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Kondo time scales for quantum dots: Response to pulsed bias potentials

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The response of a quantum dot in the Kondo regime to rectangular pulsed bias potentials of various strengths and durations is studied theoretically. It is found that the rise time is faster than the fall time, and also faster than time scales normally associated with the Kondo problem. For larger values of the pulsed bias, one can induce dramatic oscillations in the induced current with a frequency approximating the splitting between the Kondo peaks that would be present in steady state. The effect persists in the total charge transported per pulse, which should facilitate the experimental observation of the phenomenon.

The theoretical predictions[1] of consequences of the Kondo effect for the steady state conduction through quantum dots began a decade ago. At low temperatures, a narrow resonance in the dot density of states can form at the Fermi level, leading to a large enhancement of the dot's conductance, which is strongly dependent on temperature, bias, and magnetic field. Many of these effects have been recently observed by a set of experiments by several groups.[2] These successes, supplemented by the anticipation that time dependent experiments[3] are not far behind, have spurred a number of theoretical groups[4] to consider the effects expected when sinusoidal biases or gate potentials are applied. Surprisingly, the application of steps or pulses, which can provide a less ambiguous measure of time scales, have not been considered until very recently;[5] the latter work considered the time dependent change in linear response conductance when a stepped potential was applied to a gate, thereby shifting the dot into the Kondo regime. In the present work we consider the response of a dot already in the Kondo regime to a sudden change of the bias potential across the dot. We show that the physics is qualitatively different from the latter case,[5] leading to a different range of characteristic times and physical phenomena.

While Ref. 5 studied the time scale for the system to go from one equilibrium configuration to another, we study here the time scale to go from an equilibrium configuration to a nonequilibrium one; and then back to equilibrium again. We find that these latter two times scales are very different from each other, the first being much shorter—and also much shorter than the time scale of Ref. 5. Furthermore, if one applies a rectangular bias pulse large enough to split the Kondo resonance, then there appear current oscillations[6] at a frequency characterizing the splitting between the Kondo peaks. We show that these current oscillations cause oscillations in the charge transported through the dot as a function of pulse length, thus providing a clear experimental signature. The damping of the oscillations provides an additional time scale which also can be measured directly.

We model the quantum dot by a single spin degenerate level of energy $\epsilon_{\rm dot}$ coupled to leads through tunnel barriers, as illustrated schematically in the inset to Fig. 1. The Coulomb charging energy U prevents the level from being doubly occupied. We apply a time-dependent potential across the leads $v(t)=v_0$ for $0<t<\tau$ and zero otherwise, so that the energy E_k of an electron in a lead is time dependent. Here k represents all lead quantum numbers other than spin, including the labels, left and right, specifying the lead in question. Then we have $E_k=E_k(t)=\epsilon_k\pm\frac{1}{2}ev(t)$, the sign depending on which lead k refers to, where ϵ_k is the band energy. The system may be described by the following

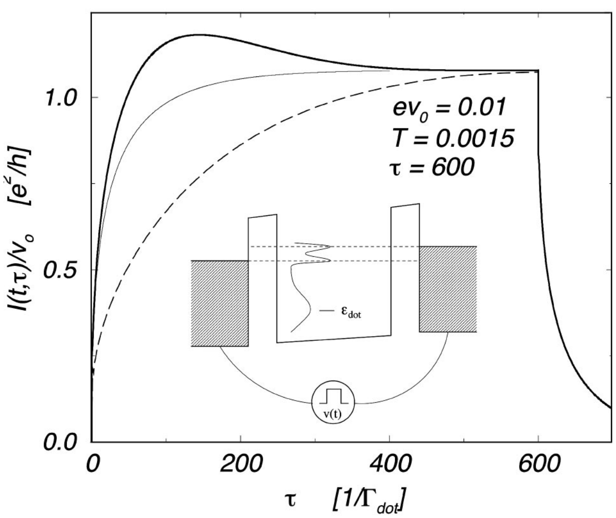

FIG. 1. The instantaneous current $I(t,\tau)$ induced by a bias pulse of height $ev_0=0.01$ beginning at $t=0$ and ending at $t=\tau=600$ (heavy solid line). The portion for $t>\tau$ is translated back to the origin and inverted (light solid line) so that the rise time and fall time can be easily compared. The area between these curves represents the excess charge forced through the dot above what would flow $[G(v_0)\tau]$ if the response to v_0 were instantaneous. The value of v_0 is small, and these curves represent the very beginning of the nonlinear response regime. As v_0 increases further, the rise time becomes shorter $[\propto 1/v_0]$, while the fall time remains long. The dashed line represents the extent to which the Kondo state would be formed (at zero bias) if the Kondo coupling were to be suddenly turned on at $t=0$ (see text). Inset: Schematic of the quantum dot during a pulse.

Anderson Hamiltonian:

$$\sum_{\sigma} \epsilon_{\rm dot} n_{\sigma} + \sum_{k\sigma} [E_k(t) n_{k\sigma} + (V_k c_{k\sigma}^{\dagger} c_{\sigma} + \text{H.c.})], \qquad (1)$$

with the constraint that the occupation of the dot cannot exceed one electron. Here $c_{\sigma}^{\dagger}$ creates an electron of spin σ in the quantum dot, with n_{σ} the corresponding number operator; $c_{k\sigma}^{\dagger}$ creates an electron in the leads.

The general features of the static equilibrium spectral density when the dot level ϵ is sufficiently below the Fermi level (taken at zero here) are well known. There is a broad resonance of half-width $\sim\Gamma_{\rm dot}$,[7] at an energy $\sim\epsilon_{\rm dot}$, plus a sharp temperature sensitive resonance at the Fermi level (the Kondo peak), characterized by the low energy scale T_K (the Kondo temperature), $T_K \simeq D' \exp(-\pi|\epsilon_{\rm dot}|/\Gamma_{\rm dot})$, where D' is a high energy cutoff.[8] Throughout this work, energies and temperatures are given in units of $\Gamma_{\rm dot}$, and times in units of $1/\Gamma_{\rm dot}$, with $\hbar=1$. Specifically, the calculations in this paper were made with $\epsilon_{\rm dot}=-2$, which leads to a Kondo temperature $T_K \sim 0.0025$. We present results only for a dot with left-right symmetry.

The current into the dot depends on the time t and parametrically on the pulse length τ, and is given by

$$I_{\rm in}(t,\tau) = ie \sum_{k\sigma} V_k \langle c_{k\sigma}^{\dagger}(t) c_{\sigma}(t) \rangle + \text{c.c.} \qquad (2)$$

It may be divided into contributions $I_{\rm left}(t,\tau)$ and $I_{\rm right}(t,\tau)$ by respectively restricting the k summations to the appropriate lead. The transport current is then $I(t,\tau)=\frac{1}{2}[I_{\rm left}(t,\tau) - I_{\rm right}(t,\tau)]$. We calculate the Keldysh propagators corresponding to the angular-bracketed expectation values in Eq. (2) for each lead, and hence obtain $I(t,\tau)$. A more experimentally accessible quantity, the total charge transported, $Q(\tau)=\int_0^{\infty} dt\, I(t,\tau)$ is also calculated. Our calculations use the noncrossing approximation (NCA), which is reliable for temperatures down to $T<T_K$.[9] Its time-dependent formulation has been developed elsewhere.[10] The finite bias on the leads is taken into account by introducing a time-dependent phase in V_k in Eq. (1).

In Fig. 1, we show the response to a long, weak bias pulse, which illustrates some features that will persist for still stronger pulses. The most important feature is that the rise time of the current is shorter than the decay time, leading to a greater charge transport than would have occurred if the response at each end of the pulse had been instantaneous.

Another comparison can also be made: how does this time scale for turning on the current compare with how long it takes to form the Kondo state itself? The latter was obtained in Ref. 5, by calculating the turn-on of the linear response current when the parameter $\epsilon_{\rm dot}$ was suddenly switched into the Kondo regime. This is illustrated by the dashed curve in Fig. 1, which is the calculation of Ref. 5 for the present parameters, with the abscissa scaled to make the curves coincide at saturation (large τ and large t, with $t<\tau$). It is clear that the formation time for the Kondo state is significantly longer.

A closer look at Fig. 1 reveals that approximately a third of the initial rise (in both curves) when the bias is turned on (and the same fraction of the initial fall when the bias is

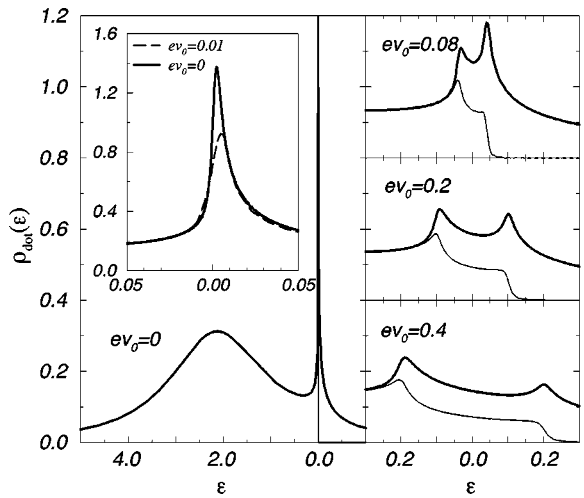

FIG. 2. Steady state spectral densities for different values of the bias. The left panel shows the whole spectral function for the bias equal to the v_0 of Fig. 1, with the Kondo region expanded in the inset; the equilibrium spectral function is also shown. The three right panels show just the Kondo region for increasing biases. The heavy curves represent the total spectral density, while the light curves represent the occupied portion. The oscillations that we find in the next two figures presumably represent transitions from the lower Kondo peak in the occupied spectra to the upper peak in the unoccupied spectra (i.e., the difference between the pairs of curves). Note that the vertical scale factors for all four panels (but, not the inset) are identical. $T=0.0015$ everywhere.

turned off) occurs essentially instantaneously on the scale of the graph. This fast response occurs on the time scale $t_0 \equiv 1/\Gamma_{\rm dot}$. This represents the part of the conductance arising from the fact that the dot level has finite width which overlaps the Fermi level, and would be present even in the absence of the Kondo peak.

The Kondo derived features from switched pulses can be partially understood by reference to the *steady state* spectral densities for a dot with a fixed bias equal to v_0. Once $ev_0 \gg \max(T_K,T)$, the Kondo peak becomes split, with a peak separation associated with the potential $\pm\frac{1}{2}v_0$ of each respective lead, as discussed by Wingreen and Meir.[1] Such spectral densities for increasing values v_0 are illustrated in Fig. 2. This splitting should provide a sharp excitation frequency $\omega \sim ev_0$ for the dot, with an electron being excited from the lower peak at energy $-\frac{1}{2}v_0$ to the upper peak at $+\frac{1}{2}v_0$. We should therefore expect current oscillations of this frequency to be induced by the rapid turn on of the voltage pulse. For larger v_0's than in Fig. 1, the frequency ω provides a fast characteristic rise time $t_1=\pi/2\omega$, and we may expect that the rise time depicted in Fig. 1, is a precursor to this effect. Figure 3, showing the results for a much larger v_0, not only confirms this interpretation, but shows how dramatic the current oscillations can be. In what follows, we refer to them as *split Kondo peak (SKP) oscillations*.

How can one experimentally distinguish SKP oscillations from those reported[11] for the resonant level model ($U=0$), which occur as each lead adjusts to its new chemical potential (NCP)? Our calculations show that, unlike the SKP oscillations which occur robustly at a frequency $\sim ev_0$, the NCP oscillations are sensitive to the details of the exciting

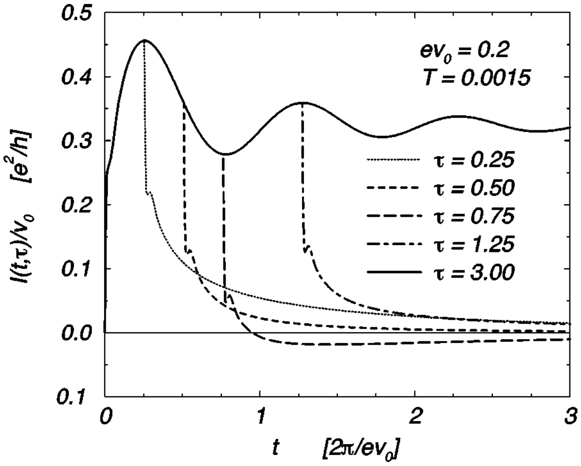

FIG. 3. The instantaneous current $I(t,\tau)$ induced by a strong pulse (an order of magnitude larger than the width of the zero bias Kondo peak) for several pulse lengths τ. The units of both τ and t are both $2\pi/ev_0$, which is almost precisely the ''period'' of the induced oscillations (although we see the frequency slightly decrease from $\omega = ev_0$ as the bias is lowered towards T_K). Note the reversal, for certain pulse lengths, of the direction of the instantaneous current after the end of the pulse.

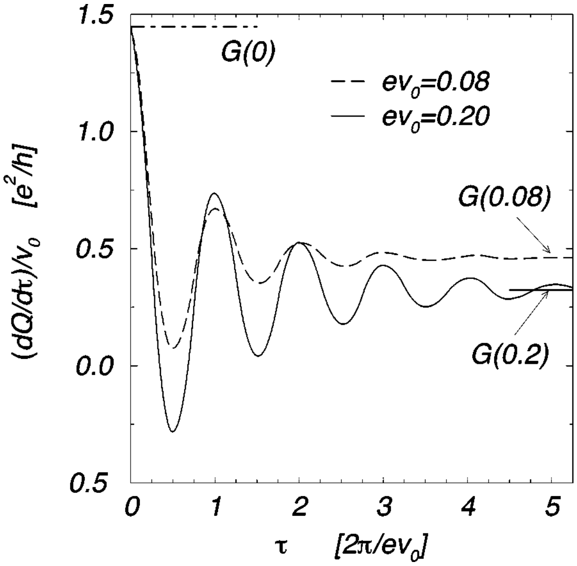

FIG. 4. $1/v_0$ times the derivative with respect to pulse length τ of the total charge Q transported during and after a pulse vs τ. The curves go to the zero bias conductance $G(0)$ at $\tau = 0$, and each go to the respective values of the finite bias conductance $G(v_0)$ for large τ.

pulse. First, with the resonant level at the Fermi level to simulate a Kondo peak, the NCP oscillation frequency is equal to ev_0 only in the special case that a pulse of magnitude $|ev_0|$ is applied to a single lead, with the other held at the original Fermi level. For equal amplitude pulses of $\pm ev_0/2$ to the respective leads, the oscillation frequency is, on the other hand, $ev_0/2$. In a more general case of asymmetrically applied pulses there are *two* oscillation frequencies, one associated with the new chemical potential of each lead. If in addition, the resonant level is not at the original Fermi level, the oscillations are even more complicated, with multiple frequency components.

The NCP oscillations are damped on the time scale $\sim 1/\Gamma_{\mathrm{dot}}$. When this becomes shorter than the oscillation period, the oscillations become overcritically damped and do not appear. We find that they only occur for $ev_0/\Gamma_{\mathrm{dot}} \geqslant 1$, and not for the much smaller values of this ratio used in the large U examples shown in this paper. SKP oscillations, on the other hand, occur only when $ev_0/\Gamma_{\mathrm{dot}} \ll 1$, and hence not simultaneously with NCP oscillations. This explains the robustness of the oscillations in the Kondo regime.

SKP oscillations can probably be excited by other types of applied waveforms, and a parametrically driven form of them is probably responsible for the resonances reported by Schiller and Hershfield.[4] However, we suspect that the well-defined robust frequency for them provided by a square pulse, plus the lack of one for NCP oscillations, may provide the key for unambiguous experimental identification.

Figure 3 shows an additional feature—the damping of the SKP current oscillations. This damping presumably arises from the fact that the split Kondo peaks in the spectral density have widths, giving rise to dephasing. These widths are substantially greater than the width of the Kondo peak in equilibrium, as the application of a bias begins to weaken the Kondo effect. The reason for this has been pin-pointed by Wingreen and Meir,[1] in a model perturbation theoretic calculation whose essential feature is that the particle-hole phase space restriction from the Pauli principle in the leads for incoherent spin processes is eased. For biases $ev_0 \gg T$, this gives a rate enhancement factor of $\sim ev_0/T$ relative to the zero bias case. For $T < T_K$, one should replace T by T_K in the above argument; in this case the width of the Kondo peak(s) would be proportional to ev_0 for $ev_0 \gg T_K$, but would saturate at $\sim T_K$ for $ev_0 \ll T_K$. Translating this into the damping time t_2 of the oscillations, gives $t_2 \propto 1/v_0$ at large biases, but with a saturation $t_2 \sim 1/T_K$ when $ev_0 < T_K$. The expected saturation as the split Kondo peaks merge removes the oscillations for $ev_0 < T_K$ (cf. Fig. 1).

Figure 3 also shows the falloff in current after the voltage pulse has been turned off. After an initial drop due to the fast non-Kondo time scale $t_0 = 1/\Gamma_{\mathrm{dot}}$, the current falls very slowly. Its decay does not follow single time scale, but it is clear that the total time required for the decay is very long. While the initial falloff after t_0 is bias dependent and relates to the time t_1, the long tail has a characteristic time $t_3 \sim 1/T_K$, the longest time in the problem. This is to be expected: the system is trying to regain an equilibrium where the Kondo peak is no longer split or broadened.

Although we only show the results for one temperature, $T = 0.0015 \sim T_K$, we have made calculations for a number of higher and lower temperatures. The results are not very sensitive to temperature when $T \leqslant T_K$. At higher temperatures, the oscillation amplitude decreases, and each time t_1, t_2, or t_3 shortens when T gets large enough to equal the reciprocal of the time in question ($T \sim 1/t_i$). The falloff time t_3 and oscillation decay time t_2 are the first ones affected. At $T \sim ev_0$, the rise time t_1 also shortens. However, the nonlinear effects persist to temperatures much higher than T_K presumably because of the logarithmic nature of the Kondo peak.

How can these effects be seen experimentally? We suggest that the quantity $Q(\tau)$ (the charge transported through the dot by a single pulse of length τ) will contain sufficient memory of the charge already transported at the time τ when the bias is turned off, to directly reflect the current oscillations and their damping. $Q(\tau)$ is proportional to the average current transported by a series of successive pulses, provided that the time between pulses is $\gg t_3$; it thus corresponds to an easily measured quantity. To restore the features to the prominence which was there before the time integration, it is suggested that $dQ(\tau)/d\tau$ be measured. In Fig. 4 we display our predicted $dQ(\tau)/d\tau$ plotted vs τ for a number of values of bias amplitude v_0. The information about the frequency and damping of the oscillations, and hence of the time scales t_1 and t_2 is preserved. Similar oscillatory structure for fixed τ occurs in plots of dQ/dv_0 vs v_0.

In conclusion, we have studied the response of a quantum dot in the Kondo regime to a large pulsed bias voltage across the leads. We find that the rise time of the instantaneous current is related to the period of current oscillations that are set up. These oscillations have a frequency corresponding to the energy difference between the split Kondo peaks. The damping rate of these oscillations is related to the widths of the split Kondo peaks. The falloff of the current when the pulse is turned off can take much longer than the rise, with a tail of length approaching $1/T_K$. These effects may be studied experimentally by measuring the total charge transported through the dot during and after a voltage pulse.

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[6] P. Coleman (private communication) has also predicted these oscillations, starting from mean field theory in the slave boson field.

[7] We define $\Gamma_{\rm dot}(\epsilon)=2\pi\Sigma_k|V_k|^2\delta(\epsilon-\epsilon_k)$, a slowly varying quantity. The notation $\Gamma_{\rm dot}$ with no energy specified will always refer to the value at the Fermi level.

[8] For $U=\infty$, $D'\simeq\sqrt{D\Gamma_{\rm dot}/4}$, where $2D$ is the effective bandwidth. The calculations here used a parabolic band of total width $9\Gamma_{\rm dot}$.

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