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Far-Infrared Exchange Resonance in Ytterbium Iron Garnet*

A. J. SIEVERS, III, AND M. TINKHAM†
Department of Physics, University of California, Berkeley, California

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An exchange resonance has been observed in a pressed powder sample of ytterbium iron garnet at 14.1±0.2 cm^{-1} at 2°K. The temperature dependences of the resonance frequency and absorption strength were measured from 2°K to 66°K. The resonant frequency at all temperatures is in good agreement with the two-sublattice theory when the appropriate spectroscopic g value $(3g_J)$ is used and when the anisotropy energy is taken into account at low temperatures. The two-sublattice model also reproduces the measured absorption strength to within a factor of two, and its temperature dependence even more precisely. Two other magnetic absorptions with temperature-independent frequencies were observed at 23.4±0.3 cm^{-1} and 26.4±0.3 cm^{-1}. At 30°K, an 11-koe field on a single crystal was found to vary the frequency and intensity of these lines, which arise from the exchange splittings of the ytterbium ground-state doublet. The contribution via dispersion theory of the infrared absorption lines to the static susceptibility at 2°K is in good accord with the dc value measured by Pauthenet. This agreement suggests that all major contributions to the static susceptibility have been measured.

I. INTRODUCTION

IN magnetic systems which are composed of more than one sublattice, it is in general possible to observe a collective transition directly related to the exchange energies connecting the different sublattices. The exchange resonance between two sublattices was first suggested and derived by Kaplan and Kittel.[1] They found the exchange resonance frequency to be

$$\omega=\lambda(\gamma_2M_1-\gamma_1M_2), \qquad (1)$$

where λ is the molecular field constant; γ_1,γ_2 the respective gyromagnetic ratios; and M_1,M_2 the sublattice magnetizations. A more detailed treatment of the situation in the garnets is given in the preceding paper,[2] hereafter referred to as I.

Pauthenet's dc susceptibility investigations[3] of the rare-earth iron garnets have indicated that for these materials a two-sublattice model in which the rare-earth ions are individually coupled to the iron sublattice is a valid approximation. This paramagnetic nature of the ytterbium sublattice gives rise to a marked temperature dependence of its magnetization, which in turn should produce a characteristic temperature dependence in the exchange resonance frequency. Thus, transmission measurements on powder samples as a function of temperature permit us to differentiate between the two types of magnetic absorptions that are observed in the far infrared, the collective exchange resonance and the quasi-ground state splittings of the single ytterbium ions in the iron exchange field, as modified slightly by spin-wave effects.

II. EXPERIMENT

The experiment consisted of transmission measurements on an ytterbium iron garnet (YbIG) single crystal and two YbIG pressed powder samples in the far infrared at low temperatures. The measurements were made under different conditions induced by variation of one of the four available parameters: (1) infrared frequency, (2) magnetic field, (3) sample thickness, and (4) sample temperature.

The far-infrared grating monochrometer used in these experiments has been described in full elsewhere.[4] Briefly, it consisted of a blazed grating arranged in a Littrow mounting with a high-pressure mercury arc as source. A light pipe carried the radiation to the low-temperature sample and bolometer detector. The burden of filtering higher-order radiation from the dispersion grating was placed on transmission filters, since only one reflection from a restrahl plate or "zero-order filter" was possible. NaCl was used as a transmission filter material, since its absorptive properties have been accurately measured from 33 cm^{-1} to 3.3 cm^{-1} by Genzel *et al.*[5] As much as 19 mm of NaCl was required to reduce the second-order intensity below one-tenth of the first order at the lowest frequencies.

A typical low-resolution transmission trace of YbIG pressed powder at 2°K in the 25-cm^{-1} region is shown in Fig. 1. The shallow wiggles superimposed on the intensity distribution are due to interference effects in the fused quartz envelope of the mercury arc source. The irregular intensity distribution vs wavelength emphasizes the necessity of recording the intensity distribution with and without a sample.

The second parameter, the magnetic field, was supplied by a 12-in. electromagnet with a 3-in. gap. This magnet could be rotated through 180°, and the maximum field was about 11 koe.

The pressed powder disk samples of YbIG were 16 mm in diameter and of thickness 0.96 mm and 1.47 mm, respectively. The irregularly shaped single crystal of YbIG was about 5.5 mm in diameter and of roughly 2 mm thickness. The crystal was oriented so that a

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[1] J. Kaplan and C. Kittel, J. Chem. Phys. **21**, 760 (1953).

[2] M. Tinkham, preceding paper Phys. Rev. **124**, 311 (1961), referred to as I.

[3] R. Pauthenet, Ann. phys. **3**, 424 (1958).

[4] R. C. Ohlmann and M. Tinkham, Phys. Rev. **123**, 425 (1961).

[5] L. Genzel, H. Happ, and R. Weber, Z. Physik **154**, 13 (1959).

magnetic field would be in the (110) plane for transmission measurements with the infrared radiation propagating along the [110] axis. Most of the transmission measurements were made with the powder samples, since they were the same size as the light pipe and hence the infrared beam. Since the single crystal had an area of only about one-fourth that of the light pipe, the use of a time constant 16 times longer was required to obtain the same signal-to-noise ratio as with the powders. This increase usually proved not to be experimentally feasible.

The sample temperature was controlled manually between 10° and 70°K, and measured with a Au+Co: Ag+Au thermocouple[6] which was glued to its edge. The sample was arranged in a chamber in series with the light pipe. The sample assembly could be rotated in and out of the infrared beam by means of a long thin-walled stainless-steel tube which carried the thermocouple leads through a vacuum seal to room temperature, as well as forming a mechanical linkage between the low-temperature system and room temperature. Sample cooling was provided by a copper braid connecting the sample mount to a copper plug brazed in the chamber wall. Since this chamber was about 25 cm from the inside bottom of the inner glass Dewar, the chamber was not always immersed in liquid helium. However, a copper braid attached to the outside end of the copper plug in the chamber wall hung two thirds of the way to the bottom of the Dewar. The sample temperature was then controlled by a heater which was also attached to the outside end of the copper plug. The lowest temperatures were maintained by completely immersing the chamber in liquid helium and pumping on the helium vapor. The intermediate temperatures, 10° to 40°K, were maintained by manually adjusting the heater when the helium was about 10 to 15 cm below the sample holder, still touching the braid. This method was found to be very satisfactory, since an adjustment of the heater current every 20 min was sufficient to hold the temperature constant to about $\frac{1}{2}$° for three hours. The high temperatures, 40° to 70°K, were obtained only when the liquid helium had dropped below the copper braid; however, the heater required frequent adjustment in this region.

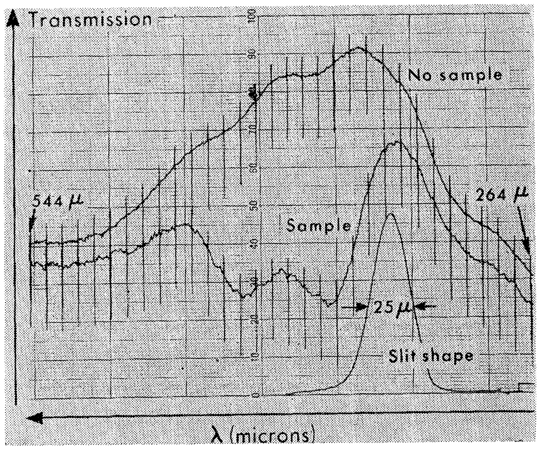

FIG. 1. Intensity distribution as a function of wavelength for a specific grating in the region of the first-order blaze wavelength, both with and without a YbIG pressed powder sample in the flux path. The slit had an effective width of 25 μ and the recorder gain was changed between the sample and no-sample run.

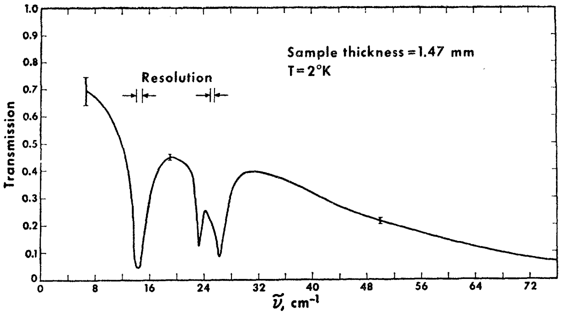

FIG. 2. Transmission through a YbIG pressed powder sample of thickness 1.47 mm at a temperature of 2°K. High-resolution data were only obtained between 22 and 28 cm^{-1}.

III. RESULTS

Although the monochrometer was capable of reaching frequencies as high as 200 cm^{-1}, the strong lattice absorptions in YbIG above 100 cm^{-1} rendered this region experimentally impractical. As the infrared frequency was decreased from 100 cm^{-1} to 6.7 cm^{-1}, the powder became increasingly transparent, as shown

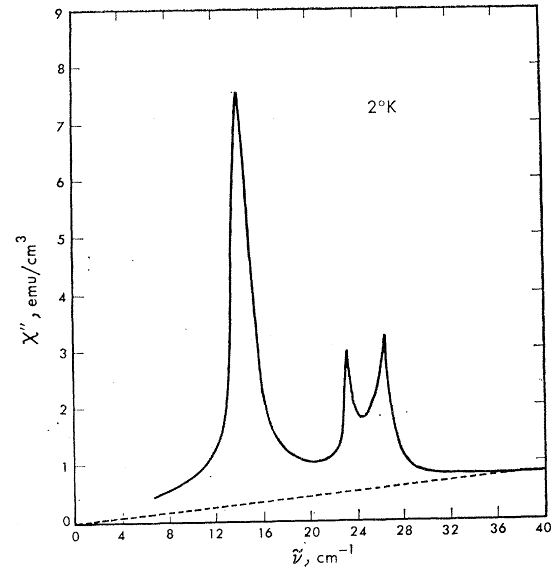

FIG. 3. The χ'' calculated from data of Fig. 2. (The ordinate scale must be multiplied by 10^{-3}). The area below the dashed line is the estimated electric absorption contribution from the high-frequency lattice modes. The integrated magnetic absorption is equal to the total area less the electric part.

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[6] G. K. White, *Experimental Techniques in Low-Temperature Physics* (Oxford University Press, New York, 1959).

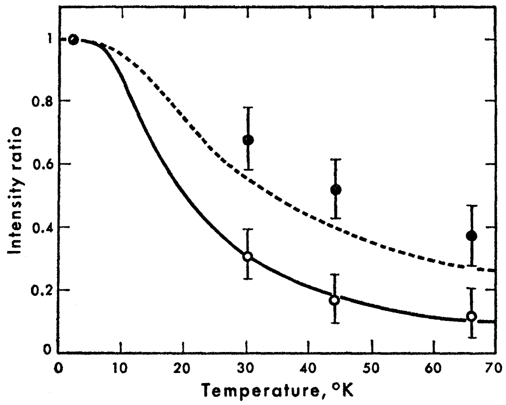

FIG. 4. Temperature dependence of absorption strengths. The strengths of the two absorptions at different temperatures are normalized to the 2°K measured values. The solid points give the measured temperature dependence of the single-ion transitions. (We have considered the doublet as one line for this comparison.) The calculated temperature dependence of a two-level system is given by a dashed curve. The open circles give the measured temperature-dependence of the exchange resonance strength, and the solid curve represents the temperature dependence calculated via the isotropic two-sublattice approximation.

in Fig. 2. The reflection loss essentially did not depend on the magnetic absorptions, since $4\pi|\chi|$ was always much smaller than one. Thus, measurements with two sample thicknesses allowed an estimate of the dielectric reflection loss and the corresponding index of refraction in the low-frequency region beyond the lattice absorptions. We found that $n=\epsilon^{\frac{1}{2}}=2.2$ was consistent with our experimental results at 2°K.

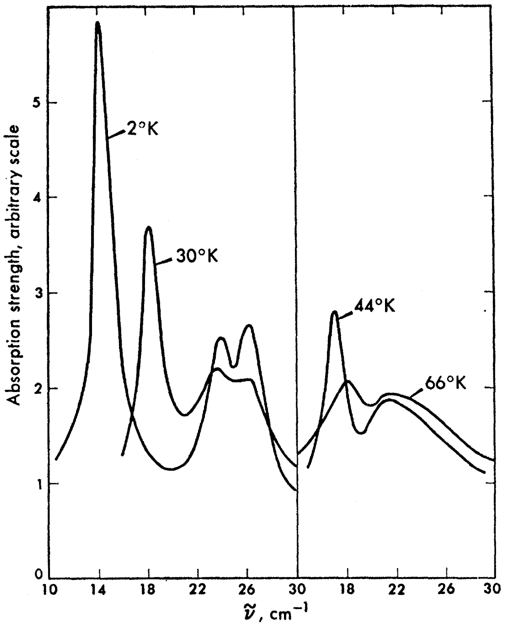

FIG. 5. Absorption strengths (χ'' on arbitrary scale) at different temperatures. Note the temperature-dependent shape of the doublet as well as the temperature-independent intrinsic linewidth of the exchange resonance below 44°K.

In Fig. 2, the magnetic absorptions appear superimposed on the low-frequency wing of the electric absorptions. This is displayed more clearly in Fig. 3, where χ'', the imaginary part of the complex susceptibility, derived from Fig. 2, is presented. At frequencies low compared to the lattice absorption, in the classical oscillator approximation, the electric χ_e'' has a linear frequency dependence. We have estimated this contribution by a dashed line in Fig. 3. The magnetic absorption around 25 cm^{-1} seems to be composed of three lines: two lines of slightly different strengths at 26.4 ±0.3 cm^{-1} and 23.4$\pm$0.3 cm^{-1} and a weak third line at 24.6$\pm$0.6 cm^{-1}. The intrinsic linewidths, or frequency intervals between those values of χ'' which are one-half of the maximum values of χ'', for these three lines are 1.4, 1.1, and 1.6 cm^{-1}, respectively. It should be mentioned that the effective spectrometer slit width had modified the true shape, since its width was not negligible. Although a detailed consideration of this refinement would surely change our estimates of the integrated absorptions (Sec. IV), we felt the quality of the data possible did not warrant such close examination of the experimental results. The absorption mechanism was revealed through the magnetic field and temperature variation.

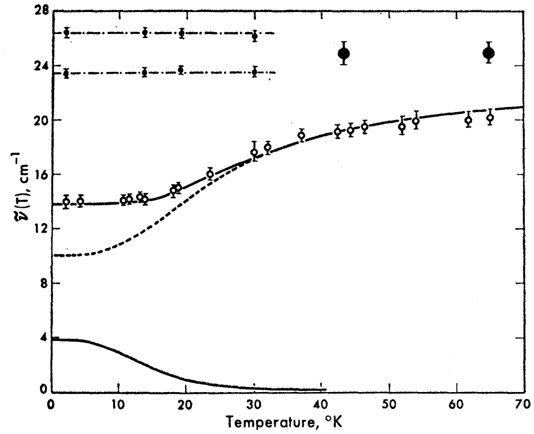

FIG. 6. Temperature dependence of the resonance frequencies for the three strong lines. The two highest frequencies are temperature independent. Above 30°K only the center of gravity of the doublet could be measured. The dashed curve gives the calculated temperature dependence of the exchange frequency in the isotropic two-sublattice approximation. The solid curve demonstrates the good agreement between theory and experiment when anisotropy energy is introduced. The low-frequency curve is the predicted "ferrimagnetic" resonance with the same model.

At 2°K the net magnetic moment in YbIG is only roughly 2% of either sublattice magnetization. A moderate (11 koe) magnetic field had no observable effect on these absorptions because of the small net magnetic moment and the large anisotropy at low temperatures. An increase in sample temperature from 2° to 66°K produced no measurable change in the frequency of these absorptions, although the intensity of the absorptions was found to decrease rapidly with increasing temperature, as shown in Fig. 4. This statement should perhaps be qualified. As the temperature was raised from 2° to 30°K, the strength of the 26.4 cm^{-1} line decreased the more rapidly, until at 30°K the two maxima were just distinguishable, as shown in Fig. 5. Therefore, at temperatures of 44°K and above we essentially measured only the center of gravity of the lines. The magnetic nature of these absorptions was demonstrated with the single crystal of YbIG. With the sample at 30°K, a field of 11 koe was found to cause a shift in the frequency, an increase in the number, and a corresponding decrease in the intensity of the absorptions. The decrease in intensity unfortunately made it impossible actually to plot the resonance frequencies as a function of crystal orientation in the magnetic field.

These absorptions have been interpreted as due to three normal modes in a more complex sublattice model treated in Sec. III of I, in which the iron sublattice appears to remain almost stationary. The two strong absorptions correspond to the ground-state splittings of the Yb^{3+} ions in the iron exchange field. The two lines are expected, since the ions are distributed in two magnetically inequivalent sites when the spontaneous magnetization is along the crystal [111] direction (easy axis). These splittings recently have been studied extensively in an elegant series of experiments by Wickersheim[7] in the near infrared, from which the single-ion splittings were inferred. The weaker line at the average frequency arises from a mode in which these two types of ions oscillate 180° out of phase with each other. It is expected to be weaker, since there is a greater averaging of transverse g values when more ions participate.

The strong absorption in the YbIG pressed powder at 14.1 ± 0.2 cm^{-1} had an intrinsic linewidth of 1.8 cm^{-1}. Again a magnetic field at 2°K had no observable effect. However, a temperature variation produced a marked change in the resonance frequency, as shown in Fig. 6. This temperature dependence qualitatively has the characteristic shape expected of an exchange resonance. However, as will be discussed later, at low temperatures better harmony was obtained between theory and experiment by introducing an anisotropy energy into the isotropic exchange model of Kaplan and Kittel. The intensity of the absorption was found to drop more strongly with increasing temperature than for the single-ion splittings, as seen in Fig. 4, so that the absorption could not be followed above 66°K. The linewidth, on the other hand, was found to be approximately constant up to 44°K, as shown in Fig. 5.

In a preliminary announcement,[8] we reported the existence of still another absorption at 7 cm^{-1}. Further experiments have shown this line to be a spurious result of the strong 14 cm^{-1} line which appeared due to inadequate filtering of second-order grating radiation.

IV. DISCUSSION

In comparing our experimental results with the isotropic exchange frequency, we have not used the Landé g in the gyromagnetic ratio of the rare-earth sublattice, but rather an effective g arising from modifications imposed on the free ion by the local crystal fields. This problem has been considered in Sec. IV of I.

The crystal-field splittings of the Yb^{3+} ion in the garnet structure have been measured by Pappalardo and Wood.[9] Their measurements show that the ground state is about 550 cm^{-1} below the next state. This splitting pattern is in agreement with the calculations of Ayant and Thomas,[10] which show that the cubic crystal field causes the ground state to be a degenerate Kramers doublet well below the other states. For a cubic crystal field at the Yb^{3+} ion, the isotropic g_{eff} for a splitting of the lowest doublet in an external magnetic field is equal to three times the Landé g factor, thus, $g_{\mathrm{eff}}=3\times8/7=24/7$. This value agrees well with the average g observed in paramagnetic resonance experiments on Yb^{3+} in diamagnetic garnets by Boakes *et al.*[11] and by Carson and White.[12] Accordingly, it is the value we have used in Fig. 6 for the comparison of the Kaplan-Kittel isotropic exchange resonance theory with the measured curve.

The effect of anisotropy on the exchange resonance has been described in Sec. V of I. With the values of K_1 and K_2 [see Eq. (27) of I] calculated by Henderson and White[13] from Wickersheim's observed anisotropic exchange splittings,[7] we have obtained a parameter-free fit to the exchange resonance by using Eqs. (34)–(36) of I. The solid curve in Fig. 6. is to be compared with the experimental points. Because of the extremely close fit to the exchange resonance, we have also shown the theoretical temperature dependence of the low-frequency "ferrimagnetic" mode which also results from the same model [Eq. (34) of I].

A further comparison between the predicted and measured results can be obtained from a comparison of the exchange resonance intensity. The integrated absorption of this resonance in the isotropic model has been obtained in Eq. (13) of I. For YbIG, this leads to

$$\int \chi_x''(\tilde{\nu})d\tilde{\nu} = 12\times10^{-3}\ (\mathrm{emu/cm^3})\mathrm{cm}^{-1}\quad \text{(calculated)}. \qquad (2)$$

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[8] A. J. Sievers, III, and M. Tinkham, Bull. Am. Phys. Soc. **6**, 160 (1961).

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[11] D. Boakes, G. Garton, D. Ryan, and W. P. Wolf, Proc. Phys. Soc. (London) **74**, 663 (1959).

[12] J. W. Carson and R. L. White, J. Appl. Phys. **31**, 53S (1960).

[13] J. Henderson and R. L. White, Phys. Rev. **123**, 1627 (1961).

We have estimated a value from the experimental data in Fig. 3,

$$\int \chi_x''(\tilde{\nu})d\tilde{\nu} = 21\times10^{-3}\ (\mathrm{emu/cm^3})\mathrm{cm^{-1}}\quad (\text{measured}). \tag{3}$$

The anisotropy at low temperatures would modify the calculated value, so that the factor of two difference is perhaps not unreasonable. Some question still remains, however, since the temperature dependence of the intensity in the same approximation has been found to agree with the experimental points, as shown in Fig. 4.

Since the contribution to the static susceptibility can be obtained from the integrated absorption quite easily if the lines are narrow, we have again made this approximation [see Eqs. (11) and (14) of I] to write

$$\chi_x(0) \cong \frac{2}{\pi}\sum_i \frac{1}{\omega_{0i}} \int \chi_x''(\omega_i)d\omega_i, \tag{4}$$

where the sum is over the magnetic absorptions. An estimate of the areas under these absorptions from Fig. 3 gave the value

$$\chi_x(0) = 1.2\times10^{-3}\ \mathrm{emu/cm^3}; \tag{5}$$

thus

$$\chi_m(0) = \chi_x(0)A/\rho = 1.2\times10^{-3}(1982/6.28) = 0.38\ \mathrm{emu/mole}.$$

Pauthenet's measured value was

$$\chi_m(0) = 0.37\ \mathrm{emu/mole}. \tag{6}$$

The excellent agreement is no doubt fortuitous, but it suggests that all major contributions to the static susceptibility have been considered.

Although more magnetic absorptions have been observed in the far infrared than originally anticipated, a six-sublattice model, discussed in I, quantitatively describes the experimental results. Also the dependence of the exchange resonance on different experimental parameters has been predicted in detail. These predictions, however, are found to be valid only if the appropriate $g(\neq g_J)$ value is used in the gyromagnetic ratio of the rare-earth sublattice.

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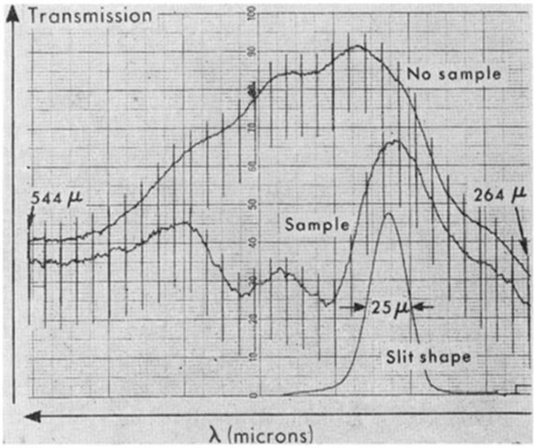

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