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Variational calculation of the dynamics of a two level system interacting with a bath

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A variational calculation of the dynamics of a two level system interacting with a bath is presented. The effective tunneling matrix element is renormalized by the interaction with the bath. For the case of a bath with ohmic dissipation (or infrared divergence), the variational calculation gives a vanishing tunneling at $T = 0$ for critical values of the coupling and gives a highly unusual temperature dependence for the tunneling rate for large kT, all of which agree with recent path integral calculations. The present method also yields results for intermediate temperatures and coupling. This indicates that a judicious choice of the zeroth order Hamiltonian to include the major part of the coupling can lead to correct results even in low order perturbation theory.

Recently, there has been considerable interest in the study of the dynamics of a tunneling system coupled to a harmonic bath. This rather simple system has been used as a model for the effect of the environment on a number of interesting chemical[1] and physical[2] systems. The model we examine is found by representing the tunneling system by a two level system, so that the Hamiltonian can be written as ($h = 1$):

$$H = K\sigma_x + \sum_l \omega_l(a_l^+ a_l + 1/2) + \sigma_z \sum_l g_l(a_l + a_l^+), \tag{1}$$

where a_l and a_l^+ are boson operators and $(1/2)\sigma_x$ and $(1/2)\sigma_z$ are the usual Pauli matrices. In this Hamiltonian, K represents the bare tunneling matrix element and g the coupling to the l th bath mode. In the limit that $K = 0$, the system is just a collection of oscillators, displaced in one direction when the system is one of the two levels and displaced in the other direction when the system is in the other of the two levels. Thus there is a twofold degenerate localized ground state with energy $E = -\Sigma_l g_l^2 \omega_l^{-1}$. In the opposite limit that $g_l = 0$, the eigenstates of the system are the symmetric and antisymmetric combinations (delocalized states) of the spin states with energies $E = \pm K$. Thus, this system exhibits a competition between the localization inherent in the interaction with the bath and the delocalization inherent in the tunneling. In the intermediate regime, the effect of the bath is to modify the tunneling matrix element and damp the oscillations.

In a recent paper, Harris and Silbey,[1] examined the rate of crossing from one localized state to the other in the weak coupling limit. At zero coupling to the bath, the system oscillates between the two states with a frequency $2K$. As the coupling increases, they found that the system could be represented as a damped oscillator whose probability $P(t)$ for being in the initially populated level obeys the oscillator equation:

$$\ddot{P}(t) + \lambda \dot{P}(t) + (2K)^2 P(t) = 0, \tag{2}$$

where[3]

$$\lambda = 2\pi \sum_l g_l^2 \coth(\beta\omega_l/2)\ \delta(2K - \omega_l). \tag{3}$$

In this approximation, for $4K > \lambda$, $P(t)$ oscillates with frequency $[(2K)^2 - (\lambda/2)^2]^{1/2}$; for $4K < \lambda$, $P(t)$ is damped with lifetime $(4K^2/\lambda)$. Although this calculation shows the competition between friction effects and tunneling, it is not flexible enough to show the complete renormalization of the tunneling frequency as predicted by Bray and Moore.[2]

Indeed, Bray and Moore and others[2] have predicted that as the coupling to the bath increases, for a model with ohmic dissipation, the renormalized tunneling rate goes to zero, even at $T = 0$. This is a surprising result at first, because one expects that, although a two level system coupled, for e.g., to a phonon bath will tunnel more slowly as the coupling increases, the rate will not go to zero (at $T = 0$) except for infinitely strong coupling. However, as pointed out by Chakravarty and Kivelson,[2] assuming ohmic dissipation leads to an infrared divergence and the pathology associated with it.[4] It turns out that this leads to the localization phenomenon.

In this paper, we examine a simple variational model for this system, used earlier for the coupling of excitons to phonons,[5,6] which exhibits the central features of the renormalization of the tunneling rate and the competition between tunneling kinetic energy and the localization potential energy. In addition, this model shows clearly the differences between the pathological cases (associated with the infrared divergence) and the nonpathological cases associated with a more exoteric environment.

The Hamiltonian is transformed using a unitary transformation

$$U = \exp\left\{-\sigma_z \sum_l \omega_l^{-1} f_l(a_l - a_l^+)\right\}. \tag{4}$$

For $K = 0, f_l = g_l$ diagonalizes H. For $K \neq 0$:

$$\begin{aligned}\tilde{H} &= UHU \\ &= \tilde{K}\sigma_z + \sum_l \omega_l(a_l^+ a_l + 1/2) + \sum_l (f_l^2 - 2f_l g_l)\omega_l^{-1} \\ &\quad + V_+\sigma_+ + V_-\sigma_- + V_0\sigma_z,\end{aligned} \tag{5}$$

where the renormalized tunneling matrix element is given by

$$\widetilde{K}=K\exp\left\{-2\sum_l f_l^2\omega_l^{-2}\coth(\beta\omega_l/2)\right\}\equiv K\exp(-2F) \tag{6a}$$

and the perturbation terms by

$$V_+=V_-^*=K\exp\left\{-2\sum_l f_l\omega_l^{-1}(a_l-a_l^+)\right\}-\widetilde{K}, \tag{6b}$$

$$V_0=\sum_l(g_l-f_l)(a_l+a_l^+). \tag{6c}$$

Since we have constructed V_i so that $\langle V_i\rangle=0$, where the average is over the bath, Bogoliubov-Peierls upper bound on the free energy is[7]

$$A_B=A_{\text{Phon}}-\beta^{-1}\ln\{2\cosh(\beta\widetilde{K})\}+\sum_l\{f_l^2-2f_lg_l\}\omega_l^{-1}. \tag{7}$$

This upper bound is optimized by varying the $\{f_l\}$ to find

$$f_l=g_l\{1+2\widetilde{K}\omega_l^{-1}\coth(\beta\omega_l/2)\tanh\beta\widetilde{K}\}^{-1}, \tag{8}$$

which is an implicit equation for the $\{f_l\}$.

The effective tunneling matrix element $\widetilde{K}$ has a relatively simple behavior as a function of temperature and coupling to the bath as long as F does not become infinite. However, in the case considered by Bray and Moore and others,[2] they assume that $J(\omega)\equiv\pi\Sigma_l g_l^2\delta(\omega-\omega_l)=\eta\omega$, where η is a friction constant; thus,

$$\sum_l g_l^2\omega_l^{-2}\coth\beta\omega_l/2=\frac{1}{\pi}\int_0^{\omega_c}d\omega\,\frac{J(\omega)}{\omega^2}\coth(\beta\omega/2)=\frac{\eta}{\pi}\int_0^{\omega_c}d\omega\,\frac{\coth(\beta\omega/2)}{\omega}, \tag{9}$$

which exhibits an infrared divergence (here ω_c is the upper limit of the bath frequencies). Thus if $f_l=g_l$ for all l, F is infinite and $\widetilde{K}$ zero. It is the assumption that $J(\omega)=\eta\omega$ which leads to the divergence; for a three-dimensional phonon bath $J(\omega)\sim\omega^3$ so that F remains finite, and increases with temperature in the normal way of a Debye–Waller factor.

To examine the effect of the infrared divergence in this variational calculation, we look first at $T=0$. In this case, we find a self-consistent equation for $\widetilde{K}$ by substituting Eq. (8) into Eq. (6a):

$$\widetilde{K}=K(1+\omega_c/2\widetilde{K})^{-2\eta/\pi}\exp\left\{\frac{2\eta}{\pi}(1+2\widetilde{K}/\omega_c)^{-1}\right\}. \tag{10}$$

For $K/\omega_c<1$, we find (our parameter $2\eta/\pi$ is equal to α of Chakravarty and Leggett[2])

$$\widetilde{K}(T=0)=K(2K/\omega_c)^{\frac{2\eta}{\pi}\left(1-\frac{2\eta}{\pi}\right)^{-1}},\quad 2\eta/\pi<1. \tag{11}$$

Thus as $2\eta/\pi\to1$, $\widetilde{K}(T=0)\to0$ as predicted by earlier workers.[2] Note however that if $K/\omega_c>1$, this does not occur. For example, in the limit of an adiabatic bath (where $\omega_c\to0$), we see from Eq. (10) that either $\widetilde{K}=K$ (i.e., $f_l=0$) or $\widetilde{K}=0$ (i.e., $f_l=g_l$). If $f_l=0$ for all l, then the third term on the right-hand side of Eq. (7), the potential energy of interaction between the tunneling system and the bath, is 0; if $f_l=g_l$ then this term equals $-\eta\omega_c$ when $J(\omega)=\eta\omega$. If we assume that this term vanishes as ω_c approaches 0, then the lowest energy solution of the variational equation is $\widetilde{K}=K$ (delocalized states) for all η; however, if we assume that this term $(-\eta\omega_c)$ remains finite as ω_c approaches 0, then there is a transition from delocalized states $(\widetilde{K}\neq0)$ to localized states $(\widetilde{K}=0)$ as η increases.

For higher temperatures, but still low compared to ω_c, the self-consistent equation for $\widetilde{K}(T)$ is

$$\widetilde{K}=Ke^{-2\eta/\beta\widetilde{K}}\left[\frac{2+2\beta\widetilde{K}\tanh\beta\widetilde{K}}{\beta\omega_c+2\beta\widetilde{K}\tanh\beta\widetilde{K}}\right]^{2\eta/\pi}, \tag{12}$$

which also agrees with Eq. (10) as $T\to0$. Equation (12) yields for $\beta\widetilde{K}\ll1$ and $\eta<K/kT$,

$$\widetilde{K}=K(2kT/\omega_c)^{2\eta/\pi}, \tag{13}$$

which is very similar to Bray and Moore [their Eq. (13)]. For $\eta>K/kT$ our calculation predicts that $\widetilde{K}=0$, and a second order calculation for the tunneling rate or relaxation must be done; we present this below.

The probability of the system remaining in one state $P(t)$ again obeys an equation similar to Eq. (2), except that K is replaced by $\widetilde{K}$ and λ by $\widetilde{\lambda}$ (determined now by the spectrum of the V_i). At $T=0$ and η small, $P(t)$ oscillates with frequency $[(2\widetilde{K})^2-(\widetilde{\lambda}/2)^2]^{1/2}$ and is damped with a rate $\widetilde{\lambda}/2$. For this case, $\widetilde{\lambda}=4\widetilde{K}\eta$ [8] so that the oscillation frequency is $2\widetilde{K}[1-\eta^2]^{1/2}$. Note that the ratio of the frequency to decay rate is $[1-\eta^2]^{1/2}/\eta$ which agrees to lowest order for the small η limit of Chakravary and Leggett.[2] For $\eta>1$, $P(t)$ has no oscillations, but is damped with rate $-[(\widetilde{\lambda}/2)^2-(2\widetilde{K})^2]+(\widetilde{\lambda}/2)$. For η larger than $\pi/2$, $\widetilde{K}=0$ and localization occurs.

For large (kT/K) and $\eta\ll K/kT$, we find $\widetilde{\lambda}=ckT$, with c an unknown constant, so that $P(t)$ is overdamped with decay rate

$$(2\widetilde{K})^2/\widetilde{\lambda}=\{(2K)^2/c\omega_c\}(2kT/\omega_c)^{(4\eta/\pi)-1}. \tag{14}$$

For large kT/K and $\eta>K/kT$, the tunneling is completely incoherent since $\widetilde{K}=0$. The probability of tunneling (or the relaxation of $P(t)$) is then given by $W=\int_0^\infty dt\,\langle V_{12}(t)V_{21}\rangle dt$, the Golden Rule expression, where V_{12} is given in Eq. (6b) with $f_l=g_l$ and $\widetilde{K}=0$. Because of the infrared divergence, great care must be taken with the integral; however it has been worked out for the x-ray singularity case.[4] We find for $\beta\omega_c\gg1$ and $\beta K\ll1$,

$$W=\frac{K^2}{\omega_c}\left[\frac{2\pi kT}{\omega_c}\right]^{(4\eta/\pi)-1}\frac{\pi\cos2\eta}{2\eta}e^{-(2.28)\eta/\pi}. \tag{15}$$

These conclusions agree with those of Chakravarty and Leggett[2] in almost all details. [Note however that their calculation predicts that the oscillations of $P(t)$ disappear at $\eta=\pi/4$ at $T=0$ rather than at $\eta=1$.] These results indicate that the present method, based on a simple variational calculation contains enough flexibility to handle correctly the competition between the localization and delocalization effects in this problem and suggests that a judicious transformation of the Hamiltonian to pick the best H_0 can lead to good results using only low order perturbation theory. In addition, this method is flexible to handle even the pathological cases associated with the infrared divergent terms.[9,10]

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[3]Our g_l is equal to $(h/2m_l\omega_l)^{1/2}c_l$ of Harris and Silbey (Ref. 1).

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[8]In Harris and Silbey (Ref. 1), q_0 in Eqs. (30) and (31) should be replaced by $2q_0^2$; the right-hand side of Eq. (36) should be multiplied by 4 and the right-hand side of Eqs. (34) and (37) by 2.

[9]A numerical path integral calculation of a two level system interacting with a phonon bath (Ref. 10) showed similar behavior to the infrared divergent case. However these authors looked at a one dimensional phonon bath with coupling constant at low frequency $g_l = (C\omega_l)^{1/2}$ which also exhibits the infrared divergence. This should be absent in the three-dimensional calculation.

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