

the cavity decay, and (ii) it describes a highly correlated atomic state containing a high degree of spin squeezing. The latter can be seen by computing the ratio between the spin fluctuations in the x and y direction: $\langle S_x^2 \rangle / \langle S_y^2 \rangle = \langle (S^+ + S^-)^2 \rangle / \langle (S^+ - S^-)^2 \rangle = (\Omega_- - \Omega_+)^2 / (\Omega_- + \Omega_+)^2$. For any $\Omega_+ \Omega_- > 0$ this ratio is smaller than 1, indicating that the dark state is spin squeezed.

To quantify the degree of useful spin squeezing, we use the phase sensitivity introduced by Wineland *et al.* [23],

$$\delta\phi = \frac{\sqrt{\langle S_x^2 \rangle}}{|\langle S_z \rangle|}. \quad (3)$$

We first focus on symmetric states with the total spin $S = N/2$, i.e., an initially spin-polarized ensemble, and expand the dark state into eigenstates of S_z as $|\psi_{\text{spin}}\rangle = \sum_m c_m |S = N/2, S_z = -N/2 + m\rangle$. Substituting this into Eq. (2) results in a recursion relation between c_m and c_{m+2} , in close analogy with squeezed light [24],

$$c_{m=2n} = \left(\frac{\Omega_+}{\Omega_-}\right)^n \binom{N/2}{n} \binom{N}{2n}^{-1/2} c_0. \quad (4)$$

Here $(:)$ are the binomial coefficients and c_0 is determined by the normalization condition $\sum_m |c_m|^2 = 1$. The phase sensitivity $\delta\phi$ of $|\psi_{\text{spin}}\rangle$ is shown by the blue solid curve in Fig. 2 as a function of the ratio Ω_+/Ω_- between the control fields. For $\Omega_+/\Omega_- = 0$, the dark state is fully polarized along the z axis, giving a phase sensitivity of $1/\sqrt{N}$ corresponding to the standard quantum limit (SQL). In the opposite limit of $\Omega_+ \rightarrow \Omega_-$, the phase sensitivity approaches the Heisenberg limit [16,25] $\delta\phi = 1/\sqrt{N(N/2 + 1)}$, indicating that the dark state corresponds to an almost maximally squeezed atomic state. A related method to generate spin squeezing in the weak coupling regime, i.e., $\Omega_+/\Omega_- \ll 1$, has been recently proposed in Ref. [26].

We now turn to the preparation of the dark state (2) and study the corresponding quantum dynamics, focusing first on the effect of the cavity decay. To this end, consider an ensemble of N four-state atoms, consisting of two (meta) stable states $|+\rangle, |-\rangle$ serving as our effective spin states and two excited states $|e_{\pm}\rangle$. The atoms are coupled to a

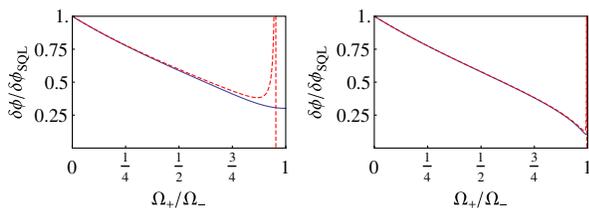


FIG. 2 (color online). Phase sensitivity of the dark state $|\psi_{\text{spin}}\rangle$. Exact numerical solution (solid blue curve) and mean-field approximation (dashed red curve) for $N = 10$ (left) and $N = 100$ (right) atoms. The optimal squeezing is obtained for $\Omega_+ \rightarrow \Omega_-$ and approaches the Heisenberg limit $\delta\phi = 1/\sqrt{N(N/2 + 1)} \sim 1/N$.

single mode of an open cavity (cf. Fig. 1) with resonance frequency ω_a , volume V , and photon escape rate κ driving the transitions $|\pm\rangle \rightarrow |e_{\pm}\rangle$ and strong control fields Ω_{\pm} with frequency ω_c driving the transitions $|\mp\rangle \rightarrow |e_{\pm}\rangle$, respectively. The atom-cavity field coupling strength is given by $g = \wp/\hbar\sqrt{\hbar\omega_a/2\epsilon_0V}$, where \wp is the dipole matrix element of the respective transition. We assume the control fields and the cavity mode to be far detuned from the resonant transition frequency $\omega_{\text{res}} = \omega_{\pm} - \omega_{e_{\pm}}$ between ground state and excited state manifold. This allows us to adiabatically eliminate the excited states, and we obtain the effective Hamiltonian (1) with $S^+ = \sum_j^N \sigma_j^+ = \sum_j^N |+\rangle_j \langle -|$, $S^- = (S^+)^{\dagger}$, and $|\Delta| = |\omega_{\text{res}} - \omega_{a,c}|$. For simplicity of the calculation we assume here that we can realize two independent Λ schemes according to Fig. 1 and neglect any possible hyperfine structure in the excited states. While this approximation is only true if the detunings from the respective excited state is smaller than the hyperfine splitting in the excited state manifold, Hamiltonian (1) is valid for more general conditions, when Raman processes mediated by multiple virtual states can be added up [27,28]. A more detailed derivation of the Hamiltonian, also including different detunings, can be found in the Supplemental Material [29]. Assuming $\kappa \gg |\Delta|$, we can neglect the occupation of the cavity mode, and using standard techniques [30] we obtain the master equation in Lindblad form after tracing over the cavity modes:

$$\frac{\partial}{\partial t} \rho = -\gamma_{\text{cav}} [\{I^{\dagger} I, \rho\}_+ - 2I\rho I^{\dagger}]. \quad (5)$$

Here $\{\cdot, \cdot\}_+$ denotes the anticommutator, $\gamma_{\text{cav}} = g^2 \Omega^2 / \Delta^2 \kappa$ with $\Omega^2 = \Omega_+^2 + \Omega_-^2$, $I = \sin\theta S^+ + \cos\theta S^-$ is the Lindblad operator accompanying the emission of a cavity photon, and we defined $\tan\theta = \Omega_-/\Omega_+$.

To compute the phase sensitivity $\delta\phi$, we use (5) to determine the time evolution of the expectation values of the spin operators S_z and S_x^2 :

$$\frac{d\langle S_z \rangle}{dt} = -\gamma_{\text{cav}} [\cos^2(\theta) \langle S^+ S^- \rangle - \sin^2(\theta) \langle S^- S^+ \rangle], \quad (6)$$

$$\begin{aligned} \frac{d\langle S_x^2 \rangle}{dt} = & -\gamma_{\text{cav}} (\sin(\theta) - \cos(\theta)) \\ & \times \langle (\sin(\theta) S^- + \cos(\theta) S^+) (S_x S_z + S_z S_x) + \text{H.c.} \rangle. \end{aligned} \quad (7)$$

Note that the dark state $|\psi_{\text{spin}}\rangle$, defined by Eq. (2), is indeed a steady state of Eqs. (6) and (7). Equations (6) and (7) involve higher moments of the spin, whose time evolution should be computed independently. To avoid this complication and obtain an analytic solution, we linearize the equations of motion around $\langle S_z \rangle \approx -N/2$. Defining the small fluctuations as $\delta S_z = S_z + N/2$ and using the approximation $\langle S_x^2 + S_y^2 \rangle = \langle \mathbf{S}^2 \rangle - \langle S_z^2 \rangle \approx N \langle \delta S_z \rangle + N/2$, we find

$$\frac{d\langle\delta S_z\rangle}{dt} = -\gamma_{\text{cav}}N[\cos(2\theta)\langle\delta S_z\rangle - \sin^2(\theta)], \quad (8)$$

$$\frac{d\langle S_x^2\rangle}{dt} = -\gamma_{\text{cav}}N\left[\cos(2\theta)\langle S_x^2\rangle + [1 - \sin(2\theta)]\frac{1}{4}\right]. \quad (9)$$

The solutions to these equations decay exponentially in time with an effective rate $\gamma_{\text{eff}} = \gamma_{\text{cav}} \cos(2\theta)N$. From Eqs. (8) and (9) we obtain the phase sensitivity in the steady state as

$$\delta\phi^2 = \frac{N[1 - \sin(2\theta)]\cos(2\theta)}{[N\cos(2\theta) - 2\sin^2(\theta)]^2}. \quad (10)$$

In Fig. 2 we compare this approximate expression (dashed red curve) with the exact result obtained from Eq. (4) (solid blue curve). The two curves significantly deviate only for $\Omega_+/\Omega_- \approx 1 - \mathcal{O}(1/N)$, or equivalently $\theta \approx \pi/4 - \mathcal{O}(1/N)$. In this regime the present linearized approximation fails because $\langle\delta S_z\rangle = \sin^2(\theta)/\cos(2\theta) \approx \mathcal{O}(N)$. Note that, approaching the limit $\Omega_+ = \Omega_-$, the steady-state spin squeezing reaches its maximal Heisenberg-limited value. But, at the same time, the effective dark-state pumping rate γ_{eff} tends to zero, making the spin squeezing process extremely slow. This renders competing processes, such as e.g., spontaneous Raman scattering of individual atoms, very important, as will be discussed below.

Up to this point, we have restricted ourselves to the manifold of total spin $S = N/2$. This approach is valid if the system is initially prepared in the maximally polarized state $|S = N/2, S_z = N/2\rangle$ because Hamiltonian (1) commutes with the total spin S^2 . Consequently, if the system is initially prepared in a state with a different total spin, the final steady state will be different and will in general contain a smaller amount of spin squeezing. This is shown in Fig. 3(a), where we compare the linearized solution with the exact numerical solution of the Lindblad equation (5) for $N = 10$ spins with different initial conditions.

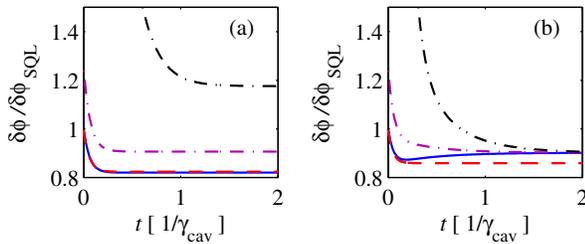


FIG. 3 (color online). Time evolution of the phase sensitivity for $N = 10$ spins and $\Omega_-/\Omega_+ = 0.2$. Numerical solution of the master equation, with fully polarized (solid curves) or randomly generated (dash-dotted curves) initial state, and mean-field approximation (dashed curves). (a) The atoms are coupled only to the cavity, i.e., $\gamma = 0$. (b) Finite scattering rate $\gamma \neq 0$ with single-atom cooperativity $\chi = 1$. Observe that the steady state does not depend on the initial state.

The linearization works well only when the spins are initially polarized, as would be expected.

So far our analysis completely disregarded the effect of spontaneous Raman scattering of the individual atoms into free space. Being a single atom process, it breaks the conservation of the total spin and thus competes with the squeezing process. To investigate the effect of spontaneous Raman scattering we introduce a spontaneous decay rate $\gamma \ll |\Delta|$ to the states $|e_{\pm}\rangle$. The corresponding master equation is easily obtained using standard techniques and reads

$$\begin{aligned} \frac{\partial}{\partial t}\rho = & -\gamma_{\text{cav}}[\{I^\dagger I, \rho\}_+ - 2I\rho I^\dagger] \\ & - \gamma_{\text{spont}} \sum_{\alpha=\sigma^\pm, \pi} \sum_{j=1}^N [\{\mathcal{L}_{j,\alpha}^\dagger \mathcal{L}_{j,\alpha}, \rho\}_+ - 2\mathcal{L}_{j,\alpha}\rho\mathcal{L}_{j,\alpha}^\dagger]. \end{aligned} \quad (11)$$

The first term on the right-hand side of Eq. (11) is equivalent to (5), whereas the second term describes the spontaneous Raman scattering of atom j into free space with rates $\gamma_{\text{spont}} = \gamma\Omega^2/\Delta^2$. The Lindblad operators are given by $\mathcal{L}_{j,\sigma^\pm} = (\cos\theta\sigma_j^\pm + \sin\theta\sigma_j^\mp)\sigma_j^\mp$ and $\mathcal{L}_{j,\pi} = \cos\theta\sigma_j^\pm + \sin\theta\sigma_j^\mp$. The numerical solution of Eq. (11) is shown in Fig. 3(b) for different initial conditions and $N = 10$ spins. The spontaneous Raman scattering couples different total spin- S manifolds and, as expected, lifts the degeneracy between them. Consequently, it leads to a mixed but nevertheless unique steady state, characterized by a unique density matrix, independent of the initial state, and hence presents a feature rather than a detrimental problem.

Still, Raman scattering leads to a reduction of the achievable phase sensitivity. To quantify this effect we obtain the contribution of the single-spin decay to the time evolution of the collective variables, by using the second row of Eq. (11), leading to

$$\frac{d\langle\delta S_z\rangle}{dt} = -\gamma_{\text{spont}}[\langle\delta S_z\rangle - \sin^2(\theta)N], \quad (12)$$

$$\frac{d\langle S_x^2\rangle}{dt} = -2\gamma_{\text{spont}}\left(1 - \frac{1}{2}\sin(2\theta)\right)\left[\langle S_x^2\rangle - \frac{1}{4}\right]. \quad (13)$$

Adding Eqs. (12) and (13) to the contribution of the collective coupling to the cavity, Eqs. (8) and (9), we find that in the steady state the competition between the two processes leads to

$$\langle\delta S_z\rangle_\infty = \frac{\sin^2(\theta)(\frac{Ng^2}{\kappa} + N\gamma)}{\frac{Ng^2}{\kappa} \cos(2\theta) + \gamma}, \quad (14)$$

$$\langle S_x^2\rangle_\infty = \frac{1}{4} \frac{\frac{Ng^2}{\kappa} [1 - \sin(2\theta)] + 2\gamma [1 - \frac{1}{2}\sin(2\theta)]}{\frac{Ng^2}{\kappa} \cos(2\theta) + 2\gamma [1 - \frac{1}{2}\sin(2\theta)]}. \quad (15)$$

For a given single-atom cooperativity $\chi = \gamma_{\text{cav}}/\gamma_{\text{spont}} = g^2/\kappa\gamma$, the optimal value of the phase sensitivity $\delta\phi$ can be obtained by numerically minimizing the corresponding expression with respect to the mixing angle θ while keeping χ fixed. The resulting curve is shown in Fig. 4 for $N = 10^6$. To gain insight into this result, we will now separately investigate the limits of small and large single-atom cooperativity.

If the single-atom cooperativity is small, but $\chi N \sim 1$, the optimal spin-squeezing is obtained for $\theta \ll \pi/4$. Expanding Eqs. (14) and (15) around $\theta \approx 0$ we obtain a phase sensitivity $\delta\phi^2 \approx \frac{1}{N}(1 - N\chi\theta + 4\theta^2)$ with the optimum value obtained for $\theta = N\chi/8$:

$$\delta\phi_{\text{opt}} = \frac{1}{\sqrt{N}} \left(1 - \frac{\chi^2 N^2}{32} \right). \quad (16)$$

It should be noted that, due to the collective enhancement by a factor of N , one gains a quadratic improvement in the phase sensitivity even for small cooperativities.

If the cooperativity is larger, i.e., $\chi N \gg 1$, the optimal squeezing is obtained for $\Omega_+ \approx \Omega_-$ and one may naively expect the linearization to fail. However, to render the problem tractable we nevertheless perform the large- N expansion and explicitly check the validity of the obtained results below. To obtain the scaling of the phase sensitivity, we expand Eqs. (14) and (15) around $\theta = \pi/4 - \epsilon$. For small $\epsilon \sim 1/\sqrt{N} \ll 1$ and neglecting terms of order $1/N$, we find $\delta\phi^2 = \frac{1}{N} \frac{1+2N\chi\epsilon^2}{2N\chi\epsilon}$, which has a minimum at $\epsilon = \sqrt{1/2N\chi}$. This leads to the optimal sensitivity

$$\delta\phi = \left(\frac{2}{\chi N^3} \right)^{1/4}. \quad (17)$$

Note that, for the optimal value of θ , the ensemble is still mostly polarized, as can be checked by explicitly computing $\langle \delta S_z \rangle \approx \frac{\chi+1}{4\chi\epsilon} \sim \sqrt{N}$. This observation provides a self-consistency check for our linearization procedure in the limit of a large ensemble, where $\sqrt{N} \ll N$. Equation (17)

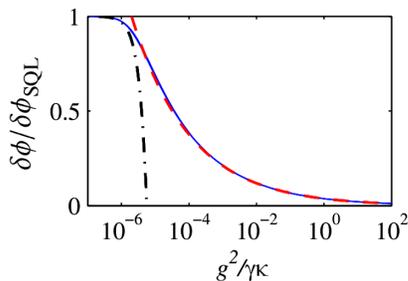


FIG. 4 (color online). Optimal phase sensitivity in the steady state, in the presence of single spin decay for $N = 10^6$. The solid curve is given by the numerical minimization of Eqs. (14) and (15). The dash-dotted and dashed curves are Eqs. (16) and (17), respectively.

shows that the achievable phase sensitivity scales as $\delta\phi \sim N^{-3/4}$ and thus offers a significant improvement over the SQL. With respect to other proposals with a similar scaling [16,19,31], our approach deterministically generates spin squeezing in a steady state by optical pumping and can thus potentially be more robust against external perturbations.

A possible experimental realization could be set up in ^{87}Rb using the clock states $|F=1, m_F=0\rangle$ and $|F=2, m_F=0\rangle$ in the $5S_{1/2}$ ground-state manifold. Circularly σ_+ -polarized cavity and control fields couple these states to the states $|F=1, m_F=+1\rangle$ and $|F=2, m_F=+1\rangle$ of the $5P_{1/2}$ manifold. In this case the inclusion of the hyperfine structure leads to an effectively larger Raman rate, as the scattering pathways via the two hyperfine states interfere constructively for suitably chosen detunings (see the Supplemental Material [29]). For current experiments [18], we have $N \approx 10^6$ and $\chi \approx 0.1$, predicting a phase sensitivity $\delta\phi/\delta\phi_{\text{SQL}} \approx 0.07$, i.e., an improvement of more than 1 order of magnitude with respect to the SQL.

We now discuss two additional effects related to the experimental realization of the proposed model. First, in a multilevel atom (such as the suggested ^{87}Rb), the spontaneous Raman scattering can lead to states outside of the effective four-state system. Because, as shown above, the squeezed steady state is (mixed but) unique and is achieved from any initial state, we can use additional optical pumping fields to repump the atoms into the correct configuration. In the case of ^{87}Rb this can be achieved through linearly polarized fields resonantly driving the $F=1 \rightarrow F'=1$, $F=2 \rightarrow F'=2$, and $\Delta m_F = 0$ transition. These fields do not couple directly to the clock states $|F=1, m_F=0\rangle$ and $|F=2, m_F=0\rangle$ and therefore act as a repumping field for the proposed four-state scheme. The repump processes can be easily incorporated in our formalism in the form of additional single-atom scattering channels [29]. These processes effectively increase the single-atom scattering rate and reduce the cooperativity χ . In addition, the use of a large detuning Δ makes it possible to obtain repumping rates that are much larger than the decay rate into the external states and to minimize the loss of atoms into these states. We conclude that our results, and in particular Eq. (17), are valid even in the presence of additional repumped decay channels, provided that the cooperativity rate χ is computed with respect to the total linewidth of the excited states.

Second, in the ideal model (1), we assumed a spatially homogeneous coupling of the atoms to the cavity mode, which can be approximately achieved only in a ring cavity. In a standing-wave cavity the off-resonant Raman coupling $g\Omega_{\pm}/\Delta$ depends on the position of the atoms and can be either positive or negative, leading to a cancellation of spin squeezing. A solution is to localize the atoms in space and choose an appropriate geometry for the control fields and

the cavity mode. This ensures that all coupling constants have the same sign, as experimentally shown in Ref. [18]. In this case, the symmetry of the collective state is preserved, leading only to a small reduction of the effective spin cooperativity.

To summarize, we presented a method for the deterministic generation of spin-squeezed states in a driven ensemble of effective two-state atoms in a strongly dissipative cavity. The generating process can be understood as optical pumping into a nonequilibrium steady state of the atom that at the same time is a dark state with respect to the cavity decay. Introducing spontaneous Raman scattering we showed that the squeezed steady state is unique and does not depend on the initial state of the system. We discussed the effect of the single atom spontaneous Raman scattering on the achievable phase sensitivity and found that it scales favorably with the single-atom cooperativity, indicating that the present method can be of direct importance for, e.g., optical atomic clocks.

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