High energy Yb:CaF$_2$ femtosecond laser for efficient terahertz generation in lithium niobate

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ABSTRACT

We investigated intense Terahertz generation in lithium niobate pumped by a powerful Yb:CaF$_2$ laser at room temperature and 25 K. This unique amplifier system delivers transform-limited pulses of variable duration (0.38-0.65 ps) with pulse energies up to 12 mJ at a central wavelength of 1030 nm. From theoretical investigations it is expected that those laser parameters are excellently suited for efficient THz generation. In this study we present experimental results on both the conversion efficiency and the THz spectral shape for a series of transform-limited pump pulse durations and crystal temperatures and discuss the optimum pump parameters for most efficient THz generation.

Keywords: High-energy THz, LiNbO3, Optical rectification, ytterbium lasers

1. INTRODUCTION

High-energy ultrashort terahertz (THz) pulses in the THz gap (frequency range at 0.1-10 THz and photon energy between 0.4 and 40 meV) allow direct access to several resonant and nonresonant excitations [1]. The key advantage of terahertz pulses with respect to visible and near-infrared lasers is that collective modes in complex materials, such as phonons, magnons and electromagnons, can be selectively excited without heat deposition. Moreover intense THz sources are proposed for new investigations such as non-resonant excitations of magnetization dynamics [2], THz-assisted attosecond pulse generation [3], initiation of catalytic reactions [4], and optical THz undulators [5], which would benefit from field strength greater than the ones currently provided from table-top sources.

Among tabletop sources, optical rectification (OR) of intense laser pulses is a prominent approach for the generation of high-energy terahertz pulses [6-11]. OR is a second order process, which is based on the difference frequency generation between components of the same femtosecond laser pulse. The efficiency of the OR depends on the nonlinear material properties such as absorption coefficient, nonlinearity, phase matching and on the characteristic of the pump laser, namely fluence, pulse duration, spectrum and wavelength. Different materials permit to generate THz over different spectral regions. While organic salt crystals offer efficient generation between 1-10 THz, the inorganic LiNbO$_3$ (LN) with tilted front pump phase matching demonstrated high-energy pulse generation below 1 THz [12, 13]. By using LN at room temperature (RT) and pump pulses with a typical duration of about 100 fs pump-to-THz conversion efficiencies up to 10^{-3} could be demonstrated [9]. It was proposed that OR in LN could be significantly improved by optimizing the Fourier-limited (FL) pump pulse duration

Moreover cooling the LN to cryogenic temperature (CT) results in lower THz absorption and higher pulse energy [14, 15]. The absorption at THz frequency in fact, hinder high-efficient OR at room temperature especially at frequencies around 1 THz. The main source of absorption is identified in the transverse optical phonon modes of the LN [15]. The phonon coupling is weaker at low temperatures, resulting in lower THz absorption and higher efficiency for OR.

Lasers with spectrum peaked around 1 μm, delivering transform-limited pulses of about 600 fs FWHM, are predicted to be ideal for driving OR in LN [16, 17]. The laser-to-THz conversion efficiency decreases in fact, for shorter FL pump due to the strong group velocity dispersion in LN as well as for longer pulses where the low intensity laser gives rise to lower OR efficiency [17].

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Recent experimental studies confirmed these predictions. The highest so far THz pulse energy (125 µJ) from OR in LN could be reached with longer duration (though non-optimal) pump pulses [10]. High conversion efficiency (3.8%) was demonstrated in congruent LN at CT using close-to-optimal pump pulse duration [11].

In this paper we present a versatile and powerful sub-picosecond Yb:CaF$_2$ laser system which is employed to explore the optimal pump parameters in efficient and broadband OR [18]. The THz generation has been performed in stoichiometric LN, which is used at room temperature (RT) as well as at cryogenic temperature (CT, 25 K). In order to find the maximum conversion efficiency we investigated OR with variable, but always Fourier limited pulse durations. We report detailed characterization of the conversion efficiency as well as the THz spectra as function of pump pulse width and LN temperature. To our best knowledge this is the first systematic study on the effects of pump pulse width and LN temperature for the THz generation. While emphasis has been put solely on enhanced efficiency in the past, the potential changes of the THz spectral shape have not been examined.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. A femtosecond laser is used to generate THz pulses by OR in LN. The laser system is based on chirped-pulse amplification (CPA) and is composed by a compact ytterbium oscillator, a grating-based stretcher-compressor and a regenerative Yb:CaF$_2$ amplifier.

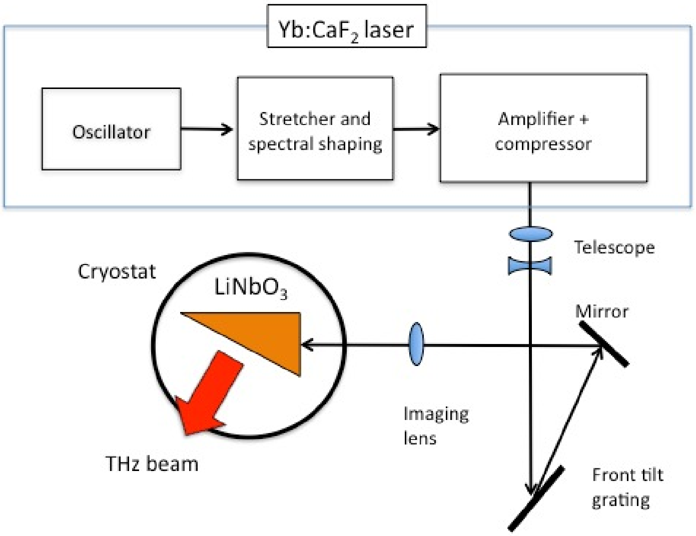

Figure 1 Experimental setup: an high energy Yb:CaF2 is used for THz generation by optical rectification in litium niobate. The laser delivers sub-ps, 15 mJ pulses with variable Fourier limit duration. After compression the beam size is reduced by a factor 3 in a telescope. The pulse front is tilted by diffraction grating to achieve phase matching THz generation in the LN crystal. The crystal is installed in a cryostat.

The broadband oscillator delivers sub-ps pulses of 1 nJ energy at a repetition rate of 54 MHz. The laser pulses are successively stretched up to 500 ps prior to the injection into the regenerative amplifier. Here the 4-mm long amplifier Yb:CaF$_2$ crystal kept at room temperature is pumped by two 80-W CW diode modules which emits at 981 nm. This compact single-stage amplifier permits to boost the pulse energy up to 30 mJ at a repetition rate of 10 Hz. The amplified pulses are then recompressed to transform limit duration in transmissive grating compressor which has an overall energy throughput of 50%. As particular feature, the laser design allows seeding the amplifier in narrowband as well as broadband mode without risk of damage while maintaining the 15 mJ pulse energy after compression. In Fig. 2 (a) different spectral widths and (b) the corresponding autocorrelation of compressed pulses are shown. The Gaussian fit shown in Fig. 2 (b) indicates that FL pulses between 380 and 650 fs FWHM can be achieved by reducing the spectral width. The corresponding spectra are shown in Fig 2 a. The spectral manipulation is implemented placing a variable slit in the Fourier plane inside the stretcher.

To increase the fluence for optimal THz generation the compressed laser beam is down-collimated by 3:1 telescope. The pump pulse front is tilted using a reflective diffraction grating to achieve the phase matched OR [10]. To recombine the different spectral components the beam at the grating is imaged on the LN crystal using a spherical lens of 250 mm focal length. In the experiment constant pulse energy up to 12 mJ over a 5 mm beam diameter is used to pump the LN crystal.

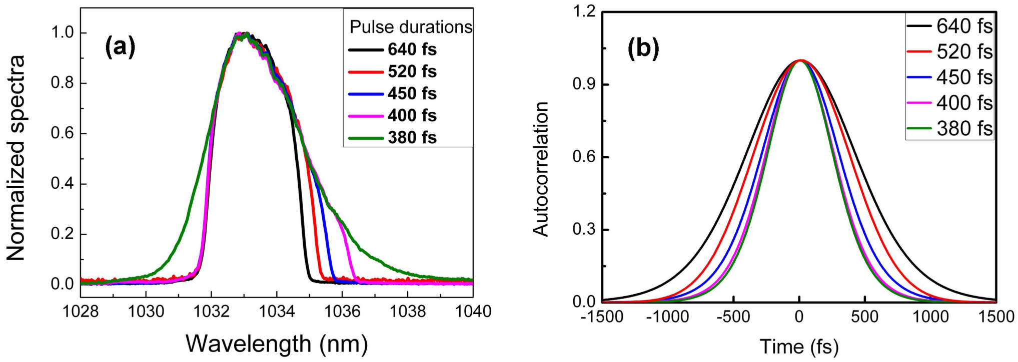

Figure 2. The laser system can deliver Fourier limit pulse with duration variable between 380 to 640 fs FWHM. A spectral mask, installed in the laser stretcher is used to filter out part of the oscillator spectrum prior to the amplification. The resulting spectra and the corresponding autocorrelation traces are shown.

The THz generation is carried out in stoichiometric $LiNbO_3$ with 0.6% MgO doping. The LN input surface is 8 by 16 mm while the crystal is cut at 63°. The lithium niobate is installed in a cryostat which allow to vary the temperature from room temperature down to 25 K. At this temperature the phase matching conditions are expected to slightly change due to 5% lower THz index of refraction at CT [13]. However adjusting of 1.3 deg the pump pulse front tilt can preserve the phase matching condition at 25 K. In the experiment, the position of the imaging lens is adjusted to achieve the optimal pulse front tilt over the full temperature range. The pump energy deposited in the LN has been calculated to produce a peak temperature of 29 deg. This modest temperature variation is not expected to alter the THz absorption coefficient and the overall OR efficiency [15]. The THz pulse is then characterized in energy by means of a calibrated $LiTaO_3$ pyroelectric detector with 3kV/W responsivity (Microtech instrument). Spectral measurements are performed with an in air THz Michelson interferometer (Fourier-transform spectroscopy) equipped with THz sensitive detector.

3. EXPERIMENTAL RESULTS

In Fig. 3 the THz energy normalized to the pump peak power is plotted as function of the laser pulse duration.

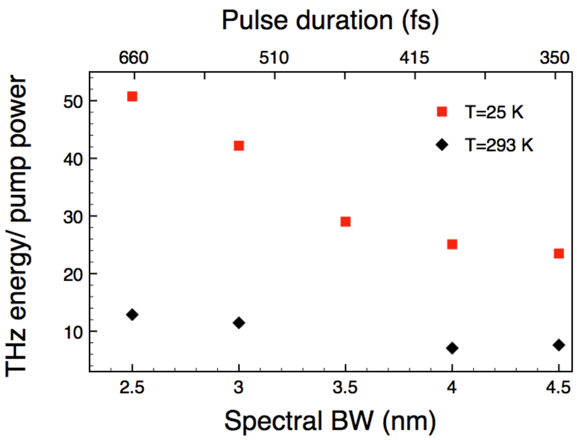

Figure 3 THz output energy normalized to the pump power as function of the laser pulse duration and spectral width for (black diamonds) room temperature and (red points) cryogenic temperature.

As numerically predicted [12], the pump-to-THz conversion efficiency increases with pulse duration. In fact, for this pulse width the group velocity dispersion in LN is smaller and the phase matching can be reached over longer crystal length. As shown in the Fig. 3. at CT the maximum efficiency is 0.36% for the 650-fs pulse, which corresponds to the longest pulse available from our laser system. For this pump duration the amount of THz pulse energy is twice as high as the one measured for the shortest pump pulse under otherwise equal conditions. The observed behavior is qualitatively reproduced over the whole temperature range explored in the experiment. The measurements indicate moreover that, for specific pulse duration, the conversion efficiency is systematically higher for cryogenically cooled LN. While higher conversion efficiency is doubtlessly whished for high energy THz applications, it needs to be clarified how the THz spectrum is affected by the pump pulse duration. For the first time, a systematic measurement of the THz spectral shape as function of temperature and pump pulse width has been performed. The spectra are measured in air, with multi-shot first order interferometer equipped with THz sensitive detector.

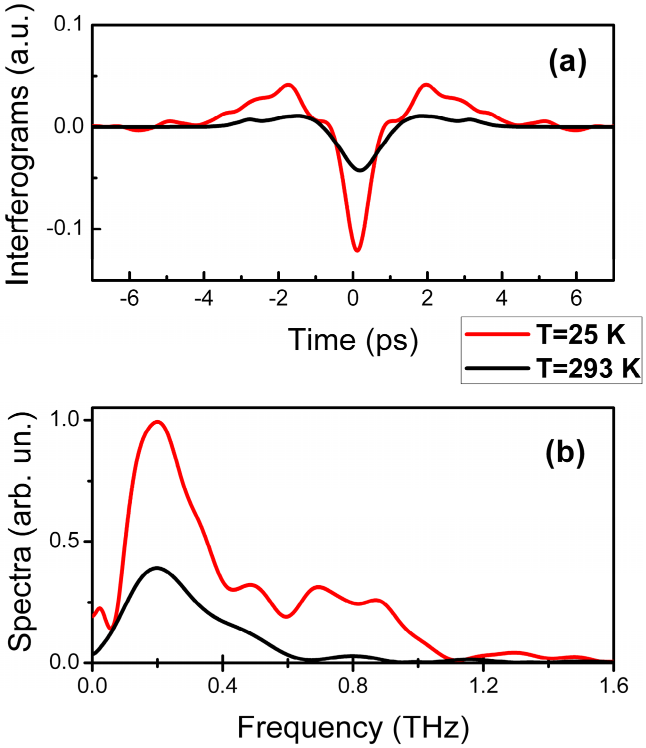

Figure 4 (a) THz interferograms and (b) retrieved spectral intensity obtained at (black curves) room and (red curves) cryogenic temperature for the shortest pump pulse duration (380 fs FWHM).

Shown in Fig. 4 (a) and (b) are THz interferograms and spectra at (black curve) RT and at (red curve) CT. Despite the curves are plotted in arbitrary units, the relative intensities are correct and can be used to estimate the improvements for the different spectral contents induced by cryogenic cooling the LN.

The spectral intensities shown in Fig. 4(b), are calculated by Fourier-transformation of the corresponding interferograms of Fig. 4 (a). For the generation of the broadest THz spectra at both temperatures we used the shortest available pump pulse (380 fs FWHM). Obviously the THz radiation covers a significantly broader range at CT with a shift in the cut-off frequency from 0.6 THz (RT) to 1.1 THz (CT). The occurrence of broader spectrum at 25 K is predicted by theory and it is due to the dramatic reduction of phonon coupling at frequency around 1 THz. The spectral modulations visible in both the spectra are associated to water absorption peaks (0.55 THz; 0.75 THz), in agreement with the data reported in the literature [18]. The corresponding interferograms indicate THz pulse duration slightly longer than 1 ps (FWHM). The results show that with LN at cryogenic temperature it is possible to fully cover the spectral region between 0.1 to 1 THz.

Reported in Fig. 5 are the THz spectra at (a) RT and (b) CT for different pump pulse durations. The relative intensities for RT and CT are accurate and permit the direct estimation of the specific effects of temperature and pump pulse duration on the THz spectral components. The figure shows clearly that shorter pump pulse gives rise to higher-frequency components. The enhancement of higher THz frequencies is more evident for cooled LN (Fig. 5b) because the high-frequency THz absorption is significantly reduced at CT. The experimental results show that broader THz spectra require shorter pump pulses, which inherently goes along with lower conversion efficiency, as shown in Fig. 3.

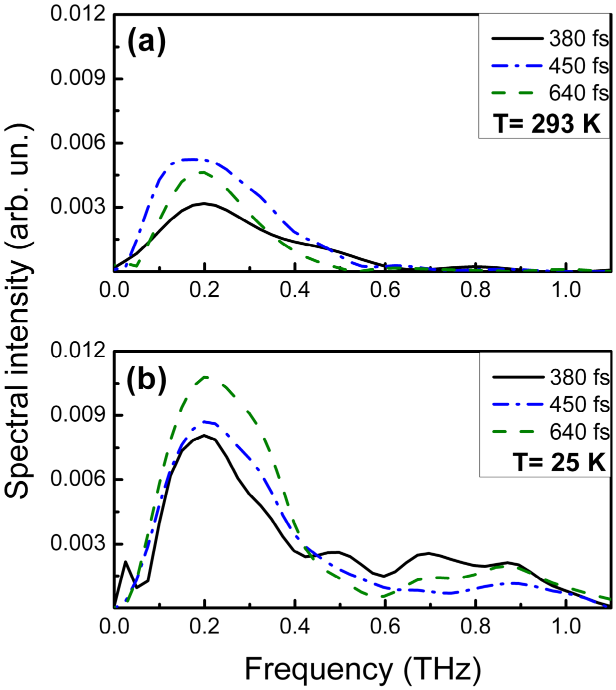

Figure 5. THz spectra obtained by different pump pulse width at (a) room and (b) at cryogenic temperature. (from ref. 18).

At CT the $LiNbO_3$ produces three times larger integrated signal as compared to RT. The maximum conversion efficiency achieved in our experiment is 0.12 % at RT and 0.36% at 25 K. Under the latter conditions the highest THz energy reached 45 μJ per pulse. The measured RT conversion efficiencies are similar to the values measured previously at longer pulses [10], and are lower than the theoretical predictions [16]. The reasons for this discrepancy needs further investigation and may be connected to impurities in the LN crystal used in the experiment [16]. The high pulse energy and the single cycle THz transient recorded in the experiment could potentially result at the beam focus in peak electric field of 2 MV/cm. This value is calculated assuming the pulse length measured with the THz interferometer. The spot size at the focus is estimated assuming a monochromatic spectrum at 0.4 THz (centroid of the CT spectrum) and a focusing system with numerical aperture of 1.

In conclusion, we experimentally explored the optimum conditions for intense THz pulse generation in $LiNbO3$ driven by a multi-mJ compact $Yb:CaF_2$ laser able to deliver variable transform limited pulses. The efficiency of OR and the emitted THz spectra are studied for various $LiNbO3$ temperatures and pump pulse durations. While the broadest THz spectra (0.1-1.2 THz) are achieved by using the shortest available pump pulses, longer pulses are beneficial for reaching highest energy conversion efficiency (0.36%) and pulse energies (45 uJ) at the cost of THz bandwidth. For all pulse durations cryogenic cooling of the LN systematically leads to higher conversion efficiencies and higher frequency components. Our experimental results indicate that the versatile $Yb:CaF_2$ laser could be an ideal tool for tailoring the THz output to the need of the specific THz application.

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