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Spin dynamics and violation of the fluctuation dissipation theorem in a nonequilibrium 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 that are out of equilibrium due to the application of a bias voltage. Both the nonequilibrium steady state and the rate constants describing the approach to the 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 the response to the 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 in their dependence on parameters, and, in general, are nonmonotonic.

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A fundamental question in quantum condensed matter physics is understanding properties of nonequilibrium many-body systems, some examples being the Kondo effect in quantum dots,[1] ultracold gases with rapidly tunable interactions,[2] and strongly driven optical lattices.[3] While there are a variety of nonperturbative techniques in place to study equilibrium systems, these methods cannot be extended to nonequilibrium systems in a straightforward way.[4] The experimental accessability[1–3] of the nonequilibrium regime of strongly correlated quantum many-body systems gives rise to the need for the further development of the formalism.

Many-body systems driven out of equilibrium are known[5,6] 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 a steady state by the response function $\chi(t-t')$ describing changes induced by weak external probes, and by the correlation function $S(t-t')$ describing the probabilities of observing various configurations of the system. An important and still incompletely understood issue is the manner in which χ and S characterizing a nonequilibrium system differ from those describing an equilibrium one. In particular, there has been considerable interest[7,8] in the possibility of establishing a generalized fluctuation dissipation theorem relating $\chi(\omega)$ to $S(\omega)$ and thereby characterizing the departures from equilibrium in terms of an effective temperature. Several systems have been identified[8] 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 B and tunneling rate Δ, coupled via a coupling J_z to a spinless resonant level (creation operator $d^\dagger$), which is itself connected to two leads (L and R) that may be at different chemical potentials. The Hamiltonian is

$$H = S_z B + \Delta S_x + J_z S_z d^\dagger d + H_{bath}, \tag{1}$$

$$H_{bath} = \sum_{k,\alpha=L,R} \epsilon_k c^\dagger_{k\alpha} c_{k\alpha} + \sum_{k,\alpha=L,R} (t_{k\alpha} c^\dagger_{k\alpha} d + \text{H.c.}). \tag{2}$$

We assume the leads are infinite reservoirs characterized by the correlators $\langle c^\dagger_{k\alpha} c_{q\beta}\rangle = \delta_{kq}\delta_{\alpha\beta}(e^{\beta(\epsilon_k-\mu_\alpha)}+1)^{-1}$. A nonequilibrium 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[6] by $\tan\delta_L = a_L J_z/[1-i\,\text{sgn}(V)a_R J_z]$, $\tan\delta_R = a_R J_z/[1+i\,\text{sgn}(V)a_L J_z]$ with $a_{L,R}=\Gamma_{L,R}/(\Gamma_L+\Gamma_R)^2$, with $\Gamma_{L,R}=\pi\rho t^2_{L,R}$ and $\rho = dk/d\epsilon_k$. We will study the properties of H at $T=0$ but out of equilibrium ($V\neq 0$) working to leading nontrivial order in Δ but to all order in J_z. The conditions under which the perturbation theory is justified will be discussed below. We will find the time scales characterizing the approach to the 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

$$\frac{d\rho(t)}{dt} = -i[H,\rho(t)], \tag{3}$$

from which the density matrix ρ_S for the local spin is obtained from taking a trace over the electronic degrees of freedom,

$$\hat{\rho}_S = \text{Tr}_{el}\rho. \tag{4}$$

We adopt a spin language, writing

$$\hat{\rho}_S = \frac{1}{2}(1 + S_z \hat{\sigma}_z) \tag{5}$$

and study S_z. When $\Delta=0$, the Hamiltonian is exactly solvable both in and out of equilibrium. For nonzero Δ one may expand Eq. (3) perturbatively in Δ. The key object in the analysis is the time-evolution operator separating two spin-flip processes,[9] $K_\pm(t)=\text{Tr}_{el}[e^{-iH(\Delta=0,S_z=\pm1/2)t}e^{iH(\Delta=0,S_z=(\mp1)/2)t}] = e^{\pm iBt}e^{-C_\pm(t)}$, where $C_+(t)=C(t)=[C_-(-t)]^*$ computed from the linked cluster theorem has the following form at zero temperature:[6]

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

$$C'(t) = \left(\frac{\delta_{eq}}{\pi}\right)^2 \ln(\xi t) + \phi'(Vt), \tag{6}$$

$$C'' = \frac{\pi}{2}\left(\frac{\delta_{eq}}{\pi}\right)^2 \mathrm{sgn}(t) + \phi''(Vt). \tag{7}$$

Here ξ is a short time cutoff, $\delta_{eq}=\delta_L+\delta_R=\arctan[J_z/(\Gamma_L+\Gamma_R)]$ is the equilibrium phase shift, and $\phi(Vt)$ is a function describing the crossover from the short-time ($Vt \ll 1$) equilibrium behavior characterized by[10] $C(t)=(\delta_{eq}/\pi)^2[\ln(\xi t)+i(\pi/2)\mathrm{sgn}(t)]$, to the long-time ($Vt \gg 1$) nonequilibrium behavior characterized by[12] $C(t)=[(\delta_L^2+\delta_R^2)/\pi^2][\ln(\xi t)+i(\pi/2)\mathrm{sgn}(t)]+\Gamma_{neq}t$ with $\Gamma_{neq}=V|\delta_L''-\delta_R''|/2\pi$. Correct treatment for the intermediate and short-time behavior of ϕ is essential for obtaining correct results for $\chi(\omega), S(\omega)$. A general analytic expression for ϕ does not exist, here we use perturbation theory to the third order in J_z to obtain,[6]

$$\begin{aligned}\phi(Vt) = {} & \frac{|\delta_L''-\delta_R''|}{2\pi} Vt\left[\left(\frac{2}{\pi}\right)\left(Si(Vt) - \frac{1-\cos(Vt)}{Vt}\right)\right] \\ & - \frac{2\,\mathrm{Re}\;\delta_L\delta_R}{\pi^2}[\gamma_e - Ci(Vt) + \ln(Vt)] \\ & - \frac{2i\;Im\;\delta_L\delta_R}{\pi^2}\left[\frac{2}{\pi}\int_0^1 du\,\sin(uVt) \right. \\ & \left. \times \frac{[(1-u)\ln(1-u) + u\ln u]}{u^2}\right].\end{aligned} \tag{8}$$

Now let us return to the evaluation of various spin observables. In terms of the symmetric and antisymmetric time-evolution operators $K_{s,a}(t)=\mathrm{Re}[K_+(t)\pm K_-(t)]$, the equation of motion for the variable S_z defined in Eq. (5) to leading nontrivial order in Δ is given by

$$\frac{dS_z}{dt} = -\int_0^t dt'[K_s(t,t')S_z(t') + K_a(t,t')]. \tag{9}$$

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

$$\tilde{K}_s(\lambda) = \Delta^2\int_0^\infty dt e^{-\lambda t}e^{-C'(t)}\cos Bt \cos C''(t), \tag{10}$$

$$\tilde{K}_a(\lambda) = \Delta^2\int_0^\infty dt e^{-\lambda t}e^{-C'(t)}\sin Bt \sin C''(t). \tag{11}$$

The above equations capture the effect of two sources of decoherence on spin dynamics; one is a Korringa-type decoherence existing even in equilibrium, while the second arising mathematically from $C'(t)$ is due to a nonzero voltage and is intrinsically nonequilibrium.

The solution to Eq. (9) can be written as[13]

$$S_z(t) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}\frac{d\lambda}{\lambda}e^{\lambda t}\frac{\lambda S_z(0) - \tilde{K}_a(\lambda)}{\lambda + \tilde{K}_s(\lambda)}. \tag{12}$$

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

$$\Gamma_{rel} = \tilde{K}_s(0) = \frac{\pi}{2}\frac{\Delta^2}{\xi}\frac{1}{\Gamma(\alpha)}\frac{\sin\left[\frac{\pi\alpha}{2} + (1-\alpha)\arctan\frac{\Gamma_{neq}}{B}\right]}{\sin\frac{\pi\alpha}{2}} \times \left(\frac{\sqrt{B^2+\Gamma_{neq}^2}}{\xi}\right)^{\alpha-1}, \tag{13}$$

where the nonequilibrium exponent $\alpha=(\delta_L/\pi)^2+(\delta_R/\pi)^2$. The most interesting situation is the relatively small phase shift limit, in which $\Gamma_{neq} \ll V$ and Eq. (13) holds even when $B \gg \Gamma_{neq}$, provided $B \ll V$. For $B \gg V$, one should set $\Gamma_{neq}=0$ in Eq. (13) and replace the nonequilibrium exponent α by the equilibrium exponent $\alpha_{eq}=(\delta_{eq}/\pi)^2$, yielding the familiar $T=0$ Korringa relaxation.

The above results of an exponential relaxation to a steady state are obtained from neglecting the λ dependence of $K_s(\lambda)$, which is justified when

$$\frac{\Gamma_{rel}}{\sqrt{\Gamma_{neq}^2+B^2}} \sim \frac{\Delta^2}{\xi^2}\left(\frac{\sqrt{B^2+\Gamma_{neq}^2}}{\xi}\right)^{\alpha-2} \ll 1, \tag{14}$$

and therefore holds in the perturbative in Δ regime, provided the voltage or the magnetic field is not too small. An analysis similar to the equilibrium case shows that Eq. (14) is also the condition for validity of perturbation theory in Δ.

At steady state and in the small Δ 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:

$$\rho = \rho_S \otimes \rho_{el} = \begin{pmatrix}\rho_\uparrow\rho_{el}^{\uparrow\uparrow} & 0 \\ 0 & \rho_\downarrow\rho_{el}^{\downarrow\downarrow}\end{pmatrix}, \tag{15}$$

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

The correlation function we study is,

$$S_{xx}(t_1,t_2) = i\langle\{S_x(t_1),S_x(t_2)\}_+\rangle = \mathrm{Tr}[\rho\{S_x(t_1),S_x(t_2)\}_+], \tag{16}$$

and the corresponding spin response function is

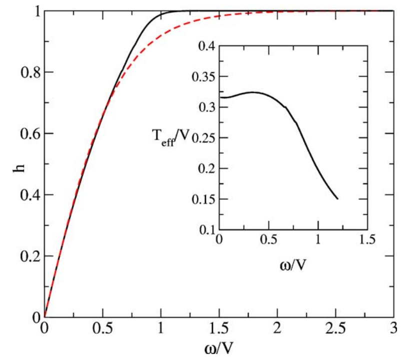

FIG. 1. (Color online) Main panel. Solid line: fluctuation-dissipation ratio (Eq. (21)) at $B=0.5V$. Dashed line: "pseudoequilibrium" result $\tanh(\omega/2T^h_{eff})$ (Eq. (22)). Inset: Plot of $T_{eff}(\omega,V)=\omega/2\tanh^{-1}h$, which shows rapid crossover from non-equilibrium behavior ($T_{eff}\sim V$) at $\omega<V$ to equilibrium behavior ($T_{eff}=0$) at $\omega>V$.

$$\chi_{xx}(t_1,t_2)=-i\theta(t_1-t_2)\langle[S_x(t_1),S_x(t_2)]\rangle$$
$$=-i\theta(t_1-t_2)\mathrm{Tr}\{\rho[S_x(t_1),S_x(t_2)]\}, \qquad (17)$$

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

$$\chi''_{xx}(\omega)=\rho_\uparrow[I(B+\omega)-I(B-\omega)]$$
$$-\rho_\downarrow[I(-B-\omega)-I(-B+\omega)], \qquad (18)$$

$$-iS_{xx}(\omega)=\rho_\uparrow[I(B+\omega)+I(B-\omega)]$$
$$+\rho_\downarrow[I(-B-\omega)+I(-B+\omega)], \qquad (19)$$

where

$$I(B)=\mathrm{Re}\left[\int_0^\infty dte^{iBt}e^{-C(t)}\right], \qquad (20)$$

and $\rho_\uparrow/\rho_\downarrow=I(-B)/I(B)$. We define the fluctuation dissipation ratio (also mentioned in Ref. 14),

$$h(\omega)=\frac{\chi''(\omega)}{iS_{xx}(\omega)}. \qquad (21)$$

In equilibrium and at $T>0$ (when $\phi(0)=1$ and $C(t)=(\delta_{eq}/\pi)\ln[(\xi/\pi T)\sinh\pi tT]$), $h(\omega)=\tanh\omega/2T$.

Out of equilibrium (and at $T=0$), $h(\omega)$ has the form shown in Fig. 1, which differs from the equilibrium solution $\mathrm{sgn}(\omega)$. The calculated $h(\omega)$ is not a tanh function (compare the 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 $T_{eff}(\omega)=\omega/2\tanh^{-1}h$. This function is plotted in the inset of Fig. 1 and is seen to have a strong ω dependence and indeed not to be monotonic. For $\omega<V/2$, T_{eff} is of the order of V and depends weakly on ω. For $\omega>V/2$, T_{eff} drops sharply and at high ω approaches the equilibrium value (here, $T=0$). The results presented in Fig. 1 show that no unique definition of "nonequilibrium effective temperature" exists; the value obtained depends on the quantity examined. Two obvious definitions are (i) from the $\omega\to0$ limit of $h(\omega)$,[11] (ii) from the population ratio $\rho_\uparrow/\rho_\downarrow$. The effective temperature from definition (i) is obtained by expanding Eqs. (18) and (19) for small ω,

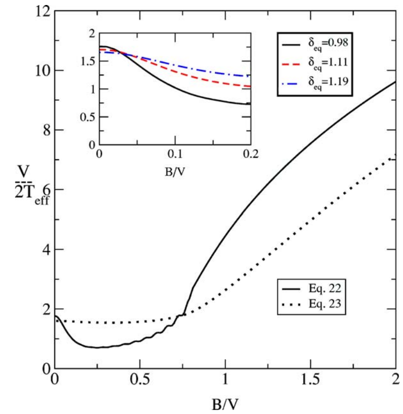

FIG. 2. (Color online) Main panel: Low-frequency effective temperature defined from $h(\omega\to0)$ (solid line) and population ratios (dashed line). Note: Nonmontonic behavior with the magnetic field. Inset: The effective temperature [from Eq. (22)] as a function of the coupling constant. Note: T^h increases weakly with δ_{eq} for $B\to0$, but decreases strongly with δ_{eq} for large B.

$$\frac{1}{T^h_{eff}}=\left.\frac{\partial h(\omega)}{\partial\omega}\right|_{\omega=0}=\sum_{\sigma=\pm}\left.\frac{\partial\ln I(x)}{\partial x}\right|_{x=\sigma B}, \qquad (22)$$

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

$$\frac{1}{T^\rho_{eff}}=\frac{1}{B}\ln\frac{\rho_\downarrow}{\rho_\uparrow}. \qquad (23)$$

Figure 2 shows the dependence of these two measures of effective temperature on the magnetic field. We see that the two curves differ in magnitude and in dependence on parameters; the variation in general is nonmonotonic. The inset shows that the magnitude and field variation of the effective temperature T^h_{eff} also depends on the coupling constant.

The nonmonotonic behavior as a function of B/V may be understood as follows. For $B\ll\Gamma_{neq}$ and for $(\delta''_L-\delta''_R)/2\pi\ll1$ so that $\Gamma_{neq}\ll V$, the integrand in Eq. (20) is dominated by $t\sim1/\Gamma_{neq}\gg1/V$. In this regime, Eq. (13) applies; from this one sees that the decoherence rate for the spin increases with the magnetic field, causing the initial downturn in Fig. 2. This behavior may also be understood as originating from the opening up of an additional scattering channel on application of a magnetic field that corresponds to the relaxation of the higher energy spin state by creating particle-hole excitations in the leads. We make this more precise by studying Eq. (20) perturbatively in tunneling amplitude ($t_{L,R}$) to find

$$\frac{1}{T^h_{eff}}=\frac{a_L^2+a_R^2+a_La_R}{(a_L^2+a_R^2)|B|+a_La_R(|B|+V)}-\frac{1}{|B|-V}. \qquad (24)$$

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 $1/2T^h_{eff}\sim2/V(1-2B/V)$, which captures the initial downturn

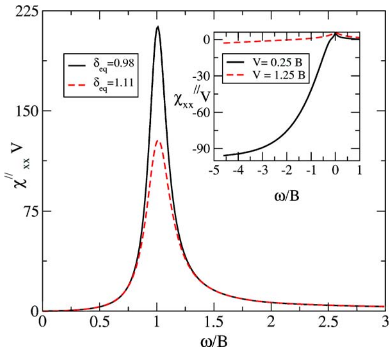

FIG. 3. (Color online) Main panel: $\chi''(\omega)$ for two different values of the spin-bath coupling strength J_z and for $B=V$. Inset: $\chi''(\omega)$ on a log-log plot for the spin-bath coupling strength corresponding to $\delta_{eq}=0.98$ and for two different degrees of departure from equilibrium.

in the plot for T_{eff}.For $B \gg \Gamma_{neq}$, on the other hand, the integrals in Eq. (20) are dominated by $t \sim 1/B \ll 1/V$. In this regime T_{eff} approaches the equilibrium value of $T_{eff} \to 0$ and therefore results in an upturn in Fig. 2. The $B \gg V$ behavior of the integral $I(B)$ was found[6] to be $I(B \gg V) \sim (V/B)^{B/V}$, 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 $n=B/V$ 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 $I(B)=(V/B)^n$ 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 $T^h_{eff} \to V/\ln(B/V)$ 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,[12] now has a non-zero 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 ω for small ω, with a slope that is inversely related to the long-time relaxation rate of the density matrix $\Gamma_{rel}=T^h_{eff}$, while at large frequencies $\omega \gg B$, $\chi''(\omega) \sim 1/\omega^{1-(\delta_{eq}/\pi)^2}$. 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 nonequilibrium ohmic spin-boson model including a nonvanishing level splitting and orthogonality effects exactly. Previous work[14] studied the zero level splitting limit, treating the orthogonality effects perturbatively. Our results agree with previous results in the appropriate limit, but also provide new information, including the nonmonotonic effective temperature and the line shape at nonvanishing level splitting. The calculated spin dynamics reveal that the nonequilibrium 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 nonzero temperature (e.g., permitting subthreshold absorption seen in Fig. 3), the analogy cannot be pushed too far. The "fluctuation dissipation" ratio is not a hyperbolic tangent and is not characterized by a unique effective temperature (c.f. Fig. 1), and the low-frequency effective temperature is itself a nontrivial function of the control parameters (c.f. Fig. 2), 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 $\delta_{eq}/\pi \to \sqrt{2}(1-\delta_{eq}/\pi)$. Our finding that nonequilibrium effects enter into different parameters in different ways suggests that the mapping will not be so simple in the nonequilibrium case. A direction for future research is to extend the analysis in this paper to an 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.[15]

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