

Thermal Transport By Coupled Magnons and Phonons in Yttrium Iron Garnet at Low Temperatures*

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(Received 22 December 1972)

It is shown that the magnetic field dependence of the thermal conductivity of yttrium iron garnet (YIG) can be accounted for if the coupled magnon-phonon modes are employed in calculating the conductivity. On the other hand, the use of independent magnon and phonon modes does not lead to satisfactory agreement between theory and experiment. Experimental results are presented for the magnetic field dependence of the thermal conductivity of YIG in the range of 0.23–1 K.

I. INTRODUCTION

Previous thermal-conduction measurements on yttrium iron garnet¹⁻³ (YIG) indicate that in the neighborhood of 1.0 K magnon conduction is as important as phonon conduction. The observed decrease in conductivity upon the application of a magnetic field is evidence that the magnon conductivity is appreciable in YIG at low temperatures. However, as pointed out by Douglass¹ and Friedberg and Harris² the simple noninteracting theory does not agree very well with the field dependence observed.

The present measurements represent an extension of the above work to lower temperatures and higher magnetic fields. As will be pointed out in Sec. II, the analysis of the data is much simplified by applying a magnetic field of sufficient strength to completely attenuate the magnon part of the conductivity.

It has been found that it is impossible to fit the data we have obtained if the magnon-phonon interaction is ignored. On the other hand, if the interaction is included in the sense that the coupled magnon-phonon modes are employed for the normal modes of the system, we will show that good agreement between calculated and measured conductivities may be achieved.

Measurements were made in fields up to 40 kOe over the temperature range 0.23–1.0 K. Over this entire temperature range the conductivity reached its minimum value for fields well below the maximum field.

We can argue, therefore, that the high-field conductivity is due only to phonon conduction. The magnon states have been driven to sufficiently high energies so that they are no longer thermally excited, and hence do not contribute to the conductivity. Thus the phonon conductivity can be subtracted out in a direct way.

If one assumes no magnon-phonon interaction

then the total conductivity is just

$$K = K_{ph} + K_m, \quad (1)$$

where K_{ph} is the phonon conductivity and K_m is the magnon conductivity. If H_s is the value of the magnetic field, above which the magnons are completely attenuated, then

$$K_{ph} = K(H_s) \quad (2)$$

and

$$K_m(H_i) = K(H_i) - K(H_s). \quad (3)$$

The magnon conductivity can be calculated separately and compared to current theories of magnon conductivity.

If, on the other hand, the magnon-phonon interaction is not zero, then one obtains coupled magnetoelastic modes which will affect the total conductivity. In this case, the high-field conductivity is still the pure-phonon conductivity; however, Eqs. (1) and (3) are no longer valid, and the separation at intermediate fields is no longer possible. It is expected that the effect of the interaction is to reduce the magnitude of the conductivity at intermediate fields. That this is indeed the case in YIG will be demonstrated.

II. THERMAL CONDUCTION IN FERROMAGNETIC INSULATORS

A. Noninteracting Theory

In this case Eq. (1) is valid and one has only to use an appropriate form for K_{ph} and K_m . We may start with the expression for the thermal conductivity obtained from a Boltzmann equation⁴

$$K = \sum_k V(k) l(k) \hbar \omega(k) \frac{\partial N(k)}{\partial T}, \quad (4)$$

where $V(k)$ is the group velocity, $N(k)$ is the distribution function for the carriers, and $l(k)$ is the

mean free path of carriers of wave vector \vec{k} . If $\omega(k)$ is independent of temperature then $\hbar\omega(k)\partial N(k)/\partial T$ is just the specific heat.

Transforming this sum to an integral, assuming isotropy, and averaging over all angles between k and the heat-flow direction, Eq. (4) becomes

$$K = \frac{1}{6\pi^2} \int_0^{\omega_m} \left(\hbar\omega \frac{\partial N(\omega)}{\partial T} \right) l(k) k^2(\omega) d\omega. \quad (5)$$

The form of Eq. (5) is obtained by using $V(k) = \partial\omega/\partial k$ and $d^3k = k^2(dk/d\omega) d\omega d\Omega$. Thus in the present form the equation is valid for both phonons and magnons. It will be particularly useful in this form later when, because of the magnon-phonon interaction, the dispersion relations cannot be expressed in simple analytic form.

In the present case of zero coupling (5) can be put in a much simpler form for both phonons and magnons: below 1.0 K in YIG the mean free path for both phonons and magnons will be boundary limited in a sufficiently perfect crystal, since the intrinsic phonon-phonon, as well as the magnon-magnon mean free paths are many times the dimensions of the crystal.

For phonons in the acoustic limit, $\omega = Ck$, where k is the phonon wave vector, and C the velocity of sound.

If we assume that C is an average over the transverse and longitudinal modes, we obtain

$$K_{ph} \propto T^3 \int_0^{\infty} x^4 \text{csch}^2(\frac{1}{2}x) \tau_{ph}(x) dx, \quad (6)$$

where $x = \hbar\omega/k_B T$ and $\tau(x) = l(x)/C$ is the total relaxation time for momentum destroying processes. The upper limit of integration is actually Θ_D/T but since $T \ll \Theta_D$, in the present case, no serious error occurs from extending the integration to infinity.

For ferromagnetic magnons in zero field, if one assumes $\hbar\omega = \alpha k^2$, Eq. (5) reduces to

$$K_m \propto T^2 \int_0^{\infty} x^3 \text{csch}^2(\frac{1}{2}x) l_m(x) dx. \quad (7)$$

If the internal field is H_i then $\hbar\omega = \alpha k^2 + g\mu_B H_i$ and Eq. (7) is modified. We have

$$k^2(\omega) = (\hbar\omega - g\mu_B H_i)/\alpha = (k_B T/\alpha)(x - h), \quad (8)$$

where $h = g\mu_B H_i/k_B T$. Referring to Eq. (5) we must replace $l(k)$ by $l(x - h)$ and the lower limit of integration becomes $g\mu_B H_i/k_B T$. Thus,

$$K_m(H_i) \propto T^2 \int_h^{\infty} (x - h) x^2 \text{csch}^2(\frac{1}{2}x) l_m(x - h) dx. \quad (9)$$

In this expression the scattering has been left in terms of the mean free path in order to eliminate the group velocity which is a complicated function of x , for magnons.

In a sufficiently high-quality crystal if $T \ll \Theta_D$, T_c , one would expect that the dominant scattering processes are those due to the boundary and point de-

fects. Thus,

$$\tau_{ph}^{-1}(x) = \tau_B^{-1} + \tau_D^{-1}(x) \quad (10a)$$

and

$$l_m^{-1}(x) = l_B^{-1} + l_D^{-1}(x), \quad (10b)$$

where τ_B^{-1} is the inverse relaxation time for boundary scattering and τ_D^{-1} is the inverse relaxation for point-defect scattering. Point-defect scattering of phonons has been treated by several authors (see, for instance, Carruthers⁴) and can be expressed as $\tau_D^{-1}(k) = d'k^4 = dT^4 x^4$ where $d = d'(k_B/\hbar C)^4$ if a simple Rayleigh law is assumed. Thus Eq. (10a) becomes

$$\tau_{ph}^{-1}(x) = C/L + dT^4 x^4 = (C/L)[1 + a(T)x^4], \quad (11)$$

where $a(T) = (Ld/C)T^4$. In the above equation we have put $\tau_B^{-1} = C/L$ where L is the Casimir length, which is of the order of the smallest dimension of the crystal.

Callaway⁵ and Callaway and Boyd⁶ have treated the case of scattering of magnons by magnetic defects. If resonance scattering is neglected they find that $l_D^{-1} = g'k^4 = gT^2 x^2$, where $g = g'(k_B/\alpha)^2$. If the internal field is not zero then $l_D^{-1} = gT^2(x - h)^2$. Thus Eq. (10b) becomes

$$l_m^{-1}(x) = L^{-1} + gT^2 x^2 \\ = L^{-1}(1 + b(T)x^2) \text{ for } H_i = 0, \quad (12a)$$

and

$$l_m^{-1}(x - h) = L^{-1}[1 + b(T)(x - h)^2] \text{ for } H_i \neq 0. \quad (12b)$$

Using Eqs. (11), (12a), and (12b) we can write Eqs. (6), (7), and (9) as

$$K_{ph} = A'T^3 \int_0^{\infty} \frac{x^4 \text{csch}^2(\frac{1}{2}x) dx}{1 + a(T)x^4}, \quad (13)$$

$$K_m(O) = B'T^2 \int_0^{\infty} \frac{x^3 \text{csch}^3(\frac{1}{2}x) dx}{1 + b(T)x^2}, \quad (14)$$

$$K_m(H_i) = B'T^2 \int_h^{\infty} \frac{x^2(x - h) \text{csch}^2(\frac{1}{2}x) dx}{1 + b(T)(x - h)^2}. \quad (15)$$

The leading term in the integrand of Eq. (15) for $x \gtrsim 1$ is e^{-x} , hence $K_m(H_i)$ should decrease approximately exponentially with field. In Sec. VI, where the experimental results are compared with the noninteracting theory outlined above, it is shown that Eq. (15) predicts a somewhat slower decrease in K_m with field than is observed. It will be shown below that it may be possible to explain this behavior if the effects of magnon-phonon interactions are taken into account.

B. Magnon-Phonon Interaction

The magnetoelastic coupling of magnons and phonons in a ferromagnetic dielectric was treated by Kittel⁷ using a classical field approach. In the limit

it of long wavelengths the results of the classical treatment should be identical to a quantum theory if the interaction is due primarily to magnetostriction. This theory ignores higher-order interactions which have been considered by Kittel and Abrahams.⁸ These may be important,⁹ but constitute a refinement that will not be considered here.

The dispersion relations obtained by Kittel⁷ are in the form of a cubic equation for ω in terms of the wave vector k :

$$(\omega - \omega')(\omega^2 \rho - C^2 k^2) + \omega_s k^2 (B_2/M_s)^2 = 0, \quad (16)$$

where

$$\omega' = H_i + (2\gamma A/M_s)k^2, \quad \omega_s = \gamma M_s,$$

and where γ is the gyromagnetic ratio, M_s is the saturation magnetization, and B_2 is the magnetoelastic coupling constant. From Eq. (16) it is seen that when $B_2 = 0$, one obtains the usual uncoupled phonon and magnon dispersion curves.

Yttrium Iron Garnet (YIG)

In recent years there has been a rather exhaustive investigation of the magnetic properties of a class of ferromagnetic materials known as the rare-earth garnets. They form compounds of general formula $5\text{Fe}_2\text{O}_3 \cdot 3M_2\text{O}_3$, where M is a trivalent rare-earth ion, or the nonmagnetic ion Y^{3+} . An excellent review of the early work on the rare-earth garnets is given by Néel *et al.*¹⁰

In yttrium iron garnet, since the ion is nonmagnetic, the magnetic properties are entirely due to the Fe^{3+} ions. The unit cell has cubic symmetry¹¹ and contains four formula units of garnet whose positive ions occupy three types of sites. The Fe^{3+} ions occupy the 16 a sites at the center of a tetrahedron of oxygen ions and the 24 d sites at the center of an octahedron of oxygen ions. The M^{3+} ions occupy the 24 c sites at the center of a distorted cube of oxygen ions. There is a strong antiferromagnetic coupling between the ions on the a sites and the d sites.

The equilibrium thermal and magnetic properties of YIG have been reported by a number of authors.¹²⁻²⁰ The complete spin-wave spectrum has been calculated by Harris.²¹ At low temperatures only the acoustic mode will be populated and for long wavelengths neglecting demagnetizing effects and anisotropy, the dispersion relation is given by $\hbar\omega = \alpha k^2$. Thus the acoustic spin-wave modes are identical to ferromagnetic modes.

It is of interest to note that Eq. (16) predicts, for $H = 0$, that the main deviation of the coupled modes from the unperturbed phonon and magnon modes occurs near the crossover frequencies. For YIG there are two in zero field but only the upper one at $\epsilon_0 = C^2 \hbar^2 / \alpha$ is important here. For YIG this is equivalent to approximately 9 K. Hence, below 1

K it is expected that the thermal conductivity in zero field will not be appreciably affected by the magnetoelastic coupling. The total conductivity, for $H = 0$ at $T < 1$ K is just $K_T = K_{ph} + K_m$, where K_{ph} and K_m are calculated using Eqs. (6) and (7).

This of course ignores the possible effect of higher-order interactions such as one-phonon-two-magnon interactions.⁸ It is obvious that these exist, otherwise it would be impossible for heat to enter the magnon system from the phonon system at $H = 0$ and $T \sim 1$ K. Nevertheless it is not necessarily inconsistent to ignore the damping due to this interaction since it can be shown⁹ that the mean free path for magnon-phonon interaction, such that sufficient heat will flow into the magnon system, is of the order of the length of the crystal.

For nonzero fields the unperturbed modes cross at two points, one at $\epsilon_1 \cong \mu H$ and one at $\epsilon_2 \cong \epsilon_0$. As the field is increased ϵ_1 increases while ϵ_2 decreases, until for some field $\epsilon_1 = \epsilon_2$. Above this field the magnon mode no longer intersects the phonon mode. Equation (16) gives the behavior of the coupled modes in the presence of external fields according to the classical theory of Kittel.⁷

The thermal conductivity in the presence of an external field can be calculated by solving Eq. (16) for the two coupled modes and using these solutions in an integral of the form of Eq. (5).

III. EXPERIMENTAL METHODS

The sample used in these studies was obtained from Dr. Van Uitert of Bell Laboratories. It was cut into rectangular shape 5.0 mm by 3.9 mm about 20 mm long.

The present measurements were carried out in a modification of the He^3 cryostat described earlier by Walton.²² Temperatures between 0.2 and 1.0 K were obtained by evaporation of liquid He^3 . The sample, which was thermally attached to the He^3 refrigerator was situated in the center of a 40-kOe superconducting solenoid.

Absolute temperatures were determined by measuring the vapor pressure of liquid He^3 (1958 He^3 scale) contained in a separate bulb thermally attached to the He^3 refrigerator. A Texas Instrument quartz pressure gauge was used to determine the vapor pressure. Speer, nominally 470 Ω , carbon resistors were used as secondary thermometers to measure the temperature and temperature gradients along the sample.

The calibration of the carbon resistors below 0.6 K was achieved by comparison with the magnetic susceptibility of a cerium manganese nitrate (CMN) salt pill. The thermal conductivity was determined by a standard steady-state technique. One end of the sample is thermally attached to the He^3 refrigerator by a phosphor-bronze clamp. A heater is similarly attached to the lower end of the sample.

Two carbon resistors are attached at intermediate points along the sample. To avoid large magneto-resistance corrections for the resistors, they are placed well out of the solenoid region, but thermally connected to the clamps by large copper wires.

The resistance was determined by a dc potentiometric technique. With a constant, equal current in both resistors, the potential across one resistor was measured as well as the difference in potential across the two resistors. The former measurement served to monitor the absolute temperature at that point on the sample. The latter measurement then determined the temperature gradient. Thus simultaneous measurements of the temperature and the gradient were made. At all temperatures the resistor current was adjusted to produce negligible self-heating.

IV. RESULTS AND COMPARISON WITH THEORY

The temperature dependence of the thermal conductivity in zero external field, as well as at 40 kOe, is shown in Fig. 1. On the assumption that the magnon and the phonon contributions to the conductivity are additive at zero field, the 40-kOe results have been subtracted from the zero-field results to yield the zero-field values of the magnon conductivity. These have also been displayed in Fig. 1.

Both the phonon and the magnon conductivities are seen from Fig. 1 to increase with temperature,

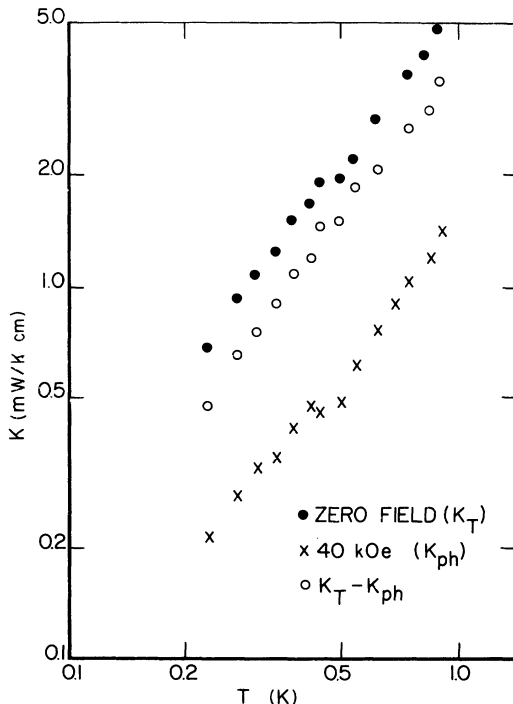


FIG. 1. Thermal conductivity of YIG as a function of temperature.

within the temperature range indicated, roughly as T^n , where $n \cong 1.5$. It is clear, therefore, that neither the magnon nor the phonon mean free paths are boundary limited, since one would expect $n = 2.0$ and 3.0 , respectively, if the main scattering was with the boundaries.

When magnetic defect scattering was included, a reasonable fit to the magnon conductivity was obtained with Eq. (14), i. e.,

$$K_m(o) = B'T^2 \int_0^\infty \frac{x^3 \operatorname{csch}^2(\frac{1}{2}x) dx}{1 + b(T)x^2},$$

where

$$B' = 1.54 \text{ mW cm}^{-1} \text{ K}^{-3}, \quad b(T) = 0.30T^2.$$

An attempt to fit the high-field phonon conductivity over the entire temperature range of the experiment was not very successful with the form given by Eq. (13), which includes only boundary plus point-defect scattering. It was found that a reasonable fit could be obtained if the effect of resonance scattering was included. The best fit was obtained by assuming the inverse relaxation time for resonance scattering could be expressed as

$$1/\tau_r \sim T^2 \omega^2 / (\omega^2 - \omega_0^2)^2, \quad (17)$$

as first suggested by Pohl.²³ With the addition of Eq. (17) to the inverse relaxation time in the denominator of Eq. (13) the phonon conductivity could be satisfactorily fitted by the relation

$$K_{ph} = AT^3 \int \frac{x^4 \operatorname{csch}^2(\frac{1}{2}x) dx}{1 + a(T)x^4 + Gx^2 / (\omega_0^2 - x^2)^2}, \quad (18)$$

where

$$A = 0.98 \text{ mW cm}^{-1} \text{ K}^{-4}, \quad a = 1.45 \times 10^{-2} T^4, \\ G = 40, \quad \omega_0 = 2.61 \times 10^{11} \text{ Hz}.$$

It should be emphasized that we attach no significance whatsoever to the resonance term. It is simply a device to reproduce the phonon-thermal-conductivity data. Our main interest is in the variation of the conductivity with magnetic field, and thus the phonon conductivity simply contributes a field-independent background.

With the zero-field temperature dependence of the phonon and magnon conductivity apparently accounted for, an attempt to understand the field dependence of the data was made. If the magnon-phonon interaction is neglected, the total conductivity at a field H is just the sum of Eqs. (15) and (18), where the constants in Eq. (15) are those found from the fit to the zero-field magnon conductivity. A comparison with the experimental results at three different temperatures is shown in Figs. 2(a), 2(b), and 2(c). The calculated conductivities are shown as the solid lines.

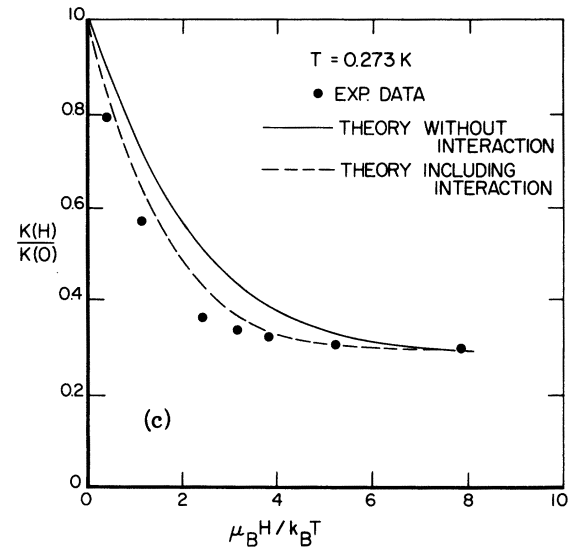
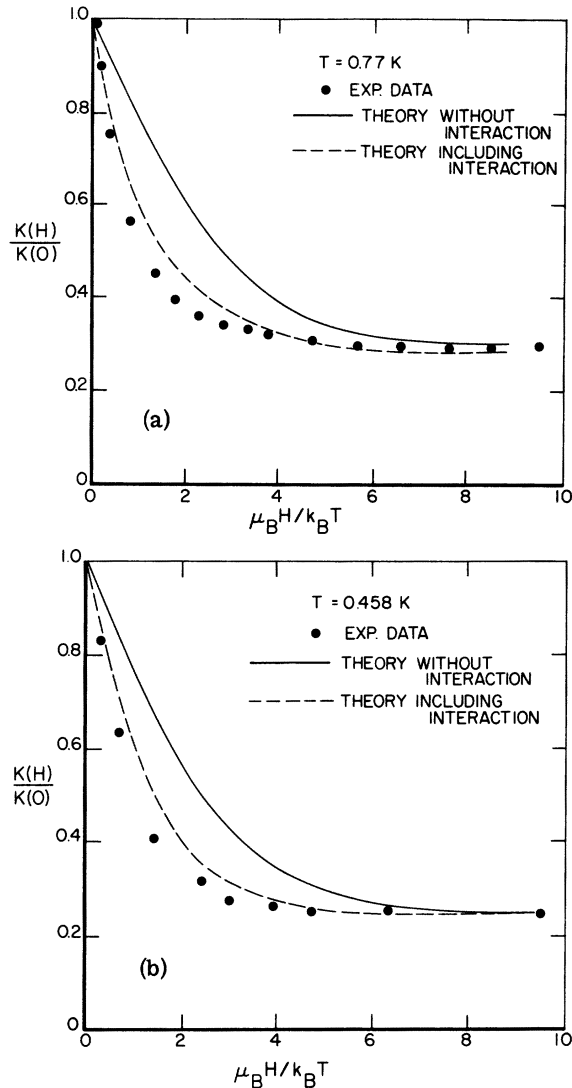


FIG. 2. Change in conductivity with magnetic field at three different temperatures. The solid line was calculated ignoring the interaction between the magnon and phonon mode. The dashed line was calculated using coupled magnetoelastic modes.

by Eq. (16). These coupled modes differ significantly from the noninteracting modes only in the neighborhood of the region where the energy and wave vector of the phonons and magnons are equal. The lower mode is phononlike for energies below the distorted region, and magnonlike above the distorted region. The reverse is true for the upper mode. There is another distorted region at higher energies, with another reversal of character of each mode.

Since there is no simple analytic form for the dispersion relations of the coupled system, it is necessary to return to Eq. (5) in order to calculate the thermal conductivity. The contributions from each of the two elastomagnon modes are summed to give the total conductivity. There is, however, the problem of what to use for the mean free path $l(k)$ in Eq. (5). Approximate expressions for both the phonon and the magnon scattering were previously determined by fitting the temperature dependence of the data. Thus in the magnonlike regions those found from Eq. (14) were used, and in the phononlike region those found from Eq. (20) were used.

Using this technique the field dependence of the conductivity was again calculated with the results displayed by the dashed line in Figs. 2.

The magnetoelastic coupling constants for YIG are temperature dependent. Using a parallel-pumping technique Nilsen, Comstock, and Walker²⁵ have measured B_2 at 4.2 K and obtain a value of 4×10^7 erg/cm³. A higher value of 10^8 erg/cm³ gives a better fit to our data and was used to obtain

The data presented in these, as well as previous figures, have been approximately corrected for demagnetizing effects using the method of Joseph and Schlomann²⁴ which is applicable to nonellipsoidal bodies. The field parameter in all these cases is then appropriately the internal field. At intermediate fields it is seen that the experimental points lie below the predicted values. This is just the expected result, since the magnon-phonon interaction, if important, will interfere with both the magnon and the phonon transport, and thus reduce the total conductivity. It is clear therefore, that the effects of the magnon-phonon interaction must be taken into consideration, and the coupled modes must be used.

If the interaction is included directly in the Hamiltonian describing the system, the noninteracting phonon and magnon modes are distorted producing elastomagnon modes. The modes of the coupled system have dispersion relations which are given

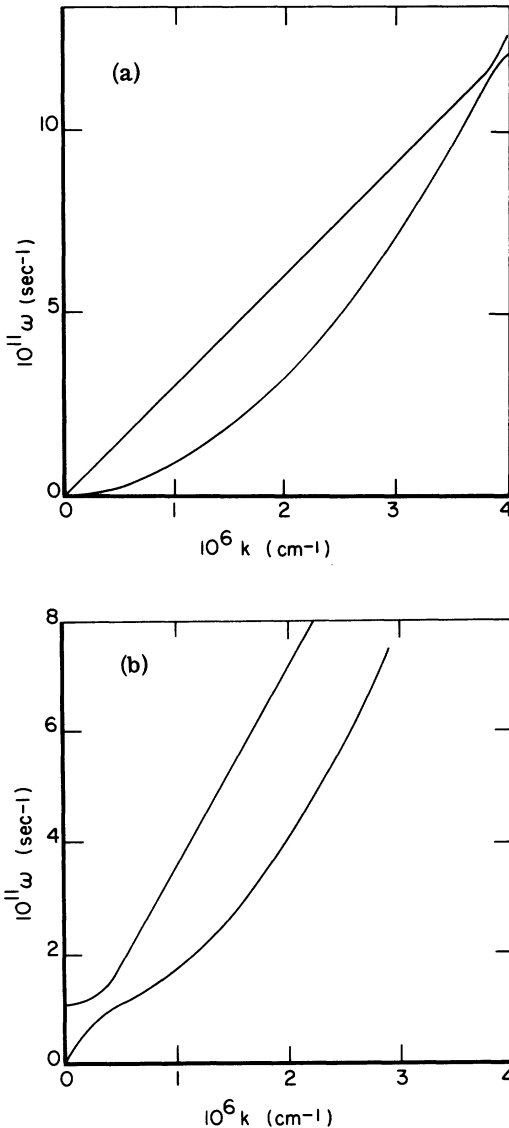


FIG. 3. Dispersion relation for coupled magnon-phonon mode calculated with $B_2 = 10^8 \text{ erg/cm}^3$ (a) for $H_i = 0$, (b) for $H_i = 5 \text{ kOe}$, (c) for $H_i = 40 \text{ kOe}$.

action is not restricted to modes of the same energy and wave vector. Inclusion of this interaction would tend to diminish the magnitude of B_2 necessary to obtain agreement between the calculated and measured curves.

V. SUMMARY

It has been shown that the thermal conductivity of YIG at low temperatures, calculated under the assumption of noninteracting magnon and phonon modes, does not agree with the experimental data at intermediate fields. However, comparison with the noninteracting theory does point to several important aspects of the data.

Comparison with the theory was done by first assuming that the high-field conductivity was due to phonons only, and then normalizing the magnitude of the magnon conductivity at zero field. The fact that the qualitative behavior of the conductivity, and particularly the field at which the conductivity saturates is correctly predicted by the theory, is strong evidence that direct conduction by the magnons is observed. However quantitative agreement with experiment is lacking.

The fact that the observed conductivity decreases faster with increasing field in the intermediate region of field, suggests that a mutual damping of phonons and magnons due to a phonon-magnon interaction is important. The effect of the interaction is primarily to produce coupled modes. When these were included, using the semiclassical theory of Kittel⁷ they accounted for this behavior within experimental error.

the results shown in Fig. 2. The value of $B_2 = 4 \times 10^7 \text{ erg/cm}^3$ would yield a correction to the uncoupled theory about $\frac{1}{3}$ as large as the one we show. The difference between our value and that obtained by Nilsen *et al.*²⁵ could reflect not only the effect of the lower temperature employed but also that of static strains to which the thermal conductivity would be much more sensitive than parallel pumping. Dispersion relations for the coupled modes calculated with this value of B_2 are shown in Fig. 3.

Higher-order magnon-phonon interactions have not been taken into account. The most important term in the Hamiltonian leading to these effects is linear in the phonon, but quadratic in the magnon operators. Thus it leads to a phonon scattering process in which one phonon interacts with two magnons, and is nonresonant in nature; i.e., the inter-

- *Experimental work performed at Oak Ridge National Laboratory, Oak Ridge, Tenn.
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PHYSICAL REVIEW B

VOLUME 8, NUMBER 3

1 AUGUST 1973

Effect of Pressure on the Kondo Temperature of Cu:Fe—Existence of a Universal Resistivity Curve

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(Received 22 January 1973)

The electrical resistivity of Cu-110-ppm Fe has been measured for temperatures from 1.3–40 K at pressures to 82 kbar. The Kondo temperature T_K is observed to increase with pressure, having doubled in value by 82 kbar. When plotted versus T/T_K , the spin-resistivity curves are found to accurately overlap for all measured pressures, thus confirming the existence of a universal resistivity curve $\rho = \rho(T/T_K)$ for Cu:Fe. Within the experimental uncertainty of 1.7%, the saturation value of the resistivity $\rho(T = 0 \text{ K})$ does not change with pressure. This indicates that to this accuracy the spin S and the potential scattering at the magnetic impurity remain constant. From the pressure dependence of T_K one obtains the volume dependence of the effective exchange parameter J_{eff} . Approximately the same volume dependence is found for a series of CuAu:Fe alloys using their known average atomic volume. The Cu:Fe high pressure and the CuAu:Fe-alloy data are discussed within the context of a simple Fermi-gas model based on the Schrieffer-Wolff transformation. The pressure dependence of the resistivity of pure copper, ρ_{phonon} , has also been studied and can be understood using the Bloch-Grüneisen formula with known values of the compressibility and the Grüneisen parameter. A method for experimentally determining deviations Δ from Matthiessen's rule in Kondo alloys is also presented. Such deviations can be very large; in fact, for $T < 30 \text{ K}$, we find that $\Delta = 1.3\rho_{\text{phonon}}$.

I. INTRODUCTION

Because of its importance to the general problem of magnetism in metals, the problem of localized magnetic states in a metallic environment has for many years been an object of considerable study. Investigations on dilute magnetic alloys have been pursued with particular intensity following Kondo's success in 1964 in explaining the resistance minimum anomaly.¹ Kondo postulated an s - d exchange interaction $\mathcal{H}_{s-d} = -J_{\text{eff}} \vec{s} \cdot \vec{S}$ between conduction elec-

trons with spin \vec{s} and magnetic impurities with spin \vec{S} and calculated the scattering to third order in perturbation theory. For antiferromagnetic (negative) values of the effective exchange parameter J_{eff} , the resultant resistivity is found to increase logarithmically with decreasing temperature. This is in approximate agreement with measurements on a large number of dilute magnetic alloys.^{2,3} Such a logarithmic divergence is, of course, not possible for arbitrarily low temperatures, as the resistivity at $T = 0 \text{ K}$ cannot exceed the unitary lim-