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Generating Long-Lived Macroscopically Distinct Superposition States in Atomic Ensembles

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We propose to create and stabilize long-lived macroscopic quantum superposition states in atomic ensembles. We show that using a fully quantum parametric amplifier can cause the simultaneous decay of two atoms, and in turn create stabilized atomic Schrödinger cat states. Remarkably, even with modest parameters these intracavity atomic cat states can have an extremely long lifetime, up to *four orders of magnitude* longer than that of intracavity photonic cat states under the same parameter conditions, reaching *tens of milliseconds*. This lifetime of atomic cat states is ultimately limited to *several seconds* by extremely weak spin relaxation and thermal noise. Our work opens up a new way towards the long-standing goal of generating large-size and long-lived cat states, with immediate interests both in fundamental studies and noise-immune quantum technologies.

Introduction.—Schrödinger cat states, which are macroscopically-distinct superposition states, express the essence of quantum mechanics. Such states are appealing not only for fundamental studies of quantum mechanics [1, 2], but also for wide applications ranging from quantum metrology [3, 4] to quantum computation [5–9]. So far, a large number of approaches [10–24] have been proposed to generate cat states. However, these cat states (especially of large size) are extremely fragile in a noisy environment, and their fast decoherence makes them no longer practical for applications. Thus, the ability to stabilize cat states, as an essential prerequisite for their various applications, is highly desirable. To address this problem, two-photon loss has been engineered [25–28], and recently experimentally demonstrated [29–33]. Such a nonlinear loss can protect cat states against photon dephasing [28, 34]; but, unfortunately, *not* against the unavoidable single-photon loss. This implies a significantly limited cat-state lifetime. Single-photon loss has been considered to be the major source of noise in fault-tolerant quantum computation based on cat states [5–9]. Thus, the stabilization of large-size cat states for a long time remains challenging.

Ensembles of atoms or spins have negligible spin relaxation, and instead their major source of noise is spin dephasing, i.e., collective dephasing, local dephasing, and inhomogeneous broadening. This motivates us to engineer the *simultaneous* decay of two atoms of an ensemble (here denoted as two-atom decay), and then use it to stabilize atomic cat states. Such cat states could have a very long lifetime since the two-atom decay can protect them against spin dephasing, in close analogy to the mechanism of using two-photon loss to suppress

photon dephasing.

To implement the two-atom decay that is still lacking and fundamentally different from two-photon loss, here we propose to exploit fully quantum degenerate parametric amplification. More importantly, the lifetime of the resulting atomic cat states can be made longer, by up to *four orders of magnitude*, than that of common intracavity photonic cat states (see Table I in [35]), i.e., equal superpositions of two opposite-phase coherent states. To ensure a fair comparison, these photonic cat states need to have the same size as our atomic cat states and also suffer from single-photon loss of the same rate as given for the signal mode. With a modest cavity decay time ($\sim 16 \mu\text{s}$), our cat state lifetime can reach ~ 20 ms. This is comparable to 17 ms [36], which is the longest lifetime of intracavity photonic cat states to date, but which was achieved with an extreme cavity decay time (~ 0.13 sec). As the cavity decay time increases, our cat state lifetime can further increase and ultimately is limited to a maximum value determined by spin relaxation and thermal noise. For a typical spin relaxation time ~ 40 sec [37, 38], we can predict a maximum cat state lifetime of ~ 3 sec.

Physical model.—The central idea is illustrated in Fig. 1(a). To consider degenerate parametric amplification in the fully quantum regime, our system, inspired by recent experimental advances [30–32, 39, 40], contains two parametrically coupled cavities: one as a pump cavity with frequency ω_p and the other as a signal cavity with frequency ω_s . We assume that the pump cavity is subject to a coherent drive with amplitude Ω and frequency ω_d . The intercavity parametric coupling J stimulates the conversion between pump single photons and pairs of signal photons. Furthermore, an ensemble of

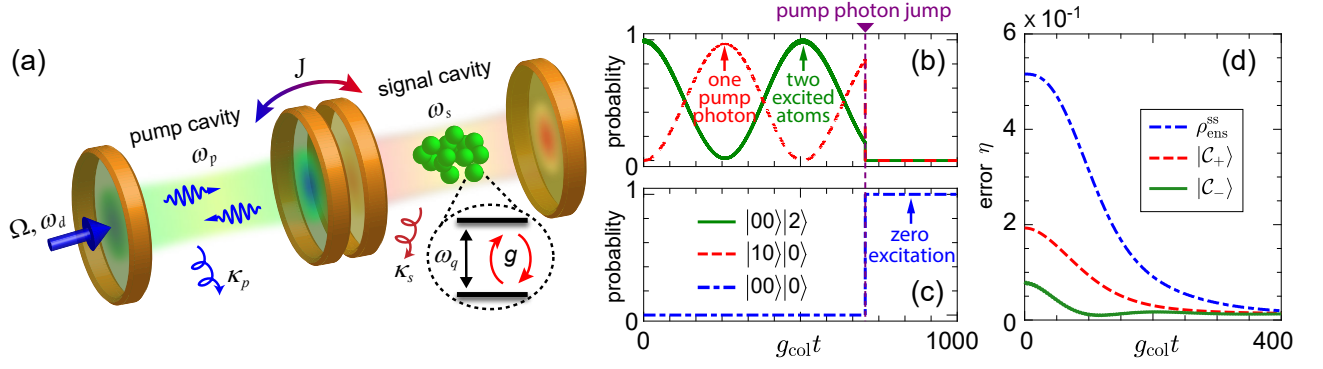


FIG. 1. (a) Schematic setup of our proposal. The pump and signal cavities are coupled via a parametric coupling J , and the atomic ensemble is coupled to the signal cavity with a single-atom coupling g . The pump cavity is subject to a coherent drive with amplitude Ω and frequency ω_d . Here, ω_p, ω_s are the resonance frequencies of the pump and signal cavities, κ_p, κ_s are their respective single-photon loss rates, and ω_q is the atomic transition frequency. (b, c) Quantum Monte-Carlo trajectory pictured through the probabilities of the system being in the states $|m_p 0\rangle|n\rangle$. Initially, only two atoms in the ensemble are excited. Here, $\kappa_p = 0.2\chi$ and $\kappa_s = \Omega = 0$. (d) Time evolution of the preparation error η for a cat size $|\alpha|^2 = 1$. Here, $\kappa_p = 5\chi$, $\kappa_s = 0.3\kappa_p$, and the ensemble is initialized in the ground state $|0\rangle$, the single-excitation state $|1\rangle$, and a spin coherent state $|\theta_0, 0\rangle$ with $\sqrt{N} \tan(\theta_0/2) = 1$, for the states $|\mathcal{C}_+\rangle$, $|\mathcal{C}_-\rangle$, and $\rho_{\text{ens}}^{\text{ss}}$, respectively. In (b)-(d), we assume that $N = 100$, $J = 3g_{\text{col}}$, and both cavities are initialized in the vacuum.

N identical two-level atoms is placed in the signal cavity, and the atomic transition, of frequency ω_q , is driven by a coupling g to the signal photon. When $2\omega_q \approx \omega_p \ll 2\omega_s$, a pair of excited atoms can jointly emit a pump photon. The subsequent loss of the pump photon gives rise to the two-atom decay, which in turn stabilizes large-size, extremely long-lived cat states in the ensemble.

The system Hamiltonian in a frame rotating at ω_d is

$$H = \sum_{i=p,s} \delta_i a_i^\dagger a_i + \delta_q S_z + J (a_p a_s^{\dagger 2} + a_p^\dagger a_s^2) + g (a_s S_+ + a_s^\dagger S_-) + \Omega (a_p + a_p^\dagger), \quad (1)$$

where a_p, a_s are the annihilation operators for the pump and signal modes, $S_\pm = S_x \pm iS_y$, $\delta_p = \omega_p - \omega_d$, $\delta_s = \omega_s - \omega_d/2$, and $\delta_q = \omega_q - \omega_d/2$. The collective spin operators are $S_\alpha = \frac{1}{2} \sum_{j=1}^N \sigma_j^\alpha$, with σ_j^α ($\alpha = x, y, z$) the Pauli matrices for the j th atom. The Lindblad dissipator, $\mathcal{L}(o)\rho = o\rho o^\dagger - \frac{1}{2}o^\dagger o\rho - \frac{1}{2}\rho o^\dagger o$, describes the dissipative dynamics determined by

$$\dot{\rho} = -i[H, \rho] + \sum_{i=p,s} \kappa_i \mathcal{L}(a_i)\rho, \quad (2)$$

where κ_p and κ_s are the photon loss rates of the pump and signal modes. Spin dephasing, spin relaxation, and thermal noise are discussed below.

We assume that $2\omega_q \approx \omega_p \approx \omega_d$, and the detuning $\Delta = \omega_s - \omega_q \gg \{g_{\text{col}}, J\}$. Here, $g_{\text{col}} = \sqrt{N}g$ represents the collective coupling of the ensemble to the signal mode. Then, we can predict a parametric coupling, $\chi = g_{\text{col}}^2 J / \Delta^2$, between atom pairs and pump single photons. Accordingly, the Hamiltonian H , after time averaging [41, 42], becomes

$$H_{\text{avg}} = \frac{\chi}{N} (a_p S_+^2 + a_p^\dagger S_-^2) + \Omega (a_p + a_p^\dagger), \quad (3)$$

which describes a third-order process. The stronger second-order process has been eliminated with an appropriate detuning between ω_p and $2\omega_q$ (see [35]). To derive H_{avg} , we have considered the low-excitation regime, where the average number of excited atoms is much smaller than the total number of atoms.

We now adiabatically eliminate the pump mode a_p , yielding an effective master equation

$$\dot{\rho}_{\text{ens}} = -i[H_{\text{ens}}, \rho_{\text{ens}}] + \frac{\kappa_{1\text{at}}}{N} \mathcal{L}(S_-)\rho_{\text{ens}} + \frac{\kappa_{2\text{at}}}{N^2} \mathcal{L}(S_-^2)\rho_{\text{ens}}, \quad (4)$$

where $H_{\text{ens}} = i\chi_{2\text{at}}(S_-^2 - S_+^2)/N$, and ρ_{ens} represents the reduced density matrix of the ensemble. Here, $\kappa_{2\text{at}} = 4\chi^2/\kappa_p$ and $\chi_{2\text{at}} = 2\Omega\chi/\kappa_p$ are the rates of the simultaneous decay and excitation of two atoms, respectively. Moreover, $\kappa_{1\text{at}} = (g_{\text{col}}/\Delta)^2 \kappa_s$ is the rate of the Purcell single-atom decay (see [35]), and we can tune it to be $\ll \kappa_{2\text{at}}$, as long as $\kappa_s \ll (g_{\text{col}}J/\Delta)^2/\kappa_p$.

We note that the methods of Refs. [43–45] can lead to a Hamiltonian formally similar to H_{avg} . However, contrary to our method, the two-atom decay *cannot* be realized in those methods assuming the strong cavity-photon loss. This is because those methods require a virtual cavity photon to mediate a third-order process, which indicates that the cavity-photon loss cannot be allowed to be strong; moreover, they also depend on a longitudinal coupling, which cannot be collectively enhanced in atomic ensembles.

To gain more insights into the engineered two-atom decay, we use the quantum Monte Carlo method [46]. In Figs. 1(b, c) we plot a single quantum trajectory with the Hamiltonian H and an initial state $|00\rangle|2\rangle$ (see [35] for more cases). Here, the first ket $|m_p m_s\rangle$ ($m_p, m_s =$

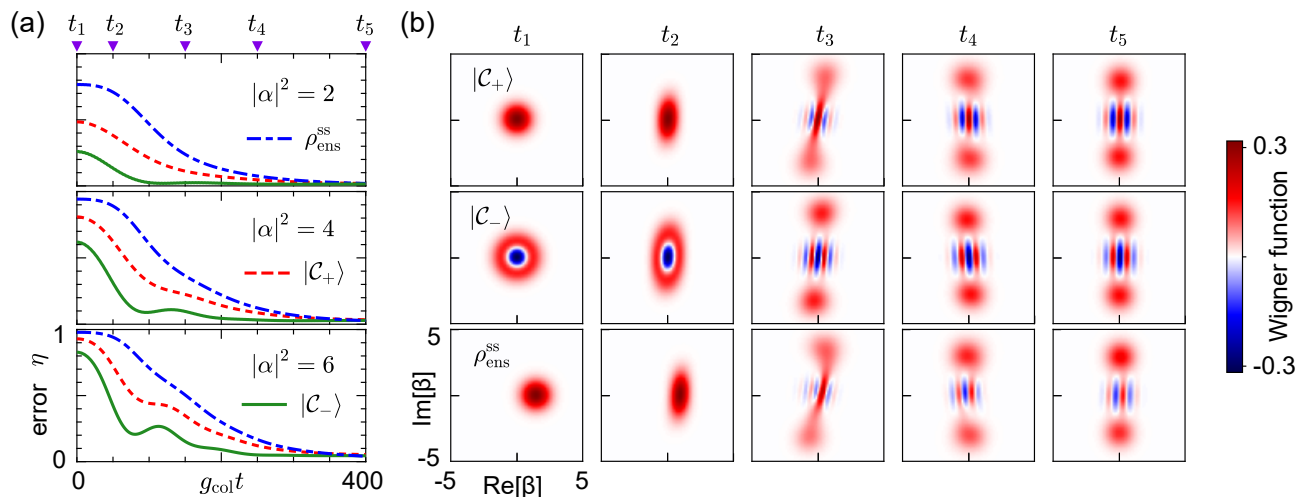


FIG. 2. (a) Time evolution of the preparation error η of the states $|C_+\rangle$, $|C_-\rangle$, and $\rho_{\text{ens}}^{\text{ss}}$ under the bosonic approximation for different cat sizes $|\alpha|^2 = 2, 4$, and 6 . The initial states are chosen as in Fig. 1(d). (b) Wigner function at times t_1, \dots, t_5 shown on top of panel (a) for the $|\alpha|^2 = 4$ cat size. The first, second, and third rows correspond to the states $|C_+\rangle$, $|C_-\rangle$, and $\rho_{\text{ens}}^{\text{ss}}$, respectively. For all plots, we set $J = 3g_{\text{col}}$, $\delta_p = J^2/(20g_{\text{col}})$, $\kappa_p = 5\chi$, and $\kappa_s = 0.3\kappa_p$.

$0, 1, 2, \dots$) in the pair refers to the cavity state with m_p pump photons and m_s signal photons, and the second $|n\rangle$ ($n = 0, 1, 2, \dots$) refers to the collective spin state $|S = N/2, m_z = -N/2 + n\rangle$, corresponding to n excited atoms in the ensemble. The non-Hermitian Hamiltonian $H_{\text{NH}} = H - \frac{i}{2}\kappa_p a_p^\dagger a_p$ drives Rabi oscillations between $|00\rangle|2\rangle$ and $|10\rangle|0\rangle$, as shown in Fig. 1(b). The Rabi oscillations are then interrupted by a quantum jump a_p . We find from Fig. 1(c) that the jump leaves the system in its ground state $|00\rangle|0\rangle$, implying that single-photon loss of the pump mode causes the two-atom decay.

Stabilized manifold of atomic cat states.—When $\kappa_{\text{1at}} = 0$, the dynamics of the effective master equation in Eq. (4) describes a pairwise exchange of atomic excitations between the ensemble and its environment, thus conserving the excitation-number parity. As demonstrated in [35], the ensemble is driven to an even cat state $|C_+\rangle = \mathcal{A}_+(\theta, \phi) + |\theta, \phi + \pi\rangle$ if initialized in an even parity state, or to an odd cat state $|C_-\rangle = \mathcal{A}_-(\theta, \phi) - |\theta, \phi + \pi\rangle$ if initialized in an odd parity state. Here, $|\theta, \phi\rangle$, where $\phi = \pi/2$ and $\theta = 2\arctan(|\alpha|/\sqrt{N})$, refers to a spin coherent state, and $\mathcal{A}_\pm = 1/\{2[1 \pm \exp(-2|\alpha|^2)]\}^{1/2}$. Moreover, $\alpha = i\sqrt{\Omega}/\chi$ is the coherent amplitude. The average number of excited atoms, $|\alpha|^2$, of the states $|C_\pm\rangle$ characterizes the cat size [36]. When assuming the initial state to be a spin coherent state $|\theta_0, \phi_0\rangle$, the steady state of the ensemble is confined into a quantum manifold spanned by the states $\{|C_+\rangle, |C_-\rangle\}$, and is expressed as $\rho_{\text{ens}}^{\text{ss}} = c_{++}|C_+\rangle\langle C_+| + c_{--}|C_-\rangle\langle C_-| + c_{+-}|C_+\rangle\langle C_-| + c_{-+}^*|C_-\rangle\langle C_+|$, where $c_{++} = \frac{1}{2}[1 + \exp(-2|\alpha_0|^2)]$ with $\alpha_0 = \sqrt{N}\exp(i\phi_0)\tan(\theta_0/2)$, $c_{--} = 1 - c_{++}$, and c_{+-} is given in [35]. To confirm these predictions, we numerically integrate [47, 48] the master equation in Eq. (2) to simulate the time evolution of the

preparation error $\eta = 1 - F$ in Fig. 1(d). Here, F is the fidelity between the actual and ideal states. It is seen that, as expected, the ensemble states are steered into a stabilized 2D cat-state manifold with a high fidelity.

In the low-excitation regime considered above, the collective spin in fact behaves as a quantum harmonic oscillator. This allows us to map S_- to a bosonic operator b , i.e., $S_- \approx \sqrt{N}b$, and thus to investigate cat states of large size ($|\alpha| \geq 2$) in large ensembles. The spin coherent state $|\theta, \phi\rangle$ accordingly becomes a bosonic coherent state $|\alpha\rangle$, such that the states $|C_\pm\rangle$ become $|C_\pm\rangle = \mathcal{A}_\pm(|\alpha\rangle + |-\alpha\rangle)$. With the master equation in Eq. (2) and under the bosonic approximation, we plot the time evolution of the preparation error η in Fig. 2(a), and the Wigner function $W(\beta)$ for different times in Fig. 2(b). We find that a cat state of size $|\alpha|^2 = 4$ is obtained after time $t \sim 250/g_{\text{col}}$, or more specifically, $t \sim 4 \mu\text{s}$, for a typical collective coupling strength $g_{\text{col}}/2\pi = 10 \text{ MHz}$ [37, 49–52].

Suppressed spin dephasing.—So far, we have assumed a model where there is no spin dephasing; however, there will always be some spin dephasing. Before discussing spin dephasing, let us first consider the rate γ of convergence, i.e., how rapidly the steady cat states can be reached. To determine γ , we introduce the Liouvillian spectral gap, $\lambda = |\text{Re}[\lambda_1]|$, of the effective master equation in Eq. (4) for $\kappa_{\text{1at}} = 0$. Here, λ_1 is the Liouvillian eigenvalue with the smallest modulus of the real part. Since the gap λ determines the slowest relaxation of the Liouvillian [53], we thus conclude that $\gamma > \lambda$. In the inset of Fig. 3(a), we numerically calculate the gap λ , and find $\lambda \approx |\alpha|^2 \kappa_{\text{2at}}$ for $|\alpha|^2 \geq 2$.

Below we consider collective spin dephasing $\gamma_{\text{col}}\mathcal{L}(S_z)\rho_{\text{ens}}$, local spin dephasing $\gamma_{\text{loc}}\sum_{j=1}^N\mathcal{L}(\sigma_j^z)\rho_{\text{ens}}$,

and inhomogeneous broadening $\frac{1}{2} \sum_{j=1}^N \delta_j \sigma_j^z$. Here, γ_{col} and γ_{loc} are the collective and local dephasing rates, respectively. Moreover, $\delta_j = \omega_j - \omega_q$, where ω_j is the transition frequency of the j th atom and ω_q can be considered as the average of transition frequencies of all the atoms [35]. We assume that the distribution of δ_j has a linewidth Δ_{inh} . The three sources of dephasing noise conserve the excitation-number parity of the superradiant subspace, where the cat states are created and stabilized. Thus, all these dephasing processes can be strongly suppressed by the two-atom decay as long as $\gamma \gg \{\gamma_{\text{col}}, \gamma_{\text{loc}}, \Delta_{\text{inh}}\}$ (i.e., $|\alpha|^2 \kappa_{2\text{at}} \gg \{\gamma_{\text{col}}, \gamma_{\text{loc}}, \Delta_{\text{inh}}\}$) (see [35] for more details). Figure 3(a) shows the dependence of such a dissipative suppression on the ratio $\kappa_{2\text{at}}/\gamma_{\text{deph}}$, assuming $\gamma_{\text{col}} = \gamma_{\text{loc}} = \Delta_{\text{inh}} \equiv \gamma_{\text{deph}}$. It is seen that for $\kappa_{2\text{at}} = 10\gamma_{\text{deph}}$, corresponding to an ensemble coherence time of $\gamma_{\text{deph}}^{-1} \sim 27 \mu\text{s}$, a steady cat state is generated, implying a significant suppression of spin dephasing. We note that in Fig. 3(a) the error η is limited by a small N , especially for $\kappa_{2\text{at}} = 10\gamma_{\text{deph}}$; and a larger N could lead to a smaller η until the bosonic approximation is well satisfied.

Cat-state lifetime.— Let us now consider the cat state lifetime τ_{at} . According to the above discussions, the effects of spin dephasing on τ_{at} can be excluded. This lifetime is thus determined by the Purcell decay rate $\Gamma_{1\text{at}} = 2|\alpha|^2 \kappa_{1\text{at}}$, such that

$$\tau_{\text{at}} = \Gamma_{1\text{at}}^{-1} = \left(\frac{\Delta}{g_{\text{col}}} \right)^2 \frac{1}{2|\alpha|^2 \kappa_s}. \quad (5)$$

Note that intracavity photonic cat states, i.e., equal superpositions of two opposite-phase coherent states, rapidly decohere into statistical mixtures due to single-photon loss. The lifetime of such photonic cat states is thus given by $\tau_{\text{ph}} = 1/2|\alpha|^2 \kappa_s$ [54]. Here, for a fair comparison, we have assumed the same cat size $|\alpha|^2$ as our atomic cat states, and the same single-photon loss rate κ_s as given for the signal cavity. It is seen that τ_{at} is longer by a factor of $(\Delta/g_{\text{col}})^2$, compared to τ_{ph} . To make $\tau_{\text{at}}/\tau_{\text{ph}}$ larger, it is essential to increase Δ/g_{col} . However, the rate $\kappa_{2\text{at}}$, which needs to be $\gg \gamma_{\text{deph}}$ as mentioned already, decreases as Δ/g_{col} increases. Thus, the ratio Δ/g_{col} has an upper bound for a given γ_{deph} . Experimentally, the coherence time, $\gamma_{\text{deph}}^{-1}$, of NV-spin ensembles has reached ~ 1 ms with spin-echo pulse sequences [55, 56], and if dynamical-decoupling techniques are employed, it can be even close to 1 sec [57]. In Fig. 3(b), the ratio $\kappa_{2\text{at}}/\gamma_{\text{deph}}$ for different γ_{deph} , as well as the ratio $\tau_{\text{at}}/\tau_{\text{ph}}$, is plotted versus Δ/g_{col} . Assuming a realistic parameter of $\gamma_{\text{deph}}^{-1} = 1$ ms, we find from Fig. 3(b) that in stark contrast to previous work on intracavity photonic cat states (see Table I in [35]), our approach can lead to *an increase in the cat state lifetime of up to four orders of magnitude* for $\kappa_{2\text{at}} \approx 15\gamma_{\text{deph}}$

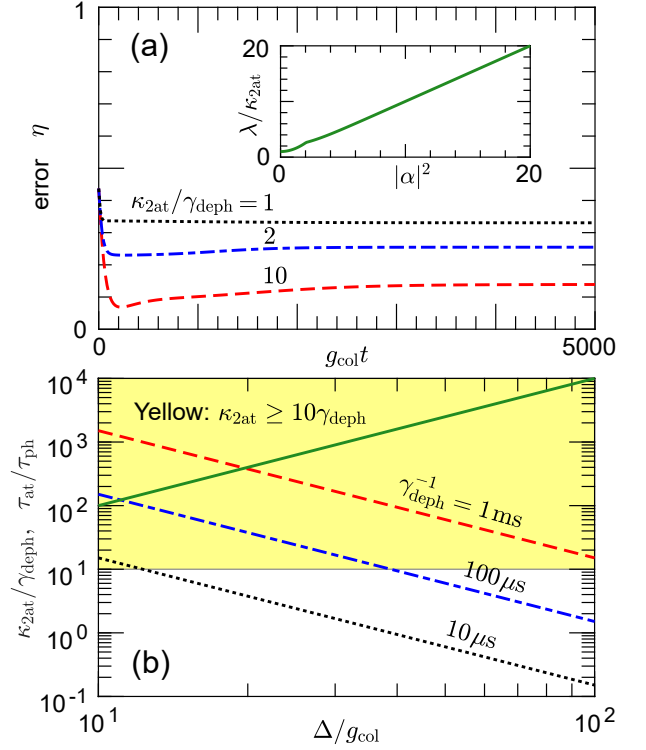


FIG. 3. (a) Effects of collective dephasing, local dephasing, and inhomogeneous broadening on the preparation error η of the state $|\mathcal{C}_+\rangle$ of size $|\alpha|^2 = 2$. We integrated the effective master equation (4), with an additional spin dephasing $\gamma_{\text{col}} \mathcal{L}(S_z) \rho_{\text{ens}}$, local spin dephasing $\gamma_{\text{loc}} \sum_{j=1}^N \mathcal{L}(\sigma_j^z) \rho_{\text{ens}}$, and inhomogeneous broadening $\frac{1}{2} \sum_{j=1}^N \delta_j \sigma_j^z$. The frequency shifts δ_j are randomly given according to a Lorentzian distribution of linewidth Δ_{inh} . For simplicity, we here set $N = 10$, $\gamma_{\text{col}} = \gamma_{\text{loc}} = \Delta_{\text{inh}} \equiv \gamma_{\text{deph}}$, and $\kappa_{1\text{at}} = 0$, so that only the effects of these dephasing processes are shown. Inset: the Liouvillian spectral gap, λ , of the master equation (4) versus the cat size $|\alpha|^2$ for $\kappa_{1\text{at}} = 0$, under the bosonic approximation. (b) Ratio $\kappa_{2\text{at}}/\gamma_{\text{deph}}$ versus the parameter Δ/g_{col} for $\gamma_{\text{deph}}^{-1} = 10 \mu\text{s}$, $100 \mu\text{s}$, and 1 ms for $\kappa_p = 5\chi$ and $J/2\pi = 30$ MHz. The yellow shaded area represents the $\kappa_{2\text{at}} \geq 10\gamma_{\text{deph}}$ regime, where spin dephasing is strongly suppressed by the two-atom decay. The solid green line shows $\tau_{\text{at}}/\tau_{\text{ph}}$ versus Δ/g_{col} . Other parameters in (a) and (b) are chosen as in Fig. 2.

and a very large cat size of $|\alpha|^2 \geq 4$. Correspondingly, for a typical single-photon loss rate of $\kappa_s/2\pi = 10$ kHz (i.e., a cavity decay time $\sim 16 \mu\text{s}$) [30], the lifetime of the $|\alpha|^2 = 4$ cat states resulting from our approach is ~ 20 ms.

As the cavity loss rate κ_s decreases, the lifetime τ_{at} further increases and ultimately reaches its maximum value, limited by spin relaxation and thermal noise (see [35] for more details). This maximum lifetime is given by $\tau_{\text{at}}^{\text{max}} = \Gamma_{\text{relax}}^{-1}$. Here, $\Gamma_{\text{relax}} = [2|\alpha|^2(1 + 2n_{\text{th}}) + 2n_{\text{th}}]\gamma_{\text{relax}}$ [58] is the cat state decay rate arising from spin relaxation with a rate γ_{relax} and thermal

noise with a thermal average boson number n_{th} . For realistic parameters of $\gamma_{\text{relax}} = 2\pi \times 4$ mHz [37, 38] and $T = 100$ mK, we can predict a maximum lifetime of $\tau_{\text{at}}^{\text{max}} \sim 3$ sec, which is *more than two orders of magnitude longer than the longest lifetime*, 17 ms, of the intracavity photonic cat states reported in Ref. [36].

Conclusions.—We have introduced a method to create and stabilize large-size, long-lived Schrödinger cat states in atomic ensembles. This method is based on the use of fully quantized degenerate parametric amplification to facilitate the simultaneous decay of two atoms, i.e., the two-atom decay. The resulting atomic cat states can last *an extremely long time, because of strongly suppressed spin dephasing, and of extremely weak spin relaxation and thermal noise.* These long-lived cat states are promising for both fundamental tests and practical applications of quantum mechanics. Our work can further stimulate more efforts to create and protect macroscopic cat states or other fragile quantum states, and also to utilize them to improve the performance of various modern quantum technologies.

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