Enhancement of Relativistic Harmonic Generation by an Optically Preformed Periodic Plasma Waveguide

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Enhancement of relativistic third-harmonic generation by using an optically preformed periodic plasma waveguide was achieved. Resonant dependence of harmonic intensity on plasma density and density modulation parameters was observed, which is a distinct characteristic of quasi-phase-matching. The results demonstrate the potential of a modulated plasma waveguide in high-field applications.

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High-order harmonic generation based on optical-field ionization (OFI) of electrons in atoms followed by recombination [1] and relativistic harmonic generation based on nonlinear quiver motion of free electrons in relativistically intense laser field [2–5] are active fields of research, as they are the most promising approaches to date for generating coherent ultrashort x-ray pulses. Yet, for a wide range of applications how to produce harmonics with short wavelength and high efficiency is still a challenge. In order to achieve sustained growth of harmonics, one needs to increase the interaction length and at the same time suppress the reverse conversion process. This can be done by guiding the laser pulse and phase matching the frequency conversion process. Rundquist *et al.* [6] has reported the use of hollow fiber to achieve guiding and phase matching of high harmonic generation under the condition of low ionization fraction. It was later demonstrated that further enhancement of high harmonic generation can be achieved by using a corrugated hollow capillary tube to attain quasi-phase-matching under the condition of high ionization fraction [7]. High-order harmonic generation from ions in a capillary-discharge plasma was also achieved [8].

Recent progress in high-power ultrashort-pulse laser technology has made available pump sources with higher and higher intensity. As a result, relativistic harmonic generation is emerging as a promising scheme for generating strong coherent x rays. However, hollow fiber and corrugated hollow capillary are not suitable for relativistic harmonic generation because optical damage occurs under relativistic intensity. A few schemes for producing a plasma waveguide have been reported [9–12], which have the capability of guiding the pump laser pulse and the harmonics simultaneously, but none of them has been applied to relativistic harmonic generation yet. It has also been proposed that a plasma density modulation may be used to achieve quasi-phase-matching of relativistic third-harmonic generation [13,14]. In this scheme periodically interlacing layers of high- and low-density plasmas in the direction of beam propagation with their widths matching the coherence lengths allow periodic correction of the relative phase between the harmonic wave and the fundamental. With a gain in the high-density layer larger than the inverse gain in the low-density layer, the harmonics can grow with increasing interaction length. To date, no experimental demonstration of quasi-phase-matched relativistic harmonic generation has been reported yet.

In this Letter, we experimentally demonstrate the enhancement of relativistic third-harmonic generation (THG) in both a uniform plasma waveguide and a periodic plasma waveguide. For the case of uniform waveguide, enhancement of harmonic generation by optical guiding was verified. For the case of periodic waveguide, resonant dependence of harmonic intensity on plasma density and density modulation parameters was observed, which is a distinct characteristic of quasi-phase-matching. Under the resonant condition, the energy of the third harmonic was enhanced by a factor of 8, and the intensity was enhanced by a factor of 50.

A 10-TW, 45-fs, 810-nm, and 10-Hz Ti:sapphire laser system based on chirped-pulse amplification was used in this experiment (upgraded from the laser system in Ref. [15]). The setup is shown in Fig. 1. Two linearly polarized laser beams from this system were used. One beam served as the longitudinal pump pulse, and the other, set to be 10 ns earlier than the pump pulse, was used as the machining beam for producing the uniform plasma waveguide and periodic plasma waveguide. The 200-mJ, 45-fs pump pulse was focused onto a gas jet by an $f/7$ off-axis parabolic mirror to a spot of 8-μm diameter in full width at half maximum (FWHM) with 80% energy enclosed in a Gaussian-fit profile. The peak intensity was 5×10^{18} W/cm^2. The spatial intensity profile of the machining beam was programmed with an amplitude modulator array composed of a liquid-crystal spatial light modulator

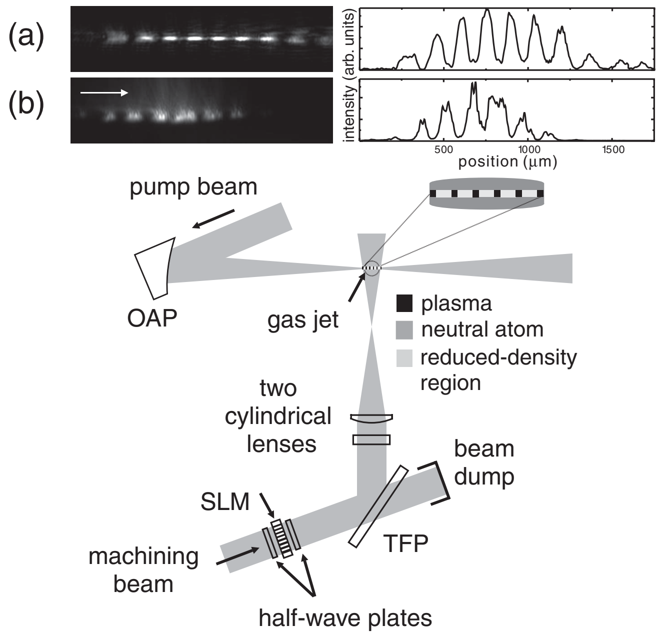

FIG. 1. Experimental setup of the pump beam and the machining beam. Inset (a) shows the intensity pattern and line out profile of the machining beam on the interaction plane. The SLM is set for a periodic pattern of 37.5-μm duty-off regions and 112.5-μm duty-on regions. Inset (b) shows the corresponding side image and line profile of Thomson scattering for a hydrogen atom density of 3.5×10^{19} cm^{-3} and a delay of 10 ns between the machining pulse and the pump pulse. Off-axis parabolic mirror (OAP). Thin-film polarizer (TFP).

(SLM), two half-wave plates, and a thin-film polarizer. Propagating perpendicular to the pump pulse, the 10-mJ, 45-fs machining pulse was imaged horizontally from the SLM onto the interaction plane by a cylindrical lens of 20-cm focal length with a demagnification factor of 8. Also, it was focused vertically to a width of 20 μm in FWHM by a cylindrical lens of 30-cm focal length. The line-shaped transverse machining beam overlapped with the propagation path of the longitudinal pump beam. The gas jet was produced by a pulsed valve with a supersonic conical nozzle. The density profile has a flattop region of 1 mm in length and a sharp boundary of 250 μm at both edges. The atom density was 3.5×10^{19} cm^{-3} at 500-psi backing pressure for hydrogen gas. The angular distribution and total energy of the third-harmonic pulse were measured with an imaging system consisting of a fused-silica spherical lens, a notch filter for eliminating the pump pulse, a bandpass filter centered at 266 nm with a bandwidth of 35 nm, and a 16-bit uv-enhanced charge-coupled device (CCD) camera. The spectrum was measured with an optical spectrometer. The SLM consisted of a horizontal array of 320 strips of 10-μm-thick nematic liquid-crystal layer, each of which was 97 μm in width and 13 mm in height with a spacing of 3 μm between strips. By setting the voltages of the SLM strips to make the polarization axis of the transmitted light perpendicular or parallel to the plane of the thin-film polarizer, the portion of the machining beam from each strip was turned on or off to determine the distribution of laser intensity on the interaction plane and thus the pattern of the fabricated structure.

The principle of plasma density-structure fabrication by laser machining was described in Ref. [16]. When a spatially varying laser intensity pattern is projected transversely onto a neutral gas jet, plasma is formed and heated at the high-intensity regions where the laser intensity exceeds the threshold of optical-field ionization. After several nanoseconds the plasma density in the ionized regions is reduced as a result of hydrodynamic expansion. When an intense pump beam is incident longitudinally, because the front foot of the laser pulse preionizes the gas, the main peak of the pump pulse encounters a plasma with longitudinal plasma density variation. Since the intensity of Thomson scattering is proportional to the product of laser intensity and electron density, side imaging of Thomson scattering from electrons was used to provide a crude measurement of the plasma density distribution along the propagation path of the pump beam. Figure 1(a) shows a periodic intensity pattern with the SLM set for 37.5-μm duty-off regions and 112.5-μm duty-on regions. Figure 1(b) shows the corresponding side image of Thomson scattering. The lengths of duty-off and duty-on regions refer to the settings in the SLM that determine the lengths of unmachined and machined regions, respectively. Although in the SLM the lengths of the duty-off regions and the duty-on regions are set to be 37.5 and 112.5 μm respectively, the actual widths of the high-density and low-density regions on the interaction plane produced by the machining beam are about 60 and 90 μm in FWHM. Such distortion is mainly due to diffraction caused by the SLM and the aberration of the focusing optics. The duration of the pump pulse was stretched to 400 fs when it was used for measuring the fabricated plasma density structure by Thomson scattering.

Inset (a) and (d) in Fig. 2 shows the angular profiles of THG at 4.6×10^{18}-cm^{-3} and 3.5×10^{19}-cm^{-3} hydrogen atom densities, respectively, without the machining beam. In inset (d) the strong reduction of THG divergence in both vertical and horizontal directions at high plasma densities is a result of relativistic self-guiding, which occurs when the plasma density exceeds a threshold such that the laser power exceeds the critical power for self-guiding [17]. Inset (b) shows the THG angular profile under the same condition as in (a) with the machining beam turned on and the SLM set for 100% duty ratio (the entire line-focus region of the machining beam is irradiated), and inset (c) shows the THG angular profile with the SLM set for 37.5-μm duty-off regions and 112.5-μm duty-on regions. In inset (b) the asymmetrical reduction of THG divergence indicates that at low density an elliptic waveguide was produced when the machining beam of 100% duty ratio was turned on. This is anticipated from the results of Ref. [10] in which a transverse line focus was used to produce a plasma waveguide with an elliptical cross section. In inset (c), further reduction of THG divergence was

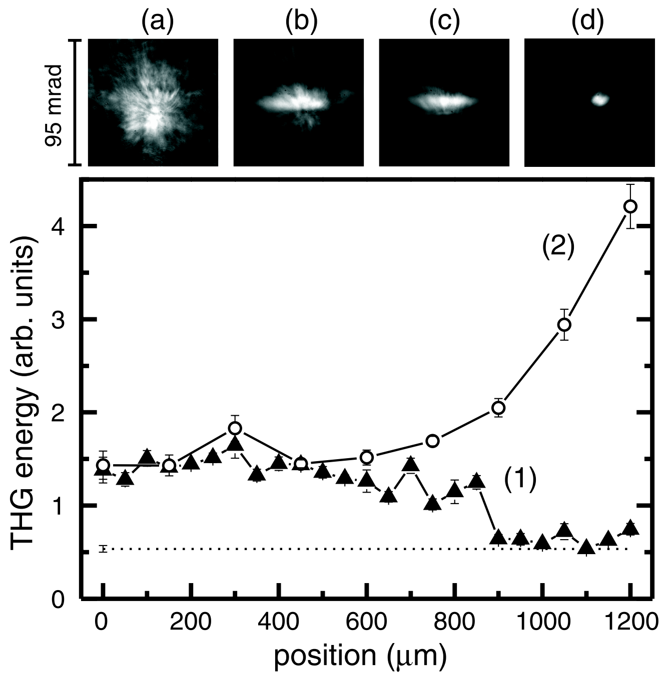

FIG. 2 (color online). Insets (a) and (d) show the angular profiles of THG at 4.6×10^{18}-cm^{-3} and 3.5×10^{19}-cm^{-3} hydrogen atom densities without the machining beam. Inset (b) shows the THG angular profile under the same condition as in (a) with the machining beam turned on and the SLM set for 100% duty ratio, and (c) shows that with the SLM set for 37.5-μm duty-off regions and 112.5-μm duty-on regions. The delay between the machining pulse and the pump pulse is 10 ns. Curve (1) shows the THG energy as a function of the boundary position between the uniform plasma section and the uniform plasma-waveguide section. Larger coordinate means shorter waveguide. Curve (2) shows the THG energy as a function of the boundary position between the periodic plasma-waveguide section and the uniform plasma-waveguide section. Larger coordinate means longer periodic plasma waveguide. The dot line denotes the THG energy for using only the pump pulse. The error bars represent the standard error of mean.

observed when periodic modulation was applied to the elliptical waveguide. As shown in inset (a), at this density relativistic self-guiding is insignificant, hence the periodic high-density regions in the periodic plasma waveguide cannot improve the guiding. The reduction of THG divergence should be the result of improved phase matching in the forward direction.

By programming the SLM, the enhancement of THG as functions of the lengths of both the uniform plasma waveguide and the periodic plasma waveguide was measured, as shown in Fig. 2. As the length of the periodic plasma waveguide is increased to the full length of the gas jet, the THG energy is enhanced by a factor of 8 with respect to that obtained with a uniform plasma (i.e., that without the machining beam), and the on-axis intensity is enhanced by a factor of 50 as the result of simultaneous enhancement of collimation by guiding and quasi-phase-matching. The data in curve (2) clearly show the anticipated function of quasi-phase-matching provided by the periodic density modulation.

Relativistic harmonic generation arises from nonlinear plasma currents driven by nonlinear quiver motion of electrons in an intense laser field [2]. The identification of relativistic third-harmonic generation in this experiment is supported by the following experimental facts. (1) By using a 1/4-wave plate installed in the path of the pump beam and a soft x-ray spectrometer, the THG energy was found to have a very different dependence on the polarization ellipticity of the pump pulse from the case of the 25th–33rd harmonics generated by OFI followed by recombination. Because for the OFI-recombination mechanism the polarization ellipticity dependence is similar for all harmonic orders, it rules out the possibility that the measured THG may come from the OFI-recombination mechanism. (2) The THG energy was not reduced when the machining pulse was used and set at 50 ps before the arrival of the pump pulse to preionize the gas. This rules out the possibility of the bound-electron harmonic generation mechanism. It also rules out again the OFI-recombination mechanism. The conversion efficiency of THG without the machining beam was measured to be 1.1×10^{-8} at 4.6×10^{18}-cm^{-3} plasma density, which is close to the efficiency calculated by using equations in Ref. [13,14].

The resonant dependence of THG on plasma density and periodic density modulation is shown in Fig. 3. Figure 3(a) shows the dependence of the THG energy on the length of the duty-off regions with a fixed 112.5-μm duty-on regions as well as the dependence on the length of the duty-on regions with a fixed 37.5-μm duty-off regions. The hydrogen atom density was 4.6×10^{18} cm^{-3}. Open circles in Fig. 3(b) shows the dependence of the THG energy on hydrogen atom density for 37.5-μm duty-off regions and 112.5-μm duty-on regions. All the dependencies display a sharp resonant behavior, which is a distinct characteristic of quasi-phase-matching. Figure 3(b) also shows the density dependence of the THG energy without [curve (1)] and with [curve (2)] the machining beam of 100% duty ratio. At low densities implementation of the plasma waveguide enhances the THG energy, but not at high densities. This is because at high densities the laser pulse is guided by relativistic self-guiding and the implementation of the plasma waveguide does not provide further enhancement by guiding. Instead, the THG energy is decreased as a result of reduction of the on-axis plasma density. At low densities the plasma waveguide can increase the THG energy by guiding but the effect is offset by the reduced on-axis plasma density. This offset effect does not affect much the periodic plasma waveguide, because under the quasi-phase-matching condition the low-density sections of the periodic plasma waveguide are the places where the reverse conversion occurs. The plasma density in these regions are meant to be reduced to cut down the reverse conversion and thus enhance the THG.

The quasi-phase-matching condition can be estimated by calculating the phase mismatch that takes into account

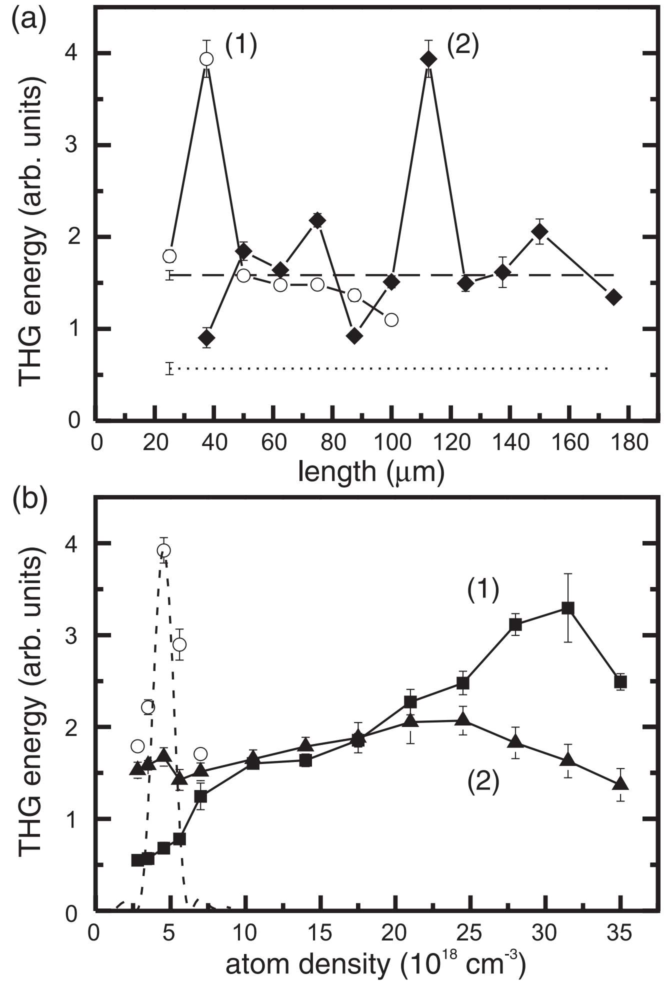

FIG. 3. (a) THG energy as a function of the length of duty-off regions with 112.5-μm duty-on regions [curve (1)] and as a function of the length of duty-on regions with 37.5-μm duty-off regions [curve (2)]. The hydrogen atom density is 4.6×10^{18} cm^{-3}. The dot line and the dashed line indicate the THG energies without the machining beam and with the machining beam of 100% duty ratio, respectively. (b) THG energy as a function of hydrogen atom density for without the machining beam [curve (1)], with the machining beam of 100% duty ratio [curve (2)], and with the machining beam of 37.5-μm duty-off regions and 112.5-μm duty-on regions (open circles). The dash line shows the theoretical prediction based on the assumption described in the context.

the plasma dispersion and waveguide dispersion for both the pump pulse and the third-harmonic pulse. At the optimal condition the plasma density in the high-density regions is 4.6×10^{18} cm^{-3} and the plasma density in the low-density regions is 20% of that in the high-density regions, measured from the interferograms without and with the machining beam of 100% duty ratio, respectively. Assuming a guided-mode radius of 6 μm estimated from the refractive index profile, the calculated phase mismatches are 5.53×10^4 m^{-1} and 3.32×10^4 m^{-1} for the high-density and low-density regions, respectively, which correspond to optimal lengths (equal to the coherent lengths) of 56.8 and 94.8 μm for the high-density and low-density regions, respectively, for achieving quasi-phase-matching. The numbers match well with the widths measured from side-scattering images. In addition, simulations based on a method similar to that used in Ref. [18] reproduce the observed sharp resonant dependence on plasma density [shown in Fig. 3(b) by a dashed line] and density modulation parameters. Under the quasi-phase-matching condition the enhancement factor should be about the length of interaction (1 mm) divided by the modulation period (150 μm), which is about 7. This number is close to that observed in the experiment.

In summary, enhancements of relativistic third-harmonic generation by using a uniform plasma waveguide and a periodic plasma waveguide were demonstrated. The resonant increase of harmonic energy for the periodic plasma waveguide and the enhancement of collimation relative to the uniform waveguide indicate that quasi-phase-matching has played a major role in the THG enhancement. On-line optimization of the resonant enhancement was made possible by the capability of programmable fabrication of longitudinal density modulation using a liquid-crystal spatial light modulator.

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