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Dynamics of the dissipative multiwell system

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The motion of a quantum particle with Ohmic damping in a tight-binding lattice is discussed. An exact series representation in powers of the tunnel matrix element Δ for moments of the probability distribution is given. The dynamics at zero and at finite temperatures in the presence of an external force is solved exactly to all orders of Δ for a particular value of the friction coefficient $\eta = \pi\hbar/d^2$, where d is the lattice constant, and also for very weak damping.

The problem of the quantum transport of a particle coupled to a heat bath occurs in many different areas of physics including superionic conductors, atoms on surfaces, and interstitials in crystals. While earlier work was mainly concerned with the significance of polaron effects, e.g., in tunneling transitions of defects in solids[1] and exciton motion in molecular crystals,[2] recent attention has focused on the influence of a frequency-independent (Ohmic) damping mechanism. This type of dissipation influences, for instance, incoherent tunneling transitions of charged interstitials in metals[3] through the interaction with the conduction electrons.[4,5] Ohmic damping is also relevant for the quantum effects in the current-voltage characteristic of Josephson devices.[6] From a theoretical point of view the case of Ohmic damping represents the case of critical dimensionality[7] in which the phase diagram at zero temperature is nontrivial both for a double well[8] and a periodic potential:[9] At zero temperature there is a transition from an extended to a localized ground state as the dimensionless friction α is raised through a critical value $\alpha_c = 1$. Further, in a cosine potential there is a duality transformation between the tight-binding limit and the weak corrugation case in which weak and strong damping are exchanged, as shown for the partition function at $T=0$ (Ref. 10) and for the full dynamics at arbitrary T.[11]

We consider a particle moving in a "washboard" potential $V(q) = V_0(q) - Fq$, where $V_0(q)$ is a periodic potential with lattice spacing d, i.e., $V_0(q) = V_0(q+d)$, and F is an external force. The frequency of small oscillations about the minimum of a well is denoted by ω_0. Following Caldeira and Leggett[12] we assume that the heat bath can be represented by a set of harmonic oscillators coupled bilinearly to the particle. The system is then governed by the translationally invariant Hamiltonian

$$H = \frac{p^2}{2M} + V(q) + \sum_j \left[\frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 \left(x_j - \frac{C_j}{m_j \omega_j^2} q \right)^2 \right] , \tag{1}$$

and the influence of the environment on the particles motion is described by the spectral density

$$J(\omega) = \frac{\pi}{2} \sum_j \frac{|C_j|^2}{m_j \omega_j} \delta(\omega - \omega_j) .$$

In the case of Ohmic dissipation $J(\omega)$ has the form $\eta\omega$ for $\omega \rightarrow 0$, where η is the viscosity coefficient.

Here we study the motion of the system described by the Hamiltonian (1) in the tight-binding limit and calculate moments of the probability distribution as functions of time, temperature, damping strength, and external force.

To determine the time evolution of the probability distribution $P_n(t)$ of the damped particle, we use a functional-integral method based on the influence-functional theory of Feynman and Vernon.[13] In this approach the reduced density matrix is given by a double path integral where the paths $q(t)$, $q'(t)$ are weighted by the factor

$$\exp \left(\frac{i}{\hbar} (S_0[q] - S_0[q']) + F[q,q'] \right) .$$

Here, $S_0[q]$ describes the action of the undamped system and $F[q,q']$ is the complex influence functional describing the frictional influence of the environment. The diagonal elements of the reduced density matrix are the occupation probabilities $P_n(t)$. In the following, we choose the initial condition $P_n(t=0) = \delta_{n,0}$. In the tight-binding case each transition between different system states is associated with an amplitude $\pm i\Delta\, dt/2$. The transition amplitude, which may be calculated by instanton methods, is already renormalized for the high-frequency modes $\omega > \omega_0$ of the heat bath.[14] Further, it is convenient to transform the double path integral into a *single* path integral over states $\{n', n''\}$ by embedding the flip times t_j, t_k in chronological order into a single time interval extending from 0 to t. We then obtain $P_n(t)$ in the form of a power series in Δ^2. For reasons of space we merely sketch the derivation here.[15]

In order to specify all the possible paths $q(t)$, $q'(t)$ contributing to a given order Δ^{2m}, we define[16] $x(t) = [q(t) + q'(t)]/2$, $y(t) = q(t) - q'(t)$ and introduce "charges" $\chi_l = \pm 1$, $\xi_l = \pm 1$ with $l = 1, \ldots, 2m$, and time-ordered flip times t_l, $t_l < t_{l+1}$. Then a path composed of $2m$ transitions is given by

$$x(t) = \frac{d}{2} \sum_{l=1}^{2m} \chi_l \Theta(t - t_l), \quad y(t) = d \sum_{l=1}^{2m} \xi_l \Theta(t - t_l) , \tag{2}$$

where d denotes the lattice constant and $\Theta(t)$ is the standard step function. Now, the sum over all paths contributing to the order Δ^{2m} of the occupation probability $P_n(t)$

is represented by a sum over all configurations $\{\chi_l\}'=\pm 1$, $\{\xi_l\}'=\pm 1$, where the prime denotes that each configuration obeys the constraints

$$\sum_{l=1}^{2m}\chi_l=2n,\quad \sum_{l=1}^{2m}\xi_l=0\ . \tag{3}$$

Further, the influence functional introduces an interaction $Q(\tau)=Q_2(\tau)+iQ_1(\tau)$ between two flips with an interval τ, which for Ohmic dissipation assumes the form[17,18]

$$\begin{aligned} Q_1(\tau)&=2\alpha\arctan(\omega_0\tau)\ ,\\ Q_2(\tau)&=2\alpha\ln\left[\frac{\hbar\beta}{\pi\tau}\sinh\left(\frac{\pi\tau}{\hbar\beta}\right)\right]+\alpha\ln(1+\omega_0^2\tau^2)\ , \end{aligned} \tag{4}$$

where $\beta=1/k_BT$ and where α is related to the Ohmic viscosity η by $\alpha=\eta d^2/2\pi\hbar$. We finally arrive at the following expression for $P_n(t)$:

$$P_n(t)=\sum_{m=|n|}^{\infty}(-1)^{m-n}(\Delta/2)^{2m}\int_0^t dt_{2m}\int_0^{t_{2m}}dt_{2m-1}\cdots\int_0^{t_2}dt_1\sum_{\{\xi_j\}'}G_m(\{t_l\},\{\xi_j\})\sum_{\{\chi_j\}'}\exp\left[i\sum_{k=1}^{2m-1}\chi_kH_{k,m}(\{t_l\},\{\xi_j\})\right]\ , \tag{5}$$

where

$$G_m=\exp\left[\sum_{j=2}^{2m}\sum_{l=1}^{j-1}\xi_j\xi_lQ_2(t_j-t_l)-i\sigma\sum_{j=1}^{2m}\xi_jt_j\right]\ ,\quad H_{k,m}=\sum_{j=k+1}^{2m}\xi_jQ_1(t_j-t_k)\ . \tag{6}$$

Here, $\hbar\sigma$ is the potential drop between neighboring wells provided by the external force $F=\hbar\sigma/d$. On introducing the generating functional

$$Z(\lambda,t)=\sum_{n=-\infty}^{\infty}e^{\lambda dn}P_n(t)\ , \tag{7}$$

the sum over all possible "charge" configurations $\{\chi_j\}'$ can be performed explicitly and moments of $P_n(t)$ are readily obtained by differentiating $Z(\lambda,t)$ with respect to λ at $\lambda=0$. Following these lines we find

$$\langle q^N(t)\rangle=d^N\sum_{m=1}^{\infty}(-1)^{m-1}\Delta^{2m}\int_0^t dt_{2m}\int_0^{t_{2m}}dt_{2m-1}\cdots\int_0^{t_2}dt_1\sum_{\{\xi_j\}'}a_m^{(N)}(\{\xi_j\},\{t_l\})G_m(\{\xi_j\},\{t_l\})\ , \tag{8}$$

where we give $a_m^{(N)}$ for $N=1$ and $N=2$ explicitly:

$$\begin{aligned} a_m^{(1)}&=\frac{i}{2}\prod_{j=1}^{2m-1}\sin(H_{j,m})\ ,\\ a_m^{(2)}&=\frac{1}{2}\sum_{l=1}^{2m-1}\cos(H_{l,m})\prod_{\substack{j=1\\ j\neq l}}^{2m-1}\sin(H_{j,m})\ . \end{aligned} \tag{9}$$

Formulas (5)–(9) are general and exact expressions for the time evolution of a damped particle in a tilted periodic potential in the range $k_BT\ll\hbar\omega_0$ and $|\sigma|\ll\omega_0$ in which excited states in the individual wells can safely be neglected.

Clearly, it is impossible to sum up the cumbersome expression (8) to all orders of Δ^2 for arbitrary damping. We shall be able, however, to perform the summation to leading order in the ratio Δ/ω_0 exactly for the damping parameter $\alpha=\frac{1}{2}$ and also for weak damping $\alpha\ll 1$.

Before studying these cases two general remarks are appropriate. First, in earlier work[19,11] the dynamics have been solved by taking solely incoherent tunneling events between neighboring wells into account. Then the occupation probabilities $P_n(t)$ obey simple master equations. This case formally corresponds to restricting the paths contributing to (5) to the strip $|y|<d$ along the main diagonal $y=0$ of the (q,q') plane. Introducing the off-diagonal measure

$$g_{j,m}=-\sum_{l=1}^{j}\xi_l=\sum_{l=j+1}^{2m}\xi_l\ , \tag{10}$$

we then have $|g_{j,m}|\le 1$ for all j and m. In this approximation the nonlinear mobility

$$\mu=\lim_{t\to\infty}\langle q(t)\rangle/Ft \tag{11}$$

is found to be $\mu=\tanh(\beta\hbar\sigma/2)d^2\Gamma/\hbar\sigma$, where Γ is the rate of incoherent relaxation in an asymmetric double well.[20] Further, the linear mobility μ_l is related to the diffusion coefficient

$$D=\lim_{t\to\infty}\langle q^2(t)\rangle/t\ , \tag{12}$$

of the unperturbed system ($\sigma=0$) by the well-known Einstein relation

$$\mu_l=D/2k_BT\ , \tag{13}$$

where $D=D_0\equiv d^2\Gamma$ and where

$$\Gamma=\frac{\sqrt{\pi}}{2}\frac{\Delta^2}{\omega_0}\frac{\Gamma(\alpha)}{\Gamma(\alpha+1/2)}\left[\frac{\pi k_BT}{\hbar\omega_0}\right]^{2\alpha-1} \tag{14}$$

is the rate of incoherent relaxation in a symmetrical double well. Hence, in this approximation both the mobility and the diffusion coefficient are found to be simply proportional to Δ^2.[21] Below, these special results will be recovered in the high-temperature limit of our more general expressions.

Second, we note that the "noninteracting blip approximation" commonly used for damped double-well systems[7,17,20,22] fails for those paths of a multiwell system which are not restricted to the strip $|y| \le d$ along the main diagonal of the (q,q') plane.

For the special value $\frac{1}{2}$ of the damping strength α (Ref. 23) the evaluation of $\langle q(t)\rangle$ to all orders of Δ and to lowest order in the ratio Δ/ω_0 is possible. The crucial point, now, is that in the corresponding limit $\omega_0 \to \infty$, $\Gamma = \pi\Delta^2/2\omega_0 = \text{const}$, only those paths of a given order Δ^{2m} contribute to $\langle q(t)\rangle$ which belong to the strips $1 \le g_{j,m} \le 2$ and $-1 \ge g_{j,m} \ge -2$ for all j and m, respectively. Then, by introducing a Laplace integral representation the series (8) for $N=1$ can be summed to yield

$$\langle q(t)\rangle = \frac{2\Gamma d}{\pi}\frac{1}{2\pi i}\int_C ds \frac{e^{st}}{s^2}\,\text{Im}\psi\left[\frac{1}{2}+\frac{\hbar\Gamma}{\pi k_B T}+\frac{i\hbar\sigma}{2\pi k_B T}+\frac{\hbar s}{2\pi k_B T}\right], \tag{15}$$

where ψ is the digamma function and C the standard Bromwich contour. This expression is exact in leading order of Δ/ω_0 for arbitrary times t, temperature T, and bias energy $\hbar\sigma$ (as long as we maintain the condition $\hbar\beta\omega_0 \gg 1$, $\omega_0 \gg |\sigma|$). At zero temperature we find, from (15),

$$\langle q(t)\rangle = \frac{\hbar\sigma}{d}\mu(T=0)t + \frac{2\Gamma d}{\pi}\left[\frac{1}{2}\sum_{n=0}^{\infty}(-1)^n\frac{(\sigma/2)^{2n+1}}{(2n+1)!}\left(\frac{\partial}{\partial\Gamma}\right)^{2n}\frac{e^{-2\Gamma t}}{\Gamma^2} - \frac{\sigma}{4\Gamma^2+\sigma^2}\right]. \tag{16}$$

Here, the second term is only relevant at short times $t \lesssim 1/\Gamma$. In the first term we have introduced the nonlinear zero-temperature mobility

$$\mu(T=0) = \mu_0(2\Gamma/\sigma)\arctan(\sigma/2\Gamma) \tag{17}$$

which is conveniently normalized by

$$\mu_0 = 1/\eta = d^2/2\pi\hbar\alpha\ . \tag{18}$$

The nonlinear mobility at finite temperature is obtained from (11) and (15) as

$$\mu = \mu_0\frac{2\Gamma}{\sigma}\,\text{Im}\psi\left(\tfrac{1}{2}+\hbar\Gamma/\pi k_B T + i\hbar\sigma/2\pi k_B T\right)\ . \tag{19}$$

At high temperatures $k_B T \gg \hbar\Gamma$ this expression simplifies to

$$\mu = \mu_0\pi(\Gamma/\sigma)\tanh(\hbar\beta\sigma/2)\ ,$$

which is the result discussed above and obtained previously.[11,19] Correspondingly, the linear mobility μ_l at arbitrary temperatures is obtained from (19) as

$$\mu_l = (\hbar\Gamma\beta\mu_0/\pi)\psi'(\tfrac{1}{2}+\hbar\Gamma\beta/\pi)$$

where ψ' is the trigamma function. At $T=0$ we then obtain the strikingly simple expression

$$\mu_l(T=0) = \mu_0\ . \tag{20}$$

This result confirms for the particular value $\alpha = \frac{1}{2}$ a recent conjecture[11] that (20) holds for all $\alpha < 1$. This conjecture is based upon the duality transformation $\mu_l = \mu_0 - \bar{\mu}_l$ where $\bar{\mu}_l$ is the linear mobility in a weak-binding model with damping parameter $\bar{\alpha} = 1/\alpha$.

Next we discuss more briefly the second moment $\langle q^2(t)\rangle$ for $\alpha = \frac{1}{2}$. We find that the paths contributing to $\langle q^2(t)\rangle$ in leading order of Δ/ω_0 are restricted by the weaker constraint $|g_{j,m}| \le 3$ for all j and m. Now, the summation problem is more delicate than before. Also the resulting expressions are more complicated for general values of t and σ, except in the most interesting limit $t \to \infty$, $\sigma = 0$, in which we find

$$\lim_{t\to\infty}\langle q^2(t)\rangle_{\sigma=0} = (2d/\beta\hbar\sigma)\lim_{t\to\infty}\langle q(t)\rangle_l\ , \tag{21}$$

verifying that for arbitrary T and arbitrary orders of Δ^2 the linear mobility μ_l is related to the diffusion coefficient by the relation (13). Hence we find, for the diffusion coefficient,

$$D = (2/\pi^2)d^2\Gamma\psi'(\tfrac{1}{2}+\hbar\Gamma/\pi k_B T)\ , \tag{22}$$

which vanishes at zero temperature and reduces to the previous result $D = d^2\Gamma$ at temperatures $k_B T \gg \hbar\Gamma$. At zero temperature and long times $\Gamma t \gg 1$ the second moment is found to grow logarithmically as $\langle q^2(t)\rangle = (2/\pi^2)d^2\ln(2\Gamma t)$.

For weak damping $\alpha \ll 1$ the relevant expansion parameter in the series (8) is $u = \hbar\beta\Gamma\pi\alpha/2$. On keeping only the leading α dependence in each term of the power series (8), we find, in the limit $t \to \infty$,

$$\langle q(t)\rangle = t\frac{d}{\hbar\beta}\sum_{m=1}^{\infty}(-u)^m\sum_{\{\xi_j\}'}\prod_{j=1}^{2m-1}\text{Im}\left[\left(g_{j,m}+i\frac{\hbar\beta\sigma}{2\pi\alpha}\right)^{-1}\right]. \tag{23}$$

In the range $\hbar\beta\sigma/\alpha \le 1$, the neglected terms are at least smaller by a factor α^2 compared to those being kept.

Recently, Zwerger[16] has solved the combinatorial problem of the path summation in (23) and obtained the nonlinear mobility in the form of a continued fraction. Here, we note that by formal analogy with a recent result[24] (23) can be transformed into an integral expression yielding for the nonlinear mobility

$$\mu/\mu_0 = 1 - \frac{(2\pi/z)f(z)}{A_+(2\pi)A_-(2\pi) - f(z)\int_0^{2\pi}dx\exp[-g(x)A_+(x)}\ , \tag{24}$$

with

$$f(z)=1-\exp(-2\pi z),\ z=\hbar\beta\sigma/2\pi\alpha\ ,$$
$$g(x)=y\cos x+zx,\ A_{\pm}(x)=\int_0^x dx'\exp[\pm g(x')]\ ,$$

and

$$y=(2\pi\hbar\alpha\Gamma/k_BT)^{1/2}\ . \tag{25}$$

From (24) the linear mobility is obtained in the form

$$\mu_l=\mu_0[1-1/I_0^2(y)]\ , \tag{26}$$

where $I_0(y)$ is a modified Bessel function. Again, this expression simplifies for $T=0$ to $\mu_l=\mu_0$. We further note that the evaluation of $\langle q^2(t)\rangle$ in the analogous approximation gives a result which is again related to $\langle q(t)\rangle_l$ by (21). Hence the diffusion coefficient is found to be

$$D=d^2\frac{k_BT}{\pi\alpha\hbar}[1-1/I_0^2(y)]\ . \tag{27}$$

At high temperatures $k_BT\gg\hbar\Gamma\alpha$ from (27) the earlier result $D=D_0=d^2\Gamma$ which behaves like $T^{2\alpha-1}$ is recovered. In the low-temperature region, $y\gg 1$, we find $D=d^2k_BT/\pi\alpha\hbar$. Hence the diffusion coefficient vanishes at $T=0$ indicating that the spreading of the probability distribution is again slowing down at zero temperature. The diffusion coefficient has a maximum $D\simeq 0.4d^2\Delta/\pi\alpha$ at $T\simeq 0.8\hbar\Delta/k_B$.

In summary, we have investigated the quantum dynamics of a multiwell system where the environmental influences are modeled by an Ohmic heat bath represented by bosons. However, some of the qualitative features found here, e.g., that the diffusion coefficient linearly increases at very low temperatures and decreases like $T^{2\alpha-1}$ at high temperatures for $\alpha<1/2$, are expected to remain unchanged for a coupling to a Fermi bath.

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[1] C. P. Flynn and A. M. Stoneham, Phys. Rev. B **1**, 3966 (1970); Yu. Kagan and M. I. Klinger, J. Phys. C **7**, 2791 (1974); H. Teichler and A. Seeger, Phys. Lett. **82A**, 91 (1981).

[2] K. Kassner and P. Reinecker, Z. Phys. B **59**, 357 (1985), and references therein.

[3] For a general review, see the *Proceedings of the Fourth Conference on Muon Spin Rotations, Uppsala, 1986* [Hyperfine Interact. **31**, (1986)].

[4] J. Kondo, Physica B **84**, 40 (1976); **125**, 279 (1984).

[5] The dissipative influence of a Fermi bath has been investigated by F. Guinea, Phys. Rev. Lett. **53**, 1268 (1984). For a two-state system a fermionic bath is very similar to a bosonic bath with an Ohmic spectral density. See L. D. Chang and S. Chakravarty, Phys. Rev. B **31**, 154 (1985).

[6] S. Chakravarty, G. L. Ingold, S. Kivelson, and A. Luther, Phys. Rev. Lett. **56**, 2303 (1986); M. P. A. Fisher, *ibid.* **57**, 885 (1986); A. Kampf and G. Schön (unpublished).

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

[8] S. Chakravarty, Phys. Rev. Lett. **49**, 681 (1982); A. Bray and M. A. Moore, *ibid.* **49**, 1546 (1982).

[9] F. Guinea, V. Hakim, and A. Muramatsu, Phys. Rev. Lett. **54**, 263 (1985); S. A. Bulgadaev, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 264 (1984) [JETP Lett. **39**, 315 (1984)]; see also Refs. 10 and 11.

[10] A. Schmid, Phys. Rev. Lett. **51**, 1506 (1983).

[11] M. P. A. Fisher and W. Zwerger, Phys. Rev. B **32**, 6190 (1985).

[12] A. O. Caldeira and A. J. Leggett, Ann. Phys. (N.Y.) **149**, 374 (1983).

[13] R. P. Feynman and F. L. Vernon, Ann. Phys. (N.Y.) **24**, 118 (1963).

[14] U. Weiss, H. Grabert, P. Hänggi, and P. Riseborough, Phys. Rev. B **35**, 9535 (1987).

[15] Further details and extensions of this work will be given elsewhere.

[16] W. Zwerger, Phys. Rev. B **35**, 4737 (1987).

[17] S. Chakravarty and A. J. Leggett, Phys. Rev. Lett. **52**, 5 (1984).

[18] For simplicity we have chosen an exponential cutoff of the modes $\omega>\omega_0$ in the frequency integral defining $Q(\tau)$. Although the exact duality transformation only holds for an algebraic cutoff (see Ref. 11) the difference is irrelevant in leading order of Δ/ω_0.

[19] U. Weiss and H. Grabert, Phys. Lett. **108A**, 63 (1985).

[20] H. Grabert and U. Weiss, Phys. Rev. Lett. **54**, 1605 (1985); M. P. A. Fisher and A. T. Dorsey, *ibid.* **54**, 1609 (1985).

[21] The inclusion of self-energy corrections is discussed by U. Eckern and F. Pelzer, Europhys. Lett. **3**, 131 (1987).

[22] U. Weiss and H. Grabert, Europhys. Lett. **2**, 667 (1986); U. Weiss, H. Grabert, and S. Linkwitz, J. Low Temp. Phys. **68**, 213 (1987).

[23] We note that in this limit our model can be mapped onto a model involving fermions. The corresponding linear mobility at $T=0$ has been calculated by F. Guinea, Phys. Rev. B **32**, 7518 (1985), and coincides with our result from Eq. (19).

[24] H. Risken, *The Fokker-Planck Equation* (Springer, Berlin, 1984), p. 289.