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Magnetic properties of iron adatoms and small iron clusters on Ag(1 0 0)

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Abstract

A Green's function embedding technique based on the fully relativistic spin-polarized Screened Korringa–Kohn–Rostoker method is used to calculate the electronic and magnetic properties of magnetic nanostructures. Strongly enhanced spin and orbital moments are obtained for an Fe adatom and for small clusters of Fe on a Ag(1 0 0) surface. As a consequence, for an Fe adatom a magnetic anisotropy energy is found that is about 10 times larger than for an Fe monolayer. Furthermore, the exchange coupling energy between two Fe adatoms is calculated in terms of the force theorem, showing a very rapid decay with increasing distance.

Keywords: Magnetic nanoparticles; Green's function technique; Orbital magnetization; Magnetic anisotropy; Exchange coupling

Magnetic nanostructures such as impurities, small clusters, islands and wires on top or in the uppermost layers of surfaces are of special interest for nano-scale technologies, in particular, regarding their challenging application to high-density magnetic recording. Therefore, the understanding of the magnetic properties of such structures, namely, the size and the orientation of spin and orbital moments, magnetic anisotropy energies and the magnetic interactions between them, is an important challenge for ab initio calculations. First attempts were already made by the Jülich and Munich groups [1,2], who applied a relativistic extension of the local density approximation (LDA) to the density functional theory within the Korringa–Kohn–Rostoker (KKR) Green's function formalism.

In this contribution, we present the first results based on a similar theoretical approach, namely, in terms of the relativistic Screened KKR method [3], which proved to be extremely useful in calculating magnetic properties, in particular, magnetic anisotropy energies (MAE) of ordered and disordered surfaces, multilayers and superstructures (for a review see Ref. [4]). Multiple scattering theory enables the treatment of a finite cluster of impurities embedded into a two-dimensional translationally invariant host,

$$\boldsymbol{\tau}_{\mathrm{clus}} = \boldsymbol{\tau}_{\mathrm{host}}\left[\mathbf{I} - (\mathbf{t}_{\mathrm{host}}^{-1} - \mathbf{t}_{\mathrm{clus}}^{-1})\boldsymbol{\tau}_{\mathrm{host}}\right]^{-1}, \tag{1}$$

where $\boldsymbol{\tau}_{\mathrm{clus}}$ and $\boldsymbol{\tau}_{\mathrm{host}}$ are the matrices of the scattering path operator for the embedded cluster and for the host, which account for all the scattering events in the respective media, while $\mathbf{t}_{\mathrm{clus}}^{-1}$ and $\mathbf{t}_{\mathrm{host}}^{-1}$ stand for the single-site scattering matrices of the impurity and host atoms, respectively. The positions of the scatterers are fixed by the parent lattice of the host, i.e., no attempt to include lattice relaxation effects was made. By solving the Poisson equation with appropriate boundary conditions, self-consistent calculations for the embedded clusters were performed. In order to elucidate charge transfer effects, a sufficient number of host atoms had to be included in the cluster. The potentials were treated within the atomic sphere approximation (ASA); for the evaluation of the Madelung potentials an expansion of the charge densities up to $\ell = 2$ was included.

We tested the stability of the calculated properties for an Fe adatom on Ag(1 0 0) by increasing the number of self-consistently calculated atoms, N_c, in the cluster from 19 to 67. Note that in all clusters 13 empty spheres (part of the vacuum region) surrounding the adatom are present and that the orientation of the magnetization was fixed to be normal to the surface. As can be seen

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from Fig. 1, a reliable convergence for the spin moment (S_z) and the orbital moment (L_z) of the Fe adatom was achieved, albeit in the case of L_z $N_c > 40$ was needed to consider. Comparing the values $S_z = 3.39$ and $L_z = 0.88\,\mu_B$ with the respective values 3.15 and $0.14\,\mu_B$ for an Fe overlayer on Ag(1 0 0), a substantial enhancement of the spin moment and, in particular, of the orbital moment is evident.

We also calculated the magnetic anisotropy energy (MAE), $E_x - E_z$, of an adatom in the spirit of the force theorem, i.e., neglecting self-consistency for the orientation of the magnetization along the x-axis. We obtained 5.63 meV for the MAE, which is more than 10 times larger than in the monolayer case (0.46 meV). Thus, in connection with a strongly enhanced orbital moment, we predict an extreme tendency to perpendicular anisotropy for an Fe adatom. Note that Cabria et al. [2] got a smaller orbital moment ($0.55\,\mu_B$) and, quite contradictory, a negative MAE (−0.98 meV) predicting an in-plane orientation for the magnetic moment of an Fe adatom on Ag(1 0 0).

We also performed self-consistent calculations for small planar clusters of Fe on Ag(0 0 1) by considering different magnetic orientations. The results for the spin and orbital moments (including also the case of the adatom) are summarized in Table 1, where the inequivalent atoms of a cluster are labelled by their coordination number n_c defined as the number of closest Fe atoms. In the case of $N_c = 5$ and 9, and for an in-plane magnetic orientation, nominally n_c can be the same although different kinds of Fe atoms are referred to, see Table 1. As seen in Table 1, the moments systematically reduce with increasing coordination number, converging more or less to the monolayer values (see above). The abrupt decrease of the orbital moment from a single adatom to a dimer is remarkable. In general, the spin moments are quite insensitive to the magnetic orientation. In contrast, the orbital moments are considerably larger for a magnetization normal to the plane than for that in plane.

Table 1
Calculated spin moments, S and orbital moments, L (in units of μ_B) for small clusters of Fe on a Ag(1 0 0) surface with magnetization perpendicular (z) and parallel (x) to the surface[a].

Cluster	n_c	S_z	L_z	S_x	L_x
	0	3.39	0.88	3.39	0.51
	1	3.31	0.32	3.31	0.20
	2	3.25	0.25	3.29	0.13
	1	3.33	0.46	3.33	0.28
	2	3.26	0.18	3.25	0.16
	4	3.13	0.15	3.13	0.12
	1	3.35	0.37	3.36	0.29
	1*			3.36	0.34
	4	3.15	0.12	3.15	0.12
	2	3.23	0.16	3.24	0.20
	3*	3.23	0.33	3.23	0.15
	3*			3.23	0.14

[a] In the first column the arrangement of the Fe atoms in the clusters is sketched. The different Fe atoms in a cluster are identified by their coordination number n_c. In the case of clusters with 5 and 9 Fe atoms, for a magnetization along the x-axes the lines labelled by n_c^* refer to atoms depicted by squares.

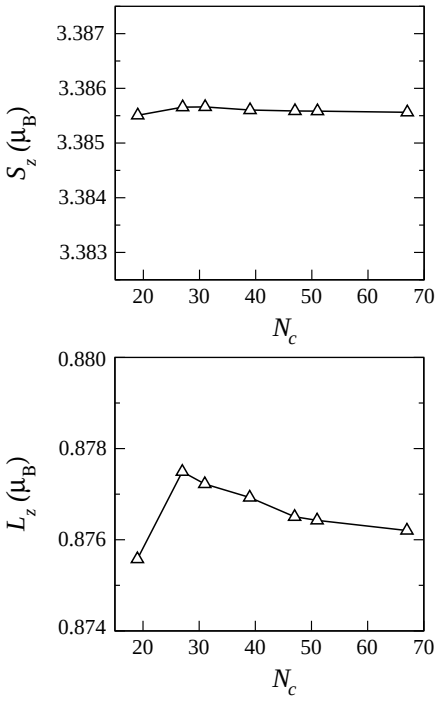

Fig. 1. Calculated spin moments (S_z) and orbital moments (L_z) of an Fe adatom on a Ag(1 0 0) surface as a function of the number of atoms in the cluster, N_c, surrounding the adatom.

Finally, we calculated the magnetic interaction ΔE_X of two Fe adatoms placed along the x-axis of the Ag(1 0 0) surface

$$\Delta E_X = E_{\uparrow\uparrow} - E_{\uparrow\downarrow}\,, \tag{2}$$

where the arrows sketch the relative orientation of the magnetic moments of the individual adatoms. Here, we again made use of the force theorem, i.e., we performed self-consistent calculations only for the $\uparrow\uparrow$ configuration (actually with the magnetization along the z-direction), then by reversing the orientation of one of the Fe moments evaluated the difference in band energies between these two particular configurations. Due to the lack of self-consistency, for near adatoms this

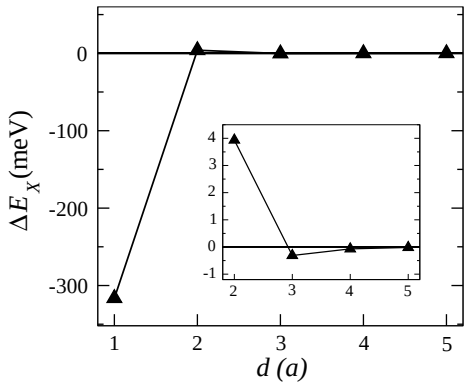

Fig. 2. Exchange coupling energy, ΔE_X (see Eq. (2)) of two Fe adatoms on a Ag(1 0 0) as a function of their distance d (measured in units of a the 2D lattice constant a). In the inset the range of $2a \leqslant d \leqslant 5a$ is shown on an enlarged scale.

approach might be quite poor, however, we believe that it provides a good estimate to the order of magnitude of the interaction. ΔE_X is shown in Fig. 2 as a function of the distance d between the two adatoms. Note that the case $d = a$, where a is the 2D lattice constant, refers to the dimer. Apparently, for this case there is a strong, direct exchange coupling of ferromagnetic character between the two Fe atoms. Increasing the separation between the two adatoms, ΔE_X rapidly decreases, even changes sign, and virtually vanishes for $d \geqslant 5a$. Thus we conclude that there is a very weak magnetic interaction between the Fe adatoms induced by the Ag host.

In conclusion, we presented a first principles, relativistic approach to deal with magnetic nanostructures on a surface of a metallic host. We performed systematic calculations for Fe clusters on Ag(0 0 1) in order to explore important magnetic properties of the system.

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