

Transport properties of a two-impurity system: A theoretical approach

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(Received 14 September 2012; published 4 March 2013)

A system of two interacting cobalt atoms, at varying distances, was studied in a recent scanning tunneling microscope experiment by Bork *et al.* [*Nature Phys.* **7**, 901 (2011)]. We propose a microscopic model that explains, for all experimentally analyzed interatomic distances, the physics observed in these experiments. Our proposal is based on the two-impurity Anderson model, with the inclusion of a two-path geometry for charge transport. This many-body system is treated in the finite- U slave boson mean-field approximation and the logarithmic-discretization embedded-cluster approximation. We physically characterize the different charge transport regimes of this system at various interatomic distances and show that, as in the experiments, the features observed in the transport properties depend on the presence of two impurities but also on the existence of two conducting channels for electron transport. We interpret the splitting observed in the conductance as the result of the hybridization of the two Kondo resonances associated with each impurity.

DOI: [10.1103/PhysRevB.87.115102](https://doi.org/10.1103/PhysRevB.87.115102)

PACS number(s): 73.23.Hk, 72.15.Qm, 73.63.Kv

I. INTRODUCTION

Two interacting magnetic impurities in a bath of conducting electrons are one of the simplest strongly correlated systems, with a rich phase diagram containing a Kondo-regime region and a spin-singlet state where the two impurities are locked into a dimer.¹ The possible quantum phase transition (QPT) between these two phases is dominated by a non-Fermi-liquid quantum critical point (QCP). The parameter that drives the system through the phase diagram is the ratio I/T_K^0 , where I is the interimpurity exchange interaction and T_K^0 is the Kondo temperature of the individual impurities (assumed to be identical). This system has received great attention both theoretically (see below) and experimentally.^{2–6} Experiments performed in two-impurity systems have been able to carry it from the Kondo screened phase to the antiferromagnetic regime, but without achieving precise control of the exchange interaction between the two impurities.^{3,4}

From the theoretical point of view, numerical renormalization-group⁷ calculations on the two-impurity Anderson model (TIAM) focused on the properties of its non-Fermi-liquid QCP⁸ and pointed out that the interimpurity hopping suppresses the critical transition.^{9,10} Several theoretical methods have been used to analyze the TIAM, such as the slave-boson formalism,^{11–17} the numerical renormalization-group,^{8,18} the logarithmic-discretization embedded cluster approximation (LDECA),¹⁹ and the noncrossing approximation.²⁰ The results obtained confirmed the replacement of the critical transition by a crossover as a consequence of the broken even-odd parity symmetry. In addition, a splitting of the zero-bias anomaly in the differential conductance (dC) with an increase of the interdot hopping was observed. This can be understood from the coherent superposition of the many-body Kondo states of each QD (forming bonding and antibonding combinations)^{12,13,17,19} or, alternatively, due to the “parity splitting” caused by direct hopping between the impurities.^{8,21,22}

Recently, a remarkable experiment has been performed where a cobalt (Co) atom, positioned at the tip of a scanning tunneling microscope (STM), is continuously approached to another Co atom adsorbed on an Au(111) surface.²³ The position of the STM tip was varied with subpicometer (pm) accuracy and hence the ratio I/T_K^0 (as defined above) could, in principle, be modified almost continuously. The results in Ref. 23 indicated that the system stayed away from the neighborhood of the QCP, as a peak in the dC, expected to appear at the QPT,^{24,25} was not observed. Starting from a larger interatomic distance, where electron transport occurs through tunneling, the dC initially showed a Fano antiresonance that, with decreasing distance, first narrowed and then evolved into a peak. This peak in the dC, upon further approaching the Co atoms, showed a splitting that was interpreted as consequence of an effective exchange interaction between the two magnetic impurities.^{5,16,26} However, surprisingly enough, this splitting was observed at energy scales smaller than T_K^0 (for details, see Fig. 6 in Ref. 23), in contradiction with theoretical results for two-impurity models that establish a critical coupling I^* greater than the Kondo temperature ($\approx 2k_B T_K^0$) at which the splitting should appear.^{1,16,26–28} In addition (Fig. 6 in Ref. 23), the QPT was washed away by a broad crossover region, the lower end of which roughly coincides with the appearance of the splitting in the dC. The origin of this wide crossover was explained by Bork *et al.*²³ as coming from the strong direct coupling between the electrodes, i.e., the STM tip and the Au substrate themselves.

The Fano antiresonance was discussed in Ref. 23 within the context of a phenomenological model and not by solving a microscopic Hamiltonian. Such analysis assumes the existence in the dC of two Fano antiresonances of Kondo origin with a superposition between them. However, this treatment is not able to obtain either the single peak at the Fermi level or its splitting when the distance between the Co atoms is reduced, as observed in the experiments. To provide an explanation for the latter feature of the dC, a microscopic model was

proposed and solved within the numerical renormalization-group formalism,²³ incorporating an indirect coupling between the Co atoms. It is then important to theoretically account for such an evolution of the dC by using a realistic and *single* microscopic model, capable of reproducing all the experimental features described above. This model provides, as well, evidence that a splitting in the dC is compatible with both impurities still being independently in the Kondo screened regime. This complete explanation is particularly important because a consistent characterization of the physics observed in such an experiment, as a function of distance (or, equivalently, interaction between the impurities), is still lacking in the literature.

In this paper, we show that the double-Co experiment described above can be completely interpreted, for all interimpurity distances studied, by a model that incorporates, as essential ingredients, *a direct hopping between the Co atoms and another one between the electronic reservoirs*. In particular, the results show to what extent the interplay between the direct and the indirect inter-impurity hoppings influences the transport properties of the system. Moreover, we show that the splitting in the conductance is compatible with a Kondo screened ground state.

II. MODEL

The Hamiltonian is written as a sum of three terms, namely,

$$H = H_{\text{imp}} + H_{\text{hyb}} + H_{\text{leads}}, \quad (1)$$

where

$$H_{\text{imp}} = \sum_{i=\alpha,\beta;\sigma} \left(\epsilon_i n_{i\sigma} + \frac{U}{2} n_{i\sigma} n_{i\bar{\sigma}} \right) \quad (2)$$

describes the isolated impurities, ϵ_i (where $i = \alpha, \beta$) being the energy of each localized impurity, U is the on-site Coulomb interaction, and $\sigma = \pm$ is the spin orientation. The contribution

$$H_{\text{hyb}} = \sum_{\sigma} t_L c_{L,1\sigma}^{\dagger} c_{\alpha\sigma} + t_R c_{R,1\sigma}^{\dagger} c_{\beta\sigma} + t_{\alpha\beta} c_{\alpha\sigma}^{\dagger} c_{\beta\sigma} + t_{LR} c_{L,1\sigma}^{\dagger} c_{R,1\sigma} + \text{H.c.} \quad (3)$$

describes the hybridization of each impurity with the first site of its adjacent metallic lead, the hybridization between both impurities, and the direct tunneling between the left (L) and the right (R) electron reservoirs (in that order). Finally,

$$H_{\text{leads}} = t \sum_{j=L,R} \sum_{i=1;\sigma}^{\infty} (c_{j,i\sigma}^{\dagger} c_{j,i+1\sigma} + \text{H.c.}) \quad (4)$$

describes the L, R leads, represented by two semi-infinite chains of noninteracting sites, with hopping t between adjacent sites. Note that the hopping between each impurity and the opposite electrode was not included, since the experiments indicated that these couplings do not play an important role in the charge transport, as no change in the dC line shape for a bare tip approaching a Co atom was observed.²³ The interaction between each impurity and the opposite electrode is indirectly included through the hopping t_{LR} between the leads. In addition, its explicit inclusion also proved unnecessary, as the experimental results could be explained with a simpler model. In fact, our results show that the hypothesis in Ref. 23,

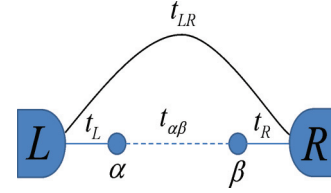


FIG. 1. (Color online) Schematic of the model studied in this work. α and β represent the two Co atoms.

just mentioned, is correct. The model is depicted in Fig. 1. The transport properties were calculated within the finite- U slave boson mean-field approximation (SBMFA),²⁹ although, for the sake of comparison, some results were obtained using the LDECA.¹⁹ Finally, for the sake of simplicity, we adopt a symmetric model (i.e., $t_L = t_R = t'$).

We define T_K^0 as the Kondo temperature for each Co atom in the two independent single-impurity Anderson models (as obtained through $t_{LR} = t_{\alpha\beta} = 0$). Taking, in units of t , $U = 0.8$, $t' = 0.25$, and $\epsilon_i = -U/2$, we obtain $T_K^0 = 0.0073$. Considering $t = 1.3$ eV for Au, one obtains $T_K^0 \approx 9.5$ meV \equiv K, roughly the same Kondo temperature measured in the experiments with Co atoms on Au.²³ T_K^0 is a reference for comparison with experiments. The values of U and t' were chosen, within the convergence parameter space of the SBMFA method, so that $U/\Gamma \sim 14$, where $\Gamma = \pi t'^2 \rho(E_F)$, $\rho(E_F)$ being the leads' density of states at the Fermi energy. This U/Γ value assures that the single-impurity system is deep inside the Kondo regime at zero temperature.

Our model includes the hopping t_{LR} between the electron reservoirs, providing a channel through which the dots couple indirectly and also a weaker direct hopping $t_{\alpha\beta}$ between the Co atoms that, as mentioned above, turns out to be essential for reproducing the experimental results. The assumption that $t_{\alpha\beta} \ll t_{LR}$ is in accordance with the hypothesis made in Ref. 23, namely, that for a vertical approach between the STM tip and the surface, the interaction between the d orbitals of the Co atoms is very weak. Assuming the hoppings to follow $t_{LR} = Ae^{\gamma z}$ and $t_{\alpha\beta} = Be^{\delta z}$, where z is a parameter representing the interimpurity distance, then a decrease in z results in an exponential increase in $t_{\alpha\beta}$ and t_{LR} (for $\gamma, \delta < 0$). After an extensive survey, by varying the parameters so as to take into account the constraint $t_{\alpha\beta} \ll t_{LR} < t$, as discussed above, and the obvious conditions $t_{\alpha\beta} \approx 0$ and $t_{LR} \ll 1$ for the largest z value used, we found that the parameters that best allow the reproduction of the experimental results are³⁰ $A = 0.4066$, $B = 0.0305$, $\gamma = -0.002534$, and $\delta = -0.00973$. This parametrization allows us to reproduce the dC experimental results for the whole range of z values [compare our Fig. 2 with Fig. 4(b) in Ref. 23].

In the SBMFA, the dC is calculated using the Keldysh formalism.³¹ For simplicity, we assume electron-hole symmetry. The dC can then be written as $dC = 4\pi^2 t^4 \text{Im}\{G_L(V/2)\} \text{Im}\{G_R(V/2)\} |G_{LR}^V(V/2)|^2$, where $G_L = G_R$ are the reservoirs' noninteracting Green's functions and $G_{LR}^V(\omega)$ is the many-body propagator from left to right, under the presence of a bias V between the L and the R reservoirs. However, it is known³² that for a two-impurity system the SBMFA results obtained for the equilibrium situation are very similar to the nonequilibrium results, as long as V is

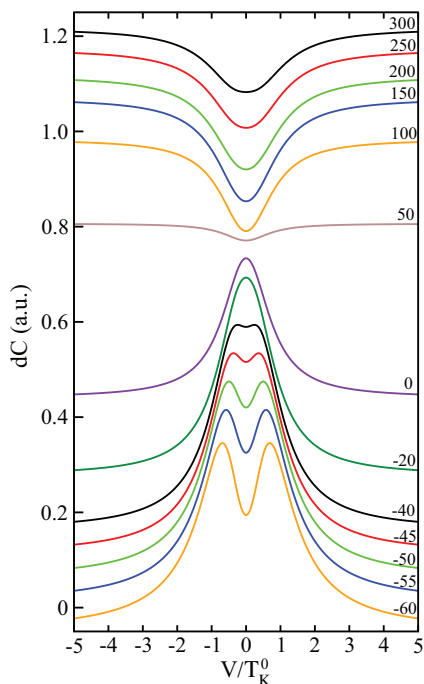


FIG. 2. (Color online) Differential conductance as a function of V/T_K^0 for $300 \leq z \leq -60$ [where z is the separation between the impurities (in pm), indicated at the right]. Each curve, calculated using SBMFA, has been shifted vertically for clarity.

smaller than a few times T_K^0 . Under these conditions, the propagator $G_{LR}^V(\omega)$ is almost independent of the external bias V . Therefore, we assume its complete independence from V and calculate dC as if the system were in equilibrium.

III. RESULTS

In Fig. 2, we present the SBMFA results for dC as a function of V/T_K^0 . The dC curves present three fundamental features that should be emphasized: (i) for negative values of z , there is a double-peak structure, displaying a splitting that decreases with increasing z ; (ii) eventually, still for negative values of z , the splitting is totally suppressed, becoming a single peak; and (iii) for higher z values, a Fano antiresonance develops centered at $V = 0$, with increasing width as z increases (see also Fig. 4). The behavior just described is qualitatively and semiquantitatively similar to that observed in the experiments by Bork *et al.* [Fig. 4(b) of Ref. 23]. However, the range of z values for which our results show a single peak is slightly larger than in the experiments, where an antiresonance line shape persists down to $z \approx -30$, while our results show a single-peak feature already at $z = 0$.

Note that a split peak in dC was also obtained by using the numerical renormalization group,²³ with a model where only the hopping between the electronic reservoirs was included. However, the overall agreement between our theoretical results and the experiments requires the inclusion, as a crucial parameter, of the *direct* interimpurity interaction $t_{\alpha\beta}$. This direct hopping opens another channel through which electrons can flow, hence, at larger distances, this results in a Fano dip, when $t_{\alpha\beta}$ is very small. The dip transforms into a peak as the ratio $t_{\alpha\beta}/t_{LR}$ increases (from 0.009

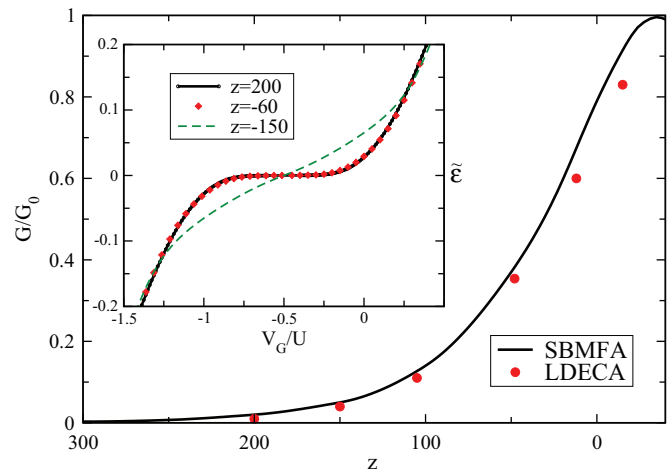


FIG. 3. (Color online) Conductance G/G_0 as a function of z . Compared to Fig. 4(a) in Ref. 23, we see the same overall behavior. Inset: Renormalized energy level $\tilde{\epsilon}_\alpha = \tilde{\epsilon}_\beta = \tilde{\epsilon}$ as a function of gate voltage V_g . The plateau at the Fermi energy for both $z = 200$ [solid (black) line] and $z = -60$ [diamond (red) line] indicates that the system stays in the Kondo regime. For larger hopping values, $t_{\alpha\beta} = 0.06$ and $t_{LR} = 0.13$, the plateau starts to be suppressed, as shown by the dashed (green) curve for $z = -150$, reflecting that the system enters a crossover regime (see text).

at $z = 300$ to 0.115 at $z = -60$). At the lower distances (larger $t_{\alpha\beta}$) the Kondo states of each impurity superpose, forming bonding/antibonding many-body states that result in a splitting in the dC, as shown in our results in Fig. 2. It is important to point out that a splitting (with varying magnitude in relation to T_K^0) in the dC has also been obtained in previous studies on similar models. In these studies, a direct hopping and/or a superexchange interaction J between the impurities has been taken in account.^{8,13,14,17,19,21,22,27} The splitting thus obtained was interpreted as a superposition of the two “independent” many-body Kondo states^{13,14,17,19} or, alternatively (but not in contradiction with the previous idea), as being caused by the parity splitting of the occupation number between even and odd channels that takes place when the direct hopping between the impurities has a magnitude comparable to the low-energy scale of the model, as in our case.^{8,21,22}

Using SBMFA, we have calculated the phase difference (not shown) between the two channels through which the current can flow, the one between the impurities via $t_{\alpha\beta}$ and the one through the reservoirs t_{LR} .³³ We obtained that this phase difference is 0 for all values of z , indicating that these two channels interfere constructively, satisfying the Onsager relation for systems of closed geometry, which establishes that this phase difference can be only 0 or π .

In Fig. 3 we present SBMFA [solid (black) line] and LDECA [filled (red) circles] results for the conductance G/G_0 as a function of z (where $G_0 = 2e^2/h$ is the quantum of conductance), which can be compared to Fig. 4(a) in Ref. 23. Since LDECA is exact at the Fermi energy,¹⁹ the excellent agreement between SBMFA and LDECA gives support to the SBMFA results shown in this paper. The main difference from the experimental results is that our G/G_0 values increase more smoothly as z decreases. This can be associated with

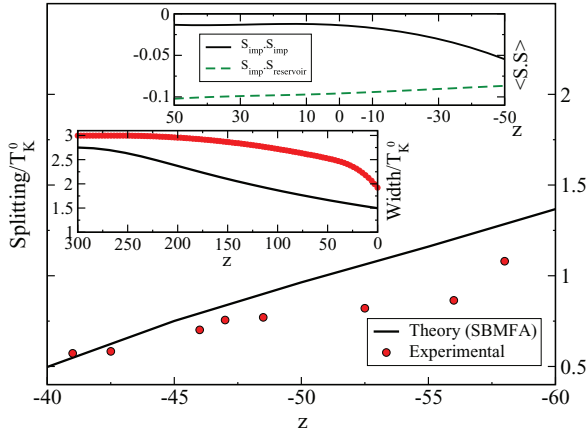


FIG. 4. (Color online) Splitting as a function of z [solid (black) curve]. Lower inset: Width of the antiresonance, as a function of distance, in units of T_K^0 . The SBMFA results (black line) are similar to those obtained in Figs. 5(b) and 5(a) in Ref. 23 [reproduced schematically here as filled (red) circles]. Upper inset: Spin-spin correlations between the impurities [solid (black) curve] and between each impurity and its adjacent reservoir [dashed (green) curve; see text].

the fact that, as expected, the experimental results are very dependent on the distance between the atoms. As stated by Bork *et al.*, “mechanical relaxation” effects should be at play when a transition from “tunneling” to “point contact” occurs as the tip gets closer to the surface.²³ Hence, at this transition the real distance z and the associated hopping parameters $t_{\alpha\beta}$ and t_{LR} are difficult to determine. Besides, other matrix elements such as t_L and t_R , assumed to be constant, may also vary at this transition.

In the SBMFA, when the system is in the Kondo regime, there is a plateau at the Fermi energy ($E_F = 0$) in the renormalized energy level of each impurity ($\tilde{\epsilon}_\alpha = \tilde{\epsilon}_\beta = \tilde{\epsilon}$), as a function of the gate voltage V_g .^{29,34} As shown in the inset in Fig. 3, for the region of interest, the plateau is perfectly defined [compare the solid (black) curve for $z = 200$ with the almost-identical diamond (red) curve for $z = -60$], indicating that the system remains in the Kondo regime, although the splitting in dC is relatively large for the lowest values of z , as shown in Fig. 4, discussed below. For larger values of $t_{\alpha\beta}$ and t_{LR} , the plateau is partially eliminated as shown in the dashed (green) curve for $z = -150$. Thus, for large hopping values there is an effective antiferromagnetic spin-spin correlation between the Co atoms that starts to suppress the Kondo regime, and the system enters a crossover region that is compatible with measurements presented in Fig. S.6 of Ref. 23. In order to confirm our SBMFA results, spin-spin correlations were calculated with LDECA. These are shown in the upper inset in Fig. 4. It can be seen that even when a splitting is present, the AF spin-spin correlation between each impurity and its adjacent reservoir (calculated between the impurity and the first site of the adjacent noninteracting chain), which may be used to characterize the Kondo effect, is dominant with respect to the AF correlation between the impurities. This result coincides with the scenario provided by the SBMFA approach: as the system stays in the Kondo regime, the splitting in dC is a consequence of the

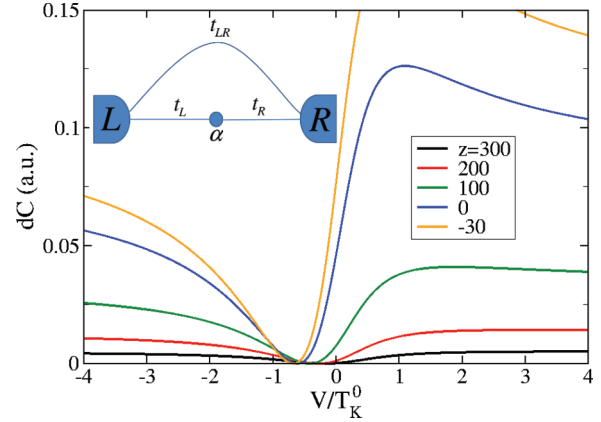


FIG. 5. (Color online) Differential conductance for a single impurity as a function of V/T_K^0 . The behavior is markedly different from the case of two atoms, as in the experiments.

hybridization of the Kondo resonances associated with each impurity.¹⁷

Figure 4 shows a comparison, as a function of z , of the SBMFA dC splitting (distance between peaks) and the experimental results from Ref. 23, both in units of their corresponding single-impurity Kondo temperature T_K^0 ($75 \text{ K} \approx 6.5 \text{ meV}$ in the experiments). The SBMFA splitting is almost linear in z . In the lower inset we compare experimental and SBMFA results for the width of the dC Fano antiresonance in the interval $300 \geq z \geq 0$, scaled by T_K^0 . We fit the SBMFA dC curve with a Fano antiresonance to extract its width. The sharper decrease in the dip width for the experimental results at $z \approx 25$ can be ascribed to the relaxation process mentioned above. We stress the fact that as the magnitude of the splitting may change when the experiment is repeated, a qualitative and semiquantitative description of the experimental results should be satisfactory.

IV. SINGLE-IMPURITY CASE

In the case of a single Co impurity, the experimental dC shows only one dip, which neither diminishes its width nor transforms into a peak as the distance between tip and surface is reduced.^{23,35} In order to study the differences between the single- and the double-impurity cases, we have carried out a z dependence study for the single-impurity model. The model is depicted in the inset in Fig. 5. The hopping between the impurity and one of the reservoirs (t_R ; modeling the STM tip) and the hopping between the two reservoirs (t_{LR}) are varied in the same way as in the double-atom case. The SBMFA results (Fig. 5) show an asymmetric antiresonance, as previously obtained.³⁶ We checked that the dip’s width does not decrease upon changing z . Besides, the dip reaches its minimum at negative values of bias voltage, as noted in Ref. 36 for an $S = 1/2$ impurity, which is our case. The value of G/G_0 at the Fermi level (not shown) is much smaller than in the two-impurity case. Therefore, these results show that the reduction of the antiresonance width, the appearance of a peak and its splitting, as observed in the experiments in Ref. 23, are a consequence of the presence of a second impurity, interacting with the first.

V. CONCLUSIONS

Summarizing, the model proposed in this paper to study the transport properties through two Co atoms in series correctly describes the behavior observed in STM experiments for the whole parameter range. In that respect, the inclusion of a direct hopping between the atoms, *besides* the one between the electron reservoirs, proves to be an essential ingredient. In addition, we find that the electrons interfere constructively along the two possible paths, but in the case of a single impurity these two same paths give completely different transport properties, as observed in the experiments. In our model, the direct and indirect couplings between the impurities result in an antiferromagnetic spin-spin correlation between them. This

interaction is not strong enough to take the system out of the Kondo regime. The splitting in the dC is indeed a consequence of the hybridization of the two Kondo resonances.

ACKNOWLEDGMENTS

We gratefully acknowledge fruitful discussions with C. A. Büsser, L. O. Manuel, A. E. Trumper, C. J. Gazza, and E. Vernek. G.B.M. acknowledges financial support from the NSF under Grants No. DMR-1107994, No. DMR-0710529, and No. MRI-0922811. E.V.A. acknowledges financial support from the Brazilian agencies CNPq (CIAM project) and FAPERJ (CNE).

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