Relaxation from fluctuating fields in a spin-1/2 system

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1 Introduction

We describe fluctuations of the local magnetic field around its mean. Split

$$\boldsymbol{B}(t) = B_0\,\hat{\boldsymbol{z}} + \boldsymbol{B}_1(t), \qquad \langle \boldsymbol{B}_1(t)\rangle = 0,$$

where $\boldsymbol{B}_1$ is not an RF-field, but a stochastic magnetic field coming from thermal motion of electrons and protons in the vicinity of the spin, i.e. a thermal bath. As a stochastic variable, it can be characterized by its moments — in this note, we will use angular brackets $\langle\cdot\rangle$ to mean an average over the distribution of $\boldsymbol{B}_1$. The first moment, i.e. the mean, is zero for all time points per definition. We further assume stationarity, meaning that the *two-point correlation function*

$$\langle B_{1\alpha}(t)B_{1\beta}(t+\tau)\rangle = C_{\alpha\beta}(\tau).$$

depends only on the time difference. Assume correlations decay on the time scale of a correlation time, τ_c, meaning $C_{\alpha\beta}(\tau)$ is appreciable mainly for $|\tau| \lesssim \tau_c$. See Fig. (1). Typically, for many liquids, τ_c is on the order of picoseconds. Define its Fourier transform, the (two-sided) spectral density

$$J_{\alpha\beta}(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau}\, C_{\alpha\beta}(\tau).$$

2 R_2: dephasing from longitudinal fluctuations

Consider a spin whose Larmor frequency in the rotating frame fluctuates due to $B_{1z}(t)$:

$$\Omega(t) = \gamma\, B_{1z}(t).$$

The transverse phase is thus

$$\phi(t) = -\int_0^t dt_1\, \Omega(t_1),$$

and the (complex) transverse magnetization is

$$M_+(t) = \left\langle e^{-i\phi(t)}\right\rangle = \left\langle \exp\left(-i\int_0^t dt_1\,\Omega(t_1)\right)\right\rangle.$$

2.1 Second-order cumulant (Gaussian/weak-fluctuation approximation)

For a Gaussian stochastic variable ϕ with zero mean, we have the general result that

$$\langle \exp(-i\phi)\rangle = \exp\left(-\frac{1}{2}\langle\phi^2\rangle\right).$$

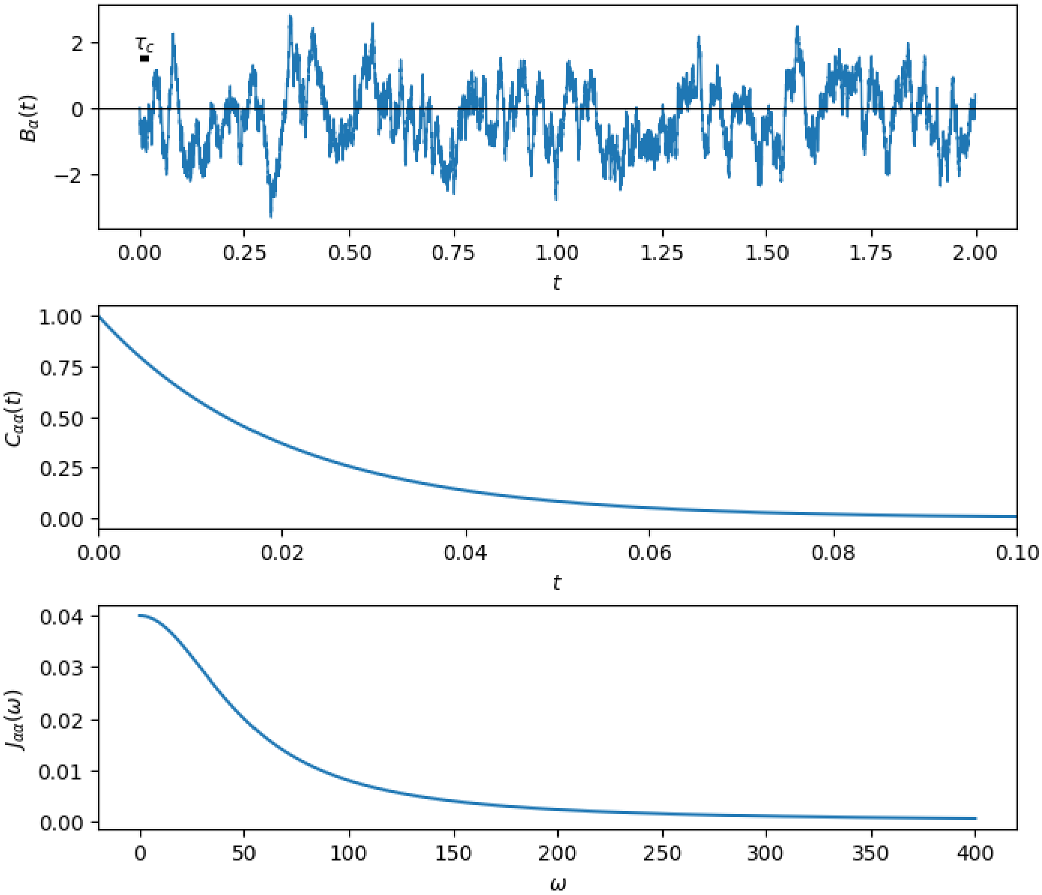

Figure 1: Top: Typical fluctuating component α of the local magnetic field, $B_\alpha(t)$. Middle: The correlation function, $C_{\alpha\alpha}(t) = \langle B_\alpha(t_0)B_\alpha(t_0+t)\rangle$, and its Fourier transform $J_{\alpha\alpha}(\omega)$, bottom. All quantities are in arbirary units.

We assume $\langle\Omega(t)\rangle = 0$, and assume that the distribution of $\Omega(t)$ is Gaussian, hence

$$\begin{aligned} M_+(t) &\approx \exp\left(-\frac{1}{2}\left\langle\left[\int_0^t dt_1\,\Omega(t_1)\right]^2\right\rangle\right) \\ &= \exp\left(-\frac{1}{2}\int_0^t dt_1\int_0^t dt_2\,\langle\Omega(t_1)\Omega(t_2)\rangle\right). \end{aligned} \tag{1}$$

Stationarity gives

$$\langle\Omega(t_1)\Omega(t_2)\rangle = \gamma^2 C_{zz}(t_2 - t_1).$$

So

$$M_+(t) \approx \exp\left(-\frac{\gamma^2}{2}\int_0^t dt_1\int_0^t dt_2\,C_{zz}(t_2-t_1)\right).$$

The integration in the exponent is over a rectangular area as shown in Fig. (2). Due to the behavior of the correlation function $C_{zz}(t_2-t_1)$, only a narrow band of width $\sim 2\tau_c$ around $t_1 \approx t_2$ (i.e. the diagonal band in the figure) contributes. Change variables to

$$t = \frac{t_1+t_2}{2}, \qquad \tau = t_2 - t_1.$$

For $t \gg \tau_c$, the only substantial contributions come from $|\tau| \lesssim \tau_c$ Then the square $[0,t]\times[0,t]$ effectively contributes an area $\sim t\times(2\tau_c)$, and one gets the standard approximation

$$\int_0^t dt_1\int_0^t dt_2\,C_{zz}(t_2-t_1)\ \approx\ t\int_{-\infty}^{\infty} d\tau\,C_{zz}(\tau).$$

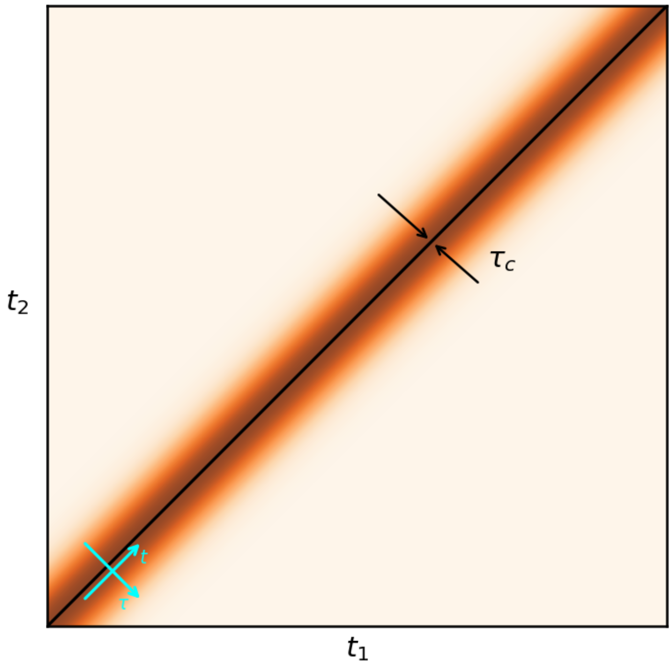

Figure 2: In the t_1–t_2 plane, the correlator $\langle\Omega(t_1)\Omega(t_2)\rangle = \gamma^2 C(t_2 - t_1)$ contributes mainly in a diagonal band of width $\sim \tau_c$. This is the geometric reason that $\int_0^t dt_1 \int_0^t dt_2\, J(t_2-t_1) \approx t \int_{-\infty}^{\infty} d\tau\, J(\tau)$ for $t \gg \tau_c$. The cyan axes at the bottom left show the transformed coordinates $t = (t_1 + t_2)/2$ and $\tau = t_2 - t_1$.

Here we extended the τ integration to $[-\infty; \infty]$ with negligible error. Therefore

$$M_+(t) \approx \exp\left(-t\frac{\gamma^2}{2}\int_{-\infty}^{\infty} d\tau\, C_{zz}(\tau)\right) = \exp\left(-t\frac{\gamma^2}{2}J_{zz}(0)\right) \equiv \exp(-R_2 t).$$

Thus we have approximately

$$R_2 \approx \frac{\gamma^2}{2} J_{zz}(0).$$

We see that transverse dephasing is sensitive to *slow* (near-zero-frequency) fluctuations of the longitudinal field.

3 R_1: population relaxation from transverse fluctuations

R_1 controls the relaxation of the z component of the magnetization/spin operator, which is related to the population of the σ_z eigenstates, the diagonal components of the density operator ρ. We therefore must include transverse fluctuations that can affect the populations i.e., induce spin flips. With $\boldsymbol{\mu} = \gamma \boldsymbol{S} = \frac{\hbar}{2}\gamma\boldsymbol{\sigma}$, where $\boldsymbol{\sigma}$ are the Pauli matrices, the Hamiltonian is

$$\mathcal{H}(t) = -\frac{\hbar\gamma}{2}\boldsymbol{B}(t)\cdot\boldsymbol{\sigma} = \mathcal{H}_0 + V(t)$$

with

$$\mathcal{H}_0 = -\frac{\hbar\gamma}{2}B_0\sigma_z = -\frac{\hbar\omega_0}{2}\sigma_z$$
$$V(t) = -(\hbar\gamma/2)\boldsymbol{B}_1(t)\cdot\boldsymbol{\sigma}.$$

The time evolution of the density matrix ρ obeys

$$\dot{\rho}(t) = -\frac{i}{\hbar}[\mathcal{H}(t), \rho(t)],$$

which simplifies in the interaction picture w.r.t. $\mathcal{H}_0$ (i.e., rotating frame), $\rho \to \rho_I = e^{(i/\hbar)\mathcal{H}_0 t}\rho e^{-(i/\hbar)\mathcal{H}_0 t}$:

$$\dot{\rho}_I(t) = -\frac{i}{\hbar}[V_I(t), \rho_I(t)]. \tag{2}$$

Decompose transverse components via $B_{1\pm} = B_{1x} \pm iB_{1y}$ and $\sigma_\pm = (\sigma_x \pm i\sigma_y)/2$. Then

$$V_I(t) = e^{(i/\hbar)\mathcal{H}_0 t}V(t)e^{-(i/\hbar)\mathcal{H}_0 t} = -\frac{\hbar\gamma}{2}\left[B_{1z}(t)\sigma_z + B_{1-}(t)e^{-i\omega_0 t}\sigma_+ + B_{1+}(t)e^{+i\omega_0 t}\sigma_-\right].$$

3.1 Lowest order

Integrate once Eq. (2) formally once giving

$$\rho_I(t) = \rho_I(0) - \frac{i}{\hbar}\int_0^t d\tau\,[V_I(\tau), \rho_I(\tau)]. \tag{3}$$

and substitute back into Eq. (2) to find:

$$\begin{aligned}\dot{\rho}_I(t) = &-\frac{i}{\hbar}[V_I(t), \rho_I(0)] \\ &-\frac{1}{\hbar^2}\int_0^t d\tau\,[V_I(t), [V_I(\tau), \rho_I(\tau)]].\end{aligned} \tag{4}$$

We now introduce three physically motivated approximations.

Vanishing first-order term (zero mean field). We assume

$$\langle V_I(t)\rangle = 0,$$

i.e. the fluctuating field has zero mean. Upon averaging over realizations of the stochastic field, the first term in Eq. (4) vanishes. Thus, after ensemble averaging, relaxation appears first at second order in the fluctuating field. We thus have

$$\dot{\rho}_I(t) \approx -\frac{1}{\hbar^2}\int_0^t d\tau\,\big\langle[V_I(t), [V_I(\tau), \rho_I(\tau)]]\big\rangle = -\frac{1}{\hbar^2}\int_0^t d\tau\,\big\langle[V_I(t), [V_I(t-\tau), \rho_I(t-\tau)]]\big\rangle \tag{5}$$

where we changed variables in the last step, $\tau \to t-\tau$. We are strictly speaking really working with the averaged state $\langle\rho_I(t)\rangle$ from here on. Within the correlation window $\tau \lesssim \tau_c$, we neglect correlations between the instantaneous system state and the bath fluctuations (weak coupling approximation, seee below).

Short-memory approximation. The commutator in the integrand essentially involves a sum of different correlation functions, $C_{\alpha\beta}(\tau)$. These field correlation functions decay rapidly for $|\tau| \gtrsim \tau_c$, and therefore the integrand is appreciable only for τ close to 0. Therefore, for times $t \gg \tau_c$ we make only a minor error by extending the upper limit of integration from t to infinity, as the integrand is approximately zero there anyway:

$$\int_0^t d\tau \;\to\; \int_0^\infty d\tau.$$

with the result that

$$\dot{\rho}_I(t) \approx -\frac{1}{\hbar^2}\int_0^t d\tau\,\big\langle[V_I(t), [V_I(\tau), \rho_I(\tau)]]\big\rangle \approx -\frac{1}{\hbar^2}\int_0^\infty d\tau\,\big\langle[V_I(t), [V_I(t-\tau), \rho_I(t-\tau)]]\big\rangle \tag{6}$$

Weak-coupling approximation. The double commutator term is already second order in V_I. If the fluctuations are weak,

$$\gamma^2 \langle B_1^2 \rangle \tau_c^2 \ll 1,$$

then $\rho_I(\tau)$ changes only slightly over a correlation time. But recall from above that the integrand is only substantially different from 0 near $|\tau - t| \lesssim \tau_c$. Hence, within this region we may approximate

$$\rho_I(\tau) \approx \rho_I(t).$$

Physically, the spin state evolves on a much slower time scale (T_1, T_2) than the bath correlation time τ_c. In fact, $T_1 \sim$1 s, $T_2 \sim$1–100ms, whereas $\tau_c \sim$1ps.

With these three approximations, we finally have the Redfield equation

$$\dot{\rho}_I(t) \approx -\frac{1}{\hbar^2} \int_0^\infty d\tau \left\langle [V_I(t), [V_I(t-\tau), \rho_I(t)]] \right\rangle.$$

where we have also taken the average over magnetic field fluctuations. We care about $M_z(t) = Tr(\sigma_z \rho_I(t))$. Upon assuming vanishing cross-correlations, i.e., $\langle B_\alpha(0) B_\beta(t) \rangle = 0$ for $\alpha \neq \beta$, one finds (see exercise) a *rate equation*

$$\dot{M}_z(t) = -(W_+ + W_-) M_z(t) + (W_- - W_+),$$

where W_+ is the rate for $|-\rangle \to |+\rangle$ and W_- for $|+\rangle \to |-\rangle$. Crucially, the oscillating factors $e^{\pm i\omega_0 t}$ generate Fourier selection:

$$W_+ = \frac{\gamma^2}{4} \int_{-\infty}^{\infty} d\tau\, e^{+i\omega_0\tau} \langle B_{1-}(0) B_{1+}(\tau) \rangle = \frac{\gamma^2}{4} J_{-+}(\omega_0), \tag{7}$$

$$W_- = \frac{\gamma^2}{4} \int_{-\infty}^{\infty} d\tau\, e^{-i\omega_0\tau} \langle B_{1+}(0) B_{1-}(\tau) \rangle = \frac{\gamma^2}{4} J_{+-}(\omega_0). \tag{8}$$

Therefore the longitudinal relaxation rate is

$$R_1 = W_+ + W_- = \frac{\gamma^2}{4} \Big[J_{+-}(\omega_0) + J_{-+}(\omega_0) \Big] = \frac{\gamma^2}{2} \Big[J_{xx}(\omega_0) + J_{yy}(\omega_0) \Big].$$

4 Intuitive summary

R_2 governs dephasing of transverse magnetization, i.e., the disperion of phases in a population of spins. Each phase is an integral over $\Omega(t)$, so the leading decay involves $\int_0^t dt_1 \int_0^t dt_2\, C_\Omega(t_2 - t_1)$. For $t \gg \tau_c$ this becomes $\sim t \int d\tau\, C_\Omega(\tau) = t\, J_\Omega(0)$. So R_2 samples **low-frequency** noise. R_1 controls the relaxation of the z component of the magnetization/spin operator, which are related to the population of the eigenstates. Changes thus require spin flips driven by B_{1x}, B_{1y}. In the interaction picture those terms oscillate as $e^{\pm i\omega_0 t}$, so the same second-order correlation integral becomes $\int d\tau\, C_\perp(\tau) e^{\pm i\omega_0\tau} = J_\perp(\omega_0)$. So R_1 samples **noise at the Larmor frequency**.

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