

# Nonlinear Response of a Kondo system: Direct and Alternating Tunneling Currents.

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(cond-mat/9710085)

## Abstract

Non - equilibrium tunneling current of an Anderson impurity system subject to both constant and alternating electric fields is studied. A time - dependent Schrieffer - Wolff transformation maps the time - dependent Anderson Hamiltonian onto a Kondo one. Perturbation expansion in powers of the Kondo coupling strength is carried out up to third order, yielding a remarkably simple analytical expression for the tunneling current. It is found that the zero - bias anomaly is suppressed by the ac - field. Both dc and the first harmonic are equally enhanced by the Kondo effect, while the higher harmonics are relatively small. These results are shown to be valid also below the Kondo temperature.

The Kondo effect in bulk materials has been extensively studied for more than three decades (for review, see [1,2]). The impressive advance in the fabrication of mesoscopic tunneling devices opens a road to explore the *non - equilibrium* Kondo physics. Experiments have been reported on crossed - wire tungsten junctions [3], quenched lithographic point contacts [4], metal and metallic glass break junctions [5] and, recently, quantum dots [6]. Similar progress has also been recorded in numerous theoretical works, [7,8]. Within this realm, theoretical interest is focused on the physics of a Kondo system subject to nonlinear time - dependent fields [9–12]. Although an appropriate experimental research has not yet been carried out, the required ranges of frequencies and temperatures have already been achieved in transport measurements so there is no real obstacle to perform the pertinent experiments.

So far, calculations of the current through a Kondo system subject to a time - dependent bias were carried out using various assumptions and approximations [9–11]. In the present work we carry out straightforward perturbation expansion of the current in powers of the coupling constant between the impurity (or the quantum dot) and the conduction bands. Indeed, perturbation theory proved its usefulness for the equilibrium Kondo model right

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from its onset [13]. Recently it was used by Sivan and Wingreen [8] for the Anderson model (in the Kondo regime) with a constant voltage bias.

Our strategy is to start from the time - dependent Anderson model Hamiltonian and then carry out a time - dependent Schrieffer - Wolff transformation to map it onto the corresponding Kondo - type Hamiltonian. The next step is to combine a specific approach suggested by Coleman [14] to treat interaction in strongly correlated systems with the Schwinger - Keldysh non - equilibrium Green's functions formalism. It makes the problem amenable for perturbation expansion which is performed below up to third order in the Kondo coupling  $J$  (sixth order in the tunneling strength). The outcome of this procedure is an analytical expression for the tunneling current, which is used to obtain two novel results pertaining to the direct and alternating currents, namely: 1) The zero - bias anomaly in the direct current is smoothed out by an external alternating field. 2) The zeroth and the first harmonic of the time - dependent current are enhanced by the Kondo effect while the other harmonics remain relatively small.

At voltages and frequencies less than the level spacing in the tunneling region a convenient way to describe a tunneling system is the Anderson Hamiltonian:

$$H_A = \sum_{k \in L, R; \sigma} \left( \epsilon_k + \Delta_{L(R)}(t) \right) a_{k, \sigma}^\dagger a_{k, \sigma} + \sum_{\sigma} \epsilon_{d, \sigma} c_{d, \sigma}^\dagger c_{d, \sigma} + \frac{1}{2} U \sum_{\sigma, \sigma' \neq \sigma} n_{\sigma} n_{\sigma'} + \sum_{k \in L, R; \sigma} \left( \tilde{V}_{kd} a_{k, \sigma}^\dagger c_{d, \sigma} + \text{h.c.} \right). \quad (1)$$

Here  $a_{k, \sigma}^\dagger (a_{k, \sigma})$  creates (annihilates) an electron with momentum  $k$  and spin  $\sigma$  in one of the two leads,  $c_{d, \sigma}^\dagger (c_{d, \sigma})$  creates (annihilates) an electron with spin  $\sigma$  in the tunneling region (hereafter we call it "dot"),  $\epsilon_k$  and  $\epsilon_{d, \sigma}$  are one - particle energies in the leads and in the dot respectively,  $U$  is the Coulomb interaction energy in the dot,  $n_{\sigma} \equiv c_{d, \sigma}^\dagger c_{d, \sigma}$ , while  $\tilde{V}_{kd}$  are transfer matrix elements between the leads and the dot. The external fields are included through the potential shifts of the leads  $\Delta_{L(R)}(t)$ :

$$\Delta_{L(R)}(t) \equiv \phi_{L(R)} + W_{L(R)} \cos(\Omega t + \alpha_{L(R)}). \quad (2)$$

The first term describes a dc potential, while the second one is due to an ac field, which, for simplicity, is assumed to be monochromatic. We note that chemical potentials of the leads are shifted by the same amount  $\Delta_{L(R)}(t)$  as the one - particle energies, so the population of energy levels in the leads does not change.

Although it is possible to carry out a perturbation expansion within the above model using the slave boson technique [8] it turns out to be very cumbersome for time - dependent problems. Instead, let us first map the Hamiltonian (1) onto a Kondo - like one using a *time - dependent version* of the Schrieffer-Wolff transformation [15]. This procedure (whose details will be explained elsewhere) appears to be straightforward albeit not trivial. Its application is limited to the Kondo regime, which is determined by the condition

$$\epsilon_{d, \sigma} < 0, \quad \epsilon_{d, \sigma} + U > 0, \quad |\epsilon_{d, \sigma}|, \epsilon_{d, \sigma} + U \gtrsim \Gamma_{\sigma}, \quad (3)$$

where  $\Gamma_{\sigma} = 2\pi \sum_{k \in L, R} |\tilde{V}_{kd}|^2 \delta(\epsilon_{d, \sigma} - \epsilon_k)$  are the widths of the energy levels in the dot. Furthermore, it is assumed that the external fields are not strong enough to draw the system out of this regime, so that:

$$|\phi_{L(R)}| \lesssim |\epsilon_{d,\sigma}|, |\epsilon_{d,\sigma} + U| \quad \Omega, W_L, W_R \lesssim |\epsilon_{d,\sigma}|, |\epsilon_{d,\sigma} + U|. \quad (4)$$

It should be stressed however that these conditions do not imply a linear response. The later is defined by the conditions  $|\phi_{L(R)}|, W_{L(R)} \ll T$  while  $T \ll |\epsilon_{d,\sigma}|, |\epsilon_{d,\sigma} + U|$ . For the sole purpose of having a more compact form of the Hamiltonian the infinite  $U$  limit is taken. The resulting Hamiltonian then reads,

$$H_K = \sum_{k \in L, R; \sigma} \epsilon_k a_{k,\sigma}^\dagger a_{k,\sigma} + 1/2 \sum_{k, k' \in L, R; \sigma} J_{k'k}(t) \left[ a_{k',-\sigma}^\dagger a_{k,\sigma} c_{d,\sigma}^\dagger c_{d,-\sigma} + a_{k',\sigma}^\dagger a_{k,\sigma} c_{d,\sigma}^\dagger c_{d,\sigma} \right], \quad (5)$$

where

$$J_{k'k}(t) = V_{k'd} V_{kd}^* \exp \left[ (\phi_{(k')} - \phi_{(k)}) t \right] \sum_{s', s=-\infty}^{+\infty} J_{s'} \left( \frac{W_{(k')}}{\Omega} \right) J_s \left( \frac{W_{(k)}}{\Omega} \right) \exp \left[ (s' - s) \Omega t + i(s' \alpha_{(k')} - s \alpha_{(k)}) \right] \cdot \left( \frac{1}{\epsilon_{k'} + \phi_{(k')} + s' \Omega - \epsilon_{d,\sigma}} + \frac{1}{\epsilon_k + \phi_{(k)} + s \Omega - \epsilon_{d,\sigma}} \right), \quad (6)$$

$V_{kd} \equiv \tilde{V}_{kd} \exp \left[ -i \left( W_{(k)}/\Omega \right) \sin \alpha_{(k)} \right]$ , the symbol  $(k)$  means “L” or “R” depending whether  $k$  belongs to the left or to the right lead, while  $J_s(\frac{W}{\Omega})$  are Bessel’s functions.

Before proceeding with the evaluation of the current, two comments are in order: i) Any procedure toward solution of the ensuing transport equations should take into account the fact that, out of the full Hilbert space, the system is projected onto a subspace  $F_1$  for which the dot is occupied by one (and only one) electron. ii) At this stage one might be tempted to express the electron creation - annihilation operators in the dot through spin operators [15], thus arriving at the familiar form [1,15] of the Kondo Hamiltonian. But then one would realize that the spin operators do not obey the usual commutation rules. In order to overcome this obstacle, fictitious (auxiliary) fermions [16] might be introduced. But this leads one back to equation (5). In other words, auxiliary fermions which sometimes regarded as artificial particles introduced to represent spins are real electrons in the dot (impurity atom) subject to the constraint specified in i).

Calculation of tunneling current starting from the Kondo Hamiltonian (5) is possible for arbitrary field strengths and frequency provided the inequalities (4) are satisfied. Yet, inspecting a typical experimental setup [6] one may consider somewhat weaker external fields and lower frequencies, so that  $|\phi_{L(R)}|, \Omega, W_L, W_R \ll |\epsilon_{d,\sigma}|$ . Expression (6) for  $J_{k'k}$  then significantly simplifies. Matrix elements  $J_{k'k}$  become time - independent if  $k$  and  $k'$  belong to the same lead. Moreover,  $J_{k'k}$  do not depend on potential shifts of every lead separately but only on their difference,  $\Delta_{LR} \equiv \Delta_L - \Delta_R \equiv \phi^{dc} + W \cos(\Omega t + \alpha)$ . For simplicity, we further assume that  $J_{k'k}$  depend only on the leads to which  $k'$  and  $k$  belong, independently of the values of  $k'$  and  $k$ .

The current  $I(t)$  is defined as the rate of change in the number of electrons in a lead. Within the interaction picture, the commutator of the number operator with the Hamiltonian (5) yields an expression for the current,

$$I(t) = \frac{e}{\hbar} \sum_{k' \in L, k \in R; \sigma} \text{Im} \left\{ J_{k'k}(t) \text{Tr}_{F_1} \left[ \rho_0 \hat{T}_p (a_{k',-\sigma}^\dagger(t) a_{k,\sigma}(t) c_{d,\sigma}^\dagger(t) c_{d,-\sigma}(t) S_p) \right] + J_{k'k}(t) \text{Tr}_{F_1} \left[ \rho_0 \hat{T}_p (a_{k',\sigma}^\dagger(t) a_{k,\sigma}(t) c_{d,\sigma}^\dagger(t) c_{d,\sigma}(t) S_p) \right] \right\}, \quad (7)$$

where  $\rho_0$  is the initial (equilibrium) density matrix, while  $S_p$  and  $\hat{T}_p$  are, respectively, the S - matrix and the time - ordering operator on a closed time - path.

We now carry out a perturbation expansion of the above expression in powers of the coupling strength  $J_{k'k}$  using the Schwinger – Keldysh non - equilibrium Green's functions technique. In order to get rid of the constraint to the subspace  $F_1$  we combine it with the method suggested by Coleman [14] for the analogous problem in the Anderson model. The perturbation diagrams are drawn in Fig. 1. Every line corresponds to a  $2 \times 2$  matrix of Green's functions (details are to be published elsewhere). It is important to note that the Hamiltonian (5) (and, consequently, the expression (7) for the current) contains not only the s - d coupling but also the usual potential scattering terms (see Ref. [1]) for explanation). As a result, all possible combinations of  $\sigma$ ,  $\sigma_1$  and  $\sigma_2$  (i.e.  $\sigma_1 = \pm\sigma$ ,  $\sigma_2 = \pm\sigma$ ) are allowed for diagram A but only one of them (namely,  $\sigma_2 = \sigma_1 = \sigma$ ) is allowed for diagram B. The spin - flip diagrams of the type B cancel.

The resulting current is given by the following formulae:

$$\begin{aligned}
I(t) &= I^{(2)}(t) + I^{(3)}(t) \\
I^{(2)}(t) &= C_2 \left[ \phi^{dc} + W \cos(\Omega t + \alpha) \right] \\
I^{(3)}(t) &= \frac{1}{2} I_0 + \sum_{n=1}^{+\infty} |I_n| \cos(n\Omega t + n\alpha + \arg I_n), \\
I_n &= C_3 \sum_{s=-\infty}^{+\infty} J_s \left( \frac{W}{\Omega} \right) J_{s+n} \left( \frac{W}{\Omega} \right) \left[ F(\phi^{dc} + s\Omega, T, D) + (-1)^n F^*(\phi^{dc} - s\Omega, T, D) \right], \\
F(\phi, T, D) &= -\text{Re} \int \int_{-D}^{+\infty} d\omega d\epsilon \frac{[f_L(\omega) - f_R(\omega)][f_L(\epsilon) + f_R(\epsilon)]}{\omega - \epsilon + i\gamma} - i\pi \frac{\phi}{2} \coth \frac{\beta\phi}{2},
\end{aligned} \tag{8}$$

where  $I^{(2)}$  and  $I^{(3)}$  express contributions of second (diagram C) and third (diagrams A and B) orders in  $J_{k'k}\rho$ , while  $C_2 \equiv \frac{e}{\hbar} \pi |\tilde{J}_{LR}|^2 \rho_L \rho_R$  and  $C_3 \equiv \frac{e}{\hbar} 5\pi |\tilde{J}_{LR}|^2 \rho_L \rho_R (\tilde{J}_{LL} \rho_L + \tilde{J}_{RR} \rho_R)$ . The quantities  $\rho_{L(R)}$  are densities of states in the leads, whereas  $J_{L(R)R(L)} = V_{L(R)} V_{R(L)}^* / |\epsilon_{d,\sigma}|$ . The cutoff  $D$  is equal to the energy difference between the chemical potential and the bottom of the conduction band, while  $f_L(\epsilon) \equiv 1 / (\exp[(\epsilon - \phi)/kT] + 1)$  and  $f_R(\epsilon) \equiv 1 / (\exp[\epsilon/kT] + 1)$  are Fermi functions in the leads (the left lead being shifted by  $\phi$ ). The integral in the expression for  $F$  can be written in the same form as in the dc result of Sivan and Wingreen [8]. The alternating field causes splitting of the energy levels in one of the leads [17]. Therefore the time - dependent current is a result of interference between “dc - like” contributions, each one with an effective bias  $\phi^{dc} \pm s\Omega$ .

An approximate evaluation of the double integral in the above formulae is possible both for the linear ( $\phi \ll T$ ) and for the non - linear ( $\phi \gg T$ ) regimes. The result is

$$F(\phi, T, D) = \begin{cases} \phi \ln \frac{D}{kT}, & \text{if } \phi \ll T \\ \phi \ln \frac{D}{|\phi|}, & \text{if } \phi \gg T \end{cases} \tag{9}$$

The terms that are not included in this expression are of order  $\phi$ . It should be stressed that in the nonlinear case on which we focus our attention here the function  $F$  does not diverge as  $T \rightarrow 0$ . Hence, our results for the nonlinear response must be valid even below the Kondo

temperature. This results from the fact that here, the non - linear bias plays the role of temperature as the largest low - energy scale.

Substitution of expression (9) into equations (8) yields simple formulae for the current. First of all, in the absence of an ac - field we obtain,

$$I \approx \phi^{dc} \cdot \begin{cases} C_2 + C_3 \ln \frac{D}{kT}, & \text{if } \phi^{dc} \ll T, \\ C_2 + C_3 \ln \frac{D}{|\phi^{dc}|}, & \text{if } \phi^{dc} \gg T. \end{cases} \quad (10)$$

We note that, to the best of our knowledge, such a simple expression for a non - equilibrium tunneling current through a Kondo system has not been found before. In the presence of a strong ( $W \gg \Omega$ ) ac - field we get,

$$\begin{aligned} I(t) &\approx I^{dc} + I^{ac}(t) \\ I^{dc} &= \phi^{dc} \left[ C_2 + C_3 \ln \frac{D}{A_{dc}(\phi^{dc}, W, \Omega, T)} \right], \\ I^{ac}(t) &= W \cos(\Omega t + \alpha) \left[ C_2 + C_3 \ln \frac{D}{A_{ac}(\phi^{dc}, W, \Omega, T)} \right], \end{aligned} \quad (11)$$

where  $A_{dc}, A_{ac}$  are of the order of  $\max(\phi^{dc}, W)$ , if  $\phi^{dc} \gg T$  or  $W \gg T$  (nonlinear response), and  $A_{dc} = A_{ac} = T$ , if  $\phi^{dc} \ll T$  and  $W \ll T$  (linear response).

In most of the relevant experiments the attention was focused on the dc. The best pronounced feature of the Kondo effect in the dc seems to be the zero - bias anomaly, i.e. appearance of a narrow peak in the differential conductance around zero bias. In Fig. 2 and 3 we show how it is altered by application of the external time - dependent field. According to formula (8), dc in an external ac - field is given by a sum of direct currents without ac - field but with effective biases  $\phi^{dc} + s\Omega$  (these currents are weighted by  $J_s^2$ ). Consequently, the zero - bias anomaly is suppressed. A picture of gradual flattening similar to Fig. 2 holds also at lower frequencies down to the adiabatic limit. At higher frequencies, however, side peaks at multiples of  $\Omega$  can be resolved (Fig. 3).

Although measurement of an alternating current with frequencies and amplitudes in the relevant range is not an easy task, it might reveal new interesting features of the Kondo effect. Expression (11) shows that only the dc and the first harmonic are enhanced by the Kondo effect. For the other harmonics, the interference of the contributions to the current in equations (8) with different effective biases is destructive. We emphasize that the nonlinear response is different from the linear one. Namely, (1) the second and the higher harmonics exist although they are not amplified by the large factor  $\ln \frac{D}{A}$ , (2) the factor  $\ln \frac{D}{T}$  is replaced by  $\ln \frac{D}{A}$ . Nevertheless, they *look* very similar in the sense that the dc and the first harmonic are much larger than the others. This is a particular feature of the interacting (Kondo) system. In the non - interacting one - level system the amplitude of the higher harmonics  $I_n$ ,  $n \geq 2$  is, generally, of the order of the amplitudes of the dc and the first one (gradually decreasing with their number as  $\Gamma/(n\Omega)$ ). In some sense the Kondo system, in contrast with the non - interacting one, behaves like an ordinary resistor (although the current is enhanced by the Kondo effect). Unlike the situation encountered in non - interacting systems, the magnitudes of the dc and the ac components are mostly governed by two independent parameters  $\phi^{dc}$  and  $W$ . Experimentally, Kondo contribution

to the tunneling current is usually revealed through a special dependence on the parameters (such as  $\ln T$  increase of the conductance or zero - bias anomaly). Our formulae (11) imply that this kind of effects can be found in the first harmonic as well as in the dc but not in higher harmonics.

In conclusion, the pertinent physics is rather rich since it combines strong correlations with nonlinearity and time dependence. We obtain (within perturbation theory) rigorous analytical formulae for the nonlinear time - dependent tunneling current in the Kondo regime. They apply for nonlinear response both below and above the Kondo temperature (for linear response they are valid only above it). In the special case of constant external field the  $I - \phi$  relation (10) encodes the main physics of the familiar zero - bias anomaly.

The first novel result of the present research can be easily tested experimentally, since it concerns the *direct* tunneling current. We find that its zero - bias anomaly is suppressed by an ac - field. Second, it is shown that both the zeroth and the first harmonics of the alternating current are strongly enhanced by the Kondo effect, while the other harmonics remain relatively small. This result is slightly more difficult to be tested experimentally. Yet, it is apparently worth the effort since it is remarkably different from what is expected for a non - interacting one - level system where all the harmonics emerge together.

As far as relation to previous relevant works is concerned, we, first, notice that our analytical results for the dc are consistent with the numerical calculations of Ref. [9]. However, being able to consider stronger ac - fields (larger ratio  $W/\Omega$ ), we find also an overall suppression of the zero - bias anomaly, besides the appearance of side peaks. As for the spectrum of the tunneling current, we are unable to validate the assumption suggested in Ref. [9] that *all* the harmonics beside the dc one can be neglected. On the other hand, we verify that the second and higher harmonics are generated but they are indeed much smaller than the dc and the first one [18]. Within a specific model, some authors [10] obtained current spectrum similar to that of a non - interacting system. We attribute the difference between this result and ours to the very peculiar choice of parameters used therein.

*Acknowledgments.* This research was supported in part by a grant from the Israel Academy of Science and Humanities under *Centers of Excellence Program*. We would like to thank Y. Meir, A. Golub, N.S. Wingreen, P. Coleman, K.A. Kikoin, G. Schön, D. Goldhaber - Gordon and E. Kogan for many helpful discussions and comments.

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- [17] An application of a uniform (time – dependent) potential is not observable even if it is arbitrary strong and arbitrary fast (see Y.Goldin and Y.Avishai, Phys. Rev. B, **55**, 16359 (1997), in appendix) so, for interpretation of the results, we can consider one of the leads fixed and the other one alternating. In the present discussion we chose the left lead to move.
- [18] It is true also below the Kondo temperature. We explained it in the text for the nonlinear case. In the linear response the function  $F(\phi, T, D)$  is linear in  $\phi$  by definition. Then it can be exactly proved that only the dc and the first harmonic are generated.

## FIGURES

FIG. 1. Diagrams for the perturbation expansion of the current (7). Solid lines stand for lead electrons, dashed lines — for dot electrons (auxiliary fermions).

FIG. 2. Suppression of the zero - bias anomaly in the direct current by an external alternating field at relatively low frequencies. Differential conductance  $dI^{dc}/d\phi^{dc}$  is given in units of  $C_3T$ , while  $D = 50$ ,  $\hbar\Omega = 5$ ,  $W$  and  $\phi^{dc}$  are in units of  $T$ .

FIG. 3. Suppression of the zero - bias anomaly in the direct current by an external alternating field at higher frequencies. Side peaks appear at multiples of  $\Omega$ . Differential conductance  $dI^{dc}/d\phi^{dc}$  is given in units of  $C_3T$ , while  $D = 200$ ,  $\hbar\Omega = 15$ ,  $W$  and  $\phi^{dc}$  are in units of  $T$ .









