Interferometric autocorrelation of an attosecond pulse train calculated using feasible formulae

Y Nabekawa\(^1\) and K Midorikawa
Laser Technology Laboratory, RIKEN, Wako-shi, Hirosawa 2-1, Saitama 351-0198, Japan
E-mail: nabekawa@riken.jp

*New Journal of Physics* **10** (2008) 025034 (25pp)
Received 17 October 2007
Published 29 February 2008
Online at [http://www.njp.org/](http://www.njp.org/)
doi:10.1088/1367-2630/10/2/025034

**Abstract.** The autocorrelation trace of an attosecond pulse train (APT) directly revealed the pulse envelope in our recent experiment on measuring the two-photon Coulomb explosion of a nitrogen molecule as a correlation signal. Although the spatial overlap of the two replicas of the APT in the correlation measurement was only achieved near the focal region owing to the spatial split of the measured APT field, which is a situation quite different from that of the correlation measurement using a Michelson interferometer, the interference fringes clearly appeared on the correlation envelope and proved the odd symmetry of the electric field to the time translation with a half-period of the driving laser field. In this paper, we show a simple and practical analysis for the propagation and the nonlinear interaction of an APT to simulate the experimental result of the interferometric autocorrelation of the spatially split APT. The spatial convolution of the focused electric field is essential for obtaining the fringes. We also discuss how the autocorrelation should be described in the context of the second-order perturbation theory within a dipole approximation.
1. Introduction

Nowadays, an autocorrelator [1] or an advanced instrument based on an autocorrelation [2] is one of the conventional tools for measuring the pulse shape of a femtosecond laser pulse. It always ensures the temporal confinement of the energy flow of the measured optical field in the region where the two replicas of the optical field are spatially overlapped in a nonlinear medium generating the correlated signal, by scanning delay between the two replicas.

In the field of attosecond science [3]–[5], however, somewhat different approaches from the autocorrelation technique have been the mainstream for determining the pulse characteristics [6]. An isolated attosecond pulse could be retrieved from the streaking electron spectrum with simultaneous irradiation of a waveform-controlled few-cycle laser pulse and its harmonic field in the cutoff region [7, 8]. Another approach to finding phase differences between adjacent harmonic fields in the plateau region is to observe the relative shift between the sideband peaks of the electron spectra [9, 10] generated by two-color above-threshold ionization (TC-ATI) [11]–[13], resulting in the determination of the pulse shape of the attosecond pulse train (APT) [14] formed by Fourier synthesis of the applied harmonic fields to TC-ATI.

We note that the two approaches rely on the fact that the field amplitude or intensity of the visible–near-infrared laser field applied at the focal point does not spatially change within the intersection with the focused attosecond pulse or APT, the beam diameter of which should be significantly smaller than that of the laser field. The interaction between the attosecond pulse/APT and the observed electrons is essentially classified into a linear or one-photon absorption process, so that the intensity distribution of the attosecond pulse/APT in space only affects the electron yield at a local position in the focal spot and does not distort the integrated electron spectrum.

We should, however, recall that the wavelength range of the attosecond pulse/APT can span more than one octave when the pulse duration becomes close to the single-cycle regime [7, 15, 16]. Hence, the beam diameter at the focal point should notably change depending on the wavelength if the spatial profiles of each wavelength component are similar before focusing. We can roughly estimate that the beam diameter of the lowest photon energy component of the near-single-cycle attosecond pulse reported in [7] is 1.8 times larger than that of the highest photon energy component, so that the intensity of the latter component at the center of the focal spot should be more than three times higher than that evaluated from the temporally...
and spatially integrated electron spectrum with one-photon adsorption. We anticipate, from this simple consideration, that the pulse shape of an attosecond pulse/APT depends on the position in the focal spot because of the dependence of the spectral intensity, which gives rise to the question ‘How do we find the real temporal shape of the focused attosecond pulse/APT?’

We do not have any ideas on solving this issue, but we claim that an autocorrelation measurement can reveal the ‘effective’ temporal profile of a focused attosecond pulse/APT. Nonlinear interaction accompanying non-resonant two-photon absorption of an attosecond pulse/APT, which is utilized for generating an autocorrelation signal, is an instantaneous response to the intensity profile in space and time, in contrast to a time-integrated response of linear interaction with one-photon absorption. Hence, the signature of the space dependence of the temporal profile should appear even after the spatial integration of the correlated signal, and the resultant trace should reflect the in situ pulse shape effectively.

In this paper, we present simple formulae describing the focusing and the autocorrelation of an APT. The numerical calculation based on these formulae clarifies the importance of the nonlinear spatial profiles of the focused APT beam in the appearance of the correlation trace and the interference fringes. We will briefly introduce the experimental conditions and the results of the interferometric autocorrelation (IAC) measurement of an APT [17, 18] in the next section, then show the modeled formulae that can reproduce the experimental condition in the subsequent section. After demonstrating the results of the numerical calculation, we will discuss how the pulse shape emerges in the autocorrelation trace depending on the nonlinear interaction, and then finally provide a summary.

2. Experiment

The experimental result of the IAC measurement has already been reported in [17, 18]. We present only the substance of the experiment in this section. The set-up is schematically shown in figure 1. The high-order harmonic fields generated from the Xe gas medium filled in a static gas cell co-propagates with the driving fundamental pulse of a Ti:sapphire laser, the energy and the pulse width of which are $\sim 15 \text{ mJ}$ and $\sim 40 \text{ fs}$, respectively. The pulse energy of the harmonic field is estimated to be more than $1 \mu\text{J}$ in total [19, 20]. The harmonic fields are reflected near the boundaries of two harmonic separator mirrors [21] such that they are spatially split into two replicas to be correlated. One of the separator mirrors is mounted on a translation stage to adjust or scan the delay between the two replicas. The undesired edge of the two reflected harmonic beams, which may contain the contribution of the dipole phase originating from the electron traveling through a long trajectory [14, 22], is truncated using an aperture with a diameter of
2 mm. A concave mirror with a radius of curvature of 200 mm focuses these harmonic beams into the molecular beam of nitrogen. The ions related to the nitrogen molecule are mass and energy resolved using an ion spectrometer to measure the time-of-flight (TOF) of accelerated ions, where the TOF axis is set to be parallel to the direction of polarization.

In figures 2(a) and (b), we show the resultant TOF mass spectrum at $m/z = 14$ and the autocorrelation image emerging on the TOF mass spectrum upon scanning delay. By extracting the area profiles of the side peaks denoted as S and S’, which correspond to the singly charged atomic nitrogen originating from the Coulomb explosion of a doubly charged molecular nitrogen, we can obtain the IAC, as shown in figure 2(c). We can recognize that the APT is certainly formed by observing the bunches at fixed intervals of a half-period of the optical cycle of the driving laser field (1.33 fs). The aspect for the peaks and the dips of interference fringes on the correlation envelope is direct evidence of the odd symmetry in the APT field with respect to a time translation of 1.33 fs.

We reported that the distribution of the harmonic fields forming the APT was in the range from 9th to 19th orders in the previous papers [17, 18]. We found, however, that the spectrum of the APT should contain harmonics lower than 9th order, through a separate measurement using a spectrograph for vacuum ultraviolet (VUV). Thus, the simulated results described in section 4 include the contribution of the harmonic fields ranging from the 5th to 19th orders.

3. Formulation

Our goal here is the reproduction of the IAC trace, depicted in figure 2(c), in a simple manner. We can easily see that in the experimental set-up, the crucial point for observing the autocorrelation and the interferometric fringes is the spatial split and the focusing of the

Figure 2. (a) TOF mass spectrum at $m/z = 14$. Side peaks denoted as S and S’ are evidence of Coulomb explosion of the molecular nitrogen ion. (b) Two-dimensional (2D) image of the correlation obtained by arranging the TOF mass spectra in accordance with delay. (c) Average area profile of the side peaks of the above image.
measured APT. Thus, we first concentrate on explaining how we should express the spatial effect and the propagation of the APT.

3.1. Propagation of APT

The basic formulae for describing an electromagnetic field are the Maxwell equations. These are always correct without any approximations. They can be reduced to a scalar wave equation of a electric field with an appropriate gauge fixing condition and the assumption that there is no charge or current in vacuum and that the linear polarization does not significantly change during the propagation. These conditions are satisfied in our experimental set-up because the density of the molecular beam or generated ions/electrons is so low that these ion/electrons can be neglected as radiation sources, and the APT is focused with a small F number (~1/100).

Thus, we can start from the well-known wave equation for the scalar electric field of an APT, $E_{apt}(\vec{r}; t)$.

$$\left( \hat{\nabla}^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E_{apt}(\vec{r}; t) = 0,$$

where we define variables of spatial coordinates and time as $\vec{r}$ and $t$, respectively. The velocity of light in vacuum is denoted as $c$. The spatiotemporal evolution of the electric field should obey equation (1), although this equation does not solely express the electric field because we do not define boundary and/or initial conditions, which are specified by the physical conditions.

The electric field is formed of positive and negative frequency parts, $E^{\pm}(\vec{r}; t)$, namely, $E_{apt}(\vec{r}; t) = E_{apt}^{+}(\vec{r}; t) + \tilde{E}_{apt}^{-}(\vec{r}; t)$, and $E_{apt}^{-}(\vec{r}; t)$ is decomposed into the frequency components or the harmonic modes $\tilde{E}_{apt}^{+}(\vec{r}; \omega_{2n+1})$ such as

$$E_{apt}^{+}(\vec{r}; t) = \sum_{n=n_{\text{min}}}^{n_{\text{max}}} \tilde{E}_{apt}^{+}(\vec{r}; \omega_{2n+1}) e^{-i\omega_{2n+1}t}.$$

The negative frequency part should be the same as the complex conjugate of $E_{apt}^{+}(\vec{r}; t)$ because $E_{apt}(\vec{r}; t)$ is a real field. The angular frequency of the $2n + 1$st order harmonic mode, $\omega_{2n+1}$, is $2n + 1$ times that of the fundamental laser field, $\omega$, and $n$ runs from $n_{\text{min}}$ to $n_{\text{max}}$ so that $2n_{\text{min}} + 1$ and $2n_{\text{max}} + 1$ are the minimum and the maximum order of the harmonic mode.

Note that we have neglected the bandwidth of each harmonic component, resulting in the train of pulses continuing infinitely in the time domain when using equation (2). This is, however, a good approximation for elucidating the basic properties of the APT near the peak of the train envelope without time-consuming numerical calculations for the finite bandwidth in each mode. The spectral decomposition in equation (2) can be applied to a wave field having a continuous spectrum merely by replacing $\omega_{2n+1}$ with $\omega$ and $\sum$ with $\int_{0}^{\infty}$. In general, the support of $\tilde{E}_{apt}^{+}(\vec{r}; \omega)$ is far from the origin on the $\omega$-axis, hence the lower bound of the integration to $\omega$ can be extended to $-\infty$ in many cases.

By substituting equation (2) into (1) and regarding the independence of each mode, we obtain the well-known Helmholtz equation,

$$\left( \hat{\nabla}^2 + k^2(\omega_{2n+1}) \right) \tilde{E}_{apt}^{+}(\vec{r}; \omega_{2n+1}) = 0,$$

with the dispersion formula for the wavenumber, $k(\omega_{2n+1}) = \omega_{2n+1}/c$. This equation is the origin of the Fresnel–Kirchhoff diffraction integration to obtain the electric field at a certain position.
from a given source electric field and its gradient on the surrounding surface. We can find any unknown APT field at any place, in principle, by executing exactly the Fresnel–Kirchhoff diffraction integration, while it is not easy or preferable to perform this kind of integration from the practical point of view. We may be able to use approximate diffraction formulae derived from this integration, which are the so-called Fresnel and Fraunhofer diffraction formulae, by assuming that a typical transverse size of the electric field is much smaller than the distance between the source and propagated fields. It is not, however, obvious whether we can apply this approximation to an APT in the single cycle-regime, because the requirement for the approximation is related only to the spatial dimensions.

Thus, we approximate equation (3) before integration such that the evolution of an APT is ensured. Brabec and Krausz found that the slowly evolving wave approximation (SEWA) of the nonlinear wave equation in a dispersive medium accurately describes the light pulse propagation down to the single-cycle regime [23]. In our simple model of the linear wave equation in a dispersionless medium, SEWA coincides with the slowly varying envelope approximation (SVEA). Applying SVEA to equation (3) with a decomposition of \( \hat{E}_{\text{apt}}^+(\vec{r}; \omega_{2n+1}) \) into a plane wave, \( e^{i(k(\omega_{2n+1})z)} \), and the slowly varying amplitude, \( \tilde{u}^+(\vec{r}; \omega_{2n+1}) \) such that

\[
\hat{E}_{\text{apt}}^+(\vec{r}; \omega_{2n+1}) \equiv \tilde{u}^+(\vec{r}; \omega_{2n+1})e^{i(k(\omega_{2n+1})z)},
\]

we obtain

\[
i \frac{\partial}{\partial z} \tilde{u}^+(\vec{r}; \omega_{2n+1}) = -\frac{\nabla_\perp^2}{2k(\omega_{2n+1})} \tilde{u}^+(\vec{r}; \omega_{2n+1}),
\]

where we define the propagation axis to be \( z \) and the transverse vector and its differential operator as \( \vec{r}_\perp \) and \( \nabla_\perp \), respectively. Because equation (5) has a form identical to that of the Schrödinger equation describing a free particle on a 2D plane, it can be a well-defined problem by imposing an initial condition for \( z \). Setting \( \tilde{u}^+(\vec{r}_\perp', z_0; \omega_{2n+1}) \) at \( z = z_0 \), the slowly varying amplitude at \( z \) is given by

\[
\tilde{u}^+(\vec{r}_\perp', z; \omega_{2n+1}) = \int d^2\vec{r}_\perp''G(\vec{r} - \vec{r}_\perp', z - z_0)\tilde{u}^+(\vec{r}_\perp', z_0; \omega_{2n+1}),
\]

where \( G(\vec{r}_\perp - \vec{r}_\perp', z - z_0) \) is the retarded propagator defined as

\[
G(\vec{r}_\perp - \vec{r}_\perp', z - z_0) \equiv \theta(z - z_0) \exp \left\{ -i\hat{H}(z - z_0) \right\} \delta^2(\vec{r}_\perp - \vec{r}_\perp').
\]

The operator \( \hat{H} \) should be \(-\nabla_\perp^2/2k(\omega_{2n+1})\). We can easily confirm that the right hand side of equation (6) is the solution of equation (5) by substituting equation (7) into (6), and \( G(\vec{r} - \vec{r}_\perp', z - z_0) \) should be the Green function to the operator \( i\partial/\partial z - \hat{H} \) satisfying the equation

\[
\left\{ i \frac{\partial}{\partial z} - \hat{H} \right\} G(\vec{r}_\perp - \vec{r}_\perp', z - z_0) = i\delta(z - z_0)\delta^2(\vec{r}_\perp - \vec{r}_\perp').
\]

We know that the right-hand side of equation (7) can be transformed as

\[
\theta(z - z_0) \exp \left\{ -i\hat{H}(z - z_0) \right\} \delta^2(\vec{r} - \vec{r}_\perp') = \theta(z - z_0)\frac{i}{2\pi} \frac{k(\omega_{2n+1})}{z - z_0} \exp \left\{ -\frac{i}{2(z - z_0)} \frac{k(\omega_{2n+1})}{2(z - z_0)} (\vec{r}_\perp - \vec{r}_\perp')^2 \right\}
\]

by referring to a standard textbook of applied mathematics [24].
Thus, we conclude that equation (9) of the near-single-cycle pulse. We can recognize, by looking at the off-axis retardation time, with experiment. We omit the superscript propagation effect between the harmonic separator mirrors and the aperture in the actual and $E_{\text{APT}}$. Two spatially divided fields on the pupil plane with two apertures, denoted as $A_{\text{APT}}$. To focus the APT, the second-order correction term field, which originates from a homogeneous solution of equation (9) to say, the same as that in the para-axial approximation of the spatial mode of a propagating laser plane, which we call the pupil plane according to the convention of optics, as $E_{\text{APT}}$. This discrepancy is, needless to say, the same as that in the para-axial approximation of the spatial mode of a propagating laser field, which originates from a homogeneous solution of equation (5). In our experimental set-up to focus the APT, the second-order correction term $-(\vec{r}_{\perp} - \vec{r}_{\perp}')^2/(z-z_0)^2$ is estimated to be only $\sim 10^{-9}$ at most, so that the error of retardation time should be much less than 1 attosecond. Thus, we conclude that equation (11) is appropriate for simply describing the propagation of an APT.

In the subsequent sections, we simplify the experimental condition as depicted in figure 3. Two spatially divided fields on the pupil plane with two apertures, denoted as $E_A^+(r_{\perp}; t)$ and $E_B^+(r_{\perp}; t - \tau)$, are focused with a length of $f$ in the calculation by neglecting the propagation effect between the harmonic separator mirrors and the aperture in the actual experiment. We omit the superscript $+$ and the subscript $\text{APT}$ for simplicity and define the delay between the two fields as $\tau$. The propagated field from each field on the focal plane, $E_A^+(\vec{r}_{\perp}, z; t)$ (or $E_B^+(\vec{r}_{\perp}, z; t - \tau)$), is obtained from equation (11) by replacing $\tilde{E}_A^+(\vec{r}_{\perp}; \omega_{2n+1})$ with $E_A^+(\vec{r}_{\perp}; \omega_{2n+1})$ (or $e^{i\omega_{2n+1}t} \tilde{E}_B^+(\vec{r}_{\perp}; \omega_{2n+1})$). The entrance hole of accelerating plates for the measurement of the TOF of ions is placed in front of the focal point, and we neglect the accumulation of the ion yield for the propagation axis because the diameter of the entrance hole is smaller than the confocal parameter of the focused APT.
3.2. Linear interference

Before analyzing the nonlinear IAC, we briefly show how the linear interference affects the ion yield with one-photon absorption, in order to distinguish our interferometer from a conventional one.

The total electric field at $z$, denoted as $E_{\text{tot}}(\vec{r}_\perp, z; t, \tau)$, is the linear sum of $E_A(\vec{r}_\perp, z; t)$ and $E_B(\vec{r}_\perp, z; t - \tau)$, namely,

$$E_{\text{tot}}(\vec{r}_\perp, z; t, \tau) = E_A(\vec{r}_\perp, z; t) + E_B(\vec{r}_\perp, z; t - \tau).$$  \hfill (12)

Although the instantaneous intensity of the total field is proportional to $|E_{\text{tot}}(\vec{r}_\perp, z; t, \tau)|^2$, the observed ion signal with one-photon absorption is proportional to the time and transversal-space integrations of the instantaneous intensity. Thus, the ion yield with one-photon absorption $Y_1(z, \tau)$ should be expressed as

$$Y_1(z, \tau) \propto \int \frac{d^2 \vec{r}_\perp}{T_1} \int_{-T_1/2}^{T_1/2} dt \left[ |E_A(\vec{r}_\perp, z; t)|^2 + |E_B(\vec{r}_\perp, z; t - \tau)|^2 + 2 \text{Re} \left\{ E_A^*(\vec{r}_\perp, z; t) E_B(\vec{r}_\perp, z; t - \tau) \right\} \right].$$  \hfill (13)

By applying equation (11) and utilizing the orthogonal properties of $e^{-i\omega_{n+1}t}$ and $e^{-i(\omega_{n+1}/c)(z-z_0)}\vec{r}_\perp \cdot \vec{r}'_\perp$ in the integrations in terms of $t$ and $\vec{r}_\perp$, the right-hand side of equation (13) is transformed to

$$\sum_{n=n_{\text{min}}}^{n_{\text{max}}} \int d^2 \vec{r}'_\perp \left| \tilde{E}_A'(\vec{r}'_\perp; \omega_{2n+1}) \right|^2 + \sum_{n=n_{\text{min}}}^{n_{\text{max}}} \int d^2 \vec{r}'_\perp \left| \tilde{E}_B'(\vec{r}'_\perp; \omega_{2n+1}) \right|^2 + 2 \sum_{n=n_{\text{min}}}^{n_{\text{max}}} \int d^2 \vec{r}'_\perp \text{Re} \left\{ \tilde{E}_A'^*(\vec{r}'_\perp; \omega_{2n+1}) \tilde{E}_B'(\vec{r}'_\perp; \omega_{2n+1}) e^{i\omega_{n+1}t} \right\}. \hfill (14)$$

Figure 3. Notations of the propagation model. Transverse spatial coordinates on the pupil plane are notated with a prime. Two pupils with a semicircular aperture on the pupil plane are distinguished with subscripts ‘A’ and ‘B’.
The interference of the two fields is expressed in the third term depending on delay \( \tau \), in this equation. The integration in terms of \( \vec{r}_\perp \) is executed on the pupil plane, so that \( \tilde{E}_{\lambda}(\vec{r}_\perp; \omega_{2n+1}) \) times \( \tilde{E}_{\lambda}(\vec{r}_\perp; \omega_{2n+1}) \) should be zero owing to the spatial division. Thus, the interference effect disappears from the ion yield with one-photon absorption, and we have merely confirmed the energy conservation of the electric fields going through an arbitrary plane perpendicular to the propagation axis because equation (14) is independent of \( z \). If we seek the linear interference, we must restrict the region of integration on the focal plane by placing a pinhole or a slit. Power et al demonstrated this type of interferometry using a visible laser pulse [25].

The most crucial result of this basic analysis is that we cannot observe the interference fringes from the product with one-photon absorption. In other words, the interference fringes themselves are the evidence of nonlinear interaction yielding the product we observe.

### 3.3. IAC

First we treat the IAC of an APT without consideration of the spatial effect in order to classify the fringe components contained in the IAC trace, and then introduce the nonlinear spatial profile that affects the fringes.

When we neglect the spatial dependence of an APT field, the electric field is expressed as \( E(t) + E(t - \tau) \) by omitting \( \vec{r}_\perp, z \), and the suffix from electric fields in equation (12). The Fourier component of \( E(t) \) at an angular frequency of \( \omega_{2n+1} \) can be written in the form of \( A_{2n+1} e^{i\phi_{2n+1}} \), where \( A_{2n+1} \) and \( \phi_{2n+1} \) are the real amplitude and the phase of the \( 2n+1 \)st mode (harmonic field), respectively. Thus, \( E(t) \) is simply expressed as [14]

\[
E(t) = \sum_{n=n_{\text{min}}}^{n_{\text{max}}} A_{2n+1} e^{i\phi_{2n+1}} e^{-i\omega_{2n+1}t}.
\]

We assume, in this section, that the transition amplitude accompanying two-photon absorption is proportional to the square of the APT field \( S_{\text{tot}}(t; \tau) \) defined as

\[
S_{\text{tot}}(t; \tau) \equiv |E(t) + E(t - \tau)|^2.
\]

The Fourier amplitude of \( S_{\text{tot}}(t; \tau) \) in terms of \( t \) is useful for finding fringes on the correlation, and can be obtained from the equation of

\[
\tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau) = \frac{1}{T_f} \int_{-T_f/2}^{T_f/2} dt S_{\text{tot}}(t; \tau) e^{i\Omega_{2(N+1)} t},
\]

where \( \Omega_{2(N+1)} \) is defined as \( 2(N+1)\omega_{\text{f}} \) and \( N \) should be restricted to within a range of integers \( 2n_{\text{min}}, 2n_{\text{min}} + 1, \ldots, 2n_{\text{max}} - 1, \) and \( 2n_{\text{max}} \) for a non-vanishing Fourier amplitude of sum frequencies. It is well known that the absolute square of \( \tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau) \) corresponds to the interferometric spectrogram [26, 27] derived from the frequency resolved optical gating (FROG [2]) technique. The autocorrelation trace is obtained by summing \( |\tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau)|^2 \) for all sum frequency modes. Before calculating the right-hand side of equation (17), we define the Fourier amplitude of the correlated part in \( S_{\text{tot}}(t; \tau) \) as

\[
\tilde{T}(\Omega_{2(N+1)}; \tau) \equiv e^{-i\Omega_{N+1} \tau} \frac{1}{T_f} \int_{-T_f/2}^{T_f/2} dt E(t) E(t - \tau) e^{i\Omega_{2(N+1)} t}.
\]

This equivalence shows the definition of \( \tilde{T}(\Omega_{2(N+1)}; \tau) \), in which we remove the principal oscillation with respect to delay \( \tau \) to manifest the slowly varying amplitude in the Fourier

---

New Journal of Physics 10 (2008) 025034 (http://www.njp.org/)
amplitude. The absolute square of $\tilde{T}(\Omega_{2(N+1)}; \tau)$ coincides with the fringe-free spectrogram of a pulse envelope in a FROG measurement, hence we can distinguish the correlation envelope from the interference fringes contained in $|\tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau)|^2$ by using $\tilde{T}(\Omega_{2(N+1)}; \tau)$.

In fact, we obtain $|\tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau)|^2$ as

$$
|\tilde{S}_{\text{tot}}(\Omega_{2(N+1)}; \tau)|^2 = 2 \left| \tilde{T}(\Omega_{2(N+1)}; 0) \right|^2 + 4 \left| \tilde{T}(\Omega_{2(N+1)}; \tau) \right|^2 + 2 \text{Re} \left\{ e^{-i\Omega_{2(N+1)} \tau} \right\} \tilde{T}(\Omega_{2(N+1)}; 0) \right|^2
$$

where we can write $\tilde{T}(\Omega_{2(N+1)}; \tau)$ in the explicit form of

$$
\tilde{T}(\Omega_{2(N+1)}; \tau) = \sum_{n'=-\ell(N)}^{\ell(N)} A_{2((N/2)-n')} e^{i\theta_{2((N/2)-n')}} A_{2((N/2)+n')} e^{i\theta_{2((N/2)+n')}} e^{2i\omega_0' \tau}. \quad (20)
$$

The limit of the running index of $n'$ in equation (20), $\ell(N)$, depends on the magnitude of $N$;

$$
\ell(N) = \begin{cases} 
N/2 - n_{\text{min}} & \text{(for } N \leq n_{\text{min}} + n_{\text{max}}) \\
n_{\text{max}} - N/2 & \text{(for } N > n_{\text{min}} + n_{\text{max}}) 
\end{cases} \quad (21)
$$

and the index $n'$ should be a half-integer when the mode index $N$ is an odd number.

We can confirm from equation (20) that $\tilde{T}(\Omega_{2(N+1)}; \tau)$ corresponds to the correlation amplitude excluding the interference fringes because $n'$ symmetrically runs across 0 and the oscillating term appears as $e^{2i\omega_0' \tau}$. The Fourier amplitudes with spectral phases are convolved with the nature of a Fourier transform. Thus, the five members in the right-hand side of equation (19) are classified into four categories; the first term is the constant background independent of $\tau$ contributed by the incoherent sum of the two electric fields. The second term expresses the correlation envelope that directly reflects the pulse shape or the spectrogram of FROG. The third term yields the interference fringes of the sum frequency mode. We find that the intensity of the sum frequency modulation should be independent of $\tau$. The linear interference fringes originate from the last two terms, the amplitudes of which change relative to $\tau$. We recognize from this analysis that all we have to do for obtaining the IAC of an APT is to calculate $\tilde{T}(\Omega_{2(N+1)}; \tau)$ in equation (20), from which we can also extract the correlation envelope and the interference fringes.

In the following consideration of the spatial effect, the interferometric spectrogram is also calculated and classified in a similar manner. We simplify the condition of the propagation described in equation (11) in order to describe this similarity. The Fourier amplitude of an APT on the pupil plane, which is focused with a length of $f$, should include the phase factor $e^{-i[(f_0+1)/2c]f_0^2}$ according to the para-axial approximation mentioned in section 3.1. We restrict ourselves to considering the electric field at the focal point, which results in $z$ being fixed to $f + z_0$. Thus, we can rewrite equations (2) and (11) to describe the Fourier amplitude of the electric field inside aperture A on the pupil plane as

$$
E_A'(\vec{r}_{1}'; \omega_{2n+1}) = V_{2n+1} e^{i\theta_{2n+1}} e^{-i\omega_{2n+1}f_0^2/(2cf)} P_A(\vec{r}_{1}'; \omega_{2n+1}). \quad (22)
$$
We define the pupil function \( P_A(\vec{r}_\perp'; \omega_{2n+1}) \) describing the truncated electric field with aperture A. The pupil function is normalized such that the maximum of \(|P_A(\vec{r}_\perp'; \omega_{2n+1})|^2\) should be unity at a certain peak position \( \hat{r}_\perp' \). The peak real amplitude and the spectral phase are denoted as \( V_{2n+1} \) and \( \phi_{2n+1} \), respectively. The pupil function may contain the phase factors for a tilted wavefront, an aberration, an astigmatism, and other phase distortions due to the surface roughness of the focusing mirror, although we do not include these phase factors in the numerical calculation described in section 4. The Fourier amplitude of the electric field inside the aperture B with delay \( \tau \), \( \vec{E}_B^\prime(\vec{r}_\perp'; \omega_{2n+1}) \), is derived by replacing A with B in the right-hand side of equation (22) and multiplying with \( e^{i\omega_{2n+1}\tau} \).

Substitution from \( \vec{E}_\text{apt}^+(\vec{r}_\perp'; \omega_{2n+1}) \) to \( \vec{E}_A^+(\vec{r}_\perp'; \omega_{2n+1}) \) or \( \vec{E}_B^+(\vec{r}_\perp'; \omega_{2n+1}) \) in equation (11) leads the electric field at the focal point
\[
E_A(\vec{r}_\perp, f; t) = \frac{i}{f} \sum_{n=n_{\text{min}}}^{n_{\text{max}}} \frac{\omega_{2n+1}}{c} V_{2n+1} e^{i\phi_{2n+1}} \times \exp \left\{ -i\omega_{2n+1} \left( t - \frac{f}{c} - \frac{(\vec{r}_\perp^2)}{2cf} \right) \right\} \vec{P}_A(\vec{k}_{2n+1}; \omega_{2n+1}),
\]
where we set \( z_0 = 0 \) without losing generality. The integration in terms of \( \vec{r}_\perp' \) agrees with the spatial Fourier transform of \( P_A(\vec{r}_\perp'; \omega_{2n+1}) \) because the quadratic phase factor \( e^{-i(\omega_{2n+1}/2cf)(\vec{r}_\perp')^2} \) is eliminated by focusing, resulting in the Fraunhofer diffraction formula. We define the spatial Fourier transform of \( P_A(\vec{r}_\perp'; \omega_{2n+1}) \) as
\[
\vec{P}_A(\vec{k}_{\perp}; \omega_{2n+1}) = \frac{1}{2\pi} \int d^2\vec{r}_\perp' P_A(\vec{r}_\perp'; \omega_{2n+1}) e^{-i\vec{k}_{\perp}\vec{r}_\perp'}. 
\]
The transversal wavevector \( \vec{k}_{2n+1} \), which is parallel to \( \vec{r}_\perp \) on the focal plane, is derived from
\[
\vec{k}_{2n+1} = \frac{\omega_{2n+1}}{cf} \vec{r}_\perp.
\]
Another electric field coming from pupil B with delay \( E_B(\vec{r}_\perp, f; t - \tau) \) is obtained by the substitutions of A with B and \( t \) with \( t - \tau \) in equation (23).

Now, we are ready to calculate the IAC of an APT in a similar manner as that for deriving equation (19). The Fourier amplitude of the square of the total electric field at \( \vec{r}_\perp \) on the focal plane expressed as
\[
\vec{S}_\text{tot}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau) = \frac{1}{T_f} \int_{-T_f/2}^{T_f/2} dt \{ E_A(\vec{r}_\perp, f; t) + E_B(\vec{r}_\perp, f; t - \tau) \}^2 e^{i\Omega_{2(N+1)}t}.
\]

We assume again that the probability amplitude of finding an ion produced with two-photon absorption, the total photon energy of which is \( \hbar \Omega_{2(N+1)} \), at position \( \vec{r}_\perp \) on the focal plane is proportional to \( \vec{S}_\text{tot}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau) \). Thus, the absolute square of \( \vec{S}_\text{tot}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau) \) corresponds to the spatial probability distribution of the ion yield, for which two photons with a total energy of \( \hbar \Omega_{2(N+1)} \) are used, on the focal plane. We observe the spatial integration of the ion yield in the actual experiment, so the interferometric spectrogram should be obtained with
\[
\int d^2 \vec{r}_\perp |\vec{S}_\text{tot}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau)|^2.
\]
To calculate the right-hand side of equation (26), it is convenient to use the Fourier transform of the correlated part of the two APT fields, that is, 
\[ \tilde{T}_{AB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; \tau) \]
\[ = -e^{-i\Omega_{2(N+1)}\tau} e^{-i\Omega_{2(N+1)}\vec{r}_\perp^2/(2cf)} \frac{1}{T_i} \int_{-T_i/2}^{T_i/2} dt E_A(\vec{r}_\perp, f ; t) E_B(\vec{r}_\perp, f ; t - \tau) e^{i\Omega_{2(N+1)} t} \]
\[ = f^{-2} \sum_{n'=-\ell(N)}^{\ell(N)} U_{2((N/2) - n')1} e^{i\phi_{2((N/2) - n')1}} U_{2((N/2) + n')1} e^{i\phi_{2((N/2) + n')1}} e^{2i\nu n' \omega_0 t} \]
\[ \times \tilde{P}_A(\vec{k}_{2((N/2) - n')1} ; \omega_{2((N/2) - n')1}) \tilde{P}_B(\vec{k}_{2((N/2) + n')1} ; \omega_{2((N/2) + n')1}), \tag{27} \]
where we define the modified field amplitude \( U_{2n+1} \equiv (\omega_{2n+1}/c) V_{2n+1} \). We note, upon comparing equation (27) with (20), that the spatial modes of \( \tilde{P}_A(\vec{k}_{2n+1} ; \omega_{2n+1}) \) and \( \tilde{P}_B(\vec{k}_{2n+1} ; \omega_{2n+1}) \) in the frequency domain are convolved in addition to the convolution of the amplitudes of \( U_{2n+1} \). Thus, in order for the correlation to appear, how the electric fields are spatially evolved into the focal plane is important.

The probability distribution in the two photon mode \( \Omega_{2(N+1)} \), which we call the nonlinear spatial profile, can be written as
\[ \left| \tilde{S}_{\text{we}}(\vec{r}_\perp, f ; \Omega_{2(N+1)}) \right|^2 = \left| \tilde{T}_{AA}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) \right|^2 + \left| \tilde{T}_{BB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) \right|^2 + 4 \left| \tilde{T}_{AB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; \tau) \right|^2 \]
\[ + 2 \text{Re} \left\{ \tilde{T}_{AA}^*(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) \tilde{T}_{BB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) e^{-i\Omega_{2(N+1)} \tau} \right\} \]
\[ + 4 \text{Re} \left\{ \tilde{T}_{AA}^*(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) \tilde{T}_{AB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; \tau) e^{i\Omega_{2(N+1)} \tau} \right\} \]
\[ + 4 \text{Re} \left\{ \tilde{T}_{BB}^*(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; 0) \tilde{T}_{AB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; \tau) e^{-i\Omega_{2(N+1)} \tau} \right\}, \tag{28} \]
which is very similar to equation (19) except for the space dependence in all the terms. Thus, the classification is in accordance with that for equation (19): (i) the constant backgrounds: the first and second terms, (ii) the correlation envelope: the third term, (iii) interferometric fringes of sum frequency mode: the fourth term, and (iv) interferometric fringes of the linear frequency mode: the fifth and sixth terms. Thus, we can analytically extract the correlation envelope without averaging the fringes [28].

The calculation of \( \tilde{T}_{AB}(\vec{r}_\perp, f ; \Omega_{2(N+1)} ; \tau) \) according to equation (27) leads to the spectrogram or correlation trace. This is an advantage for the numerical calculation because we do not need any commercial software to calculate ray traces [29] nor the accurate but time-consuming integration of the rigorous wave equation [30]. All we have to do is merely the 2D Fourier transform, which can be executed quickly with the well-established algorithm even by non-specialists in numerical calculation like us.

4. Result

We show the calculated results of the spectrograms and the traces of the IAC of APTs in this section. We must, however, give the above-mentioned formulae appropriate parameters in
accordance with the experimental conditions before carrying out the calculation. The aperture radius and the focal length are 1 and 100 mm, respectively, which are the same as those in the experiment. The spatial intensity profiles of the harmonic fields are all assumed to be the same as the Gaussian profile with a radius of 1 mm at half-maximum and the spatial phases are fixed at 0. These assumptions are reasonable for the measured spatial profile of each harmonic field using an XUV spectrograph. The spatial profiles are truncated with the aperture and divided into two pupil functions of $P_A(\vec{r}'; \omega_{2n+1})$ and $P_B(\vec{r}'; \omega_{2n+1})$. We insert a narrow blank area of a constant width of 200 $\mu$m between these two pupils, although we did not measure the gap between the two harmonic separators in the experiment. We fix the spectral phases $\phi_{2n+1}$ to be 0 to calculate the IAC of the Fourier limit APT.

The remaining unknown parameter is $V_{2n+1}$, which should be determined by observing the harmonic spectra. It is, however, not easy to specify simultaneously the relative intensities of all the harmonic fields because the frequency range is so wide that it exceeds the calibrated range of the optics and detectors. The quantum efficiency of a micro-channel plate (MCP) for the 5th-order harmonic field, for example, can significantly change depending on the environmental conditions and that of an x-ray CCD camera in the VUV region may be significantly modulated [31]. Furthermore, we know that the reflectivity of the SiC mirror is different from the measured one [21]. Thus, the intensity distribution shown in figure 4 is obtained from reasonable choices by making the harmonic spectra observed using the MCP and using the CCD camera in the XUV spectrograph. We use the square roots of these intensities for $V_{2n+1}$.

In figure 5, we first demonstrate, in a movie, the change in the linear intensity profiles of some harmonic fields depending on delay. The linear profile is calculated by eliminating the spatial integration from equation (13). We note that, in this movie, the cross-section of the focused beam differs notably depending on the harmonic frequency, while the spatial fringe due to the interference moves upward in every profile with a harmonic period.

We can recognize how these spatial distributions affect the appearance of the interferometric fringes in the IAC by considering that the nonlinear intensity profiles arise from the convolution between the linear amplitude profiles. In figure 6, we show an example of the graphical explanation of equation (28) to obtain the nonlinear spatial profile. The nonlinear spatial profile at the 28th mode, which is depicted on the right-hand side in this figure, originates from the products of two linear profiles, of which the sum number of their harmonic orders is 28 (the 9th and 19th, the 11th and 17th, and the 13th and 15th orders). We can see that the
large cross-section of the low order-harmonic beam is truncated with the small cross-section of the high-order harmonic beam by a multiplication of the linear profiles shown in each red parentheses, so that the deeper modulation of the interference fringe is expected for the lower order mode frequency.

Through calculations similar to those carried out for obtaining M28 in figure 6, we can obtain the nonlinear spatial profiles ranging from the 10th mode to the 38th mode. Four of these profiles are shown as a movie in figure 7. It is natural in this movie that the spatial fringe disappears and the peak intensity is maximized in all the profiles at a delay equal to 0, because two APTs coming from pupils A and B coincide at the focal position both for their envelopes and phases. At a delay equal to $\pm T_f/2$, however, each of the nonlinear spatial profiles splits vertically with the interference fringe appearing at the center, and the peak intensity is notably reduced. This is due to the distractive interference in every harmonic field forming two APTs with a phase difference of $\pi$, while the two APT envelopes are temporally overlapped. These spatial properties are the origins of the characteristic of the interference fringes on the top of the correlation envelope observed in figure 2(c).

After the spatial integration of the nonlinear profiles, we obtain the interferometric spectrogram shown in figure 8(a). The calculated delay range of $[-T_f/2, T_f/2]$ is sufficient for expressing the entire APT because we neglect the bandwidth of each harmonic field and the complete periodicity of $T_f$. Clear evidence of APT formation can be seen as a bunch with a period of $T_f/2$ on the correlation trace of each mode. The interferometric fringes, of
Figure 6. Graphical interpretation of equation (28). The nonlinear spatial profile at the 28th mode ($\Omega_{28}, N = 13$) is composed of three multiples of the two linear spatial profiles, the summed harmonic orders of which should be equal to 28. We show the intensity profiles in this figure for graphical convenience, although the complex amplitudes are convolved in the actual calculation. The color scale of each profile is adjusted such that the profile can be clearly seen as indicated by yellow numbers.

which periods gradually decrease with an ascending two-photon mode, also appear both on the correlation peak and in the background on this spectrogram.

We find a dip due to the interference fringes at the top of the correlation envelope of each mode at delays of $\pm T_f/2$ by comparison with the non-interferometric spectrogram, which is calculated from $\int d^2\vec{r}_\perp |\tilde{T}_{AB}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau)|^2$ in equation (28), shown in figure 9. In other words, we cannot discriminate the correlation envelopes at $\pm T_f/2$ from that at 0 delay without the interference fringe.

The ideal trace of the IAC should be derived from the sum of all the modes in figure 9, as shown in figure 10(a). We can see a significant difference in this trace from the conventional IAC trace shown in figure 10(b), which is calculated from equation (19). It is well known that the peak-to-background ratio of the IAC trace of the Fourier-limit pulse, obtained by a conventional IAC measurement, should be 8:1, and the intensity of the fringe dip near zero delay must be equal to 0. The latter characteristics can be confirmed by setting $\phi_{2((N/2)+n')1} = 0$ and $\tau = \pm T_f/2$ in equation (19). This is due to the complete destructive interference in a fictitious interferometer in which a half mirror splits the measured APT field into two replicas, and the energy flow of the
Figure 7. Available from stacks.iop.org/NJP/10/025034/mmedia. Movie of the nonlinear spatial profiles of the 16th-, 22th-, 28th- and 34th-order modes at the focal plane. The center clock indicates delay in the unit of the optical period of the fundamental laser field $T_f$. The color scale of each profile is adjusted such that the profile can be clearly seen, as indicated by yellow numbers.

Figure 8. (a) Interferometric spectrogram of the Fourier-limit APT calculated from $\int d^2\vec{r}_\perp|\tilde{S}_{\text{tot}}(\vec{r}_\perp, f; \Omega_{2(N+1)}; \tau)|^2$. We assume the Fourier limit in this calculation. (b) Interferometric autocorrelation trace derived from the sum of modes higher than or equal to the 28th mode.
two replica APTs should escape from another arm of the interferometer under the destructive interference condition. In our autocorrelation measurement with the spatial split of the measured APT field, on the other hand, the two replicas of the APT field always exist in the interaction region for ionization, so that the intensity of the correlation trace never falls to zero even when the delay corresponds to $\pm T_f/2$. We can also see that the peak-to-background ratio of the IAC trace in figure 10(a) is lower than that of the IAC trace in figure 10(b), even though we do not analytically determine it. The discrepancy between the peak-to-background ratio of the experimental data and the calculated result will be discussed in the next section.

In the actual experiment, the measured autocorrelation signal is contributed from the limited two-photon modes, of which energy exceeds the double ionization threshold of a nitrogen molecule. Thus, the autocorrelation trace to be compared with the experimental result should be the sum of the modes higher than or equal to the 28th two-photon mode [32]. These modes for the correlation trace lie within an area between the red dashed lines indicated in

**Figure 9.** (a) Non-interferometric spectrogram of the Fourier-limit APT calculated from $\int d^2\vec{r}_{\perp} |\tilde{T}_{AB}(\vec{r}_{\perp}, f; \Omega_2(N+1); \tau)|^2$. (b) Autocorrelation trace derived from the sum of modes higher than or equal to the 28th mode.

**Figure 10.** (a) IAC trace obtained from the sum of all modes in the spectrogram in figure 8(a). (b) Conventional IAC trace calculated from equation (19).
Figure 11. Upper part: APT field calculated from the spectrum shown in figure 4 in the Fourier limit. The carrier envelope phase is fixed at 0. Lower part: APT intensity envelope (blue hatched areas) and its autocorrelation trace (pink dotted curve) calculated from the intensity envelope in the Fourier limit.

We find that the full width at half-maximum based on the background of the correlation envelope in figure 9(b) is 380 attoseconds while the correlation width of the Fourier-limit pulse with a duration of 185 attoseconds is estimated to be 262 attoseconds without regarding the spatial profile, as shown in figure 11. An unusually large correlation width is not induced with the restriction of the two-photon modes because the non-IAC trace including all the two-photon modes is not appreciably different from the trace in figure 9(b), as demonstrated in the next section. Thus, it may be suggested from this comparison that the pulse width should be effectively broadened because of the different size of the cross-section for each harmonic field and the spatial integration. This is one of the candidates for answering the inquiry concerning the pulse width of the focused APT field presented in section 1.

5. Discussion

We cannot determine the chirp between the separate harmonic fields with the IAC measurement in principle, so that another technique would be required for the complete characterization of an APT. In the photoelectron analysis with non-resonant two-photon ionization for harmonic electric-field reconstruction (PANTHER) [33] utilizing the two-photon ATI [34, 35] of an argon atom, we found that the chirp among the three harmonic fields at the 11th, 13th and 15th orders is $1.3 \times 10^{-32}$ s$^2$. The IAC trace calculated with this chirp taken into consideration is compared with the experimental IAC trace in figure 12. The two traces in figure 12 appear similar, although we should note some disagreements to be considered. The first one is the difference in the backgrounds of these traces. We show the two traces with the common zero signal. Thus, we can see that the peak-to-background ratio of the measured trace is significantly lower than that of the calculated trace. This may be mainly due to the contribution of the single-photon double ionization to the background of the measured trace. The intensities of the harmonic fields at orders higher than the 27th order are lower than the detection limit of the XUV spectrograph because these harmonic fields lie in the cut-off region of the harmonic generation in a Xe gas target. However, these harmonic fields can still remain such that the number of doubly charged nitrogen molecules absorbing a single photon of these harmonics should be comparable to that absorbing two photons of the APT field. In fact, we observed electron yield with single-photon absorption of the 21st-order harmonic field in the PANTHER experiment even though the use
of a Sn filter should have eliminated this field [33]. The degradation of the peak-to-background ratio is also caused by the unbalanced split of the APT field as a result of the pointing instabilities of the driving laser and the generated harmonic fields. We have not yet, however, studied this effect in our calculation model.

The second issue may be related not to the discrepancy between the two traces but to the correlation trace itself arising from the restricted bandwidth. We collect only the two-photon modes higher than the 28th mode in both the experiment and the calculation. Does this high-pass filtering affect the autocorrelation? The answer is, of course, yes, but only a little. We compare the high-pass-filtered interferometric and non-IAC traces, which are the same as those shown in figures 8(b) and 9(b), with those including the full bandwidth, in figures 13(a) and (b). The full autocorrelations of the interferometric and non-interferometric traces, shown as light-blue dashed curves, are both similar to the high-pass-filtered autocorrelation traces (brown solid curves) except for the somewhat different visibility of the interference fringes in figure 13(a). These correspondences may result from the fact that the 28th and higher modes contain the contributions from most of the harmonic fields. The relative intensities of the 5th- and 7th-order harmonic fields excluded from the high-pass-filtered correlation should be lower than those shown in figure 4 at the focal position, owing to the cross-sections being larger than those of the higher-order harmonic fields, so that these two harmonic fields do not notably change the correlation trace. We conclude that the measured autocorrelation trace reveals at least the major characteristic of the APT formed by Fourier synthesis of the 9th- and higher-order harmonic fields.
The third issue is related to the principal assumption for the autocorrelation shown in equation (16). The transition amplitude with two-photon absorption is not simply proportional to the square of the electric field, even when the transition occurs non-resonantly. We should always keep in mind that equation (16) is only an approximation. In the context of the time-dependent second-order perturbation theory, the transition amplitude of an electron absorbing two photons $a_{lg}$ per unit time should be expressed as

$$
\frac{a_{lg}}{T_o} = (i\hbar)^{-1}T_o^{-1}\int_{-T_o/2}^{T_o/2}dt_1\int_{-T_o/2}^{T_o/2}dt_2\sum_m\mu_{lm}\mu_{mg}E(t_1)E(t_2)e^{i\omega_{m+n}t_1}e^{i\omega_{m}t_2},
$$

where $T_o$ is defined as $T_o \equiv (2M+1)T$ with a sufficiently large positive integer $M$. Although we need a quantum system containing two electrons interacting with each other to describe the double ionization, we adopt the single-electron system to simplify the problem. Note that this analysis can be applied to the two-photon ATI within the single active electron approximation utilizing the effective potential of a rare gas atom [36]. We note transition amplitudes from the initial state $|g\rangle$ to an intermediate state $|m\rangle$ and from the intermediate state to the final state $|f\rangle$ as $\mu_{mg} \equiv \langle m|\mu|g\rangle$ and $\mu_{fm} \equiv \langle f|\mu|m\rangle$, respectively. We adopt a dipole approximation of the interaction between the electron and the electric field and assume that the electric field commute with a dipole operator $\hat{\mu}$ because the wavelength of the APT is still much longer than the atomic scale even though it is in the range of XUV. Thus, we can neglect the space dependence of the electric field to investigate a single-electron response as in the theory of high-order harmonic generation [22]. The index of the intermediate state $m$ may be discrete or continuous depending on whether the electron is bound or bound-free, so that the symbol $\sum_m$ denotes the summation of the discrete index and the integration of the continuous variable. The eigenenergy of each state is notated as $\hbar\omega_o$ and the energy difference is defined as $\hbar\omega_{a\beta} \equiv \hbar\omega_{a\alpha} - \hbar\omega_{b\beta}$, where the suffixes $\alpha$ and $\beta$ can be $g$, $f$ or $m$.

By substituting equation (15) into (29) we find that the transition amplitude can be written in the form of

$$
\frac{4a_{lg}}{T_o} = \sum_{n_{\text{min}}=n_{\text{min}}}^{n_{\text{max}}=n_{\text{max}}}\sum_{n_{\text{min}}'=n_{\text{min}}}^{n_{\text{max}}'=n_{\text{max}}}\Theta(\Omega_{2,(N+1)}; \omega_{2n+1}, \omega_{2n'+1})A_{2n+1}A_{2n'+1}e^{i\phi_{2n+1}}e^{i\phi_{2n'+1}},
$$

where we define the response function $\Theta(\Omega_{2,(N+1)}; \omega_{2n+1}, \omega_{2n'+1})$ as

$$
\Theta(\Omega_{2,(N+1)}; \omega_{2n+1}, \omega_{2n'+1}) \equiv \hbar^{-2}\delta_{N(n'+n)}\left[\sum_m\frac{\mu_{lm}\mu_{mg}}{(\omega_m-\omega_b)-(\omega_{2n'+1})}\delta_{mm'}+2T_o^{-1}\delta_{nn'}\right] + \frac{1}{\pi} \int d\omega \rho(\omega) \sum_\ell \frac{\mu_\ell(\omega; \omega_b)\mu_\ell(\omega; \omega_g)}{(\omega-\omega_g)-(\omega_{2n'+1}-\epsilon i)}.
$$

The dipole matrix element from/to a continuous energy state is written as $\mu_\ell(\omega; \omega_b)/\mu_\ell(\omega; \omega_g)$ with other discrete quantum numbers $\ell$ for other degrees of freedom such as an angular momentum in equation (31). We insert the density of states $\rho(\omega)$ for consistency. The positive infinitesimal $\epsilon$ avoiding poles on the real axis results from the causality in the double integration of time. The indices of $m_r$ and $m_{nr}$ represent the resonant states satisfying $\omega_m-\omega_b = \omega_{2n'+1}$ and other states, respectively.

We can recognize from equation (31) that equation (30) is reduced to $\bar{T}(\Omega_{2,(N+1)}; \tau = 0)$ in equation (20) when the quantities in the square brackets on the right-hand side in equation (31)
Figure 14. IAC traces. Experimental result is shown by crosses and brown lines, while the calculated trace with a response function is depicted by the violet solid curve.

do not significantly change in terms of \( \omega_{2n'+1} \) (or equivalently \( \omega_{2n+1} = \Omega_2(N+1) - \omega_{2n'+1} \)) and can be regarded as constants. This is the condition that we have assumed in order to calculate the IAC of an APT. In fact, the transition amplitude depends on the angular frequency of the APT field, so that the amplitude of the correlated part described as equation (27) should be rewritten as

\[
\tilde{T}_{AB}(\vec{r}, f; \Omega_2(N+1); \tau) \propto \sum_{n'=\ell(N)}^{\ell(N)} U_{2((N/2)-n'+1)} U_{2((N/2)+n'+1)} e^{\imath \phi_2((N/2)-n'+1)} e^{\imath \phi_2((N/2)+n'+1)} \times \Theta(\Omega_2(N+1); \omega_{2((N/2)-n'+1)}, \omega_{2((N/2)+n'+1)}; \omega_{2n+1}, \omega_{2n'+1}) P_B(\vec{k}_{2((N/2)+n'+1)}, \omega_{2((N/2)+n'+1)})
\]

using the response function \( \Theta(\Omega_2(N+1); \omega_{2n+1}, \omega_{2n'+1}) \). This equation is justified by replacing \( E(t_i) \) with \( E(t_i) + E(t_i - \tau) \) \((i = 1, 2)\) in equation (29) and extracting the correlated part. The macroscopic space dependence of the electric field is discriminated in the calculation of the transition amplitude while the spatial distribution of the transition amplitude itself is included in this model. This approximated description is similar to that used to analyze the propagation effect, or in more specific terms, the phase matching of the high-order harmonic field [37, 38], in which the source of the nonlinear polarization is treated as being induced from the space-free electric field before introducing the spatial dependence of the nonlinear dipole phase due to the intensity distribution of the driving laser field.

The remaining issue is how we obtain the specific form of the response function. It is, however, not easy to model the two-photon double ionization (TPDI) in a simple form owing to the electron–electron correlation, and this issue is far beyond the scope of this paper. Actually, even the TPDI of the simplest atom, namely, helium, is still a developing topic in atomic physics [39]–[47]. Thus, we give up finding the accurate form of \( \Theta(\Omega_2(N+1); \omega_{2n+1}, \omega_{2n'+1}) \) and present a toy model instead:

\[
\Theta_{\text{toy}}(\Omega_2(N+1); \omega_{2n+1}, \omega_{2n'+1}) \sim \hbar^{-2} \delta_{N(n+n')} \langle \mu^2 \rangle \left[ \frac{1}{\omega_R - \omega_{2n'+1} - \imath \Gamma} + \frac{1}{\omega_R - \omega_{2n+1} - \imath \Gamma} \right],
\]

which contains a single resonance frequency of \( \omega_R \) and the resonance width of \( \Gamma \). We set the square of an effective dipole as \( \langle \mu^2 \rangle \).

The ionization energy for a singly charged ion is most likely to correspond to \( \hbar \omega_R \) in the double-ionization process even though there is no evidence. We have tried to elucidate how this type of response function distorts the autocorrelation trace by substituting the ionization energy
of a nitrogen molecule (15.58 eV) as $\hbar \omega_R$. The width of the resonance is set to 0.1 eV. There is insufficient reason why $\Gamma$ should be 0.1 eV, while the Rydberg series of highly excited states near the ionization threshold may be contained in this resonance width. The autocorrelation trace obtained with this response model and a given chirp is shown as the violet curve in figure 14. Note that the two photon modes are summed from the 26th mode, instead of the 28th mode, to the 38th mode to make the violet trace better fit the experimental results. The trace is more similar to the experimental trace than to the calculated trace in figure 12. In particular, the interferometric fringes in the background of the correlation envelope exhibits the near-resonance characteristic for the 11th-order harmonic field. Nikolopoulos et al [28] already found that the interferometric fringes with a period of near-resonant harmonic field emerge in the background when they realized the IAC of an APT with a conventional Michelson interferometer in the XUV region [48]. They did not, however, observe the interference fringes in the correlation trace of their measured APT [30].

The similarity between the experimental and calculated IAC traces appears more clearly in their Fourier transform. We show the magnitude squares of the Fourier transforms of the autocorrelation traces obtained from the experiment (brown curve), the calculation without a response function (dark-blue curve), and the calculation with $\Theta_{\nu n}(\omega_{2(n+1)}; \omega_{2n+1}, \omega_{2n'+1})$ in figure 15. The frequency of the spectrum is normalized with the carrier frequency of the fundamental laser field so that the numerics on the horizontal axis directly represent the harmonic orders or the two-photon modes. The main feature of the interferometric fringe components at odd orders in the experimental Fourier transform essentially agrees with that of the violet curve. We can conclude that the distinguished peak at the 11th frequency originates from some kind of resonance because this peak at this frequency in the dark-blue curve is much lower than the next (13th) peak. We note that the similar analyses with Fourier transforms in the previous papers [17, 18] may be corrected by using the more accurate (but still somewhat obscure, please refer to the second paragraph of section 4 describing the difficulty in determining widely spanned harmonic fields) spectrum shown in figure 4. We convince ourselves, on the basis of this calculated trace, that equation (32) can potentially reproduce the IAC of an APT accompanying any type of two-photon interaction. We need, however, to know more details of the form of $\Theta(\Omega_{2(N+1)}; \omega_{2n+1}, \omega_{2n'+1})$ by considering elemental interactions of electrons with the APT fields in order to recognize the IAC.
6. Summary

We formulated the propagation and the focusing of an APT in a simple and feasible manner. On the basis of this formulation and an easy assumption on the two-photon interaction, we have clarified the origin of the interference fringes on the IAC trace obtained with our spatial split interferometer. The spatial interference on the linear profile does not yield the fringe on the trace, but the interference fringes on the nonlinear spatial profile generates the fringes even after the spatial integration. The non-IAC trace, which is analytically expressed as a correlated part of the IAC trace, suggests that the effective pulse width of the APT at the focus may be longer than that expected from the spectrum. A possible correction of the IAC trace has been given in line with the manner of the time-dependent perturbation theory. A toy model of the response function reproduced the IAC trace relatively well, although we could not find an accurate description of this function. Our hope is that someone specializing in atomic/molecular physics will solve this problem.

References

[1] Ippen E P and Shank C V 1977 Ultrashort Light Pulses: Picosecond Techniques and Applications (Berlin: Springer) p 83
[2] Kane D J and Trebino R 1993 Characterization of arbitrary femtosecond pulses using frequency resolved optical gating IEEE J. Quantum Electron. 29 571–9
[3] Hentschel M et al 2001 Attosecond metrology Nature 414 509–13
[4] Drescher M et al 2001 X-ray pulses approaching the attosecond frontier Science 291 1923–7
[5] Baltuška A et al 2003 Attosecond control of electronic processes by intense light fields Nature 421 611–5
[6] Kienberger R et al 2001 Atomic transient recorder Nature 427 817–21
[7] Sansone G et al 2006 Isolated single-cycle attosecond pulses Science 314 443–6
[8] Mairesse Y and Quéré F 2005 Frequency-resolved optical gating for complete reconstruction of attosecond bursts Phys. Rev. A 71 011401
[9] Paul P M et al 2001 Observation of a train of attosecond pulses from high harmonic generation Science 292 1689–92
[10] Muller H G 2002 Reconstruction of attosecond harmonic beating by interference of two-photon transitions Appl. Phys. B 74(suppl.) S17–21
[11] Glover T E, Schoenlein R W, Chin A H and Shank C V 1996 Observation of laser assisted photoelectric effect and femtosecond high order harmonic radiation Phys. Rev. Lett. 76 2468–71
[12] Schins J M et al 1996 Cross-correlation measurement of femtosecond extreme-ultraviolet high-order harmonics J. Opt. Soc. Am. B 13 197–200
[13] Toma E S and Muller H G 2002 Calculation of matrix elements for mixed extreme-ultraviolet infrared two-photon above-threshold ionization of argon J. Phys. B: At. Mol. Opt. Phys. 35 3435–42
[14] Antoine P, L’Huillier A and Lewenstein M 1996 Attosecond pulse trains using high-order harmonics Phys. Rev. Lett. 77 1234–7
[15] Mairesse Y et al 2003 Attosecond synchronization of high-harmonic soft x-rays Science 302 1540–3
[16] López-Martens R et al 2005 Amplitude and phase control of attosecond pulse train Phys. Rev. Lett. 94 033001
[17] Nabekawa Y et al 2006 Interferometric autocorrelation of an attosecond pulse train in the single-cycle regime Phys. Rev. Lett. 97 153904
[18] Shimizu T et al 2007 Observation and analysis of an interferometric autocorrelation trace of an attosecond pulse train Phys. Rev. A 75 033817
[19] Takahashi E J, Nabekawa Y and Midorikawa K 2002 Generation of 10-µJ coherent extreme-ultraviolet light by use of high-order harmonics Opt. Lett. 27 1920–2

New Journal of Physics 10 (2008) 025034 (http://www.njp.org/)
Takahashi E J, Nabekawa Y, Mashiko H, Hasegawa H, Suda A and Midorikawa K 2004 Generation of strong optical field in soft x-ray region by using high-order harmonics *IEEE J. Select. Top. Quantum Electron.* **10** 1315–28

Takahashi E J, Hasegawa H, Nabekawa Y and Midorikawa K 2004 High-throughput, high-damage-threshold broadband beam splitter for high-order harmonics in the extreme ultraviolet region *Opt. Lett.* **29** 507–9

Lewenstein M, Balcou P, Ivanov M Y, L’Huillier A and Corkum P B 1994 Theory of high-harmonic generation by low-frequency laser fields *Phys. Rev. A* **49** 2117–32

Brabec T and Krausz F 1997 Nonlinear optical pulse propagation in the single-cycle regime *Phys. Rev. Lett.* **78** 3282–5

Namiki M 1982 Delta function and differential equations *Iwanami* (in Japanese)

Power E, Pentland J, Nees J, Hauri C P, Merano M, Lopez-Martens R and Mourou G 2006 All-reflective high fringe contrast autocorrelator for measurement of ultrabroadband optical pulses *Opt. Lett.* **31** 3514–6

Amat-Roldán I, Cormack I G, Loza-Alvarez P and Artigas D 2005 Measurement of electric field by interferometric spectral trace observation *Opt. Lett.* **30** 1063–5

Amat-Roldán I, Artigas D, Cormack I G and Loza-Alvarez P 2006 Simultaneous analytical characterisation of two ultrashort laser pulses using spectrally resolved interferometric correlations *Opt. Express* **14** 4538–51

Nikolooulos L A A, Benis E P, Tzallas P, Charalambidis D, Witte K and Tsakiris G D 2005 Second order autocorrelation of an XUV attosecond pulse train *Phys. Rev. Lett.* **94** 113905

Fuchs U, Zeitner U D and Tünnermann A 2005 Ultra-short pulse propagation in complex optical systems *Opt. Express* **13** 3852–61

Tzallas P, Charalambidis D, Papadogiannis N A, Witt K and Tsakiris G D 2003 Direct observation of attosecond bunching *Nature* **426** 267–71

Garnir H P and Lefèbvre P H 2005 Quantum efficiency of back-illuminated CCD detectors in the VUV region (30–200 nm) *Nucl. Instrum. Methods* B **235** 530–4

Okino T et al 2006 Attosecond molecular Coulomb explosion *Chem. Phys. Lett.* **432** 68–73

Nabekawa Y et al 2006 Conclusive evidence of an attosecond pulse train observed with the mode-resolved autocorrelation technique *Phys. Rev. Lett.* **96** 083901

Miyamoto N et al 2004 Observation of two-photon above-threshold ionization of rare gases by xuv harmonic photons *Phys. Rev. Lett.* **93** 083903

Furusawa K et al 2006 Photoelectron spectroscopy of two-photon ionisation of rare-gas atoms by multiple high order harmonics *Appl. Phys. B* **83** 203–11

Toma E S and Muller H G 2002 Calculation of matrix elements for mixed extreme-ultraviolet-infrared two-photon above-threshold ionization of argon *J. Phys. B: At. Mol. Opt. Phys.* **35** 3435–42

L’Huillier A, Li X F and Lompré L A 1990 Propagation effects in high-order harmonic generation in rare gases *J. Opt. Soc. Am. B* **7** 527–36

Balcou P, Salières P, L’Huillier A and Lewenstein M 1997 Generalized phase-matching conditions for high harmonics: the role of field-gradient forces *Phys. Rev. A* **55** 3204–10

Kornberg M A and Lambropoulos P 1999 Photoelectron energy spectrum in ‘direct’ two-photon double ionization of helium *J. Phys. B: At. Mol. Opt. Phys.* **32** L603–13

Nikolooulos L A A and Lambropoulos P 2001 Multichannel theory of two-photon single and double ionization of helium *J. Phys. B: At. Mol. Opt. Phys.* **34** 545–64

Nakajima T and Nikolooulos L A A 2002 Use of helium double ionization for autocorrelation of an XUV pulse *Phys. Rev. A* **36** 041402

Feng L and van der Hart H 2003 Two-photon double ionization of He *J. Phys. B: At. Mol. Opt. Phys.* **36** L1–7

Nabekawa Y, Hasegawa H, Takahashi E J and Midorikawa K 2005 Production of doubly charged helium ions by two-photon absorption of an intense sub-10-fs soft X-ray pulse at 42 eV photon energy *Phys. Rev. Lett.* **94** 043001

Hasegawa H, Takahashi E J, Nabekawa Y, Ishikawa K L and Midorikawa K 2005 Multiphoton ionization of He by using intense high-order harmonics in the soft-x-ray region *Phys. Rev. A* **71** 023407
[45] Foumouo E, Kamta G L, Edah G and Piraux B 2006 Theory of multiphoton single and double ionization of two-electron atomic systems driven by short-wavelength electric fields: An \textit{ab initio} treatment \textit{Phys. Rev. A} \textbf{94} 063409

[46] Horner D A, Morales F, Rescigno T N, Martín F and McCurdy C W 2007 Two-photon double ionization of helium above and below the threshold for sequential ionization \textit{Phys. Rev. A} \textbf{76} 030701

[47] Nikolopoulos L A A and Lambropoulos P 2007 Time-dependent theory of double ionization of helium under XUV radiation \textit{J. Phys. B: At. Mol. Opt. Phys.} \textbf{40} 1347–57

[48] Goulielmakis E, Nersisyan G, Papadogiannis N A, Charalambidis D, Tsakiris G D and Witte K 2002 A dispersionless Michelson interferometer for the characterization of attosecond pulses \textit{Appl. Phys. B} \textbf{74} 197–206