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LETTER TO THE EDITOR

Oscillatory instabilities in dc-biased quantum dots**P Coleman¹, C Hooley^{1,2}, Y Avishai³, Y Goldin³ and A F Ho⁴**¹ Center for Materials Theory, Rutgers University, Piscataway, NJ 08854, USA² School of Physics and Astronomy, Birmingham University, Edgbaston, Birmingham B15 2TT, UK³ Department of Physics, Ben Gurion University, Beer Sheva, Israel⁴ Department of Physics, Oxford University, 1 Keble Road, Oxford OX1 3NP, UK

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Online at stacks.iop.org/JPhysCM/14/L205**Abstract**

We consider a ‘quantum dot’ in the Coulomb blockade regime, subject to an arbitrarily large source–drain voltage V . When V is small, quantum dots with odd electron occupation display the Kondo effect, giving rise to enhanced conductance. Here we investigate the regime where V is increased beyond the Kondo temperature and the Kondo resonance splits into two components. It is shown that interference between them results in spontaneous oscillations of the current through the dot. The theory predicts the appearance of ‘Shapiro steps’ in the current–voltage characteristics of an irradiated quantum dot; these would constitute an experimental signature of the predicted effect.

Quantum dot devices [1] display very rich physics in and near equilibrium. In particular, experiments [2] over the past few years have confirmed a decade-old theoretical prediction that these systems exhibit the Kondo effect [3]: the low-temperature formation of a hybridization resonance between the dot and the leads to which it is connected. This resonance is the result of collective spin exchange processes between the leads and the dot, which dominate the low-temperature physics below a certain scale (the ‘Kondo temperature’). The Kondo effect is manifested as an enhancement of the dot’s conductance.

A bias voltage applied to a quantum dot modifies the electron energies in the leads, driving a current through the dot. This offers a unique opportunity to study correlated electrons out of equilibrium. Can new kinds of non-equilibrium collective phenomenon develop in driven quantum systems? In the realm of classical physics, there are many instances of transitions to new phases in response to a non-equilibrium driving force. An example is the Rayleigh–Bénard instability, where a temperature difference causes the development of convective roll patterns that spontaneously select their own spatial and temporal frequencies [4]. However, for a driven quantum system to develop new collective behaviour, it must preserve its quantum mechanical coherence. The issue of whether a bias voltage preserves the phase coherence of a quantum dot is thus a matter of considerable concern.

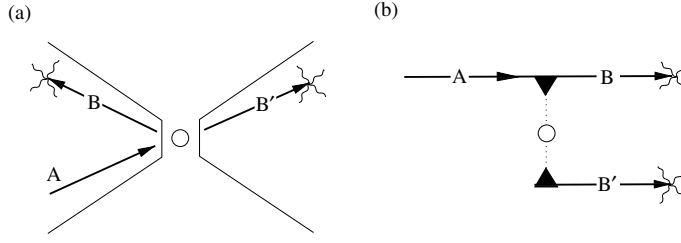


Figure 1. (a) A schematic depiction of an incoming electron wave A scattering off the quantum dot into the same (B) and the opposite (B') leads. Dephasing (indicated by wavy crosses) takes place after the scattering event. (b) The mapping of the quantum dot problem to a one-dimensional 'chiral' model. Electrons and information travel from left to right. Dephasing of electrons at B and B' cannot affect the quantum dot.

In this letter we argue that electron flow through a quantum dot modifies, but does not dephase the correlations of the dot. Central to our arguments is the observation that the physics of a quantum dot can be mapped onto a one-dimensional problem [5], where the electrons in each lead are represented by waves travelling in one direction along one-dimensional conductors, arriving at the dot from the left and leaving it to the right (figure 1). This chiral mapping is exact. Electrons scattering off the dot do not lose phase information until they suffer subsequent inelastic collisions with other electrons and magnetic impurities in the leads, which, in the chiral mapping picture, lie to the 'right' of the dot. Since information travels only from left to right, the dephasing of an outgoing electron does not affect the spin it has left behind, and thus a current of electrons through a quantum dot does not dephase the Kondo effect, but acts as a coherent driving force.

This argument is employed to examine the possibility of dynamical instabilities in a driven, yet fully coherent quantum dot. We present calculations which predict that, beyond a critical bias voltage V_c , the current flowing through the quantum dot spontaneously acquires an oscillatory component whose frequency is a non-linear function of the bias voltage.

The quantum dot system considered here consists essentially of three parts: the dot itself, and the left and right leads, to which it is connected by weak tunnelling junctions. In the Coulomb blockade regime, the occupation of the dot is fixed, and the only allowed processes involve flipping the spin of the unpaired electron on the dot. In this simple picture, a dot containing an odd number of electrons is a spin- $\frac{1}{2}$ impurity. As the temperature is lowered, spin fluctuations lead to the development of a sharp resonance peak in the interacting electron density of states: the Abrikosov–Suhl (AS) resonance. This peak occurs at an energy $\hbar\omega_1$, close to the common Fermi energy of the leads, and has a width $\Delta \sim T_K$, the Kondo temperature (see figure 2(a)).

In the description which we shall use, the coherence associated with the Kondo effect is encoded in terms of a time-dependent hybridization field $\hat{\Gamma}_\beta(t) \sim \sum_{k\sigma} c_{\beta k\sigma}^\dagger d_\sigma$ between the lead electrons and the electron on the dot. (The operator $c_{\beta k\sigma}^\dagger$ creates a lead electron with momentum k and spin σ in lead $\beta \in \{L, R\}$; d_σ annihilates a dot electron with spin σ .) Within the mean-field formalism (see below), $\hat{\Gamma}_\beta$ is replaced by its mean value $\Gamma \equiv \langle \hat{\Gamma}_\beta \rangle = \Gamma_0 e^{i\omega_1 t}$.

In the presence of a sufficiently large bias voltage V , the AS resonance splits into two peaks [6], one near each Fermi level (see figure 2(b)). $\Gamma(t)$ therefore becomes a sum of two terms, $\Gamma(t) = \Gamma_1 e^{i\omega_1 t} + \Gamma_2 e^{i\omega_2 t}$, where ω_1 and ω_2 are the level positions of the two AS peaks. Quantum interference between these two terms produces an oscillatory current at frequency $(\omega_1 - \omega_2) \sim eV/\hbar$. We emphasize that this oscillating current originates as a response to a dc bias: this is to be contrasted with the case where the driving voltage is itself time dependent [7].

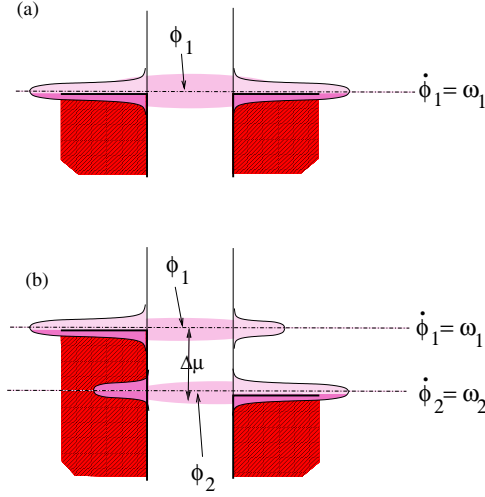


Figure 2. (a) Schematic picture of the equilibrium situation: a single Kondo resonance is formed between the electron on the dot and electrons in both leads. (b) Quantum dot under a dc bias $V > V_c$: Kondo resonances develop at each chemical potential, but *each* resonance still involves the electrons from *both* leads. Interference between the resonances drives the oscillatory current. (This figure is in colour only in the electronic version)

A natural choice for the description of a quantum dot coupled to two leads is the Anderson model. We consider a generalized version of such a model, allowing for an asymmetry between transmission and reflection amplitudes (see below). In the Coulomb blockade regime, charge fluctuations are virtual, and may be integrated out [8] to obtain an effective spin-exchange Hamiltonian:

$$\begin{aligned}
 H &= H_0 + H_R + H_T; \\
 H_0 &= \sum_{\beta k \sigma} (\epsilon_k - \mu_\beta) c_{\beta k \sigma}^\dagger c_{\beta k \sigma}, \\
 H_R &= -\frac{J}{2N} \sum_{\sigma \tau} [c_{L\sigma}^\dagger d_\sigma d_\tau^\dagger c_{L\tau} + (L \rightarrow R)], \\
 H_T &= -(1 - \eta) \frac{J}{2N} \sum_{\sigma \tau} [c_{R\sigma}^\dagger d_\sigma d_\tau^\dagger c_{L\tau} + (L \leftrightarrow R)],
 \end{aligned} \tag{1}$$

where $c_{\beta\sigma}^\dagger \equiv \sum_k c_{\beta k \sigma}^\dagger$. The spin variables σ, τ now take integer values from 1 to N , a generalization which we discuss below. Here H_0 controls the electrons in the leads, with $\mu_L - \mu_R \equiv eV$, the source–drain voltage. The remaining parts of the Hamiltonian describe the spin exchange processes which accompany electron reflection (H_R) and transmission (H_T) at the dot.

A crucial ingredient is the asymmetry parameter η . When (1) is derived directly from the Anderson model, one finds that $\eta = 0$; this reflects a symmetry between reflection and transmission processes that will never be exactly realized in a physical quantum dot system [9]. (In the discussion here, we shall take the parameter η to be independent of the voltage across the leads.) Moreover, it has been proposed [10] that, at voltages above the Kondo temperature, the dot system develops a divergent susceptibility to perturbations of the form $-\lambda O^\dagger O$, where operator O is defined by $O(t) \equiv \sum_{\sigma \tau} (c_{L\sigma}^\dagger \sigma_{\sigma \tau} c_{R\tau}) \cdot \mathbf{S}$, where \mathbf{S} is the impurity spin. The inclusion of a small non-zero η represents the effect of such terms.

Our analysis of the model (1) is based on the self-consistent determination of the hybridization fields $\hat{\Gamma}_{\beta\sigma}(t) = c_{\beta\sigma}^\dagger(t)d_\sigma(t)$. In an actual physical system, the spin index σ may take one of two values (\uparrow or \downarrow), but it is useful to consider a general case where $\sigma \in \{1, \dots, N\}$. This is because, as $N \rightarrow \infty$, the hybridization fields $\hat{\Gamma}_{\beta\sigma}(t)$ behave as well defined semi-classical variables: $\hat{\Gamma}_{\beta\sigma}(t) \rightarrow \frac{1}{N} \sum_\sigma \langle c_{\beta\sigma}^\dagger(t)d_\sigma(t) \rangle$. The use of the large- N approach deserves particular discussion. By making this choice we render the problem exactly solvable, but in doing so, we exclude inelastic scattering processes and thus emphasize the coherent aspects of the quantum dot physics. (Specifically, the lowest-order processes that produce a lifetime for the d -fermion are of $O(1/N)$, and therefore disappear in the $N \rightarrow \infty$ limit.) The phase coherent physics of the $N = 2$ Kondo effect is captured by the large- N limit [11]. Thus, if an applied voltage acts as a phase coherent driving force on the quantum dot, we expect the large- N mean-field theory to correctly capture any collective behaviour that develops at large voltage bias. Support for this approach is provided by recent calculations [10], which show that the quantum dot system remains a non-perturbative phenomenon at arbitrarily large voltages. However, the ultimate test of the procedure must surely lie in experiment.

Replacing the hybridization fields by their expectation values in the large- N limit results in the mean-field Hamiltonian

$$H = H_0 + \sum_\sigma [(\mathcal{V}_L^*(t)c_{L\sigma}^\dagger d_\sigma + d_\sigma^\dagger c_{L\sigma} \mathcal{V}_L(t)) + (L \rightarrow R)] + \frac{N|\mathcal{V}_L + \mathcal{V}_R|^2}{J(2-\eta)} + \frac{N|\mathcal{V}_L - \mathcal{V}_R|^2}{J\eta}, \quad (2)$$

where \mathcal{V}_L and \mathcal{V}_R are defined by

$$\begin{pmatrix} \mathcal{V}_L(t) \\ \mathcal{V}_R(t) \end{pmatrix} = \frac{1}{2} \begin{bmatrix} J & J(1-\eta) \\ J(1-\eta) & J \end{bmatrix} \begin{pmatrix} \Gamma_L(t) \\ \Gamma_R(t) \end{pmatrix}. \quad (3)$$

The amplitudes $\mathcal{V}_{L,R}(t)$ must be self-consistently determined from the expectation values $\Gamma_{L,R}(t)$ at finite voltage V .

Our analysis uses the Schwinger–Keldysh formalism [12], and from the practical point of view this requires an ansatz for the form of the hybridization fields $\mathcal{V}_\beta(t)$. In the present work, two ansätze are investigated: a *two-frequency ansatz* ('2F'),

$$\mathbf{v} \equiv \begin{pmatrix} \mathcal{V}_L \\ \mathcal{V}_R \end{pmatrix} = \mathcal{V}_0(\boldsymbol{\xi}_+ e^{-i\omega_0 t} + \boldsymbol{\xi}_- e^{i\omega_0 t}), \quad (4)$$

and a *one-frequency ansatz* ('1F'), where the second term on the RHS of (4) is absent. The 1F solutions correspond to states with a single AS resonance. The 2F solutions correspond to states in which the resonance has been split into two components at energies $\pm\omega_0$, giving rise to an oscillatory current.

We have supplemented this approach with detailed numerical simulations which employ the Heisenberg equations of motion to time-evolve the one-particle density matrix of the system and keep track, self-consistently, of the averages $\langle c_{\beta\sigma}^\dagger(t)d_\sigma(t) \rangle$ and $\langle c_{\alpha\sigma}^\dagger(t)c_{\beta\sigma}(t) \rangle$, which together describe the state of the whole system. The initial state is taken to be a 'high-temperature' state, i.e. one with extremely small hybridization between the dot and the leads. The fields $\mathcal{V}_\beta(t)$ are updated at each time step to ensure self-consistency. The quantity of interest is the physical current through the dot, $I(t) = 2J(1-\eta) \text{Im}(\Gamma_R(t)\Gamma_L^*(t))$.

Consider first the 1F solutions. These are *static solutions* since, with only one AS resonance present, there is no possibility of interference: the current through the dot is hence time independent. As long as $\eta \neq 0$, static solutions exist for all values of V . As V is increased from zero, the level position $\omega_0 = 0$ *does not change*, i.e. the application of a small bias neither splits nor shifts the AS peak. The width of the peak, Δ , decreases sharply as one increases the voltage (figure 3(b)). At $V = V_c \approx 2T_K/e$, the single static solution at frequency ω_0 bifurcates,

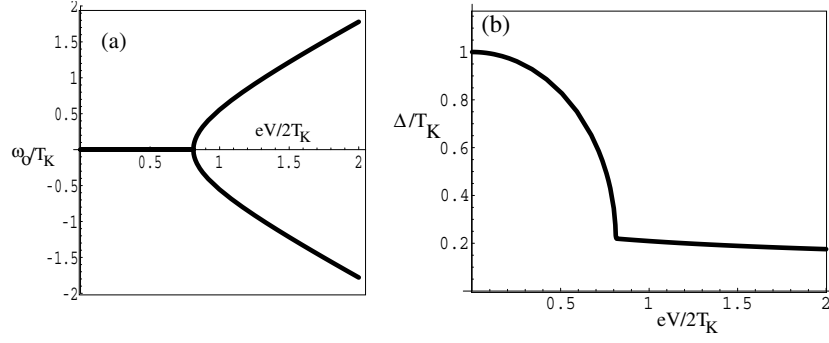


Figure 3. (a) The unstable frequency $\omega_0(V)$ governing the splitting of the AS resonance, in the case $\eta \approx 0.37$. (b) The width $\Delta(V)$ of the AS resonance for the case $\eta \approx 0.37$.

and for $V > V_c$ one finds two *degenerate* static solutions, with frequencies $\pm\omega_0$, where $\omega_0 = (eV/2\hbar)\Phi(eV/2k_B T_K)$ (figure 3(a)). At large voltages, $\Phi(x) \rightarrow 1$; thus these degenerate solutions represent an AS resonance, of width $\Delta(\eta, V)$, attached to the left or right lead.

When $V < V_c$, the numerical simulations support these results; however, for $V > V_c$, they indicate that the static solutions are unstable to the emergence of a 2F solution (4), consisting of components at both frequencies $\pm\omega_0$. Quantum interference between these two AS resonances contributes an oscillating part to the current $I(t)$ (figure 4). The oscillations are monochromatic, and their frequency is indeed $2\omega_0$ (figure 3(a)).

For the oscillating current $I(t)$, one may identify I_{\max} (the maximum instantaneous current) and I_{\min} (the minimum); one may thence define the dc and ac parts: $I_{\text{dc}} \equiv \frac{1}{2}(I_{\min} + I_{\max})$; $I_{\text{ac}} \equiv \frac{1}{2}(I_{\max} - I_{\min})$. These are plotted in figure 5 as functions of V . The appearance of alternating and direct components to the current is a manifestation of a two-frequency solution, for if the hybridization fields have a two-frequency form $\Gamma_R(t) = \Gamma_L^*(-t) = \Gamma_+ e^{-i\omega_0 t} + \Gamma_-^* e^{i\omega_0 t}$, then the current $I(t) = 2J(1 - \eta) \text{Im}(\Gamma_R(t)\Gamma_L^*(t))$ takes the form

$$\frac{I}{2J(1 - \eta)} = \text{Im}[\Gamma_+^2 + \Gamma_-^2] + 2|\Gamma_+ \Gamma_-| \sin(2\omega_0 t + \phi), \quad (5)$$

where we have written $\Gamma_+ \Gamma_- = |\Gamma_+ \Gamma_-| e^{-i\phi}$. Phase coherence between Γ_+ and Γ_- is needed to stabilize the phase ϕ inside the oscillatory term.

At small V , the I - V curve is linear, and then peaks at $eV \sim k_B T_K$. The behaviour of the differential conductance $G(V) \equiv dI_{\text{dc}}/dV$ beyond the peak is novel, as it is affected by the sharp transition into the new regime where direct and alternating currents occur together. As the voltage is increased from zero, $G(V)$ changes sign at $eV \sim k_B T_K$, exhibits a discontinuity at $V = V_c$ and eventually approaches zero from below.

We conclude with a brief discussion on the detection of this proposed oscillatory phenomenon. In close analogy to the Josephson effect, we expect that when the quantum dot is exposed to radiation, the current-voltage profile will develop Shapiro steps [13] when the oscillatory frequency is commensurate with the frequency of the incident radiation. We give here a brief derivation of the expected effect.

A modulation in the bias voltage $V(t) = V_0 + V_1 \sin \omega_{\text{in}} t$ will lead to a modulation in the separation of the Kondo resonances on opposite leads, given approximately by $2\omega(t) = 2\omega_0 + (eV_1/2\hbar) \cos \omega_{\text{in}} t$. Since this separation determines the rate of change of the oscillatory current's phase, the current in the irradiated dot will take the form

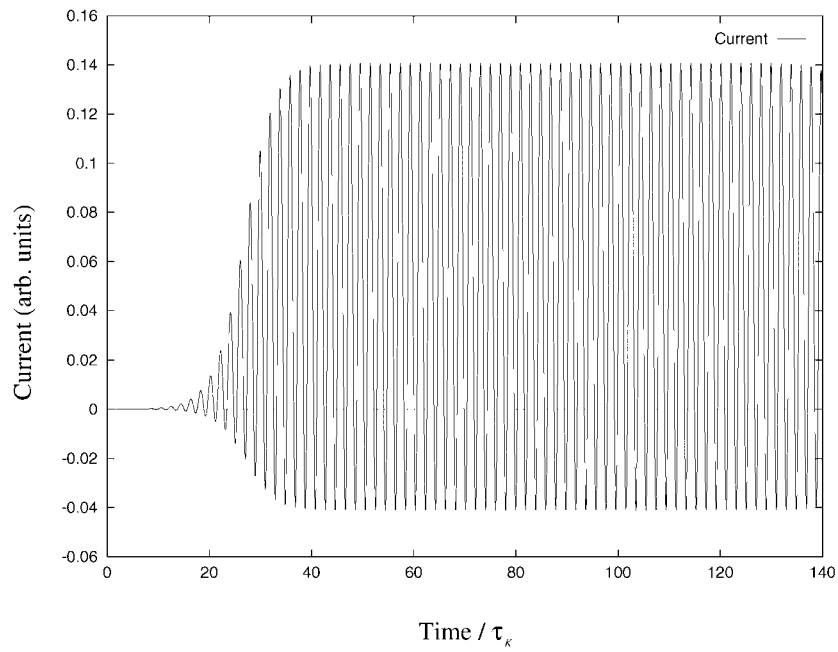


Figure 4. Tunnelling current through the dot, $I(t)$, as a function of time, for dc voltage $V = 3.5 T_K$. (Here $\eta \approx 0.46$, and time is measured in units of the Kondo time, $\tau_K \equiv \hbar/k_B T_K$.)

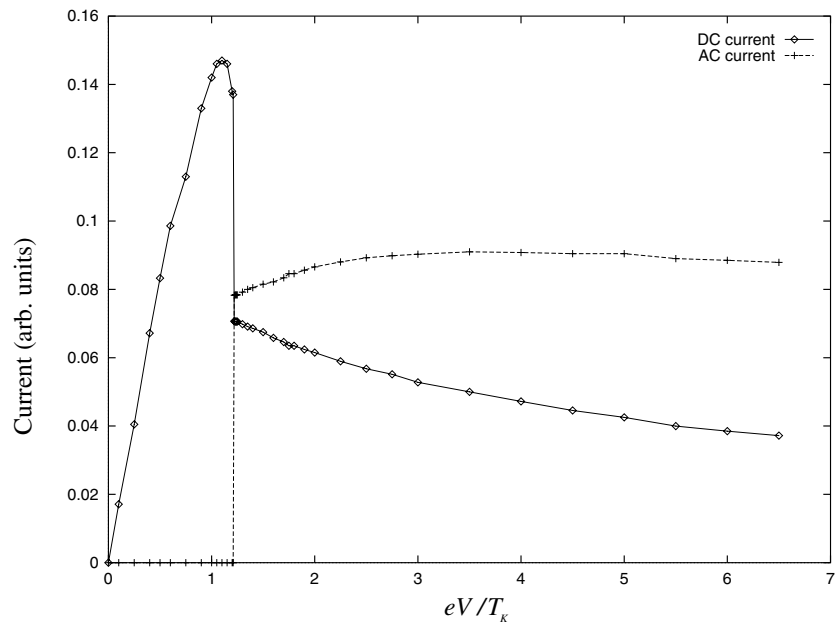


Figure 5. The direct and alternating parts of the current through the dot (I_{dc} and I_{ac}) as a function of voltage, V , in the case $\eta \approx 0.46$.

$$\begin{aligned}
 I(t) &= I_{\text{dc}} + I_{\text{ac}} \sin\left(2 \int^t \omega(t') dt'\right) \\
 &= I_{\text{dc}} + I_{\text{ac}} \sin\left(2\omega_0 t + \frac{eV_1}{\hbar\omega_{\text{in}}} \sin(\omega_{\text{in}} t) + \phi\right). \quad (6)
 \end{aligned}$$

This function contains new Fourier components at frequencies $2\omega_0 \pm n\omega_{\text{in}}$, where n is an integer, so that when $2\omega_0$ is a multiple of ω_{in} , additional contributions appear in the direct current. In a current-biased quantum dot, we expect this rectification effect to lead to Shapiro steps in the current at voltages where ω_{in} is commensurate with the separation of the Kondo resonances $2\omega_0(V)$. Experimentally, these steps should be visible in the range $T_K < V_0 < U$, where U is the charging energy of the dot. Observation of such Shapiro steps would constitute direct evidence of phase coherent current oscillations in the dc-biased quantum dot.

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