Nonequilibrium Kondo effect in asymmetrically coupled quantum dots

M. Krawiec and K. I. Wysokiński
Institute of Physics, M. Curie-Skłodowska University, ul. Radziszewskiego 10A, 20-031 Lublin, Poland

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A quantum dot asymmetrically coupled to external leads has been analyzed theoretically by means of the equation-of-motion technique and the noncrossing approximation. The system has been described by the single-impurity Anderson model. To calculate the conductance across the device the nonequilibrium Green’s function technique has been used. The obtained results show the importance of the asymmetry of the coupling for the appearance of the Kondo peak at nonzero voltages and qualitatively explain recent experiments.

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I. INTRODUCTION

Recent advances in nanotechnology have allowed the fabrication of structures containing quantum dots coupled to the external environment. The quantum dot consists of a finite number of electrons confined to a small region of space. It behaves like an impurity in a metal and allows the study of the many-body correlations between electrons. However, unlike an impurity, the parameters of which are fixed, the coupling of a quantum dot to external leads and its other parameters can be changed in a highly controlled way. Most importantly nonequilibrium transport can also be studied.

The discovery of the Kondo effect in quantum dots connected to external leads by tunnel junctions has resulted in an increased experimental interest in this many-body phenomenon. The Kondo effect in the quantum dot manifests itself at temperatures lower than the Kondo temperature $T_K$ as an increased conductance $G$ through the system. It is due to the formation of the so-called Abrikosov-Suhl or Kondo resonance at the Fermi energy. This is a many-body singlet state involving spin on the quantum dot and electrons in external leads.

The experiments have confirmed the validity of the theoretical picture. They also discovered phenomena, the explanation of which requires new theoretical ideas. In particular they have shown a nonorthodox and unexpected behavior of the systems in the Kondo regime. These are $inter alia$ absence of the odd-even parity effects expected for these systems and observation of the singlet-triplet transition in a magnetic field. Besides the nonlinear current-voltage characteristics it has been possible to measure the charge distribution which led to the conclusion of spin-charge separation in the Kondo regime, observe the evolution of the transmission phase, and detection of two different energy scales related to two stages of the spin screening process in systems with spin $S \gg 1$, with one of the Kondo temperatures as high as 4 K.

Great progress in the theoretical understanding of Kondo physics in real quantum dots has been made during last decade. The theory has concentrated on such important aspects as Kondo-driven transport in multilevel quantum dots, coupled quantum dots, double-dot structures in which the existence of the Kondo effect without spin degree of freedom and new singlet-triplet effects have been predicted, the nature (weak versus strong coupling) of the Kondo effect at high voltage, the spin-charge separation in the strongly correlated quantum dot, systems driven out of equilibrium by different means, etc.

Here we shall focus our attention on the experimental observation of the Kondo effect at nonzero source drain voltages. To state the problem in the right perspective let us recall that in systems containing a quantum dot, the Kondo effect manifests itself at low temperatures as an enhanced conductance observed at zero source-drain voltage, $V_{SD} = 0$. Occasionally, a Kondo peak in conductance appearing at nonzero voltages $V_{SD} \neq 0$ (Ref. 11) has been observed and this unusual behavior, called the anomalous Kondo effect, remains unexplained. Recently this phenomenon has been studied systematically. The authors have fabricated the dot coupled weakly to one and strongly to another lead and observed the evolution of the peak at $V_{SD} \neq 0$. The source-drain voltage $V_{SD}$, at which the peak appears, scales roughly linearly with a gate voltage, $V_g$. In the experimental setup the additional electrode determines the asymmetry in the coupling of the dot to left and right leads.

It is the purpose of this paper to study the anomalous Kondo peak observed at nonzero voltage. We shall present the results of the model calculations based on the nonequilibrium transport theory applied to the quantum dot described by the Anderson model with asymmetric coupling to the leads. As we shall see the asymmetry in the couplings is the main factor which leads to this anomalous Kondo effect. The experimentally observed shifts of the Kondo peak to higher values of $V_{SD}$ with increasing gate voltage can be satisfactorily explained by assuming that the values of the left and right barriers change together with the gate voltage, while the asymmetry in the couplings remains constant. This scenario is realized in experiment.

The organization of the rest of the paper is as follows. In Sec. II we introduce the model, give the formula for the current through the quantum dot, and discuss briefly the methods (equation of motion (EOM) with a slave boson representation of the electron operators and noncrossing approximation (NCA)) used to calculate the on-dot Green’s function relegating some technical details to the Appendix. In Sec. III we present the results of our numerical calculations of the tunneling conductance across the asymmetrically coupled single-level quantum dot in $U = \infty$ limit. Conclusions are given in Sec. IV.
II. THEORY

For the sake of simplicity we discuss here the dot with single energy level. The theoretical analysis of the transport through quantum dot usually starts with the following, Landauer-type, formula\(^2\) for the current:

\[
J = \frac{e}{\hbar} \sum_{\sigma} \int d\omega [f_{\sigma}(\omega) - f_{\bar{\sigma}}(\omega)] \\
\times \frac{G_{\sigma}^{R}(\omega)G_{\bar{\sigma}}^{R}(\omega)}{G_{\sigma}^{L}(\omega) + G_{\bar{\sigma}}^{R}(\omega)} \left( -\frac{1}{\pi} \right) \text{Im} G_{\sigma}^{r}(\omega + i0^{+}).
\]

(1)

Here \(f_{\sigma}(\omega)\) denotes the Fermi distribution function for lead \(\lambda\) with chemical potential \(\mu_{\lambda}\), \(G_{\sigma}^{r}(\omega)\) is the (retarded) impurity Green’s function, and \(G_{\sigma}^{L}(\omega) = 2\pi \sum_{\varepsilon} |V_{\lambda k}|^2 \delta(\omega - \varepsilon_{\lambda k})\) is the effective coupling of localized electrons to the conduction band.

The current \(J\) flowing across the system depends on the source-drain voltage \(V_{SD} = (\mu_{L} - \mu_{R})/e\), where \(e\) is the electron charge. The differential conductance of the system defined as \(G(V_{SD}) = dJ(V_{SD})/dV_{SD}\) is directly measured experimentally.\(^3\)

To calculate the on-dot Green’s function \(G_{\sigma}(\omega + i0^{+})\) we shall describe the dot coupled to the external leads by the single-impurity Anderson Hamiltonian\(^2\)

\[
H = \sum_{\lambda k \sigma} \varepsilon_{\lambda k} c_{\lambda k \sigma}^\dagger c_{\lambda k \sigma} + E_{d} \sum_{\sigma} d_{\sigma}^\dagger d_{\sigma} + U n_{\uparrow} n_{\downarrow} \\
+ \sum_{\lambda k \sigma} (V_{\lambda k} c_{\lambda k \sigma}^\dagger d_{\sigma} + H.c.).
\]

(2)

Here \(\lambda = R, L\) denote the right (R) or the left (L) lead in the system. Other symbols have the following meaning: \(c_{\lambda k \sigma}^\dagger (c_{\lambda k \sigma})\) denotes the creation (annihilation) operator for a conduction electron with wave vector \(k\), spin \(\sigma\) in the lead \(\lambda\), and \(V_{\lambda k}\) is the hybridization matrix element between conduction electron of energy \(\varepsilon_{\lambda k}\) in the lead \(\lambda\) and the localized electron on the dot. \(E_{d}\) is the single-particle energy at the dot. \(n_{\pm} = d_{\sigma}^\dagger d_{\sigma}\) is the number operator for electrons with spin up localized on the dot and \(U\) is the (repulsive) interaction energy between two electrons. Our calculations are restricted to very low temperatures, much smaller than the orbital level spacing in a quantum dot, so it is legitimate to consider the single energy level \(E_{d}\).

There are various methods\(^2\) of calculating the on-dot Green’s function entering the current (1). Here we shall apply two of them: the EOM method and NCA. In both cases we assume that the Coulomb repulsion \(U\) between electrons on the dot is the largest energy scale. Therefore we take the limit \(U = \infty\). The original correlated electron operators are expressed as products of auxiliary fermion and boson ones.\(^2\)

When using the EOM method we apply a mean-field-like approximation for the slave bosons and calculate all matrix elements of the Keldysh Green’s functions, including the distribution one.\(^2\) In the process we consistently decouple all elements of the higher-order Keldysh Green functions.\(^2\) The relevant formulas and some technical details can be found in the Appendix. As we shall see the method gives the correct position of the Kondo peak. However, like the standard EOM method it leads to an incorrect width of the peak and occupations. Therefore we have used the NCA, which is the generally accepted technique of solving the problem at hand.\(^3\) In the NCA one maps the infinite-\(U\) Anderson model onto the slave boson one and calculates both boson and fermion propagators. They are expressed by the coupled integral equations.\(^2\)

Where appropriate, we shall present the results obtained by both techniques.

III. NUMERICAL RESULTS

Let us first discuss the relation between the experimental parameters and those entering the model and theory. The effective couplings \(\Gamma_{L}\) and \(\Gamma_{R}\) have been estimated in Ref. 12 to be 170 and 80 \(\mu\)eV, respectively. Their values and the ratio \(\Gamma_{L}/\Gamma_{R}\approx2\) have been argued to remain constant during the measurements. The source-drain voltage \(V_{SD}\) is the difference of the chemical potentials of the external leads. The (back)gate voltage \(V_{g}\) controls the position of the on-dot energy level \(E_{d}\). As already mentioned we stick here to the \(U = \infty\) limit. In this limit there can be at most a single electron with energy \(E_{d}\) on the dot at a time.

We start the presentation of the results with a comparison of the (equilibrium) density of states of a quantum dot coupled to two leads obtained by means of the NCA and EOM approaches. This is shown in Fig. 1.

The main features of the density of states (DOS) remain the same in both approaches. However, the height and width of the Kondo peak are much larger in the NCA. Moreover, the spectral weight is shifted towards higher energies. In turn this leads to the different occupations shown in the inset of Fig. 1.

Now let us turn to the nonequilibrium (\(\mu_{L} \neq \mu_{R}\) density
of states. In this case the high-energy features to large extent remain the same as in equilibrium (see Fig. 1), so the only low-energy DOS is shown in Fig. 2. The upper panel presents results obtained via the equation-of-motion technique for the Keldysh matrix Green’s function. In the lower panel the results obtained with the noncrossing approximation are shown. The coupling is asymmetric with $G_L = 2G_R$ (dashed lines) and $G_L = \frac{1}{2}G_R$ (dotted lines). $\mu_L = -\mu_R = 0.2$ and the other parameters are the same as in Fig. 1.

The upper panel presents results obtained via the equation-of-motion technique for the Keldysh matrix Green’s function. In the lower panel the results obtained with the noncrossing approximation are shown. The coupling is asymmetric with $G_L / G_R = 2$ (dashed lines) and $\frac{1}{2}$ (dotted lines). The case of symmetric coupling $G_L = G_R$ is also shown (solid lines) for comparison. A few features have to be noted. First we see that the Kondo peak is always located at energies coinciding with those of the Fermi levels of the leads. Thus in nonequilibrium we get (in the density of states) two Kondo resonances pinned to Fermi energies of the left and right electrodes. Note also that the heights of the respective Kondo resonances strongly depend on the value of the hybridization. The overall shape of the density of states is similar. The positions of the Kondo peaks are roughly the same but they differ in width and height. The peaks obtained in EOM are much narrower and smaller. As a result the curves in Fig. 2(a) differ from that in Fig. 2(b).

These details in the energy dependence of the density of states may shortly be a matter of direct measurements. In fact it has been recently predicted theoretically that the on-dot density of states can be measured in a device containing a quantum dot coupled to three leads. The very weakly coupled third lead will act as a tunneling tip in conventional tunneling microscopes and will probe the nonequilibrium density of states. The conductance spectrum measured by this additional electrode has been shown to follow the nonequilibrium density of states, like one shown in Fig. 2.

Returning to our main subject we show in Fig. 3 the differential conductance spectrum corresponding to the same “experimental setup” as discussed previously in connection with Fig. 2.

For comparison we have also plotted in this figure the conductance through the symmetrically coupled quantum dot. In the symmetric situation ($\Gamma_L = \Gamma_R$) the Kondo resonance is located exactly at zero bias ($V_{DS} = 0$), but for $\Gamma_L > \Gamma_R$ ($\Gamma_L < \Gamma_R$) it is shifted to the negative (positive) voltages $V_{DS}$. This finding is in nice qualitative agreement with experimental studies on transport through a quantum dot in the presence of asymmetric barriers.12 While the calculated spectra within EOM and NCA show the same shifts of the
Kondo peak, their shapes are different. The NCA peaks are much higher and more symmetric in the vicinity of their maxima. For asymmetric coupling the Kondo resonance in the conductance is pinned to the position of the Fermi level of that lead which is more strongly coupled (larger $\Gamma$) to the dot. It is thus mainly the relative coupling which rules the value of the shift.

In Fig. 4 we show the systematic change of the $G(V_{DS})$ with increasing asymmetry $\Gamma_L/\Gamma_R$ of the coupling.

The upper curves in both panels correspond to $\Gamma_L/\Gamma_R = 5.5$ while lower ones are for symmetric coupling $\Gamma_L/\Gamma_R = 1$ in steps of 0.5. The increase of the asymmetry $\Gamma_L/\Gamma_R$ from 1 to 5.5 continuously moves the Kondo peak away from $V_{SD}=0$ position. We have checked that increasing the asymmetry to still higher values does not lead to bigger shifts. This is easy to understand as for large asymmetry one of the barriers is not transparent enough to produce clear Kondo resonance in the density of states.

The position of the on-dot electron energy level $E_d$ influences the anomalous Kondo peak for an asymmetric dot with $\Gamma_L/\Gamma_R > 2$ to a lesser extent. It is only important that it takes a value appropriate for observing a Kondo resonance. For all appropriate $E_d$ the shifts are of comparable magnitudes.

The data displayed in Fig. 4 qualitatively agree with those plotted in Fig. 5 of Ref. 11 and Fig. 3 of Ref. 12. However, theoretical shifts of the Kondo peak position are smaller than the experimental.

There may be additional factors which affect the position of the peaks. We have checked that the energy dependence of $\Gamma_{LR}$ introduces only small quantitative differences in the density of states and differential conductance, and does not lead to better agreement between theory and experiment. Similarly calculations within the EOM approach for finite $U$ show that finite $U$ leads to minor corrections as also does the presence of additional energy levels in the vicinity of the Fermi energy. In all cases studied one gets usual behavior with the Kondo peak located at $V_{SD}=0$ for symmetric coupling to both leads and an anomalous Kondo effect for asymmetric coupling. This proves the importance of asymmetry in the observation of the effect.

In the experimental setup\cite{15} the changes of the gate voltage $V_g$, which in the first place affect the position of the electron energy level, also modify the height of the barriers and their transparency $V_k$. This effect is of special importance in the quantum dots defined in the two-dimensional electron gas where the voltage at a single electrode couples capacitively to other electrodes.\cite{26} If we assume that (as in experiment) $\Gamma_L/\Gamma_R$ remains constant (=2) and that the decrease of the energy $E_d$ is accompanied by the simultaneous increase of the couplings $\Gamma_R$ and $\Gamma_L$, then the calculated shifts get larger.

The occurrence of the Kondo resonance is possible at low enough temperature. It is also well known\cite{2} that changes of temperature move slightly the Kondo peak. We have checked this and found that if the temperature is raised, the position of the peak moves slightly away from the $V_{SD}=0$. At finite temperature the occupation of the dot changes and the Abrikosov-Suhl resonances smear out and this leads to small

FIG. 4. The differential conductance obtained within the (a) EOM and (b) NCA for the different asymmetric couplings. The lower curve is for $\Gamma_L=\Gamma_R$ while the upper one $\Gamma_L=5.5\Gamma_R$. Other parameters are $E_d=-10$ and $T=10^{-2}$ in units of $\Gamma_R$.

FIG. 5. The NCA calculated differential conductance as a function of source-drain voltage $V_{SD}$ for various values of $E_d$ and $\Gamma_L$, $\Gamma_R$ at fixed $\Gamma_L/\Gamma_R=2$. The lowest curve corresponds to $E_d = -3$, $\Gamma_L=1$, while upper one is for $E_d = -12$, $\Gamma_L=6.4$ in units of $\Gamma_0$ equal to the initial coupling of the left lead. The temperature $T=5\times10^{-2}$ is below estimated Kondo temperatures.
changes in the position of the Kondo peak.

We thus have combined all above contributions, i.e., asymmetry in the couplings $\Gamma_L \neq \Gamma_R$, their $E_d$ dependence, and assumed high enough, but still below $T_K$, the temperature to get larger shifts of the Kondo peak.

We have shown the results in Fig. 5. The various curves have been calculated for $T=5 \times 10^{-3}$ which is below the Kondo temperature. In the figure the change of the position of the on-dot energy level is accompanied by a simultaneous change of the barrier transparency. The bottom curve in Fig. 5 corresponds to $E_d=-3\Gamma_0$, $\Gamma_0=\Gamma_L$, while the upper one corresponds to $E_d=-12\Gamma_0$, $\Gamma_L=6.4\Gamma_0$. Here $\Gamma_0$ is equal to the experimentally estimated value of the smaller of couplings. The ratio $\Gamma_R/\Gamma_L$ is kept constant and equal to 2 as estimated in Ref. 12. The data are in nice qualitative agreement with experiments. The theoretical shifts, however, are smaller than experimental by a factor of 5–10. To check whether this is due to the different asymmetry ratio we plot in Fig. 6 the results obtained for $\Gamma_R/\Gamma_L=4$. The shifts have increased.

IV. CONCLUSIONS

We have found that the emergence of the Kondo peak at nonzero voltages $V_{DS} \neq 0$ is caused by asymmetric coupling of the dot to external electrodes. These results are in qualitative agreement with experimental data on transport through a quantum dot asymmetrically coupled to the leads. The theoretical Kondo peaks in differential conductance, however, are narrower than the experimental ones. Their maxima move to nonzero $V_{SD}$ with increasing asymmetry or position of the on-dot energy level. The simultaneous change of $E_d$ and $\Gamma_L$, $\Gamma_R$ can semiquantitatively explain the experimental data. More experimental results are needed to draw firm conclusions such as the applicability of the simple Anderson model to asymmetrically coupled quantum dots. It follows from the presented studies that the asymmetry of the couplings is a necessary ingredient for explanation of the anomalous Kondo effect. Within the Anderson model one always gets a normal Kondo effect for symmetric couplings and small shifts of the Kondo peak to nonzero voltages for asymmetric couplings. Our inability to explain quantitatively the experimental data may indicate the necessity of a much better theoretical treatment of the model or even a better model for a description of these complicated systems. There is the possibility that the experimentally observed features, even though similar to, do not represent a genuine Kondo effect. In fact some recent work has seen very small shifts, consistent with the present calculations, even for quite asymmetrically coupled quantum dots.

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APPENDIX

To find the current across the system, Eq. (1), it is enough to calculate the on-dot retarded Green’s function (GF).

The NCA method to get $G^{\nu}_{\sigma}(\omega)$ has been extensively discussed previously and there is no need to repeat its derivation again. For the sake of completeness let us only note that we have adapted the formulas derived in the second paper of Ref. 2.

The EOM method to calculate the GF is straightforward and in the $U \rightarrow \infty$ limit leads to

$$G^{\nu}_{\sigma}(\omega) = \frac{1 - \langle n_{-\sigma} \rangle}{\omega - E_d - \sum_{\nu} \Sigma^{\nu}_{\sigma\nu}(\omega)},$$

with the self-energy

$$\Sigma^{\nu}_{\sigma\nu}(\omega) = \sum_{\lambda k} |V_{\lambda k}|^2 \frac{1 + f(\omega - \mu_\lambda)}{\omega - \epsilon_{\lambda k}}.$$

In Eq. (A1), $\langle n_{-\sigma} \rangle$ denotes the average on-dot occupation number of the spin-$(\sigma)$ electrons. In equilibrium one calculates $\langle n_{-\sigma} \rangle$ self-consistently from the retarded Green’s function $G^{\nu}_{\sigma}(\omega)$. Here we are dealing with a nonequilibrium situation and $\langle n_{-\sigma} \rangle$ cannot be calculated directly from $G^{\nu}_{\sigma}(\omega)$. Instead the nonequilibrium Green’s function technique has to be used. The occupation of the dot at time $t$ is expressed via the Keldysh lesser Green’s function of the dot$angle(t)$ for the Keldysh lesser Green’s function

$$\langle n_{\sigma}(t)\rangle = \langle c^\dagger_{\sigma}(t) c_{\sigma}(t)\rangle = -i G^{\nu}_{\sigma}(t, t).$$

In the steady state one gets

$$\langle n_{\sigma}(t)\rangle = -i \int_{-\infty}^{\infty} d\omega \sum_{\nu} G^{\nu}_{\sigma}(\omega).$$

This shows that consistent calculations of the retarded GF require knowledge of “lesser” one. The equation of motion for the lesser GF has been formulated by Niu et al. For the Hamiltonian of the form $H=H_0+H_I$ they derived the following general equation for the lesser GF:
\[ \langle A | B \rangle_s = g^<(\omega) \langle [A, B]_L \rangle + g^>(\omega) \langle [A, H_I] B \rangle_s \]

\[ + g^<_{\omega} \langle [A, H_I] B \rangle \tilde{\omega} \quad (A4) \]

here, \( g^{<_{\omega}}(\omega) \) is the lesser (retarded) GF of the noninteracting part \( H_0 \) of Hamiltonian.

To treat strong correlations we use the version\(^\text{24}\) of the slave boson technique and rewrite the Hamiltonian in the form

\[ H^{SB} = \sum_{\lambda k} (\epsilon_{\lambda k} - \mu_{\lambda}) c^\dagger_{\lambda k} c_{\lambda k} + \sum_{\lambda} F_{\lambda}^+ F_{\lambda} \]

\[ + \sum_{\lambda k} V_{\lambda k} \left( c^\dagger_{\lambda k} b^+ + f^+_\sigma c_{\lambda k} \right), \quad (A5) \]

where new fermionic \( (f^+_\sigma, f^-_\sigma) \) and bosonic \( (b^+ \lambda, b) \) operators have been introduced. Calculating the on-dot Green’s function \( G^\sigma_\omega(\omega) = \langle [b^+_\sigma b]_\omega \rangle \) we have taken the third term of \( H^{SB} \) as an interaction part \( H_I \) and the first two terms of it as \( H_0 \).

The average occupation number is found to be

\[ \langle n \rangle = -\frac{1}{2\pi} \sum_{\omega} \sum_{\lambda} \text{Im} \Sigma^\sigma_\omega(\omega) \langle f^+_\lambda(\omega) | G^\sigma_\omega(\omega) | f^+_\lambda(\omega) \rangle. \]

\[ (A6) \]

Note that in turn it depends on the retarded Green’s function. This closes the system of equations.

27 D. Goldhaber-Gordon (private communication).