Application of Resonance Perturbation Theory to Dynamics of Magnetization in Spin Systems Interacting with Local and Collective Bosonic Reservoirs

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Abstract

We apply our recently developed resonance perturbation theory to describe the dynamics of magnetization in paramagnetic spin systems interacting simultaneously with local and collective bosonic environments. We derive explicit expressions for the evolution of the reduced density matrix elements. This allows us to calculate explicitly the dynamics of the macroscopic magnetization, including characteristic relaxation and dephasing time-scales. We demonstrate that collective effects (i) do not influence the character of the relaxation processes but merely renormalize the relaxation times, and (ii) significantly modify the dephasing times, leading in some cases to a complicated (time inhomogeneous) dynamics of the transverse magnetization, governed by an effective time-dependent magnetic field.

1 Introduction

When quantum systems interact with their environments the effects of relaxation and decoherence occur [1–8]. In this paper we study relaxation and decoherence in quantum macroscopic systems of "effective" spins interacting simultaneously with both local and collective thermal environments. By "effective" we mean that our approach can be applied not only to magnetic spin systems, but also to many quantum systems with discrete energy levels, including recently widely discussed quantum bits (qubits) based on superconducting Josephson junctions and SQUIDs [9–14]. We also would like to mention here the research on ephaptic coupling of cortical neurons, when both local and collective electrical fields play a significant role in the synchronization dynamics of neurons [15]. We assume that spins do not interact directly among themselves, but only through their interactions with collective (energy conserving and energy exchange)

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bosonic environments ("thermal baths", "reservoirs"). The relaxation in these systems is caused by energy exchange between the environments and spins. The rate of relaxation is usually characterized by the spectral density of noise of the reservoirs at the transition frequency, ω, of spins in their effective magnetic field, and by the interaction constant between a spin and an environment [7, 9, 14]. The rate of decoherence usually has a more complicated dependence on the parameters of spins, their local environments, and interaction constants [7–9, 14, 16]. In particular, low-frequency noise ($1/f$ noise) makes a significant contribution to the decoherence rate [17–20] (see also references therein).

Usually relaxation and decoherence are unwanted effects, for example, in a quantum computer one must maintain quantum coherence for long times [5]. But dissipative effects can also be put to good use, for example, in magnetic resonance imaging (MRI) [21–23]. Indeed, in this case, different values of relaxation times (T_1) for different substances (e.g. water and biological tissues) allow one to distinguish and visualize pathological developments in tissues [21]. Dissipative effects can also be utilized, for example, to analyze and classify the influence of many types of defects and impurities, in order to improve the properties of materials [24].

Improving our understanding of relaxation and decoherence processes is important for many fields of science and for many applications. The main problem associated with dissipative effects is that there are many different sources of noise and thermal fluctuations which lead to relaxation and decoherence. We mention only some of them, electromagnetic and acoustic fluctuations (bosonic degrees of freedom), magnetic fluctuations (such as two-level systems in superconducting materials), charge defects, and non-equilibrium quasiparticles. Generally, it is impossible to eliminate all sources of noise, so some additional classification can be useful. For example, in [16] we demonstrated that, for a system of N spins interacting with bosonic environments, one can introduce clusters of reduced density matrix elements in such a way that to a given cluster corresponds a decoherence rate describing the fading of all matrix elements belonging to it. When dealing with a quantum algorithm in quantum computation, this could imply that the decay of some clusters is rapid, but – if the algorithm is built mainly on the use of slower decaying clusters – that decay may not influence significantly the fidelity of the quantum protocol.

In this paper we are mainly interested in the effects produced by simultaneous influence of both local and collective bosonic environments on the dynamics of a collective magnetization in a system of N non-interacting (paramagnetic) spins in a time-independent magnetic field. The local and collective environments include both energy conserving and energy exchange interactions with spins. This allows us to determine conditions of applicability of the Bloch equation for describing the evolution of the magnetization. We also consider two (and more) ensembles of spins with different parameters and strengths of interactions with their environments. Using our approach based on resonant perturbation theory [16], we derive explicit expressions for the time evolution of the reduced density matrix elements and, consequently, for the macroscopic magnetization. We explicitly calculate the relevant relaxation and decoherence rates. The obtained results are important for many applications including MRI and for studying collective effects in materials for superconducting qubits.

We point out that while we continue here the development of the rigorous resonance pertrubation theory [16], other approaches to open (spin) systems include the master equation and Keldysh formalisms [25, 26].

Main results of the paper

• *Single spin dynamics.* We consider a microscopic, Hamiltonian model of N spins interacting with local and collective bosonic thermal reservoirs, via energy conserving and energy exchange interactions. In Theorem 2.1 we derive rigorously an expression for the reduced density matrix of a single spin, consisting of a main term describing relaxation and dephasing, plus a remainder term which is small in the couplings *homogeneously* in time.

• *Single spin relaxation.* We show that the single-spin relaxation rate is given by

$$\gamma_{\rm relax} = \frac{1}{4}\coth(\beta\omega/2)\left\{\lambda^2 J_{g_{\rm c}}(\omega) + \mu^2 J_{g_\ell}(\omega)\right\},$$

where ω is the spin frequency, λ and μ are the strengths of the energy exchange collective and local couplings, respectively, and where $J_g(\omega)$ is the reservoir spectral density. (In some publications the spectral density of noise is defined as to include the temperature-dependent factor $\coth(\beta\omega/2)$.) Only energy-exchange couplings contribute to this rate, and the expressions for the contributions of local and collective effects are the same.

• *Single spin dephasing.* We show that the single-spin dephasing rate is given by

$$\gamma_{\rm deph} = \frac{1}{2}\gamma_{\rm relax} + \gamma_{\rm cons} + \gamma',$$

where $\gamma_{\rm cons}$ is a contribution stemming only from the energy conserving local and collective interactions, determined by the spectral density of the reservoir at zero frequency (see (3.3)). The contribution γ' encodes the effect on dephasing of a single spin due to all other spins. It is defined as follows. The time-dependence of the single spin off-diagonal density matrix elements has a very complicated, not exponentially decaying contribution coming from the collective coupling. The term γ' is defined to be the reciprocal of the time by which that quantity is reduced to half its initial value.[1]

The explicit expression of γ' is not simple (see (2.34), (2.32)). For small ratio r between the strengths of the collective to the local couplings we have $\gamma' = O(r^2)$ (independent of the number N of spins). For large collective coupling we have $\gamma' \sim$ const.$\gamma_{\rm relax}$, for a constant not depending on N.

• *Evolution of magnetization.* We consider the spins in a homogeneous magnetic field pointing in the z-direction. We show that the z-component of the total magnetization vector relaxes to its equilibrium value at the single-spin relaxation rate $\gamma_{\rm relax}$. This verifies the correctness of the usual Bloch equation (3.13) for the z-component. The equation for the transverse total magnetic field is given by a modified Bloch equation (3.15), with a time-dependent dephasing time ($T_2 = T_2(t)$) and a time-dependent effective magnetic field. For large times, the coefficients in the modified Bloch equation

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[1] For the purely energy-conserving collective coupling model (only $\varkappa \neq 0$), the quantity in question does not decay but oscillates, and so we associate $\gamma' = 0$ to this situation.

approach stationary values and give rise to the usual Bloch equation with renormalized $T_2(\infty)$ time and renormalized effective magnetic field. We show that

$$\frac{1}{T_2(\infty)} = \frac{1}{2}\gamma_{\rm relax} + \gamma_{\rm cons} + (N-1)\gamma'',$$

where $\gamma'' \geq 0$ is independent of N. [2] For small ratio r between the strengths of the collective to the local couplings we have $\gamma'' = O(r^2)$. Consequently, if $r \sim N^{-1/2}$ (large N) then the collective coupling gives a non-vanishing renormalization to the (asymptotic) T_2 time, while if $r \sim N^{-1/2-\epsilon}$ (any $\epsilon > 0$) or smaller, then no collective effect is visible in the dephasing. An interesting question is what happens for $r \sim N^{-1/2+\epsilon}$ or larger. Then the expression for $T_2(\infty)$ suggests that the collective interaction may decrease the T_2 time drastically for large N. However, this range of interaction parameters is not accessible by our perturbation theory approach, and more work in this direction is required. It is important to note here that in order to derive our rigorous result, Theorem 2.1, we need a strong smallness condition on all coupling constants (see (2.11)). As explained in Section 5, we expect that our result should hold for collective coupling constants up to size $O(N^{-1/2})$ and local coupling constants of size $O(N^0)$ (relative to the spin frequency). However, for $r = O(N^{-1/2+\epsilon})$ we do not think that usual perturbation theory can be applied, and a different approach should be taken.

We also examine the situation where we have two (or more) species of spins, A and B, each species coupled homogeneously to local and collective reservoirs (with a single collective reservoir for both species). We show that the z-component of the magnetization of either species relaxes with single-spin relaxation time (associated to that species). The transverse magnetization dephases following a modified Bloch equation with time-dependent T_2-time and effective magnetic field. For large times, the T_2-time of species A approaches the limiting value

$$\frac{1}{T_{2,A}(\infty)} = \frac{1}{2}\gamma_{{\rm relax},A} + \gamma_{{\rm cons},A} + (N_A-1)\gamma_A + N_B\gamma_B,$$

where N_A and N_B are the number of spins in each class, and γ_A, $\gamma_B \geq 0$. For small ratio r_A, r_B of the collective and local coupling constants, we have $\gamma_A = O(r_A^2)$, $\gamma_B = O(r_B^2)$. The total magnetization is the sum of that of species A and B. It is the sum of two terms decaying (relaxing and dephasing) at different rates, and so we cannot associate to it a total relaxation time or a total dephasing time.

Remarks on N-dependencies. Let L be the correlation length of a physical environment we want to describe. Our model is adequate for a system where $N/\rho_{\rm spins}$ is of the order of L^3 (here, $\rho_{\rm spins}$ is the spin density). This is so since in our Hamiltonian, the collective interaction couples all N effective spins to the *same* bath (see (2.2)). If we increase N then not all spins should interact collectively with a single reservoir, but only the amount corresponding to the given correlation length L. A more subtle Hamiltonian than (2.2) is needed to describe this situation. One may model the finite correlation length of the collective reservoir heuristically (and not entirely accurately)

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[2] For the purely energy-conserving collective coupling model (only $\varkappa \neq 0$), we have $\gamma'' = 0$, as the quantity associated to this decay rate does actually not decay, but oscillates.

by taking the collective coupling parameters $\lambda(N)$ and $\varkappa(N)$ as decreasing functions of N. In this way, the parameter r in the discussion above behaves as $r \sim \varkappa(N)^2$ (N large, $\varkappa(N)$ small) and so a finite renormalization of γ'', determined by $r \sim N^{-1/2}$, suggests a scaling $\varkappa(N) \sim N^{-1/4}$. Note however that if we impose this scaling, then the collective effects in $\gamma_{\rm relax}$ and $\gamma_{\rm deph}$ vanish for large N (since $\varkappa(N) \to 0$).

Remarks on characteristic times. As one can see from the above expressions for the single qubit relaxation ($\gamma_{\rm relax}$) and decoherence ($\gamma_{\rm deph}$) rates there are two kinds of contributions. One comes from interactions of the effective spins with their local environments (coupling constant μ). This part gives the standard expressions for relaxation and decoherence rates. For example, in the case of paramagnetic nuclear systems in liquids the corresponding decay times can be long enough: $1/\gamma_{\rm relax}$, $1/\gamma_{\rm deph}$ of the order of seconds or more, while for superconducting qubits, usually $1/\gamma_{\rm relax}$, $1/\gamma_{\rm deph} \sim 10\mu s$. The collective interactions (coupling constant λ) contribute to the standard expressions for single spin relaxation and decoherence rates as renormalizations, with the corresponding spectral densities of noise. In experiments where the dynamics of collective magnetization is observed, one should expect that the evolution of the z-component of the magnetization will evolve with a renormalized relaxation rate, as for single spins. On the other hand, the evolution of the transversal magnetization includes (i) single spin dephasing rates renormalized by collective effects (as discussed above) and (ii) an additional *collective* term (γ'') proportional to the number of spins, N. (If the collective coupling parameters are scaled correctly with N, see above, then γ'' becomes independent of N.)

The presence of collective effects will be seen in experiments on the evolution of macroscopic magnetization as the renormalizations of the decoherence and relaxation rates. In the case of superconducting qubits one of the main interests related to $1/f$ noise are experiments involving so-called two-level-system environments (TLS). They model various kinds of defects in superconducting materials [27]. Recently scientists have started to discuss possibilities of collective effects provided by TLS. The approach discussed in our paper here indicates possible renormalizations of decoherence and relaxation rates associated with collective effects of TLS environments.

2 Model, single spin dynamics

We consider N non interacting spins 1/2 coupled to local and collective bosonic heat reservoirs. The full Hamiltonian is given by

$$\begin{aligned} H &= -\hbar\sum_{n=1}^N \omega_n S_n^z + \sum_{n=1}^N H_{{\rm R}_n} + H_{\rm R} && (2.1)\\ &\quad + \sum_{n=1}^N \lambda_n S_n^x \otimes \phi_{\rm c}(g_{\rm c}) + \sum_{n=1}^N \varkappa_n S_n^z \otimes \phi_{\rm c}(f_{\rm c}) && (2.2)\\ &\quad + \sum_{n=1}^N \mu_n S_n^x \otimes \phi_n(g_n) + \sum_{n=1}^N \nu_n S_n^z \otimes \phi_n(f_n). && (2.3)\end{aligned}$$

Below we use dimensionless variables and parameters. To do so, we introduce a characteristic frequency, ω_0, typically of the order of spin transition frequency. The total Hamiltonian, energies of spin states, and temperature are measured in units $\hbar\omega_0$. The frequencies of spins, $\omega_n > 0$, bosonic excitations, $\omega(k) = c|\vec{k}|$ (where c is the speed of light), the wave vectors of bosonic excitations are normalized by ω_0/c, and all constants of interactions are measured in units ω_0. A dimensionless time is defined as $t \longrightarrow \omega_0 t$.

In (2.2), (2.3), $\omega_n > 0$ is the frequency of spin n,

$$S^z = \frac{1}{2}\left[\begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array}\right] \qquad \text{and} \qquad S^x = \frac{1}{2}\left[\begin{array}{cc} 0 & 1 \\ 1 & 0 \end{array}\right], \tag{2.4}$$

and $S_n^{z,x}$ denotes the $S^{z,x}$ of spin n. $H_{\rm R}$ is the Hamiltonian of the bosonic collective reservoir,

$$H_{\rm R} = \int_{\mathbb{R}^3} |k| a^*(k) a(k) {\rm d}^3 k, \tag{2.5}$$

and $H_{{\rm R}_n}$ is that same Hamiltonian pertaining to the n-th individual reservoir. For a square-integrable *form factor* $h(k)$, $k \in \mathbb{R}^3$, $\phi(h)$ is given by

$$\phi(h) = \frac{1}{\sqrt{2}} \int_{\mathbb{R}^3} \left\{ h(k) a^*(k) + h(k)^* a(k) \right\} {\rm d}^3 k. \tag{2.6}$$

The real numbers λ_n, $\varkappa_n$, μ_n, ν_n are coupling constants, measuring the strengths of the various interactions as follows:

λ_n	energy exchange collective coupling
$\varkappa_n$	energy conserving collective coupling
μ_n	energy exchange local coupling
ν_n	energy conserving local coupling

We introduce the maximal size of all couplings,

$$\alpha := \max_n \{ |\varkappa_n|, |\lambda_n|, |\mu_n|, |\nu_n| \}. \tag{2.7}$$

The energies of the N uncoupled spins are the eigenvalues of $H_{\vec{S}} = -\sum_{n=1}^N \omega_n S_n^z$, given by $-\frac{1}{2}\sum_{n=1}^N \omega_n \sigma_n$, where $\sigma_n \in \{1, -1\}$. We denote by $\varphi_{\underline{\sigma}} = \varphi_{\sigma_1} \otimes \cdots \otimes \varphi_{\sigma_N}$ the corresponding eigenvector. **Bohr energies** (energy differences) are thus given by

$$e(\underline{\sigma}, \underline{\tau}) = -\frac{1}{2} \sum_{n=1}^N \omega_n (\sigma_n - \tau_n). \tag{2.8}$$

Assumptions.

(A) We consider the spin frequencies $\{\omega_n\}$ to be uncorrelated in the following sense:

$$\textit{If } e(\underline{\sigma}, \underline{\tau}) = e(\underline{\sigma}', \underline{\tau}') \textit{ then } \sigma_n - \tau_n = \sigma_n' - \tau_n' \textit{ for all } n. \tag{2.9}$$

In particular, we do not allow any of the ω_n to be the equal. However, we can describe a *homogeneous magnetic field* within the constraint (2.9) by considering a distribution $\omega_n = \omega + \delta\omega_n$ for some fluctuation $\delta\omega_n$ having, say, uniform distribution in some interval. Then assumption (A) is satisfied almost surely. In view of such fluctuations, relation (2.9) is reasonable from a physical point of view, and its mathematical advantage is that it breaks permutation symmetry and hence reduces the degeneracies of the energies e.

(B) The smallest gap between different Bohr energies of the non-interacting spin energies (2.8) is

$$\Delta = \frac{1}{2} \min_{m_n, m_n'} \left\{ \left| \sum_{j=1}^{N} \omega_n (m_n - m_n') \right| \right\} \backslash \{0\}, \tag{2.10}$$

where the minimum is taken over sequences $m_n, m_n' \in \{-2, 0, 2\}$. As our approach is based on perturbation theory of Bohr energy differences, their displacement under interaction, which is of size $N^2\alpha^2$, should be small relative to Δ,

$$N^2\alpha^2 \ll \Delta. \tag{2.11}$$

For $\omega_n = \omega$ constant, we have $\Delta = \omega$. Hence for a homogeneous magnetic field $\omega_n = \omega + \delta\omega_n$ with small fluctuation $\langle\delta\omega_n\rangle/\omega \ll 1$, we have $\Delta = \omega + O(\langle\delta\omega\rangle)$, and the r.h.s. of (2.11) is independent of N. Condition (2.11) is a serious restriction on the coupling strength for large systems (big N). Our analysis uses this condition in several technical estimates of remainder terms, stemming from perturbation theory (see also [16]). However, it is seen from physical considerations, presented in Section 5, that the true condition should read $\alpha_c^2 N \ll \omega$ and $\alpha_\ell \ll \omega$, where α_c and α_ℓ are the sizes of collective and local coupling constants, and ω is the typical frequency of a spin.

(C) Regularity of form factors: denote by h any of the functions $f_{\rm c}, g_{\rm c}, f_n, g_n$ in the Hamiltonian H. Let $r \geq 0$, $\Sigma \in S^2$ be the spherical coordinates of $\mathbb{R}^3$. Then $h(r, \Sigma) = r^p {\rm e}^{-r^m} h'(\Sigma)$, with $p = -1/2 + n$, $n = 0, 1, 2, \ldots$ and $m = 1, 2$, and where h' is any angular function. (Less restrictive requirements on h are necessary only [16], but they are more technical to describe, so we restrict our attention to h satisfying this condition. The condition is needed in order to guarantee that the technical apparatus of 'spectral deformation' is applicable. This family of form factors contains the usual physical ones [8].)

Given e, (2.8), the number

$$N_0(e) = \{n \ : \ \sigma_n = \tau_n \ \text{ for any } (\underline{\sigma}, \underline{\tau}) \text{ with } e(\underline{\sigma}, \underline{\tau}) = e\} \tag{2.12}$$

depends on e alone, and the number of different configurations $(\underline{\sigma}, \underline{\tau})$ with constant value (2.8) is $2^{N_0(e)}$. There are $2^{N_0(e)}$ elements $\langle \varphi_{\underline{\sigma}}, \rho_{\underline{S}} \varphi_{\underline{\tau}} \rangle$ of the (reduced) density matrix of the spins with fixed value (2.8). As shown in [16], these elements evolve in time jointly, and independently of elements associated with any other value of (2.8). We mention that this system with $N = 2$ spins has been considered in [28] for an analysis of 2-qubit entanglement.

We consider unentangled initial states

$$\rho_0 = \rho_{\mathrm{S}_1} \otimes \cdots \otimes \rho_{\mathrm{S}_N} \otimes \rho_{\mathrm{R}_1} \otimes \cdots \otimes \rho_{\mathrm{R}_N} \otimes \rho_{\mathrm{R}}, \tag{2.13}$$

where ρ_{S_j} are arbitrary single spin states, and ρ_{R_j}, ρ_{R} are thermal equilibrium states of single reservoirs, all at temperature $T = 1/\beta > 0$.

The reduced density matrix $\rho_t^{(j)}$ of spin j is given by

$$\rho_t^{(j)} = \mathrm{Tr}^{(j)}\ \mathrm{e}^{-\mathrm{i}tH} \rho_0 \mathrm{e}^{\mathrm{i}tH},$$

the trace being taken over all spins $n \neq j$ and over all reservoirs.

Let A_j be an observable of the j-th spin, and denote its dynamics by

$$\langle A_j \rangle_t = \mathrm{Tr} \rho_t^{(j)} A_j,$$

where the trace is taken over the space of S_j. Our goal is to find a representation of $\langle A_j \rangle_t$. For a square integrable form factor $h(k) = h(|k|, \Sigma)$ (spherical coordinates of $\mathbb{R}^3$), the **spectral density** of the reservoir associated to h is given by

$$J_h(\omega) = \pi\omega^2 \int_{S^2} |h(\omega, \Sigma)|^2 \mathrm{d}\Sigma.\ ^3 \tag{2.14}$$

Decay rates are given by coupling constants squared times $J_h(\omega)\coth(\beta\omega/2)$ at values ω corresponding to Bohr frequencies of the spin system. Energy-conserving processes are associated with the Bohr frequency $\omega = 0$, and since $\coth(\beta\omega/2) \sim \omega^{-1}$ as $\omega \sim 0$, we introduce

$$\widetilde{J}_h(0) = \lim_{\omega \to 0_+} \frac{J_h(\omega)}{\omega}. \tag{2.15}$$

Define the quantities

$$\begin{aligned}
b_j &= \frac{1}{4} \frac{\mathrm{e}^{\beta\omega_j}}{\mathrm{e}^{\beta\omega_j} - 1} \left\{ \lambda_j^2 J_{g_\mathrm{c}}(\omega_j) + \mu_j^2 J_{g_j}(\omega_j) \right\} && (2.16)\\
c_j &= \mathrm{e}^{-\beta\omega_j} && (2.17)\\
Z_{\beta,j} &= \mathrm{e}^{-\beta\omega_j/2} + \mathrm{e}^{\beta\omega_j/2} && (2.18)\\
X_j &= \frac{1}{8\pi} \int_{\mathbb{R}} \frac{\lambda_j^2 J_{g_\mathrm{c}}(|u|) + \mu_j^2 J_{g_j}(|u|)}{u + \omega_j} \coth(\beta|u|/2) \mathrm{d}u && (2.19)\\
Y_j &= \frac{1}{8} \left\{ \lambda_j^2 J_{g_\mathrm{c}}(\omega_j) + \mu_j^2 J_{g_j}(\omega_j) \right\} \coth(\beta\omega_j/2) + \frac{1}{2\beta} \left\{ \varkappa_j^2 \widetilde{J}_{f_\mathrm{c}}(0) + \nu_j^2 \widetilde{J}_{f_j}(0) \right\} && (2.20)
\end{aligned}$$

We represent the density matrix of a single spin as a 2×2 matrix according to the ordered orthonormal basis $\{\varphi_1 := \varphi_{\sigma=+1}, \varphi_2 := \varphi_{\sigma=-1}\}$ of $\mathbb{C}^2$. With this notation in place we have the following result.

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[3] Let $C_h(t) = \frac{1}{2}[\langle \phi(h) \mathrm{e}^{\mathrm{i}tH_\mathrm{R}} \phi(h) \mathrm{e}^{-\mathrm{i}tH_\mathrm{R}} \rangle_\beta + \langle \mathrm{e}^{\mathrm{i}tH_\mathrm{R}} \phi(h) \mathrm{e}^{-\mathrm{i}tH_\mathrm{R}} \phi(h) \rangle_\beta]$ be the symmetrized correlation function of a reservoir in thermal equilibrium at temperature $T = 1/\beta$, with H_R and $\phi(h)$ given in (2.5) and (2.6). The Fourier transform $\widehat{C}_h(\omega) = \int_0^\infty \mathrm{e}^{-\mathrm{i}\omega t} C(t) \mathrm{d}t$, $\omega \geq 0$, is related to the spectral density by

$$\mathrm{Re}\, \widehat{C}_h(\omega) = J_h(\omega) \coth(\beta\omega/2).$$

Theorem 2.1 (Dynamics of single spin) *For any observable* A_j *of spin* j, $j = 1, \dots, N$, *and* $t \geq 0$, *we have*

$$\begin{aligned} \langle A_j \rangle_t &= Z_{\beta,j}^{-1} \operatorname{Tr} \mathrm{e}^{-\beta H_{\mathrm{S}_j}} A_j && (2.21)\\ &+ \mathrm{e}^{-tb_j(c_j+1)} \left\{ [\rho_0^{(j)}]_{11} - \frac{1}{\mathrm{e}^{-\beta\omega_j}+1} \right\} \big([A_j]_{11} - [A_j]_{22} \big) && (2.22)\\ &+ \mathrm{e}^{\mathrm{i}t(-\omega_j + X_j + \mathrm{i}Y_j)} \mathcal{C}_j(N,t)\ [\rho_0^{(j)}]_{21}\ [A_j]_{12} && (2.23)\\ &+ \overline{\mathrm{e}^{\mathrm{i}t(-\omega_j + X_j + \mathrm{i}Y_j)} \mathcal{C}_j(N,t)\ [\rho_0^{(j)}]_{21}}\ [A_j]_{21} && (2.24)\\ &+ O(\alpha^2), && (2.25) \end{aligned}$$

where the remainder term is uniform in $t \geq 0$.

The quantity $\mathcal{C}_j(N,t)$ involves the interaction parameters and the initial condition of all spins other than j,

$$\begin{aligned} \mathcal{C}_j(N,t) &= \prod_{l \neq j} \left\{ \left[\mathrm{e}^{\mathrm{i}tz_{l,j}^{+}} - \mathrm{e}^{\mathrm{i}tz_{l,j}^{-}} \right] \frac{1 + c_l \alpha_{l,j}}{1 + c_l \alpha_{l,j}^2} \big(\alpha_{l,j} + [\rho_0^{(l)}]_{11} (1 - \alpha_{l,j}) \big) + \mathrm{e}^{\mathrm{i}tz_{l,j}^{-}} \right\} && (2.26)\\ z_{l,j}^{\pm} &= \frac{1}{2} \left\{ \mathrm{i}b_l(1+c_l) \pm \sqrt{-b_l^2(1+c_l)^2 + 4a_{l,j}[a_{l,j} - \mathrm{i}b_l(1-c_l)]} \right\} && (2.27)\\ \alpha_{l,j} &= 1 + \mathrm{i}\frac{z_{l,j}^{+} - a_{l,j}}{b_l c_l} && (2.28) \end{aligned}$$

In (2.27), the square root is the principal value (cut on the negative real axis), b_l, c_l are given in (2.16), (2.17), and

$$a_{l,j} = -\frac{1}{2} \varkappa_l \varkappa_j \int_{\mathbb{R}^3} \frac{|f_{\mathrm{c}}(p)|^2}{|p|} \mathrm{d}^3 p. \tag{2.29}$$

Discussion of the factor $\mathcal{C}_j(N,t)$.

Clearly $\mathcal{C}_j(N,0) = 1$. For vanishing energy-conserving collective coupling of spin j, $\varkappa_j = 0$, we have $\mathcal{C}_j(N,t) = 1$ for all $t \geq 0$. This follows since $\varkappa_j = 0$ implies that for all l: $a_{l,j} = 0$, $z_{l,j}^{+} = \mathrm{i}b_l(1+c_l)$, $z_{l,j}^{-} = 0$ and $\alpha_{l,j} = -1/c_l$, and so (2.26) reduces to 1. This shows that if spin j does not undergo energy-conserving collective interaction, then it evolves independently of all other spins. The collective energy-*exchange* coupling (λ_j) has the same effect as the local energy-exchange coupling (μ_j) up to order $O(\alpha^2)$, see (2.20).

As soon as the collective energy-conserving couplings are switched on, $\varkappa_l \neq 0$, the analysis of $\mathcal{C}_j(N,t)$ is difficult. The factors in the product (2.26) decay with rate at least

$$\gamma_j = \min_{l \neq j} \{ \Im z_{l,j}^{+}, \Im z_{l,j}^{-} \}, \tag{2.30}$$

so for a homogeneous system (each factor the same) we have the estimate

$$|\mathcal{C}(N,t)| \leq C_N \, \mathrm{e}^{-\gamma(N-1)t} \qquad \text{with } \gamma = \gamma_j. \tag{2.31}$$

Of course, $\mathcal{C}_j$ does not depend on j anymore. This estimate says that $\mathcal{C}$ decays in time with rate $\gamma(N-1)$, but we have a prefactor depending on N. We have the upper bound $C_N \leq \mathrm{e}^{(N-1)c'}$, for

$$c' = \ln\left\{2\left|\frac{1+c\alpha}{1+c\alpha^2}(\alpha+[\rho_0]_{11}(1-\alpha)\right|+1\right\} > 0. \tag{2.32}$$

We know that for $t=0$ the true upper bound on $|\mathcal{C}(N,t)|$ corresponds to $C_N = 1$, but this does not mean at all that $|\mathcal{C}(N,t)| \leq \mathrm{e}^{-\gamma(N-1)t}$. The estimate (see (2.30), (2.31))

$$|\mathcal{C}(N,t)| \leq \mathrm{e}^{(N-1)[-\gamma t+c']} \tag{2.33}$$

shows that $|\mathcal{C}|$ decays to 1/2 (half of its initial value) no later than at time $\gamma^{-1}[\frac{\ln 2}{N-1}+c']$. We thus call

$$\gamma' = \gamma\left[\frac{\ln 2}{N-1}+c'\right]^{-1} \qquad (\gamma' \approx \gamma/c' \ \text{ for large } N) \tag{2.34}$$

the decay rate of $|\mathcal{C}(N,t)|$. Let us examine this decay rate in the two cases where

$$r = \frac{|a_l|}{b_l} \sim \frac{\varkappa^2}{\lambda^2+\mu^2} \tag{2.35}$$

is either very small or very close to one, and where we have homogeneous coupling constants (independent of l). (2.35) very small corresponds to the situation where the collective interactions ($\varkappa \approx \lambda$) are much smaller than the local ones ($\mu \approx \nu$). The situation where (2.35) is unity describes very large collective coupling relative to the local ones.

For small collective coupling, $r \approx 0$, we obtain $\gamma' \sim \text{const.} rb(1+c) = \text{const.} r\gamma_{\text{relax}}$, where const. does not depend on N. In the limit $r \longrightarrow 0$ we get $\gamma' = 0$, which is the correct behaviour as we have seen above ($\mathcal{C} = 1$ in this setting, no decay).

For large collective coupling, $r \approx 1$, we obtain $\gamma' \sim \text{const.} b$, where const. does not depend on N and b is given in (2.16).

Comparison to exactly solvable model.

If the spins interact with the reservoirs only through energy conserving channels, then $\lambda_n = \mu_n = 0$ in (2.2), (2.3). This model is *exactly solvable.* By proceeding as in [16] ("Resonance theory of decoherence and thermalization", proof of Proposition 7.4), one finds the following *exact formula* for the evolution of the reduced density matrix element of a single spin. For simplicity of notation, we take all $\varkappa_n$ to be constant $\varkappa_c$ (collective) and all ν_n constant ν_ℓ (local). We also take all local form factors equal (f_ℓ) and all collective ones too (f_c). Then we find

$$[\rho_t^{(j)}]_{21} = [\rho_0^{(j)}]_{21}\ \mathrm{e}^{-\mathrm{i}\omega_j t}\ \mathrm{e}^{-\nu_\ell^2\Gamma_\ell(t)-\varkappa_c^2\Gamma_c(t)} \sum_{\sigma_k,\, k\neq j}\ \prod_{l,\, l\neq j}^{N} [\rho_0^{(l)}]_{\sigma_k\sigma_k}\mathrm{e}^{-2\mathrm{i}\sigma_k\varkappa_c^2 S_c(t)}. \tag{2.36}$$

The sum is over $\sigma_k = \pm 1/2$, $k = 1,\ldots,N$, $k \neq j$, where $\sigma_j = 1/2$ corresponds to the energy eigenstate $\varphi_1 = [1\ 0]^T$ of S^z (see (2.4)). The decoherence functions and Lamb shift are given by

$$\Gamma(t) = \int_{\mathbb{R}^3} |f(k)|^2 \coth(\beta|k|/2) \frac{\sin^2(|k|t/2)}{|k|^2} \mathrm{d}^3 k \tag{2.37}$$

$$S(t) = -\frac{1}{2} \int_{\mathbb{R}^3} |f(k)|^2 \frac{|k|t - \sin(|k|t)}{|k|^2} \mathrm{d}^3 k. \tag{2.38}$$

Of course, the populations are time-independent in this model, $[\rho_t^{(j)}]_{ll} = [\rho_0^{(j)}]_{ll}$ for all $t \geq 0$, $l = 1, 2$. The time-dependence in the exponentials in (2.36) becomes linear for large times, $\Gamma(t) \to t\widetilde{J}(0)/2\beta$ and $\varkappa_c^2 S(t) \to ta$ as $t \to \infty$, where $\widetilde{J}(0)$ and a are given in (2.15) and (2.29) with $\varkappa_l$ replaced by $\varkappa_c$ (see [16]).

It is not hard to see that upon the replacements $\Gamma(t) \mapsto t\widetilde{J}(0)$ and $\varkappa_c^2 S(t) \mapsto ta$ the exact formula (2.36) coincides precisely with expression (2.21)-(2.24) for A s.t. $[A]_{11} = [A]_{22} = [A]_{21} = 0$, $[A]_{12} = 1$ (so that $\langle A \rangle_t = [\rho_t]_{21}$). The factor $\mathcal{C}_j(N,t) = \mathcal{C}(N,t)$ is thus identified with the sum of the product in (2.36). If all spins are initially in the same state, characterized by the population probability $0 \leq p \leq 1$ for the state with $\sigma = 1/2$, we obtain

$$\mathcal{C}(N,t) = \left[p\mathrm{e}^{-\mathrm{i}at} + (1-p)\mathrm{e}^{\mathrm{i}at}\right]^{N-1}, \qquad \text{with } a \text{ given in (2.29).}$$

Clearly $|\mathcal{C}(N,t)| \leq 1$ for all times and all N. Also, for all $n \in \mathbb{Z}$, we have $|\mathcal{C}(N, n\pi a^{-1})| = 1$ and $|\mathcal{C}(N, (n+\frac{1}{2})\pi a^{-1})| = |1-2p|^{N-1} \approx 0$ for N large and $p \neq 0, 1$. Therefore the factor $\mathcal{C}(N,t)$ oscillates in size between zero and one, with frequency $|a|/\pi$ proportional to the square of the energy-conserving collective coupling $\varkappa_c^2$.

3 Evolution of single spins and of magnetization

3.1 Single spin relaxation and dephasing times

The term on the r.h.s. of (2.21) is the equilibrium average at temperature $T = 1/\beta$. From (2.22) we obtain the relaxation rate of spin j, namely $\gamma_{\mathrm{relax},j} = b_j(c_j + 1)$. The single spin relaxation rate is

$$\gamma_{\mathrm{relax},j} = 1/\tau_{\mathrm{relax},j} = \frac{1}{4} \coth(\beta\omega_j/2) \left\{ \lambda_j^2 J_{g_\mathrm{c}}(\omega_j) + \mu_j^2 J_{g_j}(\omega_j) \right\}. \tag{3.1}$$

The single-spin relaxation time depends on the local (μ_j) and collective (λ_j) couplings in the same manner: *In the relaxation process, the collective reservoir acts as a local reservoir.*

Next we consider the dephasing time determined by (2.23), (2.24). There are two contributions to the time decay. One comes from spin j itself and is given by Y_j, the other one comes from all other spins than j and is given by $\mathcal{C}_j$. One sees from (2.27) that $\Im z_{l,j}^{\pm} \geq 0$, and that $\min\{\Im z_{l,j}^+, \Im z_{l,j}^-\} = 0 \Leftrightarrow a_{l,j} b_l = 0$. It follows that if the energy conserving collective couplings and at least one of the energy-exchange couplings (local or collective) do not vanish (so that $a_{l,j} b_l \neq 0$), then we have (2.33) with $\gamma > 0$. The single-spin dephasing rate is thus $Y_j + \gamma'$ which we can write as

$$\gamma_{\mathrm{deph},j} = \frac{1}{2}\gamma_{\mathrm{relax},j} + \gamma_{\mathrm{cons},j} + \gamma', \tag{3.2}$$

where

$$\gamma_{\text{cons},j} = \frac{1}{2\beta}\{\varkappa_j^2 \widetilde{J}_{f_c}(0) + \nu_j^2 \widetilde{J}_{f_j}(0)\} \tag{3.3}$$

is a contribution coming purely from the energy-conserving interactions, in which the local and collective couplings play the same role. The last term in expression (3.2) is due to the presence of the $N-1$ spins other than the considered one. As we have seen after (2.35), if the collective coupling is small ($r \approx 0$), then $\gamma' \sim r\gamma_{\text{relax}} << \gamma_{\text{relax}}$ and hence the last term in (3.2) is negligible. If the collective coupling is large ($r \approx 1$), then $\gamma' \sim b \sim \gamma_{\text{relax,j}}$.

Conclusions. • The single-spin relaxation rate is the sum of two contributions from the local and the collective energy-exchange interactions (3.1). The collective term has the same form as the local term, and the presence of all other spins does not influence the single spin relaxation rate.

• The single-spin dephasing rate has three contributions (3.2). One is half the relaxation rate (exchange interactions), one comes from energy conserving interactions (local and collective), and a third term which is due to the presence of all other spins. That last term (γ') is negligible for small collective coupling, and renormalizes the dephasing rate for strong collective couplings by an amount independent of the number of spins.

3.2 Evolution of magnetization

Let

$$\vec{S} = \begin{bmatrix} S^x \\ S^y \\ S^z \end{bmatrix}$$

be the total magnetization vector, where $S^{x,y,z} = \sum_{j=1}^N S_j^{x,y,z}$. It is convenient to introduce the complex (non-hermitian) observable

$$S_j^- = S_j^x - \mathrm{i}S_j^y.$$

We use Theorem 2.1 with $A_j = S_j^{x,y,z}$ to obtain

$$\begin{aligned} \langle S_j^z\rangle_t &= \frac{1}{2}\tanh(\beta\omega_j/2)[1-\mathrm{e}^{-t/\tau_{\text{relax},j}}] + \mathrm{e}^{-t/\tau_{\text{relax},j}}\langle S_j^z\rangle_0 + O(\alpha^2), & (3.4)\\ \langle S_j^-\rangle_t &= \mathrm{e}^{\mathrm{i}t(-\omega_j+X_j+\mathrm{i}Y_j)}\mathcal{C}_j(N,t)\langle S_j^-\rangle_0 + O(\alpha^2). & (3.5)\end{aligned}$$

Purely local coupling. In the absence of collective coupling ($\lambda_n = 0 = \varkappa_n$), the above equations simplify to

$$\begin{aligned} \langle S_j^z\rangle_t &= \frac{1}{2}\tanh(\beta\omega_j/2)[1-\mathrm{e}^{-t/\tau_{\text{relax},j}}] + \mathrm{e}^{-t/\tau_{\text{relax},j}}\langle S_j^z\rangle_0 + O(\alpha^2), & (3.6)\\ \langle S_j^-\rangle_t &= \mathrm{e}^{\mathrm{i}t(-\omega_j+X_j+\mathrm{i}Y_j)}\langle S_j^-\rangle_0 + O(\alpha^2). & (3.7)\end{aligned}$$

where $\tau_{\text{relax},j}$ is given by $1/\gamma_{\text{relax},j}$, see (3.1) with $\lambda_j = 0$. Also, X_j, Y_j are given in (2.19), (2.20) with $\lambda_j = \mu_j = 0$. The factor $\mathcal{C}_j(N,t)$ equals 1 (as discussed after (2.29)).

3.2.1 Homogeneous magnetic field

In this section we derive the evolution of the magnetization vector in a homogeneous magnetic field, characterized by $\omega_j = \omega + \delta\omega_j$ with $\delta\omega_j \to 0$ (see also assumption (B) after (2.11)). This is the description of an elementary volume of many spins sitting in a magnetic field with gradient much smaller than the size of the elementary volume.

We consider all spins initially in the same state. We take all local couplings to be the same, i.e., $g_j = g_\ell$ and all collective couplings to be the same, $f_j = f_{\rm c}$, and all coupling constants independent of j. In this limit, we have in formulas (3.4), (3.5)

$$\omega_j = \omega, \quad X_j = X, \quad Y_j = Y,$$

where $\tau_{\rm relax}$, X, Y are given in (3.1), (2.19), (2.20), with ω_j, f_j, g_j and all coupling constants replaced by their constant values, in particular,

$$\gamma_{\rm relax} = 1/\tau_{\rm relax} = \frac{1}{4}\coth(\beta\omega/2)\left\{\lambda^2 J_{g_{\rm c}}(\omega) + \mu^2 J_{g_\ell}(\omega)\right\}. \tag{3.8}$$

Furthermore, we have $\mathcal{C}_j(N,t) = \mathcal{C}(N,t)$, with (see (2.26))

$$\begin{aligned} \mathcal{C}(N,t) &= [\mathcal{D}(t)]^{N-1} & (3.9)\\ \mathcal{D}(t) &= \left[\mathrm{e}^{\mathrm{i}tz^+} - \mathrm{e}^{\mathrm{i}tz^-}\right]\frac{1+c\alpha}{1+c\alpha^2}\big(\alpha + [\rho_0]_{11}(1-\alpha)\big) + \mathrm{e}^{\mathrm{i}tz^-}. & (3.10)\end{aligned}$$

We sum equations (3.4) and (3.5) over j to obtain (dropping the $O(\alpha^2)$ terms)

$$\begin{aligned} \langle S^z\rangle_t &= \frac{N}{2}\tanh(\beta\omega/2)[1-\mathrm{e}^{-t/\tau_{\rm relax}}] + \mathrm{e}^{-t/\tau_{\rm relax}}\,\langle S^z\rangle_0 & (3.11)\\ \langle S^-\rangle_t &= \mathrm{e}^{\mathrm{i}t(-\omega+X+\mathrm{i}Y)}[\mathcal{D}(t)]^{N-1}\langle S^-\rangle_0. & (3.12)\end{aligned}$$

It is clear that (3.11) is the integrated version of the Bloch equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle S^z\rangle_t = -\frac{1}{\tau_{\rm relax}}\big[\langle S^z_j\rangle_t - \frac{N}{2}\tanh(\beta\omega/2)\big] \tag{3.13}$$

corresponding to the homogeneous magnetic field $\vec{B} = B_z\vec{e}_z = -\omega\vec{e}_z$, with relaxation time

$$T_1 = \tau_{\rm relax} = 1/\gamma_{\rm relax},$$

see (3.8). The Bloch equation for the *transverse magnetization* would read

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle S^-\rangle_t = -\frac{1}{T_2}\langle S^-\rangle_t + \mathrm{i}B_z\langle S^-\rangle_t. \tag{3.14}$$

However the true evolution, (3.12), is not of this form. By differentiating (3.12) we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle S^-\rangle_t = -\Gamma(t)\langle S^-\rangle_t + \mathrm{i}B(t)\langle S^-\rangle_t, \tag{3.15}$$

with

$$\Gamma(t) \;=\; \frac{1}{2}\gamma_{\rm relax} + \gamma_{\rm cons} - (N-1)\,{\rm Re}\frac{\rm d}{{\rm d}t}\ln\mathcal{D}(t), \tag{3.16}$$

$$B(t) \;=\; -\omega + X + (N-1)\,{\rm Im}\frac{\rm d}{{\rm d}t}\ln\mathcal{D}(t), \tag{3.17}$$

where $\gamma_{\rm relax}$ is the single-spin relaxation rate (3.1) and $\gamma_{\rm cons}$ is the single-spin dephasing rate due to the energy-conserving interactions (3.3).

Comparing (3.15) with (3.14) leads us to the identification of a *time-dependent dephasing time* $T_2 = 1/\Gamma(t)$ and *a time-dependent effective magnetic field* $B_z = B(t)$.

The deviation of the true equation of evolution from the Bloch equation is given by the terms ${\rm d}/{\rm d}t\ln\mathcal{D}(t)$ in (3.16), (3.17). We now estimate the size of this term for weak collective coupling, where r is small, see (2.35). It is not hard to see that

$$\left|\frac{\dot{\mathcal{D}}(t)}{\mathcal{D}(t)}\right| \leq C|r|, \qquad \lim_{t\to\infty}\frac{\dot{\mathcal{D}}(t)}{\mathcal{D}(t)} = {\rm i}z^- = {\rm i}br\tanh(\beta\omega/2) + O(r^2), \tag{3.18}$$

for a constant C independent of t (and N).

Conclusions. The Bloch equation for the total magnetization (homogeneous magnetic field) holds with relaxation time T_1 given by the single-spin relaxation rate (3.1) (no influence of the other spins). The total magnetization dephases with a time-dependent T_2-time, $T_2 = T_2(t)$. We have $1/T_2(t) = \frac{1}{2}\gamma_{\rm relax} + \gamma_{\rm cons} + (N-1){\rm Re}\frac{d}{dt}\ln\mathcal{D}(t)$, see also (3.1), (3.3). The time-dependent part $\frac{d}{dt}\ln\mathcal{D}(t)$ stems from the collective interaction. For weak collective interaction, r small (see (2.35)), we have $|\frac{d}{dt}\ln\mathcal{D}(t)| \leq C|r|$ (all times).

The term $(N-1){\rm Re}\frac{\rm d}{{\rm d}t}\mathcal{D}(t)$ in (3.16) is $O(Nr^2)$ for large times, see (3.18). If r is of the order $1/\sqrt{N}$ then this is of order one, and the collective interaction gives time-dependent modification of the dephasing time T_2 with an asymptotically renormalized value

$$1/T_2(\infty) = \frac{1}{2}\gamma_{\rm relax} + \gamma_{\rm cons} + (N-1){\rm Im}z^-, \qquad {\rm Im}z^- = O(r^2).$$

If r is smaller, of the order $N^{-1/2-\epsilon}$ (any $\epsilon > 0$) then the collective interaction has no effect in (3.16), (3.17) and the Bloch equation for transversal relaxation holds with T_2 the single-spin dephasing time $[\gamma_{\rm relax}/2 + \gamma_{\rm cons}]^{-1}$. For larger collective interaction we may get large corrections to the Bloch equation, since the last terms in (3.16), (3.17) may become large (big N). This regime does not enter the present perturbative setup, and more work on this issue is needed.

Note that in any event, since ${\rm Im}z^- \geq 0$, the collective interactions can only accelerate the dephasing process.

3.2.2 Multi-species inhomogeneity

Consider the situation where N spins are grouped into two (or more) classes A and B. We describe the situation where within each class, the spins are homogeneous. We have two magnetic fields ω_A, ω_B, two sets of coupling constants (λ_A, λ_B etc), two sets of form factors ($g_{\mathrm{c},A}$, $g_{\mathrm{c},B}$, g_A, g_B etc). Let N_A and N_B be the relative sizes,

$$N_A + N_B = N.$$

If spin j belongs to class A, then (2.26) becomes

$$\mathcal{C}_j(N,t) = [\mathcal{D}_A(t)]^{N_A-1}\,[\mathcal{D}_B(t)]^{N_B}\,, \tag{3.19}$$

with $\mathcal{D}_A(t), \mathcal{D}_B(t)$ given as in (3.10) for species A, B. Let

$$\vec{S}_A = \sum_{j\ \text{in class}\ A} \vec{S}_j$$

and, correspondingly, for the three components of this vector. We sum (3.4) and (3.5) over all indices of spins belonging to class A to obtain

$$\langle S^z_A\rangle_t = \frac{N_A}{2}\tanh(\beta\omega_A/2)[1-\mathrm{e}^{-t/\tau_{\mathrm{relax},A}}] + \mathrm{e}^{-t/\tau_{\mathrm{relax},A}}\langle S^z_A\rangle_0 + O(\alpha^2) \tag{3.20}$$

$$\langle S^-_A\rangle_t = \mathrm{e}^{\mathrm{i}t(-\omega_A+X_A+\mathrm{i}Y_A)}[\mathcal{D}_A(t)]^{N_A-1}[\mathcal{D}_B(t)]^{N_B}\langle S^-_A\rangle_0 + O(\alpha^2). \tag{3.21}$$

Hence class A relaxes with single-spin relaxation time $\tau_{\mathrm{relax},A}$ according to the usual Bloch equation (3.18). For the transverse magnetization we obtain again a modified Bloch equation with time-dependent relaxation time and effective magnetic field,

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle S^-_A\rangle_t = -\Gamma_A(t)\langle S^-_A\rangle_t + \mathrm{i}B_A(t)\langle S^-_A\rangle_t + O(\alpha^2), \tag{3.22}$$

with

$$\Gamma_A(t) = \frac{1}{2}\gamma_{\mathrm{relax},A} + \gamma_{\mathrm{cons},A} - (N_A-1)\,\mathrm{Re}\frac{\mathrm{d}}{\mathrm{d}t}\ln\mathcal{D}_A(t) - N_B\,\mathrm{Re}\frac{\mathrm{d}}{\mathrm{d}t}\ln\mathcal{D}_B(t) \tag{3.23}$$

$$B_A(t) = -\omega_A + X_A + (N_A-1)\,\mathrm{Im}\frac{\mathrm{d}}{\mathrm{d}t}\ln\mathcal{D}_A(t) + N_B\mathrm{Im}\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{D}_B(t). \tag{3.24}$$

As in the previous paragraph, we see that for weak collective coupling and large times, $\Gamma_A(t)$ converges to $\frac{1}{2}\gamma_{\mathrm{relax},A} + \gamma_{\mathrm{cons},A} + (N_A-1)\mathrm{Im}z^-_A + N_B\mathrm{Im}z^-_B$, and $B_A(t)$ converges to $-\omega_A + X_A + (N_A-1)\mathrm{Im}z^-_A$. We thus obtain the (asymptotic) dephasing rate for species A,

$$\gamma_{\mathrm{deph},A}(\infty) = \frac{1}{2}\gamma_{\mathrm{relax},A} + \gamma_{\mathrm{cons},A} + (N_A-1)\mathrm{Im}z^-_A + N_B\mathrm{Im}z^-_B.\,^{4} \tag{3.25}$$

---

[4] A straightforward generalization to s species $A_1,\ldots,A_s$ with sizes $N_{A_1}+\cdots+N_{A_s}=N$ gives the transverse relaxation (i.e., dephasing) rates

$$\gamma_{\mathrm{deph},A_j}(\infty) = \frac{1}{2}\gamma_{\mathrm{relax},A_j} + \gamma_{\mathrm{cons},A_j} + (N_{A_j}-1)\mathrm{Im}z_{A_j} + \sum_{k\neq j} N_{A_k}\mathrm{Im}z_{A_k},$$

for $j=1,\ldots,s$. We conclude that the relaxation rate of each species is a single-spin relaxation rate, while the dephasing contains collective effects. In particular, the dephasing rate of class A_j depends on all other classes.

Recall again that for small collective interaction, $\mathrm{Im} z^-_{A,B} = O(r^2_{A,B})$, (2.35).

Conclusions. The z-component of the total magnetization of each species A and B evolves according to the Bloch equation (3.13) with single-spin relaxation rates $\gamma_{\mathrm{relax},A}$ and $\gamma_{\mathrm{relax},B}$, (3.1).

The transverse total magnetization of species A evolves according to a modified Bloch equation (3.22) (similarly for B). The dephasing time becomes time-dependent (3.23), and takes the value $T_{2,A}(\infty) = 1/\gamma_{\mathrm{deph},A}(\infty)$, (3.25) for large times and small collective coupling.

The total magnetization is the sum of that of species A and B, $\langle S\rangle_t = \langle S_A\rangle_t + \langle S_B\rangle_t$. The z-component relaxes as a sum of two exponentially decaying quantities with different rates (corresponding to A and B). Therefore we cannot associate to it a total a single decay rate.

The total transverse magnetization is the sum of that of species A and B. Each contribution evolves according to the modified Bloch equation. For large times, the dephasing time approaches a renormalized constant value. Being again a sum of two terms decaying at different rates, the total transverse magnetization does not have a single decay rate.

4 Proof of Theorem 2.1

We set $j = 1$ in this proof (the case of general j is obtained merely by a change in notation). To alleviate notation, we will not keep the index $j = 1$ in the expressions in this Section. For example, we simply write $z^\pm_l$ instead of $z^\pm_{l,1}$ (see (2.27)), a_l instead of $a_{l,1}$ (see (2.29)), and so forth. In this section, we present technical details only to a degree necessary for the proof of Theorem 2.1. More background and detail is found in Appendix A.

Following the method developed in [16], the dynamics of A_1 is represented as

$$\langle A_1\rangle_t = \big\langle \psi_0, B_1\cdots B_N \mathrm{e}^{\mathrm{i}tK} A_1 \Omega_{\vec{S}} \otimes \Omega_{\vec{R}}\big\rangle . \tag{4.1}$$

The scalar product on the r.h.s. is that of the GNS Hilbert space, see (A.6) ("doubled space"). Here, $\Omega_{\vec{S}} = \Omega_{\mathrm{S}_1}\otimes\cdots\otimes\Omega_{\mathrm{S}_N}$, $\Omega_{\vec{R}} = \Omega_{\mathrm{R}_1}\otimes\cdots\otimes\Omega_{\mathrm{R}_N}\otimes\Omega_{\mathrm{R}}$ and Ω_{S_k} is the trace state of spin k, $\langle \Omega_{\mathrm{S}_k}, (A\otimes\mathbb{1})\Omega_{\mathrm{S}_k}\rangle = \frac12([A]_{11}+[A]_{22})$ given explicitly by

$$\Omega_{S_k} = \frac{1}{\sqrt{2}}\big(\varphi_{11}+\varphi_{22}\big), \tag{4.2}$$

where $\varphi_{ij} = \varphi_i\otimes\varphi_j$, and (recall the convention after (2.20)) $\varphi_1 = \varphi_{\sigma=+1}$, $\varphi_2 = \varphi_{\sigma=-1}$. The link between the initial density matrices ρ_S in (2.13) and the corresponding representing vector $\psi_S \in \mathbb{C}^2\otimes\mathbb{C}^2$ is provided by the GNS construction: $\mathrm{Tr}(\rho_S A) = \langle\psi_S, (A\otimes\mathbb{1})\psi_S\rangle$ (see e.g. [31]).

The vectors Ω_{R_k} are reservoir equilibrium states at temperature $T = 1/\beta$ (see Appendix A).

The vector ψ_0 in (4.1) is the initial state of the entire system, it is of the form $\psi_0 = \psi_{S_1}\otimes\cdots\psi_{S_N}\otimes\Omega_{\vec{R}}$, where ψ_{S_k} represents the initial state of spin k (see also

(2.13)). The B_k are unique operators (in the commutant of the algebra of observables of spin k) satisfying

$$\psi_{\mathrm{S}_k} = B_k \Omega_{\mathrm{S}_k}. \tag{4.3}$$

The existence of such unique B_k is a property of Ω_{S_k} called "cyclicity" [31].

The operator K is the Liouville operator acting on all spins and all reservoirs, satisfying

$$K\Omega_{\vec{S}} \otimes \Omega_{\vec{R}} = 0. \tag{4.4}$$

Its explicit form is rather involved, see Appendix A and [16]. For the purposes in this section, it suffices to know the representation

$$P_{\vec{R}} \mathrm{e}^{\mathrm{i}tK} P_{\vec{R}} = \sum_{e,s} \mathrm{e}^{\mathrm{i}t\varepsilon_e^{(s)}} Q_e^{(s)} + O(\alpha^2 \mathrm{e}^{-\gamma t}), \tag{4.5}$$

where $P_{\vec{R}} = |\Omega_{\vec{R}}\rangle\langle\Omega_{\vec{R}}|$ projects out all degrees of freedom of the reservoirs. The sum runs over all e of the form (2.8), i.e., eigenvalues of the operator

$$L_{\vec{S}} = H_{\vec{S}} \otimes \mathbb{1}_{\vec{S}} - \mathbb{1}_{\vec{S}} \otimes H_{\vec{S}} \tag{4.6}$$

acting on $\mathbb{C}^{2N} \otimes \mathbb{C}^{2N}$ (which are also the eigenvalues of K with $\alpha = 0$). Here, we have set $H_{\vec{S}} = -\sum_{n=1}^N \omega_n S_n^z$. For each e fixed, s indexes its splitting into $\varepsilon_e^{(s)}$, $1 \le s \le \mathrm{mult}(e)$, as an eigenvalue of K, under the perturbation (2.2) plus (2.3).[5] We have $\varepsilon_e^{(s)} \neq \varepsilon_e^{(s')}$ unless $s = s'$. The $Q_e^{(s)}$ are the (not orthogonal) spectral projections of K, and $\gamma > 0$ satisfies $0 \le \Im\varepsilon_e^{(s)} < 2\gamma < T$ (temperature).

We now describe the perturbation expansion in α of $\varepsilon_e^{(s)}$ and $Q_e^{(s)}$. Due to Assumption (2.9) the eigenspace of $L_{\vec{S}}$ associated to an eigenvalue e is obtained as follows. Associated to e are unique indices $1 \le j_1 < j_2 < \cdots < j_{N_0(e)} \le N$ (recall (2.12)) satisfying $\sigma_j = \tau_j \Leftrightarrow j \in \{j_1, \ldots, j_{N_0(e)}\}$ for any $(\underline{\sigma}, \underline{\tau})$ with $e(\underline{\sigma}, \underline{\tau}) = e$. Let $\underline{\varrho} = (\varrho_1, \ldots, \varrho_{N_0(e)}) \in \{-1, +1\}^{N_0(e)}$ and define vectors in $\mathbb{C}^2$ by

$$\xi_j^{\varrho_j} = \xi_j^{\pm} = \begin{bmatrix} 1 \\ \alpha_j^{\pm} \end{bmatrix}, \tag{4.7}$$

$$\widetilde{\xi}_j^{\varrho_j} = \widetilde{\xi}_j^{\pm} = \frac{1}{1 + c_j[(\alpha_j^{\pm})^*]^2} \begin{bmatrix} 1 \\ c_j(\alpha_j^{\pm})^* \end{bmatrix}, \tag{4.8}$$

according to whether $\varrho_j = \pm 1$. Here, c_j is given in (2.17), $\alpha_j^{\pm}$ is given in (2.28) with a_j, b_j, $z_j^{\pm}$ from (2.29), (2.16) and (2.27).

Given $e, \underline{\varrho}$, set

$$\eta_e^{(\underline{\varrho})} = \varphi_{\sigma_1,\tau_1} \otimes \cdots \otimes \xi_{j_1}^{\varrho_1} \otimes \cdots \otimes \xi_{j_{N_0(e)}}^{\varrho_{N_0(e)}} \otimes \cdots \otimes \varphi_{\sigma_N,\tau_N}, \tag{4.9}$$

$$\widetilde{\eta}_e^{(\underline{\varrho})} = \varphi_{\sigma_1,\tau_1} \otimes \cdots \otimes \widetilde{\xi}_{j_1}^{\varrho_1} \otimes \cdots \otimes \widetilde{\xi}_{j_{N_0(e)}}^{\varrho_{N_0(e)}} \otimes \cdots \otimes \varphi_{\sigma_N,\tau_N}, \tag{4.10}$$

---

[5] To be more precise, one has to use a 'spectral deformation' of the operator K in this argument, see also Appendix A and [16], but the deformation does not influence the physical results.

where at locations j_k, we replace $\varphi_{\sigma_k,\tau_k}$ by ξ (or $\widetilde{\xi}$) with the appropriate value of ϱ_k.

Let h be a form factor. We define

$$\mathcal{G}_h(u) = \int_{S^2} |h(u,\Sigma)|^2 \mathrm{d}\Sigma, \qquad \text{and} \qquad \gamma_+(h) = \lim_{u\to 0_+} u\mathcal{G}_h(u). \tag{4.11}$$

Let $\{h_n\}$ and $\{\alpha_n\}$ be form factors and coupling constants, respectively. For an eigenvalue e as in (2.8), set

$$\begin{aligned}
x_e(\{\alpha_n\},\{h_n\}) &= -\frac{1}{8}\sum_{\{n:\sigma_n\neq\tau_n\}} \alpha_n^2\sigma_n \int_{\mathbb{R}} u^2 \frac{\mathcal{G}_{h_n}(|u|)}{u+\omega_n}\coth(\beta|u|/2)\mathrm{d}u && (4.12)\\
y_e(\{\alpha_n\},\{h_n\}) &= \frac{\pi}{8}\sum_{\{n:\sigma_n\neq\tau_n\}} \alpha_n^2(\omega_n)^2\ \mathcal{G}_{h_n}(\omega_n)\coth(\beta\omega_n/2), && (4.13)\\
y'_e &= \frac{\pi}{2\beta}\sum_{\{n:\sigma_n\neq\tau_n\}} \nu_n^2\ \gamma_+(f_n), && (4.14)\\
y''_e &= \frac{\pi}{8\beta}\gamma_+(f_{\mathrm{c}})[e_0(e)]^2, && (4.15)\\
e_0(e) &= \sum_{\{n:\sigma_n\neq\tau_n\}} \varkappa_n\,(\sigma_n-\tau_n). && (4.16)
\end{aligned}$$

Note that the indices over which the sums are taken are the same for any pair of spin configurations $(\underline{\sigma},\underline{\tau})$ with $e(\underline{\sigma},\underline{\tau}) = e$. Furthermore, we define

$$\begin{aligned}
X_e &= x_e(\{\lambda_n\}, g_{\mathrm{c}}) + x_e(\{\mu_n\},\{g_n\}), && (4.17)\\
Y_e &= y''_e + y'_e + y_e(\{\lambda_n\}, g_{\mathrm{c}}) + y_e(\{\mu_n\},\{g_n\}). && (4.18)
\end{aligned}$$

Then we have:

Proposition 4.1 *Suppose that the numbers* $e+\delta_e^{(\underline{\varrho})}$, *where*

$$\delta_e^{(\underline{\varrho})} = X_e + \mathrm{i}Y_e + \sum_{k=1}^{N_0(e)} z_{j_k}^{\varrho_k}, \tag{4.19}$$

are distinct for all e *and all* $\underline{\varrho}$. *Here,* $z_{j_k}^{\pm} = z_{j_k}^{\pm}(e)$ *is given by the r.h.s. of (2.27), with* $l = j_k$ *and* $a_{l,j}$ *replaced by* $-\frac{1}{2}\varkappa_l e_0(e)\int_{\mathbb{R}^3}|f_c(p)|^2/|p|\mathrm{d}^3p$ *(see (2.29)). Then, for nonzero, small* α, *the eigenvalues of (the spectrally deformed)* K *are all simple and have the expansion*

$$\varepsilon_e^{(\underline{\varrho})} = e + \delta_e^{(\underline{\varrho})} + O(\alpha^4) \tag{4.20}$$

with corresponding eigenprojection

$$Q_e^{(\underline{\varrho})} = |\eta_e^{(\underline{\varrho})}\rangle\langle\widetilde{\eta}_e^{(\underline{\varrho})}| + O(\alpha^2). \tag{4.21}$$

We give a proof of the proposition in Section 4.1. Combining the result of the Proposition with (4.1) and (4.5) gives

$$\langle A_1\rangle_t = \sum_e \sum_{\underline{\varrho}\in\{\pm1\}^{N_0(e)}} \mathrm{e}^{\mathrm{i}t\varepsilon_e^{(\underline{\varrho})}} \left\langle \psi_{\mathrm{S}_1}\cdots\psi_{\mathrm{S}_N}, B_1\cdots B_N(|\eta_e^{(\underline{\varrho})}\rangle\langle\widetilde{\eta}_e^{(\underline{\varrho})}|)A_1\Omega_{\vec{S}}\right\rangle + O(\alpha^2), \tag{4.22}$$

with a remainder term uniformly bounded in $t \geq 0$. Since $\widetilde{\eta}_e^{(\varrho)}$ belongs to the range of the spectral projection $P(L_{\vec{S}} = e)$, and since

$$A_1\Omega_{\vec{S}} = P(L_{\mathrm{S}_2} = \cdots = L_{\mathrm{S}_N} = 0)A_1\Omega_{\vec{S}}, \tag{4.23}$$

only the terms $e \in \mathrm{spec}(L_1) = \{-\omega_1, 0, 0, \omega_1\}$ in the sum in (4.22) contribute.

Let us first consider $e = -\omega_1$. We have $N_0(\omega_1) = N - 1$,

$$\eta_{-\omega_1}^{(\varrho)} = \varphi_{+-} \otimes \xi_2^{\varrho_2} \otimes \cdots \otimes \xi_N^{\varrho_N} \quad \text{and} \quad \widetilde{\eta}_{-\omega_1}^{(\varrho)} = \varphi_{+-} \otimes \widetilde{\xi}_2^{\varrho_2} \otimes \cdots \otimes \widetilde{\xi}_N^{\varrho_N}. \tag{4.24}$$

The term with $e = -\omega_1$ in (4.22) equals

$$\sum_{\varrho_2,\ldots,\varrho_N \in \{\pm 1\}} \mathrm{e}^{\mathrm{i}t[-\omega_1 + X_{-\omega_1} + \mathrm{i}Y_{-\omega_1} + \sum_{j=2}^N z_j^{\varrho_j} + O(\alpha^4)]} \tag{4.25}$$

$$\times [\rho_0^{(1)}]_{21}[A_1]_{12} \prod_{j=2}^N \left\langle \psi_{\mathrm{S}_j}, B_j \xi_j^{\varrho_j} \right\rangle \left\langle \widetilde{\xi}_j^{\varrho_j}, \Omega_{\mathrm{S}_j} \right\rangle \tag{4.26}$$

$$= \quad \mathrm{e}^{\mathrm{i}t(-\omega_1 + X_1 + \mathrm{i}Y_1 + O(\alpha^4))} \mathcal{C}_1(N,t)\, [\rho_0^{(1)}]_{21}[A_1]_{12}, \tag{4.27}$$

where we set

$$\mathcal{C}_1(N,t) = \prod_{j=2}^N \left[\mathrm{e}^{\mathrm{i}tz_j^+} \left\langle \psi_{\mathrm{S}_j}, B_j \xi_j^+ \right\rangle \left\langle \widetilde{\xi}_j^+, \Omega_{\mathrm{S}_j} \right\rangle + \mathrm{e}^{\mathrm{i}tz_j^-} \left\langle \psi_{\mathrm{S}_j}, B_j \xi_j^- \right\rangle \left\langle \widetilde{\xi}_j^-, \Omega_{\mathrm{S}_j} \right\rangle \right]. \tag{4.28}$$

Let us analyze the factors of this product. Using (4.7) and (4.8) we have (omitting the index j)

$$\langle \psi_{\mathrm{S}}, B\xi^\varrho \rangle \left\langle \widetilde{\xi}^\varrho, \Omega_{\mathrm{S}} \right\rangle \tag{4.29}$$

$$= \quad \frac{1}{1 + c[\alpha^\varrho]^2} \langle \psi_{\mathrm{S}}, B(\varphi_{11} + \alpha^\varrho \varphi_{22}) \rangle \left\langle \varphi_{11} + c[\alpha^\varrho]^* \varphi_{22}, 2^{-1/2}(\varphi_{11} + \varphi_{22}) \right\rangle$$

$$= \quad \frac{1 + c\alpha^\varrho}{1 + c[\alpha^\varrho]^2} \left\langle \psi_{\mathrm{S}}, B\, 2^{-1/2}(\varphi_{11} + \alpha^\varrho \varphi_{22}) \right\rangle$$

$$= \quad \frac{1 + c\alpha^\varrho}{1 + c[\alpha^\varrho]^2} \langle \psi_{\mathrm{S}}, B\{|\varphi_1\rangle\langle\varphi_1| \otimes \mathbb{1} + \alpha^\varrho |\varphi_2\rangle\langle\varphi_2| \otimes \mathbb{1}\}\Omega_{\mathrm{S}} \rangle \tag{4.30}$$

$$= \quad \frac{1 + c\alpha^\varrho}{1 + c[\alpha^\varrho]^2} \big([\rho_0]_{11} + \alpha^\varrho [\rho_0]_{22} \big). \tag{4.31}$$

In the last step, we use that B commutes with $|\varphi_j\rangle\langle\varphi_j| \otimes \mathbb{1}$, and that $B\Omega_{\mathrm{S}} = \psi_{\mathrm{S}}$. Next we note the relation $\alpha^+\alpha^- = -1/c$, which can be derived readily, for instance from the fact that $|\xi^+\rangle\langle\widetilde{\xi}^+| + |\xi^-\rangle\langle\widetilde{\xi}^-| = \mathbb{1}$. A short calculation then shows that

$$\zeta := \frac{1 + c\alpha^+}{1 + c(\alpha^+)^2} = 1 - \frac{1 + c\alpha^-}{1 + c(\alpha^-)^2},$$

so that the factor in the product (4.28) becomes

$$[\rho_0]_{11} \left\{ \mathrm{e}^{\mathrm{i}tz^+} \zeta + \mathrm{e}^{\mathrm{i}tz^-}(1 - \zeta) \right\} + (1 - [\rho_0]_{11}) \left\{ \mathrm{e}^{\mathrm{i}tz^+} \alpha^+ \zeta + \mathrm{e}^{\mathrm{i}tz^-} \alpha^-(1 - \zeta) \right\}.$$

Next, collecting the terms proportional to $[\rho_0]_{11}$ and using

$$(1-\alpha^-)(1-\zeta) = -(1-\alpha^+)\zeta \ \text{ and } \ \alpha^+\zeta = 1-\alpha^-(1-\zeta),$$

we obtain formula (2.26).

One can transfer the error term down from the exponent: with $D = O(\alpha^4)$ we have $\mathrm{e}^{-tY_1+tD} - \mathrm{e}^{-tY_1} = \mathrm{e}^{-tY_1}\sum_{n\geq 1}\frac{(tD)^n}{n!}$ and hence

$$|\mathrm{e}^{-tY_1+tD} - \mathrm{e}^{-tY_1}| \leq \mathrm{e}^{-tY_1}[\mathrm{e}^{t|D|}-1] \leq \mathrm{e}^{-tY_1} t|D|\mathrm{e}^{t|D|} \tag{4.32}$$

(mean value theorem). Now for $|D| \leq C\alpha^4$ and $Y_1 \geq c\alpha^2 > 0$, the r.h.s. can be bounded from above as follows: let $\epsilon > 0$, then for $\alpha^2 \leq c\epsilon/C$, an upper bound is

$$Ct\alpha^4\mathrm{e}^{-t\alpha^2c(1-\epsilon)} \leq C\alpha^2\mathrm{e}^{-t\alpha^2c(1-2\epsilon)}\sup_{x\geq 0} x\mathrm{e}^{-xc\epsilon} = \frac{C\alpha^2}{ec\epsilon}\mathrm{e}^{-t\alpha^2c(1-2\epsilon)}.$$

This gives that if $Y_1 \geq c\alpha^2 > 0$, then for all $\epsilon > 0$ and α small enough,

$$\mathrm{e}^{-tY_1+O(\alpha^4)} = \mathrm{e}^{-tY_1} + O(\alpha^2\mathrm{e}^{-tc\alpha^2(1-\epsilon)}). \tag{4.33}$$

The remainder depends on ϵ. Taking $\epsilon = 1/2$ and $\alpha < c/(2C)$, we get

$$\mathrm{e}^{-t[Y_1+O(\alpha^4)]} = \mathrm{e}^{-tY_1} + O(\alpha^2) \tag{4.34}$$

(uniformly in $t \geq 0$). This gives the contribution (2.23).

Similarly to (4.27), one shows that the term in (4.22) with $e = \omega_1$ equals (2.24). To derive this, one checks that under the change $e \mapsto -e$, the exponent in (4.27) undergoes a complex conjugation and a sign change, and $\mathcal{C}_1$ turns into its complex conjugate.

Next we consider $e = 0$ in (4.22). We have $N_0(0) = N$ and obtain two contributions: one associated with $\varepsilon_0 = 0$, $\eta_0 = \Omega_{\vec{S}}$ (see (4.4)), $\widetilde{\eta}_0 = \widetilde{\xi}_1^+ \otimes \cdots \otimes \widetilde{\xi}_N^+$ and another contribution with $\varepsilon_0^{(-1,1,1,\ldots)}$ and

$$\eta_0^{(-1,1,1,\ldots)} = \xi_1^- \otimes \xi_2^+ \otimes \cdots \otimes \xi_N^+ \quad \text{and} \quad \widetilde{\eta}_0^{(-1,1,1,\ldots)} = \widetilde{\xi}_1^- \otimes \widetilde{\xi}_2^+ \otimes \cdots \otimes \widetilde{\xi}_N^+. \tag{4.35}$$

It is easily seen that these two contributions are (2.21) and (2.22). This completes the proof of Theorem 2.1 ■

4.1 Level shift operators and proof of Proposition 4.1

The total Liouville operator has the form

$$K = L_0 + \widetilde{W}, \tag{4.36}$$

where $L_0 = L_{\vec{S}} + L_{\vec{R}}$ is the free (non interacting) Liouville operator, and $\widetilde{W}$ contains all interactions and is of $O(\alpha^2)$ (in the notation of Appendix A, $\widetilde{W} = W - J\Delta^{1/2}WJ\Delta^{1/2}$, where we do not display the spectral deformation parameter ω here; for more detail, see Appendix A and also [16], Resonance theory of decoherence and thermalization). To every eigenvalue e of L_0 we associate the level shift operator

$$\Lambda_e = -P_e \widetilde{W} \overline{P}_e \left(L_0 - e + \mathrm{i}0_+\right)^{-1} \overline{P}_e \widetilde{W} P_e, \tag{4.37}$$

where P_e is the spectral projection of L_0 onto the eigenvalue e and $\overline{P}_e = \mathbb{1} - P_e$. The eigenvalues of Λ_e are the second order (in α) corrections to the eigenvalues of K under the analytic (in α) perturbation W of L_0, see also Section 5 of [16], Resonance theory of decoherence and thermalization. Moreover, if $\varepsilon_e^{(\varrho)} = e + \delta_e^{(\varrho)} + O(\alpha^4)$ are the eigenvalues bifurcating out of e for $\alpha \neq 0$ then the corresponding eigenprojections of K, to lowest order in α, are given by $|\eta_e^{(\varrho)}\rangle\langle\widetilde{\eta}_e^{(\varrho)}|$, where

$$\begin{aligned} \Lambda_e \eta_e^{(\varrho)} &= \delta_e^{(\varrho)} \eta_e^{(\varrho)} && (4.38) \\ [\Lambda_e]^* \widetilde{\eta}_e^{(\varrho)} &= [\delta_e^{(\varrho)}]^* \widetilde{\eta}_e^{(\varrho)} && (4.39) \end{aligned}$$

and $\left\langle \eta_e^{(\varrho)}, \widetilde{\eta}_e^{(\varrho)} \right\rangle = 1$. For more information on these facts, we refer to Section 6 of [16], Resonance theory of decoherence and thermalization. We are assuming here that all energies $\delta_e^{(\varrho)}$ are different, so that the corresponding eigenspaces are one-dimensional. This is generically true in applications, but it is not necessary for our strategy to work, see e.g. Appendix A of [16], Dynamics of collective decoherence and thermalization.

Below, we give the explicit form of the level shift operators associated with all eigenvalues e (given by (2.8)). Each level shift operators splits into a sum

$$\Lambda_e = \Lambda_e^{\mathrm{coll}} + \Lambda_e^{\mathrm{loc}} \tag{4.40}$$

of two operators associated with the local and the collective interactions. We find the spectrum and eigenvalues of the level shift operators. In view of the explanations given at the beginning of this section, this gives a proof of Proposition 4.1.

4.1.1 Collective level shift operator

Let e be an eigenvalue (2.8) and define

$$r_n = \frac{1}{4} \varkappa_n e_0(e) \int_{\mathbb{R}^3} \frac{|f_{\mathrm{c}}(k)|^2}{|k|} \mathrm{d}^3 k \tag{4.41}$$

Proposition 4.2 (Collective LSO) *The collective level shift operator associated to e is given by*

$$\Lambda_e^{\mathrm{coll}} = x_e(\{\lambda_n\}, g_{\mathrm{c}}) + \mathrm{i}[y_e'' + y_e(\{\lambda_n\}, g_{\mathrm{c}})] + \sum_{\{n:\sigma_n = \tau_n\}} M_{\mathrm{coll}}^n, \tag{4.42}$$

where M_{coll}^n acts on span$\{\varphi_{++}, \varphi_{--}\}$ *(doubled Hilbert space of n-th spin) as*

$$M_{\mathrm{coll}}^n = \mathrm{i}\frac{\pi}{4} \lambda_n^2 (\omega_n)^2 \frac{\mathcal{G}_{g_c}(\omega_n)}{\mathrm{e}^{\beta\omega_n} - 1} \begin{bmatrix} 1 & -1 \\ -\mathrm{e}^{\beta\omega_n} & \mathrm{e}^{\beta\omega_n} \end{bmatrix} - r_n \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}. \tag{4.43}$$

A proof of this proposition is obtained along the lines of Proposition 3.7 of [16] (Dynamics of collective decoherence and thermalization).

4.1.2 Local level shift operator

Proposition 4.3 (Local LSO) *The local level shift operator associated to e is given by*

$$\Lambda_e^{\rm loc} = x_e(\{\mu_n\},\{g_n\}) + \mathrm{i}[y_e' + y_e(\{\mu_n\},\{g_n\})] + \sum_{\{n:\sigma_n=\tau_n\}} M_{\rm loc}^n, \tag{4.44}$$

where $M_{\rm loc}^n$ *acts on* $\mathrm{span}\{\varphi_{++},\varphi_{--}\}$ *(doubled Hilbert space of n-th spin) as*

$$M_{\rm loc}^n = \mathrm{i}\frac{\pi}{4}\mu_n^2(\omega_n)^2 \frac{\mathcal{G}_{g_n}(\omega_n)}{\mathrm{e}^{\beta\omega_n}-1}\left[\begin{array}{cc} 1 & -1 \\ -\mathrm{e}^{\beta\omega_n} & \mathrm{e}^{\beta\omega_n}\end{array}\right]. \tag{4.45}$$

A proof of this proposition is obtained along the lines of Proposition 5.1 of [16] (Resonance theory of decoherence and thermalization).

Remarks. (1) The contributions to local and collective level shift operators coming from the energy-exchange interactions are of the same form.

(2) In the present work we consider as a reference state for each spin the trace state (4.2). In [16] (Resonance theory of decoherence and thermalization, Proposition 5.1) the level shift operators have been calculated for the spin-reference state equalling the Gibbs state at temperature β. The formulas obtained with the latter reference state become the ones for the current reference state upon replacing $\mathrm{e}^{\pm\beta H_S}$ by 1 (for $\beta=\infty$ the Gibbs state is the trace state).

4.1.3 Proof of Proposition 4.1

The explicit forms of $\Lambda_e^{\rm coll}$ and $\Lambda_e^{\rm loc}$ given in Propositions 4.2 and 4.3, and relation (4.40) yield

$$\Lambda_e = X_e + \mathrm{i}Y_e + \sum_{\{n:\sigma_n=\tau_n\}}\left\{\mathrm{i}b_n\left[\begin{array}{cc} c_n & -c_n \\ -1 & 1\end{array}\right] - r_n\left[\begin{array}{cc} 1 & 0 \\ 0 & -1\end{array}\right]\right\}. \tag{4.46}$$

The results (4.20), (4.19) and (4.9) for the eigenvalues and eigenvectors follow, and similarly for its adjoint.

Note that for $e=0$ we have $e_0(e)=0$, and the level shift operator becomes

$$\Lambda_0 = \mathrm{i}\sum_{n=1}^N b_n\left[\begin{array}{cc} c_n & -c_n \\ -1 & 1\end{array}\right]. \tag{4.47}$$

It follows that $z_n^+=0$ and $z_n^-=\mathrm{i}b_n(c_n+1)$, with

$$\xi_n^+ = \frac{1}{\sqrt{2}}\left[\begin{array}{c} 1 \\ 1\end{array}\right],\quad \xi_n^- = \left[\begin{array}{c} c_n \\ -1\end{array}\right],\quad \widetilde{\xi}_n^+ = \frac{\sqrt{2}}{1+c_n}\left[\begin{array}{c} 1 \\ c_n\end{array}\right],\quad \widetilde{\xi}_n^- = \frac{1}{1+c_n}\left[\begin{array}{c} 1 \\ -1\end{array}\right]. \tag{4.48}$$

5 Validity of perturbation expansion

The Heisenberg equations of motion corresponding to (2.1)-(2.3) are

$$\begin{aligned}
\dot S_n^z &= -\mathrm{i}[S_n^z, H] = \lambda_{\mathrm{c}} S_n^y \otimes \phi_{\mathrm{c}}(g_{\mathrm{c}}) + \mu_n S_n^y \otimes \phi_n(g_n) && (5.1)\\
\dot S_n^x &= \omega_n S_n^y - \varkappa_n S_n^y \otimes \phi_{\mathrm{c}}(f_{\mathrm{c}}) - \nu_n S_n^y \otimes \phi_n(f_n) && (5.2)\\
\dot S_n^y &= -\omega_n S_n^x - \lambda_n S_n^z \otimes \phi_{\mathrm{c}}(g_{\mathrm{c}}) + \varkappa_n S_n^x \otimes \phi_{\mathrm{c}}(f_{\mathrm{c}}) - \nu_n S_n^z \otimes \phi_n(g_n) && (5.3)\\
&\quad + \nu_n S_n^x \otimes \phi_n(f_n). && (5.4)
\end{aligned}$$

For the local and collective annihilation operators $a_n(k)$ and $a_{\mathrm{c}}(k)$ we have

$$\begin{aligned}
\mathrm{i}\dot a_n(k) &= [a_n(k), H] = |k| a_n(k) + \frac{1}{\sqrt{2}} \mu_n g_n(k) S_n^x + \frac{1}{\sqrt{2}} \nu_n f_n(k) S_n^z && (5.5)\\
\mathrm{i}\dot a_{\mathrm{c}}(k) &= |k| a_{\mathrm{c}}(k) + \frac{1}{\sqrt{2}} \lambda_n g_{\mathrm{c}}(k) \sum_{n=1}^N S_n^x + \frac{1}{\sqrt{2}} \varkappa_n f_{\mathrm{c}}(k) \sum_{n=1}^N S_n^z. && (5.6)
\end{aligned}$$

The latter two equations can be integrated,

$$\begin{aligned}
a_n(k,t) &= \mathrm{e}^{-\mathrm{i}|k|t} a_n(k,0)\\
&\quad - \frac{1}{\sqrt{2}} \int_0^t \mathrm{e}^{-\mathrm{i}|k|(t-\tau)} \left\{ \mu_n g_n(k) S_n^x(\tau) + \nu_n f_n(k) S_n^z(\tau) \right\} \mathrm{d}\tau && (5.7)\\
a_{\mathrm{c}}(k,t) &= \mathrm{e}^{-\mathrm{i}|k|t} a_{\mathrm{c}}(k,0)\\
&\quad - \frac{1}{\sqrt{2}} \int_0^t \mathrm{e}^{-\mathrm{i}|k|(t-\tau)} \left\{ \lambda_n g_{\mathrm{c}}(k) \sum_{n=1}^N S_n^x(\tau) + \varkappa_n f_{\mathrm{c}}(k) \sum_{n=1}^N S_n^z(\tau) \right\} \mathrm{d}\tau. && (5.8)
\end{aligned}$$

Remembering that $\phi(h) = \frac{1}{\sqrt{2}} \int_{\mathbb{R}^3} \{h(k) a^*(k) + \overline{h}(k) a(k)\} \mathrm{d}^3 k$ we insert (5.7), (5.8) into (5.2) to obtain

$$\begin{aligned}
\dot S_n^x(t) &= \omega_n S_n^y(t) - \varkappa_n S_n^y(t) \phi(\mathrm{e}^{\mathrm{i}|k|t} f_{\mathrm{c}})\\
&\quad + \frac{1}{2} \varkappa_n S_n^y(t) \int_{\mathbb{R}^3} \int_0^t \mathrm{e}^{-\mathrm{i}|k|(t-\tau)} \overline{f_{\mathrm{c}}}(k) \left\{ \lambda_n g_{\mathrm{c}}(k) \sum_{n=1}^N S_n^x(\tau) + \varkappa_n f_{\mathrm{c}}(k) \sum_{n=1}^N S_n^z(\tau) \right\} \mathrm{d}\tau \mathrm{d}^3 k\\
&\qquad + \mathrm{h.c.}\\
&\quad + \frac{1}{2} \nu_n S_n^y(t) \int_{\mathbb{R}^3} \int_0^t \mathrm{e}^{-\mathrm{i}|k|(t-\tau)} \overline{f_n}(k) \left\{ \mu_n g_n(k) S_n^x(\tau) + \nu_n f_n(k) S_n^z(\tau) \right\} \mathrm{d}\tau \mathrm{d}^3 k\\
&\qquad + \mathrm{h.c.} && (5.9)
\end{aligned}$$

Let us denote by $h^2(k)$ any of the products of functions of k occuring in the above integrals (e.g. $h^2(k) = \overline{f_{\mathrm{c}}}(k) g_{\mathrm{c}}(k)$ etc). Let us analyze the k-integrals in the last expression for $\dot S_n^x$. The product of form factors behaves like $h^2(k) = |k|^{p_1+p_2} \mathrm{e}^{-|k|/k_0}$, where $p_j = -1/2 + n_j$, $n_j = 0, 1, 2, \dots$ (see Assumption (C)). So

$$\int_{\mathbb{R}^3} \mathrm{e}^{\mathrm{i}|k|\tau} h^2(k) \mathrm{d}^3 k \sim \int_0^\infty r^{1+n_1+n_2} \mathrm{e}^{\mathrm{i}r\tau} \mathrm{e}^{-r/k_0} \mathrm{d}r = \partial_\tau^{1+n_1+n_2} \frac{(-\mathrm{i})^{1+n_1+n_2}}{1/r_0 + \mathrm{i}\tau},$$

which decays at least as $\frac{1}{1+\tau^2}$ (worst case $n_1 = n_2 = 0$). Together with the boundedness $|S_n^{x,y,z}(\tau)| \leq 1/2$ this implies that the integrals over τ and k in (5.9) are bounded homogeneously in $t \geq 0$, leading to

$$\dot{S}_n^x(t) = \omega_n S_n^y(t) - \varkappa_n S_n^y(t)\phi(\mathrm{e}^{\mathrm{i}|k|t} f_{\mathrm{c}}) + \varkappa_n O(\lambda_n N + \varkappa_n N) + \nu_n O(\mu_n + \nu_n).$$

For the validity of perturbation theory homogeneously in $t \geq 0$, we should impose $\varkappa_n^2 N, \varkappa_n \lambda_n N \ll \omega_n$ and $\nu_n^2, \nu_n \mu_n \ll \omega_n$. Denoting by α_{c} and α_ℓ the size of collective and local coupling parameters, we thus need $\alpha_{\mathrm{c}}^2 N \ll \omega$, $\alpha_\ell \ll \omega$, where ω is the (typical) Bohr frequency of the single spin.

A The operator K

The purpose of this Appendix is to provide some detail on explicit formulas of the operators K. For more detail, we refer to [16].

Given a form factor $g = g(k) \in L^2(\mathrm{R}^3, \mathrm{d}^3k)$, smoothed-out creation and annihilation operators are defined by $a^*(g) = \int_{\mathrm{R}^3} g(k) a^*(k) \mathrm{d}^3k$, $a(g) = \int_{\mathrm{R}^3} \overline{g(k)} a(k) \mathrm{d}^3k$, and the field operator is given by (2.6).

Description of a single reservoir. The so-called Araki–Woods representation gives the Hilbert space (GNS) representation of the infinitely extended Bose gas in thermal equilibrium [29, 30].[6] The Hilbert space is given by the bosonic Fock space over the one-particle space $L^2(\mathbb{R} \times S^2, \mathrm{d}u \times \mathrm{d}\Sigma)$,

$$\mathcal{F} = \mathcal{F}(L^2(\mathbb{R} \times S^2, \mathrm{d}u \times \mathrm{d}\Sigma)), \tag{A.1}$$

where $\mathrm{d}\Sigma$ is the uniform measure on S^2 and $\mathrm{d}u$ is the Lebesgue measure on $\mathbb{R}$. The thermal annihilation operators are

$$a_\beta(f) = a\big(\sqrt{1+\mu_\beta(u)}\,\chi_+(u) u f(u,\sigma)\big) - a^*\big(\sqrt{\mu_\beta(-u)}\,\chi_-(u) u \overline{f}(-u,\sigma)\big), \tag{A.2}$$

where $\mu_\beta(u) = (\mathrm{e}^{\beta u} - 1)^{-1}$ and $\chi_\pm$ are the indicator functions of $\mathbb{R}_\pm$ The $a_\beta^*(f)$ are obtained by taking the adjoint on the r.h.s. of (A.2). It is easy to see that the CCR are satisfied. The thermal field operator is

$$\phi_\beta(f) = \frac{1}{\sqrt{2}}(a_\beta^*(f) + a_\beta(f)) = \frac{1}{\sqrt{2}}(a^*(f_\beta) + a(f_\beta)) =: \phi(f_\beta), \tag{A.3}$$

for $f \in L^2(\mathbb{R}^3)$, where f_β is defined by

$$f_\beta(u,\sigma) := \sqrt{\frac{u}{1-\mathrm{e}^{-\beta u}}}\;|u|^{1/2} \left\{ \begin{array}{ll} f(u,\sigma) & \text{if } u \geq 0 \\ \overline{f}(-u,\sigma) & \text{if } u < 0 \end{array} \right. \tag{A.4}$$

(spherical coordinates on the r.h.s.) Note that ϕ in the r.h.s. of (A.3) is the field operator in $\mathcal{F}$. The equilibrium state is represented by the vacuum vector of $\mathcal{F}$, $\Omega_R = \Omega$. For a one-body operator O acting on wave functions of the variables (u,σ), we write

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[6] In this paper, we directly work in a spatially unitarily equivalent representation of the original representation, see [16] for details.

$\mathrm{d}\Gamma(O)=\int_{\mathbb{R}\times S^2} a^*(u,\sigma)Oa(u,\sigma)\,\mathrm{d}u\mathrm{d}\sigma$ for the second quantization of O. The dynamics of the field is generated by

$$L_R=\mathrm{d}\Gamma(u), \tag{A.5}$$

the second quantization of the operator of multiplication by u. We have $L_R\Omega_R=0$, and for $z\in\mathbb{C}$, $\mathrm{e}^{zL_\mathrm{R}}\phi_\beta(f)\mathrm{e}^{-zL_\mathrm{R}}=2^{-1/2}\big[a_\beta\big(\mathrm{e}^{-\overline{z}u}f\big)+a^*_\beta\big(\mathrm{e}^{zu}f\big)\big]$, which gives the dynamics for $z=\mathrm{i}t$.

Description of the entire system. The total (GNS) Hilbert space of N spins 1/2 with all local and collective reservoirs (c.f. (2.1)-(2.3)) is given by

$$\mathcal{H}_\mathrm{tot}=(\mathbb{C}^2\otimes\mathbb{C}^2)\otimes\cdots\otimes(\mathbb{C}^2\otimes\mathbb{C}^2)\otimes\mathcal{F}_1\otimes\cdots\otimes\mathcal{F}_n\otimes\mathcal{F}_c, \tag{A.6}$$

where each $\mathbb{C}^2\otimes\mathbb{C}^2$ is the Hilbert space of a single spin (in the GNS representation) and where $\mathcal{F}_j$, $\mathcal{F}_c$ are copies of $\mathcal{F}$, (A.1), representing the j-th local and the collective reservoirs, respectively.

The total Liouville operator L acts on $\mathcal{H}_\mathrm{tot}$ and is given by $L=L_0+W$, where $L_0=L_{\vec{S}}+L_{\vec{R}}$ with $L_{\vec{S}}$ defined in (4.6) and $L_{\vec{R}}=\sum_{n=1}^N \mathrm{d}\Gamma_n(u)+\mathrm{d}\Gamma_c(u)$. Here, $\mathrm{d}\Gamma_n(u)$ acts on $\mathcal{F}$ associated to the local reservoir n (and similarly for the collective reservoir). According to (2.2)-(2.3), the interaction operator W is the sum

$$\begin{aligned} W &= \sum_{n=1}^N \lambda_n S_n^x\otimes\mathbb{1}\otimes\phi_\mathrm{c}((g_\mathrm{c})_\beta)+\sum_{n=1}^N \varkappa_n S_n^z\otimes\mathbb{1}\otimes\phi_\mathrm{c}((f_\mathrm{c})_\beta) && \text{(A.7)}\\ &\quad+\sum_{n=1}^N \mu_n S_n^x\otimes\mathbb{1}\otimes\phi_n((g_n)_\beta)+\sum_{n=1}^N \nu_n S_n^z\otimes\mathbb{1}\otimes\phi_n((f_n)_\beta). && \text{(A.8)}\end{aligned}$$

In the above sums, $\mathbb{1}$ stands for the identity operator on the second factor of $\mathbb{C}^2\otimes\mathbb{C}^2$ associated to spin n, $(g_c)_\beta$ is as in (A.4). Of course, ϕ_n is the field operator acting on the space $\mathcal{F}$ associated to the n-th local reservoir (and similarly for the collective reservoir).

In the spirit of spectral deformation methods ("complex scaling"), we introduce the deformation group (in ω) $U(\omega)$, acting as $\mathrm{e}^{-\mathrm{i}\omega\mathrm{d}\Gamma(\mathrm{i}\partial_u)}$ on each reservoir Hilbert space $\mathcal{F}$. The spectrally deformed Liouville operator is

$$L(\omega)=U(\omega)LU(\omega)^{-1}=L_0+\omega(N_1+\cdots N_n+N_c)+W(\omega), \tag{A.9}$$

where $N_j=\mathrm{d}\Gamma_j(\mathbb{1})$ is the number operator in $\mathcal{F}$ of reservoir j, and where $W(\omega)=U(\omega)WU(\omega)^{-1}$.

Definition of the operator K. Define the following operators acting on $\mathcal{H}_\mathrm{tot}$

$$\begin{aligned} J &= J_{S_1}\otimes\cdots\otimes J_{S_N}\otimes J_{R_1}\otimes\cdots\otimes J_{R_N}\otimes J_{R_c} && \text{(A.10)}\\ \Delta &= \mathbb{1}_{\vec{S}}\otimes\mathrm{e}^{-\beta L_{R_1}}\otimes\cdots\otimes\mathrm{e}^{-\beta L_{R_N}}\otimes\mathrm{e}^{-\beta L_{R_c}} && \text{(A.11)}\end{aligned}$$

where $J_{S_j}\phi_l\otimes\phi_r=\mathcal{C}\phi_r\otimes\mathcal{C}\phi_l$, $J_{R_j}\psi_n(u_1,\sigma_1,\ldots,u_n,\sigma_n)=\overline{\psi}_n(-u_1,\sigma_1,\ldots,-u_n,\sigma_n)$, and where the action of the antilinear operator $\mathcal{C}$ is to take the complex conjugate of vector coordinates in the energy basis (associated to S^z) and $\overline{\psi}_n$ is the complex conjugate of $\psi_n\in\mathcal{F}$. The operator K is defined as

$$K=L(\omega)-U(\omega)J\Delta^{1/2}WJ\Delta^{1/2}U(\omega)^{-1}.$$

We now give the explicit contribution to K coming from the first sum in (A.7). It is then clear how to obtain the other ones. We have $J_R a^{\#}(f(u,\sigma))J_R = a^{\#}(\overline{f}(-u,\sigma))$. One obtains

$$J\Delta^{1/2} S^x_n \otimes \mathbb{1} \otimes \phi_c((g_c)_\beta) J\Delta^{1/2} = \mathbb{1} \otimes S^x_n \otimes \frac{1}{\sqrt{2}} \left[a^*_c\big((g_c)_\beta\big) + a_c\big(\mathrm{e}^{-\beta u}(g_c)_\beta\big)\right]. \quad \text{(A.12)}$$

The spectral deformation of the last expression is obtained as follows. For $\omega \in \mathrm{R}$ we have $U(\omega)a^{\#}(f)U(\omega)^{-1} = a^{\#}(f(\cdot+\omega))$, where $f(\cdot+\omega)$ is the shifted function $(u,\sigma)\mapsto f(u+\omega,\sigma)$. In order to obtain an analytic extension to complex ω, we need to take the complex conjugate of ω in the argument of the annihilation operator (since the latter is anti-linear in its argument). We thus arrive at the following contribution of the general term in the first sum of (A.7) to K:

$$\mathbb{1} \otimes S^x_n \otimes \frac{1}{\sqrt{2}} \left[a^*_c\big((g_c)_\beta(\cdot+\omega))\big) + a_c\big(\mathrm{e}^{-\beta(\cdot+\overline{\omega})}(g_c)_\beta(\cdot+\overline{\omega}))\big)\right]. \quad \text{(A.13)}$$

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