Prediction of Ferromagnetic Correlations in Coupled Double-Level Quantum Dots

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Numerical results for transport properties of two coupled double-level quantum dots (QDs) strongly suggest that under appropriate conditions the dots develop a novel ferromagnetic (FM) correlation at quarter filling (one electron per dot). In the strong coupling regime (Coulomb repulsion larger than electron hopping) and with interdot tunneling larger than tunneling to the leads, an $S = 1$ Kondo resonance develops in the density of states, leading to a peak in the conductance. A qualitative "phase diagram," incorporating the new FM phase, is presented. In addition, the necessary conditions for the FM regime are less restrictive than naively believed, leading to its possible experimental observation in real QDs.

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Strongly correlated electronic systems, such as high-T_c cuprates, heavy fermions, and manganites, display a variety of nontrivial collective states, which are difficult to analyze due to the many-body character of the interactions, and the difficulties in experimentally *controlling* the parameters determining these interactions. These problems are severe in materials that spontaneously grow in particular structures and patterns, with several effects (lattice, spin, charge, orbital) in direct competition. Therefore, the observation of a celebrated many-body effect, the Kondo effect, in a single quantum dot (QD) [1] has captured the attention of the strongly correlated community. It is conceivable that the most interesting states that are spontaneously stabilized in some materials—and are very difficult to control—could instead be artificially created in a man-made structure. In this framework, a natural first step is to analyze coupled QDs. In fact, the two-impurity Kondo problem—extensively studied since the 1980s [2]—can now be realized in a real system. Moreover, recent investigations have reported antiferromagnetic (AF) correlations between two single-level coupled QDs, in competition with Kondo correlations [3–5]. As a consequence, it is now clear that two of the most remarkable magnetic states known to exist in spontaneously grown materials—the Kondo and AF states—have already found realizations in the context of QDs. However, the other dominant magnetic state of some materials—the ferromagnetic (FM) state—has comparatively received much less attention [6]. For the dream of artificially replicating collective states using QDs to be fulfilled, a realization of FM states must be achieved. The lack of attention to FM states in QDs should not be surprising in view of the physics of FM materials, such as manganites. Here, the FM state is reached by removing electrons (doping) from an AF state. Under the constraint of having one particle per level (1/2 filling), and only one level active per QD as in most previous investigations, the double-exchange [7] generated FM state *cannot* be realized. To reach a FM state, more levels need to be active, resulting in *less* than one electron per level.

In this Letter, clear evidence is presented for the development of *ferromagnetic* correlations between two *double-level* QDs [8]: at 1/4 filling (one electron per dot), two coupled double-level QDs form a triplet state. Coupling this state to ideal metallic leads produces a Kondo resonance and a peak in the conductance. The results do not appear to be restricted to only a pair of QDs, but they seem valid for larger QD arrays. Basically here *it is reported a realization of the double-exchange mechanism using QDs*. Although the above mentioned effect is stronger if the appropriate intradot interlevel many-body interactions are added to the Hamiltonian, it is important to stress that these interactions are not necessary: considering just an intralevel Coulomb repulsion (Hubbard U) is enough to obtain qualitatively the same results, opening the possibility for the FM regime to be experimentally observable. Figure 1 schematically depicts the system analyzed here and introduces the labeling for the different tunneling parameters t' and t''. To model this system, the impurity Anderson Hamiltonian that describes the two QDs with two levels (denoted α and β) is given by

$$H_{\rm d} = \sum_{i=1,2}\Bigg\{\sum_{\sigma;\lambda=\alpha,\beta} U n_{i\lambda\sigma} n_{i\lambda\bar{\sigma}} + \sum_{\sigma\sigma'}[U' n_{i\alpha\sigma} n_{i\beta\sigma'} - J c^{\dagger}_{i\alpha\sigma} c_{i\alpha\sigma'} c^{\dagger}_{i\beta\sigma'} c_{i\beta\sigma}] + \sum_{\sigma}[V_g n_{i\alpha\sigma} + (V_g + \Delta V) n_{i\beta\sigma}]\Bigg\} + \sum_{\sigma,\lambda=\alpha,\beta} t''[c^{\dagger}_{1\lambda\sigma} c_{2\lambda\sigma} + \text{H.c.}], \quad (1)$$

where the first term represents the usual Coulomb repulsion between electrons in the same level (considered equal for both levels). The second term represents the Coulomb repulsion between electrons in different levels (the U'

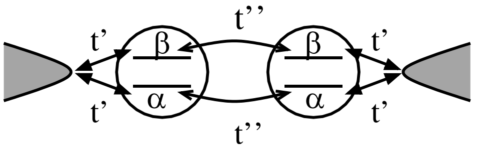

FIG. 1. Schematic representation of the two double-level QDs system and the hopping amplitudes of Eqs. (1) and (3).

notation is borrowed from standard many-orbital studies in atomic physics). The third term represents the Hund coupling ($J > 0$) that favors the alignment of spins, and the fourth term is the energy of the states regulated by the gate voltage V_g. To decrease the number of free parameters, all the calculations presented here assume the following relations: $U' = 2U/3$ and $J = U - U'$. As discussed later, the main result in this Letter does not depend on the specific values taken by U' and J. Note that α and β are separated by ΔV, and by modifying this parameter an interpolation between one- and two-level physics can be obtained. The last term represents the interdot coupling, with matrix element t''. For simplicity, we assume that there is no hopping between levels α and β. The dots are connected to the leads (represented by semi-infinite chains) by a hopping term with amplitude t', while $t = 1$ is the hopping amplitude in the leads (and energy scale). More specifically,

$$H_{\rm leads} = t\sum_{i\sigma}[c^{\dagger}_{li\sigma}c_{li+1\sigma} + c^{\dagger}_{ri\sigma}c_{ri+1\sigma} + {\rm H.c.}], \qquad (2)$$

$$H_{\rm int} = t' \sum_{\sigma,\lambda=\alpha,\beta}[c^{\dagger}_{1\lambda\sigma}c_{l0\sigma} + c^{\dagger}_{2\lambda\sigma}c_{r0\sigma} + {\rm H.c.}], \qquad (3)$$

where $c^{\dagger}_{li\sigma}$ ($c^{\dagger}_{ri\sigma}$) creates electrons at site i with spin σ in the left (right) contact. Site "0" is the first site at the left of the left dot and at the right of the right dot, for each half chain. The total Hamiltonian is $H_{\rm T} = H_{\rm d} + H_{\rm leads} + H_{\rm int}$. Note that for $V_g = -U/2 - U' + J/2 - \Delta V/2$, the Hamiltonian is particle-hole symmetric. Using the Keldysh formalism [9], the conductance through this system can be written as [10] $\sigma = \frac{e^2}{h}|t^2 G_{\rm lr}(E_{\rm F})|^2[\rho(E_{\rm F})]^2$. In practice, a cluster containing the interacting dots and a few sites of the leads is solved exactly, the Green's functions are calculated, and the leads are incorporated through a Dyson Equation embedding procedure (details of the embedding have been provided elsewhere [5,11]). In Figs. 2–4, the cluster used involved the two QDs plus one lead site at left and right.

In Fig. 2(a), results for conductance (solid line) at strong interdot tunneling ($t''/t' \gg 1$) are shown. The main feature displayed is the peak at $V_g \approx -1.0$ (at and near 1/4 filling). The occupancy for this value of V_g is approximately one electron per dot (dashed line $\times 4$) and the total spin $S_{\rm T}$ of the four levels [12] (dotted line) is ≈ 1.0. The smooth charging of the levels as the gate potential decreases (in the peak region) indicates a possible Kondo regime. This is confirmed in Fig. 2(b), where the density of states (DOS) close to the Fermi level is displayed as the gate potential

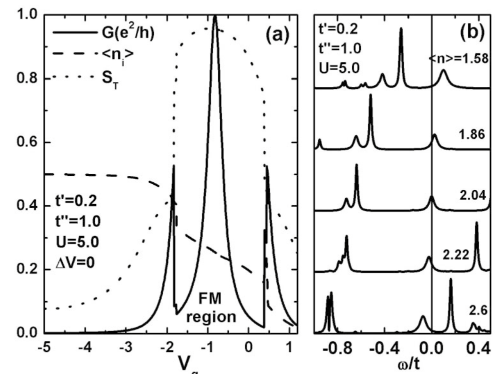

FIG. 2. Results for the strong interdot tunneling regime ($t''/t' \gg 1$) showing the presence of a *ferromagnetic* state at 1/4 filling and concomitant Kondo effect: (a) Conductance (solid line), charge occupancy per level and per spin (dashed line), and total spin $S_{\rm T}$ in the four levels of the dots (dotted line) vs gate potential V_g for two coupled double-level QDs. The values for the parameters are indicated. Note that the maximum in the conductance corresponds to a spin 1 Kondo peak that occurs when there is exactly one electron per dot and the total spin $S_{\rm T} \approx 1.0$. The other two peaks occur when there are one and three electrons in the two-dots system, corresponding to $S_{\rm T} = 1/2$ Kondo resonances. (b) DOS indicating the presence of a Kondo resonance associated with the $S_{\rm T} = 1$ Kondo peak in (a). The numbers on the right side indicate the total number of electrons inside the two quantum dots. V_g takes the values 0.0, -0.5, -0.83, -1.12, and -1.5 from the top to the bottom curve. The Fermi level is located at $\omega/t = 0.0$.

varies from 0.0 to -1.5 (top to bottom). Through this variation of V_g the two dots are charged with one additional electron (the total mean charge varies from $\approx$1.6 to 2.6). One can clearly see a Kondo resonance pinned to the Fermi level. For lower values of the gate potential (in the region at and near 1/2 filling, with two electrons per dot) the conductance is drastically reduced and the total spin $S_{\rm T}$ inside the dots reaches its minimum value, indicating the formation of a global singlet state. Calculations of the total spin in each dot indicate that this singlet state is formed by the AF coupling of two spins $S = 1$. A description of how this picture changes as the t'' decreases is shown in Fig. 3(a), where results are shown for five different values of t''. The conductance at the particle-hole symmetric point, $V_g = -5.0$, varies from zero, for $t'' = 1.0$, to 1.0 (in units of e^2/h), for $t'' = 0.08$. Figure 3(b) shows how the Kondo correlation (between the total spin in the dots and a conduction electron in the first site of one of the leads) evolves from a negligible value for $t''/t' \gg 1$ to a large value (≈ -0.65) for $t''/t' < 1$. The inset of Fig. 3(a) displays the change of the total spin (from $S_{\rm T} \approx 0.0$ to $\approx 3/4$) as t'' decreases. The two main peaks in the conductance discussed up to now were the $S_{\rm T} = 1$ Kondo peak at 1/4 filling (relevant in the strong interdot tunneling regime) and the peak at 1/2 filling (relevant in the weak interdot tunneling regime). It is interesting to discuss how these peaks evolve as ΔV increases. Figure 4(a) shows results

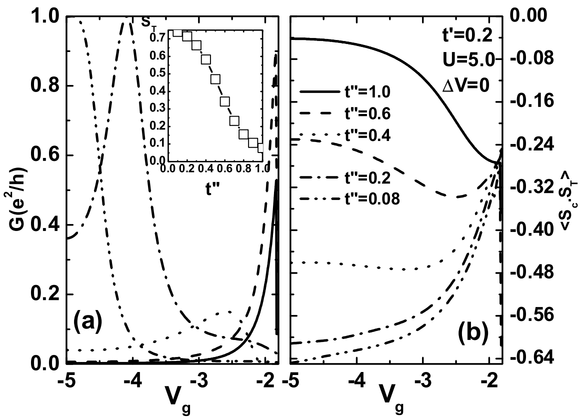

FIG. 3. (a) Conductance G vs gate potential V_g for different values of interdot tunneling [$t'' = 1.0$, 0.6, 0.4, 0.2, and 0.08, see convention in (b)]. V_g varies across the region where the fourth electron is charged into the double-dot system. Note that the conductance at $V_g = -5.0$ gradually increases from zero (at strong tunneling, $t'' = 1.0$) to the maximum value (at weak tunneling, $t'' = 0.08$). This variation indicates a transition from two $S = 1$ spins (in each dot) forming a global singlet to two uncorrelated $S = 1$ spins, each forming a Kondo resonance with the lead conduction electrons. The inset shows the variation with t'' of the total spin inside the dots. (b) Variation with t'' of the spin-spin correlation between the total spin in the double-dot and a conduction electron located in the first site of the leads.

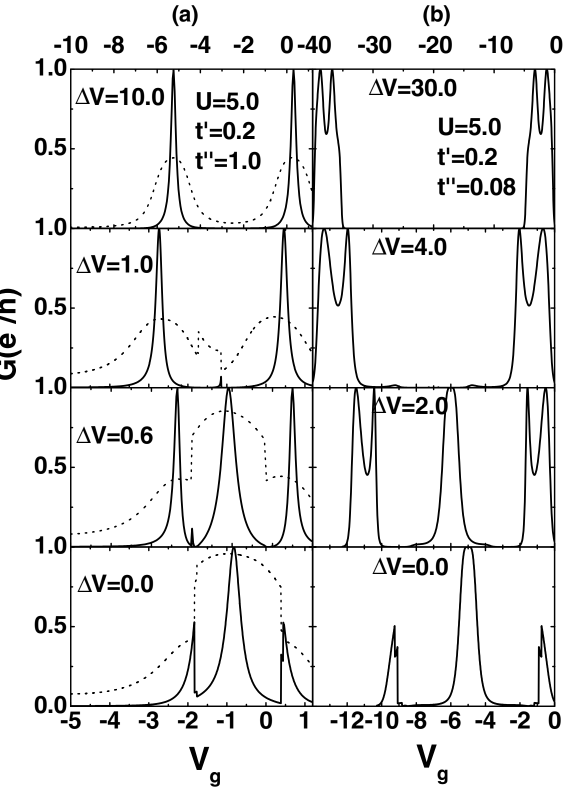

FIG. 4. Variation of the conductance with level separation ΔV. The level separation is indicated for each curve. (a) $t'' = 1.0$: The gate potential decreases down to the particle-hole symmetric value, to highlight how the $S_T = 1$ Kondo peak varies with ΔV. Note that all graphs have the same horizontal axis scale, except for the upper one (scale indicated on top). (b) $t'' = 0.08$: The gate potential varies down to the lowest value (total charging of the dots), to highlight the variation of the central peak (at the particle-hole symmetric point). Here also the upper graph has a different horizontal scale. A discussion of how the data interpolate between double- and single-level dots is given in the text.

for $t''/t' \gg 1$ ($t'' = 1.0$, $U = 5.0$ and $t' = 0.2$). The solid line displays the conductance and the dotted line displays the total spin S_T. Level separation ΔV increases from bottom to top (values for each graph are displayed in the left side). From $\Delta V = 0.0$ up to $\Delta V \approx 0.6$ the width of the conductance peak slowly decreases, as also does the maximum value of S_T. Above $\Delta V \approx 0.7$ (not shown) the narrowing of the peak accelerates (as does the decrease of S_T), until the peak has all but vanished for $\Delta V = 1.0$. For higher values of ΔV (top graph, $\Delta V = 10.0$), the conductance shows the typical Coulomb blockade profile previously discussed [5] for coupled single-level dots when $t''/t' \gg 1$.

Figure 4(b) shows the corresponding results for $t''/t' < 1$ ($t'' = 0.08$, $U = 5.0$, and $t' = 0.2$). Note that the central peak does not change appreciably from $\Delta V = 0.0$ to $\Delta V = 2.0$. In fact, changes start only above $\Delta V = 3.0$, when the central peak splits in two ($\Delta V \gtrsim 3.2$, not shown). For $\Delta V > 3.4$ the two peaks start moving farther apart from each other and become very narrow. Finally, for $\Delta V = 4.0$ the central peaks have disappeared, and the remaining structures are already similar to the single-level result for $t''/t' < 1$. The top graph ($\Delta V = 30.0$) is basically the result reported for single-level QDs at weak interdot tunneling [5] (if one discards the slight shoulders in the internal peaks).

Based on the results displayed in Fig. 4(a), a qualitative phase diagram for the strong interdot tunneling regime can be sketched. In Fig. 5, the electron occupancy is in the horizontal axis (controlled by V_g) and ΔV is in the vertical axis. The left side (indicating $1/2$ filling) is dominated by antiferromagnetism for all values of $\Delta V \lesssim U$. The singlet formed by the four levels is made of two spins $S \approx 1.0$. For $\Delta V > U$ one recovers the single-level picture. The right side of the phase diagram, which describes the evolution of the central peak in Fig. 4(a), is more interesting. For $\Delta V < t''$ one has the novel FM region, where an $S_T = 1$ Kondo effect is present. For $\Delta V > t''$ an AF region is found, with no Kondo effect.

A finite-size scaling analysis was done (results not shown) to verify how our numerical results converge with cluster size [13]. The authors found very little change in the results with increasing cluster size, giving us confidence that all the qualitative results here discussed are not caused by finite-size effects. It is also important to stress that the calculations presented in Fig. 2(a) were reproduced for $U' = J = 0.0$ [with the values of all other parameters kept the same as before ($U = 5.0$, $t'' = 1.0$, and $t' = 0.2$)] and the results obtained barely changed. This indicates that the FM correlation and the $S_T = 1$ Kondo should be experi-

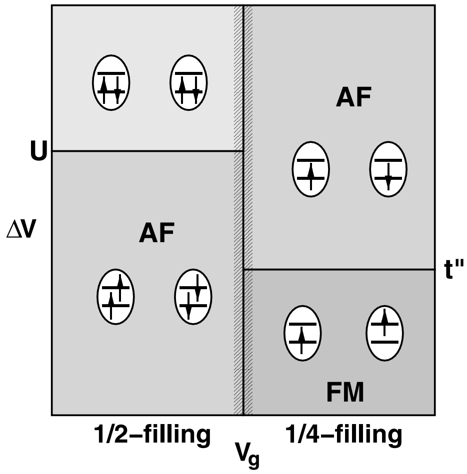

FIG. 5. Qualitative phase diagram for the strong interdot tunneling regime ($t''/t' \gg 1$). Horizontal axis indicates the level occupancy (controlled by V_g) and vertical axis indicates the level separation (controlled by ΔV). In the 1/2-filling region, displayed in the left side, one goes from an AF coupling between two $S = 1$ spins (for $\Delta V < U$) to a situation where the two lower levels are completely occupied (for $\Delta V > U$). In both cases there is no Kondo effect. On the other hand, in the 1/4-filling region (right side), for $\Delta V < t''$ one has the novel $S_T = 1$ FM Kondo region, which gives way to an AF region (with no Kondo effect) once $\Delta V > t''$. The regime with three electrons in the two dots is very narrow as a function of V_g and it is not shown.

mentally observable, since the only requirement is to have two double-level QDs with strong interdot tunneling [14].

Figure 6 qualitatively summarizes the main result presented in this Letter: (a) For double-level coupled quantum dots in the strong interdot tunneling regime at 1/4 filling, FM correlations will develop and conductance through a Kondo channel is allowed. (b) On the other hand, single-level coupled QDs will develop AF correlations in the strong interdot tunneling regime and conductance is suppressed. The results discussed in this Letter complete the analogy between QD states and magnetic phenomena in bulk materials. Previous investigations had shown that Kondo and AF states were possible in QDs. Now, at least theoretically, a regime with ferromagnetism has also been found, if more than one level per dot is active. Certainly, it would be important to confirm experimentally this prediction. Our calculations emphasizing multilevel dots present

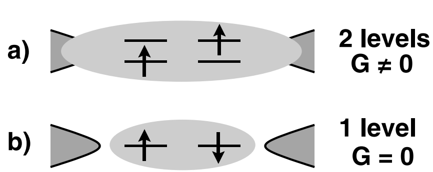

FIG. 6. Schematic representation of the main result in this Letter.

analogies with multiorbital materials such as manganites, nickelates, cobaltites, and ruthenates. These compounds have a plethora of phases, all of which could find realizations in QDs systems as well.

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[10] $G_{\mathrm{lr}}(E_F)$ is the Green's function that moves an electron from the left to the right lead, E_F is the Fermi energy, and $\rho(E_F)$ is the density of states of the leads: $\rho(\omega) = \frac{-1}{\pi}\mathrm{Im}\{\frac{(\omega+i\eta)-\sqrt{(\omega+i\eta)^2-4t^2}}{2t^2}\}$.

[11] This method was originally proposed by E. V. Anda and G. Chiappe. See, V. Ferrari *et al.* Phys. Rev. Lett. **82**, 5088 (1999); and M. A. Davidovich *et al.*, Phys. Rev. B **65**, 233310 (2002).

[12] Note that the total spin in the four levels in the two quantum dots (denoted as S_T and obtained through $\langle \vec{S}_T{}^2 \rangle_{\mathrm{cluster}} = S_T(S_T+1)$, where $\vec{S}_T = \sum_{i,\lambda} \vec{S}_{i,\lambda}$, where i labels the dots and λ labels the levels) is not a good quantum number (since the dots are not isolated). However, S_T gives a good indication on the nature of the Kondo effect ($S = 1/2$ or $S = 1$).

[13] For a finite-size scaling analysis of similar systems, see C. A. Büsser *et al.*, [Phys. Rev. B. (to be published)].

[14] One can understand why an *effective* FM J_{eff} is generated (even in the absence of the Hund J and the U' terms) by using the following argument: At 1/4 filling (two electrons in the four levels), the hopping from one lead to the adjacent dot (and vice versa) is maximized when the spins of the electrons in the levels α and β in the same QD are parallel to each other. A strong interdot tunneling then generates an effective FM coupling between the QDs. Therefore, the only restriction for the FM coupling seems to be that the leads should have just one conduction channel.