

Dependence of Faraday effect on the orientation of terbium–scandium–aluminum garnet single crystal

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To investigate the directional dependence of the Faraday effect in terbium–scandium–aluminum garnet (TSAG) single crystals, grown by the Czochralski method, the Verdet constant was measured at $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$ orientations. Extinction ratio and magnetic susceptibility were measured. From the linear dependence of the Verdet constant and inverse wavelength square $1/\lambda^2$, $\langle 111 \rangle$ direction shows the highest value of Verdet constant (for $\lambda = 649.1$ nm, $V_{av} = 8.256 \times 10^{-3}$ deg \cdot Oe $^{-1}$ \cdot cm $^{-1}$). Significant anisotropy of magnetic susceptibility was not observed. The extinction ratio of TSAG shows the highest value for $\langle 111 \rangle$ orientation 38.7 dB, which implies that it can be used as an optical isolator.

I. INTRODUCTION

In an optical communication system or optical head of optical disk memory, a semiconductor laser is used as the light source. The light reflected to the semiconductor laser causes the change in the oscillation mode, deterioration in the frequency properties, and the noise generation, and so forth. Therefore, to realize high-quality optical communications and to improve the signal to noise ratio, it is necessary to use an optical isolator, which prevents the reflected light from going back to the semiconductor laser.

Usually, magneto-optical (MO) effects are described in terms of the dielectric tensor $\hat{\epsilon}$ of the material in which the interaction between the light and the applied magnetic field or the internal magnetization of the material

takes place. In an isotropic material, the three diagonal elements are identical and, in the presence of a magnetic field along the z axis, a non-zero off-diagonal element e' appears, which couples the x - and y -components of the optical E -field (see, e.g., Refs. 1 and 2)

$$\hat{\epsilon} = \begin{pmatrix} e & e' & 0 \\ -e' & e & 0 \\ 0 & 0 & e_z \end{pmatrix}. \quad (1)$$

In general, e and e' are wavelength-dependent, but over a narrow range of wavelengths they might be treated as constants. In a transparent material, where there is no optical absorption, e is real and e' is imaginary. For diamagnetic and paramagnetic media, e' is proportional to the applied magnetic field H , while for ferro- and ferrimagnetic materials the spin-orbit coupling is the dominant source of the MO interaction, making e' proportional to the magnetization M of the medium. Because $B = H + 4\pi M$, we

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can treat the B -field inside the medium as the source of the MO effects.

When a polarized beam of light propagates in a medium along the direction of the magnetic field B , the material interacts with the right- and left-circularly polarized (RCP and LCP) components of the beam with different refractive indices, $n_{\pm} = \sqrt{e \pm ie'}$. For linearly polarized light passing through a length L of the material under the influence of a magnetic field B , the two circular polarization components suffer a relative phase-shift

$$\frac{1}{2} \theta = \frac{1}{2} \frac{\omega}{c} L(n_+ - n_-) \quad .$$

A change of relative phase between the RCP and LCP components is equivalent to a rotation of the plane of polarization by the Faraday angle θ . Materials showing Faraday rotation are used in optical isolators.

Magnetic garnets are transparent enough to transmit a big fraction of the light while producing a fairly large Faraday rotation. Ferrimagnetic materials, like bismuth-doped rare earth iron garnets, have high Faraday rotation angles and they are widely used in telecommunication. In the regions (visible and near-infrared) where the iron garnets have significant absorptions, paramagnetic garnets, showing much smaller Faraday effect, are used.

The most commonly used materials for the visible and near-infrared regions are terbium-doped glasses and terbium-gallium garnet (TGG) single crystals. The Verdet constant of TGG for $\lambda = 633$ nm was reported to be $7.68 \times 10^{-3} \text{ deg} \cdot \text{Oe}^{-1} \cdot \text{cm}^{-1}$ at room temperature.³ More promising material than TGG is terbium-aluminum garnet (TAG). Already in the 1960s it was reported to have a higher Verdet constant than TGG, for $\lambda = 635$ nm $10.33 \times 10^{-3} \text{ deg} \cdot \text{Oe}^{-1} \cdot \text{cm}^{-1}$ at room temperature.⁴

Terbium-aluminum garnet (TAG) shows strong (as for paramagnetic materials) Faraday effect and high transparency in the visible region.⁴ It can be used for the optical isolator (e.g., for the blue laser). The disadvantage of TAG comparing with TGG is that it cannot easily be grown from the melt, because it melts incongruently.⁵ To stabilize the garnet phase, terbium cations were partially substituted by smaller cations, Lu, Yb, or Tm. By this method, single crystals were obtained.^{6,7} However, when the concentration of terbium cation decreased, the values of Verdet constant also decreased. To prevent reduction of terbium cation concentration, but stabilize the garnet phase, substitution of the octahedral site was proposed. The scandium cation was reported as the substituting ion for aluminum in the octahedral site.⁸ Terbium-scandium-aluminum garnet (TSAG) was first prepared and reported by Brandle and Barns.⁹ Successful growth by the Czochralski method was presented by Yoshikawa et al.¹⁰ The value of the Verdet constant of TSAG is very close to that of TAG (for $\lambda = 670$ nm, $V_{av} = 9.033 \times 10^{-3} \text{ deg} \cdot \text{Oe}^{-1} \cdot \text{cm}^{-1}$).

Recently, optical isolators found application also in photonic devices.¹¹ At present, high-quality isolators are commercially available but only in the bulk form. There are some reports about the Faraday rotation in dispersions of nanoparticles.¹² The newest investigations consider the enhancement of the Faraday effect in the photonic band gap materials.^{13–16} The discussed terbium-scandium-aluminum garnet was obtained by the micro-pulling down method also in the form close to the form of the photonic crystals.¹⁷

Garnet belongs to the cubic system, therefore it is considered as an isotropic material. However, it is reported that ²⁷Al single-crystal NMR of yttrium-aluminum garnet (YAG) shows seven magnetically nonequivalent positions of aluminum.¹⁸ It means that not all octahedral and tetrahedral sites of YAG single crystal are equivalent in the magnetic field. If the dodecahedral site, which is occupied by terbium ion in TSAG, is also anisotropic in the magnetic field, then the value of the Verdet constant should depend on the crystallographic direction. In this paper, we report the anisotropy of the Faraday effect of TSAG crystal.

II. EXPERIMENTAL

TSAG single crystal was grown by the Czochralski method from an iridium crucible and using radio frequency generator. Starting materials were prepared by mixing of 99.99% pure Tb_4O_7 , Sc_2O_3 , and Al_2O_3 powders produced by High Purity Chemicals Co. (Inoueki, Japan) at the stoichiometric ratio of 1% Al_2O_3 : $\text{Tb}_{2.8}\text{Sc}_{2.2}\text{Al}_{3.0}\text{O}_{12}$. The growth atmosphere was Ar gas flow, and a YAG single crystal with $\langle 111 \rangle$ orientation was used as a seed.

The as-grown samples were oriented using back-reflection Laue method and cut at $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$ orientations.

The Faraday rotation angle θ of several samples of different orientation were measured for the 649.1, 660, 670, 780, and 845.7 nm laser beam wavelength, λ , in a magnetic field $H = 3200$ Oe at room temperature. The Verdet constant, V , is defined as

$$V = \theta/HL \quad , \quad (2)$$

where L is the sample length.

The extinction ratio (ER) was measured at $\lambda = 649.1$ nm in a magnetic field $H = 4800$ Oe at room temperature. Faraday rotation angle and extinction ratio were measured for three orientations of TSAG crystals. In each case, two or three samples were measured at three points. Average values of these data were used for interpretation.

The magnetization was measured with a QUANTUM DESIGN MPMS-5S SQUID magnetometer at the temperature range from 4 K to 300 K in a 1-T magnetic field.

TABLE I. Results of Verdet constant $V[\times 10^{-3} \text{ deg} \cdot \text{cm}^{-1} \cdot \text{Oe}^{-1}]$ measurements.

Orientation	Wavelength (nm)					$y = ax + b$	
	649.1	660	670	780	845.7	Slope a	Intercept b
$\langle 111 \rangle$	8.256	7.866	7.475	5.443	4.278	3.92×10^3	-1.12×10^{-3}
$\langle 110 \rangle$	7.958	7.670	7.204	5.235	4.228	3.75×10^3	-0.98×10^{-3}
$\langle 100 \rangle$	7.693	7.397	7.249	4.955	3.995	3.81×10^3	-1.32×10^{-3}

$H = 3200 \text{ Oe}$. The average values of six to nine data are presented. Slope a and intercept b of the regression lines, $y = ax + b$, where $x = \lambda^{-2}$, of Verdet constant versus inverse λ^2 of TSAG are described.

The effective number of Bohr magnetons, μ_B , of terbium cation was calculated from the Curie-Weiss law.

III. RESULTS AND DISCUSSION

Verdet constants of TSAG crystals measured in three crystallographic directions, $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$, are shown in Table I. The Verdet constant of rare earth aluminum garnets can be described by following equation¹⁹⁻²⁵

$$V = -\frac{\omega^2}{\omega_0^2 - \omega^2} \left[\frac{2\omega_0 A}{\hbar(\omega_0^2 - \omega^2)} + B + C_\chi \right] + V_{\text{gm}} \quad (3)$$

where ω is the frequency of incident light, ω_0 is the effective transition frequency, the first term is the diamagnetic contribution caused by the splitting of the excited multiplets, and the second term, B , describes the contribution of mixing of the wave functions of the ground-state and the excited-state. The third term, C_χ , where χ is the magnetic susceptibility, is the paramagnetic contribution caused by the difference in the population of the ground state multiplet. The fourth term, V_{gm} , is the gyromagnetic contribution to the Verdet constant. When the frequency is $\omega \ll \omega_0$, we can replace $\omega/(\omega_0^2 - \omega^2)$ by ω/ω_0^2 . In the case of terbium garnet, the diamagnetic contribution A is almost zero. And the contribution of the term B is 5% to 10%. For the terbium-scandium-aluminum garnet, the paramagnetic contribution is the main one. The gyromagnetic contribution, V_{gm} , does not depend on the frequency. From Eq. (3), the Verdet constant of terbium-aluminum garnet is proportional to the frequency square. In other words, the Verdet constant is proportional to the inverse wavelength square (λ^{-2}). The graphs of the Verdet constant versus λ^{-2} for the as-grown crystals are shown in Fig. 1. The Verdet constant is plotted linearly. The regression lines are described by linear relations, $y = ax + b$, where x is equal to λ^{-2} . Intercept b depends on the directions. The intercept of $\langle 110 \rangle$ direction is the biggest one, $\langle 111 \rangle$ is the middle, and $\langle 100 \rangle$ is the smallest one. This is what we expected because b represents the gyromagnetic part in Eq. (3), and it contains the spectroscopic splitting factor g , which is in general a tensor, and for the particular direction of magnetic field, its value depends on the angle that the

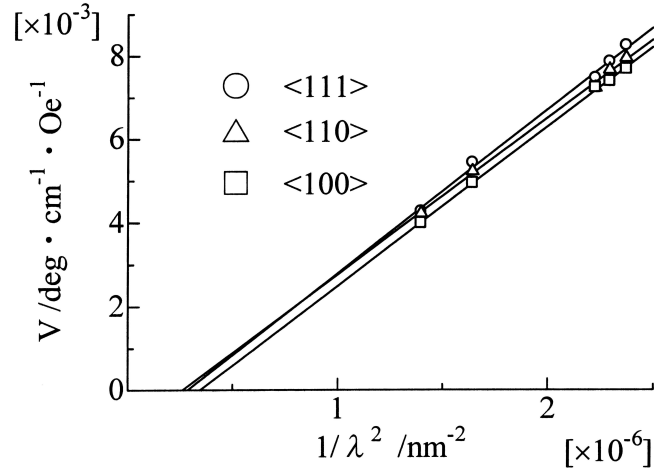


FIG. 1. λ^{-2} dependence of Verdet constant of $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$ orientations of TSAG crystals. The error bars of Verdet constant are within the mark.

external magnetic field makes with the axis defined by the local symmetry of the magnetic complex. (For example, for $\text{Gd}_3\text{Ga}_5\text{O}_{12}$, frequency-independent gyromagnetic contribution to Verdet constant is given as $C_m[(S + 1)g\mu_B / 3kT]$, where C_m is a constant [deg/cm], μ_B is Bohr magneton, S is total electron spin.) The paramagnetic contribution to the Verdet constant is directly proportional to the magnetic susceptibility. And the magnetic susceptibility depends on g^2 , therefore we should also observe a dependence of the slope a on the crystallographic direction.

$$\chi_A = \frac{Ng^2\mu_B^2}{3kT} J(J + 1) \quad ,$$

(where N is Avogadro number, μ_B is Bohr magneton and J is total angular momentum.) The slope a slightly depends on the directions. The $\langle 110 \rangle$ direction is less sensitive to λ^{-2} .

Because the difference of the slope a , which contains paramagnetic contribution C_χ , is the biggest between $\langle 111 \rangle$ and $\langle 110 \rangle$ in the as-grown samples, magnetic susceptibility of $\langle 111 \rangle$ and $\langle 110 \rangle$ as-grown samples was measured. The magnetic moment obeys Curie's law. The inverse magnetization versus temperature is shown in Fig. 2. The effective number of Bohr magnetons of Tb

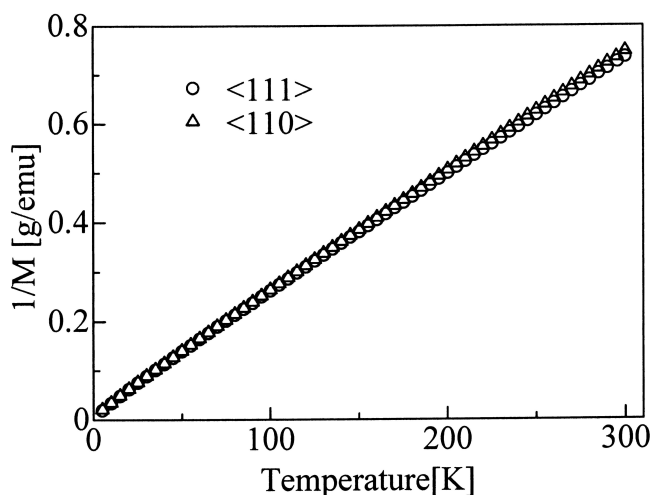


FIG. 2. Temperature dependence of inverse magnetization of TSAG $\langle 111 \rangle$ and $\langle 110 \rangle$ as-grown samples.

cation was calculated. The obtained experimental value of $\langle 111 \rangle$ orientation was $\mu_{\text{eff}\langle 111 \rangle(\text{exp.})} = 9.49\mu_{\text{B}}/\text{Tb ion}$, and that of $\langle 110 \rangle$ orientation $\mu_{\text{eff}\langle 110 \rangle(\text{exp.})} = 9.43\mu_{\text{B}}/\text{Tb ion}$. These values are higher than reported for the crystals grown by micro pulling down method, where the magnetic susceptibility was measured for the powdered crystal.⁶ The theoretical values are $\mu_{\text{eff}\langle 111 \rangle(\text{theor.})} = 9.72\mu_{\text{B}}/\text{Tb}^{3+}$ and $\mu_{\text{eff}\langle 110 \rangle(\text{theor.})} = 7.86\mu_{\text{B}}/\text{Tb}^{4+}$. The graph of inverse magnetization versus temperature for the $\langle 111 \rangle$ orientation is almost the same as that for the $\langle 110 \rangle$ orientation. The difference of the effective number of Bohr magnetons of the Tb cation is very small. Because the magnetic susceptibility does not depend on the orientation in the case of TSAG, the paramagnetic contribution to Verdet constant C_{χ} does not depend much on the orientation. The main contribution to the anisotropy dependence of Verdet constant comes from the gyromagnetic contribution V_{gm} .

The extinction ratio (ER) is the property that characterizes the ellipticity of the light transmitted through the material showing Faraday effect. It depends on the homogeneity of the material. If the extinction ratio is large, the elliptically polarized light approaches the linearly polarized light. The isolation properties of an isolator are described by the magnitude of the extinction ratio. Materials showing a higher extinction ratio value than 30 dB can be used as an optical isolator. The average values of extinction ratios of as-grown TSAG samples are as follows: $ER\langle 111 \rangle = 38.70$ dB, $ER\langle 110 \rangle = 26.8$ dB, and $ER\langle 100 \rangle = 24.2$ dB. The ER is significantly larger for the $\langle 111 \rangle$ orientation than for the other orientations. It is big enough for the optical isolator. Direction $\langle 111 \rangle$ is a special one for garnets. Garnet crystals are the most often grown in this direction. Along the $\langle 111 \rangle$ axis, the octahedral atoms are placed. There are quite a few works, especially using extended x-ray absorption fine structure,

showing that the garnet can lower its symmetry²⁶ (though it is not seen by x-ray diffraction XRD measurements) because some of the atoms from dodecahedral positions (usually yttrium or lanthanide ions), in this work terbium, can replace some atoms in octahedral positions. The symmetry is then lowered, but the threefold symmetry in the $\langle 111 \rangle$ direction stays unchanged.²⁷ From the ER results, it seems that the most stable symmetry in the distinguished direction $\langle 111 \rangle$ is the best also for avoiding the ellipticity in the Faraday effect. The other possible reason why samples cut perpendicularly to the $\langle 111 \rangle$ direction have a much higher value of ER than other samples is the good radial homogeneity of the crystal and the not-as-good homogeneity along the crystal. The crystal composition is slightly changing during growth (along the crystal) because of the noncongruent growth of TSAG. During growth, the concentration of scandium ions is slightly increasing (the distribution coefficient k_{eff} of $\text{Sc}^{3+} = 0.87$), and the concentration of aluminum ion is slightly decreasing (k_{eff} of $\text{Al}^{3+} = 1.04$) in the crystal as was presented elsewhere.¹⁰ Therefore, the homogeneity in the other direction is not perfect considering the distribution of ions in the crystal, and this is the next probable cause of lowering of the extinction ratio value. Yet even the crystals grown in a direction different from $\langle 111 \rangle$ can have a similar effect of the dependence of ER on the crystallographic direction because of the special role of the $\langle 111 \rangle$ crystallographic direction in garnet crystal, described above. Probably the only way to check it properly is to grow crystals in different orientations.

Because the Verdet constant has the highest value for the $\langle 111 \rangle$ orientation, and also the extinction ratio for this orientation is above 30 dB, we can conclude that TSAG single crystals, grown in the $\langle 111 \rangle$ crystallographic direction, can be used as an isolator when the incident light and the orientation of external magnetic field are parallel to the $\langle 111 \rangle$ orientation.

IV. CONCLUSIONS

The Verdet constant, magnetic susceptibility, and extinction ratio of TSAG single crystals in three crystal orientations, $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$, were measured. The Verdet constant slightly depends on the orientation of the crystal. For the as-grown crystal, the highest magnitude was obtained for the $\langle 111 \rangle$ orientation. The magnetization was not found to depend on the orientation. Therefore, we conclude that the main contribution to the anisotropy of the Faraday effect in TSAG single crystal comes from the gyromagnetic part.

The extinction ratio of $\langle 111 \rangle$ orientation is 38.7 dB; this orientation of TSAG crystal grown in the $\langle 111 \rangle$ direction seems to be the only one that can be used for application as an optical isolator.

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