Semiclassical analysis of the Nonequilibrium Local Polaron

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A resonant level strongly coupled to a local phonon under nonequilibrium conditions is investigated. The nonequilibrium Hartree-Fock approximation is shown to correspond to approximating the steady state density matrix by delta functions at field values to which the local dynamics relaxes in a semiclassical limit. If multiple solutions exist, all are shown to make nonvanishing contributions to physical quantities; multistability does not exist. Nonequilibrium effects are shown to produce decoherence, causing the standard expansions to converge and preventing the formation of a polaron feature in the spectral function. The formalism also applies to the nonequilibrium Kondo problem.

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\[ H_f = \lambda q d^d d; \quad H_{ph} = \frac{p^2}{2m_{ph}} + U(q) \] (3)

At \( M_{ph} = \infty \) the model is analytically solvable, with properties determined by the Green functions

\[ g_{q}(t-t') = \begin{pmatrix} g_{qR}(t-t') & g_{qR}(t-t') \cr 0 & g_{qA}(t-t') \end{pmatrix} \] (4)

where \( g(t) = \int \frac{d\omega}{2\pi} e^{-i\omega t} g(\omega) \) with

\[ g_{qR}(\omega) = \frac{1}{\omega - \epsilon_0 - \lambda q - \Sigma(\omega + i\delta)} = g_{qA}(\omega)^* \] (5)

\[ g_{qK}(\omega) = 2i \sum_{j=L,R} \left[ a_j(\omega) \tanh(\beta_j(\omega - \mu_j)) \right] , \] (6)

and \( \Sigma = \Sigma_L + \Sigma_R, \Sigma_j(z) = \sum_p \frac{i\omega}{z - \epsilon_p + \Sigma_j} \) and \( a_j = Im \Sigma_j(\omega - i\delta)/\left( (\omega - \epsilon_0 - \lambda q - Re \Sigma(\omega))^2 + Im \Sigma^2(\omega) \right) \). It is also useful to define \( b = Reg_R \) and the 'pseudoenergy'

\[ \Phi(q) = U(q) + \int_0^q dq' \lambda q' \int \frac{d \omega}{2\pi i} \frac{g_{qR}(\omega) - g_{qR} + g_{qA}}{2} \] (7)

which in equilibrium becomes the \( q \)-dependent part of the total energy. Fig. 1 shows a possible form of \( \Phi(q) \).

The equilibrium physics of this model is very well understood, and is conveniently viewed as a functional integral over trajectories \( q(t) \). In the limits \( M_{ph}, \beta \to \infty \) the integral is dominated by those paths for which \( q \) takes the definite value \( q_n \) minimizing the energy. At finite \( M_{ph} \), other paths become important: two crucial classes are the 'gaussian fluctuation' paths involving small excursions (characteristic frequency \( \omega_n \) from the mininan and, if \( \Phi_{eq}(q) \) has the form shown in Fig 2 and the barrier height \( H \) is sufficiently large, tunnelling (instanton) processes during which \( q \) goes rapidly from the vicinity of the global minimum (\( E_1 \) in the notations of Fig 2) to the higher energy minimum (here \( E_2 \), spends a time of order \( \Delta E^{-1} \equiv (E_2 - E_1)^{-1} \) near the higher minimum, and then returns to the vicinity of the global minimum. If \( H \) is sufficiently large, then tunnelling processes are rare, but when \( E_2 \approx E_1 \), the time spent in the higher minimum becomes longer than the interval
between tunnelling events and the dilute instanton approximation breaks down, signalling the formation of a polaron resonance in the density of states. In this paper we investigate the changes occurring when the system is driven out of equilibrium by application of a chemi-

cal potential difference $\Delta$ between the two leads. We present a general discussion but focus most attention on

the nonequilibrium polaron limit $H >> \omega$ with $\beta_j = \infty$ but $\Delta E/\Delta \mu$ arbitrary. In our actual calculations we also assume that departures from equilibrium are small enough that we may neglect density-of-states variations: $(\Delta \mu) \partial \ln a / \partial \omega < 1$.

To analyse the out of equilibrium behavior we use the standard extension of Feynman diagrams to the Keldysh contour which consists of a time-ordered $(-)$ branch extending from $t = -\infty$ to $t = +\infty$, followed by an anti-time-ordered $(+)$ branch extending from $+\infty$ back to $-\infty$. The Keldysh diagrams may be obtained by functional differentiation, with respect to source terms $\eta_a(t)$, of a generating functional $W[\{\eta_a\}]$ which can be formulated as a coherent state path integral:

$$W[\{\eta_a\}] = \int dq dq_- \bar{\rho}(q_+, q_-) \int Dq(t) \tilde{S}_K[-\infty_+, -\infty_-]$$

Here the functional integral $\int Dq(t)$ is over all paths beginning at $q_-$ at $t = -\infty$ on the time ordered contour and ending at $q_+$ at $t = -\infty$ on the antitime ordered contour, while $\tilde{S}_K[t_1, t_2; \eta_a(t)] = T_K e^{i \int_{t_1}^{t_2} dta(t) R[\eta_a(t)]}$ is the time evolution operator on the Keldysh contour and $a(t) = \pm 1$ according to whether the time is on $-\text{ or +\text{ branch}}$. The contributions of paths with endpoints $q_+, q_-$ are weighted by the appropriate element of the steady state density matrix $p$ which is the long time limit of an equation which may be expressed in path integral language as

$$\bar{\rho}(q_+, q_-; t; \eta) = \int dq'_+ dq'_- \int Dq'(t') \tilde{S}_K[t_+, t_0+; \bar{\rho}(q'_+, q'_-; t_0)] \tilde{S}_K[t_0-, t_-]$$

Here the integral $Dq'(t')$ is over all paths which begin at $q'_-$ on the lower contour at time $t_0$ and end at $q_-$ time $t$ on the lower contour, along with the time reversed paths which begin at $q'_+$ (t) on the upper contour and return to $q_+$ at time $t_0$. We require the long time behavior after transients have decayed. We do not find non-steady long time limits such as limit cycles or chaos: $\rho$ in Eq. 8 is the time independent solution of Eq. 9.

Because Eqs. 8, 9 involve a finite system coupled to two infinite reservoirs, the trace over electron operators may be performed. The combination of time evolution operators needed in Eqs. 8, 9 may be written as a coherent state path integral:

$$\mathcal{R}(t_2, t_1; \{q_c(t), q_q(t)\}) = e^{-i \int_{t_1}^{t_2} dt (\lambda q_c(t) n_c(t) - \lambda q_q(t) n_q(t))} \langle \rho_q(t) = \frac{q_q(t)}{2} \rangle$$

where $q_c(t) = \frac{q_q(t)}{2}$, $q_q(t) = \frac{q_q(t) - q_q(t)}{2}$, $\langle \rangle$ denotes expectation value in the reservoir defined by the trace (note this trace depends on the entire trajectory $q(t)$) and $n_{+/-} = \delta_{d} d$ on the indicated contour. In the physical problem the quantum field coupling constant $\lambda_q = \lambda$. Differentiation with respect to $\lambda_q$ and application of the usual linked cluster arguments leads to

$$\mathcal{R}(t_2, t_1; \{q_c(t), q_q(t)\}) = e^{-\int_{t_1}^{t_2} dt C(t) + \mathcal{L}_{ph+S}}$$

Here $\mathcal{L}_{ph+S}$ is the phonon action $M_{ph}(\partial_t q_c(t)) - U(2q_c^2 - U/2)$ supplemented by source terms,

$$C(t) = q_q(t) \int_0^\lambda d\lambda_q Tr [\sigma_x G_{\lambda_q}(t, t)]$$

and $G_{\lambda_q}$ solves

$$G_{\lambda_q} = g + g^* v \ast G_{\lambda_q}$$

with $v(t) = (\lambda q_c(t) 1 + \lambda q_q(t) \sigma_x)$. We have verified that an expansion in powers of $\lambda$ about the weak coupling limit $\lambda = 0$ reproduces results obtained by standard Keldysh diagrammatic analysis.

We now turn to the semiclassical analysis. In the large $M_{ph}$ limit, one expects stationary (time-independent) paths to dominate the physics. However, in the Keldysh formalism, stationary paths have $q_q = 0$, so in the absence of source terms the time evolution operator in Eq 8 is unity for all stationary paths, providing no basis for selection. Instead, the important paths are selected by the density matrix; i.e. from the solution of Eq 9.

To understand the dynamics implied by Eq 9 we first assume that $\rho(q, q')$ is strongly peaked near $q = q' = q_0$. We may then expand $\Phi = E_0 + \frac{M_{ph} \omega^2}{2} (q - q_0)^2$ and analyse Eq 9 by the usual perturbative method. In the large mass, weakly nonequilibrium limit we find relaxation, with a rate of order $\lambda^2 \omega (a_R + a_L)^2$ to a sum of functions sharply peaked about the values $q_a$ which minimize the pseudoenergy.

$$\rho(q, q') \approx \sum_a p_a r_a(q, q')$$

To leading order in the electron-phonon coupling $\lambda$ we find $r_a(q, q') = \int \frac{dp}{2\pi} \rho_p(q - q') E \rho_p(-\frac{p^2}{2M_{ph}} +$
the nearly complete cancellation of a sum of many paths approximated by its equilibrium value. However, in the antikink is infinite, so each kink must be followed by an important only for \( \Delta \mu > \omega \). Thus we expect roughly that for \( \Delta \mu > \omega_a \), the diffusion process dominates.

\[
\text{ln} R_{\text{tun}} \approx -2\sqrt{\frac{2\pi}{\omega_a \omega_\mu}} \sim \frac{H}{\omega_a};\text{ corrections become important only for } \Delta \mu \sim \omega_a.
\]

\section*{Instanton processes entering equation of motion of density matrix.}

We now consider in more detail the interesting quantum limit \( \Delta \mu < \omega_a \) for large barrier height \( H \). In this limit one would like to restrict attention to paths for which \( q(t) \) is almost always near one of the minima, with occasional tunnelling events in which \( q \) shifts from one minimum to another. The tunnelling amplitude \( R_{\text{tun}} \) is exponentially small and (for small enough \( \Delta \mu \)) is well approximated by its equilibrium value. However, in the real time path integral the smallness of \( R_{\text{tun}} \) arises as the nearly complete cancellation of a sum of many paths with large but oscillating amplitudes. We argue that for small \( \Delta \mu \) the result of performing this complicated sum (which we do not treat directly) may be represented by the 'instanton' (kink) vertices shown in Fig 2 with amplitude \( R_{\text{tun}} \). A kink or antikink comes with a factor \( i \) and a sign \( \sigma = \pm 1 \) determined by the contour it is on. We find (see below) that the action of a single kink or antikink is infinite, so each kink must be followed by an antikink. The path integral Eq 9 thus becomes

\[
\rho_a(t) = R_{\text{tun}} \int_0^t dt_f dt_i \rho_a(t_f) \rho_a(t_i) \rho_a(t_0) \rho_a(t_0) - R_{\text{tun}} \int_0^t dt_f dt_i \rho_a(t_f) \rho_a(t_i) \rho_a(t_0) + O R^4
\]

where the first term is the sum of the processes shown in panels \( a \) and \( b \) in Fig. 2, the second term is the sum of the processes shown in \( c \) and \( d \), and we have denoted explicitly only the beginning and ending values of the classical component of the field.

Differentiating Eq 14 with respect to time yields

\[
\frac{d\rho_a(t)}{dt} = -\Gamma_{a\to b}\rho_a(t) + \Gamma_{b\to a}\rho_b(t)
\]

where to order \( R_{\text{tun}}^2 \) the scattering rates are

\[
\Gamma_{a\to a'} = R_{\text{tun}}^2 Re \int_0^\infty dt \rho(t_0; \{q_{ca} \to q_{ca'}\})
\]

The integral in Eq 16 yields a quantity \( T_0 \) with dimension of time; we may neglect higher order terms if \( R_{\text{tun}} T_0 < 1 \). To evaluate Eq 16 it suffices to approximate the potential in Eq 12 by the 'telegraph' form shown in Fig 2; the equations may then be solved by standard methods. In equilibrium a crucial role is played by the \( \text{dec} \) energy arising from the imbalance in chemical potential. The tunnelling amplitude \( R_{\text{tun}} \) is well peaked about \( q_a \),  with di-
be computed perturbatively or numerically. We find

\[ \phi_2(x) = \left( \frac{2}{\pi} \right) \left( \sin(x) - \frac{1 - \cos(x)}{x} \right). \]

\( \delta C_{\text{ortho}} \) expresses the change in orthogonality effects arising because at nonvanishing \( \Delta \mu \) there is destructive interference between the left and right leads.\(^\text{11}\)

\[ \delta C_{\text{ortho}}(\omega) = \frac{2 Re \delta \rho}{\pi^2} \psi_1(\Delta \mu t) + \frac{2i Im \delta \rho}{\pi^2} \psi_2(\Delta \mu t) \]

with \( \psi_1(x) = \gamma_x - C_i(x) + \ln(x) \) and \( \psi_2(x) = \frac{2}{\pi} \int_0^1 du \sin(ux) \left[ \frac{1 - u}{\ln(1 - u) + u \ln u} \right] \).

We have used the perturbative expressions for the crossover functions to evaluate \( \Gamma \) (Eq. 10). In equilibrium and at \( \beta_j = \infty \) we find that only scattering from the higher energy to the lower energy extremum occurs (the ‘up-scattering rate’ vanishes); under nonequilibrium conditions an upscattering rate appears: of order \( \left( \frac{\Delta \mu}{\Delta E} \right) \left( \frac{\Delta \rho}{\rho} \right) \) if \( \frac{\Delta E}{\Delta \mu} >> 1 \) but of the order of the down-scattering rate as \( \frac{\Delta E}{\Delta \mu} \rightarrow 0 \). The change in the weight \( \rho_1 \) for one of the two minima as \( \Delta \mu/\Delta E \) is varied is shown in the lower inset of Fig 3. A similar result was found by Parcollet and Hooley in a ‘pseudofermion’ diagrammatic study of the nonequilibrium Kondo problem.\(^\text{13, 14}\)

The semiclassical Greens function follows as:

\[ G_R(\omega) = \sum_a \frac{\rho_a (\Delta \mu)}{\omega - \epsilon_a - g q_a - \Sigma_R(\omega)} \]

The main panel of Fig 3 shows the current computed in the standard way by inserting the Green function given in Eq. 21 into Eq 28 of Ref. 7. The inset shows the evolution of the spectral density \( A(\omega) = Im R(\omega) \).

The minimum condition \( \partial \phi / \partial q = 0 \) is equivalent to the Hartree-Fock equations discussed in recent literature,\(^\text{13, 14}\) but in these works it is assumed that at each set of parameter values, only one of the minima is occupied (\( \rho_a = 0 \) or 1); preparation conditions are argued to determine the state of the system, leading to multistability and switching, in contradiction to the results shown in Fig 3. Although the bistability discussed by \( \text{13, 14} \) does not occur, the slow dynamics governing equilibration between the minima will lead to ‘telegraph’ noise in the current.

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