Heat Transport by Spin Waves in Yttrium Iron Garnet*

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The thermal conductivity of two single-crystal samples of the ferrimagnet yttrium iron garnet is reported between 0.4 and 20°K from 0 to 20 kOe. A 70% decrease in conductivity occurs when a strong magnetic field is applied at 0.5°K, and smaller reductions at higher temperatures. The results are interpreted as evidence for heat transport by spin waves, which has never been observed in other materials. The two samples show somewhat different temperature, field-magnitude, and field-direction dependences. These differences are qualitatively discussed, and can reasonably be ascribed to impurities.

INTRODUCTION

HEAT transport in solids at low temperatures is usually described by a theory of the motion and scattering of phonons and, in metals or semiconductors, electrons and holes. However, it was suggested as early as 1936 by Fröhlich and Heitler[1] that at a few hundredths of a degree Kelvin the heat transported by collective excitations of coupled paramagnetic ions might exceed that transported by phonons. Sato,[2] in 1955, suggested that in a ferrimagnetic dielectric, where spins are strongly exchange coupled to their neighbors, the spin-wave heat transport might be important in the liquid-helium range of 1 to 4°K. More recently, theoretical papers by Akhieser and Shishkin[3] and by Bar'yakhtar and Urushadze[4] treat heat conduction in pure and impure ferrimagnetic dielectrics, respectively.

An attempt to observe spin-wave heat transport in several ferrite single crystals has been reported by Douthett and Friedberg.[5] Whereas spin waves were, perhaps, important in modifying phonon heat transport, they did not themselves make the expected contribution to heat flow, possibly because of the magnetic inhomogeneity of the crystals investigated. Recently, several preliminary reports have been made[6–8] concerning the thermal conductivity of the ferrimagnet yttrium iron garnet (YIG). This paper discusses an investigation of the various features of heat transport in YIG.

Yttrium iron garnet is a particularly advantageous material for the search for spin-wave heat transport because of its relative freedom from magnetic disorder and its low magnetic anisotropy. At temperatures below 1.6°K with no applied magnetic field the specific heat due to spin waves in YIG exceeds that of the lattice.[9] A spin wave having exchange energy Dk^2 equal to the thermal mean k_BT has a group velocity $2Dk/\hbar$ of 3.5×10^5 cm sec^{-1} at 1.6°K, which is about the same as the transverse sound velocity in YIG. If such a spin wave has a relaxation time of 10^{-6} or 10^{-7} sec, which is a reasonable extrapolation from ferrimagnetic resonance and parallel pumping data in "pure" YIG,[10,11] then its relaxation length L_k is comparable to that expected for a thermal phonon. Thus, one may expect that the spin waves contribute a thermal conductivity $K=\frac{1}{3}\sum_k C_k v_k L_k$ which, in "pure" YIG at helium temperatures, is of the same order of magnitude as that due to phonons.

Identification of the spin-wave contribution to heat transport is possible because the spin-wave heat capacity C_k can be made very small by applying a magnetic field H so strong that $g\mu H \gg k_BT$. An investigation conducted below the liquid-helium range offers the possibility of a large spin-wave contribution to the heat transport and an almost complete quenching of this contribution with the application of moderate magnetic-field intensities. In the experiments to be reported, the thermal conductivity of YIG was measured as a function of temperature from 0.4 to 20°K and as a function of magnetic-field intensity from zero to 20 kOe.

EXPERIMENTAL

Thermal conductivity was measured by conventional steady-state techniques, temperatures being measured at two points along the specimen while a known amount of power flowed along its length. As thermometers, two carbon composition radio resistors, manufactured by the Speer Carbon Company, were used. These were measured separately with a 33-cps Wheatstone bridge, and calibrated against the vapor pressures of liquid He^4 and He^3. The lowest temperatures thus obtained agreed, within the experimental accuracy of a few millidegrees, with temperatures calculated from the measured susceptibility of a cerium magnesium nitrate pill in thermal equilibrium with the He^3. Thermal re-

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TABLE I. Relaxation lengths and resonance linewidths.

	Sample 1[a]	Sample 2[b]
$L_s(0.5°)$	2.3×10^{-2} cm	0.96×10^{-2} cm
$L_p(0.5°)$	3.4×10^{-2} cm	1.3×10^{-2} cm
Dimensions	1.22 cm×0.38 cm×0.35 cm	1.27 cm×0.46 cm×0.16 cm
$\Delta H(300°)$	1.2 Oe	1.4 Oe
ΔH_{max}	2.2 Oe	8 Oe
$\Delta H(4°)$	1.8 Oe	2.0 Oe

[a] From Microwave Chemicals Lab., Inc., 282 Seventh Ave., New York, New York.
[b] From Airtron Division, Litton Industries, 200 E. Hanover Ave., Morris Plains, New Jersey.

laxation times never exceeded 30 sec at any part of the temperature range. Results were reproducible from run to run unless the sample was removed and remounted, in which case a shift of up to 10% occurred. This shift resulted from a change in the heat-flow path, which, in these short, broad samples, depended on the evenness of the thermal contact with the heater and sink at the ends of the sample. Data could always be brought into coincidence with that of a previous run by using a constant renormalization factor. Experimental methods are more fully described elsewhere.[12]

The YIG samples used in the thermal conductivity measurements were cut from single crystals grown commercially by the slow cooling of a melt. The crystals were cut into the rectangular shapes listed in Table I with a diamond saw, and one surface of each sample was then polished and etched and examined under a microscope. The only defects detected were a few small pits, which might be associated with strain centers, and several larger (0.002 to 0.025-in.) holes in each crystal. All surfaces were then ground to a uniform finish with 600-mesh silicon carbide. Table I also gives the ferrimagnetic-resonance uniform-mode linewidths measured on spheres cut from the conductivity samples. It is believed that most of the room-temperature linewidth of both samples is due to surface pits, but the temperature-dependent component probably results from paramagnetic impurities.[13]

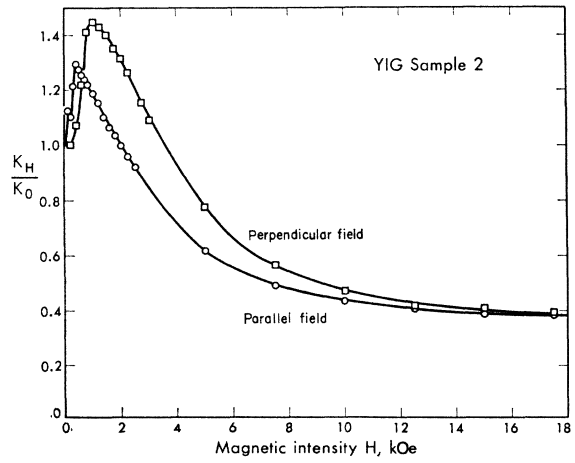

FIG. 1. Thermal conductivity at 0.5°K; the field directions are expressed relative to the direction of the heat flow.

RESULTS

At the lowest temperatures both samples exhibited the large decrease in conductivity expected upon the application of a strong magnetic field. Sample 2 showed the more complicated field dependence illustrated by Fig. 1. Let us suppose, for the moment, that the zero-field conductivity of sample 2 is the value obtained by a linear extrapolation of that portion of the curve above 1 kOe. Using this artificial zero-field conductivity, whose meaning is made clear presently, the field dependence for fields directed along the temperature gradient is as shown by Fig. 2. Similar curves are obtained for sample 1, using in this case the real zero-field conductivity. Thus, both samples showed a decrease of about 70% in thermal conductivity at 0.5°K with an applied field, and smaller decreases at higher temperatures.

This drop in thermal conductivity could be attributed to either the expected quenching of spin-wave transport

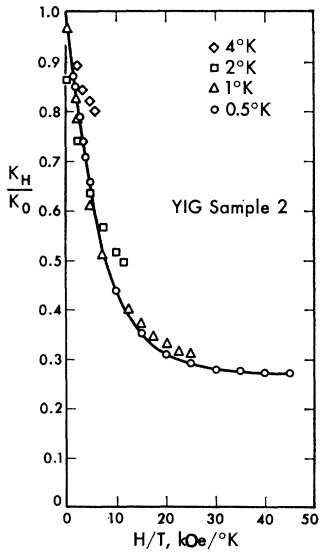

FIG. 2. Corrected field dependence at constant temperature; parallel field.

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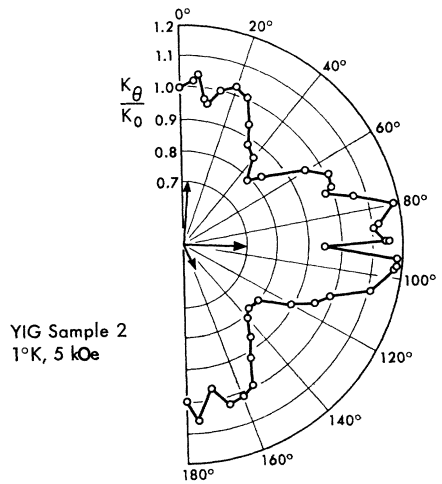

FIG. 3. Thermal conductivity vs field direction. The arrows at the center of the figure indicate the projections of the cubic axes.

by the field, or a decrease in the phonon relaxation lengths. The latter phenomenon would require a phonon relaxation rate which increases with field. It seems more likely that, as spin waves are "frozen out" by the field, the phonon relaxation rate decreases, because scattering processes involving spin waves no longer occur. Thus, even more than 70% of the zero-field heat transport may be due to spin waves at 0.5°K.

In both samples the conductivity is greater when the field is perpendicular to the temperature gradient than when the directions coincide. Figure 3 shows the relative thermal conductivity of sample 2 at 1°K with a constant internal field intensity of 5 kOe as a function of the angle between field and temperature gradient. Unfortunately, the sample was not cut accurately along the crystal axes. Projections of the cubic axes are shown by the arrows at the center of the figure. One sees that 0° and 90° are directions in which the conductivity is fairly large, whereas the easy directions of magnetization, which are the [111] and equivalent axes, are probably directions corresponding to much lower conductivity. Although no such angular-dependence curves were obtainable for sample 1, due to its larger shape anisotropy, the difference between conductivity with parallel and perpendicular fields is greater than can be accounted for by the difference in demagnetizing fields; i.e., there is a real dependence upon the internal field direction in sample 1 as well.

If we postulate that the thermal conductivity depends mainly on the magnetization direction, rather than the field direction, we can obtain a simple explanation of the low-field conductivity increase observed in sample 2, for the principal effect of a weak field is to re-orient the magnetization. It is reasonable to suppose that in the absence of external fields most of the magnetization aligns along easy directions, corresponding to low conductivity. An internal field of approximately 250 Oe is required to re-orient the magnetization in the 0° or 90° directions. The external fields required are about 375 Oe and 825 Oe for parallel and perpendicular directions, in rough agreement with the maxima of the conductivity of sample 2, as shown in Fig. 1. Thus, the conductivity value obtained by the previous linear extrapolation is the thermal conductivity which would result at zero field if sample 2 were magnetized along its length.

The two samples also have different temperatures dependences, as shown by Fig. 4. Note that the sample with lower conductivity at helium temperatures and below has the higher conductivity peak.

DISCUSSION

The behavior difference of the two samples indicates that impurities or other inhomogeneities are important scattering centers, rather than sample boundaries. Further confirmation of this is found in the absence of any significant change in field or temperature dependences upon decreasing the thickness of sample 2 from 0.16 to 0.055 cm.

However, the behavior of sample 1 is fairly well described by the theory of Sato,[2] which simply assumes a constant relaxation length L_s for all spin waves and a constant phonon relaxation length L_p. Using numerical values appropriate to YIG, the spin-wave thermal conductivity is[5,12]

$$K_s = 0.34 L_s T^2 F(H/T) \text{ W cm}^{-1} \text{ °K}^{-1}. \quad (1)$$

The function $F(H/T)$ is the normalized field dependence shown by the solid curve in Fig. 5. It can be seen that

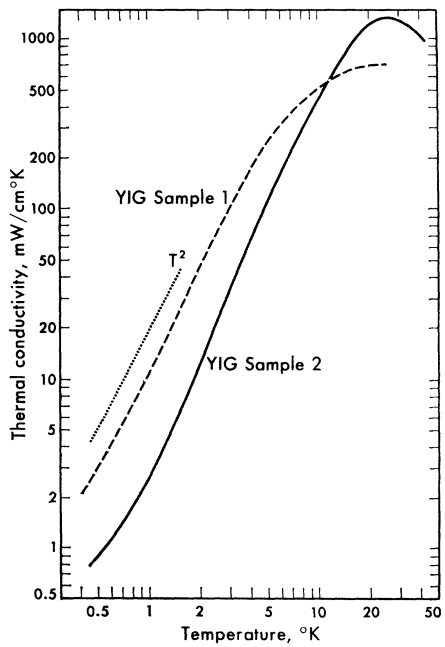

FIG. 4. Thermal conductivity at zero field.

sample 1 conforms much more closely to the predicted field dependence than does sample 2. Sample 1 also has a low-temperature zero-field conductivity which is nearly proportional to T^2, as shown in Fig. 4.

The phonon thermal conductivity for YIG, assuming equal relaxation lengths, is[12]

$$K_p = 0.24 L_p T^3 \text{ watt cm}^{-1} \text{ °K}^{-1}. \quad (2)$$

If we take the total decrease in conductivity caused by a magnetic field as equal to the zero-field spin-wave contribution and attribute the remainder to phonons, (1) and (2) yield the relaxation lengths given in Table I.

It is suggested that the lower conductivity, the low-field increase in conductivity, and the larger dependence on field direction observed with sample 2 are due to an impurity whose scattering rate depends on the direction of magnetization. Because the directional dependence of the conductivity vanishes at high fields, it is probably associated with scattering of spin waves. The ferrimagnetic linewidth results shown in Table I suggest that the scattering centers are paramagnetic impurity ions, which have often been found to cause a characteristic peak in the linewidth vs temperature curve of YIG.[13] Even in the absence of strong spin-lattice coupling, a paramagnetic impurity ion that is exchange-coupled to the ferric lattice is a powerful scattering center for low-energy spin waves.[4] The spin-lattice interaction can lead to a dependence of this scattering on magnetization direction.[12]

It might be thought that intrinsic magnon-phonon interactions would give rise to thermal resistance. However, the situation is analogous to that of phonons by themselves,[14] where thermal resistance can only arise if there are scattering processes which do not conserve wave vector, i.e., Umklapp or extrinsic processes. If processes conserving wave vector are much stronger than those which do not, it can be shown[12] that the ratio of spin-wave and phonon heat currents equals twice the ratio of the heat capacities, or, for YIG,

$$K_s/K_p = (2.6/T)^{3/2}. \quad (3)$$

Equation (3) may be more appropriate than (1) and (2) for temperatures above the He range, but in either case, we see that the spin-wave heat transport rapidly becomes negligible at higher temperatures. In the neighborhood of the conductivity maxima, the heat transport is surely due to phonons. A likely source of thermal resistance at these temperatures is the scattering of phonons by atomic Pb, known to be present in concentrations of tenths of a percent as a result of the use of a lead flux in the growth of the crystals. Thus, it is not unreasonable that the sample with the smaller low-temperature conductivity can have the greater peak conductivity.

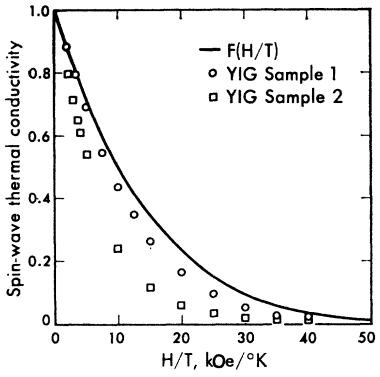

FIG. 5. Comparison of field dependences at 0.5°K; heat flow parallel to magnetic field.

The results are not well described by the theory of Bar'yakhtar and Urushadze,[4] which deals with thermal resistance arising from impurities but treats only the zero-field temperature dependence. The reason for this failure is the existence, in YIG, of a phonon-scattering mechanism, probably associated with paramagnetic impurities, which is much more effective at long wavelengths than those employed by these authors. This process is not one involving creation or destruction of spin waves, for it persists at low temperature and high fields, leading to temperature dependences of $T^{1.7}$ and $T^{1.3}$ for samples 1 and 2, respectively, at temperatures between 0.4 and 0.6°K. No explanation of these temperature dependences is offered.

ACKNOWLEDGMENTS

The author wishes to express his appreciation to Professor Charles Kittel for suggesting this investigation, and to Professor Norman Phillips for discussions concerning experimental techniques, and to Professor Klaüs Dransfeld for his interest and suggestions.

[14] R. Peierls, *Quantum Theory of Solids* (Oxford University Press, New York, 1955), p. 42.