Magnetic anisotropy of yttrium-holmium iron garnets

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The magnetization and the first constant of cubic magnetic anisotropy have been measured in $Ho_xY_{3-x}Fe_5O_{12}$ ferrite-garnets ($0 \leqslant x \leqslant 3$). It is shown that the magnetic anisotropy of these garnets at low temperatures is strongly field-dependent; at temperatures below the compensation point the anisotropy constant increases, and at temperatures above the compensation point it decreases, with increase of field. It is concluded that this dependence is caused by change of the magnetization of the rare-earth sublattice in the field. It is found that the anisotropy constant depends nonlinearly on the holmium concentration in the garnet. It is shown that the deviation from linearity is due to the contribution to the anisotropy constant from pseudodipole interaction within the rare-earth sublattice.

Rare-earth iron garnets (REIG), with the general formula $R_3Fe_5O_{12}$ (cubic structure with space group O^h_{10}-Ia3d), are three-sublattice ferrimagnets (see, for example,[1]). There is a strong negative exchange interaction between the Fe^{3+} iron ions located on the tetrahedral (d) and octahedral (a) sites; consequently, the magnetic moments of these sublattices are oriented antiparallel to each other, and in many cases they can be treated as a single resultant iron sublattice. The third sublattice is formed by R^{3+} rare-earth (RE) ions, located on dodecahedral (c) sites. Magnetic order in this sublattice is produced by the relatively weaker negative exchange interaction between the RE and the resultant iron sublattices.

Despite the fact that a considerable number of papers[2-9] have been devoted to investigation of the magnetic anisotropy of REIG, its nature and several of its peculiarities have so far not been explained. This is due to the fact that the magnetic anisotropy of REIG is made up of various components: single-ion anisotropy of the RE and iron ions, anisotropy caused by exchange interaction within the RE and iron sublattices, and also anisotropy caused by interaction between the RE and iron sublattices; and there are no definitive data on the relative contribution of all these mechanisms to the magnetic anisotropy of the REIG. The information obtained so far can be briefly summarized as follows. At room temperature, the magnetic anisotropy of REIG is almost entirely determined by the iron ions. Lowering of the temperature leads to a sharp rise in the anisotropy of most REIG; this is caused by an increase of the contribution of the RE ions to the anisotropy. In most of the papers, the anisotropy caused by the RE ions is interpreted within the framework of the single-ion model. It must be remarked, however, that because of the complicated character of the splitting of the energy levels of the RE ions in a crystalline field of rhombic symmetry, in most cases there is no successful theoretical calculation of the single-ion anisotropy of the RE ions in REIG; therefore comparison of experimental and theoretical data is mainly of qualitative character.

In order to explain the nature of the magnetic anisotropy, a number of papers[2,4,7] compare the anisotropy of pure REIG with the anisotropy of mixed REIG, in which a part of the RE ions have been replaced by nonmagnetic yttrium ions. But in the papers cited above, only yttrium garnets with small admixtures of RE ions were investigated; in most cases, therefore, it was not possible to follow the concentration dependence of the magnetic anisotropy.

For these reasons, we set ourselves the problem of studying the magnetic anisotropy of mixed REIG, of the type $R_xY_{3-x}Fe_5O_{12}$, over a broad range of concentrations ($0 \le x \le 3$). These investigations were undertaken with the aim of explaining the influence of anisotropic interactions within the RE sublattice on the magnetic anisotropy of the REIG. The present paper gives the results of measurements of the magnetic anisotropy of the system $Ho_xY_{3-x}Fe_5O_{12}$. This system was chosen because in it the crystal-lattice parameter, the interionic distances, and the angles between the ions remain practically unchanged on replacement of yttrium by holmium[10]. Hence it may be expected that the constants of the crystalline field that acts on the Ho^{3+} ions will not change with change of x. Furthermore, it can be shown by use of the data of[11] that magnetoelastic interaction gives a small contribution to the magnetic anisotropy of holmium iron garnet; therefore it can be neglected in the interpretation of the experimental data.

SPECIMENS AND METHOD OF MEASUREMENT

Ferrite monocrystals of the system $Ho_xY_{3-x}Fe_5O_{12}$, with x = 0, 0.25, 0.41, 1.05, 1.65, 1.93, 2.53, and 3.00 and weight up to 15 g, were obtained by slow (1.6 deg/h) cooling from 1250 to 950°C of a melt of weight 200–250 g and composition 38 molecular % PbO, 30.7 % $PbFe_2$, 5.5 % B_2O_3, 8.5 % (Y_2O_3 + Ho_2O_3), 17 % Fe_2O_3, 0.3 % $CaCO_3$. High-purity reagents were used. The composition of the crystals was determined on an x-ray microanalyzer JXA-5 (Japan), with accuracy 0.02 in x. The composition was also checked by a magnetic method: comparison of the magnetic properties of the monocrystals with the properties of polycrystals of REIG of nearly the same composition. For measurements of torque and magnetization, specimens were cut in the form of disks, oriented in planes of the type (110) (the plane of natural faceting of the crystal). The diameter of the disks was 3–5 mm, their thickness about 1 mm.

The magnetization along the axis of easy magnetization (the [111] axis) was measured on a vibration magnetometer over the temperature range 4.2–300°K. From these data, the saturation magnetization and paraprocess susceptibility of the REIG were determined.

Measurements of the magnetic anisotropy were made in fields up to 20 kOe, at temperatures 77–300°K, by the torque method. As is well known[12], in REIG, along with the cubic crystallographic anisotropy, there is observed a uniaxial growth-induced anisotropy. Therefore in the analysis of the angular dependence of the torque in the plane (110), the expression used was

$$L = {}^1/_8 K_1 (2\sin 2\theta + \sin 4\theta) + {}^1/_{64} K_2 (\sin 2\theta + 4\sin 4\theta - 3\sin 6\theta) + K_{\text{uniax}} \sin 2(\theta - \varphi). \quad (1)$$

Here the first and second terms describe the cubic crystallographic anisotropy, the third the uniaxial growth-induced anisotropy; θ is the angle between the magnetization and the [100] axis, φ the angle of inclination of the axis of growth-induced anisotropy to this direction. The anisotropy constants K_1, K_2, and K_{uniax} were determined by harmonic analysis of the dependence $L(\theta)$. A correction was made for "lag" of the magnetic moment behind the field direction[13].

Here we shall not consider in detail the uniaxial growth-induced anisotropy of the ferrite-garnets studied. We remark only that it was observed at compositions with $x \leq 1$ and that its value $\sim 10^3$ erg/cm^3. Annealing of the specimens at 1300°C for 12 hours led to a decrease of K_{uniax} by a factor of three or four (except for the pure ytrrium iron garnet) and had no effect on the value of the cubic anisotropy.

EXPERIMENTAL RESULTS

A. Magnetization

Figure 1a shows the temperature dependences of the saturation magnetization of the ferrites $Ho_xY_{3-x}Fe_5O_{12}$. It is seen that introduction of Ho^{3+} ions into yttrium iron garnet leads to the appearance of a magnetic compensation temperature T_c; the value of T_c increases with increase of the holmium content (Fig. 1b).

According to the model presented above for the ferromagnetism of REIG, the magnetization of a iron garnet is $M = \pm(M_{Fe} - M_R)$, where M_{Fe} and M_R are the magnetizations of the resultant iron and of the RE sublattices, and the signs + and − refer respectively to the regions above the magnetic compensation temperature ($M_{Fe} > M_R$) and below it ($M_{Fe} < M_R$). In mixed garnets of the type $R_xY_{3-x}Fe_5O_{12}$, the magnetization of the iron sublattice is independent of composition, whereas the magnetization of the RE sublattice, per mole, is $M_R = xN\mu_R$, where μ_R is the magnetic moment of the RE ion at the given temperature. Consequently

$$M = \pm(M_{Fe} - xN\mu_R). \quad (2)$$

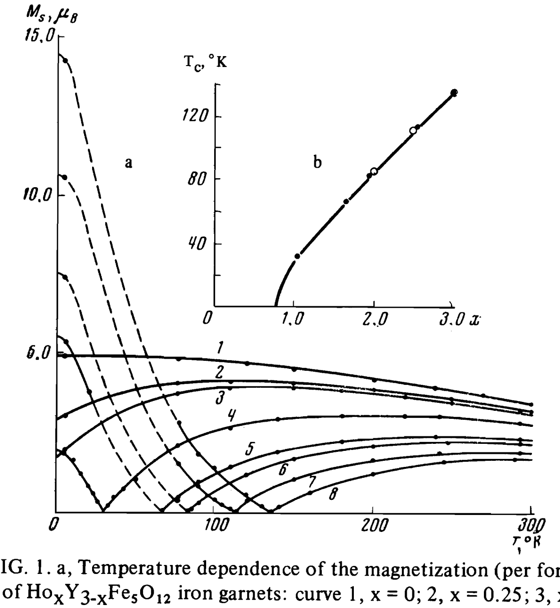

FIG. 1. a, Temperature dependence of the magnetization (per formula unit) of $Ho_xY_{3-x}Fe_5O_{12}$ iron garnets: curve 1, x = 0; 2, x = 0.25; 3, x = 0.41; 4, x = 1.05; 5, x = 1.65; 6, x = 1.93; 7, x = 2.53; 8, x = 3. b, Composition dependence of the compensation temperature of the iron garnets: ●, monocrystals; ○, polycrystals; solid curve, theoretical dependence.

If, as has already been stated, the magnetic order of the RE sublattice is caused by exchange interaction between the RE and iron ions and the exchange interaction within the RE sublattice is negligible in comparison with this intersublattice interaction, then at a given temperature the magnetic moment of the RE ion is independent of composition, and consequently the magnetization of the REIG should vary linearly with the concentration x. Our experimental data fit this model well: over the temperature range investigated, there is a linear dependence of the magnetization of $Ho_xY_{3-x}Fe_5O_{12}$ ferrites on composition. The dependence of the compensation point T_c on x also is described well by formula (2), if in it one sets M = 0 (Fig. 1b).

As has already been mentioned, the effective exchange field with which the iron sublattice acts on the RE sublattice is small (in holmium iron garnet the molecular field ~100 kOe[14]); therefore an external magnetic field of order 10 kOe significantly affects the magnetic order in this sublattice. Below the compensation temperature, when $M_R > M_{Fe}$ and the external field is directed parallel to the magnetization of the RE sublattice, it increases the magnetization M_R; but above T_c, when $M_{Fe} > M_R$ and the external field is oriented antiparallel to the magnetization of the RE sublattice, it decreases M_R. This is clearly evident from Fig. 2, which shows, on the basis of our experimental data, the field dependences of the magnetization of the RE sublattice of $Ho_3Fe_5O_{12}$ (T_c = 136°K) at various temperatures.

B. Magnetic Anisotropy

Figure 3 shows the temperature dependences of the first magnetic-anisotropy constant K_1 of $Ho_xY_{3-x}Fe_5O_{12}$ iron garnets.1) At room temperature the anisotropy is small and independent of the holmium content. This indicates that at this temperature the anisotropy is basically determined by the iron sublattices, and that the RE ions make an insignificantly small contribution to the anisotropy. With lowering of the temperature below about 200°K, the anisotropy increases, more sharply in specimens containing a larger amount of holmium. In this temperature range, the contribution of the holmium ions to the anisotropy is important.

Note that the values of the anisotropy constant plotted in Fig. 3 have been extrapolated to zero field. This is because, as our measurements showed, the anisotropy constants of holmium iron garnets depend

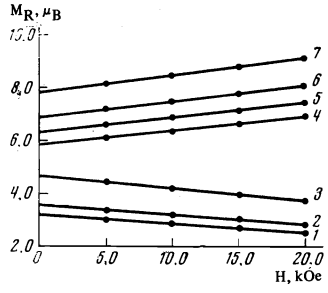

FIG. 2. Field dependence of the magnetization of the rare-earth sublattice (per formula unit) of $Ho_3Fe_5O_{12}$ ferrite-garnet at various temperatures: 1, 200°K; 2, 180°K; 3, 140°K; 4, 110°K; 5, 100°K; 6, 90°K; 7, 77°K.

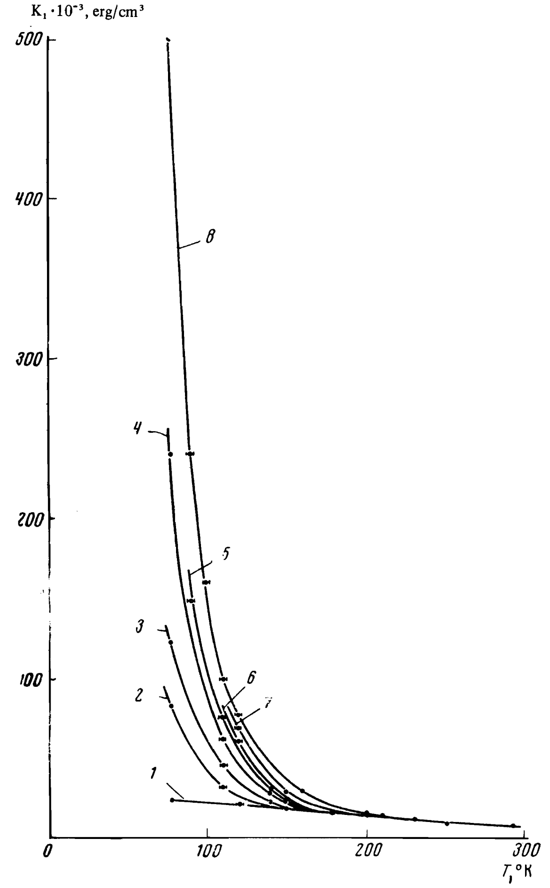

FIG. 3. Temperature dependence of the first cubic anisotropy constant of $Ho_xY_{3-x}Fe_5O_{12}$ iron garnets: curve 1, x = 0; 2, x = 0.25; 3, x = 0.41; 4, x = 1.05; 5, x = 1.65; 6, x = 1.93; 7, x = 2.53; 8, x = 3.

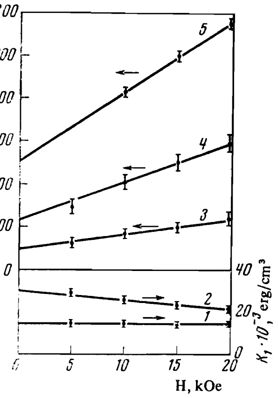

FIG. 4. Field dependence of the anisotropy constant K_1 of $Ho_3Fe_5O_{12}$ ferrite-garnet: 1, at 200°K; 2, 120°K; 3, 110°K; 4, 90°K; 5, 77°K.

strongly on field. Figure 4 shows the field dependences of the anisotropy constant of pure holmium FG at various temperatures. It is seen that for $T < T_C = 136°K$ the anisotropy constant increases, and that for $T > T_C$ it decreases, with increase of field. Similar dependences $K_1(H)$ are observed also for mixed yttrium-holmium garnets.

The change of the magnetic anisotropy of REIG with field is caused by the change in the field of the magnetization of the RE sublattice. As follows from the general theory of magnetic anisotropy[15], the anisotropy constant is a function of the magnetization; if, therefore, the magnetization changes, this leads to a change of the anisotropy constant. In an iron garnet the external field strongly affects the magnetization of the RE sublattice (see Fig. 2 above), decreasing it for $T > T_C$ and increasing it for $T < T_C$, and this leads to decrease or increase, respectively, of the anisotropy constant in the field.

As has already been mentioned, it is possible to draw a conclusion in regard to the contribution of the various mechanisms to the anisotropy of REIG by following the concentration dependence of the anisotropy constant of mixed REIG. In fact, if the anisotropy is single-ion, then it is made up additively from interactions of individual RE ions with the crystalline field of the lattice, and therefore it should depend linearly on the concentration x of RE ions. A linear $K_1(x)$ dependence should also be observed if the magnetic anisotropy is caused by interaction of the iron and RE ions. But if the anisotropy is produced by interaction within the RE sublattice, then it is proportional to the number of pair interactions of RE ions with each other and should be a quadratic function of the concentration of these ions in the iron garnet. We remark that since the value of the magnetic anisotropy depends sharply on the magnetization, the concentration dependence of the anisotropy constant of mixed REIG must be followed at a single value of the magnetic moment of the RE ion. It is therefore not permissible, as was done for example in[2], to intercompare anisotropy constants of mixed REIG for $H \neq 0$, since the field affects differently the anisotropy constant of iron garnets for which the measurement temperature is higher than the magnetic compensation temperature and of those for which the measurement temperature is lower than the magnetic compensation temperature. We have compared with each other the anisotropy constants of $Ho_xY_{3-x}Fe_5O_{12}$ extrapolated to zero field (Fig. 5). It is evident from Fig. 5 that at low temperatures in the region of large x, the $K_1(x)$ dependence deviates from linearity.

The question arises, however, whether this deviation from linearity may not have been caused by the errors of extrapolation of the $K_1(H)$ dependence to zero field. To elucidate this matter, we constructed, on the basis of the experimental data, the dependence of the contribution of the RE sublattice to the REIG anisotropy, per RE ion, on the magnetic moment of the RE ion at various fields and temperatures (Fig. 6). If the anisotropy of the iron garnets depends linearly on the RE ion content, then the contribution of the RE sublattice to the anisotropy, per RE ion, will not depend on x; but if there is a deviation from linearity, then this contribution will be different for compositions with different x. As is evident from Fig. 6, the latter situation is ob-

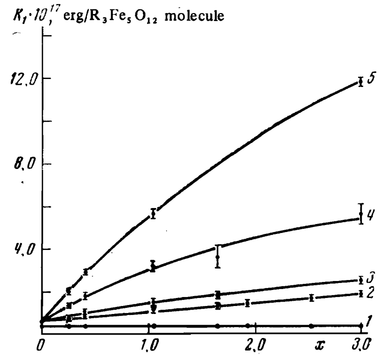

FIG. 5. Composition dependence of the anisotropy constant K, of $Ho_xY_{3-x}Fe_5O_{12}$ ferrite-garnets, at temperatures: 1, 200°K; 2, 120°K; 3, 110°K; 4, 90°K; 5, 77°K.

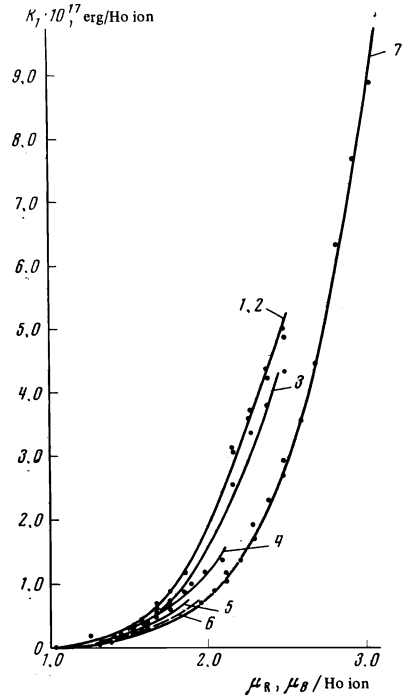

FIG. 6. Dependence of the anisotropy constant of $Ho_xY_{3-x}Fe_5O_{12}$ garnets, produced by the rare-earth sublattice (per rare-earth ion), on the magnetic moment of the rare-earth ion: 1, x = 0.25; 2, x = 0.41; 3, x = 1.05; 4, x = 1.65; 5, x = 1.93; 6, x = 2.53; 7, x = 3.

served in the system $Ho_xY_{3-x}Fe_5O_{12}$: the contribution of the RE sublattice to the anisotropy constant, per RE ion, is, for given μ_R, less for compositions with large x.

Thus it follows from our measurements that the magnetic anisotropy of the system $Ho_xY_{3-x}Fe_5O_{12}$ depends nonlinearly on the holmium concentration. This indicates, for the iron garnets investigated, an appreciable contribution to the anisotropy from anisotropic interaction between holmium ions.

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[1] Our measurements showed that for all the iron garnets investigated, K_2 is zero within the limits of error (it does not exceed 0.05 K_1).

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