Cavity cooling of an ensemble spin system

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We describe how sideband cooling techniques, prevalent in quantum optics, may be applied to large spin ensembles in magnetic resonance. Using the Tavis-Cummings model in the presence of a Rabi drive, we solve a Markovian master equation describing the joint spin-cavity dynamics to derive cooling rates as a function of ensemble size. Our calculations indicate that a spin ensemble containing roughly $10^{11}$ electron spins may be polarized to a non-thermal equilibrium state in a time many orders of magnitude shorter than the typical thermal relaxation time. The described techniques permit the efficient removal of entropy for spin-based quantum information processors and fast polarization of spin samples. The proposed application of a standard technique in quantum optics to magnetic resonance also serves to reinforce the connection between the two fields, which has only recently begun to be explored in detail due to the development of hybrid designs for manufacturing noise-resilient quantum devices.

I. INTRODUCTION

Efficient removal of entropy from a quantum system presents a significant challenge toward the development of new quantum technologies and devices. High purity quantum states that may be quickly initialized and reset are necessary for the application of quantum error correcting codes to suppress and mitigate the effects of noise and errors that naturally occur in quantum information processors, sensors, and communication devices [1]. Additionally, in spectroscopic applications the signal-to-noise ratio increases significantly with state purity, allowing for the detection of small spin ensembles.

A spin ensemble may be naively prepared in a pure state by simply moving to low temperatures, where thermal excitations are not energetic enough to cause significant transitions out of the ground state. However, the required temperatures are often impractical to obtain or require sophisticated and expensive equipment. Additionally, the time required for the spin system to reach thermal equilibrium with the environment – the energy relaxation time, $T_1$ – often becomes very long at low temperatures, limiting the rate at which spin resets and signal averaging may be applied [2].

A variety of techniques for removing entropy from a quantum system are commonly used, including dynamic nuclear polarization (DNP) [2] [3], algorithmic cooling [3], optical pumping [3], laser cooling [6–8], and microwave cooling [9] [10], among others. Recently, it was demonstrated that superconducting qubits may be prepared in an arbitrary pure state through sideband cooling by a high quality factor (high-Q) cavity [11] [12]. We discuss in this work how similar microwave cooling techniques should also be applicable to ensemble spin systems in magnetic resonance, despite the relatively small coupling between the cavity and a single spin. In particular, we present a theoretical model for how a high-Q resonator (cavity) may be used to actively drive an ensemble spin system to a highly pure, non-thermal equilibrium state on a timescale that is significantly shorter than the thermal $T_1$.

II. MATHEMATICAL MODEL

We consider an inductively driven ensemble of non-interacting spin-1/2 particles quantized in a large static magnetic field and magnetically coupled to a high-Q cavity. In the presence of the drive the spins interact with the cavity via coherent radiative processes and may be treated quantum mechanically as a single collective magnetic dipole coupled to the cavity [13]. In analogy to quantum optics, we describe the spin-cavity dynamics as being generated by the Tavis-Cummings (TC) Hamiltonian [14] [15]. Assuming the control field to be on resonance with the Larmor frequency of the spins, the spin-cavity Hamiltonian under the rotating-wave approximation (RWA) is given by

$$
H = H_0 + H_R(t) + H_I,$$

where $a^\dagger (a)$ are the creation (annihilation) operators describing the cavity, $\Omega_R$ is the strength of the drive field (Rabi frequency), $\omega_s$ is the resonant frequency of the cavity, $\omega_0$ is the Larmor resonance frequency of the spins, and $g$ is the coupling strength of the cavity to a single spin in the ensemble in units of $\hbar = 1$. Here we have used the notation that $J_\alpha = \sum_{j=1}^{N_s} a_j^{(\alpha)} / 2$ are the total angular momentum spin operators for an ensemble of $N_s$
spins. The TC Hamiltonian is also known as the Dicke model [16] and has been studied extensively for quantum optics (for a recent review see [17]).

The eigenstates of $H_0$ are the tensor product of photon-number states for the cavity and spin states of collective angular momentum in the $J_z$ direction: $|n\rangle_c |J, m_z\rangle_s$. Here $n = 0, 1, 2, \ldots$, $m_z = -J, -J + 1, \ldots, J - 1, J$, and $J = N_s/2$. The collective excitation number of the joint system is given by $N_{ex} = a^\dagger a + (J_z^+ + J_z^-)$. The interaction term $H_I$ commutes with $N_{ex}$, and hence preserves the total excitation number of the system. It drives transitions between the state $|n\rangle_c |J, m_z\rangle_s$ and states $|n+1\rangle_c |J, m_z-1\rangle_s$ and $|n-1\rangle_c |J, m_z+1\rangle_s$ at a rate of $(n+1)(J(J+1)-m_z(m_z+1))$ and $\sqrt{n}(J(J+1)-m_z(m_z+1))$, respectively.

After moving into an interaction frame defined by $H_I = \omega_c (a^\dagger a + J_z)$, the spin-cavity Hamiltonian is transformed to

$$\tilde{H}^{(1)} = e^{itH_1}H_{sc}e^{-itH_1} - H_1 \quad (2.4)$$

$$H_{0\Omega R}(t) = g(e^{-i\Delta\omega t})^2 + e^{i\Delta\omega t}J_z \quad (2.5)$$

where $\delta\omega = \omega_c - \omega_s$ is the detuning of the drive from the cavity resonance frequency and we have made a second RWA to remove the doubly rotating terms in the interaction frame. If we move into a second interaction frame of $H_2 = \delta\omega a^\dagger a + \Omega_R J_z$, the Hamiltonian transforms to

$$\tilde{H}^{(2)}(t) = H_{0\Omega R}(t) + H_{-\Omega R}(t) + H_{+\Omega R}(t) \quad (2.6)$$

$$H_{0\Omega R}(t) = g(e^{-i\Delta\omega t})^2 + e^{i\Delta\omega t}J_z \quad (2.7)$$

$$H_{-\Omega R}(t) = \frac{i}{2}g(e^{-i(\delta\omega-\Omega_R t)a^\dagger J_z^+})J_z \quad (2.8)$$

$$H_{+\Omega R}(t) = \frac{i}{2}g(e^{-i(\delta\omega+\Omega_R t)a^\dagger J_z^+})J_z \quad (2.9)$$

where $J_z^\pm = J_\pm = \pm iJ_z$ are the spin-ladder operators in the $x$-basis.

In analogy to Hartmann-Hahn in magnetic resonance cross-relaxation experiments [18-20] for $\delta\omega > 0$ we may set the cavity detuning to be close to the Rabi frequency of the drive, so that $\Delta = \delta\omega - \Omega_R/2$ is small compared to $\delta\omega$. Then, by discarding the high frequency terms in Eqn (2.6), the interaction Hamiltonian reduces to the $H_{-\Omega R}$ flip-flop exchange interaction between the cavity and spins in the $x$-basis:

$$H_I(t) = \frac{i}{2}g(e^{-i\Delta t}a^\dagger J_z^+(t) - e^{i\Delta t} a^\dagger J_z^-(t)). \quad (2.10)$$

From here we will drop the $(t)$ superscript and just note that we are working in the $J_z$ eigenbasis.

Isolating the spin-cavity exchange interaction allows efficient energy transfer between the two systems, permitting them to relax to a joint equilibrium state in the interaction frame of the control field. The coherent enhancement of the ensemble spin-cavity coupling – similar to the enhancement of the vacuum Rabi frequency for atomic ensembles, but not restricted to the single-excitation manifold [21] – enhances spin polarization at a rate that may exceed the thermal relaxation rate.

We note that the spin-cavity exchange coupling also exists in the absence of the Rabi drive, and permits cooling of the spin system by matching the resonance frequency of the spin system to the cavity resonance. However, this process is thermally driven, and thus corresponds to incoherent radiative processes that do not display the coherent collective enhancement obtained in the presence of a drive [13]. This Purcell effect in magnetic resonance systems has been previously noted and is normally small enough to be neglected due to the low probability of spontaneous emission in spin systems [22, 23].

To model the cavity-induced cooling of the spin system we use an open quantum system description of the cavity and spin ensemble. The joint spin-cavity dynamics may be modelled using the time-convolutionless (TCL) master equation formalism [24], allowing the derivation of an effective dissipator acting on the spin ensemble alone.

The evolution of the spin-cavity system is described by the Lindblad master equation

$$\frac{d}{dt}\rho(t) = \mathcal{L}_I(t)\rho(t) + \mathcal{D}_c\rho(t) \quad (2.8)$$

where $\mathcal{L}_I(t)$ is the super operator $\mathcal{L}_I(t)\rho = -i[H_I(t),\rho]$ describing evolution under the interaction Hamiltonian [27], and $\mathcal{D}_c$ is a dissipator describing the quality factor of the cavity phenomenologically as a photon amplitude damping channel [25]:

$$\mathcal{D}_c = \frac{\kappa}{2}\left(1 + \pi\right)\mathcal{D}[a] + [\pi\mathcal{D}[a^\dagger]], \quad (2.9)$$

where $\mathcal{D}[A](\rho) = 2A\rho A^\dagger - \{A^\dagger A, \rho\}$, $\pi = |tr[a^\dagger a\rho_{eq}]|$ characterizes the temperature of the bath, and $\kappa$ is the cavity dissipation rate ($\propto 1/Q$). The expectation value of the number operator at equilibrium is related to the temperature, $T$, of the bath by

$$\pi = \left(e^{-\omega_c/k_BT} - 1\right)^{-1} \Leftrightarrow T = \frac{\omega_c}{kB} \left[\ln \left(\frac{1 + \pi}{\pi}\right)\right]^{-1}. \quad (2.10)$$

where $kB$ is the Boltzmann constant.

The reduced dynamics of the spin-ensemble in the interaction frame of the dissipator [28] is given to 2nd order by the TCL master equation [20]:

$$\frac{d}{dt}\rho_s(t) = \int_0^{t-t_0} d\tau tr_c\left[\mathcal{L}_I(t)\rho_s(t) \otimes \rho_{eq}\right], \quad (2.11)$$

where $\rho_s(t) = tr_c[\rho(t)]$ is the reduced state of the spin-ensemble and $\rho_{eq}$ is the equilibrium state of the cavity. Under the condition that $\kappa \gg g\sqrt{N_s}$, the master equation [21] reduces to

$$\frac{d}{dt}\rho_s(t) = \frac{g^2}{4}\int_0^{t-t_0} d\tau e^{-\kappa\tau/2} \left(\cos(\Delta\tau)\mathcal{D}_s\rho_s(t) - \sin(\Delta\tau)\mathcal{L}_s\rho_s(t)\right). \quad (2.12)$$
where
\[ D_s = (1 + \pi) D[J_+] + \pi D[J_-] \] (2.13)
\[ \mathcal{L}_s \rho = -i [H_s, \rho] \] (2.14)
\[ H_s = (1 + \pi) J_+ J_- - \pi J_- J_+ \] (2.15)
are the effective dissipator and Hamiltonian acting on the spin ensemble due to coupling with the cavity.

Under the assumption that \( \kappa \gg g\sqrt{N_s} \), we may take the upper limit of the integral in (2.11) to infinity to obtain the Markovian master equation for the driven spin ensemble:
\[ \frac{d}{dt} \rho_s(t) = \left( \Omega_s \mathcal{L}_s + \frac{\Gamma_s}{2} D_s \right) \rho_s(t) \] (2.16)
where
\[ \Omega_s = -\frac{g^2 \Delta}{\kappa^2 + 4\Delta^2}, \quad \Gamma_s = \frac{g^2 \kappa}{\kappa^2 + 4\Delta^2}. \] (2.17)
Here \( \Omega_s \) is the frequency the effective Hamiltonian, and \( \Gamma_s \) is the effective dissipation rate of the spin-system.

If we consider the evolution of a spin state which is diagonal in the Dicke basis, \( \rho(t) = \sum_m P_m(t) \rho_m \), where \( P_m(t) = \langle J, m | \rho(t) | J, m \rangle \) is the probability of finding the system in the Dicke state \( \rho_m = | J, m \rangle \langle J, m | \) at time \( t \), the master equation (2.16) reduces to a rate equation for the populations of the Dicke states:
\[ \frac{d}{dt} P_m(t) = \Gamma_s \left( A_{m+1} P_{m+1}(t) + B_m P_m(t) \right) + C_{m-1} P_{m-1}(t) \] (2.18)
where
\[ A_m = (1 + \pi) [J(J+1) - m(m-1)] \] (2.19)
\[ C_m = \pi [J(J+1) - m(m+1)] \] (2.20)
\[ B_m = -(A_m + C_m) \] (2.21)

Defining \( \tilde{P}(t) = (P_{-J}(t), \ldots, P_J(t)) \), we obtain a matrix differential equation
\[ \frac{d}{dt} \tilde{P}(t) = \Gamma_s M \tilde{P}(t), \] (2.22)
where \( M \) is the tridiagonal matrix
\[ M = \begin{pmatrix}
B_{-J} & A_{-J+1} & 0 & 0 & 0 & \cdots & 0 \\
C_{-J} & B_{-J+1} & A_{-J+2} & 0 & 0 & \cdots & 0 \\
0 & C_{-J+1} & B_{-J+2} & A_{-J+3} & 0 & \cdots & 0 \\
\vdots & \vdots & \ddots & \ddots & \ddots & \ddots & \vdots \\
0 & \cdots & \cdots & 0 & C_{J-1} & B_J
\end{pmatrix} \] (2.23)

For a given initial state specified by Dicke-state populations \( \tilde{P}(0) \), Eqn (2.22) has the solution
\[ \tilde{P}(t) = \exp(t \Gamma_s M) \tilde{P}(0). \] (2.24)

The equilibrium state of the driven spin-ensemble satisfies \( M \cdot \tilde{P}(\infty) = 0 \), and is given by \( \rho_{eq} = \sum_m \rho_m(\infty) \rho_m \), where
\[ P_m(\infty) = \frac{\pi^{J+m}(1 + \pi)^{J-m}}{(1 + \pi)^{2J+1} - \pi^{2J+1}}. \] (2.25)

It corresponds to the non-thermal state
\[ \rho_{eq} = \frac{1}{Z} \exp \left( -\frac{\omega_s}{k_B T_{eq}} J_x \right), \quad T_{eq} = \frac{\omega_s}{k_B} \ln \left( \frac{1 + \pi}{\pi} \right) \] (2.26)

By comparing with Eqn (2.10), we have that the final effective temperature reached by the spin ensemble in the interaction frame of the Rabi drive is \( T_{eq} = \frac{\omega_s}{k_B} T_c \), where \( T_c \) is the temperature of the cavity. The total spin expectation value for the equilibrium state of the spin-ensemble is
\[ \langle J_x \rangle_{eq} = -J + \frac{\pi}{2} - \frac{(2J + 1)\pi^{2J+1}}{(1 + \pi)^{2J+1} - \pi^{2J+1}}. \] (2.27)

In the limit of \( N_s \gg \pi \), we have that the ground state population at equilibrium is given by \( P_{-J} \approx 1/(1 + \pi) \) and the final expectation value is approximately \( \langle J_x \rangle_{eq} \approx -J + \pi \). Thus, the final spin polarization will be roughly equivalent to the thermal cavity polarization.

We note that if the detuning \( \delta \omega \) were negative, matching \( \Omega_R = 2\delta \omega \) would result in the \( H_+ + \Omega_R \) term being dominant, leading to a master equation (2.16) with the operators \( J_- \) and \( J_+ \) interchanged, the dynamics of which would drive the spin ensemble towards the \( \langle J_x \rangle = J \) state. Thus, the detuning must be larger than the cavity equivalent to the thermal spin-lattice relaxation time, \( T_{1,eff} \).

III. SIMULATIONS

The triadiagonal nature of the rate matrix (2.23) allows Eqn (2.24) to be efficiently simulated for large numbers of spins. For simplicity we will consider the ideal case where the cavity is cooled to its ground state (\( \pi = 0 \)), and the spin-ensemble is taken to be maximally mixed \( (P_m(0) = 1/(2J+1) \) for \( m = -J, \ldots, J \).

The simulated expectation value of \( \langle J_x(t) \rangle \) for ensemble sizes ranging from \( N_s = 10^4 \) to \( N_s = 10^5 \) is shown in Fig. 1, normalized by \( -J \) to obtain a maximum value of 1. At a value of \( -\langle J_x(t) \rangle / J = 1 \) the total angular momentum subspace of the spin ensemble is completely polarized to the \( J_x \) ground eigenstate \( |J_x \rangle \).

The expectation value \( \langle J_x(t) \rangle \) may be fitted to an exponential to derive an effective cooling time-constant, \( T_{1,eff} \), analogous to the thermal spin-lattice relaxation time, \( T_1 \).

A fit to a model given by
\[ -\langle J_x(t) \rangle / J = 1 - \exp \left( -\frac{t}{T_{1,eff}} \right) \] (3.1)
yields the parameters $T_{1,\text{eff}} = \frac{\lambda N\gamma}{\Gamma_s}$ with $\lambda = 2.0406$ and $\gamma = -0.9981$. An approximate expression for the cooling time-constant as a function of the number of spins $N_s$ is then

$$T_{1,\text{eff}}(N_s) \approx \frac{2}{\Gamma_s N_s} = \frac{2(\kappa^2 + 4\Delta^2)}{g^2 N_s}$$ (3.2)

Eqn (3.2) shows that the cooling efficiency is maximized when the Rabi drive strength is matched to the cavity detuning ($\Delta = 0$). In this case the cooling rate and time-constant simplify to $\Gamma_s = \frac{g^4}{\kappa}$ and $T_{1,\text{eff}} = \frac{2\kappa}{g^2 N_s}$, respectively.

If we assume a cavity cooled to its ground state with $Q = 10^4$ ($\kappa/2\pi = 1$ MHz) and a spin-cavity coupling of $g/2\pi = 1$ Hz [27], the range of validity of the Markovian master equation is $N_s \ll \kappa^2/g^2 = 10^{12}$ and an ensemble containing roughly $10^{11}$ electron spins may be completely polarized with an effective $T_1$ of 3.18 $\mu$s. This polarization time is significantly shorter than the thermal $T_1$ for low-temperature spin ensembles, which normally range from seconds to days [2].

In the case where the cavity is thermally occupied, the final spin polarization is roughly equal to the thermal cavity polarization, Eqn (2.26), and the fitted effective cooling constant of the spin ensemble as a function of the thermal cavity occupation number is shown in Fig 2

We find that for cavity temperatures corresponding to $\pi < \sqrt{N_s}$ the effective cooling constant $T_{1,\text{eff}}$ is approximately equal to the zero temperature value. However when $\pi > \sqrt{N_s}$ the effective cooling constant is reduced, and approaches a value of $T_{1,\text{eff}} \approx (2\pi)^{-1}$.

IV. CONCLUSION

The ability to reduce the effective $T_1$ time of a spin ensemble by simply applying a detuned microwave drive provides an important tool for spin-based quantum information processing (for example [28-30] and references therein), where fast gated reset operations provide continual access to pure ancilla states for use in quantum error correction protocols. The ability to efficiently polarize spin samples should also provide more reliable measurements in spectroscopy by permitting fast signal averaging.

Several assumptions were made in the presented theoretical model for cavity cooling of a spin ensemble. Firstly, we have assumed that the spin ensemble has no dipolar coupling between spins. This assumption is valid for magnetically dilute samples. The case of weak dipolar coupling between spins needs to be considered in more detail, but is not expected to fundamentally change the results. Secondly, we have neglected the effects of thermal relaxation of the spin system. As the cooling effect of the cavity on the spin system relies on a coherent spin-cavity information exchange, the relaxation time of the spin system in the frame of the Rabi drive – commonly referred to as $T_{1,\rho}$ – must be significantly longer than the inverse cavity dissipation rate $1/\kappa$. Thirdly, we have assumed that the spin-cavity coupling, $g$, is spatially invariant and that the spin-cavity coupling and Rabi drive are spatially homogeneous across the spin-ensemble. Spatial inhomogeneities would lead to an incoherent distribution of cooling rates over the sample that obscures, but does not prevent, the cooling effect.

Finally, the derivation of the Markovian master equation (2.16) assumes that no correlations between the cavity and spin system accrue during the cooling process, such that there is no back action of the cavity dynamics on the spin system. This condition is enforced when the cavity dissipation rate, $\kappa$, exceeds the rate of coherent spin-cavity exchange in the lowest excitation manifold by at least an order of magnitude – i.e. $\kappa \geq 10g\sqrt{N_s}$ (see supplementary material). In this Markovian limit, the rate at which spin photons are added to the cavity is sig-
significantly less than the rate at which thermal photons are added, meaning the cooling power of the fridge necessary to maintain the thermal cavity temperature is sufficient to dissipate the spin photons without raising the average occupation number of the cavity. From eqn. (3.2) we see that, in principle, the cooling efficiency could be improved by adding more spins to make the cavity to dissipate the spin photons without raising the average occupation number of the cavity. From eqn. (3.2) we see that, in principle, the cooling efficiency could be improved by adding more spins to make $\kappa$ closer to $g\sqrt{N_s}$, but in this regime the cooling power of the fridge is no longer sufficient to prevent back action from the cavity and non-Markovian effects significantly lower the cooling rate (see supplementary material).

As a final comment, the connection between standard techniques in quantum optics and magnetic resonance used in this work has been noted previously (for example, [31,36]), but remains relatively unexplored. The many recent examples of coupling spin ensembles to high-Q resonators, microwave oscillators, and superconducting qubits (for example [27,37–43] and references therein) motivates more investigation into the relationship between the two fields.

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V. SUPPLEMENTARY MATERIAL

In this supplementary material we provide additional details on the effective cooling rate for a thermally excited cavity and the validity of the Markovian assumption used to derive the cooling rates derived in the main text.

A. Thermally excited cavity

In the Markov master equation simulations used to derive the effective cooling constant, $T_{1,\text{eff}}$, eqn. (3.2), as a function of ensemble size, $N_s$, we assumed that the cavity was initially cooled to its ground state. If we fit our simulations for a non-zero average cavity occupation number $n = \left(\frac{\omega_c}{\hbar B T_c} - 1\right)^{-1}$, where $\omega_c$ and $T_c$ are the resonant frequency and equilibrium temperature of the cavity, the effective cooling time constant of the spin ensemble was found to obey

$$T_{1,\text{eff}} \approx \begin{cases} \frac{2}{N_s} & \text{for } n < \sqrt{N_s} \\ \frac{2}{N_s} & \text{for } n \gtrsim N_s \end{cases}$$

(5.1)

This effect appears to originate from the fact that the final spin system polarization will be equal to the cavity polarization. Thus, cooling to a spin temperature that is not fully polarized requires removing fewer photons from the spin system. Given that the spin dissipation rate is independent of the cavity temperature, it takes less time to drive the spins to a state that is not fully polarized. An example of the simulated normalized spin expectation value $-\langle J_x \rangle/J$ as a function of temperature is shown in Fig 3. Here we are considering a cavity with resonant frequency of $\omega_c/2\pi = 10$ GHz for the Tavis-Cummings Hamiltonian in Eqn (2.3).

![Normalized spin expectation value](image)

FIG. 3: Normalized spin expectation value $-\langle J_x \rangle/J$ of the spin ensemble as a function of time for various equilibrium temperatures of the cavity. We consider the case of $N_s = 10, 100, 1000$ and $10,000$ spins in the ensemble, and a cavity with resonant frequency $\omega_c = 10$ GHz.

B. Validity of Markov Approximation

The validity of the cooling rates derived by the Markovian master equation, eqn. (2.16), depends on the validity of the Markov approximation used to derive the master equation. As stated in the main text, enforcing the condition that the cavity dissipation rate, $\kappa$, exceeds the rate of coherent exchange between the spins and cavity implies that
there will be no back action of the cavity dynamics on the spin system. We stated this condition as \( \kappa \geq 10g\sqrt{N} \), such that any photons transferred to the cavity are immediately dissipated in the cavity before they return to the spin system.

\[
\begin{align*}
| -J_x + 3, 0 \rangle & \xrightarrow{g\sqrt{3(N-2)}} | -J_x + 2, 1 \rangle \xrightarrow{\kappa \sqrt{2}} | -J_x + 1, 2 \rangle \xrightarrow{g\sqrt{N}} | -J_x, 3 \rangle \\
| -J_x + 2, 0 \rangle & \xrightarrow{g\sqrt{2(N-1)}} | -J_x + 1, 1 \rangle \xrightarrow{\kappa \sqrt{3}} | -J_x, 2 \rangle \\
| -J_x + 1, 0 \rangle & \xrightarrow{g\sqrt{N}} | -J_x, 1 \rangle \\
| -J_x, 0 \rangle & \xrightarrow{\kappa}
\end{align*}
\]

**FIG. 4:** Energy level diagram of the joint spin-cavity system with coherent transitions denoted by a solid line and cavity dissipation rates denoted by a curved line. States are labelled as \( | -J_x + m, n \rangle \), where \( m \) is the number of spin excitations and \( n \) is the number of cavity excitations. For the cooling dynamics to appear Markovian states of high cavity excitation number should not be significantly populated on a coarse-grained time scale.

More concretely, from the spin-cavity energy level diagram shown in fig. 4 the rate of transfer between states \( | -J_x + m, n \rangle \) and \( | -J_x + m - 1, n + 1 \rangle \) is given by \( g\sqrt{(m+1)(N-m)} \), where \( N \) is the number of spins in the ensemble. At the same time, the cavity dissipator of strength \( \kappa \sqrt{n} \) is acting to drive the spin-cavity system to the state \( | -J_x + m - 1, n \rangle \). To satisfy the Markov condition, we require the cooling dynamics to always drive the spin-cavity system toward states of low excitation number (bottom left of diagram), without significantly populating states of high excitation number (top right of diagram). This will occur if the maximum rates for coherent transfer and cavity dissipation obey the following relationship:

\[
g\sqrt{(\frac{N}{2} + 1)(N - \frac{N}{2})} \ll \kappa \sqrt{\frac{N}{2}}. \tag{5.2}
\]

Assuming that \( N \gg 1 \), such that \( \frac{N}{2} + 1 \approx \frac{N}{2} \), this condition simplifies to

\[
\kappa \gg 0.707g\sqrt{N}. \tag{5.3}
\]

As shown in fig. 5 when \( \kappa = 0.5g\sqrt{N} \) a full non-Markovian simulation of the cooling procedure yields dynamics that are much richer than predicted by the Markovian model used in the main text. In particular, coherent transfer of spin photons deposited in the cavity back to the spin system are seen as oscillations in the expectation value of \( J_x \). These memory effects reduce the cooling efficiency such that the cooling rate is initially very fast when the cavity occupation is low, then slows down significantly as higher excitations of the cavity are transferred back to the spin system. As \( \kappa \) becomes larger than \( \sqrt{2} \), the oscillations are damped out, but the Markovian master equation still does not fully agree with the full non-Markovian simulation. When \( \kappa = 10g\sqrt{N} \), the oscillations are critically damped and the Markovian master equation captures the full cooling dynamics. Thus, if the cavity dissipation rate, \( \kappa \), exceeds the rate of coherent spin-cavity exchange in the single excitation manifold by at least an order of magnitude – i.e. \( \kappa \geq 10g\sqrt{N} \) – then the Markovian master equation is valid.
FIG. 5: Comparison of the cooling dynamics by the Markovian master equation (pink dotted curve) and a full spin-ensemble cavity simulation (blue dashed curve). The normalized expectation value of $-\langle J_x(t) \rangle / J$ is plotted for $N = 5$ spins with $\kappa = 0.5, 1, 5, 10 g \sqrt{N_s}$ and cavity temperature $T = 0K$. When $\kappa \geq 10 g \sqrt{N}$ the Markovian master equation calculation agrees very well with the full simulation. Also, as predicted by eqn. (3.2), the cooling rate increases for larger $\kappa$, until the point where non-Markovian effects take over.