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) 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 80’s - 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. 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. 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 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: 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 intra-dot inter-level interactions are added to the Hamiltonian, it is important to stress that these interactions are not necessary: considering just an intra-level 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 labelling 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 $\alpha$ and $\beta$) is given by

$$H_d = \sum_{i=1,2} \left\{ \sum_{\sigma,\lambda=\alpha,\beta} U n_{i\lambda\sigma} n_{i\lambda\bar{\sigma}} + \sum_{\sigma,\sigma'} |U'| n_{i\alpha\sigma} n_{i\beta\sigma'} - J' c_{i\alpha\sigma}^\dagger c_{i\beta\sigma'}^\dagger c_{i\beta\sigma'} c_{i\alpha\sigma} + \sum_{\sigma} [V g_{i\alpha\sigma} + (V_g + \Delta V) n_{i\beta\sigma}] + \sum_{\sigma,\lambda=\alpha,\beta} t'' [c_{i\lambda\sigma}^\dagger c_{2\lambda\sigma} + h.c.] \right\},$$

(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 $U'$ 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 $\alpha$ and $\beta$ are separated by $\Delta V$, and by modifying this parameter an interpolation between one- and two-level physics can be obtained. The last term represents the inter-dot coupling, with matrix element $t''$. For simplicity, we assume that there is no hopping between levels $\alpha$ and $\beta$. 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,

$$H_{\text{leads}} = t \sum_{i,\sigma} \left[ c_{i\alpha}^\dagger c_{i+1\sigma} + c_{i\beta}^\dagger c_{i+1\sigma} + \text{h.c.} \right],$$

(2)

$$H_{\text{int}} = t' \sum_{\sigma,\lambda,\alpha,\beta} \left[ c_{1\lambda\sigma}^\dagger c_{0\sigma} + c_{2\lambda\sigma}^\dagger c_{0\sigma} + \text{h.c.} \right].$$

(3)

where $c_{i\alpha}^\dagger$ ($c_{i\beta}^\dagger$) creates electrons at site $i$ with spin $\sigma$ 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 $H_T = H_d + H_{\text{leads}} + H_{\text{int}}$. Note that for $V_g = -U/2 - U' + J/2 - \Delta V/2$, the Hamiltonian is particle-hole symmetric. Using the Keldysh formalism, the conductance through this system can be written as

$$\sigma = \frac{2}{\pi} \int [T_G(E_F)]^2 |\rho(E_F)|^2 dE_F.$$

In practice, a cluster containing the interacting dots and a few sites of the leads is solved exactly, the Green functions are calculated, and the leads are incorporated through a Dyson Equation embedding procedure (details of the embedding have been provided elsewhere). In Figs. 2-4, the cluster used involved the two QDs plus one lead site at left and right.

In Fig. 2a, results for conductance (solid line) at strong inter-dot 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 1 electron per dot (dashed line $\times 4$) and the total spin $S_T$ of the four levels (dotted line) is $\approx 1.0$. 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. 2b, where the density of states (DOS) close to the Fermi level is displayed as the gate potential 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 2 electrons per dot) the conductance is drastically reduced and the total spin $S_T$ 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. 3a, 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 $t'' = 0.08$. Fig. 3b shows how the Kondo correlation (between the total spin in the dots and a conductance function in the first site of one of the leads) evolves from a negligible value for $t''/t' \gg 1$ to a large value ($\approx -0.65$) for $t''/t' < 1$. The inset of Fig. 3a displays the change of the total spin (from $S_T \approx 0.0$ to $\approx 3/4$) as $t''$ decreases. The two main peaks in the conductance discussed up to now were the $S_T = 1$ Kondo peak at 1/4-filling (relevant in the strong inter-dot tunneling regime) and the peak at 1/2-filling (relevant in the weak inter-dot tunneling regime). It is interesting to discuss how these peaks evolve as $\Delta V$ increases. Fig. 4a shows results 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 $\Delta V$ increases from
FIG. 3: (a) Conductance $G$ vs. gate potential $V_g$ for different values of inter-dot 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.

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 $\Delta V$ (top graph, $\Delta V = 10.0$), the conductance shows the typical Coulomb blockade profile previously discussed for coupled single-level dots when $t''/t' \gg 1$.

Figure 4b 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 $t''/t' < 1$. The top graph ($\Delta V = 30.0$) is basically the result reported for single-level QDs at weak interdot tunneling (if one discards the slight shoulders in the internal peaks).

Based on the results displayed on Fig. 4a, a qualitative phase diagram for the strong inter-dot tunneling regime can be sketched. In Fig. 5, the electron occupancy is in the horizontal axis (controlled by $V_g$) and $\Delta V$ 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.

A finite size scaling analysis was done (results not shown) to verify how our numerical results converge with cluster size. The authors found out 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. 2a 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 effect should be experimentally observable, since the only requirement is to have two double-level QDs with strong inter-dot tunneling.

Figure 6 qualitatively summarizes the main result pre-
sent in this Letter: (a) For double-level coupled quantum dots in the strong inter-dot 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 inter-dot tunneling regime and conductance is suppressed. The results discussed in this paper 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 analogies with multi-orbital 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.

FIG. 5: Qualitative phase diagram for the strong inter-dot tunneling regime (t''/t' ≫ 1). Horizontal axis indicates the level occupancy (controlled by Vg) 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 ΔV < Vg) to a situation where the two lower levels are completely occupied (for ΔV > Vg). In both cases there is no Kondo effect. On the other hand, in the 1/4-filling region (right side), for ΔV < t'' one has the novel St = AF Kondo regime, which gives way to an AF region (with no Kondo effect) once ΔV > t'''. The regime with 3 electrons in the two dots is very narrow as a function of Vg and it is not shown.

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

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