

Generation of terahertz pulses with arbitrary elliptical polarization

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We employ two different methods to generate controllable elliptical polarization of terahertz (THz) pulses. First, THz pulses are generated via optical rectification in nonlinear crystals using a pair of temporally separated and perpendicularly polarized optical pulses. The THz ellipticity is controlled by adjusting the relative time delay and polarization of the two optical pulses. We generate mixed polarization states of single-cycle THz pulses using ZnTe, and elliptically polarized multicycle THz pulses in periodically poled lithium niobate crystals. Second, we generate elliptically polarized THz pulses by making a THz “wave plate” using a combination of a wire-grid THz polarizer and a mirror to transform linearly polarized multicycle THz pulses into elliptical polarization. © 2005 American Institute of Physics. [DOI: 10.1063/1.2138351]

Studies of the interaction of coherent terahertz (THz) pulses with matter, up to the present time, have generally employed linearly polarized THz waves. A wide range of new experiments, however, would be enabled if THz sources with controlled and adjustable elliptical polarization states were available. Examples for which the THz ellipticity is critical include the control of excitons in semiconductor nanostructures¹ and of molecular rotational wave packets.² Elliptically polarized THz is also potentially important to study macromolecular chiral structures such as proteins and DNA because of strong absorption in the THz regime due to collective vibrational modes.³ In spite of the potential importance, only a few efforts to control the polarization of THz pulses have been reported so far.^{4,5} The lack of established methods for generating elliptically polarized THz waves is partly because the most common and powerful sources such as photoconductive dipole antennas or optical rectification of ultrafast laser pulses in $\chi^{(2)}$ media naturally generate linearly polarized THz radiation; additionally, the majority of THz sources using femtosecond lasers produce single-cycle pulses and the ellipticity is not well defined for the wave form. On the other hand, ellipticity is well defined for multicycle pulses. In this letter, we describe the demonstration of elliptical polarization evolution in single-cycle THz pulses generated in ZnTe using pairs of linearly polarized and time-delayed femtosecond laser pulses. We also extend our recently developed technique for generating narrow-band multicycle THz pulses by optical rectification in periodically-poled lithium niobate (PPLN) crystals,^{6–8} to generate elliptically polarized multicycle THz pulses. We also demonstrate a scheme to transform multicycle THz pulses from linear to elliptical polarization using a combination of a free-standing wire-grid (WG) THz polarizer and a high reflector.

The light source for these experiments was a 76 MHz Ti:sapphire oscillator (Coherent inc., Mira 900F) producing 820 nm, 85 fs pulses. The femtosecond pulses were focused to a spot of 20 μm diameter in the nonlinear crystals. We detected the THz radiation using established electro-optic

sampling techniques: THz radiation was collimated with a pair of off-axis parabolic mirrors and focused into a 1 mm ZnTe crystal for electro-optic detection of the THz fields.⁹ The excitation beam was modulated at 1 kHz for lock-in signal detection. The instantaneous THz polarization was determined by separately measuring the two linear components of THz E -field vector.

In the two-pulse scheme, we control the relative phase of two linearly polarized THz pulses by adjusting the path length difference of the two optical pulses used to generate them. The ellipticity of the total THz field is thus a function of the relative time delay, intensity, and polarization angle of the two incident optical pump pulses. Even though the ellipticity is not well defined for broadband single-cycle THz pulses, we still can apply the two-pump-pulse scheme to generate broadband THz pulses of mixed polarization in ZnTe crystals. Since the crystal class of ZnTe is $\bar{4}3m$, the second order nonlinear susceptibility has six nonzero elements, of which only 1 is independent. Accordingly, the THz field is $\mathbf{E}_{\text{THz}} = \gamma(E_y E_z, E_z E_x, E_x E_y)$ in the crystal coordinate where the applied optical field is $\mathbf{E}_{\text{opt}} = (E_x, E_y, E_z)$ and γ is a coefficient proportional to the second order nonlinear susceptibility, d_{14} . We aligned the optical pump pulses to propagate in the $[1\bar{1}0]$ direction of the ZnTe crystal. The electric fields of the two perpendicularly polarized optical pulses can be written as

$$\mathbf{E}_1(t) = \frac{E_O(t)}{2}(1, 1, \sqrt{2}),$$

$$\mathbf{E}_2(t) = \frac{E_O(t - \tau)}{2}(1, 1, -\sqrt{2}),$$

where τ is the relative time delay between the optical pulses. Then, the generated THz fields are

$$\mathbf{E}_1^{\text{THz}}(t) = \frac{E_T(t)}{\sqrt{5}}(\sqrt{2}, \sqrt{2}, 1),$$

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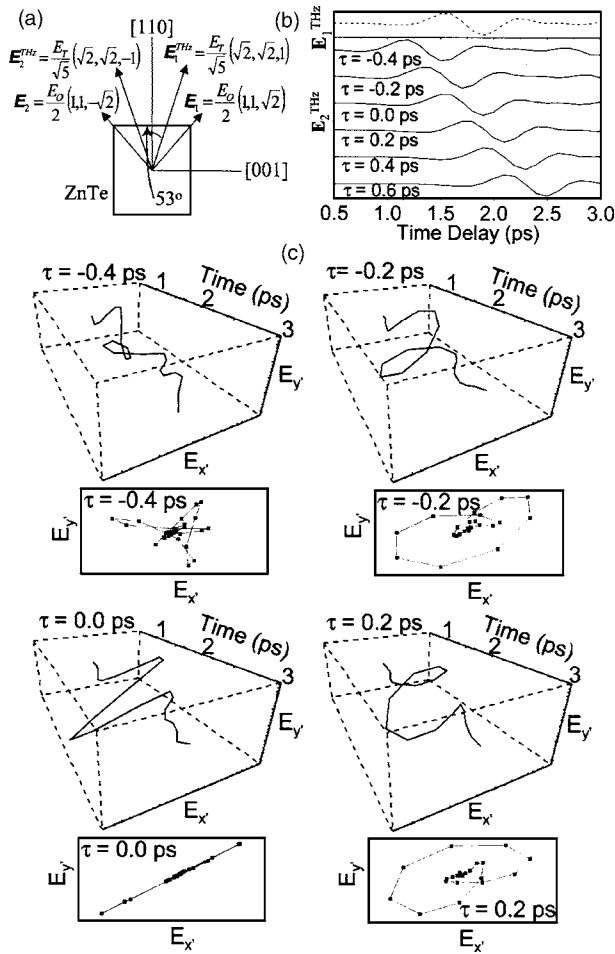


FIG. 1. Mixed polarization states of single-cycle THz pulses. (a) Optical and THz electric fields in ZnTe (110) plane. (b) Time-dependent wave forms of two linearly polarized THz pulses: the measured angle between the two THz fields is $51^\circ \pm 2^\circ$. (c) THz E -field vectors and corresponding polarization trajectories of the superposed THz fields.

$$\mathbf{E}_2^{\text{THz}}(t) = \frac{E_T(t - \tau)}{\sqrt{5}}(\sqrt{2}, \sqrt{2}, -1).$$

The arrangement of the optical and THz fields is described in Fig. 1(a). The calculated angle between the two THz field vectors is $2 \tan^{-1} \frac{1}{2} = 53^\circ$.

The measured THz fields are shown in Fig. 1(b), revealing the temporal evolution of the two THz fields. The first THz pulse ($\mathbf{E}_1^{\text{THz}}$) is featured in the top panel, and the other ($\mathbf{E}_2^{\text{THz}}$) is shown below for various relative time delays ($\tau = -0.4, 0.2, 0.0, 0.2, 0.4, \text{ and } 0.6$). The measured angle between the two THz polarizations is $51^\circ \pm 2^\circ$, close to the calculated value of 53° . Figure 1(c) shows the three-dimensional (3D) plots of the THz E -field vectors, and the corresponding polarization trajectories (projection of the E -field vector on the x' - y' plane) of the superposed THz fields in the laboratory coordinate: the THz fields are in the x' - y' plane and x' axis is in the direction of $1/\sqrt{5}(\sqrt{2}, \sqrt{2}, 1)$. The time interval between the polarization trajectory data points is 0.1 ps. At zero time delay $\tau=0.0$, the superposed THz field is evidently linearly polarized. For other time delays, the THz fields have complicated polarization trajectories. When the time delays are of the same magnitude but with opposite sign, the trajectories have the same pattern, but are rotated by 180 deg in the x' - y' plane, e.g., $\tau=-0.2$ and

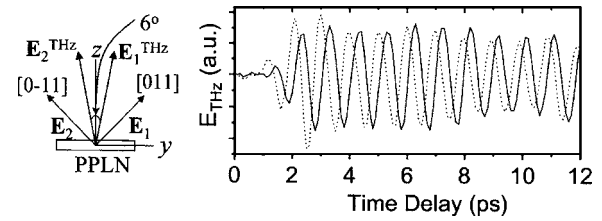


FIG. 2. Elliptically polarized narrow-band multicycle THz pulses. Two linearly and perpendicularly polarized optical pulses generate THz pulses $\mathbf{E}_1^{\text{THz}}$ (dotted line) and $\mathbf{E}_2^{\text{THz}}$ (solid line) in a PPLN crystal. The time delay (τ) between the optical pulses corresponds to 106° of the phase difference ($\phi = 2\pi\nu_{\text{THz}}\tau$, $\nu_{\text{THz}}=1.015$ THz) between the THz pulses. The angle between the polarizations of the THz pulses is 6° .

0.2. For $\tau = \pm 0.2$, the THz E -field vectors show a spiral pattern. Since the mean frequency of the single-cycle THz pulse is about 1.2 THz, $\tau = \pm 0.2$ corresponds to the relative phase of $\pm\pi/2$ at that frequency. The spiral pattern illustrates how the evolution of the electric field vector for a single-cycle pulse can lead to situations where the polarization ellipticity becomes ill defined.

In order to generate elliptically polarized multicycle THz pulses, we applied the same scheme of double-pulse excitation to optical rectification in a PPLN crystal. Figure 2 shows the polarization combination of the two linearly polarized optical and THz pulses, and corresponding THz wave forms. In the case of z -cut PPLN, this scheme works with the proper selection of the azimuthal angle of the crystal. The optical pump pulses propagate in the $[100]$ direction of the PPLN crystal. The optical pulses \mathbf{E}_1 and \mathbf{E}_2 are polarized along the $[011]$ and $[0-11]$ directions, respectively. The electric fields of the optical pulses are described as

$$\mathbf{E}_1(t) = \frac{E_O(t)}{\sqrt{2}}(0, 1, 1),$$

$$\mathbf{E}_2(t) = \frac{E_O(t - \tau)}{\sqrt{2}}(0, -1, 1),$$

where τ is the relative time delay between the optical pump pulses. Each optical pulse generates a linearly polarized multicycle THz pulse centered at 1.015 THz. The crystal class of LiNbO_3 is $3m$, so there are three independent elements of the second order nonlinear susceptibility: d_{22}, d_{31} , and d_{33} . The generated THz pulses can then be described as

$$\mathbf{E}_1^{\text{THz}}(t) = E_T(t)(0, d_{22} + 2d_{31}, d_{31} + d_{33}),$$

$$\mathbf{E}_2^{\text{THz}}(t) = E_T(t - \tau)(0, d_{22} - 2d_{31}, d_{31} + d_{33}).$$

Thus the angle between the polarizations of the THz pulses $\mathbf{E}_1^{\text{THz}}$ and $\mathbf{E}_2^{\text{THz}}$ is

$$\theta = \tan^{-1}\left(\frac{d_{22} + 2d_{31}}{d_{31} + d_{33}}\right) - \tan^{-1}\left(\frac{d_{22} - 2d_{31}}{d_{31} + d_{33}}\right).$$

The experimentally measured angle using a WG THz polarizer is 6° . The small value of the measured angle indicates that the ellipticity of the superposed THz waves is weak, and the polarization is almost linear. In the optical regime, the nonlinear susceptibility tensor components are given as $d_{22}=7.4$, $d_{31}=14$, and $d_{33}=98(10^{-9} \text{ esu})$,¹⁰ which corresponds to 28° for the calculated angle between the nonlinear polarizations. The smaller angle in the THz regime

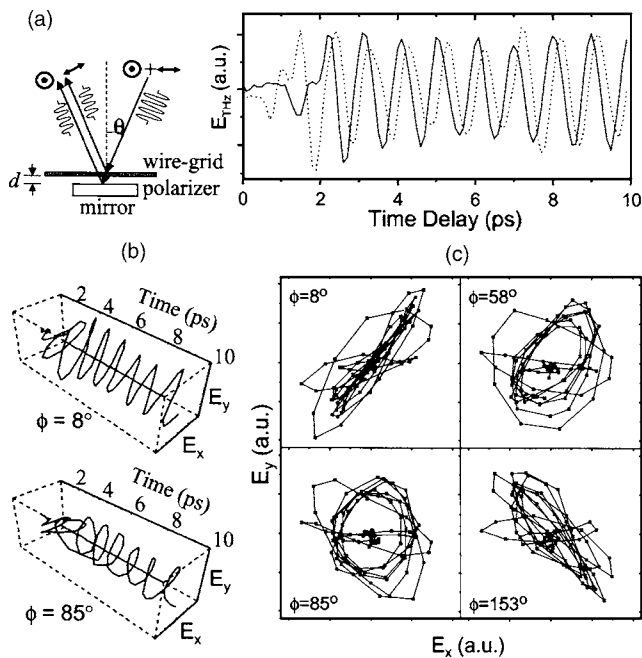


FIG. 3. THz wave plate consisting of WG polarizer and mirror controls THz ellipticity of narrow-band multicycle THz pulses. (a) Two linearly and perpendicularly polarized THz pulses E_x (dotted line: reflection from the WG polarizer) and E_y (solid line: reflection from the mirror) are shown. The phase difference between the THz pulses at $\nu_{\text{THz}} = 1.015$ THz is $\phi = -78^\circ$. (b) 3D plots of THz E-field vectors for $\phi = 8^\circ$ and 85° . (c) Polarization trajectories in the x - y plane for $\phi = 8^\circ, 58^\circ, 85^\circ$, and 153° .

indicates that the relative nonlinearity in the direction of the optic axis is stronger in the THz regime than in the optical regime: $d_{33} \gg d_{22}, d_{31}$. Since $\theta \approx 4d_{31}/d_{33}$, the measurement leads to $d_{31}/d_{33} \approx 0.03$ in the THz regime. The time delay (τ) is 0.29 ps corresponding to 106° of the phase difference between the THz waves at 1.015 THz. The superposed THz pulse can be written as

$$\hat{z}' E_{z'}^{\text{THz}} \cos(kx - \omega_{\text{THz}}t) + \hat{y}' E_{y'}^{\text{THz}} \cos(kx - \omega_{\text{THz}}t + \phi),$$

where $E_{y'}^{\text{THz}}/E_{z'}^{\text{THz}} = 0.088$ and $\phi = 53^\circ$. \hat{z}' and \hat{y}' are orthonormal unit vectors and \hat{z}' is parallel to the polarization of the THz pulse $\mathbf{E}_1^{\text{THz}}$.

Due to the relatively strong nonlinearity along the optic axis, the two-pulse scheme is not a very efficient way to generate circularly polarized THz waves. An alternative method to control the THz ellipticity is illustrated in Fig. 3(a). Elliptical polarization can be obtained from the reflection of a linearly polarized THz wave by a combination of a WG polarizer and a mirror. The mirror is placed just behind the polarizer. The mirror and the polarizer reflect two perpendicularly polarized THz waves. The THz “wave-plate” (WG polarizer+mirror) controls the ellipticity of the combined THz waves by adjusting the distance between the mirror and the polarizer and by rotating the polarizer. The THz wave form shown in Fig. 3(a) is characterized as

$$\hat{x} E_x^{\text{THz}} \cos(kz - \omega_{\text{THz}}t) + \hat{y} E_y^{\text{THz}} \cos(kz - \omega_{\text{THz}}t + 2\pi + \phi),$$

where $E_x^{\text{THz}}/E_y^{\text{THz}} = 1.0$, $\omega_{\text{THz}}/2\pi = 1.015$ THz and $\phi = -78^\circ$. The phase resolution at 1.015 THz is $\Delta\phi = 0.25^\circ$.

Figure 3(b) shows the THz E-field vectors of the multicycle narrow-band THz pulses in the x - y plane for $\phi = 8^\circ$ and 85° . The polarization for $\phi = 8^\circ$ is almost linear except the

first two periods where the two THz pulses only partially overlap. Since the reflection from the WG polarizer—linearly polarized in the x axis—arrives earlier, the trajectory shows the 45° rotation from the x axis in the first few picoseconds. The early time rotations are also shown in Fig. 3(c): the polarization trajectories in the x - y plane. As the phase increases, the polarization changes continuously: linear(8°) \rightarrow elliptical(58°) \rightarrow circular(85°) \rightarrow elliptical(135°).

In conclusion, we have demonstrated two techniques to generate elliptically polarized THz pulses. One utilizes optical rectification in nonlinear crystals using two temporally separated optical pump pulses. We control the THz ellipticity adjusting the relative time delay and polarization of the two optical pulses. We generate mixed polarization states of single-cycle THz pulses using ZnTe, and elliptically polarized multicycle THz pulses in PPLN crystals. The second is essentially a broadband THz wave-plate using a combination of a wire-grid THz polarizer and a mirror to transform linearly polarized multicycle THz pulses into elliptical polarization. The wire-grid THz wave plate is applicable to the entire THz spectral range. In general, the wave-plate method should be more practical than the two-pulse technique because the set-up is simpler and the polarization control is more flexible. We expect that THz ellipticity control techniques may be useful for coherent control of carrier dynamics in semiconductor nanostructures and of molecular dynamics. For example, single-cycle THz pulses of spiral polarization might be useful for exploring quantum chaos of the transiently kicked quantum rotor.¹¹ This problem has been rigorously studied theoretically, but only a few experimental studies have been done in atomic systems.^{12,13} A spirally polarized single-cycle THz pulse could be used as a transient rotational impact on gas phase heteropolar molecules, which are nearly ideal quantum rotors. THz time-domain spectroscopy, which can measure not only the amplitude of the rotational motion but also the phase, would be an excellent experimental scheme to investigate the quantum wavepacket dynamics.

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