Angular distributions and correlations in sequential two-photon atomic double ionization

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Abstract. Theory of the sequential two-photon double ionization of atoms in the XUV wavelength region is discussed in the context of recent experiments with the free-electron laser. Angular distributions and angular correlations of photoelectrons are analyzed within the statistical tensor formalism and examples are given for sequential two-photon double ionization of noble gases.

1. Introduction
The high intensity of the photon beams achieved in experiments with Free electron LASer in Hamburg (FLASH) has opened the possibility for studying nonlinear processes in the XUV range. Among them, the sequential two-photon double ionization (TPDI) is especially important: it is one of the simplest possible non-linear processes, associated with the consecutive absorption of two photons and the emission of two electrons. In contrast to the direct TPDI, the sequential TPDI proceeds through a particular intermediate ionic state (figure 1) and the individual energy of each of the emitted electrons is fixed by energy conservation. If the photon energy is larger than the ionization potential of the singly charged ion, the sequential TPDI is the dominant double ionization process at moderate XUV radiation intensities [1, 2, 3]. Thus, neglecting interference between the amplitudes of the two mechanisms is a reliable approximation at least for the photon energies, corresponding to the sequential process (marked by two arrows in the right panel of figure 1).

Already in the first experiments at the FLASH facility, the angular distributions of photoions and of photoelectrons have been measured [4, 5, 6, 7, 8] and the angular correlation for the two emitted photoelectrons has been reported [9]. Recently, we have developed a general theoretical framework for description of the angular distributions and angular correlations of the emitted photoelectrons and applied it to noble gas atoms [10, 11, 12]. The theory is based on the two-step mechanism of the TPDI with possible quantum evolution between the individual steps. It uses the formalism of density matrix and statistical tensors which is very well suited for the treatment of such processes (for example [13]). An alternative approach has been applied to the sequential TPDI by Kheifets [14, 15].
In this report we discuss selected topics of our recent theoretical studies of the sequential TPDI and present some new results, both analytical and numerical, for noble gases. Since the basic formalism is well documented in [10], we present only those expressions which are necessary for discussion.

![Figure 1](image)

**Figure 1.** Scheme of direct and sequential two-photon double ionization of a noble gas atom. The total kinetic energy of the two outgoing electrons ($e_1 + e_2$) shared between the electrons for the direct process (left panel) equals to the sum of discrete energies of the electrons for the sequential process (right panel).

2. Ionization by first and second photons as independent events
In order to discuss the angular distributions and angular correlations of photoelectrons in the sequential TPDI of noble gas atoms, we start with the single ionization from their outer $np^6$ shell,

$$\gamma_1 + A(np^6^1S_0) \rightarrow A^+(np^5^2P_{J_i}) + e_1^-(s_{1/2}, d_{3/2}, d_{5/2}),$$

and ionization of their single charged ions from the $np^5$ shell,

$$\gamma_2 + A^+(np^5^2P_{J_i}) \rightarrow A^{++}(np^4^3P_{0,1,2}, ^1D_2, ^1S_0) + e_2^-(s_{1/2}, d_{3/2}, d_{5/2}),$$

as independent events. The reaction (1) has been extensively studied both experimentally, with the use of synchrotron radiation, and theoretically (for a list of references see e.g. the reviews [16, 17]). We assume linearly polarized beam of radiation, spin insensitive electron detector, and use the dipole approximation to describe atomic interaction with the radiation field. The angular distribution of photoelectrons in the process (1) is of the form

$$\frac{d\sigma_1}{d\Omega_1}(J_i) = \frac{\sigma_1(J_i)}{4\pi} \left( 1 + \beta^{(1)} P_2(\cos \vartheta_1) \right),$$

where $\sigma_1(J_i)$ is the angle-integrated partial cross section for transition to the ionic state $J_i$, $\vartheta_1$ is the angle of electron emission with respect to the $z$ axis which is chosen along the polarization vector of photons, $P_2(x)$ is the second Legendre polynomial and $\beta^{(1)}$ is the asymmetry parameter. Similarly, the angular distribution of photoelectrons in ionization of randomly oriented ions to the final state $J_f$ of the double charged ion is

$$\frac{d\sigma_2}{d\Omega_2}(J_f) = \frac{\sigma_2(J_f)}{4\pi} \left( 1 + \beta^{(2)} P_2(\cos \vartheta_2) \right),$$
with obvious change of notations. Here and below the upper indices (1) and (2) refer to the ionization of the neutral atom and the ion, respectively. The parameters $\beta^{(1)}$ and $\beta^{(2)}$ are expressed in terms of the partial dipole amplitudes of photoionization processes (1) and (2), respectively. Details of the models which we have used to describe the photoionization of the outer $np^5$ shells of Ne, Ar, Kr can be found in [10, 11]. Here we present the results of calculations based on the multiconfiguration Hartree-Fock (MCHF) approximation [18], accounting for mixing of configurations in the discrete spectra of atoms and ions. Term-dependent frozen-core Hartree-Fock continuum photoelectron wave functions were employed to calculate the LS-coupled multielectron dipole ionization amplitudes. The MCHF approach does not account for the relativistic effects in the radial electron functions. In papers [10, 11] we have presented also the results of multiconfiguration Dirac-Fock (MCDF) calculations. In general they are close to those demonstrated here.

![Graph showing angular asymmetry parameters $\beta^{(1)}$ and $\beta^{(2)}$.](image)

Figure 2. Angular asymmetry parameters $\beta^{(1)}$ (upper panel) and $\beta^{(2)}$ (lower panel). The parameter $\beta^{(2)}$ is displayed for randomly oriented ions in the $np^5 2P_J$ states and the final $^3P$ state of the doubly charged ion. Ionization thresholds of the single charged ions are indicated. Experimental data for $\beta^{(1)}$: Ne [19] (crosses), Ar [20, 21] (squares), Kr [22] (triangles).

Figure 2 shows our results for $\beta^{(1)}$ and $\beta^{(2)}$ obtained in the MCHF approximation together with experimental data for $\beta^{(1)}$. The region of the photon energies is shown above ionization threshold of the single charged ion, where the sequential TPDI dominates. For the considered targets, the calculated $\beta^{(1)}$ is in good agreement with experiments. The above results demonstrate the quality of the wave functions used in the description of the first step of the sequential TPDI. No experimental data exist for the photoelectron angular distributions from ions. Note that the calculated asymmetry parameters $\beta^{(1)}$ and $\beta^{(2)}$ show similar behavior as functions of the photon energy.

If the emission of the two electrons in the sequential TPDI were independent events, the angular correlation pattern would be a product of the two angular distributions (3) and (4):

$$W(\theta_1, \varphi_1; \theta_2, \varphi_2) = \frac{d\sigma_1}{d\Omega_1(J_i)} \frac{d\sigma_2}{d\Omega_2(J_f)}$$

(5)
Our values of the alignment are in good agreement with previous calculations [23]. In the np far the alignment of the noble gas photoions in the ground ionic state, which is polarized after the first step ionization. In the simplest case, when the two steps of the sequential TPDI are connected at least through the intermediate photoelectron existence of real dynamical correlations in the emission events.

Any deviation of calculated or measured angular correlations from the IE pattern indicates the existence of real dynamical correlations in TPDI. The IE pattern depends only on the two polar angles of the photoelectrons with respect to the linear polarization vector of the radiation, $\theta_1$ and $\theta_2$, and can be calculated using the asymmetry parameters $\beta^{(1)}$ and $\beta^{(2)}$. Typical IE pattern for the sequential TPDI of the noble gas atoms near the threshold, where the both asymmetry parameters take positive and close values, is presented in figure 3. Any deviation of calculated or measured angular correlations from the IE pattern indicates the existence of real dynamical correlations in the emission events.

3. Alignment of the photoion and angular distribution of the second-step photoelectron

In fact the two steps of the sequential TPDI are connected at least through the intermediate ionic state, which is polarized after the first step ionization. In the simplest case, when the first electron is not detected, the residual photoion in the np$^5 2P_3/2$ state is characterized by the alignment, $A_{20}(2P_{3/2}) = (N_{3/2} - N_{1/2})/(N_{3/2} + N_{1/2})$, where $N_{Mi}$ is the population of magnetic sublevel of the $2P_{3/2}$ state with the projection $M_i$ on the polarization direction of the photon. (The alignment is zero for $2P_{1/2}$ states). The calculated alignment for the $2P_{3/2}$ state (figure 4) is negative, pointing to a preferential population of the $M_i = \pm 1/2$ substates. So far the alignment of the noble gas photoions in the ground np$^5 2P$ states remains unmeasured. Our values of the alignment are in good agreement with previous calculations [23]. In the nonrelativistic approximation, $A_{20}(2P_{3/2})$ reaches the value of $-1$ in the Cooper minimum, associated with zero amplitude for ionization into the Ed channel.

The angular distribution of the second photoelectron depends on the alignment $A_{20}(2P_{3/2})$ and instead of (4) it takes the form

$$W(\vartheta_2) = \sigma_1(J_i) \frac{\sigma_2(J_f)}{4\pi} \left[ 1 + \beta^{(2)} P_2(\cos \vartheta_2) + A_{20}(J_i)[a_0 + a_2 P_2(\cos \vartheta_2) + a_4 P_4(\cos \vartheta_2)] \right]$$

![Figure 3. IE angular correlation patterns for sequential TPDI of Kr (upper panels) at photon energy of 40 eV ($\beta^{(1)}=\beta^{(2)}=1.8$) and Ne (lower panels) at 44 eV ($\beta^{(1)}=0.9$, $\beta^{(2)}=0.5$). Although the values of $\beta^{(1)}$ and $\beta^{(2)}$ were taken for the states A$^+$ np$^5 2P_{3/2}$ and A$^{++}$ np$^4 3P_2$, the results are essentially similar for other state components. Lighter and darker greys indicate higher and lower values, respectively.](image-url)
\[
W_0 = \frac{1}{4\pi} \left[ 1 + \beta_2^{(2)} P_2(\cos \theta_2) + \beta_4^{(2)} P_4(\cos \theta_2) \right],
\]

where \(a_0, a_2, a_4\) are dynamical parameters, expressed in terms of the ratios of the partial amplitudes of the second step ionization [10]. First two terms in parenthesis in equation (6) correspond to photoionization from randomly oriented ion. The value of \(\beta_2^{(2)}\) differs from \(\beta^{(2)}\) in (4) due to the term proportional to the alignment. The values of \(\beta_4^{(2)}\) generally are small, after averaging over the final state \(np^4 3P, 1D, 1S\) multiplets and intermediate \(np^5 2P_{1/2,3/2}\) fine structure states [14]. When the states are resolved, the \(\beta_4^{(2)}\) coefficients can reach absolute values of 0.2-0.3, as demonstrated in figure 5. First data on noticeable values of \(\beta_4^{(2)}\) in the \(np^4\) term-resolved spectra have been reported [5, 24]. Contrary to what one might expect, the contribution of the term with \(P_4(\cos \theta_2)\) in the angular distribution (6) is maximal far from the region of maximal values of the alignment \(A_{20}\) [12]. The reason is that the partial ionization cross section in the \(d\) wave vanishes \((a_4 \sim 0, \text{see equation (6)})\) in the region of the Cooper minimum, where the alignment \(A_{20}\) takes maximum absolute values.

**Figure 4.** Alignment of the photoion \(np^5 2P_{3/2}\) states. Notations are the same as in figure 2.

**Figure 5.** Asymmetry coefficients \(\beta_4^{(2)}\) in the angular distribution of the second-step electron in sequential TDPI of the Kr atom via the intermediate \(Kr^+ 4p^5 2P_{3/2}\) state into different final states \(Kr^{++} 4p^4\), marked in the figure.

### 4. Polarization of the photoion and electron angular correlations

When the first-step photoelectron is detected, the axial symmetry of the first photoionization process breaks down. Instead, the plane of symmetry is defined by the direction of the photoemission and the direction of the photon polarization. For the plane of symmetry, the ionic
state $A^+(2P_{3/2})$ is characterized not only by the alignment, but by larger numbers of polarization parameters (statistical tensors). Thus, the statistical tensors of the ranks 1, 2, and 3, including those with nonzero projections, are generally nonvanishing for the $2P_{3/2}$ state. Only some of them are independent. Furthermore, the state $2P_{1/2}$ would possess orientation described by the statistical tensor of the first rank. The situation is quite similar to the statistical tensors (state multipoles) of atomic states excited by particle impact, when the spin unresolved scattering process defines the plane of symmetry spanned by the directions of the incident beam and scattering particle [25, 26]. In the laboratory frame with z axis along the photon polarization and arbitrary fixed x axis, the statistical tensors of the photoion can be derived as [26, 10]

$$
\rho_{k_1q_1}(J_1, J'_1; \theta_1, \varphi_1) = \pi \alpha \omega \sum_{k_1k_1'} B(k_1, k_1, k_1') \rho^7_{k_10}(k_1q_1, k_10 | k_1q_1) \sqrt{\frac{4\pi}{2k_1 + 1}} Y^*_{k_1q_1} (\theta_1, \varphi_1), \quad (7)
$$

where $\alpha$ is the fine structure constant, $\omega$ is the photon energy, $(j_1m_1, j_2m_2 | j m)$ is the Clebsch-Gordan coefficient, $Y_{kq}(\theta, \varphi)$ denotes the spherical harmonic, $\rho^7_{k_10}$ is the statistical tensor of the rank $k_1$ of the photon which in our case possesses only two nonzero components: $\rho^7_{k_10} = 1/\sqrt{3}$ and $\rho^7_{20} = -\sqrt{2/3}$. Equation (7) is slightly more general than equation (5) of [10], because of arbitrary direction of the x axis. The generalized anisotropy coefficients $B(k_1, k_1, k_1')$ are expressed in terms of dipole matrix elements of single photoionization of atoms (see equation (6) in [10] in which one should set $J_0 = 0$ for noble gas atoms). The polarization parameters of the photoion (7) depend on the angle of the photoemission $\theta_1, \varphi_1$ through the spherical harmonics $Y_{k_1q_1}(\theta_1, \varphi_1)$.

The polarized ionic state, described by the statistical tensors (7), is the initial state for the second step ionization. Applying a general theory of photoionization from polarized atoms and standard algebra of the angular momentum [26], the angular correlation function between the two photoelectrons can be written as

$$
W(\theta_1, \varphi_1; \theta_2, \varphi_2) = \pi \alpha \omega \sum_{k_1k_2k_{2'q_2j_2}} \tilde{B}(k_1, k_2, k_2') \rho_{k_1q_1}(J_1, J'_1; \theta_1, \varphi_1) h(J_1, J'_1) \rho^7_{k_20} \times (k_1q_1, k_20 | k_2q_1) \sqrt{\frac{4\pi}{2k_2 + 1}} Y^*_{k_2q_1}(\theta_2, \varphi_2), \quad (8)
$$

where the dynamical coefficients $\tilde{B}(k_1, k_2, k_2')$ are given in terms of the amplitudes of the second step ionization (see equation (13) in [10]). Using results of [27], the dynamical coefficients can be further reduced to the multielectron term-dependent amplitudes in the LS-coupling approximation. In equations (7) and (8), $k_1$ and $k_2$ can take only even values $\{0, 2, 4\}$ due to definite parities of the orbital momenta of the photoelectrons in each photoionization step. The depolarization factor $h(J_1, J'_1)$, discussed in detail in [10], accounts for the evolution of the statistical tensors (7) during the time between the two ionization steps. Extreme cases are the fully incoherent excitation of isolated states ($h(J_1, J'_1) \sim \delta_{J_1J'_1}$), fully overlapping states (pure LS-coupling states, $h(J_1, J'_1) \sim 1$), and fully coherent excitation of isolated states (scenario of the quantum beats). Here we adopt the first scenario by the following reasons. The width of the ionic ground doublet states $np^52P_{1/2,3/2}$ is determined by the field width from the incoming radiation, which in our case is smaller than the fine structure splitting $\Delta E = E_{1/2} - E_{3/2}$ even for neon ($\Delta E=0.097$ eV). Furthermore, the duration of the FLASH pulse in experiments on the angular distributions of photoelectrons in sequential TPDI was $\Delta t \geq 25$ fs [4, 5, 9], which corresponds to $\Delta \omega \leq 0.03$ eV in the energy domain. It follows that the fine structure $np^52P_{1/2,3/2}$ states can be considered as isolated and incoherently excited with a good accuracy.
Figure 6. Angular correlation functions for sequential TPDI of Kr through the intermediate $4p^5 2P_{3/2}$ and $2P_{1/2}$ states to the final Kr$^{++} 4p^4 3P_2$ state at photon energies of 40 eV (upper panels) and 60 eV (lower panels). Angles are counted from the direction of the photon polarization in the plane perpendicular to the photon beam. The electrons are emitted in the same half plane ($\varphi_1 = \varphi_2 = 0$). The 3D plots visualize the contour plots for ionization from the $2P_{3/2}$ state (second column). Lighter and darker greys indicate higher and lower values, respectively.

For neon and with confidence for heavier noble gases. Strictly speaking, consistent calculations must be performed, which are beyond the scope of the present report, for the dynamical Stark shifts and field broadening of the ionic ground state fine-structure levels to justify more rigorously the present approach. Note that the hyperfine interactions are not relevant as a reason for the depolarization, because the precession period of the angular momentum of the electronic shell due to the hyperfine interactions is a few orders of magnitude longer than the duration of the photon pulse.

In the angular correlation function (8), the dynamical coupling between the two steps of the sequential ionization is represented by summation over $k_i$. As soon as only the term with $k_i = 0$ (unpolarized ion after the first step ionization) remains, expression (8) turns into a product (5), characterizing independent photoionization events.

For the purpose of analysis, expansion of the angular correlation functions in terms of the bipolar harmonics is often convenient. Such an expansion for the function (8) reads

$$W(\vartheta_1, \varphi_1; \vartheta_2, \varphi_2) = (\pi \alpha \omega)^2 \sum_{K=0,2,4} \sum_{k_1k_2} C_{Kk_1k_2} 4\pi \{ Y_{k_1}(\vartheta_1, \varphi_1) \otimes Y_{k_2}(\vartheta_2, \varphi_2) \} K0 , \quad (9)$$

where

$$C_{Kk_1k_2} = \sum_{k_i} \sum_{k_{\gamma 1}=0,2} \sum_{k_{\gamma 2}=0,2} Z(k_1, k_2, k_{\gamma 1}, k_{\gamma 2}, k_i) (k_{\gamma 1}0, k_{\gamma 2}0 | K0) \left\{ \frac{k_{\gamma 1}}{k_2} \frac{k_{\gamma 2}}{k_1} K \right\} \rho_{k_{\gamma 1}0} \rho_{k_{\gamma 2}0} , \quad (10)$$
\[ Z(k_1, k_2, k_{\gamma_1}, k_{\gamma_2}, k_i) = \sum_{J_i, J'_i} h(J_i, J'_i) B(k_1, k_i, k_{\gamma_1}) B(k_i, k_2, k_{\gamma_2}). \]  

(11)

The zero projection of the bipolar harmonic in (9) is due to the fact that the angular correlation function depends on the difference of the azimuthal angles \( \phi_1 - \phi_2 \) but not on \( \phi_1 \) and \( \phi_2 \) separately.

Figure 6 shows examples of the angular correlation patterns in Kr, calculated according to equations (7) and (8). To reduce the number of dimensions, we consider a geometry when the electric field of the radiation and linear momenta of the emitted electrons belong to one plane. Comparing to the upper panels of figure 3, we conclude that the influence of the polarization of the intermediate \( ^2P_{1/2} \) state is negligible at 40 eV: the corresponding pattern in figure 6 is almost identical to the IE pattern in figure 3. With increasing of the photon energy the symmetry of the pattern violates, indicating a small influence of the orientation of the \( ^2P_{1/2} \) state. For \( \theta_1 = 0, \pi \) (emission of the first electron along the photon polarization), the state with \( J_i = 1/2 \) is isotropic: for the axis of symmetry defined by collinear polar vectors, the odd rank tensors of the system vanish [26]. As a result the second electron emission for this geometry remains symmetric with respect to the reflection \( \theta_2 \rightarrow \pi - \theta_2 \). In the case of the \(^2P_{3/2}\) intermediate state, the IE pattern modifies crucially. Instead of maxima when photoelectrons are emitted along the photon polarization (in the same direction or back-to-back) in the IE pattern (see figure 3), one can clearly see local minima in figure 6, right column. Besides, there is no more symmetry with respect to the reflection \( \theta_i \rightarrow \pi - \theta_i, i = 1, 2 \). These features are clear indications of the dynamical correlations between two emissions in TPDI. Some effects of correlation have been observed recently in experiments at FLASH for the Ne atom at 44 eV photon energy [9], yet with not very good statistics.

![Figure 6. Examples of the angular correlation patterns in Kr, calculated according to equations (7) and (8). To reduce the number of dimensions, we consider a geometry when the electric field of the radiation and linear momenta of the emitted electrons belong to one plane.](image)

**Figure 7.** Asymmetry coefficient \( \beta_4^{(1)} \) in the angular distribution of the first-step electron in sequential TDPI of the Kr atom via the intermediate \( \text{Kr}^+ 4p^5\,^2P_{3/2} \) state into different final states \( \text{Kr}^{++} 4p^4 \), marked in the figure.

5. **On the angular distribution of the first-step electron**

Integrating the angular correlation function (9) over the emission angles of the second electron, we obtain for the first-step electron the angular distribution of the form

\[ W(\theta_1) = \frac{W_0^{(1)}}{4\pi} \left[ 1 + \beta_2^{(1)} P_2(\cos \theta_1) + \beta_4^{(1)} P_4(\cos \theta_1) \right]. \]

(12)

This result seems to contradict the angular distribution of the first electron (3), presented earlier. However, one should take into account that expression (12) is obtained for the TPDI process when two photons are absorbed and two electrons are emitted albeit the direction of emission of the second electron is not detected. In general, two-photon absorption leads to the angular
distribution of the type (12), more complicated than the standard dipole distribution (3) inherent
in the one-photon absorption. Theory predicts that the coefficient $\beta_4^{(1)}$ for resolved multiplet
and fine structure states can be large (see figure 7). Similar to the asymmetry parameter $\beta_4^{(2)}$,
after averaging over the unresolved states, the value of $\beta_4^{(1)}$ drops down.

6. Conclusion
The theory of angular correlations in sequential TPDI predicts several effects which deserve
detailed experimental investigations, such as unusual shape of the angular distributions of pho-
toelectrons in non-coincidence experiments and the dynamical correlations between emitted
electrons connected with the polarization of the intermediate states. On the other side, when
the polarization (alignment) is negligible, the study of the spectra and angular distributions
of the second step electrons in sequential TPDI provides valuable information about the pho-
toionization of ions, the process which is difficult to investigate by standard methods with low
intensity sources. Besides, there are interesting problems in TPDI which are beyond the scope
of the present report such as the problem of quantum interference, quantum entanglement in the
continuum and concurrence. The study of TPDI is only at the beginning and promises many
interesting discoveries.

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