

Characterization of Kapton, FeBO₃ and sapphire in the THz region

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Abstract—We measure Kapton, FeBO₃ and sapphire in the terahertz region with a time-domain spectrometer and fit the measurement with Lorentz and Debye oscillators as well as scattering to extract the refractive index and absorption under 5 THz.

I. INTRODUCTION

TERAHERTZ (THz) radiation is currently widely used in diverse applications which are requiring efficient use of the THz radiation, however high losses at interfaces are inherent to the THz range. Kapton foils have proven to be an efficient anti-reflection coating in various environments [1]. Its use allows to increase the coupling efficiency at the exit interface in materials where THz radiation is generated, for example in LiNbO₃ (extraordinary refractive index of ~5 [2]) where the Fresnel losses are close to 40 %, or on the substrate of spin emitters, for example sapphire. It can also be used to increase the efficiency in experiments where THz radiation is used as a pump, such as the excitation of magnons. In all those cases, it is important to have a reliable estimation of the refractive index in the THz range. Still, experiments on thin flexible foils as well as the analysis and interpretation of the results are difficult to perform reliably. As a consequence only few reports mention optical characteristics such as their complex refractive index in the THz range.

II. METHODS

We use a THz time-domain spectrometer (TDS) from Menlo Systems and measure the transmission through the materials of interest of a single-cycle pulse extending up to 4 THz. The analytical calculation of the complex refractive index from the Fourier spectra [3] requires a precise measurement of the sample thickness and is only exact for non-absorbing samples. We instead use Fit@TDS [4] to fit the measurement with different models of the permittivity, for example the Lorentz-Drude model. This allows us to retrieve the refractive index and the absorption while ensuring that the Kramers-Kronig relations are fulfilled, and using the Fabry-Pérot interferences to increase the reliability in the estimation of the thickness.

$$\begin{array}{ccccccc} \varepsilon_{\infty} & \Delta\varepsilon_D & \tau_D \text{ (s)} & \Delta\varepsilon_L & \frac{\omega_L}{2\pi} \text{ (THz)} & \frac{\gamma_L}{2\pi} \text{ (THz)} \end{array}$$

Kapton o.	2.892	420	600	0.165	4.9	2
Kapton e.	3.317	/	/	/	/	/
FeBO ₃ o.	16.02	/	/	/	/	/
Sapphire o.	9.7	/	/	0.053	3.6	0.6

Table 1. Fit parameters for Kapton along the ordinary (o) and extraordinary (e) axes, for FeBO₃ and for sapphire.

We investigate a 125 μm-thick film of Kapton HN polyimide. The measurements were taken for 200 ps window with a resolution of 33 fs, integrating over 800 pulses. We measured the birefringence by rotating the foil in-plane and fitting the refractive index at the perpendicular angles of highest and lowest indices. The second sample is a 35 μm-thick single-crystal of the canted antiferromagnet FeBO₃. Because of the sample size and brittleness, it was loosely mounted on a 3mm-diameter metallic pinhole and measured at normal incidence with integration over 2000 pulses. The last sample is a 650 μm-thick sapphire wafer with a C-plane (0001) surface, measured over a 100 ps window with integration over 800 pulses.

III. RESULTS

Figure 1 shows one of our measurements of Kapton and its fit in time and frequency. The relative permittivity ε is constructed by Lorentz and Debye oscillators according to the equation

$$\varepsilon = \varepsilon_{\infty} + \frac{\Delta\varepsilon_L \omega_L^2}{\omega_L^2 - \omega^2 + i\omega\gamma_L} + \frac{\Delta\varepsilon_D}{1 + i\omega\tau_D}$$

whose parameters are given in Table 1. The high-frequency permittivity is equal to 3.317 along the extraordinary axis and 2.892 along the ordinary axis. As input parameter to the fit, the frequency of the Lorentz oscillator was restricted to match

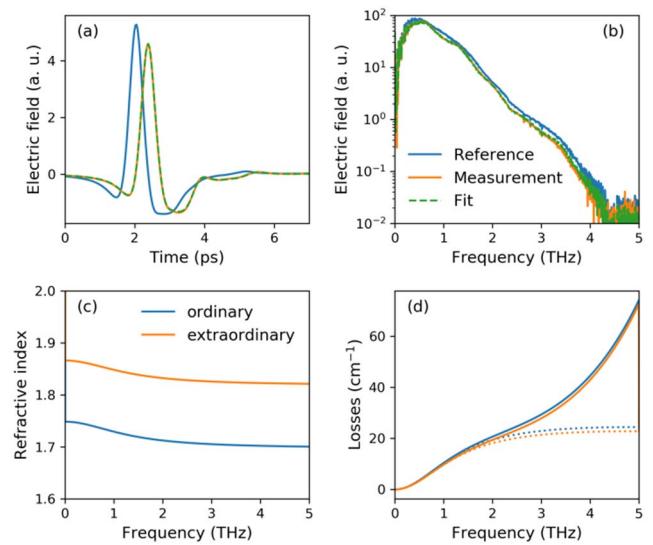


Fig. 1. Fit in time (a) and frequency domains (b) of a THz pulse transmitted through a 125 μm-thick Kapton film. The reference is the pulse transmitted through the same setup without the Kapton film. (c) and (d): retrieved refractive index and losses (absorption and scattering) of Kapton along the ordinary and extraordinary axes. The dotted lines correspond to the absorption only.

the strong peak observed in [5] and [6] at 4.9 THz while the algorithm was free to adjust $\Delta\epsilon_L$ and γ_L . The relaxation time of the Debye oscillator found is on the same order of magnitude as the one measured in [7]. In addition, scattering with a loss coefficient of $1.9 \times 10^{-40} \text{ s}^3$ is responsible for the increase of the absorption above 3 THz. The homogeneity of the sample has been checked with the help of an optical microscope: in all observed foils, a high density of bubbles or inclusions is present, which are generating scattering losses.

The resulting refractive index grows from 1.87 to 1.82 under 5 THz for the extraordinary axis, and from 1.70 to 1.75 for the ordinary axis. The losses are very close for both axes and grow between 10.4 cm^{-1} at 1 THz and 74 cm^{-1} at 5 THz.

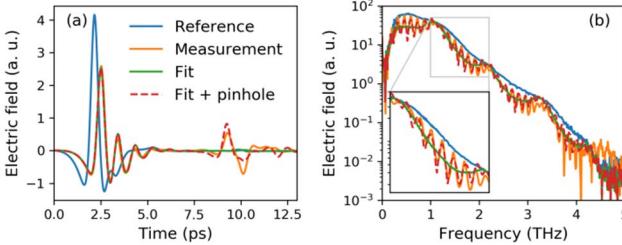


Fig. 2. Fit in time (a) and frequency domains (b) of a THz pulse transmitted through a $35 \mu\text{m}$ -thick FeBO_3 single-crystal mounted on a 3 mm metallic pinhole. The fit (green) only takes into account the transmission through the crystal and the pinhole, while the fit + pinhole (red dashed) adds a component reflected on the pinhole and the crystal separated by 1 mm of air.

The FeBO_3 crystal has its extraordinary optical and magnetic hard axis along the normal of the crystal plane so no birefringence is expected at normal incidence in the absence of magnetic fields. According to [8], the lowest peak in its Raman spectrum is the E_g phonon at 8.27 THz so no oscillator was used for the fit. The result is shown in Figure 2. As the crystal was loosely mounted in front of a metallic pinhole, we attribute the reflection appearing at 9 ps in (a) which causes the interference pattern in the spectrum (b) to a partial back-reflection of the beam on the pinhole and forward-reflection again by the crystal, which allows them to reach the detecting antenna after a delay.

The fitted relative permittivity is given in Table 1. The resulting refractive index is 4.003 in the 0–4 THz range used for the fit and there is no absorption. The losses in the transmitted pulse are due to the reflections at the interfaces of the crystal. In addition to the output from fit@TDS which only fits the main pulse, we calculated the interference of the reflection on the pinhole, adjusting the distance between the crystal and the pinhole to match the observed interference pattern. We are then able to reproduce the measured signal with a distance of 1.01 mm.

The fit of the sapphire wafer is shown in Figure 3. The measurement exhibits a clear Lorentz oscillator at 3.6 THz whose values are given in Table 1. The retrieved refractive index (Fig. 3(c)) and absorption (d) calculated from the Lorentz model are shown alongside analytical estimations following the formulas from [3]. Those formulas do not take multiple internal reflections into account, so the oscillations in the spectrum corresponding to the interference are transferred to the index and absorption.

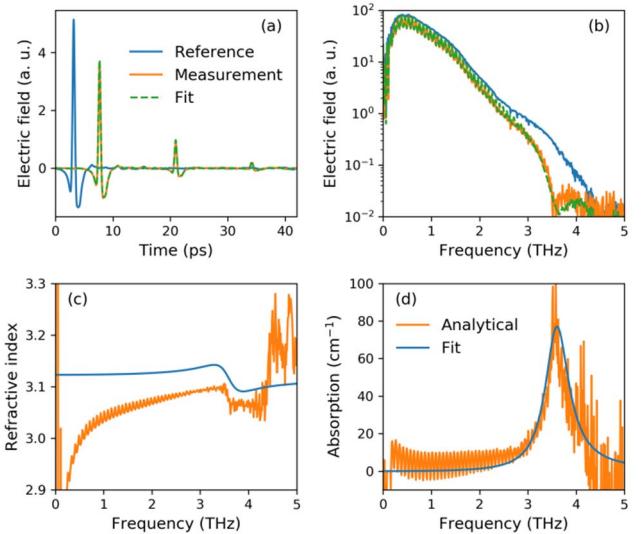


Fig. 3. Fit in time (a) and frequency domains (b) of a THz pulse transmitted through a $640 \mu\text{m}$ -thick C-plane sapphire wafer. (c) and (d): retrieved ordinary refractive index and absorption of sapphire, by analytical analysis of the spectrum and by fit@TDS..

IV. SUMMARY

We investigate the complex refractive index of Kapton, FeBO_3 and sapphire in the 0–4 THz region and use an algorithm to fit the time-domain spectrometry data to a Lorentz-Drude-Debye model, incorporating scattering losses and reflections due to the mounting of the sample. The results for the Kapton are similar to the existing published indices [5–6], which do not extend as far in the low frequency region. The presence of an absorption peak in sapphire at 3.6 THz was, to the extent of our knowledge, not yet reported in the literature.

REFERENCES

- [1]. X. Wu et al., “Temperature dependent refractive index and absorption coefficient of congruent lithium niobate crystals in the terahertz range,” *Opt. Expr.*, vol. 23, pp. 29729–29737, 2015.
- [2]. J. Lau et al., “Millimeter-wave antireflection coating for cryogenic silicon lenses,” *Appl. Opt.*, vol. 45, 3746–3751, 2006.
- [3]. P. U. Jepsen and B. M. Fischer, “Dynamic range in terahertz time-domain transmission and reflection spectroscopy,” *Opt. Lett.*, vol. 30, p. 29, 2005.
- [4]. R. Peretti et al., “THz-TDS Time-Trace Analysis for the Extraction of Material and Metamaterial Parameters,” *IEEE Trans. THz Sci. Technol.*, vol. 9, pp. 136–149, Mar. 2019.
- [5]. P. D. Cunningham et al., “Broadband terahertz characterization of the refractive index and absorption of some important polymeric and organic electro-optic materials,” *Journal of Applied Physics*, vol. 109, pp. 043505–043505–5, 2011.
- [6]. D. R. Smith et al., “Optical constants of far infrared materials. 3: plastics,” *Appl. Opt.*, vol. 14, p. 1335, 1975.
- [7]. A. Sharma, Set al., “Evaluation of dielectric relaxation parameters from TSDC analysis of pristine and ion irradiated kapton-H polyimide,” *Nucl Instrum Methods Phys Res B*, vol. 269, pp. 759–763, 2011.
- [8]. N. Koshizuka et al., “Raman Scattering by Two-Magnon Excitations in FeBO_3 ,” *J. Phys. Soc. Jpn.*, vol. 37, pp. 354–362, 1974.