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Complete temporal reconstruction of attosecond high-harmonic pulse trains

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Abstract. The method of complete reconstruction of attosecond bursts has been demonstrated for attosecond high-harmonic pulse trains. The retrieved harmonic field provided detailed information about the envelope and the individual attosecond pulses contained in the attosecond pulse train. The time–frequency analysis revealed complicated spectral chirp structures and the contribution of different quantum paths to attosecond pulse formation.

Contents

1. Introduction 2
2. Reconstruction procedure for attosecond pulse trains 3
3. Temporal reconstruction of attosecond pulse trains 7
4. Time–frequency analysis of attosecond pulse trains 9
5. Conclusion 12
Acknowledgment 12
References 12
1. Introduction

High-harmonic generation (HHG) is a powerful method to produce attosecond pulses [1]–[7]. Gaseous atoms exposed to an intense femtosecond laser field emit high-harmonic radiation at the driving laser frequency. With proper control of harmonic generation conditions, the harmonic radiation can form an attosecond pulse train containing one pulse per half-optical cycle [1]. For the application of attosecond pulses to light–matter interactions, rigorous temporal characterization of the attosecond pulses is essential for a proper understanding of the physical processes. The temporal reconstruction of the attosecond pulse train can be obtained by applying a cross-correlation technique to the photoionization of atoms using attosecond high-harmonic pulses and femtosecond laser pulses [3]–[7]. Averaged temporal characteristics can be obtained using the reconstruction of attosecond burst by interference of two-photon transition (RABITT) method [3]. For rigorous temporal characterization, the method of complete reconstruction of attosecond bursts (CRAB) was proposed using the frequency-resolved optical gating (FROG) technique [7]. The application of the CRAB method to the characterization of isolated attosecond pulses has been done [4, 6], but its application to the case of attosecond pulse trains is not straightforward due to the involvement of many individual attosecond pulses within one pulse train.

The generation of attosecond high-harmonic pulses is a result of complex processes coming from the interplay between laser propagation in an ionizing gas medium, single atom response to the laser field and macroscopic effects. An electron is tunnel-ionized and accelerated in the laser field. The electron can recombine with its parent ion, emitting harmonic radiation as a result of a repetitive process. Since the kinetic energy of the recombined electron is increasing (decreasing) for the shorter (longer) quantum paths with the recombination time, lower-order harmonics are emitted earlier (later) than higher-order harmonics and the attosecond pulse is positively (negatively) chirped within a half cycle of the laser pulse, known as an ‘atto-chirp’. The high-order harmonics can also be chirped over several cycles of the laser pulse, i.e. the frequency of a certain harmonic can evolve in time, depending on the temporal profile of the laser field, known as the ‘frequency-chirp of harmonics’ [8]. The attosecond pulse train can thus contain a complicated chirp structure. In addition, the temporal profile of an attosecond pulse train contains information about the interaction of an ejected electron with the Coulomb field of an atom or molecule in a strong laser field [9]. Consequently, it is essential to accurately characterize attosecond pulses not only to probe ultrafast phenomena but also to trace harmonic generation processes as a marker functional for quantum path analyses of HHG [10, 11], control of attosecond electron wavepackets [12] and molecular tomography [13]. Most previous studies, however, have been carried out in the frequency domain owing to measurement difficulties in the time domain [9, 14]. In the present study, the CRAB method is successfully demonstrated for rigorous temporal reconstruction of attosecond high-harmonic pulse trains. First, we examine the reconstruction process used for characterizing attosecond pulse trains. The method is then applied to reconstruct the attosecond pulse trains obtained at two different laser intensities. The reconstruction results reveal the chirp structure of attosecond pulse trains as well as the temporal profiles of the envelope and of the individual attosecond pulses, allowing a systematic examination of high-order harmonics.
2. Reconstruction procedure for attosecond pulse trains

An accurate characterization of attosecond harmonic pulses can be implemented by the CRAB method. An attosecond harmonic pulse ionizes an atom in a weak probe laser pulse, generating an electron carrying the phase information of the attosecond harmonic pulse. The electron can gain or lose its kinetic energy due to the probe laser pulse, depending on the timing of the ionization. The photoelectron spectrum thus varies depending on the time delay between the attosecond harmonic pulse and the probe laser pulse. For the attosecond pulse train that consists of odd harmonics, for example, even-order sidebands appear in photoelectron spectra that modulate twice in one optical cycle. The temporal profile of the attosecond harmonic pulse can be found from the photoelectron spectra obtained with a time-delay scan using a reconstruction algorithm, such as the principal component generalized projection algorithm (PCGPA). In this section, we discuss the procedure used for the reconstruction of attosecond harmonic pulse trains.

The ionization of an atom by a high-harmonic pulse in the presence of a probe laser pulse can be well described by the strong field approximation. The electron motion, after ionization, can be well described by the strong field approximation. The electron can be modulated twice in one optical cycle. The temporal profile of the attosecond harmonic pulse can be found from the photoelectron spectra obtained with a time-delay scan using a reconstruction algorithm, such as the principal component generalized projection algorithm (PCGPA). In this section, we discuss the procedure used for the reconstruction of attosecond harmonic pulse trains.

The ionization of an atom by a high-harmonic pulse in the presence of a probe laser pulse can be described by the strong field approximation. The electron motion, after ionization by an attosecond harmonic pulse $E_X(t)$, can be described as a free charged particle moving in the laser field by neglecting the Coulomb potential of the atom. In such an approximation, the transition amplitude $b(v, \tau)$ to the continuum state $|v\rangle$ with momentum $v$ can be represented in atomic units as [7, 15, 16]

$$b(v, \tau) = -i \int_{-\infty}^{+\infty} dt' E_a(t' - \tau) d[v + A(t')] \exp[i\phi(t')] \exp[i(\omega_e + I_p) t']$$

(1)

where $\omega_e (= \frac{v^2}{2})$ is the electron energy, $I_p$ is the ionization potential and $A(t')$ is the vector potential of the laser field. The function $\phi(t')$ is the temporal phase modulator representing the energy gain or loss of the photoelectron in the laser field [16], which is defined as

$$\phi(t') = - \int_{t'}^{\infty} dt'' [v \cdot A(t'') + A^2(t'')/2].$$

(2)

The dipole transition matrix element $d[v + A(t')]$ can be evaluated using the He photoionization cross section [17].

For the reconstruction, the PCGPA was applied [18]. PCGPA is a blind FROG technique, in which both signal $X(t)$ and gate $G(t)$ can be found for a given intensity constraint set by measured photoelectron spectra, $S(\omega, \tau)$. In PCGPA, the reconstructed spectrogram $S_R(\omega, \tau)$ is defined as

$$S_R(\omega, \tau) = \left| \int_{-\infty}^{\infty} dt \ X(t - \tau) G(t) \exp(i\omega t) \right|^2.$$  

(3)

Then, the reconstructed signal $tX(t)$ and gate $tG(t)$ are related to the attosecond pulse $E_X(x)$ and the vector potential $A(t)$ without normalization factors as

$$X(t) \approx \int_{-\infty}^{+\infty} d(\omega) \omega_k^{1/4} \left[ \int_{-\infty}^{+\infty} E_X(t') \exp(i\omega t') dt' \right] \exp(-i\omega t) d\omega,$$

$$G(t) \approx \exp \left[ -i \int_{-\infty}^{+\infty} dt \{v_0 A(t) + A^2(t)\} \right].$$

(4)

Here, a constant $v_0$ is found by the algorithm that minimizes the error between $S(\omega, \tau)$ and $S_R(\omega, \tau)$. In PCGPA, the initial values for $X^{(i=0)}(t)$ and $G^{(i=0)}(t)$ are set to unity. Here, $i$ is the
index of the iteration. The following iteration process is continued until the root-mean-square
error between $S(\omega, \tau)$ and $S_R(\omega, \tau)$ is stabilized:

1. Reconstructed spectra $S_R^{(i)}(\omega, \tau)$ are obtained using $E_X^{(i)}(t)$ and $G^{(i)}(t)$.
2. The intensity of $S_R^{(i)}(\omega, \tau)$ is set by the photoelectron spectra $S(\omega, \tau)$ obtained in the
experiment.
3. New guesses for $E_X^{(i+1)}(t)$ and $G^{(i+1)}(t)$ are found from $S_R^{(i)}(\omega, \tau)$ using the power
method [18].

There are some issues to be clarified for the application of the CRAB procedure. The use of
angle-integrated photoelectron spectra needs to be examined first. The characterization of an
attosecond pulse train requires the measurement of many photoelectron spectra owing to their
long duration compared to an isolated single attosecond pulse. A photoelectron spectrometer
with a wide acceptance angle is thus preferred for efficient collection of photoelectrons. The
wide acceptance angle, however, may cause a systematic error in the reconstruction result. For
the case of isolated single attosecond pulses, it is known that the CRAB method should be
applied for photoelectron spectra obtained with a narrow acceptance angle [15, 16, 19, 20]. For
the case of attosecond pulse trains, the applicability of angle-integrated photoelectron spectra to
the CRAB method has not been examined yet.

Before applying the reconstruction procedure to experimental data, it was tested with
simulated angle-integrated photoelectron spectra. The angle-integrated photoelectron spectrum
$\bar{S}(\omega, \tau)$ with an acceptance angle of 90° (corresponding to the solid angle of 2π) is given by

$$\bar{S}(\omega, \tau) = 2\pi \int_0^{\pi/2} S(\omega, \tau, \theta) \, d\theta.$$  

(5)

The photoelectron spectrum, $S(\omega, \tau, \theta)$, for a specific angle $\theta$ can be written as

$$S(\omega = v^2/2, \tau, \theta) = |b(v, \tau)|^2 \sqrt{\omega_c} \sin \theta.$$  

(6)

The quantity $\sqrt{\omega_c} \sin \theta$ is introduced to take into account the density of states. The angle-integrated photoelectron spectrum $\bar{S}(\omega, \tau)$, calculated by equation (5), is shown in figure 1(a). The attosecond pulse train was defined by the superposition of odd harmonics from the 17th
to the 25th order. The envelope duration of the attosecond pulse train was about 7 fs, as shown
in figure 2(b), and the duration of the individual attosecond pulse at the center of the train was
about 470 as. The intensity of the 30 fs probe laser pulse at 815 nm was $1 \times 10^{12} \text{ W/cm}^2$.

In order to apply PCGPA for the reconstruction of the attosecond pulse train near the
ionization potential $I_p = 16.16\omega_0$ ($\omega_0$ is the central frequency of the probe laser pulse, 1.52 eV),
we have slightly modified the algorithm. Since we do not have information below the ionization
threshold of the He target gas, the harmonics below the ionization threshold were ignored in the
analysis. In the reconstructed photoelectron spectra $S_R(\omega, \tau)$, the sideband appeared at $16\omega_0$
because the momentum dependence $\mathbf{v} \cdot \mathbf{A}$ in $\phi(t)$ was not considered for the gate function $G(t)$
in PCGPA. Thus, the photoelectron spectra $S_R^{(i)}(\omega, \tau)$ below the ionization threshold could not
be set to zero in procedure (2). Instead, $S_R^{(i)}(\omega, \tau)$ was assigned only for the spectral region
above $16.5\omega_0$, and the spectral amplitude of $X^{(i)}(t)$ below the ionization threshold as set to
zero after procedure (3). This modification allowed $S_R^{(i)}(\omega, \tau)$ to have a sideband $16\omega_0$ and to
satisfy the intensity distribution of $\bar{S}(\omega, \tau)$ above $16.5\omega_0$. The reconstructed spectra $S_R(\omega, \tau)$
then showed an almost identical interference pattern to $\bar{S}(\omega, \tau)$, as shown in figure 1(b).
Figure 1. (a) Simulated angle-integrated photoelectron spectra calculated with an acceptance angle of 90°. (b) Reconstructed photoelectron spectra. \(T_0\) is the period of the laser pulse \(1T_0 = 2.72\) fs.

Figure 2. (a) Temporal profiles of the original attosecond pulse (black solid line), the attosecond pulse train reconstructed using angle-integrated photoelectron spectra (red dashed line, exactly overlapping with the black solid line) and the attosecond pulse train reconstructed using angle-integrated photoelectron spectra with spectral broadening (blue dotted line). (b) Temporal profiles of probe laser pulses are shown by the black solid line, red dashed line and blue dotted line, respectively, for the cases in panel (a).

The reconstruction result gives the temporal profiles of the probe laser pulse and the attosecond pulse train. Although angle-integrated spectra were used for the reconstruction, the reconstructed attosecond pulse was almost identical to the original attosecond pulse when simulated data were used, as shown in figure 2(a). This was quite surprising, because the use of the angle-integrated photoelectron spectra caused a reconstruction error in the case of single attosecond pulses [19]. However, the simulated result in figure 2 shows that such an
Figure 3. (a) Angle-integrated photoelectron spectra and (b–f) photoelectron spectra calculated for the detection angles of 20°, 40°, 60°, 70° and 80°, respectively. The red dashed line that connects the peaks of the sidebands in (a) is shown in each figure for comparison. All spectra were normalized for clarity. (e) The number of photoelectrons generated with respect to detection angle when the probe laser is not overlapping with the attosecond pulse train ($\tau = -25T_0$, black dashed line with squares) and overlapping ($\tau = 0$, red dashed line with triangles).

angular dependence is negligible for the reconstruction of an attosecond pulse train. In order to investigate the angular dependence, photoelectron spectra $S(\omega, \tau, \theta)$, calculated for specific angles, are shown in figures 3(b)–(f) for detection angles $\theta = 20^\circ$, $40^\circ$, $60^\circ$, $70^\circ$ and $80^\circ$, and the angle-integrated spectra $\bar{S}(\omega, \tau)$ are shown in figure 3(a). For comparison, red dashed lines are drawn that connect the maxima of sidebands (even order) calculated for $\bar{S}(\omega, \tau)$. For $\theta < 70^\circ$, all photoelectron spectra show the same pattern, but the intensity of sidebands decreases because $v \cdot A$ is getting smaller as $\theta$ increases in equation (2). For $\theta > 70^\circ$, the photoelectron spectra show a different pattern that is more pronounced when $v$ is small and $\theta$ is large. For $S(\omega, \tau, \theta = 70^\circ)$, the 18th sideband is shifted by $\pi/2$, but the sidebands above the 20th are not shifted, as shown in figure 3(e). For $S(\omega, \tau, \theta = 80^\circ)$, all sidebands are shifted by $\pi/2$, as shown. Therefore, the use of the angle-integrated photoelectron spectra may cause a reconstruction error. However, the contribution of $S(\omega, \tau, \theta > 70^\circ)$ to $\bar{S}(\omega, \tau)$ was only about 4%, because the number of electrons is approximately proportional to $\cos^2 \theta \sin \theta$, as shown in figure 3(g). Thus, the angle-integrated spectra could be very similar to $S(\omega, \tau, \theta = 40^\circ)$ where the maximum contribution occurred, and the reconstruction was very accurate. In addition, the
duration of the reconstructed probe laser, 31 fs, was quite comparable to the original duration of 30 fs, as shown in figure 2(b). Consequently, the attosecond pulse train could be successfully reconstructed, even with angle-integrated photoelectron spectra.

Next, the effect of spectral broadening on the reconstruction results needs to be examined. Photoelectron spectra often become broadened due to the limited resolution of a spectrometer used in experiments. This can cause a serious problem because broadened spectra are supplied for reconstruction. We examined the reconstruction error due to the limited resolution of our spectrometer. The broadened spectrum, \( \tilde{S}_b(\omega, \tau) \), was simulated by calculating the convolution of the instrumental function \( \Gamma(\omega) \) and the original spectra \( \tilde{S}(\omega, \tau) \) as

\[
\tilde{S}_b(\omega, \tau) = \int \tilde{S}(\Omega, \tau) \Gamma(\omega - \Omega) \, d\Omega.
\]

The instrumental function contains the delta function response of the spectrometer that represents the resolution of the spectrometer. The resolution of our magnetic bottle time-of-flight (MBTOF) spectrometer, measured separately using Ne III emission lines, was less than 1.4% of the photoelectron energy (11 meV at 0.8 eV), but increased linearly with photoelectron energy. For the estimation of broadening effects, a normalized Gaussian function with the full-width at half-maximum (FWHM) of 0.014\( \omega_e \) was used. An attosecond pulse train was reconstructed with \( \tilde{S}_b(\omega, \tau) \), and the result is shown in figure 2(a). The individual attosecond pulses in the train still showed very good agreement, but the envelope duration of the train was underestimated. For a Gaussian pulse with a bandwidth of \( \Delta \omega \), the broadened bandwidth is given by \( \sqrt{(\Delta \omega)^2 + (0.014 \omega_e)^2} \). The duration error is, thus, large for narrow bandwidth harmonics. The estimation showed that the duration of the reconstructed 21st harmonic became 6.6 fs, while the original harmonic duration was 7.0 fs, showing about 6% error. Although the spectral broadening caused an underestimation in the duration of the harmonic pulse, its effect was not serious in our measurement owing to the good spectral resolution of our MBTOF spectrometer. The parameters used for the calculation of the photoelectron spectra were quite similar to the experimental result obtained with the attosecond pulse train generated at the intensity of \( 1.6 \times 10^{14} \text{ W/cm}^2 \). Consequently, the simulation result can support the reconstruction procedure applied to the experimental results presented in section 3.

3. Temporal reconstruction of attosecond pulse trains

An attosecond pulse train is produced as a result of the coherent superposition of a range of high harmonics emitted from atoms driven by an intense laser pulse. As the HHG process is an interplay between the atoms and driving laser pulse, the formation of individual attosecond pulses in the train can be significantly influenced by the deformation of the driving laser pulse in the ionizing medium. Since the complete reconstruction of high-harmonic pulses by the CRAB method gives full information about the temporal structure of the attosecond pulse train, the reconstruction result can reveal how the macroscopic effects influence the formation of attosecond pulses in the train.

The temporal characterization of high-harmonic pulses obtained from Ar has been achieved. The experiment was carried out with a 1 kHz Ti:sapphire laser, generating pulses of 33 fs duration at 815 nm. The laser pulse was split into two parts by an 80:20 beam splitter. Using a concave mirror of \( f \)-number 60, the first femtosecond pulse was focused onto a 3 mm Ar gas cell placed 6 mm after the focus for harmonic generation. The laser pulse, propagating together
Figure 4. Photoelectron spectra of He ionized by attosecond harmonic pulses with a dressed laser field. High harmonics were obtained with a driving laser pulse at an intensity of (a) $1.6 \times 10^{14}$ W/cm$^2$ and (b) $3.2 \times 10^{14}$ W/cm$^2$. The reconstructed spectra obtained using PCGPA are shown in (c) and (d) for cases (a) and (b), respectively.

With the harmonic pulse, was blocked by a 200-nm-thick Al filter. A second femtosecond laser pulse, acting as a probe pulse, was combined with the harmonic pulse using a holed mirror. The time delay between the two pulses was controlled using a piezo-electric transducer. A gold-coated toroidal mirror was used to focus both pulses onto an He target, and the energy spectrum of the photoelectrons generated from He was then measured using an MBTOF spectrometer. The time delay between two pulses was calibrated using a Michelson interferometer attached to a delay line. The time delay ‘zero’ was determined separately by finding the maximum intensity of the above-threshold ionization of Ar generated by two laser pulses—harmonic generation and probe pulses—after removing the Al x-ray filter. From this time delay setting, the calibration of harmonic emission time with respect to the driving laser pulse could be specified. After obtaining photoelectron spectra covering the full overlap between the harmonic generation pulse and the probe laser pulse, temporal reconstruction by the CRAB method was performed.

The strongest advantage of the CRAB technique is its completeness in revealing all the information of the attosecond pulses, including both individual pulses and their envelopes. For the investigation of macroscopic effects, photoelectron spectra were obtained at two laser intensities, $1.6 \times 10^{14}$ W/cm$^2$ and $3.2 \times 10^{14}$ W/cm$^2$, as shown in figures 4(a) and (b), respectively. The reconstruction was carried out by following the procedure given in section 2. The temporal profile of the probe laser pulse can also be obtained, since PCGPA is a blind FROG algorithm. The retrieved probe laser pulse is shown in figure 5. The durations of the retrieved probe pulses were 36 and 33 fs, respectively, for the low and high intensity cases, which are very similar to the duration of the laser pulse, 33 fs, obtained using a second-harmonic FROG measurement. When the maximum of the photoelectron spectrum was set to unity, the RMS error of the reconstructed photoelectron spectrum was 0.014 and 0.012 for the low- and
Figure 5. Temporal profiles of reconstructed attosecond pulse trains corresponding to the cases of low (figure 4(a)) and high intensities (figure 4(b)), as indicated by the blue and red lines, respectively. The reconstructed temporal profile of the probe laser pulses is also shown (red dashed line). The duration of each pulse in the train is shown by squares and circles for the low and high intensities, respectively.

The duration of each pulse in the train is shown by squares and circles for the low and high intensities, respectively. The temporal reconstruction of attosecond pulse trains could thus be successfully performed using the CRAB method adopting PCGPA. The temporal profile of the attosecond pulse train is also shown in figure 5. The duration of each attosecond pulse for the low intensity case is 310–360 as. This, however, is greatly reduced to 190–250 as for the high intensity case. The shorter attosecond formation can be interpreted as follows: more harmonics are generated and atto-chirp is favorably set due to the rapid increase in the electric field during a half-optical cycle in the high intensity case [21, 22].

Figure 5 also shows that, at low laser intensity, the harmonic emission peaks around the pulse center, while it occurs about three optical cycles earlier at high intensity. This is a propagation effect. A numerical simulation showed that, for the high intensity case, the propagation of a laser pulse in an ionizing medium induced the deformation of the laser pulse shape: while the leading edge remained unchanged as it propagated through a neutral medium, its intensity of the rear portion of the laser pulse became lowered due to plasma defocusing (see [23] for details). As a result, the peak intensity was decreased and the peak position was shifted to an earlier time. The overall harmonic emission followed this temporal shift, as confirmed by the calculations [23]. Thus, the reconstruction result revealed the overall features of the attosecond pulse trains.

4. Time–frequency analysis of attosecond pulse trains

Time–frequency analysis is often useful for understanding the complicated chirp structure of light pulses. Short-time Fourier transform (STFT), as a method of time–frequency analysis, was performed to visualize the chirp structures of an attosecond pulse train [24]. For a deeper investigation of macroscopic effects on harmonic generation, the STFT was calculated first with a long temporal gate of 5$\tau_0$ to observe the harmonic frequency chirp. These spectrograms are
Figure 6. Time–frequency analysis of attosecond pulse trains calculated by STFT. Panels (a) and (b) were calculated with a temporal gate of $5T_0$ for the low and high intensity cases shown in figures 4(a) and (b), respectively.

shown in figures 6(a) and (b) for the low and high intensity cases, respectively. The STFT results in figure 6 reveal clearly when each harmonic was efficiently generated. For the low intensity case, all harmonics were formed strongly around the peak of the laser pulse, as shown in figure 6(a). For the high intensity case, however, higher-order harmonics were generated earlier than lower-order harmonics, as shown in figure 6(b). This result could be due to the deformation of the laser pulse induced during its propagation through the ionizing medium. In addition, from figure 6, the duration of the attosecond pulse train was found to be much shorter than the duration of the driving laser pulse. The measured durations were 6.0 and 5.2 fs for the low and high intensity cases, respectively. Although the bandwidth of each harmonic for the high intensity case was much broader than that for the low intensity case, as shown in figures 4(a) and (b), the envelope duration did not show a significant difference. This happened because the emission of different harmonics occurred at different times for the high intensity case, spreading the emission over time.

Figure 6 also shows the time–frequency behavior of each harmonic. At low intensity, the harmonics were slightly negatively chirped. The negative harmonic chirp came from the dynamically induced chirp due to the rapidly increasing laser field—an indication of a single atom response [25]. On the other hand, at high laser intensity, the self-phase modulation (SPM) in an ionizing medium can induce a positive chirp in the leading edge of the laser pulse, which adds positive chirp to the high harmonics [26]. The positive slope of the harmonic frequency variation in figure 6(b) is a clear indication of the SPM-induced positive chirp, which makes the duration of harmonics longer than the transform-limited value. The durations of the 21st harmonic were found to be 5.8 fs ($\tau_{TL} = 5.7$ fs) and 4.1 fs ($\tau_{TL} = 3.8$ fs) for the low and high intensity cases, respectively. Consequently, the reconstruction results clearly demonstrated the influences of macroscopic effects on the formation of the attosecond pulse train.

For a detailed investigation of the instantaneous frequency composition of individual attosecond pulses in the train, STFT was again performed with a short temporal gate of $0.35T_0$, 083019 (http://www.njp.org/)
Figure 7. Time–frequency analysis of attosecond pulse trains calculated by STFT. Panels (a) and (b) were calculated with a temporal gate of $0.35T_0$ for the low and high intensity cases shown in figures 4(a) and (b), respectively. For comparison, the kinetic energy of the recombining electron calculated using the semi-classical model at 70% of the peak laser intensity is indicated for short (solid line) and long (dashed line) paths.

as shown in figures 7(a) and (b) for the low and high intensity cases, respectively. The atto-chirp (second-order phase, $\partial^2\Phi(\omega)/\partial\omega^2$, or equivalently group delay dispersion) estimated for the most intense part in the train was $1.8 \times 10^{-32} \text{s}^2$ for the low intensity case and $1.1 \times 10^{-32} \text{s}^2$ for the high intensity case. Given that the broader bandwidth of the harmonics can be added constructively when the second-order phase is small, the duration of the attosecond pulse should be shorter for the high intensity case, consistent with the result shown in figure 5.

The harmonic emission time in figures 7(a) and (b) agreed reasonably well with the results calculated with the semi-classical model, indicated as solid and dotted lines for short and long quantum paths, respectively. This is also useful in revealing HHG processes. Since the harmonic generation medium was placed after the focus, the short-path components mainly contribute to the HHG [27]. The observed harmonic component also agrees with the fact that, since the harmonic phase of long-path components varies more sensitively with variation in laser intensity, compared to that of the short-path components, the long-path harmonics become less dominant [28]. Figures 7(a) and (b) clearly show that the short-path components were dominant and formed attosecond harmonic pulses. For the low intensity case, long-path components survived, although weak, because the phase change was not so severe within the intensity
variation during the harmonic generation. The low intensity case in figure 7(a) shows a tail with a negative slope—clear evidence of long-path harmonics. The temporal reconstruction of the attosecond high-harmonic pulses thus revealed the high-harmonic emission generated by long-path components that could not be easily identified in harmonic spectra, along with a strong contribution from short-path components. Consequently, the complete temporal reconstruction by the CRAB method is a powerful tool for revealing all the details of attosecond harmonic pulses.

5. Conclusion

The complete temporal characterization of attosecond high-harmonic pulse trains has been demonstrated using the CRAB technique. Retrieval of complete high-harmonic fields was achieved, offering information about the chirp structure of each harmonic and individual attosecond pulses by the time–frequency analysis. As high-harmonic emission comes from the interactions of an electron with an atom or molecule in a strong laser field, a detailed temporal reconstruction of high-harmonic pulses will provide information pertaining to the atomic and molecular structure with attosecond time resolution. Moreover, the contribution of different quantum paths to harmonic generation could be analyzed. Consequently, the complete temporal reconstruction of attosecond high-harmonic pulse trains demonstrated here will become a powerful tool for accurate characterization of attosecond pulses and for applications such as quantum path control and molecular tomography.

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References


[22] Mairesse Y et al 2003 Science 302 1540