Conclusive Evidence of an Attosecond Pulse Train Observed with the Mode-Resolved Autocorrelation Technique

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]*RIKEN (The Institute of Physical and Chemical Research), 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*
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We report on the direct observation of an attosecond pulse train with a mode-resolved autocorrelation technique. The chirp among the three harmonic fields is specified by analyzing two-photon above-threshold ionization spectra of electrons, resulting in a pulse duration that should be shorter than 450 as, which is, to our knowledge, the first determination of the chirp in the attosecond pulse train with an autocorrelation technique. These results will open the way to full characterization of an attosecond pulse train with its envelope.

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Characterization of the temporal profile of a high-order harmonic field, which is generated with an intense laser field, is of great importance for the progress of ultrafast optical science because a high-order harmonic field can form an extremely short bunch of optical fields in the attosecond regime.

Most researchers, however, owe their success in characterizing attosecond pulses to the use of an intense visible laser pulse interacting with electrons detached from atoms with one-photon absorption of the measured harmonic field. In fact, the measurement of an isolated attosecond pulse was demonstrated in the cutoff region of high-order harmonics by virtue of this principle [1]. The first report of the observation of the attosecond pulse train [2], which is formulated with the Fourier synthesis of many modes of the harmonic field in a plateau region, and other related studies [3–6] have already shown the signatures of the relative phase differences between adjacent harmonics. We cannot, however, straightforwardly imagine the reconstructed attosecond pulse train from the sideband spectra of the electrons caused by two-color above-threshold ionization (two-color ATI) [7,8]

An autocorrelation signal given by the nonlinear interaction between matter and optical fields, on the other hand, directly reflects the shape of the pulse in the primary data, whether it is trainlike or isolated, and it is widely used for characterizing femtosecond laser pulses of visible lasers. There have been, however, only a few attempts [9–11] to apply this method to measure the pulse shape consisting of high-order harmonic fields since the first measurement of the pulse duration of the 9th-order harmonic field in 1998 [12], because the significant issue of low nonlinearity for the nonresonant two-photon interaction of harmonic fields with matter requires a high intensity ($\sim 10^{12}$ W/cm^2), which is $\sim 10^6$ times that required for the second harmonic generation of the visible laser field in a nonlinear crystal, and makes it difficult to detect the correlated signal. In addition, the energy of the harmonic pulse is restricted to a low conversion efficiency, which is typically $\sim 10^{-4}$ [13], from the fundamental laser pulse and also reduced with dissipating optics such as a metal filter.

Here, we report the direct observation of an attosecond pulse train formed by the Fourier synthesis of three harmonic fields of the 11th , 13th, and 15th orders. The electron spectra produced by the two-photon above-threshold ionization (two-photon ATI) [14] are utilized as mode-resolved signals of the autocorrelation measurements both for the pulse train and its envelope. These results clearly indicate the formation of an attosecond pulse train. The analysis of the two-photon ATI spectra, which can be considerably simplified due to the small numbers of harmonic fields, specifies that the chirp among the three harmonic fields should be smaller than 1.4×10^{-32} s^{-2}, resulting in a pulse duration of 450 as.

In our experiment, a fundamental laser pulse with a duration of 40 fs and an energy of 13 mJ is delivered from a chirped pulse amplification system of a Ti:sapphire laser, and focused into a static gas cell filled with xenon gas in a vacuum chamber. The detailed parameters are fixed according to the results reported in Ref. [15]. The pulse energies of the 11th-, 13th-, and 15th-order harmonic fields just behind the gas cell are all estimated to be higher than 1 μJ [13,15], and these high pulse energies are critical for observing the nonlinear interaction of the harmonic field without the help of another intense laser field.

The small divergence of ~ 0.5 mrad is attributed to the phase matching between the harmonic fields and the fundamental laser field [13,15]. The phase-matching effect should also take an important role in selecting the short trajectory of the electron in each xenon atom, and ensure the generation of the attosecond pulse train [16,17].

The generated harmonic fields and the fundamental laser copropagate to the next vacuum chamber including a pair of harmonic separators made of silicon [18]. The reflection near the boundaries of the two closely placed harmonic

separators spatially divides the harmonic fields. One of the divided harmonic fields can be delayed or advanced to another field by moving one of the separators fixed on a translation stage with a piezoactuator. This technique is the same as that used in the previous experiment [19,20] except for the separator material used.

The remaining portion of the fundamental laser, typically $\sim 10^{-4}$ of the incident pulse energy, and the harmonic fields with an order lower than the 9th are eliminated with a tin filter with a thickness of 0.1 μm, of which the measured transmittances of the 11th-, 13th-, and 15th-order harmonic fields are 6%, 14%, and 20%, respectively.

Two replicas of the harmonic fields are introduced in a magnetic bottle photoelectron spectrometer and focused with a concave mirror made of silicon carbide with a radius of curvature of 100 mm. Argon gas, as target atoms of two-photon ATI, is supplied with a capillary tube attached to a pulsed gas valve. Approximately half of the ejected electrons with photoionizations are guided with a magnetic field, and then go through a small bore with a radius of 700 μm. The kinetic energies of the electrons are resolved by measuring the time of flight of the electrons traveling into a flight tube behind the bore with the guidance of the magnetic field.

The harmonic spectra at the focal region of the concave mirror was determined by observing electron spectra with the one-photon absorption of an argon atom because the photon energies of the 11th- and higher-order harmonic fields exceed the ionization energy of the argon atom. The resultant relative intensity ratios of the 11th-, 13th-, and 15th-order harmonic fields, which we, respectively, denote as I_{11}, I_{13}, and I_{15} in this Letter, are 0.46:1:0.28, and the intensities of higher-order harmonic fields are negligible compared with I_{13} due to the tin filter, but measurably large compared with the two-photon ATI signals up to the 23rd order, as is described later. The reductions of the 9th- and 7th-order harmonic fields were confirmed using a VUV $\sim$ XUV spectrometer.

We can expect five peaks, from the lowest-order mode at 22nd originating from two photons of the 11th-order harmonic field, to the highest-order mode at 30th originating from two photons of the 15th-order harmonic field.

The observed two-photon ATI spectra at a fixed delay, shown in Fig. 1, exhibited four peaks. A missing peak at 22nd order mode was attributed to large signals owing to the one-photon absorption with the remaining 21st- and 23rd-order harmonic fields. There was nothing to be regarded as a two-photon ATI peak in the kinetic energy range higher than or equal to the 32nd order mode, suggesting that the harmonic fields of the 17th-order and higher did not notably participate in forming the attosecond pulse train. This result confirms the assumption that there are only three harmonic fields, in our analysis described later.

By changing the delay between the two replicas of the harmonic fields, these four two-photon ATI peaks should

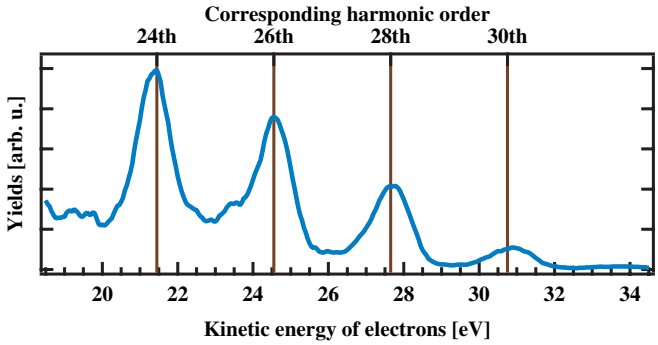

FIG. 1 (color). Typical ATI spectrum at fixed delay.

reveal the correlated signal of the temporal profile synthesized with the harmonic fields to each energy mode. In fact, we clearly observed the modulations of the two-photon ATI peaks with a period of 1.33 fs, which corresponds to half of the period of the fundamental laser field, in the 24th, 26th, and 28th modes, as shown in the three-dimensional graph of Fig. 2.

Before saying eureka, we should briefly analyze how the attosecond pulse train causes this mode-resolved autocorrelation. We define the positive frequency part of the electric field synthesized with each harmonic field mode as

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

where ω_f is the angular frequency of the fundamental laser field. Because the width of the envelope of the pulse train is

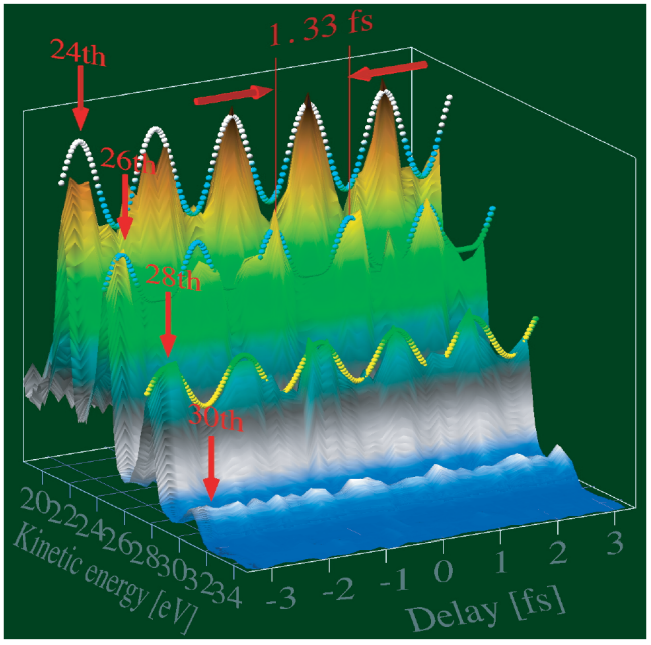

FIG. 2 (color). Energy spectra of ATI electrons depending on delay. This 3D graph corresponds to the mode-resolved autocorrelation of the synthesized harmonic field.

much longer than $T = 2\pi/\omega_f$, it is a good approximation near the peak of the envelope that the amplitude A_{2n+1} should be independent of time, that is, it should be a real constant. The odd integers of $2n_1 + 1$ and $2n_2 + 1$ are the minimum and the maximum orders of the harmonic fields, respectively.

The Fourier amplitude of the signal, $\tilde{S}^{(+)}_{\mathrm{all}}(\Omega_{2(N+1)};\tau)$, with the nonresonant two-photon process induced by two replica of the electric field with delay τ is proportional to [10] $(1/T)\int_{-T/2}^{T/2} dt\{E^{(+)}(t) + E^{(+)}(t-\tau)\}^2 e^{i\Omega_{2(N+1)}t}$, where the angular frequencies should be the discrete modes of $\Omega_{2(N+1)} \equiv 2(N+1)\omega_f$ and the integer N is restricted in the range from $2n_1$ to $2n_2$. The correlated part included in $\tilde{S}^{(+)}_{\mathrm{all}}(\Omega_{2(N+1)};\tau)$ is given as

$$\tilde{S}^{(+)}(\Omega_{2(N+1)};\tau) \propto \frac{1}{T}\int_{-T/2}^{T/2} dt E^{(+)}(t)E^{(+)}(t-\tau)e^{i\Omega_{2(N+1)}t} = \sum_{n=n_{\min}}^{n_{\max}} A_{2(N-n)+1}A_{2n+1}e^{-i\Phi_{(N,n)}}e^{i(2n+1)\omega_f\tau}, \tag{2}$$

where the phase $\Phi_{(N,n)}$ is defined as $\phi_{2(N-n)+1} + \phi_{2n+1}$, and the pair of integers $(n_{\min}, n_{\max})$ should be equal to $(n_1, N-n_1)$ for the case of $N \le n_1 + n_2$ and $(N-n_2, n_2)$ for $N > n_1 + n_2$. The observed electron spectra should reflect the absolute square of $\tilde{S}^{(+)}(\Omega_{2(N+1)};\tau)$.

The electron yields for the actual experiment are approximately proportional to $|\tilde{S}^{(+)}_{\mathrm{all}}(\Omega_{2(N+1)};\tau)|^2$, so that they include a constant background originating from $|\tilde{S}^{(+)}(\Omega_{2(N+1)};0)|^2$, and the oscillating terms corresponding to the carrier frequencies of the harmonic fields, which originate from $\tilde{S}^{(+)}(\Omega_{2(N+1)};0)\tilde{S}^{(+)^*}(\Omega_{2(N+1)};\tau)$ [21]. These are also affected by the volume integration in the focal region. The oscillating terms could not be resolved in the experiment because the surface roughnesses of the XUV optics that we used in this experiment are not expected to give a sufficient visibility of the spatial fringes of the intensity at the focal region of the harmonic field as was calculated in Ref. [21].

The odd integers of $2n_1 + 1$ and $2n_2 + 1$ are 11 and 15, respectively, in our experiment, thus, the mode-resolved electron yielding $I(\Omega_{2(N+1)};\tau) = |\tilde{S}^{(+)}(\Omega_{2(N+1)};\tau)|^2$ can be written as $I(\Omega_{22};\tau) \propto I_{11}^2$, $I(\Omega_{30};\tau) \propto I_{15}^2$, and

$$I(\Omega_{24};\tau) \propto 2I_{11}I_{13}\{1 + \cos(2\omega_f\tau)\}, \tag{3}$$

$$I(\Omega_{26};\tau) \propto \{I_{13} + 2\sqrt{I_{11}I_{15}}\cos(2\omega_f\tau)\}^2 - 8I_{13}\sqrt{I_{11}I_{15}}\sin^2(\Delta\Phi)\cos(2\omega_f\tau), \tag{4}$$

$$I(\Omega_{28};\tau) \propto 2I_{13}I_{15}\{1 + \cos(2\omega_f\tau)\}, \tag{5}$$

where we define the intensity of each harmonic field as $I_{2n+1} \equiv \alpha A_{2n+1}^2$ $(2n+1 = 11, 13, 15)$ with an appropriate constant, α, and the phase difference $\Delta\Phi \equiv (\phi_{11} + \phi_{15})/2 - \phi_{13}$.

Trivial equations for the 22nd- and 30th-order modes can be easily recognized as results that only one harmonic field is concerned in generating these modes.

The two adjacent harmonic fields responsible for generating the 24th- and 28th-order modes cause a sinusoidal modulation with the half period of the fundamental laser field as we have expected. Note, however, that the spectral phases of the harmonic fields, ϕ_{11}, ϕ_{13}, and ϕ_{15}, disappear in these equations, so that we cannot extract any relations among the spectral phases of the three fields from the measured autocorrelation traces in these two modes, while these two modes should always exhibit the perfect sinusoidal modulation whatever happens to the spectral phases. Thus, these two modes give us benchmarks determining the correlated part of the ATI signals on the constant background.

The last mode we should consider is the 26th-order mode. The intensity of $I(\Omega_{26};\tau)$ is modulated with mainly the half period of the fundamental laser field because of the first term in Eq. (4), and somewhat corrected with the second term proportional to $\sin^2(\Delta\Phi)$, which includes information on the relative phases of the three harmonic fields, so that we should be able to determine $|\Delta\Phi|$ from the measured profile of $I(\Omega_{26};\tau)$.

We have to consider what $\Delta\Phi$ is and how it affects the temporal profile of the pulse train before determining it. The general expression of the intensity of the synthesized harmonic field, denoted $I(t)$, can be easily written from the absolute square of Eq. (1) as follows:

$$I(t) = I_{11} + I_{13} + I_{15} + 2\sqrt{I_{11}I_{13}}\cos\{2\omega_f(t+\tau_g) - \Delta\Phi\} + 2\sqrt{I_{13}I_{15}}\cos\{2\omega_f(t+\tau_g) + \Delta\Phi\} + 2\sqrt{I_{15}I_{11}}\cos\{4\omega_f(t+\tau_g)\}. \tag{6}$$

Here, we define $\tau_g \equiv (\phi_{15} - \phi_{11})/(4\omega_f)$.

Obviously, the parameter τ_g is regarded as the group delay, which only induces a temporal shift of the pulse train, while the parameter $\Delta\Phi$ changes the temporal profile. In fact, we can uniquely define the spectral phase ϕ as a quadratic function to the angular frequency Ω such that it passes on the given three points of the spectral phases, ϕ_{11}, ϕ_{13}, and ϕ_{15}, at Ω_{11}, Ω_{13}, and Ω_{15}, respectively. That is, $\phi(\Omega) = \Delta\Phi/(4\omega_f^2)(\Omega - \Omega_{13})^2 + \tau_g(\Omega - \Omega_{13}) + \phi_{13}$, thus, the "chirp" at Ω_{13} should be expressed as, $\ddot{\phi}(\Omega_{13}) = \Delta\Phi/(2\omega_f^2)$. Hence, all we have to do is to determine $\Delta\Phi$, or equivalently $\ddot{\phi}(\Omega_{13})$, if we would like to determine the temporal profile of the intensity of the pulse train formed by three harmonic fields.

We fitted the benchmarks of Eqs. (3) and (5) to the measured autocorrelation traces in Fig. 3, which are obtained by averaging each mode at ± 0.5 eV, at the 24th- and 28th-order modes, respectively, with a constant back-

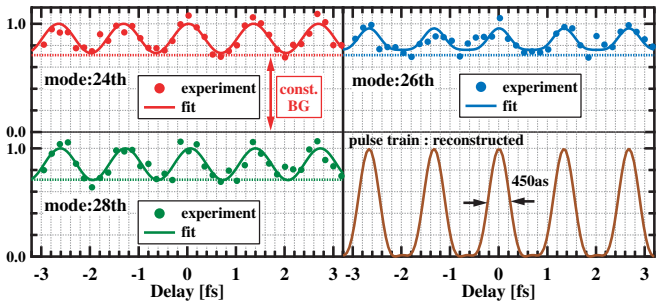

FIG. 3 (color). Autocorrelation traces at three individual modes and the pulse train reconstructed.

ground, and then found that the correlated part of the measured traces should be 0.283 ± 0.12 of the maximum intensity of each mode. The ratios among the intensities of the three harmonic fields were maintained in this procedure. With the assumption that the 26th-order mode includes the correlated part with the same ratio, $I(\Omega_{26}; \tau)$ in Eq. (4) was fitted to the measured trace in Fig. 2 at the 26th-order mode. $|\Delta\Phi|$ and $|\ddot{\phi}(\Omega_{13})|$ were 0.145 ± 0.013 radians and $1.3 \pm 0.1 \times 10^{-32}$ s^2, respectively.

Although the signs of $\Delta\Phi$ and $\ddot{\phi}(\Omega_{13})$ cannot be determined due to the nature of the second-order autocorrelation, we assume them to be a positive for the consistency with that in theories [6] and other related works of the experiment. As a result, these quantities agree well with those reported for other measurement techniques [2,4,6] under the condition that the intensity of the fundamental laser field is estimated to be $\sim 10^{14}$ W/cm^2. We conclude, by substituting the obtained numeric to Eq. (6), that the pulse duration should be 450 as, at most, at full width at half maximum (FWHM), which is almost the same as that for the Fourier limit (445 as), as was shown at right bottom of Fig. 3. This is the finest temporal structure, to our knowledge, observed by the autocorrelation technique.

The photoelectron analysis with nonresonant two-photon-ionization for harmonic electric-field reconstruction (PANTHER) from the mode-resolved autocorrelation traces mentioned above was feasible for determining the characteristic of the train feature of the harmonic field, while it clarified nothing as regards the envelope of the train, because the resolution of the kinetic energy of electrons in this measurement ($\sim$0.5 eV) is not sufficient to find the spectral phase information within each mode. This is one of the reasons we cannot specify this measuring technique as the two-photon-ionization frequency-resolved optical gating [22] demonstrated in Ref. [10]. The train envelope, however, appeared by extending the range of delay in a separate experiment, resulting these traces directly proving that the bunches of the harmonic field were temporally confined within approximately 30 fs. We sampled only delays such that the correlated signal should be the local maximum of the modulation at each delay, for reducing the acquisition time of this experiment. Here, we have determined both the duration of the pulse in the train and the upper limit of the duration of its envelope.

There are still some remaining issues regarding the full characterization of an attosecond pulse train by utilizing the mode-resolved autocorrelation technique by electron spectroscopy. The most crucial issue for fully characterizing an attosecond pulse train is the spectral broadening of the kinetic energy of electrons due to the local electric field induced by a large number of ions left with one-photon absorption, the so-called space-charge effect. This effect would be relaxed by decreasing the density of the target atoms. The reduced number of ATI electrons per laser shot would be compensated with a higher repetition rate if we could use a terrawatt-class laser system at a 1 kHz repetition rate such as one of the authors has developed [23].

Thus, we believe that this study will open a novel way to the full characterization of an intense attosecond pulse train with a method classified as a frequency-resolved optical gating [22] measurement.

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*Electronic address: nabekawa@riken.jp

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