Coherent Atom Interactions Mediated by Dark-State Polaritons

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We suggest a technique to induce effective, controllable interactions between atoms that is based on Raman scattering into an optical mode propagating with a slow group velocity. The resulting excitation corresponds to the creation of spin-flipped atomic pairs in a way that is analogous to correlated photon emission in optical parametric amplification. The technique can be used for fast generation of entangled atomic ensembles, spin squeezing, and applications in quantum information processing.

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The intriguing possibility for controlled manipulation of interacting quantum systems is the basis for a number of exciting developments in the field of quantum information science [1]. These are expected to have an impact in a broad area ranging from quantum computation and quantum communication [2] to precision measurements [3] and controlled modeling of complex quantum phenomena [4].

This Letter describes a new technique to induce effective coherent interactions between atoms in metastable states. The technique is based on a resonantly enhanced nonlinear process involving Raman scattering into a "slow" optical mode [5], which creates a pair of spin-flipped atoms and slowly propagating coupled excitation of light and matter (dark-state polariton). When the group velocity of the polariton is reduced to zero [6,7], this results in pairs of spin-flipped atoms.

The present phenomenon of spin pair creation exhibits strong similarities with optical parametric amplification (OPA), in which pairs of photons are generated that possess nonclassical correlations in photon number, quadrature component fluctuations, or polarization states [8]. In direct analogy, the present technique is capable of generating nonclassically correlated atomic ensembles and entangled spin excitations. The latter can easily be converted into corresponding states of photon wave packets "on demand," which makes the present approach most suitable for implementing protocols in quantum information processing that require a combination of deterministic sources of entangled states and long-lived quantum memory [9,10].

The present technique can also be viewed as a new mechanism for coherent "collisions" [11] between atoms mediated by light. In particular, the case when atomic pairs are excited into two different levels (as, e.g., in Fig. 1a) closely resembles coherent spin-changing interactions that occur in degenerate atomic samples [12], whereas the case when atomic pairs are stimulated into identical states (Fig. 1b) is reminiscent of dissociation of a molecular condensate [13]. To put this analogy in perspective we note that the rate of the present optically induced process can exceed that of weak interatomic interactions by orders of magnitude. Therefore the present work may open up interesting new possibilities for studying many-body phenomena of strongly interacting atoms.

Before proceeding we note that a number of proposals have been made for generating entangled states of atomic ensembles. Some are based on interatomic interactions at ultracold temperatures [14], whereas others involve mapping the states of nonclassical light fields into atoms [15], quantum nondemolition measurements of spins [16] with light, and Rydberg blockade [17]. Also note the recent experiments on number-phase squeezed states and the Mott insulator phase in Bose-Einstein condensation [18]. In contrast to these mechanisms the present approach does not require coherence of the atomic motion or sources of nonclassical light and is completely deterministic thereby significantly simplifying possible experimental realizations. We further show that the present technique can be made robust with respect to realistic decoherence processes such as spontaneous emission and leakage of slow photons from the medium. Note that the present mechanism does not rely on so-called "photon exchange" interactions discussed by Franson *et al.* [19,20].

We consider a system of N atoms (Fig. 1) interacting with two classical driving fields and one quantized mode that is initially in a vacuum state. Relevant atomic sublevels include two manifolds of metastable states (e.g., hyperfine sublevels of electronic ground state) and excited states that might be accessed by optical transitions. The atoms are initially prepared in their ground states $|g\rangle$. One of the classical fields (Rabi frequency Ω_1) is detuned from

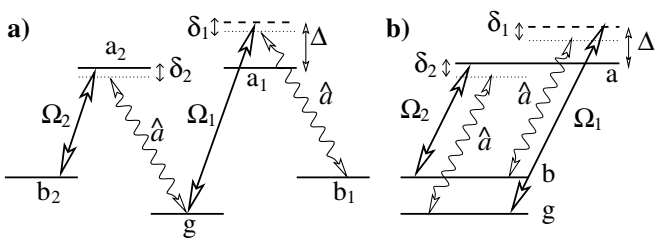

FIG. 1. Level scheme for the coherent interaction leading to pairs of atoms in (a) different final states $|b_2\rangle$ and $|b_1\rangle$, (b) the same final state $|b\rangle$.

the atomic resonance by an amount roughly equal to the frequency splitting between ground state manifolds. The other (Rabi frequency Ω_2) is resonant with an atomic transition $|b_2\rangle \rightarrow |a_2\rangle$. The quantized field can be involved in two Raman transitions corresponding to Stokes and anti-Stokes processes. Whereas the former corresponds to the usual Stokes scattering in the forward direction, the latter establishes an electromagnetically induced transparency (EIT) and slows down its group velocity. The pair excitation can be viewed as resulting from quantized photon exchange between atoms (Fig. 2) in a two-step process. The first flipped spin is created due to Stokes Raman scattering, which also results in photon emission in a corresponding Stokes mode. In the presence of EIT, this photon is directly converted into a dark-state polariton which becomes purely atomic when the group velocity is reduced to zero. This implies that atomic spins are always flipped in pairs.

In what follows we focus on a system (Fig. 1a) involving two atomic modes. Consideration of the scheme of Fig. 1b proceeds along the same lines. For conceptual simplicity we here assume that the quantized field corresponds to a single mode of a running-wave cavity with a creation operator $\hat{a}^\dagger$ and atom-field coupling constants g_1 and g_2. Generalization to multimode, i.e., traveling wave configuration is straightforward. The interaction Hamiltonian for the system of N atoms and light can be split into two parts $H = H_{\rm ram} + H_{\rm res}$, which are given by

$$H_{\rm ram} = -\hbar\Delta\Sigma_{a1a1} - \hbar\delta_1\Sigma_{b_1b_1} + [\hbar\Omega_1\Sigma_{ga1} + \hbar g_1 a^\dagger\Sigma_{b_1a1} + \text{H.c.}], \quad (1)$$

$$H_{\rm res} = \hbar\delta_2\Sigma_{b_2b_2} + \hbar\delta_2\Sigma_{a_2a_2} + [\hbar g_2 a^\dagger\Sigma_{ga2} + \hbar\Omega_2\Sigma_{b_2a2} + \text{H.c.}], \quad (2)$$

where $\Sigma_{\mu\nu} = \sum_i |\mu\rangle_{ii}\langle\nu|$ are collective atomic operators corresponding to transitions between atomic states $|\mu\rangle, |\nu\rangle$ and Δ, δ_1, and δ_2 are single and two-photon detunings as in Fig. 1.

In the limit of large detuning Δ and ignoring two-photon detunings for the moment, the Hamiltonian $H_{\rm ram}$ describes an off-resonant Raman scattering. After a canonical transformation corresponding to adiabatic elimination of the excited state $H_{\rm ram}$ becomes equivalent to

$$H_{\rm ram} = \hbar\sqrt{N}\,\frac{g_1^*\Omega_1}{\Delta}\,aS_1 + \text{H.c.}, \quad (3)$$

where we disregarded the light shift $\delta_L = |\Omega_1|^2/\Delta$ and introduced $S_1 = 1/\sqrt{N}\,\Sigma_{gb_1}$. Light shifts can be easily compensated by redefining the energy of atomic levels and will be disregarded in the remainder of this Letter. The resonant part of the Hamiltonian $H_{\rm res}$ is best analyzed in terms of dark- and bright-state polaritons [21]

$$P_D = \frac{\Omega_2 a - g_2\sqrt{N}\,S_2}{\sqrt{|g_2|^2N + |\Omega_2|^2}}, \qquad P_B = \frac{g_2\sqrt{N}\,a + \Omega_2 S_2}{\sqrt{|g_2|^2N + |\Omega_2|^2}}, \quad (4)$$

which are superpositions of photonic and atomic excitations, with $S_2 = 1/\sqrt{N}\,\Sigma_{gb_2}$. In particular, $H_{\rm res}$ has an important family of dark states:

$$|D^n\rangle \sim (P_D^\dagger)^n|g\rangle\,|\text{vac}\rangle \quad (5)$$

with zero eigenenergies. Note that all other eigenstates of $H_{\rm res}$ have, in general, nonvanishing interaction energy. Under conditions of Raman resonance and sufficiently slow excitation ("adiabatic condition") the Stokes photons emitted by Raman scattering, Eq. (3), will therefore couple solely to the dark states (5). In this case the evolution of the entire system is described by an effective Hamiltonian:

$$H_{\rm eff} = \hbar\xi(P_D S_1 + S_1^\dagger P_D^\dagger), \quad (6)$$

with $\xi = \Omega_1\Omega_2^*/\Delta \times g_1^*\sqrt{N}/\sqrt{|g_2|^2N + |\Omega_2|^2}$. The Hamiltonian (6) describes coherent generation of pairs of excitations involving polaritons P_D and spin-flipped atoms S_1. It is analogous to the "countertwisting" model of Ref. [22] which is known to result in maximal spin squeezing. Note that for a small number of excitations the spin waves and polaritons obey bosonic commutation relations and the Hamiltonian (6) is formally equivalent to that describing OPA [8].

We now consider the scenario in which the system is evolving for a time τ under the Hamiltonian $H_{\rm eff}$, after which both fields are turned off. If the procedure is adiabatic upon turn-off of the coupling fields $\Omega_{1,2}$, the polaritons are converted into pure spin excitations $P_D \rightarrow S_2$. Hence the entire procedure will correspond to the following state of the system:

$$|\Psi\rangle = \frac{1}{\cosh\xi\tau}\sum_n (\tanh\xi\tau)^n\frac{1}{n!}(P_D^\dagger)^n(S_1^\dagger)^n|g\rangle\,|\text{vac}\rangle \rightarrow \frac{1}{\cosh\xi\tau}\sum_n (\tanh\xi\tau)^n|n_{b_1}, n_{b_2}\rangle\,|\text{vac}\rangle. \quad (7)$$

Here $|n_{b_1}, n_{b_2}\rangle = 1/n!\,(S_2^\dagger)^n(S_1^\dagger)^n|g\rangle$ are Dicke-like symmetric states of atomic ensemble and we assumed $n_{b_1,b_2} \ll N$. This entangled state possesses nonclassical properties such as reduced fluctuations as compared to a state representing uncorrelated atoms.

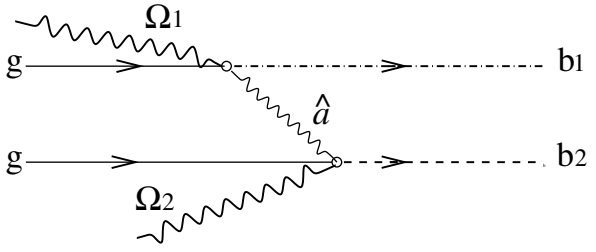

FIG. 2. Diagram illustrating coherent atom-atom interaction mediated by a dark-state polariton, leading to the creation of a pair of spin-flipped atoms.

The above analysis includes only the interaction with a single (forward-propagating) quantized radiation mode and neglects decoherence processes. We now take into account realistic decoherence mechanisms such as spontaneous emission from the excited states in all directions and decay of the cavity mode with a rate κ. The evolution of atomic operators is then described by Heisenberg-Langevin equations:

$$\dot{\Sigma}_{\mu\nu} = -\gamma_{\mu\nu}\Sigma_{\mu\nu} + \frac{i}{\hbar}[H, \Sigma_{\mu\nu}] + F_{\mu\nu}, \qquad (8)$$

where $\gamma_{\mu\nu}$ is a decay rate of coherence $\mu \rightarrow \nu$ and $F_{\mu\nu}$ are associated noise forces. The latter have zero average and are δ correlated with associated diffusion coefficients that can be found using the Einstein relations.

We proceed by adiabatic elimination of optical polarizations associated with Stokes emission. To this end we assume large single-photon detuning $\Delta \gg \gamma$ and to first order in $\hat{a}$ we obtain the following equations of motion for the metastable coherences:

$$\begin{aligned} \dot{S}_1^\dagger &= -[\bar{\gamma}_{gb} + i\delta_1]S_1^\dagger + i\frac{g_1^*\sqrt{N}\,\Omega_1}{\Delta}a + \bar{F}_{S_1}^\dagger(t), \\ \dot{S}_2 &= -[\bar{\gamma}_{gb} + i\delta_2]S_2 - i(\Omega_2/\sqrt{N})\Sigma_{ga_2} + \bar{F}_{S_2}(t), \end{aligned} \qquad (9)$$

where $\bar{\gamma}_{gb} = \gamma_{gb} + \gamma_L$, γ_{gb} is the ground state relaxation rate, and $\gamma_L = \gamma_{ag}|\Omega_1|^2/\Delta^2$ is the optical pumping rate.

To treat the resonant EIT-like interaction we first rewrite the equations of motion in terms of the dark- and bright-polariton operators (4) and proceed to adiabatically eliminate the optical coherence Σ_{ga_2} and the bright-state polariton P_B. In the relevant limit when $|g_2|^2N/\gamma_{ag}\kappa \gg 1$ and when $\eta \equiv |g_2|^2N/|\Omega_2|^2$ the ratio of vacuum light velocity to group velocity is large ($\eta \gg 1$), we find

$$\begin{aligned} \dot{P}_D &= -[\kappa/\eta + \bar{\gamma}_{gb} + i\delta_2]P_D + i\xi S_1^\dagger + \tilde{F}_D(t), \\ \dot{S}_1^\dagger &= \left[\frac{|g_1|^2}{|g_2|^2}\gamma_L - \bar{\gamma}_{gb} - i\delta_1\right]S_1^\dagger \\ &\quad - i\xi P_D + \tilde{F}_{S_1}^\dagger(t). \end{aligned} \qquad (10)$$

We note that cavity losses are strongly suppressed in the limit $\eta \gg 1$: subsequent to the large group velocity reduction [5], the polariton is almost purely atomic and the excitation leaks very slowly out of the medium.

The equation of motion for the coherence $S_1^\dagger$ contains a loss term (due to isotropic spontaneous emission) and a linear gain term (due to emission into a bright polariton) that can compensate each other. However, the linear phase-insensitive amplification is also accompanied by correspondingly increased fluctuations, represented by new Langevin forces $\tilde{F}_D(t), \tilde{F}_{S_1}^\dagger(t)$.

To quantify the resulting quantum correlations we introduce a measure of squeezing in direct analogy to the OPA case: the quadratures are defined as $X_1 = (S_1 + S_1^\dagger)/\sqrt{2}$, $Y_1 = i(S_1 - S_1^\dagger)/\sqrt{2}$. Dynamical evolution leads to the creation of correlated atomic modes and reduced fluctuations in the quadratures of the sum and difference modes $X_\pm = (X_1 \pm X_D)/\sqrt{2}$ and $Y_\pm = (Y_1 \pm Y_D)\sqrt{2}$, and for a small number of excitations these obey standard commutation relations. With the phases of the Rabi frequencies $\Omega_{1,2}$ appropriately chosen, the quadrature Y_+ becomes squeezed, i.e., $\Delta Y_+(t)^2 \leq 1/2$. To detect the resulting correlations established among atoms, the atomic excitations can be converted to photons [6]: in the nondegenerate case (Fig. 1a) twin beams/photons are generated (i.e., two light modes strongly correlated in one quadrature and anticorrelated in the other quadrature) and in the degenerate case a squeezed beam is produced (i.e., one mode with reduced fluctuations in one quadrature and increased fluctuations in the other). For a review of experimental detection of such states, see [23].

We find that squeezing is optimal under conditions of four-photon resonance ($\delta_1 = \delta_2$) and in the limit of $\eta \gg 1$ (Fig. 3). Note that the number of excitations grows exponentially with time (Fig. 3c). Specifically, in the case $g_1 = g_2$, for $\xi t > 1$, we have

$$[\Delta Y_+(t)]^2 = 1/2\left\{e^{-2\xi t} + \frac{2\kappa/\eta + 5\gamma_L + 4\gamma_{gb}}{4\xi} + \left(\frac{\kappa/\eta + \gamma_L}{4\xi}\right)^2 e^{2\xi t}\right\}, \qquad (11)$$

where we have neglected terms of higher order in γ_L/ξ and κ/ξ. The maximum amount of squeezing is obtained after an interaction time t^* such that $e^{-2\xi t^*} = (\gamma_L + \kappa/\eta)/4\xi$ and is given by $\Delta Y_+^2 = (4\kappa/\eta + 7\gamma_L + 4\gamma_{gb})/8\xi$. Since both the interaction parameter ξ and the relaxation rate of the polariton $\gamma_D = \gamma_L + \kappa/\eta$ depend on the single photon detuning Δ (Fig. 3a), we find that

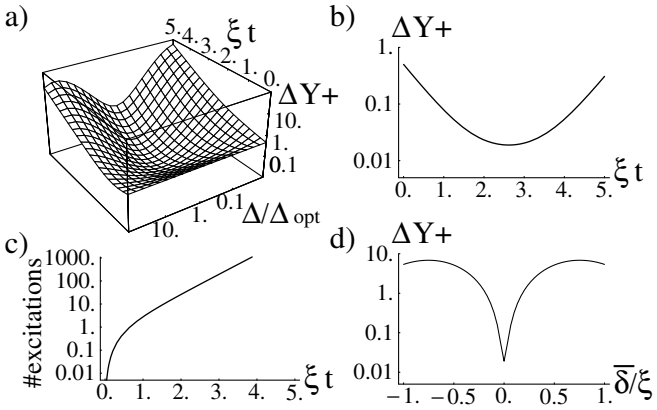

FIG. 3. (a) Quadrature variance ΔY_+^2 vs single-photon detuning Δ and interaction time ξt, (b) same for $\Delta = \Delta_{\text{opt}}$ and $\delta_1 = \delta_2$ showing maximum squeezing $\Delta Y_+^2 \simeq 0.02$ (for $\sqrt{|g_2|^2N/\gamma_{ag}\kappa} = 100$), (c) number of excitations pumped in the system vs time [same conditions as in (b)], and (d) $\Delta Y_+(t^*)^2$ vs two-photon detuning $\bar{\delta} \equiv (\delta_1 - \delta_2)/2$ for $\Delta = \Delta_{\text{opt}}$ and where t^* gives maximum squeezing.

squeezing is optimized for the single-photon detuning $\Delta_{\rm opt} = \gamma_{ag}\sqrt{\frac{7|\Omega_1|^2}{4|\Omega_2|^2}\frac{|g_2|^2N}{\gamma_{ag}\kappa}}/\sqrt{1 + \gamma_{gb}\eta/\kappa}$, and

$$\Delta Y_+^2 = \sqrt{\frac{\gamma_{ag}\kappa}{|g_2|^2N}}\sqrt{\frac{7}{4}\left(1 + \frac{\gamma_{gb}}{\kappa/\eta}\right)}. \qquad (12)$$

The factor $|g_2|^2N/\kappa\gamma$ is equal to the atomic density-length product multiplied by an empty cavity finesse and can easily exceed 10^4 even for modest values of the density-length product and cavity finesse. The coefficient $\gamma_{gb}\eta/\kappa$ is small as long as the effective group delay η/κ is smaller than the ground state relaxation time $1/\gamma_{gb}$, which is easily achievable. Note that the strong coupling regime of cavity QED $g \gtrsim \kappa\gamma$ is *not* required to achieve strong correlations; in fact, as long as $g^2N \gtrsim \kappa\gamma$ squeezing is achieved. Furthermore, although a cavity configuration was used for simplicity, the results of the present analysis remain qualitatively valid in the limit of unity finesse, i.e., free space. We consider a possible implementation of our "degenerate" scheme (Fig. 1b): levels $|g\rangle$ and $|b\rangle$ correspond to the $5^2S_{1/2}, F = 1, m_F = 1$ and $5^2S_{1/2}, F = 2, m_F = 1$ levels in ^{87}Rb (i.e., D_1 line) and level $|a\rangle$ to the $5^2P_{1/2}, F = 2, m_F = 2$ level. With all fields σ^+ polarized and atoms prepared in state $5^2S_{1/2}, F = 1, m_F = 1$ by optical pumping or magnetic state selection in atom traps, this implements the scheme of Fig. 1b. For these conditions the typical generation rate resulting in optimal squeezing $\Omega_1\Omega_2/\Delta_{\rm opt}$ can easily be on the order of a fraction of MHz. In such a case other decoherence mechanisms are negligible. Doppler shifts can also be disregarded as long as all fields are copropagating.

To summarize, we have presented a scheme based on the interaction of coherent classical light with an optically dense ensemble of atoms that leads to effective coherent spin-changing interactions involving pairs of atoms. We have shown that this process is robust with respect to realistic decoherence mechanisms and can result in rapid generation of correlated (spin squeezed) atomic ensembles. Furthermore, the resulting spin excitations can be easily converted into photons on demand, which facilitates applications in quantum information processing. Possible applications involving high-precision measurements in atomic clocks can also be foreseen. We further note that extension of this work into the domain of very large atomic density-length product or high-finesse cavities might allow one to create maximally spin-squeezed states or macroscopic quantum superpositions ("Schrodinger cat" states). This in turn might allow one to observe interaction-induced quantum phase transitions [24].

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