Observation of Carrier-Envelope Phase Phenomena in the Multi-Optical-Cycle Regime

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So far the role of the carrier-envelope phase of a light pulse has been clearly experimentally evidenced only in the sub-6-fs temporal regime. Here we show, both experimentally and theoretically, the influence of the carrier-envelope phase of a multi-optical-cycle light pulse on high-order harmonic generation. For the first time, we demonstrate that the short and long electron quantum paths contributing to harmonic generation are influenced in a different way by the pulse carrier-envelope phase.

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Recently, by using few-optical-cycle light pulses, significant developments have been achieved in the field of extreme nonlinear optics, a light-matter interaction regime where the electric field of a light pulse, rather than the intensity profile, is relevant [1]. One of the most important results, due to its general interest, is the generation and application of soft-x-ray attosecond ($1\ \text{as} = 10^{-18}$ s) pulses [2], produced by high-order harmonic generation in gas using few-optical-cycle driving pulses. In this contest, a key parameter of the light pulse electric field, which significantly influences the strong-field interaction, is the phase of the carrier frequency with respect to the envelope. Experimental evidence of the role of the carrier-envelope phase (CEP) has been obtained in strong-field photoionization, using 6-fs pulses with random CEP [3]. It is worth pointing out that CEP effects in the reported experimental measurements completely disappear for light pulse duration exceeding 8 fs. Phase-dependent changes in the spectral characteristics of high-order harmonic radiation were reported in 2003 by Baltuška *et al.,* using phase-stabilized 5-fs pulses [4], and by Nisoli *et al.,* using 6-fs pulses with random CEP [5]. The concept of CEP itself has been supposed to be well defined only for pulses whose duration is comparable with the optical period [6] (in the visible, this corresponds to pulse duration in the sub-10-fs range). The only experimental glimpse of a CEP effect induced by 20-fs pulses was reported in 1999 to justify discrepancies between theoretical and experimental results on the pressure dependence of the yield of harmonic 29 generated in an argon-filled hollow fiber [7].

In this work we demonstrate, both experimentally and theoretically, that the CEP of 20-fs pulses (multicycle regime) induces clear signatures in the spectral characteristics of high-order harmonic radiation. Moreover, we demonstrate, for the first time to our knowledge, that the short and long electron quantum paths contributing to harmonic generation are influenced in a different way by the CEP of a light pulse. The reported results lead to the general conclusion that phase-driven strong-field processes are feasible even in the multicycle regime. This observation is consistent with the results reported by Christov *et al.,* demonstrating that coherent control of electronic processes in the strong-field regime can be achieved by precisely shaping a laser pulse on an attosecond time scale [8].

When an intense light pulse is focused onto a gas medium, electrons are set free by optical field ionization and are accelerated by the pulse electric field. With a small probability such electrons can return to their parent ions and recombine to the ground state emitting the energy gained in the laser field in the form of an energetic photon. This process is repeated periodically 2 times each light period, thus leading to the generation of discrete harmonics of the fundamental radiation. Since the described physical mechanism depends on the electric field of the driving pulse, CEP can be a crucial parameter. In the experiments the harmonic beam is produced by focusing 20-fs light pulses, generated by a Ti:sapphire laser system (800-nm central wavelength, 1-kHz repetition rate), into an argon jet. To observe the harmonic radiation a high-resolution flat-field soft x-ray spectrometer and a high-resolution charge-coupled device (CCD) detector have been used [9]. 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, with a good signal-to-noise ratio. Single-shot characterization of the driving pulses has been performed using spectral phase interferometry for direct electric field reconstruction [10,11]. Average pulse durations are measured to be (20.3 ± 0.4) fs. The single-shot spectra reported in this work correspond to consecutive light pulses with almost identical temporal and spatial intensity profile, spectral amplitude, and phase. The spatial properties of the fundamental beam have been monitored, on a single-shot basis, using a CCD beam profiler. This ensures that the only difference in the driving pulses is their CEP, which randomly changes from one pulse to the other.

Figure 1(a) shows two single-shot harmonic spectra measured when the gas jet was located ~2 mm after the laser focus, for a laser peak intensity of $\sim 2 \times 10^{14}$ W/cm^2 at focus. Well resolved odd harmonics are visible; no radiation is observed between adjacent harmonic peaks. The single-shot harmonic spectra measured in this condition do not show any variation for different driving pulses and, therefore, for different values of the CEP. We point out the remarkable stability of the two single-shot harmonic spectra: this is an additional proof that the corresponding driving pulses are almost identical (apart from their CEP). Figure 1(b) displays two single-shot harmonic spectra measured, with the same intensity at focus, when the gas jet was located around the laser focus. In this case, besides the odd harmonic peaks, discrete and well resolved peaks are clearly visible in between. The spectral position of these peaks is pulse dependent. Increasing the CCD integration time in order to measure the harmonic spectrum corresponding to several extreme ultraviolet (EUV) pulses, broad and structureless peaks were observed between the sharp odd harmonic peaks. This last experimental observation is in agreement with previously reported measurements [12].

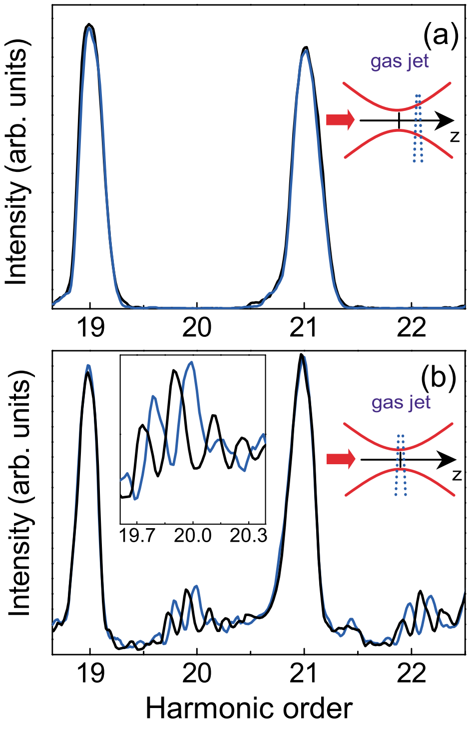

FIG. 1 (color). Measured single-shot harmonic spectra in argon generated by 20-fs driving pulses: (a) gas jet located ~2 mm after the laser focus; (b) gas jet located around the laser focus; the inset shows an enlarged view of the spectral region between two adjacent odd harmonic peaks. The black and blue curves correspond to two different driving pulses.

The experimental results can be understood in the framework of the strong-field approximation [13]. Using a nonadiabatic three-dimensional numerical propagation model [14], we have analyzed the role of CEP on the spectral characteristics of the harmonic radiation. Figure 2 shows two harmonic spectra calculated in argon assuming two different CEPs, namely, $\varphi = 0$ and $\varphi = 5\pi/8$, with the gas jet located around the laser focus. 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 parameters used in the simulations reproduce the experimental conditions. The simulations are in good agreement with the experimental results: well resolved peaks, whose position is CEP dependent, are visible between the odd harmonic peaks. When the gas jet is placed after the laser focus the calculated harmonic spectra show clean odd harmonics with no radiation between adjacent peaks, in agreement with the measurements reported in Fig. 1(a). By using the same numerical code we have investigated the effects of small fluctuations of the intensity and of the duration of the driving pulses. We have indeed calculated two sets of harmonic spectra generated by pulses with fixed CEP: the first one considering a 6% peak-to-peak intensity fluctuation of the driving pulses; the second one assuming a 10% peak-to-peak variation of the driving pulse duration (19–21 fs). It is worth pointing out that such fluctuations are much larger than the measured values. In both cases, the calculated spectra display a negligible shift of the peaks between the odd harmonics. This is a clear indication that the variation of the spectral characteristics of the EUV radiation, shown in Fig. 1(b), cannot be induced by shot-to-shot fluctuations of the intensity and of the duration of the fundamental pulses. In order to investigate more deeply the role of propagation, we have calculated the EUV spectra as a function of the propagation distance inside the gas jet. Besides an obvious increase of the EUV intensity upon increasing the gas jet thickness, the spectral shift of the peaks between the odd harmonics is almost negligible. This

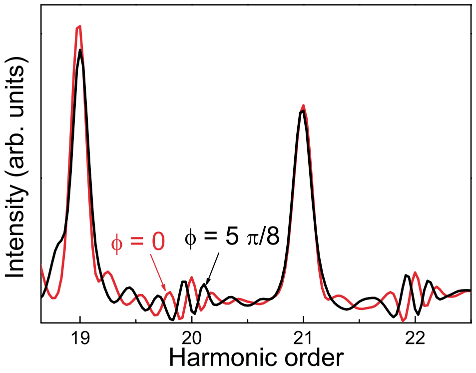

FIG. 2 (color). Harmonic spectra in argon calculated using a nonadiabatic three-dimensional numerical code for two different CEPs (pulse duration 20 fs; pulse peak intensity 2×10^{14} W/cm^2; gas jet thickness 0.2 mm; argon pressure 50 torr).

observation strongly supports the conclusion that the physical mechanisms, which determine the spectral behavior shown in Fig. 1(b), are based on the interaction of the driving pulse with a single atom. For this reason we have calculated the nonadiabatic single-atom response of the nonlinear medium.

In particular, the method of stationary phase [13,15] offers the possibility to gain a good deal of physical insight. This method requires the solution of the saddle-point equations, obtained by equating to zero the derivatives of $\phi(\mathbf{p}, t, \tau) = S(\mathbf{p}, t, \tau) - \omega t$, with respect to $\mathbf{p}$, t, and τ, where $S(\mathbf{p}, t, \tau)$ is the quasiclassical action; $\mathbf{p}$ is the canonical momentum; τ is the time spent by the electron in the continuum between the ionization instant and the recombination instant t; and ω is the angular frequency of the emitted harmonic radiation. In the calculation we consider the linearly polarized laser electric field $E(t) = E_0 \cos^2(t/\tau_0) \cos(\omega_0 t + \varphi)$. Using the stationary phase approximation, the Fourier transform of the dipole moment, $x(\omega)$, can be written as a coherent superposition of the contributions from the (complex) electron quantum paths corresponding to the (complex) saddle-point solutions $(\mathbf{p}_s, t_s, \tau_s)$ [16]. Note that for the calculation the full electric field of the laser pulse is used, so that the model is generalized to account for nonadiabatic effects. In the plateau spectral region two classes of quantum paths give the most relevant contribution to the dipole moment [17]. One class of quantum paths is characterized by a short electron return time, close to half an optical period, whereas the second one has a return time close to 1 period. The use of the saddle-point method allows one to identify the contributions from the different quantum paths. In the spirit of Feynman's path integrals [17], $x(\omega)$ can be written as follows:

$$x(\omega) = \sum_{\text{short}} C_s \exp[-i\phi(\mathbf{p}_s, t_s, \tau_s)] + \sum_{\text{long}} C_s \exp[-i\phi(\mathbf{p}_s, t_s, \tau_s)], \quad (1)$$

where $C_s = C_s(\mathbf{p}_s, t_s, \tau_s)$ is an amplitude determined by the saddle-point solutions. The first sum in Eq. (1) extends over the short quantum paths; the second sum extends over the long quantum paths. It has been demonstrated that phase matching can provide a powerful method to identify the contributions of the individual quantum paths [12,17–19]. In particular, efficient selection of the short path harmonics can be obtained when the gas jet is located after the focus of the laser beam; while, when the gas jet is located around the focus, the contribution of the long path components becomes important. So far, the role of the CEP of the driving pulses on the individual quantum paths has never been considered. The use of the saddle-point approximation allows one to point out clearly the essential physical mechanisms. Figure 3(a) shows a portion of the plateau region of the calculated single-atom harmonic spectrum in argon produced by the coherent superposition of the contributions from the short quantum paths [first sum in Eq. (1)]. The parameters used in the calculations are the following: driving pulse duration, 20 fs; pulse peak intensity, 2×10^{14} W/cm^2; argon gas. The spectrum is characterized by sharp peaks, corresponding to the odd harmonics of the fundamental radiation. Such spectrum is not affected by the CEP of the driving pulse. Figure 3(b) shows two calculated spectra produced by the long quantum paths [second sum in Eq. (1)] for two different CEPs. The spectra show several peaks, which do not correspond to harmonics of the fundamental wavelength. In this case the whole spectrum is clearly sensitive to CEP. The spectral peaks corresponding to different CEPs are frequency shifted. The

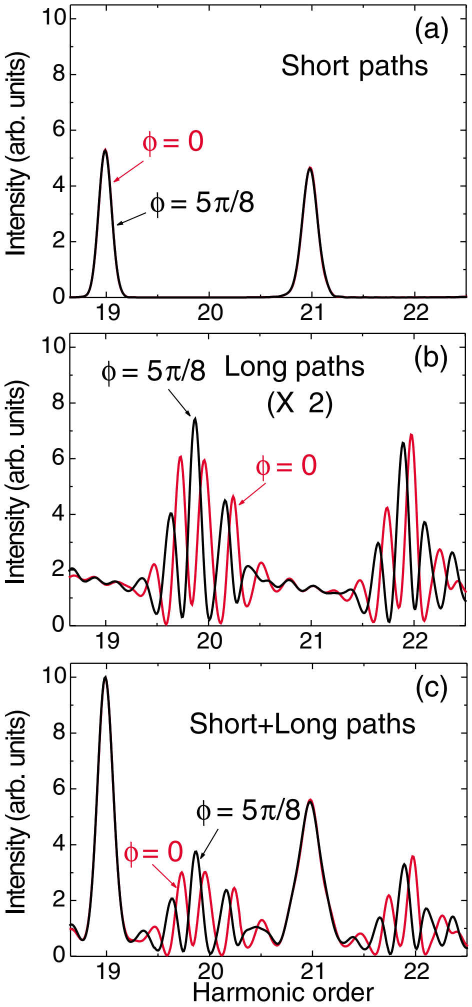

FIG. 3 (color). Portion of the plateau region of harmonic spectra calculated using the saddle-point method; the red curves correspond to $\varphi = 0$; the black curves correspond to $\varphi = 5\pi/8$. (a) Coherent superposition of the contributions from the short quantum paths. (b) Coherent superposition of the contributions from the long quantum paths; the calculated intensity has been multiplied by a factor of 2 for visual convenience. (c) Complete single-atom emission spectra produced by the coherent superposition of the contributions from the short and the long quantum paths.

complete single-atom spectrum, shown in Fig. 3(c) for two CEPs, is obtained as coherent superposition of the contributions from the short and the long quantum paths (such a spectrum does not take into account phase-matching mechanisms in the gas jet, which can increase the relative contribution from one of the two possible quantum paths). It is worth pointing out that small intensity fluctuations of the driving pulses do not significantly affect the position of the spectral peaks.

On the basis of these calculations, the interpretation of the experimental results reported in Figs. 1(a) and 1(b) is straightforward. When the gas jet is located after the laser focus [see Fig. 1(a)] the phase-matching mechanisms lead to selection of the short quantum path; since the CEP does not significantly influence such trajectories (in the plateau region), the single-shot harmonic spectra do not show significant shot-to-shot variations. On the contrary, when the gas jet is located around the laser focus, where the long quantum paths contribute significantly to the emission, the single-shot spectra show a clear CEP dependence. The spectral peaks corresponding to odd harmonics of the fundamental radiation visible in Fig. 1(b) mainly originate from the short path harmonics and, therefore, are not CEP dependent. The spectral peaks, displayed in Fig. 1(b), between adjacent odd harmonic peaks are due to contributions from the long quantum paths, which are sensitive to the CEP.

An intuitive physical interpretation of the different effect of the CEP on the short and long quantum paths is related to the different time spent by the electron in the continuum before recombination with the parent ion. In the plateau region, the electron return times $\mathrm{Re}(\tau)$ associated to the short quantum paths are even lower than half an optical period of the fundamental radiation [for the harmonics reported in Fig. 1(a) $\mathrm{Re}(\tau) \approx 0.3T$, where T is the optical period], whereas for the long paths $\mathrm{Re}(\tau) \approx T$. Since the electron trajectories are determined by the temporal evolution of the laser electric field, the long quantum paths are more sensitive to even small changes of the electric field induced by different CEPs, as a consequence of the longer time spent by the electrons in the continuum with respect to the short quantum paths.

In conclusion, we have shown, both experimentally and theoretically, that the spectral characteristics of the harmonic emission generated by light pulses in the multi-optical-cycle regime are significantly affected by the pulse carrier-envelope phase. For the first time, to our knowledge, we have demonstrated that the short and long electron quantum paths contributing to harmonic generation are influenced in a different way by the pulse carrier-envelope phase. We expect that our observations, together with the use of the single-shot measurement technique, will promote new developments in the field of coherent phase-controlled phenomena in a novel and easily accessible temporal regime.

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[1] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).

[2] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **414**, 509 (2001).

[3] G. G. Paulus, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, and S. De Silvestri, Nature (London) **414**, 182 (2001).

[4] A. Baltuška, Th. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, Ch. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch, and F. Krausz, Nature (London) **421**, 611 (2003).

[5] M. Nisoli, G. Sansone, S. Stagira, S. De Silvestri, C. Vozzi, M. Pascolini, L. Poletto, P. Villoresi, and G. Tondello, Phys. Rev. Lett. **91**, 213905 (2003).

[6] S. T. Cundiff, J. Phys. D **35**, R43 (2002).

[7] C. G. Durfee III, A. R. Rundquist, S. Backus, C. Herne, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **83**, 2187 (1999).

[8] I. P. Christov, R. Bartels, H. C. Kapteyn, and M. M. Murnane, Phys. Rev. Lett. **86**, 5458 (2001).

[9] L. Poletto, G. Tondello, and P. Villoresi, Appl. Opt. **42**, 6367 (2003).

[10] W. Kornelis, J. Biegert, J. W. G. Tisch, M. Nisoli, G. Sansone, C. Vozzi, S. De Silvestri, and U. Keller, Opt. Lett. **28**, 281 (2003).

[11] J. Biegert, W. Kornelis, G. Steinmeyer, J. W. G. Tisch, U. Keller, M. Nisoli, G. Sansone, S. Stagira, C. Vozzi, and S. De Silvestri, in *Ultrafast Phenomena XIII*, edited by R. D. Miller *et al.* (Springer-Verlag, Berlin, 2003), p. 217.

[12] D. G. Lee, H. J. Shin, Y. H. Cha, K. H. Hong, J.-H. Kim, and C. H. Nam, Phys. Rev. A **63**, 021801 (2001).

[13] M. Lewenstein, Ph. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A **49**, 2117 (1994).

[14] E. Priori, G. Cerullo, M. Nisoli, S. Stagira, S. De Silvestri, P. Villoresi, L. Poletto, P. Ceccherini, C. Altucci, R. Bruzzese, and C. de Lisio, Phys. Rev. A **61**, 063801 (2000).

[15] M. Lewenstein, P. Salières, and A. L'Huillier, Phys. Rev. A **52**, 4747 (1995).

[16] D. B. Milošević, G. G. Paulus, and W. Becker, Phys. Rev. Lett. **89**, 153001 (2002).

[17] P. Salières, B. Carré, L. Le Déroff, F. Grasbon, G. G. Paulus, H. Walther, R. Kopold, W. Becker, D. B. Milošević, A. Sanpera, and M. Lewenstein, Science **292**, 902 (2001).

[18] P. Salières, A. L'Huillier, and M. Lewenstein, Phys. Rev. Lett. **74**, 3776 (1995).

[19] M. Bellini, C. Lyngå, A. Tozzi, M. B. Gaarde, T. W. Hänsch, A. L'Huillier, and C.-G. Wahlström, Phys. Rev. Lett. **81**, 297 (1998).