RESEARCH ARTICLE

Simultaneous generation of nonlinear optical harmonics and terahertz radiation in air: polarization discrimination of various nonlinear contributions

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Abstract In this paper, we experimentally observed generation of the second and the third optical harmonics and the broadband terahertz radiation in the course of 800 nm 120 fs pulse in atmospheric air. The analysis of their polarization properties revealed unity of their nonlinear optical nature. Taking into account only the third-order nonlinear response of the neutral molecules of air, we analytically described the newly generated elliptically polarized 3d harmonic, the linear polarization of terahertz radiation and the stability of terahertz energy yield for the initial circularly polarized ω pump pulse.

Keywords terahertz, polarization, harmonics, nonlinearity

1 Introduction

When focusing femtosecond laser radiation, atmospheric air acts like nonlinear optical medium. This leads to generation of optical harmonics, waves at sum- and difference frequencies at fairly small femtosecond laser pulse energies [1]. In the case of simultaneous interaction of laser pulses at frequencies ω and 2ω in the optical beam waist, a number of symmetry forbiddances are removed and both odd and even harmonic generation [2,3] become allowed. This is also related to low frequency generation in the terahertz frequency range [4,5].

The nature and the origin of nonlinear polarization in gases interacting with powerful laser radiation are quite complex. Along with many others, major contributions are related to gas atoms which stay neutral after the interaction with intense laser radiation, and free electrons which are result of ionization of other part of atoms of the medium. These contributions can interact both constructively and destructively behaving differently in low-frequency emission properties, such as THz emission spectrum, directionality diagram and its polarization properties [6]. In our earlier works, we demonstrated that the contribution of free electrons (the transient photocurrent) works excellently in the range 0.1–8 THz, while moving further to the far infrared range needs the inclusion of the nonlinear polarizability of molecules that remains neutral after the interaction with the femtosecond pulse [7]. Similar results were obtained by the other authors who analyzed spectral properties of the THz emission from gaseous media [8,9].

In this paper, we studied experimentally the polarization of the second and the third optical harmonics and the broadband terahertz radiation generated during the interaction of two femtosecond pulses with degenerated frequencies ω and 2ω with air. While nonlinear polarization properties at second and third optical harmonics are defined mainly by the contribution of neutral atoms, the terahertz emission is in general described as the contribution of both neutral atoms and free electrons. We showed that taking into account only the third-order nonlinear response of an isotropic medium containing neutral [10] (non-ionized) atoms is enough to describe the experimentally observed polarization properties of the newly generated third harmonic, the phase-dependent linear polarization of the broadband terahertz radiation and the appearance of the crossed-polarized second harmonic radiation.

2 Experimental setup

In our experiments, we used laser radiation from Ti: Sapphire regenerative amplifier (Spectra Physics Spitfire) with the energy up to 2 mJ per pulse, pulse duration 120 fs,

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Fig. 1 Scheme of experimental setup. The dielectric mirror 1 (DM1) splits the input optical beam with 50% reflection and 50% transmission. The second harmonic pulse is generated in beta barium borate crystal (BBO) and delayed with a delay line (DL). The wave plates (WP1 and WP2) were used to control the polarization state of the beams, the mirror (M1) reflected second harmonic radiation and transmitted the fundamental radiation, the Glan prism (GP) cleaned the linear polarization of the second harmonic radiation. The dielectric mirror 2 (DM2) recombined the two beams. Lenses (L1 and L2) were used to focus and collimate the optical radiation, the photodiode (PD) detected the intensity of second harmonic radiation. Silicon filter (Si) was used to block the optical radiation and transmit terahertz radiation. The off-axis parabolic mirrors (PM1 and PM2) guided the terahertz beam into the entrance window of a Golay cell detector

wavelength 797 nm and 1 kHz repetition rate (see Fig. 1). Laser beam was divided into two arms with equal energies using 50/50 beam splitter. The first beam (called ω beam) was used for fundamental radiation, while the second one-for generation of second harmonics with 300 mkm I-type beta barium borate (BBO) crystal (2ω beam). In the 2ω beam, the residual fundamental radiation was filtered out with dielectric mirrors, 2ω polarization was adjusted with half-wave plate and cleaned with a Glan prism just before recombining the two beams. Polarization and energy of the ω beam was adjusted with a phase plate and attenuator. The temporal overlap of the beams was controlled using a delay line in the arm of 2ω beam. The beams were recombined with a dichroic mirror 2 (DM2) which reflected the second harmonic (SH) radiation.

The resulting two-color radiation was focused by the L2 lens with f = 10 cm. Its focal region is the area of interaction, where plasma channel is ignited and generation of optical harmonics and terahertz radiation takes place. Optical radiation from the breakdown region was collimated with a quartz lens L3 (f=10 cm) and guided into the slit of ActonResearch SpectraPro 500i monochromator coupled to Princeton Instruments Pixis 400 charge coupled device (CCD) camera for studying 3ω polarization or to a diffraction grating with 1200 slits/mm for studying the polarization of 2ω using a photodiode and lock-in amplifier.

The terahertz radiation was separated from the optical beam using a 0.35 mm thick Si plate, collimated and refocused using off-axis parabolic mirrors (PM1 and PM2) with aperture diameter 51 mm and effective focal distance 51 mm. The power of the terahertz radiation was measured with a Golay cell (Tydex GC-1P) able to detect radiation from 0 to 300 THz. A wire-grid analyzer was placed into the collimated terahertz beam to study its polarization. The schematic picture of the system used for measurement of terahertz radiation is shown on the inset of Fig. 1.

We stress that in all the experiments we kept the similar energies of the input ω and 2ω radiations ((0.25\pm0.01) and (0.05\pm0.005) mJ/pulse, respectively), so that we could compare the data on different nonlinear processes under study.

3 Experimental results

We investigated the polarization state of the 2ω radiation emitted from the beam focus in the case of the linear initial polarization of both ω and 2ω and zero time delay between the pulses. The $E_{2\omega}$ vector of the 2ω beam radiation was directed along the x axis while for the ω beam the angle ψ between the x axis and E_{ω} vector was varied in the 0–180° range.

The measured dependence of the energy $W_y^{2\omega}$ of SH radiation polarized along the y axis (i.e., orthogonally to the initial 2ω polarization) on the angle ψ is shown in Fig. 2. The dependence has a period of 90° and is symmetric about $\psi = 45^\circ$ and $\psi = 135^\circ$, where it takes its maximum value. This is consistent with the results obtained in argon in Ref. [11]. In case of air as a molecular gas, the birefringence is possible for nonzero time delays between the strong pump pulse and probe one [12]. This effect is caused by the molecular alignment by the pump pulse. We carried out the measurements of dependence of the energy $W_y^{2\omega}$ on angle ψ at delays of 2.8 and 4.0 ps.



Fig. 2 Measured intensity of the 2ω radiation polarized orthogonally to the initial 2ω polarization vs angle ψ between electric fields of ω and 2ω pulse at zero delay between pulses (green circles). The solid line shows the dependence of the 2ω energy at the crossed analyzer in accordance with Eq. (4) (see Section 4). Black squares and red circles show the dependence of $W_y^{2\omega}$ vs the angle ψ for 4.0 and 2.8 ps delay between ω and 2ω pulse (the moments of realignment of N₂ and O₂ molecules respectively)

These delays correspond to the first realignment of O_2 and N_2 molecules respectively. The character of the resulting dependences is similar with one at zero delay, but the efficiency of the polarization rotation is lower (see Fig. 2).

Figure 3 shows the experimental dependences of energies $W_y^{0\omega}(\psi)$ and $W_x^{0\omega}(\psi)$ of terahertz radiation polarized along the y and x axes respectively. The $W_y^{0\omega}(\psi)$ is also symmetric about $\psi = 45^\circ$ and similar to the $W_y^{2\omega}(\psi)$ dependence. The energy $W_x^{0\omega}(\psi)$ of x-

polarized terahertz radiation has its maximal value when ω and 2ω polarizations are parallel and $W_x^{0\omega}(\psi)$ decreases as the angle ψ increases from 0 to 90° (the case of orthogonally polarized ω and 2ω beams).

We also studied the polarization state of the 3ω radiation resulting from nonlinear interaction of circularly polarized fundamental radiation and linearly polarized second harmonic radiation (along x axis). A $\lambda/4$ wave plate placed before the dichroic mirror 2 (DM2) was used to achieve the circular polarization of ω beam. As the DM2 mirror has slightly different transmittance for s- and p-polarized radiation, the angle of incidence of the ω radiation on the wave plate was tuned to achieve the lowest possible ratio of the major to minor ellipse axes of the polarization ellipse which in our case reached 1.05. Intensity of the input ω beam vs the analyzer angle is shown in Fig. 4 with red circles. In this configuration of the optical fields, the third harmonic radiation is generated only [1] in the four-wave mixing process $3\omega = 2\omega + 2\omega - \omega$. We observed the elliptically polarized 3ω radiation with the major axis of the ellipse directed along the electric field vector of 2ω pulse (see Fig. 4).

In our experiments, we did not control the phase difference between the fields of the first and the second harmonics and it was fluctuating in the whole range $0-2\pi$ due to small mechanical vibrations of the beam steering elements in the ω and 2ω beams. To avoid possible averaging effects, we switched our regenerative amplifier from 1 kHz to 10 Hz regime so that the Golay cell could detect single pulse energies. In this configuration, we studied terahertz pulse energies for circular polarization of the input ω radiation and the linear polarization of the 2ω beam.

Figure 5(a) shows the measured energies of subsequent THz pulses in case when no analyzer is placed in the



Fig. 3 Energy of y- (a) and x- (b) polarized terahertz radiation vs the angle ψ between electric fields of ω and 2ω pulse. The solid curves show the dependences of the x- and y-polarized terahertz energy in accordance with Eqs. (7) and (8) (see Section 4)



Fig. 4 Polarization of the third harmonic radiation (black squares) as compared with the polarization if the initial ω radiation (red circles) and the linear 2ω polarization direction (blue line). Orange solid line shows the simulated 3ω polarization (Eqs. (10) and (11))

terahertz beam. Despite some fluctuations, the terahertz pulse energy is constant and it is substantially higher than the noise level. For the case when the analyzer is placed in the collimated terahertz beam, the pulse energies are shown in Fig. 5(b) having the same intensity scale as the Fig. 5(a). There are laser shots for which terahertz energy falls down to zero. This demonstrates random appearance of the crossed-polarized terahertz pulses. Thus, inserting the terahertz analyzer into the collimated terahertz beam does not reveal any specific polarization direction of terahertz radiation. The data shown in Figs. 5(a) and 5(b) are obtained using the same experimental conditions. Therefore, we may conclude that from one laser shot to another the terahertz pulse polarization changes randomly but remains linear. The evidence for the linear polarization is the occurrence of almost zero transmission through the analyzer (Fig. 5(b), shots 17 and 22). This random change of the terahertz polarization direction is, as expected, due to the random fluctuation of the ω to 2ω phase in our experimental setup.

4 Theoretical description of experimental results

In this section, we proposed that the observed polarization properties of the generated terahertz radiation and optical harmonics can be described using the third-order effective nonlinearity only and neglecting spatial and temporal dispersion. The general expression for the third-order susceptibility tensor derived in Ref. [13] is substantially simplified if the material dispersion is absent:

$$\chi_{ijkl}^{(3)}(t_1, t_2, t_3) = (\chi_0/3)\delta(t_1)\delta(t_2)\delta(t_3)$$
$$[\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}].$$
(1)

Here χ_0 is a constant, $\delta(t)$ is Dirac delta function and δ_{ij} is Kronecker delta. In this case, the polarization of the medium P(t) can be written as

$$P_{i}(t) = \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \chi_{ijkl}^{(3)}(t_{1}, t_{2}, t_{3}) E_{j}(t - t_{1}) E_{k}(t - t_{2}) E_{l}(t - t_{3}) dt_{1} dt_{2} dt_{3} = \chi_{0} E_{j}(t) E_{k}(t) E_{l}(t).$$
(2)

Here any repeated index symbol is summed over. A



Fig. 5 Energy of subsequent terahertz pulses generated by circularly polarized ω beam and linearly polarized 2ω beam in case of no terahertz analyzer (a) and terahertz wire-grid analyzer (b) present in the collimated terahertz beam

femtosecond pulse having two carrier frequencies propagating along the z axis induces polarization of the medium which acts as a source of the electromagnetic field under study which contains optical harmonics and THz radiation.

In our experiments, the light field of the 2ω -pulse was linearly polarized along the *x*-axis, while the polarization direction of the ω -pulse varied with the angle ψ relatively to the *x*-axis. The total two-color optical field inducing the nonlinear polarization is written as

$$E_x(t) = E_\omega(t)\cos\psi\cos\omega t + E_{2\omega}(t)\cos(2\omega t + \varphi_2),$$

$$E_y(t) = E_\omega(t)\sin\psi\cos(\omega t + \varphi_1),$$
(3)

where $E_{\omega}(t)$ and $E_{2\omega}(t)$ are the envelopes of the first and the second harmonic radiation, respectively, ψ is the angle between their polarizations, φ_2 is the phase difference between the *x*-polarized components of the ω and 2ω fields, φ_1 is phase shift between the two components of the ω field.

First we consider the generation of terahertz radiation $(0 = 2\omega - \omega - \omega \text{ process})$ and the transformation of the 2ω radiation $(2\omega = 2\omega + \omega - \omega)$ if the linearly polarized ω pulse interacts with the linearly polarized 2ω pulse, angle between the polarization directions being ψ . Using Eqs. (2) and (3), the polarization of the medium oscillating at the frequency 2ω can be written as

$$P_{y}^{(2\omega)}(t) = \frac{\chi_{0}}{2} E_{\omega}^{2}(t) E_{2\omega}(t) \cos\left(2\omega t + \varphi_{2}\right) \sin 2\psi, \quad (4)$$

$$P_x^{(2\omega)}(t) = \frac{\chi_0}{2} E_{\omega}^2(t) E_{2\omega}(t) \cos(2\omega t + \varphi_2)(2 + \cos 2\psi).$$
(5)

As follows from Eqs. (4) and (5), the dependence of energy of 2ω radiation polarized orthogonally to the initial 2ω radiation has the sine-square character: $W_y^{(2\omega)}(\psi) = W_y^{2\omega(\max)} \sin^2 2\psi$. This is in good agreement with the experimental data (Fig. 2, the solid line).

Using Eqs. (2) and (3), we can also obtain the expressions for the components of the nonlinear polarization oscillating at THz frequency:

$$P_{y}^{(0\omega)}(t) = \frac{\chi_0}{4} E_{\omega}^2(t) E_{2\omega}(t) \cos\varphi_2 \sin 2\psi, \qquad (6)$$

$$P_x^{(0\omega)}(t) = \frac{\chi_0}{4} E_{\omega}^2(t) E_{2\omega}(t) \cos \varphi_2(2 + \cos 2\psi).$$
(7)

The dependence of *y*- and *x*-components of terahertz radiation on the angle ψ is given by

$$W_{y}^{\text{THz}}(\psi) = W_{y}^{\text{THz}(\max)} \sin^{2} 2\psi, \qquad (8)$$

$$W_x^{\text{THz}}(\psi) = W_x^{\text{THz}(\max)} (2 + \cos 2\psi)^2.$$
(9)

The dependences $W_y(\psi)$ and $W_x(\psi)$ are shown in Figs. 3(a) and 3(b) along with the experimental data. As we

can see, these expressions are in good agreement with the measurements.

For the case of the nonlinear interaction of circularly polarized fundamental ω radiation and the linearly polarized second harmonic ($\varphi_1 = \pi/2$, $\psi = \pi/4$ in Eq. (3)), the components of the nonlinear polarization oscillating at 3ω can be written in form

$$P_{y}^{(3\omega)}(t) = -\frac{\chi_{0}}{12} E_{\omega}(t) E_{2\omega}^{2}(t) \sin(3\omega t + 2\varphi_{2}), \qquad (10)$$

$$P_x^{(3\omega)}(t) = \frac{\chi_0}{4} E_\omega(t) E_{2\omega}^2(t) \cos(3\omega t + 2\varphi_2).$$
(11)

We see from Eqs. (10) and (11) that the third harmonic polarization is elliptical, the long axis of the ellipse is directed along the initial second harmonic polarization. The ratio of the major to minor axis of the polarization ellipse is 9. The corresponding angular dependence of the third harmonic intensity on the analyzer angle is shown in Fig. 4 with a solid line in good agreement with the experimental data (squares in Fig. 4).

The experimentally observed terahertz emission in case of the initial two-color field (Eq. (3)) was linear with the terahertz pulse energy independent from the phase difference between ω and 2ω optical fields. The 3rd order nonlinear polarization at terahertz frequency can be obtained from Eq. (2) using the input field (Eq. (3)) with $\varphi_1 = \pi/2, \ \psi = \pi/4$:

$$P_{y}^{(0\omega)}(t) = -\frac{\chi_{0}}{2} E_{\omega}^{2}(t) E_{2\omega}(t) \sin \varphi_{2}, \qquad (12)$$

$$P_{x}^{(0\omega)}(t) = \frac{\chi_{0}}{2} E_{\omega}^{2}(t) E_{2\omega}(t) \cos\varphi_{2}.$$
 (13)

We see that terahertz radiation is expected to have the linear polarization, with the terahertz electric field direction defined by the phase difference φ_2 . The total terahertz energy yield $W^{(0\omega)} = W_x^{(0\omega)} + W_y^{(0\omega)}$ is independent from φ_2 in agreement with the experimental observations (Fig. 5).

We note the stability of the terahertz radiation energy in this configuration of the circular polarized ω and the linearly polarized 2ω pulses. This stability attracts much interest for the problem of remote terahertz generation in femtosecond filaments. We have shown in Ref. [14] that in a long filament, circular polarization of the optical radiation is stable, so that the small distortions of the ω polarization or addition of 2ω radiation do not lead to the transformation of the fundamental pulse light field polarization state. In the turbulent atmosphere, the stochastic phase shift between ω and 2ω radiations can lead to the instability of generated terahertz radiation if both ω and 2ω pulses have linear polarization. In this case, terahertz signal can even disappear at the moments when ω to 2ω phase is close to some distinct values. The use of the circularly polarized ω pump radiation allows us to avoid this effect, so that the terahertz signal intensity remains, on average, at the same energy level throughout the experiment.

5 Conclusions

In conclusion, we have experimentally investigated simultaneous generation of three nonlinear-optical processes appearing in the course of light-gas interactions enhanced by geometrical focusing of the two-color femtosecond pulses into atmospheric pressure air. We have found that the energy of 2ω radiation, polarized orthogonally to its initial direction, follows the sine-square dependence $W_v^{(2\omega)}(\psi) = W_v^{2\omega(\max)} \sin^2 2\psi$ on the angle 2ψ between the initial linear polarization directions of the ω to 2ω fields. This dependence is described by the third order polarizability of the neutral air molecules. The major contribution of the third order nonlinear response of neutrals is further confirmed by the similar sine-square dependence of the newly appearing terahertz radiation after the analyzer crossed to the initial 2ω field linear polarization direction.

If the initial ω field is circularly polarized and the 2ω field is linearly polarized, the newly appearing 3ω radiation is elliptically polarized with its major axis of the ellipse directed along the 2ω linear polarization direction. The terahertz field polarization in this case is linear. The direction of this linear polarization changes randomly from one laser shot to another following the initial ω to 2ω phase shift. At the same time, the terahertz pulse energy remains stable independently of this phase shift.

The polarization analysis of the newly generated second harmonic component, the third harmonic and the widerange terahertz radiation is well described under the assumption of the instantaneous third-order nonlinear polarizability of neutral molecules in air in the strong light field.

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tion in gases and transparent solids. In collaboration with Prof. See Leang Chin (Laval University, Quebec, Canada), she pioneered the world-leading research in femtosecond laser pulse filamentation and was the first one to theoretically explain and simulate spatiotemporal pulse transformation and conical emission accompanying filamentation in air (1996 - 2000). She continued with optimization of transport and positioning of multiple filaments on a long-range atmospheric path (2000-2006). Since 2006, she has been the leader of the research projects on filamentation nonlinear optics using terawatt Ti:Sapphire laser facility of the Center for Collective Usage of the International Laser Center of MSU. Since 2010, she has worked on the sources of THz radiation from single- and dual-color filaments. In collaboration with Prof. Xi-Cheng Zhang's (University of Rochester) and Prof. Shkurinov's (Lomonosov MSU) groups, she defined the frequency-dependent contribution to the broadband spectrum of terahertz radiation of the free carriers and neutrals from the partially ionized filament core in gases.

Now Olga Kosareva is one of the leading world-known scientists in the field of pulse propagation and filamentation, she published 79 papers in peer-reviewed scientific journals and 88 papers in conference proceedings, she was invited to deliver more than 40 invited lectures and talks, organizes annually well-recognized international conferences (Laser Physics Workshop, ICONO/LAT, COFIL). Since 2002, she is Faculty Advisor of MSU SPIE Student Chapter (about 30 students).

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Nikolay Panov – Ph.D. (2009, Lomonosov Moscow State University (MSU)), Research Scientist with the International Laser Center of Lomonosov MSU. Since 2004, he has worked on the simulations of laser beam propagation in both regular and randomly-inhomogeneous media. He performed the optimization of the threedimensional plus time numerical algorithm

and the code for simulating nonstationary multiple filament formation in air. Based on this code, the Monte-Carlo simulations of femtosecond light filamentation in atmospheric turbulence were performed on both horizontal and vertical atmospheric path with the pressure gradient. Since 2009, Nikolay Panov developed the vectorial approach for the description of the polarization rotation of the probe pulse co-propagating with a high-peak power pump pulse creating a filament in gases. This approach was successfully extended to the general four-wave mixing case and allowed to efficiently follow the polarization of a newly born terahertz radiation due to the nonlinear ω -2 ω pulse interaction in the filament.

Since 2012, Nikolay Panov has been a project leader according to the special program for young scientists and has headed the study of the terahertz frequency-angular spectrum irradiated from femtosecond optical breakdown or filament in gases. Nikolay Panov has 32 journal publications and 53 publications in conference proceedings. His honors and awards include: Dynasty Foundation award for young researches with Ph.D. degree (2012), the Rector of Lomonosov MSU award for the series of publications (2012), SPIE Scholarship (2005).



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