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Far Infrared Exchange Resonance in Rare Earth Iron Garnets

M. TINKHAM
Department of Physics, University of California, Berkeley California, U.S.A.

In the rare earth iron garnets, the exchange coupling of the rare earth ions to the iron is relatively weak (~10-50 cm^{-1}), while the iron ions are strongly coupled together. Thus the low-lying excitations observable in the far infrared depend on the iron-rare earth coupling parameter λ. Two types of excitations are seen. One is a collective mode predicted by Kaplan and Kittel in which the entire iron and rare earth sublattices precess as units, with a frequency depending on sublattice magnetization. The other type consists essentially of single-ion transitions, made possible by breakdown of simple selection rules by anisotropy, with temperature-independent frequencies. We have completed a detailed study of YbIG, which shows resonances at 14.1, 23.4, and 26.4 cm^{-1} at low temperatures. A preliminary study of ErIG shows many more absorption lines, corresponding to the greater number of low-lying crystal-field levels.

The exchange coupling between rare earth and iron ions in rare earth iron garnets is typically of order 10-50 cm^{-1} (15-75°K), while the iron ions are coupled more strongly together, corresponding to their Curie temperature of about 550°K. Thus, at low temperatures, the iron lattice acts essentially as a rigid unit, and excitations observable in the far infrared have to do with the iron-rare earth coupling constant λ.

One type of excitation is the exchange resonance predicted by Kaplan and Kittel[1]. This has the frequency

$$\omega_e=\lambda(\gamma_2 M_1-\gamma_1 M_2) \qquad (1)$$

where the subscripts 1 and 2 refer to iron and rare earth sublattices, respectively, which are assumed to precess as units. Evidently the frequency of the resonance will change markedly as M_2 falls with increasing temperature.

The other type of excitation is essentially an excitation of a single rare earth ion to a higher level. The level structure arises from a combination of the crystalline electric field and the exchange field of the iron. Many of these transitions would be forbidden if all the rare earth ions were equivalent and isotropic, but departures from isotropy are so great that they are quite strongly allowed in fact. The frequencies of these transitions are nearly temperature independent in the region of interest ($T<100$°K) because the iron exchange field is nearly constant so far below the Curie point. Thus, one can distinguish the two types of resonance experimentally by examination of the temperature dependence.

Sievers and I have studied in detail[2] the spectrum of YbIG from 100μ to 1500μ by means of transmission measurements on polycrystalline discs about 1 mm thick. Since the iron magnetization is held to a [111] direction in each crystallite by the anisotropy energy, the spectrum is characterized by the symmetry of that direction. With the exchange field in that direction, the lowest-lying Kramers doublet of the Yb ion is split by 23.4 or 26.4 cm^{-1}, according to which type of site it is on. [In a general direction, there would be 6 different splittings.] In addition to these single-ion splittings, we see a strong collective resonance at 14.1 cm^{-1}, at 2°K. As the temperature is raised, the former frequencies are unaffected, but the lattter rises, and reaches ~20 cm^{-1} by the time 60°K has been reached. Observation then becomes difficult since all the absorptions become weak and merge together.

If we average out the anisotropy to recover the overall cubic symmetry, we expect $\bar{\gamma}_2$ corresponding to $g=3g_J=24/7$. This conclusion can be reached theoretically[3], or by inspection of paramagnetic resonance data on Yb in YGaG and YAlG[4]. If we average the two single-ion exchange splittings, we find that $\overline{\lambda\gamma}M_1=24.9$ cm^{-1}. Taking magnetization data from Pauthenet[5] or the calculations of Henderson and White[6], and taking $g=2$ for the iron lattice, we can then calculate ω_e from (1) to be 10.1 cm^{-1} at $T=0$. As

M_2 falls with increasing temperature, ω_e should rise toward $\overline{\lambda\gamma_2 M_1}\approx 25\,\mathrm{cm}^{-1}$, as observed. The discrepancy between 10.1 and 14.1 cm^{-1} at low tempertures is due to neglect of anisotropy in the Kaplan-Kittel model. To correct for this, one can introduce an averaged anisotropic exchange coupling into the equations of motion via an effective $\gamma_\perp \neq \gamma_{//}$. If $(\gamma_\perp^2-\gamma_{//}^2)$ is evaluated in terms of macroscopic anisotropy constants, we find essentially perfect agreement with the 14.1 cm^{-1} frequency at $T\approx 0$, and with the entire temperature dependence of the resonance frequency. We also find satisfactory agreement between theory and experiment on the absolute intensity of the collective mode.

Subsequent to this complete study of YbIG, we have initiated a similar study of ErIG. The spectrum is similar, but more complex. At 2°K, there is a strong line at 10 cm^{-1}. This is the exchange resonance mode, and its temperature dependence is similar to that of this mode in YbIG. There is a strong doublet at 18.2 and 21.6 cm^{-1}, which is presumed to arise from two different exchange splittings of the ground doublet, as in YbIG. In addition there appear to be 8 to 10 lines in the region 30 to 100 cm^{-1}. These lines presumably arise from transitions from the ground state to higher doublets separated from it by crystal field splittings. These results are in excellent agreement with the specific heat data of Meyer and Harris[7]. They fit their data with a doublet at 16 and 24 cm^{-1}, and a group of additional levels at about 50 cm^{-1}. The average of 16 and 24 is nearly the same as the average of our values, 18.2 and 21.6 cm^{-1}, and the levels near 50 cm^{-1} correspond to our lines at 30–100 cm^{-1}.

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