Magnetization anomalies in yttrium-erbium and yttrium-holmium iron garnets in strong fields at low temperature

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The magnetization of yttrium iron garnet single crystals doped with holmium or erbium is measured in fields up to 200 kOe and at helium temperatures along different crystallographic directions. Anomalies in the magnetization of the ferrimagnets are observed in strong magnetic fields. It is shown that the anomalies in iron garnets with erbium impurities are due to the appearance of a noncollinear magnetic structure of the iron and erbium sublattices. The field dependence of the magnetization anomalies of iron garnets doped with holmium is different. It is suggested that these anomalies are due to the crossing of the ground and excited levels of the Ho^{3+} ion in the field.

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INTRODUCTION

Rare-earth iron garnets (REIG) (formula type: $R_3Fe_5O_{12}$, where R is a rare earth or yttrium) have a cubic structure (space group O_h^{10}) and can be treated as two-sublattice ferrimagnets, one made up of iron ions Fe^{3+} occupying octahedral (a) and tetrahedral (d) sites, and the other of rare-earth (RE) ions R^{3+} occupying dodecahedral (c) sites.[1] Magnetic ordering in the RE sublattice is produced by indirect, a comparatively small ($H_{eff} = 10^5$ Oe), exchange interaction of the RE and iron ions. Besides, there is a crystalline electric field of the oxygen environment (symmetry D_2) acting upon the RE ions in REIG.

Combined action of the exchange and crystalline fields produces a complex splitting of the ground multiplet of the RE ion in these compounds. An interesting feature of the multiplet energy structure of some RE ions in REIG is the presence of excited energy states close to the ground state and their possible convergence at certain orientations of magnetic moments referred to the crystallographic axes. This was first shown by a study of the anomalies of ferromagnetic resonance in RE-doped yttrium iron garnet.[2,3]

It should be noted that theoretical interpretations of anomalies of ferromagnetic resonance in REIG assume that the external field causes only rotation of the magnetization, and do not take into account the effect of the field on the magnitude and character of the splitting of the energy levels of RE ions. However, the existence of excited energy states close to the ground state may be expected to produce another effect, namely, crossing or near-crossing of the ground and of one of the excited states in a sufficiently strong field. This phenomenon (inversion of energy levels in a magnetic field) was theoretically predicted by Cooper[4] for paramagnetic RE compounds with cubic crystal fields. As already noted, besides the crystal field, the exchange interaction of the iron sublattice affects the splitting of the energy levels of RE ions in REIG. Therefore external fields comparable in strength with the exchange field are necessary to observe the effect of crossing (or near-crossing) of energy levels in these compounds.

Earlier[5] we have observed anomalies of magnetization and magnetostriction in yttrium-terbium iron garnets at 4.2 K in the fields of the order of 10^5 Oe, which we have interpreted as due to the inversion of energy levels. But such an interpretation is not unique, as noted in[5], since it is possible that a noncollinear magnetic structure of RE and iron sublattices may arise in the field.[6,7] And although the experimentally observed magnetization and magnetostriction anomalies in yttrium-terbium iron garnets differ from those theoretically predicted for noncollinear magnetic structures, we cannot exclude the theoretical possibility that the rise of the latter accounts for the phenomena observed in[5]. A better understanding of the nature of the observed anomalies calls for a systematic study of magnetic, magnetoelastic, optic and other properties of iron garnets with various RE ions in strong fields.

Experimental study of magnetization of holmium- and erbium-doped yttrium iron garnets is reported in this paper. The doping ions are chosen because their energy levels split differently in the crystal and the exchange fields. The ion Ho^{3+}, as well as the ion Tb^{3+} is a non-Kramers ion with two excited states close to the ground state in a crystal field of symmetry D_2[8] (Fig. 1a). Therefore, exchange and external fields of the order of 10^5 Oe (~ 10 cm^{-1}) would have a strong effect on the splitting of the energy levels of the Ho^{3+} ion in the iron garnet structure, and inversion of the levels in holmium-doped iron garnets in strong fields is possible. The ion Er^{3+} is a Kramers ion and its first excited state in the iron-garnet structure is separated by about several dozen cm^{-1} from the ground state[9,10] (Fig. 1b). Consequently, both exchange and external fields of the order of 10^5 Oe would have a weaker effect on the energy spectrum of ion Er^{3+} in iron garnets, and inversion of levels in the field is much less likely.

SAMPLES AND MEASUREMENT TECHNIQUE

Single crystals of yttrium-holmium and yttrium-erbium iron garnets, $Ho_xY_{3-x}Fe_5O_{12}$ ($x = 0.25$; 0.41) and $Er_xY_{3-x}Fe_5O_{12}$ ($x = 0.12$; 0.52), were grown by crystallization from the solution in the melt. The magnetiza-

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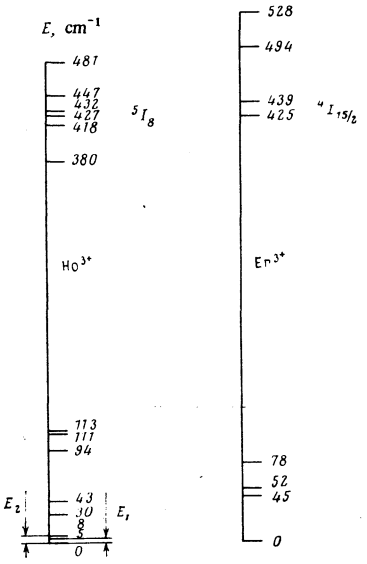

FIG. 1. Energy splitting of the ground multiplet of Ho^{3+} and Er^{3+} in yttrium iron garnet.

tion was measured on cylindrical samples cut from the single crystal, 3 mm in diameter and 4–6 mm in length, with the cylinder axis along the [111], [110] or [100] direction. The accuracy of the orientation was within 2–4°.

The magnetization was measured by the induction technique in pulsed magnetic fields with a pulse width 6 msec.[111] The measurement error of the absolute value of the magnetization was 10%; the relative error of the measured value of the magnetization (as a function of the field, temperature etc.) amounted to 3–5%.

EXPERIMENTAL DATA

Figure 2 shows field plots of the magnetization of the iron garnet $Er_{0.12}Y_{2.88}Fe_5O_{12}$ along different crystallographic directions at 4.2 K. It can be seen that technical magnetization phenomena cease in the fields of 10–20 kOe,[1)] magnetization reaches saturation in stronger fields, but in the field interval 70–150 kOe the magnetization rises again gradually along the [111] and [110] axes and more rapidly along the [100] axis (measurements in weak fields have shown that this is the direction of easy magnetization). In fields above 150 kOe the magnetization reaches saturation again. The magnetization exhibits greater hysteresis in strong fields along the [100] axis and much less along [110] and [1$\bar{1}$1].

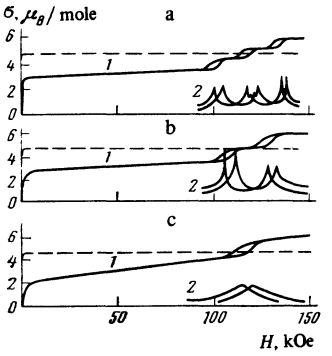

FIG. 3. Magnetization (1) and differential susceptibility (2) of a single crystal iron garnet $Ho_{0.25}Y_{2.75}Fe_5O_{12}$ at 4.2 K as a function of the field: a) H along [111], b) H along [110], c) H along [100] (solid lines); dashed lines—magnetization of yttrium iron garnet.

The field dependence of the magnetization of the iron garnet $Er_{0.52}Y_{2.48}Fe_5O_{12}$ is similar, but the field interval in which the magnetization is seen to rise is wider (60–170 kOe).

The behavior of the magnetization of yttrium-holmium iron garnets in strong fields is completely different. This can be well seen in Fig. 3, which shows the field dependences of the magnetization of iron garnet $Ho_{0.25}Y_{2.75}Fe_5O_{12}$ along different directions at 4.2 K. This iron garnet also reaches technical saturation in comparatively weak fields, its magnetization increases slightly in fields up to 100 kOe followed by a considerably sharper increase with one jump along the [100] axis, two jumps along [110], and three jumps along [111]. These jumps are subject to hysteresis, which is particularly well seen on the plots of differential susceptibility.[2)] As the temperature increases magnetization anomalies become flatter, their height as well as their hysteresis decrease, and above 16–18 K neither magnetization nor susceptibility anomalies could be observed at our measurement accuracy. The temperature dependences of the fields at which the observed jumps occurred are shown in Fig. 4.

The field dependence of the magnetization of the iron garnet $Ho_{0.41}Y_{2.59}Fe_5O_{12}$ is similar. It also shows

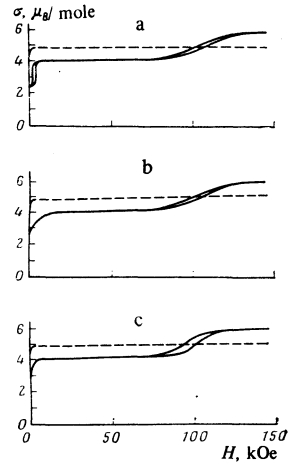

FIG. 2. Magnetization, σ, of a single-crystal iron garnet $Er_{0.12}Y_{2.88}Fe_5O_{12}$ at 4.2 K as a function of the field: a) H along [111], b) H along [110] c) H along [100] (solid lines); dashed line—magnetization of yttrium iron garnet.

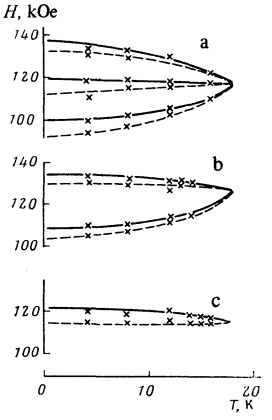

FIG. 4. Temperature dependence of the fields at which magnetization jumps occur in $Ho_{0.25}Y_{2.75}Fe_5O_{12}$ (solid lines—field increasing, dashed lines—field decreasing); a) H along [111], b) H along [110], c) H along [100].

magnetization jumps, but less pronounced than in $Ho_{0.25}Y_{2.75}Fe_5O_{12}$.

DISCUSSION

The data reported here as well as in our earlier paper[5] show that terbium-, holmium- and erbium-doped yttrium iron garnets behave differently in strong fields. As already noted, the observed magnetization anomalies can be interpreted with two models.

A. Formation of a noncollinear magnetic structure of the iron and the RE sublattices caused, due to competition between the negative exchange interaction of the sublattices, which tends to align the magnetic moments of the RE and the iron ions antiparallel to each other, on the one hand, and the external field, which tends to align them parallel to each other,[6,7] on the other.

B. Near-crossing or crossing of the energy levels of the RE ions in the magnetic field, due to the different field dependences of the different energy levels.[4]

The magnetization anomalies observed in yttrium-erbium iron garnets can be explained with the model of the field-induced noncollinear magnetic structure. As shown in[6,7], a noncollinear magnetic structure exists in an isotropic ferrimagnet in the field interval:

$$H_{c1}=H_{\text{eff}}(1-M_R/M_{Fe})\leqslant H\leqslant H_{\text{eff}}(1+M_R/M_{Fe})=H_{c2}, \qquad (1)$$

where H_{eff} is the effective molecular field of the iron sublattice acting on the RE sublattice and M_R and M_{Fe} are the sublattice magnetizations. The transitions into the noncollinear phase in an isotropic ferrimagnet are of second order, and the noncollinear phase should show a gradual increase of the magnetization. Magnetic anisotropy narrows down the interval of existence of the noncollinear phase in the easy magnetization direction and it broadens it in the hard direction by an amount on the order of $\sqrt{H_aH_{\text{eff}}}$, where H_a is the anisotropy field.[12] The anisotropy can also transform the transition into the non-collinear phase in the easy direction into a first-order phase transition. All these features (gradual increase of magnetization in the hard directions [111] and [110] and a sharper increase in the easy direction [100]; magnetization hysteresis in the latter direction) agree with our experimental data. So do the transition fields calculated from Eq. (1): $H_{c1}=82$ kOe and $H_{c2}=125$ kOe for $Er_{0.12}U_{2.88}Fe_5O_{12}$; $H_{c1}=61$ kOe and $H_{c2}=147$ kOe for $Er_{0.52}Y_{2.48}Fe_5O_{12}$ (the values of H_{eff}, M_R and M_{Fe} for the calculations were taken from[13-15]).

We note one more corroborating of the proposition that the magnetization anomalies in yttrium-erbium iron garnets are due to the formation of a noncollinear magnetic structure in the field. Since at $H<H_{c1}$ the magnetization M_R of the RE sublattice is antiparallel to the magnetization M_{Fe} of the iron sublattice and at $H>H_{c2}$ the magnetization M_R is parallel to M_{Fe}, it follows then on going through the noncollinear phase the magnetization of an iron garnet should change from $M_{Fe}-M_R$ to $M_{Fe}+M_R$, and its value at the center of the interval of existence of the noncollinear phase should be M_{Fe}, i.e., be the same as that of the yttrium iron garnet. This is in fact observed in yttrium-erbium iron garnets, as seen in Fig. 2.

The magnetization anomalies of yttrium-holmium iron garnets (Fig. 3) do not follow the usual pattern for a noncollinear magnetic structure, since their magnetization increases jumpwise along all three directions in strong fields. At the same time the character of the observed magnetization anomalies is close to that theoretically predicted for the phenomenon of crossing (or near-crossing) of energy levels: as shown in[4,16], in this case a more or less sharp rise of magnetization should occur. We believe therefore that the complex character of the magnetization anomalies in yttrium-holmium iron garnets in strong fields is due to the superposition of the two effects: formation of the noncollinear magnetic structure and inversion of energy levels in the field.

Two hypotheses can be advanced to explain the magnetization anomalies in yttrium-holmium iron garnets.

A. As the field increases, inversion of the energy levels of the ion Ho^{3+} takes place and further increase of the field brings about formation of a noncollinear magnetic structure.

B. As the field increases, formation of the noncollinear magnetic structure of iron and rare earth sublattices takes place first. The magnetization of the iron sublattice in the noncollinear phase rotates about the direction of the field as the field changes; this is accompanied by a change in orientation of the total effective field acting on the Ho^{3+} ion relative to the crystallographic axes. If the energy levels approach or cross at some directions of the total effective field, then magnetization anomalies arise (similar to the appearance of anomalies of ferromagnetic resonance in iron garnets).

The possibility in principle of the crossing of the Ho^{3+} levels in iron garnets stems from the following qualitative considerations: In a crystal field of symmetry D_2 there are two excited levels of Ho^{3+} with energies close to the ground state ($E_1=5$ cm^{-1}, $E_2=8$ cm^{-1}, Fig. 1a). The splitting pattern changes in the exchange field,[3] and the crossing of levels can occur: one of the originally excited states will become the ground state (this will take place if the rate of energy decrease in the field is greater for the excited state than for the ground state). In iron garnets with small holmium content, the magnetization of the RE sublattice is smaller than that of the iron one, therefore the external field is directed opposite to the iron-sublattice exchange field acting on Ho^{3+} ion and thus reduces the splitting caused by the exchange field. Consequently, if level "crossing" occurred in the exchange field, then level inversion will ensue in a sufficiently strong field—the character of splitting will be the same as that caused by the crystal field alone, and the magnetization jumps will appear.

The different character of the magnetization anomalies in yttrium-holmium iron garnets along different crystallographic directions is explained in the level-crossing model by the sharp dependence of the splitting on the orientation of the magnetization relative to the crystallographic axes.[2,8] Several magnetization jumps

at certain orientations of the magnetic field are caused either when the ground state crosses two consecutive excited states or when the ground state crosses one excited state for different crystallographically nonequivalent positions of RE ions with different splitting of terms due to different orientation of the oxygen dodecahedron relative to crystallographic axes.[2,8]

It is necessary to take it into account that the character of the magnetization anomalies associated with the crossing of levels in ferrimagnets is affected by the "magnetic Jahn–Teller effect,"[17,18] which causes deviation of the magnetization of the iron sublattice from the direction of the field. Therefore, it is apparently possible to consider from a single point of view both effects that account for the anomalies of magnetization in yttrium-holmium iron garnets in strong fields, i.e., crossing of the levels of the Ho^{3+} ion and formation of a noncollinear magnetic sublattice structure, (for details see[17]).

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[1)]The plot of the magnetization along the [111] axis shows a jump in weak fields. Calculations suggest that it is associated with the first-order phase transition of the saturation along the hard axis in cubic ferromagnets with a positive magnetic anistropy constant.

[2)]Our measurements have shown that the width of hysteresis depends somehow, possibly because of relaxation phenomena, on the rate of field variation.

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Establishment of equilibrium between the electron dipole pool, the nuclear spins, and the lattice of a paramagnet

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The processes that take place when equilibrium is established in the electron-nuclear spin system of ruby crystals are investigated at helium temperatures. The existence of a strong coupling between the dipole-dipole pool (DDP) of the Cr^{3+} ions and the Zeeman subsystems of the ^{27}Al ions is confirmed. The parameters of this coupling are determined, as well as the rate of spin-lattice relaxation of the DDP, whose temperature was directly determined in the course of these processes by measuring the longitudinal magnetic susceptibility due to the cross relaxation of the Cr^{3+} ions. It is shown that the coupling between the DDP and the nuclei can be broken by modulation saturation of the DDP with an alternating longitudinal field. The results indicate that an appreciable fraction (up to 30%) of the ^{27}Al nuclei is coupled to the DDP more strongly than with the remaining nuclei, and for some reason makes no contribution to the observed NMR signal. A final interpretation of the results calls for a further development of the theory of nuclear relaxation, with allowance for spin diffusion.

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1. INTRODUCTION AND FORMULATION OF THE PROBLEM

It is known that in solids there can exist a strong coupling between the electron dipole–dipole pool (DDP) of a paramagnetic impurity and the Zeeman subsystem of the nuclear spins of the lattice (Z_n), and this coupling leads to equalization of the temperatures T_d and T_{Zn} of these subsystems. This phenomenon (the so-called "direct thermal contact" between the DDP and Z_n) was theo-

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