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Generating Large Cats with Nine Lives: Long-Lived Macroscopic Schrödinger Cat 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 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 engineering large-size and long-lived cat states, with immediate interests both in fundamental studies and noise-immune quantum information processing.

Introduction.—Schrödinger cat states, i.e., quantum superpositions of two macroscopically distinct states, are appealing not only for fundamental studies of quantum mechanics [1, 2], but also for wide applications, ranging from quantum information processing [3] to quantum metrology [4, 5]. The ability to reliably create and then stabilize cat states is therefore highly desirable. So far, a large number of approaches that have been proposed for the generation of cat states rely on unitary gate operations [6–20]. However, it remains a challenge for these unitary approaches to obtain stabilized cat states (and, especially, large-size ones) in a noisy environment. To overcome this obstacle, quantum reservoir engineering [21, 22] could provide a counterintuitive route. Two-photon loss has already been engineered to stabilize photonic cat states [23–26]. Such a nonlinear loss can protect these cat states against photon dephasing [27, 28], but unfortunately not against single-photon loss, which is unavoidable and can randomly change the cat state parity. Thus, the cat state lifetime is still significantly limited. For example, single-photon loss has been considered to be the major source of noise in fault-tolerant quantum computation based on cat states [3, 28–33].

Ensembles of atoms or spins have negligible spin relaxation, and instead their major source of noise is spin dephasing. 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, we here propose to use fully quantum degenerate parametric amplification (DPA), and demonstrate that stabilized atomic cat states of large size (i.e., containing at least four excited atoms on average) can be generated in an ensemble of atoms off-resonantly coupled to the signal mode of DPA. More importantly, the lifetime of our 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 [34]), 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 ($\sim 16\ \mu$s), our cat state lifetime can reach $\sim$ 20 ms. This is comparable to 17 ms [35], which is the longest lifetime of intracavity photonic cat states to date, but which was achieved with an extreme cavity decay time ($\sim$ 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 $\sim$ 40 sec [36, 37], we can predict a maximum cat state lifetime of $\sim$ 3 sec.

Physical model.—The central idea is illustrated in Fig. 1(a). To consider DPA in the fully quantum regime, our system, inspired by recent experimental advances [3, 38–41], 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 N identical two-level atoms is placed in the signal cavity, and the atomic transition, of frequency ω_q, is driven by a

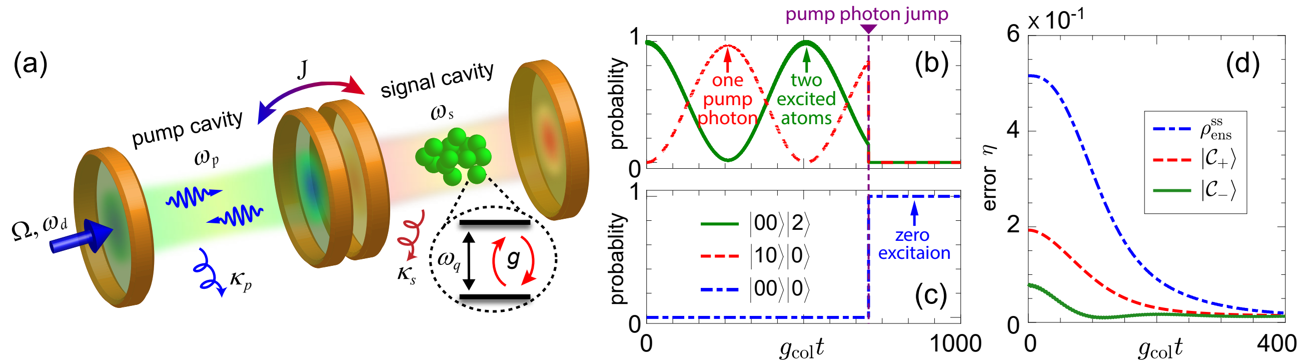

FIG. 1. (a) Schematic setup of our proposal. The pump and signal cavities are coupled via a parametric coupling J, and the atomic ensemble is coupled to the signal cavity with a single-atom coupling g. The pump cavity is subject to a coherent drive with amplitude Ω and frequency ω_d. Here, ω_p, ω_s are 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, we assumed that $\kappa_p = 0.2\chi$ and $\kappa_s = \Omega = 0$. (d) Time evolution of the preparation error η for a cat size $|\alpha|^2 = 1$. Here, we assumed that $\kappa_p = 5\chi$, $\kappa_s = 0.3\kappa_p$, and that 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 $\rho_{\rm ens}^{\rm ss}$, respectively. In (b)-(d), we assumed that $N = 100$, $J = 3g_{\rm col}$, and both cavities are initialized in the vacuum.

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 cat states for an extremely long time in the ensemble.

The system Hamiltonian in a frame rotating at ω_d is

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

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 jth 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 $\Delta = \omega_s - \omega_q \gg \{g_{\rm col}, J\}$. Here, $g_{\rm col} = \sqrt{N}g$ represents the collective coupling of the ensemble to the signal mode. Then, we can predict a parametric coupling, $\chi = g_{\rm col}^2 J/\Delta^2$, between atom pairs and pump single photons. Accordingly, the Hamiltonian H, after time averaging [42, 43], becomes

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

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 [34]). To derive $H_{\rm avg}$, we have considered the low-excitation regime, where the average number of excited atoms is much smaller than the total number of atoms. We note that other approaches [44, 45] can also obtain a Hamiltonian similar in form to $H_{\rm avg}$. Such approaches, however, rely on a longitudinal coupling which cannot be collectively enhanced, and in those cases [44, 45], the coupling χ becomes extremely weak in typical ensembles.

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

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

where $H_{\rm ens} = i\chi_{\rm 2at}\left(S_-^2 - S_+^2\right)/N$, and $\rho_{\rm ens}$ represents the reduced density matrix of the ensemble. Here, $\kappa_{\rm 2at} = 4\chi^2/\kappa_p$ and $\chi_{\rm 2at} = 2\Omega\chi/\kappa_p$ are the rates of the simultaneous decay and excitation of two atoms, respectively. Moreover, $\kappa_{\rm 1at} = (g_{\rm col}/\Delta)^2\kappa_s$ is the rate of the single-atom decay induced by single-photon loss of the signal cavity (see [34]), and we can tune it to be $\ll \kappa_{\rm 2at}$, as long as $\kappa_s \ll (g_{\rm col}J/\Delta)^2/\kappa_p$.

To gain more insights into the engineered two-atom decay, we turn to 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 [34] for more cases). Here, the first ket $|m_p m_s\rangle$ ($m_p, m_s = 0, 1, 2, \ldots$) in the pair refers to the cavity state with m_p pump photons and m_s signal photons, and the second $|n\rangle$ ($n = 0, 1, 2, \ldots$) refers to the collective spin state $|S = N/2, m_z = -N/2 + n\rangle$, corresponding to n excited

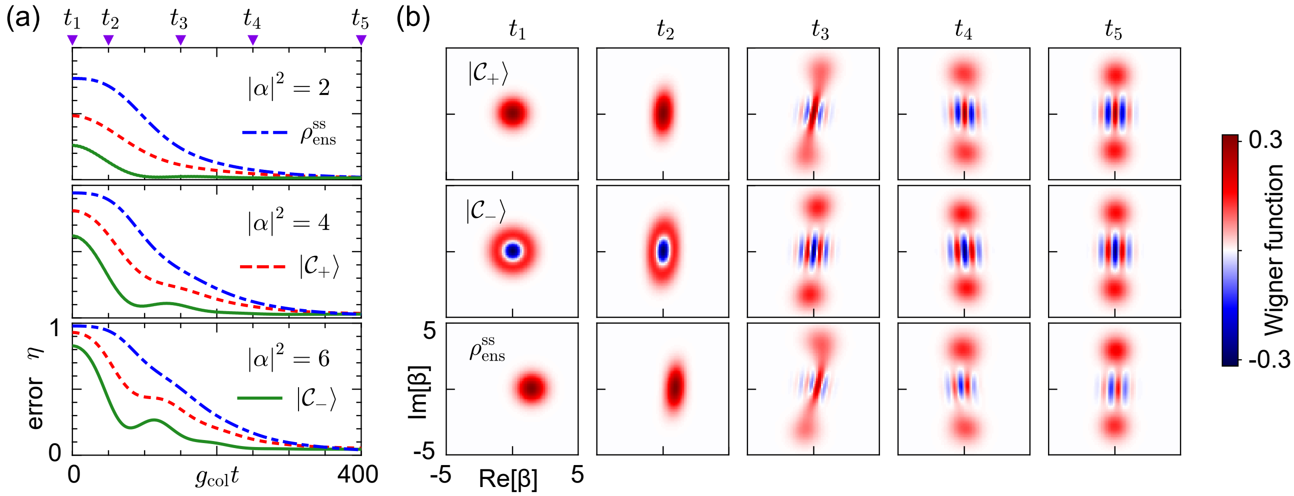

FIG. 2. (a) Time evolution of the preparation error η of the states $|\mathcal{C}_+\rangle$, $|\mathcal{C}_-\rangle$, and ρ^{ss}_{ens} under the bosonic approximation for different cat sizes $|\alpha|^2 = 2$, 4, and 6. The initial states are chosen as in Fig. 1(d). (b) Wigner function at times $t_1,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

atoms in the ensemble. The non-Hermitian Hamiltonian $H_{NH} = H - \frac{i}{2}\kappa_p a_p^\dagger a_p$ 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 $\kappa_{1at} = 0$, the dynamics of the effective master equation in Eq. (4) describes a pairwise exchange of atomic excitations between the ensemble and its environment. Because this exchange conserves the excitation number parity, the ensemble state space can be decomposed into even and odd subspaces, according to the eigenvalues ± 1 of the parity operator $P = \exp(i\pi\sum_{j=1}^{N}|e\rangle_j\langle e|)$. Here, $|e\rangle$ is the excited state of the atoms. As demonstrated in [34], the ensemble is driven to an even cat state $|\mathcal{C}_+\rangle = \mathcal{A}_+(|\theta,\phi\rangle + |\theta,\phi+\pi\rangle)$ if initialized in an even parity state, or to an odd cat state $|\mathcal{C}_-\rangle = \mathcal{A}_-(|\theta,\phi\rangle - |\theta,\phi+\pi\rangle)$ 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 $|\mathcal{C}_\pm\rangle$ characterizes the cat size [35]. 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 $\{|\mathcal{C}_+\rangle, |\mathcal{C}_-\rangle\}$, and is expressed as $\rho^{ss}_{ens} = c_{++}|\mathcal{C}_+\rangle\langle\mathcal{C}_+| + c_{--}|\mathcal{C}_-\rangle\langle\mathcal{C}_-| + c_{+-}|\mathcal{C}_+\rangle\langle\mathcal{C}_-| + c^*_{+-}|\mathcal{C}_-\rangle\langle\mathcal{C}_+|$, 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 [34]. 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.

Bosonic approximation and cat-state lifetime.— 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 ($|\alpha| \geqslant 2$) in large ensembles. The spin coherent state $|\theta,\phi\rangle$ accordingly becomes a bosonic coherent state $|\alpha\rangle$, such that the states $|\mathcal{C}_\pm\rangle$ become $|\mathcal{C}_\pm\rangle = \mathcal{A}_\pm(|\alpha\rangle + |-\alpha\rangle)$. 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 $t \sim 250/g_{col}$, or more specifically, $t \sim 4\ \mu$s, for a typical collective coupling strength $g_{col}/2\pi = 10$ MHz [36, 49–52].

So far, we have assumed a model where there is no spin dephasing. However, there will always be some spin dephasing, described by a Lindblad dissipator $\gamma_d\mathcal{L}(b^\dagger b)\rho$, with a rate $\gamma_d > 0$. 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 the rate γ, we introduce the Liouvillian spectral gap, $\lambda = |\mathrm{Re}[\lambda_1]|$, of the effective master equation in Eq. (4) for $\kappa_{1at} = 0$. 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 $\lambda \approx |\alpha|^2\kappa_{2at}$ for $|\alpha|^2 \geq 4$.

Below we show that spin dephasing, which conserves the parity operator P, can be strongly suppressed by the two-atom decay, as long as the rate of convergence is much larger than the spin dephasing rate (i.e., $\gamma \gg \gamma_{\rm deph}$, or $|\alpha|^2\kappa_{2\rm at} \gg \gamma_{\rm deph}$ due to $\gamma > \lambda$). This is like in the cases of photonic cat states stabilized by two-photon loss [27, 28]. The dissipative suppression can be understood from the quantum jump approach. The jump operator $b^\dagger b$, when acting, e.g., on the state $|\mathcal{C}_+\rangle$, excites a state $|\psi\rangle = \mathcal{A}_+ [D(\alpha) - D(-\alpha)]\,|1\rangle$, according to $b^\dagger b|\mathcal{C}_+\rangle = |\alpha|^2\,|\mathcal{C}_+\rangle + \alpha|\psi\rangle$. Here, $D(\pm\alpha) = \exp[\pm\alpha(b^\dagger + b)]$ are displacement operators. However, the state $|\psi\rangle$ still has even parity, and thus can be autonomously driven back to the state $|\mathcal{C}_+\rangle$ by the two-atom decay. Figure 3(a) shows the dependence of such a dissipative suppression on the ratio $\kappa_{2\rm at}/\gamma_{\rm deph}$. We find that for $\kappa_{2\rm at} = 10\gamma_{\rm deph}$, corresponding to an ensemble coherence time of $\gamma_{\rm deph}^{-1} \sim 27$ μs, the steady state $|\mathcal{C}_+\rangle$ is achieved with an error $\eta \sim 0.06$, implying a significant suppression of spin dephasing. There is a similar suppression mechanism for the action of $b^\dagger b$ on the state $|\mathcal{C}_-\rangle$. Furthermore, if the ensemble is initialized in a spin coherent state, the decay rate of the coherence between the states $|\mathcal{C}_\pm\rangle$ scales as $\sim \gamma_{\rm deph}\exp(-2\,|\alpha|^2)$, and is suppressed exponentially with the cat size $|\alpha|^2$. Hence, the 2D cat-state manifold stabilized by the two-atom decay is robust against spin dephasing.

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

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

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 $\tau_{\rm ph} = 1/2\,|\alpha|^2\,\kappa_s$ [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 $\tau_{\rm at}$ is longer by a factor of $(\Delta/g_{\rm col})^2$, compared to $\tau_{\rm ph}$. To make $\tau_{\rm at}/\tau_{\rm ph}$ larger, it is essential to increase $\Delta/g_{\rm col}$. However, the rate $\kappa_{2\rm at}$, which needs to be $\gg \gamma_{\rm deph}$ as mentioned already, decreases as $\Delta/g_{\rm col}$ increases. Thus, the ratio $\Delta/g_{\rm col}$ has a lower bound for a given $\gamma_{\rm deph}$. Experimentally, the coherence time, $\gamma_{\rm deph}^{-1}$, of NV-spin ensembles has reached $\sim$ 1 ms with spin-echo pulse sequences [55], and if dynamical-decoupling techniques are employed, it can be even close to 1 sec [56]. In Fig. 3(b), the ratio $\kappa_{2\rm at}/\gamma_{\rm deph}$ for different $\gamma_{\rm deph}$, as well as the ratio $\tau_{\rm at}/\tau_{\rm ph}$, is plotted versus $\Delta/g_{\rm col}$. Assuming a realistic parameter of $\gamma_{\rm deph}^{-1} = 1$ ms, we find from Fig. 3(b) that in stark contrast to previous work on

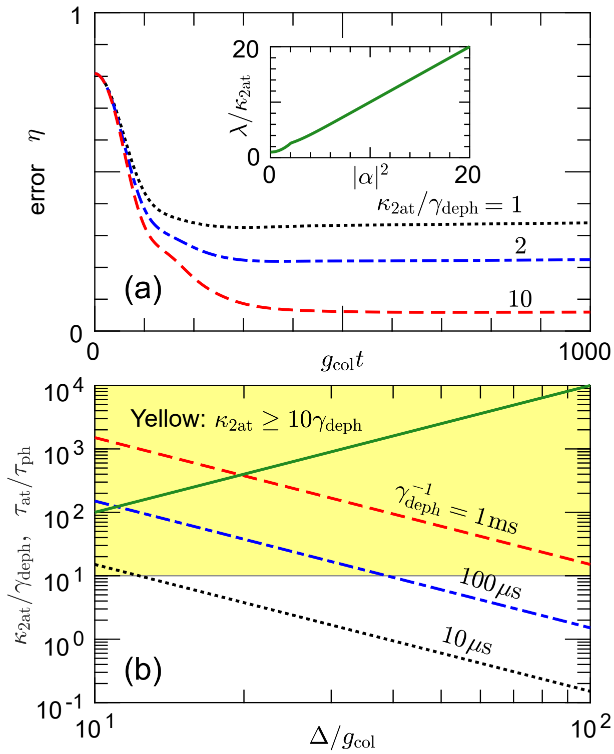

FIG. 3. (a) Effects of spin dephasing on the preparation error η of the state $|\mathcal{C}_+\rangle$ of size $|\alpha|^2 = 4$. We integrated the master equation in Eq. (2), with an additional spin dephasing $\gamma_{\rm deph}\mathcal{L}(b^\dagger b)\rho$. We set $\kappa_s = 0$, so that only the effects of spin dephasing are shown. Inset: the Liouvillian spectral gap, λ, of the master equation in Eq. (4) versus the cat size $|\alpha|^2$ for $\kappa_{1\rm at} = 0$. Here, the bosonic approximation is made for all plots. (b) Ratio $\kappa_{2\rm at}/\gamma_{\rm deph}$ versus the parameter $\Delta/g_{\rm col}$ for $\gamma_{\rm deph}^{-1} = 10$ μs, 100 μs, and 1 ms for $\kappa_p = 5\chi$ and $J/2\pi = 30$ MHz. The yellow shaded area represents the $\kappa_{2\rm at} \geq 10\gamma_{\rm deph}$ regime, where spin dephasing is strongly suppressed by the two-atom decay. The solid green line shows $\tau_{\rm at}/\tau_{\rm ph}$ versus $\Delta/g_{\rm col}$. Other parameters in (a) and (b) are set to be the same as in Fig. 2.

intracavity photonic cat states (see Table I in [34]), our approach can lead to an increase in the cat state lifetime of up to four orders of magnitude for $\kappa_{2\rm at} \approx 15\gamma_{\rm deph}$ and a large cat size of $|\alpha|^2 \geqslant 4$. Correspondingly, for a typical single-photon loss rate of $\kappa_s/2\pi = 10$ kHz (i.e., a cavity decay time ~ 16 μs) [38], 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 $\tau_{\rm at}$ further increases and ultimately reaches its maximum value limited by spin relaxation and thermal noise (see [34] for more details). This maximum lifetime is given by $\tau_{\rm at}^{\rm max} = \Gamma_{\rm relax}^{-1}$. Here, $\Gamma_{\rm relax} = [2|\alpha|^2(1 + 2n_{\rm th}) + 2n_{\rm th}]\gamma_{\rm relax}$ [57] is the cat state decay rate arising from spin relaxation with a rate $\gamma_{\rm relax}$ and thermal noise with a thermal average boson number $n_{\rm th}$. For realistic parameters of $\gamma_{\rm relax} = 2\pi \times 4$ mHz [36, 37] and $T = 100$ mK, we can predict a maximum lifetime of

$\tau_{\rm at}^{\rm max} \sim 3$ sec, which is more than two orders of magnitude longer than the longest lifetime, i.e., 17 ms, of intracavity photonic cat states reported in Ref. [35].

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 DPA to engineer 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.* We expect that these atomic cat states can find wide applications in fundamental investigations of quantum measurement and decoherence, and various quantum technologies.

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SUPPLEMENTAL MATERIAL

Here, we first compare the lifetimes of our atomic cat states and common intracavity photonic cat states. In Sec. S2, we present how to eliminate the second-order effect and as a result to make the desired third-order effect dominant. In Sec. S3, we derive the single-atom decay induced by single-photon loss of the signal cavity. In Sec. S4, we show quantum Monte-Carlo trajectories of the ensemble-cavity system. Then Sec. S5 gives the detailed derivation of atomic cat states stabilized by the two-atom decay. Finally, in Sec. S6, we discuss the effects of spin relaxation and thermal noise on the cat state lifetime, and also show the maximum cat state lifetime limited by them.

S1. Comparison of the lifetimes of our atomic cat states and intracavity photonic cat states

The cat state lifetime can be defined as the inverse cat state decoherence rate. Sec. S6 shows how to derive the cat state decoherence rate and then obtain the cat state lifetime. In this section, let us first compare the lifetime of intracavity atomic cat states resulting from our approach with that of common intracavity photonic cat states, under some realistic parameters. Our atomic cat states refer to superpositions of two spin coherent states, i.e.,

$$|\mathcal{C}_{\pm}\rangle = \mathcal{A}_{\pm}\left(|\theta,\phi\rangle + |\theta,\phi+\pi\rangle\right), \tag{S1}$$

Here, $\mathcal{A}_{\pm} = 1/\{2[1\pm\exp(-2\,|\alpha|^2)]\}^{1/2}$, and the state $|\theta,\phi\rangle$, where $\phi=\pi/2$ and $\theta = 2\arctan(|\alpha|/\sqrt{N})$, is the spin coherent state that is obtained by rotating the ground state of the ensemble by an angle θ about the axis $(\sin\phi, -\cos\phi, 0)$ of the collective Bloch sphere. For a large ensemble, we can apply the bosonic approximation, which maps the collective spin of the ensemble to a quantum harmonic oscillator. Under this approximation, the spin coherent states $|\theta,\phi\rangle$ and $|\theta,\phi+\pi\rangle$ become bosonic coherent states $|\alpha\rangle$ and $|-\alpha\rangle$, respectively, with coherent amplitudes $\pm\alpha$. The atomic cat states in Eq. (S1) likewise become

$$|\mathcal{C}_{\pm}\rangle = \mathcal{A}_{\pm}\left(|\alpha\rangle \pm |-\alpha\rangle\right). \tag{S2}$$

Furthermore, the intracavity photonic cat states refer to

$$|\mathcal{C}_{\pm}\rangle_{\rm ph} = \mathcal{A}_{\pm}\left(|\alpha\rangle_{\rm ph} \pm |-\alpha\rangle_{\rm ph}\right), \tag{S3}$$

where $|\pm\alpha\rangle_{\rm ph}$ are the photonic coherent states with coherent amplitudes $\pm\alpha$. It is seen, from Eqs. (S2) and (S3), that $|\alpha|^2$ is the average number of excited atoms or photons and, thus, can characterize the cat size.

In Table I, we list some parameters of intracavity photonic cat states $|\mathcal{C}_{\pm}\rangle_{\rm ph}$ implemented in experiments. For comparison, we also show the corresponding results of our atomic cat states $|\mathcal{C}_{\pm}\rangle$ at the end of the table. With

TABLE I. Some relevant parameters of experimentally implemented intracavity photonic cat states $|\mathcal{C}_{\pm}\rangle_{\rm ph}$. Here, $|\alpha|^2$ characterizes the cat size, T_c is the cavity photon lifetime, $\kappa_s = 1/T_c$ is the cavity photon loss rate, $\tau_{\rm exp}$ is the cat state lifetime measured in experiments, and $\tau_{\rm theor} = 1/(2\,|\alpha|^2\,\kappa_s)$ is the theoretical prediction of the cat state lifetime. For comparison, we also list at the end of the table the corresponding theoretical predictions for our atomic cat states $|\mathcal{C}_{\pm}\rangle$.

Ref.	approach type	$\|\alpha\|^2$	T_c (μs)	$\kappa_s/2\pi$ (kHz)	$\tau_{\rm exp}$ (μs)	$\tau_{\rm theor}$ (μs)
[S1]	unitary evolution	3.0	1.3×10^5	1.2×10^{-3}	1.7×10^4	2.2×10^4
[S2]	reservoir engineering	5.8	3.0	53.0	0.2	0.26
[S3]	unitary evolution	28	22.1	7.2	—	0.4
[S4]	reservoir engineering	2.4	20	8.0	—	4.1
[S5]	reservoir engineering	5	92	1.7	8	9.2
[S6]	unitary evolution	3.3	160	1.0	38.4	35
[S7]	unitary evolution	1.4	0.14	1.1×10^3	—	5.3×10^{-2}
[S8]	unitary evolution	11.3	8.1×10^3	2.0×10^{-2}	200	360
[S9]	unitary evolution	2	692	0.2	—	173
our results	reservoir engineering	4	16	10	—	2×10^4
			5.3×10^3	3.0×10^{-2}	—	2×10^6

modest parameters the lifetime of our atomic cat states is predicted to be longer, by up to *four orders of magnitude*, compared to those photonic cat states under the same parameter conditions. For a modest single-photon loss rate of $\kappa_s/2\pi = 10$ kHz (i.e., a cavity decay time of $T_c \sim 16$ μs), the lifetime of our atomic cat states can reach ~ 20 ms for a cat size of $|\alpha|^2 = 4$. This lifetime is comparable in length to that (~ 17 ms) reported in Ref. [S1] in Table I, which, to our best knowledge, is the longest lifetime of intracavity photonic cat states to date. We stress that in such a comparison our cat state lifetime is achieved with a modest cavity decay time of $T_c \sim 16$ μs. This is in stark contrast to the cat state lifetime reported in Ref. [S1], which was achieved with an extreme cavity decay time of $T_c = 0.13$ sec. This means that our approach can stabilize (*for an extremely long time*) large-size cat states, even with common setups.

When decreasing the single-photon loss rate κ_s, i.e., increasing the cavity decay time T_c, our atomic cat state lifetime can further increase. For example, a single-photon loss rate $\kappa_s/2\pi = 3.0 \times 10^{-2}$ kHz, corresponding to a cavity decay time $T_c \sim 5.3$ ms, results in a cat state lifetime of ~ 2 sec, *more than two orders of magnitude longer than the lifetime*, i.e., 17 ms, reported in Ref. [S1] in Table I. Ultimately, the maximum value of our cat state lifetime is determined by extremely weak spin relaxation and thermal noise, reaching ~ 3 sec.

The essential reason for such an improvement in the cat state lifetime is because, as shown in Fig. S1, single excitation loss of ensembles (i.e., spin relaxation) is extremely weak compared to that of cavities (i.e., single-photon loss). At the same time, spin dephasing, though stronger than photon dephasing, is greatly suppressed by the engineered two-atom decay. This is in close analogy to the mechanism of using two-photon loss to suppress photon dephasing.

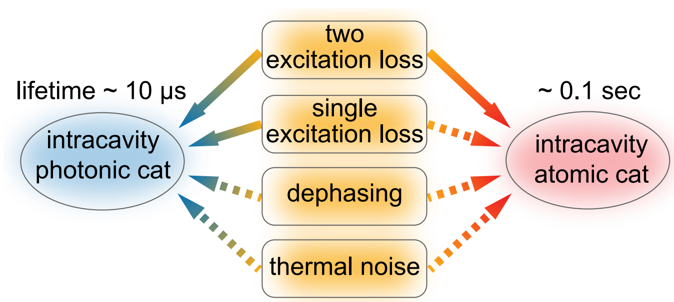

FIG. S1. Comparison of the effects of noise on intracavity photonic cat stats and our atomic cat states. Solid arrows represent the strong effects, and dashed arrows represent the extremely weak or strongly suppressible effects. While the lifetime of photonic cat states is ~ 10 μs, our atomic cat states can have a ~ 0.1 sec lifetime.

S2. Elimination of the second-order effect

The time-averaged Hamiltonian $H_{\rm avg}$ in Eq. (3) in the main article describes a third-order process, and there exists a stronger second-order process, which is described by the Hamiltonian

$$H^{(2)} = -\frac{g^2}{\Delta}\left(2a_s^\dagger a_s S_z + S_+S_-\right) - \frac{J^2}{\Delta'}\left(2a_p^\dagger a_p - a_s^\dagger a_s^\dagger a_s a_s + 4a_p^\dagger a_p a_s^\dagger a_s\right), \tag{S4}$$

where $\Delta' = 2\omega_s - \omega_p$. In order to make the third-order $H_{\rm avg}$ dominant, we need to eliminate the second-order $H^{(2)}$. Since the signal cavity is initialized in the vacuum state, the Hamiltonian $H^{(2)}$ is thus reduced to

$$H^{(2)} = -\frac{g^2}{\Delta}S_+S_- - \frac{2J^2}{\Delta'}a_p^\dagger a_p. \tag{S5}$$

We further focus our attention on the low-excitation regime, where the average number of excited atoms is much smaller than the total number of atoms. In this regime, the operator S_z can be expressed as $S_z = -N/2 + \delta S_z$, where δS_z is a small fluctuation. As a result, we find

$$S_+S_- \approx N\delta S_z, \tag{S6}$$

according to the identity $N\left(N/2+1\right)/2 = S_z^2 - S_z + S_+S_-$, and then obtain

$$H^{(2)} = -\frac{g_{\rm col}^2}{\Delta}\delta S_z - \frac{2J^2}{\Delta'}a_p^\dagger a_p. \tag{S7}$$

It is seen that the second-order process causes a Lamb shift (i.e., the first term), and a dispersive resonance shift for the pump cavity (i.e., the second term). These additional shifts can be compensated by properly detuning the pump cavity resonance ω_p from twice the atomic resonance ω_q. Hence, the second-order process can be strongly suppressed, such that the third-order process becomes dominant.

S3. Single-atom decay induced by single-photon loss of the signal cavity

Since the signal cavity is largely detuned from both the ensemble and the pump cavity, the average number of photons inside the signal cavity is thus very low. In this case, we can only consider the vacuum state $|0\rangle$ and the single-photon state $|1\rangle$ of the signal cavity. We work within the limit where $\delta_s \approx \Delta \gg \{\delta_p, \delta_q, g_{\rm col}, J\}$, and the Hamiltonian in Eq. (1) in the main article can thus be rewritten as $H = H_e + H_g + V + V^\dagger$. Here,

$$H_e = \delta_s |1\rangle\langle 1|, \tag{S8}$$

$$H_g = \delta_p a_p^\dagger a_p + \delta_q S_z + \Omega\left(a_p + a_p^\dagger\right), \tag{S9}$$

represents the interactions inside the excited- and ground-state subspaces, and

$$V = g S_- |1\rangle\langle 0| \tag{S10}$$

describes the perturbative interaction between the excited- and ground-state subspaces. Then, according to the formalism of Ref. [S10], we can define a non-Hermitian Hamiltonian $H_{\rm NH}^e = H_e - i\kappa_s|1\rangle\langle 1|/2$, and obtain an effective Lindblad dissipator for the ensemble

$$\kappa_s \mathcal{L}\left[|0\rangle_s\langle 1| \left(H_{\rm NH}^e\right)^{-1} V\right]\rho_{\rm ens} = \frac{\kappa_{\rm 1at}}{N}\mathcal{L}\left(S_-\right)\rho_{\rm ens}, \tag{S11}$$

where

$$\kappa_{\rm 1at} = \frac{\kappa_s g_{\rm col}^2}{\delta_s^2 + \kappa_s^2/4} \approx \left(\frac{g_{\rm col}}{\Delta}\right)^2 \kappa_s. \tag{S12}$$

This means that the single-photon loss process of the signal cavity gives rise to the single-atom decay of the ensemble. Importantly, the resulting decay rate $\kappa_{\rm 1at}$ is smaller than the cavity decay rate κ_s by a factor of $\left(g_{\rm col}/\Delta\right)^2$. Thus, our atomic cat states have an extremely long lifetime.

S4. Quantum Monte-Carlo trajectory for the initial states $|00\rangle|3\rangle$ and $|00\rangle|4\rangle$

The dynamics described by the time-averaged $H_{\rm avg}$ in Eq. (3) of the main article implies that pairs of atoms can jointly convert their excitations into pump single photons, and then the subsequent single-photon loss process of the pump cavity results in the simultaneous decay of two atoms, i.e., the two-atom decay.

In Fig. S2, we plot single quantum trajectories, utilizing the quantum Monte Carlo method, for the initial states $|00\rangle|3\rangle$ and $|00\rangle|4\rangle$. Here, the first ket $|m_p m_s\rangle$ $(m_p, m_s = 0, 1, 2, \ldots)$ in the pair refers to the cavity state with m_p pump photons and m_s signal photons, and the second $|n\rangle$ $(n = 0, 1, 2, \ldots)$ refers to the collective spin state $|S = N/2, m_z = -N/2 + n\rangle$, corresponding to n excited atoms in the ensemble.

For the former case, where initially the ensemble has three excited atoms, we find from Figs. S2(a, b) that two excited atoms, as a pair, decay via a single-photon loss process of the DPA pump (corresponding to a quantum jump), and one excited atom is kept in the ensemble because alone it cannot emit a single photon. If there are initially four excited atoms as shown in Figs. S2(c, d), all excited atoms, as two pairs, can decay sequentially via two single-photon loss processes of the DPA pump (corresponding to two quantum jumps).

S5. Stabilized atomic cat states by the two-atom decay

In this section we show a detailed derivation of atomic cat states stabilized by the engineered two-atom decay. We begin with the effective master equation given in Eq. (4) of the main text

$$\dot{\rho}_{\rm ens} = i\left[\rho_{\rm ens}, H_{\rm ens}\right] + \frac{\kappa_{\rm 1at}}{N}\mathcal{L}\left(S_-\right)\rho_{\rm ens} + \frac{\kappa_{\rm 2at}}{N^2}\mathcal{L}\left(S_-^2\right)\rho_{\rm ens}, \tag{S13}$$

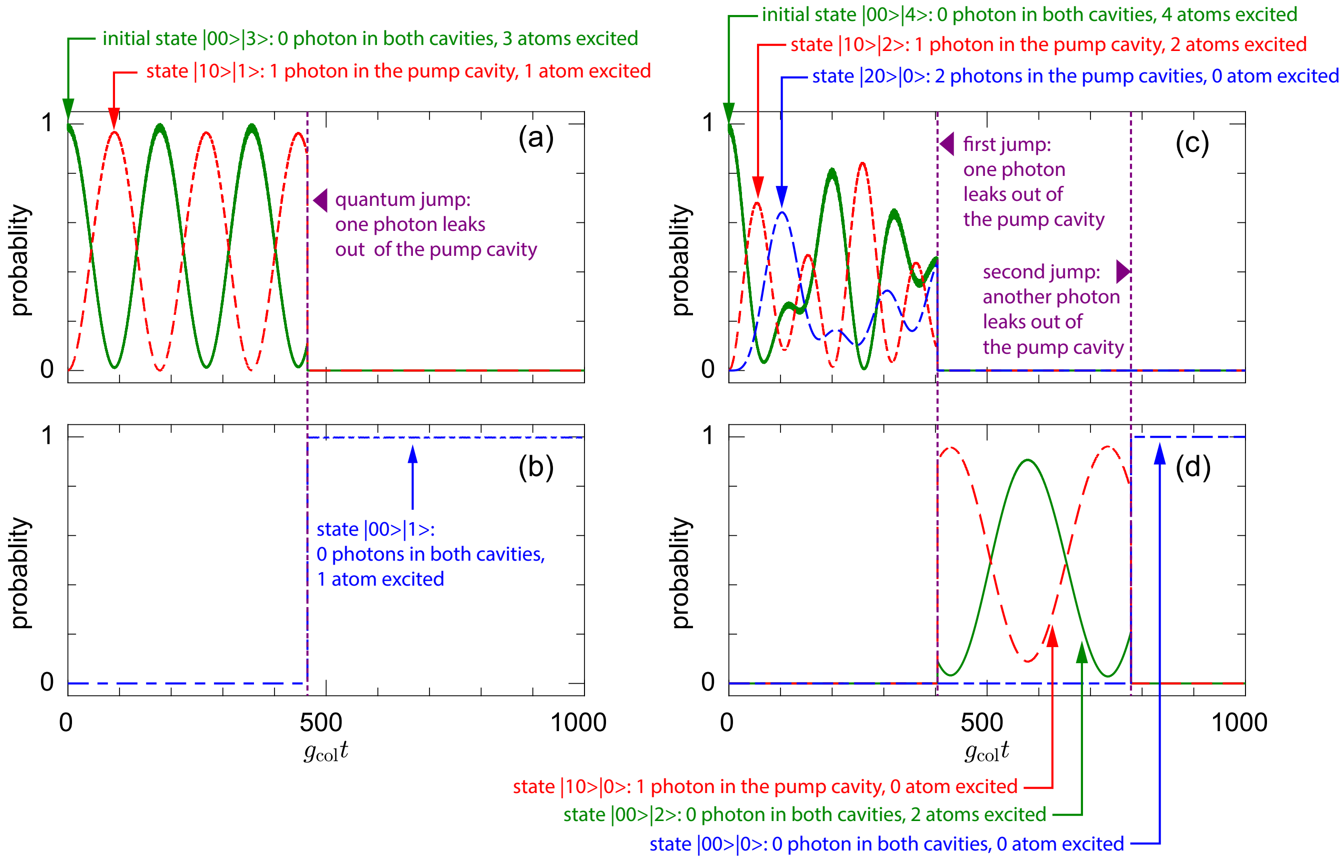

FIG. S2. Quantum Monte-Carlo trajectory pictured through the probabilities of the system being in the states $|m_p 0\rangle|n\rangle$ for the initial states (a, b) $|00\rangle|3\rangle$ and (c, d) $|00\rangle|4\rangle$. A single quantum jump a_p gives rise to the two-atom decay in the ensemble. In all plots, we used the full Hamiltonian H in Eq. (1) in the main article, and set $N = 100$, $J = 3g_{\rm col}$, $\delta_p = J^2/20g_{\rm col}$, and $\kappa_p = 0.2\chi$. In order to show more clearly the quantum jump responsible for the two-atom decay, we further set $\kappa_s = \Omega = 0$.

Here,

$$H_{\rm ens} = \frac{i}{N}\chi_{\rm 2at}\left(S_-^2 - S_+^2\right), \tag{S14}$$

$$\chi_{\rm 2at} = \frac{2\Omega\chi}{\kappa_p}, \tag{S15}$$

$$\kappa_{\rm 1at} = \left(\frac{g_{\rm col}}{\Delta}\right)^2\kappa_s, \tag{S16}$$

$$\kappa_{\rm 2at} = \frac{4\chi^2}{\kappa_p}. \tag{S17}$$

To proceed, we assume that $\kappa_{\rm 1at} = 0$, such that the single-atom decay induced by the signal cavity is subtracted. Then, we obtain in the steady state

$$\left(S_-^2 - N\alpha^2\right)|D\rangle\langle D|S_+^2 - S_+^2\left(S_-^2 - N\alpha^2\right)|D\rangle\langle D| + \text{H.c.} = 0, \tag{S18}$$

where $|D\rangle$ is the dark state of the ensemble, and

$$\alpha = i\sqrt{2\chi_{\rm 2at}/\kappa_{\rm 2at}} = i\sqrt{\Omega/\chi}. \tag{S19}$$

This indicates that the dark state $|D\rangle$ satisfies

$$\left(S_-^2 - N\alpha^2\right)|D\rangle = 0. \tag{S20}$$

We now express $|D\rangle$, in terms of the eigenstates $|S=N/2, m_z=-N/2+n\rangle$ of the collective spin operator S_z, as

$$|D\rangle = \sum_n c_n |n\rangle, \tag{S21}$$

where, for simplicity, we have defined $|n\rangle \equiv |S=N/2, m_z=-N/2+n\rangle$. Here, n refers to the number of excited atoms in the ensemble. The condition in Eq. (S20) gives two recursion relations as follows

$$c_{2n+k} = \frac{\varepsilon^n}{\sqrt{(2n+k)!}} c_k, \tag{S22}$$

where $k=0,1$. Here, we have worked within the low-excitation regime, in which $\langle S_z\rangle \approx -N/2$, such that the main contributions to the dark state $|D\rangle$ are from these components with $n \ll N$.

The recursion relation in Eq. (S22) reveals that, when the ensemble is initially in a collective spin state $|n\rangle$ with an even n, e.g., in the ground state $|0\rangle$ (i.e., a spin coherent state with all atoms in the ground state), the dark state $|D\rangle$ can be expressed as,

$$|D\rangle_e = \frac{1}{\sqrt{\cosh |\alpha|^2}} \sum_n \frac{\alpha^{2n}}{\sqrt{(2n)!}} |2n\rangle. \tag{S23}$$

Similarly, when the ensemble is initially in a collective spin state $|n\rangle$ with an odd n, e.g., in the first excited state $|1\rangle$ (i.e., a state with only one atom is excited), the dark state $|D\rangle$ becomes

$$|D\rangle_o = \frac{1}{\sqrt{\sinh |\alpha|^2}} \sum_n \frac{\alpha^{2n+1}}{\sqrt{(2n+1)!}} |2n+1\rangle. \tag{S24}$$

On the other hand, the spin coherent state $|\theta,\phi\rangle$ is defined as

$$|\theta,\phi\rangle = R(\theta,\phi)\,|0\rangle. \tag{S25}$$

Here,

$$R(\theta,\phi) = \exp(\tau S_+) \exp\left[\ln\left(1+|\tau|^2\right) S_z\right] \exp\left(-\tau^* S_-\right), \tag{S26}$$

is a rotation operator with $\tau = \exp(i\phi)\tan(\theta/2)$. In the low-excitation limit, $S_+^n|0\rangle \approx \sqrt{n! N^n}|n\rangle$, and then

$$|\theta,\phi\rangle \approx \exp\left(-N|\tau|^2/2\right) \sum_n \frac{\left(\sqrt{N}\tau\right)^n}{\sqrt{n!}} |n\rangle. \tag{S27}$$

By setting $\sqrt{N}\tau = \alpha$, we further have

$$|D\rangle_{e,o} = \mathcal{A}_\pm \left(|\theta,\phi\rangle \pm |\theta,\phi+\pi\rangle\right) = |\mathcal{C}_\pm\rangle, \tag{S28}$$

where $\mathcal{A}_\pm = 1/\{2[1\pm\exp(-2|\alpha|^2)]\}^{1/2}$. This is what we have already given in Eq. (S1).

We now consider the case when the atomic ensemble is initialized in a spin coherent state $|\theta_0,\phi_0\rangle$. In this case, the atomic ensemble evolves into a subspace spanned by the cat states $\{|\mathcal{C}_+\rangle, |\mathcal{C}_-\rangle\}$ and, thus, its steady state is

$$\rho_{\rm ens}^{\rm ss} = c_{++}|\mathcal{C}_+\rangle\langle\mathcal{C}_+| + c_{--}|\mathcal{C}_-\rangle\langle\mathcal{C}_-| + c_{+-}|\mathcal{C}_+\rangle\langle\mathcal{C}_-| + c_{+-}^*|\mathcal{C}_-\rangle\langle\mathcal{C}_+|. \tag{S29}$$

To obtain the amplitudes c_{++}, c_{--}, and c_{+-}, we follow the method in Refs. [S11, S12], and after straightforward calculations, find that

$$c_{++} = \frac{1}{2}\left[1+\exp\left(-2|\alpha_0|^2\right)\right], \tag{S30}$$

$$c_{--} = 1 - c_{++} = \frac{1}{2}\left[1-\exp\left(-2|\alpha_0|^2\right)\right], \tag{S31}$$

$$c_{+-} = -\frac{\alpha_0^* |\alpha| \exp\left(-|\alpha_0|^2\right)}{\sqrt{2\sinh\left(2|\alpha|^2\right)}} \int_0^\pi d\varphi I_0\left(\left|\alpha^2 - \alpha_0^2 \exp(i2\varphi)\right|\right) \exp(-i\varphi), \tag{S32}$$

where $\alpha_0 = \sqrt{N}\exp(i\phi_0)\tan(\theta_0/2)$, and $I_0(\bullet)$ is the modified Bessel function of the first kind.

The above results show that the ensemble states are steered into a 2D quantum manifold spanned by the cat states $|\mathcal{C}_+\rangle$ and $|\mathcal{C}_-\rangle$. In typical atomic ensembles, spin relaxation is extremely weak, such that the dominant noise source is spin dephasing. However, the engineered two-atom decay can protect the cat states of the quantum manifold against spin dephasing. As a result, these cat states have a very long lifetime even with modest parameters, and thus, can be used for fundamental studies of quantum physics. Moreover, this atomic-cat-state manifold stabilized by the two-atom decay could also be used to encode logical qubits (i.e., cat qubits) for fault-tolerant quantum computation, as an alternative to the photonic-cat-state manifold stabilized by two-photon loss [S12].

S6. Spin relaxation, thermal noise, and the maximum cat state lifetime

In the main article, we discussed the effects of spin dephasing, and also showed that it can be strongly suppressed by the engineered two-atom decay. In this section, let us consider the effects of spin relaxation and thermal noise, and also the maximum cat state lifetime limited by them. Here, we proceed with the bosonic approximation. Such an approximation maps the spin coherent states $|\theta,\phi\rangle$ and $|\theta,\phi+\pi\rangle$ to the bosonic coherent states $|\pm\alpha\rangle$, respectively. Correspondingly, the cat states $|\mathcal{C}_\pm\rangle = \mathcal{A}_\pm(|\theta,\phi\rangle \pm |\theta,\phi+\pi\rangle)$ become $|\mathcal{C}_\pm\rangle = \mathcal{A}_\pm(|\alpha\rangle \pm |-\alpha\rangle)$, as given in Eq. (S2).

Spin relaxation and thermal noise can be described by the Lindblad dissipators, $\gamma_{\rm relax}(n_{\rm th}+1)\mathcal{L}(b)\rho$ and $\gamma_{\rm relax}n_{\rm th}\mathcal{L}(b^\dagger)\rho$. Here, $\gamma_{\rm relax}$ is the spin relaxation rate, and $n_{\rm th} = [\exp(\hbar\omega_q/k_BT)-1]^{-1}$ is the thermal average boson number at temperature T. The decay rate of the cat state coherence, which is induced by single-photon loss of the signal cavity, is given by

$$\Gamma_{\rm 1at} = 2|\alpha|^2\kappa_{\rm 1at}, \tag{S33}$$

with $\kappa_{\rm 1at} = (g_{\rm col}/\Delta)^2\kappa_s$ as given in Eq. (S12). At the same time, for a thermal background at $T\neq 0$, an additional decay rate of the cat state coherence, which is induced by spin relaxation and thermal noise, is given by [S13]

$$\Gamma_{\rm relax} = \left[2|\alpha|^2(1+2n_{\rm th}) + 2n_{\rm th}\right]\gamma_{\rm relax}. \tag{S34}$$

By assuming realistic parameters $\omega_q = 2\pi\times 3$ GHz, $T = 100$ mK, $|\alpha|^2 = 4$, and $\gamma_{\rm relax} = 2\pi\times 4$ mHz [S14, S15], we have $\Gamma_{\rm relax}\approx 2\pi\times 54$ mHz, much smaller the decay rate, $\Gamma_{\rm 1at}\approx 2\pi\times 8.0$ Hz, which is obtained with $\kappa_s = 2\pi\times 10$ kHz and $\Delta/g_{\rm col} = 100$. This means that the effects of both spin relaxation and thermal noise on the cat states $|\mathcal{C}_\pm\rangle$ can be safely neglected. In this case, the lifetime of these cat states is determined only by the single-atom decay rate $\kappa_{\rm 1at}$, and is given by

$$\tau_{\rm at} = \Gamma_{\rm 1at}^{-1} = \left(\frac{\Delta}{g_{\rm col}}\right)^2 \frac{1}{2|\alpha|^2\kappa_s}. \tag{S35}$$

On the other hand, the intracavity photonic cat states $|\mathcal{C}_\pm\rangle_{\rm ph}$ in Eq. (S3) mainly suffer from single-photon loss, e.g, with a rate κ_s, and thus their lifetime is given by [S16],

$$\tau_{\rm ph} = \frac{1}{2|\alpha|^2\kappa_s}. \tag{S36}$$

It is found from Eqs. (S35) and (S36) that $\tau_{\rm at}$ is longer than $\tau_{\rm ph}$ by a factor of $(\Delta/g_{\rm col})^2$, i.e.,

$$\frac{\tau_{\rm at}}{\tau_{\rm ph}} = \left(\frac{\Delta}{g_{\rm col}}\right)^2. \tag{S37}$$

According to the analysis in the main article, the factor $(\Delta/g_{\rm col})^2$ can be tuned to be $\sim 10^4$ under modest parameters. This indicates that *the lifetime of our atomic cat states is longer than that of intracavity photonic cat states by up to four orders of magnitude for cat sizes of* $|\alpha|^2 \geq 4$.

In fact, the decoherence rate $\Gamma_{\rm 1at}$ can be further decreased with the smaller single-photon loss rate κ_s (i.e., the longer T_c). This results in a longer cat state lifetime. When $\Gamma_{\rm 1at}$ is comparable to or even smaller than $\Gamma_{\rm relax}$, the lifetime $\tau_{\rm at}$ is given by

$$\tau_{\rm at} = (\Gamma_{\rm 1at} + \Gamma_{\rm relax})^{-1}. \tag{S38}$$

For a single-photon loss rate of $\kappa_s/2\pi = 30$ Hz, we have $\Gamma_{1\mathrm{at}} = 2\pi \times 24$ mH, which is smaller than $\Gamma_{\mathrm{relax}} \sim 2\pi \times 54$ mHz. In this case, Eq. (S38) gives a cat state lifetime of $\tau_{\mathrm{at}} \sim 2$ sec. Ultimately, when decreasing the rate κ_s, the lifetime τ_{at} increases to its maximum value,

$$\tau_{\mathrm{at}}^{\mathrm{max}} = \Gamma_{\mathrm{relax}}^{-1}. \tag{S39}$$

Using the parameters given above, we can predict a maximum lifetime of $\tau_{\mathrm{at}}^{\mathrm{max}} \sim 3$ sec.

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