Coherent Polarization Control of THz Waves Generated from Asymmetrically Ionized Gases

Jianming Dai\textsuperscript{a)}, Nicholas Karpowicz\textsuperscript{b)}, and X.-C. Zhang\textsuperscript{a)} \textsuperscript{1}
\textsuperscript{a)} Center for Terahertz Research, Rensselaer Polytechnic Institute, Troy, New York 12180, USA
\textsuperscript{b)} Max-Planck Institute for Quantum Optics, Garching, Germany

E-mail: zhangxc@rpi.edu

Abstract. Unlike polarization control of optical waves, lossless control over the polarization of broadband terahertz waves remained challenging. We recently found that the polarization of terahertz waves generated from gas plasma excited by femtosecond fundamental pulse (ω) and its second harmonic (2ω) could be coherently controlled by changing the relative phase between the ω and 2ω pulses. In particular, when the ω and 2ω pulses are both circularly polarized (or close to it), the photo-excited electrons exhibit different trajectories as the relative phase between the two optical pulses changes, and subsequently terahertz polarization angle can be controlled arbitrarily through the relative phase while the intensity of the emitted terahertz wave is kept constant. This new finding may enable fast terahertz wave modulation and coherent control of nonlinear responses excited by intense terahertz waves with controllable polarization.

1. Introduction
As a promising nonlinear optical medium, femtosecond-laser-induced plasma in gases has been studied intensively over the past several years. It is not only used to generate tunable ultrashort laser pulses and coherent, extreme, ultraviolet radiation via phase-matched high-order difference-frequency mixing \cite{1, 2}, but also used to generate terahertz (THz) waves \cite{3-8}. Such an air-plasma-based THz source provides very high THz electric field, which is comparable to those obtained with large-scale facilities only accessible in some national laboratories \cite{9}. Moreover, recent results show that femtosecond laser-induced gas plasma exhibits a remarkable ability to detect pulsed (broadband) THz waves through nonlinear optical processes \cite{10, 11}. The use of air (or selected gases) as both the broadband THz wave emitter and THz wave sensor provides superior bandwidth covering the entire THz gap and well-beyond \cite{12}. Potential applications include nonlinear spectroscopy and imaging, as well as remote sensing and identification. The air-plasma-based THz source has already become a common tool in research laboratories for fundamental scientific research, such as nonlinear THz response of different materials \cite{13, 14}.

Coherent control of THz wave generation from gas plasma has been demonstrated in 2006 \cite{15} with an interferometric phase compensator, which has been used to phenomenologically determine the mechanism for THz wave generation from gas plasma. More recently, quantum mechanical models have been employed to describe the physical mechanism for THz generation from gas plasma in cases

\textsuperscript{1} To whom any correspondence should be addressed.
using phase-controlled femtosecond $\omega$ and $2\omega$ pulses and few-cycle single-color pulses, respectively, both treated with linearly polarized excitation optical pulses [16, 17]. It has been 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.

Using the quantum mechanical model described in our aforementioned work [16], we calculated the electron expectation value trajectories in the case of circularly and elliptically polarized $\omega$ and $2\omega$ beams. When linearly polarized optical excitation beams are used, the problem essentially reduces to two dimensions, and one expects the THz radiation to share the polarization of the pump pulses.

![Figure 1. Three-dimensional, quantum-mechanical simulation of the effects of changing the phase $\phi$ between the circularly-polarized fundamental and second harmonic pulses.](image)

(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. 
(d) and (e) Final electron density distribution in the $z$-$x$ plane (scaled logarithmically) for the phases $\pi$ and $\pi/2$ respectively. 
(f) Momentum space electron density distribution for phase $\pi/2$ (scaled logarithmically, bound states removed).

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 $\ell$. 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. 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 (and the laser Poynting vector was along the $y$-axis), which allowed the $m$-coupling to be confined to a single operation, $\exp(i\mathbf{A}_y)$. The exact (real, dense) operator was used rather than the infinitesimal or Padé approximants so that arbitrary ellipticities could be utilized without the build-up of rotation errors. The laser-induced coupling between the $\ell$ partial waves was performed in the velocity gauge [18]. The two-dimensional electron
polarization was continuously monitored throughout the simulation by calculating the expectation values \(<z>\) and \(<x>\) 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 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, \(\phi\), 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 \(\phi\). When left-circular excitation is used, the situation is similar, but the THz field rotation is counter-clockwise.

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. Specifically, focusing on the configuration with two-color (\(\omega\) and \(2\omega\)) optical pulse excitation, we presented both theoretical and experimental investigations of the THz polarization characteristics as the relative phase between \(\omega\) and \(2\omega\) pulses changes, with different polarization combinations 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 (\(\omega\) or \(2\omega\)) is elliptically polarized. In particular, when both \(\omega\) and \(2\omega\) 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.

2. Experimental investigation and analysis

Experimentally, a stable phase control mechanism with sufficient scan range is necessary. Instead of using the phase compensator described in reference [19] or a phase plate [20], a new phase compensator in an in-line configuration with attosecond phase-control accuracy is employed, as shown inside the dashed line in Fig. 2. A femtosecond pulse at 800 nm (\(\omega\)) generates a second harmonic pulse at 400 nm (\(2\omega\)) while passing through a type-I Beta Barium Borate (\(\beta\)-BBO) crystal. The \(\omega\) and \(2\omega\) beams, which have polarizations perpendicular to each other, pass through an \(x\)-cut birefringent plate (BP, here we use a linear crystal, \(\alpha\)-BBO) with its slow axis aligned with the \(\omega\) beam polarization (o-ray) and the fast axis aligned with the \(2\omega\) beam (e-ray) so that right after this plate the \(2\omega\) pulse is leading to the \(\omega\) pulse, as shown in the figure. A fused silica wedge pair with a small wedge angle of 3.93º is used to finely control the relative phase delay between the \(\omega\) and \(2\omega\) pulses through the relationship \(\Delta \tau = \Delta l (n_{2\omega} - n_{\omega}) \tan(\theta_w)\), where \(\Delta l\) is the step size of the mechanical translation stage, \(n_{\omega}\) and \(n_{2\omega}\) is the refractive index of the fused silica at 800 nm and 400 nm, respectively, \(\theta_w\) is the wedge angle, and \(\Delta \tau\) is the resulting optical delay step. Finally, a tunable dual-band, zero-order waveplate is used to control polarizations of the \(\omega\) and \(2\omega\) beams. In the experiments, additional quarter waveplates at 800 and 400 nm are necessary to further control the polarization of the two optical pulses.

The advantage of this in-line phase compensator is that it combines the minimal lateral displacement of our previous phase compensator and the minimal phase fluctuation of the phase plate as described in references [19, 20]. The collimated \(\omega\) and \(2\omega\) beams are focused simultaneously to ionize the gas and emit THz waves with an off-axis parabolic mirror (M2). A polyethylene or silicon lens is used to collect the emitted 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.** Schematic illustration of the experimental setup. Inside the dashed line is the in-line phase compensator. β-BBO, Beta Barium Borate crystal; BP, Birefringent Plate (α-BBO); QW, Quartz Wedges; DWP, Dual-wavelength Waveplate; the red and blue arrows indicate the polarization of the ω and 2ω beams, respectively; M1, dual-wavelength high-reflection mirror (HR at both 800 and 400 nm); M2, off-axis parabolic mirror used to focus optical beams; Lens, polyethylene or silicon lens.

Fig. 3 shows a typical phase curve obtained by changing relative phase between ω and 2ω pulses through the translation of one of the wedges while monitoring at the THz average power with a pyroelectric detector when ω and 2ω pulses are linearly polarized and parallel to each other.

**Figure 3.** Schematic illustration of the experimental setup. Inside the dashed line is the in-line phase compensator. β-BBO, Beta Barium Borate crystal; BP, Birefringent Plate (α-BBO); QW, Quartz Wedges; DWP, Dual-wavelength Waveplate; the red and blue arrows indicate the polarization of the ω and 2ω beams, respectively; M1, dual-wavelength high-reflection mirror (HR at both 800 and 400 nm); M2, off-axis parabolic mirror used to focus optical beams; Lens, polyethylene or silicon lens.
Initially, the THz polarization effect is tested with linearly polarized $\omega$ and $2\omega$ optical beams. In this case, the electron energies also change but the overall orientation does not change. Figs. 4(a) and 4(c) show the change in measured THz intensity versus THz polarizer angle and the relative phase between $\omega$ and $2\omega$ pulses with the $\omega$ and $2\omega$ beams parallel and orthogonally polarized (the $2\omega$ beam is kept vertically polarized), respectively. Figs. 4(b) and 4(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 $2\omega$ beam. However, the THz emission efficiency is about one order of magnitude higher when $\omega$ and $2\omega$ 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 [16]. 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.

When at least one of the optical beams is circularly or elliptically polarized, the electron trajectories will change their orientation as the phase between $\omega$ and $2\omega$ changes. As a consequence, the polarization of the emitted THz wave changes its orientation. Figs. 5(a) and 5(c) show the experimental results when the $\omega$ beam is left-handed and right-handed circularly polarized while the

![Figure 4](image_url)
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. Figs. 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.

Figs. 6(a) and 6(c) show the experimental results when the 2ω beam is left-handed and right-handed circularly polarized while the ω beam is elliptically polarized with the ratio between the minor axis and major axis of the ellipse (defined here as ellipticity) of about 1/7 in terms of THz intensity, respectively. Figs. 6(b) and 6(d) are the corresponding simulation results using the quantum mechanical model, which also reproduces the experimental results. We can see that when 2ω is right-handed circularly polarized the THz polarization rotates in a left-handed manner.

Figure 5. 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.
As has been simulated above, the case of both circularly-polarized $\omega$ and $2\omega$ 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. Fig. 6(a) shows the experimental results when both $\omega$ and $2\omega$ beams are right-handed elliptically polarized with their ellipticities both higher than 0.8 (which means that both the $\omega$ and $2\omega$ beams are close to circular polarization). Fig. 6(b) shows the simulation result for comparison.

Both the experimental and theoretical results indicate that the emitted THz waves are linearly polarized when the $\omega$ and $2\omega$ beams are both linearly polarized. They also indicate that when the $\omega$ and $2\omega$ 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 [21]. Furthermore, our results indicate that, as the relative optical phase changes by $2\pi$, 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 [16].

Figure 6. THz intensity versus THz polarizer angle and the relative phase between the $\omega$ and $2\omega$ pulses with left- or right-handed circularly polarized $2\omega$ pulse and with elliptically polarized $\omega$ pulse (with an ellipticity of about 1/7 in terms of optical intensity). (a) and (c) are the experimental results with left- and right-handed circularly polarized $\omega$ pulses, respectively, and (b) and (d) are the corresponding simulation results.
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 ($\omega$ and $2\omega$). We found that the polarization of the THz waves can be coherently controlled by changing the phase between the $\omega$ and $2\omega$ pulses when at least one of the optical pulses is elliptically polarized. In particular, when both $\omega$ and $2\omega$ 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. Similar results have been observed independently by two research groups [22, 23]. 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.

Acknowledgement

This work was supported in part by the U.S. Office of Naval Research (ONR), the Defense Threat Reduction Agency (DTRA), National Science Foundation (NSF), and the U.S. Department of Homeland Security through the DHS-ALERT Center under Award No. 2008-ST-061-ED0001. The views and conclusions contained in this document are those of the authors and should not be interpreted as necessarily representing the official policies, either expressed or implied, of the U.S. Department of Homeland Security.

References

[1] F. Théberge, N. Aközbek, W. Liu, A. Becker, and S. L. Chin, “Tunable ultrashort laser pulses generated through filamentation in gases”, Phys. Rev. Lett. 97, 023904 (2006).
[2] S. Meyer, H. Eichmann, T. Menzel, S. Nolte, and B. Wellegehausen, “Phase-matched high-order difference-frequency mixing in plasmas”, Phys. Rev. Lett. 76, 3336 (1996).
[3] T. Bartel, P. Gaal, K. Reimann, M. Woerner, and T. Elsaesser, “Generation of single-cycle THz transients with high electric-field amplitudes”, Opt. Lett. 30, 2805-2807 (2005).
[4] H. Hamster, A. Sullivan, S. Gordon, W. White, and R. W. Falcone, “Subpicosecond, electromagnetic pulse from intense laser-plasma interaction”, Phys. Rev. Lett. 71, 2725 (1993).
[5] S. Tzortzakis, G. Méchain, G. Patalano, Y.-B. André, B. Prade, M. Franco, A. Mysyrowicz, J.-M. Munier, M. Gheudin, G. Beaudin, and P. Encrenaz, “Coherent subterahertz radiation from femtosecond infrared filaments in air”, Opt. Lett. 27, 1944-1946 (2002).
[6] D. J. Cook, and R. M. Hochstrasser, “Intense terahertz pulses by four-wave rectification in air”, Opt. Lett. 25, 1210-1212 (2000).
[7] M. Kress, T. Töffler, S. Eden, M. Thomson, and H. G. Roskos, “Terahertz-pulse generation by photoionization of air with laser pulses composed of both fundamental and second-harmonic waves”, Opt. Lett. 29, 1120-1122 (2004).
[8] T. Löffler, M. Kress, M. Thomson, and H. G. Roskos, “Efficient Terahertz Pulse Generation in Laser-Induced Gas Plasmas”, Acta Phys. Pol. A 107, 99 (2005).
[9] Y. Shen, T. Watanabe, D. A. Arena, C.-C. Kao, J. B. Murphy, T. Y. Tsang, X. J. Wang, and G. L. Carr, “Nonlinear cross-phase modulation with intense single-cycle terahertz pulses,” Phys. Rev. Lett. 99, 043901 (2007).
[10] J. Dai, X. Xie, and X.-C. Zhang, “Detection of broadband terahertz waves with a laser-induced plasma in gases”, Phys. Rev. Lett. 97, 103903 (2006).
[11] N. Karpowicz et al., “Coherent heterodyne time-domain spectrometry covering the entire “Terahertz gap””, Appl. Phys. Lett. 92, 011131 (2008).
[12] I. Ho, X. Guo, X.-C. Zhang, “Design and performance of reflective terahertz air-biased-coherent-detection for time-domain spectroscopy”, Opt. Express 18, 2872-2883 (2010).
[13] H. Wen, M. Wiczer, and A. M. Lindenberg, “Ultrafast electron cascades in semiconductors driven by intense femtosecond terahertz pulses”, Phys. Rev. B 78, 125203 (2008).
[14] P. Gaal et al., “Nonlinear terahertz response of n-type GaAs”, Phys. Rev. Lett. 96, 187402 (2006).
[15] X. Xie, J. Dai, and X.-C. Zhang, “Coherent control of THz wave generation in ambient air”, Phys. Rev. Lett. 96, 075005 (2006).
[16] N. Karpowicz and X.-C. Zhang, “Coherent terahertz echo of tunnel ionization in gases”, Phys. Rev. Lett. 102, 093001 (2009).
[17] A.A. Silaev and N.V. Vvedenskii, “Residual-current excitation in plasmas produced by few-cycle laser pulses” Phys. Rev. Lett. 102, 115005 (2009).
[18] H.G. Muller, “An Efficient Propagation Scheme for the Time-Dependent Schrödinger Equation in the Velocity Gauge”, Laser Phys. 9, 138 (1999).
[19] J. Dai and X.-C. Zhang, “Terahertz wave generation from gas plasma using a phase compensator with attosecond phase-control accuracy”, Appl. Phys. Lett. 94, 021117 (2009).
[20] Yu Oishi et al., “Generation of extreme ultraviolet continuum radiation driven by a sub-10-fs two-color field”, Optics Express 14, 7230 (2006).
[21] Y. Zhang et al., “Non-radially polarized THz pulse emitted from femtosecond laser filament in air”, Opt. Express 16, 15483 (2008).
[22] J. Dai, N. Karpowicz, and X.-C. Zhang, “Coherent polarization control of terahertz waves generated from two-color laser-induced gas plasma”, Phys. Rev. Lett. 103, 023001 (2009).
[23] H. Wen and A. Lindenberg, “Coherent Terahertz Polarization Control through Manipulation of Electron Trajectories”, Phys. Rev. Lett. 103, 023902 (2009).