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Determination of multiphoton absorption coefficients in lithium niobate and lithium tantalate crystals by the Z-scan technique

Ph.D. Thesis

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Supervisor:

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1. Introduction

In the past two decades optical rectification of femtosecond near-infrared pulses in lithium niobate (LN or $LiNbO_3$) has become the most widespread way to generate high energy terahertz (THz) pulses in the low part of the THz spectrum (0.1 – 2 THz) leading to the generation of both high-energy broadband THz pulses [1-3] and narrowband radiation [4-6]. The success of OR can be attributed to several critical factors. One key element is the substantial difference in the indices of refraction between the near-infrared laser light used for pumping and the THz waves. This difference enables the efficient conversion of the pumped energy into THz radiation. The process of quasi-phase matching, achieved through periodic poling of the crystalline material [7], as well as velocity matching through tilted-pulse-front pumping [8] plays an important role in enhancing the conversion efficiency for both narrowband and broadband pulse generation. However, a crucial challenge associated with OR and similar frequency conversion processes is particularly related to the nonlinear absorption phenomenon. This phenomenon has been observed in various materials [9], with a specific focus on multi-photon absorption (MPA) [10,11] and free-carrier absorption (FCA) [11]. LN possesses a relatively large bandgap of around 3.8 eV

[12,13]. These properties are crucial in making LN suitable for nonlinear optical applications. In terms of its nonlinear optical properties, LN exhibits an exceptionally high damage threshold of 204 GW/cm^2 for short 1 ps pulses with a repetition rate of 10 kHz [14]. Similar to LN crystal, lithium tantalate, (LT or $LiTaO_3$), is an isomorph of LN where the Ta^{5+} ion replaces the Nb^{5+} ion [15] with a relatively large band gap of 4.9 eV [16]. It is a material of interest and promising in terms of THz generation through nonlinear optical processes also due to its excellent optical and electronic properties [17], large nonlinear coefficients, and high damage thresholds. Several studies have been conducted on the determination of MPA coefficients in LN and LT crystals using different techniques. However, the Z-scan setup originally introduced by Sheik-Bahae et al [18] is a reliable and accurate method for measuring MPA coefficients in these materials. This technique involves measuring the transmission of a material as a function of the sample's position along the optical axis. A simple transmission measurement was used to determine the 2PA [19] in LN. However, 2PA coefficients and third-order nonlinear refractive index of LT were determined using the Z-scan setup [20], also another study investigated 2PA in undoped sLT crystals [21] and congruent crystals [22]. The Z-scan technique was applied to evaluate the 3PA in LN [23] and used to investigate the scanning nonlinear

absorption in LN over the time regime of small polaron formation [24]. However, 3PA coefficients have not been determined yet in LT. In a previous study, 4PA in LN was estimated based on the saturation of THz pulse generation efficiency with increasing pump intensity [25]. Recently, a simple transmission measurement was used to determine the 4PA coefficient of stoichiometric LN [26]. Notably, 4PA of LN has not yet been measured using the Z-scan technique. These investigations into the nonlinear optical properties of LN and LT are crucial for understanding and optimizing its performance in a range of applications.

2. Objectives and Methods

LN crystals have large effective nonlinear coefficients which are advantageous for the generation of THz pulses by optical rectification. LT, which belongs to the same symmetry class as ferroelectric crystals, is also a noteworthy material for generating THz radiation. However, there exist difficulties in increasing the scale of THz pulse energies and field strengths, such as the free-carrier absorption in the THz range induced by the multiphoton absorption of the pump. The experimental work carried out during this thesis was based on the research lines being conducted at two laboratories: the High-Field Terahertz (THz) Laboratory at the Institute of Physics, Szentágothai Research Centre, and at the Ultrafast High-Intensity Light-Matter Interactions Laboratory at the Institute for Solid State Physics and Optics, Wigner Research Centre. In accordance with them, the general objective consisting of proposing and implementing feasible solutions to the challenges involved in the precise processing of materials with two different laser sources was set out.

More specifically, the following main objectives were proposed: The main aim of this work was to determine multiphoton absorption coefficients in lithium niobate and lithium tantalate crystals by the Z-scan technique consisting of two different femtosecond laser sources. My first goal was to investigate 3PA coefficients in LN crystals. I planned to use the open aperture Z-scan technique which consisted of a Ti:shappire laser (Newport-Spectra Physics) producing 40 fs long pulses at 800 nm and 1 kHz of repetition rate in LN congruent (cLN) and stoichiometric (sLN) crystals at different Mg-doping concentrations for extraordinary polarization. The peak intensity inside the crystals varied between approximately 110 and 550 GW/cm^2. The second objective of the study was the determination of 3PA coefficients in LT crystals. by using the same laser parameters with a similar process in undoped congruent, undoped, and Mg-doped stoichiometric LT crystals (cLT, sLT) for both ordinary and extraordinary polarized light. The peak intensity inside the crystals varied between approximately 120 and 480 GW/cm^2. The third objective was to measure the 4PA coefficients in LN crystals. I planned to use the open aperture Z-scan technique which consisted of an Yb laser (Pharos, Light Conversion) producing 190 fs long pulses at 1030 nm at 1 kHz repetitions rate in LN congruent (cLN) and stoichiometric (sLN) crystals at different Mg-doping concentrations for both ordinary and extraordinary polarization. The peak intensity inside the crystals was 180 GW/cm^2. The fourth objective was to identify the optimum crystal composition for efficient THz generation and other nonlinear optical processes requiring high pump intensities, I made a comparison between the 3PA coefficients in LN and LT crystals, using the same pump source at 800 nm, and a 40 fs pulse duration. This examination focused on extraordinary polarization, assuming nearly identical intensity levels: LT at I=240 GW/cm² and LN at I=255 GW/cm².

3. New Scientific Results

I. I performed open-aperture Z-scan measurements at 800 nm wavelength, 40 fs pulse duration, and 1 kHz of repetition rate in congruent (cLN, cLT) and stoichiometric (sLN, sLT) lithium niobate and lithium tantalate respectively with different concentrations of Mg doping. I carried out the measurements specifically for extraordinary polarization in LN, while also accounting for both ordinary and extraordinary polarizations in LT crystals. The peak intensity inside the crystals varied between approximately 110 and 550 GW/cm^2. I have demonstrated the possible photon absorption coefficients, since LN has a band-gap of about 3.8 eV, the maximum wavelength where 3PA is possible is about $\lambda_{3PA} = 3hc/E_g = 980\ nm$. Beyond that I considered the E_g = 4.9 eV optical band gap of LT as mentioned earlier, I expected the border wavelength between 3PA and 4PA ranges at $\lambda_P = 3hc/E_g = 761\ nm.$ I have concluded that the dominant multi-photon absorption with this pumping wavelength is three-photon absorption (3PA) [S1, S2].

II. Using the pulse propagation model by fitting a theoretical curve to the measured points I have investigated the 3PA coefficients in LN and LT. I have shown that the 3PA coefficients exhibited distinctive variations at different Mg doping concentrations with different intensities. For all the investigated LN crystals composition I have shown that both cLN and sLN displayed minima in their absorption coefficients at a specific Mg doping concentration, corresponding to the point at which photo-refraction was effectively suppressed, and have maximum absorption coefficients at about 290 GW/cm^2. In the case of LT for both cLT and sLT crystals, the transmission curves show significantly deeper values for ordinary polarization compared to extraordinary polarization. Consequently, I have shown that the calculated 3PA coefficients exhibit greater magnitudes under ordinary polarization than under extraordinary polarization, the absorption of LT crystals decreases by increasing intensities from 120 GW/cm^2 to 480 GW/cm^2. I have shown that there is only a slight difference in the absorption between the differently doped stoichiometric samples at low pumping intensities which vanishes above 240 GW/cm^2. Furthermore, the congruent LT has lower multi-photon absorption, hence more attractive for nonlinear applications like THz generation [S1, S2].

III. To investigate the possible photon absorption coefficients in lithium niobate crystals I carried out the open-aperture

Z-scan technique at 1030 nm of central wavelength, femtosecond laser pulse of 190 fs, and 1 kHz of repetition rate on congruent and stoichiometric (cLN, sLN) crystals having different Mg doping concentrations. Both ordinary and extraordinary polarizations were considered. I considered a practical pump intensity level of 180 GW/cm^2 to prioritize the measurement of the absorption coefficients at various polarizations, rather than solely focusing on variations in the coefficient at different intensity levels. I examined the pulse propagation model by fitting a theoretical curve to the measured points. I pointed out that the primary MPA occurring at this pumping wavelength is not caused by genuine 4PA. Nevertheless, effective 4PA values through the fitting, can be used for designing nonlinear optical applications at the intensities used during measurement [S1].

IV. I determined the effective four-photon absorption (4PA) coefficients in LN investigated at 1030 nm. I found that both cLN and sLN exhibited minima in their absorption coefficients at specific Mg doping concentrations (sLN: 0.67% Mg and cLN: 6.0% Mg), corresponding to the point where photo-refraction was effectively suppressed. I have shown that the z-scan curves measured at ordinary polarization indicated significantly larger absorption than those measured for extraordinary polarization. However, the magnesium doping dependence was much stronger than at 800 nm. I also verified the 4PA at 1030 nm exhibited greater nonlinear absorption than the 3PA at 800 nm under the same intensity level. I concluded this finding with the role of the interplay between the second harmonic and defect center-related polarons, shedding light on the underlying mechanisms governing these nonlinear optical phenomena [S1, S2].

V. I compared the 3PA for both LN and LT at 800 nm by supposing identical pump intensity. I have shown that larger 3PA coefficients were measured for LT than for LN, contrary to the larger bandgap of LT in comparison to LN. I concluded that LT has no advantage in comparison to LN in nonlinear optical applications if 800 nm pump pulses are used. As the explanation for this unusual phenomenon, I noted that it is consistent with the observation that in multi-photon (external) photoemission, the emission significantly increases when the excitation photon energy is at the boundary of two different orders of multi-photon photoemission [S1, S2].

List of Publications

Publications related to the dissertation

[S1] **I. Benabdelghani**, G.Tóth, G. Krizsán, G. Bazsó, Z. Szaller, N. Mbithi, P. Rácz, P. Dombi, G. Polónyi, and János Hebling, “Three-photon and four-photon absorption in lithium niobate measured by the Z-scan technique”, Optics Express. 32(5), 7030-7043 (2024).

[S2] **I. Benabdelghani**, G. Bazsó, G.Tóth, P. Rácz, P. Dombi, János Hebling, and G. Polónyi, “Three-photon absorption in lithium tantalate measured by the Z-scan technique”, Optical materials. Submitted, (2024).

Presentations

[E1] **I. Benabdelghani**, V. Gupta, A. Sharma, A. Gupta, G. Á. Polónyi, J. Hebling, J. A. Fülöp, "Terahertz pump transmission measurements in lithium niobate", 10th Jubilee Interdisciplinary Doctoral Conference, 347 297 (2021).

[E2] **I. Benabdelghani**, G. Krizsán, L. Nasi, N. M. Mbithi, J. A. Fülöp, "Measurements of four-photon absorption in lithium niobate", 1st International Conference on Sustainable Energy and Advanced Materials, (2021).

[E3] **I. Benabdelghani**, L. Nasi, G. Tóth, L. Pálfalvi, J. Hebling, G. Krizsán, "Measurement of Four Photon Absorption Coefficient in Lithium Niobate by Z-scan Technique", 9th International Conference on Applications of Femtosecond Lasers in Materials Science, (2022).

[E4] **I. Benabdelghani**, G.Tóth, G. Krizsán, N. Mbithi, G. Bazsó, P. Rácz, P. Dombi, J. Hebling, G. Polónyi, "Three-photon and Four-photon Absorption in Lithium Niobate and

Lithium Tantalate by Z-scan Technique", Lasers and Electro-Optics Europe & European Quantum Electronics Conference (CLEO/Europe-EQEC), (2023).

[E5] G. Polónyi, G. Tóth, N. Mbithi, Z. Tibai, **I. Benabdelghani**, L. Nasi, G. Krizsán, G. Illés, J. Hebling, "Investigation of Terahertz Pulse Generation in Semiconductors Pumped at Long Infrared Wavelengths", Lasers and Electro-Optics Europe & European Quantum Electronics Conference (CLEO/Europe-EQEC), (2023).

Other publications

[S3] N. M. Mbithi, G. Tóth, Z. Tibai, **I. Benabdelghani**, L. Nasi, G. Krizsán, J. Hebling, and G. Polonyi, "Investigation of terahertz pulse generation in semiconductors pumped at long infrared wavelengths," J. Opt. Soc. Am. B, 39(10), 2684-2691 (2022).

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