Generation of macroscopic singlet states in atomic ensembles

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(Dated: February 12, 2010)

We study squeezing of the spin uncertainties by quantum non-demolition (QND) measurement in non-polarized spin ensembles. Unlike the case of polarized ensembles, the QND measurements can be performed with negligible back-action, which allows, in principle, perfect spin squeezing as quantified by [G. Tóth *et al.*, Phys. Rev. Lett. 99, 250405 (2007)]. The generated spin states approach many-body singlet states, and contain a macroscopic number of entangled particles, even when individual spin is large. We introduce the Gaussian treatment of unpolarized spin states and use it to estimate the achievable spin squeezing for realistic experimental parameters. Our proposal might have applications for magnetometry with a high spatial resolution or quantum memories storing information in decoherence free subspaces.

PACS numbers: 03.65.Ud, 03.67.Mn, 32.80.Qk, 42.50.Dv

I. INTRODUCTION

Realization of large coherent quantum systems exhibiting nonclassical behavior is at the center of attention in many-body quantum experiments with cold atoms [1] and ions [2]. In a system of spin-$\frac{1}{2}$ particles spin squeezing [3, 4] is one of the most successful approaches for creating large scale quantum entanglement [1]. In a spin squeezed state, the collective spin of an ensemble of particles is almost completely set into one direction, while the variance of an orthogonal spin component is decreased below the standard quantum limit. In the special case of spin-1/2 atoms this implies atom-atom entanglement [7] [2]. In a Quantum Non-Demolition (QND) scheme, the particles interact with a light field, which is subsequently measured projecting the atoms into a squeezed state [9, 10]. Typically, the length of the squeezing dynamics is limited by losses, and a short-time approximation can be applied. In this regime, two of the spin components orthogonal to the large mean spin behave like the canonical variables x and p. The Gaussian formalism can then be used for modeling [11], and can include realistic effects of noise and imperfections [12–15, 17].

At this point the questions arise: Is it possible to realize a protocol for the creation of entanglement by squeezing the spin uncertainties that would also work for higher spins? This is important since most experiments use atoms with larger spins. The solution is not easy: Known methods for creating spin-$\frac{1}{2}$ entanglement by spin-squeezing cannot straightforwardly be generalized to higher spins, without restricting dynamics or the detection to a spin-1/2 subspace [18, 19]. Moreover, from the point of view of modeling spin systems, one might ask: Is it possible to extend the Gaussian formalism to unpolarized spin states? This is important if we want to depart from the usual setups with large collective spin.

In this paper, we show how to create and detect entanglement by QND measurement of collective spin in an unpolarized ensemble. We show how to create a many-body singlet state of atoms without requiring that they interact with each other and the system settles in a ground state of some antiferromagnetic Hamiltonian. Our proposal works even for particles with a large spin. With realistic experimental parameters for ^{87}Rb [15], the predicted squeezing dynamics are robust to decoherence, and produce a many-atom singlet state. Unlike standard spin squeezing, the method creates entanglement even in the limit of very strong interaction, which might be used in experimental implementations with cavities [20, 21], or in any multi-atomic system in which a von Neumann measurement of the

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[1] It is instructive to see the relation of spin squeezing to polarization squeezing [5, 6].

[2] For $j > 1/2$, spin squeezing can be also the result of entanglement between the spin-1/2 constituents of the particles, e.g., entanglement between the nuclear and electronic spins [8].

collective spin is possible. We demonstrate the validity of the Gaussian approximation for unpolarized spin states. For the lossless case, we confirm our finding with comparison to the exact model.

The paper is organized as follows. In Section 2, we present the spin squeezing parameter to detect the entanglement of many-body singlet states, and also discuss the properties of the singlets we aim to prepare. In Section 3, we describe the squeezing process. First, we consider the lossless case and present a model based on a Gaussian approximation. Later, we include decoherence in the model. For the lossless case, we compare our results to the results of the exact model. In the Appendix, we present the deltails of the calculations for the exact model.

II. DETECTING THE ENTANGLEMENT OF SINGLET STATES

In this paper, we will use the spin squeezing parameter

$$\xi_s^2 := \frac{(\Delta J_x)^2 + (\Delta J_y)^2 + (\Delta J_z)^2}{J}, \tag{1}$$

where J_l are the components of the collective angular momentum, and for a system of N spin-j particles we define $J := Nj$. It has already been shown in [22–26] that any state giving $\xi_s < 1$ is entangled (i.e., not fully separable). For completeness, we present briefly the proof for (1). For pure product states of the form $|\Psi_N\rangle = \otimes_{k=1}^{N} |\psi_k\rangle$, we have

$$\sum_{l=x,y,z} (\Delta J_l)^2 = \sum_{l=x,y,z} \sum_{k=1}^{N} (\Delta j_l^{(k)})^2_{|\Psi_k\rangle} \geq Nj, \tag{2}$$

where $j_l^{(k)}$ denotes the spin coordinates of particle (k) for $l = x, y, z$. Here we used that $\sum_l (\Delta j_l^{(k)})^2_{|\Psi_k\rangle} \geq j$. For a mixture of pure product states, i.e., for separable states, (2) remains true since the variance is concave in the state.

The states giving $\xi_s = 0$ are called many-body singlet states [27]. In particular, an equal mixture of all pure singlets, expected to arise in permutationally invariant systems, has intriguing entanglement properties [22, 24, 25]. The bipartite entanglement of this state has already been determined for qubits [28]. It is very mixed, yet its entanglement is robust to noise [26]. For qubits, the singlet state studied in this paper is an equal mixture of all states composed of two-qubit singlets as can be seen in figure 1.

For an imperfect realization, $N\xi_s^2$ gives an upper bound on the number of particles unentangled with other particles [24, 25]. This can be seen as follows. Let us consider a pure state of the form $|\Psi_M\rangle = \otimes_{k=1}^{M} |\psi_k\rangle \otimes |\psi_{M+1,\ldots,N}\rangle$, which have M particles unentangled with the rest. For such a state, we have

$$(\Delta J_l)^2 = \sum_{k=1}^{M} (\Delta j_l^{(k)})^2_{|\Psi_k\rangle} + [\Delta(\sum_{k=M+1}^{N} j_l^{(k)})]^2_{|\psi_{M+1,\ldots,N}\rangle}. \tag{3}$$

Hence,

$$(\Delta J_x)^2 + (\Delta J_y)^2 + (\Delta J_z)^2 \geq \sum_{k=1}^{M} (\Delta j_x^{(k)})^2_{|\Psi_k\rangle} + (\Delta j_y^{(k)})^2_{|\Psi_k\rangle} + (\Delta j_z^{(k)})^2_{|\Psi_k\rangle} \geq \frac{M}{2}. \tag{4}$$

Due to the concavity of the variance, mixing pure states with M or more unentangled particles, we still have $N\xi_s^2 \geq M$. If $N\xi_s^2$ for some quantum state is smaller than this bound, then the state cannot be obtained by preparing pure states having at least M unentangled spins and mixing them. If $N(1-\xi_s^2)$ is a large number then we can say that entanglement between macroscopic number of particles is present in the system in this sense.

Next, we will review the relevant properties of singlet states. Many-body singlet states are at the center of attention in condensed matter physics, and also in other areas of quantum physics. They appear as ground states of many fundamental spin models. There have been numerous experiments and experimental proposals for studying such states [29–32]. Surprisingly, the permutationally invariant singlet appears even in quantum gravity calculations of black hole entropy [28]. Its realization is difficult as a $T = 0$ thermal state of a spin system, since in this case the system Hamiltonian is $J_x^2 + J_y^2 + J_z^2$, which is essentially the sum of two-body interactions connecting *all* spin pairs.

Singlets play an important role in quantum information processing. They are invariant under transformations of the form $U_\phi = \exp(-i\phi J_{\vec{n}})$, which describe the effect of an external homogenous magnetic field on the spins and $\vec{n}$ is the direction of the field. ξ_s can directly be related to the decrease of fidelity of a pure state $|\Psi\rangle$ under an external magnetic field, defined as

$$\Delta F := 1 - |\langle\Psi|U_\phi|\Psi\rangle|^2. \tag{5}$$

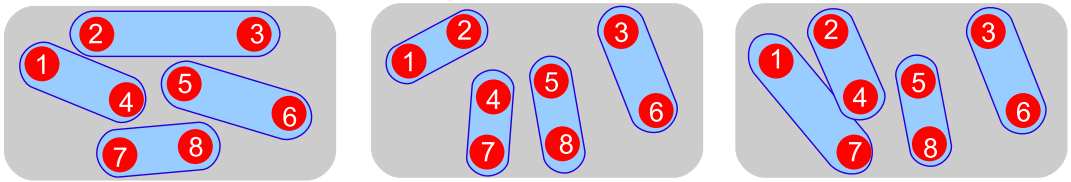

FIG. 1: For qubits, the permutationally invariant singlet is an equal mixture of all possible arrangements of two-particle singlets. Three of such arrangements are shown for eight particles. Note that the eight atoms are arranged in the same way on the figures, only the pairings are different.

Straightforward calculations show that

$$\Delta F \approx (\Delta J_{\vec{n}})^2 \phi^2 \leq \phi^2 J \xi_s^2 \tag{6}$$

for small ϕ [3]. Thus, small ξ_s indicates insensitivity to external fields, which makes singlet states applicable to encoding quantum information in decoherence free subspaces [34], for sending information independent of a reference direction [35], or possibly, for metrological applications in which insensitivity to external homogenous magnetic fields is needed, e.g., measuring the spatial gradient or fluctuations of the fields. Note that measuring field gradients is possible with two almost completely polarized atomic ensembles in an entangled state [36, 37]. A bipartite singlet can also be used for this aim [38]. However, using the singlet of a *single* atomic ensemble can lead to magnetometry with a better spatial resolution.

III. SQUEEZING OF THE COLLECTIVE SPIN UNCERTAINTIES

A. Outline of the squeezing procedure

In this paper, we consider a procedure to produce an atomic state with $\xi_s < 1$ from a non-entangled state with $\xi_s \sim 1$. For the initial state and throughout the dynamics we have $\langle \mathbf{J} \rangle = 0$. Suitable initial states include the thermal state (completely mixed state), for which $\xi_s = \sqrt{j+1}$, and pure product states of the form $|\Psi\rangle = \otimes_k |\psi_k\rangle$, with $|\langle \vec{j} \rangle_{\psi_k}| = j$ for which $\xi_s = 1$. Spin coherent states also have $\xi_s = 1$, however, for them $\langle \mathbf{J} \rangle \neq 0$. After the initial state was created, ξ_s is reduced by sequentially measuring and if necessary correcting by feedback the spin components.

Next, we will examine the dynamics of the expectation values and variances of collective spin operators and we will determine how they depend on the particle number N. In particular, $A \sim N^p$ will denote that the quantity A is proportional to N^p for large N.

The first squeezing step is a QND measurement of the atomic spin component J_x. The realization is similar to QND measurements in polarized atomic ensembles [9, 10]. The atoms interact with light, which is subsequently measured. As discussed later, this procedure can reduce the value of $(\Delta J_x)^2$ considerably but with only a minor increase in $(\Delta J_y)^2$ and $(\Delta J_z)^2$. However, the procedure results in nonzero $\langle J_x \rangle$. Fortunately, the expectation value of the spin component J_x remains small and we obtain $|\langle J_x \rangle| \lesssim \sqrt{N}$. Feedback, using the data coming from the measurement of the light can then be applied to restore $\langle J_x \rangle = 0$. This feedback step is similar to the coherent rotation in polarized spin-squeezing [39, 40], but does not require a large average polarization. However, it is now not simply a rotation, but an incoherent process. The feedback introduces negligible noise: It can be shown that for making $\langle J_x \rangle$ zero, $\sim \sqrt{N}$ atoms are affected, introducing extra noise $(\Delta J_x)^2 \sim \sqrt{N}$, which is negligible in the large N limit [4]. Alternately, instead of feedback, post-selection could be used to identify cases with low $\langle J_x \rangle$.

If the initial state was a pure product state of the type mentioned above, measurement of J_x is sufficient to produce a squeezed state with $\xi_s < 1$. For the thermal state, further squeezing steps are needed for J_y and J_z. Since $\langle \mathbf{J} \rangle = 0$,

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[3] For mixed states, $4J\xi_s^2 = 4\sum_l (\Delta J_l)^2$ bounds from above the Quantum Fisher Information $F_Q[\rho, J_{\vec{n}}]$, which gives the maximum phase estimation sensitivity in an interferometer using the quantum state ρ and unitary dynamics $\exp(-iJ_{\vec{n}}t)$. Here we used that $F_Q[\rho, J_{\vec{n}}] \leq 4(\Delta J_{\vec{n}})^2$ [33].

[4] Scattering sets the two-body correlations $\langle j_l^{(m)} j_l^{(n)} \rangle$ to zero, where m is one of the spins that went through scattering, while n in one of the non-affected spins. From $\langle J_l^2 \rangle \geq 0$, a lower bound on the average two-point correlations is $\sim -\frac{1}{N}$. Since $\sim \sqrt{N}$ atoms are affected by scattering, the number of two-point correlations set to zero by scattering is $\sim N\sqrt{N}$. Thus, the increase of $(\Delta J_l)^2$ is at most $\sim \sqrt{N}$.

the generalized uncertainty relations $(\Delta J_k)^2(\Delta J_l)^2 \geq \frac{\hbar^2}{4}\langle J_m\rangle^2$ do not enforce a measurement back-action. As such, the QND measurements can produce a state with $(\Delta J_x)^2, (\Delta J_y)^2, (\Delta J_z)^2$ all significantly reduced so that $\xi_s^2 \ll 1$.

B. Lossless case

We now describe the details of our calculations. We employ methods developed for treating the Gaussian dynamics of continuous variable systems [9–11, 14], which we adapt to the case of unpolarized ensembles of spin-j particles. First, the QND measurement is modeled in the absence of decoherence, which allows comparison with an exact calculation. Then, we incorporate decoherence due to light scattering [13, 44, 45].

For modeling the QND pulse, the atoms are described by the J_l operators, while the light pulse is characterized by the Stokes operators S_l [13, 17]. We choose the initial state to be the completely mixed atomic state, $\varrho_0 := \frac{1}{(2j+1)^N}\mathbb{1}$ and a fully-polarized optical state with $\langle \mathbf{S}\rangle = (S_0, 0, 0)$. The full system is described by the operators

$$R = \left\{ \tfrac{J_x}{\sqrt{J}}, \tfrac{J_y}{\sqrt{J}}, \tfrac{J_z}{\sqrt{J}}, \tfrac{S_x}{\sqrt{S_0}}, \tfrac{S_y}{\sqrt{S_0}}, \tfrac{S_z}{\sqrt{S_0}} \right\} \tag{7}$$

with a covariance matrix

$$\Gamma_{mn} := \tfrac{1}{2}\langle R_m R_n + R_n R_m\rangle - \langle R_m\rangle\langle R_n\rangle. \tag{8}$$

As shown by simple calculations, for large N the initial state is Gaussian for the R_k operators. That is, symmetric moments with order higher than second can be obtained from lower order ones according to the theory of Gaussian distributions, knowing that cumulants with order three and higher are zero [41]. In other words, concerning the moments of R_k, the state is completely characterized by Γ, $\langle \mathbf{S}\rangle$ and $\langle \mathbf{J}\rangle$.

The first step of the QND measurement of J_x is interaction between the atoms and the light via the Hamiltonian

$$H = \hbar\Omega J_x S_z. \tag{9}$$

This suggests a characteristic time-scale [12]

$$\tau := \tfrac{1}{\Omega\sqrt{S_0 J}}. \tag{10}$$

The dynamical equations of Γ_{mn} can be obtained from the Heisenberg equation of motion for the operators R_k given as

$$R_k^{(\mathrm{out})} = R_k^{(\mathrm{in})} - it[R_k^{(\mathrm{in})}, H], \tag{11}$$

with $\hbar = 1$. For example, the dynamics of R_5 is obtained as

$$R_5^{(\mathrm{out})} = R_5^{(\mathrm{in})} + \tfrac{\kappa}{\sqrt{S_0}} R_4^{(\mathrm{in})} R_1^{(\mathrm{in})}. \tag{12}$$

where the coupling constant is defined as $\kappa := \frac{t}{\tau}$. Hence, for the dynamics of the variance of R_5 we obtain

$$\langle (R_5^{(\mathrm{out})})^2\rangle = \langle (R_5^{(\mathrm{in})})^2\rangle + \tfrac{\kappa^2}{S_0}\langle (R_4^{(\mathrm{in})})^2 (R_1^{(\mathrm{in})})^2\rangle + \tfrac{\kappa}{\sqrt{S_0}}\langle R_1^{(\mathrm{in})}\{R_4^{(\mathrm{in})}, R_5^{(\mathrm{in})}\}_+\rangle, \tag{13}$$

where $\{A, B\}_+$ is the anticommutator of A and B. Knowing that due to symmetries of the setup for all times $\langle R_k\rangle = 0$ for $k = 1, 2, 3, 5, 6$, we obtain

$$\begin{aligned} \langle (\Delta R_5^{(\mathrm{out})})^2\rangle &= \langle (\Delta R_5^{(\mathrm{in})})^2\rangle + \tfrac{\kappa^2}{S_0}\langle (\Delta R_4^{(\mathrm{in})})^2 (\Delta R_1^{(\mathrm{in})})^2\rangle + \tfrac{\kappa^2}{S_0}\langle R_4^{(\mathrm{in})}\rangle^2\langle (\Delta R_1^{(\mathrm{in})})^2\rangle \\ &+ \tfrac{\kappa}{\sqrt{S_0}}\langle \Delta R_1^{(\mathrm{in})}\{\Delta R_4^{(\mathrm{in})}, \Delta R_5^{(\mathrm{in})}\}_+\rangle + \tfrac{2\kappa}{\sqrt{S_0}}\langle \Delta R_1^{(\mathrm{in})}\Delta R_5^{(\mathrm{in})}\rangle\langle R_4^{(\mathrm{in})}\rangle, \end{aligned} \tag{14}$$

where we used the notation $\Delta A := A - \langle A\rangle$.

Let us now consider dynamics for $t \lesssim \tau$. Knowing that $\langle R_4\rangle = \sqrt{S_0}$ for $t = 0$ and $\langle \Delta R_k \Delta R_l \Delta R_m \cdot \cdot\rangle \lesssim 1$, we can examine how the different terms depend on S_0. We find that on the right hand side of (14) the second term is of order $\frac{1}{S_0}$, the fourth term is of the order $\frac{1}{\sqrt{S_0}}$, while the rest of the term are of order 1. Thus, assuming a large number of photons, that is, a large S_0, several terms can be neglected and we obtain

$$\langle (\Delta R_5^{(\mathrm{out})})^2\rangle = \langle (\Delta R_5^{(\mathrm{in})})^2\rangle + \tfrac{\kappa^2}{S_0}\langle R_4^{(\mathrm{in})}\rangle^2\langle (\Delta R_1^{(\mathrm{in})})^2\rangle + \tfrac{2\kappa}{\sqrt{S_0}}\langle \Delta R_1^{(\mathrm{in})}\Delta R_5^{(\mathrm{in})}\rangle\langle R_4^{(\mathrm{in})}\rangle. \tag{15}$$

Long, but straightforward calculations show that several terms can also be neglected in the dynamical equation for variances of the other R_k variables, assuming large J and S_0. Hence, for the evolution of the covariance matrix

$$\Gamma_P = M\Gamma_0 M^T \tag{16}$$

is obtained, where M is identity matrix, apart from $M_{5,1} = \frac{\langle R_4^{(\mathrm{in})}\rangle}{\sqrt{S_0}}\kappa$. The evolution of the expectation values is described as

$$\langle R_k^{(\mathrm{out})}\rangle = \sum_l M_{kl}\langle R_l^{(\mathrm{in})}\rangle. \tag{17}$$

Similar analysis shows that the dynamical equations for higher order moments of R_k can also be simplified if $t \lesssim \tau$. When computing the dynamics of these moments, instead of the Heisenberg equation of motion, $R_k^{(\mathrm{out})} = \sum_l M_{kl} R_l^{(\mathrm{in})}$ can be used. Under this dynamics, which is a linear mapping between operators, the state remains Gaussian. Note, however, that this approximation breaks down for much larger times $t \sim \tau\sqrt{J}$. (See also Appendix.)

The second step of the QND process is measuring S_y of the light. The theory of Gaussian states can be used to describe the measurement, which is modeled with a projection [42] [5]

$$\Gamma_M = \Gamma_P - \Gamma_P(P_y\Gamma_P P_y)^{\mathrm{MP}}\Gamma_P^T. \tag{18}$$

Here MP denotes the Moore-Penrose pseudoinverse and $P_y = \mathrm{diag}(0,0,0,0,1,0)$. As described above, the QND measurement minimally disturbs the unmeasured components. In the Gaussian approximation the variance of the other spin components remain unchanged.

Measuring the other J_k components is analogous to the J_x case. Note that magnetic fields could be used to rotate the collective spin to facilitate the measurement of the different spin components. Finally, as we have already noted, after each squeezing step, feedback has to be used to restore the zero expectation value of the angular momentum coordinates. We have also mentioned that post-selection can be used in the place of feedback. In this case a feedback scheme do not have to be realized, however, part of the experiments must be discarded. Thus, when a QND measurement determines $\langle J_l\rangle$, only cases with $|\langle J_l\rangle| \leq B$ are retained. We define $I(f(x), L) := \int_{-L}^{L} f(x)\exp(-x^2/2\Delta^2)dx$, where Δ is the width of the distribution of values obtained when measuring $\langle J_l\rangle$. The effect of post-selection is given as $\mathrm{var}(\langle J_l\rangle)_{\mathrm{after}} = \mu\mathrm{var}(\langle J_l\rangle)_{\mathrm{before}}$ where $\mu := [I(x^2, B)/I(1, B)]/[I(x^2, \infty)/I(1, \infty)]$ and the fraction of post-selected experiments is $q := I(1, B)/I(1, \infty)$. Hence, for a moderate post-selection of $q = 0.5$ (0.75) for all the three squeezing steps, the variances decrease to 14% (37%) of their original values.

Let us now make the calculations for realistic parameters. We consider $N = 10^6$ ^{87}Rb atoms with spin $j = 1$, and for the light field $S_0 = 5 \times 10^7$. Sequential squeezing of the x, y and z spin components is shown in figure 2. The horizontal axis indicates the total interaction time, with successive intervals of up to 2τ for measurement of J_x, J_y, and J_z, respectively. Results are shown for squeezing from a thermal state and also from

$$|\Psi\rangle_0' := |+j\rangle^{\otimes\frac{N}{2}} \otimes |-j\rangle^{\otimes\frac{N}{2}}. \tag{19}$$

We obtain $\xi_s^2 = 0.32$ and $\xi_s^2 = 0.20$ for the completely mixed initial atomic state and for (19), respectively. Remarkably, the QND interaction can be solved exactly for the initial state (19), as shown in the Appendix. The results are presented in figure 2. In that calculation we find that, for large N, time $t \sim \tau \times J^{0.25}$ gives $(\Delta J_x)^2 \sim \sqrt{J}$ and the two halves of the atoms remain almost fully polarized into the $+z$ and $-z$ directions, respectively. For much longer times, the two halves are not fully polarized any more. In particular, for $t \sim \tau_2 := \tau \times \sqrt{J}$ we obtain $(\Delta J_x)^2 \sim 1$, and $\xi_s^2 = \frac{1}{2}$. Thus, we have squeezing even in the long-time (von Neumann) limit.

C. Model including losses

We now incorporate decoherence, following ideas from [13, 14, 17], adapted to our use of a correlation matrix of all the three spin components. In particular, a parameter η describes the probability that an atom suffers spontaneous

[5] Equation (18) is analogous to the formula for the behavior of the correlation matrix during a von Neumann measurement in the case of Gaussian continuous variable systems described in [11]. However, (18) is based on the theory of $SU(2)$ Wigner functions [43] rather than on the theory of Wigner functions for multimode systems. For polarized ensembles, (18) has already been used in [17].

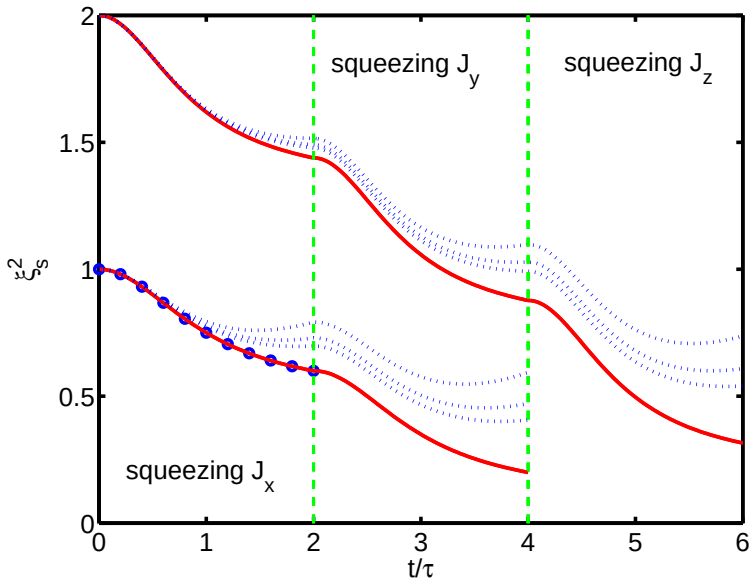

FIG. 2: Dynamics of the spin squeezing parameter ξ_s^2 as a function of the time for ^{87}Rb atoms with spin 1. Sequential QND measurements are made of the x, y and z spin components, with interaction time t up to 2τ. The initial state is the completely mixed state (upper traces) or the state of (19) (lower traces). (Solid) Gaussian approximation without losses. (Dotted) Gaussian approximation including losses with (from top to bottom) $\alpha = 50, 75, 100$. (Dots) Exact model.

excitation due to the off-resonant probe, and thus the fraction of atoms that decohere during the QND process. For simplicity, we assume that the atoms end up in the completely mixed state, which is usual for handling the effects of noise. We obtain

$$\Gamma'_P = (\mathbb{1} - \eta D) M \Gamma_0 M^T (\mathbb{1} - \eta D) + \eta(2-\eta) D \Gamma_{\rm noise}, \tag{20}$$

where $D = \mathrm{diag}(1,1,1,0,0,0)$ and $\Gamma_{\rm noise} = \mathrm{diag}(1,1,1,0,0,0) \times \frac{n_j}{j}$ [6]. Here n_j is the variance of j_x for a spin-j particle in a totally mixed state. The decoherence is connected to κ through

$$\eta = Q \frac{\kappa^2}{\alpha}, \tag{21}$$

where α is the resonant optical depth of the sample and $Q = 1$ for the spin-$\frac{1}{2}$ case. For the spin-1 case, $Q = \frac{8}{9}$ if the near-resonant intermediate state has $j = 0$ [46]. Using these techniques, we calculate the degree of squeezing as a function of the time for different, experimentally feasible, values of α [14, 15].

The results, shown in figure 2, indicate that considerable squeezing is indeed possible under realistic conditions. For the completely mixed atomic initial state, we obtained $\xi_s^2 = 0.74, 0.61$, and 0.54 for $\alpha = 50, 75$ and 100, respectively. For comparison, polarized spin states have been squeezed in variance by $\sim 50\%$ under similar conditions. Thus, our proposal is realistic with present-day technology. The values of α chosen reflect the state-of-the-art for single-pass optical probing [16, 44, 45]. The use of an optical cavity could boost the effective α by orders of magnitude [21].

IV. CONCLUSIONS

In summary, spin squeezing of unpolarized atomic ensembles by QND measurement shows several intriguing differences relative to polarized samples. The absence of a significant measurement back-action allows simultaneous squeezing of all spin components and approaches a macroscopic singlet state. Spin squeezing of this type implies entanglement of a macroscopic number of particles, for arbitrary spin. To treat this problem, we have extended the Gaussian formalism to include the dynamics of all the three spin components and their correlations for an unpolarized

[6] Compare (20) with (13) in [13].

ensemble [7]. The advantage of our approach is that it is possible to determine the area of validity for our model, and we can also incorporate von Neumann measurements. In the lossless case, the results agree with an exact calculation. Realistic calculations including decoherence indicate that production of these macroscopic singlet states should be possible with existing ensemble systems. In the future, it will be interesting to look at the possibility of storing quantum information in the decoherence free subspace [48], formed by different singlet states, obtained for $j > \frac{1}{2}$ when using different initial states for our squeezing procedure. The method of modeling large spin systems can be generalized to multiple ensembles and a series of light pulses or even for modeling many-particle quantum systems in other areas of physics.

Acknowledgments

We thank A. Acín, N. Brunner, A. Cere, J.I. Cirac, G. Giedke, O. Gühne, C. Hadley, K. Hammerer, M. Koschorreck, M. Lewenstein, and M. Napolitano for fruitful discussions. We thank the support of the EU (OLAQUI, SCALA, QICS), the National Research Fund of Hungary OTKA (Contract No. T049234), the Spanish MEC (Ramon y Cajal Programme, Consolider-Ingenio 2010 project "QOIT", project No. FIS2009-12773-C02-02, ILUMA project with Ref. FIS2008-01051).

Appendix A: Exact model for squeezing $(\Delta J_z)^2$ starting from the state (19)

1. The $j = \frac{1}{2}$ case.

We briefly describe a method that makes it possible to model exactly the QND process without using the Gaussian approximation. First, let us consider the initial state (19) for the $j = \frac{1}{2}$ case and measure J_x with the QND interaction. We divide the atoms into two groups. In the first group, initially N_1 atoms are in the $|+\frac{1}{2}\rangle_z$ state, while in the second group N_2 atoms are in the $|-\frac{1}{2}\rangle_z$ state. We define the angular momentum operators $J_{k,l}$ with $k = 1, 2$ corresponding to the two groups. We choose the two halves equal: $N_1 = N_2 = \frac{N}{2}$. Moreover, without the loss of generality we choose N_k to be even since in this case $J_{k,l}$ have integer eigenvalues between $-\frac{N_k}{2}$ and $\frac{N_k}{2}$. The initial state can be given in the $J_{k,x}$ basis by

$$|\Psi_1\rangle := \sum_j f_1(j)|j\rangle_{1,x} \tag{A1}$$

and

$$|\Psi_2\rangle := \sum_j f_2(j)(-1)^j|j\rangle_{2,x}, \tag{A2}$$

where for large particle numbers $f_m(x) \propto \exp[-\frac{x^2}{N_m}]$. Similarly, we define for the state of the light

$$|\Psi_l\rangle := \sum_j g(j)|j\rangle_{\mathrm{light},z}, \tag{A3}$$

where $g(z) \propto \exp[-\frac{z^2}{2S_0}]$. Hence, we obtain the evolution for the state of atoms and photons as

$$\begin{aligned} |\Phi(t)\rangle &= \exp(-iJ_xS_zt)|\Psi_1\rangle \otimes |\Psi_2\rangle \otimes |\Psi_l\rangle \\ &= \sum_{j_1,j_2}\sum_s e^{-i(j_1+j_2)s\Omega t} f_1(j_1)f_2(j_2)(-1)^{j_2}g(s)|j_1\rangle_x|j_2\rangle_x|s\rangle_{\mathrm{light},z}. \end{aligned} \tag{A4}$$

The projection to the $S_y = 0$ state can be incorporated into the model by introducing $w_s =: \langle S_z = s|S_y = 0\rangle$. Then, the final state of the atoms is

$$|\Psi(t)\rangle \propto \sum_{j_1,j_2} G(j_1 + j_2)f_1(j_1)f_2(j_2)(-1)^{j_2}|j_1\rangle_x|j_2\rangle_x, \tag{A5}$$

[7] Modeling lossy dynamics is also possible by writing down the infinite hierarchy of dynamical equations for many-body correlations and truncate it at second order [47].

where $G(j) := \sum_s e^{-ijs\Omega t} g(s)w(s)$. Here $G(j)$, for large systems, is essentially the Fourier transform of $g(s)w(s)$. The value of $w(s)$ matters only for $|s| \lesssim \sqrt{S_0}$ since for much larger s we have $g(s) \approx 0$. For this case, $w(s)$ is to a good approximation for successive values of s is alternating between 0 and a constant. Hence, $G(j)$ is very close to a Gaussian around $j = 0$ with a variance $\sim J(\frac{t}{\tau})^2$. Thus, $t \sim J^{0.25}\tau$ gives $(\Delta J_x)^2 \sim \sqrt{J}$, while for $t \sim \tau_2 := \tau\sqrt{J}$ the width of the Gaussian is ~ 1. That is, when computing $|\Psi(t)\rangle$, only states with $j_1 + j_2 = 0$ are selected corresponding to projecting to the $J_x = 0$ subspace. Thus, for $t \sim \tau_2$ our setup realizes a von Neumann measurement of J_x. For large systems the summation in (A5) can be replaced by integration. Using these ideas, we obtain

$$\langle J_x^2 \rangle \approx \int dj_1 dj_2 \left| \langle \Psi(t)|j_1\rangle_x |j_2\rangle_x \right|^2 (j_1 + j_2)^2. \tag{A6}$$

Due to the absolute value sign, the $(-1)^{j_2}$ term in (A5) can be neglected for this calculation, thus the integral of a smooth function must be computed numerically for $\langle J_x^2 \rangle$. The dynamics of the other two variances can be computed similarly, knowing the matrix elements of $M := J_y^2 + J_z^2$. The change of $\langle M \rangle$ during this dynamics is negligible. Note that $[J_x, M] = 0$, thus a von Neumann measurement of J_x does not change $\langle M \rangle$.

2. The $j > \frac{1}{2}$ case.

Finally, the $j > \frac{1}{2}$ case is analogous to the $j = \frac{1}{2}$ case, if we notice that starting from a product state with N_1 particles in state $|j\rangle$ and N_2 particles in state $|-j\rangle$ gives the same dynamics for $\langle J_{k,l} \rangle$, as starting from a state with $2N_1 j$ particles in state $|+\frac{1}{2}\rangle$ and $2N_2 j$ particles in state $|-\frac{1}{2}\rangle$.

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