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Dynamics of the spin-boson model in the adiabatic approximation

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Abstract. We study the dynamics of the spin-boson model using a simple mean-field-type approximation which treats phonons classically. This simple approximation allows us to cover the whole range of system parameters and to determine boundaries between regimes with different behaviours. The validity of the model is tested by its ability to recover most of the previous results. The results are also compared with the results of some more sophisticated variational approaches.

1. Introduction

A variety of important phenomena in some physical [1–18], chemical [19–24] and biological [25, 26] systems may be efficiently represented as the motion of a single particle between two equivalent minima. The simplest model which still contains most of the relevant information necessary for understanding the dynamics of such systems consists of a two-level system (TLS) coupled to a collection of harmonic oscillators simulating the influence of a dissipative environment:

$$H = -\Delta\sigma_z - \sigma_x \sum_q \lambda_q (a_q + a_{-q}^\dagger) + \sum_q \hbar\omega_q a_q^\dagger a_q. \tag{1}$$

Disregarding other applications, we shall restrict ourselves to the case of a 'particle' (spin, defect, electron, exciton, etc) in the crystal, where the lattice vibrations take over the role of the bath. Then each particular case of interest can be specified either by the choice of the explicit q-dependence of coupling parameter λ_q and phonon frequency ω_q, or by the particular choice of spectral density of states which is connected to the spin–phonon coupling as follows: $J(\omega) = \sum_q |\lambda_q|^2 \delta(\omega - \omega_q)$. As usual, Δ denotes the bare tunnelling energy, $a_q^\dagger$ (a_q) is the creation (annihilation) operator of phonon quanta while σ_i ($i = x, y, z$) are the Pauli matrices.

Depending on the ratio of the basic physical parameters ($\Delta\omega_B$ is the phonon band width and $E_B = \sum_q |\lambda_q|^2/\hbar\omega_q$ is the lattice relaxation energy or small polaron binding energy as usually referred to in the theory of self-trapping [27]) the system can display regimes with substantially different physical behaviours. Thus in the weak-coupling (WC) regime the mean position of the 'particle' follows an equation of motion identical with that of a damped harmonic oscillator [9, 21, 22], while in the strong-coupling (SC) regime a significant reduction in the tunnelling frequency Δ_{eff} appears because of phonon dressing and the system exhibits a pronounced tendency towards localization [1, 5, 10, 12, 27–29].

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The magnitude as well as the character of the dependence of Δ_{eff} on the coupling constant S ($\simeq E_B/\hbar\omega_B$) is governed by the value of the adiabatic parameter B ($\simeq 2\Delta/\hbar\omega_B$) and the particular type of spin–phonon coupling [29]. Knowledge of the behaviour of Δ_{eff} as a function of S and B is of particular interest for understanding the nature of the localization transition. In the present paper we shall not consider this problem and we shall concentrate on the system dynamics employing the (semi)classical approximation for phonons while treating the influence of quantum fluctuations perturbatively.

Bearing in mind that the spin–phonon system has been successfully examined by means of sophisticated methods such as path integral (PI) techniques [1, 14–16] and the renormalization group (RG) approach [1, 17, 18], the above-proposed typical mean-field (MF) approach could look like a step backwards. That is, since the MF approximation was applied to the spin-boson (SB) model a long time ago [10, 11], it is not very clear, at first sight, what new results can be obtained by now employing such an approximation.

Let us recall first that the above-mentioned RG and PI approaches were mainly related to the phenomenon of macroscopic quantum tunnelling [1, 14–18] where non-adiabaticity ($\Delta \ll \hbar\omega_B$) has been implicitly assumed. That is, the mapping of a realistic tunnelling system, e.g. magnetic flux trapped in an RF SQUID ring, to the spin–phonon model is rigorous in that case only.

On the other hand, in the number of so-called 'intrinsically' TLSs, parameters do not satisfy that condition. On the contrary, their values could lie in any particular region of the parameter space of the system, sometimes approaching extremely adiabatic and SC limits. In the existing literature, especially in the context of macroscopic quantum phenomena, the problem of the dynamics of the SB model under these conditions has not attracted any particular attention; so we find it interesting to deal with it now. For that purpose, the above-proposed application of the MF theory is a natural choice. That is, according to the previous studies [6, 10, 11, 16] of the equilibrium (thermodynamic) properties of the SB model, the MF approach represents a satisfactory theoretical framework for that purpose in the highly adiabatic limit ($\Delta \gg \hbar\omega_B$). Furthermore, in the SC limit we expect a significant manifestation of the (semi) classical behaviour of the phonon field. According to the non-conservation of the number of phonons, and disregarding their temperature excitation, the level of excitation of each phonon mode is exclusively influenced by the spin–phonon interaction. Consequently they become macroscopically (classically) occupied in the SC limit. Under these conditions, the presence of an excess particle may cause a local distortion of the surrounding lattice which, in turn, creates a potential well for that particle in which it can be trapped. For this reason, tunnelling dynamics can be significantly affected and even suppressed because of the above-described, typically polaronic effects, and it is our aim to examine what consequences it should have on SB dynamics. Furthermore, in order to determine the range of validity of the present adiabatic treatment, we shall compare it with the results of some previous studies. The problem is not of purely academic interest since the classical nature of the phonon field can be assigned to some realistic problems such as small-polaron dynamics [13, 27] or Kenkre's so-called non-linear dimer [30–33] which was recently proposed as a theoretical framework for a description of the energy transfer in the 'stick dimer' [34] and motion of hydrogen atoms trapped near the impurity atoms in metals [35].

2. Quasi-classical approximation

In order to achieve the above goal we have to find the equation of motion for the tunnelling probability $P(t) = \langle\sigma_x(t)\rangle$, and as the first step we separate the classical large-amplitude

(α_q) part of the phonon operators from the fluctuating part: $a_q = a_q - \alpha_q + \alpha_q = b_q + \alpha_q$. Here the classical phonon amplitudes α_q refer to that part of the phonon field which is engaged in the formation of the lattice deformation around the tunnelling particle. Under the above-emphasized (adiabatic) conditions, the lattice cannot follow the internal motion of the particle; so we can neglect the temporal fluctuation of α_q. On the other hand, the complex entity—particle plus surrounding lattice distortion—represents a stable (minimum-energy) state whose energy is lower than that in the rigid lattice by the amount gained in the creation of lattice deformation. Therefore, we shall find α_q by minimizing the ground-state (GS) energy of the system. The new operators b_q ($b_q = a_q - \alpha_q$) are a small correction to the classical part, and their influence will be treated perturbatively. Substituting the above separation of phonon operators into the slow (classical) and fluctuating part of the original Hamiltonian, we obtain an effective spin–phonon Hamiltonian which now reads

$$H = H_S + H_{SB} + H_B \tag{2}$$

where H_S denotes the effective spin Hamiltonian

$$H_S = -\Delta\sigma_z - \epsilon\sigma_x \qquad \epsilon = 2\delta E_B \tag{3a}$$

while H_{SB} and H_B denote the interaction and the Hamiltonian, respectively, of 'new' phonons:

$$H_{SB} = -(\delta - \sigma_x)\sum_q \lambda_q(b_q + b_{-q}^\dagger) \tag{3b}$$

$$H_B = \sum_q \hbar\omega_q b_q^\dagger b_q + \delta^2 E_B. \tag{3c}$$

Here δ denotes a new variational parameter introduced after specifying the classical amplitudes as $\alpha_q = \delta\lambda_q^*/\hbar\omega_q$. This is not an additional approximation but rather the anticipation of the explicit form of α_q. It can be confirmed easily by minimizing the GS energy of the system (see for example the classical approximation in [6, 10, 11]). Obviously, owing to the assumed classical nature of the phonons the original symmetry of the system is broken and the Hamiltonian of the spin subsystem is modified through the appearance of the symmetry-breaking term $\epsilon\sigma_x$ which can significantly violate the effects of tunnelling.

The above-proposed treatment is more flexible than that of Grigolini and co-workers [32, 33] who also introduced the analogous shift of phonon operators but for a fixed amount corresponding to the *ad hoc* choice $\delta = 1$ in our approach. It obviously corresponds to the maximally deformed lattice with the particle energy lowered by E_B and with an almost vanishing amplitude of internal oscillations of the particle in the well. However, such a choice could be justified in the extremely SC and adiabatic limit only, while in the intermediate region the depth of the potential well is balanced by its kinetic energy and their mutual ratio is expressed by the value of the parameter δ ranging from zero (WC) to unit (SC limit). Furthermore, our treatment also has some advantages with respect to the approaches of Feinberg and Ranninger [13] and Kenkre and co-workers [30, 31] because their non-linear equations no longer contain information on two-state properties of the original spin variables and their evolution equations should be equally valid for any spin larger than $\frac{1}{2}$.

The result of the above-described procedure is the separation of the original spin–phonon model with arbitrary SC into two weakly interacting subsystems: the relevant

subsystem (spin) and irrelevant subsystem (new phonons), where the weakness of interaction is provided by the smallness of the quantum fluctuations b_q. Therefore in deriving the equation of motion for the tunnelling amplitude $\langle\sigma_x(t)\rangle$ we can apply the perturbation procedure. The first step will be to eliminate the phonon variables from the equations of motion for relevant, i.e. spin, operators. For that purpose we shall use the method of quantum Langevin equations as interpreted by Shibata and co-workers [36–38]. Without going into details let us write their fundamental evolution equation for an operator A from the relevant system:

$$\dot{A}(t) = \exp(\mathrm{i}Lt)\,\mathrm{i}(L_S + \langle L_{SB}\rangle_B)A(0) + \exp(\mathrm{i}Lt) \times \int_0^t \mathrm{d}\tau \exp(-\mathrm{i}L_S t)(\mathrm{i}L_{SB}\exp[\mathrm{i}(I_S + I_B)\tau]\mathrm{i}L_{SB})_B A(0) + K(t) \tag{4}$$

$$K(t) = (1 - \hat{P})\exp[\mathrm{i}(L_S + L_B)t]\,\mathrm{i}L_{SB}A(0).$$

Here L denotes the Liouville operator which acts as follows: $\mathrm{i}LA = (\mathrm{i}/\hbar)[H, A]$; $\exp(\mathrm{i}Lt)\,A(0) = \exp(\mathrm{i}/\hbar Ht)\,A(0)\exp(-\mathrm{i}/\hbar Ht)$. If L is supplied with some index (S, B or SB), it means that, in (5), one should use that part of the total Hamiltonian (2) which is specified by the particular index. $K(t)$ denotes the so-called fluctuating force, while $\hat{P}$ is the projection operator: $\hat{P}\hat{O} = \mathrm{Tr}[\rho_B\hat{O}(t)] = \langle O(t)\rangle_B$ (ρ_B is the phonon density matrix). Derivation of the equations of motion is straightforward but very cumbersome and tedious; so we shall avoid this purely technical problem and write the final results only:

$$\begin{aligned}
\dot{\sigma}_x &= (2\Delta/\hbar)\sigma_y \\
\dot{\sigma}_y &= -(2\Delta/\hbar)\sigma_x + (2\epsilon/\hbar)\sigma_z - \lambda\sigma_y + K_y(t) \\
\dot{\sigma}_z &= -(2\epsilon/\hbar)\sigma_y - (\epsilon/\hbar)\lambda\sigma_x - \lambda\sigma_z + (\hbar\Omega/2\Delta)\lambda_0 + K_z(t)
\end{aligned} \tag{5}$$

where λ defines the friction constant:

$$\lambda = \left(\frac{2\Delta}{\hbar\Omega}\right)^2 \frac{1}{\hbar^2}\int_0^t \mathrm{d}\tau \sum_{q,q'} \lambda_q\lambda_{q'}\cos(\Omega t)\langle[b_q\exp(-\mathrm{i}\omega_q t) + b_q^\dagger\exp(\mathrm{i}\omega_q t)](b_{q'}(0) + b_{q'}^\dagger(0))\rangle_B. \tag{6}$$

It can be calculated explicitly in the so-called short-correlation-time limit which assumes that the bath relaxation time is very short compared with the scale of changes in $\sigma_i(t)$, so that we can extend the integration limit in (4) and (6) towards infinity. Thus we obtain

$$\lambda = \left(\frac{2\Delta}{\hbar\Omega}\right)^2 \frac{4\pi}{\hbar^2}\sum |\lambda_q|^2(\bar{\nu}_q + \tfrac{1}{2})\delta(\omega_q - \Omega) \qquad \lambda_0 = \lambda(T = 0\ \mathrm{K}) \tag{7}$$

which apart from the numerical pre-factor $2\Delta/\hbar\Omega$ is equivalent to the expression obtained by Harris and Silbey [9] in the WC limit. In our case, the biased frequency is determined by the strength of interaction and in the WC limit it should vanish so that $2\Delta/\hbar\Omega$ tends to unity and we recover the results of [9]. Here $\Omega = (2/\hbar)\sqrt{\Delta^2 + \epsilon^2}$. The fluctuating forces K_y and K_z are given by

$$\begin{aligned}
K_y(t) &= 2\sigma_z^{(0)}(t)\sum_q \lambda_q[b_q\exp(-\mathrm{i}\omega_q t) + b_{-q}^\dagger\exp(\mathrm{i}\omega_q t)] \\
K_z &= -2\sigma_y^{(0)}(t)\sum_q \lambda_q[b_q\exp(-\mathrm{i}\omega_q t) + b_{-q}^\dagger\exp(\mathrm{i}\omega_q t)].
\end{aligned} \tag{8}$$

Here $\sigma_i^{(0)}(t)$ denote the solutions of the Heisenberg equations in the absence of fluctuations:

$$\begin{aligned}
\sigma_x^{(0)}(t) &= \sigma_x[(2\epsilon)^2 + (2\Delta)^2\cos(\Omega t)]/(\hbar\Omega)^2 \\
&\quad + [2\Delta\epsilon/(\hbar\Omega)^2]\sigma_z[1-\cos(\Omega t)] + (2\Delta/\hbar\Omega)\sigma_y\sin(\Omega t) \\
\sigma_y^{(0)}(t) &= \sigma_y\cos(\Omega t) - (2/\hbar\Omega)(\Delta\sigma_x - \epsilon\sigma_z)\sin(\Omega t) \\
\sigma_z^{(0)}(t) &= \sigma_z + (\epsilon/\Delta)\sigma_x - (\epsilon/\Delta)\sigma_x^{(0)}(t).
\end{aligned} \tag{9}$$

Since our main interest is the derivation of the equation for the tunnelling probability $P(t)$, let us average the set of equations (6) over the equilibrium density matrix: $\rho = (1+\sigma_x)/2 \otimes \rho_B$. Thus, with the initial conditions $\langle\sigma_x(0)\rangle = 1$, $\langle\dot{\sigma}_x(0)\rangle = 0$ and $\langle\sigma_y(0)\rangle = \langle\sigma_z(0)\rangle = 0$, we finally obtain the set of Bloch equations

$$\begin{aligned}
\partial\langle\sigma_x(t)\rangle/\partial t &= (2\Delta/\hbar)\langle\sigma_y(t)\rangle \\
\partial\langle\sigma_y(t)\rangle/\partial t &= -(2\Delta/\hbar)\langle\sigma_x(t)\rangle + (2\epsilon/\hbar)\langle\sigma_z(t)\rangle - \lambda\langle\sigma_y(t)\rangle \\
\partial\langle\sigma_z(t)\rangle/\partial t &= -(2\epsilon/\hbar)\langle\sigma_y(t)\rangle - \lambda(\langle\sigma_z(t)\rangle + (\epsilon/\Delta)\langle\sigma_x(t)\rangle) + \lambda_0(\hbar\Omega/2\Delta).
\end{aligned} \tag{10}$$

Combining the last two equations to eliminate $\langle\sigma_z(t)\rangle$ and then eliminating $\langle\sigma_y(t)\rangle$, we finally obtain that $P(t)$ satisfies the equation of motion of a driven damped harmonic oscillator:

$$\ddot{P}(t) + \lambda\dot{P}(t) + \Omega^2 P(t) = 2\epsilon\Omega\lambda_0/\hbar^2\lambda + [(2\epsilon/\hbar)^2 - (2\epsilon\Omega/\hbar^2)(\lambda_0/\lambda)]\exp(-\lambda t) \tag{11}$$

which except for the 'driving force' term on the right-hand side is similar to those previously obtained by Harris and Silbey [9] and recently by Grigolini *et al* [32] in the WC limit. However, this time our analysis can be related to both WC and SC limits. This is determined by the value of the variational parameter δ ranging from zero to unity. Since it arises as a consequence of the assumed classical nature of phonons, its vanishing value should be related to the WC case, while $\delta \to 1$ corresponds to the SC limit and maximally expressed classical nature of the phonon field. Furthermore, since so far we have neglected the time dependence of the classical phonon amplitudes α_q which corresponds to disregarding the phonon kinetic energy as a first approximation, the validity of our approach demands that the adiabatic condition is satisfied too. In order to analyse the possibility of a localization transition let us write the solution of equation (11):

$$P(t) = A\exp(-\tfrac{1}{2}\lambda t)\cos(\bar{\Omega}t+\varphi) + [(2\epsilon/\hbar\Omega)^2 - (2\epsilon/\hbar\Omega)(\lambda_0/\lambda)]\exp(-\lambda t) + (2\epsilon/\hbar\Omega)(\lambda_0/\lambda) \tag{12}$$

where the amplitude, phase and effective tunnelling frequency satisfy

$$\begin{aligned}
A &= [1-(2\epsilon/\hbar\Omega)^2]\sqrt{1+\tan^2\varphi} \\
\tan\varphi &= (\lambda/2\bar{\Omega})[1-(\lambda_0/\lambda)^2]/[1-(2\epsilon/\hbar\Omega)^2] \\
\bar{\Omega} &= \sqrt{\Omega^2 - (\tfrac{1}{2}\lambda)^2}.
\end{aligned} \tag{13}$$

All information which can be obtained on the basis of the above equations is determined by the value of the parameter δ which follows after minimization of the GS energy of the effective spin–phonon system:

$$E_{GS} = -\sqrt{\Delta^2 + 4\delta^2 E_B^2} + \delta^2 E_B \tag{14}$$

from which we obtain

$$2\delta E_B \left[1 - 2E_B \Big/ \sqrt{\Delta^2 + 4\delta^2 E_B^2} \right] = 0. \tag{15}$$

It has two solutions: the symmetry-breaking solution $\delta = \sqrt{1 - (\Delta/2E_B)^2}$ corresponding to non-vanishing static lattice displacement and, consequently, to non-vanishing transverse spin polarization ($\langle\sigma_x\rangle \neq 0$ and $\langle\sigma_y\rangle \neq 0$) and $\delta = 0$ which preserves the symmetry. Strictly speaking, because of parity conservation of the original SB model, with H being invariant under the canonical transformation ($a_q \to -a_q$, $\sigma_x \to -\sigma_x$, $\sigma_y \to -\sigma_y$ and $\sigma_z \to \sigma_z$), both lattice distortion and transverse polarization should disappear. However, since we have focused our attention on a particular region of parameter space, i.e. the SC and adiabatic limit, we are dealing with the case where the classical nature of phonons should appear. Thus, as far as the $\Delta/2E_B < 1$ classical behaviour of phonons is more or less expressed, it can induce the localization of the 'particle' in the initial position.

This limit corresponds to the SC limit which will be clear later. If this condition is violated, $\delta = 0$ and we have the WC regime.

The second piece of information necessary to understand the system dynamics is knowledge of the friction constant and its dependence on system parameters. For that purpose we must specify the explicit forms of λ_q and ω_q.

As a first example let us consider the case of point coupling with dispersionless optical phonons: $\lambda_q = \chi(\hbar/2MN\omega_q)^{1/2}$; $\omega_q = \omega_0 =$ constant. This is in fact the well known molecular crystal (MC) model [27].

The other case that we shall consider will be the short-range coupling with acoustic phonons via the deformation potential: $\lambda_q = i\chi(q/|q|)(\hbar|q|R_0/2MN\omega_B)^{1/2}$; $\omega_q = c_0|q|$. We shall call this model the ADP model [29].

Now, one can introduce the set of dimensionless parameters, one of them (B) describing the adiabaticity and the other (S) giving the measure of the coupling strength: $S = E_B/\hbar\omega_0$; $B = 2\Delta/\hbar\omega_0$ for the MC model and $S = E_B/\hbar\omega_B q_D R_0$); $B = 2\Delta/\hbar\omega_B q_D R_0$ for the ADP model. (q_D denotes the Debye cut-off quasi-momentum and R_0 is the lattice constant.) In terms of these parameters, the condition for the applicability of the classical approximation becomes $B/4S < 1$ which, together with the above-mentioned adiabatic demands ($B \gg 1$), clearly defines the SC limit.

Let us now look at the friction constant value. For the MC model we have

$$\lambda = \begin{cases} (\Delta/2E_B)^2(4\pi\omega_0 E_B/\hbar)(\bar{v}_q + \frac{1}{2})\delta(\omega_0 - 4E_B/\hbar) & \text{for } \Delta < 2E_B \quad (16a) \\ (4\pi\omega_0 E_B/\hbar)(\bar{v}_q + \frac{1}{2})\delta(\omega_0 - 2\Delta/\hbar) & \text{for } \Delta > 2E_B. \quad (16b) \end{cases}$$

Obviously, damping has a resonant character, i.e. it exhibits a sudden jump when ω_0 approaches a certain value; otherwise it is zero. However, satisfying the resonance condition in the SC limit demands $\omega_0 = 4E_B/\hbar$ or equivalently $S = \frac{1}{8}$, which contradicts the above-emphasized SC and adiabatic conditions; so it cannot be satisfied for (16a). In contrast, in the WC case, resonance arises when $B = 1$, which is quite possible.

For the ADP model we have

$$\lambda = \left(\frac{2\Delta}{\hbar\Omega}\right)^2 \frac{4\pi E_B}{\hbar} n q_D^n \int_0^{q_D} q^n \delta\left(q - \frac{2\Omega}{\omega_B R_0}\right)(\bar{\nu}_q + \tfrac{1}{2})\, d^n q \tag{17}$$

where n is the dimensionality of the phonon subsystem. Here $n = 1$ corresponds to ohmic dissipation, while $n > 1$ defines super-ohmic dissipation. (Here, the introduction of the Debye cut-off is the natural consequence of the transition from the summation over phonon quasi-momenta to the integration.) Obviously, equation (17) can never be satisfied in the SC limit, since the argument of the δ-function in (17) is always out of the integration range. That is, substituting Ω defined in terms of δ, we see that the disappearance of the argument of the δ-function requires that $8E_B/\hbar\omega_B R_0 - q_D = 0$, implying that $S = \frac{1}{4}$, which is in sharp contradiction to the above SC and adiabatic demands. In the weak WC case,

$$\lambda = 2^n(4\pi/\hbar)E_B n B^n(\bar{\nu}_{q_0} + \tfrac{1}{2}) \qquad q_0 = 2B/R_0. \tag{18}$$

Obviously, as long as $\delta \neq 0$, damping is zero and $P(t)$ satisfies

$$P(t) = 1 - 2(\Delta/2E_B)^2 \sin^2(2E_B t/\hbar) \tag{19}$$

while, in the WC regime, $P(t)$ satisfies equation (12) with $A = (1 + \tan^2\varphi)^{1/2}$; $\tan\varphi = (\lambda/2\bar{\Omega})[1 - (\lambda_0/\lambda)^2]$ and $\bar{\Omega} = \sqrt{(2\Delta)^2 - (\frac{1}{2}\lambda)^2}$.

It follows from (19) that, as long as $2(\Delta/2E_B)^2 \leqslant 1$, the 'particle' is localized in the initial position (i.e. initial well), exhibiting harmonic oscillations inside the well and never leaving it. The amplitude of these oscillations decreases with increase in the coupling strength. In (S, B) language, the above localization condition is $S \geqslant B/\sqrt{8}$.

With proper identification of the corresponding parameters ($4E_B = \chi$ and $2\Delta = V$), our result is identical with that of Kenkre and Campbell [30] (cf equation (9) of [30]) which arises in the SC limit of their non-linear analysis. The non-dissipative character of our result (equation (19)) is the consequence of the fact that almost all phonons are frozen—engaged in the creation of the static lattice distortion—while the emission of the real phonons, the mechanism which is the cause of the dissipation, is forbidden since the energy balance ($\omega_q = \Omega$) (see equation (7)) condition is not satisfied in the SC limit. Clearly, such a picture can survive as long as the adiabatic limit is satisfied, which enables one to neglect temporal fluctuations of classical phonon amplitudes. However, the time dependence of α_q is not the only possible manifestation of non-adiabatic effects. That is, when B decreases, lattice deformation begins to follow the motion of the particle instantaneously and the well known phenomenon of a decrease in the tunnelling frequency due to phonon dressing arises. Thus, in order to determine properly the validity of the present analysis, we should distinguish the regions in the (S, B) plane where each particular type of behaviour prevails.

In the WC regime the 'particle' exhibits underdamped oscillations as long as $2\Delta/\hbar > \frac{1}{2}\lambda$. When this condition is violated, the transition to the overdamped regime occurs. Using the explicit expression for λ, we can define the overdamping condition as

$$S > [n2^{n-1}\pi B^{n-1}(\bar{\nu}_{q_0} + \tfrac{1}{2})]^{-1} \tag{20}$$

which means that the temperature stimulates transition to the overdamped regime. Thus, even when parameters of the system initially do not allow the transition to overdamping, it can be achieved when the temperature increases.

3. The range of validity

The above results obtained without taking into account the reduction in the effective tunnelling frequency due to phonon dressing clearly have limited validity. Thus, to estimate the region of parameter space where the above predictions could apply, we need to compare the present results with those previously obtained without neglecting the dressing. For that purpose, we shall use the results of our recent paper where the simple variational approach known from polaron theories [29] has been applied to study the dependence of Δ_{eff} on the system parameters S and B and type of coupling. Because of the variational character of both methods it is clear that the present (semiclassical) results should be valid in that part of (S, B) space where classical theory predicts lower estimates of the GS energy: $E_{\text{GS}}^{\text{class}} \leqslant E_{\text{GS}}^{\text{source}}$. The term 'source' was borrowed from Gross [28] and it is related to the variational approach developed in the polaron theories [27] and later successfully applied in studies of the spin–phonon model [12, 22, 28, 29]. Without going into the details which are presented in [29] let us quote

$$E_{\text{GS}}^{\text{source}} = -\Delta \exp\left(-2\sum |f_q|^2\right) - \sum \lambda_q (f_q + f_q^*) + \sum \hbar\omega_q |f_q|^2 \quad (21)$$

with f_q being the variational parameter satisfying

$$f_q = \lambda_q / [\hbar\omega_q + 2\Delta \exp(-x)] \quad (22)$$

where $x = 2\sum |f_q|^2$ defines the degree of dressing and reduction in effective tunnelling frequency. According to our previous study, the dependence of Δ_{eff} on x and the system parameters S and B is very different for each particular type of coupling; so, at this stage, we must make the particular model concrete. In the case of ohmic dissipation (ADP; $n = 1$) we have that the classical approach gives lower estimates for the GS energy if the following condition is satisfied:

$$-S - B^2/16S \leqslant -\tfrac{1}{2}B \exp(-x) - S[1 + 2B \exp(-x)]. \quad (23)$$

Let us now recall that the classical situation means the SC and adiabatic limit in which, according to our previous study, dressing is almost negligible ($x \to 0$). In that case, (23) can be written as

$$\{S[1 + (1 + 2B)^{-1/2}] - \tfrac{1}{4}B\}\{S[1 - (1 + 2B)^{-1/2}] - \tfrac{1}{4}B\} \geqslant 0. \quad (24)$$

Since we are in the SC region the first multiplier in (24) is always positive and the only limitation arises from the other multiplier which gives

$$S \geqslant \tfrac{1}{4}B[1 - (1 + 2B)^{-1/2}]^{-1}. \quad (25)$$

In this case, the equality (25) defines the so-called dressing boundary; the curve in the (S, B) plane which separates the regions in which the classical method (i.e. the present study) gives better predictions for spin–phonon dynamics than the so-called 'source' approximation. Since the 'source' method almost reproduces the known results of the PI and RG approaches in the appropriate limits (non-adiabatic) while having a wider domain of applicability when B increases [29], we can accept the present results as a reasonably good approximation for

the true dynamics of the spin–phonon system if (25) is satisfied. Analogous reasoning for the WC case leads to the condition $-\frac{1}{2}B \leqslant -\frac{1}{2}B\exp(-x) - S/(1+2B\exp(-x))$ which can be satisfied in the case of non-vanishing dressing only. This means that, in the WC case, Δ should be always substituted by $\Delta_{\text{eff}} = \Delta\exp(-x)$. However, since in this regime (small B and S), x is very small, we can consider this reduction in tunnelling frequency to be negligible.

Using analogous reasoning we can find dressing boundaries for the ADP type of coupling for $n = 2$ and 3 corresponding to the so-called super-ohmic dissipation: for $n = 2$,

$$S \geqslant \tfrac{1}{4}B\{1 - \sqrt{1 + 2B^2/(1+B) - 2B^2\ln(1+1/B)}\}^{-1}$$

and, for $n = 3$, (26)

$$S \geqslant \tfrac{1}{4}B\{1 - \sqrt{1 - 3B^2[1 + (B/1+B) - 2B\ln(1+1/B)]}\}^{-1}.$$

For the MC model we obtain the simple expression

$$S \geqslant \tfrac{1}{4}B\left\{1 - \sqrt{1 - [B/(1+B)]^2}\right\}^{-1}. \tag{27}$$

From our analysis it follows that the parameter space of the system (the (S, B) plane) is divided into three regions with substantially different physical behaviours of the system. The boundaries of these regions are defined by the two limits $S = \frac{1}{4}B$ and $S = B/\sqrt{8}$. So for $S < \frac{1}{4}B$ we encounter the WC regime where the particle exhibits underdamped oscillations with a transition to pure exponential decay $P \sim \exp(-Rt)$ when condition (20) is satisfied for ADP-type coupling, while having the resonant character (i.e. an abrupt transition to infinite damping when $2\Delta = \frac{1}{2}\lambda$) for the MC model. Here $R \sim (2\Delta)^2/\lambda$ defines the transition rate which is in full agreement with previous studies [9, 21, 22], while being in qualitative agreement with the work of Dekker [39] and Goerlich *et al* [40]. Concerning these last references, there appear to be certain differences, based on the crudeness of our approximation manifested in our extension of the integration boundary in equation (6) to infinity. In fact, in the WC limit, our bias frequency vanishes; so differences also disappear (cf equation (12) in [40] and equation (3.23) in [39]). Thus, as $S < \frac{1}{4}B$, with respect to the localization behaviour, dissipation can cause two substantially different effects. The first, characterized by a damped oscillation of the transition probability $P(t)$, represents the gradual relaxation towards $P(\infty) = 0$ which denotes a fully delocalized state. In the practical application to the problem of the tunnelling dynamics of handed molecules, this problem is called racemization and represents the gradual relaxation of the system predominantly populated with just one kind of molecule (left-handed for example) towards the mixture of equal amounts of both kinds. When the overdamping criterion is satisfied and when the system is initially localized, the localized state is essentially stabilized further owing to the very slow relaxation of $P(t)$ towards equilibrium. For $\frac{1}{4}B < S < B/\sqrt{8}$ we encounter, so to say, the intermediate-coupling (IC) region where, although being substantially violated, tunnelling is not fully suppressed. Finally for $S > B/\sqrt{8}$ the particle oscillates inside the initial well, never leaving it. The amplitudes of these localized oscillations are undamped and their amplitude vanishes as the coupling strength increases.

Our results are given in figure 1 where we have plotted the regions of parameter space with the three characteristic regions where each particular type of behaviour should be expected.

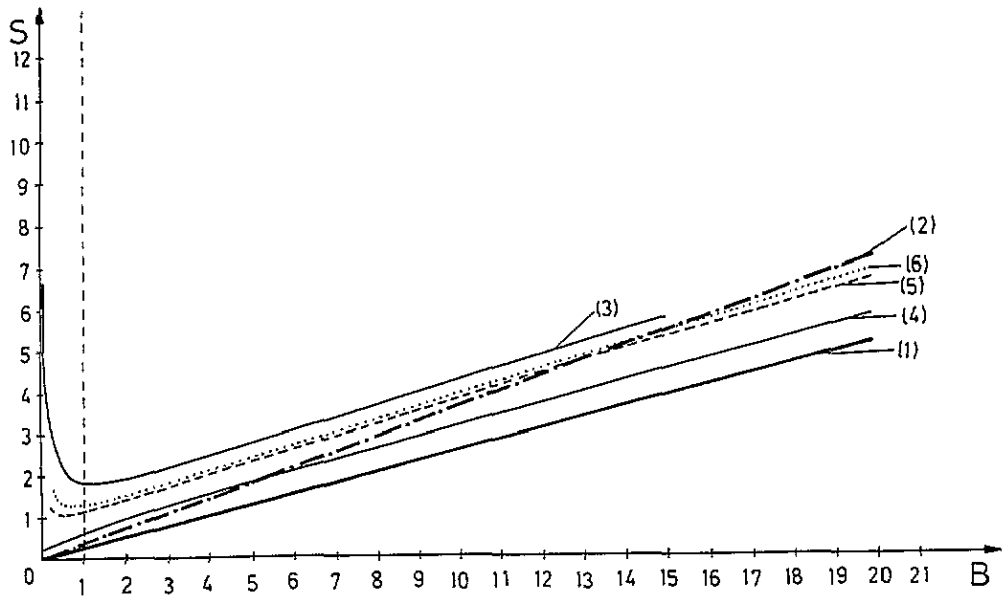

Figure 1. Characteristic curves in the (S, B) plane: curve (1), $S = \frac{1}{4}B$ WC versus IC boundary; curve (2), $B/\sqrt{8}$ SC boundary; curve (3), dressing boundary, MC model; curve (4) dressing boundary, ADP model, $n = 1$; curve (5), dressing boundary, ADP model, $n = 2$; curve (6), dressing boundary, ADP model, $n = 3$.

Clearly the validity of the above predictions is determined by the applicability of the classical approach and we estimate that our results could be accepted when conditions (25)–(27) are satisfied. Therefore, in figure 1, only for those points in parameter space lying above the so-called dressing boundaries could the above discussion apply. For the points below the dressing boundaries, the dynamics are determined by the reduction in the effective tunnelling frequency. The presence of these dressing boundaries implies a reduction in the validity of classical analysis and for each particular type of spin–phonon coupling there appears a crossing point $(S_c; B_c)$ of the dressing boundary and SC boundary which together with the adiabatic demand $(B > 1)$ define the restricted domain of applicability of the present approach. So, for example, for ohmic dissipation, the SC limit embraces all points in parameter space lying in the area $B > 1$ and above the dressing boundary, while the IC region can occur only for $B > B_c$ ($B_c \simeq 5.7$). A similar analysis for $n = 2$ gives $B_c \simeq 14$ and for $n = 3$ it gives $B_c \simeq 15$, while for the MC model it gives $B_c \simeq 21.7$.

4. Concluding remarks

Concluding this paper, let us note that the above-presented analysis embraces some of the known results while, at the same time, being complementary to some other approaches. So, for example, we have recovered the results of both WC [9] and SC [30–33] analysis. Furthermore, comparing the present results with those obtained on the basis of approaches which explicitly take into account the effects of phonon dressing, we are able to determine the regions of parameter space (the (S, B) plane) in which a particular type of system behaviour should occur. These results are given in figure 1. In this way, knowledge of the

values of system parameters S and B enables one to predict the character of the system dynamics and to understand the mechanisms and conditions which lead to the breakdown of quantum coherence in a simple TLS.

Finally, concerning the relevance of the present concept to the understanding of the dynamics of some realistic systems, we believe that the above analysis could be connected to small-polaron and related problems such as defect motion in solids [41], stabilization of optical isomers embedded in crystals [42], energy transfer in the condensed phase [4] and self-trapping in the non-linear dimer [30,31]. If we bear in mind the possibility of tuning the system parameters by some external action (pressure, for example) [43], it could be possible to observe experimentally the transition from the free to the self-trapped state in order to check the predicted behaviour.

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