Angular distributions and correlations in sequential three-photon triple atomic ionization

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Abstract. A theoretical description of the sequential three-photon triple atomic ionization is given within a model of stepwise ionization with emphasis on the angular distributions and angular correlations of the emitted electrons. The theory is applied to the photoionization of neon atom at 90.5 eV, corresponding to the experiment performed at the Free electron LASer in Hamburg (FLASH) by A. Rouzée et al (Phys. Rev. A 83 031401(R) (2011)).

1. Introduction

The new sources of VUV and X-ray radiation based on free-electron lasers in self-amplification spontaneous emission regime (FLASH in Germany, SCSS in Japan, LCLS in USA) have given a base for a number of new topics in physics. In particular, the high brightness of the modern facilities enables one to observe the ionization and the formation of positive multicharged ions. In such experiments, the ionic target is created with quite some density and continues to interact with photons from the same femtosecond laser pulse. The double ionization of atoms by two or three VUV photons has provoked a lot of scientific attention as it represents one of the simplest non-linear processes in atomic continua. The multiple ionization of atoms can proceed either directly due to the simultaneous absorption of several photons or sequentially via some intermediate states of the ion. It is well established that the second mechanism usually dominates if the laser frequency exceeds ionic ionization threshold [1, 2]. However, even if the laser frequency is not enough to ionize the ion by the single photon absorption, the sequential three-photon double ionization may become dominant at high enough intensity due to the two-photon ionization of the intermediate ion on the second step. Such a three-photon double ionization has been observed both for resonant and non-resonant second step [3, 4, 5, 6].

Starting from the first observation of sequential two-photon double ionization (2PDI) [7, 8], the experiments including studies of angular distribution and angular correlation functions of the ejected electrons has been giving fruitful information about mechanisms of atomic and ionic photoionization [9, 10, 11, 12, 13, 14]. First data on the sequential three-photon triple ionization (3PTI) (as well as on the attendant 2PDI), were obtained at FLASH with atomic neon [15],...
and were accompanied by a theoretical analysis that proves the sequential nature of the observed resonances. In this contribution we extend the theoretical analysis of sequential 3PTI with an emphasis on the angular distributions and angular correlations of emitted electrons.

2. Theory

The theoretical description is based on a stepwise model and the statistical tensor formalism [16, 17]. Statistical tensors being a result of unitary transformation of the angular momentum density matrix contain information about polarization of a quantum system and include amplitudes of transitions to particular states of the system. This approach has been successfully applied to the sequential 2PDI from the outer p-shells of the noble gas atoms [11, 18, 19, 20], d-shell of xenon [21], and three-photon resonantly enhanced sequential double ionization of argon [3]. By assuming an incoherent excitation of ionic fine-structure levels, the angular correlation function of the photoelectrons in the \( n \)-fold ionization with the last step

\[
\gamma_n + A^{(n-1)+}(\alpha_{i_{n-1}} J_{i_{n-1}}) \rightarrow A^{n+}(\alpha_{i_n} J_{i_n}) + e_n^-
\]

is expressed as

\[
W(\{\Theta_n, \Phi_n\}) = \sum_{k_{i_{n-1}} q_{i_{n-1}}} \rho_{k_{i_{n-1}} q_{i_{n-1}}} (J_{i_{n-1}}; \{\Theta_{i_{n-1}}, \Phi_{i_{n-1}}\}) \rho_{k_{n} 0}^n
\]

\[
\times (k_{i_{n-1}} q_{i_{n-1}}, k, m, 0 | k_{i_n} q_{i_n-1}) \tilde{B}_n(k_{i_{n-1}}, k, m, n) \sqrt{\frac{4\pi}{k_n}} Y_{k_n q_{i_n-1}} (\vartheta_n, \varphi_n).
\]

Here \( \{\Theta_n, \Phi_n\} = \{\vartheta_1, \vartheta_2, ..., \vartheta_n; \varphi_1, \varphi_2, ..., \varphi_n\} \) is the set of emission angles of the \( n \) electrons, \( J_{i_{n-1}}, J_{i_n} \) are the total angular momenta of the initial, \( A^{(n-1)+}(\alpha_{i_{n-1}} J_{i_{n-1}}) \), and final, \( A^{n+}(\alpha_{i_n} J_{i_n}) \), ionic states in the \( n \)-th ionization step (\( J_{i_0} = 0 \) for rare gases) and \( \alpha_{i_{n-1}}, \alpha_{i_n} \) are the sets of quantum numbers to specify these states uniquely; \( k \equiv \sqrt{2\ell + 1} \), the standard notations for Clebsch-Gordan coefficients are used and \( \rho_{k_{n} 0}^n \) are the statistical tensors of the \( n \)-th absorbed photon. In our case of linearly polarized photon beam, all photons irrespective of their number \( n \) possess only two nonvanishing statistical tensors, \( \rho_{30}^0 = 1/\sqrt{3} \) and \( \rho_{30}^0 = -\sqrt{2/3} \).

The statistical tensors of the intermediate ion \( A^{m+}(\alpha_{i_m} J_{i_m}) \) (i.e. before the \( (m+1) \)-th ionization step) are given by the recurrent relation

\[
\rho_{k_{i_m} q_{i_m}} (J_{i_m}; \{\Theta_m, \Phi_m\}) = \sum_{k_{i_{m-1}} q_{i_{m-1}}} \hat{k}_{e_m} \hat{k}_{i_m} (k_{i_m} q_{i_m}, k_{e_m} q_{e_m} | k_{i_{m-1}}) \rho_{k_{m} 0}^{n-1}
\]

\[
\times (k_{i_{m-1}} q_{i_{m-1}}, k_{e_m} 0 | k_{i_m} q_{i_m-1}) \rho_{k_{e_m} 0}^{n-1} \tilde{B}_m(k_{i_{m-1}}, k_{e_m}, k_{e_m}) \sqrt{\frac{4\pi}{k_{e_m}}} Y_{k_{e_m} q_{i_m-1}} (\vartheta_m, \varphi_m)
\]

in terms of the statistical tensors of the intermediate ion at the previous ionization step. Equation (2) is a generalization of equation (16) of [17] to the \( n \)-fold ionization, provided that the intermediate ionic states are excited incoherently. The dynamical factors \( \tilde{B}_n(k_{i_{n-1}}, k, m, n) \) depend on dipole amplitudes for the \( n \)-th ionization step. They are given by expression (13) of [17] with obvious change in notations for the angular momenta quantum numbers:

\[
\tilde{B}_n(k_{i_{n-1}}, k, m, n) = \hat{k}_{i_{n-1}} \hat{k}_m \sum_{\ell_n \ell_n' \ell_n'' \ell_n'''} (-1)^{\ell_n''+1/2} \mathcal{J}_{n}^{\ell_n \ell_n' \ell_n'' \ell_n''' \ell_n' n} J_{n}^{\ell_n \ell_n' \ell_n'' \ell_n''' \ell_n' n} \ell_n' n | k_{i_{n-1}}
\]

\[
\times \begin{pmatrix} J_{i_{n-1}} & 1 & J_{n} \\ J_{i_{n-1}}' & 1 & J_{n}' \\ k_{i_{n-1}} & k_{n} & k_{n} \end{pmatrix} D_{J_{i_{n-1}} \alpha_{n} J_{i_{n-1}} \ell_n \alpha_{n} J_{i_{n-1}}'} D_{J_{n} \alpha_{n} J_{n} \ell_n' \alpha_{n} J_{n}'}.
\]
where $D_{J_{in-1},\alpha_J}\ell_{jn}\ell_{jn}\ell_{in}\ell_{in}\ell_{in} = \langle \alpha_{in},\ell_{jn},\ell_{jn}\ell_{in}\ell_{in}|| J_{in-1} \rangle$ is the reduced dipole matrix element.

From equations (2) and (3), the anisotropy parameters $\beta_{J}^{m}$ of the angular distribution of the photoelectrons $e_{m}$

$$W(\theta_{m}) = \frac{W_{m}}{4\pi} \left(1 + \sum_{k=2,4,..}^{k_{max}} \beta_{k}^{m} P_k(\cos \theta_{m})\right) \quad (5)$$

can be found by integration over the emission angles of all except the m-th electron. In equation (5), $P_k(x)$ is the Legendre polynomial and $W_{m}$ is the angle-integrated emission probability. In neon, the maximum rank of the asymmetry parameters in equation (5) is restricted by $k_{max} = 2$ and $k_{max} = 4$ if the ionization proceeds from either an s and/or p shell, respectively. More general, $k_{max}$ can reach the value of $2n$. Integrating (2) over the emission angles of all except l-th and m-th step electrons, one obtains the angular correlation function between these two electrons. Note that electrons from different ionization steps are distinguished by their kinetic energy. It has been shown in [11, 17, 18, 19, 22] that, in contrast to some first intuition,

![Figure 1](image)

**Figure 1.** Experimental (dots) and theoretical (red bold line) electron spectra under 54.7°. Calculated contributions from 2PDI (bold dashed), and 3PTI (thin for ionization from 2p shell and thin dashed for ionization involving 2s shell). Numbers correspond to the processes in table 1. The calculated SI, 2PDI, and 3PDI spectra are convoluted with the Gaussian (FWHM = 1 eV) and independently normalized to measured intensities of the lines at 69.0 eV (process 1), 46.4 eV (process 3), and 19.4 eV (process 10), respectively.

the photoelectron angular distributions are different in single ionization (SI) and in the same ionization transition constituting the first ionization step of the 2PDI. This difference is caused by dynamical correlations between the two ionization steps of the 2PDI due to polarization of the intermediate ion. Similar correlations occur in the 3PTI and influence the angular distributions of photoelectrons due to the occurrence of some step of the photoionization. Calculations for the 2PDI from the 2p subshell in Ne [11, 17], show that the influence of the dynamical correlations on the angular distributions of the first-step photoelectrons is small in the broad interval of the photon energies. Furthermore, the correlation effect smears out to a large extent after summing up over different unresolved ionization paths. Therefore the correspondingly simplified stepwise model can be often used for comparison with experiments [15].

### 3. Numerical results and discussion

The photoelectron spectrum of neon generated by 90.5 eV pulses from FLASH and detected under the angle 54.7° with respect to the laser electric field is displayed in figure 1 together with its theoretical simulation. Details of the experiment and the spectrum analysis have been
Table 1. Processes contributing to the photoelectron spectrum with corresponding asymmetry parameters of the photoelectron angular distribution.

| $E_{kin}^{exp}$ eV | Photoionization process | Initial state | Final state | N | Type | $E_{kin}$ eV | $\beta_2$ | $\beta_4$ | $\beta_2$ | $\beta_4$ |
|-------------------|-------------------------|---------------|-------------|---|------|-------------|----------|----------|----------|----------|
| 69.0              | Ne 2p$^5$              | Ne$^+2p^5$    | 1$^c$ SI    | 68.9 | 1.41 | 0.00 | 1.41 | 0.00 |
| 49.5              | Ne$^+$2s2p$^5$         | Ne$^{2+}2s2p^53P$ | 2 | 2PDI | 51.1 | 1.30 | 0.08 | 2.00 | 0.00 |
| 46.4              | Ne$^+$2p$^5$           | Ne$^{2+}2p^35P$ | 3$^c$ 2PDI | 49.5 | 1.37 | -0.05 |
| 42.0              | Ne$^+$2s2p$^5$         | Ne$^{2+}2s2p^15S$ | 4$^c$ 2PDI | 46.4 | 1.13 | 0.14 | 1.11 | 0.01 |
| 35.5              | Ne 2p$^6$              | Ne$^+2s2p^6$  | 6 | SI | 42.1 | 1.91 | 0.16 | 2.00 | 0.00 |
| 30.7              | Ne$^+$2s5p$^{2}P$      | Ne$^{2+}2s2p^{2}D$ | 8 | 3PTI | 31.4 | 1.43 | 0.01 |
| 27.1              | Ne$^+$2p$^5$           | Ne$^{2+}2s2p^35P$ | 9 | 3PTI | 29.6 | 1.38 | 0.00 |
| 25.2              | Ne$^+$2s2p$^5$         | Ne$^{2+}2s2p^{1}D$ | 10$^c$ 3PTI | 27.1 | 1.35 | -0.05 |
| 22.0              | Ne$^+$2p$^{1}D$        | Ne$^{2+}2p^{1}S$ | 11 | 3PTI | 26.5 | 1.35 | 0.05 |
| 19.4              | Ne$^+$2p$^{1}S$        | Ne$^{2+}2p^{1}D$ | 12$^c$ 3PTI | 26.3 | 1.3 | 0.00 |
| 13.5              | Ne$^+$2s5p$^{3}P$      | Ne$^{2+}2s2p^{3}P$ | 14 | 2PDI | 24.2 | 1.29 | 0.05 | 2.00 | 0.00 |
|                   | Ne$^+$2s2p$^{3}P$      | Ne$^{2+}2s2p^{3}P$ | 15 | 3PTI | 23.3 | 1.30 | -0.07 |
|                   | Ne$^+$2s$^{6}P$        | Ne$^{2+}2s^{6}P$ | 16$^c$ 3PTI | 22.6 | 1.40 | 0.18 |
|                   | Ne$^+$2s5p$^{3}P$      | Ne$^{2+}2s2p^{3}D$ | 17$^c$ 3PTI | 22.1 | 1.31 | -0.04 | 1.30 | 0.00 |
|                   | Ne$^+$2s2p$^{3}P$      | Ne$^{2+}2s2p^{3}D$ | 18 | 3PTI | 20.9 | 1.27 | 0.00 |
|                   | Ne$^+$2s$^{6}P$        | Ne$^{2+}2s^{6}P$ | 19$^c$ 3PTI | 19.4 | 0.89 | -0.14 | 1.37 | 0.02 |
|                   | Ne$^+$2s5p$^{3}P$      | Ne$^{2+}2s2p^{3}S$ | 20 | 3PTI | 15.3 | 1.27 | 0.01 |
|                   | Ne$^+$2s2p$^{5}$       | Ne$^{2+}2s2p^{1}S$ | 21 | 2PDI | 13.7 | -0.03 | 2.00 | 0.00 |
|                   | Ne$^+$2s$^{4}P$        | Ne$^{2+}2s^{2}P$ | 22 | 3PTI | 12.8 | 1.19 | 0.00 |

* $a$ data from [15] and private communication. The energies of the lines ($E_{kin}^{exp}$) could be determined no better than up to 1 eV. The estimated accuracy of the experimental values of $\beta_2$ and $\beta_4$ is not worse than 10%.
* $b$ energies derived from spectroscopic data on the energy levels [23].
* $c$ photoionization from the 2p-shell, mostly dominating the spectrum.

In the description of photoionization in the 2s shell of neon was discussed in [25]. Last two columns of table 1 contain calculated asymmetry parameters by assuming an incoherent excitation of the ionic intermediate and final fine-structure states. When compared with experiment in the energy region in which several processes contribute, the laser pulse shape and the 2s-hole decay rates should be taken into account. These procedures have been done for some of the resonances in [15]. The $\beta_2$ parameters in different processes involving the 2p electron, both 2PDI and 3PTI, respectively. Nevertheless, as seen from table 1, not a single process may contribute to a certain observed line.

Theoretical simulation of the photoelectron spectrum was performed within the two- and three-step model of the 2PDI and the 3PTI, respectively, and using multiconfiguration Hartree-Fock method for generating photoionization amplitudes in the dipole approximation [24]. At every stage we used term-dependent wave functions for initial states and performed full self-consistent LS-coupling calculations by including the reference configuration $1s^22s^22p^3$ as well as all those with single and double replacements of 2s and 2p by virtual 3s, 3p, 3d orbitals. For the final states (ion + electron), the orbitals taken from the initial-state calculations were frozen and only continuum electron wave functions were optimized. Appropriateness of such a method for description of photoionization in the 2s shell of neon was discussed in [25].
states. The alignment of Ne$^+$ step ionization may be more pronounced [19, 22] due to larger alignment of intermediate ionic gases, as follows from the analysis of the 2PDI, the effect of the consecutive steps on the 1st explicitly provided that the consecutive photoionization steps do not occur. In heavier noble gases, as follows from the analysis of the 2PDI, the effect of the consecutive steps on the 1st step ionization may be more pronounced [19, 22] due to larger alignment of intermediate ionic states. The alignment of Ne$^+$ and Ne$^{2+}$ after the 1st and the 2nd ionization steps is only around -0.1÷-0.2 in the wide range of the photon energies, provided the photoelectron is not detected.

Table 2 illustrates the influence of the 2nd and the 3rd ionization steps on the angular distribution of the 1st step photoelectron: the anisotropy coefficients $\beta^1_k$ depend on the whole 3PTI path although the 1st step is the same for all the cases. The coefficient $\beta^1_k$ would vanish explicitly provided that the consecutive photoionization steps do not occur. In heavier noble gases, as follows from the analysis of the 2PDI, the effect of the consecutive steps on the 1st step ionization may be more pronounced [19, 22] due to larger alignment of intermediate ionic states. The alignment of Ne$^+$ and Ne$^{2+}$ after the 1st and the 2nd ionization steps is only around -0.1÷-0.2 in the wide range of the photon energies, provided the photoelectron is not detected.

For example, the alignment of the Ne$^{2+}$ 2p$^4$ 3P$_2$ ($^1D_2$) states (presented in table 2) after the second ionization step is $A_{20} = 0.05$ ($A_{20} = -0.17$), respectively, for the photon energy of 90.5 eV. The fourth rank tensors $A_{i0}$ for these Ne$^{2+}$ states are, correspondingly, 0.00 and -0.02, and should not noticeably influence the dynamics of the 3PTI.

Figure 2 displays the contour plots of the angular correlation functions between pairs of photoelectrons emitted in the 3PTI. To reduce the four-dimensional (hyper-) surfaces to two dimensions, we took the geometry as in previous discussions of the angular correlations in sequential 2PDI [11, 17, 19, 26] and measurements [11]: emission angles $\theta_j$ ($j = 1, 2, 3$) of the two electrons are counted from the direction of the photon linear polarization in the plane perpendicular to the photon beam and are in the same half plane $\varphi_i = \varphi_j = 0$ ($i, j = 1, 2, 3$).

The $e_1$-$e_2$ correlation patterns (right column) are similar to those of the 2PDI from the 4p subshell in krypton (figure 3 of [20] and figure 6 of [19]) and are close to those obtained for the 2PDI of neon (figure 3 of [26] taking into account that here we separated the intermediate $2P_{3/2}$ state). A concept of ‘true’ correlations was introduced in [20] as a difference between the angular correlation function of two electrons and the product of two independent angular distributions of the two electrons. As in [20], the $e_1$-$e_2$ contour plots for the ionization path including Ne$^{2+}$ 2p$^4$ 1D characterize a very weak ‘true’ correlations between the 1st and the 2nd step ionization, while the correlation is much more pronounced for the paths involving the Ne$^{2+}$ 2p$^4$ 3P$_2$ state. Interestingly, while the $e_1$-$e_2$ correlation patterns in the 3PTI via the Ne$^{2+}$ 2p$^4$ 1D state is characteristic for the weak ‘true’ correlations (right column, two upper panels), the corresponding $e_2$-$e_3$ correlations (left column, two upper panels) indicate strong ‘true’ correlations. The opposite situation occurs for the case of the Ne$^{2+}$ 2p$^4$ 3P$_2$ level which is involved within the 3PTI (last three rows). The ‘true’ $e_1$-$e_3$ correlations (middle column) are weak, which is not surprising because of an additional ionization step between the two emissions.
Figure 2. Contour plots of angular correlation functions between two of the three electrons, in the 3PTI of neon: e₂ and e₃ (first column); e₁ and e₃ (second column); e₁ and e₂ (third column). Here e₁, e₂ and e₃ are photoelectrons produced in the first, the second and the third ionization steps, respectively. Rows correspond to different ionization paths: terms of the ionic states, Ne⁺(2p⁵), Ne²⁺(2p⁴), and Ne³⁺(2p³) after each of the three sequential ionization steps are marked on the right as I₁, I₂, and I₃, respectively. See text for explanation of the angles.

4. Conclusion
The theory of the sequential ionization processes is extended to the description of angular distributions and angular correlation functions of photoelectrons in the n-fold n-photon ionization. For the three-photon triple ionization of neon at 90.5 eV, the angular correlation patterns between pairs of the emitted photoelectrons are calculated, as well as the angular asymmetry parameters in the angular distributions. Similar 2p-shell ionization dynamics in Ne 2p⁶, Ne⁺ 2p⁵, and Ne²⁺ 2p⁴ states and small values of the alignment of the intermediate ionic states lead to similar angular distributions of photoelectrons in one-photon single ionization, sequential two-photon double ionization and sequential three-photon triple ionization. In
particular, the influence of the consecutive ionization steps on the angular distributions of photoelectrons from preceding steps is demonstrated. Angular correlation patterns between pairs of the emitted electrons contain rich information on the dynamics of the sequential three-photon triple ionization, for example, they crucially depend on the terms of intermediate ions.

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