Measurement of Harmonic Phase Differences by Interference of Attosecond Light Pulses

G. Sansone,[1] E. Benedetti,[1] J.-P. Caumes,[1] S. Stagira,[1] C. Vozzi,[1] M. Pascolini,[2] L. Poletto,[2] P. Villoresi,[2] S. De Silvestri,[1] and M. Nisoli[1,*]

[1]*Dipartimento di Fisica, National Laboratory for Ultrafast and Ultraintense Optical Science-INFM, Politecnico, Milano, Italy*
[2]*Laboratory for Ultraviolet and X-Ray Optical Research – INFM D.E.I. – Università di Padova, Padova, Italy*

(Received 13 January 2005; published 19 May 2005)

By using a self-referencing technique, we have experimentally measured the influence of the carrier-envelope phase of femtosecond light pulses on the phase of the electric field of the radiation produced by high-order harmonic generation. We show that, in particular experimental conditions, the temporal evolution of the electric field of the attosecond pulses, is directly controlled by the carrier-envelope phase of the driving pulses.

DOI: 10.1103/PhysRevLett.94.193903 PACS numbers: 42.65.Ky, 42.50.Hz, 42.65.Re

In the last four years it has been experimentally demonstrated that attosecond pulses can be produced by using the process of high-order harmonic generation in noble gases, as theoretically suggested in 1992 [1]. Using few-cycle driving pulses the emission process can be confined to a single optical cycle, so that single x-ray subfemtosecond pulses can be generated [2,3]. In the case of multicycle driving pulses, trains of attosecond pulses, with a discrete spectrum containing the odd harmonics of the fundamental radiation, are produced [1,4]. Recently Baltuška *et al.* [5] have demonstrated that the use of driving pulses with stable carrier-envelope phase (CEP) allows one to control the spectral distribution of the soft-x-ray radiation generated by few-cycle infrared pulses. From both a fundamental and a technical point of view it is essential to understand the CEP effects on the phase of the harmonics, on the synchronization between the attosecond pulses and the generating field, and on the electric field evolution of the attosecond pulses. In this work we will present a self-referencing technique for the measurement of the effect of the CEP of the driving pulses on the phase difference between consecutive harmonics. Such a method is based on the measurement of the beat pattern produced by interference of attosecond pulses. Moreover, by using a numerical model based on the nonadiabatic saddle-point method (NASPM) [6,7], we will demonstrate that, in particular experimental conditions, the temporal evolution of the electric field of the attosecond pulses can be directly controlled by the CEP of the driving pulses. This can be regarded as a coherent control mechanism of electronic processes, obtained by shaping a light pulse on an attosecond time scale [8].

The phase difference between consecutive harmonics can be retrieved by the technique called reconstruction of attosecond beating by interference of two-photon transition (RABITT) [4,9]. RABITT is based on the measurement of photoelectron spectra generated in noble gases ionized by a superposition of odd harmonics in the presence of the fundamental infrared (IR) light of the driving beam. The phase difference between consecutive harmonics can be determined by measuring the magnitude of the generated sidebands peaks as a function of the delay between the IR pulse and the harmonic field. Such a method, as we will show in the following, does not give access to the influence of the CEP of the driving field. Therefore, we have used a different approach. The first step is the use of a physical mechanism leading to spectral broadening of the harmonic peaks, so that a beat pattern is generated between pairs of consecutive harmonics. It is well known that the phase, $\Phi(r, z, t)$, of the harmonic field at angular frequency $q\omega_0$, where ω_0 is the angular frequency of the fundamental radiation, is given by [10] $\Phi_i(r, z, t) = -\alpha_i I(r, z, t)$, where $i = 1, 2$ refers to the short ($i = 1$) and long ($i = 2$) quantum paths contributing to the qth harmonic, $I(r, z, t)$ is the intensity profile of the driving field, and α_i is the slope of the phase as a function of intensity. The temporal variation of the driving pulse intensity leads to a variation of the instantaneous frequency (harmonic chirp) $\Delta\omega_i = -\partial\Phi_i/\partial t$, which broadens the spectrum of each individual harmonic. It has been demonstrated, both theoretically [10] and experimentally [11], that the long quantum paths lead to a larger chirp and, therefore, a broader spectrum than the short paths. We recall that phase matching allows one to select the contributions from the different quantum paths [12]: when the gas jet, where harmonics are generated, is placed after the laser focus, the short quantum paths are efficiently selected, while positioning the gas jet around the laser focus leads to an increase of the contribution of the long paths. The achievable spectral broadening can be larger than the frequency separation between two consecutive odd harmonics ($\Delta\omega \geq 2\omega_0$). This process can be clearly understood using Fig. 1(a), which shows the calculated instantaneous frequency for two consecutive harmonics (q and $q + 2$) as function of time for $\Delta\omega > 2\omega_0$. The shaded area in Fig. 1(a) corresponds to the spectral region where the high-frequency side of the qth-harmonic spectrum, generated on the leading edge of the driving pulse, overlaps the low-frequency side of the spectrum of $(q + 2)$th harmonic, generated on the pulse trailing edge. The spectral superposition of temporally delayed compo-

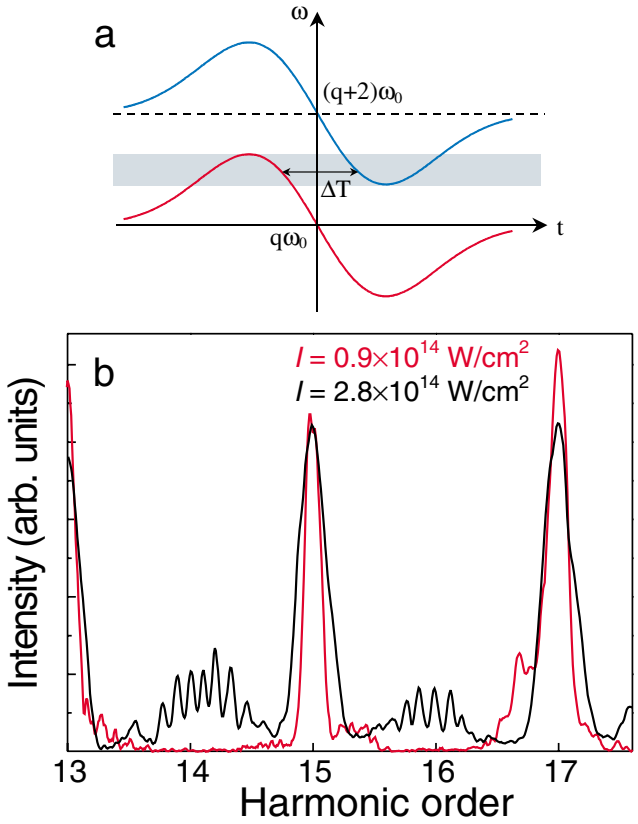

FIG. 1 (color). (a) Frequency chirp of two consecutive odd harmonics, calculated as explained in the text. The shaded area corresponds to the interference spectral region. (b) Normalized measured harmonic spectra generated in argon by 35 fs pulses, with stable CEP, at two peak intensities.

nents [delay ΔT in Fig. 1(a)] gives rise to an interference effect, which can be experimentally measured. Here we assume that the effects of ionization can be neglected, as in our measurements. It is worth mentioning that, since the harmonic emission is in general confined to the central part of the light pulse, only the two branches separated in time by ΔT [see Fig. 1(a)] should be considered. Note that, assuming a constant spectral broadening $\Delta\omega$, the delay ΔT turns out to be proportional to the duration, τ, of the driving pulse. At this point, using the algorithm of Fourier transform spectral interferometry (FTSI) [13], it is possible to retrieve from the interference pattern the phase difference between all the consecutive harmonics in the overlapping region.

In the experiments, harmonic radiation has been produced focusing femtosecond light pulses, generated by a Ti:sapphire laser system (800 nm central wavelength, 1 kHz repetition rate), into an argon jet. The CEP of the light pulses has been stabilized using an experimental setup similar to that described by Baltuška *et al.* [5]. Such a phase can be finely adjusted by introducing in the beam path a glass plate with variable thickness. At 800 nm a 2π CEP variation is induced by the addition of 52 μm of fused silica without affecting the pulse duration. We have used a pair of wedges mounted on a stepper motor. Figure 1(b) shows harmonic spectra generated by 35 fs light pulses at two intensities; the gas jet was located around the laser focus in order to increase the contributions of the long quantum paths. As expected, upon increasing the intensity, the harmonic peaks broaden and eventually overlap in the spectral region between consecutive odd harmonics, where distinct spectral peaks are formed. Without CEP stabilization the peaks between the odd harmonics are completely smeared out, and a featureless structure is measured [14]. At low intensity well-resolved odd harmonics of the fundamental radiation were observed, without any evidence of spectral structures in between. This is due to the fact that, in this case, the intensity-dependent spectral broadening is not large enough to cause an overlap between two consecutive harmonics, so that no interference is possible. To provide additional evidence for the origin of the measured spectral structures between the odd harmonics, we have investigated the effect of the driving pulse duration. Harmonic spectra generated in argon have been measured increasing the laser pulse duration, τ, from 20 fs up to 43 fs; pulse intensity has been chosen in order to obtain the same spectral broadening. The driving pulses were transform limited. The temporal delay, ΔT, between the interfering field components, calculated by taking the Fourier transform of the measured interference patterns, increases almost linearly with pulse duration from 11 fs to 23 fs. These results confirm our physical interpretation and are well reproduced by a nonadiabatic three-dimensional propagation model [15]. All our experimental results drive to the conclusion that the spectral structures between odd harmonics originate from a beat between two extreme ultraviolet (XUV) fields, emitted during two different temporal portions of the fundamental light pulse, separated by about one-half the pulse duration, the first on the leading edge and the second on the trailing edge of the driving pulse.

We have then varied the CEP, ψ, of the driving pulses moving the glass wedges. Figure 2 shows the portion between the 13th and 15th harmonics of nine generated harmonic spectra for different amounts δz of glass in the beam path, which correspond to different CEP values of the driving pulses in a range of $\pi(\psi_0 < \psi < \psi_0 + \pi)$. Each horizontal line represents a XUV spectrum measured at a fixed CEP. As clearly visible in Fig. 2, the spectral positions of the interference peaks continuously shift by changing the CEP of the driving pulses. We found that the beat pattern periodically changes for a CEP variation $\Delta\psi = \pi$. The same spectral behavior was observed for all the other pairs of consecutive odd harmonics. The experimental results are well reproduced by simulations based on the nonadiabatic three-dimensional propagation model. We have then analyzed the interference patterns measured at different CEPs using the FTSI algorithm [13]. The interference between two consecutive harmonics (q and $q + 2$), spectrally broadened due to the intensity dependence of their phases, can be written as follows:

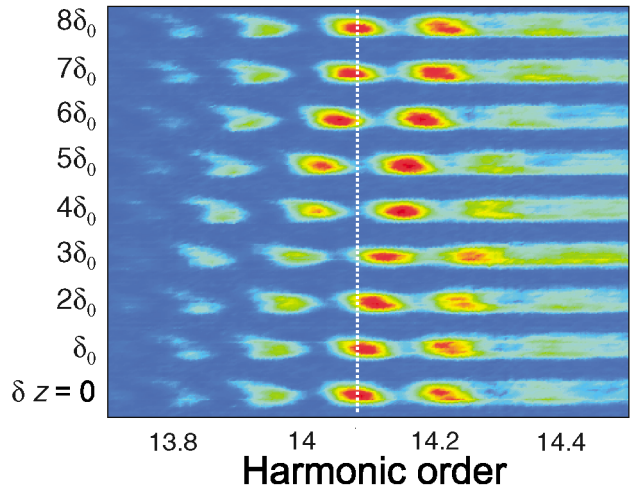

FIG. 2 (color). Portion of the harmonic spectra between 13th and 15th harmonics generated in argon by 32 fs pulses, for different amounts δz of glass in the beam path ($\delta_0 = 3.42\ \mu$m). The vertical dashed white line is a guide for the eye.

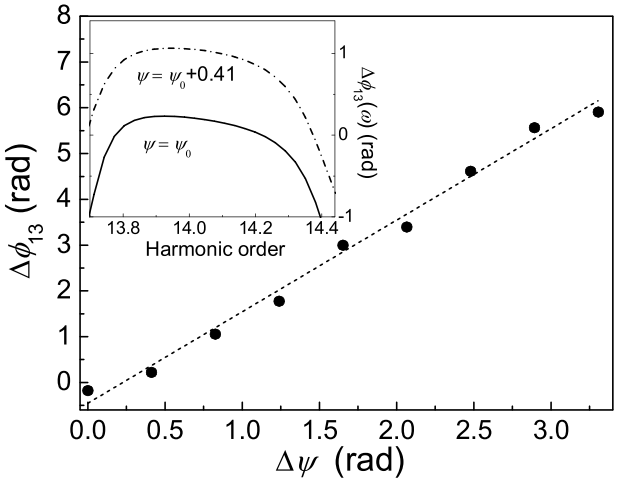

FIG. 3. $\Delta\phi_{13}(\omega = 14\omega_0)$ as a function of the CEP variation $\Delta\psi = (\pi/26)\delta z[\mu\text{m}]$ calculated from the experimental spectra reported in Fig. 2; the dashed line is a linear fit $\Delta\phi_{13} = 2\Delta\psi - 0.46$. Inset: retrieved $\Delta\phi_{13}(\omega)$ for two CEP values, corresponding to two consecutive harmonic spectra of Fig. 2.

$$S(\omega) = I_q(\omega) + I_{q+2}(\omega) + 2\sqrt{I_q(\omega)I_{q+2}(\omega)}\cos[\phi_{q+2}(\omega) - \phi_q(\omega) + \omega\Delta T], \quad (1)$$

where $I_q(\omega)$ is the intensity of qth harmonic, $\phi_q(\omega)$ is the harmonic phase, and ΔT is the time delay between the two interfering components. For a sufficiently large temporal delay ΔT, the argument of the cosine in Eq. (1) can be retrieved using the algorithm of FTSI. This algorithm can be successfully implemented since $I_q(\omega)$, $I_{q+2}(\omega)$, and $\Delta\phi_q(\omega) = \phi_{q+2}(\omega) - \phi_q(\omega)$ vary slowly compared to the linear term $\omega\Delta T$. It is thus possible to precisely determine the delay ΔT and the phase difference $\Delta\phi_q(\omega)$. Applying the FTSI to the experimental results reported in Fig. 2, we found that the delay ΔT is not appreciably affected by the CEP of the driving pulses. The inset of Fig. 3 shows the retrieved $\Delta\phi_{13}(\omega)$ for two CEP values: a CEP variation $\Delta\psi$ determines a $2\Delta\psi$ variation of $\Delta\phi_{13}(\omega)$. This is clearly shown in Fig. 3, which displays $\Delta\phi_{13}(\omega = 14\omega_0)$ as a function of $\Delta\psi$: the experimental points can be well fitted by a line (dashed line in Fig. 3) with slope 2. The same results are valid for all the other pairs of consecutive harmonics. These experimental results support the conclusion that the phase of the harmonic field is related to the CEP of the driving pulses by the following expression: $\phi_q(\omega) = \theta_q(\omega) + q\psi$, where $\theta_q(\omega)$ does not depend on ψ. This conclusion and the retrieved behavior of $\Delta\phi_q(\omega)$ are also confirmed by the results of numerical simulations based on the use of the nonadiabatic saddle-point method. The calculations demonstrate that the validity of the previous conclusion is not limited to the case of spectrally broadened harmonics generated by the contributions of the long quantum paths, but that it can be extended also to the contributions of the short paths.

Note that from a RABITT measurement it is not possible to retrieve the real phase difference between consecutive harmonics, since the dependence on the CEP of the IR light pulse is canceled. In fact, the amplitude of the sideband is modulated by the interference term $S = A\cos(2\varphi_{\text{IR}} + \phi_q - \phi_{q+2} + \Delta\phi_{\text{at}})$, where A and $\Delta\phi_{\text{at}}$ are atomic amplitude and phase, respectively, $\varphi_{\text{IR}} = \omega_0 T + \psi$, and T is the delay between the IR and the XUV pulses. Assuming $\phi_q(\omega) = \theta_q(\omega) + q\psi$, we obtain $S = A\cos(2\omega_0 T + \theta_q - \theta_{q+2} + \Delta\phi_{\text{at}})$; therefore, from the measurement of the sideband modulation it is possible to determine $\theta_q - \theta_{q+2}$: the influence of the CEP of the IR pulse is canceled. Note that this does not invalidate the reconstruction of the temporal intensity profile of the attosecond pulse train in the case of well-separated harmonics, since the measured CEP influence on the harmonic phase does not induce changes in the electric field evolution of the attosecond pulses apart from a small amplitude variation.

So far we have limited our attention to the frequency domain. To gain a good deal of physical insight we have to consider the temporal structure of the emitted XUV radiation. The nonadiabatic saddle-point method [7] allows one to obtain a clear physical picture of the process. Since the measured beat pattern is due to the long quantum paths, we have calculated the spectrum produced by the coherent superposition of the long path contributions. By taking the inverse Fourier transform it is possible to calculate the corresponding temporal structure. A train of attosecond pulses, separated by one-half the optical period is obtained, as shown by the dashed curve in Fig. 4(a). In the calculated spectrum we have then selected the beat pattern by proper spectral windows (width $\Delta\omega = 0.4\omega_0$) centered in the regions between consecutive odd harmonics. The associated temporal structure [blue curve in Fig. 4(a)] corre-

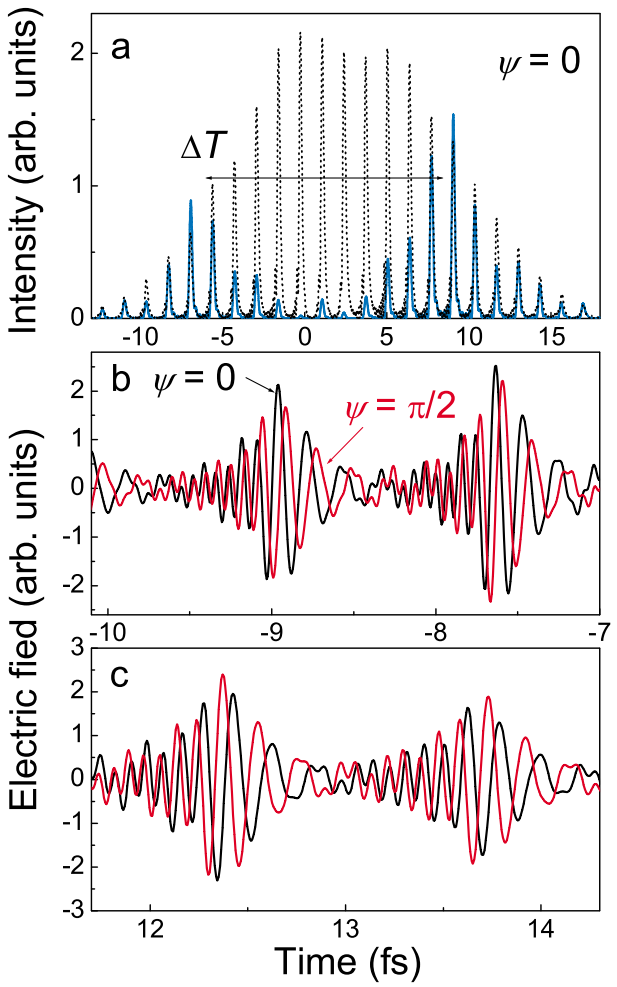

FIG. 4 (color). (a) Dashed curve: intensity profile of the attosecond pulse train generated by the long paths, calculated by using the NASPM; solid blue curve: calculated temporal structure associated to the interference pattern generated by the long paths. Calculated electric field of the attosecond pulses generated on the leading edge (b) and on the trailing edge (c) of the driving pulse, for two values of the CEP, ψ, of the IR pulse. Driving pulse duration 30 fs; peak intensity $I = 2.6 \times 10^{14}$ W/cm^2.

sponds to two trains of attosecond pulses, located one on the leading and the other on the trailing edge of the complete train, separated in time by about one-half the duration of the total train of attosecond pulses. This is the picture, in the temporal domain, of the interference effect previously described in the frequency domain: the spectral structures between the odd harmonics originate from the interference between the electric fields of such two groups of attosecond pulses. In addition, the temporal analysis offers a simple explanation of the physical origin of the carrier-envelope phase dependence of the interference pattern. We have calculated the temporal structures associated to the interference patterns for several values of CEP of the driving pulses. We found that, in agreement with our experimental results, the temporal delay, ΔT, between the interfering fields is not significantly affected by the CEP of the IR pulse. Therefore, we can conclude that the CEP dependence of the beat pattern is related to the temporal evolution of the electric field of the interfering attosecond pulses, which turns out to be directly influenced by ψ. Indeed, Figs. 4(b) and 4(c) show two different portions of the attosecond pulse train calculated for two values of ψ (the two trains have been temporally shifted to overlap the intensity envelopes of the attosecond pulses; the same shift determines the overlap of the electric fields of the corresponding IR pulses). The temporal evolution of the electric field of the attosecond pulses generated on the leading and trailing edges of the IR pulse is significantly influenced by ψ, while around the peak of the driving pulse the attosecond pulses (not shown in Fig. 4) are not influenced by ψ. We have then calculated the temporal evolution of the electric field of the attosecond pulses generated by the long quantum path at low laser intensity ($\Delta\omega < 2\omega_0$) and by the short paths. In these cases the temporal evolution of the electric field of the attosecond pulses is not significantly influenced by the CEP of the driving pulses.

In conclusion, using a self-referencing technique based on the interference of attosecond light pulses, we have measured the phase difference between consecutive harmonics in the spectral region where they overlap. This phase difference directly depends on the CEP of the driving pulses. The role of CEP on the phase of the individual harmonics has been determined. By using a numerical model based on the nonadiabatic saddle-point method, we have shown how CEP influences the temporal evolution of the electric field of the attosecond pulses.

This work was partially supported by European Community under project MRTN-CT-2003-505138 (XTRA) and by the national FISR project "SPARC."

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

[1] G. Farkas and C. Toth, Phys. Lett. A **168**, 447 (1992).
[2] I. P. Christov, M. M. Murnane, and H. Kapteyn, Phys. Rev. Lett. **78**, 1251 (1997).
[3] R. Kienberger *et al.*, Nature (London) **427**, 817 (2004).
[4] P. M. Paul *et al.*, Science **292**, 1689 (2001).
[5] A. Baltuška *et al.*, Nature (London) **421**, 611 (2003).
[6] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[7] G. Sansone *et al.*, Phys. Rev. A **70**, 013411 (2004).
[8] I. P. Christov *et al.*, Phys. Rev. Lett. **86**, 5458 (2001).
[9] P. Agostini and L. F. DiMauro, Rep. Prog. Phys. **67**, 813 (2004).
[10] M. B. Gaarde *et al.*, Phys. Rev. A **59**, 1367 (1999).
[11] P. Salières *et al.*, Science **292**, 902 (2001).
[12] P. Salières, A. L'Huillier, and M. Lewenstein, Phys. Rev. Lett. **74**, 3776 (1995).
[13] M. Takeda, H. Ina, and S. Kobayashi, J. Opt. Soc. Am. **72**, 156 (1982).
[14] G. Sansone *et al.*, Phys. Rev. Lett. **92**, 113904 (2004).
[15] E. Priori *et al.*, Phys. Rev. A **61**, 63801 (2000).