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## Analytical estimates of attosecond streaking time delay in photoionization of atoms

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#### Analytical estimates of attosecond streaking time delay in photoionization of atoms

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We present estimates of the attosecond streaking delay in photoionization of atoms based on an analytical formula. In the derivation of the formula we use that the streaking delay depends on the propagation of the photoelectron over a finite range in space. We find that the analytical estimates agree well with results of ab-initio calculations. Application of the formula provides insights into the influence of the streaking field on the field-free time delay in the analysis of streaking measurements.

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#### I. INTRODUCTION

The quest to study ultrafast dynamics in matter on their natural timescales has driven the technological development of ultrashort laser pulses over the last decades. Attosecond pulse technology (for reviews, see e.g. [1, 2]) has recently enabled the resolution of dynamical quantum processes on the timescale of electronic motion in atoms, molecules and solid state matter. A number of spectroscopic techniques have been developed (for reviews see e.g. [1, 3-7]), among which the attosecond streaking technique [8] is a prominent one.

In this application of the streak camera principle a linearly polarized isolated attosecond pulse is used along with a moderately strong femtosecond streaking pulse, usually with wavelengths in the near-infrared or infrared regime. The asymptotic momentum of the photoelectron, ionized by the attosecond pulse, depends on the vector potential of the streaking pulse and the propagation of the electron in the continuum. A streaking trace can be obtained by detecting the momentum as a function of the time delay between ionizing and streaking pulses. By comparing the streaking trace to the oscillation of the vector potential a temporal shift, called streaking time delay, can be determined (see, e.g. [9–12]).

The streaking time delay  $\Delta t_s$  is usually studied via the sum of the Wigner-Smith time delay  $\Delta t_{WS}$  [13, 14], related to the short-range part of the residual ion potential, and a term  $\Delta t_{CLC}$ , related to the coupling between the laser field and the long-range part of the Coulomb potential (e.g., [9–12, 15–25]):

$$\Delta t_s = \Delta t_{WS} + \Delta t_{CLC} \ . \tag{1}$$

Predictions based on this formula often rely on advanced numerical calculations. On the other hand the option of an accurate estimate based on analytical expressions can be useful to study the dependence of the time delay on different parameters of the pulses as well as targets. In our approach to develop such an analytical formula, in section II we set out with an alternative estimate of the streaking time delay, which consists of a sum of streaking field weighted field-free delays [26]. In the sections III and IV we then derive analytical expressions for each contribution to the field-free delay, namely the short-range, the Coulomb phase and the logarithmic term. A key element of the derivation is the fact that the streaking time delay depends on the propagation of the photoelectron over a finite range in space only [10, 26]. The predictions of the analytical estimate of the streaking time delay agree well with those of ab-initio calculations for photoemission from the outermost shell of an atom over a wide range of photoelectron energies. As an application we study in section V the influence of the streaking field on the three field-free delay contributions which provides an alternative justification of the sum formula, Eq. (1). The article ends with a brief summary. Hartree atomic units  $(e = m = \hbar = 1)$  are used throughout unless stated otherwise.

#### **II. PRELIMINARY CONSIDERATIONS**

Using a classical analysis of the propagation of the photoelectron after the liberation in the continuum it has been found that the streaking time delay can be approximated as [26]:

$$\Delta t_s \simeq \sum_{j=1}^N \frac{E_s(t_j)}{E_s(t_{ion})} \,\Delta t_{ff}^{(j)} \,. \tag{2}$$

Here,  $\Delta t_{ff}^{(j)}$  is the field-free time delay that the electron accumulates during the propagation in the time interval  $[t_j, t_{j+1}]$  after the liberation in the continuum at  $t_1 = t_{ion}$ and over the related finite-region  $[r_j, r_{j+1}]$  within the potential  $V(\mathbf{r})$  along the polarization direction of the streaking pulse.  $E_s(t_j)$  is the actual strength of the streaking field during the propagation over the given time interval and  $E_s(t_{ion})$  is the field strength at the time of liberation of the photoelectron. Since the streaking pulse has a finite duration, the sum extends over a finite number of intervals. Physically, the above formula states that the streaking delay can be considered as a sum of (streaking) field weighted field-free delays, accumulated from the moment of transition into the continuum to the end of the streaking pulse over a finite propagation distance. We note that the streaking time delay, Eq. (2), can alternatively be represented as an integral [26].

In order to obtain an analytical expression it is useful to write the piecewise field-free time delay as a difference:

$$\Delta t_{ff}^{(j)} = \Delta t_{ff}(k_{j+1}, r_{j+1}) - \Delta t_{ff}(k_j, r_j)$$
(3)

where  $\Delta t_{ff}(k,r)$  is the field-free time delay accumulated during the propagation of the photoelectron, from the origin of the transition into the continuum by the ionizing XUV pulse to r, with constant momentum  $k = k(r) = \sqrt{2(E_{asym} - V(r))}$  and  $E_{asym}$  is the asymptotic energy of the photoelectron. Since it has been previously shown [18, 26] that predictions based on classical calculations for the streaking time delay are in good agreement with those of quantum mechanical calculations, we assume a quantum-classical correspondence for the field-free time delay and further decompose the (classical) time delay into three well-known terms:

$$\Delta t_{ff}(k,r) = \Delta t_{short}(k,r) + \Delta t_{phase}(k,r) + \Delta t_{log}(k,r)$$
(4)

where each of the terms represents the derivative of a contribution to the (quantum mechanical) phase shift with respect to the photoelectron's energy.  $\Delta t_{short}(k, r)$  is the short-range contribution, while

$$\Delta t_{phase}(k,r) = \frac{1}{k} \frac{\partial}{\partial k} \arg\left[\Gamma(1+l+i\eta)\right]$$
(5)

and

$$\Delta t_{log}(k,r) = \frac{1}{k} \frac{\partial}{\partial k} \left[ \frac{Z}{k} \ln(2kr) \right]$$
(6)

with l is the angular momentum of the photoelectron,  $\eta = -Z/k$  and Z is the charge of the residual ion.

We note that  $\Delta t_{ff}(k,r)$  corresponds to the field-free time delay accumulated in the atomic potential, which is cut-off at r. In such cut-off potentials the time delay due to the Coulomb potential can be evaluated using the asymptotic forms (see e.g. [27]). The assumed quantum-classical correspondence in Eq. (4) is therefore expected to hold for cut-off distances, at which the shortrange part of the potential has vanished. Conversely, for distances close to the core of the residual ion, at which multi-electron dynamics play a dominant role, the above assumption is not longer valid. Thus, for small energies, at which the photoelectron explores the short-range part of the potential for a significant time, this may lead to discrepancies between the analytical estimates, to be derived in the next two sections, and the results of advanced ab-initio quantum calculations.



FIG. 1. (Color online) Comparison of predictions of analytical formula for the field-free time delay  $\Delta t_{ff}$ , Eq. (8), with results extrapolated from ab-initio calculations for hydrogen atom (Z = 1) [10] as a function of propagation time.

#### III. HYDROGEN-LIKE SYSTEMS: COULOMB-PHASE AND LOGARITHMIC TERMS

We first consider the case of hydrogen-like atoms and ions where the potential V = -Z/r does not contain a short-range contribution. The spectral derivative in Eq. (6) is readily determined as:

$$\Delta t_{log}(k,r) = \frac{Z}{k^3} \left[ 1 - \ln(2kr) \right] \,. \tag{7}$$

Since the streaking time delay, Eq. (2), depends on finite distances only, this term will not diverge. The Coulombphase term, Eq. (5), can be expressed in terms of the real part of the digamma function [12] or, equivalently, expanded in a sum [28], such that the field-free time delay, Eq. (4), for a hydrogenlike system is given by:

$$\Delta t_{ff}(k,r) = \frac{Z}{k^3} \left[ F(k,l) + \sum_{m=1}^{l} \frac{1}{m} - \gamma + 1 - \ln(2kr) \right]$$
(8)

where

$$F(k,l) = \sum_{n=0}^{\infty} \frac{Z^2}{(n+l+1)([k(n+l+1)]^2 + Z^2)}.$$
 (9)

The accurateness of the predictions of the analytical expression (solid line) can be seen from the comparison with previously published data extrapolated from ab-initio calculations (circles, [10]) as a function of the propagation time t after liberation of the electron in the continuum in Fig. 1. In these calculations we have used  $r = k_{asym}t$ , where  $k_{asym} = \sqrt{2E_{asym}}$ , in accordance with the analysis in [10].



FIG. 2. (Color online) Comparison of analytical estimates (solid lines) with results of ab-initio calculations (circles, [10]) of the streaking delay in hydrogen atom and helium ion. Parameters of the streaking field: peak intensity  $I_s = 1 \times 10^{12}$  W/cm<sup>2</sup>, wavelength  $\lambda_s = 800$  nm and pulse duration of three cycles. In the analytical calculations, ionization of the photoelectron at the peak of the pulse and the most probable location of the electron in the initial state has been assumed.

Calculation of the streaking delay, Eq. (2), along with Eq. (3) for hydrogen-like systems then requires to evaluate Eq. (8) at N finite distances  $r_j$ . These distances can be estimated via the recursive relation:

$$r_{j+1} = r_j + k(r_j)\delta t \tag{10}$$

with  $r_1 = r_{ion}$  and  $k(r_1) = \sqrt{2E_{asym} - V(r_1)}$  are the initial position and momentum of the photoelectron after the transition in the continuum. We note that the distances can be also determined using other standard approaches from the differential equation determining the classical trajectory r(t). In Fig. 2 we compare our analytical results for the streaking time delay for hydrogen atom and helium ion with those of ab-initio calculations [10]. A good agreement within less than one attosecond is found over the whole range of asymptotic energies of the photoelectron studied.

#### IV. MULTIELECTRON ATOMS: SHORT RANGE TERM

Next we consider atoms other than hydrogen and note that ionization of a multielectron atom, in particular in strong-field processes, is often modeled using singleactive-electron potentials, in which all electrons except the active photoelectron are assumed to be frozen during the interaction with the external fields. The corresponding single-active-electron potentials are given by:

$$V_{SAE}(\mathbf{r}) = -\frac{1}{r} + V_{short}(\mathbf{r})$$
(11)

where the short-range part,  $V_{short}$ , is typically modeled via a linear combination of Yukawa terms,  $V_{Yukawa}(\mathbf{r}) = a \exp(-\mu r)/r$ , and exponential terms,  $V_{exp}(\mathbf{r}) = a \exp(-\mu r)$  with a and  $\mu$  constant.

For the field-free time delay  $\Delta t_{ff}$  obtained by the photoelectron in such a single-active-electron potential, the sum of  $\Delta t_{phase}$  and  $\Delta t_{log}$  is given by Eq. (8) for Z = 1. Analogous to the other parts, the short-range contribution is given by the spectral derivative of the corresponding phase shift  $\delta_{short}$ :

$$\Delta t_{short}(k,r) = \frac{1}{k} \frac{\partial}{\partial k} \delta_{short}(k,r)$$
(12)

To obtain analytical estimates we make use of the distorted wave Born approximation (DWBA), which accounts for scattering effects in a reference potential  $V_{short}$  and provides good estimates as long as the difference to the real potential is small and the electron energy is not too low. Within DWBA  $\delta_{short}$  is calculated as [29]:

$$\delta_{short}(k,r) \simeq -\frac{1}{k^2} \int_0^\infty d\varrho F_{l,\eta}^2(k,r) V_{short}(r), \qquad (13)$$

with the bare Coulomb wave function

$$F_{l,\eta}(k,r) = C_l e^{i\varrho} \varrho^{l+1} {}_1 F_1 \left( l+1+i\eta; 2l+2; -2i\varrho \right)$$
(14)

where  $\varrho = kr$  and

$$C_{l} = 2^{l} e^{-\frac{\pi\eta}{2}} \frac{|\Gamma(l+1+i\eta)|}{\Gamma(2l+2)}$$
(15)

For Yukawa and exponential potential terms the integrals in Eq. (13) and the related derivatives in Eq. (12) have the following closed form solutions, as shown in the Appendix:

$$\Delta t_{Yukawa} = \frac{\delta_{Yukawa}}{k^2} \left[ f(k) + g(k) - 1 \right]$$
(16)

where the analytical forms of  $\delta_{Yukawa}$ , f and g are given in Eqs. (A2), (A8) and (A9) and

$$\Delta t_{exp} = \frac{\delta_{exp}}{k} \\ \times \left[ \frac{2i(l+1-i\eta)}{\mu - 2ik} - \frac{i\eta}{k} \log\left(1 - \frac{2i}{\mu}k\right) \right. \\ \left. + \frac{2}{k} \left( l + \frac{\pi\eta}{2} - \frac{\eta}{k} \sum_{n=0}^{\infty} \frac{1}{(l+1+n)^2 + \eta^2} \right) \right. \\ \left. + \frac{c_1(k) - c_2(k) - c_3(k) + c_4(k)}{c_0(k)} \right]$$
(17)

where the analytical forms of  $\delta_{exp}$  and  $c_i$  (i = 0, ..., 4) are given in Eq. (B3), and Eq. (B9) to Eq. (B13), respectively.

In order to test the analytical streaking formula we made use of the following form of the single-activeelectron potentials for electrons in the outermost shell of atoms:

$$V_{SAE}(\mathbf{r}) = -\frac{Z}{r} - \frac{a_1 e^{-\mu_1 r} + a_2 e^{-\mu_2 r}}{r} - a_3 e^{-\mu_3 r}, \quad (18)$$



FIG. 3. (Color online) Comparison of analytical predictions (solid lines) with results of ab-initio calculations (symbols) for the streaking time delay in the case of photoionization of (a) helium (ab-initio data from [10]) and (b) neon atom as a function of the XUV photon energy. Ab-initio results from calculations using the present SAE potential (circles, [20]) and multi-electron *B*-spline *R*-matrix calculations (diamonds, [22]) are shown. Streaking laser parameters as in Fig. 2.

where the parameters for various noble gas neutrals and ions are given in Ref. [30]. The streaking time delay can then be obtained in the same way as for hydrogenlike systems, using the analytical formulas for  $\Delta t_{short}$ ,  $\Delta t_{phase}$  and  $\Delta t_{log}$ . As can be seen in Fig. 3, the analytical predictions are in good overall agreement with results of ab-initio calculations for photoionization of helium and neon atom. We note that in the case of ionization of Ne we have considered that the  $2p \rightarrow d$  transition is strongly dominant over the  $2p \rightarrow s$  one [16].

#### V. RELATION TO WIGNER-SMITH DELAY

In this section we use the analytical formula to study the effect of the streaking field in Eq. (2) on the three contributions to the field-free time delay. To this end, we have determined

$$\Delta_i = \sum_{j=1}^{N} \left[ \frac{E_s(t_j)}{E_s(t_{ion})} - 1 \right] \Delta t_i^{(j)} \tag{19}$$

for each of the three contributions (i = short, phase, log). The corresponding results are shown in Figs. 4, 5 and 6,



FIG. 4. (Color online) Absolute difference  $\Delta_{short}$  as a function of the XUV photon energy for the contribution due to the short-range potential for (a) neon atom and (b) helium atom. Streaking laser parameters as in Fig. 2.



FIG. 5. (Color online) Absolute difference  $\Delta_{phase}$  for photoionization of (a) neon, (b) helium and (c) hydrogen. Streaking laser parameters as in Fig. 2.

respectively. The comparison clearly shows that for the short-range and the Coulomb phase terms the effect of the streaking field is significant for XUV photon energies near the threshold of photoionization only. At photoelectron energies of 10 eV or more the respective differences  $\Delta_i$  are below 1 as. On the other hand the effect of the streaking field on the logarithmic term is much larger, even for photoelectron energies of several tens of eV.

Thus, except for photoelectron energies close to the



FIG. 6. (Color online) Absolute difference  $\Delta_{log}$  for photoionization of (a) neon, and (b) helium. Streaking laser parameters as in Fig. 2.



FIG. 7. (Color online) Comparison of present analytical estimates (solid lines) for  $\Delta t_{CLC}$  with those of an alternative analytical approximation (dashed lines, [12]). Streaking laser parameters as in Fig. 2.

threshold, we can approximate Eq. (2) further as follows:

$$\Delta t_s = \sum_j \frac{E_s(t_j)}{E_s(t_i)} \left[ \Delta t_{short}^{(j)} + \Delta t_{phase}^{(j)} + \Delta t_{log}^{(j)} \right]$$
(20)

$$\simeq \sum_{j} \left[ \Delta t_{short}^{(j)} + \Delta t_{phase}^{(j)} + \frac{E_s(t_j)}{E_s(t_i)} \Delta t_{log}^{(j)} \right]$$
(21)

$$= \Delta t_{short} + \Delta t_{phase} + \sum_{j} \frac{E_s(t_j)}{E_s(t_i)} \Delta t_{log}^{(j)} \qquad (22)$$

$$=\Delta t_{WS} + \Delta t_{CLC} \tag{23}$$

where  $\Delta t_{WS} = \Delta t_{short} + \Delta t_{phase}$  represents the wellknown Wigner-Smith time delay for short range potentials, while  $\Delta t_{CLC} = \sum_j \frac{E_s(t_j)}{E_s(t_i)} \Delta t_{log}^{(j)}$  can be interpreted as the Coulomb-laser-coupling term often used in the analysis of streaking time delays. Indeed, as shown in Fig. 7, the present estimates for  $\Delta t_{CLC}$  are in good agreement with another analytical estimate  $\Delta t_{CLC} = Z[2 - \ln(E_{asym}T_{IR})]/(2E_{asym})^{3/2}$  [12], where  $T_{IR}$  is the streaking field period.

#### VI. SUMMARY

We have presented estimates of the streaking time delay for photoionization of atoms based on an analytical formula. In the derivation of the formula we have made use of the fact that the time delay depends on the propagation of the photoelectron over a finite range in space. This enabled us to evaluate the different contributions to the field-free time delay using single-active electron potentials and the distorted wave Born approximation. Analytical predictions are found to be in good agreement with results of ab-initio calculations. Application of the analytical estimates gave insights into the effect of the streaking field on the short-range, Coulomb phase and logarithmic terms in the field-free time delay and provided an alternative justification of the widely used analysis of the streaking time delay as a sum of Wigner-Smith time delay and Coulomb-laser coupling term.

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#### Appendix A: Short-range time delays for Yukawa potentials

In this section we derive an analytical formula for the field-free time delay in a short range Yukawa potential of the form

$$V_{Yukawa} = \frac{a_Y \exp[-\mu r]}{r} \,. \tag{A1}$$

Using the Distorted Wave Born Approximation the phase shift induced on a l-partial wave in the potential is given by [31]:

$$\delta_{Yukawa} \simeq \frac{a_Y}{4k\Gamma(2l+2)} |\Gamma\left(l+1+i\eta\right)|^2 \exp\left[-\pi\eta\right]$$
(A2)  
 
$$\times \exp\left[2\eta \arctan\left(\frac{2k}{\mu}\right)\right] \left(1+\frac{\mu^2}{4k^2}\right)^{-(l+1)}$$
  
 
$$\times_2 F_1\left(l+1+i\eta, l+1-i\eta; 2l+2; \frac{4k^2}{\mu^2+4k^2}\right)$$

where

$${}_{2}F_{1}(a,b;c;z) = \sum_{n=0}^{\infty} \frac{(a)_{n}(b)_{n} z^{n}}{(c)_{n} n!}$$
$$= \frac{\Gamma(c)}{\Gamma(b)\Gamma(c-b)}$$
(A3)
$$\times \int_{0}^{1} dt \ t^{b-1} (1-t)^{c-b-1} (1-zt)^{-a}$$

is the convergent Gaussian hypergeometric function, given both as series and Euler integral representation,

with

$$(a)_i = \frac{\Gamma(a+i)}{\Gamma(a)}.$$
 (A4)

is the Pochhammer symbol.

Calculation of the corresponding time delay

$$\Delta t_{Yukawa} = \frac{\partial \delta_{Yukawa}}{\partial E} = \frac{1}{k} \frac{\partial \delta_{Yukawa}}{\partial k}$$
(A5)

requires to take the derivative of the convergent Gaussian hypergeometric function, in which three of the four arguments depend on k. To this end, we make use of both representations of the function as follows:

$$\frac{\partial}{\partial k}{}_{2}F_{1}\left(a\left(k\right),b\left(k\right);c;z\left(k\right)\right) = \left(\frac{\partial z}{\partial k}\right)\frac{{}_{2}F_{1}\left(a,b;c;z\right)}{z(1-z)}\left[\left(c-b\right)\frac{{}_{2}F_{1}\left(a,b-1;c;z\right)}{{}_{2}F_{1}\left(a,b;c;z\right)} + \left(b-c+az\right)\right] + \left(\frac{\partial a}{\partial k}\right)\frac{\Gamma(c)}{\Gamma(b)\Gamma(c-b)}\int_{0}^{1}t^{b-1}\left(1-t\right)^{c-b-1}\left(1-tz\right)^{-a}\left[\log\left(\frac{1-t}{t\left(1-tz\right)}\right) + \psi^{(0)}(b) - \psi^{(0)}(c-b)\right]dt$$
(A6)

with

where we used that  $\frac{\partial a}{\partial k} = -\frac{\partial b}{\partial k}$  and  $\psi^{(0)}$  is the polygamma function.

Using this result the time delay is given as:

$$\Delta t_{Yukawa} = \frac{\delta_{Yukawa}}{k^2} \left[ f(k) + g(k) - 1 \right]$$
(A7)

$$f(k) = 2\eta \left[ \frac{\pi}{2} + \frac{2k\mu}{4k^2 + \mu^2} - \arctan\left(\frac{2k}{\mu}\right) - \frac{1}{k} \sum_{n=0}^{\infty} \frac{1}{(l+1+n)^2 + \eta^2} \right] + \frac{(2l+2)\mu^2}{4k^2 + \mu^2}$$
(A8)

and

$$g(k) = 2 \left(l+1+i\eta\right) \left( \frac{{}_{2}F_{1} \left(l+1+i\eta, l-i\eta; 2l+2; \frac{4k^{2}}{\mu^{2}+4k^{2}}\right)}{{}_{2}F_{1} \left(l+1+i\eta, l+1-i\eta; 2l+2; \frac{4k^{2}}{\mu^{2}+4k^{2}}\right)} - \frac{\mu^{2}}{4k^{2}+\mu^{2}} \right) - i\eta \int_{0}^{1} t^{l-i\eta} \left(1-t\right)^{l+i\eta} \left(1-tz\right)^{-(l+1+i\eta)} \left[ \log\left(\frac{\left(1-t\right)\left(4k^{2}+\mu^{2}\right)}{\left(4k^{2} \left(1-t\right)+\mu^{2}\right)t}\right) - \sum_{n=0}^{\infty} \frac{2i\eta}{\left(l+1+n\right)^{2}+\eta^{2}} \right] dt \quad (A9)$$

where we have replaced the polygamma function by its respective series representation. We also note that the full derivative of the hypergeometric function can alternatively be written as a series (as opposed to the integral), however the integral converges at a much higher rate than the series.

### Appendix B: Short-range time delay for exponential decay potentials

In this section we derive an analytical formula for the field-free time delay in a short range potential of the form

$$V_{exp} = a_{exp} \exp[-\mu r]. \tag{B1}$$

The corresponding phase shift  $\delta_{exp}$  has a closed form expression which can be represented in terms of the Appell  $F_2$  function as [32]

$$\delta_{exp} = \frac{a_{exp}(2l+2)|\Gamma(l+1+i\eta)|^2 \exp[-\pi\eta]k^{2l}}{\Gamma(2l+2)\mu^{2l+3}} F_2\left(2l+3; l+1+i\eta, l+1-i\eta; 2l+2, 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{\mu}\right)$$
(B2)

$$= \frac{a_{exp}(2l+2)|\Gamma(l+1+i\eta)|^2 \exp[-\pi\eta]k^{2l}}{\Gamma(2l+2)\mu^{2l+3}} \left(1 - \frac{2ik}{\mu}\right)^{-(l+1-i\eta)}$$

$$\times \left\{ F_1\left(l+1+i\eta; l+2+i\eta, l+1-i\eta; 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right) -\frac{l+1-i\eta}{2l+2} \left(\frac{2ik}{2ik-\mu}\right) F_1\left(l+1+i\eta; l+2+i\eta, l+2-i\eta; 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right) \right\}.$$
(B3)

where in the second equation we expanded the  $F_2$  function in the more common Appell  $F_1$  series as [32]

$$F_{2}\left(c+s;a,a';c,c-p;\frac{k}{h};\frac{k'}{h}\right) = \left(1-\frac{k'}{h}\right)^{-a'}\sum_{m=0}^{s+p}\frac{(a')_{m}(-s-p)_{m}}{(c-p)_{m}m!}\left(1-\frac{h}{k'}\right)^{-m} \times F_{1}\left(a;c+s-a',m+a';c;\frac{k}{h};\frac{k}{h-k'}\right)$$
(B4)

where

$$F_{1}(\alpha;\beta_{1},\beta_{2};\gamma;x,y) = \sum_{m,n}^{\infty} \frac{(\alpha)_{m+n}(\beta_{1})_{m}(\beta_{2})_{n}}{(\gamma)_{m+n}m!n!} x^{m}y^{n}$$

$$= \frac{\Gamma(\gamma)}{\Gamma(\alpha)\Gamma(\gamma-\alpha)} \int_{0}^{1} t^{\alpha-1}(1-t)^{\gamma-\alpha-1}(1-xt)^{-\beta_{1}}(1-yt)^{-\beta_{2}}dt.$$
(B5)

given both as series and Euler integral representations.

Calculation of the corresponding time delay

$$\Delta t_{exp} = \frac{\partial \delta_{exp}}{\partial E} = \frac{1}{k} \frac{\partial \delta_{exp}}{\partial k}$$
(B6)

requires to take the derivative of the Appell series, in which all of the arguments except  $\gamma$  depend on k. To this end, we make use of both representation of  $F_1$  to get:

$$\frac{\partial}{\partial k}F_{1}\left(\alpha(k);\beta_{1}(k),\beta_{2}(k);\gamma,x(k);y(k)\right) = \left(\frac{\partial x}{\partial k}\right)\frac{\alpha\beta_{1}}{\gamma}F_{1}\left(\alpha+1;\beta_{1}+1,\beta_{2};\gamma+1;x,y\right) + \left(\frac{\partial y}{\partial k}\right)\frac{\alpha\beta_{2}}{\gamma}F_{1}\left(\alpha+1;\beta_{1},\beta_{2}+1;\gamma+1;x,y\right) \quad (B7) + \frac{\partial\alpha}{\partial k}\frac{\Gamma(\gamma)}{\Gamma(\alpha)\Gamma(\gamma-\alpha)} \times \int_{0}^{1}t^{\alpha-1}\left(1-t\right)^{\gamma-\alpha-1}\left(1-tx\right)^{-\beta_{1}}\left(1-ty\right)^{-\beta_{2}}\left[\log\left(\frac{t(1-ty)}{(1-t)(1-tx)}\right) + \psi^{(0)}(\gamma-\alpha) - \psi^{(0)}(\alpha)\right]dt$$

where we used that  $\frac{\partial \alpha}{\partial k} = \frac{\partial \beta_1}{\partial k} = -\frac{\partial \beta_2}{\partial k}$ 

Using this result the time delay is given by:

$$\Delta t_{exp} = \frac{\delta_{exp}}{k} \left\{ \frac{2i(l+1-i\eta)}{\mu - 2ik} - \frac{i\eta}{k} \log\left(1 - \frac{2i}{\mu}k\right) + \frac{2}{k} \left(l + \frac{\pi\eta}{2} - \frac{\eta}{k} \sum_{n=0}^{\infty} \frac{1}{(l+1+n)^2 + \eta^2}\right) + \frac{c_1(k) - c_2(k) - c_3(k) + c_4(k)}{c_0(k)} \right\}$$
(B8)

with

$$c_{0}(k) = F_{1}\left(l+1+i\eta; l+2+i\eta, l+1-i\eta; 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right)$$
(B9)  
$$-\frac{l+1-i\eta}{2l+2} \left(\frac{2ik}{2ik-\mu}\right) F_{1}\left(l+1+i\eta; l+2+i\eta, l+2-i\eta; 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right)$$
(B9)  
$$c_{1}(k) = \frac{2i}{(2l+2)(2ik-\mu)} \left(\frac{\mu(l+1-i\eta)}{2ik-\mu} - i\eta\right) F_{1}\left(l+1+i\eta; l+2+i\eta, l+2-i\eta; 2l+2; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right)$$
(B10)

$$c_{2}(k) = \frac{2i(l+1+i\eta)}{2l+2} \left\{ \frac{l+2+i\eta}{\mu} F_{1}\left(l+2+i\eta; l+3+i\eta, l+1-i\eta; 2l+3; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right) + \frac{\mu(l+1+i\eta)}{\mu} F_{1}\left(l+2+i\eta; l+2+i\eta; l+2+i\eta; l+2-i\eta; 2l+3; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu}\right) \right\}$$
(B11)

$$c_{3}(k) = \frac{4k|l+1+i\eta|^{2}}{(2l+2)^{2}(2ik-\mu)} \left\{ \frac{l+2+i\eta}{\mu} F_{1} \left( l+2+i\eta; l+3+i\eta, l+2-i\eta; 2l+3; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu} \right) + \frac{\mu(l+2-i\eta)}{(2ik-\mu)^{2}} F_{1} \left( l+2+i\eta; l+2+i\eta, l+3-i\eta; 2l+3; \frac{-2ik}{\mu}; \frac{2ik}{2ik-\mu} \right) \right\}$$
(B12)

$$c_{4}(k) = \delta F_{1} \left( l + 1 + i\eta; l + 2 + i\eta, l + 1 - i\eta; 2l + 2; \frac{-2ik}{\mu}; \frac{2ik}{2ik - \mu} \right) - \frac{l + 1 - i\eta}{2l + 2} \left( \frac{2ik}{2ik - \mu} \right) \delta F_{1} \left( l + 1 + i\eta; l + 2 + i\eta, l + 2 - i\eta; 2l + 2; \frac{-2ik}{\mu}; \frac{2ik}{2ik - \mu} \right)$$
(B13)

where

$$\delta F_1(\alpha;\beta_1,\beta_2;\gamma;x;y) = \frac{\partial \alpha}{\partial k} \frac{\Gamma(\gamma)}{\Gamma(\alpha)\Gamma(\gamma-\alpha)}$$

$$\times \int_0^1 t^{\alpha-1} (1-t)^{\gamma-\alpha-1} (1-tx)^{-\beta_1} (1-ty)^{-\beta_2} \left[ \log\left(\frac{t(1-ty)}{(1-t)(1-tx)}\right) + \psi^{(0)}(\gamma-\alpha) - \psi^{(0)}(\alpha) \right] dt$$
(B14)

- F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009).
- [2] T. Popmintchev, M.-C. Chen, P. Arpin, M.M. Murnane, and H.C. Kapteyn, Nat. Photon. 4, 822 (2011).
- [3] L. Gallmann, C. Cirelly, and U. Keller, Annu. Rev. Phys. Chem. 63, 447 (2012).
- [4] M.J.J. Vrakking, Phys. Chem. Chem. Phys. 16, 2775 (2014).
- [5] L.-Y. Peng, W.-C. Jiang, J.-W. Geng, W.-H. Xiong, and Q. Gong, Phys. Rep. 575, 1 (2015).
- [6] K. Ramasesha, S.R. Leone, and D.M. Neumark, Annu. Phys. Rev. Chem. 67, 41 (2016).
- [7] F. Calegari, G. Sansone, S. Stagira, C. Vozzi, and M. Nisoli, J. Phys. B: At. Mol. Opt. Phys. 49, 062001 (2016).
- [8] J. Itatani, F. Quéré, G. L. Yudin, M. Y. Ivanov, F. Krausz, and P. B. Corkum, Phys. Rev. Lett. 88, 173903 (2002).
- [9] J. M. Dahlström, A. L'Huillier, and A. Maquet, J. Phys. B: At. Mol. Opt. Phys. 45, 183001 (2012).
- [10] R. Pazourek, S. Nagele, and J. Burgdörfer, Faraday Discuss. 163, 353 (2013).
- [11] A. Maquet, J. Caillat, and R. Taieb, J. Phys. B: At. Mol. Opt. Phys. 47, 204004 (2014).

- [12] R. Pazourek, S. Nagele, and R. Burgdörfer, Rev. Mod. Phys. 87, 765 (2015).
- [13] E. P. Wigner, Phys. Rev. 98, 145 (1955).
- [14] F. T. Smith, Phys. Rev. 118, 349 (1960).
- [15] M. Schultze, M. Fieß, N. Karpowicz, J. Gagnon, M. Korbman, M. Hofstetter, S. Neppl, A. L. Cavalieri, Y. Komninos, T. Mercouris, C. A. Nicolaides, R. Pazourek, S. Nagele, J. Feist, J. Burgdörfer, A. M. Azzeer, R. Ernstorfer, R. Kienberger, U. Kleineberg, E. Goulielmakis, F. Krausz, and V. S. Yakovlev, Science **328**, 1658 (2010).
- [16] A. S. Kheifets and I. A. Ivanov, Phys. Rev. Lett. 105, 233002 (2010).
- [17] C.-H. Zhang and U. Thumm, Phys. Rev. A 82, 043405 (2010).
- [18] S. Nagele, R. Pazourek, J. Feist, K, Doblhoff-Dier, C. Lemell, K. Tökési, and J. Burgdörfer, J. Phys. B: At. Mol. Opt. Phys. 44, 081001 (2011).
- [19] M. Ivanov and O. Smirnova, Phys. Rev. Lett. 107, 213605 (2011).
- [20] S. Nagele, R. Pazourek, J. Feist, and J. Burgdörfer, Phys. Rev. A 85, 033401 (2012).
- [21] J. M. Dahlström, D. Guénot, K. Klünder, M. Gisselbrecht, J. Mauritsson, A. L'Huillier, A. Maquet, and R. Taiëb, Chem. Phys. **414**, 53 (2013).

- [22] J. Feist, O. Zatsarinny, S. Nagele, R. Pazourek, J. Burgdörfer, X. Guan, K. Bartschat, and B.I. Schneider, Phys. Rev. A 89, 033417 (2014).
- [23] R. Pazourek, S. Nagele, and J. Burgdörfer, J. Phys. B: At. Mol. Opt. Phys. 48, 061002 (2015).
- [24] H. Wei, T. Morishita, and C.D. Lin, Phys. Rev. A 93, 053412 (2016).
- [25] M. Ossiander, F. Siegrist, V. Shirvanyan, R. Pazourek, A. Sommer, T. Latka, A. Guggenmos, S. Nagele, J. Feist, J. Burgdörfer, R. Kienberger, and M. Schultze, Nature Phys. 13, 280 (2017).
- [26] J. Su, H. Ni, A. Becker, and A. Jaroń-Becker, Phys. Rev. A. 88, 023413 (2013).
- [27] J.R. Taylor, *Scattering Theory*. Dover Publications Inc. (2006).
- [28] M. Abramowitz and I. Stegun, Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables, Dover Publications Inc. (1965).
- [29] A. Messiah, Quantum Mechanics, John Wiley and Sons, New York (1966).
- [30] X.M. Tong and C.D. Lin, J. Phys. B: At. Mol. Opt. Phys. 38, 15 (2005).
- [31] S. Klarsfeld, Il Nuovo Cimento 40, 1078 (1966).
- [32] N. Saad and R.L. Hall, J. Phys. A 36, 7771 (2003).