Coherent Control of Atom-Atom Interactions and Entanglement using Optical Fields

M. D. Lukin\textsuperscript{1} and P. R. Hemmer\textsuperscript{2}

\textsuperscript{1} ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138,
\textsuperscript{2} Sensors Directorate, Air Force Research Laboratory, Hanscom Air Force Base, Bedford, MA 01731

(December 29, 2021)

Abstract

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

PACS numbers: 32.80.Qk,03.67.Lx,42.50.-p
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 well 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 coherent manipulation of many-atom metastable states based on two-photon optical resonances in interacting atomic ensembles. Such collective resonances take place due to the coupling of the optical dipoles on selected, field-free transitions via a dipole-dipole interaction. Even though the mean values of the individual dipole moments involved in such atom-atom interactions vanish, this coupling can result in “hopping” of the excitation from one atom to another. Splitting of collective excited states due to this “hopping” is a dominant feature of the present problem. In such a system it is possible to induce a narrow two-photon transition between two distinct atoms or to perform a two-photon transition within one atom conditional upon the metastable state of the other. When excited with a prescribed pulse sequence these resonances can be used for the generation of entangled superpositions of metastable atomic states and conditional quantum logic. As a specific example we suggest a solid state implementation of a quantum computer, in which individual impurities or defects with long lived ground state coherences can be excited optically.

Substantial progress has been made in recent years towards the understanding of the interactions of optical fields with dense ensembles of multi-state atoms [2]. Particularly relevant studies of solid materials [3] and ultra-cold atoms [4] should be noted. Dipole-dipole induced “hopping” or transfer of the excitation between different atoms has been extensively studied in the past [5] and its manifestations have been observed e.g. in a dense thermal vapor [6]. Recent work utilizing these effects for “nano-optics” [7] might also have interesting implications for the experimental realization of the present ideas. Before proceeding we note that adiabatic following in “dark states” is the basis for a cavity QED-
based quantum logic as described in Ref. [8]. We also note very recent related proposals for quantum logic, based on the precise control of atomic positions in optical lattices [9,10].

Consider a pair of distinguishable multi-state atoms (A and B) at a fixed distance $r_{AB}$ which is smaller than the optical wavelength $\lambda$ (Fig.1a). Each of these atoms is assumed to have a single excited state, which can be coupled to several metastable states via electric dipole-allowed transitions. We assume that the atoms can only interact with each other via the coupling of dipoles on selected transitions ($|a_i\rangle \rightarrow |b_i\rangle$, $i = A, B$ in Fig.1a), whereas all the other dipoles (e.g. $|a_i\rangle \rightarrow |c_i\rangle$) do not couple to those in other atoms due to differing frequencies or polarizations. In the situations considered below, the coupled dipoles are assumed not to be driven by any external electromagnetic field, and we will examine the response of this type of system when optical fields are applied to uncoupled transitions. In cases when retardation effects can be disregarded the system can be described by the following effective Hamiltonian:

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

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

Let us first consider the case when each of the three-state atoms $i = A, B$ in Fig.1a is coupled by an independent optical field with a respective Rabi-frequency $\Omega_{1,2}$ ($\nu_{1,2}$ are oscillation frequencies), as shown. 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 will result in the level $c_A$ being emptied while level $c_B$ is filled. To get an insight into the origin of this transition let us consider collective energy levels of the
two-atom system dressed by coherent driving field $\Omega_2$ (Fig.1b). Three relevant dressed states $|0\rangle, |\pm\rangle$ with energies $\omega_0, \omega_{\pm}$ can be excited by the probe field $\Omega_1$. In the simple case of an exactly resonant driving field $\Omega_2$, close atoms $|g\rangle \gg \Omega_2$ and interacting transitions $a_i \rightarrow b_i$ of equal energies ($\omega_{ab}^A = \omega_{ab}^B$) we find

$$\omega_0 = 0, \quad |0\rangle \approx -\frac{\Omega_2|a_Ab_B\rangle + g|b_Ac_B\rangle}{|g|}$$

$$\omega_{\pm} \approx \pm |g|, \quad |\pm\rangle \approx \frac{|a_Ab_B\rangle \pm |b_Aa_B\rangle}{\sqrt{2}} + O(\frac{|\Omega_2|}{|g|}).$$

The dominant feature of the present problem is splitting of the states $|\pm\rangle$. The third dressed state $|0\rangle$ coincides nearly identically with the metastable two-atom state $|b_Ac_B\rangle$. At the same time it contains a small admixture of the state $|a_Ab_B\rangle$ and therefore can be excited from the initial state $|c_Ab_B\rangle$ by the field $\Omega_1$ resulting in the two-atom Raman transition.

These effects can be quantified by computing the response of the two-atom system assuming 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 expectation value of the induced atomic polarization:

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

Here $\Gamma_{ac} = \gamma_{ac}^A + i(\nu_1 - \omega_{ac}^A)$, $\Gamma_{bcab} = \gamma_{bc}^A + \gamma_{ab}^B + i(\nu_1 - \nu_2 - \omega_{eb}^A - \omega_{ab}^B)$, and $\Gamma_{cbcb} = \gamma_{bc}^A + \gamma_{cb}^B + i(\nu_1 - \nu_2 - \omega_{bc}^A - \omega_{eb}^B)$ are complex relaxation rates of a polarization $\langle \sigma_{ac}^A \rangle$ and coherences $\langle \sigma_{bc}^A\sigma_{ab}^B \rangle$, and $\langle \sigma_{bc}^B\sigma_{ab}^A \rangle$. $\gamma_{ij}^k$ are the coherence decay rates for the respective transitions of the individual atoms. The susceptibility spectrum corresponding to Eq. (2) is shown in Fig.1c. Note that if the $|\pm\rangle$ dressed state splitting caused by the atom-atom interaction $\sim 2|g|$ is larger that the linewidth of the atomic transitions ($\gamma$), the narrow $|c_Ab_B\rangle \rightarrow |0\rangle$ resonance (solid curve in Fig.1c) can be selectively excited without significantly populating the upper states. Excitation of this resonance with pulsed fields results in a a metastable, entangled superposition $|\Psi_{\text{final}}\rangle = \xi_{cb}|c_Ab_B\rangle + \xi_{dc}|b_Ac_B\rangle$. Here $\xi_{ij}$ are complex numbers determined by Rabi frequencies, pulse lengths and magnitude of $g$. 

4
It is easy to generalize the above analysis to the case when the atom \( A \) is initially in coherent superposition \( \ket{A} = \alpha \ket{b_A} + \beta \ket{c_A} \) and \( B \) is in the pure state \( \ket{b_B} \). The two-atom state \( \ket{b_A b_B} \) remains decoupled from the optical fields; hence \( \ket{\Psi_{\text{final}}} = \alpha \ket{b_A b_B} + \beta (\xi_{eb} \ket{c_A b_B} + \xi_{bc} \ket{b_A c_B}) \). The particular case \( (\xi_{eb} \to 0, \xi_{eb} \to 1) \) corresponds to a two-atom Raman \( \pi \) pulse that yields complete coherence transfer. The inset to Fig.1c illustrates Raman-type Rabi oscillations between the metastable states \( \ket{c_A b_B} \) and \( \ket{b_A c_B} \) corresponding to the transfer of coherence \( \sigma_{bc} \) between two atoms.

We next extend the above technique to illustrate how it can be used to perform a conditional manipulation of ground state coherences. To this end (Fig.2a) we provide atoms \( A \) and \( B \) with two or more ground state sublevels that are not coupled to other atoms. On atom \( A \) only, we also provide a pair of optical fields \( \Omega_1, \Omega_2 \) which are near resonance with the uncoupled transitions \( \ket{c_A} \to \ket{a_A} \) and \( \ket{d_A} \to \ket{a_A} \) respectively, forming a resonant Raman system, as shown in Fig.2a. As before, atom \( A \) can interact with atom \( B \) via the dipole-dipole coupling on the \( \ket{a_i} \to \ket{b_i} \) transitions. We assume that the multi-state atom \( B \) is initially prepared in a coherent superposition of the metastable states (e.g. \( \ket{B} = \alpha_B \ket{b_B} + \beta_B \ket{c_B} \)) and the fields do not drive any transitions in this atom. If atom \( B \) is in the state \( \ket{c_B} \), then its presence does not affect the two photon transition within atom \( A \) (see Fig.2b). However, if atom \( B \) is in the state \( \ket{b_B} \) the dipole-dipole coupling causes an effective splitting of the excited state (into \( \ket{\pm} \) states) resulting in a substantial slowing of the two-photon processes (Fig.2c). In such a case, Raman transitions in atom \( A \) can be effectively eliminated by choosing appropriate Rabi-frequencies and pulse durations.

Two particular examples of such interactions are conditional adiabatic passage and conditional Raman excitation with resonant pulses. In the former case, one of the ground states (say \( \ket{d_A} \)) of atom \( A \) must be initially unoccupied. Conditional adiabatic passage can be achieved by applying resonant pulses in the form \( \Omega_1 = \Omega \cos(t/T), \Omega_2 = \Omega \sin(t/T) \) with properly chosen \( \Omega \) and \( T \). In the ideal limit, the state of the system after the transfer \( t = T \pi/2 \) is governed by the operator \( U_{cAP} \):

\[
U_{cAP} = \begin{pmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1 \\
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0
\end{pmatrix}
\]
\[ U_{cAP}(\alpha_B|c_Ab_B\rangle + \beta_B|c_Ac_B\rangle) = \alpha_B|c_Ab_B\rangle + \beta_B|d_Ac_B\rangle. \]

A conditional Raman \(\pi\)-pulse can be achieved by applying identically shaped resonant pulses \(\Omega_1(t) = \bar{\Omega}_1f(t)\), \(\Omega_2(t) = \bar{\Omega}_2f(t)\) with properly chosen intensities and durations \( \int f(t)dt \bar{\Omega} = \pi \). For an arbitrary initial state the ideal limit corresponds to the unitary operation \( U_{c\pi} \):

\[
\begin{align*}
U_{c\pi}|c(d)\rangle_Ac(B\rangle &= \pm \cos \theta |c(d)\rangle_Ac(B\rangle + e^{i\phi} \sin \theta |d(c)\rangle_Ac(B\rangle, \\
U_{c\pi}|c(d)\rangle_Ab(B\rangle &= |c(d)\rangle_Ab(B\rangle,
\end{align*}
\]

where \( \tan \theta = 2|\Omega_1||\Omega_2|/(|\Omega_1|^2 - |\Omega_2|^2) \), \( \bar{\Omega} = \sqrt{|\Omega_1|^2 + |\Omega_2|^2} \) and \( e^{i\phi} = \Omega_1^*\Omega_2/(|\Omega_1||\Omega_2|) \).

As a figure of merit for such processes we compute 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 adiabatic transfer and assuming \( \omega_{ab}^A = \omega_{ab}^B \) we find that for \( |g| > \gamma \) the fidelity of adiabatic passage:

\[
F_{cAP} \approx \min \left[ \exp\left( -\frac{\pi \gamma}{2\bar{\Omega}^2 T} \right), \exp\left( -\frac{\pi \gamma \bar{\Omega}^2 T}{4|g|^2} \right) \right]
\]

In this expression, the first term corresponds to imperfect adiabatic passage and the second corresponds to unwanted transfer. It follows that the fidelity is large when \( \Omega^2 T \gg \gamma \) and \( \Omega^2 T \ll |g|^2/\gamma \) (see Fig.3a). A corresponding fidelity for Raman excitation induced by a pair of rectangular pulses with identical Rabi-frequencies is:

\[
F_{c\pi} \approx \min \left[ \exp\left( -\frac{\pi \gamma}{2\bar{\Omega}} \right), \exp\left( -\frac{\pi \gamma \bar{\Omega}^2}{4|g|^2} \right) \right].
\]

\( F_{c\pi} \) is large when \( \bar{\Omega} \gg \gamma \) and \( \bar{\Omega} \ll |g|^2/\gamma \). It follows from Eqs.(3,4) that whenever the necessary inequalities on pulse durations and Rabi-frequencies are fulfilled precise knowledge of magnitude of the dipole coupling is not required to achieve desired conditional manipulations. This is especially remarkable since the above operations can be used to implement universal logic gates such as a controlled-NOT \([12]\).

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 (see e.g. the inset to Fig.3b which shows the two-atom spectrum when cooperative relaxation is taken into account). In general, cooperative relaxation effects are typically dominated by the dipole-dipole interaction in dense small-size atomic samples [13].

It is possible to generalize the above analysis to the case of ensembles of interacting atoms consisting of two groups of atoms. Fig.3b shows an example of an absorption spectrum corresponding to a Raman transition between two atomic ensembles in the case of a homogeneous medium consisting of atoms A and B with equal densities [14]. It is clear that simultaneous many-particle interactions introduce shifts and asymmetry (resulting from effects such as local field correction [15]) 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, although its magnitude might be reduced.

In summary, collective multiphoton resonances should be observable in systems where the shifts due to the interaction between atoms exceeds the homogeneous optical linewidths and where there are several metastable states available. Systems of that kind include certain solid materials such as spectral hole burning materials and cold atoms in small traps and optical lattices [16].

As an important example we now describe a possible implementation of a quantum logic gates using the above techniques on single atoms (i.e. impurities or defects) in solid crystals. We here take optical spectral hole burning solids as an example. Possible candidate materials include Pr doped Y$_2$SiO$_5$ (Pr:YSO) [18], for which efficient Raman excitation has been realized [3], color centers such as the N-V center in diamond (N-V) [19] for which single-atom spectroscopy have been performed [20], F$_2$ centers in LiF [7] or Cs in solid helium [21]. 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 ($\gamma \sim$ 3 kHz (Pr:YSO) - 20 MHz (N-V),
\[ \gamma_{rad} \sim \gamma/(2...5)) \text{, and relatively long lived ground state coherence lifetimes } ((\pi\gamma_0)^{-1} \sim 1 \text{ ms (Pr:YSO) - 0.1 ms (N-V) } [22]). \]

Due to crystal field fluctuations there is a large \((\Delta_{inh} \sim 5 \text{ GHz (Pr:YSO) - few THz (N-V)) inhomogeneous distribution of the optical frequencies. This allows one to use spectral hole burning techniques to select atoms with optical transitions at desired frequencies. In these systems qbits can be defined as pairs of ground state atomic sublevels corresponding to spectrally selected atoms [17]. All but relevant atoms can be optically pumped into designated storage sublevels. In the current approach we choose to excite samples of a very small \((< \lambda) \text{ size by using near-field microscopy or tightly focused beams, and choose the density such that each spectral hole of interest consists of only one impurity atom. In this case it is possible to selectively address single atoms [20]. A pair of individual impurity atoms A and B can be selectively coupled by shifting the Zeeman sublevels with an H-field until one optical transition of the atom A becomes resonant with another transition of the atom B. 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 quantum logic operations on a time scale of at least } \gamma^{-1}. \text{ By choosing an appropriate concentration of impurities and sample size the excitation and a few steps of coherent manipulation of about hundred of qbits might be feasible.} \]

The authors gratefully acknowledge many useful discussions with M. Fleischhauer, V. Kharchenko and V. Sautenkov. This work was supported by the National Science Foundation.
REFERENCES

[1] See e.g. C. Williams and S. Clearwater, *Explorations in Quantum Computing*, (Springer-Verlag, New-York, 1998); A. M. Stene, Rep. Prog. Phys. 61, 117 (1998).

[2] S. E. Harris, Physics Today, 50(7), 36 (1997).

[3] B. Ham, M. S. Shahriar, and P. Hemmer, Optics Lett., 22, 1138 (1997); *ibid*, 24, 86 (1999).

[4] L. V. Hau, S. Harris, Z. Dutton, and C. Behroozi, Nature 397, 594 (1999).

[5] See e.g. Th. Föster, in *Modern Quantum Chemistry*, O. Sinanoglu Ed., (Academic Press, New-York, 1996).

[6] H. van Kampen *et al.*, Phys.Rev.A 56 3569 (1997); in a thermal vapor large collisional broadening will however tend to wash out the effects of interest here.

[7] S. K. Sekatskii, and V. S. Letokhov, JETP Lett., 65, 465 (1997).

[8] T. Pelizzari, S. A. Gardiner, J. I. Cirac, and P. Zoller, Phys.Rev.Lett. 75, 3788 (1995).

[9] G. Burennen, C.Caves, P. Jessen, and I. Deutsch, Phys. Rev. Lett. 82, 1060 (1999); this implementation utilizes dipole-dipole interaction in a way different from the present approach.

[10] D. Jaksch, H. Briegel, J. I. Cirac, C. W. Gardiner, and P. Zoller, Phys. Rev. Lett. 82, 1975 (1999).

[11] Alternatively, by tuning the optical fields such that dressed states |0⟩ and |±⟩ cross, a narrow transparency resonance (dashed curve in Fig.1c) appears due to the generation of cooperative dark state of the type |Ψ_{final}⟩. Adiabatic passage in this dark state can be also used for generation of entangled states and coherence transfer.

[12] In order to perform quantum logic with conditional adiabatic passage an auxiliary state (|e_A⟩ in Fig.2a) can be used. If this state is initially empty, a conditional swapping
of arbitrary amplitudes in the states $|c_A\rangle$ and $|d_A\rangle$ can be achieved with a three-step transfer sequence $|c_A\rangle \rightarrow |e_A\rangle$, $|d_A\rangle \rightarrow |c_A\rangle$, $|e_A\rangle \rightarrow |d_A\rangle$.

[13] M.Gross, and S. Haroche, Phys.Reports 93, 301 (1982).

[14] This is the result of an analysis based on an $N_A+N_B$ atom master equation, from which a hierarchy of evolution equations for atomic correlations has been obtained. Spectrum in Fig.3b is calculated by making a Gaussian truncation of this hierarchy, and assuming a homogeneous medium. Detailed derivation and discussion of these results will be presented elsewhere.

[15] J. Dowling and C. Bowden, Phys. Rev. Lett. 70, 1421 (1993).

[16] E.g. the present technique can be used to alleviate stringent requirements on the precise location of atoms in quantum logic gates based on optical lattices [9].

[17] Materials pertaining to spectral hole burning have recently been proposed for a cavity-QED implementation of quantum computing along the lines of Ref. [8]; P.Hemmer, M.S.Shahriar, and S. Lloyd (unpublished).

[18] R.W. Equall, R.L.Cone, R.M.Macfarlane Phys.Rev. B 47 14741 (1993).

[19] X.-F. He, N. Manson, and P. Fisk, Phys.Rev.B 47 8809 (1993); A. Lenef at al., ibid, 53 13427 (1996).

[20] A.Gauber et al., Science 276 2012 (1997).

[21] S. I. Kanorsky, S. Lang, T. Eicher, K. Winkler, and A. Weis, Phys.Rev.Lett. 81 401 (1998).

[22] $T_1$s in some of these materials are on the order of minutes or longer. This suggests that it should be possible to significantly lengthen $T_2$ using techniques such as strong applied magnetic bias fields, milliKelvin temperatures, or NMR-based dephasing cancellation.
FIGURES

FIG. 1. (a) Schematic energy levels for two-atom Raman transitions. (b) Collective dressed states corresponding to Fig.1a. (c) Susceptibility spectra (in arbitrary units) of a weak probe field for the two-atom system of Fig.1a. The dotted curve corresponds to absorption of a free atom; for the solid curve $g = 4\gamma, \nu_2 = \omega_{bc}^B, \omega_{ac}^A = \omega_{cb}^B, \Omega_2 = \gamma; \gamma_{bc}^k = 0$; for the dashed curve $\nu_2 - \omega_{ac}^B = 4\gamma$. Inset shows the time evolution of the ground state coherences (i-$\sigma_{bc}^A$, ii-$\sigma_{bc}^B$/i) corresponding to the system of Fig.1a excited at $t = 0$ by a pair of identical fields $\Omega_1 = \Omega_2 = \gamma; g = 20\gamma$.

FIG. 2. (a) Schematic energy levels for conditional two-photon manipulations. (b) Corresponding two-atom states dressed by atom-atom interaction
FIG. 3. (a) The minimum fidelity for conditional adiabatic passage as a function of the drive field Rabi frequency. For curves (i-iii) $\gamma_{bc}^k = 0$, $T = 10\gamma^{-1}$, and $g = 2\gamma, 20\gamma, 200\gamma$ respectively. For curves (iv,v) $\gamma_{bc} = 0.1\gamma$, $g = 20\gamma$ and $T = 10\gamma^{-1}, 0.7\gamma^{-1}$ respectively. (b) Typical susceptibility spectra for cooperative Raman transitions in homogeneous mixture of atoms A and B with equal densities. $N\lambda^3 = 10 \times (2\pi)^2 \gamma/\gamma_{rad}$. $\Omega = 3\gamma$, $\gamma_{bc}^k = 0.01\gamma$. 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.