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Tunable soft-x-ray radiation by high-order harmonic generation

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We have investigated the effect of free electrons on the spectral properties of high-order harmonics generated by 30-fs Ti:sapphire laser pulses in a neon gas jet. Our observations clearly indicate the possibility of continuously tuning the harmonic wavelength in the region below 20 nm, by taking advantage of the blue shift of the harmonic wavelength induced by the presence of free electrons in the gas. We have experimentally demonstrated that this allows one to cover the entire spectral region between two consecutive harmonics of the unshifted spectrum. Different amounts of blue shift are imparted by simply changing the gas jet position relative to the laser beam waist, namely, by varying the effective laser intensity experienced by the gas jet when it is moved across the focal region. We have also interpreted the experimental results in terms of a simple model of the generation process based on the tunnel ionization of an atom exposed to an ultraintense laser field.

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High-order harmonic (HOH) radiation of very intense infrared lasers represents an interesting source of high brightness vacuum ultraviolet (vuv) and extreme ultraviolet (xuv) radiation [1]. Since harmonics generated in a gas are produced in a coherent process the coherence properties of the primary laser source can be transferred to some extent to the harmonic radiation [2,3], provided that particular experimental conditions are met. Moreover, the HOH pulses last less than the laser pulse [4]. All the mentioned properties make it possible to employ HOH radiation as a xuv source with unprecedented properties in terms of coherence, brightness and peak intensity.

A well known effect caused by the use of very high intensity laser fields in HOH generation is the ionization of the gas medium, which reduces the harmonic conversion efficiency, as a consequence of neutral atom population depletion. This has led to the use of very short pumping laser pulses ($<$100 fs), since in general the gas saturation intensity for tunneling ionization increases by decreasing pulse duration [5].

Thus, for laser pulses lasting less than 100 fs, submillijoule pulse energies can efficiently generate high-order harmonics in the tunneling regime [6], provided the focusing geometry leads to intensities on target of the order of 10^{14}–10^{15} W/cm^2 and not exceeding the barrier suppression intensity of the used noble gas [7,8]. In the same time, laser systems operating in the above-mentioned pulse energy and pulse duration regime can easily operate at a relatively high repetition rate (1 kHz), thus considerably reducing data acquisition time. Another important consequence of the ionization of the gas medium where the harmonics are produced is the blue shift of the harmonics, which is originated by the temporal change in the free electron density ($\partial N_e/\partial t$). The last quantity strongly depends on laser intensity and on the time needed to ionize a considerable fraction of neutral atoms, and it has been recently shown [9] that such a time can be considerably less than the laser-pulse duration. In principle, the blue shift suffered by the harmonics, if properly controlled, can be used for a fine tuning of the coherent radiation generated in the vuv and xuv regions.

The above considerations have led us to study the process of harmonic generation in a neon gas jet by using a 30-fs Ti:sapphire laser source operating at high repetition rate. In particular, we have carefully investigated the spectral properties of the harmonics extending below the 20 nm region (corresponding to harmonic orders between 40 and 100). Special attention has been paid to the analysis of the dependence of the blue shift of the harmonic spectral peaks on the relative position of the gas jet and laser beam waist. The extent of such a shift of the harmonic wavelength can be as large as 3–4 Å in the deep plateau region, thus allowing a continuous, fine, and reliable tuning of the output harmonic wavelength in the interval between two consecutive harmonics of a reference spectrum. Finally, we have also checked our experimental results with a simple theoretical model, based on tunneling ionization of noble atoms exposed to ultrashort and ultraintense laser fields. This has allowed a better characterization of the complex dynamics of the building up of the harmonic pulse in an ionizing gas medium.

We have used a Ti:sapphire laser system with chirped-pulse amplification based on a nine-pass confocal amplifier stage and a prism compressor. This system generates 30 fs, up to 0.8 mJ laser pulses (centered at 796 nm) at 1-kHz repetition rate. Owing to a precise high-order dispersion control up to the fourth order, the system provides high-quality pulses with a symmetric intensity envelope over a range of three orders of magnitude. The steep leading edge over a high dynamic range is essential to prevent an uncontrolled formation of plasma in the gas jet before the peak of the pulse. The laser beam, having a diameter of about 0.7 cm is focused with a lens ($f=23$ cm) through an 0.5-mm-thick fused-silica window into the laser-gas interaction chamber.

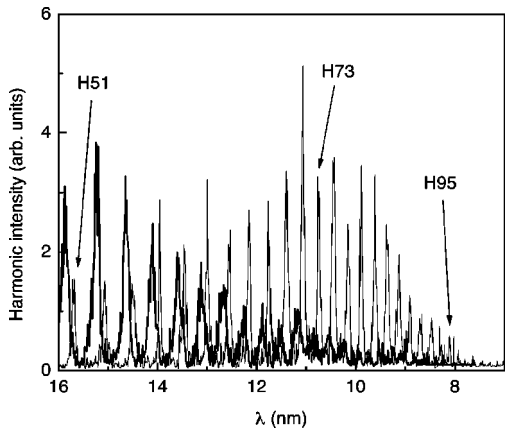

FIG. 1. Harmonic spectra in Ne with pulse energy $E_p = 300\ \mu$J at $z = 2.1$ mm (solid line) and $z = 1.6$ mm (dotted line).

We have measured a confocal parameter $b = 4$ mm. Accordingly, an estimate of the beam radius at the waist is $w_0 = (\lambda b/2\pi)^{1/2} = 22.5$ mm, which exceeds the diffraction limited value by 25%.

The gas sample (Ne) is injected into the interaction chamber by an electromagnetic valve, operating at about 100 Hz, which presently represents the limiting factor of the repetition rate of the whole apparatus. The valve opening time is set to 400 μs, and the injected gas jet has a diameter at nozzle of $\approx$0.8 mm. By varying the gas backing pressure (typically 2–5 bar), the gas pressure in the interaction region has been estimated to vary between 20 and 50 mbar. Such values, as well as the synchronization with the laser pulse, have been set so as to maximize the harmonic signal. Harmonic radiation has been analyzed with a grazing incidence (86°), Rowland mounting monochromator based on a platinum coated, 300 grooves/mm, spherical grating (2 m radius of curvature), designed for broadband efficiency in the 80–5 nm spectral range. A toroidal mirror (incidence angle 83.5°, and radii of 2.8 m in the tangential plane and 60 mm in the sagittal plane) is used to focus the harmonic beam onto the monochromator entrance slit in the tangential plane, matching the instrument aperture. It also provides focusing in the sagittal plane in a position at the center of the diffracted spectrum. By correcting the mounting astigmatism, our two-component optical instrument achieves high sensitivity and high resolving power (typically 1500), thus allowing a detailed analysis of the spectral structure of the harmonics. Finally, the detector is a channel electron multiplier with bare glass photocathode, with variable gain up to the photon counting regime.

We report in Fig. 1 two typical harmonic spectra obtained in Ne with a laser pulse energy $E_p = 300\ \mu$J in correspondence of two different positions, *z*, of the gas jet with respect to the laser beam focus (referenced at $z = 0$). We use positive values for *z* to indicate that the valve is positioned downstream the laser beam waist. For $z = 1.6$ mm, one can clearly observe up to the 95th harmonic ($\lambda_{95} = 8.38$ nm), although smaller peaks up to the 103rd harmonic can be also identified. The absence of background radiation and the high spectral resolution of the monochromator allow a clear detection of neat harmonic peaks, even in the region below 10 nm.

The main features of these spectra are (i) the larger extension of the plateau for the spectrum corresponding to a position of the gas jet closer to the laser beam waist ($z_1 = 1.6$ mm, corresponding to $I \approx 4 \times 10^{14}$ W/cm^2); (ii) a blue shift of the whole spectrum at z_1 with respect to the spectrum obtained at $z_2 = 2.1$ mm ($I \approx 3.2 \times 10^{14}$ W/cm^2). The first characteristic agrees with the existence of an optimum gas jet position (in general different from $z = 0$) that maximizes the harmonic yield, as shown by theoretical calculations [10] and similar experiments with longer laser pulse duration [11,12]. Moreover, we have also found good agreement with the maximum expected order of about 97 and 89 at z_1 and z_2, respectively, according to the cut-off law giving the highest photon energy emitted in a HOH generation process when using ultrashort ($\leqslant$30-fs) laser pulses [13].

On the other hand, the blue shift of harmonics observed when the generating medium is closer to the laser beam focus can be explained by the higher intensity experienced by the gas sample. As previously mentioned, it is originated by the temporal change in the free electron density ($\partial N_e / \partial t$) during harmonic propagation in the ionized gas medium.

We have, thus, investigated the possibility to continuously tune the harmonic wavelength in the spectral interval between two consecutive harmonics of a reference spectrum by simply changing the position of the gas jet relative to the laser beam waist. As a clear example, we report in Fig. 2 the detail of the spectral region around the 73rd harmonic ($\lambda_{73} = 10.9$ nm). The curves are normalized at the peak value. From Fig. 2, we measure a total shift of the 73rd harmonic of more than 3 Å, which is approximately the wavelength separation between the 71st harmonic (also visible on the *red* side of the upper curves) and the 73rd harmonic. In the range of *z* values from 0.4 to 2.2 mm, the laser intensity in the interaction region varies between $\approx 2.4 \times 10^{14}$ W/cm^2 and 4.9×10^{14} W/cm^2.

The effect of a rapidly ionizing atomic population on the spectral characteristics of high-order harmonics is also evident when the laser intensity is kept constant while varying the gas density. This has been verified by changing the value of the valve backing pressure (namely, the local gas pressure). Also in this case we have observed a shift of the harmonic wavelength towards the blue region of the spectrum as long as the gas pressure is increased, although the effect is less pronounced in the investigated pressure range.

As a final remark, let us observe that the blue shift introduced by changing the gas-jet-to-laser-focus relative position is only weakly dependent on the harmonic order for the harmonics in the deep plateau region ($q > 39$, in our case), and that it is much larger than the observed blue shift of the laser wavelength divided by *q*. In fact, we have also measured the fundamental blue shift in several experimental conditions, by means of a shutter which is triggered by the electromagnetic valve driver. In such a way we have been able to select only those laser pulses interacting with the gas medium. As an example, the blue shift measured for the 73rd harmonic at $I_0 \approx 4.9 \times 10^{14}$ W/cm^2 is about 0.3 nm. This would lead to a shift for the fundamental of the order of 73×0.3 nm$=$21.9 nm, while the measured fundamental blue shift at the above pumping intensity amounts to only 5 nm.

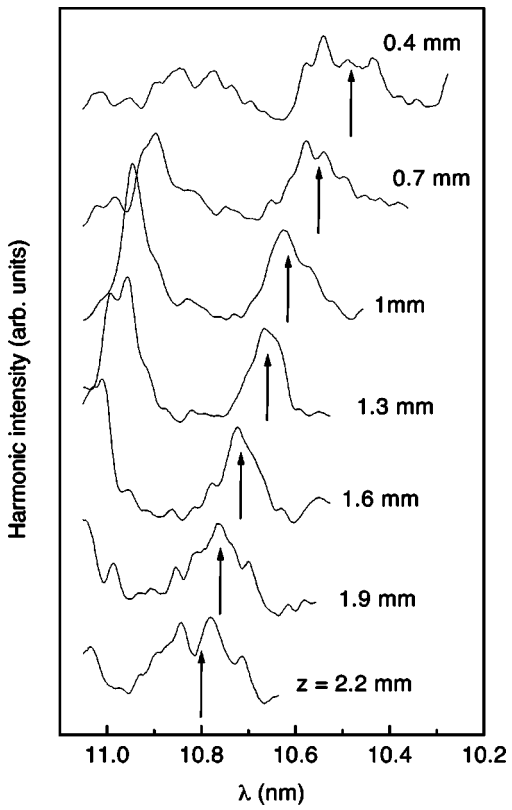

FIG. 2. Detail of the spectral region around the 73rd harmonic (the bluer peak). The experimental conditions are the same as in Fig. 1. Different curves have been obtained at different z positions (from bottom to top: $z=2.2$, 1.9, 1.6, 1.3, 1.0, 0.7, and 0.4 mm). The arrows indicate the central wavelength of the 73rd harmonic power spectrum.

In order to understand our results in terms of a simple theoretical model, we have followed the approach of Miyazaki and Takada [9], which is based on the tunneling ionization of the medium as originally treated by Keldysh [14] and Ammosov *et al.* [15]. Within the framework of this theory, the tunneling ionization rate, $W(t,E)$, for an s state (ground state of Ne) is calculated as (in atomic units)

$$W(t,E)=\left(\frac{3E}{\pi E^*}\right)^{1/2}|C_n|^2\left(\frac{2E^*}{E}\right)^{2n-1}I_p\exp\left(-\frac{2E^*}{3E}\right), \tag{1}$$

where $E(t)$ is the field amplitude at time t, $E^*=(2I_p)^{3/2}$, I_p the ionization energy, C_n a numeric constant (of the order of 2), and $n=Z(2I_p)^{-1/2}$, where Z is the residual ion charge.

According to the semiclassical picture [5,6], which has been confirmed in a large number of experiments, harmonics are emitted in the interaction between a single active electron and its parental ion in a short temporal interval (a few optical cycles) after electron tunneling. In this regard, tunnel ionization is viewed as the first necessary step for the harmonic generation process to occur. Therefore, the harmonic field generated at time t is proportional to the instantaneous electron-ion pair density, $N(t)W(t,E)$, present at time t. Next, by assuming that the problems of phase matching and of the single-atom response can be treated separately, the time-dependent intensity of the qth harmonic field, $I(t,q\omega)$, reduces to

$$I(t,q\omega)\approx|d(q\omega,t)|^2N^2(t)W^2(t,E)|F_q(\Delta k,t)|^2V(t), \tag{2}$$

where $d(q\omega,t)$ is the dipole moment at the frequency of the qth harmonic, $V(t)$ the laser-gas interaction volume at time t, $|F_q(\Delta k,t)|^2$ the phase-matching factor describing propagation effects in the ionizing medium ($\Delta k=k_q-qk_1$), and $N(t)$ the density of neutral atoms at time t.

It is worth stressing that, although our laser pulse duration is almost ten times shorter than the one used by Miyazaki and Takada [9], and only about 11 optical cycles, we still assume in our analysis that the slowly varying envelope approximation holds essentially true, and that the atomic dipole spectral response can be decomposed in discrete spectral components corresponding to odd harmonics of the fundamental frequency, as discussed by several authors [16].

Next, by following Ref. [9] we have replaced $F_q(\Delta k,t)$ in Eq. (2) by the coherence length $L_c=\pi L/(\Delta\phi_g+\Delta\phi_e)$, where L is the medium length. The phase-mismatch term due to the focusing geometry is $\Delta\phi_g\approx 2(q-1)L/b$ ($b\gg L$), and the one due to free electrons is $\Delta\phi_e=q\omega L(n_q-n_1)/c$, where c is the speed of light, and n_1 and n_q the refractive indexes at the laser and qth harmonic wavelengths, respectively, both depending on the time-varying electron density. Therefore, we only consider the harmonics emitted within a single time-dependent coherence length.

We have also assumed a constant value for the atomic dipole d, independent of time and harmonic order. Such an approximation is justified, since the atomic dipole varies very slowly both with the laser intensity in the strong field regime and with the harmonic order for harmonics belonging to the plateau region [17].

Finally, we have replaced in Eq. (2) the time-dependent interaction volume in the gas medium, $V(t)$, with a constant, nearly cylindrical volume V of gaseous medium, having length equal to gas jet diameter and base areas given by the laser cross section at the beam waist, multiplied by a function corresponding to the temporal profile of the laser intensity, $f(t)$. In particular, we have assumed $f(t)=I(t)/I_0=\mathrm{sech}^2(1.76t/\tau)$, where τ is the full-width at half-maximum laser-pulse duration.

With these assumptions, Eq. (2) can be recast as

$$I(t,q\omega)\propto N^2(t)W^2(t,E(t))|L_c|^2f(t), \tag{3}$$

which allows us to calculate the approximate time profile of a given harmonic.

Let us now observe that, due to the tunneling ionization factor, N^2W^2, of Eq. (2), harmonics are emitted well before the laser intensity $I(t)$ reaches its maximum value, given the strong depletion of the gas medium. Let us also define with $t_q(E)$ the time at which the qth harmonic intensity profile reaches its maximum value. According to Yablonovitch [18], the time-dependent spectral shift, $\delta\lambda$, of the laser fundamental wavelength λ is given by

$$\delta\lambda(t) = -\frac{e^2\lambda^3 L}{2\pi m_e c^3}\frac{\partial N_e}{\partial t}. \quad (4)$$

Therefore, the effective laser wavelength at time $t=t_q$, when most of the qth harmonic is generated, is $\lambda+\delta\lambda(t_q)$. Such a shift of the fundamental wavelength essentially induces a harmonic spectral shift $\delta\lambda_q=\delta\lambda(t_q)/q$, which dominates over the shift $\delta\lambda(t_q)/q^3$, due to the refractive index change at λ_q caused by the presence of free electrons. We point out that $\delta\lambda(t_q)$ is much larger than the overall laser blue shift measured with an optical multichannel analyzer. In fact, the laser spectra obtained after the laser-gas interaction region are time integrated. This means that the large blue shift occurring in a very short time interval around t_q (when the laser field is ionizing the medium and the slope of the free-electron density, $\partial N_e/\partial t$, is very large) is partially cancelled out by the time integration over the entire laser-pulse envelope.

By relying on this approach, we have, as an example, calculated the blue shift of the 51st and 73rd harmonics, as a function of the laser intensity for a pulse duration of 30 fs and a gas pressure of 50 Torr. Different laser intensities correspond to different z values. In spite of the simplicity of our model, which also neglects any 3D spatial effects on harmonic spectra, the main experimental features, namely, the strong dependence on laser intensity and the weak dependence on harmonic order of the induced blue shift, are reasonably well recovered. The quantitative agreement between numerical and experimental values is also rather good. This simple model, thus, confirms the dynamical interplay between the temporal evolution of the ionization degree in the medium and the blue shift undergone by the propagating harmonic radiation even in the regime of ultrashort pumping laser pulses.

In conclusion, we have demonstrated the possibility of using HOH generation to produce continuously tunable, coherent radiation in the xuv ($\approx$20 to $\approx$8 nm). Reliable tunability of the harmonic radiation, based on the blue shift undergone by harmonics while propagating in an ionizing gas medium, is obtained by simply adjusting the gas-jet position relative to the laser beam waist.

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