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Temperature-Dependent Dielectric Characterization of Magneto-Optical Tb₃Sc₂Al₅O₁₂ Crystal Investigated by Terahertz Time-Domain Spectroscopy

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Terbium scandium aluminum garnet (TSAG) crystals have been widely used in magneto-optical systems. We investigate the complex refractive index of the TSAG crystal in the terahertz frequency range using terahertz (THz) time-domain spectroscopy in the temperature range 100–300 K. It is observed that the refractive index and the absorption coefficient increase with the THz frequency. The refractive index increases with the temperature. We measure the temperature coefficient of the refractive index of the TSAG crystal in the frequency range 0.4–1.4 THz. Furthermore, the loss tangent, i.e., the ratio of experimental values of the imaginary and real part of the dielectric permittivity, is found to be almost independent of frequency. TSAG is very promising for applications in THz optoelectronics because it has a high dielectric constant, low loss, and low thermal coefficient of the dielectric constant.

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Ultrafast magnetic phenomena in magneto-optical crystals triggered by femtosecond laser pulses have attracted great interest from both fundamental physics of magnetism and potential applications in spintronics. Stupakiewicz et al. described the ultrafast all-spectral photo-magnetic recording in transparent films of the dielectric cobalt-substituted garnet (YIG-Co). By changing the polarization of the laser pulse, the net magnetization in the garnet can be steered deterministically, thus writing magnetic bits 0 and 1. Furthermore, Mikhaylovskiy et al. and Jin et al. separately reported the ultrafast inverse Faraday effect (IFE) in a paramagnetic terbium gallium garnet crystal Tb₃Ga₅O₁₂ (TGG). Based on the IFE, Gorelov et al. reported terahertz (THz) Cherenkov radiation from a moving magnetic moment produced in TGG. Recently, Subkhangulov et al. found a THz modulation of the magneto-optical Faraday effect in TGG, which is the result of the interaction of two counter-propagation laser pulses via the optical Kerr effect.

In 1997, Riordan et al. used a free-space magneto-optic sampling sensor of TGG to measure the transient magnetic component of a freely propagating THz beam. Recently, Qiu et al. reported a substantial enhancement of THz magnetic near field achieved by a combination of a tapered metallic waveguide and a micro-split-ring resonator. The magnetic near field is directly probed via the TGG crystal. Kurihara et al. proposed and experimentally demonstrated that the THz magnetic near-field-detection sensitivity of magneto-optical sampling with the TGG crystal can be drastically enhanced by cooling the crystal down to cryogenic temperatures.

Rare-earth garnet single crystals with large Faraday rotation angles and low optical absorption loss have been widely used for magneto-optical applications. Recently, a new magneto-optical crystal of terbium scandium aluminum garnet (TSAG) was successfully designed and grown. The Verdet constant of the TSAG crystal is about 20–25% higher than that of the traditionally used TGG crystal. The small thermo-optic constant makes the TSAG crystal used in high-average-power laser operations. Numerous works have been reported on the growth of TSAG crystals with the mechanics and thermal properties, while the temperature dependence of the permittivity of the TSAG crystal at THz frequencies is still inadequately demonstrated. The permittivity is a fundamental property of materials, which is related to the electronic polarizability of ions, local field inside materials, and is important for the THz Cherenkov radiation. In this work, our measurements are carried out using a THz time-domain spectroscopy (THz-TDS), which is a phase-sensitive measurement technique. THz-TDS can provide more information than conventional Fourier-transform infrared spectroscopy, by which a power spectrum is measured. THz-TDS is convenient and valuable to investigate the complex dielectric function of magneto-optical crystals.

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The THz-TDS system is purged with dry N₂ gas to reduce the THz absorption due to residual water vapor in the beam path.

Figure 2(a) is a typical time-domain waveform (blue line) observed for dry N₂ at room temperature, taken as a reference, $E_{\text{ref}}(t)$. Figure 2(b) represents the transmitted THz waveforms through the TSAG crystal with thickness of 3 mm in the time domain, taken as $E_{\text{sample}}(t)$. Using the Fourier transformation, we obtained the spectrum of the reference signal $E_{\text{ref}}(\nu)$ and the sample signal $E_{\text{sample}}(\nu)$. As shown in Fig. 2(c), the spectral range of current setup extends from 0.4 to 1.4 THz. As shown in Figs. 2(b) and 2(c), the phase delay and the attenuation of the THz electric field perpendicular and parallel to the z axis of the sample are almost the same. It is apparent that the complex refractive index within the range of 0.4–1.4 THz is independent of the polarization of THz wave along the in-plane directions. In contrast, the TSAG crystal has a high absolute value of the optical anisotropy parameter $\xi = -101 \pm 10$ at 1076 nm at room temperature. [23, 24]

We assume a plane wave impinging on a layer of the thickness $d$ at normal incidence. The transmittance is given by [25]

$$T(\nu) = \frac{E_{\text{sample}}(\nu)}{E_{\text{ref}}(\nu)} = A(\nu) \exp(-i\Delta\phi)$$

$$= T_1(\nu)T_2(\nu) \cdot \exp \left(-\frac{i(\tilde{n}(\nu) - 1)2\pi vd}{c} \right),$$

(1)
where $\tilde{n}(\nu)$ is the complex refractive index of the sample, $c$ is the speed of light in vacuum, and $T_1(\nu) = \frac{2\tilde{n}(\nu)}{1 + \tilde{n}(\nu)}$ and $T_2(\nu) = \frac{2\tilde{n}(\nu)}{1 + \tilde{n}(\nu)}$ are the Fresnel transmission coefficients of the air sample and of the sample–air interfaces, respectively. We replace the complex refractive index $\tilde{n}(\nu) = n(\nu) - ik(\nu)$ into the Fresnel transmission coefficients and then obtain

$$T(\nu) = \frac{4(n - ik)}{(1 + (n - ik))^2} \cdot \exp(-\alpha \cdot d)$$

$$\cdot \exp \left( -\frac{2\pi\nu(n - 1)}{c} \right),$$

where $\alpha = \frac{2\pi\nu}{c} \kappa$ is the absorption coefficient. Solving Eq. (2) numerically allows us to determine the complex refractive index from the experimental measurements. Due to the moderate attenuation of the THz pulse inside the sample and $\kappa \ll n$, thus $n$ can be directly determined by the phase delay of the $T(\nu)$ as $n(\omega) = \frac{\Delta T \cdot 2\pi\nu}{\Delta \nu}$ and $\alpha(\omega) = \frac{4n(\nu)}{\Pi(\nu)\cdot(1 + \Pi(\nu))}$. [28,29]

Furthermore, the complex permittivity is given by $\varepsilon = \varepsilon_r + i\varepsilon_i$, where $\varepsilon_r = n^2 - \kappa^2$, $\varepsilon_i = 2n\kappa$, and $\varepsilon_r$ is also known as the dielectric constant, indicating that the amount of energy from an external electrical field can be stored in the material. However, $\varepsilon_i$ is a measure of the amount of energy loss from the material, which is mainly attributed to the bound charge and dipole relaxation phenomena.

Figure 2(d) shows the refractive index and absorption coefficient of the TSAG crystal in the frequency range 0.4–1.4 THz, obtained at room temperature, using the above-mentioned procedure. Both the refractive index and the absorption coefficient increase gradually with the frequency. The mean value of the refractive index is nearly a constant of 3.2 within our THz frequency range, which can be used to calculate the THz surface reflectance of the sample. In addition, it should be noted that the quartz window of the cryostat will limit the whole frequency range up to around 1.4 THz, as shown in Figs. 2(a) and 2(c). The signal-to-noise ratio also drops rapidly above 1.4 THz. In further studies, we will attempt to improve the bandwidth of the THz-TDS measurement with high accuracy.

Figure 3(a) shows the THz transmission measurements at the selected temperatures. When the temperature is increased, the time delay of the THz pulse is slightly increased, as shown by the dashed line, which is due to the increase of the group velocity of the THz pulse. In addition, it can also be found that the amplitude of the THz pulses increases with the temperature. It should be mentioned that, due to the thermal expansion of the sample, the thickness of the sample depends on the temperature. However, we cannot determine the thickness of the sample as it was mounted inside the cryostat. However, the thermal expansion coefficient measured for TSAG is around $8.4 \times 10^{-6} \text{K}^{-1}$. Thus we find that a variation of the thickness for the sample in the measured temperature range is less than 2%, which has a negligible effect on the refractive index.

Figures 3(b) and 3(c) show the frequency spectra of the complex refractive index range from 0.4 to 1.4 THz at different temperatures. As shown by the arrow, the values of $\alpha(\nu)$ of TSAG are found to be temperature dependent, which is increasing to higher values with the temperature. As the temperature rises, disorders are created in the lattice and the mobility of the ions increases. The values of $\nu$ in the temperature range from 100 to 300 K for several selected frequencies of 0.8 THz, 1.0 THz and 1.2 THz are shown in the inset of Fig. 3(b). We find that $\nu$ increases slightly with the temperature. Our results suggest that $\nu$ is associated to the thermal motion of dipoles, which cannot orient themselves at low temperatures. [30,31]

$$\tau_n = \frac{1}{n} \frac{dn(T)}{dT},$$

where $n$ is the refractive index measured at room temperature. By fitting with Eq.(3), the $\tau_n$ data are obtained to be $(18.0 \pm 3.2) \times 10^{-6} \text{K}^{-1} @0.8 \text{THz}$, $(16.0 \pm 4.4) \times 10^{-6} \text{K}^{-1} @1.0 \text{THz}$ and $(14.8 \pm 4.8) \times 10^{-6} \text{K}^{-1} @1.2 \text{THz}$, indicating the relative change of $n$ as the temperature is changed. The temperature coefficients $\tau_n$ are plotted as a function of THz frequencies in Fig. 4. In contrast to TiO$_2$ and (Zr,Sn)TiO$_3$, [27] $\tau_n$ of TSAG is positive in the investigated frequency range. In addition, $\tau_n$ of TSAG is much smaller than that of Al$_2$O$_3$, showing a thermal stability for the TSAG crystal.

Finally, Fig. 5(a) shows the real and imaginary parts of the permittivity versus the frequency for different temperatures. The value of $\varepsilon_r$ increases from $\sim$10.0 to $\sim$10.4 with the frequency increasing from 0.4 to 1.4 THz. Similar to the refractive index, $\varepsilon_r$ increases with the temperature, while $\varepsilon_i$ is relatively small and featureless within our present THz range. We have further calculated the loss tangent for the engineering...
application. The loss tangent is defined as the ratio of the imaginary part to the real part of the complex permittivity, \( \tan \delta = \frac{\varepsilon_i}{\varepsilon_r} \), which is used to describe the dielectric loss. As represented in Fig. 5(b), the loss tangent of the TSAG crystal is comparably low, which slightly increases with the temperature. The difference between the tan \( \delta \) values at 300 K and at 100 K is less than 0.005, for almost all frequencies, as shown in Fig. 5(c). The loss tangent of TSAG is less than the value of high-\( \kappa \) materials such as TiO\(_2\),\(^{[27]}\) while it is similar to the value of Al\(_2\)O\(_3\) and orthorhombic perovskite YAl\(_2\)O\(_5\), which is reported to be 0.01–0.02 at frequencies from 1.5 to 3.0 THz.\(^{[32]}\) The weak loss means that TSAG crystals are good candidates for high-permittivity insulation materials in optoelectronic devices in THz frequency ranges.

In summary, we have used time-domain THz spectroscopy to study the temperature-dependent THz dielectric response of paramagnetic TSAG crystals from 0.4 THz to 1.4 THz. The experimental results show that the values of real and imaginary permittivity are found to increase with the frequency and the temperature. The present results demonstrate that TSAG is designed to provide high thermal stability. Our work enables researchers to use the temperature-dependent permittivity of TSAG to design optoelectronic devices in THz frequency more accurately.\(^{[33,34]}\) Finally, our findings also have a prospect for applications of the THz spectroscopy to other magnetic dielectric materials and magnonic metamaterials.\(^{[35]}\)

References

[8] Subkhangulov R R et al 2013 Nat. Photon. 10 111
[33] Kriegel I and Scotognella F 2017 Optik 142 249