Detuning-enhanced cavity spin squeezing

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The unconditional squeezing of the collective spins of an atomic ensemble in a laser driven optical cavity [I. D. Leroux, M. H. Schleier-Smith, and V. Vuletić, Phys. Rev. Lett **104**, 073602 (2010)] is studied and analyzed theoretically. Surprisingly, we find that the largely detuned driving laser can improve the scaling of cavity squeezing from $S^{-2/5}$ to $S^{-2/3}$, where S is the total atomic spin. Moreover, we also demonstrate that the experimental imperfection of photon scattering into free space can be efficiently suppressed by detuning.

I. INTRODUCTION

Large ensembles of atoms are good platforms for quantum information processing [1–3], due to the long coherence time of atomic energy levels and the collective enhanced interaction with light. Therefore, the atomic ensemble has been studied extensively for both fundamental physics research [4] and practical applications, such as quantum memory [1], atomic clocks [5,6], magnetometers [7,8], and gravitational wave detectors [9]. For those metrology applications, it is believed that the classical limitation of measurement precision can be broken by using quantum states of the atomic ensemble [10,11]. The squeezed spin state (SSS) [12] is one type of such a quantum correlated state with reduced fluctuations in one axis of the collective spin components, and it has therefore attracted considerable attention recently.

To prepare the SSS, a variety of experiment schemes based on light-matter interaction have been proposed and demonstrated [10]. One approach is transfer the quantum state of light to the atomic spins directly [13–16], where the degree of spin squeezing is determined by the quality of squeezed input light. Another approach is to generate the SSS by the quantum nondemolition measurement on the states of photons coupled with the atomic ensemble [17–22]. While this method has already been demonstrated by several groups, the efficiency of SSS preparation strongly depends on the performance of the detector. The last and very promising approach is cavity squeezing [23–29] without the measurement of the light field, which is based on the off-resonant interaction between atomic ensemble and light circulating in an optical resonator cavity. The spin states of the ensemble imprint their quantum fluctuations onto the light, which acts back and reduces the fluctuation of spin states.

In this paper, we theoretically study the detuning dependence of cavity spin squeezing for the experimental scheme demonstrated in Ref. [24] [Fig. 1(a)]. Comparing with the near resonance case [23], it is surprising to find that the scaling of cavity squeezing on total atomic spin can be significantly improved from $S^{-2/5}$ to $S^{-2/3}$ by large laser detuning. In

addition, we find that the spin squeezing will be enhanced if the atoms are weakly coupled to the cavity or the laser detuning is very large. From our numerical solutions and analytical analysis, the large detuning is very important because the squeezing originates from the laser induced spin state dependent geometry phase [30,31]. Finally, we study the influence of scattering of a photon into free space due to imperfect Raman scattering, and demonstrate that the optimal spin squeezing can be obtained with appropriate detuning. This improvement of spin squeezing by detuning, without the requirement of preparation or postselection of the photon state, is very feasible for experiments. The detuning-enhanced cavity spin squeezing can also be applied to other systems, such as nitrogen-vacancy centers in diamond, to prepare SSS for quantum metrology.

II. MODEL

The system [Fig. 1(b)] is an ensemble of N identical three-level atoms trapped inside an optical Febry-Pérot cavity. There are two stable ground states $|\uparrow\rangle$ and $|\downarrow\rangle$, which are coupled to the excited state $|e\rangle$ via optical transitions of frequencies $\omega_c \pm \omega_a/2$. The cavity resonance frequency ω_c is chosen so that the detunings to transitions $|\uparrow\rangle \leftrightarrow |e\rangle$ and $|\downarrow\rangle \leftrightarrow |e\rangle$ are opposite in sign but having the same magnitude $\Delta = \omega_a/2$. For simplicity, we only consider the case where the two transitions have equal single-photon Rabi frequency $2g$ and all atoms are uniformly coupled to the cavity. The Hamiltonian of the system reads ($\hbar = 1$)

$$H_{\text{cav}} = \omega_c c^\dagger c + \sum_{i=1}^{N} \left(\frac{\omega_a}{2}[|\uparrow\rangle_i\langle\uparrow|_i - |\downarrow\rangle_i\langle\downarrow|_i] + \omega_c|e\rangle_i\langle e|_i \right.$$
$$\left. + g[c|e\rangle_i\langle\uparrow|_i + c|e\rangle_i\langle\downarrow|_i + \text{H.c.}] \right). \tag{1}$$

Here, c and $c^\dagger$ are the photon annihilation and creation operators for the cavity mode, and the index i labels the individual atoms. As we are interested in the linear and dispersive regime of atom-field interactions, we assume the excited-state population is negligible. The assumption requires a large detuning $|\Delta| \gg \kappa, \Gamma, g$ and sufficiently low intracavity photon number $\langle c^\dagger c\rangle \ll (\Delta/g)^2$, where κ is the

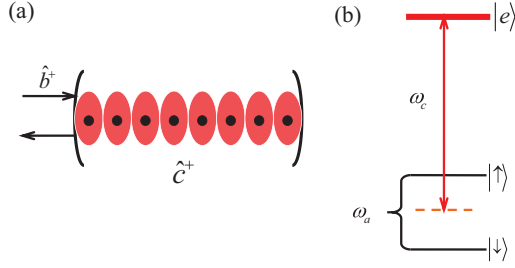*clzou321@ustc.edu.cn

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FIG. 1. (Color online) (a) Schematic illustration of an ensemble of atoms uniformly coupled to optical cavity mode c, and a laser field is driving the cavity. (b) Energy diagram of the atom; the transitions between lower states ($|\downarrow\rangle$ and $|\uparrow\rangle$) and excited state $|e\rangle$ are coupled to the cavity mode.

cavity linewidth and Γ is the excited-state decay rate. After adiabatically eliminating the excited state of the atom and considering external continuum fields [32,33], we obtain the effective Hamiltonian for the system:

$$
H_{\text{eff}} = (\delta + \Omega S_z)c^\dagger c + \int_{-\infty}^{\infty} \omega b_\omega^\dagger b_\omega d\omega
$$
$$
+ \sqrt{\kappa}[\beta_{\text{in}}^*(t)c + c^\dagger \beta_{\text{in}}(t)]
$$
$$
+ \sqrt{\frac{\kappa}{2\pi}} \int_{-\infty}^{\infty} (b_\omega^\dagger c + c^\dagger b_\omega)d\omega, \quad (2)
$$

where $\delta = \omega_c - \omega_l$ is the detuning between the resonator mode and the driving light, $S_z = \frac{1}{2}\sum_{i=1}^{N}(|\uparrow\rangle_i\langle\uparrow|_i - |\downarrow\rangle_i\langle\downarrow|_i)$ is the z component of the total spin, $\Omega = 2g^2/|\Delta|$ is the dispersive frequency shift due to spin-photon interaction, $\beta_{\text{in}}(t)$ is the driving, and b_ω ($b_\omega^\dagger$) is the annihilation (creation) operator of the continuum.

Under coherent laser driving, the intracavity field is the coherent state with spin-dependent phase shift. Assume the system is in state

$$
|\psi\rangle = \sum_m C_m e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'} |\varphi_m(t),m\rangle \prod_\omega |\beta_{\omega,m}(t)\rangle. \quad (3)
$$

Where m is the quantum number associated with S_z, which is conserved during the evolution, $|\varphi_m(t)\rangle$ is the cavity photon state and the $|\beta_{\omega,m}(t)\rangle$ is the state of the continuum. By solving the Schrödinger equation $i\frac{\partial}{\partial t}|\psi\rangle = H_{\text{eff}}|\psi\rangle$ (Appendix A), the time dependent intracavity field [30] is in the state with the complex amplitude

$$
\varphi_m(t) = -i\sqrt{\kappa}\int_0^t \beta_{\text{in}}(t')e^{-i(\delta+\Omega m)(t-t')}e^{-\kappa(t-t')/2}dt', \quad (4)
$$

and the continuum modes are in the states with the complex amplitudes

$$
\beta_{\omega,m}(t) = -i\sqrt{\frac{\kappa}{2\pi}}\int_0^t \varphi_m(t')e^{-i\omega(t-t')}dt'. \quad (5)
$$

In general, the cavity photon, continuum, and atomic spin states are entangled [Eq. (3)]. If the output field is not measured, the density matrix of the cavity photon and the

atomic spin can be written as

$$
\rho_{\text{in,atom}} = \sum_{m,n} C_m C_n^* e^{\phi_{m,n}(t)} |\varphi_m(t),m\rangle\langle\varphi_n(t),n|, \quad (6)
$$

by tracing the continuum modes out, where

$$
\phi_{m,n}(t) = -i\int_0^t \sqrt{\kappa}\text{Re}[\beta_{\text{in}}^*(t')\varphi_m(t') - \beta_{\text{in}}(t')\varphi_n^*(t')]dt'
$$
$$
- \kappa\int_0^t |\varphi_n(t')|^2 dt'/2 - \kappa\int_0^t |\varphi_m(t')|^2 dt'/2
$$
$$
+ \kappa\int_0^t \varphi_n^*(t')\varphi_m(t')dt'. \quad (7)
$$

The spin squeezing is evaluated by squeezing parameter [12]

$$
\xi_s^2 = \frac{\min(\Delta S_{\vec{n}_\perp}^2)}{S/2}, \quad (8)
$$

where $\Delta S_{\vec{n}_\perp}^2$ is the variance of spin operators along the direction perpendicular to the mean-spin direction $\vec{n}_0 = \frac{\vec{s}}{|\langle\vec{s}\rangle|}$, which is determined by the expectation values $\langle S_\alpha\rangle$, with $\alpha \in \{x,y,z\}$. For an atomic system initialized in a coherent spin state (CSS) [34] along the x axis, satisfying $S_x|\psi(0)\rangle_{\text{atom}} = S|\psi(0)\rangle_{\text{atom}}$, we have $C_m = 2^{-S}\sqrt{\frac{(2S)!}{(S-m)!(S+m)!}}$ and $\Delta S_{\vec{n}_\perp}^2 = S/2$. Thus, for squeezed spin states we have $\xi_s^2 < 1$.

III. DETUNING-ENHANCED SQUEEZING

Now, we study the cavity spin squeezing with continuous drive $\beta_{\text{in}}(t) = i\sqrt{\kappa}\beta_0$ with a small detuning $\delta = -\kappa/2$. For easier illustration, it is useful to introduce the dimensionless shearing strength [23]:

$$
Q = \frac{4S|\beta_0|^2\Omega^2 t}{\kappa}, \quad (9)
$$

which is proportional to the transformation degree from the optical field to the atomic spin. In Fig. 2(a), we plot the spin squeezing parameter ξ_s^2 as a function of shearing strength Q for various coupling Ω. It clearly shows that the spin squeezing parameter has a minimal value for certain optimal Q, and it takes a longer time for smaller coupling Ω. The minimal value of the spin squeezing parameter increases with the coupling Ω, because there are higher-order effects associated with Ω that will limit the squeezing.

To study the effect of the detuning δ on spin squeezing, we set $\delta = -x\kappa/2$, and the dimensionless shearing strength can be generalized as

$$
Q_x = 4Qx/(1+x^2)^2. \quad (10)
$$

In Fig. 2(b), we plot the spin squeezing parameter ξ_s^2 as a function of shearing strength Q_x for various detuning δ with fixed coupling strength $\Omega = 0.2$ MHz. The spin squeezing can be enhanced for both red and blue large detuning δ. Since the larger detuning means that it is difficult for the driving light to enter into the cavity, the larger input power or longer interaction time is required. It can be seen from Figs. 2(a) and 2(b) that the atomic spin can be squeezed more than once until the atomic spin is fully uncorrelated. This oscillation
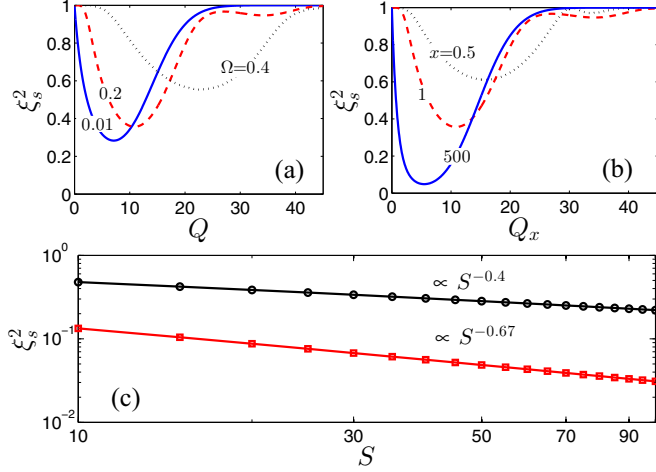
FIG. 2. (Color online) (a) The squeezing parameter ξ_s^2 as a function of shearing strength Q for $S = 50$, $\delta = -\kappa/2$ and various coupling $\Omega = 0.01$, 0.2, 0.4 MHz. (b) The squeezing parameter ξ_s^2 as a function of shearing strength Q_x for $S = 50$, $\Omega = 0.2$ MHz and various detuning $\delta = -x\kappa/2$, $x = 0.5$, 1, 500. (c) The optimal squeezing parameter ξ_s^2 as a function of the atomic spin S for $\Omega = 0.01$ MHz and the detuning $\delta = -x\kappa/2$, $x = 1$ (black), $x = 500$ (red). The other parameters are $\kappa = 4$ MHz and $\beta_0 = 1$.

behavior is due to the competition between the effective spin squeezing interaction, higher-order effects, and decoherence.

In Fig. 2(c), we plot the optimal spin squeezing as a function of the number of spins S, and the optimal spin squeezing is the minimum value of $\xi_s^2(Q_x)$. The black line shows the optimal squeezing parameter $\xi_s^2 \propto S^{-2/5}$ with the small detuning $\delta = -\kappa/2$, as obtained in Ref. [23]. When we choose the large detuning $\delta = -250\kappa$, the optimal squeezing parameter is obtained as the red line, which satisfies $\xi_s^2 \propto S^{-2/3}$. Obviously, the spin squeezing is greatly enhanced by the detuning, approaching the fundamental limitation of the one-axis spin squeezing [12].

IV. MECHANISM

The Hamiltonian Eq. (2) implies that the atom-photon interaction induces a spin state dependent geometric phase $\int dt \langle \varphi_m(t), m | \frac{\partial}{\partial t} | \varphi_m(t), m \rangle$ [30,31]. The spin squeezing is caused by the accumulated geometric phase difference $\phi_{m,n}$ between the different spin states $|m\rangle$ and $|n\rangle$. For continuous laser driving and long interaction time $t \gg \kappa^{-1}$, the intracavity field transient behavior can be neglected. The steady cavity field for detuning $\delta = -x\kappa/2$ can be written as

$$\varphi_m = \frac{\kappa \beta_0}{\kappa/2 + i(\delta + \Omega m)}. \tag{11}$$

From Eq. (7), we solve the phase factor as

$$\phi_{m,n}(t) = i \frac{|\varphi_m|^2 |\varphi_n|^2 \Omega^2 t}{\kappa \beta_0^2} \left\{ \frac{\frac{\kappa^2}{4} + \delta^2}{\Omega \kappa}(n - m) + \frac{\delta}{\kappa}(n^2 - m^2) \right.$$
$$\left. + \frac{\Omega}{\kappa} nm(n - m) + \frac{i(n - m)^2}{2} \right\}. \tag{12}$$

The first term accounts for the coefficient that is approximately proportional to Q_x, and the terms within the brace are the linear, quadratic, and higher-order couplings of the spin z component. The quadratic term corresponds to spin squeezing interaction S_z^2, while the last two terms give rise to disorder and decoherence of spin states. It is obvious that the detuning is essential in the cavity induced spin squeezing, as there is no squeezing at all for zero detuning $\delta = 0$. The parameters $\frac{\delta}{\kappa}$ should be as large as possible to make the squeezing effect outperform the undesired effects, i.e., $\frac{\delta}{\kappa} \gg 1$ and $\frac{\delta}{\kappa} \gg \frac{\Omega}{\kappa}$ should be satisfied. This can explain the results of the dependence of optimal spin squeezing on δ and Ω shown in Figs. 2(a) and 2(b).

(1) For very large Q or Q_x, the disorder and decoherence dominate over the coherent process.

(2) Larger δ help to suppress both disorder and dissipation.

(3) Smaller Ω can suppress the high-order terms and thus can enhance the squeezing.

For a more intuitive understanding, we obtain the spin squeezing parameter ξ_s^2 from the Heisenberg equation [23] under certain approximation

$$\frac{\Omega}{\kappa}|Sz|\frac{1 + |x|}{1 + x^2} \leqslant \frac{\Omega}{\kappa}\sqrt{S/2}\frac{1 + |x|}{1 + x^2} \ll 1, \tag{13}$$

$$1 \ll |Q_x| \ll S, \tag{14}$$

$$\xi_s^2 = \frac{1}{Q_x^2} + \frac{2}{Q_x x} + \frac{Q_x^4}{24 S^2}, \quad x \neq 0. \tag{15}$$

When $(5/2)^{5/4}12^{-1/4}S^{-1/2} \leqslant x \ll 12^{1/6}S^{1/3}$, we obtain the optimal cavity squeezing $\xi_{s,\min}^2 = (5/2)12^{-1/5}S^{-2/5}x^{-4/5}$ at the point $Q_x = 12^{1/5}S^{2/5}x^{-1/5}$. When the detuning is very large $x \gg 12^{1/6}S^{1/3}$, the squeezing limit is $\xi_{s,\min}^2 = (3/2)12^{-1/3}S^{-2/3}$ with $Q_x = 12^{1/6}S^{1/3}$. The detuning is the source of the effect nonlinear interactions between the atomic spin and the optical mode, and the part $2/(Q_x x)$ is the photon shot noise [23]. The large detuning means that the $1/Q_x^2$ is the main factor of spin squeezing rather than the photon shot noise, and we can improve the scaling of cavity squeezing to $(3/2)12^{-1/3}S^{-2/3}$ with sufficient detuning by suppressing the photon shot noise.

V. IMPERFECTIONS

In previous studies, we have neglected the scattering of the photon into free space, which is an unavoidable process that deteriorates squeezing performance [22]. Any atom scattering the photon into free space will acquire a random phase, so that it no longer contributes to the mean spin length. The Raman transitions $|\uparrow\rangle \to |e\rangle \to |\downarrow\rangle$ or $|\downarrow\rangle \to |e\rangle \to |\uparrow\rangle$ reduce the correlation between the time average $\overline{S_z}$ during the cavity squeezing process. The average photon number emitted into free space per atom is given by (Appendix B)

$$R_x = Q_x(1 + x^2)/(8xS\eta), \tag{16}$$

which depends on the single-atom cooperativity $\eta = 4g^2/(\kappa\Gamma)$. This expression indicates that very large collective cooperativity $S\eta \gg 1$ is required to suppress the scattering of the cavity photon into free space. We extend the

solution previously obtained in Ref. [22] to the large detuning, and obtain the spin squeezing parameter:

$$\xi_s^2 = \frac{\langle \widetilde{S_y}^2 \rangle + \langle S_z^2 \rangle - \sqrt{\langle \widetilde{S_y}^2 \rangle - \langle S_z^2 \rangle + W^2}}{S}, \quad (17)$$

where $W = \langle \widetilde{S_y} S_z + S_z \widetilde{S_y} \rangle$ and the mean values of spin operators $\widetilde{S}$ are solved approximately in the rotating frame as

$$\langle \widetilde{S_y} \rangle = \langle S_z \rangle = 0, \quad \langle S_z^2 \rangle = \frac{S}{2}, \quad (18)$$

$$\langle \widetilde{S_y}^2 \rangle = \frac{S}{2}[1 + Se^{-4R_x}(1 - e^{-U})], \quad (19)$$

$$\langle \widetilde{S_y} S_z + S_z \widetilde{S_y} \rangle = S(1 - R_x)Q_x e^{-V}, \quad (20)$$

with parameters $U = \frac{2Q_x}{xS} + \frac{Q_x^2(1-2R_x/3)}{S}$, $V = \frac{Q_x}{2xS} + 2R_x + \frac{Q_x^2(1-2R_x/3)}{4S}$.

Although ξ_s^2 is a complicated function of η, x, and Q_x due to imperfection, the spin squeezing can be optimized for a given η by adjusting the laser detuning and pump power and interacting time. Figure 3(a) shows the squeezing parameter ξ_s^2 as a function of Q_x for various values of the cooperativity η and fixed large detuning $x = 200$, and in Fig. 3(b) the optimized spin squeezing parameters for certain Q_x are calculated against detuning x for given cooperativity η. These results indicate that the squeezing parameter is very sensitive to the value of the

FIG. 3. (Color online) (a) The squeezing parameter ξ_s^2 as a function of Q_x for fixed detuning $x = 200$, and single-atom cooperativity η=2, 20, ∞. (b) The optimal squeezing parameter ξ_s^2 as a function of detuning x for the various cooperativity $\eta = 1, 2, 20$. (c) The solid lines are optimal squeezing parameter ξ_s^2 as a function of the cooperativity η for the fixed detuning $x = 1$ (green) and optimized detuning (black), and the dashed lines are results for ideal condition $\eta = \infty$ for $x = 1$ (red) and $x = 200$ (blue). The atomic spin is $S = 10^4$.

cooperativity η, and better spin squeezing can be achieved for larger η and appropriate detuning x. Shown in Fig. 3(c) is the optimal squeezing parameter ξ_s^2 as a function of η. Green and black solid lines are the results for fixed detuning ($x = 1$) and optimized detuning. With increasing η, ξ_s^2 is reduced and tends to be saturated at a certain value. Compared with the fixed detuning, the optimal detuning is always better. When the cooperativity is not too small, $\eta > 0.1$, the squeezing by optimized detuning can be even better than the result of fixed detuning with $\eta = \infty$.

To lowest-order expansion of $R_x \ll 1$ and ignoring curvature effects for the moment, the asymptotic solution of the squeezing parameter [Eq. (17)] can be written as

$$\xi_s^2 = Q_x^{-2} + \frac{2}{Q_x x} + \frac{Q_x(x^2 + 1)}{6xS\eta}. \quad (21)$$

When δ is very small, the squeezing variance suppressed by the square of the shearing strength is neglected. Consequently, there is an optimum shearing strength $Q_{\text{scatt}} = \sqrt{12S\eta/(x^2 + 1)}$, to achieve the optimum squeezing $\xi_s^2 = \sqrt{\frac{4(x^2+1)}{3S\eta x^2}}$. For very large detuning that satisfies $x \gg 12^{1/6}S^{1/3}$, we have optimum squeezing $\xi_s^2 = 3(\frac{1+x^2}{12xS\eta})^{2/3}$ for a shearing strength $Q_{\text{scatt}} = (\frac{12xS\eta}{1+x^2})^{1/3}$. The squeezing is thus possible even for a very weakly coupled resonator and atoms with single photon-atom coupling cooperativity $\eta \ll 1$, as long as the collective cooperativity $S\eta \gg 1$. Similar results can also be found for spin squeezing with the near-resonance laser input [23,35].

In practical terms, we should also consider the spin dephasing during the preparation of SSS. For simplicity, we assume that spin dephasing is Markovian with the pure dephasing rate T^{-1}. Including it in the master equation (Appendix C), we obtain the modified squeezing parameter $\xi_s'^2 = \xi_s^2 + \frac{t}{T}$ under the approximation $\frac{t}{T} \ll 1$. When the single spin dephasing rate is large, obviously the larger input power is the better choice rather than longer interaction time. In addition, the large detuning also means that the second cavity mode (Appendix D) may be activated, and our analysis shows that the opposite detuning for the second cavity mode has a bad effect on the spin squeezing. However, the effective coupling between the spin and the second cavity mode is very small, and the bad effect can be ignored. We also verify that the effect of the second cavity mode can be ignored by an example using the achievable parameters (Appendix D) from the Vuletić group's experiment [24].

VI. CONCLUSION

We have theoretically analyzed unconditional squeezing of the collective spin of an atomic ensemble in a driven optical cavity. We find that strong atom-cavity coupling weakens the spin squeezing, while large detuned laser driving can improve the scaling of spin squeezing to $S^{-2/3}$, which is the ultimate limit of the ideal one-axis twisting spin squeezing. The imperfection of light scattering into free space can also be efficiently suppressed by optimal detuning. The detuning-enhanced cavity spin squeezing can be tested experimentally and be applied for quantum metrology based on the SSS.
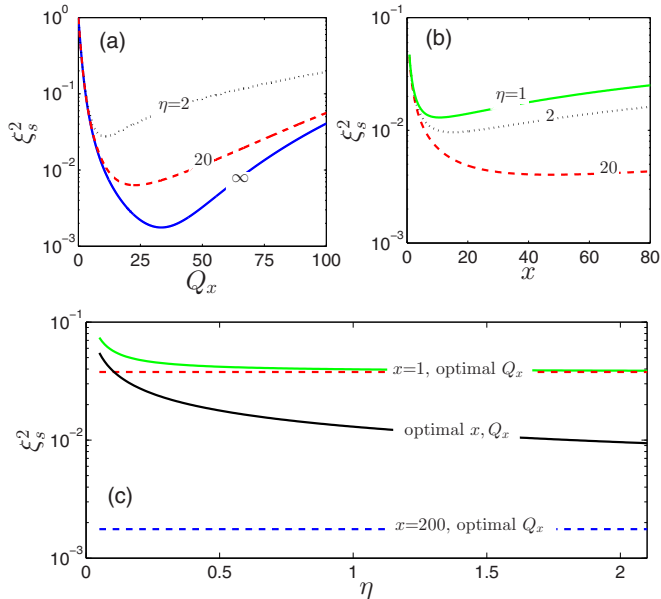
APPENDIX A: DYNAMICS OF QUANTUM STATES

The dynamics of the quantum state satisfy Schrodinger equation

$$i\frac{\partial}{\partial t}|\psi\rangle = H_{\text{eff}}|\psi\rangle. \tag{A1}$$

Substituting the effective Hamiltonian [Eq. (2)] and quantum state [Eq. (3)] into the above equation, the right- and left-hand sides of the equation become

$$H_{\text{eff}}|\psi\rangle = \sum_m C_m \left\{ (\delta+\Omega m)c^\dagger c + \sqrt{\kappa}[\beta_{\text{in}}^*(t)c + c^\dagger\beta_{\text{in}}(t)] + \int_{-\infty}^\infty \omega' b_{\omega'}^\dagger b_{\omega'} d\omega' + \sqrt{\frac{\kappa}{2\pi}}\int_{-\infty}^\infty (b_{\omega'}^\dagger c + c^\dagger b_{\omega'})d\omega' \right\}$$

$$\times e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}|\varphi_m(t),m\rangle\prod_\omega |\beta_{\omega,m}(t)\rangle, \tag{A2}$$

$$i\frac{\partial}{\partial t}|\psi\rangle = \sum_m C_m \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t)\varphi_m(t))e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}|\varphi_m(t),m\rangle\prod_\omega |\beta_{\omega,m}(t)\rangle$$

$$+ i\sum_m C_m e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}\left(\frac{\partial}{\partial t}|\varphi_m(t),m\rangle\right)\prod_\omega |\beta_{\omega,m}(t)\rangle$$

$$+ i\sum_m C_m e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}|\varphi_m(t),m\rangle\frac{\partial}{\partial t}\prod_\omega |\beta_{\omega,m}(t)\rangle, \tag{A3}$$

where the cavity field follows

$$\varphi_m(t) = -i\sqrt{\kappa}\int_0^t \beta_{\text{in}}(t')e^{-i(\delta+\Omega m)(t-t')}e^{-\kappa(t-t')/2}dt'. \tag{A4}$$

Multiplying $\langle\varphi_m(t),m|$ on both sides of the Schrodinger equation, we have

$$\langle\varphi_m(t),m|H_{\text{eff}}|\psi\rangle = C_m e^{i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}\left\{ (\delta+\Omega m)|\varphi_m(t)|^2 + \int_{-\infty}^\infty \omega' b_{\omega'}^\dagger b_{\omega'} d\omega' + \sqrt{\frac{\kappa}{2\pi}}\int_{-\infty}^\infty (b_{\omega'}^\dagger\varphi_m(t) + \varphi_m^*(t)b_{\omega'})d\omega' \right.$$

$$\left. + \sqrt{\kappa}[\beta_{\text{in}}^*(t)\varphi_m(t) + \varphi_m^*(t)\beta_{\text{in}}(t)] \right\}\prod_\omega |\beta_{\omega,m}(t)\rangle, \tag{A5}$$

and

$$\langle\varphi_m(t),m|i\frac{\partial}{\partial t}|\psi\rangle = C_m e^{-i\int_0^t \text{Re}(\sqrt{\kappa}\beta_{\text{in}}^*(t')\varphi_m(t'))dt'}\left\{ \text{Re}[\sqrt{\kappa}\beta_{\text{in}}^*(t)\varphi_m(t)] \right.$$

$$\left. + i\langle\varphi_m(t),m|\left(\frac{\partial}{\partial t}|\varphi_m(t),m\rangle\right) + i\frac{\partial}{\partial t} \right\}\prod_\omega |\beta_{\omega,m}(t)\rangle. \tag{A6}$$

Note that

$$\langle\varphi_m(t),m|\frac{\partial}{\partial t}|\varphi_m(t),m\rangle = \frac{1}{2}\left(\varphi_m^*(t)\frac{\partial\varphi_m(t)}{\partial t} - \varphi_m(t)\frac{\partial\varphi_m^*(t)}{\partial t}\right) = \frac{1}{2}\sqrt{\kappa}[-i\varphi_m^*(t)\beta_{\text{in}}(t) - i\varphi_m(t)\beta_{\text{in}}^*(t)] - i(\delta+\Omega m)|\varphi_m(t)|^2, \tag{A7}$$

is the *geometry phase* of the system. Since

$$i\frac{\partial}{\partial t}\prod_\omega |\beta_{\omega,m}(t)\rangle = \left\{ (\delta+\Omega m)|\varphi_m(t)|^2 - i\langle\varphi_m(t),m|\left(\frac{\partial}{\partial t}|\varphi_m(t),m\rangle\right) + \int_{-\infty}^\infty\left[\omega' b_{\omega'}^\dagger b_{\omega'} + \sqrt{\frac{\kappa}{2\pi}}(b_{\omega'}^\dagger\varphi_m(t) + \varphi_m^*(t)b_{\omega'})\right]d\omega' \right.$$

$$\left. + \sqrt{\kappa}[\beta_{\text{in}}^*(t)\varphi_m(t) + \varphi_m^*(t)\beta_{\text{in}}(t)]/2 \right\}\prod_\omega |\beta_{\omega,m}(t)\rangle, \tag{A8}$$

we get

$$i\frac{\partial}{\partial t}\prod_\omega |\beta_{\omega,m}(t)\rangle = \left(\int_{-\infty}^\infty\left\{\omega' b_{\omega'}^\dagger b_{\omega'} + \sqrt{\frac{\kappa}{2\pi}}[b_{\omega'}^\dagger\varphi_m(t) + \varphi_m^*(t)b_{\omega'}]\right\}d\omega'\right)\prod_\omega |\beta_{\omega,m}(t)\rangle. \tag{A9}$$

Therefore, we obtain the solution of the continuum modes with the complex amplitudes as

$$\beta_{\omega,m}(t) = -i\sqrt{\frac{\kappa}{2\pi}} \int_0^t \varphi_m(t')e^{-i\omega(t-t')}dt'. \tag{A10}$$

Based on these results, the density operator of the inner cavity and the atomic spin can be calculated by

$$\rho_{\text{in,atom}} = \text{Tr}_{\text{out}}(|\psi\rangle\langle\psi|) = \sum_{m,n} C_m C_n^* e^{-i\sqrt{\kappa}\int_0^t \text{Re}(\beta_{\text{in}}^*(t')\varphi_m(t')-\beta_{\text{in}}(t')\varphi_n^*(t'))dt'} |\varphi_m(t),m\rangle\langle\varphi_n(t),n| \prod_\omega \langle\beta_{\omega,n}|\beta_{\omega,m}\rangle. \tag{A11}$$

Finally, the density operator of the inner cavity and the atomic spin can be written as

$$\rho_{\text{in,atom}} = \sum_{m,n} C_m C_n^* e^{\phi_{m,n}(t)} |\varphi_m(t),m\rangle\langle\varphi_n(t),n|, \tag{A12}$$

with the phase factor

$$\phi_{m,n}(t) = -i\sqrt{\kappa}\int_0^t \text{Re}[\beta_{\text{in}}^*(t')\varphi_m(t') - \beta_{\text{in}}(t')\varphi_n^*(t')]dt' - \kappa\int_0^t |\varphi_n(t')|^2 dt'/2 - \kappa\int_0^t |\varphi_m(t')|^2 dt'/2 + \kappa\int_0^t \varphi_n^*(t')\varphi_m(t')dt'. \tag{A13}$$

APPENDIX B: SCATTERING INTO FREE SPACE

By excluding the continuum modes, we obtain the effective Hamiltonian:

$$\widetilde{H}_{\text{eff}} = (\delta + \Omega S_z)c^\dagger c + \sqrt{\kappa}[\beta_{\text{in}}^*(t)c + c^\dagger\beta_{\text{in}}(t)]. \tag{B1}$$

The master equation reads

$$\frac{d\rho}{dt} = i[\rho,\widetilde{H}_{\text{eff}}] + \sum_{i=1}^N \Gamma[L(|\uparrow\rangle_i\langle e|_i)\rho + L(|\downarrow\rangle_i\langle e|_i)\rho]$$
$$+ \kappa L(c)\rho, \tag{B2}$$

where Lindblad superoperator

$$L(\hat{o})\rho = \hat{o}\rho\hat{o}^+ - \tfrac{1}{2}\hat{o}^+\hat{o}\rho - \tfrac{1}{2}\rho\hat{o}^+\hat{o}. \tag{B3}$$

The equations of motion are

$$\frac{dc}{dt} = -i(\delta + \Omega S_z)c - \frac{\kappa}{2}c - i\sqrt{\kappa}\beta_{\text{in}}, \tag{B4}$$

$$\frac{dS_z}{dt} = -\frac{\Gamma g^2 c^+ c}{\Delta^2}S_z, \tag{B5}$$

$$\frac{dS_+}{dt} = i\Omega c^+ S_+ c - \frac{\Gamma g^2 c^+ c}{\Delta^2}S_+. \tag{B6}$$

Then, the steady-state solution is

$$c \approx \frac{i\sqrt{\kappa}\beta_{\text{in}}}{-i(\delta + \Omega\overline{S_z}) - \frac{\kappa}{2}}, \tag{B7}$$

$$S_z \approx e^{-2R_x}S_z(0), \tag{B8}$$

$$S_+ \approx e^{2i\Omega\frac{\beta_0^2}{(1+x^2)^2}[1+x^2+\frac{4\Omega\overline{S_z}x}{\kappa}+\frac{2\Omega(x+i)}{\kappa}]t-2R_x}S_+(0), \tag{B9}$$

where $\overline{S_z} = \int S_z$, $\beta_{\text{in}} = i\sqrt{\kappa}\beta_0$, and $R_x = Q_x(1+x^2)/(8xS\eta)$. In the rotating frame, by eliminating the quickly linear rotating along z, we obtain the mean value of operators

as

$$\widetilde{S_+} = e^{-2i\Omega\frac{\beta_0^2}{(1+x^2)^2}[1+x^2+\frac{2\Omega x}{\kappa}]t}S_+$$
$$= e^{-Q_x/(2Sx)}e^{iQ_x\overline{S_z}/S}e^{-2R_x}S_+(0), \tag{B10}$$

$$\widetilde{S_+}^2 = e^{-Q_x(2+i)/(Sx)}e^{2iQ_x\overline{S_z}/S}e^{-4R_x}S_+^2(0). \tag{B11}$$

These results can be approximated as

$$\langle\widetilde{S_y}\rangle \approx \langle S_z\rangle = 0, \quad \langle S_z^2\rangle = \frac{S}{2}, \tag{B12}$$

$$\langle\widetilde{S_y}^2\rangle \approx \frac{S}{2}[1 + Se^{-4R_x}(1 - e^{-U})], \tag{B13}$$

$$\langle\widetilde{S_y}S_z + S_z\widetilde{S_y}\rangle \approx S(1 - R_x)Q_x e^{-V}, \tag{B14}$$

where $U = \frac{2Q_x}{xS} + \frac{Q_x^2(1-2R_x/3)}{S}$, $V = \frac{Q_x}{2xS} + 2R_x + \frac{Q_x^2(1-2R_x/3)}{4S}$.

APPENDIX C: SINGLE SPIN DEPHASING

If we consider the effect of single spin dephasing, the master equation is written as

$$\frac{d\rho}{dt} = i[\rho,H_{\text{eff}}] + \sum_{i=1}^N \Gamma[L(|\uparrow\rangle_i\langle e|_i)\rho + L(|\downarrow\rangle_i\langle e|_i)\rho]$$
$$+ \kappa L(c)\rho + \frac{1}{2T}\sum_i L(\sigma_i^z)\rho, \tag{C1}$$

where the single spin dephasing T^{-1} is assumed to be Markovian for simplicity. By including the dephasing, Eq. (B6) is changed to

$$\frac{dS_+}{dt} = i\Omega c^+ S_+ c - \frac{\Gamma g^2 c^+ c}{\Delta^2}S_+ - \frac{1}{T}S_+. \tag{C2}$$

With a similar approach to derive Eq. (B11), we obtain the expected value of operators as

$$\widetilde{S}_+ = e^{-Q_x/(2Sx)+iQ_x\overline{S_z}/S-2R_x-\frac{t}{T}}S_+(0),$$

$$\widetilde{S}_+^2 = e^{-Q_x(2+i)/(Sx)+2iQ_x\overline{S_z}/S-4R_x-\frac{2t}{T}}S_+^2(0). \quad (C3)$$

Usually, the decoherence is very small. Thus, we can obtain the spin squeezing under the approximation $\frac{t}{T} \ll 1$ as

$$\xi_s'^2 = \xi_s^2 + \frac{t}{T}. \quad (C4)$$

APPENDIX D: MULTIPLE CAVITY MODES

When the detuning is very large, it becomes comparable with the free spectral range of the cavity ($\Delta\omega_{\mathrm{FSR}}$), and multiple cavity modes should be involved in the interaction with the atomic ensemble. Assuming the nth cavity mode as c_n with resonance frequency ω_n, the full Hamiltonian of the system reads

$$H_{\mathrm{cav}} = \sum_n \omega_n c_n^\dagger c_n$$
$$+ \sum_{i=1}^N \left[\frac{\omega_a}{2}(|\uparrow\rangle_i\langle\uparrow|_i - |\downarrow\rangle_i\langle\downarrow|_i) + \omega_c|e\rangle_i\langle e|_i \right]$$
$$+ \sum_{n=1}\sum_{i=1}^N g(c_n|e\rangle_i\langle\uparrow|_i + c_n|e\rangle_i\langle\downarrow|_i + \mathrm{H.c.}). \quad (D1)$$

Under the ordinary condition that $|\Delta \pm (\omega_n - \omega_c)| \gg \kappa, \Gamma, g$, we can obtain the dispersive frequency shift due to the nth cavity mode:

$$\Omega_n = \frac{2g^2\Delta}{\Delta^2 - (\omega_n - \omega_c)^2}. \quad (D2)$$

It is easy to obtain the $Q_{x,n}$ [Eq. (10)] for the nth cavity mode by using $\delta_n = \omega_n - \omega_l$. Then, we get the solutions $U = \frac{2}{S}\sum_{n=1}\frac{Q_{x,n}}{x_n} + \frac{(\sum_{n=1}Q_{x,n})^2[1-2(\sum_{n=1}R_{x,n})/3]}{S}$, $V = \frac{1}{2S}\sum_{n=1}\frac{Q_{x,n}}{x_n} + 2\sum_{n=1}R_{x,n} + \frac{(\sum_{n=1}Q_{x,n})^2[1-2(\sum_{n=1}R_{x,n})/3]}{4S}$,

and

$$\sum_{n=1}\frac{Q_{x,n}}{x_n} = \sum_{n=1}\frac{16S|\beta_0|^2\Omega_n^2 t}{\kappa\left(1+x_n^2\right)^2}, \quad (D3)$$

$$\sum_{n=1}R_{x,n} = \sum_{n=1}\frac{2|\beta_0|^2\Omega_n^2 t}{\kappa\eta\left(1+x_n^2\right)}, \quad (D4)$$

$$\sum_{n=1}Q_{x,n} = \sum_{n=1}\frac{16S|\beta_0|^2\Omega_n^2 x_n t}{\kappa\left(1+x_n^2\right)^2}. \quad (D5)$$

If we only consider two cavity modes for $\omega_1 = \omega_c$ and ω_2 satisfies the condition $|\Delta \pm (\omega_{1,2} - \omega_c)| \gg \kappa, \Gamma, g$, the decoherence terms are enhanced due to the additional cavity mode. For the opposite detuning x_1, x_2, the spin squeezing term is weakened. However, due to $\Omega_1 \gg \Omega_2$ for the second cavity mode, $|\omega_2 - \omega_c| = \Delta\omega_{\mathrm{FSR}} \gg \Delta$. For the Fabry-Perot cavity, $\Delta\omega_{\mathrm{FSR}} = \pi\frac{C}{L}$ with C the speed of light and L the length of the cavity. For a practical experiment, it very easy to engineer the cavity length to satisfy the condition $\Delta\omega_{\mathrm{FSR}} \gg \Delta$. Then, the relation

$$\left|\frac{Q_{x,1}}{Q_{x,2}}\right| = \frac{|x_1|\left(1+x_2^2\right)^2}{|x_2|\left(1+x_1^2\right)^2}\frac{[\Delta^2 - (\omega_2 - \omega_c)^2]^2}{\Delta^4}. \quad (D6)$$

If the condition $\Delta\omega_{\mathrm{FSR}} \gg \Delta$ is not well satisfied, we should have $|x_2| \gg |x_1|$. So, $|\frac{Q_{x,1}}{Q_{x,2}}| \gg 1$ is satisfied for practical experiments, and we can neglect the effect of the second cavity mode.

To verify the feasibility of the single mode approximation, we take the Vuletić group's experiment [24] as an example. In the experiment, the parameters are $\Delta \approx 3.4$ GHz, $\Delta\omega_{\mathrm{FSR}} \approx 5.6$ MHz, and cavity linewidth $\kappa = 4$ MHz. Following the discussion above, the detuning of the laser to the nth cavity mode is $\omega_n - \omega_l = -x_n\kappa/2$. For large detuning to the nth mode, let $x_1 = 100$, and the detunings to $(n-1)$th and $(n+1)$th modes are $x_{n-1} = 2900$ and $x_{n+1} = 2700$. Therefore, we have $|\frac{Q_{x,n}}{Q_{x,n\pm1}}| \sim 10^4 \gg 1$, and therefore the effect of other cavity modes can be neglected.

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