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Distance dependence of tunneling in dissipative systems

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We investigate the distance dependence of the rate of tunneling between two sites a distance r apart interacting with a thermal bath of phonons characterized by a density of states of the form $g(\omega)\sim\omega^{d-1}$. We show that in such systems, the correction to the tunneling rate is of the form $\exp[-(r/r_0)^{3-d}]$, where r_0 is a characteristic length. Ohmic dissipation corresponds to $d=1$ and a Gaussian correction $\exp[-(r/r_0)^2]$ to the tunneling rate arises as predicted by one of us [P. Phillips, J. Chem. Phys. **84**, 976 (1986)].

Consider an electron tunneling between two sites a distance r apart in a condensed phase. Phillips[1] has recently pointed out that, if the condensed phase is characterized by *Ohmic* dissipation,[2-5] the distance dependence of the tunneling rate will change from the standard exponential decay $\exp(-r/r_0)$ to $\exp[-(r/r_0)^2]$ at large r/r_0, where r_0 is some characteristic length. It was shown[1] that this change produces a strong qualitative effect on the transport of electrons in Ohmic systems (such as the transport of electrons among localized impurity sites on a metal surface) that should be experimentally observable. In this paper, we discuss the distance dependence of the tunneling rate for a thermal bath of phonons. We show here that, in such systems, the correction to the tunneling rate is of the form $\exp[-(r/r_0)^{3-d}]$, where d is the dimension of the bath. The $d=1$ phonon bath has the characteristic spectral density of an Ohmic dissipative system. Hence, the subsequent dissipation correction $\exp[-(r/r_0)^2]$ agrees with earlier results.[1]

The starting point for our analysis is the Euclidean Lagrangian for an electronic system described by the continuous coordinate q interacting linearly with a dissipative phonon bath:[1-5]

$$\mathcal{L}_E=\tfrac{1}{2}M\dot{q}^2-V(q)+\tfrac{1}{2}\sum_j(\dot{x}_j^2-\omega_jx_j^2)-q\sum_jC_j(q)x_j-q^2\sum_j|C_j(q)|^2/\omega_j^2\ , \tag{1}$$

where $V(q)$ is the potential energy of our electronic system. $V(q)$ will be taken to be some continuous function of q having two degenerate minima at $\pm q_0/2$. In this expression, x_j is the phonon coordinate for the jth phonon of frequency ω_j and $C_j(q)$ is the electron-phonon coupling constant for an electron at q with the jth phonon mode. We have written the phonon coordinates in mass-weighted form. The standard spin-boson Hamiltonian for a two-state system can be obtained from (1) by projecting out the lowest states of $V(q)$. This limit is valid if k_BT is small, relative to the well depths at the minima of $V(q)$, and the "vibrational" level spacing in each well. The effective Euclidean action is obtained by integrating over the phonon degrees of freedom and approximating the q dependence of $C_j(q)$ by a two valued function $C_j(\pm q_0/2)$, valid for a deep double-well potential in the instanton approximation:

$$S_{\rm eff}(q(t))=\int_{-\infty}^{\infty}[\tfrac{1}{2}M\dot{q}^2+V(q)]dt+\tfrac{1}{2}\int_{-\infty}^{\infty}dt'\int_0^{\beta}dt\,\alpha(t-t')[q(t)-q(t')]^2\ , \tag{2}$$

where

$$\alpha(t-t')=\frac{1}{2\pi}\int_0^{\infty}J(\omega)\exp(-\omega|t-t'|)d\omega\ , \tag{3}$$

and the spectral density of the bath

$$J(\omega)=\frac{\pi}{2}\sum|\Delta C_j|^2/\omega_j\delta(\omega-\omega_j)\ , \tag{4}$$

where ΔC_j is $C_j(q_0/2)-C_j(-q_0/2)$. The standard form[2] for the probability per unit time of tunneling between the minima of $V(q)$

$$\Gamma=Be^{-S_{\rm eff}(q_{cl}(t))} \tag{5}$$

involves the effective action evaluated along the least action path $q_{cl}(t)$, which is determined from the equations of motion of (2). The prefactor B is a function of T, in general, and accounts for excursions about the classical path. For Ohmic systems, the standard form yields zero, so that a more careful analysis including corrections to $q_{cl}(t)$ is used.[2]

We can find the distance dependence of Γ from (2)–(5) once we know the distance dependence of $J(\omega)$, which is determined by the ΔC_j. Since we are interested in the low-frequency behavior of $J(\omega)$,[1-5] only the low-frequency delocalized phonons are important, so that[6] $C_j(q)$ is proportional to $\exp(i\mathbf{k}_j\cdot\mathbf{q})$ where $\mathbf{k}_j$ is the phonon wave vector, and

$$\Delta C_j=\lambda q_0^{-1}[\exp(i\mathbf{k}_j\cdot\mathbf{q}_0/2)-\exp(-i\mathbf{k}_j\cdot\mathbf{q}_0/2)]\ , \tag{6}$$

where λ is independent of phonon frequency in the deformation potential approximation.[6] If (6) is now substituted into (4) and (3) we find

$$J(\omega)=\pi\frac{\lambda^2}{q_0^2}\sum_j\sin^2(\mathbf{k}_j\cdot\mathbf{q}_0/2)\omega_j^{-1}\delta(\omega-\omega_j) \tag{7}$$

and

$$\alpha(t-t')=\frac{1}{2}\frac{\lambda^2}{q_0^2}\sum_j \sin^2(\mathbf{k}_j\cdot\mathbf{q}_0/2)\omega_j^{-1}e^{-\omega_j|t-t'|} \ . \quad (8)$$

In the Debye approximation $\omega_j=|\mathbf{k}_j|c$, where c is the speed of sound, and the density of phonon frequencies $\rho(\omega)$ is proportional to ω^{d-1}, with d the dimension of the system. The angular dependence (i.e., $\mathbf{k}_j\cdot\mathbf{q}_0$) in these integrals produces slightly different forms for the final integral depending on dimension; however, the q_0 dependence is essentially the same and is manifested most simply by assuming that $\mathbf{k}_j\cdot\mathbf{q}_0$ can be replaced by $|\mathbf{k}_j|\,|\mathbf{q}_0|$. Then

$$J(\omega)\sim\frac{\lambda^2}{q_0^2}\sin^2(\omega)(q_0/2c)\omega^{d-2} \ , \quad (9)$$

and

$$\alpha(t-t')\sim\int_0^{\omega_D}d\omega\frac{\lambda^2}{q_0^2}\sin^2\left(\frac{\omega q_0}{2c}\right)\omega^{d-2}e^{-\omega|t-t'|} \ , \quad (10)$$

where ω_D is the Debye (or cutoff) frequency for the phonons. Note for small ω, $J(\omega)\sim\omega^d$, so that the one-dimensional phonon bath is an Ohmic system, while higher dimensional phonons are not. By transforming to the variables $x=\omega q_0/c$, $\tau=tc/q_0$, we find

$$\alpha(\tau-\tau')=Aq_0^{1-d}\lambda^2\int_0^{\omega_D q_0/c}dx\,x^{d-2}\sin^2(x/2)e^{-x|\tau-\tau'|} \ . \quad (11)$$

(A is a numerical factor arising from the density of states) or

$$\alpha(\tau-\tau')=q_0^{1-d}\alpha'(\tau-\tau') \quad (12)$$

and for the dissipation part of S_{eff},

$$S_{\text{eff}}^{(\text{diss})}=Aq_0^{3-d}\int_{-\infty}^{\infty}d\tau'\times\int_0^{\beta c/q_0}d\tau\,\alpha'(\tau-\tau')[Q(\tau)-Q(\tau')]^2 \ , \quad (13)$$

where $Q(\tau)=q(q_0\tau/c)/q_0$. Because all the variables are now scaled, we conclude that the general result for the distance dependence of the tunneling rate is for low temperature, i.e., for $\beta c/q_0\to\infty$,

$$\Gamma_{\text{diss}}\propto\exp(-fq_0^{3-d}) \ . \quad (14)$$

For the Ohmic case (one-dimensional phonons) the correction is Gaussian,[1] while for higher dimensional phonons (super Ohmic cases), the correction is weaker.

This behavior can also be found from the spin-boson model[3-5] of this system. The Hamiltonian in this case is

$$H=K\sigma_x+\sum_j\omega_j(b_j^\dagger b_j+\tfrac{1}{2})+\sigma_z\sum_j g_j(b_j+b_j^\dagger) \ . \quad (15)$$

Here, K is the bare tunneling matrix element, ω_j is the phonon frequency for the jth mode, and g_j is the coupling constant. To make contact with (1), we must have

$$g_j=\Delta C_j q_0\left(\frac{1}{2\omega_j}\right)^{1/2} \ . \quad (16)$$

In the strong coupling limit or overdamped limit, the rate of tunneling is found by transforming to the small polaron basis and doing second order perturbation theory. We find[4,6]

$$\Gamma=\text{Re}\int_0^{+\infty}dt\,K^2(e^{\phi(t)}-e^{-\phi}) \quad (17)$$

with

$$\psi(t)=4\sum_j q_j^2\omega_j^{-2}\left[-i\sin(\omega_j t)+[\cos(\omega_j t)-1]\coth\left(\frac{\beta\omega_j}{2}\right)\right]$$
$$=\frac{4}{\pi}\int_0^\infty d\omega\,q_0^2\frac{J(\omega)}{\omega^2}\left[-i\sin(\omega t)+[\cos(\omega t)-1]\coth\left(\frac{\beta\omega_j}{2}\right)\right] \quad (18)$$

and

$$\phi=\frac{4}{\pi}\int_0^\infty d\omega\,q_0^2\frac{J(\omega)}{\omega^2}\coth\left(\frac{\beta\omega}{2}\right) \ . \quad (19)$$

Note that for Ohmic dissipation, even though $\phi=\infty$, $\psi(t)$ is finite and well defined for all temperatures. In fact, for $J(\omega)=\eta\omega e^{-\omega/\omega c}$, the integral in (18) can be done to yield

$$\phi(t)_{\text{Ohmic}}=q_0^2(4\eta/\pi)\left\{i\tan^{-1}(\omega_c t)-\tfrac{1}{2}\ln(1+\omega_c^2t^2)-\left[\frac{\beta}{\pi t}\sinh\left(\frac{\pi t}{\beta}\right)\right]\right\} \ , \quad (20)$$

agreeing exactly with Chakravarty and Leggett.[3] In the case $J(\omega)$ is given by (9) for $0\le\omega\le\omega_D$ ($d>1$), i.e., for acoustic phonons, it is easiest to evaluate (18) for strong coupling by the method of steepest descent. Equivalently, we can expand the integrand in (18) around $t=0$, keeping only quadratic terms:

$$\psi(t)=-it\Omega-t^2\delta/2 \ ,$$
$$\Omega=\frac{4}{\pi}\int_0^{\omega_D}q_0^2\frac{J(\omega)}{\omega}d\omega \ ; \quad (21)$$
$$\delta=\frac{4}{\pi}\int_0^{\omega_D}q_0^2J(\omega)\coth\left(\frac{\beta\omega}{2}\right)d\omega \ .$$

Then

$$\Gamma\cong\left(\frac{K^2}{\delta^{1/2}}\right)\exp-\left(\frac{\Omega^2}{2\delta}+K^2\right)\exp(-\phi) \ . \quad (22)$$

Using (9), we find $\Omega\sim q_0^{2-d}$, $\delta\sim q_0^{1-d}$, and $\phi\sim q_0^{3-d}$, so that the exponents in (22) vary as q_0^{3-d}, again assuming

$\beta c/q_0 \gg 1$ and $q_0\omega_D/c \gg 1$. The latter follows if q_0 is much larger than the nearest-neighbor distance, and the former for sufficiently low temperature. Combining this with the $d=1$ (Ohmic) result, we have $\log\Gamma \sim q_0^{3-d}$.[7]

Note that the essential aspects of our argument are (1) that $C_j(q_0) \sim \exp(i\mathbf{k}_j \cdot \mathbf{q}_0/2)$ and (2) that the density of phonon modes is proportional to ω^{d-1}. The first of these is true whenever a delocalized mode (like a phonon) interacts with a localized particle. The second is a feature of phonon systems; for electron-hole pair modes, or other excitations, the frequency dependence of the density of states is quite different. This will lead to other distance dependences. However, in the Ohmic case, a q_0^2 dependence as found by Phillips[1] is expected.

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[7]A more careful analysis of the $d=3$ result indicates that the exponent will vary as $\ln(q_0\omega_D/c)$, so that the dissipative correction in this case is algebraic.