

Attosecond streaking measurements

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Abstract. A detailed analysis of the attosecond streaking method is performed. For a class of measurements a consistent quantum theory of streaking is put forward, which allows a close analogy with the intuitive classical picture. In particular, an ‘electron release distribution’ in time and momentum can be defined. The mathematical and practical limitations of recovery of the release distribution from a limited set of measurements are discussed. Recovery procedures for monotonic and oscillatory streak fields are described. A simple formula for time-resolution is derived and confirmed by numerical experiments.

1. Introduction

Attosecond measurements can give us unprecedented insight into the dynamics of valence shell electrons in atoms and small molecules, because currently accessible time scales match the typical time scales of electronic motion. The measurements are made possible by the rapid developments of (a) fully controlled few cycle laser pulses [1–3], (b) extreme ultraviolet (XUV) pulse trains with durations of the individual peaks down to 130 attoseconds [4, 5], and single XUV pulses with currently a minimum duration of 250 attoseconds [6, 7], which are (c) time-locked to each other on the attosecond time scale [5, 7]. A set of pioneering experiments have demonstrated that the new tools can be employed for the controlled modification of photoelectron spectra [8], measurement of electronic relaxation [9] and for XUV pulse diagnostics [7].

At the present time, most attosecond measurements are based on the ‘streak camera’ principle [10–12], which, in a generalized version, was dubbed the ‘atomic transient recorder’ [7]. The technique has an intuitive interpretation in terms of classical mechanics, but for a correct interpretation of observed spectra and the underlying electronic dynamics as well as for analysis of the achievable time-resolution a consistent quantum mechanical description of the complete time-resolved measurement is needed. In the present contribution we make a few steps in that direction. After a layout of the measurement principle in terms of classical mechanics we construct a quantum theory of streaking measurement, which is applicable to a wide class of experiments. Within this theory, we point

out which dynamical information can be maximally extracted from a streaking measurement and where exactly it exceeds information obtainable from time-independent measurements. An analysis of the time-resolution of the method is made by a simple intuitive argument, which is confirmed by quantum mechanical simulations.

2. The streaking principle

Suppose an electron, treated as a classical particle, is released from an atom at a time t with an initial momentum $\mathbf{p}_i$ in the presence of a laser pulse with electric field $\mathcal{E}(t)$. Between its release and detection the electron experiences a boost of

$$\Delta \mathbf{p} = e \int_t^\infty \mathcal{E}(t') dt'. \quad (1)$$

Here e denotes the electron charge ($e < 0$). For convenience, we define the integral as $\mathbf{A}(t) = e \int_t^\infty \mathcal{E}(t') dt'$, which is proportional to the vector potential (in the Coulomb gauge) of the laser pulse. The finally observed momentum is therefore

$$\mathbf{p} = \mathbf{p}_i + \mathbf{A}(t). \quad (2)$$

Here $\mathbf{p}$ is nothing but the canonical momentum of the electron, which is conserved during free propagation in the laser field. The at first sight surprising fact that a free electron is accelerated by a laser pulse arises, because the electron is set free *during* the laser pulse. If the electron were set free *before* the pulse, it would indeed not be accelerated as $\mathbf{A}(-\infty) = \int_{-\infty}^\infty \mathcal{E}(t) dt = 0$ for a freely propagating laser pulse. If electrons are released during an extended period of time the *time* distribution of the release process is mapped into a *momentum* distribution of the measured electrons, as illustrated in figure 1 for a linearly polarized streak field with $\mathbf{p}_i \parallel \mathbf{A}(t)$. This is the basic principle of the attosecond streaking measurement.

Clearly, a unique reconstruction of the release time distribution from a single measurement requires that the electrons are released with a well-defined initial momentum $\mathbf{p}_i$. However, electrons are generally released with an initial *momentum*

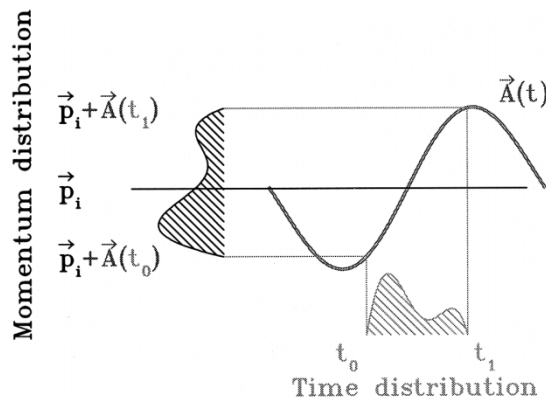


Figure 1. Mapping time distributions into momentum distributions. Electrons are released between times t_0 and t_1 with a certain time-dependent intensity profile and constant initial momentum $\mathbf{p}_i$. The laser field with vector potential $\mathbf{A}(t)$ shifts the momenta to different final momenta $\mathbf{p} = \mathbf{p}_i + \mathbf{A}(t)$, depending on the release time t . For clarity, it is assumed that $\mathbf{p} \parallel \mathbf{A}(t) \forall t$.

distribution $n_{\text{cl}}(\mathbf{p}_i, t)$, as in XUV photoionization, where spectral width and a possible chirp of the XUV pulse are reflected in a broad and time-dependent distribution of initial electron energies. In this more general case, the observed electron spectra are given by the integral

$$\sigma(\mathbf{p}) = \int_{-\infty}^{\infty} n_{\text{cl}}(\mathbf{p} - \mathbf{A}(t), t) dt, \quad (3)$$

where we have expressed the initial release momentum through the conserved canonical momentum $\mathbf{p}_i = \mathbf{p} - \mathbf{A}(t)$. The field free release distribution can be fully recovered, when a series of measurements with different laser pulses $\mathbf{A}(t)$ is performed. Each measurement provides a different ‘tomographic projection’ of $n_{\text{cl}}(p_i, t)$ on the momentum axis from which the two-dimensional distribution can be recovered (cf. figure 2).

For short release times that fit into half an optical period of the streak field, an explicit formula for the tomographic reconstruction of $n_{\text{cl}}(\mathbf{p}_i, t)$ can be given. For simplicity, we again restrict our analysis to the $\mathbf{p}_i$ -component parallel to $\mathbf{A}$ and consider the corresponding scalars: p_i , A and $\mathcal{E}$. We assume that A is a monotonic function of the form $A(t) = \mathcal{E}_0 a(t)$ and instead of varying the delay we vary the *amplitude* of the electric field $\mathcal{E}_0$. If we have measurements for the whole range of $\mathcal{E}_0 = -\infty \dots \infty$, it is easy to verify that the release distribution $n_{\text{cl}}(p_i, t)$ is obtained from the streaked spectra $\sigma(p, \mathcal{E}_0)$ by the inverse Radon transform

$$\begin{aligned} n_{\text{cl}}(p_i, t) = & \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} dq \int_{-\infty}^{\infty} d\mathcal{E}_0 \int_{-\infty}^{\infty} dp' |q\dot{a}(t)| \\ & \times \exp(i[qp' - qp - qE_0 a(t)]) \sigma(p', E_0). \end{aligned} \quad (4)$$

This shows that we may, in principle, *fully* reconstruct the release dynamics from a sufficiently large set of streaking measurements. In practice, the applicable field strength $\mathcal{E}_0$ is limited by the requirement that the investigated process not be severely disturbed by the streak field. This constraint limits the accuracy of

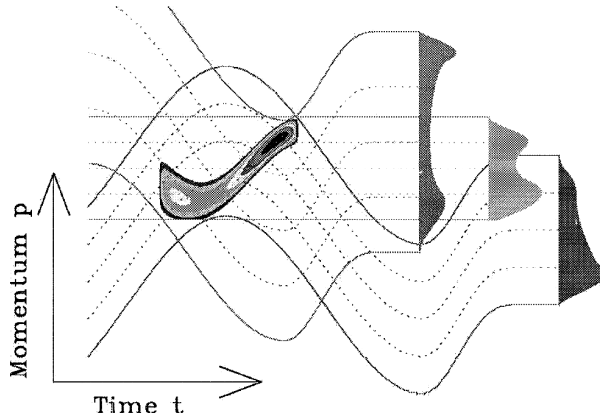


Figure 2. Tomographic cuts through the time-momentum distribution of electron release by a set of streaking measurements. For each delay, electrons observed at momentum $\mathbf{p}$ are accumulated along different time-momentum cuts (solid and dashed lines) through the release distribution $n_{\text{cl}}(\mathbf{p}_i, t)$. The lines indicate the laser vector potential $\mathbf{A}(t)$ which relates the momentum at release time t to the observed (canonical) momentum by $\mathbf{p}_i = \mathbf{p} - \mathbf{A}(t)$.

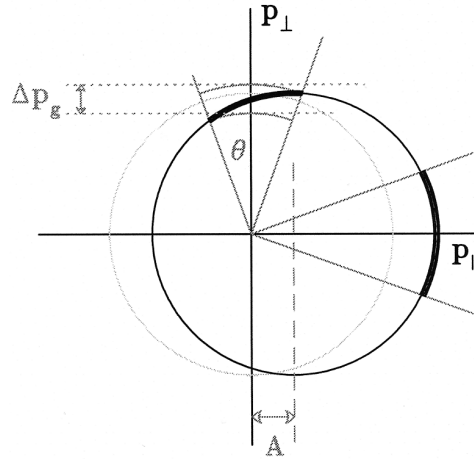


Figure 3. Geometrical effects in a streaking measurement. Streaking shifts the circle corresponding to equal emission energies (light grey) in momentum space by $\mathbf{A}$ to a circle of observed momenta (black). Parallel to the laser field polarization, the electron energies are maximally boosted. In the perpendicular direction the boost is much smaller, but when collecting electrons over a finite angle θ momentum spectra experience a purely geometrical broadening by Δp_g .

reconstruction of $n_{cl}(p_i, t)$, in particular the time-resolution that can be achieved, which will be discussed below after the introduction of quantum theory.

The principle laid out for the case $\mathbf{p} \parallel \mathbf{A}$ can be readily generalized to other experimental geometries. For example, in the first streaking measurements [6, 13] electrons were detected perpendicular to the laser polarization direction. As in that case, not only the magnitude of emitted electron momentum is changed, but also its direction, effects of the measurement geometry are superimposed on the simple boosting picture explained above. If one assumes (as in photoionization), a rather well-defined initial photoelectron energy E_i , the momenta of all emitted electrons lie on a sphere in momentum space with radius $(2mE_i)^{1/2}$. Streaking shifts this sphere as a whole by $\mathbf{A}(t_i)$, leading to maximum acceleration in the forward direction and maximum deceleration in the backward direction. The effect of the boost is minimal in the perpendicular direction—it is of the order $\sim |\mathbf{A}(t)|^2$ provided that $E_i \gg |\mathbf{A}(t)|^2$. When, as in any experiment, electrons are collected from a finite opening angle, a *geometrical* broadening of the detected electron spectrum occurs, even when the unidirectional spectrum is not broadened. The effect is weakest parallel to the laser polarization and maximal in the perpendicular direction (cf. figure 3). Provided the measurement geometry is well known, these geometrical effects can be easily corrected for. We will therefore disregard all geometrical effects in the further discussion and concentrate on the case $\mathbf{p} \parallel \mathbf{A}$.

3. Quantum theory

In the above discussion we have tacitly assumed that a clear distinction can be made between electrons that are ‘inside’ the atom, where they are not affected by the streaking laser field, and electrons that are ‘outside’ the atom, which are

essentially free. We formulate the analogous assumption for quantum theory by writing the full time-dependent wave function in momentum space and in direction $\mathbf{A} \parallel \mathbf{p} := p$ in the form

$$\Psi(p, t) = \Phi_{\text{in}}(p, t) + \Phi_{\text{out}}(p, t), \quad (5)$$

where $\Phi_{\text{in}}(p, t)$ is the (quasi-)bound content of the wave function at time t , whereas $\Phi_{\text{out}}(p, t)$ does not feel the binding potential and behaves like a free electron wave function. Such a splitting will usually be acceptable, if the binding forces of the excited electronic system are large compared to the laser force and if the electron release energies are far from threshold, as has been the case in streaking experiments so far [7, 9]. We further assume that Φ_{out} can be written as the integral

$$\Phi_{\text{out}}(p, t) = \int_{-\infty}^t \chi(p, t') \exp\left(\frac{i}{2} p^2 t'\right) dt', \quad (6)$$

i.e. the free part of the wave function $\Phi_{\text{out}}(t)$ is accumulated as the coherent sum of wave function parts $\chi(t')$ released at all times before t . To illustrate what is meant by this, we recall that the single XUV photoionization amplitude is given by

$$\Phi_{\text{out}}(p, t = \infty) \approx \int_{-\infty}^{\infty} \mathcal{E}_{\text{XUV}}(t') d(p) \exp(-iE_0 t') \exp\left(\frac{i}{2} p^2 t'\right) dt', \quad (7)$$

where $\mathcal{E}_{\text{XUV}}(t)$ is the full (carrier and envelope) field of the XUV pulse, $d(p)$ is the bound free dipole matrix element and $\exp(-iE_0 t)$ are the quantum phase oscillations corresponding to the initial state energy E_0 . In this case we simply have $\chi(p, t) = \mathcal{E}_{\text{XUV}}(t) d(p) \exp(-iE_0 t)$. A suitable $\chi(p, t)$ was recently defined also for the Auger decay [14].

A streak field affects Φ_{out} by shifting the momenta after release *and* modifying the quantum phases of each contribution $\chi(p, t)$ depending on the ‘release time’ t in the form

$$\Phi_{\text{out}}(p, t) = \int_{-\infty}^t \chi(p - A(t'), t') \exp\left(\frac{i}{2} \int_{-\infty}^{t'} [p - A(t'')]^2 dt''\right) dt'. \quad (8)$$

From that we obtain the observed streaked spectra

$$\begin{aligned} \sigma_A(p) &= |\Phi_{\text{out}}(p, t = \infty)|^2 \\ &= \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt' \exp\left[\frac{i}{2} \int_t^{t'} [p - A(t'')]^2 dt''\right] \chi^*(p - A(t), t) \chi(p - A(t'), t'), \end{aligned} \quad (9)$$

which by the coordinate transformation $(t, t') \rightarrow (T, \tau) = ((t + t')/2, t - t')$ takes the form

$$\sigma_A(p) = \int_{-\infty}^{\infty} dT n_{\text{qu}, A}(p, T), \quad (10)$$

with the quantum equivalent of the classical electron release distribution

$$\begin{aligned} n_{\text{qu}, A}(p, T) &= \int_{-\infty}^{\infty} d\tau \exp\left[-\frac{i}{2} \int_{-\tau/2}^{\tau/2} [p - A(T + t'')]^2 dt''\right] \\ &\times \chi^*(p - A(T + \tau/2), T + \tau/2) \chi(p - A(T - \tau/2), T - \tau/2). \end{aligned} \quad (11)$$

Within the assumptions made about our release process, namely that electrons outside the atom are essentially free and that the electric field does not become too large, n_{qu} behaves under streaking like its classical counterpart n_{cl}

$$n_{\text{qu,A}}(p, T) \approx n_{\text{qu,0}}(p - A(T), T). \quad (12)$$

This can be shown by separating phase and modulus of χ in the form

$$\chi(p, T) = \eta(p, T) \exp[-i\Omega(p, T)T] \quad (13)$$

and assuming that both η and Ω slowly vary under streaking. For η as a function of $p - A(T \pm \tau/2)$ this is generally fulfilled far from threshold and in the absence of sharp resonances in the continuum, which is equivalent to assuming that the unbound electron is essentially free. Slow variation of Ω more specifically means that the total derivative of $\Omega(p - A(T \pm \tau), T \pm \tau)$ with respect to τ remains small compared to the emission momentum $|\mathcal{E}\partial_p\Omega + \partial_T\Omega| \ll |\mathcal{E}p|$. It means that the ‘instantaneous emission energies’ $\Omega(p, T)$ do not vary too much compared to their absolute value. For given T and p , contributions to integral (11) then only come from a narrow range of τ around the stationary point τ_0 defined by

$$\left\{ \frac{d}{d\tau} \int_{-\tau/2}^{\tau/2} [p - A(T + t'')]^2 dt'' - \Omega\tau \right\} \bigg|_{\tau_0} = 0. \quad (14)$$

Expanding the rest of the integrand to the lowest order in τ one obtains the behaviour (12).

The purpose of this derivation was to point out that, as n_{qu} behaves in the same way under streaking as n_{cl} , it can be reconstructed from a series of streaking measurements by the inverse Radon transform (4). Since the transformation behaviour (12) contains certain approximations, reconstruction by inverting the Radon transform introduces a certain amount of errors. An unproven conjecture is that an exact reconstruction may be possible also for the quantum case.

4. Time-independent versus time-resolved measurement

The quantum release distribution n_{qu} contains rich time and energy information about the release process. As pointed out below, n_{qu} assumes the approximate form of a Wigner function, when the p dependence over the range of release energies of χ can be neglected, which allows a complete reconstruction of χ . The release distribution also enables us to clearly identify additional information gained in time-resolved measurements which *in principle* cannot be obtained in a time-independent measurement of traditional spectroscopy.

The most elementary object of observation in time-independent spectroscopy is a single line $\sigma_1(E)$, which determines the spectral amplitude of a release process up to an unknown spectral phase

$$\tilde{\Phi}_1(E) = [\sigma_1(E)]^{1/2} \exp[i\varphi(E)]. \quad (15)$$

The Fourier transform $\Phi_1(t) = \mathcal{F}[\tilde{\Phi}_1](t)$ contains the complete timing information about the decay process. It is common to assume that we know the time characteristics of the release, e.g. exponential decay with a Lorentzian line shape, in which case the line width is the only unknown parameter and one can determine the duration of the decay. In that case one can write $\varphi(E) \approx \beta(E) + t_1 E$,

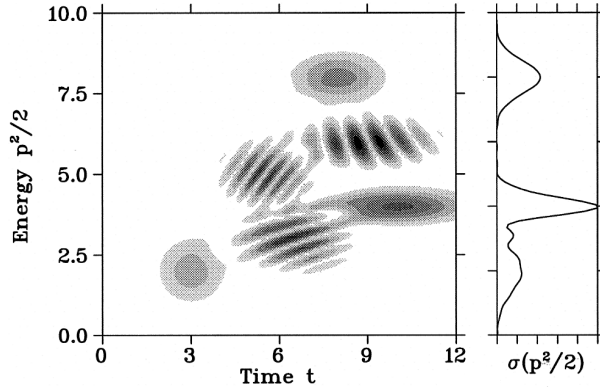


Figure 4. The electron release distribution n_{qu} for three decay lines at energies $E_n = 2, 8, 4$ and times $t_n = 3, 8, 10$. The oscillating areas are typical for Wigner functions and they add up to 0 in those parts of the time-independent spectrum where the lines do not overlap (box on the right). Note that n_{qu} is plotted with respect to energy $p^2/2$ rather than momentum p .

where $\beta(E)$ is characteristic for the decay and t_1 determines *when* the decay happens, information that is usually of little interest for a single line. This changes when there is a second line σ_2 in the spectrum, as the difference between the times $t_1 - t_2$ gives us important information about the dynamics of the underlying process, telling us which of the two decays happens first. This information is not accessible by a time-independent measurement, as can be easily seen when the two lines do not overlap. In order to relate the phases of Φ_1 and Φ_2 we need a third process that can be kept coherent over a sufficiently wide spectral range to have overlap and interfere with both lines, i.e. a time-resolved measurement. As an illustration we show in figure 4 n_{qu} for a system which generates three lines at different times with the release function $\chi(p, t) = \sum_n \exp[(t - t_n)^2/w_n^2 - iE_n t]$.

5. Explicit reconstruction of release dynamics

A complete measurement of the electron release dynamics means that we can recover the ‘released wave packet’ $\chi(p, t)$ defined above. If we could reconstruct this function from experimental data, we would indirectly measure the outgoing wavefunction of the electron $\Phi_{\text{out}}(p, t)$. Several methods have already been proposed for such reconstruction [15]. One of them, dubbed ‘attosecond SPIDER’ [16], allows for explicit reconstruction of the wavepacket, but it assumes that two identical time-delayed replicas of the electronic wavepacket are available in experiments. Another technique, which is called ‘attosecond FROG’ [15], is free from this assumption, but the method is not explicit, as it requires an iterative procedure to retrieve the wavepacket. Here we propose reconstruction techniques, which are both explicit and applicable to a single electronic wavepacket.

While the question whether transformation (11), which relates $\chi(p, t)$ to the quantum time-momentum distribution, can be exactly inverted in the general case remains open, we can point out two important particular cases, when such inversion can be performed analytically: linear streaking, which we consider in this section, and oscillatory streaking, which will be considered in section 7. Together

with the tomographic reconstruction of the time–momentum distribution (4) it gives us a recipe on how to reconstruct a wavepacket.

Let us assume that the electronic wavepacket is so short in time that the electric field of the probe pulse $\mathcal{E}(t)$ can be considered constant during the release time, so that the vector potential can be approximated by

$$A(t) = -\mathcal{E}_0 t. \quad (16)$$

If we assume further that the function $\chi(p, t)$ can be factorized

$$\chi(p, t) = d(p)\chi(t) \exp(-i\Omega t), \quad (17)$$

like in the photoionization example above, then n_{qu} assumes a form very similar to the Wigner distribution

$$W(t, \omega) = \int_{-\infty}^{\infty} d\tau \chi^*\left(t - \frac{\tau}{2}\right) \chi\left(t + \frac{\tau}{2}\right) \exp(-i\omega\tau). \quad (18)$$

There are just two more approximations we have to make in order to express $n_{\text{qu,A}}(p, T)$ as a Wigner transform of the wavepacket. First, we assume that the streak field is weak enough to neglect terms $\sim \mathcal{E}_0^2 \tau^3$. Second, we assume that $d(p - A(t))$ is not affected much by the streak field, which makes it independent of time:

$$d(p - A(t)) \approx d(p). \quad (19)$$

Under these approximations

$$n_{\text{qu}}(p, T) = |d(p)|^2 W\left(T, \Omega - \frac{(p + \mathcal{E}_0 T)^2}{2}\right). \quad (20)$$

The function $d(p)$ is the dipole matrix element for the transition from the bound state to the continuum state with the momentum p . From equation (20) we see that in order to reconstruct the Wigner transform of the wavepacket we do not have to know the complex dipole matrix element, but only its squared module, which can be measured experimentally.

Once the Wigner distribution is known, it is easy to invert it. In order to do this we consider the inverse Fourier transform of $W(\tau, \omega)$ with respect to the frequency:

$$\tilde{W}(\tau, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} W(\tau, \omega) \exp(-i\omega t) d\omega = \chi^*\left(\tau - \frac{t}{2}\right) \chi\left(\tau + \frac{t}{2}\right),$$

and notice that

$$\chi(t) = \frac{\tilde{W}\left(\frac{t}{2}, t\right)}{[\tilde{W}(0, 0)]^{1/2}} \exp(i\varphi),$$

where φ is a constant phase that cannot be retrieved.

There is one important issue to be discussed in this context. Equation (10) requires a scan over the strength of the streak field from $-\infty$ to ∞ , but the maximal streak field is limited by several conditions: it may not disturb the investigated quantum system and it must be small enough for the approximations above to be valid. In order to analyse the effect of the final strength of the streak field and to build a reliable reconstruction procedure, we have to consider the 2D

Fourier transform of the distribution $n_{\text{qu}}(t, p)$:

$$\tilde{n}(w, u) = \int_{-\infty}^{\infty} dt \exp(iwt) \int_{-\infty}^{\infty} dp \exp(iup) n_{\text{qu}}(t, p). \quad (21)$$

According to the Fourier slice theorem the Fourier transform $S(u, \mathcal{E}_0)$ of the spectrum $\sigma(p, \mathcal{E}_0)$ with respect to p is equal to the values of $\tilde{n}(w, u)$ on the line $u = -\mathcal{E}_0 w$:

$$S(u, \mathcal{E}_0) = \int_{-\infty}^{\infty} dp \exp(ipu) \sigma(p, \mathcal{E}_0) = \tilde{n}(-\mathcal{E}_0 u, u). \quad (22)$$

So, varying the field strength $\mathcal{E}_0$ from minus infinity to plus infinity one can record $\tilde{n}(w, u)$:

$$\tilde{n}(w, u) = S\left(u, -\frac{w}{u}\right). \quad (23)$$

The distribution $n_{\text{qu}}(t, p)$ is then reconstructed with the aid of the inverse 2D Fourier transform:

$$n_{\text{qu}}(t, p) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} dw \exp(-iwt) \int_{-\infty}^{\infty} du \exp(-iup) \tilde{n}(w, u). \quad (24)$$

A finite strength of the streak field means that $\tilde{n}(w, u)$ cannot be reconstructed for some values of w and u , as shown schematically in figure 5. It is easy to show that, when we know $\tilde{n}(u, w)$ on any finite cone as indicated in figure 5, in mathematical principle we can reconstruct $\tilde{n}$ everywhere, provided the investigated process has a finite duration, i.e. $n_{\text{qu}}(p, t) = 0$ for $|t| > t_{\text{max}}$, because the function $\tilde{n}(w, u)$ is analytic in w and can therefore be uniquely continued across the gap of our measurement.

There remains the practical problem to obtain an optimal analytic continuation from experimental data with finite accuracy. Because of analyticity, we know that polynomial interpolations are legitimate and should converge to the exact result. In numerical simulations we used the spline interpolation, however, higher order

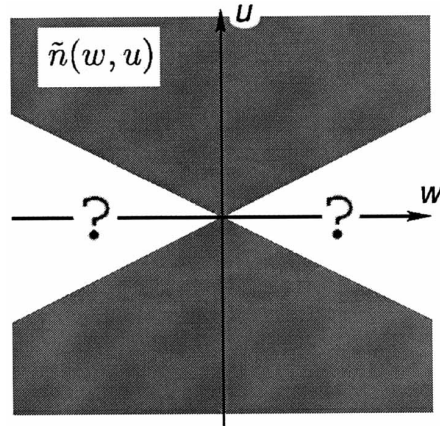


Figure 5. If the strength of the streak field varies within a finite range, there is a region where $\tilde{n}(w, u)$ cannot be reconstructed from electron spectra. In particular, if $\mathcal{E}_0$ varies from $-\mathcal{E}_{\text{max}}$ to $\mathcal{E}_{\text{max}}$, then (23) leaves $\tilde{n}(w, u)$ undefined on the cone $|u| < |w|/\mathcal{E}_{\text{max}}$.

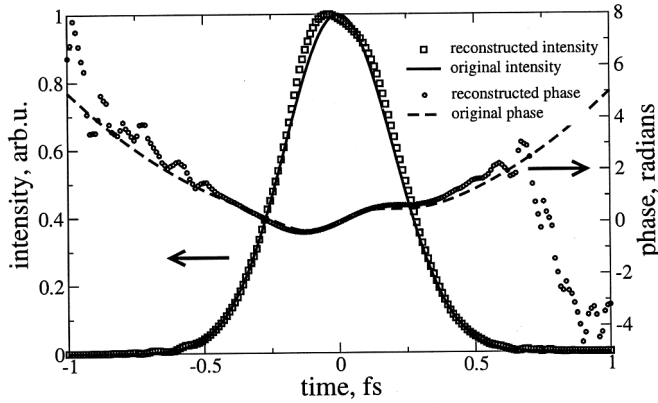


Figure 6. An example of reconstruction of a chirped electronic wavepacket in the regime of linear streaking. The parameters of the original wavepacket are given in the text.

methods or algorithms exploiting in addition the redundancy contained in the specific form of n_{qu} may further improve reconstruction [17].

In figure 6 a reconstruction example is shown. In this example we reconstructed the following wavepacket:

$$\chi(t) = \exp \left[-\frac{t^2}{2T_0^2} + i \left(\frac{t^2}{5T_0^2} + \sin \left(\frac{t}{T_0} \right) \exp(-t^2/T_0^2) \right) \right]$$

with $T_0 = 0.2$ fs. The maximal intensity of the streak field was chosen to be equal to $10^{13} \text{ W cm}^{-2}$ and the gap in $\tilde{n}(w, u)$ was filled with the aid of the spline interpolation.

6. Time resolution

A simple intuitive estimate for the time resolution by a *linear* streaking measurement uses the quantum mechanical time–energy uncertainty relation. Say we want to resolve a given time interval Δt . That means that we should shift the parts of the spectrum produced at the beginning and at the end of the interval far enough such that their contributions do not overlap in the final streaked spectrum. Obviously, how much we need to shift depends on the width the two separate contributions to the spectrum. That width itself is at least as big as given by the time–energy uncertainty

$$\Delta t \Delta E_u \approx \hbar. \quad (25)$$

The streaking shift near the peak with the maximum field strength $\mathcal{E}_0$, where the vector potential is approximately linear $A(t) \sim \mathcal{E}_0 t$, is given by

$$\Delta E_s(\Delta t) = \frac{1}{2} [p_i + \mathcal{E}_0 \Delta t]^2 - \frac{1}{2} p_i^2 \approx \mathcal{E}_0 p_i \Delta t. \quad (26)$$

From the requirement

$$\Delta E_u < \Delta E_s, \quad (27)$$

one obtains the fundamental quantum limitation

$$\Delta t > \left(\frac{\hbar}{E_0 p_i} \right)^{1/2}. \quad (28)$$

A similar estimate was obtained in [11] for the special case of photoionization by two XUV pulses.

It is useful to express the time resolution (28) in terms of the maximum photoelectron energy shift $\Delta E_{\max}$ by a streak field compared to the field free position. For a continuous-wave laser field $\mathcal{E}(t) = \mathcal{E}_0 \sin(\omega t)$ the energy shift for an electron release with initial momentum p_i is $\Delta E_{\max} = \mathcal{E}_0 p_i / \omega$, by which (28) assumes the form

$$\Delta t > \left(\frac{\hbar}{\omega \Delta E_{\max}} \right)^{1/2}. \quad (29)$$

Inserting the parameters of the recent streaking measurements [7], where an 800 nm laser was used for streaking and energy shifts of $\Delta E_{\max} = \pm 20$ eV could be achieved, one sees that currently release dynamics on the scale of ~ 100 as may be resolved.

The time-resolution limitation by the maximum streak field is seemingly in contradiction to the preceding section, where it was pointed out that any finite range of streak fields is sufficient for the complete reconstruction of $n_{\text{qu}}(p, t)$ and therefore of χ . However, the mathematical statement assumes error free electron spectra and exact knowledge of the streak field. From a mathematical point of view, we can observe any minuscule structure with an arbitrarily blunt instrument, if our knowledge of the instrument is not subject to errors. However, a meaningful definition of time resolution requires an error analysis of the measurement.

Rather than attempting an analytic description of the error, we show that numerical experiments using the quantum theory of streaking reproduce the resolution formula with high accuracy. White noise is added to simulated streaked spectra and the Fourier transform of the release wave function $\chi(t)$ is analysed for sensitivity to noise. Our simulations show that, if the noise is small, the spectral phase of the wavepacket is accurately reconstructed for small frequencies with an abrupt growth of the standard deviation at some threshold frequency (figure 7(a)). In this case it is natural to define the time resolution of the reconstruction method through the width of the spectral region, in which the wavepacket is reconstructed reliably:

$$\Delta t = \frac{\pi}{\omega_{\max} - \omega_{\min}}. \quad (30)$$

In figure 7(b) (over page) the width of the spectral region predicted by equation (28) is compared with the width calculated numerically for a chirped Gaussian wavepacket. The reconstruction of the wavepacket, used for this example, was performed as described in section 5. The region of reliable reconstruction of the spectral phase was defined as the region in which the standard deviation of the phase is smaller than 0.1 rad, confirming our simple analytic formula with surprising accuracy.

Better time resolution can be achieved, when we have additional information about the underlying process. For example, when we know that the release wavepacket can be described by a central frequency with some extra chirp,

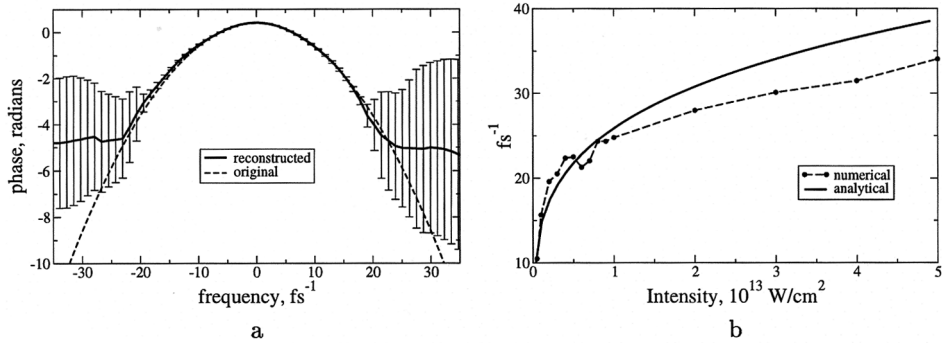


Figure 7. (a) Deviations of the reconstructed spectral phase caused by 1% noise added to $\sigma(p)$. (b) The width of the spectral region in which the spectral phase is reliably reconstructed in the presence of the noise (marked as ‘numerical’) in comparison with $\pi/\Delta t$, where Δt is given by equation (28) (marked as ‘analytical’).

streaking provides a very sensitive indicator of the chirp. In that case, in principle, measurements with only two different fields may suffice to recover the complete chirp [18]. Such an overdetermination of the streaking measurement explains the very small errors of a few tens of attoseconds on the XUV pulse duration reported in [7].

7. Oscillatory streak field

In this regime the duration of an electronic wavepacket is assumed to be comparable to the oscillation period of the streak field, and we use the following expression for the vector potential of the field:

$$A(t) = A_0 \sin(\omega_0 t). \quad (31)$$

Like in the case of linear streaking discussed in section 5, let us assume that the wavepacket is factorizable:

$$\chi(p, t) = d(p)\chi(t) \exp(i\Omega t), \quad (32)$$

where the fast linear change of the phase is written as a separate term (the physical meaning of Ω is the average electron’s kinetic energy without the streak field). We will also use assumption (19), considering the dipole matrix element to be independent of time even in the time-dependent streak field.

Unlike the case of linear streaking, we will change not the strength of the streak field, but the delay τ_d between the examined wavepacket and the streak field, which is equivalent to changing the phase of the field. It can be shown that for a moderate intensity of the streak field $A_0^2 \ll 8\pi\omega_0$ the momentum distribution of the electrons can be written as

$$\sigma(p, A_0, \tau_d) \approx \left| d(p) \sum_{k=-\infty}^{\infty} i^k \exp \left[-i \left(\frac{p^2}{2} + k\omega_0 \right) \tau_d \right] J_k \left(\frac{pA_0}{\omega_0} \right) \tilde{\chi} \left(\frac{p^2}{2} - \Omega + k\omega_0 \right) \right|^2, \quad (33)$$

where J_n are the Bessel functions and $\tilde{\chi}(\omega)$ stands for the Fourier transform of the wavepacket $\chi(t)$. The function $\sigma(p, A_0, \tau_d)$ has to be periodic with respect to τ_d due

to the periodicity of the streak field. Hence, it can be expanded into the Fourier series and the coefficients of this expansion are given by the integral

$$\int_0^{2\pi/\omega_0} \sigma(p, \tau_d, A_0) \exp(iq\omega_0\tau_d) d\tau_d = -\frac{2\pi i}{\omega_0} |d(p)|^2 \sum_n \left[J_{n+q} \left(\frac{pA_0}{\omega_0} \right) J_n \left(\frac{pA_0}{\omega_0} \right) \right. \\ \left. \times \tilde{\chi} \left(\frac{p^2}{2} - \Omega + (n+q)\omega_0 \right) \tilde{\chi}^* \left(\frac{p^2}{2} - \Omega + n\omega_0 \right) \right], \quad (34)$$

where q is an integer number. Analysing this equation one can see that the problem of reconstruction of $\tilde{\chi}(\omega)$ resembles the SPIDER reconstruction [19]. Indeed, the power spectrum $|\tilde{\chi}(\omega)|^2$ can be measured in experiments, so that the reconstruction problem is reduced to the reconstruction of the spectral phase $\varphi(\omega) = \arg[\tilde{\chi}(\omega)]$. Each term of the sum in equation (34) represents the interference of two replicas of the wavepacket (sidebands) shifted by the streaking frequency ω_0 . In the regime, when contribution from no more than three sidebands is significant at each separate frequency, the phase difference $\theta(\omega) = \varphi(\omega - \omega_0) - \varphi(\omega)$ can be reconstructed analytically. The only essential difference from the SPIDER is that in our case the frequency shift ω_0 is comparable to the spectral width of the wavepacket, which we want to reconstruct, and the SPIDER reconstruction requires the shift to be much smaller than the frequency range, on which the spectral phase changes significantly. However, one can solve this problem by making two experiments with slightly different streaking frequencies. Indeed, if we know the functions

$$\begin{aligned} \theta_1(\omega) &= \varphi(\omega - \omega_1) - \varphi(\omega), \\ \theta_2(\omega) &= \varphi(\omega - \omega_2) - \varphi(\omega), \end{aligned} \quad (35)$$

and if the difference between the frequencies $\Delta\omega = \omega_2 - \omega_1$ is small, then

$$\frac{\theta_2(\omega + (\Delta\omega/2)) - \theta_1(\omega - (\Delta\omega/2))}{\Delta\omega} = -\frac{\varphi(\omega + (\Delta\omega/2)) - \varphi(\omega - (\Delta\omega/2))}{\Delta\omega} \\ \approx -\varphi'(\omega), \quad (36)$$

$$\frac{\theta_2([(\omega_1 + \omega_2)/2] - \omega) - \theta_1([(\omega_1 + \omega_2)/2] + \omega)}{\Delta\omega} = -\frac{\varphi(-\omega + (\Delta\omega/2)) - \varphi(-\omega - (\Delta\omega/2))}{\Delta\omega} \\ \approx -\varphi'(-\omega), \quad (37)$$

which gives us the derivative of the spectral phase (also known as the group delay). It is interesting to note that this method does not require strong probe fields. In fact, the approximations involved favour weak probe fields, the lower limit being set by our ability to experimentally observe the streaking effect and its dependence on the frequency of the streak field.

An example of pulse reconstruction in this regime is shown in figure 8. The wavepacket was specified in the frequency domain:

$$\tilde{\chi}(\omega) = \exp \left[-1.7\omega^2/\omega_0^2 + i(\cos(1.5\pi\omega/\omega_0) + 0.5 \sin(2\pi\omega/\omega_0)) \right]$$

with $\omega_0 = 2.51 \text{ fs}^{-1}$. The two streaking frequencies were 2.51 and 2.36 fs^{-1} and in both cases the intensity of the streak field was equal to $3 \times 10^{10} \text{ W cm}^{-2}$.

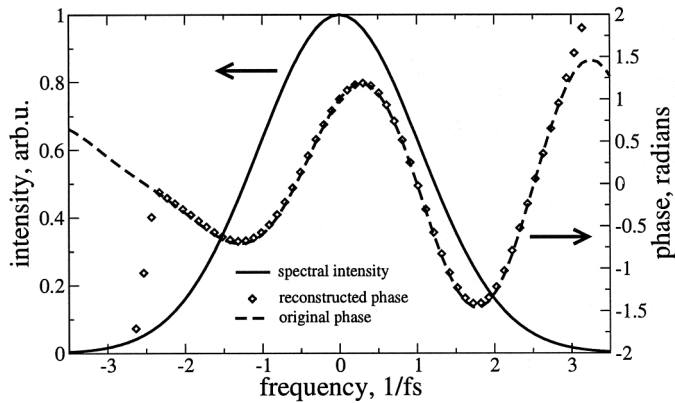


Figure 8. An example of reconstruction of a chirped electronic wavepacket in the regime of oscillatory streaking. The parameters of the original wavepacket are given in the text. For this reconstruction we assume that the power spectrum of the wavepacket $|\tilde{\chi}(\omega)|^2$ is known, and the spectral phase is retrieved under the assumption that at each frequency ω at most three sidebands contribute to the observed spectral intensity.

8. Summary and open questions

We have given an account of our present understanding of the attosecond streaking method. A key assumption in all present applications of the method is that electrons are either in the atom or free, and that intermediate states are negligible. This assumption underlies both the classical and quantum theories of streaking, and it basically excludes modification of the electron release process by the field, limiting the influence of the field to a boost of free electron motion. Under this condition a quantum release distribution was defined in analogy with the intuitive classical release distribution n_{cl} . Both densities, in good approximation, have the same behaviour under streaking and therefore both can be recovered from measured spectra by the same or similar reconstruction procedures. In the quantum case the release distribution bears great similarity to a Wigner function and takes the exact shape of the Wigner function of the temporal release amplitude, when the momentum dependence of the release amplitude is weak. In that case it is easy to show that not only the electron release distribution, but also the underlying quantum mechanical amplitude can be recovered from a streaking measurement, thus providing the *maximal possible information* about the electron release dynamics. Limitations of the applicable field strength limit the time resolution of a streaking measurement in a straightforward way, which we capture in a simple time-resolution formula. Using the notion of ‘release distribution’ one can easily point out what is the edge of time-resolved measurements over time-independent methods: it is—tautologically—time resolution. A simple example of non-overlapping spectral lines with different timing is instructive for understanding that time-resolved measurements cannot just do some things ‘better’ than time-independent methods, but that timing, *in principle*, is not accessible by stationary methods.

Although our overall understanding of attosecond streaking becomes rather clear by now, several important issues remain to be resolved. On a technical level, optimal reconstruction mechanisms need to be defined that allow the recovery of the theoretical maximum of dynamical information from noisy measurement data

obtained with limited electric field strength. This may also lead to quantitative improvements of time resolution, although we do not expect a fundamental reduction of the limits given by our simple formula. Both advanced analytic continuation techniques and exploitation of the specific structure of the electron release distribution may help us to approach that goal. Similarly, the deviations of the quantum streaking behaviour from the classical behaviour must be corrected for in the reconstruction process. Here the analogy to the classical system seems to indicate that this is possible, but proof and practical implementation are lacking. Resolution of this issue seems particularly important, as systems with interesting dynamics may not allow factorization of the type $\chi(p, t) \sim d(p)\chi(t)$, which is assumed for most of the practical examples given here.

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