Interferometric Autocorrelation of an Attosecond Pulse Train in the Single-Cycle Regime

Yasuo Nabekawa,[1,*] Toshihiko Shimizu,[1] Tomoya Okino,[2,1] Kentaro Furusawa,[1] Hirokazu Hasegawa,[1] Kaoru Yamanouchi,[2,1] and Katsumi Midorikawa[1]

[1]*Laser Technology Laboratory, RIKEN, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan*

[2]*Department of Chemistry, School of Science, The University of Tokyo, 7-3-1 Hongo Bunkyo-ku, Tokyo 113-0033, Japan*

(Received 19 May 2006; published 13 October 2006)

We report on the direct observation of the phase locking of the attosecond pulse train (APT) via interferometric autocorrelation in the extreme ultraviolet region. APT is formed with Fourier synthesis of high-order harmonic fields of a femtosecond laser pulse. Time-of-flight mass spectra of N^+, resulting from the Coulomb explosion of N_2 absorbing two photons of APT, efficiently yield correlated signals of APT. The measured autocorrelation trace exhibits that the duration of the pulse should be only 1.3 periods of the extreme ultraviolet carrier frequency. A few interference fringes within the short pulse duration clearly show two types of symmetry, which ensure the phase locking between pulses in APT.

DOI: 10.1103/PhysRevLett.97.153904 PACS numbers: 42.65.Re, 33.80.Rv, 42.65.Ky

The phase locking of an optical pulse train has aroused great interest in recent years because it has become a key issue in ultrafast optical science [1,2] and frequency metrology [3–7]. Because the spectra of high-order harmonic fields in the wavelength range of extreme ultraviolet (XUV) generated with an intense femtosecond laser pulse intrinsically exhibit a comblike structure, these harmonic fields form a train of extremely short bunches of an optical field, which is conventionally called an attosecond pulse train (APT) [8–11].

APT has a unique feature that cannot be seen in an isolated attosecond pulse [2,12]; this feature concerns a phase relationship in the pulse sequence. The importance of this feature can be recognized by comparing the positive frequency part of the optical field of APT, $E^{(+)}(t)$, to that of a mode-locked and frequency offset (ν_{FO}) -stabilized laser, which is now the established instrument for connecting optical frequency to radio frequency in order to define the frequency standard [3,5]. We can approximately express $E^{(+)}(t)$ as [8]

$$E^{(+)}(t) = \sum_{n=n_1}^{n_2} A_{2n+1} e^{-2\pi i(2n+1)\nu_f t - i\phi_{2n+1}}, \qquad (1)$$

where $(2n + 1)$ corresponds to the harmonic order, and A_{2n+1} and ϕ_{2n+1} are, respectively, the field amplitude and the phase of the $2n + 1$th harmonic field. The frequency of the fundamental laser field is denoted as ν_f. We neglect the variation of the phase and amplitude depending on that of the intensity of the driving laser field [13], because we restrict ourselves to describing APT within a range much shorter than the pulse duration of the driving laser field.

This equation is the same as that which describes the optical field of the mode-locked laser, of which the repetition frequency is $2\nu_f$ and ν_{FO} is set to half the repetition frequency ν_f, so that the time translation symmetries, namely, $E^{(+)}(t + qT_f) = E^{(+)}(t)$ and $E^{(+)}(t + (q + 1/2)T_f) = e^{-i\pi}E^{(+)}(t) = -E^{(+)}(t)$ $(q = \pm 1, \pm 2, \ldots)$, with respect to T_f ($\equiv 1/\nu_f$) are common to both optical fields. Although the locking property of the phase in a mode-locked laser was demonstrated with the interferometric correlation signal of two-photon absorption in a photodiode [5], similar properties of the phase in the APT have not been revealed yet because of some intrinsic and technical difficulties due to the specific features of XUV light: (i) The shorter wavelength requires the higher precision of the optical elements in an interferometer; (ii) the pulse duration should be short enough to determine the relative phase of the interference fringes to the correlation envelope; (iii) it is very difficult to observe a nonlinear phenomenon that can be utilized for correlation measurement in the XUV region.

Our recent studies to generate intense high-order harmonic fields [14,15], however, make it possible to overcome the third issue. We reported on the measurement of the pulse envelope of the 27th-order harmonic field [16,17] and that of APT formed with three harmonic fields [11] by autocorrelation. In addition, the measured pulse duration of APT satisfied the second condition mentioned above.

In this Letter, we describe the demonstration of phase locking in APT observed by interferometric autocorrelation (IAC). The detected signals of the atomic fragment ion of singly charged nitrogen (N^+) with two-photon absorption of the harmonic fields exhibit interferometric fringes on the correlation trace of the envelope. We can clearly see the locked phase in the fringes on the top of the pulse envelope due to the short pulse duration nearly attaining a single-cycle period of the carrier frequency of the principal harmonic field.

The setup for the experiment is similar to that reported in previous experiments [11,16,17] other than that we utilize N_2 molecules with two-photon absorption of high-order harmonic fields at orders from 9th to 19th, of which the relative intensities are measured to be $1 : 0.67 : 0.33 : 2.1 \times 10^{-2} : 3.9 \times 10^{-2} : 8.4 \times 10^{-3}$. In order to generate these harmonic fields, a driving laser pulse with an energy of 13 mJ and a duration of 40 fs, which is delivered from a

chirped pulse amplification system of a Ti:sapphire laser, is focused in a 10-cm-long gas cell filled with Xe [14,15]. The loosely focused laser pulse with a focal length of 5 m enables us to obtain harmonic fields with an energy range of 10 μJ under the phase-matching condition. The intensity of the driving laser field near the focal region is estimated to be $\sim 10^{14}$ W/cm^2, and that of the 11th-order harmonic field at the focal point is estimated be 3×10^{14} W/cm^2 [11,14,18,19]. We expected from the results of a separate experiment [20] that the cross sections of diatomic molecules for absorbing two photons of XUV light are generally larger than those of single atoms having a similar ionization potential to diatomic molecules. This is the reason we adopted N_2, instead of the argon atom (Ar) used in the previous experiment [11], as a nonlinear medium.

The broad range of octave spanning harmonic modes can be achieved without using any metallic filter, which is commonly used for removing a fundamental laser field. The smaller aperture (2 mm diameter) and the longer focal length (100 mm) than those adopted in the previous experiment [11] sufficiently reduce the intensity of the residual fundamental laser field reflected by the beam separator [18] such that there is no nonlinear interaction with N_2. In fact, we did not observe the signal of the two-color above-threshold ionization [21] of Ar, whose ionization potential is comparable to that of N_2, in a similar experiment using electron spectroscopy.

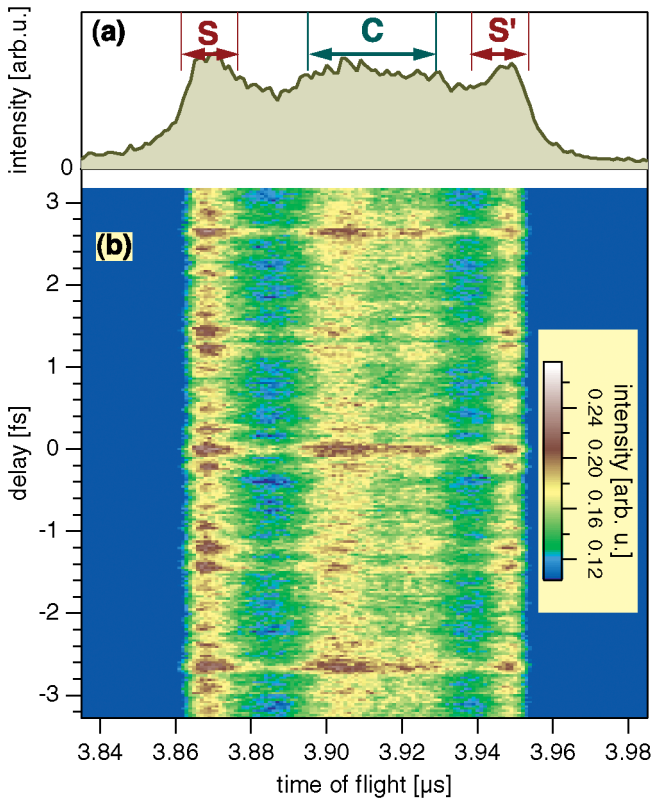

FIG. 1 (color). Time-of-flight mass spectrum at $m/z = 14$. (a) Typical spectrum of ions from N_2 with two-photon absorption of high-order harmonic fields. (b) Variation of spectrum by translating delay between two replicas of harmonic fields. Bunches with a period of 1.33 fs and fringes with a much shorter period reveal the interferometric autocorrelation of harmonic fields. The color scale of the intensity is shown in the inset.

A beam of N_2 was introduced through a skimmer from a pulsed gas valve. Ions from N_2 yielded at the focus of harmonic fields are accelerated with two stages of static electric fields between three plates of electrodes to minimize the aberration of the ion spectrum or, more specifically, to satisfy the Wiley-Mclaren condition for the time of flight (TOF) of ions. The flight length of the ions to an ion detector (microchannel plate) was 50 cm. The ambient pressure of the TOF chamber is approximately 10^{-6} Torr. The somewhat poor vacuum of the chamber is due to stray Xe gas from the chamber for harmonic generation.

A typical time-of-flight mass spectrum at $m/z = 14$ is shown in Fig. 1(a). The side peak denoted S and S' in this figure should be assigned to the fragment ion of N^+ because the kinetic energy of this fragmentary ion is estimated to be 5 eV, which corresponds to the release of the potential energy of Coulomb force stored in a doubly charged ion of N_2 (N_2^{2+}). Thus, these peaks verify that there is, at least, an ionization pathway via a dissociative potential of N_2^{2+}. By considering the simulated potential curves for N_2 and related ions [22,23], we can expect that two photons are needed for this ionization pathway. The sum of the harmonic orders including these two photons should be larger than or equal to 28 ($=9 + 19$, $11 + 17$, and so on). It is not easy to identify the contents of the central peak denoted C in Fig. 1(a) because there are two possible ion species and several ionization pathways for these ion yields. Details of these ionization processes are beyond the scope of this Letter and will be discussed elsewhere.

According to our previous research on the autocorrelation of high-order harmonic fields in the XUV [11] or soft-x-ray region [16,17], TOF mass spectra at $m/z = 14$ were recorded at each delay time between the two replicas of the high-order harmonic field, which resulted from the spatial division of the harmonic field with two harmonic beam separators. The two-dimensional image plot in Fig. 1(b) is the resultant TOF mass spectrum. We acquired and averaged the TOF mass spectrum yielded with 25 shots of a fundamental laser pulse and scanned the delay every 27 attoseconds from -3 to 3 fs. The scanning was repeated 40 times, and the accumulated 2D data of these repeated scannings resulted in Fig. 1(b).

We can see clear bunches, on both the sides (S, S') and the center (C) of the spectrum, at every 1.33 fs, which are proof of the attosecond pulse train formed with the synthesized harmonic field. These bunches also show that there exists a nonlinear interaction related to these ion yields, because the period of the train is exactly the same as the half period of the optical cycle of the fundamental laser field T_f ($=2.67$ fs) and the linear interaction cannot provide the correlated signal of the pulse envelope.

A remarkable feature of the fringes along the delay axis in the entire image, however, may be more attractive than the attosecond pulse train itself. We plot the averaged trace within S and S' in Fig. 2(a) to see the characteristic of the fringe.

First, we verified that the fringe originated from the interference between two replicas of the harmonic field by extracting the principal frequency of the fringe by Fourier transformation. The magnitude square of the Fourier amplitude of the trace in Fig. 2(a) reveals some distinct peaks as shown in Fig. 2(b). Since the frequency is normalized with the carrier frequency of the fundamental laser field, peaks at 9, 11, and 13 should be assigned to originate from the interference fringe of the 9th, 11th, and 13th harmonic fields, respectively, while peaks at even numbers of 2 and 4 should correspond to the difference frequency between the adjacent and the next adjacent harmonic field modes to form the envelope of the train. The highest peak at $11\nu_f$, which may seem inconsistent with the relative intensities of the harmonic fields, is mainly due to the restriction of the harmonic-order pair that induces a two-photon Coulomb explosion of N_2.

We note that the interference in the time domain itself is worth observing in optical science, because we cannot straightforwardly construct the Michelson or Mach-Zender interferometer for XUV light due to the absence of a partial mirror without dispersion in this wavelength range.

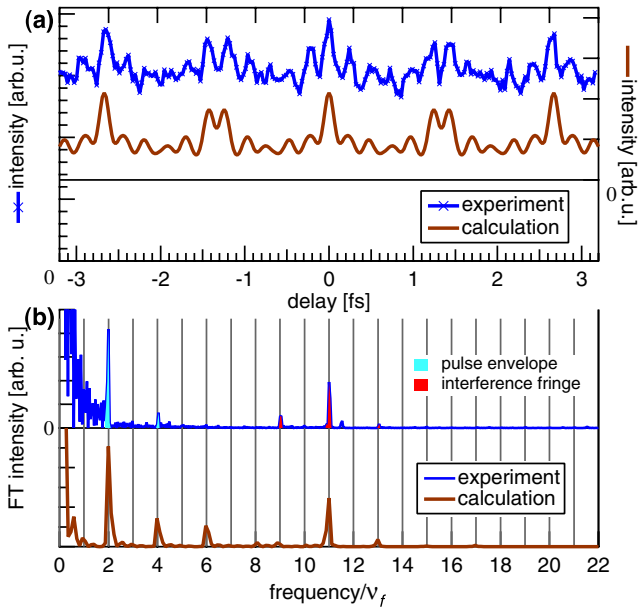

FIG. 2 (color online). Interferometric autocorrelation trace and its Fourier analysis. (a) A trace of the dark-blue solid line with crossing markers is derived by averaging data in Fig. 1(a) in S and S'. (b) Square magnitude of the Fourier amplitude of the dark-blue upper trace in (a) [dark-blue curve in the upper part of (b)]. The light-blue peaks at frequencies of $2\nu_f$ and $4\nu_f$ are assigned to modes of the pulse envelope, while the red peaks at $9\nu_f$, $11\nu_f$, and $13\nu_f$ emerge from interference fringes. The calculated correlation trace and its Fourier transform are shown as brown solid curves in the lower parts of both figures, respectively.

Second, we found that two kinds of symmetry of the fringe to the pulse envelope clearly appear in the interferometric autocorrelation trace in Fig. 2(a). One is a symmetry having the peak of the fringe on the top of the pulse envelope of delays at -2.67, 0, and 2.67 fs, and another is that having the bottom of the fringe on the top of the pulse envelope at delays of -1.33 and 1.33 fs. Although the trace in Fig. 2(a) arises from a second-order nonlinear interaction, the π-flipped nature of the phase in the fringe can be qualitatively explained by considering that the linear interference between two harmonic fields delayed each other, because the primary part of the fringe frequency in the interferometric autocorrelation should not coincide with the sum frequency but with the frequency itself of the measured field, as is shown in Fig. 2(b). The symmetry of the fringes in the second-order IAC trace of the APT will be discussed elsewhere [24]. The intensity of the linear interference $I_q(\tau)$ between the optical field $E^{(+)}(t)$ and the replica of $E^{(+)}(t)$ that separated with qT_f ($q = \pm 1, \pm 2, \ldots$) is expressed as

$$I_q(\tau) \propto \int_{-T_f/2}^{T_f/2} dt |E^{(+)}(t) + E^{(+)}(t + qT_f - \tau)|^2 = 2 \sum_{n=n_1}^{n_2} |A_{2n+1}|^2 [1 + \cos\{2\pi(2n+1)\nu_f \tau\}], \quad (2)$$

where we used Eq. (1). On the other hand, we have

$$I_{q+(1/2)}(\tau) \propto 2 \sum_{n=n_1}^{n_2} |A_{2n+1}|^2 [1 - \cos\{2\pi(2n+1)\nu_f \tau\}] \quad (3)$$

for the interference to the optical field at the half integer of T_f. The sign exchange in front of the cosine function in Eqs. (2) and (3) should be superposed to the pulse envelope of the autocorrelation trace, resulting in peaks or dips on the top of the pulse envelope. In addition, the similarity of the fringe at ± 2.67 fs to that at 0 fs of the measured trace ensures that there is no notable phase slip, at least within the time range of $\pm T_f$. Thus, we conclude that the symmetries of the fringe on the interferometric autocorrelation derived in this experiment directly prove the antiphase of the pulse to the next pulse in the attosecond pulse train formed with the high-order harmonic fields. We should mention, to avoid misunderstanding about the phase, that the absolute phase of the APT is still uncertain from the IAC trace, while we have confirmed that the pulse-to-pulse relationship of the phase is locked.

The π-flipped phase or the sign exchange of the optical pulse to that of the next pulse is not merely a mathematical property of Eqs. (2) and (3). The three step model of the high-order harmonic generation [25,26] tells us that an electron ionized from an atom at a certain phase of the fundamental laser field returns to the atom accompanied by

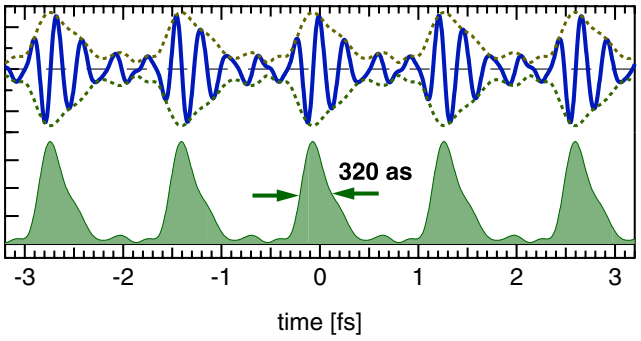

FIG. 3 (color online). Estimated intensity profile of the attosecond pulse train (dark-green curve with hatched area) and the optical field (dark-blue curve).

the emission of a pulse of a high-order harmonic field every half-cycle period. In this model, the trajectory of the electron, moving along the direction of the electric field of the fundamental laser, determines the phase of the emitted harmonic field. Therefore, our finding of the π-flipped phase in the attosecond pulse train verifies that harmonic pulses are certainly from electrons detached from opposite sides at every half-cycle period of the fundamental laser field.

Note that the clear observation of phase locking in the actual experiment is due to the small number of interference fringes emerging from near single-cycle pulses in the APT. We estimated the duration of the pulse by comparing the calculated results of the interferometric autocorrelation of the harmonic fields with group-delay dispersion (GDD) and its Fourier transform to the measured results. Details of this calculation will be reported elsewhere. We plot the calculated autocorrelation trace and its magnitude square of Fourier amplitude as solid blue curves in Figs. 2(a) and 2(b), respectively. Setting the GDD to 1.3×10^{-32} [s^2], which is the measured numeric value with the separate experiment [11], there is fairly good agreement between the experimental observation and the calculated result of both autocorrelation and Fourier transform. The increased GDD to 2.0×10^{-32} [s^2] reduced the height of the envelope with the remaining interference fringe in the autocorrelation so that the intensity at $2\nu_f$ in the Fourier transform is almost equal to that at $11\nu_f$. Therefore, 1.3×10^{-32} [s^2] is the reasonable GDD in practice, although we might take into account the intrinsic response of N_2 to two-photon absorption, as was mentioned in Refs. [27,28] for rare-gas atoms, in order to determine GDD in more detail.

The envelope and optical field of the attosecond pulse train, evaluated from the measured intensity ratio of the harmonic fields from the 9th to 19th orders and the estimated GDD, are shown in Fig. 3. The duration of the pulse envelope is 320 attoseconds in full width at half maximum, which corresponds to only an $\sim$1.3-cycle period of the principal carrier frequency of the 11th harmonic field. Thus, we can clearly see the relative phase of interference fringes to pulse envelopes.

In conclusion, we have shown clear evidence of the locked phase in an attosecond pulse train by interferometric autocorrelation. The detection of fringes that emerged from the interference of a pulse sequence in the XUV range enables us to determine the phase difference between pulses, so that we would obtain the instantaneous shape of the optical field in the attosecond pulse train by combining this result with those obtained by other techniques for measuring the chirp [9,11,29] and by using a driving laser field whose absolute phase is stabilized [1,2]. Thus, an optical synthesizer of XUV light comes into the realm of reality.

We thank Dr. E. J. Takahashi at RIKEN for his support in generating intense harmonic fields. This study was financially supported by MEXT through a Grant-in-Aid for Scientific Research on Priority Areas, for Young Scientists (A) and for Young Scientists (B).

*Electronic address: nabekawa@riken.jp

[1] A. Baltuška *et al.*, Nature (London) **421**, 611 (2003).
[2] R. Kienberger *et al.*, Nature (London) **427**, 817 (2004).
[3] J. Reichelt *et al.*, Opt. Commun. **172**, 59 (1999).
[4] H. R. Telle *et al.*, Appl. Phys. B **69**, 327 (1999).
[5] D. H. Jones *et al.*, Science **288**, 635 (2000).
[6] C. Gohle *et al.*, Nature (London) **436**, 234 (2005).
[7] R. J. Jones *et al.*, Phys. Rev. Lett. **94**, 193201 (2005).
[8] P. Antoine *et al.*, Phys. Rev. Lett. **77**, 1234 (1996).
[9] P. M. Paul *et al.*, Science **292**, 1689 (2001).
[10] P. Tzallas *et al.*, Nature (London) **426**, 267 (2003).
[11] Y. Nabekawa *et al.*, Phys. Rev. Lett. **96**, 083901 (2006).
[12] T. Sekikawa *et al.*, Nature (London) **432**, 605 (2004).
[13] K. Varjú *et al.*, Phys. Rev. Lett. **95**, 243901 (2005).
[14] E. J. Takahashi *et al.*, Opt. Lett. **27**, 1920 (2002).
[15] E. Takahashi *et al.*, Phys. Rev. A **68**, 023808 (2003).
[16] Y. Nabekawa *et al.*, Phys. Rev. Lett. **94**, 043001 (2005).
[17] H. Hasegawa *et al.*, Laser Phys. **15**, 812 (2005).
[18] E. J. Takahashi *et al.*, Opt. Lett. **29**, 507 (2004).
[19] E. J. Takahashi *et al.*, IEEE J. Sel. Top. Quantum Electron. **10**, 1315 (2004).
[20] K. Hoshina *et al.*, J. Phys. B **39**, 813 (2006).
[21] T. E. Glover *et al.*, Phys. Rev. Lett. **76**, 2468 (1996).
[22] E. W. Thulstrup and A. Andersen, J. Phys. B **8**, 965 (1975).
[23] R. W. Wetmore and R. K. Boyd, J. Phys. Chem. **90**, 5540 (1986).
[24] In fact, the fringes corresponding to the carrier frequencies of the harmonic fields in the IAC trace should originate from the term $\tilde{S}^{(+)}(\Omega_{2(N+1)};0)\tilde{S}^{(+)^*}(\Omega_{2(N+1)};\tau)$, where the mode-resolved correlation amplitude $\tilde{S}^{(+)}(\Omega_{2(N+1)};\tau)$ was defined as Eq. (2) in Ref. [11]. The factor $e^{i(2n+1)\omega_f\tau}$ in $\tilde{S}^{(+)}(\Omega_{2(N+1)};\tau)$ necessarily yields the odd parity of the fringes to the translation of the delay with $(q+1/2)T_f$.
[25] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[26] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[27] T. Nakajima and S. Watanabe, Phys. Rev. A **70**, 043412 (2004).
[28] L. A. A. Nikolopoulos *et al.*, Phys. Rev. Lett. **94**, 113905 (2005).
[29] T. Sekikawa *et al.*, Phys. Rev. Lett. **88**, 193902 (2002).