Scattering of Magnons and Phonons in the Thermal Conductivity of Yttrium Iron Garnet*

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Expressions have been derived for magnon-phonon scattering relaxation times for different cases as a function of temperature and wave vector using the theory of Sinha and Upadhyaya. The appropriate relaxation time is used in the determination of the effective relaxation time for magnons, and the thermal conductivity of yttrium iron garnet has been calculated in the temperature range 0.5 to 20°K using a quadratic dispersion law for magnons and a Debye spectrum for phonons.

I. INTRODUCTION

IN insulators the transport of energy takes place through phonons, and heat conduction is adequately described by the interaction between the phonons, impurities, and imperfections. In ferromagnetic insulators at sufficiently low temperatures magnons can also act as the carrier of heat energy. This possibility was investigated by Sato[1] in 1955. Assuming that there is no interaction between magnons and phonons and that at sufficiently low temperatures the mean free paths of the magnons and phonons are boundary-limited and comparable in magnitude, Sato[1] found that the thermal conductivity due to magnons varies as T^2 and is greater than phonon thermal conductivity, which varies as T^3. Douthett and Friedberg[2] studied the low-temperature thermal conductivities of ferrite crystals, which behave like insulators at liquid-hydrogen and liquid-helium temperatures and for which the dispersion relation for the spin-wave spectra for the important branch at low temperatures is similar to that of a ferromagnet. Using a magnon dispersion law $\hbar\omega_\lambda = Dk_\lambda^2 + g\beta H$, where the second term is the Zeeman term, they showed that the magnon contribution to thermal conductivity is given by

$$K_m(H) = \frac{l_s k_B^3 T^2}{3\pi h D} \sum_{n=1}^{\infty} \left[\left(\frac{\beta g H}{k_B T} \right)^2 \frac{1}{n} + \left(\frac{4\beta g H}{k_B T} \right) \frac{1}{n^2} + \frac{6}{n^3} \right] e^{-n g \beta H / k_B T}, \quad (1)$$

which, for $H=0$, reduces to

$$K_m(0) = 0.765 l_s k_B^3 T^2 / hD. \quad (2)$$

Here l_s is the magnon free path and is assumed to be constant. For a constant phonon free path l_p, the phonon conductivity is given by

$$K_{ph}(0) = 326 k_B^4 l_p T^3 / h^3 v_S^2, \quad (3)$$

where v_S is the average sound velocity.

If it is assumed that $l_p = l_s$ and one takes $v_S = 5\times10^5$ cm/sec, $D \simeq 10^{-28}$ erg cm^2, the thermal conductivity at 2°K in a field of 24 kOe is expected to be reduced by $\Delta k/k_0 \simeq -0.3$. Friedberg and Harris[3] studied thermal conductivities of single crystals of rods of yttrium iron garnet (YIG) in the temperature range 1.3 to 40°K and in an external field varying from 0 to 24 kOe. They found that if the magnetic field of 24 kOe is applied parallel to the [100] or [111] axis, the reduction in the thermal conductivity in the liquid-helium region is of the same order as mentioned above. Below 6°K, Friedberg and Harris showed that the thermal conductivity is well represented by $K(0) = AT^2 + BT^3$, where the first term represents the contribution due to magnons, and the second, the contribution due to phonons. The constants A and B, which are identified as $A = 0.765 l_s k_b^3/h_0$ and $B = 326 k_b^4 l_p / h^3 v_S^2$, are known from the extrapolated intercept and the slope of the plot $K(0)/T^2$ versus T. Taking $v_S = 5\times10^5$ cm/sec, $D = 0.83\times10^{-28}$ erg cm^2, they obtained $l_s = 6.0\times10^{-3}$ cm and $l_p = 5.8\times10^{-3}$ cm. The assumption that the field affects only the magnon contribution was tested by calculating $K_m(H)/T^2$ with the help of Eq. (1) and using $l_s = 6.0\times10^{-3}$ cm as obtained above, and then plotting $K(H)/T^2 - K_m(H)/T^2$ versus T, where $K(H)$ is the thermal conductivity in the presence of magnetic field. This plot is a straight line with slope $K(0)/T^2$ and zero intercept, which suggests strongly that $K(H) - K_m(H) = K_{ph} = K_{ph}(0)$ and that K_{ph} is unaffected by the field.

Similar results were also obtained by Luthi[4] and Douglass.[5] The magnon contribution has been also detected in EuS.[6]

The above analysis is, however, based on the neglect of the magnon-phonon interaction, and the magnons or phonons are assumed to be scattered only by the grain or specimen boundaries. At temperatures above the helium range the thermal conduction in YIG is mainly due to phonons. In the temperature region

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where the contributions of magnons and phonons are comparable, one cannot neglect the interaction of magnons and phonons. In the present paper we have obtained the expression for the frequency and temperature dependence of magnon-phonon scattering relaxation time from the theory of the magnon-phonon interaction discussed by Sinha and Upadhyaya.[7] We have used this relaxation time to explain the thermal conductivity of the YIG sample in the temperature range 0.4 to 20°K as studied by Douglass.[5] The measurements of Douglass strongly suggest that the thermal-conductivity results are sensitive to impurities, and hence the present analysis also incorporates the scattering of magnons and phonons by defects.

II. THEORY

At low temperatures, particularly in the liquid-helium range, magnons contribute significantly towards the thermal conductivity in a magnetic crystal. The total thermal conductivity may be assumed to be a sum of separate contributions of magnons and phonons:

$$K=K_{\mathrm{m}}+K_{\mathrm{ph}}. \tag{4}$$

A calculation of the magnon thermal conductivity has been proposed by Callaway[8] in a way analogous to the calculation of the phonon thermal conductivity. The magnon thermal conductivity K_{m} is given by

$$K_{\mathrm{m}}=\frac{k_B}{3(2\pi)^3\hbar^2}\int\left(\frac{E}{k_BT}\right)^2\frac{\exp(E/k_BT)}{[\exp(E/k_BT)-1]^2}\times(\nabla_k E)^2\tau(\mathbf{k}_\lambda)d\mathbf{k}_\lambda, \tag{5}$$

where k_B is the Boltzmann constant, T is the absolute temperature, and $E=E(\mathbf{k}_\lambda)$ is the energy of the spin wave of wave vector $\mathbf{k}_\lambda$. Here $\tau(\mathbf{k}_\lambda)$ is the effective relaxation time of the spin wave of wave vector $\mathbf{k}_\lambda$ and is given by

$$1/\tau(\mathbf{k}_\lambda)=1/\tau_{\mathrm{mb}}+1/\tau_{\mathrm{mm}}+1/\tau_{\mathrm{mp}}+1/\tau_{\mathrm{md}}, \tag{6}$$

where τ_{mb} is the relaxation time for the boundary scattering of magnons, τ_{mm} is the relaxation time for magnon-magnon scattering, τ_{mp} is the relaxation time for the magnon-phonon scattering, and τ_{md} is the relaxation time for the scattering of magnons due to magnetic defects.

The energy $E(\mathbf{k}_\lambda)$ is given by[9]

$$E(\mathbf{k}_\lambda)=(\epsilon_{k_\lambda}+g\beta H)\{1+\phi_k\sin^2\theta_k\}^{1/2}, \tag{7}$$

where H is the external magnetic field and ϵ_{k_λ} is given by

$$\epsilon_{k_\lambda}=2S\sum_n J_n-2S\sum_n J_n e^{i\mathbf{k}_\lambda\cdot\mathbf{R}_n}. \tag{8}$$

Here $\mathbf{R}_n$ is a direct lattice vector, J_n is an effective exchange integral which is same for all vectors $\mathbf{R}_n$ of the same type, θ_k is the angle between vector $\mathbf{k}_\lambda$ of the spin wave and the magnetization $\mathbf{M}$ of the specimen, and ϕ_k is given by

$$\phi_k=4\pi g\beta M/(\epsilon_{k_\lambda}+g\beta H). \tag{9}$$

The relaxation time for magnon-magnon scattering is given by[10]

$$\tau_{\mathrm{mm}}^{-1}=BT^{5/2}k_\lambda^2, \tag{10}$$

where

$$B=\frac{3\zeta(\frac{3}{2})av_{\mathrm{m}}}{Z\omega S^2}\left(\frac{3k_B}{2\pi JSZ\mu}\right)^{5/2}. \tag{11}$$

Here a is the lattice constant, v_{m} is the magnon velocity, Z is the number of nearest neighbors, S is the spin of the magnetic ion, J is the exchange integral between the neighboring magnetic ions, and ω and μ are given by

$$\begin{aligned}\omega&=(1,\ 3\times2^{-5/3},\ 2^{-1/3}),\\ \mu&=(1,\ 3\times2^{-4/3},\ 2^{1/3})\end{aligned} \tag{12}$$

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

The relaxation time due to scattering of magnons by magnetic defects is given by[7]

$$\tau_{\mathrm{md}}^{-1}=N_{\mathrm{md}}A_{\mathrm{m}}k_\lambda^4=N_{\mathrm{md}}A_{\mathrm{m}}(k_BT/D)^2x^2=A'T^2x^2, \tag{13}$$

where N_{md} represents the number of magnetic defects and $x=E_\lambda|_{k_BT}=Dk_\lambda^2/k_BT$. The parameter A_m is given by

$$A_{\mathrm{m}}=\frac{V_0^2v_{\mathrm{m}}}{4\pi^2}\left[\left(\frac{J'}{J}\right)^2\left(1-\frac{S'}{S}\right)^2+\frac{4}{3}\left(1+\frac{J'}{J}-2\frac{J'S'}{JS}\right)\right]. \tag{14}$$

Here V_0 is the atomic volume, v_{m} is the magnon velocity, S is the spin of the atom which constitutes the magnetic defect and is coupled to the nearest neighbors with an exchange integral J', and J and S refer to the host lattice.

III. RELAXATION TIME DUE TO MAGNON-PHONON SCATTERING

Sinha and Upadhyaya have given a theory of magnon-phonon interaction in magnetic crystals. The Taylor expansion of the crystal-field potential of the neighboring ions provides the perturbation which mixes some excited orbital states with the ground orbital states of the magnetic ions. Starting with the one-electron wave function defined in the presence of this perturbation, the total Hamiltonian is expressed in an occupation-number representation by the method of second quantization. Such a representation provides the interaction term of the Hamiltonian. Using the relationships between the conventional spin operators and the creation and annihilation operators of one-electron states, expressions are obtained for the magnon-phonon interaction term, which is further simplified by express-

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ing the relative displacements between the ions in terms of creation and annihilation operators of phonons, and using Holstein-Primakoff spin deviation and other operators.

The magnon-phonon interaction for the one-phonon process is given by

$$H_{\rm int}=\frac{4}{\sqrt{N}}\sum_{h\lambda qp} 2S\,{}^{\alpha}J(\mathbf{R}_h)[e^{i\mathbf{k}_\lambda\cdot\mathbf{R}_h{}^0}-e^{i(\mathbf{k}_\lambda-q)\cdot\mathbf{R}_h{}^0}+e^{-i\mathbf{q}\cdot\mathbf{R}_h{}^0}-1]\times\mathbf{G}_{qp}a_{\lambda-q}{}^{\dagger}a_\lambda(b_{qp}{}^{\dagger}-b_{-qp}), \quad (15)$$

where $\mathbf{R}_h=\mathbf{R}_l-\mathbf{R}_m=\mathbf{R}_{lm}$, and

$${}^{\alpha}J(\mathbf{R}_{lm})\equiv\sum_{\alpha}\frac{\langle\phi_\alpha\phi_m|G_{12}|\phi_m\phi_l\rangle\langle\phi_\alpha|\mathbf{V}^h|\phi_l\rangle}{(E_\alpha-E_l)}. \quad (16)$$

Here $G_{12}=1/r_{12}$, where r_{12} is the distance between the electrons 1 and 2, ϕ_l and ϕ_m represent the unperturbed ground-state wave functions for the magnetic ions l and m, α is the excited state, and $\mathbf{V}^h$ is given by

$$V=V_0+\sum_h\left(\frac{\partial V}{\partial\mathbf{R}_h}\right)_0\cdot\delta\mathbf{R}_h+\sum_{hh'}\frac{1}{2}\left(\frac{\partial^2V}{\partial\mathbf{R}_h\partial\mathbf{R}_{h'}}\right)\delta\mathbf{R}_h\delta\mathbf{R}_{h'}$$
$$=V_0+\sum_h\mathbf{V}^h\cdot\delta\mathbf{R}_h+\tfrac{1}{2}\sum_{hh'}V^{hh'}\delta\mathbf{R}_h\delta\mathbf{R}_{h'}, \quad (17)$$

where V_0 is the static crystal-field potential due to nearest-neighbor ions at the ith electron when it is localized at the ion at $\mathbf{R}_l{}^0$ and is given by $V_0=\sum_{m\neq l}v(\mathbf{r}_i-\mathbf{R}_m{}^0)$. The relative displacement $\delta\mathbf{R}_h$ between the ions l and m is given by

$$\delta\mathbf{R}_h=\frac{1}{\sqrt{N}}\sum_{qp}(-i)\mathbf{e}_{qp}\left(\frac{\hbar}{2\omega_{qp}M}\right)^{1/2}(b_{qp}{}^{\dagger}-b_{-qp})\times(e^{i\mathbf{q}\cdot\mathbf{R}_l{}^0}-e^{i\mathbf{q}\cdot\mathbf{R}_m{}^0})$$
$$=\frac{1}{\sqrt{N}}\sum_{qp}\mathbf{G}_{qp}(b_{qp}{}^{\dagger}-b_{-qp})(e^{i\mathbf{q}\cdot\mathbf{R}_l{}^0}-e^{i\mathbf{q}\cdot\mathbf{R}_m{}^0}). \quad (18)$$

Here $\mathbf{k}_\lambda$ is a magnon wave vector. In deriving the magnon-phonon interaction only normal processes have been assumed and the following momentum conservation relation has been used for the one-phonon process:

$$\mathbf{k}_\lambda'+\mathbf{q}-\mathbf{k}_\lambda=0. \quad (19)$$

Two-phonon Raman processes are found to be unimportant at low temperatures.

The relaxation time due to magnon-phonon scattering is given by

$$\tau_{\lambda qp}{}^{-1}\sim\frac{2\pi}{\hbar}|\phi_{\lambda qp}|^2\times\frac{e^{E_\lambda/k_BT}}{(e^{E_\lambda/k_BT}-1)(e^{E_{\lambda-q}/k_BT}-1)(e^{E_{qp}/k_BT}-1)}\times\delta(E_{\lambda-q}+E_{qp}-E_\lambda), \quad (20)$$

where

$$\phi_{\lambda qp}=\frac{4}{\sqrt{N}}\sum_h 2S\,{}^{\alpha}J(\mathbf{R}_h)\times[e^{i\mathbf{k}_\lambda\cdot\mathbf{R}_h{}^0}-e^{i(\mathbf{k}_\lambda-q)\cdot\mathbf{R}_h{}^0}-1+e^{-iq\cdot\mathbf{R}_h{}^0}]\mathbf{G}_{qp}. \quad (21)$$

Here it has been assumed that $\hbar\omega_{qp}\sim k_BT$ and that $\Delta T/T=(T_s-T_l)/T$ (where $T_s=T$) is a constant and independent of temperature. Using

$$(\partial V/\partial\mathbf{R}_h)\cdot\delta\mathbf{R}_h\sim|\partial V/\partial\mathbf{R}_h||\delta\mathbf{R}_h|,\quad|\mathbf{e}_{qp}|\sim1, \quad (22)$$

and

$$G_{qp}\simeq(\hbar/2\omega_{qp}M)^{1/2},$$

one obtains

$$\phi_{\lambda qp}\simeq\frac{4}{\sqrt{N}}\left(\frac{\hbar}{2\omega_{qp}M}\right)^{1/2}(i)(-2k_\lambda qa^2\cos\theta_{\lambda q})\times[2S\,{}^{\alpha}J(\mathbf{R}_h)] \quad (23)$$

from the δ-function relation

$$E_{\lambda-q}+E_{qp}-E_\lambda=0, \quad (24)$$

i.e.,

$$2k_\lambda q\theta_C k_B a^2(q|2k_\lambda-\cos\theta_{\lambda q}+\Theta_D/2\Theta_C k_\lambda a)=0,$$

where

$$\Theta_D=\hbar v_S/k_Ba.$$

Hence

$$\cos\theta_{\lambda q}=q/2k_\lambda+\Theta_D/2\Theta_C k_\lambda a. \quad (25)$$

The relaxation time is obtained in two special cases, $E_\lambda\ll k_BT$ and $E_\lambda>k_BT$.

Case I: $E_\lambda\ll k_BT$

$$\tau_{\lambda qp}{}^{-1}=\frac{64\pi S^2({}^{\alpha}J)^2}{NMv_S\Theta_C\Theta_D{}^2}\frac{qT^3}{k_\lambda{}^2}. \quad (26)$$

Case II: $E_\lambda>k_BT$, $E_q<k_BT$, $E_{\lambda-q}=E_\lambda-E_q<k_BT$

$$\tau_{\lambda qp}{}^{-1}=\frac{64\pi aS^2({}^{\alpha}J)^2}{NMv_S\Theta_C\Theta_D}\frac{q^2T^2}{k_\lambda{}^2}. \quad (27)$$

Since $\tau_{\rm mp}{}^{-1}(q)=\sum_{k\lambda}\tau_{\lambda qp}{}^{-1}$ and $\tau_{\rm mp}{}^{-1}(k_\lambda)=\sum_q\tau_{\lambda qp}{}^{-1}$, one obtains the following expressions for magnon-phonon relaxation times for the two cases:

Case I:

$$\tau_{\rm mp}{}^{-1}(q)=\frac{32S^2({}^{\alpha}J)^2\Omega k_m}{\pi Mv_S\Theta_C\Theta_D{}^2}qT^3,\qquad \tau_{\rm mp}{}^{-1}(k_\lambda)=\frac{8S^2({}^{\alpha}J)^2\Omega q_D{}^4}{\pi Mv_S\Theta_C\Theta_D{}^2}k_\lambda{}^{-2}T^3. \quad (28)$$

Case II:

$$\tau_{\rm mp}{}^{-1}(q)=\frac{32aS^2({}^{\alpha}J)^2\Omega k_m}{\pi Mv_S\Theta_C\Theta_D}q^2T^2,\qquad \tau_{\rm mp}{}^{-1}(k_\lambda)=\frac{32aS^2({}^{\alpha}J)^2\Omega q_D{}^5}{5\pi Mv_S\Theta_C\Theta_D}k_\lambda{}^{-2}T^2. \quad (29)$$

Here Ω is the volume of unit cell and k_m is the maximum value of the magnon wave vector $\mathbf{k}_\lambda$. Similarly, q_D represents the maximum wave vector for phonons.

IV. MAGNON AND PHONON CONTRIBUTIONS TO THE THERMAL CONDUCTIVITY OF YIG

A correct estimate of the magnon-phonon interaction and the scattering of magnons and phonons due to impurities and boundaries is necessary to give an accurate description of the low-temperature thermal conductivity of a real magnetic crystal. Unfortunately, the information about the nature and concentration of magnetic and nonmagnetic impurities and the quantities such as Θ_C, Θ_D, ${}^{\alpha}J$, and k_m is not available. However, an attempt has been made to study the temperature dependence of thermal conductivity of YIG by adjusting the relevant parameters and making the following simplified assumptions:

(1) A quadratic dispersion law ($E_\lambda = Dk_\lambda^2$) is taken for magnons, and phonons are assumed to have a Debye spectrum consisting of one (average) acoustic branch.

(2) For the calculation of magnon thermal conductivity, magnons are assumed to be scattered by boundaries, phonons, and impurities.

(3) For the calculation of phonon thermal conductivity the contribution due to scattering of phonons by magnons is considered negligible in the temperature range where phonon thermal conductivity is appreciable.

(4) For the scattering of magnons and phonons due to defects a Rayleigh-type scattering is assumed. Since the Rayleigh scattering parameter A is independent of temperature and frequency, calculations can be performed for different values of A'/A'' where A' and A'' correspond to magnons and phonons, respectively. Since the temperature dependence of the thermal conductivity is independent of the parameters A' and A'', they are taken to be the same for the sake of using a minimum number of adjustable parameters.

The magnon thermal conductivity can be expressed as

$$K_{\mathrm{m}} = \frac{D^{-1/2}k_B^{7/2}T^{5/2}}{3\pi^2\hbar^2}\int_0^\infty \frac{x^{7/2}e^x\tau(k_\lambda)dx}{(e^x-1)^2}, \qquad (30)$$

where

$$x = \hbar\omega_\lambda/k_BT = Dk_\lambda^2/k_BT.$$

The upper limit may be safely taken as infinity at the temperatures under consideration. The effective relaxation time $\tau(\mathbf{k}_\lambda)$ for magnons is given by

$$\tau^{-1}(\mathbf{k}_\lambda) = \tau_{\mathrm{mb}}^{-1}+\tau_{\mathrm{mp}}^{-1}+\tau_{\mathrm{md}}^{-1} = \beta(DTx)^{1/2}+(\alpha DT)x^{-1}+A'T^2x^2, \qquad (31)$$

where

$$\beta = 2l_b^{-1}k_B^{1/2}\hbar^{-1/2}$$

and

$$\alpha = \frac{32aS^2({}^{\alpha}J)^2\Omega q_D^5}{5\pi v_S M\Theta_C\Theta_D k_B}.$$

The phonon thermal conductivity K_{ph} is given by

$$K_{\mathrm{ph}} = CT^3\int[y^4e^y/(e^y-1)^2]\tau_{\mathrm{ph}}dy, \qquad (32)$$

where

$$C = (k_B/2\pi^2v_S)(k_B/\hbar)^3,$$

$$y = \hbar\omega_{qp}/k_BT,$$

and the different symbols have their usual meaning. The total relaxation time τ_{ph} is given by

$$\tau_{\mathrm{ph}}^{-1} = \tau_{pb}^{-1}+\tau_{pi}^{-1} = V_s/l_p+A''y^4T^4, \qquad (33)$$

where l_p is the relaxation length of phonons for boundary scattering, $y = E_{qp}/k_BT = \hbar\omega_{qp}/k_BT$, and $A'' = A(k_B/\hbar)^4$.

The experimental data for the thermal conductivity of YIG have been taken from Douglass, and sample 1 is considered for the present calculations. It is assumed that at 20°K the whole contribution to thermal conductivity comes from phonons. The parameter A, which is so adjusted that the calculated phonon thermal conductivity agrees with the experimental data at 20°K, is 9.797×10^{-42} sec^3.

According to Klemens, the scattering parameter A is given by

$$A = 12V_0^2S_a^2N_d/4\pi v_S^3, \qquad (34)$$

where V_0 is the atomic volume, N_d is the number of scattering centers per unit volume, v_S is the average phonon velocity, and S_a is a dimensionless scattering parameter related to the scattering cross section which includes terms corresponding to the deviation ΔM of the mass M, the force constant F_c, and the nearest-neighbor distance R_n. The expression for S_a^2 has the form

$$S_a^2 = a_1\left(\frac{\Delta M}{M}\right)^2+\left(b_1\frac{\Delta F_c}{F_c}+C_1\frac{\Delta R_n}{R_n}\right)^2. \qquad (35)$$

Using $M = 738.01\times1.67\times10^{-24}$ g for $Y_3Fe_2(FeO_4)_3$ and $\rho = 5.17$, one obtains $V_0 = 239\times10^{-24}$ cm^3. Substituting this value of V_0 and $v_S = 4.57\times10^5$ cm/sec in Eq. (34), one finds $N_dS_a^2 = 1.72\times10^{19}$ cm^{-3} for the product of point-defect concentration and the square of the scattering parameter S_a, which is related to the scattering cross section. If one takes $N_d = 10^{18}$ cm^{-3}, one finds $S_a^2 = 17.2$.

The scattering cross section σ can be expressed as $\sigma = 1/N_dl$, where N_d is the concentration of scattering centers and l is the corresponding mean free path. σ can be calculated if the matrix elements connecting the initial and scattered states are known, since the collision rate

$$\tau^{-1} = Nv_S\sigma = v_S/l. \qquad (36)$$

From Klemens's theory of point-defect scattering,

$$W_d/T = 3(2\pi)^3V_0^2N_dS_a^2/0.90hv_S^2, \qquad (37)$$

where W_d is the phonon thermal resistance at a temperature T due to point-defect scattering. If one writes in the Debye approximation

$$K=\tfrac{1}{3}C_{\rm ph}v^2\tau, \tag{38}$$

one has

$$W_d=\frac{3}{C_{\rm ph}v^2}\tau^{-1}=\frac{3}{C_{\rm ph}v_S^2}N_dv_S\sigma$$
$$=\frac{3(2\pi)^3V_0^2N_dS_a^2T}{0.90hv_S^2}, \tag{39}$$

which gives

$$\sigma=(2\pi)^2V_0^2S_a^2C_{\rm ph}T/0.90hv. \tag{40}$$

Using $C_{\rm ph}=142$ erg deg^{-1} cm^{-3} and $S_a^2=17.2$, as obtained previously for $N_d=10^{18}$ cm^{-3}, one obtains the scattering cross section $\sigma=17.16\times1.65\times10^{-21}$ cm^{-2}.

In the present calculations we have taken $A'=A''$ for the sake of minimizing the number of adjustable parameters. This means that

$$A(k_B/\hbar)^4=N_{\rm md}A_{\rm m}(k_B/D)^2.$$

If the phonon velocity is same as the magnon velocity, one obtains, after substituting for A and $A_{\rm m}$,

$$N_{\rm md}=\frac{192\pi^5S_a^2N_dD^2k_B^2}{h^4v_S^4\left[\left(\frac{J'}{J}\right)^2\left(1-\frac{S'}{S}\right)^2+\frac{4}{3}\left(1+\frac{J'}{J}-2\frac{J'S'}{JS}\right)\right]}.$$

Substituting $S_a^2N_d=1.72\times10^{19}$ cm^{-3}, $D=0.81\times10^{-28}$ erg cm^2, $v_S=4.57\times10^5$ cm/sec, $J'/J=0.1$, and $S'/S=\frac{1}{5}$, one finds $N_{\rm md}=1.05\times10^{18}$ cm^{-3}. This value of $N_{\rm md}$ is quite reasonable, which shows that $A'\sim A''$. A lower value of $N_{\rm md}$ would require that $A'<A''$.

The parameter α is adjusted so that at 2°K the total thermal conductivity $(K_{\rm m}+K_{\rm ph})$ agrees with the observed thermal conductivity. For the calculation of phonon thermal conductivity only scattering of phonons by boundary and point defect is considered, and phonon-phonon scattering, which is negligible near the conductivity maximum (about 20°K), is not considered for $T\leqslant20$°K. The present calculation reveals that although at 0.5°K the magnon contribution is as high as 46% of the total thermal conductivity, the phonon contribution in general dominates at temperatures greater than 0.5°K. Scattering of phonons by magnons is also negligible as far as the calculation of phonon thermal conductivity is concerned.

The value of α which has been obtained by adjustment is 1×10^{36} sec g^{-1} deg^{-1} cm^{-4}. The parameter α is proportional to the square of the derivative of the exchange interaction, ${}^{\alpha}J(\mathbf{R}_h)$, where ${}^{\alpha}J(\mathbf{R}_h)$ is given by

$${}^{\alpha}J(\mathbf{R}_h)=\sum_{\alpha}\frac{\langle\phi_\alpha\phi_m|G_{12}|\phi_m\phi_l\rangle\langle\phi_\alpha|(\partial V/\partial\mathbf{R}_h)_0|\phi_l\rangle}{(E_\alpha-E_l)}.$$

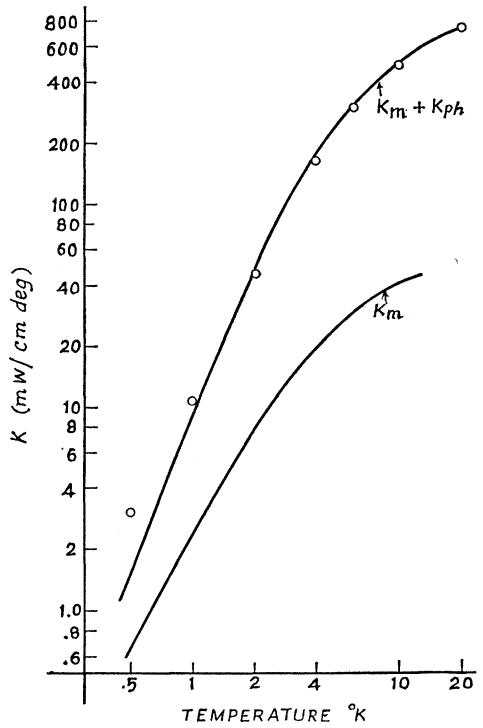

FIG. 1. Thermal conductivity of YIG. Circles represent the experimental results. Solid curves give the theoretical results obtained for the total thermal conductivity $K_{\rm m}+K_{\rm ph}$ and magnon thermal conductivity $K_{\rm m}$. Parameters for the theoretical curve: $l_{bm}=1.15\times10^{-2}$ cm; $l_{bp}=3.4\times10^{-2}$ cm; $V_S=4.57\times10^5$ cm sec^{-1}; $A'=A''=2.875\times10^3$ sec^{-1} deg^{-4}; $\alpha=1\times10^{36}$ sec g^{-1} deg^{-1} cm^{-4}; $D=0.81\times10^{-28}$ erg cm^2.

The factor ${}^{\alpha}J(\mathbf{R}_h)$ when expressed in terms of the experimentally adjusted parameter α is given by

$$({}^{\alpha}J)^2=\frac{5\pi\alpha v_SM\Theta_C\Theta_Dk_B}{32a^4S^2q_D^5},$$

which reduces to

$$({}^{\alpha}J)^2=\frac{5\pi\alpha v_S^6M\Theta_C\hbar^5}{32a^4S^2k_B^4\Theta_D^4},$$

where

$$q_D=k_B\Theta_D/\hbar v_S.$$

Substituting $\alpha=10^{36}$ sec g^{-1} deg^{-1} cm^{-4}, $v_S=4.57\times10^5$ cm/sec, $M=738.01\times1.67\times10^{-24}$ g, $\Theta_C=1000$°K (used by Sinha and Upadhyaya), $a=12.376\times10^{-8}$ c, $S=\frac{5}{2}$, $\Theta_D=538$°K, one finds ${}^{\alpha}J=1.27\times10^{-2}$ dyn. This value of ${}^{\alpha}J$ may be compared with the value of

$$\langle\psi_{3dz^2}|(\partial V/\partial\mathbf{R}_h)_0|\psi_{4pz}\rangle\sim1.25\times10^{-3}\text{ dyn}$$

which was obtained by Sinha and Upadhyaya for iron.

The results of calculation are shown in Fig. 1, and a good agreement betweeen theory and experiment has been obtained in the entire temperature range. The reasonable agreement between theory and experiment in the temperature range 0.5 to 2°K, where the magnon

contribution is appreciable, justifies the use of our expression for the temperature and wave-vector dependence of magnon-phonon relaxation time. In view of the fact that phonon contribution dominates over the magnon contribution at temperatures beyond 2°K, the accuracy of the magnon contribution to thermal conduction beyond 2°K cannot be checked; for example, the magnon contribution at 10°K is about 10% of the total conductivity.

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Optical Properties of Free Electrons in ZnO

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The optical properties of indium-doped ZnO crystals, with free-carrier concentrations ranging from 10^{19} to 5×10^{19} cm^{-3}, have been studied at room temperature. The condition of the crystal surfaces has been shown to influence radically the reflectivity in the near infrared. The optical effective mass for free electrons has been calculated from the dispersion in the near infrared to be $m_{opt}=(0.265\pm0.015)m$, which is in close agreement with other determinations. Experiments with polarized radiation revealed no measurable anisotropy of effective mass. The "polaron" effective mass in ZnO is estimated and briefly discussed.

I. INTRODUCTION

OPTICAL properties of semiconductors have been very instrumental in determining energy-band structures and effective masses. In particular, analysis of the exciton spectrum and fundamental absorption edge of zinc oxide (ZnO) has revealed that the minimum in the conduction band is very nearly spherical and at $k=0$.[1,2] The effective mass of electrons in the conduction band has been the subject of much discussion. Values as high as $0.5m$ have been reported for the density-of-states effective mass[3] and as low as $0.06m$ for the "inertial" effective mass from infrared reflectivity.[4] Hutson[5] has discussed the history of the effective mass of electrons in ZnO and has reached statisfactory agreement between most determinations with the possible exception of $0.06m$ reported by Collins and Kleinman.[4]

It is the purpose of this paper to report on the determination of the electron effective mass of ZnO from dispersion in the infrared due to free carriers. This work is similar to that by Collins and Kleinman,[4] but it results in more close agreement with other determinations.

II. SAMPLE PREPARATION AND EXPERIMENTAL APPARATUS

The single crystals of ZnO used in these experiments were grown from the vapor phase in these laboratories. The crystals were rough-cut to thin plates approximately 0.05-cm thick with an area of approximately 1 cm^2 to facilitate doping and optical measurements. Each was oriented so that the c axis was in the plane of the plate. Subsequent to the doping and optical measurements the crystals were cut to dimensions appropriate to electrical measurements.

The free-carrier electron concentrations of the plates were controlled by doping with indium at various equilibrium temperatures, as discussed by Thomas.[6] The time required for uniform doping was, in general, about twice as long as that calculated for simple diffusion in a thin plate using the diffusion rates given by Thomas.[6] The progress of the diffusion was intermittently checked by visually observing, with a microscope, gradients in transmission of red light through the crystals parallel to the crystal plate faces. These gradients in transmission were due to gradients in the free-carrier adsorption, which was appreciable in the red portion of the spectrum for high carrier concentrations. Because the absorption coefficient is proportional to the concentration of free carriers,[7] transmission gradients give a rough estimate of the uniformity of doping. When no visual non-uniformities of doping could be observed, the crystals were mechanically polished for further optical and electrical measurements. The uniformity of doping was further checked by repeated optical and electrical measurements on crystals for which the thickness had been substantially reduced by grinding.

The free-carrier concentrations were determined from

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