Nonequilibrium Transport through Double Quantum Dots: Kondo Effect versus Antiferromagnetic Coupling

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We theoretically study the nonequilibrium transport properties of double quantum dots, in both series and parallel configurations. Our results lead to novel experimental predictions that unambiguously signal the transition from a Kondo state to an antiferromagnetic spin-singlet state, directly reflecting the physics of the two-impurity Kondo problem. We prove that the nonlinear conductance through parallel dots directly measures the exchange constant J between the spins of the dots. In serial dots, the nonlinear conductance provides an upper bound on J.

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Introduction.—It is now well established that quantum dots (QD's) [1] are artificial realizations of the Anderson model [2] and, then, behave as Kondo impurities at very low temperatures [3]. Recent and ongoing research studying different aspects [4] of the Kondo effect in QD's has renewed the interest in this important problem of condensed matter physics. In view of these and recent experiments studying quantum coherence in double quantum dots (DQD's) [5], it is thus a timely question to ask what happens when two QD's in the Kondo regime are coupled [6].

The interest in studying DQD's is twofold: (i) The rapid developments in the fields of spintronics and quantum information processing (QIP) have made it desirable to understand the behavior of spins which are confined to nanostructures. In a serial DQD with two electrons, the interdot coupling t_C and the intradot on-site Coulomb interaction U generate many-body states. For an isolated DQD, the ground state is a spin singlet. This ground state is an entangled state with possible applications in QIP [7]. The excitation energy to the closest triplet state is given by the antiferromagnetic (AF) exchange constant $J = t_C/2[\sqrt{(U/t_C)^2 + 16} - (U/t_C)] \sim 4t_C^2/U$. (ii) DQD's coupled to external leads are fully tunable, allowing one to investigate in a well controlled manner different regimes of interest. If only spin fluctuations are important, this system can be regarded as an artificial version of the two-impurity Kondo problem [8]. Early studies of this problem by Jones *et al.* [9] demonstrated that the competition between the Kondo effect and antiferromagnetism appears as a quantum critical phenomenon when $J \simeq 2.2T_K^0$ (T_K^0 is the Kondo temperature of each single impurity) when there is an even-odd parity symmetry. When this symmetry is broken, the critical transition is replaced by a crossover [10–12]. This competition determines the behavior of different strongly correlated electron systems like, e.g., heavy-fermion systems [2]. Importantly, DQD's, unlike bulk metals with magnetic impurities, allow study of this problem at the level of *two fully tunable single magnetic impurities*. The question of whether and how this competition manifests in the nonequilibrium transport properties through DQD's is nontrivial. It will be addressed in this Letter.

Transport through DQD's in the Kondo regime has already received some theoretical attention [8,13–15], but a study of the nonequilibrium transport properties when there is an interplay between antiferromagnetism and Kondo effect has been lacking. We study two different experimental realizations of a DQD system: serial [Fig. 1(a)] and parallel [Fig. 1(b)] configurations. Our main findings can be summarized in Figs. 2(b) (serial DQD) and 3 (parallel DQD) where we prove that the nonlinear conductance $\mathcal{G} \equiv dI/dV_{dc}$ directly reflects the physics of the Kondo state (KS) $\rightarrow$ AF transition: in both configurations, the key feature of this transition is that the zero-bias anomaly in $\mathcal{G}$ splits upon changing from $J/T_K < (J/T_K)_c \simeq 2.5$ to $J/T_K > (J/T_K)_c \simeq 2.5$. This can be accomplished by reducing the Kondo temperature of the DQD (T_K). Importantly, the KS $\rightarrow$ AF transition manifests differently for each case: in serial cases, the splitting (Δ) provides an upper bound on J. For parallel cases, Δ is

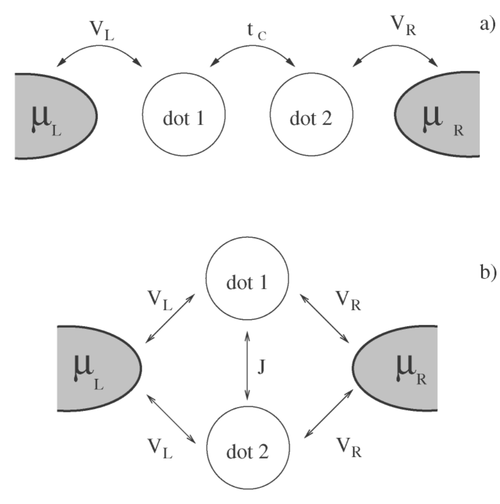

FIG. 1. (a) DQD's in series. (b) DQD's in parallel.

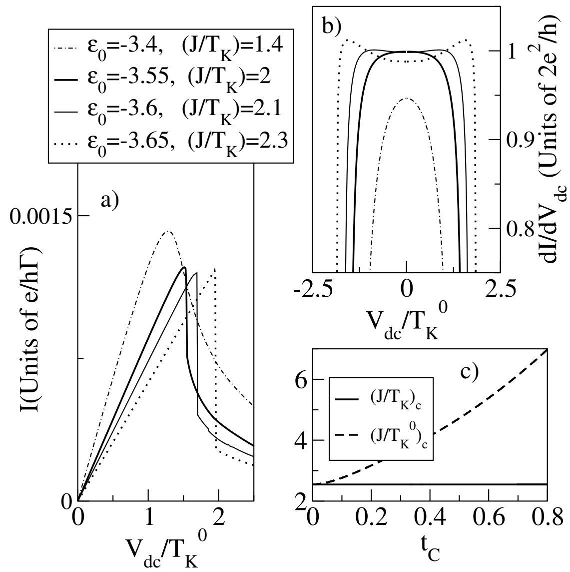

FIG. 2. Serial DQDs with $t_C = 0.5$ and $J = 25 \times 10^{-4}$. (a) I-V characteristics for different ϵ_0's corresponding to $T_K^0 = 1.4 \times 10^{-3}, 1.2 \times 10^{-3}, 1.0 \times 10^{-3}, 8.6 \times 10^{-4}, 7.4 \times 10^{-4}, 6.3 \times 10^{-4}$. (b) Nonlinear differential conductance $\mathcal{G} \equiv dI/dV_{dc}$ for the same values. At $(J/T_K) \lesssim (J/T_K)_c \simeq 2.5$ the ZBA splits. (c) Dependence of $(J/T_K^0)_c$ on t_C (dotted line). The line $(J/T_K)_c = 2.5$ (solid) is also shown.

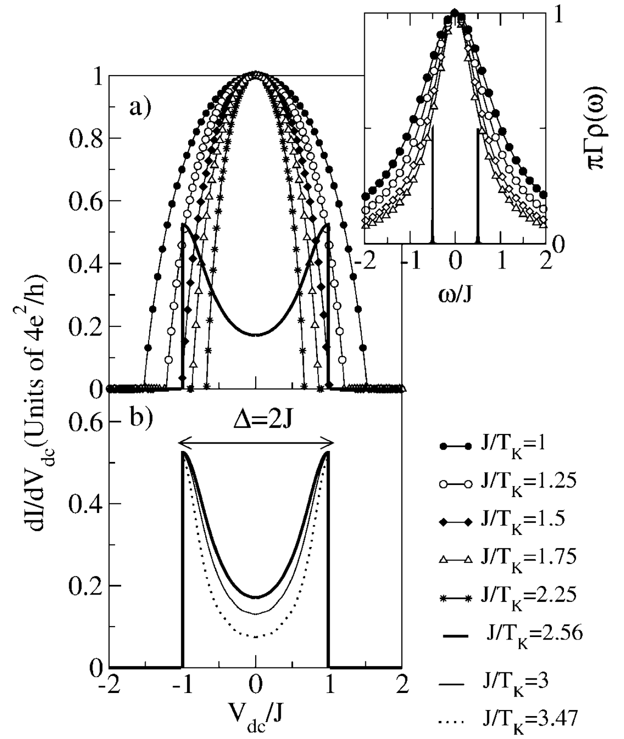

FIG. 3. Parallel DQD's with $J = 25 \times 10^{-4}$. (a) dI/dV_{dc} for different $(J/T_K) < (J/T_K)_c \simeq 2.5$. The dI/dV_{dc} curves show a ZBA with $\mathcal{G}_0 = 4e^2/h$. The width of the ZBA decreases as T_K decreases, namely, as the ratio (J/T_K) increases. At $(J/T_K) \approx (J/T_K)_c$ the ZBA splits. (b) dI/dV_{dc} for different $(J/T_K) > (J/T_K)_c \simeq 2.5$. The splitting is always $\Delta = 2J$ allowing one to measure J experimentally. Inset: Abrupt change of the DOS at the transition.

always $2J \sim 5(T_K)_c$ (Fig. 4). This would allow one *to extract J experimentally* from $\mathcal{G}$ (Fig. 3).

Serial DQD's (Model I).—DQD's in series are modeled by using a $(N = 2)$ fold-degenerate two-impurity Anderson model with an extra interdot tunneling term. Each QD is attached to a different electron reservoir with chemical potentials μ_L and μ_R, respectively. We assume that U is sufficiently large so that (i) double occupancy on each QD is forbidden, but (ii) there is an effective AF spin coupling due to virtual double occupancy, namely, $J\vec{\mathbf{S}}_1 \cdot \vec{\mathbf{S}}_2$, where $\vec{\mathbf{S}}_{1,2}$ are the usual SU(N) spin operators and $J = 4t_C^2/U > 0$ [8,13]. The total Hamiltonian is then $\mathcal{H}_I = \mathcal{H}_I^{\rm SB} + \mathcal{H}_{\rm AF}$, where condition (i) allows us to use an auxiliary slave-boson (SB) representation [16]:

$$\mathcal{H}_I^{\rm SB} = \sum_{k_\alpha \in \{L,R\}\sigma} \epsilon_{k_\alpha} c^\dagger_{k_\alpha \sigma} c_{k_\alpha \sigma} + \sum_{i \in \{1,2\},\sigma} \epsilon_{i\sigma} f^\dagger_{i\sigma} f_{i\sigma} + \frac{t_C}{N} \sum_\sigma (f^\dagger_{1\sigma} b_1 b_2^\dagger f_{2\sigma} + f^\dagger_{2\sigma} b_2 b_1^\dagger f_{1\sigma})$$
$$+ \frac{V_L}{\sqrt{N}} \sum_{k_L \sigma} (c^\dagger_{k_L\sigma} b_1^\dagger f_{1\sigma} + f^\dagger_{1\sigma} b_1 c_{k_L\sigma}) + (L \to R, 1 \to 2) + \sum_{i \in \{1,2\}} \lambda_i \Big(\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} + b_i^\dagger b_i - 1 \Big). \qquad (1)$$

Condition (ii) gives $\mathcal{H}_{\rm AF} = \frac{J}{N} \sum_{\sigma,\sigma'} f^\dagger_{1\sigma} f_{1\sigma'} f^\dagger_{2\sigma'} f_{2\sigma}$. In Eq. (1), $c^\dagger_{k_\alpha,\sigma}(c_{k_\alpha,\sigma})$ are the creation (annihilation) operators for electrons in the reservoir α. In the SB representation, the annihilation operator $d_{i\sigma}$ $(i \in 1, 2)$ for electrons in each dot is decomposed into the SB operator $b_i^\dagger$ which creates an empty state and a pseudofermion operator $f_{i\sigma}$ which annihilates the singly occupied state with spin σ: $d_{i\sigma} \to b_i^\dagger f_{i\sigma}$ $(d^\dagger_{i\sigma} \to f^\dagger_{i\sigma} b_i)$. This replacement is exact provided that $\hat{Q}_i = \sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} + b_i^\dagger b_i = 1$ is fulfilled in each dot. The two constraints are enforced in (1) by two Lagrange multipliers λ_i.

Parallel DQD's (Model II).—Parallel dots can be fabricated to have both electrostatic and interdot tunnel couplings [17]. We choose to treat the cleanest case where the interdot tunneling is negligible [18]. Thus, the exchange J comes only from a strong electrostatic interdot coupling. This way, parallel DQD's can be described with the model Hamiltonian $\mathcal{H}_{\rm II} = \mathcal{H}_{\rm II}^{\rm SB} + \mathcal{H}_{\rm AF}$, where $\mathcal{H}_{\rm II}^{\rm SB}$ can be obtained from $\mathcal{H}_{\rm I}^{\rm SB}$ by coupling each dot to *two leads* and by eliminating the interdot tunneling term.

Our analysis of $\mathcal{H}_{\rm I}$ and $\mathcal{H}_{\rm II}$ is based on two mean field (MF) approximations. First, we use the so-called slave-boson mean field theory (SBMFT) [8,10,13,14,16], which consists of the replacement $b_i(t)/\sqrt{N} \to \langle b_i \rangle/\sqrt{N} = \tilde{b}_i$. The neglect of fluctuations around $\langle b_i(t) \rangle$ is exact in the limit $N \to \infty$, and corresponds to $\mathcal{O}(1)$ in a $1/N$ expansion. At $T = 0$, it correctly describes spin fluctuations (Kondo regime). Second, the AF interaction is decoupled by introducing a valence bond operator $\chi_{12} \equiv -\frac{J}{N}\sum_\sigma f^\dagger_{1\sigma} f_{2\sigma}$. At large N we may ignore its fluctuations ($\chi_{12} \to \bar{\chi}_{12} = \langle \chi_{12} \rangle$) such that $\mathcal{H}_{\rm AF} \to \sum_\sigma f^\dagger_{2\sigma} f_{1\sigma} \bar{\chi} + \bar{\chi}^* \sum_\sigma f^\dagger_{1\sigma} f_{2\sigma} + \frac{N}{J} ||\bar{\chi}||^2$, where $\bar{\chi} \equiv \bar{\chi}_{12} = \bar{\chi}^*_{21}$ [19]. By making these two MF approximations, we render the Hamiltonian quadratic in the fermion operators. The problem, though quadratic, is far from being trivial for $\tilde{b}_1$, $\tilde{b}_2$, λ_1, λ_2, $\bar{\chi}$, and $\bar{\chi}^*$ and depend self-consistently on fermionic nonequilibrium Green's functions (NGF's). Following Ref. [14], we obtain nonlinear self-consistent equations relating the MF parameters with the NGF's. For model I they read

$$\tilde{V}_{L(R)} \sum_{k_{L(R)},\sigma} \langle c^\dagger_{k_{L(R)}\sigma}(t) f_{1(2)\sigma}(t) \rangle + \tilde{t}_C \sum_\sigma \langle f^\dagger_{2(1)\sigma}(t) f_{1(2)\sigma}(t) \rangle + \lambda_{1(2)} \tilde{b}^2_{1(2)} = 0, \tag{2a}$$

$$\tilde{b}^2_{1(2)} + \frac{1}{N} \sum_\sigma \langle f^\dagger_{1(2)\sigma}(t) f_{1(2)\sigma}(t) \rangle = \frac{1}{N}, \tag{2b}$$

$$\bar{\chi} = -\frac{J}{N} \sum_\sigma \langle f^\dagger_{1\sigma} f_{2\sigma} \rangle. \tag{2c}$$

where $\tilde{V}_{L(R)} = V_{L(R)} \tilde{b}^2_{1(2)}$ and $\tilde{t}_C = t_C \tilde{b}_1 \tilde{b}_2$: namely, the original couplings are strongly renormalized by Kondo correlations (i.e., by the MF bosons). The NGF's ($i, j \in 1, 2$), $G^<_{i,j\sigma}(t - t') = -i\langle f^\dagger_{i\sigma}(t') f_{j\sigma}(t) \rangle$, $G^<_{1(2),k_{L(R)}\sigma}(t - t') = -i\langle c^\dagger_{k_{L(R)}\sigma}(t') f_{1(2)\sigma}(t) \rangle$, are obtained by applying the analytic continuation rules of Ref. [20] to the equation of motion of the time-ordered GF's along a complex contour (Keldysh, Kadanoff-Baym, etc.). This allows us to close the set of Eqs. (2). In equilibrium they reduce to Eq. (4) in Ref. [8]. Model II is solved similarly. The current is obtained from the NGF's [21].

Results.—To simplify, we consider henceforth that $V_L = V_R = V_0$ and $\epsilon_{1\sigma} = \epsilon_{2\sigma} = \epsilon_0$. All energies are given in units of $\Gamma(\epsilon) = \pi \sum_{k_\alpha} |V_0|^2 \delta(\epsilon - \epsilon_{k_\alpha}) \equiv \Gamma(\epsilon_F)$ for $-D \leq \epsilon \leq D$ (D is the half-bandwidth and serves as a high energy cutoff). Previous studies [8,13] of the linear transport properties through DQD's in series have already yielded information about the KS $\to$ AF transition. By comparing their ground state energies [13] $\epsilon^{\rm GS}_K - \epsilon^{\rm GS}_{\rm AF} = J/4 - 2T_K/\pi$, the transition can be estimated to appear at $(J/T_K)_c = 8/\pi \simeq 2.5$. Using $T_K \sim T^0_K e^{\alpha t_C}$, with $\alpha = \tan^{-1}(t_C)$ [8,13], we get $(J/T^0_K)_c \sim 2.5 e^{\alpha t_C}$ [Fig. 2(c)]. Figure 2 shows that the KS $\to$ AF transition [12] can be directly measured in the $\mathcal{G} \equiv dI/dV_{dc}$ curves [Fig. 2(b)] of serial DQD's with $t_C < 1$ (for $t_C > 1$, J plays little role [13]). For $(J/T_K) < (J/T_K)_c$, $\mathcal{G}$ has a zero-bias anomaly (ZBA), reflecting Kondo physics. Upon making ϵ_0 more negative, namely, increasing the ratio (J/T_K) by reducing T^0_K at fixed J, the singlet formation quenches the Kondo effect, and the ZBA smoothly splits to two peaks at finite V_{dc} [10,12,13]. The singlet and the Kondo state coexist in a coherent fashion when $(J/T_K) \approx (J/T_K)_c$ (thick solid line) and $\mathcal{G}_0 \equiv dI/dV_{dc}|_{V_{dc}=0} = 2e^2/h$ (unitary limit) [8,13]. Importantly, the splitting appears before the AF singlet completely develops, $(J/T_K) \lesssim (J/T_K)_c$ [note that the previous estimation of $(J/T_K)_c \simeq 2.5$ from $\epsilon^{\rm GS}_K = \epsilon^{\rm GS}_{\rm AF}$ assumes a complete singlet formation, namely, $||\bar{\chi}|| = J/2$]; this can be attributed to the small interdot tunneling contribution to $\Delta = 4(||\bar{\chi}|| + \tilde{t}_C)$. This prevents us from extracting the value of $||\bar{\chi}||$ from Δ. Nonetheless, the fact that the ZBA splits for $t_C < 1$ [14] is a clear indication that $||\bar{\chi}|| \neq 0$. When $(J/T_K) \gg (J/T_K)_c$ the singlet is completely formed, $||\bar{\chi}|| = J/2$ [10]. Since $J \lesssim \Delta/2$, the splitting of the nonlinear conductance provides an upper bound on J. Experimentally, observation of splitting in $\mathcal{G}$, at a small t_C, as T^0_K is reduced would provide a "smoking gun" for spin-singlet formation. The reverse process, an increase of T^0_K (at fixed t_C), should change the split $\mathcal{G}$ into a ZBA.

In parallel DQD's [Fig. 1(b)] the AF interaction is due to electrostatic coupling rather than tunneling, thereby greatly simplifying the interpretation of the results. This is shown in Fig. 3. For $(J/T_K) < (J/T_K)_c$ [Fig. 3(a)], $\mathcal{G}$ exhibits a ZBA with $\mathcal{G}_0 = 4e^2/h$ (each dot acts as a unitary Kondo channel). As expected, the width of the ZBA decreases as the ratio (J/T_K) grows upon reducing T_K. When $(J/T_K) \approx (J/T_K)_c$ (thick solid line), the dI/dV changes abruptly signaling the Kondo to AF state transition: $\mathcal{G}_0$ drops sharply [22] while the maximum $\mathcal{G}$ appears at finite voltages for which $V_{dc} = J$, namely, $\Delta = 2J$. Importantly, further decrease of T_K, namely, increase of the ratio (J/T_K), *does not change* Δ [Fig. 3(b)], *allowing one to*

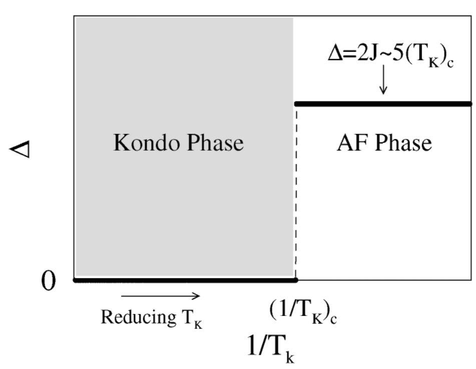

FIG. 4. Phase diagram Δ vs $1/T_K$. At $(1/T_K)_c$, Δ jumps from zero to $2J \sim 5(T_K)_c$. The line $1/T_K \equiv (1/T_K)_c$ separates the Kondo and antiferromagnetic phases.

measure J experimentally. We mention in passing that to access ways of measuring J is of great importance for QIP applications [7]. Also, the robustness of the splitting allows one to anticipate that for $T \neq 0$, $\mathcal{G}_0$ vs T would have a nonmonotonic behavior with a maximum around $T \sim J$ [23]. The underlying physical picture can be understood in terms of the density of states (DOS) (Fig. 3, inset), whereby there is a Kondo state (peak at $\omega = 0$) for $(J/T_K) < (J/T_K)_c$. For $(J/T_K) \approx (J/T_K)_c$, two narrow peaks abruptly form at $\omega = \pm J/2$ indicating the formation of a spin singlet. This scenario does not change so long as $(J/T_K) > (J/T_K)_c$. This has to be compared with the smooth appearance of the splitting in Fig. 2(b). Our results can be interpreted as follows: unlike in serial DQD's, the order parameter characterizing the transition to the AF state can be directly extracted from $\mathcal{G}$ (using $\Delta = 4||\bar{\chi}||$). This way, we propose a phase diagram Δ vs $1/T_K$ which unambiguously signals the transition: using $(J/T_K)_c \sim 2.5$ together with $\Delta = 2J$ we conclude that Δ exhibits, upon reducing T_K, a first order jump. The jump occurs at $1/(T_K)_c$ (which is J dependent) and goes from zero to $2J \sim 5(T_K)_c$ (Fig. 4). Observation of this feature in the proposed phase diagram would constitute direct evidence of the KS to AF singlet transition in parallel DQD's.

In closing we have demonstrated that the transport through DQD's directly reflects the physics of the two-impurity Kondo problem. We give a series of experimental predictions that: (i) unambiguously signal the KS $\rightarrow$ AF transition, and (ii) show how to measure the exchange constant J between the spins of the dots. As the relevant physics occurs at energy scales of the order of T_K we believe that our predictions could be tested experimentally.

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