UNIVERSITY OF PÉCS

Doctoral School of Physics

Nonlinear Optics and Spectroscopy Programme

Linear terahertz spectroscopic investigation of oxide crystals

PhD Thesis

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PÉCS, 2016

1. PRELIMINARIES AND OBJECTS

Oxid crystals have been investigated in the last decades in a wide range of scientific fields due to their promising application possibilities. Previous experiments have shown that lithium niobate (LN) and paratellurite (α-TeO_2) crystals can be used to generate THz pulses as well [1, 2].

Lithium niobate (LN) is a nonlinear optical material used in the THz frequency range due to its extremely large second order nonlinear optical tensor element (d_{33}) [3]. It can be applied for high-energy THz pulse generation [1], nonlinear THz spectroscopy and THz nonlinear optics [4]. For designing experiments the knowledge of absorption coefficients and refractive indices in the THz range is of key importance.

In order to get information about the effect of Mg doping on THz absorption and refraction, one decade ago far-infrared Fourier transform (FIR FT) spectroscopy measurements were performed on a series of undoped and Mg doped (with 1.2, 6.1 and 8.4 mol% Mg concentrations) cLN and undoped and Mg doped (with 0.7, 1.5 and 4.2 mol% Mg concentrations) sLN crystals in the 0.9-4.6 THz frequency range with extraordinary polarization [5]. However the determined absorption coefficient and refractive index values have some uncertainty due to the lensing effect caused by the unwanted curved surfaces of the used crystals.

The object of this work was to refine the results obtained by FIR FT measurements and determine the absorption coefficient and refractive index spectra with ordinary polarization as well.

α-TeO_2 has attracted considerable attention because of its remarkable acousto-optical [6] and electro-optical properties [7]. α-TeO_2 crystallizes in the D_4 space group, with tetragonal symmetry. Single crystals are colorless and transparent in the 0.33-6.5 μm region [8][8]. It is a positive uniaxial material showing optical activity for light propagating along the optical axis [8][8] and has a large second-order nonlinear optical coefficient in the transparent frequency region (values of d_{14} = 0.59 pm/V at 1064 nm) [9].

Sotome and co-workers have only recently reported the generation of THz radiation in α-TeO_2 single crystals irradiated with 800 nm wavelength femtosecond laser pulses [10]. They performed also FIR FT and terahertz time-domain spectroscopy (THz-TDS) measurements to determine the optical properties of the crystal. They have determined the refractive index and absorption coefficient functions of the α-TeO_2 crystal by estimation of the complex dielectric function with the composition of N Lorentz oscillators. The value of N was 8 for ordinary and 4 extraordinary polarizations. Reflectance functions calculated from the Lorentz function were fitted to the measured FIR FT reflectance data for both ordinary and extraordinary polarization.

However there is significant mismatch between the results of the THz-TDS transmission measurements and the curves derived from the Lorentz model fitted to the reflectance curves measured by Sotome et al. In order to fit precisely the measured THz-TDS data in the 0.25 – 2 THz frequency range and also to be consistent with the high frequency FIR FT reflectance data (determined by

Sotome et al.) the modification of some Lorentz parameters was needed.

The object of this work was the verification of the previous measured absorption coefficient and refractive index spectra by THz-TDS measurements and the determination of new Lorentz parameters from which more accurate absorption coefficient and refractive index spectra can be calculated with consideration of the FIR FT and THz-TDS results.

2. METHODS

The index of refraction $n(\nu)$ and absorption coefficient $\alpha(\nu)$ spectra of LN and α-TeO_2 were determined by THz time domain spectroscopy (THz-TDS) in the 0.25 – 2.5 THz and in the 0.25 – 2 THz frequency range respectively. For the examination a

TERA K8 terahertz spectrometer (Menlo Systems) was used. To avoid the absorption of water vapor nitrogen gas was circulated in the spectrometer keeping the humidity around 7% [11].

In the course of the measurements the time dependence of the THz electric field was monitored with and without the sample. From the Fourier transformed electric field functions a complex transfer function was determined in the frequency domain from which the refractive index and absorption coefficient spectra were determined. The determination of the material parameters from the temporal curves was performed by the TeraMat software (Menlo Systems) belonging to the spectrometer. Since the software is unable to take into account the Fabry-Perot effect the time window was shut before the arrival of the first reflected pulse at 34 ps. In order to verify the data obtained by the TeraMat software an evaluation method was improved by adopting an optimizing algorithm [12-14] to our case.

The thickness of the LN and α-TeO_2 samples was measured with micrometer screw with 10 μm accuracy. Because of the well-known birefringence of these crystals [15, 16] measurements were performed with beam polarization both parallel (extraordinary polarization) and perpendicular (ordinary polarization) to the optical axis of the crystal. Since the polarization of the THz antenna source was not perfectly linear, a week beating-like modulation [17, 18] was observed in the spectra. In order to avoid this effect a wire grid polarizer was placed between the source and the sample.

3. NEW SCIENTIFIC ACHIEVEMENTS

I. I determined the absorption coefficient and refractive index spectra of undoped and Mg doped (with 0.7, 1.5 and 4,2 mol % Mg concentrations) stoichiometric lithium niobate crystals with terahertz time-domain spectroscopy in the 0.25 - 2.5 THz frequency range for ordinary and extraordinary polarizations. According to the results of the terahertz time-domain spectroscopy measurements I worked out a procedure to refine the far-infrared Fourier transform spectroscopy results of reference [10] of the PhD thesis [S1, S2].

II. I determined the absorption coefficient and refractive index spectra of undoped and Mg doped (with 0.7, 1.5 and 4,2 mol % Mg concentrations) congruent lithium niobate crystals with terahertz time-domain spectroscopy in the 0.25 - 2.5 THz frequency range for ordinary and extraordinary polarizations. According to the results of the terahertz time-domain spectroscopy measurements I worked out a procedure to refine the far-infrared Fourier transform spectroscopy results of reference [10] of the PhD thesis [S2].

III. I determined the absorption coefficient and refractive index spectra of α-TeO_2 crystals in the 0.25 - 2.5 THz frequency range for ordinary and extraordinary polarizations [S3].

IV. I modelled the complex dielectric constant of α-TeO_2 crystals with the composition of corresponding number of Lorentz oscillators for ordinary and extraordinary polarizations. In contrast to reference [43] of the PhD thesis the model successfully fits with the results of the terahertz time-domain spectroscopy measurements.

4. ARTICLES RELATED TO THE TOPIC OF THIS THESIS

[S1] K. Lengyel, Á. Péter, L. Kovács, G. Corradi, L. Pálfalvi, J. Hebling, **M. Unferdorben**, G. Dravecz, I. Hajdara, Zs. Szaller, K. Polgár, „Growth, defect structure, and THz application of stoichiometric lithium niobate", Appl. Phys. Rev. **2**, 040601 (2015)

[S2] **M. Unferdorben**, Zs. Szaller, I. Hajdara, J. Hebling and L. Pálfalvi, „Measurement of refractive index and absorption coefficient of congruent and stoichiometric lithium niobate in the terahertz range", J. Infrared Milli. Terahz. Waves **36**, 1203 (2015)

[S3] **M. Unferdorben**, A. Buzády, J. Hebling, K. Kiss, I. Hajdara, L. Kovács, Á. Péter, L. Pálfalvi, „Index of refraction and absorption coefficient spectra of paratellurite in the terahertz region", J. Infrared Milli. Terahz. Waves (2016) DOI: 10.1007/s10762-016-0261-1

5. References

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