High-Resolution THz Transmission and Birefringence Measurements in Ferroelectric $LiNbO_3$

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Abstract—We investigate single-crystal, ferroelectric $LiNbO_3$ as a polarization transformer in the region 400 GHz to 1.0 THz, and extract the THz birefringence and attenuation coefficient from high-resolution photomixer spectrometer data. We also report narrow absorption resonances whose frequency is practically independent of sample thickness and whose strength decreases between 400 GHz and 1.0 THz. These are preliminarily associated with bulk-polaron oscillations

I. Introduction

Optically anisotropic crystals are of growing interest in the THz field for applications as frequency downconverters, parametric oscillators and amplifiers, and birefringent devices. In all of these applications, it is important to know the anisotropic refractive indices and the optical attenuation coefficient in the THz region. By implementing a commercial, ultra-wideband, coherent photomixing spectrometer, we have characterized $LiNbO_3$ as a polarization transformer in the THz region. In the process, we have determined the optical birefringence, $\Delta n = n_e - n_o$, and the optical attenuation coefficient α at spot frequencies between 400 and 1000 GHz.

$LiNbO_3$ has the Perovskite crystal structure with an underlying rhombohedral Bravais lattice. As such, it is inherently non-centrosymmetric and optically birefringent. The material in this study is in the form of undoped, congruent, single-crystal, x-cut, 4-inch diam, $LiNbO_3$ substrates poled along the z axis. A benefit of $LiNbO_3$ compared to other anisotropic crystals (e.g. sapphire) in the THz region is the larger birefringence. In a study using time-domain spectroscopy, it was found that at 300 GHz $\varepsilon_y \cong 45$ and $\varepsilon_z \cong 26$ at 300 GHz, yielding $\Delta n = n_y - n_z \sim 1.6$ [1]. The tradeoff for this Δn is a larger attenuation coefficient, α.

II. Experimental Technique

To measure the THz transmission of the $LiNbO_3$ across a very wide spectral bandwidth, we implemented a coherent photomixing transceiver. This is a commercialized system whose transmit function is based on the difference-frequency generation with two single-frequency distributed-feedback diode (DFB) lasers and a GaAs ultrafast photomixer element under constant-voltage bias. The receive function is based on the same two DFB lasers driving a second photomixer under zero bias and having transimpedance readout. The block diagram of the transceiver is shown in Fig. 1 where the location of the $LiNbO_3$ substrate is shown in the collimated portion of the THz arm. Although higher resolution is achievable, the frequency resolution in the present experiments was set by the laser-temperature-controlled frequency step of of 0.5 GHz. This has been found to be adequate for all of the experiments conducted to date with this instrument on solid-state samples at room temperature.

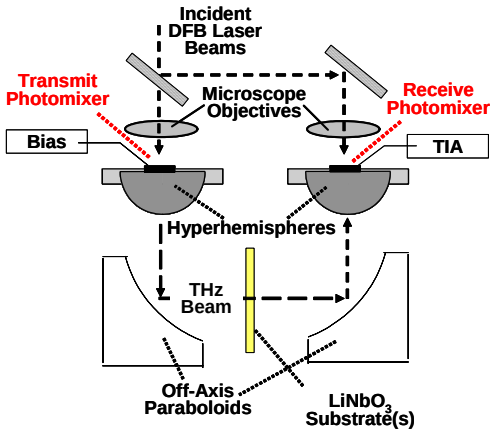

Fig. 1.Experimental front-end of THz coherent photomixing spectrometer used to measure the THz transmission.

The THz transmit beam in the present experiment is incident normally on the $LiNbO_3$ substrate and has predominantly right-hand circular (RHC) polarization. The receiver is designed to accept the same circular polarization, so the $LiNbO_3$ can act as a transformer, changing the transmitted polarization with increasing frequency to linear, then left-hand circular (LHC), then orthogonal linear, and then right-hand circular once again. The frequency at which RHC polarization is converted to LHC should create a large drop in received power because of the polarization mismatch.

III. Results

Fig. 2 shows the experimental results for transmitted power vs frequency between 400 GHz and 1.0 THz for a 1-mm-thick substrate. Fig. 3 shows the experimental results over the same range for two 1-mm-thick substrates placed face-to-face and aligned so that the x, y, and z axes in each crystal were parallel. The instrumental response is shown by the "background" spectrum in Fig. 2 having ~16 dB drop between 400 GHz and 1.0 THz associated primarily with the intrinsic rolloff of the transmit and receive photomixers. Also apparent in both figures are the water lines centered around 556, 752 GHz, and 980 GHz respectively, in decreasing order of strength. These are present because the spectrometer is operated in free space with an optical path length of ~18 in.

Of greater interest are the other narrow attenuation resonances evident in Figs. 2 and 3. The strongest of these are denoted by vertical red arrows in Fig. 3 and separated by roughly 160 GHz in Fig. 2 (single 1-mm-thick substrate). Similar deep minima are evident in Fig. 3 (two 1-mm-thick substrates) and denoted by green vertical arrows, but have a much smaller separation of ~80 GHz. The fact that the separation between the deep minima decreases by a factor of two with doubled sample thickness suggests that these arise

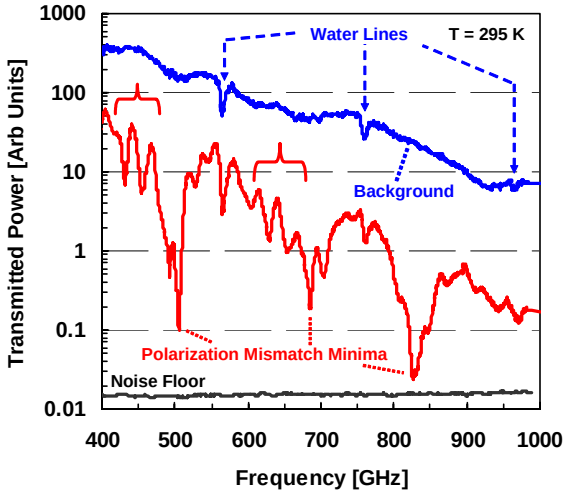

Fig. 1.Transmission through 1.0-mm thick, x-cut $LiNbO_3$ substrate, along with the photomxing spectrometer background spectrum and noise floor. The horizontal curly brackets label regions of absorption signatures attributed only to bulk effects.

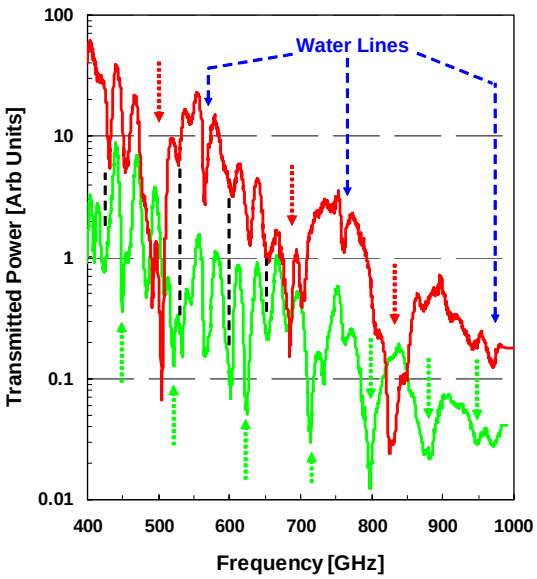

Fig. 2. Transmission through one (red) and two (green) 1.0-mm-thick $LiNbO_3$ substrates butted face-to-face. The vertical red (green) arrows denote the polarization mismatch frequencies for the single (double) substrate(s). The black dashed lines denote transmission minima that appear at the same frequency in both cases.

from polarization transformation.

To confirm this, we note that the separation in frequency between neighboring minima at frequencies f_1 and f_2 is $\{f_2\Delta n_2 - f_1\Delta n_1\} = c/d$, where d is the thickness of the substrate. By doing a linear fit to the minima in each sample between 400 GHz and 1.0 THz, we obtain $\Delta n = 1.5$ at 400 GHz and 1.7 at 1.0 THz. This agrees well with the time-domain results [1].

The data at or near the polarization matching points half-way between the minima allows a calculation of the attenuation constant through the single-pass relation $T \cong (1-R)^2 e^{-\alpha d}$ where R is the air-$LiNbO_3$ power reflection. At the match points just below the 556 and 752 GHz waterlines of Fig. 1, we see $T \cong -10$ dB and -13 dB, respectively. So assuming R = 0.5 (average between the y and z axes), we find $\alpha = 9$ and 16 cm^{-1}, respectively.

The other interesting attenuation resonances in Figs. 2 and 3 are the weaker transmission minima that occur most clearly at the lower frequency end, where they are separated by ~30 GHz, and almost disappear by the upper end. For at least four of these (denoted by vertical dashed black lines in Fig. 3), the center frequencies *do not depend significantly on the $LiNbO_3$ thickness*. We first suspected that they must be caused by some standing wave effect in the system, such as free-space reflections between the transmit or receive photomixers and the front or back faces of the $LiNbO_3$ substrates, respectively. However, the free spectral range for such a standing wave would be $\Delta f = c/2L$, where L is the free-space separation. Using L = 9 inch, this yields $\Delta f = 0.66$ GHz, which is comparable to the instrumental resolution and clearly too small to be relevant here.

IV. Conclusion

Our results suggest that the thickness-independent resonances are associated with a surface or bulk effect in the $LiNbO_3$, such as phonon-polaritons or phonon–polarons. An intriguing possibility is photon-assisted real-space polaron oscillations, which we can only address qualitatively here. Polarons are known to exist in crystalline $LiNbO_3$ at room temperature because of its ionic character and its large difference between the dc (ε_0) and high-ν-limit (ε_∞) dielectric constants, particularly ε_{11} [2]. This allows bound polarons to exist at ≥300 K on certain atomic sites, and to participate in the dc electrical transport by phonon-assisted hopping from site to site [3]. If the binding potential is sufficiently deep, it can support multiple bound energy levels and associated wavefunctions, which can oscillate back-and-forth in real space between neighboring sites similar to what occurs to the N atom in molecular NH_3 but not limited by tunneling transport and not hampered inertially by the mass of the N nucleus. Hence we would expect such oscillations to occur at a higher ν than NH_3 (24 GHz), and comparable to the typical polaronic binding energy of order 1 meV [3] (242 GHz), so the THz region is conceivable. Of course, the entire argument is incumbent on having a long enough polaron lifetime τ to satisfy $\omega\tau >> 1$, which requires $\tau >> 0.3$ ps at ν = 500 GHz. Until recently, such lifetimes were thought unlikely in the solid state at 300 K. However, with the discovery of τ >10 ps for sub-THz vibrational modes in lactose monohydrate [4] and certain other solids, this is not out of the realm of possibility, especially in highly crystalline and pure samples such as the present $LiNb0_3$.

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