PAPER

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Effect of attochirp on attosecond streaking time delay in photoionization of atoms

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Abstract

We present a theoretical analysis of the effect of the attochirp on the streaking time delay, intrinsic to photoionization of an atom by an attosecond laser pulse at extreme ultraviolet wavelengths superposed by a femtosecond streaking pulse. To this end, we determine the expectation value of the delay in a chirped pulse using a recently developed model formula. Results of our calculations show that the attochirp can be relevant for photoemission from the $3p$ shell in argon atom at frequencies near the Cooper minimum, while it is negligible if the photoionization cross section as a function of frequency varies smoothly.

Keywords: attosecond, ionization, laser pulses

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

1. Introduction

The progress of ultrashort laser pulse technology towards the development of isolated attosecond pulses and attosecond pulse trains (for reviews, see e.g., [1, 2]) has enabled studies of electron dynamics in atomic, molecular and condensed matter on their natural time scale (e.g., [3–6]). Various spectroscopic methods (see e.g., [7–11]) have been developed to perform attosecond measurements. Among these attosecond streaking [12], reconstruction of attosecond beating by interference of two-photon transitions (RABBITT, [13]) and atto-ARPES (angle-resolved photoemission spectroscopy) [14] have been applied to measure ultrashort time delays, given as the energy derivative of the phase accumulated by the electron in photoionization.

The streaking time delay is determined by superposing an attosecond extreme ultraviolet (XUV) pulse, which ionizes the target, with a femtosecond (near-)infrared laser pulse, which interacts with the emitted electron and shifts the energy (or momentum) of the electron. By varying the relative time delay of the two pulses a streaked energy spectrum is obtained. An absolute time delay is given via comparison of the spectrum with the vector potential of the femtosecond streaking pulse, or a relative time delay can be extracted by comparison between the spectra for electrons originating from different states within the target [3, 4] or from different targets [15].

The interpretation of attosecond time delay measurements has generated much interest, since only a fraction of the relative delay of 21 as measured for the promotion of photoelectrons from the $2p$ and $2s$ orbitals in neon atom [4] could be accounted for in theoretical predictions [4, 16–19]. Results of these and further theoretical studies (e.g., [20–34]) are often analyzed via a sum of the Wigner–Smith delay [35–37] and a term related to the coupling between streaking laser field and the long-range Coulomb potential. Besides the difficulty to account for the full electron dynamics, e.g. multi-electron effects, in the theoretical analysis and calculations, the sensitivity of the measured results on the retrieval method and the laser parameters have been considered as reasons for the discrepancy between theory and experiment.

Recently, it has been suggested that the streaking time delay [15] and other time measurements such as RABBITT [38, 39] may depend on the chirp of the attosecond XUV pulse. In the case of streaking this was related to the presumption that different times for the transition of the photoelectron in the continuum during the interaction with the ionizing pulse would correspond to different kinetic energies

of the photoelectron. Previous theoretical analysis focused on analyzing the measurement method to retrieve the Wigner–Smith time delay from streaked energy spectra in chirped pulses [33, 39]. It was demonstrated that the spectrogram itself is sensitive to the attochirp of the XUV pulse and for chirped pulses the retrieved time delays may depend on the accuracy of the retrieval algorithm and/or the central momentum approximation, in which the momentum of the photoelectron is approximated by the center momentum of the unstreaked spectrum.

In this work we present results of a complementary study of the attochirp effect on the attosecond streaking time delay. Instead of focusing on the method to extract the Wigner–Smith time delay from an observed streaked energy spectrum, we study how the streaking time delay itself, intrinsic to the photoionization process in the superposition of the two pulses, depends on the chirp of the attosecond XUV pulse. To his end, in the next section we use a recently developed model formula [40] based on previous classical electron trajectory calculations [41], that provides an efficient method to estimate the time delay. The formula is then applied to determine the expectation value of the delay in a chirped laser pulse. In section 3 we present results for neon and argon atoms for different values of the chirp parameters. We discuss the significance of phenomena such as the Cooper minimum [42], since it has been previously pointed out that the time delay is particularly sensitive near the Cooper minimum [43]. The articles ends with a brief summary. Hartree atomic units ($e = m = \hbar = 1$) are used throughout unless stated otherwise.

2. Modeling chirp effect on streaking time delay

Recently, we have proposed an efficient estimate of the streaking time delay Δt_s in photoionization of atoms via an analytical formula [40, 41]

$$\Delta t_s(\omega, t_i) = \sum_{j=1}^{N} \frac{E_s(t_j)}{E_s(t_i)} \Delta t_{\rm ff}^{(j)}(\omega), \tag{1}$$

where $E_s(t)$ is the amplitude of the streaking field at time t, t_i is the time of transition of the photoelectron in the continuum and

$$\Delta t_{\rm ff}^{(j)}(\omega) = \Delta t_{\rm ff}(\omega; k_{j+1}, r_{j+1}) - \Delta t_{\rm ff}(\omega; k_j, r_j) \tag{2}$$

with $\Delta t_{\rm ff}(\omega; k, r)$ is the field-free time delay accumulated by the photoelectron during the propagation from the origin of the transition into the continuum by the ionizing XUV pulse to r with momentum $k = k(r) = \sqrt{2(E_{\rm asym} - V(r))}$. Here, $E_{\rm asym} = \omega - I_p$ is the asymptotic energy of the photoelectron, ω is the photon energy of the ionizing XUV field, and I_p is the ionization potential of the atom. The field-free time delay can be decomposed into three well-known terms:

$$\begin{aligned}\Delta t_{\rm ff}(\omega; k, r) = \Delta t_{\rm short}(\omega; k, r) + \Delta t_{\rm phase}(\omega; k, r) \\ + \Delta t_{\rm log}(\omega; k, r),\end{aligned} \tag{3}$$

where $\Delta t_{\rm short}$, $\Delta t_{\rm phase}$ and $\Delta t_{\rm log}$ are the contributions due to the short-range part of the potential, the Coulomb phase and the logarithmic term. Equation (1) shows that in a laser pulse of finite duration the time delay is accumulated over a finite distance, which allows for the analytical estimates of all three contributions [40]. For photoelectron energies $E_{\rm asym}$ not too close to the threshold the effect of the streaking field on the first two contributions is negligible and equation (1) can be further approximated by

$$\Delta t_s(\omega, t_i) = \Delta t_{\rm WS}(\omega) + \sum_{j=1}^{N} \frac{E_s(t_j)}{E_s(t_i)} \Delta t_{\rm log}^{(j)}(\omega), \tag{4}$$

where $\Delta t_{\rm WS} = \Delta t_{\rm short} + \Delta_{\rm phase}$ represents the Wigner–Smith time delay [35–37] and the second term can be interpreted as the Coulomb-laser-coupling term often part of the commonly used analysis of the streaking time delay [16, 19–34]. The Wigner–Smith time delay can be either estimated analytically [40] or determined using literature values based on advanced calculations (e.g., [43, 44]). In the present work we have used the data for the Wigner–Smith time delay obtained by Kheifets [44]. The logarithmic term can be calculated analytically using

$$\Delta t_{\rm log}^{(j)}(\omega) = Z\left[\frac{1 - \ln(2k_{j+1}r_{j+1})}{k_{j+1}^3} - \frac{1 - \ln(2k_j r_j)}{k_j^3}\right], \tag{5}$$

where Z is the charge of the residual ion. The finite distances r_j can be estimated via the recursive relation:

$$r_{j+1} = r_j + k(r_j)\delta t \tag{6}$$

with $r_1 = r_{\rm ion}$ is the initial position of the photoelectron after the transition in the continuum and $k(r_1) = \sqrt{2E_{\rm asym} - V(r_1)}$. In our calculations the interval $\delta t = t_{j+1} - t_j$ has been varied until convergence for Δt_s is achieved, and r_1 is chosen as the most probable radius of the electron in the state of the atom. For the potentials we used single-active-electron models.

In figure 1 we present results of the analytical estimates (lines) for the streaking time delay for photoemission from neon atom ((a) $2p$ and (b) $2s$) and argon atom ((c) $3p$ and (d) $3s$) as a function of the XUV central frequency. In the calculations we assumed a Gaussian XUV laser pulse with FWHM pulse duration of $\tau_{\rm FHWM} = 40$ as and used a streaking pulse with $\sin^2$ -envelope consisting of 3 cycles. The maxima of the two fields were set to coincide in time.

Estimates for the streaking delays for three different times of transition of the photoelectron in the continuum during the XUV laser pulse, i.e. $t_i = -20$ as (dash-dotted line), $t_i = 0$ (solid line), and $t_i = 20$ as (dashed line), are compared. The comparison shows that the streaking time delay does not depend significantly on the time of transition, since the strength of the streaking pulse does not vary much over the interval of 40 as. The estimates of the delay for the neon atom are in good agreement with results from *ab initio* simulations (diamonds, [19]). The current model formula however does not provide an improvement concerning the discrepancies between experimental data and theoretical predictions, e.g. concerning the relative delay between photoemission from $2p$ and $2s$ orbitals in neon atom. We note that predictions of the model formula are expected [40, 41] to be less reliable for XUV central frequencies near the ionization

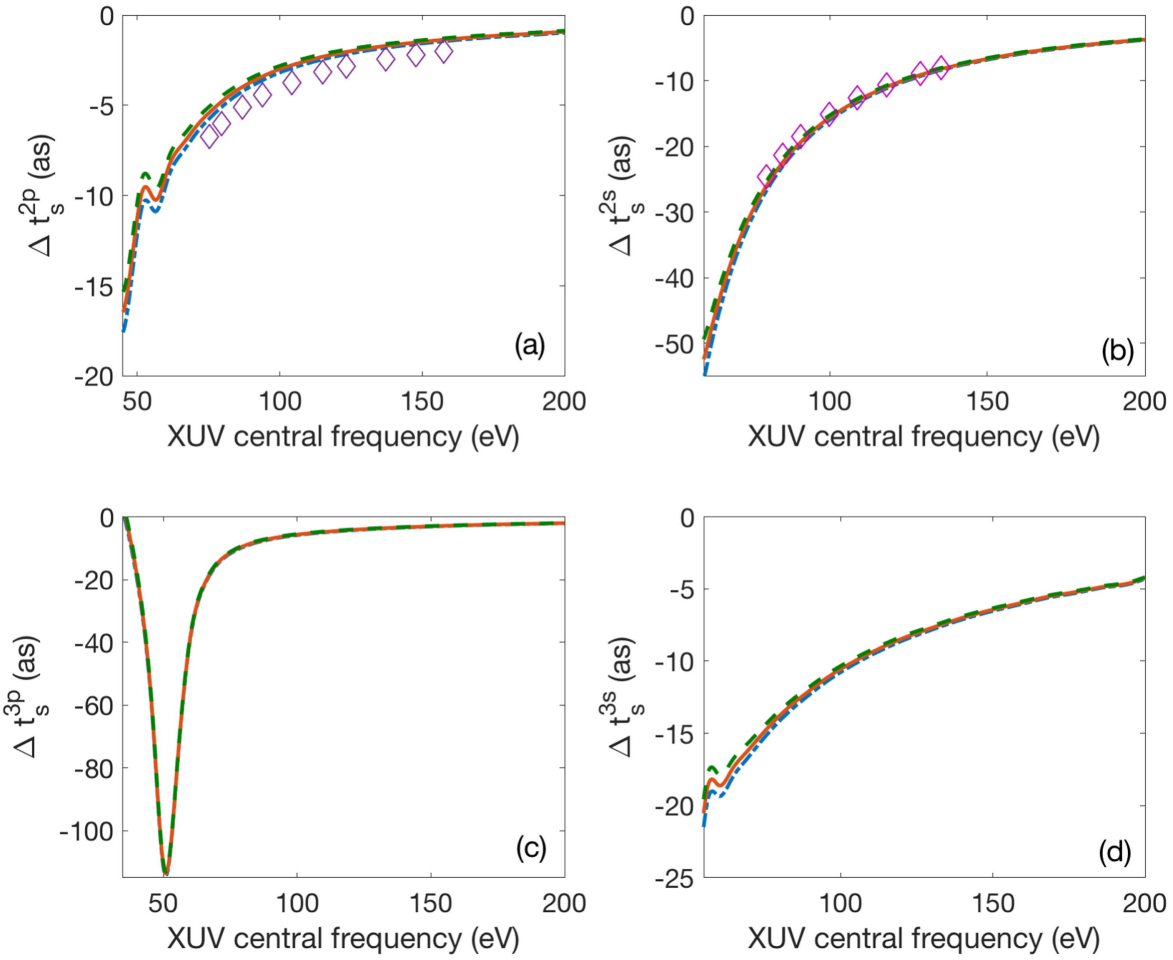

Figure 1. Estimates of the streaking delay for neon ((a) 2*p*, (b) 2*s*) and argon ((c) 3*p*, (d) 3*s*) given by equation (4), as a function of the central XUV frequency for times of the transition in the continuum, $t_i = -\tau_{\mathrm{FWHM}}/2$ (dash-dotted line), $t_i = 0$ (solid line), and $t_i = \tau_{\mathrm{FWHM}}/2$ (dashed line) are compared. Also shown are results from *ab initio* calculations (diamonds) from [19]. In all calculations a streaking pulse of wavelength $\lambda_s = 800$ nm, with $\sin^2$ -envelope consisting of 3 cycles, and peak intensity $I_s = 1 \times 10^{12}$ W cm^{-2} has been considered.

threshold. We therefore restrict ourselves in the frequency range of the attosecond pulse in the present analysis.

While the time delays for Ne and Ar(3*s*) increase rather smoothly as a function of XUV frequency, a pronounced minimum appears around the Cooper minimum for Ar(3*p*). The minimum arises from the Wigner–Smith time delay and is related to the change of sign in the dipole matrix element [43–47].

To study the effect of the attochirp using equation (4) we determine the expectation value of the streaking time delay in a chirped laser pulse as

$$\langle \Delta t_s \rangle(\omega_0, b) = \int \mathrm{d}\omega \int \mathrm{d}t_i P(\omega, \omega_0, t_i, b) \Delta t_s(\omega, t_i), \qquad (7)$$

where $P(\omega, \omega_0, t_i, b)$ is the probability distribution for a transition at a certain frequency ω at a given time t_i; ω_0 is the central frequency of the pulse, and b is a parameter which describes the chirp of the pulse (see equation (11) below). Since the dependence of $\Delta t_s(\omega, t_i)$ on t_i during the interaction with the XUV pulse is negligible (see figure 1), we can further approximate

$$\langle \Delta t_s \rangle(\omega_0, b) \simeq \int \mathrm{d}\omega \Delta t_s(\omega) P_{\mathrm{avg}}(\omega, \omega_0, b), \qquad (8)$$

where $P_{\mathrm{avg}}(\omega, \omega_0, b)$ is the time averaged probability distribution. In our studies below we consider a linear chirped laser Gaussian pulse for which [48]

$$P_{\mathrm{avg}}(\omega, \omega_0, b) = N\sigma(\omega) \int_{-\infty}^{\infty} g(\omega, \omega_0, t, b) U(t) \mathrm{d}t, \qquad (9)$$

where $\sigma(\omega)$ is the photoionization cross section and

$$U(t) = \exp\left[\frac{-(16\ln 2)^2 t^2}{\tau^2}\right] \qquad (10)$$

is the temporal intensity distribution with $\tau/8\sqrt{\ln(2)}$ being the FWHM pulse duration of the Gaussian pulse. The instantaneous spectral distribution of the pulse

$$g(\omega, \omega_0, t, b) = \exp\left[-\frac{(\omega - \omega_0 - bt)^2}{\gamma^2/4\ln 2}\right], \qquad (11)$$

with $\gamma^2 = \Gamma^2 - \frac{\tau^2 b^2}{4}$, where $\Gamma = \frac{1}{\tau}[64(\ln 2)^2 + \tau^4 b^2]^{1/2}$ and N is a normalization factor.

Without chirp (i.e., $b = 0$) the instantaneous spectral distribution g is time-independent and P_{avg} is given by the photoionization cross section weighted by a Gaussian distribution centered about the central frequency ω_0 (solid lines

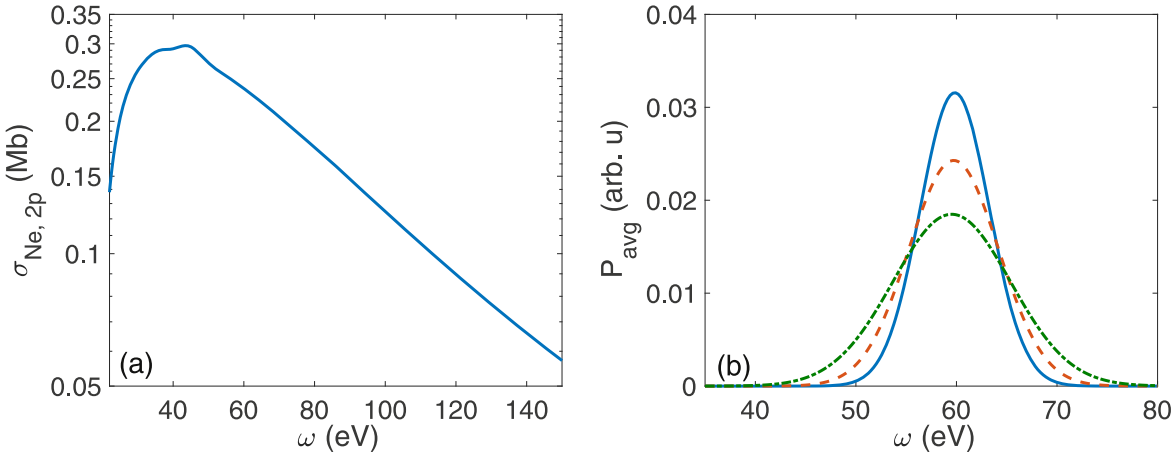

Figure 2. (a) Neon $2p$ photoionization cross section [44] and time averaged probability for (b) $\omega_0 = 60$ eV. The chirp is taken to be $b = 0$ eV fs^{-1} (solid line), $b = 15$ eV fs^{-1} (dashed line), and $b = 25$ eV fs^{-1} (dash-dotted) and $\tau = 450$ as.

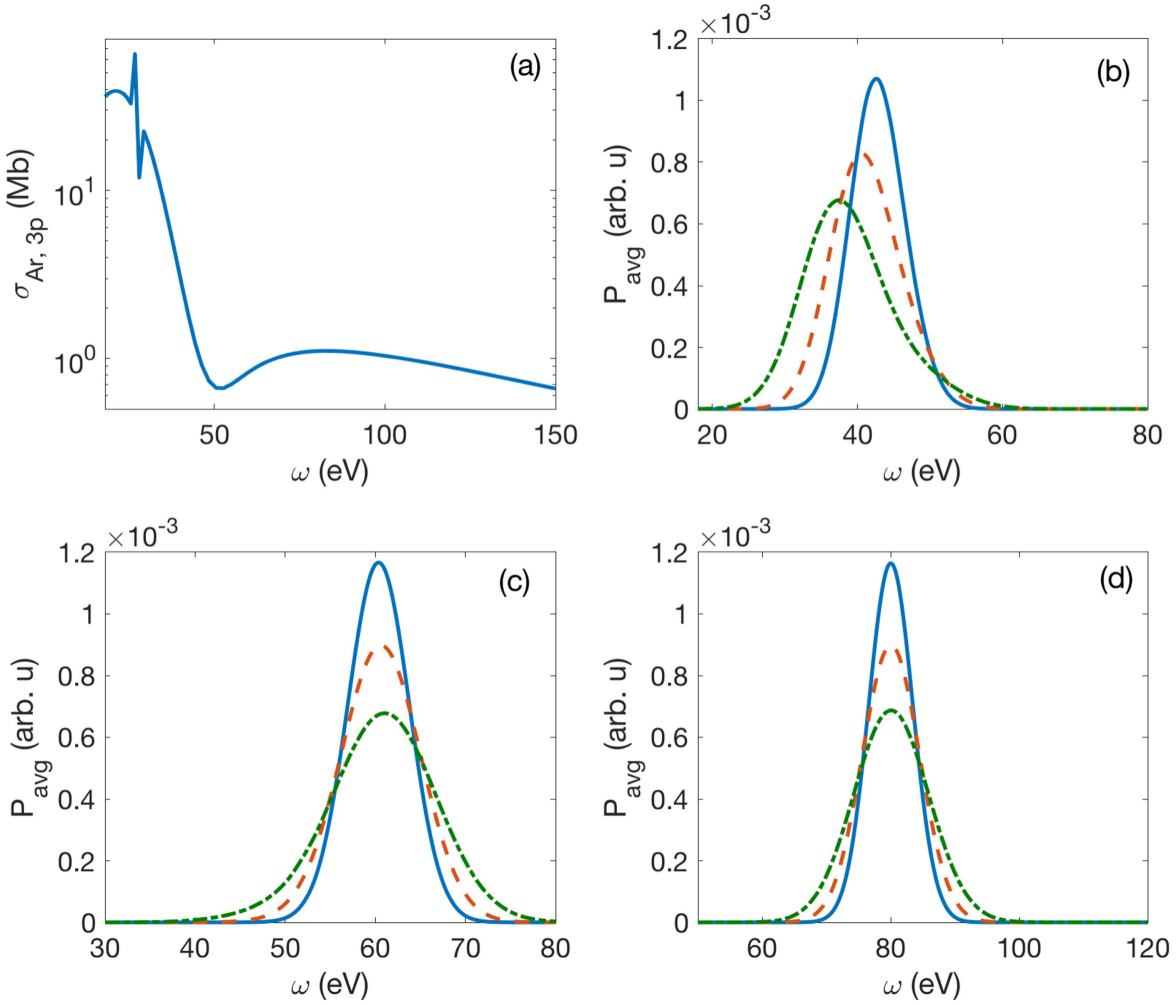

Figure 3. Same as figure 2 but for (a) argon $3p$ photoionization cross section [44]; time averaged probability for (b) $\omega_0 = 45$ eV, (c) $\omega_0 = 60$ eV, and (d) $\omega_0 = 80$ eV.

in figures 2(b) and 3(b)–(d)). For photoemission from Ne($2p$) (and the s-states, not shown) the cross section as a function XUV frequency is rather smooth (figure 2(a)). Consequently, a temporal chirp in the pulse causes a broader and less intense time averaged probability distribution while the position of the maximum in the distribution does not shift significantly independent of the central frequency ω_0 (figure 2(b)).

In contrast, in the case of Ar($3p$) (figure 3) the variation in the photoionization cross section near the frequency ω_C for the Cooper minimum leads to a shift of the maximum of P_{avg}.

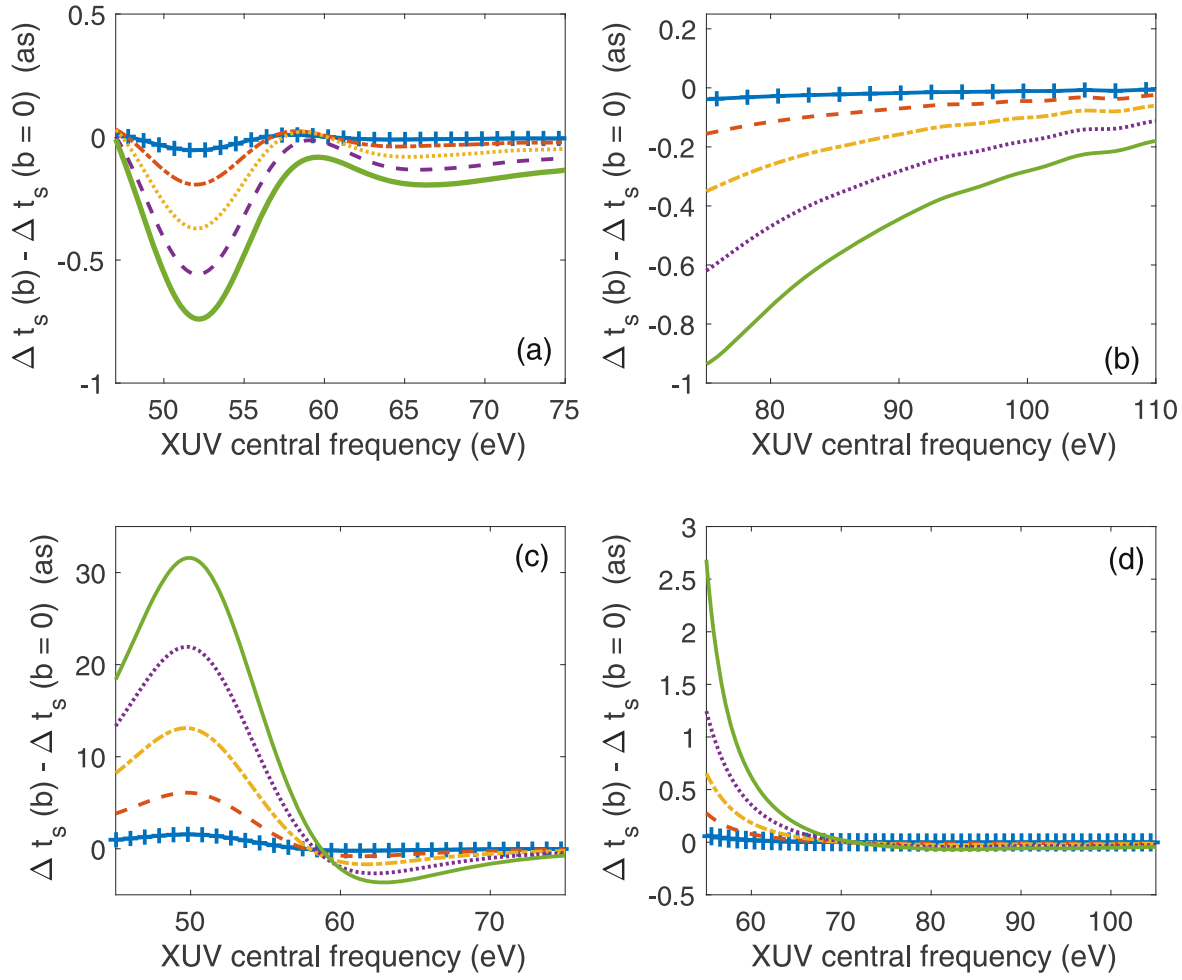

Figure 4. Change in streaking time delay for photoionization of Ne from $2p$ (a) and $2s$ (b) shells and of Ar from $3p$ (c) and $3s$ (d) in chirped laser pulses (with respect to an unchirped pulse) for $b = 5$ eV fs^{-1} (line with plus signs), $b = 10$ eV fs^{-1} (dashed), $b = 15$ eV fs^{-1} (dash-dot), $b = 20$ eV fs^{-1} (dotted), and $b = 25$ eV fs^{-1} (solid).

For central frequencies close below ω_C (figure 3(b)) the maximum appears at smaller frequencies than the central frequency ω_0 as the chirp of the pulse increases due the rapid decrease of the photoionization cross section. We note that the effect of Fano resonances below 30 eV on the present results cannot be further analyzed, since the resonances appear outside the frequency range of applicability of our model formula. At frequencies just above ω_C (figure 3(c)) we observe a small effect of the chirp due to the increase of the photoionization cross section. On the other hand, at frequencies significantly above ω_C the distribution of P_{avg} for Ar($3p$) changes in a similar way (figure 3(d)) as in the case of Ne($2p$) (or the s-states).

Before proceeding we note that for very large chirp

$$\lim_{b\to\infty} g(\omega, \omega_0, t_i, b) = \exp\left[\frac{-16\ln(2)}{3}\left(\frac{t}{\tau}\right)^2\right]. \qquad (12)$$

In this limit the time averaged probability distribution

$$\lim_{b\to\infty} P_{avg}(\omega, \omega_0, b) = N\sigma(\omega)\frac{\tau}{8}\sqrt{\frac{3\pi}{\ln 2}} \qquad (13)$$

and the expectation value of the streaking delay, equation (8), become independent of the central frequency ω_0. As test calculations have shown however, this limit is only reached approximately for unrealistic values of the chirp parameter b of a few hundred eV fs^{-1}.

3. Effect of Cooper minimum on streaking time delays in chirped pulses

As can be seen from equation (8) the effect of the attochirp on the streaking time delay arises from the convolution of the time averaged probability distribution $P_{avg}(\omega, \omega_0, b)$ and the variation of the delay with XUV photon energy ω. In figure 4 we present predictions for the difference between the streaking time delays obtained in a chirp pulse and an unchirped pulse for (a) Ne($2p$), (b) Ne($2s$) as well as for (c) Ar($3p$) and (d) Ar($3s$). In each case the difference increases with increase of the chirp parameter b and we observe variations as a function of XUV central frequency. For neon the effect is however even for the largest attochirp considered ($b = 25$ eV fs^{-1}) smaller than 1 as over the whole range of frequencies. This is due to the rather smooth variation of both

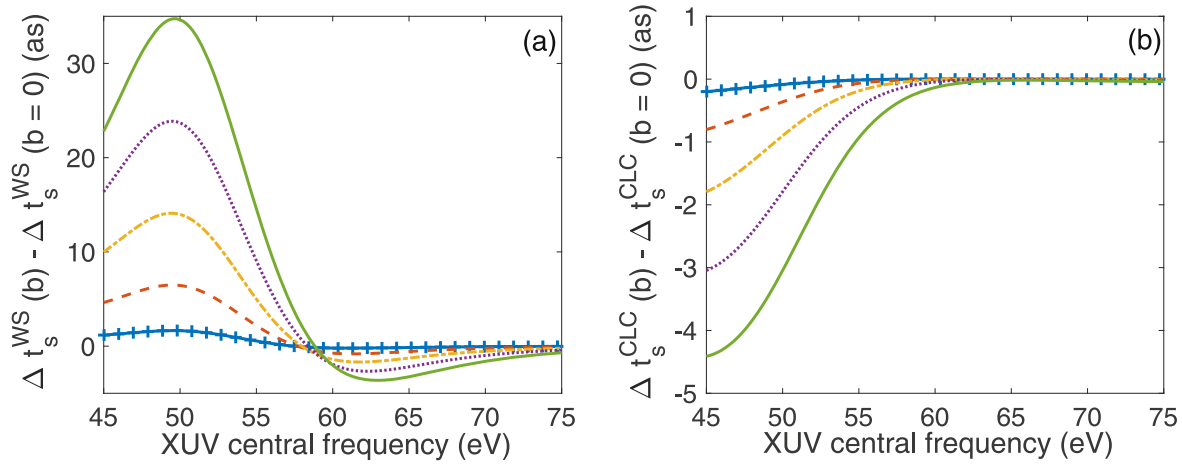

Figure 5. Comparison of the contributions from the (a) Wigner–Smith and (b) Coulomb-laser coupling terms for the change in the streaking delay for photoionization of Ar from 3*p* shell. Lines and symbols as in figure 4(c).

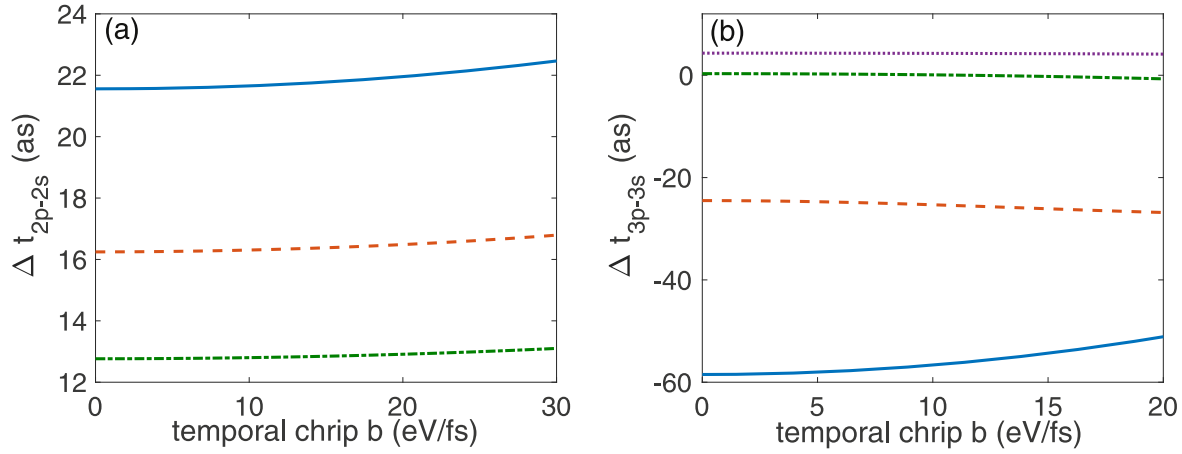

Figure 6. Difference of streaking time delays for photoemission (a) from 2*p* and 2*s* in neon at XUV central frequencies of 80 eV (solid), 90 eV (dashed), and 100 eV (dash-dotted) and (b) from 3*p* and 3*s* in argon at XUV central frequencies of 55 eV (solid), 60 eV (dashed), 70 eV (dash-dot), and 80 eV (dotted) as a function of the chirp parameter *b*.

the streaking time delays and the cross section for photoemission from the 2*p* and 2*s* subshells.

According to our results in the previous section, the influence of the chirp is much larger when the time delay and the cross section changes rapidly as a function of the laser frequency. Consequently, we observe significant changes of up to a couple of tens of attoseconds in the streaking time delay at small XUV frequencies for photoemission from the 3*p* shell in Ar, with a pronounced maximum centered at the Cooper minimum (figure 4(c)). This confirms previous expectations [43] that time delays and related effects are most pronounced near a Cooper minimum. The effect is larger at frequencies lower than ω_C than for $\omega > \omega_C$, in agreement with the results for P_{avg} in figure 3. In contrast, the changes in the streaking time delay for ionization of Ar(3*s*) as well as of Ar(3*p*) at frequencies significantly above the Cooper minimum are as small as for photoionization of Ne atom. We have further analyzed the impact of the two contributions to the streaking delay, namely Wigner–Smith term and Coulomb-laser coupling term, on the change observed in figure 4. The data in figure 5 show that the significant change for the Ar(3*p*) photoemission primarily results from the Wigner–Smith contribution (figure 5(a)). This is due to the fact that the Wigner–Smith term varies more rapidly than the Coulomb-laser coupling term around the Cooper minimum.

Finally, we present in figure 6 the relative time delays for photoemission from the different subshells in (a) Ne and (b) Ar as a function of the chirp parameter *b*. As expected, in the case of Ne it is found that an attochirp has a negligible effect on the intrinsic value of the relative streaking time delay. In particular, the relative time delay at 100 eV remains at about 12–13 as independent of the value of the chirp parameter. On the other hand, for Ar atom the relative delay can vary by several attoseconds for XUV laser frequencies near the Cooper minimum.

In conclusion, we have presented an analysis of the attochirp effect on the streaking time delay using a model formula which provides an efficient estimate of the delay. Our analysis relates to the intrinsic streaking time delay obtained in the superposition of both ionizing and streaking pulse, and provides complementary information to previous studies in which the effect of the attochirp on retrieved data has been

studied. The streaking time delay in chirped pulses is found to be significant for photoemission from Ar(3*p*) at frequencies near the Cooper minimum. On the other hand, if the photoionization cross section of the target subshell varies smoothly as a function of the XUV frequency, the effect of the attochirp on the streaking time delay appears to be negligible.

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