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Generation of tripartite states of flying electronic qubits and their characterization by energy measurements

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A scheme for generating states of three free electrons entangled with respect to their spins is suggested. It consists of sequential ejection of two Auger electrons (e_2 and e_3) in the nonradiative decay of an inner-shell vacancy created due to the emission of photoelectron e_1 from an atom, say A. In the absence of spin-orbit interaction, the entanglement among the spin angular momenta of the flying (e_1, e_2, e_3) is generated simply by the Coulomb interaction experienced by them inside A. Their states are classified according to the hierarchic structure suggested by Dür, Cirac, and Tarrach [Phys. Rev. Lett. **83**, 3562 (1999); Dür and Cirac, Phys. Rev. A **61**, 042314 (2000)]. The generation of fully separable, 1-electron biseparable, fully inseparable, or "$1 \rightarrow 2$ entangled" tripartite (in addition to various kinds of bipartite) states is shown to be completely determined only by the spin multiplicities of the electronic states of A and of its ionic species $(A^{+^*}, A^{2+^*}, A^{3+})$ participating in the suggested scheme. The entanglement of three electrons is Greenberger-Horne-Zeilinger type. The experimental characterization of these states is fully achieved merely by the measurements of the energies of (e_1, e_2, e_3), without requiring any entanglement witness or other similar protocols hitherto developed in quantum information.

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I. INTRODUCTION

Some of the protocols, hitherto developed in quantum information (QI), require for their successful implementations entangled states of two or more particles shared among different parties located at distant places (see, for example, [1–3]). In the case of discrete variable entanglement, each of these particles must at least be a qubit, i.e., it must have—at least—two independent states simultaneously accessible to it. Due to the intense efforts made during the last two decades, the generation, detection, and quantification of bipartite entanglement of two qubits are now well understood—both experimentally as well as theoretically.

However, our present knowledge of these properties for the entangled states of three or more particles is yet far from satisfactory and/or complete. The investigations carried out hitherto have already shown that the entanglement of more than two particles is not merely an extension of its bipartite counterpart but has instead several new as well as different properties which are advantageous, for example, in superdense coding [4], quantum cloning [5], teleportation [6], "$1 \rightarrow N$ quantum telecloning" (i.e., quantum teleportation with one sender and N receivers) [7], etc. Indeed, Koike *et al.* [8] already experimentally demonstrated "$1 \rightarrow 2$ quantum telecloning" of optical coherent states. The multipartite entangled states, in addition, are needed also to study QI at more fundamental levels. Among the experimentally studied entanglement of three or more qubits in the form of photons [9], trapped ions [10], or cold atoms on an optical lattice [11], the parametric down conversion (PDC) [9] is probably—hitherto—the most successful and widely used method for producing entangled states of two or more photonic qubits.

Although, photons are known to be excellent carriers of information—nothing can travel faster than a photon—they are, however, not suitable for storing information for a long time and detection of a photon, in addition, always leads to its destruction unless one is prepared to perform quantum nondemolition-type [12] extremely difficult experiments. On the other hand, particles with nonzero rest mass can interact and be detected without being destroyed, in addition to being capable of storing information for long durations. Even, otherwise, it is always desirable to perform the above-mentioned [4–8] and other similar beautiful experiments with particles possessing rest mass different from zero.

At the minimum unit of multipartite entanglement, it requires entangled states of three qubits of rest mass different from zero shared among parties at separately addressable distant locations. Two of the possible ways for achieving this are: first, one tries to entangle qubits which are already at distant places. Son *et al.* [13] entangled two remote qubits with the help of a bipartite continuous variable state. Dos Reis and Sharma [14] proposed this [13] field-mediated approach to generate shared tripartite entanglement between two-trapped atoms present together in the same cavity with one party and a single-trapped atom in a remote laboratory owned by another party. However, it remains yet to be seen whether this field-mediated approach can entangle qubits kept at more than two different locations. The other method of producing minimum unit of shared multipartite entanglement consists of sending one qubit each to all the three parties from a single source of tripartite entanglement. This, in fact, is the approach used by Koike *et al.* [8] for generating tripartite photonic entanglement of continuous variables shared by three different remote parties participating in their $1 \rightarrow 2$ quantum telecloning experiment. The success of this second approach depends, obviously, on the weakness of the coupling of the shared tripartite entanglement with the environment so that the dissipation and decoherence have minimal effects, while the three qubits are flying away to their respective destinations from their common source of generation.

It has already been proposed [15] that electron spin as a qubit can be used in quantum computation [1–3]. Consequently, the creation of a solid-state quantum computer based on spin qubits has already been suggested [1–3,16] in spintronics (i.e., active manipulation of the spin degree of freedom of electrons in solid-state environment) [17]. For use of electrons in quantum communication [1–3,18], one requires, on the other hand, two or more 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 [18–21] for electron entanglers have hitherto been made in spintronics. For example, extractions of a Cooper pair from a superconductor [20] or of the singlet ground state of a quantum dot with an even number of electrons [21] are some of the methods which have already been investigated in spintronics.

There are several 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, the presently available synchrotron radiation is very widely and effectively being used for ejecting an inner-shell electron from matter into the continuum. This departed electron is called photoelectron. The resulting positively charged ion (known as photoion) with a vacancy in one of its inner shells is in a highly unstable state. Auger [22] discovered that this excited ion relaxes either radiatively by emitting a photon or nonradiatively in the emission of a secondary (called Auger) electron. For light elements (atomic number less than 26), nonradiative decay is the dominant process for photoions with a vacancy in their *K* shell [23]. The validity of this two-step model [called two-step double photoionization (2-DPI)] for the sequential emission of a photoelectron and an Auger electron is well established if the lifetime of the excited photoion is longer than the time taken for photoionization or if the kinetic energy of the photoelectron is more than that of the Auger electron. Auger spectroscopy has since long been in use both in basic research and material science [24]. The coincidence detection of both electrons emitted in 2-DPI has proved to be useful in studying solid as well as diluted species and go beyond the two-step description [25–28]. Yet, the entanglement properties of the spin state of a photo-Auger-electron pair emitted from an atom [29] or a rotating linear molecule [30] in 2-DPI were investigated only very recently.

A vacancy in the first accessible inner shell of matter usually decays by the emission of a single Auger electron [31]; but the decay of a vacancy in the deep inner shell may lead to the emission of two or more Auger electrons. These more than one secondary electrons are emitted sequentially in a cascade Auger (CA) decay giving rise to a structured energy spectrum. Here, each Auger electron has a discrete kinetic energy—independent of the energy of the ionizing photon—which is determined from the energy difference between the initial, intermediate, and final states. The CA emission has hitherto been observed [32–37] in various kinds of gases (e.g., in the decay of a vacancy in the 3*d* [32,33] or 4*d* [33–35] shell of Xe, 3*d* shell of Kr [33], 2*p* shell of Ar [36], 4*d* shell of Cs [37], etc.). The CA decay is usually described by an extension of the two-step model used for 2-DPI by introducing consecutive decay steps, each emitting one more electron until the internal energy of the residual ion is no longer sufficient to emit further secondary electrons. The validity of this multistep model for describing CA is, however, applicable if intermediate levels with sufficiently long lifetime exist. If these intermediate levels do not exist, two or more secondary electrons have to be simultaneously emitted. This process was called [38] "double Auger" decay for the simultaneous emission of two Auger electrons.

In the present paper, we analyze the entanglement properties of the spin state of three particles consisting of a photoelectron (hereafter called e_1) plus two secondary electrons (e_2 and e_3) emitted in the CA decay of an atom, say *A*. [Unless stated otherwise, hereafter, we call this process as three-step triple photoionization (3-TPI).] Thus, this paper presents a realistic study which can readily be performed in a laboratory of the spin state of three flying qubits, possessing rest mass different from zero, generated from a single source. In addition, we show that there is no coupling at all between the entanglement of (e_1,e_2,e_3) and the environment external to the target atom *A*. The three electrons (e_1,e_2,e_3) can, therefore, be made to fly (with a suitable combination of electric and magnetic fields) to any locations in any desired directions (for performing experiments such as 1→2 quantum telecloning [7,8], etc.) without affecting there entanglement properties due to the harmful effects of dissipation and decoherence.

The entanglement of (e_1,e_2,e_3) is analyzed according to the hierarchic classification suggested in Refs. [39,40] by calculating the partial transpose [41,42] of their tripartite state with respect to each of the three electrons and concurrences [43,44] of the three density matrices (DMs) obtained by tracing the tripartite DM over the photoelectron e_1 or either of the two Auger electrons (e_2,e_3). Simple conditions based on the spins of the atomic species participating in a 3-TPI process of any generic atom *A* are developed for identifying analytically tripartite states of (e_1,e_2,e_3) which [39,40] are fully separable, 1-qubit biseparable, fully inseparable, or "1→2 entangled." Pairs of (e_1,e_2), (e_2,e_3), and (e_3,e_1) in Bell (i.e., pure, maximally entangled), maximally chaotic, Werner (i.e., mixed entangled or separable), and non-Werner states are also shown to exist. Expressions involving spins of the atomic species are obtained to characterize these states and to calculate their concurrences [43,44] for quantifying entanglement of a bipartite state formed of electrons (e_1,e_2), (e_2,e_3), or (e_1,e_3). A brief account of some of these results was presented in Ref. [45].

Section II contains preliminaries related to various definitions used in the rest of this paper and introduces the density operator (DO) for a spin state of (e_1,e_2,e_3) formed in 3-TPI of an atom *A*. A detailed derivation of this tripartite DO is given in the Appendix of this paper. The Appendix also gives a simple method for calculating from this DO the (8×8) DM for a state of the three electronic qubits (e_1,e_2,e_3) formed in the 3-TPI of any generic atom *A*. In Sec. III, we analyze the properties of this DM and show as to why the entanglement properties of a tripartite state of (e_1,e_2,e_3) generated using the scheme suggested in this paper are not coupled to the environment external to the atom *A* and, hence, immune to dissipation and decoherence effects. The classification of these states according to the hierarchic scheme suggested in [39,40] and calculation of their properties is presented in Sec. IV. In the last subsection of the Sec. IV, we suggest experiments on 3-TPI which can realistically be performed, for example, on Xe [33–35] and Cs [37] atoms in a laboratory for generating tripartite states of (e_1,e_2,e_3) possessing desired properties. Finally, Sec. V contains conclusions of the present study.

II. PRELIMINARIES

Let us assume that $|0\rangle$ represents the antisymmetrized initial electronic state of our atomic target A. If $|1\rangle$, $|2\rangle$, and $|f\rangle$ are taken to be the antisymmetrized electronic states of the respective excited photoion A^{+^*}, excited dication A^{2+^*}, and of the triply charged residual positive ion A^{3+} of this target then the three well-known consecutive steps in a 3-TPI process are [32–37]

$$\gamma_r||\vec{\ell}_r| = 1, m_r\rangle + A|0\rangle \longrightarrow A^{+^*}|1\rangle + e_1(\mu_1,\hat{u}_1,\vec{k}_1), \tag{1a}$$

$$A^{+^*}|1\rangle \longrightarrow A^{2+^*}|2\rangle + e_2(\mu_2,\hat{u}_2,\vec{k}_2), \tag{1b}$$

and

$$A^{2+^*}|2\rangle \longrightarrow A^{3+}|f\rangle + e_3(\mu_3,\hat{u}_3,\vec{k}_3). \tag{1c}$$

Here, in the first step (1a), γ_r (with the subscript "r" standing for radiation) represents the incident photon of frequency ν_r and, hence, energy $E_r = h\nu_r$. This photon is absorbed by one of the inner-shell electrons of the target atom A leading to the latter's ionization. For photons possessing such required energies, it is usually sufficient [46] to treat the ionization step (1a) in the electric-dipole ($E1$) approximation. Then, $|\vec{\ell}_r| = 1$ represents the angular momentum of γ_r in the $E1$ approximation. The parameter m_r in (1a) specifies the polarization of the incident radiation: $m_r = 0$ for linear polarization, $m_r = +1$ or -1 for a photon possessing right or left circular polarization, respectively; unpolarized electromagnetic radiation, on the other hand, is taken to be an even mixture of the waves with $m_r = \pm 1$. Further, e_1 in Eq. (1a) is the photoelectron and (e_2,e_3) in Eqs. (1b) and (1c) are the two Auger electrons.

The propagation vector of the emitted i (=1 to 3) 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 $\epsilon_i = \hbar^2 k_i^2/2m$. Also in Eq. (1), $\mu_i (= \pm\frac{1}{2})$ is the projection of the spin angular momentum of the ejected electron e_i along its own direction of quantization $\hat{u}_i = (\vartheta_i,\varphi_i)$. Unless stated otherwise, each of the vectors $\vec{k}_i$ and $\hat{u}_i$ and others used in this paper are defined with respect to the photon-fixed right-handed coordinate system $OXYZ$ (see Fig. 1) centered at the atom A. The polar axis of this system is along the direction of the electric field vector if the photon absorbed in Eq. (1a) is linearly polarized; however, for circularly polarized or unpolarized ionizing radiation, its direction of incidence defines the OZ axis of our frame of reference. Rotations by the Euler angles [48] $\omega_i = (\varphi_i,\vartheta_i,0)$ will put the OZ axis along the quantization direction $\hat{u}_i$ of the spin of the electron e_i emitted in the 3-TPI process (1).

Let us represent by (E_0,E_1,E_2,E_f) the energies of the electronic states $(|0\rangle,|1\rangle,|2\rangle,|f\rangle)$ of $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in the 3-TPI process (1). Energy should obviously be conserved in each of the three steps of the process (1) separately. This means, while kinetic energy $\epsilon_1 = h\nu_r - (E_1 - E_0)$ of the photoelectron e_1 varies with the frequency of the ionizing radiation in Eq. (1a); energies $\epsilon_2 = (E_1 - E_2)$ of the Auger electron e_2 in Eq. (1b) and $\epsilon_3 = (E_2 - E_f)$ of the Auger electron e_3 in Eq. (1c) are completely independent of the energy of the absorbed photon and are totally determined from those of (A^{+^*},A^{2+^*}) and (A^{2+^*},A^{3+}), respectively.

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 $(\gamma_r + A)$ system in the step (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, ρ_0 of the target atom A, and ρ_r of the ionizing radiation γ_r. The DO for the photoionization of the atom A, taking place in the first of the three-step process (1), is given by (see, for example, [49,50])

$$\rho_1 = \mathcal{K}_{p_1} F_{p_1} \rho_i F^{\dagger}_{p_1}. \tag{2a}$$

The photoionization operator F_{p_1} and the quantity $\mathcal{K}_{p_1}$ (both in the $E1$ approximation) are obtained, for example, from [51]. The DO for the complete three-step process (1) can now readily be written to be

$$\begin{aligned}\rho_f &= \mathcal{K}_{a_3} F_{a_3} (\mathcal{K}_{a_2} F_{a_2} \rho_1 F^{\dagger}_{a_2}) F^{\dagger}_{a_3} \\ &= \mathcal{K}[F_{a_3} F_{a_2} F_{p_1}](\rho_0 \otimes \rho_r)[F_{a_3} F_{a_2} F_{p_1}]^{\dagger}.\end{aligned} \tag{2b}$$

Here, (F_{a_2},F_{a_3}) are the operators for the emission of the Auger electrons (e_2,e_3) in the (second and third) steps of the three-step process (1). These operators and their associated quantities $(\mathcal{K}_{a_2},\mathcal{K}_{a_3})$, occurring in Eq. (2b), are given elsewhere [52]. Although, the explicit forms of none of $(\mathcal{K}_{p_1},\mathcal{K}_{a_2},\mathcal{K}_{a_3})$ are required for the following discussions, it should—nevertheless—be mentioned that they depend, among other things, on the energies $(\epsilon_1,\epsilon_2,\epsilon_3)$ of electrons (e_1,e_2,e_3) but do not involve any of the angle- or spin-related quantities of $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in the process (1). On the right-hand side of the second equation (2b), we have defined—for brevity—$\mathcal{K} \equiv \mathcal{K}_{a_3}\mathcal{K}_{a_2}\mathcal{K}_{p_1}$. The form (2) of the DO is equivalent to those given elsewhere (see, e.g., [53]) for other more than two-step processes, e.g., three-stage cascade of γ rays in nuclear physics, etc.

In order to perform the desired investigations, we need to calculate a matrix for the DO ρ_f defined in Eq. (2). Electrons (e_1,e_2,e_3) emitted in the 3-TPI may have same $[\epsilon_1 = \epsilon_2 = \epsilon_3 (ie, |\vec{k}_1| = |\vec{k}_2| = |\vec{k}_3|)]$ or different $[\epsilon_1 \neq \epsilon_2 \neq \epsilon_3 (ie, |\vec{k}_1| \neq |\vec{k}_2| \neq |\vec{k}_3|)]$ energies [subject to satisfying three different conservation conditions, one for each step in Eq. (1)] and move in any three directions $\hat{k}_1$, $\hat{k}_2$, and $\hat{k}_3$. Our study requires a density matrix which is diagonal in energies as well as in the directions of propagation (i.e., diagonal in $\vec{k}_1$, $\vec{k}_2$, and $\vec{k}_3$). But this DM must necessarily be nondiagonal with respect to the components (μ_1,μ_2,μ_3) along $(\hat{u}_1,\hat{u}_2,\hat{u}_3)$ of the spin angular momenta of (e_1,e_2,e_3). The Appendix of this paper describes the calculation of such a DM without taking spin-orbit interaction (SOI) into account.

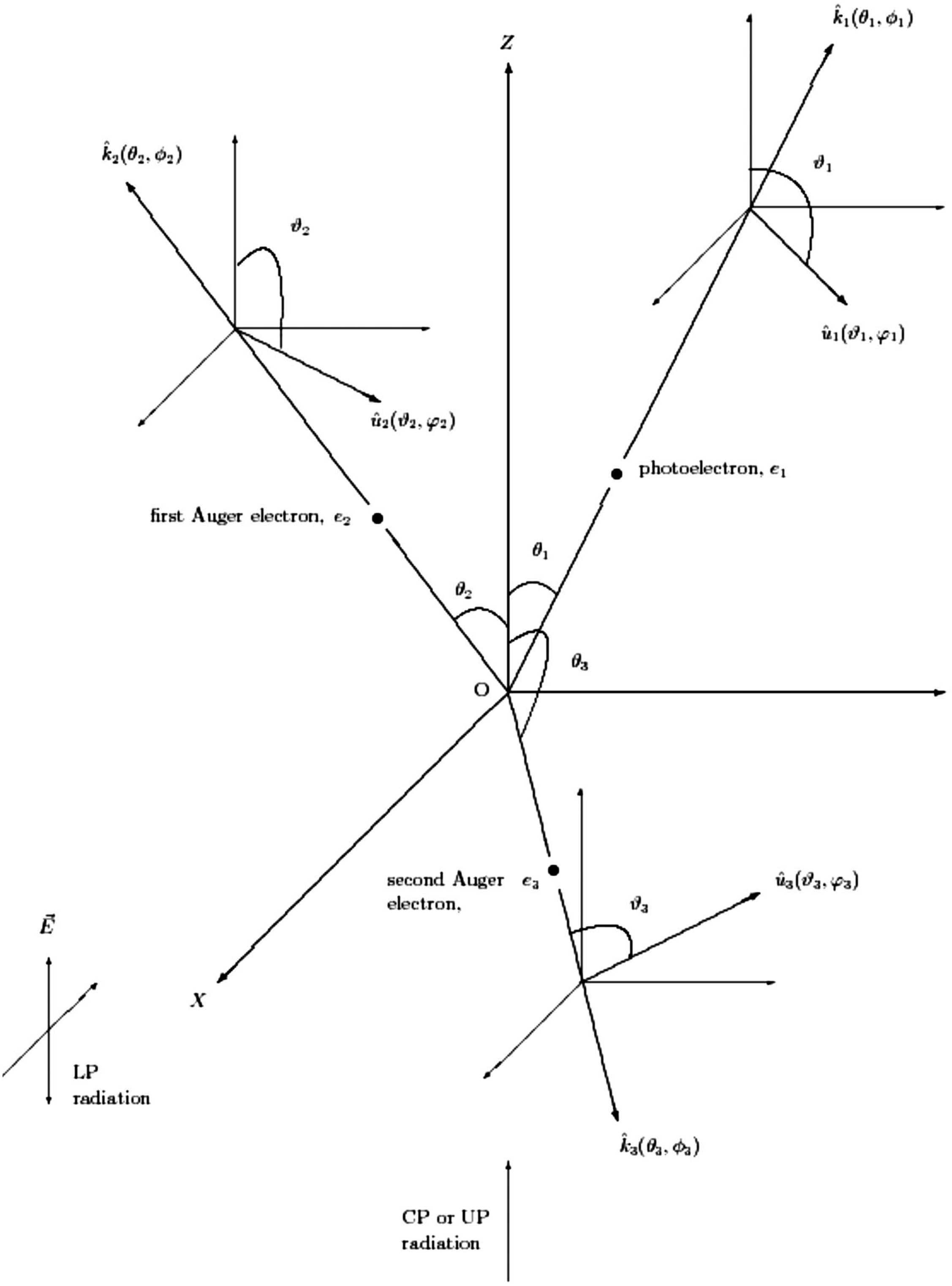

FIG. 1. Coordinate system showing the photon-fixed frame of reference *OXYZ* and the propagation directions $(\hat{k}_1, \hat{k}_2, \hat{k}_3)$ as well as the spin-quantization directions $(\hat{u}_1, \hat{u}_2, \hat{u}_3)$ of the photoelectron e_1 as well as of the first and of the second Auger electrons e_2 and e_3, respectively. These three electrons are sequentially ejected from the atom A situated at the origin O of our coordinate system, following the absorption of a single photon. The OZ axis is the quantization direction for the electronic states $(|0\rangle, |1\rangle, |2\rangle, |f\rangle)$ of the atomic species $(A, A^{+^*}, A^{2+^*}, A^{3+})$ participating in the 3-TPI process (1). Each electron enters its own Mott detector [47] oriented along $[\hat{u}_1(\vartheta_1, \varphi_1), \hat{u}_2(\vartheta_2, \varphi_2), \hat{u}_3(\vartheta_3, \varphi_3)]$ for $[e_1, e_2, e_3]$. These detectors [47] record whether the spin of the ejected electron e_i is up or down with respect to its own quantization direction $\hat{u}_i$.

III. DENSITY MATRIX FOR TRIPARTITE SPIN STATE OF ELECTRONS (e_1,e_2,e_3) EMITTED IN THE 3-TPI PROCESS (1)

The two most important forces experienced by the constituents of an atom are the electrostatic Coulomb interaction among nucleus and electrons and those which may arise due to spins of the electrons [in A, A^{+^*}, A^{2+^*}, A^{3+}, and of (e_1,e_2,e_3)] participating in one or more of the steps of the process (1). Most of the important properties of any atom A are predominantly determined from its Coulomb interaction as it is always much stronger in comparison to the weak perturbative spin-dependent forces [e.g., SOI]. Moreover, almost all [32–37], both experimental and theoretical, studies of atomic 3-TPI have hitherto been performed by taking only the Coulomb interactions into account. In our present study as well, we disregard all the spin-dependent forces which may exist inside an atom. This, in other words, means that the entanglement properties of the tripartite spin state of (e_1,e_2,e_3) formed in the 3-TPI process (1) in a generic atom A will due purely be to the electrostatic Coulomb forces acting among the atomic constituents. It can, therefore, be called as Coulombic spin entanglement of (e_1,e_2,e_3). Thus, the environment or conditions external to the atom A play no role whatsoever in the generation of the tripartite entanglement being discussed in this paper. The generated entanglement should, therefore, be free from the harmful effects of decoherence and dissipation as the three electrons ejected in the process (1) recede from the residual atomic ion A^{3+}.

Further, both the $E1$ photoionization and two Auger emission operators F_{p_1} and (F_{a_2},F_{a_3}), respectively, are spin independent. In the absence of such spin-dependent interactions which—in the case of an atom—is primarily SOI, the orbital and spin angular momenta in each of the three steps of the process (1) are separately conserved. These conservation conditions are expressed in Eqs. (A1)–(A3) in the Appendix of this paper. This, in other words, means that the L-S (i.e., Russell-Saunders) coupling [46] naturally becomes applicable in each step of Eq. (1).

We have derived an expression for the desired DM. Our derivation, whose details are given in the Appendix of this paper, 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 3-TPI process (1) in any atom. The final expression (A9) for the DM can be written as a product of two independent terms which describe two entirely different physical situations. It is obvious from Eq. (A10) that the first term [i.e., $d^4\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3$] on the right-hand side of the DM (A9) depends upon—among other things—the orbital angular momentum (L_0,L_1,L_2,L_f) of each of $(A,A^{+^*},A^{2+^*},A^{3+})$, phase shifts, energies $(\epsilon_1,\epsilon_2,\epsilon_3)$, and directions $(\hat{k}_1,\hat{k}_2,\hat{k}_3)$ of emission of electrons (e_1,e_2,e_3), including the polarization (m_r) of the photon absorbed in the first of the three-step process (1). This term, in addition, contains both the $E1$ amplitude (A12a) (determined by the electronic states $|0\rangle$ of A, $|1\rangle$ of A^{+^*}, and the continuum orbital of e_1) for photoionization (1a), as well as the amplitudes (A12b) and (A12c) [obtained, respectively, from the (electronic states $|1\rangle$ of A^{+^*}, $|2\rangle$ of A^{2+^*} and continuum orbital of e_2) and (electronic states $|2\rangle$ of A^{2+^*}, $|f\rangle$ of A^{3+}, and continuum orbital of e_3)] for the Auger emissions in steps (1b) and (1c), respectively. Thus, $d^4\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3$ in Eq. (A9) very much depends upon both the kinematics and dynamics of the three-step process (1). But it includes neither the spins nor the quantization directions $(\hat{u}_1,\hat{u}_2,\hat{u}_3)$ of any of the three emitted electrons (e_1,e_2,e_3) or of the atomic species $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in the 3-TPI process (1). Hence, $d^4\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3$ [i.e., expression (A10)] in the DM (A9) describes purely angular correlation between the three electrons (e_1,e_2,e_3) in the L-S coupling. Its value is always *positive* and it acts merely as a *multiplicative* factor in the DM (A9) for a given Auger process (1).

The second term [i.e., $\sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}$] present on the right-hand side of the DM (A9) is defined in Eq. (A11). Unlike the angular correlation in Eq. (A10), $\sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}$ represents a (8×8) matrix. It contains neither any of those physical quantities (e.g., m_r, L_0, L_1, L_2, L_f, $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3$, etc.) nor any of the three dynamical amplitudes (A12a)–(A12c) which are present in the angular correlation $d^4\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3$. On the other hand, $\sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}$ is completely determined from Eq. (A11) by the spins of all the seven particles (i.e., A, A^{+^*}, A^{2+^*}, A^{3+^*}, e_1, e_2, and e_3) participating in the three steps of the process (1), in addition to the quantization directions $(\hat{u}_1,\hat{u}_2,\hat{u}_3)$ [see Fig. 1]—and the components (μ_1,μ_2,μ_3) along these unit vectors—of the spins of electrons (e_1,e_2,e_3). Hence, the second term on the right-hand side of the DM (A11) describes purely spin correlation between the three electrons (e_1,e_2,e_3) ejected sequentially in the 3-TPI process (1) in the absence of SOI. Equation (A13) contains simplified expressions for an easy calculation of this DM.

IV. CHARACTERIZATION OF THE SPIN ENTANGLEMENT OF (e_1,e_2,e_3)

A. Methodology

Dür *et al.* [39] fully classified arbitrary three-qubit mixed states with respect to their separability and distillability properties. Later on, Dür and Cirac [40] generalized this [39] scheme in the form of a hierarchic classification for arbitrary multi-qubit, mixed states based on the separability properties of certain partitions. Both of these schemes provide sufficient conditions for separability and distillability for tripartite [39] and multipartite [40] arbitrary states of qubits. Sharma and Sharma [54], on the other hand, suggested a K-way negativities approach for characterizing multipartite entanglement of qubits.

The classification schemes developed in [39,40,54] are based on the stringent Peres [41]-Horodecki *et al.* [42] condition of positive partial transposition for the separability of a bipartite state of two qubits. Although, the partial transpose (PT) of any operator may be different in different bases, however, its eigenvalues do not change with a change in the basis. By using random local operations, one can always convert [39,40] any three-qubit state to the form of Eq. (3) in [39] [or Eq. (6) in [40]]. In this paper, we have used the classification scheme suggested in Refs. [39,40] for characterizing the entanglement properties of a tripartite state of the flying electronic qubits (e_1,e_2,e_3) generated in the 3-TPI process (1).

It has already been discussed elsewhere in this paper that $d^4\sigma(m_r)/d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3$ in Eq. (A9) describes purely angular correlation of (e_1,e_2,e_3) in the L-S coupling and always has a single positive value for a given experimental configuration. Consequently, the properties of the Coulomb generated spin entanglement among (e_1,e_2,e_3) in the present case will completely be determined by the spin-correlation matrix (A11). We, therefore, do not write in the following the angular correlation (A10) explicitly and represent—for brevity—the tripartite DM simply by the second term present on the right-hand side of Eq. (A9). That is, unless stated otherwise, we imply

$$\begin{aligned}&\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f|f;\\ &\quad\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle\\ &\quad\Leftrightarrow \sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}\end{aligned} \tag{3a}$$

and write, for brevity,

$$\sigma(S;\omega)_{\mu;\mu'} \equiv \sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}. \tag{3b}$$

Here, and in the following, $S\equiv(S_0,S_1,S_2,S_f)$ and each of ω,μ,μ' stands for the triods $(\omega_1,\omega_2,\omega_3)$, (μ_1,μ_2,μ_3), and (μ_1',μ_2',μ_3'), respectively. The PT [41] of this DM with respect to the photoelectron e_1, first Auger electron e_2, and the second Auger electron e_3 are, respectively, given by

$$\begin{aligned}\sigma^{T_{e_1}} &\equiv \langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f^{T_1}|f;\\ &\quad\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle\\ &= \sigma(S;\omega)_{\mu_1'\mu_2\mu_3;\mu_1\mu_2'\mu_3'},\end{aligned} \tag{4a}$$

$$\begin{aligned}\sigma^{T_{e_2}} &\equiv \langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f^{T_2}|f;\\ &\quad\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle\\ &= \sigma(S;\omega)_{\mu_1\mu_2'\mu_3;\mu_1'\mu_2\mu_3'},\end{aligned} \tag{4b}$$

and

$$\begin{aligned}\sigma^{T_{e_3}} &\equiv \langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f^{T_3}|f;\\ &\quad\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle\\ &= \sigma(S;\omega)_{\mu_1\mu_2\mu_3';\mu_1'\mu_2'\mu_3}.\end{aligned} \tag{4c}$$

According to the classification scheme suggested in Refs. [39,40], the positivity or otherwise of $\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}$, and of $\sigma^{T_{e_3}}$ decides about the full separability, biseparability, and full inseparability of the tripartite spin state $\sigma(S;\omega)$ of (e_1,e_2,e_3) emitted in the 3-TPI process (1).

It is possible that in a particular experiment, one may be interested in any two [say, (e_i,e_j) with $i\neq j$] of the three (e_1,e_2,e_3) electrons emitted in the process (1), or, the party holding electron e_k (with $k\neq i,j$) decides not to cooperate with the other two. Such kinds of situations will be described by the reduced DOs and DMs

$$\begin{aligned}\sigma(S;\omega_1,\omega_2) &\equiv \mathrm{Tr}_3[\sigma(S;\omega)],\\ &\quad\text{or}\quad \sigma(S;\omega_1,\omega_2)_{\mu_1\mu_2;\mu_1'\mu_2'}\\ &\quad= \sum_{\mu_3}\sigma(S;\omega)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3};\end{aligned} \tag{5a}$$

$$\begin{aligned}\sigma(S;\omega_2,\omega_3) &\equiv \mathrm{Tr}_1[\sigma(S;\omega)],\\ &\quad\text{or}\quad \sigma(S;\omega_2,\omega_3)_{\mu_2\mu_3;\mu_2'\mu_3'}\\ &\quad= \sum_{\mu_1}\sigma(S;\omega)_{\mu_1\mu_2\mu_3;\mu_1\mu_2'\mu_3'};\end{aligned} \tag{5b}$$

$$\begin{aligned}\sigma(S;\omega_1,\omega_3) &\equiv \mathrm{Tr}_2[\sigma(S;\omega)],\\ &\quad\text{or}\quad \sigma(S;\omega_1,\omega_3)_{\mu_1\mu_3;\mu_1'\mu_3'}\\ &\quad= \sum_{\mu_2}\sigma(S;\omega)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2\mu_3'}\end{aligned} \tag{5c}$$

of the resulting bipartite state. These are the only three bipartite states which can be formed from a tripartite state of (e_1,e_2,e_3). Each of the states (5a)–(5c) will determine also the properties of a two-qubit state formed in a certain bipartite split [39,40] of a tripartite state. In order to study the properties of each of these three bipartite states, we have written it, wherever possible, in the form of a Werner state [55] and calculated its concurrence [43,44]. It has helped us in studying the properties also of the bipartite part of a biseparable state of (e_1,e_2,e_3). The state (5a) is identical to that obtained by us in Eq. (A15) in Ref. [29] for studying spin-entanglement properties of two electrons emitted in 2-DPI. Equations (A17a)–(A17c) herein give explicit expressions for the DMs representing the bipartite states (5a)–(5c).

B. Results

An application of the rules [46,48] of vector coupling of angular momenta of three spin-$\frac{1}{2}$ particles in quantum mechanics suggests that the total spin of (e_1,e_2,e_3) can be $j=\frac{1}{2},\frac{3}{2}$. How many or which of these two values will contribute to a given process (1) will be determined from the spin conservation conditions (A1)–(A3), as well as from the two 6-j and one 9-j symbols [46,48] present in Eq. (A11). The required conditions to be satisfied are $|S_0-S_1|$, $|S_1-S_2|$, and $|S_2-S_f|=1/2$. These, in turn, imply that S_0 of A and S_f of A^{3+} can differ by 1/2 and/or 3/2. Then the DM (A11) will represent states of (e_1,e_2,e_3) for all those allowed values of j which lie in the range $|S_0-S_f|$ to $|S_0+S_f|$ for given spins (S_0,S_1,S_2,S_f). Consequently, there will be states of (e_1,e_2,e_3) with $j=1/2$ or 3/2 or mixture of states with $j=1/2$ and 3/2.

The rank [3] [i.e., the number of nonzero eigenvalues of $\sigma(S;\omega)_{\mu;\mu'}$] of the state (3) of (e_1,e_2,e_3) with total spin angular momentum j was always found to be equal to $(2j+1)$. In other words, the 3-TPI process (1) can generate spin states of (e_1,e_2,e_3) possessing rank two (for $j=1/2$), four (for $j=3/2$), or six (for $j=1/2$ and 3/2). A pure state is always of rank one [50]. In other words, (e_1,e_2,e_3) herein will never be in a pure tripartite state. However, it is always possible that one or more of the bipartite state (5) formed in the process (1) is pure. We will show in the following that all the properties of a tripartite state of (e_1,e_2,e_3) relevant to a quantum information study are readily determined from the spins (S_0,S_1,S_2,S_f) of $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in Eq. (1).

We have calculated the tripartite states $\sigma(S;\omega)$ defined in Eq. (3), for a large number of 3-TPI process (1) with different permissible values of (S_0,S_1,S_2,S_f). Before studying these states according to the schemes suggested in [39,40] or elsewhere (e.g., in Refs. [56,57]), we would like to first mention some general rules based on the allowed values of (S_0,S_1,S_2,S_f), specifying the properties of the bipartite state (5) formed from a given tripartite state of (e_1,e_2,e_3). It is useful because, in addition to other reasons, almost all the procedures [39,40,54,56,57] hitherto suggested for investigating entanglement of a three-qubit state are mostly based on the properties of its bipartite states which can be formed according to the procedure explained in Eq. (5).

1. Properties of the bipartite states formed by any two of the three electrons (e_1,e_2,e_3)

Any bipartite state (5) formed of the qubits (e_i,e_j) can, in general, be represented by

$$\sigma(S;\omega_i,\omega_j)=p_{ij}\sigma_1(\omega_i,\omega_j)+(1-p_{ij})\sigma_0. \qquad (6)$$

Here, $\sigma_1(\omega_i,\omega_j)$ is a pure maximally entangled singlet state (i.e., one of the four Bell states [1–3]) of the ith and jth spin-$\frac{1}{2}$ particles defined in Eq. (A18); whereas, $\sigma_0=I/4$, with I a (4×4) unit matrix, is a maximally chaotic state [58] which is also called the white-noise states [59]. The p_{ij} (≤ 1 always) present in Eq. (6) is called a mixing parameter as it determines the amount of σ_0 mixed with σ_1. Thus, $\sigma(S;\omega_i,\omega_j)$ in Eq. (6) becomes the Bell state $\sigma_1(\omega_i,\omega_j)$ for $p_{ij}=1$ or the maximally chaotic state σ_0 if $p_{ij}=0$. For $0<p_{ij}<1$, on the other hand, Eq. (6) represents a Werner state [55]; otherwise, i.e., with $p_{ij}<0$, $\sigma(S;\omega_i,\omega_j)$ is a non-Werner state. Electrons (e_i,e_j) in the state (6) are always entangled if $p_{ij}>1/3$. The eigenvalues of the bipartite state (6) and of its PT are [29]

$$(1-p_{ij})/4,(1-p_{ij})/4,(1-p_{ij})/4,(1+3p_{ij})/4 \qquad (7a)$$

and

$$(1+p_{ij})/4,(1+p_{ij})/4,(1+p_{ij})/4,(1-3p_{ij})/4, \qquad (7b)$$

respectively.

It is obvious that a value of the mixing parameter p_{ij} will very much depend upon the spin quantum numbers (S_0,S_1,S_2,S_f) of the electronic states $(|0\rangle,|1\rangle,|2\rangle,|f\rangle)$ of $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in the 3-TPI process (1). The mixing parameter p_{12} in Eq. (6) for the bipartite state $\sigma(S;\omega_1,\omega_2)$ [defined in Eqs. (5a) and (A17a)] formed of the photoelectron e_1 and first (i.e., e_2) of the two Auger electrons is given by

$$p_{12}=S_1/[3(S_1+1)] \ \text{ for } \ S_0=S_2=S_1+\frac{1}{2}, \quad S_1\geq 0, \qquad (8a)$$

$$p_{12}=(S_1+1)/(3S_1) \ \text{ for } \ S_0=S_2=S_1-\frac{1}{2}, \quad S_1\geq \frac{1}{2}, \qquad (8b)$$

$$p_{12}=-\frac{1}{3} \quad \text{for}|S_0-S_2|=1. \qquad (8c)$$

Similarly, for the bipartite state $\sigma(S;\omega_2,\omega_3)$ [defined in Eqs. (5b) and (A17b)] consisting of both Auger electrons (e_2,e_3), we have

$$p_{23}=S_2/[3(S_2+1)] \ \text{ for } \ S_1=S_f=S_2+\frac{1}{2}, \quad S_2\geq 0, \qquad (9a)$$

$$p_{23}=(S_2+1)/(3S_2) \ \text{ for } \ S_1=S_f=S_2-\frac{1}{2}, \quad S_2\geq \frac{1}{2}, \qquad (9b)$$

$$p_{23}=-\frac{1}{3} \ \text{ for } \ |S_1-S_f|=1. \qquad (9c)$$

The mixing parameter for the remaining bipartite state $\sigma(S;\omega_1,\omega_3)$ specified by Eqs. (5c) and (A17c) and consisting of the photoelectron e_1 and the second Auger electron e_3 is

$$p_{13}=\Delta\left(S_0S_1\frac{1}{2}\right)\Delta\left(S_1S_2\frac{1}{2}\right)\Delta\left(S_2S_f\frac{1}{2}\right)$$
$$\times\frac{1}{3}(1-\delta_{0S_1})(1-\delta_{0S_2})\mathbb{P}_{13}. \qquad (10)$$

Here, $\mathbb{P}_{13}$ and Δ are given by Eqs. (A14) and (A15), respectively, while, δ_{ab} [48] is the Kronecker delta function.

It is interesting to find that each of the mixing parameters p_{12} in Eq. (8) and p_{23} in Eq. (9) is determined by the spins of only three of the four atomic species $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in the 3-TPI process (1). Namely, the former contains (S_0,S_1,S_2) of (A,A^{+^*},A^{2+^*}) and the latter involves (S_1,S_2,S_f) of $(A^{+^*},A^{2+^*},A^{3+})$. But, the mixing parameter p_{13} in Eqs. (10) and (A14) contains spins of all the four atomic particles present in Eq. (1). Equations (8b) and (9b) are the respective conditions for each of the pairs (e_1,e_2) and (e_2,e_3) to form an entangled state; otherwise, both or either of the states $\sigma(S;\omega_1,\omega_2)$ and $\sigma(S;\omega_2,\omega_3)$ is separable which may [for $0<p_{ij}<1/3$ from Eq. (8a) or Eq. (9a)] or may not [for $p_{ij}<0$ from Eq. (8c) or Eq. (9c)] be a Werner state. Similarly, one can determine from a value of p_{13} calculated from

TABLE I. Properties of the bipartite states (6) which can be formed from a given tripartite state (3) of electrons (e_1,e_2,e_3) generated in the 3-TPI process (1).

	$\sigma(S;\omega_1,\omega_2)$ of electrons (e_1,e_2)	$\sigma(S;\omega_2,\omega_3)$ of electrons (e_2,e_3)	$\sigma(S;\omega_1,\omega_3)$ of electrons (e_1,e_3)
(i,j) in Eq. (6) and in Eq. (A18)	(1, 2)	(2, 3)	(1, 3)
Maximally chaotic state σ_0	Eq. (8a): $p_{12}=0$; $S_0=\frac{1}{2}$, $S_1=0$, $S_2=\frac{1}{2}$	Eq. (9a): $p_{23}=0$; $S_1=\frac{1}{2}$, $S_2=0$, $S_f=\frac{1}{2}$	Eq. (10): $p_{13}=0$; (S_0,S_1,S_2,S_f) with Eqs. (A1)–(A3)
Bell state: Eq. (A18), singlet state of two spin-$\frac{1}{2}$ particles	Eq. (8b): $p_{12}=1$; Eq. (11a): $\mathcal{C}_{12}=1$; $\sigma_1(\omega_1,\omega_2)$; $S_0=0$, $S_1=\frac{1}{2}$, $S_2=0$	Eq. (9b): $p_{23}=1$; Eq. (11b): $\mathcal{C}_{23}=1$; $\sigma_1(\omega_2,\omega_3)$; $S_1=0$, $S_2=\frac{1}{2}$, $S_f=0$	Eq. (10): $p_{13}=1$; Eq. (11c): $\mathcal{C}_{13}=1$; $\sigma_1(\omega_1,\omega_3)$; (S_0,S_1,S_2,S_f) with Eqs. (A1)–(A3)
Separable Werner state	$0<p_{12}<\frac{1}{3}$; Eq. (8a): $p_{12}=S_1/[3(S_1+1)]$; $S_0=S_2=S_1+\frac{1}{2}$ with $S_1>0$	$0<p_{23}<\frac{1}{3}$; Eq. (9a): $p_{23}=S_2/[3(S_2+1)]$; $S_1=S_f=S_2+\frac{1}{2}$ with $S_2>0$	$0<p_{13}<\frac{1}{3}$; Eq. (10): p_{13}; (S_0,S_1,S_2,S_f) with Eqs. (A1)–(A3)
Nonseparable Werner state	$\frac{1}{3}<p_{12}<1$; Eq. (8b): $p_{12}=(S_1+1)/(3S_1)$; Eq. (11a): $\mathcal{C}_{12}=1/(2S_1)$; $S_0=S_2=S_1-\frac{1}{2}$ with $S_1>\frac{1}{2}$	$\frac{1}{3}<p_{23}<1$; Eq. (9b): $p_{23}=(S_2+1)/(3S_2)$; Eq. (11b): $\mathcal{C}_{23}=1/(2S_2)$; $S_1=S_f=S_2-\frac{1}{2}$ with $S_2>\frac{1}{2}$	$\frac{1}{3}<p_{13}<1$; Eq. (10): p_{13}; Eq. (11c): $\mathcal{C}_{13}$; (S_0,S_1,S_2,S_f) with Eqs. (A1)–(A3)
Separable non-Werner state	Eq. (8c): $p_{12}=-\frac{1}{3}$; $\lvert S_0-S_2\rvert=1$	Eq. (9c): $p_{23}=-\frac{1}{3}$; $\lvert S_1-S_f\rvert=1$	Eq. (10): $p_{13}<0$; (S_0,S_1,S_2,S_f) with Eqs. (A1)–(A3)

Eqs. (10) and (A14), whether $\sigma(S;\omega_1,\omega_3)$ is an entangled or separable Werner state, or a product non-Werner state of electrons (e_1,e_3).

Furthermore, the concurrence [43,44] for the entangled state (6) of electrons (e_1,e_2) with mixing parameter (8) is

$$\mathcal{C}_{12}=(2S_1)^{-1}. \tag{11a}$$

In the case of the mixing parameter (9), the concurrence of the state (6) representing Auger electrons (e_2,e_3) becomes

$$\mathcal{C}_{23}=(2S_2)^{-1}. \tag{11b}$$

The concurrence for the bipartite state (5c) of the pair of electrons (e_1,e_3) is, on the other hand, obtained from

$$\mathcal{C}_{13}=\max\left[0,\frac{3p_{13}-1}{2}\right] \tag{11c}$$

with p_{13} given by Eq. (10). Substitution in Eq. (11c) of the mixing parameters p_{12} from Eq. (8b) or of p_{23} from Eq. (9b) will give us the concurrence (11a) or Eq. (11b), respectively.

Table I contains a summary of the properties of all the three bipartite spin state (5) which can be formed from a given tripartite state (3) of (e_1,e_2,e_3) emitted in the 3-TPI process (1).

2. *Fully separable states of* (e_1,e_2,e_3)

Our studies showed that for all the states of (e_1,e_2,e_3) with $\lvert S_0-S_f\rvert=3/2$, each of the three PTs defined in Eqs. (4a)–(4c), $\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}$, and $\sigma^{T_{e_3}}\geq 0$. These are the states of rank four. Their bipartite reduced DMs (5a)–(5c) are obtained on taking $p_{ij}=-1/3$ in Eq. (6) for $(i,j)=(1,2)$, (2, 3), or (1, 3). It has already been discussed elsewhere in this paper that although Eq. (6) is not in the form of a Werner state [55] with this value of the mixing parameter, it, nevertheless, describes a mixed, separable, and bipartite spin state of any two of the three electrons (e_1,e_2,e_3) ejected in the 3-TPI process (1). Thus, all rank four states generated in the process (1) with $\lvert S_0-S_f\rvert=3/2$ are of the fully separable form [39,40] $\lvert e_1\rangle\otimes\lvert e_2\rangle\otimes\lvert e_3\rangle$.

In addition to this, we found that tripartite states of (e_1,e_2,e_3) generated in Eq. (1) for $\lvert S_0-S_f\rvert=1/2$ and $S_0+S_f\geq 3/2$ provided $S_0>S_1$ and $S_2<S_f$ were also fully separable. Rank of these states is six because now the total spin angular

momentum j of (e_1,e_2,e_3) can simultaneously be 1/2 and 3/2. However, unlike in the case of the fully separable states of rank four, the bipartite state (5) formed from the presently generated states of (e_1,e_2,e_3) can be of Werner type as well—including also the white-noise state [58,59] σ_0 present in Eq. (6). These states are as follows:

(a) $S_0{=}S_2{=}S_1{+}1/2{=}S_f{-}1/2$. Auger electrons (e_2,e_3) are again in the state (6) with $p_{23}{=}{-}1/3$, i.e., a non-Werner state. But, each of two pairs formed with the photoelectron e_1 [i.e., (e_1,e_2) and (e_1,e_3)] is in a state usually different from that of (e_2,e_3) with the mixing parameters given by the respective Eqs. (8a) and (10) which, in the present case, are equal, i.e., $p_{12}{=}p_{13}$. Thus, each of the three pairs of emitted electrons in the present case is in a product state and, hence, (e_1,e_2,e_3) are in a fully separable state [39,40]. For example, for $(S_0{=}S_2{=}1/2,S_1{=}0,S_f{=}1)$, the two pairs (e_1,e_3) and (e_1,e_2) are in the maximally chaotic bipartite state σ_0. For $(S_0{=}S_2{=}1,S_1{=}1/2,S_f{=}3/2)$, on the other hand, the state of (e_1,e_3) and of (e_1,e_2) is Eq. (6) with $p_{13}{=}p_{12}{=}1/9$ obtained from Eq. (8a) or Eq. (10), while (e_2,e_3) continue to be in the states (6) and (9c).

(b) $S_1{=}S_f{=}S_0{-}1/2{=}S_2{+}1/2$. Now, Eq. (6), with $p_{12}{=}{-}1/3$, happens to be a state of the qubits (e_1,e_2), while the separable states (6) and (9a) or Eqs. (6) and (10) describe each of the pairs (e_2,e_3) and (e_1,e_3). For example, the latter two pairs are in the state (6) with $p_{13}{=}p_{23}{=}0$ or 1/9 for $(S_0,S_1,S_2,S_f){=}(1,1/2,0,1/2)$ or (3/2, 1, 1/2, 1), respectively; whereas, (e_1,e_2) are always in the non-Werner state (6) and (8c) in each of these two and other similar situations.

3. One-qubit biseparable states of (e_1,e_2,e_3)

These are the tripartite state (3) of (e_1,e_2,e_3) which have only one of the three PTs (4a)–(4c) positive [39,40]. We found that some of the states of rank two (i.e., $j{=}1/2$) and some of rank six (i.e., $j{=}1/2$ and 3/2), while none of rank four (i.e., $j{=}3/2$) has this property. We discuss, in the following, all the three possibilities [39,40] which may arise in this case.

(a) $\sigma^{T_{e_1}}{\geq}0$. All tripartite state (3) generated in the 3-TPI process (1) with $S_1{=}S_f{=}S_0{-}1/2{=}S_2{-}1/2$ have this characteristic. These states [39,40] can be written as e_1-(e_2,e_3). The entangled Werner state of (e_2,e_3) is Eqs. (6) and (9b). Here, for example, with $(S_0,S_1,S_2,S_f){=}(1/2,0,1/2,0)$, (e_2,e_3) are in the Bell state (A18); whereas, each of the remaining two pairs (e_1,e_2) and (e_1,e_3) is in the maximally chaotic state σ_0 (i.e., p_{12} [Eq. (8a)] $=0{=}p_{13}$ [Eq. (10)]). For $(S_0,S_1,S_2,S_f){=}(1,1/2,1,1/2)$, on the other hand, the respective states of (e_2,e_3), (e_1,e_3), and (e_1,e_2) are obtained from Eqs. (6) and (9b) with $p_{23}{=}2/3$ (i.e., a mixed, entangled Werner state), Eqs. (6) and (10) with $p_{13}{=}{-}2/9$ (i.e., a mixed, separable non-Werner state), and from Eqs. (6) and (8a) with $p_{12}{=}1/9$ (i.e., a mixed, separable Werner state). According to [40], the mixed entangled state $\sigma(S;\omega_2,\omega_3)$ can be distilled to the Bell state $\sigma_1(\omega_2,\omega_3)$ because, in the present case, both $\sigma^{T_{e_2}}$ are $\sigma^{T_{e_3}}$ are negative.

(b) $\sigma^{T_{e_3}}{\geq}0$. These are e_3-(e_1,e_2) states produced whenever $S_1{=}S_f{=}S_0{+}1/2{=}S_2{+}1/2$ in the 3-TPI process (1). Equations (6) and (8b) represent the nonseparable state of (e_1,e_2). On taking $(S_0,S_1,S_2,S_f){=}(0,1/2,0,1/2)$ in the process (1), for example, one will generate Bell state $\sigma_1(\omega_1,\omega_2)$ [Eq. (A18)] for electrons (e_1,e_2) and white-noise states σ_0 for the remaining two pairs (e_2,e_3) and (e_1,e_3) of ejected electrons; but, 3-TPI with $(S_0,S_1,S_2,S_f){=}(1/2,1,1/2,1)$ will give us states $[\sigma(S;\omega_1,\omega_2),\sigma(S;\omega_2,\omega_3),\sigma(S;\omega_1,\omega_3)]$ with $p_{12}{=}2/3$ [Eqs. (6) and (8b)], $p_{23}{=}1/9$ [Eqs. (6) and (9a)], and $p_{13}{=}{-}2/9$ [Eqs. (6) and (10)]. The presence of the PTs $\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}$ <0 suggests [40] that one can distill $\sigma(S;\omega_1,\omega_2)$ from a mixed to a maximally entangled pure state of (e_1,e_2).

(c) $\sigma^{T_{e_2}}{\geq}0$. Except in the case of fully separable tripartite states of (e_1,e_2,e_3) already discussed herein in the Sec. IV B 2, $\sigma^{T_{e_2}}$ was always less than zero for any 3-TPI process (1). This, in other words, means [39,40] that the process (1) is incapable of generating tripartite states of (e_1,e_2,e_3) which are one qubit biseparable to the form e_2-(e_1,e_3).

4. Fully inseparable states of (e_1,e_2,e_3)

In all other cases, the tripartite states of (e_1,e_2,e_3) have all the three PTs $(\sigma^{T_{e_1}},\sigma^{T_{e_2}},\sigma^{T_{e_3}})$ negative. According to [40], one can now distill a Greenberger-Horne-Zeilinger (GHZ) [60] state $|\mathrm{GHZ}\rangle$. Here, the reduced DMs (5a)–(5c) showed that entangled bipartite states are either of the photoelectron e_1 with the Auger electrons (e_2,e_3), or of e_3 with (e_1,e_2). Thus, one can connect [61] either $[\sigma(S;\omega_1,\omega_2),\sigma(S;\omega_1,\omega_3)]$ or $[\sigma(S;\omega_1,\omega_3),\sigma(S;\omega_2,\omega_3)]$ to produce a $|\mathrm{GHZ}\rangle$ state [60]. This also means that the first Auger electron e_2 is never simultaneously entangled with (e_1,e_3).

These correspond to the situation wherein one electron is entangled with the remaining two electrons present in a tripartite state of (e_1,e_2,e_3). Properties of such states was discussed by Dür [56] and by Lohmayer *et al.* [57]. It is related also to the concept of entanglement splitting suggested by Bruß [62] and has a direct relevance to 1→2 quantum telecloning [7,8]. It can further be viewed [62] as a channel bifurcation with one input side and two output sides.

The 3-TPI process (1) is capable of producing tripartite states of rank two (i.e., $j{=}1/2$) or of rank six (i.e., $j{=}1/2$ and 3/2) with 1→2 entanglement. It completely depends on the spins (S_0,S_1,S_2,S_f) of the electronic states $(|0\rangle,|1\rangle,|2\rangle,|3\rangle)$ of the species $(A,A^{+^*},A^{2+^*},A^{3+})$ participating in a given process (1). In addition, the 1→2 entanglement will be present whether in $[(e_1,e_2),(e_1,e_3)]$ or in $[(e_1,e_3),(e_2,e_3)]$, as well as concurrences of each pair, are also decided by the spins (S_0,S_1,S_2,S_f) only.

(a) 1→2 entanglement for (e_1,e_2) and (e_1,e_3). Process (1) generates tripartite states $\sigma(S;\omega)$ of (e_1,e_2,e_3) with this property for $S_0{=}S_2{=}S_1{-}1/2{=}S_f{+}1/2$. Here, the state $\sigma(S;\omega_2,\omega_3)$ of the two Auger electrons (e_2,e_3) is always separable and non-Werner with $p_{23}{=}{-}1/3$ [Eqs. (6) and (9c)]. But, both $\sigma(S;\omega_1,\omega_2)$ and $\sigma(S;\omega_1,\omega_3)$ are in the Werner state (6) with the mixing parameters p_{12} [Eq. (8b)], $=p_{13}$ [Eq. (10)], and $\equiv p_1$, say. Consequently, the concurrences of these two states are also equal and are given by the respective Eqs. (11a) and (11c), i.e., $\mathcal{C}_{12}{=}\mathcal{C}_{13}{=}(2S_1)^{-1}{\equiv}\mathcal{C}_1$, say. On account of the above-mentioned conditions to be satisfied by (S_0,S_1,S_2,S_f) in the present case, the minimum permissible value of the spin S_1 of A^{+^*} participating in the process (1) is now one. This gives, as shown in [44], $\mathcal{C}_{12}^2$

$+\mathcal{C}_{13}^2=(2S_1^2)^{-1}<1$. The entanglement molecule [56], in the present case, can obviously be either linear of the shape e_2-e_1-e_3 (e.g., CO_2) or in the form of H_2O with photoelectron e_1 at the position of the oxygen atom and one each of the Auger electrons (e_2,e_3) situated at the locations of two hydrogen atoms.

For example, for a rank-two tripartite state (3) generated on taking $(S_0,S_1,S_2,S_f)=(1/2,1,1/2,0)$ in the process (1), $p_1=2/3$ and $\mathcal{C}_1=1/2$. Similarly, with $(S_0,S_1,S_2,S_f)=(3/2,2,3/2,1)$, one generates a rank-six tripartite state of (e_1,e_2,e_3) with $p_1=1/2$ and $\mathcal{C}_1=1/4$. Thus, in the present case, the amount of the pure maximally entangled states $\sigma_1(\omega_1,\omega_2)$ and $\sigma_1(\omega_1,\omega_3)$, contributing to the respective Werner states $\sigma(S;\omega_1,\omega_2)$ and $\sigma(S;\omega_1,\omega_3)$, as well as the strength of their entanglement, decreases with an increase in the spin S_1 of the electronic state $|1\rangle$ of A^{+^*} participating in the process (1).

(b) $1\rightarrow 2$ entanglement for (e_1,e_3) and (e_2,e_3). Here, the second Auger electron e_3 forms entangled bipartite states with the photoelectron e_1 and with the first Auger electron e_2. The entanglement molecule [56] can, therefore, be either of the shape e_1-e_3-e_2 or in the form of an isosceles triangle whose two equal sides are e_3-e_1 and e_3-e_2, wherein e_1 and e_2 remain unconnected (i.e., not entangled). For it to happen in the 3-TPI process (1), one should have $S_1=S_f=S_0+1/2=S_2-1/2$. Then, each of the two pairs of entangled electrons (e_2,e_3) and (e_1,e_3) is in identical Werner state (6) with the mixing parameter p_{23} [Eq. (9b)] $=p_{13}$ [Eq. (10)] $\equiv p_3$, say. These states have concurrences $\mathcal{C}_{23}$ [Eq. (11b)] $=\mathcal{C}_{13}$ [Eq. (11c)] $\equiv\mathcal{C}_3$, say. On the other hand, product states (6) and (8c) of the first two emitted electrons is always non-Werner. A 3-TPI process (1) with $(S_0,S_1,S_2,S_f)=(0,1/2,1,1/2)$ will produce a rank-two $1\rightarrow 2$ entanglement state of this kind with $p_3=2/3$ and $\mathcal{C}_3=1/2$. In addition to this, some of the examples of rank-six tripartite states possessing this property are $(S_0,S_1,S_2,S_f)=(1/2,1,3/2,1)$ and (1, 3/2, 2, 3/2) with $(p_3,\mathcal{C}_3)=(5/9,1/3)$ and (1/2, 1/4).

The properties of the tripartite states of (e_1,e_2,e_3) discussed in Sec. IV B herein are summarized in the Table II.

C. Some possible experiments for producing various tripartite states of (e_1,e_2,e_3)

One can give numerous realistic examples, which can readily be implemented in a laboratory, for generating various kinds of tripartite states of flying electronic qubits using the 3-TPI process (1). In this subsection, we analyze two of those experiments [32–37] wherein CA decay, following inner-shell photoionization, has been observed in contexts totally different from those considered in this paper. The following two examples are for 3-TPI in a closed-shell atom and in an open-shell atom.

1. Xe 3d or 4d photoionization

Xe($3s^23p^63d^{10}4s^24p^64d^{10}5s^25p^6\ {}^1S$) is one of the most widely studied [32–35] atoms for CA decay. Electron spectra of Xe has been measured both for $3d$ [32,33] and $4d$ [33–35] photoionization. The process (1) involving [$3d^{10}\ {}^1S$, $3d^{-1}\ {}^2D$, $4d^{-1}5p^{-1}\ {}^1P$ (or $4d^{-1}5p^{-1}\ {}^1D$, or $4d^{-1}5p^{-1}\ {}^1F$), $5p^{-3}\ {}^2P$ (or $5p^{-3}\ {}^2D$)] states of [Xe, Xe^{+^*}, Xe^{2+^*}, and Xe^{3+}] belongs to Sec. IV B 3(b) with $\sigma^{T_{e_3}}\geq 0$. Herein, (e_1,e_2) are in the Bell state $\sigma_1(\omega_1,\omega_2)$ [Eq. (A18)] and each of the two remaining pairs is in the maximally chaotic state σ_0. On the other hand, transitions involving the states [$3d^{10}\ {}^1S$, $3d^{-1}\ {}^2D$, $4d^{-1}5p^{-1}\ {}^3P$ (or $4d^{-1}5p^{-1}\ {}^3D$, or $4d^{-1}5p^{-1}\ {}^3F$), $5p^{-3}\ {}^2P$ (or $5p^{-3}\ {}^2D$)] generate, according to Sec. IV B 4(b), the fully inseparable state of (e_1,e_2,e_3). Here, while (e_1,e_2) are in the non-Werner states (6) and (8c), each of the two remaining pairs (e_1,e_3) and (e_2,e_3) forms mixed, entangled Werner state (6) with $p_3=2/3$ [Eq. (9b)] and $\mathcal{C}_3=1/2$ [Eq. (10)]. This process, therefore, produces $1\rightarrow 2$ entanglement of the form e_1-e_3-e_2. The two entangled bipartite states $\sigma(S;\omega_1,\omega_3)$ and $\sigma(S;\omega_2,\omega_3)$ can be connected [40], if desired, to form a GHZ state. However, with [Xe, Xe^{+^*}, Xe^{2+^*}, and Xe^{3+}] participating in the 3-TPI process (1) with the states [$3d^{10}\ {}^1S$, $3d^{-1}\ {}^2D$, $4d^{-1}5p^{-1}\ {}^3P$ (or $4d^{-1}5p^{-1}\ {}^3D$, or $4d^{-1}5p^{-1}\ {}^3F$), $5p^{-3}\ {}^4S$] one will have (e_1,e_2,e_3) in a fully separable state of rank four discussed in the Sec. IV B 2.

2. Cs 4d photoionization

Osmekhin *et al.* [37] very recently observed CA decay in Cs $4d$ ionization. Unlike the inert gas atoms Xe, Kr, and Ar, whose electron spectra for the 3-TPI process (1) have earlier been experimentally observed [32–36], Cs atom has an unfilled outer most orbital in its electronic configuration. The states of [Cs, Cs^{+^*}, Cs^{2+^*}, and Cs^{3+}] involved in the 3-TPI process (1) are [37]

$$[4d^{10}5s^25p^66s^1\ {}^2S, 4d^{-1}5s^25p^66s^1\ {}^1D,\\ 4d^{10}5s^05p^66s^1\ {}^2S, 4d^{10}5s^{-1}5p^{-1}6s^0\ {}^1P].$$

It is an example of Sec. IV B 3(a) wherein both of the Auger electrons (e_2,e_3) are in the $\sigma_1(\omega_2,\omega_3)$ Bell state (A18), but each of the two remaining bipartite states $\sigma(S;\omega_1,\omega_2)$ and $\sigma(S;\omega_1,\omega_3)$ is maximally chaotic. The other possibility is with the states

$$[4d^{10}5s^25p^66s^1\ {}^2S, 4d^{-1}5s^25p^66s^1\ {}^1D,\\ 4d^{10}5s^05p^66s^1\ {}^2S, 4d^{10}5s^{-1}5p^{-1}6s^0\ {}^3P]$$

participating in Eq. (1). This will generate, according to Sec. IV B 2(a), a fully separable tripartite state in which (e_1,e_2) as well as (e_1,e_3) form white-noise state σ_0, but (e_2,e_3) are in the non-Werner state (6) with $p_{23}=-1/3$. Yet, one more possibility is with the transitions

$$[4d^{10}5s^25p^66s^1\ {}^2S, 4d^{-1}5s^25p^66s^1\ {}^3D,\\ 4d^{10}5s^05p^66s^1\ {}^2S, 4d^{10}5s^{-1}5p^{-1}6s^0\ {}^1P].$$

In this case, (e_1,e_2,e_3) form a fully inseparable state discussed in Sec. IV B 4(a) with $1\rightarrow 2$ entanglement of the photoelectron e_1 with each of the two Auger electrons e_2 and e_3 such that $p_1=2/3$ [Eq. (8b) or Eq. (10)] and $\mathcal{C}_1=1/2$ [Eq. (11a) or Eq. (11c)] for the mixing parameter and concurrence of the entangled bipartite states $\sigma(S;\omega_1,\omega_2)$ and $\sigma(S;\omega_1,\omega_3)$. The two Auger electrons (e_2,e_3) are now in a non-Werner state with $p_{23}=-1/3$. Another 3-TPI process,

TABLE II. Properties of the tripartite states (3) of electrons (e_1,e_2,e_3) generated in the 3-TPI process (1).

Partial transpose [Eq. (4)]	States of rank two (j [a] $=\frac{1}{2}$): $\lvert S_0-S_f\rvert=\frac{1}{2}$, $S_0+S_f=\frac{1}{2}$	States of rank four (j [a] $=\frac{3}{2}$): $\lvert S_0-S_f\rvert=\frac{3}{2}$, $S_0+S_f\geq\frac{3}{2}$	States of rank six (j [a] $=\frac{1}{2}$ and $\frac{3}{2}$): $\lvert S_0-S_f\rvert=\frac{1}{2}$, $S_0+S_f\geq\frac{3}{2}$
$\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}$, $\sigma^{T_{e_3}}>0$:[b]	None	All[c]	Those with[d] or with[e]
$\sigma^{T_{e_1}}>0$(with $\sigma^{T_{e_2}}$, $\sigma^{T_{e_3}}<0$): 1-qubit biseparable states [39,40]e_1-(e_2,e_3)	$S_1=S_f=S_0(=\frac{1}{2})-\frac{1}{2}=S_2-\frac{1}{2}$: (e_2,e_3) in the Bell state (A18); (e_1,e_2) and (e_1,e_3) in white noise σ_0	None	$S_1=S_f=S_0(>\frac{1}{2})-\frac{1}{2}=S_2-\frac{1}{2}$: (e_2,e_3) in the entangled, Werner state [(6) and (9b)] with concurrence (11b); (e_1,e_2) in [(6) and (8a)] and (e_1,e_3) in [(6) and (10)] separable bipartite states
$\sigma^{T_{e_3}}>0$ (with $\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}<0$): 1-qubit biseparable states [39,40]e_3-(e_1,e_2)	$S_1=S_f=S_0(=0)+\frac{1}{2}=S_2+\frac{1}{2}$: (e_1,e_2) in the Bell state (A18); (e_2,e_3) and (e_1,e_3) in white noise σ_0	None	$S_1=S_f=S_0(>0)+\frac{1}{2}=S_2+\frac{1}{2}$: (e_1,e_2) in the entangled, Werner state [(6) and (8b)] with concurrence (11a); (e_2,e_3) in [(6) and (9a)] and (e_1,e_3) in [(6) and (10)] separable bipartite states
$\sigma^{T_{e_2}}>0$:[f]	None	None	None
$\sigma^{T_{e_1}}$, $\sigma^{T_{e_2}}$, $\sigma^{T_{e_3}}<0$: Fully inseparable states [39,40]	$1\rightarrow2$ entanglement: (a)[g] $S_0(=\frac{1}{2})=S_2=S_1-\frac{1}{2}=S_f+\frac{1}{2}$; (b)[h] $S_1(=\frac{1}{2})=S_f=S_0+\frac{1}{2}=S_2-\frac{1}{2}$	None	$1\rightarrow2$ entanglement: (a)[g] $S_0(>\frac{1}{2})=S_2=S_1-\frac{1}{2}=S_f+\frac{1}{2}$; (b)[h] $S_1(>\frac{1}{2})=S_f=S_0+\frac{1}{2}=S_2-\frac{1}{2}$

[a] Total spin angular momentum $j=\lvert\vec{(\frac{1}{2})}_1+\vec{(\frac{1}{2})}_2+\vec{(\frac{1}{2})}_3\rvert$ of electrons (e_1,e_2,e_3) ejected in 3-TPI process (1).

[b] Fully separable states [39,40].

[c] (e_1,e_2), (e_2,e_3), (e_1,e_3) in the product, non-Werner, state (6) with $p_{12},p_{23},p_{13}=-\frac{1}{3}$;.

[d] $S_0=S_2=S_1+\frac{1}{2}=S_f-\frac{1}{2}$: (e_2,e_3) in the product, non-Werner states [(6) and (9c)]; but, each of (e_1,e_2) and (e_1,e_3) is in the same separable, Werner states [(6) and (8a)] or [(6) and (10)].

[e] $S_1=S_f=S_0-\frac{1}{2}=S_2+\frac{1}{2}$: now the separable state of (e_1,e_2) is [(6) and (8c)], i.e., non-Werner; whereas, both (e_2,e_3) and (e_1,e_3) belong to the same separable Werner states [(6) and (9a)] or [(6) and (10)].

[f] One-qubit biseparable state e_2-(e_1,e_3). The 3-TPI process (1) is incapable of generating such states.

[g] (e_1,e_2) and (e_1,e_3) in identical, entangled Werner states [(6) and (8b)] or [(6) and (10)] with concurrence (11a) or (11c); (e_2,e_3) in product, non-Werner states [(6) and (9c)].

[h] (e_2,e_3) and (e_1,e_3) in identical, entangled Werner states [(6) and (9b)] or [(6) and (10)] with concurrence (11b) or (11c); (e_1,e_2) in product, non-Werner states [(6) and (8c)].

possible in the present target, is [Cs, Cs^{+^*}, Cs^{2+^*}, and Cs^{3+}] participating in Eq. (1) with the states

$$[4d^{10}5s^25p^66s^1\ {}^2S,4d^{-1}5s^25p^66s^1\ {}^3D,$$
$$4d^{10}5s^05p^66s^1\ {}^2S,4d^{10}5s^{-1}5p^{-1}6s^0\ {}^3P].$$

According to Sec. IV B 3(b), it is a one-qubit biseparable state of the form e_3-(e_1,e_2). Accordingly, the entangled state of (e_1,e_2) is Eqs. (6) and (8b); whereas, (e_2,e_3) are in the separable Werner states (6) and (9a), but electrons (e_1,e_3) are in the non-Werner states (6) and (10). One may also have [Cs, Cs^{+^*}, Cs^{2+^*}, and Cs^{3+}] participating in Eq. (1) with the states

$$[4d^{10}5s^25p^66s^1\ {}^2S,4d^{-1}5s^25p^66s^1\ {}^3D,$$
$$4d^{10}5s^05p^66s^1\ {}^4S,4d^{10}5s^{-1}5p^{-1}6s^0\ {}^3P].$$

Here again, (e_1,e_2,e_3) are in a fully inseparable state discussed in Sec. IV B 4(b). In the present case, however, the second Auger electron forms entangled bipartite states $\sigma(S;\omega_1,\omega_3)$ and $\sigma(S;\omega_2,\omega_3)$ with $p_3=5/9$ and $\mathcal{C}_3=1/3$. The bipartite, product state of photoelectron e_1, and first Auger electron e_2 is non-Werner corresponding to $p_{12}=-1/3$. Finally, the transitions with the states

$$[4d^{10}5s^2 5p^6 6s^1\ {}^2S, 4d^{-1}5s^2 5p^6 6s^1\ {}^3D,$$
$$4d^{10}5s^0 5p^6 6s^1\ {}^4S, 4d^{10}5s^{-1}5p^{-1}6s^0\ {}^5S]$$

generate (e_1,e_2,e_3) in a fully separable mixed state of rank four discussed in Sec. IV B 2 of this paper.

V. CONCLUSIONS

This paper presents the study of entanglement generation and characterization of three flying electronic qubits (e_1,e_2,e_3) from a single atomic source A. These electrons are sequentially produced in the inner-shell photoionization and in the CA decay of the consequent vacancy. The entanglement properties are generated purely due to the Coulomb interaction experienced by (e_1,e_2,e_3) inside the atom and, hence, have no coupling at all with the environment external to it. Thus, the generated entanglement is least prone to the harmful effects of dissipation and decoherence while (e_1,e_2,e_3) fly away from the residual triply charged positive ion A^{3+}. A simple expression for the (8×8) DM has been derived for a tripartite state of (e_1,e_2,e_3). This DM is shown to be completely determined merely from the spins (S_0,S_1,S_2,S_f) of the electronic states $(|0\rangle,|1\rangle,|2\rangle,|f\rangle)$ of the species $(A,A^{+^*},A^{2+^*},A^{3+})$, as well as of the electrons (e_1,e_2,e_3) participating in this three-step process. Although, DM involves also the directions of quantization of the spins of (e_1,e_2,e_3), the entanglement properties of the generated tripartite states of electronic qubits are totally independent of these directions.

Here, one is able to generate—depending upon the spins (S_0,S_1,S_2,S_f)—fully separable, 1-qubit biseparable, fully inseparable, and $1\rightarrow 2$ entangled tripartite states of (e_1,e_2,e_3). The last of these can be connected to produce a GHZ state. Extremely simple analytical expressions have been given in terms of the spins (S_0,S_1,S_2,S_f) to characterize the properties of these tripartite states as well as of the bipartite state which can be formed when any party holding one of the three electrons (e_1,e_2,e_3) refuses to cooperate with the other two. A bipartite state so formed can be any thing from a maximally chaotic to a Bell state, including mixed entangled Werner states, or mixed separable Werner/non-Werner states. The properties (e.g., mixing parameter, concurrence, etc.) of these bipartite states too are completely determined from the spins (S_0,S_1,S_2,S_f) using the expressions given herein. We have suggested also several realistic three-step processes in Xe and Cs atoms which can be the basis of a potential experiment for generating in a laboratory these tripartite states of (e_1,e_2,e_3). Such entangled states can be used for implementing quantum communication protocols (e.g., $1\rightarrow 2$ quantum telecloning, etc.) and for quantum computation with particles possessing rest mass different from zero.

The present paper, thus, provides an entanglement factory for generating various kinds of tripartite (as well as bipartite) states of flying electronic qubits. Experimental characterization of these states does not require use of any protocols [1–3] hitherto developed in quantum information. Merely measurements of the energies of these qubits using three different electron spectrometers is sufficient to determine the electronic states $(|0\rangle,|1\rangle,|2\rangle,|f\rangle)$ and, hence, spin multiplicities, in the L-S coupling of $(A,A^{+^*},A^{+^*},A^{3+})$ participating in a given 3-TPI process (1). With this knowledge of the spins (S_0,S_1,S_2,S_f), the expressions derived herein are sufficient to completely specify all the relevant entanglement properties of a spin state of (e_1,e_2,e_3).

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APPENDIX: DENSITY MATRIX FOR THE TRIPARTITE STATE OF A PHOTOELECTRON AND TWO AUGER ELECTRONS EMITTED SEQUENTIALLY FROM AN ATOM IN THE ABSENCE OF SPIN-ORBIT INTERACTION

This appendix contains a derivation of the DM explained in Sec. III and used in the discussion in Sec. IV of this paper. In order to analyze the nonlocal spin-correlation (i.e., spin entanglement) of the tripartite state formed by the photoelectron e_1 and two Auger electrons (e_2,e_3), it is necessary that our DM for the three-step process (1) be both angle and spin resolved. In the following derivation, we take SOI into account neither in the bound electronic states $(|0\rangle,|1\rangle,|2\rangle,|f\rangle)$ of $(A,A^{+^*},A^{2+^*},A^{3+})$ nor in the continuum of any of the ejected electrons (e_1,e_2,e_3). Only the electrostatic Coulomb forces for particles participating in 3-TPI process (1) are thus included. In addition, neither the photoionization operator F_{p_1} in the $E1$ approximation nor any of the Auger emission operators (F_{a_2},F_{a_3}) depend upon any spin variables. In such situations, L-S coupling [46] becomes applicable. Consequently, orbital and spin angular momenta are individually conserved in each of the three steps (1a) and (1c) of the process (1). We, therefore, have

$$\vec{\ell}_r + \vec{L}_0 = \vec{L}_1 + \vec{\ell}_1, \quad \vec{S}_0 = \vec{S}_1 + \vec{s}_1\left[=\left(\frac{1}{2}\right)_1\right], \tag{A1}$$

$$\vec{L}_1 = \vec{L}_2 + \vec{\ell}_2, \quad \vec{S}_1 = \vec{S}_2 + \vec{s}_2\left[=\left(\frac{1}{2}\right)_2\right], \tag{A2}$$

$$\vec{L}_2 = \vec{L}_f + \vec{\ell}_3, \quad \vec{S}_2 = \vec{S}_f + \vec{s}_3\left[=\left(\frac{1}{2}\right)_3\right]. \tag{A3}$$

Here, $(\vec{L}_0,\vec{L}_1,\vec{L}_2,\vec{L}_f)$ and $(S_0\equiv|\vec{S}_0|,\ S_1\equiv|\vec{S}_1|,\ S_2\equiv|\vec{S}_2|,\ S_f\equiv|\vec{S}_f|)$ are the respective orbital and spin angular momenta of $(A,A^{+^*},A^{2+^*},A^{3+})$; whereas, $(\vec{\ell}_1,\vec{\ell}_2,\vec{\ell}_3)$ and $(s_1\equiv|\vec{s}_1|,\ s_2\equiv|\vec{s}_2|,\ s_3\equiv|\vec{s}_3|)$ are those of the electrons (e_1,e_2,e_3) participating in the 3-TPI process (1). Further, we use the symbols M_{L_0}, M_{L_1}, M_{L_2}, M_{L_f}, M_{S_0}, M_{S_1}, M_{S_2}, M_{S_f} for representing, along our quantization (i.e., OZ-) axis (shown in Fig. 1), projections of the corresponding angular momenta defined in Eqs. (A1)–(A3). One can now readily write [46]

$$|0\rangle = |\vec{L}_0\vec{S}_0 M_{L_0} M_{S_0}\rangle, \tag{A4a}$$

$$|1\rangle = |\vec{L}_1\vec{S}_1 M_{L_1} M_{S_1}\rangle, \tag{A4b}$$

$$|2\rangle = |\vec{L}_2\vec{S}_2 M_{L_2} M_{S_2}\rangle, \tag{A4c}$$

$$|f\rangle = |\vec{L}_f\vec{S}_f M_{L_f} M_{S_f}\rangle, \tag{A4d}$$

for the electronic stats of A, A^{+^*}, A^{2+^*}, and A^{3+}, respectively, in the L-S coupling.

Further, in Eq. (1) and elsewhere in this paper, $(|\mu_1\hat{u}_1\vec{k}_1\rangle^-, |\mu_2\hat{u}_2\vec{k}_2\rangle^-, |\mu_3\hat{u}_3\vec{k}_3\rangle^-)$ are the continuum spin orbitals of the ejected electrons (e_1, e_2, e_3). Here, a minus superscript means that these orbitals individually satisfy asymptotic incoming wave boundary conditions [63] appropriate for photoionization (1a) as well as for Auger emissions (1b) and (1c).

In order to calculate the desired DM, we first need to know the DO ρ_i present in Eq. (2). It, for a noninteracting system of an unpolarized atom (in the L-S coupling) plus a polarized photon, is given by (see, for example, [64])

$$\rho_i = \rho_0 \otimes \rho_r = \frac{1}{(2L_0+1)(2S_0+1)} \sum_{M_{L_0} M_{S_0}} |0;1m_r\rangle\langle 0;1m_r|, \tag{A5}$$

where we have defined $|0;1m_r\rangle \equiv |0\rangle|1m_r\rangle$. An element (which is diagonal in $\hat{u}_i$ and $\vec{k}_i$ but nondiagonal in μ_i, with $i=1$ to 3) of the DO defined in Eq. (2b) is

$$\begin{aligned}\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle \\ = \mathcal{K} \sum_{M_{L_f} M_{S_f}} \langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|(F_{a_3}F_{a_2}F_{p_1})\rho_i(F_{a_3}F_{a_2}F_{p_1})^\dagger|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle.\end{aligned} \tag{A6}$$

In this expression, we have summed over all the degenerate Zeeman components [46] of the electronic state $|f\rangle$ of the residual triply charged positive stable ion A^{3+} formed in the final step (1c) after the sequential emission of (e_1, e_2, e_3) in the process (1). On substituting Eq. (A5) and using the completeness of the electronic state $|1\rangle$ of A^{+^*} and $|2\rangle$ of A^{2+^*}, the above DM becomes

$$\begin{aligned}&\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle \\ &= \frac{\mathcal{K}}{(2L_0+1)(2S_0+1)} \sum_{\substack{M_{L_0}M_{L_1}M_{L_1}'M_{L_2}M_{L_2}'M_{L_f} \\ M_{S_0}M_{S_1}M_{S_1}'M_{S_2}M_{S_2}'M_{S_f}}} \langle f;\mu_3,\hat{u}_3,\vec{k}_3|F_{a_2}|2\rangle\langle 2;\mu_2,\hat{u}_2,\vec{k}_2|F_{a_1}|1\rangle\langle 1;\mu_1,\hat{u}_1,\vec{k}_1|F_{p_1}|0;1m_r\rangle \\ &\quad\times\langle 0;1m_r|F_{p_1}^\dagger|1';\mu_1',\hat{u}_1,\vec{k}_1\rangle\langle 1'|F_{a_1}^\dagger|2';\mu_2',\hat{u}_2,\vec{k}_2\rangle\langle 2'|F_{a_2}^\dagger|f;\mu_3',\hat{u}_3,\vec{k}_3\rangle.\end{aligned} \tag{A7}$$

Here, we have abbreviated $|1'\rangle \equiv |\vec{L}_1\vec{S}_1 M_{L_1}' M_{S_1}'\rangle$ and $|2'\rangle \equiv |\vec{L}_2\vec{S}_2 M_{L_2}' M_{S_2}'\rangle$ for the states of the excited photoion A^{+^*} and excited dication A^{2+^*}, respectively. These are degenerate to the states defined in the respective Eqs. (A4b) and (A4c). Further in Eq. (A7), $|1;\mu_1,\hat{u}_1,\vec{k}_1\rangle = \mathcal{A}(|1\rangle|\mu_1,\hat{u}_1,\vec{k}_1\rangle^-)$ is an antisymmetrized state of the excited photoion A^{+^*} and of the photoelectron e_1, and so on.

Expression (A7) represents an eight-dimensional matrix for a system of three qubits which contains, among other things, complete information on photoelectron e_1 and both Auger electrons (e_2, e_3) ejected sequentially in the 3-TPI process (1) taking place in the L-S coupling in the absence of SOI. The nondiagonal $(\mu_i' \neq \mu_i)$ elements in Eq. (A7) represent the coherent effects; whereas, diagonal $(\mu_i' = \mu_i)$ elements describe angular distribution of spin resolved (e_1, e_2, e_3). It is obvious that the DM (A7) is Hermitian, i.e.,

$$\begin{aligned}&\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle \\ &= \langle f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3|\rho_f|f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_3;\mu_3,\hat{u}_3,\vec{k}_3\rangle^*.\end{aligned} \tag{A8}$$

In order to proceed further, one needs to evaluate the matrix elements of the $E1$ photoionization operator F_{p_1} and of both of the Auger emission operators F_{a_2} and F_{a_3} present on the right-hand side of Eq. (A7). The procedures to be used for evaluating the matrix elements of F_{p_1} and of F_{a_2} have already been explained in detail in the appendix of [29(b)]. The final results given in Eqs. (A9) and (A11) therein [29(b)] can directly be used in the present case as well for the matrix elements of these two operators with, of course, a proper change in notations. A procedure, identical to that used for calculating a matrix element of F_{a_2} in Ref. [29], needs to be adopted for the second Auger operator F_{a_3} as well. These three matrix elements and their respective Hermitian conjugates are then substituted in the DM (A7). The consequent expression is simplified with a heavy use of Racah algebra [48]. Finally, the DM for the 3-TPI process (1) can be shown to reduce to the following form:

$$\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2;\mu_3,\hat{u}_3,\vec{k}_3|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2;\mu_3',\hat{u}_3,\vec{k}_3\rangle = \frac{d^4\sigma(m_r)}{d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3}\sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}. \tag{A9}$$

Here,

$$\begin{aligned}\frac{d^4\sigma(m_r)}{d\epsilon_1 d\hat{k}_1 d\hat{k}_2 d\hat{k}_3} ={}& (-1)^{L_0+L_2+L_f+m_r}\frac{(2L_1+1)(2L_2+1)\mathcal{K}}{(4\pi)^{3/2}(2L_0+1)}\sum_{\substack{\ell_1\ell_1'C_1\gamma_1CL_r\\ \ell_2\ell_2'C_2\gamma_2\mathbb{L}\\ \ell_3\ell_3'C_3\gamma_3\mathbb{L}'}}(-1)^{\ell_1+\ell_2+C+C_3+\mathbb{L}'+\gamma_1}\\ &\times(2C+1)(2L_r+1)\sqrt{(2C_1+1)(2C_2+1)(2C_3+1)}\begin{pmatrix}1 & 1 & L_r\\ m_r & -m_r & 0\end{pmatrix}\begin{pmatrix}\ell_1 & \ell_1' & C_1\\ 0 & 0 & 0\end{pmatrix}\\ &\times\begin{pmatrix}\ell_2 & \ell_2' & C_2\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}\ell_3 & \ell_3' & C_3\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}C_2 & C_3 & C\\ \gamma_2 & \gamma_3 & \gamma_1\end{pmatrix}\begin{pmatrix}C_1 & C & L_r\\ \gamma_1 & -\gamma_1 & 0\end{pmatrix}\begin{Bmatrix}1 & 1 & L_r\\ \mathbb{L} & \mathbb{L}' & L_0\end{Bmatrix}\begin{Bmatrix}\ell_3 & \ell_3' & C_3\\ L_2 & L_2 & L_f\end{Bmatrix}\\ &\times\begin{Bmatrix}\ell_1 & \ell_1' & C_1\\ L_1 & L_1 & C\\ \mathbb{L} & \mathbb{L}' & L_r\end{Bmatrix}\begin{Bmatrix}\ell_2 & \ell_2' & C_2\\ L_2 & L_2 & C_3\\ L_1 & L_1 & C\end{Bmatrix}Pe_1(L_1\ell_1;L_01;\mathbb{L})[Pe_1(L_1\ell_1';L_01;\mathbb{L}')]^*\\ &\times Ae_2(L_2\ell_2;L_1)[Ae_2(L_2\ell_2';L_1)]^*Ae_3(L_f\ell_3;L_2)[Ae_3(L_f\ell_3';L_2)]^*[Y_{C_1}^{\gamma_1}(\hat{k}_1)]^*[Y_{C_2}^{\gamma_2}(\hat{k}_2)]^*[Y_{C_3}^{\gamma_3}(\hat{k}_3)]^*,\end{aligned} \tag{A10}$$

and

$$\begin{aligned}\sigma(S_0;S_1;S_2;S_f;\hat{u}_1,\hat{u}_2,\hat{u}_3)_{\mu_1,\mu_2,\mu_3;\mu_1',\mu_2',\mu_3'} ={}& (-1)^{S_0-S_1-S_2+S_f+\mu_1'+\mu_2'+\mu_3'-1/2}(2S_1+1)(2S_2+1)\sum_{\substack{P_1P_2P_3p_1p_2p_3\\ p_1'p_2'p_3'}}(-1)^{P_1+P_3}(2P_1+1)(2P_2+1)\\ &\times(2P_3+1)\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & P_1\\ \mu_1 & -\mu_1' & p_1\end{pmatrix}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & P_2\\ \mu_2 & -\mu_2' & p_2\end{pmatrix}\begin{pmatrix}\frac{1}{2} & \frac{1}{2} & P_3\\ \mu_3 & -\mu_3' & p_3\end{pmatrix}\begin{pmatrix}P_1 & P_2 & P_3\\ p_1' & p_2' & p_3'\end{pmatrix}\\ &\times\begin{Bmatrix}\frac{1}{2} & \frac{1}{2} & P_1\\ S_1 & S_1 & S_0\end{Bmatrix}\begin{Bmatrix}\frac{1}{2} & \frac{1}{2} & P_3\\ S_2 & S_2 & S_f\end{Bmatrix}\begin{Bmatrix}S_1 & S_1 & P_1\\ \frac{1}{2} & \frac{1}{2} & P_2\\ S_2 & S_2 & P_3\end{Bmatrix}[\mathcal{D}^{P_1}_{p_1p_1'}(\omega_1)]^*[\mathcal{D}^{P_2}_{p_2p_2'}(\omega_2)]^*[\mathcal{D}^{P_3}_{p_3p_3'}(\omega_3)]^*.\end{aligned} \tag{A11}$$

Further, in the expression (A10)

$$Pe_1(L_1\ell_1;L_01;\mathbb{L}) = (-i)^{\ell_1}e^{i\sigma_{\ell_1}}(-1)^{\ell_1}(2\mathbb{L}+1)\sqrt{2\ell_1+1}\langle(L_1\ell_1)\mathbb{L}|F_{p_1}|(L_01)\mathbb{L}\rangle \tag{A12a}$$

is the photoionization matrix element in the $E1$ approximation for the first step (1a) in the 3-TPI process (1); whereas,

$$Ae_2(L_2\ell_2;L_1) = (-i)^{\ell_2}e^{i\sigma_{\ell_2}}(-1)^{\ell_2}\sqrt{2\ell_2+1}\langle(L_2\ell_2)L_1|F_{a_2}|L_1\rangle \tag{A12b}$$

and

$$Ae_3(L_3\ell_3;L_2) = (-i)^{\ell_3}e^{i\sigma_{\ell_3}}(-1)^{\ell_3}\sqrt{2\ell_3+1}\langle(L_f\ell_3)L_2|F_{a_3}|L_2\rangle \tag{A12c}$$

are the respective amplitudes for the emission of Auger electrons e_2 and of e_3 in the second step (1b) and third step (1c) of Eq. (1).

A very good check on the accuracy of the procedure used in this appendix is that when the second Auger electron (i.e., e_3) remains unobserved (i.e., on integrating over $\hat{k}_3$ and summing over μ_3), relations (A9)–(A11) herein reduce to those obtained earlier in Eqs. (A14) and (A15) in Ref. [29(b)] for studying spin entanglement between a photo-Auger-electron pair in the absence of SOI in 2-DPI.

The following expressions provide a simple method for calculating Eq. (A11) for any values of the spins (S_0,S_1,S_2,S_f):

$$\sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_2,\omega_3)_{\mu_1\mu_2\mu_3;\mu_1'\mu_2'\mu_3'}$$

$$= \Delta\left(S_0S_1\frac{1}{2}\right)\Delta\left(S_1S_2\frac{1}{2}\right)\Delta\left(S_2S_f\frac{1}{2}\right)\left[\frac{1}{8}I_8 + (-1)^{S_0-S_2}(2S_1+1)\frac{3}{2}\begin{Bmatrix} S_0 & \frac{1}{2} & S_1 \\ 1 & S_1 & \frac{1}{2} \end{Bmatrix}\begin{Bmatrix} S_2 & \frac{1}{2} & S_1 \\ 1 & S_1 & \frac{1}{2} \end{Bmatrix}\varsigma(\omega_1,\omega_2)\right.$$

$$+ (-1)^{S_1-S_f}(2S_2+1)\frac{3}{2}\begin{Bmatrix} S_1 & \frac{1}{2} & S_2 \\ 1 & S_2 & \frac{1}{2} \end{Bmatrix}\begin{Bmatrix} S_f & \frac{1}{2} & S_2 \\ 1 & S_2 & \frac{1}{2} \end{Bmatrix}\varsigma(\omega_2,\omega_3)$$

$$\left. + (-1)^{S_0+S_f-1/2}(2S_1+1)(2S_2+1)\frac{3}{2}\begin{Bmatrix} S_0 & \frac{1}{2} & S_1 \\ 1 & S_1 & \frac{1}{2} \end{Bmatrix}\begin{Bmatrix} S_f & \frac{1}{2} & S_2 \\ 1 & S_2 & \frac{1}{2} \end{Bmatrix}\begin{Bmatrix} \frac{1}{2} & S_1 & S_2 \\ 1 & S_2 & S_1 \end{Bmatrix}\varsigma(\omega_1,\omega_3)\right] \tag{A13a}$$

$$=\Delta\left(S_0S_1\frac{1}{2}\right)\Delta\left(S_1S_2\frac{1}{2}\right)\Delta\left(S_2S_f\frac{1}{2}\right)\left[\frac{1}{8}I_8 + \frac{1}{4}\mathbb{P}_{13}\varsigma(\omega_1,\omega_3) + \left[\frac{3}{4} + S_1(S_1+1) - S_0(S_0+1)\right]\left[\frac{3}{4} + S_1(S_1+1) - S_2(S_2+1)\right]\right.$$
$$\left. \times[4S_1(S_1+1)]^{-1}\varsigma(\omega_1,\omega_2) + \left[\frac{3}{4} + S_2(S_2+1) - S_1(S_1+1)\right]\left[\frac{3}{4} + S_2(S_2+1) - S_f(S_f+1)\right][4S_2(S_2+1)]^{-1}\varsigma(\omega_2,\omega_3),\right], \tag{A13b}$$

with

$$\mathbb{P}_{13} = \left[\frac{3}{4} + S_1(S_1+1) - S_0(S_0+1)\right]\left[\frac{3}{4} + S_2(S_2+1) - S_f(S_f+1)\right]\left[S_1(S_1+1) + S_2(S_2+1) - \frac{3}{4}\right][2S_1S_2(S_1+1)(S_2+1)]^{-1} \tag{A14}$$

and $\Delta(S_0S_1\frac{1}{2})$, etc., express the triangular condition [48] to be satisfied by the three angular momenta $(S_0,S_1,\frac{1}{2})$, etc. Namely [48],

$$\Delta\left(S_0S_1\frac{1}{2}\right) = \begin{cases} 1 & \text{if } |S_0 - S_1| \le \frac{1}{2} \le S_0 + S_1 \\ 0 & \text{otherwise.} \end{cases} \tag{A15}$$

Further in Eq. (A13), each of I_8, $\varsigma(\omega_1,\omega_2)$, $\varsigma(\omega_2,\omega_3)$, $\varsigma(\omega_1,\omega_3)$ is a (8×8) matrix. While I_8 is a unit matrix, but

$$\varsigma(\omega_1,\omega_2)_{\mu_1,\mu_2,\mu_3;\mu_1',\mu_2',\mu_3'} = \delta_{\mu_3\mu_3'}(-1)^{\mu_1'+\mu_2'}\sum_{nm_1m_2}(-1)^n\begin{pmatrix} \frac{1}{2} & \frac{1}{2} & 1 \\ \mu_1 & -\mu_1' & m_1 \end{pmatrix}\begin{pmatrix} \frac{1}{2} & \frac{1}{2} & 1 \\ \mu_2 & -\mu_2' & m_2 \end{pmatrix}[\mathcal{D}^1_{m_1n}(\omega_1)]^*[\mathcal{D}^1_{m_2,-n}(\omega_2)]^*, \tag{A16}$$

with similar expressions for the remaining $\varsigma(\omega_2,\omega_3)$ and $\varsigma(\omega_1,\omega_3)$ present in both Eqs. (A13a) and (A13b), is a nondiagonal matrix.

Equation (A13) can be used to calculate also the PT (4a)–(4c) of the DM $\sigma(S;\omega)_{\mu;\mu'}$ with respect to any of the three electrons (e_1,e_2,e_3) emitted in the 3-TPI process (1).

The analysis of the entanglement properties of a tripartite state of (e_1,e_2,e_3) occasionally requires a knowledge also of the reduced DMs which can be formed from $\sigma(S;\omega)_{\mu;\mu'}$. Each of the three reduced DMs, which are possible in the present case, are defined in Eqs. (5a)–(5c) herein. On substituting in Eq. (5a) any of the Eqs. (A11) and (A13a), or Eq. (A13b), the reduced DM for the photoelectron e_1 and the first Auger electron e_2 are obtained to be

$$\begin{aligned}\sigma(S;\omega_1,\omega_2)_{\mu_1\mu_2;\mu_1'\mu_2'} &\equiv \sigma(S_0,S_1,S_2;\omega_1,\omega_2)_{\mu_1\mu_2;\mu_1'\mu_1'}\\ &= (-1)^{S_0+S_2-2S_1+\mu_1'+\mu_2'}(2S_1+1)\sum_{Ppp_1p_2}(-1)^{P+p}(2P+1)\begin{pmatrix}1/2 & 1/2 & P\\ \mu_1 & -\mu_1' & p_1\end{pmatrix}\begin{pmatrix}1/2 & 1/2 & P\\ \mu_2 & -\mu_2' & p_2\end{pmatrix}\begin{Bmatrix}1/2 & 1/2 & P\\ S_1 & S_1 & S_0\end{Bmatrix}\\ &\quad\times\begin{Bmatrix}1/2 & 1/2 & P\\ S_1 & S_1 & S_2\end{Bmatrix}[\mathcal{D}^P_{p_1,p}(\omega_1)]^*[\mathcal{D}^P_{p_2,-p}(\omega_2)]^*.\end{aligned}\tag{A17a}$$

With a proper identification of the present spin quantum numbers (S_0,S_1,S_2) with those (S_0,S_e,S_f) used in Ref. [29] for studying entanglement in 2-DPI, the above expression (A17a) becomes—of course—identical to that derived earlier in Eq. (A15) [29(b)].

Substitutions of any of the forms (A11) and (A13a) or Eq. (A13b) of the spin-correlation DM in Eqs. (5b) and (5c) give us the reduced DMs

$$\begin{aligned}\sigma(S;\omega_2,\omega_3)_{\mu_2\mu_3;\mu_2'\mu_3'} &\equiv \sigma(S_1,S_2,S_f;\omega_2,\omega_3)_{\mu_2\mu_3;\mu_2'\mu_3'}\\ &= (-1)^{S_1+2S_2+S_f+\mu_2'+\mu_3'}(2S_2+1)\sum_{Ppp_2p_3}(-1)^{P+p}(2P+1)\begin{pmatrix}1/2 & 1/2 & P\\ \mu_2 & -\mu_2' & p_2\end{pmatrix}\begin{pmatrix}1/2 & 1/2 & P\\ \mu_3 & -\mu_3' & p_3\end{pmatrix}\begin{Bmatrix}1/2 & 1/2 & P\\ S_2 & S_2 & S_f\end{Bmatrix}\\ &\quad\times\begin{Bmatrix}1/2 & 1/2 & P\\ S_2 & S_2 & S_1\end{Bmatrix}[\mathcal{D}^P_{p_2,p}(\omega_2)]^*[\mathcal{D}^P_{p_3,-p}(\omega_3)]^*\end{aligned}\tag{A17b}$$

and

$$\begin{aligned}\sigma(S;\omega_1,\omega_3)_{\mu_1\mu_3;\mu_1'\mu_3'} &\equiv \sigma(S_0,S_1,S_2,S_f;\omega_1,\omega_3)_{\mu_1\mu_3;\mu_1'\mu_3'}\\ &= (-1)^{S_0+S_f-1/2+\mu_1'+\mu_3'}(2S_1+1)(2S_2+1)\sum_{Ppp_1p_3}(-1)^{p}(2P+1)\begin{pmatrix}1/2 & 1/2 & P\\ \mu_1 & -\mu_1' & p_1\end{pmatrix}\begin{pmatrix}1/2 & 1/2 & P\\ \mu_3 & -\mu_3' & p_3\end{pmatrix}\\ &\quad\times\begin{Bmatrix}1/2 & 1/2 & P\\ S_1 & S_1 & S_0\end{Bmatrix}\begin{Bmatrix}1/2 & 1/2 & P\\ S_2 & S_2 & S_f\end{Bmatrix}\begin{Bmatrix}S_1 & S_1 & P\\ S_2 & S_2 & \frac{1}{2}\end{Bmatrix}[\mathcal{D}^P_{p_1,p}(\omega_1)]^*[\mathcal{D}^P_{p_3,-p}(\omega_3)]^*,\end{aligned}\tag{A17c}$$

respectively. Here, the DM (A17b) describes the bipartite state of electrons (e_2,e_3) and Eq. (A17c) that of (e_1,e_3).

Each of the reduced DMs (A17a)–(A17c) can be expressed in the form of the Werner state (6) with $(i,j)=(1,2)$, (2, 3), and (1, 3), respectively. Equations (8a)–(8c), (9a)–(9c), and (10) give the mixing parameters p_{12}, p_{23}, and p_{13} for the bipartite state (A17a) of (e_1,e_2), Eq. (A17b) of (e_2,e_3), and Eq. (A17c) of (e_1,e_3), respectively.

We finally give, for completeness and for the convenience of the reader of this paper, the explicit form of the pure maximally entangled bipartite state $\sigma_1(\omega_i,\omega_j)$ of electrons (e_i,e_j) present in the expression (6) herein. It is one of the Bell states of two qubits. In the present case, it is a singlet state of two spin-$\frac{1}{2}$ particles. The following expressions are adapted from Eqs. (9) and (10) in Ref. [29(b)] wherein they were originally derived:

$$4\sigma_1(\omega_i,\omega_j)=\begin{array}{ccccc}
\mu_i,\mu_j/\mu_i',\mu_j' \rightarrow & \frac{1}{2},\frac{1}{2} & \frac{1}{2},-\frac{1}{2} & -\frac{1}{2},\frac{1}{2} & -\frac{1}{2},-\frac{1}{2} \\
\Downarrow & & & & \\
\frac{1}{2},\frac{1}{2} & 1-\hat{u}_i\cdot\hat{u}_j & \begin{array}{c} c_is_j-s_ic_jc \\ -is_is \end{array} & \begin{array}{c} s_ic_j-c_is_jc \\ +is_js \end{array} & \begin{array}{c} (1-c_ic_j)c-s_is_j \\ -i(c_i-c_j)s \end{array} \\
\frac{1}{2},-\frac{1}{2} & \begin{array}{c} c_is_j-s_ic_jc \\ +is_is \end{array} & 1+\hat{u}_i\cdot\hat{u}_j & \begin{array}{c} -(1+c_ic_j)c-s_is_j \\ +i(c_i+c_j)s \end{array} & \begin{array}{c} -s_ic_j+c_is_jc \\ -is_js \end{array} \\
-\frac{1}{2},\frac{1}{2} & \begin{array}{c} s_ic_j-c_is_jc \\ -is_js \end{array} & \begin{array}{c} -(1+c_ic_j)c-s_is_j \\ -i(c_i+c_j)s \end{array} & 1+\widehat{u_i}\cdot\widehat{u_j} & \begin{array}{c} -c_is_j+s_ic_jc \\ +is_is \end{array} \\
-\frac{1}{2},-\frac{1}{2} & \begin{array}{c} (1-c_ic_j)c-s_is_j \\ +i(c_i-c_j)s \end{array} & \begin{array}{c} -s_ic_j+c_is_jc \\ +is_js \end{array} & \begin{array}{c} -c_is_j+s_ic_jc \\ -is_is \end{array} & 1-\hat{u}_i\cdot\hat{u}_j
\end{array} \tag{A18a}$$

with the definitions

$$\begin{gathered} s\equiv\sin(\varphi_j-\varphi_i), \quad c\equiv\cos(\varphi_j-\varphi_i), \\ i\equiv\sqrt{(-1)}, \quad \hat{u}_i\cdot\hat{u}_j=c_ic_j+s_is_jc, \\ s_i\equiv\sin\vartheta_i, \quad s_j\equiv\sin\vartheta_j, \quad c_i\equiv\cos\vartheta_i, \quad c_j\equiv\cos\vartheta_j. \end{gathered} \tag{A18b}$$

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