Spin dynamics and violation of the fluctuation dissipation theorem in a non-equilibrium ohmic spin boson model.

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We present results for the dynamics of an impurity spin coupled to a magnetic field and to two ohmic baths which are out-of equilbrium due to the application of a bias voltage. Both the non-equilibrium steady state and the rate constants describing the approach to steady state are found to depend sensitively on the relative strengths of a magnetic field and a voltage dependent decoherence rate. Computation of physical quantities including the frequency dependent ratio of response to correlation functions and the probabilities of the two spin states allows the extraction of voltage dependent effective temperatures. The temperatures extracted from different quantities differ from one another in magnitude and their dependence on parameters, and in general are non-monotonic.

A fundamental question in quantum condensed matter physics is understanding properties of non-equilibrium many body systems, some examples being the Kondo effect in quantum dots, ultra-cold gases with rapidly tunable interactions, and strongly driven optical lattices. While there are a variety of non-perturbative techniques in place to study equilibrium systems, these methods cannot be extended to non-equilibrium systems in a straightforward way. The experimental accessibility of the non-equilibrium regime of strongly correlated quantum many body systems gives rise to the need for developing the formalism further.

Many body systems driven out of equilibrium are known to acquire a steady state that may be quite different in character from their ground state properties, with the details of the steady state depending on the nature of the correlations, as well as on the way in which the system is driven out of equilibrium. One may characterize a system in steady state by the response function \( \chi(t) \) describing changes induced by weak external probes, and by the correlation function \( S(t) \) describing the probabilities of observing various configurations of the system. An important and still incompletely understood issue is the manner in which \( \chi \) and \( S \) characterizing a non-equilibrium system differ from those describing an equilibrium one. In particular there has been considerable interest in the possibility of establishing a generalized fluctuation-dissipation theorem relating \( \chi(\omega) \) to \( S(\omega) \) and thereby characterizing the deviations from equilibrium in terms of an effective temperature. Several systems have been identified where such a generalized fluctuation dissipation theorem is found to hold, with the extracted temperature often sensitive to the observables being studied.

In this paper we study the dynamics of the out of equilibrium ohmic spin-boson model. This model describes a two state system (which we represent in spin notation) with level splitting \( 2B \) and tunneling rate \( \Delta \), coupled via a coupling \( J_z \) to a spin-less resonant level (creation operator \( d_\uparrow \)), which is itself connected to two leads (\( L \) and \( R \)) that may be at different chemical potentials. The Hamiltonian is

\[
H = S_z B + \Delta S_x + J_z S_z d_\uparrow d + H_{\text{bath}}
\]

\[
H_{\text{bath}} = \sum_{k,\alpha=L,R} \epsilon_k c_{k\alpha}^\dagger c_{k\alpha} + \sum_{k,\alpha=L,R} \left( t_{k\alpha} c_{k\alpha}^\dagger d + h.c. \right)
\]

We assume the leads are infinite reservoirs characterized by the correlators \( \langle c_{k\alpha}^\dagger c_{q\beta} \rangle = \delta_{kq} \delta_{\alpha\beta} \left( e^{\beta (\epsilon_k - \mu_\alpha)} + 1 \right)^{-1} \), and a non-equilibrium state occurs when \( \mu_L - \mu_R = V \neq 0 \). Crucial parameters of the model are the left and right channel phase shifts \( \delta_{L,R} \) defined by \( \tan \delta_{L,R} = \frac{a_{L,R}}{a_{\text{inc}} \pm \Gamma_{L,R}} \), \( \tan \delta_{R} = \frac{a_{L}}{a_{R}} \tan \delta_{L} \) with \( a_{L,R} = \frac{V_{\text{rel}}}{\Gamma_{L,R}} \). We will study properties of \( H \) at \( T = 0 \) but out of equilibrium \( (V \neq 0) \) working to leading nontrivial order in \( \Delta \) but to all order in \( J_z \). The conditions under which perturbation theory is justified will be discussed below. We will find the time scales characterizing the approach to steady state, the response and correlation functions and the generalized fluctuation dissipation ratio.

In order to study the spin-dynamics, the appropriate starting point is the density matrix for the full Hamiltonian

\[
\frac{d\rho(t)}{dt} = -i [H, \rho(t)]
\]

from which the density matrix \( \rho_S \) for the local spin is obtained from taking a trace over the electronic degrees of freedom.

\[
\dot{\rho}_S = Tr_{\text{el}} \rho
\]

We adopt a spin language, writing

\[
\dot{\rho}_S = \frac{1}{2} (1 + S_z \hat{\sigma}_z)
\]
FIG. 1: Main panel: Effective distribution function derived from Eq. 24 at $B = 0.05V$. The dashed line is tanh($\frac{\theta}{2T_{\text{eff}}}$). At equilibrium $h(\omega) = \text{sgn}(\omega)$. Inset: Plot of $T_{\text{eff}}(\omega, V) = \frac{2\text{Im}h(\omega) - 1}{\omega}$ which shows a rapid crossover from non-equilibrium behaviour ($T_{\text{eff}} \sim V$) to equilibrium behaviour ($T_{\text{eff}} = 0$) at $\omega \sim V/2$.

and study $S_z$. When $\Delta = 0$, the Hamiltonian is exactly solvable both in and out of equilibrium. For non-zero $\Delta$ one may expand Eq. 3 perturbatively in $\Delta$. The key object in the analysis is the time-evolution operator separating two spin-flip processes $K_{\pm}(t) = T_{\text{rel}}\left[e^{-\frac{i}{\hbar}H(\Delta = 0, \delta_{\pm} \pm 1/2)H}e^{\frac{i}{\hbar}H(\Delta = 0, \delta_{\pm} \pm 1/2)H}\right] = e^{\pm i B t e^{-C_z(t)}}$ where $C_{\pm}(t) = C(t) = (C_{\mp}(-t))^*$ computed from the linked cluster theorem has the following form at zero temperature,

$$C(t) = C'(t) + iC''(t)$$

$$C'(t) = (\frac{\delta_{eq}}{\pi})^2 \log(\xi t) + \phi'(Vt)$$

$$C''\pi = \frac{\pi}{2} (\frac{\delta_{eq}}{\pi})^2 \text{sgn}(t) + \phi''(Vt)$$

Here $\xi$ is a short time cut-off, $\delta_{eq} = \delta_L + \delta_R = \arctan\left(\frac{J_1}{1 + J_1}\right)$ is the equilibrium phase shift, and $\phi(Vt)$ is a function describing the crossover from the short-time ($Vt \ll 1$) equilibrium behaviour characterized by $\xi_{eq}$,

$C(t) = (\frac{\delta_{eq}}{\pi})^2 \sqrt{\log(\xi t) + i\frac{\pi}{2} \text{sgn}(t)}$, to the long time ($Vt \gg 1$) non-equilibrium behaviour characterized by $\xi_{neq}$

$C(t) = (\frac{\delta_{eq}}{\pi})^2 \sqrt{\log(\xi t) + i\frac{\pi}{2} \text{sgn}(t)} + \Gamma_{\text{neq}} t$ with $\Gamma_{\text{neq}} = V + \frac{\delta_{neq}^2}{2\pi^2}$. Correct treatment for the intermediate and short time behaviour of $\phi$ is essential for obtaining correct results for $\chi(\omega), S(\omega)$. A general analytic expression for $\phi$ does not exist, here we use perturbation theory to third order in $J_z$ to obtain,

$$\phi(Vt) = \frac{\delta_{neq}^2}{2\pi^2} \sqrt{\log(\xi t) + i \frac{\pi}{2} \text{sgn}(t)} - \frac{2i \text{Im}\delta_{eq}\delta_{neq}}{\pi^2} \left[\text{Ci}(Vt) + \log(Vt)\right] - \frac{2i \text{Im}\delta_{eq}\delta_{neq}}{\pi^2} \left[\frac{\pi}{2} \int_0^1 du \sin(uVt)\right] \left[1 - \left(1 - u\right) \log(1 - u) + u \log(u)\right]$$

Now let us return to the evaluation of various spin observables. In terms of the symmetric and anti-symmetric time-evolution operators $K_{s,a}(t) = \text{Re}[K_{\mp}(t) \pm K_{\mp}(t)]$, the equation of motion for the variable $S_z$ parameterized in Eq. 4 to leading non-trivial order in $\Delta$ is given by

$$\frac{dS_z}{dt} = -\int_0^t dt'[K_s(t, t')S_z(t') + K_a(t, t')]$$

The Laplace transform of the two scattering rates $K_{s,a}$ defined by $\tilde{K}_{s,a}(\lambda) = \int_0^\infty dt e^{-\lambda t} \text{Re}e^{-C_z(t)}$ have the form,

$$\tilde{K}_s(\lambda) = \Delta^2 \int_0^\infty dt e^{-\lambda t} e^{-C_z(t)} \cos \omega t \cos C''(t)$$

$$\tilde{K}_a(\lambda) = \Delta^2 \int_0^\infty dt e^{-\lambda t} e^{-C_z(t)} \sin \omega t \sin C''(t)$$

The above equations capture the effect of two sources of decoherence on spin dynamics, one is a Korringer type decoherence existing even in equilibrium, while the second arising mathematically from $C_z(t)$ is due to a non-zero voltage and is intrinsically non-equilibrium.

The solution to Eq. 9 can be written as

$$S_z(t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{d\lambda}{\lambda} e^{\lambda t} \left[\tilde{K}_s(\lambda) - \tilde{K}_a(\lambda)\right]$$

Eq. 12 allows straightforward analysis of the long-time behaviour. As $t \to \infty$, the integral is dominated by the pole at $\lambda = 0$ and the residue gives $S_z(\infty) = S_z(t \to \infty) = -\tilde{K}_s(0)$. Consideration of $S_z(t) - S_z(\infty)$ then yields the rate at which the system approaches steady state. In the small $\Delta$ limit, and if at least one of $B, \Gamma_{\text{neq}}$ is not too small, the result is exponential relaxation with rate $\Gamma_{\text{rel}} = K_s(0)$. The value of $\Gamma_{\text{rel}}$ depends crucially on whether the dominant time scales in Eq. 10 are large or small relative to $V^{-1}$. If both $B$ and $\Gamma_{\text{neq}}$ are less than $V$, one finds (for compactness we write for the symmetric case $t_L = t_R$)

$$\Gamma_{\text{rel}} = \tilde{K}_s(0) = \frac{\Delta^2}{\pi^2} \frac{\sin \left[\frac{\pi}{2} (\frac{1}{\Gamma_{\text{rel}}} - 1) \arctan \frac{\pi}{2} \right]}{\sin \left[\frac{\pi}{2} \right]} \left(\sqrt{B^2 + \Gamma_{\text{neq}}^2} \delta_{neq}\right)^{\alpha - 1}$$

where the non-equilibrium exponent $\alpha = \left(\frac{\delta_{neq}}{\pi}\right)^2$, yielding the familiar $T = 0$ Korringer relaxation. The above results of an exponential relaxation to steady state is obtained from neglecting the $\lambda$ dependence of $K_s(\lambda)$, which is justified when

$$\frac{\Gamma_{\text{rel}}}{\Gamma_{\text{neq}}} \sim \frac{\Delta^2}{\xi^2} \left(\sqrt{B^2 + \Gamma_{\text{neq}}^2} \delta_{neq}\right)^{\alpha - 2} \ll 1$$
and therefore holds in the perturbative in \( \Delta \) regime provided the voltage or the magnetic field is not too small. Analysis similar to the equilibrium case shows that Eq. 14 is also the condition for validity of perturbation theory in \( \Delta \).

At steady state and in the small \( \Delta \) limit, the density matrix for the full system is an incoherent superposition of spin up times the electronic state appropriate to spin-up and spin-down times the electronic state appropriate to spin-down, which may be expressed follows,

\[
\rho = \rho_{S} \otimes \rho_{\text{el}} = \begin{pmatrix} \rho_{\uparrow \uparrow} & 0 \\ 0 & \rho_{\downarrow \downarrow} \end{pmatrix}
\]

where \( \rho_{\uparrow \uparrow} \) is the density matrix corresponding to Hamiltonian \( H \) with \( S_z = 1/2 \) and \( \Delta = 0 \). Likewise \( \rho_{\downarrow \downarrow} \) is the density matrix for \( S_z = -1/2 \) and \( \Delta = 0 \), while \( \rho_{\uparrow \downarrow} = \frac{1}{2}(I \pm S_z) \). We now calculate the response and correlation functions appropriate to this state and also study the fluctuation-dissipation relation between them.

The correlation function we study is,

\[
S_{xx}(t_1, t_2) = \langle [S_x(t_1), S_x(t_2)] \rangle = Tr [\rho(S_x(t_1), S_x(t_2))] + \]

and the corresponding spin response function is,

\[
\chi_{xx}(t_1, t_2) = \langle -i\theta(t_1 - t_2)\{[S_x(t_1), S_x(t_2)]\} \rangle = -i\theta(t_1 - t_2)Tr [\rho[S_x(t_1), S_x(t_2)]]
\]

where the density matrix \( \rho \) is evaluated to leading order in the spin flip term \( \Delta \) and hence given by Eq. 15. The Fourier transform of the imaginary part of the response and correlation functions are,

\[
\chi''_{xx}(\omega) = \rho_{\uparrow} [I(B + \omega) - I(B - \omega)] - \rho_{\downarrow} [I(-B - \omega) - I(-B + \omega) ]
\]

\[
-i\chi'_{xx}(\omega) = \rho_{\uparrow} [I(B + \omega) + I(B - \omega)] + \rho_{\downarrow} [I(-B - \omega) + I(-B + \omega)]
\]

where

\[
I(B) = Re \left[ \int_{0}^{\infty} dt e^{itB} e^{-C(t)} \right]
\]

and \( \frac{\rho_{\uparrow}}{\rho_{\downarrow}} = \frac{I(-B)}{I(B)} \). We also consider the fluctuation-dissipation ratio (also mentioned in \( \delta \))

\[
h(\omega) = \frac{\chi''_{xx}(\omega)}{i\chi'_{xx}(\omega)}
\]

In equilibrium and at \( T > 0 \) (when \( \phi(0) = 1 \) and \( C(t) = \left( \frac{\omega}{2\pi} \right) \ln \left( \frac{\sinh \pi T}{\sin \pi t} \right) \)), \( h(\omega) = \tanh \frac{\omega}{2T} \).

Out of equilibrium (and at \( T = 0 \)), \( h(\omega) \) has the form shown in Fig. 1 which differs from the equilibrium solution \( sgn(\omega) \). The calculated \( h(\omega) \) is not a tanh function (compare dashed line), and therefore a generalized fluctuation dissipation theorem encompassing all frequencies does not exist. However we may define a frequency dependent effective temperature via \( T_{eff}(\omega) = \frac{\omega}{\partial h(\omega)/\partial \omega} \). This function is plotted in the inset of Fig. 1 and is seen to have a strong \( \omega \) dependence and is indeed non-monotonic. For \( \omega < V/2 \), \( T_{eff} \) is seen to be of the order of \( V \) and to depend weakly on \( \omega \). For \( \omega > V/2 \), \( T_{eff} \) drops sharply and at high \( \omega \) approaches the equilibrium value (here, \( T = 0 \)).

The results presented in Fig. 1 show that no unique definition of “non-equilibrium effective temperature” exists, the value obtained depends on the quantity examined. Two obvious definitions are (i) from the \( \omega \) \( \rightarrow \) 0 limit of \( h(\omega) \), (ii) from the population ratio \( \rho_{\uparrow}/\rho_{\downarrow} \). The effective temperature from definition (i) is obtained by expanding Eq. 18 and Eq. 19 for small \( \omega \),

\[
\frac{1}{T_{eff}} = \frac{\partial h(\omega)}{\partial \omega} |_{\omega=0} = \sum_{\sigma=\pm} \frac{\partial \ln I(\omega)}{\partial \omega} |_{x=\sigma B}
\]

while definition (ii) for the effective temperature leads to the expression

\[
\frac{1}{T_{eff}} = \frac{1}{B} \ln \frac{\rho_{\uparrow}}{\rho_{\downarrow}}
\]

Fig. 2 shows the dependence of these two measures of effective temperature on magnetic field. We see that the two curves differ in magnitude and in dependence on parameters; the variation in general is non-monotonic. The inset shows that the magnitude and field variation of the effective temperature (plots are for \( T_{eff} \)) also depends on coupling constant.

The non-monotonic behaviour as a function of \( B/V \) may be understood as follows. For \( B \ll \Gamma_{\text{neq}} \) and for \( \frac{\delta \phi}{2\pi} \ll 1 \) so that \( \Gamma_{\text{neq}} \ll V \), the integrand in Eq. 20 is dominated by \( t \sim 1/\Gamma_{\text{neq}} \gg 1/V \). In this regime, Eq. 19 applies; from this one sees that the decoherence rate for the spin increases with magnetic field causing the initial
the integral
to
δ
a log-log plot for spin-bath coupling strength corresponding
We make this more precise by studying Eq. 20 perturba-
state by creating particle-hole excitations in the leads.
corresponds to the relaxation of the higher energy spin
energy spin state where the energy for populating it is sup-
results in an upturn in Fig. 2. The
proaches the equilibrium value of
\[ \frac{1}{T_{eff}^B} = \frac{a_L^2 + a_R^2}{(a_L^2 + a_R^2)(|B| + V)} \] (24)
For the special case of symmetric couplings \( a_L = a_R \)
which corresponds to the case in Fig. 2, and for \( B \ll V \),
one finds \( \frac{1}{2T_{eff}^B} \sim \frac{1}{2} (1 - \frac{2h}{\omega}) \) which captures the initial
downturn in the plot for \( T_{eff} \).

For \( B \gg \Gamma_{neq} \) on the other hand the integrals in Eq. 20
is dominated by \( t \sim 1/B \ll 1/V \). In this regime \( T_{eff} \)
approaches the equilibrium value of \( T_{eff} \rightarrow 0 \) and therefore
results in an upturn in Fig. 2. The \( B \gg V \) behaviour of
the integral \( I(B) \) was found to be \( I(B \gg V) \sim \left( \frac{V}{B} \right)^{\frac{1}{2}} \),
the physical significance of which may be understood as follows.
\( I(B) \) represents the population of the high energy spin state where the energy for populating it is supplied
by the voltage source. The ratio \( n = \frac{B}{V} \) represents the optimal number of bath electrons that can be transmitted
across the voltage source in order to excite the higher energy spin state, while \( I(B) = \left( \frac{V}{B} \right)^{n} \) is simply
the probability for doing so. Plugging this expression for
\( I(B) \) into Eq. 22 the effective temperature is found to approach zero as \( T_{eff}^B \rightarrow \frac{1}{\ln \frac{B}{V}} \) in the regime \( B/V \gg 1 \).

Let us now turn to the discussion of the spin response function itself. The imaginary part of the response function is plotted for different coupling strengths and ratio of \( B/V \) in Fig. 3. The line-shape (main panel: Fig. 3) in addition to having the familiar asymmetric form of an x-ray response function, now has a finite weight at \( |\omega| < |B| \), which is forbidden at zero temperatures in equilibrium. The coupling constant (main panel) and voltage (inset panel) dependence of the broadening is illustrated in Fig. 3. \( \chi''(\omega) \) is linear in \( \omega \) for small \( \omega \), with a slope that is inversely related to the long-time relaxation rate of the density matrix \( \Gamma_{rel} = T_{eff}^B \), while at large frequencies \( \omega \gg B \), \( \chi''(\omega) \sim \frac{1}{\omega^{1 - \frac{B}{\omega}}} \). These two different frequency regimes appear as a change in slope of the plots in the inset of Fig. 3.

In conclusion, we have studied the non-equilibrium ohmic spin-boson model including a non-vanishing level splitting and orthogonality effects exactly. Previous work\(^1\) studied the zero level splitting limit, treating the orthogonality effects perturbatively. Our results agree with previous results in the appropriate limit, but provide significant new information including the non-monotonic effective temperature and the line-shape at non-vanishing level splitting. The calculated spin dynamics reveal that the non-equilibrium regime can be quite complex because of the interplay between various voltage and magnetic field dependent relaxation mechanisms. While departures from equilibrium are qualitatively similar to a non-zero temperature (e.g. permitting sub-threshold absorption of Fig. 3), the analogy cannot be pushed too far. The "fluctuation-dissipation" ratio is not a hyperbolic tangent and indeed is not characterized by a unique effective temperature (c.f. Fig. 1), and the low-frequency effective temperature is itself a non-trivial function of the control parameters (c.f. Fig. 4), and is different depending on the quantity used to evaluate it. In equilibrium, the spin boson and Kondo models are related by the simple mapping \( \frac{\delta^B}{\Gamma_{eff}} \rightarrow \sqrt{2} \left( 1 - \frac{\delta}{\Gamma} \right) \). Our finding that non-equilibrium effects enter into different parameters in different ways suggests that the mapping will not be so simple in the non-equilibrium case. A direction for future research is to extend the analysis in this paper to arbitrary number of spin flip processes, and to perform an Anderson-Yuval-Hamann type renormalization group treatment for the out of equilibrium spin boson and Kondo models\(^6\). This work was supported by NSF DMR-0431350.

1. S. De Franceschi, R. Hanson, W. G. van der Wiel, J. M. Elzerman, J. J. Wijckema, T. Fujisawa, S. Tarucha and L. P. Kouwenhoven, Phys. Rev. Lett., 89, 156801 (2002).
2. C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett., 92, 040403 (2004).
3. G. Grynberg and C. Robilliard, Physics Reports, 355, 335 (2001).
4. A. Kamenev, cond-mat/0412296
5. D. A. Abanin and L. S. Levitov, Phys. Rev. Lett., 94, 186803 (2005); E. Yuzbashyan, Boris L. Altshuler, Vadim

FIG. 3: Main panel: \( \chi''(\omega) \) for two different values of the spin-bath coupling strength \( J_z \) and for \( B = V \). Inset: \( \chi''(\omega) \) on a log-log plot for spin-bath coupling strength corresponding to \( \delta_{eq} = 0.98 \) and for two different degrees of departure from equilibrium.
B. Kuznetsov, Victor Z. Enolskii, cond-mat/0505493 A. Kaminski, Yu. V. Nazarov, and L. I. Glazman, Phys. Rev. B, 62, 8154 (2000).

A. Mitra, I. Aleiner and A. J. Millis, Phys. Rev. Lett., 94, 076404 (2005)

Giovanni Gallavotti, Chaos 14, 680 (2004); F. Zamponi, L. F. Cugliandolo, J. Kurchan, cond-mat/0504750 S. Schuler, T. Speck, C. Tietz, J. Wrachtrup, and U. Seifert, Phys. Rev. Lett., 94, 180602 (2005).

F. Zamponi, G. Ruocco, and L. Angelani Phys. Rev. E, 71, 020101 (R) (2005); Pierre Gaspard, J. Chem. Phys., 120, 8898 (2004).

P. W. Anderson and G. Yuval, Phys. Rev. Lett., 23, 89 (1969).

P. Nozieres and C. T. De Dominicis, Phys. Rev., 178, 1097 (1969).

Suzanne M. Fielding and Peter Sollich, AIP Conf. Proc., 553, 81 (2001).

Tai-Kai Ng, Phys. Rev. B, 54, 5814 (1996); B. Muzykantskii, N. d'Ambrumenil, and B. Braunecker, Phys. Rev. Lett., 91, 266602 (2003).

Grabert and Weiss, Phys. Rev. Lett., 54, 1605 (1985); M. P. A. Fisher and A. T. Dorsey, Phys. Rev. Lett, 54, 1609 (1985).

W. Mao, P. Coleman, C. Hooley, and D. Langreth, Phys. Rev. Lett., 91, 207203 (2003).

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