Generation of Elliptically Polarized Terahertz Waves from Laser-Induced Plasma with Double Helix Electrodes

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By applying a helical electric field along a plasma region, a revolving electron current is formed along the plasma and an elliptically polarized far-field terahertz wave pattern is observed. The observed terahertz wave polarization reveals the remarkable role of velocity retardation between optical pulses and generated terahertz pulses in the generation process. Extensive simulations, including longitudinal propagation effects, are performed to clarify the mechanisms responsible for polarization control of air-plasma-based terahertz sources.

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Electromagnetic pulses in the terahertz (THz) frequency range play a pivotal role in material spectroscopy, biomedical diagnosis, and homeland security [1]. However, coherently exciting and controlling a circularly polarized THz wave has proved to be a challenge, primarily because most pulsed THz sources are based on optical rectification [2] or transient dipole radiation [3]. Elliptically or circularly polarized THz waves are potentially important to study macromolecular chiral structures, such as proteins and DNA, or to excite spin dynamics in solid state materials. Typically, a Fresnel prism is used as a quarter wave plate for THz waves to switch linear polarization to circular polarization [4]. The introduced material absorption and Fresnel loss limit the bandwidth and intensity of THz waves. The latest advances in plasma-based THz sources [5–12] have made it possible to observe and even control the linear polarization state of intense and broadband THz waves through a coherent manipulation of the motion of ionized electrons electrically [7,12] or optically [13,14]. In all these experiments the trajectory of released electrons, which contributes to the THz polarization state, is determined by the direction of the external field or the relative phase of two-colored optical fields. Although elliptical imperfections of the THz polarization state from these plasma sources have been reported [13,15], insightful physical and theoretical explanations are required for further polarization control and manipulation.

In this Letter, we present a combined theoretical and experimental study of the elliptical polarization properties of generated THz waves from a laser-induced plasma with a pair of helical electrodes. We demonstrate that temporal propagation effects substantially influence the THz polarization and are necessary to understand the THz wave generation process. In both experiments and simulations, we observe remarkable changes of the elliptical THz polarization state with various electrode designs. Such ellipticity is a result of a sensitive dependence on the velocity mismatch between the optical and generated THz pulses. Extensive numerical simulations were performed combining a transient current model with pulse velocity retardation, which includes the longitudinal propagation effects responsible for polarization control of THz waves.

In the laser-induced ionization process, electrons released from molecules or atoms exhibit a net drifting current after passage of the laser pulses when they experience an asymmetric electric field [10,11,16]. This drifting current, which contributes to the far-field THz radiation, is aligned with the direction of an external dc field [12,15] or asymmetric optical field [13,14]. In our experiment with a pair of helical electrodes, the induced net electron current revolves along the filament due to the longitudinal variation of the applied dc electric field. This revolution occurs during the travel of the ionization front, which is associated with the propagation of intense laser pulses. Furthermore, due to the velocity mismatch between the THz wave and optical beam, we find that the produced THz wave will travel ahead of the optical excitation, which will eventually lead to a phase difference between the produced *s* and *p* components of the THz pulse in the far field due to the retarded radiation. We demonstrate that the THz polarization state can be coherently manipulated by varying the electrode design and longitudinal properties of plasma. For example, the handedness can be controlled directly through the control of the handedness of electrodes and the ellipticity can be controlled by electrode design.

A Ti-sapphire amplified laser system, which can deliver laser pulses of 40 fs and 3 mJ at a repetition rate of 1 kHz, is used in this experiment. The laser pulses were split into pump beam and probe beam by a 95% to 5% beam splitter. The optical pump beam was focused by a lens with a 400 mm focal length to produce an ionization region in air of over 40 mm in length. Figure 1 illustrates the

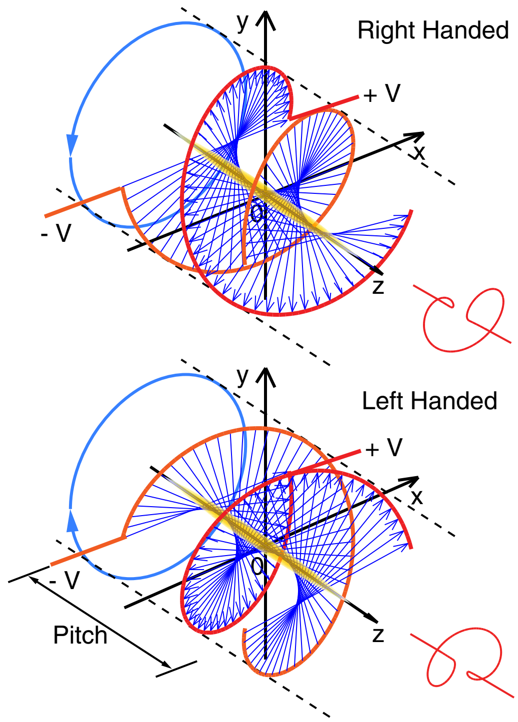

FIG. 1 (color online). Illustration of double helix electrodes and corresponding electric field applied on plasma region with different handedness. The laser pulses are focused to ionize air where a pair of double helix electrodes is positioned. The trajectory of electrons (arrows) follows the external electric field direction, resulting in an elliptically polarized THz wave in the far field. The laboratory coordinate and definition of handedness are shown in the figure. The origin is defined at the center of the plasma.

revolving electric field in the laboratory coordinates. A pair of helical electrodes made from copper wires with a 1 mm diameter were mounted along the plasma region. The two copper wires were twisted for only one period along the grooves on a plastic mount, which has an inner diameter of 4 mm and an outer diameter of 6 mm. The inter spacing is around 5 mm and the length of one pitch is 30 mm. The pitch length was chosen to optimize the ellipticity of produced THz waves under current experimental conditions. The bias field was provided by a high voltage modulator, which delivers bipolar square waves with a frequency of 500 Hz and a tunable amplitude up to 3 kV. The generated THz wave was collected by a 90° off-axis parabolic mirror and then focused again by another parabolic mirror. A 1 mm thick high resistivity silicon wafer was inserted between two parabolic mirrors to block the residual optical beam. The optical probe beam went through a time-delay stage and was focused through a hole in the second parabolic mirror by a lens with 150 mm focal length onto a 1 mm thick (110) ZnTe crystal to resolve the temporal information of THz waves. To analyze the polarization of THz waves, a wire grid THz polarizer was used in the collimated THz beam path. A combination of a half wave plate and a polarizer was inserted into the optical pump and probe beam to rotate their polarizations. The x and y axes in Fig. 1 represent p and s polarizations, respectively. Note that the applied electrical field originates along the x axis in the laboratory coordinate and revolves clockwise or anticlockwise depending on the handedness.

In order to clarify that the effect is related to pump beam polarization, we performed initial measurements by rotating the polarization of the pump beam using a half wave plate. The variation of the peak-to-peak amplitude of the p component of THz radiation was independent of pump beam polarization, in agreement with Ref. [15].

An elliptically polarized wave can be decomposed into an arbitrary set of mutually orthogonal component waves with their polarization perpendicular to each other and with a fixed phase relationship. By recovering the THz electric fields obtained at two orthogonal directions, it is feasible to reconstruct the electron trajectories within the plasma. In our experiment, the polarization of THz waves was obtained by measuring the THz electric fields in two orthogonal directions measured by an electric-optic sampling technique. An elliptical THz polarization trajectory with an ellipticity $e = 0.5$ was observed on the X-Y plane and is shown in Figs. 2(a) and 2(b). In the presence of the external helical electric field, the handedness of elliptically polarized THz waves can be manipulated by the longitudinally revolving direction of the dc field. A THz wave generated from a pair of linear electrodes is also shown in Fig. 2(c) for comparison.

When applying an external bias field to the plasma, the driven motion of electrons will result in a far-field THz radiation whose polarization is aligned to the external electric field direction. The electron density $\rho(t)$ of the plasma, which satisfies $\frac{\partial\rho(t)}{\partial t} = W_{\rm ST}(t)[\rho_0 - \rho(t)]$, where ρ_0 is the molecular gas density. $W_{\rm ST}(t)$ is the ionization rate calculated from the static tunneling model, which can be expressed as, $W_{\rm ST}(t) = \frac{\alpha}{\hat{E}_\omega(t)}\exp\left(-\frac{\beta}{\hat{E}_\omega(t)}\right)$ [17], where $\hat{E}_\omega(t) = E_\omega(t)/E_a$ is the electric field of fundamental beam in atomic units, $E_a = m^2e^5/(4\pi\varepsilon_0)^3\hbar^4$, $\alpha = 4\omega_a r_H^{5/2}$, $\beta = 2r_H^{3/2}/3$, $\omega_a = me^4/(4\pi\varepsilon_0)^2\hbar^3$, $r_H = U/U_H$ is the relative molecular ionization potential normalized with that of hydrogen, and m, and e are the electron mass and charge, respectively. Here, the direction of applied electric field is alternating along the plasma, and can be expressed as: $\vec{E}_{\rm DC}(z) = E_0\cos(\gamma z)\vec{x} \pm E_0\sin(\gamma z)\vec{y}$, where E_0 is the amplitude of electric field and $\gamma = 2\pi/L$ is the alternating frequency which is related to the pitch length of electrodes L. The sign of the second term is determined by the handedness of the electrode structure. For an averaged electron-ion collision time τ, the velocity of electrons born at time

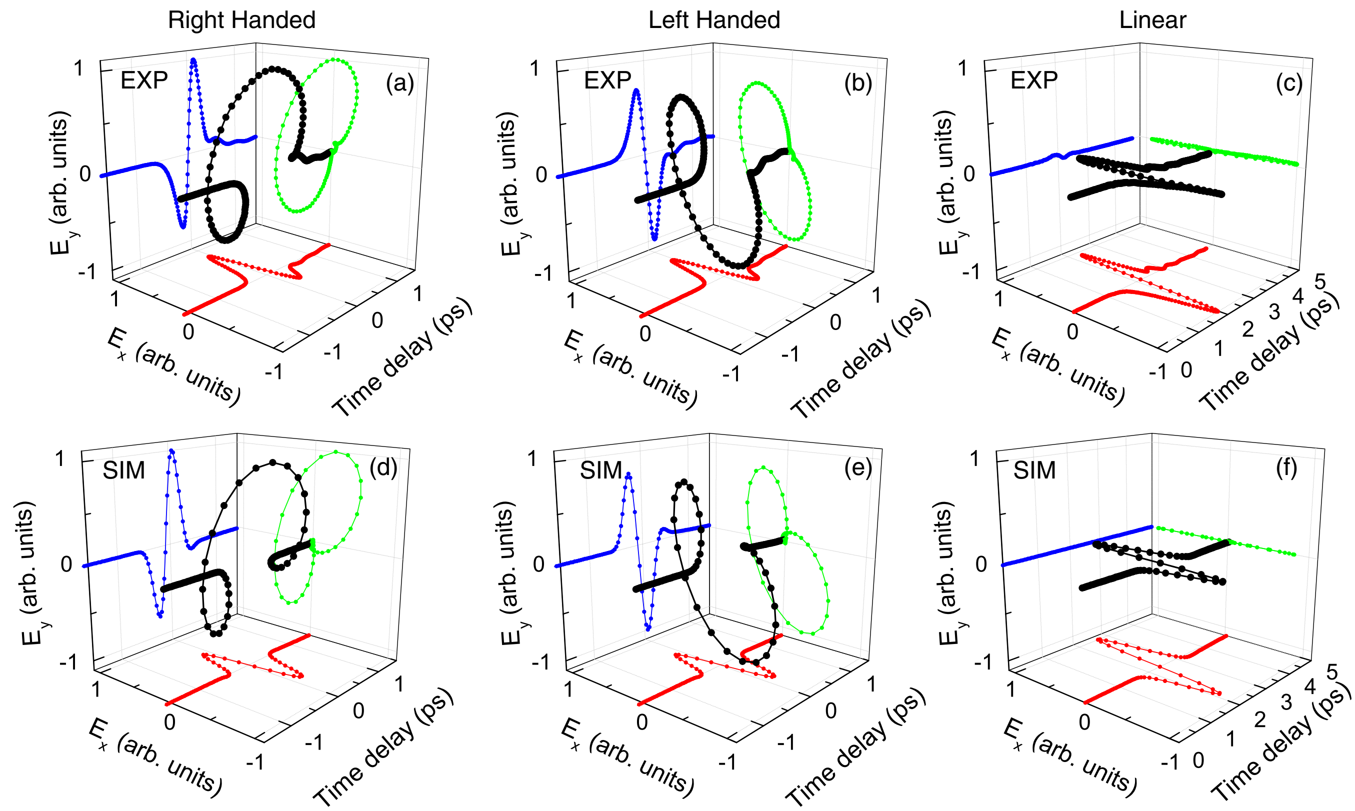

FIG. 2 (color online). Temporal evolution of electric field vector of (a) right-handed, (b) left-handed elliptically polarized, and (c) linear polarized THz pulses. Simulated temporal evolution of electric field vector of (d) right-handed, (e) left-handed elliptically polarized, and (f) linear polarized THz pulses. EXP: experimental results. SIM: simulation results.

$t = t_0$ can be described by $\overrightarrow{v}(z, t, t_0) = -\frac{e}{m}\int_{t_0}^{t}[E_{\rm DC}(z) + \overrightarrow{E}_\omega(z, t')]H(t' - \tau)dt'$, where $H(t' - \tau)$ is the Heaviside function [11].

The transient current contributing to THz radiation is

$$\vec{J}(z, t) = e\int_{-\infty}^{t}\overrightarrow{v}(z, t, t_0)\dot{\rho}(t_0)dt_0. \qquad (1)$$

We used the retarded solution for the one-dimensional Maxwell equation

$$\overrightarrow{A}(t) = \frac{\mu_0}{4\pi R}\int\overrightarrow{J}(z, t)e^{ik_{\rm THz}(R-z)}dz, \qquad (2)$$

to describe the effect of longitudinal propagation on the far-field THz radiation pattern. $\vec{A}(t)$ is the vector potential for THz waves, μ_0 is the vacuum permeability, R is the distance from origin to field point position, $k_{\rm THz} = \Omega/v_{\rm THz}$ is the wave number, Ω is the angular frequency of THz waves, and $v_{\rm THz}$ is the speed of THz waves in air.

The far-field THz radiation is then

$$\vec{E}_{\rm THz}(t) \propto \frac{e^{ik_{\rm THz}R}}{R}\int_{-L/2}^{L/2}\frac{\partial\vec{J}(z, t - k_{\rm opt}z/\omega)}{\partial t}e^{-ik_{\rm THz}z}dz, \qquad (3)$$

where $t - k_{\rm opt}z/\omega$ describes the propagation of optical pulses in air.

In Eq. (3), the mismatch between $k_{\rm opt}$ and $k_{\rm THz}$ will contribute to the vector properties of the far-field THz radiation. We modeled the dynamic process under our experimental conditions by considering a focused Gaussian beam, whose peak value along the z axis can be expressed as $E_\omega(z, t) = E_\omega\frac{w_0}{w(z)}\exp(-2t^2/\tau_0^2)$. Here $w(z) = w_0[1 + (\lambda z/\pi w_0^2)^2]^{1/2}$ represents the transverse dimension of the optical field, w_0 is the beam waist, and λ is the wavelength.

In our experiment, we estimated the velocity mismatch by measuring the time delay between two detected THz waves generated at the beginning and end of the plasma. The corresponding difference in index of refraction was $\Delta n = n_{\rm opt} - n_{\rm THz} = 1.08 \times 10^{-4}$, which was subsequently used in our simulations. Moreover, we ignore the effects of dispersion due to propagation through air on the optical and THz pulse individually, but we include the relative dispersive effects between them. By considering the pulse duration and the focusing condition of laser beams, we estimated the power density in the focus region to be 10^{15} W/cm^2 and the induced plasma density to be within the range of 10^{15}–10^{16} cm^{-3}, assuming a focal spot diameter of 100 μm. The simulated results for various electrodes are shown in Figs. 2(d)–2(f) for comparison. Because of the velocity mismatch between optical pulses and THz pulses during propagation, within the coherent length, the constructed far-field radiation shows the arbitrary elliptical polarization. The simulated results agree well with our experimental findings, and the polarization state of the output THz pulse demonstrates a handedness dependence on the electric field.

We noted that as we move the electrodes away from the center of the plasma, the constructed far-field THz radiation shows different ellipticity. Figures 3(a) and 3(b) show a series of measured x and y components of the THz field while moving the electrode position longitudinally along the plasma with right-handed and left-handed electrodes, respectively. A case with linear electrodes is also shown in Fig. 3(c) for comparison. The calculated results are also shown in Figs. 3(d)–3(f) with the corresponding condition of applied electric field. The black curves reveal the polarization properties of far-field THz radiation at various positions, which define the relative position between the center of the plasma and the electrodes. We found that the far-field THz polarization state depends not only on the applied electric field, but also on the uniformity of the plasma.

To clarify elliptically polarized THz waves generated from double helix electrodes, we compared the results with that obtained by a Fresnel prism made of high-density polyethylene (HDPE). Figures 4(a) and 4(b) plot the electric vector of right-handed and left-handed circularly polarized THz waves obtained with the Fresnel prism, respectively. The input polarization was controlled by a THz polarizer. Two main characteristics of circularly or elliptically polarized beams are the amplitude ratio E_x/E_y and the phase difference between x and y components $\varphi_x - \varphi_y$. For perfect circularly polarized broadband pulses, $E_x(\omega)/E_y(\omega) = 1$ and $\varphi_x(\omega) - \varphi_y(\omega) = \pm\pi/2$ for all frequencies. We extracted the amplitude ratio and the phase difference between x and y components through Fourier transform of experimental data and plotted the results in Figs. 4(c) and 4(d) for the Fresnel prism, and in Figs. 4(e)–4(h) for plasma generation. Because of the large index of refraction and absorption of HDPE, the E_x/E_y and $\varphi_x - \varphi_y$ have a relatively large fluctuation around the optimized number. In contrast, with double helix electrodes, the phase difference and amplitude ratio have minimal fluctuations across the spectral range, providing an elliptical polarized THz wave. The plasma-based method also eliminates absorption, and Fresnel loss issues. With further optimization of the electrodes, plasma profile, and conversion efficiency, this technique may provide an intense and broadband elliptically polarized THz wave.

To produce nearly circularly polarized THz waves with our method, the plasma longitudinal profile and the electrode pitch length are two critical parameters. The E_x/E_y of produced THz pulses is related to the plasma longitudinal profile since the generation efficiency is consequentially associated to the plasma density. In addition, $\varphi_x - \varphi_y$ is strongly dependent on the pitch length of the revolving

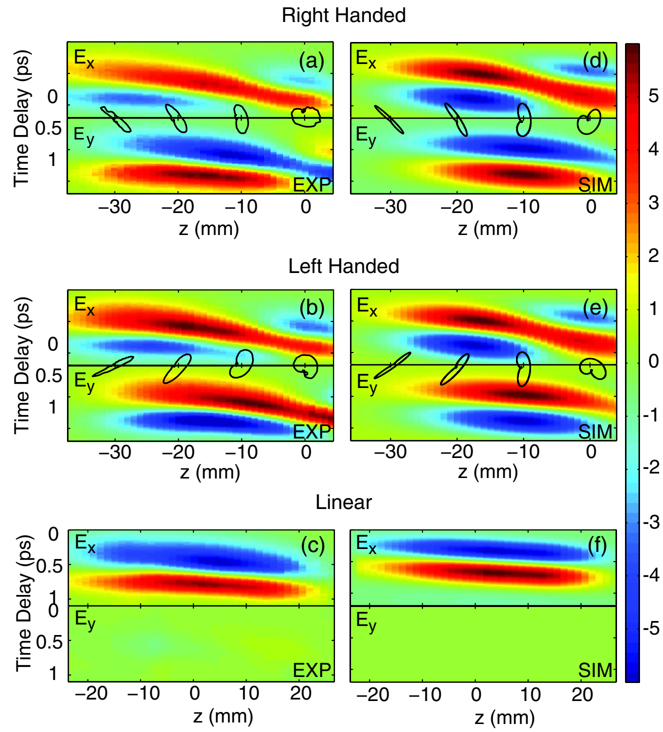

FIG. 3 (color online). The measured far-field THz waves in x (upper) and y (lower) directions at various electrode positions from pairs of (a) right-handed, (b) left-handed helical, and (c) linear electrodes. The calculated far-field THz waves in x and y directions at various electrode positions from (d) right-handed, (e) left-handed helical, and (f) linear electrodes are also shown for comparison. EXP: experimental results. SIM: simulation results.

electric field since the phase difference is provided from the velocity mismatch.

In conclusion, we demonstrate that the polarization of far-field THz radiation can be significantly modified through manipulation of revolving electron transient current in a laser filament with a pair of helical electrodes. An elliptically polarized far-field THz wave pattern was observed and demonstrated to be highly sensitive to the handedness of the applied electric field and the longitudinal position of the electrodes. The velocity mismatch between optical excitation and propagation of THz waves is verified to be the mechanism through which the ellipticity is created. This finding provides new perceptions of plasma-based THz emitters, opens interesting possibilities for coherent control of the polarization properties of plasma-based THz emission over a broader spectral range, and offers a useful diagnostic tool for biomolecular and spintronics studies in the THz frequency range.

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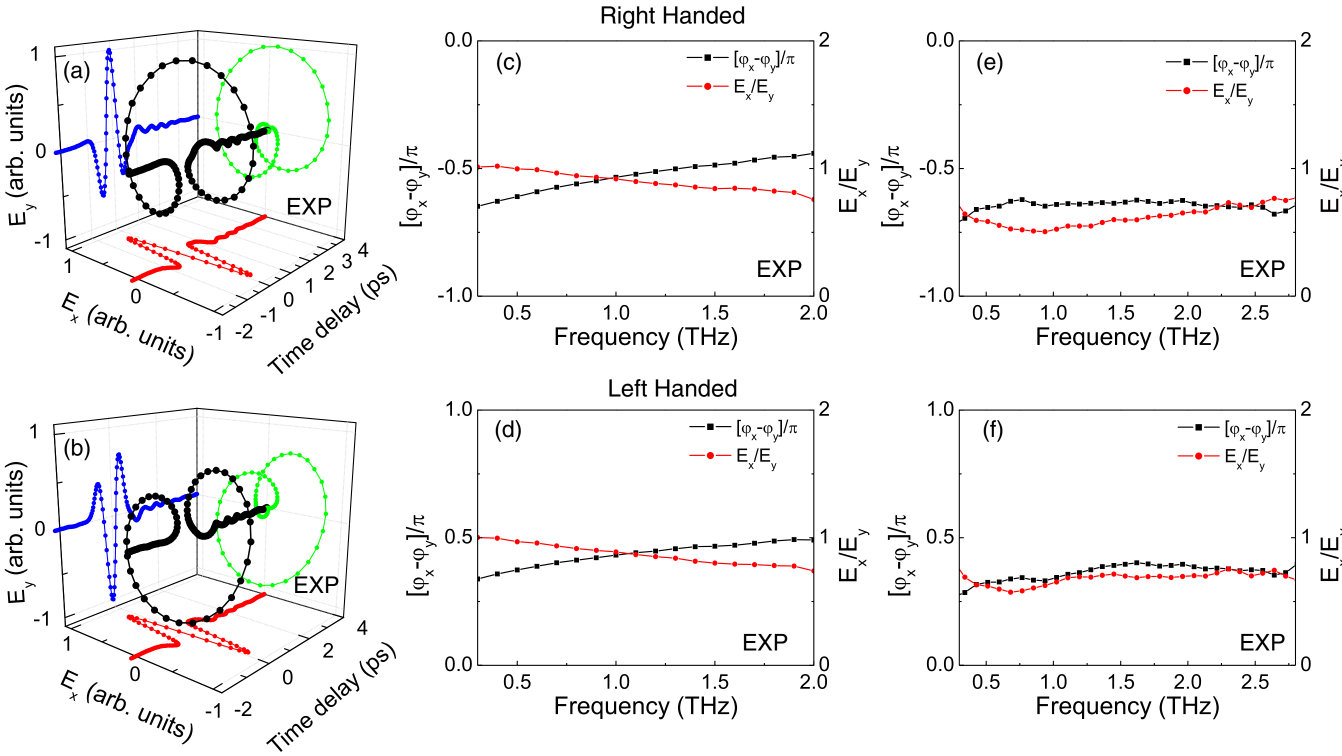

FIG. 4 (color online). Measured temporal evolution of electric field vector of (a) right-handed and (b) left-handed circular polarized THz pulses with a HDPE Fresnel prism. The phase difference and amplitude ratio between x and y components for (c) right-handed and (d) left-handed circular polarized THz pulses in frequency domain. The (e) phase difference and (f) amplitude ratio of elliptically polarized THz pulses generated from double helix electrodes are shown for comparison. EXP: experimental results.

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