Magnetic anisotropy and magnetostriction of neodymium-substituted yttrium iron garnets

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The anisotropy and magnetostriction constants of five neodymium-substituted yttrium iron garnet single crystals are measured by the ferromagnetic resonance technique at a frequency of 8.4 GHz and in the 4.2–560 °K temperature range. At temperature between 4.2 and 25 °K an anomalous resonance field peak is observed in the [110] direction. The experimental results are compared with the single- and double-ion theories of anisotropy and magnetostriction.

The present paper describes an investigation of the influence of the Nd^{3+} ions on the magnetic anisotropy and magnetostriction of yttrium iron garnet (YIG). The magnetic anisotropy of the rare-earth-substituted YIG can be explained by allowing for the spin-orbit interaction. The presence of small amounts of rare-earth ions, which have a free orbital moment L, increases strongly the anisotropy and magnetostriction of YIG. The free Nd^{3+} ions are in the $^4I_{9/2}$ state and have an orbital moment L = 6. Therefore, even a small admixture of these ions may raise considerably the anisotropy and magnetostriction.

We studied the magnetic anisotropy and magnetostriction of five single crystals of the $Y_{3-x}Nd_xFe_5O_{12}$ system (x = 0.0, 0.006, 0.015, 0.03, 0.09). These crystals were grown by crystallization from a molten solution. Their composition was deduced with the aid of an x-ray microanalyzer and a polycrystalline standard sample. The measurements in the 4.2–560°K range were carried out by the ferromagnetic resonance method using polished spheres of ≈0.05 cm in diameter. The samples were oriented by x-ray diffraction to within ±1°.

Figure 1 shows the dependence of the resonance field on the angle between the direction of magnetization and the [100] axis in the (110) plane. We can see that the resonance field has an anomalous peak along the [110] direction. The amplitude of this peak is a function of the temperature and its disappears at 25°K. Dillon and Nielsen[1] were the first to observe a similar phenomenon for other rare-earth ions in YIG. Such peaks were attributed by Kittel[2] to the crossing or approach of energy levels of the rare-earth ion under the influence of the crystal and exchange fields.

Figures 2 and 3 shows the temperature dependences of the ratios K_1/M_S and K_2/M_S for all the investigated samples; here, K_1 and K_2 are the first and second anisotropy constants in the expansion of the free energy of the magnetic anisotropy in terms of the direction cosines, and M_S is the saturation magnetization. We can see that small amounts of Nd^{3+} increase considerably the anisotropy of YIG. For all the compositions the values of K_1 and K_2 are negative and $|K_2|$ is approximately twice as large as $|K_1|$.

The anisotropy constants were calculated from the formulas[3]

$$f=2.8\left(H_{100}-\frac{2K_1}{M_s}\right),\quad f=2.8\left(H_{111}+\frac{4}{3}\frac{K_1}{M_s}+\frac{4}{9}\frac{K_2}{M_s}\right),$$
$$f=2.8\left\{\left(H_{110}-\frac{K_1}{M_s}-\frac{1}{2}\frac{K_2}{M_s}\right)\left(H_{110}+2\frac{K_1}{M_s}\right)\right\}^{1/2}, \quad (1)$$

where H_{100}, H_{111}, and H_{110} are the resonance values of the field along the [100], [111], and [110], respectively; f is the ferromagnetic resonance frequency. The resonance fields H_{100} and H_{111} were determined at two frequencies (f_1 = 8.36 GHz and f_2 = 9.62 GHz) for the samples with x = 0.006 and x = 0.015; this was done in the temperature range 4.2–25°K, which corresponded to the range of existence of the anomalous peak along the [110] direction. These measurements enabled us to calculate K_1 and K_2 from Eq. (1) without recourse to the value of H_{110}.

In theoretical investigations it is usual to expand the free energy in terms of spherical harmonics. This gives rise to an anisotropy constant $\tilde{K}_4$, which is related to K_1 and K_2 by:

$$\tilde{K}_4(T)\approx K_1(T)+{}^1/_{11}K_2(T).$$

The temperature dependence $\tilde{K}_4(T)$ was calculated using the one-ion model of the magnetic anisotropy:[4]

$$\tilde{K}_4(T)=\tilde{K}_4(0)\hat{I}_{9/2}[\mathcal{L}^{-1}(m_n)], \quad (2)$$

where $\hat{I}_{9/2}$ is a hyperbolic Bessel function whose argument is the reciprocal of the Langevin function of the

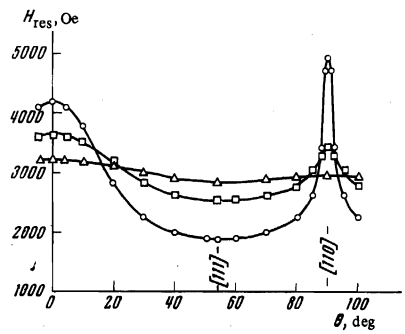

FIG. 1. Dependences of the resonance field of $Y_{3-x}Nd_xFe_5O_{12}$ single crystals on the angle between the direction of the saturation magnetization and the [100] axis in the (110) plane. Frequency f = 8.360 GHz; T = 4.2°K; Δ–x = 0.0; □–x = 0.006; ○–x = 0.015.

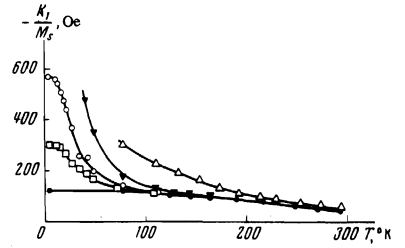

FIG. 2. Temperature dependences of K_1/M_S for $Y_{3-x}Nd_xFe_5O_{12}$ single crystals: ●–x = 0.00; □–x = 0.006; ○–x = 0.015; ▼–x = 0.03; Δ–x = 0.09.

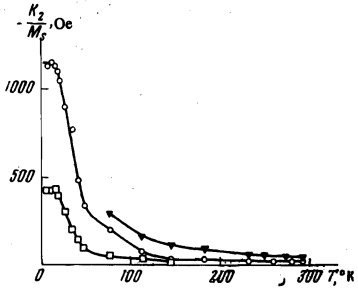

FIG. 3. Temperature dependences of K_2/M_S for $Y_{3-x}Nd_xFe_5O_{12}$ single crystals: □—x = 0.006; ○—x = 0.015; ▼ = 0.03.

magnetization of a given sublattice. Following Henderson and White,[5] who studied the magnetic properties of YIG, we assumed that the magnetization of the Nd sublattice varies in accordance with the law

$$m_n \sim \text{th}\,(\Delta E / 2kT), \tag{3}$$

where ΔE is the average exchange splitting of the principal Nd^{3+} doublet in the yttrium garnet lattice. Employing the results of Clarke,[6] we assumed that $\Delta \approx 55\ \text{cm}^{-1}$.

The theoretical curves calculated from Eq. (2) for one Nd^{3+} ion are plotted in Fig. 4. The dashed curve in the same figure is the theoretical dependence $\tilde{K}_4(T)$ derived using the two-ion interaction model:[7]

$$\tilde{K}_4(T) \approx \tilde{K}_4(0)\, m_{Re}^3 m_{Fe}^3,$$

where m_{Re} and m_{Fe} are the reduced magnetizations of the rare-earth and iron sublattices, respectively. The points in Fig. 4 represent the experimental values of $\tilde{K}_4(T)/\tilde{K}_4(0)$.

The values of the anisotropy constants at 0°K can be calculated if we use the standard equation for the angular dependence of the exchange splitting:[8]

$$\Delta E(l, m, n) = (\Delta E_x^2 l^2 + \Delta E_y^2 m^2 + \Delta E_z^2 n^2)^{1/2}, \tag{4}$$

where l, m, and n are the direction cosines of the magnetization measured from the local orthorhombic axes associated with an Nd^{3+} ion; ΔE_X, ΔE_Y, and ΔE_Z are the splittings along the x, y, and z axes deduced from the temperature dependence of the width of the ferromagnetic resonance curve given by Clarke.[6] In the garnet structure the Nd^{3+} ions may generally occupy six inequivalent sites. If the magnetization lies in the (110) plane, there are only four inequivalent sites. Going over from one type of site to another by rotating the local system of coordinates, we find that the general formula (4) yields the splitting for each of the inequivalent sites:

$$\begin{aligned}
\Delta E_1 &= (\Delta E_x^2 \cos^2\theta + \Delta E_y^2 \sin^2\theta)^{1/2},\\
\Delta E_2 &= (\Delta E_x^2 \cos^2\theta + \Delta E_z^2 \sin^2\theta)^{1/2},\\
\Delta E_3 &= [{}^1/_2 \Delta E_x^2 \sin^2\theta + \Delta E_y^2({}^1/_2\cos^2\theta\\
&\quad + {}^1/_4 \sin^2\theta - 2^{-1/2}\sin\theta\cos\theta)\\
&\quad + \Delta E_z^2({}^1/_2\cos^2\theta + {}^1/_2\sin^2\theta + 2^{-1/2}\sin\theta\cos\theta)]^{1/2},\\
\Delta E_4 &= [{}^1/_2 \Delta E_x^2 \sin^2\theta + \Delta E_y^2({}^1/_2\cos^2\theta + {}^1/_2\sin^2\theta\\
&\quad + 2^{-1/2}\sin\theta\cos\theta) + \Delta E_z^2({}^1/_2\cos^2\theta + {}^1/_4\sin^2\theta - 2^{-1/2}\sin\theta\cos\theta)]^{1/2},
\end{aligned} \tag{5}$$

where θ is the angle between the magnetization and the [100] axis in the (110) plane.

Knowing the splitting (5), we can find the energy at 0°K:

$$F(\theta) = \frac{1}{2}\sum_i \Delta E_i.$$

The summation is carried out over all six inequivalent sites. The dependence $F(\theta)$ yields the values of the anisotropy constants at 0°K:

$$\begin{aligned}
K_1 &= 4[F([110]) - F([100])],\\
K_2 &= 9[3F([111]) + F([100]) - 4F([110]),
\end{aligned}$$

where F([111]), F([100]), and F([110]) are the values of the anisotropy energy of a cubic crystal for the magnetization along the [111], [100], and [110] axes, respectively.

The calculated values $K_1 = -5\ \text{cm}^{-1}$ and $K_2 = -24\ \text{cm}^{-1}$ are in satisfactory agreement with the experimental values $K_1 = -8\ \text{cm}^{-1}$ and $K_2 = -20\ \text{cm}^{-1}$.

Measurements of the ferromagnetic resonance in the samples with x = 0.006 and x = 0.015 under pressure (the method was described by Smith and Jones[9]) were used to calculate the magnetostriction constants λ_{100} and λ_{111}. The value of λ_{111} was deduced from measurements along the [111] axis. The dependences $\lambda_{100}(T)$ and $\lambda_{111}(T)$ are plotted in Fig. 5. We can see that the substitution with neodymium alters significantly only the constant λ_{100}, which has a positive sign for Nd^{3+}. It is interesting to note that the strong rise of λ_{100} of the neodymium-substituted samples begins from about 25°K, i.e., when an anomalous peak appears in the ferromagnetic resonance curve.

We compared the temperature dependence of the magnetostriction constant λ_{100} of the sample with x = 0.015 and the corresponding dependence calculated in the one-ion approximation:[10]

$$\lambda_{100}(T) = \lambda_{100}(0)\hat{I}_{1/2}[\mathcal{L}^{-1}(m_n)],$$

and in the two-ion approximation:[11]

$$\lambda_{100}(T) = \lambda_{100}(0)\, m_{Re} m_{Fe}.$$

The discrepancies were found to be of a qualitative nature, as in the case of the anisotropy constants (Fig. 4). It is likely that, in the case of ions having a

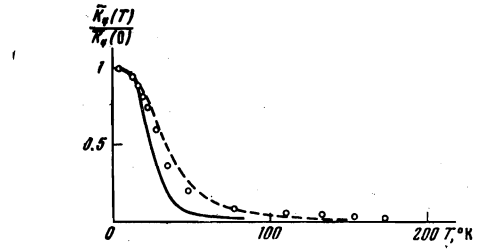

FIG. 4. Temperature dependences of $\tilde{K}_4(T)/\tilde{K}_4(0)$: the points represent the experimental values and the continuous and dashed curves correspond to the one-ion and two-ion models.

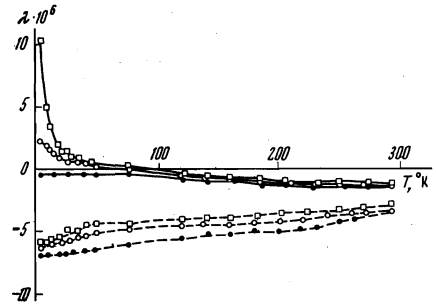

FIG. 5. Temperature dependences of the magnetostriction constants λ_{100} and λ_{111} of $Y_{3-x}Nd_xFe_5O_{12}$ single crystals: ●—x = 0.0; ○—x = 0.006; □—x = 0.015; the dashed curves represent λ_{111} and the continuous curves λ_{100}.

nonzero orbital moment, the temperature dependences of the anisotropy and magnetostriction constants cannot be explained without simultaneous allowance for the one-ion and two-ion interaction mechanisms.

It should be noted that discrepancies may also originate from the approximate nature of Eq. (3), which ignores the dependence of ΔE on the direction and anisotropy of the g factor. The temperature dependence of ΔE is not significant because considerable changes of ΔE occur near the Curie point, which is much higher than the temperatures employed in our investigation.

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