Appl. Phys. B 71, 581–584 (2000) / Digital Object Identifier (DOI) 10.1007/s003400000389

Applied Physics B
Lasers
and Optics

Intense femtosecond laser-induced second-harmonic generation in atmospheric-pressure air

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Received: 23 May 2000/Published online: 16 August 2000 –

Abstract. Second-harmonic generation (SHG) in atmospheric-pressure air is experimentally studied using single focused linear-polarized Ti:sapphire intense femtosecond laser pulses at 810 nm. The efficiency of SHG is found to reach a maximum at the optical breakdown threshold of $\approx 2.9 \times 10^{14}\,\mathrm{W/cm^2}$. The spectral distribution and polarization property of the second harmonic are investigated. The contribution to SHG from electric-field-induced third-order mixing plays the main role even after the optical breakdown had occurred.

PACS: 33.80.Rv; 42.65.Ky; 52.40.Nk

Recently, much attention has been concentrated on high-order harmonic generation by means of interaction between intense laser pulses and gaseous media. The results of these investigations are important for developing sources of coherent short-wavelength radiation. Meanwhile, such research provides an opportunity to reveal specific features of nonlinear optical interactions in intense light fields. Usually, even-order harmonics are observed when a high-power ultrafast laser pulse is focused into a gas although these harmonics are strictly forbidden in centrosymmetric media under the dipole approximation. The second harmonic (SH), as the lowest even-order harmonic, has been observed in metal vapor or in gaseous media [1–15]. Various mechanisms, including resonant high-order multipole transitions [8], third-order susceptibility $\chi^{(3)}$ with external-applied or resonance-enhanced ionization-induced dc electric field [9, 10], inhomogeneous plasma [11, 12] and collision-assisted mechanism [13–15], have been proposed.

When an intense ultrafast (picosecond or femtosecond) laser beam is focused into a gas in the absence of any resonant effects and external-applied electric or magnetic fields, the medium in the focus is usually a mixture of neutral molecules (or atoms) and plenty of plasma. Fedotov et al. [3] investigated the second-harmonic generation (SHG) of 90-fs 798-nm Ti:sapphire laser pulses in atmospheric-pressure air. They measured the efficiency of the SH at different pulse energies of the fundamental light; the maximum efficiency was estimated to be 10^{-6}. The self-action of light was proven to noticeably modify the efficiency. Liu et al. [4] measured scaling of SHG in hydrogen with different pressures by focusing 1-ps/1.053-μm Nd:glass laser pulses and obtained a square dependence. In this paper, we present a systematic study on SHG with focused intense fs laser pulses in atmospheric-pressure air. Both the spectral distributions of the SH signal and the fundamental light after interaction are investigated in detail for the first time. It appears that the SH signal is generated from self-phase-modulated fundamental light and experiences self-phase modulation itself after its generation. The efficiency of the SH is also measured for various fundamental light intensities exhibiting a maximum at the optical breakdown threshold. This indicates that the saturation of the SH is due to plasma formation.

1 Experimental setup

The experimental setup for SHG is based on a Ti:sapphire chirped-pulse amplification system (TSA-10, Spectra-Physics Inc., USA) delivering 810-nm pulses with a repetition rate of 10 Hz, a pulse duration of 110 fs, and a maximum energy of 10 mJ per pulse. As shown in Fig. 1, the fundamental light with linear polarization is focused to a spot of 50 μm

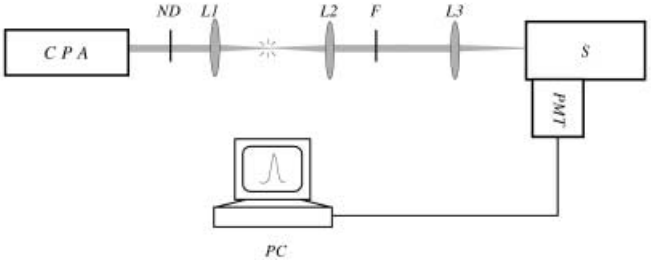

Fig. 1. Experimental setup. CPA: chirp-pulsed amplification laser system; ND: neutral-density optical filter; L1–3: focal lenses; F: band filter; S: spectrometer; PMT: photomultiplier tube; PC: personal computer

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diameter (about two-times diffraction limited) with a lens L1 ($f = 20$ cm). The maximum focal intensity corresponds to 4×10^{15} W/cm^2. In the experiments, we varied the energy of the fundamental light by inserting different neutral-density optical filters (ND). The optical breakdown in air manifested itself by the appearance of a spark in the focus. Radiation emerging from the focus was recollimated by a quartz lens L2. Spectral components around the fundamental frequency were then filtered by an appropriate band filter (F). Radiation transmitted through F was collimated with another quartz lens L3 and fed into a spectrometer. The SH signal was detected by a photomultiplier tube (PMT, R456, Hamamatsu, Japan) and processed by a linked personal computer (PC).

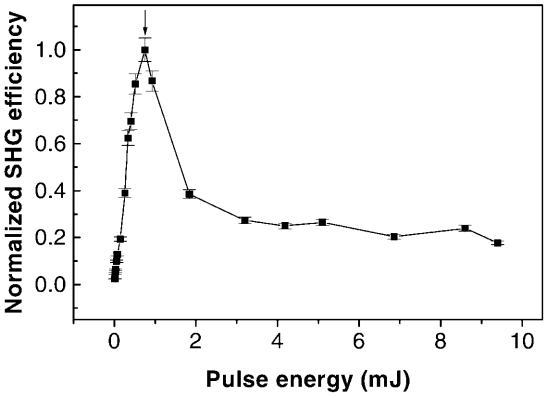

Fig. 3. Normalized SHG efficiency as a function of pulse energy of fundamental light

2 Results and discussions

Figure 2 shows the energy of the SH signal as a function of the pulse energy of the fundamental light. The energy increases with increasing fundamental pulse energy. A remarkable feature revealed from Fig. 2 is that there exists a distinctive inflexion as indicated by an arrow in Fig. 2. It was confirmed that the inflexion just corresponded to the visible optical breakdown threshold. This inflexion point at 2.9×10^{14} W/cm^2 (pulse energy 0.75 mJ) is consistent with that of about 2×10^{14} W/cm^2 as obtained in [4]. Such an inflexion point suggests that the interaction mechanism between the laser pulse and air experiences some changes when the pulse energy passes through the inflexion. When the fundamental pulse intensity is lower than the optical breakdown threshold, the fundamental light interacts with neutral molecules and a few ionized particles generated through multiphoton ionization, whereas for intensities higher than the threshold, the fundamental light interacts with a highly-ionized plasma. The slope below the optical breakdown threshold is 1.9 ± 0.1, which is in reasonable agreement with 2.05 ± 0.1 reported in [3]. We found that this value has a slight dependence on the focal length of L1. For instance, when a lens of 30 cm focal length was employed, the slope below the optical breakdown threshold was 1.6 ± 0.1. Figure 3 shows the efficiency of the SH signal normalized to its peak value as a function of the pulse energy of the fundamental light. The efficiency has a maximum just at the optical breakdown threshold (the absolute maximum efficiency is about $10^{-7}-10^{-6}$), as shown by an arrow in the figure.

Investigation of the spectral distribution of the SH signal is helpful in understanding the relevant processes involved in SHG. We measured the spectral distribution of the SH signal at different pulse energies of the fundamental light. For high fundamental pulse energies (Fig. 4a,b), the spectrum of the SH signal is asymmetric and exhibits structures that are characteristic of self-phase modulation [3]. This spectral modulation may simultaneously result from two mechanisms. One is that the SH signal is generated with modulated fundamental light; the other is that the SH signal experiences modulation after generation. In the experiments, we directly measured the spectrum of the fundamental light after interaction. As shown in Fig. 5 at high pulse energy, the spectrum is greatly blue-broadened while there is nearly no evident broadening in the red region. This indicates that the spectrum variation is solely due to self-phase modulation in the produced

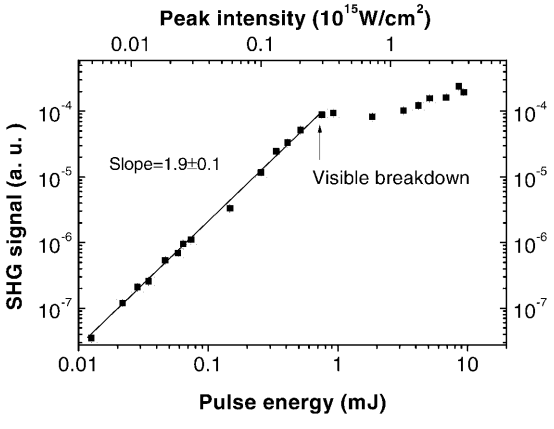

Fig. 2. SHG signal as a function of pulse energy of fundamental light

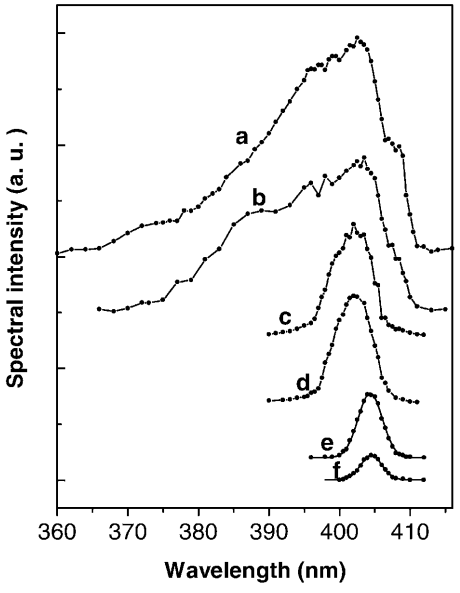

Fig. 4. Spectral properties of the SH signal at different fundamental pulse energies: (a) 9.4 mJ; (b) 6.8 mJ; (c) 0.92 mJ; (d) 0.72 mJ (optical breakdown threshold); (e) 0.025 mJ; (f) 0.0025 mJ. *Solid curves* in (e) and (f) are Gaussian-fitted lines

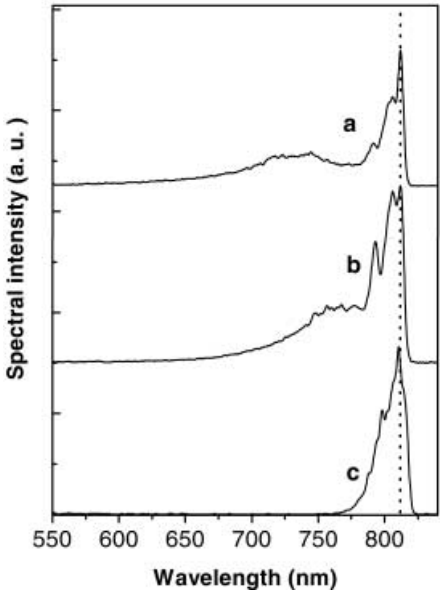

Fig. 5. Modulated fundamental laser spectra at different fundamental pulse energy: (a) 9.4 mJ; (b) 6.8 mJ; (c) 0.72 mJ (optical breakdown threshold). The *dotted line* indicates the fundamental center wavelength of 810 nm

plasma [16–21]. Thus, the blue-broadening of the fundamental light may account for the long blue-side of the SH signal shown in Fig. 4a,b. At the same time, the blueshift of the peak of the SH signal and the shoulder in the red side (≈ 407 nm) suggest that the second harmonic itself also experiences self-phase modulation. Decreasing the energy of the fundamental light causes the spectrum of the SH signal to become more symmetric and narrow spectrally. At the optical breakdown threshold or below, the structures of the SH signal vanish and the spectral profile becomes increasingly smoother and more and more Gaussian, as shown in Fig. 4d–f.

To illustrate the origin of the SH signal, we checked its polarization above and below the optical breakdown threshold by putting a polarizer (analyzer) before the spectrometer. The SH linear component was found to increase when the optical breakdown occurred. Figure 6 shows the dependence of the normalized SH signal on the angle between the polarizer axis and the fundamental light polarization. When the angle varies from 0° to 90°, the SH signal varies by a factor of 1/0.32 at

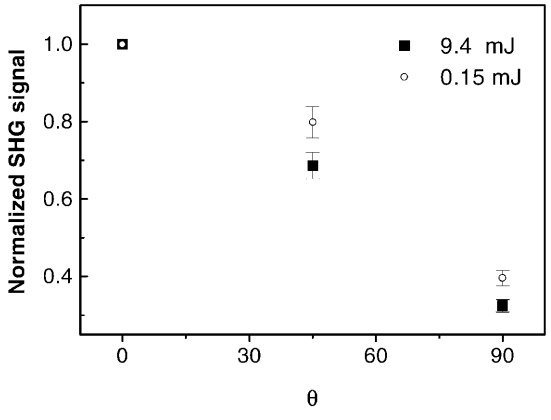

Fig. 6. Polarization properties of SHG. θ is the angle between the polarizer axis and the fundamental light polarization

a pulse energy 9.4 mJ, and 1/0.4 at a pulse energy of 0.15 mJ. The SH signal is not linearly polarized, neither above nor below the optical breakdown threshold. This indicates that the third-order nonlinear mixing via atomic nonlinear susceptibility ($\chi^{(3)}$) between the laser field and induced-electric-field due to ionization is undoubtedly involved in SHG in the parameter range of our study. Such nonlinear mixing would result in radial polarization of SHG [5–7], and in most cases, the SH signal is nonuniformly radially polarized [1].

When the laser intensity is higher than 10^{11} W/cm^2, atoms and molecules can be ionized although likely with a low ionization rate [22]. Figure 4d–f demonstrates the influence of the ionization. The spectral width of the SH signal becomes wider and the center wavelength gets more blueshifted upon increasing the fundamental pulse energy. When the fundamental light intensity (or energy) exceeds the optical breakdown threshold, air molecules are ionized in large numbers and an absorbing, refracting, and scattering plasma is formed. Then another mechanism begins to contribute to SHG. The second-order response of the nonuniform plasma could also give rise to SHG. For a nonuniform plasma, there exists a net charge density that would give a 2ω polarization (ω is the angular frequency of the fundamental light):

$$\boldsymbol{P}^{(2)}(2\omega) = \frac{2n_e e^3}{4m^2\omega^2}\,\frac{\boldsymbol{E}(\boldsymbol{E}\cdot\nabla \ln n_e)}{\varepsilon_p}, \tag{1}$$

where $\varepsilon_p = 1 - \frac{\omega_p^2}{\omega^2}$ is the square of the refractive index, n_e the electron density, e the charge of an electron with mass m, ω_p the plasma frequency, and $\boldsymbol{E}$ the laser field. No bulk 2ω radiation can occur if n_e is uniform. The SH directly radiated by $\boldsymbol{P}^{(2)}(2\omega)$ has the same linear polarization as that of the fundamental, as shown by (1). The occurrence of more parallel SH components (with respect to the fundamental laser polarization) at fundamental pulse energies exceeding the optical breakdown threshold indicated that the second-order polarization due to nonuniform plasma begins to contribute to SHG. However, it is difficult to accurately distinguish the 2ω contribution radiated by $\boldsymbol{P}^{(2)}(2\omega)$ from that radiated by $\boldsymbol{P}^{(3)}(2\omega)$ even when taking into account their different polarization properties. Although $\boldsymbol{P}^{(2)}(2\omega)$ would play an important role at very high ionization levels [11], SH generated by $\boldsymbol{P}^{(2)}(2\omega)$ is potentially much smaller than that by $\boldsymbol{P}^{(3)}(2\omega)$, as suggested by Fig. 6.

As high-order multipole contributions (quadrupole and magnetic-dipole) from neutral gaseous media can only be detected in very resonant cases [1], they are negligible in our experiment. Additionally according to thermodynamics, the average collision time between two molecules is about 154 ps for 1-atm air at room temperature, which is three orders of magnitude longer than the laser pulse duration of 110 fs. Thus, the collision-assisted mechanism only contributes marginally to SHG.

When the fundamental light intensity exceeded the optical breakdown threshold, a lot of plasma was produced, which leads to a new medium to interact with the laser pulses. This may be the reason why the SH begins to saturate and its efficiency decreases when the fundamental light intensity exceeds the optical breakdown threshold. Rapid plasma formation would greatly broaden the spectrum of fundamental light and decrease the laser intensity. Meanwhile, the plasma can effectively defocus the fundamental light and further lower

the laser intensity [19]. The generated ions have a lower polarizability than the neutrals and hence contribute less effectively to SHG. Moreover, plenty of free electrons would change the refractive index and remarkably alter the phase-matching condition [23] between the laser and induced electric fields resulting in low SH efficiency.

3 Conclusion

We have studied the SHG with focused intense laser pulses in atmospheric-pressure air by measuring its spectral and polarization properties. The spectral investigation of the SH signal and the fundamental light suggests that the SH is generated by self-phase-modulated fundamental light and also experiences self-phase modulation itself. The polarization property of the SH signal shows that the main contribution is due to third-order mixing between the laser and ionization-induced electric fields. A smaller contribution comes from the second-order polarization of the non-uniform plasma produced when the fundamental pulse intensity exceeds the optical breakdown threshold. The efficiency of the SH signal displays a maximum at the optical breakdown threshold. The results also reveal that plasma formation is disadvantageous for both efficiency and spectral quality of the SH signal.

Acknowledgements. The work was supported by the National Climbing Project (97.3) of China under Grant No. G1999075207 and the National Natural Science Foundation of China under grant No. 19884001 and 19525412.

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