In-Plane Transverse Susceptibility of (111)-Oriented Iron Garnet Films

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Iron garnet films with various compositions $Y_3Fe_5O_{12}$ (YIG), $(LuPr)_3Fe_5O_{12}$ (LuPrIG), and $Tm_3(FeSc)_5O_{12}$ (TmScIG) were grown by liquid phase epitaxy on gadolinium gallium garnet substrates. The angular dependences of in-plane transverse magnetic susceptibility $\chi'(\varphi)$ were investigated to estimate their applicability as a fluxgate core. The $\chi'(\varphi)$ dependence permitted the determination of the anisotropy field H_C in the film plane. Experimentally obtained values of H_C show good agreement with theoretical values and correspond to 2.5, 0.92, and 0.03 Oe for YIG, LuPrIG, and TmScIG epitaxial films, respectively. The influence of garnet composition on mechanisms of decreasing of induced in-plane anisotropy field was determined. TmScIG films exhibit about 100-fold reduction in the anisotropy in the plane of the film as compared with films of pure YIG.

Index Terms—**Epitaxial iron garnet film, fluxgate magnetometer, magnetic anisotropy, magnetic susceptibility.**

I. Introduction

PRINCIPAL point in determining the sensitivity of fluxgate magnetometer is the choice of material for the active media. Iron garnet films showed a high sensitivity in rotational fluxgate magnetometer with saturated core [1]. Magnetization reversal process was carried out in this magnetometer by circular rotation in the film's plane.

Thus, the sensitivity of this magnetometer was determined by transverse magnetic susceptibility in the film plane $\chi'(\varphi)$, defined crystallographic anisotropy of magnetic material.

As was shown in [2] for a cubic magnet magnetization reversal, the (111) plane is isotropic along the first cubic anisotropy constant, i.e., expansion of the free energy of the magnet in the rotational angle of the plane (111) begins with the second cubic anisotropy constants. However, this conclusion has been made on the assumption that the magnetization vector never leaves the plane.

In reality, the path of movement upon rotation of the magnetization vector in the plane (111) is more complicated, the crystallographic anisotropy displays the magnetization vector of the plane, and there are more anisotropic forces proportional to the angle of the exit plane. Analytical expressions for the magnetic susceptibility tensor in the static case were obtained in [3] and for transverse magnetic susceptibility $\chi'(\varphi)$ that corresponds to the tensor component $\chi'_{\varphi\varphi}$ of the magnetic susceptibility tensor in polar coordinates in [1].

The purpose of this paper is the investigation of transverse susceptibility in different iron garnet films to check the applicability of analytical expressions obtained in [1] and [3] and to minimize the in-plane anisotropy H_C in (111) film orientation due to crystallographic anisotropy.

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TABLE I
Melt Composition for Growth Iron Garnet Films

Mole ratio	$Y_3Fe_5O_{12}$	$Tm_3Fe_{4.3}Sc_{0.7}O_{12}$	$Lu_{2.15}Pr_{0.85}Fe_5O_{12}$
Fe_2O_3/Y_2O_3	29.9	—	—
Fe_2O_3/Tm_2O_3	—	30.0	—
Fe_2O_3/Sc_2O_3	—	30.0	—
$Fe_2O_3/(Lu_2O_3+Pr_6O_{11})$	—	—	29.6
Lu_2O_3/Pr_6O_{11}	—	—	9.3
PbO/B_2O_3	15.6	15.6	16.9
(Garnet oxides/ All oxides)·100%	13.8	13.8	17.0

II. Growth of Epitaxial Iron Garnet Films

The iron garnet films with nominal composition $Y_3Fe_5O_{12}$, $Lu_{2.15}Pr_{0.85}Fe_5O_{12}$, and $Tm_3Fe_{4.3}Sc_{0.7}O_{12}$ were grown in Scientific Research Company Carat by standard isothermal liquid phase epitaxy (LPE) method from super-cooled solution of garnet components dissolved in a solvent melt on horizontal rotating substrate. The YIG films were grown as reference samples. The Sc and Pr ions were chosen due their strong influence on magnetic anisotropy [4]–[6]. Compositions of epitaxial films were chosen to ensure minimization of crystal lattice mismatch between epitaxial film and substrate.

All technological experiments were carried out on air using the Garnet-3 (LPAI, France) five-heating-zone LPE furnace. Crucible temperature was maintained with accuracy ±0.1 °C. Substrates of the gadolinium gallium garnet (GGG) ($Gd_3Ga_5O_{12}$) single crystal with (111)-orientation, 3′ diameter and thickness of 460–490 μm having both surfaces of epitaxial grade polishing were used.

The garnet-forming oxides (Y_2O_3, Tm_2O_3, Pr_6O_{11}, Fe_2O_3, and Sc_2O_3) and melt components (PbO and B_2O_3) with purity better than 5N were used. The mole ratios (Blank–Nielsen coefficients) used for growth of epitaxial films is summarized in Table I.

The garnet-forming oxides weighed in appropriate ratio were premelted into platinum crucible with internal diameter

120 and 180 mm height together with flux components. Crucible content was maintained in growth furnace during some hours at temperature to be 100–150 °C over the saturation temperature in respect to garnet phase to dissolve the garnet-forming component completely and homogenize the melts. After reliable homogenization the melt temperature was slowly decreased till the growth one that was 5–35 °C down the saturation point, and a prepared GGG substrate held horizontally was dipped into the fluxed melt and held there for appropriate time depending on the film thickness needed. The substrate was reversely rotated with the rate of 60 r/min during growth process. The rotation rate was increased up to 200–400 r/min after withdrawal from solution to remove any residual flux from the layer surface. The thickness of grown films was determined by the weighting method, considering computed film density.

The iron garnet epitaxial films with thickness about 9 μm were grown at growth rate changed from 0.5 to 1.1 μm/min and temperature region 930 °C–980 °C.

III. In-Plane Transverse Susceptibility of Grown Epitaxial Films

The real part of in-plane complex susceptibility $\chi = \chi' - i\chi''$ of the iron garnet film is obtained by solving the Landau–Lifshitz–Gilbert equation

$$\chi' = \frac{M}{H}\left(1 - \frac{h_x}{H}\sin\varphi - \frac{h_y}{H}\cos\varphi - \frac{Ch_z}{H}\cos 3\varphi - \frac{H_c}{H}\cos 6\varphi\right) \quad (1)$$

$$H_C = \frac{K_2}{3M} - \frac{2\left(K_1 + \frac{K_2}{6}\right)^2}{M(4\pi M^2 - 2K_u + MH)} \quad (2)$$

$$C = \frac{3\sqrt{2}\left(K_1 + \frac{K_2}{6}\right)}{4\pi M^2 - 2K_u + MH} \quad (3)$$

where M is the garnet magnetization, K_u is constant of uniaxial anisotropy, K_1 and K_2 are the constants of cubic anisotropy, φ is the angle between the [211] crystallographic direction and the projection of the magnetization on the (111) plane, H is the saturating field, and h_x, h_y, and h_z are the orthogonal components of the external constant field h.

As follows from (1) by measuring the angular dependence of $\chi'(\varphi)$ it is possible to determine H_C as a signal at the six harmonic in φ. To define a constant C a constant field h_z should be applied normal to the film and detect a signal at the third harmonic in φ.

The Fourier components from dependence $\chi'(\varphi)$

$$\chi'_{S1} = \frac{1}{\pi}\int_0^{r2} \chi'(\varphi)\ \sin\varphi\, d\varphi = \frac{h_x}{H^2}M \quad (4)$$

$$\chi'_{C1} = \frac{1}{\pi}\int_0^{r2} \chi'(\varphi)\ \cos\varphi\, d\varphi = \frac{h_y}{H^2}M \quad (5)$$

$$\chi'_{C3} = \frac{1}{\pi}\int_0^{r2} \chi'(\varphi)\ \cos 3\varphi\, d\varphi = \frac{Ch_z}{H^2}M \quad (6)$$

$$\chi'_{C6} = \frac{1}{\pi}\int_0^{r2} \chi'(\varphi)\ \cos 6\varphi\, d\varphi = \frac{H_C}{H^2}M. \quad (7)$$

Thus, by measuring the Fourier components from angular dependence of the transverse magnetic susceptibility $\chi'(\varphi)$ at different h_x, h_y, and h_z allows to define the material constant C and the induced in-plane anisotropy field H_C.

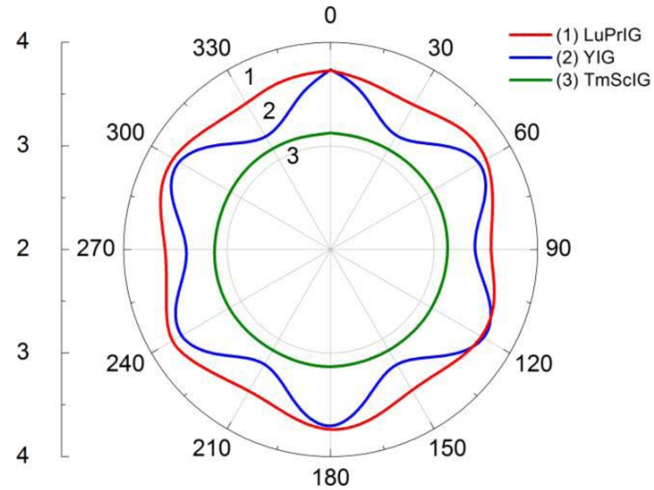

Fig. 1. Experimental angular dependence of transverse susceptibility $\chi'(\varphi)$ in a rotating field $H = 4$ mT in plane (111) for some iron garnets.

From the standpoint of the magnetometer, the obtained relations (4–7) enable us to construct an algorithm of simultaneous measurement of all three components of the external constant field h_x, h_y, and h_z by measuring the Fourier components of the transverse magnetic susceptibility $\chi'(\varphi)$.

The H_C magnitude represents the anisotropy field in the plane of rotation of the film and may even be zero for nonzero value of the constant C. It allows removing the uninformative six harmonic frequencies from the signal of the pump, while maintaining the sensitivity of the sensor in three directions.

The angular dependence of the magnetic susceptibility was measured by radiofrequency method [8] using Agilent E4980A impedance meter, which was connected to the resonator with iron garnet film placed in the Helmholtz coil system. The experimental angular dependence of $\chi'(\varphi)$ in a saturating field $H = 4$ mT for YIG, LuPrIG, and TmScIG films present on Fig. 1. The average value of the magnetic susceptibility of these garnet films differ in value due to different values of the magnetizations M. The magnitude of the sixth harmonic is proportional to H_C.

To determine the constants C the dependence of χ'_{C3} (h_z) can be used. The experimental dependence of χ'_{C3} (h_z) for YIG, LuPrIG, and TmScIG films present in Fig. 2.

The values of the constant C and the in-plane anisotropy field H_C defined from experiment and calculated according (2) and (3) shown in Table II. Values of anisotropy constant K_1 and K_u taken from sources [5]–[7]. For calculation magnetization $4\pi M$ for substituted iron garnets the extended by Dione the Neel molecular field theory for ferrimagnetism has been used [9]–[11].

Experimentally obtained values of anisotropy field H_C show good agreement with theoretical values. According to these results one can conclude that a decrease of the induced anisotropy field in the (111) plane in Pr- and Sc-substituted iron garnet films occur under different mechanism.

For Sc-substituted iron garnet decrease constant K_1 and as a consequence the decline H_C by quadratic law.

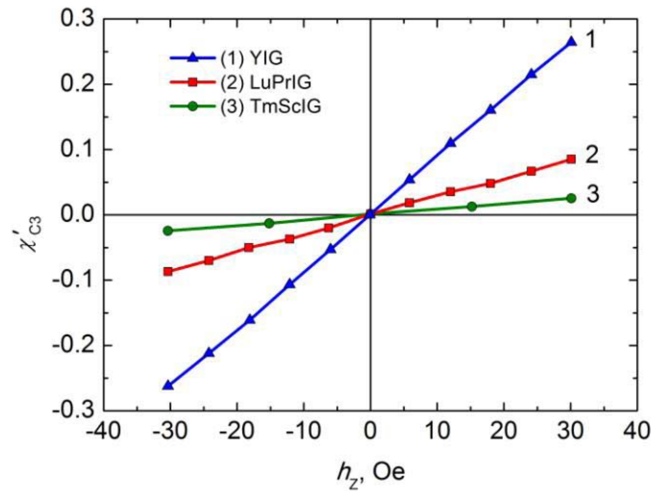

Fig. 2. Measured dependence of χ'_{C3} versus magnetic field h_z, applied normal to the film plane for different garnet.

TABLE II

COMPARISON BETWEEN EXPERIMENT AND THEORY

Symbol	$Y_3Fe_5O_{12}$	$Tm_3Fe_{4.3}Sc_{0.7}O_{12}$	$Lu_{2.15}Pr_{0.85}Fe_5O_{12}$
K_1, erg/cm3	$-6\cdot10^3$	$-0.6\cdot10^3$	$-6\cdot10^3$
K_u, erg/cm3	0	0	$-1.5\cdot10^5$
$4\pi M$, G	1750	1600	1800
H_C, Oe (theory)	2.500	0.026	0.900
H_C, Oe (experiment)	2.500	0.030	0.920
C (theory)	0.095	0.012	0.045
C (experiment)	0.100	0.011	0.036

For Pr-substituted iron garnet linear decrease of induced in-plane anisotropy field mainly due to increase uniaxial anisotropy $K_u < 0$. As one can observe from Fig. 1, cubic anisotropy compensation in TmScIG films ensures homogeneity of magnetization rotation of the garnet core, and thus can reduce magnetic noise of fluxgate magnetometer.

IV. CONCLUSION

Experimental data showed full compliance with the formulas obtained in [1] and [3]. Reduction of K_1 by a factor 10 leads to two orders of magnitude decrease of the in-plane anisotropy H_C since the anisotropy in the film plane orientation (111) depends on square value of the first cubic anisotropy constant K_1. All three used material had a negligible value of sixth order cubic anisotropy constant K_2, but is of considerable interest synthesis of materials with a positive nonzero value of K_2. In this case, according to (2) and (3) can be a value of zero at H_C with nonzero value C, to build a 3-D magnetometer. Such values of the constant K_2 can be achieved by adding Ir^{4+} and Ru^{4+} ions in the iron garnet composition [12].

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REFERENCES

[1] P. M. Vetoshko, M. V. Valeiko, and P. I. Nikitin, "Epitaxial iron garnet film as an active medium of even-harmonic magnetic field," *Sens. Actuators A, Phys.*, vol. 106, nos. 1–3, pp. 270–273, 2003.

[2] S. Chikazumi, *Physics of Ferromagnetism*. New York, NY, USA: Oxford Univ. Press, 1997.

[3] M. V. Valeiko, P. M. Vetoshko, A. J. Perlov, and A. Y. Toporov, "Effect of anisotropy constants on the magnetic susceptibility of the materials with cubic crystal structure," (in Russian), *Solid-State Phys.*, vol. 36, no. 10, pp. 3067–3070, 1994.

[4] I. I. Syvorotka, I. M. Syvorotka, and I. V. Kityk, "Surface morphological changes and magnetic properties of Sc-substituted $Y_3Fe_5O_{12}$ epitaxial films deposited on the GGG substrate," *J. Magn. Magn. Mater.*, vol. 322, no. 21, pp. 3314–3319, 2010.

[5] S. E. G. Slusky, J. F. Dillon, Jr., C. D. Brandle, M. P. Norelli, and V. J. Fratello, "Magnetic properties of praseodymium iron garnet and neodymium iron garnet," *Phys. Rev. B, Condens. Matter*, vol. 34, no. 11, pp. 7918–7923, 1986.

[6] P. Hansen, C.-P. Klages, and K. Witter, "Magnetic and magneto-optic properties of praseodymium- and bismuth-substituted yttrium iron garnet films," *J. Appl. Phys.*, vol. 60, no. 2, pp. 721–727, 1986.

[7] A. Gangulee and R. J. Kobliska, "Magnetocrystalline anisotropy in epitaxially grown $(Gd,Tm,Y)_3(Fe,Ga)_5O_{12}$ garnet thin films," *J. Appl. Phys.*, vol. 51, no. 6, pp. 3333–3337, 1980.

[8] N. N. Evtihiev and B. N. Naumov, Eds., *Elements and Bubble Magnetic Domains Devices*, Moscow, Russia: Radio i Svyaz, 1987.

[9] C. D. Brandle, S. Blank, C. Brandle, and S. Blank, "Magnetic moments for mixed substituted rare earth iron garnets," *IEEE Trans. Mag.*, vol. 12, no. 1, pp. 14–18, Jan. 1976.

[10] G. F. Dionne, "Molecular-field coefficients of rare-earth iron garnets," *J. Appl. Phys.*, vol. 47, no. 9, pp. 4220–4221, Sep. 1976.

[11] P. Roschmann and P. Hansen, "Molecular field coefficients and cation distribution of substituted yttrium iron garnets," *J. Appl. Phys.*, vol. 52, no. 10, pp. 6257–6269, 1981.

[12] P. Hansen and R. Krishnan, "Anisotropy and magnetostriction of 4d and 5d transition-metal ions in garnets and spinel ferrites," *J. Phys. Colloques*, vol. 38, no. C1, pp. C1-147–C1-155, 1977.