Chapter 2

Description of the nanoscale spin-resonator system

1 Average Hamiltonian

We begin by obtaining an interaction-frame Hamiltonian for a system consisting of a torsional mechanical resonator coupled to a collection of isochronous spins that interact only with the resonator. The field at the spins is the sum of a static applied field and the field of the magnetic mechanical resonator. Let θ be the resonator's angular coordinate, with equilibrium position corresponding to $\theta = 0$, and let $\mathbf{B}_a$ and $\mathbf{B}_h(\theta)$ represent the applied field and the field of the resonator, respectively. We define

$$\mathbf{B}(\theta) = \mathbf{B}_a + \mathbf{B}_h(\theta)$$

to be the total field at the spins, and simplify notation by letting $\mathbf{B}$, $\mathbf{B}_h$, and $d\mathbf{B}/d\theta$ stand for $\mathbf{B}(0)$, $\mathbf{B}_h(0)$, and $\{d\mathbf{B}/d\theta\}(0)$, respectively. The positive z-axis is chosen to lie in the direction of $\mathbf{B}$, and the x-axis is chosen so that $d\mathbf{B}/d\theta$ lies in the xz-plane; i.e., $d\mathbf{B}/d\theta$ has nonzero components along only the x-axis and the z-axis. The Hamiltonian is written in units of rad/s as

$$H = -\gamma \mathbf{I} \cdot \mathbf{B}(\theta) + H_{\text{osc}}, \tag{2.1}$$

where H_{osc} is the Hamiltonian for the harmonic oscillator, γ is the gyromagnetic ratio, and $\mathbf{I}$ is the spin operator. In analyzing the Hamiltonian, we approximate $\mathbf{B}(\theta)$ by its first-order Taylor series:

$$\mathbf{B}(\theta) \approx \mathbf{B} + \left(\frac{d\mathbf{B}}{d\theta}\right)\theta. \tag{2.2}$$

Our first-order approximation to $\mathbf{B}(\theta)$ is completely characterized by the three constants B_z, $dB_x/d\theta$, and $dB_z/d\theta$. For oscillators which have

$$B_z(\theta) = B_z(-\theta),$$

the derivative $dB_z/d\theta$ is zero at $\theta = 0$, and we limit the discussion to oscillators having this property. The first-order approximation to $\mathbf{B}(\theta)$ is

$$B_x(\theta) = \frac{dB_x}{d\theta}\theta, \tag{2.3}$$

$$B_y(\theta) = 0, \tag{2.4}$$

and

$$B_z(\theta) = B_z. \tag{2.5}$$

Equations 2.3 through 2.5 allow us to express the Hamiltonian as

$$H = \left(-\gamma\frac{dB_x}{d\theta}\right) I_x\theta + H_0,$$

where H_0 would be the Hamiltonian for a system in which the spins and oscillator are uncoupled:

$$H_0 = \omega_0 I_z + H_{\text{osc}} \tag{2.6}$$

$$\omega_0 \equiv -\gamma B_z. \tag{2.7}$$

Making the substitutions

$$\theta = \frac{1}{\sqrt{2}\beta}\left(a + a^\dagger\right),$$
$$I_x = \frac{1}{2}\left(I_+ + I_-\right)$$

yields

$$H = H_0 + g\left(I_+ a^\dagger + I_- a + I_+ a + I_- a^\dagger\right). \tag{2.8}$$

In (2.8), I_+, I_- are the respective raising and lowering operators for the spins, $a^\dagger$ and a are the respective raising and lowering operators for the mechanical oscillator, and the constants β and g are given by

$$\beta \equiv \sqrt{\frac{I_h \omega_h}{\hbar}},$$
$$g \equiv \frac{-\gamma}{2\sqrt{2}\beta}\frac{dB_x}{d\theta}, \tag{2.9}$$

where I_h is oscillator's moment of inertia and ω_h is the mechanical frequency.

Using the operator $\exp\left(-iH_0 t/\hbar\right)$ to switch to the interaction frame and applying the identities

$$e^{i\omega_h t a^\dagger a} a e^{-i\omega_h t a^\dagger a} = a e^{-i\omega_h t},$$
$$e^{i\omega_0 t I_z} I_+ e^{-i\omega_0 t I_z} = I_+ e^{i\omega_0 t}$$

transforms the Hamiltonian to

$$\tilde{H} = g\left[e^{i(\omega_h+\omega_0)t} I_+ a^\dagger + e^{-i(\omega_h+\omega_0)t} I_- a + e^{-i(\omega_h-\omega_0)t} I_+ a + e^{i(\omega_h-\omega_0)t} I_- a^\dagger\right].$$

If the gyromagnetic ratio is positive, resonance between the Larmor and mechanical frequencies corresponds to the condition

$$\omega_h = -\omega_0, \tag{2.10}$$

since $\omega_h > 0$ and $\omega_0 < 0$. At resonance, the average Hamiltonian, which we denote by V, is

$$V = g\left(I_+ a^\dagger + I_- a\right). \tag{2.11}$$

This Hamiltonian is often referred to as the Jaynes-Cummings Hamiltonian. It has been studied extensively in quantum optics, since it governs the interaction between a two-level atom and a mode of the electromagnetic field [4]. In the current context, it can be interpreted as governing an interaction in which one rotating component of the resonator's transverse field is resonant with the Larmor frequency and induces transitions between spin eigenstates. This resonant transverse field can be considered roughly analogous to the applied transverse field which rotates spins during an NMR pulse.

The use of the first-order expression (2.2) as an approximation to $\mathbf{B}(\theta)$ yields a model in which B_z does not vary as the mechanical resonator moves. The model excludes physical effects caused by fluctuations in B_z associated with the mechanical motion, such as the resonator's contribution to transverse spin relaxation by "secular broadening" (i.e., transverse relaxation due to fluctuations in the longitudinal field). To include such effects in our analysis, we expand the field to second order in θ, limiting the discussion to resonators for which the properties

$$B_x(\theta) = -B_x(-\theta)$$
$$B_y(\theta) \equiv 0$$

imply that

$$\frac{d^2 B_x}{d\theta^2} = \frac{d^2 B_y}{d\theta^2} = 0$$

at $\theta = 0$. The average Hamiltonian in the interaction frame is then

$$V' = g\left(I_+ a^\dagger + I_- a\right) + f I_z\left(a^\dagger a - n_{\mathrm{th}}\right), \tag{2.12}$$

where

$$f = -\gamma \frac{d^2 B_z}{d\theta^2} \frac{\hbar}{2 I_h \omega_h}$$

and $n_{\rm th}$ is the thermal number of quanta in the resonator. In deriving (2.12), we have used the interaction frame defined by $H_0 = \omega_0 I_z + \omega_h \left(a^\dagger a + 1/2 \right)$, where

$$\omega_0 = -\gamma B_z + f \left(n_{\rm th} + 1/2 \right). \tag{2.13}$$

The terms proportional to f in (2.12) and (2.13) arise because the value of B_z depends on the number of quanta in the resonator. In the case where

$$\frac{d^2 B_z}{d\theta^2} < 0,$$

for instance, the value of B_z is greatest when the resonator is in equilibrium position, and motion away from equilibrium decreases B_z. In the interaction-frame Hamiltonian, the resonator's contribution to B_z consists of terms which vary at frequency $\pm 2\omega_h$ as well as a time-independent term that depends on the the number of quanta $a^\dagger a$ in the resonator. Fluctuations in $a^\dagger a$ away from the thermal value $n_{\rm th}$ correspond to a fluctuating value of B_z at the spins. In the presence of this fluctuating field, the mean value of the Larmor frequency is given by (2.13). Using this value of ω_0 in defining H_0 ensures that $\langle V' \rangle = 0$, where the average is taken over the thermal reservoir that damps the resonator.

2 Equations of motion for spin operators

2.1 Reduced master equation for the spins

In the case where the sample consists of spins 1/2, evolution under the Hamiltonian (2.11) can be characterized using results available in the quantum optics literature. When a single atom interacts with an undamped electromagnetic mode of a resonant cavity, an initial state function that has n quanta in the mode and that has the

spin in its excited state evolves in time by periodically exchanging a single quantum between spin and mode at frequency $2g\sqrt{n+1}$ [4]. When N atoms are present in the undamped cavity, with all atoms initially excited, the system evolves "quasi-periodically," as the excitation initially present in the spins is transferred between the atoms and the resonant mode with a frequency of order [5]

$$g \times \text{ field amplitude } \approx g\sqrt{\langle a^\dagger a \rangle}, \tag{2.14}$$

where $\langle a^\dagger a \rangle$ is the mean number of quanta in the cavity mode, including thermal quanta and quanta donated by the atoms to the cavity mode. For a cavity at zero Kelvins, this frequency is of order $g\sqrt{N}$, where N is the number of atoms in the cavity [5]. If the cavity mode is weakly damped, oscillations in the excitation of the atoms gradually decay as quanta are dissipated from the mode. Increasing the strength of the damping eventually suppresses the oscillations, and the atoms decay monotonically when the rate constant for dissipation of quanta is large compared to the frequency at which quanta would cycle between atoms and the resonator in the absence of damping [5]. Since the rate constant for dissipation of quanta can be written as $2/\tau_h$, where τ_h is the decay time of the mechanical resonator's position coordinate (or "ringdown time"), the condition that guarantees oscillations will be suppressed is

$$g\sqrt{\langle a^\dagger a \rangle} \ll \frac{2}{\tau_h}. \tag{2.15}$$

In this regime, the evolution of the atomic system can be described by a reduced master equation which does not explicitly include the resonator's degrees of freedom [5], and the resonator can be considered a reservoir which damps the atomic system. For a resonator at zero Kelvins, the condition (2.15) which allows the use of a reduced master equation is written more explicitly as [6]

$$g\sqrt{N} \ll \frac{2}{\tau_h}. \tag{2.16}$$

These results can be carried over directly to a system consisting of spins 1/2 which

evolve under a spin Hamiltonian H_s while interacting with a damped mechanical resonator. The evolution of the spin system is governed by the master equation [5]

$$\frac{d}{dt}\rho_s = -i\left[H_s, \rho_s\right] - \frac{1}{2}R_0\left(n_{\rm th}+1\right)\left[I_-I_+, \rho_s\right]_+ + R_0\left(n_{\rm th}+1\right)I_+\rho_s I_- \qquad (2.17)$$
$$- \frac{1}{2}R_0 n_{\rm th}\left[I_+I_-, \rho_s\right]_+ + R_0 n_{\rm th} I_-\rho_s I_+,$$

where

$$R_0 = 2g^2\tau_h.$$

The anticommutator $\left[\cdot,\cdot\right]_+$ is defined by

$$\left[A, B\right]_+ = AB + BA.$$

Both H_s and the spin density matrix ρ_s are expressed in the interaction frame in which the Hamiltonian H_0 of equation (2.6) has been eliminated, and the resonator field is assumed to be identical at all spins. Note that spin-lattice interactions are not included, since (2.17) is derived by considering an undamped system of atoms which interact with a damped electromagnetic mode. At very low temperatures, where the spin-lattice relaxation is "frozen out," equation (2.17) can be used to investigate the question of whether spin-resonator relaxation governed by the Hamiltonian of equation (2.11) can efficiently cool the spins toward thermal equilibrium with the resonator.

Note that (2.17) was derived by adding the term $-i\left[H_s, \rho_s\right]$, which governs unitary evolution under H_s, to a relaxation superoperator derived under the assumption that $H_s = 0$. The discussion in this thesis is limited to the regime in which this step is valid. To characterize this regime, note first that resonator-induced relaxation depends on weak correlations which develop between spins and resonator. The resonator "remembers" an interaction with the spins for a time period of order τ_h. In a simple visualization of the relaxation, we can consider that a spin-resonator correlation survives during a period of order τ_h and is then annihilated. The new spin-resonator correlation which then develops is determined by the instantaneous

state of the spins. Spin relaxation can thus be visualized as occurring during time periods of order τ_h, with the relaxation during a given time period depending only on the state of the spins at the beginning of that period. If the time scale of the spin evolution associated with H_s is long compared to τ_h, then there is little error in adopting the point of view that the spins are at every instant relaxing just as they would if H_s were absent, while H_s slowly modulates the spin state. In this regime, we can obtain a master equation by adding the unitary term $-i\left[H_s, \rho_s\right]$ to the relaxation superoperator Λ derived under the assumption that $H_s = 0$. During a time step during which evolution due to H_s is negligible, for instance, such a master equation correctly predicts that relaxation is governed by Λ, while the presence of the unitary term allows for the slow modulation of the spin state.

This argument can be formalized by considering the general derivation given in reference [7] of the master equation for a system A coupled to a reservoir R. The interaction Hamiltonian can be written as

$$V = V_A V_R,$$

where V_A acts on A and V_R acts on R. In the absence of the coupling V, the lab-frame Hamiltonians H_A and H_R govern A and R, respectively. These Hamiltonians are eliminated from the evolution equations by a switch from the lab frame to an interaction frame, and second-order time-dependent perturbation theory is used to obtain an interaction-frame expression for the evolution of A and R during a time step Δt. A partial trace is taken over the reservoir degrees of freedom, and the resulting expression is simplified using the assumption that $\Delta t \gg \tau_h$ and the assumption that the reservoir is only weakly perturbed from thermal equilibrium by the interaction with A. A similar derivation can be carried out in the case where the switch from the lab frame to the interaction frame does not completely eliminate the Hamiltonian H_A but rather leaves a "secular" term H_s. A second-order expression for the evolution of the full density matrix for A and R yields terms quadratic in V, terms which are proportional to V and H_s, as well as terms quadratic in H_s. A partial trace over the

reservoir degrees of freedom eliminates the terms which are linear in V, due to the assumption that the thermal average of V_R over the reservoir states is zero. (This condition can always be achieved by adding a term $\langle V_R \rangle V_A$ to the Hamiltonian H_A and then defining $V = V_A V_R'$, where $V_R' = V_R - \langle V_R \rangle$.) The terms depending on the square of V are unaffected by the presence of H_s, and they yield the same relaxation superoperator that would be obtained in the absence of H_s. The remaining terms yield a second-order approximation to the unitary evolution of A associated with H_s. If this second-order approximation is valid throughout the time step Δt, then the resulting master equation for A includes the same relaxation superoperator which would be obtained in the absence of H_s, along with the additional term $-i\,[H_s, \rho_s]$. The relaxation superoperator can therefore be calculated without consideration of H_s if the evolution associated with H_s is sufficiently slow that it can be approximated by second-order perturbation theory during the time step Δt.

We consider a simple example in which $H_s = 0$. Multiplying the master equation (2.17) by I_z and taking the trace gives the derivative of $\langle I_z \rangle (t)$:

$$\frac{d}{dt} \langle I_z \rangle = R_0 \left(n_{\rm th} + 1\right) \langle I_- I_+ \rangle - R_0 n_{\rm th} \langle I_+ I_- \rangle \,. \tag{2.18}$$

$$= -R_0 \left(2n_{\rm th} + 1\right) \langle I_z \rangle + R_0 \left\langle I_x^2 + I_y^2 \right\rangle . \tag{2.19}$$

If only a single spin is present, then

$$I_x^2 = I_y^2 = 1/4,$$

and we obtain

$$\frac{d}{dt} \langle I_z \rangle = -R_0 \left(2n_{\rm th} + 1\right) \left(\langle I_z \rangle - \frac{1/2}{2n_{\rm th} + 1} \right) .$$

This equation describes the exponential relaxation of $\langle I_z \rangle$ toward thermal equilibrium with the resonator, and the rate constant is

$$R_h = R_0 \left(2n_{\rm th} + 1\right) .$$

Since $R_h = R_0$ at when $n_{\rm th} = 0$, we can consider R_0 to be the rate constant in the limiting case $T \to 0$.

The interaction-frame equations of motion for the transverse spin components are

$$\frac{d}{dt}\langle I_x\rangle = -\frac{1}{2}R_h\langle I_x\rangle - R_0\left\langle\frac{1}{2}\left(I_xI_z + I_zI_x\right)\right\rangle, \tag{2.20}$$

$$\frac{d}{dt}\langle I_y\rangle = -\frac{1}{2}R_h\langle I_y\rangle - R_0\left\langle\frac{1}{2}\left(I_yI_z + I_zI_y\right)\right\rangle. \tag{2.21}$$

For a sample consisting of a single spin 1/2, we have

$$I_xI_z + I_zI_x = I_yI_z + I_zI_y = 0,$$

and

$$\frac{d}{dt}\langle I_x\rangle = -\frac{1}{2}R_h\langle I_x\rangle, \tag{2.22}$$

$$\frac{d}{dt}\langle I_y\rangle = -\frac{1}{2}R_h\langle I_y\rangle. \tag{2.23}$$

We can interpret the transverse relaxation with rate constant $R_h/2$ as lifetime broadening associated with the spin transitions induced by the resonator.

2.2 Full master equation for the spin-resonator system

In analyzing resonator-induced relaxation, it is often convenient to use a master equation which includes the resonator's degrees of freedom. If the effects of the spin lattice are neglected, the full master equation for the spin-resonator density matrix ρ in the interaction frame is [5]

$$\frac{d}{dt}\rho = -i\left[H_s + V, \rho\right] + \Lambda\rho, \tag{2.24}$$

where V is the average Hamiltonian (2.11) governing the spin-resonator interaction, and Λ is the relaxation superoperator for the damped mechanical resonator [8]:

$$\begin{aligned}\Lambda\rho = &-\frac{n_{\rm th}+1}{\tau_h}\left[a^\dagger a,\rho\right]_+ + 2\frac{n_{\rm th}+1}{\tau_h}a\rho a^\dagger \\ &-\frac{n_{\rm th}}{\tau_h}\left[aa^\dagger,\rho\right]_+ + 2\frac{n_{\rm th}}{\tau_h}a^\dagger\rho a.\end{aligned} \tag{2.25}$$

Equations (2.19) through (2.21) can be obtained for arbitrary values of I from the full master equation by using a method presented in Appendix A to derive a "coarse-grained" derivative, i.e., the average rate of change during a time step Δt which is long compared to τ_h but short compared to the time needed for spin relaxation. The error associated with the use of the rotating-frame approximation (i.e., the use of an average Hamiltonian obtained by neglecting the off-resonant components of the transverse field) to obtain equations of motion for spin operators can be estimated by replacing the average Hamiltonian used during the time step Δt with a Magnus expansion [9]. The average Hamiltonian (2.11) is the zero-order term in this expansion, and the first-order term is smaller than the average Hamiltonian by a factor of order g/ω_h.

The method given in Appendix A can be used to correct the spin equations of motions to include "secular broadening" associated with the fluctuations in B_z caused by the mechanical motion. Replacing (2.11) by (2.12) as the interaction Hamiltonian does not affect the equation of motion for $\langle I_z\rangle$, but equations (2.20) and (2.21) become

$$\begin{aligned}\frac{d}{dt}\langle I_x\rangle &= -\frac{1}{2}R_h\langle I_x\rangle - R_0\left\langle\frac{1}{2}\left(I_xI_z+I_zI_x\right)\right\rangle \\ &\quad -\frac{1}{2}f^2\tau_h n_{\rm th}\left(n_{\rm th}+1\right)\langle I_x\rangle, \\ \frac{d}{dt}\langle I_y\rangle &= -\frac{1}{2}R_h\langle I_y\rangle - R_0\left\langle\frac{1}{2}\left(I_yI_z+I_zI_y\right)\right\rangle \\ &\quad -\frac{1}{2}f^2\tau_h n_{\rm th}\left(n_{\rm th}+1\right)\langle I_y\rangle.\end{aligned}$$

Appendix B uses a numerical example to demonstrate that

$$f^2\tau_h n_{\rm th}\left(n_{\rm th}+1\right) \ll R_h$$

for the low-temperature nanoscale regime of interest. In this regime, (2.11) may be used as the interaction Hamiltonian, since the corrections introduced by the switch from (2.11) by (2.12) are negligible.

A similar approach can be used to derive equations of motion in the case where the spins' Larmor frequency is separated from the mechanical frequency by an offset β:

$$\omega_0 = -\omega_h + \beta.$$

Appendix C shows that if the spins all experience the same off-resonant field, the rate of longitudinal relaxation is given by

$$\frac{d\langle I_z\rangle}{dt} = \left\{R_0\left(n_{\text{th}}+1\right)\langle I_- I_+\rangle - R_0 n_{\text{th}}\langle I_+ I_-\rangle\right\}\frac{1}{1+\left(\beta\tau_h\right)^2}. \tag{2.26}$$

A rate equation for longitudinal relaxation is also given for the case where the resonator's field varies across the sample. These results can be used to estimate the sample volume which can be cooled toward thermal equilibrium by a mechanical resonator.

3 Spontaneous and stimulated transitions

Agarwal has shown that spontaneous emission from a two-level atom into the vacuum is governed by the operator which is written in our notation as $I_- I_+$ [10]. Since (2.18) can be expressed as

$$\frac{d}{dt}\langle I_z\rangle = R_0\langle I_- I_+\rangle + R_0 n_{\text{th}}\langle I_- I_+\rangle - R_0 n_{\text{th}}\langle I_+ I_-\rangle, \tag{2.27}$$

it is natural to interpret the terms $R_0\langle I_- I_+\rangle$, $R_0 n_{\text{th}}\langle I_- I_+\rangle$, and $R_0 n_{\text{th}}\langle I_+ I_-\rangle$ as characterizing processes analogous to spontaneous emission, stimulated emission, and stimulated absorption, respectively. More precisely, the term $R_0\left(n_{\text{th}}+1\right)$ and $R_0 n_{\text{th}}\langle I_+ I_-\rangle$ are expected to give the respective rates at which the spins donate energy to an oscillator and receive energy from it.

The conjecture can be verified using the general formulas derived in reference [7] for the coefficients of a master equation. The interaction-frame master equation is written as

$$\frac{d}{dt}\rho_{ab}(t) = \sum_{c,d} \exp\left\{i\left(\omega_{ab} - \omega_{cd}\right)t\right\} \mathcal{R}_{abcd}\,\rho_{cd}(t),$$

where ρ_{ij} is an element of the density matrix expressed in the energy eigenbasis, and $\mathcal{R}_{abcd}$ is a constant which characterizes the rate of transfer from ρ_{cd} to ρ_{ab}. The eigenfrequency of spin eigenstate $|a\rangle$ is denoted by ω_a, and the difference of two such eigenfrequencies by

$$\omega_{ab} = \omega_a - \omega_b.$$

A spin transition from state $|b\rangle$ to state $|c\rangle$ changes the respective populations ρ_{bb}, ρ_{cc} of the states, and $\mathcal{R}_{ccbb}$ is the rate constant for this transition. In the case where the transition $b \longrightarrow c$ involves the donation of a quantum from the spins to the resonator, we find by applying the general formulas of reference [7] that

$$\mathcal{R}_{ccbb} = g^2 \left|\langle c\left|I_+\right|b\rangle\right|^2 \int_{-\infty}^{\infty} \exp\left(i\omega_{bc}\tau\right) \left\langle a\left(\tau\right) a^\dagger\left(0\right)\right\rangle d\tau. \tag{2.28}$$

The correlation function $\left\langle a\left(\tau\right) a^\dagger\left(0\right)\right\rangle$ appearing in the integrand can be approximated as

$$\begin{aligned}\left\langle a\left(\tau\right) a^\dagger\left(0\right)\right\rangle &= \left\langle a\left(\tau\right) a^\dagger\left(0\right)\right\rangle \exp\left(-i\omega_h\tau\right) \exp\left(-\tau/\tau_h\right) \\ &= \left(n_{\text{th}}+1\right) \exp\left(-i\omega_h\tau\right) \exp\left(-\tau/\tau_h\right).\end{aligned}$$

When the difference frequency ω_{bc} is resonant with the mechanical frequency, the integrand of (2.28) is equal to $\left(n_{\text{th}}+1\right)\exp\left(-\tau/\tau_h\right)$, and we obtain

$$\mathcal{R}_{ccbb} = 2g^2\tau_h\left(n_{\text{th}}+1\right)\left|\langle c\left|I_+\right|b\rangle\right|^2. \tag{2.29}$$

In the case where the Hamiltonian governing the spins is $\omega_0 I_z$, the energy eigenstates can be chosen to consist of angular momentum manifolds, with I_+ and I_-

the raising and lowering operators within each manifold. In this basis, $\langle c\,|I_+|\,b\rangle$ is nonzero only if $I_+\,|b\rangle = |c\rangle$, and in this case

$$\begin{aligned} |\langle c\,|I_+|\,b\rangle|^2 &= 1 \\ &= \langle b\,|I_-I_+|\,b\rangle\,, \end{aligned}$$

and

$$\mathcal{R}_{ccbb} = R_0\,(n_{\rm th}+1)\,\langle b\,|I_-I_+|\,b\rangle\,.$$

Summing over all transitions $b \longrightarrow c$ for which $\langle c\,|I_+|\,b\rangle$ is nonzero shows that the rate at which quanta are donated to the resonator is

$$\sum_{\langle c|I_+|b\rangle\neq 0} \mathcal{R}_{ccbb}\rho_{bb} = R_0\,(n_{\rm th}+1)\,\langle I_-I_+\rangle\,.$$

Since temperature $T_h = 0$ gives $n_{\rm th} = 0$, we find that $R_0\,\langle I_-I_+\rangle$ is the rate of spontaneous emission, while

$$R_0\,(n_{\rm th}+1)\,\langle I_-I_+\rangle - R_0\,\langle I_-I_+\rangle = R_0 n_{\rm th}\,\langle I_-I_+\rangle$$

is the rate of stimulated emission. Similar arguments can be used to demonstrate that $R_0 n_{\rm th}\,\langle I_+I_-\rangle$ is the rate at which quanta are donated to the spins, i.e., the rate of stimulated absorption.

The contribution of spontaneous and stimulated transitions to longitudinal relaxation can be highlighted by expressing equation (2.27) in the form

$$\frac{d}{dt}\,\langle I_z\rangle = -2R_0 n_{\rm th}\,\langle I_z\rangle - R_0\left\{\langle I_z\rangle - \left\langle I_x^2 + I_y^2\right\rangle\right\}.$$

The term proportional to $n_{\rm th}$ is due to stimulated transitions. In the absence of spontaneous emission, stimulated transitions would cause $\langle I_z\rangle$ to relax exponentially to zero. The remaining contribution is due to spontaneous emission, which drives $\langle I_z\rangle$ toward the instantaneous value of $\left\langle I_x^2 + I_y^2\right\rangle$. In the case where a single spin

1/2 interacts with the resonator, spontaneous emission drives $\langle I_z \rangle$ toward 1/2. In the general case, spin-spin correlations affect the value of $\left\langle I_x^2 + I_y^2 \right\rangle$ and hence the contribution of spontaneous emission to the relaxation of $\langle I_z \rangle$.

4 Physical interpretation of the cooling process

Reference [7] presents a physical interpretation of the energy exchange which occurs between a system $\mathcal{A}$ and a thermal reservoir $\mathcal{R}$ which is weakly-coupled to $\mathcal{A}$ and damps its motion. Two types of processes contribute: 1) Processes in which system $\mathcal{A}$ responds linearly to fluctuations in $\mathcal{R}$, and 2) Processes in which $\mathcal{R}$ responds linearly to the motion of $\mathcal{A}$ and damps this motion. In the case where $\mathcal{A}$ is a single atom and $\mathcal{R}$ is an isotropic and homogeneous radiation field, stimulated emission and absorption are shown to depend on the first type of process, while spontaneous emission is shown to include equal contributions from both types. In particular, the response of the atom to vacuum fluctuation and the response of the electromagnetic field to the motion of the electrons contribute equally to spontaneous emission. The atom continually loses energy as the radiation field responds to its motion (the "radiation reaction"), while the atom can either gain or lose energy when acted upon by vacuum fluctuations. If a two-level atom is in its excited state, vacuum fluctuations and the radiation reaction both transfer energy from the atom to the field at an equal rate. When the atom is in its ground state, however, vacuum fluctuations tend to induce atomic transitions to the excited state, thereby increasing the atom's energy, while the energy transfer due to the radiation reaction cancels the effect of the vacuum fluctuations.

The derivations used in justifying this interpretation can be adapted to yield a similar interpretation of the energy exchanges between a single spin 1/2 and a damped mechanical resonator. Consider as an example a problem in which the initial spin density matrix ρ_s is diagonal in the product-state eigenbasis. It follows from (2.17) that ρ_s will remain diagonal as the spin relaxes, since the derivative of ρ_s is itself diagonal. (This can be verified directly using matrix representations of

I_- and I_+.) In this case, the mean transverse dipole is zero during the relaxation, and it is the fluctuations in the transverse dipole which drive the mechanical motion. The fluctuations are damped as they drive the resonator, and the resulting transfer of energy from spin to resonator is a mechanical analog of the radiation reaction. When $T_h = 0$ and the spin is in its ground state, no transitions occur, since spin fluctuations drive the resonator and donate energy to it at the same rate that zero-point fluctuations return energy to the spin. If the spin is in the excited state, spin fluctuations and zero-point motion both contribute equally to spontaneous emission.

This interpretation is consistent with the idea that transverse spin fluctuations continue to occur even when a system is in its ground state. Consistent with this interpretation is the convention that the mean square fluctuation of a complex operator $T = T_1 + iT_2$ be defined as [11]

$$\begin{aligned} |\Delta T|^2 &\equiv \frac{1}{2}\left\langle TT^\dagger + T^\dagger T\right\rangle - \left\langle T\right\rangle\left\langle T^\dagger\right\rangle \\ &= (\Delta T_1)^2 + (\Delta T_2)^2 . \end{aligned} \tag{2.30}$$

Under this convention, the mean square fluctuations in the resonator's complex amplitude in thermal equilibrium are

$$|\Delta a|^2 = \left|\Delta a^\dagger\right|^2 = n_{\rm th} + 1/2,$$

while the thermal fluctuations in the transverse spin are given by

$$|\Delta I_+|^2 = |\Delta I_-|^2 = \left\langle I_x^2 + I_y^2\right\rangle .$$

For a system consisting of a resonator at $T_h = 0$ and a spin 1/2 in its ground state, we have

$$\begin{aligned} |\Delta a|^2 &= 1/2, \\ |\Delta I_+|^2 &= 1/2. \end{aligned}$$

Definition (2.30) is not appropriate in all cases, however. Although a spin 1/2 in its ground state coupled to a mode at $T_h = 0$ can be visualized as actively exchanging energy with the mode, no net radiation into or out of the mode will be detectable. Note also that the results presented in 2.3 imply that the radiation emitted by transverse spin fluctuations is characterized by the operator $I_- I_+$, rather than by $|\Delta I_+|^2$. In studying "radiative transverse fluctuations" of a mechanical oscillator or a spin system, an alternative to definition (2.30) may be used. If we let T denote the complex operator which removes a quantum from the radiating system (i.e., I_+ in the case of radiating spins or a in the case of a radiating mode), the radiative fluctuations of the spins and the resonator are characterized by

$$|\Delta' T|^2 \equiv \left\langle T^\dagger T \right\rangle - \langle T \rangle \left\langle T^\dagger \right\rangle . \tag{2.31}$$

For a resonator or a spin system in a thermal state, definition (2.31) yields the respective operators

$$\begin{aligned} \left\langle a^\dagger a \right\rangle &= n_{\text{th}}, \\ \langle I_- I_+ \rangle &= \left\langle I_x^2 + I_y^2 \right\rangle - \langle I_z \rangle . \end{aligned}$$

At zero Kelvins, the radiative fluctuations defined by these operators are zero.

The physical interpretation of energy exchange given in reference [7] can also be used to explain the appearance of the resonator's ringdown time τ_h in the rate constant

$$R_0 = 2g^2 \tau_h. \tag{2.32}$$

When subject to a time-dependent input $x(t)$, the response $y(t)$ of a linear system can be expressed as

$$y(t) = \int_{-\infty}^{+\infty} x(t')\, h(t - t')\, dt', \tag{2.33}$$

where $h(t - t')$ gives the response at time t to a unit impulse applied at time t'. In using (2.33) to explain the appearance of τ_h in equation (2.32), we consider the

two types of process which contribute to energy exchange between the spins and the damped resonator. For processes in which the spins respond linearly to resonator fluctuations, the linear system of equation (2.33) is the spin sample, the input $x(t)$ is the fluctuating field of the mechanical resonator, and τ_h is the correlation time of $x(t)$. Short τ_h limits the rate of stimulated emission and absorption by limiting the time period during which the spins experience the steady periodic field that induces transitions. For processes in which the damped resonator responds linearly to spin motion and damps this motion (i.e., the mechanical analog of the "radiation reaction" mentioned above), the decay time of the impulse response $h(t)$ is τ_h, and so τ_h limits the time period during which the mechanical response can "ring up."

Note that the derivation of equation (2.32) depends on the assumption that τ_h is much shorter than the correlation time of the transverse sample dipole, and as a result, τ_h is the only correlation time which appears in this equation. If the transverse spin correlation time is short enough to have a significant effect in determining the time period during which the linear response of the spins and resonator can accumulate, then the rate constant R_0 must be modified to take account of the effects of spin fluctuations. This could be done by adding a superoperator for spin relaxation to the spin-resonator master equation and then performing a derivation similar to that of Appendix A. The discussion in section 2 of chapter 7 illustrates a method of including the effects of spin relaxation in an approximate way without using an explicit expression for the spin relaxation superoperator.

5 Semiclassical model

The motion of spin systems can often be visualized using a semiclassical model in which each spin component has a definite value at all times. Such a model can be considered a formalization of the "finger physics" pictures that are used to visualize spin evolution. We present a semiclassical model of the spin-resonator system which can be used to visualize the spin-resonator interaction and also to distinguish the relaxation governed by (2.18) from so-called radiation damping [12]. The semiclassical

spin $\mathbf{I}^c$ is governed by the equation

$$\frac{d}{dt}\mathbf{I}^c = \gamma \mathbf{I}^c \times \mathbf{B}, \tag{2.34}$$

i.e., $\mathbf{I}^c$ precesses around the instantaneous field at frequency

$$\omega_0 = -\gamma B.$$

The magnetic dipole associated with $\mathbf{I}^c$ is

$$\mu^c = \gamma \hbar \mathbf{I}^c.$$

A classical damped mechanical resonator is coupled to the spin system by the Hamiltonian

$$W = -\mu^c \cdot \mathbf{B}(\theta),$$

and the torque exerted on the resonator by the dipole is

$$-\frac{\partial W}{\partial \theta} = \frac{dB_x}{d\theta}\mu_x^c.$$

Precession of $\mathbf{I}^c$ around the applied field causes μ_x^c to vary sinusoidally. The mechanical oscillator thus responds to a resonant driving torque, and energy can be transferred from the spins to the oscillator by this driving torque. The resonant rotating component of the oscillator's field simultaneously exerts a torque on the spins and causes spin rotation toward or away from the static applied field. As in the derivative of the quantum mechanical equations of motion, coarse-grained relaxation equations can be found by integrating the motion over a time step Δt which is long compared to τ_h but short compared to spin relaxation time.

Appendix D derives an equation of motion for $\langle I_z^c \rangle$, where the average is taken

over a statistical ensemble of spin-resonator systems:

$$\frac{d\langle I_z^c\rangle}{dt} = -2R_0\frac{\langle E_h^c\rangle}{\hbar\omega_h}\langle I_z^c\rangle + R_0\left\langle I_-^c I_+^c\right\rangle. \tag{2.35}$$

In (2.35), I_+^c, I_-^c are defined as

$$I_\pm^c = I_x^c \pm iI_y^c,$$

and $\langle E_h^c\rangle$ is the mean thermal energy of the classical resonator. From equation (2.18), we can obtain a formally equivalent equation for longitudinal relaxation of the quantum mechanical system:

$$\frac{d\langle I_z\rangle}{dt} = -2R_0\frac{\langle E_h\rangle}{\hbar\omega_h}\langle I_z\rangle + R_0\langle I_-I_+\rangle, \tag{2.36}$$

where

$$\langle E_h\rangle = \hbar\omega_h n_{\text{th}}$$

can be considered the mean thermal energy of the quantum resonator, with zero-point energy excluded.

The formal equivalence between (2.35) and (2.36) masks the fact that the commutation properties of the quantum operators can yield distinctly nonclassical relaxation in the quantum system. Writing the two equations in the form

$$\frac{d\langle I_z^c\rangle}{dt} = -2R_0\frac{\langle E_h^c\rangle}{\hbar\omega_h}\langle I_z^c\rangle + R_0\left\langle (I_x^c)^2 + \left(I_y^c\right)^2\right\rangle, \tag{2.37}$$

$$\frac{d\langle I_z\rangle}{dt} = -2R_0\frac{\langle E_h\rangle}{\hbar\omega_h}\langle I_z\rangle + \left\{R_0\left\langle I_x^2 + I_y^2\right\rangle - R_0\langle I_z\rangle\right\} \tag{2.38}$$

highlights the failure of the semiclassical model to characterize correctly the spontaneous emission which is responsible for polarizing the spins in the quantum system at low temperatures. As shown in section 3, the terms in curly brackets on the right side of (2.38) are due to spontaneous emission. Section 4 interprets spontaneous emission as including equal contributions from the spins' response to zero-point fluctuations

of the resonator's field, and the resonator's response to transverse spin fluctuations that are present even in a perfectly polarized sample. Since these phenomena are not present in the semiclassical model, it is not surprising that spontaneous emission is not correctly characterized by this model. Equations (2.35) and (2.36) show that discrepancies between the quantum and semiclassical models can be expected when

$$\frac{\langle E_h \rangle}{\hbar \omega_h} = n_{\mathrm{th}}$$

has order of magnitude unity or less, as well as when $\left\langle I_x^2 + I_y^2 \right\rangle$ differs significantly from the value that would be calculated if each spin component had a definite value simultaneously.

As an illustration, we compare the semiclassical phenomenon called "radiation damping" with the longitudinal polarization of a spin 1/2 by a cold resonator. Magnetization precessing in an inductive coil excites current oscillations within the coil circuit, and radiation damping occurs when the field generated within the coil by the oscillating current is strong enough to rotate the magnetization into alignment with the static applied field, thereby shortening the precession period. In the case where spins are coupled to a mechanical rather than an inductive resonator, the analogous phenomenon occurs when the mechanical response to precessing spins creates a resonant field which rotates the spins.

Abragam has derived a rate equation for radiation damping using a model equivalent to our semiclassical model [12]. Adopting the language and notation of our semiclassical model, we can say that Abragam derives the rate equation by assuming that a single semiclassical dipole μ^c interacts with a classical resonator in which thermal fluctuations can be neglected, i.e., a classical resonator at temperature $T_h = 0$. The equation of motion for $\langle I_z^c \rangle$ during radiation damping can therefore be obtained from (2.37) by setting $\langle E_h^c \rangle$ to zero:

$$\frac{d \langle I_z^c \rangle}{dt} = R_0 \left\langle (I_x^c)^2 + \left(I_y^c\right)^2 \right\rangle . \tag{2.39}$$

Equation (2.39) implies that $d\langle I_z^c\rangle/dt$ is zero if the semiclassical dipole is aligned with the negative z-axis, and that $d\langle I_z^c\rangle/dt$ takes on its maximum value when the dipole lies in the transverse plane. This is consistent with the fact that it is the transverse dipole which drives the mechanical motion and thereby induces the resonant field responsible for radiation damping.

For purposes of comparison, note that the quantum mechanical equation of motion is

$$\frac{d\langle I_z\rangle}{dt} = R_0\left\langle I_x^2 + I_y^2\right\rangle - R_0\langle I_z\rangle \tag{2.40}$$

when the resonator is at zero Kelvins. Consider an example in which the spin sample consists of a single spin 1/2. Since

$$I_x^2 + I_y^2 = 1/2,$$

equation (2.40) reduces to

$$\frac{d\langle I_z\rangle}{dt} = -R_0\left(\langle I_z\rangle - 1/2\right). \tag{2.41}$$

The evolution governed by (2.41) is distinctly different from that governed by (2.39). If the spin is aligned with the negative z-axis, for instance, $d\langle I_z\rangle/dt$ takes on its maximum possible value, and the derivative decreases linearly as $\langle I_z\rangle$ is increased, reaching zero when the spin is oriented along the positive z-axis. Since $I_x^2 + I_y^2$ is a constant, $d\langle I_z\rangle/dt$ depends only on $\langle I_z\rangle$. In particular, since a polarized spin 1/2 initially precessing in the transverse plane has the same initial value of $\langle I_z\rangle$ as a spin which is initially completely unpolarized, the evolution of $\langle I_z\rangle$ is the same in both cases.

The most striking difference between the semiclassical and quantum models is that only the quantum mechanical model allows for the polarization of a sample of spins which all experience the same field. Equation (2.34) implies that the derivative $d\mathbf{I}^c/dt$ is perpendicular to $\mathbf{I}^c$, and it follows that this equation of motion describes rotation of $\mathbf{I}^c$. The magnitude $|\mathbf{I}^c|$ cannot change as a result of the spin-resonator

interaction. The quantum model does allow polarization, as can be seen from equation (2.41). For a single spin 1/2, relaxation of $\langle I_z \rangle$ toward thermal equilibrium proceeds independently of I_x and I_y. As we will see in chapter 3, resonator-induced polarization is also possible for a system of N spins.

6 Polarization of spins using an inductive resonator

Reference [13] demonstrates that mechanical detection of NMR signals is more sensitive than inductive detection for sufficiently small samples, with mechanical detection typically becoming more sensitive when the sample radius is on the order of tens of microns to hundreds of microns. We extend this result by presenting a numerical estimate which suggest that a nanoscale inductive resonator would not efficiently cool a nanoscale spin sample.

6.1 Rate constant for longitudinal relaxation

An classical LC circuit is governed by the Hamiltonian

$$H = \frac{p^2}{2L} + \frac{q^2}{2C},$$

where q is charge, L is inductance, and C is capacitance. The inductive oscillator has the coordinate q, and the conjugate momentum is

$$p = L\dot{q},$$

where $\dot{q}$ is the current flowing in the circuit. For a sufficiently long solenoid, the solenoid's field at the spins, which we will denote by B_y, is

$$\begin{aligned} B_y &= \mu_0 n \dot{q} \\ &= \frac{\mu_0 n}{L} p, \end{aligned}$$

where n is the number of turns per unit length. We will assume that the inductive resonator's field at the spins can be written in the form

$$\mathbf{B}_s(p) = \left(0, \frac{dB_y}{dp}p, 0\right),$$

with

$$\frac{dB_y}{dp} = \frac{\mu_0 n}{L}.$$

This assumption yields the following Hamiltonian (in units of rad/s) for the spin-resonator system:

$$H = \omega_0 I_z + \omega_h \left(a^\dagger a + 1/2\right) - \gamma I_y \frac{dB_y}{dp} p.$$

Substituting

$$p = i\sqrt{\frac{L\hbar\omega_h}{2}}\left(a^\dagger - a\right)$$

and

$$I_h = \frac{1}{2i}\left(I_+ - I_-\right)$$

into the Hamiltonian gives

$$\begin{aligned} H = {} & \omega_0 I_z + \omega_h \left(a^\dagger a + 1/2\right) \\ & - \frac{\gamma}{2}\frac{dB_y}{dp}\sqrt{\frac{L\hbar\omega_h}{2}}\left(a^\dagger - a\right)\left(I_+ - I_-\right). \end{aligned}$$

In the interaction frame, the term in the Hamiltonian which survives averaging is

$$V = -\frac{\gamma}{2}\frac{dB_y}{dp}\sqrt{\frac{L\hbar\omega_h}{2}}\left(I_+ a^\dagger + I_- a\right). \tag{2.42}$$

Equation (2.42) shows that for both inductive and mechanical resonators, the spin-resonator Hamiltonian in the interaction frame takes the same form. For the inductive resonator, we define

$$g_{\text{induct}} = -\frac{\gamma}{2}\frac{dB_y}{dp}\sqrt{\frac{L\hbar\omega_h}{2}}.$$

Longitudinal relaxation of the spins will be governed by the rate constant

$$R_{\text{induct}} = 2g^2_{\text{induct}}\tau_{\text{induct}}\left(2n_{\text{th}}+1\right), \tag{2.43}$$

where the ringdown time τ_{induct} of the inductive resonator is given by

$$\tau_{\text{induct}} = 2L/R,$$

with R the resistance in the inductive circuit. Equation (2.43) can be written more explicitly as

$$R_{\text{induct}} = \left(\gamma\mu_0 n\right)^2 \frac{\hbar\omega_h}{2R}\left(2n_{\text{th}}+1\right).$$

6.2 Comparison of mechanical and inductive resonators

In comparing mechanical and inductive resonators, we first consider the way in which g^2 scales with size. If ω_h varies as $1/r$, we find that

$$\begin{aligned} g^2_{\text{mech}} &\propto \frac{1}{I_h\omega_h} \\ &\propto 1/r^4, \end{aligned}$$

and

$$\begin{aligned} g^2_{\text{induct}} &\propto \frac{n^2\omega_h}{L} \\ &\propto 1/r^4. \end{aligned}$$

However, if ω_h is assumed to be determined by a fixed field which does not vary during the scaling, we obtain

$$\begin{aligned} g^2_{\text{mech}} &\propto 1/r^5, \\ g^2_{\text{induct}} &\propto 1/r^3. \end{aligned}$$

At a given frequency, the the strength of the spin-resonator coupling depends more strongly on size if the resonator is mechanical.

The spin-relaxation rate depends on the resonator ringdown time as well as the coupling strength, and a quantitative comparison of spin relaxation rates for mechanical and inductive resonators is not possible because the dependence of mechanical ringdown time on size and temperature is poorly understood. We can, however, make simple estimates which suggest that inductive resonators would not efficiently cool spins. The resistance R of a coil is scale invariant if the skin depth is smaller than the radius of the wire used in the windings of the coil, while R scales as $1/r$ in the regime where the current flows uniformly through the wire [13]. Cooling an exceptionally pure conductor to a temperature of a few Kelvins or below can increase its conductivity by a factor of up to 10^6 [14], which would yield a skin depth for copper of a few nanometers at 200 MHz. In order to make an estimate advantageous to inductive cooling, we assume that R is scale invariant, setting aside the question of whether a nanoscale inductor of this purity could be fabricated. This assumption yields

$$\tau_{\text{induct}} = 2L/R \propto r$$

and

$$g^2_{\text{induct}} \tau_{\text{induct}} \propto 1/r^2. \tag{2.44}$$

The longitudinal relaxation rate constant R_{induct} for the example coils presented in reference [13] were calculated using the assumption that the resonator's conductivity increased by a factor of 10^6 over that of room-temperature copper due to cooling of the coil to mK temperatures:

$$R_{\text{induct}} \approx 3 \times 10^{-11}\,\text{s}^{-1}\,.$$

These example coils have length and diameter of order 50 μm. Scaling down these dimensions by a factor of 10^3 under the assumption that (2.44) holds would increase R_{induct} by six orders of magnitude, yielding a rate constant that is negligible compared

to the value

$$R_h \sim 1\,\mathrm{s}^{-1}$$

obtained from numerical examples presented in section 6 of chapter 5.