Dynamical magnetic phase diagrams of a magnetic material with strong anisotropy

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(Submitted 15 January 1992)
Zh. Eksp. Teor. Fiz. **102**, 262–270 (July 1992)

The absorption of microwave radiation in a $Ho_{0.4}Y_{2.6}Fe_5O_{12}$ single crystal at $\mathbf{H}\|[111]$ in the temperature range from 4.2 to 150 K is studied within broad ranges of frequencies (40–96 GHz) and magnetic fields (up to 250 kOe). Dynamical magnetic phase diagrams are constructed in the $(H_{res}-T)$ plane, with H_{res} the resonance field. In addition to the branches of the phase diagram known from static magnetic measurements a whole family of new branches has been discovered. A model is suggested for a qualitative description of the discrepancies between the results of static and dynamical measurements that is based on the assumption of a dynamical magnetic Jahn–Teller effect being present in the compound under investigation.

INTRODUCTION

The present study deals with the results of an experimental investigation of the transformation, induced by an external magnetic field, of the magnetic structure of multisublattice ferromagnets with strong crystallographic anisotropy. The fact that the anisotropy is large means that the effective fields of magnetic anisotropy in investigated materials are commensurable with, or are even higher than, certain effective fields of the exchange interaction between the sublattices of the magnetic material.

The investigated objects were rare-earth iron garnets (REIG) of the $Ho_xY_{3-x}Fe_5O_{12}$ system, whose elementary cell contains eight formula units. The magnetic structure of REIG constitutes a set of eight magnetic sublattices: two nonequivalent iron sublattices (the *d*- and *a*-sites in the cell) and six nonequivalent rare-earth sublattices (*c*-sites). The exchange interaction of the antiferromagnetic type between the iron sublattices is very strong ($H_{exch}^{Fe-Fe}\approx 2\cdot 10^6$ Oe; see Refs. 1 and 2). Consequently, in substantially weaker fields these sublattices can be considered a common sublattice with the resulting moment $\mathbf{M}_{Fe}$ equal to the difference in the moments of the *d*- and *a*-sublattices. The exchange interaction second in strength, also of the antiferromagnetic type, is the one between the iron and rare-earth subsystems ($H_{exch}^{R-Fe}\approx 10^5$ Oe; see Ref. 2), which causes the formation in a zero magnetic field of the RE structure in the shape of two cones whose axis points in the [111] direction, with all six RE moments approaching the edges of the cubic elementary cell as the concentration of holmium ions decreases.[3] The latter makes it possible to represent the RE magnetic structure of an iron garnet in the form of three sublattices whose moments are oriented along the [100], [010], and [001] directions, respectively, so that the resulting moment $\mathbf{M}_R$ is directed along the body diagonal of the cube.

The exchange interaction between RE sublattices is even weaker (by a factor of ten and is not taken into account in the present study. Thus, within the chosen approximation, the magnetic system of REIG consists of the exchange-ordered iron subsystem and the paramagnetic rare-earth subsystem in the strong magnetizing field generated by the iron magnetic moments.

The field dependences of the magnetization of single crystals of $Ho_xY_{3-x}Fe_5O_{12}$ at low temperatures measured earlier,[4] showed that the behavior of the RE magnetic subsystem can be described fairly well on the assumption that the ordering of holmium ions is of the Ising nature. This means that the RE magnetic moments are rigidly "fixed" to the local Ising axes (directions of the [100] type) and can only change the sign of their projection onto these axes in the process of transformation of the magnetic structure. Here, in a growing external field, the moment $\mathbf{M}_R$ changes its direction in a discrete manner from one type-[111] direction to another. Such jumps in $\mathbf{M}_R$ are accompanied by a change in the initial orientation of $\mathbf{M}_{Fe}$. As a result, in fields of the order of effective exchange between the iron and RE subsystems the magnetization of the entire garnet experiences jumps whose number depends on the orientation of the external field with respect to the crystallographic axes.

The mechanism of all such magnetic phase transitions is the same: the external field compensates H_{exch}^{R-Fe} in one of the nonequivalent *c*-sites, so that the state of the respective RE ion becomes degenerate. The degeneracy is lifted by the change in sign of the projection of the RE moment and, hence, the change in the orientation of $\mathbf{M}_R$ (see Ref. 5). Here it is important to mention the uniqueness of the holmium-yttrium system: no other structural phase transitions induced by a field exist in it, that is, far from H_{exch}^{R-Fe} no anomalies in the behavior of magnetization are observed.

STATEMENT OF THE PROBLEM AND THE EXPERIMENTAL RESULTS

The model suggested by Silant'ev *et al.*[4] for describing the static magnetic properties of holmium-yttrium ferrite garnets at low temperatures made it possible to describe for various orientations of the external field the magnetic phase diagrams of these crystals in the "field–temperature" and "field–concentration" planes not only qualitatively but in many cases even quantitatively. Yet the dynamics of the magnetic system of crystals under transitions induced by a field remained until recently practically unknown not only for the system under discussion but for all REIG, though the anomalies in the static properties could have been expected to influence the dynamical characteristics of such crystals, say, the behavior of the magnetic resonance lines.

Indeed, the very first experiments in absorption of microwave radiation in holmium-yttrium iron garnets in

 0038-5646/92/070138-04$05.00

strong magnetic fields[6,7] revealed that the magnetic resonance pattern in such crystals is very complex and differs markedly from the "classical" resonance of weakly anisotropic magnetic materials. It was discovered, for one thing, that in addition to the branches of ferromagnetic and exchange (the analog of AFMR in ferrites) resonances, there is a large number of microwave-radiation absorption lines in no way correlated (in the field) with the above-mentioned structural magnetic phase transitions. In Ref. 7 it was suggested that a fraction of these resonances (not related to phase transitions or to any anomalies in the static magnetization or susceptibility) can be explained by the sizable "softening" in some fields of the initial rigid magnetic structure of the RE magnetic subsystem (i.e., the emergence of transverse components in the RE moments) and the formation of a special dynamical state of the crystal's entire magnetic system. It was assumed that in this state such coupled vibrations are excited as do not change the initial type of the magnetic structure. This explains the absence of noticeable magnetization variations in fields that are distant in magnitude from $H_{\text{exch}}^{\text{R-Fe}}$ and in which a new dynamical state of the crystal exists.

To obtain additional arguments in favor of the hypothesis put forward in Ref. 7, magnetic resonance studies have been conducted in a broad range of wavelengths of electromagnetic radiation in a $Ho_{0.4}Y_{2.6}Fe_5O_{16}$ single crystal, with the external magnetic field aligned with the [111] axis of the cubic crystal, at temperatures ranging from 4.2 to 150 K. The latter temperature is much higher than the critical temperature $T_c \approx 25$ K that limits the region where static phase H–T diagrams exist for the given composition;[4] for $T > T_c$ reversal of magnetization of a RE ion takes place without jumps, via smooth demagnetization to zero and, subsequently, smooth magnetization.[5] The goal of the present investigation was to build dynamical phase diagrams in this plane and compare them with static diagrams so as to substantiate the assumption that far from $H_{\text{exch}}^{\text{R-Fe}}$ the magnetic system of the crystal can exist in stable dynamical states.

Magnetic resonance studies were conducted at several fixed frequencies in the 40 to 96 GHz range on a millimeter-wave reflectance spectrometer in pulsed magnetic fields up to 250 kOe (the method is detailed in Ref. 6). The single crystals used in the present investigation were earlier used in the experiments conducted by Silant'ev *et al.*[4] to measure the field and temperature dependences of magnetization and had the shape of rectangular parallelepipeds that measured 1.5×1.5 mm^2 and were 6–8-mm long. Temperature scanning was maintained by a flow cryostat. The set of radiation frequencies used in the study was not accidental: it encompassed the most interesting sections on the frequency vs resonance-field dependences obtained in Ref. 7 at $T = 4.2$ K.

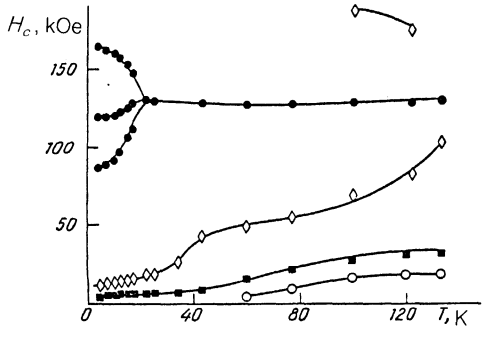

FIG. 1. Temperature curves for the fields H_c corresponding to the peaks in the absorption of microwave radiation in a $Ho_{0.4}Y_{2.8}Fe_5O_{12}$ single crystal at $\mathbf{H} \| [111]$ and obtained at a probing frequency of 40.5 GHz. Different labels stand for different absorption lines.

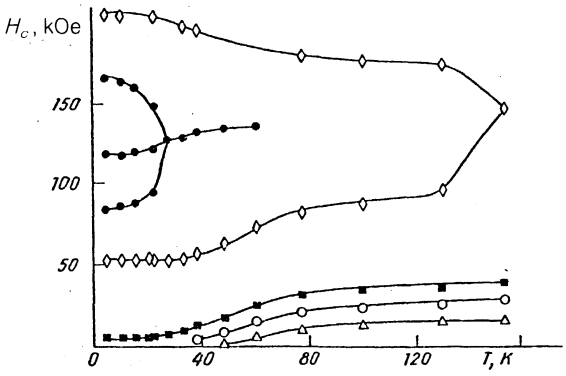

FIG. 2. The same as in Fig. 1 but at $\nu = 50.7$ GHz.

The microwave-radiation absorption spectra were analyzed with the aim of determining the fields H_{res} corresponding to the peaks of absorption, with the result that dynamical H_{res}–T phase diagrams were set up for each frequency. Such diagrams for the five frequencies of the probing radiation are depicted in Figs. 1–5. Different labels in the figures correspond to different absorption lines. The static phase diagram obtained in Ref. 4 from the results of magnetization measurements practically coincides with one of the sections of the dynamical diagram in Fig. 3 (the curves with ●). The only difference is that instead of H_{res} in Fig. 3 we have equal magnitudes of the fields of magnetic phase transitions. It is important to note that the static diagrams contain no other branches. The branches existing at temperatures below T_c are related to the sudden reversal of magnetization of a RE ion at one of the nonequivalent sites.

Comparison of the static diagram with dynamical diagrams suggests the following conclusions. First, all diagrams contain similar branches (type A) that separate the states with different orientations of $\mathbf{M}_R$ (the ● in the figures) and are rigidly linked in the fields to the points of magnetic phase transitions. Below T_c such transitions occur, as noted ear-

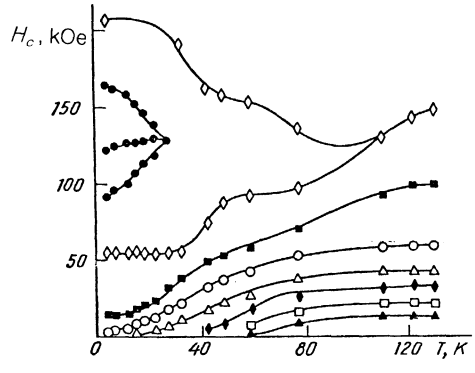

FIG. 3. The same as in Fig. 1 but at $\nu = 70.8$ GHz.

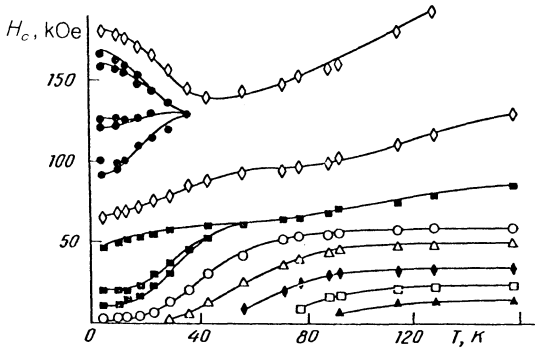

FIG. 4. The same as in Fig. 1 but at $\nu = 89.6$ GHz.

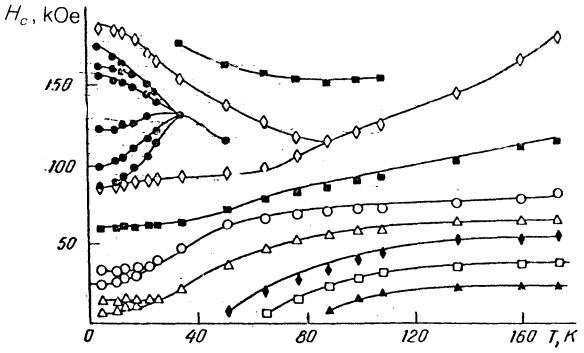

FIG. 5. The same as in Fig. 1 but at $\nu = 93.6$ GHz.

lier, because of the instability of the magnetic structure of the ferrite garnet in fields of the order of H_{exch}^{R-Fe}, where the ground state of 4*f*-ions is degenerate or close to it. The degeneracy is lifted by the deformation of the initial magnetic structure of the crystal; in other words, what is known as the static magnetic Jahn–Teller effect occurs.[3,8] In contrast to the static diagram, however, some dynamical diagrams contain continuations of these branches above T_c, in the region where in static conditions there is no jump in the moment. A characteristic feature of the absorption lines corresponding to *A*-branches (at low temperatures) is the presence of a soft mode (i.e., absorption of microwave radiation in a field of the given magnitude is possible at any frequency used in experiments from the very low to the very high) and the symmetric arrangement of H_{res} with respect to $H_{exch}^{R-Fe} \approx 125$ kOe (see Ref. 7). In addition, as the frequency grows, there is observed moderate splitting (in field strength) of some of the branches into two or three closely located.

Second, the dynamical diagrams have branches of type *B* (the ◇ and ■ in Figs. 1–5) encompassing the domain of the static diagram. What is evident is the existence of some kind of correlation between branches of the two types: the temperature dependences of *B*-branches are similar in shape to those of *A*-branches. The only difference is that the critical temperature T_c^* for the *B*-branches is much higher (≈ 120 K) and diminishes as the frequency grows; more precisely, as the probing frequency increases, the *B*-branches move closer to the static diagram. It should be especially noted that the characteristic features of type *B* resonances (the symmetric arrangement of H_{res} with respect to H_{exch}^{R-Fe} and the presence of a soft mode) are identical to the features of the resonances corresponding to the *A*-branches (see Ref. 7) but that the absorption lines for the *B*-branches are much wider (by a factor of ten) than those for the *A*-branches. Therefore, it proved relatively easy to single out the *A*- and *B*-branches among the multitude of resonances simply because of their specific field dependence (more precisely, the absence of dependence on the field under frequency variations at low temperatures) and because of their symmetric position with respect to H_{exch}^{R-Fe}.

Finally, there is one more group of branches (type *C*), for which the H_{res} are practically equidistant in field strength, are temperature independent for $T > 90$ K, and rapidly drop to zero as the temperature is lowered. These branches are related either to excitation in the sample of some sort of dimensional vibrations (magnetostatic modes) or to the interference of the signal reflected from the sample and the one passing through the sample, since the sample dimensions are commensurable with the wavelength of the radiation employed in the experiment. Attempts to single out a predominant mechanism in the conditions of a specific experiment by studying the evolution of these branches under changes in the sample's orientation in the waveguide proved unsuccessful. Minor variations in the dimensions of the sample were found to affect both mechanisms simultaneously, and it proved impossible to use sufficiently thin crystals (compared with the wavelength) to get rid of the interference reliably because of the drastic reduction in signal strength from the detector. Analysis of the evolution of the branches as the frequency increased did not provide a full answer either: both mechanisms of absorption-line formation in low fields lead to about the same increase in the number of absorption peaks in the observed spectra. Therefore, we will leave for future discussion the extent to which each mechanism affects the observed phenomenon and the possible alternatives, especially since the more interesting resonance phenomena (*B*-branches) are fully distinguishable from *C*-lines in the REIG under investigation.

DISCUSSION

As noted earlier, we can assume it proved that there is a static magnetic Jahn–Teller effect in holmium-yttrium iron garnets in fields corresponding to *A*-branches. It would be a mistake, however, to link the discovered resonance absorption of microwave radiation at magnetic phase transition points with the stimulated transitions between the ground and the first excited level of a RE ion (the analog of EPR), as noted in Ref. 7, because of the presence in the spectrum of the holmium ion of a sizable gap (≈ 5 cm^{-1}) between the ground and the nearest excited level caused by the splitting of the levels of the holmium ion in the crystal field. It appears that even at the highest frequencies used in the experiment this barrier between the ground and excited states of Ho^{3+} cannot be surmounted. The only solution is to recall that the deformation of the surroundings of the Jahn–Teller ion (in our case, the direction in which the moment of the iron subsystem is deflected) is not, in general, static.[9] Indeed, almost always there is a whole set of different deformations (in accordance with the symmetry of the problem; in our case there are three) corresponding to the same energy of the magnetic system of the crystal. The true wave functions (normal modes) are linear combinations of the wave func-

tions Ψ_k that depend on the specific direction of deflection of $\mathbf{M}_{Fe}$ and the corresponding variation in the orientation of $\mathbf{M}_R$. These linear combinations describe the combined motion of the RE subsystem with the motion of the iron moment. Such states are known as vibronic.[3,9] In this case they correspond to localized spin-wave modes incorporating the combined motion of the moment of a RE ion and the moments of the matrix. Thus, generally in the magnetic Jahn–Teller effect purely electronic degeneracy is also replaced with vibronic. Physically vibronic degeneracy describes the tunneling of the system between different energy minima corresponding to deformations in the picture of a "single" static Jahn–Teller effect. The latter has meaning if the tunneling frequency ν_{JT} is much lower than the characteristic frequencies of the problem. This condition is always assumed met in the cooperative Jahn–Teller effect, in which the tunneling frequency tends to zero. Hence, the soft mode discovered in experiments at low temperatures in fields that are close in strength to H_{exch}^{R-Fe} (the A-branches in dynamical phase diagrams) is most likely connected with these very tunneling transitions, that is, with the static magnetic Jahn–Teller effect.

The similarity in behavior of B- and A-branches as the temperature and probing frequency change and the presence of a soft resonance mode in fields symmetric with respect to H_{exch}^{R-Fe} unquestionably point to the closeness of the mechanisms causing these resonances. The statistical data,[4] however, makes it clear that there is no sizable deformation of the magnetic structure of the given garnet in fields corresponding to B-lines. Hence, such absorption cannot be linked with the magnetic Jahn–Teller effect. It is also known that the distance between the ground level and the first excited level of the holmium ion far from H_{exch}^{R-Fe} is great—no inversion of the ion state is possible. But in the vicinity of such a field such inversion is possible.

If we assume, however, that B-lines are caused by the dynamical magnetic Jahn–Teller effect rather than the static effect, then all the experimental data can be interpreted in a simple manner. Far from the exchange field there is also tunneling between some of the normal modes related to the simultaneous deflection of $\mathbf{M}_R$ and $\mathbf{M}_{Fe}$ from their initial positions. The only difference of B-lines and A-lines is that tunneling between the degenerate states for B-branches occurs at frequencies ν_{JT} exceeding those used in the experiments and that $\mathbf{M}_R$ does not change the sign of its projection on the local quantization axis while in motion.

The discovered soft mode in B-lines at low temperatures is the result of such tunneling, and the characteristic features of such a resonance must be close to the resonances of A-lines, as was revealed in the experiments. Moreover, it becomes clear why the phase diagrams obtained at frequencies low compared to ν_{JT} and, for one thing, the static phase diagram ($\nu_{probe} = 0$) do not contain B-branches: in the course of the measurements all variations in the magnetic system average out and the resulting variation of magnetization is zero. As the probing frequency is raised, the B-branches become more evident and evolve in such a manner that for $\nu_{probe} \gg \nu_{JT}$ they merge with the static line, since in this regime the transition from a truly dynamical Jahn–Teller effect to the static effect takes place. It is this change in the behavior of these branches that was observed in the experiments: the dynamical diagrams were attracted to the static (Figs. 1–5).

The similarity between the temperature dependence of B- and A-branches makes it possible in this case to say that the field induces in the garnet a new state that is truly dynamical and has a critical temperature $T_c^* > T_c$. The difference in the temperatures is due to the fact that formation of the dynamical states involves higher levels of the holmium ion, separated from the ground level by $\Delta E \approx kT_c^*$. In other words, far from H_{exch}^{R-Fe} the ground state of the Ho^{3+} ion is not a truly Ising state; there is a strong admixture of excited states, which determines the appearance of transverse components in the RE magnetic moment, that is, the initial rigid RE magnetic structure "softens."

On the other hand, all static diagrams in the "field-concentration" and "field-temperature" planes and the field dependences of magnetization for holmium-yttrium iron garnets are described fairly well within the framework of the Ising model[3–5] not only qualitatively but also quantitatively if we allow for only the lowest (on the energy scale) quasidoublet in the spectrum of the Ho^{3+} ion. This dilemma can be resolved by assuming that as the field strength grows, a transition from a strongly anisotropic non-Ising magnetic material to an Ising one occurs. The reason for such a transformation is the decrease in the splitting of the quasidoublets of the holmium ion with increasing field strength due to compensation of the exchange field by the external field, as a result of which the admixture of excited states to states of the lower quasidoublet (which is considerable in low fields because of the large splitting of both the main and the excited quasidoublets) also diminishes. Consequently, near H_{exch}^{R-Fe} a situation is realized in which the spectrum of Ho^{3+} constitutes a set of nearly degenerate quasidoublets, and the lowest quasidoublet can be considered isolated at sufficiently low temperatures to within a fairly good approximation. It is this essentially that underlies the basis for the Ising behavior of holmium ions for $H \approx H_{exch}^{R-Fe}$.

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Translated by Eugene Yankovsky