

Gas medium ionization and harmonic wavelength tunability in high-order harmonic generation with ultrashort laser pulses

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Abstract

We have investigated the effect of free electrons on the spectral properties of high-order harmonics generated in a neon gas jet by a 30 fs Titanium:Sapphire pumping laser with intensities in the range $5\text{--}10 \times 10^{14}$ W/cm². The main feature of our observations concerns the possibility of continuously tuning the harmonic wavelength in the spectral region 20–7 nm, by taking advantage of the blue shift of harmonic wavelengths induced by the presence of free electrons to cover the entire spectral region between two consecutive harmonics of the unshifted spectrum. Different amounts of blue shift, which can be as large as 0.3–0.4 nm, are imparted to the given harmonic by simply changing the gas-jet-laser-beam-waist relative position. We have also interpreted our experimental results with a simple model for the generation process based on the “barrier suppression” ionization of an atom exposed to an ultraintense laser field.

1. INTRODUCTION

High-order harmonics (HOH) of very intense infrared laser pulses represent an interesting source of high brightness vacuum ultraviolet (VUV) and extreme ultraviolet (XUV) radiation (Protopapas *et al.*, 1997). Since harmonics generated in gas are produced in a coherent process, it can be shown that the coherence properties of the primary laser source can be transferred to some extent to the harmonic radiation (Ditmire *et al.*, 1996; Bellini *et al.*, 1998), provided that particular experimental conditions are met. Moreover, it can be shown that HOH pulses last less than the laser pulse (Glover *et al.*, 1996). All the mentioned properties of HOH radiation make it possible to employ it as a XUV radiation source with unprecedented properties in terms of coherence, brightness, and peak intensity.

When using very high laser fields, the ionization of the gas sample occurring during the generation process typically reduces the harmonic conversion efficiency, as a consequence of the depletion of the neutral atom population.

Nevertheless, Corkum (1993), and Schafer and coworkers (Schafer *et al.*, 1993) have proposed a simple model for the harmonic generation process, where tunneling ionization plays a fundamental role. According to such a model, an electron escapes to the continuum through the potential barrier created by the atomic and the radiation field potentials, gains energy from the laser field and, within one optical cycle, recombines with the parent atom to the ground state, emitting a high-energy photon. This semiclassical model accounts for the main features of the harmonic generation process and is in agreement with a full quantum-mechanical treatment of the interaction of an atomic system with a very intense radiation field (Lewenstein *et al.*, 1994). It also accounts for the drop in the harmonic conversion efficiency when the laser intensity exceeds a limit value (depending on the laser wavelength, the pulse duration, and the atomic species) corresponding to the barrier suppression. In this case, the atomic potential well is completely suppressed and electrons immediately escape from the parent atom. Suppression of the potential well prevents the free electrons from recombination and, thus, no high-energy photon can be emitted.

For an effective use of the laser pulse energy, it is worth using very short pulses (<100 fs), because in general the

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saturation intensity for the ionization process increases for decreasing pulse duration. Thus, for laser pulses lasting less than 100 fs, submillijoule pulse energies can efficiently generate high order harmonics in the tunneling regime (Christov *et al.*, 1997), provided the focussing geometry leads to an intensity on the gas target of the order of 10^{14}W/cm^2 and not exceeding the barrier suppression intensity. At 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 (typically 1 kHz).

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 (Miyazaki & Takada, 1995) that such a time can be considerably less than the laser pulse duration. In principle, the blue shift undergone 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, z , 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 barrier suppression 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.

2. EXPERIMENT

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 a 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 laser beam is focused with a lens ($f = 23 \text{ cm}$) through a 0.5 mm thick fused-silica window into the laser-gas interaction chamber. We have measured a confocal parameter $b = 4 \text{ mm}$. Accordingly, an estimate of the beam

radius at the waist is $w_0 = (\lambda b/2\pi)^{1/2} = 22.5 \mu\text{m}$, which exceeds the diffraction limited value by 25%.

The gas sample (Ne) is injected into the interaction chamber by an electromagnetic valve (up to 100 Hz repetition rate) producing a jet diameter at nozzle of 0.8 mm, and operating with a valve opening time of 300 μs . 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 nm–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 spectral resolution (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.

3. EXPERIMENTAL RESULTS

In Figure 1 we report a typical spectrum obtained in Ne with $E_p \approx 520 \mu\text{J}$. Only the short wavelength region of the spectrum is shown. We can clearly observe up to the 95th harmonic ($\lambda_{95} = 8.4 \text{ nm}$), although one can identify smaller peaks up to the 103rd harmonic. The large value of the signal-to-background ratio is due both to the absence of back-

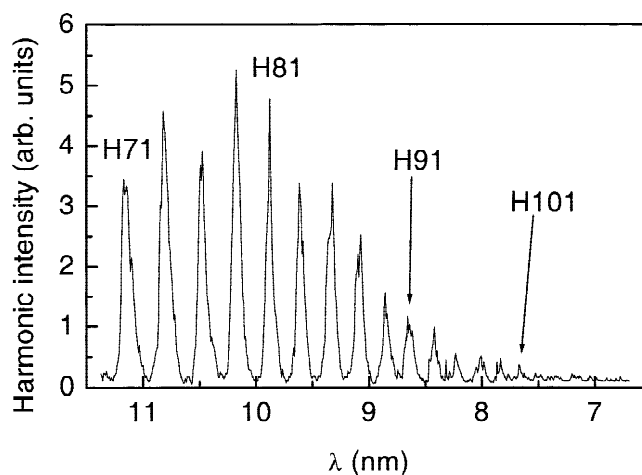


Fig. 1. Harmonic spectrum in Ne with pulse energy $E_p = 520 \mu\text{J}$ at $z = 1.6 \text{ mm}$.

ground radiation and to the high spectral resolution of the monochromator. In order to maximize the harmonic photon yield in the short wavelength region, we positioned the gas jet at a distance $z = 1.6$ mm downstream to the laser beam waist. The existence of an optimum gas jet position (different from $z = 0$) is in agreement with theoretical calculations (Salières *et al.*, 1995) and with similar measurements performed by several authors (Sakai & Miyazaki, 1994; de Lisio *et al.*, 1998).

In Figure 2 we show two spectra in the same experimental conditions except for a different position, z , of the gas jet with respect to the laser beam focus (placed at $z = 0$). The main features emerging from the figure are: (1) a larger extension of the plateau for the spectrum corresponding to a position of the gas jet closer to the laser beam waist (namely $z_1 = 1.6$ mm); (2) a blue-shift of the whole spectrum with respect to the spectrum obtained at $z_2 = 2.1$ mm. The two positions z_1 and z_2 correspond to a laser intensity (in the gas sample) of $\approx 6.7 \times 10^{14}$ W/cm² and $\approx 5.3 \times 10^{14}$ W/cm², respectively.

The maximum observed harmonic order of about 97 in the first case and 89 in the second one are in good agreement with the maximum expected order according to the cutoff law giving the highest photon energy emitted in a HOH generation process when using ultrashort laser pulses (<30 fs) (Chang *et al.*, 1997).

The blue shift of harmonics when the generating medium is closer to the laser beam focus can be explained by the higher intensity experienced by the gas sample. In fact, it is originated by the temporal change in the free electron density ($\partial N_e / \partial t$) which is the larger the higher the laser intensity and the shorter the time interval necessary for the ionization of a considerable fraction neutral atoms. It has been shown (Miyazaki & Takada, 1995) that in similar experimental con-

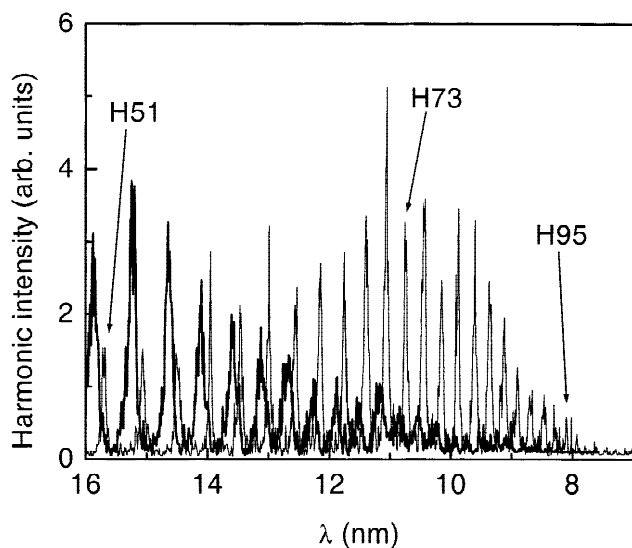


Fig. 2. Harmonic spectra in Ne with pulse energy $E_p = 520 \mu\text{J}$ at $z = 2.1$ mm (thick line) and $z = 1.6$ mm (thin line).

ditions, such a time interval lasts considerably less than the laser pulse.

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 Figure 3 the detail of the spectral region around the 73rd harmonic ($\lambda_{73} = 10.9$ nm). The curves are normalized at the peak value. From Figure 3, we measure a total shift of the 73rd harmonic of more than 3 \AA , 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 mm to 2.2 mm, the laser intensity in the interaction region varies between $\approx 5 \times 10^{14}$ W/cm² and $\approx 10^{15}$ W/cm².

The effect of a rapidly ionizing atomic population on the spectral characteristics of high-order harmonics is also evi-

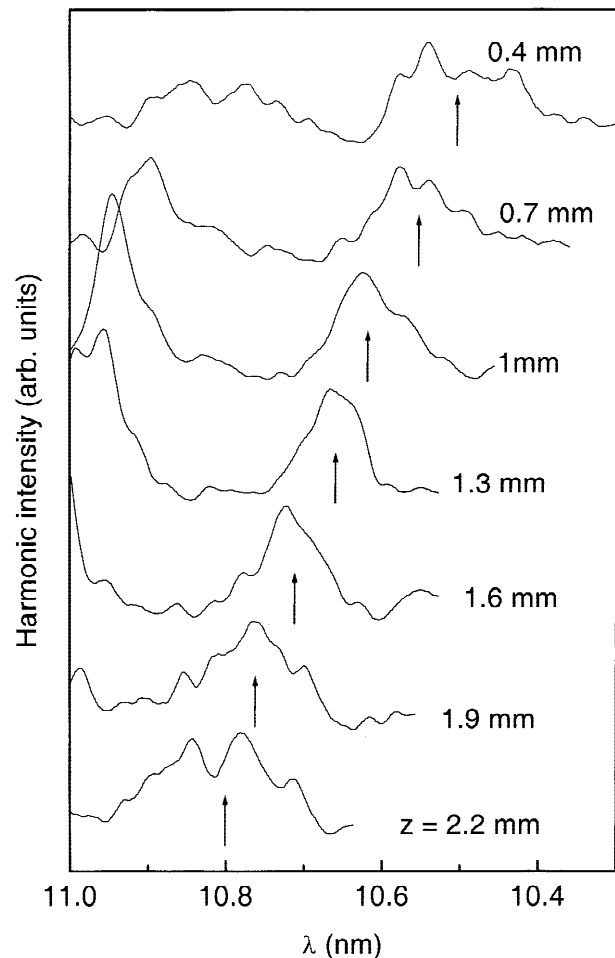


Fig. 3. Detail of the spectral region around the 73rd harmonic (the bluer peak). The experimental conditions are the same as Figure 1. Different curves have been obtained at different z positions (from bottom to top: $z = 2.2, 1.9, 1.6, 1.3, 1.0, 0.7, 0.4$ mm). The arrows indicate the central wavelength of the 73rd harmonic power spectrum.

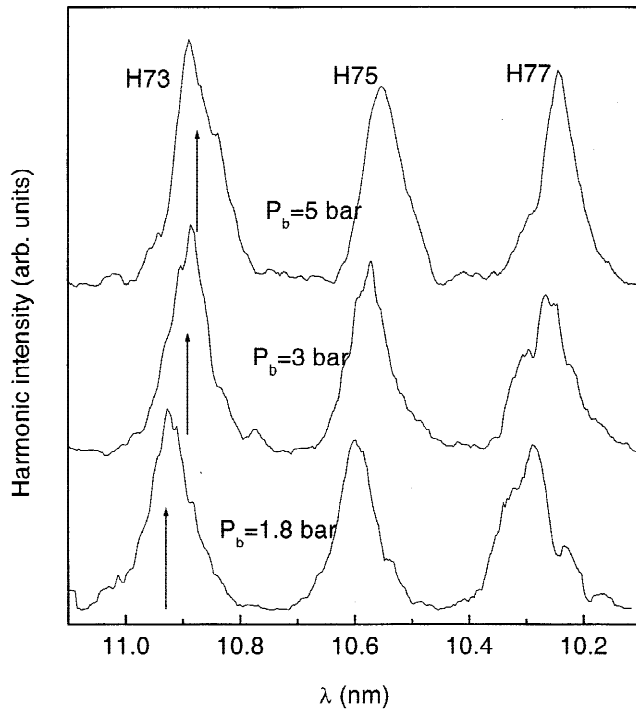


Fig. 4. Detail of the spectral region around the 73rd harmonic. Different curves have been obtained at different backing pressures, P_b (from bottom to top: $P_b = 1.8, 3, 5$ bar).

dent when the laser intensity is kept constant while varying the gas density. Figure 4 shows three curves obtained at different values of the valve baking pressure (namely, at three different local gas pressures). Also in this case we observe a shift of the harmonic wavelength towards the blue region of the spectrum as long as the gas backing pressure increases,

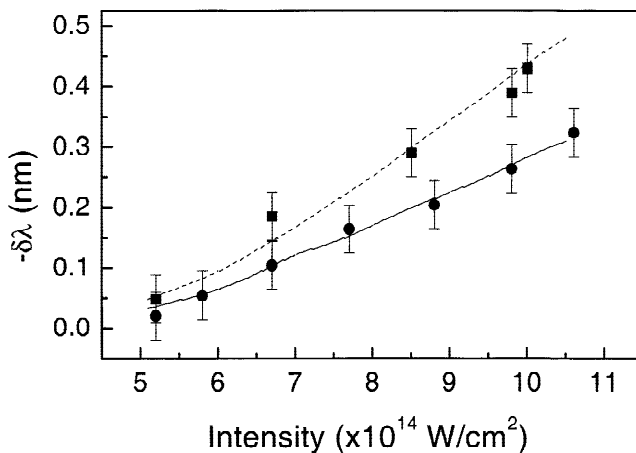


Fig. 5. Calculated blue shifts for the 51st (dashed line) and 73rd (solid line) versus laser intensity for a Ne local pressure of 50 Torr. The corresponding experimental points (squares for H51 and circles for H73) are also reported.

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 . As an example, the blue shift measured for the 73rd harmonic at $I_0 \approx 10^{15}$ W/cm² 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 blue shift of the fundamental wavelength amounts to only 5 nm, as determined by an Optical Multichannel Analyzer (OMA). A possible explanation is the following: An OMA performs a *time average* of the power spectrum over the entire laser pulse duration, whereas the *instantaneous* blue shift depends on the *instantaneous* ionization rate. If a given harmonic is generated in a time interval considerably shorter than the laser pulse duration, its blue shift must reflect what the *instantaneous* blue shift (and, in turn, the *instantaneous* ionization rate) was at the instant when the harmonic was generated. With this picture in mind, it is reasonable to expect that the fundamental wavelength blue shift deduced from the harmonic blue shift can be much larger than the *time averaged* direct measurement.

4. THEORETICAL BACKGROUND

In order to understand the most relevant features of our experimental results, that is, the considerable and somehow surprising blue shift undergone by the harmonics in the deep plateau region, we have interpreted these results in terms of a simple theoretical model, following the approach of Miyazaki and Takada (1995), which was based on the ADK-tunneling ionization of the medium as originally treated by Keldysh (1965) and Ammosov *et al.* (1986).

Since the laser intensities of our experiment ($5 - 10 \times 10^{14}$ W/cm²) approach the barrier suppression intensity of Ne ($\approx 8.6 \times 10^{14}$ W/cm²), we have used the ionization rate formula originally proposed by Krainov (1997) and Bauer and Mulser (1999), which incorporates barrier suppression ionization in the ADK theory.

As a consequence, the ionization rate, $W[E(t)]$, is calculated as (in atomic units)

$$W[E(t)] = \frac{4\sqrt{3}}{\pi n} \frac{E(t)}{(2E(t))^{1/3}} \left(\frac{4eI_p^{3/2}}{E(t)n} \right)^{2n} \times \int_0^\infty Ai^2 \left(x^2 + \frac{2I_p}{(2E(t))^{2/3}} \right) x^2 dx \quad (1)$$

where Ai denotes the Airy function, $E(t)$ is the field amplitude at time t , I_p is the ionization potential, e is the Euler's number, and $n = Z(2I_p)^{-1/2}$ Z being the residual ion charge. Since we limit ourselves to the case of singly ionized atoms ($Z = 1$).

Thus, by assuming that the problems of phase matching and of the single-atom response can be treated separately, the time dependent intensity of the q th harmonic field, $I(t, q\omega)$, reduces to:

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

where $d(q\omega, t)$ is the dipole moment at the frequency of the q th 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 , given by

$$N(t) = N_0 - N_e(t) = N_0 \exp \left[- \int_{-\infty}^t W(t', E(t')) dt' \right], \quad (3)$$

N_0 being the initial atomic density.

It is worth stressing that, although our laser pulse duration is almost ten times shorter than the one used by Miyazaki and Takada (1995), 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 (Salières *et al.*, 1998).

Next, by following Miyazaki and Takada (1995) 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 q th 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 moment, d , substantially independent of time and harmonic order. Such an approximation is based on the behavior of the atomic dipole moment strength as a function of the laser intensity within the adiabatic approximation (Lewenstein *et al.*, 1995). For harmonics in the deep plateau, in fact, the dipole moment strength rapidly increases with laser intensity up to a saturation value which, in neon, is reached at about $2 - 3 \times 10^{14}$ W/cm² (Lewenstein *et al.*, 1995). Then the dipole strength saturates, although rapid and intense oscillations are calculated. These oscillations, due to quantum interference of different electron trajectories, are washed out by propagation effects [see, e.g., Kan *et al.*, 1995 and Lewenstein *et al.*, 1995]. They can, thus, be neglected in the macroscopic response of the gaseous medium. Since high order harmonics require rather high laser intensities to be generated, they are produced only at intensities corresponding to the saturated response of a single dipole moment. As an example, in our experimental conditions the 73rd har-

monic in Ne is generated at intensities $\geq 4.9 \times 10^{14}$ W/cm², according to the *cut-off* law. Such an intensity is already well beyond the intensity threshold for the saturation of the dipole moment intensity response.

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 a height 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 $f(t) = I(t)/I_0 = \text{sech}^2(1.76t/\tau)$, where τ is the FWHM laser pulse duration.

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

$$I(t, q\omega) \propto N^2(t) W^2[t, E(t)] |L_c|^2 f(t), \quad (4)$$

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

Let us now observe that, due to the tunnelling ionization factor, $N^2 W^2$, 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 q th harmonic intensity profile reaches its maximum value. According to Yablonovitch (1988), the time-dependent spectral shift, $\delta\lambda$, of the laser fundamental wavelength λ is given by

$$\delta\lambda(t) = - \frac{e^2 \lambda^3 L}{2\pi m_e c^3} \frac{\partial N_e}{\partial t}. \quad (5)$$

Therefore, the effective laser wavelength at time $t = t_q$, when most of the q th 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 $\delta\lambda_{tot}$ of the pumping laser pulse, that we have 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. The overall fundamental blue shift results from integration of the contributions to the spectral shift, each weighted with a coefficient proportional to the instantaneous laser pulse intensity:

$$\delta\lambda_{tot} = \frac{\int_{-\infty}^{\infty} I(t) \delta\lambda(t) dt}{\int_{-\infty}^{\infty} I(t) dt}. \quad (6)$$

From this expression, it turns out that the overall spectral shift of the laser pulse, $\delta\lambda_{tot}$, is much smaller than the maximum value, $\delta\lambda(t_q)$, obtained when the ionization rate is maximum. Thus, the measured harmonic blue shifts are much more relevant than it can be expected just by dividing the time integrated fundamental blue-shift by the harmonic order, namely, $\delta\lambda_{tot}/q$.

By relying on this approach, we have, as an example, calculated the blue shift of the 51st (solid line) and 73rd (broken line) harmonics, reported in Figure 3 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. We also report in the same figure the corresponding experimental points as obtained from Figure 2 and similar measurements performed on the 51st harmonic. The main experimental features, namely the strong dependence on laser intensity and the weak dependence on harmonic order of the induced blue shift, are fully recovered by the model, and also the quantitative agreement between numerical and experimental values is 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.

5. CONCLUSIONS

In conclusion, we have demonstrated the possibility of using HOH generation to produce continuously tunable, coherent radiation in the XUV (≈ 20 nm– ≈ 7 nm). Due to the high spectral resolution of the monochromator employed in our experiment and to a good signal-to-noise-ratio, all harmonic spectra do not exhibit any remarkable background in the free spectral range between harmonics, which remain well distinguished even at very high orders. This achieves good reliability of our measurements. Tunability of 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. Blue shift of harmonics, though less remarkable, is also observed when increasing the atomic density of the medium by increasing the valve backing pressure at a fixed gas jet position and laser intensity.

The results have been interpreted with a simple model based on the barrier suppression ionization of atoms due to an intense laser field, namely, a pure intensity effect. However, we expect that other factors, such as the atomic dipole phase and the intrinsic laser pulse chirp, not accounted for

by the present model, can play a role in contributing to the observed blue shift. Moreover, the phase mismatch due to free electrons is introduced in our model in terms of a 1D, uniformly ionized medium much shorter than the laser confocal parameter. Thus, transverse effects due to different relative positions of gas jet and laser beam waist are obviously overlooked. Nevertheless, in spite of its simplicity, our model is able to reproduce the main qualitative features of our observations and also to lead to a reasonably good quantitative agreement with measurements.

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