Spin polarization in the double photoionization of atoms

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In this paper we develop a theoretical framework for studying spin polarization of one or both of the two electrons emitted in the one-photon, one-step, double ionization of an atom. The expressions derived herein are in the form of an incoherent sum over the angular momentum j_t exchanged between the unobserved initial and final angular momenta. It then naturally facilitates an analysis of spin-resolved double-photoionization processes in terms of the parity-favored and -unfavored transitions. Several photon-propagation and electron-detection configurations are considered in which it becomes simpler to study the angular-spin or spin-spin correlation between two photoelectrons. The approach suggested in this paper has been used to analyze spin-resolved double photoionization in the $6s^2$ subshell of atomic ytterbium. [S1050-2947(97)04508-3]

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I. INTRODUCTION

Since the pioneering work of Byron and Jochain [1], there have been several experimental as well as theoretical studies of double photoionization (DPI) in atomic targets (see, for example, Refs. [2–15]). In this process, either absorption [1–10] of a single photon in an electric dipole ($E1$) approximation or single-photon Compton scattering [11–15] simultaneously ejects two electrons from an atom. In most of these papers [1–15], usually integrated cross-section and/or angular distributions of the two photoelectrons have been studied.

The two photoelectrons, moving in a continuum, should not only share between themselves photon energy in excess of the ionization potential and influence each other's angular distribution through mutual Coulombic repulsion but affect, in principle, also the orientations of their spins. This means that the role played by the spin-orbit interaction (SOI) in DPI processes should also be investigated. Moreover, in order to be able to extract complete [16] information from any experimental data on DPI, it is necessary that the initial state of the reactants (here an atom plus the radiation field) should be state selected and the internal properties of the reaction products (a doubly charged atomic photoion and the two photoelectrons, in the present case) should be analyzed.

Therefore, if one wants to properly study electron-electron correlations in DPI, it is necessary to analyze, in addition to their energy, both the angular distribution as well as spin polarization of the two photoelectrons. Such investigations will also constitute one step towards performing a 'complete' experiment on DPI.

However, because of a substantial loss [approximately by a factor of about 1000 (Ref. [17])] of intensity in a Mott detector used to detect the direction of a photoelectron's spin, measurements on spin polarization are extremely difficult to perform and are probably beyond the reach of currently available experimental facilities for DPI. However, the probable nonfeasibility of angle- and spin-resolved DPI measurements in the near future should not deter one from studying such processes at least theoretically, even at present. This observation becomes particularly significant in view of the fact that the correlation between the spins of a pair of electrons moving in a continuum has been analyzed already in another kind of experiment on double ionization.

For example, several recent investigations have shown that the fragmentation patterns characterized by the energy partitioning, angular distribution, and spin polarization of photoelectrons and of Auger electrons ejected sequentially in the one-photon, two-step, double ionization of atoms [18–20] and of molecules [20–22] are much more complicated than those in which spins of the outgoing electrons are not detected. Such angle- and spin-resolved studies are much richer sources of information [18–22] on the dynamics of double ionization as well as on electron-electron correlations.

In this paper we therefore present a study of atomic DPI including an analysis of the spin polarization of both photoelectrons. In Sec. II an angle- and spin-correlation function for DPI has been derived by taking into account the SOI in the initial electronic state of the atom, say, A, in the electronic state of the doubly charged residual photoion A^{2+}, and in the two continua of both photoelectrons. In order to obtain a simplest possible expression for this complicated function, we have adapted the angular-momentum transfer method to DPI. This method was originally proposed by Fano and Dill [23,24] for the purely angle-resolved, one-photon, single-ionization of atoms and molecules, and later generalized by us [25–27] to those experiments in which the spin polarization of the single outgoing photoelectron is analyzed as well. The present adaptation of the angular-momentum transfer method to angle- and spin-resolved DPI in atoms helps in an analysis of this process in terms of parity-favored and -unfavored transitions [24], already carried out by us [27] for single-photoelectron spectroscopy.

In Sec. II several photon-propagation and electron-detection configurations also have been investigated for which the angular- and spin-correlation functions for DPI take particularly simpler forms. Therein, we suggest also various simple alternative experiments involving the detection of the direction and/or spin of one or both of the two photoelectrons.

The theory developed in this paper is used in Sec. III to study the spin polarization of electrons ejected from the $6s^2$ subshell of atomic Yb in its DPI. Therein we show how the SOI in the continuum of one photoelectron affects not only its own properties but also those of its companion photoelectron. It happens even if only one of the two ejected photo-

electrons is observed. Finally, Section IV contains the conclusions of this paper.

II. ANGULAR AND SPIN CORRELATIONS BETWEEN ELECTRONS EJECTED IN DPI

The process of interest to us in this paper can schematically be represented by

$$h\nu_r(|\vec{l}_r|=1)+A(J_0M_0)\rightarrow A^{2+}(J_fM_f)+e_1(\vec{k}_1;\mu_1\hat{u}_1)+e_2(\vec{k}_2;\mu_2\hat{u}_2). \quad (1)$$

Here $E_r=h\nu_r$ and $|\vec{l}_r|=1$ are, respectively, the energy and angular momentum of the photon absorbed by atom A in the $E1$ approximation. $|J_0M_0\rangle$ and $|J_fM_f\rangle$ are the bound electronic states of A with energy E_0 and of the residual doubly charged photoion A^{2+} possessing energy E_f, respectively. M_0 and M_f are the respective projections along the polar axis of the space (or photon) frame of reference of the total angular momentum $\vec{J}_0$ of A and $\vec{J}_f$ of A^{2+}. The quantization axis is in the direction of the electric vector of the linearly polarized (LP) radiation present in Eq. (1). If the ionizing radiation happens to be circularly polarized (CP) or unpolarized (UP), its direction of incidence then defines the polar axis of the photon frame. Also in Eq. (1), $\vec{k}_1[k_1,\hat{k}_1(\theta_1,\phi_1)]$ and $\vec{k}_2[k_2,\hat{k}_2(\theta_2,\phi_2)]$ are the propagation vectors of two photoelectrons ejected simultaneously with respective energies $\varepsilon_1=\hbar^2k_1^2/2m$ and $\varepsilon_2=\hbar^2k_2^2/2m$. We therefore have $\varepsilon_1+\varepsilon_2=h\nu_r-(E_f-E_0)$ from energy conservation. Projections $\mu_1\hbar$ and $\mu_2\hbar$ (with $\mu_1,\mu_2=\pm\frac{1}{2}$) of the spin angular momenta of e_1 and e_2 ejected in DPI are taken to be in the directions $\hat{u}_1(\theta_1',\phi_1')$ and $\hat{u}_2(\theta_2',\phi_2')$, respectively.

Following the procedure explained in Ref. [26] for angle- and spin-resolved photoelectron spectroscopy of the single ionization of atoms, the required final state describing the $A^{2+}+e_1+e_2$ system produced in the DPI in Eq. (1) can be written as

$$\begin{aligned}\Psi^-_{f;\vec{k}_1\mu_1;\vec{k}_2\mu_2}=&\sum_{\substack{l_1,j_1,m_{l_1},m_{j_1},\nu_1,j\\ l_2,j_2,m_{l_2},m_{j_2},\nu_2,m_j}} i^{(l_1+l_2)}e^{-i(\sigma_{l_1j_1}+\sigma_{l_2j_2})}\\ &\times(-1)^{1-l_1-l_2+j_2-j_1-m_{j_1}-m_{j_2}-m_j}\\ &\times\sqrt{(2j_1+1)(2j_2+1)(2j+1)}\\ &\times\begin{pmatrix} l_1 & \frac{1}{2} & j_1\\ m_{l_1} & \nu_1 & -m_{j_1}\end{pmatrix}\begin{pmatrix} l_2 & \frac{1}{2} & j_2\\ m_{l_2} & \nu_2 & -m_{j_2}\end{pmatrix}\\ &\times\begin{pmatrix} j_1 & j_2 & j\\ m_{j_1} & m_{j_2} & -m_j\end{pmatrix}Y^{m_{l_1}*}_{l_1}(\hat{k}_1)Y^{m_{l_2}*}_{l_2}(\hat{k}_2)\\ &\times D^{1/2*}_{\mu_1\nu_1}(\omega_{u_1})D^{1/2*}_{\mu_2\nu_2}(\omega_{u_2})\Phi^-_{f;jm_j}. \quad (2)\end{aligned}$$

This expression is obtained by including the SOI in the $|J_fM_f\rangle$ state of A^{2+} and also in the continuum of e_1 and e_2 specified by the total angular momenta

$$\vec{j}_1=\vec{l}_1+\frac{\vec{1}}{2} \quad (3a)$$

and

$$\vec{j}_2=\vec{l}_2+\frac{\vec{1}}{2}, \quad (3b)$$

respectively. The channel function $\Phi^-_{f;jm_j}$ in Eq. (2) represents the doubly charged photoion A^{2+} in its fth state and the two photoelectrons with total angular momentum

$$\vec{j}=\vec{j}_1+\vec{j}_2 \quad (3c)$$

having its projection m_j along the polar axis of the space frame. The minus superscript on a function such as Φ and Ψ in Eq. (2) indicates that it asymptotically satisfies the incoming wave boundary conditions appropriate [28] for photoionization. Also in Eq. (2), $\sigma_{l_1j_1}$ and $\sigma_{l_2j_2}$ are the Coulomb phases [29] for the l_1th and l_2th partial waves of the photoelectrons e_1 and e_2, respectively; the D's are the well-known rotational harmonics with ω_{u_1} $(\equiv\phi_1',\theta_1',0)$ and ω_{u_2} $(\equiv\phi_2',\theta_2',0)$ the Euler angles for spin-polarization directions $\hat{u}_1$ and $\hat{u}_2$, respectively [30].

Let $|1m_r\rangle$ represent the state of the absorbed photon in the $E1$ approximation with $m_r=-1$, 0, and 1 for left circular polarization, linearly polarized and right circular polarization, respectively. Then the transition amplitude for the DPI process (1) is the matrix element $\langle\Psi^-_{f;\vec{k}_1\mu_1;\vec{k}_2\mu_2}|F|\Phi_0;1m_r\rangle$ of the $E1$ operator F defined in Ref. [29] in both length and velocity approximations. Here $\Phi_0\equiv|J_0M_0\rangle$ is the bound electronic state of atom A taking the SOI into account.

The angular distribution of spin-resolved photoelectrons emitted in process (1) is given by

$$\frac{d^3\sigma(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}=\frac{K}{2J_0+1}\sum_{M_0,M_f}\langle\Psi^-_{f;\vec{k}_1\mu_1;\vec{k}_2\mu_2}|F|\Phi_0;1m_r\rangle^2, \quad (4)$$

where $K=3\Pi(e^2/\alpha_0E_r)^2$ with α_0 the dimensionless fine-structure constant. The definitions of the operator F in the $E1$ length and velocity approximations given in Eqs. (7) and (8) in Ref. [29], respectively, along with the value of the constant K in Eq. (4) herein, are in conformity with Bethe and Salpeter [31] such that Eq. (4), after integration over $\hat{k}_1$ and $\hat{k}_2$, gives the cross section for DPI in cm^2. Expression (4) has been summed over M_f and averaged over M_0.

Now we substitute state (2) into Eq. (4) and expand the photoionization matrix element $\langle\Phi^-_{f;jm_j}|F|\Phi_0;1m_r\rangle$ present in the consequent expression in the form [26]

$$\begin{aligned}\langle\Phi^-_{f;jm_j}|F|\Phi_0;1m_r\rangle=&\sum_J(-1)^{1+j-J_0-J_f-2M}(2J+1)\\ &\times\begin{pmatrix} 1 & J_0 & J\\ -m_r & -M_0 & M\end{pmatrix}\\ &\times\begin{pmatrix} J_f & j & J\\ M_f & m_j & -M\end{pmatrix}\langle J_fj|F(J)|J_01\rangle. \quad (5)\end{aligned}$$

Here $\vec{J}=\vec{l}_r+\vec{J}_0=\vec{J}_f+\vec{j}$ $(=\vec{j}_1+\vec{j}_2)$ is the total angular mo-

mentum for process (1) and is a constant of the motion. On recoupling the angular momenta present in Eq. (5) according to the procedure used in Ref. [26], we find

$$\langle \Phi^{-}_{f;jm_j}|F|\Phi_0;1m_r\rangle=\sum_{j_t,m_t}(-1)^{-J_0-J_f-M_0-m_j}(2j_t+1)\times\begin{pmatrix}1 & j & j_t\\ m_r & -m_j & m_t\end{pmatrix}\times\begin{pmatrix}J_f & J_0 & j_t\\ M_f & -M_0 & m_t\end{pmatrix}\times\langle J_f j|F(j_t)|J_0 1\rangle, \tag{6}$$

with

$$\langle J_f j|F(j_t)|J_0 1\rangle=\sum_J(-1)^J(2J+1)\begin{Bmatrix}1 & J_0 & J\\ J_f & j & j_t\end{Bmatrix}\times\langle J_f j|F(J)|J_0 1\rangle. \tag{7}$$

In expressions (6) and (7),

$$\vec{j}_t=\vec{l}_r-\vec{j}=\vec{J}_f-\vec{J}_0 \tag{8}$$

is the angular momentum transferred between the unobserved reactants (A and A^{2+}) and from photon ($|\vec{l}_r|=1$) to the two photoelectrons whose spins are being analyzed. Definition (8) is in accordance with the scheme proposed in Refs. [25–27] for analyzing the spin polarization of an electron ejected in single photoionization. The expression for the angular distribution (4), obtained after substituting the final state (2) and the matrix element (6), is simplified using somewhat tedious Racah algebra and can be written in the form

$$\frac{d^3\sigma(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}=\frac{K}{2J_0+1}\sum_{j_t}\sum_{\substack{l_1,l_2\\ l_1',l_2'}}\sum_{\substack{j_1,j_2,j\\ j_1',j_2',j'}}G(l_r=1;m_r;j_t;(j_1j_2)j;(j_1'j_2')j';l_1l_1';l_2l_2';\mu_1\hat{u}_1;\mu_2\hat{u}_2;\hat{k}_1\hat{k}_2)\times d_{l_1l_2}(J_f(j_1j_2)j;J_0 1;j_t)d^{*}_{l_1'l_2'}(J_f(j_1'j_2')j';J_0 1;j_t), \tag{9a}$$

with the geometrical factor

$$\begin{aligned}G(l_r=1;m_r;j_t;(j_1j_2)j;(j_1'j_2')j';l_1l_1';l_2l_2';\mu_1\hat{u}_1;\mu_2\hat{u}_2;\hat{k}_1\hat{k}_2)&=(-1)^{1+\mu_1+\mu_2+m_r}\frac{K}{2J_0+1}\sum_{\substack{L_1,L_2,M_{L_1},M_{L_2}\\ S_1,S_2M_{s_1},M_{s_2}}}\sum_{\substack{J_1,J_2\\ J_T,M}}(-1)^{l_1'+l_2'+j+j_t+S_1+S_2}(2J_1+1)(2J_2+1)(2J_T+1)(2j_t+1)\\ &\times\sqrt{(2L_1+1)(2L_2+1)(2S_1+1)(2S_2+1)}\begin{pmatrix}l_1 & l_1' & L_1\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}l_2 & l_2' & L_2\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & S_1\\ \mu_1 & -\mu_1 & 0\end{pmatrix}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & S_2\\ \mu_2 & -\mu_2 & 0\end{pmatrix}\\ &\times\begin{pmatrix}L_1 & S_1 & J_1\\ M_{L_1} & M_{S_1} & M\end{pmatrix}\begin{pmatrix}L_2 & S_2 & J_2\\ M_{L_2} & M_{S_2} & -M\end{pmatrix}\begin{pmatrix}J_1 & J_2 & J_T\\ M & -M & 0\end{pmatrix}\begin{pmatrix}1 & 1 & J_T\\ m_r & -m_r & 0\end{pmatrix}\begin{Bmatrix}1 & 1 & J_T\\ j & j' & j_t\end{Bmatrix}\begin{Bmatrix}l_1 & \frac{1}{2} & j_1\\ l_1' & \frac{1}{2} & j_1'\\ L_1 & S_1 & J_1\end{Bmatrix}\\ &\times\begin{Bmatrix}l_2 & \frac{1}{2} & j_2\\ l_2' & \frac{1}{2} & j_2'\\ L_2 & S_2 & J_2\end{Bmatrix}\begin{Bmatrix}j_1 & j_2 & j\\ j_1' & j_2' & j'\\ J_1 & J_2 & J_T\end{Bmatrix}Y^{M_{L_1}}_{L_1}(\hat{k}_1)Y^{M_{L_2}}_{L_2}(\hat{k}_2)Y^{M_{S_1}}_{S_1}(\hat{u}_1)Y^{M_{S_2}}_{S_2}(\hat{u}_2)\end{aligned} \tag{9b}$$

and the reduced amplitudes

$$d_{l_1l_2}(J_f(j_1j_2)j;J_0 1;j_t)=(-i)^{l_1+l_2}e^{i(\sigma_{l_1j_1}+\sigma_{l_2j_2})}\sqrt{(2l_1+1)(2l_2+1)(2j_1+1)(2j_2+1)(2j+1)}\langle J_f j|F(j_t)|J_0 1\rangle \tag{10}$$

determined by the dynamics of the DPI process (1). The geometrical factor (9b) can be written also in terms of the bipolar harmonics [32] in two different ways by combining either $Y_{L_1}^{M_{L_1}}(\hat{k}_1)$ with $Y_{S_1}^{M_{S_1}}(\hat{u}_1)$ and $Y_{L_2}^{M_{L_2}}(\hat{k}_2)$ with $Y_{S_2}^{M_{S_2}}(\hat{u}_2)$, or $Y_{L_1}^{M_{L_1}}(\hat{k}_1)$ with $Y_{L_2}^{M_{L_2}}(\hat{k}_2)$ and $Y_{S_1}^{M_{S_1}}(\hat{u}_1)$ with $Y_{S_2}^{M_{S_2}}(\hat{u}_2)$.

We see that the correlation function (9) contains an incoherent sum over the angular momentum transfer $\vec{j}_t$ defined in Eq. (8). It can therefore be analyzed according to parity-favored and -unfavored transitions [23,24]. In the present case, these are characterized, respectively, by $+1$ and -1 values of $(-1)^{1+j_t-j}$. We have, from Eq. (8), $j_t=j,\ j\pm1$. The parity-favored transitions are therefore those for which $j_t=j\pm1$, whereas $j_t=j$ corresponds to unfavored transitions. Then, in view of relation (3c) and for given j_1 and j_2, the total number of parity-unfavored transitions is $2\min(j_1,j_2)+1$, whereas parity-favored transitions are $2[2\min(j_1,j_2)+1]$ for $j_1\neq j_2$ and $2[2\min(j_1,j_2)+1]-1$ for $j_1=j_2$. In the case of single photoionization, on the other hand, there will be only one $j_t=j$ and two $j_t=j\pm1$ parity-unfavored and -favored transitions, respectively. Thus the presence of two, rather than one, photoelectrons in the continuum has increased considerably the number of both parity-favored and -unfavored transitions contributing to a DPI process. The parity-unfavored transitions are known to reflect the influence of electron-ion final-state interactions and have already been observed by Langer *et al.* [33] in the spin-unresolved, nonresonant single photoionization of argon.

On account of the emission of two electrons in process (1), both of the quantum numbers J_o and J_f will simultaneously be finite, as either integers or half integers. Consequently, the angular-momentum transfer j_t defined by relation (8) will be an integer taking only $2\min(J_o,J_f)+1$ values. This in turn implies that due to the triangular condition $\Delta(|\vec{l}_r|=1,\ j,j_t)$, j too cannot be other than a finite integer. However, Eq. (3c) suggests that j_1 and j_2, both of which are half integers on account of Eqs. (3a) and (3b), can have any number of values satisfying the inequalities $|j_1-j_2|\leqslant j\leqslant j_1+j_2$. But we know from Eqs. (3a) and (3b) that $j_1=l_1\pm\frac{1}{2}$ and $j_2=l_2\pm\frac{1}{2}$, respectively. Hence, once j_1 and j_2 are fixed, l_1 and l_2 will be determined automatically. In conclusion, while spherical harmonics $Y_{S_1}^{M_{S_1}}(\hat{u}_1)$ and $Y_{S_2}^{M_{S_2}}(\hat{u}_2)$ corresponding to $S_1=(0,1)$ and $S_2=(0,1)$ contribute to expression (9) a larger number of both $Y_{L_1}^{M_{L_1}}(\hat{k}_1)$ and $Y_{L_2}^{M_{L_2}}(\hat{k}_2)$, with L_1 and L_2 determined from the conditions $|l_1-l_2'|\leqslant L_1\leqslant l_1+l_2'$ and $|l_2-l_2'|\leqslant L_2\leqslant l_2+l_2'$, respectively, will be present in that expression.

The angular distribution (9) for two spin-resolved photoelectron is extremely complicated. It is possible to parametrize by writing it in the form

$$\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}=(-1)^{\mu_1+\mu_2}\sum_{S_1,S_2,S_T,M}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & S_1\\ -\mu_1 & \mu_1 & 0\end{pmatrix}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & S_2\\ -\mu_2 & \mu_2 & 0\end{pmatrix}$$
$$\times A_{S_TM}^{S_1S_2}(m_r;\vec{k}_1;\vec{k}_2)\mathcal{Y}_{S_TM}^{S_1S_2}(\hat{u}_1,\hat{u}_2), \tag{11}$$

where the $\mathcal{Y}$'s are the bipolar harmonics [32]. An expression for the coefficients A is readily obtained by comparing Eq. (11) with Eq. (9). The advantage of writing the angular- and spin-correlation functions in the form of Eq. (11) lies in the fact that they can now be completely characterized by 12 parameters present in the expression (7) obtained by Chandra and Chrakraborty [21] to study angular and spin correlation between a photoelectron and an Auger electron emitted sequentially in one-photon, two-step, double ionization. Similar to Ref. [21], these 12 parameters herein also depend upon the experimental geometry through the propagation vectors $\vec{k}_1$ and $\vec{k}_2$ of the two photoelectrons. Expressions for these parameters are readily obtained by replacing $A_{S_TM}^{S_1S_2}$ in Eqs. (8a)–(8l) given in Ref. [21] by those extracted from Eq. (11) in this paper. Each of the 12 parameters can be analyzed in terms of parity-favored and -unfavored transitions.

There are several experimental geometries in which expression (9) [or (11)] takes particularly simpler forms. Several of such configurations are briefly discussed in the rest of this section.

(i) Both photoelectrons are taken to be polarized longitudinally to their respective directions of propagation, i.e., $\hat{u}_1\|\hat{k}_1$ and $\hat{u}_2\|\hat{k}_2$. Then, using Eq. (4.6.5) from Ref. [30], one writes

$$Y_{L_1}^{M_{L_1}}(\hat{k}_1)Y_{S_1}^{M_{S_1}}(\hat{u}_1)\rightarrow Y_{L_1}^{M_{L_1}}(\hat{k}_1)Y_{S_1}^{M_{S_1}}(\hat{k}_1)$$
$$=\sum_{A_1,a_1}\sqrt{\frac{(2L_1+1)(2S_1+1)(2A_1+1)}{4\pi}}$$
$$\times\begin{pmatrix}L_1 & S_1 & A_1\\ 0 & 0 & 0\end{pmatrix}$$
$$\times\begin{pmatrix}L_1 & S_1 & A_1\\ M_{L_1} & M_{S_1} & a_1\end{pmatrix}Y_{A_1}^{a_1*}(\hat{k}_1) \tag{12a}$$

and

$$Y_{L_2}^{M_{L_2}}(\hat{k}_1)Y_{S_2}^{M_{S_2}}(\hat{u}_1)\rightarrow Y_{L_2}^{M_{L_2}}(\hat{k}_1)Y_{S_2}^{M_{S_2}}(\hat{k}_1)$$
$$=\sum_{A_2,a_2}\sqrt{\frac{(2L_2+1)(2S_2+1)(2A_2+1)}{4\pi}}\times\begin{pmatrix}L_2 & S_2 & A_2\\ 0 & 0 & 0\end{pmatrix}\times\begin{pmatrix}L_2 & S_2 & A_2\\ M_{L_2} & M_{S_2} & a_2\end{pmatrix}Y_{A_2}^{a_2^*}(\hat{k}_2). \quad (12b)$$

After substituting Eq. (12) into Eq. (9b), sums over (M_{L_1},M_{S_1}) and (M_{L_2},M_{S_2}) are readily performed analytically using the unitarity [30] of 3-j symbols. The resulting geometrical factor will be considerably simpler than that given in Eq. (9b). The consequent final form of the correlation function (9) suggests two very interesting photon-propagation and electron-detection configurations that can be used to perform angle- and spin-resolved double-photoionization studies.

(a) One (say, e_2) of the two longitudinally polarized photoelectrons is observed along the polar axis of the photon frame. Then $\hat{k}_2=(\theta_2=0,\phi_2)$, yielding [30]

$$Y_{A_2}^{a_2^*}(\hat{k}_2)=\sqrt{(2A_2+1)/4\pi}\,\delta_{0a_2}. \quad (12c)$$

After using Eqs. (12a)–(12c), expression (9) reduces to the series of Legendre polynomials

$$\frac{d^2\sigma_{12}(m_r;\mu_1\mu_2)}{d\varepsilon_1 d\theta_1}\equiv\left.\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}\right|_{\hat{u}_1\|\hat{k}_1,\hat{u}_2\|\hat{k}_2,\hat{k}_2\|\hat{z}}$$
$$=\sum_{J_1}A_{J_1}(m_r;\mu_1 k_1;\mu_2 k_2)P_{J_1}(\cos\theta_1). \quad (13)$$

The argument θ_1 of the Legendre polynomials is the angle between two departing electrons polarized along their respective directions of propagation. The photoelectron e_2 in Eq. (13) is, however, always observed in the direction of the polar axis of the space frame. The expression for the coefficient A_J can readily be obtained and J_1 is shown from Eq. (9b) to be determined by $\max(0,|L_1-S_1|,|J_2-J_T|)\leq J_1\leq\min(2j_1,L_1+S_1,J_2+J_T)$. Since $|l_1-\frac{1}{2}|\leq j_1\leq l_1+\frac{1}{2}$, $0\leq L_1\leq 2l_1$, $S_1=(0,1)$, $|L_2-S_2|\leq J_2\leq L_2+S_2$, and $J_T=0-2$ with $0\leq L_2\leq 2l_2$ and $S_2=(0,1)$, we therefore have $\max(0,|2l_2-1|$ and $|2l_2-3|)\leq J_1\leq\min(2l_1+1,2l_2+3)$. Usually, in an atomic photoionization experiment, the initial target A and the final ion A^{2+} are in parity eigenstates. The pair of electrons escaping in DPI also will have a definite parity $(-1)^{l_1+l_2}$. But neither of the photoelectrons can be assumed to separately possess a definite parity. On the other hand, many pairs of l_1+l_2 may contribute to the two-electron continuum state such that l_1+l_2 is always either even or odd (i.e., both l_1 and l_2 have the same or opposite parities) for a given transition (1).

(b) Let us detect two longitudinally polarized photoelectrons in opposite directions, i.e., if $\hat{k}_1\equiv\hat{k}(\theta,\phi)$, then $\hat{k}_2(\theta_2,\phi_2)=-\hat{k}$, i.e., $\theta_2=\pi-\theta$ and $\phi_2=\pi+\phi$. The cross section for angle- and spin-resolved DPI of an atomic target in this collinear configuration is completely characterized by just three geometry independent parameters A, B, and C in the form of the expansion

$$\frac{d^2\sigma_{12}^{\|}(m_r;\mu_1\mu_2)}{d\varepsilon_1 d\theta}\equiv\left.\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}\right|_{\hat{u}_1\|\hat{k}_1,\hat{u}_2\|\hat{k}_2,\hat{k}_1\|\hat{k},\hat{k}_2\|-\hat{k}}$$
$$=A-2m_r B\cos\theta+\frac{1}{2}(2-3m_r^2)CP_2(\cos\theta). \quad (14)$$

Here θ is the angle that the line joining two longitudinally polarized photoelectrons makes with the polar axis of the photon frame. The correlation in this geometry will always be determined by a single parameter A, whatever the states $|J_0M_0\rangle$ and $|J_fM_f\rangle$ of A and A^{2+}, respectively, may be if the absorbed photon is LP and the photoelectrons are observed in opposite directions at the magic angle $\theta_m=54.7°$, for which $P_2(\cos\theta_m)=0$. The detailed expressions for the three parameters A, B, and C can readily be obtained. These expressions are not relevant to the present discussion and therefore have not been given in this paper. In order to extract the remaining parameters B and C from the experimental measurements, one can follow exactly the same procedure as described in Ref. [21] for angular and spin correlations between a pair of photoelectrons and Auger electrons sequentially emitted in opposite directions.

(ii) Another simple experiment on DPI may consist of observing, say, electron e_1 angle and spin resolved, while only the spin of e_2 is analyzed. The corresponding correlation function is given by

$$\frac{d^2\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1}\equiv\int\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}\,d\hat{k}_2.$$

One now substitutes Eq. (9) into the above relation, performs an integration over $\hat{k}_2$ using the orthonormality of the spherical harmonics, and simplifies the consequent expression with the help of Racah algebra. The final result can be written as

$$\frac{d^2\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1}=\frac{1}{2}\frac{d^2\sigma_1(m_r;\mu_1\hat{\mu}_1)}{d\varepsilon_1 d\hat{k}_1}+f_{12}(m_r;\vec{k}_1;\mu_1\hat{u}_1;\mu_2\hat{u}_2). \quad (15)$$

Here $d^2\sigma_1(m_r;\mu_1\hat{u}_1)/d\varepsilon_1 d\hat{k}_1$ is the angular distribution of the spin-resolved electron e_1, which is ejected in the DPI of an atom but is observed without the simultaneous detection

of the electron e_2. This expression, which is applicable to noncoincident experiments on DPI, will be explored further in Eq. (19). The function f_{12} depends, among other things, on the propagation vector $\hat{k}_1$ of e_1 and on the spin quantization directions $(\hat{u}_1,\hat{u}_2)$ of both of the photoelectrons. We thus see that the effect of not detecting the direction of ejection of one of the two photoelectrons splits the correlation function (9) into two parts: The first term on the right-hand side of Eq. (15) describes the angular distribution of the spin-resolved e_1 photoelectron observed in the noncoincident experiment and the second term f_{12} represents the contribution arising due to the analysis of the spin of the photoelectron e_2 whose direction of departure from the atomic target is not detected. With suitable choices of experimental geometries, both of the forms present in Eq. (15) can be simplified.

(iii) A still simpler experiment involving detection of the spins of both of the photoelectrons will consist of measuring spin-resolved integrated photocurrents produced in DPI. The corresponding pure spin-correlation function is given by

$$\frac{d\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1}\equiv\int\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}\times d\hat{k}_1 d\hat{k}_2.$$

On substituting Eq. (9) and simplifying, we get

$$\begin{aligned}\frac{d\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1}=\frac{1}{4}\frac{d\sigma_{12}}{d\varepsilon_1}\{&1-2m_r\mu_1\gamma_1\cos\theta_1'\\&-2m_r\mu_2\gamma_2\cos\theta_2'+4\mu_1\mu_2\\&\times[\gamma_{10}P_1(\hat{u}_1\cdot\hat{u}_2)+m_r\gamma_{11}\\&\times\sin\theta_1'\sin\theta_2'\sin(\phi_1'-\phi_2')\\&+(3m_r^2-2)\gamma_{12}(\sin\theta_1'\sin\theta_2'\cos\\&\times(\phi_1'-\phi_2')-2\cos\theta_1'\cos\theta_2')]\}.\end{aligned}\tag{16}$$

That is, a complete characterization of pure spin correlation between two simultaneously ejected photoelectrons requires just six parameters: the spin-unresolved, integrated, total photocurrent $d\sigma_{12}/d\varepsilon_1$ and five other parameters γ_1, γ_2, γ_{10}, γ_{11}, and γ_{12}. Although these parameters depend on the energies of the photoelectrons e_1 and e_2, they are totally independent of the experimental geometry. The form of the spin-correlation function (16) is identical to that obtained by us to describe the spin correlation between photoelectrons and Auger electrons emitted from an atom [20], from a ''rotationless'' molecule [21] belonging to one of the 32 point groups and from [22] rotating linear molecules. One can readily obtain expressions for the six parameters present in Eq. (16) in terms of the ''reduced'' amplitudes defined by relation (10). Even without knowing the detailed explicit expressions for these parameters, one can easily deduce the following interesting properties of the spin-correlation function (16)

(a) Let us perform a noncoincident experiment on DPI detecting only, say, electron e_1. This gives

$$\frac{d\sigma_1(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1}\equiv\sum_{\mu_2}\frac{d\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1}=\frac{1}{2}\frac{d\sigma_1}{d\varepsilon_1}[1-2m_r\mu_1\gamma_1\cos\theta_1'].\tag{17a}$$

Similarly, on specializing Eq. (16) to the detection of the electron e_2, one finds

$$\frac{d\sigma_2(m_r;\mu_2\hat{u}_2)}{d\varepsilon_2}\equiv\sum_{\mu_1}\frac{d\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1}=\frac{1}{2}\frac{d\sigma_2}{d\varepsilon_2}[1-2m_r\mu_2\gamma_2\cos\theta_2'].\tag{17b}$$

We thus see that for complete specification of the spin-resolved noncoincident experiment detecting only one of the two simultaneously ejected photoelectrons, one needs merely two parameters, either $(d\sigma_1/d\varepsilon_1,\gamma_1)$ or $(d\sigma_2/d\varepsilon_2,\gamma_2)$. (Here $d\sigma_1/d\varepsilon_1$ and $d\sigma_2/d\varepsilon_2$ are the spin-unresolved integrated photocurrents when the respective electrons e_1 and e_2 are observed in noncoincident experiments on DPI.) Expressions (17a) and (17b) are formally identical to that obtained [25] for the spin-resolved, integrated photocurrent produced in single photoionization. However, when the spin orientations of both of the photoelectrons are measured simultaneously, the number of parameters in Eq. (16) increases from 2 to 6. The spins of the two photoelectrons departing from an atom interfere with each other, affecting their mutual orientations in space. This interference effect between the quantization directions of two photoelectrons is represented by three parameters γ_{10}, γ_{11}, and γ_{12} in Eq. (16). These may therefore be called three spin-interference parameters for DPI.

(b) Neither of the noncoincident integrated photocurrents in Eq. (17) will be spin dependent if the incident light in Eq. (1) causing DPI is either LP or UP. Thus, for the noncoincident integrated photocurrent to depend on spin orientation, it is necessary that it be emitted by the absorption of CP light. This result is identical to that [34] of the spin-resolved, integrated photocurrent produced in single photoionization. The spin-resolved, integrated Auger current emitted in the decay of a vacancy created by the absorption of a photon also has been shown [35,36] to have a similar behavior with respect to the polarization of the absorbed light.

The spin-correlation function (16), on the other hand, does not become independent of spin even for LP or UP radiation. Thus, in order to study the correlation between the orientation of the spins of two photoelectrons emitted in DPI, without detecting their directions of propagation, not only

CP light may be used. The simultaneously measured integrated current of both photoelectrons emitted by the absorption of LP or UP light may also depend upon the orientation of the spins of both of these electrons due to presence in Eq. (16) of the two (γ_{10} and γ_{12}) of the three spin-interference parameters.

The procedure adopted in Ref. [21] for extracting all six parameters present in a function describing the spin correlation between photoelectrons and Auger electrons can be used also in the present case of DPI. The degree of simultaneous spin polarization of both photoelectrons can be defined analogously to that introduced in Ref. [21] for a photoelectron–Auger electron pair.

(iv) It has been mentioned already in the Introduction that experiments involving the simultaneous measurement of the spin orientation of two electrons moving in the continuum of an atom or molecule are difficult to perform due to serious intensity problems. Therefore, a much simplar experiment than those suggested hitherto will be the one in which both photoelectrons are angle resolved and the spin of only one of the two observed electrons is analyzed. Consequently, while we are studying angle- and spin-resolved photoelectron spectroscopy of, say, electron e_1, electron e_2 is detected only with its direction of propagation without any measurements of its spin. The appropriate correlation function for such experiments is

$$\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2} \equiv \sum_{\mu_2} \frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}.$$

We substitute distribution (9) in the above expression and perform a sum over μ_2 analytically. Then, after some additional simplifications, we get

$$\frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2} = \frac{1}{2}\frac{d^3\sigma_{12}(m_r)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2} + g_{12}(m_r;\mu_1\hat{u}_1;\vec{k}_1;\vec{k}_2). \tag{18}$$

Here $d^3\sigma_{12}(m_r)/d\varepsilon_1 d\hat{k}_1 d\hat{k}_2$ is the well-known angular correlation function between two spin-unresolved photoelectrons ejected in DPI. Thus the correlation between an angle- and spin-resolved photoelectron and an angle-resolved photoelectron, both of which are ejected simultaneously in DPI (1), is described by the pure angular correlation between the two ejected electrons plus another function that arises due to the analysis of the spin of either of them.

The correlation described by Eq. (18) is certainly easier than those represented by earlier functions. The angular correlation function $d^3\sigma_{12}(m_r)/d\varepsilon_1 d\hat{k}_1 d\hat{k}_2$, which is the first term on the right-hand side of Eq. (18), has been studied theoretically as well as experimentally by a number of groups [1–10]. The results of those studies can be used directly in Eq. (18) as well. A further calculation of g_{12} will be sufficient to describe the correlation (18) theoretically. The difference between the measurements of $d^3\sigma_{12}(m_r;\mu_1\hat{u}_1)/d\varepsilon_1 d\hat{k}_1 d\hat{k}_2$ and $d^3\sigma_{12}(m_r)/d\varepsilon_1 d\hat{k}_1 d\hat{k}_2$, on the other hand, will immediately give us the experimental value of the function g_{12}, which represents the influence of the detection of the spin of one of the two photoelectrons on their angular correlation.

(v) Next we consider the noncoincident angle- and spin-resolved spectroscopy of, say, the photoelectron e_1. The corresponding cross section is given by

$$\frac{d^2\sigma_1(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1} \equiv \sum_{\mu_2} \int \frac{d^3\sigma_{12}(m_r;\mu_1\hat{u}_1;\mu_2\hat{u}_2)}{d\varepsilon_1 d\hat{k}_1 d\hat{k}_2}\, d\hat{k}_2.$$

This is probably the simplest experiment that one can perform in a DPI process involving the detection of both spin and the propagation directions of one of the two ejected photoelectrons, while the other electron remains unobserved. Now we substitute Eq. (9) into the above expression, integrate over the direction $\hat{k}_2$ of e_2, and sum over its spin component μ_2. This gives, after using some complicated but straightforward, Racah algebra

$$\begin{aligned}\frac{d^2\sigma_1(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1} = (-1)^{1/2+\mu_1+m_r}\frac{K}{2J_0+1}\sum_{\substack{l_1,l_1',l_2\\ j_1,j_1',j_2}}\sum_{\substack{L_1,S_1,J_1,M\\ j,j',j_t}}(-1)^{l_1'+j_1-j_2+j+j'+j_t+S_1}(2j_t+1)(2J_1+1)\\ \times\sqrt{(2L_1+1)(2S_1+1)}[(2l_2+1)(2j_2+1)]^{-1}\begin{pmatrix} l_1 & l_1' & L_1\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} \frac{1}{2} & \frac{1}{2} & S_1\\ \mu_1 & -\mu_1 & 0\end{pmatrix}\\ \times\begin{pmatrix} L_1 & S_1 & J_1\\ M & -M & 0\end{pmatrix}\begin{pmatrix} 1 & 1 & J_1\\ m_r & -m_r & 0\end{pmatrix}\begin{Bmatrix} 1 & 1 & J_1\\ j & j' & j_t\end{Bmatrix}\begin{Bmatrix} j & j' & J_1\\ j_1' & j_1 & j_2\end{Bmatrix}\\ \times\begin{Bmatrix} l_1 & \frac{1}{2} & j_1\\ l_1' & \frac{1}{2} & j_1'\\ L_1 & S_1 & J_1\end{Bmatrix} d_{l_1 l_2}(J_f(j_1 j_2)j;J_0 1;j_t)d^*_{l_1' l_2}(J_f(j_1' j_2)j';J_0 1;j_t)Y^M_{L_1(\hat{k}_1)}Y^{-M}_{S_1}(\hat{u}_1).\end{aligned} \tag{19}$$

In this expression, $J_1=0-2$; $S_1=0,1$; and $L_1=0-3$. However, in view of the discussion presented earlier in the paper, l_1+l_2 and $l_1'+l_2$ in Eq. (19) always have the same parities for a given transition leading to DPI in an atom. This means that l_1 and l_1' should simultaneously be either even or odd. This requirement in turn implies that L_1 should always be even for the first 3-j symbol in Eq. (19) not to vanish identically. Consequently, the allowed values of L_1 are 0 and 2. Distribution (19) can now be expanded in the form

$$\frac{d^2\sigma_1(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1}=\frac{1}{8\pi}\frac{d\sigma_1}{d\varepsilon_1}\{1+\tfrac{1}{2}(2-3m_r^2)\beta_1 P_2(\cos\theta_1)-2m_r\mu_1\gamma_1\cos\theta_1'+2m_r\mu_1[P_2(\cos\theta_1)\times\cos\theta_1'+\tfrac{1}{2}P_2^1(\cos\theta_1)\sin\theta_1'\cos\times(\phi_1-\phi_1')]\delta_1+\tfrac{2}{3}(3m_r^2-2)\mu_1\xi_1 P_2^1\times(\cos\theta_1)\sin\theta_1'\sin(\phi_1'-\phi_1)\}. \tag{20}$$

Here

$$\frac{d\sigma_1}{d\varepsilon_1}=\frac{K}{3(2J_0+1)}\sum_{l_1,l_2,j_1,j_2,j_t}(2j_t+1)[(2l_1+1)(2l_2+1)\times(2j_1+1)(2j_2+1)(2j+1)]^{-1}|d_{l_1l_2}\times(J_f(j_1j_2)j;J_01;j_t)|^2, \tag{21a}$$

$$\beta_1=-2\left(\frac{d\sigma_1}{d\varepsilon_1}\right)^{-1}\sqrt{-\tfrac{5}{6}}\frac{K}{(2J_0+1)}\times\sum_{\substack{l_1,l_1',l_2\\ j_1,j_1',j_2}}\sum_{j,j',j_t}(-1)^{j_1'+j_1+j_2+j+j'+j_t}\begin{pmatrix}l_1 & l_1' & 2\\ 0 & 0 & 0\end{pmatrix}\times\begin{Bmatrix}1 & 1 & 2\\ j & j' & j_t\end{Bmatrix}\begin{Bmatrix}j & j' & 2\\ j_1' & j_1 & j_2\end{Bmatrix}\begin{Bmatrix}l_1 & l_1' & 2\\ j_1' & j_1 & \frac{1}{2}\end{Bmatrix}\times d_{l_1l_2}(J_f(j_1j_2)j;J_01;j_t)d^*_{l_1'l_2}(J_f(j_1'j_2)j';J_01;j_t), \tag{21b}$$

$$\gamma_1=(-1)^{-1/2}\left(\frac{d\sigma_1}{d\varepsilon_1}\right)^{-1}\frac{K}{4\pi(2J_0+1)}\times\sum_{\substack{l_1,l_2\\ j_2,j_t}}\sum_{\substack{j_1,j_1'\\ j,j'}}(-1)^{l_1+j_2+j+j'+j_t}(2j_t+1)[(2l_1+1)\times(2l_2+1)(2j_2+1)]^{-1}\begin{Bmatrix}l_1 & \frac{1}{2} & j_1\\ 1 & j_1' & \frac{1}{2}\end{Bmatrix}\begin{Bmatrix}j_1 & j_2 & j\\ j' & 1 & j_1'\end{Bmatrix}\times\begin{Bmatrix}1 & 1 & 1\\ j & j' & j_t\end{Bmatrix}d_{l_1l_2}(J_f(j_1j_2)j;J_01;j_t)d^*_{l_1'l_2}\times(J_f(j_1'j_2)j';J_01;j_t), \tag{21c}$$

$$\delta_1=-\frac{K\sqrt{30}}{2J_0+1}\left(\frac{d\sigma_1}{d\varepsilon_1}\right)^{-1}\times\sum_{\substack{l_1,l_1',l_2\\ j_1,j_1',j_2}}\sum_{j,j',j_t}(-1)^{l_1'+j_1+j_2+j+j'+j_t}(2j_t+1)[(2l_2+1)(2j_2+1)]^{-1}\begin{pmatrix}l_1 & l_1' & 2\\ 0 & 0 & 0\end{pmatrix}\begin{Bmatrix}j_1 & j_2 & j\\ j' & 1 & j_1'\end{Bmatrix}\times\begin{Bmatrix}1 & 1 & 1\\ j & j' & j_t\end{Bmatrix}\begin{Bmatrix}l_1 & \frac{1}{2} & j_1\\ l_1' & \frac{1}{2} & j_1'\\ 2 & 1 & 1\end{Bmatrix}\times d_{l_1l_2}(J_f(j_1j_2)j;J_01;j_t)d^*_{l_1'l_2}(J_f(j_1'j_2)j';J_01;j_t), \tag{21d}$$

$$\xi_1=i5\sqrt{\frac{3}{2}}\frac{K}{(2J_0+1)}\left(\frac{d\sigma_1}{d\varepsilon_1}\right)^{-1}\times\sum_{\substack{l_1,l_1',l_2\\ j_1,j_1',j_2}}\sum_{j,j',j_t}(-1)^{l_1'+j_1+j_2+j+j'+j_t}(2j_t+1)\times[(2l_2+1)(2j_2+1)]^{-1}\begin{pmatrix}l_1 & l_1' & 2\\ 0 & 0 & 0\end{pmatrix}\begin{Bmatrix}j_1 & j_2 & j\\ j' & 2 & j_1'\end{Bmatrix}\times\begin{Bmatrix}1 & 1 & 2\\ j & j' & j_t\end{Bmatrix}\begin{Bmatrix}l_1 & \frac{1}{2} & j_1\\ l_1' & \frac{1}{2} & j_1'\\ 2 & 1 & 2\end{Bmatrix}\times d_{l_1l_2}(J_f(j_1j_2)j;J_01;j_t)d^*_{l_1'l_2}(J_f(j_1'j_2)j';J_01;j_t). \tag{21e}$$

Both expressions (19) and (20) are formally identical to the previously [26,37] derived angular distribution of spin-resolved electrons ejected in the single photoionization of atoms. Similar to Eq. (19), the expression given in Ref. [26] also is in the angular-momentum transfer scheme defined by Eq. (8) herein, with appropriate modifications for single photoionization. This formal equivalence between Eqs. (19) and (20) and those equations derived elsewhere [26,37] is, however, an expected result. This also means that the analysis of the angle- and spin-resolved photoelectron spectroscopy of single ionization given in Refs. [26, 27] in terms of the parity-favored and -unfavored transitions becomes exactly applicable also to the expression (19) derived in this paper.

Expansion (20) means that, similar to the angle- and spin-resolved photoelectron spectroscopy of single ionization, the spin-resolved angular photocurrent in a noncoincident experiment, detecting only one of the two electrons ejected in DPI is completely characterized by five geometry independent parameters $d\sigma_1/d\varepsilon_1$, β_1, γ_1, δ_1, and ξ_1. The detailed expressions (21a)–(21e) for these parameters are naturally different from those obtained in the case of single photoionization. These have been given herein for their use in the

future as well as in Sec. III of the present study. With the help of these expressions, one can analyze the spin polarization of electron e_1 emitted in DPI using the procedure described in detail in Refs. [26, 27] for angle- and spin-resolved photoelectron spectroscopy of single ionization.

(vi) The simplest possible experiment that can be performed in DPI involving a spin analysis is the measurement of the noncoincident, spin-resolved, integrated photocurrent. For electron e_1, it is obviously given by

$$\sigma_1(m_r;\mu_1\hat{u}_1)=\int \frac{d^2\sigma_1(m_r;\mu_1\hat{u}_1)}{d\varepsilon_1 d\hat{k}_1}\,d\hat{k}_1 .$$

This yields an expression identical to that given in Eq. (17a). The parameter γ_1 present in Eq. (17a) is the same as the one occurring in Eq. (20) with the explicit form given by Eq. (21c). The degree of polarization of this noncoincident integrated current of electrons e_1 is

$$P_1(m_r;\hat{u}_1)=\frac{\sigma_1(m_r;\frac{1}{2}\hat{u}_1)-\sigma_1(m_r;-\frac{1}{2}\hat{u}_1)}{\sigma_1(m_r;+\frac{1}{2}\hat{u}_1)+\sigma_1(m_r;-\frac{1}{2}\hat{u}_1)}$$
$$=-m_r\gamma_1\cos\theta_1' . \qquad (22)$$

Thus, in order to have a nonzero degree of spin polarization in a noncoincident experiment on DPI, it is necessary that the absorbed photon be CP and the detected electron be spin resolved in other than the X-Y plane of the space frame. This degree is then proportional to the parameter γ_1. Expression (22) and other properties mentioned after it are identical to those [25,34,37] already found for the spin-polarized integrated photocurrent in the single ionization of unpolarized atoms. Expressions identical to Eq. (22) and its associated properties have been shown [35,36] to be applicable also to the spin polarization of the integrated Auger current produced in the decay of a vacancy created by the absorption of electromagnetic radiation.

III. APPLICATION

Let us consider as an example for the application of the framework developed in the preceding section DPI in the $6s$ subshell of Yb, that is,

$$h\nu_r+\mathrm{Yb}(4f^{14}6s^2\ {}^1S_0)\rightarrow \mathrm{Yb}^{2+}(4f^{14}6s^0\ {}^1S_0)+e_1(\vec{k}_1;\mu_1\hat{u}_1)+e_2(\vec{k}_2;\mu_2\hat{u}_2). \qquad (23)$$

There is more than one reason for choosing Yb as a test case in this study of spin correlation in DPI. First, for example, both Yb and Yb^{2+} are in their $^1S_0$ states, each with a closed-shell electronic configuration. Therefore, bound electrons neither in Yb nor in Yb^{2+} contribute to the SOI. The spin polarization of both photoelectrons ejected in DPI (23) will therefore be caused by the SOI in the continua of e_1 and e_2. The interesting thing to investigate here will be whether and how the SOI in the continuum of one photoelectron influences the properties of the companion photoelectron, in addition to affecting its own polarization. If the SOI plays such an important role in the DPI of this atom, it should then hopefully be observable since Yb, being a heavy atom, should give rise to a large SOI for electrons moving even in its continuum.

Second, Svensson *et al.* [38] have experimentally measured and theoretically calculated both integrated cross sections σ and the angular asymmetry parameter β for the photoelectron emitted in the single ionization in each of the $4f$ and $6s$ subshells of atomic Yb. In Ref. [39], this angle-resolved study was extended further to analyze also the spin of the ejected electron. Namely, Svensson *et al.* [39] have measured the spin-polarization parameter [40] γ for $6s$ electrons and reported both experimental as well as theoretical results for the three parameters γ, δ, and ξ [see Eq. (20)] for the single photoionization in the $4f$ subshell of Yb. Although the SOI in the bound electrons of $Yb^+(4f^{13}6s^2\ {}^2F_{5/2,7/2})$ as well as in the continuum of the photoelectron will contribute to the polarization of a $4f$ electron ejected in the single ionization of Yb, the polarization of the $6s$ electron ejected even in the single photoionization of this atom can take place by the presence of the SOI only in the continuum, as there is no fine-structure splitting in $Yb^+(4f^{14}6s^1\ {}^2S_{1/2})$. Therefore, a comparison of the angle- and spin-resolved photoelectron spectroscopy of a noncoincident experiment on DPI in the $6s$ subshell of Yb with that of its single ionization will be a direct measure of the role played by the SOI in the continuum of the unobserved electron (say, e_2) on the polarization of the detected e_1 photoelectron. In particular, γ_1 gives, according to Eq. (22), the degree of polarization of the integrated photocurrent in a noncoincident experiment on DPI. Its comparison with γ (see Ref. [40]) measured by Sevensson *et al.* [39] for single photoionization in the $6s$ subshell of Yb will give the above-mentioned information.

In order to proceed further, we note from Eq. (8) that, on account of both J_0 and $J_f=0$ in the example of Eq. (23), $j_t=0$, which in turn gives $j=1$. These make $(-1)^{1+j_t-j}=+1$, i.e., only a single parity-favored transition contributes to the DPI in the $6s^2$ subshell of Yb. The triangular condition, suggested by relation (3c), can now be used to find the angular momenta of photoelectrons e_1 and e_2, which are now considered. This gives $(j_1,j_2)=(\frac{1}{2},\frac{1}{2})$, $(\frac{1}{2},\frac{3}{2})$, $(\frac{3}{2},\frac{1}{2})$, $(\frac{3}{2},\frac{3}{2})$, etc. One further concludes from Eqs. (3a) and (3b) that $l_1,l_2=0$ and 1 for $j_1,j_2=\frac{1}{2}$; $l_1,l_2=1$ and 2 for $j_1,j_2=\frac{3}{2}$; etc. Since l_1+l_2 must always be odd in the present example of an $E1$ bound-free transition in DPI, one therefore has $(l_1,l_2)=(0,1)$ and $(1,0)$ for $(j_1,j_2)=(\frac{1}{2},\frac{1}{2})$; $(l_1,l_2)=(1,2)$ and $(2,1)$ for $(j_1,j_2)=(\frac{1}{2},\frac{3}{2})$ and $(\frac{3}{2},\frac{1}{2})$, respectively; and $(l_1,l_2)=(1,2)$ and $(2,1)$ also for $(j_1,j_2)=(\frac{3}{2},\frac{3}{2})$.

We next need to know the reduced applitude (10). To this end, we first calculate the $E1$ matrix element $\langle\Phi^-_{f;jm_j}|F|\Phi_0;1m_r\rangle$ present in Eq. (4) and used in relations (5) and (6). If one neglects the core relaxation effects by assuming that the one-electron states of Yb not directly involved in photoionizing transitions remain unchanged, then Yb can be treated as a two-electron atom and its antisymmetrized bound state is given by

$$\Phi_0(x_1,x_2)=\frac{1}{\sqrt{2}}\,6s^2(\vec{r}_1,\vec{r}_2)[\alpha(1)\beta(2)-(1\rightleftarrows 2)]. \qquad (24a)$$

In this expression, x_1 and x_2 represent space and spin coordinates of the two electrons occupying the $6s^2$ subshell of Yb. The spatial part of the state (24a) is described by the product

$$6s^2(\vec{r}_1,\vec{r}_2)=\frac{1}{r_1 r_2}P(6s;r_1)P(6s;r_2)Y_0^0(\hat{r}_1)Y_0^0(\hat{r}_2) \tag{24b}$$

of two one-electron orbitals. Here the P's are the radial functions and Y_l^m are the usual spherical harmonics. Also in Eq. (24a), α and β are the two spinors.

The antisymmetrized state of two continuum electrons with their total angular momentum j and its projection m_j can be written as

$$\begin{aligned}\Phi^-_{f;jm_j}(x_1,x_2)=&(-1)^{j_2-j_1+m_j}\sqrt{(2j+1)/2}\\ &\times\sum_{m_{j_1},m_{j_2}}\begin{pmatrix} j_1 & j_2 & j\\ m_{j_1} & m_{j_2} & -m_j\end{pmatrix}\\ &\times[\langle x_1|j_1m_{j_1}\rangle^-\langle x_2|j_2m_{j_2}\rangle^-\\ &-\langle x_2|j_1m_{j_1}\rangle^-\langle x_1|j_2m_{j_2}\rangle^-],\end{aligned} \tag{25a}$$

with

$$\begin{aligned}\langle x_1|j_1m_j\rangle^-=&(-1)^{1/2+l_1-m_{j_1}}r_1^{-1}\sqrt{(2j_1+1)}P^-(\varepsilon_1l_1j_1;r_1)\\ &\times\sum_{m_1,m_{s_1}}\begin{pmatrix} l_1 & \frac{1}{2} & j_1\\ m_1 & m_{s_1} & -m_{j_1}\end{pmatrix}Y_{l_1}^{m_1}(\hat{r}_1)\chi_{m_{s_1}}(1)\end{aligned} \tag{25b}$$

and a similar expression for $\langle x_2|j_2mj_2\rangle^-$. In Eq. (25b), $P^-(\varepsilon_1l_1j_1;r_1)$ represents the radial part of the spatial function of a photoelectron. A minus superscript on P means that it satisfies incoming-wave boundary conditions [28] appropriate for photoionization. It is normalized on the energy scale such that

$$\int_0^\infty P^{-*}(\varepsilon_1'l_1j_1;r_1)P^-(\varepsilon_1l_1j_1;r_1)dr_1=\delta(\varepsilon_1-\varepsilon_1').$$

This normalization is consistent [31(b)] with the definition of the $E1$ dipole operator given in Ref. [29] and with that of the constant K used in Eq. (4) and elsewhere in this paper. Also in Eq. (25b), $m_s=\pm\frac{1}{2}$, with $\chi_{1/2}=\alpha$ and $\chi_{-1/2}=\beta$. Also, we know from Eqs. (6) and (7) in Ref. [29] that in the dipole length approximation

$$\begin{aligned}&\langle\Phi^-_{f;jm_j}|F|\Phi_0;1m_r)\\ &\qquad=A^{(1)}\sqrt{4\pi/3}\left\langle\Phi^-_{f;jm_j}\middle|\sum_{i=1}^{2}r_iY_1^{m_r}(\hat{r}_i)\middle|\Phi_0\right\rangle,\end{aligned} \tag{26a}$$

with

$$A^{(1)}=\left(\frac{4\pi}{3}\alpha_0^3E_r^3/e^4\right)^{1/2}. \tag{26b}$$

Next, we substitute states (24) and (25) into Eq. (26) and simplify the resulting expression using Racah algebra. This yields

$$\begin{aligned}\langle\Phi^-_{f;jm_j}|F|\Phi_0;1m_r\rangle=&\frac{1}{2}\sqrt{\frac{2j+1}{\pi}}\Bigg\{(-1)^{j_2-(1/2)}\sqrt{2j_2+1}\,\delta_{(1/2)j_1}\delta_{0l_1}\delta_{1l_2}\sum_{m_{j_2}}\Bigg[\begin{pmatrix}\frac{1}{2} & j_2 & j\\ \frac{1}{2} & m_{j_2} & -m_j\end{pmatrix}\begin{pmatrix}1 & \frac{1}{2} & j_2\\ m_r & -\frac{1}{2} & -m_{j_2}\end{pmatrix}\\ &+\begin{pmatrix}\frac{1}{2} & j_2 & j\\ -\frac{1}{2} & m_{j_2} & -m_j\end{pmatrix}\begin{pmatrix}1 & \frac{1}{2} & j_2\\ m_r & \frac{1}{2} & -m_{j_2}\end{pmatrix}\Bigg]I_0(\varepsilon_1l_1j_1)I_1(\varepsilon_2l_2j_2)\\ &+(-1)^{-j_1-1/2}\sqrt{2j_1+1}\,\delta_{(1/2)j_2}\delta_{1l_1}\delta_{0l_2}\sum_{m_{j_1}}\Bigg[\begin{pmatrix}j_1 & \frac{1}{2} & j\\ m_{j_1} & -\frac{1}{2} & -m_j\end{pmatrix}\begin{pmatrix}1 & \frac{1}{2} & j_1\\ m_r & \frac{1}{2} & -m_{j_1}\end{pmatrix}\\ &+\begin{pmatrix}j_1 & \frac{1}{2} & j\\ m_{j_1} & \frac{1}{2} & -m_j\end{pmatrix}\begin{pmatrix}1 & \frac{1}{2} & j_1\\ m_r & -\frac{1}{2} & -m_{j_1}\end{pmatrix}\Bigg]I_1(\varepsilon_1l_1j_1)I_0(\varepsilon_2l_2j_2)\Bigg\}.\end{aligned} \tag{27}$$

Here

$$I_0(\varepsilon lj)=\int_0^\infty P^{-*}(\varepsilon lj;r)P(6s;r)dr \tag{28a}$$

is the overlap integral between the radial function of a bound electron occupying the $6s$ subshell before photoionization and that of a photoelectron. The radial functions present in the integral on the right-hand side of Eq. (28a) are defined in Eqs. (25b) and (24b), respectively. On the other hand,

$$I_1(\varepsilon lj)\equiv A^{(1)}\int_0^\infty P^{-*}(\varepsilon lj;r)rP(6s;r)dr \tag{28b}$$

is the one-electron radial dipole integral.

It is obvious from the right-hand side of Eq. (27) that the values of $(l_1,l_2)=(0,1)$ and $(1,0)$ only need to be considered

in the present application of DPI. In view of the previously mentioned fact that $j=1$ in this case, the values of (j_1,j_2) that will make a nontrivial contributions to the DPI (23) are therefore $(\frac{1}{2},\frac{1}{2})$ plus $(\frac{1}{2},\frac{3}{2})$ for $(l_1,l_2)=(0,1)$ and $(\frac{1}{2},\frac{1}{2})$ plus $(\frac{3}{2},\frac{1}{2})$ for $(l_1,l_2)=(1,0)$.

The total angular momentum $(\vec{J}=\vec{l}_r+\vec{J}_0=\vec{J}_f+\vec{j})$ for the process (23) is $J=1$. Then, on specializing Eqs. (5) and (7) to the present case, we find

$$\langle \Phi^-_{f;jm_j}|F|\Phi_0;1m_r\rangle=\delta_{m_rm_j}\langle J_f=0;j=1|F(J=1)|J_0=0;1\rangle \tag{29a}$$

and

$$\langle J_f=0;j=1|F(j_t=0)|J_0=0;1\rangle = -\sqrt{3}\langle J_f=0;j=1|F(J=1)|J_0=0;1\rangle, \tag{29b}$$

respectively. The combination of Eqs. (27) and (29) in Eq. (10) expresses the reduced amplitude d in terms of the radial integral, defined in Eqs. (28).

With the help of the d's so obtained, one can study the angular and spin correlations between two electrons emitted simultaneously from the $6s^2$ subshell of Yb. It can be done in any geometry or experimental arrangement, some of which have been discussed briefly in Sec. II. Similarly, one can take the photon absorbed in Eq. (23) of any polarization. However, for the reasons mentioned at the beginning of this section, we calculate hereafter only the parameter γ_1, which gives, according to Eq. (22), the degree of polarization P_1 of the integrated photocurrent of electron e_1 observed in a noncoincident experiment on DPI performed without detecting photoelectron e_2. On substituting the above-obtained reduced amplitude d, along with the appropriate values of the various quantum numbers, one finds from expressions (21a) and (21c)

$$\begin{aligned}\gamma_1=\tfrac{1}{3}\{&3(|I_1(\varepsilon_2,1,\tfrac{3}{2})|^2-|I_1(\varepsilon_2,1,\tfrac{1}{2})|^2)\rho+|I_1(\varepsilon_1,1,\tfrac{1}{2})|^2\\&-5|I_1(\varepsilon_1,1,\tfrac{3}{2})|^2+4\,\mathrm{Re}[e^{i[\sigma_{1(1/2)}(\varepsilon_1)-\sigma_{1(3/2)}(\varepsilon_1)]}\\&\times I_1(\varepsilon_1,1,\tfrac{1}{2})I_1^*(\varepsilon_1,1,\tfrac{3}{2})]\}\{|I_1(\varepsilon_1,1,\tfrac{1}{2})|^2\\&+2|I_1(\varepsilon_1,1,\tfrac{3}{2})|^2+[|I_1(\varepsilon_2,1,\tfrac{1}{2})|^2\\&+2|I_1(\varepsilon_2,1,\tfrac{3}{2})|^2]\rho\}^{-1}.\end{aligned} \tag{30}$$

Here $\sigma_{1(1/2)}(\varepsilon_1)$ and $\sigma_{1(3/2)}(\varepsilon_1)$ are the Coulomb phases for the photoelectron e_1 observed with energy ε_1 and

$$\rho=|I_0(\varepsilon_1,0,\tfrac{1}{2})|^2/|I_0(\varepsilon_2,0,\tfrac{1}{2})|^2 \tag{31}$$

is the ratio of the square of the overlap integral (28a) for the observed photoelectron e_1 to that of the unobserved photoelectron e_2.

Let us assume that there is no SOI in the continuum of the detected electron e_1. This means $\sigma_{1(1/2)}(\varepsilon_1)=\sigma_{1(3/2)}(\varepsilon_1)$ and $I_1(\varepsilon_1,1,\frac{1}{2})=I_1(\varepsilon_1,1,\frac{3}{2})\equiv I(\varepsilon_1,1)$, say. Then Eq. (30) becomes

$$\gamma_1=\frac{|I_1(\varepsilon_2,1,\frac{3}{2})|^2-|I_1(\varepsilon_2,1,\frac{1}{2})|^2}{3|I(\varepsilon_1,1)|^2\rho^{-1}+|I_1(\varepsilon_2,1,\frac{1}{2})|^2+2|I_1(\varepsilon_2,1,\frac{3}{2})|^2}, \tag{32a}$$

which is not necessarily zero. Thus the photoelectron e_1 detected in a noncoincident experiment on DPI in the $6s^2$ subshell of Yb may be spin resolved even if no SOI is included in its continuum. In this case, it is the SOI in the continuum of the undetected photoelectron e_2 that contributes to the polarization of e_1. This will happen if and only if the radial dipole integrals $I_1(\varepsilon_2,1,\frac{1}{2})$ for $(l_2=1,\ j_2=\frac{1}{2})$ and $I_1(\varepsilon_2,1,\frac{3}{2})$ for $(l_2=1,\ j_2=\frac{3}{2})$ are not equal to each other and at least one of these is not zero.

The noninclusion of the SOI in the continuum of e_2, rather than e_1, will make $I_1(\varepsilon_2,1,\frac{1}{2})=I_1(\varepsilon_2,1,\frac{3}{2})\equiv I(\varepsilon_2,1)$, say. This reduces Eq. (30) to the form

$$\begin{aligned}\gamma_1=\tfrac{1}{3}\{&|I_1(\varepsilon_1,1,\tfrac{1}{2})|^2-5|I_1(\varepsilon_1,1,\tfrac{3}{2})|^2\\&+4\,\mathrm{Re}[e^{i[\sigma_{1(1/2)}(\varepsilon_1)-\sigma_{1(3/2)}(\varepsilon_1)]}I_1(\varepsilon_1,1,\tfrac{1}{2})I_1^*(\varepsilon_1,1,\tfrac{3}{2})]\}\\&\times[|I_1(\varepsilon_1,1,\tfrac{1}{2})|^2+2|I_1(\varepsilon_1,1,\tfrac{3}{2})|^2+3|I(\varepsilon_2,1)^2\rho]^{-1}.\end{aligned} \tag{32b}$$

The degree of polarization (22) even in this case obviously does not vanish as long as any of the radial dipole integrals $I_1(\varepsilon_1,1,\frac{1}{2})$ for $(l_1=1,\ j_1=\frac{1}{2})$ and $I_1(\varepsilon_1,1,\frac{3}{2})$ for $(l_1=1,\ j_1=\frac{3}{2})$ is different from zero.

Finally, let us consider that the SOI is included in the continuum of neither photoelectron e_1 nor photoelectron e_2. This means that $I_1(\varepsilon_1,1,\frac{1}{2})=I_1(\varepsilon_1,1,\frac{3}{2})$, with $\sigma_{1(1/2)}(\varepsilon_1)=\sigma_{1(3/2)}(\varepsilon_1)$ and $I_1(\varepsilon_2,1,\frac{1}{2})=I_1(\varepsilon_2,1,\frac{3}{2})$. Then, expressions (30) and (32) show that $\gamma_1=0$. Thus the integrated photocurrent observed in a noncoincident experiment on the DPI in the $6s$ subshell of Yb will certainly be unpolarized if the SOI is not taken into account in the continua of either of the emitted electrons.

According to Eq. (32a), the SOI present in the continuum of an electron affects not only its own behavior but also that of its companion electron, which is simultaneously emitted along with it in a DPI. This behavior of a pair of photoelectrons in DPI is very different from that of a photoelectron and an Auger electron emitted sequentially. In the latter case, it has been found [20–22] that the SOI in the continuum of one electron does not make any contribution to the polarization of the other electron if the photoelectron and Auger electron are observed independently. Therefore, the motions of two electrons emitted in DPI are correlated not only by the sharing between them of the energy of the absorbed photon in excess of the ionization potential and by the mutual Coulomb repulsion, but also by the SOI present in their continua. Consequently, experiments on DPI are more stringent tests of theoretical models for studying electron-electron correlations. Even the noncoincident experiments on DPI are potentially a richer source of information than a single-photoionization process.

If it is only the SOI that is responsible for the spin polarization of a photoelectron, then the γ (see Ref. [40]) mea-

sured by Svensson *et al.* [39] for electrons emitted in the single photoionization of the 6*s* subshell of Yb should, in principle, be equal to the γ_1 given by Eq. (32b) in a noncoincident experiment on DPI in the same subshell of the same target. It should be so for the simple reason that the latter expression includes, similarly to the case of single photoionization [39] the SOI in the continuum of only the observed of the two emitted electrons. A difference in the values of γ (for single photoionization) and γ_1 [given by Eq. (32b)] will, consequently, mean that either the SOIs in the two cases are different and/or there are factors other than the SOI in the continuum of the observed electron that also contribute to the spin polarization of electrons in a noncoincident experiment on DPI. Similarly, expression (32a) provides a method to find out quantitatively the contribution made by the SOI in the continuum of the unobserved photoelectron to the polarization of the observed electron in a noncoincident experiment on DPI.

Finally, expression (30) is derived when the light absorbed in Eq. (23) is CP. It has been done for the simple reason that the degree (22) of spin polarization of the integrated photocurrent observed in a noncoincident experiment vanishes identically if the incident photon is LP or UP. However, as mentioned in Sec. II, it is not necessarily so if one is studying the degree of spin polarization of the angle-resolved photocurrent in a noncoincident experiment on DPI or detecting both of the outgoing electrons simultaneously. In that case, absorbed light may be LP, UP, or CP. the photoionization matrix element (27) can be used even for LP or UP electromagnetic radiations inducing DPI.

IV. CONCLUSION

This paper presents a study of the spin polarization of electrons ejected in the DPI of an atom. An angular- and spin-correlation function for DPI in atoms has been derived. Various geometrical configurations and experimental arrangements are discussed when it becomes simpler to perform such measurements. The simplest possible experiment that one can easily carry out in DPI is one in which only one of the two photoelectrons is detected. The angle- and spin-resolved photoelectron spectroscopy of such noncoincident experiments on DPI is shown to be formally identical to that of the single-photoionization studies performed hitherto very successfully both theoretically and experimentally. This identity means that, while one of the two electrons emitted in a DPI remains unobserved, the spin-resolved angular distribution of the observed electron is completely characterized by five energy-dependent parameters that are not affected by the geometry of an experiment. It also means that the well-developed theoretical analysis [25–27] of angle- and spin-resolved photoelectron spectroscopy of single photoionization is applicable also to angle- and spin-resolved noncoincident experiments on DPI. For example, the integrated photocurrent of the observed electron in a noncoincident experiment will have no spin selection if the absorbed light is either LP or UP and its degree of polarization for CP light is proportional to a single parameter. Such spin-resolved, noncoincident measurements on DPI are much simpler to perform, and within the reach of the existing experimental facilities, than those in which spins of both of the photoelectrons are analyzed.

The theoretical framework developed in this paper is applied to a real physical system, namely, DPI in the 6*s* subshell of atomic Yb. Because both Yb and the doubly charged residual photoion Yb^{2+} formed after DPI are in their $^1S_0$ state, each with a closed-shell electronic structure, neither of these has any SOIs. The spin polarization of the two photoelectrons in this can therefore be caused by the SOI only in the continua. Our analysis shows that the SOI in the continuum of one electron affects not only its own behavior but also that of its companion electron ejected simultaneously along with the former. Thus photoelectrons produced in DPI share not only the energy of the absorbed photon in excess of the ionization potential but also the SOI present in the continuum of each of the ejected electrons, in addition to mutually experiencing Coulomb repulsion. It happens in coincident as well as in noncoincident experiments on DPI. This is very different from that found in those noncoincident experiments in which either a photoelectron or a subsequently emitted Auger electron is observed independently. In the latter case, the energy of the absorbed photon in excess of the ionization potential is completely carried away by the photoelectron without sharing it with the Auger electron and the SOI in the continuum of the unobserved electron does not affect any of the properties of the observed electron at all. Thus, even noncoincident experiments involving spin analysis in DPI provide a greater wealth of information on electron-electron correlations than those in which one does not detect the spin of the observed electron.

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