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Generation of high power tunable multicycle teraherz pulses

Zhao Chen,[a] Xibin Zhou, Christopher A. Werley, and Keith A. Nelson[b]
Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

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We demonstrate generation of high-power, multicycle, and frequency-tunable terahertz pulses with microjoule energies by tilting the intensity front of a quasi-sinusoidal intensity-modulated optical waveform. The spatiotemporally shaped waveform undergoes difference-frequency mixing in lithium niobate, generating a THz phonon-polariton wave whose electromagnetic component is coupled out to free space. The narrowband THz spectrum is tunable between 0.3–1.3 THz, with adjustable bandwidths generally less than 0.1 THz. At 10 Hz and 1 kHz repetition rates, 10 μJ and 1 μJ THz pulse energies are achieved, respectively, over a broad frequency range. © *2011 American Institute of Physics*. [doi:10.1063/1.3624919]

Intense single-cycle terahertz (THz) pulses with microjoule energies[1] have become an essential tool for the emerging field of nonlinear THz optics and spectroscopy.[2–5] The THz generation technique is based on nonresonant optical excitation of coherent phonon-polaritons through impulsive stimulated Raman scattering in ferroelectric crystals such as lithium tantalate (LT) and lithium niobate (LN),[6–8] with the THz waveform determined through optical rectification of the pump pulse. Collinear matching between the group velocity of the near-infrared pump laser light and the phase velocity of the THz phonon-polariton wave cannot be achieved because of the high THz dielectric constant and refractive index of the ferroelectric crystals, in contrast to crystals like ZnTe which have lower THz refractive index but also considerably lower nonlinear coefficients than LN. Noncollinear velocity matching is possible by using sequences of spatially and temporally shifted optical pulses[9,10] or, most expediently, using a diffraction grating to tilt the intensity front of a single pump laser pulse.[11,24] The electromagnetic component of the phonon-polariton wave can be coupled out of the crystalline generation medium to produce a free-space THz pulse.

For many applications, frequency-tunable multicycle THz pulses would be preferred over typical broadband, near-single-cycle pulses. For example, the multicycle pulse can excite a selected resonance without influencing neighboring modes or simplify the identification of new frequency components generated in nonlinear THz processes. Well defined multi-cycle terahertz pulses can be excited inside ferroelectric crystals by optical waveforms that are shaped temporally[13] or spatially[7,12,14] or both,[10,14] including a tilted intensity front with the tilt angle set to match the THz phase velocity over a narrow rather than broad frequency range in bulk LN or in a LN slab waveguide.[15,24] However, none of these approaches is optimized for high-energy THz pulses since each cycle of the multicycle THz field is not pumped continually by the optical pulse or pulse sequence. Temporal pulse shaping has been used to generate a multiple-pulse optical waveform which was then spatiotemporally shaped with a tilted intensity front,[16] but the temporal shaping was not very versatile. However, if optimized, this approach should allow generation of high-energy multicycle THz pulses.

An effective method has been demonstrated for temporal shaping of an optical waveform for multicycle THz generation.[17] The chirp-and-delay approach yields a quasi-sinusoidal optical intensity modulation with a specified frequency and number of cycles. An optical pulse with center frequency ω_0 is chirped at rate β and split into two parts which are recombined interferometrically with a relative time delay τ, so that the instantaneous superposed frequencies $\omega = \omega_0 + \beta t$ and $\omega = \omega_0 + \beta(t - \tau)$ are swept linearly across the pulse duration. If the time delay is relatively small compared to the stretched pulse duration, the interference between two pulses leads to a quasi-sinusoidal intensity modulation at the constant difference frequency $\Omega = \beta\tau = 2\tau/T_0T_1$, where T_0 is the transform limited $1/e$ field half-width and T_1 is the stretched pulse $1/e$ field half-width.[17] The number of cycles within the full width at half maximum (FWHM) of the intensity envelope (Gaussian in profile if the transform-limited pulse was) can be written as $N = \sigma\tau$, where σ is the optical pulse bandwidth. Therefore, the modulation frequency can be continuously tuned by varying the chirp and/or the delay while the number of cycles can be independently controlled by varying only the delay. Chirp-and-delay optical modulation has been used for THz generation by optical rectification in semiconductor photoconductive antennas,[17,18] in ZnTe,[19] and in a synchrotron electron bunch.[20] However, the pulse energies and peak fields were relatively low.

In this letter, we demonstrate generation of high power multicycle frequency-tunable terahertz pulses by combining chirp-and-delay temporal shaping with tilted intensity front spatiotemporal shaping to tilt the intensity front of a quasi-sinusoidal intensity-modulated optical waveform. This represents an extension of spatiotemporal shaping of the optical field from a particularly simple form (single pulse that undergoes a linear spatial and temporal sweep at a speed set by the tilted intensity front) to a form that is still relatively simple

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[a] Electronic mail: zhaochen@mit.edu.
[b] Electronic mail: kanelson@mit.edu.

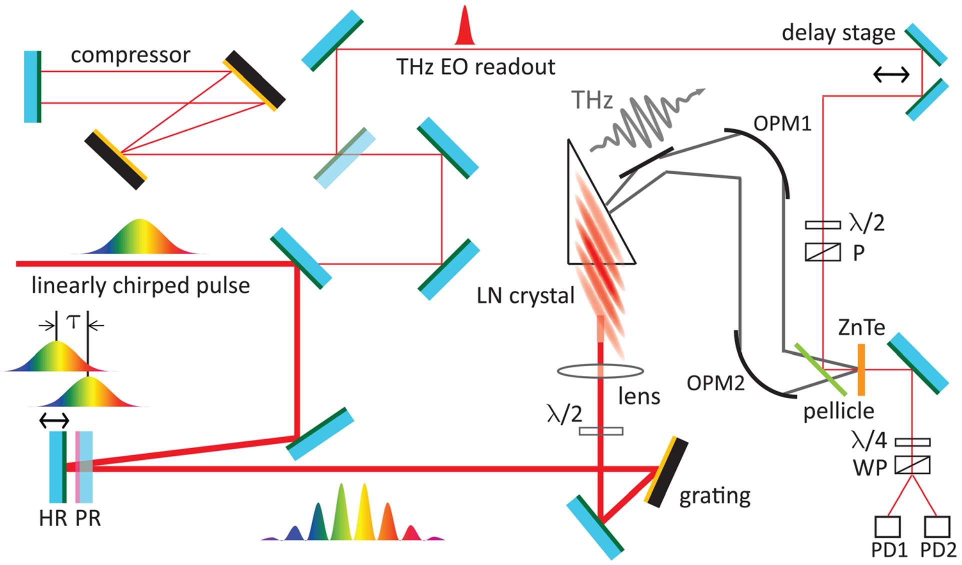

FIG. 1. (Color online) Schematic illustration of the experimental setup. Linearly chirped pulses were spatially overlapped with a varied time delay through a homemade Gires-Tournois etalon, and the intensity front was tilted through a grating-lens combination. A transform limited readout pulse was used to measure the THz field profile through electro-optic sampling and also to measure the chirp-and-delay optical waveform through cross-correlation. $\lambda/2$, half waveplate; $\lambda/4$, quarter waveplate; P, polarizer; OPM, off-axis parabolic mirror; PD, photodiode; and WP, wollaston prism.

(temporally intensity-modulated waveform that undergoes a linear spatial and temporal sweep) and that is well adapted for generation of an important class of THz waveforms—short multicycle pulses with well defined, tunable frequencies—with the high pulse energies and field amplitudes that noncollinear velocity matching in LN can provide. Completely reconfigurable spatiotemporal shaping of the optical field can be employed for generation of arbitrary THz waveforms in LN,[8] but the present approach is simpler and supports higher optical pulse energies for generation of intense frequency-tunable THz pulses.

The experimental layout is shown in Fig. 1. Near-infrared optical pulses with 6 mJ energy at 1 kHz repetition rate were produced from a two-stage commercial Ti:sapphire amplifier system, and the compressor grating spacing was reduced to generate positively chirped pulses with durations around 10 ps. A 1% beam splitter was used to split off the THz readout pulse which was subsequently recompressed to the transform limit by a separate compressor. A homemade Gires-Tournois etalon made by a partial reflector (38% reflection efficiency) and a high reflector was used to introduce the variable time delay between chirped pulses. Each of the first and second reflections has 38% of the incident light, which is the primary contribution to the intensity modulation. Each higher reflection has 38% of the previous one among the rest of the pulses with total 24% of the incident light. The advantages of this arrangement compared with a Michelson interferometer are common path optics for interferometric stability of the superposed chirp-and-delayed pulses and negligible pump power loss because multiple reflections all propagate collinearly and contribute to difference-frequency mixing in the THz generation crystal. A third-order cross-correlation between the chirp-and-delayed output and a transform limited readout pulse is shown in Fig. 2. The Fourier transform (Fig. 2 inset) reveals some second harmonic of the selected difference frequency due to the superposition of multiple reflections in the etalon.

The chirp-and-delayed waveform was sent to the same tilted intensity front setup as reported earlier.[1] A 2000 line/mm grating was used to achieve the appropriate intensity front tilt in the −1 diffraction order, and a single 80 mm focal length spherical lens was used to image the grating surface onto the LN crystal. The beam was about 10 mm in diameter at the grating and 5 mm diameter at the LN surface. Approximately, 2/3 of the optical pulse energy reached the LN crystal. The input surface of the LN crystal was AR coated for 800 nm, and the crystal was specially shaped in order to couple the THz phonon-polariton wave at normal incidence into air. The THz radiation was directed and focused by two off-axis parabolic mirrors into a ZnTe crystal for electro-optic sampling measurement or into a Microtech pyroelectric detector for pulse energy measurement.

With 6 mJ near-infrared optical pulses at 1 kHz repetition rate, multicycle terahertz pulses were generated with energies up to 1.5 μJ. The THz center frequencies were tuned through adjustment of the etalon mirror separation, and at each separation, the THz output was optimized through adjustment of the intensity front tilt angle to optimize velocity matching for the selected THz frequency. The adjustments were small because the phonon-polariton dispersion curve in this frequency range is nearly linear in LN. The THz frequency tuning range was between 0.3 and 1.3 THz, and the bandwidths of the pulses were generally less than 0.1 THz. Fig. 3 shows THz waveforms throughout this tuning range. The second harmonic components of the optical waveform modulations were mostly filtered out by the THz

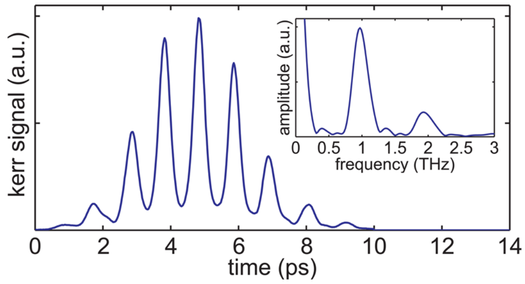

FIG. 2. (Color online) Third-order cross-correlation measurement between the chirp-and-delay etalon output and a transform limited readout pulse through optical Kerr effect in silica glass. The Fourier spectrum of the intensity modulations is shown in the inset.

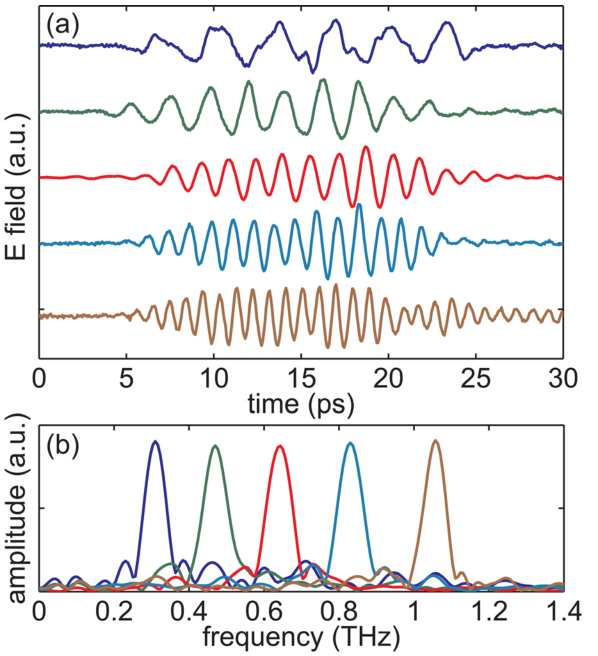

FIG. 3. (Color online) (a) THz field traces measured through electro-optic sampling and (b) the normalized Fourier spectra, generated with 6 mJ near-infrared chirp-and-delay tilted intensity front waveforms at 1 kHz repetition rate.

generation process. Comparison between the optical intensity modulation frequencies measured through cross-correlation as in Fig. 2 and the THz frequencies measured through electro-optical (EO) sampling as in Fig. 3 showed good agreement. We note, however, that for each chirp-and-delay setting of the optical modulation frequency, adjustment of the 1-lens imaging system to optimize the THz output energy caused a significant change in the THz frequency, up to 20% at the highest frequencies. We believe that this is due to a small but significant amount of chirp among the diffracted frequency components in the tilted intensity front near the image plane, as has been discussed.[21,22] This would effectively change the chirp-and-delay parameters and, therefore, would change the optical modulation frequency to a value somewhat different from that measured without the tilted intensity front.

In order to generate higher multicycle pulse energies, some experiments were conducted using a 10 Hz, 35 mJ optical pump pulse with the same chirp-and-delay and tilted intensity front setup. Fig. 4 shows the resulting waveform and Fourier spectrum of a multicycle pulse centered at 0.50 THz. The THz output had over 10 μJ energy. The phase shift in the field trace and the dip in the corresponding Fourier spectrum around 0.56 THz are due to water vapor absorption.

In summary, we have demonstrated generation of multicycle frequency-tunable THz pulses with energies up to 10 μJ, with the narrowband THz spectrum tunable between 0.3–1.3 THz. We believe higher energies are possible since we used the same focusing for the pump light that we normally use for an unmodulated short pump pulse, even though the modulated pump is far longer in duration and lower in intensity. With optimal focusing, the saturation regime should be reached and the optical-to-THz conversion efficiency in the two schemes should be comparable, yielding spectral brightness

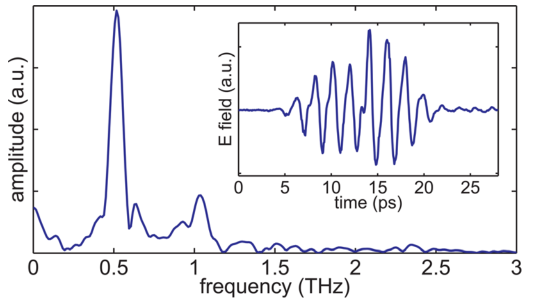

FIG. 4. (Color online) A THz field trace measured through electro-optic sampling and the normalized Fourier spectrum, generated with a 35 mJ near-infrared chirp-and-delay tilted intensity front waveform at 10 Hz repetition rate.

approximately proportional to the number of cycles in the THz pulse.[23] Cooling of the LN crystal to reduce THz attenuation[24] could provide further improvements in conversion efficiency and THz tuning range.

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[1] K.-L. Yeh, M. C. Hoffmann, J. Hebling, and K. A. Nelson, Appl. Phys. Lett. **90**, 171121 (2007).
[2] J. Hebling, K.-L. Yeh, M. C. Hoffmann, and K. A. Nelson, IEEE J. Sel. Top. Quantum Electron. **14**, 345 (2008).
[3] M. C. Hoffmann, J. Hebling, H. Y. Hwang, K.-L. Yeh, and K. A. Nelson, J. Opt. Soc. Am. B. **26**, A29 (2009).
[4] M. C. Hoffmann, N. C. Brandt, H. Y. Hwang, K.-L. Yeh, and K. A. Nelson, Appl. Phys. Lett. **95**, 231105 (2009).
[5] M. Jewariya, M. Nagai, and K. Tanaka, Phys. Rev. Lett. **105**, 203003 (2010).
[6] G. P. Wiederrecht, T. P. Dougherty, L. Dhar, K. A. Nelson, D. E. Leaird, and A. M. Weiner, Phys. Rev. B **51**(4), 916 (1995).
[7] T. F. Crimmins, N. S. Stoyanov, and K. A. Nelson, J. Chem. Phys. **117**, 2882 (2002).
[8] T. Feurer, N. S. Stoyanov, D. W. Ward, J. C. Vaughan, E. R. Statz, and K. A. Nelson, Annu. Rev. Mater. Res. **37**, 317 (2007).
[9] R. M. Koehl and K. A. Nelson, J. Chem. Phys. **114**, 1443 (2001).
[10] T. Feurer, J. C. Vaughan, and K. A. Nelson, Science **299**, 374 (2003).
[11] J. Hebling, G. Almasi, I. Kozma, and J. Kuhl, Opt. Express **10**, 1161 (2002).
[12] T. Feurer, J. C. Vaughan, T. Hornung, and K. A. Nelson, Opt. Lett. **29**, 1802 (2004).
[13] D. W. Ward, J. D. Beers, T. Feurer, E. R. Statz, N. S. Stoyanov, and K. A. Nelson, Opt. Lett. **29**, 2671 (2004).
[14] H. Kawashima, M. W. Wefers, and K. A. Nelson, Ann. Rev. Phys. Chem. **46**, 627 (1995).
[15] K.-H. Lin, C. A. Werley, and K. A. Nelson, Appl. Phys. Lett. **95**, 103304 (2009).
[16] K.-L. Yeh, J. Hebling, M. C. Hoffmann, and K. A. Nelson, Opt. Comm. **13**, 3567 (2008).
[17] A. S. Weling and D. H. Auston, J. Opt. Soc. Am. B **13**, 2783 (1996).
[18] R. Yano and H. Gotoh, Jpn. J. Appl. Phys. **44**, 8470 (2005).
[19] J. R. Danielson, A. D. Jameson, J. L. Tomaino, H. Hui, J. D. Wetzel, Y.-S. Lee, and K. L. Vodopyanov, J. Appl. Phys. **104**, 033111 (2008).
[20] S. Bielawski, C. Evain, T. Hara, M. Hosaka, M. Katoh, S. Kimura, A. Mochihashi, M. Shimada, C. Szwaj, T. Takahashi, and Y. Takashima, Nature Phys. **4**, 390 (2008).
[21] J. A. Fülöp, L. Pálfalvi, G. Almási, and J. Hebling, Opt. Express **18**, 12311 (2010).
[22] A. A. Maznev, T. F. Crimmins, and K. A. Nelson, Opt. Lett. **23**, 1378 (1998).
[23] A. Weling and T. F. Heinz, J. Opt. Soc. Am. B **16**, 1455 (2004).
[24] J. Hebling, A. G. Stepanov, G. Almási, B. Bartal, and J. Kuhl, Appl. Phys. B: Lasers Opt. **78**, 593 (2004).