Modifying the polarization state of terahertz radiation using anisotropic twin-domains in LaAlO$_3$


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Abstract: Polarization-resolved terahertz (THz) time-domain spectroscopy was utilized to examine the complex refractive index of lanthanum aluminate (LaAlO$_3$), a rhombohedrally distorted perovskite that exhibits crystallographic twin domains. The uniaxial anisotropy of the refractive index was quantified. The ellipticity of THz radiation pulses after transmission through single domains indicated that LaAlO$_3$ can be used as a quarter- or half-wave plate. The effective anisotropy of [001]-oriented LaAlO$_3$ was found to be reduced when the material exhibited multiple, narrow twin domains.

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Fig. 1. (a) Rhombohedral distortion along [111] makes LaAlO₃ uniaxially birefringent. (b) Refractive index ellipsoid with optical axis along [111] (black line). The red line shows the $z = 0$ cross section, repeated in subplot (h). The THz refractive indices for different crystal orientations ([111], [110], and [001]) are shown in (c), (e), and (g), while (d), (f), and (h) show corresponding cross sections through the refractive index ellipsoid and measurement axis orientations. In (c), (e) the solid blue lines show $n_{xx}$, and the dashed lines show $n_{yy}$. In (g) the solid and dashed blue lines show $n_{xx}$ and $n_{yy}$, while the thick red line gives $n_{yy}$.

Fig. 2. (a) Polarization-resolved THz electric field pulses for (a) the reference and (b) the LaAlO₃ sample [001]₄ (500 μm thick). (c) Amplitude spectra of data in (c). (d) Polarization state of THz radiation transmitted through sample A drawn on a unit Poincaré sphere.

This highlights the complexity of analyzing anisotropic media with THz-TDS, which standardly measures only one
polarization channel (e.g. determining just $\hat{n}_{xx}$). Other systems exhibiting anisotropy in the THz range have been reviewed recently [19–21].

To probe further the elliptical THz polarization after sample [001]$_A$, Fig. 2(d) shows a Poincaré sphere representation of the polarization state. Here, $S_1$, $S_2$, and $S_3$ are the normalized Stokes parameters, and the THz radiation varies from vertically polarized (V) at low frequency, through right-hand circular (R) at 1.94 THz to horizontal (H) and left-hand circular (L) at 2.30 THz.

The signed ellipticity $\epsilon(\omega) = \tan(\chi(\omega))$, where $\chi(\omega)$ is the angle of the major axis of the polarization ellipse to the $x$ axis, can be used to quantify the development of the polarization with frequency [23]. In Fig. 3(a) the ellipticity is shown (solid line) for sample [001]$_A$ with the THz electric field along [010], corresponding to the data in Figs. 2(c) and 2(d). Here, $\epsilon = 0$ corresponds to linear polarization, as observed at low and high frequencies and at 2.1 THz. Right-handed ($\epsilon = +1$) and left-handed ($\epsilon = -1$) circular polarization states can also be witnessed. To confirm that this polarization modification arises from optical anisotropy this sample was reduced in thickness from 500 to 387 $\mu$m by mechanical polishing. The resulting ellipticity of the transmitted THz beam (dashed line) is shifted upward in frequency by 30%, commensurate with the reduction in thickness.

The modification of the polarization state of THz radiation by [001]-oriented LaAlO$_3$ depends sensitively upon the presence of twin domains in the crystal, as discussed in the following. Polarization microscopy of the pristine (as-received) sample [001]$_A$ at 550 nm revealed large twins, with domain walls running along [010] and domains up to 600 $\mu$m in width [Fig. 3(b)]. The color scale denotes the optical orientation, the angle of the slow axis from the horizontal. The domains are comparable in width to the diffraction-limited spot size of the THz beam, which samples only one or two domains. Sample [010]$_B$ exhibited smaller twin domains [Fig. 3(d)] of about 30 $\mu$m width. In this case the THz beam probes multiple (>10) domains with orientations differing by 90°. No substantial polarization rotation occurs in the range below 2 THz, as evidenced by the ellipticity $\epsilon$ reported in Fig. 3(c), which is similar with (solid line) and without (dashed line) the sample. Averaging propagation along the fast and slow axes of multiple domains produces an effective medium with reduced anisotropy (lower effective anisotropy).

For a medium with birefringence $\Delta n$ and thickness $d$, the phase delay $\theta$ between the fast and slow axes at angular frequency $\omega$ is given by $\theta = \Delta n \omega d/c$. For a half-wave plate ($\theta = \pi$) at 2.1 THz with thickness 0.5 mm (as for sample [001]$_A$), therefore, we would predict $\Delta n_x = 0.143$. Here, the subscript denotes that the birefringence was obtained from the frequency at which $\theta = \pi$. However the birefringence calculated from the refractive index data in Fig. 1(g) was lower, at $\Delta n = n_{y'y'} - n_{x'x'} = 0.070$, perhaps as a consequence of probing more than one domain. The reduced effective anisotropy of sample [001]$_B$ requires a larger frequency to create $\theta = \pi$: as witnessed in Fig. 3(c), this condition is not satisfied below 3.4 THz.

The THz ellipticity $\epsilon$ reported in Fig. 3(a) does not vary linearly with frequency, as found, for instance, for $x$-cut quartz [23]. If this was the case the material would act as a quarter-wave plate ($\theta = \pi/2$) at half the frequency that it acts as a half-wave plate. The nonlinearity of $\epsilon$ with frequency is the consequence of a frequency-dependent $\Delta n$, which results from the different oscillator frequencies and strengths of the $A_1^u$ and $E_1^u$ modes created by the rhombohedral distortion. Upon lowering the sample temperature below room temperature, enhancing the octahedral rotation [4], the frequency at which $\theta = \pi$ is lowered, as the transmission spectra in Fig. 4(a) indicate. A reduced temperature thus enhances the anisotropy, leading to a greater $\Delta n_x$, as shown in Fig. 4(b).

In summary, polarization-resolved THz-TDS was utilized to examine the distorted perovskite LaAlO$_3$, which was found to be optically anisotropic. The modification of the polarization state of THz radiation was demonstrated using [001] oriented crystals. LaAlO$_3$ may, therefore, be used to fabricate THz quarter- and half-wave plates, although a precise control of twin domain size is required to achieve the desired birefringence. Rather than a linear change in the THz ellipticity with frequency...
(as for $x$-cut quartz waveplates), LaAlO$_3$ exhibits a rapid change in ellipticity as a result of a frequency-dependent birefringence.

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References