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Variational calculation of the tunneling system interacting with a heat bath. II. Dynamics of an asymmetric tunneling system

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The dynamics of tunneling of an asymmetric double well interacting with a heat bath is reexamined in the binary collision dilute gas phase limit and investigated in the harmonic bath case. (a) In both cases dynamic and static asymmetries are additive. (b) In both cases, when the tunneling amplitude is not renormalized to a value of zero, the effect of asymmetry on the approach to equilibrium due to incoherent tunneling only quantitatively differs from the symmetric double well. (c) When the renormalized tunneling amplitude is zero, the asymmetric tunneling turns dephasing into population relaxation. (d) Variational methods again give results in agreement with renormalization group and instanton calculations.

I. INTRODUCTION

Recently we discussed the dynamics of tunneling in a double well.[1] When the medium was an ideal gas of harmonic oscillators, our theory showed that the double well dynamics was well described by a set of renormalized Bloch equations. The theory was in agreement with various path integral, instanton, and renormalization group treatments of the same problem.[2]

Although our work was explicitly planned to deal with a "weakly" asymmetric double well, our theory was developed for the symmetric double well. It therefore behooves us, both because of our earlier statement, and because of the intrinsic interest in asymmetric double wells, to generalize our previous theory to include asymmetric tunneling.

In the preceding paragraph we used the phrase "weak asymmetry." What we mean by this term is that the asymmetry is small compared to the natural excitation frequency of either well. Thus, at low temperature, we may treat the double well as a two-state system, rather than a spin system with more than two components.

In order to set the stage, in the next section we briefly review and criticize an earlier theory of gas phase tunneling in a weakly asymmetric double well.[3] We then turn to the main problem, asymmetric tunneling in the presence of a harmonic bath at finite temperature, and, in particular, the case of an "ohmic" density of oscillator states.[2(b)] In both these sections we examine the roles played by static and dynamic asymmetries, and show how they are essentially additive in effect. In the following section of this paper we discuss the physical nature of our results by looking at our theory at zero temperature. Finally, we conclude by discussing the effect of asymmetry on low temperature tunneling in general.

II. TUNNELING IN THE GAS PHASE

In this section we review and criticize earlier work on gas phase tunneling.[1(a),3] The fundamental assumptions of this theory, aside from the two state model of the double well, were that the duration of a collision was rapid compared with the tunneling frequency K and static asymmetry frequency Δ. In addition, it was assumed that the rotations of the tunneling molecule were equilibrated after each collision. No persistence of rotations was allowed. According to this theory, the change in the overall density matrix by a collision was given by

$$\rho(t) = S^{+}\rho(t)S - 1\,, \tag{2.1}$$

where S is the S matrix for the energy shell collisions, and was given by

$$S = S_L|L\rangle\langle L| + S_R|R\rangle\langle R|\,. \tag{2.2}$$

The S matrix acts separately in the left and right wells. By "on the energy shell," we meant only that the collisions with left- and right-handed molecules conserved energy. This is a crucial conceptual point to which we will return in a moment. The above assumptions along with the usual assumption of a uniform rate of collision, or invocation of generalized Ehrenfest theorems,[4] is sufficient to derive Bloch equations for the averaged Pauli spin operators. These equations are

$$\dot{P} = \mathbf{V}\times\mathbf{P} - \lambda'(\mathbf{P}_x\hat{x} + \mathbf{P}_y\hat{y})\,, \tag{2.3}$$

where $P_i(t)$ are given by the following equations:

$$P_x(t) = \mathrm{Tr}\,\rho(t)[|L\rangle\langle R| + |R\rangle\langle L|] \equiv \mathrm{Tr}\,\rho(t)\sigma_x\,, \tag{2.4a}$$

$$P_y(t) = i\,\mathrm{Tr}\,\rho(t)[|L\rangle\langle R| - |R\rangle\langle L|] \equiv \mathrm{Tr}\,\rho(t)\sigma_y\,, \tag{2.4b}$$

$$P_z(t) = \mathrm{Tr}\,\rho(t)[|L\rangle\langle R| - |R\rangle\langle L|] = \mathrm{Tr}\,\rho(t)\sigma_z\,. \tag{2.4c}$$

V is given by

$$V = 2K\hat{x} + [2\Delta + \lambda'']\hat{z}\,. \tag{2.4d}$$

K is the natural tunneling frequency, Δ is the static asymmetry, and $\lambda = \lambda' + i\lambda''$ is the damping parameter given essentially by

$$\lambda = \sum_i w(i)\frac{nv_i\pi}{k_i^2}\sum_j\sum_f\frac{(2J+1)}{(2J_i+1)(2J_f+1)} \times [1 - (S_{if}^{J})^{+}S_{if}^{J}]\,, \tag{2.5}$$

where i and f refer to all initial and final states of the two molecules, including relative momentum, but excluding angular momentum J. It may be shown that λ is real if the medium is parity symmetric. Thus, the imaginary part of λ is a direct measure of medium asymmetry and is manifested exactly as an additional free motion. It is clear, therefore, that this theory predicts that there is no way of distinguishing static from dynamically induced asymmetry given one set of medium conditions. Of course, if one is able to look at the tunneling *in vacuo*, there is an absolute measure of static versus dynamic asymmetry. A similar result obtains when the tunneling particle interacts with a harmonic solid (see below).

Of much greater importance is the homogeneous nature of the Bloch equation. The population of left and right become equal as time goes to infinity; thus the equations predict an infinite ratio of temperature to static splitting and tunneling frequency. A concomitant of the fact that the theory predicts a set of homogeneous equations is that no population difference between left and right may arise at any time if the tunneling density matrix is completely incoherent, that is the unit matrix. This result is, of course, at variance with the exact solutions of the equations of motion if asymmetries are statically or dynamically applied after $t=0$. The lack of inhomogeneous terms in the Bloch equations is a result of the impact nature of the S matrix which feel the left- and right-handed molecules only in a superposition sense. No energy is allowed to be exchanged between left and right through collisions so true equilibrium can never arise. Hence what we meant by the S matrix being on the energy shell is just that it conserves the energy of the collisions for left- and right-handed molecules separately. We may add that the correct conservation of energy is sufficient to allow the correct equilibrium even when the coupling between left and right is neglected in the S matrix after energy conservation is explicitly accounted for. The point will be made manifest in the next section where the exact weak coupling Bloch equation is used.

Assuming our theory is correct, that is, the term $(K^2+\Delta^2)/k_BT$ is very small, we may now examine how the approach to equilibrium is modified by an asymmetry. To do this we write a single cubic equation for the difference between left and right population, namely

$$\dddot{P}_z + 2\lambda'\ddot{P}_z + (K^2+\Delta^2+\lambda''^2+\lambda'^2)\dot{P}_z + \lambda' K^2 P_z = 0\,. \tag{2.6}$$

It is clear that the presence of $\Delta^2+\lambda''^2$ changes the onset of the critical damping; however we are interested in overdamping. Once the system is well overdamped, we have

$$\lambda'^2 \gg \Delta^2 + K^2 + \lambda''^2 \tag{2.7}$$

so that Eq. (7) reduces to

$$\lambda'^2\left(1+\frac{\Delta^2+K^2+\lambda''^2}{\lambda'^2}\right)\dot{P}_z + \lambda' K^2 P_z = 0\,, \tag{2.8}$$

whose solution is

$$P_z(t) = P_z(0)\exp\{-K^2 t/\lambda' \times [1+(\Delta^2+K^2+\lambda''^2)/\lambda'^2]\}\,. \tag{2.9}$$

We see that the rate of approach to complete incoherence is only quantitatively affected by the asymmetry. Indeed, even if $\Delta \simeq \lambda'$ the rate would only be lowered by a factor of 2. These results are very similar to those found in the renormalized weak coupling limit presented in the next section. The reason that asymmetry plays only a quantitative rather than a qualitative role is that the competition between fluctuations in localization and tunneling are what gives rise to the incoherence. Weak asymmetry only rotates the axis where the competition is played out. In a sense this is what happens in the renormalization of the tunneling frequency itself.

III. HAMILTONIAN OF A TWO-LEVEL SYSTEM INTERACTING WITH A HARMONIC BATH

We now consider a condensed medium, represented by a large number of independent harmonic oscillators, coupled linearly to our two-level system. This the famous "spin boson Hamiltonian," ubiquitous in condensed matter physics. H is given by

$$H = \Delta\{|L\rangle\langle L| - |R\rangle\langle R|\} + K\{|L\rangle\langle R| + |R\rangle\langle L|\} + (1/2)\sum_j\{P_j^2+\omega_j^2 Q_j^2\} + \sum_i (g_i^L|L\rangle\langle L| + g_j^R|R\rangle\langle R|)Q_j\,. \tag{3.1}$$

The inequivalence of $|L\rangle$ and $|R\rangle$ appear in two places. The static term Δ is identical to that which appeared in Sec. II. The fact that the coupling between well and bath is no longer even under inversion (i.e., $g_j^L \neq -g_j^R$) represents the dynamic aspect of the inequivalence. However, the linear coupling allows the distinction between static and dynamic asymmetry to be removed entirely.

To exhibit this, we simply rewrite the coupling term in the Hamiltonian

$$\sum_j Q_j[g_j^L|L\rangle\langle L| + g_j^R|R\rangle\langle R|] = 1/2\sum_j (g_j^R+g_j^L)Q_j[|L\rangle\langle L| + |R\rangle\langle R|] + 1/2\sum_j (g_j^L-g_j^R)Q_j(|L\rangle\langle L| - |R\rangle\langle R|)\,. \tag{3.2}$$

The first term, symmetric in the two sites, can be removed by shifting the equilibrium position of the Q_j,

$$Q_j^{\text{new}} = Q_j + (1/2)(g_j^L+g_j^R)/\omega_j^2\,. \tag{3.3}$$

This shifts all energies by a constant amount, and leaves a final Hamiltonian like that of Eq. (3.1) except that the new static splitting is

$$\Delta^{\text{new}} = \Delta + \frac{1}{2}\sum_j [(g_j^R)^2 - (g_j^L)^2]/\omega_j^2 \tag{3.4}$$

and the new dynamic coupling constant is

$$g_j^{\text{new}} = \tfrac{1}{2}(g_j^L - g_L^R)\,. \tag{3.5}$$

Thus

$$H^{\text{new}} = \Delta^{\text{new}}(|L\rangle\langle L| - |R\rangle\langle R|) + K(|L\rangle\langle R| + |R\rangle\langle L|) + (1/2)\sum_j (P_j^2 + \omega_j^2 Q_j^2) + \sum_j g_j^{\text{new}} Q_j(|L\rangle\langle L| - |R\rangle\langle R|) . \tag{3.6}$$

Thus, an asymmetric coupling to the bath modes ($g_j^L \neq -g_j^R$) can be made into a symmetric coupling at the cost of changing the static energy asymmetry.

We shall use this Hamiltonian [Eq. (3.6)] as the starting point in our description of the tunneling dynamics in a low temperature harmonic bath, but will deplete the superscript "new".

IV. THE VARIATIONAL RENORMALIZED TUNNELING FREQUENCY

As we mentioned in the Introduction, we must proceed in two distinct steps if we wish to determine the dynamics of our tunneling system. In this section we carry out the first step which is the minimalization of the Helmoltz free energy A by a unitary transformation. Since the method is identical to that used in our previous paper (and many other places) our presentation will be brief.

We perform the unitary transformation on H using the operator U, that is, we write

$$\tilde{H} = U^+ H U , \tag{4.1}$$

where U is the unitary operator which shifts the origins of the oscillator, namely,

$$U = \exp\left\{ i \sum_j f_j/\omega^2{}_j \, P_j [|L\rangle\langle L| - |R\rangle\langle R|] \right. . \tag{4.2}$$

The set $\{f_i\}$ are determined variationally by finding an upper, or Bogoliubov, bound on A. The resulting $\tilde{H}$ is given by

$$\tilde{H} = \Delta\,[|L\rangle\langle L| - |R\rangle\langle R|] + K\chi^+|R\rangle\langle L| + K\chi|L\rangle\langle R| + \frac{1}{2}\sum_j (P_j^2 + \omega_j^2 Q_j^2) - S + \sum_j (g_j - f_j) \times Q_j[|L\rangle\langle L| - |R\rangle\langle R|] , \tag{4.3}$$

where

$$\chi = \exp \sum_j (2if_j \, P_j/\omega_j^2) ; \tag{4.4a}$$

and

$$S = \frac{1}{2}\sum_j (f_j^2 - 2f_j \, g_j)/\omega_j^2 . \tag{4.4b}$$

To make the Bogoliubov upper bound[5] on A as straightforward as possible, we add and subtract the thermal average of χ over the bath to χ so that

$$\tilde{H} = \Delta\,[|L\rangle\langle L| - |R\rangle\langle R|] + \tilde{K}\,[|L\rangle\langle R| + |R\rangle\langle L|] + H_{\text{bath}} - S + (K\chi - \tilde{K})|L\rangle\langle R| + (K\chi^+ - \tilde{K})|R\rangle\langle L| + \sum_i (g_i - f_i)Q_i[|L\rangle\langle L| - |R\rangle\langle R|] \equiv \tilde{H}_0 + \tilde{V} . \tag{4.5}$$

Here

$$\tilde{K} = K \exp - \sum_j (f_j^2/\omega_j^3)\coth(\beta\omega_j/2) ,$$

and $\langle \tilde{V}\rangle_{\text{bath}} = 0$. Now the Bogoliubov bound on A is (since $\langle V\rangle = 0$),

$$A_B = -\beta^{-1} \ln \operatorname{Tr} e^{-\beta\tilde{H}_0} \geqslant A , \tag{4.6}$$

where the trace is over the complete set of states of the combined system. We find, neglecting the free energy of the bath modes which is unchanged by the interaction with the tunneling system,

$$A_B = -\beta^{-1} \ln 2\cosh\beta(\Delta^2 + \tilde{K}^2)^{1/2} - S . \tag{4.7}$$

Minimizing with respect to f yields

$$f_i = g_i \left\{ 1 + \frac{2\tilde{K}^2 \coth(\beta\omega_i/2)\tanh\beta(\Delta^2 + \tilde{K}^2)^{1/2}}{\omega_i(\Delta^2 + \tilde{K}^2)^{1/2}} \right\}^{-1} \tag{4.8}$$

agreeing with our earlier result when $\Delta = 0$.

In the limit that T is large compared to ω_i, which is true for the low frequency modes even at low temperatures, we see that

$$f_i = g_i/\{1 + 4\tilde{K}^2 \tanh\beta\Delta/\beta\Delta\omega_i^2\} , \tag{4.9}$$

where we have assumed that $\Delta \gg \tilde{K}$. For the case that $T \gg \Delta$ also, we find

$$f_i = g_i/[1 + 4\tilde{K}^2/\omega_i^2] \tag{4.10}$$

and the self-consistent equation for $\tilde{K}$ is

$$\tilde{K} = K \exp - \sum_j (f_j^2/\omega_j^3)\coth(\beta\omega_j/2) = K \exp - \frac{1}{\pi}\int_0^{\omega_c} d\omega \, I(\omega)\coth(\beta\omega)/2)[\omega + \tilde{4}K^2/\omega]^{-2} \cong K \exp - \frac{2}{\beta\pi}\int_0^{\omega_c} d\omega \, I(\omega)\frac{\omega}{[\omega^2 + 4\tilde{K}^2]^{-2}} , \tag{4.11}$$

where

$$I(\omega) = \pi \sum_j (g_j^2/\omega_j)\delta(\omega - \omega_j) .$$

For the infrared divergent case, $I(\omega) = \eta\omega e^{-\omega/\omega_c}$, and we find $\tilde{K} = 0$ for all such temperatures.

For the case $T \ll \Delta$ we find

$$f_i = g_i[1 + 4\tilde{K}^2 T/\omega_i^2\Delta]^{-1} \tag{4.12}$$

so that

$$\tilde{K} = K \exp - \frac{2}{\beta}\int d\omega \, I(\omega)\{\omega[\omega^2 + 4\tilde{K}^2 T/\Delta]^{-2}\} . \tag{4.13}$$

Again for the infrared divergent case, $\tilde{K} = 0$ for all but the lowest temperatures. Note that for finite T, even $I(\omega) \sim \omega^2$ at low temperature can lead to an infrared divergence (because of the $\coth\beta\omega/2$ term) so that $\tilde{K} = 0$ is a possibility in this case also. For the general case of $I(\omega)$ leading to a nondivergent integral $\tilde{K} = 0$ and $\tilde{K}$ is a function of T.

V. DYNAMICS

We now turn to the dynamics and relaxation of the two level system interacting with the harmonic bath. In the pre-

vious section, we have found that a good zeroth order Hamiltonian can be constructed by performing a variational unitary transformation on the original H. After choosing the f to minimize the free energy, we have a new Hamiltonian as given by Eq. (4.5) which has a static (noninteracting) part, $\tilde{H}_0$, and a fluctuating part $\tilde{V}$:

$$\tilde{H}_0 = \Delta\,[|L\rangle\langle L| - |R\rangle\langle R|] + \tilde{K}\,[|L\rangle\langle R| + |R\rangle\langle L|] - S + H_{\text{bath}}\,, \quad (5.1a)$$

$$\tilde{V} = (K\chi - \tilde{K})|L\rangle\langle R| + (K\chi^{+} - \tilde{K})|R\rangle\langle L| + \sum_j (g_i - f_i)Q_i\,[|L\rangle\langle L| - |R\rangle\langle R|]\,, \quad (5.1b)$$

or

$$\tilde{V} \equiv V_{RL}|R\rangle\langle L| + V_{LR}|L\rangle\langle R| + V_{LL}|L\rangle\langle L| + V_{RR}|R\rangle\langle R| \quad (5.2)$$

in an obvious notation.

In the basis of the eigenstates of $\tilde{H}_0$, we may write

$$\tilde{H}_0 = (\Delta^2 + \tilde{K}^2)^{1/2}[|a\rangle\langle a| - |b\rangle\langle b|] - S + H_{\text{bath}} \quad (5.3a)$$

$$= \Omega\,[|a\rangle\langle a| - |b\rangle\langle b|] + H_{\text{bath}} - S \quad (5.3b)$$

and

$$\tilde{V} = V_{aa}|a\rangle\langle a| + V_{bb}|b\rangle\langle b| + V_{ab}|a\rangle\langle b| + V_{ba}|b\rangle\langle a|\,, \quad (5.4)$$

where

$$V_{aa} = \left(\frac{1+\cos 2\theta}{2}\right)V_{LL} + \left(\frac{1-\cos 2\theta}{2}\right)V_{RR} + \frac{\sin 2\theta}{2}(V_{LR} + V_{RL})\,, \quad (5.5a)$$

$$V_{bb} = \left(\frac{1-\cos 2\theta}{2}\right)V_{LL} + \left(\frac{1+\cos 2\theta}{2}\right)V_{RR} - \frac{\sin 2\theta}{2}(V_{LR} + V_{RL})\,, \quad (5.5b)$$

$$V_{ab} = V_{ba}^{+} = -\frac{\sin 2\theta}{2}(V_{LL} + V_{RR}) + \left(\frac{1+\cos 2\theta}{2}\right)V_{LR} - \left(\frac{1-\cos 2\theta}{2}\right)V_{RL}\,, \quad (5.5c)$$

where $\tan 2\theta = \tilde{K}/\Delta$. In this basis, we may calculate the relaxation of the density matrix elements in the usual weak coupling or Redfield[6] approximations, since we have, by variational construction chosen a perturbation $\tilde{V}$ which is assumed small. If this has not worked, i.e., the perturbation is still not weak, we must go beyond weak coupling. However, since the unitary transformations we have chosen is exact in the limit that $g_i/K = 0$ or ∞, and interpolates between these limits giving an upper bound on the free energy, we expect that weak coupling will be sufficient.

The dynamics and relaxation of the two level system are best discussed by considering the equations of motion of the reduced density matrix σ,

$$\sigma(t) = \text{Tr}_{\text{bath}}\,\rho(t)\,. \quad (5.6)$$

We will assume that the initial state of the system is such that the bath and the two level system are uncorrelated and that the bath is in Boltzmann equilibrium:

$$\rho(0) = \sigma(0)e^{-\beta H_{\text{bath}}}\,. \quad (5.7)$$

The standard techniques then yield, in the *eigenstates* of $\tilde{H}_0$ representation:

$$\dot{\sigma}_{nm} = -i\omega_{nm}\sigma_{nm}(t) - \sum_{pq} R_{nm,pq}\sigma_{pq}(t)\,, \quad (5.8)$$

where the elements of the relaxation matrix R are given by the standard formulas.[6]

Since the trace of $\sigma(t)$ is unity, independent of time, the equations of motion for $\sigma_{nm}(t)$ can be written as three equations in the population difference, $\sigma_{aa}(t) - \sigma_{bb}(t)$, and the coherences, $\sigma_{ab}(t) + \sigma_{ba}(t)$, $\sigma_{ab}(t) - \sigma_{ba}(t)$. Because the matrix elements obey detailed balance,[6] these equations automatically have the correct equilibrium solutions. We note in passing that, in general, the equations of motion can be rewritten as a third order differential equation in any single variable as in Eq. (2.6).

We are concerned with the decay of the difference between $|L\rangle$ and $|R\rangle$ population or $P_z(t) = \sigma_{LL}(t) - \sigma_{RR}(t)$. Note however that in the eigenstate representation,

$$P_z(t) = (\cos 2\theta)[\sigma_{aa}(t) - \sigma_{bb}(t)] - (\sin 2\theta)[\sigma_{ab}(t) + \sigma_{ba}(t)] \quad (5.9)$$

so that the decay of $P_z(t)$ is determined in general by *both* the decay of the populations, $\sigma_{aa} - \sigma_{bb}$, and the coherence, $\sigma_{ab} + \sigma_{ba}$, in the eigenstate representation. In the limit that $\tilde{K}/\Delta \gg 1$, $P_z(t)$ will be the coherence in the eigenstate representation (i.e., $\sigma_{ab} + \sigma_{ba}$). In the opposite limit that $\Delta/\tilde{K} \gg 1$, $P_z(t)$ will be given by the population in the eigenstate representation.

Instead of discussing the general case, we will be concerned only with these limiting cases.

(a) $\tilde{K}/\Delta \ll 1$. (5.10)

In this case, $f_i \approx g_i$ for (almost) all modes, and $|a\rangle \approx |L\rangle$ and $|b\rangle \approx |R\rangle$. The equation of motion of $P_z(t)$ becomes approximately

$$\dot{P}_z(t) = -(1+A)\Gamma P_z - (1-A)\Gamma\,, \quad (5.11)$$

where $A = \exp(-2\beta\Delta)$. Since $|L\rangle$ has a higher zeroth order energy than $|R\rangle$, the decay rate of P_z is directly related to the *population relaxation* rate which is given (using the standard form for the matrix R) by the Golden rule formula

$$\Gamma = \int_{-\infty}^{+\infty} d\tau\, e^{-i2\Delta\tau}\langle V_{LR}(\tau)V_{RL}(0)\rangle\,. \quad (5.12)$$

Using the form of V_{LR} given above, we find (as in polaron theory[7])

$$\Gamma = \frac{1}{2}K^2\int_{-\infty}^{+\infty} d\tau\{e^{\phi(\tau)-\phi(0)} - e^{-\phi(0)}\}\,e^{-i2\Delta\tau}\,, \quad (5.13)$$

where

$$\phi(\tau) = \frac{4}{\pi}\int_0^{\infty} d\omega\,\frac{I(\omega)}{\omega^2} \times \left[i\sin\omega\tau + \coth\frac{\beta\omega}{2}(\cos\omega\tau - 1)\right]\,, \quad (5.14)$$

$$I(\omega)=\pi\sum_j \frac{g_i^2}{\omega_j}\delta(\omega-\omega_j)\,. \tag{5.15}$$

For the case of an ohmic bath[3] where $I(\omega)=\eta\omega e^{-\omega/\omega_c}$, the integral can be done and we find (for $2\eta/\pi>1$ so that $\tilde{K}=0$)

$$\Gamma=\frac{1}{2}\int_{\infty}^{\infty} d\tau\, e^{-i2\Delta\tau}(1-i\omega_c\tau)^{-4\eta/\pi}\left[\frac{\beta}{\pi\tau}\sinh\frac{\pi\tau}{\beta}\right]^{-4\eta/\pi} \tag{5.16}$$

so that for $\beta\Delta\gg 1$ and $\Delta/\omega_c\ll 1$, we find

$$\Gamma=\frac{K^2}{\omega_c}\frac{\pi}{\gamma(4\eta/\pi)}\left(\frac{2\Delta}{\omega_c}\right)^{(4\eta/\pi)-1}\left[1+\frac{\pi^2}{6}\left(\frac{1}{2\beta\Delta}\right)^2\left(\frac{4\eta}{\pi}\right)\right.$$
$$\left.\times\left(\frac{4\eta}{\pi}-1\right)\left(\frac{4\eta}{\pi}-2\right)+O(\beta^{-4})\right], \tag{5.17}$$

where $\gamma(x)$ is the gamma function of x. This agrees exactly with the result of Fisher and Dorsey found using a path integral method.[8] We want to emphasize that, when looked at in the eigenstate representation, this result can be seen to be simply the normal population relaxation from an upper level ($|L\rangle$) to a lower level ($|R\rangle$) via the Fermi Golden Rule rate. The first, or T independent term, is simply the spontaneous emission, as expressed in Eq. (5.11). In the ohmic dissipation case for $2\eta/\pi>1$, $\tilde{K}=0$ so that any fluctuations in the matrix element connecting $|L\rangle$ and $|R\rangle$ will cause transitions, thus enhancing the rate.

In the limit that $\beta\Delta\ll 1$, the result for Γ reduces to the $\Delta=0$ result, so that $\Gamma\sim(T/\omega_c)^{4\eta/\pi-1}$ in the ohmic case.

(b) $\tilde{K}/\Delta\gg 1$.

In this case $\theta\simeq\pi/4$ so that the eigenstates are those for $\Delta=0$, and so $P_z(t)$ is equal to $\sigma_{ab}+\sigma_{ba}$. The results of our earlier analysis[1] apply.

VI. ZERO TEMPERATURE

In order to illustrate the method presented in Sec. IV further, we briefly discuss the zero temperature case. Consider the lowest two states in the system with the variational wave functions found by displacing all the modes in the bath in one direction on the left and in the opposite direction on the right:

$$|\tilde{L}\,0\rangle=|L\rangle\prod_i \phi_i^{(0)}(Q_i+f_i/\omega^2{}_i)\,, \tag{6.1}$$

and

$$|\tilde{R}\,0\rangle=|R\rangle\prod_i \phi_i^{(0)}(Q_i-f_i/\omega^2{}_i)\,. \tag{6.2}$$

These states are the lowest energy eigenstates of the Hamiltonian (3.6) with $K=0$ and $g_j=f_j$ and are the variational localized states. With this physical picture in mind we may therefore diagonalize the full H (i.e. $g_j\neq f_j, K\neq 0$) in this basis and determine the minimum energy. The matrix elements of H are

$$\langle\tilde{L}\,0|H|\tilde{L}\,0\rangle=\Delta-\sum_i(g_i f_i/\omega^2{}_i-1/2 f_i^2/\omega^2{}_i)\,, \tag{6.3}$$

$$\langle\tilde{R}\,0|H|\tilde{R}\,0\rangle=-\Delta-\sum_i(g_i f_i/\omega^2{}_i-1/2 f_i^2/\omega^2{}_i)\,, \tag{6.4}$$

and most importantly

$$\langle\tilde{L}\,0|H|\tilde{R}\,0\rangle=K\exp-\sum f_j^2/\omega_j^3\equiv\tilde{K}\,. \tag{6.5}$$

The term multiplying K in Eq. (6.5) is the Franck–Condon factor for the variationally shifted oscillator states. Equation (6.5) expresses the fact that the tunneling amplitude is suppressed by the coupling to the bath. The energy of the lowest state of the Hamiltonian is then

$$E=-(\Delta^2+\tilde{K}^2)^{1/2}-\sum g_i f_i/\omega_i^2+1/2\sum f_i^2/\omega_i^2$$
$$\equiv-(\Delta^2+\tilde{K}^2)^{1/2}-S\,. \tag{6.6}$$

Minimizing E with respect to f_i leads to

$$f_i=g_i\{1+2\tilde{K}^2/[\omega_i(\Delta^2+\tilde{K}^2)^{1/2}]\}^{-1} \tag{6.7}$$

(we have assumed $\tilde{K}>0$). Equation (6.7) is the zero temperature limit of Eq. (4.8). The physical picture which emerges from the calculation is that the tunneling matrix element is decreased by a Franck–Condon factor for all the modes in the bath; however, the low frequency modes are not displaced very much and so their contribution to lowering $\tilde{K}$ (i.e., to the Franck–Condon factor) is small. As the coupling to the bath increases, $\tilde{K}$ decreases and more modes are displaced, thus decreasing $\tilde{K}$ further. In three dimensional systems with phonon modes, $\tilde{K}$ finally decreases to the value calculated for $f_j=g_j$, that is, all modes fully displaced. In systems with an infrared divergence, $\tilde{K}\to 0$ at the same finite value of g found in other calculations using path integral methods.

Note that the modes are more likely to be displaced if $\Delta\neq 0$ than if $\Delta=0$. In particular, for $\Delta\gg K$, we find $f_i=g_i$ for almost all the modes. Only those with small frequencies such that

$$\omega_i\ll\tilde{K}^2/\Delta \tag{6.8}$$

are not displaced much. Hence, even a weakly asymmetric double well (but such that $\Delta\gg\tilde{K}$) will have fully displaced modes for almost all frequencies. In the case of phonon modes, this yields a Franck–Condon factor which is independent of Δ and equal to its strong coupling value. The amplitude of the tunneling oscillations will be reduced from

$$K^2/(K^2+\Delta^2)\simeq K^2/\Delta^2 \tag{6.9}$$

to

$$\tilde{K}^2/(\tilde{K}^2+\Delta^2)\simeq\tilde{K}^2/\Delta^2\,, \tag{6.10}$$

i.e., by the Franck–Condon factor of the bath modes. Note in general that the FC factor is zero only if $g\to\infty$.

The possibility that the Franck–Condon factor is exactly zero for $g_i<\infty$ is raised in the infrared divergent case treated by Leggett, Chakravarty, Zwerger, and others.[2] In this case, the strong coupling FC factor is given by

$$\exp-\sum_j g_j^2/\omega_j^3=\exp-\int\frac{d\omega}{\omega^2}\sum\frac{g_i^2}{\omega_i}\delta(\omega-\omega_i)$$
$$=\exp-\frac{1}{\pi}\int\frac{d\omega}{\omega^2}I(\omega)\,. \tag{6.11}$$

But $I(\omega) = \eta\omega e^{-\omega/\omega_c}$ in the IR divergent case, whereupon

$$\exp - (\eta/\pi) \int_0^\infty \frac{d\omega}{\omega} e^{-\omega/\omega_c} = 0 \tag{6.12}$$

so that the tunneling matrix element can become zero due to the low frequency modes. Note that the variational FC factor is given in the IR divergent case by

$$\begin{aligned}\exp - \sum_i f_i^2/\omega_i^3 &= \exp - \frac{1}{\pi} \int d\omega \frac{I(\omega)}{\omega^2} \\ &\quad \times [1 + 2\tilde{K}^2/\omega(\Delta^2 + \tilde{K}^2)^{1/2}]^{-2} \\ &= \exp - (\eta/\pi) \int_0^\infty d\omega\, e^{-\omega/\omega_c} \\ &\quad \times [\omega + 2(\Delta^2 + \tilde{K}^2)^{-1/2}\tilde{K}^2]^{-2} \\ &\simeq [\tilde{K}^2/\omega_c\Delta]^{\eta/\pi},\end{aligned} \tag{6.13}$$

where we have assumed $\omega_c \gg \tilde{K}$, and $\Delta \gg \tilde{K}$. These assumptions lead to the variationally determined self-consistent values of K, namely,

$$\tilde{K} = 0, \tag{6.14}$$

and

$$\tilde{K} = K\,[K^2/\omega_c\Delta]^{\eta/\pi(1-2\eta/\pi)^{-1}}. \tag{6.15}$$

Thus the tunneling amplitude approaches zero as $\eta \to \pi/2$, from below. When $\Delta \ll \tilde{K}$ (or zero) we have the earlier result

$$\tilde{K} = (K/\omega_c)^{2\eta/\pi(1-2\eta/\pi)^{-1}}.$$

Thus, we see that the presence of a large Δ only reduces the value of $\tilde{K}$ for $\eta < \pi/2$, but once the transition point is reached, $\tilde{K} = 0$.

VII. CONCLUSIONS

There are a number of conclusions that may be reached from our study of the influence of a medium on the tunneling in a weakly asymmetric double well. First, static and dynamic asymmetries are additive in their effects, in both the binary collision, dilute gas, and the harmonic oscillator medium cases. Secondly, the role of the asymmetry in the tunneling dynamics is only quantitative at finite temperature as long as one is not in a region where the tunneling amplitude $\tilde{K}$ vanishes. When $\tilde{K}$ is zero, as in the ohmic density of states case, the low temperature tunneling dynamics of the asymmetric well differs qualitatively from that of the symmetric well. In the symmetric case at $T = 0$, the localization is total in the renormalized golden rule limit, but absolutely symmetric. That is, if the system begins in $|L\rangle$ it stays in $|L\rangle$ forever. Similarly, if it begins in $|R\rangle$ it stays in $|R\rangle$ forever. In a sense we must say that localization really is not symmetry breaking. Any incoherent mixture of $|L\rangle$ and $|R\rangle$ could be created. It is just that their mirrors would be equally probable, and stay in the original conditions forever. As T is raised, there will be incoherent tunneling from $|L\rangle$ and $|R\rangle$ and vice versa.

When the wells are *asymmetric*, there is a Golden rule rate which is spontaneous emission (plus induced emission at higher temperatures) for a transition from the higher energy well to the lower energy well, and induced emission backwards, exactly like the Einstein A and B coefficients. Like the symmetric case, $\tilde{K} = 0$ has rendered everything incoherent, but the broken symmetry has created populations which incoherently tunnel into one another via the transformed interaction. At zero temperature, since only spontaneous emission can occur, the higher energy state alone decays, and the lower state is stable forever. Thus, the medium has truly broken the mirror symmetry; for in the absence of the medium, even though the tunneling is asymmetric, the mirror tunneling is identical. At zero temperature in the asymmetric well, when $\tilde{K} = 0$, the medium has truly destroyed the mirror.

Finally, it seems clear that all the instanton type of calculations on the double well tunneling coupled to harmonic baths give the identical results as our simple variational method. The reason is that all the "heavy" work in the instanton calculation has gone into creating an undistorted, renormalized, oscillating, two state system. That is the easy work of the variational methods.

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