Strong-coupling theory for the spin-phonon model

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We present a strong-coupling approach to the spin-phonon model with a cubic spectral density, which relies on a matrix representation for the reduced time evolution operator on the four-dimensional space of pseudospins. The 4×4 self-energy matrix Σ is written as an infinite series in powers of bath fluctuation operators, and the lowest-order approximation is evaluated explicitly. The diagonal element Σ_{zz} corresponds to the result of the *noninteracting blip approximation* (NIBA); retaining the full matrix is equivalent to taking into account certain blip-blip interactions. Contrary to NIBA, this approach agrees with a rigorous result for the quadratic term of the rate. As a main result, we find a crossover to incoherent motion at a temperature T^*, which is compared to previous theoretical work and discussed in view of experimental data for various two-level tunneling systems. [S0163-1829(98)04201-5]

I. INTRODUCTION

Tunneling defects in various materials are well described in terms of quantum diffusion of a bistable system. As examples we note two level-systems in oxide glasses,[1,2] submicron metallic wires,[3,4] amorphous metals,[5,6] impurity ions on off-center positions in alkali halides (KCl:Li, KBr:CN, ...),[7,8] and interstitial hydrogen in niobium.[9,10]

These different situations are accounted for by a double-well potential for some collective coordinate q. At low temperatures only the ground states in the two wells, $|R\rangle$ and $|L\rangle$, are relevant. Then all quantum features reduce to a tunnel frequency $\Delta_{\rm b}$ which reads in WKB approximation $\Delta_{\rm b}=\omega_0\exp(-\sqrt{2mV_0d^2}/\hbar)$ with the particle mass m, the potential barrier V_0, and the distance of the wells d.

The quantum states $|R\rangle$ and $|L\rangle$ give rise to a two-level system whose operators are conveniently expressed in terms of Pauli matrices,

$$\sigma_z\equiv|L\rangle\langle L|-|R\rangle\langle R|,\quad \sigma_x\equiv|R\rangle\langle L|+|L\rangle\langle R|; \tag{1.1}$$

the discrete coordinate $q=\frac{1}{2}d\sigma_z$ takes the values $\pm\frac{1}{2}d$.

The simplest dynamic model is given by such a two-state pseudospin system whose reduced coordinate σ_z is linearly coupled to a heat bath,[11]

$$H'=\frac{1}{2}\hbar\Delta_{\rm b}\sigma_x+\frac{1}{2}\sigma_z\sum_k\hbar\lambda_k(b_k+b_k^\dagger)+\sum_k\hbar\omega_k b_k^\dagger b_k, \tag{1.2}$$

where the bath operators obey Bose commutation relations, $[b_k,b_{k'}^\dagger]=\delta_{kk'}$. (This paper is confined to the symmetric case where the two minima are degenerate, i.e., we discard an asymmetry energy.) The heat bath is entirely characterized by the spectral function

$$J(\omega)=\frac{\pi}{2}\sum_k\lambda_k^2\delta(\omega-\omega_k). \tag{1.3}$$

Equations (1.2) and (1.3) state the so-called spin-boson problem which, with an appropriate choice for the spectral function $J(\omega)$, accounts for various situations in solid-state physics and chemistry.[11] We discuss atomic tunneling only, where the frequency $\Delta_{\rm b}$ never exceeds a few Kelvin and hence is much smaller than the cutoff of the spectral function. Then $J(\omega)$ shows a simple power-law behavior in the relevant frequency range.[11] Two particularly interesting cases are defined by the linear and cubic spectral functions.

Ohmic damping. The linear case $J(\omega)=\pi K\omega$ leads to a frequency-independent damping function at low frequency and finite temperature; for this reason it is usually referred to as the case of Ohmic dissipation. It arises, as a most prominent example, from electron-hole excitations in a metal. As a most striking feature, a logarithmic infrared singularity arises in any order of perturbation theory from the linear frequency dependence. As a consequence, one finds a nonanalytical temperature dependence for the tunnel frequency, $\tilde{\Delta}_0\propto T^K$. At higher temperature there is a crossover from damped oscillations to overdamped motion,[11–17] with a relaxation rate that, in the weak-coupling regime $K\ll 1$, decreases with rising temperature, $\Gamma\propto T^{2K-1}$.

Phonon damping. The other case of physical relevance is realized in insulating materials where acoustic phonons provide the most efficient damping mechanism. In terms of the Debye model, the elastic waves obey the dispersion relation $\omega_{sk}=v_s|k|$, where s labels transverse and longitudinal polarization. When subsuming the wave vector k and the branch index s in the label k in Eq. (1.3), we recover the well-known expression

$$J(\omega)=\pi\alpha\omega^3, \tag{1.4}$$

where the coupling parameter α has dimension (frequency)$^{-2}$. It is related to material constants through

$$\alpha=\frac{1}{2\pi^2\hbar\varrho}\left(\frac{\gamma_{\rm l}^2}{v_{\rm l}^5}+\frac{2\gamma_{\rm t}^2}{v_{\rm t}^5}\right)\equiv\frac{1}{2\pi^2\hbar\varrho}\frac{3\gamma^2}{v^5}, \tag{1.5}$$

where v and γ are appropriate average values of sound velocities $v_{\rm t}$ and $v_{\rm l}$ and deformation potentials $\gamma_{\rm t}$ and $\gamma_{\rm l}$. The-

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cubic frequency dependence arises from the coupling constant, $\lambda_k \propto \sqrt{\omega_k}$, and the Debye density of states, $\Sigma_k \delta(\omega - \omega_k) = \text{const} \times \omega^2$. For further use we define a temperature

$$T_0 = (2\alpha)^{-1/2}(\hbar/\pi k_\mathrm{B}). \tag{1.6}$$

In the isotropic Debye model, a frequency cutoff for each phonon branch s is given by $v_s k_\mathrm{D}$. For later use we define the corresponding Debye temperatures Θ_s for N atoms in a volume V,

$$k_\mathrm{B}\Theta_s = \hbar v_s (6\pi^2 N/V)^{1/3}, \tag{1.7}$$

and the appropriate average value Θ,[18]

$$3/\Theta^3 = 1/\Theta_\mathrm{l}^3 + 2/\Theta_\mathrm{t}^3. \tag{1.8}$$

A heat bath with cubic spectral density leads to damping phenomena which are basically different from Ohmic dissipation. Whereas the latter case is determined by an infrared singularity, the cubic spectral function does not cause any anomaly at low frequency; yet as to the weight of thermal phonons, the increasing $J(\omega)$ causes a strong enhancement with temperature.

Equation (1.4) is not the only possible spectral density for phonon coupling. For an impurity site with inversion symmetry, the couplings vary as $\lambda_k \propto \omega_k^{3/2}$, which leads to $J(\omega) \propto \omega^5$, instead of the cubic law. The linear coupling potential in Eq. (1.2) can be considered as the first term of an expansion in powers of the elastic strain field. Taking into account the quadratic coupling potential would give rise to an Ohmic contribution to the bath spectral density[19–22] and hence add a different damping mechanism.

In this paper we address only the case of a cubic spectral function which, e.g., describes linar coupling to sound waves of defects in glasses. The model parameters are given by the tunnel frequency Δ_b, temperature T, the Debye temperature Θ, and the coupling strength α or the temperature T_0. Typical values for the latter are $T_0 \approx 5$ K for tunneling systems in amorphous SiO_2. On the other hand, the quantity $\hbar\Delta_\mathrm{b}/k_\mathrm{B}$ hardly exceeds a few K, thus satisfying

$$\alpha\Delta_\mathrm{b}^2 \ll 1. \tag{1.9}$$

We will frequently take advantage of this relation and drop small terms accordingly. There is no restriction with respect to temperature. Yet in order to simplify certain integrals, we will in general assume $T \ll \Theta$.

The dissipative dynamics of the spin-phonon model was tackled by path integral and functional integral methods,[11,17,23] diagrammatic perturbation theory,[24,25] and mode-coupling theory.[26–29] There seems to be general agreement on the behavior at low temperature. In this limit all mentioned works find weakly damped tunneling oscillations; both damping and relaxation rates involve the direct or one-phonon process only.

Yet with rising temperature, multiphonon processes are no longer small as compared to the direct process. For this domain, contradictory results have been obtained in different approaches. The controversial point may be cast in the question whether or not the thermal motion destroys the coherent tunnel oscillations.

On the one hand, it was claimed that damping of a two-level system by phonons is always weak, resulting in coherent tunneling motion at all relevant temperatures, i.e., $T \ll \Theta$.[11] On the other hand, several works indicated a crossover to incoherent motion at a much lower temperature; each of them, however, found a different relaxation rate in the incoherent regime.[24,25,29–31]

We close this introductory section with a short outline of the paper. In Sec. II we perform a canonical transformation which converts the Hamiltonian H' to that of a two-state polaron. Separating the static and fluctuating parts of the polaron operators $B_\pm$ provides the basis for the subsequent perturbation theory. In Sec. III, the relevant dynamic quantities and the reduced propagator are defined. The perturbation theory for the latter is set up in Sec. IV, where we develop a formal series expansion for the self-energy matrix in terms of the quantum Liouville operator; as an essential ingredient, this requires to define commutators and anticommutators as response and correlation operators, respectively. In Secs. V and VI we calculate explicitly the lowest-order approximation of the self-energy and the corresponding bath spectral functions.

Evaluating the propagator matrix in terms of a pole approximation in Sec. VII, permits us to derive the explicit time evolution in Sec. VIII. As a main result we find that, already in the noninteracting blip approximation (NIBA), there is a crossover to incoherent motion, and that in the coherent regime blip-blip interactions lead to a significant correction of the damping rate. In the final sections we discuss and summarize our results.

II. STRONG-COUPLING APPROACH

A perturbative treatment of the Hamiltonian (1.2) amounts to a power-series expansion in terms of λ_k^2 and provides an approximation that is valid for the weak-coupling limit. A calculation of the lowest-order term $\propto \lambda_k^2$ and the correction $\propto \lambda_k^4$ revealed, however, that the corresponding expansion parameter increases with temperature and that the perturbative approach breaks down at the temperature T_0.[31]

A proper treatment of the two-state dynamics then requires a strong-coupling approach, including terms of any order of the coupling parameter T^2/T_0^2. The canonical transformation

$$S = \exp\left[-\frac{1}{2}\sigma_z \sum_k u_k (b_k - b_k^\dagger) \right] \tag{2.1}$$

provides a representation that proves to be an appropriate starting point. Here we have defined

$$u_k = \lambda_k / \omega_k. \tag{2.2}$$

Applying Eq. (2.1) on the Hamiltonian (1.2) yields with $H = e^S H' e^{-S}$,

$$H = \frac{1}{2}\Delta_\mathrm{b}(\sigma_+ B_- + \sigma_- B_+) + \sum_k \hbar\omega_k b_k^\dagger b_k, \tag{2.3}$$

where we have dropped a constant and used

$$B_{\pm}=\exp\left[\pm\sum_k u_k(b_k-b_k^\dagger)\right]. \quad (2.4)$$

(The ladder operators $\sigma_+=|L\rangle\langle R|$ and $\sigma_-=|R\rangle\langle L|$ fulfill $\sigma_x=\sigma_++\sigma_-$.) Such a form is well known from studies on polaron motion[32] and dissipative two-state dynamics.[11]

The Hamiltonian contains the non-Hermitian operators $B_\pm$ and $\sigma_\pm$. For our purpose it becomes convenient to separate the average of $B_\pm$,

$$B=\langle B_+\rangle=\langle B_-\rangle, \quad (2.5)$$

from the fluctuations and to chose Hermitian combinations of the latter,

$$\xi_{\rm g}=\frac{1}{2}(B_++B_--2B), \quad \xi_{\rm u}=\frac{1}{2i}(B_+-B_-). \quad (2.6)$$

In order to set up a perturbation theory, we separate the Hamiltonian (2.3) into two parts,

$$H=H_0+H_1, \quad (2.7)$$

the first of which describes the uncoupled system,

$$H_0=\frac{1}{2}\hbar\tilde{\Delta}_0\sigma_x+\sum_k \hbar\omega_k b_k^\dagger b_k, \quad (2.8)$$

with the reduced tunnel energy

$$\tilde{\Delta}_0=B\Delta_{\rm b}; \quad (2.9)$$

the second term contains the interaction,

$$H_1=\frac{1}{2}\hbar\Delta_{\rm b}\sigma_x\xi_{\rm g}+\frac{1}{2}\hbar\Delta_{\rm b}\sigma_y\xi_{\rm u}. \quad (2.10)$$

The treatment of the two-state dynamics in this paper relies on a perturbation expansion in terms of the spin-phonon coupling H_1.

III. INITIAL STATE AND DYNAMIC QUANTITIES

Following Leggett *et al.*,[11] we consider a particle which dwells in the left well at $t=0$ and which evolves in time according to Eq. (2.7). As a consequence, the statistical operator at $t=0$ factorizes,

$$\rho=\rho_{\rm S}\rho_{\rm B}, \quad (3.1)$$

where the pseudospin part $\rho_{\rm S}=\frac{1}{2}(1+\sigma_z)$ projects on the quantum state $|L\rangle$. The remaining factor, $\rho_{\rm B}=e^{-\beta H_{\rm B}}/{\rm tr}(e^{-\beta H_{\rm B}})$, describes the heat bath in thermal equilibrium, with $H_{\rm B}=\Sigma_k\hbar\omega_k b_k^\dagger b_k$. The average with respect to Eq. (3.1) is denoted by angular brackets,

$$\langle\ \dots\ \rangle={\rm tr}(\ \dots\ \rho). \quad (3.2)$$

Time evolution is written in terms of the quantum Liouville operator $\mathcal{L}$, whose action on the density operator is given by the von Neumann equation

$$\dot{\rho}=-(i/\hbar)[H,\rho]\equiv-i\mathcal{L}\rho. \quad (3.3)$$

Formal integration yields

$$\rho(t)=e^{-i\mathcal{L}t}\rho. \quad (3.4)$$

Because of the factorization property of ρ, the thermal average over spin and bath degrees of freedom may be performed separately. Accordingly the time-dependent spin polarization may be written as

$$\langle\sigma_\alpha(t)\rangle=\langle\sigma_\alpha\mathcal{U}(t)\rangle, \quad (3.5)$$

where we have defined the reduced propagator

$$\mathcal{U}(t)=\langle e^{-i\mathcal{L}t}\rangle_{\rm B}, \quad (3.6)$$

the subscript B indicating a partial trace over bath coordinates, $\langle\cdots\rangle_{\rm B}={\rm tr}_{\rm B}(\cdots\rho_{\rm B})$.

The dissipative two-state dynamics is entirely determined by the time-dependent expectation values of σ_z and σ_x. Following Leggett *et al.*,[11] we define

$$P(t)=\langle\sigma_z(t)\rangle \quad (3.7)$$

as the time-dependent expectation value of the reduced two-state coordinate σ_z. With Eq. (3.1) we find the initial value $P(t=0)=1$. For zero-phonon coupling, $\lambda_k=0$, it shows coherent oscillations with the bare tunnel frequency $\Delta_{\rm b}$, $P(t)=\cos(\Delta_{\rm b}t)$. Taking into account the phonon coupling will lead to a reduced frequency, $\tilde{\Delta}_0$, and a loss of phase coherence in terms of an exponential damping factor.

The second quantity of interest is given by the expectation value

$$R(t)=\langle\sigma_x(t)\rangle. \quad (3.8)$$

In the case of zero-phonon coupling, we have $R(t)=0$ for all times, since σ_x is diagonal in the energy eigenstates of the uncoupled system. Taking into account the interaction with phonons results in a finite lifetime of the spin states. Then the average $R(t)$ provides two relevant quantities: In the long-time limit, it tends towards the equilibrium spin polarization, $R(t\to\infty)=\langle\sigma_x\rangle_{\rm eq}$. The corresponding relaxation time determines the lifetime of the spin states.

Note that we have not transformed the Pauli matrices; the Hamiltonian (2.7) is written in terms of the original operators σ_α. Since the reduced coordinate σ_z commutes with Eq. (2.1), $[S,\sigma_z]=0$, its time evolution is the same when calculated with respect to H' or H. Due to $[S,\sigma_x]\neq 0$, this statement does not hold true for σ_x. Equation (3.8) describes relaxation of a dressed two-state system in the adiabatic limit.

For later convenience we note the equations of motion for pseudospin operators, $\dot{\sigma}_\alpha=i\mathcal{L}\sigma_\alpha$, in terms of the reduced tunnel frequency $\tilde{\Delta}_0$ and the bath fluctuation operators (2.6)

$$\dot{\sigma}_x=\Delta_{\rm b}\xi_{\rm u}\sigma_z,$$

$$\dot{\sigma}_y=-\tilde{\Delta}_0\sigma_z-\Delta_{\rm b}\xi_{\rm g}\sigma_z,$$

$$\dot{\sigma}_z=\tilde{\Delta}_0\sigma_y+\Delta_{\rm b}\xi_{\rm g}\sigma_y-\Delta_{\rm b}\xi_{\rm u}\sigma_x. \quad (3.9)$$

IV. PERTURBATION SERIES

The perturbation series for $\mathcal{U}(t)$ is set up by splitting the Liouville operator according to Eq. (2.7) into two parts

$\mathcal{L}=\mathcal{L}_0+\mathcal{L}_1$, with $\hbar\mathcal{L}_0A=[H_0,A]$, etc., and by expanding the time evolution operator in terms of $\mathcal{L}_1$,

$$e^{-i\mathcal{L}t}=e^{-i\mathcal{L}_0t}-i\int_0^t d\tau e^{-i\mathcal{L}_0(t-\tau)}\mathcal{L}_1e^{-i\mathcal{L}_0\tau}-\int_0^t d\tau\int_0^\tau d\tau' \times e^{-i\mathcal{L}_0(t-\tau)}\mathcal{L}_1e^{-i\mathcal{L}_0(\tau-\tau')}\mathcal{L}_1e^{-i\mathcal{L}_0\tau'}+\cdots. \quad (4.1)$$

In order to obtain the reduced propagator (3.6), we divide $\mathcal{L}_0$ further in spin and bath parts, $\mathcal{L}_0=\mathcal{L}_S+\mathcal{L}_B$, which act on any composite operator A as $\mathcal{L}_SA=\frac{1}{2}\tilde{\Delta}_0[\sigma_x,A]$ and $\hbar\mathcal{L}_BA=[H_B,A]$. Since $\mathcal{L}_0$ does not contain the coupling term, the unperturbed time evolution factorizes,

$$e^{-i\mathcal{L}_0t}=e^{-i\mathcal{L}_St}e^{-i\mathcal{L}_Bt}. \quad (4.2)$$

The operator $\mathcal{U}(t)$ can be represented as a 4×4 matrix, which acts on a space spanned by the identity operator σ_0 and the three Pauli matrices σ_x, σ_y, σ_z,

$$\mathcal{U}_{ij}(t)=\frac{1}{2}\mathrm{tr}[\sigma_i\mathcal{U}(t)\sigma_j], \quad (4.3)$$

with $i,j=0,x,y,z$. [The element σ_0 is necessary in order to obtain a closed algebra with respect to multiplication, $\sigma_i\sigma_0=\sigma_i$ and $\sigma_i^2=\sigma_0$; note $\mathrm{tr}(\sigma_0)=2$.]

In order to link up with Eq. (4.3) we are going to represent all quantities appearing in the perturbation series as matrices. The unperturbed time evolution of spin operators is given by

$$\breve{\mathcal{U}}_{ij}(t)=\frac{1}{2}\mathrm{tr}(\sigma_ie^{-i\mathcal{L}_St}\sigma_j); \quad (4.4)$$

integrating the equation of motion (3.9) for zero-phonon coupling, one finds in a straightforward fashion

$$\breve{\mathcal{U}}(t)=\begin{pmatrix}1&0&0&0\\0&1&0&0\\0&0&\cos(\tilde{\Delta}_0t)&-\sin(\tilde{\Delta}_0t)\\0&0&\sin(\tilde{\Delta}_0t)&\cos(\tilde{\Delta}_0t)\end{pmatrix}. \quad (4.5)$$

The factors $\mathcal{L}_1$ in Eq. (4.1) involve composite operators whose spin parts develop according to Eq. (4.4). Yet each factor $\mathcal{L}_1$ acting as a commutator with the whole object to its right, gives rise to a more complicated action on the bath variables.

We note a general relation for composite operators S_iB_i with $[S_i,B_j]=0$,

$$[S_1B_1,S_2B_2]=\frac{1}{2}[S_1,S_2]\{B_1,B_2\}+\frac{1}{2}\{S_1,S_2\}[B_1,B_2], \quad (4.6)$$

where square brackets denote the commutator, and curly brackets the anticommutator.[33] The factors $\mathcal{L}_1$ in Eq. (4.1) are just commutators of H_1 with the expression to the right. According to Eq. (2.10), there are two terms, the first one with $S_1=\sigma_x$ and $B_1=\xi_g$ and the second one with $S_1=\sigma_y$ and $B_1=\xi_u$. The spin part of $\mathcal{L}_1$ may be represented by a 4×4 matrix,

$$i\Omega_{ij}=\frac{1}{2}\mathrm{tr}_S(\sigma_i\mathcal{L}_1\sigma_j), \quad (4.7)$$

whose entries still act on the bath variables. Using the commutator relations for composite operators and those for $\sigma_0,\sigma_x,\sigma_y,\sigma_z$, we calculate

$$i\Omega_{ij}=\mathcal{X}_{ij}\mathcal{B}_g^j+\mathcal{Y}_{ij}\mathcal{B}_u^j, \quad (4.8)$$

where $\mathcal{X}$ and $\mathcal{Y}$ are matrix elements of σ_x and σ_y,

$$\mathcal{X}_{ij}=\frac{1}{2}\mathrm{tr}(\sigma_i\sigma_x\sigma_j), \quad \mathcal{Y}_{ij}=\frac{1}{2}\mathrm{tr}(\sigma_i\sigma_y\sigma_j). \quad (4.9)$$

The parts acting on the bath variables read explicitly

$$\mathcal{B}_g^j=\begin{cases}\mathcal{R}_g & \text{for } j=0,x\\ \mathcal{C}_g & \text{for } j=y,z,\end{cases} \quad \mathcal{B}_u^j=\begin{cases}\mathcal{R}_u & \text{for } j=0,y\\ \mathcal{C}_u & \text{for } j=x,z,\end{cases}$$

where we have defined response and correlation operators

$$\mathcal{R}_s\hat{B}=\frac{1}{2}\Delta_b[\xi_s,\hat{B}], \quad \mathcal{C}_s\hat{B}=\frac{1}{2}\Delta_b\{\xi_s,\hat{B}\}. \quad (4.10)$$

Here $\hat{B}$ is an arbitrary function of b_k and $b_k^\dagger$, and $s=g,u$ labels the even and odd fluctuation operators (2.6). Putting together Eqs. (4.8)–(4.10) we find the matrix

$$i\Omega=\begin{pmatrix}0&\mathcal{R}_g&\mathcal{R}_u&0\\\mathcal{R}_g&0&0&i\mathcal{C}_u\\\mathcal{R}_u&0&0&-i\mathcal{C}_g\\0&-i\mathcal{C}_u&i\mathcal{C}_g&0\end{pmatrix}. \quad (4.11)$$

When inserting Ω_{ij} in the perturbation series (4.1), we still have to account for the time evolution of the bath operators. To that purpose we define

$$\Omega_{ij}(t)=e^{i\mathcal{L}_Bt}\Omega_{ij}e^{-i\mathcal{L}_Bt}. \quad (4.12)$$

Inserting Eqs. (4.5) and (4.12) in Eq. (4.1) and taking the thermal average with respect to the bath we find a corresponding series for the time evolution operator $\mathcal{U}(t)$,

$$\mathcal{U}_{ij}(t)=\breve{\mathcal{U}}_{ij}(t)+\int_0^t d\tau\breve{\mathcal{U}}_{ik}(t-\tau)\langle\Omega_{kl}(\tau)\rangle\breve{\mathcal{U}}_{lj}(\tau)+\int_0^t d\tau\int_0^\tau d\tau'\breve{\mathcal{U}}_{ik}(t-\tau)\langle\Omega_{kl}(\tau)\times\breve{\mathcal{U}}_{lm}(\tau-\tau')\Omega_{mn}(\tau')\rangle\breve{\mathcal{U}}_{nj}(\tau')+\cdots, \quad (4.13)$$

where the summation labels $k,l,m,\ldots$ take the values $0,x,y,z$. Since $\breve{\mathcal{U}}$ does not depend on the bath variables, the influence of the heat bath is accounted for by the correlation functions of n operators Ω,

$$\langle\Omega_{ij}(\tau_1)\Omega_{kl}(\tau_2)\cdots\Omega_{pq}(\tau_n)\rangle, \quad (4.14)$$

with $n=1,2,3,\ldots$ and $\tau_1\geqslant\tau_2\geqslant\tau_3\ldots$.

Equation (4.13) gives the exact time evolution of the initial state (3.1). The average $\langle\ldots\rangle$ does not involve the matrices $\breve{\mathcal{U}}$, since these depend on neither spin nor phonon operators. Our treatment relies on an expansion of Eq. (4.14) in terms of irreducible, or connected, correlations of order $1,\ldots,n$ and truncating at finite order.

Since the linear term vanishes, there are no reducible contributions in the second and third orders,

$$\langle\Omega(\tau_1)\rangle_c=\langle\Omega(\tau_1)\rangle=0, \tag{4.15}$$

$$\langle\Omega(\tau_1)\Omega(\tau_2)\rangle_c=\langle\Omega(\tau_1)\Omega(\tau_2)\rangle, \tag{4.16}$$

$$\langle\Omega(\tau_1)\Omega(\tau_2)\Omega(\tau_3)\rangle_c=\langle\Omega(\tau_1)\Omega(\tau_2)\Omega(\tau_3)\rangle. \tag{4.17}$$

As to the fourth order, we have to subtract reducible, or nonconnected, terms of second order according to

$$\begin{aligned}&\langle\Omega(\tau_1)\Omega(\tau_2)\Omega(\tau_3)\Omega(\tau_4)\rangle_c\\&=\langle\Omega(\tau_1)\Omega(\tau_2)\Omega(\tau_3)\Omega(\tau_4)\rangle\\&\quad-\langle\Omega(\tau_1)\Omega(\tau_2)\rangle_c\langle\Omega(\tau_3)\Omega(\tau_4)\rangle_c.\end{aligned} \tag{4.18}$$

After an appropriate change of time integrations and recollecting terms, we find an integral equation for the propagator $\mathcal{U}(t)$,

$$\mathcal{U}_{ij}(t)=\check{\mathcal{U}}_{ij}(t)-\int_0^t d\tau\int_0^\tau d\tau'\check{\mathcal{U}}_{ik}(t-\tau)\Sigma_{kl}(\tau-\tau')\mathcal{U}_{lj}(\tau'), \tag{4.19}$$

where the self-energy is given as an infinite series

$$\Sigma_{ij}(t)=\Sigma_{ij}^{(1)}(t)+\Sigma_{ij}^{(2)}(t)+\Sigma_{ij}^{(3)}(t)+\cdots. \tag{4.20}$$

The terms of nth order involve the irreducible part of n factors $\Omega(\tau_n)$ with $(n-1)$ factors $\check{\mathcal{U}}(\tau_n-\tau_{n-1})$ interposed. Because of Eq. (4.15), the linear term vanishes,

$$\Sigma_{ij}^{(1)}(t-t')=-\langle\Omega_{ij}(t')\rangle_c\delta(t-t')=0; \tag{4.21}$$

that of second-order reads, with $k,l=0,x,y,z$,

$$\Sigma_{ij}^{(2)}(t-t')=\langle\Omega_{ik}(t)\Omega_{lj}(t')\rangle_c\,\check{\mathcal{U}}_{kl}(t-t'). \tag{4.22}$$

In a similar fashion, we find the third-order term

$$\begin{aligned}\Sigma_{ij}^{(3)}(t-t')=&-\int_{t'}^t d\tau\langle\Omega_{ik}(t)\Omega_{lm}(\tau)\Omega_{nj}(t')\rangle_c\\&\times\check{\mathcal{U}}_{kl}(t-\tau)\check{\mathcal{U}}_{mn}(\tau-t').\end{aligned} \tag{4.23}$$

The summation labels k,l,m,n run over $0,x,y,z$. We have used the translational invariance with respect to $t\rightarrow t+t_0$, $t'\rightarrow t'+t_0$. In Eqs. (4.22) and (4.23) all matrix indices are indicated, since we have changed the order of the various factors Ω and $\check{\mathcal{U}}$.

As to terms of higher order, we have to integrate over the $(n-2)$ inner time arguments and to sum over $(2n-2)$ indices. We note the general expression for $n\geqslant 3$,

$$\begin{aligned}\Sigma^{(n)}(\tau_1-\tau_n)=&(-1)^n\int_{\tau_n}^{\tau_1}d\tau_2\cdots\int_{\tau_n}^{\tau_{n-2}}d\tau_{n-1}\langle\Omega(\tau_1)\\&\times\check{\mathcal{U}}(\tau_1-\tau_2)\Omega(\tau_2)\check{\mathcal{U}}(\tau_2-\tau_3)\cdots\Omega(\tau_{n-1})\\&\times\check{\mathcal{U}}(\tau_{n-1}-\tau_n)\Omega(\tau_n)\rangle_c,\end{aligned} \tag{4.24}$$

where Σ, Ω, and $\check{\mathcal{U}}$ are matrices and where the usual matrix product $(\Omega\check{\mathcal{U}}\cdots\Omega)_{ij}=\Omega_{ik}\check{\mathcal{U}}_{kl}\cdots\Omega_{mj}$ is understood. The bath correlation $\langle\Omega(\tau_1)\cdots\Omega(\tau_n)\rangle_c$ contains the irreducible, or connected, part of Eq. (4.14) only.

V. SECOND-ORDER APPROXIMATION

The series (4.20) constitutes an expansion in terms of the connected bath correlations (4.15)–(4.18). We are going to retain the lowest-order term Eq. (4.16) only, and to evaluate the second-order term of the self-energy, as given in Eq. (4.22). We start with a few observations which restrict the number of finite matrix elements of $\Sigma^{(2)}$.

(i) There is no bath operator to the left of the factor $\Omega_{lj}(t)$ in Eq. (4.22). Since the response operators $\mathcal{R}_s$ vanish when there is unity to its left, we have

$$\langle\mathcal{R}_{s'}(t)\mathcal{C}_s(t')\rangle=0=\langle\mathcal{R}_{s'}(t)\mathcal{R}_s(t')\rangle$$

for any s,s'.

(ii) The fluctuation operator ξ_u is odd in terms of bath operators $b_k,b_k^\dagger$, and ξ_g is even. Thus two-times bath correlations are finite only for $s=s'$,

$$\langle\xi_{s'}(t)\xi_s(t')\rangle=\langle\xi_s(t)\xi_s(t')\rangle\delta_{ss'}.$$

The time evolution of the fluctuation operators reads as $\xi_s(t)=e^{iH_Bt/\hbar}\xi_s e^{-iH_Bt/\hbar}$, according to Eq. (4.12).

(iii) The Hamiltonian (2.7) is invariant under the ''parity'' transformation $\sigma_z\rightarrow-\sigma_z$, $\sigma_y\rightarrow-\sigma_y$, $b_k\rightarrow-b_k$, implying a useful symmetry property. Both the propagator $\mathcal{U}$ and the self-energy matrix Σ are block diagonal with respect to the pairs of labels $0,x$ and y,z.

Taking into account these selection rules and inserting the matrices for Ω and $\check{\mathcal{U}}$ in Eq. (4.22), we find that the matrix Σ is block diagonal with six finite entries. The terms involving the labels y,z read as

$$\Sigma_{zz}^{(2)}(t)=\Gamma_u(t)\check{\mathcal{U}}_{xx}(t)+\Gamma_g(t)\check{\mathcal{U}}_{yy}(t), \tag{5.1}$$

$$\Sigma_{yz}^{(2)}(t)=-\Gamma_g(t)\check{\mathcal{U}}_{zy}(t)=-\Sigma_{zy}^{(2)}(t), \tag{5.2}$$

$$\Sigma_{yy}^{(2)}(t)=\Gamma_g(t)\check{\mathcal{U}}_{zz}(t), \tag{5.3}$$

where the symmetrized bath correlations

$$\Gamma_s(t-t')=\langle\mathcal{C}_s(t)\mathcal{C}_s(t')\rangle=\frac{1}{2}\Delta_b^2\langle\{\xi_s(t),\xi_s(t')\}\rangle \tag{5.4}$$

are readily obtained by inserting Eq. (4.10) in Eq. (4.16).

The obvious relation $\langle\sigma_0(t)\rangle=1$ requires $\Sigma_{00}=0=\Sigma_{0x}$. As a consequence, the upper submatrix contains only two finite entries,

$$\Sigma_{xx}^{(2)}(t)=\Gamma_u(t)\check{\mathcal{U}}_{zz}(t), \tag{5.5}$$

$$\Sigma_{x0}^{(2)}(t)=i\chi_{\rm u}(t)\check{\mathcal{U}}_{zy}(t). \tag{5.6}$$

The matrix Σ is not symmetric; the off-diagonal entry involves the bath response function

$$\chi_{\rm u}(t-t')=\langle\mathcal{C}_{\rm u}(t)\mathcal{R}_{\rm u}(t')\rangle=\frac{1}{2}\Delta_{\rm b}^2\langle[\xi_{\rm u}(t),\xi_{\rm u}(t')]\rangle. \tag{5.7}$$

VI. THE PROPAGATOR MATRIX

To proceed further we take the Laplace transform of Eq. (4.19), use the convolution theorem, and solve for the propagator matrix $\mathcal{U}$,

$$\mathcal{U}(z)=-[-\check{\mathcal{U}}^{-1}(z)+\Sigma(z)]^{-1}. \tag{6.1}$$

As a consequence of the conservation law mentioned above, the matrix $\mathcal{U}_{\rm ij}$ is block diagonal and splits in two 2×2 matrices,

$$\mathcal{U}(z)=\begin{pmatrix}\mathcal{V}(z) & 0\\ 0 & \mathcal{W}(z)\end{pmatrix}, \tag{6.2}$$

where $\mathcal{V}$ involves $0,x$, and $\mathcal{W}$ the indices y,z only.

As to the self-energy matrix, when inserting the uncoupled propagator (4.5) and using the convolution theorem, we find in a straightforward fashion

$$\Sigma_{zz}(z)=\Gamma_{\rm u}(z)+\frac{1}{2}[\Gamma_{\rm g}(z+\tilde{\Delta}_0)+\Gamma_{\rm g}(z-\tilde{\Delta}_0)], \tag{6.3}$$

$$\Sigma_{yz}(z)=\frac{i}{2}[\Gamma_{\rm g}(z+\tilde{\Delta}_0)-\Gamma_{\rm g}(z-\tilde{\Delta}_0)], \tag{6.4}$$

$$\Sigma_{yy}(z)=\frac{1}{2}[\Gamma_{\rm g}(z+\tilde{\Delta}_0)+\Gamma_{\rm g}(z-\tilde{\Delta}_0)], \tag{6.5}$$

$$\Sigma_{xx}(z)=\frac{1}{2}[\Gamma_{\rm u}(z+\tilde{\Delta}_0)+\Gamma_{\rm u}(z-\tilde{\Delta}_0)], \tag{6.6}$$

$$\Sigma_{x0}(z)=\frac{1}{2}[\chi_{\rm u}(z+\tilde{\Delta}_0)-\chi_{\rm u}(z-\tilde{\Delta}_0)]. \tag{6.7}$$

Here and in the remainder of this paper, we drop the label for the second-order approximation. The spectra $\Gamma''_{\rm u}$ and $\Gamma''_{\rm g}$ are symmetric functions of frequency, whereas $\chi''_{\rm u}$ is antisymmetric.

The unperturbed propagator of the upper block in Eq. (6.2) is given by $\check{\mathcal{V}}(z)=-1/z$. Inserting the two entries of the self-energy matrix, we have

$$\mathcal{V}(z)=-\begin{pmatrix} z & 0\\ \Sigma_{x0}(z) & z+\Sigma_{xx}(z)\end{pmatrix}^{-1}. \tag{6.8}$$

Before writing down the lower block $\mathcal{W}$, we note that at small frequency, the symmetric spectra $\Gamma''_{\rm u}$ and $\Gamma''_{\rm g}$ are well approximated by a constant. For this reason, the off-diagonal matrix elements Σ_{yz} and Σ_{zy} are, at small z, much smaller than the diagonal ones, and may be dropped. Then the self-energy of $\mathcal{W}$ is diagonal,

$$\mathcal{W}(z)=-\begin{pmatrix} z+\Sigma_{yy}(z) & -i\tilde{\Delta}_0\\ i\tilde{\Delta}_0 & z+\Sigma_{zz}(z)\end{pmatrix}^{-1}. \tag{6.9}$$

It seems worthwhile noting the Laplace transform of the quantity $P(t)$, defined in Eq. (3.7), which is given by the lower diagonal element $P(z)=\mathcal{W}_{zz}(z)$. Inversion of the 2×2 matrix results in

$$P(z)=-\left(z+\Sigma_{zz}(z)-\frac{\tilde{\Delta}_0^2}{z+\Sigma_{yy}(z)}\right)^{-1}. \tag{6.10}$$

A. Pole approximation

In order to proceed further we simplify the frequency dependence of the propagator $\mathcal{U}(z)$. To that purpose we note that the self-energy spectra $\Sigma''_{ij}(\omega)$ are constant at small frequencies, whereas the reactive parts Σ' are roughly linear in z. Accordingly we evaluate the dissipative parts $\Sigma''(z)$ at the resonances and keep the frequency dependence of the reactive parts,

$$\Sigma(z)=\Sigma'(z)+i\Sigma''(z_0). \tag{6.11}$$

The values of z_0 are given by the zeroes of $\det(\check{\mathcal{U}}(z)^{-1})=z^2(z^2-\tilde{\Delta}_0^2)$. From Eqs. (6.8) and (6.9) it is clear that $\mathcal{V}(z)$ involves the double pole at $z=0$, whereas $\mathcal{W}(z)$ shows two poles at $z=\pm\tilde{\Delta}_0$.

As shown in the Appendix, the reactive part is small and, therefore, of little consequence. Here we evaluate the dissipative part of the self-energy matrix. From Eqs. (6.3)–(6.7) we obtain

$$\Sigma''_{zz}(\tilde{\Delta}_0)=\gamma_{\rm u}+\frac{1}{2}(1+c)\gamma_{\rm g}, \tag{6.12}$$

$$\Sigma''_{yy}(\tilde{\Delta}_0)=\frac{1}{2}(1+c)\gamma_{\rm g}, \tag{6.13}$$

$$\Sigma''_{xx}(0)=\gamma_{\rm u},\quad \Sigma''_{x0}(0)=\gamma_0, \tag{6.14}$$

where we have defined the rates

$$\gamma_{\rm u}=\Gamma''_{\rm u}(\tilde{\Delta}_0),$$

$$\gamma_{\rm g}=\Gamma''_{\rm g}(0),$$

$$\gamma_0=\chi''_{\rm u}(\tilde{\Delta}_0), \tag{6.15}$$

and the additional temperature factor

$$c=\beta\hbar\tilde{\Delta}_0\sinh(\beta\hbar\tilde{\Delta}_0)^{-1}. \tag{6.16}$$

In Eq. (6.9) we have neglected the off-diagonal parts Σ''_{yz} and Σ''_{zy}. Considering the relation

$$\Sigma''_{yz}(\pm\tilde{\Delta}_0)=\pm\frac{i}{2}(1-c)\gamma_{\rm g}=-\Sigma''_{zy}(\pm\tilde{\Delta}_0), \tag{6.17}$$

we find that the factor $(1-c)$ is significant at low temperatures $\hbar\tilde{\Delta}_0\ll k_{\rm B}T$ only. In Eq. (7.18) we will see that the rate $\gamma_{\rm g}$ is relevant at high temperatures $T\gg T_0$ only. Since the weak-coupling condition (1.9) assures the inequality $\hbar\widetilde{\Delta}_0 \ll k_B T_0$, we conclude that the off-diagonal entries (6.17) are negligible indeed.

VII. DAMPING RATES

The damping rates defined in Eq. (6.15) may be written in terms of the spectra of the two bath correlation functions

$$\langle B_\pm(t)B_\mp(t')\rangle = B^2 e^{\varphi(t-t')},$$

$$\langle B_\pm(t)B_\pm(t')\rangle = B^2 e^{-\varphi(t-t')}, \tag{7.1}$$

where $B_\pm$ are defined in Eq. (2.4). The phase is given by the coupled phonon propagator[34]

$$\varphi(t) = \sum_k u_k^2[n_k e^{i\omega_k t} + (1+n_k)e^{-i\omega_k t}], \tag{7.2}$$

with the Bose occupation numbers

$$n_k \equiv n(\omega_k) = [e^{\beta\hbar\omega_k} - 1]^{-1}. \tag{7.3}$$

In the long-time limit, both functions (7.1) tend towards the constant B^2. According to the definition of the fluctuation operators (2.6), this constant has been removed in the self-energy. With Eq. (2.9) and the definition of ξ_α, we obtain

$$\Gamma_g(t) = \frac{1}{2}\widetilde{\Delta}_0^2[\cosh(\varphi(t)) + \cosh(\varphi(-t)) - 2],$$

$$\Gamma_u(t) = \frac{1}{2}\widetilde{\Delta}_0^2[\sinh(\varphi(t)) + \sinh(\varphi(-t))],$$

$$\chi_u(t) = \frac{1}{2}\widetilde{\Delta}_0^2[\sinh(\varphi(t)) - \sinh(\varphi(-t))]. \tag{7.4}$$

Since Γ_g contains only even powers of φ and Γ_u only odd ones, we are led to consider the function $\Gamma(t) = \Gamma_g(t) + \Gamma_u(t)$,

$$\Gamma(t) = \frac{1}{2}\widetilde{\Delta}_0^2[e^{\varphi(t)} + e^{\varphi(-t)} - 2], \tag{7.5}$$

and to separate even and odd terms later.

In view of Eq. (6.5) we need to calculate the spectrum $\Gamma''(\omega)$ which is given by the Fourier transform

$$\Gamma''(\omega) = \frac{1}{4}\widetilde{\Delta}_0^2 \int_{-\infty}^{\infty} dt\; e^{i\omega t}[e^{\varphi(t)} + e^{-\varphi(t)} - 2]. \tag{7.6}$$

It is worth noting that this spectrum is closely related to the damping function obtained from the noninteracting blip approximation.[11,30] In the present approach, the lowest diagonal element of the self-energy matrix, Σ_{zz}, is given by $\Gamma(z)$, whereas the remaining entries involve its even or odd parts only.

As to the the response function χ_u, its spectrum is related to odd part Γ''_u by a fluctuation-dissipation theorem,

$$\chi''_u(\omega) = \tanh\left(\frac{1}{2}\beta\hbar\omega\right)\Gamma''_u(\omega). \tag{7.7}$$

The coupled phonon propagator $\varphi(t)$ is not invariant under time reversal but rather satisfies the relation

$$\varphi(-t) = \varphi(t - i\beta\hbar). \tag{7.8}$$

It turns out convenient to define the function

$$\overline{\varphi(t)} = \varphi\left(t - \frac{1}{2}i\beta\hbar\right), \tag{7.9}$$

which is symmetric under time reversal and reads explicitly as

$$\overline{\varphi(t)} = \sum_k u_k^2 \frac{\cos(\omega_k t)}{\sinh(\beta\hbar\omega_k/2)}. \tag{7.10}$$

Following Grabert,[17] we are going to rewrite Eq. (7.6) in terms of $\overline{\varphi(t)}$.

Since $\Gamma(t)$ is an analytic function of time, we may shift the time integration from the real axis into the complex t plane. As the phonon density of states vanishes in the limit of zero frequency, the phase varies rapidly with time for $t\to\infty$. Thus the Fourier integral (7.6) is determined by thermal frequencies; the integration contour for large t is immaterial, and, with $t \to t - \frac{1}{2}i\beta\hbar$, we have instead of the spectrum (7.6)

$$\Gamma''(\omega) = \frac{1}{2}\widetilde{\Delta}_0{}^2\cosh\left(\frac{1}{2}\beta\hbar\omega\right)\int_{-\infty}^{\infty} dt\; e^{i\omega t}[e^{\overline{\varphi(t)}} - 1]. \tag{7.11}$$

When replacing the sum over k in $\overline{\varphi(t)}$ by an integral and pushing the cutoff frequency ω_D to infinity, one obtains the phase[17]

$$\overline{\varphi(t)} = 2\alpha\left(\frac{\pi}{\hbar\beta}\right)^2 \frac{1}{\cosh(\pi t/\beta\hbar)^2}. \tag{7.12}$$

In this limit, $\varphi(t)$ diverges at $t=0$, as does $\overline{\varphi(t = \frac{1}{2}i\hbar\beta)}$, leading to an essential singularity in $\Gamma(t)$ that is, however, of no consequence to our purpose.

Since the Fourier transformation of $[e^{\overline{\varphi(t)}} - 1]$ cannot be performed in closed form, we expand the exponential in a power series, and transform each term separately,

$$\Gamma''(\omega) = \widetilde{\Delta}_0^2\cosh\left(\frac{1}{2}\beta\hbar\omega\right)\frac{\beta\hbar}{\pi}\sum_{n=1}^{\infty}\frac{\varphi_0^n}{n!}A_n(\beta\hbar\omega). \tag{7.13}$$

For notational convenience we have split off a prefactor in powers of

$$\varphi_0 = 2\alpha(\pi/\hbar\beta)^2 = \frac{T^2}{T_0^2}; \tag{7.14}$$

the remaining Fourier integral arising from the exponential series is given by

$$A_n(\beta\hbar\omega) = \frac{\pi}{\hbar\beta}\int_{-\infty}^{\infty} dt e^{i\omega t}\frac{1}{\cosh(\pi t/\beta\hbar)^{2n}}. \tag{7.15}$$

In the following we consider a few particular cases with respect to frequency and temperature.

Low-temperature expansion

The parameter φ_0 plays the role of a dimensionless coupling constant. At low temperatures, $T \ll T_0$, it is much smaller than unity, and the series (7.13) may be restricted to its first term. When inserting the value of the Fourier integral $A_1(x) = \frac{1}{2} x \sinh(\frac{1}{2}x)^{-1}$, we obtain

$$\Gamma''(\omega) = \frac{1}{2}\beta\hbar\omega\coth\left(\frac{1}{2}\widetilde{\Delta}_0^2\beta\hbar\omega\right)\frac{\beta\hbar}{\pi}\varphi_0 \quad (\varphi_0 \ll 1). \tag{7.16}$$

This result has been derived previously in Ref. 11, where the series (7.13) has been truncated at lowest order. In physical terms this means that multiphonon processes have been discarded. In view of the expansion parameter $\varphi_0 = T^2/T_0^2$ it is clear that this approximation breaks down as temperature approaches T_0.

By separating odd and even powers of φ_0, setting $\omega = \widetilde{\Delta}_0$ and using Eq. (7.14), we find

$$\gamma_{\mathrm{u}} = \pi\alpha\widetilde{\Delta}_0^3\coth\left(\frac{1}{2}\beta\hbar\widetilde{\Delta}_0\right)[1 + O(\varphi_0^2)], \tag{7.17}$$

whose first term is the well-known one-phonon rate. Its counterpart γ_{g} has to be evaluated at $\omega = 0$,

$$\gamma_{\mathrm{g}} = 2\pi\alpha\widetilde{\Delta}_0^2(k_{\mathrm{B}}T/\hbar)\frac{\varphi_0}{3}[1 + O(\varphi_0^2)], \tag{7.18}$$

and starts with a term proportional to $\alpha^2 T^3$.

The series at zero frequency

In view of Eq. (6.15) we need to evaluate $\Gamma''(\omega)$ at $\omega = \widetilde{\Delta}_0$ and $\omega = 0$. For temperatures well above $\hbar\widetilde{\Delta}_0/k_{\mathrm{B}}$, the argument of the function (7.15) is much smaller than unity. Since $A_n(x)$ depends very weakly on x for $|x| \ll 1$, we may put $\omega = 0$ in Eq. (7.13),

$$\Gamma''(0) = \widetilde{\Delta}_0^2\frac{\beta\hbar}{\pi}\varphi_0 F(\varphi_0), \tag{7.19}$$

where the infinite series has been absorbed in the factor

$$F(\varphi_0) = \sum_{n=1}^{\infty}\frac{\varphi_0^{n-1}}{n!}A_n, \tag{7.20}$$

with the coefficients $A_n \equiv A_n(0)$. Evaluating the integral (7.15) at $\omega = 0$, one finds[35]

$$A_n = 4^{n-1}\frac{(n-1)!^2}{(2n-1)!}. \tag{7.21}$$

One easily verifies $F(\varphi_0) \to 1$ for $\varphi_0 \to 0$. With the definition of φ_0, the first few terms of the series read

$$\Gamma''(0) = 2\pi\alpha\widetilde{\Delta}_0^2(k_{\mathrm{B}}T/\hbar) \times\left[1 + \frac{1}{3}\varphi_0 + \frac{4}{45}\varphi_0^2 + \frac{2}{105}\varphi_0^3 + \cdots\right]. \tag{7.22}$$

The linear correction $[1 + \frac{1}{3}\varphi_0]$ has been derived in Ref. 30. When expanding the coth function in powers of its inverse argument, we find that Eqs. (7.16) and (7.22) agree with respect to the leading contribution.

Saddle-point integration

At high temperatures, $\varphi_0 \gg 1$, the spectrum at zero frequency, $\Gamma''(0)$, may be evaluated by saddle-point integration. When truncating the expansion of $\overline{\varphi(t)} = \varphi_0 - \frac{1}{2}\varphi_2 t^2 + \cdots$ after the quadratic term and evaluating the Gaussian integral, we find Holstein's diffusion rate[32]

$$\Gamma_{\mathrm{SPI}} = \widetilde{\Delta}_0^2\frac{\beta\hbar}{\pi}\frac{1}{\sqrt{4\varphi_0/\pi}}e^{\varphi_0} \quad (\varphi_0 \gg 1). \tag{7.23}$$

Here we have already used the relation $\varphi_2 = \varphi_0^2/\alpha$ [Ref. 30 and Eq. (1.6)]. For dissipative two-state dynamics, Eq. (7.23) has been first derived by Pirc and Gosar.[24]

Eq. (7.23) is valid for $T_0 \ll T \ll \Theta$. In the opposite limit $T > \Theta$, the rate has been evaluated by Niu.[25]

Interpolation formula

For practical purposes, the series expansion (7.20) is not very convenient, since it converges slowly. From Stirling's formula, $n! \approx (n/e)^n\sqrt{2\pi n}$ for large n, we find that the main contributions to the series stem from terms with n of the order of φ_0. Thus for temperatures about ten times larger than T_0, several hundred terms have to be retained in order to assure convergence of the series (7.20).

For this reason we propose a simple interpolation formula for the factor $F(\varphi_0)$,

$$F(\varphi_0) = \frac{e^{\varphi_0} - 1}{\varphi_0}(1 + 4\varphi_0/\pi)^{-1/2}, \tag{7.24}$$

that correctly describes the limits $F(\varphi_0) \to 1$ for $\varphi_0 \to 0$ and $\varphi_0 F(\varphi_0) \to \sqrt{\pi/4\varphi_0}e^{\varphi_0}$ for $\varphi_0 \gg 1$. The error in the intermediate range about $\varphi_0 \approx 1$ does not exceed 15%.

The frequency-dependent prefactor of the linear term is relevant at very low temperatures $k_{\mathrm{B}}T \leq \hbar\widetilde{\Delta}_0$. At higher T it tends towards unity, $\frac{1}{2}x\coth(\frac{1}{2}x) \to 1$ for $x \ll 1$. For this reason we may write

$$\Gamma''(\omega) = \pi\widetilde{\Delta}_0{}^2\alpha\omega\coth\left(\frac{1}{2}\widetilde{\Delta}_0\beta\hbar\omega\right)F(\varphi_0), \tag{7.25}$$

which holds true for frequencies $\hbar|\omega| \leq \max(T, T_0)$.

For further convenience, we give approximate expressions for the rates γ_{u} and γ_{g}, resulting from the interpolation formula (7.24),

$$\gamma_{\mathrm{u}} = \pi\alpha\widetilde{\Delta}_0^3\coth\left(\frac{1}{2}\beta\hbar\widetilde{\Delta}_0\right)\frac{\sinh(\varphi_0)}{\varphi_0\sqrt{1 + 4\varphi_0/\pi}}, \tag{7.26}$$

$$\gamma_{\mathrm{g}} = 2\pi\alpha\widetilde{\Delta}_0^2(k_{\mathrm{B}}T/\hbar)\frac{\cosh(\varphi_0) - 1}{\varphi_0\sqrt{1 + 4\varphi_0/\pi}}. \tag{7.27}$$

At low temperatures, $T \ll T_0$, these rates tend towards the correct expressions (7.17) and (7.18), whereas in the opposite limit $T \gg T_0$ they satisfy $\gamma_{\mathrm{u}} = \frac{1}{2}\Gamma_{\mathrm{SPI}} = \gamma_{\mathrm{g}}$.

In order to complete the temperature dependence of the damping spectra, we note the value of B as defined in Eq. (2.5). Inserting the spectral function (1.4) in

$$B=\exp\left[-\sum_k u_k^2(1+2n_k)\right], \tag{7.28}$$

we obtain[34]

$$B=\exp\left[-\frac{1}{2}\alpha\omega_{\rm D}^2-\frac{1}{6}\varphi_0\right]. \tag{7.29}$$

By separating the argument of the exponential we rewrite the renormalized tunnel frequency (2.9) as

$$\tilde{\Delta}_0=\Delta_0\exp\left[-\frac{1}{6}\varphi_0\right]. \tag{7.30}$$

Here the constant Δ_0 contains the zero-temperature Debye-Waller factor, $\Delta_0=\Delta_{\rm b}\exp(-\frac{1}{2}\alpha\omega_{\rm D}^2)$. The thermal motion reduces the tunnel frequency further to the value $\tilde{\Delta}_0$.

Thereby, the above rates acquire an additional factor $e^{-\varphi_0/3}$ that reduces, e.g., Eq. (7.23) to

$$\Gamma_{\rm SPI}=\Delta_0^2\frac{\beta\hbar}{\pi}\frac{1}{\sqrt{4\varphi_0/\pi}}\exp\left(\frac{2}{3}\varphi_0\right)\quad(\varphi_0\gg 1). \tag{7.31}$$

VIII. TIME EVOLUTION

Equations (6.8) and (6.9) provide the damped spin propagator in terms of the self-energy. Inserting the rates (6.15) and matrix inversion gives the upper 2×2 block,

$$\mathcal{V}(z)=-\frac{1}{z(z+i\gamma_{\rm u})}\begin{pmatrix} z+i\gamma_{\rm u} & 0\\ -i\gamma_0 & z\end{pmatrix}, \tag{8.1}$$

which shows two poles in the complex plane. Upon inverse Laplace transformation, the relaxation pole at $z=-i\gamma_{\rm u}$ gives rise to an exponentially decaying contribution, whereas the undamped pole at $z=0$ leads to a constant,

$$\mathcal{V}(t)=\begin{pmatrix} 1 & 0\\ -\gamma_0/\gamma_{\rm u} & 0\end{pmatrix}+e^{-\gamma_{\rm u}t}\begin{pmatrix} 0 & 0\\ \gamma_0/\gamma_{\rm u} & 1\end{pmatrix}. \tag{8.2}$$

The latter feature assures conservation of the trace of the density operator, and the initial condition $\mathcal{V}(t=0)=1$.

With the ratio $\gamma_0/\gamma_{\rm u}=\tanh(\beta\hbar\tilde{\Delta}_0/2)$ we obtain the function (3.8),

$$R(t)=-\tanh(\beta\hbar\tilde{\Delta}_0/2)[1-e^{-\gamma_{\rm u}t}]. \tag{8.3}$$

The relaxation rate contains only odd powers of the coupling constant α; in the long-time limit, $R(t)$ tends towards the thermal expectation value $\langle\sigma_x\rangle_{\rm eq}$.

Now we turn to the lower submatrix in Eq. (6.2). As discussed below Eq. (6.17), we set $c=1$. When inserting the rates in Eq. (6.10), the diagonal element $P(z)=\mathcal{W}_{zz}(z)$ reads

$$P(z)=-\frac{z+i\gamma_{\rm g}}{(z+i\gamma_{\rm g})(z+i\gamma_{\rm u}+i\gamma_{\rm g})-\tilde{\Delta}_0^2}. \tag{8.4}$$

Calculating the roots of the quadratic form in the denominator and taking the inverse Laplace transformation, we obtain

$$P(t)=\sum_{\pm}\frac{\Gamma_\pm-\gamma_{\rm g}}{\Gamma_\pm-\Gamma_\mp}\exp(-\Gamma_\pm t), \tag{8.5}$$

whose complex frequencies are given by

$$\Gamma_\pm=\gamma_{\rm g}+\frac{1}{2}\gamma_{\rm u}\pm\sqrt{\frac{1}{4}\gamma_{\rm u}^2-\tilde{\Delta}_0^2}. \tag{8.6}$$

These three formulas constitute, together with the rates (7.26) and (7.27), the main results of this paper.

The argument of the square root changes sign as a function of temperature. At low temperature, the relation $\tilde{\Delta}_0>\frac{1}{2}\gamma_{\rm u}$ gives rise to two complex poles in Eq. (8.4) and to underdamped oscillations of $P(t)$. With increasing temperature, the system passes through the aperiodic case $\tilde{\Delta}_0=\frac{1}{2}\gamma_{\rm u}$ and finally reaches the range of incoherent motion, where both poles of Eq. (8.4) are purely imaginary, i.e., where $\Gamma_\pm$ are real. Because of the different dynamical behavior, we deal separately with the two cases.

Damped oscillations: $\frac{1}{2}\gamma_{\rm u}<\tilde{\Delta}_0$

In view of the temperature dependence of both rate and tunnel energy, this range may be labeled as a weak-coupling or low-temperature case.

The roots of the denominator of Eq. (8.4) exhibit both real and imaginary parts, $\Gamma_\pm=\Gamma_t\pm i\omega_{\rm t}$, where the effective tunnel frequency $\omega_{\rm t}$ and the damping rate are defined as

$$\omega_{\rm t}=\sqrt{\tilde{\Delta}_0^2-\frac{1}{4}\gamma_{\rm u}^2},\quad \Gamma_t=\frac{1}{2}\gamma_{\rm u}+\gamma_{\rm g}. \tag{8.7}$$

Accordingly we find damped oscillations,

$$P(t)=\frac{\cos(\omega_{\rm t}t+\delta)}{\cos(\delta)}\exp(-\Gamma_t t), \tag{8.8}$$

with a phase shift defined by $\tan\delta=(\gamma_{\rm u}/2\omega_{\rm t})$. At zero temperature, the tunnel frequency $\omega_{\rm t}$ is almost identical to Δ_0. In the range $T\approx T_0$ it decreases exponentially according to Eq. (7.30), and finally vanishes at the crossover temperature T^*.

At very low temperatures the rate is dominated by the one-phonon contribution to $\gamma_{\rm u}$,

$$\Gamma_t=\frac{\pi}{2}\alpha\tilde{\Delta}_0^3\coth(\hbar\tilde{\Delta}_0/2k_{\rm B}T)+O(\alpha^2). \tag{8.9}$$

With rising temperature, multiphonon terms become more important; in order to permit a comparison with the result of Sec. IV, we give the power series resulting from Eqs. (7.17) and (7.18),

$$\Gamma_t=\pi\alpha\tilde{\Delta}_0^2(k_{\rm B}T/\hbar)\left[1+\frac{2}{3}\varphi_0+\frac{4}{45}\varphi_0^2+\frac{4}{105}\varphi_0^3+\cdots\right]. \tag{8.10}$$

Note that the rate of phase memory loss, Γ_t, involves both $\gamma_{\rm u}$ and $\gamma_{\rm g}$, whereas the phase shift depends on the odd-order terms $\gamma_{\rm u}$ only.

***Incoherent tunneling:* $\frac{1}{2}\gamma_u > \tilde{\Delta}_0$**

For the aperiodic case $\frac{1}{2}\gamma_u = \tilde{\Delta}_0$, the two poles $-i\Gamma_\pm$ merge on the imaginary axis, $\Gamma_+ = \Gamma_-$. When increasing the temperature further, the rate $\frac{1}{2}\gamma_u$ exceeds $\tilde{\Delta}_0$. Then both poles of Eq. (8.4) are purely imaginary; accordingly the motion is best described as incoherent tunneling between the two states $\sigma_z = \pm 1$ with two different relaxation rates $\Gamma_\pm$.

Even for very high temperatures, the smaller rate Γ_- differs from Γ_+ merely by a factor of 2, $\Gamma_- = \gamma_g \approx \frac{1}{2}(\gamma_g + \gamma_u) = \frac{1}{2}\Gamma_+$. Due to the exponential increase of the rates, however, the amplitude of the term involving Γ_- in Eq. (8.5) vanishes rapidly. As a consequence, $P(t)$ is well described by the simpler function

$$P(t) = e^{-\Gamma t} \quad \text{with} \quad \Gamma = \Re\Gamma_+ . \tag{8.11}$$

Well above the crossover temperature, the rate Γ is identical to the NIBA result

$$\Gamma = 2\pi\alpha(k_B T \tilde{\Delta}_0^2/\hbar) F(\varphi_0), \tag{8.12}$$

where the last factor can be taken either as the exact series (7.20) or as the approximate expression (7.24).

Regarding the temperature dependence, there is a competition between the increase of the last factor and the decrease of the renormalized tunnel energy $\tilde{\Delta}_0$. Yet a glance at Eq. (7.30) shows that the sum of the arguments in the exponentials is positive, resulting in an exponential increase of the rate. With the constant Δ_0 it reads as

$$\Gamma = \frac{\hbar\Delta_0^2}{2\pi^{1/2}k_B}\frac{T_0}{T^2}\exp\left(\frac{2}{3}\frac{T^2}{T_0^2}\right) \quad \text{for} \quad T \gg T_0 . \tag{8.13}$$

The crossover temperature

The crossover to incoherent motion described above occurs at $2\tilde{\Delta}_0 = \gamma_u$, requiring a value for φ_0 larger than unity. Accordingly we have $\gamma_u = \frac{1}{2}\Gamma$ and find with the tunnel energy (7.30) and

$$T_0 = (2\alpha)^{-1/2}(\hbar/\pi k_B)^2$$

an implicit equation for the crossover temperature T^*,

$$T^* = T_0\sqrt{\frac{6}{5}\ln(4\sqrt{\pi}k_B T^{*2}/T_0\hbar\Delta_0)}. \tag{8.14}$$

For temperatures well below T_0, the tunnel energy is given by the constant Δ_0, whereas it decreases exponentially above. For most physical systems one finds $\Delta_0 \ll k_B T_0$. Therefore the crossover temperature T^*, which is defined by $\omega_t = 0$ in Eq. (8.7), is significantly larger than T_0.

IX. DISCUSSION

The strong-coupling approach of this paper relies on a perturbative treatment with respect to the bath fluctuation operators ξ_α. Already in lowest-order approximation, the resulting tunnel frequency and damping rates comprise terms of any order in the coupling parameter α or, more precisely, in the dimensionless quantity φ_0.

The restricted range of validity of previous work[11] comes from the fact that the quantity $\alpha\Delta_0^2$ has been treated as expansion parameter, thereby neglecting terms of higher orders in the temperature-dependent parameter φ_0. Implicitly, in Ref. 11 our function $F(\varphi_0)$, as defined in Eq. (7.20), has been replaced by the value $F(\varphi_0) = 1$ for $\varphi_0 \ll 1$.

Formally, the expansion of the self-energy (4.20) may be written as a series in powers of the tunnel frequency Δ_b. Yet it is not *a priori* clear whether this series involves a small parameter which would assure convergence. We have calculated the lowest-order corrections arising from $\Sigma^{(3)}$ and $\Sigma^{(4)}$ and we have found they are negligible. Although this is not a rigorous proof for the validity of our retaining $\Sigma^{(2)}$ only, we may reasonably assume that the second-order term provides a controlled approximation.

Besides the dissipative part of the self-energy matrix, we have, in the Appendix, considered its real part. As a main result, we find that the derivative of $\Gamma'(\omega)$ is much smaller than unity [cf. Eq. (A20)], resulting in $\mathcal{Z}$ factors close to unity.

Comparison with previous work

Weak-coupling perturbation theory. In a recent investigation based on the Hamiltonian (1.2), the self-energy of the spin propagator was expanded in powers of λ_k.[31] By calculating the contribution up to fourth order, the two-phonon correction for the transverse damping rate was obtained,

$$\Gamma_t = \pi\alpha\mathcal{Z}\Delta_b^2(k_B T/\hbar)\left[1 + \frac{2}{3}\varphi_0 + O(\varphi_0^2)\right], \tag{9.1}$$

where $\mathcal{Z} = [1 + \alpha\omega_D^2 + \frac{2}{3}\pi^2\alpha/(\hbar\beta)^2 + \cdots]^{-1}$ is identical to the lowest-order terms of B^2 as given in Eq. (7.29). Comparison with the low-temperature result Eq. (8.10) reveals a perfect agreement for both the explicit temperature dependence and the renormalization of the tunnel frequency.

Regarding the longitudinal rate γ_u in Eq. (8.3), it contains terms of odd order only. It seems worth noting that perturbation theory confirms this result as well.[36] Therefore we conclude that the present approach provides the exact results for the first two terms of the power series of the rates. This is not an irrelevant statement, since the results obtained previously by various authors differ significantly with respect to the second-order term.

Imaginary-time approach. The series (7.20) can be related to Grabert's result for the incoherent rate arising from a composite spectral density that accounts for coupling to both phonons and conduction electrons. Eq. (18) of Ref. 17 depends on φ_0 through the hypergeometric series ${}_1F_1(K, K+\frac{1}{2}; \varphi_0)$, divided by the Kondo parameter K. Considering the phonon correction factor, and taking the limit of zero K, we recover the series (7.20),

$$\lim_{K\to 0}\frac{1}{K}\left[{}_1F_1\left(K, K+\frac{1}{2}; \varphi_0\right) - 1g\right] = \varphi_0 F(\varphi_0),$$

where the "1" in brackets removes, according to Holstein, the "diagonal transition."[32]

Comparison with the blip expansion

Much work on the spin-boson model relies on the *noninteracting blip approximation* (NIBA).[11,37] In a field-theoretical language, a blip consists of an instanton pair, which describe the motion along the classical trajectory in an inverted double-well potential. In terms of the Hamiltonian (2.3) a blip corresponds to the scattering from one well to the other and back to the original well, while the dressed particle drags its phonon cloud.

A blip expansion for the two-state dynamics is most easily derived by formal integration of the equation of motion of the spin operators σ_z and $\sigma_\pm$.[15,30] Imposing the initial condition $P(t=0)=1$ and taking the thermal average, one obtains an infinite series for Eq. (3.7),

$$P(t)=1-\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle K(\tau_1,\tau_2)\rangle+\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2 \times\int_0^{\tau_2}d\tau_3\int_0^{\tau_3}d\tau_4\langle K(\tau_1,\tau_2)K(\tau_3,\tau_4)\rangle+\cdots, \tag{9.2}$$

where a blip is described by the kernel[30]

$$K(t,t')=\frac{1}{2}\Delta_b^2[B_+(t)B_-(t')+B_-(t)B_+(t')]. \tag{9.3}$$

(The operators $B_\pm$ shift the phonon coordinates between the potential minima corresponding to $\sigma_z=\pm1$.)

The exact series (9.2) cannot be evaluated as such. The approximation developed in Ref. 11 relies on neglecting the blip-blip interactions. This is equivalent to replacing in Eq. (9.2) the kernel $K(t,t')$ by its average value,

$$\langle K(t-t')\rangle=\tilde{\Delta}_0{}^2e^{\varphi(t-t')}, \tag{9.4}$$

which results in the integral equation[30]

$$P_{\rm NIBA}(t)=1-\int_0^t d\tau\int_0^{\tau}d\tau'\langle K(\tau-\tau')\rangle P_{\rm NIBA}(\tau'). \tag{9.5}$$

Evaluating the kernel, applying a Markov approximation, and discarding an insignificant difference in the frequency argument of the self-energy, we obtain

$$P_{\rm NIBA}(z)=-[z+\Sigma_{zz}(\tilde{\Delta}_0)-\tilde{\Delta}_0{}^2/z]^{-1}. \tag{9.6}$$

As to the damping rate in the coherent regime, one finds, instead of Eq. (8.7),

$$\Gamma_{\rm NIBA}=\frac{1}{2}(\gamma_{\rm u}+\gamma_{\rm g}). \tag{9.7}$$

In view of the weak-coupling result (9.1) the leads us to the conclusion that NIBA gives the correct terms of odd order in the rate. Yet it fails with respect to those of even order, where it misses a factor of 2; cf. Ref. 30. Formally, there is a similar discrepancy in the overdamped regime, where we have $\Gamma_{\rm NIBA}=(\gamma_{\rm u}+\gamma_{\rm g})$. Well beyond the crossover temperature, however, we find $\Re\Gamma_+=\Gamma_{\rm NIBA}$, i.e., blip-blip interaction is immaterial.

Calculating lowest-order corrections to NIBA requires one to evaluate in the series (9.2) correlations of blips at different times. Since any factor $\langle K\rangle$ is already accounted for by Eq. (9.4), we define the blip fluctuation

$$\delta K(t,t')=K(t,t')-\langle K(t-t')\rangle. \tag{9.8}$$

Then the lowest-order corrections arise from correlations of two such factors,

$$\langle\,\delta K(\tau_1,\tau_2)\,\delta K(\tau_3,\tau_4)\rangle, \tag{9.9}$$

It turns out that $\langle K(t,t')\rangle$ does not contain all two-time correlations of fluctuation operators

$$\xi_\pm=B_\pm-B. \tag{9.10}$$

In particular, NIBA misses the correlation of adjacent fluctuation operators belonging to *different* factors K,

$$\Delta_b^4B^2\langle\xi_{\beta_2}(\tau_2)\xi_{\beta_3}(\tau_3)\rangle, \tag{9.11}$$

which is, however, present in Eq. (9.9). This term has been evaluated in Ref. 30.

In order to recover the present result (6.10), however, one has to resum an infinite series of blip-blip interactions. As to the third-order term

$$\langle\,\delta K(\tau_1,\tau_2)\,\delta K(\tau_3,\tau_4)\,\delta K(\tau_5,\tau_6)\rangle, \tag{9.12}$$

we have to retain correlations of adjacent operators according to

$$\Delta_b^6B^2\langle\xi_{\beta_2}(\tau_2)\xi_{\beta_3}(\tau_3)\rangle\langle\xi_{\beta_4}(\tau_4)\xi_{\beta_5}(\tau_5)\rangle, \tag{9.13}$$

with $\beta_n=\pm$. A few comments on these blip-blip interactions are in order. (i) There are no bath correlations from τ_{2n-1} to τ_{2n}. (ii) Since the labels β_{2n} and β_{2n+1} are uncorrelated, the resulting bath correlation function involves even powers of the phase φ only, i.e., it is proportional to $\cosh[\varphi(\tau_{2n}-\tau_{2n+1})]$. (iii) Equation (9.11) contains one factor $\cosh[\varphi(\tau_2-\tau_3)]$, the three-blip term factorizes in two such factors; the n-blip correction involves $(n-1)$ such factors. As a consequence, all these corrections give rise to time convolutions in the series (9.2).

By summing an *infinite* number of such blip-blip correlations, integrating over all intermediate times, and taking the Laplace transform, one obtains $P(z)$ as given in Eq. (6.10). In comparison with the NIBA result (9.6), the blip-blip corrections have added Σ_{yy} in the denominator of the last term. The partial summation of Ref. 30 corresponds to linearizing the additional term in Eq. (6.10) as $\tilde{\Delta}_0^2[z+\Sigma_{yy}]^{-1}\rightarrow\tilde{\Delta}_0^2/z-\Sigma_{yy}\tilde{\Delta}_0^2/z^2$.

Finally we note that this resummation scheme is not unique. It may formally be improved by retaining additional correlations. A perturbation expansion in powers of φ, however, indicates that these extra terms are insignificant. Together with the perturbative approach of Ref. 30, these considerations lead us to the conclusion that Eq. (6.10) constitutes the proper solution for the spin-phonon model with cubic bath spectral density.

Experimental relevance

Here we discuss a few measurements of the phonon-driven damping rate of two-state systems in both conducting and insulating materials. In metallic compounds, there is competition between damping by conduction electrons and phonons; at low temperatures, the former are dominant, whereas the phonon mechanism prevails at high T. There are at least two examples where the strong increase of the damping rate due to the coupling to lattice vibrations has been observed, namely bistable defects in Bi wires and quantum diffusion of interstitial hydrogen in $Nb(OH)_x$.

Tunneling of bistable defects in submicrometer Bi wires causes significant conductance fluctuations (Refs. 3,4, and references cited therein). When measuring the resistance of the wire as a function of time, one observes the ''telegraph noise'' of a two-state system. For temperatures below 1 K, the tunneling motion is strongly affected by the interaction with conduction electrons;[12,11] its rate decreases with T as $\Gamma \propto T^{2K-1}$. For a given tunneling system, Chun and Birge[4] find a Kondo parameter $K=0.16$ and a tunnel energy $\hbar\Delta_0/k_B=1.9\times10^{-7}$ K. Above 1 K, however, phonon coupling becomes predominant and results in an exponentially increasing rate; from Fig. 1(b) of Ref. 4 we obtain for the temperature scale T_0 a value of about 1 K.

The quantum motion of interstitial hydrogen trapped by an oxygen impurity in niobium is well described as a two-state tunneling system. Inelastic neutron scattering at low temperature ($T<10$ K) revealed coherent motion with a tunnel frequency $\hbar\Delta_0/k_B=2.4$ K; the damping by conduction electrons is described by a Kondo parameter $K=0.055$.[9] Between 10 and 60 K, incoherent motion with a rate $\Gamma \propto T^{2K-1}$ has been observed by quasielastic neutron scattering.[10] Above 60 K, coupling to thermal motion of the lattice leads to a strongly increasing rate; from Fig. 3 of Ref. 10 we derive a value for T_0 of about 25 K.

Now we turn to insulating materials, where phonons provide the only damping mechanism. Substitutional lithium defects in potassium chloride form tunneling states with an energy splitting of 1.65 K for ^{6}Li and 1.1 K for the lighter isotope ^{7}Li. From sound velocity measurements below 10 K, Hübner *et al.* derived an elastic deformation potential $\gamma=0.04$ eV.[38]

Most dynamic experiments on tunneling systems in glasses involve linear-response functions with respect to a time-dependent elastic or electric field. Since the configurational average involves the broad distribution for the parameters of the two-state systems, the observed acoustic and dielectric properties do not permit rigorous conclusions. On the other hand, nonlinear response functions arise from a well-defined subensemble of tunneling systems and thus allow a more thorough comparison with theory.

As an example we mention two-pulse echoes observed by Hunklinger and Arnold.[2] The resonance condition singles out defects with an energy splitting $E=\hbar\omega_t$, where $\omega_t/2\pi=760$ MHz is the oscillation frequency of the applied elastic waves. Since such an experiment probes mainly systems with small asymmetry energy, we have $E\approx\hbar\Delta_0$, resulting in $\hbar\Delta_0/k_B\approx35$ mK. From a fit of Eq. (8.6) to these data we have obtained $\gamma=2.6$ eV. (Hunklinger and Arnold[2] derived a slightly different value $\gamma=3$ eV.) The data cover only the

TABLE I. Parameters for tunneling defects in various materials. For a-Bi and $Nb(OH)_x$, the crossover temperature T_0 is taken from Fig. 1(b) of Ref. 4 and Fig. 3 of Ref. 10; the elastic deformation γ is calculated according to Eq. (9.14). For the insulating systems, γ is derived from measured values for the low-temperature damping rates, and T_0 is obtained from Eq. (9.14).

	ϱ (g/cm^3)	$v_{l,t}$ (km/s)	γ (eV)	T_0 (K)	$\hbar\Delta_0/k_B$ (K)
$Nb(OH)_x$	8.4	5.1/2.1	0.2	25	2.4
KCl:Li	2.0	3.9/2.4	0.04	98	1.65/1.1 [a]
Bi	9.8	2.3/1.1	1.6	0.85	1.9×10^{-7}
a-SiO_2	2.2	5.8/3.8	2.6	4.8	0.035

[a]1.65 for the lighter isotope ^{6}Li, and 1.1 for ^{7}Li impurities.

temperature range where the rate is dominated by the direct, or one-phonon, process. From Eq. (8.12) we expect an exponential increase of the rate above a few K.

According to Eqs. (1.5) and (1.6), the elastic deformation potential γ and the temperature T_0 are related through

$$k_B T_0=\gamma^{-1}\sqrt{\frac{1}{3}\hbar^3\varrho v^5} \tag{9.14}$$

in an unambiguous fashion, since the average sound velocity v and the mass density ϱ are known.

In Table I we compare these parameters for a few crystalline and amorphous solids with tunnel defects. Starting from measured values of the elastic deformation potential γ for the insulating systems a-SiO_2 (Ref. 2) and KCl:Li,[38] we calculate the temperature T_0 where incoherent motion arising from phonon damping is supposed to set in. For $Nb(OH)_x$ and defects in mesoscopic Bi wires, we proceed in the opposite way: The values for T_0 have been taken from fits to Fig. 1(b) of Ref. 4 and Fig. 3 of Ref. 10; then we calculate the deformation potential according to Eq. (9.14).

The values obtained for defect atoms in crystalline materials are very similar. Both interstitial hydrogen in Nb and Li impurities in KCl are weakly coupled to elastic waves; the elastic deformation of about 50 meV corresponds to a crossover temperature T_0 of about 100 K. On the other hand, the configurational defects display an elastic deformation energy of several eV, leading to incoherent tunneling at a few Kelvin. This distinction between tunneling systems in crystalline and amorphous materials seems to be valid in general: Values similar to those for a-SiO_2 have been reported for other oxide glasses as a-GeO_2 and a-B_2O_3,[39] whereas the numbers given for KCl:Li are characteristic for a whole class of doped alkali halides.[8]

A final remark concerns the phonon overlap matrix element (7.29), which accounts for the dressing effect and the reduction of the tunnel frequency, according to Eq. (2.9). At zero temperature we have

$$B_0=\exp[-(\Theta/2\pi T_0)^2]. \tag{9.15}$$

With typical values for the Debye temperature Θ of a few hundred K, we find $B_0\approx1$ for substitutional and interstitial defects, i.e., the phonon dressing effect for impurities in crystals is weak.

The situation is very different for the configurational defects. According to Table I, the Debye temperature is by one or two orders of magnitude larger than T_0, resulting in a very small factor B_0. The tunnel energy Δ_0 measured for mesoscopic Bi wires is by about seven orders of magnitudes smaller than those for impurities in crystals, confirming qualitatively the law (9.15).

When applying this law to tunneling systems in glasses, one encounters an inconsistency of the data of Table I with experimental findings. The values for Θ and T_0 would indicate a dressing effect of many orders of magnitudes, which is not compatible with the observed tunnel energies up to $\hbar\Delta_0/k_B \approx 4$ K. Most probably, the phonon model used here does not apply to amorphous solids. In particular, it would seem that in glasses the high-frequency phonons are replaced by strongly damped local oscillators, thereby reducing considerably the effective Debye temperature Θ. Thus the present treatment would overestimate the dressing effect arising from high-frequency modes.

Finally we recall that we have treated the cubic part of the phonon bath spectral density only. As mentioned in the introductory section, an inversion symmetry or quadratic coupling to the elastic amplitude may change the power law of the leading term. In most cases, however, the cubic part would seem to be the relevant one. For tunneling systems in oxide glasses, the observed shift of the sound velocity maximum with applied frequency, $T_{\mathrm{max}} \propto \omega^{1/3}$, provides an experimental confirmation of this statement.[40]

X. SUMMARY AND CONCLUSION

We have developed a strong-coupling approach to the spin-phonon model with cubic bath spectral density, which is based on a perturbation series in terms of the quantum Liouville operator $\mathcal{L}$. Starting from the static part of the polaron operators $B_\pm$, we have treated their fluctuations ξ_g and ξ_u as a perturbation [cf. Eq. (2.6)].

As a crucial step we have decomposed the interaction part of the Liouvillian in commutators and anticommutators according to Eq. (4.6), and thus obtained the self-energy in terms of correlation functions of the bath response and correlation operators (4.10). The resulting expression for the self-energy corresponds to a series in powers of the tunnel frequency Δ_b. In order to evaluate the second-order contribution explicitly, we have calculated the spectra of the relevant bath correlations (7.1).

After applying a pole approximation, we have derived explicit expressions for the damping rates. The detailed comparison with the results of weak-coupling perturbation theory and the blip expansion in Sec. IX confirmed the validity of the present approach. Here we briefly summarize the main results.

(i) The damping rate, as arising from the noninteracting blip approximation, has been given as an infinite series, which permits one to recover previous results by taking appropriate limits; cf. Eqs. (7.16)–(7.23).

(ii) Contrary to the Ohmic case, the damping rates cannot be given in closed form. We propose an approximate formula for the bath correlation spectrum, Eq. (7.25), which interpolates smoothly between the one-phonon spectrum at low temperature and the result from saddle-point integration at high temperature.

(iii) The present work confirms a crossover from coherent tunneling to incoherent motion at a temperature T^*, as derived previously in Ref. 30. According to Eq. (8.14), T^* is essentially determined by T_0. This crossover is already present when treating the two-state dynamics in NIBA and retaining the whole series (7.19), i.e., retaining multiphonon processes of any order.

(iv) In order to obtain a controlled approximation for the cubic bath spectrum, one has to go beyond NIBA and retain certain blip-blip interactions. It turns out that the present approach corresponds to an infinite partial summation of such corrections. The resulting Eq. (6.10) constitutes the proper solution for the spin-phonon model.

(v) Phonon coupling affects the two-state dynamics in two ways: The dressing effect reduces the effective tunnel frequency, whereas at higher temperatures, phonon-assisted tunneling results in an exponentially increasing rate. For several materials, we have derived the values of the crossover temperature T_0, and we have discussed the relevance of our results for configurational defects and impurity atoms in various systems.

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APPENDIX A

Spectral representation

Because of the different conventions that can be found in the literature, we give the Laplace transformation as it is used in this paper,

$$f(z) = i\int_{-\infty}^{\infty} dt e^{izt} f(t) \quad (\Im z > 0). \tag{A1}$$

For $\Im z \to 0$, the function $f(z)$ may be written as

$$f(\omega + i0) = f'(\omega) + i f''(\omega), \tag{A2}$$

where the spectral function f'' is given by the Fourier transform,

$$f''(\omega) = \frac{1}{2}\int_{-\infty}^{\infty} dt e^{i\omega t} f(t), \tag{A3}$$

and f' by the Kramers-Kronig relation

$$f'(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega' \frac{f''(\omega')}{\omega' - \omega}. \tag{A4}$$

For real and symmetric functions $f(t)$, both $f'(\omega)$ and $f''(\omega)$ are real, i.e., Eq. (A2) separates real and imaginary parts of $f(z)$.

Reactive part Γ'

Here we consider the reactive part of the self-energy matrix. Since the Kramers-Kronig integral (A4) is determined by high-frequency contributions, we need to use the exact expression rather than the approximate (7.25), when evaluating $\Gamma'(\omega)$.

We expand the exponential in Eq. (7.6) in a power series in $\varphi(t)$ and take the Fourier transform of each term,

$$\Gamma''(\omega)=\frac{1}{2}\tilde{\Delta}_0^2\sum_{n=1}^{\infty}\frac{1}{n!}[\kappa_n''(\omega)+\kappa_n''(-\omega)], \qquad \text{(A5)}$$

where we use the shorthand notation

$$\kappa_n{}'(\omega)=\frac{1}{2}\int_{-\infty}^{\infty}dte^{i\omega t}\varphi(t)^n. \qquad \text{(A6)}$$

Using the symmetry properties of Eq. (A4), we easily find that the real part $\Gamma'(\omega)$ is given by a corresponding series,

$$\Gamma'(\omega)=\tilde{\Delta}_0^2\sum_{n=1}^{\infty}\frac{1}{n!}\gamma_n(\omega) \qquad \text{(A7)}$$

with

$$\gamma_n(\omega)=\frac{1}{2}[\kappa_n'(\omega)-\kappa_n'(-\omega)]. \qquad \text{(A8)}$$

Due to the frequency dependence of $\varphi''(\omega)$, the Kramers-Kronig integral is determined by high frequencies. At moderate frequency and temperature, i.e., $T\ll\Theta$ and $|\omega|\ll\omega_{\mathrm{D}}$, we may replace $\Gamma'(\omega)$ by its value at $T=0$; finite-temperature corrections are small.

Hence we consider the zero-temperature limit for the coupled phonon spectrum,

$$\varphi''(\omega)=\begin{cases}2\pi\alpha\omega & \text{for } 0\leqslant\omega\leqslant\omega_{\mathrm{D}}\\ 0 & \text{else}\end{cases} \quad (T=0). \qquad \text{(A9)}$$

According to Eq. (A6), the Kramers-Kronig integral $\kappa_n'(\omega)$ may be written in terms of an n-fold convolution of $\varphi''(\omega)$. After inserting the zero-temperature expression (A9) we obtain

$$\gamma_n(\omega)=\omega(2\alpha)^n\int_0^{\omega_{\mathrm{D}}}d\omega_1\cdots\int_0^{\omega_{\mathrm{D}}}d\omega_n \times\frac{\omega_1\cdots\omega_n}{(\omega_1+\ldots+\omega_n)^2-\omega^2}. \qquad \text{(A10)}$$

The first two terms are easily integrated,

$$\gamma_1(\omega)=\omega 2\alpha\ln(\omega_{\mathrm{D}}/\omega), \qquad \text{(A11)}$$

$$\gamma_2(\omega)=\omega(2\alpha)^2\omega_{\mathrm{D}}^2\left[\ln(2)-\frac{1}{2}\right], \qquad \text{(A12)}$$

where we have neglected corrections of the order $\omega/\omega_{\mathrm{D}}$. The presence of the logarithmic factor renders the first term a bit particular; because of the factors $\omega_1\cdots\omega_n$ in Eq. (A10), there is no such factor in the higher orders, as shown explicitly in Eq. (A12) for the quadratic term.

For this reason, we may expand the integrand in powers of ω^2, and integrate the term of zero order,

$$\gamma_n(\omega)=\omega(2\alpha)^n\omega_{\mathrm{D}}^{2(n-1)}I_n[1+O(\omega/\omega_{\mathrm{D}})]. \qquad \text{(A13)}$$

Here we have substituted $x_i\equiv\omega/\omega_{\mathrm{D}}$ and defined the integral

$$I_n=\int_0^1dx_1\cdots\int_0^1dx_n\frac{x_1\cdots x_n}{(x_1+\ldots+x_n)^2}. \qquad \text{(A14)}$$

With increasing n, the coefficients I_n tend towards zero; we give those for $n=2,3,4$,

$$I_2=\ln 2-\frac{1}{2}, \qquad \text{(A15)}$$

$$I_3=\frac{3}{8}[\ln(3)-1], \qquad \text{(A16)}$$

$$I_4=\frac{9}{4}\ln 3-\frac{28}{9}\ln 2-\frac{11}{36}, \qquad \text{(A17)}$$

which indicate a rapid convergence of the series (A7).

In order to obtain an upper bound for that series, we resort to the following approximations for the terms of order $n\geqslant 2$. Since the integrand is positive, discarding the term $x_2+\cdots+x_n$ in the denominator provides a strict upper limit for I_n. Then the n integrals factorize, resulting in the inequality

$$I_n<2^{1-n} \quad \text{for} \quad n\geqslant 2. \qquad \text{(A18)}$$

Inserting Eqs. (A11) and (A18) in the expression for $\Gamma'(\omega)$ and using $W_0=\alpha\omega_{\mathrm{D}}^2$ and $\Delta_0^2=\Delta_{\mathrm{b}}^2e^{-W_0}$, we obtain

$$\Gamma'(\omega)<2\omega\alpha\Delta_{\mathrm{b}}^2\left[e^{-W_0}[\ln(\omega_{\mathrm{D}}/\omega)-1]+\frac{1-e^{-W_0}}{W_0}\right]. \qquad \text{(A19)}$$

In physical terms, $\Gamma'(\omega)$ describes a frequency shift due to phonon coupling. It is negligible if $\Gamma'(\Delta_0)\ll\Delta_0$ or, equivalently, if $\partial_\omega\Gamma'(\omega)\ll 1$ for $\omega=\Delta_0$. In this case, the $\mathcal{Z}$ factor $[1+\partial_\omega\Gamma']^{-1}$ is close to unity.

With the weak-coupling condition (1.9) it is clear that the second term in brackets is irrelevant. When inserting $\omega=\Delta_0$ we find for the derivative of the first term

$$2\alpha\Delta_{\mathrm{b}}^2e^{-W_0}\left[\ln(\omega_{\mathrm{D}}/\Delta_{\mathrm{b}})+\frac{1}{2}W_0-2\right].$$

Rewriting the argument of the logarithm as $(\omega_{\mathrm{D}}/\Delta_{\mathrm{b}})=\sqrt{\alpha\omega_{\mathrm{D}}^2/\alpha\Delta_{\mathrm{b}}^2}$, we see that this quantity is small in fact.

In summary we have shown that the derivative of $\Gamma'(\omega)$ at $\omega=\Delta_0$ is much smaller than unity,

$$\partial_\omega\Gamma'(\omega)|_{\omega=\Delta_0}\ll 1, \qquad \text{(A20)}$$

which justifies our neglecting the reactive part of the self-energy in Sec. VI.

In deriving Eq. (A20) we heavily relied on the weak-coupling condition (1.9). A more thorough investigation of the real part Γ' for the opposite case would seem most interesting.

[1] J. Jäckle, Z. Phys. **257**, 212 (1972).

[2] S. Hunklinger and W. Arnold, in *Physical Acoustics*, edited by R. N. Thurston and W. P. Mason (Academic, New York, 1976); S. Hunklinger and A. K. Raychaudhuri, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (Elsevier, Amsterdam, 1986), Vol. IX.

[3] B. Golding, N. M. Zimmerman, and S. N. Coppersmith, Phys. Rev. Lett. **68**, 998 (1992).

[4] K. Chun and N. O. Birge, Phys. Rev. B **48**, 11 500 (1993).

[5] G. Weiss, W. Arnold, K. Dransfeld, and H. J. Güntherodt, Solid State Commun. **33**, 111 (1980).

[6] *Amorphous Solids – Low-Temperature Properties*, edited by W. A. Phillips (Springer, Berlin, 1981).

[7] G. Baier, C. Enss, and M. von Schickfus, Phys. Rev. B **40**, 9868 (1989).

[8] A. Würger, *From Coherent Tunneling to Relaxation*, Springer Tracts in Modern Physics Vol. 135 (Springer, Berlin, 1997).

[9] H. Wipf, D. Steinbinder, K. Neumaier, P. Gutsmiedel, A. Magerl, and A.-J. Dianoux, Europhys. Lett. **4**, 1379 (1987).

[10] D. Steinbinder, H. Wipf, A. Magerl, D. Richter, A.-J. Dianoux, and K. Neumaier, Europhys. Lett. **6**, 535 (1988).

[11] A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, Rev. Mod. Phys. **59**, 1 (1987).

[12] J. Kondo, Physica B & C **84**, 40 (1976); **84**, 207 (1976); **125**, 279 (1984); **126**, 377 (1984).

[13] H. Grabert and U. Weiss, Phys. Rev. Lett. **54**, 1605 (1985); H. Grabert, L. Linkwitz, S. Dattagupta, and U. Weiss, Europhys. Lett. **2**, 631 (1986).

[14] M. P. A. Fisher and A. T. Dorsey, Phys. Rev. Lett. **54**, 1609 (1985).

[15] C. Aslangul, N. Pottier, and D. Saint-James, J. Phys. (Paris) **47**, 1657 (1986).

[16] U. Weiss, *Quantum Dissipative Systems*, Series in Modern Condensed Matter Physics, Vol. 2 (World Scientific, Singapore, 1993).

[17] H. Grabert, Phys. Rev. B **46**, 12 753 (1992).

[18] C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963).

[19] Yu. Kagan and M. I. Klinger, J. Phys. C **7**, 2791 (1974).

[20] J. Kondo, Hyperfine Interact. **31**, 117 (1986).

[21] Yu. Kagan and N. V. Prokof'ev, Sov. Phys. JETP **69**, 1250 (1989).

[22] T. Regelmann, L. Schimmele, and A. Seeger, Z. Phys. B **95**, 441 (1994).

[23] H. Grabert, U. Weiss, and H. R. Schober, Hyperfine Interact. **31**, 147 (1986); H. Grabert, Phys. Rev. B **46**, 12 753 (1992).

[24] R. Pirc and P. Gosar, Phys. Kondens. Mater. **9**, 377 (1969).

[25] Q. Niu, J. Stat. Phys. **65**, 317 (1991).

[26] R. Beck, W. Götze, and P. Prelovsek, Phys. Rev. A **20**, 1140 (1979).

[27] W. Zwerger, Z. Phys. B **53**, 53 (1983); **54**, 87 (1983).

[28] W. Götze and G. M. Vujicic, Phys. Rev. B **38**, 9398 (1988).

[29] P. Neu, Ph.D. thesis, Universität Heidelberg, 1994; P. Neu and A. Würger, Z. Phys. B **95**, 385 (1994).

[30] A. Würger, Phys. Rev. Lett. **78**, 1759 (1997).

[31] A. Würger, J. Phys.: Condens. Matter **9**, 5543 (1997).

[32] T. Holstein, Ann. Phys. (N.Y.) **8**, 343 (1959).

[33] H. Horner (unpublished).

[34] G. D. Mahan, *Many-Particle Physics* (Plenum, New York, 1981).

[35] I. S. Gradsteyn and I. M. Ryzhik, *Table of Integrals, Series, and Products* (Academic, London, 1980).

[36] A. Würger (unpublished).

[37] U. Weiss, H. Grabert, and S. Linkwitz, J. Low Temp. Phys. **68**, 213 (1987).

[38] M. Hübner, C. Enss, and G. Weiss (private communication).

[39] S. Rau, C. Enss, S. Hunklinger, P. Neu, and A. Würger, Phys. Rev. B **52**, 7179 (1995).

[40] J. Classen, C. Enss, C. Bechinger, G. Weiss, and S. Hunklinger, Ann. Phys. (Leipzig) **3**, 315 (1994).