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Einstein-Podolsky-Rosen-Bohm correlation in photoelectron–Auger-electron coincidence spectroscopy of atoms

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We present a theoretical analysis of spin-entanglement between a photoelectron and an Auger electron sequentially ejected from an atom (say, A) following the absorption of a single photon. Entanglement, both without and with spin-orbit interaction has been analyzed. In the former case, entanglement in a photo-Auger electron pair is generated only by the electrostatic Coulomb forces inside an atom and completely characterized merely by the multiplicities of the electronic states of A, A^{+^*}, A^{2+} participating in the process, without using any protocols already suggested in the literature for this purpose. The presence of both the Coulomb and spin-orbit interactions, on the other hand, couples the entanglement of this bipartite state with the dynamics and kinematics of the whole process, as well as with the properties of the absorbed photon. In such cases, therefore, it is not possible to predict, *a priori*, the photo-Auger electron entanglement. These discussions have quantitatively been illustrated by several examples.

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I. INTRODUCTION

The present studies of entanglement, albeit in a different context, started with the presentation of a paper by Einstein, Podolsky, and Rosen [1] almost 70 years ago on bipartite states. Later, Ref. [1] was interpreted by Bohm [2] for discrete variables of two spin-half particles. Although, an innumerable number of papers have discussed entanglement since then in one context or the other; however, the realization that entanglement is a resource [3] which should be available in order to successfully put to technological use several protocols developed in recent years in quantum information (QI) science came only a few years ago. Moreover, further investigations [4,5] have shown that entanglement can even affect macroscopic thermodynamical properties (e.g., magnetic susceptibility or heat capacity of solids). Consequently, there has been an upsurge in both theoretical and experimental activities for developing methods for the generation and detection of nonseparable (i.e., entangled) states of two or more particles. Each particle in an entangled (or EPR [1]) pair must have at least two independent states simultaneously accessible to it. Such particles, each with two states, have come to be known as qubits in QI.

It has already been proposed [6] that electron spin as a qubit can be used in quantum computation [7]. Consequently, creation of a solid-state quantum computer, based on spin qubits, has already been suggested [7,8] in spintronics (i.e., active manipulation of the spin degree of freedom of electrons in a solid state environment) [9]. For use of electrons in quantum communication [7,10] one requires, on the other hand, two entangled, but mobile, electrons which are separately addressable because of their spatial separation. In order to have on-chip quantum communication, a number of theoretical proposals [10–13] for electron entanglers have hitherto been made in spintronics. For example, extraction of a Cooper pair from a superconductor [12] or of the singlet ground state of a quantum dot with an even number of electrons [13] are some of the methods which have already been investigated in spintronics.

There are several simple, well known processes in atomic and molecular physics which are capable of producing two or more spatially separated electrons moving freely in space. These processes can equally take place in solid, liquid, and gaseous phases of matter. For example, in one-step double photoionization (1-DPI) [14], two electrons are simultaneously ejected following the absorption of a single photon in a target. It is analogous to the parametric down conversion [15] in quantum optics. "Shake-off" [16] and "knockout" [17] are the two main mechanisms [18] which have primarily been suggested for 1-DPI. In both of these mechanisms, the incident photon is absorbed by only one of the electrons bound in the atom or molecule, resulting in its departure with a high speed from the target. On the other hand, the ejection of the second electron in 1-DPI is a consequence [18] of the soft [16] or hard binary [17] collision with the first one. The two electrons, coming out from a target in 1-DPI [14], may or may not be *equivalent* (i.e., with the same values of the principal quantum number n and orbital angular quantum number ℓ in an atom) [19], but they are from the same many-body electron state of the atom and share between themselves energy of the absorbed photon in excess to that needed to eject both of them. The spin-entanglement properties of two such photoelectrons emitted together from an atom [20] or a rotating linear molecule [21] have already been analyzed in detail.

Another, more commonly used and well-known process which can give us two electrons in the continuum is the two-step double photoionization (2-DPI) [22]. In the first step of this process, absorption of a single photon ejects an electron (say, e_1) from an inner shell of the target, forming an excited photoion. The subsequent, nonradiative decay of this excited photoion leads to the ejection of one more (say, e_2) electron. The two sequentially emitted particles are called [22] photoelectron and Auger electron, respectively. Thus, unlike in 1-DPI, e_1 and e_2 in 2-DPI come out from the many-body states of the initial target and of its excited photoion, respectively [22]. Second, in 2-DPI, it is only the energy of the e_1 which varies with that of the ionizing radiation. But the energy of the Auger electron changes with that of the excited photoion and/or of the residual dication formed after the departure of e_1 and e_2. The photoelectron and Auger electron in 2-DPI are, in general, nonequivalent [19,22]. In this paper, we analyze the spin-entanglement properties of a photo-Auger-electron pair, sequentially departed from an atom in 2-DPI.

On the basis of the interactions one takes into account, studies of both 1-DPI and 2-DPI in atomic targets can conveniently be divided in two broad classes: One possibility is to consider only the electrostatic Coulomb forces experienced by the atomic constituents. In the second possibility, the spin-dependent force which, in the case of atoms, is primarily spin-orbit interaction (SOI), is also included in a study of the proposed process. This paper presents investigations of spin-entanglement between two electrons ejected from an atom in 2-DPI both without and with SOI.

In Sec. II, we introduce relevant notations, conventions, and the density operator (DO) to be employed in this paper for the study of 2-DPI of an atom following the absorption of a single photon. A successful technological application of QI crucially depends upon the availability of at least one simple, but stringent, criterion for deciding, theoretically as well as experimentally, whether a given state is separable or not and also to quantify the amount of its entanglement. Therefore, Sec. II also contains a brief description of the criteria and of different measures of entanglement currently in vogue. In Sec. III, this DO is used first to derive an expression for a density matrix (DM) when SOI is not taken into account in any parts of the two-step process (1). An analysis of this DM pertaining to the spin-entanglement properties of photo-Auger-electron pair is also presented therein. (A brief account of some of the results discussed in the Sec. III herein was given in Ref. [23].) On the other hand, derivation of a DM needed to study the desired entanglement when SOI is included in each part of 2-DPI process (1) is contained in Sec. IV. In addition to other things, therein we discuss the relevant properties of this DM. A quantitative application of this DM with SOI to 2-DPI in Xe is also presented in Sec. IV and the results are compared with those obtained without SOI. Finally, Sec. V contains Conclusions of the present study.

II. PRELIMINARIES

A. Density operator

Let us assume that $|A\rangle$ represents the antisymmetrized initial electronic state, with energy E_0, of our atomic target A. If $|A^{+^*}\rangle$ and $|A^{2+}\rangle$ are taken to be the antisymmetrized electronic states of the respective excited photoion A^{+^*} (possessing energy E_e) and of the dication A^{2+} (with energy E_f) of this target, then the two well-known consecutive steps in Auger emission are [22]

$$\gamma(|\vec{\ell}_r|=1,m_r)+|A\rangle \rightarrow |A^{+^*}\rangle + e_1(\mu_1\hat{u}_1\vec{k}_1) \qquad (1a)$$

and

$$|A^{+^*}\rangle \rightarrow |A^{2+}\rangle + e_2(\mu_2\hat{u}_2\vec{k}_2). \qquad (1b)$$

Here, in the first step (1a), γ represents the incident photon of frequency ν_r and, hence, energy $E_r=h\nu_r$, with the subscript r standing for radiation. This photon is absorbed by one of the inner-shell electrons of the target atom A leading to its (i.e., atom's) ionization. For photons possessing such required energies, it is usually sufficient [19] to treat the ionization step (1a) in the electric dipole ($E1$) approximation. Then, $|\vec{\ell}_r|=1$ represents the angular momentum of the photon in the $E1$ approximation. The parameter m_r in (1a) specifies the polarization of γ: $m_r=0$ for linear polarization (LP), $m_r=+1$ and -1 for a photon possessing circular polarization (CP) with positive helicity (PH) and negative helicity (NH), respectively; unpolarized (UP) electromagnetic radiation, on the other hand, is taken to be an even mixture of the waves with $m_r=\pm 1$. Further, e_1 and e_2 in (1a) and (1b) are the photoelectron and the Auger electron, respectively. The propagation vector of the $i(=1,2)$th electron (e_i) is represented herein by $\vec{k}_i=[k_i,\hat{k}_i(\theta_i,\phi_i)]$ such that its kinetic energy is given by $\epsilon_i=\hbar^2k_i^2/2m$. Also in (1), $\mu_i(=\pm\frac{1}{2})$ is the projection of the spin angular momentum of the ejected electron e_i along the direction of quantization $\hat{u}_i=(\vartheta_i,\varphi_i)$.

Energy should obviously be conserved in each of the two steps of the process (1) separately. This means, while kinetic energy $\epsilon_1=h\nu_r-(E_e-E_0)$ of the photoelectron e_1 varies with the frequency of the ionizing radiation in (1a); energy $\epsilon_2=(E_e-E_f)$ of the Auger electron e_2 in (1b) is completely independent of the energy of the absorbed photon and is totally determined from that of A^{+^*} and A^{2+}. Unless stated otherwise, the polar axis of our right-handed coordinate system (see Fig. 1) is along the direction of the electric field vector if absorbed photon is LP; however, for CP or UP ionizing radiation, its direction of incidence defines the OZ axis of our frame of reference.

The incident photon and the target atom are completely uncorrelated before the interaction between the two takes place. This, in other words, means that the DO for the combined (photon+atom) system in Eq. (1a) is separable before the absorption of the photon and is simply a direct product $\rho_i=\rho_0\otimes\rho_r$ of the two DOs, $\rho_0=|A\rangle\langle A|$ of the target atom A and $\rho_r=|1,m_r\rangle\langle 1,m_r|$ of the ionizing radiation. Here, $|1,m_r\rangle$ denotes the state of a photon in the $E1$ approximation with its polarization specified by the parameter m_r. Now the DO for the photoionization of the atom A, taking place in the first of the two-step process (1), is given by (see, for example, Refs. [24,25])

$$\rho_1 = \mathcal{K}_p F_p \rho_i F_p^{\dagger}. \qquad (2a)$$

The photoionization operator F_p and the quantity $\mathcal{K}_p$, both in the $E1$ approximation, are obtained, for example, from Ref. [26]. The DO for the complete two-step process (1) can now readily be written to be

$$\rho_f = \mathcal{K}_a F_a \rho_1 F_a^{\dagger} = \mathcal{K} F_a F_p(\rho_0\otimes\rho_r)F_p^{\dagger}F_a^{\dagger}. \qquad (2b)$$

The Auger emission operator F_a and the quantity $\mathcal{K}_a$, occurring in (2b), are given elsewhere [27]. Although, the explicit

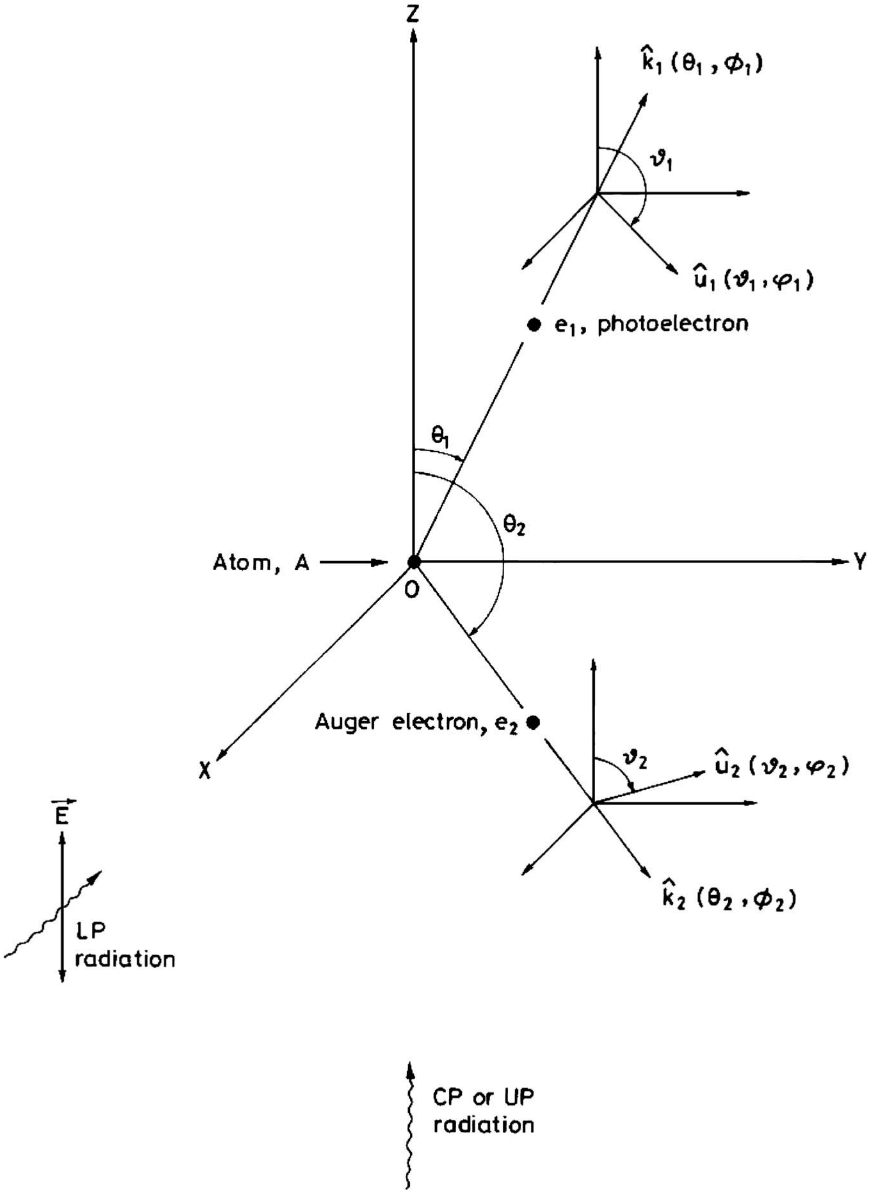

FIG. 1. Coordinate system showing the photon-fixed frame of reference OXYZ and the propagation directions $(\hat{k}_1,\hat{k}_2)$ as well as the spin quantization directions $(\hat{u}_1,\hat{u}_2)$ of the photoelectron e_1 and of the Auger electron e_2. These two electrons are sequentially ejected from the atom A, following the absorption of a single photon, situated at the origin O of our coordinate system. The OZ axis is the quantization direction for the electronic states of the atomic species A, A^{+^*}, A^{2+} participating in the two-step process (1). Each electron enters its own Mott detector [61], oriented along $\hat{u}_1(\vartheta_1,\varphi_1)$ for e_1 and $\hat{u}_2(\vartheta_2,\varphi_2)$ for e_2. These detectors record whether an ejected electron's spin is up or down with respect to its own quantization direction.

forms of neither $\mathcal{K}_p$ nor $\mathcal{K}_a$ are required for the following discussions, it should, nevertheless, be mentioned that they depend, among other things, on the respective energies ϵ_1 and ϵ_2, but do not involve any of the angle or spin related quantities of the species participating in the processes (1). In the second equation on the right-hand side of (2b), we have defined, for brevity, $\mathcal{K}\equiv\mathcal{K}_a\mathcal{K}_p$. The form (2) of the DO is equivalent to those given elsewhere (see, e.g., Refs. [22,28]) for other two-step processes like two-stage cascade of γ rays, etc.

To perform the desired investigations, we need to calculate a matrix for the DO ρ_f defined in (2). Electrons (e_1,e_2) emitted in 2-DPI may have the same $[\epsilon_1=\epsilon_2$ (i.e., $k_1=k_2$)] or different $[\epsilon_1\neq\epsilon_2$ (i.e., $k_1\neq k_2$)] energies [subject to satisfying two different conservation conditions, one for each step in (1)] and move in any two directions $\hat{k}_1$ and $\hat{k}_2$. Our study requires a density matrix (DM) which is diagonal in energies as well as in the directions of propagation (i.e., diagonal in $\vec{k}_1$ and $\vec{k}_2$). But this DM must necessarily be nondiagonal with respect to the components (i.e., μ_1,μ_2) of the spin angular momenta of the photoelectron and Auger electron (e_1,e_2), respectively. The appendixes to this paper describe calculations of such DMs without and with SOI, respectively.

B. Criteria and measures for entanglement

A successful technological application of the QI science crucially depends upon the availability of simple, but stringent, criteria for deciding, theoretically as well as experimentally, whether a given state is separable or entangled and also for quantifying the amount of its entanglement. One of the several conditions [29] for the separability of a bipartite state of qubits is that the partial transpose (PT), with respect to either of the two particles, of its DM must remain positive [30,31]. (Here, positivity of a DM means that none of its eigenvalues should be less than zero [30,31].) This condition was initially found [30] to be necessary but later shown [31] to be sufficient as well. It, consequently, is a very stringent criteria which, nevertheless, is extremely easy to use. Also, it is equally applicable to both pure and mixed states of two particles each defined in a two-dimensional Hilbert space. In our present theoretical study we have, therefore, tested the nonseparability (i.e., entanglement) or otherwise of the spin-bipartite states of two electrons by using the necessary as well as sufficient, stringent Peres-Horodecki [30,31] condition.

Various methods [32] have hitherto been suggested for theoretically quantifying the entanglement of a pair of qubits. In the present study, we have quantified the degree of entanglement of (e_1,e_2) emitted in the process (1) by calculating each of those three measures which are most commonly being used in the current theoretical discussions of the properties of entangled states. Namely, the negativity [33–35] $\mathcal{N}(\rho_f)$, and [36–38] concurrence $\mathcal{C}(\rho_f)$ as well as entanglement of formation $\mathcal{E}_F(\rho_f)$ of a given density matrix ρ_f.

(i) The negativity of a bipartite state ρ_f of qubits is defined as [33–35]

$$\mathcal{N}(\rho_f)=\max[0,-2(\text{minimum eigenvalue of the PT of }\rho_f)]. \tag{3}$$

Thus, negativity is the degree to which a given state violates the Peres-Horodecki condition [30,31] for it to be separable. Vidal and Werner [35] have shown that $\mathcal{N}(\rho_f)$ is a good entanglement measure. The PT of a DM can have at most one negative eigenvalue [33]. Consequently, negativity (3) can vary from zero to unity for a separable to a maximally entangled bipartite state, respectively.

(ii) The concurrence

$$\mathcal{C}(\rho_f)=\max(0,\sqrt{\lambda_1}-\sqrt{\lambda_2}-\sqrt{\lambda_3}-\sqrt{\lambda_4}) \tag{4a}$$

for a DM ρ_f was first introduced in Refs. [36–38]. Here, λ_i's are the successively decreasing eigenvalues of the matrix

$$R=\rho_f\widetilde{\rho_f}, \tag{4b}$$

with the spin-flipped state of ρ_f given by

$$\widetilde{\rho_f}=(\sigma_y\otimes\sigma_y)(\rho_f)^*(\sigma_y\otimes\sigma_y)\quad\text{and } \sigma_y=\begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}, \tag{4c}$$

and $(\rho_f)^*$ represents complex conjugate of the DM ρ_f. Although, negativity and concurrence can differ for a mixed entangled state; for a pure bipartite state of qubits, however, these two have equal values [32]. Concurrence too has been used [32,39] as a measure of entanglement. The pure state of two qubits is separable if its concurrence is zero, it is maximally entangled if its concurrence is one; concurrence has a value between zero and one, otherwise.

(iii) The third quantity currently in use for calculating the degree of nonseparability of a bipartite state of qubits is the entanglement of formation. The concept was originally introduced by Bennett *et al.* [40]. Hill and Wootters [37] conjectured an explicit formula for it for a special class of DMs. The following expression

$$\mathcal{E}_F(\rho_f)=-\frac{1+\sqrt{1-\mathcal{C}(\rho_f)^2}}{2}\log_2\left(\frac{1+\sqrt{1-\mathcal{C}(\rho_f)^2}}{2}\right)-\frac{1-\sqrt{1-\mathcal{C}(\rho_f)^2}}{2}\log_2\left(\frac{1-\sqrt{1-\mathcal{C}(\rho_f)^2}}{2}\right) \tag{5}$$

for the entanglement of formation was proved [41] for an arbitrary state of two qubits. It is obvious that $\mathcal{E}_F(\rho_f)$ is an increasing function of the concurrence $\mathcal{C}(\rho_f)$.

Entanglement witness [31,42–45], on the other hand, is probably the only protocol among those presently available in QI science which has hitherto been very successfully implemented for experimental characterization of the entangled states of two [46,47] or more photons [48]. Originally, it was theoretically suggested by Horodecki *et al.* [31] and subsequently studied in Refs. [42–45].

III. EINSTEIN-PODOLSKY-ROSEN-BOHM CORRELATION IN A PHOTO-AUGER-ELECTRON PAIR IN THE ABSENCE OF SPIN-ORBIT INTERACTION

A. Density matrix

Let us first investigate the spin-entanglement properties of the bipartite state formed by sequentially emitted photoelectron e_1 and Auger electron e_2, disregarding all forces which may arise due to spins of the electrons [in A, in A^{+^*}, in A^{2+}, and of (e_1,e_2)] participating in one or both of the steps of the process (1). The entanglement between (e_1,e_2) will, thus, be generated purely by the electrostatic Coulomb forces acting among the nucleus and electrons in the target atom A. It can, therefore, be called as Coulombic spin-entanglement of (e_1,e_2). Moreover, both the $E1$ photoionization and the Auger emission operators F_p and F_a, respectively, are spin-independent. In the absence of such spin-dependent interactions which, in the case of atoms, is primarily SOI, the orbital and spin angular momenta in each of the two steps of the process (1) are separately conserved. These conservation conditions are expressed in Eqs. (A1a), (A2a), (A1b), and (A2b), respectively, in Appendix A. This, in other words, means that the L-S (i.e., Russell-Saunders) [19] coupling naturally becomes applicable in each step of (1).

We have derived an expression for the desired DM. Our derivation, whose details are given in the Appendix A, is completely general, rigorous, and independent of all dynamical models (except that it is in the L-S coupling as no SOI is taken into account) which can be used in a theoretical study of the 2-DPI process (1) in any atom. The final expression (A13) for the DM can be written as a product of two independent terms which describe two entirely different physical situations. It is obvious from Eqs. (A14a) that the first term [i.e., $d^3\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2$] on the right-hand side of the DM (A13) depends upon, among other things, the orbital angular momentum of each of (A,A^{+^*},A^{2+}); phase shifts, energies (ϵ_1,ϵ_2), and directions $(\hat{k}_1,\hat{k}_2)$ of emission of both of the emitted electrons $(e_1,\ e_2)$; polarization (m_r) of the photon absorbed in the first of the two processes (1). This term, in addition, contains both the $E1$ amplitude (A14b) (determined by the electronic states $|A\rangle$ and $|A^{+^*}\rangle$, plus the continuum orbital of e_1) for photoionization (1a) and the amplitude (A14c) (obtained from the electronic states $|A^{+^*}\rangle$ and $|A^{2+}\rangle$, including the continuum orbital of e_2) for Auger emission in (1b). Thus, $d^3\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2$ in (A13) very much depends upon the dynamics as well as the kinematics of the two-step process (1). But it includes neither the spins nor the quantization directions $(\hat{u}_1,\hat{u}_2)$ of any of the two emitted electrons (e_1,e_2), or of the target atom A, excited photoion A^{+^*}, and the dication A^{2+}. However, it has an implicit dependence on the spins (S_0,S_e,S_f) of each of the three atomic species participating in (1). For, energies of the respective electronic states $(|A\rangle, |A^{+^*}\rangle, |A^{2+}\rangle)$ depend on their multiplicities $[(2S_0+1),(2S_e+1),(2S_f+1)]$. Hence, $d^3\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2$ in the DM (A13) describes purely angular correlation between the photoelectron e_1 and the Auger electron e_2 in the *L-S* coupling scheme, expressed in Eqs. (A1) and (A2), for the angular momenta of the particles involved in 2-DPI (1). Its value is always *positive* and it acts merely as a *multiplicative* factor in the DM (A13) for a given Auger process (1).

The second term [i.e., $\sigma(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'}$], present on the right-hand side of the DM (A13), is defined in Eq. (A15). Unlike the angular correlation, $\sigma(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'}$ represents a 4×4 matrix. It contains neither any of those physical quantities $(eg,\epsilon_1,\epsilon_2,\hat{k}_1,\hat{k}_2,m_r,$ etc.) nor the dynamical amplitudes (A14b) and (A14c) which affect the angular correlation $d^3\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2$. On the other hand, $\sigma(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'}$, is completely determined from Eq. (A15) by the spins of the five particles (i.e., A, A^{+^*}, A^{2+}, e_1, e_2) which participate in the two steps of the process (1), in addition to the quantization directions $(\hat{u}_1,\hat{u}_2)$ and by the components (μ_1,μ_2) along these unit vectors of the spins of the photoelectron e_1 and Auger electron e_2. Hence, the second term on the right-hand side of the DM (A13) describes purely spin correlation between the two electrons (e_1,e_2), ejected sequentially in the process (1) in the absence of SOI.

B. Are photoelectron and Auger electron in a pair entangled?

In order to definitively test the spin-entanglement properties of a photo-Auger electron pair (e_1,e_2), produced in the 2-DPI (1) in the absence of SOI, we have applied to our DM (A13) the necessary and sufficient, stringent Peres-Horodecki condition [30,31]. It has already been discussed elsewhere in this paper that $d^3\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2$ in (A13) describes purely angular correlation of (e_1,e_2) in the *L-S* coupling and always has a single positive value for a given experimental configuration. Consequently, the properties of the electrostatically generated spin-entanglement between (e_1,e_2) in the present case will completely be determined by the spin correlation matrix (A15). We, therefore, do not write in the following the angular correlation (A14a) explicitly and represent, for brevity, the DM simply by the second term present on the right-hand side of (A13). That is, unless stated otherwise, in the present subsection we write

$$\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2\rangle \to \sigma(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'}. \tag{6}$$

[In various equations in this paper, we have interchangeably used letters $(0,e,f)$ for (A,A^{+^*},A^{2+}) in side Dirac's bra and ket notations, for brevity.] Then

$$\gamma_1(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'} \equiv \sigma(S_0;S_e;S_f;\hat{u}_1;\hat{u}_2)_{\mu_1'\mu_2,\mu_1\mu_2'} \tag{7a}$$

and

$$\gamma_2(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'} \equiv \sigma(S_0;S_e;S_f;\hat{u}_1;\hat{u}_2)_{\mu_1\mu_2',\mu_1'\mu_2} \tag{7b}$$

are the PTs of (6) with respect to the photoelectron e_1 and the Auger electron e_2, respectively.

According to the spin conservation requirements (A1b) and (A2b), applicable in the present case of *L-S* coupling, we must have $S_e=S_0\pm\frac{1}{2}$ and $S_e=S_f\pm\frac{1}{2}$, respectively. The same two respective conditions are obtained by applying the triangular rules for the coupling of two of the angular momenta so that neither of the 6-*j* symbols, present in (A15), vanish identically [49]. These requirements, in other words, mean that the respective spins (S_0,S_e,S_f) of the electronic states $(|A\rangle,|A^{+^*}\rangle,|A^{2+}\rangle)$ participating in the processes (1) are not totally independent; they, instead, are related by the conditions $S_0=S_f=|S_e\pm\frac{1}{2}|$, or $|S_0-S_f|=1$ with $S_0=|S_e\pm\frac{1}{2}|$ and $S_f=|S_e\mp\frac{1}{2}|$. Let us study spin-entanglement between photoelectron e_1 and Auger electron e_2 in both of these situations occurring when SOI is not taken into account.

(i) $S_0=S_f=|S_e\pm\frac{1}{2}|$: Here, one needs to consider the following three cases separately.

(a) $S_0=S_f=1/2$ with $S_e=0$. That is, multiplicities of each of the electronic states of A and A^{2+} is two; whereas, the excited photoion A^{+^*} is in its singlet electronic state. The DM obtained from (A15) and (6) is given by

$$\sigma\Big(S_0=\tfrac{1}{2};S_e=0;S_f=\tfrac{1}{2};\hat{u}_1,\hat{u}_2\Big)_{\mu_1\mu_2,\mu_1'\mu_2'} = \tfrac{1}{4}\delta_{\mu_1\mu_1'}\delta_{\mu_2\mu_2'} \equiv (\sigma_0)_{\mu_1\mu_2,\mu_1'\mu_2'}, \tag{8}$$

where δ_{ab} is the Kronecker delta function [49]. Equation (8) represents a constant matrix with each of its diagonal elements equal to 1/4. Obviously, each of the four eigenvalues of the PT of (8) is also equal to 1/4, i.e., greater than zero. Hence, according to the Peres-Horodecki condition [30,31], the DM (8) represents a mixed separable state of (e_1,e_2). (This particular form of a bipartite state has been called [46] a maximally chaotic state.) One of the simplest possible examples of this result can be the two-step DPI $\mathrm{B}(1s^2 2s^2 2p^1\ {}^2P)\to\mathrm{B}^{+^*}(1s^1 2s^2 2p^1\ {}^1P)$

$\rightarrow B^{++}(1s^2 2s^1 2p^0\ {}^2S)$ in the ground electronic configuration of a boron atom. The sequentially emitted photoelectron e_1 and the Auger electron e_2 form a separable spin state represented by the DM (8) in the absence of SOI.

(b) $S_0=S_f=0$ with $S_e=1/2$. It, in other words, means that while the electronic state of A and of A^{2+} is singlet, that of A^{+^*} is doublet. The DM, calculated from Eqs. (6) and (A15), can in this case be written as

$$4\sigma(S_0=0;S_e=1/2;S_f=0;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'}$$

$$\begin{array}{lllll}
\mu_1,\mu_2/\mu_1',\mu_2'\Rightarrow & \frac{1}{2},\frac{1}{2} & \frac{1}{2},-\frac{1}{2} & -\frac{1}{2},\frac{1}{2} & -\frac{1}{2},-\frac{1}{2} \\
\Downarrow & & & & \\
\frac{1}{2},\frac{1}{2} & 1-\hat{u}_1\cdot\hat{u}_2 & c_1s_2-s_1c_2c & s_1c_2-c_1s_2c & -s_1s_2+(1-c_1c_2)c \\
 & & -is_1s & +is_2s & -i(c_1-c_2)s \\
\frac{1}{2},-\frac{1}{2} & c_1s_2-s_1c_2c & 1+\hat{u}_1\cdot\hat{u}_2 & -s_1s_2-(1+c_1c_2)c & -s_1c_2+c_1s_2c \\
 & +is_1s & & +i(c_1+c_2)s & -is_2s \\
-\frac{1}{2},\frac{1}{2} & s_1c_2-c_1s_2c & -s_1s_2-(1+c_1c_2)c & 1+\hat{u}_1\cdot\hat{u}_2 & -c_1s_2+s_1c_2c \\
 & -is_2s & -i(c_1+c_2)s & & +is_1s \\
-\frac{1}{2},-\frac{1}{2} & -s_1s_2+(1-c_1c_2)c & -s_1c_2+c_1s_2c & -c_1s_2+s_1c_2c & 1-\hat{u}_1\cdot\hat{u}_2 \\
 & +i(c_1-c_2)s & +is_2s & -is_1s &
\end{array}$$

$$\equiv 4\sigma_1(\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'} \tag{9}$$

with the definitions

$$s\equiv\sin(\varphi_2-\varphi_1),\quad c\equiv\cos(\varphi_2-\varphi_1)$$

$$i\equiv\sqrt{(-1)},\quad \hat{u}_1\cdot\hat{u}_2=c_1c_2+s_1s_2c,$$

$$s_1\equiv\sin\vartheta_1,\quad s_2\equiv\sin\vartheta_2,\quad c_1\equiv\cos\vartheta_1,\quad c_2\equiv\cos\vartheta_2. \tag{10}$$

The PT of this DM (9) is readily obtained using either of the definitions (7).

We have calculated eigenvalues of (9) and of its PT (with respect to the Auger electron e_2). Both, (9) and its PT, were diagonalized using procedures explained in Refs. [50–52]. The eigenvalues obtained for (9) are (0, 0, 0, 1). The fact that only one of the four eigenvalues is nonzero means [25] that the DM (9) represents a pure spin state of (e_1,e_2). The eigenvalues of the PT of (9) came out to be $(\frac{1}{2}, \frac{1}{2}, -\frac{1}{2}, \frac{1}{2})$. These eigenvalues clearly mean [30,31] that the spin state of (e_1,e_2), represented by the DM (9), is entangled. In this case, each of the negativity [Eq. (3)] and concurrence [Eq. (4a)] is equal to 1. Furthermore, the degree of nonseparability (i.e., participation ratio or Schmidt number) [53] for this pure state is maximum, i.e., $K=2$, which is that of a Bell state [7]. In conclusion, the DM (9) represents a pure and maximally entangled bipartite spin-state of photoelectron e_1 and Auger electron e_2 emitted in the two consecutive steps of the process (1) in the absence of SOI.

Although, the elements both in the matrix (9) and in its PT obviously depend upon the spherical angles (ϑ_1,φ_1) and (ϑ_2,φ_2) of the respective spin quantization directions $\hat{u}_1$ of photoelectron e_1 and $\hat{u}_2$ of Auger electron e_2; it is, however, very remarkable that their eigenvalues are totally independent of these angles. This means, whatever may be the spin quantization directions $(\hat{u}_1,\hat{u}_2)$ [and also the propagation directions $(\hat{k}_1,\hat{k}_2)$], spin state of (e_1,e_2) always possesses the properties discussed in the preceding paragraph.

Second, Refs. [20,21] analyze the spin-entanglement of two electrons ejected in 1-DPI of an atom and of a rotating linear molecule, respectively, both in the absence as well as in the presence of spin-dependent interactions. The spin correlation DM obtained in Eq. (A12) in Ref. [20], or in Eq. (A10) in Ref. [21], in the absence of SOI, although identical to each other, are however very different from that given in Eq. (A15) in the present paper. But, each of these three density matrices reduces to an identical form on specializing the first two to the transition $S_0=S_f=0$ in Refs. [20,21] and the third to $S_0=S_f=0$ with $S_e=1/2$ in the present paper (compare Eqs. (9a) in Ref. [20] [for $\hat{u}_1(\vartheta_1=\pi/2,\varphi_1)$, $\hat{u}_2(\vartheta_2=\pi/2,\varphi_2)$], (A12a) in Ref. [21], and (9) herein). It has been discussed in detail in Refs. [20,21] that the two electrons ejected simultaneously in a $S_0=S_f=0$ transition must necessarily be in a singlet [i.e., $(|\frac{1}{2}\rangle_1|-\frac{1}{2}\rangle_2-|-\frac{1}{2}\rangle_1|\frac{1}{2}\rangle_2)/\sqrt{2}$] spin state. The identicalness of the present DM (9) to that of (9a) in Ref. [20] [for $\hat{u}_1(\vartheta_1=\pi/2,\varphi_1)$, $\hat{u}_2(\vartheta_2=\pi/2,\varphi_2)$] as well as to (A12a) in Ref. [21] simply means that the photoelectron e_1 and Auger electron e_2 also form a singlet spin state for the transition presently under consideration. Thus, $\sigma_1(\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2;\mu_1'\mu_2'}$ in (9) represents a bipartite singlet state of two spin-half qubits.

Among the several possible simple examples for this particular case are

$$\mathrm{C}(1s^22s^22p^2\;{}^1S/{}^1D)\to \mathrm{C}^{+^*}(1s^12s^22p^2\;{}^2S/{}^2D)\to \mathrm{C}^{2+}(1s^22s^22p^0\;{}^1S);\mathrm{O}(1s^22s^22p^4\;{}^1S/{}^1D)\to \mathrm{O}^{+^*}(1s^12s^22p^4\;{}^2S/{}^2D)$$
$$\to \mathrm{O}^{2+}(1s^22s^22p^2\;{}^1S/{}^1D);\mathrm{Ne}(1s^22s^22p^6\;{}^1S)\to \mathrm{Ne}^{+^*}(1s^12s^22p^6\;{}^2S)\to \mathrm{Ne}^{2+}(1s^22s^22p^4\;{}^1S/{}^2D).$$

Although, SOI in O and Ne atoms is probably not as negligible as in B and C atoms, nevertheless the photoelectron e_1 and Auger electron e_2 emitted in these transitions do form maximally entangled, pure singlet spin state in the absence of SOI.

(c) Let us now consider the process (1) for the remaining values of $S_0=S_f$. Namely, $S_0=S_f=S_e+1/2$ with $S_e>0$, or $S_0=S_f=S_e-1/2$ with $S_e>1/2$. The DMs for all these cases too are readily calculated from Eqs. (6) and (A15). All these DMs can easily be shown to be represented by the following simple equation:

$$\sigma_p(\hat{u}_1,\hat{u}_2)_{S_0=S_f}=p\left(S_0=S_f=S_e\pm\tfrac{1}{2};S_e\right)\sigma_1(\hat{u}_1,\hat{u}_2)+(1-p)\sigma_0 \tag{11}$$

with the parameter

$$p\left(S_0=S_f=S_e\pm\tfrac{1}{2};S_e\right)=\begin{cases}0 & \text{for } S_e=0,\\ \dfrac{1}{3S_e(S_e+1)}\left(\dfrac{3}{4}+S_e(S_e+1)-S_0(S_0+1)\right)^2 & \text{for } S_e>0.\end{cases} \tag{12}$$

and the respective matrices σ_0 and $\sigma_1(\hat{u}_1,\hat{u}_2)$ given by Eqs. (8) and (9). It is obvious from Eq. (12) that the parameter p is always positive, can assume zero value, and, on account of the restriction $|S_0-S_e|=1/2$, $p\le 1$. Eq. (12) further gives, $p(S_0=1/2;S_e=0;S_f=1/2)=0$ and $p(S_0=0;\ S_e=1/2;\ S_f=0)=1$. With these two allowed values of p, the DM (11) rightly reproduces the maximally chaotic state (8) for $p=0$ and the maximally entangled state (9) for $p=1$.

On using Refs. [50–52] once again for the diagonalization of a matrix, we find that

$$\tfrac{1}{4}(1-p),\tfrac{1}{4}(1-p),\tfrac{1}{4}(1-p),\tfrac{1}{4}(1+3p) \tag{13a}$$

are the eigenvalues of the DM (11). The condition that all the eigenvalues of a physically acceptable DM must necessarily be greater than or equal to zero, requires $-1/3\le p\le 1$. But, in the present case we find that, on account of Eq. (12), negative values for p are not admissible. Let us now look at the eigenvalues

$$\tfrac{1}{4}(1+p),\tfrac{1}{4}(1+p),\tfrac{1}{4}(1-3p),\tfrac{1}{4}(1+p) \tag{13b}$$

of the PT (with respect to the Auger electron e_2) of the DM (11). An application of the Peres-Horodecki [30,31] condition now immediately suggests that p must be greater than 1/3 for (11) to represent a nonseparable spin state.

Thus, for the DM (11) to represent a spin state of a photo-Auger-electron pair (e_1,e_2) emitted in 2-DPI with $S_0=S_f=S_e\pm 1/2$ and $S_e\ge 0$, in the absence of SOI, one must have $0\le p\le 1$. All these states are mixed. Those with $0\le p\le 1/3$ are separable; whereas, for an entangled pair of (e_1,e_2), $p>1/3$. For a given value of the triod (S_0,S_e,S_f), one can readily calculate from Eq. (12) the value of p, and hence find out the nature of the state (11). Some of the illustrative examples for this case are $\mathrm{N}(1s^22s^22p^3\;{}^4S)\to \mathrm{N}^{+^*}(1s^12s^22p^3\;{}^3S)\to \mathrm{N}^{2+}(1s^22s^12p^2\;{}^4P)$, with $p(S_0=3/2,S_f=3/2,S_e=1)=1/6$ obtained from Eq. (12). That is, (e_1,e_2) in this case are in a mixed separable state. On the other hand, the photoelectron e_1 and the Auger electron e_2 sequentially emitted in the transition $\mathrm{N}(1s^22s^22p^3\;{}^2P/{}^2D)\to \mathrm{N}^{+^*}(1s^12s^22p^3\;{}^3P/{}^3D)\to \mathrm{N}^{2+}(1s^22s^22p^1\;{}^2P)$ are represented by Eq. (11) corresponding to $p(S_0=S_f=1/2,S_e=1)=2/3$ which is a mixed entangled state of the two qubits.

In fact, for the two cases described by Eqs. (11) and (12), one can readily write that

$$p(S_e) = \begin{cases} \dfrac{S_e}{3(S_e+1)} & \text{for } S_0 = S_f = S_e + \dfrac{1}{2} \text{ and } S_e \geq 0, \quad (14a) \\ \dfrac{S_e+1}{3S_e} & \text{for } S_0 = S_f = S_e - \dfrac{1}{2} \text{ and } S_e \geq \dfrac{1}{2}. \quad (14b) \end{cases}$$

These simple relations clearly show that transitions (1) corresponding to (14a) always have $p < 1/3$, i.e., (e_1, e_2) are in a product spin-state; whereas, those satisfying (14b) are described by $p > 1/3$ and, hence, produce (e_1, e_2) in an entangled state.

Further, the negativity (3) of the states (11) is readily obtained from the eigenvalues (13b) of its partial transpose. However, in order to obtain the concurrence of this state, one needs to use the DM (11) in Eqs. (4). Both of these measure of the entanglement come out to be equal to each other in the present case and are given by

$$\mathcal{N}(\rho_f) = \mathcal{C}(\rho_f) = \begin{cases} \max(0, (3p-1)/2), & (15a) \\ 0 & \text{for } S_0 = S_f = S_e + 1/2 \text{ and } S_e \geq 0, \quad (15b) \\ (2S_e)^{-1} & \text{for } S_0 = S_f = S_e - 1/2 \text{ and } S_e \geq \tfrac{1}{2}, \quad (15c) \end{cases}$$

Substitution of the concurrence $\mathcal{C}(\rho_f)$, given in (15), in the expression (5) immediately gives us the entanglement of formation $\mathcal{E}_F(\rho_f)$ for the state (11).

Figure 2 shows variations of the mixing parameter p, negativity $\mathcal{N}(\rho_f)$, concurrence $\mathcal{C}(\rho_f)$, and of the entanglement of formation $\mathcal{E}_F(\rho_f)$ with the spin quantum number S_e of the photoion A^{+^*}. Both p and all the three different measures of entanglement have expected behavior with S_e. Namely, (e_1, e_2) with $S_0 = S_f = S_e + 1/2$ are always in separable states. These two electrons, on the other hand, always form an entangled state for $S_0 = S_f = S_e - 1/2$. Even in this case, however, the degree of entanglement decreases very rapidly with increasing S_e from its maximum possible value of unity for $S_e = 1/2$. Thus, states of (A, A^{+^*}, A^{2+}) with higher multiplicities produce (e_1, e_2) with lower degree of entanglement.

(ii) $|S_0(=|S_e \pm 1/2|) - S_f(=|S_e \mp 1/2|)| = 1$: In this case, the DM (A15) is readily shown to become

$$\sigma_p(\hat{u}_1, \hat{u}_2)_{|S_0 - S_f| = 1} = -\tfrac{1}{3}\sigma_1(\hat{u}_1, \hat{u}_2) + \tfrac{1}{3}\sigma_0. \quad (16)$$

The eigenvalues [50–52] of this matrix and of its PT are, respectively, (1/3, 1/3, 1/3, 0) and (1/6, 1/6, 1/2, 1/6). Thus (16) represents a mixed and separable state of the electrons (e_1, e_2). A relevant example for the present case can be

$$\mathrm{C}(1s^2 2s^2 2p^2\ {}^1S/{}^1D) \rightarrow \mathrm{C}^{+^*}(1s^1 2s^2 2p^2\ {}^2S/{}^2D) \rightarrow \mathrm{C}^{2+}(1s^2 2s^0 2p^2\ {}^3P)$$

.

It is obvious from the above discussions that Eqs. (11)–(16) completely characterize the properties of the quantum entanglement (QE), generated by the purely electrostatic Coulomb forces inside an atom, of any spin state of the photoelectron e_1 plus the Auger electron e_2 emitted from all possible electronic states $|A\rangle$, $|A^{+^*}\rangle$, $|A^{2+}\rangle$ participating in the two-step process (1) in the absence of SOI in the *L*-*S* coupling. These properties of this Coulombic spin-entanglement are totally decoupled form the photoionization and the Auger decay dynamics as well as kinematics, including properties of the ionizing radiation, and are completely predictable. They simply depend upon the spins of the three electronic states of the atomic species involved in this two-step process. Spin conservation conditions suggest that there are only three possibilities: (i) $|S_0 - S_f| = 1$ with $S_0 = S_e \pm 1/2$ and $S_f = S_e \mp 1/2$; (ii) $S_0 = S_f = S_e + 1/2$; (iii) $S_0 = S_f = S_e - 1/2$. In (i) and (ii), there is no entanglement at all; whereas, photo-Auger electrons are always entangled in (iii). In case (i), the spin state of(e_1, e_2) is always expressible as $-\sigma_1(\hat{u}_1, \hat{u}_2)/3 + \sigma_0/3$. But, in cases (ii) and (iii), $p\sigma_1(\hat{u}_1, \hat{u}_2) + (1-p)\sigma_0$ represents the spin state of (e_1, e_2). We have shown in this paper that $0 \leq p < 1/3$ given by $p = S_e/[3(S_e+1)]$, with $S_e \geq 0$ for (ii); whereas, $1/3 < p \leq 1$ obtained from $p = (S_e+1)/[3S_e]$, with $S_e \geq 1/2$ for (iii). There are no values of S_e which can give us $p = 1/3$. In arriving at these results, (A, A^{+^*}, A^{2+}) have been described in *L*-*S* coupling purely by the electrostatic Coulomb forces.

Bipartite states of qubits represented by $p\sigma_1(\hat{u}_1, \hat{u}_2) + (1-p)\sigma_0$, with $0 \leq p \leq 1$, are called Werner [54] states in QI. Obviously, one can always calculate values of p from the expressions derived herein. These expressions contain only the total spin S_e of A^{+^*}. According to the discussions given herein, cases (ii) and (iii) produce Werner [54] states. Our analysis further shows that a Werner state is entangled if and only if $p > 1/3$, which corresponds to our case (iii). However, the photo-Auger-electron pair in our case (i) comes out in a spin state represented by $-\sigma_1(\hat{u}_1, \hat{u}_2)/3 + \sigma_0/3$. Thus, Werner [54] states represent only a set of bipartite spin states formed by two electrons ejected in the process (1).

Further, case (iii) for the entanglement of (e_1, e_2) means that multiplicities of the states $|A\rangle$ and $|A^{++}\rangle$ must be equal and one less than that of $|A^{+^*}\rangle$ whose spin S_e should be different from zero. It has already been shown elsewhere in this paper that for $S_e = 1/2$ and $p = 1$, (e_1, e_2) are in the maximally entangled, singlet spin state. For this (i.e., $S_0 = S_f = 0, S_e = 1/2$) particular choice in case (iii), while each of $|A\rangle$ and $|A^{2+}\rangle$ is singlet, but $|A^{+^*}\rangle$ is a doublet state. As no SOI or any other interaction which can flip the spins of the departing electrons has presently been taken into account, the spin con-

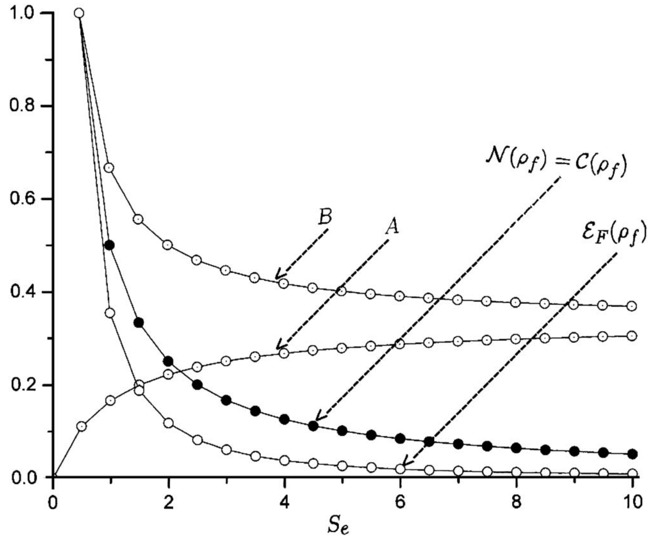

FIG. 2. Variations of the parameter p [Eqs. (14)], negativity $\mathcal{N}(\rho_f)$ [Eq. (3)], concurrence $\mathcal{C}(\rho_f)$ [Eq. (4a)], and of the entanglement of formation $\mathcal{E}(\rho_f)$ [Eqs. (5)] with the spin S_e of the excited photoion AB^{+^*} [Eqs. (1)]. As explained in Eqs. (15), here we have $\mathcal{N}(\rho_f)=\mathcal{C}(\rho_f)$ for all values of S_e considered in this paper. Curve A, p [from Eq. (14a)] $<$ 1/3, always; curve B, p [from Eq. (14b)] $>$ 1/3, always; curve C, negativity and concurrence from Eqs. (15); curve E, entanglement of formation from Eqs. (5) and (15).

servation conditions discussed in the preceding paragraphs then demand that e_1 in A and $e_2$in A^{+^*} should have opposite spins even before their emissions from the atom. However, such simple explanations for the spin orientations of ejected electrons while they were bound may not be applicable with other multiplicities of $|A\rangle$, $|A^{+^*}\rangle$, $|A^{2+}\rangle$ which will give values of $p<$ 1 with (e_1,e_2) in a mixture of maximally entangled (σ_1) and maximally chaotic (σ_0) states. For, in such situations $S_e>$ 1/2 and, hence, although $S_0=S_f$, none of A,A^{2+}, and (e_1,e_2) is in a singlet state, nor A^{+^*} is in a doublet state.

The other interesting and important thing about the Eqs. (11)–(16) is that one can *a priori* determine a value of p and, hence, select the appropriate photoionizing and Auger transitions in the process (1), according to one's requirements of the degree of mixing of the maximally chaotic state σ_0 with the pure, maximally entangled state $\sigma_1(\hat{u}_1,\hat{u}_2)$, in order to produce a spin-entangled state of (e_1,e_2) with a desired degree of entanglement. In other words, the Auger spectroscopy, described by Eq. (1), provides a simple and commonly available method for producing physically acceptable, all possible bipartite states of electrons with a tunable degree $p=(2S_e)^{-1}$ [with $S_0=S_f=S_e-1/2$, and $S_e\geq 1/2$] of their spin entanglement. Hence, spin S_e of the excited photoion A^{+^*} can be called also a tuning parameter for the degree of entanglement in the present context.

Using the expressions derived herein, one can always theoretically determine all properties of the spin state of a photo-Auger electron pair produced in 2-DPI in the absence of SOI. The experimental characterization of these states merely requires measurements of the energies of the photoelectron e_1 and of the Auger electron e_2, in order to determine $(|A\rangle,|A^{+^*}\rangle,|A^{2+}\rangle)$ participating in the process and, hence, calculate spins (S_0,S_e,S_f) from the multiplicities of each of these three electronic states in the L-S coupling. Use of any protocol, e.g., entanglement witness [31,42–45], etc., for the experimental characterization of the Coulombic entanglement of (e_1,e_2) is not needed at all. In addition, the present method can be used also as a test bed for the existing or new protocols which may be developed in the future for characterizing the entanglement properties of bipartite states of qubits.

IV. EINSTEIN-PODOLSKY-ROSEN-BOHM CORRELATION IN A PHOTO-AUGER-ELECTRON PAIR IN THE PRESENCE OF SPIN-ORBIT INTERACTION

A. Density matrix

On the inclusion of the spin-dependent forces in the process (1), the QE between (e_1,e_2) will be generated by a combination of the electrostatic Coulomb and spin-orbit interactions (CSOIs). In the presence of SOI, neither the total orbital nor total spin angular momenta is individually conserved in either of the two steps of the process (1). This renders the L-S coupling inapplicable in the presence of SOI [19]. Instead, it is now the sum of the total orbital and spin angular momenta which is separately conserved in each of the two steps of the process (1). The appropriate conservation conditions for total angular momenta valid for the present case are those given in the two equations (B1) in the Appendix B. This means, one now needs to work in j-j coupling scheme [19]. In Appendix B, we have, therefore, derived expressions in j-j coupling for the elements of our DO defined in Eq. (2b). Equation (B14) is the required DM for the present purpose. This expression is completely general and independent of all dynamical models which can be used in a calculational study of photo-Auger electron coincidence spectroscopy of atoms.

Each of the elements of the DM (B14a) contains some or all of the $A^{S_1S_2}_{N_{S_1}N_{S_2}}(m_r;\vec{k}_1,\vec{k}_2)$ defined in Eq. (B14b). The total number of such A's is 16. (Each of S_1 and S_2 can independently take two values, i.e., 0 and 1; whereas, $-S_1\leq N_{S_1}\leq S_1$ and $-S_2\leq N_{S_2}\leq S_2$.) For the purpose of the present and future applications, we have given in Eqs. (B16) the explicit expressions for the 16 elements constituting the DM (B14). The requirement that a physically acceptable DM must necessarily be Hermitian imposes conditions (B17) on the coefficients A's, defined in (B14b). This, obviously, means that only 10 of the 16 coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}$ are independent, including those four which are pure real. In view of (B17), one finds that the trace (B18) of our DM is always a pure real quantity.

Let us compare the DMs (A13) and (B14) obtained without and with SOI, respectively, in the process (1). Obviously, the two are completely different in many ways: (i) The present DM (B14) is much more complicated than the previous DM (A13). (ii) Unlike (A13), it is not at all possible to write (B14) as a product of two independent terms. That is, one cannot separate the DM (B14) into angular and spin parts. This, in other words, means that, unlike in the absence of SOI, both photoionization and Auger dynamics now play important roles in determining the spin-entanglement properties of the emitted electrons (e_1,e_2). Therefore, a knowledge of the frequency and polarization of the photon absorbed in (1a), of the linear momenta $(\vec{k}_1,\vec{k}_2)$ [i.e., both energies (ϵ_1,ϵ_2) and directions of motion $(\hat{k}_1,\hat{k}_2)$] of two emitted electrons, the $E1$ matrix element (B15a) for photoionization, as well as of the Auger emission amplitude (B15b), among others, is essential for studying the entanglement properties of a photo-Auger-electron pair in the presence of SOI. Thus, CSOIs have coupled the spin entanglement of (e_1,e_2) with the dynamics and the kinematics, including the properties of the absorbed photon, in (1). (iii) Consequently, unlike the DM (A13), it is not possible to analytically analyze (B14). Now, one cannot learn *a priori* anything at all about the entanglement of (e_1,e_2). A knowledge of both photoionization and of Auger amplitudes is necessary in order to be able to say anything about the properties of the spin state of (e_1,e_2) generated by the CSOIs. That is, one must study each atom individually with the specific transition one is interested in.

B. Example for photo-Auger-electron entanglement with spin-orbit interaction

Let us consider a particular example of photoionization

$$h\nu_r + \mathrm{Xe}(4d^{10}5s^25p^6\ {}^1S_0) \rightarrow \mathrm{Xe}^{+^*}(4d^95s^25p^6\ {}^2D_{5/2}) + e_1(p_{3/2};f_{5/2;7/2}) \qquad (17a)$$

followed by the Auger emission

$$\mathrm{Xe}^{+^*}(4d^95s^25p^6\ {}^2D_{5/2}) \rightarrow \mathrm{Xe}(4d^{10}5s^05p^6\ {}^1S_0) + e_2(d_{5/2}) \qquad (17b)$$

in Xe atom. We have selected this two step process for studying the effects of CSOIs on the spin-entanglement of (e_1,e_2) for several reasons:

(i) Kammerling *et al.* [55] have measured the average probabilities for the emission of a single Auger electron from each of the two hole states $4d_{3/2}$ and $4d_{5/2}$ of the excited photoion Xe^{+^*} to be 0.783 and 0.825, respectively. They [55] have further shown that these probabilities for the nonradiative, spontaneous decay of Xe^{+^*} depend little on the energy of the photon absorbed by Xe. Thus the probability for the emission of (e_1,e_2) in the above process in Xe very high.

(ii) It has recently been shown experimentally [56] that nonrelativistic description of photoionization of as heavy a target as Xe is both sufficient and satisfactory.

(iii) Kämmerling and Schmidt [57] have experimentally studied this two-step process and used their measurements to extract magnitudes and phases of the complex amplitudes for photoionization channels $4d_{5/2}\rightarrow p_{3/2}$, $4d_{5/2}\rightarrow f_{5/2}$, $4d_{5/2}\rightarrow f_{7/2}$. The experimentally extracted values [57] are in agreement with those calculated by Johnson and Cheng [58] in the relativistic random-phase approximation [59]. In the notation of our Eq. (B7c), the complex photoionization amplitudes taken from Ref. [57] and used in the present analysis are

$$\begin{aligned}
\langle J_e=5/2, j_1=3/2|F(J=1)|J_0=0,1\rangle &\equiv D_1 = d_1e^{i\Delta_1},\\
\langle J_e=5/2, j_1=5/2|F(J=1)|J_0=0,1\rangle &\equiv D_2 = d_2e^{i\Delta_2},\\
\langle J_e=5/2, j_1=7/2|F(J=1)|J_0=0,1\rangle &\equiv D_3 = d_3e^{i\Delta_3}
\end{aligned} \qquad (18a)$$

with

$$\begin{aligned}
d_1 &= 0.138\ \text{a.u.}, & &\\
d_2 &= -0.131\ \text{a.u.}, & \Delta_1-\Delta_2 &= 3.04\ \text{rad},\\
d_3 &= -0.474\ \text{a.u.}, & \Delta_2-\Delta_3 &= 1.35\ \text{rad},
\end{aligned} \qquad (18b)$$

where, a.u. stands for atomic units.

(iv) The Auger transition in (17b) is very simple as it is determined by a single (namely, $d-$) partial wave only.

(v) Both, Xe and Xe^{2+} being in their ${}^1S$ electronic state, SOI is always zero for these two species in the present case. However, SOI will very much be present in the ${}^2D$ electronic state of Xe^{+^*}, and affect photoionization as well as Auger emission in (17).

(vi) Both, the photoelectron in (17a) and the Auger electron in (17b) are represented by higher than s partial waves. SOI should, therefore, be present in the continua of both of these two ejected electrons as well.

(vii) Analysis presented in Sec. III herein shows that if one does not take the SOI into account in (17), then (e_1,e_2) are in the maximally entangled, pure, singlet spin state with each of negativity (3), concurrence (4a), and the entanglement of formation (5) equal to their maximum possible value, i.e., equal to 1 (see Fig. 2). Moreover, these properties of the Coulombic spin-entanglement of (e_1,e_2) are not affected at all by the respective dynamics of photoionization of Xe or of Auger emission from Xe^{+^*}, or by the momenta $(\vec{k}_1,\vec{k}_2)$, directions $(\hat{u}_1,\hat{u}_2)$ of spin quantization of (e_1,e_2), or by a change in the polarization of the ionizing radiation. It will, therefore, be very easy to detect even small deviations in the entanglement properties of (e_1,e_2) due to the inclusion of the SOI in the process (17).

Each of the photoelectron e_1 and the Auger electron e_2 can come out from their respective parents Xe and Xe^{+^*} in an infinite number of possible directions. Moreover, these directions of their emissions are quite independent of each other. Let us select diametric emission of (e_1,e_2), i.e., photoelectron and Auger electron are receding from the residual dication Xe^{2+} in (17) in opposite directions. In this collinear experimental geometry, if we take

$$\hat{k}_1(\theta_1,\phi_1) \parallel \hat{k}(\theta,\phi), \qquad (19)$$

obviously when

$$\hat{k}_2(\theta_2,\phi_2) \parallel -\hat{k}$$

with

$$(\theta_2 = \pi - \theta, \phi_2 = \pi + \phi).$$

Consequently, the coefficients A's, present in the DM (B14), will now depend upon (θ,ϕ) only. The present geometry for the emission of (e_1,e_2) also means that the polar axis of our PF and the line joining these two electrons always lie in one plane which will, henceforth, be called as the emission plane.

The spins of each of the photoelectron e_1 and the Auger electron e_2 can be quantized in all possible directions in space, quite independently of each other. But, entanglement is well known [7] to be independent of the local choice of the basis and coordinate system. Hence the choice of the spin quantization directions $(\hat{u}_1,\hat{u}_2)$ of (e_1,e_2) should not change entanglement between these two electrons. In the present study, we have considered, for simplicity, that each of the photoelectron and Auger electron has its spin quantized longitudinally to its respective direction of free motion in space. Namely,

$$\hat{u}_1 \| \hat{k}_1 \| \hat{k}, \text{ i.e., } (\vartheta_1 = \theta, \varphi_1 = \phi), \tag{20}$$

$$\hat{u}_2 \, \| \, \hat{k}_2 \, \| -\hat{k}, \text{ i.e., } (\vartheta_2 = \pi - \theta, \varphi_2 = \pi + \phi).$$

This, consequently, means that the final form of DM (B14) will contain only two angles specifying the direction $\hat{k}(\theta,\phi)$.

The last thing remaining to decide is the polarization of the ionizing photon which may be used for initiating the 2-DPI process (17). In order to investigate the effects of light polarization on the QE properties of (e_1,e_2), generated in the presence of CSOIs, we have considered four possibilities. Namely, the electromagnetic wave ionizing Xe is LP, CP with PH, CP with NH, or is UP. These are the most common polarizations of electromagnetic waves generally used in various experiments in a laboratory.

The following contains a brief description of our results for the entanglement properties of (e_1,e_2) emitted in the process (17) in the above-mentioned [i.e., (19)] possible experimental geometry of $(\hat{k}_1,\hat{k}_2)$ for each of the four polarizations of the incident light.

1. Incident light linearly polarized

In order to specialize the general expressions (B16) for the elements of the required DM to this case, we first need to know the 16 coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}(m_r;\vec{k}_1,\vec{k}_2)$ present therein. These are calculated from (B14b) by substituting m_r=0 for LP incident radiation, relation (19) for diametric emission of the photoelectron e_1 and Auger electron e_2, and amplitudes (18) for the photoionization process (17a). The resulting expressions are given in Eqs. (B19). These A's obviously satisfy the conditions (B17) needed for the resulting DM to be Hermitian. Thus, each of the 16 A's reduces to be a function of the angles (θ,ϕ) which define the orientation in space of the line joining e_1 and e_2 receding from the residual dication Xe^{2+} in opposite directions.

In order to obtain the required DM, one now merely needs to substitute in (B16) both angles (20) and coefficients (B19). Unless stated otherwise, we denote the resulting DM by $\rho_{\text{LP}}(\theta,\phi)$. It is then diagonalized using the procedures explained in Refs. [50–52]. Its eigenvalues are

$$0,0,0,$$

$$[19.532 - 15.4730(\sin\theta)^2 + \{16.4860(\sin\theta)^4 + 39.654(\sin 2\theta)^2 + 381.52(\cos\theta)^4\}^{1/2}] \times 10^{-4}. \tag{21a}$$

Thus, only one of the four eigenvalues in (21a) is nonzero. This, obviously, means [25] that $\rho_{\text{LP}}(\theta,\phi)$ represents a pure state of (e_1,e_2). The eigenvalues of the PT of $\rho_{\text{LP}}(\theta,\phi)$, on the other hand, are obtained to be

$$\begin{aligned}
&[4.0602(\sin\theta)^2 + 2.1609\sin 2\theta + 19.532(\cos\theta)^2] \times 10^{-4},\\
&[4.0602(\sin\theta)^2 - 2.1609\sin 2\theta + 19.532(\cos\theta)^2] \times 10^{-4},\\
&[16.4852(\sin\theta)^4 - 7.042(\sin 2\theta)^2 + 381.504(\cos\theta)^4]^{1/2} \times 10^{-4},\\
&-[16.4852(\sin\theta)^4 - 7.042(\sin 2\theta)^2 + 381.504(\cos\theta)^4]^{1/2} \times 10^{-4}.
\end{aligned} \tag{21b}$$

Here, the last of the four eigenvalues (21b) is always less than zero, for all allowed values (between 0 and π) of the polar angle θ. Hence, in view of the Peres-Horodecki [30,31] condition, $\rho_{\text{LP}}(\theta,\phi)$ represents an entangled state. One, therefore, concludes that (e_1,e_2), ejected in the 2-DPI (17) using LP light, are always in a pure entangled state.

Before proceeding further, it is probably appropriate to point out here that the eigenvalues (21) should be divided by the trace (B18) in order for them to belong to those of a normalized DM $\rho_{\text{LP}}(\theta,\phi)$ and its PT, respectively. After such a normalization, the eigenvalues (21a) will simply become (0, 0, 0, 1). Second, none of the eigenvalues (21) depend upon the azimuthal angle ϕ of the dimetric emission direction $\hat{k}(\theta,\phi)$. This, in other words, means that (e_1,e_2), emitted in 2-DPI (17) with LP light are always in a pure entangled state for all possible orientations of the emission plane. In the remaining discussion in the present Sec. IV B 1, we, therefore, do not further write the angle ϕ for brevity.

In order to study the degree of entanglement of (e_1,e_2), we have next calculated the negativity $\mathcal{N}[\rho_{\text{LP}}(\theta)]$ using (after normalization) the eigenvalues (21b) in Eq. (3); whereas,

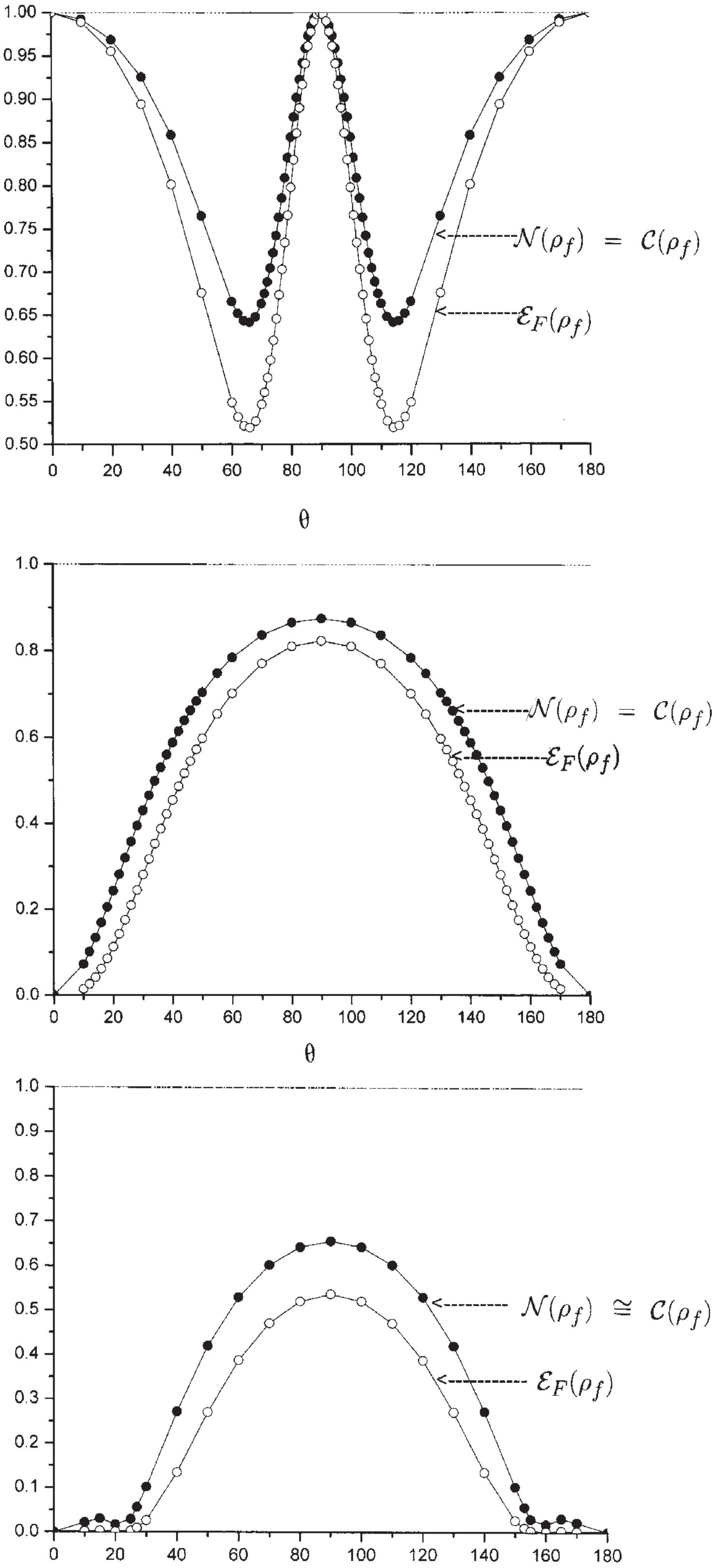

FIG. 3. Each of the three parts of this figure shows the values of the three measures of entanglement calculated in this paper for a photo-Auger-electron pair emitted from Xe in the 2-DPI process (17) in the presence of SOI. In each part, the upper, broken horizontal line represents these measures when no SOI is taken into account in (17). For this case, all the three measures have the same value which is equal to one corresponding to a maximally entangled state of (e_1, e_2). The negativity $\mathcal{N}(\rho_f)$, concurrence $\mathcal{C}(\rho_f)$, and entanglement of formation $\mathcal{E}_F(\rho_f)$ are calculated using the respective Eqs. (3), (4a), and (5). (a) All three measures in this figure are calculated when the ionizing radiation in (17a) is LP. On account of the pure nature of the bipartite state of (e_1, e_2), negativity and concurrence in this case have identical values. (b) This figure is for ionization in (17a) by a CP photon with NH. Here again, both negativity and concurrence have equal values. Also, as discussed in the text of this paper (see Sec. IV B 3), this figure represents values of all three measures of entanglement also for ionization in (17a) due to the absorption of a CP photon with PH. (c) This contains three measures of entanglement for ionizing radiation in (17a) to be UP. In this case, (e_1, e_2) were found to be in a mixed bipartite state. Consequently, negativity [Eq. (3)] and concurrence [Eq. (4a)] do not have the same values which are, however, indistinguishable on the scale of this figure.

concurrence $\mathcal{C}[\rho_{\mathrm{LP}}(\theta)]$ and entanglement of formation $\mathcal{E}_F[\rho_{\mathrm{LP}}(\theta)]$ are obtained from the respective Eqs. (4a) and (5) for the DM $\rho_{\mathrm{LP}}(\theta)$. In the present case, we always found [50(b)] that $\mathcal{N}[\rho_{\mathrm{LP}}(\theta)]=\mathcal{C}[\rho_{\mathrm{LP}}(\theta)]$, for all values of the angle θ.

Figure 3(a) shows our results for each of the measures $\mathcal{N}[\rho_{\mathrm{LP}}(\theta)]$, $\mathcal{C}[\rho_{\mathrm{LP}}(\theta)]$, and $\mathcal{E}_F[\rho_{\mathrm{LP}}(\theta)]$ as a function of the polar angle θ. {In view of Ref. [50(b)], curves for $\mathcal{N}[\rho_{\mathrm{LP}}(\theta)]$ and $\mathcal{C}[\rho_{\mathrm{LP}}(\theta)]$ are coincident in Fig. 3(a).} The broken horizontal line at the top of this figure represents these three measures for the Coulombic spin-entanglement of (e_1,e_2), i.e., when no SOI is taken into account in the 2-DPI (17). (This result is the one shown in Fig. 2 for the mixing parameter $p=1$.) Thus, while the introduction of SOI in (17) has no effect on the pure nature of the state of (e_1,e_2); however, its entanglement properties now very much depend upon the directions of motion of (e_1,e_2). All the three measures of entanglement, considered herein, have their maximum possible values of unity for the diametric emission of two electrons (e_1,e_2) either in the directions of the electric field vector of the LP ionizing radiation (i.e., $\theta=0,\pi$ in Fig. 1) or in a plane perpendicular to it (i.e., $\theta=\pi/2$). For other values of the polar angle θ, these measures vary significantly, their minimum value being for $\theta=65.50°$, where (e_1,e_2) are barely entangled. Thus, SOI tends to dilute the amount of entanglement between (e_1,e_2) making it highly anisotropic with respect to the directions of emission of two electrons. This entanglement is, otherwise, maximum in all directions of motion of (e_1,e_2) in the absence of SOI.

2. Incident light circularly polarized with negative helicity

Let us now study spin entanglement between (e_1,e_2) when incident light in (17) is CP with NH, i.e., left circularly polarized (LCP). For this, we first substitute in (B14b) the values of $(\hat{k}_1,\hat{k}_2)$ from Eq. (19) and $m_r=-1$, along with the quantities given in Eq. (18). The resulting values of each of the 16 coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}(-1;\theta,\phi)$ are given in Eq. (B20). These A's also satisfy the required Hermiticity conditions (B17).

Substitutions of the coefficients (B20) as well as of the angles (20) in Eqs. (B16) immediately give us the DM, say, $\rho_{\text{NH}}(\theta,\phi)$ describing the state of a photo-Auger-electron pair receding from Xe^{++} in opposite directions when the photon absorbed in (17) is LCP. A diagonalization of this DM with the processes [50–52] shows that three of its four eigenvalues are zero. This, in other words, means [25] that (e_1,e_2) are in a pure state. That is, whether one uses LP or LCP light for ionization in (17a), the sequentially emitted (e_1,e_2) are always in a pure spin state. Moreover, similar to the case of LP, the fourth, nonzero eigenvalue of the present DM was also a function only of the polar angle of $\hat{k}(\theta,\phi)$.

The three of the four eigenvalues of the PT of the current DM were always positive for $0\leq\theta\leq\pi$ but the fourth eigenvalue was greater than, or equal to, zero for $\theta=0$ and π only, remaining negative for other allowed values of this angle. Accordingly [30,31], the present photo-Auger-electron pair is in an entangled state for all directions of their diametrically opposite motions, except when they are moving parallel to the incident LCP light.

Next, we calculate [50(b)] each of the three measures of the entanglement defined in Eqs. (3)–(5). These, for the present case, are shown in Fig. 3(b). A comparison of Figs. 3(a) and 3(b) shows the effects of change of ionizing radiation from LP to LCP on the entanglement properties of (e_1,e_2) ejected in the 2-DPI (17). The curves in Fig. 3(b) for CSOIs are very different in each of their characteristics from those given in Fig. 3(a) for ionization by LP light when CSOIs is taken into account, or that obtained without SOI. Unlike in Fig. 3(a), (e_1,e_2) in the present case are never in a maximally entangled state: The maximum entanglement achieved by the photo-Auger-electron pair in the present case corresponds to concurrence $\mathcal{C}[\rho_{\text{NH}}(\theta)]\simeq0.90$ and entanglement of formation $\mathcal{E}_F[\rho_{\text{NH}}(\theta)]\simeq0.80$. Thus, the use of LCP radiation for ionization in (17a) with CSOIs tends to dilute the entanglement from those of its values with $m_r=0$, or without SOI for any polarization of the ionizing radiation. Similar to the case of ionization by LP light, the entanglement in Fig. 3(b) too varies significantly [but in a way which is very different from that in Fig. 3(a)] with the change in the directions of two oppositely moving (e_1,e_2).

3. Incident light circularly polarized with positive helicity

In order to investigate the effects on the entanglement of (e_1,e_2) of the PH of the CP light with the inclusion of CSOIs, the photon absorbed in (17a) is now right circularly polarized (RCP). Taking $m_r=+1$ in (B14b) [and substituting (18) plus (19)] gives us the 16 coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}(+1;\theta,\phi)$. The values of these A's are given in Eq. (B21). The DM, called $\rho_{\text{PH}}(\theta,\phi)$ for this case, is calculated by using in Eq. (B16) the coefficients (B21) and directions (20). The diagonalization [50–52] of $\rho_{\text{PH}}(\theta,\phi)$ and of its PT showed that this DM represents a pure state of (e_1,e_2) which is entangled everywhere except when the two electrons are ejected vertically in opposite directions. This behavior of the state of (e_1,e_2) is similar to that found in Sec. IV B 2 for ionization in (17) by LCP. We then calculated [50(b)] $\mathcal{N}[\rho_{\text{PH}}(\theta)]$, $\mathcal{C}[\rho_{\text{PH}}(\theta)]$, and $\mathcal{E}_F[\rho_{\text{PH}}(\theta)]$. The values of concurrence and of entanglement of formation obtained in the present case were found to be identical to those calculated in the Sec. IV B 2 for the DM $\rho_{\text{NH}}(\theta,\phi)$ obtained for ionization in (17) by LCP and shown in Fig. 3(b). This simply means that as far as entanglement properties of (e_1,e_2) are concerned, the helicity of the CP photon absorbed in (17) does not matter at all. That is, entanglement between a photoelectron and an Auger electron, sequentially emitted in 2-DPI (17), does not exhibit any dichroic properties.

4. Incident light unpolarized

Last, we investigate the entanglement between (e_1,e_2) when the ionizing radiation in (17) is UP and CSOIs are taken into account. An unpolarized electromagnetic wave can be looked upon to be an even mixture of LCP and RCP radiations. The corresponding DM, say, $\rho_{\text{UP}}(\theta,\phi)$ is now readily obtained on dividing by 2 the algebraic sum of the DMs $\rho_{\text{NH}}(\theta,\phi)$ and $\rho_{\text{PH}}(\theta,\phi)$ calculated in Secs. IV B 2 and IV B 3 for the same geometry.

On diagonalizing [50–52] the resulting DM $\rho_{\text{UP}}(\theta)$, we found that it has more than one eigenvalue to be nonzero. This means [25], electrons emitted in the 2-DPI when light used in (17) is UP, are not in a pure state. This result is totally opposite to those hitherto obtained in the present paper. Thus while (e_1,e_2) emitted in (17) without SOI for any polarization of the ionizing radiation or in the presence of SOI with LP, LCP or RCP photon, are always in a pure state, the same two electrons form a mixed spin state when the absorbed light is UP and the SOI is taken into account. Moreover, one of the four eigenvalues of the PT of this new DM $\rho_{\mathrm{UP}}(\theta)$ became negative in the range $\theta \simeq 25°$ to $\theta \simeq 155°$ only. That is, photoelectron and Auger electron are in a mixed spin-entangled state when the propagation direction $\hat{k}(\theta,\phi)$ for their diametric emission was lying in the above-specified range. We next calculated the $\mathcal{N}[\rho_{\mathrm{UP}}(\theta)]$, $\mathcal{C}[\rho_{\mathrm{UP}}(\theta)]$, and $\mathcal{E}_F[\rho_{\mathrm{UP}}(\theta)]$ for the present DM. These three measures are shown in Fig. 3(c). Unlike in the previous three cases, discussed in Secs. IV B 1, IV B 2, and IV B 3, now $\mathcal{N}[\rho_{\mathrm{UP}}(\theta)] \neq \mathcal{C}[\rho_{\mathrm{UP}}(\theta)]$ for the simple reason [50(b)] that $\rho_{\mathrm{UP}}(\theta)$ does not represent a pure state. However, the difference between these two measures is not visible on the scale of Fig. 3(c). On comparing Figs. 3(a)–3(c), one will obviously notice that the degree of entanglement is minimum when the light used in (17) is UP. The UP ionizing radiation in (17a) has reduced the degree of entanglement between (e_1,e_2) further than that it was diluted when the incident photon is CP. Also, the range of direction $\hat{k}(\theta,\phi)$ over which (e_1,e_2), receding from Xe^{2+}, are entangled in the present case is smaller than that found for ionization by LP, LCP, or RCP radiations.

V. CONCLUSIONS

Photoelectron and Auger electron, before their sequential emission, were part of the same atom experiencing various interactions with its other constituents and between themselves. Are these interactions experienced by the two electrons, plus their common fraternity, sufficient to entangle them while they are moving freely outside the atom? The present paper has attempted to theoretically investigate this question both without and with SOI taken into account.

When SOI is excluded, only the electrostatic Coulomb forces are present. Consequently, both total orbital and total spin angular momenta are individually conserved in each step of a 2-DPI process. These four conservation conditions completely separate the DM into its angular and spin parts. But, it is only the spin part of the DM which determines the QE properties of (e_1,e_2). These properties are found to be totally independent of all dynamics and kinematics. Instead, these are merely determined by the total spins of each of (A,A^{+^*},A^{2+}) participating in any 2-DPI. The spin conservation conditions suggest that there are only three possibilities. But, (e_1,e_2) are entangled if and only if the condition $S_0 = S_f = S_e - 1/2$, with the obvious requirement $S_e \geq 1/2$, is satisfied. The degree of entanglement in this case is simply given by $p = 1/(2S_e) > 1/3$, always. Thus, electrostatic Coulomb forces can produce EPR pairs of (e_1,e_2) possessing only discrete degrees of nonseparability given by the above expressions and decoupled from the dynamics as well as kinematics, including the properties of the ionizing radiation, of the process (1). The experimental determination of this Coulombic entanglement requires a knowledge of (S_0,S_e,S_f). This is readily obtained by measuring energies of the photoelectron e_1 and the Auger electron e_2 in the L-S coupling in order to identify the electronic states of (A,A^{+^*},A^{2+}) participating in 2-DPI. In the remaining two of the three above-mentioned possibilities, (e_1,e_2) are never entangled. In one of these three cases, (e_1,e_2) are neither in a Werner [54] state nor entangled. The expressions derived herein completely specify all properties of both Werner as well as non-Werner bipartite Coulombic spin-states of (e_1,e_2) produced in the 2-DPI process (1).

The spin-orbit energy is well known [60] to be a consequence of Dirac equation in relativistic quantum mechanics and is of the order of $(v/c)^2$ times the potential energy, where v is the speed of the particle. It is thus a small relativistic correction to the electrostatic Coulomb force inside an atom. Even this weak interaction is known to produce significant effects (e.g., fine structure splitting of energy levels [19], polarization of photoelectrons [61], etc.,) in atoms. Most of such effects are, however, present in single (or independent)-particle picture of an atom which, of course, get modified by the many-body interactions representing localized correlation. In this paper we have shown that this weak SOI is capable of significantly influencing even the physical phenomena like QE which not only does not have a single-particle analogue but also violates the principle of local realism [1].

In the presence of CSOIs, unlike in the case when SOI is not taken into account, only the sums of total orbital and total spin angular momenta are conserved in each of the two steps of 2-DPI. Thus, the number of conservation conditions for angular momenta is now reduced from four to two. Second, inclusion of SOI requires [19] that one should use the j-j coupling scheme for the angular momenta, rather than the Russell-Saunders coupling applicable in the absence of SOI. These changes render the DM to be nonseparable in angular and spin parts. Consequently, the QE properties of a photo-Auger-electron pair now heavily depend not only on both the dynamics and kinematics of 2-DPI, but also on the properties of the light used to ionize the atom. Now, one can no longer *a priori* determine the entanglement between (e_1,e_2) without first doing the dynamical calculations theoretically or using any of the protocols (e.g., entanglement witness [31,42–45], etc.) hitherto successfully used experimentally. The specific example considered herein show that the SOI reduces the degree of entanglement between (e_1,e_2) compared to that when this interaction is not taken into account. Entanglement now continuously changes both with the directions of motions of (e_1,e_2) and with the polarization of the absorbed photon, including, of course, the states of (A,A^{+^*},A^{2+}) participating in 2-DPI. In addition, changing the ionizing radiation from LP/CP to UP, converts a pure state to a mixed state of (e_1,e_2).

Thus, purely Coulomb interaction can produce (e_1,e_2) in spin-states possessing a degree of entanglement given by the analytical expression $1/2S_e$; it is otherwise totally independent of all other features of the process (1) and is completely predictable. In order to change the degree of this Coulombic spin-entanglement, one or more of the electronic states participating in the 2-DPI (1) needs to be changed. The inclusion of SOI, on the other hand, couples this entanglement with the dynamics and kinematics of the process (1), and with the properties of the absorbed photon. The degree of this entanglement varies continuously [even for the same transitions participating in 2-DPI (1)] and one can choose its value between zero (for a product state) to one (for a maximally entangled state) by a proper choice of the polarization of the ionizing radiation and of the direction of emission of (e_1, e_2). All mixed states of (e_1, e_2), generated without or with SOI, have degree of entanglement less than 1. A pure, Coulombic spin-state of (e_1, e_2) is always maximally entangled; whereas, the inclusion of SOI can give us a photo-Auger-electron pair in a pure state with its degree of entanglement varying from zero to one.

Attempts [62] are currently being made to theoretically develop relativistic QI theory based on the QE of particles with rest mass different from zero moving with speeds comparable to that of a photon. Harshman [63], citing mutually contradictory results of such previous studies, has argued, using relativistic both quantum mechanics and entangled bases, that spin entanglement of massive, relativistic particles depends on their linear momenta. The present analysis, on the other hand, is for a pair of electrons moving with speeds much less than that of light in the framework of the nonrelativistic quantum mechanics. It shows that QE properties of (e_1, e_2) will always depend, among other things, on these electrons' linear momenta whenever SOI is taken into account; otherwise, it is totally unaffected by these and other such physical quantities.

Cinelli *et al.* [64] showed that two photons produced in spontaneous parametric down conversion (SPDC) in a nonlinear optical crystal are polarization-entangled if they are emitted only in certain directions. This spatial characteristics of entangled photon has subsequently been used [65] to engineer polarization-momentum hyperentangled two-photon states. This property, where the polarization and momentum degrees of freedom each store one copy of the bipartite photon-state produced in SPDC, has recently been used in an experiment [66] to directly determine one of the several entanglement measures, namely concurrence [36–38], with a single, local measurement on just one photon. These developments suggest that, analogous to the experiments [64,65] with photons, it should be possible to engineer the spin-momentum entanglement between two spin-entangled electrons produced by 2-DPI in the presence of SOI. This raises the subsequent possibility of experimental determination of the spin-entanglement of a bipartite electronic state with a single measurement, similar to the one done in Ref. [66] for photons. This will make the gedanken experiment, originally performed on electrons by Bohm [2] on EPR [1] proposition, to carry out in a laboratory.

Finally, it is probably obvious from the discussions presented herein that whenever there is entanglement between the sequentially emitted photoelectron e_1 and the Auger electron e_2, without or with SOI, it is due completely as well as exclusively to the internal forces and/or the electronic states of (A, A^{+^*}, A^{2+}) participating in the process (1). External conditions and/or laboratory environments have little role to play in it. Also, generation of the suggested entanglement does not require any special external preparations, whatsoever, which may lead to the coupling of the spins of (e_1, e_2) with the environment out side the target or other sources of decoherence and/or dissipation. Consequentially, the present entanglement is least prone to the harmful effects of decoherence and dissipation, usually produced by external conditions.

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APPENDIX A: DENSITY MATRIX FOR THE ANGLE- AND SPIN-RESOLVED, SEQUENTIAL EMISSION OF A PHOTOELECTRON AND AN AUGER ELECTRON FROM AN ATOM WITHOUT TAKING SPIN-ORBIT INTERACTION INTO ACCOUNT

This appendix contains a derivation of the DM used in the discussion in Sec. III of this paper. In order to analyze the Einstein-Podolsky-Rosen-Bohm correlation (i.e., spin-entanglement) of the bipartite state formed by the photoelectron e_1 and the Auger electron e_2, it is necessary that our DM for the two-step process (1) be both angle and spin resolved. In the following derivation, we do not take SOI into account not only in none of the bound electronic states $|A\rangle$, $|A^{+^*}\rangle$, and $|A^{2+}\rangle$, but also in the continuum of either of the two ejected electrons (e_1, e_2). Only the electrostatic Coulomb forces for particles participating in 2-DPI process (1) are thus included. In addition, neither the photoionization operator F_p in the $E1$ approximation, nor the Auger emission operator F_a, depends upon any spin variables. In such situations, L-S coupling [19] becomes applicable. Consequently, each of the orbital angular momenta and spin angular momenta are individually conserved in step (1a) as well in the step (1b) of the process(1).

We, therefore, have

$$\vec{\ell}_r + \vec{L}_0 = \vec{L}_e + \vec{\ell}_1 \tag{A1a}$$

and

$$\vec{L}_e = \vec{L}_f + \vec{\ell}_2 \tag{A2a}$$

for the conservation of the orbital angular momenta in the respective two steps of (1). Here, $(\vec{L}_0, \vec{L}_e, \vec{L}_f)$ are the total orbital angular momenta of (A, A^{+^*}, A^{2+}), respectively; whereas, $\vec{\ell}_1$ and $\vec{\ell}_2$ are those of the respective photoelectron e_1 and Auger electron e_2. Similarly, for the conservation of the spin angular momenta we have

$$\vec{S}_0 = \vec{S}_e + \vec{s}_1 \left[= \left(\frac{\vec{1}}{2} \right)_1 \right] \tag{A1b}$$

and

$$\vec{S}_e = \vec{S}_f + \vec{s}_2 \left[= \left(\frac{\vec{1}}{2} \right)_2 \right]. \tag{A2b}$$

The quantities $(|\vec{S}_0|, |\vec{S}_e|, |\vec{S}_f|, |\vec{s}_1|, |\vec{s}_2|)$ in the above two equations represent, respectively, total spins of each of the species $(A, A^{+^*}, A^{2+}, e_1, e_2)$ participating in the Auger emission process (1). Further, we use the symbols

$M_{L_0}, M_{L_e}, M_{L_f}, M_{S_0}, M_{S_e}, M_{S_f}$ for representing, along our quantization axis (specified in Fig. 1), projections of the corresponding angular momenta defined in Eqs. (A1) and (A2). One can now readily write [19] $|A\rangle = |\vec{L}_0\vec{S}_0 M_{L_0} M_{S_0}\rangle, |A^{+^*}\rangle = |\vec{L}_e\vec{S}_e M_{L_e} M_{S_e}\rangle$, and $|A^{2+}\rangle = |\vec{L}_f\vec{S}_f M_{L_f} M_{S_f}\rangle$ for the electronic states of A, A^{+^*}, and A^{2+}, respectively, in the L-S coupling.

In the present as well as in the following appendix, $|\mu_1\hat{u}_1\vec{k}_1\rangle^-$ and $|\mu_2\hat{u}_2\vec{k}_2\rangle^-$ are the continuum spin orbitals of the photoelectron e_1 and Auger electron e_2, respectively. A minus superscript on these means that each of the continuum orbitals of e_1 and of e_2 individually satisfies asymptotic incoming wave boundary conditions [67] appropriate for photoionization as well as for Auger emission.

In order to calculate the desired DM, we first need to know the DO ρ_i present in Eqs. (2). It, for a non-interacting system of an unpolarized atom (in the L-S coupling) plus a polarized photon, is given by (see, for example Ref. [20])

$$\rho_i = \rho_0 \otimes \rho_r = \frac{1}{(2L_0+1)(2S_0+1)} \sum_{M_{L_0} M_{S_0}} |0;1m_r\rangle\langle 0;1m_r|, \tag{A3}$$

where we have defined $|0;1m_r\rangle \equiv |0\rangle|1m_r\rangle$. [In various equations in this and the following appendix, we have interchangeably used letters $(0,e,f)$ for $A, A^{+^*}, A^{2+})$ inside Dirac's bra and ket notations, for brevity.] An element (which is diagonal in $\hat{u}_i$ and $\vec{k}_i$, but nondiagonal in μ_i, with $i=1$, 2) of the DO defined in Eq. (2b) is

$$\begin{aligned}\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle &= \mathcal{K}\sum_{M_{L_f} M_{S_f}} \langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|F_a F_p \rho_i F_p^\dagger F_a^\dagger \\ &\quad \times |f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle.\end{aligned} \tag{A4}$$

In this expression we have summed over all the degenerate states $|A^{2+}\rangle$ of A^{2+} in the L-S coupling. On substituting (A3) and using the completeness of the states $|A^{+^*}\rangle$ of A^{+^*}, the above DM takes the following form:

$$\begin{aligned}&\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle \\ &= \frac{\mathcal{K}}{(2L_0+1)(2S_0+1)} \sum_{\substack{M_{L_0}M_{L_e}M_{L_e}'M_{L_f} \\ M_{S_0}M_{S_e}M_{S_e}'M_{S_f}}} \langle f;\mu_2\hat{u}_2\vec{k}_2|F_a|e\rangle \\ &\quad \times \langle e;\mu_1\hat{u}_1\vec{k}_1|F_p|0;1m_r\rangle\langle 0;1m_r|F_p^\dagger|e';\mu_1'\hat{u}_1\vec{k}_1\rangle \\ &\quad \times \langle e'|F_a^\dagger|f;\mu_2'\hat{u}_2\vec{k}_2\rangle.\end{aligned} \tag{A5}$$

Here, $|e\rangle$ and $|e'\rangle$ represent degenerate states of the excited photoion A^{+^*}, i.e., $|e\rangle = |\vec{L}_e\vec{S}_e M_{L_e} M_{S_e}\rangle$ and $|e'\rangle = |\vec{L}_e\vec{S}_e M_{L_e}' M_{S_e}'\rangle$. Further in (A5), $|e;\mu_1\hat{u}_1\vec{k}_1\rangle = \mathcal{A}(|A^{+^*}\rangle|\mu_1\hat{u}_1\vec{k}_1\rangle^-)$ is an antisymmetrized state of the excited photoion A^{+^*} and of the photoelectron e_1, with $\mathcal{A}$ an antisymmetrization operator. Likewise, $|f;\mu_2\hat{u}_2\vec{k}_2\rangle = \mathcal{A}(|A^{2+}\rangle|\mu_2\hat{u}_2\vec{k}_2\rangle^-)$ is an antisymmetrized state of the dication A^{2+} and of the Auger electron e_2.

Expression (A5) represents a four-dimensional matrix for a 2×2 system which contains, among other things, complete information on photoelectron e_1 and Auger electron e_2 ejected sequentially in the two-step process (1) taking place in the L-S coupling in the absence of SOI. The nondiagonal $(\mu_1' \neq \mu_1, \mu_2' \neq \mu_2)$ elements in (A5) represent the coherent effects; whereas, diagonal $(\mu_1' = \mu_1, \mu_2' = \mu_2)$ elements describe angular distribution of spin-resolved e_1 and e_2. For diagonal elements, (A5) simplifies to a form identical to that given elsewhere ([22,28], see, for example, Eq. (14.4) in Ref. [22]). It is obvious that the DM (A5) is Hermitian, i.e.,

$$\begin{aligned}&\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle \\ &\quad = \langle f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2\rangle^*.\end{aligned} \tag{A6}$$

The next task for us is to evaluate the matrix elements of the $E1$ photoionization operator F_p and of the Auger emission operator F_a occurring on the right-hand side of (A5). In order to calculate the matrix elements of F_p, one needs to introduce the couplings suggested in Eq. (A1a) for the orbital and in (A1b) for the spin angular momenta for photoionization (1a) in L-S coupling. We, therefore, have for the couplings expressed on the left-hand side of Eqs. (A1a),

$$\begin{aligned}|0;1m_r\rangle &= \sum_{LM_L} (-1)^{1-L_0-M_L}\sqrt{2L+1}\begin{pmatrix} L_0 & 1 & L \\ M_{L_0} & m_r & -M_L \end{pmatrix} \\ &\quad \times |(L_0 1)LM_L;S_0M_{S_0}\rangle.\end{aligned} \tag{A7}$$

Here, $\begin{pmatrix} .. & .. & .. \\ .. & .. & .. \end{pmatrix}$ is a 3-j symbol [49]. Similarly, angular momentum couplings shown on the right-hand side of Eqs. (A1) give

$$\begin{aligned}&|e;\mu_1\hat{u}_1\vec{k}_1\rangle \\ &\quad = (-1)^{(1/2)-L_e-S_e} \sum_{\ell_1 m_1 \nu_1} \sum_{\substack{LM_L \\ SM_S}} i^{\ell_1}(-1)^{\ell_1-M_L-M_S} e^{-i\sigma_{\ell_1}} \\ &\quad\quad \times \sqrt{(2L+1)(2S+1)}\begin{pmatrix} L_e & \ell_1 & L \\ M_{L_e} & m_1 & -M_L \end{pmatrix} \\ &\quad\quad \times \begin{pmatrix} S_e & \frac{1}{2} & S \\ M_{S_e} & \nu_1 & -M_S \end{pmatrix} \times [Y_{\ell_1}^{m_1}(\hat{k}_1)]^* \\ &\quad\quad \times [\mathcal{D}_{\mu_1\nu_1}^{1/2}(\omega_1)]^* \left|(L_e\ell_1)LM_L;\left(S_e\frac{1}{2}\right)SM_S\right\rangle^-.\end{aligned} \tag{A8}$$

In Eq. (A8), σ_{ℓ_1} is the Coulomb phase [26] for the ℓ_1th partial wave of the photoelectron e_1; $\mathcal{D}$ are the well-known rotational harmonics [49] with $\omega_1(\varphi_1, \vartheta_1, 0)$ the Euler angles which rotate the polar axis of the space-frame (shown in Fig. 1) into the spin-polarization direction $\hat{u}_1$. Further in (A8), the coupled state $|(L_e\ell_1)LM_L;(S_e\frac{1}{2})SM_S\rangle^-$ represents the excited photoion A^{+^*} in its $|A^{+^*}\rangle$ electronic state and the photoelectron e_1 with its respective orbital and spin angular momenta $\vec{\ell}_1$and $\vec{s}_1$ [see, Eqs. (A1)]. The $E1$ photoionization matrix element needed in the DM (A5) is now readily calculated to be

$$\begin{aligned}\mathcal{P}_e &\equiv \langle e;\mu_1\hat{u}_1\vec{k}_1|F_p|0;1m_r\rangle\\ &= (-1)^{\frac{1}{2}+L_0+L_e+S_e+M_{S_0}}\sqrt{2S_0+1}\sum_{\substack{\ell_1 m_1 \nu_1\\ LM_L}}(-i)^{\ell_1}(-1)^{\ell_1}e^{i\sigma_{\ell_1}}\\ &\quad\times(2L+1)\begin{pmatrix}L_e & \ell_1 & L\\ M_{L_e} & m_1 & -M_L\end{pmatrix}\begin{pmatrix}L_0 & 1 & L\\ M_{L_0} & m_r & -M_L\end{pmatrix}\\ &\quad\times\begin{pmatrix}S_e & \frac{1}{2} & S_0\\ M_{S_e} & \nu_1 & -M_{S_0}\end{pmatrix}Y_{\ell_1}^{m_1}(\hat{k}_1)\mathcal{D}^{1/2}_{\mu_1\nu_1}(\omega_1)\\ &\quad\times\langle(L_e\ell_1)L|F_p|(L_0 1)L\rangle. \end{aligned} \tag{A9}$$

In arriving at this result, use has been made of the conservation conditions (A1). Although, not explicitly specified, the bracket $\langle(L_e\ell_1)L|F_p|(L_0 1)L\rangle$ in the above equation depends upon the multiplicities $(2S_0+1)$ and $(2S_e+1)$ of the eigenstates of A and A^{+*}, respectively, in the L-S coupling. This, in turn, affects the energy $\epsilon_1 = h\nu_r - (E_e - E_0)$ available to the photoelectron e_1.

For calculating the Auger emission amplitudes present in (A5), one needs

$$\begin{aligned}|f;\mu_2\hat{u}_2\vec{k}_2\rangle &= (-1)^{1/2}\sum_{\substack{\ell_2 m_2\nu_2\\ AaBb}} i^{\ell_2}(-1)^{\ell_2-L_f-S_f-a-b}\\ &\quad\times e^{-i\sigma_{\ell_2}}\sqrt{(2A+1)(2B+1)}\times\begin{pmatrix}L_f & \ell_2 & A\\ M_{L_f} & m_2 & -a\end{pmatrix}\\ &\quad\times\begin{pmatrix}S_f & \frac{1}{2} & B\\ M_{S_f} & \nu_2 & -b\end{pmatrix}\\ &\quad\times[Y_{\ell_2}^{m_2}(\hat{k}_2)]^*[\mathcal{D}^{1/2}_{\mu_2\nu_2}(\omega_2)]^*\left|(L_f\ell_2)Aa;\left(S_f\frac{1}{2}\right)Bb\right\rangle^{-}\end{aligned} \tag{A10}$$

to represent the $(A^{2+}+e_2)$ system. Here, σ_{ℓ_2} is the Coulomb phase for the ℓ_2th partial wave of the Auger electron e_2; $\omega_2(\varphi_2,\vartheta_2,0)$ are the Euler angles which rotate the polar axis of the space frame (shown in Fig. 1) into the spin-polarization direction $\hat{u}_2$ of e_2. Further in (A10), $|(L_f\ell_2)Aa;(S_f\frac{1}{2})Bb\rangle^{-}$ is a state of the dication A^{2+} and Auger electron e_2 coupled according to the scheme expressed in Eqs. (A2). The Auger decay matrix element, needed in the DM (A5), is now readily calculated to be

$$\begin{aligned}\mathcal{A}_e &\equiv \langle f;\mu_2\hat{u}_2\vec{k}_2|F_a|A^{+^*}\rangle\\ &= (-1)^{-(1/2)+L_f+S_f+M_{L_e}+M_{S_e}}\sqrt{(2L_e+1)(2S_e+1)}\\ &\quad\times\sum_{\ell_2 m_2\nu_2}(-i)^{\ell_2}(-1)^{-\ell_2}e^{i\sigma_{\ell_2}}\begin{pmatrix}L_f & \ell_2 & L_e\\ M_{L_f} & m_2 & -M_{L_e}\end{pmatrix}\\ &\quad\times\begin{pmatrix}S_f & \frac{1}{2} & S_e\\ M_{S_f} & \nu_2 & -M_{S_e}\end{pmatrix}Y_{\ell_2}^{m_2}(\hat{k}_2)\mathcal{D}^{1/2}_{\mu_2\nu_2}(\omega_2)\\ &\quad\times\langle(L_f\ell_2)L_e|F_a|L_e\rangle\end{aligned} \tag{A11}$$

The derivation of (A11) depends on the application of the angular momentum conservation conditions (A2); the bracket $\langle(L_f\ell_2)L_e|F_a|L_e\rangle$ in the above equation implicitly involves the respective multiplicities $(2S_e+1)$ and $(2S_f+1)$ of the eigenstates of A^{+^*} and A^{2+} in the L-S coupling which determines the energy $\epsilon_2 = E_e - E_f$ of the Auger electron e_2.

Next, we need to calculate the DM (A5). This is obtained by substituting in (A5), the $E1$ photoionization and Auger decay amplitudes $\mathcal{P}_e$ and $\mathcal{A}_e$, respectively. The simplification of the consequent expression for the DM involves a heavy use of Racah algebra. In order to make this process easier, we first calculate the following two expressions separately:

$$\begin{aligned}&\frac{1}{2S_0+1}\sum_{M_{L_0}\,M_{S_0}}\mathcal{P}_e(\mathcal{P}'_e)^\dagger\\ &\quad\equiv\frac{1}{2S_0+1}\sum_{M_{L_0}\,M_{S_0}}\langle e;\mu_1\hat{u}_1\vec{k}_1|F_p|0;1m_r\rangle\\ &\qquad\times\langle 0;1m_r|F_p^\dagger|e';\mu'_1\hat{u}_1\vec{k}_1\rangle\end{aligned} \tag{A12a}$$

and

$$\sum_{M_{L_f}\,M_{S_f}}\mathcal{A}_e(\mathcal{A}'_e)^\dagger\equiv\sum_{M_{L_f}\,M_{S_f}}\langle f;\mu_2\hat{u}_2\vec{k}_2|F_a|e\rangle\langle e'|F_a^\dagger|f;\mu'_2\hat{u}_2\vec{k}_2\rangle. \tag{A12b}$$

Evaluation of (A12a) involves substitution of (A9) and of its Hermitian conjugate, single use of the identity (6.2.8) from Edmonds [49], and some other minor simplifications; whereas, calculation of (A12b) requires substitution of (A11) and of its Hermitian conjugate, application of the identity (6.2.8) two times, including other simplifications.

The final, simplified forms of (A12a) and of (A12b) thus obtained are then substituted in the DM (A5) and the remaining sums over $(M_{L_e}, M'_{L_e}, M_{S_e}, M'_{S_e})$ are evaluated using identities (14.42), (3.7.8), and (6.2.8). The first identity is taken from de Shalit and Talmi [68], while the last two are given in Ref. [49]. These and some other simplifications help us in writing the final expression for the DM in the following product of two independent terms:

$$\begin{aligned}&\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\,\hat{u}_2\,\vec{k}_2|\rho_f|f;\mu'_1\,\hat{u}_1\,\vec{k}_1;\mu'_2\,\hat{u}_2\,\vec{k}_2\rangle\\ &\quad=\frac{d^3\sigma(m_r)}{d\epsilon_1\mathrm{d}\hat{k}_1\mathrm{d}\hat{k}_2}\sigma(S_0;S_e;S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu'_1\mu'_2}.\end{aligned} \tag{A13}$$

Here,

$$\frac{d^3\sigma(m_r)}{d\epsilon_1 d\hat{k}_1 d\hat{k}_2} = (-1)^{m_r+L_0+L_e+L_f}\frac{(2L_e+1)\mathcal{K}}{4\pi(2L_0+1)} \sum_{\substack{\ell_1\ell_1'L_1LL' \\ \ell_2\ell_2'L_2ML_r}} (-1)^{\ell_1'+L+L_2}(2L_r+1)\sqrt{(2L_1+1)(2L_2+1)}\begin{pmatrix} \ell_1 & \ell_1' & L_1 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} \ell_2 & \ell_2' & L_2 \\ 0 & 0 & 0 \end{pmatrix}$$
$$\times\begin{pmatrix} 1 & 1 & L_r \\ m_r & -m_r & 0 \end{pmatrix}\begin{pmatrix} L_2 & L_1 & L_r \\ M & -M & 0 \end{pmatrix}\times\begin{Bmatrix} 1 & 1 & L_r \\ L & L' & L_0 \end{Bmatrix}\begin{Bmatrix} \ell_2 & \ell_2' & L_2 \\ L_e & L_e & L_f \end{Bmatrix}\begin{Bmatrix} \ell_1 & \ell_1' & L_1 \\ L & L' & L_r \\ L_e & L_e & L_2 \end{Bmatrix}$$
$$\times[Y_{L_1}^{-M}(\hat{k}_1)]^*[Y_{L_2}^{M}(\hat{k}_2)]^* P_e(L_e\ell_1;L_01;L)[P_e(L_e\ell_1';L_01;L')]^* A_e(L_f\ell_2;L_e;L_e)[A_e(L_f\ell_2';L_e;L_e)]^* \tag{A14a}$$

with

$$P_e = (-i)^{\ell_1}(-1)^{\ell_1}e^{i\sigma_{\ell_1}}(2L+1)\sqrt{2\ell_1+1}\langle(L_e\ell_1)L|F_p|(L_01)L\rangle \tag{A14b}$$

containing the $E1$ photoionization amplitude $\langle(L_e\ell_1)L|F_p|(L_01)L\rangle$ and

$$A_e = (-i)^{\ell_2}(-1)^{\ell_2}e^{i\sigma_{\ell_2}}\sqrt{2\ell_2+1}\langle(L_f\ell_2)L_e|F_a|L_e\rangle \tag{A14c}$$

depending upon the Auger decay amplitude $\langle(L_f\ell_2)L_e|F_a|L_e\rangle$.

On the other hand, the second term on the right-hand side of (A13) is given by

$$\sigma(S_0,S_e,S_f;\hat{u}_1,\hat{u}_2)_{\mu_1\mu_2,\mu_1'\mu_2'} = (-1)^{S_0+S_f-2S_e+\mu_1'+\mu_2'}(2S_e+1)\sum_{snm_1m_2}(-1)^{s+n}(2s+1)\begin{pmatrix} 1/2 & 1/2 & s \\ \mu_1 & -\mu_1' & m_1 \end{pmatrix}\begin{pmatrix} 1/2 & 1/2 & s \\ \mu_2 & -\mu_2' & m_2 \end{pmatrix}$$
$$\times\begin{Bmatrix} 1/2 & 1/2 & s \\ S_e & S_e & S_0 \end{Bmatrix}\begin{Bmatrix} 1/2 & 1/2 & s \\ S_e & S_e & S_f \end{Bmatrix}[\mathcal{D}^s_{m_1,n}(\varphi_1,\vartheta_1,0)]^*[\mathcal{D}^s_{m_2,-n}(\varphi_2,\vartheta_2,0)]^*. \tag{A15}$$

Here, $\begin{Bmatrix} .. & .. & .. \\ .. & .. & .. \end{Bmatrix}$ and $\begin{Bmatrix} .. & .. & .. \\ .. & .. & .. \\ .. & .. & .. \end{Bmatrix}$ are 6-j and 9-j symbols [49], respectively.

APPENDIX B: DENSITY MATRIX FOR THE ANGLE- AND SPIN-RESOLVED, SEQUENTIAL EMISSION OF A PHOTOELECTRON AND AN AUGER ELECTRON FROM AN ATOM TAKING SPIN-ORBIT INTERACTION INTO ACCOUNT

In the following, we present details of the derivation of the DM used in Sec. IV for studying spin entanglement between a photoelectron e_1 and an Auger electron e_2 emitted when both electrostatic Coulomb as well as spin-orbit interactions in each of the two steps of the process (1) are fully taken into account. [However, both the $E1$ operator for the photoionization step (1a) and the interelectronic interaction ejecting the Auger electron in (1b) are still independent of the spins.] In such a physical situation, neither the orbital nor the spin angular momentum will individually be conserved in any of the steps in (1), i.e., L-S coupling will not be applicable [19]. Hence, one cannot use either of the Eqs. (A1) or (A2). On the other hand, the total angular momenta will now be conserved [19]. These are given [19] by

$$\vec{\ell}_r + \vec{J}_0 = \vec{J}_e + \vec{j}_1\left\{=\vec{\ell}_1 + \vec{s}_1\left[=\left(\tfrac{\vec{1}}{2}\right)_1\right]\right\} \tag{B1a}$$

and

$$\vec{J}_e = \vec{J}_f + \vec{j}_2\left\{=\vec{\ell}_2 + \vec{s}_2\left[=\left(\tfrac{\vec{1}}{2}\right)_2\right]\right\} \tag{B1b}$$

for the two respective steps of the process (1). Here, $\vec{J}_0 = \vec{L}_0 + \vec{S}_0$, $\vec{J}_e = \vec{L}_e + \vec{S}_e$, and $\vec{J}_f = \vec{L}_f + \vec{S}_f$ are the total angular momenta of A, A^{+*}, and of A^{2+}, respectively. Let us represent by (M_0, M_e, M_f) the projections of $(\vec{J}_0, \vec{J}_e, \vec{J}_f)$ along our space quantization axis (see Fig. 1). Then Ref. [19], $|A\rangle = |J_0M_0\rangle$, $|A^{+*}\rangle = |J_eM_e\rangle$, and $|A^{2+}\rangle = |J_fM_f\rangle$ are the respective electronic states of each of the three atomic species participating in the process (1). Thus, j-j coupling [19] describes the physical situation arising from the inclusion of the SOI in our study of the process (1). Unless mentioned otherwise, the other symbols, used in this appendix, have their meanings the same as specified elsewhere in this paper.

The DO [corresponding to the one in Eq. (A3)] for representing the noninteracting system of a photon and unpolarized atom is now given by

$$\rho_i = \rho_0 \otimes \rho_r = \frac{1}{(2J_0+1)}\sum_{M_0}|0;1m_r\rangle\langle 0;1m_r|. \tag{B2}$$

An element of the required DO (2b) then becomes

$$\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle$$
$$= \mathcal{K}\sum_{M_f}\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|F_aF_p\rho_iF_p^{\dagger}F_a^{\dagger}|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle. \tag{B3}$$

This equation includes sum over all the Zeeman levels [19] of A^{++} which remain unresolved as well as unobserved. Substitution of Eq. (B2) in (B3) yields

$$\begin{aligned}
&\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|\rho_f|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle \\
&\quad=\frac{\mathcal{K}}{(2J_0+1)}\sum_{M_0\ M_f}\langle f;\mu_1\hat{u}_1\vec{k}_1;\mu_2\hat{u}_2\vec{k}_2|F_aF_p|0;1m_r\rangle \\
&\qquad\times\langle 0;1m_r|F_p^{\dagger}F_a^{\dagger}|f;\mu_1'\hat{u}_1\vec{k}_1;\mu_2'\hat{u}_2\vec{k}_2\rangle \qquad \text{(B4)}
\end{aligned}$$

On twice using in (B4) the completeness relation $\Sigma_{M_e}|J_eM_e\rangle\langle J_eM_e|=1\equiv\Sigma_{M_e}|e\rangle\langle e|$ for the magnetic states of the excited photoion A^{+*}, one finds that

$$\begin{aligned}
&\frac{\mathcal{K}}{(2J_0+1)}\sum_{M_0\ M_e\ M_e'\ M_f}\langle f;\mu_2\hat{u}_2\vec{k}_2|F_a|e\rangle \\
&\qquad\times\langle e;\mu_1\hat{u}_1\vec{k}_1|F_p|0;1m_r\rangle\langle 0;1m_r|F_p^{\dagger}|e';\mu_1'\hat{u}_1\vec{k}_1\rangle \\
&\qquad\times\langle e'|F_a^{\dagger}|f;\mu_2'\hat{u}_2\vec{k}_2\rangle. \qquad \text{(B5)}
\end{aligned}$$

In analogy with the DM (A6), suitable for *L*-*S* coupling, the present DM (B5), appropriate for *j*-*j* coupling, is Hermitian as well.

In order to calculate the matrix elements of the photoionization and of Auger emission operators F_p and F_a, respectively, we follow procedures similar to those used in Appendix A with the difference that unlike therein, we now need to work in the *j*-*j* coupling. Consequently, we now have [according to the left-hand side of Eq. (B1a)],

$$\begin{aligned}
|0;1m_r\rangle&=\sum_{JM_J}(-1)^{1-J_0-M_J}\sqrt{2J+1}\begin{pmatrix}J_0 & 1 & J\\ M_{J_0} & m_r & -M_J\end{pmatrix} \\
&\qquad\times|(J_01)JM_J\rangle \qquad \text{(B6a)}
\end{aligned}$$

for the (photon + atom) system before the interaction between the two takes place. Similarly, taking the coupling of the angular momenta on the right-hand side of Eq. (B1b) into account, we express

$$\begin{aligned}
|e;\mu_1\hat{u}_1\vec{k}_1\rangle&=(-1)^{(1/2)-J_e}\sum_{\substack{\ell_1m_1\nu_1\\ j_1m_{j_1}}}\sum_{JM_J}i^{\ell_1}(-1)^{\ell_1+j_1-m_{j_1}-M_J} \\
&\qquad\times e^{-i\sigma_{\ell_1j_1}}\sqrt{(2j_1+1)(2J+1)}\begin{pmatrix}\ell_1 & \frac{1}{2} & j_1\\ m_1 & \nu_1 & -m_{j_1}\end{pmatrix} \\
&\qquad\times\begin{pmatrix}J_e & j_1 & J\\ M_e & m_{j_1} & -M_J\end{pmatrix} \\
&\qquad\times[Y_{\ell_1}^{m_1}(\hat{k}_1)]^*[\mathcal{D}_{\mu_1\nu_1}^{1/2}(\omega_1)]^*|(J_ej_1)JM_J\rangle^- \qquad \text{(B6b)}
\end{aligned}$$

in terms of the *j*-*j* coupled states $|(J_ej_1)JM_J\rangle^-$ of the $(A^{+^*}+e_1)$ system.

In order to obtain an expression for the matrix element of the transition (1a) in *j*-*j* coupling, we use states (B6) and find

$$\begin{aligned}
\mathcal{P}_e&\equiv\langle e;\mu_1\hat{u}_1\vec{k}_1|F_p|0;1m_r\rangle \\
&=(-1)^{-1/2}\sum_{\substack{\ell_1m_1\nu_1\\ j_1m_{j_1}}}(-i)^{\ell_1} \\
&\qquad\times(-1)^{\ell_1+m_{j_1}}e^{i\sigma_{\ell_1j_1}} \\
&\qquad\times\sqrt{2j_1+1}\begin{pmatrix}\ell_1 & \frac{1}{2} & j_1\\ m_1 & \nu_1 & -m_{j_l}\end{pmatrix}Y_{\ell_1}^{m_1}(\hat{k}_1)\mathcal{D}_{\mu_1\nu_1}^{1/2}(\omega_1) \\
&\qquad\times\langle\Psi_{e;j_1m_{j_1}}^-|F_p|0;1m_r\rangle. \qquad \text{(B7a)}
\end{aligned}$$

Here we have defined

$$\begin{aligned}
\langle\Psi_{e;j_1m_{j_1}}^-|F_p|0;1m_r\rangle&\equiv(-1)^{1-J_0-J_e+j_1}\sum_{JM_J}(-1)^{2M_J}(2J+1) \\
&\qquad\times\begin{pmatrix}J_0 & 1 & J\\ M_0 & m_r & -M_J\end{pmatrix} \\
&\qquad\times\begin{pmatrix}J_e & j_1 & J\\ M_e & m_{j_1} & -M_J\end{pmatrix}\langle J_ej_1\|F(J)\|J_01\rangle \\
&\qquad\qquad \text{(B7b)}
\end{aligned}$$

with

$$\langle(J_ej_1)JM_J|F_p|(J_01)J'M_{J'}\rangle=\langle J_ej_1\|F(J)\|J_01\rangle\delta_{JJ'}\delta_{M_JMJ'} \qquad \text{(B7c)}$$

obtained by using the conservation condition (B1a) for the total angular momentum applicable to the step (1a) of the process (1). In order to obtain a simplified form for (B7), and hence for the required DM, let us rewrite the above expression using the angular momentum transferred [69] $j_t=\ell_r-j_1=J_e-J_0$ from absorbed photon to the photoelectron e_1 observed along with its spin polarization. [Here, the excited photoion A^{+^*} remains unobserved [69]. The definition of the angular momentum transfer is readily obtained from the conservation condition (B1a).] This scheme of recoupling of the angular momenta can be shown [69] to reduce $\langle\Psi_{e;j_1m_{j_1}}^-|F_p|0;1m_r\rangle$ to the following form:

$$\begin{aligned}
\langle\Psi_{e;j_1m_{j_1}}^-|F_p|0;1m_r\rangle&=(-1)^{-J_0-J_e-M_0-m_{j_1}}\sum_{j_tm_t}(2j_t+1) \\
&\qquad\times\begin{pmatrix}1 & j_1 & j_t\\ m_r & -m_{j_1} & m_t\end{pmatrix}\begin{pmatrix}J_e & J_0 & j_t\\ M_e & -M_0 & m_t\end{pmatrix} \\
&\qquad\times\langle J_ej_1\|F(j_t)\|J_01\rangle, \qquad \text{(B8a)}
\end{aligned}$$

where

$$\begin{aligned}
\langle J_ej_1\|F(j_t)\|J_01\rangle&\equiv\sum_J(-1)^J(2J+1)\begin{Bmatrix}1 & J_0 & J\\ J_e & j_1 & j_t\end{Bmatrix} \\
&\qquad\times\langle J_ej_1\|F(J)\|J_01\rangle. \qquad \text{(B8b)}
\end{aligned}$$

Substitution of (B8a) in (B7a) and some subsequent simplifications lead to the following:

$$\mathcal{P}_e = (-1)^{-(1/2)-J_0-J_e-M_0} \sum_{\substack{\ell_1 m_1 \nu_1 \\ j_1 m_{j_1}}} \sum_{j_t m_t} (-i)^{\ell_1} (-1)^{\ell_1} e^{i\sigma_{\ell_1 j_1}} (2j_t+1)$$
$$\times \sqrt{2j_1+1} \begin{pmatrix} \ell_1 & \frac{1}{2} & j_1 \\ m_1 & \nu_1 & -m_{j_l} \end{pmatrix} \begin{pmatrix} 1 & j_1 & j_t \\ m_r & -m_{j_1} & m_t \end{pmatrix}$$
$$\times \begin{pmatrix} J_e & J_0 & j_t \\ M_e & -M_0 & m_t \end{pmatrix} Y_{\ell_1}^{m_1}(\hat{k}_1) \mathcal{D}^{1/2}_{\mu_1 \nu_1}(\omega_1)$$
$$\times \langle J_e j_1 | F(j_t) | J_0 1 \rangle, \tag{B9}$$

the final form of the matrix element for the transition (1a) in $E1$ approximation in the j-j coupling.

Let us next evaluate the matrix element for the Auger transition (1b). Taking the angular momentum coupling expressed on the right-hand side of Eq. (B1b) into account, a state of the $(A^{2+}+e_2)$ system is given by

$$|f;\mu_2 \hat{u}_2 \vec{k}_2\rangle = (-1)^{(1/2)-J_f} \sum_{\substack{\ell_2 m_2 \nu_2 \\ j_2 m_{j_2}}} \sum_{J M_J} i^{\ell_2} (-1)^{\ell_2 + j_2 - m_{j_2} - M_J} e^{-i\sigma_{\ell_2 j_2}}$$
$$\times \sqrt{(2j_2+1)(2J+1)} \begin{pmatrix} \ell_2 & \frac{1}{2} & j_2 \\ m_2 & \nu_2 & -m_{j_2} \end{pmatrix}$$
$$\times \begin{pmatrix} J_f & j_2 & J \\ M_f & m_{j_2} & -M_J \end{pmatrix}$$
$$\times [Y_{\ell_2}^{m_2}(\hat{k}_2)]^* [\mathcal{D}^{1/2}_{\mu_2 \nu_2}(\omega_2)]^* |(J_f j_2) J M_J\rangle^-. \tag{B10}$$

Here, $|(J_f j_2) J M_J\rangle^-$ is the j-j coupled state of $(A^{2+}+e_2)$. The above expression can also be written in analogy with the state (B6b) for the $(A^{+^*}+e_1)$ system. Now the Auger decay matrix element is readily calculated to be

$$\mathcal{A}_e \equiv \langle f;\mu_2 \hat{u}_2 \vec{k}_2 | F_a | A^{+^*}\rangle = (-1)^{-(1/2)+J_f+M_e} \sum_{\substack{\ell_2 m_2 \nu_2 \\ j_2 m_{j_2}}} (-i)^{\ell_2}$$
$$\times (-1)^{\ell_2 - j_2 + m_{j_2}} e^{i\sigma_{\ell_2 j_2}} \sqrt{(2j_2+1)(2J_e+1)}$$
$$\times \begin{pmatrix} \ell_2 & \frac{1}{2} & j_2 \\ m_2 & \nu_2 & -m_{j_2} \end{pmatrix} \begin{pmatrix} J_f & j_2 & J_e \\ M_f & m_{j_2} & -M_e \end{pmatrix}$$
$$\times Y_{\ell_2}^{m_2}(\hat{k}_2) \mathcal{D}^{1/2}_{\mu_2 \nu_2}(\omega_2) \langle (J_f j_2) J_e | F_a | J_e \rangle \tag{B11}$$

in the j-j coupling. In arriving at (B11), use has been made of the conservation condition (B1b) in the following form:

$$\langle (J_f j_2) J M_J | F_a | J_e M_e \rangle = \langle (J_f j_2) J_e | F_a | J_e \rangle \delta_{J J_e} \delta_{M_J M_e}.$$

The final step in the calculation of the DM for the process (1) in the j-j coupling requires the substitutions of the matrix elements (B9) and (B11) in (B5) and simplification of the subsequent expression using Racah algebra. However, one of the simpler ways to do it is to first separately evaluate and simplify the following two expressions which are simultaneously present in (B5):

$$\sum_{M_0} \mathcal{P}_e (\mathcal{P}'_e)^\dagger \equiv \sum_{M_0} \langle e;\mu_1 \hat{u}_1 \vec{k}_1 | F_p | 0;1 m_r \rangle \langle 0;1 m_r | F_p^\dagger | e';\mu'_1 \hat{u}_1 \vec{k}_1 \rangle \tag{B12}$$

and

$$\sum_{M_f} \mathcal{A}_e (\mathcal{A}'_e)^\dagger \equiv \sum_{M_f} \langle f;\mu_2 \hat{u}_2 \vec{k}_2 | F_a | e \rangle \langle e' | F_a^\dagger | f;\mu'_2 \hat{u}_2 \vec{k}_2 \rangle. \tag{B13}$$

Simplification procedure for (B12) involves, among other things, (i) substitution of (B9) and of its Hermitian conjugate, (ii) double use of the identity (14.42) given in Ref. [68], and (iii) a single application of Eq. (2.19) from Ref. [70]. In order to simplify (B13), on the other hand, one merely needs, *inter alia*, to substitute (B11) and twice use Eq. (14.42) from de Shalit and Talmi [68].

The simplified forms of each of (B12) and (B13), thus obtained, are then substituted in the DM (B5) and the remaining sums over M_e, M'_e are readily evaluated using the orthonormality of 3-j symbols [see, for example, Eq. (3.7.8) in [49]]. A few other simplifications help us in writing the DM for the process (1) in j-j coupling when SOI is taken into account in the following final form:

$$\langle f;\mu_1 \hat{u}_1 \vec{k}_1;\mu_2 \hat{u}_2 \vec{k}_2 | \rho_f | f;\mu'_1 \hat{u}_1 \vec{k}_1;\mu'_2 \hat{u}_2 \vec{k}_2 \rangle = (-1)^{\mu'_1+\mu'_2} \sum_{\substack{S_1\, M_{S_1}\, N_{S_1} \\ S_2\, M_{S_2}\, N_{S_2}}} \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & S_1 \\ \mu_1 & -\mu'_1 & M_{S_1} \end{pmatrix} \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & S_2 \\ \mu_2 & -\mu'_2 & M_{S_2} \end{pmatrix}$$
$$\times [\mathcal{D}^{S_1}_{M_{S_1} N_{S_1}}(\omega_1)]^* [\mathcal{D}^{S_2}_{M_{S_2} N_{S_2}}(\omega_2)]^* A^{S_1 S_2}_{N_{S_1} N_{S_2}}(m_r;\vec{k}_1 \vec{k}_2), \tag{B14a}$$

where we have defined

$$A^{S_1S_2}_{N_{S_1}N_{S_2}}(m_r;\vec{k}_1,\vec{k}_2) = (-1)^{1+m_r+J_0+2J_e+J_f}\frac{(2J_e+1)\mathcal{K}}{4\pi(2J_0+1)} \sum_{\substack{\ell_1\ell_1'j_1j_1'L_1M_{L_1}J_1J_2j_t \\ \ell_2\ell_2'j_2j_2'L_2M_{L_2}ML_rj_t'}} (-1)^{\ell_1'+\ell_2-j_2'+j_t} \times (2S_1+1)(2S_2+1)(2J_1+1)(2J_2+1)(2L_r$$

$$+1)\sqrt{(2L_1+1)(2L_2+1)}\begin{pmatrix} \ell_1 & \ell_1' & L_1 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} \ell_2 & \ell_2' & L_2 \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L_1 & S_1 & J_1 \\ M_{L_1} & N_{S_1} & -M \end{pmatrix}\begin{pmatrix} L_2 & S_2 & J_2 \\ M_{L_2} & N_{S_2} & M \end{pmatrix}\begin{pmatrix} 1 & 1 & L_r \\ m_r & -m_r & 0 \end{pmatrix}$$

$$\times\begin{pmatrix} J_1 & J_2 & L_r \\ -M & M & 0 \end{pmatrix}\begin{Bmatrix} J_e & J_e & J_2 \\ j_t & j_t' & J_0 \end{Bmatrix}\begin{Bmatrix} J_e & J_e & J_2 \\ j_2 & j_2' & J_f \end{Bmatrix}\begin{Bmatrix} \ell_1 & \ell_1' & L_1 \\ \frac{1}{2} & \frac{1}{2} & S_1 \\ j_1 & j_1' & J_1 \end{Bmatrix}\begin{Bmatrix} \ell_2 & \ell_2' & L_2 \\ \frac{1}{2} & \frac{1}{2} & S_2 \\ j_2 & j_2' & J_2 \end{Bmatrix}\begin{Bmatrix} 1 & 1 & L_r \\ j_1 & j_1' & J_1 \\ j_t & j_t' & J_2 \end{Bmatrix}$$

$$\times[Y^{M_{L_1}}_{L_1}(\hat{k}_1)]^*[Y^{M_{L_2}}_{L_2}(\hat{k}_2)]^* P_e(J_ej_1;J_01;j_t;\ell_1)[P_e(J_ej_1';J_01;j_t';\ell_1')]^* A_e(J_fj_2;J_e;\ell_2)[A_e(J_fj_2';J_e;\ell_2')]^* \qquad \text{(B14b)}$$

with

$$P_e(J_ej_1;J_01;j_t;\ell_1) = (-i)^{\ell_1}e^{i\sigma_{\ell_1j_1}}(-1)^{\ell_1+j_t}(2j_t+1)\sqrt{(2\ell_1+1)(2j_1+1)}\langle J_ej_1|F(j_t)|J_01\rangle \qquad \text{(B15a)}$$

and

$$A_e(J_fj_2;J_e;\ell_2) = (-i)^{\ell_2}e^{i\sigma_{\ell_2j_2}}\sqrt{(2\ell_2+1)(2j_2+1)}\langle (J_fj_2)J_e|F_a|J_e\rangle \qquad \text{(B15b)}$$

containing the photoionization matrix element $\langle J_ej_1|F(j_t)|J_01\rangle$ and the Auger decay element $\langle (J_fj_2)J_e|F_a|J_e\rangle$, respectively, both defined in the *j*-*j* coupling in Eqs. (B8b) and (B11).

The four diagonal elements of the matrix (B14) are

$$(\rho_f)_{\frac{1}{2},\frac{1}{2};\frac{1}{2},\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle = -\tfrac{1}{2}A^{00}_{00} - g(\hat{u}_1;\vec{k}_1,\vec{k}_2) - g(\hat{u}_2;\vec{k}_1,\vec{k}_2) - h(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),(\rho_f)_{\frac{1}{2},-\frac{1}{2};\frac{1}{2},-\frac{1}{2}}$$

$$\equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle = -\tfrac{1}{2}A^{00}_{00} - g(\hat{u}_1;\vec{k}_1,\vec{k}_2) + g(\hat{u}_2;\vec{k}_1,\vec{k}_2) + h(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),(\rho_f)_{-\frac{1}{2},\frac{1}{2};-\frac{1}{2},\frac{1}{2}}$$

$$\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle = -\tfrac{1}{2}A^{00}_{00} + g(\hat{u}_1;\vec{k}_1,\vec{k}_2) - g(\hat{u}_2;\vec{k}_1,\vec{k}_2) + h(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),(\rho_f)_{-\frac{1}{2},-\frac{1}{2};-\frac{1}{2},-\frac{1}{2}}$$

$$\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle = -\tfrac{1}{2}A^{00}_{00} + g(\hat{u}_1;\vec{k}_1,\vec{k}_2) + g(\hat{u}_2;\vec{k}_1,\vec{k}_2) - h(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2). \qquad \text{(B16a)}$$

Here, we have defined

$$g(\hat{u}_1;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{2\sqrt{3}}\left(-\tfrac{1}{\sqrt{2}}s_1e^{-i\varphi_1}A^{10}_{10} + c_1A^{10}_{00} + \tfrac{1}{\sqrt{2}}s_1e^{i\varphi_1}A^{10}_{-10}\right), g(\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{3}}\left(-\frac{1}{\sqrt{2}}s_2e^{-i\varphi_2}A^{01}_{01} + c_2A^{01}_{00}\right.$$

$$\left. + \frac{1}{\sqrt{2}}s_2e^{i\varphi_2}A^{01}_{0-1}\right), h(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{6}\left(-\frac{1}{\sqrt{2}}c_1s_2e^{-i\varphi_2}A^{11}_{01} + c_1c_2A^{11}_{00} + \frac{1}{\sqrt{2}}c_1s_2e^{i\varphi_2}A^{11}_{0-1} + \frac{1}{2}s_1s_2e^{-i(\varphi_1+\varphi_2)}A^{11}_{11}\right.$$

$$\left. - \frac{1}{\sqrt{2}}s_1c_2e^{-i\varphi_1}A^{11}_{10} - \frac{1}{2}s_1s_2e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1} - \frac{1}{2}s_1s_2e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11} + \frac{1}{\sqrt{2}}s_1c_2e^{i\varphi_1}A^{11}_{-10} + \frac{1}{2}s_1s_2e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1}\right). \qquad \text{(B16b)}$$

Each of the 12 nondiagonal elements of the DM (B14) is, on the other hand, given by

$$(\rho_f)_{\frac{1}{2},\frac{1}{2};\frac{1}{2},-\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle$$
$$= -G_-(\hat{u}_2;\vec{k}_1,\vec{k}_2) - H^{(1)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$(\rho_f)_{\frac{1}{2},\frac{1}{2};-\frac{1}{2},\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle$$
$$= -G_-(\hat{u}_1;\vec{k}_1,\vec{k}_2) - H^{(2)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$(\rho_f)_{\frac{1}{2},\frac{1}{2};-\frac{1}{2},-\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle$$
$$= -H^{(3)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2);$$

$$(\rho_f)_{\frac{1}{2},-\frac{1}{2};\frac{1}{2},\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle$$
$$= G_+(\hat{u}_2;\vec{k}_1,\vec{k}_2) + H^{(4)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$(\rho_f)_{\frac{1}{2},-\frac{1}{2};-\frac{1}{2},\frac{1}{2}} \equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle$$
$$= H^{(5)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$
\begin{aligned}
(\rho_f)_{\frac{1}{2},-\frac{1}{2};-\frac{1}{2},-\frac{1}{2}} &\equiv \left\langle f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= -G_-(\hat{u}_1;\vec{k}_1,\vec{k}_2)+H^{(2)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2);
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},\frac{1}{2};\frac{1}{2},\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= G_+(\hat{u}_1;\vec{k}_1,\vec{k}_2)+H^{(6)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},\frac{1}{2};\frac{1}{2},-\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= H^{(7)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},\frac{1}{2};-\frac{1}{2},-\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= -G_-(\hat{u}_2;\vec{k}_1,\vec{k}_2)+H^{(1)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2);
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},-\frac{1}{2};\frac{1}{2},\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= -H^{(8)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},-\frac{1}{2};\frac{1}{2},-\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= G_+(\hat{u}_1;\vec{k}_1,\vec{k}_2)-H^{(6)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),
\end{aligned}
$$

$$
\begin{aligned}
(\rho_f)_{-\frac{1}{2},-\frac{1}{2};-\frac{1}{2},\frac{1}{2}} &\equiv \left\langle f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;-\tfrac{1}{2}\hat{u}_2\vec{k}_2\middle|\rho_f\middle|f;-\tfrac{1}{2}\hat{u}_1\vec{k}_1;\tfrac{1}{2}\hat{u}_2\vec{k}_2\right\rangle \\
&= G_+(\hat{u}_2;\vec{k}_1,\vec{k}_2)-H^{(4)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2), \qquad \text{(B16c)}
\end{aligned}
$$

where

$$
\begin{aligned}
G_-(\hat{u}_1;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{\sqrt{6}}\Bigg(&\frac{1}{2}(1-c_1)e^{-i\varphi_1}A^{10}_{10}-\frac{1}{\sqrt{2}}s_1A^{10}_{00} \\
&+\frac{1}{2}(1+c_1)e^{i\varphi_1}A^{10}_{-10}\Bigg),
\end{aligned}
$$

$$
\begin{aligned}
G_-(\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{\sqrt{6}}\Bigg(&\frac{1}{2}(1-c_2)e^{-i\varphi_2}A^{01}_{01}-\frac{1}{\sqrt{2}}s_2A^{01}_{00} \\
&+\frac{1}{2}(1+c_2)e^{i\varphi_2}A^{01}_{0-1}\Bigg),
\end{aligned}
$$

$$
\begin{aligned}
G_+(\hat{u}_1;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{\sqrt{6}}\Bigg(&\frac{1}{2}(1+c_1)e^{-i\varphi_1}A^{10}_{10}+\frac{1}{\sqrt{2}}s_1A^{10}_{00} \\
&+\frac{1}{2}(1-c_1)e^{i\varphi_1}A^{10}_{-10}\Bigg),
\end{aligned}
$$

$$
\begin{aligned}
G_+(\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{\sqrt{6}}\Bigg(&\frac{1}{2}(1+c_2)e^{-i\varphi_2}A^{01}_{01}+\frac{1}{\sqrt{2}}s_2A^{01}_{00} \\
&+\frac{1}{2}(1-c_2)e^{i\varphi_2}A^{01}_{0-1}\Bigg),
\end{aligned}
$$

$$
\begin{aligned}
H^{(1)}(\hat{u}_2,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{3\sqrt{2}}[&\mathcal{H}^{(1)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(1)}_2 \\
&\times(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(1)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \\
&+\mathcal{H}^{(1)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)],
\end{aligned}
$$

$$
\begin{aligned}
H^{(2)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{3\sqrt{2}}[&\mathcal{H}^{(2)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(2)}_2 \\
&\times(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(2)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \\
&+\mathcal{H}^{(2)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)],
\end{aligned}
$$

$$
\begin{aligned}
H^{(3)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{3}\Bigg\{&\mathcal{H}^{(3)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)-\frac{1}{2\sqrt{2}}s_1[(1+c_2) \\
&\times e^{i\varphi_2}A^{11}_{0-1}+(1-c_2)e^{-i\varphi_2}A^{11}_{01}]+\tfrac{1}{4}(1+c_1) \\
&\times(1+c_2)e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1}+\tfrac{1}{4}(1-c_1)(1 \\
&-c_2)e^{-i(\varphi_1+\varphi_2)}A^{11}_{11}+\tfrac{1}{4}(1-c_1)(1 \\
&+c_2)e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1}+\tfrac{1}{4}(1+c_1)(1 \\
&-c_2)e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}\Bigg\},
\end{aligned}
$$

$$
\begin{aligned}
H^{(4)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{3\sqrt{2}}[&\mathcal{H}^{(4)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(4)}_2 \\
&\times(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(4)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \\
&+\mathcal{H}^{(4)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)],
\end{aligned}
$$

$$
\begin{aligned}
H^{(5)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{3}\Bigg\{&\mathcal{H}^{(5)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)-\frac{1}{2\sqrt{2}}s_1[(1-c_2) \\
&\times e^{i\varphi_2}A^{11}_{0-1}+(1+c_2)e^{-i\varphi_2}A^{11}_{01}]+\tfrac{1}{4}(1+c_1) \\
&\times(1-c_2)e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1}+\tfrac{1}{4}(1-c_1)(1 \\
&+c_2)e^{-i(\varphi_1+\varphi_2)}A^{11}_{11}+\tfrac{1}{4}(1-c_1)(1 \\
&-c_2)e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1}+\tfrac{1}{4}(1+c_1)(1 \\
&+c_2)e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}\Bigg\},
\end{aligned}
$$

$$
\begin{aligned}
H^{(6)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{3\sqrt{2}}[&\mathcal{H}^{(6)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(6)}_2 \\
&\times(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\mathcal{H}^{(6)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \\
&+\mathcal{H}^{(6)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)],
\end{aligned}
$$

$$
\begin{aligned}
H^{(7)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{3}\Bigg(&\mathcal{H}^{(7)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2)+\frac{1}{2\sqrt{2}}s_1[(1+c_2) \\
&\times e^{i\varphi_2}A^{11}_{0-1}+(1-c_2)e^{-i\varphi_2}A^{11}_{01}]+\tfrac{1}{4}(1-c_1) \\
&\times(1+c_2)e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1}+\tfrac{1}{4}(1+c_1)(1
\end{aligned}
$$

$$- c_2)e^{-i(\varphi_1+\varphi_2)}A^{11}_{11} + \tfrac{1}{4}(1+c_1)(1 + c_2)e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1} + \tfrac{1}{4}(1-c_1)(1 - c_2)e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}\Big),$$

$$H^{(8)}(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{3}\Big\{\mathcal{H}^{(8)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) + \frac{1}{2\sqrt{2}}s_1[(1-c_2) \times e^{i\varphi_2}A^{11}_{0-1} + (1+c_2)e^{-i\varphi_2}A^{11}_{01}] + \tfrac{1}{4}(1-c_1) \times (1-c_2)e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1} + \tfrac{1}{4}(1+c_1)(1 + c_2)e^{-i(\varphi_1+\varphi_2)}A^{11}_{11} + \tfrac{1}{4}(1+c_1)(1 - c_2)e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1} + \tfrac{1}{4}(1-c_1)(1 + c_2)e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}\Big\}, \tag{B16d}$$

with

$$\mathcal{H}^{(1)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\frac{1}{\sqrt{2}}c_1s_2A^{11}_{00} + \frac{1}{2}s_1s_2(e^{-i\varphi_1}A^{11}_{10} - e^{i\varphi_1}A^{11}_{-10}),$$

$$\mathcal{H}^{(1)}_2(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{2}c_1[(1+c_2)e^{i\varphi_2}A^{11}_{0-1} + (1-c_2)e^{-i\varphi_2}A^{11}_{01}],$$

$$\mathcal{H}^{(1)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_1(1+c_2)(e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1} - e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1}),$$

$$\mathcal{H}^{(1)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_1(1-c_2)(-e^{-i(\varphi_1+\varphi_2)}A^{11}_{11} + e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11});$$

$$\mathcal{H}^{(2)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\frac{1}{\sqrt{2}}s_1c_2A^{11}_{00} + \frac{1}{2}s_1s_2(e^{-i\varphi_2}A^{11}_{01} - e^{i\varphi_2}A^{11}_{0-1}),$$

$$\mathcal{H}^{(2)}_2(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{2}c_2[(1+c_1)e^{i\varphi_1}A^{11}_{-10} + (1-c_1)e^{-i\varphi_1}A^{11}_{10}],$$

$$\mathcal{H}^{(2)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_2(1+c_1)(e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1} - e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}),$$

$$\mathcal{H}^{(2)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_2(1-c_1)(-e^{i(\varphi_1+\varphi_2)}A^{11}_{11} + e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1});$$

$$\mathcal{H}^{(3)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2}s_1s_2A^{11}_{00} - \frac{1}{2\sqrt{2}}s_2[(1+c_1)e^{i\varphi_1}A^{11}_{-10} + (1-c_1)e^{-i\varphi_1}A^{11}_{10}];$$

$$\mathcal{H}^{(4)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\mathcal{H}^{(1)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$\mathcal{H}^{(4)}_2(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{2}c_1[(1+c_2)e^{-i\varphi_2}A^{11}_{01} + (1-c_2)e^{i\varphi_2}A^{11}_{0-1}],$$

$$\mathcal{H}^{(4)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\frac{1}{2\sqrt{2}}s_1(1+c_2)(e^{-i(\varphi_1+\varphi_2)}A^{11}_{11} - e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}),$$

$$\mathcal{H}^{(4)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_1(1-c_2)(e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1} - e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1});$$

$$\mathcal{H}^{(5)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\mathcal{H}^{(3)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2);$$

$$\mathcal{H}^{(6)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\mathcal{H}^{(2)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2),$$

$$\mathcal{H}^{(6)}_2(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \tfrac{1}{2}c_2[(1-c_1)e^{i\varphi_1}A^{11}_{-10} + (1+c_1)e^{-i\varphi_1}A^{11}_{10}],$$

$$\mathcal{H}^{(6)}_3(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_2(1-c_1)(e^{i(\varphi_1+\varphi_2)}A^{11}_{-1-1} - e^{-i(\varphi_2-\varphi_1)}A^{11}_{-11}),$$

$$\mathcal{H}^{(6)}_4(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv \frac{1}{2\sqrt{2}}s_2(1+c_1)(-e^{-i(\varphi_1+\varphi_2)}A^{11}_{11} + e^{i(\varphi_2-\varphi_1)}A^{11}_{1-1});$$

$$\mathcal{H}^{(7)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\frac{1}{2}s_1s_2A^{11}_{00} - \frac{1}{2\sqrt{2}}s_2[(1-c_1)e^{i\varphi_1}A^{11}_{-10} + (1+c_1)e^{-i\varphi_1}A^{11}_{10}];$$

$$\mathcal{H}^{(8)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2) \equiv -\mathcal{H}^{(7)}_1(\hat{u}_1,\hat{u}_2;\vec{k}_1,\vec{k}_2). \tag{B16e}$$

The Hermiticity of (B14) requires that

$$A^{00}_{00} = (A^{00}_{00})^*, \quad A^{10}_{00} = (A^{10}_{00})^*,$$

$$A^{01}_{00} = (A^{01}_{00})^*, \quad A^{11}_{00} = (A^{11}_{00})^*;$$

$$A^{11}_{1-1} = (A^{11}_{-11})^*, \quad A^{11}_{11} = (A^{11}_{-1-1})^*;$$

$$A^{11}_{10} = -(A^{11}_{-10})^*, \quad A^{11}_{01} = -(A^{11}_{0-1})^*;$$

$$A^{10}_{10} = -(A^{10}_{-10})^*, \quad A^{01}_{01} = -(A^{01}_{0-1})^*. \tag{B17}$$

We further have from (B16a),

$$\mathrm{Tr}(\rho_f) = -2A^{00}_{00}. \tag{B18}$$

Here, Tr stands for the trace of a matrix.

The 16 coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}(m_r;\vec{k}_1,\vec{k}_2)$, calculated from (B14b), by substituting Eqs. (18) and (19), in addition to m_r=0, are

$$A^{10}_{00}(\theta,\phi)=0, \quad A^{01}_{00}(\theta,\phi)=0,$$

$$A^{11}_{1-1}(\theta,\phi)=-0.005\,860\cos^2\theta,$$

$$A^{11}_{-11}(\theta,\phi)=-0.005\,860\cos^2\theta,$$

$$A^{10}_{10}(\theta,\phi)=i0.000\,837\sin 2\theta e^{i\phi},$$

$$A^{10}_{-10}(\theta,\phi)=i0.000\,837\sin 2\theta e^{-i\phi},$$

$$A^{01}_{01}(\theta,\phi)=-i0.000\,837\sin 2\theta e^{i\phi},$$

$$A^{01}_{0-1}(\theta,\phi)=-i0.000\,837\sin 2\theta e^{-i\phi},$$

$$A^{11}_{10}(\theta,\phi)=0.001\,211\sin 2\theta e^{i\phi},$$

$$A^{11}_{-10}(\theta,\phi)=-0.001\,211\sin 2\theta e^{-i\phi},$$

$$A^{11}_{01}(\theta,\phi)=-0.001\,211\sin 2\theta e^{i\phi},$$

$$A^{11}_{0-1}(\theta,\phi)=0.001\,211\sin 2\theta e^{-i\phi},$$

$$A^{11}_{11}(\theta,\phi)=-0.001\,218\sin^2\theta e^{2i\phi},$$

$$A^{11}_{-1-1}(\theta,\phi)=-0.001\,218\sin^2\theta e^{-2i\phi},$$

$$A^{00}_{00}(\theta,\phi)=-0.001180-0.000774\cos 2\theta,$$

$$A^{11}_{00}(\theta,\phi)=0.002321+0.003539\cos 2\theta. \tag{B19}$$

Each of the A's in (B19) [as well as in (B20) and (B21)] is to be multiplied by $(|A_e(J_f=0,j_2=5/2;J_e=5/2,\ell_2=2)|^2\mathcal{K})/(4\pi)$. Here, the Auger decay amplitude $A_e(J_f,j_2;J_e,\ell_2)$ are defined in Eq. (B15b); whereas, constant $\mathcal{K}$, present in (B14b) has already been explained elsewhere in the present communication. This multiplication, however, has not explicitly been shown in Eqs. (B19) [including (B20) and (B21)] for brevity.

The following coefficients

$$A^{00}_{00}(\theta,\phi)=-0.000\,792\,817+0.000\,386\,796\cos 2\theta,$$

$$A^{01}_{0-1}(\theta,\phi)=(0.000\,598\,331+i0.000\,418\,455)\sin 2\theta e^{-i\phi},$$

$$A^{01}_{00}(\theta,\phi)=-0.000\,142\,918+0.000\,846\,167\cos 2\theta,$$

$$A^{01}_{01}(\theta,\phi)=(-0.000\,598\,331+i0.000\,418\,455)\sin 2\theta e^{i\phi},$$

$$A^{10}_{-10}(\theta,\phi)=(-0.000\,101\,058-i0.000\,418\,455)\sin 2\theta e^{-i\phi},$$

$$A^{10}_{00}(\theta,\phi)=-0.000\,142\,918\cos 2\theta+0.000\,846\,167,$$

$$A^{10}_{10}(\theta,\phi)=(0.000\,101\,058-i0.000\,418\,455)\sin 2\theta e^{i\phi},$$

$$A^{11}_{0-1}(\theta,\phi)=(-0.001\,036\,34-i0.000\,724\,786)\sin 2\theta e^{-i\phi},$$

$$A^{11}_{00}(\theta,\phi)=0.001\,160\,39-0.002\,378\,45\cos 2\theta,$$

$$A^{11}_{01}(\theta,\phi)=(0.001\,036\,34-i0.000\,724\,786)\sin 2\theta e^{i\phi},$$

$$A^{11}_{-10}(\theta,\phi)=(0.000\,175\,038+i0.000\,724\,786)\sin 2\theta e^{-i\phi},$$

$$A^{11}_{10}(\theta,\phi)=(-0.000\,175\,038+i0.000\,724\,786)\sin 2\theta e^{i\phi},$$

$$A^{11}_{-1-1}(\theta,\phi)=0,$$

$$A^{11}_{-11}(\theta,\phi)=-2*(0.001\,160\,39+i0.001\,025)\sin^2\theta,$$

$$A^{11}_{1-1}(\theta,\phi)=-2*(0.001\,160\,39-i0.001\,025)\sin^2\theta,$$

$$A^{11}_{11}(\theta,\phi)=0, \tag{B20}$$

are obtained from Eq. (B14b) again. These are for CP electromagnetic radiation with NH (i.e., $m_r=-1$). In order to obtain expressions (B20) for the 2-DPI process (17) in Xe, we have made use of Eqs. (18) and (19).

Last, we need the coefficients $A^{S_1S_2}_{N_{S_1}N_{S_2}}(+1;\vec{k}_1,\vec{k}_2)$ also for the ionizing radiation in (17) to be RCP (i.e., CP with PH) for the diametric emissions of photoelectron and Auger electron. For this, we again repeat the methodologies used in calculating (B19) and (B20), but this time with $m_r=+1$. The resulting 16 coefficients now come out to be

$$A^{00}_{00}(\theta,\phi)=-0.000\,792\,817+0.000\,386\,796\cos 2\theta,$$

$$A^{01}_{0-1}(\theta,\phi)=(-0.000\,598\,331+i0.000\,418\,455)\sin 2\theta e^{-i\phi},$$

$$A^{01}_{00}(\theta,\phi)=0.000\,142\,918-0.000\,846\,167\cos 2\theta,$$

$$A^{01}_{01}(\theta,\phi)=(0.000\,598\,331+i0.000\,418\,455)\sin 2\theta e^{i\phi},$$

$$A^{10}_{-10}(\theta,\phi)=(0.000\,101\,058-i0.000\,418\,455)\sin 2\theta e^{-i\phi},$$

$$A^{10}_{00}(\theta,\phi)=0.000\,142\,918\cos 2\theta-0.000\,846\,167,$$

$$A^{10}_{10}(\theta,\phi)=(-0.000\,101\,058-i0.000\,418\,455)\sin 2\theta e^{i\phi},$$

$$A^{11}_{0-1}(\theta,\phi)=(-0.001\,036\,34+i0.000\,724\,786)\sin 2\theta e^{-i\phi},$$

$$A^{11}_{00}(\theta,\phi)=0.001\,160\,39-0.002\,378\,45\cos 2\theta,$$

$$A^{11}_{01}(\theta,\phi)=(0.001\,036\,34+i0.000\,724\,786)\sin 2\theta e^{i\phi},$$

$$A^{11}_{-10}(\theta,\phi)=(0.000\,175\,038-i0.000\,724\,786)\sin 2\theta e^{-i\phi},$$

$$A^{11}_{10}(\theta,\phi)=(-0.000\,175\,038-i0.000\,724\,786)\sin 2\theta e^{i\phi},$$

$$A^{11}_{-1-1}(\theta,\phi)=0,$$

$$A^{11}_{-11}(\theta,\phi)=-2(0.001\,160\,39-i0.001\,025)\sin^2\theta,$$

$$A^{11}_{1-1}(\theta,\phi)=-2(0.001\,160\,39+i0.001\,025)\sin^2\theta,$$

$$A^{11}_{11}(\theta,\phi)=0. \tag{B21}$$

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[50] (a) The DMs and their PTs, formed in this communication, were diagonalized using commercially available softwares MATHEMATICA [51] and MATLAB [52]. These two softwares gave results which were in complete agreement with each other. Moreover, in order to further check the correctness of diagonalizations and of other computations done herein using Refs. [51,52], the calculations were performed—wherever feasible—by hand as well. The manual results too agreed with those obtained using MATHEMATICA [51] and MATLAB [52]. (b) The negativity $\mathcal{N}(\rho_f)$ and concurrence $\mathcal{C}(\rho_f)$ are defined by two completely different as well as independent expressions (3) and (4), respectively. It has theoretically been shown [32] that for a pure bipartite state ρ_f, these two measures of entanglement have exactly equal values. Our calculations of $\mathcal{N}(\rho_f)$ and $\mathcal{C}(\rho_f)$, discussed in the Secs. IV B 1, IV B 2, IV B 3, and IV B 4, are in complete conformity with this [32] theoretical prediction. This provides an additional check, both on the correctness as well as on the internal consistency, of the calculations (including diagonalization of various matrices) performed using [51,52] and/or manually in this communication.

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