High average power second harmonic generation in air

Martynas Beresna,[1,a)] Peter G. Kazansky,[1] Yuri Svirko,[2] Martynas Barkauskas,[3] and Romas Danielius[3]
[1]*Optoelectronics Research Centre, University of Southampton, Southampton SO17 1BJ, United Kingdom*
[2]*Department of Physics and Mathematics, University of Joensuu, Joensuu FI-80101, Finland*
[3]*Light Conversion Ltd., Vilnius, LT-10223, Lithuania*

(Received 29 May 2009; accepted 17 August 2009; published online 22 September 2009)

We demonstrate second harmonic vortex generation in atmospheric pressure air using tightly focused femtosecond laser beam. The circularly polarized ring-shaped beam of the second harmonic is generated in the air by fundamental beam of the same circular polarization, while the linear polarized beam produces two-lobe beam at the second harmonic frequency. The achieved normalized conversion efficiency and average second harmonic power are two orders of magnitude higher compared to those previously reported and can be increased up to 20 times by external gas flow. We demonstrate that the frequency doubling originates from the gradient of photoexcited free electrons created by pondermotive force. © *2009 American Institute of Physics*.
[doi:10.1063/1.3232235]

Progress in high power ultrashort pulse lasers has provided deeper insight in physics of light-matter interaction. At high intensities atoms in gaseous media are ionized and plasma in the interaction volume is formed giving rise to such phenomena as coherent x-ray generation and nonlinear Thomson scattering.[1] Particularly attractive medium due to its abundance is ambient air, in which an intense femtosecond pulse can emit terahertz radiation[2] or control atmospheric discharge.[3] Moreover, it has been demonstrated that in gaseous media, under radiation forces photoelectrons[4] can give rise also to the frequency doubling,[5] what is otherwise forbidden under the dipole approximation. Most of the experiments on second harmonic generation (SHG) in gaseous media or metal vapor were carried by focusing high energy pulses with low numerical aperture (NA) optics. On the contrary, here we demonstrate SHG in atmospheric pressure air using tightly focused femtosecond laser beam with linear and circular polarizations. The conversion efficiency of 4×10^{-6} is achieved, corresponding to two orders of magnitude increase in normalized efficiency (which here is defined as efficiency divided by the pump peak power) and average second harmonic (SH) power compared to previously reported.[6] The SHG mechanism is explained in terms of the pondermotive force induced free electron gradient. Further, generation of SH vortex in accordance with angular momentum conservation is demonstrated.

Experiments were performed with Yb:KGW (Yb-doped potassium gadolinium tungstate) based femtosecond system (Pharos, Light Conversion Ltd.) that delivered pulses of 270 fs with repetition rate of 200 kHz at 1025 nm. The power delivered to the focus was varied from 0.15 to 2.5 W with an achromatic half-wave plate and Glan polarizer (Fig. 1). Consequently, the maximum radiation intensity in the focal spot was 10^{15} W/cm^2 (i.e., it was above the air ionization threshold of about $\sim10^{14}$ W/cm^2) producing plasma with concentration of about 10^{17} cm^{-3}. Linear polarization (LP) of the incident beam was rotated with the second achromatic half-wave plate, which was replaced with an achromatic quarter-wave plate when circular polarization (CP) was studied. The beam after being reflected from a dichroic mirror was focused with an infinity-corrected objective (Mitutoyo $\times$100/0.7 NA M Plan Apo NIR HR). Additionally, the effect of focusing on the SH generation was investigated using lower NA (0.2 and 0.55) objectives. Light after the focus was collected with microscope condenser. The focal spot was observed through dichroic mirror with coupled-charge device (CCD) camera.

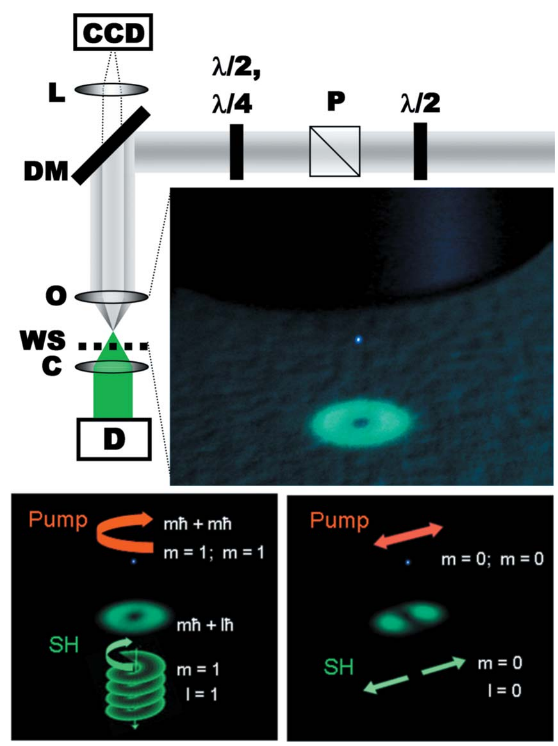

FIG. 1. (Color online) (Top) Experimental setup for generating SH in air: Half-wave plate and Glan polarizer (P), dichroic mirror (DM), objective (O), condenser (C), CCD camera (CCD), white screen (WS), power meter (D). (Inset) Image of SH pattern for circular polarization. On top of the photography the objective's edge can be seen. (Bottom) SH vortex generated with circularly polarized pump in accordance with angular momentum conservation (Left). Two-lobe SH pattern with π shifted electric fields for linearly polarized pump (Right).

a)Electronic mail: mxb@orc.soton.ac.uk.

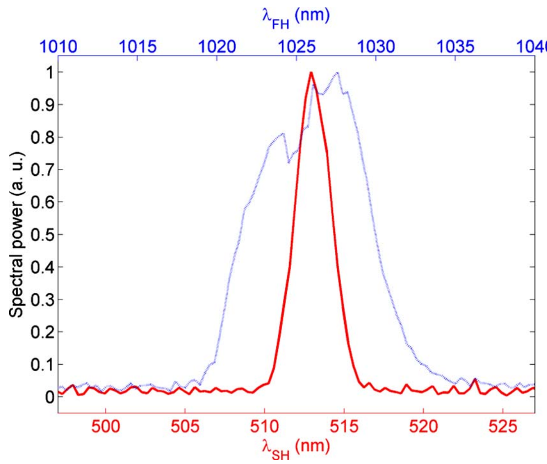

FIG. 2. (Color online) Spectra of the first and second harmonics.

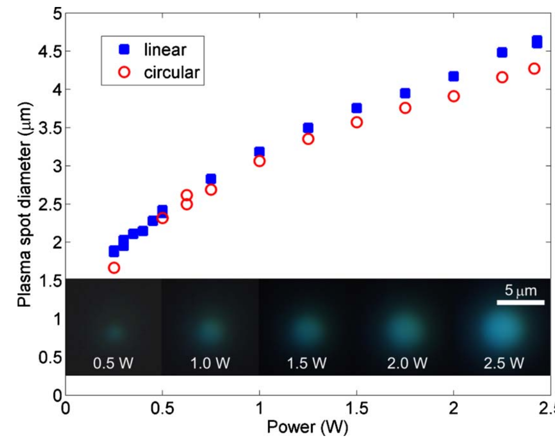

FIG. 4. (Color online) Plasma spot diameter d_{FWHM} dependence on pump power. (Inset) CCD images of plasma spot evolution with pump power increase. Numeric values indicate pump power delivered to the focus.

Once the laser beam was focused with the objective small bluish spherical spot appeared in the focus with coherent green light emerging from it was viewed on the white screen placed after the beam focus (Fig. 1). The spectral analysis confirmed that the observed green light is SH of the fundamental laser frequency (Fig. 2). The shape of SH pattern depended on the fundamental beam polarization. LP produced two lobes aligned along the polarization direction. For the CP light the doughnut shape was observed. We observed a quadratic dependence of the SH generation on fundamental beam power with approximately 1.4 times stronger signal for CP (Fig. 3). The estimated maximum SH conversion efficiency was about 4×10^{-6}.

The third harmonic, however, visualized via blue luminescence of the paper screen was much weaker than the SHG. This can be explained by the fact that a circularly polarized fundamental beam cannot produce third harmonic radiation in isotropic medium and the tight focusing of the pump. Only when the fundamental beam was focused with lower NA objectives and linearly polarized, we observed blue luminescence. This demonstrates crucial role of NA in the SHG in air.

The power of the fundamental beam measured behind the focal plane was a linear function of the incident power with the same slope for LP and CP. This indicates negligible plasma absorption.

The diameter of the ionized area d_{FWHM} was estimated by fitting with the Gaussian function the spot profile measured with the CCD camera (Fig. 4). It increased with pump power from about 1.8 μm, corresponding to the spot size in the focus, to 4.6 μm. For the CP d_{FWHM} was about 7% smaller than for LP. This change, however, did not affect SH far field pattern size that supports our assumption of the ponderomotive force being responsible for the observed frequency doubling.

Another interesting observation was a significant (about three times) growth of the SH signal accompanied by the plasma spot diameter increase when a weak stream of nitrogen gas (Fig. 3) or compressed air was pointed at the focal area. The strong signal increase was observed at pressure below 0.1 bar (the lowest pressure measurable with our setup). Yet higher gas pressure had produced merely several percent additional increase in the SH signal. The nitrogen stream was accompanied with yellow-orange light tail (emerging from plasma spot and pointing in the direction of gas flow) and strong odor typical for nitrogen dioxide (NO_2). This glow can be attributed to chemiluminescence.

Even stronger effect was noticed with compressed hydrofluorocarbon (HFC) gas flow from air duster (Fig. 5). This time the twenty and fourfold increase in the SH signal was measured at the pump powers of 0.65 and 2.45 W, re-

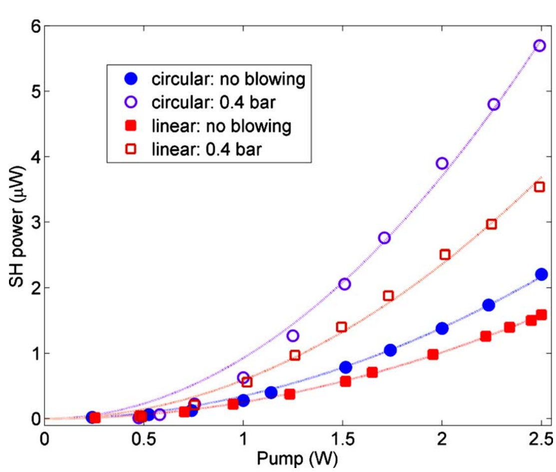

FIG. 3. (Color online) SH power as a function of the pump power. Dashed lines show fit of data with quadratic function.

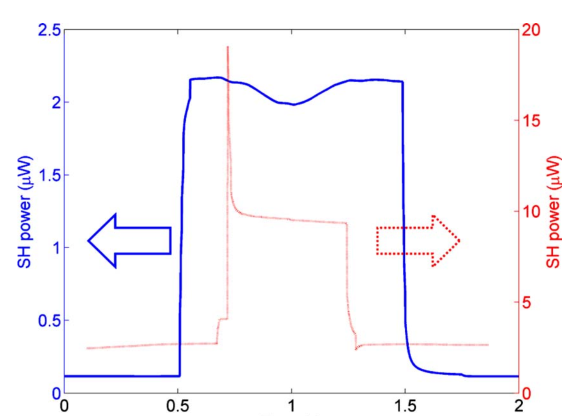

FIG. 5. (Color online) SH power change when compressed HFC gas is blown at the focus spot for two pump powers. The SH increases about 20 times at 0.65 W pump power (continuous blue) and about 4 times at 2.45 W (dotted red).

spectively. Consequently, the measured power of signal was as high as 10 μW. Additional sharp peak was observed in the wake front of SH power. If, however, laser beam was physically interrupted, while gas was blowing, the SH signal wake front was without this peak. Currently, the origin of this peak needs to be further investigated.

The described above phenomenon also supports the assumption that the pondermotive force dominates the SHG process. Indeed, the pondermotive force pushes electrons until attraction between separated positive and negative charges counterbalance this force. The external gas flow drags positive ions out of the focus weakening the electric field strength and leading to stronger electron concentration gradient and thus to the enhanced SH emission.

Demonstrated results are explained as follows. SH could be generated in isotropic gaseous medium if the plasma in the irradiated volume is inhomogeneous.[7] Two mechanisms can give rise to the nonzero gradient of the electron concentration.[8] First one is due to the inhomogeneous distribution of the electron density produced by the Gaussian fundamental beam in the focal area. We assume that at our laser intensities the ionization is saturated and the ionized region is bigger than the pump spot size, thus this mechanism does not explain the observed SHG. The second mechanism is based on the pondermotive force,

$$f_e = -\frac{e^2}{4m\omega^2}\nabla_\perp \boldsymbol{E}^2, \tag{1}$$

where $\nabla_\perp$ is gradient perpendicular to the beam propagation direction. This force is directed out of the center of the pump and thus creates inhomogeneous distribution of electrons pushing them from the center of the focused beam and giving rise to the SHG.

Frequency doubling in the isotropic plasma with density n_e can be described in terms of the second-order polarization:[4,7]

$$\vec{P}(2\omega) = \chi\left[(\vec{E}\cdot\nabla)\vec{E} + \frac{\iota\omega}{c}\vec{E}\times\vec{B}\right] + \frac{e\vec{E}(\nabla\cdot\vec{E})}{8\pi m\omega^2}, \tag{2}$$

where $\chi = n_e e^3/4m^2\omega^4$ By using condition of the plasma neutrality $\nabla(\varepsilon_p \boldsymbol{E}) = 0$, where $\varepsilon_p = 1-\omega_p^2/\omega^2$ is the plasma dielectric constant and $\omega_p = \sqrt{4\pi n_e e^2/m}$ is the plasma frequency, Eq. (2) can be written as[4]

$$\vec{P}(2\omega) = \chi\left[\frac{1}{2}\nabla E^2 + \frac{2(\vec{E}\cdot\nabla \ln n_e)\vec{E}}{\varepsilon_p}\right]. \tag{3}$$

In the nonuniform plasma, the second term on the right-hand side dominates the SHG,[4] i.e., the SH beam will be copolarized with the fundamental one. Therefore, since in the cylindrically symmetric fundamental beam ∇n_e is perpendicular to the propagation direction, the LP fundamental beam produces SH radiation in the form of two lobes. The lobes are oriented along the polarization azimuth and with electric field phase shifted by π with respect to one another. The CP fundamental beam produces the SH radiation, which is co-circular polarized and forms a ring around the beam axis.

The later case is particularly interesting and deserves further insight because of the restrictions imposed by the momentum conservation law. Specifically, in the SHG process, a pair of the photons of the fundamental beam with angular momentum $2\hbar$ creates only one SH photon with angular momentum $\hbar$ (Fig. 1). Thus the remaining angular moment should be either transferred to the plasma in the focal area or to the orbital angular momentum of the SH beam creating an optical vortex.[9,10] However, the ring shaped pattern (typical for Laguerre–Gaussian TEM_{01^*} mode) observed in the far-field strengthens assumption of phase singularity presence and an optical vortex generation.

The CP of the SH was confirmed by placing a linear polarizer in the SH beam. This led to the signal decrease by a half and was independent of the polarizer orientation. This observation coincides with the results on SH generation in helium gas.[8]

Assuming interaction length of about 2 μm, we estimate the effective second order nonlinearity in our experimental conditions to be 0.03 pm/V, which is of the same order as in poled silica fibers.[11,12] Increasing the interaction length to 1 mm, e.g., using specially designed capillary or photonic crystal fiber,[13] can result in achieving practical conversion efficiencies in access of 50%, which particularly interesting for deep UV generation where the use of nonlinear crystals such as beta barium borate (b-BaB_2O_4 or BBO) is restricted.

In conclusion, we demonstrated high average power ($\sim$10 μW) SHG in ambient air with subpicosecond pulses at high repetition rate using tight focusing conditions. The efficient generation of circularly polarized SH vortex in air and the technique of efficiency increase via gas blow could find applications in optical trapping and manipulation.

P.G.K. dedicates this paper to the memory of V. Serikov (Ref. 14). The work was supported by the Physical Sciences Research Council (EPSRC) and Academy of Finland (Grant No. 131165).

[1] Y. Y. Lau, F. He, D. P. Umstadter, and R. Kowalczyk, Phys. Plasmas **10**, 2155 (2003).
[2] C. D'Amico, A. Houard, M. Franco, B. Prade, and A. Mysyrowicz, Phys. Rev. Lett. **98**, 235002 (2007).
[3] R. Ackermann, K. Stelmaszczyk, P. Rohwetter, G. Méjean, E. Salmon, and J. Kasparian, Appl. Phys. Lett. **85**, 5781 (2004).
[4] D. S. Bethune, Phys. Rev. A **23**, 3139 (1981).
[5] T. Mossberg, A. Flusberg, and S. R. Hartmann, Opt. Commun. **25**, 121 (1978).
[6] Y.-D. Qin, H. Yang, C.-J. Zhu, and Q. Gong, Appl. Phys. B: Lasers Opt. **71**, 581 (2000).
[7] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
[8] M. Mori, E. Takahashi, and K. Kondo, Phys. Plasmas **9**, 2812 (2002).
[9] K. Dholakia, N. B. Simpson, M. J. Padgett, and L. Allen, Phys. Rev. A **54**, R3742 (1996).
[10] A. Bahabad and A. Arie, Opt. Express **15**, 17619 (2007).
[11] W. Margulis, F. Laurell, and B. Leschel, Nature (London) **378**, 699 (1995).
[12] P. G. Kazansky and V. Pruneri, Phys. Rev. Lett. **78**, 2956 (1997).
[13] R. F. Cregan, B. J. Mangan, J. C. Knight, T. A. Birks, P. St. J. Russell, P. J. Roberts, and D. C. Allan, Science **285**, 1537 (1999).
[14] V. V. Serikov and K. Nanbu, J. Appl. Phys. **82**, 5948 (1997).