Angular correlation theory for double photoionization in a rare gas atom: ionization by polarized photons

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This is a sequel to an earlier article on the theory of angular correlation for double photoionization. Here we consider the two-step double photoionization of a rare gas atom under the influence of a polarized photon beam described by appropriate Stokes parameters. Cylindrical mirror analyzers (CMA) are used to detect the outgoing electrons. Theoretical values of the correlation function are obtained for linearly polarized light. Two different situations are handled. The statistical theory we obtained good agreement with the experimental results of K"ammerling and Schmidt.

In the present paper we take the incident photon beam to be polarized. The rare gas atoms receiving the photon beam no longer remain randomly oriented, but become aligned. If a photon of adequate energy is absorbed by an atom, a photo-electron is emitted from one of its inner shells, leaving the atom singly ionized. This ion subsequently de-excites by emitting an Auger electron from one of its outer shells. We are left with a doubly ionized atom and two electrons in the continuum. For the purpose of the present study, we have taken the two electrons to be successively emitted electrons, their emissions being adequately separated in time.

Using the Wigner-Eckart theorem, the matrix element of the density operator for the initial atomic state can be expressed as

\[ \langle J_a M_a \alpha_a | \rho | J'_a M'_a \alpha'_a \rangle = \sum_{k_a \kappa_a} (-1)^J_e M'_e C_{M_e M'_e k_a}^{J_a J'_a k_a} \rho_{k_a \kappa_a} (J_a \alpha_a, J'_a \alpha'_a). \]  

Here the statistical tensor \( \rho_{k_a \kappa_a} \) is an irreducible tensor of rank \( k_a \), which transforms according to the \((2k_a + 1)\) dimensional irreducible representation \( D^{k_a} \) of the rotation group. In Eq.\( C_{M_e M'_e k_a}^{J_a J'_a k_a} \) is a Clebsch-Gordan coefficient satisfying the triangle rule \( k_a = J_a + J'_a \) and \( \kappa_a \) is the projection of \( k_a \). Using the unitarity property of Clebsch-Gordan coefficients we get

\[ \sum_{k_a} C_{M_e M'_e k_a}^{J_a J'_a k_a} C_{M_e' M'_e' k_a}^{J_a' J'_a' k_a} = \delta_{M_e M'_e} \delta_{M'_e M'_e'} \delta_{J_e J'_e} \delta_{J'_e J'_e'} \delta_{k_a k_a'}. \]
\[ \rho_{k_\alpha \alpha_a} (J_a \alpha_a, J'_a \alpha_a') = \sum_{M_a M'_a} (-1)^{M_a - M'_a} C_{M'_a M_a \alpha_a}^{J'_a J_a} \langle J_a M_a \alpha_a | \rho | J'_a M'_a \alpha_a' \rangle . \]  

(3)

We assume that the initial state is formed after the randomly oriented rare gas atom absorbs a photon. Then the density matrix of the initial state becomes

\[
\rho_{k_\alpha \alpha_a} (J_a \alpha_a, J'_a \alpha_a') = 3(2J_0 + 1) \sum_{k_0 n_0 k_\gamma} \sqrt{2k_\gamma + 1} C_{k_0 k_\gamma \alpha_a \alpha_a}^{J_a J'_a} \rho \left( \langle J_a \alpha_a | \rho | J'_a \alpha_a' \rangle \right). 
\]

(4)

This equation satisfies the triangle rule \(k_0 = J_0 + J'_0\), where \(J_0\) and \(J'_0\) are the angular momentum quantum numbers of the randomly oriented atom before absorption of the photon and its virtual counterpart respectively. Here \(\rho_{k_0 n_0} (J_0, J_0')\) represents the density matrix of the randomly oriented atom and can be expressed as

\[
\rho_{k_0 n_0} (J_0, J_0') = \frac{1}{\sqrt{2J_0 + 1}} \delta_{k_0 0} \delta_{n_0 0} \langle J_0 || j_1 || J_0 || J_0' || J_0 || j_2 || J_0 || j_2' || J_0' \rangle^*. 
\]

(5)

Here the symbol \(\langle || \rangle\) stands for a reduced matrix element.

In Eq. (4) the expression \(\rho_{k_\alpha \alpha_a} (1, 1, 1)\) represents the density matrix of the photon with its polarization properties. Its elements are

\[
\begin{align*}
\rho_{10}^0 &= \frac{1}{\sqrt{3}} & \rho_{10}^1 &= \frac{2}{\sqrt{3}} & \rho_{10}^2 &= \frac{1}{\sqrt{3}} \\
\rho_{21}^1 &= 0 & \rho_{21}^2 &= 0 & \rho_{21}^3 &= -\frac{1}{2} (S_1 + i S_2).
\end{align*}
\]

(6)

\(S_1, S_2\) and \(S_3\) are the Stokes parameters describing the polarization of the photon. Then Eq. (4) yields

\[
\rho_{k_a n_a} (J_a J'_a) = \frac{3(-1)^{J_a + k_a + 1}}{\sqrt{2J_a + 1}} \sum_{j_1 j_2} \rho_{k_a n_a} (1, 1) \times \langle J_a || j_1 || J_a || J_a' || J_a || j_2 || J_a || j_2' || J_a' \rangle^*. 
\]

(7)

We define the angular correlation function as the expectation value of the efficiency operator. Following the same notation as in reference [1] we can write it as

\[
\bar{\varepsilon} = \sum_{J_a J'_a \alpha_a \alpha_a'} \rho_{k_a n_a} (J_a \alpha_a, J'_a \alpha_a') \varepsilon_{k_a n_a}^* (J_a \alpha_a, J'_a \alpha_a'). 
\]

(8)

Some simplification gives

\[
\bar{\varepsilon} = \sum_{J_a J'_a, k_a, k_a'} \rho_{k_a n_a} (J_a, J'_a) \varepsilon_{k_a n_a} (J_a, J'_a) \varepsilon_{k_a' n_a'} (J_a, J'_a) \times C_{k_a k_a' \gamma}^{J_a J'_a} \times \left( J_a || j_1 || J_a || j_1' || J_a || j_2 || J_a || j_2' || J_a' \right)^*. 
\]

(9)

where the summation extends over \(J_a, J'_a, k_a, k_a', j_1, j_1', j_2, j_2', k_a, k_a', \gamma, k_1, k_2, k\). In DPI experiments the detectors usually used are cylindrical mirror analyzers (CMA) which have cylindrical symmetry with respect to the axis of the detector. Details of the choice of detectors are given in reference [1]. The efficiency tensor component now becomes

\[
\varepsilon_{k_a n_a} (j_1 j_1') = \sum_{\gamma} z_{k_a}(i) c_{k_a \gamma} (j_1 j_1') D_{k_a n_a} (|R_i \rangle). 
\]

(10)

Since the residual doubly ionized state is unobserved, the corresponding quantum numbers are averaged over. This gives

\[
\varepsilon_{k_a n_a} (j_1 j_1') = \sqrt{2J_a + 1} \delta_{k_a 0} \delta_{n_a 0} \delta_{j_1 j_1'}. 
\]

(11)

Then Eq. (8) becomes
\[ \tau = \sum_{n_a} \rho_{n_a} (J_a, J'_a) \sqrt{2 J_a + 1} C_{n_a n_a} C_{0 n_a} \sqrt{2 J_a + 1} \sqrt{2 J_a + 1} \times \left( \begin{array}{ccc} J_c & j_2 & J_0 \\ J_c' & j_2' & J_0' \\ 0 & k_2 & k_3 \end{array} \right) \times \left\{ \begin{array}{ccc} J_0 & j_1 & J_a \\ J_0' & j_1' & J_a' \\ k_3 & k_1 & k_0 \end{array} \right\} \times z_{k_1} (1) c_{k_1} \kappa_{i} (j_1 j_1') z_{k_2} (2) c_{k_2} \kappa_{i} (j_2 j_2') D^{k_1}_{k_{i} n_{i}} (J_1) D^{k_2}_{k_{i} n_{i}} (J_2). \tag{12} \]

Here we have used the relation

\[ D^{k_1}_{k_{i} n_{i}} (R_1) D^{k_2}_{k_{i} n_{i}} (R_2) = \sum_{k} C_{k_{i} k_{i} k_{j} k_{j}} C_{k_{i} k_{i} k_{j} k_{j}} D^{k_{i}}_{k_{i} n_{i}} (R). \tag{13} \]

to get the actual angular dependence of the angular correlation function. In Eq. (13) the Euler rotation \( R = (\beta \gamma \beta \gamma) \). This geometrical dependence of the tensor matrix element is separated out from the dynamics by using the Wigner-Eckart theorem. As a result, the dynamics of the DPI process resides in the reduced matrix elements and the geometrical dependence is contained in the angular part.

We define

\[ \zeta = \sqrt{2 J_c + 1} \sqrt{2 k_2 + 1} \sqrt{2 J_a + 1} \sqrt{2 J_a' + 1} \sqrt{2 J_a'' + 1} \tag{14} \]

and

\[ \xi = \langle J_b \| j_1 \| J_a \rangle \langle J_b \| j_1' \| J_a' \rangle^* \langle J_c \| j_2 \| J_a \rangle \langle J_c \| j_2' \| J_a' \rangle^*. \tag{15} \]

Then the expectation value of the efficiency operator in Eq. (12) becomes

\[ \tau \sim \sum (-1)^{J_a} \zeta \xi \left( \begin{array}{ccc} J_c & j_2 & J_0 \\ J_c' & j_2' & J_0' \\ 0 & k_2 & k_3 \end{array} \right) \times \left( \begin{array}{ccc} J_0 & j_1 & J_a \\ J_0' & j_1' & J_a' \\ k_3 & k_1 & k_0 \end{array} \right) \times C_{k_{i} k_{i} k_{j} k_{j}} (1) z_{k_2} (2) \rho_{k_3} (1, 1) c_{k_1} (j_1 j_1') \times c_{k_2} (j_2 j_2') C_{k_{i} k_{i} k_{j} k_{j}} C_{k_{i} k_{i} k_{j} k_{j}} D^{k_{i}}_{k_{i} n_{i}} (R). \tag{16} \]

A. Attenuation corresponding to polarization sensitivity of a detector

The electron detector may or may not be sensitive to the spin state of the incoming electron. The attenuation of the signal due to the detector will depend on this sensitivity. The factor \( c_{k_{i} \kappa_{i}} (j_{i} j_{i}') \) describes this property. We shall now consider two different cases.

1. Case-I : Detectors insensitive to electron polarization

If the detectors (CMAs) are insensitive to the spin polarization of electrons then the projection \( \kappa_{i} \) of the \( k_{i} \)th component of the angular momentum is effectively zero, i.e. the electrons are emitted symmetrically with respect to the axis of the detector. Hence the attenuation factor can be written as

\[ c_{k_{i} \kappa_{i}} (j_{i} j_{i}') = \frac{\sqrt{2 j_i} + 1 \sqrt{2 j_i'} + 1}{4\pi} (-1)^{j_i - \frac{1}{2} + k_{i}} e^{j_{i} j_{i}' k_{i}} \left( \begin{array}{ccc} \frac{1}{2} & \frac{1}{2} \end{array} \right). \tag{17} \]

2. Case-II : Detectors sensitive to electron polarization

In reference [8] we defined \( c_{k_{i} \kappa_{i}} (j_{i} j_{i}') \) as the attenuation factor due to the change in the state of polarization of an electron caused by the detector. When the detectors are insensitive to electron polarization, one takes the average over the electron spin and its projection. Now consider the case where the detectors are sensitive to electron polarization. In this case the spin sensitivity of the detectors is described by a tensor of the form \( c_{k_{i} \kappa_{i}} (s_i, s_i) \). The attenuation factor then turns out to be

\[ c_{k_{i} \kappa_{i}} (j_{i} j_{i}') = c_{k_{i} \kappa_{i}} (l_{i} l_{i}') c_{k_{i} \kappa_{i}} (s_i, s_i) \sqrt{2 k_{i} + 1} \sqrt{2 k_{i} + 1} \times \left( \begin{array}{ccc} l_i & l_i' & k_{i} \\ s_i & s_i & k_{i} \end{array} \right) C_{k_{i} k_{i} k_{i}} \left( \begin{array}{ccc} \frac{1}{2} j_i \frac{1}{2} j_i' \end{array} \right). \tag{18} \]
where
\[ c_{k_10}(l,l') = \frac{\sqrt{2l + 1}\sqrt{2l' + 1}}{4\pi} (-)^{l'} C_{00}^{l'l} k_1, \tag{19} \]

and \( c_{k_1\kappa,s_i}(s_i,s_i) \) can be expressed in terms of the Stokes parameters describing the spin polarization of the electron to be detected\[[3]\]. The factor \( c_{k_1\kappa,s_i}(s_i,s_i) \) picks out electrons with a particular spin projection and may be called a Stern-Gerlach operator. Its components are
\[ c_{00} = \frac{-\sqrt{2}}{2}, \quad c_{10} = \frac{S_z^*}{2}, \quad c_{11} = -(S_y^* - iS_x^*), \quad c_{1-1} = -(S_y^* + iS_x^*). \tag{20} \]

Here \( S_y^*, S_x^* \) and \( S_z^* \) are Stokes parameters describing the polarization of the electron. For polarization insensitive detectors one has \( S_y^* = S_x^* = S_z^* = 0 \), and the attenuation factor reduces to Eq.(13).

The lifetime of the singly ionized state is very small. Depending on the photon energy there may be a situation where it is impossible to differentiate between the photo- and Auger electrons simply by energy analysis. Then, to distinguish between the two electrons it is necessary to measure the electron spin, i.e. their polarization. For spin analysis of the electrons we have to use a Stern-Gerlach type experimental set-up. Here the factor \( c_{k_1\kappa,s_i}(s_i,s_i) \) serves exactly that purpose, i.e. picks out electrons with a particular spin projection. This type of experiment is known as 'energy- and angle-resolved coincidence experiment' and is being done by Schmidt and his co-workers\[[3]\].

In general, for DPI of atoms using polarized photon of sufficient energy, one can distinguish the photo- and the Auger electrons by differential energy analysis. In that case determination of electron spin is meaningless. Then, if the spin is unobserved, one can take the average over the spin projection. In that case the projection \( \kappa_i \) of the \( k_i \)th component of the angular momentum is zero and the attenuation factor \( c_{k_i\kappa_i}(j_1j_2') \) turns out to be \( c_{k_i0}(j_1j_2') \).

III. CALCULATION AND RESULTS

In reference\[[1]\] we treated DPI in the xenon atom due to unpolarized light. In this paper we are concerned with the same xenon atom with the difference that DPI occurs due to a polarized light source. A randomly oriented xenon atom is irradiated with a polarized photon beam of energy 94.5 eV. As a result, the xenon atom no longer remains randomly oriented but acquires the polarization of the photon beam. This leads to photoionization in the \( 4d_{5/2} \) shell followed by a subsequent \( N_0 → O_{23}O_{23}^{-} S_0 \) Auger decay. We use the dipole approximation, the letters \( e, f \) and \( q \) for the three possible photoionization channels\[[1]\]. These are characterized by \( e) 4d_{5/2} → ε_p f_{7/2}, f) 4d_{5/2} → ε_p f_{5/2} \) and \( g) 4d_{5/2} → ε_p p_{3/2} \) respectively. And the Auger transition is characterized by the wave \( ε_A 4d_{5/2} \). The same selection rules for photoionization and Auger transitions hold good as in the case of unpolarized light.

In experiments for measuring angular correlation one usually chooses detectors which are insensitive to the spin polarization of electrons. In such a case \( \kappa_1 = \kappa_2 = \kappa'_1 = \kappa'_2 = \kappa = \kappa' = 0 \), and \( D_{00}^0(\beta_1\beta_2) = P_k(\cos \theta) \). Then Eq.(14) becomes
\[ \tau \sim \sum (-1)^{J_k} \zeta_k \left\{ \begin{array}{ccc} J_e & J_f & J_o \\ j_1 & j_2 & J_o \\ 0 & k_2 & k_2 \end{array} \right\} z_{k_1}(1)z_{k_2}(2)P_k(1,1) \]
\[ \times C_{k_2k_1k}^{k_2k_1k} c_{k_10}(j_1j'_2)c_{k_20}(j_2j'_2)P_k(\cos \theta). \tag{21} \]

The summation extends over \( k_1, k_2 \) and \( k \).

In the limiting case of unpolarized photons Eq.(21) reduces to a simple form. Using Eq. (27) and some properties of \( 9-j \) symbols and Racah coefficients\[[1]\], we get
\[ \tau = \sum_k z_k(1)z_k(2)(-1)^{j_1+j_2} c_{k0}(j_1j'_2)c_{k0}(j_2j'_2) \]
\[ \times \langle J_e \mid j_1 \parallel J_o \rangle \langle J_f \mid j_2 \parallel J_o \rangle^* \langle J_e \mid j_2 \parallel J_o \rangle \langle J_f \mid J_o \parallel J_o \rangle^* \]
\[ \times w(J_e,J_f;j_1j'_2;J_o,k_{J_o})w(J_e,J_f;j_2j'_2;J_o,k_{J_o})P_k(\cos \theta). \tag{22} \]

Note that this is identical with Eq.(25) of reference\[[1]\], as it should be.

Experiments on the xenon atom were carried out by Schmidt and his co-workers using 94.5 eV synchrotron radiation\[[3]\]. They used a perpendicular plane geometry to describe the process. The collision frame \( x, y, z \) is attached to the target where the \( z \) axis coincides with the direction of the photon beam. The arbitrary polarization of the incident beam from the synchrotron is described by the Stokes parameters \( S_1, S_2 \) and \( S_3 \). Both \( S_1 \) and \( S_2 \) refer to the same quantity, but with differently oriented axes. One can make \( S_2 = 0 \) by choosing the \( x \) axis of the collision frame to coincide with the direction of maximum linear polarization, i.e. the major axis of the polarization ellipse. To compare our results with experimental values we use the same polar and azimuthal angles in the perpendicular plane geometry. Fig.1 shows the perpendicular plane geometry described above, \( e_1 \) and \( e_2 \) being the directions of emission of the photo- and the Auger electron respectively. \( \theta \) is the angle between their directions of emission. We have calculated the theoretical value of the angular correlation function for two different cases.
(i) The photo-electron is observed in a fixed direction and the Auger electron spectrometer is turned around to get the angular distribution of the Auger electrons with respect to the photo-electron. Here the maximum allowed value of $k$ is $2j_2$.

(ii) The second one is the complementary case, i.e. the Auger electron is observed in a fixed direction and the photo-electron spectrometer is turned around to get the angular distribution of the photo-electrons with respect to the Auger electron. Here the maximum allowed value of $k$ is $2j_{1\text{,max}}$, $j_{1\text{,max}}$ is the maximum value of $j_1$ for the possible photoionization channels.

The value of $k$ gives the highest order of the Legendre polynomials occurring in the correlation function. Interchannel interaction of the different photo-electron channels contributes to the angular correlation pattern by introducing the different terms, however, the total intensity remain unchanged. This inter channel interaction is treated as it was in reference [1].

As in reference [2] we have defined the angular correlation function to be the angular part of the expectation value of the efficiency operator. Solid lines represents the theoretically calculated plot and the dots represents the experimental plot [3]. For a linearly polarized incident photon beam the angular correlation function for our case turns out to be

\begin{equation}
W(\theta) \sim 1 + 1.314P_2(\cos \theta) + 1.100P_4(\cos \theta). 
\end{equation}

\[ \text{eq:23} \]

\begin{equation}
\text{i) Case 1: } S_1 = 1, S_2 = 0, S_3 \text{ unknown. The photo-electron is observed in a fixed direction and the Auger electron spectrometer is turned around to get the angular distribution of the Auger electron with respect to the photo-electron.}
\end{equation}

\begin{equation}
W(\theta) \sim 1 + 0.817P_2(\cos \theta) + 0.602P_4(\cos \theta) + 0.570P_6(\cos \theta). 
\end{equation}

\[ \text{eq:24} \]

\begin{equation}
\text{ii) Case 2: } S_1 = 1, S_2 = 0, S_3 \text{ unknown. The Auger electron is observed in a fixed direction and the photo-electron spectrometer is turned around to get the angular distribution of the photo-electron with respect to the Auger electron.}
\end{equation}

\begin{equation}
W(\theta) \sim 1 + 1.314P_2(\cos \theta) + 1.100P_4(\cos \theta) .
\end{equation}

In both the cases one of the electron spectrometers is kept fixed along the direction of the electric field vector (x-axis). The index $k$ in the general theoretical expression for $\mathbf{\mathcal{T}}$ depends on the angular momenta of the emitted electrons. Hence, the structure of the angular correlation pattern depends on this index. If higher order angular momenta are involved the angular correlation pattern has more structure. This is clear from the figures 2 and 3. Since the distribution of the photo-electron with respect to the fixed Auger electron direction involves higher order angular momenta, the angular correlation patterns has more structure.

Figure 1: Perpendicular plane geometry used in the experiments by Schmidt and his co-workers
Figure 2. Angular correlation pattern for xenon due to a linearly polarized photon beam ($S_1 = 1, S_2 = 0, S_3$ unknown) of 94.5 eV ($4d_{5/2}$ photoionization followed by $N_5 - O_{23}O_{23}^1 S_0$ Auger decay). The photo-electron is observed in a fixed direction[3].

Figure 3. Angular correlation pattern for xenon due to a linearly polarized photon beam ($S_1 = 1, S_2 = 0, S_3$ unknown) of 94.5 eV ($4d_{5/2}$ photoionization followed by $N_5 - O_{23}O_{23}^1 S_0$ Auger decay). The Auger electron is observed in a fixed direction[3].

Acknowledgment: One of the author (CS) would like to acknowledge the financial support provided by the University Grants Commission of India.

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