Dynamics of a Tunneling Magnetic Impurity: Kondo Effect Induced Incoherence

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We study how the formation of the Kondo compensation cloud influences the dynamical properties of a magnetic impurity that tunnels between two positions in a metal. The Kondo effect dynamically generates a strong tunneling impurity-conduction electron coupling, changes the temperature dependence of the tunneling rate, and may ultimately result in the destruction of the coherent motion of the particle at zero temperature. We find an interesting two-channel Kondo fixed point as well for a vanishing overlap between the electronic states that screen the magnetic impurity. We propose experiments where the predicted features could be observed.

Introduction.—Tunneling of a heavy particle or some collective degree of freedom in a dissipative environment has been the subject of intense theoretical and experimental research in the past and is by now reasonably well understood [1–7]. Maybe the most intriguing case is that of *Ohmic* dissipation, where the particle usually couples to electron-hole excitations of a metallic environment. In this case the bare tunneling amplitude Δ_0 of the particle is strongly renormalized due to the dissipative environment, and becomes temperature dependent. In the simplest scenario, where the tunneling occurs between two sites (two level system, TLS), the effective tunneling displays a power-law behavior over a wide range of temperatures, $\Delta(T) \sim \Delta_0 (T/\omega_0)^\alpha$, with ω_0 a high-energy cutoff of the order of the Debye frequency, and α a dimensionless coupling constant [1,2]. The renormalization of the tunneling amplitude is a consequence of Anderson's orthogonality catastrophe [8]: At any position the presence of the particle generates a screening cloud that consists of an *infinite* number of electron-hole excitations. The formation of this huge "electronic polaron-cloud" slows down the particle, increases its mass, and thus decreases its tunneling amplitude. Depending on the specific value of the coupling α, the dynamics of the particle can be of three different kinds: (a) If the coupling is small the particle moves with slightly damped *coherent* oscillations between the two positions at $T = 0$. (b) For $1/2 < \alpha < 1$ the motion of the particle becomes *incoherent* while for even larger couplings (c) the particle becomes localized and cannot move from one well to the other.

Here we discuss the very interesting but poorly understood case of a tunneling *magnetic impurity* coupled to an Ohmic environment [9,10]. Possible examples of such a system include a magnetic impurity tunneling between a scanning tunneling microscope (STM) tip and a metallic surface [11], a Kondo impurity in an amorphous region [12], a spin-1/2 quantum soliton interacting with a metallic environment [13], charge tunneling in double quantum-dot systems [14,15], or a Kondo impurity on a metallic surface. In all these cases the spin of a magnetic impurity couples through an exchange interaction to the local spin density fluctuations of the conduction electrons, providing an Ohmic dissipative environment. Unlike the usual Caldeira-Leggett model, this coupling becomes renormalized due to many-body effects as the temperature is lowered, and leads to the dynamical formation of a Kondo compensation cloud below the Kondo temperature T_K. Since the impurity has to drag the compensation cloud with itself, this results in an increased dissipation and a renormalized tunneling amplitude below T_K. In the present paper we study this interplay between the magnetic Kondo effect and the orbital motion of the TLS. Electron assisted processes involving simultaneous tunneling and electron scattering have a negligible effect compared to direct tunneling [16]. Therefore, in contrast to Refs. [9,10] we neglect them.

Model.—We consider the simplest possible case, where tunneling takes place between two positions only, $\mathbf{R}_\pm$. Furthermore, though we also discuss the role of asymmetry to some extent, we mostly focus on spatially symmetrical TLS's. We show, in particular, that the Kondo effect associated with the magnetic degrees of freedom leads to a strong *temperature dependence* of the exponent α, and may eventually induce an incoherent state. Destroying the Kondo cloud with a magnetic field, one can decrease the dissipation, increase the tunneling rate, and eventually drive the particle back to the coherent regime. We also find that under very special circumstances the orbital motion may lead to the appearance of a *two-channel Kondo* (2CK) state where the impurity tunnels very fast back and forth and forms a Kondo state with the conduction electrons at *both* positions. This new type of 2CK fixed point appears in the *real spin sector* and has nothing to do with the orbital Kondo effect [17] debated in Ref. [16].

We describe the TLS by the tunneling Hamiltonian

$$H_{\text{tun}} = -(\Delta_0 T^x + \Delta_z T^z), \tag{1}$$

where the two pseudospin states $T^z = \pm 1/2$ correspond to the two tunneling positions, Δ_0 is the tunneling matrix element, and Δ_z describes the asymmetry of the TLS.

We assume that the tunneling particle interacts with the local electronic spin density only at its actual position:

$$H_{\text{int}} = \sum_{q=\pm} J_q P_q \vec{S} (\Psi_q^\dagger \vec{\sigma} \Psi_q). \qquad (2)$$

Here $P_\pm = 1/2(1 \pm 2T^z)$ projects out the TLS states at positions $\mathbf{R}_\pm$, $J_\pm$ is the exchange coupling at these positions, $\vec{S}$ denotes the spin operator of the impurity, and $\vec{\sigma}$ stands for the Pauli matrices. The field operators, $\Psi_{\pm,\mu}^\dagger = \int e^{i\mathbf{kR}_\pm} c_{\mathbf{k}\mu}^\dagger d^3\mathbf{k}/(2\pi)^3$, create conduction electrons at $\mathbf{R}_\pm$ with spin μ. Including electron-assisted processes in Eq. (2) has not changed our results [14].

Consider the case of a symmetric TLS with $J_- = J_+ = J$ and $\Delta_z = 0$. The relevant conduction electron degrees of freedom can be represented simply by one-dimensional fermion fields [18,19], $c_{p\alpha\mu}$, obeying canonical anticommutation relations, $\{c_{p\alpha\mu}, c_{p'\alpha'\mu'}^\dagger\} = 2\pi\delta(p - p')\delta_{\alpha\alpha'}\delta_{\mu\mu'}$:

$$H_{\text{el}} = \sum_{\alpha=e,o} \sum_{\mu=\uparrow,\downarrow} \int_{-k_F}^{k_F} \frac{dp}{2\pi} v_F p c_{p\alpha\mu}^\dagger c_{p\alpha\mu}. \qquad (3)$$

The radial momentum p is measured from the Fermi momentum k_F, v_F is the Fermi velocity ($v_F = k_F = \hbar = 1$), $\alpha = \{e, o\}$ is the parity, and μ denotes the spin. In terms of the fields $\Psi_{\alpha,\mu} \equiv \int_{-k_F}^{k_F} c_{p\alpha\mu} dp/\sqrt{2\pi}$, Eq. (2) can be rewritten as

$$H_{\text{int}} = \frac{g}{2} \vec{S}(1 + F)\Psi_e^\dagger \vec{\sigma} \Psi_e + \frac{g}{2} \vec{S}(1 - F)\Psi_o^\dagger \vec{\sigma} \Psi_o$$
$$+ g\vec{S}\sqrt{1 - F^2} T^z(\Psi_e^\dagger \vec{\sigma} \Psi_o + \Psi_o^\dagger \vec{\sigma} \Psi_e). \qquad (4)$$

Here $F = \sin(k_F d)/k_F d$ measures the overlap of the states $\Psi_\pm$, with $d = |\mathbf{R}_+ - \mathbf{R}_-|$ the tunneling distance, and $g = Jk_F^2/2\pi^2$. For $d = 0$ this Hamiltonian obviously reduces to the single-channel Kondo model.

Numerical Renormalization Group.—We used Wilson's numerical renormalization group (NRG) [20] to determine the effective temperature-dependent tunneling amplitude $\Delta(T)$ and the $T = 0$ impurity spin and pseudospin spectral functions $\varrho_\mathcal{O}^i = -\text{Im}\{\chi_\mathcal{O}^i(\omega)\}$, (with $\chi_\mathcal{O}^i(\omega)$ the Fourier transform of the retarded response function, $O = \{T, S\}$.) In this technique one constructs a series of Hamiltonians, H_N, which are diagonalized iteratively. Having obtained the many-body eigenstates and energies of H_N one can calculate physical quantities at an energy scale $T, \omega \sim \omega_N \sim \Lambda^{-(N+1)/2}$, with $\Lambda \approx 3$ a discretization parameter. Our results were obtained by keeping the lowest 250 states in each iteration. To obtain accurate results we exploited invariance under (i) parity (ii) global spin rotations, and (iii) a hidden SU(2) symmetry, related to electron-hole symmetry [21]. [For calculations in the presence of magnetic field we used only the U(1) component of the two SU(2) symmetries, and kept ~700 states.]
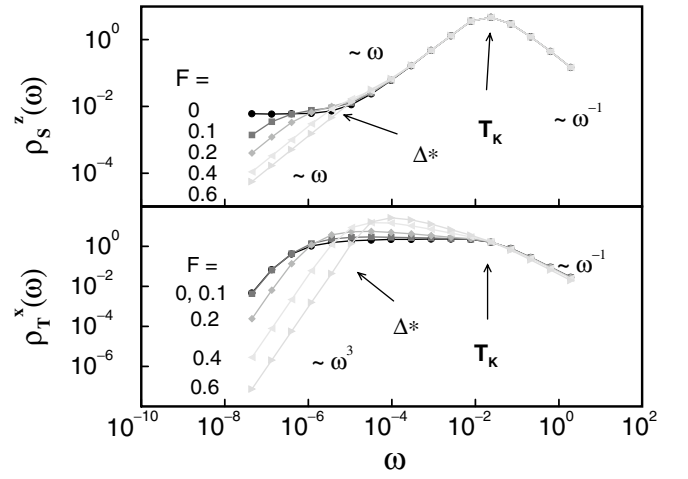

FIG. 1. Logarithmic plot of various spectral functions discussed in the text for $\Lambda = 3$, $g = 0.144$, and $\Delta_0 = 2.31 \times 10^{-5}$. The energy scales T_K and Δ^* are also indicated.

Results.—The spectral functions ϱ_S and ϱ_T^x are shown in Fig. 1. In the somewhat peculiar case of $F = 0$, where Ψ_+ and Ψ_- do not overlap, we can observe two distinct crossovers: The first takes place at the Kondo energy $T_K \approx e^{-1/2g}$ and corresponds to the formation of a Kondo state at the *actual* position of the TLS. Above T_K all spectral functions behave as $1/\omega$, indicating that all correlation functions are constant for times shorter than $1/T_K$. Below T_K the spin spectral function becomes linear corresponding to a Fermi liquid impurity susceptibility $\sim 1/T_K$. The logarithmic slope of ϱ_T^x changes at T_K: This change is related to the dynamical renormalization of the tunneling amplitude by the formation of the Kondo compensation cloud.

As also confirmed by a detailed analysis of the finite size spectrum [14], at time scales $\sim 1/T_K \ll 1/\Delta_0$ tunneling events are very rare, and the particle is essentially immobile. However, Δ_0 is a relevant perturbation, and leads to a second crossover at a renormalized tunneling amplitude Δ^*, where the TLS freezes into the even tunneling state.

For $F = 0$ a two-channel Kondo state is formed below Δ^*, as confirmed by the analysis of the finite size spectra [14]. This is most easily understood by observing that the last term of Eq. (4) flips the TLS between the $T^x = \pm 1/2$ states, and can therefore be dropped below Δ^*. Then H_{int} becomes simply the two-channel Kondo Hamiltonian with couplings $g_{e/o} = g(1 \pm F)$, and for $F = 0$ a two-channel Kondo state is formed in the spin sector [22]. The spin spectral function becomes constant below Δ^*, implying the logarithmic divergence of the spin-susceptibility, $\chi_S(T < \Delta^*) \sim \Delta^* \ln(T_K/T)/T_K^2$. The external magnetic field and asymmetry are both relevant operators at this two-channel Kondo fixed point [23], which is thus extremely unstable, and probably rather difficult to access experimentally. For any finite overlap $g_e \neq g_o$, a third crossover occurs to a Fermi liquid state at an energy $T^* < \Delta^*$. For generic F's this second crossover takes place almost simultaneously
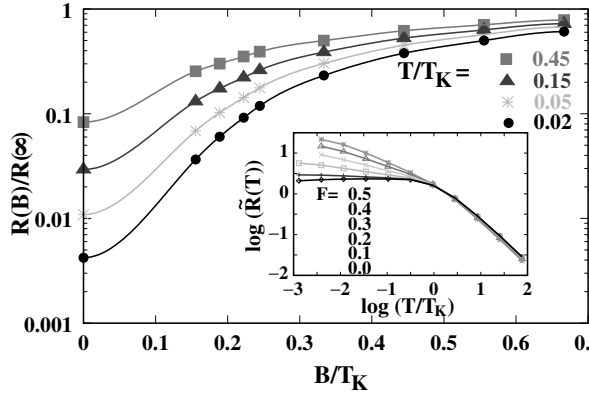
FIG. 2. Magnetic field dependence of the tunneling rate for $F = 0.2$ and several values of T/T_K. Inset: Temperature dependence of the normalized tunneling rate $\tilde{R}(T) = R(T)/\Delta_0^2$. F decreases from top to bottom. We used $\Lambda = 3$, $g = 0.144$, and $\Delta_0 = 2.31 \times 10^{-5}$ in both figures.

with the crossover at Δ^* and only a small kink remains from the two-channel Kondo behavior at $F = 0$.

While the spectral functions help us to understand the behavior of the model, experimentally it is next to impossible to measure them. Quantities of real interest are the effective tunneling amplitude at temperature T, $\Delta(T)$, and the tunneling rate $R(T)$. We determined $\Delta(T)$ directly from the Δ_0 induced ground state splitting and also indirectly from the spectral function $\varrho_T^x(\omega \sim T)$ using scaling arguments, with identical results [14]. The temperature dependent tunneling rate [1], $R(T) \sim \Delta(T)^2/T$, calculated in this way is shown in the inset of Fig. 2. It can be experimentally determined by performing real-time measurements [7]. As the most striking consequence of the Kondo effect, the logarithmic slope of $R(T)$ changes at $T \approx T_K$.

In some experiments it is difficult to change the external temperature. However, one can suppress the Kondo effect by applying an external magnetic field $H_{\text{tun}} \rightarrow H_{\text{tun}} - BS^z$, and thereby increasing $R(T)$ by several orders of magnitude, as shown in Fig. 2.

Another quantity of theoretical interest is the *effective dissipation strength* α, that we can define as

$$\frac{d \ln \Delta(\omega)}{d \ln \omega} = \alpha(g, \omega, F), \qquad [\Delta(\omega_0) = \Delta_0]. \quad (5)$$

In the regime of interest $\omega \gg \Delta^*$, one can show that α is a universal function of ω/T_K, and F, that can be also related to the logarithmic derivative of ϱ_T^x as [14] $\alpha(\omega) \approx \frac{1}{2}(\frac{d \ln \varrho_T^x}{d \ln \omega} + 1)$. For large frequencies $\alpha \approx 0$, meaning that the tunneling amplitude remains unrenormalized above T_K (see Fig. 3a). Below T_K, on the other hand, the dissipation strength scales to an overlap-dependent constant, $\alpha_<$, that coincides with the Anderson orthogonality exponent $K \equiv 2(\frac{1}{\pi} \text{atan}\sqrt{\frac{1}{F^2} - 1})^2$ for a maximally strong scatterer with a phase shift $\delta = \pi/2$ (see inset) [24].

The physical picture behind this is as follows: It takes about a time of $\sim 1/T_K$ to build up the Kondo compensation cloud at the impurity's actual position. Therefore the motion of the impurity is essentially decoupled from the heat bath at energy scales $\omega > T_K$. However, once formed, this compensation cloud acts as a maximally strong potential scatterer in agreement with Nozières' Fermi liquid picture [25], and leads to a strong dissipation.

The energy scale Δ^* is determined by the condition $\Delta(\omega = \Delta^*) \approx \Delta^*$ [1], leading to the expression

$$\Delta^* = \Delta_0 \left(C \frac{\Delta_0}{T_K} \right)^{\alpha_</(1-\alpha_<)}, \quad (6)$$

with C a constant of the order of unity. The constant $\alpha_<$ also characterizes the dissipative nonequilibrium dynamics of the TLS below T_K [1]. In Fig. 3 we show the rescaled spectral function $\Delta^{*2}\varrho_T^z(\omega)/\omega$, related to the real part of the retarded response function. Without dissipation, $\alpha_< = 0$ ($F = 1$), the tunneling of the TLS is entirely coherent: The TLS oscillates between the two positions without damping, and the spectral function consists of two Dirac delta's. For $F < 1$ the coherence peak broadens: The oscillations become exponentially damped and at very long time scales (at $T = 0$) the correlation function behaves as $\langle T^z(t)T^z(0)\rangle \sim 1/t^2$. For even smaller values of overlap the peak becomes completely invisible, implying that the formation of the Kondo compensation cloud suppresses the coherent oscillations.
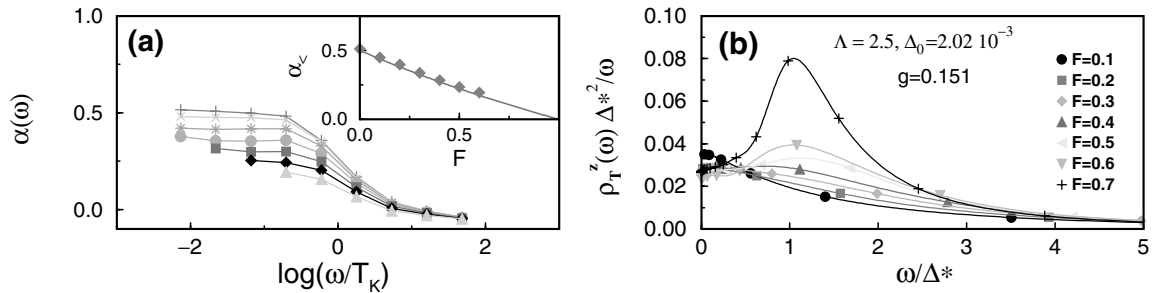

FIG. 3. (a) Energy and overlap dependence of the anomalous dimension α of the dimensionless tunneling amplitude for $F = 0$, 0.1, 0.2 0.3, 0.4, 0.5, and 0.6 (top to bottom), as determined from the logarithmic derivative of the spectral function ϱ_T^x. (b) Rescaled spectral functions and the evolution of the coherence peak with increasing overlap. We computed Δ^* from Eq. (6) with $C = 0.0916$ and $T_K = e^{-1/2g}$.

Realistic situations.—In general, the TLS model is more complex than the one we discussed until now. The TLS may not be symmetrical, $\Delta_z \neq 0$, and the couplings $J_\pm$ in Eq. (2) may not be equal. The difference between $J_\pm$ leads to *two* subsequent Kondo effects at T_K's associated with the two positions, and consequently changes in the logarithmic slope of $R(T)$ twice, while a finite Δ_z generates a new energy scale, below which the TLS freezes into one of the states $T_z = \pm 1/2$ [14].

In Eq. (2) we only took into account the exchange interaction. In reality, the TLS and the electrons interact through a local potential scattering as well. However, for a Kondo impurity this potential scattering is relatively small ($\sim J$) in the exchange scattering-channel and since it remains essentially unrenormalized, its effect can be neglected compared to that of the exchange interaction. Potential scattering in *other* scattering channels may, however, be still present and shift α by a temperature-independent value $\alpha(T) \rightarrow \alpha(T) + \alpha_>$. Therefore, for small overlaps the Kondo effect may even drive the TLS from a coherent ground state to an incoherent state with $\alpha > 1/2$ (see Fig. 3a). The value of $\alpha_<$ can also be considerably larger for spin $S > 1/2$ impurities [14].

Possible experiments.—An interesting experimental realization is provided by a Kondo impurity on a metallic surface. In a generic situation, the Kondo impurity is too heavy, and the barrier height and distance between two neighboring lattice positions are too large to obtain reasonable tunneling probabilities. However, one can actually *tune* the barrier height by placing an STM tip above the impurity and applying a voltage on it. Approximating the barrier shape by a sine function, we can estimate the tunneling rate. Assuming a tunneling distance of $d = 1.47$ Å, corresponding to tunneling on a 111 Cu surface, taking the mass of Co as an example, and an attempt frequency of $\omega_0 \sim 100$ K, we find that a tunneling rate of $R \sim 1$ Hz corresponds to $\Delta_0 \sim 0.710^{-5}$ K and a barrier height of $V_0 \approx 120$ K. With these parameters the motion of the particle is entirely dominated by quantum tunneling below $T^* \sim 4$ K.

Therefore, if one gradually decreases the barrier height at $T = 1$ K until the impurity starts to hop between neighboring positions, one is safely in the tunneling regime, where our theory applies. One could monitor the motion of the magnetic impurity by placing it on a nanowire, and extract the tunneling rate from the time dependent conductance fluctuations, just as in point contact experiments [7,12]. In an STM experiment it is rather difficult to change the temperature, however, for a suitable magnetic impurity with a surface Kondo temperature of $T_K < 10$ K one can destroy the Kondo effect by applying a magnetic field, and thereby increase the tunneling rate as shown in Fig. 2.

Our discussion can be easily generalized to the case of a spinless TLS that happens to be close to a Kondo impurity. In this case Friedel oscillations generated by the TLS modify the local density of states at the Kondo impurity and thus the exchange coupling will depend on the position of the TLS, leading to a Hamiltonian similar to Eq. (2). In this case we predict that formation of the Kondo state on the magnetic impurity may strongly suppress the tunneling rate of the TLS. This effect could be measured performing real time measurements on magnetically doped disordered point contacts [7,12], where both the magnetic field and temperature dependence could be tested.

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