Non-radially Polarized THz Pulse Emitted From Femtosecond Laser Filament in Air

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Abstract: Femtosecond laser filament could produce THz wave at forward direction. In our experiment, THz pulse emitted from a femtosecond laser filament has been investigated. It is found that the polarization of the studied THz pulse mainly appears as elliptical. This observation supplements the previous conclusion obtained by C. D'Amico et al. that THz wave emitted by a filament is radially polarized. The mechanism of generating elliptically polarized THz wave has been interpreted by either four-wave optical rectification or second order optical rectification inside the filament zone where centro-symmetry of the air is broken by the femtosecond laser pulse.

Keywords: ultrafast nonlinear optics, spectroscopy, Terahertz, nonlinear optics, four-wave mixing

1. Introduction

Terahertz (THz) radiation has attracted great research interests in view of its widely spanned applications such as remote-sensing, chemical spectroscopy, biomedical diagnostics and threat detection [1-3]. Air is particularly interesting for THz generation in term of the simplicity and being free of optical damage [4-9]. However, due to the water vapor absorption, THz wave suffers great loss during the propagation in the atmosphere. Up to now, THz generation through laser filamentation process seems to be the best solution for avoiding this strong attenuation [6-9]. By simply manipulating the position of a filament, THz pulse could be transported to a remotely located target [10]. In fact, this idea has been proven by C. D'Amico et al. recently [6]. The detected THz wave in their work was radially polarized and confined in a forward cone. They have interpreted it as a transition-Cherenkov radiation from space charge moving at light velocity in the wake of a femtosecond laser filament.

In this paper, we have also investigated the THz radiation from a femtosecond laser filament in air. The experimental evidences imply that the polarization of the observed THz pulse differs from the previous reports. It consists of elliptically polarized component. An intuitive explanation about the generation mechanism of elliptically polarized THz pulse has also been proposed. In details, two orthogonally polarized THz wave could be generated by either four-wave rectification [8] or second order rectification due to the spatial asymmetry of filament plasma. Taking into account the optical birefringence brought forth by femtosecond laser pulse [11], the output THz pulse from the filament could be elliptically polarized depending on the amplitude and phase difference of two orthogonal polarization components.

2. Experimental setup

In our experiment, a 1 *kHz*, 800 *nm*, 45 *fs* Ti-sapphire laser beam was split into two paths. One was used as the pump for the THz wave generation, and the other was the probe for an electro-optic sampling (EOS) setup. Fig. 1 schematically illustrates the experimental configuration. The pump beam with energy of 1.15 *mJ*/pulse was focused by a plano-convex lens with focal length of 50 *cm* in ambient air, producing a 3-*cm* long filament. The emitted THz pulse from this filament was collimated by a parabolic mirror (PM1) with a 4-*mm* diameter hole at the center. Most fundamental pulse energy passed through this hole. The collimated THz pulse was focused by another parabolic mirror (PM2) into a 1-*mm*-thick <110> oriented ZnTe crystal (Zomega Terahertz Corporation). The [0,0,1] axis (Z axis) of the ZnTe crystal was oriented horizontally throughout the experiment. The diameter and focal length of these two parabolic mirrors were 5 *cm* and 10 *cm*, respectively. The diagnosis of the THz pulse was realized by a standard time-resolved EOS method [12]. In this case, the probe beam joined the THz beam by a pellicle beam-splitter (PBS, R=45% for 800 *nm*). Two beams propagated collinearly through the ZnTe crystal. In addition, a Teflon plate with 5 *mm* thickness and 75 *mm* diameter was placed between the parabolic mirrors to block residual fundamental light.

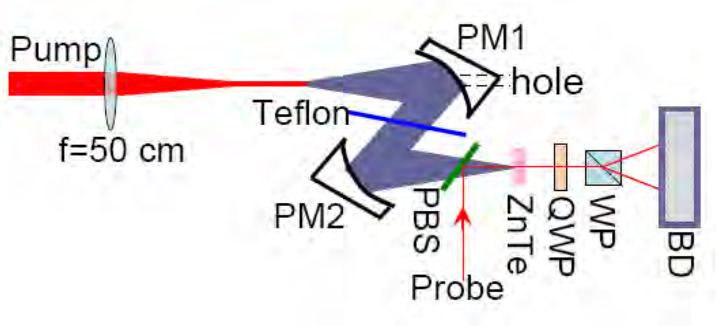

Fig. 1 Schematic illustration of the experimental setup. Time delay of the probe beam can be varied by a delay line which is not specified here.

3. Results and discussions

Fig. 2 shows typical THz electric field waveform obtained in our experiment. The corresponding THz spectrum is given in the inset of Fig. 2. Note that the oscillations after the first cycle of the THz signal in Fig. 2 are resulted from the strong water vapor absorption in air. Fig. 3 presents the peak-to-peak amplitude of the THz electric field (solid squares) at different polarization orientation of the pump beam. In Fig.3, θ is defined as the angle of the polarization of the pump beam with respect to the Z axis of the ZnTe crystal. The variation of θ was experimentally achieved by putting a zero-order half-wave plate (HWP) before the convex lens, while the polarization of probe beam was fixed horizontally. As shown in Fig. 3, the maximum signal was obtained when $\theta = 0^o$, while the minimum signal was observed when $\theta = 90^o$. Based on this result, we could deduce that the observed THz pulse can not be totally attributed to the transition-Cherenkov radiation from the axial current excited by the ponderomotive force of a short laser pulse which is independent on the polarization orientation of the driving laser pulses [6].

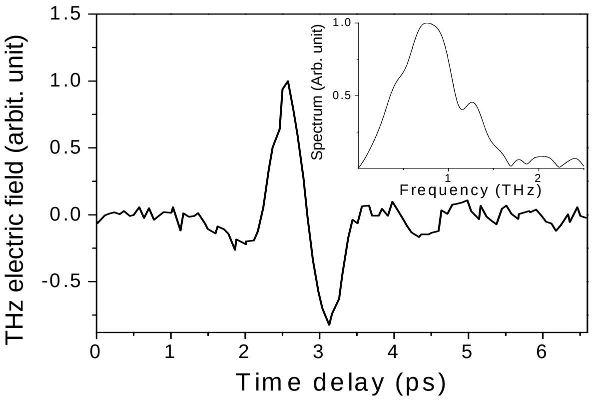

Fig. 2 Typical experimental THz electric field waveform and the corresponding THz spectrum in the inset.

Next, we will verify if the observed THz pulse is linearly polarized. Recalling that by using EOS method to diagnose THz wave, the change of the signal as a function of the angle between the THz electric field polarization and the [0, 0, 1] axis of the ZnTe crystal obeys the following rule [13]:

$$S_{signal} \propto E_{THz}(\cos\phi \sin 2\alpha + 2\sin\phi \cos 2\alpha) \tag{1}$$

Where ϕ and α correspond to the angles of the THz wave polarization and the probe beam polarization with respect to the [0,0,1] axis of the ZnTe crystal, respectively, while E_{THz} denotes the magnitude of the THz electric field which is assumed to be independent of θ in the calculation. It is reasonable because all the experimental conditions except of the pump beam polarization were kept constant. Since $\alpha = 0^o$ in our experiment, Eq. (1) can be further shortened as:

$$S_{signal} \propto 2E_{THz} \sin\phi \tag{2}$$

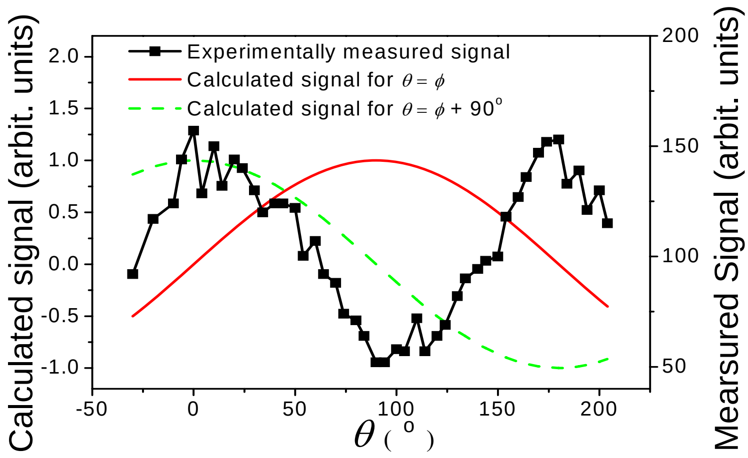

Fig. 3 Solid squares (right label): measured peak-to-peak amplitude of THz electric field as a function of θ, and θ is defined as the angle of the pump pulse polarization with respect to the [0,0,1] axis of the ZnTe crystal); red solid line (left label): calculated orientation dependent EOS signal of assumed linearly polarized THz wave according to Eq. (2) (θ = ϕ, ϕ being the angle between the THz wave polarization and the [0,0,1] axis of ZnTe crystal); green dashed line (left label): the same as the red solid line but θ = ϕ + 90^{o}, i.e. polarization of the THz wave is perpendicular to that of the pump beam.

Our first attempt was to study the situation that the polarization of the generated THz pulse was the same as the pump beam, namely, θ = ϕ. In this case, the calculated EOS signal as a function of θ is indicated in Fig. 3 by the red line (left label). Immediately, one finds that it is different to the experimental results. Furthermore, another linear polarization state has also been considered. In this later case, the polarization of THz pulse is perpendicular to that of the pump beam (θ = ϕ + 90°). The corresponding plot is shown as green line in Fig. 3. It is peaked at θ = 0° and arrives at its minimum when θ = 180° . It does not fit the experimental results either. Cleary, the assumption of linear polarization fails to reproduce the experimental results. Hence, the discrepancy between the calculations and experimental results has rule out the possibility that the THz polarization is linear, which could be related to ponderomotive THz emission with the presence of pre-formed plasma [4, 5]. Then, there is only one choice left for us. The THz pulse generated by the filament in our experiment is elliptically polarized.

However, it is not trivial for us to directly compare the evolution of θ dependent EOS signal of an elliptically polarized THz pulse with the theoretical prediction as what has been done for linear polarization in Fig. 3. On the other hand, according to Eq. (2), when the incident THz wave is elliptically polarized, the EOS setup only response to the projection of the THz electric field on the vertical axis since the Z axis of the ZnTe crystal is oriented horizontally. It is equivalent to the transmission through a polarizer whose optical axis is along the vertical direction in the experiment. We have known that for an elliptically polarized wave, the tip of its electric field vector describes an ellipse in any fixed intersecting plane. Besides, it can be resolved into an arbitrary set of mutually orthogonal component waves with their polarization planes perpendicular to each other. Therefore, by recomposing the THz electric fields obtained at two orthogonal directions (i.e., θ and θ + 90^{o}), it is feasible to derive the elliptical polarization trajectory if the THz pulse is elliptically polarized. Typical

THz electric fields experimentally measured at two orthogonal directions ($\theta = 15^o$ and $\theta = 105^o$). We define the THz electric field obtained at $\theta = 15^o$ as X and the electric field at $\theta = 105^o$ as Y. When they are plotted in an X-Y graph, a closed polarization trajectory is observed (solid square in Fig. 4). Each solid square in Fig. 4 corresponds to the measured THz amplitudes of two orthogonal directions at one specified time delay and two successive solid squares are separated by 100 *fs*. This polarization trajectory essentially follows an elliptical shape. The fact that the experimental polarization trajectory is not a perfect ellipse might be due to the broad bandwidth of near single-cycle THz pulse and energy fluctuation of the laser. Thus, we have confirmed that an elliptically polarized THz pulse has been observed in our experiment accompanying the occurrence of femtosecond laser filamentation in air.

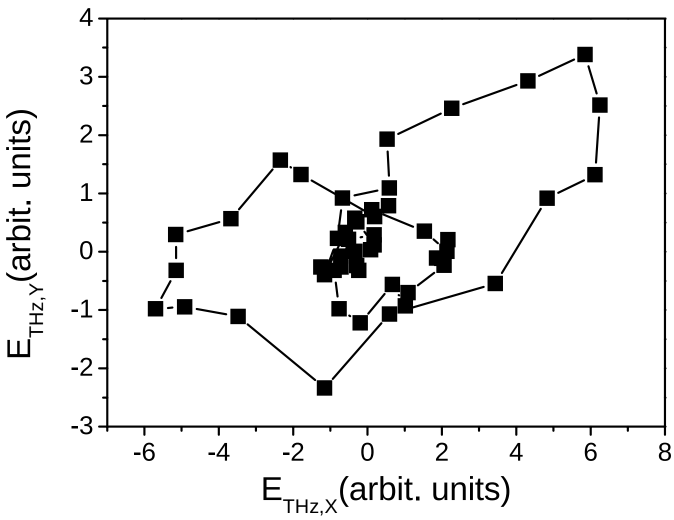

Fig. 4 Measured THz polarization.

Note that the depolarization of THz wave was observed in ref. 5 by using 50 *mJ*, 120 *fs* laser pulses and with strong external focusing (f = 5 *cm*). The authors of ref. 5 explained this phenomenon by severe laser beam distortion and breakup. However, we did not observe beam breakup and distortion during our experiment. Therefore, their hypothesis could not explain our observation. On the other hand, it is known that THz wave could be generated by femtosecond laser pulse in air through the four wave rectification process [8]:

$$\Omega_{THz} = \omega_1 + \omega_2 - \omega_3 \tag{3}$$

Eq. (3) hints that the optical frequencies ω_1, ω_2 and ω_3 should satisfy $\omega_1 + \omega_2 \cong \omega_3$. Since in our work, we did not use the popular ω-2ω scheme [8, 14-16], it is required that the pump laser spectrum need to be octave-spanned [17]. Anyway, this condition can be easily fulfilled because in the course of filamentation, the laser spectrum could cover an extremely broad range, extending from ultraviolet to infrared [18-20]. Hence, THz wave with polarization parallel to the pump polarization (referred to x direction in the following discussion) can be obtained in air via

$$E_{THz,x} \propto \chi^{(3)}_{xxxx} E_{\omega_3} E^*_{\omega_1} E^*_{\omega_2} e^{i\Delta kL} \quad (4)$$

where $\Delta k = k_1 + k_2 - k_3$ describes the phase matching condition and L is the effective interaction length. On the other hand, the generation of orthogonally polarized THz wave could be realized mathematically through

$$E_{THz,y} \propto \chi^{(3)}_{yxxx} E_{\omega_3} E^*_{\omega_1} E^*_{\omega_2} e^{i\Delta kL} \quad (5)$$

Note that $\chi^{(3)}_{yxxx} = 0$ for an isotopic material like air. However, X. Xie et al. have observed non-vanished $\chi^{(3)}_{yxxx}$ when plasma is produced in air using femtosecond Ti-sapphire laser pulses. They have attributed it to the spatial asymmetry of laser induced plasma [8]. Accordingly, THz pulse could be generated along both orthogonal axes. Two components would travel at different phase velocities due to the substantial difference in the non-linear refractive indices generated by the femtosecond laser pulse along its polarization axis and the orthogonal axis, respectively [11]. Finally, an elliptically polarized THz pulse could be generated at the output of the filament. The shape of the polarization ellipse depends on both the accumulated phase delay and the amplitudes of the two polarization components.

The argument outlined above is mainly focused on the third order nonlinear optical susceptibility. It is worth mentioning that due to the spatial non-uniformity of the laser induced plasma, second order nonlinear optical process such as second harmonic generation have been observed [21, 22] using longer laser pulses and lower intensities (order of 10^{12} W/cm^2) than the clamped intensity inside a filament (~$5\times10^{13} W/cm^2$) [20]. This could provide another alternative hypothesis of the generation mechanism of the elliptically polarized THz pulse in our work. The second order optical rectification could be realized in this case through $\omega_{THz} = \omega_{800+THz} - \omega_{800}$ along both orthogonal axes leading to the detected THz pulse output.

4. Conclusions

In conclusion, we have observed elliptically polarized THz emission from a filament induced by intense femtosecond laser pulses. Two orthogonal polarization components may be generated simultaneously through four-wave rectification in view of non-zero susceptibility tensor elements $\chi^{(3)}_{xxxx}$ and $\chi^{(3)}_{yxxx}$. Though $\chi^{(3)}_{yxxx}$ is vanished in normal air, it could be non-negligible in the presence of plasma [8]. The spatial non-uniformity of plasma might also make second order optical rectification feasible giving rise to THz wave generation along both the laser polarization and the orthogonal polarization. In this case, further study is under the way to clarify the role of the involved nonlinear optical processes. On the other hand, significant birefringence induced by the femtosecond laser pulse will lead to phase delay between the laser pulse polarization axis and the orthogonal axis. This in term produces elliptically polarized THz pulse at the output of the filament. It has been widely reported that the filament length can be easily controlled by varying the pump laser parameters such as pulse energy, chirp and focal length [18-20]. Consequently, the phase delay is potentially controllable, realizing controllable polarization of the generated THz wave. This can be potentially helpful to the control of excitons in semiconductor nanostructures [23] and of molecular rotational wave packets [24].

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