Cavity Cooling of an Ensemble Spin System

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We describe how sideband cooling techniques 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 the coupled angular momentum subspaces of a spin ensemble containing roughly 10^{11} electron spins may be polarized in a time many orders of magnitude shorter than the typical thermal relaxation time. The described techniques should permit 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 recently begun to be explored in further detail due to the development of hybrid designs for manufacturing noise-resilient quantum devices.

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Efficient removal of entropy from a quantum system is essential for the development of robust 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]. For 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 fluctuations are not energetic enough to cause significant excitation 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 [2,3], algorithmic cooling [4], optical pumping [5], laser cooling [6–8], and microwave cooling [9–11], 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 [12,13]. 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 each coupled angular momentum subspaces of an ensemble spin system to a state with purity equal to that of the cavity on a time scale significantly shorter than the thermal T_1 of the spins. Our model is motivated by recent studies that describe magnetic resonance in terms of quantum optics (for example, [14–19]).

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 error correcting spin-based quantum information processors (see, for example, [20–22] and references therein) and should also find applications in spectroscopy by permitting faster signal averaging. These techniques may also find use in enhancing quantum memories for microwave photons based on coupling spin ensembles to superconducting devices (see, for example, [23–27] and references therein).

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 [28]. In analogy to quantum optics, we describe the spin-cavity dynamics as being generated by the Tavis-Cummings (TC) Hamiltonian [29,30]. Assuming a linearly oscillating control field resonant with the Larmor frequency of the spins, the spin-cavity Hamiltonian is given by $H = H_0 + H_R(t) + H_I$, with

$$H_0 = \omega_c a^\dagger a + \omega_s J_z, \tag{1}$$

$$H_R(t) = 2\Omega_R \cos(\omega_s t) J_x, \tag{2}$$

$$H_I = 2g(a^\dagger + a)J_x, \tag{3}$$

where $a^\dagger(a)$ are the creation (annihilation) operators describing the cavity, Ω_R is the strength of the drive field (Rabi frequency), ω_c is the resonant frequency of the cavity, ω_s 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 $\mu_0 J_\alpha \equiv \sum_{j=1}^{N_s} \sigma_\alpha^{(j)}/2$ are the total angular momentum spin operators for an ensemble of N_s spins.

The state-space V of a spin ensemble of N_s identical spins may be written as the direct sum of coupled angular momentum subspaces $V = \bigoplus_{J=j_0}^{N_s/2} V_J^{\oplus n_J}$ where $j_0 = 0(1/2)$ if N_s is even (odd). V_J is the state space of a spin-J particle with dimension $d_J = 2J+1$, and there are n_J degenerate subspaces with the same total spin J [31]. Since the TC Hamiltonian has a global SU(2) symmetry it will not couple between subspaces in this representation. The largest subspace in this representation is called the Dicke subspace and consists of all totally symmetric states of the spin ensemble. It corresponds to a system with total angular momentum $J = N_s/2$. The TC Hamiltonian restricted to the Dicke subspace is known as the Dicke model [32] and has been studied extensively for quantum optics (for a recent review, see [33]).

The eigenstates of H_0 are the tensor product of photon-number states for the cavity and spin states of collective angular momentum of each total-spin subspace 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 indexes the coupled angular momentum subspace V_J. The collective excitation number of the joint system for each subspace is given by $N_{\mathrm{ex}} = a^\dagger a + (J_z + J)$. 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 $\sqrt{n+1}\sqrt{J(J+1) - m_z(m_z-1)}$ and $\sqrt{n}\sqrt{J(J+1) - m_z(m_z+1)}$, respectively.

After moving into a rotating frame defined by $H_1 = \omega_s(a^\dagger a + J_z)$, the spin-cavity Hamiltonian is transformed to

$$\tilde{H}^{(1)} = \delta\omega a^\dagger a + \Omega_R J_x + g(a^\dagger J_- + a J_+), \tag{4}$$

where $\delta\omega = \omega_c - \omega_s$ is the detuning of the drive from the cavity resonance frequency and we have made the standard rotating wave approximation (RWA) to remove any time-dependent terms in the Hamiltonian [2].

If we now move into the interaction frame of $H_2 = \delta\omega a^\dagger a + \Omega_R J_x$, the Hamiltonian transforms to

$$\begin{aligned}
\tilde{H}^{(2)}(t) &= H_{0\Omega_R}(t) + H_{-\Omega_R}(t) + H_{+\Omega_R}(t),\\
H_{0\Omega_R}(t) &= g(e^{-i\delta\omega t} a + e^{i\delta\omega t} a^\dagger) J_x,\\
H_{-\Omega_R}(t) &= \frac{ig}{2}(e^{-i(\delta\omega - \Omega_R)t} a J_+^{(x)} - e^{i(\delta\omega-\Omega_R)t} a^\dagger J_-^{(x)}),\\
H_{+\Omega_R}(t) &= \frac{ig}{2}(e^{-i(\delta\omega + \Omega_R)t} a J_-^{(x)} - e^{i(\delta\omega+\Omega_R)t} a^\dagger J_+^{(x)}),
\end{aligned} \tag{5}$$

where $J_\pm^{(x)} \equiv J_y \pm i J_z$ are the spin-ladder operators in the x basis.

In analogy to Hartmann-Hahn matching in magnetic resonance cross-relaxation experiments [34–36] for $\delta\omega > 0$ we may set the cavity detuning to be close to the Rabi frequency of the drive, so that $\Delta = \delta\omega - \Omega_R$ is small compared to $\delta\omega$. By making a second RWA in the interaction frame of H_2, the interaction Hamiltonian reduces to the $H_{-\Omega_R}$ flip-flop exchange interaction between the cavity and spins in the x basis:

$$H_I(t) = \frac{ig}{2}(e^{-i\Delta t} a J_+^{(x)} - e^{i\Delta t} a^\dagger J_-^{(x)}). \tag{6}$$

This RWA is valid in the regime where the detuning and Rabi drive strength are large compared to the time scale, t_c, of interest ($\delta\omega$, $\Omega_R \gg 1/t_c$, [37]). From here we will drop the (x) superscript and just note that we are working in the J_x 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 [38]—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 theoretically 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 a set of incoherent radiative processes that may not be described by a single Hamiltonian [28]. This Purcell effect in magnetic resonance systems has been previously noted and is normally small enough to be neglected [39,40].

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 modeled using the time-convolutionless (TCL) master equation formalism [41], allowing the derivation of an effective dissipator acting on the spin ensemble alone. Since the spin-subspaces V_J are not coupled by the TC Hamiltonian, the following derivation holds for all values of J in the state-space factorization.

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

$$\frac{d}{dt}\rho(t) = \mathcal{L}_I(t)\rho(t) + \mathcal{D}_c\rho(t), \tag{7}$$

where $\mathcal{L}_I$ is the superoperator $\mathcal{L}_I(t)\rho = -i[H_I(t), \rho]$ describing evolution under the interaction Hamiltonian (6), and $\mathcal{D}_c$ is a dissipator describing the quality factor of the cavity phenomenologically as a photon amplitude damping channel [42]:

$$\mathcal{D}_c = \frac{\kappa}{2}((1+\bar{n})\mathcal{D}[a] + \bar{n}\mathcal{D}[a^\dagger]), \tag{8}$$

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

$$\bar{n} = (e^{\omega_c/k_B T} - 1)^{-1} \Leftrightarrow T_c = \frac{\omega_c}{k_B}\left[\ln\left(\frac{1+\bar{n}}{\bar{n}}\right)\right]^{-1}, \tag{9}$$

where k_B is the Boltzmann constant.

The reduced dynamics of the spin-ensemble in the interaction frame of the dissipator (8) is given to second order by the TCL master equation [43]:

$$\frac{d}{dt}\rho_s(t) = \int_0^{t-t_0} d\tau\, \mathrm{tr}_c[\mathcal{L}_I(t)e^{\tau\mathcal{D}_c}\mathcal{L}_I(t-\tau)\rho_s(t)\otimes\rho_{\mathrm{eq}}], \tag{10}$$

where $\rho_s(t) = \mathrm{tr}_c[\rho(t)]$ is the reduced state of the spin-ensemble and ρ_{eq} is the equilibrium state of the cavity. Under the condition that $\kappa \gg g\sqrt{N_s}$, the master equation (10) reduces to

$$\frac{d}{dt}\rho_s(t) = \frac{g^2}{4}\int_0^{t-t_0} d\tau e^{-\kappa\tau/2} \times (\cos(\Delta\tau)\mathcal{D}_s\rho_s(t) - \sin(\Delta\tau)\mathcal{L}_s\rho(t)), \tag{11}$$

where

$$\mathcal{D}_s = (1+\bar{n})\mathcal{D}[J_-] + \bar{n}\mathcal{D}[J_+], \tag{12}$$

$$\mathcal{L}_s\rho = -i[H_s, \rho], \tag{13}$$

$$H_s = (1+\bar{n})J_+J_- - \bar{n}J_-J_+ \tag{14}$$

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 (10) to infinity to obtain the Markovian master equation for the driven spin ensemble:

$$\frac{d}{dt}\rho_s(t) = \left(\Omega_s\mathcal{L}_s + \frac{\Gamma_s}{2}\mathcal{D}_s\right)\rho_s(t), \tag{15}$$

where

$$\Omega_s = -\frac{g^2\Delta}{\kappa^2 + 4\Delta^2}, \qquad \Gamma_s = \frac{g^2\kappa}{\kappa^2 + 4\Delta^2}. \tag{16}$$

Here Ω_s is the frequency of the effective Hamiltonian and Γ_s is the effective dissipation rate of the spin-system [37].

We consider the evolution of a spin state that is diagonal in the coupled angular momentum basis, $\rho(t) = \sum_J \sum_{m=-J}^{J} P_{J,m}(t)\rho_{J,m}$. Here the sum over J is summing over subspaces V_J, and $P_{J,m}(t) = \langle J, m|\,\rho(t)\,|J, m\rangle$ is the probability of finding the system in the state $\rho_{J,m} = |J, m\rangle\langle J, m|$ at time t. In this case the master equation (15) reduces to a rate equation for the state populations:

$$\frac{d}{dt}P_{J,m}(t) = \Gamma_s(A_{J,m+1}P_{J,m+1}(t) + B_{J,m}P_{J,m}(t) + C_{m_J-1}P_{m_J-1}(t)), \tag{17}$$

where

$$A_{J,m} = (1+\bar{n})[J(J+1) - m(m_J - 1)], \tag{18}$$

$$C_{J,m} = \bar{n}[J(J+1) - m(m_J + 1)], \tag{19}$$

$$B_{J,m} = -(A_m + C_m). \tag{20}$$

Defining $\vec{P}_J(t) = (P_{J,-J}(t), \ldots, P_{J,J}(t))$, we obtain the following matrix differential equation for each subspace V_J:

$$\frac{d}{dt}\vec{P}_J(t) = \Gamma_s M_J \vec{P}_J(t), \tag{21}$$

where M_J is the tridiagonal matrix

$$M_J = \begin{pmatrix} B_{J,-J} & A_{J,-J+1} & 0 & 0 & 0 & \cdots & 0 \\ C_{J,-J} & B_{J,-J+1} & A_{-J+2} & 0 & 0 & \cdots & 0 \\ 0 & C_{J,-J+1} & B_{J,-J+2} & A_{J,-J+3} & 0 & \cdots & 0 \\ \vdots & & & \ddots & & & \vdots \\ 0 & & \cdots & & 0 & C_{J,J-1} & B_{J,J} \end{pmatrix}. \tag{22}$$

For a given state specified by initial populations $\vec{P}_J(0)$, Eq. (21) has the solution

$$\vec{P}_J(t) = \exp\,(t\Gamma_s M_J)\vec{P}_J(0). \tag{23}$$

The equilibrium state of each subspace V_J of the driven spin-ensemble satisfies $M_J \cdot \vec{P}_J(\infty) = 0$, and is given by $\rho_{J,\mathrm{eq}} = \sum_{m=-J}^{J} P_{J,m}(\infty)\rho_{J,m}$, where

$$P_{J,m}(\infty) = \frac{\bar{n}^{J+m}(1+\bar{n})^{J-m}}{(1+\bar{n})^{2J+1} - \bar{n}^{2J+1}}. \tag{24}$$

The total spin expectation value for the equilibrium state of each subspace of the spin ensemble is

$$\langle J_x\rangle_{\mathrm{eq}} = -J + \bar{n} - \frac{(2J+1)\bar{n}^{2J+1}}{(1+\bar{n})^{2J+1} - \bar{n}^{2J+1}}. \tag{25}$$

If we consider the totally symmetric Dicke subspace in the limit of $N_s \gg \bar{n}$, we have that the ground state population at equilibrium is given by $P_{N_s/2,-N_s/2} \approx 1/(1+\bar{n})$ and the final expectation value is approximately $\langle J_x \rangle_{\text{eq}} \approx -N_s/2 + \bar{n}$. Thus, the final spin polarization in the Dicke subspace will be roughly equivalent to the thermal cavity polarization.

We note that if the detuning $\delta\omega$ were negative, matching $\Omega_R = \delta\omega$ would result in the $H_{+\Omega_R}$ term being dominant, leading to a master equation (15) 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 linewidth to prevent competition between the $H_{-\Omega_R}$ and $H_{+\Omega_R}$ terms, which would drive the spin system to a high entropy thermally mixed state.

The tridiagonal nature of the rate matrix (22) allows Eq. (23) to be efficiently simulated for large numbers of spins. For simplicity we will consider the cooling of the Dicke subspace in the ideal case where the cavity is cooled to its ground state ($\bar{n} = 0$), and the spin ensemble is taken to be maximally mixed in the basis of the spin-J subspace [$P_m(0) = 1/(2J+1)$ for $m = -J, \ldots, J$].

The simulated expectation value of $\langle J_x(t) \rangle$ for the Dicke subspace with total spin $N_s/2$ ranging from $N_s = 10^3$ to 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 Dicke subspace of the spin ensemble is completely polarized to the J_x ground eigenstate $|J, -J\rangle$.

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

$$-\frac{\langle J_x(t) \rangle}{J} = 1 - \exp\left(-\frac{t}{T_{1,\text{eff}}}\right) \tag{26}$$

yields the parameters $T_{1,\text{eff}} = \lambda(2J)^\gamma / \Gamma_s$ with $\lambda = 2.0406$ and $\gamma = -0.9981$. An approximate expression for the cooling time constant for the spin subspace V_J as a function of J is then

$$T_{1,\text{eff}}(J) \approx \frac{1}{\Gamma_s J} = \frac{\kappa^2 + 4\Delta^2}{g^2 \kappa J}, \tag{27}$$

showing 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 $\Gamma_s = g^2/\kappa$ and $T_{1,\text{eff}} = \kappa / g^2 J$, respectively.

In the case where the cavity is thermally occupied, the final spin polarization is roughly equal to the thermal cavity polarization, and for cavity temperatures corresponding to $\bar{n} < \sqrt{2J}$ the effective cooling constant $T_{1,\text{eff}}$ is approximately equal to the zero temperature value [37].

To achieve this result experimentally, one must choose parameters that adhere to the two RWA's used to isolate the spin-cavity exchange term of Eq. (6). Under the condition that $\delta\omega \approx \Omega_R$, this requires that $g\sqrt{N_s} \ll \kappa \ll \Omega_R$, $\delta\omega \ll \omega_c, \omega_s$ [37]. For example, assuming an implementation using x-band pulsed electron spin resonance ($\omega_c/2\pi \approx \omega_s/2\pi = 10$ GHz), with samples that typically contain from roughly $N_s = 10^6$ spins to $N_s = 10^{17}$ spins [44,45], experimentally reasonable values are $\Omega_R/2\pi = 100$ MHz, $Q = 10^4$ ($\kappa/2\pi = 1$ MHz) [46–48], and $g/2\pi = 1$ Hz [47].

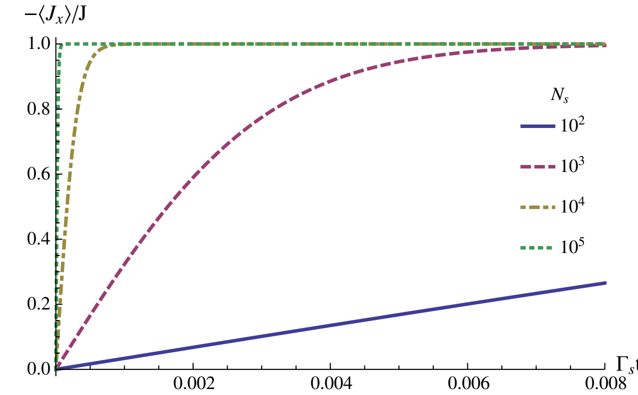

FIG. 1 (color online). Simulated evolution of the normalized expectation value of $-\langle J_x(t) \rangle / J$ for the Dicke subspace of a cavity-cooled spin ensemble. The time axis is scaled by the effective dissipation rate, Γ_s, for the spin-ensemble given in Eq. (16).

For these parameters, the range of validity of the Markovian master equation is $N_s \ll \kappa^2/g^2 = 10^{12}$ and the Dicke subspace of an ensemble containing roughly 10^{11} electron spins may be polarized with an effective T_1 of 3.18 μs. This polarization time is significantly shorter than the thermal T_1 for low-temperature spin ensembles, which normally range from seconds to days [2].

Several assumptions were made in the presented theoretical model for cavity cooling of a spin ensemble. First, we have assumed that the spin ensemble is magnetically dilute such that no coupling exists between spins. Any spin-spin interaction that breaks the global SU(2) symmetry of the TC Hamiltonian will connect the spin-J subspaces in the coupled angular momentum decomposition of the state space. Such an interaction may be used as an additional resource that should permit complete polarization of the full ensemble Hilbert space. Second, 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 $1/\kappa$. Third, we have assumed that the spin-cavity coupling and Rabi drive are spatially homogeneous across the spin ensemble. Inhomogeneities may be compensated for by numerically optimizing a control pulse that implements an effective spin-locking Rabi drive of constant strength over a range of spin-cavity coupling and control field amplitudes [49].

Finally, the derivation of the Markovian master equation (15) 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, κ, exceeds the rate of coherent spin-cavity exchange in the lowest excitation manifold by at least an order of magnitude —i.e., $\kappa \geq 10g\sqrt{N_s}$ [37]. In this Markovian limit, the rate at which spin photons are added to the cavity is 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 Eq. (27) we see that, in principle, the cooling efficiency could be improved by adding more spins to make κ 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 strong non-Markovian effects significantly lower the cooling rate.

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