

Low-Temperature Thermal Conductivity of Ferromagnetic Insulators Containing Impurities

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The spin-wave thermal conductivity of ferromagnetic insulators with magnetic scattering is investigated. The role of magnon-magnon interactions is clarified. The magnetoelastic coupling is treated by the Green-function technique.

I. INTRODUCTION

IN an insulator at low temperatures with ferro- or ferrimagnetic properties, heat conduction by spin waves is as important as heat conduction by phonons. In ferromagnetic metals, the heat transport by spin waves also plays a role. The situation in metals requires an analysis different from the one given here, because of the electronic contribution to the heat transport, and the mutual scattering of spin waves and electrons. There is ample experimental evidence for spin-wave (magnon) heat conduction in insulators.¹⁻³ The theory of this phenomenon has led to a controversy expressed in a few papers dealing with this subject.⁴⁻⁶ Therefore, in Sec. III, we set out to resolve this controversy, the essence of which is as follows:

A first team of authors^{4,5} showed by solving the linearized Boltzmann equation that the interaction of spin waves with one another will result in a reduction of the heat flux carried by spin waves. Thus magnon-magnon interaction itself leads to thermal resistance.

The other group⁶ argued that, as a result of Peierls' theorem,⁷ spin-wave interactions cannot alter a steadily drifting spin-wave distribution which carries a heat current but which in other aspects is thermodynamically stable. Hence the thermal resistance due to these interactions should be zero.

Anticipating the results of our analysis, we arrive at the following conclusion: The two treatments of the problem have different ranges of validity. A steadily drifting distribution may indeed exist in principle, in accord with Callaway and Boyd.⁶ However, dynamical conditions rarely permit the establishment of such a distribution.

The possibility of this distribution does not basically depend on the presence of magnetic anisotropy or on external field, which create an energy gap in the spin-wave spectrum, as asserted in Ref. 6. Rather is it due to a kind of condensation phenomenon, analogous to Bose-Einstein condensation.

The drifting distribution, however, carries a very small heat current and is only established when the mean free path of mutual spin-wave interactions is short compared to the sample dimensions or some other scattering distance.

Consequently, when discussing spin-wave heat conduction, one has to distinguish between two regimes: The first regime is characterized by

$$l_{ss} \ll \bar{l}$$

and the second regime by the opposite limit. l_{ss} is the magnon-magnon scattering free path, while \bar{l} is the actual mean free path of magnons determined by the cooperation of all scattering mechanisms.

The estimate given in Sec. IV (see Table I) demonstrates that in garnets the magnon-magnon scattering mean free path is so large that the spin waves cannot possibly reach thermal equilibrium in samples of ordinary dimensions at low temperatures, i.e., temperatures for which the phonon thermal conductivity is small compared with the magnon contribution. In some other classes of materials, which have a high Debye temperature but a low Curie temperature, the magnon-magnon scattering becomes important in the temperature range close to the Curie point.

The long scattering free path indicates that for thermal-conduction phenomena, boundary and impurity scattering play a more important role than the magnon-magnon scattering. For this reason, the theory of magnon thermal conductivity including impurity scattering (regime 1) will be given. Estimates⁸ of the magnon-phonon relaxation time at 10°K are $\tau_{sp} \approx 10^{-6}$ sec.

The heat current in regime 2 is practically zero unless there is an energy gap in the magnon spectrum. Further, there is no proportionality between heat current and temperature gradient. It is difficult to prove that the heat current in this limit can be calculated using the linearized Boltzmann equation, as will be explained in Sec. III.

In some cases, the transition region between regimes 1 and 2 may be reached in experiments. Then the thermal conductivity by spin waves will be found smaller than the theoretical prediction, which is based on the regime-1 calculation.

⁸ K. P. Sinha and U. N. Upadhyaya, Phys. Rev. **127**, 432 (1962).

¹ D. Douthett and S. A. Friedberg, Phys. Rev. **121**, 1662 (1961).

² B. Lüthi, J. Phys. Chem. Solids **23**, 35 (1962).

³ R. L. Douglass, Phys. Rev. **129**, 1132 (1963).

⁴ A. Quattropani, Physik Kondensierten Materie **1**, 125 (1963).

⁵ A. Quattropani and W. Baltensperger, Helv. Phys. Acta **34**, 780 (1961).

⁶ J. Callaway and R. Boyd, Phys. Rev. **134**, A1655 (1964).

⁷ See, for example, R. E. Peierls, *Quantum Theory of Solids* (Oxford University Press, London, 1956), 2nd ed., p. 42.

The magnon-magnon scattering free path is proportional to k^{-2} (k =magnon wave vector). Therefore those spin waves carrying the most heat (high k values) have the shortest free paths, and would necessitate a transition-region treatment. We estimated their share of the heat flow and found it to be small.

II. THERMAL CONDUCTIVITY OF THE FREE-MAGNON GAS

The same boundary conditions are adopted for the medium as in the phonon heat-conduction problem (cf. Ref. 9). The plane boundaries at $x=-a$ and $x=a$, perpendicular to the x axis are considered as black bodies which emit and absorb magnons according to Planck's law. The boundaries are kept at temperatures T_1 and T_2 , respectively. The energy $\epsilon(k)$, which appears in Eq. (II.2), of Ref. 9 is now taken, appropriate to spin waves,

$$\epsilon(\mathbf{k}) = \alpha k^2. \quad (2.1)$$

Anisotropy and external and demagnetizing fields are dropped because their effect on the dominant spin-wave modes is slight above liquid-helium temperature. (At liquid-helium temperature, important magnetoresistance effects arise which can be treated easily, including the Zeeman term $\mu \mathbf{H}_{\text{ext}}$ in (2.1). Also in some highly anisotropic substances, the magnetic anisotropy energy may be as high as 10–100°K times Boltzmann's constant. Our calculations do not apply for such materials.)

The heat flux of the free-magnon gas is given by

$$Q = Q^+ - Q^-, \quad (2.2)$$

where Q^+ and Q^- are the fluxes in the positive and negative x directions, respectively. It is

$$Q^+ = \Omega \int d^3k f_{\mathbf{k}}(-a) \epsilon(\mathbf{k}) v_x(\mathbf{k}), \quad (2.3)$$

and Q^- is given by a similar expression involving $f_{\mathbf{k}}(a)$, the distribution function of incoming spin waves at the other boundary. The magnon velocity is denoted by $v(\mathbf{k})$. It is $\Omega = (2\pi)^{-3}V$, where V is the volume of the sample. The latter is taken as unity; hence $\Omega = (2\pi)^{-3}$ with the dimensions of (length)³.

We use

$$v_x(\mathbf{k}) = (2\alpha/\hbar)k \cos\beta, \quad \beta = \angle(\mathbf{k}, x), \quad (2.4)$$

and

$$K_n = \int k^n f_{\mathbf{k}}^0(\alpha k^2) dk = \frac{1}{2} \left(\frac{k_B T}{\alpha} \right)^{(n+1)/2} J_{(n+1)/2-1}. \quad (2.5)$$

⁹ P. Erdős, Phys. Rev. **138**, A1200 (1965). The idea of considering the currents entering through the boundaries, to determine the conductivity of the medium through the interaction with scattering centers was first introduced by R. Landauer (International Conference on the Electronic Properties of Metals at Low Temperatures, 1958, Geneva, New York, and Conference on Statistical Mechanics and Irreversibility, Queen Mary College, London, 1960).

J_n is defined and tabulated in Ref. 9, footnote 9, and is a number of order 1–10. Thus we obtain

$$Q^+ = (2\pi\Omega\alpha^2/\hbar)^{1/2} (k_B T_1/\alpha)^3 J_2 \quad (2.6)$$

and

$$Q^- = (\pi\Omega\alpha^2/\hbar) J_2 [(k_B T_1/\alpha)^3 - (k_B T_2/\alpha)^3]. \quad (2.7)$$

For $\Delta T = (T_1 - T_2) \ll T = \frac{1}{2}(T_1 + T_2)$, this expression reduces to

$$Q = (3\pi\Omega J_2 k_B \alpha / \hbar) (k_B T / \alpha)^2 \Delta T = 0.462 C \bar{v} \Delta T, \quad (2.8)$$

where the specific heat of spin waves

$$C = 5\pi J_{3/2} \Omega k_B (k_B T / \alpha)^{3/2}, \quad (2.9)$$

and their average velocity

$$\bar{v} \equiv \langle v^2 \rangle_{\text{av}}^{1/2} = (2/\hbar) (J_{3/2} k_B T \alpha / J_{1/2})^{1/2} \quad (2.10)$$

has been inserted. The thermal conductivity of the free-spin-wave gas between the two boundaries at a distance $2a$ apart hence becomes

$$\kappa = Q/2a\Delta T = 0.462 C \bar{v} (2a). \quad (2.11)$$

This formula represents the analog of (II.7) of Ref. 9, derived for phonons, and it is reminiscent of the gas-kinetic result, where $2a$ plays the role of the mean free path of spin waves.

The actual distribution function of spin waves is given by (II.8) of Ref. 9, where the index s is to be omitted, and ϵ is given by (2.1).

Let us determine the *equivalent drifting distribution*, as defined in Sec. II, Ref. 9. This is a Bose distribution with constant drift velocity. To this end, we determine the momentum P (in the x direction), associated with the magnon assembly.

$$\begin{aligned} P &= \Omega \int d^3k \hbar k \cos\beta f^0(\epsilon) = \frac{1}{2} \pi \Omega J_1 \hbar \left[\left(\frac{k_B T_1}{\alpha} \right)^2 - \left(\frac{k_B T_2}{\alpha} \right)^2 \right] \\ &= \Omega J_1 \hbar \left(\frac{k_B T}{\alpha} \right)^2 \frac{\Delta T}{T}. \end{aligned} \quad (2.12)$$

According to the definition of the equivalent drifting distribution, we should construct a thermodynamically stable magnon distribution function with the same total energy E and momentum \mathbf{P} as found above for the actual distribution function. In contrast to the case of phonons, for spin waves, this endeavor is confronted with some difficulties, which are explained in the next section. These difficulties are precisely the same ones which led to the controversy mentioned in the previous section.

III. DRIFTING BOSE GAS WITH QUADRATIC DISPERSION LAW

In contrast to a material Boson gas with quadratic dispersion law the rest mass of spin waves—and consequently their chemical potential—is zero. In this respect spin waves resemble phonons. The resemblance

does not carry further, however, because phonons have a linear, and magnons a quadratic, dispersion law. These properties lead to an important peculiarity of the spin-wave gas, and indeed of any Bose system with zero rest mass and a quadratic dispersion law.

This peculiarity can immediately be manifested if one writes down the distribution function $f_{\mathbf{k}}$ of such a gas, distributed homogeneously in space, and drifting with an average velocity $\boldsymbol{\lambda}$ as a whole.¹⁰

$$f_{\mathbf{k}} = [\exp(\alpha k^2 - \boldsymbol{\lambda} \cdot \hbar \mathbf{k}) - 1]^{-1}, \quad \beta = 1/k_B T. \quad (3.1)$$

It appears that for any nonzero value of the drift velocity $\boldsymbol{\lambda}$ there exists a sphere in \mathbf{k} space, whose equation is

$$\alpha k^2 - \boldsymbol{\lambda} \cdot \hbar \mathbf{k} = 0 \quad (3.2)$$

of diameter $\hbar \lambda / \alpha$, touching the origin, such that for every \mathbf{k} inside this sphere the distribution function becomes negative.

The appearance of negative occupation numbers is, of course, a warning, which indicates that something is wrong. But it is premature, and even incorrect to conclude⁴ that a drifting distribution cannot exist. A more careful analysis is required.

Consider a system of Bose-type elementary excitations with a quadratic dispersion law in a crystal of cube shape of volume $V = L^3$ (or periodic boundary conditions). The available levels in reciprocal lattice space are given by

$$k_i = (2\pi/L)n_i, \quad n_i = 0, 1, \dots, N, \quad i = x, y, z,$$

where N is the total number of spin sites. Assume that the total energy E_0 , momentum P_0 is not changed in the processes which occur, due to the sufficiently weak interaction among the excitations. [The interactions with the lattice (umklapp processes, etc.) are also assumed to be negligible.] The interactions will then create a thermal equilibrium, characterized by maximum entropy S , subject to the auxiliary conditions

$$\begin{aligned} F_1 &\equiv E_0 - \sum \epsilon_{\mathbf{k}} f_{\mathbf{k}} = 0, \\ F_2 &\equiv \mathbf{P}_0 - \hbar \sum \mathbf{k} f_{\mathbf{k}} = 0. \end{aligned} \quad (3.3)$$

The number of particles must not remain constant. It is irrelevant whether or not \mathbf{P}_0 actually represents the momentum of the system: its conservation follows from the conservation of quasimomentum \mathbf{k} in the individual processes of interaction.

Anticipating trouble, we introduce a new variable $a_{\mathbf{k}}$ through

$$f_{\mathbf{k}} = a_{\mathbf{k}}^2, \quad (3.4)$$

to make sure that no negative occupation numbers occur. In the usual way, the entropy

$$S/k_B = \sum_{\mathbf{k}} [(f_{\mathbf{k}} + 1) \ln(f_{\mathbf{k}} + 1) - f_{\mathbf{k}} \ln f_{\mathbf{k}}]$$

¹⁰ Cf. L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon Press, Ltd., London, 1958), p. 205.

of a noninteracting Bose gas is extremized with respect to the $a_{\mathbf{k}}$, using Lagrangian multipliers β , $-\boldsymbol{\lambda} \cdot \beta$:

$$(\partial/\partial a_{\mathbf{k}})(S/k_B - \beta F_1 + \beta \boldsymbol{\lambda} \cdot \mathbf{F}_2) = 0,$$

which gives

$$a_{\mathbf{k}} \ln[(a_{\mathbf{k}}^2 + 1)/a_{\mathbf{k}}^2] - \beta \epsilon_{\mathbf{k}} a_{\mathbf{k}} + \beta \boldsymbol{\lambda} \cdot \hbar \mathbf{k} a_{\mathbf{k}} = 0.$$

The two solutions hereof are

$$a_{\mathbf{k}}^2 = f_{\mathbf{k}} = [\exp(\beta(\epsilon_{\mathbf{k}} - \boldsymbol{\lambda} \cdot \hbar \mathbf{k}) - 1)]^{-1} \quad \text{for } \epsilon_{\mathbf{k}} > |\boldsymbol{\lambda} \cdot \hbar \mathbf{k}| \quad (3.5)$$

and

$$f_{\mathbf{k}} = 0 \quad \text{for } \epsilon_{\mathbf{k}} < |\boldsymbol{\lambda} \cdot \hbar \mathbf{k}|.$$

It is important to note that the principle of maximum entropy has been applied in a coordinate system in which the crystal is at rest, and not in a coordinate system drifting with the Boson gas, because the problem lacks Galilean invariance. The excitations are referred to the lattice, which defines a preferred coordinate system. There is no *a priori* reason to believe that any extremum principle is available in a coordinate system drifting with respect to the lattice. It should also be noted that there is no room to introduce any nonzero chemical potential.

This distribution function should be discerned from that which describes the magnon gas in a moving reference frame R' . Suppose that R' moves with velocity $\boldsymbol{\lambda}$ with respect to the frame R , in which the body and the magnon gas are at stationary thermal equilibrium. The distribution function in R' is obtained by the application of the Doppler transformation formulas for the wave vector and frequency to the magnons in plane-wave states. Hence

$$\mathbf{k}' = \mathbf{k}$$

and

$$\epsilon' = \hbar \omega' = \hbar \omega \left| 1 - \frac{\boldsymbol{\lambda}}{\omega/k \cos \beta} \right| = |\epsilon_{\mathbf{k}} - \boldsymbol{\lambda} \cdot \hbar \mathbf{k}| = |\epsilon_{\mathbf{k}'} - \boldsymbol{\lambda} \cdot \hbar \mathbf{k}'|.$$

It follows

$$f_{\mathbf{k}'} = (\exp \beta |\epsilon_{\mathbf{k}'} - \boldsymbol{\lambda} \cdot \hbar \mathbf{k}'| - 1)^{-1}.$$

The solution (3.5) seemingly depends only on the two parameters β and $\boldsymbol{\lambda}$, and therefore should be determined uniquely by E and \mathbf{P} . However, the dependence of β and $\boldsymbol{\lambda}$ on E and \mathbf{P} is very complicated, and therefore one cannot immediately decide whether or not the diameter of the sphere bounding the unoccupied states exceeds $2\pi/L$ or not. In the former case, there actually exist unoccupied states $f_{\mathbf{k}} = 0$; in the latter case, there are no such states.

With reference to Fig. 1, let us denote the radius of the unoccupied sphere in wave-vector space by r , and the coordinate of the first occupied state by q_0 . The drift velocity is assumed parallel to k_x . In Fig. 1(a), q_0 is shown by a filled-in square.

We wish to show, by scrutinizing the variational principle in more detail, that the actual maximum of the

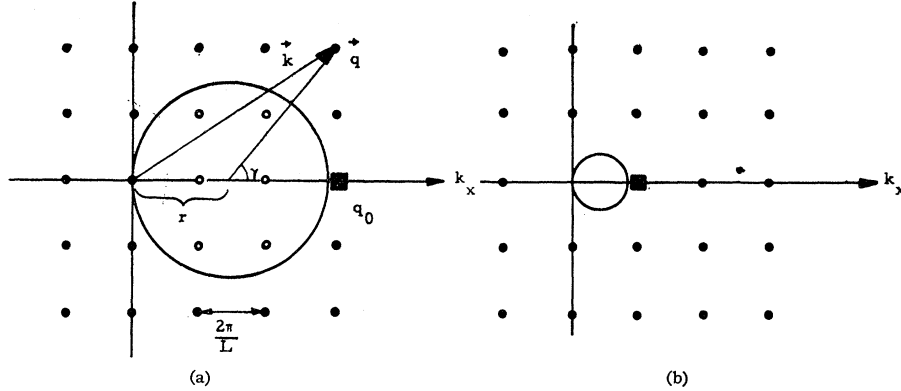


FIG. 1. Cross-sectional diagrams of k -space occupancy of the magnon assembly. The filled-in circles represent occupied states, the open circles empty states. The other symbols are explained in the text.

entropy S is reached when $q_0 = 2\pi/L$, i.e., the diameter of the unoccupied sphere is smaller than the first reciprocal lattice vector. Hence, there may exist metastable distributions such that there is a sphere of unoccupied state touching the origin, but the interaction among the spin waves will relax this distribution toward a distribution with no unoccupied states close to the origin [Fig. 1(b)].

To see this, first assume that the contrary is true: the state \mathbf{k}_l is empty, whereas the states \mathbf{k}_m and \mathbf{k}_n are filled. If by the interaction a spin wave is created in state k_l and two destroyed in \mathbf{k}_m and \mathbf{k}_n , conserving energy and momentum:

$$k_l^2 = k_m^2 + k_n^2, \quad \mathbf{k}_l = \mathbf{k}_m + \mathbf{k}_n,$$

then the change of entropy becomes

$$k_B^{-1} \delta S = k_B^{-1} \left(\frac{\partial S}{\partial f} \delta f_l + \frac{\partial S}{\partial f_m} \delta f_m - \frac{\partial S}{\partial f_n} \delta f_n \right) = \beta \alpha (k_m^2 + k_n^2) - \lambda \hbar (\mathbf{k}_m + \mathbf{k}_n) = -\beta \alpha k_l^2 + \lambda \cdot \hbar \mathbf{k}_l \geq 0,$$

where the derivatives are taken setting f equal to the equilibrium distribution function ($f_l = 0$), and $\delta f_l = -\delta f_m = -\delta f_n = 1$.

Since $\delta S > 0$, unless $l=0$, it follows that in a true equilibrium there are no empty states close to the origin. It follows

$$q_0 + r = 2\pi/L.$$

Since $r > \pi/L$, the drift velocity can never exceed the value

$$\lambda_0 = 2\pi\alpha/\hbar L.$$

If the sample is arbitrarily large, λ_0 and also the heat flow will be arbitrarily small. Of course, in a sample of any dimension, the above estimate must be supplemented by the remark that only spin waves with wavelengths smaller than their free path may be considered as realistic.

We can therefore make the following qualitative picture of what would happen in a heat-conduction experiment in a material with strong magnon-magnon interaction. As a temperature difference is applied to

the sample, spin waves will tend to decay into long-wavelength modes, i.e., they will crowd into the low-momentum states. However, since the interaction cross section decreases with decreasing spin-wave momentum, the final result will be a spin-wave distribution which is displaced towards smaller k values, the amount of displacement becoming less, as k decreases. The value of the corresponding thermal conductivity cannot be estimated on the basis of the linearized Boltzmann equation, because the deviations from equilibrium may be exceedingly large.

An interesting consequence of the shift of the distribution to low-momentum values in the presence of a temperature difference would be a decrease in the saturation magnetization. The latter is proportional to the total number of spin waves, which increases in the relaxation process.

It should again be emphasized that, in actual fact, the magnon-magnon interaction is so weak in the region of magnon thermal conductivity that the above relaxation process, and thereby its influence on thermal conductivity, is completely negligible.

IV. ESTIMATE OF THE MAGNON-MAGNON RELAXATION TIME

In the theory of thermal conductivity by spin waves the magnitude of the magnon-magnon relaxation time plays a decisive role, as discussed in Sec. III.

The magnon-magnon relaxation time τ_k , and the associated mean free path $l_k = v_k \tau_k$ (v_k = absolute value of the magnon-group velocity) may be readily estimated on the basis of the differential cross-section formula

$$\sigma_{\mathbf{k}\mathbf{k}'} = \sigma_0 k^2 k'^2 \cos^2 \theta, \quad (4.1)$$

derived by Dyson.¹¹ In this formula θ is the angle between \mathbf{k} and \mathbf{k}' , and

$$\sigma_0 = V^2/8\pi S^2, \quad (4.2)$$

where V is the volume of the unit cell, S the spin attached to a lattice point, and all spins are alike. $\sigma_{\mathbf{k}\mathbf{k}'}$ gives the cross section for the scattering of two spin

¹¹ F. Dyson, Phys. Rev. **102**, 1230 (1956).

waves with wave vectors \mathbf{k} and \mathbf{k}' into all other directions. It is convenient to express σ_0 with the help of the saturation magnetization M_s per unit volume. It is

$$\sigma_0 = (1/8\pi)(1/\delta^2)(\mu_B/M_s)^2, \quad (4.3)$$

where μ_B is the Bohr magneton, and

$$\delta = 1, \frac{1}{2}, \frac{1}{4} \quad (4.4)$$

for simple, body-centered and face-centered cubic lattices, respectively.

The reciprocal relaxation time of a spin wave with wave vector \mathbf{k} is given by

$$\frac{1}{\tau_k} = \Omega v_k \int \sigma_{\mathbf{k}\mathbf{k}'} f(k') d^3 k'. \quad (4.5)$$

Using the equilibrium Bose distribution function $f^0(\epsilon_k)$ for the spin waves, and (4.1) for the scattering cross section, the integral is easily evaluated, and gives

$$\begin{aligned} \frac{1}{\tau_k} &= \frac{2\pi}{3} J_{3/2} \Omega \sigma_0 k^2 v_k \left(\frac{k_B T}{\alpha} \right)^{5/2} \\ &= 0.6 \times 10^{-3} \left(\frac{\mu_B}{M_s \delta} \right)^2 k^2 v_k \left(\frac{k_B T}{\alpha} \right)^{5/2}. \end{aligned} \quad (4.6)$$

The free path of a spin wave of wave vector \mathbf{k} is then

$$\begin{aligned} l_k &= 1.67 \times 10^3 (M_s \delta / \mu_B)^2 k^{-2} (k_B T / \alpha)^{-5/2} \\ &= 2.19 \times 10^{17} k^{-2} T^{-5/2}. \end{aligned} \quad (4.7)$$

The last member in this and subsequent equations indicates typical values in cgs units, calculated for yttrium iron garnet (YIG) with $M_s = 200$ G, $\alpha = 0.83 \times 10^{-28}$ erg cm².

If we define the average squared wave vector of the thermal equilibrium spin-wave system by

$$\langle k^2 \rangle_{\text{av}} = \bar{\epsilon} / \alpha = (1/\alpha)(E/N), \quad (4.8)$$

where $E = 2\pi\Omega J_{3/2} \alpha (k_B T / \alpha)^{5/2}$ is the total energy and $N = 2\pi\Omega J_{1/2} (k_B T / \alpha)^{3/2}$ the total number of spin waves, then

$$\langle k^2 \rangle_{\text{av}} = 0.77 k_B T / \alpha. \quad (4.9)$$

Inserting this value in (4.7) we obtain the mean free path

$$\begin{aligned} \bar{l}_{ss} &= 2.17 \times 10^3 (M_s \delta / \mu_B)^2 (k_B T / \alpha)^{-7/2} \\ &= 1.71 \times 10^5 T^{-7/2}. \end{aligned} \quad (4.10)$$

Averaging $1/\tau_k$ (4.6) over the equilibrium distribution gives, with the definition $\bar{\tau} = (\langle \tau^{-1} \rangle_{\text{av}})^{-1}$

$$\begin{aligned} \bar{\tau} &= 0.80 \times 10^3 (\hbar / \alpha) (M_s \delta / \mu_B)^2 (k_B T / \alpha)^{-4} \\ &= 0.624 T^{-4}. \end{aligned} \quad (4.11)$$

The numerical coefficients in \bar{l}_{ss} and $\bar{\tau}$ have only an order of magnitude significance, because they depend on the way averages are taken. Table I shows a few representative values.

TABLE I. Mean free path of spin waves in yttrium iron garnet limited by magnon-magnon scattering only.

T (°K)	1	4	16	100	400
\bar{l}_{ss} (cm)	1.71×10^5	1.34×10^3	10.4	1.71×10^{-2}	1.34×10^{-4}
$\bar{\tau}$ (sec)	0.624	2.44×10^{-3}	0.952×10^{-5}	0.624×10^{-8}	2.44×10^{-11}

V. REVIEW OF PREVIOUS THERMAL-CONDUCTIVITY CALCULATIONS

At this point, it is of interest to review the calculations of Ref. 4.

The basic reason why its results are not of general validity but describe only one class of substances is because there is only one parameter of the dimension of a length which appears in the theory, namely, the lattice parameter. It is important to consider also the ratio of the magnon-magnon scattering length to the total magnon free path. (In a pure sample the latter may be set equal to the length of the sample.)

The distribution functions are drastically different, according to whether $l_{ss}/\bar{l} \gg 1$ or $l_{ss}/\bar{l} \ll 1$.

In the former case, the distribution does not have the time in passing through the sample to decay into a collection of low-momentum magnons. Then an equivalent drifting distribution may be defined by the following artifice to avoid divergent integrals: One introduces into the dispersion relation a small positive parameter δ subject only to the restriction

$$\delta > \hbar^2 \lambda^2 / 4\alpha. \quad (5.1)$$

(This is the essence of the method of Callaway and Boyd.⁶) The equivalent distribution function is set up as

$$f_k^{\text{eq}} = \frac{1}{e^{\beta(\alpha k^2 - \lambda \cdot \hbar \mathbf{k} + \delta)} - 1}, \quad (5.2)$$

which is always positive. The free parameters β and α are determined from the condition $E_{\text{true}} = E_{\text{eq}}$ and $\mathbf{P}_{\text{true}} = \mathbf{P}_{\text{eq}}$. The exact value of the cutoff parameter δ is of little importance, since E_{eq} and \mathbf{P}_{eq} depend only weakly on δ as long as the drift velocity is small compared with the average thermal velocity of the magnons. The results obtained with this equivalent drifting distribution therefore represent very good approximations.

We may also write

$$f_k^{\text{eq}} = f_k^0 \left[1 - \frac{e^{\beta(\alpha k^2 + \delta)}}{e^{\beta(\alpha k^2 + \delta)} - 1} \beta \lambda \cdot \hbar \mathbf{k} + O((\lambda \cdot \mathbf{k})^2) \right], \quad (5.3)$$

where the dominant term is the one linear in k .

Hence, if the true distribution function is written as

$$f_k = f_k^0 [1 + a_i(T) k_i + b_{ij}(T) k_i k_j + \dots], \quad (5.4)$$

the dominant term in the expansion will also be the one linear in k ; because f_k is only slightly different from f_k^{eq} . Hence the linear terms $a_i(T) k_i$ cannot be dropped as in Ref. 4.

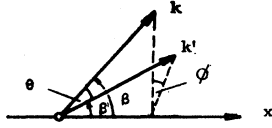


FIG. 2. Definition of the variables used in the calculation of the magnon collision operator.

In the opposite limit of $l_{ss}/\bar{l} \ll 1$, the series development of Quattropani is correct since the drift term, linear in k , is no longer dominant and may be dropped. Whether or not the other terms are small and the expansion converges sufficiently well can only be judged by comparison of the results with experiment.

VI. CALCULATION OF THE MAGNON THERMAL CONDUCTIVITY IN THE PRESENCE OF IMPURITY SCATTERING

As explained in the previous sections, the distribution function can be split into an equivalent drifting distribution plus a correction by the introduction of an artificial cutoff parameter. The dependence of the results on the cutoff parameter is in general, difficult to establish.

Instead, we split the distribution function into a free-magnon distribution, plus a small correction due to impurities.

We write the magnon distribution function as a sum of three terms

$$f_{\mathbf{k}} = f_{\mathbf{k}}^0 + \varphi_{\mathbf{k}} + f_{\mathbf{k}}^1 \quad (6.1)$$

with

$$f_{\mathbf{k}}^0 = \{ \exp[\epsilon(k)/k_B T] - 1 \}^{-1}, \quad \epsilon(k) = \alpha k^2, \\ \varphi_{\mathbf{k}} = f_{\mathbf{k}}^0(T) [f_{\mathbf{k}}^0(T) + 1] [\epsilon(k)/k_B T^2]^{1/2} \Delta T S(\beta). \quad (6.2)$$

Here $S(\beta) = \text{sgn} \cos \beta$. The free-magnon distribution under the boundary conditions explained in Sec. II is represented by $f_{\mathbf{k}}^0 + \varphi_{\mathbf{k}}$, and $f_{\mathbf{k}}^1$ is the unknown correction to the distribution function due to the scattering. The collision operator $L_{\mathbf{k}}\{f\}$ represents the change in the number of magnons with wave vector \mathbf{k} per unit volume and unit time due to impurity scattering. It may be expressed by the scattering amplitude $g_{\mathbf{k}}(\theta)$ which depends on the angle θ between \mathbf{k} and \mathbf{k}' , and on k , through

$$L_{\mathbf{k}}\{f\} = \Omega N \int (f_{\mathbf{k}} - f_{\mathbf{k}'}) |g_{\mathbf{k}}(\theta)|^2 |v_{\mathbf{k}}| d\Omega'. \quad (6.3)$$

(The factor $|v_{\mathbf{k}}|$ appears because the square of the scattering amplitude—which is the differential scattering cross section $d\sigma/d\Omega$ —is usually referred to unit incident flux instead of one particle, as here required.) N is the number of scattering centers per unit volume.

The scattering amplitude, produced by magnetic point defects, has been calculated by Callaway and Boyd⁶ for simple, body-centered and face-centered cubic lattices for nearest-neighbor exchange interactions. They found that up to the second order in k only s - and p -wave scattering occurs, and the scattering

amplitude is given by

$$g_{\mathbf{k}}(\theta) = (d + b \cos \theta) k^2,$$

where

$$d = -(V/4\pi)(J'/J)(1 - S'/S), \quad (6.4)$$

and

$$b = (V/4\pi)2(1 - J'S'/JS).$$

Here S and S' are the spins, J and J' the nearest-neighbor exchange integrals of the magnetic atoms and the isolated magnetic defects, respectively. V is the volume of the unit cell. Certain complicated structure-dependent corrections, which are small when no resonance scattering occurs, have been omitted.

The collision operator now takes the form

$$L_{\mathbf{k}}\{f\} = \Omega N \int (f_{\mathbf{k}} - f_{\mathbf{k}'}) (d^2 + b^2 \cos^2 \theta + 2db \cos \theta) 2\alpha k^5 \\ \times \sin \beta' d\beta' d\phi, \quad (6.5)$$

where $\cos \theta$ is given by

$$\cos \theta = \cos \beta \cos \beta' + \sin \beta \sin \beta' \cos \phi. \quad (6.6)$$

The angles β , β' and ϕ are defined in Fig. 2. Using (6.1), (6.2), and (6.6), the collision operator (6.5) may be evaluated:

$$L_{\mathbf{k}}\{f\} = L_{\mathbf{k}}\{f^0\} + L_{\mathbf{k}}\{\varphi\} + L_{\mathbf{k}}\{f^1\},$$

with

$$L_{\mathbf{k}}\{f^0\} = 0,$$

$$L_{\mathbf{k}}\{\varphi\} = 8\pi\Omega N \alpha^2 k^7 f_{\mathbf{k}}^0 (f_{\mathbf{k}}^0 + 1) (\Delta T / 2k_B T^2) \\ \times [(d^2 + \frac{1}{3}b^2)S(\beta) - 2db \cos \beta],$$

and

$$L_{\mathbf{k}}\{f^1\} = 8\pi\Omega N \alpha^2 k^5 \left\{ f_{\mathbf{k}}^1(\beta, x) [d^2 + \frac{1}{3}b^2] \right. \\ \left. - \frac{1}{2} \int_0^\pi f_{\mathbf{k}}^1(\beta', x) [d^2 + b^2 \cos^2 \beta \cos^2 \beta' \right. \\ \left. + \frac{1}{2}b^2 \sin^2 \beta \sin^2 \beta' + 2db \cos \beta \cos \beta'] \sin \beta' d\beta' \right\}.$$

Here we explicitly noted the dependence of $f_{\mathbf{k}}^1$ on k , β , and x . The Boltzmann equation to be solved is now

$$-\alpha k \cos \beta \partial f_{\mathbf{k}}^1(\beta, x) / \partial x = L_{\mathbf{k}}\{\varphi\} + L_{\mathbf{k}}\{f^1\}. \quad (6.7)$$

The structure of the collision operator suggests that the unknown function $f_{\mathbf{k}}^1(\beta, x)$ consists both of an odd and an even component with respect to reflections on the plane $\beta = \frac{1}{2}\pi$. Therefore it is not possible to annul the integral appearing in $L_{\mathbf{k}}\{f^1\}$ for $x=0$, as when solving the corresponding equation for phonons. For this reason we consider s - and p -wave scattering separately.

VII. S- AND P-WAVE SCATTERING. FINAL RESULTS

A. S-Wave Scattering

Pure s -wave scattering is characterized by

$$b = 0$$

in the expression (6.4) for the scattering amplitude. The Boltzmann equation (6.7) then reduces to

$$\begin{aligned}
 & -\cos\beta(\partial f_k^1(\beta, x)/\partial x) \\
 & = 8\pi\Omega N\alpha k^6 f_k^0(f_k^0+1)(\Delta T/2k_B T^2)d^2 S(\beta) + 8\pi\Omega N k^4 d^2 \\
 & \quad \times \left\{ f_k^1(\beta, x) - \frac{1}{2} \int_0^\pi f_k^1(\beta', x) \sin\beta' d\beta' \right\}. \quad (7.1)
 \end{aligned}$$

This equation is of the same form as the corresponding equation [Ref. 9, Eq. (III.7)] for phonon heat transport except for a different k dependence. Hence the exact distribution function may be found for $x=0$ by the same method as used there. It is

$$\begin{aligned}
 f_k^1(\beta, 0) & = S(\beta)\alpha k^2 f_k^0(f_k^0+1)(\Delta T/2k_B T^2) \\
 & \quad \times [\exp\{-8\pi\Omega N d^2 k^4 a/|\cos\beta|\} - 1]. \quad (7.2)
 \end{aligned}$$

The heat flow which, by symmetry, is in the x direction, is given by

$$Q = Q^0 + Q^1, \quad (7.3)$$

where Q^0 is the contribution of $f_k^0 + \varphi_k$, whereas Q^1 is the contribution from f_k^1 . The contribution of f_k^0 vanishes, hence

$$\begin{aligned}
 Q^0 & = \Omega \int v_{k,x} \epsilon(k) \varphi_k d^3 k \\
 & = 2\pi\Omega\alpha^3 \frac{\Delta T}{k_B T^2 \hbar} \int k^7 f_k^0(f_k^0+1) S(\beta) \cos\beta \sin\beta d\beta dk. \quad (7.4)
 \end{aligned}$$

The heat flow Q^1 , due to the change f_k^1 in the distribution function by the presence of impurities is

$$\begin{aligned}
 Q^1 & = \Omega \int v_{k,x} \epsilon(k) f_k^1(\beta, 0) d^3 k \\
 & = 2\pi\Omega\alpha^3 \frac{\Delta T}{k_B T^2 \hbar} \int k^7 f_k^0(f_k^0+1) \left[\exp\left\{-\frac{Bk^4}{|\cos\beta|}\right\} - 1 \right] \\
 & \quad \times S(\beta) \cos\beta \sin\beta d\beta dk \quad (7.5)
 \end{aligned}$$

with

$$B = 8\pi\Omega N d^2 a. \quad (7.6)$$

Adding the two contributions, and making use of $S(\beta)\cos\beta = |\cos\beta|$ the total heat flow is

$$\begin{aligned}
 Q & = 2\pi\Omega\alpha^3 \frac{\Delta T}{k_B \hbar T^2} \int_0^{\pi/2} \int_0^\infty k^7 f_k^0(f_k^0+1) \\
 & \quad \times \exp\left\{-\frac{Bk^4}{|\cos\beta|}\right\} \cos\beta \sin\beta d\beta dk. \quad (7.7)
 \end{aligned}$$

In the *strong-impurity-concentration* limit, the result of integration over the wave vector may be taken from

Appendix A (A10), and we obtain

$$\begin{aligned}
 Q & = 2\pi\Omega\alpha\hbar^{-1}k_B\Delta T 12.38B^{-1} \int_0^{\pi/2} \cos^2\beta \sin\beta d\beta \\
 & = 12.38 \frac{2\pi}{3} \Omega\alpha\hbar^{-1}k_B\Delta T B^{-1} \\
 & = 0.104\alpha k_B\Delta T/Nd^2a\hbar. \quad (7.8)
 \end{aligned}$$

The thermal conductivity κ is deduced from $Q = \kappa\Delta T/a$:

$$\kappa = 0.104(\alpha k_B/Nd^2\hbar). \quad (7.9)$$

We recall that d is the s -wave magnetic scattering amplitude determined by the exchange integrals and spins of magnetic defects, as given by (6.4). N is the impurity concentration, $2a$ is the distance of the boundaries, and α is the constant which appears in the spin-wave dispersion relation $\epsilon = \alpha k^2$.

This result, of course, also holds if the scattering is of other than magnetic-point-defect origin, as long as it is isotropic and the scattering amplitude depends quadratically on the wave vector k .

It is interesting to note that the spin-wave thermal conductivity does not show any size effect (in contrast to the phonon thermal conductivity). Neither does it depend on temperature in the limit of high-impurity concentration N . It is inversely proportional to the impurity concentration, again different from the phonon conductivity.

In the limit of *weak impurity scattering*, the angular integration has to be carried out first. By the same method as applied in Ref. 9, Eq. (V10), we find

$$\begin{aligned}
 Q & = \pi\Omega\alpha^3 \frac{\Delta T}{T^2} \int_0^\infty k^7 f_k^0(f_k^0+1)(1-Bk^4) dk \\
 & = \frac{7}{2}\pi J_{5/2} \Omega\alpha k_B \Delta T \left(\frac{k_B T}{\alpha}\right)^2 \left[1 - \frac{11}{7} \frac{J_{9/2}}{J_{5/2}} \left(\frac{k_B T}{\alpha}\right)^2 B \right]. \quad (7.10)
 \end{aligned}$$

Inserting the numerical values and the definition of B , the heat flow is given by

$$Q = 0.110k_B\Delta T(k_B T/\alpha)^2 [1 - 2.28Nd^2a(k_B T/\alpha)^2]. \quad (7.11)$$

In this limit, the usual T^2 -dependent thermal conductivity is diminished by a T^6 -dependent term, which is proportional to the impurity concentration.

B. P-Wave Scattering

In the case of pure p -wave scattering, the Boltzmann equation (7.7) can be solved in the same way as for pure s -wave scattering: In all results the square of the s -wave scattering amplitude d^2 has to be replaced by one-third of the square of the p -wave scattering amplitude $\frac{1}{3}b^2$.

A solution in the case of combined s - and p -wave scattering (including the interference terms) has not been worked out.

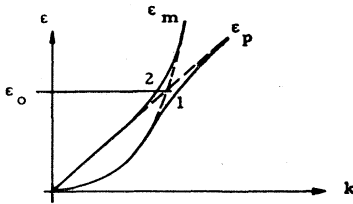


FIG. 3. Energy versus wave vector k for phonons ϵ^p , magnons ϵ^m in a ferromagnetic insulator (dashed lines). The splitting at ϵ_0 is due to the magnetoelastic coupling, and gives rise to elastomagnons, whose dispersion relation is denoted by the solid lines 1 and 2.

C. Thermal Conductivity of Antiferromagnetic Spin Waves

Antiferromagnetic spin waves have a linear dispersion relation of the form

$$\epsilon = \alpha |k|.$$

Consequently, their contribution to the thermal conductivity is determined by the same expressions as for phonons, with due replacement of the scattering cross section for phonons by that for antiferromagnetic spin waves.

VIII. INFLUENCE OF THE MAGNETOELASTIC COUPLING ON HEAT CONDUCTION

In a ferroelectric, heat is predominantly conducted by magnons at low temperatures and by phonons at high temperatures. In Fig. 3, the broken lines show the magnon and phonon branches of the excitation spectrum. (The phonon branch is triply degenerate.) In the region below the intersection ϵ_0 , the magnon branch has the lower energy, and has therefore a higher population than the phonon branch. Above ϵ_0 the situation is reversed.

The point of intersection is determined by equating the magnon energy

$$\epsilon_m = \alpha k^2 \quad (8.1)$$

and the phonon energy

$$\epsilon_p = c\hbar k; \quad (8.2)$$

hence it is given by

$$\epsilon_0 = c^2\hbar^2/\alpha. \quad (8.3)$$

Typically, for yttrium iron garnet and transverse phonons

$$\epsilon_0 = 12 \times 10^{-16} \text{ erg.}$$

This value should be compared with the thermal average energies of magnons and phonons,

$$\bar{\epsilon}_m = 0.77 k_B T = 1.06 \times 10^{-16} T \text{ erg} \quad (8.4)$$

and

$$\bar{\epsilon}_p = 0.27 k_B T = 0.373 \times 10^{-16} T \text{ erg,} \quad (8.5)$$

calculated with a Debye spectrum. The *transition region* from magnon to phonon conductivity thus may very roughly be marked off by those two temperatures

for which $\bar{\epsilon}_m = \epsilon_0$ and $\bar{\epsilon}_p = \epsilon_0$. For yttrium iron garnet this region is 11–33°K.

It is well known¹² that magnetoelastic coupling lifts the degeneracy of the two excitation branches at ϵ_0 and produces a gap Δ . The true elementary excitations are elastomagnons whose dispersion relation is shown by the solid lines 1 and 2 of Fig. 3.

It follows that the thermal properties of the body in the transition region are not determined by independent contributions from magnons and phonons, but from elastomagnons. In most cases, the elastomagnetic coupling (i.e., the gap) is small compared to ϵ_0 . The separate phonon and magnon picture is then still adequate if, in addition, the damping of both types of excitation is small.

The magnetoelastic coupling may affect the thermal conductivity even at temperatures below or above the transition region. For example, suppose that the phonons are strongly damped (for instance, through isotope scattering), whereas the magnons may be considered undamped. Then, by virtue of the magnetoelastic coupling, magnons will continuously transform into phonons of the same wave vector. Hence there will be a damping introduced into the magnon system, too, which has to be taken into account in the calculation of the mean free path and thermal conductivity. This effect will be small if the magnetoelastic interaction has a strongly resonant character, because only magnons of one particular energy ϵ_0 may be damped by the phonons. However, if the resonance is broadened by virtue of strong phonon damping, then the effect may be large enough to be of importance.

All that has been stated about the damping of magnons through phonons also remains valid for the reverse process, and the results apply equally to phonon thermal conductivity.

In Secs. IX, X, XI, and XII we derive all the properties of the magnetoelastic coupling which we need for the estimation of their influence on thermal properties. They are also of importance for ultrasonic attenuation and related phenomena.

IX. DETERMINATION OF THE ENERGY SPECTRUM OF ELASTOMAGNONS

The Hamiltonian of the magnon-phonon system in a ferroelectric in second-quantized form is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}, \quad (9.1)$$

$$\mathcal{H}_0 = \sum \epsilon_k^m a_k^\dagger a_k + \sum \epsilon_{ks}^p b_{ks}^\dagger b_{ks}, \quad (9.2)$$

a_k and b_{ks} being the destruction operators of magnons and phonons, respectively, of wave vector k and polarization index s . We have $\epsilon_k^m = \alpha k^2$, $\epsilon_{ks}^p = c_s \hbar k$. For a cubic Heisenberg ferromagnet $\alpha = 2\hbar J S a^2$, where J is the exchange integral between neighboring spins S ,

¹² E. A. Turov and Yu P. Irkin, Phys. Metals Metallog. (U.S.S.R.) (English transl.) 3, 15 (1956).

and a is the lattice constant. c_s is the sound velocity, the subscript s refers to the polarization. It is assumed that at the temperatures under consideration, the relevant k 's are considerably inside the first Brillouin zone.

To obtain \mathcal{H}_{int} , we note the well-known fact that elastic deformation changes the magnetization of a ferroelectric medium, and vice versa. Hence there exists a magnon-phonon interaction. Phenomenologically this interaction may be written as follows¹³:

$$\mathcal{H}_{\text{int}} = \int d^3x \left\{ \gamma_{iklm} M_i M_k u_{lm} + \gamma_{mikl}' \frac{\partial M_r}{\partial x_i} \frac{\partial M_r}{\partial x_k} u_{lm} \right\}, \quad (9.3)$$

with the strain tensor

$$u_{ik} = \frac{1}{2} (\partial u_i / \partial x_k + \partial u_k / \partial x_i),$$

$u_i(\mathbf{x}, t)$ is the vector of elastic displacement at the point \mathbf{x} at time t , $M_i(\mathbf{x}, t)$ is the vector of magnetization per unit volume. γ_{iklm} and γ_{mikl}' are the tensors of magnetoelastic coupling, to be specified later.

The expression (9.3) is the most general magnetoelastic Hamiltonian, which is (a) linear in the strain tensor, (b) not higher than of the second order in the magnetization and its gradient. Experience shows that this Hamiltonian is adequate to describe all magnetoelastic phenomena.

The derivation of this formula on the atomic scale has been successfully achieved,¹⁴ although the calculation of the magnetoelastic tensors from first principles has only been done for a few substances and with limited success.

We note merely that the first term of (9.3) arises from the strain dependence of the magnetic anisotropy energy. Microscopically, this dependence is largely due to the fact that the crystalline potential at the site of the magnetic ion is modulated by the lattice distortion. On the other hand, this potential acts on the spin of the magnetic ion via spin-orbit coupling. In a few instances, where the anisotropic exchange or the dipole-dipole interaction between the magnetic ions is important, it is the strain dependence of the latter, which gives rise to the first term in (9.3).

The second term of (9.3) reflects the strain dependence of the inhomogeneous magnetization, and is microscopically due to the strain dependence of the exchange interaction. Elastic waves modulate the interatomic distance and, therefore, also modulate the exchange constants, which are determined by the overlap of the wave functions of neighboring ions.

It may easily be verified that only the first term gives rise to first-order processes (i.e., involving one magnon and one phonon at a time). Hence, the second term will be dropped.

¹³ A. I. Akhiezer, V. G. Baryakhtar, and M. I. Kaganov, *Usp. Fiz. Nauk* **71**, 533 (1960) [English transl.: *Soviet Phys.—Uspekhi* **3**, 567 (1961)].

¹⁴ J. Kanamori, in *Magnetism*, edited by G. T. Rado and H. Suhl, (Academic Press Inc., New York, 1963), Vol. I, p. 127.

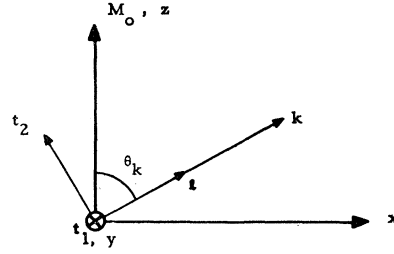


FIG. 4. Coordinate system (x, y, z) which defines the direction of saturation magnetization M_0 , the direction of propagation k of the magnetoelastic wave, and the longitudinal (l) and two transverse (t_1, t_2) directions of phonon polarization, as well as the angle θ .

Although second-order processes have a lower probability of occurrence than first-order processes (they depend on the second power of the density of magnons, rather than on the first), this may be compensated by the larger phase-space volume available for the former, as well as by the fact that the exchange interaction (and its strain dependence) is more important than the spin-orbit interaction. Therefore, only if we consider resonance phenomena, is it safe to drop the second term in (9.3), which has a nonresonant character.

For an isotropic medium

$$\gamma_{mikl} M_i M_k = \gamma_0 M^2 \delta_{ik} + \gamma_1 M_i M_k; \quad (9.4)$$

hence

$$\mathcal{H}_{\text{int}} = \int d^3x \{ \gamma_0 M^2 u_{ii} + \gamma_1 M_i M_k u_{ik} \}. \quad (9.5)$$

γ_0 and γ_1 are the magnetostriction constants.

We write $\mathbf{M} = \mathbf{M}_0 - \mathbf{m}$, \mathbf{M}_0 is the saturation magnetization, parallel to the z axis, \mathbf{m} is the deviation from \mathbf{M}_0 due to spin waves, and introduce the Holstein-Primakoff¹⁵ spin-wave operators¹⁶ a_k through

$$\begin{aligned} m_x &= (\mu M_0)^{1/2} \sum_k [a_k(t) e^{ik \cdot r} + a_k^\dagger(t) e^{-ik \cdot r}], \\ m_y &= i(\mu M_0)^{1/2} \sum_k [a_k(t) e^{ik \cdot r} - a_k^\dagger(t) e^{-ik \cdot r}], \end{aligned} \quad (9.6)$$

and

$$m_z = -\mu \sum_{kk'} a_k^\dagger a_{k'} e^{i(k'-k) \cdot r}.$$

μ is the magnetic moment of the atom.

The operator of elastic displacement $u(\mathbf{r}, t)$ at a point \mathbf{r} in the medium is expressed by the phonon operators b_{ks} (s = longitudinal l , transverse t_1 , transverse t_2), as

$$u(\mathbf{r}, t) = \hbar \sum_{ks} (2\rho \epsilon_{ks}^p)^{-1/2} \times \mathbf{e}_{ks} [b_{ks}(t) e^{ik \cdot r} + b_{ks}^\dagger(t) e^{-ik \cdot r}]. \quad (9.7)$$

ρ is the density of the medium and \mathbf{e}_{ks} the unit vector of polarization.

¹⁵ T. Holstein and H. Primakoff, *Phys. Rev.* **58**, 1098 (1940).

¹⁶ The demagnetizing field is negligible for spin waves of thermal energies as long as $k_B T / \mu M_0 \gg 2\pi$, therefore spin waves of opposite wave vector are not coupled to each other. a_k are thus the true spin wave operators.

In the interaction Hamiltonian we restrict ourselves to the terms which give rise to first-order processes. Consequently, terms containing the product of several operators a_k are dropped, and using the operator expansion (9.7) and (9.6) the Hamiltonian (9.5) becomes

$$\mathcal{H}_{\text{int}} = -\gamma_1 \sum_{ks} (\mu M_0^3 \hbar^2 / 2\rho \epsilon_{ks}^p)^{1/2} \times [(i\epsilon_{ks}^x + \epsilon_{ks}^y)k^z + \epsilon_{ks}^z(i\epsilon_{ks}^x + \epsilon_{ks}^y)] b_{ks} a_k^\dagger + \text{H.c.} \quad (9.8)$$

The superscripts refer to the coordinate axis. Owing to the neglect of second-order terms, γ_0 does not appear in \mathcal{H}_{int} .

It is convenient to define the coordinate axis, polarization directions, and the angle θ as in Fig. 4.

We set

$$\gamma_1 (\mu M_0^3 \hbar^2 / 2\rho)^{1/2} = K \quad (9.9)$$

and obtain

$$\mathcal{H}_{\text{int}} = -K \sum_k \left(\frac{ik}{(\epsilon_{kl})^{1/2}} \sin 2\theta_k b_{kl} + \frac{k}{(\epsilon_{kt_1})^{1/2}} \cos \theta_k b_{kt_1} + \frac{ik}{(\epsilon_{kt_2})^{1/2}} \cos 2\theta_k b_{kt_2} \right) a_k^\dagger + \text{H.c.} \quad (9.10)$$

It is possible to diagonalize the Hamiltonian (9.10) including \mathcal{H}_{int} , but the general results are rather unwieldy. We therefore make the simplifying assumption that the longitudinal and transverse sound velocities are equal

$$c_l = c_{t_1} = c_{t_2} \equiv c. \quad (9.11)$$

A glance at (9.10) presents us with a Bogoliubov¹⁷ canonical transformation to new Bose operators b_{k3} and b_{k4} defined through

$$\begin{aligned} b_{k4} &= \cos 2\theta_k b_{kt_2} + \sin 2\theta_k b_{kl}, \\ b_{k3} &= -\sin 2\theta_k b_{kt_2} + \cos 2\theta_k b_{kl}. \end{aligned} \quad (9.12)$$

\mathcal{H}_0 has the same form in terms of b_{k3} and b_{k4} as it had in terms of b_{kl} and b_{kt_2} , but b_{k3} drops out of \mathcal{H}_{int} . A consecutive canonical transformation

$$\begin{aligned} b_{k1} &= u_k b_{k4} + v_k b_{kt_1}, \quad u_k = (1 + \cos^2 \theta_k)^{1/2}, \\ b_{k2} &= -v_k^* b_{k4} + u_k b_{kt_1}, \quad v_k = -i \cos \theta_k (1 + \cos^2 \theta_k)^{-1/2}, \end{aligned} \quad (9.13)$$

decouples also b_{k2} , and leaves

$$\begin{aligned} \mathcal{H} &= \sum \epsilon_k^m a_k^\dagger a_k + \sum_{j=1}^3 \sum_k \epsilon_k^p b_{kj}^\dagger b_{kj} - K \sum_k (ki / (\epsilon_k^p)^{1/2}) \\ &\quad \times (1 + \cos^2 \theta_k)^{1/2} (b_{kt_1}^\dagger a_k - b_{kl} a_k^\dagger). \end{aligned} \quad (9.14)$$

At this point it is convenient to introduce the double-time, temperature-dependent Green's functions¹⁸

¹⁷ See, for example, N. N. Bogoliubov, Zh. Expt. Theor. Phys. SSSR 34, 58 (1958) [English transl.: Soviet Phys.—JETP 34, 41 (1958)].

¹⁸ For the detailed definition of the Green's function symbolism the reader should consult D. N. Zubarev, Usp. Fiz. Nauk 71, 71 (1960) [English transl.: Soviet Phys.—Uspekhi 3, 320 (1960)].

for phonons and magnons

$$G_k^m \equiv G_k^m(t-t', \beta) \equiv \langle\langle a_k(t); a_k^\dagger(t') \rangle\rangle,$$

and

$$G_k^p \equiv G_k^p(t-t', \beta) \equiv \langle\langle b_{k1}(t); b_{k1}^\dagger(t') \rangle\rangle, \quad (9.15)$$

where the double angular bracket denotes the (advanced or retarded) Green's function of the enclosed Heisenberg operators, averaged with respect to the grand canonical ensemble,¹⁹ and $\beta = 1/k_B T$. The Dyson equation of motion for the Green's function

$$i\dot{G}_k^m = \delta(t-t') \langle\langle [a_k(t), a_k^\dagger(t')] \rangle\rangle + \langle\langle [a_k(t), \mathcal{H}]; a_k^\dagger(t') \rangle\rangle \quad (9.16)$$

involves the grand canonical averages (denoted by single brackets) of commutators with \mathcal{H} . Going over to the Fourier transform with respect to time

$$G_k(E) = (2\pi)^{-1} \int_{-\infty}^{\infty} G_k(t-t'; \beta) e^{iE(t-t')} d(t-t'),$$

(9.16) and (9.14) yields

$$E G_k^m(E) = (1/2\pi) + \epsilon_k^m G_k^m(E) + g_k F_k(E), \quad (9.17)$$

with

$$g_k = [iKk / (\epsilon_k^p)^{1/2}] (1 + \cos^2 \theta_k)^{1/2}, \quad (9.18)$$

where $F_k(E)$ is the Fourier transform of the newly introduced mixed-type Green's function

$$F_k(t-t'; \beta) = \langle\langle b_{k1}(t); a_k(t') \rangle\rangle.$$

The latter may be eliminated by means of its equation of motion, which gives

$$E F_k(E) = \epsilon_k^p F_k(E) + g_k^* G_k^m(E);$$

hence

$$G_k^m(E) = (1/2\pi) [(E - \epsilon_k^m - M_k^m(E))^{-1}], \quad (9.19)$$

where

$$\begin{aligned} M_k^m(E) &= |g_k|^2 / (E - \epsilon_k^p), \\ |g_k|^2 &= (K^2 k / c\hbar) (1 + \cos^2 \theta_k). \end{aligned} \quad (9.20)$$

The energies of the elastomagnons are given by the poles of

$$G_k^m(E), \quad \text{i.e., by } E - \epsilon_k^m - M_k^m(E) = 0. \quad (9.21)$$

The two solutions of (9.21) are

$$\begin{aligned} E_k^{1,2} &= \frac{1}{2} (\epsilon_k^m + \epsilon_k^p) \\ &\quad \pm \frac{1}{2} [(\epsilon_k^m - \epsilon_k^p)^2 + 4K^2 k (1 + \cos^2 \theta_k) / c\hbar]^{1/2}. \end{aligned} \quad (9.22)$$

For $\epsilon_{k_0}^m = \epsilon_{k_0}^p = c^2 \hbar^2 / \alpha$ there is an energy gap Δ between the two branches,

$$\Delta_k = E_{k_0}^1 - E_{k_0}^2 = 2K ((1 + \cos^2 \theta_k) / \alpha)^{1/2}. \quad (9.23)$$

In terms of the gap we shall write $|g_k|^2 = 4\Delta_k^2 \epsilon^p / \epsilon_0$.

¹⁹ In this section the temperature dependence is irrelevant, and the averaging over the ensemble may be discarded. It will be used, however, in XI.

Expressed by the two elastomagnon operators and the two uncoupled phonon operators b_{k2} and b_{k3} , \mathcal{H} is diagonal. The Green's function of the phonon $G_k^p(E)$ is obtained from (9.19) simply by interchanging the superscripts m and p .

X. POLARIZATION OF THE COUPLED PHONON

It is interesting to establish the polarization of a phonon, which is produced by a magnon through the magnetoelastic coupling. This amounts to the determination of the state of polarization of

$$b_{k1} = (1 + \cos^2\theta_k)^{-1/2} [\cos 2\theta_k b_{kt_2} + \sin 2\theta_k b_{kt_1} - i \cos\theta_k b_{kt_1}].$$

Using the procedure of Appendix B, we readily find the Cartesian components of Stokes' vector

$$\mathbf{P} = (1 + \cos^2\theta_k)^{-1} \times \{\cos^2\theta_k - \cos^2 2\theta_k, 0, 2 \cos\theta_k \cos 2\theta_k\}. \quad (10.1)$$

The squared amplitude of the longitudinal polarization component can be directly read off:

$$|a_l|^2 = \sin^2 2\theta_k / (1 + \cos^2\theta_k). \quad (10.2)$$

The transverse component is elliptically polarized. Since $P_2 = 0$, the axes of the ellipse are parallel to t_1 and t_2 . The ratio of the corresponding semi-axis a and b is

$$a/b = \cos\theta_k / \cos 2\theta_k. \quad (10.3)$$

The maximum amount of longitudinal polarization is obtained for $d|a_l|^2/d\theta = 0$, which gives for the angle θ_0 between the direction of propagation of the magnon and the direction of magnetization

$$\theta_0 = 50^\circ.$$

It is easily verified that this result holds true, even if the longitudinal and transverse sound velocities are different.

From (9.12) and (9.13) we see, that for

$$\theta_k = 45^\circ, 135^\circ$$

the transverse phonons t_2 , polarized in the plane $(\mathbf{k}, \mathbf{M}_0)$ do not couple to the magnons. For

$$\theta_k = 0^\circ, 90^\circ, 180^\circ$$

the longitudinal phonons do not couple, whereas for

$$\theta_k = 90^\circ$$

the transverse phonons t_1 , polarized perpendicular to the plane $(\mathbf{k}, \mathbf{M}_0)$ do not couple to the magnons.

Of special interest are the cases of pure circular polarization. According to (B6) they are obtained as eigenstates of P_3 . Since $P_2 = 0$, the necessary condition for circular polarization is

$$P_1 = \cos^2\theta_k - \cos^2 2\theta_k = 0,$$

which yields

$$\theta_{\text{circ}} = 0^\circ, 60^\circ, 120^\circ, 180^\circ.$$

P_3 is positive for 0° and 120° and negative for 60° and 180° , hence the former and the latter two differ in their sense of polarization. This means, that phonons of the corresponding sense of circular polarization couple to the magnons, those of the opposite sense pass through the medium without interaction.

Thus the medium is birefringent with respect to the transverse phonons, as was first pointed out by Kittel.²⁰ When the wave propagates under an angle of 60° or 120° to the direction of magnetization, the circular transverse polarization may combine with longitudinal polarization, yielding conically polarized waves. Of course, the longitudinal component may be experimentally eliminated.

The difference of the group velocities of the ordinary and the extraordinary sound rays is easily obtained from the expression of the energy (9.22), if we assume that the frequency of sound is sufficiently different from the crossing frequency. In this case

$$(\epsilon_k^m - \epsilon_k^p)^2 \gg 4K^2k(1 + \cos^2\theta_k)/c\hbar$$

and the square root may be expanded to yield for the acoustic branch

$$E \simeq \epsilon_k^p + [K^2k(1 + \cos^2\theta_k)/c\hbar(\epsilon^m - \epsilon^p)].$$

The magnitude of the group velocity of the coupled phonons is

$$v = -\frac{1}{\hbar} \frac{\partial E}{\partial k} \simeq c + \frac{K^2(1 + \cos^2\theta_k)}{c\hbar^2(\epsilon_k^m - \epsilon_k^p)}.$$

For any of the directions of circular polarization of the coupled phonon, the plane of polarization of linearly polarized sound will be rotated by the angle

$$\varphi = v - c = K^2(1 + \cos^2\theta_k)/c\hbar^2(\epsilon^m - \epsilon^p)$$

per unit length. This rotation has often been used²¹ to determine the magnetoelastic coupling constant K , and to deduce γ_1 .

XI. THERMAL-EQUILIBRIUM DISTRIBUTION OF MAGNONS AND PHONONS

Keeping in mind that all energy-dependent quantities depend on the wave vector, too, the subscripts k and \mathbf{k} will be suppressed in the sequel, except where ambiguity might arise.

While the two branches of elastomagnons have a Bose distribution

$$n^{1,2} = [\exp(\beta E^{1,2}) - 1]^{-1} \quad (11.1)$$

with $E^{1,2}$ given by (9.22), the distribution function of bare magnons (and phonons) is obtained as the integral over E of the spectral density function $J^m(E)$. The

²⁰ Ch. Kittel, Phys. Rev. **110**, 836 (1958).

²¹ H. Matthews and R. C. LeCraw, Phys. Rev. Letters **8**, 397 (1962); B. Lüthi, Appl. Phys. Letters **6**, 240 (1965).

latter is defined (for magnons) as

$$J^m(E) = (i/(e^{\beta E} - 1)) \lim_{\Delta \rightarrow 0} [G^m(E+i\Delta) - G^n(E-i\Delta)]. \quad (11.2)$$

To obtain $J^m(E)$, it is rewarding to factorize the Green's function (9.19) as

$$G^m(E) = \frac{1}{2\pi(E^2 - E^1)} \left(\frac{\epsilon^m - E^1}{E - E^1} \frac{E^2 - \epsilon^m}{E - E^2} \right),$$

and use the identity

$$\lim_{\Delta \rightarrow 0} \left(\frac{1}{x+i\Delta} - \frac{1}{x-i\Delta} \right) = -2\pi i \delta(x).$$

The result being, for the number of magnons with wave vector \mathbf{k} ,

$$n_{\mathbf{k}}^m = \int_{-\infty}^{\infty} J(E) dE = \frac{\epsilon^m - E^1}{E^2 - E^1} \frac{1}{e^{\beta E^1} - 1} + \frac{E^2 - \epsilon^m}{E^2 - E^1} \frac{1}{e^{\beta E^2} - 1}. \quad (11.3)$$

The distribution function for that branch of phonons which is coupled to the magnons is obtained by replacing the superscript m by p in (11.3).

XII. MUTUAL DAMPING OF MAGNONS AND PHONONS

The knowledge of the Green's functions $G^m(E)$ and $G^p(E)$ enables us to write down the damping of one type of quasiparticle, if the damping of the other type is known.

Consider the case where the damping of the phonons due to some unspecified mechanism is given by a relaxation time τ^p , which might depend on the wave vector \mathbf{k} . As usual, we introduce the damping γ^p , assigning an imaginary part to the energy

$$\begin{aligned} \epsilon^p &= \bar{\epsilon}^p + i\gamma^p, \\ \gamma^p &= \hbar/\tau^p. \end{aligned} \quad (12.1)$$

The damping of the elastomagnons is given by the imaginary part of their energy

$$\bar{E}^{1,2} = \bar{E}^{1,2} + i\gamma^{1,2}. \quad (12.2)$$

Using Eqs. (9.20), (9.21), and (9.23), we have

$$\bar{E}^{1,2} + i\gamma^{1,2} - \epsilon^m - \frac{4\Delta^2 \bar{\epsilon}^p / \epsilon^0}{\bar{E}^{1,2} + i\gamma^{1,2} - \bar{\epsilon}^p - i\gamma^p} = 0. \quad (12.3)$$

To obtain a simple expression for $\gamma^{1,2}$ we consider two separate energy regions I, II.

(I) ϵ^m sufficiently far from ϵ^0 . Then the energy difference between one of the branches E^i and ϵ^p is

much bigger than the gap:

$$\frac{4\Delta^2 \bar{\epsilon}^p / \epsilon^0}{(E^i - \bar{\epsilon}^p)^2 + (\gamma^i - \gamma^p)^2} < \Delta.$$

The elastomagnon branch i then practically coincides with the magnon branch, so that $\gamma^i = \gamma^m$, $E^i = \epsilon^m$ and the imaginary part of (12.3) gives

$$\gamma^m = \frac{4\Delta^2 \bar{\epsilon}^p / \epsilon^0}{(\epsilon^m - \bar{\epsilon}^p)^2 + (\gamma^p)^2} \gamma^p.$$

(II) At the crossover $\epsilon^m = \epsilon^p = \epsilon^0$, (12.3) gives

$$\gamma^{1,2} = \gamma^p / 2.$$

We see that the damping of the phonons introduces a damping of order of magnitude equal to that due to the magnons in the energy region ϵ^0 . This region itself has a considerable width, if the phonon damping is strong.

A. Numerical Estimates

With $\gamma_1 = b_2/M_0^2$, where $b_2 = 7.4 \times 10^6$ erg cm⁻³, a typical value for the energy gap is $\bar{\Delta} \sim 7 \times 10^{-18}$ erg for YIG.²¹ (Δ_k averaged with respect to the angle θ_k is denoted by $\bar{\Delta}$.) Hence, $\bar{\Delta}/\epsilon^0 = 0.6\%$. It follows that if the phonon damping is less than (say) 1%, no influence on the thermodynamic properties of the magnon system can be detected.

This conclusion is, of course, not generally true: Under circumstances of very strong phonon damping, the magnon heat conductivity will be noticeably reduced. The reverse effect is probably more realistic: Very strong spin-wave damping is observed in certain materials, due to Van Vleck-type²² or other fast-relaxation mechanisms. If the material also shows strong magnetostriction, then the phonon heat conductivity will very likely be noticeably smaller than expected on the basis of calculations involving phonons only. The amount of damping may then be estimated from (12.3).

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APPENDIX A

An asymptotic expression is required for

$$J_2(m, n; B, T) = \int_0^\infty f_k^0 (f_k^0 + 1) e^{-B k^m k^n} dk, \quad (A1)$$

$$f_k^0 = \{\exp[(\alpha k^2/k_B T) - 1]\}^{-1}, \quad (A2)$$

for

$$B / \left(\frac{\alpha}{k_B T} \right)^m \gg 1. \quad (A3)$$

²² J. H. Van Vleck, Phys. Rev. **123**, 58 (1961).

Deliberate use will be made of the formulas derived in Ref. 9, Appendix.

In contrast to f_k^0 , used in Ref. 9, Appendix, f_k^0 here has a quadratic k dependence.

We set

$$k = (k_B T / \alpha)^{1/2} z^{1/2}, \quad B(k_B T / \alpha)^{m/2} = C \quad (\text{A4})$$

and write

$$J_2(m, n; B, T) = \left(\frac{k_B T}{\alpha} \right)^{(n+1)/2} \frac{1}{2} \int_0^\infty f_k^0 (f_k^0 + 1) e^{-C z^{m/2}} \times z^{(n-1)/2} dz = \frac{1}{2} \left(\frac{k_B T}{\alpha} \right)^{(n+1)/2} J_2(m, n; C). \quad (\text{A5})$$

Integrating by parts, we obtain

$$J_2(m, n; C) = -\frac{1}{2} C m K\left(\frac{1}{2}m, \frac{1}{2}(n+m-3); C\right) + \frac{1}{2}(n-1)K\left(\frac{1}{2}m, \frac{1}{2}(n-3); C\right). \quad (\text{A6})$$

$K(a, b; C)$ is defined by Ref. 9, (A6), and has been calculated for large C by asymptotic methods to yield Ref. 9, (A8). Hence

$$J_2(m, n; C) = M_2(m, n) C^{-(n-3)/m}, \quad (\text{A7})$$

with

$$M_2(m, n) = -(2\pi)^{1/2} \left\{ \left(\frac{n+m-5}{m} \right)^{(n+m-3)/m-3/2} e^{-(n+m-5)/m} - \left(\frac{n-5}{m} \right)^{(n-3)/m-3/2} m(n-1) e^{-(n-5)/m} \right\}. \quad (\text{A8})$$

Collecting the results, we arrive at

$$J_2(m, n; B, T) = \frac{1}{2} M_2(m, n) B^{-(n-3)/m} (k_B T / \alpha)^2. \quad (\text{A9})$$

Integrals which appear in the formula for spin-wave thermal conductivity involve $m=4$, $n=7$. Evaluation of (A8) and (A9) gives

$$M_2(4, 7) = 24.75,$$

and

$$J_2(4, 7; B, T) = 12.38 B^{-1} (k_B T / \alpha)^2. \quad (\text{A10})$$

APPENDIX B. PHONON POLARIZATION

Consider the creation operators of phonons of three mutually perpendicular directions of polarization b_{k1}^\dagger , b_{k2}^\dagger , b_{k3}^\dagger . The subscripts 1 and 2 denote the transverse directions, l refers to the longitudinal direction. The momentum index \mathbf{k} will be suppressed in the sequel. Let us define the operator, constructed from transverse phonon operators.

$$X_{ik} = b_i^\dagger b_k - \delta_{ik} \sum_j b_j^\dagger b_j, \quad i, k, j = 1, 2. \quad (\text{B1})$$

We shall show that the (four-component) operator $X = (X_{ik})$ describes the polarization of a phonon state—in analogy to the corresponding operator of the electromagnetic field. For this purpose we introduce Stokes' vector operator $\mathbf{P} = (P_1, P_2, P_3)$ and will show that its eigenstates correspond to certain simple polarization

states of the phonon with eigenvalues ± 1 . Stokes' vector is defined by

$$X = \mathbf{P} \cdot \boldsymbol{\sigma}, \quad \boldsymbol{\sigma} = \text{Pauli spin matrix vector}. \quad (\text{B2})$$

Hence

$$\begin{aligned} P_1 &= b_1^\dagger b_1 - b_2^\dagger b_2, \\ P_2 &= b_1^\dagger b_2 + b_2^\dagger b_1, \\ P_3 &= i(b_2^\dagger b_1 - b_1^\dagger b_2). \end{aligned} \quad (\text{B3})$$

It is

$$P^\dagger \cdot P = (n_1 + n_2)(n_1 + n_2 + 2), \quad (\text{B4})$$

where $n_i = b_i^\dagger b_i$ is the number operator of phonons with polarization i . Hence the eigenvalues of the operator $P^\dagger P$ are

$$N(N+2),$$

where N is the total number of transverse polarized phonons.

Using Eq. (B3) and the commutation relations for the b_i , it is easily shown that

$$\begin{aligned} P_1 b_1^\dagger |0\rangle &= b_1^\dagger |0\rangle, \\ P_1 b_2^\dagger |0\rangle &= -b_2^\dagger |0\rangle, \end{aligned}$$

i.e., a one-phonon state with linear polarization in the 1 direction is the eigenstate of P_1 with eigenvalue 1, whereas a one-phonon state with linear polarization in the 2 direction is the eigenstate of the same operator with the eigenvalue -1 .

States of linear polarization directed along the bisector of the angle between 1 and 2 are created by the operators

$$b_{\pi/4}^\dagger = (1/\sqrt{2})(b_1^\dagger + b_2^\dagger) \quad \text{and} \quad b_{-\pi/4}^\dagger = (1/\sqrt{2})(b_1^\dagger - b_2^\dagger).$$

These states are found to be eigenstates of P_2 ;

$$\begin{aligned} P_2 b_{\pi/4}^\dagger |0\rangle &= b_{\pi/4}^\dagger |0\rangle, \\ P_2 b_{-\pi/4}^\dagger |0\rangle &= -b_{-\pi/4}^\dagger |0\rangle. \end{aligned}$$

To obtain the eigenstates of P_3 one has to construct phonon states of right and left circular polarization²³ with the help of the operators

$$b_{\circlearrowright}^\dagger = (1/\sqrt{2})(b_2^\dagger + i b_1^\dagger)$$

and

$$b_{\circlearrowleft}^\dagger = (1/\sqrt{2})(b_2^\dagger - i b_1^\dagger). \quad (\text{B5})$$

It is

$$\begin{aligned} P_3 b_{\circlearrowright}^\dagger |0\rangle &= -b_{\circlearrowright}^\dagger |0\rangle, \\ P_3 b_{\circlearrowleft}^\dagger |0\rangle &= b_{\circlearrowleft}^\dagger |0\rangle. \end{aligned} \quad (\text{B6})$$

²³ Since b^+ represents the Fourier amplitude of the sound vibration, $b^+ = b_0^+ e^{-i\omega t}$, the operators $b_{\circlearrowright}^\dagger$ and $b_{\circlearrowleft}^\dagger$ create phonons of circular polarization. This becomes evident by inspecting the real part of the amplitude, e.g.,

$$\begin{aligned} R(b_{\circlearrowright}^\dagger) &= (1/\sqrt{2})R(b_{20}^+ e^{-i\omega t} + i b_{10}^+ e^{-i\omega t}) \\ &= (1/\sqrt{2})(b_{20}^+ \cos \omega t + b_{10}^+ \sin \omega t). \end{aligned}$$

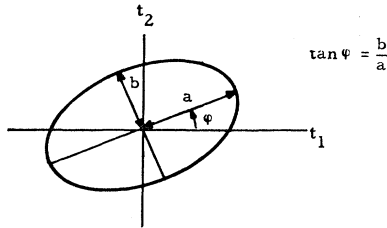


FIG. 5. Definition of the quantities a , b , and φ which characterize elliptical phonon polarization.

Let us now turn to the determination of the polarization of a general one-phonon state Φ given by

$$\begin{aligned} \Phi &= (a_1 b_1^\dagger + a_2 b_2^\dagger + a_i b_i^\dagger) |0\rangle, \\ |a_1|^2 + |a_2|^2 + |a_i|^2 &= 1. \end{aligned} \quad (\text{B7})$$

The norm P of the expectation value of the vector operator P is then found to be $(|a_1|^2 + |a_2|^2)^2$, hence

$$P = |\langle \Phi^\dagger | \mathbf{P} | \Phi \rangle| = 1 - |a_3|^2. \quad (\text{B8})$$

The amplitude of the longitudinal component is obtained from Eqs. (B7) and (B4) as

$$|a_3|^2 = 1 - |\langle \Phi^\dagger | \mathbf{P} | \Phi \rangle| = 1 - \frac{1}{3} \langle \Phi^\dagger | \mathbf{P} \cdot \mathbf{P} | \Phi \rangle. \quad (\text{B9})$$

The amplitude of the transverse components enter into the expectation values of the Stokes' parameters through

$$\begin{aligned} P_1 &= \langle \Phi^\dagger | P_1 | \Phi \rangle = a_1^* a_1 - a_2^* a_2, \\ P_2 &= \langle \Phi^\dagger | P_2 | \Phi \rangle = a_1^* a_2 + a_2^* a_1, \\ P_3 &= \langle \Phi^\dagger | P_3 | \Phi \rangle = -i a_1^* a_2 + i a_2^* a_1. \end{aligned} \quad (\text{B10})$$

In general, the transverse component of the phonon is elliptically polarized with parameters α and φ defined in Fig. 5. These parameters can be expressed by the expectation values of the Stokes' parameters as follows:

$$\tan 2\varphi = P_2/P_1, \quad (\text{B11})$$

$$\tan \alpha = P_3/[P + (P^2 - P_3^2)^{1/2}]. \quad (\text{B12})$$

Exciton Complexes and Donor Sites in 33R SiC

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Polytype 33R, which, in some respects, may be considered as part 6H, part 15R, proves to be intermediate between those polytypes in many properties, among which are the exciton energy gap (3.003 eV), absorption strength, exciton binding energies (two kinds), and nitrogen donor ionization energies (0.15 to 0.23 eV). The latter three appear to be consequences of an intermediate value of electron effective mass. Nitrogen-exciton complexes (Lampert complexes) are observed in the low-temperature photoluminescence at ten of the eleven inequivalent donor sites. In this polytype, it is possible to assign spectral lines to particular sites, which we distinguish by a simple code. Both four-particle and three-particle complexes are observed, yielding phonon energies and, in addition, exciton binding energies which may be compared with those at corresponding sites in 6H and 15R. The comparison suggests that the 15R electron effective mass is about half that of 6H. For three-particle complexes, the effective-mass approximation appears to be inadequate. Thermally excited states are observed for both kinds of complexes. 33R is a member of a special series of SiC polytypes, each of which may be characterized as part 6H, part 15R. Some properties of the luminescence of higher members of this series are predicted, and their large zones and conduction-band minima are discussed.

I. INTRODUCTION

FOR a study of donor properties, SiC has the advantage that the same donor (e.g., nitrogen) can be studied with slight changes in neighbor arrangements (at inequivalent sites in a polytype), or with slight changes in band structure^{1,2} (by comparing polytypes).

We are now reporting on 33R SiC, in which we have observed photoluminescence due to nitrogen-exciton

complexes (Lampert³ complexes) at ten of the eleven inequivalent donor sites. Such luminescence has previously been reported for the single site⁴ of cubic SiC, for two sites¹ in 4H, three sites^{5,6} in 6H, four (of a possible five)⁷ in 15R, and six (of a possible seven)⁸ in 21R, a total of 26 sites in six polytypes. Within a given

¹ Lyle Patrick, W. J. Choyke, and D. R. Hamilton, Phys. Rev. **137**, A1515 (1965).

² Certain band properties appear to be similar in all polytypes. For example, the valence-band maximum at $\mathbf{k}=0$ is thought to be affected very little by polytype changes, and the conduction-band maxima are thought to be always on mirror planes at the large zone boundary.

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⁸ D. R. Hamilton, Lyle Patrick, and W. J. Choyke, Phys. Rev. **138**, A1472 (1965).