Angular resolved time delay in photoemission

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Abstract
We investigate theoretically the relative time delay of photoelectrons originating from different atomic subshells of noble gases. This quantity was measured via attosecond streaking and studied theoretically by Schultze et al. (2010 Science 328 1658) for neon. A substantial discrepancy was found between the measured and the calculated values of the relative time delay. Several theoretical studies were put forward to resolve this issue, e.g., by including correlation effects. In the present paper we explore a further aspect, namely the directional dependence of time delay. In contrast to neon, for argon target a strong angular dependence of the time delay is found near the Cooper minimum.

Keywords: time delay, angle-resolved, argon, neon

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

1. Introduction
The development of attosecond light sources paved the way for new exciting opportunities for studying time-resolved dynamical processes. Experiments on the attosecond electron dynamics in atomic, molecular and condensed matter systems confirmed the feasibility of this technique for a wide class of systems [1–7]. Thereby, an attosecond streaking metrology is employed for tracing the dynamics [8–11]: an extreme ultraviolet (XUV) pulse with a duration of a few hundred attoseconds plays the role of the pump and a phase-controlled few-cycle infrared (IR) pulse acts as the probe pulse. The coherent XUV pulse is characterized by a moderate intensity, short wavelength and a Keldysh parameter $\gamma \gg 1$. Valence electrons, which are emitted due to the action of the XUV field, are accelerated to different final momenta in the presence of the IR field. The asymptotic momenta depend on the vector potential of the IR field while the electron recedes from the residual ion allowing, thus, to access time information via momentum measurements.

The experiment in [11] reports on the time delay in the photoemission of electrons from the 2p subshell relative to those from the 2s subshell. A series of theoretical works were developed to quantitatively reproduce the measurements (for a review of the time delay concept we refer to [6] and references therein). For details of the time delay theory/interpretation we refer to the review articles [12, 13]. The experiment yielded a relative time delay of $21 \pm 5$ as. Within an independent electron model Schultze et al were able to calculate a delay of 6.37 as from matrix elements obtained by means of the state-specific expansion approach [11]. Ivanov and Kheifets reached a delay of 6.2 as by calculating scattering phases and dipole matrix elements in the Hartree–Fock (HF) approximation [14]. Accounting for first order correlation correction via the random phase approximation with exchange (RPAE) added 2.2 as. Another factor that may influence the time delay is the interaction between the photoelectron and the IR field. The measured delays can be generally divided in two parts. The first is the Wigner-like delay $\tau_W$ originating from the XUV photoionization process alone [15]. The second term stems from the interaction of the liberated photoelectron with the IR laser and the field of the residual ion and is referred to as Coulomb-laser coupling.
delay $\tau_{\text{CLC}}$ [16–18]:

$$
\tau = \tau_W + \tau_{\text{CLC}}.
$$

Zhang and Thumm [19] showed for the one dimensional hydrogen model that the resulting delay is independent of the IR pulse intensity. Nagele et al [16] confirmed this statement by propagating the full three dimensional Schrödinger equation numerically. They found that the Wigner-delay is accessible by streaking techniques only if distortions of both the initial-state entrance channel and the final-state exit channels (e.g. initial state polarization by IR or combined Coulomb-IR fields) are precisely accounted for. Recently, Moore et al [20] found a value of $10.2 \pm 1.3$ as for relative delay between the 2p and 2s subshells of argon for photon energies in the range of 34–40 eV. This measurement was repeated later by Guénot et al [22], whose results agree very well, except for that obtained at the highest energy of 40 eV. On the basis of the RPAE model the time delay in argon was calculated theoretically [18, 22, 23]. Recently, it was revisited by Dixit et al [24] who used the time-dependent density functional theory.

The angular dependence of the time delay has received relatively less attention. E.g., it was calculated for hydrogen atom showing a strong variation of the time delay around ±90° with respect to the laser polarization axis [25]. H$_2$ was studied in [26, 27].

In the experiment the time-of-flight detector collects all electrons within a certain solid angle. We introduce the angle $\theta_k$ between the asymptotic direction of the momentum of the photoelectron and the laser polarization axis. The latter is chosen to be parallel to the z-axis. The aim here is to investigate to which extent the angular dependence influences the resulting time delay with reference to its value by $\theta_k = 0°$. We analyze whether the calculations allow for some trends in the angle-dependent time delay. To address this point we study all possible propagation directions of the photoelectrons and calculate the corresponding angle-resolved time delays for neon and argon.

We concentrate on the angular dependence of the Wigner time delay $\tau_W$, which enters the time delay in equation (1) and the delay measured in attosecond streaking experiments. The corrections to the Wigner time delay through the Coulomb-laser coupling effect can be evaluated according to [28, 29]. Neon was chosen because of the experiment by Schultze et al [11] and the large number of theoretical results for the relative time delay between photoemissions from the 2p and 2s states; argon on the other hand is interesting due to the presence of a Cooper minimum in the spectra. Unless otherwise stated, atomic units are used throughout.

2. Theoretical model

The wave function representing a photoelectron wave packet is given by

$$
\Phi(r, t) = \int \! dk \; a(k, t) \psi_k^{(-1)}(r)e^{-i\varepsilon_k t},
$$

where $\psi_k^{(-1)}(r)$ stand for a set of continuum wave functions of the system and $a(k, t)$ are the corresponding projection coefficients. The projection coefficients corresponding to photoionization of a bound state indexed $i$ are in general angle-dependent and can be evaluated as [30]

$$
a_i(k) = -i \int_{-\infty}^{\infty} \! dt' \left\{ \psi_k^{(-1)}(r)H_{\text{int}} \right\} e^{-i\varepsilon_k t'},
$$

where $\varepsilon_i$ is the energy eigenvalue of the bound state $\mathcal{Y}_i$ and $\varepsilon_k = k^2/2$ is the kinetic energy. We employ the length gauge $H_{\text{int}} = E(t)$ for the interaction with the laser electric field $E(t)$. The field vanishes for $|t| \to \infty$ and we can express the coefficients as

$$
a_i(k) = -i D_i(k) \mathcal{E}(\varepsilon_k - \varepsilon_i),
$$

which is a product of the matrix element $D_i(k) = \langle \psi_k^{(-1)} | \mathcal{Y}_i \rangle$ and the Fourier transform (pulse spectral width) $\mathcal{E}(\varepsilon_k - \varepsilon_i)$ of the pulse electric field $E(t)$. The matrix element $D_i(k)$ describes the transition from the bound state $\mathcal{Y}_i$ to the continuum state $|\psi_k^{(-1)}\rangle$ with the kinetic energy $\varepsilon_k$. We express the wave function of the initial state as $\mathcal{Y}(r) = R_{\ell m}(r)Y_{\ell m}(\Omega_r)$, where $R_{\ell m}(r)$ and $Y_{\ell m}(\Omega_R)$ are the radial and spherical parts of the wave functions indexed by the usual quantum numbers $\ell_i$ and $m_i$. For the continuum states we expand in partial waves [17, 31, 32]

$$
\psi_k^{(-1)}(r) = \sqrt{(2\pi)^3} \sum_{l=0}^{+\infty} \sum_{m=-l}^{+l} i^{l+1} R_{l\ell}(r)e^{-i\delta_{j(k)}}
$$

$$
\times Y^*_{lm}(\Omega_k)Y_{lm}(\Omega_r).
$$

The radial wavefunctions are normalized as $\langle R_{l\ell}|R_{l'\ell'}\rangle = \delta(\ell_k - \ell_k')$. For the bound states we use a self-consistent HF method [33], while the continuum states are evaluated in the HF frozen core approximation [34]. Furthermore, we introduce the scattering phases which are given by $\delta_{j}(k) = \sigma_{\ell}(k) + \eta_{\ell}(k)$, where $\sigma_{\ell}(k) = \arg \left[ \Gamma \left( \ell + 1 - 1/2k \right) \right]$ is the Coulomb phase shift [35]. $\eta_{\ell}(k)$ is a phase correction originating from the short range deviation of the atomic potential from pure Coulomb potential [17].

Using the partial wave expansion and performing the angular integration we obtain for the matrix element $D_{ij}^{HF}(k)$
in the HF approximation:

\[ D_{1}^{\text{HF}}(k) = \sqrt{\frac{(2\pi)^3}{k}} \sum_{\ell,n,l=1}^{\infty} e^{i\Omega_{\text{HF}}(n)} Y_{\ell m}(\Omega_k) \times \left( \begin{array}{cc} \ell & 1 \\ -m & 0 \\ 1 & m \end{array} \right) d_{\ell,n,l}(k). \] (6)

The reduced dipole matrix elements \( d_{\ell,n,l}(k) = \langle kl|D|nl\rangle \) are given by

\[ d_{\ell,n,l}(k) = \sqrt{(2\ell + 1)(2\ell' + 1)} \left( \begin{array}{ccc} \ell & 1 & \ell' \\ 0 & 0 & 0 \end{array} \right) \times \int_{0}^{\infty} dr r^{3} R_{\ell l}(r) R_{\ell' l'}(r). \] (7)

In the RPAE model electron correlation effects are treated to the lowest order. The reduced dipole matrix element \( d_{\ell,n,l}(k) \) is replaced by a screened matrix element \( D_{\ell,n,l}(k) = \langle kl\parallel D\parallel nl\rangle \) which accounts for correlation between the various valence subshells. This matrix element is defined by the self-consistent equation \[31, 32\]

\[ D_{\ell,n,l}(k) = d_{\ell,n,l}(k) + \lim_{\epsilon \to 0} \sum_{n'l'} \sum_{n,l} \int \frac{dk'}{2\pi} \left[ \frac{D_{\ell,n',l'}(k')}{(n'\ell' + 1) |V|k' l' n,l} \right] \frac{\omega_{XUV} - \epsilon_{k} + \epsilon + i\epsilon}{|\omega_{XUV} + \epsilon_{k} - \epsilon|} + \frac{D_{n',l',l}(k')}{(n\ell + 1) |V|n\ell' n',l'} \right]. \] (8)

The indices i and j stand for valence orbitals and the sum/integral sign indicates summation over discrete excited states with energies \( \omega_{k} = \omega_{\ell l'} \) as well as integration over the continuum states with the energy \( \omega_{k} = k^2/2 \). The matrix element \( D_{\ell,n',l'}(k') \) in the third line of equation (8) means \( \langle n'\ell' |\parallel D\parallel k\ell' \rangle \). The reduced Coulomb matrix elements \( \langle n'\ell |\parallel V|\parallel kl\ell' \rangle \) and \( \langle k'\ell |\parallel V|\parallel nl\ell' \rangle \) belong to the time-forward and time-reversed scattering processes, which include both direct and exchange parts \[31, 32\]. Important for the phase information is that the integration in the time-forward term contains a pole with the consequence that the reduced matrix element is complex and acquires therefore an extra phase. Thus, \( D_{\ell,n,l}(k) \) can be expressed as \( D_{\ell,n,l}(k)e^{i\delta_{RPAE}(k)} \). The RPAE dipole matrix element has now the form

\[ D_{1}^{\text{RPAE}}(k) = \sqrt{\frac{(2\pi)^3}{k}} \sum_{\ell,n,l=1}^{\infty} e^{i\Omega_{RPAE}(k)} Y_{\ell m}(\Omega_k)\left| d_{\ell,n,l}(k) \right|. \] (9)

The reduced dipole matrix element includes both direct and exchange parts [31, 32]. The matrix element \( d_{\ell,n,l}(k) \) is therefore an extra phase. Thus, \( D_{1}^{\text{RPAE}}(k) \) provides a direct connection between the time delay and scattering phases \( \delta_{\ell}^{\text{HF}}(k) + \delta_{\ell}^{\text{RPAE}}(k) \), which strongly influence the dipole matrix element. By writing the spherical harmonics \( Y_{\ell m}(\Omega_k) \) as \( N_{\ell}^{m} p_{\ell}^{m}(\cos \theta_{k})e^{im\phi_{k}} \), the phase of the dipole matrix element is cast as

\[ \mu_{\ell}(\epsilon_{k}, \Omega_{k}) = \frac{\partial_{\epsilon_{k}}}{} \mu_{\ell}(\epsilon_{k}, \Omega_{k}). \] (10)

where \( \mu_{\ell}(\epsilon_{k}, \Omega_{k}) = \arg \left(D_{1}^{\text{RPAE}}(k) \right) \). The energy derivative of the photoionization amplitude of the respective subshell i, i.e.

\[ \tau_{\ell}^{\text{vm}}(\epsilon_{k}, \Omega_{k}) = \frac{\partial_{\epsilon_{k}}}{} \mu_{\ell}(\epsilon_{k}, \Omega_{k}). \] (11)

where we used

\[ S_{\ell \pm 1}(k, \theta_{k}) = \left( \begin{array}{ccc} \ell_{i} & 1 & \ell_{i} \\ -m & 0 & m \end{array} \right) D_{\ell_{i} \pm 1,n_{i}}(k) \times \left( N_{\ell_{i} \pm 1}^{m_{i}} p_{\ell_{i} \pm 1}^{m_{i}}(\cos \theta_{k}) \right). \] (12)

and

\[ \phi_{\ell_{i} \pm 1}(k, \theta_{k}) = \delta_{\ell_{i} \pm 1}^{\text{HF}}(k) + \delta_{\ell_{i} \pm 1}^{\text{RPAE}}(k) \] (13)

Equation (11) in this form is only suitable for the photoemission from a np state with \( \ell_{i} = 1 \) and \( m_{i} = 0 \). One can see immediately that in this case the phase of the dipole matrix element \( \mu_{\ell_{i}}(\epsilon_{k}, \Omega_{k}) \) is only dependent on the angle \( \theta_{k} \) but not on \( \phi_{k} \) (since \( m_{i} \) is zero). This means that the time delay \( \tau_{W} \) as the energy derivative has an angular dependence which is strongly influenced by the ratio \( S_{\ell_{i} \pm 1}/S_{\ell_{i} \pm 1} \) and the scattering phases \( \delta_{\ell_{i} \pm 1}^{\text{HF}}(k) + \delta_{\ell_{i} \pm 1}^{\text{RPAE}}(k) \).

In the cases of photoemission from a ns state \( (\ell_{i} = 0 \) and \( m_{i} = 0 \)) and photoemission from a np state with \( m_{i} = 1 \) equation (11) reduces to \( \mu_{\ell_{i}}(\epsilon_{k}, \Omega_{k}) = \phi_{\ell_{i} \pm 1}(\epsilon_{k}, \phi_{k}) \), because \( S_{\ell_{i} \pm 1}(\epsilon_{k}, \theta_{k}) \) vanishes. In these special cases the time delay as the energy derivative of \( \mu_{\ell_{i}}(\epsilon_{k}, \Omega_{k}) \) is only characterized by the properties of the scattering phase in equation (13). Therefore, the HF and RPAE theory predicts no angular dependence of these state-specific time delays. In all cases the phase factor \( m_{i} \phi_{k} \) which depends on the polar angle \( \phi_{k} \) has no influence on the time delay. Therefore, we can state that \( \tau_{W}^{\text{vm}}(\epsilon_{k}, \Omega_{k}) \equiv \tau_{W}^{\text{vm}}(\epsilon_{k}, \theta_{k}) \).

The time delays defined as the energy derivative of the phase of the photoionization amplitude equation (10) are in general energy-dependent. To find the characteristic time delay corresponding to the photon frequency \( \hbar \omega_{XUV} \) we have to average over the ionization probability \( w_{i}(\epsilon_{k}, \theta) = |a_{i}(k)|^{2} \), which has also an angular dependence due to the angular dependence of \( a_{i}(k) \). These probabilities are peaked at the center of energy (COE) \( \epsilon_{\text{COE}} = \hbar \omega_{XUV} + \epsilon_{i} \) but are not sharp because of the spectral width of the pulse due to the short duration. Therefore we can define the time delays corresponding to photoionization from the ns and np
subshells by averaging over the probabilities:

\[
\tau_W^\phi(\theta_k) = \frac{\int d\mathbf{r}_1 \ |W_{0,0}^f(\mathbf{r}_1, \theta_k)\rangle \langle W_{0,0}^f|}{\int d\mathbf{r}_2 \ |W_{0,0}^p(\mathbf{r}_2, \theta_k)\rangle \langle W_{0,0}^p|}.
\]

\[
\tau_W^s(\theta_k) = \frac{\int d\mathbf{r}_1 \ \sum_{m=-1}^1 W_{1,m}^f(\mathbf{r}_1, \theta_k) \langle W_{1,m}^f|}{\int d\mathbf{r}_2 \ \sum_{m=-1}^1 W_{1,m}^p(\mathbf{r}_2, \theta_k) \langle W_{1,m}^p|}.
\]

(14)

The relative time delay is defined as \(\tau_W^{\phi - s} \equiv \tau_W^\phi - \tau_W^s\).

3. Angular dependence of the time delay of neon

The considered XUV field is modeled as

\[
E_{\text{XUV}}(t) = E_0 \cos\left(\frac{\pi t}{2T_{\text{XUV}}}\right) \cos(\alpha \omega_{\text{XUV}} t)
\]

for times \(t\) within the interval \([-T_{\text{XUV}}, T_{\text{XUV}}]\) and vanishing otherwise. In view of the experiment \([11]\) the parameter \(T_{\text{XUV}}\) for Ne is chosen such that the pulse full width at half maximum (FWHM) is 182 as. The amplitude of the electric field of XUV field is 0.12 a.u. which corresponds to a peak intensity of \(5 \times 10^{14}\) W cm\(^{-2}\).

In panel (a) of figure 1 we show explicitly the time delays corresponding to the 2s and the three possible 2p initial states of neon in dependence on the angle \(\theta_k\) for a photon energy of 106 eV, which was used in the experiment of Schultze \textit{et al.} \([11]\). Although the RPAE is a more advanced theory because it treats intershell correlation we compare its predictions of the angular dependence with a full three dimensional SAE (single active electron) propagation. Previous studies showed that in Ne the intershell correlation has no significant impact on the 2p photoionization process \([14, 23]\), which is according to equation (11) the origin of the angular dependence of the relative time delay \(\tau_W^{\phi - 2s}\). Thus we can compare both results qualitatively. The numerical simulation is realized with the matrix iteration method \([37]\). As an atomic potential for Ne we use the optimized effective single-particle potential as in \([38]\). The time dependent wave function is expanded in spherical harmonics, i.e.

\[
\Psi(\mathbf{r}, t) = \sum_{L=0}^L \sum_{m=-L}^L R_l(r) Y_{lm}(\Omega_t).
\]

For \(t \to -\infty\) we define that \(\Psi(\mathbf{r}, t) = \Psi(\mathbf{r})\). Thus, every initial state is propagated from \(t = -T_{\text{XUV}}\) to \(T_{\text{XUV}}\) in the presence of the laser field. After the photoelectron wave packet is fully formed, the solution \(\Psi(\mathbf{r}, t > T_{\text{XUV}})\) is projected on a set of field-free continuum wave functions \(\phi_k^\phi(r)\) and we obtain the projection coefficients \(a_i(k)\) (compare with equation (3)) corresponding to the bound state \(i\), which can be further analyzed to extract the time delay information.

We remark that the results presented in this work reflect the angular dependence of the Wigner time delay \(\tau_W\) only. Information on angular dependencies associated the Coulomb-laser coupling time delay \(\tau_{\text{CLC}}\) that originates from the two-photon matrix element \([17]\) and includes effects of the interaction of the photoelectron with the IR-field and the residual ion are beyond the scope of the present work. So, we cannot rule out any additional angular dependencies related to CLC effects.

The RPAE results indicate a strong angular dependence of the time delay which corresponds to the 2p photoionization channel with \(m_i = 0\). However the time delays corresponding to the 2s and 2p with \(m_i = \pm 1\) channels show no angular...
The time delay of the photoionization process corresponding to the 2p initial state with $\{\ell = 1, m = 0\}$ shows substantial variations. The pronounced sharp structures around $\pm 57^\circ$ emerge due to the vanishing contribution of the typically dominating transition to the $\ell = 2$ partial wave [39], that means the corresponding phase $\delta_{\ell=2}(k) = \delta^{HF}_{\ell=2}(k) + \delta^{RPAE}_{\ell=2}(k)$ mainly determines the time delay. From equation (11) follows that the phase $\mu_\ell(\varepsilon_k, \theta_k)$ is then mainly determined by the scattering phase $\delta_{\ell=2}(k) = \delta^{HF}_{\ell=2}(k) + \delta^{RPAE}_{\ell=2}(k)$, whose energy derivative is negative. The RPAE calculations and full numerical simulation deliver comparable results for the angular dependence of the 2p time delay corresponding to $\{\ell = 1, m = 0\}$.

While the effect of the RPAE on the delays related to photoionization from the 2p subshell is subsidiary [23], we find that the energy derivative of the additional phase $\delta_{\ell=2}^{RPAE}(k)$ has a significant influence on the resulting delay $\tau^{RPAE}_{\ell=2}$. This leads to the observed discrepancy between the RPAE results and the full numerical simulation regarding the relative time delay including the contributions of all four possible initial states. The panel (b) of figure 1 shows the angular dependence of the relative delay. The results illustrate that the strong effect of the angular dependence corresponding to the 2p initial state with $\{\ell = 1, m = 0\}$ is nearly compensated by the constant time delays corresponding to $\{\ell = 1, m = \pm 1\}$. This is underpinned by the fact that the sum in the denominator of equation (14) can be expressed by

$$\sum_{m=-1}^{1} \sum_{\ell,m} W_{\ell,m}(\varepsilon_k, \theta_k) \propto 1 + \beta_{2p}(\varepsilon_k) P_2(\cos \theta_k),$$

which describes the angular dependence of the photoionization process. From the inset in the panel (b) we know that $\beta_{2p}(\varepsilon_k)$ is approximately 1.5 around $\hbar \omega_{\text{XUV}} = 106 \text{eV}$, meaning that

$$\frac{1}{1 + \beta_{2p}(\varepsilon_k) P_2(\cos \theta_k)}$$

has two maxima at $\pm 90^\circ$. This explains why the relative time delay grows slowly when approaching $\theta_k = \pm 90^\circ$. Thus, the numerator of equation (14) which is determined by

$$\sum_{m=-1}^{1} \sum_{\ell,m} W_{\ell,m}(\varepsilon_k, \theta_k) \tau^{\text{rel}}_{\ell,m}(\varepsilon_k, \theta_k)$$

shows a very weak angular dependence. The value of the relative time delay $\tau^{2p-2s}_{\ell=2}(\theta_k)$ in the forward direction $\theta_k = 0^\circ$ is 4.82 as, which is comparable to the 4.5 as from SAE simulations performed by Schulzke et al [11]. The value for the RPAE calculations is 8.19 as, which is in good agreement with the results of Kheifets et al [14, 23] and serves as a good check for our calculations.

The panel (c) of figure 1 shows the relative time delay of neon as a function of the XUV photon energy for different asymptotic directions $\theta_k$. While the effect of the RPAE on the delays related to photoionization from the 2p subshell is subsidiary, we find that the energy derivative of the additional phase $\delta_{\ell=2}^{RPAE}(k)$ has a significant influence on the resulting delay $\tau^{RPAE}_{\ell=2}$. This leads to the observed discrepancy between the RPAE results and the full numerical simulation regarding the relative time delay including the contributions of all four possible initial states. The panel (b) of figure 1 shows the angular dependence of the relative delay. The results illustrate that the strong effect of the angular dependence corresponding to the 2p initial state with $\{\ell = 1, m = 0\}$ is nearly compensated by the constant time delays corresponding to $\{\ell = 1, m = \pm 1\}$. This is underpinned by the fact that the sum in the denominator of equation (14) can be expressed by

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The panel (c) of figure 1 shows the relative time delay of neon as a function of the XUV photon energy for different asymptotic directions $\theta_k$ up to $45^\circ$, exhibiting a very smooth angular dependence. An averaging of the relative time delay over the interval $\theta_k \in [-45^\circ, 45^\circ]$ at a photon energy of $106 \text{eV}$ leads to a relative time delay of $\tau^{2p-2s}_{\ell=2} = 8.41$ as while a smaller acceptance angle of $\theta_{\text{max}} = 20^\circ$ results in a time delay of $\tau^{2p-2s}_{\ell=2} = 8.24$ as.

4. Angular dependence of the time delay of argon

In case of argon we used the same shape and field amplitude of the electromagnetic perturbations. To reach improved
results we used experimental bound state energies instead of the HF eigenvalues [23]. Thus, the energy difference between the 3s and 3p states is 13.48 eV [40]. To avoid accidental photoionization of both initial states we had to use a pulse with a longer duration in comparison to neon, which means that the energy spectrum is narrower. In case of argon a FHWM of 300 as is sufficient.

The panel (a) of figure 2 shows the angular independent time delay corresponding to photoionization from the 3s initial state. Electronic correlations as implemented in RPAE have marked effects that show up in the time delay. In particular the presence of the well-known Cooper minimum (in contrast to Ne) results in a distinctive feature [23]. This Cooper minimum does not be reproduced correctly by a SAE calculation. Thus, the full numerical simulation with a SAE model potential is not performed.

In panel (b) of figure 2 the time delay of the 3p photoionization is depicted, which is calculated according to equation (14). The expected Cooper minimum around the photon energy of 50 eV [41, 42] is correctly reproduced. Substantial variations in the time delay with the photon energy as well as a distinctive angular dependence are observed. We find a less pronounced peak for the 3p subshell in comparison to the 3s subshell (transitions to the final states with \( \ell = 0 \) and \( \ell = 2 \) are possible in the former case). This can be explained by the interference between the normally weak \( \ell = 1 \rightarrow \ell = 0 \) transition, which becomes stronger near the Cooper minimum, and the normally dominant \( \ell = 1 \rightarrow \ell = 2 \) transition [39]. Thus, the resulting time delay does not fall below \(-100\) as. The discrepancies between the various energy-dependent curves for different asymptotic directions \( \theta_k \) vanish for larger photon energies.

In the region around the Cooper minimum the time delays corresponding to photoionization from 3p subshell are negligible despite the positive energy derivative of the HF scattering phase of the dominant \( \ell = 1 \rightarrow \ell = 2 \) transition. This is due to the additional phase which is provided by the RPAE correction and contributes with a negative \( \pi \)-jump at the Cooper minimum producing a local and very distinctive negative time delay [23]. For larger angles, the peaks around the Cooper minimum become less prominent. This characteristic can be explained by the decreasing values of the associated Legendre polynomial \( P^0_3(\cos(\theta_k)) \) (see equation (11)) with increasing \( \theta_k \), which weakens the transition \( \ell = 1 \rightarrow \ell = 2 \) further and increases the influence of the scattering phases \( \delta_\ell^{HF} = 0 + \delta_\ell^{RPAE} \). The inset in figure 2(b) shows the time delay of the 3p photoionization process angle-resolved for three different photon energies. In case of neon the time delay was nearly independent on the angle in the range between \(-45^\circ \) and \( 45^\circ \) (see figure 1(b)), while in the case of argon we find a strong angular variation at energies in the vicinity of the Cooper minimum (50 and 60 eV). For higher energies the situation is similar to the case of neon.

In figure 2(c) the full relative delay \( \delta_{\ell = 3s}^{i - 3p} \) in dependence on the photon energy for different asymptotic directions is depicted. The graph reveals a large peak around 42 eV, which originates from the prominent peak of the 3s delay at the corresponding Cooper minimum. Characteristic for this feature is a very weak angular dependence. The larger variations with angle, originating from the 3p contribution, show up primary at larger photon energies. The results for the forward direction, i.e. \( \theta_k = 0^\circ \), are in good agreement with the calculations of Dahlström et al [28] and Kheifets [23]. We marked the photon energies (SB 22–SB 26) corresponding to the results of the interferometric experiments [18, 22] and see that for these energies there is no significant angular dependence of the relative delay, which could influence the result of the measurement.

In contrast, at the photon energy of 51 eV the value of the relative time delay in forward direction amounts to 105 as, while an acceptance angle of \( 20^\circ \) of the detector results in an averaged relative delay of 92 as. A larger acceptance angle of \( 45^\circ \) leads to a relative time delay, averaged over all possible photoemission directions, of 68 as.

5. Conclusion

In the present work, we studied the angular dependence of the photoemission time delay from the valence shells of neon and argon. Results were obtained within the RPAE to include intershell correlations. We found that the existence of a Cooper minimum has a strong impact on the angular dependence of time delay. From an experimental point of view the angular dependence has almost no relevance in the case of neon because the effect is nearly not existent around the forward direction. In the case of argon and for photon energies around the Cooper minimum corresponding to the 3p photoionization process the effect of the angular dependence of the Wigner time delay is evident and can have a sizable influence on the measured delay in this region.

Note added. Very recently we became aware of the publication of an independent work by J M Dahlström and E Lindroth [43] who calculated, among other quantities, the angular resolved time delay for the 3p photoionization in argon with the aid of the perturbation theory based on two-photon matrix elements. The results in directions of \( 0^\circ \) and \( 45^\circ \) are comparable to ours which reflects the minor influence of the IF field on the angular characteristics of the time delay.

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