

Characterization of terbium containing cubic zirconia crystal for high power laser applications

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Abstract

A magneto-active terbium containing cubic zirconia crystal is an attractive material for creating Faraday isolators thanks to a high value of its Verdet constant and low absorption coefficient. The optical anisotropy parameter of this crystal which shows the distribution of orientations of the axes of thermally induced birefringence in a thermally loaded optical element has negative value. It means that there exists a dedicated orientation of crystallographic axes, at which the axes of thermally induced birefringence are arranged in the one direction and thermally induced depolarization may be reduced substantially. For assessing potential of this crystal for high-power applications we have investigated its temperature dependence of the Verdet constant that proved to correspond to the paramagnetic behavior. Thermo-optical characteristics are also of principal importance when operating with highpower laser radiation. We present results of measurements of thermo-optical characteristics Q and P which determines values of polarization and phase distortions of passing laser radiation. Measured values can be used to determine the characteristics of Faraday isolators based terbium containing cubic zirconia crystals in various implementations of their optical schemes for various parameters of laser radiation.

Keywords Magneto-active materials · Thermal effects · Birefringence · Depolarization

1 Introduction

A solid state laser with high peak and high average power is currently an indispensable tool for numerous basic and applied studies. With an increase of available laser radiation power, the need to shield the laser from back reflected radiation increases as well. Faraday isolators (FI) can solve this task but the potential of using them in high-power lasers is restricted by relatively high absorption in magneto-optical elements (MOE) and the related negative thermal effects.

Nonuniform temperature distribution over MOE cross-section gives rise to elastic stresses and to birefringence caused by the photoelastic effect. The passing radiation

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polarization is distorted by the induced birefringence, hence, part of the radiation passes through the isolator on backward passage, thus reducing the isolation ratio. Nonuniform MOE heating also leads to formation of a thermal lens, i.e., to wavefront distortion of the passing radiation. Therefore, magneto-active media that could be effectively used for creating Faraday isolators used in laser systems with high average power are currently actively sought for. Of particular interest in this search are cubic crystals with negative optical anisotropy parameter. The optical anisotropy parameter ξ is a combination of the components of piezo-optical tensor π_{ij} which determines the orientation of the axes of thermally induced birefringence in a thermally loaded optical element (Joiner et al. 1977):

$$\xi = \frac{\pi_{44}}{\pi_{11} - \pi_{12}} \tag{1}$$

It is known that when the optical anisotropy parameter takes on a negative value, there exists a dedicated orientation of the MOE crystallographic axes [[C]], at which the axes of thermally induced birefringence are arranged in one direction, independent of the point of the MOE cross-section. This allows eliminating thermally induced depolarization (Joiner et al. 1977). In a Faraday isolator, circular birefringence is present in addition to thermally induced linear birefringence, so thermally induced depolarization cannot be fully eliminated, but its magnitude may be reduced substantially.

Until recently, nothing was known about the existence of magneto-active media with a negative optical anisotropy parameter, so they could not be considered in the context of creating FIs. However, investigations of a terbium scandium aluminum garget (TSAG) crystal showed that it possesses this property ($\xi_{TSAG} = -101$) in addition to very good thermo-optical characteristics (Mironov and Palashov 2014; Snetkov et al. 2015). Based on this crystal, an FI with an isolation ratio of 35.4 dB at a laser radiation power of 1470 W was created (Yasuhara et al. 2016). That result was achieved without schemes compensating thermally induced depolarization and cryogenic cooling and is, at the present, the best one for FIs operating at room temperature. At the same time, the advantages of negative optical anisotropy parameter cannot be used in full measure for the TSAG crystal because of a very high absolute value of this parameter. Thermally induced depolarization in crystals with high absolute value of parameter ξ increases sharply when the crystals are placed in magnetic field (Yasuhara et al. 2016).

Another promising magneto-active crystal of this type is NTF (Mironov et al. 2015). It has very good thermo-optical characteristics, like a TSAG crystal, and optical anisotropy parameter $\xi_{\text{NTF}} = -0.37$. A combination of these characteristics makes it a perspective medium for Faraday isolators with multi kilowatt permissible operating power.

Another magneto-active crystal with negative optical anisotropy parameter was revealed quite recently. It is a terbium containing cubic zirconia (TCZ) crystal. This material has a crystal lattice structure of fluorite type with the content of the basic components being 60% of Tb₂O₃ and 40% of ZrO₂ and optical anisotropy parameter $\xi_{TCZ} = -0.29$. Also, it possesses relatively low absorption, good spectral characteristics, and a high value of Verdet constant (Mironov and Palashov 2018). So it is highly interesting to investigate its thermo-optical properties at high-power laser radiation level.

In the presented work we measured the magneto-optical figure of merit μ of the TCZ crystal, that determines FI's maximum permissible operating power, and the constant *P*

characterizing thermal lens value. These characteristics were measured by the method which allows using a crystal sample with an orientation different from [001]. We also studied the temperature dependence of the Verdet constant needed for the development of cryogenically cooled isolators.

2 Temperature dependence of the Verdet constant

One of the proven ways to reduce thermally induced depolarization in FIs and to increase substantially their maximum permissible operating power is to cool their magneto-optical elements (Zheleznov et al. 2012).

Cryogenic FIs allow operation at laser radiation power many times higher than the operating power of isolators functioning at room temperature. This becomes possible primarily due to an increase of the Verdet constant of the used magneto-active media on cooling and, hence, a shortening of MOE length. Thus, the temperature dependence of Verdet constant is an important characteristic of a magneto-optical medium in terms of assessing its applicability for high-power Faraday isolators.

The results of measuring this dependence for a TCZ crystal for several wavelengths are presented in Fig. 1.

The measurement results show that the temperature dependence of Verdet constant is well approximated by the dependence ~ 1/T (dashed lines), which corresponds to paramagnetic behavior. Thus, MOE cooling to cryogenic temperatures will result in a ~ 3.5-fold increase of the TCZ Verdet constant and in the corresponding MOE shortening. The related decrease of heat generation will lead to reduction of thermally induced depolarization caused by the photoelastic effect and of thermal lens strength (Zheleznov et al. 2012).

3 Magneto-optical figure of merit

One of important characteristics of a magneto-optical medium used in FIs for isolating high-power laser radiation is the magneto-optical figure of merit.

 $\mu = \frac{V\kappa}{\alpha O}$

Fig. 1 Temperature dependence of the Verdet constant of TCZ at 405 nm (rhombs), 660 nm (triangles), 782 nm (circles) and 1064 nm (squares)



(2)

where α and κ are the absorption and thermal conductivity coefficients, and Q is the thermo-optical constant that is a combination of the components of piezo-optical and elastic compliance tensors and tensor of thermal expansion (Khazanov et al. 1999).

The magneto-optical figure of merit is a convenient criterion for comparing magneto-active media as it is a combination of thermo-optical and magneto-optical characteristics. The higher the magneto-optical figure of merit of the medium, the higher the maximum permissible operating power of the Faraday isolator is (Khazanov et al. 1999). This conclusion follows from the assumption that thermally induced depolarization induced by the photoelastic effect is the principal limiting factor specifying the isolation ratio of the device.

The depolarization degree of the radiation transmitted through an optical element is defined by

$$\gamma = \frac{P_d}{P_0},\tag{3}$$

where P_d is the power of the depolarized radiation component and P_0 is the power of the basic radiation component.

Magneto-optical figure of merit is usually measured based on the analysis of the magnitude of the depolarized component of radiation that has passed through a crystal sample cut in the [001] orientation (Snetkov et al. 2012). The studied TCZ crystal was grown in the orientation greatly differing from [001], so we could not cut from it a sample having a volume sufficient for studying. Therefore, we used the measurement method described in Mironov et al. (2017). This method is also based on analyzing the structure of depolarized radiation component, but in this case a sample of arbitrary known orientation can be used.

The studied TCZ sample was 8 mm long. The crystal was oriented along the [100 37 121] direction that may be characterized by the Euler angles $\tilde{\alpha}=20.4^{\circ}$, $\tilde{\beta}=41.5^{\circ}$ (Koechner and Rice 1971). When the optical element is rotating around its axis, there is a position in which there is no depolarized radiation component in the center of the laser beam, and the distribution pattern of the depolarized radiation takes on a form of the "Maltese cross".

For an optical element cut with arbitrary known orientation, the degree of thermally induced depolarization in the absence of magnetic field for the position in which the distribution pattern of the depolarized radiation has a form of the "Maltese cross" is found in the form (Mironov et al. 2017):

$$\gamma = k_{\alpha\beta\xi} \cdot \frac{H}{8} \left(\frac{L}{\lambda} \frac{\alpha Q}{\kappa} P_0 \right)^2, \tag{4}$$

where *H* is the numerical coefficient determined by the intensity profile of the laser beam (equal to 0.137 for Gaussian distribution), $k_{\alpha\beta\xi}$ is the coefficient the value of which depends on sample orientation (Euler angle values) and on the value of the optical anisotropy parameter:

$$k_{\alpha\beta\xi} = d_2 - \frac{a_1^2 - b_1^2}{a_1^2 + b_1^2} d_3 - \frac{2a_1b_1}{a_1^2 + b_1^2} d_6,$$
(5)

where

$$d_{2} = \frac{1}{2} \left(a_{2}^{2} + 2a_{3}^{2} + b_{3}^{2} \right) (1 - \xi)^{2} + \left(a_{2} + b_{3} \right) (1 - \xi) \xi + \xi^{2},$$

$$d_{3} = \frac{1}{2} \left(a_{2}^{2} - b_{3}^{2} \right) (1 - \xi)^{2} + \left(a_{2} - b_{3} \right) (1 - \xi) \xi,$$

$$d_{6} = a_{3} \left(a_{2} + b_{3} \right) (1 - \xi)^{2} + 2a_{3} (1 - \xi) \xi,$$

$$a_{1} = -\sin^{2} \tilde{\beta} \cdot \left\{ \cos^{2} \tilde{\beta} - \cos^{2} \tilde{\alpha} \cdot \sin^{2} \tilde{\alpha} \cdot \left(\cos^{2} \tilde{\beta} + 1 \right) \right\},$$

$$b_{1} = \frac{1}{4} \sin 4 \tilde{\alpha} \cdot \cos \tilde{\beta} \cdot \sin^{2} \tilde{\beta},$$

$$a_{2} = 1 - \sin^{2} \tilde{\beta} \cdot \cos^{2} \tilde{\beta} - \cos^{2} \tilde{\alpha} \cdot \sin^{2} \tilde{\alpha} \cdot \left(\cos^{2} \tilde{\beta} + 1 \right),$$

$$a_{3} = -\frac{1}{4} \sin 4 \tilde{\alpha} \cdot \cos \tilde{\beta} \cdot \left(\cos^{2} \tilde{\beta} + 1 \right),$$

$$b_{3} = \sin^{2} 2 \tilde{\alpha} \cdot \cos^{2} \tilde{\beta}$$
(6)

We have measured the laser power dependence of the degree of thermally induced depolarization γ in the absence of magnetic field in the position of the studied sample of terbium containing zirconia when the structure of depolarized radiation component has the form of the Maltese cross. The result is presented in Fig. 2.

By substituting into formula (5) values of the Euler angles of the studied sample and of the optical anisotropy parameter of the TCZ crystal $\xi_{TCZ} = -0.29$ found in (Mironov and Palashov 2018) we obtain for our case $k_{\alpha\beta\xi} = 0.056$. It is clear from the results of measurements that the degree of radiation depolarization of about 10^{-3} as a function of power is already well approximated by quadratic dependence (dashed line). The depolarization value of 10^{-3} corresponds to the laser radiation power of 24 W. By substituting these and other necessary data into formula (4) we can find the value of parameter $\alpha Q/\kappa$ of terbium containing zirconia for the radiation wavelength $\lambda = 1075$ nm: $(\alpha Q/\kappa)_{TCZ} = 5.6 \times 10^{-6}$ W⁻¹.

Substituting the obtained value into formula (2) and using the Verdet constant value measured in Mironov and Palashov (2018) that is $V_{TCZ} = 48.5$ rad/T m for the radiation wavelength $\lambda = 1075$ nm yields the value of magneto-optical figure of merit $\mu_{TCZ} = 8.7 \times 10^6$ rad W/(T m) which is rather low. For comparison, the magneto-optical figure of merit of the TGG crystal used for creating a Faraday isolator with a maximum permissible operating power of 650 W (Mironov et al. 2013) is 86 times higher.

Fig. 2 Degree of radiation depolarization passing through the studied TCZ sample measured as a function of its power



Making use of the measurement results of we can estimate the value of maximum permissible operating power of a Faraday isolator based on a TCZ crystal with [[C]] orientation. The [[C]] orientation for the TCZ crystal was found in (Mironov and Palashov 2018). It is characterized by the values of Euler angles $\tilde{\alpha} = 45^{\circ}$, $\tilde{\beta} = 61.7^{\circ}$ and equivalent to them. The expression for the degree of thermally induced depolarization in the Faraday isolator based on a magneto-active crystal of this orientation was derived in Snetkov (2018). Estimates show that, for a TCZ crystal with the use of a magnetic system having a maximum magnetic field value of 2.5 T (Mironov et al. 2013), the isolation ratio of 30 dB will be achieved at laser radiation with a maximum power of ~ 130 W.

The low value of magneto-optical figure of merit is caused by a high value of thermooptical constant Q that is determined by the components of piezo-optical and elastic compliance tensors and tensor of thermal expansion (Khazanov et al. 1999). The value of this thermo-optical constant of the TCZ crystal may be found from the measured parameter ($\alpha Q/\kappa$), as we measured the α/κ ratio by the method described in Volkov et al. (2018): (α/κ)_{TCZ} = 1.5 × 10⁻¹ W⁻¹ K. In this way we obtain Q_{TCZ} = 3.7 × 10⁻⁵ K⁻¹, which is an order of magnitude more than the corresponding value for a TGG crystal (Khazanov et al. 2004).

With such a low value of magneto-optical figure of merit the value of thermally induced depolarization becomes essential, even at low laser radiation power. This is a drawback for creating FIs for high-power lasers. This material, however, may be a useful tool for investigating specific features of thermal effects. For instance, no experimental studies of thermally induced polarization distortions in magneto-active crystals cut in the [[C]] orientation in the presence of magnetic field have been performed so far. This is explained, in particular, by the need to use high-power radiation, as during propagation along this direction depolarization reduces significantly by tens and even hundreds times. With the use of a TCZ crystal such experiments will be possible. They will permit to figure out the behavior of depolarization distortions of radiation propagating in crystals cut in the [[C]] orientation, and determine needed accuracy of aligning of magneto-optical elements so as to meet the requirements to isolators and, as a result, will allow passing from experimental research to commercial production of Faraday devices. Closeness of the value of optical anisotropy parameter of a TCZ crystal to that of the perspective NTF crystal (Mironov et al. 2015) will enable using the obtained results for developing high-power Faraday isolators on its basis.

4 Thermal lens

Another effect of heat generation in a Faraday isolator MOE is wavefront distortion of laser radiation, or thermal lens. For a laser beam with a Gaussian intensity distribution profile, an expression for thermal lens strength in a cubic crystal of arbitrary known orientation, under the assumption that it is determined only by changes in the index of refraction caused by its dependence on temperature and photoelastic effect, was obtained in (Snetkov 2015):

$$\frac{1}{F} = -\frac{1}{4\pi r_0^2} \frac{\alpha}{\kappa} L P_0 \Big[P - Q(\xi - 1) \Big\{ \frac{1}{2} K_{13} + K_{24} \Big\} \Big],\tag{7}$$

where r_0 is beam radius, and *P* is the thermo-optical constant characterizing the value of thermal lens which, similarly to the constant *Q*, is also a combination of piezo-optical and elastic compliance tensors and tensor of thermal expansion (Snetkov et al. 2013). The coefficients K_{13} and K_{24} are determined by the sample orientation and specified by the Euler angles:

$$K_{13} = -\sqrt{a_1^2 + b_1^2 \cdot \cos\left(2\varphi - 4\theta *\right)}$$

$$K_{24} = \frac{1}{4}\sin^2 2\tilde{\alpha} \cdot \sin^2 \tilde{\beta} + \frac{1}{4}\left(\cos^2 \tilde{\alpha} + \sin^4 \tilde{\alpha}\right) \cdot \sin^2 2\tilde{\beta}$$
(8)

where φ is the polar angle of the cylindrical reference frame related to the optical element. The dependence of focal length on angular coordinate of the MOE cross-section point through which a laser beam passes leads to astigmatism of the thermal lens. Averaging over the entire laser beam, i.e. over all possible values of coordinate φ in the $[0,2\pi]$ range, zeros the term related to K_{13} , as a result of which the expression for the averaged focal length F_{av} of the thermal lens takes on the form.

$$\frac{1}{F_{av}} = -\frac{1}{4\pi r_0^2} \frac{\alpha}{\kappa} L P_0 \left[P - Q(\xi - 1) \cdot K_{24} \right]$$
(9)

The coefficient K_{24} determined by (8) changes in the interval [0,1/3]. The value 0 corresponds to the [001] and equivalent orientations, 1/3 corresponds to [111], and 1/4 corresponds to the [110] and equivalent orientations. For the orientation of the sample under study this value is $K_{24} \approx 0.27$.

For determining the focal length of the thermal lens the diameter of the laser beam that has passed through the studied sample was scanned and the coordinate of the beam waist was found. An Yb-fiber laser with the wavelength $\lambda = 1075$ nm and a maximum operating power of 50 W was used in the experiment. The measured dependence of the thermal lens strength for a Gaussian beam having radius $r_0 = 0.83$ mm on the beam power is plotted in Fig. 3.

The obtained data are well approximated by a linear dependence. Using the slope angle of this dependence and substituting into formula (9) the Euler angles values corresponding to the considered sample, the measured values of the coefficient of optical anisotropy parameter ξ and of the parameter $\alpha Q/\kappa$ we find the value of the parameter $\alpha P/\kappa$ of terbium containing zirconia for the wavelength $\lambda = 1075$ nm: $(\alpha P/\kappa)_{TCZ} = 2.33 \times 10^{-5} \text{ W}^{-1}$, from which we can also find the constant *P* for the TCZ crystal: $P_{TCZ} = 1.55 \times 10^{-4} \text{ K}^{-1}$. It is also much higher than for the TGG crystal.

Fig. 3 Thermal lens strength *versus* laser beam power



5 Conclusion

The thermo-optical properties of a terbium containing zirconia (TCZ) crystal determining the value of polarization and phase distortions of high-power laser radiation passing through it have been investigated.

We studied the dependence of the degree of thermally induced depolarization and thermal lens strength on the laser radiation power. The material constants Q and P determining values of these effects for the new material were found. The measurements were performed on a crystal sample with the orientation differing from [001] following the method developed by our team earlier. The thermo-optical constants had the following values: $Q_{\text{TCZ}}=3.7 \times 10^{-5} \text{ K}^{-1}$ and $P_{\text{TCZ}}=1.55 \times 10^{-4} \text{ K}^{-1}$. The measurement of thermally induced depolarization enabled direct measurement of magneto-optical figure of merit which, along with the optical anisotropy parameter, determines the maximum permissible operating power of the Faraday isolator. The magneto-optical figure of merit of the studied crystal proved to be rather low, $\mu_{\text{TCZ}}=8.7 \times 10^6$ rad W/(T m) for the laser radiation wavelength $\lambda = 1075$ nm.

We also investigated the temperature dependence of the Verdet constant of the TCZ crystal. It coincided to a good accuracy with the inversely proportional dependence, which corresponds to paramagnetic behavior.

The low value of magneto-optical figure of merit of the TCZ crystal is the shortcoming for the creation of Faraday isolators, but it allows investigating the features of thermally induced depolarization of the laser radiation that has passed through the MOE elements cut with orientations close to [[C]]. In particular, closeness of the optical anisotropy parameter value of the TCZ crystal to that of the NTF crystal (highly promising for high-power FIs) will enable using the obtained results for the development of Faraday isolators.

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