Effects of Carrier-Envelope Phase Differences of Few-Optical-Cycle Light Pulses in Single-Shot High-Order-Harmonic Spectra

M. Nisoli,* G. Sansone, S. Stagira, and S. De Silvestri
National Laboratory for Ultrafast and Ultraintense Optical Science-INFM, Dipartimento di Fisica, Politecnico, Milano, Italy

C. Vozzi
INFM-Dipartimento di Fisica, Universitá degli Studi, Milano, Italy

M. Pascolini, L. Poletto, P. Villoresi, and G. Tondello
Laboratory for Ultraviolet and X-Ray Optical Research-INFM, D.E.I., Universitá di Padova, Padova, Italy
(Received 30 April 2003; published 20 November 2003)

For the first time single-shot harmonic spectra generated by few-optical-cycle pulses have been measured. Clear carrier-envelope phase effects have been observed in the cutoff harmonic spectral structure. Results have been interpreted in terms of the nonadiabatic single-atom response of the nonlinear medium excited by few-optical-cycle pulses.

DOI: 10.1103/PhysRevLett.91.213905 PACS numbers: 42.65.Ky, 42.50.Hz, 42.65.Re

Progress in femtosecond laser technology has led to the generation of laser pulses whose envelope varies on a time scale comparable to that of the electromagnetic field itself. Recently, pulses as short as 3.8 fs have been generated, with energy in the microjoule range, thus approaching the single-cycle regime in the visible spectral region [1]. Under these conditions, the phase of the carrier frequency with respect to the envelope [carrier-envelope phase (CEP)] determines the variation of the laser electric field in time. Since all effects in strong-field laser interaction are driven by the electromagnetic field of the laser, the CEP is important for many different topics in laser physics: generation of attosecond pulses, strong-field photoionization, production of coherent soft x rays, and frequency metrology [2]. Experimental evidences of the role of the CEP have been obtained for the first time in strong-field photoionization, using pulses with random CEP [3]. Phase-dependent changes in the spectral characteristics of the high-order harmonic radiation in the cutoff region have been recently observed, using phase-stabilized pulses [4]. The role of the CEP of few-optical-cycle pulses on high-order harmonic generation has been theoretically investigated [5–9].

In this work, for the first time, *single-shot* harmonic spectra generated by few-optical-cycle pulses have been measured. This experimental approach, together with the excellent stability of the driving pulses, allows one to investigate the role of the CEP of the driving pulses employing a direct method. The experimental results have been interpreted in the framework of a three-dimensional nonadiabatic numerical code [7]. An intuitive picture of the physical processes is given, based on the nonadiabatic single-atom response of the nonlinear medium excited by few-optical-cycle pulses [10].

Twenty-five–fs pulses are coupled into an argon-filled capillary consisting of a 60-cm-long fiber with an inner diameter of 0.5 mm at the entrance side and 0.3 mm at the exit side. The emerging pulses are then compressed by chirped mirrors (five bounces, 30 fs^2 each). Typical pulse duration ranges from 5 to 7 fs. Single-shot, 1-kHz-rate pulse characterization of the compressed pulses has been performed using spectral phase interferometry for direct electric field reconstruction (SPIDER) [11]. These measurements demonstrate the remarkable shot-to-shot stability of the spectral phase, of the temporal profile, and of the spectrum of the compressed pulses. This is of paramount importance for the analysis of the single-shot harmonic spectra. In the experiments the harmonic beam is produced by focusing sub-10-fs laser pulses into a neon jet. The laser-gas interaction length is $\sim$1 mm, with a gas pressure of $\sim$50 torr. To observe the harmonic emission we have used a high-resolution flat-field soft x-ray spectrometer and a high-resolution charge-coupled device (CCD) detector. The spectrometer has been calibrated in order to provide an absolute measurement of the emitted photon flux. The gas jet, synchronized with the driving pulses, was operated at a 10-Hz repetition rate with a 300-μs valve opening time. The integration time of the CCD detector was set to 85 ms: in this way single-shot spectra can be measured. It is worth pointing out both the high brightness of the harmonic source [12] and the design of the spectrometer which allows one to efficiently collect the generated extreme ultraviolet (XUV) photons. These two conditions are essential in order to perform measurements on a single-shot basis, with a good signal-to-noise ratio.

Figure 1(a) shows two single-shot harmonic spectra, for a laser peak intensity on the gas jet of $\sim 6.2 \times 10^{14}$ W/cm^2. Figure 1(b) displays a harmonic spectrum measured in the same experimental condition, using a 10-s integration time, thus corresponding to 100 XUV shots. The single-shot spectra show discrete harmonic peaks,

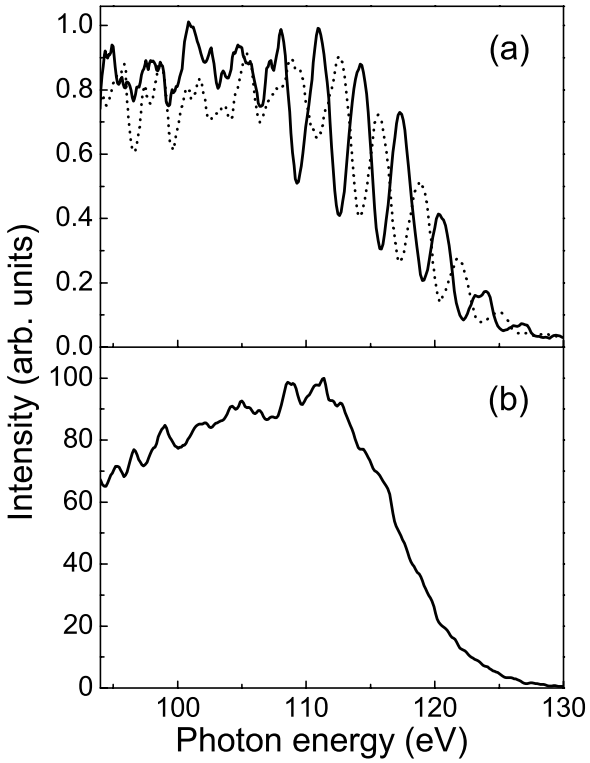

FIG. 1. Measured harmonic spectra in neon generated by 6-fs driving pulses: (a) single-shot harmonic spectra; (b) time-integrated harmonic spectrum corresponding to 100 XUV shots.

with different amplitude, up to the cutoff region. In the cutoff region, the harmonic peaks of the single-shot spectra corresponding to different driving pulses are frequency shifted, while in the plateau region (not shown in Fig. 1) the harmonic peaks are not frequency shifted. This pulse-dependent frequency shift of the harmonic peaks explains why the time-integrated spectrum, which corresponds to several driving pulses with random CEPs, appears as a structureless continuum. It is worth pointing out that the single-shot technique and the excellent stability of the compressed pulses guarantee that the only difference in the driving pulses is their CEP, which randomly changes from one pulse to the other. We have checked that, by using 25-fs driving pulses, the single-shot harmonic spectra do not show changes from one pulse to the other. As a consequence, the integrated harmonic spectrum, corresponding to a few hundred laser shots, shows discrete and well resolved peaks up to the cutoff region. Therefore we can conclude that the frequency shift of the harmonic peaks in the cutoff region is an effect related to the use of few-optical-cycle pulses.

We have then performed a theoretical study of the observed spectral properties of high-order harmonic radiation, using a nonadiabatic three-dimensional numerical propagation model [7]. Figure 2 shows the cutoff region of two harmonic spectra calculated in neon assuming a driving pulse duration of 6 fs, and two different CEPs, namely, $\varphi = 0$ and $\varphi = \pi/2$. We have assumed the following expression for the electric field of the linearly polarized light pulse: $E(t) = A(t)\cos(\omega_0 t + \varphi)$. The other parameters of the simulations are the following: pulse peak intensity on the gas jet 6.2×10^{14} W/cm^2, gas-jet thickness 1 mm, neon pressure 50 torr, and gas jet located 2 mm after the laser focus. The relative frequency shift of the harmonic peaks is clearly visible. In order to simulate the effect of the randomly changing absolute phase, we have averaged the harmonic spectra calculated for eight different CEPs ranging from $\varphi = 0$ to $\varphi = 7\pi/8$: as expected, the harmonic peaks are almost completely smeared out, in agreement with the experimental result shown in Fig. 1(b). Upon increasing the duration of the driving pulses, well resolved harmonic peaks progressively appear in the cutoff region of the averaged spectrum. This is due to the fact that the relative spectral shift of the harmonic peaks for different CEPs decreases upon increasing the driving pulse duration.

Using the numerical simulations it is possible to conclude that the small intensity fluctuations of the driving pulses cannot induce the observed frequency shift. We have indeed calculated the harmonic spectra generated by pulses with fixed CEP, and a 5% shot-to-shot peak intensity fluctuation, larger than the measured one. Apart from a variation of the harmonic intensity, the frequency shifts of the harmonic peaks are negligible. We have also verified that the chirp of the driving pulse, generated by plasma-induced self-phase modulation in the gas jet, which, in principle, could introduce a variation in the harmonic spectral characteristics [13], does not have an appreciable effect. To this purpose, we have calculated the harmonic spectra assuming a low gas pressure (0.5 torr), in order to have a completely negligible chirp on the driving pulse. Also in this case the calculated harmonic spectra clearly display a phase-dependent harmonic shift.

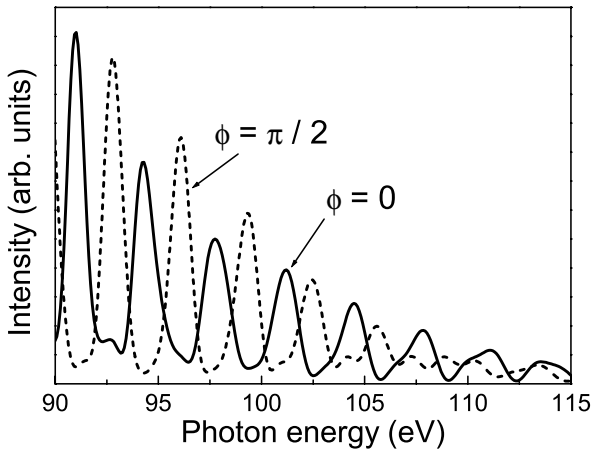

FIG. 2. Harmonic spectra in neon calculated using a nonadiabatic three-dimensional numerical code for two different CEPs of the 6-fs driving pulse.

It is well-known that propagation can strongly influence the spectral characteristics of the harmonic radiation. Moreover, as pointed out by Durfee *et al.* for high-order harmonic generation in hollow waveguides [14], CEP influences the phase-matching mechanisms, due to a CEP-dependent ionization rate. Using the three-dimensional model we investigated the effects of the plasma dispersion on the CEP-dependent spectral shift, performing simulations with different gas pressures (ranging from 0.5 to 50 torr). We observed that, besides an overall blueshift of the spectrum at increasing gas pressure (in agreement with previous results [15]), the spectral shift of a given *harmonic* peak for two different CEPs is almost independent of gas pressure. This suggests that the phase-dependent harmonic shift is a single-atom effect. For this reason we have calculated the nonadiabatic single-atom response to the few-optical-cycle driving field in the strong-field approximation (SFA) [10]. Figure 3 shows the cutoff region of the calculated spectra for two CEPs, namely, $\varphi = \pi/4$ (solid curve) and $\varphi = \pi/2$ (dashed curve), assuming a pulse duration of 6 fs. The harmonic shift is clearly visible, thus providing an additional evidence of the single-atom nature of the observed phenomenon.

A simple physical understanding of the origin of the CEP dependence of the spectral characteristics of the harmonic radiation in the cutoff region can be obtained in the framework of the so-called two-step model [16,17], which can be recovered, as a quasiclassical limit, in the fully quantum-mechanical treatment of the SFA model [10]. This approach has been used by various groups to point out the nonadiabatic origin of important aspects of harmonic radiation, such as the role of the chirp of the driving pulses on the dipole phase [13], which offers the possibility for a coherent control of the spectrum of the high harmonic emission [18,19], and the blueshift of high-order harmonics in an intense laser field [15]. The

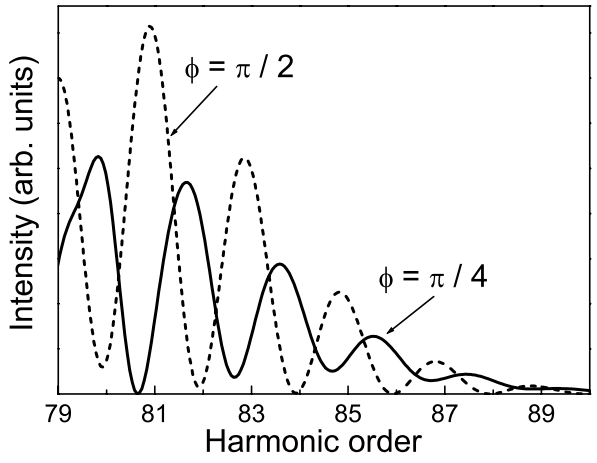

FIG. 3. Nonadiabatic single-atom spectra in neon for two different CEPs of the 6-fs driving pulse.

SFA predicts that for a plateau harmonic two quantum paths, which are associated to two classical electron trajectories, give the most relevant contribution to the dipole moment. One trajectory has a short return time, close to half an optical period, while the second one has a return time close to one period. For a harmonic in the cutoff region, only one quantum path contributes to the generation process, with a return time of approximately half a laser period. The phase associated to each quantum path is given by the following expression [20,21]:

$$\Theta = \omega t_r - \frac{1}{\hbar} S(p_{\mathrm{st}}, t_0, t_r), \tag{1}$$

where ω is the angular frequency of the harmonic radiation, $S(p_{\mathrm{st}}, t_0, t_r)$ is the semiclassical action along the considered stationary action trajectory, and p_{st} is the stationary action value of the momentum of the electron for which the trajectory starting from the atomic core at t_0 returns to it at time t_r. In agreement with the quasiclassical picture, the SFA model predicts that a given harmonic photon energy, $\hbar\omega$, is emitted only at those instants, t_r, at which the electrons returning to the parent ions have the appropriate kinetic energy, $E_k = \hbar\omega - I_p$ [10]. Since when the gas jet is located after the focus the electron trajectory with the short return time is selected [8], we have considered only the short electron quantum paths contributing to the emission in the cutoff region. In the case of few-optical-cycle pulses, only up to two emission processes contribute to the generation in this spectral region. Using Eq. (1) we have calculated, for each electron kinetic energy (and therefore for each harmonic photon energy), the phases, Θ_1 and Θ_2, associated to the two emission processes involved in harmonic generation in the cutoff region. The modulation of the generated (cutoff) harmonic spectrum is determined by the phase difference $\Delta\Theta = \Theta_2 - \Theta_1$, reported in Fig. 4 for three different absolute phases. $\Delta\Theta$ increases almost linearly with frequency: $\Delta\Theta = \alpha\omega$; this leads to the generation of *harmonic* peaks separated by the frequency interval $\Delta\omega = 2\pi/\alpha$. As shown in Fig. 4, $\Delta\Theta$ depends on the CEP of the driving pulses. Depending on pulse peak intensity, the slope, α, of $\Delta\Theta$ and therefore $\Delta\omega$, can be a function of the CEP of the few-optical-cycle driving pulses. Moreover, the $\Delta\Theta$ curves associated with different CEPs are displaced from each other, thus leading to a phase shift of the induced frequency modulation of the harmonic spectrum. This is pictorially shown in the inset of Fig. 4, which displays the modulation of the resultant spectrum induced by the three reported phase differences.

It is worth pointing out the differences with respect to the case of long driving pulses (adiabatic approach). In the adiabatic case the phase accumulated by the electron wave packet during propagation in the laser field between t_0 and t_r, given by the second term in Eq. (1), is the same for consecutive emission processes. Therefore the

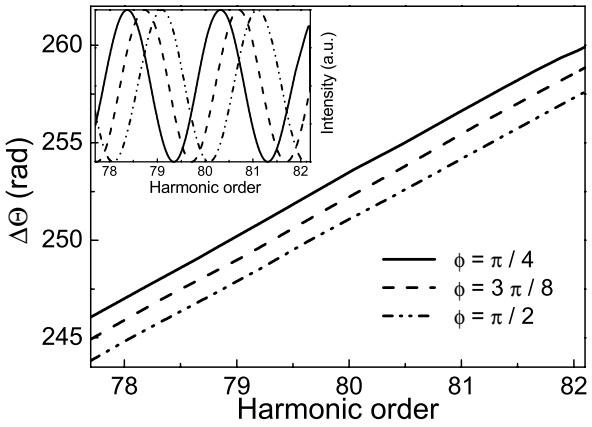

FIG. 4. Phase difference $\Delta\Theta$, as described in the text, for three CEPs. Inset: resulting modulation of the harmonic spectra.

modulation of the resultant spectrum is simply determined by $\Delta\Theta = \omega(t_{r2} - t_{r1}) = \omega T_0/2$, where t_{r1} and t_{r2} are two consecutive electron return instants, and $T_0 = 2\pi/\omega_0$ is the fundamental laser period, independently of the CEP of the pulse. This leads to the generation of the odd harmonics of the fundamental frequency, separated by the frequency interval $\Delta\omega_0 = 2\omega_0$. The calculated $\Delta\Theta$ curves are the same for different pulse CEPs; therefore, the harmonic peaks are not frequency shifted. In this case such peaks really correspond to (odd) harmonics of the fundamental frequency. In the nonadiabatic case, the electron wave packets contributing to the same harmonic photon energy accumulate different phases in the continuum; therefore, the corresponding contribution to $\Delta\Theta$ is nonzero. Moreover, in this case the temporal separation between two consecutive return instants with the same electron kinetic energy, $t_{r2} - t_{r1}$, is no longer constant but slightly depends on the corresponding kinetic energy. These nonadiabatic effects lead to the shift of the harmonic peaks in the cutoff region. In this case, such peaks are no longer associated to real harmonics of the fundamental frequency.

In conclusion, in this Letter we have reported for the first time on *single-shot* measurements of harmonic spectra generated by few-optical-cycle pulses. Clear CEP effects have been observed in the measurements. In particular, a CEP-dependent frequency shift of the *harmonic* peaks has been measured. We have ascribed such behavior to the nonadiabatic single-atom response of the nonlinear medium.

This work was partially supported by the European Community's Human Potential Programme under Contract No. HPRN-CT-2000-00133 (ATTO) and by INFM under the project CLUSTERS.

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

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