

Generation and control of spin-orbit entanglement in atomic pairs

Xuan Li^{1,*} and Moshe Shapiro^{1,2}¹*Department of Chemistry, The University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1*²*Department of Chemical Physics, The Weizmann Institute, Rehovot 76100, Israel*

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We propose a method for producing entangled spin and orbital states in pairs of atoms resulting from the dissociation of diatomic molecules. The method employs the interference between a one-photon and a two-photon process to break the backward-forward symmetry of the dissociation processes. The spin entanglement of distant atom pairs makes use of excited spin-orbit-coupled (molecular) intermediate states. When the dissociating molecules are confined to a one-dimensional optical lattice, the method can be used to generate long sequences of (spin- and orbit-) entangled pairs of atoms.

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I. INTRODUCTION

The controlled generation of entangled states, which is essential for many quantum information applications such as superdense coding, quantum teleportation, quantum computing, and quantum cryptography [1–4], is a challenging task. High-order entanglements, which involve either many particles [5], multiple dimensions [6], or many degrees of freedom [7], are even more difficult to produce. Previous works have primarily concentrated on, in addition to two-level atomic systems, cavity QED systems [8], excitons in semiconductor microcrystallites [9], two-atom autoionizing systems [10], three-spin chain systems [11], nonlinear quantum scissors devices based on Kerr-like systems [12], trapped ions [13], linear optical models [14], quantum-dot systems [15], and microcavities [16].

In general we can categorize entangled states as being composed of states of the same degree of freedom $|i\rangle \otimes |j\rangle$ or being composed of states belonging to different degrees of freedom. Trapped-ion W states [17,18], in which one correlates between quantum bits of different ions associated with the same type of state, are an example of the former. States composed of spin and orbital angular momenta of the same particle are an example of the latter. Such states are the $S_z(1) \otimes S_z(2) \otimes P_z(1) \otimes P_z(2)$ states that have been realized for photons [19].

In this paper we propose and develop a method for producing both types of entangled states in atoms (or ions). The method employs the interference between a one-photon and a two-photon dissociation process [20–24] to produce a Greenberger-Horne-Zeilinger (GHZ) type of state [25], such as $|0000\rangle + |1111\rangle$, involving spin and orbital angular momenta of *different* atoms. In such states a measurement of S_z of one particle yields, on top of the spin state of the other particle, the orbital information of both particles.

In the present scenario the one-photon process is used to execute a spin-conserving excitation to a dissociative state, while the two-photon process is used to excite the molecule to a spin-orbit-coupled (molecular) intermediate state, from which it is transferred to a dissociative state of *different* spin compared to the ground state. Lacking the directional

information, the above two pathways of producing the final photofragments interfere, allowing for coherent control of the directionality of the entangled states. We aid in the control of the makeup of these states by using an additional laser that changes the two-photon phase by ac Stark shifting the position of the intermediate states. In the context of molecules dissociated in an optical lattice [26,27], this method can produce arbitrarily long chains of pairs of entangled GHZ atoms.

II. THEORY

We consider the interaction of a system, composed of $|E_0\rangle$, the initial bound state of energy E_0 ; $|E_i\rangle$, an intermediate bound state of energy E_i ; and $|E, n^-\rangle$ ($n = 1, \dots, N$), a set of N -fold degenerate scattering states of continuous energies E , with three external laser fields $\epsilon_j \equiv \epsilon_j \hat{\epsilon}_j$ ($j = 1, 2, 3$) where ϵ_j is the amplitude and $\hat{\epsilon}_j$ is the polarization of each laser field. The total Hamiltonian is given as $H = H_M + H_{\text{int}}$, where H_M is the radiation-free material Hamiltonian and $H_{\text{int}} = -\sum_j \hat{\mu} \cdot \epsilon_j \hat{\epsilon}_j$ is the radiation-matter interaction term, with μ denoting the electric dipole operator. The above (nuclear plus electronic) eigenstates of H_M ,

$$[H_M - E_0]|E_0\rangle = [H_M - E_i]|E_i\rangle = [H_M - E]|E, n^-\rangle = 0, \quad (1)$$

$$n = 1, \dots, N,$$

are used to expand $|\Psi\rangle$, the solution of the time-dependent Schrödinger equation

$$i\hbar \partial |\Psi\rangle / \partial t = H |\Psi\rangle$$

as

$$|\Psi\rangle = a_0 e^{-iE_0 t} |E_0\rangle + a_i e^{-iE_i t} |E_i\rangle + \sum_n \int dE b_{E,n} e^{-iEt} |E, n^-\rangle. \quad (2)$$

The $\epsilon_{j=1,2,3}$ laser fields are respectively tuned to be in near resonance with the $|E_0\rangle \rightarrow |E, n^-\rangle$, $|E_0\rangle \rightarrow |E_i\rangle$, and $|E_i\rangle \rightarrow |E, n^-\rangle$ transitions. By substituting Eq. (2) into the time-dependent Schrödinger equation, using atomic units ($\hbar = 1$), and invoking the rotating-wave approximation, we obtain

*xli3536@chem.ubc.ca

the set of coupled equations

$$\begin{aligned}
 \dot{a}_0 &= i\mu_{0,i}\varepsilon_2 a_i e^{-i(\Delta_2 t - \phi_2)} \\
 &+ i \sum_n \int dE \varepsilon_1 e^{-i(\Delta_1 t - \phi_1)} \mu_{0,n} b_{E,n}, \\
 \dot{a}_i &= i\mu_{i,0}\varepsilon_2 a_0 e^{i(\Delta_2 t - \phi_2)} \\
 &+ i \sum_n \int dE \varepsilon_3 e^{-i(\Delta_3 t - \phi_3)} \mu_{i,n} b_{E,n}, \\
 \dot{b}_{E,n} &= i\mu_{n,0}\varepsilon_1 a_0 e^{i(\Delta_1 t - \phi_1)} \\
 &+ i\mu_{n,i}\varepsilon_3 a_i e^{i(\Delta_3 t - \phi_3)}, \quad n = 1, \dots, N
 \end{aligned} \tag{3}$$

in which the transition dipole matrix elements are defined as $\mu_{0,n} \equiv \langle E_0 | \hat{\mu} \cdot \hat{\mathbf{e}}_1 | E, n^- \rangle$, $\mu_{i,n} \equiv \langle E_i | \hat{\mu} \cdot \hat{\mathbf{e}}_3 | E, n^- \rangle$ ($n = 1, \dots, N$), and $\mu_{0,i} \equiv \langle E_0 | \hat{\mu} \cdot \hat{\mathbf{e}}_2 | E_i \rangle$. The three detuning factors are defined as $\Delta_1 = E - E_0 - \omega_1$, $\Delta_2 = E_i - E_0 - \omega_2$, and $\Delta_3 = E - E_i - \omega_3$.

By invoking first- and second-order perturbation theory, we can express the final states' coefficients at the end of the pulse as

$$b_{E,n} = \Omega_n^{(1)} + \Omega_n^{(2)},$$

where

$$\begin{aligned}
 \Omega_n^{(1)} &\equiv i e^{-i\phi_1} \mu_{n,0}(E) \epsilon_1(\Delta_1), \\
 \Omega_n^{(2)} &\equiv -\mu_{n,i}(E) \mu_{i,0} e^{-i\phi_2 - i\phi_3} \epsilon_{2,3}(\Delta_2, \Delta_3), \quad n = 1, \dots, N,
 \end{aligned} \tag{4}$$

and

$$\begin{aligned}
 \epsilon_1(\Delta_1) &= \int_{t_0}^{\infty} dt \varepsilon_1(t) e^{i\Delta_1 t}, \\
 \epsilon_{2,3}(\Delta_2, \Delta_3) &= \int_{t_0}^{\infty} dt \varepsilon_3(t) e^{i\Delta_3 t} \int_{t_0}^t dt' \varepsilon_2(t') e^{i\Delta_2 t'}.
 \end{aligned} \tag{5}$$

Equation (4) leads to the possible control of the relative channel weights and phases $\Omega_n^{(1)}/\Omega_n^{(2)} = |\Omega_n^{(1)}/\Omega_n^{(2)}| \exp(i\alpha_n)$ by tuning the laser phase difference $\phi_1 - \phi_2 - \phi_3$. The scenario outlined here is depicted schematically in Fig. 1.

In order to illustrate the method we now study the dissociation of the F_2 molecule (Fig. 2). The electronic state associated with $|E_0\rangle$ is chosen as $X^1\Sigma_g^+$ and the electronic state associated with $|E_i\rangle$ is chosen as $a^3\Pi_{1u}$. We consider four continuum channels ($n = 1, 2, 3, 4$) belonging to the $A^1\Pi_{1u}$ and $b^3\Pi_{1g}$ states. The final dissociative state is a result of two possible quantum routes, the one-photon process $X \xrightarrow{\hbar\omega_1} A$ and the two-photon process $X \xrightarrow{\hbar\omega_2} a \xrightarrow{\hbar\omega_3} b$.

The use of n of Eq. (2) to denote (among other quantum numbers) the physically interesting backward b and forward f directions amounts to working with symmetry-broken states. Such states are coupled to the initial state by both the one-photon and the two-photon processes. In order to compute the transition amplitudes, however, we need to correlate these symmetry-broken states with the usual symmetry-adapted states, e.g., the gerade and ungerade states or the spin triplet and singlet states. The spin parts of the symmetry-adapted final dissociative states $A^1\Pi_{1u}$ and $b^3\Pi_{1g}$ are given as

$$\begin{aligned}
 S(u) &= [\alpha(f)\beta(b) - \beta(f)\alpha(b)]/\sqrt{2}, \\
 S(g) &= [\alpha(f)\beta(b) + \beta(f)\alpha(b)]/\sqrt{2}.
 \end{aligned} \tag{6}$$

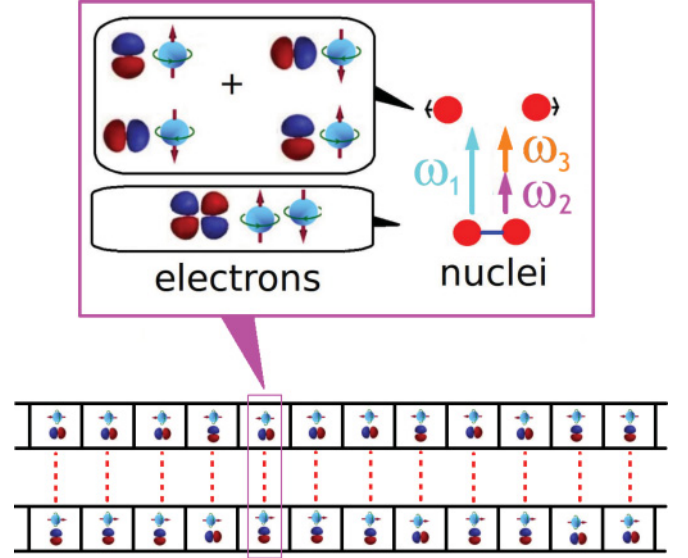


FIG. 1. (Color online) Schematic process of the controlled entanglement in spin and orbital angular momenta and directionality of the dissociated atomic pairs.

The symmetry-adapted orbital part of the two dissociative states is obtained in the asymptotic region using the linear combination of atomic orbitals approximation

$$\begin{aligned}
 \mathcal{O}(A^1\Pi_{1u}, \Omega = \Gamma = 1) &\xrightarrow{R \rightarrow \infty} [|\mathcal{O}_1\rangle + |\mathcal{O}_2\rangle] \\
 &\equiv [Y_{1,1}(f)Y_{1,0}(b) + Y_{1,0}(f)Y_{1,1}(b)]
 \end{aligned} \tag{7}$$

for the u state and

$$\begin{aligned}
 \mathcal{O}(b^3\Pi_{1g}, \Omega = \Gamma = 1) &\xrightarrow{R \rightarrow \infty} [|\mathcal{O}_1\rangle - |\mathcal{O}_2\rangle] \\
 &\equiv [Y_{1,1}(f)Y_{1,0}(b) - Y_{1,0}(f)Y_{1,1}(b)]
 \end{aligned} \tag{8}$$

for the g state, where $Y_{l,m}(f)$ and $Y_{l,m}(b)$ are spherical harmonics about the center of the forward- and backward-scattered atoms, respectively. Here Ω and Γ are defined as the projection

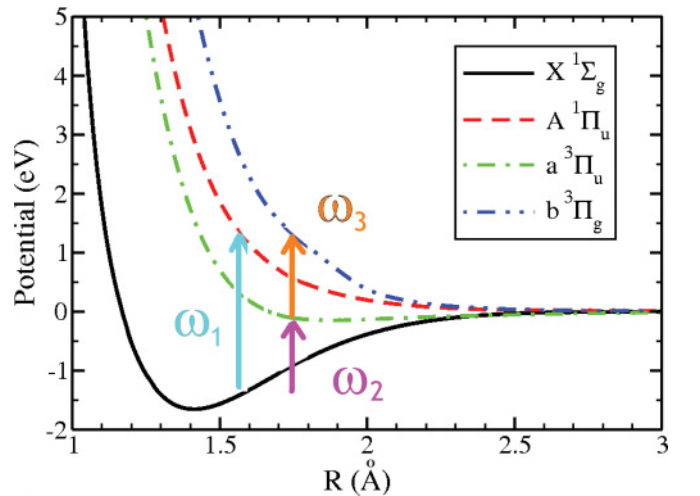


FIG. 2. (Color online) Four potentials involved in the one-photon ω_1 versus two-photon ω_2 and ω_3 photodissociation processes of the F_2 molecule.

of the total $\vec{L} + \vec{S}$ and orbital angular momenta \vec{L} , respectively, on the molecular \hat{e}_z axis. Note that the $\Omega = \Gamma = +1$ states are chosen specifically by applying left circularly polarized light for the ω_1 and ω_2 laser fields; in contrast, the ω_3 field has a linear polarization to define both the space-fixed frame and the body-fixed frame due to the strong field alignment effect in the polarization direction [28]. Therefore, the correlation between our previously defined symmetry-broken states in Eq. (2) and these symmetry-related states can be described as

$$\begin{aligned}
 |E, 1^- \rangle &\xrightarrow{R \rightarrow \infty} |E\rangle \{Y_{1,1}(f)Y_{1,0}(b)\alpha(f)\beta(b)\} \\
 &= |E\rangle \{[\mathcal{O}(u) + \mathcal{O}(g)][\mathcal{S}(u) + \mathcal{S}(g)]\}, \\
 |E, 2^- \rangle &\xrightarrow{R \rightarrow \infty} |E\rangle \{Y_{1,0}(f)Y_{1,1}(b)\beta(f)\alpha(b)\} \\
 &= |E\rangle \{[\mathcal{O}(u) - \mathcal{O}(g)][\mathcal{S}(u) - \mathcal{S}(g)]\}, \\
 |E, 3^- \rangle &\xrightarrow{R \rightarrow \infty} |E\rangle \{Y_{1,1}(f)Y_{1,0}(b)\beta(f)\alpha(b)\} \\
 &= |E\rangle \{[\mathcal{O}(u) + \mathcal{O}(g)][\mathcal{S}(u) - \mathcal{S}(g)]\}, \\
 |E, 4^- \rangle &\xrightarrow{R \rightarrow \infty} |E\rangle \{Y_{1,0}(f)Y_{1,1}(b)\alpha(f)\beta(b)\} \\
 &= |E\rangle \{[\mathcal{O}(u) - \mathcal{O}(g)][\mathcal{S}(u) + \mathcal{S}(g)]\},
 \end{aligned} \tag{9}$$

where $|E\rangle$ is a plane wave of energy E describing the free relative motion of the two nuclei after dissociation. The final dissociative part of the system's wave function in the asymptotic region can thus be expressed as

$$\begin{aligned}
 \psi = \int dE e^{-iEt} \{ &b_{E,1} |E, 1^- \rangle + b_{E,2} |E, 2^- \rangle + b_{E,3} |E, 3^- \rangle \\
 &+ b_{E,4} |E, 4^- \rangle \}.
 \end{aligned} \tag{10}$$

In the absence of (coherent) control, all four dissociative components exist. As a result, as in a W -state case, a measurement of the electronic spin state of the forward-going atom determines only the spin state of backward-going atom. However, we can exercise coherent control on the coefficients of the four dissociative components of Eq. (10) by varying the relative phases and the relative magnitudes of $\Omega_n^{(1)}(E)$ and $\Omega_n^{(2)}(E)$ of Eq. (4),

$$\begin{aligned}
 P_n &= \int dE \{ |\Omega_n^{(1)}|^2 + |\Omega_n^{(2)}|^2 + 2|\Omega_n^{(1)}\Omega_n^{(2)}| \cos(\alpha_n) \}, \\
 n &= 1, \dots, 4,
 \end{aligned} \tag{11}$$

by tuning relative phases α_n . In more detail, P_n can be controlled using two experimental knobs: the relative phase between the laser fields $\phi_1 - \phi_2 - \phi_3$, which directly controls α_n , and the ratio of electric field strengths $\varepsilon_1/\varepsilon_2\varepsilon_3$, which directly controls the $|\Omega_n^{(1)}|/|\Omega_n^{(2)}|$ ratio, as shown in Eq. (4). It follows from Eq. (11) that $\Omega_1^{(m)}$ ($m = 1, 2$) are not completely independent of one another. In fact, $\Omega_1^{(m)} = -\Omega_2^{(m)}$, $\Omega_3^{(m)} = -\Omega_4^{(m)}$, and $\alpha_1 = \alpha_2 = \alpha_3 + \pi = \alpha_4 + \pi$. Hence it follows from Eq. (11) that $P_1 = P_2$ and $P_3 = P_4$.

In spite of the above limitation we can still control the makeup of the spin-orbit GHZ (SOGHZ) states. It follows

from Eq. (11) that for $\alpha_1 = 0$,

$$\begin{aligned}
 |\psi\rangle \rightarrow \psi_A = \int dE e^{-iEt} b_E |E\rangle [&Y_{1,1}(f)Y_{1,0}(b)\alpha(f)\beta(b) \\
 &- Y_{1,0}(f)Y_{1,1}(b)\beta(f)\alpha(b)],
 \end{aligned}$$

and for $\alpha_1 = \pi$,

$$\begin{aligned}
 |\psi\rangle \rightarrow \psi_B = \int dE e^{-iEt} b_E |E\rangle [&Y_{1,0}(f)Y_{1,1}(b)\alpha(f)\beta(b) \\
 &- Y_{1,1}(f)Y_{1,0}(b)\beta(f)\alpha(b)].
 \end{aligned} \tag{12}$$

This is a remarkable result because we generate in this method multiple entanglements in which, contrary to ordinary spin entangled states (e.g., the spin Bell states), where a measurement of the spin of one atom does not reveal the orbital state of either atom, measurement of the spin of one atom yields information of the spin *and* orbital states of *both* atoms.

To demonstrate the controllability that can be attained, we compute the branching ratio for producing different SOGHZ states $(P_1 + P_2)/(P_1 + P_2 + P_3 + P_4)$ for ψ_A and $(P_3 + P_4)/(P_1 + P_2 + P_3 + P_4)$ for ψ_B , as a function of the experimental knobs. We study first the periodic dependence of the branching ratio on α_1 , which is directly determined by $\phi_1 - \phi_2 - \phi_3$, with fixed laser intensities and zero detuning $\Delta_2 = 0$. We note that the two-photon resonance condition $\omega_1 = \omega_2 + \omega_3$ must be satisfied in order to ensure that the two quantum pathways are indistinguishable from one another. Figure 3 depicts a case where $I_1 = 3 \times 10^9$ W/cm², $I_2 = I_3 = 2 \times 10^{11}$ W/cm², and the temporal width is 1 ps. When $\alpha_1 = 0$, due to destructive interference, P_3 and P_4 are suppressed to a minimum, thereby giving rise to the ψ_A entangled state of Eq. (12). When $\alpha_1 = \pi$, P_1 and P_2 are suppressed to a minimum and the ψ_B in Eq. (12) is formed. By tuning α_1 over the $[0 - \pi]$ range, the branching ratio $(P_1 + P_2)/(P_1 + P_2 + P_3 + P_4)$ is seen to vary between 4.6% and 95.4%, an essentially complete controllability.

A different parameter, which is related to the intensities of the optical fields used, is defined as

$$R = S/S_0 \equiv (I_1/I_2I_3)/(I_{10}/I_{20}I_{30}), \tag{13}$$

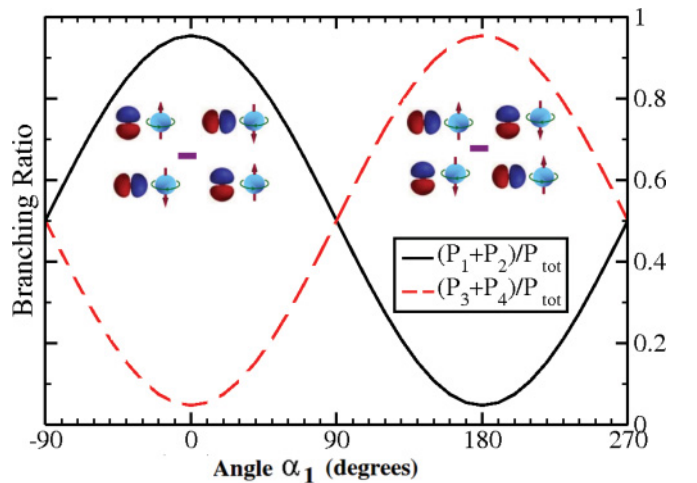


FIG. 3. (Color online) Branching ratios for the multiple-entanglement states ψ_A and ψ_B ; $I_{10} = 3 \times 10^9$ W/cm², $I_{20} = I_{30} = 2 \times 10^{11}$ W/cm², and $\Delta_t = 1$ ps.

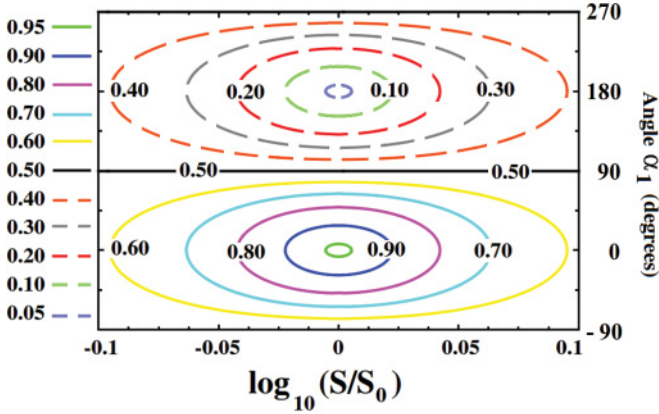


FIG. 4. (Color online) Two-dimensional contour plots for the branching ratios of the ψ_A entangled state; $\log_{10}(S/S_0)$ is defined by Eq. (13).

where I_{10} , I_{20} , and I_{30} are given in Fig. 3. This parameter, which is directly related to the $\Omega_n^{(1)}/\Omega_n^{(2)}$ ratio, describes the strength of the two competing routes. A two-dimensional contour plot of $(P_1 + P_2)/(P_1 + P_2 + P_3 + P_4)$ as a function of α_1 and $\log_{10}(R) = \log_{10}(S/S_0)$ is given in Fig. 4. When $S/S_0 \approx 1$, the two-photon pathway becomes comparable in strength to the one-photon pathway, thus enabling maximal interference and hence the most extensive range of control.

The creation of the above states can be observed experimentally by applying a Stern-Gerlach splitter to the dissociated atoms. If the two pathways are not controlled, four product states result, leading the $j_z (= m_s + m_l)$ quantum numbers to assume the $\pm 1/2$ and $\pm 3/2$ values. However, when the process is controlled by tuning $\phi_1 - \phi_2 - \phi_3$ such that $\alpha_1 = 0$, only two possible j_z values survive, $-1/2$ and $3/2$, leading to the appearance of only two beams in the Stern-Gerlach machine.

A natural extension of the current theory is to form an array of entangled atom pairs in a one-dimensional optical lattice [26,27]. In order to vary the makeup of the entangled states from one site to another an additional control knob must be applied [27]. One option is to vary the molecular phase α_1 by inducing a nonzero detuning Δ_2 with a spatial dependence by ac Stark shifting the energy of the intermediate state $|E_i\rangle$. Using second-order perturbation theory, it is easy to see that when Δ_2 is larger than the energy bandwidth of the laser fields, the phase of $\Omega_n^{(2)}$ is flipped by π , $\Omega_n^{(2)}(\Delta_2 > 0) = -\Omega_n^{(2)}(\Delta_2 < 0)$. The spatial variation of the detuning Δ_2 thus changes α_1 and switches the final dissociation direction from f to b . Changing phases of internal transitions has been proved to be an effective method to control molecular dynamics externally [26,27,29]. The result of such an additional knob is the ability to control the

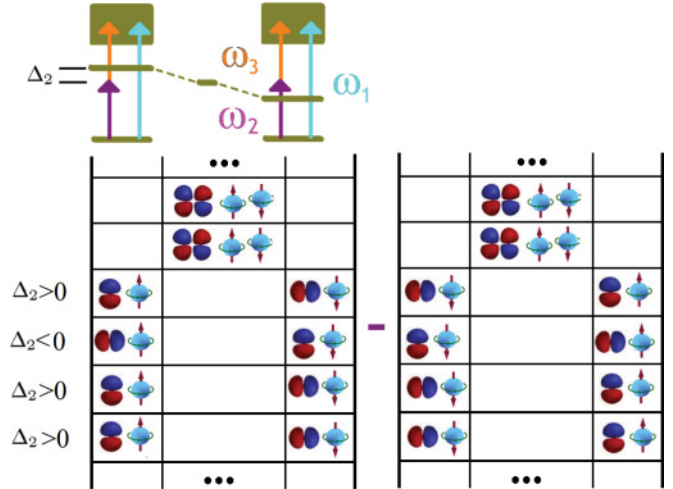


FIG. 5. (Color online) Controlled formation of arrays of entanglement by utilize the ac Stark shift to control Δ_2 and thus α_1 .

makeup of each entangled state in a long sequence of dissociated atom pairs, as shown in Fig. 5.

III. CONCLUSION

In this study we have developed a means to produce multiple-entanglement states in atomic pairs. This states entangle both spin and orbital angular momenta of different atoms. The makeup of these states is controlled by using the interference between one-photon and two-photon pathways. By breaking in this manner the spatial symmetry and using spin-orbit-coupled excited molecular states, we attain a full entanglement of spin and orbital angular momenta and directionality of different atoms. By ac Stark shifting the energy of intermediate states, we can also control the makeup of long chains of atom-pair entangled states in one-dimensional optical lattices.

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