetization given by $4\pi M_{\text{eff}} = -456$ G. At room temperature ($T = 300$ K) and taking $J = 2.42$ meV, we obtain Figure 6.4. Note that the peak of $\Gamma(\omega)$ occurs at the intersection between the NV resonance frequency ω_- and the ferromagnetic resonance $\omega_{\mathbf{k}=\mathbf{0}}$, indicating the point where both materials resonate simultaneously. This ferromagnetic resonance is given by the dispersion relation in equation (4.15) evaluated at $\mathbf{k} = \mathbf{0}$. In this case, the angle of the NV anisotropy axis, relative to $\hat{\mathbf{z}}$, is $\theta_H = 2\pi/5$, and it's aligned with the external magnetic field $\mathbf{H}_0$, which is to say, $\theta = \theta_H$ and $\theta'_B = 0$.

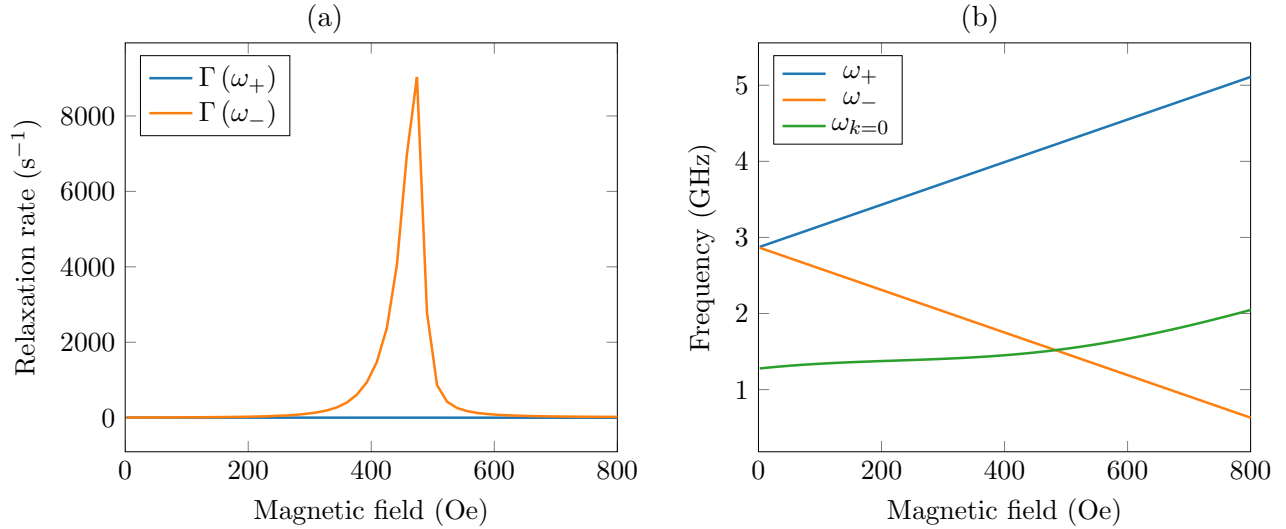


Figure 6.4: Relaxometry of a NV center located at distance $z = 239 \text{ nm}$ and with angle $\theta = 2\pi/5$ on a YIG (8 nm) sample with $4\pi M_{eff} = -456 \text{ G}$. (a) Relaxation rates $\Gamma(\omega_{\pm})$ versus external magnetic field. (b) NV and YIG resonance frequencies versus external magnetic field.

Our results are slightly different when we take a YIG film of $t_{\text{film}} = 12 \text{ nm}$ thick with $4\pi M_{eff} = -1489 \text{ G}$, as shown in Figure 6.5. Here, the ferromagnetic resonance $\omega_{\mathbf{k}=0}$ intersects the ω_+ frequency, instead of ω_- , resulting in a peak reached by $\Gamma(\omega_+)$.

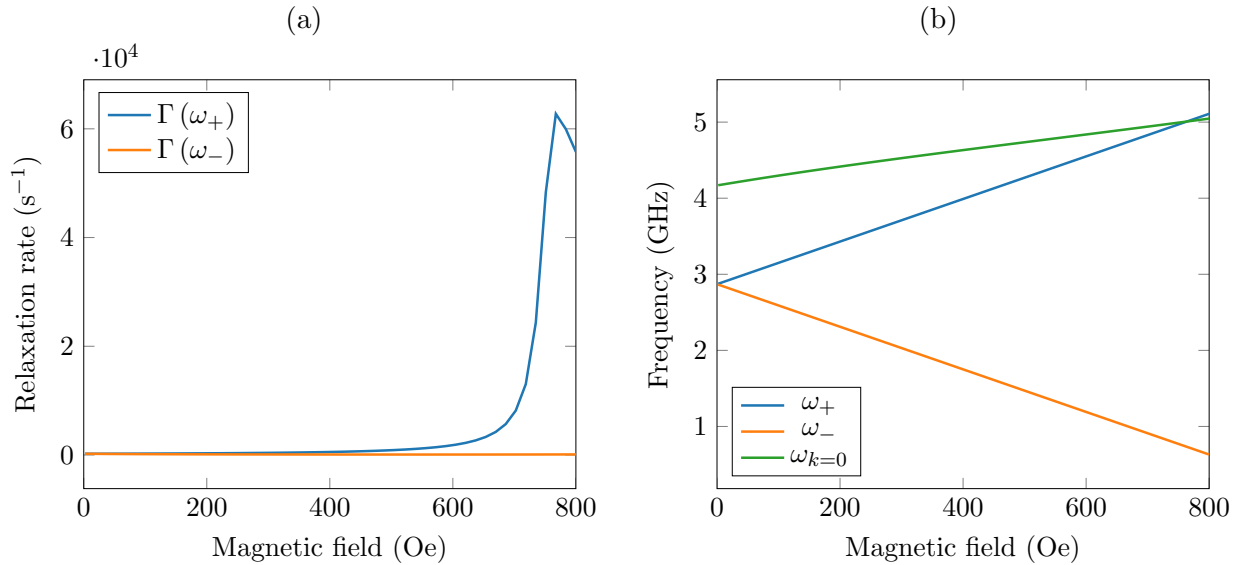


Figure 6.5: Relaxometry of a NV center located at distance $z = 114 \text{ nm}$ and with angle $\theta = 61\pi/180$ on a YIG (12 nm) sample with $4\pi M_{eff} = -1489 \text{ G}$. (a) Relaxation rates $\Gamma(\omega_{\pm})$ versus external magnetic field. (b) NV and YIG resonance frequencies versus external magnetic field.

Comparing with experimental data in the literature [17], we observe excellent agreement between this model and the measured relaxation rates fits very well the measured points (see Figure 6.6). Doing this experiment and fitting the curves of $\Gamma(\omega_{\pm})$, one can extract the saturation magnetization M_S , the effective magnetization M_{eff} and the exchange constant J .

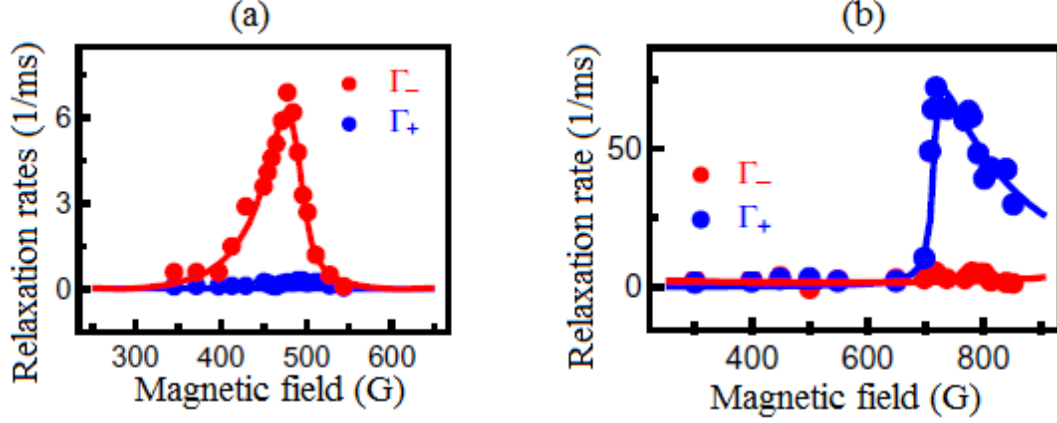


Figure 6.6: Experimental data for NV relaxation rates versus magnetic field H_0 , using both (a) YIG (8 nm) located at $z = 239$ nm with $4\pi M_{eff} = -456$ G, and (b) YIG (12 nm) located at $z = 114$ nm with $4\pi M_{eff} = -1489$ G [17].

6.3 Spin-spin correlations in antiferromagnetic systems

6.3.1 Transverse spin-spin correlation for AF systems

First, we calculate the transverse spin-spin correlation functions $C_{xx} = C_{yy}$, which by fluctuation-dissipation theorem (3.13) is given by

$$C_{xx}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi''_{xx}(\mathbf{k}, \omega), \quad (6.8)$$

where the linear response function in real space is

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{S}_x(\mathbf{r}, t), \hat{S}_x(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_x(\mathbf{r}, t), \hat{S}_x(\mathbf{r}', 0)] \rangle_{eq}. \end{aligned} \quad (6.9)$$

Here, σ is the dipole surface density $\hat{S}_x = (\hat{S}^+ + \hat{S}^-)/2$ and $\hat{S}_y = (\hat{S}^+ - \hat{S}^-)/(2i)$, so we may write the linear response function as

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{4\hbar} \theta(t) \langle [\hat{S}^+(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \\ &\quad + [\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^-(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

Note that the HP transformation for the spin operators $\hat{S}^\pm(\mathbf{r}, t)$ depends on the position $\mathbf{r}$. For instance, if $\mathbf{r}$ lies on the sublattice L_1 , we must expand $\hat{S}^+(\mathbf{r}, t)$ as $\sqrt{2s}\hat{a}(\mathbf{r}, t)$, while if $\mathbf{r} \in L_2$, we use $\hat{S}^+(\mathbf{r}, t) = \sqrt{2s}\hat{b}^\dagger(\mathbf{r}, t)$. Since $C_{\alpha\beta}(\mathbf{k}, \omega)$ is the Fourier transform of $C_{\alpha\beta}(\mathbf{r}, t)$ (and therefore, an integral over all the positions in the sample is performed), it is necessary to step on every case and sum them all. For example, if $\mathbf{r} \in L_1$ and $\mathbf{r}' \in L_1$, the needed HP transformations are for the sublattice L_1 , expressed in equation (5.4):

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2}\theta(t) \langle [\hat{a}(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \\ & + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

Now, expanding every raising and lowering operator in the Fourier space, we have

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2N}\theta(t) \sum_{\mathbf{k}' \in 1\text{BZ}} \sum_{\mathbf{k}'' \in 1\text{BZ}} \langle [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \rangle_{eq} \end{aligned}$$

and taking the Fourier transform for the relative position vector $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ (or equivalently², for $\mathbf{r}$ evaluating $\mathbf{r}' = \mathbf{0}$), in the convenient limit $\int d^2\mathbf{R} \rightarrow \frac{1}{\sigma} \sum_{\mathbf{R}}$, we obtain

$$\chi_{xx}(\mathbf{k}, t) = \frac{i\hbar\sigma^2}{2}\theta(t) \sum_{\mathbf{k}' \in 1\text{BZ}} \left\langle [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] \right\rangle_{eq}$$

The first and the last commutators vanish, while the two remaining must be expanded using the Bogoliubov transformations³

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2}\theta(t) \sum_{\mathbf{k}' \in 1\text{BZ}} \langle u_{\alpha\mathbf{k}}u_{\alpha\mathbf{k}'} [\hat{\alpha}_{\mathbf{k}}(t), \hat{\alpha}_{\mathbf{k}'}^\dagger(0)] - v_{\alpha\mathbf{k}}v_{\alpha\mathbf{k}'} [\hat{\beta}_{-\mathbf{k}'}(0), \hat{\beta}_{-\mathbf{k}}^\dagger(t)] \\ & - u_{\alpha\mathbf{k}}u_{\alpha\mathbf{k}'} [\hat{\alpha}_{\mathbf{k}'}(0), \hat{\alpha}_{-\mathbf{k}}^\dagger(t)] + v_{\alpha\mathbf{k}}v_{\alpha\mathbf{k}'} [\hat{\beta}_{\mathbf{k}}(t), \hat{\beta}_{-\mathbf{k}'}^\dagger(0)] \rangle_{eq} \end{aligned}$$

Now, using the characteristic raising and lowering operators evolution in the Heisenberg picture $\hat{\alpha}_{\mathbf{k}}(t) = \hat{\alpha}_{\mathbf{k}}(0) \exp(-i\omega_{\alpha\mathbf{k}}t - \eta t)$ and $\hat{\beta}_{\mathbf{k}}(t) = \hat{\beta}_{\mathbf{k}}(0) \exp(-i\omega_{\beta\mathbf{k}}t - \eta t)$, with $\eta > 0$, and remembering the commutation relations $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}'}^\dagger] = [\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$, we have

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2}\theta(t) [u_{\alpha\mathbf{k}}^2 \exp(-i\omega_{\alpha\mathbf{k}}t) - v_{\alpha\mathbf{k}}^2 \exp(i\omega_{\beta\mathbf{k}}t) \\ & - u_{\alpha\mathbf{k}}^2 \exp(i\omega_{\alpha\mathbf{k}}t) + v_{\alpha\mathbf{k}}^2 \exp(-i\omega_{\beta\mathbf{k}}t)] \exp(-\eta t) \end{aligned}$$

²In the long run, the result will be the same as using $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, since the system has translational symmetry.

³One can check this by expanding $\hat{a}_{\mathbf{k}}$ and $\hat{b}_{\mathbf{k}}$ with the Bogoliubov transformation, in a similar fashion to the method shown here, as the Bogoliubov coefficients are defined such that $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}}] = [\hat{\alpha}_{\mathbf{k}}^\dagger, \hat{\alpha}_{\mathbf{k}}^\dagger] = [\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}}] = [\hat{\beta}_{\mathbf{k}}^\dagger, \hat{\beta}_{\mathbf{k}}^\dagger] = 0$, and the mixed commutators are zero as well.

Finally, taking the remaining temporal Fourier transform, considering $\omega_{\alpha\mathbf{k}} > 0$ and $\omega_{\beta\mathbf{k}} > 0$, we finally obtain

$$\chi''_{xx}(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[u_{\alpha\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + v_{\alpha\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right]$$

with $\eta > 0$ a damping parameter for the spin wave. The other cases, like $\mathbf{r}, \mathbf{r}' \in L_2$ or the mixed one ($\mathbf{r} \in L_1$ and $\mathbf{r}' \in L_2$) are developed in Appendix D. The general procedure idea is quite similar to the one shown here. Adding up all the possible susceptibilities, we obtain

$$\chi''_{xx}(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[(u_{\alpha\mathbf{k}} - v_{\alpha\mathbf{k}})^2 \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + (u_{\beta\mathbf{k}} - v_{\beta\mathbf{k}})^2 \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right],$$

so, according to dissipation-fluctuation theorem (3.13), the transverse spin-spin correlation function $C_{xx}(\mathbf{k}, \omega) = C_{yy}(\mathbf{k}, \omega)$ for easy-plane anisotropic systems (like NiO) is:

$$C_{xx}(\mathbf{k}, \omega) = \frac{\hbar\sigma s k_B T}{\omega} \left[(u_{\alpha\mathbf{k}} - v_{\alpha\mathbf{k}})^2 \frac{\eta}{\eta^2 + (\omega_{\alpha\mathbf{k}} - \omega)^2} + (u_{\beta\mathbf{k}} - v_{\beta\mathbf{k}})^2 \frac{\eta}{\eta^2 + (\omega_{\beta\mathbf{k}} - \omega)^2} \right] \quad (6.10)$$

where σ is the dipole surface density, η is a spin-wave damping factor and the Bogoliubov coefficients $u_{\alpha(\beta)\mathbf{k}}$ and $v_{\alpha(\beta)\mathbf{k}}$ are given by (5.16). For uniaxial anisotropy (like MnF_2), the spin-spin transverse correlators are analogous, being the only difference that $u_{\alpha\mathbf{k}} = u_{\beta\mathbf{k}} = u_{\mathbf{k}}$ and $v_{\alpha\mathbf{k}} = v_{\beta\mathbf{k}} = v_{\mathbf{k}}$ and they are given by equations (5.6).

The transverse spin-spin correlators (6.10) corresponds to a sum of Lorentzians, one of them reaches its peak at $\omega = \omega_{\alpha\mathbf{k}}$, while the other one, at $\omega = \omega_{\beta\mathbf{k}}$ (see Figure 6.1). Those limits can be interpreted as NV-magnon *resonances*. Far away from both peaks, the correlator decays to 0. However, we mentioned before that, for both MnF_2 and NiO, the dispersion relation $\omega_{\alpha(\beta)\mathbf{k}}$ is of the order of THz, while ω is just of GHz, which is to say, $\omega_{\alpha(\beta)\mathbf{k}} \gg \omega$. Therefore, the spin-spin transverse correlator C_{xx} doesn't contribute significantly to the integral in equation (3.12). This is just the opposite as the ferromagnetic case, for materials like YIG [17], whose magnon frequencies are of similar order to the NV resonance frequency.

6.3.2 Longitudinal spin-spin correlation for AF systems

A realistic calculation of $\Gamma(\omega)$ needs the longitudinal correlation $C_{zz}(k, \omega)$. Doing this by equation (3.14) is a much longer procedure, due to the bi-linear form of the HP transformations (5.4) and (5.5). Here, the desired spin-spin correlator is

$$C_{zz}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi''_{zz}(\mathbf{k}, \omega) \quad (6.11)$$

where the response linear function is given by

$$\begin{aligned}\chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_z(\mathbf{r}, t), \hat{s}_z(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_z(\mathbf{r}, t), \hat{S}_z(\mathbf{r}', 0)] \rangle_{eq}.\end{aligned}$$

As in the transverse correlator computation, it is necessary to split the procedure in several cases for the positions $\mathbf{r}$ and $\mathbf{r}'$, and finally sum them all. So, we have to calculate the linear response functions in four parts, after applying the HP (5.4) and (5.5) transformations for each case:

$$\begin{aligned}\chi_{zz}^{(11)}(\mathbf{r}, \mathbf{r}'; t) &= i\hbar\sigma^2\theta(t) \langle [\hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}(\mathbf{r}', 0)] \rangle_{eq} \\ \chi_{zz}^{(12)}(\mathbf{r}, \mathbf{r}'; t) &= -i\hbar\sigma^2\theta(t) \langle [\hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0) \hat{b}(\mathbf{r}', 0)] \rangle_{eq} \\ \chi_{zz}^{(21)}(\mathbf{r}, \mathbf{r}'; t) &= -i\hbar\sigma^2\theta(t) \langle [\hat{b}^\dagger(\mathbf{r}, t) \hat{b}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}(\mathbf{r}', 0)] \rangle_{eq} \\ \chi_{zz}^{(22)}(\mathbf{r}, \mathbf{r}'; t) &= i\hbar\sigma^2\theta(t) \langle [\hat{b}^\dagger(\mathbf{r}, t) \hat{b}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0) \hat{b}(\mathbf{r}', 0)] \rangle_{eq}.\end{aligned}$$

Each of these response functions is associated with a many-particle correlation function. The required steps to calculate these terms are similar to those explained in Section 6.1.2, being the only difference that now it is necessary to expand the raising and lowering operators using the Bogoliubov transformations. The full procedure can be found in Appendix D. For example, we can split $\chi_{zz}^{(11)}$ in four terms using the product rule. Next, expanding the operators $\hat{a}$ and $\hat{a}^\dagger$ in the Fourier space, and using the Bogoliubov transformations, we obtain the imaginary part of the response function in $\mathbf{k}$ space. The final susceptibility in Fourier space is the sum of all the possible cases for $\mathbf{r}, \mathbf{r}'$:

$$\chi_{zz}'' = \chi_{zz}''^{(11)} + \chi_{zz}''^{(12)} + \chi_{zz}''^{(21)} + \chi_{zz}''^{(22)}.$$

After finding all of these terms and reducing them in the limit $\eta \rightarrow 0^+$, thanks to the identity

$$\lim_{\eta \rightarrow 0^+} \frac{1}{\pi} \frac{\eta}{\eta^2 + (\omega - \omega_0)^2} = \delta(\omega - \omega_0),$$

and taking the continuous limit $\sum_{\mathbf{k}' \in 1\text{BZ}} \rightarrow \frac{N}{\sigma(2\pi)^2} \int d^2\mathbf{k}'$, we obtain

$$\begin{aligned}\chi_{zz}''(\mathbf{k}, \omega) &= \frac{\hbar}{4\pi} \int_{1\text{BZ}} d^2\mathbf{k}' \{ \\ &\quad [u_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega_{\alpha\mathbf{k}'}) + v_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\ &\quad \times [n_B(\hbar\omega_{\alpha\mathbf{k}'} - \mu_\alpha) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \mu_\alpha)] \\ &\quad + [u_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega_{\beta\mathbf{k}'}) + v_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\ &\quad \times [n_B(\hbar\omega_{\beta\mathbf{k}'} - \mu_\beta) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \mu_\beta)] \},\end{aligned}$$

being μ_α and μ_β the chemical potentials for the respective modes α and β . In general, α and β magnons behave statistically similar to particle-antiparticle couples, in the sense that $\mu_\alpha = -\mu_\beta = \mu$ [41]. These chemical potentials increase in magnitude as the drive magnetic field $\mathbf{B}_{AC}(t)$, used to generate spin excitations and control the NV spin state, does. So, the longitudinal spin-spin correlator, according to dissipation-fluctuation theorem (3.13), is

$$\begin{aligned}
C_{zz}(\mathbf{k}, \omega) = & \frac{\hbar k_B T}{2\pi\omega} \int_{\text{1BZ}} d^2\mathbf{k}' \{ \\
& [u_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega_{\alpha\mathbf{k}'}) + v_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\alpha\mathbf{k}'} - \mu_\alpha) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \mu_\alpha)] \\
& + [u_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega_{\beta\mathbf{k}'}) + v_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\beta\mathbf{k}'} - \mu_\beta) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \mu_\beta)] \}
\end{aligned} \tag{6.12}$$

This is the quantity that dominates the relaxation rates in antiferromagnetic systems. Note that the Dirac deltas cancel out one of the two integrals present here. Those deltas represent energy conservation for the possible magnon pairs, exchanging energy $\hbar\omega$ along with the NV center.

Unfortunately, computing the integral (6.12) might not be easy. In fact, for the dispersion relations of antiferromagnetic magnons, this integral cannot be exactly calculated, and numerical methods are required. However, dispersion relations of the form

$$\hbar\omega_{\alpha(\beta)\mathbf{k}} = \frac{\hbar^2 k^2}{2m_{\alpha(\beta)}} + m_{\alpha(\beta)} c_{\alpha(\beta)}^2 + U_{\alpha(\beta)}$$

allow the integral to be explicitly solved, giving the result

$$\begin{aligned}
C_{zz}(k, \omega) = & \frac{(k_B T)^2}{2\omega} \{ m_\alpha \left[\frac{\hbar\tilde{\omega}}{\mu_\alpha} \left(\frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4(V_\alpha - \mu_\alpha) A_{\alpha(\beta)} k^2}} - \frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4V_\alpha A_{\alpha(\beta)} k^2}} \right) \right. \right. \\
& \left. \left. (\mu_\alpha \rightarrow \mu_\alpha - \hbar\omega) \right) \right. \\
& \left. + \frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4(V_\alpha - \mu_\alpha) A_{\alpha(\beta)} k^2}} - \frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4(V_\alpha + \hbar\omega - \mu_\alpha) A_{\alpha(\beta)} k^2}} \right. \\
& \left. \left. (\omega \rightarrow -\omega) \right] \right\} + (\alpha \leftrightarrow \beta)
\end{aligned} \tag{6.13}$$

$$\begin{aligned}
& + \frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4(V_\alpha - \mu_\alpha) A_{\alpha(\beta)} k^2}} - \frac{1}{\sqrt{(\hbar\omega - A_{\alpha(\beta)} k^2)^2 + 4(V_\alpha + \hbar\omega - \mu_\alpha) A_{\alpha(\beta)} k^2}} \\
& (\omega \rightarrow -\omega) \} + (\alpha \leftrightarrow \beta)
\end{aligned} \tag{6.14}$$

being $V_{\alpha(\beta)} = m_{\alpha(\beta)} c_{\alpha(\beta)}^2 + U_{\alpha(\beta)}$ the full energy gap (considering both mass term and magnetic field isolated term, if any) and $A_{\alpha(\beta)} = \hbar^2 / (2m_{\alpha(\beta)})$. So, it is possible to realize the approximations (5.14) and (5.20) already presented in Chapter 5. In Figure 6.7 we plot both correlation functions (transverse

and longitudinal) of an easy-axis antiferromagnet, using this non-relativistic approximation (5.14), and how this is compared to the numerical calculation (which uses the exact dispersion relation (5.7)).

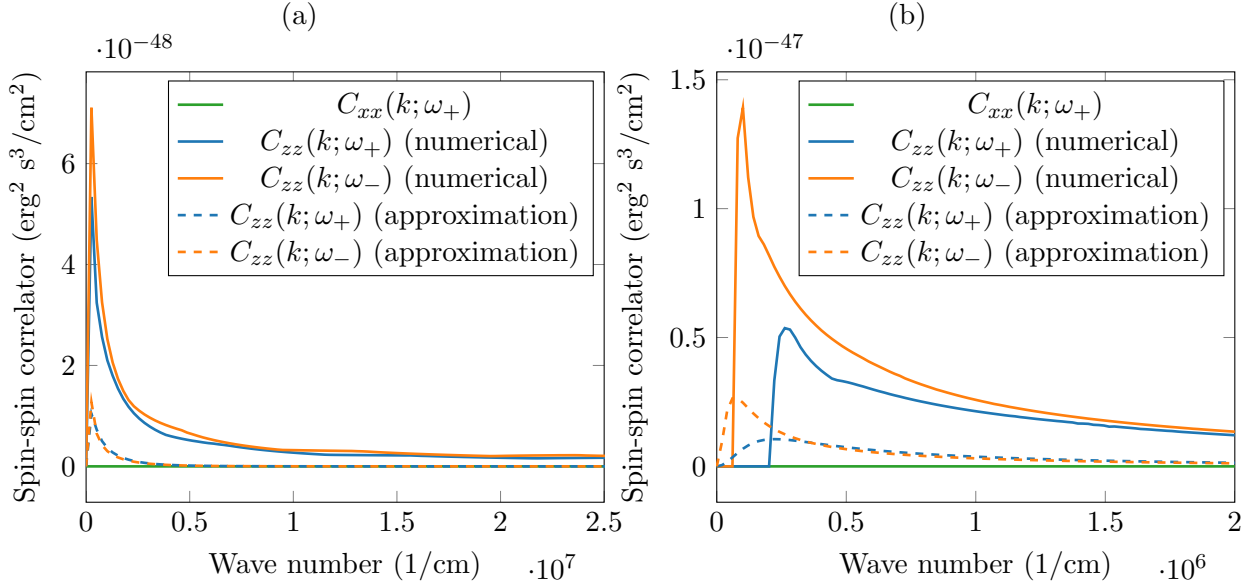


Figure 6.7: Spin-spin correlation function for an easy-axis antiferromagnetic sample, using numerical methods for the dispersion relation (5.7), and using the non-relativistic approximation which leads to equation (6.14). (a) Graph using as domain $[0, \pi/a]$. (b) The same graph, but using a zoomed domain $[0, 10/z]$.

We note that $C_{zz} \gg C_{xx}$, which confirms our hypothesis that C_{zz} dominates the relaxation rates. The approximation (6.14) is in the same order of magnitude than the numerical simulation, being the first one smaller in general (because the dispersion relation is bigger). However, the numerical simulation reveals an interesting behavior that the approximation formula does not: for small k , the correlation function is exactly 0. This means that spins with small wave vectors $\mathbf{k}$ does not correlate, or more precisely, won't contribute to the real spin-spin correlation.

The numerical simulations for the longitudinal correlation were done in Python with the following procedure:

1. In equation (6.12), we evaluate at $\mathbf{k} = k\hat{\mathbf{y}}''$ (in this way, we choose the y'' direction so that it coincides with $\mathbf{k}$). Therefore, $\mathbf{k} + \mathbf{q} = q_x\hat{\mathbf{x}}'' + (q_y + k)\hat{\mathbf{y}}''$.
2. We separate the expression in four terms with integrals. For example, one of the four terms is

$$\int_{-\pi/a}^{\pi/a} \int_{-\pi/a}^{\pi/a} dq_x dq_y u_{\alpha\mathbf{q}}^2 \delta(\omega - \omega_{\alpha(\mathbf{q}+\mathbf{k})} + \omega_{\alpha\mathbf{q}}) [n_B(\hbar\omega_{\alpha\mathbf{q}}) - n_B(\hbar\omega_{\alpha\mathbf{q}} + \hbar\omega)]$$

For each term, we find numerically the roots of the expression inside the Dirac delta, in this case, $\omega - \omega_{\alpha(\mathbf{q}+\mathbf{k})} + \omega_{\alpha\mathbf{q}}$. Let one of these roots to be q_y^* and $\mathbf{q}^* = q_x \hat{\mathbf{x}}'' + q_y^* \hat{\mathbf{y}}''$, so the integral previously shown reduces to

$$\int_{-\pi/a}^{\pi/a} dq_x \sum_{q_y^*} u_{\alpha\mathbf{q}^*}^2 \left| \frac{\partial \omega_{\alpha\mathbf{q}}}{\partial q_y} \right|_{q_y=q_y^*} - \left| \frac{\partial \omega_{\alpha(\mathbf{q}+\mathbf{k})}}{\partial q_y} \right|_{q_y=q_y^*} \right|^{-1} [n_B(\hbar\omega_{\alpha\mathbf{q}^*}) - n_B(\hbar\omega_{\alpha\mathbf{q}^*} + \hbar\omega)]$$

In this program, we divide the one-dimensional projection of the first Brillouin zone $[-\frac{\pi}{a}, \frac{\pi}{a}]$ in 10 subintervals, and if a change of sign in the total energy $\hbar(\omega - \omega_{\alpha(\mathbf{q}+\mathbf{k})} + \omega_{\alpha\mathbf{q}})$ is detected in one of those subintervals, Brent's method is used to find the respective root (*scipy.optimize.brentq*) [42].

- Finally, we realize the integral in q_x . Particularly, graphs in Figure 6.7 were created by doing the integration with the function *scipy.integrate.quad* of Python. For each value of k and ω , this calculation of $C_{zz}(k, \omega)$ takes approximately 10 seconds. Therefore, each subfigure (a) and (b) in Figure 6.7 took several minutes to generate, as we used 100 different values of k to plot (50 for ω_+ , and the other 50 for ω_-).

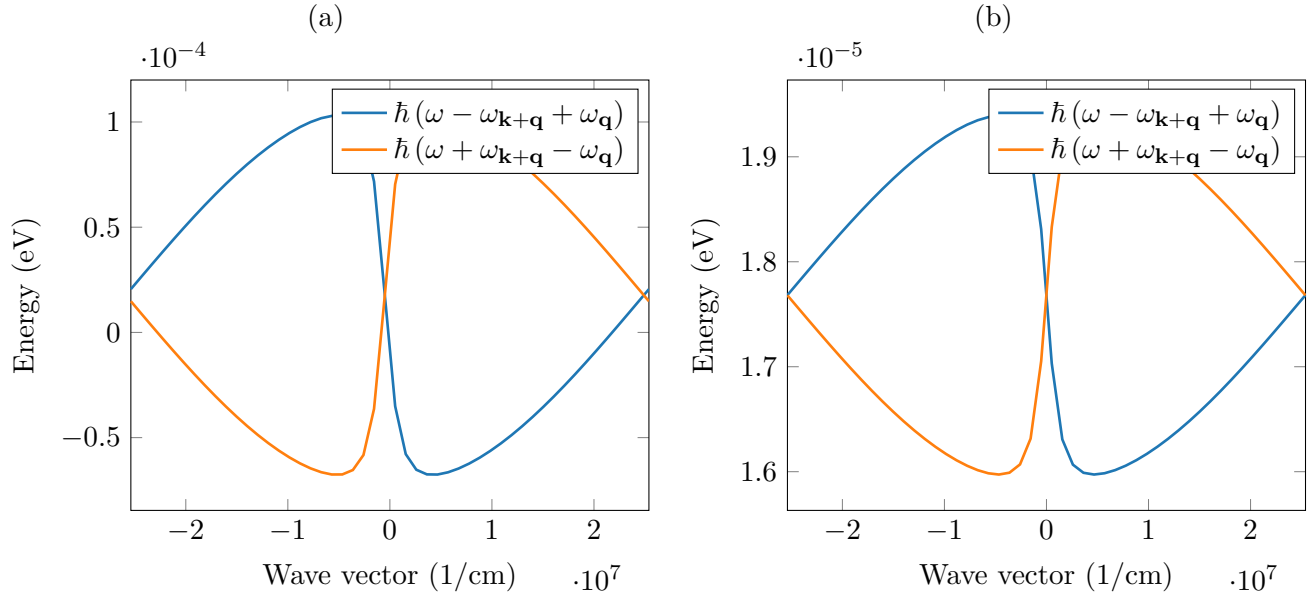


Figure 6.8: Two-magnon total energy $\hbar\omega - \hbar\omega_{\alpha(\mathbf{q}+\mathbf{k})} + \hbar\omega_{\alpha\mathbf{q}}$ restricted to be zero in the Dirac deltas of equation (6.12), versus q_y . α mode only. (a) Plot generated using $q_x = 0$ and $k = 5/z$, one can see two roots. (b) Plot generated using $q_x = 0$, $k = 0.1/z$, no roots at all. For both cases, $z = 50$ nm and $\omega = D_{\text{NV}} + \gamma H_0$ ($H_0 = 500$ G).

6.4 Relaxation rates using antiferromagnetic samples

In the antiferromagnetic case, the relaxation rates is dominated by the longitudinal spin-spin correlation function C_{zz} given by equation (6.12). As none of the dispersion relations mentioned in Chapter 5 depend on the magnon angle ϕ_k , using the simplified version (3.12) of $\Gamma(\omega)$, with $C_{xx} \rightarrow 0$, is enough. Creating the simulations for $\Gamma(\omega_{\pm})$ is already a highly demanding computational task if we use the numerical methods mentioned in previous sections; it may take hours or even full days to plot a single graphic.

In the model of this work, it is enough to indicate the parameters of anisotropy D_z and D_x and the exchange constant J_2 to give a full description of the sample. It is a common practice in literature to directly report the effective fields

$$H_E = \frac{2sZ|J_2|}{\hbar\gamma} \quad , \quad H_{Ax} = \frac{2sD_x}{\hbar\gamma} \quad , \quad H_{Az} = \frac{2sD_z}{\hbar\gamma}$$

defined in Chapter 5. First, we evaluate the obtained expression for $\Gamma(\omega_{\pm})$ in (3.12) considering the easy-axis antiferromagnet MnF_2 , as its parameters $H_E = 526 \text{ kOe}$, $H_{Az} = 8.2 \text{ kOe}$ and $H_{Ax} = 0$ are well-known [5]. Simulations for these relaxation rates were done numerically using the procedure mentioned in the previous sections to compute C_{zz} , and the results are shown in Figure (6.9a), for temperature⁴ $T = 50 \text{ K}$ and neglecting chemical potential. These relaxation rates are of the order of 10 s^{-1} , which can be barely detected using relaxometry. NV centers have intrinsic relaxation rates whose magnitude order oscillate around 10 s^{-1} . Effects in diamond like strain, temperature and accumulation of NV centers can reduce or elevate even more this intrinsic relaxation rate [43], but hardly it goes down to the order of $1 \times 10^{-1} \text{ s}^{-1}$. Thus, MnF_2 is almost out of the sensitivity limits of NV centers, even using a relatively short NV-to-sample distance of $z = 30 \text{ nm}$.

Nevertheless, it is possible to elevate relaxation rates' orders of magnitude by manipulating the magnon chemical potential μ . Experimentally, this can be done by applying a drive magnetic field $\mathbf{B}_{AC}(t)$ [32]. This radiation, if resonates with one of the antiferromagnetic resonance frequencies, either $\omega_{\alpha, \mathbf{k}=0} = V_{\alpha}$ or $\omega_{\beta, \mathbf{k}=0} = V_{\beta}$, pumps respectively α or β magnons into the sample [41], and therefore more intense spin fluctuations. In this case, we assume the drive field to be left-handed circularly polarized⁵ and transverse

⁴According to (6.12) and (6.14), higher temperatures would result in higher relaxation rates $\Gamma(\omega_{\pm})$. However, MnF_2 presents a Neel temperature of $T_N = 66.5 \text{ K}$ [33], so making these simulations at room temperature $T = 300 \text{ K}$ would be unrealistic.

⁵A left-handed polarized, frequency $\omega_{\beta, \mathbf{k}=0}$ microwave triggers β -magnon creation with $\mu > 0$, while right-handed polarized drive fields with frequency $\omega_{\alpha, \mathbf{k}=0}$ pump α -magnons with $\mu < 0$. Here, we consider only the first case as $\omega_{\beta, \mathbf{k}=0} < \omega_{\alpha, \mathbf{k}=0}$.

to the equilibrium orientation of the spins, so

$$\mathbf{B}_{AC}(t) = B_{AC} [\cos(\omega_{\beta, \mathbf{k}=\mathbf{0}}t)\hat{\mathbf{x}} - \sin(\omega_{\beta, \mathbf{k}=\mathbf{0}}t)\hat{\mathbf{y}}].$$

Given this microwave, it is possible to show that $\mu \propto B_{AC}^2$ [32, 41]. Thus, the greater drive magnetic field, the greater μ . Particularly, if μ is near the magnon condensation point ($\mu \approx V_\beta$ for $\mu < V_\beta$), the orders of magnitude of the relaxation rates increase drastically, as shown in Figure (6.9b), where we have evaluated⁶ MnF_2 at $\mu = 0.985mc^2$. Elevating the drive magnetic field is the key for obtaining measurable relaxation rates using antiferromagnetic systems.

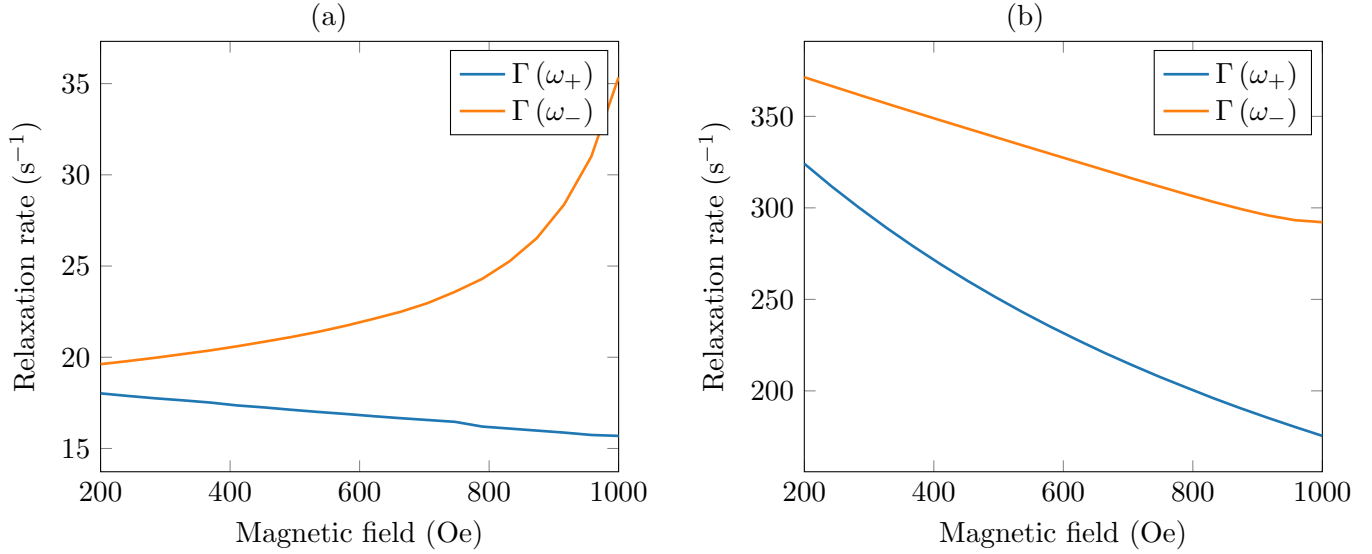


Figure 6.9: Relaxation rates $\Gamma(\omega_{\pm})$ versus external magnetic field H_0 , at temperature $T = 50$ K, using a MnF_2 sample at distance $z = 30$ nm from the NV center. (a) For chemical potential $\mu = 0$. (b) $\mu = 0.985mc^2$. For both cases, equation (6.12) was used for the longitudinal spin-spin correlation and numerical integration was performed.

The *only* problem of this drive field technique is that we require μ close to mc^2 , whose values may be extremely large. For instance, $mc^2/\hbar \sim \text{THz}$ for MnF_2 , so reaching a chemical potential of $0.985mc^2$ would require a field amplitude of $B_{AC} \sim mc^2/(\hbar\gamma) > 5$ T. However, for easy-axis antiferromagnets with low anisotropy, such as RbMnF_3 [34], using drive fields would be a good addition to increase $\Gamma(\omega_{\pm})$.

On the other hand, using as a sample the easy-plane antiferromagnet NiO , whose parameters are also well-known (exchange field $H_E = 9684$ kOe, easy-axis anisotropy field $H_{Az} = 0.11$ kOe and hard-axis

⁶Actually, β -magnon energy gap is $V_\beta = mc^2 - \hbar\gamma H_0$. However, for MnF_2 , $\hbar\gamma H_0$ is smaller than $0.015mc^2$ even at $H_0 = 1000$ Oe, so $0.985mc^2$ is a good choice for the chemical potential, near to mc^2 and does not intersect the magnon dispersion relations $\hbar\omega_{\alpha(\beta)\mathbf{k}}$

anisotropy field $H_{Ax} = 6.35$ kOe) [19], we obtain a similar graph, but with extremely low relaxation rates, as illustrated in Figure (6.10b). These values for $\Gamma(\omega_{\pm})$ are definitely unreadable and will be overshadowed by the NV intrinsic relaxation rate, whose magnitude order oscillates around 10 s^{-1} , and increasing the relaxation rates controlling chemical potential is unpractical because of the high drive fields required. We conclude that NiO is not a material that can be used for this experiment. In terms of the sample, these problems occur mainly because NiO has a very high effective exchange field H_E , which *freezes* the system dynamics, making it inert against the NV center.

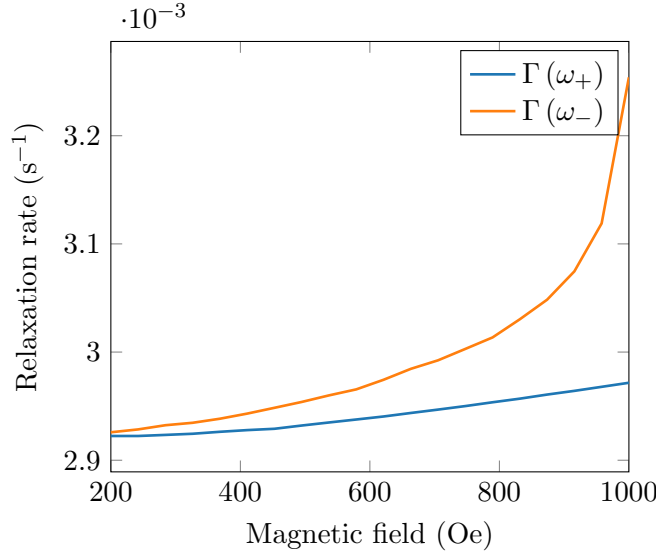


Figure 6.10: Relaxation rates $\Gamma(\omega_{\pm})$ versus external magnetic field H_0 , at temperature $T = 300$ K, using a NiO sample at a distance $z = 100$ nm. Equation (6.12) was used for the longitudinal spin-spin correlation and numerical integration was performed.

A valid question that now arises is: for what kind of antiferromagnetic materials will this technique work. One can conclude that antiferromagnetic materials should have a low $|J_2|$, and therefore, a low exchange field $H_E = 2sZ|J_2|/(\hbar\gamma)$ defined in equation (5.22). In fact, this is the main influence on the values that $\Gamma(\omega)$ takes (see Figure 6.11). Other parameter that one should be careful about is the anisotropy constant D_z which defines how big the gap $m_{\alpha(\beta)}c_{\alpha(\beta)}^2$ is in the dispersion relation $\hbar\omega_{\alpha(\beta)\mathbf{k}}$. So, the product $\sqrt{J_2 D_z}$, which defines the spin-flop field $H_{SF} := \sqrt{2H_E H_{Az} - H_{Az}^2} \approx \sqrt{2H_E H_{Az}}$ should not be so big. A material that stands out for its relatively low spin-flop (which can be smaller than 1 T [44]) is the hematite $\alpha\text{-Fe}_2\text{O}_3$, already mentioned in Chapter 5 when discussed phase transitions, specifically Morin transition.

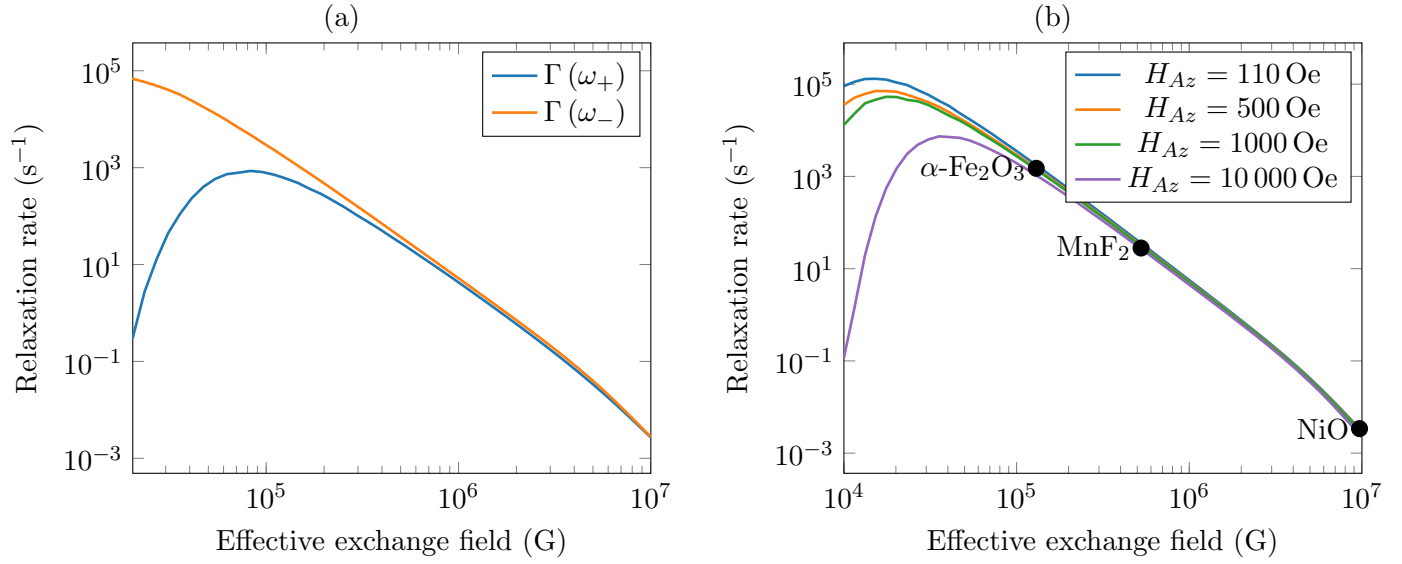


Figure 6.11: Relaxation rates $\Gamma(\omega_{\pm})$ versus effective exchange field H_E , using NV-to-sample distance $z = 50$ nm. (a) Both relaxation rates, using fixed anisotropy fields $H_{Az} = 2sD_z/(\hbar\gamma) = 500$ Oe and $H_{Ax} = 0$. (b) Only $\Gamma(\omega_-)$, for several values of H_{Az} and fixed $H_{Ax} = 0$. Both graphs are shown in $\log_{10} - \log_{10}$ scale, as we want to analyze changes in the relaxation rates order of magnitude.

The Morin temperature $T_M = 263$ K defines a phase transition from which the system rotates its easy-axis, as explained in Chapter 5. For $T < T_M$, the system behaves as a easy-axis antiferromagnet, while for $T > T_M$, it is in a canted phase. For this material in the antiferromagnetic (AF) phase, we take the parameters $H_{Az} = 0.5$ kOe and $H_E = 130$ kOe, so that $H_{SF} \approx \sqrt{2H_E H_{Az}} \approx 1.1$ T coincides with the value we take as reference for the spin-flop field [18]. In general, this Morin transition may change the value of the anisotropy field H_{Az} , so we took $H'_{Az} = 5$ Oe for $T > T_M$. Simulations for the relaxation rates $\Gamma(\omega_{\pm})$ are shown in Figure (6.12a), for temperature $T < T_M$.

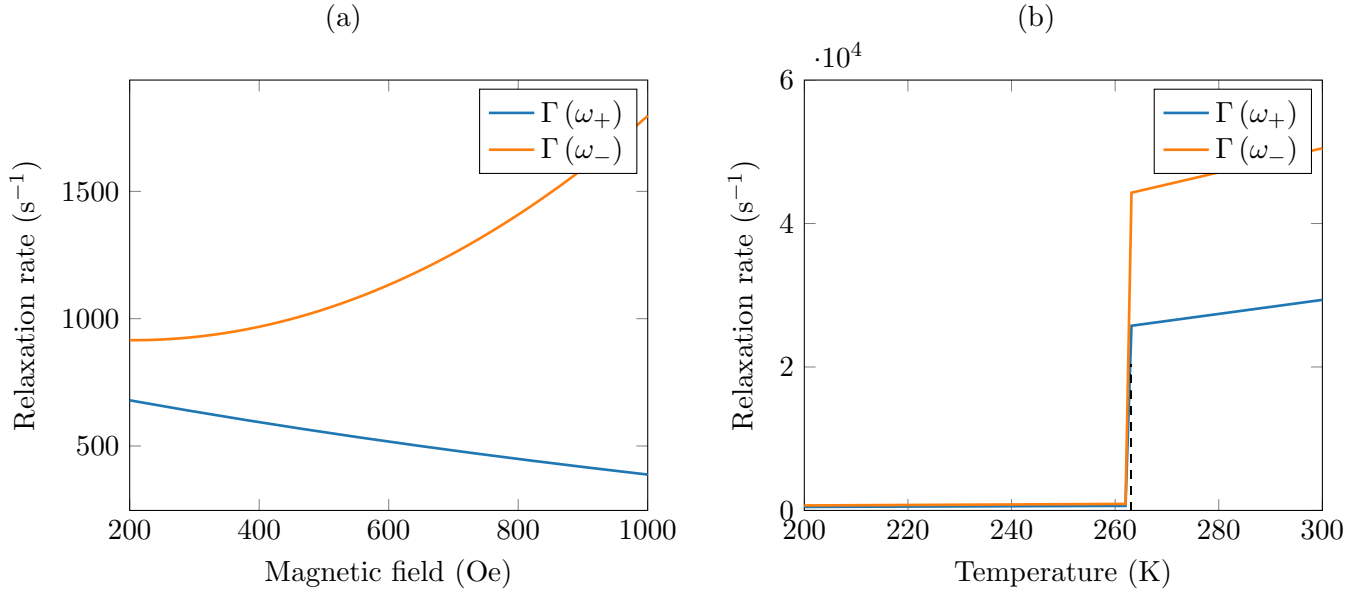


Figure 6.12: Relaxation rates $\Gamma(\omega_{\pm})$ simulations for a NV center located at $z = 50$ nm from a hematite $\alpha\text{-Fe}_2\text{O}_3$ sample. (a) Relaxation rates versus magnetic field H_0 at temperature $T = 250$ K. (b) Relaxation rates versus temperature T in a fixed magnetic field $H_0 = 500$ Oe. Here, equation (5.7) was used for the dispersion relation in the antiferromagnetic phase ($T < T_M$), while (5.25) was used in canted phase ($T > T_M$). For both graphs, numerical integration was performed for the longitudinal spin-spin correlation (6.12).

The previous plot was created considering a fixed temperature. An interesting question is: how the NV center reacts as T approaches to the Morin transition at $T = T_M$. Figure (6.12b) responds to this question by displaying a plot of the relaxation rates $\Gamma(\omega_{\pm})$ taking T as a variable. We observe that $\Gamma(\omega_-)$ increases drastically when $T = T_M$, while $\Gamma(\omega_+)$ barely reacts. In this way, NV centers can detect phase transitions of antiferromagnetic materials, and quantitatively measuring their relaxation rates one can determine at which temperature these transitions occur, as well as the fitting parameters $H_E \propto J_2$ and $H_{Az} \propto D_z$.

In summary, relaxation rates can be calculated for both ferromagnetic and antiferromagnetic systems. The main goal of this chapter to calculate the relaxation rates $\Gamma(\omega)$ using these systems as samples, and we did it using the equation (3.12) found in Chapter 3, along with the formalism presented in Chapters 4 and 5 to describe magnetic materials. We calculated the spin-spin correlation functions for each case. In ferromagnets, as the resonance frequency $\omega_{\mathbf{k}=0}$ is of the order of GHz (similar to $\omega_{\pm}$), the relaxation rates $\Gamma(\omega_{\pm})$ are dominated by the transverse spin-spin correlation C_{xx} , and therefore $\Gamma_{\pm}(\mathbf{H}_0)$ peaks at the

magnetic field $\mathbf{H}_0$ where $\omega_+ = \omega_{\mathbf{k}=\mathbf{0}}$ or $\omega_- = \omega_{\mathbf{k}=\mathbf{0}}$. In this way, it is possible to characterize ferromagnetic samples by reading their dispersion relation via relaxometry, obtaining with this fitting parameters like the effective magnetization M_{eff} , the saturation magnetization M_S and the spin-spin exchange constant $J > 0$. On the other hand, antiferromagnetic systems have typically resonance frequencies of the order of THz, so the relaxation rates are dominated by the longitudinal spin-spin correlation function C_{zz} . We observed that not always it is possible to characterize antiferromagnetic samples using NV center relaxometry. Some materials with extremely high effective exchange field H_E , like NiO, are inert against the NV center.

Relaxometry with nitrogen-vacancy centers is a novel technique and its possibilities are still being studied. For future work, it would be interesting to study the interaction between a NV center and antiferromagnetic material with other types of geometry. For instance, triangular-lattice antiferromagnets, like Cs_2CuCl_4 (see Figure 6.13) [45], present non-trivial spin orientations and can form what is called a “frustrated system”. We expect the relaxometry technique to detect this sample, as $s = 1/2$ and its exchange constants J and J' are of the order of $J = 0.37 \text{ meV}$ and $J'/J = 0.34$ [46]. In general, the interaction between NV centers and low-dimension systems (quasi-1D and 2D materials) is a whole open world.

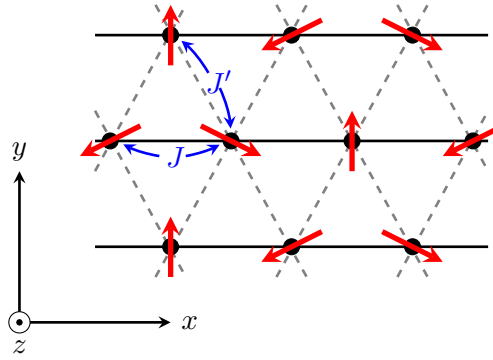


Figure 6.13: Crystal structure and spin orientation in the triangular-lattice antiferromagnet Cs_2CuCl_4 .

Other interesting topic that arises from the analysis of hematite ($\alpha\text{-Fe}_2\text{O}_3$) is the temperature dependence of the relaxation rates. We have already seen that a plot of $\Gamma(\omega_{\pm})$ versus temperature T is able to reveal phase transitions (in this case, Morin transitions). Some materials, like ferrimagnets, present magnetic properties with interesting temperature dependences. Particularly, ferrimagnets exhibit two spin sublattices, just as antiferromagnets, but with one spin orientation dominating over the other (in terms of equations (5.3) and (5.15), $\mathbf{S}(\mathbf{r}_1) > \mathbf{S}(\mathbf{r}_2)$), producing a net magnetization [6]. However, there exists a compensation temperature where both magnetic moments are equal and cancel each other,

creating a similar system to antiferromagnets, but with lower resonance frequencies that can intersect $\omega_{\pm}$. In this way, NV center relaxometry would be able to differentiate between a natural antiferromagnet and a compensated ferrimagnet, just by characterizing their resonance frequencies. In general, this is the quality that makes relaxometry a powerful characterization technique.

Chapter 7

Conclusions and outlook

As conclusion, nitrogen-vacancy centers provide an excellent way for measuring physical quantities of magnetic materials. The main goal of this work was to describe theoretically how the NV center reacts against several types of magnetic materials (either ferro or antiferromagnetic). This was done by calculating the relaxation rates $\Gamma(\omega_{\pm})$ using quantum perturbation theory, in such a way those $\Gamma(\omega_{\pm})$ were expressed as a function of the magnetic perturbations. These perturbations are actually produced by spin perturbations, whose correlation functions are purely determined by the physical properties of the sample. In this way, the work of calculating $\Gamma(\omega_{\pm})$ was reduced to finding spin-spin correlation functions.

We found that, for ferromagnetic systems whose resonance was of the order of GHz, the transverse spin-spin correlation dominates over the longitudinal, so the relaxation rates peak at the field where the NV center and the sample resonate simultaneously, occurring one-magnon creation-annihilation processes. On the other hand, in antiferromagnetic systems, whose magnon energy gap was of the order of THz, the main contribution to $\Gamma(\omega_{\pm})$ was the longitudinal spin-spin correlation. Unlike using ferromagnetic systems, relaxometry with antiferromagnetic does not exhibit a characteristic peak in its relaxation rates, being this the main difference between both materials. An analytic and exact calculation of the relaxation rates under these circumstances was not possible, and was necessary to develop approximations or numerical methods. Moreover, we found that not all materials produce readable relaxation rates. Due to its extremely high spin coupling parameter, NiO is almost inert against this relaxometry technique.

Despite these difficulties, we found that hematite ($\alpha\text{-Fe}_2\text{O}_3$) presents required parameters for using as a readable sample. We developed simulations of how the relaxation rates vary as a function of both the external magnetic field and the temperature, and found that NV centers can read phase transitions (in this case, Morin transition) that occurs in this sample.

We achieved the goal of characterizing the relaxation of NV centers near ferromagnetic and antiferromagnetic materials, finding the mentioned differences between both cases. It would be interesting to see how this NV center reacts against ferrimagnetic materials, which exhibit interesting temperature dependence, or other spin geometries in antiferromagnetic systems. For instance, triangular-lattice antiferromagnets, like Cs_2CuCl_4 , present non-trivial spin orientations and can form a frustrated system. In general, the study of interaction between NV centers and magnetic materials like ferromagnets, ferrimagnets and antiferromagnets (in all of their possible geometries and dimensions) is an open and interesting topic to explore.

Appendix A

Interaction between magnetic dipoles

Two magnetic dipoles, with dipolar magnetic moments $\boldsymbol{\mu}_1$ and $\boldsymbol{\mu}_2$, will have an interaction energy [22] given by the well-known hamiltonian (in CGS units):

$$H_{12} = \frac{\boldsymbol{\mu}_1 \cdot \boldsymbol{\mu}_2}{r^3} - \frac{3(\boldsymbol{\mu}_1 \cdot \mathbf{r})(\boldsymbol{\mu}_2 \cdot \mathbf{r})}{r^5},$$

where $\mathbf{r}$ is the vector joining μ_2 and μ_1 . It is possible to use the macrospin approximation $\boldsymbol{\mu}_1 = g_s \mu_B \mathbf{S}_1 / \hbar$, being $g_s \approx 2$, μ_B the Bohr magneton, $\hbar$ the Planck's constant and $\mathbf{S}_1$ is the spin angular momentum. Similarly for $\boldsymbol{\mu}_2 = g_s \mu_B \mathbf{S}_2 / \hbar$.

$$\Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{r^5} (r^2 \mathbf{S}_1 \cdot \mathbf{S}_2 - 3(\mathbf{S}_1 \cdot \mathbf{r})(\mathbf{S}_2 \cdot \mathbf{r})).$$

Expanding $\mathbf{S}_1 = S_{1x}\hat{\mathbf{x}} + S_{1y}\hat{\mathbf{y}} + S_{1z}\hat{\mathbf{z}}$, $\mathbf{S}_2 = S_{2x}\hat{\mathbf{x}} + S_{2y}\hat{\mathbf{y}} + S_{2z}\hat{\mathbf{z}}$ and $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}} + z\hat{\mathbf{z}}$, we get

$$\Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{r^5} [r^2(S_{1x}S_{2x} + S_{1y}S_{2y} + S_{1z}S_{2z}) - 3(S_{1x}x + S_{1y}y + S_{1z}z)(S_{2x}x + S_{2y}y + S_{2z}z)]$$

$$\begin{aligned} \Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{r^5} [& S_{1x}S_{2x}(r^2 - 3x^2) + S_{1y}S_{2y}(r^2 - 3y^2) + S_{1z}S_{2z}(r^2 - 3z^2) \\ & - 3xy(S_{1x}S_{2y} + S_{1y}S_{2x}) - 3xz(S_{1x}S_{2z} + S_{1z}S_{2x}) - 3yz(S_{1y}S_{2z} + S_{1z}S_{2y})] \end{aligned}$$

In terms of the *total* spin angular momentum $\mathbf{S} = \mathbf{S}_1 + \mathbf{S}_2$,

$$\begin{aligned} \Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{r^5} \frac{1}{2} [& S_x^2(r^2 - 3x^2) + S_y^2(r^2 - 3y^2) + S_z^2(r^2 - 3z^2) \\ & - 3xy(S_xS_y + S_yS_x) - 3xz(S_xS_z + S_zS_x) - 3yz(S_yS_z + S_zS_y)] \end{aligned}$$

$$\Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{r^5} \frac{1}{2} \begin{bmatrix} S_x & S_y & S_z \end{bmatrix} \begin{bmatrix} (r^2 - 3x^2)S_x - 3xyS_y - 3xzS_z \\ -3xyS_x + (r^2 - 3y^2)S_y - 3yzS_z \\ -3xzS_x - 3yzS_y + (r^2 - 3z^2)S_z \end{bmatrix}$$

$$\Rightarrow H_{12} = \frac{g_s^2 \mu_B^2}{2r^5} \begin{bmatrix} S_x & S_y & S_z \end{bmatrix} \begin{bmatrix} r^2 - 3x^2 & -3xy & -3xz \\ -3xy & r^2 - 3y^2 & -3yz \\ -3xz & -3yz & r^2 - 3z^2 \end{bmatrix} \begin{bmatrix} S_x \\ S_y \\ S_z \end{bmatrix}$$

Using expectation values (as x , y and z are random variables), and re-writing this as a matricial product,

$$\hat{H}_{12} = \frac{1}{\hbar} \hat{\mathbf{S}}^\dagger \mathbf{D} \hat{\mathbf{S}}.$$

Here, $\hat{\mathbf{S}}$ is the array containing the spin operators $\hat{S}_x$, $\hat{S}_y$ y $\hat{S}_z$ while $\mathbf{D}$ is the matrix formed by the average values of the projections (x , y y z) of the relative positions between the two magnetic dipoles. In the chosen coordinate system, this is the so-called zero-field splitting tensor.

Appendix B

Spin-spin correlation in Fourier space

In Chapter (3), one needs to calculate a factor in the form $\langle \delta s_\alpha(\mathbf{k}, t) \delta s_\beta(\mathbf{k}', 0) \rangle$ and express it in terms of the spin-spin correlation functions $C_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \langle \hat{s}_\alpha(\mathbf{r}, t) \hat{s}_\beta(\mathbf{r}', 0) \rangle$. For simplicity, we will use summation convention within this Appendix, only for latin letters (like n or m). First, we expand this product as Fourier transforms

$$\langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle = \int \int \langle \hat{s}_n(\mathbf{r}', t) \hat{s}_m(\mathbf{r}'', 0) \rangle \exp(-i\mathbf{k} \cdot \mathbf{r}') \exp(-i\mathbf{k}' \cdot \mathbf{r}'') d^2\mathbf{r}' d^2\mathbf{r}''$$

Inside the integral, this average should not depend on the separate positions $\mathbf{r}'$ and $\mathbf{r}''$, but the difference $\mathbf{r} - \mathbf{r}''$, which we will call $\mathbf{R}$. Therefore,

$$\langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle = \int \int \langle \hat{s}_n(\mathbf{r} - \mathbf{r}'', t) \hat{s}_m(0, 0) \rangle \exp(-i\mathbf{k} \cdot \mathbf{r}') \exp(-i\mathbf{k}' \cdot \mathbf{r}'') d^2\mathbf{r}' d^2\mathbf{r}''$$

and one can realize the substitution $\mathbf{r}' = \mathbf{R} + \mathbf{r}''$, obtaining

$$\begin{aligned} \langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle &= \int \int \langle \hat{s}_n(\mathbf{R}, t) \hat{s}_m(0, 0) \rangle \exp(-i\mathbf{k} \cdot \mathbf{R}) \exp[-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{r}''] d^2\mathbf{R} d^2\mathbf{r}'' \\ \Rightarrow \langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle &= \int \int \langle \hat{s}_n(\mathbf{R}, t) \hat{s}_m(0, 0) \rangle \exp(-i\mathbf{k} \cdot \mathbf{R}) \exp[-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{r}''] d^2\mathbf{r}'' d^2\mathbf{R} \\ \Rightarrow \langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle &= \int \langle \hat{s}_n(\mathbf{R}, t) \hat{s}_m(0, 0) \rangle \exp(-i\mathbf{k} \cdot \mathbf{R}) \int \exp[-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{r}''] d^2\mathbf{r}'' d^2\mathbf{R} \end{aligned}$$

The internal integral with variable $\mathbf{r}''$ corresponds to a 2D Dirac delta, while the other one with $\mathbf{R}$ is a Fourier transform. So,

$$\langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle = (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') \mathcal{F}_{\mathbf{R}} \{ \langle \hat{s}_n(\mathbf{R}, t) \hat{s}_m(\mathbf{0}, 0) \rangle \}(\mathbf{k}),$$

or, in terms of the 2D Fourier transform $\mathcal{C}_{nm}(\mathbf{k}, t)$ of the spin-spin correlation function,

$$\langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle = (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') \mathcal{C}_{nm}(\mathbf{k}, t). \quad (\text{B.1})$$

Now, let's suppose that we have to calculate terms of the form

$$f_{\ell p}(\mathbf{k}, \mathbf{k}') = D_{\ell n}(\mathbf{k}) D_{pm}(\mathbf{k}') \langle \hat{s}_n(\mathbf{k}, t) \hat{s}_m(\mathbf{k}', 0) \rangle, \quad (\text{B.2})$$

being $D_{\alpha\beta}(\mathbf{k})$ a complex matrix with $D_{\alpha\beta}(-\mathbf{k}) = D_{\alpha\beta}^*(\mathbf{k})$, which is exactly what we have in Chapter 3, specifically in equation (3.8). Using the result (B.1) in this definition,

$$\begin{aligned} f_{\ell p}(\mathbf{k}, \mathbf{k}') &= D_{\ell n}(\mathbf{k}) D_{pm}(-\mathbf{k}) (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') C_{nm}(\mathbf{k}, t), \\ \Rightarrow f_{\ell p}(\mathbf{k}, \mathbf{k}') &= D_{\ell n}(\mathbf{k}) D_{pm}^*(\mathbf{k}) (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') C_{nm}(\mathbf{k}, t) \end{aligned}$$

Let $\alpha, \beta = x, y, z$ two fixed coordinate indexes. If $m \neq n$, we have two terms (one with $n = \alpha$ and $m = \beta$, and the other with $n = \beta$ and $m = \alpha$):

$$f_{\ell p}(\mathbf{k}, \mathbf{k}') = (2\pi)^2 \delta^{(2)}(\mathbf{k} + \mathbf{k}') \{ D_{\ell\alpha}(\mathbf{k}) D_{p\beta}^*(\mathbf{k}) C_{\alpha\beta}(\mathbf{k}, t) + D_{\ell\beta}(\mathbf{k}) D_{p\alpha}^*(\mathbf{k}) C_{\beta\alpha}(\mathbf{k}, t) \} \quad (\text{B.3})$$

If both α and β are x or y (which is to say, the transverse coordinates), then $D_{\alpha\beta}^*(\mathbf{k}) = D_{\alpha\beta}(\mathbf{k})$ and the correlation $C_{\alpha\beta}(\mathbf{k}, \omega)$ (in Fourier space-time, with frequency ω) can be calculated using the fluctuation-dissipation theorem

$$C_{\alpha\beta}(\mathbf{k}, \omega) = \frac{2\hbar}{1 - \exp\left(-\frac{\hbar\omega}{k_B T}\right)} \chi''_{\alpha\beta}(\mathbf{k}, \omega), \approx \frac{2k_B T}{\omega} \chi''_{\alpha\beta}(\mathbf{k}, \omega)$$

where $\chi''_{\alpha\beta}$ is the imaginary part of the linear response function $\chi_{\alpha\beta}$, given in the real space by

$$\begin{aligned} \chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_\alpha(\mathbf{r}, t), \hat{s}_\beta(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_\alpha(\mathbf{r}, t), \hat{S}_\beta(\mathbf{r}', 0)] \rangle_{eq}. \end{aligned}$$

So, expanding $\hat{S}_x = (\hat{S}^+ + i\hat{S}^-)/2$ and $\hat{S}_y = (\hat{S}^+ - i\hat{S}^-)/(2i)$, we found

$$\begin{aligned} \chi_{xy}(\mathbf{r}, \mathbf{r}'; t) &= \frac{\sigma^2}{4} \theta(t) \langle [\hat{S}^+(\mathbf{r}, t) + i\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0) - i\hat{S}^-(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{xy}(\mathbf{r}, \mathbf{r}'; t) &= \frac{\sigma^2}{4} \theta(t) \langle [\hat{S}^+(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] - i[\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \\ &\quad + i[\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^-(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

Clearly, $[\hat{S}^+(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] = [\hat{S}^-(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] = 0$; they commute at $t = 0$, and they will do at every time. One can prove this by using Holstein-Primakoff transformations (4.4), (5.4) or (5.5) as appropriate. So,

$$\chi_{xy}(\mathbf{r}, \mathbf{r}'; t) = \frac{i\sigma^2}{4} \theta(t) \langle [\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] - [\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \rangle_{eq}$$

In a similar way, and using the same recent arguments,

$$\chi_{yx}(\mathbf{r}, \mathbf{r}'; t) = \frac{i\sigma^2}{4} \theta(t) \left\langle \left[\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0) \right] - \left[\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0) \right] \right\rangle_{eq}$$

Thus, we observe that $\chi_{xy} = -\chi_{yx}$, and consequently, $C_{xy}(\mathbf{k}, \omega) = -C_{yx}(\mathbf{k}, \omega)$ and $\mathcal{C}_{xy}(\mathbf{k}, t) = -\mathcal{C}_{yx}(\mathbf{k}, t)$, which in equation (B.3) results $f_{\ell p}(\mathbf{k}, \mathbf{k}') = 0$ when both α and β are x or y .

On the other hand, if one of the coordinates (namely α) is z and the other $\beta = x, y$, we have to calculate

$$\begin{aligned} \chi_{z\beta}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \left\langle \left[\hat{s}_z(\mathbf{r}, t), \hat{s}_\beta(\mathbf{r}', 0) \right] \right\rangle_{eq} \\ \Rightarrow \chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \left\langle \left[\hat{S}_z(\mathbf{r}, t), \hat{S}_\beta(\mathbf{r}', 0) \right] \right\rangle_{eq}. \end{aligned}$$

Again, expanding $\hat{S}_x = (\hat{S}^+ + i\hat{S}^-)/2$ or $\hat{S}_y = (\hat{S}^+ - i\hat{S}^-)/(2i)$,

$$\chi_{z\beta}(\mathbf{r}, \mathbf{r}'; t) = \frac{\sigma^2}{4} \theta(t) \left\langle \left[\hat{S}_z(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0) \pm \hat{S}^-(\mathbf{r}', 0) \right] \right\rangle_{eq}$$

and by Holstein-Primakoff transformations (in any of their forms), it is clear that $\chi_{z\beta}(\mathbf{r}, \mathbf{r}'; t) = 0$. In general, correlators with an odd number of annihilation operators always result 0.

Finally, we conclude that identity (B.1) holds, and only diagonal terms with $n = m$ contribute to the sum in (B.2).

Appendix C

Fluctuation-dissipation theorem

In this work, we define a fluctuation $\delta\hat{A}(t)$ as every slight, time-dependent deviation of a quantity A respect to its equilibrium, time-independent canonical average $\langle\hat{A}\rangle_{eq}$, which is to say, $\delta\hat{A}(t) = \hat{A}(t) - \langle\hat{A}\rangle_{eq}$. Correlation functions in the form $F(t, \tau) = \langle\delta\hat{A}(t)\delta\hat{A}(t+\tau)\rangle$ are also constantly used here, where the variable t might represent time, position vectors, or even space-time events x , just as in the case of the so-mentioned spin-spin correlation function $C_{zz}(x, x') = \langle\delta\hat{s}_z(x)\delta\hat{s}_z(x')\rangle$ of the main text. Some of the most important properties of those correlation functions are [27]:

1. In steady state, systems acquire time translation symmetry, which for correlators implies that

$$F(t, \tau) = \langle\delta\hat{A}(t)\delta\hat{A}(t+\tau)\rangle = F(\tau) \text{ for each } t.$$

2. In steady state, $F(0) = \langle\delta\hat{A}(t)^2\rangle > 0$.

3. The correlation F is an even function:

$$\begin{aligned} F(\tau) &= \langle\delta\hat{A}(t)\delta\hat{A}(t+\tau)\rangle \\ \Rightarrow F(\tau) &= \langle\delta\hat{A}(t-\tau)\delta\hat{A}(t-\tau+\tau)\rangle \\ \Rightarrow F(\tau) &= F(-\tau). \end{aligned}$$

4. For each t , we have $|F(t)| \leq F(0)$. Proof: consider $\langle|\delta\hat{A}(t_1) \pm \delta\hat{A}(t_2)|^2\rangle = \langle\delta\hat{A}^2(t_1)\rangle + \langle\delta\hat{A}^2(t_2)\rangle \pm 2\langle\delta\hat{A}(t_1)\delta\hat{A}(t_2)\rangle$, so

$$\langle|\delta\hat{A}(t_1) \pm \delta\hat{A}(t_2)|\rangle = 2[F(0) \pm F(t)],$$

but $\langle|\delta\hat{A}(t_1) \pm \delta\hat{A}(t_2)|^2\rangle \geq 0$, so

$$|F(t)| \leq F(0).$$

5. Fluctuations in general have a *characteristic time* τ^* in which they occur. For $\tau \gg \tau^*$, fluctuations $\delta\hat{A}(t)$ and $\delta\hat{A}(t + \tau)$ become statistically independent, which is to say,

$$\langle \delta\hat{A}(t) \delta\hat{A}(t + \tau) \rangle \approx \langle \delta\hat{A}(t) \rangle \langle \delta\hat{A}(t + \tau) \rangle = 0, \text{ for } \tau \gg \tau^*.$$

In general, fluctuations produce time-dependence in system's hamiltonian, which is not studied in stationary ensemble theory (like canonical ensemble with partition function). That's why using the partition function to calculate averages $\langle \dots \rangle$, including correlations, won't work. Our primary aim in this Appendix is to state a general way to calculate out-of-equilibrium (*perturbed*) expectation values $\langle \dots \rangle$, like $\langle \hat{A}(t) \rangle$. The fundamental theory that enables us to study and calculate these quantities is the *linear response theory*, which defines what is called the *linear response function*. Let two observables, with their respective self-adjoint operators $\hat{A}$ and $\hat{B}$ in Schrödinger picture. This last observable operator $\hat{B}$ is what gives time-dependence to the hamiltonian $\hat{H}$, modulated by an external agent $f(t)$ adiabatically switched on at $t \rightarrow -\infty$, such that the hamiltonian takes the form

$$\hat{H}(t) = \hat{H}^{(0)} - f(t) \hat{B},$$

where $\hat{H}^{(0)}$ is the time-independent part of the hamiltonian, without considering fluctuations. Therefore, the linear response function χ_{AB} is defined in a way such that¹

$$\langle \hat{A}(t) \rangle = \langle \hat{A} \rangle_{eq} + \int_{-\infty}^{\infty} \chi_{AB}(t - t') f(t') dt'. \quad (\text{C.1})$$

This response function plays a similar role to Green's function in inhomogeneous linear differential equations: it allows us to determine how $\hat{A}$ evolves in presence of an external agent f . In this case, $f(t)$ connects $\hat{A}$ and $\hat{B}$. What we pretend here is to determine this linear response function χ_{AB} .

To begin with, we expand the canonical average

$$\langle \delta\hat{A}(t) \rangle = \langle \hat{A}(t) \rangle - \langle \hat{A} \rangle_{eq} = \text{Tr } \hat{A} \hat{\rho}(t) - \text{Tr } \hat{A} \hat{\rho}_{eq} = \text{Tr } \hat{A} \delta\hat{\rho}(t), \quad (\text{C.2})$$

where $\hat{\rho}(t) = \rho_{eq} + \delta\hat{\rho}(t)$ is the perturbed density operator and $\delta\hat{\rho}(t)$ its fluctuation with respect to the equilibrium operator ρ_{eq} , caused by the coupling term $-f(t) \hat{B}$. In order to determine $\delta\hat{\rho}(t)$, we utilize

¹One may write the first term either $\langle \hat{A} \rangle_{eq}$ or $\langle \hat{A}(t) \rangle_{eq}$, explicitly evaluated at time t . As $\hat{A}(t) = \exp\left(\frac{iH^{(0)}t}{\hbar}\right) \hat{A} \exp\left(-\frac{iH^{(0)}t}{\hbar}\right)$ and $[\hat{H}^{(0)}, \hat{\rho}_{eq}] = 0$, we have $\text{Tr } \hat{A}(t) \hat{\rho}_{eq} = \text{Tr } \hat{A} \hat{\rho}_{eq}$ and it is enough to use operators in Schrödinger picture in $\langle \dots \rangle_{eq}$. By the way, if explicit time dependence is written along the operators $\hat{A}(t)$ or $\hat{B}(t)$, we assume they are in the interaction picture.

Liouville-von Neumann equation:

$$\begin{aligned} i\hbar \frac{\partial \hat{\rho}}{\partial t} &= [\hat{H}(t), \hat{\rho}(t)] \\ \Rightarrow \frac{\partial}{\partial t} [\hat{\rho}_{eq} + \delta \hat{\rho}(t)] &= \frac{1}{i\hbar} \left([\hat{H}^{(0)}, \hat{\rho}_{eq}] - f(t) [\hat{B}, \hat{\rho}_{eq}] + [\hat{H}^{(0)}, \delta \hat{\rho}(t)] - f(t) [\hat{B}, \delta \hat{\rho}(t)] \right) \\ \Rightarrow \frac{\partial}{\partial t} [\delta \hat{\rho}(t)] &\approx \frac{1}{i\hbar} \left([\hat{H}^{(0)}, \delta \hat{\rho}(t)] - f(t) [\hat{B}, \hat{\rho}_{eq}] \right), \end{aligned}$$

where we used $i\hbar \partial \hat{\rho}_{eq} / \partial t = 0$ (and consequently, $[\hat{H}^{(0)}, \hat{\rho}_{eq}] = 0$) and the term $f(t) [\hat{B}, \delta \hat{\rho}]$ was neglected at first order approximation. This results in a linear differential equation for $\delta \hat{\rho}(t)$, which can be solved by multiplying the integrating factors $\exp(i\hat{H}_0 t / \hbar)$ to right, and $\exp(-i\hat{H}_0 t / \hbar)$ to left, obtaining

$$\delta \hat{\rho}(t) = \frac{i}{\hbar} \int_{-\infty}^t f(t') \exp\left(-\frac{i\hat{H}^{(0)}(t-t')}{\hbar}\right) [\hat{B}, \hat{\rho}_{eq}] \exp\left(\frac{i\hat{H}^{(0)}(t-t')}{\hbar}\right) dt'.$$

Next, using (C.2),

$$\langle \delta \hat{A}(t) \rangle = \frac{i}{\hbar} \int_{-\infty}^t \langle [\hat{A}(t), \hat{B}(t')] \rangle_{eq} f(t') dt',$$

being $\hat{A}(t)$ and $\hat{B}(t)$ the operators in the interaction picture. Comparing this result with (C.1) we read the linear response function to be

$$\chi_{AB}(t, t') = \frac{i}{\hbar} \theta(t - t') \langle [\hat{A}(t), \hat{B}(t')] \rangle_{eq},$$

where θ is the Heaviside theta function.

Usually (as just in the case of this work), one needs the Fourier transform of the linear response function, given by

$$\mathcal{F}_t \left\{ \langle \delta \hat{A}(t) \rangle \right\}(\omega) = \mathcal{F}_t \{ \chi_{AB}(t) \}(\omega) \mathcal{F}_t \{ f(t) \}(\omega) = [\chi'(\omega) + i\chi''(\omega)] \mathcal{F}_t \{ f(t) \}(\omega),$$

where χ' y χ'' are the real and imaginary part of $\mathcal{F}_t \{ \chi_{AB}(t) \}$, respectively. Physically, the imaginary part indicates how much energy is being dissipating along the fluctuation process triggered by the external agent $f(t)$. Using the identity $\chi'' = (\mathcal{F}_t \{ \chi_{AB}(t) \} - \mathcal{F}_t \{ \chi_{AB}(t) \}^*) / (2i)$, we obtain

$$\chi''_{AB}(\omega) = \frac{1}{2\hbar} \int_{-\infty}^{\infty} \langle [\hat{A}(t), \hat{B}(0)] \rangle_{eq} \exp(i\omega t) dt. \quad (C.3)$$

The last remaining question is: how this linear response function is related to the correlation function. One can answer this question by expanding the commutator $\langle [\hat{A}(t), \hat{B}(0)] \rangle_{eq}$ using Boltzmann distribution:

$$\langle \hat{A}(t) \hat{B}(t') \rangle_{eq} = \frac{\text{Tr} [\hat{A}(t) \hat{B}(t') \exp(-\beta \hat{H}^{(0)})]}{Z} = \frac{\text{Tr} [\hat{A}(t) \exp(-\beta \hat{H}_0) \exp(\beta \hat{H}_{(0)}) \hat{B}(t') \exp(-\beta \hat{H}_{(0)})]}{Z},$$

where Z is the canonical partition function and $\beta = 1/(k_B T)$, with $T > 0$ the temperature and k_B the Boltzmann constant. Now, we utilize the time evolution relation in Heisenberg picture²

$$\exp\left(i\hat{H}^{(0)}\tau/\hbar\right)\hat{B}(t')\exp\left(-i\hat{H}^{(0)}\tau/\hbar\right) = \hat{B}(t' + \tau) \quad (\text{C.4})$$

with $\tau = -i\hbar\beta$, obtaining

$$\begin{aligned} \left\langle \hat{A}(t) \hat{B}(t') \right\rangle_{eq} &= \frac{\text{Tr}(\hat{B}(t' - i\hbar\beta) \hat{A}(t) \exp(-\beta H))}{Z} \\ \Rightarrow \left\langle \hat{A}(t) \hat{B}(t') \right\rangle_{eq} &= \left\langle \hat{B}(t') \hat{A}(t + i\hbar\beta) \right\rangle_{eq} \end{aligned}$$

Finally, inserting this result into (C.3),

$$\begin{aligned} \chi''_{AB}(\omega) &= \frac{1}{2\hbar} (1 - \exp(-\beta\hbar\omega)) \int_{-\infty}^{\infty} \left\langle \hat{A}(t) \hat{B}(0) \right\rangle_{eq} \exp(i\omega t) dt, \\ \Rightarrow \chi''_{AB}(\omega) &= \frac{1}{2\hbar} \left[1 - \exp\left(-\frac{\hbar\omega}{k_B T}\right) \right] S_{AB}(\omega) \end{aligned}$$

with $S_{AB}(\omega) = \int_{-\infty}^{\infty} \left\langle \hat{A}(t) \hat{B}(0) \right\rangle \exp(i\omega t) dt$. This is the so-called fluctuation-dissipation theorem.

²Actually, Heisenberg picture's time evolution is described by $\exp\left(i\hat{H}\tau/\hbar\right)\hat{B}(t')\exp\left(-i\hat{H}\tau/\hbar\right) = \hat{B}(t' + \tau)$, using the full hamiltonian $\hat{H} = \hat{H}^{(0)} + \hat{H}'$. If $[\hat{H}^{(0)}, \hat{H}'] = 0$, we may separate $\exp\left(i\hat{H}\tau/\hbar\right) = \exp\left(i\hat{H}^{(0)}\tau/\hbar\right)\exp\left(i\hat{H}'\tau/\hbar\right)$ and cancel the factors with $\hat{H}'$, because $\hat{H}'(t)$ and $\hat{B}(t)$ commute at any given instant t by definition. Time evolution (C.4) is valid up to first order in perturbations.

Appendix D

Spin-spin correlations in antiferromagnets

According to dissipation-fluctuation theorem, it is possible to relate the correlation function $C_{\alpha\beta}$ with the susceptibility $\chi_{\alpha\beta}(\mathbf{k}, \omega)$, as

$$C_{\alpha\beta}(\mathbf{k}, \omega) = \frac{2\hbar}{1 - \exp(-\beta\hbar\omega)} \chi''_{\alpha\beta}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi''_{\alpha\beta}(\mathbf{k}, \omega), \quad (\text{D.1})$$

where $\chi''(\mathbf{k}, \omega)$ is the imaginary part of the Fourier transform of the response function. The most straightforward method to calculate this type of susceptibilities is using the dissipation-fluctuation formula in terms of the commutator

$$\chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; t) = \frac{i}{\hbar} \theta(t) \langle [\hat{s}_\alpha(\mathbf{r}', t), \hat{s}_\beta(\mathbf{r}, 0)] \rangle_{eq}. \quad (\text{D.2})$$

To keep going, it is necessary to specify which coordinates α, β are we using. Each case will give us a different susceptibility.

D.1 Transverse spin-spin correlator

In this case, the desired spin-spin correlator is

$$C_{xx}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi''_{xx}(\mathbf{k}, \omega) \quad (\text{D.3})$$

where the linear response function in real space is given by

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_x(\mathbf{r}, t), \hat{s}_x(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_x(\mathbf{r}, t), \hat{S}_x(\mathbf{r}', 0)] \rangle_{eq}. \end{aligned} \quad (\text{D.4})$$

Here, $\hat{S}_x = (\hat{S}^+ + \hat{S}^-)/2$ and $\hat{S}_y = (\hat{S}^+ - \hat{S}^-)/(2i)$, so we may write the linear response function as

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\sigma^2}{4\hbar} \theta(t) \langle [\hat{S}^+(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^+(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \\ & + [\hat{S}^-(\mathbf{r}, t), \hat{S}^+(\mathbf{r}', 0)] + [\hat{S}^-(\mathbf{r}, t), \hat{S}^-(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

Note that the HP transformation for the spin operators $\hat{S}^\pm(\mathbf{r}, t)$ depends on the position $\mathbf{r}$; if $\mathbf{r}$ lies on the sublattice L_1 , we must expand $\hat{S}^+(\mathbf{r}, t)$ as $\sqrt{2s}\hat{a}(\mathbf{r}, t)$, while if $\mathbf{r} \in L_2$, we use $\hat{S}^+(\mathbf{r}, t) = \sqrt{2s}\hat{b}^\dagger(\mathbf{r}, t)$. Since $C(\mathbf{k}, \omega)$ is the Fourier transform of $C(\mathbf{r}, t)$ (and therefore, an integral over all the positions in the sample is performed), it is necessary to step on every case and sum them all.

D.1.1 First case: same sublattice (1)

In this situation, using the HP transformations for the sublattice L_1 , we may write the susceptibility as

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2} \theta(t) \langle [\hat{a}(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \\ & + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}(\mathbf{r}', 0)] + [\hat{a}^\dagger(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

Now, expanding every raising and lowering operator in the Fourier space, we have

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2N} \theta(t) \sum_{\mathbf{k}'} \sum_{\mathbf{k}''} \langle [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{a}_{\mathbf{k}'}^\dagger(t), \hat{a}_{\mathbf{k}''}^\dagger(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \rangle_{eq} \end{aligned}$$

and taking the Fourier transform for the relative position vector $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ (or equivalently¹, for $\mathbf{r}$ evaluating $\mathbf{r}' = \mathbf{0}$), in the convenient limit $\int d^2\mathbf{R} \rightarrow \frac{1}{\sigma} \sum_{\mathbf{R}}$, we obtain

$$\chi_{xx}(\mathbf{k}, t) = \frac{i\hbar\sigma^2}{2} \theta(t) \sum_{\mathbf{k}'} \left\langle [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{\mathbf{k}}(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}(0)] + [\hat{a}_{-\mathbf{k}}^\dagger(t), \hat{a}_{\mathbf{k}'}^\dagger(0)] \right\rangle_{eq}$$

The first and the last commutators vanish, while the two remaining must be expanded using the Bogoliubov transformations²

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2} \theta(t) \sum_{\mathbf{k}'} \langle u_{\alpha\mathbf{k}} u_{\alpha\mathbf{k}'} [\hat{\alpha}_{\mathbf{k}}(t), \hat{\alpha}_{\mathbf{k}'}^\dagger(0)] - v_{\alpha\mathbf{k}} v_{\alpha\mathbf{k}'} [\hat{\beta}_{-\mathbf{k}'}(0), \hat{\beta}_{-\mathbf{k}}^\dagger(t)] \\ & - u_{\alpha\mathbf{k}} u_{\alpha\mathbf{k}'} [\hat{\alpha}_{\mathbf{k}'}(0), \hat{\alpha}_{-\mathbf{k}}^\dagger(t)] + v_{\alpha\mathbf{k}} v_{\alpha\mathbf{k}'} [\hat{\beta}_{\mathbf{k}}(t), \hat{\beta}_{-\mathbf{k}'}^\dagger(0)] \rangle_{eq} \end{aligned}$$

¹In the long run, the result will be the same as using $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, since the system has translational symmetry.

²One can check this by expanding $\hat{a}_{\mathbf{k}}$ and $\hat{b}_{\mathbf{k}}$ with the Bogoliubov transformation, in a similar fashion to the method shown here, as the Bogoliubov coefficients are defined such that $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}}] = [\hat{\alpha}_{\mathbf{k}}^\dagger, \hat{\alpha}_{\mathbf{k}}^\dagger] = [\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}}] = [\hat{\beta}_{\mathbf{k}}^\dagger, \hat{\beta}_{\mathbf{k}}^\dagger] = 0$, and the mixed commutators are zero as well.

Now, using the characteristic raising and lowering operators evolution in the Heisenberg picture $\hat{\alpha}_{\mathbf{k}}(t) = \hat{\alpha}_{\mathbf{k}}(0) \exp(-i\omega_{\alpha\mathbf{k}}t - \eta t)$ and $\hat{\beta}_{\mathbf{k}}(t) = \hat{\beta}_{\mathbf{k}}(0) \exp(-i\omega_{\beta\mathbf{k}}t - \eta t)$, with $\eta > 0$, and remembering the commutation relations $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}'}^\dagger] = [\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$, we have

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2} \theta(t) [u_{\alpha\mathbf{k}}^2 \exp(-i\omega_{\alpha\mathbf{k}}t) - v_{\alpha\mathbf{k}}^2 \exp(i\omega_{\beta\mathbf{k}}t) \\ & - u_{\alpha\mathbf{k}}^2 \exp(i\omega_{\alpha\mathbf{k}}t) + v_{\alpha\mathbf{k}}^2 \exp(-i\omega_{\beta\mathbf{k}}t)] \exp(-\eta t) \end{aligned}$$

Finally, taking the remaining temporal Fourier transform, considering $\omega_{\alpha\mathbf{k}} > 0$ and $\omega_{\beta\mathbf{k}} > 0$, we finally obtain

$$\chi_{xx}''(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[u_{\alpha\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + v_{\alpha\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right] \quad (\text{D.5})$$

D.1.2 Second case: same sublattice (2)

This case is quite similar to the last one. Here, we need to use the HP transformations for the sublattice L_2 , writing the susceptibility as

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2} \theta(t) \langle [\hat{b}^\dagger(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0)] + [\hat{b}^\dagger(\mathbf{r}, t), \hat{b}(\mathbf{r}', 0)] \\ & + [\hat{b}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0)] + [\hat{b}(\mathbf{r}, t), \hat{b}(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

As before, we expand every raising and lowering operator in the Fourier space, we have

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2N} \theta(t) \sum_{\mathbf{k}'} \sum_{\mathbf{k}''} \langle [\hat{b}_{\mathbf{k}'}^\dagger(t), \hat{b}_{\mathbf{k}''}^\dagger(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{b}_{\mathbf{k}'}^\dagger(t), \hat{b}_{\mathbf{k}''}(0)] \exp i(-\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{b}_{\mathbf{k}'}(t), \hat{b}_{\mathbf{k}''}^\dagger(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} - \mathbf{k}'' \cdot \mathbf{r}') \\ & + [\hat{b}_{\mathbf{k}'}(t), \hat{b}_{\mathbf{k}''}(0)] \exp i(\mathbf{k}' \cdot \mathbf{r} + \mathbf{k}'' \cdot \mathbf{r}') \rangle_{eq} \end{aligned}$$

and taking the Fourier transform for the position for $\mathbf{r}$ evaluating $\mathbf{r}' = \mathbf{0}$, in the convenient limit $\int d^2\mathbf{R} \rightarrow \frac{1}{\sigma} \sum_{\mathbf{R}}$, we obtain

$$\chi_{xx}(\mathbf{k}, t) = \frac{i\hbar\sigma^2}{2} \theta(t) \sum_{\mathbf{k}'} \left\langle [\hat{b}_{-\mathbf{k}}^\dagger(t), \hat{b}_{\mathbf{k}'}^\dagger(0)] + [\hat{b}_{-\mathbf{k}}^\dagger(t), \hat{b}_{\mathbf{k}'}(0)] + [\hat{b}_{\mathbf{k}}(t), \hat{b}_{\mathbf{k}'}^\dagger(0)] + [\hat{b}_{\mathbf{k}}(t), \hat{b}_{\mathbf{k}'}(0)] \right\rangle_{eq}$$

The first and the last commutators vanish, while the two remaining must be expanded using the Bogoliubov transformations:

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2} \theta(t) \sum_{\mathbf{k}'} \langle u_{\beta\mathbf{k}} u_{\beta\mathbf{k}'} [\hat{\alpha}_{\mathbf{k}}(t), \hat{\alpha}_{\mathbf{k}'}^{-\dagger}(0)] - v_{\beta\mathbf{k}} v_{\beta\mathbf{k}'} [\hat{\beta}_{\mathbf{k}'}(0), \hat{\beta}_{-\mathbf{k}}^\dagger(t)] \\ & - u_{\beta\mathbf{k}} u_{\beta\mathbf{k}'} [\hat{\alpha}_{-\mathbf{k}'}(0), \hat{\alpha}_{-\mathbf{k}}^\dagger(t)] + v_{\beta\mathbf{k}} v_{\beta\mathbf{k}'} [\hat{\beta}_{-\mathbf{k}}(t), \hat{\beta}_{-\mathbf{k}'}^\dagger(0)] \rangle_{eq} \end{aligned}$$

Now, using the characteristic raising and lowering operators evolution in the Heisenberg picture $\hat{\alpha}_{\mathbf{k}}(t) = \hat{\alpha}_{\mathbf{k}}(0) \exp(-i\omega_{\alpha\mathbf{k}}t - \eta t)$ and $\hat{\beta}_{\mathbf{k}}(t) = \hat{\beta}_{\mathbf{k}}(0) \exp(-i\omega_{\beta\mathbf{k}}t - \eta t)$, with $\eta > 0$, and remembering the commutation relations $[\hat{\alpha}_{\mathbf{k}}, \hat{\alpha}_{\mathbf{k}'}^\dagger] = [\hat{\beta}_{\mathbf{k}}, \hat{\beta}_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'}$, we have

$$\begin{aligned} \chi_{xx}(\mathbf{k}, t) = & \frac{i\hbar\sigma^2}{2} \theta(t) [u_{\beta\mathbf{k}}^2 \exp(-i\omega_{\alpha\mathbf{k}}t) - v_{\beta\mathbf{k}}^2 \exp(i\omega_{\beta\mathbf{k}}t) \\ & - u_{\beta\mathbf{k}}^2 \exp(i\omega_{\alpha\mathbf{k}}t) + v_{\beta\mathbf{k}}^2 \exp(-i\omega_{\beta\mathbf{k}}t)] \exp(-\eta t) \end{aligned}$$

Finally, taking the remaining temporal Fourier transform, considering $\omega_{\alpha\mathbf{k}} > 0$ and $\omega_{\beta\mathbf{k}} > 0$, we finally obtain

$$\chi_{xx}''(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[u_{\beta\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + v_{\beta\mathbf{k}}^2 \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right] \quad (\text{D.6})$$

D.1.3 Third case: mixed sublattices

It is easy to see that both mixed cases give the same contribution to the total susceptibility, so we'll consider only $\mathbf{r} \in L_1$ and $\mathbf{r}' \in L_2$ and count it twice. Here, this linear response function is expanded by

$$\begin{aligned} \chi_{xx}(\mathbf{r}, \mathbf{r}'; t) = & \frac{i\hbar\sigma^2}{2} \theta(t) \langle [\hat{a}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0)] + [\hat{a}(\mathbf{r}, t), \hat{b}(\mathbf{r}', 0)] \\ & + [\hat{a}^\dagger(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0)] + [\hat{a}^\dagger(\mathbf{r}, t), \hat{b}(\mathbf{r}', 0)] \rangle_{eq} \end{aligned}$$

and doing the same procedure as before, we find

$$\chi_{xx}''(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[u_{\alpha\mathbf{k}} u_{\beta\mathbf{k}} \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + v_{\alpha\mathbf{k}} v_{\beta\mathbf{k}} \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right] \quad (\text{D.7})$$

D.1.4 Final transverse spin-spin correlator

Adding up the results (D.7) (twice), (D.5) and (D.6), we obtain

$$\chi_{xx}''(\mathbf{k}, \omega) = \frac{s\hbar\sigma^2}{2} \left[(u_{\alpha\mathbf{k}} - v_{\alpha\mathbf{k}})^2 \frac{\eta}{(\omega - \omega_{\alpha\mathbf{k}})^2 + \eta^2} + (u_{\beta\mathbf{k}} - v_{\beta\mathbf{k}})^2 \frac{\eta}{(\omega - \omega_{\beta\mathbf{k}})^2 + \eta^2} \right],$$

so, according to dissipation-fluctuation theorem (D.1),

$$C_{xx}(\mathbf{k}, \omega) = \frac{\hbar\sigma s k_B T}{\omega} [(u_{\alpha\mathbf{k}} - v_{\alpha\mathbf{k}})^2 \frac{\eta}{\eta^2 + (\omega_{\alpha\mathbf{k}} - \omega)^2} + (u_{\beta\mathbf{k}} - v_{\beta\mathbf{k}})^2 \frac{\eta}{\eta^2 + (\omega_{\beta\mathbf{k}} - \omega)^2}]. \quad (\text{D.8})$$

D.2 Longitudinal spin-spin correlator

In this situation, the desired spin-spin correlator is

$$C_{zz}(\mathbf{k}, \omega) \approx \frac{2k_B T}{\omega} \chi_{zz}''(\mathbf{k}, \omega)$$

where the response linear function is given by

$$\begin{aligned}\chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i}{\hbar} \theta(t) \langle [\hat{s}_z(\mathbf{r}, t), \hat{s}_z(\mathbf{r}', 0)] \rangle_{eq} \\ \Rightarrow \chi_{zz}(\mathbf{r}, \mathbf{r}'; t) &= \frac{i\sigma^2}{\hbar} \theta(t) \langle [\hat{S}_z(\mathbf{r}, t), \hat{S}_z(\mathbf{r}', 0)] \rangle_{eq}.\end{aligned}$$

As in the transverse correlator computation, it is necessary to split the procedure in several cases for the positions $\mathbf{r}$ and $\mathbf{r}'$, and finally sum them all.

D.2.1 First case: same sublattice (1)

First, if $\mathbf{r}, \mathbf{r}' \in L_1$, the spin operators $\hat{S}_z$ can be expanded using HP transformations, so

$$\chi_{zz}^{(11)}(\mathbf{r}, \mathbf{r}'; t) = i\hbar\sigma^2\theta(t) \langle [\hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}, t), \hat{a}^\dagger(\mathbf{r}', 0) \hat{a}(\mathbf{r}', 0)] \rangle_{eq}.$$

This is a many-particle correlator. We can split it up in four terms using the product rule. Next, expanding the operators $\hat{a}$ y $\hat{a}^\dagger$ in the Fourier space, and using the Bogoliubov transformations, we obtain the imaginary part of the response function in $\mathbf{k}$ space:

$$\begin{aligned}\chi_{zz}^{(11)}(\mathbf{k}, \omega) &= \frac{\hbar\sigma}{N} \sum_{\mathbf{k}'} \{ \\ &|u_{\alpha\mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\alpha\mathbf{k}'} - \omega_{\alpha(\mathbf{k}'-\mathbf{k})} + \omega)^2} \\ &- |u_{\alpha\mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(-\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\alpha\mathbf{k}'} + \omega_{\beta(\mathbf{k}'-\mathbf{k})} + \omega)^2} \\ &- |v_{\beta\mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\alpha(\mathbf{k}'-\mathbf{k})} + \omega_{\beta\mathbf{k}'} - \omega)^2} \\ &+ |v_{\beta\mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(-\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\beta(\mathbf{k}'-\mathbf{k})} - \omega_{\beta\mathbf{k}'} + \omega)^2} \\ &+ |v_{\beta\mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\beta\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega)^2} \\ &- |v_{\beta\mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(-\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\beta\mathbf{k}'} - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega)^2} \\ &- |u_{\alpha\mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\alpha\mathbf{k}'} + \omega)^2} \\ &+ |u_{\alpha\mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(-\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\alpha\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega)^2} \} \end{aligned}$$

where we used $\langle \hat{\alpha}_{\mathbf{k}}^\dagger \hat{\alpha}_{\mathbf{k}} \rangle_{eq} = n_B(\hbar\omega_{\alpha\mathbf{k}})$ and $\langle \hat{\beta}_{\mathbf{k}}^\dagger \hat{\beta}_{\mathbf{k}} \rangle_{eq} = n_B(\hbar\omega_{\beta\mathbf{k}})$, with $n_B(\varepsilon) = \left[\exp\left(\frac{\varepsilon}{k_B T}\right) - 1 \right]^{-1}$ the Bose-Einstein distribution.

D.2.2 Second case: mixed sublattices

On the other hand, if $\mathbf{r} \in L_1$ and $\mathbf{r}' \in L_2$, it is necessary to use the HP transformations (5.4) y (5.5) in a mixed way. The needed commutator is

$$\chi_{zz}^{(12)}(\mathbf{r}, \mathbf{r}'; t) = -i\hbar\sigma^2\theta(t) \left\langle \left[\hat{a}^\dagger(\mathbf{r}, t) \hat{a}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0) \hat{b}(\mathbf{r}', 0) \right] \right\rangle$$

The procedure to calculate $\chi_{zz}^{(12)}$ in Fourier space is analogous to $\chi_{zz}^{(11)}$, obtaining

$$\begin{aligned} \chi_{zz}^{(12)}(\mathbf{k}, \omega) = \frac{\hbar\sigma}{N} \sum_{\mathbf{k}'} \{ & \frac{2\eta n_B (-\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\alpha(\mathbf{k}'-\mathbf{k})} + \omega_{\beta\mathbf{k}'})^2} \\ & - u_{\beta\mathbf{k}'} u_{\beta(\mathbf{k}'-\mathbf{k})} v_{\alpha(\mathbf{k}'-\mathbf{k})} v_{\beta\mathbf{k}'} \frac{2\eta n_B (\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\beta\mathbf{k}'} - \omega_{\beta(\mathbf{k}'-\mathbf{k})})^2} \\ & - u_{\alpha\mathbf{k}'} u_{\alpha(\mathbf{k}'-\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\alpha(\mathbf{k}'-\mathbf{k})} \frac{2\eta n_B (-\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\alpha(\mathbf{k}'-\mathbf{k})} - \omega_{\alpha\mathbf{k}'})^2} \\ & + u_{\alpha\mathbf{k}'} u_{\beta(\mathbf{k}'-\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\beta(\mathbf{k}'-\mathbf{k})} \frac{2\eta n_B (\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega - \omega_{\alpha\mathbf{k}'} - \omega_{\beta(\mathbf{k}'-\mathbf{k})})^2} \\ & + u_{\alpha\mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} \frac{2\eta n_B (-\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\alpha\mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})})^2} \\ & - u_{\alpha\mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} \frac{2\eta n_B (\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\alpha\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})})^2} \\ & - u_{\beta\mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\beta\mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} \frac{2\eta n_B (-\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\beta\mathbf{k}'})^2} \\ & + u_{\beta\mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\beta\mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} \frac{2\eta n_B (\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega + \omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta\mathbf{k}'})^2} \} \end{aligned}$$

In a similar way, if $\mathbf{r}, \mathbf{r}' \in L_2$, the needed HP transformations are entirely (5.5), because the many-particle commutator now is

$$\chi_{zz}^{(22)}(\mathbf{r}, \mathbf{r}'; t) = i\hbar\sigma^2\theta(t) \left\langle \left[\hat{b}^\dagger(\mathbf{r}, t) \hat{b}(\mathbf{r}, t), \hat{b}^\dagger(\mathbf{r}', 0) \hat{b}(\mathbf{r}', 0) \right] \right\rangle$$

In the Fourier space, what we get is

$$\begin{aligned} \chi_{zz}''^{(22)}(\mathbf{k}, \omega) = \frac{\hbar\sigma}{N} \sum_{\mathbf{k}'} \{ & |u_{\beta\mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(\alpha\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\beta\mathbf{k}'} - \omega_{\beta(\mathbf{k}'-\mathbf{k})} + \omega)^2} \\ & - |u_{\beta\mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(-\alpha\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\beta\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'-\mathbf{k})} + \omega)^2} \\ & - |v_{\alpha\mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(\alpha\hbar\omega_{\beta(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\beta(\mathbf{k}'-\mathbf{k})} + \omega_{\alpha\mathbf{k}'} - \omega)^2} \\ & + |v_{\alpha\mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 \frac{2\eta n_B(-\alpha\hbar\omega_{\alpha(\mathbf{k}'-\mathbf{k})})}{4\eta^2 + (\omega_{\alpha(\mathbf{k}'-\mathbf{k})} - \omega_{\alpha\mathbf{k}'} + \omega)^2} \\ & + |v_{\alpha\mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(\alpha\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\alpha\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega)^2} \\ & - |v_{\alpha\mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(-\alpha\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\alpha\mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega)^2} \\ & - |u_{\beta\mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(\alpha\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta\mathbf{k}'} + \omega)^2} \\ & + |u_{\beta\mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \frac{2\eta n_B(-\alpha\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})}{4\eta^2 + (\omega_{\beta\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega)^2} \} \end{aligned}$$

The final susceptibility in Fourier space is the sum of all these contributions:

$$\chi_{zz}'' = \chi_{zz}''^{(11)} + \chi_{zz}''^{(12)} + \chi_{zz}''^{(21)} + \chi_{zz}''^{(22)}.$$

Now, we take the limit $\eta \rightarrow 0^+$ in equation (D.2.2), in order to make it easier to reduce. This will transform the lorentzian into Dirac delta distributions, by the identity

$$\lim_{\eta \rightarrow 0^+} \frac{1}{\pi} \frac{\eta}{\eta^2 + (\omega - \omega_0)^2} = \delta(\omega - \omega_0).$$

Note that, as $\omega > 0$, as well as $\omega_{\alpha\mathbf{k}}$, factors like $\delta(\omega + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega_{\beta\mathbf{k}'})$ are always zero and terms that

contain them vanish. This simplifies enormously the final expression for the susceptibility, resulting in

$$\begin{aligned}
\chi''_{zz}(\mathbf{k}, \omega) = & \frac{\pi \hbar \sigma}{N} \sum_{\mathbf{k}'} \{ \\
& |u_{\alpha \mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 n_B(\hbar \omega_{\alpha(\mathbf{k}'-\mathbf{k})}) \delta(\omega_{\alpha \mathbf{k}'} - \omega_{\alpha(\mathbf{k}'-\mathbf{k})} + \omega) \\
& + |v_{\beta \mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'-\mathbf{k})}|^2 n_B(-\hbar \omega_{\beta(\mathbf{k}'-\mathbf{k})}) \delta(\omega_{\beta(\mathbf{k}'-\mathbf{k})} - \omega_{\beta \mathbf{k}'} + \omega) \\
& - |v_{\beta \mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'+\mathbf{k})}|^2 n_B(-\hbar \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \delta(\omega_{\beta \mathbf{k}'} - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega) \\
& - |u_{\alpha \mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 n_B(\hbar \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \delta(\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\alpha \mathbf{k}'} + \omega) \\
& - u_{\beta \mathbf{k}'} u_{\beta(\mathbf{k}'-\mathbf{k})} v_{\beta \mathbf{k}'} v_{\beta(\mathbf{k}'-\mathbf{k})} n_B(\hbar \omega_{\beta(\mathbf{k}'-\mathbf{k})}) \delta(\omega + \omega_{\beta \mathbf{k}'} - \omega_{\beta(\mathbf{k}'-\mathbf{k})}) \\
& - u_{\alpha \mathbf{k}'} u_{\alpha(\mathbf{k}'-\mathbf{k})} v_{\alpha \mathbf{k}'} v_{\alpha(\mathbf{k}'-\mathbf{k})} n_B(-\hbar \omega_{\alpha(\mathbf{k}'-\mathbf{k})}) \delta(\omega + \omega_{\alpha(\mathbf{k}'-\mathbf{k})} - \omega_{\alpha \mathbf{k}'}) \\
& + u_{\alpha \mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\alpha \mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} n_B(-\hbar \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \delta(\omega + \omega_{\alpha \mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \\
& + u_{\beta \mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\beta \mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} n_B(\hbar \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \delta(\omega + \omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta \mathbf{k}'}) \\
& - u_{\alpha \mathbf{k}'} u_{\alpha(\mathbf{k}'-\mathbf{k})} v_{\alpha \mathbf{k}'} v_{\alpha(\mathbf{k}'-\mathbf{k})} n_B(\hbar \omega_{\alpha(\mathbf{k}'-\mathbf{k})}) \delta(\omega + \omega_{\alpha \mathbf{k}'} - \omega_{\alpha(\mathbf{k}'-\mathbf{k})}) \\
& - u_{\beta \mathbf{k}'} u_{\beta(\mathbf{k}'-\mathbf{k})} v_{\beta \mathbf{k}'} v_{\beta(\mathbf{k}'-\mathbf{k})} n_B(-\hbar \omega_{\beta(\mathbf{k}'-\mathbf{k})}) \delta(\omega + \omega_{\beta(\mathbf{k}'-\mathbf{k})} - \omega_{\beta \mathbf{k}'}) \\
& + u_{\beta \mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\beta \mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} n_B(-\hbar \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \delta(\omega + \omega_{\beta \mathbf{k}'} - \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \\
& + u_{\alpha \mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\alpha \mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} n_B(\hbar \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \delta(\omega + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\alpha \mathbf{k}'}) \\
& + |u_{\beta \mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'-\mathbf{k})}|^2 n_B(\hbar \omega_{\beta(\mathbf{k}'-\mathbf{k})}) \delta(\omega_{\beta \mathbf{k}'} - \omega_{\beta(\mathbf{k}'-\mathbf{k})} + \omega) \\
& + |v_{\alpha \mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'-\mathbf{k})}|^2 n_B(-\hbar \omega_{\alpha(\mathbf{k}'-\mathbf{k})}) \delta(\omega_{\alpha(\mathbf{k}'-\mathbf{k})} - \omega_{\alpha \mathbf{k}'} + \omega) \\
& - |v_{\alpha \mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 n_B(-\hbar \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \delta(\omega_{\alpha \mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega) \\
& - |u_{\beta \mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'+\mathbf{k})}|^2 n_B(\hbar \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \delta(\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta \mathbf{k}'} + \omega) \}
\end{aligned}$$

We simplify even more this formula by taking advantage of the reciprocal lattice periodicity. For instance, for terms that depend on $\mathbf{k}' - \mathbf{k}$, we replace $\mathbf{k}' \rightarrow \mathbf{k}' + \mathbf{k}$ in the sums that contain them. Also, we take

the continuous limit $\sum_{\mathbf{k}'} \rightarrow \frac{N}{\sigma(2\pi)^2} \int d^2\mathbf{k}'$, resulting

$$\begin{aligned}
\chi''_{zz}(\mathbf{k}, \omega) = & \frac{\hbar}{4\pi} \int d^2\mathbf{k}' \{ \\
& |u_{\alpha\mathbf{k}'}|^2 |u_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \delta(\omega + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\alpha\mathbf{k}'}) \\
& \times [n_B(\hbar\omega_{\alpha\mathbf{k}'}) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& + |v_{\beta\mathbf{k}'}|^2 |v_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \delta(\omega + \omega_{\beta\mathbf{k}'} - \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \\
& \times [n_B(-\hbar\omega_{\beta\mathbf{k}'}) - n_B(-\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& - u_{\beta\mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\beta\mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} \delta(\omega + \omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta\mathbf{k}'}) \\
& \times [n_B(\hbar\omega_{\beta\mathbf{k}'}) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& - u_{\alpha\mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} \delta(\omega + \omega_{\alpha\mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \\
& \times [n_B(-\hbar\omega_{\alpha\mathbf{k}'}) - n_B(-\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& - u_{\alpha\mathbf{k}'} u_{\alpha(\mathbf{k}'+\mathbf{k})} v_{\alpha\mathbf{k}'} v_{\alpha(\mathbf{k}'+\mathbf{k})} \delta(\omega + \omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \omega_{\alpha\mathbf{k}'}) \\
& \times [n_B(\hbar\omega_{\alpha\mathbf{k}'}) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& - u_{\beta\mathbf{k}'} u_{\beta(\mathbf{k}'+\mathbf{k})} v_{\beta\mathbf{k}'} v_{\beta(\mathbf{k}'+\mathbf{k})} \delta(\omega + \omega_{\beta\mathbf{k}'} - \omega_{\beta(\mathbf{k}'+\mathbf{k})}) \\
& \times [n_B(-\hbar\omega_{\beta\mathbf{k}'}) - n_B(-\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& + |u_{\beta\mathbf{k}'}|^2 |u_{\beta(\mathbf{k}'+\mathbf{k})}|^2 \delta(\omega + \omega_{\beta(\mathbf{k}'+\mathbf{k})} - \omega_{\beta\mathbf{k}'}) \\
& \times [n_B(\hbar\omega_{\beta\mathbf{k}'}) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& + |v_{\alpha\mathbf{k}'}|^2 |v_{\alpha(\mathbf{k}'+\mathbf{k})}|^2 \delta(\omega + \omega_{\alpha\mathbf{k}'} - \omega_{\alpha(\mathbf{k}'+\mathbf{k})}) \\
& \times [n_B(-\hbar\omega_{\alpha\mathbf{k}'}) - n_B(-\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \}
\end{aligned}$$

Finally, using the identity $u_{\alpha\mathbf{k}'} u_{\alpha\mathbf{k}''} - v_{\alpha\mathbf{k}'} v_{\alpha\mathbf{k}''} = 1$, we obtain

$$\begin{aligned}
\chi''_{zz}(\mathbf{k}, \omega) = & \frac{\hbar}{4\pi} \int d^2\mathbf{k}' \{ \\
& [u_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega_{\alpha\mathbf{k}'}) + v_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\alpha\mathbf{k}'} - \mu_\alpha) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \mu_\alpha)] \\
& + [u_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega_{\beta\mathbf{k}'}) + v_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\beta\mathbf{k}'} - \mu_\beta) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \mu_\beta)] \},
\end{aligned}$$

where we have introduced the chemical potentials μ_α and μ_β subtracting them to every argument of the n_B functions. So, the longitudinal spin-spin correlator, according to dissipation-fluctuation theorem

(D.1), is

$$\begin{aligned}
C_{zz}(\mathbf{k}, \omega) = & \frac{\hbar k_B T}{2\pi\omega} \int d^2\mathbf{k}' \{ \\
& [u_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha(\mathbf{k}'+\mathbf{k})} + \omega_{\alpha\mathbf{k}'}) + v_{\alpha\mathbf{k}'}^2 \delta(\omega - \omega_{\alpha\mathbf{k}'} + \omega_{\alpha(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\alpha\mathbf{k}'} - \mu_\alpha) - n_B(\hbar\omega_{\alpha(\mathbf{k}'+\mathbf{k})} - \mu_\alpha)] \\
& + [u_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta(\mathbf{k}'+\mathbf{k})} + \omega_{\beta\mathbf{k}'}) + v_{\beta\mathbf{k}'}^2 \delta(\omega - \omega_{\beta\mathbf{k}'} + \omega_{\beta(\mathbf{k}'+\mathbf{k})})] \\
& \times [n_B(\hbar\omega_{\beta\mathbf{k}'} - \mu_\beta) - n_B(\hbar\omega_{\beta(\mathbf{k}'+\mathbf{k})} - \mu_\beta)] \}.
\end{aligned}$$

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