Generation of 1.5 µJ single-cycle terahertz pulses by optical rectification from a large aperture ZnTe crystal

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Abstract: We demonstrate the generation µJ-level, single-cycle terahertz pulses by optical rectification from a large-aperture ZnTe single crystal wafer. Energies up to 1.5 µJ per pulse and a spectral range extending to 3 THz were obtained using a 100 Hz Ti:sapphire laser source and a 75-mm-diameter, 0.5-mm-thick, (110) ZnTe crystal, corresponding to an average power of 150 µW and an energy conversion efficiency of 3.1 x 10^{-5}. We also demonstrate real-time imaging of the focused terahertz beam using a pyroelectric infrared camera.

OCIS codes: (260.3090) Infrared, far; (300.6270) Spectroscopy, far-infrared; (300.6500) Spectroscopy, time-resolved; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

The terahertz (THz) band (~ 0.1 - 10 THz) has recently drawn much attention due to its potential for both fundamental physics and an increasingly larger variety of applications. For example, many molecules exhibit distinctive absorptive and dispersive properties in the terahertz spectral range. In particular, the recent progress in both time-domain and time-resolved THz spectroscopy as well as THz imaging are providing unprecedented insight into the nature of molecular vibrations, carrier dynamics in semiconductors, protein kinetics, and other applications [1-3]. However, the applicability of THz sources is still critically dependent on the power available with current technology, which has prompted much research in developing pulsed high-intensity broadband THz sources. Furthermore, even though there has been some work on nonlinear terahertz electrodynamics [4-11], the ultrafast nonlinear optical properties of most materials in the terahertz frequency region remain relatively unexplored, mainly due to the lack of picosecond pulsed high-intensity THz sources. Synchrotron-based sources can generate picosecond THz pulse energies on the order of several 10's of microjoules [12, 13] and more recently up to 100 μJ [11], but their availability may be limited for comprehensive THz experiments. On the other hand, some of the highest THz pulse energies published to date using table-top laser sources and large-aperture photoconductive switches (along with the estimated peak electric fields at the focus) have been 0.8 μJ (~ 150

#86123 - $15.00 USD Received 6 Aug 2007; revised 24 Sep 2007; accepted 25 Sep 2007; published 27 Sep 2007

kV/cm) [14] and 0.4 μJ (~ 350 kV/cm) [15]. However, there are several drawbacks associated with this method. First, this technique requires a pulsed high voltage bias (~10 kV) in order to avoid excessive power dissipation in the switch associated with dc biasing, since the emitted THz amplitude scales linearly with bias voltage [15,16]. Second, the THz amplitude saturates at relatively low excitation fluences [16, 17]. Third, the high-voltage switching circuit can produce significant electrical noise, and the GaAs surface is susceptible to degradation due to high-voltage arcing effects at the electrodes. Other schemes have employed four-wave mixing in air (with an estimated THz pulse energy of ~30 nJ) [18] and laser-wakefield-accelerated electron bunches [19] to generate THz pulses with peak electric fields up to 400 kV/cm. Terahertz pulses with peak electric fields up to 150 kV/cm have also been reported recently from an ultrafast ionized air source [20]. In contrast, optical rectification (OR) in large-aperture nonlinear crystals represent an interesting alternative for high-intensity THz pulse generation, which is typically much easier to implement than most other schemes. In particular, in comparison to large-aperture photoconductive sources, OR enables the generation of the broad bandwidth THz pulses [21] required for time-resolved spectroscopic applications. Optical rectification in zinc-blende crystals such as ZnTe [22] and GaP [23] is commonly used by many groups as a way to realize OR THz pulse sources. THz pulse energies up to 0.6 nJ have been observed from a large-aperture (25 mm diameter) ZnTe emitter using 400 μJ excitation pulses from a 1 kHz amplified laser source (energy conversion efficiency of ~1.5 x 10^{-6}) [24]. It was also found that larger aperture OR emitters helped reduce saturation effects due to two-photon absorption of the pump beam and free-carrier absorption of the THz pulse within the ZnTe crystal [24].

In this article, we report high-energy THz pulse generation by optical rectification in a large aperture (75 mm diameter) ZnTe single crystal wafer at the Advanced Laser Light Source (ALLS). Single-cycle THz pulses with energies as high as 1.5 μJ per pulse and with a spectral range extending to 3 THz were observed using 48 mJ, 800 nm, 30 fs excitation pulses (energy conversion efficiency of ~3.1 x 10^{-5}) at a repetition rate of 100 Hz, which represents, to the best of our knowledge, the highest THz pulse energy ever measured from a ZnTe optical rectification source. The THz pulses have also been fully characterized in spectrum, temporal duration, and electric field amplitude. We note that even though our measured THz pulse energies exceed the highest published THz pulse energies to date reported by You *et al.* [14] of 0.8 μJ and Budiarto *et al.* [15] of 0.4 μJ from large-aperture photoconductive emitters, optical rectification in $LiNbO_3$ using tilted pulse front excitation has led to near single-cycle THz pulses with energies of 0.24 μJ (energy conversion efficiency of 5 x 10^{-4}) [25,26], and more recently up to 10 μJ using 20 mJ, 800 nm excitation pulses at a repetition rate of 10 Hz [27]. It is clear that high-intensity THz pulse generation by optical rectification is proving to be a promising technique for realizing nonlinear THz pulse experiments and developing novel applications requiring high-energy THz pulses.

2. Experimental

The ALLS beam line used in these experiments provided 800 nm, 30 fs laser pulses with energies as high as 48 mJ (at the ZnTe crystal position) at a repetition rate of 100 Hz. The experimental setup, as shown in Fig. 1, is comprised of three main parts: a THz generation chamber held under vacuum (~10^{-6} torr), an 800-nm probe beam propagating in air, and a dry-N_2-purged electro-optic (EO) sampling detection line. The THz emitter consisted of a 0.5-mm-thick (110) ZnTe single crystal wafer with a diameter of 75 mm (Nikko Materials). To minimize saturation effects and damage to the crystal surface, the 800-nm pulse was expanded to about 40 mm FWHM (36-cm^2 at $1/e^2$) at the surface of the ZnTe emitter. In addition, the ZnTe crystal was rotated in its mount to maximize the THz emission via OR [22]. Any remaining 800-nm light transmitted through the ZnTe wafer was blocked using a black polyethylene absorber which was transparent to the THz radiation. The emitted THz pulse is first focused by a gold-coated off-axis parabolic reflector (4" clear aperture, 6" focal length) in

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vacuum through a 2-mm-thick polypropylene window, and is then re-collimated by another 4" off-axis parabolic reflector outside the vacuum chamber. To facilitate steering of the THz beam, the beam size was reduced by a factor of 2 using 2" clear aperture, 3" focal length, off-axis parabolic reflectors, thereby preserving the numerical aperture of the terahertz optics. A chopper positioned at the first focus allowed modulation of the THz beam. Two 4" diameter wire-grid polarizers were also used to control the intensity and the polarization of the THz beam and were primarily used for characterizing the linearity of the detection system.

One difficulty that pulsed THz experiments often face is the lack of a truly calibrated and sensitive energy detector in the appropriate frequency range. To obtain a reliable reading of the total THz pulse energy emitted from the ZnTe source, we used a pyroelectric detector (Microtech Instruments) with a sensitivity of 2100 V/W at 1 THz, calibrated by the manufacturer at terahertz frequencies between 0.1 and 3 THz. A second pyroelectric detector (Coherent-Molectron) with a specified sensitivity of 2624 V/J at 1.06 μm provided more routine measurements of the total THz pulse energy by reading the amplitude of the output voltage pulse on an oscilloscope. We calibrated the response in the terahertz frequency range using the first pyroelectric detector (Microtech Instruments) and found that the sensitivity of the Coherent-Molectron pyroelectric detector at 1 THz was approximately 1.8 times less than the specified sensitivity at 1.06 μm. Such a reduction in sensitivity from the infrared to the terahertz frequency range can be expected due to the nature of the thin absorbing layer on the surface of the pyroelectric detector element [15].

Free-space electro-optic (EO) sampling in a second (110) ZnTe crystal [28,29] with a thickness of 20 μm, mounted on top of a 0.5 mm thick (100) backing crystal, was used to detect the THz pulse wave form, which could be scanned using a delay stage. A lock-in amplifier connected to the output of the balanced photodiodes and referenced to the chopper was used to acquire the THz wave forms.

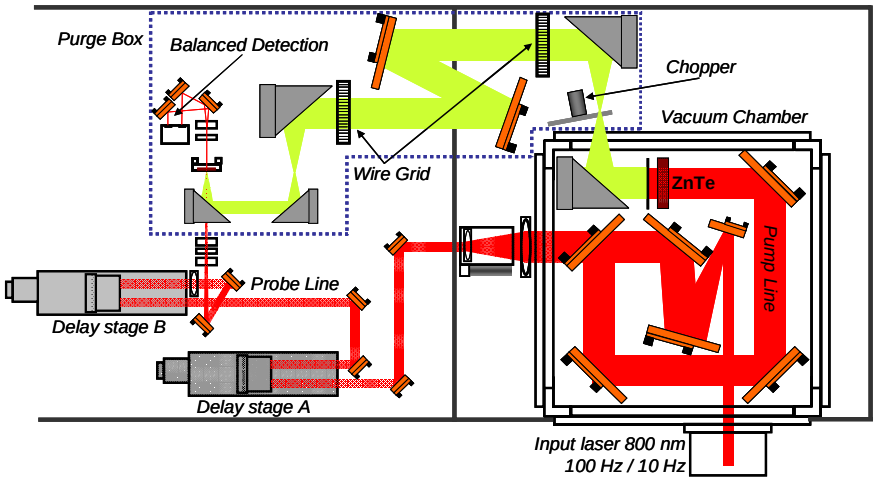

Fig. 1. Schematic of the high-power THz pulse source at the Advanced Laser Light Source.

3. Results

3.1. Energy measurements

Figure 2 shows the total THz pulse energy generated from the large-aperture ZnTe wafer as a function of the normally incident excitation laser fluence (pump energy) between 28 μJ/cm^2 (1 mJ) and 1.33 mJ/cm^2 (48 mJ). At the maximum pump energy of 48 mJ the THz pulse energy

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is 1.5 μJ, corresponding to an energy conversion efficiency of 3.1 x 10^{-5} and an average THz power of 150 μW (100 Hz repetition rate).

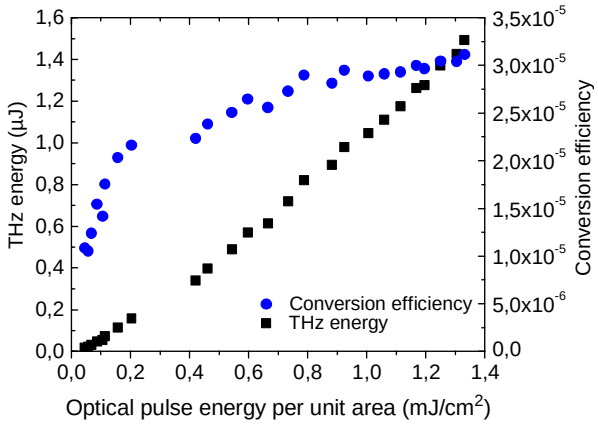

Fig. 2. Total THz pulse energy emitted from the large-aperture ZnTe source (measured just outside the vacuum chamber) as a function of the incident 800 nm laser pulse energy per unit area. The corresponding energy conversion efficiency is also shown.

For an optical rectification process we would normally expect the THz pulse energy to increase quadratically with incident pump fluence. However, at high pump fluences two-photon absorption in the ZnTe wafer leads to depletion of the pump beam as well as free-carrier absorption of the THz beam, which limits the THz output energy [24, 30-32]. Löffler *et al*. [24] showed that the onset of this saturation effect in ZnTe occurred at pump fluences close to 50 μJ/cm^2. Below this fluence, Löffler, *et al*. observed that the THz pulse energy scaled quadratically with pump energy, whereas above this fluence an approximately linear behavior was seen. The pump fluences used here (Fig. 2) are typically well above the saturation fluence of 50 μJ/cm^2 reported by Löffler, *et al*., and so we do not expect to see a quadratic increase in the THz pulse energy as a function of 800 nm pump energy over the entire range. Instead we see a slightly superlinear increase of the THz pulse energy as a function of the 800 nm excitation pulse energy, which is consistent with the results of Löffler *et al*. above the saturation fluence for ZnTe where two-photon absorption effects become significant. Even if locally the beam experiences complete saturation where the maximum pump fluence is achieved, the THz emission continues to grow at the (lower intensity) wings, which may contribute to the non-trivial behavior shown in Fig. 2. The exact nature of this dependence is still under investigation. It is worth noting, however, that Löffler *et al*. observed an energy conversion efficiency in ZnTe that saturated at a maximum value of 1.5 x 10^{-6} at pump fluences above pump 150 μJ/cm^2. In our case we see from Fig. 2 a maximum energy conversion efficiency of 3.1 x 10^{-5}, which is 20 times higher than that reported by Löffler *et al*. Furthermore, the energy conversion efficiency plotted in Fig. 2 continues to increase even at the maximum fluence of 1.4 mJ/cm^2. A possible explanation for this may be due to the lower repetition rate (100 Hz) of our laser source compared to that of Löffler *et al*. (1 kHz). The lower repetition rate may give residual free carriers enough time to recombine between subsequent pulses, thereby keeping the background free carrier concentration much lower than if we operated at 1 kHz.

3.2. Azimuthal angle dependence of the THz field

The azimuthal angle dependence of the emitted THz pulse energy was measured to confirm that the THz emission was due to optical rectification. Let θ be the angle between the Y axis ([001] direction) of our (110) cut ZnTe crystal and the polarization vector of the incident

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optical field. The THz field, E_{THz}, will be proportional to the following vector in the XY reference system, where X is the [-110] direction:

$$\begin{pmatrix} E_{THz\,X} \\ E_{THz\,Y} \end{pmatrix} \propto \begin{pmatrix} \sin(2\theta) \\ \sin^2(\theta) \end{pmatrix}, \tag{1}$$

The THz energy will thus have an angular dependence of the type $\sin^2(2\theta)+\sin^4(\theta)$ [22]. Figure 3 shows the experimentally measured THz energy as a function of the azimuthal angle θ, which is found to be in good agreement with this theoretical dependence.

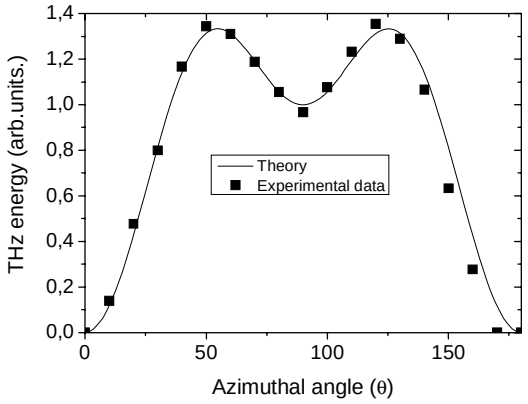

Fig. 3. Azimuthal angle dependence of the THz pulse energy generated from the ZnTe source. The solid line is the theoretically predicted response for an optical rectification source according to Eq. (1).

In general, the polarization direction of the THz beam is given by:

$$\alpha = \tan^{-1}\left(\frac{2}{\tan(\theta)}\right) \tag{2}$$

with respect to the Y direction. When θ=90°, then α=0° and the THz beam is orthogonally polarized with regards to the optical beam. When $\tan(\theta) = \sqrt{2}$, $\alpha = \theta = 54.7^0$, and the THz energy reaches its maximum. All the results presented below were obtained fixing the crystal orientation according to this last condition.

3.3. Electric field evaluation and imaging of THz spot size

Figure 4(a) shows the single-cycle THz pulse wave form measured through EO sampling in the 20 μm thick ZnTe detection crystal. This acquisition was done after purging with N_2 to remove water vapor, with the THz beam propagating 3 m from the source to the detector. The THz pulse energy measured at the detection crystal position was 0.76 μJ. The corresponding THz frequency spectrum, shown in Fig. 4(b), is centered at 0.6 THz and extends all the way to 3 THz. We note that the energy in the THz pulses emitted from the ZnTe OR source is spread over a broader frequency range compared to $LiNbO_3$ OR sources. By carrying out the Hilbert transform of the electric field pulse, we estimate a 0.8 ps (FWHM) pulse width at the detector position, as shown in the inset of Fig. 4(a).

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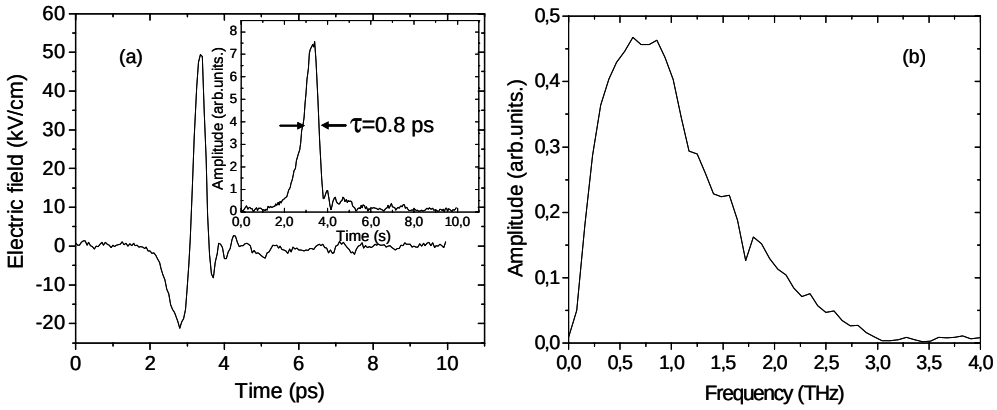

Fig. 4. (a). THz pulse wave form obtained in a N_2-purged environment using a 20 μm thick ZnTe EO detection crystal. The THz pulse energy measured at the position of the ZnTe detection crystal was equal to 0.76 μJ. The inset shows the corresponding Hilbert transform of the wave form. (b) Frequency spectrum of the THz pulse wave form shown in (a).

From the modulation of the balanced photodetectors A and B in the EO detection system, one can measure the phase shift θ induced in the ZnTe detection crystal by the peak electric field of the THz pulse. This can be used in turn to determine the peak THz electric field, E_{THz}, using the formula:

$$\frac{A-B}{A+B} = \sin\theta = \frac{2\pi}{\lambda} n_0^3 r_{41} E_{THz} L , \qquad (3)$$

where $n_0 = 2.85$ is the index of refraction of ZnTe at $\lambda = 800$ nm, $r_{41} = 4.04$ pm/V is the electro-optical coefficient of ZnTe [28,29] and L = 20 μm is the detector thickness. For an observed maximum modulation of (A-B)/(A+B) = 6.16%, we estimate the peak electric field to be 41.9 kV/cm. If we account for Fresnel reflection losses of about 15%, then the peak THz field becomes 49.4 kV/cm, as shown in Fig. 4(a).

Since we have characterized both the energy and electric field of the THz source, we can compare the two to check for consistency. In Fig. 5, we plot the total THz pulse energy and the square of the measured THz peak electric field, showing good agreement between the two. (The reason no data is plotted for laser energies between 5 and 15 mJ in Figs. 2 and 5 is due to a discontinuity in the attenuation system of the Ti:Sapphire laser when changing between fluence ranges.) Since both the electro-optic and pyroelectric measurements exhibit similar scaling with pump energy, as demonstrated in Fig. 5, the peak electric field at the focus of the 1.5 μJ THz pulses must be about 69 kV/cm.

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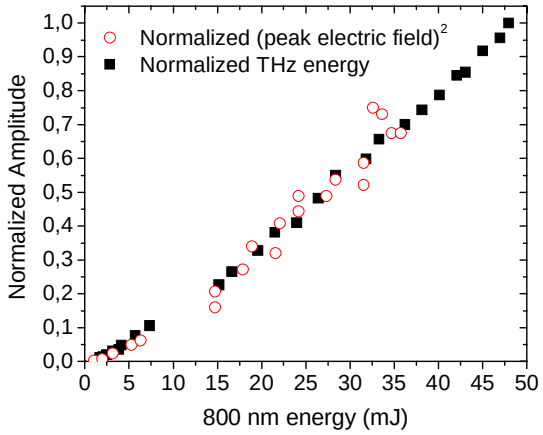

Fig. 5. The square of the THz peak electric field normalized to the total THz pulse energy as a function of the laser energy.

To further characterize the THz beam, a $BaSrTiO_3$ (BST) pyroelectric infrared camera (Electrophysics model PV320-L2Z) was used to image the THz beam at the focus. This camera operates with an internal 10 Hz chopper and has a 320×240 pixel imaging array with a pixel spacing of 48.5 μm. In Fig. 6(a), we show an image obtained with focused 0.76 μJ THz pulses. Figure 6(b) shows the corresponding line-profiles of the THz spot in the horizontal and vertical directions. Assuming a Gaussian profile, we evaluate the THz beam spot size at the focus to be 1.57×1.91 mm, for a total THz spot area of 2.36 mm^2. The electric field at the focus can be estimated from [33]:

$$W = \frac{\tau A \left| E_{THz}^{pk} \right|^2}{2\eta_0}, \qquad (4)$$

where W is the THz energy, τ is the pulse width, A is the THz beam area evaluated at the focus, E_{THz}^{pk} is the electric field peak and η_0 is the impedance of free space. Using Eq. 4, along with the beam spot size analysis discussed above, a value of 174 kV/cm for 0.76 μJ of THz energy is calculated, which is not in agreement with the value of 49 kV/cm measured directly by electro-optic sampling. It is not exactly clear why these two results disagree with each other, and we are still investigating this discrepancy. A complete integration of the THz power spectrum over the entire beam spot could be more accurate, and Eq. 4 may be too simple to estimate the peak field for a given THz pulse energy when the THz field consists of a broad-band, single cycle pulse.

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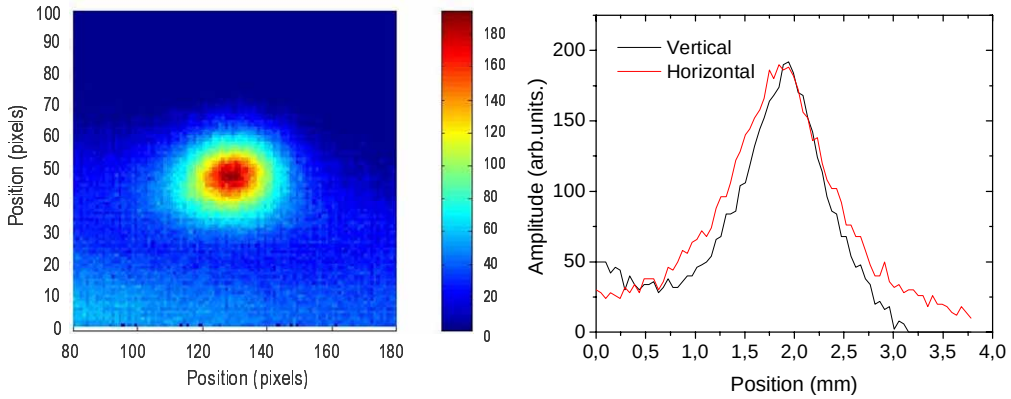

Fig. 6. (a). Real time Image of the THz pulse focus obtained with an infrared pyroelectric camera (Electrophysics model PV320-L2Z). The THz pulse energy at the focal plane was 0.76 uJ. (b) Line profiles of the THz spot size from the image in (a) in the horizontal and vertical directions.

3. Conclusion

In conclusion, we have demonstrated and characterized a high energy THz pulse source using optical rectification in a large aperture ZnTe crystal. Energies up to 1.5 μJ are observed in single-cycle THz pulses with a 3 THz spectral range. This is, to the best of our knowledge, the highest THz pulse energy ever measured from a ZnTe optical rectification source. Approximately linear (slightly superlinear) scaling of the THz pulse energy is observed for laser excitation fluences up to 1.3 mJ/cm^2. We also successfully performed real-time imaging of the THz spot at the focus using a pyroelectric infrared camera. The high intensity THz pulse source will be used for exploring the nonlinear terahertz interactions in materials and for THz imaging applications.

Acknowledgments

We wish to acknowledge financial support from NSERC, NSERC Strategic Projects, and INRS. L. Razzari wishes to acknowledge a Marie Curie Outgoing International Fellowship (contract n. 040514). We are also thankful to Jean-Philippe Moreau for technical assistance with the laser source, and to Francois Martin for providing the Electrophysics camera.

#86123 - $15.00 USD Received 6 Aug 2007; revised 24 Sep 2007; accepted 25 Sep 2007; published 27 Sep 2007