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Analysis of absorption time delays in streaking of resonant and non-resonant two-photon ionization

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Abstract

We theoretically study the determination of the time delay between the subsequent absorption of photons in two-photon ionization with the attosecond streaking technique. An analytical model for the absorption time delay based on analysis of the process using perturbation theory is obtained. Model predictions at central frequencies that are resonant and non-resonant with an intermediate bound state agree well with results of numerical streaking simulations, if the intermediate state is well-isolated from other states and the effect of the streaking field on the intermediate state is negligible.

Keywords: attosecond physics, ultrafast laser science, time delays

(Some figures may appear in colour only in the online journal)

1. Introduction

The development of attosecond (1 as $= 10^{-18}$ s) laser pulse technology (for review, see e.g., [1, 2]) opens the perspective to temporally resolve electron dynamics in matter, such as atoms, molecules and solids (e.g., [3–7]). Attosecond streaking [8] is one of the methods to obtain temporal information about ultrafast electronic processes such as photoionization. In the streak camera technique a linearly polarized isolated attosecond pulse is applied along with a moderately strong femtosecond near-infrared laser pulse. The photoelectron, liberated into the continuum by the attosecond pulse, experiences a momentum shift due to the presence of the near-infrared laser pulse. Since the magnitude of the shift depends on the vector potential at the time of ionization and the subsequent propagation of the electron wave packet, a streaking trace of the momentum, or energy, of the photoelectron as a function of the time delay between the streaking pulse and the ionizing XUV pulse can be obtained. By comparing the streaking trace to the oscillation of the vector potential a temporal shift, called streaking time delay, can be obtained (see e.g., [9–11]). The streaking time delay can be further analyzed (e.g., [4, 11–23]) via a sum of the Wigner–Smith time delay [24, 25] and a Coulomb-laser-coupling time delay, or by classical electron trajectory calculations [14, 19, 26, 27].

Recently, it has been proposed to apply the streaking principle to the temporal resolution of electron dynamics in two-photon resonant ionization [28, 29] and double ionization [21]. In both cases it has been shown that the time delay between the subsequent absorption of the two photons can be retrieved from the measurement of the streaking time delay, and it depends linearly on the duration of the ionizing pulse. In order to understand whether or not these results can be used for advanced time-resolved observations or spectroscopic measurements, further analysis is needed. In this article we therefore study the streaking of two-photon ionization at resonant and non-resonant frequencies. To this end we consider the ionization via a resonance using perturbation theory and numerical simulations, mainly within a model system.

We make use of the following parametrization of the streaking time delay Δt_s, as proposed for a two-photon

absorption process before [28]:

$$\Delta t_s = \Delta t_{abs} + \Delta t_c. \quad (1)$$

Here, Δt_c is the time delay acquired by a photoelectron after liberation in the continuum with a certain final kinetic energy. This contribution has been interpreted as the time delay accumulated by the electron during the propagation in the combined fields generated by the Coulomb potential of the parent ion and the streaking field, as compared to the propagation of a free electron, and is therefore labeled 'continuum time delay' (Δt_c). It occurs in both single-photon ionization and two-photon ionization and is rather well understood in terms of the Wigner–Smith time delay and a Coulomb-laser coupling term, as mentioned above. Since results of numerical simulations for the streaking time delay for resonant two-photon absorption have been found to be significantly larger than theoretical predictions for Δt_c [28], it has been proposed that there is a second contribution to the time delay that is acquired during the transition from the initial state to the continuum due to the absorption of the two photons and that has been labeled 'absorption time delay' (Δt_{abs}). The latter contribution may be interpreted as the time of propagation in intermediate states between two successive one-photon transitions. Similar suggestions for additional time delays (beyond the continuum time delay) have been put forward in the interpretation of time delays in sequential double ionization via absorption of two photons [21] and in photoionization involving a Fano resonance [29].

The paper is organized as follows: in the next section we obtain a model formula for the absorption time delay based on an analysis of two-photon ionization using perturbation theory and briefly review the determination of the delay from numerical streaking simulations. Then, we present predictions of the model formula for two-photon transitions at central frequencies that are resonant and non-resonant with an intermediate bound state and compare those with results of simulations. The article ends with a brief summary. We make use of Hartree atomic units ($e = m_e = \hbar = 1$) unless stated otherwise.

2. Absorption time delay through intermediate resonance

2.1. Analysis using perturbation theory

In this section, we present a model for the absorption delay of a two-photon ionization process from an initial bound state $|i\rangle$ into a final continuum state $|f\rangle$ with respect to parameters of the ionizing attosecond pulse. Assuming the ionizing pulse is sufficiently weak, the amplitude c_f of the final state can be written within second order perturbation theory as [30]:

$$c_f(\omega) = \not{\Sigma}_m \int_{-\infty}^{\infty} \mu_{fm} e^{i\delta_f t} E(t) \int_{-\infty}^{t} \mu_{mi} e^{i\delta_m t'} E(t') dt' dt, \quad (2)$$

where μ_{jk} is the transition dipole moment between states $|j\rangle$ and $|k\rangle$, $E(t)$ is the field envelope and ω is the central frequency of the XUV pulse, $\delta_f = E_f - E_m - \omega$, and $\delta_m = E_m - E_i - \omega$ with $E_f = E_i + 2\omega$. The sum with integral represents the expansion of the intermediate state over all field-free states of the target atom or molecule. Equation (2) can be solved analytically for a Gaussian field envelope $E(t) = E_0 e^{-t^2/2T^2}$ [30]

$$c_f = \pi E_0^2 T^2 \not{\Sigma}_m \mu_{fm}\mu_{mi}\left[e^{-\delta_m^2 T^2} - i\frac{2}{\sqrt{\pi}} F(\delta_m T)\right], \quad (3)$$

where $F(x) = e^{-x^2}\int_0^x e^{-t^2} dt$ is Dawson's integral. Assuming that the bandwidth of the attosecond pulse predominantly covers only one intermediate (resonant) state $|r\rangle$ we further approximate [30]

$$c_f = \pi E_0^2 T^2 \left[\mu_{fr}\mu_{ri} e^{-\delta_r^2 T^2} - i\frac{2}{\sqrt{\pi}} \not{\Sigma}_m \mu_{fm}\mu_{mi} F(\delta_m T)\right], \quad (4)$$

since the Gaussian term decays quickly as a function of detuning and therefore contributes for the resonant state only.

The absorption time delay is related to the phase of the two-photon transition amplitude as

$$\phi_f = \arg c_f = \arctan\left[\frac{\mathrm{Im}(c_f)}{\mathrm{Re}(c_f)}\right]. \quad (5)$$

Before proceeding, we note that it is only the absolute value of the dipoles which are of concern. The dipole matrix element $\mu_{fm} = |\mu_{fm}|\, e^{i\theta_{fm}}$, involving the transition into the continuum state, has a phase θ_{fm} whose derivative with respect to energy is merely the widely discussed Wigner–Smith delay. This delay, and therefore the corresponding phase, is accounted for in the continuum delay Δt_c, or equivalently does not contribute to the absorption delay. On the other hand, the dipole matrix elements μ_{mi} between bound states are, in general, real. Therefore, we proceed to get the phase related to the absorption time delay as follows:

$$\phi_{abs} = \arctan\left[-\frac{2}{\sqrt{\pi}} \not{\Sigma}_m \frac{|\mu_{fm}\mu_{mi}|\, F(\delta_m T)}{|\mu_{fr}\mu_{ri}|\, e^{-\delta_r^2 T^2}}\right] \quad (6)$$

$$= -\arctan\left[\frac{2}{\sqrt{\pi}}\left(e^{\delta_r^2 T^2} F(\delta_r T) + \not{\Sigma}_{m\neq r} \frac{|\mu_{fm}\mu_{mi}|\, F(\delta_m T)}{|\mu_{fr}\mu_{ri}|\, e^{-\delta_r^2 T^2}}\right)\right], \quad (7)$$

where we have separated the contribution by the resonant state from those by the other states. Assuming that the latter contributions are small, we make a Taylor expansion by considering the closest (dipole allowed) non-resonant state $|b\rangle$ in the sum over m only and obtain:

$$\phi_{abs} = \phi_{abs}^{(\mathrm{res})} + \phi_{abs}^{(\mathrm{non-res})} \quad (8)$$

with

$$\phi_{abs}^{(\mathrm{res})} = -\arctan\left[\frac{2}{\sqrt{\pi}} e^{\delta_r^2 T^2} F(\delta_r T)\right] \quad (9)$$

$$\phi_{abs}^{(\mathrm{non-res})} = -\frac{a_b(\delta_b, \delta_r, T)}{1 + \mathrm{erfi}^2(\delta_r T)} + \frac{2}{\sqrt{\pi}} \frac{a_b^2(\delta_b, \delta_r, T) e^{\delta_r^2 T^2} F(\delta_r T)}{(1 + \mathrm{erfi}^2(\delta_r T))^2}, \quad (10)$$

where

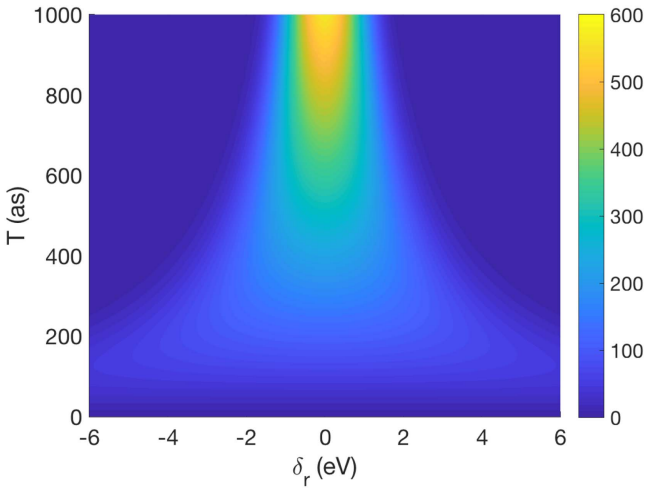

Figure 1. Absorption delay $\Delta t_{abs}^{(\mathrm{res})}$ (in attoseconds) as a function of detuning δ_r and pulse duration T.

$$a_b(\delta_b, \delta_r, T) = \frac{|\mu_{fb}\mu_{bi}|}{|\mu_{fr}\mu_{ri}|} F(\delta_b T) e^{\delta_r^2 T^2}, \tag{11}$$

$\mathrm{erfi}(\delta_r T) = \frac{2}{\sqrt{\pi}} F(\delta_r T) e^{\delta_r^2 T^2}$, and the above expansion was made about $a_b = 0$. The absorption time delay is given by the spectral derivative of ϕ_{abs}:

$$\begin{aligned}\Delta t_{abs}(\delta_r, \delta_b, T) &= \Delta t_{abs}^{(\mathrm{res})}(\delta_r, T) + \Delta t_{abs}^{(\mathrm{non-res})}(\delta_b, \delta_r, T) \\ &= \frac{\partial \phi_{abs}^{(\mathrm{res})}}{\partial E_f} + \frac{\partial \phi_{abs}^{(\mathrm{non-res})}}{\partial E_f} = \frac{\partial \delta_r}{\partial E_f}\frac{\partial \phi_{abs}^{(\mathrm{res})}}{\partial \delta_r} + \frac{\partial \phi_{abs}^{(\mathrm{non-res})}}{\partial E_f} \\ &= \frac{T}{\sqrt{\pi}} \frac{e^{(\delta_r T)^2}}{1 + \mathrm{erfi}^2(\delta_r T)} + \frac{\partial \phi_{abs}^{(\mathrm{non-res})}}{\partial E_f},\end{aligned} \tag{12}$$

where we used $\delta_r = E_r - E_i - \omega = E_r - \frac{1}{2}(E_i + E_f)$. The first term is the contribution from the resonant intermediate state and the second term is the approximative correction due the closest (dipole allowed) non-resonant state. In the present analysis we obtained the latter correction by calculating the corresponding phase and performing the derivative numerically. As we will show below (see figure 3) the correction term is, as expected, typically small and we neglected it in the main part of our analysis (unless mentioned otherwise).

Equation (12) shows that the absorption delay $\Delta t_{abs}^{(\mathrm{res})}(\delta_r, T)$ depends on the detuning from the resonance δ_r and the duration of the ionizing laser pulse T, while it is independent of other laser or target parameters. In figure 1, we present the predictions for $\Delta t_{abs}^{(\mathrm{res})}(\delta_r, T)$ as a function of δ_r and T. As one would expect, for large durations (i.e., small energy bandwidth of the pulse) the absorption delay is non-negligible for small detunings from the resonance only, while the detuning range over which $\Delta t_{abs}^{(\mathrm{res})}(\delta_r, T)$ is significant increases with decrease of the pulse duration.

2.2. Streaking delay simulations

In order to test if the predictions of the model formula, equation (12), are reliable we have performed numerical simulations of a streaking measurement. Since according to our analytical model the general trends for the absorption time delay through a single intermediate resonance (i.e. when $\Delta t_{abs}^{(\mathrm{non-res})} \simeq 0$) are independent of the specific form of the atomic potential, we have chosen a model system for our simulations.

We used the standard Crank–Nicolson method to numerically solve the time-dependent Schrödinger equation (TDSE) on a grid in space and time,

$$i\frac{\partial \Psi(x, t)}{\partial t} = \left[\frac{p^2}{2} + V(x) + (E_{\mathrm{XUV}}(t) + E_s(t))x\right]\Psi(x, t), \tag{13}$$

where p is the momentum operator and $E_i(t) = E_{0,i}(t)\cos(\omega_i t + \phi_i)$ with central frequency ω_i and carrier-envelope phase ϕ_i for both the ionizing (E_{XUV}) and the streaking (E_s) fields. The field amplitudes are given by $E_{0,s}(t) = E_{0,s}\cos^2(\omega t/2N)$ and $E_{0,\mathrm{XUV}}(t) = E_{0,\mathrm{XUV}}e^{-t^2/2T^2}$ with N is the number of cycles of the streaking pulse and the full width half maximum duration of the XUV pulse given as $\Delta t_{1/2} = \sqrt{2\ln(2)}T$.

For the main part of our analysis we have considered a one-dimensional (1D) potential:

$$V(x) = -\frac{Z}{\sqrt{x^2 + a}}, \tag{14}$$

where Z is the effective charge and a is a soft-core parameter. Choosing $Z = 3.0$ and $a = 0.15$, the energies of the three lowest states are $E_1 = -5.32$, $E_2 = -2.31$, and $E_3 = -1.30$. These eigenstates are well separated such that one can selectively study the two-photon absorption into the continuum via a single intermediate resonance. In order to keep the duration of the XUV pulse shorter than the oscillation period of the streaking pulse, we have used a three-cycle streaking field of 2400 nm in wavelength with $I_s = 1.0 \times 10^{11}$ W cm^{-2} and $\phi_s = -\pi/2$. In order to test the predictions of the perturbative analysis in case of nearby additional states, we have also made use of previously obtained data for the absorption delay for resonant two-photon ionization of helium atom [28].

The streaking trace was obtained by changing the relative delay τ between the centers of these two pulses. For each delay τ, the wave function was propagated on the grid for a sufficiently long time until both laser pulses have ceased and the ionizing wave packet could be clearly separated from the remaining bound part of the wave function. The extensions of the grid were chosen to be $[-7000, 7000]$ such that the outgoing wave packet remains on the grid and does not reach the boundaries. The momentum distributions were obtained by performing a Fourier transform of the ionizing wave packet. The final momentum k_f of the photoelectron was obtained as the expectation value of the spectrum. The streaking time delay Δt_s is then quantified by fitting the streaking trace obtained in quantum simulations to

$$k_f(\tau) = k_0 - \alpha A_s(\tau + \Delta t_s), \tag{15}$$

where A_s is the vector potential of the streaking pulse and α is a fitting parameter. In test calculations we have varied the propagation time, i.e. propagated the ionizing wave packet to

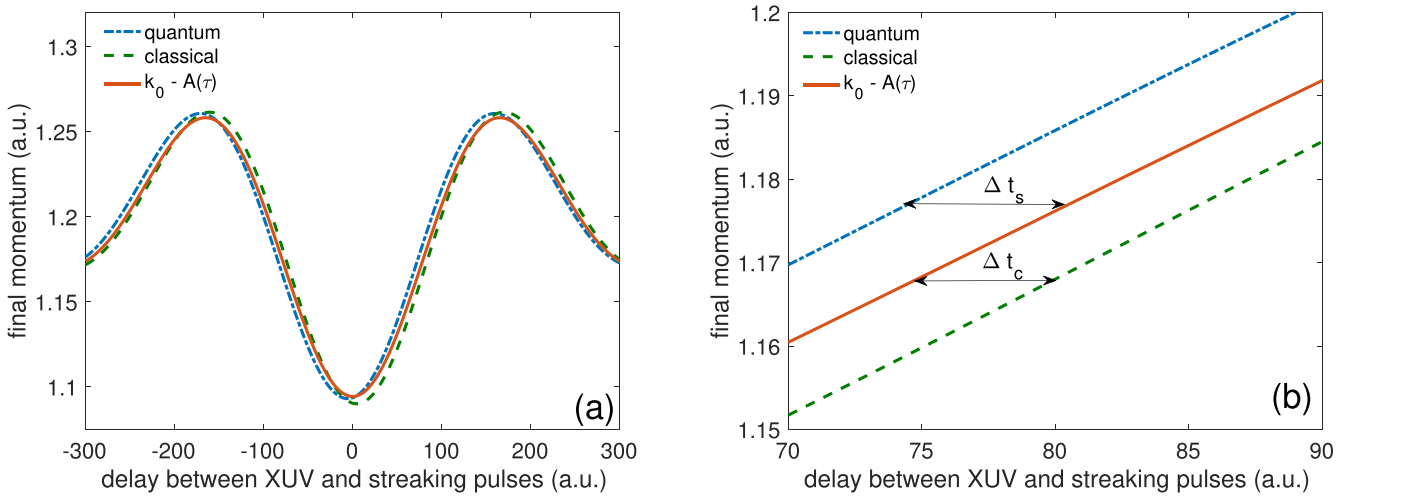

Figure 2. Three streaking traces for a resonant two-photon ionization from numerical streaking simulations (red solid curve), classical electron trajectory calculations (green dashed line), and the reference free-particle streaking formula $k_f^{\rm free}(\tau) = k_0 - A_s(\tau)$ (blue dashed–dotted curve). Panel (b) is an enlargement of the traces in panel (a) to show the streaking (Δt_s) and continuum (Δt_c) time delay. Laser parameters used in the simulations are: $I_{\rm XUV} = 1.0 \times 10^{13}$ W cm^{-2}, $\omega_{\rm XUV} = 81.81$ eV, $T = 550$ as, and $\phi_{\rm XUV} = -\pi/2$; $I_s = 1.0 \times 10^{11}$ W cm^{-2}, $\lambda_s = 2400$ nm, $N_s = 3$, and $\phi_{\rm s} = -\pi/2$.

different distances, until the variations in the photoelectron spectrum, and in particular the retrieved expectation value and streaking time delay, were small (in case of the time delay much smaller than a fraction of an attosecond). This also indicates that the error on the streaking time delay due to the plane wave approximation (in the Fourier transform procedure) can be considered to be negligible.

To obtain the absorption time delay from the numerical results for the streaking delay we need to retrieve the contribution due to the continuum time delay. As discussed in the introduction, from the well-studied streaking of photoionization several techniques are known to obtain Δt_c. One of these techniques makes use of the fact that the propagation in the continuum can be often well described by classical analysis [14]. We have therefore used a classical trajectory method to determine the continuum time delay. To this end, we consider the Newton equation for the dynamics of the electron in the field generated by the Coulomb potential of the residual ion and the streaking field of the linearly polarized laser. The corresponding differential equation has been solved using the fourth-order Runge–Kutta method. As initial conditions, we have chosen the most probable position of the electron in the ground state of the potential and the initial momentum of the electron due to the absorption of a photon in the Coulomb potential. Alternatively, one can sample the initial conditions in Monte-Carlo calculations [14]. The delay between XUV pulse and streaking pulse corresponds to the instant of the appearance of the photoelectron in the continuum during the streaking pulse. We have therefore varied this initial time instant to obtain a classical streaking trace in terms of the classical final momentum. The continuum time delay Δt_c can be then retrieved from this classical streaking trace in the same way as Δt_s from the quantum streaking simulations. For further details of this classical trajectory calculations we refer to [26]. An example of the procedure is shown in figure 2 for a resonant two-photon ionization of the model system. The absorption time delay is then given as $\Delta t_{abs} = \Delta t_s - \Delta t_c$ (see equation (1)).

3. Results and discussion

In this section we compare results from streaking simulations with the predictions within perturbation theory for ionization at central frequencies that are resonant or non-resonant with the first excited state of the model system. Limits as well as special cases will be considered.

3.1. Two-photon ionization at resonant central frequency

Considering resonant two-photon ionization at the central frequency of the pulse, i.e. detuning $\delta_r = 0$, we have performed streaking simulations using the model potential and an ionizing pulse with $\omega = 3.01 = E_2 - E_1$. In figure 3(a), we present the numerically determined absorption time delay $\Delta t_{abs} = \Delta t_s - \Delta t_c$ (circles) as a function of the pulse duration T. The numerical results are compared with the theoretical model predictions for the absorption time delay Δt_{abs} (equation (12), solid line), including both resonant and non-resonant contributions, and the resonant contribution $\Delta t_{abs}^{(\rm res)}$ (dashed line) only. Note that in the case of resonant central frequency $\lim_{\delta_r \to 0} \Delta t_{abs}^{(\rm res)} = \frac{T}{\sqrt{\pi}}$. The absorption time delays extracted from the numerical simulations indeed reproduce the linear dependence on the pulse duration as predicted by perturbation theory. Although the correction from the non-resonant contribution is small (see solid line versus dashed line), its inclusion further improves the agreement between predictions and numerical data.

In figure 3(b), we compare predictions from perturbation theory to TDSE simulations of the absorption delay in 3D helium atom taken from [28]. In the calculations of the dipole matrix elements for the helium atom, we used bound state

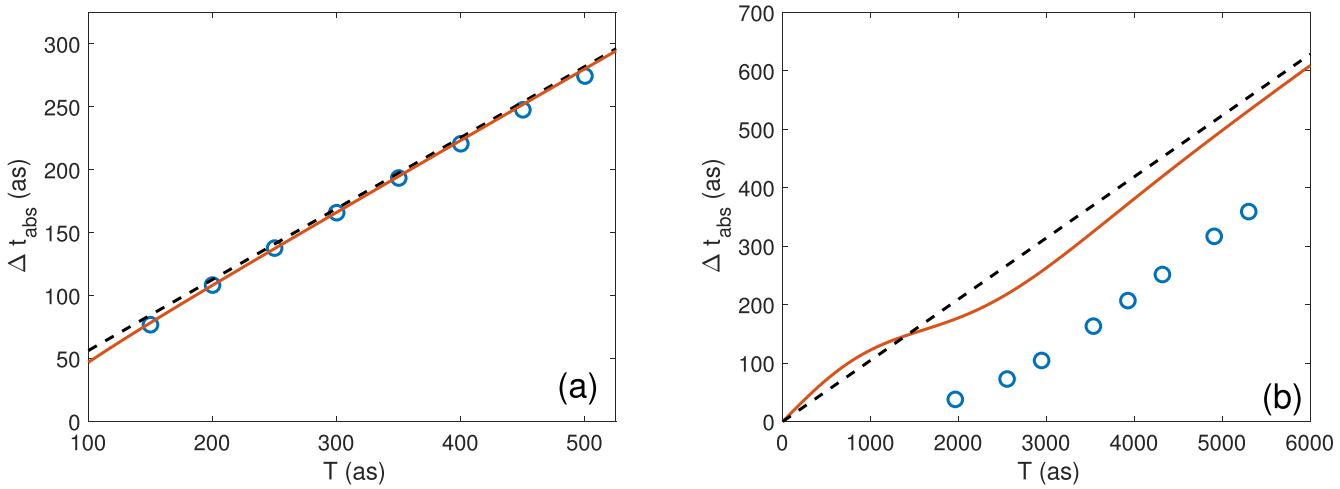

Figure 3. Absorption time delay from numerical simulations (blue circles) and model predictions for $\Delta t_{abs}^{(\mathrm{res})} + \Delta t_{abs}^{(\mathrm{non-res})}$ (red solid line) and $\Delta t_{abs}^{(\mathrm{res})}$ (black dashed line) as a function of pulse duration for a Gaussian XUV pulse tuned into resonance with the first excited state of (a) the model system and (b) helium atom (data for numerical simulations taken from [28]). Laser parameters: $I_{\mathrm{XUV}} = 1.0 \times 10^{13}$ W cm^{-2} (resonant), $\phi_{\mathrm{XUV}} = -\pi/2$; $I_s = 1.0 \times 10^{11}$ W cm^{-2}, $\lambda_s = 2400$ nm, $N_s = 3$, and $\phi_{\mathrm{s}} = -\pi/2$.

Table 1. Results of simulations for the absorption delay (in as) in the model system as a function of the pulse length T (in as) of a two-photon resonantly ionizing pulse and streaking pulses at different wavelengths λ (in nm), intensities I (in W cm^{-2}). Also shown are the predictions $\Delta t_{abs}^{(\mathrm{res})} = T/\sqrt{\pi}$ (in as) and $\Delta t_{abs}^{(\mathrm{res})} + \Delta t_{abs}^{(\mathrm{non-res})}$ (in as) from perturbation theory.

T	$\lambda_s = 2400$ $I_s = 10^{11}$	$\lambda_s = 3200$ $I_s = 10^{11}$	$\lambda_s = 4800$ $I_s = 10^{10}$	$\Delta t_{abs}^{(\mathrm{res})}$	$\Delta t_{abs}^{(\mathrm{res})} + \Delta t_{abs}^{(\mathrm{non-res})}$
250.0	137.79	134.34	140.08	141.05	137.40
350.0	193.47	190.52	195.96	197.47	194.86
450.0	247.66	244.87	250.49	253.89	251.86

wavefunctions in the Coulomb field of an effective charge $Z_e = 27/16$ and the plane wave approximation for the continuum states. The comparison reveals that while the numerical results qualitatively reproduce the linear dependence of the absorption time delay on the pulse duration, there is an almost constant offset. This offset was interpreted before as the influence of the streaking field on the resonant $2p$ state [28], which is not taken into account in the present perturbation model. In the present comparison we however also observe that in the case of the helium atom consideration of the non-resonant correction term (see solid line versus dashed line) has a much larger effect than for the model system (figure 3(a)), where the states are more separated in energy. Thus, it is likely that in the case of helium atom further improvement may require to consider more than one non-resonant state in the analysis, which is beyond the scope of the present analysis.

As mentioned above, in our perturbative model analysis of the absorption time delay we have neglected the influence of the streaking pulse. In case of the model system with the transition through a strongly bound resonant state, this assumption is confirmed by the results of our numerical simulations. For example, in table 1 it is shown that, although the streaking and continuum time delays (not shown) vary with a change of either the wavelength or the intensity of the streaking pulse, the variation of the extracted absorption time delays is less than 5 as. Similarly, we have found that the absorption time delay changes by less than 1 as for a variation of the XUV pulse intensity between 10^{12} and 5×10^{13} W cm^{-2}. This implies that by accurate determination of the continuum time delay one is able to obtain the absorption time delay, independent of the parameters of the applied streaking pulse, if the intermediate resonant state is well-isolated from other states and the effect of the streaking field on the intermediate state is negligible.

3.2. Two-photon ionization at small detunings

For the case of a well-isolated intermediate state, as in the case of the 1D model system, we have also considered how $\Delta t_{abs}^{(\mathrm{res})}$ changes for small detunings, i.e. $\delta_r \neq 0$. The predictions of the perturbative analysis for the absorption delay $\Delta t_{abs}^{(\mathrm{res})}$ as a function of pulse duration T for various detunings δ_r are shown in figure 4. We note that for the 1D model system, the contribution $\Delta t_{abs}^{(\mathrm{non-res})}$ is less than 10 as even for the largest δ_r considered.

One can see that for small T, or more generally small product $\delta_r T$, the absorption delay approximately depends linearly on the pulse duration, as in the on-resonant case. This can be understood by expanding the model formula in a

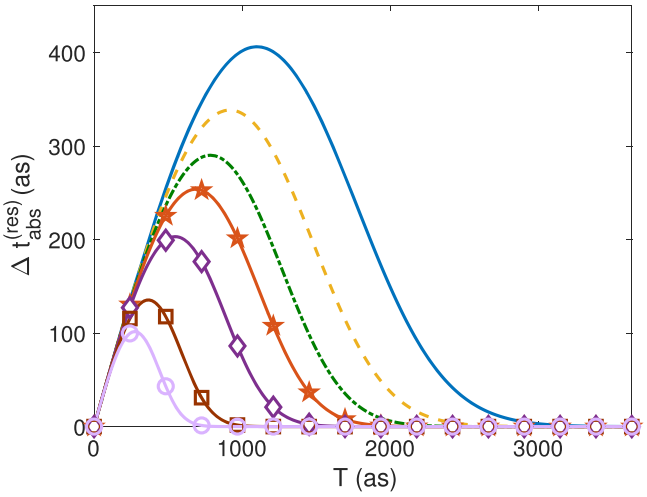

Figure 4. Absorption delay $\Delta t_{abs}^{(\mathrm{res})}$ as a function of pulse duration for detunings $\delta_r = 0.68$ eV (solid line), 0.82 eV (dashed line), 0.95 eV (dash–dot line), 1.09 eV (stars), 1.36 eV (diamonds), 2.04 eV (boxes), and 2.72 eV (circles).

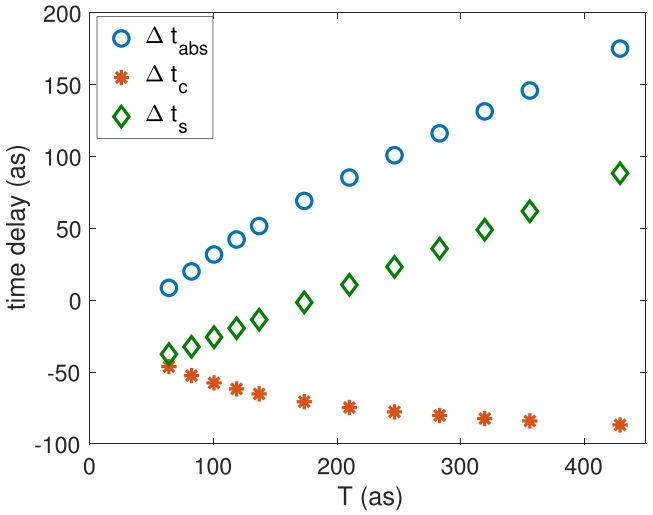

Figure 5. Absorption delay Δt_{abs}, obtained for the 1D model system, as a function of XUV pulse duration, for a detuning δ_r chosen such that the resonance is at FWHM of the XUV pulse.

Taylor series for $x = \delta_r T$ about $x = 0$:

$$\Delta t_{abs}^{(\mathrm{res})}(x) \simeq \frac{1}{\sqrt{\pi}\,\delta_r}\left[x + \left(1 - \frac{4}{\pi}\right)x^3 + O(x^5)\right]. \tag{16}$$

For a given detuning δ_r and sufficiently small T, the first term of the expansion is dominant and the absorption delay is approximately determined by $\Delta t_{abs}^{(\mathrm{res})} = T/\sqrt{\pi}$. Physically, this corresponds to the situation that the frequency for the resonant transition lies well within the bandwidth of the ionizing laser pulse.

On the other hand, we see that the model prediction for the absorption delay $\Delta t_{abs}^{(\mathrm{res})}$ goes to zero for large pulse duration, independent of the detuning δ_r. Physically, this corresponds to the situation that the energy bandwidth of the pulse is sufficiently small and the contributions at the resonant frequency become negligible. In this case the streaking delay is determined by the continuum time delay only, as discussed before [28]. This can be also shown by taking the limit of $\Delta t_{abs}^{(\mathrm{res})}$ for $T \to \infty$ as:

$$\lim_{T\to\infty} \Delta t_{abs}^{(\mathrm{res})}(\delta_r, T) = \frac{1}{\sqrt{\pi}} \lim_{T\to\infty} \frac{\frac{d}{dT}(Te^{\delta_r^2 T^2})}{\frac{d}{dT}(1 + \mathrm{erfi}^2(\delta_r T))} \tag{17}$$

$$= \lim_{T\to\infty} \frac{1 + 2\delta_r^2 T^2}{2\delta_r\,\mathrm{erfi}(\delta_r T)} \tag{18}$$

$$= \lim_{T\to\infty} \frac{\delta_r T^2}{\mathrm{erfi}(\delta_r T)} = 0, \tag{19}$$

where we have repeatedly applied L'Hospital's rule.

Besides these limits, further analysis of $\Delta t_{abs}^{(\mathrm{res})}$ reveals another interesting dependence: if the detuning δ_r is inversely proportional to the pulse duration T, i.e. $\delta_r T = \text{const.}$, the absorption delay depends linearly on T. Since the half-width of a pulse with Gaussian envelope is given by $\Delta\omega_{1/2} = \frac{\sqrt{2\ln 2}}{T}$, we therefore expect $\Delta t_{abs}^{(\mathrm{res})} \propto T$ for $\delta_r = \Delta\omega_{1/2}$. In figure 5, we present results of TDSE simulations for the streaking, continuum and absorption delay as a function of the pulse duration for this case using the 1D model system. Indeed, the linear relationship of $\Delta t_{abs}^{(\mathrm{res})}$ on the pulse duration is captured by the *ab-initio* calculations of Δt_{abs} for pulse durations $T > 200$ as. For shorter pulse durations we observe a deviation from the linear trend, since the corresponding broad bandwidth of the pulse covers more than one resonance.

4. Summary

We have studied streaking of two-photon ionization by determining the absorption time delay, which accounts for the delay between the subsequent absorption of the two photons during the transition of the photoelectron from the ground state into the continuum. To this end, we have used perturbation theory to obtain a model formula for the absorption delay in the case of a well-isolated intermediate resonance. Predictions of the model formula, at central frequencies that are resonant and non-resonant with the first excited state of a model system, are in good agreement with the results of numerical streaking simulations. Comparisons with numerical data for the helium atom indicate that further improvement may require the careful consideration of non-resonant states as well as the effect of the streaking pulse on the resonant intermediate state.

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