Coherent Polarization Control of Terahertz Waves Generated from Two-Color Laser-Induced Gas Plasma

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(Received 4 May 2009; published 10 July 2009)

Electrons ionized from an atom or molecule by circularly or elliptically polarized femtosecond ω and 2ω pulses exhibit different trajectory orientations as the relative phase between the two pulses changes. Macroscopically, the polarization of the terahertz wave emitted during the ionization process was found to be coherently controllable through the optical phase. This new finding can be completely reproduced by numerical simulation and may enable fast terahertz wave modulation and coherent control of nonlinear responses excited by intense terahertz waves with controllable polarization.

DOI: 10.1103/PhysRevLett.103.023001 PACS numbers: 32.80.Fb, 32.80.Qk, 42.65.−k, 52.38.−r

Intense terahertz (THz) wave generation during the gas ionization processes induced by femtosecond pulses at 800 (ω) and 400 nm (2ω) is attracting more attention due to the remarkable bandwidth and intensity it provides, as well as its potential applications in nonlinear THz spectroscopy and remote sensing and imaging [1–16]. This THz source has already become a common tool in research laboratories for fundamental scientific research, such as nonlinear THz responses of different materials [17,18]. In recent works, quantum mechanical models have been employed to describe the physical mechanism for THz generation from gas plasma in the cases using phase-controlled ω and 2ω pulses and few-cycle single-color pulses, respectively, both treated with linearly polarized excitation optical pulses [14,15]. In addition, we have shown that the full THz emission process takes place in two steps: first, a broadband pulse is produced through the asymmetric ionization due to the laser-atom interaction, and then a second step, an "echo" is produced by the interaction of the ionized wave packets with the surrounding gas and plasma [14].

Theoretically, the electrons exhibit different trajectories after being ionized from an atom or molecule by circularly or elliptically polarized optical pulses (ω plus 2ω), in comparison to the case when the pulses are linearly polarized. However, no theoretical predication has been specified for the case of terahertz generation with circularly or elliptically polarized excitation ω and 2ω beams, although Wu *et al.* have applied a semiclassical model to the case with circularly polarized, single-color, few-cycle optical excitation [13]. Using the quantum mechanical model described in our previous work [14], we calculated the electron expectation value trajectories in the case of circularly and elliptically polarized ω and 2ω beams. When linearly polarized optical excitation is used, the problem essentially reduces to two dimensions, and one expects the THz radiation to share the polarization of the pump pulses.

When circularly or elliptically polarized optical fields are applied, the laser-atom interaction requires three dimensions, since the optical field is capable of coupling states with differing values of the z-projection of the angular momentum (m) in addition to the angular momentum ℓ. This was calculated by representing the electron wave function as a series of partial waves in spherical coordinates, with a spatial radial dimension and momentum-space angular dimensions, and numerically solving the time-dependent Schrödinger equation [19]. The simulations were performed using hydrogen for simplicity. The system of coordinates was rotated dynamically such that the vector potential of the laser was always aligned with the z axis (the laser Poynting vector was along the y axis), which allowed the m coupling to be confined to a single operation, $\exp(i\theta L_y)$. The exact (real, dense) operator was used rather than the infinitesimal or Padé approximants so that arbitrary ellipticities could be utilized without the buildup of rotation errors. The laser-induced coupling between the ℓ partial waves was performed in the velocity gauge [19]. The two-dimensional electron polarization was continuously monitored throughout the simulation by calculating the expectation values $\langle z\rangle$ and $\langle x\rangle$ at each time step. This polarization completely describes the THz radiation produced by the first step of the emission process (ionization), and also determines the direction of the remaining emission processes. The simulation includes coupling between bound states as well as ionized states, so all effects due to four-wave mixing [1,5] and other results of the perturbation theory framework are reproduced. However, the generation of terahertz radiation is by far dominated by nonperturbative asymmetric ionization.

The results of such a simulation are shown in Fig. 1, which plots the consequence of changing the relative phase between the fundamental and second harmonic carrier waves, ϕ, when the optical pulses are both right circularly polarized. We can see that instead of the intensity modulation observed with linearly polarized excitation, the THz intensity remains constant, but the polarization angle rotates with ϕ. When left-circular excitation is used, the situation is similar, but the THz field rotation is counterclockwise.

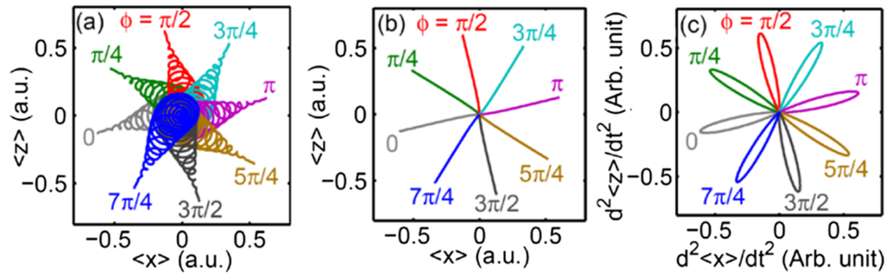

FIG. 1 (color online). Three-dimensional, quantum mechanical simulation of the effects of changing the phase ϕ between the circularly polarized fundamental and second harmonic pulses. (a) Electron expectation value trajectories in the dual-color field. (b) Electron trajectories with the laser-driven quiver motion removed. (c) Second time-derivative of the trajectories, showing the effective polarization of the emitted radiation.

In this Letter, motivated by the above physical picture and some preliminary experimental results indicating that the THz electric field detected by polarization-sensitive electric-optic sampling does not increase when the THz power detected with a pyroelectric detector is tripled during the optimization of the THz emission, we performed systematic experiments to test the polarization behavior of THz waves generated from gas plasma. We performed both theoretical and experimental investigations of the THz polarization characteristics as the relative phase between the ω and 2ω pulses changes, with different combinations of the polarizations of the two pulses.

We found that the polarization of the THz waves is coherently controllable through the phase when at least one of the optical pulses (ω or 2ω) is elliptically polarized. In particular, when both ω and 2ω beams are circularly polarized (or close to it), the THz polarization angle can be rotated arbitrarily simply by changing the phase, with the THz amplitude kept unchanged. Our results not only give a clearer picture about the behavior of the THz emission from gas plasma but also add to the THz air source a more attractive feature that may lead to fast THz wave modulation devices and enable coherent control of nonlinear responses excited by intense THz waves.

In order to test the theoretical predictions, a stable phase-control mechanism with sufficient scan range is necessary. Instead of using the phase compensator described in Ref. [16] or a phase plate [20], a new phase compensator in an inline configuration with attosecond phase-control accuracy is employed, as shown inside the dashed line in Fig. 2(a). A femtosecond pulse at 800 nm (ω) generates a second harmonic pulse at 400 nm (2ω) while passing through a type-I Beta Barium Borate (β-BBO) crystal. The ω and 2ω beams, which have perpendicular polarizations, pass through an x-cut birefringent plate (BP, here we use α-BBO) with its slow axis aligned with the ω beam polarization (o ray) and the fast axis aligned with the 2ω beam (e ray) so that right after this plate, the 2ω pulse leads the ω pulse [20], as shown in the figure. A fused silica wedge pair with a wedge angle of 3.93° is used to finely control the phase delay between the ω and 2ω pulses through the relationship $\Delta\tau = \Delta l(n_{2\omega} - n_{\omega})\tan(\theta_w)$, where Δl is the step size of the mechanical translation stage, n_ω and $n_{2\omega}$ are the refractive indices of the fused silica at 800 and 400 nm, respectively, θ_w is the wedge angle, and $\Delta\tau$ is the resulting optical delay step. Finally, a tunable dual-band wave plate is used to control polarizations of the ω and 2ω beams. In the experiments, additional quarter wave plates at 800 and 400 nm are necessary to further control the polarization of the two pulses.

The advantage of this inline phase compensator is that it combines the minimal lateral displacement of our previous

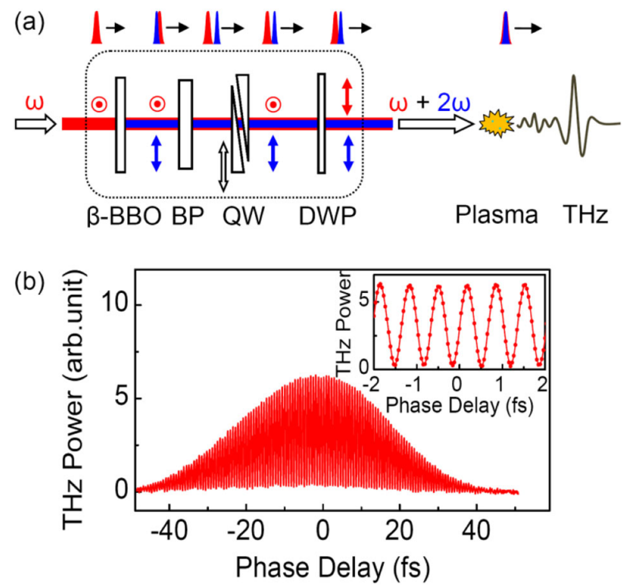

FIG. 2 (color online). (a) Schematic illustration of the experimental setup. Inside the dashed line is the inline phase compensator. β-BBO, Beta Barium Borate crystal; BP, Birefringent Plate (α-BBO); QW, Quartz Wedges; DWP, Dual-wavelength Wave plate; the upper and lower arrows near the laser beams indicate the polarization of the ω and 2ω beams, respectively. (b) A typical phase curve obtained by changing relative phase between the ω and 2ω pulses through the change of the insertion of one of the wedges while monitoring the THz average power with a pyroelectric detector when the ω and 2ω pulses are linearly polarized and parallel to each other; the inset shows a zoomed-in portion of the phase curve.

compensator and the minimal phase fluctuation of the phase plate as described in Refs. [16,20]. Both ω and 2ω beams are focused to ionize the gas and emit THz waves. A broadband THz polarizer is used to analyze the polarization of the emitted THz waves, and a pyroelectric detector is used to monitor the transmitted THz power through the THz polarizer as it rotates. Figure 2(b) shows a phase curve obtained by changing the phase between ω and 2ω pulses through the translation of one wedge while monitoring the THz power with a pyroelectric detector when ω and 2ω pulses are linearly polarized and parallel to each other.

Initially, the THz polarization effect is tested with linearly polarized ω and 2ω optical beams. In this case, the electron energies also change, but the overall orientation does not change. Figures 3(a) and 3(c) show the change in measured THz intensity versus THz polarizer angle and the relative phase between ω and 2ω pulses with the ω and 2ω beams parallel and orthogonally polarized (the 2ω beam is kept vertically polarized), respectively. Figures 3(b) and 3(d) are the corresponding simulation results using the quantum mechanical model. Both experimental results and theoretical simulation show the THz polarization in this case essentially follows the polarization of the 2ω beam. However, the THz emission efficiency is about 1 order of magnitude higher when ω and 2ω are parallel polarized than when they are orthogonally polarized. When the polarizations of the two pulses are aligned, the second harmonic has the effect of causing destructive interference of the electron wave packets in one direction and constructive interference in the other, resulting in one of the electron "beams" emerging from the atom being effectively switched off [14]. When the two pulses have orthogonal polarizations, the relatively weak second harmonic can only "pull" the symmetric pair of beams to one side or the other, resulting in a much smaller net polarization.

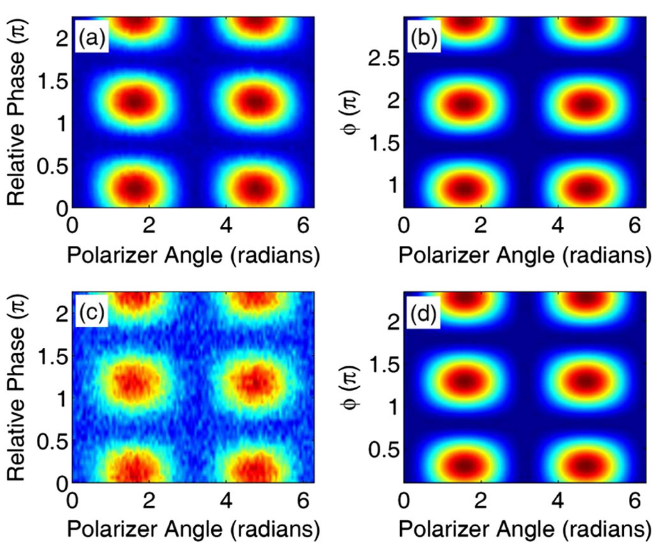

FIG. 3 (color online). THz intensity versus THz polarizer angle and the relative phase between the ω and 2ω pulses with linear polarization. (a) and (c) are experimental results with the two pulses parallel and orthogonally polarized, respectively; (b) and (d) are the corresponding simulation results.

When at least one of the optical beams is circularly or elliptically polarized, the electron trajectories will change their orientation as the phase between ω and 2ω changes. As a consequence, the polarization of the emitted THz wave changes its orientation. Figures 4(a) and 4(c) show the experimental results when the ω beam is left-handed and right-handed circularly polarized while the 2ω beam is elliptically polarized with the ratio between the minor axis and major axis of the ellipse (defined here as ellipticity) of about $1/11$ in terms of THz intensity, respectively. Figures 4(b) and 4(d) are the corresponding simulation results using the quantum mechanical model, which reproduces the experimental results. We can see that when ω is right-handed circularly polarized, the THz polarization rotates in a right-handed manner.

As has been simulated above, the case of both circularly polarized ω and 2ω beams leads to the result that when the relative phase between the pulses changes, the polarization of the emitted THz beam rotates while the intensity or the electric field of the THz wave is kept unchanged. This particular situation is very important for some applications, such as THz modulation devices. Figure 5(a) shows the experimental results when both ω and 2ω beams are right-handed elliptically polarized with their ellipticities both higher than 0.8 (which means that both the ω and 2ω beams are close to circular polarization). Figure 5(b) shows the simulation result for comparison.

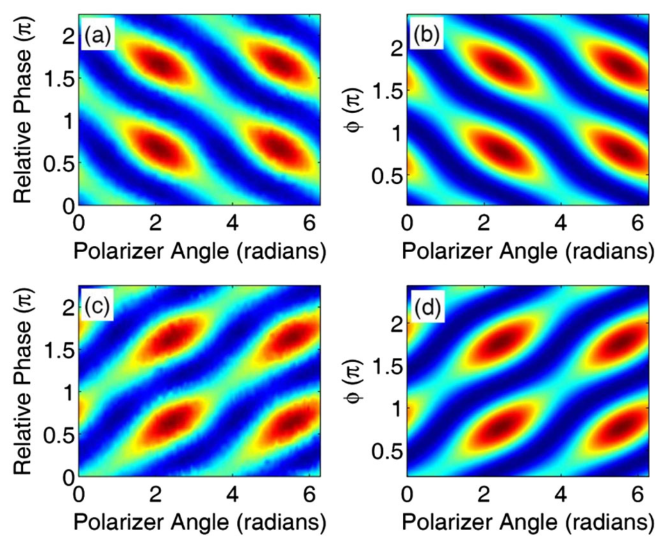

FIG. 4 (color online). THz intensity versus THz polarizer angle and the relative phase between the ω and 2ω pulses with left- or right-handed circularly polarized ω pulse and with elliptically polarized 2ω pulse (with an ellipticity of about $1/11$ in terms of optical intensity). (a) and (c) are the experimental results with left- and right-handed circularly polarized ω pulses, respectively, and (b) and (d) are the corresponding simulation results.

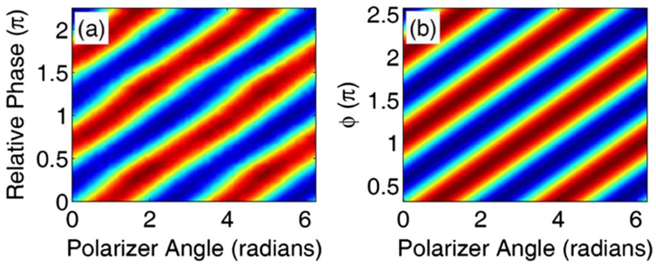

FIG. 5 (color online). THz intensity versus THz polarizer angle and relative phase between ω and 2ω pulses. (a) The experimental result when both ω and 2ω beams are right-handed elliptically polarized with their ellipticities both higher than 0.8 (which means that both of the ω and 2ω beams are close to circular polarization), and (b) the simulation result with ideally circularly polarized optical beams. Both indicate the THz polarization can be rotated while keeping the amplitude of the THz wave constant.

Both the experimental and theoretical results indicate that the emitted THz waves are linearly polarized when the ω and 2ω beams are both linearly polarized. They also indicate that when the ω and 2ω beams are circularly or elliptically polarized the polarization of THz waves can be slightly elliptical but is very close to linear polarization with ellipticity less than $1/20$ in terms of THz intensity. This result is basically consistent with the recent result reported by S. L. Chin's group [11]. Furthermore, our results indicate that, as the relative optical phase changes by 2π, the polarization direction of the THz wave rotates one complete circle accordingly when at least one optical pulse is elliptically polarized. With the above theoretical and experimental analysis, we have a much clearer picture about the THz generation during the gas ionization process. Our demonstration further verified the validity of the quantum mechanical model in our previous work [14].

In conclusion, we presented both theoretical and experimental investigations of the polarization characteristics of the THz waves generated from the gas plasma excited by dual-color optical pulses (ω and 2ω). We found that the polarization of the THz waves can be coherently controlled by changing the phase between the ω and 2ω pulses when at least one of the optical pulses is elliptically polarized. In particular, when both ω and 2ω beams are circularly polarized, the THz polarization angle can be rotated arbitrarily by simply changing the phase between the two optical pulses, with the THz amplitude kept constant. The demonstration adds a new feature into this novel THz gas source and should enable fast THz wave modulation and coherent control of nonlinear responses excited by intense THz waves with controllable polarization.

This work is partially supported by the Office of Naval Research, the Defense Threat Reduction Agency, and the National Science Foundation.

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