

# Generation and characterization in a laboratory of $\mathbb{C}^2 \otimes \mathbb{C}^d$ states of flying electrons and ions with negative or positive partial transpose possessing free or bound entanglement

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In this paper, we show that bipartite states of an electronic qubit and ionic qudit, produced in a laboratory in as simple an experiment as photoionization in electric dipole approximation, can be separable or can possess either free or bound entanglement. The dimension of the Hilbert space of the qudit depends upon the electronic state  $|1^+\rangle$  of the residual photoion  $A^{1+}$ : In the absence of spin-orbit interaction (SOI), when Russell-Saunders coupling is applicable, it is equal to the spin multiplicity of  $|1^+\rangle$ , but becomes  $(2J + 1)$  in  $j$ - $j$  coupling, where  $J$  is the total angular momentum of  $|1^+\rangle$  when SOI is taken into account. In the case of  $L$ - $S$  coupling, all the properties (relevant to a study in quantum information) of a qubit-qudit state can readily be predicted merely with a knowledge of the spins of the atomic target and of the residual photoion which can experimentally be verified by measuring them using an entanglement witness, or any other such protocols, or by doing a full quantum-state tomography.

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## I. INTRODUCTION

The mode of information which uses quantum mechanical laws for its generation, coding, processing, etc., has come to be known as quantum information (QI) [1–3]. In analogy to the classical information, QI too needs bits. But, a bit in QI is a quantum system which has, at least, one observable requiring two-, or higher-, dimensional space for its characterization. A two-dimensional quantum system (e.g., a spin- $\frac{1}{2}$  particle, a two-level atom, etc.) has come to be called in QI as a quantum bit or qubit [4]; likewise, a *qutrit* is a three-dimensional quantum system (e.g., a spin-1 particle, a three-level atom, etc.). In general, a  $d$ -dimensional quantum system (with  $d \geq 4$ ) is called a *qudit* [5]. Applications of qudits provide an extension of the binary logic used both in QI and in classical information. The Hilbert space of  $N_d$  qudits contains the space of  $N_2$  qubits. It has, consequently, been suggested [6] that the use of qudits can reduce the number  $N_2$  of qubits, needed for a computation, by a factor of  $\ln_2 d$ . In addition, it has recently been shown [7–10] that the use of nonseparable (i.e., entangled) states of two qutrits in QI helps in developing quantum communication protocols which provide greater security in quantum cryptography [7], quantum communication complexity [8], and weak coin flipping [9,10]. Following these [7–10] and other similar developments, Vaziri *et al.* [11] have experimentally realized entanglement concentration of orbital angular momentum entangled photons in three dimensions.

Probably, only the bipartite states of photons, entangled in higher than two-dimensional space, have hitherto been successfully generated [11]. These photons are produced [11] in parametric down conversion. Although a photon is an excellent carrier of information, it is, however, not suitable for its long-term storage. Also, a photon is immediately destroyed as soon as one tries to detect it, unless a quantum nondemolition [12] type extremely difficult experiment is being performed. For these and other similar reasons, it will certainly be desirable to produce entangled states of two qudits made of atoms, molecules, etc., which, in addition to being amenable to easy detection without leading to their destruction, can be used also for storing the information for a long period.

In this paper, we discuss a very simple process in atoms for generating bipartite states of an electronic qubit (say,  $e_p$ ) and an ionic qudit (say,  $A^{1+}$ ). These two particles are entangled with respect to their either spin angular momenta in the absence of spin-orbit interaction (SOI), or spin of  $e_p$  and total angular momentum  $J_{1+}$  of  $A^{1+}$  when SOI too is taken into account. In addition to providing a qubit-qudit entanglement of two flying particles, both of which are readily detectable without any one being destroyed, the proposed study has several useful features: For example, there have recently been many experimental studies of quantum entanglement between a single photon and a single free atom in the continuously infinite Hilbert space (see, for example, [13,14]), as well as between a photonic and a trapped ionized (e.g., [15]), or neutral (e.g., [16]), atomic qubit in the finite-dimensional Hilbert space. Such studies have been initiated for fulfilling one of the outstanding goals in QI theory, namely, the faithful mapping of quantum information between a stable quantum memory and a reliable quantum communication channel [15,17]. This study too provides an alternative approach for achieving the same goal [15,17] of the QI theory by using an electronic, rather than a photonic, quantum communication channel. Although an electron can not travel as fast as a photon, there are nevertheless many other obvious advantages in using the former, in place of the latter, as a quantum communication channel.

Another interesting thing about the method proposed herein is that, in some of the cases discussed herein, one can analytically *a priori* determine or predict the presence (along with its other related properties), or absence, of entanglement between the spins of a ( $e_p, A^{1+}$ ) pair. Later, these properties of an entangled state of a ( $e_p, A^{1+}$ ) system can experimentally be verified by measuring them with an entanglement witness [18] or any other such protocols (e.g., [19]) hitherto developed in QI theory.

Furthermore, the recent research has shown that entangled states can also be of two types (see, for example, [3,20] and references therein): namely, the free entangled states and bound entangled states. A nonseparable state with entanglement which can be concentrated or purified is said

to possess free entanglement; if, on the other hand, such a procedure is not possible with a nonseparable state, it is then said to have bound entanglement. Concentration of entanglement means [3,20–25] combining the entanglement of several of those pure states which possess partial (i.e., less than maximum possible) entanglement into a single, but maximally entangled, pure state. More often than not, states which have partial entanglement are impure, i.e., mixed. The process of generating a maximally entangled pure state from several partially entangled mixed states is called entanglement purification or distillation (see, for example, [3,20,22,25–29]). The successful implementation of the protocols hitherto developed for concentration or purification of entanglement involves [3,20] only local operations and classical communications (LOCC) on separately addressable (i.e., distant) qubits of partially entangled pure or mixed states, respectively. Bennett *et al.* [28] have suggested that the task performed by these protocols in quantum information is equivalent to the reliable transmission of data through a noisy channel in classical information.

Although bound entangled states [3,20] too are nonseparable, their entanglement, however, can not [30] be concentrated or purified to generate a pure maximally entangled state. This kind of nondistillable entanglement is called [3,20,30] bound entanglement. Horodecki *et al.* [3,20] have discussed several consequences of bound entanglement. It has, for example, been shown [31] that, with the support of a bound entangled state, one can perform quantum teleportation [32] via a pair of particles which, otherwise, have an insufficient amount of free entanglement. Since their discovery, there have been considerable efforts for the generation and detection of states with bound entanglement (see, for example, [3,20,33–43] and references therein). In this paper, we show that photoionization in  $L$ - $S$  coupling is capable of producing qubit-qudit pairs possessing bound entanglement in a laboratory.

Section II contains a brief description of the required density operator. This operator is then used in Sec. III to obtain a density matrix (DM) needed to study the quantum informatics properties of a spin state of a photoelectron-photoion pair in Russell-Saunders coupling when SOI is not taken into account. Therein, we also discuss the properties of this DM. The respective Secs. III A and III B show how this DM can be used to generate states possessing free or bound entanglement between the spins of a photoelectron  $e_p$  and photoion  $A^{1+}$ . Entanglement between a photoelectron's spin angular momentum and photoion's total angular momentum  $J_{1+}$ , on the inclusion of SOI, is described in Sec. IV. Finally, Sec. V contains conclusions of the present investigations. The density matrices derived in Secs. III and IV can readily be reduced to obtain well-known expressions for angular distribution of photoelectrons in  $L$ - $S$  and in  $j$ - $j$  couplings, respectively, as well as that needed to study statistical tensorial properties (i.e., state multipoles) [44–46] of a photoion.

## II. DENSITY OPERATOR

Equation (1) represents the well-known process of ionization of an atom  $A$  caused due to the absorption of a single photon  $\gamma_r$  (the subscript “ $r$ ,” unless stated otherwise, stands

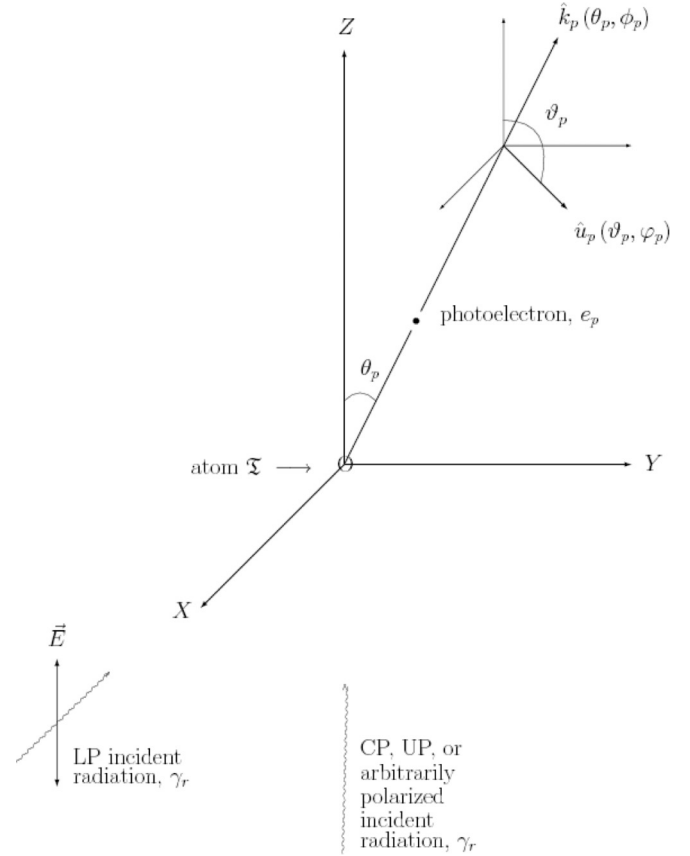


FIG. 1. Space-fixed (SF) [also called laboratory-fixed (LF)] frame of reference  $OXYZ$ . Its origin “O” is at the atomic target  $A$ . As explained in the text, direction of its polar  $OZ$  axis is shown in this figure to be determined by the polarization of the incident radiation  $\gamma_r$  which ionizes  $A$  in the processes (1). [According to that mentioned elsewhere in this paper, LP: linearly polarized  $\gamma_r$ ; CP: (right/left) circularly polarized  $\gamma_r$ ; UP: unpolarized  $\gamma_r$ .] Here,  $\hat{k}_p(\theta_p, \phi_p)$  and  $\hat{u}_p(\vartheta_p, \varphi_p)$  are, respectively, the directions of propagation and of spin quantization of the photoelectron  $e_p$  emitted in Eq. (1), whereas the angular momenta of the target  $A$  as well as those of its residual photoion  $A^{1+}$  are assumed to be quantized along the  $OZ$  axis.

for ionizing, incident, or absorbed radiation):

$$\gamma_r + A \longrightarrow A^{1+} + e_p. \quad (1)$$

Here,  $A^{1+}$  is the residual photoion formed after the ejection of the photoelectron  $e_p$  from the atom  $A$ . The respective kets  $|0\rangle$  and  $|1^+\rangle$  represent the antisymmetrized electronic states of  $A$  and of the photoion  $A^{1+}$  possessing the energies  $E_0$  and  $E_{1^+}$ . The propagation vector  $\vec{k}_p = (k_p \equiv |\vec{k}_p|, \theta_p, \phi_p)$  of the photoelectron  $e_p$  is such that its kinetic energy is given by  $\epsilon_p = \hbar^2 k_p^2 / 2m$ ; also,  $\mu_p = (\pm \frac{1}{2})$  is the projection of the spin of  $e_p$  along  $\hat{u}_p(\vartheta_p, \varphi_p)$  (both  $\vec{k}_p$  and  $\hat{u}_p$  are shown in Fig. 1). Thus, energy

$$\epsilon_p = h\nu_r - (E_{1^+} - E_0) \quad (2)$$

of  $e_p$  varies with the frequency  $\nu_r$  of the photon absorbed in Eq. (1). In the following discussion, unless stated otherwise, ket  $|1, m_r\rangle$  is used to suggest [47,48] that the process (1) is in the electric dipole approximation ( $E1$ ) specified by  $\gamma_r$ 's angular momentum  $|\ell_r| = 1$  and polarization  $m_r$ . Then,

$m_r = +1$  or  $-1$  is for photons with positive or negative helicity [also called photons with right circular polarization (RCP) or left circular polarization (LCP)], respectively; whereas,  $m_r = 0$  corresponds to a linearly polarized (LP) photon. [An unpolarized (UP) electromagnetic wave is taken to be an even mixture of photons with negative and positive helicities.] Accordingly, a photon absorbed in the process (1) in the state  $|1, +1\rangle$  or  $|1, -1\rangle$  is incident, but a photon in the  $|1, 0\rangle$  state has its electric field vector along the polar (i.e., OZ) axis of the space-fixed (SF) [or, laboratory-fixed (LF)] coordinate system OXYZ shown in Fig. 1.

Processes taking place in an atomic or a molecular target can, in general, be divided in two broad classes: namely, those that (a) are completed in a single step or (b) require more than one step for their completion. For example, photoionization in Eq. (1) is a one-step process because electron  $e_p$  is ejected from  $A$  in the same step (i.e., from the same electronic state of  $A$ ) in which photon  $\gamma_r$  is absorbed by this atom. Therefore, the process (1) has been called herein as a one-step single photoionization (1-SPI). The other well-known one-step process is double photoionization wherein simultaneous ejection of two electrons from an atom or a molecule takes place in the same step in which a photon is absorbed by this target. Such processes have appropriately been named [49,50] as one-step double photoionization (1-DPI). The simplest example of a process requiring more than one step for its completion is, on the other hand, the radiative, or nonradiative, decay of an excited state of an atom or a molecule. This is a two-step process: its first step consists of creation of an excited state, say, due to the absorption of a photon. The additional step of this two-step process requires decay of the excited state to the ground state of the original atom by fluorescence, or of the target's singly charged positive ion by ejecting an electron (called Auger electron). The resonant Auger decay [51] of an atom, for example, is a two-step single photoionization (2-SPI) as it consists of spontaneous nonradiative decay of an excited state of an atom created in a preceding step due to the absorption of a photon. Thus, in 2-SPI, an (Auger) electron comes out from an electronic state of the atom which is different from that in which a photon was absorbed.

It is shown in Appendix A [Eqs. (A4) and (A9a)] that the most general form of a density operator for any one-step process taking place due the absorption of a single photon is [49,50]

$$\rho_f = \mathcal{K}_p F_p \rho_1 F_p^\dagger. \quad (3a)$$

Here, while the constant  $\mathcal{K}_p$  and the photoionization operator  $F_p$  have been defined in Appendix A,

$$\rho_1 = \rho_0 \otimes \rho_r, \quad (3b)$$

with  $\rho_0 = |0\rangle\langle 0|$  and  $\rho_r = |1, m_r\rangle\langle 1, m_r|$  being the respective density operators [49,50] for the ionizing radiation  $\gamma_r$  and for the target atom  $A$ , assumed to be uncorrelated before the interaction between the two takes place.

The two important forces experienced by the constituents of an atom are due to the Coulomb interaction of electrons among themselves as well as with their nucleus, and the SOI. The latter of the two is significant primarily for heavy atoms [52,53]. In the absence of SOI, all the quantum informatics properties of a

$(e_p, A^{1+})$  pair we want to investigate in this paper will be due only to the Coulomb forces occurring in an atom. It is for this reason that the entanglement generated in these circumstances has appropriately been called (in Sec. III) as Coulombic entanglement. On the inclusion of SOI, the entanglement properties of a  $(e_p, A^{1+})$  system, on the other hand, are very much controlled by the same physical variables (e.g., the total angular momentum) which determine fine-structure (FS) levels [52,53] in an atom. The entanglement studied in the presence of SOI in Sec. IV has, consequently, been named as FS entanglement.

### III. COULOMBIC ENTANGLEMENT BETWEEN AN ELECTRONIC QUBIT AND AN IONIC QUDIT

On the exclusion of SOI from the atom  $A$ , its residual photoion  $A^{1+}$ , and from the continuum of the photoelectron  $e_p$ , the most suitable scheme for coupling the angular momenta relevant to the photoionization process (1) is the Russell-Saunders (i.e.,  $L$ - $S$ ) coupling [52,53]. Thus, in this section,  $L$ - $S$  coupling becomes applicable to the whole of the process (1). In addition, due to the spin-independent nature [54,55] of the photoionization operator  $F_p$  [see, for example, Eqs. (A7) and (A9)] in the density operator (3), both the total orbital and total spin angular momenta, before and after the emission of  $e_p$ , are individually conserved, i.e.,

$$\vec{\ell}_r + \vec{L}_0 = \vec{L}_{1+} + \vec{\ell}_p \quad (4a)$$

and

$$\vec{S}_0 = \vec{S}_{1+} + \vec{s}_p. \quad (4b)$$

Here,  $(\vec{L}_0, \vec{L}_{1+})$  and  $(\vec{S}_0, \vec{S}_{1+})$  are, respectively, the total orbital and spin angular momenta—quantized along the OZ-axis in Fig. 1—of the electronic states  $(|0\rangle, |1^+\rangle)$  of  $(A, A^{1+})$  participating in the process (1), whereas  $(\vec{\ell}_p, \vec{s}_p)$  (with  $|\vec{s}_p| = 1/2$ ) are (orbital, spin) angular momenta of the photoelectron  $e_p$  in Eq. (1), quantized along  $\hat{u}_p$  in the same figure.

The most general form of the density operator  $\rho_0 \otimes \rho_r$ , which represents in Eq. (3) an unpolarized atom  $A$  in the  $L$ - $S$  coupling plus a noninteracting photon, can be written as [49,50]

$$\begin{aligned} \rho_1 &\equiv \rho_0 \otimes \rho_r \\ &= \frac{1}{(2L_0 + 1)(2S_0 + 1)} \sum_{M_{L_0} M_{S_0}} |0; 1, m_r\rangle\langle 0; 1, m_r|. \end{aligned} \quad (5)$$

Here,  $|0; 1, m_r\rangle \equiv |0\rangle |1, m_r\rangle$  represents a state of the noninteracting (atom + photon) system;  $M_{L_0} (= \hat{e}_z \cdot \vec{L}_0)$  and  $M_{S_0} (= \hat{e}_z \cdot \vec{S}_0)$  are the projections of the respective angular momentum vectors  $\vec{L}_0$  and  $\vec{S}_0$  along the polar axis of our coordinate system in Fig. 1. Further, taking the target atom to be unpolarized, we have averaged in Eq. (5) over all the degenerate Zeeman components of  $A$  in its ground electronic state  $|0\rangle$ . Consequently, one can now write [52,53]  $|0\rangle \equiv |L_0 S_0 M_{L_0} M_{S_0}\rangle$  for the target atom  $A$  in  $L$ - $S$  coupling in the absence of SOI. Similarly,  $|1^+\rangle \equiv |L_{1+} S_{1+} M_{L_{1+}} M_{S_{1+}}\rangle$  for the ground electronic state of the residual photoion  $A^{1+}$ . Here, we have  $M_{L_{1+}} (= \hat{e}_z \cdot \vec{L}_{1+})$  and  $M_{S_{1+}} (= \hat{e}_z \cdot \vec{S}_{1+})$ .

In the entanglement between the spins of photoelectron  $e_p$  and of the residual photoion in the process (1), the Zeeman components ( $M_{L_{1+}} = -L_{1+}, -L_{1+} + 1, \dots, 0, \dots, L_{1+} - 1, L_{1+}$ ) of  $A^{1+}$  due to the space quantization of its orbital angular momentum  $L_{1+}$  are not observed. The required DM, obtained from (3), can therefore be written as

$$\begin{aligned} & \langle L_{1+} S_{1+} M_{S_{1+}}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | L_{1+} S_{1+} M'_{S_{1+}}; \mu'_p \hat{u}_p \vec{k}_p \rangle \\ &= \mathcal{K}_p \sum_{M_{L_{1+}}=-L_{1+}}^{L_{1+}} \langle L_{1+} S_{1+} M_{L_{1+}} M_{S_{1+}}; \\ & \quad \mu_p \hat{u}_p \vec{k}_p | (F_p \rho_1 F_p^\dagger) | L_{1+} S_{1+} M_{L_{1+}} M'_{S_{1+}}; \\ & \quad \mu'_p \hat{u}_p \vec{k}_p \rangle. \end{aligned} \quad (6a)$$

This, on substituting (5), finally becomes

$$\begin{aligned} & \langle L_{1+} S_{1+} M_{S_{1+}}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | L_{1+} S_{1+} M'_{S_{1+}}; \mu'_p \hat{u}_p \vec{k}_p \rangle \\ &= \frac{\mathcal{K}_p}{(2L_0 + 1)(2S_0 + 1)} \sum_{M_{L_0} M_{S_0} M_{L_{1+}}} \\ & \quad \times \langle 1^+; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1m_r \rangle \\ & \quad \times \langle 0; 1m_r | F_p^\dagger | L_{1+} S_{1+} M_{L_{1+}} M'_{S_{1+}}; \mu'_p \hat{u}_p \vec{k}_p \rangle. \end{aligned} \quad (6b)$$

Expression (6b) is evaluated in Appendix B. In its final simplified form, given in Eq. (B2), it is shown to be written as a product of two independent terms. In the derivation of (B2), only the Coulomb interactions among the constituents of an atom have been taken into account. Expression (B2) has, therefore, been called as Coulombic DM.

The first term (B3), present on the right-hand side of the Coulombic DM (B2), contains, among other things, the state of polarization (specified by  $m_r$ ) of the ionizing radiation used in Eq. (1) and the kinematics (i.e., direction of propagation  $\hat{k}_p$ , energy  $\epsilon_p$ ) of the photoelectron  $e_p$ . It, in addition, has total orbital angular momenta ( $L_0, L_{1+}$ ) of ( $A, A^{1+}$ ), the dynamical amplitudes (B3b) for the  $E1$  photoionization (calculated using the bound electronic states of  $A, A^{1+}$ , and the continuum wave function of the photoelectron  $e_p$ ). Also, this first term [i.e., (B3)] in Eq. (B2) has an implicit dependence upon the spins ( $S_0, S_{1+}$ ) because, in  $L$ - $S$  coupling, the energies ( $E_0, E_{1+}$ ) of the electronic states ( $|0\rangle, |1^+\rangle$ ) of ( $A, A^{1+}$ ) depend upon their respective spin multiplicity. It is, however, totally independent of the quantization directions  $\hat{u}_p$  as well as of the spin of the photoelectron  $e_p$  emitted in Eq. (1). Hence,  $d^2\sigma(m_r)/d\epsilon_p d\hat{k}_p$  in Eq. (B3) describes purely angular correlation between ( $e_p, A^{1+}$ ). Expression (B3) is, indeed, the spin-unresolved angular distribution [56] for electrons emitted in photoionization of an atom in  $E1$  approximation in  $L$ - $S$  coupling. Its presence in the Coulombic DM (B2) is merely as a positive multiplicative factor which does not have any explicit dependence on any spin quantum numbers.

Let us now consider the second term (B4), present on the right-hand side of the Coulombic DM (B2). Unlike the angular distribution  $d^2\sigma(m_r)/d\epsilon_p d\hat{k}_p$ , (B4) does not contain any of those physical variables which are present in Eq. (B3) and, hence, is totally independent of both the kinematical and dynamical effects contributing to the process (1). Moreover,

(B4) is *completely determined* by the spins of all the three particles (i.e.,  $A, A^{1+}, e_p$ ) involved in the process (1) and by the direction  $\hat{u}_p$  of the spin quantization of the photoelectron  $e_p$ . Hence, the second term (B4), in the Coulombic DM (B2), represents purely spin correlation between ( $e_p, A^{1+}$ ).

The separation obtained in Eq. (B2) of the DM into its two parts describing purely angular and purely spin correlations between ( $e_p, A^{1+}$ ) in the absence of SOI in Russell-Saunders coupling is completely rigorous and independent of all dynamical models. It is well known (see, for example, [44,52–54]) that the state of an atom or a molecule, in the absence of SOI, is a product of its spatial and spin parts. A DM too represents [45] the state of a system. The separation of angular and spin parts in Eq. (B2) for the 1-SPI process (1) in the present case, and for other such processes elsewhere (e.g., [49,50]), is also a consequence of the exclusion of SOI, i.e., consideration of the Hamiltonians (A5) and (A6a).

In expression (B4), although the allowed values for each of  $\mu_p$  and  $\mu'_p$  are  $\pm\frac{1}{2}$ , the spin magnetic quantum numbers  $M_{S_{1+}}$  and  $M'_{S_{1+}}$  of the photoion  $A^{1+}$  can each take  $(2S_{1+} + 1)$  values  $-S_{1+}, -S_{1+} + 1, \dots, S_{1+} - 1, S_{1+}$ . Consequently, (B4) is a matrix of size  $[2(2S_{1+} + 1) \times 2(2S_{1+} + 1)]$  representing spin correlation between the electronic qubit  $e_p$  and  $d$  [ $=2(2S_{1+} + 1)$ ]-dimensional ionic qudit  $A^{1+}$ . Hence, the dimensionality  $d$  of the qudit can be chosen according to one's requirement by considering the atomic target  $A$  and photoion  $A^{1+}$  in Eq. (1) of appropriate spins  $S_0$  and  $S_{1+}$ , respectively.

The diagonal terms (i.e., those with  $\mu_p = \mu'_p$  and  $M_{1+} = M'_{1+}$ ) in the DM (B2) describe spin-resolved angular distribution of photoelectrons ejected in the process (1) in  $L$ - $S$  coupling, whereas the nondiagonal terms (i.e., those with  $\mu_p \neq \mu'_p$  or  $M_{1+} \neq M'_{1+}$ ) represent coherent effects in the DM (B2). The other interesting thing about the spin-correlation matrix (B4) is that each of its  $[2(2S_i + 1) \times 2(2S_{1+} + 1)]$  elements is readily calculated once the the spins ( $S_0, S_{1+}$ ) of the electronic states ( $|L_0 S_0 M_{L_0} M_{S_0}\rangle, |L_{1+} S_{1+} M_{L_{1+}} M_{S_{1+}}\rangle$ ) of ( $A, A^{1+}$ ), participating in the photoionization process (1) in  $L$ - $S$  coupling, are known.

It is obvious from the spin-conservation condition (4b), as well as from the 6- $j$  symbol [57] present in Eq. (B4), that spins ( $S_0, S_{1+}$ ) of ( $A, A^{1+}$ ) in the process (1) in the absence of SOI are related by  $S_0 - S_{1+} = \pm\frac{1}{2}$ . The only two possibilities are therefore (i)  $S_0 = S_{1+} - \frac{1}{2}$  or (ii)  $S_0 = S_{1+} + \frac{1}{2}$ , with  $S_{1+} \geq \frac{1}{2}$  in both cases. (For  $S_{1+} = 0$ , multiplicity of the electronic state  $|1^+\rangle$  of the photoion  $A^{1+}$  will be one and, hence, it will simply become a single-state system.)

In the following, we investigate the spin-entanglement between ( $e_p, A^{1+}$ ) in both of these cases. This property of the qubit-qudit system will be completely determined by the matrix (B4) as the angular correlation in Eq. (B2) is always *positive* and acts as a *multiplicative* factor. Therefore, unless stated otherwise, we write the Coulombic DM (B2) as

$$\begin{aligned} & \langle L_{1+} S_{1+} M_{S_{1+}}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | L_{1+} S_{1+} M'_{S_{1+}}; \mu'_p \hat{u}_p \vec{k}_p \rangle \\ & \longrightarrow \sigma(S_0; S_{1+}; \hat{u}_p)_{\mu_p, M_{S_{1+}}; \mu'_p, M'_{S_{1+}}}. \end{aligned} \quad (7)$$

In order to study the entanglement properties of a ( $e_p, A^{1+}$ ) pair, one needs to calculate the partial transpose [58,59] of the DM (7) with respect to either of its two subsystems, i.e.,



photoelectron  $e_p$  or photoion  $A^{1+}$ . These are given by

$$\begin{aligned} \sigma^{T_e}(S_0; S_{1+}; \hat{u}_p)_{\mu_p, M_{S_{1+}}; \mu'_p, M'_{S_{1+}}} \\ = \sigma(S_0; S_{1+}; \hat{u}_p)_{\mu'_p, M'_{S_{1+}}; \mu_p, M_{S_{1+}}} \end{aligned} \quad (8a)$$

and

$$\begin{aligned} \sigma^{T_{1+}}(S_0; S_{1+}; \hat{u}_p)_{\mu_p, M_{S_{1+}}; \mu'_p, M'_{S_{1+}}} \\ = \sigma(S_0; S_{1+}; \hat{u}_p)_{\mu_p, M'_{S_{1+}}; \mu'_p, M_{S_{1+}}} \end{aligned} \quad (8b)$$

respectively. Although the matrices (8a) and (8b) are certainly different, their eigenvalues will, nevertheless, be always identical because one is the (full) transpose of the other. Consequently, one can calculate either of the two partial transposes (8) for studying the Coulombic entanglement properties of a  $(e_p, A^{1+})$  pair generated in the process (1) in the absence of SOI in  $L$ - $S$  coupling. In this study, we have always calculated, unless stated otherwise, the partial transpose of the DM (7) with respect to the photoelectron  $e_p$ , i.e., used Eq. (8a).

### A. Free Coulombic entanglement

Let us consider first [i.e., (i)  $S_0 = S_{1+} - \frac{1}{2}$ , with  $S_{1+} \geq \frac{1}{2}$ ] of the two cases mentioned earlier herein. For this case, the DM (7) readily simplifies to

$$\begin{aligned} \sigma \left( S_0 = S_{1+} - \frac{1}{2}; S_{1+}; \hat{u}_p \right)_{\mu_p, M_{S_{1+}}; \mu'_p, M'_{S_{1+}}} \\ = \frac{1}{2(2S_{1+} + 1)} \delta_{\mu_p \mu'_p} \delta_{M_{S_{1+}} M'_{S_{1+}}} + (-1)^{\frac{1}{2} + \mu'_p + S_{1+} - M_{S_{1+}}} \\ \times \sqrt{\frac{3(S_{1+} + 1)}{2S_{1+}(2S_{1+} + 1)}} \sum_{mn} \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & 1 \\ \mu_p & -\mu'_p & m \end{pmatrix} \\ \times \begin{pmatrix} S_{1+} & S_{1+} & 1 \\ M_{S_{1+}} & -M'_{S_{1+}} & -n \end{pmatrix} [D_{mn}^1(\omega_p)]^* \end{aligned} \quad (9)$$

with  $\omega_p = (\varphi_p, \vartheta_p, 0)$ . It is not possible to diagonalize the DM (9) without specifying a value of the spin  $S_{1+}$  of the photoion  $A^{1+}$  participating in the photoionization (1). Consequently, we individually calculated (9) for each of  $S_{1+} = (\frac{1}{2}, 1, \frac{3}{2}, 2, \frac{5}{2}, 3)$  corresponding to  $S_0 = (0, \frac{1}{2}, 1, \frac{3}{2}, 2, \frac{5}{2})$ , respectively. It, in other words, means that one is here looking at the bipartite spin states of the qubit of the photoelectron  $e_p$  and the qudit of the photoion  $A^{1+}$  defined in the Hilbert spaces of dimensions  $d = (2, 3, 4, 5, 6, 7)$ , while the spin multiplicities of the electronic states of  $A$  in Eq. (1) are  $(1, 2, 3, 4, 5, 6)$ . The diagonalization of each of these six DMs and of their partial transpose (PT) gave us the following eigenvalues (EVs):

$$\text{DM(9)} \begin{bmatrix} 2(S_{1+} + 1) & \text{EVs} = 0 \\ 2S_{1+} & \text{EVs} = (2S_{1+})^{-1} \end{bmatrix}, \quad (10a)$$

PT (8a) of DM (9)

$$\begin{bmatrix} 2(S_{1+} + 1) & \text{EVs} = (2S_{1+} + 1)^{-1} \\ 2S_{1+} & \text{EVs} = -[2S_{1+}(2S_{1+} + 1)]^{-1} \end{bmatrix}. \quad (10b)$$

The eigenvalues (10) mean that a spin state of  $(e_p, A^{1+})$ , formed in a photoionization process (1) taking place in an unpolarized atom when its constituents experience only the Coulomb interaction, has the following properties:

(a) The total number of eigenvalues in each of the two cases in Eq. (10) is equal to  $2(2S_{1+} + 1)$ , i.e., the dimensionality of the DM (9).

(b) The sum of all the eigenvalues in Eq. (10a), as well as in Eq. (10b), is always equal to one, implying that the DM (9) is normalized to unit trace.

(c) According to (10a), all density matrices (9), other than those with  $(S_0 = 0, S_{1+} = \frac{1}{2})$ , have more than one nonzero eigenvalue. Consequently [45],  $(e_p, A^{1+})$  are in a pure spin state if the photoion is also a qubit; otherwise, for all values of the spin  $S_0 = S_{1+} - \frac{1}{2}$  of  $A$ , with  $S_{1+} \geq 1$  of  $A^{1+}$ ,  $(e_p, A^{1+})$  always form a mixed state.

(d) Some of the eigenvalues (10b) of the partial transpose of the DM (9) are always negative. Thus, each of the spin states (9), generated in the photoionization (1) with  $(S_0 = S_{1+} - \frac{1}{2}, \text{ where } S_{1+} \geq \frac{1}{2})$ , has negative partial transpose (NPT). These are called [3,20] NPT states.

(e) A straightforward application of the very stringent, necessary, and sufficient condition [58,59] for the separability of a bipartite state shows that, in the present case,  $(e_p, A^{1+})$  are always entangled with respect to their spin angular momenta.

(f) Negativity [60–62], concurrence [63], and entanglement of formation [63] (EoF) are the three measures currently in vogue for quantifying the amount of entanglement present in a nonseparable state. Of the three, however, negativity is the only measure which can be calculated for a bipartite state of other than one or both qubits. It is defined as [62]

$$\mathcal{N}(\rho) = 2 \max \left\{ 0, -\sum [\text{negative eigenvalues of partial transpose of } (\sigma)] \right\}. \quad (11)$$

On substituting the eigenvalues (10b) in the definition (11), one immediately obtains

$$\begin{aligned} \mathcal{N} \left[ \sigma \left( S_0 = S_{1+} - \frac{1}{2}; S_{1+}; \hat{u}_p \right) \right] = \frac{2}{2S_{1+} + 1} \quad \text{with} \\ S_{1+} \geq \frac{1}{2} \end{aligned} \quad (12)$$

for the negativity of the state (9) of a  $(e_p, A^{1+})$  system.

(g) Thus, negativity is one for a state with  $(S_0 = 0, S_{1+} = \frac{1}{2})$  and asymptotically goes to  $(S_{1+})^{-1}$  with increasing spin of the electronic state  $|1^+\rangle$  of the photoion  $A^{1+}$  generated in the process (1). In other words,  $(e_p, A^{1+})$  are maximally entangled if the photoion formed in Eq. (1) is also a qubit; otherwise, an increase in the dimensions  $(2S_{1+} + 1)$  of the Hilbert space of the ionic qudit weakens the entanglement of its spin angular momentum with that of the electronic qubit.

(h) In view of the properties (c) and (g),  $(e_p, A^{1+})$  are in a pure and maximally entangled state when  $S_0 = 0$  and  $S_{1+} = \frac{1}{2}$ . It, nevertheless, is an obvious result. In this case, the ground electronic states  $(|0\rangle, |1^+\rangle)$  of  $(A, A^{1+})$  participating in the photoionization process (1) are (singlet, doublet). Then, the conservation condition (4b) demands that the sum of the

spin angular momenta of (electronic, ionic) qubits ( $e_p, A^{1+}$ ) should also be zero, i.e., these two particles should be in a singlet spin state which is pure as well as maximally entangled. Indeed, it can readily be verified by explicitly calculating all the 16 elements of the DM (9) for  $S_0 = 0$  and  $S_{1+} = \frac{1}{2}$ . On specializing, in the resulting matrix, the spin quantization direction  $\hat{u}_p(\vartheta_p, \varphi_p)$  of photoelectron  $e_p$  along the polar axis of the SF in Fig. 1 (i.e., taking  $\vartheta_p = 0$ ), one finds that it reduces to the  $(4 \times 4)$  matrix given in Ref. [1] for the singlet state of two spin- $\frac{1}{2}$  particles. This is one of the pure and maximally entangled four Bell states [1–3].

(i) Eigenvalues of the DM (9) and of its partial transpose (8a) are only of two different varieties in each of (10a) and (10b), respectively. These are, almost, evenly distributed [with values  $[0$  and  $(2S_{1+})^{-1}$  in Eq. (10a)],  $[(2S_{1+} + 1)^{-1}$  and  $-(2S_{1+}(2S_{1+} + 1))^{-1}$  in Eq. (10b)].

(1) The preceding point (h) gives one of the plausible reasons for this even distribution of the eigenvalues in Eq. (10a). The conservation of the spin angular momentum for the Coulombic Hamiltonians (A5) and (A6a) requires that, whenever ( $S_0 = 0, S_{1+} = \frac{1}{2}$ ), ( $e_p, A^{1+}$ ) have to be in a singlet spin state which is pure. It means, for these values of ( $S_0, S_{1+}$ ) in the photoionization process (1), three of the four eigenvalues of the DM (9) must be zero, while the remaining fourth eigenvalue must be unity. This requirement determines in Eq. (10a) how many of the eigenvalues of the DM (9) should be 0 even for  $S_{1+} > \frac{1}{2}$ . The number of nonzero eigenvalues will then obviously be [(total number of eigenvalues) – (number of zero eigenvalues)]. This, in the present case, is equal to  $2S_{1+} [= 2(2S_{1+} + 1) - 2(S_{1+} + 1)]$ . Obviously, as required, the sum of all the nonzero eigenvalues in Eq. (10a) is unity.

(2) It has theoretically been shown [64] that the partial transpose of an entangled bipartite state of qubits can have only one of its four eigenvalues to be negative, while, in the case of a qubit-qudit bipartite entangled state, the number of negative eigenvalues of the DM (9) may be more than one. This is one of the possible reasons for the distribution to certain values in Eq. (10b) of the eigenvalues of the partial transpose of the DM (9).

(j) Some of the simplest possible examples of the result discussed in (h) are photoionization of  $\text{He}(1s^2 1S) \rightarrow \text{He}^{1+}(1s^1 2S)$ ,  $\text{Be}(1s^2 2s^2 1S) \rightarrow \text{Be}^{1+}(1s^2 2s^1 2S)$ ,  $\text{C}(1s^2 2s^2 2p^2 1S/1D) \rightarrow \text{C}^{1+}(1s^1 2s^2 2p^1 2P)$ , etc. The SOI is obviously absent in these electronic states of He,  $\text{He}^{1+}$ , Be,  $\text{Be}^{1+}$ , and C; it certainly is negligible also for  $\text{C}^{1+}$  and for the photoelectron  $e_p$  moving in the continuum of these photoions of some of the lightest atoms. In each of these cases, ( $e_p, A^{1+}$ ) are in a singlet spin state which is pure and maximally entangled.

(k) The examples of mixed ( $e_p, A^{1+}$ ) states wherein their spin angular momenta have less than maximum possible Coulombic entanglement (i.e., are partially entangled) include  $\text{F}(1s^2 2s^2 2p^5 2P) \rightarrow \text{F}^{1+}(1s^1 2s^2 2p^4 3P)$ . Here, ( $e_p, \text{F}^{1+}$ ) are in a mixed Coulombic state of a qubit-qudit system whose negativity is  $\frac{2}{3}$ . Yet, another example of mixed and partial Coulombic entanglement is  $\text{O}(1s^2 2s^2 2p^4 3P) \rightarrow \text{O}^{1+}(1s^1 2s^2 2p^3 4S)$  with negativity equal to  $\frac{1}{2}$ . That is, spins of ( $e_p, \text{O}^{1+}$ ) are in a mixed and partially entangled Coulombic state of a qubit-qudit system. Each of these, and

other, Coulombic bipartite states can readily be produced and detected in a laboratory.

Earlier, Kim *et al.* [65] have considered spin entanglement of ( $e_p, A^{1+}$ ) generated in the photoionization of only the singlet and triplet electronic states of an atom  $A$  described in the independent particle model [52,53] by two *equivalent* (i.e.,  $ns^2$ ), or *nonequivalent* (i.e.,  $ns^1 n's^1$ ), bound electrons in  $L$ - $S$  coupling. The analysis given in this section, on the other hand, is completely general, independent of all dynamical models, and applicable to any allowed states ( $|0\rangle, |1^+\rangle$ ) of ( $A, A^{1+}$ ), irrespective of their electronic configurations, participating in the photoionization process (1) in Russell-Saunders coupling in the absence of SOI.

## B. Bound Coulombic entanglement

In this section, we investigate the second [i.e., (ii)  $S_0 = S_{1+} + \frac{1}{2}$ , with  $S_{1+} \geq \frac{1}{2}$ ] of the two possibilities arising from the spin-conservation condition (4b). On specializing the DM (7) to this case, one finds

$$\begin{aligned} & \sigma \left( S_0 = S_{1+} + \frac{1}{2}; S_{1+}; \hat{u}_p \right)_{\mu_p, M_{S_{1+}}; \mu'_p M'_{S_{1+}}} \\ &= \frac{1}{2(2S_{1+} + 1)} \delta_{\mu_p \mu'_p} \delta_{M_{S_{1+}} M'_{S_{1+}}} + (-1)^{\mu'_p - \frac{1}{2} + S_{1+} - M_{S_{1+}}} \\ & \times \sqrt{\frac{3S_{1+}}{2(S_{1+} + 1)(2S_{1+} + 1)}} \sum_{mn} \begin{pmatrix} 1/2 & 1/2 & 1 \\ \mu_p & -\mu'_p & m \end{pmatrix} \\ & \times \begin{pmatrix} S_{1+} & S_{1+} & 1 \\ M_{S_{1+}} & -M'_{S_{1+}} & -n \end{pmatrix} [\mathcal{D}_{mn}^1(\varphi_p, \vartheta_p, 0)]^* \quad (13a) \\ &= \frac{1}{2(S_{1+} + 1)} \delta_{\mu_p \mu'_p} \delta_{M_{S_{1+}} M'_{S_{1+}}} \\ & - \frac{S_{1+}}{S_{1+} + 1} \sigma \left( S_0 = S_{1+} - \frac{1}{2}; S_{1+}; \hat{u}_p \right)_{\mu_p, M_{S_{1+}}; \mu'_p M'_{S_{1+}}} \quad (13b) \end{aligned}$$

In Eq. (13b), the required DM (13a) has been written in terms of the DM (9) obtained earlier for  $S_0 = S_{1+} - \frac{1}{2}$  with  $S_{1+} \geq \frac{1}{2}$ . The relationship (13b) between the two density matrices is true also for their partial transpose as well as for their diagonalized forms.

Using (13b) and (10a), one readily obtains the following eigenvalues:

$$\begin{aligned} 2(S_{1+} + 1) \quad \text{EVs} &= [2(S_{1+} + 1)]^{-1}, \\ 2S_{1+} \quad \text{EVs} &= 0 \end{aligned} \quad (14a)$$

for the DM (13a). Similarly,

$$\begin{aligned} 2(S_{1+} + 1) \quad \text{EVs} &= [2(S_{1+} + 1)(2S_{1+} + 1)]^{-1}, \\ 2S_{1+} \quad \text{EVs} &= (2S_{1+} + 1)^{-1} \end{aligned} \quad (14b)$$

are the eigenvalues of the partial transpose of the DM (13a) and are gotten by combining Eqs. (13b) and (10b). The eigenvalues (14) show that a spin state of ( $e_p, A^{1+}$ ) formed in the present case has the following properties:

(a) Properties (a) and (b), mentioned in Sec. III A for the case [(i)  $S_0 = S_{1+} - \frac{1}{2}$ , with  $S_{1+} \geq \frac{1}{2}$ ] are applicable even in the present case.

(b) For all values of  $(S_0, S_{1+})$  appropriate for the DM (13), the number of nonzero eigenvalues in Eq. (14) is always greater than one. Hence, none of the spin state of  $(e_p, A^{1+})$ , generated in the present case is pure.

(c) None of the eigenvalues (14b) of the partial transpose of the DM (13) is negative. Thus, each of the spin states of  $(e_p, A^{1+})$ , generated in the photoionization process (1) with  $(S_0 = S_{1+} + \frac{1}{2}$  and  $S_{1+} \geq \frac{1}{2})$ , has positive partial transpose (PPT).

(d) Peres [58] has shown that if the DM of a composite system has PPT, its quantum state is then necessarily separable. Horodecki *et al.* (see, for example, [3,20,59]), while analyzing Peres's suggestion, found that it [58] is also a sufficient condition for the separability, provided the product of the dimensions of the Hilbert spaces of the subsystems does not exceed six. It, in other words, means [58,59] that a PPT state of a qubit-qubit, or a qubit-qutrit, system is always separable. Hence,  $(e_p, A^{1+})$  generated in the photoionization process (1) with  $(S_0 = 1, S_{1+} = \frac{1}{2})$  or  $(S_0 = \frac{3}{2}, S_{1+} = 1)$ , have no Coulombic entanglement at all. Examples include  $C(1s^2 2s^2 2p^2 3P) \rightarrow C^{1+}(1s^2 2s^2 2p^1 2P)$ . Here,  $C^{1+}$  is obviously a two-level system and, according to the eigenvalues (14),  $(e_p, C^{1+})$  are in a mixed product state. Similarly, in the photoionizing transition  $N(1s^2 2s^2 2p^3 4S) \rightarrow N^{1+}(1s^1 2s^2 2p^2 3P)$ , the residual photoion  $N^{1+}$  forms with the photoelectron  $e_p$  a qubit-qutrit system which too is in a mixed, separable state.

(e) Horodecki [66] has shown that a PPT state with dimension of the Hilbert space of its composite system more than six may also be nonseparable, i.e., entangled. However, the entanglement present in such PPT states has come to be known (see, for example, [3,20,30] and references therein) as bound entanglement whose properties are very different from that, called free entanglement, of a NPT state discussed in Sec. III A.

(f) In view of the discussion given in (e) herein, for a PPT state (13) of  $(e_p, A^{1+})$  to possess bound entanglement, it is necessary that  $2(2S_{1+} + 1) > 6$ , i.e., the spin multiplicity of its residual photoion  $A^{1+}$ , formed in the photoionization process (1), must be more than three, i.e., the spin angular momentum of its electronic state  $|1^+\rangle$  must at least be  $\frac{3}{2}$ . It, in other words, means that  $A^{1+}$  must, at least, be a four-state system.

(g) Many inequalities (see, for example, Refs. [66–69] or review articles [3,20,70]) have been obtained, the satisfaction, or violation, of which is suggested to decide the presence, or absence, of bound entanglement in a given PPT bipartite state. In order to see whether the PPT states (13) with  $2(2S_{1+} + 1) > 6$  are nonseparable, we have used two different criteria based upon the rank [71] and range [72] of the DM (13). According to the first of these two criteria [3,20,67–69], for the PPT state (13) with  $S_{1+} > 1$  to be nonseparable, its rank must satisfy the inequality

$$\text{Rank} \left[ \sigma \left( S_0 = S_{1+} + \frac{1}{2}; S_{1+} > 1; \hat{u}_p \right) \right] \geq \text{Max} \{ \text{Rank}[\rho^{e_p}], \text{Rank}[\rho^{1+}] \}. \quad (15)$$

Here,

$$\rho^{e_p} = \sum_{\mu_{S_{1+}}} \sigma \left( S_0 = S_{1+} + \frac{1}{2}; S_{1+} > 1; \hat{u}_p \right)_{\mu_p, M_{S_{1+}}; \mu_p', M_{S_{1+}}'} \quad (16a)$$

and

$$\rho^{1+} = \sum_{\mu_p} \sigma \left( S_0 = S_{1+} + \frac{1}{2}; S_{1+} > 1; \hat{u}_p \right)_{\mu_p, M_{S_{1+}}; \mu_p, M_{S_{1+}}'} \quad (16b)$$

are the reduced matrices, calculated from the DM (13), for the photoelectron  $e_p$  and the residual photoion  $A^{1+}$ , respectively. Both of these reduced matrices were found to be diagonal of the respective sizes  $(2 \times 2)$  and  $[(2S_{1+} + 1) \times (2S_{1+} + 1)]$ . Each of the diagonal elements of  $\rho^{e_p}$  is  $\frac{1}{2}$  and that of  $\rho^{1+}$  is  $(2S_{1+} + 1)^{-1}$ , always. Hence, ranks of the two respective reduced matrices are two and  $(2S_{1+} + 1)$ . But, according to (14a), DM (13) is of rank  $2(S_{1+} + 1)$ . Thus, in the present case, the PPT state (13) always satisfies the bound (15) for  $S_{1+} \geq \frac{3}{2}$  and, hence, possess bound entanglement.

(h) Let us take  $|v_n\rangle$  to be the eigenvector of the  $n$ th nonzero eigenvalue (14a) of the PPT DM (13) for  $S_{1+} \geq \frac{3}{2}$ . Then, the range

$$\text{Range} \left[ \sigma \left( S_0 = S_{1+} + \frac{1}{2}; S_{1+} > 1; \hat{u}_p \right) \right] = \sum_{n=1}^{2(S_{1+}+1)} C_n |v_n\rangle \quad (17)$$

of this state is the linear combination of the  $2(S_{1+} + 1)$  eigenvectors of the nonzero eigenvalues given in Eq. (14a). In Eq. (17),  $C_n$  are the coefficients of this linear combination. The range criterion for the nonseparability of the PPT bipartite state (13) requires [34,66] that the following  $2(2S_{1+} + 1)$  coupled simultaneous equations

$$\begin{pmatrix} x_1 \\ x_2 \end{pmatrix} \otimes \begin{pmatrix} y_{S_{1+}} \\ y_{S_{1+}-1} \\ \dots \\ \dots \\ y_{-S_{1+}-1} \\ y_{-S_{1+}} \end{pmatrix} = \sum_{n=1}^{2(S_{1+}+1)} C_n |v_n\rangle \quad (18)$$

should have only trivial solutions. The DM (13) was found to meet this requirement for all the  $(e_p, A^{1+})$  states with  $S_{1+} \geq \frac{3}{2}$ .

(i) Hence, a spin state of  $(e_p, A^{1+})$ , generated in the photoionization process (1) in the  $L$ - $S$  coupling with  $S_0 = S_{1+} + \frac{1}{2}$  and  $S_{1+} \geq \frac{3}{2}$  is always nonseparable, although it possesses PPT. One of the several examples of such photoionizing transitions is  $\text{Fe}(1s^2 2s^2 2p^6 3s^2 3p^6 4s^2 3d^6 5D) \rightarrow \text{Fe}^{1+}(1s^2 2s^2 2p^6 3s^2 3p^6 4s^2 3d^5 4D/4G)$  wherein  $(e_p, \text{Fe}^{1+})$  form a PPT state of qubit-qutrit (with  $d = \frac{3}{2}$ ) possessing bound entanglement. Although the SOI is negligible in none of the species  $(\text{Fe}, \text{Fe}^{1+}, e_p)$  participating in the  $E1$  process (1), nevertheless, the present example very well illustrates as to how one can generate states of an electronic qubit and ionic qutrit possessing bound Coulombic entanglement. It further shows that this entanglement can readily be predicted with a knowledge of the spins  $(S_0, S_{1+})$  of  $(A, A^{1+})$  obtained simply by measuring the energy of the photoion in a laboratory in the  $L$ - $S$  coupling.

Although both Secs. III A and III B discuss properties of the Coulombic entanglement between the spins of  $(e_p, A^{1+})$ , however, the results obtained in the two cases are quite different. The Coulombic DM (7) [i.e., (B4)] contains, among other things, spins  $(S_0, S_{1+})$  of  $(A, A^{1+})$  in one of its two 3- $j$  symbol and one 6- $j$  symbol. It is therefore very natural that, with a change in the relationship between the values of  $S_0$  and  $S_{1+}$  used to arrive at (13a), the PPT DM should be very different from that [i.e., expression (9)] for the NPT states. This indeed is obvious from the expression (13b) for the former given in terms of the latter [i.e., DM (9)]. Consequently, the properties of the Coulombic entanglement of a spin state of  $(e_p, A^{1+})$  generated in the photoionization process (1) in Sec. III B are very different from those discussed in Sec. III A, although the physical assumption (i.e., absence of SOI) involved in the two cases is the same.

Thus, purely Coulombic interaction, experienced by the constituents of an atom, is capable of generating (qubit, qudit) spin states of  $(e_p, A^{1+})$  with bound entanglement for appropriate values of the spins  $(S_0, S_{1+})$  of the electronic states  $(|0\rangle, |1^+\rangle)$  of  $(A, A^{1+})$  participating in as simple a process as photoionization in the absence of SOI.

#### IV. FINE-STRUCTURE ENTANGLEMENT BETWEEN AN ELECTRONIC QUBIT AND AN IONIC QUDIT

In this section, we study the  $E1$  process (1) taking SOI into account, i.e., using the Hamiltonians (A5) and (A6b). Then,  $j$ - $j$  coupling [52,53] of angular momenta is, obviously, the most appropriate choice to work with. If  $\vec{J}_0 = \vec{L}_0 + \vec{S}_0$  and  $\vec{J}_{1+} = \vec{L}_{1+} + \vec{S}_{1+}$  are the total angular momenta of atomic

target  $A$  and of its residual photoion  $A^{1+}$  participating in Eq. (1), then the condition for their conservation becomes

$$\vec{\ell}_r + \vec{J}_0 = \vec{J}_{1+} + \vec{j}_p \left[ = \vec{\ell}_p + \vec{s}_p \left( = \frac{\vec{1}}{2} \right) \right]. \quad (19)$$

Here,  $\vec{j}_p$  is the total angular momentum of the photoelectron  $e_p$ . Thus, two conditions [(4a) and (4b)] for the conservation of (orbital, spin) angular momenta individually in  $L$ - $S$  coupling are replaced by a single condition (19) in  $j$ - $j$  coupling. Although the conservation condition (2) for energy in the process (1) still remains the same, however,  $(E_0, E_{1+})$  are now the energies of  $(A, A^{1+})$  in their ground electronic states  $(|0\rangle = |J_0 M_0\rangle, |1^+\rangle = |J_{1+} M_{1+}\rangle)$  in  $j$ - $j$  coupling [52,53]. Here,  $M_0 = \hat{e}_z \cdot \vec{J}_0$  and  $M_{1+} = \hat{e}_z \cdot \vec{J}_{1+}$  are the projections of the total angular momenta  $\vec{J}_0$  and  $\vec{J}_{1+}$ , respectively, along the polar OZ axis of the SF shown in Fig. 1.

#### A. Density matrix

It has already been mentioned in this paper that the expression (3) for the DM is not only applicable to any one-step photoabsorption process, but also completely general (i.e., independent of all dynamical models, or angular momentum coupling schemes, etc.). It can therefore be readily used for calculating the required DM even in the present case of  $j$ - $j$  coupling when the photoionization process (1) is taking place in the presence of SOI, in addition to the Coulomb forces experienced by the constituents of an atom [i.e., Hamiltonians (A5) and (A6b) are used].

Consequently, the DM for the process (1) in an unpolarized atom  $A$  in  $j$ - $j$  coupling becomes

$$\langle J_{1+} M_{1+}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | J_{1+} M'_{1+}; \mu'_p \hat{u}_p \vec{k}_p \rangle = \frac{\mathcal{K}_p}{2J_0 + 1} \sum_{M_0} \langle 1^+; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1 m_r \rangle \langle 0; 1 m_r | F_p^\dagger | J_{1+} M'_{1+}; \mu'_p \hat{u}_p \vec{k}_p \rangle. \quad (20)$$

In order to proceed further, we need to know the photoionization matrix element present on the right-hand side of (20) such that it meets both of the conservation requirements (2) and (19). It can be shown to be given by [73]

$$\begin{aligned} \langle 1^+; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1 m_r \rangle &= (-1)^{-\frac{1}{2}} \sum_{\ell_p m_{\ell_p} j_p m_{j_p} \nu_p} (-i)^{\ell_p} (-1)^{\ell_p + m_{j_p}} e^{i\sigma_{\ell_p j_p}} \sqrt{2j_p + 1} \begin{pmatrix} \ell_p & \frac{1}{2} & j_p \\ m_{\ell_p} & \nu_p & -m_{j_p} \end{pmatrix} \\ &\times Y_{\ell_p}^{m_{\ell_p}}(\hat{k}_p) \mathcal{D}_{\mu_p \nu_p}^{\frac{1}{2}}(\omega_p) \langle J_{1+} M_{J_{1+}}; j_p m_{j_p} | F_p | J_0 M_{J_0}; 1 m_r \rangle^-. \end{aligned} \quad (21a)$$

The ‘‘minus’’ superscript on the Dirac’s bracket on the right-hand side of (21a) means that the continuum orbital of the photoelectron in  $j$ - $j$  coupling satisfies the incoming wave boundary conditions [74] appropriate for a photoionization process. Further, in Eq. (21a), we have defined [73]

$$\begin{aligned} \langle J_{1+} M_{J_{1+}}; j_p m_{j_p} | F_p | J_0 M_{J_0}; 1 m_r \rangle^- &\equiv (-1)^{1 - J_0 - J_{1+} + j_p} \sum_{J M_J} (-1)^{2M_J} \\ &\times (2J + 1) \begin{pmatrix} J_0 & 1 & J \\ M_0 & m_r & -M_J \end{pmatrix} \begin{pmatrix} J_{1+} & j_p & J \\ M_{J_{1+}} & m_{j_p} & -M_J \end{pmatrix} \langle J_{1+} j_p | F(J) | J_0 1 \rangle^- \end{aligned} \quad (21b)$$

and

$$\langle (J_{1+} j_p) J M_J | F_p | (J_0 1) J' M'_J \rangle^- = \delta_{(J J')} \delta_{(M_J M'_J)} \langle J_{1+} j_p | F(J) | J_0 1 \rangle^- \quad (21c)$$

for satisfying the angular momentum conservation condition (19).



Next, we substitute (21a) and its Hermitian conjugate in Eq. (20) and simplify the resulting expression by analytically evaluating as many sums as possible using Racah algebra. In its fully simplified form, the DM can finally be written as

$$\begin{aligned} & \langle J_{1+} M_{1+}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | J_{1+} M'_{1+}; \mu'_p \hat{u}_p \vec{k}_p \rangle \\ &= (-1)^{\mu'_p + M_{1+}} \sum_{J_T M_{J_p} S_p M_{S_p} N_{S_p}} \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & S_p \\ \mu_p & -\mu'_p & M_{S_p} \end{pmatrix} \begin{pmatrix} J_{1+} & J_{1+} & J_T \\ M_{1+} & -M'_{1+} & M_{J_p} \end{pmatrix} \left[ \mathcal{D}_{M_{S_p} N_{S_p}}^{S_p}(\omega_p) \right]^* A(S_p N_{S_p}; J_T M_{J_p}; m_r; \vec{k}_p), \end{aligned} \quad (22a)$$

where

$$\begin{aligned} A(S_p N_{S_p}; J_T M_{J_p}; m_r; \vec{k}_p) &= (-1)^{\frac{1}{2} + m_r + J_0 - J_{1+} + J_T} (2J_T + 1)(2S_p + 1) \frac{\mathcal{K}_p}{(2J_0 + 1)\sqrt{4\pi}} \sum_{\ell_p j_p J} \sum_{L_p M_{L_p}} (-1)^{\ell'_p + j_p + j'_p + J + L_r} \\ & \quad \sum_{\ell'_p j'_p J'} \sum_{J_p L_r} \\ & \quad \times (2L_r + 1)(2J_p + 1)\sqrt{2L_p + 1} \begin{pmatrix} \ell_p & \ell'_p & L_p \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & 1 & L_r \\ m_r & -m_r & 0 \end{pmatrix} \begin{pmatrix} L_p & S_p & J_p \\ M_{L_p} & N_{S_p} & M_{J_p} \end{pmatrix} \\ & \quad \times \begin{pmatrix} J_p & L_r & J_T \\ M_{J_p} & 0 & -M_{J_p} \end{pmatrix} \begin{Bmatrix} 1 & 1 & L_r \\ J & J' & J_0 \end{Bmatrix} \begin{Bmatrix} \ell_p & \ell'_p & L_p \\ \frac{1}{2} & \frac{1}{2} & S_p \\ j_p & j'_p & J_p \end{Bmatrix} \begin{Bmatrix} j_p & j'_p & J_p \\ J & J' & L_r \\ J_{1+} & J_{1+} & J_T \end{Bmatrix} \left[ Y_{M_{L_p}}^{L_p}(\hat{k}_p) \right]^* \\ & \quad \times [(-i)^{\ell_p} (-1)^{\ell_p + j_p} e^{i\sigma_{\ell_p j_p}} (2J + 1)\sqrt{(2\ell_p + 1)(2j_p + 1)} \langle J_{1+} j_p | F(J) | J_0 1 \rangle^{-1}] \\ & \quad \times [(-i)^{\ell'_p} (-1)^{\ell'_p + j'_p} e^{i\sigma_{\ell'_p j'_p}} (2J' + 1)\sqrt{(2\ell'_p + 1)(2j'_p + 1)} \langle J_{1+} j'_p | F(J') | J_0 1 \rangle^{-1}]^* \end{aligned} \quad (22b)$$

with amplitudes  $\langle J_{1+} j_p | F(J) | J_0 1 \rangle$  given in Eq. (21b).

### 1. Properties of the density matrix

Let us briefly analyze the properties of the present DM (22), obtained in  $j$ - $j$  coupling, *vis a vis* the one derived earlier in  $L$ - $S$  coupling and given in Eqs. (B2)–(B4). Although both of these two density matrices represent the residual photoion  $A^{1+}$  with angle- and spin-resolved photoelectron  $e_p$  generated in the same process (1) of photoionization, however, their structures and levels of complexity are quite different from each other. The reason for these differences is, obviously, the presence of SOI in the derivation of (22) which, otherwise, is absent in the Coulombic DM (B2) derived in Appendix B and discussed in Sec. III.

The FS levels [52,53] of the atomic target  $A$  and of the residual photoion  $A^{1+}$  are known to be determined by their respective total angular momenta  $\vec{J}_0$  and  $\vec{J}_{1+}$ . Here, too, we find that any properties of a  $(e_p, A^{1+})$  system, calculated from (22), will depend, among other things, upon  $(\vec{J}_0, \vec{J}_{1+})$ . It will, therefore, be quite appropriate to call (22) as fine-structure DM. Any physical property calculated using this DM will also be prefixed by the word FS, e.g., FS entanglement, etc.

Unlike the Coulombic DM (B2), the FS DM (22) can not be written as a product of two terms representing angular correlation and spin correlation. In the expression (22), on the other hand, angular and spin terms are intermixed in a way that they can not be separated. It is a consequence of the presence of the SOI in the Hamiltonians (A5) and (A6b). Moreover, unlike the earlier case, now the entanglement between the spin of electronic qubit and the total angular momentum  $J_{1+}$  of

the ionic qudit depends upon many things, e.g., kinematics (i.e., directions of motion  $\vec{k}_p$  and of spin quantization  $\hat{u}_p$  of photoelectron), dynamics [i.e., phases and amplitudes of the photoionization matrix elements (21)], characteristics (i.e., frequency  $\nu_r$  and polarization  $m_r$ ) of the ionizing radiation  $\gamma_r$ , orbital angular momentum  $\ell_p$  of  $e_p$ , etc., in addition to the total angular momenta  $(J_0, J_{1+})$ . Thus, the inclusion of SOI in the photoionization process (1) has resulted in coupling the  $(e_p, A^{1+})$  entanglement with many physical variables, including both the kinetics and dynamics of this process.

This property of the fine-structure entanglement is seen even in the other photon-induced processes studied elsewhere (see, for example, [49,50]). Consequentially, unlike the Coulombic entanglement, it is not possible to predict or predetermine the properties of the FS entanglement merely with a knowledge of the electronic states  $(|J_0 M_{J_0}\rangle, |J_{1+} M_{J_{1+}}\rangle)$  of  $(A, A^{1+})$  participating in the photoionization (1) in the  $j$ - $j$  coupling. One is now required to have dynamical parameters in order to be able to calculate the properties of the FS entanglement using the DM (22).

The dimensionality of the Hilbert space of the photoion  $A^{1+}$ , which will be entangled with the electronic qubit  $e_p$ , is determined by its total angular momentum  $J_{1+}$  and is equal to  $(2J_{1+} + 1)$ . Thus, one of the simplest possible single-step process, namely, photoionization, is capable of realistically generating in a laboratory qubit-qubit (for  $J_{1+} = \frac{1}{2}$ ), qubit-qutrit (for  $J_{1+} = 1$ ), and qubit-qudit (for  $J_{1+} \geq \frac{3}{2}$ ) pairs of flying (electron + ion), both in the presence or absence of SOI.

Furthermore, the  $2(2J_{1+} + 1)$  diagonal elements (given by  $\mu'_p = \mu_p$  and  $M'_{1+} = M_{1+}$ ) of the FS DM (22) are the

cross sections for angle- and spin-resolved photoelectron spectroscopy in the  $E1$  approximation with the photoion observed in one of its  $|J_{1+} M_{1+}\rangle$  electronic state in the  $j$ - $j$  coupling. The remaining nondiagonal elements (with  $\mu_p' \neq \mu_p$  or  $M_{1+}' \neq M_{1+}$ ) in Eq. (22) represent, on the other hand, the coherence effects in the photoionization (1) in the  $j$ - $j$  coupling.

The  $(2 \times 2)$  reduced matrix

$$\begin{aligned} & \langle \mu_p \hat{u}_p \vec{k}_p | \rho^{(e_p)} | \mu_p' \hat{u}_p \vec{k}_p \rangle \\ &= \sum_{M_{1+}=-J_{1+}}^{J_{1+}} \langle J_{1+} M_{1+}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | J_{1+} M_{1+}; \mu_p' \hat{u}_p \vec{k}_p \rangle \end{aligned} \quad (23)$$

describes angle- and spin-resolved photoelectron  $e_p$  emitted in photoionization (1) in the  $j$ - $j$  coupling. Its two diagonal terms are the angular distribution of photoelectrons moving in the direction  $\vec{k}_p$  with their spins having components  $\mu_p = \pm \frac{1}{2}$  along  $\hat{u}_p$ . The expression for the diagonal elements in Eq. (23) can readily be shown to be identical to that already given in Ref. [75] in terms of the total angular momentum  $J [= |\vec{J}|]$  with  $\vec{J} = \vec{\ell}_r + \vec{J}_0 = \vec{J}_{1+} + \vec{j}_p$ , see Eq. (19)], or equivalent to that derived by Chandra [73] in terms of the angular momentum  $\vec{j}_i \equiv \vec{\ell}_r - \vec{j}_p = \vec{J}_{1+} - \vec{J}_0$  transferred from absorbed photon  $\gamma_r$  to the photoelectron  $e_p$  observed with its spin.

On reducing the FS DM (23) over the variables related to the photoelectron  $e_p$ , we obtain

$$\begin{aligned} & \langle J_{1+} M_{1+} | \rho^{1+} | J_{1+} M_{1+}' \rangle \\ &= \int \left( \sum_{\mu_p=-\frac{1}{2}}^{+\frac{1}{2}} \langle J_{1+} M_{1+}; \mu_p \hat{u}_p \vec{k}_p | \rho_f \right. \\ & \quad \left. | J_{1+} M_{1+}'; \mu_p' \hat{u}_p \vec{k}_p \rangle \right) d\hat{k}_p. \end{aligned} \quad (24)$$

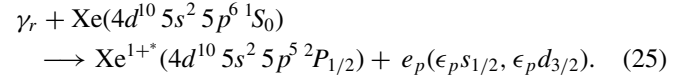
This matrix is of size  $[(2J_{1+} + 1) \times (2J_{1+} + 1)]$ . It can be used to calculate *state multipoles* (also called *statistical tensors*) [44–46] for  $A^{1+}$  in the  $j$ - $j$  coupling. These state multipoles describe [44–46], among other things, orientation and alignment of the photoion  $A^{1+}$ .

Earlier, Radtke *et al.* also have derived expressions for the density matrices for the process (1) taking relativistic and nondipolar effects [76] plus electron-electron interaction [77] into account. Unlike the presently obtained DM (22), the expressions obtained in Refs. [76,77] are for an experimental geometry wherein the polar axis is along the direction of propagation of the photoelectron  $e_p$  (i.e., OZ axis in Fig. 1 is taken along  $\hat{k}_p$  by Radtke *et al.* [76,77]) and the photoelectron therein is longitudinally polarized (i.e.,  $\hat{u}_p \parallel \hat{k}_p$ ), with SOI always taken into account. Recent studies (e.g., [78]) have, however, shown that the nonrelativistic description of the process (1) for as heavy an atom as Xe is both sufficient and accurate. Moreover, the presence of nondipolar terms, which are of second order, in the photoionization operator  $F_p$  in Eq. (3a) [specified in Eqs. (A7) and (A9) in the  $E1$  approximation] is not expected to have any significant effects on the entanglement properties of a  $(e_p, A^{1+})$  pair.

Prior to Radtke *et al.* [76,77], Fedorov *et al.* [79] had investigated narrowing of the wave packets of the photoelectron  $e_p$  and of the residual photoion  $A^{1+}$  in a coincident observation of these two particles in the photoionization process (1). They [79] argued that this type of localization of two wave packets is directly related to the  $(e_p, A^{1+})$  entanglement and, hence, to the Einstein-Podolsky-Rosen [80] localization.

## B. Example

For an application of the DM (22) and to compare the properties of the entanglement generated in a  $(e_p, A^{1+})$  pair with and without the SOI, let us consider the following photoionization process in a Xe atom:



Angle- and spin-resolved photoelectron spectroscopy of this process has been studied both theoretically [81] and experimentally [82].

As far as Coulombic entanglement for the process (25) is concerned, it corresponds to  $(S_0 = 0, S_{1+} = 1/2)$ . According to the discussion given in Sec. III A, spin angular momenta of  $(e_p, A^{1+})$ , in this case, are maximally entangled in the pure singlet state of two qubits in the absence of SOI. This Coulombic entanglement between the spins of  $(e_p, A^{1+})$  is completely independent of all those physical variables (e.g., those related to the characterization of  $\gamma_r$ , kinematics of  $e_p$ , dynamics of photoionization, etc.), other than  $(S_0, S_{1+})$ , which one may need to specify the photoionization process (25) in the absence of SOI.

On the inclusion of SOI, the photoion  $A^{1+}$  has total angular momentum  $J_{1+} = \frac{1}{2}$  in the process (25). Consequently, even in the case of FS entanglement,  $(e_p, A^{1+})$  form a bipartite state of two qubits. Further, entanglement is well known [1–3,20] to be independent of the direction of spin quantization. For simplicity, we have therefore taken the photoelectron  $e_p$  to be polarized longitudinally along its direction of propagation, i.e.,

$$\hat{u}_p \parallel \hat{k}_p \implies \vartheta_p = \theta_p \quad \text{and} \quad \varphi_p = \phi_p \quad (26)$$

in our present calculation.

In order to study FS entanglement of  $(e_p, A^{1+})$  for the example (25), we need to know magnitudes (i.e.,  $d_1, d_3$ ) and the phases (i.e.,  $\delta_1, \delta_3$ ) of both of the  $E1$  amplitudes

$$D_1 \equiv \langle J_{1+} = \frac{1}{2}, j_p = \frac{1}{2} | F(J=1) | J_0 = 0, 1 \rangle = d_1 e^{i\delta_1} \quad (27a)$$

and

$$D_3 \equiv \langle J_{1+} = \frac{1}{2}, j_p = \frac{3}{2} | F(J=1) | J_0 = 0, 1 \rangle = d_3 e^{i(\delta_1 - \Delta)} \quad (27b)$$

with

$$\Delta \equiv \delta_1 - \delta_3 \quad (27c)$$

contributing to the FS DM (22) through (21c). These quantities were extracted from the data given in Refs. [81,82] on the process (25) as follows.

Angular distribution of spin-resolved photoelectrons emitted in any  $E1$  process (1) can be completely characterized by five parameters [75,81,82], namely,  $(\sigma, \beta, \xi, \eta, \zeta)$ . We have derived analytical expressions for each of these five parameters from our reduced matrix (23). On specializing these expressions to the process (25), we could exactly reproduce those given in Refs. [81,82] provided we replace  $\Delta$ , wherever it is present in our expressions for any of the parameters  $(\sigma, \beta, \xi, \eta, \zeta)$ , by  $\pi - \Delta$ .

The three dynamical parameters (i.e.,  $d_1, d_3, \Delta$ ), specified in Eqs. (27), were extracted for 10 different values of energy  $E_r$  of the ionizing photon  $\gamma_r$  from the graphs for  $(\sigma, \beta, \xi, \eta, \zeta)$  given as a function of  $E_r$  in Ref. [81]. These dynamical parameters were then used in the DM (22) for calculating all 16 elements for each of the 4 polarizations of the absorbed photon  $\gamma_r$ : linear ( $m_r = 0$ ), left circular ( $m_r = -1$ ), right circular ( $m_r = +1$ ), and unpolarized (i.e., an even mixture of  $m_r = \pm 1$ ). In this way, 40 different density matrices (22) (10 for each of these 4 different kinds of polarization of  $\gamma_r$ ) were calculated. These density matrices were found to have the following features:

(a) Although, for the experimental geometry (26), each element of these matrices, as well as of their partial transpose, involves both of the spherical angles specifying the direction of propagation  $\vec{k}_p$  ( $\theta_p, \phi_p$ ) of the photoelectron  $e_p$ , however, their eigenvalues were always independent of the azimuthal angle  $\phi_p$  and, hence, contained only the polar angle  $\theta_p$ . The trace of this DM was normalized to unity by dividing it, wherever necessary, by the sum of these eigenvalues.

(b) For all  $\vec{k}_p$ 's considered by us, each DM (their total number being 30) always has 3 of its 4 eigenvalues 0 for any of the LP, RCP, or LCP  $\gamma_r$ ; whereas, the remaining 10 density matrices, corresponding to an UP  $\gamma_r$ , have more than 1 nonzero eigenvalue. This, in other words, means [45] that  $(e_p, A^{1+})$  FS states [generated due to the absorption of a LP, RCP, or RCP  $\gamma_r$  in Eq. (25) with specifications (26) and (27)] are always pure, while absorption of an UP  $\gamma_r$  in the same process produces a mixed state of this system. According to (a) herein, all nonzero eigenvalues of the 40 density matrices, as well as of their partial transpose, depend only on the polar angle  $\theta_p$ .

(c) For each of these 40 FS states, we have calculated negativity [60–62]  $\mathcal{N}(\rho_f)$ , concurrence [63]  $\mathcal{C}(\rho_f)$ , and EoF [63]  $\mathcal{E}(\rho_f)$ , the three measures of entanglement currently in vogue. It has theoretically been shown that for a pure bipartite state of qubits, negativity and concurrence are always identical [62]. Our results for ionization in Eq. (25) by a LP, RCP, or LCP  $\gamma_r$  were always in agreement with this prediction. Although, negativity and concurrence were different when  $\gamma_r$  in Eq. (25) is UP because these are now for a mixed FS state of an entangled  $(e_p, A^{1+})$  system; however, on the scale of the Figs. 2–4, these two measures for ionization by an UP photon are almost identical. Consequently, in Figs. 2–4, the concurrence curve also represents the negativity for each polarization of  $\gamma_r$  used by us.

(d) It is obvious that, on account of the second 3- $j$  symbol present in Eq. (22b), a DM for RCP (i.e.,  $m_r = +1$ ) ionizing radiation should be different from that for LCP (i.e.  $m_r = -1$ ) for the same  $E_r$ . Our calculations showed that the eigenvalues of these two different density matrices were always the

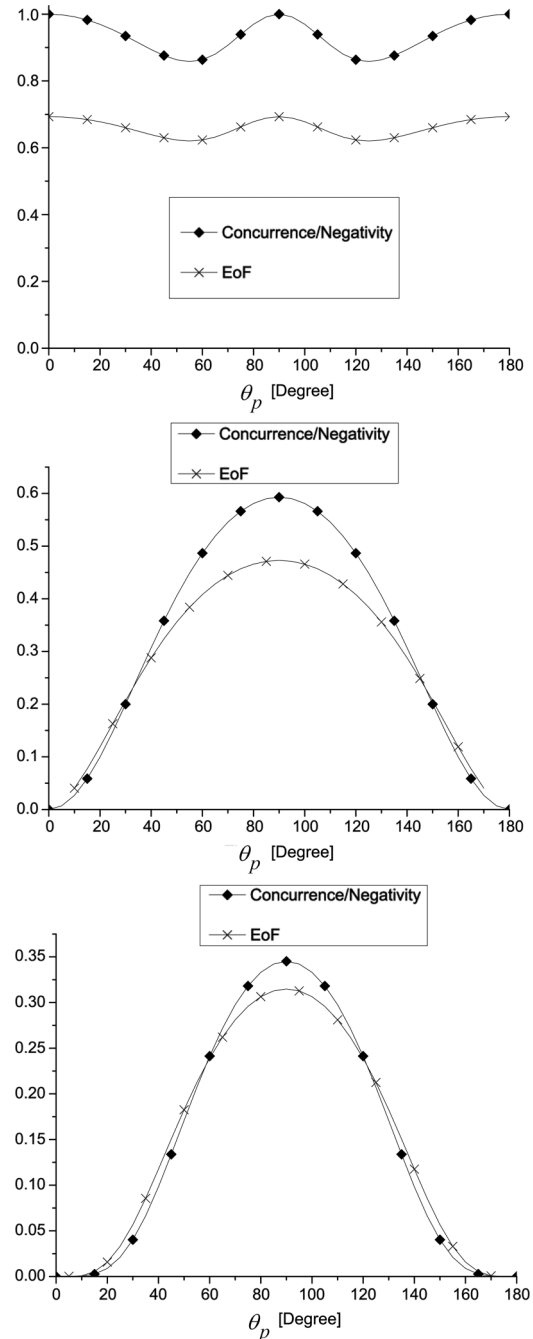


FIG. 2. Variations with the polar angle  $\theta_p$  [defined in Eq. (26)] of the concurrence and EoF for FS entanglement between spin of photoelectron  $e_p$  and angular momentum  $J_{1+}$  of the residual photoion  $A^{1+}$  generated in the  $E1$  process (25). See point (c) in the present Sec. IV B. These measures have been calculated for energy  $E_r = 0.5$  a.u. of the ionizing radiation  $\gamma_r$ , and the experimental geometry (26). The dynamical amplitudes (27) are extracted from the data given in Ref. [81]. Use of LCP (i.e.,  $m_r = -1$ )  $\gamma_r$  in Eq. (25) gave the values of negativity and concurrence and of EoF identical to those obtained when a RCP ( $m_r = +1$ )  $\gamma_r$  is used for ionizing a Xe atom. This, in other words, means that there is no circular dichroism [83] in the FS  $(e_p, A^{1+})$  entanglement, generated in the process (25). (a) Ionizing radiation in Eq. (25) LP [i.e.,  $m_r = 0$  in the FS DM (22)]; (b) ionizing radiation in Eq. (25) left and right CP [i.e.,  $m_r = +1$  or  $-1$  in the FS DM (22)]; (c) ionizing radiation in Eq. (25) UP.

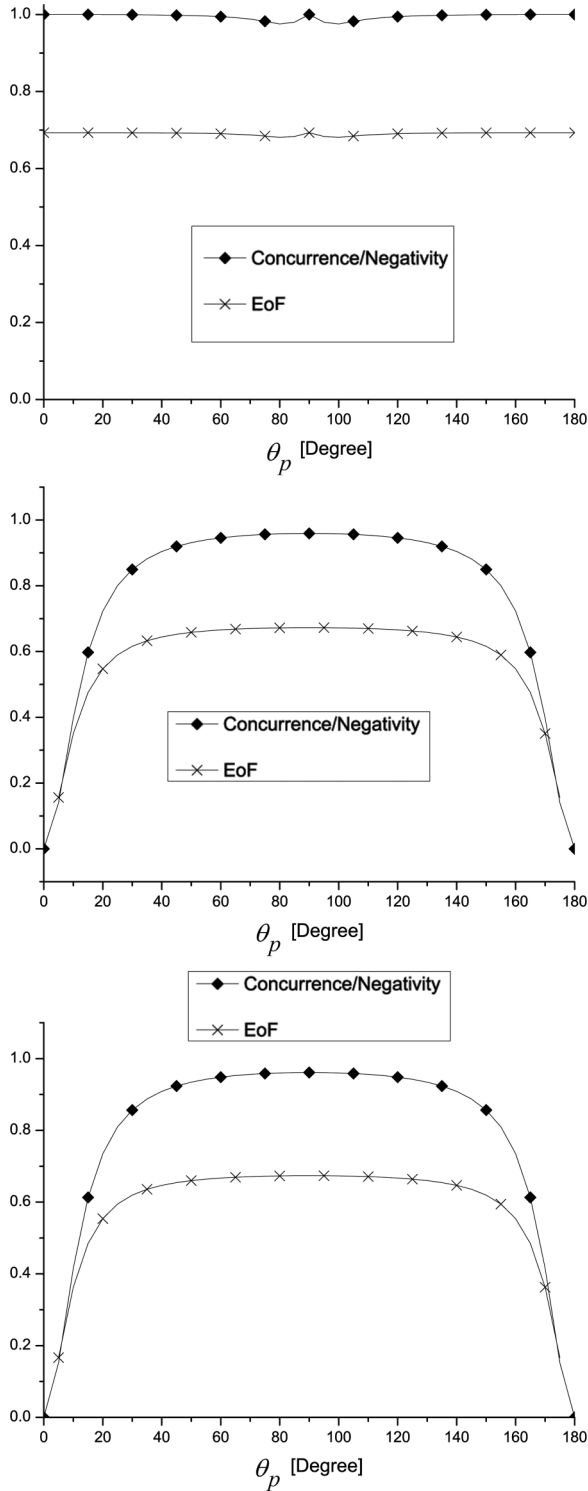


FIG. 3. Same as Fig. 2, but for energy  $E_r = 1.0$  a.u. of the ionizing radiation  $\gamma_r$  in Eq. (25).

same. Also, each of the three entanglement measures for a left circularly polarized  $\gamma_r$  [i.e.,  $m_r = -1$  in Eq. (22)] was always identical to that when the ionizing radiation in Eq. (25) has right circular polarization [i.e.,  $m_r = +1$  in Eq. (22)]. Thus, in the present case, none of the three measures of entanglement calculated by us shows any circular dichroism (see, for example, [83]) in the process (25) with

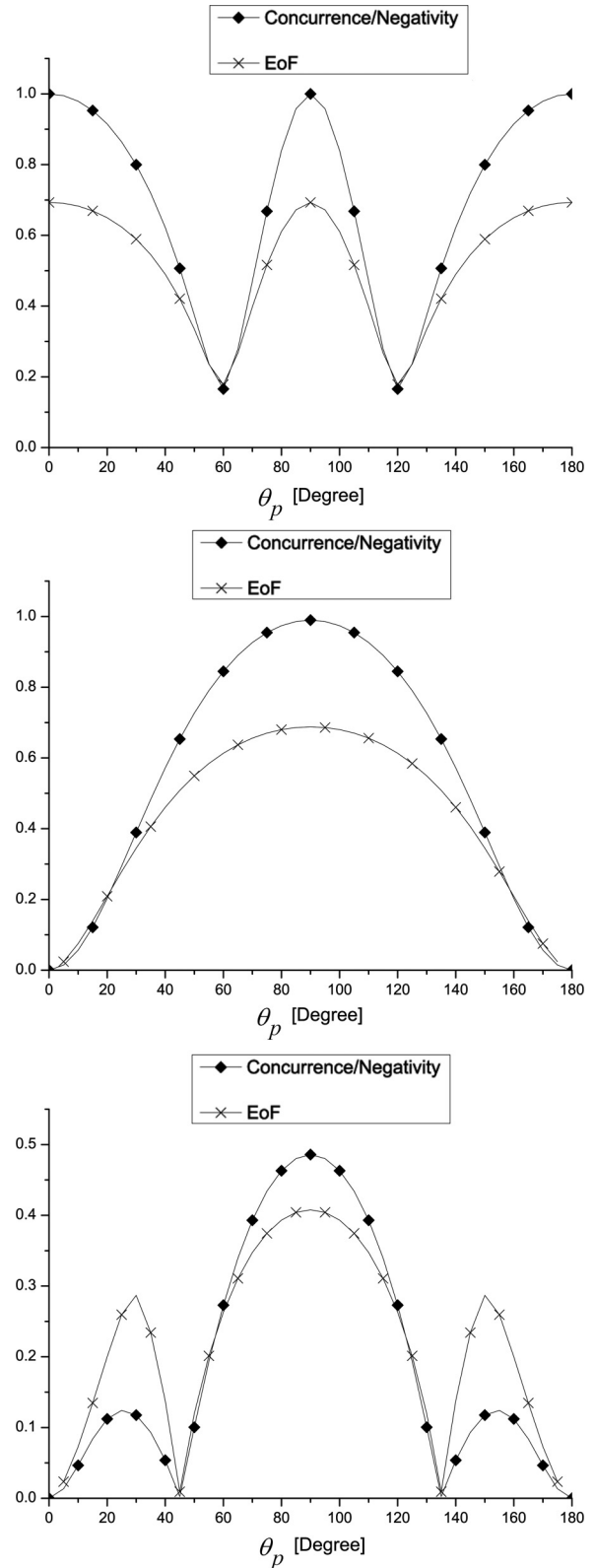


FIG. 4. Same as Fig. 2, but for energy  $E_r = 3.0$  a.u. of the ionizing radiation  $\gamma_r$  in Eq. (25).

specifications [(26) and (27)]. Figures 2–4, therefore, contain concurrence and EoF only for ionization in Eq. (25) by linearly polarized, (right or left) circularly polarized, and unpolarized radiation.



(e) Both concurrence and negativity and EoF for photon energy  $E_r = 0.50$  a.u. are shown together in each of the Figs. 2(a)–2(c) for ionization by  $m_r = 0, +1/-1$ , and unpolarized light, respectively. Figures 3(a)–3(c) and 4(a)–4(c) contain the same information for  $E_r = 1.0$  and 3.0 a.u., respectively.

(f) One may wonder that if an UP electromagnetic wave is an even mixture of RCP plus LCP radiation, then why are the measures of entanglement shown in Figs. 2–4 for UP and CP  $\gamma_r$  in Eq. (25) not always the same? It has already been mentioned in the item (d) herein that the density matrices, obtained from (22) for LCP and RCP light, are different. Consequently, a DM for UP light, obtained on dividing by two the sum of these two different density matrices, need not be equal to either of them. Consequently, measures of entanglement shown in Figs. 2–4 for UP and CP ionizing radiation in Eq. (25) may not always be the same.

(g) Figures 2–4 clearly show the dependence of the FS entanglement between spin angular momentum of the photoelectron  $e_p$  and total angular momentum  $J_{1+}$  of the photoion  $A^{1+}$  [both particles produced in the 1–SPI process (25)] on the energy  $E_r$  and polarization of  $\gamma_r$ , direction of motion of  $e_p$ , and on the dynamics of the process (25). In particular, a heavy dependence on polarization as well as energy of the photon absorbed in the process (25), and on the direction of motion of the photoelectron  $e_p$ , is very obvious from Figs. 2 ( $E_r = 0.5$  a.u.) and 3 ( $E_r = 1.0$  a.u.) wherein the entanglement measures for LP light are very different from those for CP or UP radiation, except in Fig. 4 ( $E_r = 3.0$  a.u.), where one finds that variations with  $\theta_p$  for LP [Fig. 4(a)] and for UP [Fig. 4(c)]  $\gamma_r$  are more prominent than those shown in Figs. 2 and 3, although variations with  $\theta_p$  for CP ionizing radiation in Eq. (25) do not change much for the three energies  $E_r$  considered herein. Obviously, this behavior of the entanglement measures shown in Figs. 2–4 is due to a very complex dependence of the FS DM (22) on the characteristics of the  $\gamma_r$  absorbed in Eq. (25) as well as on both the kinematics and dynamics of this process in the presence of SOI.

(h) Thus, the presence of SOI in a photoionization process (1) strongly couples the entanglement between the emitted photoelectron and residual photoion with the characteristics of the ionizing radiation, experimental geometry, and dynamics of this process. It is very unlike the Coulombic entanglement generated between the same two particles in the same process of photoionization but in the absence of SOI.

## V. CONCLUSIONS

Photoelectric effect is an extremely simple phenomena in science: its theoretical explanation by Einstein [84] has had profound effects on the development of science in general and of physics in particular. It has certainly played a very important role in the development of quantum theory as it led to suggest the wave-particle duality first of light and, subsequently, of matter. In this paper, we have tried to show that the same photoelectric effect can also be a very powerful tool (or, in other words, an entanglement factory) for producing various kinds of entangled states of a free electron and a free ion. Here, one can realistically generate in a very simple manner in a laboratory qubit-qudit bipartite states which can be pure or mixed, possess free or bound entanglement.

From the DMs [(7) and (22)] derived herein, one can theoretically determine properties of the (Coulombic, FS) entanglement of a ( $e_p, A^{1+}$ ) pair generated in the 1–SPI process (1). In the FS entanglement, one necessarily requires dynamical amplitudes for the photoionization process (1). But, properties of the Coulombic entanglement can readily be predicted merely with a knowledge of the spins ( $S_0, S_{1+}$ ) of ( $A, A^{1+}$ ) which can experimentally be obtained simply by measuring energy of the  $e_p$  in a photoionization experiment in the  $L$ - $S$  coupling. However, verification of the properties of either or both of the Coulombic and FS entanglements, obtained from the theoretical procedures described in this paper, will certainly require experimental measurements. It can be done using an entanglement witness [18], or any other such protocols [19], or by performing a full quantum-state tomography involving both the photoelectron  $e_p$  and the photoion  $A^{1+}$ .

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## APPENDIX A: DERIVATION OF EQ. (3a) FOR DENSITY OPERATOR FOR A PHOTON-INDUCED ONE-STEP PROCESS

Equation (3a) for the density operator for any one-step process is readily derived [45] using the concept of scattering operator ( $S$  operator) or scattering matrix ( $S$  matrix) from quantum theory of scattering (QTS). In QTS (see, for example, [85,86]), the  $S$  operator and  $S$  matrix connect the initial state (say,  $|\Psi_1\rangle$ ) of the combined system of a projectile and a target with the final state (say,  $|\Psi_f\rangle$ ), formed after the reaction between the two has taken place, of the particles receding from each other. Both  $|\Psi_1\rangle$  and  $|\Psi_f\rangle$  describe the noninteracting states, i.e., separated by infinite distances, of their respective members. Then, one has [45,85,86]

$$|\Psi_f\rangle = S |\Psi_1\rangle. \quad (\text{A1})$$

This immediately leads us to

$$\tilde{\rho}_f = S \rho_1 S^\dagger \quad (\text{A2a})$$

with the definitions

$$\rho_1 \equiv |\Psi_1\rangle\langle\Psi_1| \quad (\text{A2b})$$

and

$$\tilde{\rho}_f \equiv |\Psi_f\rangle\langle\Psi_f| \quad (\text{A2c})$$

for the two respective density operators appearing in Eq. (A2).

The diagonal elements of the  $S$  matrix include also those contributions for which  $|\Psi_1\rangle$  and  $|\Psi_f\rangle$  are not different. In order to remove such contributions, one defines [45,85,86] a

transition operator

$$T = S - 1. \quad (\text{A3})$$

Consequently, the density operator [45]

$$\rho_f = T \rho_1 T^\dagger \quad (\text{A4})$$

provides information only about those reaction products which are different from the ones represented by  $\rho_1$ . Equation (A4) is applicable to all those processes which are completed in a single step, e.g., those explained in Sec. II. It can readily be generalized to two-step (see, for example, [87,88]), three-step (e.g., [89]), and other multistep processes. The form of the transition operator  $T$  will, naturally, be determined by the kind of the reaction being considered, which may be different for different steps in a multistep process, giving rise to the desired products represented by the final-state DO  $\rho_f$ .

In the present one-step process (1), the final reaction products ( $e_p, A^{1+}$ ) are generated due to the absorption of a single photon  $\gamma_r$  by the atom  $A$ . The semiclassical Hamiltonian for this system is given by (see, for example, [47,54,55,85])

$$H = H_0 + \mathcal{F}. \quad (\text{A5})$$

Here,

$$H_0 = -\frac{\hbar^2}{2m_e} \sum_{i=1}^{n_e} \nabla_i^2 - \sum_{i=1}^{n_e} \frac{Ze^2}{r_i} + \sum_{i<j}^{n_e} \frac{e^2}{|\vec{r}_i - \vec{r}_j|} \quad (\text{A6a})$$

and

$$H_0 = -\frac{\hbar^2}{2m_e} \sum_{i=1}^{n_e} \nabla_i^2 - \sum_{i=1}^{n_e} \frac{Ze^2}{r_i} + \sum_{i<j}^{n_e} \frac{e^2}{|\vec{r}_i - \vec{r}_j|} + \sum_{i=1}^{n_e} \xi(r_i) \vec{\ell}_i \cdot \vec{s}_i \quad (\text{A6b})$$

are the Coulombic (i.e., without SOI) and FS (i.e., with SOI) forms of the Hamiltonian of the target atom  $A$ , respectively. In Eqs. (A6), symbols, not explained herein, have their usual (see, e.g. [52,53]) meanings. The operators

$$\mathcal{F} = \sqrt{\frac{4\pi\alpha^3 E_r^3}{3e^4}} \sum_{i=1}^{n_e} \hat{\xi}_{m_r} \cdot \vec{r}_i \quad (\text{A7a})$$

and

$$\mathcal{F} = \sqrt{\frac{4\pi\alpha^3 E_r a_0^2}{3}} \sum_{i=1}^{n_e} \hat{\xi}_{m_r} \cdot \vec{\nabla}_i, \quad (\text{A7b})$$

present on the right-hand side of the Hamiltonian (A5), represent the interaction of the atomic electrons (their number being  $n_e$ ) with the incident electromagnetic radiation in the  $E1$  length and velocity approximations, respectively. In Eqs. (A7),  $\vec{r}_i$  is the position vector and  $\vec{\nabla}_i [ = (-1)^{1/2} \vec{p}_i / \hbar ]$  is the linear momentum of the  $i$ th bound atomic electron, and  $\alpha$  and  $a_0$  are, respectively, the FS constant and Bohr radius. Further, in the last two equations,  $\hat{\xi}_{m_r}$  is a spherical unit vector [57] which specifies polarization of the electromagnetic wave incident in

the process (1). Then,

$$\begin{aligned} \hat{\xi}_{-1} &= +\frac{1}{\sqrt{2}}(\hat{e}_x - i\hat{e}_y), \quad \hat{\xi}_0 = \hat{e}_z, \quad \text{and} \\ \hat{\xi}_{+1} &= -\frac{1}{\sqrt{2}}(\hat{e}_x + i\hat{e}_y) \end{aligned} \quad (\text{A8})$$

for the respective LCP, LP, and RCP radiations. Here, ( $\hat{e}_x, \hat{e}_y, \hat{e}_z$ ) are unit vectors along the ( $OX, OY, OZ$ ) axes of the SF shown in Fig. 1.

The transition operator  $T$ , needed in Eq. (A4) for the photoionization process (1), is given by [47,55]

$$T = \sqrt{\mathcal{K}_p} F_p \quad (\text{A9a})$$

with

$$F_p = \sqrt{\frac{m_e}{\hbar^2}} \mathcal{F} \quad (\text{A9b})$$

and

$$\mathcal{K}_p = 3\pi \left( \frac{e^2}{\alpha E_r} \right)^2. \quad (\text{A9c})$$

Use of these relations gives [54]

$$\frac{d\sigma}{d\Omega} = |\langle \Psi_f | T | \Psi_1 \rangle|^2 \quad (\text{A10a})$$

$$= \mathcal{K}_p |\langle \Psi_f | F_p | \Psi_1 \rangle|^2 \quad (\text{A10b})$$

angular distribution of photoelectrons ejected into the solid angle  $d\Omega$  about their direction of propagation in a bound-free transition  $\Psi_1 \rightarrow \Psi_f$ . Here,  $\Psi_f$ , which represents photoelectron  $e_p$  moving in the field of  $A^{1+}$  after leaving  $A$ , satisfies incoming wave boundary conditions [74] appropriate for any photoionization process. Furthermore [54,55],  $\Psi_f$  has normalization (see, for example, Eq. (17) in Ref. [55]) corresponding to the unit density of states in the continuum of  $e_p$  and orthonormality of the bound states of the photoion  $A^{1+}$ .

## APPENDIX B: DENSITY MATRIX FOR ONE-STEP SINGLE PHOTOIONIZATION OF AN ATOM IN THE ABSENCE OF SPIN-ORBIT INTERACTION

This Appendix contains a derivation of the DM needed in Sec. III for studying the Coulombic entanglement between the spins of photoelectron  $e_p$  and photoion  $A^{1+}$  produced in the single-step process (1) resulting from the absorption of a photon  $\gamma_r$  by the target atom  $A$ . The following derivation, although independent of all dynamical models, does not take SOI into account either in the bound electronic states ( $|0\rangle, |1^+\rangle$ ) of ( $A, A^{1+}$ ) or in the continuum of  $e_p$ . It, in other words, means that only the Coulomb interactions plus, of course, the interaction with the incident light in the  $E1$  approximation [both specified by the Hamiltonians (A5) and (A6a)] experienced by the constituents of an atom, are included in the present derivation and, hence, it is carried out in the  $L$ - $S$  coupling wherein both of the conservation conditions (4) become applicable.

It is obvious from Eq. (6b) that derivation of the needed DM requires a knowledge of the photoionization

amplitude  $\langle 1^+; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1 m_r \rangle$  calculated in the  $L$ - $S$  coupling. Details of this calculation are already given

in Eqs. (A7)–(A9) in Ref. [87]. The final desired expression [87] is

$$\begin{aligned}
 \langle 1^+; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1 m_r \rangle &\equiv \langle L_{1^+} S_{1^+} M_{L_{1^+}} M_{S_{1^+}}; \mu_p \hat{u}_p \vec{k}_p | F_p | 0; 1 m_r \rangle \\
 &= (-1)^{\frac{1}{2}+L_0+L_{1^+}+S_{1^+}+M_{S_0}} \sqrt{2S_0+1} \sum_{\ell_p m_p \nu_p L M_L} (-i)^{\ell_p} (-1)^{\ell_p} e^{i\sigma_{\ell_p}} (2L+1) \\
 &\quad \times \begin{pmatrix} L_{1^+} & \ell_p & L \\ M_{L_{1^+}} & m_p & -M_L \end{pmatrix} \begin{pmatrix} L_0 & 1 & L \\ M_{L_0} & m_r & -M_L \end{pmatrix} \begin{pmatrix} S_{1^+} & \frac{1}{2} & S_0 \\ M_{S_{1^+}} & \nu_p & -M_{S_0} \end{pmatrix} \\
 &\quad \times Y_{\ell_p}^{m_p}(\hat{k}_p) \mathcal{D}_{\mu_p \nu_p}^{1/2}(\omega_p) \langle (L_{1^+} \ell_p) L | F_p | (L_0 1) L \rangle. \tag{B1}
 \end{aligned}$$

Equation (B1) has been arrived at by using

$$\langle (L_{1^+} \ell_p) L M_L; (S_{1^+} \frac{1}{2}) S M_S | F_p | (L_0 1) L' M_{L'}; S_0 M_{S_0} \rangle^- = \delta_{L L'} \delta_{M_L M_{L'}} \delta_{S S_0} \delta_{M_S M_{S_0}} \langle (L_{1^+} \ell_p) L | F_p | (L_0 1) L \rangle$$

to meet the requirements arising from the conservations conditions (4). Here, the superscript “minus” means that the continuum wave function representing the photoelectron  $e_p$  in Eq. (1) satisfies the incoming wave boundary conditions [74] appropriate for photoionization. For brevity, however, this “minus” superscript has not been shown on the amplitude  $\langle (L_{1^+} \ell_p) L | F_p | (L_0 1) L \rangle$  present on the right hand side of Eq. (B1) and elsewhere in this paper.

Next, we substitute the amplitude (B1) and its Hermitian conjugate in Eq. (6b). The resulting expression is simplified by analytically evaluating as many sums as possible present therein. It requires, for example, use of (a) the addition theorems (i.e., Eqs. (4.3.2) and (4.6.5) from [57]) for rotational and spherical harmonics, (b) identity (5) given on page 453 in Ref. [90] for converting a single sum of the product of two 3- $j$  symbols into a product of two 3- $j$  and one 6- $j$  symbols summed over two variables, (c) Eq. (14.42) from [91] which transforms a quadruple sum of the product of four 3- $j$  symbols into a double sum containing two 3- $j$  and one 9- $j$  symbols, (d) Eq. (3.7.9) [57] for changing a phase factor into a 3- $j$  symbol, (e) orthogonality (3.7.7) [57] of 3- $j$  symbols, and (f) relation (6.4.14) [57] for writing a 9- $j$  symbol (whose one of the nine arguments is zero) in terms of a 6- $j$  symbol. These and some other simplifications lead one to write the DM (6b) in the following form:

$$\langle L_{1^+} S_{1^+} M_{S_{1^+}}; \mu_p \hat{u}_p \vec{k}_p | \rho_f | L_{1^+} S_{1^+} M'_{S_{1^+}}; \mu'_p \hat{u}_p \vec{k}_p \rangle = \frac{d^2 \sigma(m_r)}{d\epsilon_p d\hat{k}_p} \sigma(S_0, S_{1^+}; \hat{u}_p)_{\mu_p, M_{S_{1^+}}; \mu'_p, M'_{S_{1^+}}}. \tag{B2}$$

Here, we have defined

$$\begin{aligned}
 &\frac{d^2 \sigma(m_r)}{d\epsilon_p d\hat{k}_p} \\
 &= (-1)^{L_0+L_{1^+}+m_r} \frac{\mathcal{K}_p}{4\pi(2L_0+1)} \sum_{\ell_p \ell'_p L_r L L'} (2L_r+1) \begin{pmatrix} \ell_p & \ell'_p & L_r \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & 1 & L_r \\ m_r & -m_r & 0 \end{pmatrix} \left\{ \begin{matrix} 1 & 1 & L_r \\ L' & L & L_0 \end{matrix} \right\} \left\{ \begin{matrix} \ell_p & \ell'_p & L_r \\ L' & L & L_{1^+} \end{matrix} \right\} \\
 &\quad \times \langle L_{1^+} \ell_p | \mathcal{F}(L) | L_0 1 \rangle \langle L_{1^+} \ell'_p | \mathcal{F}(L') | L_0 1 \rangle P_{L_r}(\cos \theta_p), \tag{B3a}
 \end{aligned}$$

with

$$\langle L_{1^+} \ell_p | \mathcal{F}(L) | L_0 1 \rangle = (-i)^{\ell_p} e^{i\sigma_{\ell_p}} (-1)^{\ell_p} (2L+1) \sqrt{2\ell_p+1} \langle (L_{1^+} \ell_p) L | F_p | (L_0 1) L \rangle \tag{B3b}$$

and

$$\begin{aligned}
 &\sigma(S_0; S_{1^+}; \hat{u}_p)_{\mu_p, M_{S_{1^+}}; \mu'_p, M'_{S_{1^+}}} \\
 &= (-1)^{\mu'_p+S_0+M_{S_{1^+}}} \sum_{Q m n} (2Q+1) \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & Q \\ \mu_p & -\mu'_p & m \end{pmatrix} \begin{pmatrix} S_{1^+} & S_{1^+} & Q \\ M_{S_{1^+}} & -M'_{S_{1^+}} & -n \end{pmatrix} \left\{ \begin{matrix} \frac{1}{2} & \frac{1}{2} & Q \\ S_{1^+} & S_{1^+} & S_0 \end{matrix} \right\} [\mathcal{D}_{m n}^Q(\omega_p)]^*. \tag{B4}
 \end{aligned}$$

In both Eqs. (B2) and (B4),  $M'_{S_{1^+}}$  ( $=\hat{e}_z \cdot \vec{S}_{1^+}$ ) (similar to  $M_{S_{1^+}}$ ) also is a projection of the spin angular momentum of the photoion  $A^{1^+}$  along the polar axis of the SF shown in Fig. 1. Further, in Eq. (B3a),  $P_{L_r}(\cos \theta_p)$  is the Legendre polynomial [57] of order  $L_r$  and argument  $\cos \theta_p$ ; (B3b) are the dynamical amplitudes [87] related to the matrix elements (B1) for the photoionization process (1).

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