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Irreversible adiabatic decoherence of dipole-interacting nuclear-spin pairs coupled with a phonon bath

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We study the quantum adiabatic decoherence of a multispin array, coupled with an environment of harmonic phonons, in the framework of the theory of open quantum systems. We follow the basic formal guidelines of the well-known spin-boson model, since in this framework it is possible to derive the time dependence of the reduced density matrix in the adiabatic time scale, without resorting to coarse-graining procedures. However, instead of considering a set of uncoupled spins interacting individually with the boson field, the observed system in our model is a network of weakly interacting spin pairs; the bath corresponds to lattice phonons, and the system-environment interaction is generated by the variation of the dipole-dipole energy due to correlated shifts of the spin positions, produced by the phonons. We discuss the conditions that the model must meet in order to fit within the adiabatic regime. By identifying the coupling of the dipole-dipole spin interaction with the low-frequency acoustic modes as the source of decoherence, we calculate the decoherence function of the reduced spin density matrix in closed way, and estimate the decoherence rate of a typical element of the reduced density matrix in one- and three-dimensional models of the spin array. Using realistic values for the various parameters of the model we conclude that the dipole-phonon mechanism can be particularly efficient to degrade multispin coherences, when the number of active spins involved in a given coherence is high. The model provides insight into the microscopic irreversible spin dynamics involved in the buildup of quasiequilibrium states and in the coherence leakage during refocusing experiments in nuclear magnetic resonance of crystalline solids.

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I. INTRODUCTION

Quantum dynamics of dipole-interacting spin ensembles in solids arouses great interest in various fields of modern physics, both fundamental and applied. Particularly, nuclear spins provide suitable model systems to face frontier problems on the physics of many-body systems, such as the very foundations of statistical mechanics [1]. Decoherence and irreversibility are essential pieces for the understanding of the complex dynamics that precedes equilibrium, and they are ultimately linked with basic open questions such as the emergence of thermodynamic equilibrium from the underlying microscopic unitary quantum dynamics [2,3] and the measurement problem of quantum mechanics [4–7]. Within this context is the challenging problem of explaining the irreversible coherence loss observed in refocusing experiments and the mechanism that enables spins in crystalline solids to attain a quasiequilibrium state over an early time scale—in their transit to equilibrium—long before the process governing thermalization may have acted [8–10].

In the field of applications, considerable effort is devoted to manipulating and using quantum spin systems for new applications, such as quantum computations and quantum information processing [11]. A common characteristic of these developments is that they all need the occurrence of quantum coherence between different states of a many-spin system [12]. Thus, a better understanding of the sources of the environment-induced destruction of coherent superposition of states, that is, of decoherence, becomes fundamental both in the search of scalable quantum devices and in the characterization of many-body quantum systems.

A very sensitive technique to probe the nuclear-spin dynamics along a wide range of well-differentiated time scales is nuclear magnetic resonance (NMR). Here, we present a treatment of the irreversible decoherence of solids in NMR that considers the interacting spins and the environment as a composite system evolving under purely quantum rules. Both basic and applied research fields would benefit from the development of Hamiltonian models, which capture the essential physics and are amenable to rigorous analysis of the many-body nature of the quantum interacting particles coupled to a quantum environment. To advance in this direction it is necessary to adopt a realistic model for the solid, which allows us to both derive a detailed theoretical description from first principles and to elaborate a quantitative analysis of the results. Particularly, an adequate theoretical model may pave the way for investigating the quantum dynamics of an open system of interacting particles, as well as the connection between decoherence and the system-bath entanglement. A theoretical approach based on the usual spin-boson model [13–15], adapted to take account of the spin interactions could meet those expectations.

In this work we study the adiabatic decoherence of a system of dipole-coupled spins interacting with a phonon bath. The kind of system we contemplate is that of a crystal lattice where the observed system is an ordered array of pairs of spins 1/2. The term "adiabatic" means that spins and environment do not exchange energy in the average. We follow the basic formal guidelines of the well-known spin-boson model [16], however, instead of considering a set

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of uncoupled spins interacting individually with the boson field [17,18], the observed system in our model is a network of weakly interacting spin pairs, the boson bath corresponds to the lattice phonons at temperature T, and the system-environment interaction is generated by the variation of the dipole-dipole energy due to correlated shifts of the spin positions from the equilibrium ones, produced by lattice phonons. For the sake of simplicity we keep only the main part of this interaction energy, that is, the one coming from the variation of the local (intrapair) energy. In other words, we consider a spin system whose closed-system dynamics is that of an array of spin pairs, which are magnetically coupled with all the other pairs. The openness comes from the variation of the dipole intrapair energy, which in turn correlates the different pairs through their collective dynamics.

The strategy is based on calculating the exact quantum dynamics of an initial state of the observed system to derive the adiabatic decoherence function of this model. In order to compare with the experiment we estimate the characteristic decoherence time scale of a typical element of the reduced density matrix. The basic ideas are applied to one- and three-dimensional (3D) models of oriented spin pairs. The comparison between the two cases provides insight into the role of dimension and complexity on the decoherence of quantum many-body systems.

Our calculation is intended to determine if this pair-boson coupling can act as an effective source of irreversible decoherence, able to adiabatically bring an out-of-equilibrium complex system of weakly coupled pairs to a state of internal quasiequilibrium. The occurrence of diagonal states representing quasiequilibrium states in solid-state NMR [8,19–23] is often postulated on the basis of the spin temperature hypothesis [24–27]. The spin dynamics associated with these states was described in the framework of spin thermodynamics, where the statistical properties of the spin system are formally described by extending the tools of statistical mechanics to the quasiequilibrium states [8]. Thus, a Boltzmann operator involving the various constants of motion or quasi-invariants is postulated, as an extension of the equilibrium grand canonical ensemble technique. However, the theoretical explanation of the transient processes occurring in the way of the system towards quasiequilibrium still represents a challenge for quantum theories of interacting many-body systems. Explaining the decay of the off-diagonal density matrix elements could provide additional insight on the idea behind the spin temperature assumption. Similarly, it can contribute to the understanding of the influence of the environmental degrees of freedom on the generation and the decay of multi-spin correlation in the spin dynamics of the solid state [28–30]. From a fundamental point of view, this example illustrates the way that the einselection (environment induced selection) [31] operates to suppress the quantum oscillations of an initial coherent state of a many-body spin system, along the adiabatic time scale.

Section II contains a brief review of the main general procedures of the theory of open quantum systems used in the following sections. In Sec. III the spin system is defined, together with the Hamiltonians of the system, bath and spin-bath interaction. Section IV contains a derivation of the decoherence function in a one-dimensional model and an estimation of a characteristic decoherence time scale. A discussion on the decoherence function of a three-dimensional array, and its dependence on the various lattice parameters is presented in Sec. V.

II. OPEN QUANTUM SYSTEMS

As usual when describing an open quantum system in contact with an external bath, we write the total Hamiltonian as

$$\mathcal{H} = \mathcal{H}_S + \mathcal{H}_B + \mathcal{H}_{SL}, \tag{1}$$

where the system of interest is described (in isolation) by $\mathcal{H}_S$, the bath by $\mathcal{H}_B$, and $\mathcal{H}_{SL}$ represents the system-bath interaction.

In this work we are interested in studying the evolution of coherences along an early time scale where the effects of relaxation are not yet appreciable, by calculating the matrix elements of the reduced density matrix for a given model, in exact way (without adopting a coarse-grained time scale). Then, we assume that the spin and the bath Hamiltonians satisfy the adiabatic condition

$$[\mathcal{H}_S,\mathcal{H}_{SL}] = 0 \ , \tag{2}$$

which means that the mean value of the spin energy, $\langle\mathcal{H}_S\rangle$, is a conserved quantity in this time scale.

To model the interaction Hamiltonian we follow Ref. [15] and write

$$\mathcal{H}_{SL} = \sum_A \Lambda^A \otimes P^A$$

as the sum of products of general Hermitian operators, Λ^A, of the observed system (spins) and operators P^A, which act on the Hilbert space of the bath. The only restriction imposed to $\mathcal{H}_{SL}$ is condition (2).

The properties of the observed system are expressed by a reduced density operator σ, which is obtained by tracing the density operator ρ of the whole system over the bath variables [32], that is,

$$\sigma = \mathrm{Tr}_B\{\rho\}.$$

The strategy for studying decoherence involves calculating the time dependence of σ. The unitary dynamics of ρ is driven by the time evolution operator $U(t) = \exp\{-it\mathcal{H}\}$, which, owing to condition (2) can be factorized as

$$U(t) = e^{-it\mathcal{H}_S} e^{-it(\mathcal{H}_B+\mathcal{H}_{SL})} = V_0(t)\, V(t), \tag{3}$$

where we introduced a rigid lattice time evolution operator $V_0(t) \equiv e^{-it\mathcal{H}_S}$, which acts on the spin variables only and an operator $V(t) \equiv e^{-it(\mathcal{H}_B+\mathcal{H}_{SL})}$, which acts on both the spin and lattice variables. In accordance with this evolution operator $U(t)$, the diagonal part of the density matrix in the common eigenbasis of $\mathcal{H}_S$ and $\mathcal{H}_{SL}$ remains constant with time.

Let us further define an operator $V_m(t)$ acting on the lattice variables only as

$$V(t)|m\rangle = |m\rangle V_m(t),$$

where $|m\rangle$ is an eigenvector of the set $\{\Lambda^A\}$. The time dependence of an element of the reduced density matrix is

$$\sigma_{mn}(t) = \langle m|\mathrm{Tr}_B\{U(t)\rho(0)\, U^\dagger(t)\}|n\rangle. \tag{4}$$

We now make the usual assumption that the initial condition can be factorized as $\rho(0) = \sigma(0) \otimes \rho_B$, then,

$$\sigma_{mn}(t) = \langle m|V_0(t)\sigma(0)V_0^\dagger(t)|n\rangle \, \mathrm{Tr}_B\{V_m(t)\rho_B \, V_n^\dagger(t)\}. \quad (5)$$

This very general expression may now be applied to particular cases by selecting the operators $\mathcal{H}_S$,$\mathcal{H}_{SL}$, and $\mathcal{H}_B$. The factor that involves $\sigma(0)$ in Eq. (5) describes the complex (unitary) dynamics of a closed interacting system, while the trace over the bath variables allows the occurrence of nonunitary behavior. Our aim is to calculate the second factor in the case of an array of dipole-interacting pairs of spin 1/2, coupled to a bath of phonons. In Sec. III A we consider a one-dimensional system, which, in spite of its simplicity, already contains the important basic elements that reflect both the interacting (many-body) character of the observed system and the correlated nature of the environment. In Sec. V we discuss a three-dimensional model to investigate the role of the spin-system complexity on decoherence, through the quantum system-environment correlation.

III. MODEL SYSTEM

A. Linear chain of spin pairs

We consider a system of dipole coupled nuclear spin 1/2 pairs in a strong, static magnetic field and represent the model solid as a monoatomic linear chain with a basis. The primitive cell of this one-dimensional Bravais lattice with parameter a, contains two identical atoms of mass m at a distance $d < a/2$. There are two elastic constants K and G between neighboring atoms (with $K > G$), see Fig. 1.

In this model each spin interacts with both an external magnetic field (Zeeman energy) and with all the other spins of the sample through the dipolar interaction. The spin-pair chain is immersed in a phonon bath, which describes the small amplitude displacement of nuclei around their equilibrium positions in the crystal, which in turn perturbs their magnetic dipole-dipole interaction.

Hereafter, we write the system Hamiltonians in units of $\hbar$. The bath energy of this chain model can be written as the sum of uncoupled oscillators of frequency ω_k (neglecting the zero point energy) as

$$\mathcal{H}_B = \sum_k \omega_k b_k^\dagger b_k, \quad (6)$$

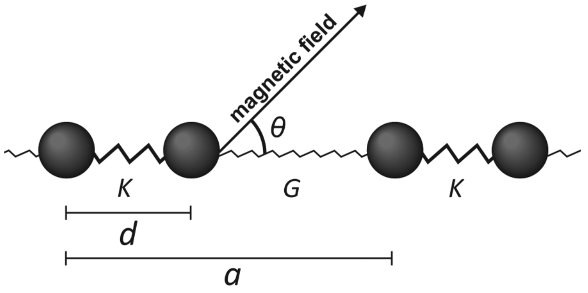

FIG. 1. Model solid used in the calculations. We assume a monoatomic chain with lattice parameter a, with two atoms per unit cell, separated a distance d. The spin system consists of a nuclear spin 1/2 per atom. The chain forms an angle θ with the direction of an external magnetic field. Each spin interacts with all other spins of the sample through dipolar interaction.

where the sum runs over the first Brillouin zone and b_k, $b_k^\dagger$ are the annihilation and creation operators, respectively, which satisfy $[b_k,b_k^\dagger] = 1$. The dispersion relation of a 1D chain is [33]

$$\omega_k{}^2 = \frac{K+G}{m} \pm \frac{1}{m}\sqrt{K^2 + G^2 + 2\,KG\cos(ka)}, \quad (7)$$

which admits an optical and an acoustic branch.

The Zeeman Hamiltonian is

$$\mathcal{H}_Z = \sum_i \omega_0 I_z^i, \quad (8)$$

where I_z^i is the z component of the angular momentum operator of the ith spin; $\omega_0 = \gamma B_0$ is the Larmor frequency, γ is the proton gyromagnetic ratio, and B_0 is the strength of the static magnetic field, which is applied along the z axis.

As regards the dipolar term, $\mathcal{H}_D$, we first adopt the high magnetic field approximation, since we assume that the norm of the Hamiltonians satisfy $\|\mathcal{H}_Z\| \gg \|\mathcal{H}_D\|$ and therefore keep only the secular part of the dipole Hamiltonian $\mathcal{H}_D^0$, which commutes with $\mathcal{H}_Z$ [34].

$$\mathcal{H}_D^0 = \frac{1}{2}\sum_{i\neq j}\sqrt{\frac{2}{3}}\,\Omega_D(r_{ij})T_{20}^{i,j}, \quad (9)$$

with $\Omega_D(r_{ij})$ defined as

$$\Omega_D(r_{ij}) = 3\frac{\mu_0\gamma^2\hbar}{8\pi r_{ij}^3}[1 - 3\cos^2(\theta)], \quad (10)$$

where θ is the angle between the external field and the chain direction and r_{ij} is the distance between the interacting nuclei i and j. $T_{20}^{i,j}$ is the zero component of a normalized irreducible spherical tensor of rank two, which in terms of the spin angular momentum operators is

$$T_{20}^{i,j} = \frac{1}{\sqrt{6}}\big[3I_z^iI_z^j - \mathbf{I}^i \cdot \mathbf{I}^j\big]. \quad (11)$$

Owing to the geometry (chain of pairs) selected in this work, a hierarchy of dipolar couplings arises, which naturally suggests splitting the secular dipolar Hamiltonian into two terms, one involving the interaction between spins belonging to the same cell, which we call intrapair, and the other representing the interpair interactions, $\mathcal{H}_D^0 = \mathcal{H}_{Da} + \mathcal{H}_{De}$. In order to stress the pair character of the spin system we label the spin pairs with indices $A,B,\ldots$ and distinguish spins within the same pair with a second index $u,v = 1$ or 2. In this way, the intrapair Hamiltonian can be written as

$$\mathcal{H}_{Da} = \sum_A \mathcal{H}_D^A = \sum_A \sqrt{\frac{2}{3}}\,\Omega_D(r_A)T_{20}^{A1,A2}, \quad (12)$$

while the interpair term involves the sum over spins belonging to different cells

$$\mathcal{H}_{De} = \frac{1}{2}\sum_{\substack{A\neq B\\ u,v}}\sqrt{\frac{2}{3}}\,\Omega_D(r_{Au,Bv})T_{20}^{Au,Bv} \quad (13)$$

It is convenient for the forthcoming reasoning, to make use of the fact that $\mathcal{H}_{De}$ can in principle be separated into a term $\widehat{\mathcal{H}}_{De}$ which commutes with $\mathcal{H}_{Da}$ and another, $\widetilde{\mathcal{H}}_{De}$, which does not. The expression for the truncated interpair dipole-dipole coupling $\widehat{\mathcal{H}}_{De}$, suggested in Ref. [35], which has been shown to adequately represent the spin dynamics in NMR experiments on hydrated salts, is reproduced in the Appendix. From now on, we keep only the commuting term $\widehat{\mathcal{H}}_{De}$.

We have so far defined the spin interactions and the bath energy. The connection between these two subsystems is provided by the change in the dipolar energy generated by the lattice vibration. It is worth to notice that it will only affect the dipolar and not the Zeeman interaction. We now assume that this perturbation is small, so that the dipolar energy can be expanded around the equilibrium value of the internuclear distances. In the case of the intrapair interaction, we have

$$\mathcal{H}_{Da} \approx \sum_A \mathcal{H}_D^A \bigg|_{r_A=d} + \sum_A \frac{d\mathcal{H}_D^A}{dr_A}\bigg|_{r_A=d} (r_A - d). \tag{14}$$

The first term of Eq. (14) is the rigid lattice value of the dipolar intrapair Hamiltonian, which from now on we call $\mathcal{H}_{Da}^R$. The second term is the first-order correction due to lattice vibration. The interpair contribution could, in principle, also be written as a rigid lattice term $\mathcal{H}_{De}^R$ plus the fluctuating terms, however, in this work we will disregard such fluctuations. Since the aim of this work is to examine if the pair-boson interaction may in fact be a source of decoherence, at this stage, and for the sake of simplicity, we consider only intrapair variations. This assumption is also based on the fact that $\|\mathcal{H}_{De}\|$ is smaller than $\|\mathcal{H}_{Da}\|$ because of the r^{-3} dependence. Then, the dipolar Hamiltonian of this model is

$$\mathcal{H}_D \approx \left[\mathcal{H}_{Da}^R + \widehat{\mathcal{H}}_{De}^R\right] + \sum_A \frac{d\mathcal{H}_D^A}{dr_A}\bigg|_{r_A=d} (r_A - d). \tag{15}$$

Let us now write $(r_A - d)$ in terms of the displacements from equilibrium, $\mu_{A,u}$, of each spin $u = 1,2$ at molecule A, since they can be easily related to the phonon creation and annihilation operators $b_{k,s}$ and $b_{k,s}^\dagger$ of mode k and branch s of an N primitive cells chain, as [33]

$$\mu_{A,u} = \frac{1}{\sqrt{N}} \sum_{k,s} \sqrt{\frac{\hbar}{2m\omega_{k,s}}} (b_{k,s} + b_{-k,s}^\dagger) e^{ik[Aa+(u-1)d]}, \tag{16}$$

where $[Aa + (u-1)d]$ is the equilibrium position of spin u in pair A. Then we obtain

$$r_A - d = \mu_{A,2} - \mu_{A,1} = \sum_k \left(g_k^{A*} b_k + g_k^A b_k^\dagger\right), \tag{17}$$

where we omitted the branch index s to simplify notation, and defined the coupling constants

$$g_k^A = e^{-ikAa} g(\omega_k), \tag{18}$$

with

$$g(\omega_k) = (1 - e^{-ikd}) \sqrt{\frac{\hbar}{2m\omega_k N}}. \tag{19}$$

At this stage we need to write the Hamiltonian of our model system in the spirit of Eq. (1). Keeping in mind that the energies involved in this problem are given by Eqs. (6), (8), and (15), we define the Hamiltonian of the spin system as the sum of $\mathcal{H}_Z$ and the term in square brackets in Eq. (15)

$$\mathcal{H}_S = \mathcal{H}_Z + \mathcal{H}_{Da}^R + \widehat{\mathcal{H}}_{De}^R, \tag{20}$$

and leave the second term of Eq. (15), having both spin and lattice variables, as the system-bath interaction

$$\begin{aligned}\mathcal{H}_{SL} &= \mathcal{H}_D - [\mathcal{H}_{Da}^R + \widehat{\mathcal{H}}_{De}^R] \\ &= \sum_A \Lambda^A \otimes \sum_k \left(g_k^{A*} b_k + g_k^A b_k^\dagger\right),\end{aligned} \tag{21}$$

where

$$\Lambda^A = \frac{d\mathcal{H}_{Da}}{dr}\bigg|_{r_A=d} = -\frac{\Omega_D(r_A = d)}{d} \sqrt{6}\, T_{20}^{A1,A2} \tag{22}$$

is an operator acting on the spin variables only. Therefore, the spin-lattice coupling is given by the anisotropic dipole-phonon Hamiltonian (21).

It is worth to mention that $\mathcal{H}_S$ involves the static magnetic dipole interactions between all the spins, both intrapair and interpair. In this way, the time evolution under $\mathcal{H}_S$ in Eq. (5) reflects the complexity of a multiply connected dipolar network. It is only in $\mathcal{H}_{SL}$ where we neglected the variation of the interpair interaction with the displacements. Even so, $\mathcal{H}_{SL}$ entangles the spin system with an environment having a dense spectrum of degrees of freedom. The variations associated to $\mathcal{H}_{De}$, which are not considered in this work, would add an extra source of decoherence, acting in a longer time scale.

B. Adiabatic hypothesis

Formally, the commutation of $\mathcal{H}_S$ and $\mathcal{H}_{SL}$ is not a general rule for the different models that can be proposed for describing natural physical situations. Indeed, from a mathematical viewpoint, Eq. (2) represents a very restrictive condition [6]. This work addresses the kind of systems where this requirement is met, at least within an early time scale, where thermal effects are still not important. From the viewpoint of the perturbation theory, the main contribution of the system-environment coupling along such a time scale is given by the commutative part of $\mathcal{H}_{SL}$ with respect to $\mathcal{H}_S$. That is, there is a time scale where the time evolution of the open system can be well represented by keeping only the commutative part of the spin-lattice Hamiltonian since the effect of the noncommutative part becomes relevant over a longer time scale. The first stage then is to find a commutative part for the particular interaction Hamiltonian of interest; the second, to find out if such adiabatic interaction is in fact a mechanism capable to allow the quantum phases loss.

In the particular case of the Hamiltonians of Eqs. (20) and (21), the commutator that is relevant for the discussion on the validity of the adiabatic hypothesis (2) is

$$\left[\mathcal{H}_{SL}, \widehat{\mathcal{H}}_{De}^R\right] = \left[\sum_A \Lambda^A \phi_A, \sum_{B \neq C} \overline{\Omega_D}(r_{B,C}) \mathcal{T}^{BC}\right], \tag{23}$$

where, in order to be concise, we used

$$\phi_A \equiv \sum_k g(\omega_k)(e^{ikAa} b_k + e^{-ikAa} b_k^\dagger), \tag{24}$$

and the interpair Hamiltonian as in Eq. (A1). Then

$$\begin{aligned}
&[\mathcal{H}_{SL},\widehat{\mathcal{H}}^{R}_{De}]\\
&\quad=\sum_{B\neq C}\overline{\Omega_D}(r_{BC})\sum_{A}[\phi_A\Lambda_A,\mathcal{T}^{BC}]\\
&\quad=\sum_{B\neq C}\overline{\Omega_D}(r_{BC})(\phi_B[\Lambda_B,\mathcal{T}^{BC}]+\phi_C[\Lambda_C,\mathcal{T}^{BC}])\\
&\quad=\sum_{B\neq C}\overline{\Omega_D}(r_{BC})(\phi_B-\phi_C)[\Lambda_C,\mathcal{T}^{BC}], \qquad (25)
\end{aligned}$$

where, in the last row we used that

$$[\Lambda^{B},\mathcal{T}^{BC}]=-[\Lambda^{C},\mathcal{T}^{BC}],$$

because of the definition of $\Lambda^{B}\propto T_{20}^{B1,B2}$ as in Eq. (22), and $\mathcal{T}^{BC}$ as in Eq. (A2). From Eq. (24) we see that

$$\begin{aligned}
\phi_B-\phi_C=\sum_{k}g(\omega_k)\{&b_k e^{ikBa}(1-e^{ik(C-B)a})\\
&+b_k^{\dagger}e^{-ikBa}(1-e^{-ik(C-B)a})\}. \qquad (26)
\end{aligned}$$

Notice that the commutator (25) differs from zero because of the phase factors of Eq. (26), however, the sum over k can be thought as partitioned into two contributions corresponding to phonon modes of short and long wavelengths, relative to some characteristic value $\bar{k}$ of the wave number, such that $|\bar{k}(C-B)a|\ll 1$ and therefore

$$e^{ik(C-B)a}\simeq 1,\quad \forall\, k\leqslant\bar{k}.$$

Then,

$$\mathcal{H}_{SL}=\mathcal{H}_{SL}^{(k<\bar{k})}+\mathcal{H}_{SL}^{(k>\bar{k})}\,. \qquad (27)$$

Let us now assume that a short time scale exists, where the coupling with the environment is controlled by the low-energy term $\mathcal{H}_{SL}^{(k<\bar{k})}$, so that

$$\left[\mathcal{H}_{S},\mathcal{H}_{SL}^{(k<\bar{k})}\right]=0, \qquad (28)$$

satisfying the adiabatic condition. Accordingly, in such time scale the time evolution operator of Eq. (5) can be written as

$$U(t)=e^{-it(\mathcal{H}^{R}_{Da}+\widehat{\mathcal{H}}^{R}_{De})}e^{-it(\mathcal{H}_{B}+\mathcal{H}_{SL}^{(k<\bar{k})})}\,. \qquad (29)$$

Therefore, the adiabatic hypothesis (28) leads to retaining only the terms with lower values of k in the interaction Hamiltonian, which implies keeping in the dynamics only the lower part of the frequency dependence of the acoustic branch, which satisfies $\omega_k=ck$.

In Sec. IV we analyze the potential of this adiabatic propagator to generate significant decoherence of the spin states. It is worth emphasizing that introducing the adiabatic hypothesis, which allowed writing the evolution operator as in Eq. (29), is equivalent to defining a characteristic time scale where the dynamics of the observed system evolves without energy exchange with the environment, far earlier than the processes ruled by thermal fluctuations.

On physical grounds, it is sound to assume that the typical time scale for decoherence of a system is short in comparison with its relaxation time scale [4,15,16]. For example, in the case of solid-state NMR, the nuclear spins in solids are weakly coupled to the lattice degrees of freedom, which usually makes the processes involving energy exchange between spins and the lattice to have very long relaxation times compared to the time scale of the coherent quantum many-body dynamics. Indeed, abundant experimental evidence in solid-state NMR shows that it is possible to prepare a variety of quasiequilibrium states whose relaxation can be described in terms of a spin temperature (that is, they behave as thermodynamic systems in the sense that they can be represented by a Boltzmann distribution with a temperature parameter different to that of the lattice) [24,36,37]. In other words, there is an evident distinction between two time scales, the shorter one distinguished by the vanishing of the coherence between levels with different energies and the buildup of the quasiequilibrium states; the longer one signed by relaxation to equilibrium with the external reservoir.

IV. DYNAMICS OF THE LINEAR-CHAIN MODEL

In this section we calculate the exact coherence dynamics given by Eq. (5), using the Hamiltonians of the particular system defined in Eqs. (6), (20), and (21) and the evolution operator of Eq. (29). We also assume that the initial state of the composite system is separable and that the bath is in a thermal state, described by independent density matrices Θ_k

$$\rho(0)=\sigma(0)\otimes\prod_{k}\Theta_k \qquad (30)$$

with

$$\begin{aligned}
\Theta_k&=Z_k^{-1}e^{-\hbar\beta\omega_k b_k^{\dagger}b_k}\\
Z_k&=(1-e^{-\beta\omega_k\hbar})^{-1}.
\end{aligned} \qquad (31)$$

In accordance with our definition of $\mathcal{H}_{SL}$, there exists an eigenbasis $\{|m\rangle\}$ whose elements can be written as direct products of states of the different pairs,

$$|m\rangle=|m\rangle_1\otimes|m\rangle_2\otimes\ldots|m\rangle_N\equiv|m_1,m_2,\ldots,m_N\rangle, \qquad (32)$$

where each $|m_A\rangle$ is any of the four states of pair A, that is, eigenstates of $\mathcal{H}_D^{A}$ and $\mathcal{H}_Z^{A}$,

$$|m_A\rangle\in\{|1,1\rangle,|1,0\rangle,|1,-1\rangle,|0,0\rangle\}. \qquad (33)$$

The corresponding eigenvalues of Λ^{A} are

$$\Lambda^{A}|m_A\rangle=\lambda_{m_A}|m_A\rangle.$$

The action of $\mathcal{H}_{SL}$ over a state $|m\rangle$ is

$$\begin{aligned}
\mathcal{H}_{SL}|m\rangle&=\sum_{A}\Lambda^{A}\otimes\sum_{k}\left(g_k^{A*}b_k+g_k^{A}b_k^{\dagger}\right)|m_1,\ldots,m_N\rangle\\
&=\sum_{A}\lambda_{m_A}\sum_{k}(e^{ikAa}g_k^{*}b_k+e^{-ikAa}g_k b_k^{\dagger})|m\rangle\\
&=\sum_{k}(\lambda_{m,k}^{*}\,g_k^{*}\,b_k+\lambda_{m,k}\,g_k\,b_k^{\dagger})|m\rangle, \qquad (34)
\end{aligned}$$

where we defined

$$\lambda_{m,k}\equiv\sum_{A}e^{-ikAa}\lambda_{m_A}. \qquad (35)$$

to emphasize that the sum over A in Eq. (34) links the eigenvalue of each pair with its position in the chain.

We now introduce the notation

$$\mathcal{H}_{SL}(m) = \sum_k (\lambda^*_{m,k} g^*_k b_k + \lambda_{m,k} g_k b^\dagger_k), \tag{36}$$

which allows writing

$$e^{-it(\mathcal{H}_B+\mathcal{H}_{SL})}|m\rangle = e^{-it[\mathcal{H}_B+\mathcal{H}_{SL}(m)]}|m\rangle \tag{37}$$

(remember that $\mathcal{H}_B|m\rangle = |m\rangle\mathcal{H}_B$). From Eqs. (6) and (36), the exponent at Eq. (37) can be written as a sum over different modes k as

$$\mathcal{H}_B + \mathcal{H}_{SL}(m) = \sum_k \gamma_{k,m}, \tag{38}$$

where

$$\gamma_{k,m} \equiv \omega_k b^\dagger_k b_k + \lambda^*_{m,k} g^*_k b_k + \lambda_{m,k} g_k b^\dagger_k. \tag{39}$$

Since terms with different k commute, the time dependence of the density operator of Eq. (5) becomes

$$\begin{aligned}\sigma_{mn}(t) &= \langle m|V_0(t)\sigma(0)V^\dagger_0(t)|n\rangle \\ &\quad\times \prod_k \{\mathrm{Tr}_k[e^{-i\gamma_{k,m}t}\,\Theta_k\, e^{i\gamma_{k,n}t}]\},\end{aligned} \tag{40}$$

where the trace over the phonon modes k comes from the trace over the states of the heat bath. Eq. (40) has the same structure of Eq. (2.17) from Ref. [18], but here the coefficients $\gamma_{k,m}$ have a definite meaning in terms of the lattice parameters. The key problem now is how to calculate such trace. Some authors solve spin-boson problems by calculating an explicit expression for the time evolution operator [14,38], an alternative strategy uses the coherent states for the harmonic oscillator to calculate the traces [18,39]. We adopted the latter strategy.

Coherent states $|z\rangle$ are eigenstates of the annihilation operator b_k [40]. The trace of an operator in the basis $\{|z\rangle\}$ can be calculated with the following integral [41],

$$\mathrm{Tr}(O) = \int d^2z\langle z|O|z\rangle, \tag{41}$$

with

$$d^2z = \frac{1}{\pi} d(\mathrm{Re}\, z)\, d(\mathrm{Im}\, z). \tag{42}$$

Using that

$$1 = \int d^2z|z\rangle\langle z|,$$

we get for arbitrary k

$$\begin{aligned}&\mathrm{Tr}_k[e^{-i\gamma_{k,m}t}\,\Theta\, e^{i\gamma_{k,n}t}] \\ &\quad= \frac{1}{Z_k}\int d^2z_1 d^2z_2 d^2z_3 \langle z_1|e^{-i\gamma_{k,m}t}|z_2\rangle \\ &\qquad\times \langle z_2|e^{-\beta\hbar\omega_k b^\dagger_k b_k}|z_3\rangle\langle z_3|e^{i\gamma_{k,n}t}|z_1\rangle.\end{aligned} \tag{43}$$

It is well known that [41]

$$\langle z_2|e^{-\beta\hbar\omega_k b^\dagger_k b_k}|z_3\rangle = e^{z^*_2(e^{-\beta\hbar\omega_k}-1)z_3}\langle z_2|z_3\rangle. \tag{44}$$

In order to calculate the two other matrix elements of the complex exponential operators in Eq. (43), it is convenient to introduce the Bogoliubov shifted operators

$$\eta = b + \frac{\lambda_m g}{\omega}, \tag{45}$$

which allows writing

$$\gamma_m = \omega\eta^\dagger\eta - \frac{|\lambda_m|^2|g|^2}{\omega} \tag{46}$$

to obtain

$$\begin{aligned}\langle z_1|e^{-i\gamma_m t}|z_2\rangle &= e^{it\frac{|\lambda_m|^2|g|^2}{\omega}}\langle z_1|z_2\rangle \\ &\quad\times e^{\left(e^{-i\omega t}-1\right)(z^*_1+\frac{\lambda^*_m g^*}{\omega})(z_2+\frac{\lambda_m g}{\omega})}.\end{aligned} \tag{47}$$

In this way, calculation of the trace in Eq. (43) implies calculating six Gaussian integrals. The result is analogous to the one reported by Ref. [18], that is,

$$|\sigma_{mn}(t)| = |\langle m|V_0(t)\sigma(0)V^\dagger_0(t)|n\rangle|e^{-\Gamma_{(1)mn}(t)}, \tag{48}$$

where $\Gamma_{(1)mn}(t)$ is the decoherence function

$$\begin{aligned}\Gamma_{(1)mn}(t) &= \sum_k \left|\lambda_{m,k} - \lambda_{n,k}\right|^2 \frac{|g(\omega_k)|^2}{\omega^2_k} \\ &\quad\times (1-\cos\omega_k t)\coth\left(\frac{\beta\hbar\omega_k}{2}\right),\end{aligned} \tag{49}$$

where ω_k corresponds to the acoustic branch because of the adiabatic approximation, as was commented in the paragraph after Eq. (29).

It is worth emphasizing that a relevant feature of having treated the chain of pairs as an open quantum system is that the decoherence rate [see Eq. (49)] is parameterized by the eigenvalues of the spin part of the interaction Hamiltonian. The consequence of considering a correlated (observed) system is the occurrence of the phase factor involved in $|\lambda_{m,k} - \lambda_{n,k}|$.

A. Analysis of the decoherence function

In this section we work in the expression of the decoherence function in order to analyze its time dependence and estimate a decoherence time in terms of realistic values of the lattice parameters.

Let us write Eq. (49) as

$$\Gamma_{(1)mn}(t) = \left(\frac{\Omega_D}{d}\right)^2 \sum_k \mathcal{E}_{(1)mn}(k)\, h(k,t)\, f(k), \tag{50}$$

where we defined three functions $\mathcal{E}_{(1)mn}(k)$, $h(k,t)$, and $f(k)$ to consider their behavior separately. It is worth to recall that the maximum value of k is π/a.

$$h(k,t) \equiv \frac{1-\cos(ckt)}{c^2k^2} = \frac{t^2}{2}\mathrm{sinc}^2\left(\frac{ckt}{2}\right), \tag{51}$$

which is peaked at $k=0$ and its first minimum depends on time as $k_h = \frac{2\pi}{ct}$.

$$\begin{aligned}f(k) &\equiv \coth\left(\frac{\beta\hbar ck}{2}\right)|g(\omega_k)|^2 \\ &= \coth\left(\frac{\beta\hbar ck}{2}\right) 2\,(1-\cos kd)\,\frac{\hbar}{2mckN}\end{aligned} \tag{52}$$

there exists an interval $(0,k_f)$ where

$$\coth\frac{\beta\hbar ck}{2}\sim\left(\frac{\beta\hbar ck}{2}\right)^{-1}, \tag{53a}$$

$$1-\cos(kd)\sim\frac{(kd)^2}{2}, \tag{53b}$$

and the function $f(k)$ can be approximated by its small-k value, $\tilde{f}(k)$

$$\tilde{f}\sim\frac{d^2k_BT}{mNc^2}. \tag{54}$$

The function

$$\begin{aligned}\mathcal{E}_{(1)mn}(k)&\equiv\left(\frac{d}{\Omega_D}\right)^2\left|\lambda_{m,k}-\lambda_{n,k}\right|^2\\&=\left(\frac{d}{\Omega_D}\right)^2\left|\sum_A e^{-ikAa}(\lambda_{m_A}-\lambda_{n_A})\right|^2\end{aligned} \tag{55}$$

has a complex dependence on its variables. Its dependence on the spin states $|m\rangle$ and $|n\rangle$ also includes the particular spatial distribution of the spins involved in the transition $m\to n$. It is worth noticing that the phase factor in Eq. (55) arises as a consequence of the central assumption of our model, which is to consider that the pairs are correlated through the lattice vibrations. This characteristic distinguishes our proposal from the view of single (uncorrelated) particles coupled to a boson reservoir.

A direct calculation of $\mathcal{E}_{(1)mn}$ would involve a sum over the 4^N spin states, and the knowledge of their spatial distribution. Therefore, we must seek convenient approximations to estimate the possible generic outcomes. The four eigenvalues λ_{m_A} of operator Λ^A [from Eq. (22)] of each A pair are

$$\begin{aligned}\Lambda^A|1,1\rangle&=0.5\frac{\Omega_D}{d}|1,1\rangle\\\Lambda^A|1,0\rangle&=-1\frac{\Omega_D}{d}|1,0\rangle\\\Lambda^A|1,-1\rangle&=0.5\frac{\Omega_D}{d}|1,-1\rangle\\\Lambda^A|0,0\rangle&=0.\end{aligned} \tag{56}$$

From Eq. (55) it is clear that $\mathcal{E}_{(1)mm}=0$ and accordingly $\Gamma_{(1)mm}=0$, which is a consequence of (2). This characteristic is similar to the effect of eigenselection observed in Refs. [42] and [43]. However, from Eq. (56) we see that $\mathcal{E}_{(1)mn}=0$ for some particular states, such as $|m\rangle=|1,1\rangle\otimes|1,1\rangle\ldots\ |1,1\rangle$ and $|n\rangle=|1,-1\rangle\otimes|1,-1\rangle\ldots\ |1,-1\rangle$. In a more general case $\mathcal{E}_{(1)mn}$ may involve up to N nonzero terms. Let us call M the number of the terms that contribute to the sum over A, that is, the number of active spin pairs whose eigenvalues change when going from states $|m\rangle$ to $|n\rangle$. For the sake of simplicity, let us assume that the active spins are uniformly distributed throughout the lattice, at distance $P\,a$, and that the coefficients $(\lambda_{m_A}-\lambda_{n_A})\sim\frac{\Omega_D}{d}$. In such a case

$$\mathcal{E}_{(1)mn}\sim\left|\sum_{A=0}^{M-1}e^{-ikAPa}\right|^2=\frac{1-\cos(MkPa)}{1-\cos(kPa)}. \tag{57}$$

It is worth noticing that $\mathcal{E}_{(1)mn}$ is a squared sinclike function, centered around $k=0$, with a smooth decay throughout the relevant k range and whose first minimum occurs at $k_{\mathcal{E}}=\frac{2\pi}{MPa}$.

In the continuum limit we integrate over $k>0$ due to the parity of the integrand, thus, Eq. (50) transforms into

$$\begin{aligned}\Gamma_{(1)mn}(t)&=\frac{\alpha\Omega_D^2}{d^2}\int_0^{k_f}\mathcal{E}_{(1)mn}(k)\frac{(ct)^2}{2}\mathrm{sinc}^2\left(\frac{ckt}{2}\right)G(k)dk\\&=C_{(1)}\int_0^{k_f}\frac{(ct)^2}{2}\frac{1-\cos(MkPa)}{1-\cos(kPa)}\mathrm{sinc}^2\left(\frac{ckt}{2}\right)dk,\end{aligned} \tag{58}$$

where

$$\alpha\equiv\frac{2d^2k_BT}{mNc^4};\qquad C_{(1)}=\frac{\alpha\Omega_D^2Na}{d^2\pi} \tag{59}$$

and

$$G(k)dk=\frac{Na}{\pi}dk$$

is the number of states between k and $k+dk$ in a linear chain. It is now useful to compare the cutoff values k_h, $k_{\mathcal{E}}$, and k_f [of functions $h(k,t)$, $\mathcal{E}_{(1)mn}(k)$, and $f(k)$, respectively] in order to determine the behavior of the integrand of Eq. (58). Then we assign some reasonable values to the different constants involved. We choose $d=0.1$nm, which, for example, is of the order of magnitude of the distance between hydrogen nuclei in the water molecules in hydrated salts; the interpair distance $a=10d$; $m=1.66\times10^{-27}$kg, is the mass of a hydrogen atom, $\Omega_D=300$ kHz corresponds to a typical dipolar frequency in a solid, $T=300$ K and for the speed of sound in a solid we use $c=3000$ m/s.

In this way, Eq. (53a) is satisfied provided $k\ll\frac{2}{\beta\hbar c}=2.6\times10^{10}m^{-1}$, while the condition imposed by Eq. (53b) is $k\ll\frac{1}{d}=10^{10}m^{-1}$. Then, we can set $k_f\sim10^9m^{-1}$. The cutoff related with $h(t)$ is $k_h=\frac{2\times10^{-3}}{t}m^{-1}$, which satisfies $k_h\ll k_f$ for times larger than 0.1 ns. To evaluate $k_{\mathcal{E}}$ we have to assign some value for M and P. An estimation of M over a large random selection of states indicates that $M=10^3$ is a reasonable and representative value for a chain with $N\approx10^6$. Assigning $P\sim100$, we have $k_{\mathcal{E}}\sim6\times10^4$, which satisfies $k_h\ll k_{\mathcal{E}}$ for times larger than 0.1 μs. In order to illustrate this relationship, Fig. 2 compares $\mathcal{E}_{(1)mn}(k)$ with $h(k,t)$ as a function of the wave number, at different times. It can be seen that for times as short as a few tenths of microseconds, $h(k,t)$ dominates the k dependence of the integrand of Eq. (58) and that $\mathcal{E}_{(1)mn}(k)$ can be approximated by $\mathcal{E}_{(1)mn}(0)$.

These relations allow extending the upper limit of Eq. (58) to infinity to obtain an estimation of the time scale of this irreversible mechanism

$$\begin{aligned}\Gamma_{(1)}(t,M)&=C_{(1)}M^2\int_0^{\infty}\frac{1-\cos(ckt)}{k^2}dk\\&=\frac{C_{(1)}\pi c}{2}M^2\,t\simeq1.66\times10^{-2}\frac{1}{\mathrm{s}}\,M^2\,t.\end{aligned} \tag{60}$$

Equation (60) has an explicit dependence on temperature, however, there can also be an implicit dependence in other parameters for example due to the finite phonon lifetime and the speed of sound in the particular system.

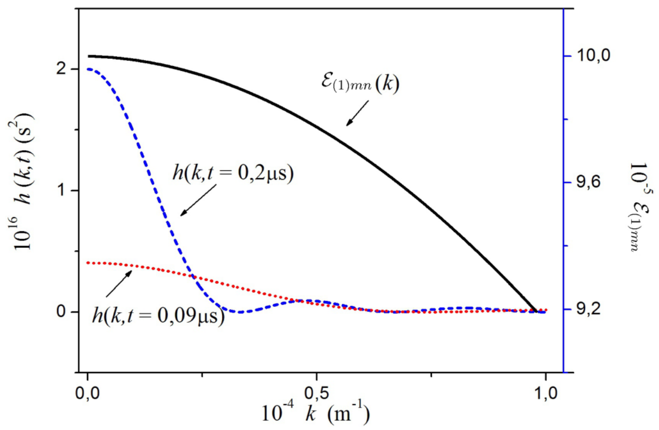

FIG. 2. Plot of functions $\mathcal{E}_{(1)mn}(k)$ (solid) and $h(k,t)$ as a function of the wave number, at different times: 0.2 μs (dashed) and 0.09μs (dotted).

Figure 3(a) shows a plot of the dependence of the decay function $D_{(1)}(t,M) \equiv e^{-\Gamma_{(1)}(t,M)}$ on time and on the number M of active spins, correlated by the phonon field. It can be seen that the function attenuates within the millisecond range, that is, an experimentally accessible time scale, provided $M \gtrsim 750$. Oppositely, decoherence becomes ineffective for small values of M. The limit $M = 1$ represents the traditional problem of single spin pairs coupled to a boson environment, where, as expected, the lattice phonons cannot induce pure dephasing [24]. Figure 3(b) shows a cut of Fig. 3(a) for the selected value $M = 1000$.

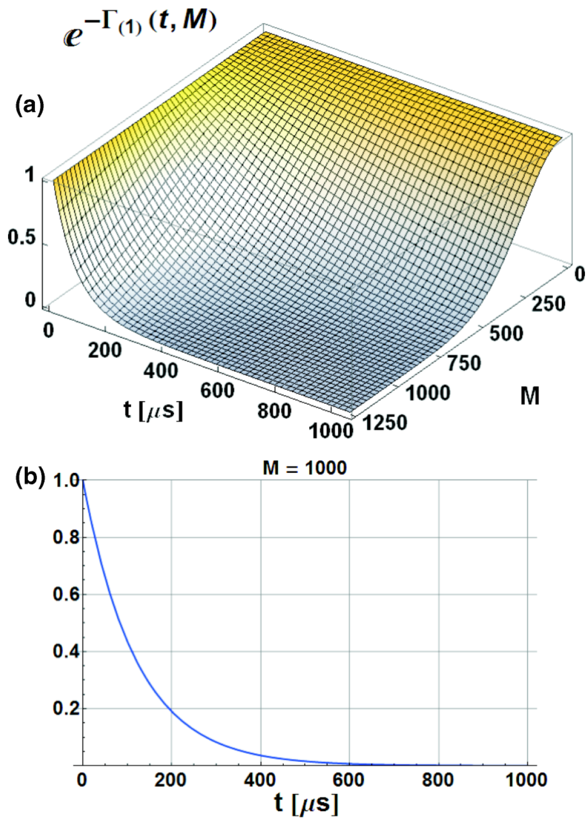

FIG. 3. Decay function $D_{(1)}(t,M) \equiv e^{-\Gamma_{(1)}(t,M)}$ in a linear chain of dipole interacting spin pairs. (a) Variation of $D_{(1)}(t,M)$ with time and the number M of active spin pairs, we can see that the function vanishes in the range of $t < 1000\mu s$ for values of $M \gtrsim 750$. (b) Cut of (a) for $M = 1000$.

Then, we can conclude that the coupling between spin pairs and phonons in a linear chain provides a mechanism of irreversible decoherence whose time scale depends on the degree of correlation of the involved states (parameterized by M).

Although we made a realistic approximation, it is important to show that the decoherence function diverges, independently of such approximations and of the chosen values of the parameters.

Lemma. $\lim_{t\to\infty} \Gamma_{(1)mn}(t) = \infty$ if $|\lambda_{m,k} - \lambda_{n,k}| \neq 0$.

Proof. In order to find a lower bound for the integral of Eq. (58), since the integrand is positive, we see that for an arbitrary wavelength ϵ,

$$\Gamma \propto \int_0^{\epsilon} (\ldots)dk + \int_{\epsilon}^{\pi/a} (\ldots)dk > \int_0^{\epsilon} (\ldots)dk. \tag{61}$$

By setting ϵ small enough, it is possible to replace the integrand by its low k value, which results in

$$\begin{aligned} \int_0^{\epsilon} (\ldots)dk &\propto \int_0^{\epsilon} \frac{1-\cos(ckt)}{k^2} dk \\ &= \frac{-1+\cos(\epsilon ct)+\mathrm{Si}(\epsilon ct)\ \epsilon ct}{\epsilon} \end{aligned} \tag{62}$$

where Si stands for the sine integral. Since the second line of Eq. (62) tends to infinity for increasing times, then

$$\begin{aligned} \Gamma_{(1)mn}(t) &> \frac{-1+\cos(\epsilon ct)+\mathrm{Si}(\epsilon ct)\ \epsilon ct}{\epsilon} \to \infty, \\ &\text{if } t \to \infty. \end{aligned} \tag{63}$$

This interesting result shows that the matrix elements of Eq. (40) attenuate because the lower bound for $\Gamma(t)$ attains large values as time grows, so proving that decoherence due to the interaction of a chain of dipole-coupled pairs of spins with a phonon bath is possible.

V. DECOHERENCE IN A 3D ARRAY OF SPIN PAIRS

Let us now extend the analysis to a three-dimensional array of pairs in order to inquire on the effect of dimensionality on the characteristics of the decoherence function. We consider a regular array of pairs of atoms, which bear the observable spins 1/2, which shares similar characteristics with the chain of pairs of Sec. III A. That is, a spin system having strong dipole-dipole coupling between spins of the same pair, while a weak coupling between different pairs. An example of such a structure can be a hydrated salt, where the water molecules are regularly distributed and adopt only few different orientations. For simplicity, we consider a case where the pairs are equally oriented within the unit cell so that the intrapair vectors are parallel, as in potassium oxalate monohydrate [44].

Let us define an ordered array whose symmetry is given by the primitive vectors $\hat{a}$, $\hat{b}$, $\hat{c}$. The observed pairs are placed at the lattice sites labeled by $\vec{A} = (A_a, A_b, A_c)$. The internuclear vector of pair $\vec{A}$ is $\vec{r}_{\vec{A}} = \vec{r}_{\vec{A}1} - \vec{r}_{\vec{A}2} = (r^x_{\vec{A}}, r^y_{\vec{A}}, r^z_{\vec{A}})$. If we call $\vec{r}_{0\vec{A}}$ the equilibrium intrapair vector and assume that all pairs are oriented along z, $\vec{r}_{0\vec{A}} = (0,0,d)$, $\forall \vec{A}$. With this notation, the corresponding intrapair interaction Hamiltonian for this 3D object, analogous to Eq. (14), is

$$\mathcal{H}_{Da} \approx \sum_{\vec{A}} \mathcal{H}_D^{\vec{A}}\bigg|_{\vec{r}_{\vec{A}}=\vec{r}_{0\vec{A}}} + \sum_{\vec{A}} \vec{\nabla}\mathcal{H}_D^{\vec{A}}\bigg|_{\vec{r}_{\vec{A}}=\vec{r}_{0\vec{A}}} \cdot (\vec{r}_{\vec{A}} - \vec{r}_{0\vec{A}}). \quad (64)$$

The dipolar Hamiltonian depends both on $r_{\vec{A}}^{-3}$ ($r_{\vec{A}} \equiv |\vec{r}_{\vec{A}}|$) and on the angle $\theta_{\vec{A}}$ between $\vec{r}_{\vec{A}}$ and the external magnetic field. However, the angular variations modify the dipolar interaction in a higher order of approximation than the one considered in Eq. (14), which allows us to consider only the variation with respect to the intrapair distance (we keep only the first order in the displacement from equilibrium and therefore on the creation and annihilation operators)[45].

Under this assumption, the gradient symbol in Eq. (64) is $\vec{\nabla} = (\frac{d}{dr_{\vec{A}}^x}, \frac{d}{dr_{\vec{A}}^y}, \frac{d}{dr_{\vec{A}}^z})$. The analog to Eq. (16) is

$$\vec{r}_{\vec{A}} - \vec{r}_{0\vec{A}} = \sum_{\vec{k}} \left(g_{\vec{k}}^{\vec{A}*} b_{\vec{k}} + g_{\vec{k}}^{\vec{A}} b_{\vec{k}}^{\dagger}\right)\hat{z}, \quad (65)$$

with $\hat{z}$ a unit vector, and the coupling constants of the 3D array

$$g_{\vec{k}}^{\vec{A}} = e^{i\vec{k}\cdot\vec{r}_{\vec{A}1}}(1 - e^{ik_z d})\sqrt{\frac{\hbar}{2mN\omega_k}}.$$

Notice that $\vec{k}$ is a reciprocal lattice vector, and that the scalar product of Eq. (64)

$$\vec{\nabla}\mathcal{H}_D^{\vec{A}}\bigg|_{\vec{r}_{\vec{A}}=\vec{r}_{0\vec{A}}} \cdot \hat{k} = \frac{d\mathcal{H}_D^{\vec{A}}}{dr_{\vec{A}}}\bigg|_{r_{\vec{A}}=d} \cos\theta_{\vec{k}} \quad (66)$$

introduces the angle $\theta_{\vec{k}}$ between $\hat{k}$ and $\hat{z}$.

The interaction Hamiltonian of this 3D model is then

$$\mathcal{H}_{SL} = \sum_{\vec{A}} \Lambda^{\vec{A}} \otimes \sum_{\vec{k}} \left(g_{\vec{k}}^{\vec{A}*} b_{\vec{k}} + g_{\vec{k}}^{\vec{A}} b_{\vec{k}}^{\dagger}\right)\cos\theta_{\vec{k}}, \quad (67)$$

where both the sum over $\vec{k}$ and over $\vec{A}$ are now triple sums, which run over the reciprocal and the direct lattice indices, respectively. Using a similar procedure to that of Sec. IV, we can deduce the decoherence function

$$\Gamma_{(3)mn}(t) = \sum_{\vec{k}} \left|\lambda_{m,\vec{k}} - \lambda_{n,\vec{k}}\right|^2 \frac{|g(\omega_{\vec{k}})|^2}{\omega_{\vec{k}}^2} \times \left(1 - \cos\omega_{\vec{k}} t\right) \coth\left(\frac{\beta\hbar\omega_k}{2}\right) \cos^2\theta_{\vec{k}}, \quad (68)$$

where $\omega_{\vec{k}} = ck$, with $k \equiv |\vec{k}|$.

Similarly to the 1D case, we define functions $h(k,t)$ and $f(k)$, which now depend on the magnitude of the reciprocal lattice vectors. The counterpart of $\mathcal{E}_{(1)mn}(k)$ takes the form

$$\mathcal{E}_{(3)mn}(\vec{k}) \equiv \left(\frac{d}{\Omega_D}\right)^2 \left|\sum_{\vec{A}} e^{-i\vec{k}\cdot\vec{A}}(\lambda_{m\vec{A}} - \lambda_{n\vec{A}})\right|^2, \quad (69)$$

where $\vec{A} = (A_x a_x, A_y a_y, A_z a_z)$. The integer index A_r, with $r \equiv x,y,z$, runs over the whole sample, and a_r is the separation between the spin pairs along x, y, or z. In Eq. (69), the phase factor depends both on the magnitude and orientation of $\vec{k}$, which suggests that the spatial distribution of active spin pairs is even more significant in the 3D case.

Another important difference with respect to the 1D calculation arises when considering the continuum case, since the density of states is now

$$G_{(3)}(\vec{k})\, d^3k = \frac{Na^3}{\pi^3} k^2 \sin\theta_{\vec{k}}\, dk\, d\theta_{\vec{k}}\, d\phi,$$

then

$$\Gamma_{(3)mn}(t) = \frac{\alpha\Omega_D^2}{d^2} \iiint \mathcal{E}_{(3)mn}(\vec{k}) \sin^2\left(\frac{ckt}{2}\right) G_{(3)}(\vec{k})\, d^3k, \quad (70)$$

with α as in Eq. (59), but with N as the total amount of spin pairs in the 3D array.

Integrating over the angular variables

$$\Gamma_{(3)mn}(t) = C_{(3)} \int_0^{k_f} \overline{\mathcal{E}}_{(3)mn}(k)\ \sin^2\left(\frac{ckt}{2}\right) dk, \quad (71)$$

where

$$C_{(3)} = C_{(1)} \frac{a^2}{\pi^2},$$

and we defined

$$\overline{\mathcal{E}}_{(3)mn}(k) \equiv \int_0^{2\pi} d\phi \int_0^{\pi} d\theta_k\, \mathcal{E}_{(3)mn}(\vec{k})\ \sin\theta_{\vec{k}} \cos^4\theta_{\vec{k}}, \quad (72)$$

In order to proceed with the integration in Eq. (70) we need an expression for the dependence of $\mathcal{E}_{(3)mn}$ on k,θ, and ϕ for the chosen states m and n. Then, we follow a similar reasoning that led us to Eq. (57) and consider the case where the difference of eigenvalues of the states of all the active spin pairs are equal to $\frac{\Omega_D}{d}$. In this case, Eq. (72) takes the form

$$\overline{\mathcal{E}}_{(3)MP}(k) = \int_0^{\pi} d\theta_k\, S_{(z)MP}(k,\theta_k)\ \sin\theta_{\vec{k}} \cos^4\theta_{\vec{k}} \times \int_0^{2\pi} d\phi\, S_{(x)MP}(k,\theta_{\vec{k}},\phi)\, S_{(y)MP}(k,\theta_{\vec{k}},\phi), \quad (73)$$

where we defined

$$S_{(r)MP}(k,\theta_{\vec{k}},\phi) \equiv \left|\sum_{A_r=0}^{M-1} e^{-i\, k_r A_r P_r a_r}\right|^2 = \frac{1 - \cos(Mk_r Pa)}{1 - \cos(k_r Pa)},$$

with $r \equiv x,y,z$ and $k_z = k\cos\theta_{\vec{k}}$, $k_x = k\sin\theta_{\vec{k}}\cos\phi$, $k_y = k\sin\theta_{\vec{k}}\sin\phi$. For simplicity we consider that $a_x = a_y = a_z = a$, the number M of active spin pairs is the same along the different axes, and that they are uniformly separated by P pairs.

The point to highlight is that the presence of angular variables prevents the simultaneous occurrence of a maximum in the three functions $S_{(x)}$, $S_{(y)}$, and $S_{(z)}$, except for $k = 0$, consequently the outcome of the integral over the angular variables, which we now call $\overline{\mathcal{E}}_{(3)MP}(k)$, is a globally decreasing function of k, which vanishes as k grows. An analytical solution of Eq. (73) is complex, but it can also be performed numerically. The dotted line (blue) in Fig. 4 is the plot of $\overline{\mathcal{E}}_{(3)MP}(k)/M^6$ with $M = P = 12000$. The solid line (red) is

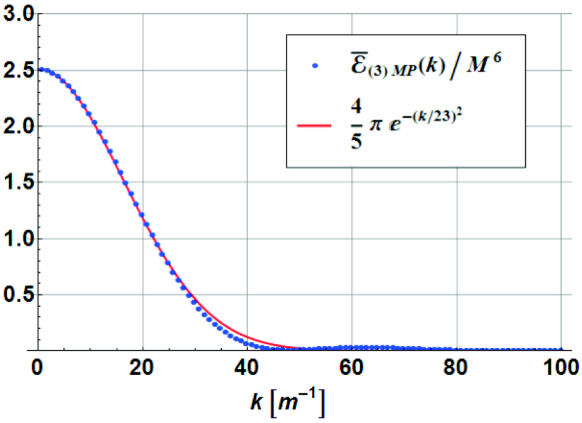

FIG. 4. Dotted line (blue): numerical calculation of the function $\overline{\mathcal{E}}_{(3)MP}(k)/M^6$ for $M = P = 12000$. Solid line (red): function $\frac{4}{5}\pi e^{-(k/k_c)^2}$ with $k_c = 23m^{-1}$. The similarity between the two functions indicates that the numerical result can be well represented by a Gaussian function.

a Gaussian fit, which practically coincides with the numerical result, showing that $\overline{\mathcal{E}}_{(3)MP}(k)$ can be well represented by a Gaussian function.

It is important to note that Eq. (71) is the integral of a sinusoidal function modulated by $\overline{\mathcal{E}}_{(3)mn}(k)$. As depicted in Fig. 4, this is a peak function that acts as a low-pass filter with a cutoff frequency k_c, which depends on M and P. With the aim of analyzing the general behavior of the decoherence function, we represent Eqs. (72) and (73) by a Gaussian function of the form

$$\overline{\mathcal{E}}_{(3)MP}(k) = \overline{\mathcal{E}}_{(3)MP}(0)\, e^{-(k/k_c)^2}, \tag{74}$$

where $\overline{\mathcal{E}}_{(3)MP}(0) = \frac{4}{5}\pi M^6$, is the value at $k = 0$.

Now, using Eq. (74) in Eq. (71), we obtain the decoherence function corresponding to this model

$$\widetilde{\Gamma}_{(3)}(t,M,k_c) = C_{(3)} \frac{\pi^{3/2}}{5} M^6\, k_c \left(1 - e^{-\frac{1}{4}c^2 k_c^2 t^2}\right). \tag{75}$$

Accordingly, the decay function $D_{(3)}(t,M,k_c) \equiv e^{-\widetilde{\Gamma}_{(3)}(t,M,k_c)}$ depends on time, on the number of active pairs M and on an effective cutoff wave number k_c. In Figs. 5(a) and 5(b) we show the time dependence of $\widetilde{\Gamma}_{(3)}(t,M,k_c)$ and $D_{(3)}(t,M,k_c)$, respectively, for $M = 12000$ and $k_c = 3 \times 10^{-9}\pi/a \simeq 9.425m^{-1}$.

Since the values of M and k_c have a complex dependence on the states m,n and on the spatial distribution of the active pairs, it is convenient to illustrate the behavior of the decay function in a wide range of variation of the three variables. Figure 6(a) shows the variation of $D_{(3)}$ with k_c and M for $t = 100\mu s$ (which is a representative time scale in NMR experiments), to highlight the attenuation due to irreversible decoherence. We can see that $D_{(3)}$ appreciably attenuates in the contour bounded by $M > 11000$ and $k_c > 3m^{-1}$. Figure 6(b) illustrates the dependence on t and k_c for $M = 12000$, while Fig. 6(c) shows the dependence on t and M for $k_c = 3 \times 10^{-9}\pi/a \simeq 9.425m^{-1}$.

These results show that in the 3D case the decoherence function has a different time behavior than in the 1D case. The decoherence function does not grow indefinitely with time but instead it converges to a constant value which depends on M and k_c. That is, it depends on the degree of correlation of the involved states in a more intricate way than in the 1D case. However, we observe that there exists a range of values of the parameters M and k_c where the decay function attains very low values in a given time scale. Thus, we can conclude that the coupling between phonons and a 3D array of dipole-interacting spin pairs provides a mechanism of irreversible decoherence.

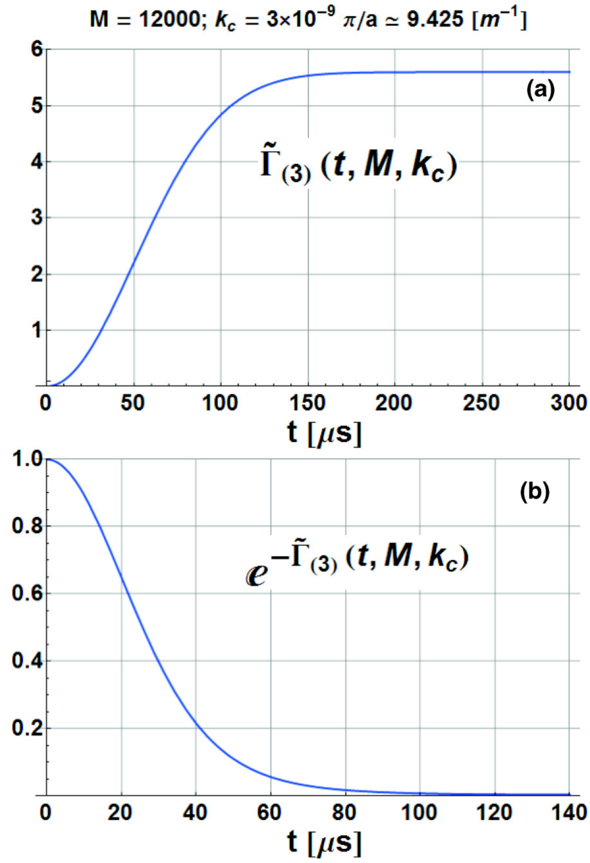

FIG. 5. (a) Time dependence of the decoherence function $\widetilde{\Gamma}_{(3)}(t,M,k_c)$ for a 3D array of dipolar interacting spin pairs, for the values of $M = 12000$ and $k_c = 3 \times 10^{-9}\pi/a \simeq 9.425m^{-1}$. (b) Decay function $D_{(3)}(t,M,k_c)$ corresponding to (a).

VI. DISCUSSION

The main result of this work is the finding that the microscopic spin dynamics of an array of weakly interacting spin pairs, quantum mechanically coupled with a phonon bath, can undergo irreversible, adiabatic decoherence over an intermediate time scale, earlier than the one dictated by thermal processes. The starting point is a generalization of the usual spin-boson model that allowed us to include the interacting nature of the observed system as a main feature. This approach, though simple, enables us to display the effect of the system-environment quantum correlation on the adiabatic decoherence. In this framework it was possible to derive the time dependence of the reduced density matrix without resorting to coarse-graining procedures for 1D and 3D models.

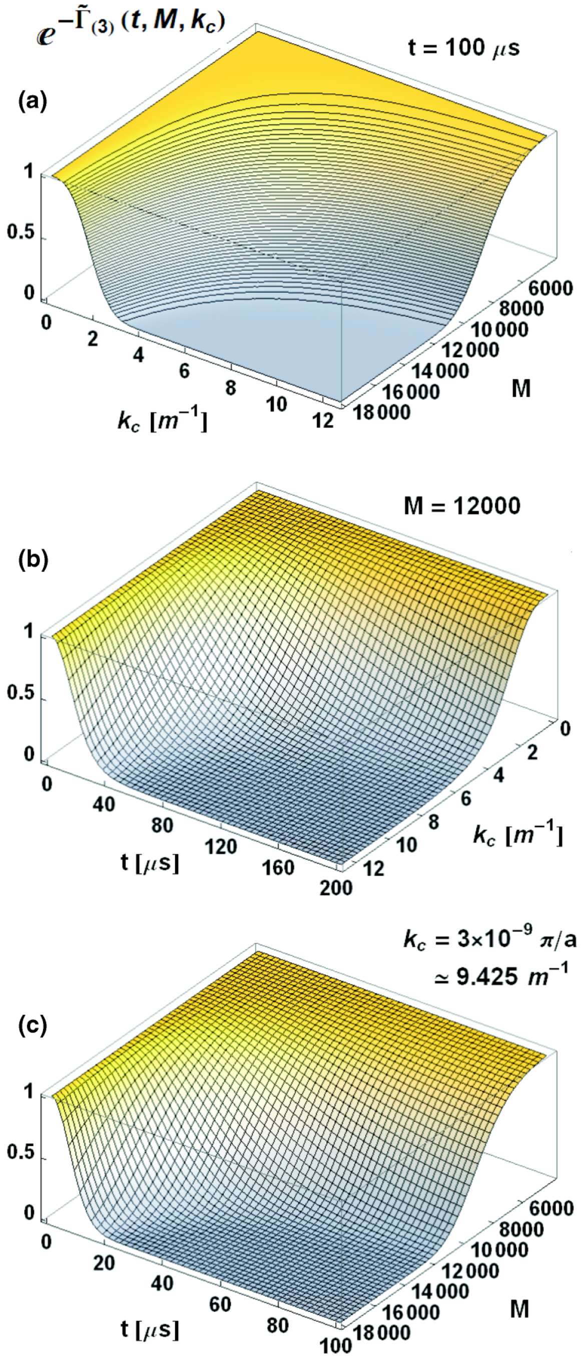

FIG. 6. Decay function $D_{(3)}(t,M,k_c) \equiv e^{-\Gamma_{(3)}(t,M,k_c)}$ for different ranges of its variables. (a) Dependence on the cutoff wave number k_c and the number M of active spin pairs at a fixed time $t = 100\mu s$. The function appreciably attenuates in the region bounded by the contour $M > 11000$ and $k_c > 3m^{-1}$. (b) Dependence on time and k_c for fixed $M = 12000$. (c) Dependence on time and M for fixed $k_c = 3 \times 10^{-9}\pi/a \simeq 9.425m^{-1}$. In (a) and (c) the lowest shown value on the M axis is 4000.

The traditional spin-boson model considers individual spins interacting with a boson field. The spin-environment coupling is represented through a Hamiltonian linear in the boson variables, which strictly satisfies the adiabatic condition. In the continuum limit, the product of the density of states and the coupling factors is condensed in a spectral density $J(\omega)$, and the effect of the interaction is thought of as a quantum noise disturbing the two-level system [15,46].

Alternatively, to take into account the many-body character of the open system, our proposal introduces the coupling with the environment through the small variations of the dipole-dipole interaction produced by acoustic phonons. The functions $\mathcal{E}_{(1)mn}(k)$ and $\mathcal{E}_{(3)mn}(\vec{k})$ represent the correlation between spin variables and phonon phases and reflect the properties of the distribution of spin eigenvalues throughout the network. The dependence on the phonon wave numbers is in principle different for every election of the states m and n. This model, based on using a spin-environment coupling, which naturally involves physical magnitudes, allows obtaining reliable estimates of decoherence rates and paves the way to the calculation of measurable quantities.

The functions $\mathcal{E}_{mn}$ arise as a signature of decoherence of an open quantum many-body system. They contain in a mingled way the complexity of the observed system and the correlation of the environment. We noted that the influence of complexity strongly depends on the dimensionality of the spin array. In fact, in one dimension the decoherence rate depends on the number of active spin pairs, which determines the scale of the decoherence function. In the 3D case, the correlation introduces a new ingredient: a low-frequency cutoff whose value regulates the efficiency of the irreversible decoherence. Clearly, this parameter has a microscopic origin and is related with the entanglement between the spin and lattice variables. This differs from the noninteracting models, where a cutoff is introduced to modulate the frequency dependence of the spectral density.

It is worth to mention that in spite of having excluded the variation of the inter-pair dipolar interactions from the interaction Hamiltonian, the calculated dynamics of reduced density operator still reflects the mechanical correlation of the different pairs established by the phonon displacement field. A similar discussion was given previously [42,43] in the study of decoherence in nematic liquid crystals, where the adiabatic decoherence function reflects the influence on a given molecule exerted by the other molecules, through their quantum mechanical interaction, even when molecules are magnetically isolated in the average.

It can be seen from Eqs. (5) and (48) that the decoherence mechanism and the multispin character of the observed system are complementary aspects of the irreversible dynamics of a system observable. Because of the multispin character of $\mathcal{H}_S$, states $|m\rangle$ and $|n\rangle$ are not eigenstates of the evolution operator $V_0(t)$, then the closed-system dynamics represented by $V_0(t)\sigma(0)V_0^{\dagger}(t)$, connects eigenstates of H_{SL} where the number of spins that change their state in an $m \rightarrow n$ transition can be large. This correlation grows with time [28,47] at a rate that depends on the particular dipolar network. On other hand, the decoherence function Γ_{mn} also depends on the number of active spins and is responsible for the irreversible decay of the reduced matrix element σ_{mn}. This microscopic irreversibility could convey an irreversible behavior to an observable of the spin system $\langle O(t)\rangle = \mathrm{Tr}[\sigma(t)O]$.

The estimation of the decoherence time scale was made by using realistic values of the various parameters involved. The order of magnitude obtained in Eq. (60) and Eq. (75), showed consistent with decoherence rates measured in NMR refocusing experiments, such as the magic echo and MREV pulse sequences [28,43]. Also, the obtained values are compatible with the time scale associated with the generation of quasiequilibrium [9].

The fact that the decoherence function is controlled by the low-frequency modes, as discussed in Sec. IV A, is completely consistent with the adiabatic hypothesis formulated at the beginning, by which the relevant spin-environment coupling in the early time scale corresponds to the long wavelength portion of the anisotropic dipole-phonon Hamiltonian. Ultimately, the occurrence of observable irreversible adiabatic decoherence along this time scale will depend on basic features of the system and its environment, as complexity of the dipole network, symmetry properties, and dimensionality of the lattice, etc.

The explanation of irreversible decoherence in solid-state NMR has remained as an open question for a long time. Particularly, the mechanism by which nuclear spins are able to achieve a state of quasiequilibrium continues to be elusive nowadays. The conclusions obtained in this work may contribute to this field. Also, treating spin coherence in the framework of open quantum systems can provide a complementary view on the dynamics of multiple quantum coherences in solid-state NMR [17,48], since including the system-environment quantum correlation allows exposing the relation between complexity and decoherence rates. Particularly, it was observed that coherences decay faster with increasing cluster size [49], which could be explained with the dependence on M of the decay function of our approach.

In NMR experiments the observed signal decay during the evolution under spin interactions is usually interpreted as a consequence of the transformation of single-quantum coherence terms into unobservable multiple-spin, single-quantum coherence terms, during the unitary evolution of the initial spin density matrix in a closed system [50,51]. The spin dynamics of an ideally isolated system is reversible and could, in principle, be perfectly retraced by means of an adequate experiment. This contrasts with the viewpoint of this work, where we conclude that the spin-environment quantum correlation can lead to a microscopic leakage of information. This subtle, purely quantum mechanism can produce an irreversible macroscopic effect whose efficiency depends on the correlation growth and the attenuation effects, which are ultimately governed by the spin-environment interaction Hamiltonian. We show that the coupling to low-energy phonon modes can in fact contribute to the adiabatic decoherence of a dipole interacting spin system, suggesting that this mechanism should be considered as a source of authentic irreversible attenuation of refocused echoes in NMR experiments.

The effectiveness of phonons to mediate spin decoherence contrasts with the null expected influence of dipole-phonon interaction in spin relaxation and thermalization [24,45,47]. This fact should not be surprising since decoherence and thermalization are processes of very different nature. Thermalization is generally described by Markovian master equations for the reduced density matrix that evolves in a coarse-grained time scale, with a rate controlled by thermal fluctuations of the spin-lattice interaction. By contrast, adiabatic decoherence is a full-quantum process that has as its main ingredient the quantum correlation between a many-body system and its environment, represented by the commutation properties of the interaction and bath Hamiltonians.

In summary, the model presented allows the estimation of decoherence rates in terms of parameters of the system. The system-environment coupling is introduced through the variations that the phonon field produces on the intrapair interaction energy, considered as the main contribution. Though the interaction Hamiltonian is a sum of individual terms, the strong correlation that exists between the pairs due to the phonon field is a decisive feature of the decoherence function. All the nonsecular terms of the interaction Hamiltonian not covered by this model could contribute to decoherence in a longer time scale. This example of decoherence of a many-body system allowed us to expose the role of complexity in the einselection process within the adiabatic time scale. Extending the description to the longer time scale beyond adiabaticity (still shorter than relaxation) would demand a comprehensive theory, such as the one proposed in Ref. [6].

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APPENDIX

Truncated inter-pair dipole-dipole coupling, from Ref. [35]

$$\widehat{\mathcal{H}}_{De} = \sum_{A\neq B} \overline{\Omega_D}(r_{A,B})\mathcal{T}^{AB}, \tag{A1}$$

with

$$\begin{aligned}\mathcal{T}^{AB} \equiv\ & 2T_{10}^{A}T_{10}^{B} + \tfrac{1}{2}\big(T_{11}^{A}T_{1-1}^{B} + T_{1-1}^{A}T_{11}^{B}\big)\\ & +4\big(T_{21}^{A}T_{2-1}^{B} + T_{2-1}^{A}T_{21}^{B}\big),\end{aligned} \tag{A2}$$

where the spherical tensors are

$$\begin{aligned}T_{10}^{A} &= I_z^{A1} + I_z^{A2}\\ T_{1\pm1}^{A} &= \mp\frac{1}{\sqrt{2}}(I_{\pm}^{A1} + I_{\pm}^{A2})\\ T_{2\pm1}^{A} &= \mp\frac{1}{2}[I_{\pm}^{A1}I_z^{A2} + I_z^{A1}I_{\pm}^{A2}],\end{aligned} \tag{A3}$$

and $\overline{\Omega_D}(r_{A,B})$ is an averaged dipolar coupling:

$$\begin{aligned}\overline{\Omega_D}(r_{A,B}) = \tfrac{1}{4}[&\Omega_D(r_{A_1,B_1}) + \Omega_D(r_{A_1,B_2})\\ &+ \Omega_D(r_{A_2,B_1}) + \Omega_D(r_{A_2,B_2})].\end{aligned} \tag{A4}$$

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