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THE EFFECT OF VANADIUM IONS ON THE MAGNETIC ANISOTROPY AND MAGNETOSTRICTION OF YTTRIUM IRON GARNET

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The anisotropy constants K and the magnetostriction constants λ have been measured by ferromagnetic resonance method. Experimental data on the effect of vanadium ions on the anisotropy and magnetostriction of YIG in the range from 4.2 up to 300 K are reported. It is shown that the anisotropy constants K and magnetostriction constants λ for vanadium doped YIG are different from those for pure YIG, in particular in the low temperature range.

1. Introduction

YIG is known as a ferromagnet and has the garnet crystal structure. Yttrium iron garnet itself is specially interesting, mainly because of its magnetic properties: very narrow resonance linewidth and the possibility of application in microwave technique.

The symmetry of the garnet lattice is cubic, space group $Oh^{10}-Ia3d$, with eight formula units in the unit cell [1]. There are 24 dodecahedral Y^{3+} sites, 16 octahedral Fe^{3+} sites and 24 tetrahedral Fe^{3+} sites.

The effect of the metal ions of the first transition group as Mn, Fe, Co, Ni, Cu [2–9], the second as Ru [10, 11] and the third as Ir [12] introduced to the garnet on the magnetic properties are well known. The influence of these ions on the anisotropy constants K and on the magnetostriction constants λ depends on the particular crystallographic site. However, up to now the influence of vanadium ions doping in garnets on the anisotropy and magnetostriction has not been investigated. This problem is very complicated. The vanadium ions introduced to the garnets can occur in divalent, trivalent, and tetravalent states with three, two and one $3d$ electrons, respectively, V^{5+} also occur but is not of interest here. According to Ref. [13] the vanadium ions are expected to occupy both octahedral and tetrahedral sites V^{3+} and V^{4+} and dodecahedral sites V^{2+}. Thus all V^{2+}, V^{3+} and V^{4+} vanadium ions can contribute to the anisotropy and magnetostriction constants. In this paper we report the effect of vanadium ions on the anisotropy constants K as well as on the magnetostriction constants λ in Yttrium iron garnet.

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2. Theory of measurements

Anisotropy

The magnetocrystalline anisotropy is defined in terms of the dependence of the free energy on the direction of magnetization. For a cubic ferromagnet the free energy per unit volumes is usually described by the following equation:

$$F[hkl] = K_0 + K_1 s + K_2 p + K_3 s^2 + K_4 sp + \ldots, \tag{1}$$

where

$$s = \alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2; \quad p = \alpha_1^2\alpha_2^2\alpha_3^2,$$

and α_i are the direction cosines of the magnetization direction related to the crystallographic axes.

The general resonance condition is given by [14]

$$\omega = \gamma H_{\text{eff}} \tag{2}$$

where γ is the gyromagnetic ratio, ω is the angular frequency and H_{eff} is the effective value of the constant magnetic field, given by [15]

$$H_{\text{eff}} = \{[H_0 + (N_x - N_z)M_s + H_x^k]\,[H_0 + (N_y - N_z)M_s + H_y^k]\}^{1/2}, \tag{3}$$

where H_0 is the external field, here assumed to be along z direction, N_x, N_y, and N_z are the shape dependent demagnetizing factors, and H_x^k and H_y^k are the components of magnetic anisotropy field and described as

$$H_x^k = \frac{1}{M_s}\frac{\partial^2 F}{\partial \theta^2}, \quad H_y^k = \frac{1}{M_s \sin^2\theta}\frac{\partial^2 F}{\partial \varphi^2}, \tag{4}$$

where θ, φ represent the orientation of the magnetization.

If the higher-order terms in equation (1) can be neglected and the resonance field is in the (110) plane, the anisotropy constants K_1, K_2 and K_3 can be calculated from equations (1), (2) and (3)

$$\frac{\omega}{\gamma} = H_{\text{res}}[100] + \frac{2K_1}{M_s}, \tag{5}$$

$$\frac{\omega}{\gamma} = H_{\text{res}}[111] - \frac{4K_1}{3M_s} - \frac{4K_2}{9M_s} - \frac{8K_3}{9M_s}, \tag{6}$$

$$\frac{\omega}{\gamma} = \left\{H_{\text{res}}[110] - \frac{2K_1}{M_s} - \frac{K_3}{M_s}\right\}^{1/2}\left\{H_{\text{res}}[110] + \frac{K_1}{M_s} + \frac{K_2}{2M_s} + \frac{K_3}{2M_s}\right\}^{1/2}, \tag{7}$$

$$\frac{\omega}{\gamma} = \left\{H_{\text{res}}[112] - \frac{K_2}{3M_s}\right\}^{1/2}\left\{H_{\text{res}}[112] - \frac{K_1}{M_s} + \frac{K_2}{18M_s} - \frac{K_3}{18M_s}\right\}^{1/2}, \tag{8}$$

where $H_{\text{res}}[hkl]$ is the resonance field in the $[hkl]$ direction in the (110) plane.

Magnetostriction

The magnetostriction constants λ_{100} and λ_{111} have been measured by the method of ferromagnetic resonance developed by Smith and Jones [16, 17]. The free energy F is given by the following standard relation for Zeeman, anisotropy and stress energy:

$$F = -HM + K_1(\alpha_x^2\alpha_y^2 + \alpha_y^2\alpha_z^2 + \alpha_z^2\alpha_x^2) + K_2\alpha_x^2\alpha_y^2\alpha_z^2$$

$$+\tfrac{3}{2}\sigma\lambda_{100}(\alpha_x^2\beta_x^2 + \alpha_y^2\beta_y^2 + \alpha_z^2\beta_z^2) + 3\sigma\lambda_{111}(\alpha_x\alpha_y\beta_x\beta_y + \alpha_y\alpha_z\beta_y\beta_z + \alpha_z\alpha_x\beta_z\beta_x), \tag{9}$$

where α_i, β_i represent the direction cosines of the magnetization $\vec{M}$ and uniaxial compressive stress σ, respectively, as referred to the cubic or $\langle 100\rangle$ axes of the crystal.

The effective value of the constant magnetic field from (2) is given by

$$H_{\text{eff}} = \{[(H_0+\delta H)+(N_x-N_z)M_s+H_x^k+H_x^{me}]\,[(H_0+\delta H)+(N_y-N_z)M_s+H_y^k+H_y^{me}]\}^{1/2}, \tag{10}$$

where H_x^{me}, H_y^{me} represent the components of field due to the magnetostriction strains, δH represents shift of the resonance field induced by compressional stress σ which has been applied along the [110] direction of the spheres.

The relation between λ_{hkl} and the shifts $\delta H_{\text{res}}[hkl]$ in the $[hkl]$ direction is given by

$$\lambda_{100} = -\tfrac{2}{3}\left(\frac{M_s}{\sigma}\right)\delta H_{\text{res}}[100], \tag{11}$$

$$\lambda_{111} = -\tfrac{4}{9}\frac{M_s}{\sigma}\left(1+\frac{K_2}{3M_sH_{\text{res}}[110]}\right)^{-1}$$

$$\left[\left(1-\frac{2K_1-K_2}{4M_sH_{\text{res}}[110]}\right)\delta H_{\text{res}}[110]+\tfrac{1}{2}\left(1-\frac{2K_1}{M_sH_{\text{res}}[110]}\right)\delta H_{\text{res}}[110]\right] \tag{12}$$

or

$$\lambda_{111} \approx -\tfrac{4}{9}\frac{M_s}{\sigma}(\delta H_{\text{res}}[110]+\tfrac{1}{2}\,\delta H_{\text{res}}[100]) \tag{12a}$$

for

$$\left(\frac{K_1}{M_s}\right), \left(\frac{K_2}{M_s}\right) \ll H_{\text{res}}[110].$$

The magnetostriction constant λ_{111} is also measured by the method of ferromagnetic resonance with the magnetic field along the $\langle 111\rangle$-axis developed by Dionne [18]

$$\lambda_{111} \approx -\tfrac{2}{3}\left(\frac{M_s}{\sigma}\right)\delta H_{\text{res}}[111]. \tag{12b}$$

To calculate σ the effective area of the spheres is taken as $\frac{2}{3}\pi r$ [19].

3. Experimental

Crystal growth and polish

The investigated samples were pure YIG crystals and vanadium doped crystals. The single crystals were grown by crystalisation from the high temperature solution (flux-technology) [20]. Crystals obtained from flux with addition of the V_2O_5 are large with well shaped surfaces. Weight of the best crystal is about 4g. Vanadium concentrations measured by the chemical analysis method were less than 50 ppm.

The samples used in the measurements were polished spheres with diameter of about 0.9 mm and oriented in a (110) plane with an accuracy of 1°. The measurements were carried out by the ferromagnetic resonance method at 9 GHz in the temperature range $4.2 \leqslant T \leqslant 300$ K.

Anisotropy

In our calculations the spontaneous magnetization M_s for vanadium doped YIG taken into account is assumed to be the same as that for pure YIG. Because such very small concentration of vanadium ions has negligible influence on the spontaneous magnetization value M_s. In Fig. 1 the experimental resonance field is plotted as a function of

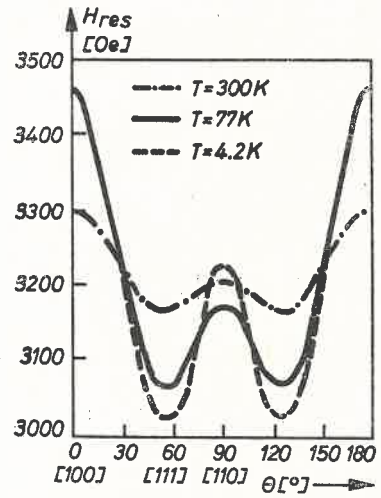

Fig. 1. Angular dependence of H_{res} in the (110) plane of vanadium doped YIG at 4.2 K, 77 K and 300 K temperatures

rotating angle in the (110) plane. The experimental points are precisely on the theoretical curve calculated from equations (1)—(3), where the three anisotropy constants K_1, K_2 and K_3 are taken into account. Some results for anisotropy constants K_1, K_2 and K_3 of the vanadium doped YIG are compared in Table I. The value presented in Table I show that at the room temperature, the anisotropy constants K_2 and K_3 for vanadium doped YIG are negligibly small, the anisotropy constant K_1 has the same sign and order of magnitude as K_1 constant for YIG [5, 21] ($K_1(300\text{ K}) = -6.1\times10^3$ erg/cm^3). The absolute

values of all anisotropy constants increase with the decreasing temperature from 300 up to 77 K. At 77 K the values of the anisotropy constants K_2 and K_3 cannot be neglected. (At 77 K, $K_1 = -24.4\times10^3$ erg/cm^3, $K_2 = -6.1\times10^3$ erg/cm^3 and $K_3 = 7.1\times10^3$ erg/cm^3). The above results are in contrast with the opinion given by authors [21–23], where they conclude that the constants K_2 and K_3 for pure YIG are negligibly small in the whole

TABLE I

Anisotropy constants K_1, K_2 and K_3 of the vanadium doped YIG at 300 K, 77 K and 4.2 K

T (K)	$\frac{K_1}{M_s}$ (Oe)	$\frac{K_2}{M_s}$ (Oe)	$\frac{K_3}{M_s}$ (Oe)
300	−43	−2	+2
77	−125	−31	+36
4.2	−112	−148	+17

temperature range. The absolute values of the anisotropy constants K_1 and K_3 decrease not significantly, however, value of K_2 strongly increases with the decrease of temperature from 77 up to 4.2 K. In particular at 4.2 K the value of K_2 is higher than that of K_1 (K_2(4.2 K) $= -28.3\times10^3$ erg/cm^3).

Magnetostriction

In our measurements the applied stress σ is about 10^7 dyn/cm^2. Some values of magnetostriction constants λ_{100} and λ_{111} obtained from Eqs. (11) and (12) for pure YIG and vanadium doped YIG are compared in Table II.

The values given in Table II show that the signs of λ_{100} and λ_{111} constants in pure YIG are negative in the whole temperature range. The magnetostriction constants λ_{100} and λ_{111} for pure YIG obtained in our experiment have the same values as λ_{100} and λ_{111} in the measurements reported elsewhere [5, 24–26] ($\lambda_{100} = -1.56\pm0.10\times10^{-6}$; $-0.55\pm0.14\times10^{-6}$ and $\lambda_{111} = -2.5\pm0.1\times10^{-6}$; $-4.45\pm0.14\times10^{-6}$ at 300 K and 4.2 K, respectively).

TABLE II

Magnetostriction constants of the pure YIG and vanadium doped YIG at 300 K, 77 K and 4.2 K

Material	T (K)	$\frac{\lambda_{100}}{M_s}\times10^9$ (Gauss^{-1})	$\frac{\lambda_{111}}{M_s}\times10^9$ (Gauss^{-1})
YIG	300	-10.8 ± 0.7	-18.0 ± 0.7
	77	-3.5 ± 0.7	-21.9 ± 0.7
	4.2	-2.8 ± 0.7	-22.6 ± 0.7
YIG+V	300	-5.6 ± 0.7	-14.1 ± 0.7
	77	-2.8 ± 0.7	-17.0 ± 0.7
	4.2	$+4.9\pm0.7$	-24.0 ± 0.7

In the temperature range from 300 to 77 K the values of the λ_{100} and λ_{111} constants in pure YIG are higher than those in vanadium doped YIG. However, the changes of the magnetostriction constants for vanadium doped YIG are more rapid than those for pure YIG with the decrease of the temperature from 77 to 4.2 K. At 4.2 K the absolute value of λ_{111} constant in vanadium doped YIG is higher than that in pure YIG (λ_{111} (4.2 K) = $= -4.73 \pm 0.14 \times 10^{-6}$). In particular the sign of λ_{100} constant varies from negative (at 77 K) to positive (at 4.2 K). ($\lambda_{100} = -0.55 \pm 0.14 \times 10^{-6}$ and $+0.87 \pm 0.14 \times 10^{-6}$ at 77 K and 4.2 K, respectively).

Conclusion

As it has been shown in the preceding section, the anisotropy constants K and magnetostriction constants λ for vanadium doped YIG are different from those for pure YIG, in particular in the low temperature range. These differences can be caused by the vanadium ions in the octahedral and tetrahedral sites of YIG.

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