Investigation of resonance properties of an Ising ferrimagnet in strong magnetic fields

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Absorption of microwaves in mixed yttrium–holmium iron-garnet single crystals were investigated at temperatures 4.2 and 77 K and in fields oriented along the [111] axis. A number of absorption lines beside the ferrimagnetic resonance—broad ($\Delta H \approx 30$ kOe) and narrow ($\Delta H \approx 2$ kOe)—were observed in fields from 30 to 160 kOe at frequencies from 35 to 60 GHz. Some oscillation modes of the magnetic subsystem of the crystal in a zero field were found to have an anomalously narrow gap [$\nu(0) \approx 45$ GHz]. Most of the results are well described qualitatively by the model of inversion of the holmium-ion ground state.

INTRODUCTION

The linear dynamics of the magnetic subsystem of rare-earth iron garnets (REIG) have been investigated to date much less than its stotic properties.[1] Only one of the many oscillation modes of this subsystem, ferrimagnetic resonance (FMR) has been investigated, and as a rule at frequencies up to 30 GHz and in weak magnetic fields.[2]

Recent investigations of magnetic resonance in terbium–yttrium iron garnets[3] have revealed in strong magnetic fields the presence, on top of FMR, of several absorption lines in the frequency range 30–100 GHz. The number of these lines correlates rigorously with the number of phase transitions induced by an external magnetic field. This circumstance, as well as the field dependences of the magnetic-resonance frequencies, were interpreted in the context of a model with intersection of the levels of the rare-earth (RE) ion. However, the large width of the resonance lines in fields on the order of the field H_{exc} of the exchange interaction between the RE and iron sublattices has made it impossible to observe all the regularities predicted by this model for resonant absorption of electromagnetic radiation upon inversion of the RE ion states. It was proposed[3] that the appreciable resonance linewidth is due to the presence of a substantial interaction of two lower levels of the Tb^{3+} ion, a fact explaining also why the Ising approximation[4] cannot be used to describe the properties of terbium–yttrium iron garnets. The chosen objects of a detailed verification of the model of resonance phenomena in REIG with level inversion were therefore Ising magnets—mixed yttrium–holmium iron garnets with small substitution of yttrium for holmium. The magnetic properties of such compounds are well described by taking into account only two noninteracting singlets, i.e., in the effective-spin $S = 1/2$ approximation.[1]

PROCEDURE, SAMPLES, AND EXPERIMENTAL RESULTS

Magnetic resonance was investigated in pulsed magnetic fields up to 200 kOe using a reflecting spectrometer, at an approximate field-pulse duration 40 msec. The waveguide channel was an assembly of standard 8-mm elements. The detector was an InSb single crystal cooled with liquid helium. The microwave sources were reflex klystrons and backward-wave tubes, and the working-frequency range was 33–70 GHz. The signals from the detector and from a probing field coil wound on top of the waveguide near the shot-circuiting bottom were fed to a DL1200 single-process recorder, and next in digital form to an IBM AT PC computer. The experiments in the pulsed fields were performed at 4.2 and 77 K. The FMR in dc fields (up to 8 kOe) was investigated with a "Brücker" EPR spectroscope at temperatures from 20 to 300 K and at a frequency 9.45 GHz.

The investigated objects were $Ho_{0.2}Y_{2.8}Fe_5O_{12}$ single-crystals previously used[5] for magnetic measurements. The external field was always parallel to the [111] axis of the cubic crystal, and the error of orienting the sample relative to the external field did not exceed 5°. The samples used for experiments in pulsed fields were cylinders 3 mm in diameter and 0.5, 2.5, and 5 mm long, whereas in dc fields we used cubes with approximately 0.3 mm on a side.

A characteristic field dependence of microwave absorption at $T = 4.2$ K is shown in Fig. 1. The dependence of the magnetic-resonance frequencies on the external magnetic field, plotted on the basis of an analysis of the absorption spectra, are given in Fig. 2. The frequencies of lines of type

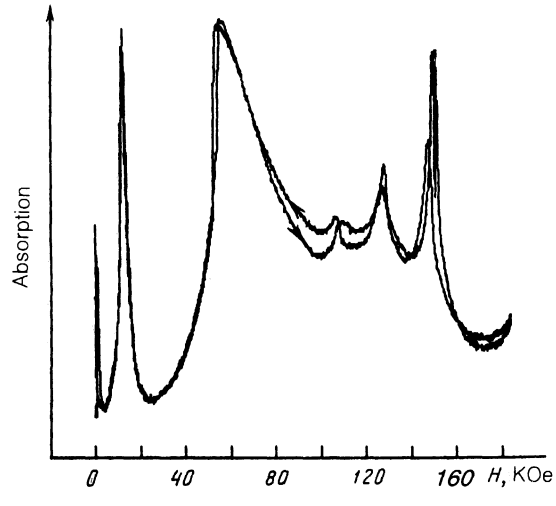

FIG. 1. Absorption of 40.09-GHz signal reflected from a sample for increasing and decreasing magnetic field, $T = 4.2$ K. The arrows indicate the direction of the change of the external magnetic field.

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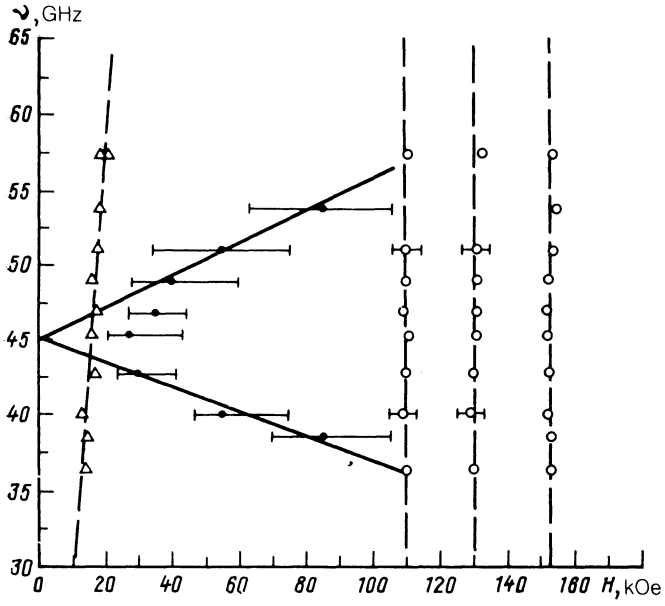

FIG. 2. Field dependences of the absorption maxima in single-crystal $Ho_{0.2}Y_{2.8}Fe_5O_{12}$ at $\mathbf{H}\|[111]$ and $T = 4.2$ K; Δ—absorption line of type "A"; ●—type "B"; ○—type "C." The horizontal segments indicate the half-widths of the resonance lines.

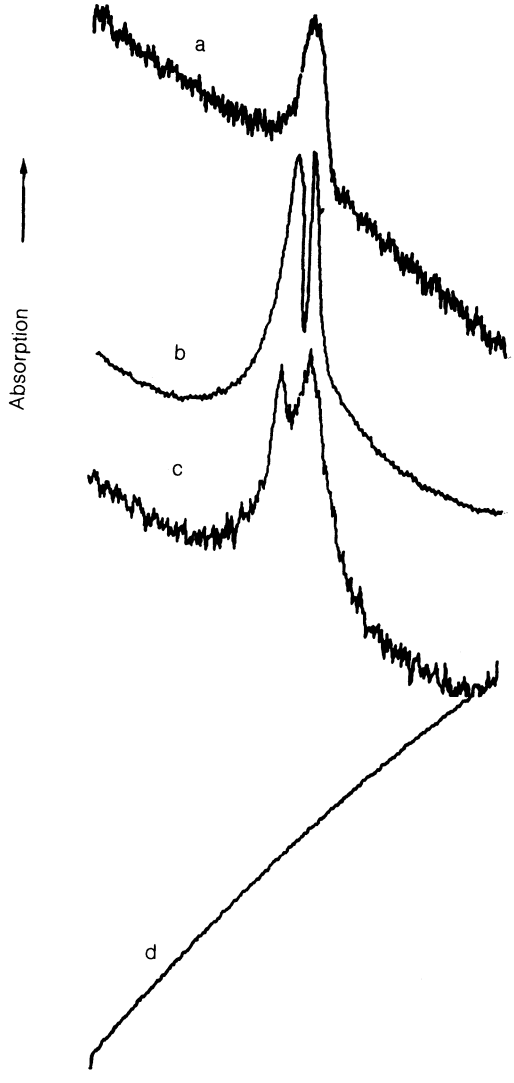

FIG. 3. Variations of absorption line of type "C" at $H = 152$ kOe and with increase of frequency ($T = 4.2$ K). a—36.42 GHz, b—40.09 GHz, c—51 GHz, d—variation of external magnetic field with time near $H = 152$ kOe.

"A" are linear in the magnetic field, and extrapolation to $H = 0$ (dashed) yields $\nu = 0$. The resonance for the line "B" is in first approximation likewise a linear function; as the field is decreased, the (frequency) spacing between the lines in a group decreases, and extrapolation to $H = 0$ (solid line in Fig. 2) yields the energy gap for oscillations of this type: $\Delta\nu = 45$ GHz. The positions of the resonance lines in the third group are practically independent of the field, and a frequency splitting into two very narrow lines is observed for the most intense line (Fig. 3).

The field dependences of the absorption maxima at $T = 77$ K are shown for various frequencies in Fig. 4. Just as at $T = 4.2$ K, an absorption band with $\nu = \gamma H$ is present (γ is the gyromagnetic ratio). A line of type "B" is likewise observed, and to first approximation the gap width is the same as for $T = 4.2$ K. In place of three lines one broad line exists in fields of order $H_{exc} \approx 125$ kOe one broad line whose center almost agrees with one of the type "C" lines at helium temperature (see Figs. 3 and 4). In addition, one more absorption line was observed in a 60 kOe field, was likewise not shifted over the field when the frequency was varied, and split into two at high frequency.

Results of the investigation of magnetic resonance in the smallest samples in a constant field, at various temperatures and microwave power levels, are shown in Fig. 5. When the temperature is lowered the FMR line shifts towards weaker fields, and its amplitude decreases sharply, so that to observe a distinct absorption the microwave-power level was changed by five orders when the temperature was varied from 300 to 20 K. For the same reason, apparently, no resonance absorption could be observed in samples less than 1 mm thick in fields of the order of H_{exc}. It was therefore necessary to use samples comparable in dimension with the wavelength, which in turn complicated significantly the picture of the observed phenomena, owing to interference and to size-effect resonances. To exclude the influence of the former, the experimental frequencies were chosen such that the resonances were not subject to significant dispersion. The size-effect resonances were investigated by comparing the obtained for samples of equal thickness. Figures 2 and 4 show thus only those lines which were observed for all samples independently of size.

DISCUSSION OF RESULTS

It is logical to interpret the $\nu = \gamma H$ dependence for lines of type "A" in Figs. 2 and 4 as FMR, the more so because the obtained γ are close to the values given in Ref. 2 for the same temperatures.

The absorption line centers at $H \approx H_{exc}$ and $T = 4.2$ K agree exactly with the fields of the phase transitions due to reorientation of the RE magnetic moment investigated in Refs. 5 and 6, and are not shifted when the frequency is varied. With increase of frequency, some lines (the most intense ones) are split into two—symmetrically about the field of the magnetic phase transition, which is due to inversion of

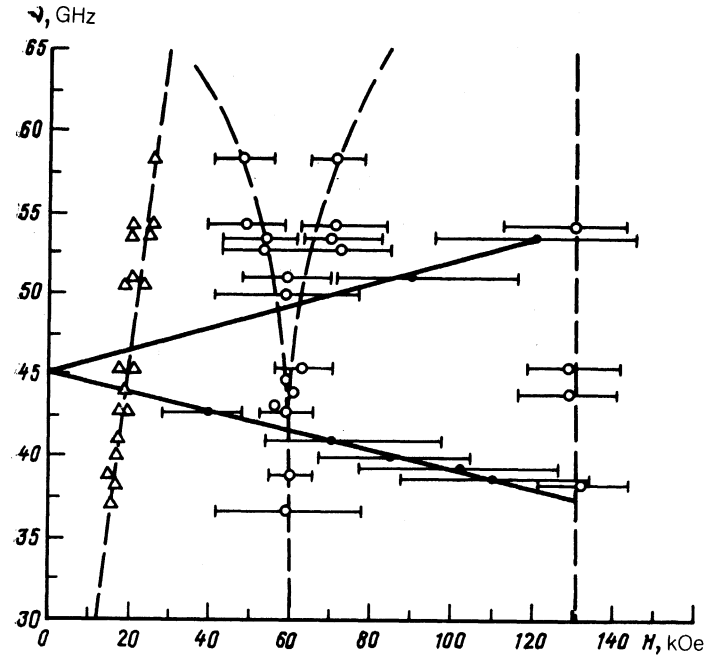

FIG. 4. The same as Fig. 2, for $T = 77$ K.

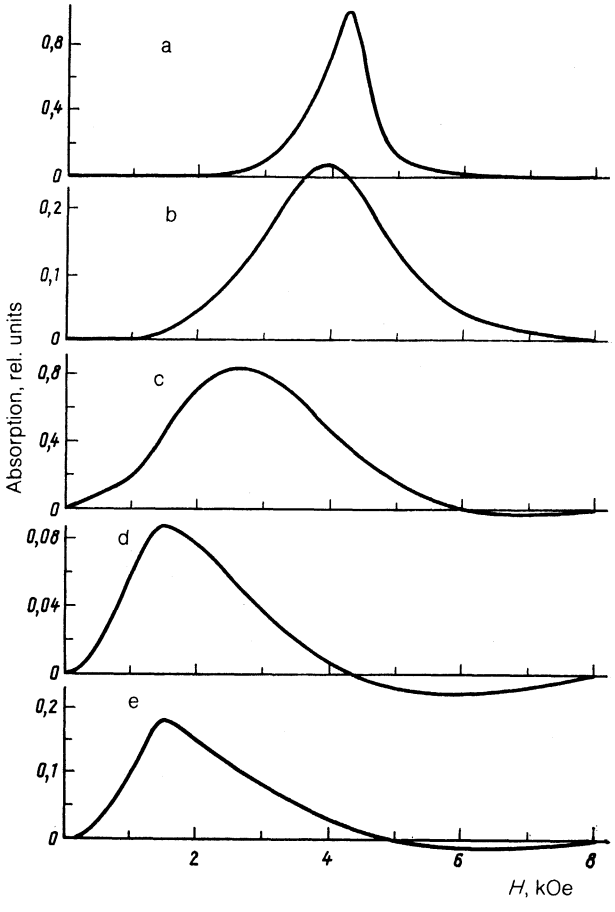

FIG. 5. Evolution of FMR line in $Ho_{0.2}Y_{2.8}Fe_5O_{12}$ with change of temperature (H||[111]; a—300 K (2 μW); b—150 K (20 μW); c—100 K (2 mW); d—77 K (20 mW); e—40 K (200 mW). All the curves are reduced to a single scale, and the parentheses contain the microwave power used to observe the resonance. Curves c–e are shifted along the ordinate axis so that the absorption zero is taken to be the absorption in a zero magnetic field.

the ground state of the Ho^{3+} ion. This should precisely be the character of the field dependences of the magnetic-resonance frequencies according to the model proposed in Ref. 3 for resonance phenomena in crossings of energy levels of an RE ion in an REIG. It can therefore be stated that the microwave absorption maxima observed at $H \approx H_{exc}$ are the results of induced transitions of the Ho^{3+} ion from the ground state to an excited one—the analog of EPR in a magnetically ordered crystal. On the other hand, the intensity difference of lines of type "C" is apparently due to the change of the sample temperature by the adiabatic demagnetization of the RE subsystem of the crystal, a process consisting at helium temperature of three magnetization jumps at fields $H \approx H_{exc}$.

Broad absorption lines at $H \approx H_{exc}$, which are likewise not shifted over the field with change of frequency, were observed also at $T = 77$ K, much higher than the critical $T_{cr} = 18$ K above which smooth reversal of the magnetization of the RE subsystem takes place without formation of canted phases.[6] Near the inversion point of the holmium-ion levels there is nevertheless a small difference between the populations of the ground and excited states, and an external microwave field will induce resonance transitions between them. Raising the temperature enhances the lattice vibrations, and this is the cause of the significant broadening of the line of type "C" at $T = 77$ K. In turn, the latter explains why such resonances are not split when the frequency is raised.

At 77 K (see Fig. 4) there is observed one more $\nu(H)$ mode at $H \approx 60$ kOe, and its form is typical of the described resonance phenomena accompanying RE-ion level crossing. The splitting of the ground quasidoublet of Ho^{3+} in such fields[7] is much larger than the quantum energy of the radiation used in this experiment, so that it is impossible to attribute the observed absorption maxima to transitions within this quasidoublet. It is known,[7] however, that in a zero

field the second excited level is located close to the first-excited one (15–20 cm^{-1}), and the dependence of the latter will be strong enough because of the high temperature. It can therefore be assumed that the observed resonance is due to the crossing, in an approximate field 60 kOe, of the first and second excited levels of the Ho^{3+} ion.

The interpretation of absorption lines of type "B" meets with the greatest difficulty. It is known that in the field region in which these lines are observed there are no magnetization of susceptibility singularities whatever,[5,6] so that it is impossible to relate the observed absorption to some reorientation process in the magnetic structure of the garnet. The vanishing of resonance lines of type "C" in the frequency region where the field dependences $\nu(H)$ of lines of type "B" and "C" cross indicates that their common "source" is a change of a holmium-atom state. The available experimental data, however, are insufficient to explain the origin of lines of type "B." It is necessary for this purpose to obtain investigations, similar to those described above, of magnetic resonance in samples of other compositions, and study the influence of the magnetic field orientation in its properties.

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