Generation of 10 μJ ultrashort terahertz pulses by optical rectification

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Generation of near single-cycle pulses centered at 0.5 THz frequency with up to 10 μJ energy, 100 μW average power, and 5.0 MW peak power was demonstrated by tilting the intensity front of a femtosecond optical pump pulse from a 10 Hz Ti:sapphire laser to match the phonon-polariton phase velocity to the group velocity of the pump pulses in a lithium niobate crystal. Terahertz pulse intensity as high as 10 MW/cm^2 was achieved. The photon conversion efficiency was 45% and the calculated peak electric field strength at the focus of an off-axis parabolic mirror was 250 kV/cm. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2734374]

There exists a strong and yet unmet need for high energy ultrashort terahertz pulses for applications in imaging, security control, and nonlinear terahertz spectroscopy. Until recently, only free electron laser sources have been able to generate terahertz pulses that have at least 1 μJ of energy.[1] Generation of terahertz pulses with 0.5 μJ energy by large area photoconductive switches was demonstrated one decade ago.[2] The generation of single-cycle terahertz pulses via four-wave mixing of the fundamental and the second harmonic of 25 fs pulses from a Ti:sapphire amplifier in air plasma was recently reported.[3] Although the energy of the terahertz pulses was only around 30 nJ, the created peak terahertz electric field was as high as 400 kV/cm because of the relatively high (3 THz) main frequency and the good focusability. While optical rectification has long provided an accessible means to generate terahertz pulses,[4–6] until recently their energies have been well under 100 nJ.

Matching between the optical group velocity and the terahertz phase velocity is crucial for efficient optical rectification. In materials such as ZnTe and GaP, collinear phase matching is possible with appropriate pump wavelengths.[4–6] For bulk high-dielectric-constant materials including lithium niobate (LN) and other ferroelectrics, which offer attractive electro-optic coefficients and other parameters, velocity matching of the terahertz phonon-polariton wave that is generated inside the crystal cannot be achieved collinearly. For such a situation, tilting of the pump pulse front by diffraction off a grating has been proposed[7] and applied[8,9] to reach velocity matching in $LiNbO_3$. Very recently, this method generated terahertz pulses with 240 nJ of energy.[10] This approach is advantageous compared to discrete tilting of the pump pulse front.[11–14]

In this letter we present results on one order of magnitude further upscaling of terahertz energy. The present results were obtained using a 10 Hz laser system, but it is clear that substantial further improvement at high repetition rates is also possible. In addition, the present approach could be combined with phonon-polariton amplification by temporally and spatially shifted pump field components that are generated through spatiotemporal femtosecond pulse shaping[11–14] to produce intense terahertz fields with arbitrary wave forms.

A setup similar to that described in Ref. 9 was used. A Ti:sapphire amplifier system produced near-infrared, 20 mJ pulses at a 10 Hz repetition rate. The intensity front of these pulses was tilted by a 2000 line/mm grating and imaged with a 60 mm lens, with a demagnification of 2, onto the input surface of the 0.6% MgO doped stoichiometric $LiNbO_3$ (sLN) crystal. A $\lambda/2$ plate was used to rotate the horizontally polarized pump light from the grating to vertical polarization, parallel to the optic axis of the sLN crystal. The pump spot on the crystal surface was 6 mm in diameter. After losses at the grating and elsewhere, approximately 16 mJ of optical energy reached the crystal. A Scientech P09 pyroelectric detector calibrated for 1 μm with 9 mm active diameter was used to measure the emitted terahertz energy. Single-shot imaging of polaritons[14,15] was used to characterize the temporal profile of the terahertz pulses after they were coupled out of the sLN crystal in which they were generated and into an adjacent 100-μm-thick sLN crystal, as depicted in Fig. 1. A frequency-doubled probe laser beam was expanded to a large area at the imaging crystal and projected through a 1:1 imaging system onto a Princeton Instrument Acton Pixis charge coupled device (CCD) camera.

We were able to generate terahertz pulses with energy up to 9.9±0.3 μJ. The optimal energy was not observed at the shortest possible optical pump pulse duration due to strong nonlinear effects in sLN. Adjustment of the amplifier com-

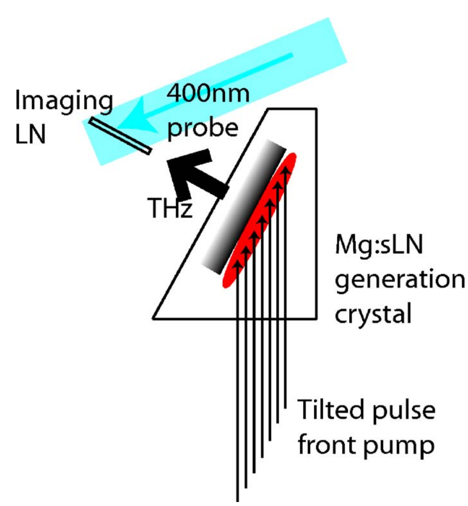

FIG. 1. (Color online) Experimental geometry for the generation and detection of terahertz radiation.

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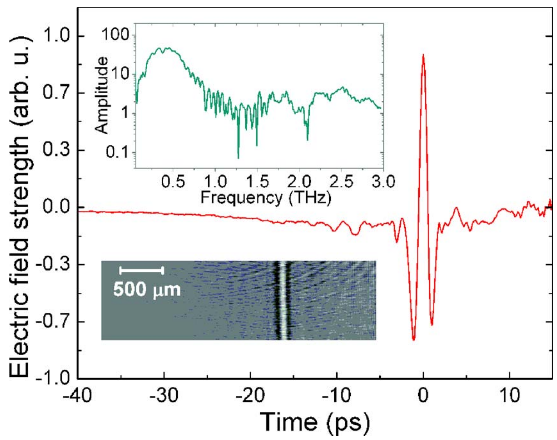

FIG. 2. (Color online) Temporal profile and spectrum of the terahertz pulse, determined from the grayscale image shown.

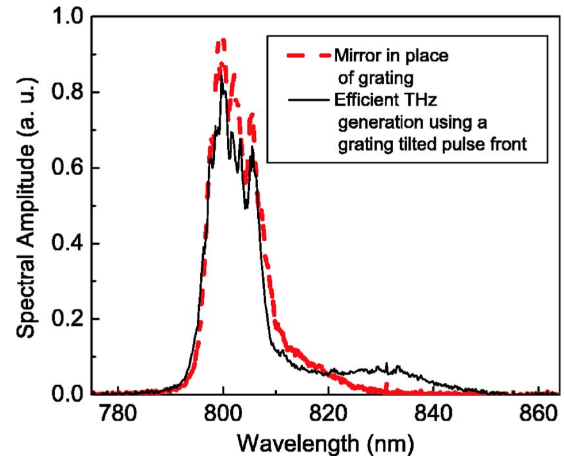

FIG. 3. (Color online) Spectra of the pump pulses scattered from the LN crystal with optimized (solid curve) and reduced (dashed curve) terahertz generations.

pressor settings for maximum terahertz energy yielded an optical pulse duration of about 400 fs (measured by single-shot autocorrelation). Figure 2 shows a raw CCD image of the terahertz pulse measured under these conditions. The reconstructed terahertz pulse profile recorded 10 mm from the generation crystal and the calculated spectral amplitude are also shown. The pulse was near single cycle with maximal spectral amplitude near 0.5 THz.

The pump-to-terahertz energy conversion efficiency calculated from the measured terahertz energy was 6.0×10^{-4}. The photon conversion efficiency was evaluated by taking into account the peak terahertz frequency of 0.5 THz, which yielded the value of 45%, eclipsing the earlier report[10] of 10% photon efficiency obtained at lower pump energy. (Note that this value could exceed 100%, in principle.[16]) The peak power generated was 5.0 MW, and the average power was 100 μW. The peak intensities of the pump and terahertz pulses were high, more than 1×10^{11} and $1 \times 10^{7} \mathrm{W/cm^2}$, respectively. Using the experimentally measured energy, pulse duration, and focused spot size of the terahertz beam, the terahertz field at the focus of a parabolic mirror was calculated to be 250 kV/cm. As a consequence of the high pump intensity, despite the small nonlinear index of refraction in LN ($n_2 = 5.3 \times 10^{-15} \mathrm{cm^2/W}$),[17] self-phase modulation of the pump light was still observed. This was monitored by measuring the spectrum of the pump light that was scattered from the back surface of the sLN crystal.

The high energy of the generated terahertz pulses causes a significant change to the optical pump spectrum. Figure 3 depicts the spectra in the cases of optimized and reduced terahertz generations, the latter effected through replacement of the grating with a mirror such that the pump pulse front was no longer tilted for velocity matching. A very pronounced broadening on the red side of the spectrum[18] was observed in the case of optimized terahertz generation. Less efficient terahertz generation led to greatly reduced spectral broadening, corroborating the cascading process[16] via difference-frequency generation (in our case optical rectification) as the principal cause of the spectral shift. A significant reduction in redshifting was also observed by rotating the pump polarization from the optic axis direction at which terahertz generation is maximized.

To provide a quantitative estimate of the extent of redshifting, we defined the average wavelength $\bar{\lambda}$ as the first moment of the measured pulse spectrum $S(\lambda)$ using the formula

$$\bar{\lambda} = \frac{\int \lambda S(\lambda) d\lambda}{\int S(\lambda) d\lambda}. \tag{1}$$

Average wavelength values of 803.7 and 806.3 nm were obtained for the input optical pump and the scattered optical output spectra, yielding a spectrum-averaged redshift of 2.6 nm in the case of our most efficient terahertz generation. The terahertz spectrum has an average frequency [calculated as the first moment of the frequency spectrum, as in Eq. (1)] of 0.66 THz. The generation of a 0.66 THz photon is accompanied by the simultaneous appearance of a photon with wavelength about 1.4 nm longer than that of the original pump photon which is consumed. Should terahertz generation alone account for the pump redshift, the photon conversion efficiency would be 190% (i.e., 2.6 nm/1.4 nm=1.9). However, the efficiency measured outside of the LN crystal was 45%, with the corresponding efficiency inside the crystal before reflective loss at the air interface being 80%. The remaining large discrepancy might be attributed to terahertz intensity-dependent index and absorption[19] effects as well as ordinary terahertz absorption[20] in the LN crystal. We note that the pump spectrum is not uniformly shifted to the red. This may reflect the fact that the extent of redshifting may not be uniform across the tilted pulse front due to different terahertz amplitudes encountered and different path lengths in the crystal.

There remain ways for further enhancement of the terahertz energy. A three- to fivefold increase of the terahertz energy was demonstrated[9] and theoretically confirmed[21] by cooling the LN, which significantly reduced the terahertz absorption. Further improvement may also be possible through optimization of the pump pulse duration and bandwidth. Recent results[21,22] have indicated an optimal transform limited bandwidth that is comparable to the terahertz frequency that is being driven. In the present case it is possible that further scaling of the crystal size and beam size to reduce the terahertz intensity will lead to higher overall terahertz output energy and, after focusing in air, higher field amplitude. Even

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at the present level of terahertz energy, the inclusion of cylindrical focusing will lead to better focusing and a higher field amplitude. The combination of the tilted pulse front with femtosecond pulse shaping will enable generation of narrow band or shaped terahertz fields with high energies and field amplitudes.

In conclusion, upscaling of terahertz pulse energy generated via a tilted intensity front of an 800 nm pump pulse has been demonstrated with a 10 Hz laser system. 10 μJ of energy in 0.5 THz pulses with field strength of 250 kV/cm can now be routinely obtained in a convenient tabletop setup. The terahertz pulse energy and field strength generated through optical rectification, hitherto available only in large facility sources such as free electron lasers, open up exciting experimental opportunities in nonlinear terahertz spectroscopy and terahertz coherent control. Our results also suggest possible approaches to improvement of terahertz signal levels in imaging and remote sensing applications.

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