Spin-Phonon Interactions in Magnetic Materials

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Abstract

A theoretical study is given of spin (magnon)-phonon interactions in antiferromagnetic materials. A perturbation technique has been applied in which the linear terms of the ions displacements from equilibrium have been retained in the Heisenberg operator. The procedure gave rise to two types of two magnon-one phonon interactions, namely conversion (C) and radiation (R) processes. Both Normal and Umklapp magnon-phonon interactions have been taken into account. The latter was not considered in any earlier treatment. The transition probabilities of these interactions have been derived by applying a modified approach that differs from those used hitherto.

The roles of magnons and phonons as heat carriers and as sources of thermal resistance have been taken into consideration. A detailed study of thermal conduction in antiferromagnets has been undertaken. The calculations are based on the two transport Boltzmann equations of phonons and magnons. The scattering mechanism in the two equations is taken to be due to the two types (C and R) of the two magnon-one phonon interactions.

The equivalent Boltzmann equation that results due to the combination of the resulting two linearized Boltzmann equations has been obtained. It has been shown that the exact magnon-phonon collision operator involved in this equation is linear, Hermitian and negative semi-definite. Its effect on the energy and wavevector for both Normal and Umklapp processes has been inferred. The exact operator was then replaced by a model operator which possesses the same important properties. The effect of other scattering processes that either phonons or magnons can undergo in antiferromagnets has been taken into consideration by using the relaxation time approximation. The thermal current due to both magnons and phonons has been calculated. The applied approach has yielded a new expression for thermal conductivity that includes terms which represent the contributions of magnons and phonons. It further includes terms that represent the

effect of both Normal and Umklapp magnon-phonon interactions. The latter type of terms is analogous to the terms obtained in earlier treatments on three-phonon interactions.

The expression obtained for the thermal conductivity has been utilized to fit the experimental data of the antiferromagnet $FeCl_2$. The parameters involved in the relaxation times have been chosen to give the best fit to the experimental results. The theoretical results obtained by using the new expression showed a good quantitative agreement with the experimental measurements and have reasonably described the dip in the thermal conductivity curve.

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1.1 Review of the Spin (Magnon)-Phonon Interactions

Spin-phonon interactions play an essential role in many of the phenomena that occur in magnetic materials. They determine a variety of magnetic and thermal properties and affect the critical parameters and phase transitions in these materials. Phonons are the quanta of energy of lattice vibrations while the quanta of energy of spin waves in magnetic materials are magnons. The terminologies of spin-phonon interactions and magnon-phonon interactions are thus equivalent. In the present work we are mainly concerned with the effect of this type of interactions on thermal conduction.

The main types of heat carriers in solids are phonons, electrons and magnons. Their contributions to the thermal current differ from one material to another and depend strongly on the temperature range under consideration and on the applied external fields. Also, the thermal scattering mechanism arises due to the interactions between these types of carriers and the other interactions in which they are involved. The roles played by phonons and electrons were discussed in many context and the results seem to be most likely settled (Ziman [72], Smith *et al* [61], Parrott and Stuckes [51], Berman [3] and Brüesch

[5]). On the other hand, magnons still need further investigations and speculations. Their thermal transport and scattering properties are governed by the different interactions which they can undergo in these materials. The interaction of magnons with magnetic defects was considered by Callaway [7] and by Callaway and Boyd [8]. Also, the spin scattering by dislocations in III-V semiconductors was studied by Jena [30] and with long range impurities in semiconductor heterostructures by Bleibaum [4]. The influence of electron-magnon interactions on the properties of ferromagnetic materials was investigated by Nolting and Nolting [50]. For this purpose they employed a Green function method. Metcalfe and Rosenberg [43] , [44] studied the magnetothermal resistivity of antiferromagnetic crystals at low temperatures ($T<4K$). They also considered the effect of an applied magnetic field on their results. They calculated both the contributions from phonons and magnons by considering only boundary scattering which can be represented by a constant mean free path. However, as regards thermal conduction the most important type of interactions in which magnons are involved is the magnon-phonon interaction. The effect of spin-phonon coupling on the specific heat (Sears [56]) and on the thermal conductivity (Elliott and Parkinson [22]) of paramagnetic materials was explored by using a Green function approach. As regards ferromagnets Walton et al [64] explained the magnetic field dependence of their thermal conductivity measurements of 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 [31] investigated further the role of the interactions between magnons and acoustic and optical transverse phonons in the ferromagnet terbium. Their results were then used to interpret the experimental measurements of inelastic neutron scattering in this material. Also, considerable effort has been devoted to study the magnon-phonon interaction by Wesselinowa [66] , [67] and Wesselinowa and Apostolov [68]. The Green function techniques and the random phase approximation were mainly used in these works. Recently, Wesselinowa [69] and Wesselinowa and Apostolova [70] extended the

study to cover the cases of ferromagnetic thin films and ferromagnetic nanoparticles.

In recent years great attention has been given to study the effect of magnon-phonon interactions on some important phenomena. Cheng et al [12] , [13] developed the magnon-phonon interaction on the basis of a two-dimensional Heisenberg ferromagnetic system and by using the Green function theories. They found that this interaction plays a significant role for the softening of the transverse acoustic phonon dispersion branch and for the damping of both transverse and longitudinal acoustic phonons. Furthermore, they have shown [14], [11], [15] that the interaction leads to magnon softening and magnon damping. The latter is mainly caused due to the interactions of magnons with optical phonons. Moreover, the calculations of the magnon and phonon damping and the softening due to magnon-phonon interactions were considered by Woods [71] while the softening in the magnon dispersion relation due to this type of interactions has been pointed out by Kirby et al [32]. Also, Wan [65] interpreted the peak anomaly which occurs in the variation of the specific heat of the ferromagnetic Ni_2MnGa alloy with temperature to be due to the one magnon-one transverse acoustic phonon interactions. The effects of spin-phonon interactions on the transport and magnetic properties of $R_{1-x}A_xMnO_3$ (R is a rare earth ion) as well as their effects on the role of oxygen isotopes and on the ferromagnetic transition temperature were explored by Huang et al [29]. The investigation was performed by using a perturbation technique and the mean field theory. Pires [52] studied the magnon-phonon interaction in the one dimensional quantum ferromagnetic XY model by using two different approaches, a low temperature approximation for the memory function and the perturbation expansion in the Green function formalism. The results have been extended in Malard and Pires [41] to be applied to the case of one dimensional antiferromagnets.

The situation for antiferromagnetic materials is much more complicated than ferromagnets as in the most simplest case the spins are divided into two sublattices. Slack and Newman [59] and Slack [60] included the effect of magnon-phonon interactions in their calculations of the thermal conductivity of antiferromagnetic materials by using a relaxation time which depends on the magnetic order parameter. In spite of the simplicity of their expression it was used successfully by Mikhail [47] to fit the thermal conductivity measurements of Morelli et al [48] on single-crystal $La_2CuO_{4+\Delta}$ along the direction [001], in particular the sharp minimum anomaly which occurs at the Néel temperature. The role of the magnon-phonon interaction in La_2CuO_4 and related cuprates was also considered by Kochelaev [36]. The same type of anomalous behavior was also observed in the thermal conductivity measurements of other antiferromagnetic materials such as $FeCl_2$ (Laurence and Petitgrand [38] and Tiwari and Ram [63]) and $UNi_{0.5}Sb_2$ (Mucha et al [49]). This, in turn, emphasizes the importance of magnon-phonon interactions in explaining these anomalies. Also, Gustafson and Walker [26] measured the thermal conductivity of the antiferromagnets $RbMnF_3$ and MnF_2. Their results in zero magnetic field showed no evidence for magnon conductivity. In the presence of a magnetic field, however, the measurements exhibited an appreciable dependence on the strength and direction of the field. They attributed this dependence to be due to the coupling between spins and phonons. More recently Sales et al [55] reported a strong dependence of the thermal conductivity and heat capacity of the antiferromagnet $K_2V_3O_8$ on the applied magnetic field. They suggested that the production of a new magnetic ground state may be responsible for this dependence. The effect of the magnon-phonon interactions on other phenomena in antiferromagnetic materials has also been investigated. Lovesey [39], [40] studied the effect of the magnon-phonon interaction on inelastic neutron scattering in FeF_2. Choi et al [16] explored the role of spin-phonon coupling in the Raman-scattering measurements on $Sr_{1-x}Ba_xCu_2(Bo_3)_2$. Moreover, the quantum magnetostriction oscillations in a two-dimensional antiferromagnet with spin-phonon interaction in a magnetic field were studied by Aplesnin [1]. For such purpose he applied the Monte Carlo method in the nonadiabatic approximation. Also, the influence of the magnon-phonon interactions on the infrared measurements in a variety of antiferromagnetic materials has been investigated by different groups of researchers

(Rudolf et al [53], [54], Djokic et al [19] and Hsu et al [28]). Special care has further been given to investigate the magnon-phonon coupling mechanism in layered antiferromagnetic superconductors (Sugai et al [62] , Knoll et al [35]).

Theoretical studies of magnon-phonon interactions in antiferomagnets were given by Gluck [24] and Dixon [17]. Dixon [17] and Dixon et al [18] developed a model of two magnon-one phonon interactions in antiferromagnetic materials. The Hamiltonian operator used in the calculations consists of four parts that represent the isotropic exchange, the Ising like anisotropic exchange, the uniaxial single-ion anisotropy and Zeeman interaction with the applied magnetic field. He obtained two types of processes, the first is the conversion processes (C-processes) in which two magnons are created (or annihilated) on the expense of destroying (or creating) a phonon while the second is the radiation processes (R-processes) in which a magnon is created (or annihilated) on the expense of destroying (or creating) a magnon and a phonon. These types of processes were then represented by relaxation times in the phonon scattering mechanism. The contribution of magnons to the heat current was accordingly neglected.

1.2 Outline of the Present Work

The present work is concerned with studying the magnon-phonon interactions in antiferromagnetic materials and with deriving the corresponding expression for the thermal conductivity. It was felt that a detailed calculation should be undertaken in which the contributions of magnons and phonons to the thermal current as well as to the thermal scattering mechanism are taken into consideration. The starting point of the calculations is the two transport Boltzmann equations of phonons and magnons. The scattering mechanism in the two equations due to the magnon-phonon interactions is described in detail in terms of the two magnon-one phonon, C and R processes introduced by Dixon [17] and Dixon et al [18]. Both Normal and Umklapp processes are taken into account. The latter was not considered in any other previous work on magnon-phonon interactions. The effect of the other scattering processes has also been included by using the relaxation time approximation. The resulting two linearized Boltzmann equations have then been combined to give one equivalent equation. The equivalent 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 model operators used here are of the same type introduced by Simons [57] , [58] for three-phonon interactions. The thermal current due to both magnons and phonons has then been calculated and the corresponding expression for the thermal conductivity has been derived. The obtained expression is analogous to those obtained in Mikhail [45] and Mikhail and Madkour [46] for the phonon thermal conductivity. The application of the model to fit the experimental data of the thermal conductivity of the antiferromagnet $FeCl_2$ (Laurence and Petitgrand [38]) has shown a good quantitative agreement.

The present work is organized as follows: In chapter 2, the basic and general relations of spin operators are given in section 2.1 while the formulation of the Heisenberg Hamiltonian has been presented in section 2.2. The method applied is based on the fact that the Heisenberg Hamiltonian arises due to the exchange interactions between the spins on the different lattice sites of the system. The spin waves have then been discussed in sections 2.3 and 2.4 from the classical and quantum mechanical points of view. The spin waves in the cases of ferromagnets and antiferromagnets have consequently been studied within the frame-work of quantum mechanics in sections 2.5 and 2.6 respectively. The concept of magnons as the quanta of energy of spin waves has been introduced. An alternative approach for the case of antiferromagnets is also given in section 2.7. This approach was originally considered by Feder and Pytte [23] and Dixon [17]. In section 2.7, it has been found that the dispersion relation of magnons in antiferromagnets consists of two branches in the presence of an external magnetic field.

Chapter 3 is devoted to study the magnon-phonon interactions.

Following Dixon [17] we apply a perturbation technique in which the linear terms of the ions displacements from equilibrium have been retained in the Heisenberg operator. The procedure gives rise to the two types (C and R) of two magnon-one phonon processes. The coefficient of the R-processes given in this section has not been reported anywhere before. The transition probabilities of the different magnon-phonon interactions have been derived in section 3.2. We started by the time dependent perturbation theory and applied an approach which seems to be new and more accurate than was done hitherto. We believe that we have amended the expression used for the C-processes transition probability in Dixon [17]. Also, both Normal and Umklapp processes are considered. The Boltizmann equations of phonon and magnons have been derived in section 3.3 by considering the two magnon-one phonon interactions (C and R, Normal and Umklapp processes). The combination of the resulting two linearized Boltzmann equations in one net equation has been performed in section 3.4. The procedure has led to the definition of the exact magnon-phonon interactions collision operator. It has then been shown that this operator is linear, Hermitian and negative semi-definite. Its effect on the energy and wavevector for both Normal and Umklapp processes has been derived.

The model operator technique has been exhibited and used for the calculation of the thermal conductivity in chapter 4. Model operators for magnon-phonon Normal and Umklapp interactions have been obtained in section 4.1. According to Simons [58], the model operators are formulated in such a way that they possess the same important properties proved in section 3.4 for the exact operator. The effects of other scattering mechanisms which either phonons or magnons are involved have been included in section 4.2 by using relaxation times. It has been found that this is important for the calculation of thermal conductivity. In section 4.3 the thermal conductivity has been calculated. The model operators derived in section 4.1 together with the relaxation times given in section 4.2 have been utilized. The contributions of phonons and magnons to the thermal current have been taken into account. The expression obtained for the thermal conductivity includes terms which represent magnons and phonons and others which represent the effect of Normal and Umklapp magnon-phonon interactions. The latter types of terms resemble the analogous terms of Normal and Umklapp three-phonons interactions in the expressions for phonon thermal conductivity obtained in Mikhail [45] and Mikhail and Madkour [46].

The expression obtained in section 4.3 for the thermal conductivity has been used in chapter 5 to fit the experimental results of the antiferromagnet $FeCl_2$ (Laurence and Petitgrand [38]). Some basic relations and assumptions are given in section 5.1. Analytical expressions for the basic quantities which are involved in the magnon-phonon model operator have been derived in section 5.2. The inner products that determine the expression for thermal conductivity have been expressed in section 5.3 in terms of integrals over the magnitudes of magnons and phonons wavevectors. These integrals can be evaluated numerically. The numerical results have been performed and displayed in section 5.4. The parameters involved in the relaxation times have been treated as fitting parameters. Their values were chosen to give the best fit to the experimental results. The comparison between the theoretical and experimental results displayed in Fig. 2 showed a reasonable quantitative agreement.