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FAST TRACK COMMUNICATION

Sequential two-photon double ionization of noble gas atoms

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Online at stacks.iop.org/JPhysB/40/F313**Abstract**

We develop a formalism which describes sequential two-photon double ionization (TPDI) of a closed-shell atom. We apply this formalism to calculate the angular asymmetry parameters of sequential TPDI of the Ne 2p and Ar 3p valence subshells. Comparison with the latest experimental data is made.

In recent years, strong-field ionization of noble gas atoms has been explored extensively using the Free electron LASer at Hamburg FLASH). Wabnitz *et al* (2005) studied multiple ionization of Ar and Xe atoms at the photon energy of $\omega = 13$ eV. Moshhammer *et al* (2007) reported few-photon multiple ionization of Ne and Ar at $\omega = 38.8$ eV. Sorokin *et al* (2007) measured direct, sequential and resonant multi-photon ionization and excitation processes in Ne at $\omega = 42.8$ eV. Using an alternative high harmonic generation technique, Benis *et al* (2006) observed two-photon double ionization (TPDI) of Ar and Kr atoms by superposition of harmonics. Besides the total ionization rate, the latest experiments are aiming to obtain various differential cross-sections. Very recently, Braune *et al* (2007) reported angular anisotropy β parameters in sequential TPDI of Ne at 47.5 eV and Ar at 38 eV. Recoil ion momentum-resolved measurements of Moshhammer (2007) on Ne at 45 eV can also be interpreted in terms of these β parameters.

On the theoretical side, little has been done so far to support these angular-resolved measurements. The only related work is a calculation of Kazansky and Kabachnik (2006) who reported angular distributions of the Auger 2p and 3d electrons in Ar and Kr in coincidence with photoelectrons in femtosecond and attosecond pulse regimes.

In the meantime, a theoretical framework to evaluate β parameters in sequential TPDI of noble gas atoms can be readily developed. In a sequential regime, the TPDI process takes place as subsequent one-photon single ionization (OPSI) of the neutral atom and the singly charged ion. A competing non-sequential TPDI process, driven by electron correlations, is negligible in femtosecond pulse regime (Moshhammer 2007, Braune *et al* 2007). The photoelectron angular distribution in OPSI can be evaluated for an arbitrary atomic target (Jacobs and Burke 1972, Dill *et al* 1975). The only missing link with the present measurements of β parameters in sequential TPDI of noble gases is coupling of the angular momenta of two photons, two

photoelectrons and two residual ions. This coupling can be worked out using the graphical angular momentum summation technique outlined, e.g., by Varshalovich *et al* (1988).

In the present communication, we perform this task and derive the expressions for β parameters in sequential TPDI of a closed-shell atomic target. We use these expressions to evaluate the angular anisotropy parameters for TPDI of the outer valence subshells of Ne and Ar in a wide photon energy range. We make a comparison of the calculated parameters with available experimental data at few selected photon energies.

We illustrate our graphical method by first deriving well-known expressions of the β_2 parameter in OPSI of an arbitrary atomic target. We write the matrix element of the dipole operator between the initial and final many-electron states using notations of Sobelman (1972) in which we omit the spin variables:

$$\langle l_0^n L_0 M_0 | D | l_0^{n-1} [L_a] l L M \rangle = \sqrt{n} G_{L_a}^{L_0} (-1)^{L-M} \begin{pmatrix} L & 1 & L_0 \\ -M & \mu & M_0 \end{pmatrix} \langle l_0^n L_0 || D || l_0^{n-1} [L_a] l L \rangle. \quad (1)$$

Here $D = e \sum_{i=1}^n r_i$ is the dipole operator, e is the polarization vector and $G_{L_a}^{L_0}$ is a fractional parentage coefficient. The expression for the reduced dipole matrix element is given by Chen *et al* (2003):

$$\langle l_0^n L_0 || D || l_0^{n-1} [L_a] l L \rangle = \sqrt{\frac{n}{2}} G_{L_a}^{L_0} \hat{L}_0 \hat{L} (-1)^{L+L_a+l_0+1} \begin{Bmatrix} 1 & L_0 & L \\ L_a & l & l_0 \end{Bmatrix} \langle v_0 || d || v \rangle, \quad (2)$$

where the one-electron reduced dipole matrix element is defined according to Amusia (1990):

$$\langle v_0 || d || v \rangle = (-1)^{l >} \sqrt{l >} \int r^2 dr R_{n_0 l_0}(r) R_{kl}(r). \quad (3)$$

Here $l >$ is greater than l_0 and l . In equations (1) and (2) we use the standard notations for the $3j$ and $6j$ symbols. For a closed-shell system, the factor preceding the one-electron reduced dipole matrix element in equation (2) is equal to 1.

Using these notations, we can write the differential, with respect to the photoelectron momentum, cross-section as

$$\frac{d\sigma}{d\mathbf{k}} = \frac{1}{2L_0 + 1} \sum_{M_0} \left| \sum_{\substack{LM \\ lm}} \hat{L} (-1)^{L+L_a+l} \begin{pmatrix} L & 1 & L_0 \\ -M & \mu & M_0 \end{pmatrix} \begin{pmatrix} L_a & l & L \\ M_a & m & -M \end{pmatrix} D_{lL} Y_{lm}(\hat{\mathbf{k}}) \right|^2. \quad (4)$$

Here $\mu = 0, 1$ is associated with the linear and circular polarizations, respectively. The hat symbol denotes $\hat{L} = \sqrt{2L+1}$. Following Jacobs and Burke (1972), we introduce in equation (4) a shortcut for the phase-modulated and normalized dipole matrix element

$$D_{lL} \equiv 2\pi (\omega/c)^{1/2} \exp(i\pi/2 + \delta_l) \langle l_0^n L_0 || D || l_0^{n-1} [L_a] l L \rangle. \quad (5)$$

The product of two spherical harmonics in equation (4) can be transformed using the Clebsch-Gordan series given by equation (5.6.9) (equation (9) of section 5.6) of Varshalovich *et al* (1988)

$$Y_{lm}(\hat{\mathbf{k}}) Y_{l'm'}(\hat{\mathbf{k}}) = \frac{1}{4\pi} \hat{l} \hat{l}' \sum_{JM_J} \hat{j}^2 \begin{pmatrix} l & l' & J \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l' & J \\ m & m' & -M_J \end{pmatrix} Y_{JM_J}(\hat{\mathbf{k}}). \quad (6)$$

Thus, equation (4) contains the sum of five $3j$ symbols running over six angular momentum projections which can be exhibited graphically by the left-hand side diagram of figure 1

The sum can be readily evaluated using equation (12.1.13) of (Varshalovich *et al* 1988) which leads to the standard photoelectron angular distribution

$$d\sigma/d\mathbf{k} = (4\pi)^{-1} d\sigma/dE [1 + \beta_2 P_2(\cos \theta)]$$

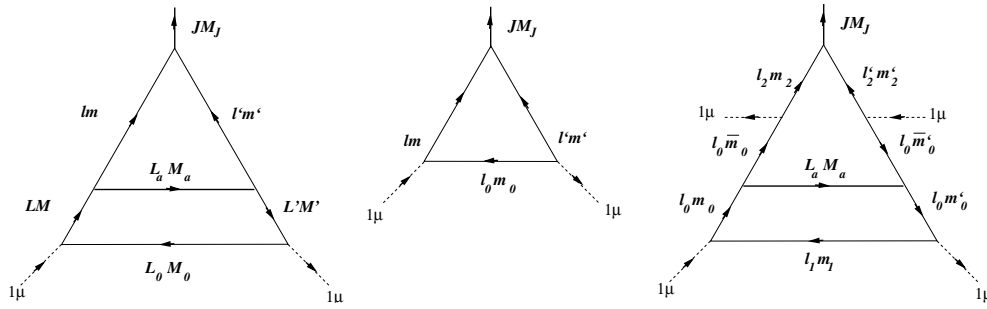


Figure 1. Angular momentum coupling schemes for one-photon single ionization (left and centre) and two-photon double ionization (right). The vertex denotes a $3j$ symbol. The sign of the angular momentum projection is indicated by the arrow, incoming with the plus sign and outgoing with the minus sign.

with $\beta_2 = A_2/A_0$. The coefficients A_J are given for linear polarization ($\mu = 0$) by the following expression:

$$A_J = \frac{(-1)^{L_a+L_0}}{2L_0+1} \hat{j}^2 \sum_{\substack{L' \\ l'}} \hat{l}' \hat{L}' \hat{L} \\ \times \begin{pmatrix} J & l & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} J & 1 & 1 \\ L_0 & L & L' \end{Bmatrix} \begin{Bmatrix} J & l & l' \\ L_a & L' & L \end{Bmatrix} \text{Re}\{D_{lL} D_{l'L'}^*\}$$

Equation (7) is equivalent to equations (3)–(4) by Jacobs and Burke (1972).¹

When the electron–ion interaction is isotropic, we can neglect the effect of the total spherical symmetry of the ionized atom on the photoelectron and factor out the explicit L -dependence of the dipole matrix elements from equations (2) and (5):

$$D_{lL} \equiv d_l \hat{L} \hat{L}_0 (-1)^L \begin{Bmatrix} 1 & L_0 & L \\ L_a & l & l_0 \end{Bmatrix}. \quad (7)$$

The sum over the total angular momenta L, L' can now be carried over using successively equations (12.2.18) and (12.2.7) of Varshalovich *et al* (1988). This finally leads to

$$A_J = \frac{1}{2l_0+1} \hat{j}^2 \sum_{l, l'=l_0 \pm 1} \hat{l}' \begin{pmatrix} J & l & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} 1 & 1 & J \\ l & l' & l_0 \end{Bmatrix} \text{Re}\{d_l d_{l'}^*\} \quad (8)$$

The asymmetry parameter $\beta_2 = A_2/A_0$ derived from equation (8) does not contain atomic or ionic quantum numbers. The fractional parentage coefficient occurs both in A_0 and A_2 and cancels out. Thus the β_2 parameter can be expressed solely by the one-electron dipole matrix elements of equation (3). This result can be obtained straightforwardly if we consider the photoionization in the independent electron approximation and describe it by the angular momentum coupling scheme exhibited by the central diagram of figure 1. Explicit formulae for the $3j$ and $6j$ symbols can be used to transform equation (8) into a closed-form expression for β_2 (Amusia 1990).

¹ Notations of Jacobs and Burke (1972) are ambiguous as the angular momenta of the atom and the ion are both denoted by the same symbol. When unambiguous notations are employed, the equivalence of the original equations and the present equation (7) becomes evident.

Now we extend our formalism to the case of sequential TPDI. In analogy to equation (4), cross-section of this process can be written as

$$\begin{aligned} \frac{d\sigma}{d\mathbf{k}_1 d\mathbf{k}_2} = & \frac{1}{2L_0 + 1} \sum_{M_0 M_a} \left| \sum_{L_i M_i} \sum_{L_1 M_1} \sum_{\substack{L_2 M_2 \\ l_1 m_1 \\ l_2 m_2}} (-1)^{L_1 + L_i + l_1} \begin{pmatrix} L_1 & 1 & L_0 \\ -M_1 & \mu & M_0 \end{pmatrix} \begin{pmatrix} L_i & l_1 & L_1 \\ M_i & m_1 & -M_1 \end{pmatrix} \right. \\ & \times D_{l_1 L_1} Y_{l_1 m_1}(\mathbf{k}_1) (-1)^{L_2 + L_a + l_2} \begin{pmatrix} L_2 & 1 & L_i \\ -M_2 & \mu & M_i \end{pmatrix} \\ & \left. \times \begin{pmatrix} L_a & l_2 & L_2 \\ M_a & m_2 & -M_2 \end{pmatrix} D_{l_2 L_2} Y_{l_2 m_2}(\mathbf{k}_2) \right|^2. \end{aligned} \quad (9)$$

Here we adopt the following angular momentum coupling scheme:

$$l_0^n [L_0] + \gamma \rightarrow l_0^{n-1} [L_i] + k_1 l_1 L_1, \quad l_0^{n-1} [L_i] + \gamma \rightarrow l_0^{n-2} [L_a] + k_2 l_2 L_2.$$

By integrating equation (9) over $d\mathbf{k}_1$ and employing the Clebsch–Gordan series (6), we end up with the expression containing the sum of nine $3j$ symbols running over eleven angular momentum projections. This sum can be exhibited by a triangular diagram similar to the one on the left of figure 1 but with a larger number of ‘ladder steps’. Varshalovich *et al* (1988) do not provide a direct summation formula for this case. Nevertheless, the summation can be carried over using reduction formulae (12.1.10) and (12.1.11) which reduce the number of ladder steps by 1. As a result, the angular distribution of the photoelectron is given by the Legendre polynomial expansion $d\sigma/d\mathbf{k}_2 = (4\pi)^{-1} d\sigma/dE_2 [1 + \beta_2 P_2(\cos \theta_2) + \beta_4 P_4(\cos \theta_2)]$, where the angular anisotropy coefficients $\beta_J = A_J/A_0$ can be derived from the following coefficients:

$$\begin{aligned} A_J = & \hat{j}^2 \sum_{l_1 l_2 l'_2} (-1)^{L_0 + L_a + l_1} \hat{l}_2 \hat{l}'_2 \sum_{\substack{l_1 l'_1 \\ l_2 l'_2}} \sum_{L_i L'_i} \begin{pmatrix} l_2 & l'_2 & J \\ 0 & 0 & 0 \end{pmatrix} \text{Re} \{ \mathcal{D}_{l_1 l_2 L_1 L_2} \mathcal{D}_{l_1 l'_2 L'_1 L'_2}^* \} \begin{Bmatrix} L_2 & L'_2 & J \\ l_2 & l_2 & L_a \end{Bmatrix} \\ & \times \sum_{KK'} \hat{K}^2 \hat{K}'^2 (-1)^{K+K'} \begin{pmatrix} 1 & K & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & K' & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} K & K' & J \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} 1 & 1 & K \\ L_1 & L'_1 & L_0 \end{Bmatrix} \begin{Bmatrix} L_i & L'_i & K \\ L'_1 & L_1 & l_1 \end{Bmatrix} \begin{Bmatrix} K & L'_i & L_i \\ K' & 1 & 1 \\ J & L'_2 & L_2 \end{Bmatrix}. \end{aligned} \quad (10)$$

Here we denoted $\mathcal{D}_{l_1 l_2 L_1 L_2} = D_{l_1 L_1} D_{l_2 L_2}$ for brevity of notations and set $\mu = 0$.

For a closed atomic shell, $L_0 = 0$ and $L_i = L'_i = l_0$, $L_1 = L'_1 = 1$. Further simplification of equation (10) can be achieved if we neglect the dependence of the wavefunction of the second photoelectron on the total angular momentum of the singly ionized atomic system L_2 . Then the explicit L_2 dependence of the matrix element $D_{l_2 L_2}$ can be factored out using equation (7), and the summation over L_2, L'_2 can be carried over. This leads to the anisotropy parameters $\beta_J = A_J/A_0$ which can be extracted from the following coefficients:

$$\begin{aligned} A_J = & \hat{j}^2 \sum_{l_1 l_2 l'_2} (-1)^{l_1 + l_2 + l'_2} \hat{l}_2 \hat{l}'_2 \begin{pmatrix} l_2 & l'_2 & J \\ 0 & 0 & 0 \end{pmatrix} |d_{l_1}|^2 \text{Re} \{ d_{l_2} d_{l'_2}^* \} \\ & \times \sum_{KK'} \hat{K}^2 \hat{K}'^2 (-1)^{K'} \begin{pmatrix} 1 & K & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & K' & 1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} K & K' & J \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} 1 & 1 & K \\ l_0 & l_0 & l_1 \end{Bmatrix} \begin{Bmatrix} l_0 & l_0 & K \\ l_0 & l_0 & L_a \end{Bmatrix} \begin{Bmatrix} K & l_0 & l_0 \\ K' & 1 & 1 \\ J & l_2 & l_2 \end{Bmatrix}. \end{aligned} \quad (11)$$

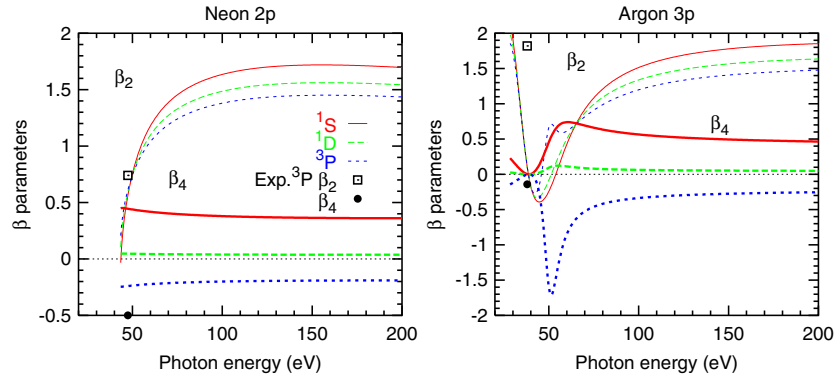


Figure 2. The angular anisotropy β -parameters for Ne (left) and Ar (right). The experimental data are from Braune *et al* (2007).

As in the case of equation (8), the β parameters extracted from equation (11) do not contain the ionic or atomic quantum numbers except for the total angular momentum of the doubly charged ion L_a . Equation (11) can be derived straightforwardly if we consider an independent electron TPDI process in which the angular momenta of the two holes couple to the total angular momentum L_a . This process is exhibited graphically by the right-hand side diagram of figure 1.

We use equation (11) to evaluate the β_J parameters of TPDI of Ne 2p and Ar 3p valence subshells. Isotropy of the system containing the doubly charged ionic core and the photoelectron, which is essential for application of equation (11), can be judged, at least partly, from the properties of the discrete excited states of the singly charged ion. Inspection of the Ne II energy levels (Moore 1949) shows that $2p^4[L_a S_a]nl\}LS$ manifold is indeed governed by the quantum numbers $L_a S_a$ and depends very weakly on LS . This is less so in Ar II. Therefore the present Ar results should be treated as qualitative rather than quantitative. A more accurate calculation using equation (10) is needed to get the Ar β -parameters on the same level of accuracy as for Ne.

In practical computations, we obtain the one-electron dipole matrix elements for the neutral atom by running the RPAE computer code (Amusia and Chernysheva 1997). The random phase approximation with exchange (RPAE) provides a very accurate description of single-photon one-electron ionization processes in noble gas atoms (Amusia and Cherepkov 1975). The β_2 parameters derived from the RPAE dipole matrix elements using equation (8) are in excellent agreement with experimental data of Braune *et al* (2007). For Ne at 47.5 eV, $\beta_2 = 0.98$ as compared with the experimental value of 1.00. For Ar at 38 eV, $\beta_2 = 1.84$ as compared with the experimental value of 1.88.

In principle, the RPAE method can be modified to describe ionization of the singly charged ion with an open np^5 shell (Cherepkov and Chernysheva 1977). This, however, would require an explicit account for anisotropy of the ionized system which we neglect when substituting equation (10) with equation (11). Therefore, we opted for a less computationally demanding Hartree–Fock method when calculating the one-electron dipole matrix elements for the singly charged ion. The radial orbitals $R_{n_0 l_0}$ and R_{kl} entering equation (3) are calculated using the self-consistent and frozen-core Hartree–Fock codes, respectively (Chernysheva *et al* 1976, 1979).

Results of our computations are shown in figure 2 for Ne (left) and Ar (right). In the same figure we indicate the numerical values of the experimental anisotropy parameters

$\beta_2 = 0.74$, $\beta_4 = -0.5$ for Ne at 47.5 eV and $\beta_2 = 1.82$, $\beta_4 = -0.14$ for Ar at 38 eV as reported by Braune *et al* (2007) for the ^3P final state of the doubly charged ion. These results are in fair agreement with the present calculation. However, more experimental data across a wider photon energy range are clearly needed to gauge the quality of the present theoretical model and its numerical implementation.

It is noteworthy that the β_2 parameters depend weakly on the symmetry of the doubly charged ion. Conversely, the β_4 parameters demonstrate a very strong dependence with very small absolute values for ^1D state and relatively large in magnitude and opposite in sign values for ^3P and ^1S states. This behaviour can be understood from equation (11). The statistical average $\sum_{L_a} (2L_a + 1)A_J$ is proportional to δ_{K0} which eliminates the coefficients A_4 and reduces A_0 to a single-photon one-electron value (8). Although the statistical average does not apply directly to the ratio A_J/A_0 , one would expect the term average value of β_4 close to zero which is indeed the case for Ne for which β_2 shows little term dependence. For Ar, β_2 shows strong term dependence near the threshold, and the statistical average of β_4 deviates from zero in this photon energy range.

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