Generation of largely elliptically polarized terahertz radiation from laser-induced plasma

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Abstract: A novel all-optical control scheme is proposed to continuously tune the THz radiation polarization, where the driving laser is based on a three-pulse configuration with adjustable time delays or intensity ratio. With this scheme, not only is the circularly polarized THz radiation realized, the continuous tuning from circular polarization to linear polarization can also be obtained conveniently just by adjusting time delays or intensity ratio. Moreover, the left or the right chirality of THz radiation can be transformed between each other with suitable time delays.

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References and links

1. Introduction

Broadband ultra-short terahertz (THz) pulses can be generated from laser-induced gas plasma by a two-color field ($\omega$ and 2$\omega$) with the potential to achieve a high intensity and broad bandwidth THz field beyond the damage threshold of bulk materials [1–5]. As the 2$\omega$ pulse overlaps with the $\omega$ pulse, the superposed asymmetric optical field drifts the photoelectrons to produce the directional transient photocurrent, which emits enhanced broadband THz radiation [5]. The waveform of generated THz radiation is mainly determined by the intense driving laser and the complicated spatiotemporal dynamics within filaments [6, 7]. Among the above studies, the polarization state of the generated THz radiation is one of the key concerns. One reason is that an elliptically polarized THz wave with a large ellipticity is demanding in the fields of imaging the macromolecular chiral structures of proteins and DNA [8], probing the spin dynamics in solid state materials [9], providing insights on the underlying physics of laser filamentation [10–12] and so on.

As for the THz waves generated from two-color laser-induced gas plasma, the polarization state of THz emission can be optically controlled by varying the relative phase or polarization states of the driving two-color laser field [13–15]. In addition, the proof-of-principle experiments with a two-color pulse have been also demonstrated to control the THz polarization direction when the phase difference of $\omega$ and 2$\omega$ pulse changes [13, 14]. Such a polarization rotation is attributed to the manipulated electron trajectories by the two-color field. However, the corresponding ellipticity variation has not been clarified. You et al [15] investigated the mechanism of elliptically polarized THz emission in two-color laser-induced plasma and found that the THz polarization evolves from linear to limited elliptical by increasing the plasma length, owing to the combined effects of polarization rotation of local plasma and the velocity mismatch of laser and THz. Chen et al [16] measured the polarization...
of THz radiation by setting the polarization of \( \omega \) and \( 2\omega \) orthogonal, and found that the ellipticity can increase from 0.1 to 0.4 with the pulse energy increasing from 0.2 mJ to 1.6 mJ, together with the rotation of polarization orientation. However, as for a two-color laser field control, the polarization state of the generated THz wave is always close to linear, i.e., its ellipticity being much smaller than one [13–15]. So far, all-optical control of THz radiation ellipticity, especially to obtain a circularly polarized THz wave from laser-induced plasma, is still a challenge.

In this work, a novel polarization control scheme based on a three-pulse configuration with adjustable time delays and intensity ratio is proposed. The results indicate that a full control of the ellipticity of THz emission can be realized, and the polarization state can be tuned from circular to linear by adjusting the time delays. Circularly polarized THz wave is possible as long as suitable time delays and intensity ratio of three pulses are chosen. Besides, the chirality of THz emission can be controlled with suitable time delays.

2. Three-pulse configuration and numerical model

We introduce a three-pulse configuration for the driving laser propagating along the \( z \) direction, which is formally similar to the general two-color pulse configuration [13, 17]: a \( 2\omega \) pulse is superposed to a \( \omega \) pulse, but here as for the fundamental \( \omega \) pulse, suitable time delays and intensity ratio are included to its two orthogonal components in the \( x\)-\( y \) plane. In some sense, the three-pulse configuration is a suitable transformation from the general two-color one. This transformation can be conveniently realized experimentally by introducing another \( \omega \) pulse to a general two-color field [13] with a suitable time delay. The driving laser components thus can be written as,

\[
E_x(t) = E_{\omega x} \exp \left[ -2 \ln 2 \frac{(t-t_{dx})^2}{\tau_{\omega}^2} \right] \cos \left[ \omega (t-t_{dx}) + \varphi \right],
\]

\[
E_y(t) = E_{\omega y} \exp \left[ -2 \ln 2 \frac{(t-t_{dy})^2}{\tau_{\omega}^2} \right] \cos \left[ \omega (t-t_{dy}) + \varphi \right]
\]

here, a Gaussian pulse profile is assumed with a pulse duration of \( \tau_{\omega} \) (full width at half maximum (FWHM) of laser intensity of fundamental pulse \( \omega \)) and a peak amplitude \( E_i \) \( (i = \omega x, \omega y, 2\omega) \). The fundamental frequency is \( f_\omega = \omega / 2\pi \approx 375 \text{ THz} \) at wavelength 800 nm and its second harmonic frequency is \( f_{2\omega} = 750 \text{ THz} \). \( t_{dx} \) and \( t_{dy} \) are the adjustable time delays, achieved by an inline phase compensator [13], between the two orthogonal components of \( \omega \) field and \( 2\omega \) component, respectively. Thus, \( \varphi_r = \varphi + \omega (t_{dy}-t_{dx}) \) is the phase retardation between \( x \) and \( y \) components of \( \omega \) field, and \( \theta_r = \theta + 2\omega t_{dx} \) is the phase difference between the \( \omega \) and \( 2\omega \) along \( x \) axis, which both can be adjusted just by inserting a fused silica wedge pair conveniently. It should be noted that \( t_{dx} \) and \( t_{dy} \) are two key parameters as shown in follows for the generation of a largely elliptically polarized THz wave.

Under this three-pulse configuration for a driving laser, the THz emission is evaluated based on the transient photocurrent model in the local current (LC) limit [5, 10]. And tunneling ionized electrons result in a two-dimensional (2D) directional photocurrent \( J_e \) [11] given by \( J_e = J_{ex} \hat{x} + J_{ey} \hat{y} \), where \( J_{ex} \) and \( J_{ey} \) are the current density components directed along the \( x \) and \( y \) axis, respectively.
\[ \frac{\partial_t J_{e\omega}}{e} + \nu_e J_{e\omega} = \frac{e^2}{m} \rho_e E, \\]
\[ \frac{\partial_t J_{e\omega}}{e} + \nu_e J_{e\omega} = \frac{e^2}{m} \rho_e E, \]

here, \( \nu_e \) is the phenomenological electron-ion collision rate (\( \nu_e = 1/190 \text{ fs}^{-1} \) [10] at atmospheric pressure). \( e, m \) and \( \rho_e \) denote electron charge, electron mass, and electron density, respectively. The electron density \( \rho_e \) is determined by the ionization rate equation \( \partial_t \rho_e(t) = W(t) \left[ \rho_a - \rho_e(t) \right] \), with \( \rho_a = 2.7 \times 10^9 \text{ cm}^{-3} \) being the initial neutral gas density at atmospheric pressure and \( W(t) \) indicating the static tunneling ionization rate given by the ADK theory [3]:

\[ W(t) = 4\omega_a \left( \frac{U_{N_2}}{U_H} \right)^{5/2} \left[ \frac{E_a}{E_L(t)} \right] \exp \left( -\frac{2E_a}{3E_L(t)} \left( \frac{U_{N_2}}{U_H} \right) \right)^{3/2} \]  

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where \( E_L(t) = \sqrt{E_\omega^2(t) + E_2^2(t)} \) is the driving laser, \( \omega_a = 4.134 \times 10^{16} \text{ s}^{-1} \) is the atomic frequency, \( E_a = 5.14 \times 10^9 \text{ V/cm} \) is the atomic field amplitude, and \( U_H \) and \( U_{N_2} \) are ionization potentials of hydrogen atoms and nitrogen molecules, respectively. Here, for a general description convenience, the plasma recombination and electron attachment of neutral molecules and other trivial effects are ignored because those events functionalize much slowly compared to the pulse duration considered [17, 18]. After obtaining the current \( J_e \) from Eq. (1), the generated THz field is given by [5]

\[ E_{\text{THz}} \propto \frac{dJ_e}{dt}. \]

It has been found both theoretically and experimentally that there basically are two physical mechanisms for terahertz radiation generation in a femtosecond plasma filament: one lies in the nonlinear polarization of neutrals and the other in the free electron photocurrent [19–21]. The well description based on the instantaneous third-order nonlinear response of neutral molecules in air in the strong field [20] shows a good agreement with the simulations with the THz spectral peaks at the tens of THz [19–21]. However, here the concerned frequency range is near to zero, that is, the so-called low-frequency range of several THz, which is guaranteed by a filter with a cutoff frequency of 5 THz for the THz time-domain spectroscopy (THz-TDS) [22], then the contribution from the free-electron photocurrent dominates. This is the reason why we have chosen the photocurrent model as the basis of the theoretical simulation.

Moreover, in our simulation, both the relative phase between \( \omega \) and \( 2\omega \) and the refractive indexes of \( \omega \) and \( 2\omega \) are safely assumed constant over the entire plasma length of typically less than 10 mm, as shown experimentally [1, 2, 4, 5, 23]. Compared with the cases of long plasma filaments ranging from a few centimeters to tens of meters, each point of which along the propagation axis serving as key mechanisms responsible for pushing the THz emission off axis and leading to ring formation in the far field [21, 24], the \( \omega-2\omega \) phase mismatch and plasma dispersion are not dominated phenomenologically in a low dispersion gas such as air. At the same time, the velocity mismatch between optical and THz pulses in plasma is negligible as well [15], therefore we can eliminate their influences on THz polarization. Additionally, it is not necessary to consider the filament dynamics and unidirectional pulse propagation effects [10]. Thus, the theoretical model here does really not include the propagation effects and thus also without the corresponding pulse nonlinear transformation effects [25, 26].
3. Simulated results and discussions

In the following numerical demonstration, the fundamental pulse intensity is set as $2 \times 10^{14}$ W/cm$^2$, which should be strong enough to ensure the generation of a sufficient amount of electrons to keep a reasonable conversion efficiency to THz radiation from driving fields. As for its second harmonic, the intensity is around $4 \times 10^{13}$ W/cm$^2$ if a 20% frequency-doubling conversion efficiency is assumed. First, one can assume $|t_{dx}| = |t_{dy}| = 20$ fs (half of $\tau_{s}$) as well as a suitable intensity ratio among the $x$ and $y$ components of $\omega$ field and the $2\omega$ component $r = (E_{\omega})^2 : (E_{2\omega})^2 = 1 : 3 : 0.2$, and phase parameters $\varphi_{r} = 268^\circ$ and $\theta_{r} = 0^\circ$, as shown in Figs. 1(a) and (b), the THz polarization state is circular. In addition, with other time delays (also as shown in Fig. 2), a largely elliptically polarized THz with ellipticity larger than 0.5 can be realized, which is impossible in other all-optical control schemes [1-2, 4-5, 13-15, 17, 27]. Both the theoretical and experimental results show that the polarization of emitted THz waves are linear or close to linear when $\omega$ and $2\omega$ beams are both linearly polarized or when $\omega$ and $2\omega$ beams are circularly or elliptically polarized [13, 14, 20]. In all these cases, only the field amplitudes and the relative phase difference can be adjusted. Instead here, the time delays are introduced to three components of laser fields. Due to the time delays not being zero, a formal three-pulse configuration is formed beyond the general two-color configuration [1-2, 4-5, 13-15, 17, 27]. Interestingly, as shown in the following, with suitable time delays and amplitude ratio combination, any elliptically polarized THz wave can be realized, where a continuous variation of time delays of three field components can lead to the continuous tuning of THz ellipticity. Another reason is that at the THz generation experiments in the above references, only the azimuthal angles of the BBO crystal for the second harmonic generation (SHG) can be adjusted to optimize the efficiency of THz conversion, which means that the intensity ratio is optimized to a higher THz yield but with a smaller ellipticity.

More interesting thing is to control the handedness of THz wave. When $t_{dx} = +20$ fs, $t_{dy} = -20$ fs, i.e. the $x$ component of $\omega$ field propagates faster than the $y$ component, thus the chirality of the circularly polarized THz waves is left-handed, as shown in Fig. 1 (a). In contrast, when $t_{dx} = -20$ fs, $t_{dy} = +20$ fs, i.e. the $y$ component of $\omega$ filed is faster than the $x$ component, the opposite chirality of THz radiation is obtained as shown in Fig. 1(b). The accordingly circular polarization trajectories on the $x$-$y$ plane are both well fitted to a circle with an ellipticity (defined as the ratio of the THz amplitudes along minor and major axes) of $e = 1$. In fact, it satisfies the two characteristics of circularly polarized beams, i.e. the amplitude ratio $E_{x}^{\text{THz}} / E_{y}^{\text{THz}} = 1$ and the phase difference between $x$ and $y$ components $\varphi_{x}^{\text{THz}} / \varphi_{y}^{\text{THz}} = \pm \pi / 2$. Note that the elliptically polarized THz field in the laboratory coordinate can revolve anticlockwise (left-handed) or clockwise (right-handed). One can see the handedness of an elliptically polarized THz generated here can be manipulated by only adjusting the sign of the time delays. As a direct comparison, when there are no time delays [13–15], i.e. $t_{dx} = 0$ fs, $t_{dy} = 0$ fs, a linearly polarized THz wave generated as shown in Fig. 1(c).

Second, in the above demonstration, $|t_{dx}| = |t_{dy}|$ is kept. Now if breaking this limit to an asymmetric form, one cannot acquire a circularly polarized THz, and an elliptically polarized
THz wave occurs instead, as shown in Fig. 2(a), when time delays set as $t_{dx} = 30, 25, 20, 15, 10$ fs in turn, and correspondingly $t_{dy} = -10, -15, -20, -25, -30$ fs with other parameters same with those in Fig. 1.

From Fig. 2 (a), it can be seen that the closer the two time delays are, the more equal the amplitudes of the two orthogonal components one can obtain, and the larger the ellipticity of the THz emission is. Only when the time delays are symmetric, i.e. $|t_{dx}| = |t_{dy}|$, the circularly polarized THz pulse can be achieved.

Additionally, as shown in Fig. 2(b), when the time delays are equal with different signs, i.e. $t_{dx} = -t_{dy}$, one time delay increased from $-23$ fs to $23$ fs, the relationship between THz ellipticity and the time delay clearly demonstrates this point. The ellipticity increases as the $|t_{dx}|$ increases, and when $t_{dx} = \pm 20$ fs, i.e. $t_{dx} = 0.5 \tau_\omega$, the ellipticity reaches maximum $e = 1$. After that, the ellipticity decreases. The polarization state can be tuned from circular to linear by adjusting the time delays. The maximum THz ellipticity from the simulation results could not completely be one owing to the simulation data treatment method. Therefore, the time delays of introducing a supplemental pulse to the two-color field can be applied to realize the full control of the THz polarization, especially a large elliptically polarized THz wave.

Besides the time delays, the intensity ratio of the three pulses, i.e., $r = (E_{x0})^2 : (E_{y0})^2 : (E_{2\omega})^2$, can be also tuned freely. This can be achievable in experiments when intensities of the three pulses can be adjusted with neutral absorption-type density filters. One found that the circularly polarized THz pulse can be also acquired through adjusting intensity ratio even though with the above asymmetric time delays. The intensity ratio plays a critical role on the amplitudes of two THz components, because it determines the ionization rate and then determines the electron density. For example, when $t_{dx} = +22$ fs, $t_{dy} = -18$ fs, $(E_{x0})^2 : (E_{y0})^2 : (E_{2\omega})^2 = 1:2.88:0.205$, and $t_{dx} = +25$ fs, $t_{dy} = -15$ fs, $(E_{x0})^2 : (E_{y0})^2 : (E_{2\omega})^2 = 1:2.66:0.205$, the circularly polarized THz pulse can be also achieved. Figure 3 shows the comparison of circularly and elliptically polarized THz waves on the $x$-$y$ plane with different intensity ratios when $t_{dx} = +25$ fs and $t_{dy} = -15$ fs.
Fig. 3. Largely elliptically polarized THz waves on the x-y plane under the same time delays ($t_{dx} = +25$ fs and $t_{dy} = -15$ fs) but with different intensity ratios.

Last but not least, without time delays, i.e. $t_{dx} = t_{dy} = 0$, the proposed three-pulse configuration should go back to the 2D photocurrent model in two-color photoionization [14, 17], and the yield of THz wave should be much higher than that with time delays, as THz yield is much sensitive to time delays experimentally demonstrated in references [13] and [27]. As for the three-pulse configuration, the relationship between THz yield and time delays is also investigated. As shown in Fig. 4, when the two components of $\omega$ field do not separate away from each other in temporal domain, i.e. $t_{dx} = t_{dy}$, but both separated from $2\omega$ field by within 10 fs, the THz yield does not decrease too much. However, when the two components of $\omega$ field split in time domain, the THz yield sharply declines with the delays increasing. Owing to this, some experiments were carried out by introducing a phase compensator to improve the terahertz generation [13, 27]. According to the simulation result in Fig. 4, one can see that compensating time delays in the experiments is an optimal method to improve terahertz generation.

In addition, the feasibility of the three-pulse scheme for an experimental realization can be proved. In the asymmetric time delays of $t_{dx} = +22$ fs and $t_{dy} = -18$ fs, one found that this situation can be achieved by the general two-color experiments of THz generation which are carried out through applying a type-I beta barium borate ($\beta$-BBO) crystal (usually a BBO crystal thickness is $l = 100 \mu$m) after the focusing lens [5, 13, 17]. And the asymmetric time delays are calculated according to the refractive indexes of the BBO crystal along the extraordinary ($e$) and ordinary ($o$) axes by the Sellmeier equations [28]. In this experimental case of $l = 100 \mu$m, a phase retardation of $\phi_r = 268^\circ$ for the $\omega$ components along $e$ and $o$ axes is acquired [17]. In addition, when the laser polarization angle related to the $e$ axis of the BBO crystal, i.e. azimuthal angle, is $\alpha = 55^\circ$, the magnitude of the THz emission is maximized [5, 13, 17]. Under this fixed azimuthal angle, the $\omega$ pulse can be decomposed into
two orthogonal components polarized along the \(o\) and the \(e\) axes because of the birefringence of the BBO crystal, i.e., \((E_{\omega o})^2: (E_{\omega e})^2 = 1:1.2\). Here, the produced \(2\omega\) by type-I phase matching is polarized along the \(e\) axis. In this special case, as shown in Fig. 5, as the relative phase \(\theta\) changes by \(2\pi\), which can be achieved by moving the position of the BBO crystal experimentally, the major axis of the THz ellipse rotates a complete circle. Simultaneously the polarization direction of the THz pulse rotates counter-clockwise, and the polarization state changes from linear to elliptical. When the relative phase is \(\theta_r = 162^\circ\), the ellipticity of THz waves reaches to the maximum of 0.5 approximately, shown as the violet bold solid line plotted in Fig. 5. When \(\theta_r = 135^\circ\), the polarization of generated THz waves is well fitted to an ellipse with an ellipticity of \(e \approx 0.25\), which is consistent with the previous experimental report [17]. However, further detailed investigation shows that no matter what values of phase retardation \(\phi_r\) of \(E_{\omega x}\) and \(E_{\omega y}\), and phase difference \(\theta_r\) of \(E_{\omega x}\) and \(E_{2\omega x}\) are, the polarization of THz wave cannot be circular at all in this asymmetric time delays and fixed intensity ratio just owing to the intensity ratio compared with that of \(1:2.88:0.205\).

\[
E_x(t) = E_{\omega x} \exp \left[ -2\ln 2 \left( \frac{t-t_{\omega 1}}{\tau_x} \right)^2 \right] \cos \left[ \omega (t-t_{\omega 1}) + \theta_1 \right] + E_{2\omega x} \exp \left[ -2\ln 2 \left( \frac{t-t_{\omega 1}}{\sqrt{2}\tau_x} \right)^2 \right] \cos \left[ 2\omega (t-t_{\omega 1}) + \theta_1 \right],
\]

\[
E_y(t) = E_{\omega y} \exp \left[ -2\ln 2 \left( \frac{t-t_{\omega 1}}{\tau_y} \right)^2 \right] \cos \left[ \omega (t-t_{\omega 1}) + \phi \right] + E_{2\omega y} \exp \left[ -2\ln 2 \left( \frac{t-t_{\omega 1}}{\sqrt{2}\tau_y} \right)^2 \right] \cos \left[ 2\omega (t-t_{\omega 1}) + \phi \right],
\]

(5)

where the meanings of the variables in Eq. (5) are similar to that in Eq. (1). Then there would be more adjustable parameters to influence the THz polarization. For example, one can easily get a largely elliptically polarized THz wave with the ellipticity \(e = 0.77\) as shown in Fig. 6 (a) when \(I_{\omega x}: I_{\omega y}: I_{2\omega x}: I_{2\omega y} = 1:0.72:0.72, t_{\omega 1} = 24\) fs, \(t_{\omega 2} = 20\) fs, \(t_{\omega 1} = -24\) fs, \(t_{\omega 2} = -20\) fs, \(\varphi = \phi + \omega(t_{\omega 1} - t_{\omega 2}) = 0^\circ, \theta_1 = \theta_1 + 2\omega(t_{\omega 1} - t_{\omega 2}) = 0^\circ, \theta_2 = \theta_2 + 2\omega(t_{\omega 1} t_{\omega 2}) = 120^\circ\), i.e., the fundamental field is linear while the second harmonic is elliptical. Even when the fundamental field and the second harmonic are both elliptical, the realization of a large THz ellipticity is still possible. For example, when choosing the parameters of \(I_{\omega x}: I_{\omega y}: I_{2\omega x}: I_{2\omega y} =\)
1:0.64:0.4:0.576, t_{dx1} = 6 fs, t_{dx2} = 6 fs, t_{dy1} = −20 fs, t_{dy2} = −21 fs, φ_r = φ + ω(t_{dx1}−t_{dy1}) = 268°, θ_{r1} = θ_1 + 2ω(t_{dx1}−t_{dy1}) = 0°, θ_{r2} = θ_2 + 2ω(t_{dx1}−t_{dy2}) = 90°, even a circularly polarized THz wave (e=1) is generated as shown in Fig. 6 (c). And the polarization of Fig. 6(a) and (b) and the corresponding fitting curves are shown in Fig. 6(b) and (d), respectively.

4. Conclusions

In conclusion, we proposed all-optical coherent control scheme of the polarization states of ultrafast THz waves using the three-pulse configuration with adjustable time delays and intensity ratio. In the case of the three-pulse configuration of a supplemental pulse introduced to two-color laser fields, the ellipticity and the chirality of THz emission are fully controllable including the generation of circularly polarized THz waves, which requires optimized time delays, optimized intensity ratio. The simulation of three-pulse configuration based on photocurrent model can explain the previous experimental observation of elliptical THz generation and its limitation of higher ellipticity. The finding provides a basis on understanding the experiments of laser-induced gas plasma THz source and the application of waveform-controlled THz emission.

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