Interaction between Kondo impurities in a quantum corral

G. Chiappe\textsuperscript{a} and A. A. Aligia\textsuperscript{b}

\textsuperscript{a} Departamento de Física, FCEyN Universidad de Buenos Aires,
Pabellón I, Ciudad Universitaria, (1428) Buenos Aires, Argentina.

\textsuperscript{b} Centro Atómico Bariloche and Instituto Balseiro, Comisión Nacional de Energía Atómica, 8400 Bariloche, Argentina.

(Received November 20, 2018)

We calculate the spectral densities for two impurities inside an elliptical quantum corral, using exact diagonalization in the relevant Hilbert subspace and embedding into the rest of the system. For one impurity, the space and energy dependence of the change in differential conductance \( \Delta dI/dV \) observed in the quantum mirage experiment is reproduced. In presence of another impurity, \( \Delta dI/dV \) is very sensitive to the hybridization between impurity and bulk. The impurities are correlated ferromagnetically between them. A hopping \( \gtrsim 0.15 \) eV between impurities destroys the Kondo resonance.
In recent years, the manipulation of single atoms on top of a surface using scanning tunneling microscopy (STM) was made possible, and quantum corrals have been assembled by depositing a closed line of atoms or molecules on noble metal surfaces. The local conduction spectral density of states $\rho_d(\omega)$, measured by differential conductance $dI/dV$ reveals patterns that remind the wave functions of two-dimensional noninteracting electrons under the corresponding confinement potential. In a recent experiment, a Co atom has been placed at a focus of an elliptic quantum corral, and the corresponding Kondo feature is observed not only at that position, but also at the other focus, where a “mirage” is formed as a consequence of the quantum interference. Several variants of this experiment, some of them involving several impurities (Co atoms) and eventually mirages inside the corral, are being performed. The main features of the observed space and voltage dependence of $dI/dV$ have been reproduced by several theories. Since in Refs. the density of states per spin at the impurity $\rho_d(\omega)$ is assumed rather than calculated, these theories cannot account for the interaction between impurities. In Ref. the Kondo effect is absent, and perturbation theory in the Coulomb repulsion $U$ is restricted to small values of $U$.

The aim of the present work is to present a theory of the quantum mirage which is able to reproduce the experimental results for the case of one impurity and give reliable predictions when more than one impurity is inside the corral. We show that experiments with two impurities can elucidate the role of the direct hybridization between the impurity and the bulk $V_b$. Scattering theories obtained an excellent agreement with experiment assuming that the resonant level width due to hybridization with bulk states $\delta_b$ is as large as that due to the surface $\delta_s$. On the other hand, the larger density of $s$ and $p$ states at the surface and the rapid decay of the hybridization matrix elements with distance suggest that $\delta_b$ is negligible, and the experiment for one impurity can also be explained if $V_b = 0$. A calculation of $\delta_b$ has not been made and experimentally the situation is still unclear. The role of $V_b$ is not only crucial for a correct theory of the mirage experiment, but also for the general understanding of the interaction between metallic surfaces and adsorbates. Since actually $\delta_b$ was introduced as a phenomenological parameter which takes into account the electrons lost in the scattering process, one expects that if $\delta_b = \delta_s$, the interaction between impurities is roughly a fourth of that for $V_b = 0$ if the same total width $\delta_b + \delta_s$ is kept.

We obtain the ground state of the Anderson model in a cluster which contains one or two impurities and the relevant conduction states inside a hard wall ellipse using the Lanczos method. These states are then mixed with bulk states using an embedding method. This embedding is essential to describe the low energy physics.

The Hamiltonian can be written as:

$$H = \sum_{j\sigma} \varepsilon_j c_{j\sigma}^\dagger c_{j\sigma} + E_d \sum_i d_{i\alpha}^\dagger d_{i\alpha} + U \sum_i d_{i\uparrow}^\dagger d_{i\uparrow} d_{i\downarrow}^\dagger d_{i\downarrow} + \sum_{ij\sigma} V[\varphi_j(R_i) d_{i\sigma}^\dagger c_{j\sigma} + H.c.] + H'$$

(1)

Here $c_{j\sigma}^\dagger$ creates an electron on the $j$th conduction eigenstate of a hard wall elliptic corral with wave function $\varphi_j(r)$ and $d_{i\alpha}^\dagger$ is the corresponding operator for the impurity at site $R_i$. The hybridization of these states with bulk states of the same symmetry is described by $H'$. We assume that each of the impurity and conduction states mixes with a different continuum of bulk states:

$$H' \approx t \sum_{j} (c_{j\downarrow} b_{j\alpha} + H.c.) + V_b \sum_{i\alpha} (d_{i\alpha}^\dagger b_{i\sigma} + H.c.).$$

(2)

The $b_{i\alpha}$ represent bulk states for which the unperturbed density is 0.05 states/eV, similar to the density of bulk $s$ and $p$ states. Approximation is justified by comparison of the non-interacting Green functions for hard wall corrals and more realistic boundary potentials.

The dressed matrix $G$ describing the one-particle Green function is calculated by solving the Dyson equation $G = g + gH'G$, where $g$ is the corresponding matrix for $H' = 0$. This equation is exact for $U = 0$ or $H' = 0$, and in the general case represents an infinite sum of particular diagrams in perturbation theory in $H'$ (the chain approximation). The $\varphi_j(r)$ are obtained as described elsewhere. We choose the ellipse with eccentricity $c = 1/2$ and size such that the state $j = 42$ lies at $\epsilon_F$. The change in $dI/dV$ ($\Delta dI/dV$) after an impurity is placed inside the corral is determined by the conduction states which lie near $\epsilon_F$ and have a strong amplitude $|\varphi_j(R_i)|$ at the impurity position. For $R_i$ at one focus they are $j = 32, 35, 42$ and 51. We have also included $j = 24$ and 62, although this inclusion leads to negligible changes in the results. We took the impurity parameters $E_d = -1$ eV and $U = 3$ eV. We consider first the case $V_b = 0$ and one impurity at the left focus ($R_i = (-0.5a, 0)$). The value $V = 0.04$ eV was chosen to lead to the observed width of $\Delta dI/dV$. The remaining parameter $t$ controls the amplitude of the mirage at the right focus.

In Fig. 1(a) we represent the resulting impurity spectral density $\rho_d(\omega)$ for two values of $t$. A clear Kondo
peak is obtained and for $t \gtrsim 0.3$ eV its width is very weakly dependent on $t$. Instead, for $t \to 0$, the peak splits into two very narrow peaks out of $\epsilon_F$. In contrast to $\rho_d(\omega)$, the magnitude of the change in the conduction density $\Delta \rho_c(r, \omega)$ at the empty focus ($r = -R_i$) is quite sensitive to $t \gtrsim 0.3$ eV: as $t$ increases, the width of the conduction states increases, the weight of the states 32, 35 and 51 (odd under the reflection through the minor axis of the ellipse $\sigma$) decreases as a consequence of the negative interference of these states with the even state 42. The differential conductance $dI/dV$ at zero temperature is proportional to the density $\rho_1$ of the state $| \phi_1 \rangle$

$$ f_\sigma(r) = \sum_j \varphi_j(r) c_{j\sigma} + q d_{j\sigma}. $$

$q$ is related to Fano’s interference parameter and represents the effect of a direct tunneling from the tip to the impurity. Therefore, it is relevant only very near the impurity. For $q = 0$, $\rho_f(r, \omega) = \rho_c(r, \omega)$. In Fig. 1 (b) we represent the effect of adding the impurity on $\rho_f(\pm R_i, \omega)$ ($\Delta \rho_f \sim \Delta dI/dV$). At the impurity site $R_i$, $\Delta \rho_c(r, \omega)$ is asymmetric and smaller at the right of the valley. This is a consequence of the asymmetry of the hybridization around $\epsilon_F$ ($| \varphi_{51}(R_i) | > | \varphi_{35}(R_i) |$). A symmetric line shape, as observed in the experimental $\Delta dI/dV$ is restored for $q \sim 1$. The effect of this $q$ is consistent with the fact that on a clean surface, $\Delta dI/dV$ is larger at the right of the peak. Another nice fact is that the minimum of $\Delta \rho_f$ for $q = 1$ lies at the experimental position 1 meV. At the right focus ($r = -R_i$) we obtain a similar valley, although slightly asymmetric and shifted to the left. Increasing $t$ from 0.4 to 0.5, the magnitude of this valley is strongly reduced (its minimum is shifted above $-5/eV$) but its shape and width is retained. At the impurity position there are no significant changes.

The space dependence of $\Delta \rho_f$ for $q = 0$ is represented in Fig. 2. As in the experimental $\Delta dI/dV$, the main features of $| \varphi_{42}(r) |^2$, attenuated at the right focus, are displayed. Thus, the theory reproduces the space and energy dependence of $\Delta dI/dV$ observed in the experiment. All results so far agree semiquantitatively with perturbative calculations. To see how the results change if $\delta_b \equiv \delta_a$ is assumed, we have reduced $V_b$ by a factor $\sqrt{2}$. This should reduce $\delta_b$ by a factor 2. Increasing $V_b$ from zero to 1.2 eV, the original width of $\rho_d$ is restored. The intensity is reduced by a factor $\sim 2$ (due to the strong energy dependence of $\delta_a$). $\Delta \rho_c$ turns out to be $\sim 4$ times smaller. The additional factor 2 can be understood from the fact that the change in conduction electron Green function is proportional to $V^2 G_d(\omega)$, where $G_d(\omega)$ is the impurity Green function. Except for these factors, the results are surprisingly similar to the previous ones. Some of them will be displayed in Fig. 4.

We now turn to the case of two impurities, one at each focus, for $V_b = 0$. The spectral density for one of these impurities is represented in Fig. 3 (a). Comparison with the previous case (Fig. 1), shows that the peak around $\epsilon_F$ broadens (by a factor $\sim 1.5$), loses intensity and shifts to lower energies. In addition, another very narrow peak appears $\sim 13$ meV below $\epsilon_F$. An analysis of the energy dependence of the density of the individual conduction states shows that the broad peak around $\epsilon_F$ is due to hybridization with even states (mainly 42), while the narrow peak reflects the hybridization of the impurity states with odd states (mainly 51). The difference in $dI/dV$ with respect to the empty corral is however, not so different as in the previous case. This is due to the effect of the unperturbed Green functions of the conduction states and is also present in the one impurity case. Nevertheless, a decrease in the amplitude and a broadening of the depression should be observed in $\Delta dI/dV$ and seems in qualitative agreement with recent experiments. The space dependence is similar to that for one impurity (Fig. 2) but it is of course, symmetric under reflection through the minor axis $\sigma$, and not attenuated at the right focus. Qualitatively, the shape of $\rho_d$ can be understood looking at the non-interacting case $U = 0$, $E_d \sim \epsilon_F$. In this case, for one impurity, the Kondo peak is replaced by a Lorentzian near $\epsilon_F$. For two impurities, a change of basis of the $d$ orbitals to $e_{\sigma} = (d_{1\sigma} + d_{2\sigma})/\sqrt{2}$, $o_{\sigma} = (d_{1\sigma} - d_{2\sigma})/\sqrt{2}$, separates the problem into those corresponding to even and odd states under $\sigma$. The even state hybridizes mainly with conduction state 42, to form a resonance near $\epsilon_F$, roughly twice wider than for one impurity due to the larger effective hybridization. Instead, the odd state $o_{\sigma}$ is displaced towards lower energies due to hybridization with state 51. The interactions should modify the quantitative details of this picture. However, we expect that it remains qualitatively valid, as suggested by the above mentioned energy distribution of the different conduction states.

To gain insight into the nature of the ground state, we have also calculated spin-spin correlation functions for $t = 0$. A reliable method to include $H'$ in these calculations has not been developed yet. For one impurity we obtain $\langle S_i \cdot s_{42} \rangle = -0.73$, where $S_i$ is the spin of the impurity $i$ and $s_{j}$ is the spin of the conduction state $j$. This value is close to the minimum possible one $-3/4$. For $j \neq 42$, $\langle S_i \cdot s_{j} \rangle$ are very small, but this, and the large magnitude of $\langle S_i \cdot s_{42} \rangle$, are affected to a certain degree by the neglect of $H'$ in this calculation. The space dependence of $\langle S_i \cdot s(r) \rangle$, where $s(r)$ the conduction spin at position $r$ follows closely $| \varphi_{42}(r) |^2$. For two impurities we find $\langle S_1 \cdot s_{42} \rangle = -0.47$ and $\langle S_1 \cdot S_2 \rangle = 0.21$. In the limit of large $U$, one expects that the main features of the spin dynamics for $V_b = 0$ are described by the Hamiltonian $H_0 = J(S_1 + S_2) \cdot s_{42}$, where $J > 0$ is the Kondo coupling. The ground state of this Hamiltonian
is a doublet in which the impurity spins are correlated ferromagnetically between them \((\langle S_1 \cdot S_2 \rangle = 1/4)\) and antiferromagnetically with state 42 \((\langle S_1 \cdot S_{42} \rangle = -1/2)\). These values are near to those we find. The effect of the hybridization of state 42 with bulk states can be modelled by a tight binding Hamiltonian in terms of Wilson’s orbitals. \(H_0\) is the strong coupling fixed point of Wilson’s renormalization group. An analysis of the stability of this fixed point using perturbation theory as in Ref. [21] leads to the conclusion that the ground state is a doublet for \(V_b = 0\). However, we expect that as soon as \(V_b \neq 0\), the doublet is screened at a very low temperature.

For the set of parameters corresponding to \(\delta_b \cong \delta_s\), \(\rho_4(\omega)\) is much more similar to the one impurity case, although a structure reminiscent of a splitting is also present near its maximum. In contrast to the case of \(V_b = 0\), when a second impurity is added, the depression in \(\Delta dI/dV\) at one impurity site \(R_i\) increases and its width is roughly the same (see Fig. 4). Comparison with results when \(t\) is increased from 0.4 eV to \(t = 0.5\) eV (not shown) suggests that when \(\delta_b \cong \delta_s\), \(\Delta dI/dV\) at \(\pm R_i\) for two impurities is roughly the sum of the results at \(R_i\) and \(-R_i\) for one impurity. This is what one would expect if the interaction is very small.

Coming back to the case \(V_b = 0\), we have also verified that qualitatively similar features in \(\Delta dI/dV\) are obtained at one focus, if one impurity is placed there and the second impurity is put at another extremum of \(\varphi_{42}(r)\), like \((0.22a,0)\) (instead of placing it at the other focus). In this case, the spectral densities at \((0.22a,0)\) have some additional structure due to an important admixture of the state 41. [22] In contrast, if both impurities are placed close to the same focus and near each other, a moderate hopping \(t \sim 0.15\) eV or larger between them is sufficient to destroy the Kondo resonance. In particular \(\Delta dI/dV\) becomes flat and featureless near \(\epsilon_F\).

In summary, we have studied the spectral density for impurities inside a quantum corral, using a many-body approach which treats exactly the correlations in the impurities and their hybridization with the relevant conduction states at the surface, and treats approximately the hybridization with bulk states. We have been able to reproduce the main features of the mirage experiment for one impurity inside the corral. The experiment for one impurity cannot determine the relative importance of the direct hybridization of the impurity with bulk states, unless the tunneling matrix elements and other details are known accurately. Instead, for two impurities inside the corral, the differential conductance is very sensitive to this hybridization. For the parameters of the experiment, the spins of both impurities are antiferromagnetically coupled with the conduction electrons, and ferromagnetically correlated between them provided they are placed sufficiently far apart, so that the hopping between them can be neglected. If this hopping is larger than 0.15 eV, there is a tendency to form a singlet state between both impurity spins and the Kondo resonance disappears. To our knowledge, this is the first theory which is able to describe the line shape of the differential conductance when more than one Kondo impurity is inside the quantum corral.

This work benefitted from PICT 03-00121-02153 of ANPCyT and PIP 4952/96 of CONICET. We are partially supported by CONICET.

[1] D.M. Eigler and E.K. Schweizer, Nature 344, 524 (1990).
[2] M.F. Crommie, C.P. Lutz, and D.M. Eigler, Science 262, 218 (1993).
[3] E.J. Heller et al., Nature 363, 464 (1994).
[4] H.C. Manoharan, C.P. Lutz, and D.M. Eigler, Nature 403, 512 (2000).
[5] H.C. Manoharan, PASI Conference, Physics and Technology at the Nanometer Scale (Costa Rica, June 24 - July 3, 2001).
[6] O. Agam and A. Schiller, Phys. Rev. Lett. 86, 484 (2001).
[7] G.A. Fiete et al., Phys. Rev. Lett. 86, 2392 (2001).
[8] D. Porras, J. Fernández-Rossier, and C. Tejedor, Phys. Rev. B 63, 155406 (2001).
[9] M. Weissmann and H. Bonadeo, Physica E 10, 44 (2001).
[10] A.A. Aligia, Phys. Rev. B 64, 121102(R) (2001).
[11] A.A. Aligia, cond-mat/0110081.
[12] A. Euceda, D.M. Bylander, and L. Kleinman, Phys. Rev. B 28, 528 (1983).
[13] V. Ferrari et al., Phys. Rev. Lett. 82, 5088 (1999); C.A. Büsser et al., Phys. Rev. B 62, 9907 (2000).
[14] A numerical diagonalization without embedding has been performed to study mirages on a spherical surface [K. Hallberg, A. Correa, and C.A. Balseiro, cond-mat/0106082].
[15] W.B. Thimm, J. Kroha, and J. von Delft, Phys. Rev. Lett. 82, 2143 (1999).
[16] Here the wave functions are adimensional and normalized as \(\int dx dy \varphi_a(r) \varphi_b(r)/(ab) = \delta_{ij}\), where \(a(b)\) is the semimajor (semiminor) axis of the ellipse.
[17] A. Correa, K. Hallberg, and C.A. Balseiro, unpublished.
[18] E.V. Anda, J. Phys. C 14, L1037 (1981); W. Metzner, Phys. Rev. B 43, 8549 (1991).
[19] O.Újsághy et al., Phys. Rev. Lett. 85, 2557 (2000).
[20] A. Schiller and S. Hershfield, Phys. Rev. B 61, 9036 (2000).
[21] R. Allub and A.A. Aligia, Phys. Rev. B 52, 7987 (1995).
[22] This calculation was made replacing the states 24 and 62, which practically do not change the results, with states 41 and 49, which have an important hybridization with the impurity at \((0.22a,0)\).
Fig. 1: (a) Impurity spectral density as a function of energy for two values of \( t \). (b) Change in the density of the mixed state \( f_\sigma \) (Eq. (3)) at the impurity site (left focus) for two values of \( q \) and at the other focus for \( t = 0.4 \) eV.

Fig 2: Contour plot of \( \Delta \rho_c(r, \omega) \) for \( t = 0.4 \) eV and \( \omega = 10 \) meV.

Fig 3: (a) Impurity spectral density for one impurity at each focus and two values of \( t \). (b) Change in the density of the mixed state \( f_\sigma \) after addition of both impurities (Eq.(3)), at one impurity site for two values of \( q \). Parameters are \( V = 0.04eV \), \( V_b = 0 \) and \( t = 0.4 \) eV.

Fig 4: \( \Delta \rho_c(r, \omega) \) as a function of \( \omega \) for the case of one impurity at the left focus (full and dashed lines) or one impurity at each focus (dashed dot dot line). Parameters are \( V = 0.04eV/\sqrt{2} \), \( V_b = 1.2 \) eV and \( t = 0.4 \) eV.
Fig. 1
Fig. 3
Fig. 4

$\Delta \rho_c (1/eV)$ vs $\omega (eV)$

- **left focus**
- **right focus**
- **2 imp**

$t = 0.4$