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Noncollinear six-wave mixing of femtosecond laser pulses in air

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Noncollinear six-wave mixing of femtosecond light pulses in air by two crossed laser beams has been demonstrated. It was shown that within a wide range of laser intensities this process leads to efficient third-order-harmonic generation, which indicates the importance of higher-order optical nonlinearities in intense light-matter interactions. A direct comparison of the experiment and theory allowed us to estimate unambiguously the magnitude of the quintic optical nonlinearity $\chi^{(5)}_{xyyyyx}(-3\omega;\omega,\omega,\omega,\omega,-\omega)$ of air as 2×10^{-49} (m/V)^4. We expect that this technique could be useful for multiphoton spectroscopy and laser frequency conversion, as well as for ultrashort light-pulse characterization and verification of theoretical models describing propagation of powerful laser pulses in gaseous media.

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I. INTRODUCTION

Optical harmonic generation is a well-established technique to produce coherent radiation in spectral regions, where conventional lasers are not available [1]. In particular, third-order-harmonic generation (THG) in gaseous media is a source of coherent radiation in the ultraviolet (uv) and vacuum ultraviolet (vuv) [2]. However, efficient harmonic generation requires phase matching, which cannot be achieved in isotropic normally dispersive media, such as air and other gases. In addition, THG is a third-order nonlinear optical process, and therefore is most efficient at very high light intensity, which usually is achieved by focusing laser pulses [3]. Unfortunately, at high laser intensities, along with the THG, a number of competing processes, linear and nonlinear, such as Kerr-induced self-focusing and filamentation, conical emission, phase modulation, multiphoton or tunneling ionization, plasma absorption and defocusing, diffraction, and group-velocity dispersion are taking place [4–8]. Moreover, even under conditions of perfect phase matching, light generated prior to the focus destructively interferes with that generated beyond the focus and the total THG efficiency becomes negligible in the tight-focusing limit [9]. Nevertheless, it has recently been demonstrated that during filamentation of femtosecond laser pulses in air and other gases the THG efficiency can be as high as 0.2% [5,10]. However, the observed complicated spectral structure and high divergence of the generated radiation strongly limits the number of potential applications of such a light source [5,11].

On the other hand, the higher-order nonlinear polarization terms induced in a medium by intense femtosecond laser pulses may become comparable to or larger than the lower-order ones [12–14]. Thus, recently it was suggested [15] that conical third-order-harmonic emission generated by tightly focused femtosecond laser pulses in sodium vapor can be a result of noncollinear six-wave mixing (SWM). In addition, Bertrand *et al.* [16] have demonstrated that the noncollinear high-order-harmonic generation (HHG) also can be fully understood in terms of nonlinear wave interaction. However, recently a series of reports on third-order-harmonic (TH) enhancement by use of additional laser beams have been published [17–20]. Although such increase of THG efficiency has been explained mainly by the influence of laser-induced gas plasma, a full theoretical model of THG by a few laser beams still needs to be developed. This motivated us to study the role of quintic optical nonlinearity during frequency tripling of femtosecond laser pulses in air, when five laser photons are mixed together to create a TH photon [Fig. 1(a)]. In order to avoid or minimize the influence of other nonlinear optical processes, such as plasma-related effects and beam filamentation, we have chosen a moderate-power noncollinear two-beam pump configuration. Apart from other advantages this method allows one to control phase-matching conditions of nonlinear interactions via a simple change of the beam crossing angle [21]. Therefore in this paper we report a phase-matched frequency tripling of femtosecond laser pulses by crossed laser beams. In contrast to previous reports, where two crossed beams were also used for THG [17–20], this research was performed at very small laser-beam crossing angles, since our preliminary plane-wave analysis revealed that only within this region (at about 10 mrad) could phase-matching conditions for the SWM in air be satisfied.

II. THEORETICAL MODEL

For the theoretical treatment of noncollinear SWM we assume that the pump pulses propagate in slightly different directions along wave vectors $\vec{k}_1$ and $\vec{k}_1'$. The noncollinear third-order-harmonic generation in air can be described via the fifth-order susceptibility tensor $\chi^{(5)}_{q,p_1,p_2,p_3,p_4,p_5}(-3\omega;\omega,\omega,\omega,\omega,-\omega)$. The indices q and p_j $(j = 1,\dots,5)$ denote the polarization of the TH and pump waves, respectively. The wave-vector diagram illustrating phase-matched frequency tripling in isotropic normally dispersive media is sketched in Fig. 1(b). One can see that in this case the TH wave vectors are directed outward with respect to the intersecting crossed laser beams. Note that in the plane-wave approximation $n_1 \cos\alpha = n_3 \cos\beta$ and $5n_1 \sin\alpha = 3n_3 \sin\beta$. So noncollinear phase matching occurs when the fundamental beams intersect at an angle $2\alpha_0 \approx \sqrt{9\Delta n/2} \approx 10$ mrad, where $\Delta n = n_3 - n_1$, and n_1 and n_3 are the refractive indices of air for the fundamental and

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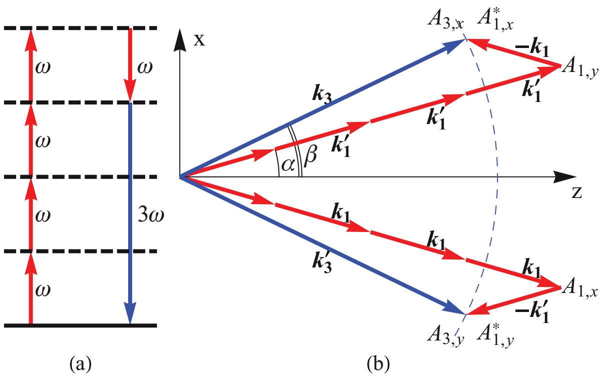

(a) (b)

FIG. 1. (Color online) (a) Photon energy diagram, and (b) wave-vector diagram of noncollinear SWM in air.

TH waves, respectively. In addition, based on the symmetry properties of the $\chi^{(5)}$ tensor one can deduce that in our experimental configuration frequency tripling via six-wave mixing is possible only through the fifth-order susceptibility tensor component $\chi^{(5)}_{xxxxxx} = \chi^{(5)}_{yyyyyy}$ for both pump beams of the same polarization and through the component $\chi^{(5)}_{xyyyyx} = \chi^{(5)}_{yxxxxy}$ for orthogonally polarized laser beams. Note that in the former case the polarizations of all interacting waves are the same, while in the latter, the polarizations of each TH beam should be orthogonal to that of the adjacent pump beam. Since light waves of linear orthogonal polarizations do not interfere and, thus, do not create the spatial intensity modulations which result in more efficient ionization of air molecules, most of our experiments were performed with orthogonally polarized laser beams.

A rigorous theoretical analysis of the propagation of an intense ultrashort laser pulse in gases at high pressure requires the inclusion of the full spatiotemporal dynamics of both the fundamental and TH pulses. However, under the circumstances of the experiment it seems that the effects related to the pulse group velocity and dispersion can be ruled out. Therefore our noncollinear six-wave-mixing model is described under the nondepleted pump approximation by a set of equations which can be written for slowly varying complex amplitudes in the retarded coordinate system ($z \to z, t \to t - z/v_g(\omega)$) as

$$\frac{\partial A_{3,x}}{\partial z} + \frac{1}{2ik_3}\Delta_\perp A_{3,x} = i\sigma e^{i\Delta kz} A^4_{1,y}(\alpha) A^*_{1,x}(-\alpha), \tag{1}$$

$$\frac{\partial A_{3,y}}{\partial z} + \frac{1}{2ik_3}\Delta_\perp A_{3,y} = i\sigma e^{i\Delta kz} A^4_{1,x}(-\alpha) A^*_{1,y}(\alpha), \tag{2}$$

where $A_{j,p}$ is the slowly varying complex amplitude for the fundamental ($j = 1$) or TH ($j = 3$) beam polarized along $p = x$ or y axis. $\sigma = D^{(5)} k_3 \chi^{(5)}/(2n_3^2)$, $D^{(5)} = 5$ is the degeneracy factor, $\chi^{(5)}$ denotes either $\chi^{(5)}_{x,y,y,y,y,x}$ or $\chi^{(5)}_{y,x,x,x,x,y}$, $\Delta k = 3k_1 \cos\alpha - k_3$, and 2α is the crossing angle of the pump beams. The pump pulse in its own coordinate system is described by the Gaussian function

$$A_{1,p}(x,y,z) = \frac{a_{1,p}}{1 + iz/L_d} \exp\left[-\frac{1}{1 + iz/L_d}\frac{x^2 + y^2}{w^2} - \frac{t^2}{\tau^2}\right], \tag{3}$$

where $a_{1,p}$ is the amplitude, $L_d = k_1 w^2/2$ is the diffraction length, w is the beam waist radius, and τ is the pulse width. The slowly varying amplitude of the tilted beam is defined as $A_{1,p}(\alpha) \equiv A_{1,p}(x\cos\alpha - z\sin\alpha, y, z\cos\alpha + x\sin\alpha)$. The far-field structure of the TH is calculated by first numerically solving Eqs. (1) and (2) for $A_{3,p}$ ($p = x, y$) in the pump-beam overlapping region $0 \leqslant z \lesssim 2w/\sin\alpha$, and then calculating the spectrum

$$S_{3,p}(k_x,k_y) = \int_{-\infty}^{\infty} dx\, e^{-ik_x x} \int_{-\infty}^{\infty} dy\, e^{-ik_y y} A_{3,p}(x,y). \tag{4}$$

This simple model gave us an excellent agreement between the theoretical predictions and experimental observations even for the pump power $P > P_{\rm cr}$[1] needed for self-focusing and filamentation in air [22].

III. EXPERIMENTAL RESULTS AND DISCUSSION

For the experiments we have used a 1 kHz repetition rate femtosecond Ti:sapphire laser system delivering 100 fs (FWHM) light pulses centered at 800 nm with a pulse energy of 2.1 mJ. The laser pulse energy could be varied by an attenuator composed of a zero-order half-wave plate and a broadband polarizer. The fundamental laser beam was divided into two almost equal parts by a thin beam splitter. An additional attenuator was used to adjust the energy of one of the beams. A half-wave plate inserted in the other beam path allowed us to obtain orthogonal polarizations of these two beams. Both laser beams were focused by a lens of 100 cm focal length and the front parts of the filaments were overlapped at a small crossing angle in the focal plane of the lens. The time delay between the incidence of two pulses in the overlap region could be varied mechanically by an optical delay line. For the energy measurements performed by calibrated power meters the fundamental and TH beams were separated by three successive dielectric harmonic separators and calibrated optical filters. Far-field patterns of TH radiation generated in air were observed on a fluorescent paper screen placed about 1.5 m beyond the beam intersection point. For large delay times and pulse energies of more than 500 μJ each laser beam produced a weak TH signal consisting of a central spot surrounded by a ring [5,11]. (5–10)-cm-long filaments, appearing at almost the same pump power level, could also be observed. When the fundamental pulses overlapped in space and time, as predicted by the theory [see Fig. 2(b)], two bright TH beams outside the crossed pump were observed on a screen for a wide range of beam crossing angles. Typical experimentally registered far-field patterns and polarizations of the pump and TH produced by the noncollinear six wave interaction are shown in Fig. 2(c). As can be seen, the TH signal consists of two orthogonally polarized spots at the locations well predicted theoretically. An even better match between the experimental data and theory can be seen in the dependence of the normalized energy of the TH generated noncollinearly on the crossing angle 2α of the pump beams [Fig. 3(a)]. From this correspondence, taking the absolute values of the TH pulse energy, we have estimated the value

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[1] The critical power needed for self-focusing and filamentation in air is $P_{\rm cr} = \lambda_1^2/(2\pi n_1 n'_2) \approx 2.6$ GW, where $n'_2 = 4 \times 10^{-19}$ cm^2/W, $n_1 = 1 + 2.7 \times 10^{-4}$, and $\lambda_1 = 0.8$ μm.

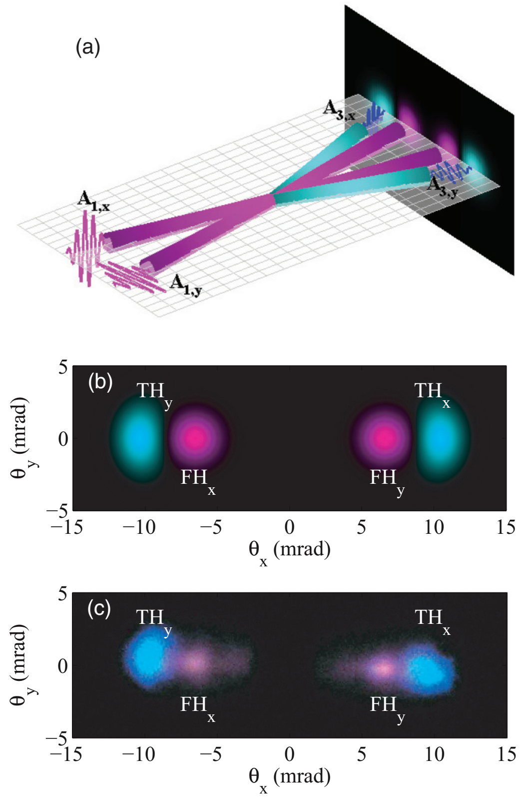

FIG. 2. (Color online) (a) Experimental configuration illustrating a two-beam six-wave mixing in air with the far-field patterns of the fundamental and TH for a beam crossing angle of 13 mrad (the total energy of both laser pulses was about 1.9 mJ); (b) numerical simulations; and (c) the experimentally obtained data.

of $\chi^{(5)}_{xyyyyx}(-3\omega;\omega,\omega,\omega,\omega,-\omega) \approx 2 \times 10^{-49}$ $(\mathrm{m/V})^4$. Note that the TH signal could be easily registered for low laser pulse energies (of less than 200 μJ for each pump beam), when no apparent filamentation and laser-induced plasma formation evidence could be observed, which indicates that plasma-related effects play little role in our experiment. The experimentally registered dependencies of the TH signal on the pump-pulse energy also showed a good agreement with theoretical predictions. Thus, as can be shown from Eqs. (1) and (2), while the total TH yield should be proportional to the fifth power of the pump intensity, the power of each TH beam should scale as the product of $I_{1,x}I^4_{1,y}$ and $I_{1,y}I^4_{1,x}$, where $I_{1,p}$ corresponds to the intensity of the pump beam. This has been confirmed experimentally [see Figs. 4(a) and 4(b)] over the full range of available laser powers and additionally indicates that during such noncollinear SWM each TH photon results from the combination of four photons from one laser beam and a single photon from the other.

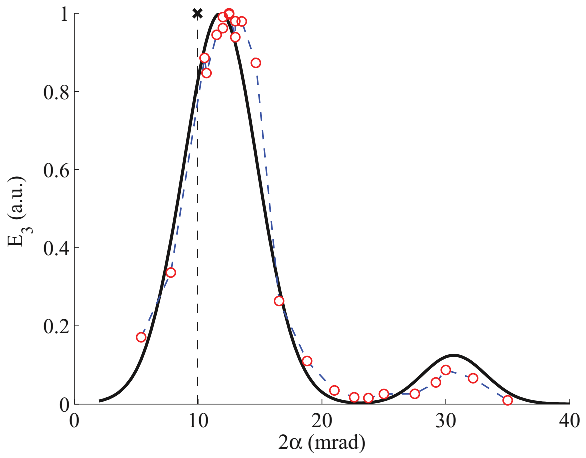

FIG. 3. (Color online) Dependence of TH yield of noncollinear SWM on the beam crossing angle 2α. The vertical dashed line corresponds to the plane-wave approximation.

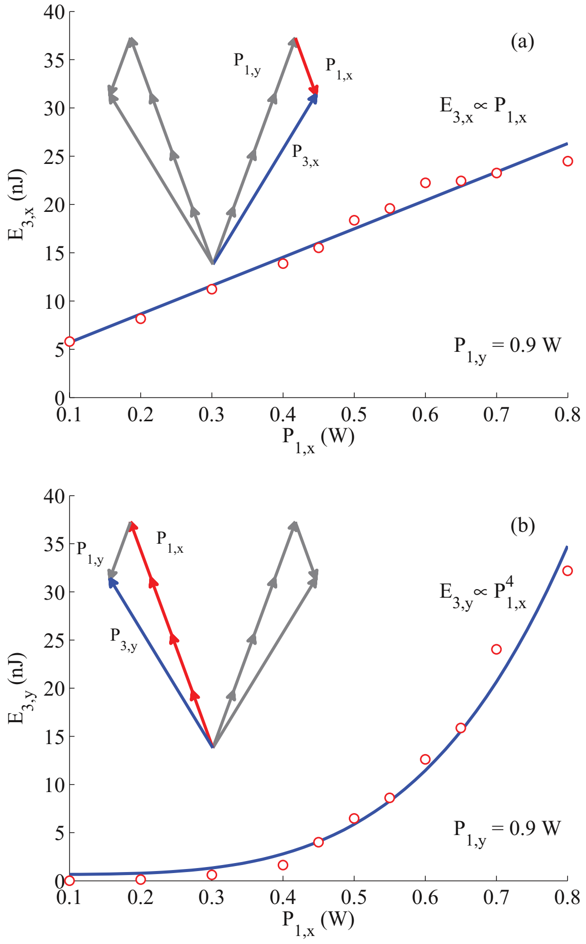

FIG. 4. (Color online) Dependence of the single-beam TH yield of noncollinear SWM on a single laser-beam power for (a) constant and (b) varying adjacent laser beam energies.

At the maximum laser-pulse energy of about 2 mJ the total TH output could reach over 200 nJ, which corresponds to a conversion efficiency of about 10^{-4}. Note, however, that the maximum TH pulse energy generated by a single laser beam, focused by a lens of the same focal length, did not exceeded a few nanojoules; thus, under the given experimental conditions, the noncollinear THG was about by two orders of magnitude more efficient.

The results of numerical calculations and the experiment show that we have observed phase-matched noncollinear SWM leading to third-order-harmonic generation in air. The fact that all the observed characteristics of noncollinear THG well correspond to the theoretical predictions indicates that within the range of laser parameters used (laser wavelength, pulse duration and energy, beam intensity, and focusing conditions) a perturbative approximation of nonlinear optics is still valid. On the other hand, this process should also be relevant at extremely high laser intensities, when the perturbative expansion of the polarizability is no longer valid, and higher-order processes may be as probable as the direct THG process. Since the dispersion of gases is small, the output generated from such a SWM process can be phase matched under a broad range of experimental conditions, whenever tight pump focusing into nonlinear media takes place or when the laser divergence exceeds some critical angle (necessary to achieve phase-matched SWM). Note here that even when the process of six-wave mixing is phase mismatched for collinear interacting waves, vectorial phase matching still can be satisfied since the focused fundamental beam contains some angular spread of its components. Thus, the phase matching in most cases is expected to be wideband and SWM can also be used to generate ultrashort or broadband light pulses. In addition, the two-beam noncollinear SWM technique could easily be adopted for the characterization of powerful ultrashort pulses, i.e., for the measurement of background free fifth-order autocorrelations, which can be asymmetric and would thus yield additional information on the asymmetry of the pulse shape, which is not available from the common second-order autocorrelations [23]. The noncollinear scheme of THG also offers the advantage of separating the fundamental and generated light spatially, which usually in the uv and vuv spectral regions is a serious technical problem. Furthermore, such a noncollinear third-order-harmonic generator can act as a tunable all-optical beam splitter, providing a well-controlled TH beam intensity ratio and desirable output polarizations.

IV. CONCLUSIONS

We have demonstrated a phase-matched frequency tripling of femtosecond laser pulses through noncollinear six-wave mixing in air. The application of two crossed laser beams of orthogonal polarizations has allowed us to separate well the quintic optical process from the lower-order ones, and to estimate roughly the $\chi^{(5)}(-3\omega;\omega,\omega,\omega,\omega,-\omega)$ value in air. Note that, though our results deal only with the one type of quintic optical nonlinearity responsible for six-wave mixing, one may expect that other types of higher-order nonlinearities are also playing an important role in a number of nonlinear optical processes, such as harmonic and supercontinua generation, when powerful ultrashort laser pulses are propagating through gaseous media.

Finally, note that the proposed noncollinear SWM could be further optimized and opens avenues not only for efficient frequency tripling over a wide range of wavelengths, but also for a variety of scientific and technological applications including all-optical photonic beam splitters and switches, the characterization of powerful ultrashort laser pulses, remote sensing, and spectroscopy of atmospheric pollutants.

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