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Quantum-field coherent control: Preparation of broken-symmetry entangled states

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We show that entangled radiation-matter states with *broken symmetries* can be prepared by using nonclassical light in the coherent control techniques. We demonstrate the method by realizing the entanglement in degenerate continuum electronic momentum states of opposite directionality and discrete states of opposite handedness in chiral molecules. When the material system is excited simultaneously by classical light and quantum light in a state with *several* semiclassical phases, the interference conditions guide the system to such entangled radiation-matter states.

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In recent years, intriguing entangled states of light or matter, such as the Greenberger-Horne-Zeilinger (GHZ) or the *W* states, have been suggested and experimentally prepared [1–3]. Hybrid radiation-matter entangled states in atoms [4–6] and states in continua [7] have also been investigated. These fascinating studies have shown among other things that the nonclassicality and entanglement of quantized light and matter fields can be *converted* one to another. Thus, light in squeezed states can prepare entangled atomic states [8] and entangled spin states [9] can give rise to spin squeezed states [10].

It could also be very interesting to consider preparation of entangled quantum states by optical interference techniques, known for their unique selection properties. Coherent control (CC) [11] methodologies can selectively populate *degenerate* quantum states in material systems, by relying on phase-sensitive quantum interference processes, induced by several classical light fields. In recent experiments [12], exceptional spectroscopic results have been obtained when CC techniques were combined with the use of entangled light [13].

Here, we introduce a quantum interference approach, termed "Quantum-field coherent control" (QCC), which is complementary to this new avenue. It could be used to prepare entangled radiation-matter states from degenerate *broken-symmetry* material states. Such discrete or continuous degenerate states cannot be excited individually by single optical transitions. However, since these states lack sharp parities, they could be simultaneously excited by both one-photon and two-photon processes from the same initial state, and addressed *individually* according to the relative phase of the two light fields used [14–16]. In the QCC approach, one of the two light fields is assumed to be *quantized* and prepared in a state with *several* semiclassical phases [13]. Then, a radiation-matter entangled state can be created, where each of its components contains one of the broken-symmetry material states associated with a nonclassical light state of one of the effective phases. Moreover, as in the classical case, the final state can be controlled by the *relative* phase of the classical and nonclassical light sources.

We present the QCC approach first on systems with *continuous* broken-symmetry states excited by two *weak* light fields. In the classical (1+2) CC [14–16], which we quantize below, a (one-dimensional) semiconductor of any crystal symmetry is simultaneously irradiated by two classical light fields, described by the Hamiltonian in the velocity gauge ($H_i \propto \mathbf{A}\cdot\mathbf{p}$) [16]. The first, with the vector potential $A_1 e^{-i\omega_1 t - i\phi_1}$, induces *one-photon* transitions, while the second, of the form $A_2 e^{-i\omega_2 t - i\phi_2}$, induces *two-photon* transitions. Since $\omega_1 = 2\omega_2$, both transitions resonantly link the same initial continuous $|\pm k_v\rangle$ states, of a filled valence band, to the $|\pm k_c\rangle$ states, of an empty conduction band (in vertical transitions, $k_c \approx k_v$).

In the velocity gauge, the population generated in the conduction band is proportional to the square of the sum of the one-photon and two-photon transition amplitudes, given in the rotating wave approximation (RWA) as [14–16] [$p_{ij}(k_c) = \langle k_i|\mathrm{p}|k_j\rangle$ with $i,j=c,v$],

$$P(k_c) \propto \left| A_1 p_{cv} e^{-i\phi_1} + A_2^2 \sum_{i=c,v} \frac{p_{ci} p_{iv}}{E_c - E_i - \hbar\omega_2} e^{-2i\phi_2} \right|^2 \propto A + B\,\mathrm{sgn}(k_c)\cos(\phi_1 - 2\phi_2). \quad (1)$$

Since $E_c - E_v = 2\hbar\omega_2$, the two-photon matrix element is proportional to [16] $-p_{cc}p_{cv}/\hbar\omega_2 + p_{cv}p_{vv}/\hbar\omega_2 = (p_{vv} - p_{cc})p_{cv}/\hbar\omega_2$. Therefore, the product of the one-photon and two-photon matrix elements in the square from Eq. (1) is $\propto |p_{cv}|^2(p_{cc} - p_{vv})$. The intraband p_{cc} and p_{vv} elements, proportional to the velocities of carriers, change sign when k_c changes sign, i.e. $p_{ii}(k_c) = -p_{ii}(-k_c)$, which leads to the $\mathrm{sgn}(k_c)$ factor in Eq. (1). Thus, the injection rates into the $|\pm k_c\rangle$ states can be different, depending on the $\phi_1 - 2\phi_2$ *relative phase* of the two fields, which results in the generation of a dc electric current, $j_c \propto \int k_c P(k_c) dk_c \neq 0$ ($j_v \neq j_c$), in the conduction (valence) band of the semiconductor. In this way, by exciting the states of one directionality more, we can *dynamically* break the system symmetry.

In the (1+2) QCC scenario, schematically shown in Fig. 1, we quantize one of the light fields used and prepare it in a nonclassical state with *several* effective phases. We could use the experimentally feasible squeezed states [17]: in a squeezed vacuum one can assign two effective "phases" mutually shifted by π to two time regions with large quantum fluctuations [13]. Here, we assume that the one-photon process is driven by the "cat (or macroscopic quantum superpo-

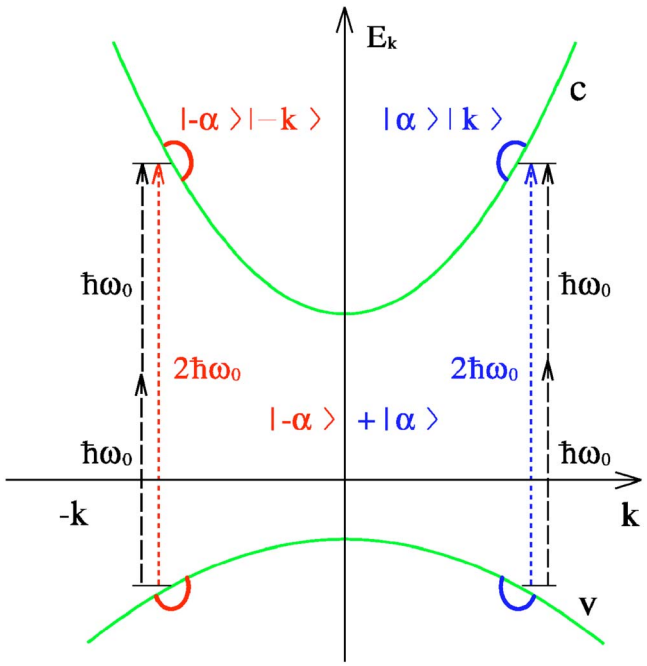

FIG. 1. (Color online) Scheme of the creation of entangled broken-symmetry radiation-matter states in a semiconductor by the QCC method. The states are generated due to quantum-classical interferences of the one-photon light source, using the nonclassical $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ cat state, with two effective phases, and the classical two-photon source.

sition) state" $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ with similar properties [18], which in principle could also be realized experimentally [19]. It is composed of two $|\pm\alpha\rangle$ coherent states, whose *effective phases* are shifted by π.

The probability $P(k_c,\alpha')$ of populating the material eigenstate $|k_c\rangle$ with the light field being in the coherent state $|\alpha'\rangle$ can be evaluated from Eq. (1), where in RWA the one-photon term is changed as [13],

$$A_1 p_{cv} e^{-i\phi_1} \rightarrow \langle\alpha'|\left(\sum_n p_{cv}|n-1\rangle\langle n| + h.c.\right)|\mathcal{C}\rangle. \qquad (2)$$

Choosing, for simplicity, $\alpha\approx\alpha'$ to be real, and using the fact [20] that $\langle n|\alpha'\rangle\propto(\alpha')^n/\sqrt{n}$, we can write it as

$$P(k_c,\alpha') \propto A' + B'\ \mathrm{sgn}(k_c)\mathrm{sgn}(\alpha')\cos(2\phi_2). \qquad (3)$$

Here, the $\mathrm{sgn}(\alpha')$ term derives from the fact that

$$\langle\alpha'|n\pm1\rangle\langle n|\mathcal{C}\rangle = [1+(-1)^n](\alpha')^{n\pm1}\alpha^n/\sqrt{n(n\pm1)}.$$

Hence, only *even* n (*odd* $n\pm1$) terms are nonzero, which all preserve the sign of α'.

The apparently small difference between Eq. (1) and Eq. (3), i.e. the *correlation* of the signs in α' and k_c, has far reaching consequences. By setting the fields such that $A'=B'$ and $\phi_2=0$, we obtain that $P(k_c,\alpha')=0$ if $\mathrm{sgn}(\alpha')\neq\mathrm{sgn}(k_c)$. Therefore, by starting from the initial state of the occupied valence band, $\rho^0_{vv}=\Sigma_{k_v}|k_v\rangle\langle k_v|$, we can prepare entangled states that associate the left and right momenta $|-k_{c,v}\rangle$ and $|k_{c,v}\rangle$ states (currents) with the $|-\alpha\rangle$ and $|\alpha\rangle$ coherent states, respectively. These states can be described by the density operator,

$$\rho_{ii} \approx \sum_{k_i<0} c_{k_i}|k_i\rangle|-\alpha\rangle\langle-\alpha|\langle k_i| + \sum_{k_i>0} c_{k_i}|k_i\rangle|\alpha\rangle\langle\alpha|\langle k_i|,$$

with $i=c(v)$ denoting the conduction (valence) band Bloch electron (hole) states. Since the (1+2) CC can also inject spin currents in semiconductors [21], the QCC could similarly prepare entangled spin currents.

In an analogous way, we can consider hybrid quantum-classical-field photoemission of electrons from discrete atomic or molecular states into a continuum of free-electron momentum states $|\pm k\rangle$. If the one-photon light source is in the $|\mathcal{C}\rangle$ cat (or other) nonclassical state, we can prepare the $|\alpha\rangle|k\rangle+|-\alpha\rangle|-k\rangle$ radiation-matter entangled states. Their preparation could be tested by measuring the light phase and correlating its value with the direction of the outgoing photoemitted electrons.

The presented example demonstrates the QCC method in the *weak-field* limit, where broken-symmetry material states in a *continuum* are entangled with nonclassical radiation states. We now extend the approach to the *strong-field* limit, where we apply the QCC on *discrete* broken-symmetry material states. We apply adiabatic passage methods (AP) [22,23], which can completely transfer populations, and are able to prepare radiative Fock states [24] and entangled material states [25]. The strong-field QCC is based on the combined CC and AP methods [28], which can selectively and completely populate degenerate discrete states of opposite symmetries.

Here, we demonstrate the strong-field QCC in chiral molecules of the left (L) or right (D) handedness (L or D "enantiomers") [26,27]. We can take advantage of the fact that small chiral molecules, such as D_2S_2 [28], can be prepared in their individual broken-symmetry ro-vibronic states and transferred to most of the other states by one-photon processes. We just consider *one* enantiomer, where three of its discrete levels are coupled by light, as shown in Fig. 2 (left). As in Fig. 1, this scheme relies on phase-sensitive interferences between one-photon and two-photon excitation processes [28].

In the excitation, we first resonantly couple the $|2\rangle$ and $|3\rangle$ levels by a "dump" pulse of the Rabi frequency Ω_{23}, from a nonclassical light field. This is followed by two "pump" pulses, starting simultaneously but ending differently, of Rabi frequencies Ω_{12} and Ω_{13}, from two classical light fields. They couple the (initially populated) $|1\rangle$ state to the (empty) $|2\rangle$ and $|3\rangle$ states, respectively. Here, for every pair of i,j levels, $\Omega_{i,j}(t)\equiv\mu_{i,j}\mathcal{E}_{i,j}(t)/\hbar$, with $\mu_{i,j}$ being the transition-dipole elements and $\mathcal{E}_{i,j}(t)$ the related optical fields. Since we excite just one enantiomer, we cannot take advantage of the fact that the signs of $\mu_{i,j}$ depend on the chirality, as in Ref. [28].

If all the three used pulsed fields were classical, the initial population would follow first an adiabatic null state and then *diabatically* cross over to one of the two remaining adiabatic eigenstates [28], as schematically shown by arrows in Fig. 2 (right). The choice of the state would depend on whether the

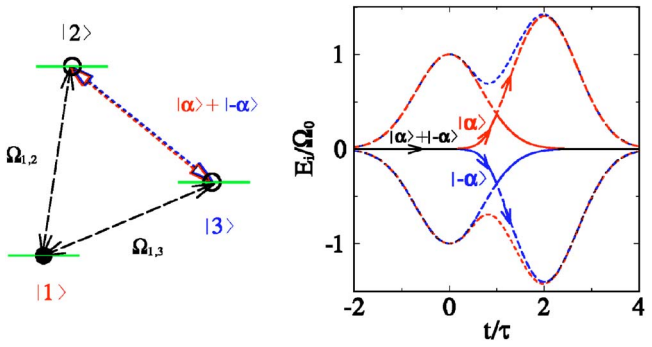

FIG. 2. (Color online) Left panel: The strong-field QCC in three levels of a chosen enantiomer. The pulsed light field coupling the $|2\rangle$ and $|3\rangle$ levels is *quantized* and prepared in the $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ cat state, while the other two pulsed fields that couple the $|1\rangle$ with the $|2\rangle$ and $|3\rangle$ levels are classical. Right panel: Time evolution of the eigenenergies $E_i(i=1-3)$ in this three-level system, coupled by three classical fields [28]. The population follows the $E_1=0$ eigenstate up to the diabatic crossing region, where it switches to one of the other eigenstates (E_2-upper or E_3-lower), depending on whether $\varphi=0$ or π. These two situations are shown by a set of three red and blue curves, respectively. In the QCC, the $\mathcal{E}_{23}$ field is in the nonclassical $|\mathcal{C}\rangle$ cat state, so we cannot define the eigenstates in such a simple way, but we can still schematically draw the diagram. Then the population splits in two parts, where each part follows one of the above classical paths. This leads to the preparation of the entangled $|1\rangle|\alpha\rangle+|3\rangle|-\alpha\rangle$ state.

phase φ of the product $\Omega_{12}\Omega_{23}\Omega_{31}$ of the three Rabi frequencies equals 0 or π.

In contrast, the strong-field QCC is again based on the fact that if a nonclassical $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ cat state is used as the $\mathcal{E}_{23}$ field, each of its two $|\pm\alpha\rangle$ coherent field components, with effective phase shifted by π, drive the system along one of the two classical paths. In Fig. 2 (right), we illustrate this schematically by showing that the population *bifurcates* at the diabatic crossing and half of it follows each of the paths, which is characteristic only of a system allowing for a "cyclic" coupling. As a result, each coherent-state component is entangled with a different eigenstate, and the enantiomer-light system eventually crosses over to the entangled radiation-matter $|1\rangle|\alpha\rangle+|3\rangle|-\alpha\rangle$ state.

We can describe this process quantitatively by invoking a multilevel Jaynes-Cummings model [20]. We use the $|k,n\rangle$ basis, where $|k=1-3\rangle$ are the molecular states of one enantiomer and n denotes the number of photons in the nonclassical dump field. The system wave function,

$$|\Psi(t)\rangle=\sum_{k=1}^{3}\sum_{n=0}^{\infty}b_{k,n}(t)|k,n\rangle, \tag{4}$$

obeys the Schrödinger equation, written in atomic units ($\hbar=1$) as, $d/dt|\Psi(t)\rangle=-iH|\Psi(t)\rangle$. In the RWA, we obtain the following equations for the $b_{k,n}$ coefficients ($k=1-3, n=0,\ldots,\infty$):

$$\dot{b}_{1,n}=-i(\Omega_{12}(t)b_{2,n}+\Omega_{13}(t)b_{3,n}),$$

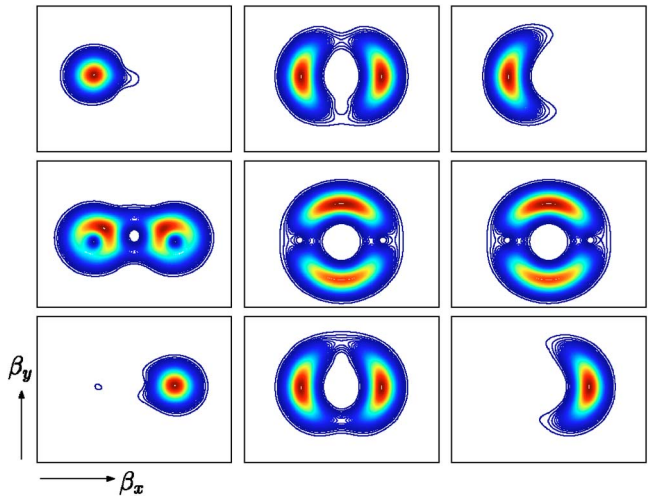

FIG. 3. (Color online) Photon distributions associated with the $|k=1-3\rangle$ states (top-bottom line) of a single enantiomer, presented in the $\Delta\beta_x=\pm6.4$, $\Delta\beta_y=\pm5$ range. Left column: We use the $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ cat state with $|\alpha|^2=7$. The material populations are $p_{1-3}=0.47,0.07,0.46$. The asymmetry of the distributions signals entanglement. Middle column: We use the $|n=7\rangle$ Fock state and do not perform the post-crossing rotation; the populations are $p_{1-3}=0.22,0.61,0.17$. Right column: After executing the post-crossing rotation, the populations remain similar $p_{1-3}=0.2,0.6,0.2$, but the asymmetry in the photon distributions again yields entanglement.

$$\dot{b}_{2,n}=-i(\Omega_{21}(t)b_{1,n}+g_Q(t)\sqrt{n}b_{3,n-1}),$$

$$\dot{b}_{3,n}=-i(\Omega_{31}(t)b_{1,n}+g_Q(t)\sqrt{n+1}b_{2,n+1}). \tag{5}$$

We choose the Rabi frequencies as, $g_Q(t)=g_0f(t)$ with $f(t)=\exp[-t^2/\tau^2]$, $\Omega_{12}(t)=\Omega_0f(t-2\tau)$ and $\Omega_{13}(t)=\Omega_0[f(t-2\tau)+f(t-4\tau)\exp\{-i\Omega_0tf(t-6\tau)\}]$. Notice that $\Omega_{13}(t)$ is prolonged with respect to $\Omega_{12}(t)$ and chirped, as in the classical limit [28]. This guarantees that after the diabatic crossing the level $|2\rangle$ is completely depopulated and the molecular basis is rotated as follows $(|1\rangle+|3\rangle)\rightarrow|1\rangle$ and $(|1\rangle-|3\rangle)\rightarrow|3\rangle$. The adiabatic passage occurs whenever the adiabatic conditions [23,24] $\Omega_0\tau\gg1$ and $2g_0\sqrt{n+1}\tau\gg1$ are met, i.e. if we use $\Omega_0=1$, $g_0=0.1$, $\tau=30$, and $n\approx|\alpha|^2\approx5-10$.

From the numerical solution of Eqs. (5) for $|\Psi(t)\rangle$, we can obtain the density operator $\rho(t)=|\Psi(t)\rangle\langle\Psi(t)|$. The system entanglement can be examined from its diagonal matrix elements $\rho_{k,k}^{\beta,\beta}(t)\equiv\langle k,\beta|\rho(t)|k,\beta\rangle$ $(k=1-3)$ in the (overcomplete) coherent-state $|\beta\rangle$ basis [20], evaluated in terms of the Fock-state matrix elements as

$$\rho_{k,k}^{\beta,\beta}(t)=\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\langle\beta|n\rangle b_k^n(t)(b_k^m(t))^*\langle m|\beta\rangle. \tag{6}$$

We can plot the elements as a function of the real and imaginary parts of β, i.e. $\beta_x=\mathrm{Re}\,\beta$ and $\beta_y=\mathrm{Im}\,\beta$.

In Fig. 3, we present the final photon distributions $\rho_{k,k}^{\beta,\beta}$ in the $|k=1-3\rangle$ molecular states. In the left panels, we use light in the $|\mathcal{C}\rangle=|\alpha\rangle+|-\alpha\rangle$ cat state with $|\alpha|^2=7$, which falls in the above adiabatic regime. The diabatic passage goes along two

different paths, initially giving the $|\Psi\rangle \approx (|1\rangle + |3\rangle)|\alpha\rangle + (|1\rangle - |3\rangle)|-\alpha\rangle = |1\rangle|\mathcal{C}\rangle + |3\rangle|\hat{\mathcal{C}}\rangle$ entangled state, where $|\hat{\mathcal{C}}\rangle = |\alpha\rangle - |-\alpha\rangle$. Thus, when the $|1\rangle \pm |3\rangle \rightarrow |1\rangle, |3\rangle$ or equivalently the $|\mathcal{C}\rangle, |\hat{\mathcal{C}}\rangle \rightarrow |\pm\alpha\rangle$ rotation is performed, the distributions assume extremal values at $\beta \approx \pm\alpha$, where $\rho_{1,1}^{\alpha,\alpha} = \rho_{3,3}^{-\alpha,-\alpha} \neq 0$ and $\rho_{1,1}^{-\alpha,-\alpha} = \rho_{3,3}^{\alpha,\alpha} \approx 0$. Therefore, we obtain the final $|1\rangle|\alpha\rangle + |3\rangle|-\alpha\rangle$ entangled state, while little population is left in the $|2\rangle$ state. The system dynamics can also be controlled by the phases of the two classical fields. For excitation with the $|\mathcal{C}\rangle$ cat state, this would roughly yield entangled classical solutions with phases $\varphi = \varphi_0, \varphi_0 + \pi$. The possibility of such a control is only lost if the nonclassical state has *no* dominant phase.

This is the case of Fock states, shown for $|n=7\rangle$ in the remainder of Fig. 3. In the middle column, calculated without a post-crossing rotation [no chirping of $\mathcal{E}_{13}(t)$], the $|1\rangle$ and $|3\rangle$ levels have practically the same populations and the associated photon distributions have the same shape, since each of the two *diabatic* passages "slices out" about the same "piece" from the Fock state. Here, *adiabatic* passages, that take part at $\varphi = \pi/2, 3\pi/2$ internal phases [28], in addition to the *diabatic* passages around the $\varphi = 0, \pi$ internal phases, cut out a big part of the Fock state and associate it with the $|2\rangle$ level.

The post-crossing rotation is done in Fig. 3 (right). We can see that the populations of the $|1\rangle$ or $|3\rangle$ levels remain the same, but the photon distributions are *mirror images* of one another, signaling radiation-matter entanglement. The resulting light states are similar to "banana-shaped" states and related multiphase $\Sigma_n |e^{i2\pi/n}\alpha\rangle$ coherent states, generated in the Kerr nonlinearity [13]. These states can be tested by measuring the phase of the nonclassical light and correlating it with the luminescence from the excited molecular levels. If the QCC schemes in chiral molecules include also *achiral* higher electronic states [28], we could prepare highly nonclassical $|\Psi\rangle \approx |3\rangle_L|\alpha\rangle + |3\rangle_D|-\alpha\rangle$ entangled states mixing L and D handedness on a single molecule.

The QCC methodology is very promising, since it could be used to prepare entangled states in a number of broken-symmetry systems. The actual experiments might be realized in high-Q microwave cavities with long photon lifetimes [29]. In the strong-field QCC, where the pulsed optical fields do not act *simultaneously* and the system is more susceptible to decoherence, we might prepare the entangled states by exciting low-energy vibrational states in chiral molecules cooled to low temperatures [28].

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