Double quantum dots: interdot interactions, co-tunneling, and Kondo resonances without spin

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Abstract

We show that through an interdot off-site electron correlation in a double quantum-dot (DQD) device, Kondo resonances emerge in the local density of states without the electron spin-degree of freedom. We identify the physical mechanism behind this phenomenon: rather than forming a spin singlet in the device as required in the conventional Kondo physics, we found that exchange of electron position between the two quantum dots, together with the off-site Coulomb interaction, are sufficient to induce the Kondo resonance. Due to peculiar co-tunneling events, the Kondo resonance in one dot may be pinned at the chemical potential of the other one.

72.15.Qm, 73.40.Gk, 73.23.Hk
The recent discovery of Kondo effect in semiconductor quantum dots (QD) [1,2] has generated tremendous interests both theoretically and experimentally. When a QD is contacted by two metallic leads, Kondo effect arises as a result of coherent superposition of co-tunneling processes [2,3] such that, at low temperatures, a very narrow Kondo resonance emerges in the density of states of the QD at the free electron chemical potential $\mu$, and a spin singlet is formed in the whole system of the QD plus the leads. Kondo effect has so far been investigated in many systems and its understanding provided important insights to strongly interacting electrons in nanostructures. In most previous theoretical investigations, the physical systems under consideration all possess three common and fundamental ingredients: the existence of co-tunneling (i.e. exchange) events, the role of the electron spin degrees of freedom, and finally the on-site Coulomb interaction $U_o$. Because the QD Kondo effect is a result of many-body correlation, the existence of a great deal of co-tunneling events and the Coulomb interaction are crucial. However, must it be the on-site interaction? Are spin degrees of freedom essential? Although no exception has been found so far, these questions are intriguing because their understanding will provide a clear picture to the deciding physical factors of QD Kondo phenomenon. Our investigation found that QD Kondo effect does occur even without considering the spin degrees of freedom and without the on-site Coulomb energy — provided that there are spatial degrees of freedom and there is an off-site interaction. It is the purpose of this letter to report our investigations on these issues.

In particular, we investigate a two-probe double QD (DQD) device in which the spin degeneracy is neglected. The two QDs provide a spatial degree of freedom for co-tunneling processes (see below) and there is an off-site e-e interaction between the dots. It should be mentioned that conventional Kondo effect, i.e. the Kondo effect caused by the three fundamental ingredients mentioned in the last paragraph, in DQD systems have been the subject of several recent studies [5-7]. These investigations considered effects of the intradot on-site interaction $U_o$ and/or interdot magnetic spin exchange. The latter is not an independent effect as it is determined by the (intradot) interaction $U_o$ and the interdot tunnel
coupling \[ \text{[7]} \]. The present work, however, focuses on Kondo effect induced by the interdot off-site interaction \( U \) without the spin degrees of freedom. Our main findings are: (i) At low temperatures there are Kondo resonances in the local density of states (LDOS) of the DQD which occur without the entire system forming a spin singlet; (ii) If the interdot tunnel coupling is zero, the Kondo resonance in one QD is pinned at the chemical potential of the lead that is attached to the other dot; (iii) At a non-zero interdot tunnel coupling, this Kondo resonance is split so that sharp features including peaks and abruptly drops emerge in the conductance of the device. These results are significant because they show that the Kondo resonance can occur by an off-site interaction even without the electron spin-degrees of freedom.

Our system of DQD coupled with two leads is described by the following Hamiltonian,

\[
H = H_{DQD} + H_T + \sum_{\alpha = L,R} H_{\alpha},
\]

where\( H_{\alpha} = \sum_k \epsilon_{\alpha k} \hat{a}_\alpha^\dagger \hat{a}_{\alpha k} \) describe the noninteraction left/right lead; \( H_T = \sum_{k,\alpha} (t_{\alpha} \hat{a}_{\alpha k}^\dagger \hat{d}_\alpha + H.c.) \) denotes the tunneling between the left (right) QD and the left (right) lead. \( H_{DQD} = \sum_\alpha \epsilon_{\alpha} \hat{d}_\alpha^\dagger \hat{d}_\alpha + U_{d_L} \hat{d}_L^\dagger \hat{d}_L + U_{d_R} \hat{d}_R^\dagger \hat{d}_R + t_C (\hat{d}_L^\dagger \hat{d}_R + H.c.) \) models the coupled double QD, in which each QD has only a single level without spin index. The purpose of neglecting the spin index is to emphasize that fact that for this system, Kondo effect occurs without the need of spin-degrees of freedom. Realistically, if a high magnetic field is applied to the QDs, leading the spin separation being larger than the QD energy level spacing \[ \text{[8]} \], the opposite spin state essentially does not affect tunneling at low bias because its contribution is actually weaker than that of the first excited QD state. In this situation the spin-degrees of freedom can be neglected. Finally, when bias voltage \( V \) is not very high, only one eigenstate on each QD is active, therefore the on-site interaction \( U_o \) of each QD can be absorbed into its single particle level \( \epsilon_\alpha \) \( (\alpha = L, R) \) \[ \text{[9]} \] and only the interdot off-site interaction \( U \) will be considered.

Our theoretical analysis is from the Keldysh nonequilibrium Green’s functions. The current from the left lead flowing into the left QD can be expressed as \( (\hbar = 1) \) \[ \text{[10]} \]:

\[
I = -2e I m \int \frac{d \epsilon}{2 \pi} \Gamma_L \{ f_L(\epsilon) G_{LL}^r(\epsilon) + \frac{1}{2} G_{LL}^< (\epsilon) \},
\]

where \( f_\alpha(\epsilon) \) is the Fermi distribution of the \( \alpha \) lead, and \( \Gamma_\alpha = 2\pi \sum_k |t_\alpha|^2 \delta(\epsilon - \epsilon_k) \). The Green’s function \( G_{LL}^{r, <} (\epsilon) \) are the Fourier transformation
of \( G^r_{LL}(t) \), and \( G^r_{LL}(t) \equiv -i\theta(t) < \{d_L(t), d_L^\dagger(0)\} > \), \( G^i_{LL}(t) \equiv i < d_L^\dagger(0)d_L(t) > \). Using the standard equation of motion technique and taking the familiar decoupling approximation \([11]\), we have solved \( G^r(\epsilon) \) in the infinite \( U \) limit \( (U \to \infty) \) to be:

\[
\begin{pmatrix}
\epsilon - \epsilon_L - \Sigma^0_L - \Sigma^c_L - \Sigma^b_R - \Sigma^d - t_R - \Sigma^d \\
- \Sigma^d - \epsilon_R - \Sigma^0_R - \Sigma^c_R - \Sigma^b_L - \Sigma^d - t_C - \Sigma^d
\end{pmatrix}
\begin{pmatrix}
G^r_{LL} \\
G^r_{RR}
\end{pmatrix}
= \begin{pmatrix}
1 - n_R < d_R^\dagger d_R > \\
< d_R^\dagger d_R > 1 - n_L
\end{pmatrix}
\begin{pmatrix}
G^r_{LR} \\
G^r_{RL}
\end{pmatrix}
\]

where \( \Sigma^d = \Sigma^d_L + \Sigma^d_R \). \( \Sigma^0(\epsilon) = \sum_k |t_\alpha|^2/(\epsilon - \epsilon_{ak} + i0^+) \) is the lowest-order self-energy for a noninteraction system; the higher-order self-energy \( \Sigma^b, \Sigma^c, \Sigma^d \) are:

\( \Sigma^b(\epsilon) = \sum_k [(\epsilon - \epsilon_{ak})(\epsilon + \Delta\epsilon - \epsilon_{ak}) - 2|t_C|^2]B_{ak}(\epsilon), \Sigma^c(\epsilon) = \sum_k 2|t_C|^2B_{ak}(\epsilon), \) and \( \Sigma^d(\epsilon) = \sum_k (\epsilon + \Delta\epsilon - \epsilon_{ak})t_CB_{ak}(\epsilon), \)

with \( B_{ak}(\epsilon) = |t_\alpha|^2f_\alpha(\epsilon_{ak})/(\epsilon - \epsilon_{ak})/[(\epsilon - \Delta\epsilon - \epsilon_{ak})(\epsilon + \Delta\epsilon - \epsilon_{ak}) - 4|t_C|^2] \) and \( \Delta\epsilon = \epsilon_L - \epsilon_R \). The quantity \( n_\alpha = < d_\alpha^\dagger d_\alpha > \) in Eq.\((1)\) is the intradot electron occupation number, \( < d_R^\dagger d_R > \) and \( < d_R^\dagger d_L > \) are the interdot correlation due to the interdot tunnel coupling and the interaction. They must be calculated self-consistently: \( < d_R^\dagger d_\beta > = -i \int \frac{d\epsilon}{2\pi} G^i_{\beta\alpha}(\epsilon) \). It should be mentioned that although the equation of motion method only gives a correct qualitative physics \([12]\), it is sufficient for the purpose here.

Next we solve the Keldysh Green’s function \( G^<(\epsilon) \). For interacting systems, \( G^<(\epsilon) \) cannot be obtained from equation of motion without introducing additional assumptions \([12, 13]\). In previous work, various approximations were developed including the use of an ansatz for interacting lesser and greater self-energy \([13]\); the noncrossing approximation \([12]\), and so on. Any of them could be sufficient for our purposes. However, we note that since only \( \int d\epsilon G^<(\epsilon) \) is actually needed for calculating both the electric current and for iterating the self-consistent equation, one does not need to solve \( G^<(\epsilon) \) itself. We found that quantity \( \int d\epsilon G^<(\epsilon) \) can actually be solved \emph{exactly}, allowing us to bypass any approximation involved in computing \( G^<(\epsilon) \). We now present this technical advance which is quite general and can be applied to many other systems. As stated above, \( \int d\epsilon G^<_{\beta\alpha}(\epsilon) \sim < d_\alpha^\dagger d_\beta > \), we therefore proceed to first derive the operator \( d_\alpha^\dagger(t)d_\beta(t) (\alpha, \beta = L, R) \) over time \( t \), and then take average \( < \frac{d}{dt}[d_\alpha^\dagger(t)d_\beta(t)] > = 0 \). The last equality is true because we consider steady state
transport. On the other hand, the time derivative can be explicitly calculated using the Heisenberg equation, and we obtain

\[
(\epsilon_\alpha - \epsilon_\alpha - i \frac{\Gamma_L + \Gamma_R}{2}) < d_{\alpha}^\dagger d_{\alpha} > + t_C < d_{\alpha}^\dagger d_{\alpha} > - t_C < d_{\alpha}^\dagger d_{\alpha} > = \int \frac{d\epsilon}{2\pi} \Gamma_\alpha f_\alpha G_{\alpha\alpha}^r - \int \frac{d\epsilon}{2\pi} \Gamma_\alpha f_\alpha G_{\alpha\alpha}^a
\]

(2)

\[
- t_C < d_{\alpha}^\dagger d_{\bar{\alpha}} > + t_C < d_{\alpha}^\dagger d_{\bar{\alpha}} > - i\Gamma_{\bar{\alpha}} < d_{\alpha}^\dagger d_{\bar{\alpha}} > = \int \frac{d\epsilon}{2\pi} \Gamma_{\bar{\alpha}} f_{\bar{\alpha}} [G_{\alpha\bar{\alpha}}^r - G_{\alpha\bar{\alpha}}^a]
\]

(3)

where \( \bar{\alpha} = R \) for \( \alpha = L \), or \( \bar{\alpha} = L \) for \( \alpha = R \). From Eqs.(2,3) and having already solved \( G^r(\epsilon), < d_{\alpha}^\dagger d_{\beta} > (i.e. \int d\epsilon G_{\beta\alpha}^\leq(\epsilon)) \) can be exactly solved and current can now be obtained without further difficulty. In our numerical evaluations, we assume square bands of width \( 2W \) and symmetric coupling barriers so that \( \Gamma_L(\epsilon) = \Gamma_R(\epsilon) = \Gamma \theta(W - |\epsilon|) \), with \( W = 1000 \gg \max(k_B T, eV, \Gamma) \). Here \( V = \mu_L - \mu_R \) is the bias voltage. We also fix \( \Gamma = 1 \) as energy unit and let \( \mu_L = -\mu_R \).

First, we investigate LDOS of the DQD device. [14] Because the LDOS of the right dot is similar to that of the left one, we only discuss the latter. Consider equilibrium situation (bias \( V = 0 \)) and the case where the two dots have equal level position (\( \Delta \epsilon \equiv \epsilon_L - \epsilon_R = 0 \)), the LDOS is shown as the right inset of Fig.1d in which various curves correspond to different values of the interdot tunnel coupling \( t_C \). When there is no tunnel coupling (\( T_C = 0 \), thin solid line), besides a broad main peak at \( \epsilon \sim -1.5 \) (not shown) which corresponds to the renormalized left dot level \( \epsilon_L \), a sharp peak emerges at energy \( \epsilon = \mu \) (now \( \mu = \mu_L = \mu_R \)). This peak has the typical Kondo characteristic: it only exists at low temperature and is pinned at \( \epsilon = \mu \). Turning on the tunnel coupling (\( t_C \neq 0 \)) between the two dots, the Kondo peak is split into two smaller peaks at \( \epsilon = \mu \pm 2t_C \) and whose weight are \( \frac{1}{2} \) [15]. Setting the off-site Coulomb energy \( U = 0 \), the Kondo peaks disappear. Clearly, it is the interdot off-site Coulomb interaction \( U \) that induces the Kondo peak.

The origin of the Kondo peaks at \( t_C = 0 \) is from an interesting co-tunneling process which is shown in the left inset of Fig.1d. When \( \epsilon_L, \epsilon_R < \mu_L, \mu_R < \epsilon_L + U, \epsilon_R + U \), the DQD has only one electron (e.g. it occupies the right dot) and the first order tunneling is blocked
by the off-site interaction. However, due to Heisenberg uncertainty principle, higher-order co-tunneling processes consisting of two virtual tunneling events can still take place. In such a process, the electron in the right dot tunnels to the Fermi level of the right lead, followed by an electron in the left lead at the Fermi level tunneling into the left dot, on a very short time scale $\sim \hbar/\left(\mu_R - \epsilon_R\right)$. This co-tunneling process, in effect, moved the original electron from the right dot to the left one. At low temperatures, a frequent exchange of electrons between the two dots occur through this process even though $t_C = 0$, and a coherent superposition of all possible co-tunneling processes of this type produces the Kondo resonance discussed in the last paragraph in which a very narrow peak emerges at $\epsilon = \mu$ in the LDOS curve of left (right) dot. We emphasize that this mechanism of inducing a Kondo resonance is qualitatively different from the co-tunneling process that induces the conventional Kondo effect by the on-site interaction: in that case the local spin is flipped after the co-tunneling process.

Next, we discuss the non-equilibrium situation ($V \neq 0$) keeping $\Delta \epsilon = 0$. Fig.1d shows the left dot’s LDOS. At $t_C = 0$, a narrow Kondo peak emerges (thin solid line). However, its position is at $\epsilon = \mu_R$, not at $\mu_L$. In other words, the Kondo peak of one dot occurs at the chemical potential of the lead of the other dot! This is a very surprising result indeed. It is also qualitatively different from the Kondo resonance induced by intradot interaction $U_o$ (not the interdot $U$) where the peak is pinned to the chemical potential of the lead attached to the same dot [3]. When $t_C \neq 0$, states $\epsilon_L$ and $\epsilon_R$ hybridize into two “molecular” states $\epsilon^\pm = \frac{\epsilon_L + \epsilon_R}{2} \pm \frac{\Delta E}{2}$ expanding to the entire device at $\Delta \epsilon = 0$ [16], where $\Delta E = \sqrt{\Delta \epsilon^2 + 4t_C^2}$. The corresponding left dot’s LDOS are plotted in Fig.1d. The main features are: (i) The Kondo peak at $\epsilon = \mu_R$ when $t_C = 0$ is now split into three peaks, at $\epsilon = \mu_R$ and $\mu_R \pm \Delta E$ with weights $\frac{1}{2}$ and $\frac{1}{4}$ respectively. (ii) A dip emerges at $\epsilon = \mu_L$ and two more peaks are at $\mu_L \pm \Delta E$. Most importantly, all these (five) peaks and the dip have Kondo characteristics: (1) they are pinned at $\mu_L/R$ and $\mu_L/R \pm \Delta E$; (2) they disappear at high temperature; on the other hand, at lower temperature they become higher/lower for the peak/dip (See Fig.2); (3) they all disappear when $U = 0$. 

6
In fact, the five Kondo peaks and the dip are results of many-body correlation due to the off-site interaction $U$. They originate from the co-tunneling processes between the molecular state $\epsilon^\pm$ and the two leads which are shown in Fig.1a-c. For example, Fig.1a describes the following co-tunneling process. An electron initially occupies $\epsilon^+$ and tunnels to the Fermi level of the right lead. At almost the same moment another electron in the left lead, having energy $\mu_R - \Delta E$, tunnels into the other molecular state $\epsilon^-$. At low temperatures, a coherent superposition of many co-tunneling events of this type gives rise to the Kondo resonance at $\mu_R - \Delta E$ in the LDOS. Exactly the same way, the co-tunneling events shown in Fig.1c induce a Kondo peak at $\mu_R + \Delta E$. Fig.1b shows a different type co-tunneling process. Here an electron occupies level $\epsilon^\pm$ initially. It then tunnels to the Fermi level of the right lead, closely followed by another electron in the left lead with energy $\mu_R$ which tunnels into same level $\epsilon^\pm$. This co-tunneling process does not induce electron exchange between the molecular states $\epsilon^\pm$, therefore it cannot induce a Kondo peak at the DOS of the molecular states. However, molecular states $\epsilon^\pm$ are linear combinations of the original dot states $\epsilon_L$ and $\epsilon_R$, so in this co-tunneling process, electrons may still exchange between $\epsilon_L$ and $\epsilon_R$, which gives rise to a Kondo peak at $\epsilon = \mu_R$ in the left dot’s LDOS and a dip at $\epsilon = \mu_R$ in the right dot’s LDOS.

The peculiar Kondo resonances discussed so far are for equal dot states level positions, i.e. for $\Delta \epsilon = \epsilon_L - \epsilon_R = 0$. They however persist even when $\Delta \epsilon \neq 0$. As $\Delta \epsilon$ increases from zero, the molecular states $\epsilon^\pm$ gradually localize to one of the dots and they asymmetrically couple to the two leads. If $\Delta \epsilon > 0$, $\epsilon^+/\epsilon^-$ are coupled stronger to the left/right lead, and weaker to the other lead (Fig.1a-c). Therefore, the co-tunneling processes of Fig.(1a,1b) will be weakened while that of Fig.1c enhanced. Correspondingly, in the left dot’s LDOS, the Kondo peaks at $\mu_R + \Delta E$ is increased, and others will be reduced (see inset of Fig.2). In the limiting case of $t_C/\Delta \epsilon \to 0$, only the Kondo peak at $\mu_R + \Delta E$ survives in the left dot’s LDOS. It is not difficult to find that approximately the weights and positions for each left dot’s Kondo peak are: 

\[
\frac{1}{2}(1 - \frac{\Delta \epsilon}{\Delta E}) - \frac{|t_C|^2}{\Delta E^2} \text{ for Kondo peak at } \mu_R - \Delta E; \quad \frac{2|t_C|^2}{\Delta E^2} \text{ for Kondo peak at } \mu_R; \quad \frac{1}{2}(1 + \frac{\Delta \epsilon}{\Delta E}) - \frac{|t_C|^2}{\Delta E^2} \text{ for Kondo peak at } \mu_R + \Delta E; \quad \frac{|t_C|^2}{\Delta E^2} \text{ for Kondo peak at } \mu_L - \Delta E;
\]
$-\frac{2|t_C|^2}{\Delta E^2}$ for Kondo dip at $\mu_L$; and $\frac{|t_C|^2}{\Delta E^2}$ for Kondo peak at $\mu_L + \Delta E$.

The current $I$ and differential conductance $dI/dV$ are substantially influenced by the Kondo resonances. The left inset of Fig.3 show $dI/dV$ versus $V$ for several different interdot coupling $t_C$. Since we fixed $\Delta \epsilon = 0$, $dI/dV$ must be symmetric while $I$ must be antisymmetric across zero bias which is what we found. The main characteristics are: (1) A very narrow peak is exhibited at $V = 0$ in $dI/dV$, as a result of the co-tunneling process of Fig.1b. Increasing $t_C$, the height of this peak first increases sharply and then reaches a maximum $\sim 0.6e^2/h$ (the unitary value is $e^2/h$) at $t_C \sim 0.18\Gamma$, followed by a slow decrease. (2) $dI/dV$ abruptly drops at $V = \pm \Delta E$. If $t_C$ is large, $dI/dV$ exhibits another peak at $V = \pm \Delta E$ followed by the drop. Because at $V = \pm \Delta E$, the left lead’s chemical potential $\mu_L$ aligns with the left dot’s Kondo resonance at $\mu_R \pm \Delta E$, while $\mu_R$ aligns with the right dot’s Kondo resonance at $\mu_L \mp \Delta E$. (3) Increasing temperature, the $dI/dV$ features at $V = 0$ and $V = \pm \Delta E$ gradually diminish, and the overall value of $dI/dV$ greatly reduces (see the right inset of Fig.3). At high temperature, e.g. $T = 0.5$, $dI/dV$ becomes very small and very weakly dependent on bias.

When $\Delta \epsilon \neq 0$, $dI/dV$ is no longer perfectly symmetric as shown in Fig.3. With increasing $\Delta \epsilon$, electron tunneling between the left and right dots becomes more difficult and molecular states $\epsilon^{\pm}$ will localize to one of the dots. As a consequence, the features of $dI/dV$ at $V = 0$ and $\pm \Delta E$ are reduced. Specifically, for $\Delta \epsilon > 0$ the abrupt drop of $dI/dV$ at $V = -\Delta E$ will gradually disappear, because the co-tunneling process of Fig.1a becomes weaker. On the other hand, the abrupt drop at $V = \Delta E$ survives although it becomes weaker. Furthermore, as discussed above, $\Delta \epsilon > 0$ enhances the co-tunneling process of Fig.1c, due to this enhancement a new peak in $dI/dV$ arises at $V = \Delta E$ for large $\Delta \epsilon$.

In summary, we investigated Kondo effect in a DQD device with an off-site electron correlation provided by the interdot interaction $U$. A number of co-tunneling processes have been identified between the molecular states and the leads, and electrons may be moved from one dot to the other due to these processes. Rather than flipping the electron spin, the
interdot exchange of electrons together with the off-site electron correlation and coherent superpositions of the co-tunneling processes, induce several Kondo resonances without the electron spin-degrees of freedom. We note that although the intradot interaction $U_o$ should be larger than $U$, the Kondo temperature is not monotonic in these parameters. It is actually quite probable that the Kondo resonance determined by $U$ is more prominent than that induced by $U_o$. Therefore, if spin, intradot and interdot interactions are all present, more complicated Kondo phenomenon should result.

**Acknowledgments:** We gratefully acknowledge financial support from NSERC of Canada and FCAR of Québec.
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10
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FIGURES

FIG. 1. (a)-(c) are schematic diagrams of co-tunneling processes between the two molecular states and the two leads. (d) and its right inset show the left dot’s LDOS vs energy $\epsilon$ at different tunnel coupling $t_C$ with $\epsilon_L = \epsilon_R = -2.5$, $T = 0.005$. In (d), $\mu_L = -\mu_R = 0.2$; in its right inset $\mu_L = -\mu_R = 0$. The left inset of (d) is a schematic diagram for the co-tunneling process at $t_C = 0$.

FIG. 2. Left dot’s LDOS vs $\epsilon$ at different temperature $T$. Inset: data for different $\Delta \epsilon$ and $\epsilon_L/\epsilon_R = -2.5 \pm \Delta \epsilon/2$. $t_C = 0.05$ and other parameters are the same as those of Fig.(1d).

FIG. 3. The conductance $dI/dV$ vs the bias $V$ at different $\Delta \epsilon$. $\epsilon_L = -2$, $\epsilon_R = -2 - \Delta \epsilon$, $T = 0.001$, and $t_C = 0.05$. Left inset: $dI/dV$ vs $V$ at different $t_C$ with $\epsilon_L = \epsilon_R = -2$ and $T = 0.001$. Different curves correspond to $t_C = 0.025$, 0.05, 0.1, and 0.2 from bottom to top. Right inset: $dI/dV$ vs $V$ at different temperature $T$ where $t_C = 0.05$ and $\epsilon_L = \epsilon_R = -2$. 
Fig. 1
Fig. 2
Fig. 3