Theory of angular correlation for double photoionization in a rare gas atom due to linearly polarized light

Chiranjib Sur and Dipankar Chattarji
Department of Physics, Visva-Bharati, Santiniketan 731 235, INDIA

In this paper we have tried to establish a theory for angular correlation between the photo- and Auger electrons in the two-step double photoionization of a rare gas atom. The basic finding of this paper is that a general theoretical expression for the angular correlation function is established in the problem of double photoionization of atoms in the presence of linearly polarized monochromatic light. We consider the case of a xenon atom and the incident light is of 94.5 eV with arbitrary linear polarization. The theoretical results are compared with the experiments done by Kämmerling and Schmidt [J.Phys.B, 26, 1141(1993)].

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In a recent paper we gave a theory for angular correlation in the double photoionization (DPI) of a rare gas atom [1]. We took DPI to be a two step process [2]. The atom is irradiated by a beam of unpolarized photons and emits a photo-electron. This is the first step. After a finite time interval the singly ionized atom de-excites by emitting an Auger electron. The time delay between the two steps decouples them energetically. However, the angular momenta of the photo-electron and the Auger electron interact to yield their final angular correlation. We defined the angular correlation $W$ as a function of $\theta$, the angle between the directions of emission of the two electrons.

The object of the present paper is to obtain a general expression for the angular correlation function for DPI of a rare gas atom due to a linearly polarized photon beam described by Stokes parameters $S_1$, $S_2$ and $S_3$ [3]. The atom is initially randomly oriented, but becomes aligned after the absorption of a photon. It is also energetically excited, and emits a photo-electron from one of its inner shells. The photo-electron leaves the interaction region in $\sim 10^{-18}$ sec. The singly ionized atom with an inner shell vacancy will now tend to de-excite by emitting an Auger electron from one of the outer shells [4]. A typical Auger lifetime of say the xenon ($Z = 54$) O-shell is $\sim 10^{-15}$ sec. The photo- and Auger ionizations are thus widely separated in time.

We now come to the basic problem of calculating the angular correlation between the photo-electron and the Auger electron. In a natural way, we can define the angular correlation function $W(\theta)$ to be the probability that the angle between their directions of emission is $\theta$. On what factors should it depend, and how can we formulate a general approach to its investigation? The following points may be noted.

(i) Obviously, $W(\theta)$ should be independent of the choice of coordinates, because basically it is a function of the angle $\theta$. This calls for a tensor formalism.

(ii) Since $W$ has the nature of a probability, we can only evaluate it by counting the number of coincident events in which the angle between the directions of emission has a particular value $\theta$, and so on. Thus the question arises of a set of detectors which can concurrently detect the photo-electron and the Auger electron. In other words, we have to define a detection operator to describe such operations quantum mechanically.

(iii) The detectors may or may not be completely dependable in actual practice. This raises the question of efficiency of a particular detector.

(iv) Thus we can think of two kinds of quantum mechanical operators to describe our problem: (a) a density operator $\rho$ which describes the probability of occurrence of an electron in a particular state, and (b) an efficiency operator $\varepsilon$ which describes the probability that our detector will actually detect the electron in that state.

We can thus visualize the angular correlation function as a scaler which is simply related to the product of the two tensor operators $\rho$ and $\varepsilon$ as defined above. Evidently a contraction of these tensor operators will be called for to give the scalar $W(\theta)$. One virtue of this theory is that we are invoking the statistical properties of a complete ensemble and no perturbation series of any kind is involved.

Let us now look a little more carefully at the angular correlation function $W(\theta)$. When we say that it is the probability that the angle between the directions of emission of the two electrons is $\theta$, we actually mean two things. (i) The two electrons are in states which are compatible with their directions of emission making an angle $\theta$ with each other, and (ii) the efficiencies of the two electron detectors are such that the angle between the directions of emission may be inferred to be $\theta$. Taking these two factors into account we define

$$\tau = \sum_{J_aJ'_a\alpha_a\alpha'_a} \rho_{\alpha_a\alpha'_a} (J_a\alpha_a, J'_a\alpha'_a) \varepsilon_{\alpha_a\alpha'_a} (J_a\alpha_a, J'_a\alpha'_a),$$  \hfill (1)
$W(\theta)$ being the angular part of $\tau$.

As before [4], we denote the initial state (photon+atom) by the set of quantum numbers $(J_a M_a \alpha_a)$, or by virtual quantum numbers $(J_a' M_a' \alpha_a')$. $(J_a, M_a)$ or $(J_a', M_a')$ are angular momentum quantum numbers, and $\alpha_a, \alpha_a'$ stand for the set of remaining quantum numbers. Similarly for the intermediate and final states.

In DPI experiments, a Cylindrical Mirror Analyser (CMA) is used for detection and energy analysis of the electrons. It imparts to the electron a cylindrical symmetry regardless of its angle of incidence with respect to the axis.

As for the orbital angular momentum of the incident photo- or Auger electron, this is fixed by arranging to receive only those incident electrons which have the characteristic energy. This implies that the detector is receiving all the photo- or Auger electrons, as the case may be, having the characteristic energy and orbital angular momentum. In other words, the CMA is acting not only as a differential energy analyser, but also as an angle-integrated device. For a given orbital angular momentum which labels a photo- or Auger electron, the conjugate angle of incidence becomes completely indeterminate. It is this indeterminacy which gives a statistical character to the distribution of the two electrons. That in its turn manifests itself in their angular correlation.

We decompose the density matrix $\rho$ of the photo-absorbed rare gas atom in terms of the density matrix of the unobserved doubly ionized state and of the two continuum electrons. The choice of detectors is discussed in detail in reference [4]. The expression for the efficiency tensor component is

$$
\varepsilon_{\alpha' \alpha}^f (j_1 j'_1) = \sum_{\kappa} z_\kappa (i) c_{\kappa \alpha} (j_1 j'_1) D_{\kappa \alpha}^{\dagger} (\mathbf{R}_1). \tag{2}
$$

Since the residual doubly ionized state is unobserved we can write

$$
\varepsilon_{\alpha' \alpha}^f (j_1 j'_1) = \sqrt{2 J_a + 1} \delta_{\kappa_\alpha 0} \delta_{\alpha' \alpha} \delta_{j_1 j'_1}. \tag{3}
$$

Some simplification yields the result

$$
\tau = \sum_{\kappa} \rho_{\alpha' \alpha} (J_a, J_a') \sqrt{2 J_a + 1} C_{\kappa_\alpha 0}^{\dagger} C_{\kappa_\alpha 0} \sqrt{2 J_a + 1} \frac{1}{2 J_a + 1} \frac{1}{2 J_a + 1} \frac{1}{2 J_a + 1} \frac{1}{2 J_a + 1} \frac{1}{2 J_a + 1} \frac{1}{2 J_a + 1}
\times \sum_{\kappa} z_\kappa (i) c_{\kappa \alpha} (j_1 j'_1) c_{\kappa' \alpha'} (j_2 j'_2) D_{\kappa \alpha}^{\dagger} (\mathbf{R}_1) D_{\kappa' \alpha'}^{\dagger} (\mathbf{R}_2). \tag{4}
$$

In experiments for measuring the angular correlation one usually chooses detectors insensitive to the spin polarization of electrons. Then Eq.(4) turns out to be

$$
\bar{\varepsilon}(\theta) \sim \sum (-1)^{j_1} \zeta \left\{ \frac{J_a}{j_1} \frac{j_2}{j_b} \frac{J_a}{j_1} \frac{j_2}{j_b} \frac{J_a}{j_1} \frac{j_2}{j_b} \right\} \sum_{\kappa} z_\kappa (1) z_{\kappa_\alpha} (2) \rho_{\alpha' \alpha} (1, 1)
\times C_{\kappa_\alpha 0}^{\dagger} C_{\kappa_\alpha 0} \left( j_1 j'_1 \right) c_{\kappa_\alpha 0} (j_2 j'_2) P_\alpha (\cos \theta), \tag{5}
$$

where $\rho_{\alpha' \alpha} (1, 1)$ is the density matrix describing the photon, and

$$
\zeta = \sqrt{2 J_a + 1} \sqrt{2 J_a + 1} \sqrt{2 J_a + 1} \sqrt{2 J_a + 1} \sqrt{2 J_a + 1}
\times \sum_{\kappa} z_\kappa (1) z_{\kappa_\alpha} (2) \rho_{\alpha' \alpha} (1, 1)
\times C_{\kappa_\alpha 0}^{\dagger} C_{\kappa_\alpha 0} \left( j_1 j'_1 \right) c_{\kappa_\alpha 0} (j_2 j'_2) P_\alpha (\cos \theta), \tag{6}
$$

and

$$
\xi = \langle J_b \parallel j_1 \parallel J_a \rangle \langle J_b \parallel j_1 \parallel J_a \rangle^* \langle J_b \parallel j_2 \parallel J_a \rangle \langle J_b \parallel j_2 \parallel J_a \rangle^*. \tag{7}
$$

Now consider a randomly oriented $^1S_0$ xenon atom which is irradiated by a 94.5 eV photon beam of arbitrary linear polarization. This leads to photoionization in the $4d_{5/2}$ shell followed by a subsequent $N_5 - O_{23}O_{23}^+$ $^1S_0$ Auger decay [4]. We use the dipole approximation, and the letters $e, f$ and $g$ for the three possible photoionization channels: $e) 4d_{5/2} \rightarrow \varepsilon_e f_{7/2}$, $f) 4d_{5/2} \rightarrow \varepsilon_f f_{5/2}$ and $g) 4d_{5/2} \rightarrow \varepsilon_g f_{5/2}$ respectively. The Auger transition is characterised by the partial wave $\varepsilon_A d_{5/2}$. The same selection rules for photoionization and Auger transitions hold good as in reference [4]. Since the photoionization process is described by three possible channels, Eq.(4) will be modified by the addition of interference terms. The total intensity will, however, remain unchanged, as we have discussed in reference [4]. Then the expectation value of the efficiency operator turns out to be

$$
\bar{\varepsilon}(\theta) = \bar{\varepsilon}_e(\theta) + \bar{\varepsilon}_f(\theta) + \bar{\varepsilon}_g(\theta) + \bar{\varepsilon}_ef(\theta) + \bar{\varepsilon}_fg(\theta) + \bar{\varepsilon}_eg(\theta). \tag{8}
$$

Here
Here the maximum allowed value of electron spectrometer is turned around to get the angular distribution of the Auger electron with respect to the photo-electron.

\[ \varepsilon_e(\theta) \sim \sum z_{\ell_1} (1) z_{\ell_2} (2) \rho_{\ell_2,\ell_2}^2 (1,1) \begin{cases} J_e & J_2 & J_b \\ J_e & J_2 & J_b \\ 0 & k_2 & k_b \end{cases} \]

\[ \times \begin{pmatrix} J_b & J_1 & J_a \\ J_b & J_1 & J_a \end{pmatrix} C_{k_2,k_1,k_a}^{0,k_2,k_b} \]

\[ \times |(J_b || J_a|^2)^0 (|J_c || J_2|^2, k_2, k_1, k_0) c_{k_1,0}(J_j^2 J_j^1) P_k(\cos \theta), \]

and the summation extends over \( k_a, k_1, k_2 \) and \( k \). The expectation values \( \varepsilon_e(\theta) \) and \( \varepsilon_p(\theta) \) have the same form with \( \{ j_1^e \rightarrow j_1^1, k_1^e \rightarrow k_2^1 \} \) and \( \{ j_1^i \rightarrow j_1^f, k_1^i \rightarrow k_1^f \} \) respectively. The quantity \( \varepsilon_{ij}(\theta) \) is an interference term arising from interaction between different photoionization channels \( i \) and \( j \) (\( i, j = e, f, g \) with \( i \neq j \)).

\[ \varepsilon_{ij}(\theta) \sim \sum z_{\ell_1} (1) z_{\ell_2} (2) \rho_{\ell_2,\ell_2}^2 (1,1) \begin{cases} J_e & J_2 & J_b \\ J_e & J_2 & J_b \\ 0 & k_2 & k_b \end{cases} \]

\[ \times \begin{pmatrix} J_b & J_1 & J_a \\ J_b & J_1 & J_a \end{pmatrix} C_{k_2,k_1,k_a}^{0,k_2,k_b} \]

\[ \times |(J_b || J_a|^2)^0 (|J_c || J_2|^2, k_2, k_1, k_0) c_{k_1,0}(J_j^2 J_j^1) \times c_{k_2,0}(j_2 j_2) P_k(\cos \theta) \]

The coefficient \( k \) is even and satisfies the triangle rule \( k = k_1 + k_2 \).

We have calculated the theoretical values of the angular correlation function for two possible cases. The electric field vector is along the \( x \) axis and a perpendicular plane geometry is used to describe the process. We choose the collision frame \( x, y, z \) attached to the sample and the \( z \) axis coincides with the direction of incident the photon beam. Since the Stokes parameters \( S_1 \) and \( S_2 \) refer to the same quantity, but with differently oriented axes, one can take \( S_2 = 0 \). This is done by selecting 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. The same polar and azimuthal angles are chosen in the experiment. Here \( c_1 \) and \( c_2 \) are the directions of propagation of the photo-electron and the Auger electron respectively. (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 electron with respect to the photo-electron. Here the maximum allowed value of \( k \) is \( 2j_2 \). This is our first case. (ii) The second case 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-electron with respect to the Auger electron. Here the maximum allowed value of \( k \) is \( 2j_1,\text{max} \). This value gives the highest order of the Legendre polynomials occurring in the correlation function.

The angular correlation function in DPI due to polarized photons now becomes

i) Case 1 : 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,

\[ W(\theta) \sim 1 + 1.314 P_2(\cos \theta) + 1.100 P_4(\cos \theta) \]

ii) Case 2 : 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,

\[ W(\theta) \sim 1 + 0.817 P_2(\cos \theta) + 0.602 P_4(\cos \theta) + 0.570 P_6(\cos \theta) \]

If the incident photon is unpolarized Eq.11 reduces to Eq.(25) of reference 4.

The solid line in the figures represents the theoretical curve and the dotted points are experimental values of \( W(\theta) \). Fig 1 gives the polar plot of the angular correlation function for unpolarized photons. Figures 2 and 3 for linearly polarized photons.
Fig. 1: Polar plot of angular correlation function $W(\theta)$ for 2-step double photoionization of xenon due to unpolarized photon beam of 94.5 eV ($4d_{5/2}$ photoionization followed by $N_5 - O_{2,3}O_{2,3}^1S_0$ Auger decay). The photo-electron is observed in a fixed direction.

Fig. 2: Angular correlation pattern for 2-step double photoionization of xenon due to a linearly polarized photon beam of 94.5 eV ($4d_{5/2}$ photoionization followed by $N_5 - O_{2,3}O_{2,3}^1S_0$ Auger decay). The photo-electron is observed in a fixed direction.
Fig.3: Angular correlation pattern for 2-step double photoionization of xenon due to a linearly polarized photon beam of 94.5 eV (4d5/2 photoionization followed by N5 = O2,3O2,31S0 Auger decay). The Auger electron is observed in a fixed direction.

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[1] D. Chattarji and C. Sur, Phys. Rev. A 65, 012702 (2001).
[2] D. Chattarji and C. Sur, J. El. Spec. and Rel. Ph. 114-116, 153(2001).
[3] Max Born and Emil Wolf, Principles of Optics, 30 (Pergamon Press, Oxford, 1975).
[4] D. Chattarji, The Theory of Auger Transitions (Academic Press, London, 1976).
[5] K. Blum, Density Matrix Theory and Applications (Plenum Press, New York, 1981).
[6] D. Chattarji and C. Sur, [physics/0108075]
[7] B. Kümmerling and V. Schmidt, J.Phys.B 26, 1141(1993).