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Development of high energy THz sources based on contact grating

PhD Thesis

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1 PRELIMINARIES AND OBJECTS

High-intensity and high-field-strength THz pulses are required for nonlinear THz spectroscopy [1], particle manipulation [2], and many other (medical, security) applications [3,4]. Optical rectification (OR) of femtosecond laser pulses in nonlinear crystals is an efficient method for the generation of picosecond THz pulses [5,6]. THz generation by OR in collinear geometry is possible in ZnTe when pumped at 0.8 μm [7]. However, two-photon absorption limits the useful pump intensity [8-10], and thereby the pump-to-THz conversion efficiency [7].

An alternative material for OR is $LiNbO_3$ (LN), since its nonlinear coefficient is very high and at 0.8 μm pumping two-photon absorption is not possible for this material. The refractive index of LN is much higher in the THz range than in the near-IR, so collinear velocity matching is not possible.

However, velocity matching condition can be fulfilled with tilted-pulse-front-excitation (TPFE) [11]. In previous realisations of TPFE the diffraction grating introducing the pulse front tilt and the nonlinear crystal were separate elements, with imaging optics in between. In such setups, reflection losses and imaging errors result in significant limitation of the THz generation efficiency [12]. One can overcome this limitation by placing the grating in contact with the input surface of the nonlinear crystal [13]. This is the so called contact grating (CG) setup. Eliminating in this way the distortions introduced by the imaging also leads to improved THz beam quality. In addition, larger pump beam size and pump energy can be used resulting in larger THz output energy. The diffraction efficiency of LN surface-relief gratings was investigated by Nagashima and Kosuge [14]. According to their calculations, for LN binary gratings with air-filled profile the diffraction efficiency can not be higher than 40%, but it could be increased up to 90% by filling the grating profile with fused silica [14]. However, realisation of such a setup seems to be technically very difficult.

Pumping of ZnTe at longer wavelength can be realized when the TPFE is applied. Significantly higher efficiency can be expected by pumping ZnTe at longer wavelengths [12], since only higher-order multi-photon absorption (MPA) will be effective, allowing for higher pump intensity. This behaviour makes ZnTe and some other semiconductors (such as GaAs and GaP) to potential candidates for high-energy THz pulse generation [12]. Pump wavelengths longer than 0.8 μm require TPFP of ZnTe, but the necessary pulse-front-tilt angle is only about 30° or smaller for pump wavelengths up to about 2.0 μm, which is much smaller than that for LN (63°). The smaller pulse front tilt angle and the corresponding smaller angular dispersion [15], together with the smaller refractive index of

ZnTe are advantageous features which might enable the realization of efficient CG THz sources with large pumped area.

In the first part of this work a setup is proposed where the LN CG profile is filled with refractive index matching liquid (RIML) instead of solid material. The proposed setup is introduced and the diffraction efficiencies calculated for different RIMLs with optimal parameters of the binary grating profiles are presented. A practical setup where both the RIML front surface and LN out-coupling surface are tilted in order to optimise the in- and out-coupling is suggested and analysed.

In the second part of this work a numerical study is presented on adopting the CG approach to OR in semiconductors (such as ZnTe, GaAs and GaP). Detailed CG design parameters will be given and the CG performance will be analysed. The results of our numerical simulations will be presented with emphasis on diffraction efficiency and optimal device parameters for different grating profiles. The expected performance in THz generation and possible limiting factors will be discussed.

2 METHODS

In a TPFE setup the pumping efficiency is strongly influenced by the diffraction efficiency of the grating. Transmission gratings with high diffraction efficiency can be realised with binary and sinusoidal structures [16] fabricated on dielectric surfaces [17]. Such gratings can be created on LN surface by reactive ion etching [18]. Appropriate relief grating structures on semiconductors (for example ZnTe) can be manufactured for example by laser ablation [19] or ion etching [20]. The former may be better suited for sinusoidal, while the latter for binary CGs. The diffraction efficiency of the setup has been determined by using the GSolver software (Grating Solver Development Company). The software uses the rigorous coupled-wave analysis (RCWA) method [21]. This method is able to solve Maxwell's equations numerically for a homogeneous and periodic structure.

The out-coupling surface of LN and ZnTe should be tilted by appropriate angle in order to minimise Fresnel losses for the THz output. The propagation distances inside LN and ZnTe for different parts of the diffracted pump beam –which also has angular dispersion– are different. The angular dispersion results in increased group velocity dispersion which should be avoided. In case of LN CG tilting the RIML input surface is used to make possible achieving the required incidence angle on the grating and to minimise Fresnel losses. Because of the tilting of the input surface, the outer part of the pump beam travels a long distance in the RIML. Therefore, I analysed the ultrashort pulse propagation in LN, ZnTe and RIML.

3 NEW SCIENTIFIC ACHIEVEMENTS

I. Based on model calculations I showed that higher than 90% diffraction efficiency can be reached for LN surface relief grating (CG) by filling the grating profile with appropriate RIML. Examining a lot of RIML's with a wide range of refractive index I have concluded that diffraction efficiency maximum can be realised by filling up binary grating profile fabricated on LN crystal surface with a matching liquid where its refractive index is equivalent of the crown glass (BK7). [S1]

II. I designed a grating structure that can be fabricated with current manufacturing technologies where the tilting of RIML and LN surfaces minimises the Fresnel loss and avoids angular dispersion. The refractive index of the applied RIML is equivalent of the crown glass, the optimal profile parameters are as follows: grating period of $d = 0.35\,\mu\text{m}$, filling factor of

$f = 0.4$ and relative depth of $h = 0.5$ μm. For these parameters the diffraction efficiency can be as high as 99% for the -1st diffraction order, and the velocity matching between the pump pulse and the generated THz wave is fulfilled at the same time. These conditions ensure an efficient near single cycle THz pulse generation above the mJ energy level. [S1]

III. The imperfection of the ion etching during the manufacturing can influence the final grating parameters; which may differ from the optimal value that I proposed. I determined tolerance ranges of different parameters for realisation such a setup where a 10% loss in diffraction efficiency is acceptable. Maximum reachable diffraction efficiency is most significantly influenced among the inspected parameters by the wall steepness of the binary grating. Therefore, great care has to be taken on minimisation these angles during realisation. [S1]

IV. According to my results a highly efficient THz source can be realised with a ZnTe based CG setup. The results of numerical calculations show that diffraction efficiencies can be as high as 80% and 90% with sinusoidal and binary CG profiles, respectively. At 1.7 µm pumping wavelength angle of incidence are 35° and 17.5° in case of optimal parameters. Based on the new model the semiconductor CG can be pumped at 1.4 – 1.7 µm pump wavelength range. The selected wavelength range enables to increase the useful pump intensity significantly beyond that possible at the more commonly used 0.8 µm wavelength, since at longer wavelengths only higher than two-photon absorption is present. [S2]

V. My calculations show that maximum achievable diffraction efficiency exceeds 75% even for normal incidence both for binary and sinusoidal gratings. A ZnTe CG pumped with normal incidence holds promise to become a particularly compact, robust and alignment-free source of high-energy few-cycle THz pulses. In this setup effective pumping beam diameter is limited only by the achievable crystal dimensions. Consequently, high pump energies can be used without significant temporal and spatial distortion of the generated pulse. [S2]

VI. The realisation of the CG technology can also be feasible with other types of semiconductors such as GaAs and GaP. To show the effectiveness of the technology I carried out detailed numerical calculations on these semiconductors too. The results show that the developed method for CG analysis is effective and simple to implement. [S2]

4 ARTICLES RELATED TO THE TOPIC OF THIS THESIS

[S1] **Z. Ollmann**, JA Fülöp,J Hebling,G Almási „Design of a high-energy terahertz pulse source based on ZnTe contact grating" OPTICS COMMUNICATIONS 315: pp. 159-163. (2014)

[S2] **Z. Ollmann**, J. Hebling, G. Almási „Design of a contact grating setup for mJ-energy THz pulse generation by optical rectification" APPLIED PHYSICS B - LASERS AND OPTICS 108:(4) pp. 821-826. (2012)

5 OTHER ARTICLES

[S3] JA Fülöp, **Z. Ollmann**, Cs Lombosi, C Skrobol, S Klingebiel, L Pálfalvi, F Krausz, S Karsch, J Hebling „Efficient generation of THz pulses with 0.4 mJ energy" OPTICS EXPRESS 22:(17) pp. 20155-20163. (2014)

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