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Phase-matched third-harmonic generation by nonlinear phase shift in a hollow fiber

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Abstract. Third-harmonic generation in a hollow glass fiber has been demonstrated. Ti:sapphire laser pulses 100 fs with a moderate intensity of around $1 \times 10^{14}\,\mathrm{W/cm^2}$ are introduced into a 1.8-cm hollow fiber in room air. From the observation of the third-harmonic beam quality and spectra, it is clarified that the harmonics are pumped at the leading edge of the fundamental pulse. The model calculation accounting for the nonlinear refractive index change of air shows a quantitative agreement with the experimental results.

High-order harmonic generation (HHG) in gases has been studied intensively during the last decade [1–3]. These studies have demonstrated that HHG is a promising method of achieving a tabletop, high-brightness, coherent, extreme-ultraviolet radiation source. In order to improve HHG efficiency, reduction of the phase mismatch between the pump waves and harmonic waves is crucial. Unfortunately, studies so far have not attempted to address the phase matching issue in a plasma- and neatral-gas mixture, since the investigation of the phase-matching condition is difficult due to complicated phenomena such as self-focusing and plasma defocusing [4] in the interaction region with the tight focusing geometry employed.

Recently, several techniques to control the propagation conditions of intense ultrafast laser pulses were investigated. These include the formation of plasma columns in gases based on bulk plasma motions [5], relativistic electron oscillation [6], balance between self-focusing and diffraction of a laser beam [7, 8], and reflection from a plasma lining wall [9]. With a glass-coated hollow-core fiber, Nisoli et al. [10] demonstrated a long channeling of intense, fs–laser pulses at an intensity of $10^{13}\,\mathrm{W/cm^2}$. Here we propose the use of the hollow fiber in the HHG experiment. With this new technique, due to the flat phase front of the driving (pumping) laser, one can avoid undesirable variations of phase-matching factors which originate from phase shifting around the focus. This will facilitate the clear and easy finding of phase-matching conditions. Furthermore, under suitable conditions, very long interaction lengths can be obtained without defocusing.

Since various measurements can easily be made using conventional UV optics and the precise measurement of spectra and intensity distribution can be made in the UV region, we concentrated our efforts on the investigation of the third-harmonic generation (THG). The availability of experimentally reported data of third-order nonlinear susceptibility $\chi^{(3)}_{3\omega}$ and refractive indices [11] can also make it possible to carry out the model calculation. Previously reported THG studies have dealt with the conversion efficiency, spectral characteristics, and spatial characteristics, in order to obtain a clue to the efficient HHG. Siders et al. [12] used loosely focused 80-fs, 800-nm pulses in subatmospheric-pressure noble gases with pump intensities $\approx 10^{16}\,\mathrm{W/cm^2}$, to obtain third-harmonics. They observed a strong blueshifting only at the beam core which was well collimated by the self-channeling of the pump pulse. Backus et al. [13] reported that whole beam self-channeling of 22 fs-Ti:sapphire laser pulses in room air demonstrated a highly efficient THG ($\approx 0.1\%$). They mentioned that the excitation pulse should be negatively chirped and the strongest third-harmonic output was obtained with loosely focusing optics $f/30$. Fedotov et al. [14] studied experimentally the effect of temporal and spatial self-action of light on the THG in room air, and speculated that the self-focusing of light at the interaction region was responsible for the saturated conversion. In spite of these studies, the underlying mechanism of THG, especially for the conversion efficiency, is not yet fully understood because of the complexity in the interaction region.

In this paper we used a hollow fiber to investigate the THG of 100-fs pulses of a Ti:sapphire laser in atmospheric room air. Since the phase front of both guided and generated beams are in a plane in the hollow fiber, in other words it is not possible for a curved phase front to couple with the hollow fiber, the analysis of the experimental result becomes straightforward. We made the spatial and spectral characterization of third-harmonic light. Our observations clarified the view that harmonics are generated at the leading edge of the pumping pulse. A model calculation assuming that the interaction occurs at ≈ 10 fs before the pump pulse peak shows good agreement with the experimental result.

1 Experimental

The laser pulses used in this study were generated by a mode-locked Ti:sapphire oscillator pumped by an Ar-ion laser. After the 80-fs pulses were temporally stretched to 250 ps and regeneratively amplified to 5 mJ at a 10-Hz repetition rate, they were recompressed to 100 fs assuming sech^2 shape. The recompressed pulses were focused to a diameter of 75 µm by a $f = 50$ cm achromatic lens in room air. The hollow fiber front was placed at the focal point. Angular and transverse positioning of the hollow waveguide were performed by monitoring the amount and mode pattern of the transmitted light [9].

The linearly polarized, 100-fs, 780-nm laser pulses at intensities up to $2.2 \times 10^{14}\,\text{W/cm}^2$ were coupled into a 100-µm-diameter glass-coated hollow fiber in room air. Measurements of THG beam quality and conversion efficiency were performed as follows. The output light from the hollow fiber was recollimated by a 30-cm-focal-length fused-silica lens. After recollimation by the lens, fundamental and third-harmonic beams were separated according to wavelength by passing through a LiF prism and then could be spatially discriminated. The third-harmonic light was further selected by two additional mirrors, which reflected $> 99\%$ of the 264-nm light but transmitted $\approx 99\%$ of the fundamental light. We verified, using a color filter, that this optical arrangement completely eliminated the fundamental light. The joule meter has a detection limit of 0.01 µJ. The setup is shown in Fig. 1.

The hollow fiber used in this study was a fused-silica capillary supplied by GL Science Inc. and was not fabricated for optical guiding. They have a 100-µm bore diameter surrounded by 375-µm-diameter fused-silica which is polymer-coated on the outer surface in order to keep it straight. The fiber length was 1.8 cm, i.e. ten times as long as the measured confocal parameter and short enough for neglecting propagation losses of the pump laser. When a Gaussian beam is coupled into a cylindrical hollow fiber at its beam waist, only the EH modes in the waveguide are excited [10]. The propagation of electromagnetic waves under the cylindrical boundary conditions has been studied in detail [15]. The maximum coupling efficiency for the EH_{11} mode is calculated to be 98.1%, which is obtained at $w_0/a = 0.64$, where w_0 is a waist of the diameter of the input beam and a is the bore diameter of the hollow waveguide. The coupling efficiency was measured at 75% for an input energy of less than 0.1 mJ. Coupling losses were gradually increased with the increase of input laser energy. The measured efficiency was decreased to 50 % for a 0.5-mJ input, 32% for 1.5 mJ, and 28% for 2.0 mJ. These values were lower than the theoretical efficiency of 90% calculated with the experimental value of $w_0/a = 0.75$. The losses were due to the distortion of the pump-beam pulse front caused by self-focusing in room air prior to coupling.

The fiber did not undergo any damage during the experiment, which was performed for several hours, when the incident beam and hollow fiber were properly aligned on the axis. This is explained by the theoretical prediction that no optical energy can exist in a boundary region [15]. In order to avoid any damage, the filter was removed while carefully observing the output beam pattern after alignment was performed with a very low-intensity laser by inserting a neutral density filter.

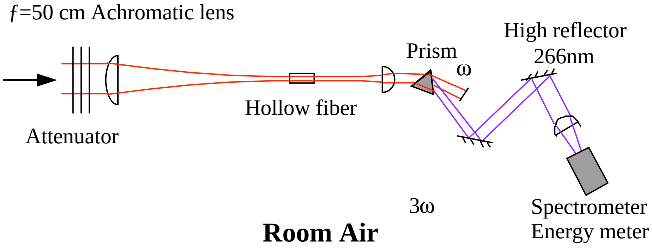

Fig. 1. Schematic of third-harmonic generation experiment in a hollow fiber including detection apparatus

2 Spatial and spectral characteristics of the THG

Measurement of the third-harmonic characteristics was performed with and without the hollow fiber. First, we measured the beam quality of the third-harmonic light through a hollow fiber using a CCD camera, as shown in Fig. 2a. The slightly oval shape of the profile obtained was due to the astigmatism of the prism. Both the fundamental and the third-harmonic beams from the fiber were linearly polarized and were single mode. However the third-harmonic profile in Fig. 2b. generated in free space was strongly disturbed and featured a bright red halo as reported before [14]. By focusing more-intense laser pulses ($5 \times 10^{14}\,\text{W/cm}^2$) in free space, we observed a white-light continuum.

The observed third-harmonic spectra with and without the fiber are shown in Fig. 3. All the spectra generated without the fiber were redshifted as shown by dashed lines in Fig. 3. This redshift is induced by spectral upchirp of the self-phase-modulation of the pump laser pulses. Since the experiments were conducted in air, the self-phase-modulation-induced spectral upchirp could not be avoided. This effect is well known and does not modify the temporal shape of the pump pulse [16].

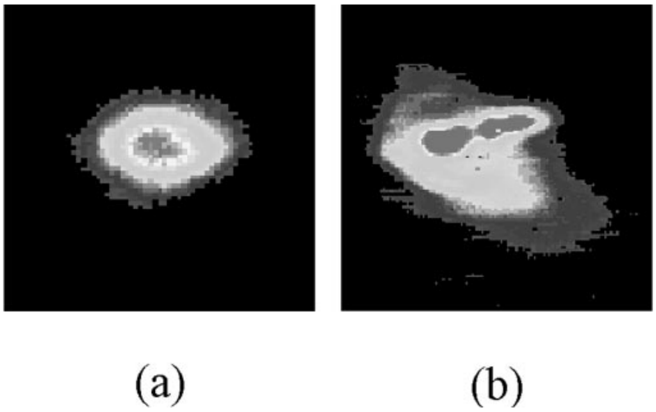

Fig. 2a,b. Spatial patterns of a third-harmonic light beam emerging from the 1.8-cm hollow fiber (**a**) and in free space (**b**). Detection was made with a CCD camera after passing the lightthrough a prism and two high reflectors

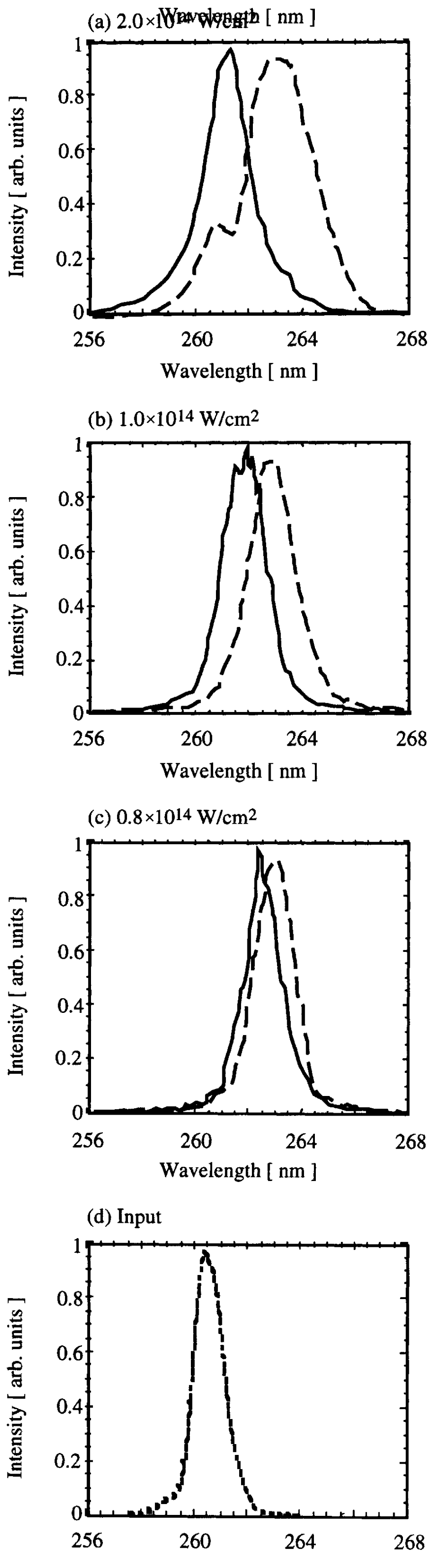

Fig. 3a–d. Spatially integrated third-harmonic spectra for three pump intensities. Spectra of third harmonics generated in free space are shown by *dashed lines*, and after the propagation in the 1.8-cm fiber are shown by *solid lines*. The pump intensities are **a** $2.0 \times 10^{14}\,\mathrm{W/cm^2}$, **b** $1.0 \times 10^{14}\,\mathrm{W/cm^2}$, and **c** $0.8 \times 10^{14}\,\mathrm{W/cm^2}$. Note that the spectra measured in free space were considered to be third-harmonic spectra at the fiber entrance in order to determine the ionization rate. The calculated unshifted third-harmonic-generated spectrum of the fundamental is shown in **d**

Third-harmonic spectra after propagation through the fiber are indicated by solid lines in Fig. 3. We found that third-harmonic spectra are blueshifted compared to those obtained in free space though they are still redshifted compared to the calculated unshifted third-harmonic-generated spectrum of the fundamental. The amount of blueshifting increased with higher pump intensity. The measured shifts were (a) 2.0 nm at a pump intensity of $2.0 \times 10^{14}\,\mathrm{W/cm^2}$, (b) 1.0 nm at $1.0 \times 10^{14}\,\mathrm{W/cm^2}$, and (c) 0.5 nm at $0.8 \times 10^{14}\,\mathrm{W/cm^2}$.

Siders et al. [12] observed two peaks in the modulated third-harmonic spectra, and also reported that blueshifted and unshifted third-harmonic pulses were separated spatially. The spatial and spectral characterization with a prism disclosed that the guided third-harmonic pulse in a plasma channel was strongly blueshifted, whereas the pulse in the spatial wing was unshifted. Third-harmonic spectra observed by Backus et al. [13] are very similar to our result shown in Fig. 3. All the specta of third-harmonics were uniformly blueshifted so they did not have spatial variations. Since only the third-harmonic pulses pumped on axis propagate in the guide, and the central part of the THG pulse beam was strongly amplified, the spatial distribution of the third-harmonic spectrum emerging from the guided region became uniform.

Generally speaking, an intense ultrashort pulse experiences a temporal variation of gradients of electron densities, which makes the shape of a self-modulated spectrum very complicated [17]. However this is not true for THG, since some experimental results reported by some other groups [2, 12–14] and this study indicated no spectral modulation of third-harmonic. This fact indicates that the ionization rate of the plasma during THG is constant [18].

A blueshift is caused by a rapid change of the refractive index because of the rapid ionization of atmospheric gases during an interaction with intense fs-laser pulses. The blueshift is expressed as a function of the rate of ionization and the interaction length [19], so the ionization rate $\mathrm{d}N_\mathrm{e}/\mathrm{d}t$ can be estimated from the observed blueshift $\Delta\lambda_\mathrm{blue}$. Here we neglect the frequency shift that is due to depletion of the neutrals since it is roughly two orders of magnitude smaller than that from the increase of the free electrons [17]. We obtained the interaction electron density N_e, simply by multiplying the rate $\mathrm{d}N_\mathrm{e}/\mathrm{d}t$ by one half of the pulse width $\Delta t = 50$ fs which was used in this experiment.

Next, in order to determine when the THG occurred in the pump pulse, we compared the experimentally estimated electron density with the numerically calculated one. The numerical calculation of the temporal evolution of electron density was performed using the ADK ionization model [19]. The result indicated that the electron densities estimated from the observed amount of blueshift must be a few percent of the calculated densities at the pulse peak. This result indicates that the interaction was made at ≈ 10 fs before the pulse peak. This calculation supports a prevailing interaction model in which harmonics should be pumped at the leading edge of the pump pulse.

3 Conversion efficiency

The measurement of energy conversion efficiency in a 1.8-cm hollow fiber was carried out. We averaged five data of measurements. Each measurement was performed for one second

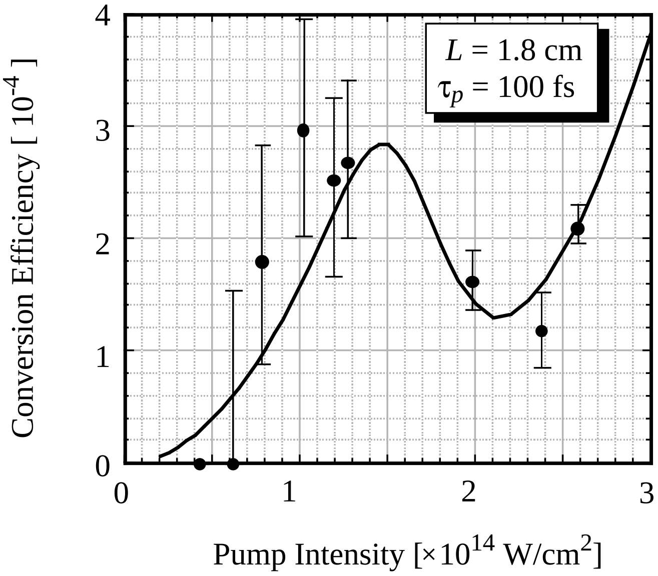

Fig. 4. Experimental plots of energy conversion as a function of pump laser intensity along with model-calculation results considering dispersion of air and plasma, and the nonlinear refractive index change of air

at 10 Hz. The results are plotted in Fig. 4. This measurement revealed that the conversion efficiency reached a peak value of 3×10^{-4} at a pump intensity of $1\times10^{14}\,\mathrm{W/cm^2}$, and then decreased to 1.0×10^{-4} at $2.4\times10^{14}\,\mathrm{W/cm^2}$. After that, it increased again. Although a similar structure of the conversion efficiency as a function the pump intensity was found in other literature [12, 14], this fact has not been explained in detail. To explain the characteristics of the conversion efficiency, we performed a model calculation. The third-harmonic conversion efficiency $\eta_{3\omega}$ for a plane wave is written as [20]

$$\eta_{3\omega} = \frac{1}{3^{3/2}} \frac{\omega_3^2}{n_3 n_1^3 c^4 \varepsilon_0^2} |\chi_{3\omega}^{(3)}|^2 I_1^2 L^2 \,\mathrm{sinc}^2(\Delta k L/2). \qquad (1)$$

SI units are used. $n_i (i = 1, 3)$ are the refractive indices of the fundamental and third-harmonic waves, c is the velocity of light in vacuum, ε_0 is the vacuum permittivity, $\chi_{3\omega}^{(3)}$ is the third-order nonlinear susceptibility, I_i is the pump laser intensity, and L is the hollow fiber length.The amount of phase mismatch is given by

$$\Delta k = 2\pi(n_3/\lambda_3 - 3n_1/\lambda_1). \qquad (2)$$

The refractive index for air $n_{i,\mathrm{air}}$ is calculated using Sellmeier's equation [21], and those for free-electron gases $n_{i,\mathrm{fe}}$ are calculated from the plasma frequency. The total refractive index is expressed by

$$n_i = n_{i,\mathrm{air}} + n_{i,\mathrm{fe}}. \qquad (3)$$

It is unlikely that phase mismatching could be decreased by increasing the pump laser intensity in air or in plasma without resonance, which will be explained below. The phase mismatch caused by neutral air is $+5.4\,\mathrm{cm^{-1}}$, and decreases with increasing electron density N_e because of the ionization by pump laser pulses, whereas the positive phase mismatch results from the free electrons, which are initially zero and increase very rapidly with the increase in N_e. Therefore, the total positive phase mismatch is expected to increase rapidly from a minimum value of $5.4\,\mathrm{cm^{-1}}$ to $100\,\mathrm{cm^{-1}}$ when I_1 exceeds $1\times10^{14}\,\mathrm{W/cm^2}$. This contradicts the experimental result. So we introduce the nonlinear refractive index which plays an important role in the nonlinear phenomena caused by an intense laser field such as self-phase modulation. The nonlinear index, $n_{i,\mathrm{air}}$ in (3) is given by

$$n_{i,\mathrm{air}} = n_{i,0} + n_{i,2} I_i, \qquad (4)$$

where n_0 is the linear refractive index and $n_{i,2}$ is the nonlinear refractive index $5\times10^{-15}\,\mathrm{m^2/W}$ [7]. The nonlinear refractive index change is effective when the pump laser intensity is higher than $1\times10^{14}\,\mathrm{W/cm^2}$, because the nonlinear term exceeds the linear one. The linear refractive indices $n_{i,0}$ and $n_{i,\mathrm{fe}}$, the nonlinear refractive index $n_{i,2}$, and the third-order nonlinear susceptibility $\chi_{3\omega}^{(3)}$ are all linear functions of the electron density. As mentioned previously, the electron density is a function of pump laser intensity and all parameters are expressed as a function of the pump intensity. The amount of phase mismatch in (2) is calculated as a function of the pump intensity and is shown in Fig. 5. To clarify the effect of the nonlinear index change, the calculation without n_2 is also shown by a dashed curve.

We have calculated the conversion efficiency as a function of the pump intensity using (1) which was rewritten in $(5\times10^{-38}) I_1^2 \,\mathrm{sinc}^2(\Delta k/2)$ in SI units from [11]. For a numerical calculation, it is assumed that harmonics are pumped at the front of the pump pulse where the interaction intensity is 90% of the peak intensities. As can been seen in Fig. 4, the model calculation showed a very good quantitative agreement. Phase matching, where a peak efficiency is given, is achieved at $1\times10^{14}\,\mathrm{W/cm^2}$, and then the conversion efficiency is decreased because of phase mismatching induced by the rapid ionization. At $1\times10^{14}\,\mathrm{W/cm^2}$, the conversion efficiency once again increases because the ionization begins to saturate and the nonlinear refractive index compensates for

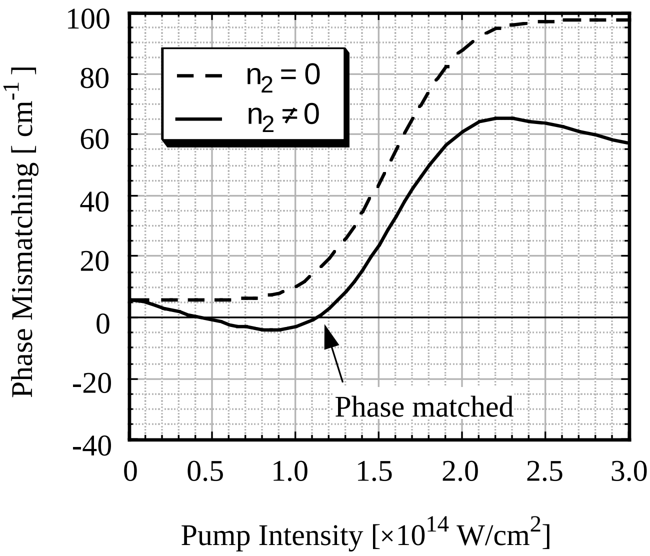

Fig. 5. Numerical calculation of phase mismatching as a function of pump laser intensity.The dashed curve is obtained by neglecting the nonlinear refractive index n_2, and the solid curve is obtained by taking account of the nonlinear refractive index change

the mismatch, as shown in Fig. 5. Since the conversion efficiency is sensitive to the amount of phase matching, and the phase matching is sensitive to the interaction electron density and the pump laser intensity, the slight variation of the pump intensity caused large fluctuations in the outputs in the experiment. This makes the error bars at $\leq 1 \times 10^{14}\ \mathrm{W/cm^2}$ much larger than those at a higher intensity in Fig. 4.

4 Conclusion

We have demonstrated third-harmonic generation in a hollow fiber in room air. The beam quality of the third harmonic was dramatically improved with a hollow fiber. Spectral characterization showed that all the spectra of third-harmonics were uniformly blueshifted. We conducted a model calculation of the energy conversion efficiency by introducing an intensity-dependent refractive index and electron density. The results of the calculation indicated that phase matching was achieved with a pump intensity of $1 \times 10^{14}\ \mathrm{W/cm^2}$, and showed quantitative agreement with the measurement. The results shown here encourage further investigation of higher-order-harmonic generation in the hollow fiber, which may lead to the understanding of the role of macroscopic phase matching in higher-order-harmonic generation. Further experiments, including the use of a gas-filled hollow fiber isolated in vacuum in order to avoid both the coupling loss due to self-focusing before the fiber and absorption after the interaction in the fiber, are under investigation.

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