Thermo-Optical and Magneto-Optical Characteristics of Terbium Scandium Aluminum Garnet Crystals

Ilya L. Snetkov, Ryo Yasuhara, Aleksey V. Starobor, Evgeniy A. Mironov, and Oleg V. Palashov

Abstract—Magnetoactive materials are of considerable current interest, primarily for applications in nonreciprocal Faraday devices used for polarization control, optical isolation, optical switching, and modulation. The need for such devices is growing with laser power enhancement. They reduce risk of self-excitation of the amplifiers and optical elements damage and are a handy tool for optical equalization. However, at high average power of radiation these devices are subject to thermally induced effects that impair their operability and lead to increased losses and to the formation of phase distortions in the transmitted radiation. One of the methods to reduce thermally induced effects is to use in Faraday devices new magnetoactive materials with better thermo-optical properties. This paper is devoted to the study of thermo-optical and magneto-optical characteristics of a unique magnetoactive material—a terbium scandium aluminum garnet (TSAG) crystal. The TSAG has an extraordinary value of optical anisotropy parameter ξ, a Verdet constant 25% higher than the traditionally used terbium gallium garnet crystal and the highest magneto-optical figure-of-merit known in magnetoactive materials at the moment.

Index Terms—Magneto-optical materials, Faraday effect, thermo-optic effects.

I. INTRODUCTION

With the increase of average power of CW and pulse periodic lasers over the past few years, the problem of reducing thermal effects arising in their optical elements is becoming increasingly more important. Heat release caused by partial radiation absorption in the bulk of the laser optical element results in average temperature increase, formation of a temperature gradient and associated mechanical stresses. This leads to parasitic effects, such as thermal lens (due to the temperature dependence of refractive index and element length) and thermally induced birefringence (due to the photoelastic effect). In turn, these parasitic thermal effects deteriorate the quality of laser radiation and increase laser losses upon the whole [1]–[4], thus being one of the causes restricting further enhancement of laser average power. Among the important optical elements of the laser are Faraday devices used for isolating optical radiation, controlling its polarization, providing multipass amplification schemes and compensating thermally induced depolarization. These devices are among those in which thermal self-action of laser radiation is most pronounced due to relatively high absorption (∼10−3 1/cm) in magneto-optical elements (MOEs) and the need to use relatively long MOEs (∼1-2 cm). Thermally induced birefringence arising in magneto-optical elements fully determines the efficiency of the devices at high average power of transmitted radiation [5], [6]. Thus, search for methods to reduce and compensate thermal effects in Faraday devices [7], as well as search and study of the properties of new magneto-active materials with better mechanical and thermo-optical properties are of current importance.

Today, one of the most popular and well studied magneto-active materials for Faraday devices used in high average power lasers is terbium gallium garnet (TGG) by virtue of a rather high Verdet constant (35-40 rad/(T·m) at the wavelength of 1 micrometer [8]), relatively high thermal conductivity (4.4-5.3 W/(m·K) [8]) and a possibility to grow high-quality large-size (30 mm) crystals using the Czochralski method [7], or a possibility to make high-quality ceramic elements [9]–[12]. However, other materials with better magneto-optical properties have long been known. One of the promising magneto-active materials is terbium scandium aluminum garnet (TAG) – a terbium aluminum garnet (TAG) in which part of Tb and Al ions are substituted by Sc to stabilize garnet phase. The synthesis of a stable Garnet phase in TAG was first reported in the early eighties [13]. Later, the influence of the composition and growth conditions on the quality of the obtained crystals was studied [14], [15]. A possibility of crystal growth by different methods was demonstrated and the influence of crystal composition on the value of Verdet constant was investigated [16]. The impact of annealing and the atmosphere in which it is performed on the value of Verdet constant was analyzed in the work [17].
The Verdet constant at several wavelengths was measured in some researches [14], [17]–[20]. The Verdet constant in TSAG was compared with that in TGG [14], [18] and in TAG [19], and it was shown that they are close in TSAG and TAG and are 15-20% higher than in TGG. Also, the temperature dependence of TSAG magnetization [16], [17], [20] and the reflection–transmission spectra by which dielectric permittivity was calculated as a function of wavelength were measured [21], [22]. At the same time, thermo-optical properties of TSAG and its use for a Faraday isolator (FI) were not studied until recently. The first FI with a traditional properties of TSAG and its use for a Faraday isolator (FI) was reported [21], [22]. At the same time, thermo-optical characterizations, which indicates that the depolarization was thermally increased, the depolarization showed a quadratic dependence profile was employed as a source [Fig. 1(c)]. Its radiation was adjusted to minimum power (θ = 0), and when the angle between them was 45 degrees.

Thermally induced depolarization was measured by a standard technique used for heating the studied sample and reading the thermally induced birefringence, which led to the polarization distortions of radiation. Distribution of polarization distortions was registered by CCD camera 6. Calcite wedge prism 1 ensured linearity of the polarization and high contrast better than 2·10⁻⁶ throughout the power range. Fused silica wedges 4 were used to attenuate radiation. Glan prism 5 was adjusted to minimum power Plaser of the laser signal. By rotating the Glan prism by an angle of 90° we measured the power of the laser signal Plaser in the main polarization.

The integral thermally induced depolarization γ = Pdlaser/Plaser was calculated by the ratio of the power in two positions of Glan prism. The results of measurements are presented in Fig. 2.

The integral thermally induced depolarization as a function of normalized absorbed power are plotted by dotted lines in Fig. 2(a). At low power, the depolarization in the crystal was very low (less than 2·10⁻⁶), which was determined by the contrast of the measuring scheme, indicating a high optical quality of the sample. As the power was increased, the depolarization showed a quadratic dependence on the normalized absorbed power, and the structure of local depolarization distribution was a Maltese cross in both positions, which indicates that the depolarization was thermally increased.
induced [Fig. 2(b) and 2(c)]. The maximum of the thermally induced depolarization corresponded to the crystal position in which the direction of incident radiation polarization coincided with the direction of one of the crystallographic axes (θ = 0), hence |ξ| > 1 [26], [28]. From (1) it follows that by the ratio of thermally induced depolarizations in two positions it is possible to find the absolute value of parameter ξ

\[
ξ = \frac{τ(θ = 0)}{τ(θ = 45°)}^{1/2},
\]

that was |ξ| = 101 ± 10 for the sample under consideration. The sign of the parameter ξ was determined by the behavior of local depolarization distribution in the course of crystal rotation relative to the radiation propagation direction. When the studied sample was continuously rotated in the experiment, the Maltese cross was also rotating continuously, which is a consequence of the negative sign of the parameter ξ [28]. Thus, ξTSAG = −101 ± 10.

Two bright features of the magneto-optical TSAG crystal distinguishing it from the other widely used TGG crystal are worthy of mention (see Table 1). The first is a large absolute value of parameter ξ that results in the difference of thermally induced depolarizations in the two positions of the [001] oriented crystal by more than four orders of magnitude (!) and, as a consequence, in the dramatic significance of the choice of orientation of the crystallographic axes of the used samples. The magnitude of thermally induced depolarization in the [111] orientation differs from τ[001](θ = 45°) by the multiplier (1 + 2ξ)2/9 [31], and for the ceramic element by the multiplier (2 + 3ξ2)2/25 [32], which is on the order of several thousands in both cases for the TSAG material because of a large value of ξ. The numerical computation of γ(p) for the [111] single crystal and TSAG ceramics is presented in Fig. 2(a) by green and black dashed curves, respectively, where the advantage of the [001] orientation is quite pronounced. Note also a higher sensitivity of the value of depolarization to the angle θ. The turn of the TSAG crystal with [001] orientation by an angle of 1 degree from the minimum position increases depolarization by a factor of ~10, which greatly enhances requirements to accuracy of its alignment.

The other feature is the negative sign of parameter ξ. The authors of [25], [28], [33] showed that materials with negative ξ have the orientation of crystallographic axes for which the directions of eigenpolarizations of arising thermally induced birefringence do not depend on transverse coordinates. The direction of this orientation depends on absolute value of parameter ξ. Therefore, by setting equal directions of radiation polarization and one of eigenpolarizations it is possible to fully eliminate thermally induced depolarization of transmitted radiation. For the obtained value of parameter ξ, this orientation is close to [001], differing from it by ~6 degrees only.

### III. THERMO-OPTICAL CONSTANTS Q AND P

The thermo-optical constant Q characterizes the magnitude of thermally induced birefringence of material [5], [26] and according to Eq. (2) makes a substantial contribution to the aspect ratio between the normalized heat release power p and the real laser power P_{laser}. It enters in (1) for thermally induced depolarization along with the absorption coefficient \( \alpha_0 \) and thermal conductivity \( \kappa \). The smaller \( \alpha_0 |Q|/\kappa \), the less thermally induced depolarization of transmitted radiation is. According to (1) and (2), from the dependence of γ on laser radiation power (Fig. 1) the value of \( \alpha_0 |Q|/\kappa \) for TSAG was found to be 1.44·10⁻⁹ W. The thermal conductivity coefficient of the studied TSAG sample was measured at room temperature using the method based on phase-shifting interferometry proposed in the work [34]. It was \( \kappa = 3.6 ± 0.3 \) W/(m·K), which is 20% less than the thermal conductivity coefficient of the TGG crystal reported in [34] (\( \kappa = 4.2 ± 0.3 \) W/(m·K)). Making use of the obtained value of thermal conductivity, \( \alpha_0 |Q|/\kappa \) was assessed to be 5.2·10⁻⁹ 1/(m·K). For comparison, in the TGG crystal \( Q = −17·10⁻⁷ \) 1/K [8], and the absorption coefficient \( \alpha_0 \) varies from 1.3·10⁻³ 1/cm to 4.8·10⁻³ 1/cm [7], [8], hence, \( \alpha_0 |Q| \) may take on values from 2.2·10⁻⁷ 1/(m·K) to 8.1·10⁻⁷ 1/(m·K).

As was mentioned above, thermally induced depolarization in materials is proportional to γ ∼ (\( \alpha_0 |Q|/\kappa \))^2 (see (1) and (2)). Comparison of the values obtained for TSAG and TGG (\( \alpha_0 = 1.3·10⁻³ \) 1/cm, \( Q = −17·10⁻⁷ \) 1/K, \( \kappa = 5 \) W/(m·K) [35], [36]) for crystals with the same length and laser radiation power gives γTSAG/γTGG = 942(1), i.e., by virtue of the small value of \( \alpha_0 |Q| \) the TSAG single crystal in the minimum position introduces about three orders of magnitude less thermally induced depolarization than the TGG single crystal of a good optical quality.

Thus, still another specific feature of TSAG compared to TGG is essentially small value of Q, which leads to a large value of the aspect ratio between the normalized heat release power p and the real laser power P_{laser}. For our sample it is

<p>| Table 1: Thermooptical Properties of the TSAG and TGG Crystals |
|-----------------|------|----------------|----------------|---------|</p>
<table>
<thead>
<tr>
<th>ξ</th>
<th>κ</th>
<th>α₀</th>
<th>α₀P</th>
<th>P/Q</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSAG</td>
<td>-101</td>
<td>3.6</td>
<td>0.052</td>
<td>2.1</td>
</tr>
<tr>
<td>TGG</td>
<td>2.25</td>
<td>4.25</td>
<td>2.2-8.1</td>
<td>2.2-8.1</td>
</tr>
</tbody>
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Fig. 2. (a) Thermally induced depolarization as a function of normalized absorbed power \( p = \exp TSAG_{\theta}(\theta = 45°) \) and \( p = \exp TSAG_{\theta}(\theta = 0°) \); for the ceramic element by the multiplier \( (2−10) \).
described by the following expression:

$$P_{\text{laser}}/P \approx 9.6 \times 10^4 \text{ W}$$.

A small absolute value of $Q$ and a large absolute value of $\xi$ may be attributed to the closeness of TSAG piezo-optical coefficients $\pi_{11}$ and $\pi_{12}$. As follows from (2), the difference between $\pi_{11}$ and $\pi_{12}$ determines the sign of the thermo-optical characteristic $Q$, which was found using the procedure described in our early paper [32]. Changes in the local depolarization distribution occurring with small variation of the ellipticity of incident radiation polarization were measured. The experiment demonstrated that the signs of $Q_{\text{TSAG}}$ and $Q_{\text{TGG}}$ coincide and are negative [37], [38].

Making use of the method of phase-shifting interferometry analogously to the work [32] we measured the thermally induced phase distortions introduced by our TSAG crystal with [001] orientation and, for comparison, under the same conditions in two TGG crystals from different producers, both with [001] orientation but having different absorption and length (TGG 1: $L = 9.1$ mm, $a_0 = 1.37 \times 10^{-3}$ 1/cm; TGG 2: $L = 9.3$ mm, $a_0 = 2.3 \times 10^{-3}$ 1/cm). Heating was performed by an Yb-fiber laser with maximum power of 50 W, Gaussian intensity profile and radius at which the intensity drops to $1/e$ at the location of the studied sample $r_h = 0.58$ mm. The results of the experiment are presented in Fig. 3. One can see in the figure that the sample under consideration introduces the same lens as TGG 1 single crystal.

The theoretical dependence of thermal lens force on laser radiation power neglecting distortion of optical surfaces is described by the following expression:

$$F = \frac{a_0 L P_{\text{laser}}}{2 \pi r_h^2} [P - X (1 - \xi) Q],$$

$$P = \frac{dn_0}{dT} + a_T n_0^3 \frac{E}{4} \frac{1}{1 - v} (\pi_{11} + 3 \pi_{12}),$$

where $P$ is the thermo-optical constant responsible for the average lens between two eigenpolarizations; $X$ is the coefficient taking on the values $X_{[001]} = 0$, $X_{[111]} = 1/3$, $X_{\text{ceramic}} = 1/5$; and $dn_0/dT$ is the temperature coefficient of the index of refraction. From the results obtained in experiment and (4) the $a_0 P/k$ ratio was estimated to be $5.7 \times 10^{-3}$ 1/W. With allowance for the thermal conductivity, $a_0 P$ for TSAG was $2.1 \times 10^{-6}$ 1/(m-K). Knowing the value of $a_0 Q$, from the measurements of thermally induced depolarization we found $P/[Q] = 395$. We present for comparison analogous values for TGG: $P/[Q] = 10$ [8]; $a_0 P$ varies from $2.2 \times 10^{-6}$ 1/(m-K) to $8.1 \times 10^{-6}$ 1/(m-K), from which it follows that due to the closeness of constants $P$ the thermal lenses in TSAG and TGG are close. If we assume, as before, that the absorption of the studied TSAG sample is $a_0 = 1.3 \times 10^{-3}$ 1/cm, then for equal lengths of the elements, laser beam radii and absorption, the TSAG single crystal with [001] orientation will introduce only a 26% larger lens than the TGG single crystal with [001] orientation. If the absorption in the studied TSAG single crystal is larger, then the estimates will be better and the difference between the thermal lens forces will reduce.

It is also worthy of notice that for the TSAG single crystal ($1 - \xi$)$a_0 Q = -5.3 \times 10^{-7}$ 1/(m-K). Consequently, according to (4) the thermal lens in a TSAG single crystal with [111] orientation and TSAG ceramics will be 33% and 20% larger, respectively, than in a single crystal with [001] orientation.

**IV. VERDET CONSTANT AND MAGNETO-OPTICAL FIGURE OF MERIT**

The Verdet constant $V$ is an important parameter of magneto-active materials. The Verdet constants of TSAG and TGG single crystals were measured under the same conditions at room temperature for several wavelengths using a magnetic system made of Nd-Fe-B magnets with maximum field of 1.4 T on the axis. The crystals were arranged inside the magnetic system that was placed between two crossed polarizers. By the angle of rotation of the second polarizer after the crystal, we measured the angle of Faraday rotation of the plane of polarization to an accuracy of $\pm 2$ minutes. Knowing the magnetic field distribution in the system, crystal position and its length we determined the Verdet constant at each of the considered wavelengths. The results of the experiment are presented in Fig. 4 and in Table II. For comparison, the plots also contain data for the Verdet constant obtained by other authors. The Faraday polarization rotation in TSAG and TGG crystals occurs in the same direction. The Verdet constant of the TSAG crystal in the studied wavelength range is, on the average, 25% larger than of the TGG crystal [Fig. 4(b)], which is in a good agreement with results of other researches and is close to the value of $V$ of the TAG material [39].

Different authors use different expressions:

$$V = A + B \lambda^2$$ [40];

$$V = A + B \lambda^2 + C \lambda^4$$ [41];

$$V = A \lambda^B (A = 6 \times 10^6 \text{ min/(Oe-cm)}; B = -2.5375$$ [42] for...
approximation of the experimentally obtained dependences of Verdet constant on wavelength (Fig. 4(a) dashed curves). The experimental and theoretical curves for the Verdet constant versus the radiation wavelength for the TGG single crystal obtained in the present work (squares) and taken from other literature sources are presented in Fig. 4(a). The approximations proposed in the works [40], [41] are in a good agreement with the experimental data only in the 500-900 nm region and poorly describe the dependence of the Verdet constant on the wavelengths more than 1100 nm (they differ from the experimental value by a factor of 2 already at 1310 nm).

At the same time, the dispersion of the Faraday rotation was described theoretically in paramagnetic and diamagnetic materials [43]–[45] and may be written in the form

\[ V = V_{\text{para}} + V_{\text{diam}}, \]

\[ V_{\text{para}}(\lambda) = \frac{A}{\lambda_2 - \xi}, \]

\[ V_{\text{diam}}(\lambda) = \frac{1}{2} \left[ B + \frac{C}{\lambda_2 - \lambda_0^2} \right], \]

where \( A, B, \) and \( C \) are free parameters, \( \lambda_1 \) is the effective transition of the wavelength, and \( \lambda_0 \) is mean ultraviolet resonance wavelength. In the works enumerated above, the Verdet constant dispersion describes separately \( V_{\text{para}} \) or \( V_{\text{diam}} \), as in the studied wavelength ranges in the materials under consideration one component markedly prevails over the other. TGG and TSAG are paramagnetic materials, but when dispersion of their Verdet constants is described only by means of \( V_{\text{para}} \), the experimental data for \( V(\lambda) \) obtained in the shortwave and longwave parts of the spectrum are described poorly and the increase of the \( V_{\text{TSAG}}/V_{\text{TGG}} \) ratio with increasing wavelength cannot be described at all [Fig. 4(b)]. Both terms, \( V_{\text{para}} \) and \( V_{\text{diam}} \), were obtained for approximation in this work. The method of least squares was used to determine the coefficients \( A, B, C, \lambda_1, \) and \( \lambda_0 \) for TGG and TSAG materials that agreed well (not worse than 5%) with the experimental data throughout the studied wavelength range that coincided with the transparency range of these materials. The found parameters of the studied materials are the following: \( A = 6.70 \times 10^7 \text{ rad-mm}^2/(\text{T-m}), \)

| \( \mu \cdot 10^6 \), at \( \lambda = 1075 \text{ nm} \) rad W/(T m) | \( [\nu(\lambda, \text{nm})], \text{rad/(T m)} \) |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| TSAG | 32.1 | 599 | 256.6 | 165.8 | 91.2 | 46.2 | 27.9 | 15.8 |
| TGG  | 1.1  | 466.5 | 209.1 | 135.8 | 75.9 | 37  | 20.7 | 8.9  |

Note that our experiments demonstrated that the \( V_{\text{TSAG}}/V_{\text{TGG}} \) ratio increased with increasing wavelength [Fig. 4(b)]. This makes the use of the TSAG material instead of the traditional TGG at wavelengths exceeding 1100 nm still more attractive.

Knowing the Verdet constant \( V \) of the material and the value of \( \alpha_0|Q/\kappa \) obtained earlier it is possible to determine the parameter of magneto-optical figure of merit \( \mu \) that characterizes the medium from the viewpoint of the isolation ratio at high average power [5]:

\[ \mu = |V\kappa/\alpha_0|Q|. \tag{7} \]

The larger \( \mu \), the better the magneto-active medium is. The authors of some works use the magneto-optical figure of merit \( \mu^* = V/\alpha_0 \), that gives a worse description of the quality of magneto-active material from the viewpoint of the isolation ratio at high average power, because it neglects important parameters, such as thermal conductivity and thermo-optical constant \( \kappa \). Some magneto-optical glasses have comparable \( \mu^* \) with the TGG crystal, but the Faraday isolators based on these glasses provide a much worse isolation ratio at high average power because of an order of magnitude lower thermal conductivity. According to (8), the magneto-optical figure of merit for the TSAG material \( \mu = 32.1 \times 10^8 \text{ rad-W/(T-m)} \) for the wavelength of 1070 nm at room temperature, which is a factor of \( \sim 30 \) more than \( \mu \) of the TGG single crystal under the same conditions (1.1 \times 10^9 \text{ rad-W/(T-m)} [46] and a factor of \( \sim 320 \) more than \( \mu \) of the best known magneto-optical glass (0.1 \times 10^9 \text{ rad-W/(T-m)} [8]. At high average power, from the viewpoint of a possible isolation ratio, TSAG is better than TGG as much as TGG is better than magneto-optical glass.

V. Conclusion

Thermo-optical and magneto-optical properties of the promising magneto-active TSAG material manufactured by Oxide Corporation have been studied at room temperature. The parameter of optical anisotropy \( \zeta \) was measured for the first time; the thermo-optical characteristics \( P \) and \( Q \) were estimated; the thermal conductivity was determined; the Verdet constant was measured in a wide wavelength range and the approximation describing the obtained experimental results to an accuracy not worse than 5% throughout the transparency range of the material was found; and the magneto-optical figure of merit of the studied material was calculated. The obtained characteristics were compared with analogous parameters for the broadly used magneto-active material TGG.
As compared to TGG, the TSAG single crystal possesses four specific features. The first of them is a very high absolute value of parameter $\xi = -101\pm 10$ that is more than an order of magnitude higher than all the known values for other crystals ($\xi_{\text{TGG}} = 2.2$ [31]; $\xi_{\text{YAG}} = 3.2$ [47]; $\xi_{\text{CaF}_2} = -0.47$ [28]), which makes the choice of crystal orientation extremely important.

The second feature is the negative sign of parameter $\xi$ from which it follows that this material has the orientation of the crystallographic axes at which thermally induced depolarization does not arise. Also, based on the results of the crystallographic axes at which thermally induced depolarization without using reciprocal polarization elements, such as a quartz rotator or a half-wave plate, thus opening up opportunities for implementing new schemes of Faraday isolators with compensation. To the best of our knowledge, TSAG is the first magneto-active material with a negative sign of parameter $\xi$.

The third feature is a very small value of the thermo-optical characteristic $Q$, which in the [001] crystal orientation, gives thermally induced depolarization about 3 orders of magnitude less that in the TGG single crystal, other things being equal.

The fourth feature is a Verdet constant 25% larger than the value for TGG, which will allow using in Faraday devices 20% shorter magneto-optical elements, reducing thermally induced effects still more. Large Verdet constant and small value of thermo-optical characteristic $Q$ leads to the fact that magneto-optical figure of merit of TSAG $\sim 30$-times higher as compared to TGG will make TSAG a unique magneto-active material for Faraday devices operating at high average power of laser radiation. The next step of our investigation will be devoted to measurement of temperature dependences of the characteristics of this material during cooling and to creation and investigation of a Faraday isolator based on a TSAG crystal with [001] orientation.

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REFERENCES


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