Orbital Kondo Spectroscopy in a Double Quantum Dot System

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We calculate the nonequilibrium conductance of a system of two capacitively coupled quantum dots, each one connected to its own pair of conducting leads. The system has been used recently to perform pseudospin spectroscopy by controlling independently the voltages of the four leads. The pseudospin is defined by the orbital occupation of one or the other dot. Starting from the SU(4) symmetric point of spin and pseudospin degeneracy in the Kondo regime, for an odd number of electrons in the system, we show how the conductance through each dot varies as the symmetry is reduced to SU(2) by a pseudo-Zeeman splitting, and as bias voltages are applied to any of the dots. We analyze the expected behavior of the system in general, and predict characteristic fingerprint features of the SU(4) → SU(2) crossover that have not been observed so far.

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The Kondo effect is one of the most studied phenomena in strongly correlated condensed matter systems [1] and is still a subject of great interest. Originally observed in systems of magnetic impurities in metals, the Kondo effect has reappeared more recently in the context of semiconductor quantum-dot (QD) systems, with a single “impurity”, in which an unprecedented control of the parameters could be achieved [2–5]. The effect is characterized by the emergence of a many-body singlet ground state formed by the impurity spin and the conduction electrons in the Fermi sea. The binding energy of this singlet is of the order of the characteristic Kondo temperature $T_K$ below which the effects of the “screening” of the impurity spin manifest in different physical properties.

The role of the impurity spin can be replaced by other quantum degree of freedom (pseudospin) that distinguishes degenerate states, such as orbital momentum. A particularly interesting case is when both a two-fold orbital degeneracy and spin degeneracy are present, leading to an SU(4) Kondo effect [6–19]. This exotic Kondo effect has been observed in different systems, such as quantum dots in carbon nanotubes [10,11], silicon nanowires [16], and organic molecules deposited on Au(111) [18].

Recently, a double QD with strong interdot capacitive coupling, and each QD tunnel-coupled to its own pair of leads has been experimentally [14–20] and theoretically [14,21] studied [see Fig. 1 (a)]. The occupation of one QD or the other plays the role of the pseudospin. These occupations, the tunneling matrix elements and the voltages at the four leads can be controlled independently. While the spin degeneracy can be broken by a magnetic field, this also affects the conduction leads. Instead, a pseudo-Zeeman splitting can be applied on the QDs solely, opening the exciting possibility to explore in detail the orbital structure of the SU(4) Kondo state and how it is changed as the pseudo-Zeeman field reduces the symmetry to SU(2).

So far, the theoretical study of this system has been concentrated in equilibrium properties, for which accurate techniques like numerical renormalization group (NRG) and density-matrix renormalization group can be applied. A much richer physics is expected in the nonequilibrium situation, which arises for finite bias voltages between the leads connected to any of the QDs in the experiment, because of the presence of inelastic processes. Unfortunately, the theoretical treatment is much more difficult in this case. For one QD, the experimental study at finite bias voltages [22] allowed the test of universality and scaling relations within different nonequilibrium theories [23,24]. Here we use the Keldysh formalism within the non-crossing approximation (NCA) [25,20], which reproduces well the scaling relations mentioned above [27] and was also successfully used to interpret experimental results on a controlled crossover between SU(4) and SU(2) Kondo states driven by magnetic field in a nanoscale Si transistor [14], and quantum phase transitions involving singlet and triplet states [28].

Here we report calculations of the conductances through both QDs in the general nonequilibrium case. We describe in particular nontrivial changes in the conductance through one QD as a voltage is applied to the other. We also describe how the spectral densities evolve under application of different bias voltages. Fingerprints of the SU(4) → SU(2) crossover are predicted.

Our starting model is the SU(4) Anderson model which mixes a singlet configuration with two degenerate spin doublets $|\sigma\rangle$ ($i = 1$ or 2) corresponding to one additional electron (or hole) in QD $i$, through couplings $\Gamma_1 = \Gamma_2$ to a continuum of extended states. It is described for example in Ref. [17] replacing valley by QD index. The symmetry is reduced to SU(2) by a pseudo-Zeeman splitting $\delta =$
that application of \( V_2 \) has a stronger effect on decreasing \( G_2 \) than \( V_1 \). In fact for \( V_2 = 0 \) and any \( V_1 \), one expects that the spin Kondo effect on QD2 still remains, although weakened, and this is consistent with our results. As in the usual SU(2) Kondo effect, \( G_1(V_1) \) drops to \( G_1(0)/2 \) at a bias voltage such that \( eV_1 \approx T_K^{SU(2)} \), where \( T_K^{SU(2)} \) is the Kondo temperature for \( \delta = 0 \) as discussed below. For our parameters, \( T_K^{SU(2)} \approx 0.02 \) and it increases to near 0.3 if \( E_1 \) is changed from -4 to -2. Since experimentally temperatures \( T \approx 0.1 \) can be reached, and \( E_1 \) can be tuned, a wide range of ratios \( G/\rho \) is accessible.

Non-trivial correlation effects between both QDs are apparent in the fact that \( G_1 \) increases on the lines \( V_1 = \pm V_2 \). This is related to the evolution of the spectral densities \( \rho_i(\omega) \) as both \( V_1 \) are varied. We find that keeping \( V_2 = 0 \) and increasing \( V_1 \) (or conversely) the Kondo peak at \( \omega = 0 \) in \( \rho_1(\omega) \) is weakened and two peaks at \( \omega \approx \pm eV_1/2 \) split from it. In the general case, when both \( V_1 \neq 0 \), four peaks are present in both \( \rho_i(\omega) \) for \( \omega \approx \pm eV_i/2 \). When \( V_1 = \pm V_2 \) these peaks merge in two more intense peaks and therefore an increase in both \( G_i \) is expected.

In Fig. 3 we show how the \( G_i \) change when a finite pseudo-Zeeman splitting \( \delta \) is introduced. It is known that the spectral density of the dot with lower energy \( \rho_1(\omega) \) has still the Kondo peak near \( \omega = 0 \) and an additional peak for \( \omega \approx -\delta \), while \( \rho_2(\omega) \) has only a peak for \( \omega \approx \delta \) (see Fig. 6). As a consequence, only \( G_1 \) has a peak near \( V_1 = V_2 = 0 \), while \( G_2 \) is vanishingly small at that point. The energy scale of the variation of \( G_1 \) with \( V_1 \) is again given by the Kondo temperature \( T_K \), but it is smaller than that of the SU(4) case. We have found that the binding energy of the singlet ground state obtained from a simple variational calculation can be described by the following expression:

\[
\delta = \frac{1}{4}.
\]
in Fig. 3). This can be understood from the onset of co-
tral density is found to be $1$.

The dependence of $\rho_1$ at equilibrium and low temperatures, both $\rho_1(\omega)$ have a Kondo peak slightly above the Fermi energy (which we take as the origin of energies) for $\delta < T_K^{SU(4)}$ \cite{14}, while for $\delta > T_K^{SU(4)}$, as seen in Fig. 4 (a), the Kondo peak in $\rho_1(\omega)$ moves to the Fermi energy and an inelastic peak near $-\delta$ appears (\(\delta = 0.5\) in the figure). The width of the Kondo peak is $\sim T_K(\delta)$. Instead $\rho_2(\omega)$ has only an inelastic peak near energy $\delta$. We find that the width of both inelastic peaks is of the order of $T_K^{SU(4)}$ for small $\delta$ (but $\delta > T_K^{SU(4)}$ in order to ensure that the inelastic peak is split from the Kondo peak) and increases with increasing $\delta$. This behavior is reminiscent of the evolution of the peaks of the ordinary SU(2) Kondo model under an applied magnetic field, which has been studied by Bethe ansatz techniques \cite{31}.

The equilibrium spectral densities can be investigated by orbital spectroscopy controlling the parameters so that the configuration is similar to that used in scanning tunneling spectroscopy (STS). Specifically if $V_{S1} \gg \Gamma_{D1}$, and only the potential at one of the drains $V_{D1}$ is displaced from the Fermi level, then the dots are in equilibrium with the source leads and for $T \ll T_K(\delta)$, $G_i \propto \rho_i(eV_i)$. Our calculations show that a ratio $\Gamma_{S1}/\Gamma_{D1} = 9$ or drain (D1) lead and an electron from S2 or D2 jumps to QD2 is inhibited because of the energy cost $\delta$. However, when $e|V_2|$ reaches $2\delta$, an event of this type becomes possible, in which as a net result an electron flows from S2 to D2 or conversely depending on the sign of $V_2$, and another electron moves from QD1 to QD2 with a possible spin flip. In a second event the electron of QD2 jumps to its lead of less energy and an electron from S1 or D1 jumps to QD1, leaving the QDs in the same charge configuration as initially. This results in an increase of the current flow $I_2$ and thus to a peak in the conductance $G_2$. On average $I_1 = 0$. However, a small $|V_1|$ breaks the symmetry between S1 and D1 in the above events, leading to a large $G_1$ also. A similar reasoning can be followed for non zero $V_1$.

More insight into the structure of the nonequilibrium conductance is obtained from the spectral densities $\rho_i(\omega)$. At equilibrium and low temperatures, both $\rho_i(\omega)$ have a Kondo peak slightly above the Fermi energy (which we take as the origin of energies) for $\delta < T_K^{SU(4)}$ \cite{14}, while for $\delta > T_K^{SU(4)}$, as seen in Fig. 4 (a), the Kondo peak in $\rho_1(\omega)$ moves to the Fermi energy and an inelastic peak near $-\delta$ appears (\(\delta = 0.5\) in the figure). The width of the Kondo peak is $\sim T_K(\delta)$. Instead $\rho_2(\omega)$ has only an inelastic peak near energy $\delta$. We find that the width of both inelastic peaks is of the order of $T_K^{SU(4)}$ for small $\delta$ (but $\delta > T_K^{SU(4)}$ in order to ensure that the inelastic peak is split from the Kondo peak) and increases with increasing $\delta$. This behavior is reminiscent of the evolution of the peaks of the ordinary SU(2) Kondo model under an applied magnetic field, which has been studied by Bethe ansatz techniques \cite{31}.

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is enough to reach this STS regime. This property was used to study the equilibrium spectral density and to compare it with that resulting from an NRG calculation for a case in which both spin and pseudospin Zee-
man terms were present [19]. However, this destroys the Kondo effect and the two-peak structure like that shown in Fig. 4 (a) remains unexplored. In Fig. 5 we show the evolution of the spectral density starting from the SU(4) case and increasing δ for parameters reached experimentally in recent work [19, 20], in particular Γ₁ = 20 µeV, T = 23 mK. While the peaks become sharper at lower temperature, the displacement of the Kondo peak to the Fermi energy from above, and the splitting of the inelastic peak as δ increases can be clearly seen. Due to the limitations of resolution of NRG at finite energies [32] [33] our NCA results are a useful complement at equilibrium [38] and have the advantage that they can be extended to the nonequilibrium situation.

In general and particularly for a symmetric voltage drop the spectral densities change under application of bias voltages $V_i$. Assuming as a first crude approximation that the $\rho_i(\omega)$ are fixed, one expect that $G_1(V_1)$ has a peak at $V_1 = 0$ corresponding to the Kondo peak in $\rho_1(\omega)$, and two peaks at $V_1 = \pm 2\delta/e$ corresponding to the inelastic peak of $\rho_1(\omega)$. This is in fact what happens for $V_2 = 0$ [see Fig. 4 (b)] but not for $V_2 = 0$ [see Fig. 3 (a)]. Similarly one expects only inelastic peaks at $V_2 = \pm 2\delta/e$ for $G_2(V_2)$, as it happens for $V_1 = 0$ but not for $V_1 \neq 0$.

The differences with the expected behavior for rigid bands when both $V_i \neq 0$ are due to changes in the spectral weight with respect to the equilibrium case. To illustrate these changes we consider the nonequilibrium situation represented in Fig. 4 (d) of Ref. [20] in which the coupling to the source leads is larger and the voltages are applied only in one of these sources $S_1$, keeping the other three voltages at zero. Specifically we keep $\Gamma_1 = 1$ but use $\Gamma_{s1}/\Gamma_{p1} = 3$ and $\Gamma_{s2}/\Gamma_{p2} = 12$, as described in the supplementary material of Ref. [20]. We also changed $E_1 = -3$ and $\delta = 1$ to correspond approximately to the experimental parameters. The evolution of $\rho_1(\omega)$ with $V_{S2}$ is shown in Fig. 6 (a). At equilibrium ($V_{S2} = 0$), the spectral density of QD1 has the two peaks mentioned above. The inelastic peak can be understood as a mixture of the ground state for zero hopping with an excited state in which the electron at QD1 is displaced to QD2 and an an electron from S2 is displaced to D1. Both states are connected in second order in the lead-QDs hopping. The excitation energy is $\delta$. As a consequence of this mixture, when an electron is destroyed in QD1, there is a finite probability of leaving an excited state with energy $\delta$. This leads to a peak at $-\delta$ in $\rho_1(\omega)$. When the chemical potential at S2 is increased, the excitation energy decreases and the peak displaces towards the Fermi energy. When this potential reaches $\delta$, the inelastic peak merges with the elastic one and this leads to a peak in $G_1(V_1)$ at $V_1 = 0$, even at temperatures above $T_K(\delta)$ for which the original elastic peak disappears. This agrees with the result presented in Fig. 4 (d) of Ref. [20]. We obtain a qualitative agreement with experiment, but the ratio of intensities is larger in our case. This might be due uncertainties in the ratio $E_1/\Gamma_1$ or to fluctuations in $\delta$ introduced by decoherence effects [33].

A similar reasoning as above can be followed for a symmetric voltage drop and brings an alternative explanation of the increase in intensity along the lines $eV_1 = \pm eV_2 \pm 2\delta$ displayed in Fig. 6.

In Fig. 6 (b) we show how $\rho_2(\omega)$ changes with $V_{S2}$. In contrast to $\rho_1(\omega)$, much of the spectral weight lies above the Fermi energy. Therefore its magnitude is proportional to the amount of the singlet configuration without particles in the ground state, or in other words, to the degree of intermediate valence. We observe that $\rho_2(\omega)$ increases as $V_{S2}$ approaches $\delta$.

In summary, we predict the values of the conductance through any of two capacitively coupled QDs as the voltage through any of them is varied. We believe that our results are important to stimulate further experimental research along the lines of recent pseudospin-resolved transport measurements [19, 20]. In particular, the presence of three peaks in $G_1(V_1)$ for $V_2 = 0$ (or two peaks in an STS configuration) with one of them at $(V_2) = 0$ is characteristic of the SU(4) → SU(2) crossover [34]. This has
not been observed in experiment yet 19, 20. However, giving the large experimental possibilities of tuning the parameters and the particular sensitivity of $T_K(\delta)$ with the pseudo Zeeman splitting $\delta$ we believe that it can be observed in the near future. An experimental study of the dependence of the lowest energy scale $T_K$ with the pseudo-Zeeman splitting $\delta$ would also contribute to our present understanding of the $SU(4) \rightarrow SU(2)$ crossover.

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