Ab initio Study of Mirages and Magnetic Interactions in Quantum Corrals

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The state of the art *ab initio* calculations of quantum mirages, the spin polarization of surface-state electrons, and the exchange interaction between magnetic adatoms in Cu and Co corrals on Cu(111) are presented. We find that the spin polarization of the surface-state electrons caused by magnetic adatoms can be projected to a remote location and can be strongly enhanced in corrals, compared to an open surface. Our studies give clear evidence that quantum corrals could permit one to tailor the exchange interaction between magnetic adatoms at large separations.

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As the physical size of a system approaches atomic dimensions, quantum effects are known to play a significant role. One of the most striking illustrations of the quantum behavior in atomic-scale nanostructures is the observation of the electronic confinement of surface-state electrons in the Fe corral constructed in an atom-by-atom fashion on a copper(111) surface [1]. The structures which confine the electrons on surfaces can be built using the manipulation into the required geometry of individual adsorbed atoms by the scanning tunneling microscope (STM) [2]. Altering the size and shape of artificial structures, one could affect their quantum states. The controllable modification of quantum states could permit one to manipulate individual spins, their dynamics, and interactions, and could be of great importance for the development of quantum nanodevices.

Recent remarkable experiments of Manoharan *et al.* [3] have shown that the electronic structure of adatoms can be projected to a remote location exploiting quantum confinement of electronic states in corrals. Results of this experiment have been explained by Fiete *et al.* [4] using the scattering theory. They have demonstrated that the mirage at the empty focus of the elliptical corral is the result of resonant scattering of electrons from the magnetic adatom and scattering from the adatoms of the walls of the corral. There have also been several important theoretical studies related to quantum corrals and the mirage experiments [5–10].

Although the above-mentioned works have provided an appealing picture of quantum mirages and interactions in quantum corrals, a full understanding of the behavior of surface-state electrons in man-made nanostructures and their response to magnetic adatoms requires first-principles calculations. In this Letter we present a fully *ab initio* study of quantum mirages, the spin polarization, and the exchange interaction between magnetic adatoms in corrals. We concentrate on $3d$ adatoms in elliptical Cu and Co corrals on Cu(111). We demonstrate that the interaction of magnetic adatoms with the confined surface-state electrons of corrals leads to significant changes in electronic and magnetic states of corrals, and could produce a mirage at a remote location. We show that the spin polarization of surface-state electrons caused by magnetic adatoms placed in the corral focus is projected to an empty focus. Our study presents clear evidence that the long-range exchange interaction between magnetic adatoms is strongly affected by confined electronic states of corrals. The possibility of tailoring the exchange interaction by modifying the corral geometry is demonstrated. The spin polarization of the electron gas in the empty focus of the Co corral used in the experimental setup of Manoharan *et al.* [3] is revealed.

Adatoms and corrals destroy the two-dimensional (2D) periodicity of the ideal surface. Heller *et al.* [11] have shown in their studies of "quantum stadium" that the multiple-scattering approach is physically motivated to treat the electronic states of an arbitrary corral geometry and arbitrary placed adatoms in 2D systems. Therefore, we believe that an *ab initio* method based on the multiple-scattering theory is well suited for calculations of magnetic adatoms in quantum corrals. Our approach is based on the density functional theory (DFT) in the local spin density approximation and multiple-scattering approach using the Korringa-Kohn-Rostoker Green's function method for adatoms and clusters on surfaces [12]. Although the DFT does not account for properties of dynamical origin like the Kondo effect, it is an accurate method to determine static quantities [13]. Therefore, our calculations are related to electronic and magnetic properties of quantum corrals above the Kondo temperature.

We treat an ideal surface as an infinite 2D perturbation of bulk. Taking into account the 2D periodicity of the ideal surface, we calculate the structural Green's function by solving a Dyson equation self-consistently [12]. This function is then used as the reference Green's function in the Dyson equation for the self-consistent calculations of the Green's function of the corral (with or without adatoms) in a real space representation:

$$G^{nn'}_{LL'}(E) = \mathring{G}^{nn'}_{LL'}(E) + \sum_{n''L''} \mathring{G}^{nn''}_{LL''}(E)\Delta t^{n''}_{L''}(E) G^{n''n'}_{L''L'}(E), \quad (1)$$

where $G^{nn'}_{LL'}(E)$ is the energy-dependent structural Green's function matrix of the surface with the corral, and $\mathring{G}^{nn''}_{LL''}$ the corresponding matrix for the ideal surface; $\Delta t^n_L(E) = t^n_L(E) - \mathring{t}^n_L(E)$ describes the difference in the scattering properties at site n between the t matrices of the surface with the corral and the ideal surface [12]. The summation in (1) is over lattice sites and angular momenta for which the perturbation $\Delta t^n_L(E)$ is significant [14].

First, we consider the elliptical Cu corral on Cu(111) (Fig. 1). The quantum interference between the electron waves traveling towards the Cu atoms forming the corral wall and the backscattered ones leads to the confinement of the surface-state electrons inside the corral [15]. The energy-resolved local density of states (LDOS) at one of the corral's foci is presented in Fig. 1(a). It is seen that the LDOS exhibits a series of resonant peaks indicating the quantum confinement. The spatial distribution of the LDOS at the Fermi energy is presented in Fig. 1(b). The standing wave patterns outside and inside the corral are caused by the quantum interference of surface-state electrons scattered by atoms of the corral wall [16].

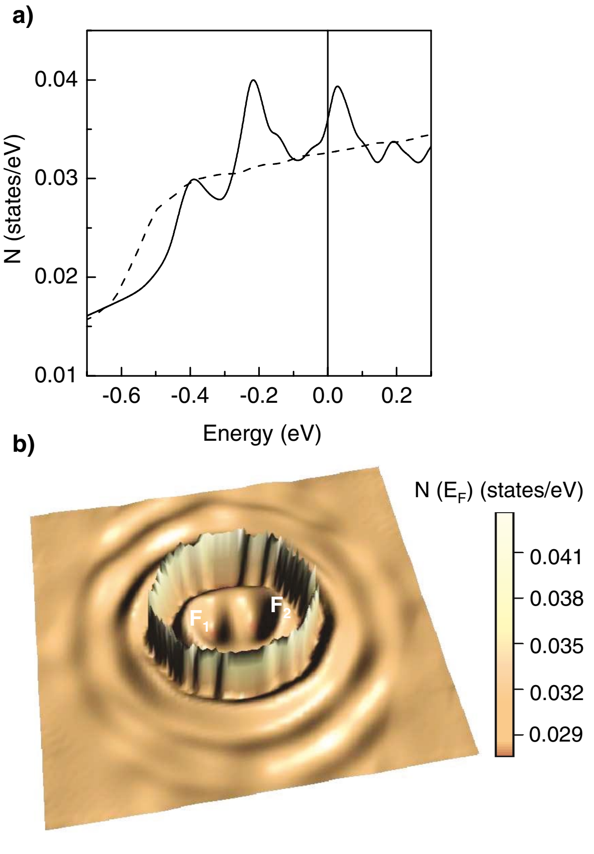

FIG. 1 (color). (a) The LDOS at the corral focus. (b) Quantum interference patterns inside and outside of the corral. The elliptical Cu corral with semiaxis $a = 25$ Å and eccentricity $\varepsilon = 0.5$ on Cu(111) is presented; the distance between the nearest Cu atoms in the corral walls is equal to the nearest neighbor separation on the Cu(111) surface. The LDOS of an open Cu(111) surface is shown by the dashed line.

If a magnetic adatom is placed at the focus of the corral, the resonance scattering of the surface-state electrons by the adatom and the corral walls leads to striking changes in the LDOS. As an example, we show in Fig. 2 our calculations for the Co adatom. Strong changes in the LDOS near E_{F} in the empty focus are resolved [see Fig. 2(a)]. Comparing this LDOS with the one for empty corral [Fig. 1(a)], it is evident that resonances act as waveguides for the projection of the electronic structure of the magnetic adatom to an empty focus [3]. The change in the LDOS (close to the E_{F}) at the empty focus clearly demonstrates the mirage effect [Fig. 2(b)]. To the best of our knowledge, the above result is the first fully *ab initio* confirmation of the projection of the electronic structure of the magnetic adatom to a remote location.

Another very important consequence of the quantum confinement in the corral concerns the spin polarization of the 2D electron gas. We place the magnetic Co adatom at the focus of the Cu corral and calculate the energy-resolved spin polarization at the empty focus. Results shown in Fig. 3(a) reveal a strong enhancement of the spin polarization in the empty focus of the corral compared

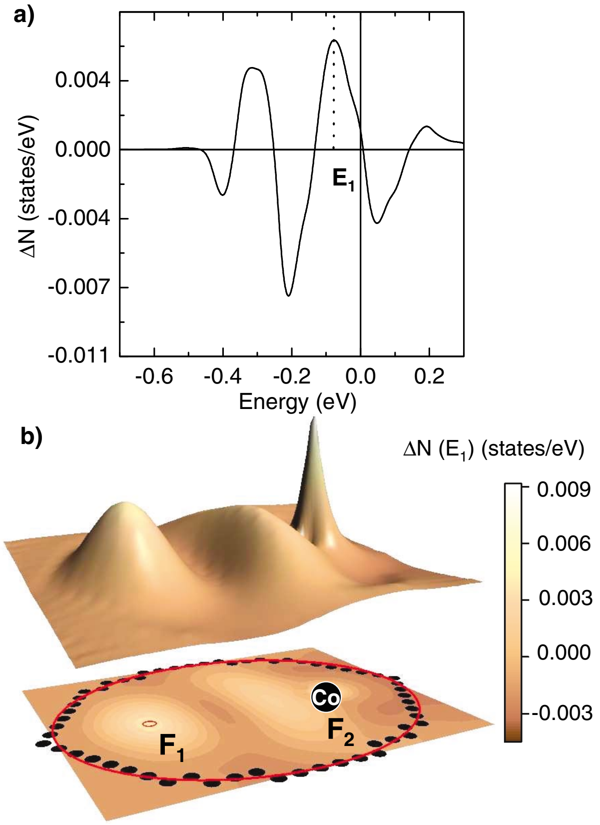

FIG. 2 (color). Mirage effect: Co adatom in the Cu corral. (a) The changes in the LDOS in the empty corral focus with respect to the Cu corral without the Co adatom are depicted. (b) Changes in the LDOS at the energy E_1 inside the corral; the LDOS of the single Co adatom on an open Cu(111) surface has been subtracted from the image.

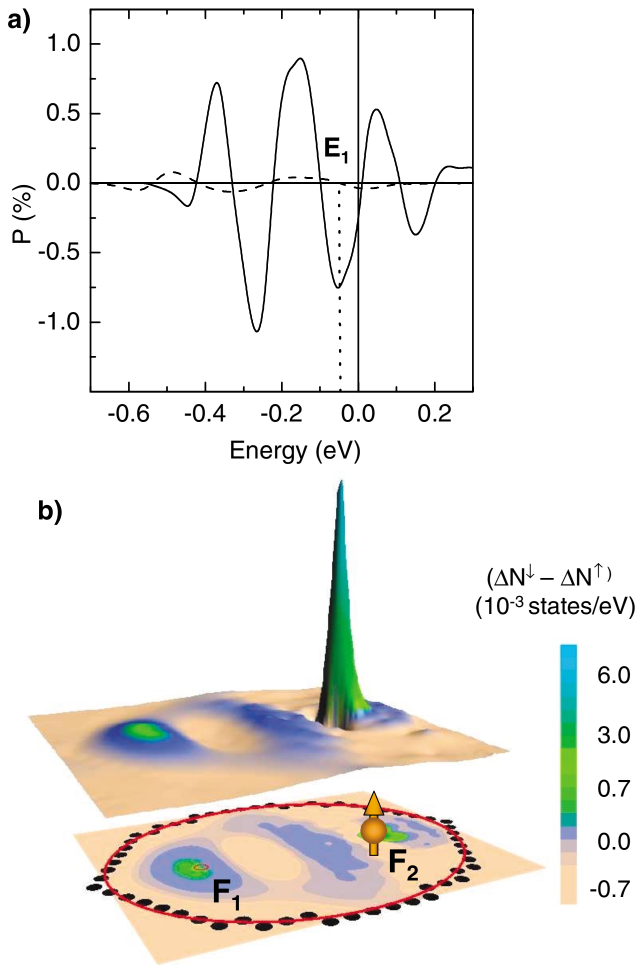

FIG. 3 (color). Enhancement of the spin polarization at the empty focus of the Cu corral. The magnetic Co adatom is placed at the right focus. (a) The spin polarization is determined as $(N^\uparrow - N^\downarrow)/(N^\uparrow + N^\downarrow)$, where $N^\uparrow$ and $N^\downarrow$ are the LDOS for majority and minority electrons, respectively. The spin polarization around the Co adatom on the open Cu(111) surface is shown by the dashed line. (b) $\Delta N^\downarrow$ and $\Delta N^\uparrow$ are determined by the difference between LDOS at the energy E_1 between the corral with the Co adatom and the single Co adatom on the open Cu(111).

to that around the Co adatom on an open Cu(111) surface. Our calculations clearly demonstrate that the spin polarization of surface-state electrons at the empty focus is very close to that near the magnetic adatom [Fig. 2(b)]. In other words, the spin polarization is projected to the second focus by the quantum states of the corral. We emphasize that the corral walls are nonmagnetic and, therefore, the spin polarization at the second focus is caused only by the spin dependent scattering of the surface-state electrons by the magnetic adatom. Our results unambiguously prove that tailoring the spin polarization of 2D electron gas could be achieved in artificial atomic structures by exploiting the quantum confinement of surface-state electrons.

To give clear evidence that quantum corrals can be used for controlled modification of magnetic interactions, we perform *ab initio* calculations for the exchange interaction between $3d$ adatoms inside the Cu corral. For large interatomic separations the exchange interaction energies are very small (meV and μeV). Therefore, there is the problem of subtracting huge total energy values to obtain the resulting small interaction energies. However, it has been proved that applying the force theorem [16,17] and using the single-particle energies, instead of total energies, one can resolve very small interaction energies at large atomic distances with high accuracy.

We have calculated the exchange interaction between $3d$ adatoms in the Cu corral on Cu(111) for different adatom-adatom separations. In the absence of the corral, i.e., for an open surface, the exchange interaction between magnetic adatoms at large distances is dominated by the surface-state electrons. However, the quantum corral drastically influences the interaction between magnetic adatoms. This is well seen in Fig. 4 where our calculations for $3d$ adatoms placed in the corral foci are presented. In order to demonstrate the effect of the corral geometry on the exchange interaction, we show our calculations for the Cu corrals of different eccentricities. These striking results reveal that the exchange interaction in the corral is strongly enhanced compared to an open surface, and can switch from the ferromagnetic coupling to the antiferromagnetic one by modifying the corral geometry. We believe that these calculations provide the clearest evidence of tailoring the magnetic interactions between adatoms at large distances by constructing appropriate corrals.

Finally, we apply our method for calculations of the spin polarization in the Co corral used in the experimental setup of Manoharan *et al.* [3]. A net spin polarization of the

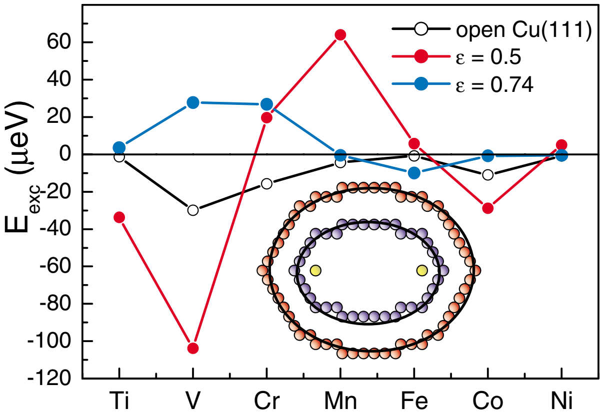

FIG. 4 (color). The exchange interaction between magnetic adatoms inside the Cu corrals of different eccentricities; the distance between foci is fixed. The exchange interaction on an open Cu(111) is presented by the black line. Negative energies mean that the spins of both adatoms are ferromagnetically coupled, while positive energies correspond to an antiferromagnetic coupling.

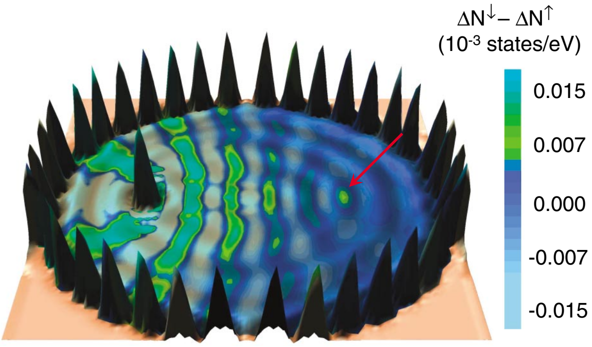

FIG. 5 (color). The LDOS at the Fermi energy on the Co adatom and the Co atoms of the corral walls are shown. The spin polarization of surface-state electrons inside the Co corral is presented in color: $\Delta N^{\downarrow}$ and $\Delta N^{\uparrow}$ are determined by the difference between LDOS near the Fermi energy (+10 meV) of the Co corral with the Co adatom, the empty Co corral, and the single Co adatom on the open Cu(111). The mirage in the empty focus is marked by the red arrow. The geometrical parameters of the corral are the same as in the experimental setup of Ref. [3], i.e., semiaxis $a = 71.3$ Å and eccentricity $\varepsilon = 0.5$.

electron gas in the Co corral was suggested in this work as the possible reason for the quantum mirage in an empty focus. First, we have found the LDOS for the minority and majority electrons inside the Co corral with the Co adatom placed in the corral focus. Calculations have been performed for energies close to the Fermi energy (E_F + 10 meV). Then, we have repeated the calculations for an empty Co corral. In fact, the difference between the two calculations presents the effect of the Co adatom on the spin polarization of the electron gas inside the corral. However, the spin polarization in the empty focus is found to be significantly smaller than the spin polarization on the Co adatom. Therefore, to make a clear presentation of the magnetic mirage, i.e., enhanced spin polarization at the empty focus, the spin polarization of the single Co adatom on an open Cu(111) surface has been removed from the image shown in Fig. 5. The oscillations of the spin polarization are well seen in Fig. 5. The enhancement of the spin polarization in an empty focus is revealed.

In summary, we have presented the first *ab initio* studies of quantum mirages and the magnetic interactions in quantum corrals. While we have used particular systems, Cu and Co corrals on Cu(111) to illustrate several effects of the quantum confinement of surface-state electrons, the main conclusions of our work are independent of the specific systems. It is generally true that the spin polarization of surface electrons caused by magnetic adatoms can be projected to a remote location by quantum states of corrals, and the exchange interaction between magnetic atoms can be manipulated at large distances.

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[14] Multiple scattering of surface-state electrons inside the corral (including the interaction with bulk states) is taken into account in our approach. Atomic relaxations of atoms of the corral walls and adatoms inside the corral practically do not influence the main results of our work. Our studies have proved that only a limited number of atoms near the corral wall and near the adatoms placed inside the corral contribute to $\Delta t_L^n(E)$ in Eq. (1). Therefore, even very large corrals can be calculated fully *ab initio* using our approach. Self-consistent and non-self-consistent calculations give essentially the same results, provided that the self-consistent potentials of single adatoms on Cu(111) are used in the non-self-consistent calculations.
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