Out-of-Equilibrium Kondo Effect: Response to Pulsed Fields

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The current in response to a rectangular pulsed bias potential is calculated exactly for a special point in the parameter space of the nonequilibrium Kondo model. Our simple analytical solution shows the all essential features predicted by the non-crossing approximation, including a hierarchy of time scales for the rise, saturation, and fall-off of the current; current oscillations with a frequency of \( eV/h \); and the instantaneous reversal of the fall-off current for certain pulse lengths. Rich interference patterns are found for a nonzero magnetic field (either dc or pulsed), with several underlying time scales. These features should be observable in ultra small quantum dots.

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The recent observation of the Kondo effect in ultra small quantum dots has focused considerable attention on the Kondo effect in mesoscopic systems. Acting as tunable Anderson impurities, quantum-dot devices offer an outstanding opportunity to test some of the basic notions of the Anderson impurity model. For example, the crossover from the local-moment to the mixed-valent and empty-impurity regimes, the \( \pi/2 \) phase shift associated with the Kondo effect, and the evolution of the Kondo effect with dc and ac bias potentials. Most recently, an intriguing scenario for the response of a quantum dot to a rectangular pulsed bias potential was put forward by Plihal et al. Using a time-dependent version of the non-crossing approximation, these authors predicted a hierarchy of time scales for the rise, saturation, and fall-off of the current through the dot, featuring a rich interplay between the Kondo temperature, the applied voltage bias, and the temperature. For larger values of the pulsed bias \( eV \), oscillations were predicted in the time-dependent current, with a characteristic frequency of \( \omega = eV/h \). These current oscillations were attributed to the sharp excitation frequency between the two split Kondo peaks induced by the \( eV \) bias potential.

In this paper, we exploit an exactly solvable nonequilibrium Kondo model to obtain a simple analytic description of the Kondo-assisted current in response to rectangular pulsed fields. Our solution captures all the essential features reported by Plihal et al., including the hierarchy of time scales for the rise, saturation, and fall-off of the current; the \( eV/h \) oscillations in the time-dependent current; and the instantaneous reversal of the current for certain pulse lengths after the pulse has ended. Furthermore, we are able to include also a nonzero magnetic field which itself can be pulsed, revealing complex interference patterns between the bias and magnetic field. Most importantly, since our solution is analytic and exact, it provides a benchmark result for the response of a quantum dot to pulsed fields.

We begin with a few general remarks on the solvable model used. An extension of the Toulouse and Emery-Kivelson solution of the equilibrium Kondo problem to nonequilibrium, this model is expected to correctly describe the strong-coupling regime of the nonequilibrium Kondo effect. Indeed, previous applications of the model to dc and ac bias potentials have shown all the qualitative features of Kondo-assisted tunneling: A zero-bias anomaly that splits in an applied magnetic field; Fermi-liquid characteristics in the low-\( T \) and low-\( V \) differential conductance; Side peaks in the differential conductance at \( eV = \pm n\hbar\omega \), for an ac drive of frequency \( \omega \). At the same time, since the solvable point involves strong couplings, this model is incapable of describing weak-coupling features such as the logarithmic temperature dependence of the conductance at elevated \( T \).

Introducing the actual physical system, it consists of two noninteracting leads of spin-1/2 electrons, interacting via a spin-exchange coupling with a spin-1/2 impurity moment \( \vec{\tau} \) placed in between the two leads. The impurity moment can represent either an actual magnetic impurity or a quantum dot with a single unpaired electron. Each lead is subject to a separate time-dependent potential, such that a time-dependent voltage bias forms across the junction. Since the interaction with the impurity is local in space (s-wave scattering), one can reduce the conduction-electron degrees of freedom that couple to the impurity to one-dimensional fields \( \psi_{\alpha\sigma}(x) \), where \( \alpha = R, L \) labels the lead (right or left) and \( \sigma = \uparrow, \downarrow \) specifies the spin orientations. In terms of these one-dimensional fields, coupling to the impurity takes place via the local spin densities \( s_{\alpha\beta} = \frac{1}{2} \sum_{\sigma,\sigma'} \psi_{\alpha\sigma}^\dagger(0)\sigma_{\sigma\sigma'}\psi_{\beta\sigma'}(0) \).

As discussed in detail in Refs. 9 and 11, the exactly solvable model corresponds to a particular choice of the spin-exchange couplings, described by the Hamiltonian

\[
\mathcal{H} = \mathcal{H}_0 - \sum_{\alpha=L,R} eV_{\alpha}(t)\hat{N}_{\alpha} - \mu_B g_i H(t)\tau^z + \sum_{\alpha,\beta=L,R} \alpha,\beta \in L, R)
\]

\[
\mathcal{J}_{\perp}^{\alpha\beta} \left\{ s_{\alpha\beta}^x \tau^x + s_{\alpha\beta}^y \tau^y \right\},
\]

(1)
\[ \mathcal{H}_0 = \hbar v_F \sum_{\alpha=L,R} \sum_{\sigma=\uparrow,\downarrow} \int_{-\infty}^{\infty} \psi_{\alpha \sigma}^\dagger(x) \frac{\partial}{\partial x} \psi_{\alpha \sigma}(x) dx + J^z (s^z_{LL} + s^z_{RR}) \tau^z. \]

(2)

Here the inter-lead longitudinal couplings have been set equal to zero, while the intra-lead longitudinal couplings take the value \( J^z = 2\pi \hbar v_F \). The three transverse couplings \( J^{LL}, J^{RR}, \) and \( J^{LR} = J^{RL} \) are arbitrary. \( \hat{N}_a \) and \( V_a(t) \) in the second term of Eq. (1) are the number operator and time-dependent potential on lead \( \alpha \), respectively, while \( H(t) \) is a local magnetic field acting on the impurity spin.

The basic approach we take is identical to that of Ref. [1] where the same system was solved for ac drives. The first step is to convert the interacting nonequilibrium problem to a noninteracting one. To this end, we first decompose the Hamiltonian into an unperturbed part and a perturbation, where the unperturbed part consists of \( \mathcal{H}_0 \) and the time-dependent voltage \( V_a(t) \). The initial density matrix — characterizing the distribution of the unperturbed system before tunneling has been switched on — is equal to \( \rho_0 = e^{-\beta \mathcal{H}_0} / \text{Tr} \{ e^{-\beta \mathcal{H}_0} \} \).

The Hamiltonian, its unperturbed part, and the initial density matrix are then converted to quadratic forms via the following sequence of steps: (i) Bosonizing the fermion fields; (ii) Converting to new boson fields corresponding to collective charge (c), spin (s), flavor (f), and spin-flavor (sf) excitations; (iii) Performing a canonical transformation; (iv) Refermionizing the new boson fields by introducing four new fermion fields, \( \psi_{\mu}(x) \) with \( \mu = c, s, f, sf; \) and (v) Representing the impurity spin (which has been mixed by the canonical transformation with the conduction-electron spin degrees of freedom) in terms of two Majorana fermions: \( \hat{\alpha} = -\sqrt{2} \tau_y \) and \( \hat{b} = -\sqrt{2} \tau_x \). For \( J^z = 2\pi \hbar v_F \), one thus arrives at the following representation of the Hamiltonian:

\[
\mathcal{H}' = \sum_{\mu=c,s,f,sf} \sum_k \epsilon_k \psi_{\mu,k}^\dagger \psi_{\mu,k} - eV_c(t) \hat{N}_c + eV(t) \hat{N}_f + i \mu g_b \hbar \hat{\alpha} + i \frac{J^+}{2\sqrt{\pi} a L} \sum_k (\psi_{sf,k}^\dagger + \psi_{sf,k}) \hat{b} + i \frac{J^{LR}}{2\sqrt{\pi} a L} \sum_k (\psi_{sf,k}^\dagger - \psi_{sf,k}) \hat{\alpha} + i \frac{J^{-}}{2\sqrt{\pi} a L} \sum_k (\psi_{sf,k}^\dagger + \psi_{sf,k}) \hat{\alpha}. \]

(3)

Here \( J^\pm \) are equal to \((J_{LL}^\pm + J_{RR}^\pm)/2\); the energies \( \epsilon_k \) are equal to \( \hbar v_F k \); \( \hat{N}_c \) and \( \hat{N}_f \) are the number operators for the charge and flavor fermions, respectively; \( V_c(t) \) and \( V(t) \) are equal to \( V_L(t) + V_R(t) \) and \( V_R(t) - V_L(t) \), respectively; \( \alpha \) is an ultraviolet momentum cutoff, corresponding to a lattice spacing; and \( L \) is the effective size of the system (i.e., \( k \) is discretized according to \( k = 2\pi n/L \)). Within this mapping, the Hamiltonian term \( \mathcal{H}_0 \) is reduced to the free kinetic-energy part of Eq. (2). \( \mathcal{H}'_{kin} \), and so \( \rho_0 \) is transformed to \( e^{-\beta \mathcal{H}'_{kin}} / \text{Tr} \{ e^{-\beta \mathcal{H}'_{kin}} \} \).

Having converted the problem to a noninteracting form, one can sum exactly all orders in the perturbation theory to obtain the current. The solution features two basic energy scales, \( \Gamma_a = [(J_{LL}^{LR})^2 + (J^{-})^2]/4\pi a v_F \) and \( \Gamma_b = (J^+)^2/4\pi a v_F \), which determine the width of the different Majorana spectral functions, and thus play the role of Kondo temperatures. The conventional one-channel Kondo effect is best described by the case where \( \Gamma_a = \Gamma_b \), for which only a single Kondo scale emerges.

Following Plihal et al. [3] we begin with the case of a pulsed bias potential and a zero magnetic field. Explicitly, we assume that a bias potential of \( eV \) is switched on at time \( t = 0 \), and then switched off again at time \( t = \tau \). Denoting \((J_{LL}^{LR})^2/4\pi a v_F \) by \( \Gamma_1 \), the resulting time-dependent current \( I(t) \) flowing from right to left is conveniently expressed via the auxiliary function

\[
J(t; V_1, V_2) = \frac{\epsilon \Gamma_1}{\pi \hbar} \sum_{n=0}^{\infty} e^{-(2n+1)\pi k_B T + \Gamma_1 t/k} \times 
\left( \frac{1}{n + \frac{1}{2} - \frac{eV + eV_2}{2\pi k_B T}} - \frac{1}{n + \frac{1}{2} + \frac{eV + eV_2}{2\pi k_B T}} \right).
\]

(4)

For \( t \leq 0, I(t) \) is obviously zero. For \( 0 < t \leq \tau \) it equals

\[
I_1(t) = I_{dc}(V) - \text{Im} \left\{ e^{-ieV t / h} J(t; 0, V) \right\},
\]

(5)
while for $\tau \leq t$ it is given by

$$I_2(t) = \text{Im} \{\mathcal{J}(t-\tau;0,V)\} - \text{Im} \{e^{-ieV\tau/h}\mathcal{J}(t;0,V)\}.$$  

(6)

Here $I_{dc}(V)$ is the steady-state current for an $eV$ dc voltage bias:

$$I_{dc}(V) = \frac{e\Gamma_1}{\pi\hbar} \text{Im} \left\{ \psi \left( \frac{1}{2} + \frac{\Gamma_a + ieV}{2\pi k_B T} \right) \right\}.$$  

(7)

The relevant time scales for the evolution of the current are easily read off from Eqs. (3) and (4). As proposed by Plihal et al. (see Ref. 13, Fig. 3) is strikingly good. In fact, we are even able to reproduce the reversal of the fall-off current for certain pulse lengths. To understand the origin of this somewhat surprising effect, we go back to Eq. (6) with a sufficiently large $v$, such that each term in Eq. (6) is asymptotically given by the solvable point. On the other hand, our solution gives a very transparent picture of the numerical calculations of Plihal et al. (see Ref. 13, Fig. 3) is strikingly good. In fact, we are even able to reproduce the reversal of the fall-off current for certain pulse lengths. To understand the origin of this somewhat surprising effect, we go back to Eq. (6) with a sufficiently large $\tau$, such that each term in Eq. (6) is asymptotically given by

$$I_2(t) \sim \frac{2k_B T \Gamma_1}{\hbar V} e^{-(t-\tau)/t_d} \left[ 1 - e^{-\alpha t_d} \cos \left( \frac{2\pi \tau}{t_v} \right) + \frac{2\pi t_d}{t_v} e^{-\alpha t_d} \sin \left( \frac{2\pi \tau}{t_v} \right) \right].$$  

(8)

Thus, since $t_d \gg t_v$ and $\tau \sim t_v$ under the assumptions above, the asymptotic fall-off current is dominated by the last term in Eq. (8). Consequently, the sign of $I_2(t)$ oscillates with $\sin(2\pi \tau/t_v)$, precisely as seen in Fig. 3.

Although our solution clearly captures the essential findings of the non-crossing approximation, there are two major omissions to be noted. First, working with a Kondo impurity rather than an Anderson impurity, our model lacks the short charge-fluctuation time scale $(t_0 = \hbar/\Gamma_{\text{dot}})$ in the notations of Ref. 13, which governs the very early response of a quantum dot to the abrupt change in the applied voltage bias. Second, for $0 < t \leq \tau$, our solution does not show the exact decrease in saturation time for large voltage bias. This decrease, which stems from the dissipative lifetime induced by the bias potential, is not captured by the solvable point. On the other hand, our solution gives a very transparent picture for the role of a temperature. As the temperature is increased, $t_d$ crosses over from $\hbar/\Gamma_a$ to $\hbar/\pi k_B T$, which re-
bias potential is the current for a zero magnetic field and an effective panel: Solid, dashed, dotted, and long-dashed lines correspond to $\muBgH/\Gamma_a = 1, 2, 4$, respectively. Lower, $\pm eV$, and a rectangular pulsed magnetic field of magnitude $H$. Upper panel: Solid, dashed, dotted, and long-dashed lines correspond to $\muBgH/\Gamma_a = 0, 1, 2, 4$, respectively. In both cases, the pulse duration is $t_\tau = 5h/|eV \pm \muBgH|$, and the temperature is equal to $k_BT/\Gamma_a = 0.1$. $I_{dc}(V,H)$ marks the steady-state current for a dc voltage bias of $eV$ and a dc magnetic field $H$. Remaining the only relevant time scale for $k_BT \gg eV$. Thus, all response times for the current are determined by the temperature in this limit.

An important advantage of the solvable point with $\Gamma_a = \Gamma_i$ is the ability to incorporate an arbitrary time-dependent magnetic field. Specifically, we find that the current $I(t)$ for a general voltage bias $eV(t)$ and an arbitrary field $H(t)$ is equal to $\frac{1}{2} [I_+(t) + I_-(t)]$, where $I_\pm(t)$ is the current for a zero magnetic field and an effective bias potential $eV_\pm(t) = eV(t) \pm \muBgH(t)$. The latter bias has a simple physical interpretation. When an electron tunnels by flipping the impurity spin, the effective potential barrier it sees is equal to $eV_\pm(t)$, depending on the initial impurity state, and the direction of tunneling.

Using the above decomposition of $I(t)$ into $I_\pm(t)$, we have computed the current for the two opposite cases of (i) a dc magnetic field $H$ and a rectangular pulsed bias potential of magnitude $eV$, and (ii) a dc bias potential of $eV$ and a rectangular pulsed magnetic field of magnitude $H$. In both cases, the pulse duration was taken to be $t_\tau$, with the corresponding field being equal to zero for $t < 0$ and $t > t_\tau$. The calculation of $I_\pm(t)$ in each of these cases requires the generalization of Eqs. (7): (8) to a pulsed bias potential of the form $V(t) = V_2$ for $0 \leq t \leq t_\tau$ and $V(t) = V_1 \neq 0$ otherwise, which gives

$$I_1(t) = I_{dc}(V_2) - \text{Im} \left\{ e^{-i eV_2 t/\hbar} \mathcal{J}(t; V_1, V_2) \right\} ,$$

$$I_2(t) = I_{dc}(V_1) + \text{Im} \left\{ e^{-i eV_1 (t-\tau)/\hbar} \mathcal{J}(t-\tau; V_1, V_2) \right\} .$$

For $t \leq 0$, the corresponding current is obviously $I_{dc}(V_1)$.

The resulting $I(t)$ curves are depicted in Fig. 3. As is clearly seen, the combination of a bias and a magnetic field produces nontrivial interference patterns with several underlying time scales. These include $t_{dc}, t_\tau = 2\pi h/|eV \pm \muBgH|$, and either $\tau_h = 2\pi h/\muBgH$ or $\tau_0$, depending on whether one is dealing with a pulsed bias potential or a pulsed magnetic field. In particular, for a pulsed bias potential and moderately large dc magnetic fields, there are current oscillations in the fall-off current, with a characteristic frequency of $\omega = \muBgH/\hbar$. Such effects should be observable in ultra small quantum dots.

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