PHYSICAL REVIEW A **70**, 060306(R) (2004)

Generation of tunable entangled states of two electrons and their characterization without entanglement witness

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(Received 26 July 2004; published 30 December 2004)

In this Rapid Communication, we show that a simple process of two-step double ionization taking place in an atom A, following the absorption of a single photon, produces a spin-entangled state of two electrons. The degree of entanglement of this state can be tuned to the desired value by selecting appropriate total spin quantum numbers of the electronic states of each of the three atomic species (i.e., A, A^{+}, A^{2+}) participating in the process in Russell-Saunders coupling. These entangled states are readily characterized by measuring only energies of two emitted electrons, without requiring the entanglement witness, or any other such protocol.

DOI: 10.1103/PhysRevA.70.060306 PACS number(s): 03.67.−a

The rapidly developing field of quantum information is based on the existence of nonseparable (i.e., entangled) states of more than one particles in quantum mechanics. Consequently, the production and characterization of entangled states of two or more particles is currently of great interest. Although, the recently implemented [1,2] entanglement witness (EW) [3] is, hitherto, probably the only protocol available for characterization and/or detection of entanglement; there have nevertheless been several methods proposed for generating two entangled particles in which neither [1,2,4], both [5–7], or one of the two [8] possesses rest mass different from zero. Among these [1,2,4–8], however, the parametric down conversion (PDC) [9] has so far been the most successful and widely used method for producing a pair of entangled photons. Even in demonstrations on realization of an EW, the experiments were performed on nonseparable states of two [1] or more [2] polarized photons produced by PDC.

Many applications of entanglement, nevertheless, require nonseparable states of two or more particles possessing rest mass different from zero. For such particles can interact and be detected, unlike photons, without being destroyed. In this Rapid Communication, we discuss a very simple process in atoms for generating states of two electrons, say (e_1, e_2), which are entangled with respect to their spins. The proposed method, in addition, has several distinct advantages over the other procedures [4–9] hitherto suggested for generating nonseparable states of two or more particles.

For example, (i) it can produce two electrons (e_1, e_2) in a maximally chaotic [1(a)] $\sigma_0 \equiv I/4$ state, in a pure, maximally entangled, singlet spin state $\sigma_1(\hat{u}_1, \hat{u}_2)$, or in a state which is a mixture of these two possible extreme states of (e_1, e_2). Here, I is a unit matrix, $\hat{u}_1(\vartheta_1, \varphi_1)$ and $\hat{u}_2(\vartheta_2, \varphi_2)$ are the spin quantization directions of (e_1, e_2), respectively. (ii) The degree $p(=0-1)$ of entanglement of the state constituted by mixing σ_0 and σ_1 can be predetermined, and hence "tuned," according to one's requirements. The proposed method thus produces two electrons with a tunable degree of spin-entanglement. (iii) Such entangled states are completely characterized by measuring merely energies of (e_1, e_2), without using any EW protocol or detecting the spins of the electrons.

Let us consider emission of (e_1, e_2) from an atom A in two different, but sequential, steps shown in Eq. (1). There, $E_r = h\nu_r$ is the energy of the photon absorbed in the electric dipole $(E1)$ approximation; photoelectron e_1 is ejected from one of the inner shells of A; the subsequent nonradiative, spontaneous decay of the excited photoion A^{+^*} in the second step in (1) emits e_2, called Auger electron,

$$h\nu_r|1,m_r\rangle + A|0\rangle \to A^{+^*}|e\rangle + e_1(\mu_1,\hat{u}_1,\vec{k}_1),$$

$$A^{+^*}|e\rangle \to A^{2+}|f\rangle + e_2(\mu_2,\hat{u}_2,\vec{k}_2). \qquad (1)$$

Here, the propagation vector of the $i(=1,2)$th electron e_i is $\vec{k}_i = (k_i, \theta_i, \phi_i)$ such that its kinetic energy is given by $\epsilon_i = \hbar^2 k_i^2/2m$; also, $\mu_i\left(=\pm\frac{1}{2}\right)$ is the projection of the spin of e_i along $\hat{u}_i(\vartheta_i, \varphi_i)$. The respective kets $|0\rangle$, $|e\rangle$, and $|f\rangle$ in Eqs. (1) represent the antisymmetrized electronic states of A, A^{+^*}, and of the dication A^{2+} possessing the energies E_0, E_e, and E_f. Thus in (1), while energy $\epsilon_1 = h\nu_r - (E_e - E_0)$ of e_1 varies with that of the photon absorbed; energy $\epsilon_2 = (E_e - E_f)$ of e_2 is fixed and is completely determined from that of A^{+^*} and A^{2+}. Ket $|1, m_r\rangle$ in Eq. (1) specifies [10] the polarization of the absorbed photon: $m_r = +1$ and -1 are for photons with positive and negative helicities, respectively; whereas $m_r = 0$ corresponds to a linearly polarized photon. (An unpolarized electromagnetic wave is taken to be an even mixture of photons with negative and positive helicities.) Accordingly, a photon in a $|1, +1\rangle$ or $|1, -1\rangle$ state is incident, but a photon in $|1, 0\rangle$ state has its electric field vector, along the polar (i.e., OZ) axis of our coordinate system.

Both steps in the process (1) are completely described by the density operator

$$\rho_f = \mathcal{K}_2 F_2 \rho_1 F_2^\dagger, \quad \text{with [6,7]} \quad \rho_1 = \mathcal{K}_1 F_1 (\rho_0 \otimes \rho_r) F_1^\dagger \qquad (2)$$

being the density operator for the first step of photoionization in (1). In ρ_1, $F_1 = \sqrt{m/\hbar^2}F$ is the photoionization operator with the operator F in the $E1$ approximation defined, for example, in [11]; $\rho_r = |1, m_r\rangle\langle 1, m_r|$ and $\rho_0 = |0\rangle\langle 0|$ are the respective density operators [6,7] of the ionizing radiation and of the unpolarized atom A, assumed to be uncorrelated before the interaction between the two takes place. ρ_f in (2), on the other hand, contains the Auger emission operator F_2 [12]. The quantities $\mathcal{K}_1$ and $\mathcal{K}_2$, whose explicit forms are not of

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interest for the present discussion, are given in Refs. [11,12], respectively.

In this Rapid Communication, we want to investigate entanglement properties of (e_1,e_2) without considering spin-orbit interaction (SOI) in any of the species A,A^{+^*},A^{2+}, and in the continua of the two sequentially ejected electrons. It, in other words, means that Russell-Saunders (i.e., *LS*) coupling becomes applicable to whole of the process (1). Moreover, in the absence of SOI and due to the spin-independent nature of both of the operators F_1 [11] and F_2 [12], the total spin before and after each of the two steps in the process (1) is conserved, i.e.,

$$\vec{S}_0=\vec{S}_e+\vec{s}_1 \quad \text{and} \quad \vec{S}_e=\vec{S}_f+\vec{s}_2. \tag{3}$$

Here, $S_0,\ S_e$, and S_f are the spins of the respective electronic states $|0\rangle$ of A, $|e\rangle$ of A^{+^*}, and $|f\rangle$ of A^{2+} participating in the process (1), whereas s_i [with $|\vec{s}_i|=(1/2)_i$] is the spin angular momentum of the electron e_i in (1) quantized along $\hat{u}_i$.

Now the most general form of the density operator $(\rho_0 \otimes \rho_r)$, which represents in (2) an unpolarized atom A in the *LS* coupling plus a noninteracting photon, can be written as

$$(\rho_0\otimes\rho_r)=\frac{1}{(2L_0+1)(2S_0+1)}\sum_{M_{L_0}\,M_{S_0}}|0;1,m_r\rangle\langle 0;1,m_r|. \tag{4}$$

Here, $|0;1,m_r\rangle\equiv|0\rangle|1,m_r\rangle$ represents a state of the noninteracting (photon + atom) system; L_0 is the total orbital angular momentum of A in its $|0\rangle$ electronic state; M_{L_0} and M_{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. Further in (4), we have averaged over all the degenerate states of A. The density matrix (DM), calculated for the operator (2) in the *LS* coupling, simplifies to a product of two independent terms in the following form

$$\begin{aligned}\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2\rangle \\ =\frac{d^3\sigma(m_r)}{d\epsilon_1\,d\hat{k}_1\,d\hat{k}_2}\sigma_p(\hat{u}_1,\hat{u}_2)_{\mu_1,\mu_2;\mu_1',\mu_2'}.\end{aligned} \tag{5}$$

The first term $d^3\sigma(m_r)/d\epsilon_1\,d\hat{k}_1\,d\hat{k}_2$ on the right-hand side of (5) contains, among other things, the directions of propagation $(\hat{k}_1,\hat{k}_2)$, energies (ϵ_1,ϵ_2) as well as phase shifts of two emitted electrons (e_1,e_2), and the state of polarization of the ionizing radiation. It, in addition, has total orbital angular momenta (L_0,L_e,L_f) of (A,A^{+^*},A^{2+}), products of the dynamical amplitudes for $E1$ photoionization (calculated using the electronic states of A and of A^{+^*}) and for Auger emission (determined by the electronic states of A^{+^*} and of A^{2+}). Also, the first term in (5) has an implicit dependence upon the spins (S_0, S_e, S_f) of the atomic species (i.e., A,A^{+^*},A^{2+}). It is, however, totally independent of the quantization directions $\hat{u}_i$ as well as of the spins of the electrons (e_1,e_2) emitted in (1). Hence, $d^3\sigma(m_r)/d\epsilon_1\,d\hat{k}_1\,d\hat{k}_2$ in (5) describes purely angular correlation between (e_1,e_2). Its explicit form, given elsewhere [13], is not needed for the present discussion.

The second term

$$\begin{aligned}&\sigma_p(\hat{u}_1,\hat{u}_2)_{\mu_1,\mu_2;\mu_1',\mu_2'}\\ &\quad=(-1)^{S_0+S_f-2S_e+\mu_1'+\mu_2'}(2S_e+1)\sum_{s\,n\,m_1\,m_2}(-1)^{s+n}(2s+1)\\ &\qquad\times\begin{pmatrix}1/2 & 1/2 & s\\ \mu_1 & -\mu_1' & m_1\end{pmatrix}\begin{pmatrix}1/2 & 1/2 & s\\ \mu_2 & -\mu_2' & m_2\end{pmatrix}\begin{Bmatrix}1/2 & 1/2 & s\\ S_e & S_e & S_0\end{Bmatrix}\\ &\qquad\times\begin{Bmatrix}1/2 & 1/2 & s\\ S_e & S_e & S_f\end{Bmatrix}\\ &\quad\times[\mathcal{D}^s_{m_1,n}(\varphi_1,\vartheta_1,0)]^*[\mathcal{D}^s_{m_2,-n}(\varphi_2,\vartheta_2,0)]^*,\end{aligned} \tag{6}$$

on the right-hand side of (5) is, on the other hand, a (4×4) matrix which does not contain any of those physical variables which are present in the angular correlation and, hence, is totally independent of the dynamical effects contributing to the process (1). Obviously, matrix (6) is *completely determined* by the spins of all the five particles (i.e., A,A^{+^*},A^{2+},e_1, and e_2) involved in the process (1) and by the directions $(\hat{u}_1,\hat{u}_2)$ of spin quantization of both of the ejected electrons (e_1,e_2). The variable s, present in (6), is a dummy summation index. Hence, the second term (6), in the DM (5), represents a purely spin correlation between (e_1,e_2). The subscript "p" to σ in (5), and elsewhere in this Rapid Communication, is a parameter $p=p(S_0;S_e;S_f)$ containing the dependence of the spin correlation matrix (6) on the three spin quantum numbers $S_0,\ S_e$, and S_f [see, for example, Eqs. (9a) and (9b)]. Further in (6), following the definitions given by Edmonds [14], each (with two rows and three columns) of the two big parentheses and two curly brackets, and $\mathcal{D}'$s are the 3-j symbols, 6-j symbols, and the rotational harmonics, respectively.

Although it is not possible to give the details of the derivations of Eqs. (5) and (6) in this Rapid Communication for the reasons of space, the separation present in (5) of the DM into its two parts describing purely angular and purely spin correlations between (e_1,e_2) in the absence of SOI is, nevertheless, completely rigorous and independent of all dynamical models.

It is obvious from the spin conservation conditions (3), as well as from the two 6-j symbols [14] present in (6), that each of the spins S_0 and S_f must be equal to $S_e\pm 1/2$. The only two possibilities are, therefore, $S_0=S_f$ and $|S_0-S_f|=1$. In the following, we investigate the spin entanglement between (e_1,e_2) in both of these cases. This property of the two electrons will be completely determined by the matrix (6) as the angular correlation in (5) is always *positive* and acts as a *multiplicative* factor. Therefore, unless stated otherwise, we write the DM (5) as

$$\begin{aligned}\langle f;\mu_1,\hat{u}_1,\vec{k}_1;\mu_2,\hat{u}_2,\vec{k}_2|\rho_f|f;\mu_1',\hat{u}_1,\vec{k}_1;\mu_2',\hat{u}_2,\vec{k}_2\rangle \\ \to\sigma_p(\hat{u}_1,\hat{u}_2)_{\mu_1,\mu_2;\mu_1',\mu_2'}.\end{aligned} \tag{7}$$

(i) $S_0=S_f(=S_e\pm 1/2)$.

(a) Let us first consider the process (1) for the case when $S_0=1/2,S_e=0,S_f=1/2$. The DM, obtained from (6) and (7), is given by

$$\sigma_p(\hat{u}_1,\hat{u}_2)_{\mu_1,\mu_2;\mu_1',\mu_2'} = (1/4)\,\delta_{\mu_1\mu_1'}\delta_{\mu_2\mu_2'} \equiv (\sigma_0)_{\mu_1,\mu_2;\mu_1',\mu_2'} \tag{8a}$$

with δ_{ab} the Kronecker delta function [14]. This is a diagonal matrix with each of the four eigenvalues of (8a) and of its partial transpose (PT) equal to 1/4. It represents [1(a)] a maximally chaotic state, corresponding to a mixed separable state, of the two electrons (e_1,e_2). One of the simplest possible examples of this result can be the two-step DPI $B(1s^2\,2s^2\,2p^1\,{}^2P) \to B^{+^*}(1s^1\,2s^2\,2p^1\,{}^1P) \to B^{2+}(1s^2\,2s^1\,2p^0\,{}^2S)$ in the ground electronic configuration of a boron atom. The sequentially emitted (e_1,e_2) in this case form the σ_0 state.

(b) Next we look at the transition (1) for $S_0=0, S_e=1/2, S_f=0$. The DM, obtained from Eqs. (6) and (7), for this case is

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

$$= \begin{array}{c|cccc} \mu_1,\mu_2/\mu_1',\mu_2' \Rightarrow \\ \Downarrow & \frac{1}{2},\frac{1}{2} & \frac{1}{2},-\frac{1}{2} & -\frac{1}{2},\frac{1}{2} & -\frac{1}{2},-\frac{1}{2} \\ \hline \frac{1}{2},\frac{1}{2} & 1-\hat{u}_1\cdot\hat{u}_2 & \begin{array}{c} c_1s_2 - s_1c_2c \\ -is_1s \end{array} & \begin{array}{c} s_1c_2 - c_1s_2c \\ +is_2s \end{array} & \begin{array}{c} -s_1s_2 + (1-c_1c_2)c \\ -i(c_1-c_2)s \end{array} \\ \frac{1}{2},-\frac{1}{2} & \begin{array}{c} c_1s_2 - s_1c_2c \\ +is_1s \end{array} & 1+\hat{u}_1\cdot\hat{u}_2 & \begin{array}{c} -s_1s_2 - (1+c_1c_2)c \\ +i(c_1+c_2)s \end{array} & \begin{array}{c} -s_1c_2 + c_1s_2c \\ -is_2s \end{array} \\ -\frac{1}{2},\frac{1}{2} & \begin{array}{c} s_1c_2 - c_1s_2c \\ -is_2s \end{array} & \begin{array}{c} -s_1s_2 - (1+c_1c_2)c \\ -i(c_1+c_2)s \end{array} & 1+\hat{u}_1\cdot\hat{u}_2 & \begin{array}{c} -c_1s_2 + s_1c_2c \\ +is_1s \end{array} \\ -\frac{1}{2},-\frac{1}{2} & \begin{array}{c} -s_1s_2 + (1-c_1c_2)c \\ +i(c_1-c_2)s \end{array} & \begin{array}{c} -s_1c_2 + c_1s_2c \\ +is_2s \end{array} & \begin{array}{c} -c_1s_2 + s_1c_2c \\ -is_1s \end{array} & 1-\hat{u}_1\cdot\hat{u}_2 \end{array} \tag{8b}$$

with $\hat{u}_1\cdot\hat{u}_2 = c_1c_2 + s_1s_2c$, and the definitions: $i \equiv \sqrt{(-1)}$, $s_1 \equiv \sin\vartheta_1$, $s_2 \equiv \sin\vartheta_2$, $c_1 \equiv \cos\vartheta_1$, $c_2 \equiv \cos\vartheta_2$, $s \equiv \sin(\varphi_2 - \varphi_1)$, $c \equiv \cos(\varphi_2 - \varphi_1)$.

The respective eigenvalues $(0,0,0,1)$ of (8b) and $(\frac{1}{2},\frac{1}{2},-\frac{1}{2},\frac{1}{2})$ of its PT are completely independent of the angles which specify the spin quantization directions $(\hat{u}_1,\hat{u}_2)$ of (e_1,e_2). These eigenvalues clearly show that the DM (8b) represents a pure and entangled state of (e_1,e_2). One further finds, from the reduced matrix of (8b), that the degree of nonseparability (participation ratio or Schmidt number) [15] for this pure state is maximum, i.e., $K=2$, which is that of a Bell state. In conclusion, DM (8b) represents a pure and maximally entangled state of two spin-half particles corresponding [7] to a singlet spin state of e_1 and e_2. Among the several possible, simple examples for this case can be the two-step DPI in C, O, inert gases, etc. For example, $\mathrm{C}(1s^2\,2s^2\,2p^2\,{}^1S/{}^1D) \to \mathrm{C}^{+^*}(1s^1\,2s^2\,2p^2\,{}^2S/{}^2D) \to \mathrm{C}^{2+}(1s^2\,2s^2\,2p^0\,{}^1S)$. Here, e_1 and e_2 are in the σ_1 state.

(c) The density matrices for other transitions involving the processes (1) with $S_0=S_f$ are also obtained from Eqs. (6) and (7) and can be shown to be given by

$$\sigma_p\,(\hat{u}_1,\hat{u}_2)_{S_0=S_f} = p(S_0=S_f,S_e)\,\sigma_1(\hat{u}_1,\hat{u}_2) + (1-p)\,\sigma_0. \tag{9a}$$

with the parameter

$$p(S_0=S_f;S_e) = \begin{cases} 0 & \text{for } S_e = 0 \\ \dfrac{1}{3S_e(S_e+1)}\left[\frac{3}{4} + S_e(S_e+1) - S_0(S_0+1)\right]^2 & \text{for } S_e > 0. \end{cases} \tag{9b}$$

It is obvious from Eq. (9b) that $p \geqslant 0$, always; in addition, on account of $|S_0 - S_e| = 1/2, p \leqslant 1$. Also, $p(S_0=1/2;S_e=0;S_f=1/2)=0$ and $p(S_0=0;S_e=1/2;S_f=0)=1$. With each of these two allowed extreme values of p, the DM (9a) rightly reproduces states (8a) and (8b) for $p=0$ and $p=1$, respectively. The eigenvalues of (9a) and of its partial transpose, in terms of p, are $\frac{1}{4}(1-p)$, $\frac{1}{4}(1-p)$, $\frac{1}{4}(1-p)$, and $\frac{1}{4}(1+3p)$ for the DM; $\frac{1}{4}(1+p)$, $\frac{1}{4}(1+p)$, $\frac{1}{4}(1-3p)$, and $\frac{1}{4}(1+p)$ for the PT of (9a). Thus, for all allowed values (0 to 1) of p, specified by Eq. (9b), each of the four eigenvalues of $\sigma_p(\hat{u}_1,\hat{u}_2)|_{S_0=S_f}$ is always greater than or equal to zero; on the other hand, one [i.e., $\frac{1}{4}(1-3p)$] of the four eigenvalues of the PT of this DM becomes negative for $p \geqslant 1/3$. That is, if p in Eqs. (9a) and (9b) exceeds 1/3 (but remaining less than 1), the corresponding DM represents an entangled state which is a mixture of the states σ_0 and σ_1.

States of two or more particles which can be expressed in the form of Eq. (9a), with $0 \leqslant p \leqslant 1$ are known [1(a),3] as Werner [16] states. As p determines the amount of mixing of $\sigma_1(\hat{u}_1,\hat{u}_2)$ with σ_0, it is therefore called [1(a),3] the "mixing parameter" or the "probability" for the Werner state (9a). The important, as well as interesting, thing here is that a value of

p in Eq. (9b) depends only on the total spins of each of the species A, A^{+^*}, and A^{2+} participating in the process (1). One can, therefore, determine the values of p *a priori* and, hence, select the appropriate photoionizing and Auger transitions in the process (1), according to one's requirements of the degree of mixing of the $\sigma_1(\hat{u}_1,\hat{u}_2)$ and σ_0 states in order to produce a spin-entangled state of (e_1,e_2). In other words, DPI in Eq. (1) provides a method for producing Werner states (9a) and (9b) of two electrons with a tunable degree of their spin entanglement. p can, therefore, be called also a tuning parameter.

One can give several examples to illustrate the above-mentioned points. While the transitions $N(1s^2\ 2s^2\ 2p^3\ {}^4S) \rightarrow N^{+^*}(1s^1\ 2s^2\ 2p^3\ {}^3S) \rightarrow N^{2+}(1s^2\ 2s^1\ 2p^2\ {}^4P)$, with $p(S_0=3/2, S_f=3/2, S_e=1)=1/6$, produce (e_1,e_2) in a mixed product Werner state, the two electrons emitted sequentially in transitions $N(1s^2\ 2s^2\ 2p^3\ {}^2P/{}^2D) \rightarrow N^{+^*}(1s^1\ 2s^2\ 2p^3\ {}^3P/{}^3D) \rightarrow N^{2+}(1s^2\ 2s^2\ 2p^1\ {}^2P)$ are in an entangled Werner state with $p(S_0=S_f=1/2, S_e=1)=2/3$.

The other interesting thing about the method proposed herein is that one needs not use EW protocol or any other method (e.g., measurement of the spins of the ejected electrons), for that matter, for detecting and/or characterizing the nonseparability of the state of (e_1,e_2). A simple measurement of the energies of the photoelectron and of Auger electron will readily identify the electronic states of each of the species A, A^{+^*}, and of A^{2+} in the LS coupling. This will, in turn, determine their spins S_0, S_e, and S_f, respectively. One can, subsequently, calculate the degree p of spin entanglement of (e_1,e_2) using Eq. (9b).

(ii) $|S_0-S_f|=1$ (with $S_0=S_e\pm 1/2, S_f=S_e\mp 1/2$).

The DM (7) in this case can be shown from Eq. (6) to reduce to the following form:

$$\sigma_p\,(\hat{u}_1,\hat{u}_2)_{|S_0-S_f|=1} = -\,(1/3)\sigma_1(\hat{u}_1,\hat{u}_2) + (4/3)\sigma_0. \qquad (10)$$

The eigenvalues of this matrix and of its PT are, respectively, $(1/3,1/3,1/3,0)$ and $(1/6,1/6,1/2,1/6)$. Thus (10) represents a mixed and separable state of the electrons (e_1,e_2). A relevant example for the present case can be the two-step DPI $C(1s^2\ 2s^2\ 2p^2\ {}^1S/{}^1D) \rightarrow C^{+^*}(1s^1\ 2s^2\ 2p^2\ {}^2S/{}^2D) \rightarrow C^{2+}(1s^2\ 2s^0\ 2p^2\ {}^3P)$. Further, $\sigma_p(\hat{u}_1,\hat{u}_2)|_{|S_0-S_f|=1}$ is not a Werner state as the coefficient of the pure, maximally entangled spin state σ_1 in (10) is always negative.

Hence, production of (e_1,e_2) with a desired degree of entanglement requires simultaneous existence of the Russell-Saunders states $(|0\rangle,|e\rangle,|f\rangle)$ with the appropriate values of their respective spins (S_0,S_e,S_f) for the given atomic species. One can always, in general, select or prepare an atom A in the needed state $|0\rangle$; generation of the right state of A^{+^*} will, however, require a proper value of the energy of the absorbed photon to cause the $|0\rangle \rightarrow |e\rangle$ ionizing $E1$ transition. But, formation of the state $|f\rangle$ of A^{2+} will depend on the various competing channels available for the nonradiative, spontaneous decay of the excited phtoion A^{+^*}. If more than one triod $(|0\rangle,|e\rangle,|f\rangle)$ of the appropriately allowed states exist in a single atom, it will then be possible to obtain different pairs of (e_1,e_2) possessing correspondingly tuned, different degrees of entanglement in a single experiment with that atomic species. Otherwise, different atoms, with appropriate triod $(|0\rangle,|e\rangle,|f\rangle)$ of states need to be used in different experiments for producing (e_1,e_2) with different, desired degrees of entanglement.

A final point, which probably also needs to be discussed herein, is that in the present study we have not taken SOI in either of the two steps of the process (1) into account. Inclusion of SOI will mean that neither of the two Eqs. (3) will be valid. This will, consequently, lead to a situation wherein DM (5) cannot be separated into its angular and spin parts, and hence cannot be written in the form of the product (5). The dynamical effects will play a very important role in the spin entanglement between sequentially emitted (e_1,e_2) in the presence of SOI. An immediate important consequence, among others, of this on the process (1) will, therefore, be that one can no longer determine *a priori* whether (e_1,e_2) are entangled or not. The effects of SOI in B, C, N, etc. atoms, considered for various examples herein, are well known to be negligibly small. These effects certainly become important for heavier atoms.

This work was supported, in part, by the Council of Scientific & Industrial Research, New Delhi, India, under Grant No. 03(0952)/02/EMR-II.

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