

# Quantum Entanglement via Optical Control of Atom-Atom Interactions

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Two-photon optical transitions combined with long-range dipole-dipole interactions can be used for the coherent manipulation of multiatom collective states. We show that it is possible to induce optical resonances accompanied by the generation of entangled superpositions of such atomic states. Resonances of this kind can be used to implement quantum logic gates using optically excited single atoms (impurities) in the condensed phase.

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Exciting recent developments in the field of quantum information and quantum computing stimulated an intensive search for coherent physical processes which could be used to manipulate coupled quantum-mechanical systems in a prescribed fashion [1]. Although it is clear that so-called universal quantum-mechanical computers are beyond the abilities of current technology, even small-scale devices consisting of few interacting quantum bits are currently of significant fundamental interest.

The present Letter describes a new method for the coherent generation and manipulation of entangled metastable states of interacting pairs of atoms. Our approach is based on the two-photon excitation of resonant transitions modified by a dipole-dipole interaction. This allows one to coherently control the evolution of interacting pairs of atoms and can be used, in particular, for quantum entanglement and conditional quantum logic.

Specifically, in the present Letter (i) we predict the existence of narrow *collective resonances* which correspond to two-photon transitions between multiatom collective metastable states, (ii) we show how these resonances can be exploited for robust generation of quantum entanglement, and (iii) we describe a specific solid state implementation of a quantum computer, involving optically excited impurities or defects with long lived ground-state coherences.

Substantial progress has been made in recent years towards the understanding of coherent interactions of optical fields with multistate atoms [2]. Particularly relevant studies of solid materials [3] and ultracold atoms [4] should be noted. The collective resonances described in the present Letter originate from a process known as dipole-dipole induced “hopping” or transfer of the optical excitation between atoms [5]. Manifestations of such a hopping have been observed, e.g., in a dense thermal vapor [6]. Before proceeding we note that two-atom dark states are the basis for a cavity QED-based quantum logic [7]. We also note very recent quantum logic proposals involving atom-atom interactions in optical lattices [8,9].

Figures 1a and 2a show examples of how dipole coupling in a two-level system can be used for the manipu-

lation of collective metastable states. Here, two distinguishable, stationary atoms (*A* and *B*) are separated by a fixed distance  $r_{AB}$  [10], which is smaller than the optical wavelength  $\lambda$ . Each atom is assumed to have a single excited state that can be coupled to several metastable (or ground) states via electric dipole-allowed transitions. We assume that interactions between atoms can occur only via the coupling of dipoles on selected transitions ( $|a_i\rangle \rightarrow |b_i\rangle$ ,  $i = A, B$  in Figs. 1a and 2a), whereas dipoles on the other transitions (e.g.,  $|a_i\rangle \rightarrow |c_i\rangle$ ) do not interact with other atoms due to differing frequencies or polarizations. In cases when retardation effects can be disregarded, the system can be described by the following effective Hamiltonian:

$$H = \sum_{i=A,B} (H_i^a + V_i^{a-f}) - [\hbar g(\vec{r}_{AB})\sigma_{ab}^A \sigma_{ba}^B + \text{H.c.}], \tag{1}$$

where  $H_i^a$  corresponds to free atoms,  $V_i^{a-f}$  describes the interaction of each atom with components of the electromagnetic field,  $\sigma_{\alpha\beta}^i = |\alpha_i\rangle\langle\beta_i|$  are dipole operators, and  $g(\vec{r}) = \hbar^{-1}\phi_{a\rightarrow b}^A \phi_{a\rightarrow b}^B / r^3 (3z^2/r^2 - 1) = 3/2(2\pi)^{-3} \sqrt{\gamma_{a\rightarrow b}^A \gamma_{a\rightarrow b}^B} \lambda^3 / r^3 (3z^2/r^2 - 1)$  is the coupling constant between interacting dipoles. Here  $\phi_{i\rightarrow j}^k$

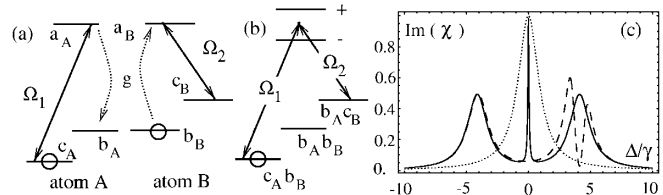


FIG. 1. (a) Schematic energy levels for two-atom Raman transitions. (b) Collective states corresponding to (a). (c) Susceptibility spectra (in arbitrary units) of probe absorption for the system of (a).  $\Delta = \nu_1 - \omega_{ab}^A$  is the detuning of the field  $\Omega_1$  from resonance. The dotted curve corresponds to absorption of a free atom; for the solid curve  $g = 4\gamma$ ,  $\nu_2 = \omega_{ac}^B$ ,  $\omega_{ab}^A = \omega_{ab}^B$ ,  $\Omega_2 = \gamma$ ,  $\gamma_{bc}^k = 0$ ; for the dashed curve  $\nu_2 - \omega_{ac}^B = 4\gamma$ .

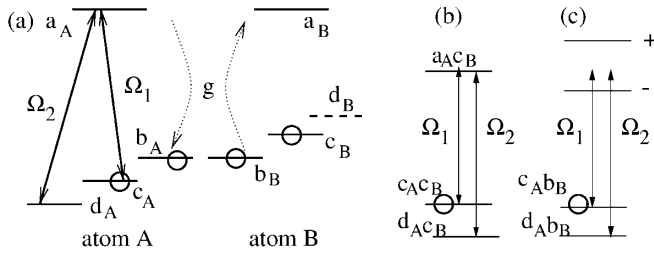


FIG. 2. (a) Schematic energy levels for conditional Raman manipulations. (b),(c) Corresponding two-atom states dressed by the dipole-dipole interaction.

are single-atom dipole matrix elements corresponding to the  $i \rightarrow j$  transition of the  $k$ th atom, and  $\gamma_{i \rightarrow j}^k$  are the corresponding radiative decay rates.

We first consider the case (Fig. 1a) when each of the three-state atoms  $i = A, B$  is coupled by an optical field with a respective Rabi frequency  $\Omega_{1,2}$  ( $\nu_{1,2}$  are the oscillation frequencies). These fields are tuned to resonance with the transitions  $|a_i\rangle \rightarrow |c_i\rangle$  and atoms  $A$  and  $B$  are initially prepared in their lowest metastable states  $c_A$  and  $b_B$ , respectively. This configuration corresponds to a Raman transition between two different atoms which, if excited, will result in the level  $c_A$  being emptied while level  $c_B$  is filled. This transition occurs via the hopping of the optical excitation indicated by dotted lines in Fig. 1a.

To get an insight into the origin of this transition let us consider a subset of the collective energy levels of the two-atom system (Fig. 1b). The dipole-dipole interaction causes splitting of the excited states  $|a_A b_B\rangle$  and  $|b_A a_B\rangle$  into symmetric and antisymmetric superpositions  $|\pm\rangle$ . In the case of equal frequencies on the coupled transitions ( $\omega_{ab}^A = \omega_{ab}^B = \omega$ ) they are

$$|\pm\rangle = \frac{|a_A b_B\rangle \pm |b_A a_B\rangle}{\sqrt{2}}, \quad \omega_{\pm} = \omega \pm |g|. \quad (2)$$

When the splitting is large the resonant optical fields excite selectively a Raman transition from the initial state  $|c_A b_B\rangle$  to the final state  $|b_A c_B\rangle$ .

The effects discussed in the present Letter are quantified by considering the evolution of the many-atom density matrix  $\rho$  [2]:  $\dot{\rho} = L\rho + i\hbar[H, \rho]$ , where the Liouvillian  $L$  describes decoherence processes and dissipation due to, e.g., spontaneous emission.

We first illustrate the optical properties of the two-atom system shown in Fig. 1a by computing the response to a cw driving field  $\Omega_2$  and a weak probe field  $\Omega_1$ . For a moment we disregard cooperative relaxation (assuming that dephasing dominates in the relaxation of the optical coherences) and we find for the steady state expectation value of the induced atomic polarization

$$\langle \sigma_{ac}^A \rangle = i\Omega_1 \frac{\Gamma_{bcab}\Gamma_{bccb} + |\Omega_2|^2}{\Gamma_{ac}(\Gamma_{bcab}\Gamma_{bccb} + |\Omega_2|^2) + g^2\Gamma_{bccb}}. \quad (3)$$

Here  $\Gamma_{ac} = \gamma_{ac}^A + i(\nu_1 - \omega_{ac}^A)$ ,  $\Gamma_{bcab} = \gamma_{bc}^A + \gamma_{ab}^B + i(\nu_1 - \omega_{cb}^A - \omega_{ab}^B)$ , and  $\Gamma_{bccb} = \gamma_{bc}^A + \gamma_{cb}^B + i(\nu_1 - \nu_2 - \omega_{bc}^A - \omega_{cb}^B)$  are complex relaxation rates of the polarization  $\langle \sigma_{ac}^A \rangle$  and the coherences  $\langle \sigma_{bc}^A \sigma_{cb}^B \rangle$  and  $\langle \sigma_{bc}^A \sigma_{cb}^B \rangle$ , respectively.  $\gamma_{ij}^k$  are the coherence decay rates for the respective transitions of the individual atoms. The absorption spectrum corresponding to Eq. (3) is shown in Fig. 1c. Note that if the  $|\pm\rangle$  state splitting is larger than the linewidth of the atomic transitions (solid curve in Fig. 1c where  $\gamma = \gamma_{ab}^k = \gamma_{ac}^k$ ), the narrow two-photon resonance can be easily resolved. This two-photon resonance occurs when the optical difference frequency matches the sum of the two ground-state transition frequencies for atoms  $A$  and  $B$ , ( $\omega_{cb}^A + \omega_{bc}^B$ ). This is the spectroscopic signature of the *coherent collective interaction*.

It is easy to see how selective excitation of these narrow lines can be used for the manipulation of collective metastable states. When a pair of resonant pulses is applied, the amplitude of the state  $|c_A b_B\rangle$  undergoes Rabi oscillations to the state  $|b_A c_B\rangle$  and back with an effective Rabi frequency  $\tilde{\Omega} = \Omega_1 \Omega_2 / g$ . In the limit of large splitting  $|g| \gg \gamma$ , the state vector is thus given by  $|\Psi(t)\rangle = \cos\theta |b_A c_B\rangle + i \sin\theta |c_A b_B\rangle$ , where  $\theta = \int_0^t d\tau \tilde{\Omega}(\tau)$ . The particular case ( $2\theta = \pi/2$ ) corresponds to the generation of a maximally entangled state. Alternatively, by tuning the optical fields close to resonance with either of  $|\pm\rangle$  states, a narrow transparency resonance (dashed curve in Fig. 1c) appears due to the generation of cooperative dark states. Adiabatic following [2] in such a dark state can be used for generation of entanglement and, in particular, for transfer of arbitrary ground-state coherence from atom  $A$  to atom  $B$ . In analogy to cavity-based coherence transfer schemes [7], this process can be generalized to perform quantum logic gates by adding more degrees of freedom to the atom.

Although the entanglement mechanism outlined above would work well for a pair of atoms with well known positions, it is not easy to use it for a prescribed excitation of atom pairs with random spacing: a small uncertainty or variation in the atomic separation  $\vec{r}_{AB}$  will lead to a corresponding change in  $g$  and therefore in  $\tilde{\Omega}$ . We now present an alternate approach, which allows us to avoid this problem and to generate atomic entanglement in a robust way by using *conditional* transitions within a single atom. In this approach the splitting induced by dipole coupling is used to modify the Raman transition rate in just one of the atoms. Here (see Fig. 2a), we provide atoms  $A$  and  $B$  with three or more ground-state sublevels. On atom  $A$  only, we also provide a pair of optical fields  $\Omega_{1,2}$  which are near resonance with the transitions  $|c_A\rangle \rightarrow |a_A\rangle$  and  $|d_A\rangle \rightarrow |a_A\rangle$ , respectively. As before, atom  $A$  can interact with atom  $B$  via dipole-dipole coupling on the  $|a_i\rangle \rightarrow |b_i\rangle$  transitions.

The basic idea is as follows. If atom  $B$  is in the state  $|c_B\rangle$ , then its presence does not affect the two-photon transition within atom  $A$  (see Fig. 2b). However, if atom  $B$  is in the

state  $|b_B\rangle$ , the dipole-dipole coupling causes an effective splitting of the excited state (into  $|\pm\rangle$  states) resulting in a substantial slowing of the two-photon processes (Fig. 2c). Note that since the two-photon process occurs here within one atom, the narrow resonances corresponding, e.g., to Fig. 1 are absent.

To implement quantum entanglement in this system, we consider the process which we term *conditional Raman adiabatic passage*. In this process each of the atoms  $A$  and  $B$  in Fig. 2a is prepared initially in a coherent superposition of the respective states  $|b_{A,B}\rangle$ ,  $|c_{A,B}\rangle$  and sub-levels  $|d_{A,B}\rangle$  are empty. Resonant pulses of the form  $\Omega_1 = \Omega \sin(t/T)$ ,  $\Omega_2 = \Omega \cos(t/T)$  are applied at  $t = 0$  for the duration  $t_d = T\pi$ . In this case, the resonantly coupled subsystem (Fig. 2b) is trapped in a time-dependent “dark” [2] state  $|D\rangle \sim \Omega_2^*(t)|c_{ACB}\rangle - \Omega_1^*(t)|d_{ACB}\rangle$  and therefore adiabatically follows the evolution of the fields. At the same time, in the case of a large splitting corresponding to Fig. 2c, no evolution of the state  $|c_A b_B\rangle$  takes place. In a time  $t_d$ , this pulse sequence effectively transfers the amplitude of state  $|c_{ACB}\rangle$  to state  $|d_{ACB}\rangle$  and back again with the opposite sign. Hence the following transformation results:

$$|c_{ACB}\rangle \rightarrow -|c_{ACB}\rangle, \quad |c_A b_B\rangle \rightarrow |c_A b_B\rangle, \quad (4)$$

$$|b_{ACB}\rangle \rightarrow |b_{ACB}\rangle, \quad |b_A b_B\rangle \rightarrow |b_A b_B\rangle. \quad (5)$$

In the language of quantum information theory, this operation corresponds to a universal logic gate [1].

It is clear that this ideal limit cannot always be achieved for realistic systems. As a relevant figure of merit we compute the minimal fidelity  $F = \min\langle\Psi_f|\rho_f|\Psi_f\rangle$ , where  $\rho_f$  is the actual atomic density matrix after the pulse sequence, and  $|\Psi_f\rangle$  is a target state achieved in the ideal limit. Disregarding dephasing of the lower level coherence during the operation, and assuming  $\omega_{ab}^A = \omega_{ab}^B$ , we find that for  $|g| > \gamma$  the fidelity is given by

$$F \approx \min\left[\exp\left(-\frac{\pi\gamma}{\Omega^2 T}\right), \exp\left(-\frac{\pi\gamma\Omega^2 T}{2|g|^2}\right)\right]. \quad (6)$$

In this expression, the first term arises from imperfect adiabatic passage for the resonant subset of levels (Fig. 2b) and the second corresponds to unwanted transfer for the off-resonant subset (Fig. 2c). It follows that the fidelity is large when  $\Omega^2 T \gg \gamma$  and  $\Omega^2 T \ll |g|^2/\gamma$ , as illustrated in Fig. 3a. This figure also demonstrates [curve (iv)] the effect of lower-state coherence dephasing on fidelity. The correction caused by this dephasing scales (for the two-atom case) as  $\gamma_{bc} T$ .

A very important conclusion that follows from Eq. (6) is that whenever the necessary inequalities are fulfilled, the precise value of the magnitude of the dipole coupling  $g$  is not important to achieve the desired conditional manipulations. Hence, the process of entanglement generation by conditional two-photon transitions is robust with respect to variations and uncertainty in atomic positions.

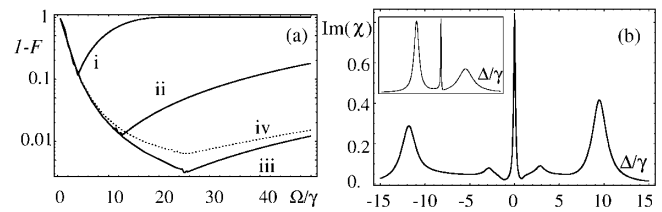


FIG. 3. (a) The minimum fidelity for a phase gate as a function of the drive field Rabi frequency. For curves (i)–(iii)  $\gamma_{bc}^k = 0$ ,  $T = 4\gamma^{-1}$ , and  $g = 10\gamma$ ,  $100\gamma$ ,  $400\gamma$ , respectively. Curve (iv) is computed using parameters characteristic of a pair of N-V atoms confined to a  $\sim 25$  nm region:  $g = 400\gamma$ ,  $\gamma_{bc} = 10^{-3}\gamma$ ,  $\gamma = 10$  MHz. (b) Susceptibility spectra for cooperative Raman transitions in a homogeneous mixture of atoms  $A$  and  $B$  with equal densities.  $N\lambda^3 = 10 \times (2\pi)^2\gamma/\gamma_{\text{rad}}$ .  $\Omega = 3\gamma$ ,  $\gamma_{bc}^k = 0.01\gamma$ . The inset demonstrates the influence of superradiant effects. All optical transitions are assumed to be radiatively broadened with identical decay rates. Other parameters correspond to those of the solid curve in Fig. 1c.

The above analysis disregards the effects of cooperative relaxation such as superradiance. This is justified since the effect of cooperative relaxation on collective two-photon transitions is not significant even if the optical dephasing is dominated by radiative decay. This is illustrated by the inset in Fig. 3b, which shows the two-atom absorption spectrum when cooperative relaxation is taken into account. It is apparent that the narrow collective resonance is not affected. In general, cooperative relaxation effects are dominated by the dipole-dipole interactions in dense small-size atomic samples [11].

It is possible to generalize the above analysis to the case of ensembles of interacting atoms consisting of two groups of atoms. Consider for instance a Raman transition between two homogeneous atomic ensembles in the form of interpenetrating cubic lattices. The absorption spectrum of this medium is shown in Fig. 3b [12]. From this, it is clear that simultaneous many-particle interactions introduce shifts and asymmetry (resulting from effects such as local field correction [13]), as well as additional broad absorption plateaus in between the split peaks. The origin of the latter can be understood as an emerging exciton band. It is important to note, however, that a narrow resonance corresponding to a many-atom Raman transition can still be easily resolved. Pulsed excitation of such resonances would result in highly entangled many-atom superpositions, which are of interest for the realization of ideas such as quantum error correction.

In summary, collective multiphoton resonances should be observable in systems of stationary (frozen) atoms or molecules where the shifts due to the interaction exceed the relevant optical linewidths. Suitable systems that may be used to implement logic gates include impurities in spectral hole burning materials, quantum dots in semiconductors, and cold atoms (in ground or Rydberg states) in small traps and optical lattices. As an example we now describe a possible implementation based on single atoms

(i.e., impurities or defects) in optical spectral hole burning solids. Particular candidate materials include Pr doped  $\text{Y}_2\text{SiO}_5$  (Pr:YSO) [3], color centers such as the N-V center in diamond (N-V) [14] for which single-atom spectroscopy has been performed [15], or Cs in solid helium [16]. The ground state of such materials consists of multiplets of degenerate sublevels. When cooled to liquid helium temperatures, such impurities can display homogeneous optical linewidths which are close to radiative broadening and relatively long lived ground-state coherence lifetimes ( $\sim 0.1$ – $1$  ms [17]).

Because of fluctuations of the crystal field there is a large inhomogeneous distribution of the optical frequencies (up to tens THz). This allows one to use spectral hole burning techniques to select atoms with optical transitions at the desired frequencies. Here qubits can be defined as pairs of ground-state atomic sublevels corresponding to spectrally selected atoms. In the current approach we choose to excite samples of a very small ( $\ll \lambda$ ) size by using, e.g., techniques of near-field microscopy [18]. The defect density can be chosen such that there is one impurity atom per fairly large spectral band  $\Delta_h$ , which is much larger than the homogeneous width but still much smaller than the inhomogeneous width. It is then possible to selectively address single atoms [15] in different parts of the inhomogeneous spectrum; hence independent single-bit operations can be performed.

In order to perform logic gates between a pair of individual impurity atoms  $A$  and  $B$  with known spectroscopic properties, the Zeeman sublevels can be shifted with an H field until one of the optical transitions in atom  $A$  becomes resonant with a transition in atom  $B$ . This is possible since different Zeeman sublevels are shifted by different amounts and sometimes in opposite directions. Zeeman frequency shifts in the above mentioned impurities are such that magnetic fields on the order of few tesla are appropriate. Once such “alignment of levels” is achieved, the two-atom multiphoton transitions described above can be used to perform logic operations on a time scale of  $\sim \gamma^{-1}$  (up to tens of nanoseconds). For realistic experimental parameters, reasonable values of fidelity can be anticipated. Numerical calculations [curve (iv) in Fig. 3a] show that for a pair of impurities with strong optical transitions, such as N-V centers confined to a few tens of nm-sized volume,  $F > 0.99$  is feasible. This number is comparable to the best values estimated for other quantum computation schemes [1,9].

It is important to note the possibilities for scaling the present quantum logic device. The total number of qubits is limited here by the ratio of the total inhomogeneous linewidth to the width of the typical band containing a single spectral hole ( $\Delta_h$ ). The limit to the later arises from the need to suppress errors (phase shifts) caused by off-resonant couplings which scale inversely with  $\Delta_h/\gamma$  [19]. In a typical material a ratio of  $\Delta_h/\gamma \sim 10^3$ – $10^4$

corresponds to about a hundred qubits. Typical values of the lower-level dephasing can, in principle, allow for a few steps of coherent manipulation with a relatively large number of particles. At the same time, it is important to consider the ideas of quantum error correction in the context of the present approach.

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