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Temperature-dependent birefringence in a (110)-oriented NdGaO₃ single crystal probed by terahertz time-domain spectroscopy

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Abstract

We report a time-domain spectroscopy study of a (110)-oriented NdGaO₃ (NGO) single-crystal specimen in the 0.3–2.6 THz range from room temperature down to 70 K. Measurements were performed for two orthogonal in-plane orientations of the incident electric field in order to resolve the anisotropic optical response of the crystal. From the complex transmission, the refractive index and extinction coefficient were retrieved at each temperature, and the corresponding complex permittivity was analyzed using a Lorentz-based description of the phonon response combined with the Debye model. NGO exhibits a small birefringence in the investigated spectral range, together with a low-loss response, both dependent on temperature and relative orientation of the electric field. Resonance-like absorption features near 1.4 and 2.5 THz are observed, with a polarization-dependent amplitude. These results provide a useful reference dataset for future THz investigations of thin films and heterostructures grown on NGO specimens, especially when specimen anisotropy and temperature-dependent optical properties must be considered.

Keywords:

THz-spectroscopy, NdGaO₃, cryogenic measurements, birefringence, dichroism

1. Introduction

Neodymium gallate (NdGaO_3 , NGO) is an orthorhombic perovskite oxide whose anisotropic crystal structure gives rise to orientation-dependent optical properties, including birefringence and dichroism [1, 2]. In recent years, NGO has attracted interest not only as an optical material in its own right but also as a technologically relevant substrate for the epitaxial growth of complex oxides and related thin films. The use of NGO as a substrate is well documented across several thin-film platforms, including oxide perovskites, nickelates, fluorite-type oxides, and high-temperature superconducting materials [3-9]. This broad applicability makes NGO particularly relevant for THz spectroscopy, since substrate-induced anisotropy, dielectric dispersion, and temperature dependence may all influence the apparent response of the film under investigation. A reliable reference characterization of NGO is therefore valuable not only from the viewpoint of lattice dynamics, but also for the practical analysis of substrate-supported THz measurements [10]. The anisotropic optical response of NGO has previously been investigated in the infrared [11] and at microwave frequencies, including under cryogenic conditions [12], whereas THz studies remain comparatively limited [13-16]. In particular, THz birefringence and dichroism have been reported for selected crystallographic orientations at room temperature [14], while other studies have addressed THz phase and dielectric properties of NGO single crystals [15, 16]. However, a systematic temperature-dependent investigation of the complex dielectric response of (110)-oriented NGO for the two orthogonal in-plane THz polarizations is still lacking. This missing information becomes especially relevant when thin films grown on NGO are studied under cryogenic conditions, where both the substrate transparency and its polarization-dependent refractive index may vary measurably and complicate the extraction of the intrinsic film response.

In this work, we investigate the complex dielectric response of a (110)-oriented NGO single crystal between 0.3 and 2.6 THz over a wide temperature range extending from room temperature down to 70 K. By retrieving the refractive index, extinction coefficient, and complex permittivity as a function of frequency, temperature, and polarization, we provide a reference description of the THz anisotropy of NGO that can support future studies of thin films and heterostructures deposited on this material.

2. Experimental Methods

Transmission measurements were performed using a customized terahertz time-domain spectroscopy (THz-TDS) system designed by Teravil and driven by a 1060 nm EKSPLA femtosecond fiber laser. The optical pulse train was split into two arms with controlled optical power and delivered to two photoconductive antennas: one acting as the emitter and the other as the detector. THz pulses were generated at the biased emitter antenna, while the detector antenna sampled the transmitted THz transient as a function of the delay between the two optical pulses. A motorized delay line in the emitter arm controlled the relative arrival time between the excitation and sampling pulses, thus enabling reconstruction of the THz waveform in the time domain.

The temporal scan window is 120 ps, corresponding to a frequency resolution of approximately 8 GHz. Off-axis parabolic mirrors were used to collimate and focus the beam onto the sample, resulting in a beam waist of about 6 mm at the sample position. In order to probe the polarization dependence of the electrodynamic response, the sample was rotated so that the incident THz electric field is aligned along two orthogonal in-plane directions associated with two distinct crystallographic orientations of the NGO specimen, as discussed below. All measurements were

carried out in a dry nitrogen atmosphere in order to suppress absorption by water vapor along the THz propagation path.

Low-temperature measurements were performed using an optical cryostat (Microstat-He, Oxford Instruments) equipped with low-loss polymethylpentene (TPX) windows. The sample was positioned at the focal point of the THz beam inside the cryostat. Temperature control was achieved using an Oxford Mercury ITC system, and the measurements were collected between room temperature and 70 K during both cooling and warming cycles.

For a given temperature T , the complex transmission function $\tilde{T}(T, \omega) = \tilde{E}_s(T, \omega)/\tilde{E}_r(\omega)$ is obtained, where $\tilde{E}_s(T, \omega)$ and $\tilde{E}_r(\omega)$ are the Fourier transform of the sample and of the reference waveform (acquired in free space and at room temperature), respectively.

NGO is treated as a non-magnetic, wide-gap dielectric medium ($\mu_r \approx 1$). Under these conditions the complex refractive index $\tilde{n} = n + ik$ can be retrieved from the transmission modelled through the Fresnel coefficients and propagation factors. The complex permittivity $\tilde{\epsilon} = \epsilon' + i\epsilon''$ is then obtained from the refractive index through the relation $\tilde{\epsilon} = \tilde{n}^2$ [17].

NGO exhibits a negligible electrical conductivity and a complex permittivity typically governed by the low-frequency dipole relaxation and high-frequency lattice vibrations [2]. For such an insulating material, the frequency-dependent electrodynamic response can be described using the Debye model, which allow to study the relaxation of the bound-charge polarization. Similarly, the incorporation of Lorentz oscillating terms leads to the study of the contribution of optical phonons. Therefore, a combined Debye–Lorentz model can provide a physically meaningful description of the dielectric behavior of insulating materials over the THz spectral range [18, 19], including the directional and thermal dependence of the Debye relaxation terms and of the Lorentzian resonance features.

Accordingly, in the present analysis the permittivity can be phenomenologically described using the complex function:

$$\tilde{\epsilon}(\omega) = \epsilon_{\infty} + \tilde{\epsilon}_{Debye}(\omega) + \tilde{\epsilon}_{Lorentz}(\omega) \quad (1)$$

where

$$\tilde{\epsilon}_{Debye}(\omega) = \frac{\Delta\epsilon}{1-i\omega\tau} \quad (2)$$

and

$$\tilde{\epsilon}_{Lorentz}(\omega) = \sum_j \frac{S_j}{\omega_{0,j}^2 - \omega^2 - i\Gamma_j\omega}. \quad (3)$$

In Eq. (1) ϵ_{∞} is the asymptotic permittivity, whereas in eq. (2) $\Delta\epsilon$ and τ correspond to the dipolar strength and relaxation time, respectively. In eq. (3), S_j , $\omega_{0,j}$ and Γ_j represent the strength, the resonance angular frequency and damping constant of the i^{th} oscillator.

3. Results

The NGO single crystal investigated in this work is a commercially available (110)-oriented specimen with a thickness of 530(2) μm and lateral dimensions of approximately $10 \times 10\text{mm}^2$. Crystallographic characterization was first performed in order to establish the orientation of the crystal with respect to the incident THz electric field. The XRD pattern of NGO is shown in Fig. 1(a) and exhibits a well-defined diffraction peak at $2\theta \approx 46.7^\circ$, corresponding to the (004) reflection, consistent with the orthorhombic crystal structure with space group Pbnm. The schematic drawings in Figs. 1(b) and 1(c) illustrate the geometrical relation between the specimen surface, the crystallographic axes, and the two orthogonal in-plane THz polarizations used throughout the measurements. In the adopted configuration, the specimen surface is parallel to the (110) plane, while rotating the sample by 90° changes the in-plane orientation of the incident

electric field with respect to the crystallographic axes. This geometry provides the basis for probing the anisotropic THz response of the crystal.

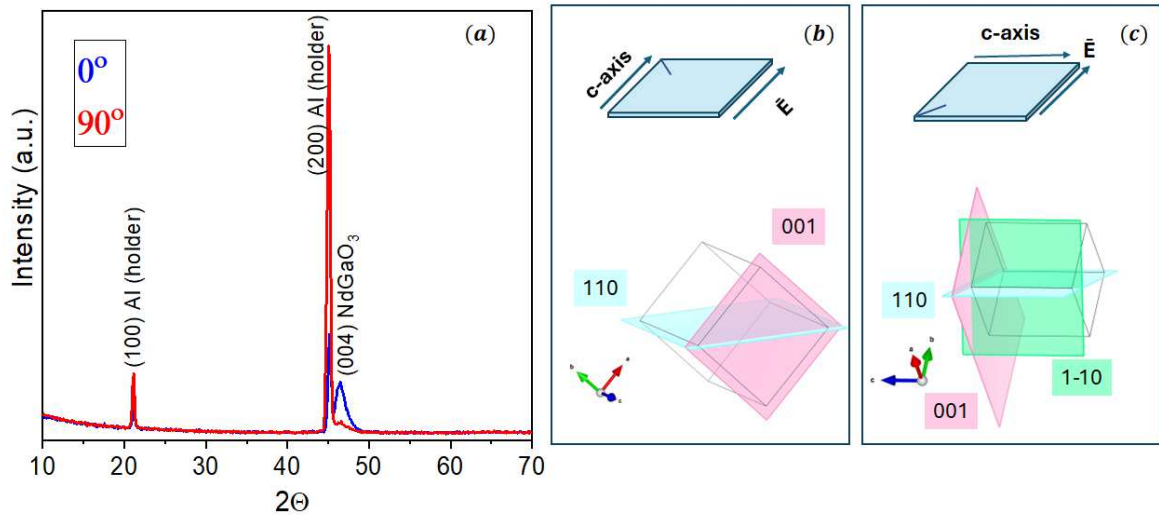


Fig. 1. (a): XRD pattern of the NGO specimen. (b) and (c): schematic views of the crystallographic planes and field orientation for the two orthogonal in-plane polarization configurations, corresponding to $E \parallel c$ and $E \perp c$, respectively.

Representative time-domain waveforms measured at the two boundary temperatures of the investigated range, namely 294 K and 70 K, are shown in Fig. 2 for the two orthogonal field orientations. In both configurations, the transmitted pulse at low temperature is anticipated with respect to the room-temperature signal, indicating a reduction in the refractive index upon cooling. At the same time, the peak-to-peak amplitude increases at low temperature, pointing to a progressive reduction in absorption losses. This behavior is summarized in the inset of Fig. 2, where the normalized peak-to-peak amplitude is plotted as a function of temperature for both polarizations.

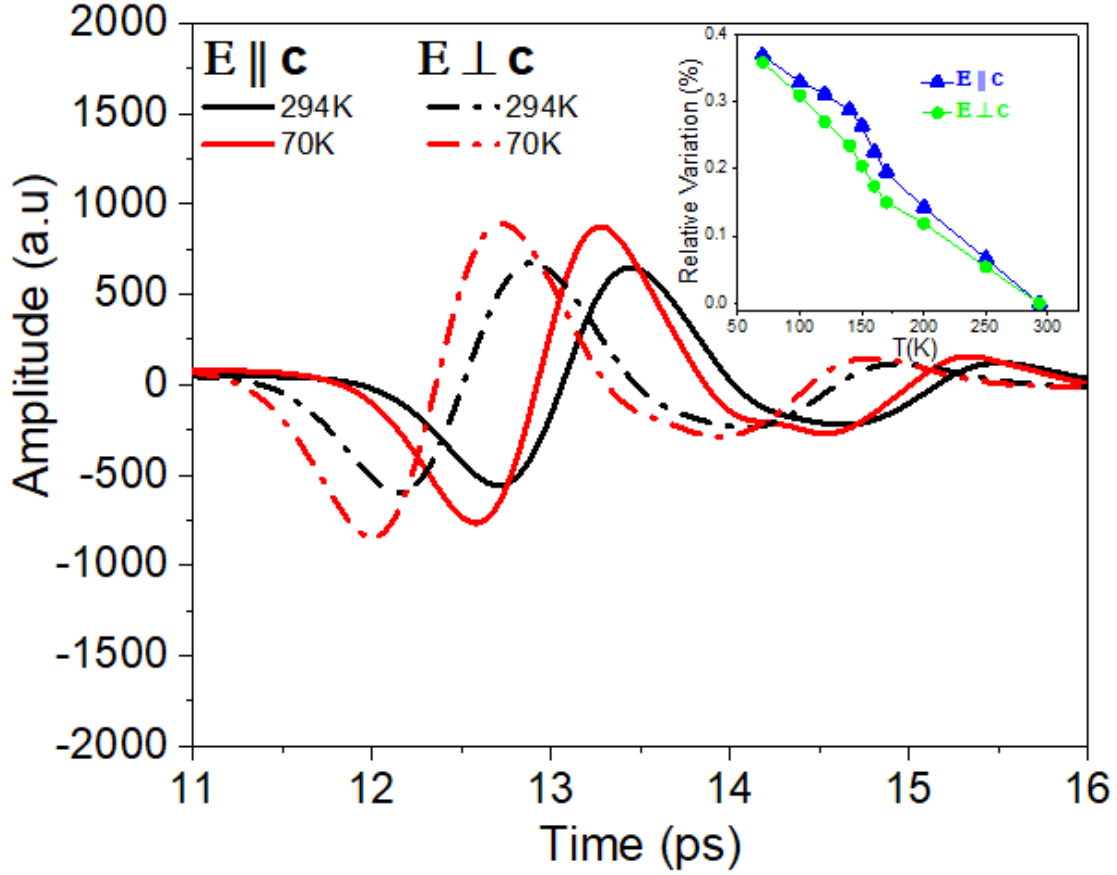


Fig. 2. Time-domain THz electric field measured at 294 K and 70 K for the two orthogonal field orientations $E \parallel c$ and $E \perp c$. The inset shows the normalized peak-to-peak amplitude as a function of temperature for both configurations.

The modulus of the transmission, reported in Fig. 3, confirms the same trend in the frequency domain. Across most of the investigated spectral window, NGO behaves as a weakly absorbing dielectric. In addition, shallow resonance-like features are observed around 1.4 THz and 2.5 THz. The temperature dependence is not identical for the two field orientations, already indicating a measurable anisotropy in the THz optical response.

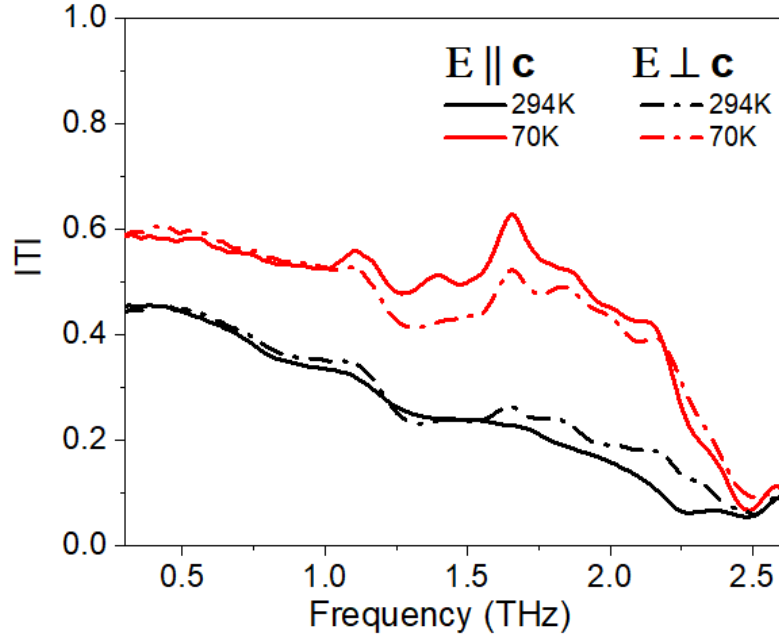


Fig. 3. Modulus of the transmission as a function of frequency at 294 K and 70 K for the two orthogonal field orientations $E \parallel c$ and $E \perp c$.

The refractive index spectra retrieved from the transmission data are shown in Fig. 4. For both crystal orientations, the real part of the refractive index decreases slightly as the temperature is lowered. At 1 THz, the refractive index changes from about 4.78(4) to 4.68(4) along one direction and from 4.67(3) to 4.57(3) along the orthogonal one. These values are broadly consistent with previous THz studies on NGO [15] and confirm the presence of a weak but reproducible birefringence in the investigated spectral range.

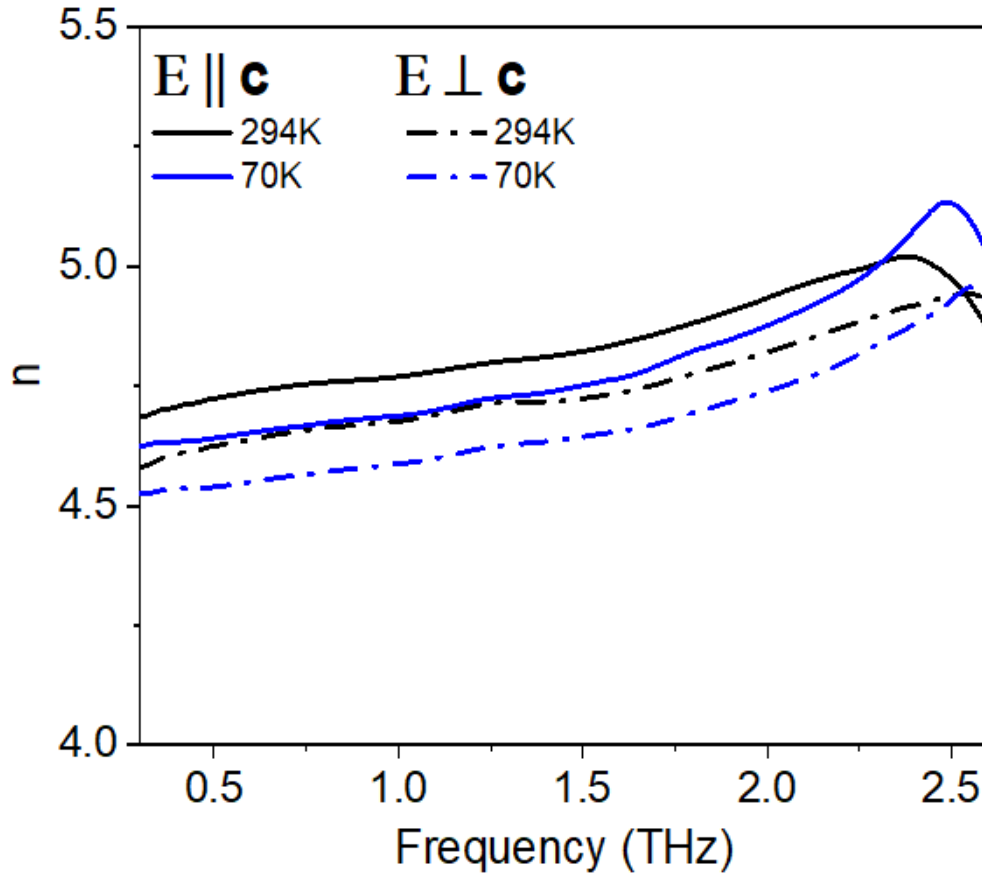


Fig. 4. Refractive index as a function of frequency at different temperatures for the two orthogonal field orientations $E \parallel c$ and $E \perp c$.

To make the thermal evolution more evident, the refractive index at selected frequencies is plotted as a function of temperature in Fig. 5 for the two orthogonal field orientations. In all cases, n decreases monotonically upon cooling, with only modest absolute variations over the explored temperature range. The effect remains systematic across the full frequency interval, whereas the separation between the two polarization configurations persists from room temperature down to 70K.

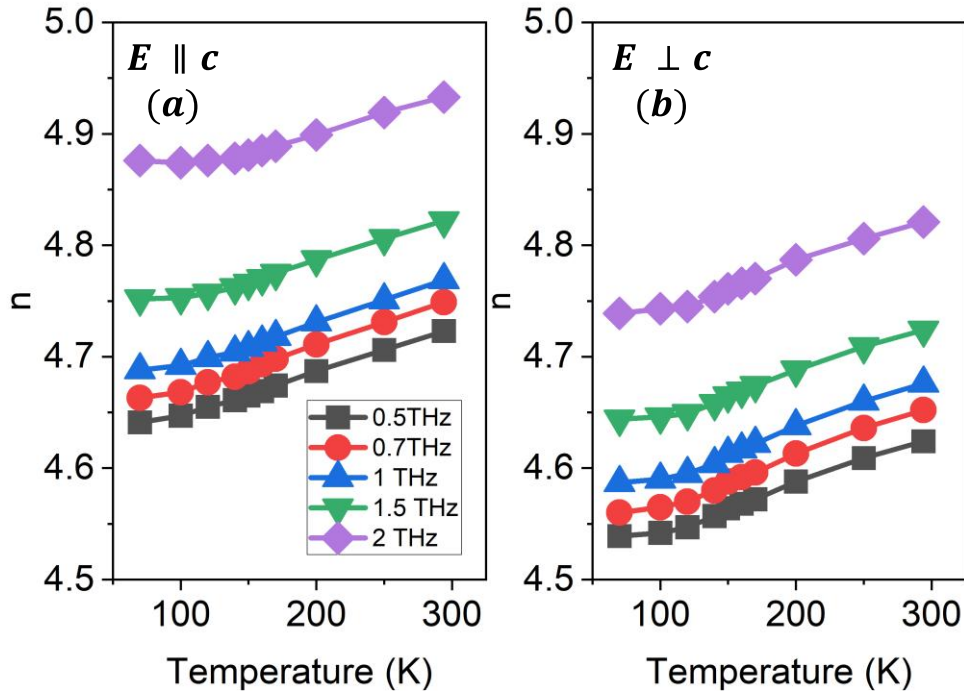


Fig. 5. Temperature dependence of the refractive index at selected frequencies for (a) $E \parallel c$ and (b) $E \perp c$.

The extinction coefficient spectra are reported in Fig. 6. For both polarizations, k remains small throughout the explored frequency range, as expected for an insulating perovskite oxide in the THz domain. The overall loss level decreases on cooling with a polarization-dependent behavior, and resonance-like absorption features are again observed near 1.4 THz and 2.5 THz. This may suggest that the dominant absorption mechanism is phonon-related rather than electronic in origin. This is consistent with the use of a Lorentzian description of the dielectric response, as it will be shown in the following.

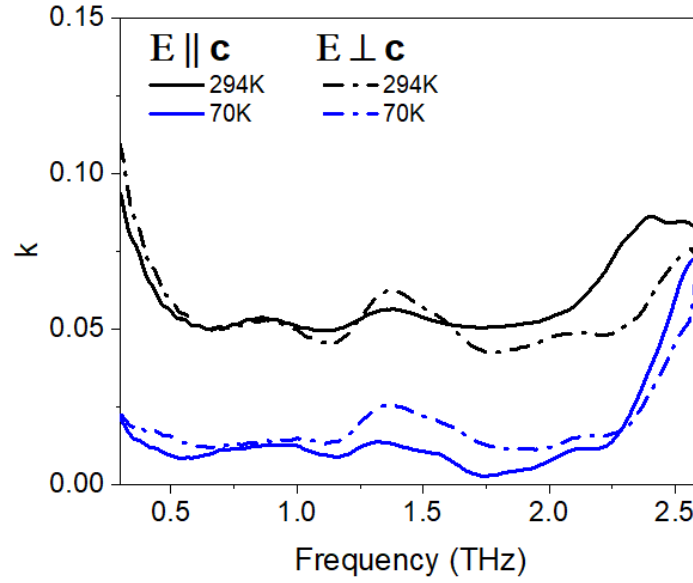


Fig. 6. Extinction coefficient as a function of frequency at different temperatures for the two orthogonal field orientations $E \parallel c$ and $E \perp c$.

The temperature dependence of the extinction coefficient at selected frequencies is shown in Fig. 7. In general, k decreases almost linearly with decreasing temperature, except at the lowest temperatures, where slight saturation effects begin to emerge. The trend in the $E \perp c$ polarization is somewhat more broadened, reasonably signaling an enhanced temperature sensitivity of the losses along the ab plane.

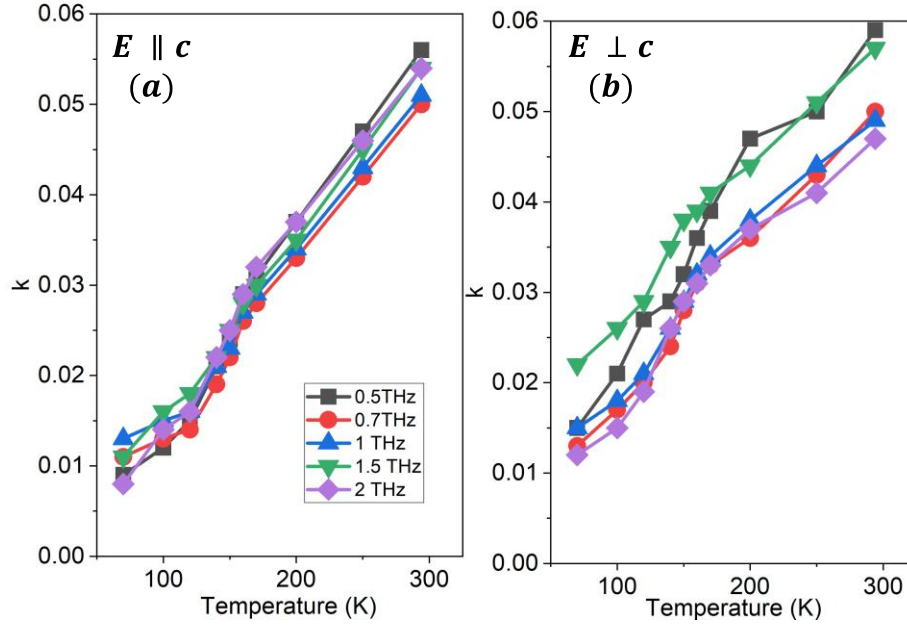


Fig. 7. Temperature dependence of the extinction coefficient at selected frequencies for (a) $E \parallel c$ and (b) $E \perp c$.

Further insight into the electrodynamic response is obtained from the complex permittivity, whose real and imaginary parts are reported in Fig. 8 for the two field orientations and for the two boundary temperatures. The real part ϵ' is only weakly dispersive across most of the investigated range, whereas the imaginary part ϵ'' exhibits clearer resonance-like contributions centred near 1.4 THz and 2.5 THz. Both the amplitude and the linewidth of this loss features show a weak but measurable dependence on temperature and polarization.

The experimental spectra are reproduced reasonably well by the model introduced in Eqs. (1)–(3), and the corresponding fitting parameters are summarized in the Table. I.

Table 1: Fitting parameters obtained from the analysis of the complex permittivity spectra.

	T (K)	$\Delta\epsilon$	ϵ_∞	τ (ps)	ω_{01} (rad/s)	ω_{02} (rad/s)	S_1	S_2	γ_1 (rad/s)	γ_2 (rad/s)
$\bar{E} \parallel \bar{c}$	70	1	22	2.95	8.67×10^{12}	1.65×10^{13}	0.027(1)	0.101(1)	$4.34(8) \times 10^{12}$	$2.48(4) \times 10^{12}$
$\bar{E} \perp \bar{c}$	70	1	22	3	8.8×10^{12}	1.6×10^{13}	0.054(1)	0.159(3)	$2.64(9) \times 10^{12}$	$1.52(4) \times 10^{12}$
$\bar{E} \parallel \bar{c}$	294	3	22	1.9	9.3×10^{12}	1.57×10^{13}	0.208(4)	0.203(5)	$6.51(12) \times 10^{12}$	$4.56(10) \times 10^{12}$
$\bar{E} \perp \bar{c}$	294	4	22	2.25	9.11×10^{12}	1.61×10^{13}	0.157(2)	0.153(4)	$4.56(8) \times 10^{12}$	$4.35(9) \times 10^{12}$

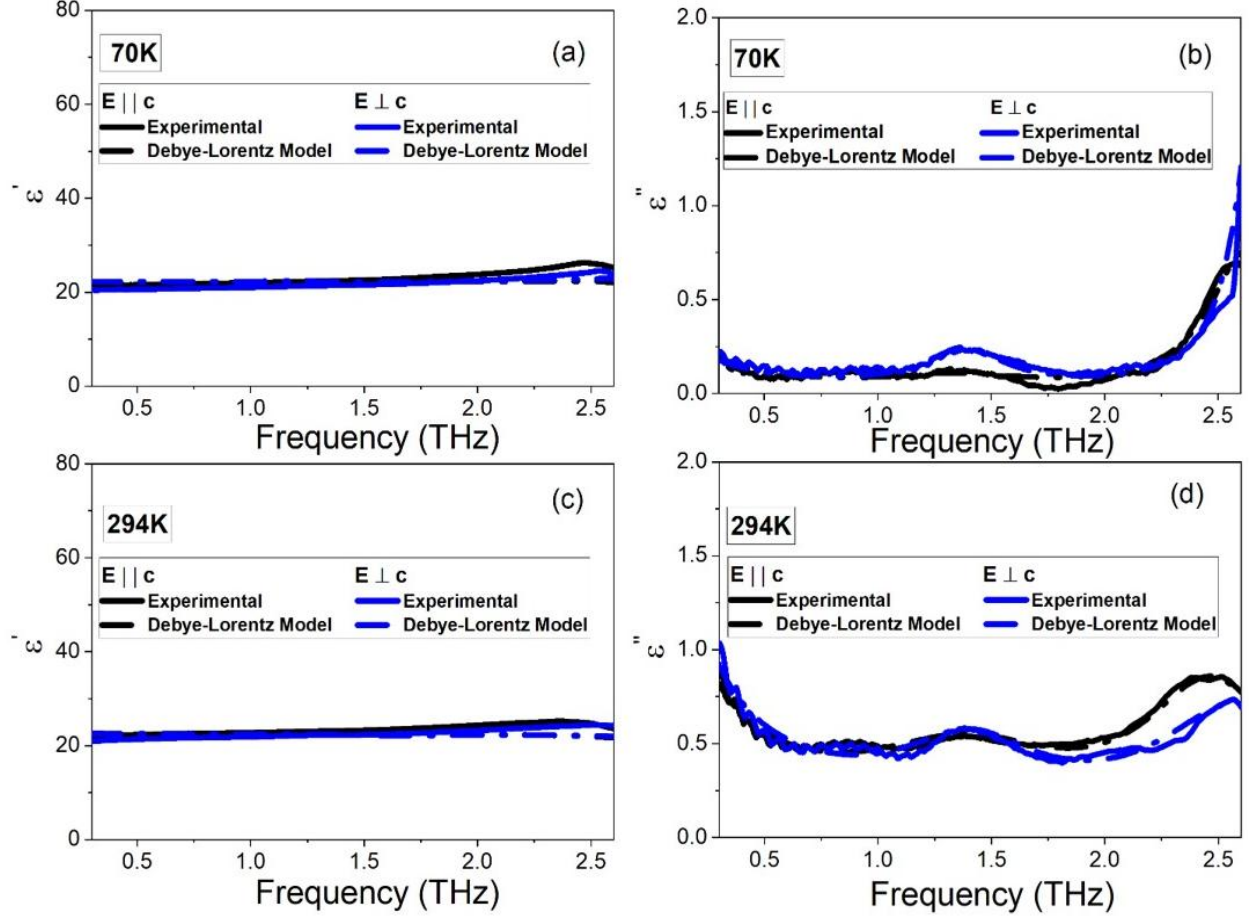


Fig. 8. Real (a, c) and imaginary (b, d) parts of the complex permittivity of NGO as a function of frequency for the two orthogonal field orientations $E \parallel c$ and $E \perp c$ and for the two boundary temperatures. Solid curves represent the experimental data, while dashed curves correspond to the model fits.

4. Discussion

The results show that the THz response of (110)-oriented NGO is weakly anisotropic, yet clearly polarization dependent over the whole investigated temperature range. Both the refractive index

and the extinction coefficient differ for the two orthogonal in-plane orientations of the incident electric field, confirming a measurable birefringent and dichroic response in the 0.3–2.6 THz interval. This is consistent with earlier work on NGO, where anisotropic dielectric behavior has been observed at microwave frequencies and cryogenic temperatures [12], orientation-dependent refractive indices have been reported in the infrared region [11], and birefringence, dichroism, and phonon-related electrodynamic features have been identified in the THz range [14, 16, 20, 21].

A second clear result is the systematic temperature dependence of the optical response. Upon cooling, the refractive index decreases slightly for both polarizations, while the extinction coefficient and the dielectric losses are reduced. This behavior is consistent with a progressive weakening of phonon-related damping at low temperature. Conversely, at higher temperature, stronger lattice anharmonicity is expected to enhance phonon-phonon scattering, thereby increasing absorption and reducing transparency [20, 22, 23].

The resonance-like absorption feature observed near 1.4 THz also admits a plausible physical interpretation. This feature is more pronounced for the $E \perp c$ configuration, indicating a stronger coupling of that polarization when the electric field lies in the ab plane. Indeed, such a resonance has been observed in [24] and ascribed to $B_{2u} + B_{3u}$ phonon modes.

The resonance at 2.5 THz can be recognized as a vibration belonging to the A_{1g} symmetry group [25]. Since this contribution lies emerges at the high-frequency edge of the measured window, only its low-frequency side is fully sampled, and its resonance frequency and damping cannot be determined with the same accuracy as those of the 1.4 THz mode. Nevertheless, it is described by a second Lorentz oscillator and interpreted as a partially resolved higher-frequency lattice excitation. Its presence in both polarization configurations, albeit with different amplitudes, further supports the conclusion that this mode involves the symmetry of the whole crystal cell.

Such an assignment is consistent with the richer spectrum of infrared-active phonon modes expected for orthorhombic NGO [20–22].

In this sense, the adopted two-oscillator description provides a compact phenomenological representation of a phonon-dominated dielectric response, without invoking electronic transport contributions.

The THz absorption in the explored spectral range appears to be largely phonon-driven, with the resonance-like contributions reflecting the oscillation of localized ionic charges at low-energy transverse-optical or quasi-optical phonon frequencies, as phenomenologically described by the Lorentzian terms used in the fit [16, 20]. The stronger absorption observed at room temperature is therefore compatible with increased phonon population, stronger damping, and temperature-induced modifications of the octahedral distortion pattern [24, 26].

The anisotropy of the extinction coefficient and imaginary permittivity can be understood in the same framework. Since the THz field probes two orthogonal in-plane orientations, it couples to different projections of the polar lattice dynamics. As a result, even a relatively weak structural anisotropy is enough to produce measurable differences in the absorption spectra, especially near low-energy phonon resonances. The persistence of these differences over the whole investigated temperature range shows that the birefringent and dichroic character of NGO remains relevant also under cryogenic conditions.

Overall, the present results support a picture in which the NGO behaves in the THz range as a low-loss anisotropic dielectric whose electrodynamic response is governed predominantly by lattice-related mechanisms and their temperature evolution. In this sense, the observed birefringence, dichroism, and polarization-dependent losses should not be regarded as secondary details of the specimen, but rather as intrinsic features that can affect both the interpretation of THz spectra in

film/substrate systems and, potentially, the performance of devices incorporating NGO as a supporting crystalline platform.

5. Conclusion

In summary, we have characterized the temperature-dependent THz response of a (110)-oriented NdGaO₃ single crystal in the 0.3–2.6 THz range for two orthogonal in-plane polarizations of the incident electric field. The measurements show that NGO behaves as a high-index, low-loss anisotropic dielectric, exhibiting a weak but reproducible birefringence and loss anisotropy throughout the investigated temperature interval. Upon cooling, the refractive index decreases slightly, while the extinction coefficient and the corresponding dielectric losses are reduced, leading to an overall increase in transparency. Polarization-dependent resonance-like absorption features are visible near 1.4 THz and 2.5 THz over the explored temperature range, consistent with low-energy phonon contributions.

Overall, these results show that the birefringence and dichroism of NGO, including their temperature dependence, should be taken into account whenever this material is used as a specimen, not only for the THz characterization of thin films and heterostructures, where the specimen response may complicate the extraction of the intrinsic film properties, but also in possible device-oriented applications, where the anisotropic specimen response may influence the performance of the final structure. The present study therefore provides a useful reference framework for future THz investigations of NGO-based film/substrate systems.

Declarations

Ethical Approval: not applicable

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Availability of data and materials: data are available upon reasonable request.

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