Resonant microwave absorption in Ising systems: holmium–yttrium iron garnets

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Dynamic properties of the magnetic system of a $Ho_{0.4}Y_{2.6}Fe_5O_{12}$ single crystal have been studied in strong magnetic fields. In addition to the absorption lines associated with magnetic phase transitions which are familiar from static measurements, there is a large number of resonances far from these points. These resonances occur in fields below the exchange field and also in fields well above the exchange field. The results are interpreted on the basis of a model of an Ising ordering of holmium ions.

Recent studies of magnetic resonances in terbium–yttrium[1] and holmium–yttrium[2] iron garnets revealed, along with the ferromagnetic resonance, several absorption lines at frequencies in the range 30–110 GHz in strong magnetic fields. The number of these lines was correlated with the number of phase transitions induced by an external magnetic field. This circumstance, along with the field dependence of the magnetic-resonance frequencies, was interpreted on the basis of a model of a crossing of levels of the rare-earth ion. For the holmium–yttrium garnets, it has been observed that there are also absorption lines which are totally unrelated to magnetic-structure transitions.

To obtain additional information on the absorption spectra of rare-earth iron garnets in strong magnetic fields and information on the effect of the field orientation and the concentration of rare-earth ions on these spectra, we have carried out an experimental study of magnetic resonances in $Ho_{0.4}Y_{2.6}Fe_5O_{12}$ single crystals over broad ranges of the frequency (30–110 GHz), the field (up to 300 kOe), and the temperature (from 4.2 to 160 K). The external magnetic field was oriented along three principal crystallographic directions: [100], [110], and [111]. In the present letter we are reporting only the results obtained at liquid-helium temperature with the field oriented along the [111] or [100] axis.

The procedure used for the experiments carried out in pulsed magnetic fields is described in detail in Ref. 2. The test samples were some single crystals which had been used previously[3] for static magnetic measurements. The samples were rectangular parallelepipeds with a base of 1.5 × 1.5 mm and a length of 6–8 mm.

Figures 1 and 2 show the magnetic-resonance frequencies versus the strength of the external field, $\nu(H)$, at 4.2 K. These curves were constructed from an analysis of the absorption spectra. We see that for the different orientations of the external field there are different numbers of narrow absorption lines (group A) in fields close to the

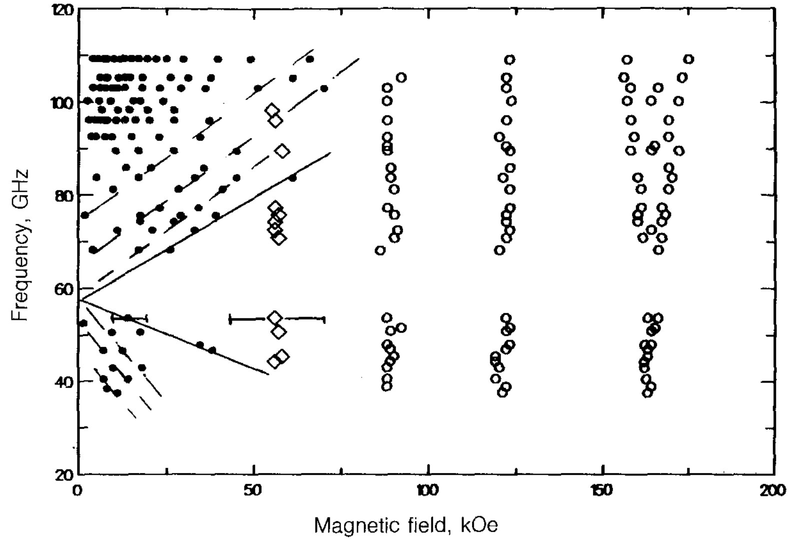

FIG. 1. Field dependence of the frequencies corresponding to the maxima of resonant microwave absorption in a $Ho_{0.4}Y_{2.6}Fe_5O_{12}$ single crystal with $H \| [111]$ at $T = 4.2$ K. Open circles—lines of type *A*; rhombi—lines of type *B*; filled circles—lines of type *C*. The horizontal error bars show the typical half-width of the resonance lines.

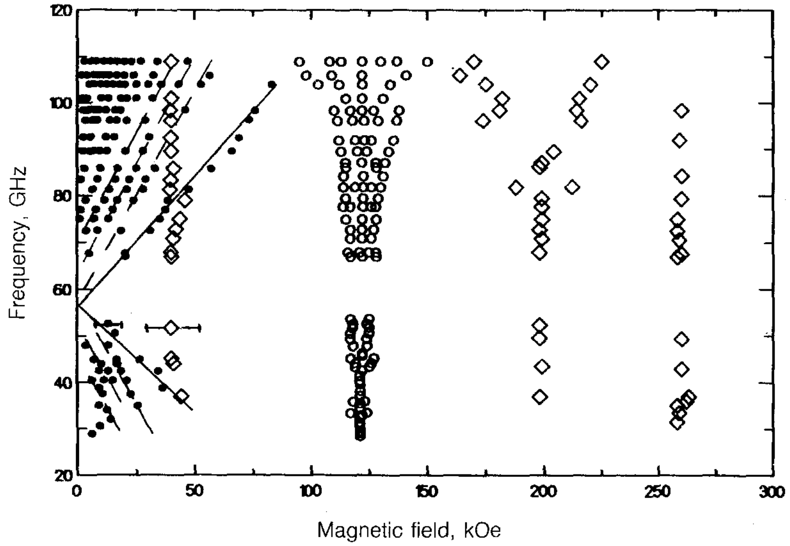

FIG. 2. Field dependence of the frequencies corresponding to the maxima of the resonant microwave absorption in a $Ho_{0.4}Y_{2.6}Fe_5O_{12}$ single crystal with $H \| [100]$ at $T = 4.2$ K. Open circles—lines of type *A*; rhombi—lines of type *B*; filled circles—lines of type *C*. The horizontal error bars show the typical half-width of the resonance lines.

field of the exchange interaction between the rare-earth and iron magnetic subsystems of the crystal ($H_{ex} \simeq 125$ kOe). The centers of these narrow absorption lines correspond precisely to the centers of magnetic phase transitions due to a magnetization reversal of the rare-earth ions. The positions of these lines are essentially independent of the field at all frequencies. For some of these lines, we observe a splitting into two very narrow lines (this type of splitting had been observed previously[2] in $Ho_{0.2}Y_{2.8}Fe_5O_{12}$) or into more than two very narrow lines ($\Delta H \simeq 1$ kOe). This behavior of $\nu(H)$, but without the splitting of lines, is also characteristic of resonances in fields both well below and far above H_{ex}. In a first approximation, these resonances are symmetric with respect to H_{ex} (group B). These lines differ markedly in width from the lines of group A ($\Delta H \simeq 25$ kOe), and their positions along the field scale are not related to any changes in the static magnetic characteristics of the rare-earth iron garnet. The absorption lines of a third type (C) are observed in fields below 60 kOe. For the resonances of this type, we were able to plot $\nu(H)$ curves in only certain cases (solid lines $C1$ in Figs. 1 and 2 are dashed lines $C2$). The reason was that the experiments which we are reporting here were aimed at learning about resonance effects in strong fields, so we did not obtain enough information to determine the resonance picture in weak fields. It was found that as the frequency increased, at $\nu > 90$ GHz, a set of additional absorption maxima appeared in fields below 20 kOe. Nevertheless, it was found possible to establish that the energies of the excitations corresponding to the $C1$ lines are identical at $H = 0$. This energy gap is anomalously small for rare-earth iron garnets ($\Delta E \simeq 57$ GHz), and it is essentially isotropic.

As we know, in fields such that lines A are present, an instability of the magnetic structure of the crystal occurs as a result of a crossing of levels of the rare-earth ions.[5] It is thus logical to link the resonant microwave absorption near H_{ex} with a reversal involving the ground state of the rare-earth ion, i.e., with a change in the sign of its projection onto the easy axis. However, even when the levels of the low-lying doublet are as close together as possible, there remains a gap of about 5 cm^{-1} between them. This gap is due to the crystal field.[5] Consequently, the resonances at $H \simeq H_{ex}$ cannot be interpreted as transitions between states of the holmium ion, since the frequencies used in the experiments were considerably lower. However, the process by which the magnetic moment of the rare-earth ion rotates ($\vec{M}_k^R$, $k = 1,...,6$) is accompanied by an excursion of the magnetic moment of the iron subsystem ($\vec{M}^{Fe}$) from the field direction. The observed absorption maxima would then logically be associated with transitions between the ground and metastable states in the iron subsystem, which exist only near phase transitions, along with corresponding reorientations of $\vec{M}_k^R$. There is a continuous spectrum of these transitions, and the curves of $\nu(H)$ would be similar to those observed experimentally (see the rightmost line in group A in Fig. 1). The absence of any apparent splitting of the other lines of this group as the frequency is raised can be explained on the basis of the substantial intrinsic width of these lines.

The most interesting experimental result is the observation of absorption lines of type B, for which the nature of the $\nu(H)$ curves is the same as that of the lines near H_{ex}. The symmetry of the positions of these lines with respect to H_{ex} suggests that holmium ions participate in their formation: Their states are essentially identical. However, the distance between the two lower levels of the rare-earth ion in such fields is large, and we can say nothing about just which transitions would be induced between them. In addition, the $\nu(H)$ behavior (in comparison with that of the lines of group A) indicates a participation of the iron subsystem of the crystal in this process. Since there are no sharp or substantial changes in the magnetization in the fields in which the absorption lines of B are found, one might suggest that in this case there is a substantial softening of the original rigid rare-earth magnetic structure, accompanied by an increase in the transverse components of $\vec{M}_k^R$ due to the interaction of the main quasidoublet of the Ho^{3+} ion with higher-lying levels. In this case the average magnitude of the projection of $\vec{M}^{Fe}$ onto the field would also decrease, because of an increase in the amplitude of its oscillations, but the difference between $\vec{M}^{Fe}$ and $\Sigma\vec{M}_k^R$ would remain the same. The presence of the lines of type B can thus be linked with the formation of a dynamic state in the magnetic system of the rare-earth iron garnet in which oscillation modes are excited without a change in the nature of the magnetic structure of the iron garnet.

The curves of $\nu(H)$ for the resonances of type $C1$ are similar to those which have been seen previously[2] in $Ho_{0.2}Y_{2.8}Fe_5O_{12}$, but the energy gap at $H=0$ in this case is slightly greater (the only change from Ref. 2 was in the concentration of rare-earth ions). This circumstance, combined with the observation that this absorption disappears at the frequencies and fields at which the $\nu(H)$ curves for groups A and C cross, is clear evidence that the presence of lines C is a consequence of oscillations of $\vec{M}_k^R$. These oscillations in turn cause oscillations in the "soft" iron subsystem. One might suggest that these coupled oscillations are similar to the oscillations which are excited at an antiferromagnetic resonance in an antiferromagnet.[4] Further evidence for this conclusion comes from the nature of the field dependence of the $C1$ lines.

At this point we do not have enough information to interpret the other lines of type C. All we can do is offer the following suggestions. One possible reason for the absorption of the radiation in this case might be the excitation of size resonances in the sample. An indication of such resonances comes from the uniform spacing of the absorption maxima along the field scale at comparatively low frequencies. Further evidence comes from the appearance of additional absorption lines between these lines as the frequency is raised. In addition, we cannot rule out the possibility that the reason for the existence of a large fraction of the C lines is the excitation of oscillations of an antiferromagnetic-resonance type. Evidence for this idea comes from the analogy between the $\nu(H)$ curve for the $C1$ lines and the $\nu(H)$ curves for the resonances of group $C2$ (the dashed lines in Figs. 1 and 2). That result, however, should be regarded as no more than tentative, since a definitive determination of the field dependence of the $C2$ lines will require some more-careful experiments, with an even smaller step along the frequency scale in fields up to 4 T.

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