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Effect of Spins on Some Phenomena in Magnetic Materials

A thesis Submitted

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Abstract

The role played by magnon-phonon interactions in two important phenomena in magnetic materials has been investigated.

The first phenomenon is thermal conduction in antiferromagnetic materials. Thus the first part of the thesis is concerned with performing a theoretical study of thermal conductivity in antiferromagnets. The study has the advantage that the three phonon interactions as well as the magnon phonon interactions have been represented by model operators that preserve the important properties of the exact collision operators. A new expression for thermal conductivity has been derived that involves the same terms obtained in our previous work in addition to two new terms. These two terms represent the conservation and quasi-conservation of wavevector that occur in the three-phonon Normal and Umklapp processes respectively. They gave appreciable contributions to the thermal conductivity and have led to an excellent quantitative agreement with the experimental measurements of the antiferromagnet $FeCl_2$.

The second phenomenon is magnon squeezing. Very important results concerning the time evolution operator and the time dependence of the operators involved that arise due to magnon-phonon interactions in antiferromagnets have been derived. However, the study of this phenomenon has been carried out mainly in the second part of the thesis in ferromagnetic materials. The squeezed states of dressed magnons in ferromagnets have been investigated. No effective Debye cutoff frequency has been assumed unlike what has been done hitherto. Instead, the results have been expressed throughout in terms of the reduced temperature. The effect of dressed magnon-phonon interactions on the formulation of these states has been studied. It has been shown that the magnon-phonon interactions play a significant role in determining the squeeze factor and the variation of the dressed magnon effective mass with temperature.

The main results of Chapters 2 and 4 have been published in

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Chapter 1

Introduction

1.1 Review of Magnon-Phonon Interactions

Spins play an important role in many physical phenomena in magnetic materials, such as determining a variety of magnetic and thermal properties and affecting the critical parameters and phase transitions in these materials. Magnons are the quanta of energy of spin waves in magnetic materials. Their effect appears mainly through their interactions with magnetic defects, electrons and phonons. Thus the magnon-phonon interactions present one of the essential types of interactions that affect substantially the properties of magnetic materials. The thesis is devoted to study two important phenomena in which the magnon-phonon interactions give an important contribution.

The first phenomenon is heat conduction in magnetic materials. For paramagnetic and ferromagnetic materials the situation is simpler than antiferromagnets. Elliott and Parkinson [23] explored the effect of magnon-phonon interactions on the thermal conductivity of Paramagnetic materials. Also Walton *et al* [78] explained the magnetic field dependence of their thermal conductivity measurements of the ferromagnet yttrium iron garnet (YIG) at low temperatures ($T=0.23-1$ K) by taking into account both magnon and phonon contributions to the

thermal current and considering a resonant one magnon one phonon interaction. Jensen and Hourmann [39] investigated further the role of the magnon-phonon interactions in the ferromagnet terbium. Also, considerable effort has been devoted to study the magnon-phonon interactions in ferromagnets by Wesselinowa [82], [83] and Wesselinowa and Apostolov [84]. Their results were extended more recently to cover the cases of ferromagnetic thin films and nanoparticles (Wesselinowa [85] and Wesselinowa and Apostolova [86]). Moreover, in last years great attention has been given to study the effect of magnon-phonon interactions on some important phenomena in ferromagnets [11-15], [87], [41], [79], [37], [63].

In antiferromagnetic materials the situation is much more complicated. Slack and Newman [69] and Slack [70] considered the effect of magnon-phonon interactions using the relaxation time approximation. They introduced a simple expression for the relaxation time that depends on the magnetic order parameter. Mikhail [52] employed the expression obtained in Refs. [69], [70] for the magnon-phonon interactions relaxation time. He succeeded to fit the measurements of Morelli *et al* [54] on single crystal $La_2CuO_{4+\Delta}$ along the direction [001] and produced the sharp minimum anomaly that occurs at the Néel temperature. The magnon-phonon interactions were further investigated in La_2CuO_4 and related cuprates by Kochelaev [45]. Similar anomalies were detected in the measurements of thermal conductivity of other antiferromagnetic materials such as $FeCl_2$ (Laurence and Petitgrand [46] and Tiwari and Ram [76]) and $UNi_{0.5}Sb_2$ (Mucha *et al* [55]). Mikhail *et al* [53] re-stressed the importance of magnon-phonon interactions that were used to give a reasonable explanation of the anomalies displayed in the thermal conductivity measurements of Ref. [46].

Also, Gustafson and Walker [28] measured the thermal conductivity of the antiferromagnets $RbMnF_3$ and MnF_2 in the presence and absence of magnetic fields. They attributed the appreciable dependence of their measurements on the strength and direction of the field due to the coupling between spins and phonons. More recently Sales *et al* [64] reported a strong dependence of the thermal conductivity and heat capacity of the antiferromagnet $K_2V_3O_8$ on the applied magnetic

field. Moreover, measurements have been performed (Oleaga *et al* [56]) on the thermal conductivity, specific heat and thermal diffusivity to study the critical behaviour of the antiferromagnetic transition in $KCoF_3$ and $KNiF_3$. We believe that the magnon-phonon interactions should play an appreciable role in investigating their measurements on thermal conductivity.

The importance of magnon-phonon interactions in antiferromagnets has been investigated recently in some fields other than thermal conduction. In this connection, Malard and Pires [49] has extended the results of Pires [63] on magnon-phonon interactions by using memory functions and perturbation expansions to be applied to the case of one dimensional antiferromagnets. Also, Khadikova *et al* [40] have investigated the magnon-phonon interaction in the one dimensional Heisenberg spin-ladder antiferromagnet $La_5Ca_9Cu_{24}O_{41}$. For this purpose they performed the dynamical fluorescent micro-thermal imaging (FMI) experiment and modelled the dynamic heat transport experiment using a two temperature model approach. They took both the crystal as well as the polymer fluorescent heat imaging layer into account.

In view of the importance of magnon-phonon interactions in antiferromagnets several theoretical studies were performed to explore the effect of these interactions on the calculation of thermal conductivity by Gluck [26], Dixon [20], Dixon *et al* [21] and Mikhail *et al* [53]. Dixon [20] developed a model of two magnon-one phonon interactions in antiferromagnetic materials. He obtained two types of processes, the first is the conversion processes (C-processes) in which two magnons are created (or annihilated) and a phonon is destroyed (or created) while the second type is the radiation processes (R-processes) in which a magnon is created (or annihilated) and a magnon and a phonon are destroyed (or created). They represented these types of interactions by relaxation times in the phonon scattering mechanism while the contribution of magnons to the heat current was neglected. A very recent theoretical investigation has been performed on the thermal conductivity of local moment models with strong spin-orbit coupling by Stamokostas *et al* [74].

Mikhail *et al* [53] performed a detailed study of thermal conduction in antiferromagnets where the contributions of magnons and phonons to the thermal scattering and thermal current were taken into account. They started by the two transport Boltzmann equations of phonons and magnons. The magnon-phonon interactions were described in detail in terms of the two magnon-one phonon C and R processes introduced in Dixon [20] and Dixon *et al* [21]. Both Normal and Umklapp processes were taken into consideration. The resulting collision operator of magnon-phonon interactions has consequently been replaced for Normal and Umklapp processes by a model operator which possesses the same important properties as the exact operator. The other scattering processes were treated as resistive processes by using the relaxation time approximation. This technique may not be suitable to deal with three-phonon interactions since it does not take into consideration the effect of the wavevector conservation and quasi-conservation that occur respectively in the Normal and Umklapp processes of this type of phonon interactions. As a result the expression for thermal conductivity derived in Mikhail *et al* [53] did not involve terms that correspond to the second term of the Callaway model [7] and to the modification considered in Mikhail [50].

The second phenomenon is magnon squeezing. In general squeezing is strongly related to the Heisenberg uncertainty principle which was postulated in 1927 [31]. Nevertheless, it still stands against any philosophy that may seem to change its concept. It simply states that two incompatible physical quantities whose quantum operators do not commute cannot be simultaneously measured. The discrepancy in the measurements should be greater than a quantity which is proportional to the modulus of their commutator bracket.

In the past three decades squeezing states have been discovered, mainly in optical systems. In these states, the discrepancy in the measurements of one of the two incompatible variables decreases below the limit allowed by the uncertainty principle on the expense that the discrepancy in the other variable increases so that the uncertainty principle remains inviolate.

The squeezing states have first been investigated for photons in

models which deal with the interaction of an atom of at least two energy levels with an electromagnetic field in a cavity (Jaynes Cummings model [38]). The photon squeezed states have been demonstrated later in Condensed matter by Slusher *et al* [71] and Artoni and Birman [1]. The discovery of squeezing states has been accompanied by the reduction of noise in photon propagation processes (Walls and Milburn [77]) which has led to new branches of quantum optical technology namely quantum information, communication and computation (Refs. [48], [19], [22], [72], [66], [75]).

This has also encouraged researcher to investigate squeezing states in systems of other bosonic quasi-particles. The squeezed states of phonons has been explored by Hu and Nori [33-36], Levi [47] and Garrett *et al* [25]. Also, the case of polaritons has been discussed in [2, 3]. Moreover, in antiferromagnets magnons can generate self-squeezing states due to the interference between the spin waves that results from the two distinct sublattices. These states have been investigated by Peng [58], [60], [62], Peng and Hao [61].

In ferromagnets, the situation is much more complicated in view of the presence of one lattice. However, the consideration of the fourth order terms in the Heisenberg Hamiltonian gave rise to possible interactions between two magnons and accordingly to the creation of a new type of magnons which are called dressed magnons. The squeezed states of dressed magnons have been studied by Peng [59], Wang *et al* [81], Wang and Cheng [80], Cheng *et al* [16] and Cheng [18].

1.2 Outline of the Present Work

The present work is concerned with studying the magnon-phonon interactions in antiferro- and ferro- magnets. It's motivation is two folds. The first is to extend the results of Mikhail *et al* [53] to deal with three-phonon interactions in a more accurate manner. In Ref. [53] these interactions have been treated by considering a relaxation time technique. In the present work they will be taken into consideration by using an approach in which the conservation conditions are

preserved. Alternative techniques were used by Hamilton [30], Simons [67], [68], Srivastave [73] and Mikhail and Madkour [51]. Simons [68] introduced a model operator which has the advantage that it possesses the same important properties as the exact collision operator. It satisfies the conservation of the wavevector in the three-phonon Normal processes and accordingly gives rise to the second term of the Callaway expression for thermal conductivity [7]. It was also applied in Mikhail [50] to produce an adequate modification for the thermal conductivity Callaway expression that represents the quasi-conservation of the wavevector in the three-phonon Umklapp processes. Mikhail and Madkour [51] constructed a model operator that was mainly based on the isotropic approach introduced in Parrott [57] and Hamilton and Parrott [29]. It possesses the same essential properties as the Simons Model [68]. It was also used to derive the modification that occurs in the thermal conductivity expression due to the three-phonon Umklapp processes quasi-conservation of wavevector. The relaxation times as well as the related quantities were calculated from their complicated exact expressions that depend on the third order elastic constants.

Here, the Simons model [68] will be utilized to represent the three-phonon Normal and Umklapp processes since the operators that were used in Mikhail *et al* [53] to represent the magnon-phonon interactions were formulated in a similar manner. The use of the isotropic model of Mikhail and Madkour [51] has been postponed to be considered in a future article. The expression derived for thermal conductivity involves the same terms obtained in Mikhail *et al* [53] that represent the contributions of magnons and phonons and the conservation conditions of magnon-phonon interactions, in addition to the required new two terms. The first of these terms stand for the second term in the Callaway expression for thermal conductivity [7] while the second represents the modification that results due to the consideration of the wavevector quasi-conservation in the three-phonon Umklapp processes. It resembles the modification terms that were derived in Mikhail [50] and Mikhail and Madkour [51].

The second motivation for the work is to investigate the effect of magnon-phonon interactions on the presence of squeezed states of the

dressed magnons in ferromagnets. The same technique used in Ref. [53] has been applied to study magnon-phonon interactions in ferromagnets. It has, then, been shown that the effect of these interactions on the squeezed states of dressed magnons becomes significant if the basic parameters involved in the calculations are suitably chosen.

The thesis is arranged in the following way. In Chapter 2 the model calculation of thermal conductivity in antiferromagnets is given. The Hamiltonian operator that represents magnon-phonon interactions has been derived in Section 2.1. The study gives rise to the two types of processes, namely the conversion (C) and radiation (R) processes that were originally introduced in Dixon [20]. The Boltzmann equations of magnons and phonons have been constructed in Section 2.2 so that the magnon-phonon C and R processes were taken into consideration. In Section 2.2.1 the model operator technique was utilized to deal with both Normal and Umklapp three phonon interactions. The solution of the phonon linearized Boltzmann equation was accordingly derived. Section 2.2.2 is devoted to solve the magnon linearized Boltzmann equation by using the model operators introduced in Mikhail *et al* [53]. The corresponding new expression for thermal conductivity of antiferromagnetic materials has been deduced in Section 2.3. The application of the results to calculate the thermal conductivity of the antiferromagnet $FeCl_2$ has been performed in Section 2.4. The comparison of the results with the experimental measurements of Laurence and Petitgrand [46] has been displayed and the contributions due to the two new terms have been presented and discussed.

Finally, the effect of R-processes has been studied in a more rigorous way in Section 2.5. It has been confirmed analytically that these processes can be neglected without large errors. In previous studies they have been ignored due to the reason that the two magnons in a process of this type must belong to the same branch and thus the probability of its occurrence is very small.

Chapter 3 is concerned with deriving a suitable expression for the time evolution operator that results due to magnon-phonon interactions in antiferromagnets. This operator has then been utilized to determine the time dependence of the two operators $\hat{b}_{1\mathbf{k}}$, $\hat{b}_{2\mathbf{k}}$ that were