PHYSICAL REVIEW A, VOLUME 65, 041803(R)

Spin squeezing of atoms by the dipole interaction in virtually excited Rydberg states

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(Received 7 May 2001; revised manuscript received 31 August 2001; published 10 April 2002)

We show that the interaction between Rydberg atomic states can provide continuous spin squeezing of atoms with two ground states. The interaction prevents the simultaneous excitation of more than a single atom in the sample to the Rydberg state, and we propose to utilize this blockade effect to realize an effective collective spin Hamiltonian $J_x^2 - J_y^2$. With this Hamiltonian the quantum-mechanical uncertainty of the spin variable $J_x - J_y$ can be significantly reduced.

DOI: 10.1103/PhysRevA.65.041803 PACS number(s): 42.50.−p, 32.80.−t

The sizable dipole interaction between atoms that have been transferred with pulsed laser fields to highly excited Rydberg states has been proposed [1,2] as a mechanism for entanglement operation on the state of neutral atoms. A "Rydberg blockade" effect realized by the dipole interaction prevents more than one atom to enter a Rydberg state at a time. Hence, the evolution of one atom can be conditioned on the state of another one as required for a two-qubit gate in a quantum computer [1]. The Rydberg blockade effect can be used in a multistep procedure to prepare any collective symmetric state of an entire atomic ensemble [2].

In this paper, we propose to use the Rydberg blockade effect in an easier way that uses only continuous laser fields to realize the particular collective states called spin squeezed states. Spin squeezing refers to the collective spin $\vec{J} = \Sigma_i \vec{S}_i$ of a collection of spin 1/2 particles, for which the Heisenberg inequality assures $\Delta J_x \Delta J_y \geq |\langle J_z \rangle|/2$, ($\hbar = 1$). A state whose mean spin is along z and in which the width of the distribution of J_x is reduced so that $\Delta J_x < \sqrt{|\langle J_z \rangle|/2}$ is called spin squeezed. The spin notation represents the state of an ensemble of two-level atoms, where the two states are represented as the $S_z = \pm 1/2$ eigenstates of a spin 1/2 particle. Spin squeezing is a useful property since reduced spin fluctuations imply an improvement of the counting statistics for the number of atoms in specific states, i.e., improved resolution in spectroscopy and in atomic clocks [3,4].

Recently, a number of proposals for spin squeezing and atomic noise reduction has been made involving absorption of broadband squeezed light [5,6], collisional interactions in two-component condensates [7,8], and quantum nondemolition detection of atomic populations [9–11]. In the work presented here, an atomic gas is illuminated with lasers that couple long-lived states $|a\rangle$ and $|b\rangle$ to a Rydberg state $|r\rangle$. The lasers are far detuned so that the population in the Rydberg state is small and their effect is described by an effective Hamiltonian H acting on the states $|a\rangle$ and $|b\rangle$. We first show how nonlinearities appear in the simple case of the lightshift produced by a single laser. The Hamiltonian J_z^2 is realized and squeezing will occur. This Hamiltonian, however, has the drawback that the squeezing axis depends on the interaction time and on the total number of atoms. Thus, we propose a way to realize the Hamiltonian $J_x^2 - J_y^2$ that enables stronger squeezing and which also presents the advantage that the squeezing axis is stationary [12].

Let us consider the situation depicted in Fig. 1, where an ensemble of N atoms is illuminated by a laser field detuned by Δ from resonance of the transition $|a\rangle \to |r\rangle$. If the internal state of the atoms is initially symmetric with respect to exchange of atoms, we can consider only the symetric states and a basis is formed by the states $|n_a, n_r\rangle$, where n_a is the number of atoms in the state $|a\rangle$, n_r is the number of atoms in the state $|r\rangle$, and the remaining $N - n_a - n_r$ atoms populate the state $|b\rangle$. The state $|n_a, 0\rangle$ is coupled with the amplitude $\sqrt{n_a}\Omega$ to $|n_a - 1, 1\rangle$ which, in turn, is coupled to the state $|n_a - 2, 2\rangle$ with the amplitude $\sqrt{2}\sqrt{n_a - 1}\Omega$. If the laser is sufficiently weak, the population in the state with $n_r > 0$ is very small, and the only effect of the laser is to shift the energy of the states $|n_a, 0\rangle$. The expression for the light shift to fourth order in the laser field amplitude is

$$\Delta E_{n_a} = -n_a \frac{\Omega^2}{\Delta} + n_a^2 \frac{\Omega^4}{\Delta^3} - \frac{1}{2\Delta} \frac{2n_a(n_a - 1)\Omega^4}{\Delta^2}, \tag{1}$$

where the last term is due to a two photon transition to the state $|n_a - 2, 2\rangle$. The terms proportional to n_a^2 in ΔE_{n_a} cancel and the light shift is proportional to n_a as expected for non-

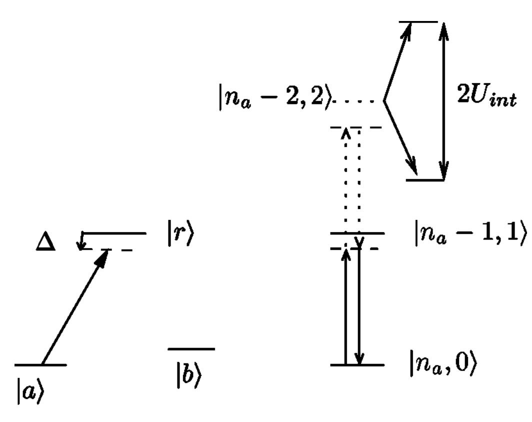

FIG. 1. Laser configuration and relevant states for calculation of the light shift to fourth order in the presence of a single laser. (a) the energy levels of a single atom. (b) the energy levels of a collection of atoms: the upper part of the figure shows how interaction causes an upward or downward shift U_{int} of the state with two Rydberg excited atoms.

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interacting atoms. Indeed, the energy of the state $|a\rangle$ of each atom does not depend on the state of the other atoms. In the picture suggested by Fig. 1(b), the absence of nonlinearities for noninteracting atoms is due to destructive interference between processes involving states with at most one atom in the Rydberg state and processes involving states with several atoms in the Rydberg state.

Let us now assume that atoms in the Rydberg state interact so that the energy of the states $|n_a-2,2\rangle$ is shifted by $\pm U_{int}\gg\Delta$. Then, the two photon contribution to the light shift is negligible and the light shift of $|n_a,0\rangle$ is given by the two first terms of Eq. (1),

$$\Delta E_{n_a} = -n_a\frac{\Omega^2}{\Delta} + n_a^2\frac{\Omega^4}{\Delta^3}. \tag{2}$$

By removing the interference path with more than one atom in the Rydberg state, the"Rydberg Blockade" leads to a nonlinear interaction. Note that the light shift (2) is independent of the precise interaction strength between Rydberg excited atoms. This implies that as long as the interaction is strong enough to substantially increase the detuning, i.e., for atoms with a wide range of spatial separations, the light shift is given by Eq. (2). Writing $n_a=J_z+N/2$, we see that the quadratic light shift in n_a results in an effective Hamiltonian containing a term in J_z^2. Such a Hamiltonian, applied to an initial coherent spin state directed in the (x,y) plane, gives squeezing [12]. The terms linear in J_z in the Hamiltonian are responsible for a rotation of the spin. The addition of a second laser, affecting the atomic state $|b\rangle$, enables us to realize a rotation independent of the number of atoms.

A better Hamiltonian to produce squeezing is

$$H=2\Omega_{\mathrm{eff}}(J_x^2-J_y^2)=\Omega_{\mathrm{eff}}(a^2b^{+2}+b^2a^{+2}). \tag{3}$$

H corresponds to the transfer of atoms to $|b\rangle$ in pairs, and it is thus analogous to the Hamiltonian for production of squeezed light that creates and annihilates photons in pairs. If this Hamiltonian is applied to an ensemble of atoms initially in $|a\rangle$, the spin variance $\langle J_{-\pi/4}^2\rangle=\langle(e^{i\pi/4}a^+b+e^{-i\pi/4}b^+a)^2\rangle$ is reduced. We propose to realize the Hamiltonian (3) in the following way.

As shown in Fig. 2(a), Raman couplings between $|a\rangle$ and $|b\rangle$ are introduced by three laser fields with two Stoke fields, Ω_1 and Ω_2, detuned symmetrically around the Raman resonance by the amount $\pm\Delta'$. The idea is now that a single atom will not make the transition between states $|a\rangle$ and $|b\rangle$ because it is not resonant, but *two* atoms can simultaneously make the transition $|aa\rangle\leftrightarrow|bb\rangle$ since this process occurs resonantly if one atom emits a Stokes photon stimulated by Ω_1 and the other emits a photon stimulated by Ω_2.

Consider two atoms initially in the product state $|aa\rangle$ illuminated by lasers with equal couplings for both atoms as depicted in Fig. 2(b). If the atoms do not interact, there is no way they can exchange the energy mismatch of the stimulated emissions in the fields Ω_1 and Ω_2 and the effective coupling to $|bb\rangle$ vanishes. As in the previous proposal, this can be understood in terms of the destructive interferences between paths involving the state $|rr\rangle$ and the other paths. Figure 2(b) only shows the paths for which stimulated emission in the field Ω_1 occurs first.

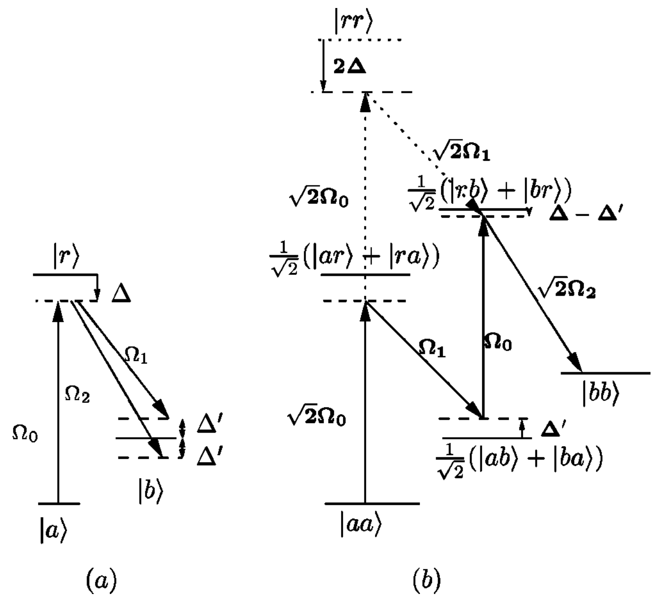

FIG. 2. (a) Energy levels in a single atom and transitions induced by laser fields Ω_i, $i=0,1,2$ to couple the "spin" states $|a\rangle$ and $|b\rangle$ via the intermediate Rydberg state $|r\rangle$. (b) Transition paths transfering two atoms from the state $|aa\rangle$ to the state $|bb\rangle$. The first path (solid lines) does not use the state $|rr\rangle$, the second path (dotted lines) does. If the atoms interact in the state $|rr\rangle$, so that this level is shifted by an amount much larger than Δ, the amplitude of the dotted path becomes negligible, and a net coupling appears from $|aa\rangle$ to $|bb\rangle$.

By contrast, if the interaction between the atoms shifts the energy of the state $|rr\rangle$ by $\pm U_{int}\gg\Delta$, the amplitude of the paths involving the intermediate state $|rr\rangle$ [dotted line in Fig. 2(b)] is suppressed compared to the paths represented by the solid line in the figure, the destructive interference is suppressed, and the coupling between $|aa\rangle$ and $|bb\rangle$ is now

$$\Omega_c=-\frac{4\Omega_0^2\Omega_1\Omega_2}{\Delta(\Delta-\Delta')(\Delta+\Delta')}. \tag{4}$$

A similar four photon transition has been used to entangle ions in ion traps [13,14], where the suppression of destructive interference arises from the Coulomb interaction that lifts the degeneracy of collective vibrational modes. We note that the emergence of a resonant transition due to removal of interfering transition paths also has analogies in spectrocopy on gases, where different mechanisms for pressure induced resonances work by similar mechanism [15,16].

As U_{int} scales as $1/r^3$, the coupling between $|aa\rangle$ and $|bb\rangle$ is given by Eq. (4) as long as the distance between the atoms is smaller than a given critical distance d_0 for which $U_{\mathrm{int}}\gg\Delta$. Thus, in an atomic sample with a size smaller than d_0 the transfer of atoms from $|a\rangle$ to $|b\rangle$ is represented by the squeezing Hamiltonian (3) with $\Omega_{\mathrm{eff}}=\Omega_c/2$. Terms involving more than two atoms at a time would be of higher order in the Rabi frequencies of the lasers and are neglected.

We now turn to an analysis of the time required to obtain substantial spin squeezing. The coupling between states with n_b and n_b+2 atoms transfered to $|b\rangle$ is about the same as the one between harmonic-oscillator number states introduced by the squeezing Hamiltonian $N\Omega_{\rm eff}(b^2+b^{+2})$, as long as n_b is much smaller than the total number of atoms N. Thus, we expect the squeezing to evolve as

$$\langle J^2_{-\pi/4}\rangle(t)=e^{-4N\Omega_{\rm eff}t}\langle J^2_{-\pi/4}\rangle(0) \tag{5}$$

and the mean number of atoms in $|b\rangle$ to follow

$$\overline{n_b}=\sinh^2(2N\Omega_{\rm eff}t). \tag{6}$$

For ease of presentation we introduce the amount of squeezing, $S=(N/4)/\langle J^2_{-\pi/4}\rangle$. Solving numerically the evolution produced by the Hamiltonian (3), we find that these simple analytical expressions are accurate up to 5% as long as $\overline{n_b}<0.05N$ and that the maximum squeezing obtained is about $S\simeq N/2$.

The amplitudes for the excitations of Rydberg states from state $|a\rangle$ and state $|b\rangle$ are proportional to $\sqrt{n_a}\Omega_0$ and $\sqrt{n_b}\Omega_{1,2}$, respectively. Therefore, to justify the elimination of the Rydberg state, the coupling amplitudes should obey

$$\begin{aligned}\sqrt{N}\Omega_0&\ll\Delta,\\ \sqrt{S/4}\Omega_1&\ll\Delta+\Delta',\\ \sqrt{S/4}\,\Omega_2&\ll\Delta-\Delta'.\end{aligned} \tag{7}$$

Here, we used that, for intermediate times so that $1\ll\overline{n_b}\ll N$, $\overline{n_b}\simeq S/4$. Assuming that these inequalities are all fulfilled by an order of magnitude, and taking $\Delta\pm\Delta'\sim\Delta$, the time required to obtain the squeezing S is about

$$T\simeq\frac{1}{16}\frac{10^4}{\Delta}S\ln(S). \tag{8}$$

T is almost linear in the squeezing parameter S, and does not depend on the total number of atoms. The coherence time of the ground states $|a\rangle$ and $|b\rangle$ may be of the order of seconds, and the spin squeezing will be limited only by incoherent effects such as spontaneous emission and thermal field absorption by the small Rydberg state population. To estimate the effect of such incoherent processes, we consider the simple case of loss of atoms, which represents atomic decay to states different from $|a\rangle$ and $|b\rangle$. If the atom i has been lost, the spin variance of the remaining atoms is $\langle J_x'^2\rangle=\langle(J_x-S_{x_i})^2\rangle$ where S_{x_i} is the spin of the lost particle. Due the permutation symmetry of the atomic state, $\langle\Sigma_j S_{x_j}S_{x_i}\rangle=(1/N)\langle J_x^2\rangle$, and thus $\langle J_x'^2\rangle=\langle J_x^2\rangle(1-2/N)+\frac{1}{4}$. After the loss of n_L atoms, we thus have the reduced squeezing

$$S'=\frac{(N-n_L)/4}{\langle J_x'^2\rangle}\simeq S\frac{1}{1+\dfrac{n_LS}{N}}, \tag{9}$$

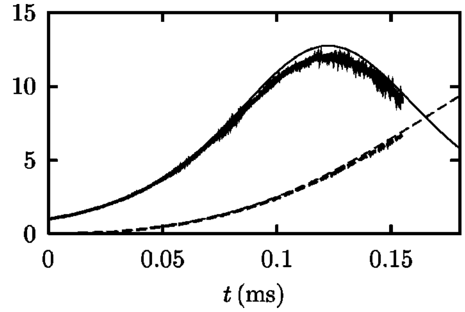

FIG. 3. Evolution of the squeezing factor $S=(N/4)/\langle J^2_{-\pi/4}\rangle$ as a function of time. Thick lines: numerical evolution with six lasers as explained in the text with $\Delta=50$ MHz, $\Delta'=20$ MHz and $|\Omega_0|=|\Omega_1|=|\Omega_2|=1.1$ MHz. Thin lines: evolution according to the Hamiltonian (3). The dashed lines show the evolution of the number of atoms in the state $|b\rangle$ (3).

where S is the value of the squeezing before the losses. To have a negligible effect of losses on the squeezing, we require $n_LS/N\ll1$. The sensiblility of squeezing to losses increases as the squeezing increases as expected since strong squeezing corresponds to strong correlations and entanglement of the particles [17]. With a population of the Rydberg state of about 10^{-2}, which corresponds to the inequalities (7) fulfilled by a factor 10, the expression (8) for the squeezing time implies that the loss rate Γ should obey

$$\frac{\Delta}{\Gamma}\gg\frac{10^2S^2\ln(S)}{16N}. \tag{10}$$

For Rydberg atoms with $n\sim50$, an overestimate for the spontaneous emission rate and the interaction with the black body field yields $\Gamma\sim10$ kHz [2]. Thus, to obtain a squeezing of a factor about 10 with 20 atoms, we require $\Delta\gg6$ MHz. For $\Delta=50$ MHz the interaction energy between the Rydberg atom is much larger than Δ for a distance between atoms $r\ll d_0=3$ μm [2]. To avoid Doppler broadening of Δ it is necessary to use a cold atomic sample. With a density of atoms of 2×10^{11} atoms/cm^3, realized in atomic ensembles obtained from a magneto-optical trap, the number of atoms in a volume of d_0^3 is about 20. Thus, squeezing by a factor about 10 can be obtained.

The coupling introduced by the lasers is well represented by the Hamiltonian (3), but as seen in our first proposal for spin squeezing, the dipole Blockade effect is accompanied by a nonlinearity in the lightshift of the states and the resulting nonlinear terms inhibit the evolution towards states with significant squeezing. To cancel these lightshifts, we propose to add three other lasers of the same intensity but with opposite detuning of the laser fields indicated in Fig. 2(a). These lasers contribute to both the lighshift and the two-atom Raman coupling. If the two added Stokes fields are dephased, respectively, by $+90°$ and $-90°$ with the original ones, the net effect is a vanishing lightshift but a nonvanishing Raman coupling. Figure 3 shows the calculated evolution of 20 atoms illuminated by six lasers with appropriate relative phases. Only states with $n_r<2$ have been taken into account in the calculation since they are the only states relevant in the presence of a strong interaction between Rydberg atoms. The numerical results follow the results of the simple quadratic spin Hamiltonian (3) with a small discrepancy due to even higher-order terms in the lightshift. The maximum squeezing factor is approximatly half of the number of interacting atoms.

It is experimentally relevant to analyze also the case of a macroscopic sample, where the Rydberg blockade is effective only for the n nearest neighbors of a given atom. The Hamiltonian for a large ensemble of atoms can be modeled

$$H=2\sum_{i\neq j}\omega_{ij}(S_{x_i}S_{y_j}+S_{y_i}S_{x_j}),\tag{11}$$

where $\omega_{ij}=\Omega_{\mathrm{eff}}$ if the distance between the atoms i and j is smaller than d_0 and vanishes otherwise. (We have introduced a phase shift of the states $|a\rangle$ and $|b\rangle$ so that the squeezing occurs along y.) The time derivative of $\langle J_y^2\rangle$ is

$$\frac{d}{dt}\langle J_y^2\rangle=-4\sum_{i,j,k}\omega_{ij}\langle S_{y_k}S_{z_i}S_{y_j}+S_{z_i}S_{y_j}S_{y_k}\rangle.\tag{12}$$

Initially, all the atoms are in $|a\rangle$. There is no correlation between atoms and Eq. (12) gives $(d/dt)\langle J_y^2\rangle(t=0)=-4n\Omega_{\mathrm{eff}}\langle J_y^2\rangle(t=0)$. Thus, the initial behavior is similar to that of a small ensemble of n atoms. For later times, numerical simulations in the case of a static ensemble show that to a good approximation, the squeezing evolves similarly to that in an entire ensemble with n atoms all interacting with each other and the maximum squeezing is of the order of $n/2$. If now the atoms move around sufficiently quickly ($v_{rms}\gg 4n\Omega_{\mathrm{eff}}d_0$) so that they are brought constantly in contact with new neighbors with whom they are not entangled, stronger squeezing could be expected. Indeed, in each term of Eq. (12), the atoms i and j are not correlated, and because $\langle S_{y_i}\rangle=0$ and $\langle S_{z_i}\rangle\simeq 1/2$, $\langle J_y^2\rangle$ continues to decrease exponentially as $e^{-4n\Omega_{\mathrm{eff}}t}$ beyond the minimum obtained for n atoms.

In summary, we have proposed a mechanism to produce spin squeezed states of atoms by use of a Rydberg blockade effect induced by cw laser fields. Our calculations show that reduction of the collective spin noise by a factor larger than 10 is possible with current experimental parameters. Note that many other interaction mechanisms may produce a similar blockade of destructive interference. Due to the interference blockade, bichromatically driven quantum transitions via intermediate states with enhanced interparticle interactions, will in general lead to pairwise transitions and nonlinear collective dynamics of the ensemble.

Isabelle Bouchoule thanks the European Community for financial support.

[1] D. Jaksch *et al.*, Phys. Rev. Lett. **85**, 2208 (2000).
[2] M.D. Lukin *et al.*, Phys. Rev. Lett. **87**, 037901 (2001).
[3] D.J. Wineland, J.J. Bollinger, W.M. Itano, and D.J. Heinzen, Phys. Rev. A **50**, 67 (1994).
[4] G. Santarelli *et al.*, Phys. Rev. Lett. **82**, 4619 (1999).
[5] A. Kuzmich, K. Mølmer, and E.S. Polzik, Phys. Rev. Lett. **79**, 4782 (1997).
[6] J. Hald *et al.*, Phys. Rev. Lett. **83**, 1319 (1999).
[7] A. Sørensen *et al.*, Nature (London) **409**, 63 (2001).
[8] U.V. Poulsen and K. Mølmer, Phys. Rev. A **64**, 013616 (2001)
[9] A. Kuzmich, L. Mandel, and N.P. Bigelow, Phys. Rev. Lett. **85**, 1594 (2000).
[10] A. Kuzmich, N.P. Bigelow, and L. Mandel, Europhys. Lett. **42**, 481 (1998).
[11] K. Mølmer, Eur. Phys. J. D **5**, 301 (1999).
[12] M. Kitagawa and M. Ueda, Phys. Rev. A **47**, 5138 (1993).
[13] A. Sørensen and K. Mølmer, Phys. Rev. Lett. **82**, 1971 (1999).
[14] C.A. Sackett *et al.*, Nature (London) **404**, 256 (2000).
[15] G.V. Varada and G.S. Agarwal, Phys. Rev. A **45**, 6721 (1992).
[16] G. Grynberg and P.R. Berman, Phys. Rev. A **41**, 2677 (1990).
[17] A. Sørensen and K. Mølmer, Phys. Rev. Lett. **86**, 4431 (2001).