

## Large spin effects in Coulomb blockade of Fe/MgO/Fe tunnel junctions

W. Wulfhekel,\* A. Ernst, J. Henk, P. Bruno, and J. Kirschner  
 Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

F. Zavaliche  
 Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany  
 and Materials Research Science and Engineering Center, University of Maryland, Building 090 College Park, Maryland 20742, USA

C. C. Kuo  
 Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany  
 and Department of Physics, National Sun Yat-Sen University, Kaohsiung 804, Taiwan

Minn-Tsong Lin  
 Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany  
 and Department of Physics, National Taiwan University, Taipei 106, Taiwan  
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The Coulomb blockade across Fe and Pd nanoislands grown on single-crystalline MgO insulator films on Fe(001) was investigated with scanning tunneling microscopy. In  $I(U)$  spectra of individual islands, the characteristic steps of the Coulomb staircase were observed as a function of island size. A detailed analysis of the Coulomb staircase energetics in terms of the island capacitance reveals the expected linear behavior for electrostatic interactions. For ferromagnetic Fe islands, however, a significant offset of 1 eV was found, which is missing in the case of nonmagnetic Pd islands. This effect is explained by the spin dependence of the electronic transport across the MgO barrier in combination with a quasihalf-metallic density of states of the Fe islands, as corroborated by first-principles electronic-structure calculations.

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With decreasing size of electronic devices, classical concepts of electronic transport become questionable because of quantum-size effects. In particular, in electronic devices of nanometer size, significant effects due to single electrons can show up, e.g., charging effects which govern the electrical properties of a device.<sup>1</sup> A paradigm is single-electron transport: The transfer of single electrons between two leads of a single-electron transistor was shown by Geerligs *et al.*, allowing single electrons to hop via a small conductive island.<sup>2</sup> Although this phenomenon was already observed in granular metallic materials more than three decades ago,<sup>1,3,4</sup> the use of modern lithographic processes, needed for the controlled fabrication of devices, allowed to study the transport of single electrons in full detail.<sup>5-7</sup>

In nanostructured double tunneling junctions, transport is governed by Coulomb blockade (CB). A nanostructured tunnel junction consists of small islands or clusters situated between two leads and separated from the latter by tunnel barriers. Applying a bias voltage  $U$  between the leads, the island can be charged by single electrons. Regarding the island as a capacitor with capacitance  $C$ , an increase from  $n$  to  $n+1$  electrons on the island results in an additional potential of  $e/C$  with  $e$  denoting the electron's charge. The transfer of an electron through the island is inhibited when the potential of the charged island is larger than the applied bias voltage. Hence, in the current-voltage characteristic  $I(U)$  of a tunnel junction, kinks appear at  $U_g(n) = (2n+1)e/(2C)$ . In particular for  $n=0$  (equilibrium charge), the current is blocked for  $-e/(2C) \leq U \leq +e/(2C)$ . A fractional influence charge  $Q_0$  can shift the positions of the Coulomb staircase but does not influence the step spacing.<sup>8</sup>

The Coulomb staircase was observed by many research groups and is well understood. A straightforward way to study this effect is using scanning tunneling microscopy (STM) of metallic islands grown on a thin insulating film which itself is deposited on a metal substrate.<sup>9</sup> In this configuration, the metallic substrate and the metallic tip of the STM are the two leads, and the metallic island is the nanoparticle, being electrically decoupled from the substrate and the STM tip by the insulator film and by the vacuum, respectively. The striking advantage of STM, as compared to other local probes of the CB, is to combine imaging with controlled recording of the  $I(U)$  characteristics of individual nanoscale islands.<sup>9</sup> With regard to spin-dependent transport, a question arises on which effects might show up upon replacing a nonmagnetic island by a magnetic one. For an ensemble of magnetic clusters, the Coulomb staircase is difficult to observe in detail because of the size distribution and of randomly oriented magnetic moments. Desmicht and co-workers designed a point-contact electrode in order to probe the CB in individual Co clusters prepared by electron beam lithography.<sup>10</sup> In contrast to this approach, STM provides precise information on the size of each cluster, and a set of clusters can be studied in one experiment.

In this paper, we focus on the Coulomb staircase in small Fe islands and illuminate the strong influence of the electron's spin on the transport properties. *A priori*, the role of the spin is not obvious and may alter the overall characteristics of the device.<sup>11,12</sup> We report on a spin-related effect of order of 1 eV originating from a combination of a quasihalf-metallic behavior of the Fe islands and a partial blocking of one spin channel. To elucidate spin-related effects, the Cou-

lomb staircase in the single-crystalline model system Fe/MgO/Fe was investigated and compared with that of nonmagnetic Pd islands on MgO/Fe.

The CB experiments were carried out on crystalline Fe islands grown on MgO insulator films, the latter deposited on Fe(001) surfaces. In a first step, flat and clean Fe surfaces were prepared in ultra-high vacuum.<sup>13</sup> Subsequently, MgO films between 3 monolayers (ML) and 5 ML thick were deposited by molecular beam epitaxy (MBE), resulting in flat and single-crystalline insulator films (for details of the growth procedure, see Ref. 13). Because the surface free energy of Fe is larger than that of MgO, Fe does not wet the MgO film and deposition of Fe by MBE leads to the formation of nanometer-sized Fe clusters.<sup>13</sup> The average lateral size of these clusters can be controlled by deposition temperature and Fe coverage.

After preparation, the sample was cooled down to 25 K and subsequently imaged *in situ* with STM using W tips. With the bias voltage  $U$  chosen large enough, electrons can tunnel from the tip into the conduction band of the MgO film<sup>13</sup> and the MgO film as well as the metallic Fe islands are imaged [Fig. 1(a)]. Due to the half-integer ML coverage, the surfaces of both the third and fourth atomic layers of MgO are visible [black and dark grey in Fig. 1(a)]. On top of the MgO film, Fe islands appear as small protrusions (white). Their height ranges between 1 and 3 ML (0.14 and 0.43 nm) and their lateral dimensions between 0.5 and 2 nm. These Fe islands are used as magnetic nanostructures for CB in the present STM experiment.

To obtain the  $I(U)$  characteristics of the islands, the feedback loop of the STM was opened in each pixel of the image and the bias voltage  $U$  was ramped. Simultaneously, the tunnel current  $I$  was recorded as a function of  $U$ . An  $I(U)$  curve recorded with the STM tip on top of the bare MgO film is depicted in Fig. 1(d) (dotted line). The current is very small unless  $U$  exceeds the band gap of MgO, indicating its good insulating quality.<sup>13,14</sup> With the STM tip on top of an Fe island, however, the  $I(U)$  curve changes significantly [solid line Fig. 1(d)]. For negative bias voltage the tunneling current on top of an island is larger since the first tunneling from the substrate to the island is proportionally enhanced by the island area. In particular, the  $I(U)$  curves show characteristic kinks, i.e., the signature of the Coulomb staircase (indicated by arrows). The electrostatic potential  $U_g$  upon charging an Fe island with a single electron can be directly obtained from the  $I(U)$  curve. It corresponds to the separation of Coulomb stairs in the current [ $U_g = (2.6 \pm 0.1)$  eV for the island indicated in Fig. 1(b)]. We emphasize that the bias dependence of other Fe islands shows similar effects, especially Coulomb stairs for larger  $n$  [see inset of Fig. 1(d)]. As an alternative to local  $I(U)$  curves, the current at  $U = -3.8$  V was plotted *versus* the position of the STM tip. Here, the metallic Fe islands show up as black in Fig. 1(c), as they display a negative tunnel current while the MgO insulator shows a vanishing current. These local current images were used to determine the lateral extension of the Fe islands with higher precision than by topographic images [Fig. 1(a)]. For example, the diameter of the island in the center of Figs. 1(b) and 1(c) was measured to be  $(1.5 \pm 0.2)$  nm. Hence, STM allows us to ef-

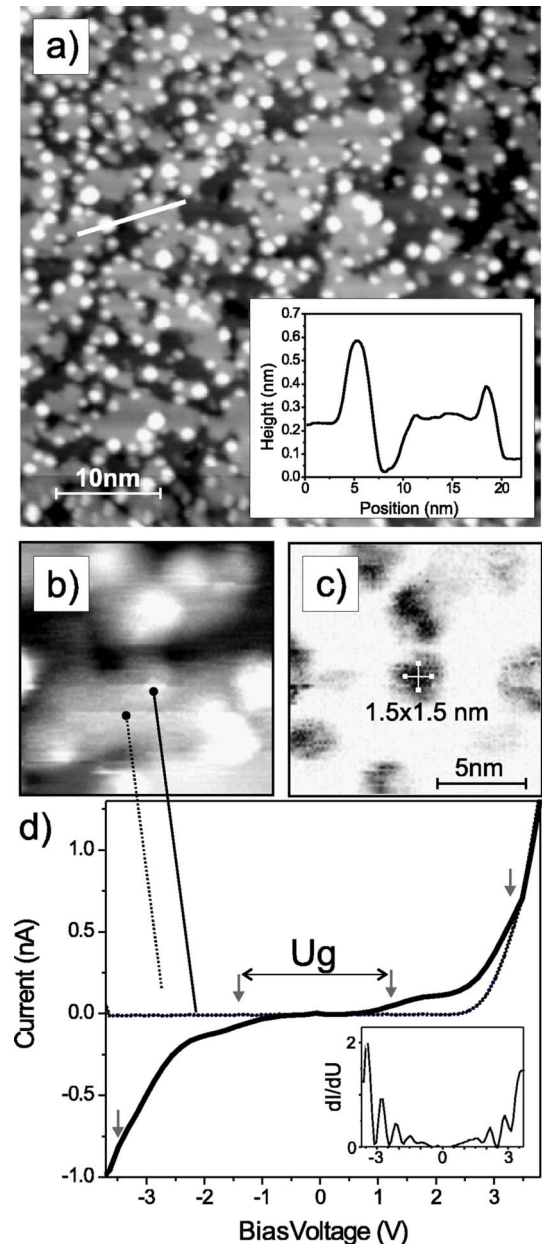


FIG. 1. (Color online) (a) Topography of Fe islands ( $\approx 0.1$  ML Fe) on a 3.5 ML MgO film on Fe(001) taken with  $I = 1$  nA and  $U = 3.8$  V. The inset shows a line profile. (b) Magnified image of several Fe islands. (c) Map of the local tunnel current at  $-3.8$  V. (d)  $I(U)$  characteristics measured with the STM tip on top of the bare MgO insulator (dotted) and an Fe island on MgO (solid). The inset shows  $dI(U)/dU$  curves of a larger island.

ficiently determine the gap  $U_g$  and size of Fe islands. A similar investigation was performed for nonmagnetic Pd islands instead of magnetic Fe islands.

The total capacitance of an Fe island is given by the sum of the substrate island and the island-tip capacitance. For the STM parameters chosen for tip stabilization, the width of the island-tip tunnel barrier can be estimated to about 0.8 nm.<sup>15</sup> This width is similar to the thickness of the MgO films, ranging between 0.6 and 1 nm. In contrast to the vacuum tunnel barrier, the MgO tunnel barrier shows an induced po-

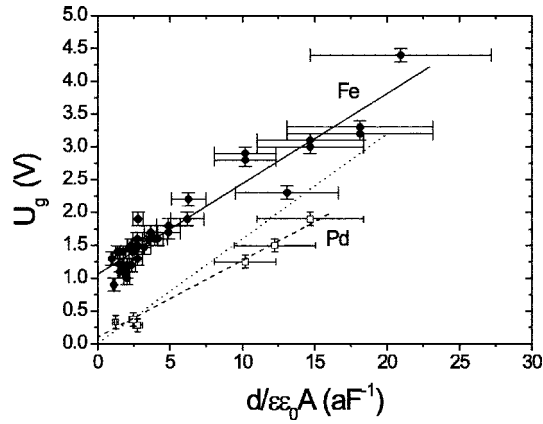


FIG. 2. Observed gap  $U_g$  for Fe and Pd islands on MgO films on Fe(001) versus the inverse estimated island capacitance and linear fits. For comparison, the estimated relation  $(ed)/(\epsilon_0 A) \approx e/C$  is shown as a dotted line.

larization due to the dielectric constant of MgO ( $\epsilon_{\text{MgO}}=9.6$ ). This has two consequences. First, it leads to a minute voltage drop between substrate and island if the tip is biased. In turn, the major potential drop occurs across the vacuum barrier (about 90% of the total drop). Second, the total island capacitance is dominated by the substrate-island capacitance, being ten times larger than the island-tip capacitance. Therefore, we restrict ourselves in the following discussion to the voltage drop across the vacuum barrier and assume that the total island capacitance is given by the substrate-island capacitance. As Hanna and Tinkham have shown, such large asymmetry in the capacitances leads to a Coulomb gap around zero bias, which is independent of  $Q_0$ .<sup>8</sup>

Having determined the area  $A$  of an island, its capacitance  $C$  can be approximated by that of a plate capacitor  $C = \epsilon_{\text{MgO}} \epsilon_0 A/d$ , where  $\epsilon_0$  and  $d$  are the electrical permittivity constant and the MgO spacer thickness, respectively. Since the electrostatic energy  $E_C$  is proportional to  $e^2/C$ , plotting the experimentally observed  $U_g = E_C/e$  versus  $1/C$  should result in a straight line through the origin with slope  $e$ .

The experimental gap  $U_g$  is displayed as a function of the inverse estimated island capacitance for Fe and Pd islands in Fig. 2. The Coulomb gap of nonmagnetic Pd islands shows the expected behavior: A linear fit of  $U_g$  versus  $1/C$  (dashed line) goes within the error margin through the origin and its slope is close to the expected one (dotted line). In fact, the slope is slightly less, which might be explained by the assumptions, i.e., neglect of the island-tip capacitance, neglect of edge effects in the substrate-island capacitance, or due to a slightly different dielectric constant of a MgO film with respect to that of MgO bulk.

Within the error bars, the slope for the Fe islands (solid line in Fig. 2) is identical to that of the Pd islands. This finding provides evidence that the MgO spacer decouples electronically both the Fe and the Pd islands from the Fe substrate.<sup>16,17</sup> The most striking difference with respect to the Pd result, however, is that the linear fit does not hit the origin, but shows a considerable offset of about 1 V. Note that data for small  $1/C$  in Fig. 2 are associated with large islands, whose relative size error is small. This proves that the offset

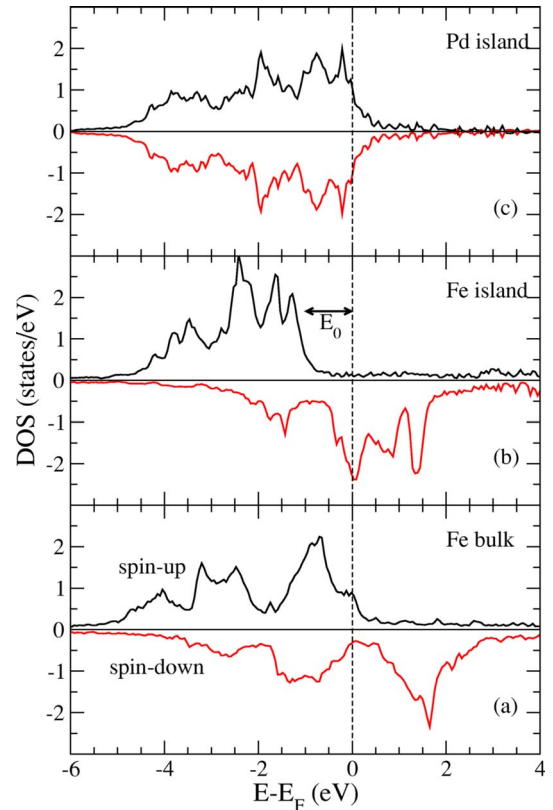


FIG. 3. (Color online) Density of states of Fe and Pd films on MgO/Fe(001). (a) Spin-resolved density of states (DOS) of Fe bulk. (b) and (c) As (a), but for the surface layers of 2 ML films of Fe and Pd deposited on 4 ML Mg/Fe(001), respectively. The energy shift  $E_0=1$  eV discussed in the text is indicated by a horizontal arrow in (b).

is not due to errors in the determination of the island sizes.

Summarizing at this point, we find a striking difference in the Coulomb blockade of magnetic Fe islands compared to that of nonmagnetic Pd islands, namely, an offset of the gap plotted versus inverse capacitance for Fe islands (Fig. 2). Since the slopes of the  $U_g(1/C)$  curve of Fe and Pd islands are identical, this unexpected behavior of the Fe islands suggests an explanation in terms of spin-resolved electronic properties.

In the following, we restrict ourselves without loss of generality to the case of islands charged by a single additional electron ( $n=0 \rightarrow n=1$ ). Without CB, a bias voltage  $U > 0$  opens an “energy window” for tunneling in which occupied electronic states in the source lead tunnel into unoccupied ones in the drain lead. In the drain lead, this energy window extends from the Fermi level  $E_F$  to  $E_F + eU$ . The current  $I$  from the source to the drain lead is given by  $I = GU$ ,  $G$  being the conductance of the device. In the CB regime, the tunneling electrons have to overcome the electrostatic energy  $E_C$  of the island. Hence, the energy window becomes effectively reduced in size, namely,  $I = G(U - U_g)$  for  $U > U_g$ . For  $U \leq U_g$ ,  $I$  vanishes due to the CB.

The findings for the Fe islands can be modeled by an additional term  $U_0 = 1$  V. Assuming that  $U_0$  is independent of the island size, the gap of Fe islands can be expressed as

$$U_g(C) = \frac{e}{C} + U_0 = \frac{e}{\epsilon_{\text{MgO}}\epsilon_0 A} d + U_0. \quad (1)$$

For Pd islands, note that  $U_0=0$  V implies that  $U_g \rightarrow 0$  V for  $1/C \rightarrow 0$  (Fig. 2). The spin-dependent case of the Coulomb staircase can be treated simply within a two-current model, i.e., the total current  $I$  is the sum of the currents of spin-up ( $\uparrow$ ) and spin-down ( $\downarrow$ ) electrons,  $I=I_\uparrow+I_\downarrow$ .

To determine the spin-dependent energetics of the charged Fe and Pd islands, we performed *ab initio* calculations for both Fe and Pd films 2 ML thick deposited on 4 ML MgO on Fe(001), using a self-consistent multiple-scattering approach. The geometric structure was taken from x-ray diffraction experiments.<sup>18</sup> As boundary conditions the substrate was treated as a semi-infinite bulk Fe and the area above the islands as a vacuum region. The laterally infinitely extended films correspond to the limit of large islands (small  $d/A$  in Fig. 2), i.e., the most relevant case here.

Our total energy calculations showed that the magnetizations of the Fe underlayers and of the Fe islands are parallel (exchange energy  $\approx 0.3$  meV/atom). While the Fe substrate shows a significant density of states (DOS) at the Fermi level in both spin channels [Fig. 3(a)], the DOS of the Fe island reveals prominent spectral weight only in the  $\downarrow$  channel [Fig. 3(b)]. Considerable weight in the  $\uparrow$  channels shows up at energies less than  $-1$  eV and is related to  $\Delta_1$  states. At energies larger than  $-1$  eV, the  $\uparrow$  DOS is almost constant and comparably small. Although it is not equal to zero, this property can be regarded as half-metallic or as close to half-metallic.<sup>23</sup> In contrast to the Fe film, the Pd film displays

significant spectral weight in both spin channels at energies around  $E_F$ .

This difference has a direct impact on the electron transport. The Fe islands are quasihalf-metallic, i.e., a band gap for  $\uparrow$  electrons shows up at the Fermi level but not for  $\downarrow$  electrons. Hence, to have  $I_\uparrow$  nonvanishing,  $\uparrow$  electrons have to overcome the binding energy  $E_0$  and the electrostatic energy, i.e.,  $I_\uparrow=G_\uparrow(U-U_0-e/C)$  for  $U>U_0+e/C$  and  $U_0=E_0/e$ .<sup>24</sup> In contrast, the  $\downarrow$  electrons have to overcome only the electrostatic potential:  $I_\downarrow=G_\downarrow(U-e/C)$  for  $U>e/C$ . This leads to a complete CB for  $U<e/C$  (i.e., both  $\uparrow$  and  $\downarrow$  currents are inhibited). One would observe a first kink at the onset of the  $\downarrow$  channel (at  $e/C$ ) and a second kink at the onset of the  $\uparrow$  channel in the  $I(U)$  spectrum (at  $e/C+U_0$ ). Further, if  $G_\downarrow$  is small compared to  $G_\uparrow$ , one would observe mainly the kink at  $U=e/C+U_0$ , which results in an offset by  $U_0$  in the  $U(1/C)$  data. Indeed, *ab initio* calculations have shown that  $G_\downarrow \ll G_\uparrow$  due to selective transmission of  $\Delta_1$  electrons of Fe through the MgO film.<sup>19-21</sup> These theoretical predictions were confirmed by the observation of giant tunneling magnetoresistances in this system.<sup>22</sup> Concluding, the large asymmetry in the spin-resolved conductances together with the significantly shifted energetic position of the  $\Delta_1^\downarrow$  band (with respect to the  $\Delta_1^\uparrow$  band) explains the observed offset of 1 eV in  $U_g$ .

In summary, the experimental findings can be explained if the Fe islands are quasihalf-metallic and a fundamental band gap in the density of states (DOS) for  $\uparrow$  electrons is present at energies of  $E_F-1$  eV and above. No such anomaly was observed for Pd, in agreement with the considerable DOS at energies close to  $E_F$  in both spin channels.

\*Corresponding author. Electronic address: wulf@mpi-halle.de

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- <sup>23</sup>The half-metallic DOS of the Fe islands is independent of the oxidation state of the lower Fe/MgO interface.
- <sup>24</sup>The value of  $E_0$  is independent of the island size for clusters larger than about 50 atoms.