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Lattice thermal conductivity of antiferromagnetic insulators

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The effect of two-magnon–one-phonon scattering processes on the lattice thermal conductivity of antiferromagnets is studied. Transition matrix elements are obtained by expansion in lattice displacements of the exchange and anisotropy for an anisotropic Heisenberg Hamiltonian. Relaxation times for the phonons are determined by conventional perturbation theory and lattice thermal conductivities are then calculated in the Debye approximation. Results are obtained in all three field-induced phases for fields parallel to the easy axis for sublattice magnetization. It is found that in the spin-flop phase, the coefficients of the transformation to magnon operators make a substantial contribution to the magnetic field dependence.

I. INTRODUCTION

The thermal conductivity of magnetic insulators has attracted the attention of many experimenters over the last twenty years.[1–23] Despite this activity there has been only moderate success in relating the observations to microscopic models of processes of heat conduction and thermal resistance. Qualitatively, it has long been understood that magnons may affect the thermal conductivity either by providing a mechanism for transport or by providing scattering processes to impede heat transfer by the phonons. In many experimental studies the analysis of the results has not proceeded beyond this qualitative stage except to suggest which magnon-phonon processes might be in important ones and to identify a contribution of the magnons to the thermal transport. It has been suggested also in both thermal-transport and ultrasonic-propagation studies that magnetic domains may provide a strong mechanism for scattering phonons.[10,11,24] Several workers have attempted to interpret the thermal conductivity in zero magnetic field by detailed microscopic models of the magnon contribution to the transport and/or magnon-phonon scattering.[14,16,17,19,20,24,33] A much more convincing case can be made for the validity of a proposed model if it accounts for the magnetic field dependence, as well as the temperature dependence, of the thermal conductivity. To our knowledge this has been achieved only for two magnetic materials, yttrium iron garnet [YIG (Ref. 14)] and MnF_2 (Refs. 22 and 23). In the first instance[14] Walton, Rives, and Khalid were able to account for both the temperature dependence and field dependence of the thermal conductivity of a YIG crystal with a model for magnon thermal conductivity and a resonant one-magnon–one-phonon interaction. While this is the most successful representation to date of magnon effects in the thermal conductivity, the lattice thermal conductivity of this sample was much smaller than the boundary limit, probably as a result of macroscopic defects introduced in the flux growth process. In the case of MnF_2 (Refs. 22 and 23) it has been possible to account for the thermal conductivity without either scattering or conduction by the magnons. The anomalous effects were found to arise by resonant scattering from the tunneling states of OH^- impurities. No successful quantitative interpretation of a nonzero effect of magnons on the magnetic field dependence of the thermal conductivity in an antiferromagnet has yet appeared.

There are several reasons for this failure, both experimental and theoretical. On the experimental side strongly coupled impurities, such as OH^- in MnF_2, may mask the intrinsic effects of magnon-phonon interactions or magnon heat conduction. Hartmann[21] has found that the magnetic field dependence of the thermal conductivity of $RbMnF_3$ is controlled to a great extent by the presence of trace concentrations of transition-metal impurities. This has led to some suspicion that impurities may be so dominant that it might not be possible to observe the intrinsic effects of magnons.

Theoretical attention has been directed largely toward the effects of magnons as heat carriers[25–28] rather than as a source of thermal resistance.[28–33] It now appears, however, that magnons manifest themselves as a major source of thermal resistance at least as often as they contribute to the heat current. To date most theoretical models have focused on magnets with high Curie and Néel temperatures, such as the iron garnets and ferrites. The magnons are then adequately treated at low temperatures by a small wave-vector approximation with the magnon energies expanded to terms quadratic in the wave vector. Such an approximation breaks down for magnetic materials that order only at cryogenic temperatures. In these, magnons may be excited over a substantial portion of the Brillouin zone even at liquid-helium temperatures.

Most recently experimental interest has been directed toward antiferromagnets that order only at cryogenic temperatures. The materials studied have included MnF_2 and $RbMnF_3$,[13,21–23] for which one has detailed dispersion relations from neutron scattering and nearly exhaustive determinations of the equilibrium magnetic properties. The motivation here has been to study systems that are thoroughly understood and for which reliable numerical calculations might be made including many of the detailed properties of the materials.[22,23] This program has been impeded by the impurity problems noted above.[21–23]

A second group of antiferromagnets that have received recent experimental attention are those that order only at liquid-helium temperatures.[8,10–12,19] Interest here derives from the accessibility of the field-induced phases with modest superconducting magnets and the interest in critical behavior near T_N. The thermal conductivity in this group of materials is characterized by rapid, or even apparently discontinuous, changes as the phase boundaries are crossed in applied magnetic fields. Effects of this kind were first observed by Donaldson and Edmonds[5] in their original study of the thermal conductivity of antiferromagnets in applied fields. Here, too, samples have shown specimen-dependent thermal conductivities, especially those grown from high-temperature fluxes. While this indicates that impurities also pose a problem here, the similarities among the behavior of the compounds in this group, particularly those features associated with changes in magnetic phase, strongly suggest an intrinsic mechanism or mechanisms by which the magnetic states scatter phonons.[5,8,10,11,19] There are semiquantitative indications that there is also a contribution to the heat transport from the magnons in some of these materials,[11,19] reinforcing the picture of intrinsic effects of the magnons as significant.

It is important to know the effect of the intrinsic magnon-phonon interactions upon the temperature dependence and field dependence of the lattice thermal conductivity. Otherwise, it is not possible to separate the magnon contribution to the thermal conductivity in a quantitative way when they contribute both to scattering and transport. Neither can it be determined whether the magnetic field effects that are observed arise primarily from impurities or from intrinsic magnon-phonon interactions, except in the case of an identifiable dominant impurity whose effect on the phonons can be calculated, such as OH^- in MnF_2.[22,23] It can be anticipated that intrinsic magnon effects might be especially important at low temperatures for those antiferromagnetics that order in the liquid-helium range. Here virtually the entire magnon spectrum can be thermally populated, and, as noted above, there are experimental indications that much of the observed scattering arises from instrinsic mechanisms. The effect of intrinsic magnon-phonon interactions on the lattice thermal conductivity of materials such as these is the primary concern of this study.

Previous studies of intrinsic magnon-phonon interactions in antiferromagnets have concentrated on the one-magnon–one-phonon resonant process.[31,32] It has been shown that the Hamiltonian for the magnons and phonons, including this resonant interaction, can be diagonalized to obtain coupled magnetoelastic normal modes. The alteration in the normal mode spectrum from that of uncoupled magnons and phonons is insignificant except in a narrow band near resonance. In this band the normal modes have characteristics of both magnons and phonons. The relaxation time for the coupled modes can be obtained from the relaxation times for uncoupled phonons and magnons as described by Kittel[29] for a ferromagnet. Unless the magnon relaxation time is very much smaller than that for the phonons in this band, the thermal conductivity due to the coupled modes will not differ substantially from that of uncoupled phonons and magnons. These interactions have been invoked to account for the temperature dependence of the thermal conductivity in $FeCl_2$,[16] and $CoCl_2 \cdot 6H_2O$.[19] It should be noted, also, that because the Hamiltonian can be diagonalized including these one-magnon–one-phonon interactions they do not contribute to the relaxation time of the normal modes in first-order perturbation theory.

The question of the effect of magnon-phonon processes on the lattice thermal conductivity of a given material may conveniently be considered in three parts. What are the interactions responsible for magnetic order in this material? What magnon-phonon processes arise from the modulation of these interactions by the lattice vibrations? Which of these magnon-phonon processes have a significant effect on the lattice thermal conductivity? The answer to the first of these questions determines the band of magnon energies available to scatter phonons and the interactions available for modulation. The answer to the second gives the scattering amplitudes and selection rules for the possible magnon-phonon processes, including their dependence on temperature and magnetic field. The last question relates to the relative magnitudes of the scattering amplitudes and to population factors that allow consideration to be limited to a few low-order processes, at least at low temperatures.

In this paper a simple model of two-magnon–one-phonon scattering is developed from a model Hamiltonian commonly used to describe the static properties of antiferromagnets, especially in the field-induced phases. The interaction responsible for the ordered magnetic state in this model system are isotropic exchange, Ising-like anisotropic exchange, uniaxial single-ion anisotropy, and the Zeeman interaction with an applied field. The motivation for studying this particular model is threefold. First, it is one of the simplest Hamiltonians exhibiting field-induced phase transitions. Second, it has frequently been used by experimentalists to interpret measurements of the static properties of antiferromagnets. Thus, one has access to a parameterization of the magnetic interactions in such materials as $MnCl_2 \cdot 4H_2O$, $GdVO_4$, and $CoCl_2 \cdot 6H_2O$ independent of any measurements of thermal conductivity. Finally, the transformations to magnon operators are known, making it possible to follow the scattering amplitudes of the magnon-phonon interactions from one phase to another. The scattering amplitudes and phonon relaxation times for these processes have been obtained without recourse to the long-wavelength approximation for the magnons. This permits the inclusion of the effects of the finite extent of the magnon energy bands. The energy-conservation surface in reciprocal space has been obtained for these processes in the limit that the sound velocity is much greater than the magnon group velocities. This approximation is appropriate to magnets ordering only at liquid-helium temperatures. For such materials the sound velocity is typically 10^2—10^3 times larger than that of the magnons. The phonon relaxation times are related to the lattice thermal conductivity via the conventional Debye model.[34] Results are obtained in the antiferromagnetic phase and in the two field-induced phases for fields applied along the easy axis for sublattice magnetization. Methods of distinguishing experimentally between these the effects of these two-magnon—one-phonon processes and those of other magnon-phonon processes are discussed.

The model is applied in the following paper[35] (hereafter referred to as Paper II) to the analysis of the thermal conductivity of $MnCl_2 \cdot 4H_2O$, a material in which thermal resistance has previously been shown to be the dominant effect of the magnons.[8,36]

II. METHOD

A uniaxial antiferromagnet described by the anisotropic Heisenberg Hamiltonian

$$\begin{aligned}\mathcal{H}_S = {} & \sum_{\langle\alpha,\beta\rangle} J(\vec{\alpha}-\vec{\beta})S(\vec{\alpha})\cdot S(\vec{\beta}) \\ & + \sum_{\langle\alpha\beta\rangle} K(\vec{\alpha}-\vec{\beta})S_Z(\vec{\alpha})S_Z(\vec{\beta}) \\ & - \sum_{\alpha} L(\vec{\alpha})S_Z^2(\vec{\alpha}) - \sum_{\beta} L(\vec{\beta})S_Z^2(\vec{\beta}) \\ & - \mu H\left(\sum_{\alpha} S_Z(\vec{\alpha}) + \sum_{\beta} S_Z(\vec{\beta})\right)\end{aligned} \tag{1}$$

is considered. $\vec{\alpha}$ and $\vec{\beta}$ are lattice sites on the α and β sublattices, respectively; and the sum over $\langle\alpha,\beta\rangle$ is over nearest-neighbor pairs. Ising-like exchange anisotropy has been included in the K term, and the L terms represent single-ion anisotropy. The magnetic field H is applied parallel to the Z axis which is the easy axis for sublattice magnetization. The equilibrium properties for such a system have been treated in the spin-wave approximation by several authors.[37–39] It is found that at sufficiently low temperatures a transition occurs to a spin-flop phase as the field is increased. At still higher fields a second transition to an aligned paramagnetic state occurs.

The phonons are described in the harmonic approximation by

$$\mathcal{H}_P = \sum_{q,s,} \hbar\omega_{q,s}(c_{q,s}^{\dagger}c_{q,s} + \tfrac{1}{2}) \ , \tag{2}$$

where $c_{q,s}^{\dagger}$ and $c_{q,s}$ are, respectively, creation and annihilation operators for phonons with wave vectors $\vec{q}$ and polarization s.

It is the interactions between the phonons and the magnons that is of interest here. Because of the dependence of the exchange and anisotropy parameters of the spin Hamiltonian on the relative positions of the magnetic ions and their surroundings, the energy of a given spin configuration is modulated by the lattice vibrations. This gives rise to interactions between the phonons and magnons. These interactions are introduced into the model by letting $\vec{\alpha} = \vec{\alpha}_0 + \vec{u}_\alpha$ and $\vec{\beta} = \vec{\beta}_0 + \vec{u}_\beta$, where $\vec{\alpha}_0$ and $\vec{\beta}_0$ are the equilibrium positions of the ions and $\vec{u}_\alpha$ and $\vec{u}_\beta$ are their displacements from equilibrium. The spin Hamiltonian can be expanded in the displacements as

$$\mathcal{H}_S = \mathcal{H}_S^0 + \mathcal{H}' \ . \tag{3}$$

Here $\mathcal{H}_S^0$ is the Hamiltonian with the exchange and anisotropies evaluated at equilibrium. At low temperatures one-phonon processes will dominate; thus, only first-order terms in the displacements need be retained in the interaction Hamiltonian $\mathcal{H}'$. Then

$$\begin{aligned}\mathcal{H}' = {} & \sum_{\langle\alpha,\beta\rangle} \vec{J}'\cdot(\vec{u}_\alpha - \vec{u}_\beta)\vec{S}(\vec{\alpha})\cdot\vec{S}(\vec{\beta}) \\ & + \sum_{\langle\alpha,\beta\rangle} \vec{K}'\cdot(\vec{u}_\alpha - \vec{u}_\beta)S_Z(\vec{\alpha})S_Z(\vec{\beta}) \\ & + \sum_{\alpha}\sum_{\gamma,x} \vec{L}'_{\gamma x}\cdot(\vec{u}_\alpha - \vec{u}_{\gamma x})S_Z^2(\alpha) \\ & + \sum_{\beta}\sum_{\gamma,x} \vec{L}_{\gamma,x}\cdot(\vec{u}_\beta - \vec{u}_{\gamma,x})S_Z^2(\vec{\beta}) \ ,\end{aligned} \tag{4}$$

where $\vec{\gamma}$ denotes the cell and $\vec{x}$ the basis vector of each ion, nonmagnetic as well as magnetic; $\vec{J}'$ and $\vec{K}'$ are gradients with respect to $\vec{u}_\alpha$, evaluated at equilibrium; and $\vec{L}'_{\gamma x}$ is the gradient with respect to $\vec{u}_{\gamma x}$, evaluated at equilibrium. The magnon-phonon interactions are obtained by transforming the spin operators into magnon variables by the Holstein-Primakoff transformations[39,40] that diagonalize $\mathcal{H}_S^0$ and expressing the lattice displacements in phonon variables as

$$\vec{u}_{\gamma x} = -i \sum_{q,s} \vec{e}_{qsx} (\hbar / 2 N m_x \omega_{q,s})^{1/2} \times (c_{q,s}^\dagger - c_{-q,s}) \exp(i \vec{q} \cdot \vec{\gamma}) \ . \qquad (5)$$

N is the number of *chemical* unit cells, $\vec{e}_{qsx}$ is a unit vector in the direction of the displacement of an ion at basis vector $\vec{x}$ due to the $(\vec{q}, s)$ phonon, and m_x is the mass of this ion.

In terms of the magnon variables the equilibrium Hamiltonian takes the form

$$\mathcal{H}_S^0 = \sum_k (\hbar \omega_{1k} b_{1k}^\dagger b_{1k} + \hbar \omega_{2k} b_{2k}^\dagger b_{2k}) \ . \qquad (6)$$

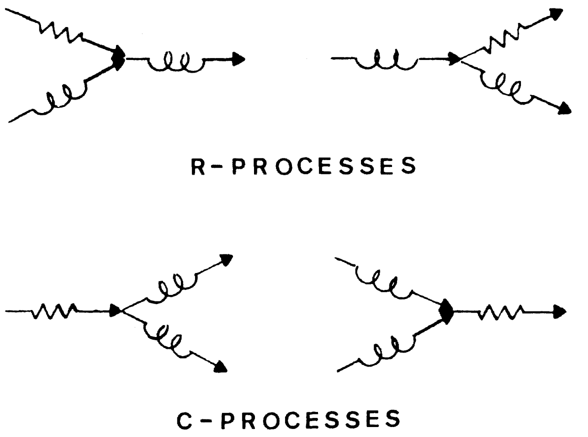

FIG. 1. Classes of two-magnon—one-phonon decay modes. For magnets with magnon group velocity smaller than the second velocity, the decay channels for R processes with both magnons on the same branch are forbidden.

Since terms bilinear in the magnon and phonon operators can be removed by the simultaneous diagonalization discussed in the Introduction,[29,31,32] the lowest-order contribution from $\mathcal{H}'$ to the phonon relaxation time comes from the three-boson (two-magnon—one-phonon) processes shown schematically in Fig. 1. These are designated as conversion processes (C processes) and radiation processes (R processes). The contribution from these to the interaction Hamiltonian is

$$\mathcal{H}_3' = \sum_{q,s} \left| \sum_{kk'} \sum_{ij} [C_{sij}(\vec{q}, \vec{k}, \vec{k}') b_{ik}^\dagger b_{jk'}^\dagger + C_{sij}^* (\vec{q}, \vec{k}, \vec{k}') b_{ik} b_{jk'} + R_{sij}(\vec{q}, \vec{k}, \vec{k}') b_{ik}^\dagger b_{jk'}] (c_{qs}^\dagger - c_{-q,s}) \right| \ ,$$

where i and j go over the magnon branches. The requirement that scattering processes satisfy conservation of energy and conservation of wave vector up to a reciprocal-lattice vector restricts the contribution of these terms to the phonon relaxation time. When the group velocity of the magnons is smaller than that of the phonons, R processes cannot satisfy these selection rules unless the two magnons lie on different branches of the excitation spectrum. This is similar to the well known restriction on three-phonon processes for longitudinal modes.[41] This restriction will be true for most materials in which the high-field phases are experimentally accessible. Additional selection rules arise from the vanishing of the C_{sij} and R_{sij} matrix elements in the various phases. These selection rules and the relation of C_{sij} and R_{sij} to the gradients J', K', L', and to the transformations to magnon operators are obtained in Sec. III.

The relaxation times associated with these processes are obtained by conventional time-dependent perturbation theory as

$$\tau_R^{-1}(q,s) = 2\pi\hbar^{-2} \sum_k |R_{s21}(\vec{q}, \vec{k}-\vec{q}, \vec{k})|^2 [n_1(\vec{k}-\vec{q}) + 1] n_2(\vec{k}) n_s^{-1}(\vec{q}) \delta(\omega_{qs} + \omega_{1k-q} - \omega_{2k}) \qquad (8)$$

and

$$\tau_c^{-1}(q,s) = 2\pi\hbar^{-2} \sum_k |C_{sij}(\vec{q}, \vec{q}-\vec{k}, \vec{k})|^2 n_1(\vec{q}-\vec{k}) n_i(\vec{k}) n_s^{-1}(\vec{q}) \delta(\omega_{q,s} - \omega_{q-k} - \omega_k) \ , \qquad (9)$$

where $n_i(\vec{k})$ is the occupation number on the ith magnon branch.

The sum over $\vec{k}$ in these expressions is nonzero only over the energy conservation surface is reciprocal space. A knowledge of the magnon spectrum is required to determine this surface fully. This spectrum is known for the model considered here,[37,39,42–44] and an approximate treatment of it to obtain the energy-conservation surface is considered in Sec. IV.

A full expansion of the interaction Hamiltonian $\mathcal{H}'$ also contains three-magnon—and four-magnon—one-phonon terms. In the temperature range where the spin-wave approximation for the magnon energies is valid without renormalization, these higher-order terms in $\mathcal{H}'$ should be negligible compared to $\mathcal{H}_3'$

since it is similar terms trilinear and quadralinear in the magnon operators that lead to renormalization by magnon-magnon interactions. In both cases this neglect is justified by the smallness of the magnon population. By similar reasoning processes involving more than one phonon have been neglected in this low-temperature approximation.

Before proceeding it should be noted again that the form of $\mathcal{H}_s$ has been chosen for simplicity rather than completeness. Real magnetic materials will have magnetic dipole forces, and perhaps antisymmetric exchange,[45,46] in addition to the isotropic exchange and Ising-like anisotropy considered here. Even if these are small enough in the equilibrium configuration of the lattice that $\mathcal{H}_s^0$ is an adequate parameterization of the spin Hamiltonian to account for the static properties of the crystal, it is not always true that they must be small in the lower symmetry induced by the lattice vibrations. There may also be other nonaxial contributions to the single-ion anisotropy. Although these may make substantial contributions to $\mathcal{H}'$ in specific materials, the exchange interactions included in this model are ubiquitous and very sensitive to interionic separation. Thus, they can be expected to be among the dominant sources of magnon-phonon interaction most frequently encountered. Although it is explicitly supposed that other sources of magnon-phonon interaction are not present for the model system considered here, the method can be applied to systems with much more complex interactions by calculating the coefficients of the transformation to magnon operators numerically.[44]

III. MATRIX ELEMENTS AND SELECTION RULES

In this section the procedure for obtaining the dispersion relations for the magnons[37,39,44] is outlined to establish a notation for the transformation to magnon operators. Both the eigenvalues of $\mathcal{H}_s^0$ and the matrix elements of the diagonalizing transformations are exhibited. These are used to calculate the matrix elements C_{sij} and R_{sij} of the two-magnon–one-phonon processes of $\mathcal{H}_3'$. By careful attention to the source of the interactions and the transformations to magnon operators these scattering amplitudes can be followed as a function of applied field. They are used in Sec. IV to obtain numerical results for the thermal conductivity.

A. Antiferromagnetic phase

The Holstein-Primakoff transformations for the antiferromagnetic phase are well known.[43,44] Spin deviation operators are introduced for each sublattice,

$$S^+(\vec{\alpha}) = (2S)^{1/2} f_\alpha(S) a_\alpha \ ,$$

$$S^-(\vec{\alpha}) = (2S)^{1/2} a_\alpha^\dagger f_\alpha(S) \ ,$$

$$S_z(\vec{\alpha}) = S - a_\alpha^\dagger a_\alpha \ ,$$

and

$$S^+(\vec{\beta}) = (2S)^{1/2} a_\beta^\dagger f_\beta(S) \ ,$$

$$S^-(\vec{\beta}) = (2S)^{1/2} f_\beta(S) a_\beta \ ,$$

$$S_z(\vec{\beta}) = -S + a_\beta^\dagger a_\beta \ ,$$

where

$$f_{\alpha,\beta}(S) = (1 - a_{\alpha,\beta}^\dagger a_{\alpha,\beta}/2S)^{1/2} \ .$$

If magnon-magnon interactions are ignored, this factor can be truncated to $f_{\alpha,\beta}(S) \approx 1$. Applying the Fourier transformations

$$a_{1k} = (2/N)^{1/2} \sum_\alpha a_\alpha \exp(-i\vec{k}\cdot\vec{\alpha}) \ ,$$

$$a_{2k} = (2/N)^{1/2} \sum_\beta a_\beta \exp(-i\vec{k}\cdot\vec{\beta}) \ ,$$

$$a_{1k}^\dagger = (2/N)^{1/2} \sum_\alpha a_\alpha^\dagger \exp(i\vec{k}\cdot\vec{\beta}) \ ,$$

$$a_{2k}^\dagger = (2/N)^{1/2} \sum_\beta a_\beta^\dagger \exp(i\vec{k}\cdot\vec{\beta})$$

and retaining only terms quadratic in the a's,

$$\begin{aligned}\mathcal{H}_S^0 \approx{}& SJz \sum_k \gamma(\vec{k})(a_{1k}a_{2\,-k} + a_{1k}^\dagger a_{2\,-k}^\dagger) \\ &+ (SJz + SKz + 2SL) \sum_k (a_{1k}^\dagger a_{1k} + a_{2k}^\dagger a_{2k}) \\ &+ \mu H \sum_k (a_{1k}^\dagger a_{1k} - a_{2k}^\dagger a_{2k}) \ . \end{aligned} \tag{10}$$

Here z is the number of nearest-neighbor magnetic ions and

$$\gamma(\vec{k}) = z^{-1} \sum_\rho \exp(-i\vec{k}\cdot\vec{\rho}) \ ,$$

where $\vec{\rho}$ denotes the relative position vectors of the nearest-neighbor magnetic ions. This quadratic Hamiltonian is diagonalized by the transformation to magnon operators

$$\begin{aligned} b_{1k}^\dagger &= l_1(\vec{k})a_{2k}^\dagger + l_2(\vec{k})a_{1\,-k} \ , \\ b_{2k}^\dagger &= l_1(\vec{k})a_{1k}^\dagger + l_2(\vec{k})a_{2\,-k} \ , \\ b_{1k} &= l_1(-\vec{1})a_{2k} + l_2(-\vec{k})a_{1\,-k}^\dagger \ , \\ b_{2k} &= l_1(-\vec{k})a_{1k} + l_2(-\vec{k})a_{2\,-k}^\dagger \ . \end{aligned} \tag{11}$$

Letting $A = SJz + SKz + 2SL$ and $d_k = SJz\gamma(k)$, the magnon frequencies are

$$\hbar\,\omega_{1,2k} = (A^2 - d_k^2)^{1/2} \mp \mu H \tag{12}$$

and the transformation coefficients are

$$l_1(\vec{\mathrm{k}}) = \cosh(\chi_k) \ , \quad l_2(\vec{\mathrm{k}}) = \sinh(\chi_k)$$

with

$$\tanh(\chi_k) = [A - (A^2 - d_k^2)^{1/2}]/d_k \ .$$

One notes that these transformation coefficients are independent of magnetic field.

The application of these transformations to the interaction terms $\mathcal{H}'$ is straightforward. The scattering amplitude for one-magnon–one-phonon processes vanishes in this phase as do the R_{s12} matrix elements. These may be seen physically as a result of angular momentum conservation in this model since each magnon carries a quantum of spin-angular momentum. For similar reasons the only nonvanishing matrix elements for C processes are the C_{s12} terms. These can be conveniently expressed as a sum of contributions from the modulation of isotropic exchange, anisotropic exchange, and single-ion anisotropy,

$$C_{s12}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') = C_{s12}^{J}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') + C_{s12}^{K}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') + C_{s12}^{L}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') \ .$$

In the long-wavelength limit for the phonons these have the form

$$C_{s12}^{J}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') = A_{q,s}^{J}\{l_1(\vec{\mathrm{k}}')l_2(\vec{\mathrm{k}})\phi_0(\vec{\mathrm{q}}) - \tfrac{1}{2}[l_1(\vec{\mathrm{k}}')l_1(-\vec{\mathrm{k}}) + l_2(\vec{\mathrm{k}}')l_2(-\vec{\mathrm{k}})]_{-k}\phi(\vec{\mathrm{q}})\}\Delta(\vec{\mathrm{q}}-\vec{\mathrm{k}}-\vec{\mathrm{k}}') \ , \quad (13a)$$

$$C_{s12}^{K}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') = A_{q,s}^{K}(\vec{\mathrm{k}}')l_2(\vec{\mathrm{k}})\phi_0(\vec{\mathrm{q}}) \ , \quad (13b)$$

$$C_{s12}^{L}(\vec{\mathrm{q}},\vec{\mathrm{k}},\vec{\mathrm{k}}') = A_{q,s}^{L}(\vec{\mathrm{k}}')l_2(\vec{\mathrm{k}})\phi_0(\vec{\mathrm{q}}) \ , \quad (13c)$$

where $\Delta(\vec{\mathrm{q}}-\vec{\mathrm{k}}-\vec{\mathrm{k}}')$ is a Kronecker delta and

$$A_{q,s}^{J} = (8S^2\hbar)^{1/2}\vec{\mathrm{J}}'\cdot\vec{\mathrm{e}}_{q,s}N^{-1/2} \ , \quad (14)$$

$$A_{q,s}^{K} = (8S^2\hbar)^{1/2}\vec{\mathrm{K}}'\cdot\vec{\mathrm{e}}_{q,s}N^{-1/2} \ , \quad (15)$$

$$A_{q,s}^{L} = (2S^2\hbar)^{1/2}\vec{\mathrm{e}}_{q,s}\cdot\sum_{x}\vec{\mathrm{L}}_{\rho x}N^{-1/2} \ , \quad (16)$$

$$\phi_k(\vec{\mathrm{q}}) = (\omega_{q,s})^{-1/2}\sum_{\rho}\vec{\mathrm{q}}\cdot\vec{\rho}\exp(i\vec{\mathrm{k}}\cdot\vec{\rho}) \ . \quad (17)$$

For R processes the only nonvanishing matrix elements are R_{s11} and R_{s22}. For the materials of present interest these cannot conserve energy and wave vector. Thus, the only three-boson processes that contribute to the relaxation of long-wavelength phonons in the antiferromagnetic phase are those in which a phonon decays into or forms from a pair of magnons, one from each of the two branches.

B. Spin-flop phase

The equilibrium Hamiltonian for the spin-flop phase has been studied by Wang and Callen[37] and by Feder and Pytee.[39] Because the spins on the two sublattices make angles $\theta(H)$ and $-\theta(H)$ with the applied field, the transformations to diagonalize $\mathcal{H}^0$ are somewhat more complicated than in the antiferromagnetic phase.

A coordinate system rotated about the x axis by θ for the α sublattice and by $-\theta$ for the β sublattice is introduced. With the spin operators in the rotated coordinate system denoted by a tilde,

$$S_X(\vec{\alpha}) = \tilde{S}_X(\vec{\alpha}) \ ,$$

$$S_Y(\vec{\alpha}) = \cos\theta\,\tilde{S}_Y(\vec{\alpha}) - \sin\theta\,\tilde{S}_Z(\alpha) \ ,$$

$$S_Z(\vec{\alpha}) = \sin\theta\,\tilde{S}_Y(\vec{\alpha}) + \cos\theta\,\tilde{S}_Z(\vec{\alpha}) \ ,$$

$$S_X(\vec{\beta}) = \tilde{S}_X(\vec{\beta}) \ ,$$

$$S_Y(\vec{\beta}) = \cos\theta\,\tilde{S}_Y(\vec{\beta}) + \sin\theta\,\tilde{S}_Z(\vec{\beta}) \ ,$$

$$S_Z(\vec{\beta}) = -\sin\theta\,\tilde{S}_Y(\vec{\beta}) + \cos\theta\,\tilde{S}_Z(\vec{\beta}) \ ,$$

and the Holstein-Primakoff transformations are applied to the spin components in the rotated coordinates.

The quadratic Hamiltonian in the Fourier transforms of the spin-deviation operators is

$$\begin{aligned}\mathcal{H}_{S2}^{0} = {} & A\sum_{k}\sum_{j} a_{jk}^{\dagger}a_{jk} \\ & + \tfrac{1}{2}B\sum_{k}\sum_{j}(a_{jk}a_{j\,-k} + a_{jk}^{\dagger}a_{j}^{\dagger}{}_{-k}) \\ & + \sum_{k} d_k(a_{1k}a_{2\,-k} + a_{1k}^{\dagger}a_{2\,-k}^{\dagger}) \\ & + \sum_{k} e_k(a_{1k}a_{2k}^{\dagger} + a_{1k}^{\dagger}a_{2k}) \ .\end{aligned}$$

where

$$\begin{aligned} A = {} & -SJz - SKz + 2SL \\ & + 2\sin^2\theta(SJz + SKz/2 - 3SL/2) + \mu H\cos\theta \ ,\end{aligned}$$

$$B = LS\sin^2\theta \ ,$$

$$d_k = Sz(J + K/2)\sin^2\theta\,\gamma(\vec{\mathrm{k}}) \ ,$$

$$e_k = SJz\gamma(k) - d_k \ ,$$

and the angle θ is determined by

$$\cos\theta = \mu H/(2SJz + SKz - 2SL)$$

at equilibrium. In the following θ is taken to be fixed by this constraint.

The quadratic Hamiltonian is diagonalized by the transformations[44]

$$a_{jk}^{\dagger}=\sum_i [V_{ji}(\vec{k})b_{ik}+W_{ji}(\vec{k})b_{i\,-k}]$$

and

$$a_{jk}=\sum_i [W_{ji}^{*}(\vec{k})b_{i\,-k}^{\dagger}+V_{ji}^{*}(\vec{k})b_{ik}]\ ,$$

giving the magnon energies

$$\hbar\omega_{1k}=[(A+B-d_k-e_k)(A-B+d_k-e_k)]^{1/2} \tag{18}$$

and

$$\hbar\omega_{2k}=[(A+B+d_k+e_k)(A-B-d_k+e_k)]^{1/2} \tag{19}$$

Defining[37]

$$l_{1f}(\vec{k})=[(A-B+d_k-e_k)/\hbar\omega_{1k}]^{1/2}$$

and

$$l_{2f}(\vec{k})=[(A-B-d_k+e_k)/\hbar\omega_{2k}]^{1/2}\ ,$$

the transformation coefficients are

$$V_{11}(\vec{k})=2^{-3/2}[l_{1f}(\vec{k})+l_{1f}^{-1}(\vec{k})]=-V_{21}(\vec{k})\ , \tag{20}$$

$$V_{12}(\vec{k})=2^{-3/2}[l_{2f}(\vec{k})+l_{2f}^{-1}(\vec{k})]=V_{22}(\vec{K})\ , \tag{21}$$

$$W_{11}(-\vec{k})=2^{-3/2}[l_{1f}(\vec{k})-l_{1f}^{-1}(\vec{k})]=-W_{21}(-\vec{k})\ , \tag{22}$$

$$W_{12}(-\vec{k})=2^{-3/2}[l_{2f}(\vec{k})-l_{2f}^{-1}(\vec{k})]=W_{22}(-\vec{k})\ . \tag{23}$$

Note that these depend on applied field, unlike those in the lower phase.

As before, the magnon-phonon interactions are obtained by applying these transformations to the interaction Hamiltonian H', Eq. (4). In the spin-flop phase, one obtains a term bilinear in the magnon and phonon operators as well as one linear in the phonons and quadratic in the magnons. This extra term allows resonant magnon-phonon conversion. As noted above, such terms may be removed by simultaneous diagonalization of the magnon and phonon Hamiltonians, leading to coupled magnetoelastic modes.[29,31,32] The alteration in the normal mode spectrum is insignificant except in a narrow band near resonance. In this band the modes have characteristics of both magnons and phonons. Unless the magnon relaxation time is much smaller than that for phonons in this band, the thermal conductivity due to the coupled modes will not substantially differ from that of uncoupled phonons and magnons. Also, the two-magnon—one-phonon processes act over a broad band of frequencies and tend to mask the resonant conversion process, if it lies in the same band. Consideration of magnon relaxation times and heat conduction by the magnons is beyond the scope of this paper; the effect of the resonant conversion processes is deferred to a separate publication, except to note that they may affect the thermal conductivity in cases of fast magnon relaxation.

The leading contribution to the phonon relaxation time is still the two-magnon—one-phonon scattering described by $\mathcal{H}_3'$, Eq. (7). In the spin-flop phase all the C and R processes that can satisfy energy and wave-vector conservation survive the transformation to magnon operators.

The matrix elements are again expressible as a sum

$$C_{sij}(\vec{q},\vec{k},\vec{k}')=\sum_{X=J,K,L} C_{sij}^{X}(\vec{q},\vec{k},\vec{k}')$$

and

$$R_{sij}(\vec{q},\vec{k},\vec{k}')=\sum_{X} R_{sij}^{X}(\vec{q},\vec{k},\vec{k}')\ .$$

The contributions from the individual interactions take the form

$$\begin{aligned}C_{sij}^{X}(\vec{q},\vec{k},\vec{k}')=A_{q,s}^{X}[&\phi_0(\vec{q})T_{1ij}^{X}(\vec{k},\vec{k}')\\&+\phi_{-k'}(\vec{q})T_{2ij}^{X}(\vec{k},\vec{k}')]\\&\times\Delta(\vec{q}-\vec{k}-\vec{k}')\ ,\end{aligned} \tag{24}$$

and

$$\begin{aligned}R_{sij}^{X}(\vec{q},\vec{k},\vec{k}')=A_{q,s}^{X}[&\phi_0(\vec{q})T_{3ij}^{X}(\vec{k},\vec{k}')\\&+\phi_{k'}(\vec{q})T_{4ij}^{X}(\vec{k},\vec{k}')]\\&\times\Delta(\vec{q}-\vec{k}+\vec{k}')\ .\end{aligned} \tag{25}$$

The coefficients $A_{q,s}^{X}$ and $\phi_k(\vec{q})$ are defined as before by Eqs. (14)–(17). The T_{mij}^{X} factors are bilinear combinations of the coefficients of the transformations to magnon operators. These are more complex than the corresponding factors in the antiferromagnetic phase and are tabulated in the Appendix.

One finds that for modulation of single-ion anisotropy the T_{1ij}^{L} and T_{3ij}^{L} factors vanish if $i=j$. Thus, this interaction does not couple phonons to pairs of magnons on the same branch. However, such processes are possible for exchange modulation. The simple connection between angular momentum conservation and selection rules has been lost because the axes of quantization for the two sublattices are not coincident with the z axis about which spin angular momentum is conserved. Now the magnon operators create and destroy spin deviations on both sublattices. As was the case in the lower phase, the R_{s11} and R_{s22} terms cannot produce first-order processes that conserve energy and wave vector.

C. Paramagnetic phase

In the paramagnetic phase the two sublattices can no longer be distinguished by the direction of sublattice magnetization. The magnetic unit cell is then the same as the chemical unit cell. If this unit cell contains only a single magnetic ion, the spin Hamiltonian is diagonal in the a_k operators.[39] Thus, the only possible two-magnon—one-phonon processes are R processes. These are forbidden by energy and wave-vector conservation for low-group-velocity magnons since there is only a single-magnon branch. For materials with magnon group velocities greater than those for phonons, the paramagnetic phase at low temperatures occurs at inaccessibly high magnetic fields. Thus, in this case no two-magnon—one-phonon processes occur for materials of experimental interest.

If each chemical cell contains magnetic ions on both the α and β sublattices, a different situation occurs. Two distinct magnon branches remain, and the spin Hamiltonian is that for the spin-flop phase with $\theta=0$. The C_{sij} matrix elements vanish; the R_{s12} matrix elements are given by Eq. (25). Thus, the scattering changes continuously across the upper phase boundary.

IV. NUMERICAL RESULTS FOR MAGNETS WITH LOW T_N

A. Approximate evaluation of the relaxation times.

In the case of present interest the group velocity of the phonons is much greater than that for the magnons. If the spatial anisotropy of the dispersion relations is neglected, the energy conservation surfaces in reciprocal space will be approximately spherical over most of the Brillouin zone ($k \gg q$). The sum over $\vec{k}$ in Eqs. (8) and (9) may be approximated as an integral over a sphere. The energy δ function for R processes takes the form

$$\delta(\omega_{q,s}+\omega_{1\,k-q}-\omega_{2k})=\delta(k-k_c)/v' \ ,$$

where k_c is the value of k that conserves energy and

$$v'=|v_1(k-q)-v_2(k)| \ .$$

Thus, from Eq. (8)

$$\tau_R^{-1}=(V/v'2\pi\hbar^2)|R_{s21}(q,k_c,k_c-q)|^2 \times k_c^2(n_1+1)n_2n_s^{-1} \ ,$$

where V is the sample volume. Similar expressions are easily obtained for the C processes.

Because of the highly dispersive nature of the magnons and the fact that the phonons interact with the whole magnon band, a simple algebraic expression for the relaxation times is not readily obtained. Consequently, these have been evaluated numerically to obtain thermal conductivities.

Because the gradients of the exchange and anisotropy are not generally well known, it is convenient to combine these and the other constants in the matrix elements into a single parameter by defining

$$\mathcal{A}^J=A^J_{q,s}V/(2\pi\hbar^2)^{1/2} \ ,$$

$$\mathcal{A}^K=A^K_{q,s}V/(2\pi\hbar^2)^{1/2} \ ,$$

$$\mathcal{A}^L=A^L_{q,s}V/(2\pi\hbar^2)^{1/2} \ .$$

Each scattering amplitude $\mathcal{A}^X$ may then be treated as a parameter to compare the model with experiment.

B. Calculation of thermal conductivities

Thermal conductivities have been calculated using the conventional Debye approximation,[34]

$$K=(k_B/2\pi^2v)(k_BT/\hbar)^3\int_0^{\Theta/T}\tau x^4e^x(e^x-1)^{-2}\,dx \ ,$$

where $x=\hbar\,\omega_{q,s}/k_BT$, and the other symbols have their usual meaning. In this approximation the distinction between phonon polarizations is neglected, and v is an average velocity of sound. The relaxation time τ is the effective relaxation time due to all phonon scattering processes. This can be obtained from the relaxation times for both magnetic and nonmagnetic scattering by the usual addition of reciprocal relaxation times

$$\tau^{-1}=\sum_j\tau_j^{-1} \ ,$$

where the sum is over different scattering processes. In the results presented below, the only nonmagnetic scattering is that due to specimen boundaries.[47] The velocity of sound has been taken as 3×10^5 cm/sec and the boundary-scattering length as 0.3 cm. It has been assumed that the Debye temperature is high enough that the upper limit of the integral may be taken as infinite. These approximations should hold satisfactorily in high quality crystals at temperatures below 1 K.

Figure 2 shows the temperature dependence of the thermal conductivity in the absence of a field for several values of exchange and anisotropy typical of magnets with Néel points in the liquid-helium range of temperatures. The T^3 factor in Eq. (26) has been divided out to display the relaxation effects more prominently. One notes that, as expected, the temperature of maximum scattering shifts with the magnon energy band. The effect of varying the coupling between the magnons and phonons is shown in Fig. 3 for exchange modulation. Here the temperature of maximum scattering is seen to be rather insensitive to the coupling strength although the magnitude of the effect changes greatly. This behavior is also

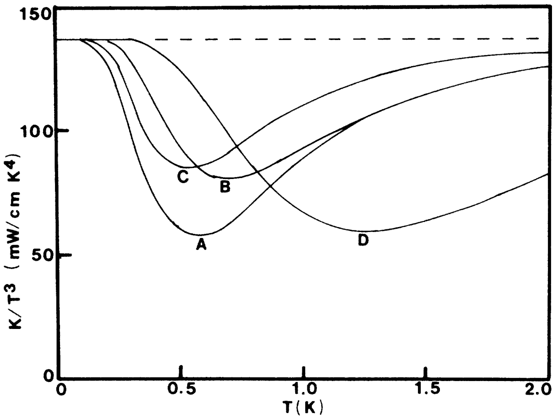

FIG. 2. Thermal conductivity calculated as a function of temperature in zero field for several values of exchange and anisotropy. Curve A: $J=0.126$ K, $K=0$, $L=0.02$ K. Curve B: $J=0.126$ K, $K=0$, $L=0.059$ K. Curve C: $J=0.916$ K, $K=0$, $L=0.059$ K. Curve D: $J=0.275$ K, $K=0$, $L=0.059$ K. In each case the coordination number z has been taken as six; and the scattering amplitudes were taken as $\mathfrak{a}_J=1.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$, $\mathfrak{a}_K=0$, and $\mathfrak{a}_L=0$. The dashed line is the boundary scattering limit.

found for modulation of the anisotropies.

The physical basis for this behavior can readily be understood from the shape of the Debye distribution of heat carriers and the selection rules for magnon-phonon interactions. The Debye distribution, $x^4e^x(e^x-1)^{-2}$, is peaked at energies near $4k_BT$. Roughly two thirds of the thermal energy is transported by phonons in the band between $2k_BT$ and $6k_BT$. In zero field the two-magnon branches are degenerate. Selection rules for C processes dictate that these two-magnon—one-phonon processes can scatter only those phonons that lie in the band between twice the smallest magnon energy and twice the largest magnon energy. For the model used here these extremes of magnon energy occur at zone center and on the zone boundary, respectively. No R processes are possible if the sound velocity is larger than the group velocities of the magnons. Thus, one expects the maximum effect of two-magnon—one-phonon scattering to occur at a temperature near $T_2=\epsilon_{\mathrm{av}}/4k_B$ where ϵ_{av} is the mean energy of the magnon band. It is important to note that the selection rules used here derive solely from conservation of energy. Thus, the argument holds independent of the mechanism by which the magnons and phonons interact as long as C processes dominate.

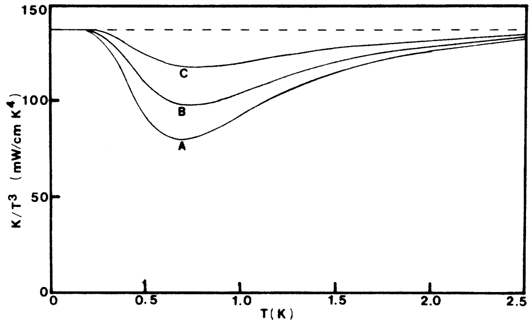

FIG. 3. Thermal conductivity calculated as a function of temperature in zero field for several values of the scattering amplitude. Curve A: $\mathfrak{a}_J=1.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$. Curve B: $\mathfrak{a}_J=0.25(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$. Curve C: $\mathfrak{a}_J=0.10(\mathrm{cm}^3\mathrm{K/sec}^2)^{1/2}$. In each case $J=0.126$ K; $K=0$; $L=0.059$ K; and $z=6$.

In the numerical examples presented in Figs. 2 and 3 the magnon energies calculated from Eq. (12) give a band of phonons scattered by C processes that extends from approximately $3k_BT$ to $5k_BT$ at the temperature of maximun scattering in each case.

It is of interest to compare the effect expected from the resonant magnon-phonon conversion process. For systems ordering only at liquid-helium temperatures the resonance is near zone center. Thus, the maximum effect of the resonant conversion process should occur near $T_1=\epsilon_0/4k_B$ where ϵ_0 is the energy of the zone-center magnon. It should be possible, therefore, to distinguish experimentally whether the resonant conversion (one-magnon—one-phonon) process or C processes dominate in such materials. Similar arguments can also be made for higher-order magnon-phonon interactions.

At the highest temperatures the dominant contribution to the scattering comes from the highest-energy magnons. Magnon-magnon interactions, which have not been included in this model, will lower the magnon energies as the Néel point is approached. Thus, the model tends to overestimate the effect of the two-magnon—one-phonon processes near T_N.

In Figs. 4 and 5 the magnetic field dependence of the thermal conductivity is displayed for two representative temperatures assuming that modulation of a single magnetic interaction dominates the scattering. The coupling coefficients have been chosen to give equal thermal conductivities in zero field. Several features are apparent. The thermal conductivity has only a weak dependence on field in the antiferromagnetic phase. It is highly structured in the spin-flop phase with the details of the structure determined by the particular magnetic interaction being modulated. For exchange modulation the scattering persists into the paramagnetic phase if there is more than one magnetic ion per chemical cell. The most prominent feature is the discontinuity seen at the antiferromagnetic to spin-flop phase boundary at 0.5 K. This structure is almost absent at 0.85 K for the exchange and anisotropy used in the calculation.

These features result from the magnon energies and their relation to the Debye distribution and from the changes in selection rules as the system passes

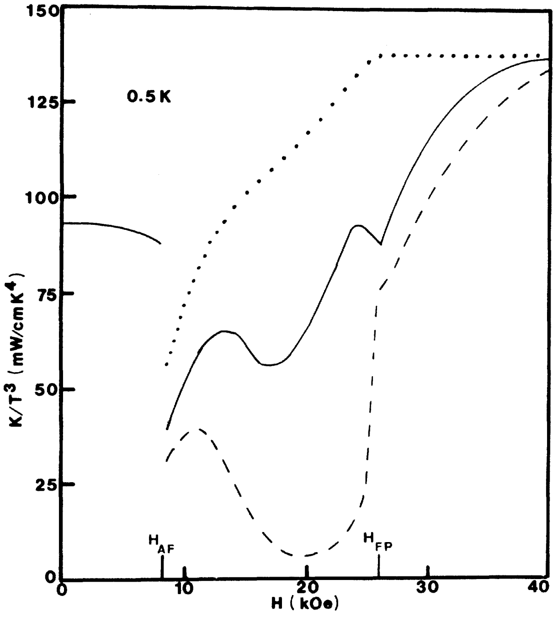

FIG. 4. Comparison of the magnetic field dependence of the thermal conductivity calculated for the three types of magnon-phonon coupling. The solid curve is isotropic exchange modulation, $\mathcal{A}_J = 1.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$; the dashed curve is anisotropic exchange modulation, $\mathcal{A}_K = 15.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$, the dotted curve is single-ion anisotropy modulation, $\mathcal{A}_L = 55.0\ (\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$. The values of the scattering amplitudes have been chosen to give the same thermal conductivity in the antiferromagnetic phase. The phase boundary between the antiferromagnetic and the spin-flop phases is indicated by H_{AF}; that between the spin-flop and paramagnetic phases, by H_{FP}.

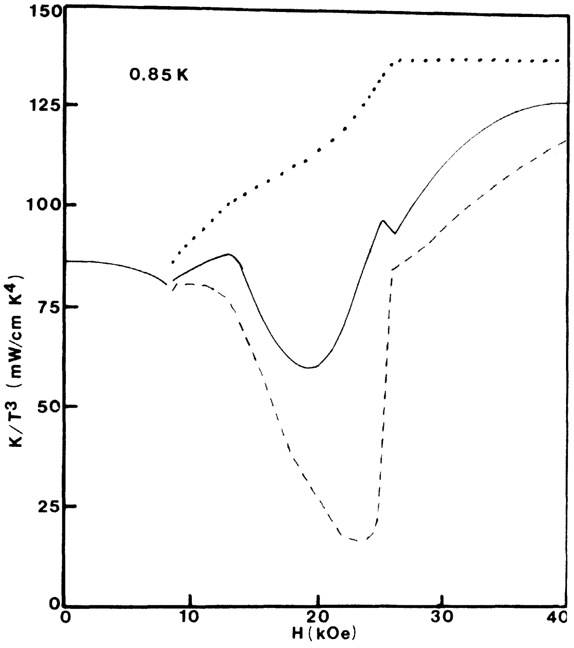

FIG. 5. Comparison of the magnetic field dependence of the thermal conductivity calculated for the three types of magnon-phonon coupling. The solid curve is isotropic exchange modulation, $\mathcal{A}_J = 1.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$; the dashed curve is anisotropic modulation, $\mathcal{A}_K = 15.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$, the dotted curve is single-ion anisotropy modulation, $\mathcal{A}_L = 55.0(\mathrm{cm}^3\,\mathrm{K/sec}^2)^{1/2}$. The values of the scattering amplitudes have been chosen to give the same thermal conductivity in the antiferromagnetic phase. The phase boundaries are indicated by H_{AF} and H_{FP}.

from one phase to another. In the antiferromagnetic phase the effect of a field parallel to the easy axis is to raise the energy of all magnons on one branch by the Zeeman energy and to lower all those on the other branch by the same amount. Selection rules prohibit all two-magnon–one-phonon processes except those C processes involving one magnon from each branch. Thus, the band of phonons that can interact with the magnons is not changed by the field as long as the system remains in the antiferromagnetic phase. The only source of magnetic field dependence is then the scattering amplitude C_{s12} and the magnon occupation numbers appearing in Eq. (9). It was shown in Sec. III A that the C_{s12} scattering amplitudes are independent of field. The entire field-dependence results from the product of occupation numbers for the two-magnon branches. Since one branch is raised in energy by the same amount that the other is lowered, the field dependence in these two occupation numbers tends to cancel; this cancellation would be exact if the occupation number were a Boltzmann factor rather than a Bose-Einstein function. Hence, only a weak field dependence results with its main effect occurring as the energy of magnons on the lower branch becomes small compared to $k_B T$.

With so little intrinsic field dependence it is easy to see that impurity effects might dominate the magnetic field dependence in this phase.[21,23] It can also be seen that if the magnons contributed substantially to the heat current then the proportionality to the magnon occupation number expected for such a contribution[25] should be easily observable. It should likewise be noted that contributions from magnon-phonon interactions that do not conserve spin angular momentum would have a strong dependence on field and thus are readily distinguishable from those processes allowed by the selection rules of the present model.

At the phase transition to the spin-flop phase both the magnon energies and the selection rules change discontinuously. One important effect is that the band of phonons that can participate in two-magnon–one-phonon processes no longer has a finite lower limit. For the numerical examples in Figs. 4 and 5 this band of phonons extends from 0 to 3.50 K in contrast to its location between 2.19 and 4.37 K in the antiferromagnetic phase. At 0.5 K the additional phonons scattered by the magnons includes important heat carriers. This produces the sharp drop in thermal conductivity as the system passes into the spin-flop phase. At 0.85 K these phonons are not important carriers, and there is little change as the phase boundary is crossed. At still higher temperatures the thermal conductivity of this model system rises on passing into the spin-flop phase. This is because the maximum magnon energy is smaller here than was the case in the antiferromagnetic phase. This has not been presented in a figure since the effects of magnon-magnon interactions and higher-order magnon-phonon interactions can be expected at these higher temperatures to alter the detailed results obtained in the two-magnon—one-phonon approximation used here. Nevertheless, this qualitative behavior could be expected if two-magnon—one-phonon scattering were important.

As the field is increased within the spin-flop phase one of the magnon branches increases its energy, especially near zone center, while the other branch remains at zero energy at zone center and changes only slightly at other wave vectors. The field dependence of the thermal conductivity arises from the magnon occupation numbers, from the increased width of the band of phonons that undergo two-magnon—one-phonon scattering, and from the field dependence of the transformations to magnon operators. At sufficiently low temperatures the increased width of the band of scattered phonons will be unimportant compared to the exponential variation in the occupation numbers. This is the case in the numerical example shown in Fig. 4. Since the effect of the occupation numbers is to decrease the scattering monotonically as the field is increased, the inflections in the thermal conductivity are seen to result from the field dependence of the transformations. The differences between the scattering induced by modulation of isotropic exchange, Ising exchange, and single-ion anisotropy arise from the different combinations of transformation coefficients that enter their scattering amplitudes. Thus, one sees that it is essential to know what combinations of spin operators occur in the interaction coupling the magnons and phonons and to carry out the details of the transformation to magnon operators. Otherwise, important contributions to the field dependence will be neglected.

At the higher temperature represented in Fig. 5 the upper limit of the band of phonons that participate in two-magnon—one-phonon processes lies near the peak in the Debye distribution. The increase in this upper limit as the higher-magnon branch is raised in energy by the magnetic field now can have a significant effect on the lattice thermal conductivity. This is responsible for the prominent dip found in the middle of the spin-flop phase for exchange modulation. The recovery of the thermal conductivity as the upper phase boundary is approached results from the vanishing of the scattering amplitudes for C processes. This structure is less prominent for modulation of single-ion anisotropy as this mechanism does not couple phonons to pairs of magnons on the same branch, and hence the band of interacting phonons broadens more slowly as the field increases.

If the broadening of the band of interacting phonons is a major source of the increased thermal resistance, then the renormalization of the magnons to lower energies by magnon-magnon interactions will substantially alter the field dependence of the thermal conductivity. This effect can be taken into account quantitatively only if experimental determinations of the temperature dependence of the magnon energies are available. When these are not available, the temperature above which magnon renormalization becomes significant can be estimated from the temperature dependence of the magnetic phase boundaries[38,39] or from the range of temperature over which noninteracting spin-wave theory can account for equilibrium properties such as the heat capacity. No attempt has been made here to include magnon renormalization effects because of the lack of agreement between theory[38,39] and experiment[48,49] for the temperature dependence of the magnetic phase boundaries.

The dip occurring at the upper phase boundary for exchange modulation is worthy of separate note. When observed experimentally, such structures have usually been attributed to critical fluctuations at this second-order phase transition.[11,19] Here it is found that two-magnon—one-phonon scattering can produce such a structure so that the invocation of a separate mechanism to account for such a dip may be unnecessary.

As noted earlier there is no two-magnon—one-phonon scattering in the paramagnetic phase if there is only one magnetic ion per chemical cell. In this case the thermal conductivity is independent of field in the paramagnetic phase except for the possible effect of four-magnon—one-phonon processes which can arise in this model but have not been treated here. If two-magnon—one-phonon scattering due to exchange modulation is the dominant process in the spin-flop phase, the thermal conductivity will increase discontinuously at this phase transition.

The numerical results for the paramagnetic phase are shown for more than one magnetic ion per cell in Figs. 4 and 5. Here the thermal conductivity varies continuously across the phase boundary. This is because the two-magnon branches are still distinct and R processes are possible. The field dependence in this phase arises from the exponential variation of the magnon occupation numbers. Note that at sufficiently high fields the magnon-phonon scattering is negligible, making possible the assessment of the effect of nonmagnetic scattering mechanisms.

Although the existing experimental results are complicated by effects of impurities and heat conduction by the magnons, it is possible to recognize many of the more prominent features of the predictions of this model in the data on $CoCl_2 \cdot 6H_2O$,[5] $MnCl_2 \cdot 4H_2O$,[8] and $GdVO_4$,[11] three antiferromagnets that have been studied into the field-induced phases. Each of these exhibits a weak field dependence in the antiferromagnetic phase with a strong reduction in the thermal conductivity on crossing the phase boundary to the spin-flop phase, an exception to this being the features at the lowest temperatures in $GdVO_4$. These were interpreted[11] as arising from a magnon heat current. In $MnCl_2 \cdot 4H_2O$, where there seem to be little heat transport by the magnons,[8] and above 0.25 K in $GdVO_4$, the thermal conductivity in the spin-flop phase exhibits the inflections predicted by the present model. As these features differ substantially from those expected for the one-magnon—one-phonon resonant conversion process, particularly as regards the relative magnitude of the thermal conductivity in the two phases, it seems likely that two-magnon—one-phonon processes are more important than resonant conversion in producing the observed field dependence. The available data do not entirely exclude the possibility that higher order processes may be also significant at some temperatures and fields.

The second velocities in MnF_2 are larger than the group velocities of the magnons.[13] The selection rules found here are thus applicable, even though the technique of numerical integration over the energy-conservation surface fails because all the excitation velocities are comparable. Sanders and Walton[23] have successfully accounted for the thermal conductivity of MnF_2 in the antiferromagnetic phase without any scattering or heat conduction by the magnons. This observation is consistent with the present model for the scattering. The lowest magnon energy is 12.54 K in MnF_2 (Ref. 50); thus, the lowest-energy phonon that can be scattered is at 25.08 K. This lower limit is independent of field. At temperatures of 4 K and below this phonon energy is greater than $6k_BT$. Hence, the phonons that could be scattered carry less than 20% of the heat. Moreover, the magnons available for scattering have only small wave vectors. The prediction of this model as applied to MnF_2 is that the intrinsic magnon-phonon scattering should be weak and independent of field without regard to the magnitude of the coupling constant for two-magnon—one-phonon processes. It can be argued with some confidence that higher-order processes should be small because of the small population of magnons at these temperatures.

V. SUMMARY

A simple model of the effect of two-magnon—one-phonon scattering on the lattice thermal conductivity of antiferromagnets has been presented. It has been shown that in the spin-flop phase important terms in the field dependence arise from the coefficients of the transformation to magnon variables. Consequently, in this phase the symmetry of the spin operators in the magnon-phonon coupling has a strong influence on the intrinsic field dependence. In the antiferromagnetic phase there is a much weaker field dependence. It is found to be identical for modulation of exchange or anisotropy, up to the magnitude of the coupling coefficient. The weakness of the intrinsic field dependence probably accounts for the sensitivity of the field dependence to impurity effects at low fields.

The results of this model are in qualitative agreement with measurements on $CoCl_2 \cdot 6H_2O$,[5] $MnCl_2 \cdot 4H_2O$,[8] $GdVO_4$,[11] and MnF_2.[23]

In the following paper a quantitative comparison is made for $MnCl_2 \cdot 4H_2O$, an antiferromagnet in which there seems to be little heat conduction by the magnons in any of the magnetic phases.

APPENDIX

The transformations to magnon operator introduce important wave vector and field-dependent factors into the two-magnon phonon-relaxation times. These appear in the transition matrix elements as $T^X_{mij}(\vec{k},\vec{k}')$. They are tabulated below for the several phases:

A. Antiferromagnetic phase

$$T^J_{112} = T^K_{112} = T^L_{112} = l_1(\vec{k}')l_2(\vec{k}) \ ,$$

$$T^J_{212} = -\tfrac{1}{2}[l_1(\vec{k}')l_1(-\vec{k}) + l_2(\vec{k}')l_2(-\vec{k})] \ ,$$

$$T^K_{212} = T^L_{212} = 0 \ .$$

B. Spin-flop phase

$$T^J_{1ij} = (1-\delta_{ij})(\sin^2\theta - \tfrac{1}{2})V_{ij}(\vec{k})W_{1i}(-\vec{k}') \;,$$

$$\begin{aligned}T^J_{2ij} = \tfrac{1}{4}\{&\sin^2\theta[V1i(\vec{k})V_{2j}(\vec{k}') + W_{1i}(-\vec{k})W_{2j}(-\vec{k}')]\\ &+\cos^2\theta[V_{1i}(\vec{k})W_{2j}(-\vec{k}') + W_{1i}(-\vec{k})V_{2j}(\vec{k})]\} \;,\end{aligned}$$

$$T^J_{3ij} = (1-\delta_{ij})(2\sin^2\theta - 1)V_{1i}(\vec{k})V^*_{1j}(\vec{k}') \;,$$

$$\begin{aligned}T^J_{4ij} = \tfrac{1}{2}\{&\sin^2\theta[V_{1i}(\vec{k})W^*_{2j}(-\vec{k}') + W_{1i}(-\vec{k})V^*_{2j}(\vec{k}')]\\ &+\cos^2\theta[V_{1i}(\vec{k})V^*_{2j}(\vec{k}') + W_{1i}(-\vec{k})W^*_{2j}(-\vec{k}')]\} \;,\end{aligned}$$

$$T^K_{1ij} = (1-\delta_{ij})\cos^2\theta\, V_{1i}(\vec{k})W_{1j}(-\vec{k}') \;,$$

$$T^K_{2ij} = \sin^2\theta\tfrac{1}{4}[V_{1i}(\vec{k})V_{2j}(\vec{k}') + W_{1i}(-\vec{k})W_{2j}(-\vec{k}') - V_{1i}(\vec{k})W_{2j}(-\vec{k}') + W_{1i}(-\vec{k})V_{2j}(\vec{k}')] \;,$$

$$T^K_{3ij} = (1-\delta_{ij})\cos^2\theta\, V_{1i}(\vec{k})V^*_{1j}(\vec{k}') \;,$$

$$T^K_{4ij} = \sin^2\theta\tfrac{1}{2}[V_{1i}(\vec{k})W^*_{2j}(-\vec{k}') + W_{1i}(-\vec{k})V^*_{2j}(\vec{k}') - V_{1i}(\vec{k})V^*_{2j}(\vec{k}') - W_{1i}(-\vec{k})W^*_{2j}(-\vec{k}')] \;,$$

$$\begin{aligned}T^L_{1ij} = (1-\delta_{ij})\{&\sin^2\theta\tfrac{1}{2}[V_{1i}(\vec{k})V_{2j}(-\vec{k}') + W_{1i}(-\vec{k})W_{2j}(-\vec{k}') - V_{1i}(\vec{k})W_{2j}(-\vec{k}')\\ &+ W_{1i}(-\vec{k})V_{2j}(\vec{k}')] + 4\cos^2 V_{1i}(\vec{k})W_{1j}(-\vec{k}')\} \;,\end{aligned}$$

$$\begin{aligned}T^L_{3ij} = (1-\delta_{ij})\{&\sin^2\theta[V_{1i}(\vec{k})W^*_{2j}(-\vec{k}') + W_{1i}(-\vec{k})V^*_{2j}(\vec{k}') - V_{1i}(\vec{k})V^*_{2j}(\vec{k}')\\ &- W_{1i}(-\vec{k})W^*_{2j}(-\vec{k}')] + 8\cos^2\theta\, V_{1i}(\vec{k})V^*_{1j}(\vec{k}')\} \;.\end{aligned}$$

C. Paramagnetic phase

These are the same as those in the spin-flop phase with $\theta = 0$. The W_{ij} transformation coefficients vanish; thus, all the T^X_{1ij} and T^X_{2ij} factors vanish in this phase.

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