Spectral Features and Modeling of High-Order Harmonics Generated by Sub-10-fs Pulses

P. Villoresi, P. Ceccherini, L. Poletto, and G. Tondello
Istituto Nazionale Fisica della Materia, Laboratorio di Elettronica Quantistica–D.E.I., Università di Padova, Padova, Italy

C. Altucci
Istituto Nazionale Fisica della Materia, Dipartimento di Chimica, Università della Basilicata, Potenza, Italy

R. Bruzzese and C. de Lisio
Istituto Nazionale Fisica della Materia, Dipartimento di Scienze Fisiche, Università di Napoli "Federico II,", Napoli, Italy

M. Nisoli,* S. Stagira, G. Cerullo, S. De Silvestri, and O. Svelto
Istituto Nazionale Fisica della Materia, Centro di Elettronica Quantistica e Strumentazione Elettronica–C.N.R., Dipartimento di Fisica, Politecnico, Milano, Italy
(Received 30 November 1999)

Harmonic radiation generated in a neon gas jet by sub-10-fs laser pulses was investigated both experimentally and theoretically. The spectral profile of the harmonics with respect to the order, their intensity and relative spectral shifts were measured as a function of the position of the gas jet. The results point out spectral features typical of the quasi-single-cycle excitation regime. A nonadiabatic three-dimensional numerical model was developed, which provides harmonic spectra in remarkable agreement with the experiments.

PACS numbers: 42.65.Ky, 42.50.Hz, 42.65.Re

High-order harmonics generated in noble gases provide a unique source of high brightness, coherent extreme ultraviolet radiation [1–3], due to a tabletop size apparatus and a very short duration. When an intense short laser pulse interacts with an atomic gas, tunneling ionization occurs followed by acceleration of the ionized electron by the laser field and generation of high-order harmonics during the recombination process. Upon decreasing the excitation pulse duration, the harmonic conversion efficiency is expected to increase [4–6] and higher photon energies (i.e., shorter wavelengths) are also expected because the electrons are released into stronger laser fields. Generation of coherent emission up to the "water window" (4.4–2.3 nm) was indeed demonstrated in helium by using 26-fs [3], and sub-10-fs [7] pulses obtained by the hollow fiber compression technique [8,9]. The advent of high-peak power few-optical-cycle (sub-10-fs) laser pulses extends the scenario of the harmonic generation process, pointing out a number of new physical problems to be addressed both from an experimental and a theoretical point of view.

In this Letter we investigate the spectral properties of harmonic radiation generated by sub-10-fs laser pulses, namely, the linewidths of the harmonic profiles, the conversion efficiency, and the spectral shift of the harmonic peaks in different experimental conditions. We present a three-dimensional numerical model, which takes into account nonadiabatic effects, typical of the few-optical-cycle regime.

The Ti:sapphire laser system used for the experiments generates 30 fs, up to 0.8-mJ laser pulses (centered at 795 nm) at a 1-kHz repetition rate. Sub-10-fs pulses are generated by the hollow fiber compression technique [8–10]. The amplitude and phase of the compressed pulses have been measured with spectral phase interferometry for direct electric field reconstruction [11,12]. Typical pulse duration ranges from 5 to 7 fs, and the spectral phase is nearly constant over the pulse spectrum. The sub-10-fs pulses are focused onto the gas jet by a 25-cm focal length silver mirror. The laser-gas interaction length is ~1 mm, with a gas pressure of ~50 torr. Harmonic radiation is analyzed with a grazing incidence (86°), Rowland mounting monochromator based on a platinum-coated, 300-grooves/mm, spherical grating (2-m radius of curvature).

Figure 1(a) shows a typical harmonic spectrum obtained in neon, for a laser peak intensity of $\sim 9 \times 10^{14}$ W/cm^2 at the focus. The spectrum shows discrete and well-resolved harmonics up to the 97th order (~80 Å). The integrated emission in the spectral region 12–40 nm has been estimated to be 3 pJ per pulse, corresponding to a conversion efficiency of 2×10^{-8}. This is the first measurement of discrete high-order harmonic emission with sub-10-fs driving pulses. By using sub-10-fs pulses a quasicontinuum harmonic spectrum has been reported by Schnürer *et al.* in neon [13]. For these measurements the laser-gas interaction geometry was different from that used in our experiment in terms of interaction lengths and gas pressure (~0.5 bar). Taking advantage of the well-resolved harmonic spectrum we have analyzed the wavelength dependence of the harmonic peak linewidths. The experimental results are depicted by the filled circles in the upper panel of Fig. 2. The linewidth (at $1/e$ of the harmonic peak value) was evaluated by fitting the spectral profile of a single harmonic with a Gaussian profile.

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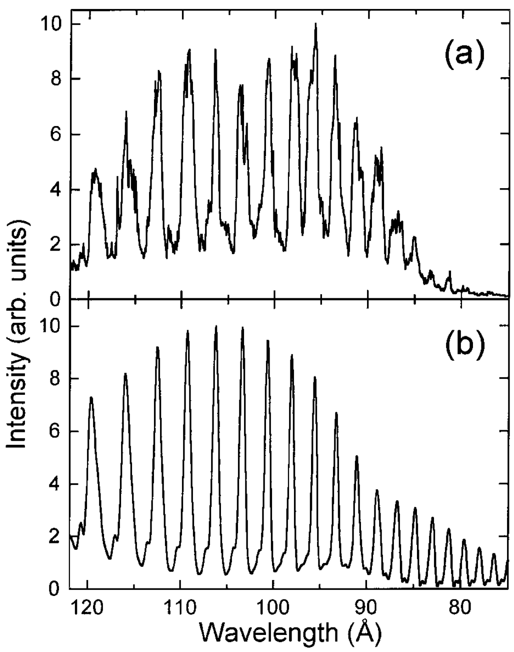

FIG. 1. Harmonic spectrum in neon generated by 7-fs excitation pulses: (a) experimental result; (b) computer simulation obtained using the numerical model described in the text.

The contribution of the instrumental function was then quadratically subtracted. To understand the role of pulse duration on the spectral behavior, we have performed measurements using 30-fs excitation pulses in the same focusing geometry. To this purpose the hollow fiber was evacuated, in order to avoid pulse spectral broadening, and the chirped-mirror compressor was removed. The measured linewidths of the harmonics generated by 30-fs pulses are depicted by the triangles in the lower panel of Fig. 2. The harmonic order dependence of the linewidths is strikingly different in the two cases: for 30-fs pulses the linewidth increases by a factor of 4 in the 12–26 nm wavelength range, while for the 7-fs pulses, it remains nearly constant with harmonic order.

When the interaction geometry is changed by moving the gas jet along the beam axis, remarkable variations have been observed in the harmonic spectrum produced by pulses with duration from 1 ps [1] down to 30 fs [14]. This behavior is related to the phase-matching and plasma contributions in the harmonic generation process [15,16]. In the sub-10-fs regime an experimental study of the influence of the gas jet position on the conversion efficiency is still lacking. Figure 3(a) shows the measured spectrum of the 53rd and 55th harmonics for different positions of the gas jet. Since the exact determination of the beam waist position is affected by a significant experimental error, in Fig. 3(a) each spectrum is identified by the displacement δ of the jet position from that corresponding to maximum blueshift of the harmonic peaks (δ increases in the direction of propagation of the excitation beam). Ranging from $\delta = 3$ mm to $\delta = 0$, we measured a maximum blueshift

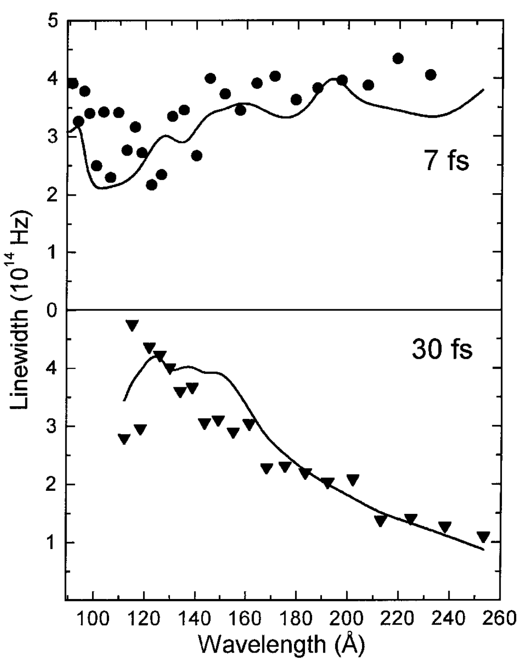

FIG. 2. Linewidths of the harmonics generated by 7-fs (upper panel) and 30-fs (lower panel) pulses. The points represent the experimental data, while the curves are the results of computer simulations.

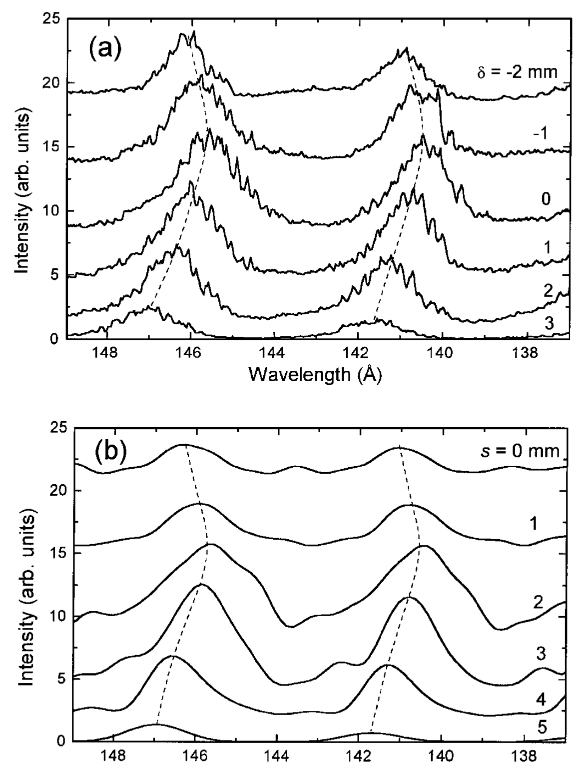

FIG. 3. Details of 53rd and 55th harmonics generated by 7-fs pulses for different gas jet positions: (a) experimental results; (b) calculated spectra. The dashed lines are a guide to the eye indicating the harmonic spectral shift.

of ~1.4 and ~1.2 Å for the 53rd and 55th harmonics, respectively. It is worth pointing out that this spectral shift turns out to be ~3 times smaller than that measured, in the same spectral region, in the case of 30-fs excitation pulses with a peak intensity (in the focus) ~1.8 times smaller [14]. Figure 4 shows as filled circles the normalized integral of two harmonic peaks (corresponding to the 55th and 85th order) as a function of δ. Both curves present a maximum located at $\delta = 0$.

The experimental results have been analyzed in terms of a three-dimensional nonadiabatic numerical propagation model. Previous calculations have either used a three-dimensional treatment in the adiabatic approximation [17–19] or a nonadiabatic approach in a one-dimensional model [4,20]. For few-optical cycle pulses the adiabatic approach is no longer valid, because when there is a significant envelope variation within a single optical cycle it is not meaningful to calculate the response to an instantaneous intensity but only to the entire driving pulse. A relevant aspect of our numerical investigation is that three-dimensional effects are essential to achieve a good agreement with the experimental data. In our model the single-atom response is calculated using the strong field model [21], generalized to account for nonadiabatic effects, using the full electric field of the laser pulse to calculate the nonlinear dipole moment, $p_{nl}(r, z, t)$, where z is the propagation coordinate and r is the transverse coordinate. Gas ionization and ground state depletion are calculated using the Ammosov-Delone-Krainov theory [22], which can still correctly provide the free electron density, $n_e(r, z, t)$, at laser intensities not exceeding 10^{15} W/cm^2 (see, e.g., Ref. [23]). The propagation of the fundamental beam is calculated by solving the following equation:

$$\nabla_\perp^2 E - \frac{2}{c}\frac{\partial^2 E}{\partial z' \partial t'} = \frac{\omega_p^2(r, z', t')}{c^2} E, \qquad (1)$$

where we have used the moving coordinate frame $z' = z$ and $t' = t - z/c$, and $\omega_p(r, z', t') = [n_e(r, z', t')e^2/(\varepsilon_0 m_e)]^{1/2}$ is the plasma frequency. In Eq. (1) the term $\partial^2 E/\partial z'^2$ has been omitted. This approximation, termed slowly evolving wave approximation, holds true independently of laser pulse duration [24]. Equation (1) takes into account both temporal plasma-induced phase modulation and spatial plasma lensing effects on the fundamental beam. For the propagation of the harmonic beam the following equation is solved:

$$\nabla_\perp^2 E_h - \frac{2}{c}\frac{\partial^2 E_h}{\partial z' \partial t'} = \mu_0 \frac{\partial^2}{\partial t'^2}\{[n_0 - n_e(r, z', t')] \times p_{nl}[E(r, z', t')]\}, \qquad (2)$$

where n_0 is the initial neutral atom density, and the dipole moment, p_{nl}, is calculated at each position inside the jet using the fundamental field derived from the previous equation. The effect of gas absorption for the harmonic beam was also included.

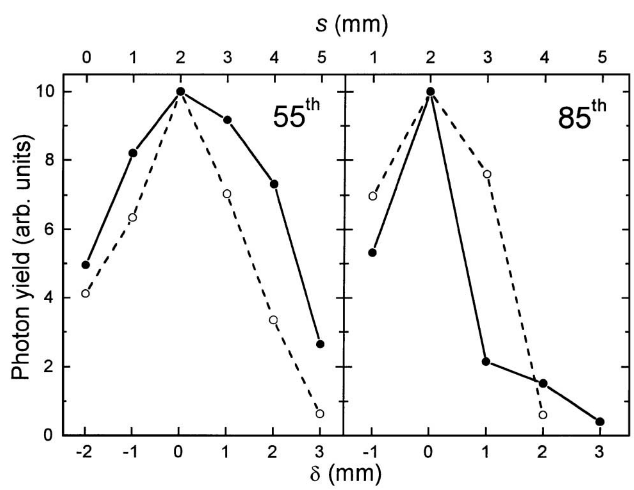

FIG. 4. Normalized integral of 55th and 85th harmonics generated by 7-fs pulses for different positions of the gas jet (δ is the displacement of the jet position from that corresponding to maximum blueshift of the harmonic peaks; s is the position of the gas jet with respect to the laser beam waist). Filled circles and solid lines correspond to experimental data; hollow circles and dashed lines correspond to the results of the numerical model.

Figure 1(b) shows the calculated harmonic spectrum using the same experimental parameters of measured spectrum in Fig. 1(a). The computer simulation is in remarkably good agreement with the experimental result, in terms of the widths and relative amplitudes of the harmonic peaks (note that the experimental harmonics close to the cutoff are broadened by the instrumental function). The simulation shows that the increased efficiency measured at wavelengths shorter than ~11.5 nm is mainly due to the reduction of neon absorption in this spectral region. The same model was applied to simulate harmonic spectra using 30-fs pulses. The upper and lower panels of Fig. 2 show by solid lines the calculated linewidths in the case of 7- and 30-fs pulses, respectively. Also in this case the numerical simulation appropriately reproduces the main features of the experimental results. For 30-fs driving pulses, the wavelength dependence of the linewidths can be at least partially understood in terms of the different duration of the emission of the harmonics: Those in the plateau are emitted over several optical cycles, while those approaching the cutoff are emitted only over a few cycles, close to the laser peak intensity. For 7-fs excitation pulses, the linewidths remain nearly constant since all the harmonics are emitted over a few cycles.

Concerning the spectral behavior versus jet position, Fig. 3(b) displays portions of calculated harmonic spectra around the 55th harmonic, for different positions, s, of the gas jet with respect to the laser beam waist (s increases in the direction of propagation of the excitation beam). We see an overall good agreement between calculated and experimental spectra [see Fig. 3(a)], if we assume that the experimental spectrum labeled with $\delta = 0$ corresponds to a gas jet position located 2 mm downstream the beam waist ($s = 2$ mm). The calculated conversion efficiencies for the 55th and 85th harmonics, reported as dashed lines in Fig. 4, are also in qualitative agreement with the experimental data. The maximum blueshift is not obtained where the intensity is maximum (i.e., when the gas jet is located in the beam waist), contrary to what is observed for longer (30-fs) pulses [14]. In the case of 30-fs pulses the harmonic blueshift has been interpreted in terms of plasma-induced shifting of the fundamental wavelength [14]. The numerical model suggests that, in the case of few-optical-cycle pulses, the physical mechanisms leading to harmonic spectral shift are different. To test this assumption we performed additional simulations neglecting the influence of free electrons on the fundamental beam. The calculated harmonic spectra show a behavior similar to that reported in Fig. 3(b): This confirms that plasma-induced effects do not play a relevant role. Therefore we attribute the spectral shift in the sub-10-fs regime to a nonadiabatic effect [25,26]. An electron that enters the continuum while the laser intensity is increasing or decreasing experiences an additional acceleration or deceleration that modifies the dipole phase related to the electron trajectory with respect to the case of an excitation with quasiconstant field amplitude (long pulses). Phase-mismatch mechanisms, due to longitudinal and radial variations of intensity and phase of the driving pulse, suppress some of the emitted spectral components, giving rise to the harmonic peaks. By changing the relative position of the gas jet and the beam waist, different shifts of the emission spectra are expected because the phase-mismatch conditions are varying.

In conclusion, we have investigated, both experimentally and theoretically, the spectral characteristics of harmonic radiation generated by sub-10-fs laser pulses. We have found that the harmonic spectrum shows discrete and well-resolved peaks, whose linewidths remain almost constant as a function of the harmonic order. On the contrary, in the case of 30-fs pulse-driven harmonic radiation the linewidth decreases upon decreasing harmonic order. The photon yield and the harmonic spectral shift as a function of the position of the gas jet have also been measured. A numerical model employing a nonadiabatic three-dimensional treatment has been developed. The results of the numerical simulations are in remarkable agreement with the experimental data.

We acknowledge valuable contributions of E. Priori for the development of the numerical model, and of P. Barbiero for his help in the data analysis. This study was partially supported within the framework of the Istituto Nazionale per la Fisica della Materia under the project "Femtosecond soft-x-ray generation by high energy laser pulses."

*Email address: mauro.nisoli@fisi.polimi.it

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