Controlling the Spin Polarization of Nanostructures on Magnetic Substrates

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It is shown that, by utilizing spin-selective quantum interference, the spin polarization of nanostructures deposited on a magnetic substrate with a surface state can be strongly modulated locally and energetically by an appropriate structural design. This finding is deduced from state-of-the-art *ab initio* calculations and interpreted within an analytical model. We present results for hexagonal Cu corrals and mesoscopic triangular Co islands on Co-covered Cu(111). These systems are experimentally feasible, and the effect should be detectable with current technology.

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When magnetic thin films and/or nanoclusters are deposited on a nonmagnetic substrate charge transfer, multiple electronic scattering and interference phenomena generally govern the electronic and the magnetic properties of the combined system. For example, surface states of nonmagnetic noble metal substrates, such as Cu(111), turn spin polarized and attain a different character when Co nanoislands are added to the substrate surface [1,2]. Such spin-polarized surface states (SP-SS) can serve as spin-transport channels across the vacuum barrier to or from another magnetic material, an issue which is of central importance for spintronic applications based on spin-polarized tunneling [3]. Indeed, experiments by Wiesendanger *et al.* [4] indicate that the vacuum tunneling magnetoresistance (TMR) is maximized by tunneling to a SP-SS. Further evidence of the prominent role of SP-SS has come from recent spin-polarized scanning tunneling microscopy studies of Rusponi *et al.* [5] on Co double-layer islands on Pt(111). Hence, it is highly desirable to envisage new general schemes and realistic techniques for controlling the character of SP-SS as well as the spin-polarization of deposited nanoparticles. This is particularly so as several experimental and theoretical studies have also evidenced the existence of SP-SS on transition metal substrates [6–9].

In this Letter, we point out a way for the modulation and control of SP-SS based on an appropriate design of the nanostructure geometry, such as the size of hexagonal corrals, and islands. In particular, we show, using *ab initio* calculations supported and interpreted within an analytical model, that the spin polarization at the Fermi level of two Co monolayers (ML) on Cu(111) can be modified spatially and energetically in a controlled manner by building on top of the sample Cu corrals or triangular Co islands with a specified size. This phenomena, which rests on spin-dependent quantum interference and, hence, is of a general validity, can be, for instance, utilized to control the TMR. Our goal is the spatial and energetic tuning of the spin polarization of Co thin films and nanoislands on Cu(111) by means of quantum confinement. While the possibility of the modulation of the spin-split local density of states (LDOS) may be expected from general arguments based on interferences and size quantization in confined systems [10], in a realistic situation size-quantized levels may be broadened or even washed out because the *spatially confined* SP-SS contains, in general, a range of wave vector components that may couple strongly to bulk states, leading thus to an enhanced decay rate. In addition, such states ''leak'' generally outside the confinement due to the discreteness and the finite strength of the confining potential. Hence, it is essential from both a conceptual and an applied point of view to perform accurate first principle calculations under realistic conditions to explore the role of the size quantization and its potential to be utilized in future applications, a task which is tackled here for the first time.

Our *ab initio* approach is based on the density functional theory and the Korringa-Kohn-Rostoker Green's function (GF) method for low-dimensional systems [11]. We perform self-consistent calculations of GF of two Co monolayers on Cu(111) surface. Adatoms, islands, or other artificial structures on the surface destroy the 2D periodicity of the ideal substrate. Therefore, the GF is calculated in a real space formulation. The structural GF of the ideal 2D substrate is used as a reference GF to calculate the GF of the perturbed system by means of the Dyson equation (technical details are in Refs. [11–14]).

We concentrate here on systems consisting of 2 ML Co on Cu(111) and Co triangular islands on Cu(111), both of which have been experimentally realized [1,2,15]. Scanning tunneling spectroscopy (STS) experiments and *ab initio* calculations evidence the existence of a pronounced peak at about 0.3 eV below the Fermi level [1]. The minority d states (referred to as $\downarrow$) and some small minority s-p contribution give rise to this peak. In addition, a mostly unoccupied free-electron-like surface state of s-p majority character (referred to as $\uparrow$) exists [1]. This state plays a vital role for the quantum interference pattern that is observed on Co nanoislands.

First, we demonstrate that the quantum confinement of the surface-state electrons in hexagonal corrals constructed on 2 ML Co/Cu(111) can generate new spin-polarized states near the Fermi level. We calculate the LDOS inside

the Cu corrals of different sizes. As an example, we depict in Fig. 1 the spatial distribution of the majority and the minority LDOS of surface-state electrons at the Fermi level inside the Cu corral. These results endorse the spin selectivity of the quantum confinement of surface electrons. The mainly energetically localized minority electrons are hardly influenced by the corrals. The standing waves are exclusively observed for the majority-surface states [16]. Hence, in contrast to LDOS(↓) we expect a corral-size-dependent modulation of LDOS(↑) due to the formation of standing waves upon coherent multiple scattering [17].

In Fig. 2, we show the energy resolved LDOS of the surface electrons in the Cu corrals on 2 ML Co/Cu(111). In all cases, the minority LDOS show strong features below the Fermi level. This localized peak is mainly determined by d states and its energetic position is hardly affected by the presence of corrals. The peak in the minority LDOS above the Fermi level at the energy of about 0.5 eV is found to occur due to the s-d hybridization with the Co monolayers. Our analysis shows that this peak is also well localized. In contrast, the free-electron-like nature of the majority-surface-state electrons on Co monolayers acts as follows: In the LDOS for unoccupied majority states interference-induced sharp peaks emerge close to the Fermi level. Increasing the corral size, the unoccupied majority electronic states move to lower energies and can cross the Fermi level.

For an understanding of the general features and trends of the energy and size-dependent changes in LDOS(↑), we utilize a simple model based on the assumption that the majority electrons are confined by the circular potential $V(r) = 0$ for $r < R_0$ and $V(r) = \infty$ for $r = R_0$. Here R_0 is the closest distance from the Cu adatoms to the center of the hexagons. The motivation of this model relies on the structure of LDOS(↑) shown in Fig. 1: LDOS(↑) significantly decreases at the corral boundary, indicating thus a substantial height of $V(r = R_0)$ on the scale of E_F.

The single particle wave functions associated with the potential $V(r)$ are then $\psi_{\uparrow,nm}(\bar{r}, \phi) = N_{nm} J_m(k_n(m)\bar{r}) e^{im\phi}$, where J_m is the Bessel function and the two quantum numbers m and n quantify the angular (ϕ) and the radial motion, respectively. $\bar{r}$ is the distance measured in units of R_0. $k_n(m)$ is the n node of J_m. The single particle energies in units of $\hbar^2/(2m^* R_0^2)$ are then $E_{nm} = k_n^2(m)$, where for higher order nodes the relation applies $k_n(m) \approx n\pi + (m - 1/2)\pi/2$. The normalization constant is given by $N_{nm} = 2/\sqrt{2\pi}[J_{m+1}(k_n(m))]^2$. The spatial structure of LDOS(↑) at the energy E_{nm} is determined by $|\psi_{\uparrow,nm}(\bar{r}, \phi)|^2$. For illustration, let us focus at the center of the corrals. All $|\psi_{\uparrow,nm=0}|^2$ are peaked at $\bar{r} = 0$, whereas the centrifugal potential removes the majority-state electron density from $\bar{r} = 0$ for $m \neq 0$. This results in an enhancement or depression of LDOS(↑) at the respective corral state energies, a fact which is nicely demonstrated in Fig. 2 for the region around E_F: For small corrals ($R_0 < 16.6$ Å), the lowest energy $E_{n=1m=0}$ is above E_F, and the corral acts in effect as a single scattering center (composite adatom) for the

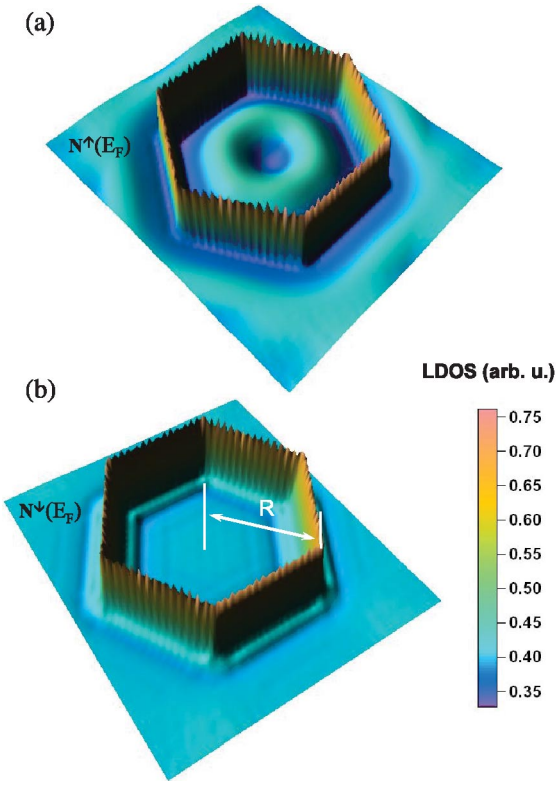

FIG. 1 (color). Spin-polarized standing waves: the LDOS at the Fermi level for (a) the majority and (b) the minority surface-state electrons in hexagonal corral on Cu atoms deposited on 2 ML Co/Cu(111). The corral extension is quantified by $R =$ 35.8 Å, as indicated in the figure.

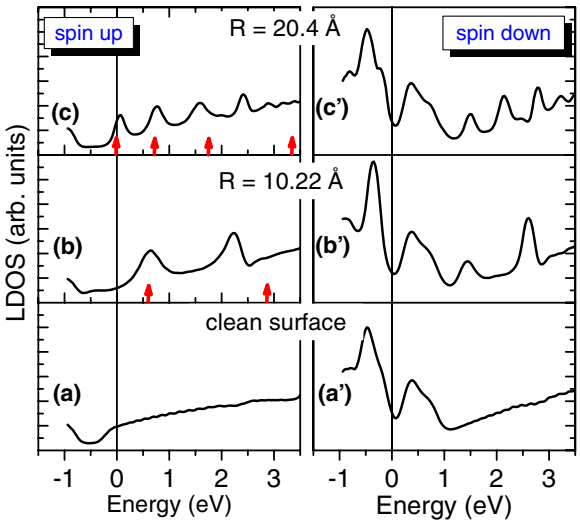

FIG. 2 (color online). LDOS for (b),(c) the majority and (b′),(c′) the minority surface-state electrons at the center of hexagonal Cu corrals; (a),(a′) show LDOS of the surface-state electrons on an open 2 ML Co/Cu(111). Predictions of the analytical model for the peak positions in LDOS(spin-up) are marked by arrows.

surrounding electrons. Inside the small corrals, LDOS(↑) is depleted, and, hence, the spin polarization which we define as $P = [\mathrm{LDOS}(\uparrow) - \mathrm{LDOS}(\downarrow)]/[\mathrm{LDOS}(\uparrow) + \mathrm{LDOS}(\downarrow)]$ tends to -1 within the corrals [note that LDOS(↓) is not affected by the confinement as evidenced by Fig. 2, i.e., at E_F, LDOS(↓) $\approx$ const $\forall$ R_0]. This behavior is confirmed by our *ab initio* calculations in Fig. 3 showing the spin polarization at the center of corrals. When $E_{nm=0}$ crosses E_F as R_0 varies [which occurs at the radii $R_{0n} = 7k_n(m = 0)$ Å, $n = 1, 2, \ldots$], LDOS(↑) is enhanced at $\bar{r} = 0$, and, hence, the spin polarization changes to positive (these particular R_0 values are marked by arrows in Fig. 3). The relative heights of the peaks in LDOS(↑) ($\bar{r} = 0$) for varying R_0 but for $E_{nm=0} = E_F$ are determined by $\mathcal{R} = |N_{nm=0}/N_{n+1m=0}| = |[J_1(k_n(0))]/[J_1(k_{n+1}(0))]|$. Hence, we observe in Fig. 3 that the peak in P at $R_0 = 44$ Å is lower than at $R_0 = 19.2$ Å. In between the peaks, LDOS(↑) is either depleted in the whole corral or removed from $\bar{r} = 0$ by the centrifugal force, and, hence, P drops below its value for a clean substrate, as observed in Fig. 3. Our model predicts that for $E > E_F$ the otherwise flat LDOS(↑) at $\bar{r} = 0$ is modulated due to the confinement by discrete (delta-type) peaks (indicated by arrows in Fig. 2) occurring at $E_{nm=0}$ and depressions otherwise. The relative height ratios of these peaks is governed by $\mathcal{R}$. In reality, the peaks stand for resonances that decay due to energy-dependent coupling to bulk states and to states outside the corral. Since the peaks are nicely separated in energy, their lifetimes might be measurable by means of STS [18]. The fact that the simple model describes to a certain extent the positions of the low-energy peaks in Fig. 2 (see also Fig. 3) is an indicator of the high potential barrier at $r = R_0$ (on the scale of E_F). In Fig. 2, we observe that the width of the size-quantized peaks is larger for smaller corrals, which can be traced back to the fact that a spatially more localized state contains a larger range of momentum components that may couple to bulk states and, hence, leads to a larger decay rate.

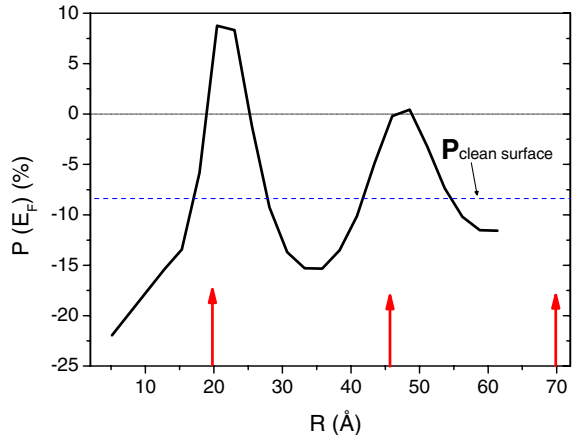

FIG. 3 (color online). Oscillations of the spin polarization of the surface-state electrons at the Fermi level and at the center of the hexagonal corrals. The spin polarization of surface electrons on the open 2 ML Co/Cu(111) surface is shown by a dashed line. The arrows mark the predictions of the analytical model for the maximal spin polarization as a function of R.

Full view on the spatial distribution of the spin polarization P at E_F calculated by our *ab initio* method is shown in Fig. 4. The size of corrals is found to significantly affect the quantum interference patterns. Since LDOS(↓) at E_F does not depend on R, we can assign the structures in P to equivalent structures in LDOS(↑) at E_F, which is given by $|\psi_{\uparrow,nm}(\bar{r}, \phi)|^2$. For example, for Fig. 4(a), E_F lies between the levels $E_{n=1m=0}$ and $E_{n=2m=0}$. Hence, we expect a negative P at $\bar{r} = 0$ and a less structured radial distribution. In Fig. 4(b), E_F almost coincides with $E_{n=2m=0}$, and, hence, we observe two circles of P where $|\psi_{\uparrow,n=2m=0}(\bar{r}, \phi)|^2$ is maximal. In between, $|\psi_{\uparrow,n=2m=0}(\bar{r}, \phi)|^2$ has a node and, hence, the negative polarization. The structures of Fig. 4(c) are readily explained along the same line by noting that in this case E_F lies in between $E_{n=2,m=0}$ and $E_{n=3,m=0}$. The modulation of the angular distributions of P cannot be explained within our simple model. However, they are readily understood in terms of the appropriate symmetry and the associated quantum numbers which are the irreducible representa-

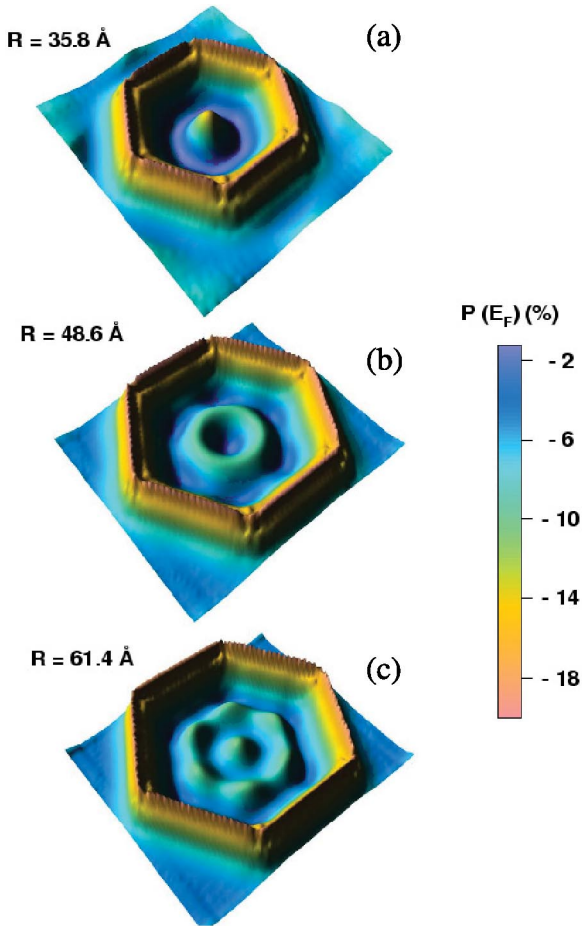

FIG. 4 (color). The spatial distribution of the spin polarization of surface-state electrons in the corrals.

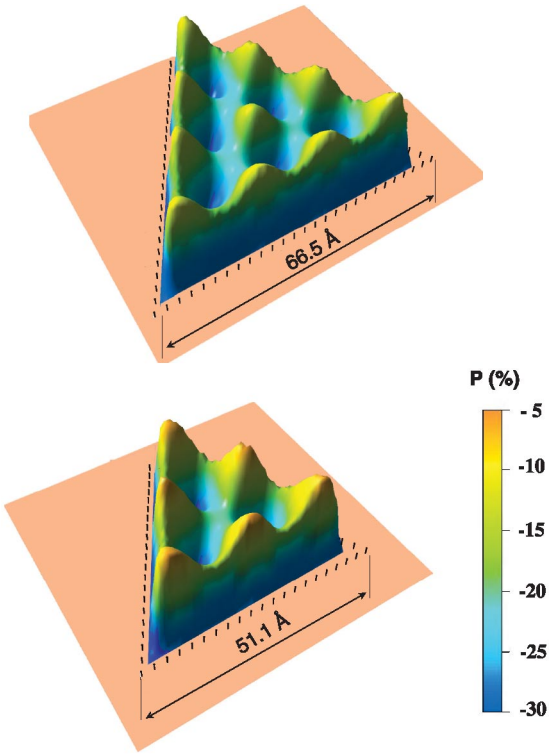

FIG. 5 (color). The spin polarization of surface-state electrons on triangular Co islands on Cu(111); calculations are performed for $E = 0.5$ eV above the Fermi level [1].

tions of the C_6 group of the hexagon [19]. The discreteness of the hexagons plays obviously no roles for the interference-induced pattern because λ_F is larger than the interatomic distance of the Cu-Cu adatoms.

In view of an experimental verification, we consider a recently realized system [1,2,15]. At lower coverage, Co grows on Cu(111) as compact islands of triangular shape. Our *ab initio* calculations for equilateral triangular islands (Fig. 5) evidence that the size of the island has a pronounced influence on the spatial distribution of the spin polarization of the Co islands. This dependence is rendered comprehensible upon assuming the majority-state electrons only to be subject to the lateral confining potential present at the boundary of the island, i.e., at the Co-vacuum interface, where LDOS(↑) dies out. The energy spectrum, in units of $\hbar^2/(2m^*l^2)$, where l is the edge length, is then given by $E_{qp} = 16\pi^2(p^2 + q^2 - qp)/9$, $1 \leq q \leq p/2$, $p, q \in \mathbb{N}^+$ [20]. Hence, the smallest l where we expect to see size enhancement of P at, e.g., $E = 0.5$ eV, is $l = 25.2$ Å, in which case P is positively increased at the center of the triangle [where LDOS(↑) is peaked] but otherwise is rotationally structureless, except for the triangular edges where LDOS(↑) diminishes, and, hence, P acquires a large negative value. A desired topography of P on the island is achieved by varying l or the shape of the triangle, as demonstrated in Fig. 5. The structures observed in this figure are readily understood from the structure of LDOS(↑) of the model system at the respective energy [while assuming LDOS(↓) to be unaffected by the quantum confinement]. The above results unambiguously prove that the size of islands drastically affects the spin polarization of surface electrons. Additionally, our calculations reveal a strongly inhomogeneous distribution of the spin polarization on islands.

In summary, using a reliable state-of-the-art *ab initio* method, we demonstrated for realistic systems the size-dependent tuning of the spin polarization of nanostructures on a magnetic substrate. The results are interpreted and analyzed within an appropriate analytical model. The present findings point out the possibility of modulating the spin polarization locally and energetically by engineering the shape and the size of the surface-deposited nanostructures.

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