Spin squeezing of atoms by Rydberg blockade

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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 one 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 reduced significantly.

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The sizable dipole interaction between atoms which 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 requested for a two-qubit gate in a quantum computer [3]. The Rydberg blockade effect can also be used to change the state of an entire atomic ensemble, one atom at a time, and can potentially be used to produce a single atom 'gun' [4].

In this letter, we show that a combination of the Rydberg blockade effect and continuous laser fields can produce particular entangled states of an ensemble of atoms, the so-called spin squeezed states. Spin squeezing refers to the collective spin $\vec{J} = \sum_i \vec{S}_i$ of a collection of spin 1/2 particles, for which the Heisenberg inequality assures $\Delta J_x \Delta J_y \geq |\langle J_x \rangle|/2$, ($h = 1$). A state whose mean spin is along $z$ and in which the width of the distribution of $J_z$ is reduced so that $\Delta J_z \leq \sqrt{\langle J_z^2 \rangle}/2$ is called spin squeezed. The spin notation symbolizes the state of an ensemble of two-level atoms, where the two states are represented as the components $\pm 1/2$ along $z$ of a spin 1/2 particle, and 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 [5].

Recently, a number of proposals for spin squeezing and atomic noise reduction has been made involving absorption of broad band squeezed light [6,7], collisional interactions in two-component condensates [8,9], and quantum non-demolition detection of atomic populations [10,11]. In the work presented here, an atomic gas is illuminated with lasers which couple long lived states $|a\rangle$ and $|b\rangle$ to a Rydberg state $|r\rangle$. The lasers are far detuned so that the 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 non-linearities appear in the simple case of the light-shift produced by a single laser. The hamiltonian $J_x^2$ is realised 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 realise the hamiltonian $J_x^2 - J_y^2$, which 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 $\Delta$ from resonance of the transition $|a\rangle \rightarrow |r\rangle$. If the internal state of the atoms is initially symmetric with respect to exchange of atoms, we can consider only the symmetric 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 $|b\rangle$. 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 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 $\sqrt{2}\sqrt{n_a - 1}\Omega$. The expression of the light shift to fourth order in the laser field amplitude is therefore

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

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 $\Delta E_{n_a}$ cancel and the light shift is proportional to $n_a$ as expected for non 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 non-linearities for non interacting 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):

$$\Delta E_{n_a} = -\frac{n_a \Omega^2}{\Delta} + \frac{n_a^2 \Omega^4}{\Delta^4}. \quad (2)$$
By removing interference paths with more than one atom in the Rydberg state, the “Rydberg Blockade” leads to the non-linear 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. (4). 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 realise a rotation independent of the number of atoms \[^{13}\].

A better hamiltonian to produce squeezing is

$$H = 2\Omega_{\text{eff}} (J_z^2 - J_0^2) = \Omega_{\text{eff}} (a^2 b^* + b^2 a^*)^2.$$  \hspace{1cm}(3)

It corresponds to the transfer of atoms to $|b\rangle$ in pairs, and it is thus analogous to the hamiltonian for production of squeezed light which creates and annihilates photons in pairs. If this hamiltonian is applied to an ensemble of atoms initially in $|\alpha\rangle$, the spin variance $\langle J_z^2 \rangle = \langle (e^{i\pi/4} a + e^{-i\pi/4} b + a^* b^*)^2 \rangle$ is reduced. Furthermore, the best squeezing we can achieve with such a hamiltonian is larger than the one we can achieve with $J_z^2$. We propose to realize the hamiltonian (3) in the following way.

As shown Fig. 2(a), Raman couplings between $|\alpha\rangle$ and $|\beta\rangle$ are introduced by three laser fields with two Stokes fields, $\Omega_1$ and $\Omega_2$, 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 $|\alpha\rangle$ and $|\beta\rangle$ because it is not resonant, but two atoms can simultaneously make the transition $|\alpha\alpha\rangle \leftrightarrow |\beta\beta\rangle$ since this process occurs resonantly if one atom emits a Stokes photon stimulated by $\Omega_1$ and the other emits a photon stimulated by $\Omega_2$.

Consider two atoms initially in the product state $|\alpha\alpha\rangle$ 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 $\Omega_1$ and $\Omega_2$ and the effective coupling to $|\beta\beta\rangle$ vanishes. As in the previous proposal, this can be understood in terms of the destructive interferences between paths involving the state where both atoms are in $|\alpha\rangle \langle \beta\beta|$ and the other paths. Fig. 2(b) shows the paths for which stimulated emission in the field $\Omega_1$ occurs first.

In contrast, if the interaction between the atoms shifts the energy of the state $|\alpha\alpha\rangle$ by $\pm U_{\text{int}} \gg \Delta$, the amplitude of the paths involving the intermediate state $|\alpha\alpha\rangle$ (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 $|\alpha\alpha\rangle$ and $|\alpha\beta\rangle$ is

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

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

\[ |\alpha\rangle \quad |\beta\rangle \quad |\alpha\alpha\rangle \quad |\beta\beta\rangle \quad |\gamma\rangle \quad |\gamma\gamma\rangle \]

\[ \Delta \quad |\gamma\rangle \quad \Delta \quad \Delta' \]

\[ \begin{align*}
|\alpha\rangle & \quad |\beta\rangle \\
|\alpha\alpha\rangle & \quad |\beta\beta\rangle \\
|\gamma\rangle & \quad |\gamma\gamma\rangle
\end{align*} \]

\[ H = 2\Omega_{\text{eff}} (J_z^2 - J_0^2) = \Omega_{\text{eff}} (a^2 b^* + b^2 a^*)^2 \]

\[ \langle J_z^2 \rangle = \langle (e^{i\pi/4} a + e^{-i\pi/4} b + a^* b^*)^2 \rangle \]

\[ \text{Eqs.}(2,3) \]
and $|bb\rangle$ is now

$$\Omega_c = -\frac{4\Omega_1^2\Omega_2}{\Delta(\Delta - \Delta')(\Delta + \Delta')}.$$  \hspace{1cm} (4)

A similar four photon transition has been used to entangle ions in ion traps \cite{14, 15}, where the suppression of destructive interference arises from the Coulomb interaction which 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 spectroscopy on gasses, where different mechanisms for pressure induced resonances work by similar mechanisms \cite{16, 17}.

As $U_{int}$ scales as $1/r^3$, the coupling between $|aa\rangle$ and $|bb\rangle$ is given by \cite{18} as long as the distance between the atoms is smaller than a given critical distance $d_0$ for which $U_{int} \gg \Delta$. 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 \cite{18} with $\Omega_{eff} = \Omega_c/2$.

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 squeezing between states with $n_b$ and $n_b + 2$ atoms transferred to $|b\rangle$ is about the same as the one between harmonic oscillator number states introduced by the squeezing Hamiltonian $N\Omega_{eff}(b^2 + b^{+2})$, as long as $n_b$ is much smaller than the total number of atoms $N$. Thus, we expect the squeezing to evolve as

$$\langle J^2_{\pi/4} \rangle(t) = e^{-4N\Omega_{eff}t} \langle J^2_{\pi/4} \rangle(0)$$  \hspace{1cm} (5)

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

$$n_b = \sinh^2(2N\Omega_{eff}t).$$  \hspace{1cm} (6)

For ease of presentation we introduce the amount of squeezing, $S = (N/4)/\langle J^2_{\pi/4} \rangle$. For intermediate times so that $1 \ll n_b = N$, the squeezing verifies $S \approx 4n_b$. Solvings numerically the evolution produced by the Hamiltonian \cite{18}, we find that these simple analytical expressions are accurate up to $5\%$ as long as $n_b < 0.05N$ and that the maximum squeezing obtained is about $S \approx N/2$.

The amplitudes for the excitations of Rydberg states from state $|a\rangle$ and state $|b\rangle$ are proportional to $\sqrt{n_0}\Omega_0$ and $\sqrt{n_b}\Omega_{1,2}$, respectively. To justify the elimination of the Rydberg state, the coupling amplitudes should therefore obey

$$\begin{cases} \sqrt{N}\Omega_0 \ll \Delta' \\ \sqrt{S/4}\Omega_1 \ll \Delta + \Delta' \\ \sqrt{S/4}\Omega_2 \ll \Delta - \Delta' \end{cases}$$  \hspace{1cm} (7)

Here, we took $S \approx 4n_b$. Assuming that these inequalities are all fulfilled by an order of magnitude, and taking $\Delta'$ and $\Delta$, the time required to obtained the squeezing $S$ is about

$$T \approx \frac{1}{16}\frac{10^4}{\Delta} S \ln(S).$$  \hspace{1cm} (8)

$T$ is almost linear in the squeezing parameter $S$, and does not depend on the total number of atoms. $T$ should be short enough so that incoherent effects such as spontaneous emission or thermal field absorption do not alter the squeezing significantly. To estimate the effect of such incoherent processes, we consider the simple case of loss of atoms, which represents, for example, atoms decaying to a ground state different from $|a\rangle$ and $|b\rangle$ after spontaneous emission. If the atom $i$ has been lost, the spin variance of the remaining atoms is $\langle J^2_x \rangle = (\langle J_x^0 - S_x \rangle)^2$ where $S_x$ is the spin of the lost particle. Due to the permutation symmetry of the atomic state, $\langle \sum_j S_xj S_xj \rangle = \frac{1}{N}\langle J^2_x \rangle$, and we get

$$\langle J^2_x \rangle = \langle J^2_x \rangle (1 - \frac{2}{N}) + \frac{1}{4}.$$  \hspace{1cm} (9)

After the loss of $n_L$ atoms, we thus have the reduced squeezing

$$S' = (N - n_L)/4 \approx S \cdot \frac{1}{1 + n_L/S}$$  \hspace{1cm} (10)

where $S$ is the value of the squeezing before the losses. To have a negligible effect of losses on the squeezing, we require $n_LS/N \ll 1$. The sensibility of squeezing to losses increases as the squeezing increases. This behavior is expected as a strong squeezing corresponds to strong correlations and entanglement of the particles \cite{20}. With a population of the Rydberg state of about $10^{-2}$, which correspond to the inequalities \cite{18} fulfilled by a factor 10, the expression \cite{18} for the squeezing time implies that the atom decay or loss rate $\Gamma$ should obey

$$\frac{\Delta}{\Gamma} \gg \frac{10^2 S \ln(S)}{4}$$  \hspace{1cm} (11)

For Rydberg atoms with $n \sim 50$, an overestimate for the spontaneous emission rate and the interaction with the black body field yields $\Gamma \sim 10 \text{kHz}$ \cite{20}. Thus, to obtain a squeezing of a factor 10, we require $\Delta > 6 \text{MHz}$. For $\Delta = 50 \text{MHz}$ the interaction energy between the Rydberg atom is much larger than $\Delta$ for a distance between atoms $d \leq 3\mu\text{m}$ \cite{20}. With a density of atoms of $2 \times 10^{13}\text{atom/cm}^3$, realised in atomic ensembles obtained from a magneto-optical trap, each atom interacts with about 20 other atoms. Thus, a squeezing by a factor about 10 can be obtained.

The coupling introduced by the lasers is well represented by the Hamiltonian \cite{18}, but as seen in the previous section, the dipole Blockade effect is accompanied by a non linearity in the light shift of the states and the resulting nonlinear terms inhibit the evolution towards
states with significant squeezing. To cancel these light-shifts, 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 lightshift and the two-atom Raman coupling. If the two added Stokes fields are dephased respectively by +90 and −90 degrees with the original ones, the net effect is a vanishing light-shift but a non-vanishing 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, in the presence of a strong interaction between Rydberg atoms, they are the only states relevant. The numerical results follow the results of the simple quadratic spin Hamiltonian \( H \) with a small discrepancy due to even higher order terms in the lightshift. The maximum squeezing factor is within a factor 2 equal to the total number of interacting atoms.

In summary, we have proposed a mechanism to produce spin squeezed states of atoms by use of a Rydberg blockade effect induced by \( \text{cw} \) laser fields. 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 pair-wise transitions and non-linear collective dynamics of the ensemble. For the Rydberg blockade mechanism we have analyzed the allowed range of parameters, and our calculations show that reduction of the collective spin noise by a factor about 10 is possible with current experimental parameters. These results were obtained for the case of an ensemble of size smaller than \( d_0 \) so that the interaction energy between any two atoms in the Rydberg state is sufficient to ensure the blockade effect, \( U_{\text{int}} \gg \Delta \). It is experimentally relevant to analyze the alternative case of
a spatially large sample, where the Rydberg blockade is effective only for the nearest neighbours of a given atom, \( n \ll N \). In that case each atom gets entangled with atoms in its neighbourhood which in turn gets entangled with atoms further away, etc. Simulations of such a coupling show that to a good approximation, the squeezing evolves similar to that in an entire ensemble with \( n \) atoms all interacting with each other\[13\]. The expected amount of squeezing is thus on the order of \( n/2 \) and, squeezing factors around \( S = 10 \) should be realistic in macroscopic samples. If the atoms move around during the illumination, as in a gas, they are in a time integrated sense brought into contact with a larger number of other atoms. If their speeds are large enough to constantly introduce new interacting pairs, i.e., if \( v_{\text{rms}} T >> d_0 \), the squeezing factor will continue to grow as \( e^{4n\Omega_{\text{eff}}t} \) beyond the maximum allowed for \( n \) atoms.

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