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BACHELOR THESIS

Magnon kinetics in thin-film magnetic insulators

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Abstract

By means of the Boltzmann equation we study the time evolution of the magnon distribution function for a magnetic insulator in contact with a normal metal. We consider a quasi-equilibrium realized by dc pumping of magnons, including Gilbert damping and magnon-magnon scattering processes. We show that the magnon distribution is well described using a Bose-Einstein distribution ansatz with time dependent magnon temperature and chemical potential. Furthermore we show that this model breaks down for low energies, and we calculate the crossover energy to be $\epsilon^* \approx 2.5$K. In order to study low-energy behavior, we derive the scattering rate due to magnon-magnon collisions. Starting from a Heisenberg exchange Hamiltonian, we exploit the Holstein-Primakoff transformation to find the magnon scattering amplitude. Using Fermi's golden rule, the scattering rate is derived.

Contents

Chapter 1

Introduction

Magnetic properties of materials ultimately arise due to specific orientations of electron spin [1]. In ferromagnetic materials, the magnetic ground state is occupied when all spins are aligned. For this state to be reached, the temperature of the system must be at absolute zero. At temperatures above 0K, excitations will emerge due to the availability of thermal energy. Such excitations are called spin waves, which can propagate through the material.

Within the language of quantum mechanics, a spin wave can be described as a quasi-particle called a magnon. Magnons carry energy as well as linear and angular momentum. They obey the Bose-Einstein statistics, as a consequence of their bosonic nature. Similar to other bosons, magnons can undergo Bose-Einstein condensation, in which a macroscopic occupation of the ground state is reached.

In order to realize such a phase of matter, a certain critical magnon density must be reached. There are different methods with which this can be achieved. Microwave pumping has been expected [2] and observed [3] to create magnon Bose-Einstein condensates. Another proposed method is the so called 'dc electronic pumping' [4]. Consider a conducting metal attached to a magnetic insulator. By applying a current to the conductor, electrons will scatter at the interface of the materials, transferring momentum to the insulator. This will cause a slight disturbance of the insulator spin density, by conservation of angular momentum. Therefore, magnons are effectively injected into the magnetic insulator.

Ref. [5] predicts the existence of dc pumped magnon Bose-Einstein condensates. The steady-state behavior of a thin film insulating magnet is studied, with a magnon distribution driven by the combination of a thermal gradient and electric potential. The thermal magnons undergo quasi-equilibration, as magnons are created and annihilated continuously.

In this thesis we go beyond the work of Ref. [5], which considers the system in quasi-equilibrium, and study the full time evolution of the magnon distribution function by means of the Boltzmann equation. We include Gilbert damping, which decreases the magnon density due to interactions with phonons, as well as elastic magnon-magnon scattering, which plays an important role in the equilibration of the magnon cloud. As in Ref. [5], we limit ourselves to a thin film magnet system, enabling us to use a spatially uniform magnon temperature.

In chapter 2, the theoretical framework for magnon dynamics is drawn and explored, deriving regimes in which different interactions dominate the Boltzmann equation. In chapter 3, we solve the Boltzmann equation with a Bose-Einstein distribution as an ansatz, and find equations for a time dependent magnon chemical potential and temperature. We find regions wherein the chemical potential converges to a non-zero value, thus showing a quasi-equilibrium state. In chapter 4 we derive the explicit form of magnon-magnon scattering processes, starting from a Heisenberg model Hamiltonian. Chapter 5 summarizes our results and offers an outlook for future research.

Chapter 2

Theoretical framework

Spin waves are a quantum mechanical phenomenon. They are excitations of the magnetic ground state of a system (see Fig. 2.1). So as to describe them properly, we look at these excitations as quasiparticles called magnons. Using the Holstein-Primakoff transformation (see chapter 4), the spin operators can be expressed in terms of magnon creation and annihilation operators, since magnons are bosonic particles.

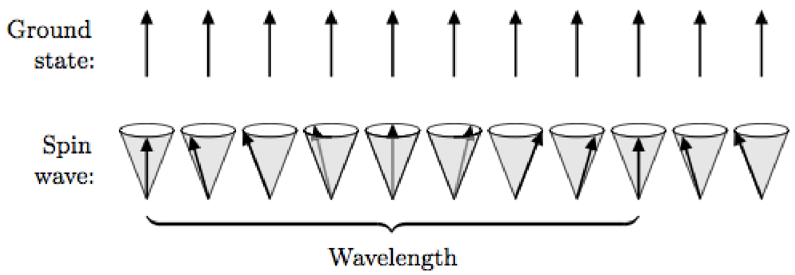

Figure 2.1 – In the ferromagnetic ground state, all spins are aligned. When excited, the individual spins precess around this ground state, creating spin waves. Copyright: Addison-Wesley 2000.

The system studied in this paper consists of a conducting metal attached to a thin film ferromagnetic insulator. See Fig. 2.2. Magnons are created by dc electronic pumping through the conductor. As magnons are bosons, they are described by Bose-Einstein statistics, with a proper magnon distribution function. This approach of describing magnons will not capture all quantum mechanical effects, such as interference between magnons, however it will give more insight into the nonequilibrium macroscopic behavior within the magnetic film. Due to the fact that we are studying a thin film, we approximate the magnon temperature to be spatially uniform. As a result, our magnon distribution function depends on time and energy only.

We define the magnon distribution $g(\epsilon, t)$ with the following formula:

$$\int d\epsilon D(\epsilon) g(\epsilon, t) = n_m, \tag{2.1}$$

with $D(\epsilon)$ the density of states and n_m the density of magnons. Assuming a gapless quadratic magnon dispersion in terms of the spin stiffness J_s, the magnon density of states is $D(\epsilon) = \sqrt{\epsilon}/4\pi^2 J_s^{3/2}$ [6].

In order to study the dynamics of the magnon distribution, we must consider the different interactions involved. Pumping of electrons causes magnetic excitations in the insulator, therefore increasing the magnon density. There is a decrease in density due to Gilbert damping. Furthermore, we include particle number preserving magnon-magnon scattering, which is important for the thermalization of the magnons. This results in a

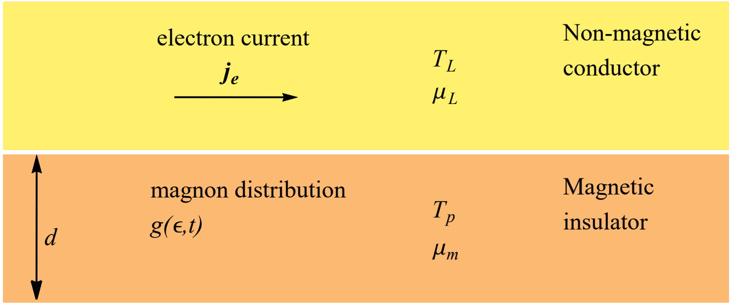

Figure 2.2 – A schematic representation of the setup studied. A non-magnetic conductor, with respective electronic temperature and spin accumulation T_L, μ_L, is placed on top of a ferromagnetic insulating film of thickness d. Within the magnetic film, a non-zero magnon chemical potential μ_m arises, which relaxes to the phonon temperature T_p.

Boltzmann equation of the following form:

$$\frac{\partial g(\epsilon,t)}{\partial t} = \Gamma_e + \Gamma_G + \Gamma_{mm}, \tag{2.2}$$

with Γ_e representing the electron-magnon interaction, Γ_G the Gilbert damping and Γ_{mm} the magnon-magnon scattering. The explicit forms of the interactions are given below (see Refs. [5] and [7]).

$$\begin{aligned}
\Gamma_e &= -\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi s d}\right)(\epsilon-\mu_L)\left[g(\epsilon,t) - n_B\left(\frac{\epsilon-\mu_L}{k_B T_L}\right)\right], \\
\Gamma_G &= -\left(\frac{2\alpha}{\hbar}\right)\epsilon\left[g(\epsilon,t) - n_B\left(\frac{\epsilon}{k_B T_p}\right)\right], \\
\Gamma_{mm} &= -\frac{2\pi}{\hbar}\iiint d\epsilon' d\epsilon'' d\epsilon''' D(\epsilon')D(\epsilon'')D(\epsilon''')|V|^2\delta(\epsilon+\epsilon'-\epsilon''-\epsilon''') \\
&\qquad \times \{g(\epsilon,t)g(\epsilon',t)\left(g(\epsilon'',t)+1\right)\left(g(\epsilon''',t)+1\right) \\
&\qquad -g(\epsilon'',t)g(\epsilon''',t)\left(g(\epsilon,t)+1\right)\left(g(\epsilon',t)+1\right)\}.
\end{aligned} \tag{2.3}$$

Here $g_{\uparrow\downarrow}$ is the real part of the spin mixing conductance, s the spin density and d the thickness of the magnetic film. The interactions are described by Bose-Einstein distributions $n_B(x) = (e^x - 1)^{-1}$, with electronic temperature and spin accumulation in the conductor respectively, T_L, μ_L, and phonon temperature T_p.

Steady states for each interaction are reached for Boltzmann distributions with the appropriate chemical potential and temperature. Before trying to solve the equation, it is insightful to calculate the different energy regimes of the processes. For the magnon-magnon scattering a relaxation-time approximation is used:

$$\Gamma_{mm}(\epsilon,t) \simeq -\frac{1}{\tau(\epsilon)}\left[g(\epsilon,t) - n_B\left(\frac{\epsilon-\mu_m}{k_B T_m}\right)\right]. \tag{2.4}$$

Furthermore we use $1/\tau(\epsilon) = g\epsilon^4$ with $g \approx 10^{97}\mathrm{J}^{-4}\mathrm{s}^{-1}$. For the derivation of this energy dependence, see chapter 4.

The electron interaction term and the Gilbert damping term have the same energy dependence when neglecting μ_L. This is valid for high energies. Equating these terms then yields a crossover thickness:

$$d^* = \frac{g_{\uparrow\downarrow}}{2\alpha\pi s} \approx 10^{-6}\mathrm{m}. \tag{2.5}$$

The values for the constants are taken from Ref. [7], and are appropriate for a Pt-YIG setup. When the thickness d of the ferromagnet is small enough ($d < d^*$), the Gilbert damping term will be dominated by the electron pumping, maintaining a non-zero magnon density. Physically, this is due to the fact that lattice interactions are volume effects, while the electron interaction is a surface effect. The ratio of the volume and the surface determines the ratio of the strengths of these interactions.

Neglecting the effect of Gilbert damping, we can equate the resulting terms to find a crossover energy

$$\epsilon^* = \left(\frac{g_{\uparrow\downarrow}}{\hbar\pi s d g}\right)^{\frac{1}{3}} \approx 10^{-23}\text{J}, \tag{2.6}$$

which corresponds to a temperature of approximately 2.5K. For energies higher than ϵ^*, magnon-magnon scattering dominates over electron-magnon interactions at the interface. This will cause the magnons to thermalize and reach a quasi-equilibrium state. We thus expect that for energies larger than ϵ^* the distribution function will be well described by a Bose distribution function with nonzero chemical potential.

Chapter 3

Time dependent Bose-Einstein distribution

In this chapter we solve the Boltzmann equation assuming a Bose-Einstein distribution for $g(\epsilon, t)$. We will work with the following ansatz:

$$g(\epsilon, t) = \frac{1}{e^{\frac{\epsilon - \mu_m(t)}{k_B T_m(t)}} - 1}, \tag{3.1}$$

assuming that μ_m and T_m are time dependent. For convenience, we will write $(k_B T_m)^{-1} = \beta_m$. Furthermore, we introduce an energy gap Δ in the density of states and magnon dispersion. This model offers a good description of the high-energy regime, in which magnon-magnon interaction dominates. The magnon-magnon term will not contribute to the equation since our ansatz is a steady state solution of this interaction, i.e. Γ_{mm} in Eq. (2.3) is zero for the above distribution function. In order to solve the Boltzmann equation, it is convenient to split it into two separate differential equations for the magnon number density and magnon energy. The equations are derived in the sections below. We will limit ourselves to linear response.

3.1 Magnon density equation

Taking a time derivative of Eq. (2.1), we get

$$\frac{\partial n_m}{\partial t} = \frac{\partial}{\partial t} \int d\epsilon D(\epsilon) g(\epsilon, t) = \int d\epsilon D(\epsilon) \frac{\partial}{\partial t} g(\epsilon, t). \tag{3.2}$$

Since we assume only μ_m and T_m to be time dependent, we expect the derivative of the magnon density to have the following form:

$$\frac{\partial n_m}{\partial t} = A \frac{\partial \mu_m}{\partial t} + B \frac{\partial T_m}{\partial t}. \tag{3.3}$$

Assuming T_m to be constant, we can write:

$$\frac{\partial n_m}{\partial t} = \frac{\partial}{\partial t} \int d\epsilon D(\epsilon) g(\epsilon, t) = \frac{\partial \mu_m}{\partial t} \int d\epsilon D(\epsilon) \frac{\partial}{\partial \mu_m} g(\epsilon, t) = -\frac{\partial \mu_m}{\partial t} \int d\epsilon D(\epsilon) \frac{\partial}{\partial \epsilon} g(\epsilon, t). \tag{3.4}$$

Using partial integration, shifting the integral and using substitution of variables we can simplify this integral. We repeat this procedure taking μ_m constant instead of T_m. The final result is:

$$\begin{aligned} \frac{4\pi^2}{J_s^{3/2}} \frac{\partial n_m}{\partial t} = {} & \dot{\mu}_m \left[\tfrac{1}{2} \sqrt{\pi} \beta_m^{-1/2} Li_{-\frac{1}{2}}(e^{(\mu_m - \Delta)\beta_m}) \right] \\ & + \dot{T}_m k_B \left[\tfrac{3}{4} \sqrt{\pi} \beta_m^{-1/2} Li_{\frac{1}{2}}(e^{(\mu_m - \Delta)\beta_m}) + \tfrac{\Delta}{2} \sqrt{\pi} \beta_m^{1/2} Li_{-\frac{1}{2}}(e^{(\mu_m - \Delta)\beta_m}) \right], \end{aligned} \tag{3.5}$$

with $Li_n(z)$ the polylogarithmic function.

Now we will work out the right hand side of Eq. (3.2) by substituting our ansatz.

$$\int d\epsilon D(\epsilon)\frac{\partial}{\partial t}g(\epsilon,t) = \frac{J_s^{3/2}}{4\pi^2}\int_\Delta^\infty \sqrt{\epsilon-\Delta} \\ \times\left[\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi sd}\right)(\epsilon-\mu_L)\left[g(\epsilon,t)-n_B\left((\epsilon-\mu_L)\beta_L\right)\right]-\left(\frac{2\alpha}{\hbar}\right)\epsilon\left[g(\epsilon,t)-n_B\left(\epsilon\beta_p\right)\right]\right]. \tag{3.6}$$

With the same methods as before, we find

$$\begin{aligned}\frac{1}{J_s^{3/2}}\frac{\partial n_m}{\partial t} &= \\ &-\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi sd}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_m^{-5/2}Li_{\frac{5}{2}}(e^{(\mu_m-\Delta)\beta_m})+\frac{1}{2}\sqrt{\pi}(\Delta-\mu_L)\beta_m^{-3/2}Li_{\frac{3}{2}}(e^{(\mu_m-\Delta)\beta_m})\right] \\ &+\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi sd}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_L^{-5/2}Li_{\frac{5}{2}}(e^{(\mu_L-\Delta)\beta_L})+\frac{1}{2}\sqrt{\pi}(\Delta-\mu_L)\beta_L^{-3/2}Li_{\frac{3}{2}}(e^{(\mu_L-\Delta)\beta_L})\right] \\ &-\left(\frac{2\alpha}{\hbar}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_m^{-5/2}Li_{\frac{5}{2}}(e^{(\mu_m-\Delta)\beta_m})+\frac{1}{2}\sqrt{\pi}\Delta\beta_m^{-3/2}Li_{\frac{3}{2}}(e^{(\mu_m-\Delta)\beta_m})\right] \\ &+\left(\frac{2\alpha}{\hbar}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_p^{-5/2}Li_{\frac{5}{2}}(e^{-\Delta\beta_p})+\frac{1}{2}\sqrt{\pi}\Delta\beta_p^{-3/2}Li_{\frac{3}{2}}(e^{-\Delta\beta_p})\right].\end{aligned} \tag{3.7}$$

In order to solve the equations analytically, we need to linearize them. We assume the magnon temperature T_m relaxes to the phonon temperature T_p. We also linearize with respect to the electron temperature and the chemical potentials. We get:

$$\begin{aligned}T_m(t) &= T_p+\delta T_m(t) \\ T_L &= T_p+\delta T_L \\ \mu_m(t) &= 0+\mu_m(t) \\ \mu_L &= 0+\mu_L\end{aligned} \tag{3.8}$$

After linearization, we find as our final equation for the magnon density:

$$\begin{aligned}&\dot{\mu}_m\left[\frac{1}{2}\sqrt{\pi}\beta_p^{-1/2}Li_{-\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\ &+\dot{T}_m k_B\left[\frac{3}{4}\sqrt{\pi}\beta_p^{-1/2}Li_{\frac{1}{2}}(e^{-\Delta\beta_p})+\frac{\Delta}{2}\sqrt{\pi}\beta_p^{1/2}Li_{-\frac{1}{2}}(e^{-\Delta\beta_p})\right] = \\ &-(\mu_m-\mu_L)\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi sd}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_p^{-3/2}Li_{\frac{3}{2}}(e^{-\Delta\beta_p})+\frac{\Delta}{2}\sqrt{\pi}\beta_p^{-1/2}Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\ &-(\mu_m)\left(\frac{2\alpha}{\hbar}\right)\left[\frac{3}{4}\sqrt{\pi}\beta_p^{-3/2}Li_{\frac{3}{2}}(e^{-\Delta\beta_p})+\frac{\Delta}{2}\sqrt{\pi}\beta_p^{-1/2}Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\ &-(T_m-T_L)k_B\left(\frac{g_{\uparrow\downarrow}}{\hbar\pi sd}\right)\left[\frac{15}{8}\sqrt{\pi}\beta_p^{-3/2}Li_{\frac{5}{2}}(e^{-\Delta\beta_p})+\frac{3\Delta}{2}\sqrt{\pi}\beta_p^{-1/2}Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) \right. \\ &\left. +\frac{\Delta^2}{2}\sqrt{\pi}\beta_p^{1/2}Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\ &-(T_m-T_p)k_B\left(\frac{2\alpha}{\hbar}\right)\left[\frac{15}{8}\sqrt{\pi}\beta_p^{-3/2}Li_{\frac{5}{2}}(e^{-\Delta\beta_p})+\frac{3\Delta}{2}\sqrt{\pi}\beta_p^{-1/2}Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) \right. \\ &\left. +\frac{\Delta^2}{2}\sqrt{\pi}\beta_p^{1/2}Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right],\end{aligned} \tag{3.9}$$

where $\dot{x}$ denotes the temporal derivative of variable x.

3.2 Magnon energy equation

In our system, the mean energy per volume is defined as

$$u_m = \int d\epsilon D(\epsilon)\epsilon g(\epsilon,t). \tag{3.10}$$

Using the same methods as for the magnon density equation, we can derive a differential equation for $T_m(t)$ and $\mu_m(t)$, and linearize it to enable us to solve it analytically. We find

$$\begin{aligned}
&\dot{\mu}_m \left[\tfrac{3}{4}\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p}) + \tfrac{\Delta}{2}\sqrt{\pi}\beta_p^{-1/2} Li_{-\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\
&+\dot{T}_m k_B \left[\tfrac{15}{8}\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) + \tfrac{3\Delta}{2}\sqrt{\pi}\beta_p^{-1/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p}) \right. \\
&\qquad \left. + \tfrac{\Delta^2}{2}\sqrt{\pi}\beta_p^{1/2} Li_{-\frac{1}{2}}(e^{-\Delta\beta_p})\right] = \\
&-(\mu_m - \mu_L)\left(\tfrac{g_{\uparrow\downarrow}}{\hbar\pi s d}\right)\left[\tfrac{15}{8}\sqrt{\pi}\beta_p^{-5/2} Li_{\frac{5}{2}}(e^{-\Delta\beta_p}) + \tfrac{3\Delta}{2}\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) \right. \\
&\qquad \left. + \tfrac{\Delta^2}{2}\sqrt{\pi}\beta_p^{-1/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\
&-(\mu_m)\left(\tfrac{2\alpha}{\hbar}\right)\left[\tfrac{15}{8}\sqrt{\pi}\beta_p^{-5/2} Li_{\frac{5}{2}}(e^{-\Delta\beta_p}) + \tfrac{3\Delta}{2}\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) \right. \\
&\qquad \left. + \tfrac{\Delta^2}{2}\sqrt{\pi}\beta_p^{-1/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p}\right] \\
&-(T_m - T_L)k_B\left(\tfrac{g_{\uparrow\downarrow}}{\hbar\pi s d}\right)\left[\tfrac{105}{16}\sqrt{\pi}\beta_p^{-5/2} Li_{\frac{7}{2}}(e^{-\Delta\beta_p}) + \tfrac{15}{8}\Delta\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{5}{2}}(e^{-\Delta\beta_p}) \right. \\
&\qquad \left. - \tfrac{3\Delta^2}{4}\sqrt{\pi}\beta_p^{-1/2} Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) - \tfrac{\Delta^3}{2}\sqrt{\pi}\beta_p^{1/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right] \\
&-(T_m - T_p)k_B\left(\tfrac{2\alpha}{\hbar}\right)\left[\tfrac{105}{16}\sqrt{\pi}\beta_p^{-5/2} Li_{\frac{7}{2}}(e^{-\Delta\beta_p}) + \tfrac{15}{8}\Delta\sqrt{\pi}\beta_p^{-3/2} Li_{\frac{5}{2}}(e^{-\Delta\beta_p}) \right. \\
&\qquad \left. - \tfrac{3\Delta^2}{4}\sqrt{\pi}\beta_p^{-1/2} Li_{\frac{3}{2}}(e^{-\Delta\beta_p}) - \tfrac{\Delta^3}{2}\sqrt{\pi}\beta_p^{1/2} Li_{\frac{1}{2}}(e^{-\Delta\beta_p})\right].
\end{aligned} \tag{3.11}$$

3.3 Results

The differential equations can be expressed in the following form:

$$\begin{aligned}
\hbar(\partial_\mu n\dot{\mu}_m + \partial_T n\dot{T}_m) &= G(\mu_m - \mu_L) + S(T_m - T_L) + G_p(\mu_m) + S_p(T_m - T_p); \\
\hbar(\partial_\mu u\dot{\mu}_m + \partial_T u\dot{T}_m) &= \Pi(\mu_m - \mu_L) + \kappa(T_m - T_L) + \Pi_p(\mu_m) + \kappa_p(T_m - T_p),
\end{aligned} \tag{3.12}$$

with G the interface spin conductance, S the Seebeck coefficient, Π the interface spin Peltier coefficient, κ the interface heat coefficient, and $G_p, S_p, \Pi_p, \kappa_p$ similar transport coefficients describing the magnon-phonon interactions. We note that the phonon processes are not interfacial, but take place inside the magnetic insulator. Therefore they are not really transport coefficients. We find that up to linear order $\Pi = TS$, in accordance with Onsagers reciprocity relations.

It is insightful to rewrite the equations into a dimensionless form prior to solving them. This will make the physical properties more transparent. We find

$$\begin{aligned}
\partial_{\tilde{t}}\tilde{\mu}_m + \eta_1\partial_{\tilde{t}}\tilde{T}_m &= \eta_2(\tilde{\mu}_m - 1) + \eta_3(\tilde{T}_m - \tilde{T}_L) + \eta_4\tilde{\mu}_m + \eta_5(\tilde{T}_m - \tilde{T}_p); \\
\partial_{\tilde{t}}\tilde{\mu}_m + \nu_1\partial_{\tilde{t}}\tilde{T}_m &= \nu_2(\tilde{\mu}_m - 1) + \nu_3(\tilde{T}_m - \tilde{T}_L) + \nu_4\tilde{\mu}_m + \nu_5(\tilde{T}_m - \tilde{T}_p),
\end{aligned} \tag{3.13}$$

where we made use of the following substitutions:

$$\begin{aligned}
&\tilde{t} = \frac{-G}{\hbar \partial_\mu n} t \equiv \frac{t}{t_0}, \\
&\tilde{\mu}_m = \frac{\mu_m}{\mu_L}, \quad \tilde{T}_m = \frac{k_B T_m}{\mu_L}, \quad \tilde{T}_p = \frac{k_B T_p}{\mu_L}, \quad \tilde{T}_L = \frac{k_B T_L}{\mu_L} \\
&\eta_1 = \frac{1}{k_B}\frac{\partial_T n}{\partial_\mu n}, \quad \eta_2 = -1, \quad \eta_3 = \frac{-S}{G k_B}, \quad \eta_4 = \frac{-G_p}{G}, \quad \eta_5 = \frac{-S_p}{G k_B} \\
&\nu_1 = \frac{1}{k_B}\frac{\partial_T u}{\partial_\mu u}, \quad \nu_2 = \frac{-\partial_\mu n}{\partial_\mu u}\frac{\Pi}{G}, \quad \nu_3 = \frac{-1}{k_B}\frac{\partial_\mu n}{\partial_\mu u}\frac{\kappa}{G}, \quad \nu_4 = \frac{-\partial_\mu n}{\partial_\mu u}\frac{\Pi_p}{G}, \quad \nu_5 = \frac{-1}{k_B}\frac{\partial_\mu n}{\partial_\mu u}\frac{\kappa_p}{G}.
\end{aligned} \tag{3.14}$$

The dimensionless magnon chemical potential is shown as a function of time in Fig. 3.1. At $t = 0$, magnon pumping starts. We see that the chemical potential rises to a non-zero value. This is evidence for a quasi-equilibrium distribution of magnons.

The behavior of the characteristic time t_0 is shown at the bottom of Fig. 3.1. In the limit where $\Delta \to 0$, t_0 diverges. As a consequence, t/t_0 will vanish and the magnon chemical potential will remain constant. This is an nonphysical situation resulting from the fact that our ansatz in Eq. (3.1) does not hold for energies below ϵ^*. Below this energy magnon-magnon interaction is dominated by electron pumping, preventing the magnons from internally equilibrating. The magnon distribution is then poorly described by a Bose-Einstein distribution.

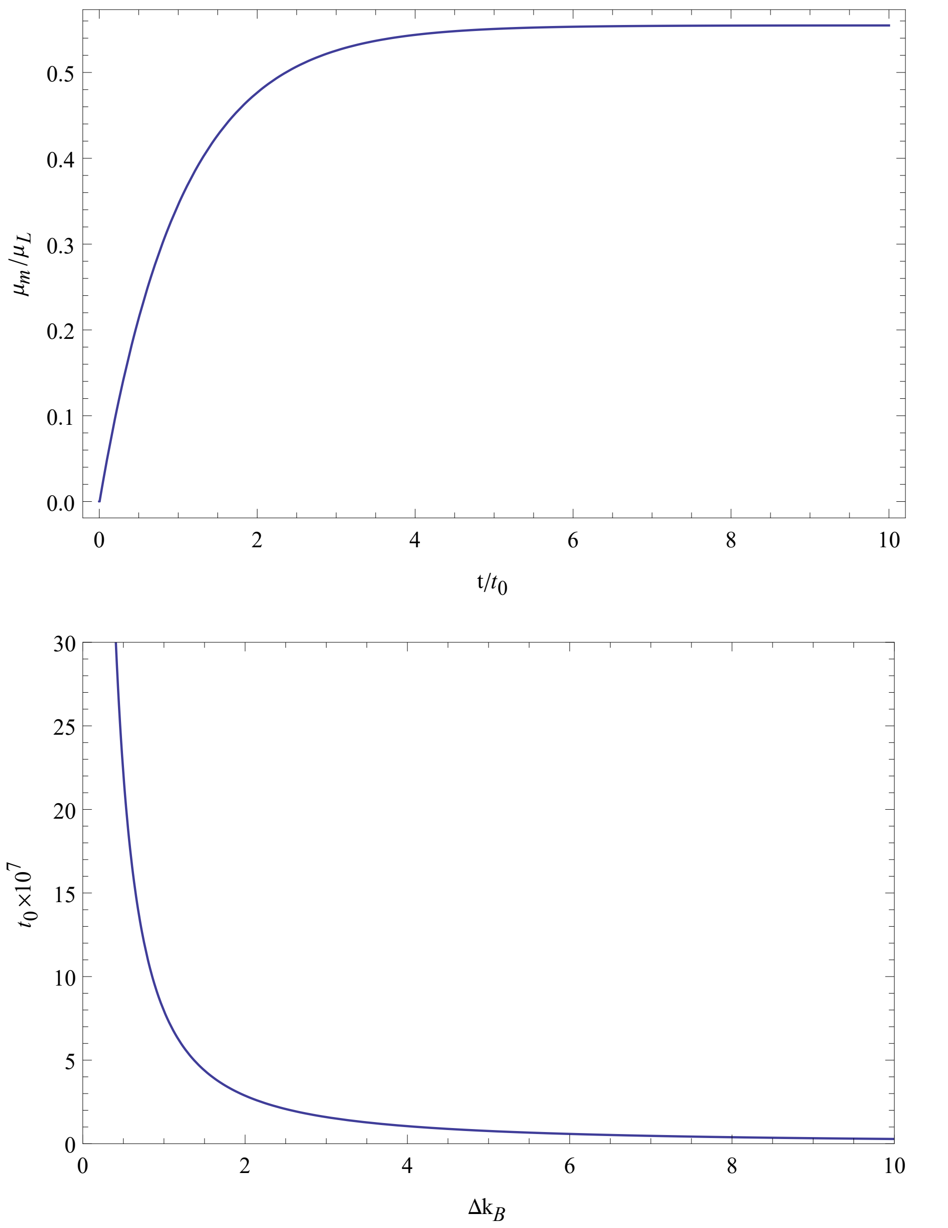

Figure 3.1 – Time evolution of the magnon chemical potential μ_m (top) and the characteristic time t_0 as a function of the energy gap Δ (bottom). We used $d = 10^{-6}$m$, T_p = T_L = 300$K. For the calculation of μ_m we used $\Delta = 1\text{K} \times k_B$.

Chapter 4

Magnon-magnon interactions

In this chapter we derive the general form of the magnon-magnon interaction, which enables us to study the behavior of the Boltzmann equation for lower energies. We are interested in interactions that conserve the number of magnons. These processes redistribute the energy of the magnons, thus causing thermalization of the system. This makes Bose-Einstein condensation of magnons possible.

The derivations in this chapter are built on previous research. In Ref. [8] the fourth order term of the Hamiltonian is derived. For Fermi's golden rule, Ref. [9] was used. Furthermore, Ref. [10] and Ref. [11] were used for the derivation of the collision integral.

We consider a Heisenberg model with exchange interactions, with spins of magnitude S on a cubic lattice with constant a. The Hamiltonian has the following form:

$$\mathcal{H} = -h\sum_{\mathbf{m}} S^z_{\mathbf{m}} - \frac{1}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}} S_{\mathbf{m}} \cdot S_{\mathbf{n}}, \tag{4.1}$$

where h is the coupling to an external magnetic field in the z-direction, while the second term embodies the nearest neighbor interaction between two spins at lattice positions $\mathbf{r_m} = a\mathbf{m}$ and $\mathbf{r_n} = a\mathbf{n}$. It is convenient to express this Hamiltonian in terms of the magnon creation and annihilation operators, $b^\dagger$ and b respectively. We use the Holstein-Primakoff transformation to rewrite the spin operators

$$\begin{aligned} S^+ &= \sqrt{2S}\sqrt{1-\frac{b^\dagger b}{2S}}\,b \approx \sqrt{2S}\left(b - \frac{b^\dagger b b}{4S} + O(b^5)\right), \\ S^- &= \sqrt{2S}b^\dagger\sqrt{1-\frac{b^\dagger b}{2S}} \approx \sqrt{2S}\left(b^\dagger - \frac{b^\dagger b^\dagger b}{4S} + O(b^5)\right), \\ S^z &= S - b^\dagger b, \end{aligned} \tag{4.2}$$

with $S^+ = S^x + iS^y$ and $S^- = S^x - iS^y$. After substitution of this transformation, we split the terms of different orders to find

$$\begin{aligned} \mathcal{H}_0 &= -hSN - \tfrac{1}{2}S^2 J_0 N, \\ \mathcal{H}_2 &= h\sum_{\mathbf{m},\mathbf{n}} \delta_{\mathbf{m},\mathbf{n}} b^\dagger_{\mathbf{m}} b_{\mathbf{n}} - S\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}(1-\delta_{\mathbf{m},\mathbf{n}}) b^\dagger_{\mathbf{m}} b_{\mathbf{n}}, \\ \mathcal{H}_4 &= \frac{1}{4}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left(b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} + b^\dagger_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} b_{\mathbf{n}} - 2 b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}}\right). \end{aligned} \tag{4.3}$$

For a full derivation of the Hamiltonian, see appendix A. Note that there are no first and third order terms in the Hamiltonian. The magnon interaction processes we are interested in are described by the fourth order terms. Fourier transforming $\mathcal{H}_4$ using $b_{\mathbf{m}} = \sum_{\mathbf{k}} b_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_m}/N$ where $\mathbf{k}$ is the magnon wave vector and N is the number of lattice sites, and normal ordering afterwards, reads

$$\begin{aligned}\mathcal{H}_4 =&\frac{1}{2Na^3}\sum_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} b^\dagger_{\mathbf{k1}} b^\dagger_{\mathbf{k2}} b_{\mathbf{k3}} b_{\mathbf{k4}} \delta_{\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4} \Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} a^3 \\ &+ \frac{1}{2N}\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b^\dagger_{\mathbf{k}} 2 \sum_{\alpha} \left(\cos(a\alpha\cdot\mathbf{k}') - \cos(a\alpha\cdot(\mathbf{k}-\mathbf{k}'))\right).\end{aligned} \tag{4.4}$$

We find the magnon-magnon scattering amplitude to be

$$\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} = J_0 \sum_{\alpha} \left(\cos(a\alpha\cdot\mathbf{k}_1) + \cos(a\alpha\cdot\mathbf{k}_4) - 2\cos(a\alpha\cdot(\mathbf{k}_4-\mathbf{k}_2))\right), \tag{4.5}$$

with $\alpha = \{\hat{x}, \hat{y}, \hat{z}\}$. Considering the case in which $\mathbf{k}_i$ is small, we can approximate the amplitude

$$\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} \approx \frac{J_0 a^2}{2}\left(-\mathbf{k}_1^2 - \mathbf{k}_4^2 + 2(\mathbf{k}_4-\mathbf{k}_2)^2\right). \tag{4.6}$$

Before we continue, we will rewrite this expression into a more convenient form. By using the following identities

$$\begin{aligned}\mathbf{k}_1^2 &= \mathbf{k}_1\cdot(\mathbf{k}_3+\mathbf{k}_4-\mathbf{k}_2) = -\mathbf{k}_1\cdot\mathbf{k}_2 + \mathbf{k}_1\cdot\mathbf{k}_3 + \mathbf{k}_1\cdot\mathbf{k}_4, \\ \mathbf{k}_2^2 &= \mathbf{k}_2\cdot(\mathbf{k}_3+\mathbf{k}_4-\mathbf{k}_1) = -\mathbf{k}_1\cdot\mathbf{k}_2 + \mathbf{k}_2\cdot\mathbf{k}_3 + \mathbf{k}_2\cdot\mathbf{k}_4, \\ \mathbf{k}_4^2 &= \mathbf{k}_4\cdot(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3) = -\mathbf{k}_3\cdot\mathbf{k}_4 + \mathbf{k}_1\cdot\mathbf{k}_4 + \mathbf{k}_2\cdot\mathbf{k}_4,\end{aligned} \tag{4.7}$$

we can rewrite the amplitude

$$\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} \approx \frac{J_0 a^2}{2}\left(-\mathbf{k}_1\cdot\mathbf{k}_2 - \mathbf{k}_3\cdot\mathbf{k}_4 + \mathbf{k}_2\cdot(\mathbf{k}_3-\mathbf{k}_4) - \mathbf{k}_3\cdot(\mathbf{k}_1-\mathbf{k}_2)\right). \tag{4.8}$$

The last two terms in this expressions will cancel, since we sum over all values for $\mathbf{k}_i$ in the Hamiltonian. Therefore, we may leave them out, resulting in the following expression:

$$\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} \approx \frac{J_0 a^2}{2}\left(-\mathbf{k}_1\cdot\mathbf{k}_2 - \mathbf{k}_3\cdot\mathbf{k}_4\right). \tag{4.9}$$

Using Fermi's golden rule, we calculate the rate of the scattering of magnons.

$$\begin{aligned}\Gamma_{mm}[f] = \frac{\pi}{\hbar}\frac{1}{(2\pi)^9} \iiint & d\mathbf{k}_2 d\mathbf{k}_3 d\mathbf{k}_4 |\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} a^3|^2 \\ &\times \delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\delta(\epsilon_1+\epsilon_2-\epsilon_3-\epsilon_4) \\ &\times \left[f(\mathbf{k}_3,t)f(\mathbf{k}_4,t)\left(1+f(\mathbf{k}_1,t)\right)\left(1+f(\mathbf{k}_2,t)\right)\right. \\ &\left. - f(\mathbf{k}_1,t)f(\mathbf{k}_2,t)\left(1+f(\mathbf{k}_3,t)\right)\left(1+f(\mathbf{k}_4,t)\right)\right],\end{aligned} \tag{4.10}$$

with ϵ_i the energy of magnon i. The second line is a statistical factor which ensures that in equilibrium a Bose-Einstein distribution is reached. The bosonic nature of the magnons is reflected in the $(1 + f(k,t))$ terms, which would have been $(1 - f(k,t))$ if we were dealing with fermions.

Next, we derive the the energy dependence of Γ_{mm} using the relaxation-time approximation. This approximation amounts to

$$\Gamma_{mm} \approx -\frac{1}{\tau_{\mathbf{k}}}\left[g(\epsilon,t) - n_B\left(\frac{\epsilon-\mu_m}{k_B T_m}\right)\right], \tag{4.11}$$

with $g(\epsilon, t)$ the magnon distribution as before. The relaxation time has the following form

$$\begin{aligned}\frac{1}{\tau_{\mathbf{k}}} = \frac{\pi}{\hbar}\frac{a^6}{(2\pi)^9}\iiint & d\mathbf{k}_2 d\mathbf{k}_3 d\mathbf{k}_4 |\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}}|^2 \\ & \times \delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\delta(\epsilon_1+\epsilon_2-\epsilon_3-\epsilon_4) \\ & \times [n_B(\epsilon_2)(1+n_B(\epsilon_3))(1+n_B(\epsilon_4))]\,.\end{aligned} \tag{4.12}$$

By substituting $\mathbf{k}_i = \mathbf{x}_i/\Lambda$, with $\Lambda = \frac{h}{\sqrt{2\pi m k_B T}}$ the De Broglie wavelength, we can make the integrals dimensionless. We find

$$\begin{aligned}\frac{1}{\tau_{\mathbf{k}}} = \frac{1}{\Lambda^8}\frac{2m}{\hbar^2}\frac{\pi}{\hbar}\frac{a^6}{(2\pi)^9}\frac{J_0^2 a^4}{4}\iiint & d\mathbf{x}_2 d\mathbf{x}_3 d\mathbf{x}_4 |\Gamma_{\mathbf{x1},\mathbf{x2},\mathbf{x3},\mathbf{x4}}|^2 \\ & \times \delta(\mathbf{x}_1+\mathbf{x}_2-\mathbf{x}_3-\mathbf{x}_4)\delta(\mathbf{x}_1^2+\mathbf{x}_2^2-\mathbf{x}_3^2-\mathbf{x}_4^2) \\ & \times [n_B(\epsilon_2)(1+n_B(\epsilon_3))(1+n_B(\epsilon_4))]\,.\end{aligned} \tag{4.13}$$

Now we define $J_0 = k_B T_c$. Using $\hbar^2/2m = J_0 a^2$, we can rewrite the expression for the scattering time

$$\begin{aligned}\frac{1}{\tau_{\mathbf{k}}} = \frac{1}{\hbar}\frac{1}{(k_B T_c)^3}(k_B T)^4\frac{\pi}{(2\pi)^{13}}\frac{1}{64}\iiint & d\mathbf{x}_2 d\mathbf{x}_3 d\mathbf{x}_4 |\Gamma_{\mathbf{x1},\mathbf{x2},\mathbf{x3},\mathbf{x4}}|^2 \\ & \times \delta(\mathbf{x}_1+\mathbf{x}_2-\mathbf{x}_3-\mathbf{x}_4)\delta(\mathbf{x}_1^2+\mathbf{x}_2^2-\mathbf{x}_3^2-\mathbf{x}_4^2) \\ & \times [n_B(\epsilon_2)(1+n_B(\epsilon_3))(1+n_B(\epsilon_4))] \\ & \equiv g\cdot(k_B T)^4,\end{aligned} \tag{4.14}$$

with

$$g = \frac{1}{\hbar}\frac{1}{(k_B T_c)^3}\times(\text{some numerical factor}) \approx 10^{97}\mathrm{J}^{-4}\mathrm{s}^{-1}, \tag{4.15}$$

approximating the numerical factor to be of order 1. We conclude that the magnon-magnon scattering process depends on the energy as ϵ^4. This justifies our use of this approximation in chapter 1.

Now we will calculate the collision term explicitly. Due to the fact that we are dealing with a thin film, our magnon distribution is assumed to be spatially uniform. Therefore, we may use $f(\mathbf{k}, t) = g(\epsilon(\mathbf{k}), t))$. Starting from Eq. (4.10), we make a phase space projection of the collision integral

$$\Gamma_{mm}(\epsilon, t) = \int \frac{d\mathbf{k}}{(2\pi)^3}\delta(\epsilon - E(\mathbf{k}))\Gamma_{mm}[f]. \tag{4.16}$$

We want to express the collision integral in terms of the energy. In order to accomplish this, we use the following relation

$$\frac{1}{(2\pi)^3}\int d\mathbf{k} = \int d\epsilon D(\epsilon) = \int d\epsilon\left(\frac{1}{(2\pi)^3}\int d\mathbf{k}\delta(\epsilon - E(\mathbf{k}))\right). \tag{4.17}$$

The collision integral then reads

$$\begin{aligned}\Gamma_{mm}(\epsilon_1, t) = \frac{\pi}{\hbar}\frac{a^6}{(2\pi)^{12}}\iiint & d\epsilon_2 d\epsilon_3 d\epsilon_4 \delta(\epsilon_1+\epsilon_2-\epsilon_3-\epsilon_4) \\ & [g(\epsilon_3,t)g(\epsilon_4,t)\left(1+g(\epsilon_1,t)\right)\left(1+g(\epsilon_2,t)\right) \\ & -g(\epsilon_1,t)g(\epsilon_2,t)\left(1+g(\epsilon_3,t)\right)\left(1+g(\epsilon_4,t)\right)] \\ \times & \iiiint d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 d\mathbf{k}_4 |\Gamma_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}}|^2 \delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4) \\ & \delta(\epsilon_1 - E_1(\mathbf{k}))\delta(\epsilon_2 - E_2(\mathbf{k}))\delta(\epsilon_3 - E_3(\mathbf{k}))\delta(\epsilon_4 - E_4(\mathbf{k})).\end{aligned} \tag{4.18}$$

Now we focus on working out the momentum integrals. We transform to total momenta $\mathbf{K} = \mathbf{k}_1 + \mathbf{k}_2, \mathbf{K}' = \mathbf{k}_3 + \mathbf{k}_4$ and relative momenta $\mathbf{q} = (\mathbf{k}_1 - \mathbf{k}_2)/2, \mathbf{q}' = (\mathbf{k}_3 - \mathbf{k}_4)/2$. The momentum conserving delta function now yields $\mathbf{K} = \mathbf{K}'$. We rewrite the delta functions and transform to spherical coordinates.

$$\begin{aligned} \delta(\epsilon_1 - E_1) = \delta(\epsilon_1 - \tfrac{\hbar^2 \mathbf{k}_1^2}{2m}) &= \frac{2m}{\hbar^2}\delta(\tfrac{2m\epsilon_1}{\hbar^2} - \tfrac{K^2}{4} - q^2 - \mathbf{K}\cdot\mathbf{q}) \\ &= \frac{2m}{\hbar^2}\frac{1}{Kq}\delta(\tfrac{1}{Kq}(\tfrac{2m\epsilon_1}{\hbar^2} - \tfrac{K^2}{4} - q^2) - \cos(\theta')). \end{aligned} \tag{4.19}$$

The other delta functions are transformed in a similar fashion. Furthermore we rewrite the scattering amplitude

$$|\Gamma_{\mathbf{k}1,\mathbf{k}2,\mathbf{k}3,\mathbf{k}4}|^2 = |\Gamma_{\mathbf{K},\mathbf{q},\mathbf{q}'}|^2 = \frac{J_0^2 a^4}{4}|q^2 + q'^2 - \tfrac{1}{2}K^2|^2. \tag{4.20}$$

The momentum integrals now take the following form

$$\begin{aligned} (2\pi)^2\left(\frac{2m}{\hbar^2}\right)^4 \int d\mathbf{K}\frac{1}{K^4} \iint dq dq' |\Gamma_{\mathbf{K},\mathbf{q},\mathbf{q}'}|^2 \iint du du' \\ \times\delta(\tfrac{1}{Kq}(\tfrac{2m\epsilon_1}{\hbar^2} - \tfrac{K^2}{4} - q^2) - u') \\ \delta(\tfrac{1}{Kq}(\tfrac{2m\epsilon_2}{\hbar^2} - \tfrac{K^2}{4} - q^2) + u') \\ \delta(\tfrac{1}{Kq}(\tfrac{2m\epsilon_3}{\hbar^2} - \tfrac{K^2}{4} - q^2) - u) \\ \delta(\tfrac{1}{Kq}(\tfrac{2m\epsilon_4}{\hbar^2} - \tfrac{K^2}{4} - q^2) + u). \end{aligned} \tag{4.21}$$

where $u(u')$ is the cosine of the angle between $\mathbf{K}$ and $\mathbf{q}(\mathbf{q}')$. Performing the integrals over u and u' results in a product of Heaviside Step functions, setting integration boundaries for q and q'. Subsequently, we can perform the integration over the orientation of $\mathbf{K}$. This gives

$$\begin{aligned} &(2\pi)^2(4\pi)\left(\frac{2m}{\hbar^2}\right)^4 \int dK \frac{1}{K^4}\int_{u_1}^{u_2} dq \int_{u_1'}^{u_2'} dq' |\Gamma_{\mathbf{K},\mathbf{q},\mathbf{q}'}|^2, \\ &u_1 = \max\left(\max\left(\frac{K}{2} - \frac{2m\epsilon_1}{\hbar}, -\frac{K}{2} + \frac{2m\epsilon_1}{\hbar}\right), \max\left(\frac{K}{2} - \frac{2m\epsilon_2}{\hbar}, -\frac{K}{2} + \frac{2m\epsilon_2}{\hbar}\right)\right), \\ &u_2 = \frac{K}{2} + \frac{\sqrt{2m}}{\hbar}\min\left(\sqrt{\epsilon_1}, \sqrt{\epsilon_2}\right), \\ &u_1' = \max\left(\max\left(\frac{K}{2} - \frac{2m\epsilon_3}{\hbar}, -\frac{K}{2} + \frac{2m\epsilon_3}{\hbar}\right), \max\left(\frac{K}{2} - \frac{2m\epsilon_4}{\hbar}, -\frac{K}{2} + \frac{2m\epsilon_4}{\hbar}\right)\right), \\ &u_2' = \frac{K}{2} + \frac{\sqrt{2m}}{\hbar}\min\left(\sqrt{\epsilon_3}, \sqrt{\epsilon_4}\right). \end{aligned} \tag{4.22}$$

From this point, the Boltzmann equation can be solved numerically. We leave this as a starting point for future work.

Chapter 5

Conclusions and outlook

We studied the behavior of the magnon distribution for a dc-pumped thin film magnetic insulator. First of all we found different regimes that characterize our setup. For a film of thickness $d \approx 10^{-6}$m, Gilbert damping and spin pumping are equal in strength. In insulators of this dimension or smaller, a quasi-equilibrium magnon distribution can be realized. For large energies, the Boltzmann equation was solved analytically. The results confirm the existence of a nonzero magnon chemical potential, converging to an equilibrium value. Finally, the magnon-magnon interaction was studied. We provided a derivation of its contribution to the Boltzmann equation. In time-relaxation approximation, the interaction was found to behave as ϵ^4, dominating other processes in the high energy regime.

The analysis provided in this thesis focuses on higher energies only. This is due to the fact that the derivation of the explicit form of the magnon-magnon interaction is still incomplete. When complete, this result would learn us more about the nature of the magnon equilibration. Future work could focus on continuing with the derivation where we left off. Using the result, low energy behavior could be studied, giving us more insight in Bose-Einstein condensation of magnons. This is crucial for a broader understanding of magnon dynamics.

We have limited ourselves to linear response. This enables us to approach the problem analytically, but keeps us from capturing the whole physical picture. Numerical evaluation could deal with the full Boltzmann equation, and could be used to check the validity of our approximation.

In this research, magnon-phonon scattering was not accounted for. Future research could improve on our work by deriving the contribution of this process to the Boltzmann equation in the same manner as magnon-magnon scattering was treated in this thesis. By adding this interaction, a more complete picture of magnon kinematics could be acquired.

Appendix A

Derivation of $\mathcal{H}_4$

We consider a Heisenberg model with exchange interactions, with spins of magnitude S on a cubic lattice with constant a. The Hamiltonian has the following form:

$$\mathcal{H} = -h\sum_{\mathbf{m}} S^z_{\mathbf{m}} - \frac{1}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}} S_{\mathbf{m}} \cdot S_{\mathbf{n}}. \tag{A.1}$$

We use the Holstein-Primakoff transformation to rewrite the spin operators:

$$\begin{aligned} S^+ &= \sqrt{2S}\sqrt{1-\frac{b^\dagger b}{2S}}\,b \approx \sqrt{2S}\left(b - \frac{b^\dagger b b}{4S} + O(b^5)\right), \\ S^- &= \sqrt{2S}b^\dagger\sqrt{1-\frac{b^\dagger b}{2S}} \approx \sqrt{2S}\left(b^\dagger - \frac{b^\dagger b^\dagger b}{4S} + O(b^5)\right), \\ S^z &= S - b^\dagger b, \end{aligned} \tag{A.2}$$

with $S^+ = S^x + iS^y$ and $S^- = S^x - iS^y$. Filling this in, we get:

$$\begin{aligned} \mathcal{H} &= -h\sum_{\mathbf{m}}(S - b^\dagger_{\mathbf{m}} b_{\mathbf{m}}) - \frac{1}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}[S^x_{\mathbf{m}}S^x_{\mathbf{n}} + S^y_{\mathbf{m}}S^y_{\mathbf{n}} + S^z_{\mathbf{m}}S^z_{\mathbf{n}}] \\ &= -h\sum_{\mathbf{m}}(S - b^\dagger_{\mathbf{m}} b_{\mathbf{m}}) - \frac{1}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\Big[\left(\tfrac{S^+ + S^-}{2}\right)_{\mathbf{m}}\left(\tfrac{S^+ + S^-}{2}\right)_{\mathbf{n}} \\ &\qquad + \left(\tfrac{S^+ - S^-}{2i}\right)_{\mathbf{m}}\left(\tfrac{S^+ - S^-}{2i}\right)_{\mathbf{m}} + (S - b^\dagger b)_{\mathbf{m}}(S - b^\dagger b)_{\mathbf{n}}\Big] \\ &= -h\sum_{\mathbf{m}}(S - b^\dagger_{\mathbf{m}} b_{\mathbf{m}}) - \frac{1}{8}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\big[(S^+ + S^-)_{\mathbf{m}}(S^+ + S^-)_{\mathbf{n}} \\ &\qquad - (S^+ - S^-)_{\mathbf{m}}(S^+ - S^-)_{\mathbf{n}} \\ &\qquad + 4(S^2 - S(b^\dagger_{\mathbf{m}} b_{\mathbf{m}} + b^\dagger_{\mathbf{n}} b_{\mathbf{n}}) + b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}})\big] \\ &= -h\sum_{\mathbf{m}}(S - b^\dagger_{\mathbf{m}} b_{\mathbf{m}}) - \frac{1}{8}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\big[2S^+_{\mathbf{m}}S^-_{\mathbf{n}} + 2S^-_{\mathbf{m}}S^+_{\mathbf{n}} \\ &\qquad + 4(S^2 - S(b^\dagger_{\mathbf{m}} b_{\mathbf{m}} + b^\dagger_{\mathbf{n}} b_{\mathbf{n}}) + b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}})\big] \end{aligned} \tag{A.3}$$

$$\begin{aligned} S^+_{\mathbf{m}}S^-_{\mathbf{n}} &= 2S\left(b_{\mathbf{m}}b^\dagger_{\mathbf{n}} - \frac{b_{\mathbf{m}}b^\dagger_{\mathbf{n}}b^\dagger_{\mathbf{n}}b_{\mathbf{n}}}{4S} - \frac{b^\dagger_{\mathbf{m}}b_{\mathbf{m}}b_{\mathbf{m}}b^\dagger_{\mathbf{n}}}{4S}\right), \\ S^-_{\mathbf{m}}S^+_{\mathbf{n}} &= 2S\left(b^\dagger_{\mathbf{m}}b_{\mathbf{n}} - \frac{b^\dagger_{\mathbf{m}}b^\dagger_{\mathbf{n}}b_{\mathbf{n}}b_{\mathbf{n}}}{4S} - \frac{b^\dagger_{\mathbf{m}}b^\dagger_{\mathbf{m}}b_{\mathbf{m}}b_{\mathbf{n}}}{4S}\right). \end{aligned}$$

Splitting the terms of different orders, we obtain:

$$\begin{aligned}
\mathcal{H}_0 &= -hS\sum_{\mathbf{m}} - \frac{S^2}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}} = -hSN - \tfrac{1}{2}S^2 J_0 N, \\
\mathcal{H}_2 &= h\sum_{\mathbf{m}} b^\dagger_{\mathbf{m}} b_{\mathbf{m}} - \frac{S}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left[b_{\mathbf{m}} b^\dagger_{\mathbf{n}} + b^\dagger_{\mathbf{m}} b_{\mathbf{n}} - b^\dagger_{\mathbf{m}} b_{\mathbf{m}} - b^\dagger_{\mathbf{n}} b_{\mathbf{n}})\right] \\
&= h\sum_{\mathbf{m},\mathbf{n}} \delta_{\mathbf{m},\mathbf{n}} b^\dagger_{\mathbf{m}} b_{\mathbf{n}} - \frac{S}{2}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left[2b^\dagger_{\mathbf{m}} b_{\mathbf{n}} + \delta_{\mathbf{m},\mathbf{n}} - 2\delta_{\mathbf{m},\mathbf{n}} b^\dagger_{\mathbf{m}} b_{\mathbf{n}}\right],
\end{aligned} \tag{A.4}$$

which is equal to the following expression if we drop the 0^{th} order term:

$$\begin{aligned}
\mathcal{H}_2 &= h\sum_{\mathbf{m},\mathbf{n}} \delta_{\mathbf{m},\mathbf{n}} b^\dagger_{\mathbf{m}} b_{\mathbf{n}} - S\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}(1-\delta_{\mathbf{m},\mathbf{n}}) b^\dagger_{\mathbf{m}} b_{\mathbf{n}}, \\
\mathcal{H}_4 &= -\frac{1}{8}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left[-b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} - b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} - b^\dagger_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} b_{\mathbf{n}} - b^\dagger_{\mathbf{m}} b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b_{\mathbf{n}} + 4 b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}}\right] \\
&= \frac{1}{8}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left(2b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} + 2b^\dagger_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} b_{\mathbf{n}} - 4b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} + [b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}}, b_{\mathbf{m}}] + [b^\dagger_{\mathbf{n}} b_{\mathbf{n}} b_{\mathbf{n}}, b^\dagger_{\mathbf{m}}]\right) \\
&= \frac{1}{4}\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}\left(b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} + b^\dagger_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} b_{\mathbf{n}} - 2b^\dagger_{\mathbf{m}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}}\right),
\end{aligned} \tag{A.5}$$

where the last two 2^{nd} cancel. There are no first order or third order contributions to the Hamiltonian.

Next, we will Fourier transform the expressions for $\mathcal{H}_2$ and $\mathcal{H}_4$ using $b_{\mathbf{m}} = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}} b_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_{\mathbf{m}}}$, with $\mathbf{k}$ the magnon wavevector and N the number of lattice sites. We transform the different terms one by one:

$$\begin{aligned}
\sum_{\mathbf{m},\mathbf{n}} \delta_{\mathbf{m},\mathbf{n}} b^\dagger_{\mathbf{m}} b_{\mathbf{n}} &= \sum_{\mathbf{m}} b^\dagger_{\mathbf{m}} b_{\mathbf{m}} \\
&= \frac{1}{N}\sum_{\mathbf{m}}\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} e^{-i\mathbf{k}\cdot\mathbf{m}a} e^{i\mathbf{k}'\cdot\mathbf{m}a} \\
&= \frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} \sum_{\mathbf{m}} e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{m}a} \\
&= \sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} \delta_{\mathbf{k}',\mathbf{k}} \\
&= \sum_{\mathbf{k}} b^\dagger_{\mathbf{k}} b_{\mathbf{k}}.
\end{aligned} \tag{A.6}$$

$$\begin{aligned}
\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}}(1-\delta_{\mathbf{m},\mathbf{n}}) b^\dagger_{\mathbf{m}} b_{\mathbf{n}} &= J_0 \sum_{\mathbf{m}}\sum_{\alpha} b^\dagger_{\mathbf{m}}(b_{\mathbf{m}+\alpha} + b_{\mathbf{m}-\alpha}) \\
&= \frac{J_0}{N}\sum_{\mathbf{m}}\sum_{\alpha}\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} e^{-i\mathbf{k}\cdot\mathbf{m}a}(e^{i\mathbf{k}'\cdot(\mathbf{m}+\alpha)a} + e^{i\mathbf{k}'\cdot(\mathbf{m}-\alpha)a}) \\
&= \frac{J_0}{N}\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} \sum_{\mathbf{m}} e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{m}a} \sum_{\alpha}(e^{i\mathbf{k}'\cdot\alpha a} + e^{-i\mathbf{k}'\cdot\alpha a}) \\
&= J_0 \sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b_{\mathbf{k}'} \delta_{\mathbf{k},\mathbf{k}'} \sum_{\alpha} 2\cos(a\alpha\cdot\mathbf{k}') \\
&= 2J_0 \sum_{\mathbf{k}} b^\dagger_{\mathbf{k}} b_{\mathbf{k}} \sum_{\alpha} \cos(a\alpha\cdot\mathbf{k}).
\end{aligned} \tag{A.7}$$

$$
\begin{aligned}
\sum_{\mathbf{m},\mathbf{n}} J_{\mathbf{mn}} b_{\mathbf{m}} b^\dagger_{\mathbf{n}} b^\dagger_{\mathbf{n}} b_{\mathbf{n}} &= J_0 \sum_{\mathbf{m}} \sum_{\alpha} b_{\mathbf{m}} (b^\dagger_{\mathbf{m}+\alpha} b^\dagger_{\mathbf{m}+\alpha} b_{\mathbf{m}+\alpha} + b^\dagger_{\mathbf{m}-\alpha} b^\dagger_{\mathbf{m}-\alpha} b_{\mathbf{m}-\alpha}) \\
&= \frac{J_0}{N^2} \sum_{\mathbf{m}} \sum_{\alpha} \sum_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} b_{\mathbf{k1}} b^\dagger_{\mathbf{k2}} b^\dagger_{\mathbf{k3}} b_{\mathbf{k4}} e^{i\mathbf{k1}\cdot\mathbf{m}a} \\
&\qquad \times (e^{-i\mathbf{k2}\cdot(\mathbf{m}+\alpha)a} e^{-i\mathbf{k3}\cdot(\mathbf{m}+\alpha)a} e^{i\mathbf{k4}\cdot(\mathbf{m}+\alpha)a} \\
&\qquad + e^{-i\mathbf{k2}\cdot(\mathbf{m}-\alpha)a} e^{-i\mathbf{k3}\cdot(\mathbf{m}-\alpha)a} e^{i\mathbf{k4}\cdot(\mathbf{m}-\alpha)a}) \\
&= \frac{2J_0}{N} \sum_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} b_{\mathbf{k1}} b^\dagger_{\mathbf{k2}} b^\dagger_{\mathbf{k3}} b_{\mathbf{k4}} \delta_{\mathbf{k1}+\mathbf{k2}-\mathbf{k3}-\mathbf{k4},0} \\
&\qquad \times \sum_{\alpha} \cos(a\alpha \cdot (\mathbf{k}4 - \mathbf{k}2 - \mathbf{k}3)).
\end{aligned}
\tag{A.8}
$$

We find the following expressions:

$$
\begin{aligned}
\mathcal{H}_2 &= \sum_{\mathbf{k}} \left(h - 2SJ_0 \sum_{\alpha} \cos(a\alpha \cdot \mathbf{k}) \right) b^\dagger_{\mathbf{k}} b_{\mathbf{k}}, \alpha = \{\hat{x}, \hat{y}, \hat{z}\}, \\
\mathcal{H}_4 &= \frac{1}{2N} \sum_{\mathbf{k1},\mathbf{k2},\mathbf{k3},\mathbf{k4}} b^\dagger_{\mathbf{k1}} b^\dagger_{\mathbf{k2}} b_{\mathbf{k3}} b_{\mathbf{k4}} \delta(\mathbf{k}_3 + \mathbf{k}_4 - \mathbf{k}_1 - \mathbf{k}_2) \\
&\qquad \times \sum_{\alpha} (\cos(a\alpha \cdot \mathbf{k}_1) + \cos(a\alpha \cdot \mathbf{k}_4) - 2\cos(a\alpha \cdot (\mathbf{k}_4 - \mathbf{k}_2))) \\
&\quad + \frac{1}{2N} \sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}} b^\dagger_{\mathbf{k}} 2 \sum_{\alpha} (\cos(a\alpha \cdot \mathbf{k}') - \cos(a\alpha \cdot (\mathbf{k} - \mathbf{k}'))) \,,
\end{aligned}
\tag{A.9}
$$

in which the second line of $\mathcal{H}_4$ represents corrections to the second order term of the hamiltonian, which arise due to normal ordering of the annihilition and creation operators.

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