Cavity Quantum Electrodynamics Effects with Nitrogen Vacancy Center Spins in Diamond and Microwave Resonators at Room Temperature

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Cavity quantum electrodynamics (C-QED) effects, such as Rabi splitting, Rabi oscillations and superradiance, have been demonstrated with nitrogen vacancy center spins in diamond in microwave resonators at cryogenic temperature. In this article we explore the possibility to realize strong collective coupling and the resulting C-QED effects with ensembles of spins at room temperature. Thermal excitation of the individual spins by the hot environment leads to population of collective Dicke states with low symmetry and a reduced collective spin-microwave field coupling. However, we show with simulations that the thermal excitation can be compensated by spin-cooling via optical pumping. The resulting population of Dicke states with higher symmetry implies strong coupling with currently available high-quality resonators and enables C-QED effects with potential applications in quantum sensing and quantum information processing.

Introduction

Cavity quantum electrodynamics (C-QED) studies the interaction between quantum emitters and cavity photon modes, and addresses both the foundations of quantum mechanics [1] and proposals for quantum information processing [2] and quantum metrology [3]. C-QED effects, such as Rabi splitting [4–6], Rabi-oscillations [7] and superradiance [8], have been realized with negatively charged nitrogen vacancy (NV–) center spins in diamond in microwave resonators at cryogenic temperature. Although the NV– center spins have a spin-1 degree of freedom, normally only two of the spin states, say those with projections \( m = 0 \) and \( m = +1 \), are coupled resonantly to the resonator, and thus the spins can be treated effectively as two-level systems. As illustrated in Fig. 1(a), at low temperature the spin ensemble can be highly polarized and can couple strongly with lumped-element microwave resonators with high Q-factors.

The low temperature restricts the application of the C-QED effects in quantum sensing and quantum information processing, and it would be desirable if the collective coupling could be achieved at room temperature. In this article, we study how the temperature affects the quantum state of the spin ensemble, and we propose to utilize optical pumping to counteract the thermal heating to achieve strong spin polarization and strong coupling, and to observe the resulting C-QED effects at room temperature with a high-quality dielectric resonator, see Fig. 1(b). Note that the strong collective coupling may have been present in the recent room-temperature experiments on continuous wave maser [9] and dispersive readout of NV– spins [10, 11] and C-QED effects with the spins in pentacene’s lowest triplet states [12].

![Figure 1. C-QED systems with a NV center spin ensemble.](image)

Panel (a) shows a 3D lumped element resonator operating in a cryogenic environment at 25 mK (left), as used in [8], where the spin-ensemble is in equilibrium with the cooled environment (right). Panel (b) shows a sapphire dielectric resonator operating at room temperature, as used in [9], and a diamond illuminated by 532 nm laser light (left), which counteracts the heating of the spins by the hot environment (right), leading to a higher population in the lower 0 spin state than in the upper +1 spin state. The upwards (downwards) arrows in the panels (a,b) represent the thermal excitation, thermal decay, and the spin-cooling \( \eta_s \) via optical pumping.

Nitrogen Vacancy Center Spin Ensemble

Nitrogen vacancy centers are formed by replacing two adjacent carbon atoms with a nitrogen atom and a vacancy in the diamond lattice. Negatively charged NV– centers have a
spin-triplet electronic ground state, which contains three spin states with projection $m = +1, 0, -1$ along the axis defined by the nitrogen atom and the vacancy. The $\pm 1$ spin states have higher energy than the 0 state, and can be split and shifted by a static magnetic field. In this way, the transition between one of the shifted states and the 0-state can be tuned into resonance with a microwave resonator. Furthermore, we can align the magnetic field along one of four possible nitrogen-vacancy orientations, and ensure that only the associated 0, $+1$ spin states couple resonantly with the microwave resonator. These spin states can be viewed as the eigenstates of an effective pseudo 1/2-spin, and $N$ NV$^-$ centers with this orientation can be viewed as an ensemble of $N$ pseudo spins.

Dicke Representation of Collective States  The collective coupling of the 1/2-spin ensemble with the quantized electromagnetic field inside the resonator is conventionally described by the so-called Dicke states $|J, M\rangle$. The half-integer or integer number $J \leq J_0 = N/2$ refers to the eigenvalues $J(J+1)$ of the collective spin operator $J^2$, and the number $M$ within the range $-J \leq M \leq J$ describes the degree of the spin excitation. For each $J$, the Dicke states of different $M$ form a vertical ladder, and the states with different $J$ form the horizontally shifted ladders, constituting a triangular space [Fig. 2(a)]. Due to permutation symmetry, for each pair of $J, M$, there are a total of $N!(2J+1)/[(J+J_0+1)!(J_0-J)!]$ states, i.e., a degeneracy in the Dicke state energy diagrams, and they couple identically with the cavity field.

The uniform coupling to the microwave resonator and the individual (but identical) incoherent excitation and decay processes can be consistently and effectively treated as transitions among the Dicke states [14][16]. In Fig. 2(a), we show how the interaction with the thermal environment causes quantum jumps that change $J$ by $\pm 1$ and $M$ by $0, \pm 1$ (red and blue arrows), while the spin-cooling via optical pumping adds a term $\eta_s$ to the rate $(1+\eta_s)\gamma_s$ of downward jumps (blue arrows). Here, $\eta_s = (\hbar \omega_s/k_B T - 1)^{-1}$ is the thermal equilibrium phonon excitation at the spin transition energy $\hbar \omega_s$ and temperature $T$ ($k_B$ is Boltzmann’s constant). A pure single spin dephasing rate $\chi_s$ is incorporated to describe the phase noise and inhomogeneous broadening of the transition frequencies, and leads to quantum jumps to states with unchanged $M$ and different $J$ (horizontal black arrows), while the coherent collective coupling with the resonator (with $g_s$ as the coupling to single spin) causes transitions between states of different $M$ and same $J$ (orange arrows). The actual rates also depend on the $J$ and $M$ and for combinatorial reasons the jump probability is larger towards the Dicke states with reduced $J$ (and increased degeneracy) as reflected by the thickness of the lines, see Appendix A in [14] for precise expressions.

In the presence of the thermal processes and the spin cooling, each spin is in a mixed steady state with the upper state population, $p = \eta_s^{1/2} \gamma_s/\eta_s + \gamma_s(1+2\eta_s^{1/2})$. This is equivalent to a distribution on the Dicke states [20], with average values $M, J$ of the quantum numbers defined by the corresponding operator expectation values, $M = J_0(2p - 1)$, and $J(J+1) = (2p - 1)^2 J_0(J_0 + 1) + 6p(p - 1)J_0$. For many spins, the relative fluctuations of these quantities are small, and we may thus depict the location of the ensemble steady-state as points in the Dicke state diagram in Fig. 2(b).

Without the optical spin-cooling $\eta_s = 0$, the spin ensemble occupies Dicke states with smaller $J$ for temperature above $T > 10$ K, and occupies the states with larger $J$ (lower-right corner) only for $T < 0.01$ K, see the dots along the edge of the left triangle in Fig. 2(b). As the collective spin-resonator coupling scales with $\sqrt{J}$, the spin-ensemble has to be held at very low temperature to ensure the strong collective coupling. In contrast, with sufficient optical spin-cooling (values of $\eta_s$ up to 1 MHz should be achievable in experiments [19]) to compensate the thermal process, the spin-ensemble can be prepared in Dicke states with larger $J$, see the dots along the rightmost triangle in Fig. 2(b), enabling the strong collective coupling.
coulping. The spin-ensemble cooling at room temperature as discussed here is fundamentally different from the rapid dephasing-induced and Purcell-enhanced spin cooling via a cavity mode, coupled to a cold environment as proposed in [27]. In the numerical calculations presented below, we shall employ a mean-field approach as in [19–21], and use the Dicke state representation only for visualization and interpretation of the results.

Quantum Master Equation To assess the C-QED effects with the NV− spin ensemble-microwave resonator system at room temperature, we must solve the quantum master equation for the reduced density operator \( \hat{\rho} \),

\[
\frac{\partial}{\partial t} \hat{\rho} = -\frac{i}{\hbar} \left[ \hat{H}_c + \hat{H}_d + \hat{H}_s + \hat{H}_{s-c}, \hat{\rho} \right] - \kappa_s \left[ (1 + n_{th}^s) D [\hat{a}] \hat{\rho} + n_{th}^s D [\hat{a}^+] \hat{\rho} \right] - \gamma_s \left[ (1 + n_{th}^s) \sum_j D [\hat{a}_j^2] \hat{\rho} + n_{th}^s \sum_j D [\hat{a}_j^1] \hat{\rho} \right] - \eta_s \sum_j D [\hat{a}_j^1] \hat{\rho} - 2\chi_s \sum_j D [\hat{a}_j^2] \hat{\rho},
\]

where \( \hat{H}_c = h \omega_c \hat{a}^\dagger \hat{a} \) describes the microwave resonator with frequency \( \omega_c \), photon creation \( \hat{a}^\dagger \) and annihilation operator \( \hat{a} \), \( \hat{H}_d = h \Omega \sqrt{N} \hat{a}^\dagger \hat{a} \) and \( h.c. \) describes the driving of the resonator by a microwave probe field with amplitude \( \Omega \), frequency \( \omega_d \) and the resonator coupling coefficient \( \sqrt{N} \) \( (\kappa_s \) is the resonator loss rate due to the same coupling). The spin Hamiltonian \( \hat{H}_s = h \omega_s \sum_{j=1}^N \hat{\sigma}_j^z \) involves the projection operator \( \hat{\sigma}_j^z \) on the upper level of the \( j \)th spin, and the spin-resonator interaction Hamiltonian \( \hat{H}_{s-c} = h g_s \left( \hat{a}^\dagger \sum_j \hat{\sigma}_j^z + \sum_j \hat{\sigma}_j^2 \right) \) depends on the lowering \( \hat{\sigma}_j^z \) and raising operators \( \hat{\sigma}_j^2 \) of the spin transitions.

The second line of Eq. 1 describes the thermal emission and excitation of the resonator with a rate \( \kappa \) and a thermal equilibrium photon number \( n_{th}^s = \left[ e^{h \omega_s / k_B T} - 1 \right]^{-1} \) at temperature \( T \). Here, the Lindblad superoperator for any operator \( \hat{\sigma} \) is defined as \( D [\hat{\sigma}] \hat{\rho} = \frac{1}{2} \left( \hat{\sigma}^\dagger \hat{\rho} + \hat{\rho} \hat{\sigma}^\dagger - \hat{\sigma} \hat{\rho}^\dagger - \hat{\rho}^\dagger \hat{\sigma} \right) \). The third and fourth line describe the thermal process, the optical spin-cooling and the dephasing of the spins, as already discussed in Fig. 2. Note that the single dephasing rate modeling of the inhomogeneous broadening is expected to offer a good approximation to the general behavior of the spin ensemble, which can be verified with a more delicate modeling with subensembles of different transition frequencies [8] [19] [28].

To solve Eq. 1, we follow the mean-field approach [20] [21] (also known as the cluster-expansion method [22]) to derive the equation \( \frac{\partial}{\partial \tau} \langle \hat{\sigma} \rangle = \text{tr} \left\{ \left( \frac{\partial}{\partial \tau} \hat{\rho} \right) \hat{\sigma} \right\} \) for the mean value \( \langle \hat{\sigma} \rangle = \text{tr} \left\{ \hat{\rho} \hat{\sigma} \right\} \) of any operator \( \hat{\sigma} \), and truncate the resulting equation hierarchy by approximating the mean values of products of many operators with those of fewer operators. To automatically derive the mean-field equations to second order and solve these equations numerically, we make use of the QuantumCumulant.jl package [23], and present our code and the resulting equations in App. A and B.

The obtained equations involve first-order mean quantities, i.e. the resonator field amplitude \( \langle \hat{a} \rangle \), the spin coherence \( \langle \hat{\sigma}^z \rangle \) and the spin upper-level population \( \langle \hat{\sigma}^x \rangle \), and second-order mean quantities, i.e. the resonator photon number \( \langle \hat{a}^\dagger \hat{a} \rangle \), photon correlation \( \langle \hat{a} \hat{a}^\dagger \rangle \), and the spin-photon correlations \( \langle \hat{a}^\dagger \hat{\sigma}^z \rangle, \langle \hat{\sigma}^x \rangle, \langle \hat{\sigma}^{x^2} \rangle \), the spin-spin correlations \( \langle \hat{\sigma}_1^{z} \hat{\sigma}_2^{z} \rangle, \langle \hat{\sigma}_1^{z} \hat{\sigma}_2^{x} \rangle, \langle \hat{\sigma}_1^{x} \hat{\sigma}_2^{z} \rangle, \langle \hat{\sigma}_1^{x^2} \hat{\sigma}_2^{x^2} \rangle \), as well as their complex conjugate quantities.

To simulate a system with a trillion spins, we have assumed the same parameters \( \omega_s, \gamma_s, n_{th}^s, \eta_s, \chi_s \) for all the NV− spins, so that we can utilize the aforementioned quantities (with subscripts 1 or 2) to represent \( \langle \hat{\sigma}_j^{mn} \rangle, \langle \hat{\sigma}_j^{mn} \rangle, \langle \hat{\sigma}_j^{mn} \rangle, \langle \hat{\sigma}_j^{mn} \rangle, \langle \hat{\sigma}_j^{mn} \rangle \), \( (m, n, m', n' = 1, 2) \), which have the same values for all the spins \( j \) and spin pairs \( (j, j') \). Although the mean-field approach does not deal directly with the Dicke states, the aforementioned mean values do allow us to calculate the average of the Dicke state quantum numbers [19–21], as, \( M = N \langle \hat{a}^2 \rangle + 1/2 \) and \( J = \sqrt{\frac{3}{2} N + N(N-1) \langle \hat{a}^2 \hat{a}^x \rangle + \langle \hat{a}^2 \hat{a}^x \rangle - \langle \hat{a}^2 \rangle + 1/2} \).

Rabi Oscillations and Splitting at Room Temperature To observe collective Rabi oscillations, we drive the optically cooled spin ensemble with a resonant square microwave pulse of 1 \( \mu \)s duration, as in the experiment [27], and calculate the photon number inside the resonator, see Fig. 3(a). We observe that the photon number increases and decays with an oscillatory amplitude behavior when the driving field switches on and off. Without the optical spin cooling \( \eta_s = 0 \) we observe no Rabi oscillations (black curve), while already for a weak rate \( \eta_s = 5 \times 10^2 \) (blue dashed line), oscillations appear and become faster for higher rates \( \eta_s = 10^3, 10^4 \) Hz (red dash-dotted line and green solid line), which confirms the increased coupling to the more symmetric states with larger \( J \) [inset of Fig. 3(a)]. We also note that the oscillations end earlier for larger values of \( \eta_s \).
Stimulated Superradiance Pulses at Room Temperature

We now study the transient excitation and subsequent decay of the spin-ensemble by the collective coupling to the cavity mode at room temperature [Fig. 4]. In our simulations, we drive the resonator with a strong microwave field to excite the spin-ensemble [marked by "(1)"], which is initially cooled by optical pumping to Dicke states with different degrees of symmetry. Then, we switch off the microwave driving, and the spin ensemble dephases and decays towards lower excitation states with reduced values of $J$, leading to the radiation pulse [marked by "(2)"]. Finally, the optical spin-cooling and dephasing re-initialize the spin-ensemble into states with larger $J$ [marked by "(3)"]. The simulated photon number dynamics [Fig. 4(a)] is similar to the results observed in experiments at cryogenic temperature [8], but it oc-
Rabi splitting and stimulated superradiance can be re-
compatible with existing experimental setups, we show 
coupling to a microwave resonator. Using parameters 
in Dicke states with high symmetry and strong collective 
cryogenic environment.

which is much faster than the thermal cooling within a 
ment and prepare and maintain an NV 
\[\text{spin polarization at room temperature and the resulting} \]
alized at room temperature. The realization of strong 
spin polarization at room temperature and the resulting 
collective coupling may enable further applications and 
and self-stimulated spin echoes \cite{21} in ambient 

Conclusions In summary, we have demonstrated the-
theoretically that optical pumping can counteract thermal-
ization of spin states in a room temperature environ-
ment and prepare and maintain an NV$^-$ spin-ensemble in 
Dicke states with high symmetry and strong collective 
coupling to a microwave resonator. Using parameters compatible with existing experimental setups, we show that C-QED effects, such as collective Rabi oscillations, Rabi splitting and stimulated superradiance can be re-
curs here with spin cooling in an apparatus maintained 
at room temperature. We note that due to the strong 
spin dephasing in our system, right after the emission 
[Fig. 4(b)] the population of the upper spin state de-
cays first to some finite value around 0.5, equivalent to 
Dicke states with low symmetry and small $J$. Our results 
also show that the radiation pulses become stronger and 
shorter for larger values of $\eta_s$. Because of the involvement 
of the stimulated emission, the large number of photons 
and the exploration of collective Dicke states, the radiation 
is qualified as stimulated superradiance. With the 
optical pumping, after the radiation pulse the spin en-
semble is cooled to the desired Dicke states with higher 
symmetry within $4\mu$s to $300\mu$s for $\eta_s = 10^6$ Hz to $10^4$ Hz, 
which is much faster than the thermal cooling within a 
cryogenic environment.

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AUTHOR CONTRIBUTIONS

Yuan Zhang and Qilong Wu contribute equally to this work. All the authors contribute to the writing of the manuscript.

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Appendix A: Julia Code to Derive and Solve Mean-Field Equations

In Fig. 5 (a), we show the Julia code used to derive the second order mean-field equations from the master equation \([1]\). In lines 1 and 2, we import the QuantumCumulants.jl package, and then define the symbols to represent the complex numbers. In lines 3 and 4, we define the Hilbert space for the two-level spin, and the special Hilbert space for \(N\) two-level spins, which utilizes the symmetry of identical spins and is restricted to the second order. In lines 5 and 6, we define the Fock space for microwave photons in the resonator (a quantized harmonic oscillator), and the product Hilbert space for the spins-resonator system. In lines 7 and 8, we define the photon annihilation operator \(a\) (and its conjugate \(a'\)), and the projection, raising and lowering operators of the spins. For example, \(\sigma(2,2)[1], \sigma(2,2)[2]\) are the upper-level projection operators of the first and second spin, and \(\sigma(1,2)[1], \sigma(1,2)[2]\) are the lowering operators of the two representative spin. Note that the lower-level projection operators \(\sigma(1,1)[1], \sigma(1,1)[2]\) are equal to \(1 - \sigma(2,2)[1], 1 - \sigma(2,2)[2]\), and are thus not directly considered in the program. In the line 9, we define the system Hamiltonian in a frame rotating with the driving field frequency, where the function \(\text{sum}(\sigma(i, j)) = \sigma(i, j)[1] + \sigma(i, j)[2]\) gives the sum of the spin operators. In lines 10 and 11, we define the operators and the rates used to specify the Lindblad terms in the master equation. In lines 12 and 13, we specify the photon number operator \(a'a'\) and the high-level projection operator of the first spin, and then derive the mean-field equations in second order for the expectation value of these operators. The program replaces the sum of spins (spin pairs) by \(N\) \((N(N-1))\) times the values for a single spin (spin pair), and apply the third-order cumulant approximation \((\hat{o}\hat{p}\hat{q}) \approx \langle \hat{o} \rangle \langle \hat{p} \hat{q} \rangle + \langle \hat{a} \hat{q} \rangle \langle \hat{p} \rangle + \langle \hat{q} \rangle \langle \hat{o} \hat{p} \rangle - 2 \langle \hat{q} \rangle \langle \hat{o} \hat{p} \rangle \langle \hat{q} \rangle\) (for any operators \(\hat{o}, \hat{p}, \hat{q}\)) to account correctly for expectation values up to second order. In the last line

Figure 5. Julia codes used to derive and solve our mean field equations. Panel (a) shows the code to derive the symbolic equations for the second order mean-field quantities from the master equation \([1]\). We work in a frame rotating with the driving field frequency \(\Delta_o = \omega_o - \omega_d\), \(\Delta_s = \omega_s - \omega_d\). Panel (b) shows the code to convert the symbolic equations to the numerical ones, and to solve these equations with numerical methods.
14, we identify other first and second order mean-field quantities appearing in the derived equations, and we derive the equations for these quantities to finally form a closed set of equations, see the Appendix B.

In Fig. 5(b), we show the Julia code to solve the mean-field equations. In line 1, we specify the thermal energy at room temperature, and the Planck constant. In lines 2 and 3, we specify the resonator frequency, the photon loss rate, and the thermal photon number. In lines 4 and 5, we specify the spin transition frequency, the spin-relaxation rate, the spin cooling rate, the dephasing rate, and the thermal phonon coupling rate. In line 6, we specify the number of spins and the spin-resonator coupling. In line 7, we specify the frequency and amplitude of driving field, and the frequency detunings. In lines 8-11, we initialize the mean-field quantities that all vanish except the mean photon number $u[0]$, the equilibrium population of higher spin level $u[2]$, and the spin-spin correlation $\langle \sigma_2^2 \sigma_2^2 \rangle u[12]$. In lines 12 and 13, we define the array of symbolic parameters, and their numerical values. In line 14, we import the OrdinaryDiffEq.jl, ModelingToolkit.jl and Plots.jl package for ordinary differential equations, the symbolic-numerical computation and the plot function. In lines 15-17, we convert the symbolic equations to a form recognized by the ModelingToolkit.jl module, and we define the ordinary differential equation problem by giving the equations, the initial values, the simulation time and the parameters, and finally solve the problem with Rouge-Kutta method. In lines 18-20, we extract the mean photon number and the population of the upper spin level, and plot their numerical solutions. The code given here can be modified to produce all the results shown in the main text and the appendix.

Appendix B: Mean Field Equations in Second-order

Our analysis reveals that the three first-order mean-field quantities for the resonator field amplitude $\langle a^\dagger \rangle$, the spin coherence $\langle \sigma_1^2 \rangle$, and the population of the upper spin level $\langle \sigma_2^2 \rangle$, obey the following equations:

$$\partial_t \langle a^\dagger \rangle = i \Delta_e \langle a^\dagger \rangle + i \Omega \sqrt{n_1} + i N_g s \langle \sigma_2^2 \rangle,$$

$$\partial_t \langle \sigma_1^2 \rangle = -i \Delta_s \langle \sigma_1^2 \rangle - i g_s \langle a \rangle + 2ig_s \langle a \sigma_2^2 \rangle,$$

$$\partial_t \langle \sigma_2^2 \rangle = n_s \gamma_s - [\gamma_s (1 + 2n_s)] \langle \sigma_2^2 \rangle + ig_s \langle (a^\dagger \sigma_1^2 - \langle a \sigma_2^2 \rangle \rangle. $$

Here and in the following, we work in a frame rotating with the driving field frequency $\omega_d$, and define the frequency detunings $\Delta_e = \omega_e - \omega_d$, $\Delta_s = \omega_s - \omega_d$, and the complex frequency detunings $\Delta_e = \Delta_e - i \kappa/2$ and $\Delta_s = \Delta_s - i \kappa/2$. We encounter two second order mean-field quantities, i.e. the mean photon number $\langle a^\dagger a \rangle$, the photon correlation $\langle aa \rangle$, the spin-photon correlations $\langle a^\dagger \sigma_1^2 \rangle$, $\langle a^\dagger \sigma_2^2 \rangle$, and the spin-spin correlations $\langle \sigma_1^2 \sigma_2^2 \rangle$, $\langle \sigma_1^2 \sigma_2^2 \rangle$, $\langle \sigma_1^2 \sigma_1^2 \rangle$. The equations for the photon number and correlation read

$$\partial_t \langle a^\dagger a \rangle = \kappa_s (n_e^a - \langle a^\dagger a \rangle) + i \Omega \sqrt{n_1} (\langle a^\dagger a \rangle) + i N_g s (\langle a \sigma_2^2 \rangle - \langle a^\dagger \sigma_2^2 \rangle),$$

$$\partial_t \langle a \rangle = -2i \Delta_e \langle a \rangle - 2i \Omega \sqrt{n_1} \langle a \rangle - 2i N_g s \langle a \sigma_2^2 \rangle.$$
By examining the mean-field equations, we note that the spin ensemble occupies states near the lower boundary of the Dicke space, which permits us to apply the Holstein-Primakoff approximation \cite{17, 19} and approximate these states as the occupation number states of quantized harmonic oscillators. As a result, we can approximate the Hamiltonian of the spin-ensemble as $\hat{H}_s \approx \sum _J \hat{H}_{s,J}$ with $\hat{H}_{s,J} = J \omega_s \hat{b}_J^\dagger \hat{b}_J$, where $\hat{b}_J^\dagger, \hat{b}_J$ are the bosonic creation and annihilation operator for given $J$ and the spin ensemble-resonator coupling Hamiltonian as $\hat{H}_{a-c} \approx \sum _J \hat{H}_{a-c,J}$ with $\hat{H}_{a-c,J} = \hbar \sqrt{2 J g_s (\hat{a} \hat{b}_J + \hat{b}_J^\dagger \hat{a})}$, where the coupling strength $\sqrt{2 J g_s}$ depends on the numbers $J$. Since $\hat{H}_s$ is sum of $\hat{H}_{s,J}$ over the different $J$, the system response can be viewed as the sum of response for sub-system with given $J$ weighted by the population on the corresponding Dicke ladder. For given $J$, we can diagonalize the approximate Hamiltonian $\hat{H}_{s,J} + \hat{H}_{a-c,J} + \hat{H}_{a-c,J}$ as $\sum _{n=\pm} \hbar \omega_n \hat{h}_n^\dagger \hat{h}_n$ with the frequencies $\omega_n = \frac{1}{2} (\omega_s + \omega_c \pm \chi)$ and the operators $\hat{h}_n = \sqrt{1 + \frac{\omega_n - \omega_c}{\chi}} \left[ \frac{\pm \sqrt{\omega_n}}{\sqrt{2}} + \frac{g_s \sqrt{2 J}}{\chi} (\hat{a} \hat{b}_J + \hat{b}_J^\dagger \hat{a}) \right]$ of the hybrid modes. Here, the abbreviation is defined as $\chi = \sqrt{8 g_s^2 J + (\omega_s - \omega_c)^2}$ for the particular case with $\omega_s = \omega_c$, we obtain $\chi = 2 \sqrt{2 J g_s}$ and $\omega_n - \omega_c = \pm \sqrt{2 J g_s}$.

Appendix C: Simulations Parameters

In this appendix, we summarize the parameters used for the simulations in Fig. 3 and Fig. 4 in the main text. For Fig.3, we utilize the parameters compatible with the Rabi oscillation experiment at cryogenic temperature \cite{7}: the resonator has the frequency $\omega_c = 2 \pi \times 2.69$ GHz, the photon loss rate $\kappa_c = 2 \pi \times 0.8$ MHz ($\kappa_1 = \kappa_c / 2$), and the coupling with single spin $g_s = 2 \pi \times 12$ Hz; the spins have the transition frequency $\omega_s = \omega_c$, the spin-lattice relaxation rate $\gamma_s = 2 \pi \times 0.157$ Hz, and the dephasing $\chi_s = 2 \pi \times 2.6$ MHz. In addition, the number of spins is $N = 2.5 \times 10^{12}$. For Fig. 3(a), the driving microwave field drives resonantly the resonator ($\omega_d = \omega_c$) with the amplitude $\Omega = 2 \pi \times 10^6$ Hz$^{-1/2}$ Hz$^{-1/2}$. For Fig. 3(b), the driving microwave field has the amplitude $\Omega = 2 \pi \times 10^6$ Hz$^{-1/2}$ Hz$^{-1/2}$. For Fig. 3, we utilize the parameters compatible with the stimulated superradiance pulse experiment at cryogenic temperature \cite{8}: the resonator has the frequency $\omega_c = 2 \pi \times 3.18$ GHz, the photon loss rate $\kappa_c = 2 \pi \times 13.8$ MHz ($\kappa_1 = \kappa_c / 2$), and the coupling with single spin $g_s = 2 \pi \times 83.1$ mHz; the spins have the transition frequency $\omega_s = \omega_c$, the spin-lattice relaxation rate $\gamma_s = 2 \pi \times 0.157$ Hz, and the dephasing rate $\chi_s = 2 \pi \times 4.7$ MHz. The number of the spins is $N = 1.5 \times 10^{10}$, and the driving microwave field drives the resonator resonantly ($\omega_d = \omega_c$) with the field amplitude $\Omega = 2 \pi \times 17 \times 10^{10}$ Hz$^{-1/2}$ and duration of 28 ns.

Appendix D: Hybrid Modes under the Holstein-Primakoff Approximation

In Fig. 3 of the main text, we show that the collective interaction of the optically cooled spin ensemble with the resonator field leads to Rabi oscillations and cavity mode splittings at room temperature. To gain more insights into the physics, we note that the spin ensemble occupies states near the lower boundary of the Dicke space, which permits us to apply the Holstein-Primakoff approximation \cite{17, 19} and approximate these states as the occupation number states of quantized harmonic oscillators. As a result, we can approximate the Hamiltonian of the spin-ensemble as $\hat{H}_s \approx \sum _J \hat{H}_{s,J}$ with $\hat{H}_{s,J} = J \omega_s \hat{b}_J^\dagger \hat{b}_J$, where $\hat{b}_J^\dagger, \hat{b}_J$ are the bosonic creation and annihilation operator for given $J$ and the spin ensemble-resonator coupling Hamiltonian as $\hat{H}_{a-c} \approx \sum _J \hat{H}_{a-c,J}$ with $\hat{H}_{a-c,J} = \hbar \sqrt{2 J g_s (\hat{a} \hat{b}_J + \hat{b}_J^\dagger \hat{a})}$, where the coupling strength $\sqrt{2 J g_s}$ depends on the numbers $J$. Since $\hat{H}_s$ is sum of $\hat{H}_{s,J}$ over the different $J$, the system response can be viewed as the sum of response for sub-system with given $J$ weighted by the population on the corresponding Dicke ladder. For given $J$, we can diagonalize the approximate Hamiltonian $\hat{H}_{s,J} + \hat{H}_{a-c,J} + \hat{H}_{a-c,J}$ as $\sum _{n=\pm} \hbar \omega_n \hat{h}_n^\dagger \hat{h}_n$ with the frequencies $\omega_n = \frac{1}{2} (\omega_s + \omega_c \pm \chi)$ and the operators $\hat{h}_n = \sqrt{1 + \frac{\omega_n - \omega_c}{\chi}} \left[ \frac{\pm \sqrt{\omega_n}}{\sqrt{2}} + \frac{g_s \sqrt{2 J}}{\chi} (\hat{a} \hat{b}_J + \hat{b}_J^\dagger \hat{a}) \right]$ of the hybrid modes. Here, the abbreviation is defined as $\chi = \sqrt{8 g_s^2 J + (\omega_s - \omega_c)^2}$ for the particular case with $\omega_s = \omega_c$, we obtain $\chi = 2 \sqrt{2 J g_s}$ and $\omega_n - \omega_c = \pm \sqrt{2 J g_s}$.

Appendix E: Sensing with the Rabi Splitting Signal

In this appendix, we show the dependence of the Rabi splittings on the spin transition frequency $\omega_s$, see Fig. 6(c), which can be potentially useful for sensing of magnetic fields and other related quantities. Fig. 6(a) shows that the peaks blue-shift as $\omega_s$ increases over the resonator frequency, and the maximum of the peaks becomes also stronger. From the analysis in Appendix D, we obtain the frequencies of the hybrid modes $\omega_n = \frac{1}{2} (\omega_s + \omega_c \pm \sqrt{8 g_s^2 J + (\omega_s - \omega_c)^2})$ leading to these peaks. For smaller frequency detuning $|\omega_s - | \ll 2 \sqrt{2 J g_s}$, these frequencies can be approximated as $\omega_n \approx \frac{1}{2} (\omega_s + \omega_c \pm 2 \sqrt{2 J g_s})$. The linear scaling of these frequency with $\omega_s$ can be used to infer the change of the spin transition frequency. Furthermore, we note that the sum of the frequencies of the two resonant Rabi peaks $\omega_+ + \omega_- = \omega_s + \omega_c$ are independent of the collective coupling $\sqrt{2 J g_s}$ and are also proportional to $\omega_s$. Thus, using the frequency sum, we can infer the spin transition frequency $\omega_s$ more faithfully.

Besides measuring the frequencies of peaks, we can also determine the change of photon number and hence transmission as a function of the spin transition frequency $\omega_s$ for different values of the frequency $\omega_d$ of the microwave probe field. Such results are shown in Fig. 6(b), and we see that, when the probe field is resonant with the cavity ($\omega_d = \omega_c$, the red solid line), the photon number is minimal for $\omega_s = \omega_c$ and increases linearly and smoothly with increasing detuning $\omega_s - \omega_c$. In contrast,
Figure 6. Sensing with Rabi splitting signals. Panel (a) shows the intra-resonator photon number, and hence the cavity transmission, as function of the frequency detuning $\omega_d - \omega_c$ of the probe field from the resonator for different values of the spin detuning $\Delta = \omega_s - \omega_c$. Here, from the left (green, dot-dashed) curve to the right (blue, dashed and red, solid) ones, the detuning is $\Delta/(2\pi) = -2, 0, 2$ MHz, respectively. Panel (b) shows the photon number, and hence the cavity transmission, as function of the spin detuning $\Delta = \omega_s - \omega_c$ for $\omega_d = \omega_c$ (red solid line), $\omega_d = \omega_c - 2\pi \times 1.59$ MHz (green dot-dashed line) and $\omega_d = \omega_c + 2\pi \times 1.59$ MHz (blue dashed line). The driving field strength is $\Omega = 2\pi \times 5 \times 10^7$ Hz, and the other parameters are the same as those used in Fig. 3(b).

when the probe field is detuned from the resonator (e.g. $\omega_d - \omega_c = \pm 2\pi \times 1.59$ MHz, the green dot-dashed and blue dashed lines), the photon transmission shows sharp peaks when $\omega_s = \omega_c \pm 2\pi \times 125$ MHz, and relatively broad dips around $\omega_s = \omega_c$. Note that the detuning of these peaks to the resonator is much larger than the dephasing rate induced by the inhomogeneous broadening $\chi = 2\pi \times 2.6$ MHz. The different behaviors in the resonant and off-resonant case may be beneficial for the wide-band, and narrow-frequency sensing of the spin transition frequency.