

very small necks on a multiply-connected surface. The saturation value of the magnetoresistance ratio extrapolated from curve a of Fig. 5 and referred to  $\rho_{\min}$  is about 1000. According to Falicov and Sievert,<sup>10</sup> the saturation value due to orbits produced by magnetic breakdown is

$$\rho_{\text{sat}}/\rho_0 = 1 + C\omega_b\tau, \quad (5)$$

where  $\omega_b$  is the breakdown cyclotron frequency from Eq. (4) and  $C$  is a constant of order unity. If the open orbits in iron result from magnetic breakdown of closed orbits, we may therefore attribute the high value of the

<sup>10</sup> L. M. Falicov and P. R. Sievert, *Phys. Rev. Letters* **12**, 558 (1964). In this paper, Eq. (5) is given for the cases when open orbits break down to form closed orbits, and when a compensated metal becomes uncompensated due to magnetic breakdown of hole orbits for form electron orbits. But the expression is also correct for the case of interest where closed orbits break down to form open orbits [L. M. Falicov (private communication)].

saturation value to a high value of  $\omega_b\tau$ , and write

$$\rho_{\text{sat}}/\rho_{\min} \sim C\omega_b\tau/D_\infty, \quad (6)$$

where  $D_\infty$  is the fraction of the Fermi surface traversed by open orbits.

Alternatively, if the open orbits are associated with very small necks, corresponding to a fraction  $D \sim 10^{-3}$ , then the saturation value of the magnetoresistance ratio is independent of  $\bar{\tau}$ . Unfortunately, it is not possible to make a reliable estimate of the saturation value of the magnetoresistance by extrapolating the field dependence curves in Figs. 3 and 4. Thus, the final conclusion as to the origin of the open orbits awaits further investigation of still higher purity samples.

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### Spin-Wave Thermal Conductivity of Ferromagnetic EuS\*

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Experimental and theoretical investigations have been made of the spin-wave thermal conductivity of the ferromagnet europium sulfide. Measurements of the thermal conductivity from 1.5–4°K have been made on compressed powders. The theoretical temperature dependence of the spin-wave thermal conductivity below 4°K has been calculated for several external magnetic fields, assuming boundary scattering and considering first- and second-neighbor interactions. With no external magnetic field, the experimental conductivity can be approximately represented by  $\mathcal{K} = 1.2T^2$  mW/cm(°K) which is reasonably close to the theoretical temperature dependence. However, an external field of 9 kG produced a drop in conductivity ranging from 2 or 3% at 4°K to 25% at 1.5°K; according to theory the conductivity should decrease by 11% at 4°K and 31% at 1.5°K. This disagreement between experiment and theory is quite marked at higher temperatures and will require further study for its resolution.

#### I. INTRODUCTION

HEAT transport in ferromagnetic insulators might be expected to occur by means of lattice vibrations and spin waves. A spin-wave thermal conductivity has recently been observed in ferrimagnetic yttrium iron garnet by Douglass<sup>1</sup> and by Lüthi.<sup>2</sup> Scattering of phonons by spin waves has been assumed by Douthett and Friedberg<sup>3</sup> to explain their measurements of ferrite thermal conductivities. Apparently no observations of spin-wave thermal conductivity have been reported for a ferromagnet. Assuming that magnons and phonons have the same constant scattering length, a rough estimate of the thermal conductivity of europium sulfide (ferromagnetic below about 16°K) in the liquid-

helium temperature range shows that the spin-wave part should be much larger than the phonon part, as is the case with the specific heat.<sup>4</sup> The experiments discussed below were done in an attempt to discover whether or not spin waves do contribute appreciably to the thermal conductivity of EuS at these temperatures.

#### II. EXPERIMENTAL PROCEDURE

All of the rectangular specimens were cut from pellets compressed at 10 kbar. Single crystals have not been grown, as yet, due to the high temperatures required for growth by sublimation techniques. Earlier measurements of thermal conductivity in the liquid-helium temperature range were made of pressed pellets receiving no heat treatment after compression.<sup>5</sup> The

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<sup>1</sup> R. L. Douglass, *Phys. Rev.* **129**, 1132 (1963).

<sup>2</sup> B. Lüthi, *Phys. Chem. Solids* **23**, 35 (1962).

<sup>3</sup> D. Douthett and S. A. Friedberg, *Phys. Rev.* **121**, 1662 (1961).

<sup>4</sup> D. C. McCollum, Jr. and J. Callaway, *Phys. Rev. Letters* **9**, 377 (1962).

<sup>5</sup> R. L. Wild and D. C. McCollum, Jr., *Bull. Am. Phys. Soc.* **8**, 208 (1963).

data in Fig. 1 were obtained on specimens which had been heat treated at  $\approx 1600^\circ\text{C}$  in a vacuum of  $10^{-6}$  Torr for about 12 h. This heat treatment caused an increase in thermal conductivity by a factor of about 10 over the previous samples.

The thermal conductivity measurements were made by fairly standard techniques. Two carbon resistance thermometers were calibrated using a helium vapor-pressure bulb and then used to measure a steady-state temperature difference between two points on the specimen, of which one end was connected to the helium bath and the other to a heater. The thermometers were fitted to the expression  $T = A(\log_{10}R)/(\log_{10}R - B)^2$  by choosing a best value for the constant  $B$  and drawing a smooth curve of  $A$  versus  $R$  to take into account any small deviations of  $A$  from a constant value. The measurements in the presence of a magnetic field were made with a longitudinal external field of 10.3 kG. An estimate of 1.3 kG for the demagnetizing field was obtained by use of the demagnetizing factor of the ellipsoid that could be inscribed in the rectangular specimen, which was 10 mm long and 2.2 by 5 mm in cross section. Thus the resultant of these fields was about 9 kG.

The largest source of random error was an uncertainty of about 2% in temperature differences. A much larger systematic error, possibly  $\pm 10\%$ , could have been caused by the difficulty in determining the length between thermometers, because the specimens were small and the thermometers made contact over a finite area.

### III. THEORY

In this section, we discuss the calculation of the thermal conductivity of a spin-wave system in the low-temperature limit in the presence of a magnetic field. General formulas for the thermal conductivity have been given previously by Callaway and Boyd.<sup>6</sup> In the present case, it will be assumed that certain correction terms produced by normal magnon-magnon scattering processes can be neglected. Since the mean free path for magnon-magnon scattering varies as  $T^{-5/2}$ ,<sup>7</sup> this approximation should be valid in the temperature range of interest in these experiments. With this assumption, the thermal conductivity may be expressed as

$$\mathcal{K} = \frac{K}{3(2\pi)^3 \hbar^2} \int \left( \frac{E}{KT} \right)^2 \frac{e^{E/KT}}{(e^{E/KT} - 1)^2} (\nabla_{\mathbf{k}} E)^2 \tau(\mathbf{k}) d^3\mathbf{k}, \quad (1)$$

in which  $K$  is Boltzmann's constant,  $T$  is the absolute temperature,  $E = E(\mathbf{k})$  is the energy of a spin-wave mode of wave vector  $\mathbf{k}$ , and  $\tau(\mathbf{k})$  is the relaxation time for such a spin-wave mode.

The thermal conductivity depends on a magnetic

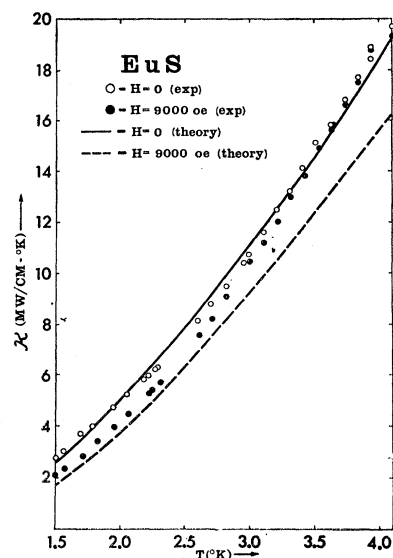


FIG. 1. Thermal conductivity versus temperature for europium sulfide.

field through the function  $E(\mathbf{k})$ , which may be written as<sup>8,9</sup>

$$E(\mathbf{k}) = (\epsilon_{\mathbf{k}} + g\beta H) \{1 + \varphi_{\mathbf{k}} \sin^2 \theta_{\mathbf{k}}\}^{1/2}, \quad (2)$$

in which  $H$  is the external magnetic field acting on the specimen, and  $\epsilon_{\mathbf{k}}$  is the "energy band" for spin waves. The latter quantity is expressed as<sup>10</sup>

$$\epsilon_{\mathbf{k}} = E_0 - 2S \sum_n J_n e^{i\mathbf{k} \cdot \mathbf{R}_n}. \quad (3)$$

In Eq. (3),  $\mathbf{R}_n$  is a direct lattice vector,  $J_n$  is an effective exchange integral, which is the same for all vectors  $\mathbf{R}_n$  of the same type, and

$$E_0 = 2S \sum_n J_n. \quad (4)$$

We will include only nearest and next-nearest neighbors in our use of Eq. (3). The remaining quantities in Eq. (2) are  $\theta_{\mathbf{k}}$ , which is the angle between the wave vector  $\mathbf{k}$  of the spin wave and the magnetization  $M$  of the specimen, and  $\varphi_{\mathbf{k}}$ , which is given by

$$\varphi_{\mathbf{k}} = 4\pi g\beta M / (\epsilon_{\mathbf{k}} + g\beta H). \quad (5)$$

Other quantities have their usual significance.

To simplify the calculation, we will ignore the dependence of the spin-wave energy on the direction of propagation, replace  $\sin^2 \theta_{\mathbf{k}}$  by its average value  $\frac{2}{3}$ , and expand the radical in (2). Only the first term is retained, so that (2) simplifies to

$$E(\mathbf{k}) = \epsilon_{\mathbf{k}} + g\beta(H + \frac{4}{3}\pi M). \quad (6)$$

This is the principal approximation which will be made in the evaluation of the thermal conductivity according to (1). To estimate the error involved, we note first that if the relaxation is by boundary scattering (which is characterized by a mean free path independent

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

<sup>7</sup> F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

<sup>8</sup> S. H. Charap and E. L. Boyd, Phys. Rev. **133**, A811 (1964).

<sup>9</sup> T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

<sup>10</sup> J. Callaway, Phys. Rev. **132**, 2003 (1963).

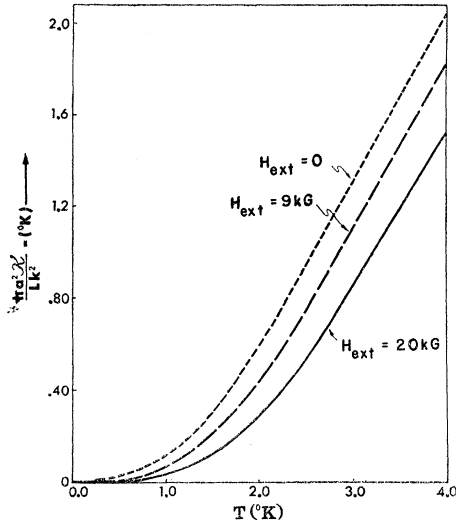


FIG. 2. Theoretical spin-wave thermal conductivity versus temperature.

of temperature), the maximum of the integrand occurs for  $E = 2.4KT$ . Using  $4\pi g\beta M/K \approx 2^\circ\text{K}$  as given by Charap and Boyd, the term  $\varphi_{\mathbf{k}} \sin^2\theta_{\mathbf{k}}$  at the maximum of the integrand is approximately  $4/7T$ . This is approximately 0.4 at the lowest temperatures ( $1.5^\circ\text{K}$ ) considered in this work. Hence the expansion employed should not be a source of significant errors.

It is necessary next to consider possible scattering mechanisms. Processes which may be of significance include boundary scattering, defect scattering, spin-wave-spin-wave scattering, and spin-wave phonon scattering. Of these, we believe that boundary scattering is likely to be the most important. This is suggested by the physical constitution of the samples, which are sintered powders, and by the approximate  $T^2$  temperature dependence of the conductivity. Scattering of spin waves by defects may be the next most important factor; but this would tend to make the thermal conductivity depend on the temperature much less strongly than  $T^2$ ,<sup>11</sup> and this is not evident in our observations. Magnon-magnon scattering should be weak as discussed above, and magnon-phonon scattering, while deserving careful examination, is somewhat restricted by the difficulty of satisfying momentum and energy-conservation requirements.

We will therefore consider only boundary scattering. This is characterized by a constant mean free path, which we denote by  $L$ . We have

$$\mathcal{K} = \frac{KL}{3(2\pi)^3\hbar} \int \left( \frac{E(\mathbf{k})}{KT} \right)^2 \frac{e^{E/KT}}{(e^{E/KT} - 1)^2} |\nabla_{\mathbf{k}\epsilon_{\mathbf{k}}}| d^3k. \quad (7)$$

If  $\epsilon_{\mathbf{k}}$  is approximated by  $Ck^2$ , the integral of Eq. (7)

<sup>11</sup> J. Callaway, Phys. Rev. 132, 2003 (1963).

may be evaluated by expanding the denominator according to the binomial theorem. The result is a series given by Douthett and Friedberg:

$$\mathcal{K} = \frac{LK^3T^2}{\pi^2\hbar C} \sum_{n=1}^{\infty} e^{-n\mathcal{K}} \left[ \frac{1}{n^3} + \frac{2\mathcal{K}}{3n^2} + \frac{1}{6} \frac{\mathcal{K}^2}{n} \right], \quad (8)$$

in which

$$\mathcal{K} = (g\beta/KT)(H + \frac{4}{3}\pi M). \quad (9)$$

However, in view of the fact that the approximation of  $\epsilon_{\mathbf{k}}$  by  $Ck^2$  is demonstrably inadequate to explain the specific heat of EuS, we have evaluated Eq. (7) numerically using Eqs. (6) and (3). The integral includes the exact Brillouin zone, and was performed using exchange integrals obtained by Charap and Boyd. The quantity  $4\pi M$  was taken as 14 kOe in agreement with those authors. Results are shown for several fields in Fig. 2. We note that the conductivity is reduced by the field, and that the curves for different fields are quite similar.

#### IV. RESULTS AND DISCUSSION

The theoretical and experimental results are shown in Fig. 1. An approximate fit to the experimental points in zero external field is given by  $\mathcal{K} = 1.2T^2$  mW/cm-deg. The theoretical prediction, represented by the solid line, has been adjusted to fit the experimental data to  $2^\circ\text{K}$  and it can be seen that the agreement between experiment and theory is fairly good. At  $1.5^\circ\text{K}$  the calculation yields  $\mathcal{K}\hbar a^2/LK^2 = 0.299$ , where  $L$  is the mean free path and the lattice constant  $a$  is  $5.96 \text{ \AA}$ . Inserting the experimental value of  $\mathcal{K}$  in this expression, the mean free path is found to be  $1800 \text{ \AA}$ . In the presence of an external magnetic field experiment and theory differ. Experimentally, a resultant external field of 9 kG causes a drop in thermal conductivity ranging from 25% at  $1.5^\circ\text{K}$  to 2 or 3% at  $4^\circ\text{K}$ . The theory predicts a drop of 31% at  $1.5^\circ\text{K}$  and 11% at  $4^\circ\text{K}$ , as shown by the dashed curve in Fig. 1.

It would appear that most of the thermal conductivity at the lowest temperatures is due to spin waves. However, the observation that the decrease in thermal conductivity produced by an external magnetic field is always smaller than the decrease given by spin-wave theory makes it impossible to rule out a phonon contribution. There may be phonon and magnon contributions with different mean free paths; scattering processes that depend on magnetic-field strength may occur. Further experiments in higher magnetic fields and attempts to produce single crystals of EuS are planned.

*Note added in proof.* Friedberg and Harris have measured the thermal conductivity of YIG and found equal scattering lengths for phonon and magnon components. (S. A. Friedberg and E. D. Harris, Proc. Eighth International Conference on Low Temp. Physics, p. 302.)