Magnetic anisotropy and magnetostriction of yttrium-terbium iron garnets

K. P. Belov, A. K. Gapeev, R. Z. Levitin, A. S. Markosyan, and Yu. F. Popov

Moscow State University
(Submitted June 18, 1974)
Zh. Eksp. Teor. Fiz. **68**, 241–248 (January 1975)

The magnetic anisotropy and magnetostriction of monocrystals of the iron garnets $Tb_xY_{3-x}Fe_5O_{12}$ have been measured over the temperature range 78–300 °K. It was discovered that at nitrogen temperatures the first anisotropy constant K_1 varies nonmonotonically with the terbium-ion concentration; it is positive for specimens with small x and negative for specimens with large x. Comparison of the data on magnetic anisotropy with the data on magnetostriction has shown that anomalous $K_1(x)$ dependence is due to the contribution of magnetoelastic interaction to the anisotropy. At 78 °K, for compounds with large x, the contribution of magnetoelastic interaction to the first magnetic-anisotropy constant exceeds the measured value of K_1 and decreases sharply with decrease of the terbium-ion concentration. This leads to the result that the anisotropy constant of the undeformed lattice, $K_{10} = K_1 - \Delta K_1^{me}$, is positive for all yttrium-terbium iron garnets. It was found that the first magnetic-anisotropy constant K_{10} of the undeformed lattice and the magnetostriction constant λ_{100} vary nonlinearly with terbium-ion concentration; this is evidence of an appreciable contribution to these constants from pseudodipole interaction between the terbium ions.

1. In[1] we studied the magnetic anisotropy of yttrium-holmium ferrites with garnet structure. It was discovered that the anisotropy of these garnets receives an important contribution from anisotropic exchange interaction between the holmium ions. It was also shown that magnetoelastic interaction does not affect the anisotropy of yttrium-holmium iron garnets, since the magnetostriction of these ferrimagnets is small.

A different situation should occur for mixed yttrium-terbium iron garnets. At nitrogen temperature, the magnetostriction of terbium iron garnet is an order of magnitude larger than for other rare-earth iron garnets[2]; and as was shown in[3,4], in this temperature range the contribution made by magnetoelastic interaction to the first constant of cubic magnetic anisotropy of terbium iron garnet is comparable with the measured value of the constant. The large value of the anisotropic magnetoelastic interaction should lead to a number of peculiarities of the magnetic anisotropy in mixed yttrium-terbium iron garnets.

Presented below are the results of measurements of the magnetic anisotropy and magnetostriction of yttrium-terbium iron garnets. These measurements were undertaken in order to determine the influence of magnetoelastic interaction on the magnetic anisotropy energy of yttrium-terbium iron garnets, and also to explain the nature of the magnetic anisotropy and magnetostriction of these ferrimagnets.

2. Single-crystal iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$ ($0 \le x \le 3$) were grown and analyzed by the methods that we have described in[1]. The methods of measuring the magnetization and the magnetic anisotropy are also described there.

The magnetostriction constants λ_{111} and λ_{100} were calculated from the general formula for the magnetostriction of a cubic crystal,

$$\lambda = {}^3/_2\lambda_{100}[\alpha_1^2\beta_1^2+\alpha_2^2\beta_2^2+\alpha_3^2\beta_3^2-{}^1/_3] + 3\lambda_{111}[\alpha_1\alpha_2\beta_1\beta_2+\alpha_1\alpha_3\beta_1\beta_3+\alpha_2\alpha_3\beta_2\beta_3] \quad (1)$$

(α_i are the direction cosines of the measurement direction, β_i the direction cosines of the vector magnetization), on the basis of measurements of the longitudinal and transverse magnetostriction along the directions [111] and [100]. The measurements were made in pulsed magnetic fields by means of a remote piezoelectric pickup[5]. Check measurements were made of the angular dependence of the transverse magnetostriction in the crystallographic planes (111), (100), and (110); they showed that in the iron garnets studied, the magnetostriction is described within the limits of error (~10%) by formula (1), and consequently the higher-order magnetostriction constants (in terms containing the direction cosines of the magnetization to the fourth degree) are zero.

3. Figure 1 shows the temperature dependences of the first (K_1) and second (K_2) cubic-anisotropy constants of yttrium-terbium iron garnets[1)]. Analysis of the data presented in Fig. 1 shows that certain peculiarities of the magnetic anisotropy of yttrium-terbium and of yttrium-holmium iron garnets coincide.

First, the anisotropy of yttrium-terbium iron garnets at room temperature depends little on the terbium concentration. This shows that in this temperature range the principal contribution to the magnetic anisotropy comes from the iron ions At low temperatures there is a strong dependence of the anisotropy constants on the terbium concentration. Therefore in this temperature range the principal contribution to the anisotropy comes from the terbium ions. We remark that for pure terbium iron garnet, our data agree with Pearson's results[6].

Second, it follows from our measurements that the anisotropy constants of yttrium-terbium iron garnets depend strongly on field. For this reason, Fig. 1 shows values of the anisotropy constants extrapolated to zero field. This phenomenon, which we discovered earlier in yttrium-holmium iron garnets, is due to the influence of the field on the magnetization of the rare-earth sublattice (for more details see[1]).

Noticeable also are the following differences of the magnetic anisotropy of yttrium-terbium iron garnets from the anisotropy of yttrium-holmium iron garnets.

First, the second anisotropy constant K_2 of these ferrimagnets at low temperatures is an order of mag-

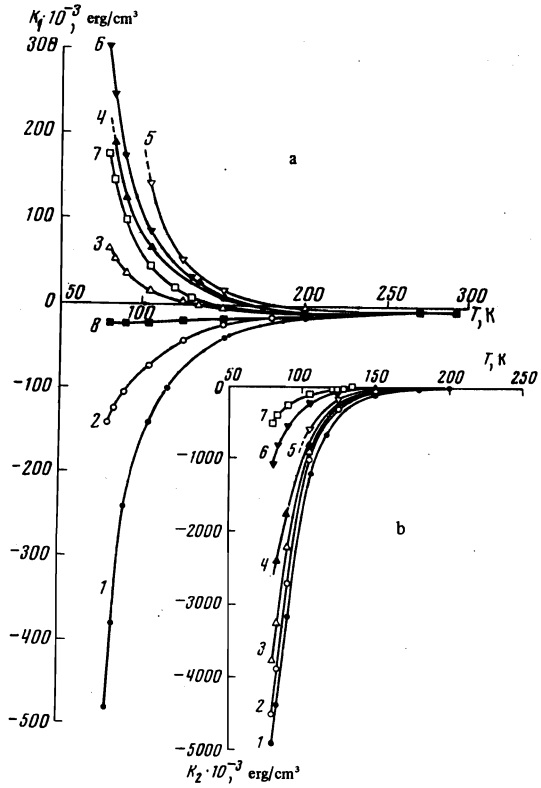

FIG. 1. Temperature dependence of the first cubic-anisotropy constant k_1 (a) and of the second constant K_2 (b) for iron garnets $Tb_xY_{3-x}Fe_5O_{12}$: 1, x = 3.0; 2, x = 2.54; 3, x = 2.12; 4, x = 1.65; 5, x = 1.17; 6, x = 0.50; 7, x = 0.26; 8, x = 0.00.

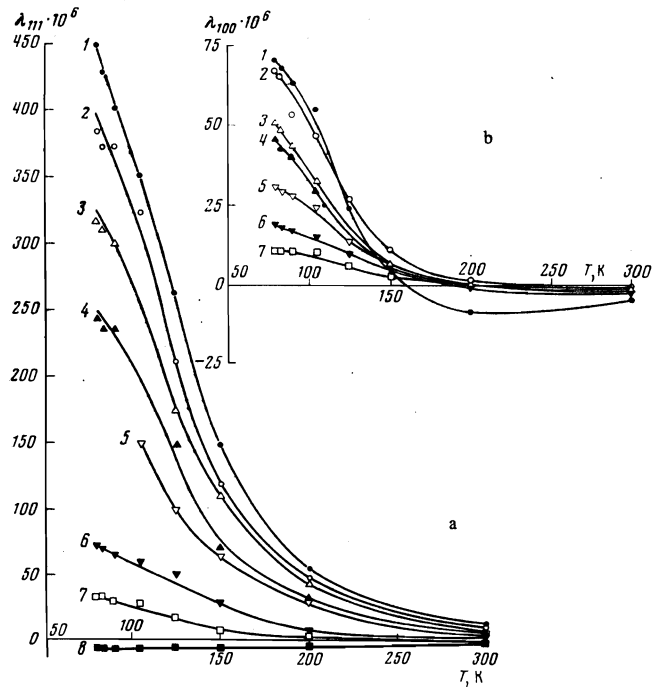

FIG. 2. Temperature dependence of the magnetostriction constants λ_{111} (a) and λ_{100} (b) for iron garnets $Tb_xY_{3-x}Fe_5O_{12}$: 1, x = 3.0; 2, x = 2.54; 3, x = 2.12; 4, x = 1.65; 5, x = 1.17; 6, x = 0.50; 7, x = 0.26; 8, x = 0.00.

nitude larger than the first constant K_1 (the second anisotropy constant of yttrium-holmium iron garnets is zero[1]).

Second, and this is of the greatest interest from our point of view, while the first magnetic-anisotropy constant of ferrites with a large terbium concentration ($3 \geq x \geq 2.54$) is negative over the whole temperature interval studied (that is, it behaves like the K_1 of yttrium-holmium iron garnets), the first constant of mixed yttrium-terbium garnets with $x < 2.54$ is positive at low temperatures and changes sign with rise of temperature.

Thus, in contrast to yttrium-holmium iron garnets, the first magnetic-anisotropy constant of yttrium-terbium iron garnets at low temperatures depends nonmonotonically on the terbium concentration and changes sign on replacement of terbium by yttrium[2)]. Since in this temperature range the anisotropy, as has already been mentioned, is determined principally by the rare-earth sublattice, it follows from our data that the contribution of the terbium ions to the first anisotropy constant of yttrium-terbium iron garnets changes sign on replacement of terbium by yttrium.

The change of sign of the first magnetic-anisotropy constant of mixed yttrium-terbium iron garnets with $x \leq 2.12$, upon increase of temperature, can be understood if one takes account of the fact that, as was pointed out above, the principal contribution to the anisotropy at high temperatures comes from the iron ions. This contribution is negative (see the data for yttrium iron garnet in Fig. 1), and K_1 in mixed ferrites vanishes at the temperature at which the anisotropy components from the terbium and iron ions compensate each other.

A possible reason for the unusual concentration dependence of the first anisotropy constant of yttrium-terbium iron garnets is the effect of magnetoelastic interaction. In order to clear up this question, we made measurements of the magnetostriction of these ferrites.

4. Figure 2 shows the temperature dependences of the magnetostriction constants λ_{111} and λ_{100} of yttrium-terbium iron garnets. Our measurements showed (in agreement with data on polycrystals[7]) that the magnetostriction of monocrystals of these iron garnets depends strongly on field; therefore Fig. 2 gives values of the magnetostriction constants extrapolated to zero field. The explanation of this phenomenon is completely analogous to the explanation of the field dependence of the anisotropy constants (see above).

As follows from Fig. 2, at low temperatures the magnetostriction of ferrites with a large terbium concentration is large and decreases on replacement of terbium by yttrium. Noticeable is the large anisotropy of the magnetostriction of yttrium-terbium iron garnets: λ_{111} is almost an order of magnitude larger than λ_{100}[3)].

With rise of temperature, λ_{111} and λ_{100} decrease; but even at room temperature, there is a dependence of the magnetostriction constants on terbium content. This shows that at this temperature the terbium ions have an appreciable influence on the magnetostriction of yttrium-terbium iron garnets.

5. The measured magnetic-anisotropy energy consists of two parts: the magnetic anisotropy of the undeformed crystalline lattice, and the additional anisotropy caused by magnetoelastic interaction.

As follows from Kittel's calculations[8], the magnetoelastic contribution to the first constant of cubic anisotropy is related to the magnetostriction constants λ_{111} and λ_{100} and to the elastic constants c_{11}, c_{12}, and c_{44} and

can be described in the form

$$\Delta K_1^{me}=9/4[(c_{11}-c_{12})\lambda_{100}^2-2c_{44}\lambda_{111}^2]. \quad (2)$$

We have carried out a calculation of the magnetoelastic contribution to the first magnetic-anisotropy constant of yttrium-terbium iron garnets, using our data for the magnetostriction constants and the elastic constants of yttrium iron garnet from the paper of Clark and Strakna[9][4)].

It was found that this contribution was negative for all the yttrium-terbium iron garnets studied by us; at low temperatures, for ferrites with a large terbium concentration, it exceeds in absolute value our experimental value of the first magnetic-anisotropy constant K_1. With decrease of the terbium concentration, ΔK_1^{me} decreases; and for specimens with small x, it becomes less than the value of the total anisotropy constant K_1. The sharp variation of the magnetoelastic contribution to the anisotropy is due to the fact that ΔK_1^{me} depends quadratically on the magnetostriction constants λ_{111} and λ_{100}, which decrease with decrease of the terbium content (Fig. 2).

Figure 3 shows the temperature dependences of K_{10}, the first magnetic-anisotropy constant of an undeformed crystal (without the magnetoelastic contribution to the anisotropy) of yttrium-terbium iron garnets. We remark that since K_{10} is the difference of two large quantities ($K_{10} = K_1 - \Delta K_1^{me}$), the errors of its determination are very large, especially for compounds with a large terbium content. Nevertheless it is evident from these data that at low temperatures the constant K_{10} is positive for all the ferrites investigated (the change of sign of the constant with rise of temperature is due, as has already been mentioned, to the influence of the iron ions).

Thus, it has been shown experimentally that the anomalous concentration dependence of the first constant of cubic anisotropy of yttrium-terbium iron garnets is caused by the presence in these ferrimagnets of a large negative magnetostrictive contribution to the magnetic-anisotropy energy.

We remark that in the general case, the magnetoelastic interaction should also have an effect on the second magnetic-anisotropy constant. But as was shown in[11], the magnetoelastic contribution to K_2 is determined by higher-order magnetostriction constants (in terms of the fourth degree in the direction cosines of the magnetization), and in the ferrites investigated these constants are zero.

6. As we have already pointed out[1], the anisotropy produced in iron garnets by the rare-earth (RE) ions may receive contributions from the following mechanisms: single-ion anisotropy, which describes the anisotropic interaction of the orbital moment of a Re ion with the crystalline field of the lattice; anisotropic exchange interaction between RE and iron ions; and anisotropic exchange interaction of RE ions with each other. In mixed iron garnets, the single-ion anisotropy depends linearly on the concentration of RE ions (if the parameters of the crystalline field do not change on replacement of RE ions by yttrium); the contribution to the anisotropy due to rare earth–iron exchange interaction also changes linearly with concentration; but the anisotropy due to rare earth–rare earth exchange interaction depends quadratically on the RE ion content.

Figure 4 shows the dependences of K_{10} (after subtraction of the magnetoelastic component) and K_2, for yttrium-terbium iron garnets, on the terbium concentration, at various temperatures. As is seen from the figure, at low temperatures K_{10} varies extremely nonlinearly with the terbium concentration. This indicates that anisotropic exchange interaction between the terbium ions makes a substantial contribution to the anisotropy. We remark that in the yttrium-terbium iron garnet system the crystal-lattice constant decreases somewhat on replacement of terbium by yttrium (the lattice constants of yttrium and terbium iron garnets differ by 0.5%[12]), and this can lead to a change of the crystalline-field parameters and consequently to a nonlinear dependence of the single-ion anisotropy on the concentration.

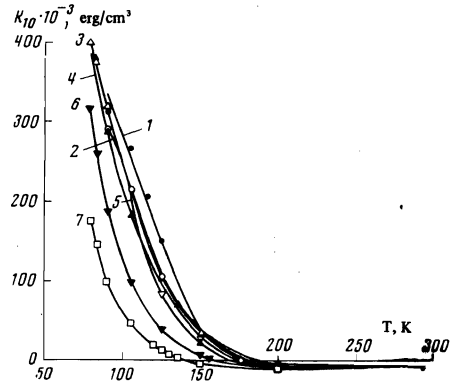

FIG. 3. Temperature dependence of $K_{10} = K_1 - \Delta K_1^{me}$ for iron-garnets $Tb_xY_{3-x}Fe_5O_{12}$: 1, x = 3.0; 2, x = 2.54; 3, x = 2.12; 4, x = 1.65; 5, x = 1.17; 6, x = 0.50; 7, x = 0.26.

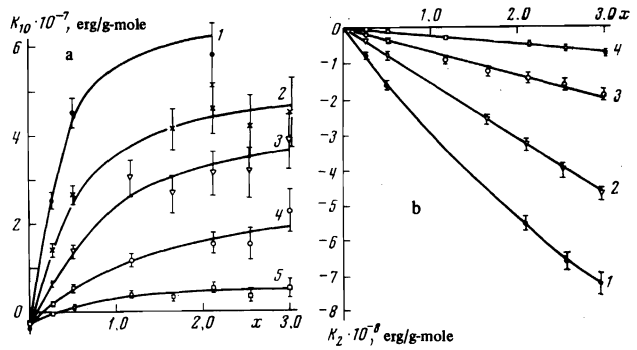

FIG. 4. Dependence on terbium concentration of the cubic-anisotropy constants $K_{10} = K_1 - \Delta K_1^{me}$ (a) and K_2 (b) for iron garnets $Tb_xY_{3-x}Fe_5O_{12}$: 1, 79.5 K; 2, 90 K; 3, 105 K; 4, 125 K; 5, 150 K.

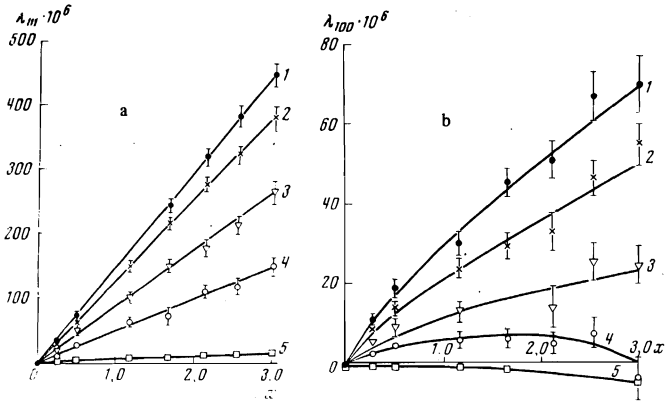

FIG. 5. Dependence on terbium concentration of the magnetostriction constants λ_{111} (a) and λ_{100} (b) for iron garnets $Tb_xY_{3-x}Fe_5O_{12}$: 1, 79.5 K; 2, 105 K; 3, 125 K; 4, 150 K; 5, 300 K.

At large x, there occurs a slight departure of the $K_2(x)$ dependence from a straight line (Fig. 4). This departure may be due both to the influence of anisotropy produced by exchange interaction and to change of the crystalline-field parameters on change of x. It will become possible to answer this question more exactly when information has been obtained about the influence of small changes of the interatomic distances in the garnet structure upon the crystalline-field parameters.

What was said above about the concentration dependence of the various contributions to the anisotropy can be carried over completely to the magnetostriction.

Figure 5 shows the dependences $\lambda_{111}(x)$ and $\lambda_{100}(x)$ of yttrium-terbium iron garnets at various temperatures. It is seen that within the limits of accuracy of the measurements, there is a linear dependence of λ_{111} on x. This indicates that terbium-terbium interaction makes no contribution to this constant. In contrast to this, λ_{100} depends nonlinearly on the terbium concentration; this is evidence of a large contribution to λ_{100} from terbium-terbium interaction.

In closing, we thank T. M. Perekalina for help in the computer processing of the torque curves, and B. V. Mill', under whose supervision the single crystals that we investigated were grown.

[1)] As in other garnets, in the specimens of yttrium-terbium garnets studied by us there is a growth-induced uniaxial anisotropy. It is not discussed here.

[2)] A positive magnetic-anisotropy constant K_1 was observed by Pearson [6] at helium temperatures in a specimen of yttrium garnet with small (~0.19%) admixtures of terbium.

[3)] We note that because $\lambda_{100} \ll \lambda_{111}$, the error of determination of the magnetostriction constant λ_{100} from the experimental data increases, since it is necessary to distinguish a small quantity (λ_{100}) against the background of a large (λ_{100}).

[4)] It was shown in [10] that the elastic constants of terbium and yttrium iron garnets are close in value.

[1] A. K. Gapeev, R. Z. Levitin, A. S. Markosyan, B. V. Mill', and T. M. Perekalina, Zh. Eksp. Teor. Fiz. 67, 232 (1974) [Sov. Phys.-JETP 40, 117 (1975)].
[2] V. I. Sokolov and Than Duc Hien, Zh. Eksp. Teor. Fiz. 52, 1485 (1967) [Sov. Phys.-JETP 25, 986 (1967)].
[3] S. Iida, J. Phys. Soc. Jap. 22, 1201 (1967).
[4] K. P. Belov and V. I. Sokolov, in the collection Magnitnye i kristallokhimicheskie issledovaniya ferritov (Magnetic and Crystal-Chemical Investigations of Ferrites), MGU, 1971, p. 42.
[5] B. K. Ponomarev and R. Z. Levitin, Prib. Tekh. Eksp. No. 3, 188 (1966) [Instrum. Exp. Tech. No. 3, 708 (1966)].
[6] R. Pearson, J. Appl. Phys. 33, 1236 (1962).
[7] K. P. Belov, R. Z. Levitin, and Yu. F. Popov, Zh. Eksp. Teor. Fiz. 59, 1985 (1970) [Sov. Phys.-JETP 32, 1076 (1971)].
[8] C. Kittel, Rev. Mod. Phys. 21, 541 (1949) [Russ. transl. in: Fizika ferromagnitnykh oblastei (Physics of Ferromagnetic Domains), IIL, 1951, p. 56).
[9] A. E. Clark and R. E. Strakna, J. Appl. Phys. 32, 1172 (1961).
[10] R. C. LeCraw and R. L. Comstock, in Physical Acoustics, ed. W. P. Mason, Vol. III, Part B, p. 127; Academic Press, 1965 (Russ. transl., Mir, 1968, p. 156).
[11] R. Birss, Symmetry and Magnetism, North-Holland, Amsterdam, 1964.
[12] G. P. Espinosa, J. Chem. Phys. 37, 2344 (1962).

Translated by W. F. Brown, Jr.
30