Highly Efficient, Phase-Matched High-Harmonic Generation by a Self-Guided Laser Beam

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We demonstrate a phase-matched high-harmonic generation of 100 fs Ti:sapphire laser pulses by a self-guided beam in 30 Torr of neon gas. Phase matching of high harmonics is experimentally examined by changing the propagation length up to 7 mm. Phase-matched propagation over 7 mm magnifies the conversion efficiency around the 49th harmonic (in the cutoff region) by 40 times. On the other hand, harmonics around the 25th order (in the plateau region) peak at 4 mm, resulting in an enhancement of 4. This scheme enhances the conversion efficiencies in the cutoff region up to 10^{-6}, corresponding to a few nJ per high-harmonic pulse in the 76 eV region. [S0031-9007(99)08446-X]

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A high-harmonic generation (HHG) in an ionizing medium [1–4] has distinguishing characteristics among the methods for the generation of coherent extremely ultraviolet (XUV). However, the obtained energy conversion efficiencies from the driving laser to harmonics below 20 nm have so far been limited to 10^{-8}. Macroscopic phase-matching issues are the key to the breakthrough.

Three groups have reported the phase-matching technique of high harmonics using hollow fibers [5–7]. We reported that the phase matching of HHG in a hollow fiber was assisted by the nonlinear phase shift of the driving laser, which canceled out the plasma dispersion [5]. Durfee *et al.* [6] reported that phase matching was achieved by the effective refractive index of the waveguide. Schnürer *et al.* [7] mentioned that the effect of the plane phase front might lead an improved conversion efficiency. All of the obtained results indicated that the interaction length was increased to more than 1 cm, and the HHG efficiencies increased by more than 1 order of magnitude. The original idea for the use of the fiber comes from the need to control the propagation conditions of high-intensity lasers, such as the interaction length, interaction gas pressure, and interaction laser intensity. The most important point is the determination of the coherent length (optimum interaction length).

The limitation of a self-channeling [8] technique is its interaction length. When the laser intensity exceeds 10^{14} W/cm^2, which is required to produce high harmonics, self-focusing is no longer in balance with plasma defocusing, resulting in the elongation of the confocal parameter to 10 times or less [9] to that in the vacuum. The harmonic generation of intense femtosecond pulses by a self-guided beam has so far been reported only for the third (lowest-order) harmonic generation [9,10]. The results indicated that spatially confined intense third harmonics were obtained with high conversion efficiency.

In this Letter, we demonstrate the HHG of Ti:sapphire laser pulses by a self-guided beam. Laser pulses of 5 mJ were introduced into a gas cell filled with neon at a pressure of <50 Torr. As a result of phase-matched propagation, harmonics in the cutoff region were magnified by 2 orders. High-harmonic conversion efficiencies of 10^{-6} were demonstrated by the phase-matched interaction, producing harmonics with an energy of $>$nJ in the cutoff region around the 49th harmonic. The phase matching was discussed in terms of intrinsic phases of a high-harmonic generation which originates in electron trajectories [11].

The laser pulses used in this study were generated by a chirped-pulse amplification-based Ti:sapphire laser producing a 100 fs temporal length with 5 mJ energy at a 10 Hz repetition rate. The measured beam waist was 40 μm (in vacuum), and the measured confocal parameter was 3.5 mm with a $f = 50$ cm achromatic lens. Therefore, the focusing intensity in the vacuum was 1.5×10^{15} W/cm^2, but the interaction intensity in the channel should be lowered to a few 10^{14} W/cm^2 as a result of a resistance of focusing by a plasma-induced negative lens effect. A schematic of the experimental setup is shown in Fig. 1. The gas cell had two 300-μm-diameter pinholes on each end surface of the bellow arms. The pinholes isolated vacuum and gas-filled regions. The outside of the gas-filled region was maintained at a pressure below 10^{-4} Torr, as the cell was pressurized with neon atomic gas at 50 Torr. The spectrograph employed consisted of a flat-field platinum-coated grating, a microchannel plate, a photocathode, and charge-coupled devices. This optical arrangement can cover the region from the 23rd to 51st harmonic of 800 nm. The spectrograph unavoidably picked up the second-order diffraction of higher-order harmonics. The obtained spectra were corrected for the sensitivity of the grating and the microchannel plate.

Laser propagation was transversely seen by observing a visible light column (plasma channel) of an orange color. The plasma channel length was approximately 8 mm (2.5× the confocal parameter), which was confirmed by changing the focal point in the gas cell and observing

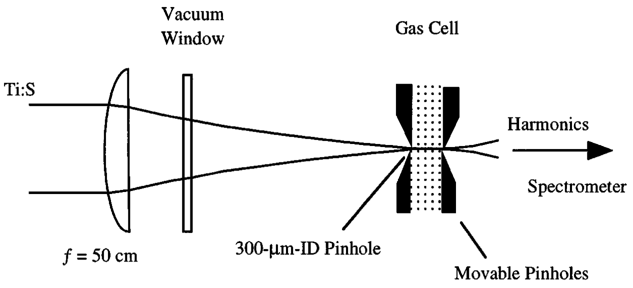

FIG. 1. Schematic top view of the differential-pumped gas cell. The focusing lens ($f = 50\,\mathrm{cm}$) is located on the outside of the vacuum window.

the end position. The transmitted fundamental beam pattern shrank to half that in the vacuum at 10 Torr, indicating a decrease of the beam divergence. This is the well-known phenomenon of laser beam self-guiding [8,9]. The degradation of the driving laser spatial mode (1.6× diffraction limited) resulted in no evidence of self-guiding. In our experiment with pressures of 10 to 55 Torr, the transmitted fundamental beam profile, as well as the energy throughput, indicated no significant changes.

Harmonics were not generated when the gas cell was evacuated to 10^{-3} Torr. In Fig. 2, we show the harmonic yield as a function of gas pressure. The distance between two pinholes, which determines propagation length, was set at 1 mm, which is the typical length in HHG experiments. The focal point was adjusted to within 1 mm in front of the entrance pinhole of the static gas cell where the maximum XUV emission was obtained. Pressurizing the gas cell from 10 to 40 Torr, the emitted high harmonic spectra showed a plateau nature, and the entire plateau region grew and then saturated at 30 Torr. As the harmonics increased, the noisy background relatively decreased so that the signal-to-noise ratio was clearly improved. For a gas pressure higher than 40 Torr, the emission decreased by 1 order of magnitude and no notable change was found in the distribution.

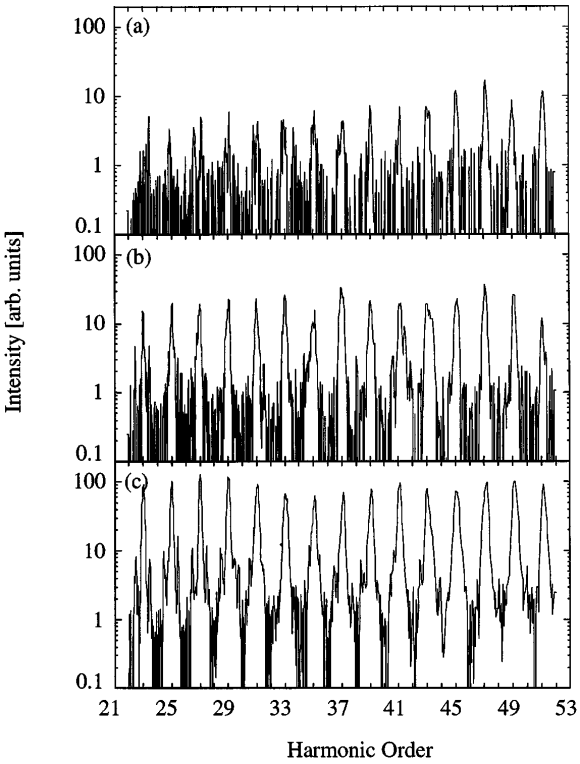

FIG. 2. Experimentally observed high-harmonic intensity as a function of photon energy in units of fundamental frequency. 100 fs, 5 mJ Ti:sapphire laser pulses at 800 nm interacted with neon in a 1-mm-long static gas cell under gas pressures of (a) 10 Torr, (b) 20 Torr, and (c) 40 Torr.

Figure 3 shows the dependence of a harmonic intensity on the propagation length at 30 Torr. Vertical axes are of the same scale for all the data shown in Figs. 2 and 3.

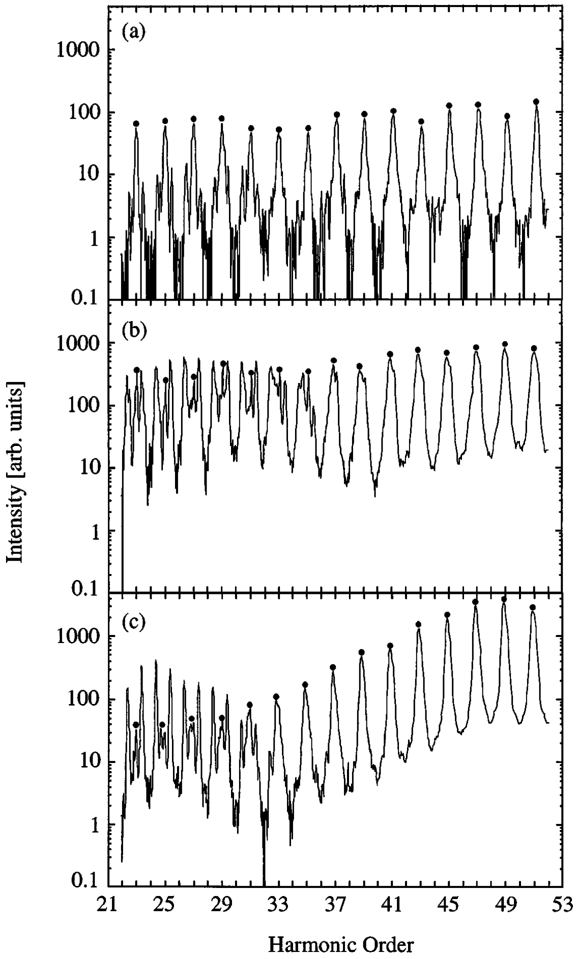

FIG. 3. Experimentally observed high-harmonic emission from a self-guided beam as a function of photon energy in units of fundamental frequency. A driving laser formed an 8-mm self-guiding beam in 30 Torr neon gas, and the spectra were obtained at interaction lengths of (a) 1 mm, (b) 4 mm, and (c) 7 mm. Filled dots indicate the first-order diffracted harmonics.

When the guiding length was elongated to 4 mm and more, the cutoff order was equal to 71. Using the relation $h\nu_c = I_p + 3.17U_p$ [12], the interaction intensity was estimated to be 1.5×10^{14} W/cm^2. This estimation for a single-atom response is a somewhat lower value. All harmonics from the 23rd to the 51st were equally magnified. It can also be observed in the figure that the spectral shapes of higher-order harmonics are broader than those of lower-order harmonics. It must be noted that the second-order diffraction disturbed the observations in the plateau region. Significant changes in the harmonic distribution gradually emerged when the interaction length was increased beyond 4 mm. Only harmonics near the cutoff order further increased with the interaction length, but the harmonics in the plateau region decreased. As the length was increased to 7 mm, the cutoff harmonics were enhanced to 4 times that at 4 mm (40 times of that at 1 mm), and the plateau harmonics were decreased to $\frac{1}{10}$ times that at 4 mm ($\frac{1}{2}$ times of that at 1 mm). For 7 mm, we measured an emitted photon number of 10^8 per harmonic pulse at the 49th harmonic (76 eV), corresponding >nJ energy per harmonic, indicating a conversion efficiency of 10^{-6}. Upon further increasing the variable gas cell length up to where the emission of the plasma channel disappears, the harmonic signals dropped to below the detection level. This disappearance may be caused by a strong inhomogeneity of the plasma distribution near the output of the plasma channel.

In order to understand the macroscopic phase-matching mechanism in HHG, we examine the spectral characteristics of high harmonics. Figure 4 shows two harmonics, the 25th harmonic representing the harmonics in the plateau region and the 49th harmonic representing those in the cutoff region. Both are normalized to clarify the spectral shape. Note that the second-order diffraction which appears in (a) must be neglected. The modulation of the spectral shape of the 25th harmonic, which is slightly seen even at 1 mm, was enhanced at longer interaction lengths. The single spectral peak broke into several apparent peaks. On the other hand, the 49th-harmonic spectrum was smooth with a good signal-to-noise ratio at all interaction lengths.

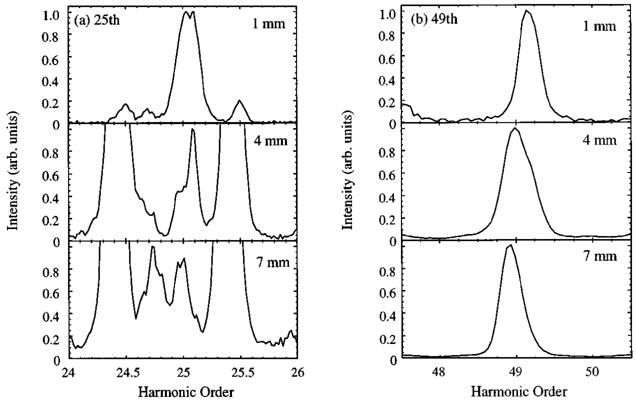

FIG. 4. The spectral characteristics of the (a) 25th (in the plateau region) and (b) 49th (in the cutoff region) harmonics for various interaction lengths. The second-order diffraction of the 49th and 51st harmonics appears at the 24.5th and 25.5th harmonics in (a).

With increasing propagation length, the peak frequency of the 49th harmonic was “redshifted,” yielding stronger emission. This might be caused by self-phase modulations (SPM) in the leading edge of the driving pulse, where the harmonics were pumped [2]. The result obtained here contradicts our former result [5] that the high harmonics were blueshifted in an argon gas. We assume that the reason is not due to the difference in the guiding scheme, but to the use of different gas species. Since neon atoms are harder to ionize, the ionization rate is much smaller, so that the SPM-induced up chirp should be stronger than the blueshift. A propagation loss would be another reason for the redshift. Since an ionization rate decreases with the propagation length, the harmonics generated at 7 mm showed a larger redshift. The remaining questions are why the spectral modulation was produced only for harmonics in the plateau and why the cutoff harmonics are enhanced markedly.

Recent advances of theoretical calculations based on the macroscopic response of the collections of atoms [11,13] have indicated the phase-matching process strongly depends on the driving laser intensity, because the trajectories of electrons which generate harmonic light are influenced by the pump laser field and the trajectories determine the initial harmonic phases. The variation of the intrinsic phase as a function of the wavelength changes its spectral structure. When the intrinsic phase exhibits a rapid variation, the harmonic spectrum results in a noisy structure. When the intrinsic phase shows a slow variation, the harmonic spectral structure results in good contrast and is smooth.

Although pumping laser energy exists even in an unguided region, the coupling between pumping and generating fields is limited to the axial direction [5]. Therefore, in this interaction scheme, phase variation on the optical axis [11] could be minimized. When the intrinsic phase is constant over the entire propagation length, the interactions are “phase matched.”

Since the intrinsic phase is much more stable for the harmonics in the cutoff region than in the plateau region [14], the phase-matched propagation is realized preferentially for the cutoff harmonics. The phase error of the plateau harmonics escalates with propagation, leading to the strong spectral modulation of the plateau harmonics. This is the main reason that the effective coherent lengths of the cutoff harmonics were larger than those of the plateau harmonics.

In conclusion, we demonstrated a highly efficient high-order harmonic generation by phase-matched propagation in a self-guided laser beam. Harmonics near the cutoff order were enhanced by 2 orders of magnitude compared to those in the plateau as a result of the phase-matched propagation for 7 mm. On the other hand, those in the plateau began to dephase at 4 mm resulting in poor conversion. The results were well explained by considering intrinsic phase-matching effects. The quantitative interpretation deserves further consideration. High-harmonic conversion efficiency of 10^{-6} was demonstrated by a phase-matched generation, producing $>$nJ harmonics in the cutoff region around the 49th harmonic.

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