

Magnetic properties of a Cr trimer on Au(1 1 1) surface

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Available online 27 April 2007

Abstract

A Cr trimer on Au(1 1 1) surface has been studied by means of a relativistic Green's function embedding technique [B. Újfalussy, B. Lazarovits, L. Szunyogh, G.M. Stocks, P. Weinberger, Phys. Rev. B 70 (2004) 100404(R)]. In terms of the magnetic force theorem we fit parameters of an extended Heisenberg model. We then solve the Landau–Lifshitz–Gilbert equations in order to find the ground-state and to study the spin-dynamical properties of the cluster.

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Keywords: Magnetic properties; Magnetic force theorem; Cluster; Cr trimer

1. Introduction

Magnetic nanostructures on nonmagnetic substrates have recently been in the focus of theoretical and experimental investigations due to their novel properties and their promising applications in high density magnetic data recording. The development in scanning tunneling microscopy and the ability to build clusters with well-controlled structures have opened a new insight into the local interactions within magnetic nanoclusters. Systems exhibiting magnetic frustration such as chromium clusters are of particular interest to theoretical [2–5] and to experimental studies [6,7]. The half-filled valence configuration of Cr yields a large magnetic moment and strong interatomic bonding, leading in turn to magnetic frustration and complex spin phenomena. The simplest system exhibiting such properties is a trimer.

The coupling between the magnetic and electronic degrees of freedom of magnetic clusters and the conducting substrate can lead to a sharp resonance in the differential conductance at low bias which is the main signature of the Kondo effect resulting from the screening of the adatom spin by the surrounding conduction electrons [8,9]. The

lack or presence of this resonance provides additional information on the magnetic structure of a cluster.

According to the resonance properties of the differential conductance of the STM two magnetically different structure can be distinguished. A linear chain and an equilateral triangle displayed featureless spectra while an isosceles triangle exhibited a narrow resonance at the Fermi energy [7].

In the present paper the magnetic properties of a Cr trimer with equilateral structure has been studied. Recently, the magnetic properties of noncollinear structures of supported chromium clusters have been investigated theoretically within the phenomenological Anderson [2,10] model and from first principles [3]. However, according to our knowledge, this work is the first fully relativistic study on supported equilateral chromium trimers.

2. Theoretical approach

Within multiple-scattering theory, the matrix of the so-called scattering path operator (SPO), $\tau_{\mathcal{C}}$, corresponding to a finite cluster \mathcal{C} embedded into a host system can be obtained from the following Dyson equation:

$$\tau_{\mathcal{C}}(\varepsilon) = \tau_h(\varepsilon)[\mathbf{I} - (\mathbf{t}_h^{-1}(\varepsilon) - \mathbf{t}_{\mathcal{C}}^{-1}(\varepsilon))\tau_h(\varepsilon)]^{-1}, \quad (1)$$

where $\mathbf{t}_h(\varepsilon)$ and $\tau_h(\varepsilon)$ denote the single-site scattering matrix and the SPO matrix for the unperturbed host confined to

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the sites in \mathcal{C} , respectively, while $\mathbf{t}_{\mathcal{C}}$ comprises the single-site scattering matrices of the embedded atoms. Once $\tau_{\mathcal{C}}$ is derived, all quantities of interest, the charge and magnetization densities and the spin and orbital moments can be calculated [1]. In our calculations the chromium atoms are situated at the hollow sites on the top of an FCC Au(1 1 1) surface. In order to find the self consistent potentials a cluster consisting of the trimer and the sites in the first nearest neighbor shell is embedded into the interface region of the Au(1 1 1) surface. The magnetization was assumed to be collinear and perpendicular to the substrate. In a procedure based on the magnetic force theorem the band energy of the cluster has been determined in various magnetic configurations using the potentials provided by the self-consistent calculation. The result here of is then fitted by the following classical vector spin model:

$$H = \frac{1}{2} \sum_{i \neq j} \sigma_i \mathbf{J}_{ij} \sigma_j + \sum_i \sigma_i \mathbf{K}_i \sigma_i + \frac{1}{2} Q_1 \sum_{i \neq j} (\sigma_i \sigma_j)(\sigma_i \sigma_j) + \frac{1}{2} Q_2 \sum_{i \neq j \neq k} (\sigma_i \sigma_j)(\sigma_i \sigma_k), \quad (2)$$

where σ_i is a unit vector parallel to the magnetization at site i , \mathbf{J}_{ij} and \mathbf{K}_i are the exchange and anisotropy tensors, respectively, and Q_1 , Q_2 describe a bi-quadratic coupling between the spins. The symmetry of the system constrains the matrix element of the exchange and anisotropy matrices. Obviously, \mathbf{J}_{13} , \mathbf{J}_{23} as well as \mathbf{K}_2 and \mathbf{K}_3 can be obtained by a 120° rotation from \mathbf{J}_{12} and \mathbf{K}_1 , respectively. Further investigations pointed out that there are only three independent matrix elements of the anisotropy tensor. In the adiabatic description the time evolution of the magnetization can be obtained from the Landau Lifshitz Gilbert (LLG) equation:

$$\frac{\partial \mathbf{M}}{\partial t} = -\frac{\gamma}{1 + \alpha^2} \mathbf{M} \times \mathbf{B}_{\text{eff}} - \frac{\alpha \gamma}{1 + \alpha^2} \frac{1}{M} \mathbf{M} \times (\mathbf{M} \times \mathbf{B}_{\text{eff}}), \quad (3)$$

where γ is the gyromagnetic ratio and α is a dimensionless Gilbert damping factor. The effective field \mathbf{B}_{eff} driving the motion of the magnetic moments can be derived from Eq. (2) as the derivative of the energy with respect of the magnetization. Solving the LLG equations with an appropriate damping parameter the magnetization will converge to the ground state, or, if it exists, to a metastable state of the system.

3. Results and discussion

For a clear physical interpretation of the ground state the exchange tensor \mathbf{J}_{ij} is decomposed into an isotropic term $J_{ij} = \frac{1}{3} \text{Tr}(\mathbf{J}_{ij})$, a symmetric $\mathbf{J}_{ij}^S = \frac{1}{2}(\mathbf{J}_{ij} + \mathbf{J}_{ij}^\dagger) - J_{ij} \mathcal{I}$ and an antisymmetric part $\mathbf{J}_{ij}^A = \frac{1}{2}(\mathbf{J}_{ij} - \mathbf{J}_{ij}^\dagger)$. This latter term is responsible for the Dzyaloshinsky–Moriya (DM) interaction. The electronic structure calculations resulted in a large antiferromagnetic isotropic coupling of $J = 136$ meV as it is expected. The next contributions in magnitude are the bi-quadratic terms $Q_1 = 4$ meV, $Q_2 = 13$ meV. Since they are

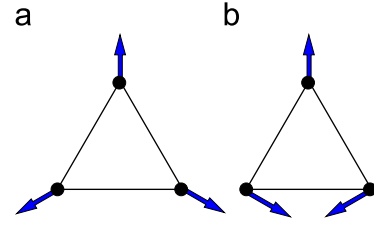


Fig. 1. Two ground state configurations of the trimer: (a) $\kappa_z = 1$ and (b) $\kappa_z = -1$.

invariant against $O(3)$ rotations they do not introduce any change in the magnetic configuration determined by isotropic exchange only. Both the symmetric anisotropic exchange \mathbf{J}^S and the on-site anisotropy \mathbf{K} are below 1 meV and prefer in-plane orientation. The length of the DM vectors are $D = 1.5$ meV. The triangular system with antiferromagnetic coupling is a basic example of a frustrated system. Confining the motion of the vector spins in the plane of the triangle there are two classes of stationary states with different chirality (see Fig. 1) as is detailed by Kawamura in Ref. [11]. The chirality of the system can be characterized by a chirality vector $\boldsymbol{\kappa} = \frac{2}{3\sqrt{3}} \sum_{(ij)} (\sigma_i \times \sigma_j)$ where the summation runs over the three directed bonds forming a triangle. Obviously the $\boldsymbol{\kappa}$ vector is perpendicular to the plane of the triangle for in-plane magnetization. In three dimensions there is no longer a discrete chiral degeneracy since the two spin configurations can be transformed into each other by continuous 3d spin rotation. However, if the anisotropy is large enough to keep the magnetization in the plane the degeneracy will still be present. Starting from random orientations the simulations resulted in one of the configurations shown in Fig. 1 with a negligible canting off the plane. At the two states the system has local minima but the degeneracy is lifted due to the anisotropy. Structure (b) has lower energy and the splitting is $\Delta = 5.7$ meV. In order to find the origin of the splitting we repeated our calculations without taking the DM interaction into account. The two configurations can be transformed into each other canting the vector spins at the endpoints of a side of the triangle towards the side as it is shown in Fig. 2.

As can be clearly seen from Fig. 2 neglecting the DM interaction states with different chirality have the same energy. In the DM interaction, $E_{\text{DM}} = \sum_{(ij)} \mathbf{D}_{ij}(\sigma_i \times \sigma_j)$, only the z components of the \mathbf{D}_{ij} vectors play a role which must be the same for all the pairs due to the symmetry of the system. The contribution of the DM interaction to the energy can be expressed by the chirality vector as

$$E_{\text{DM}} = \frac{3\sqrt{3}}{2} D_z \kappa_z. \quad (4)$$

Now it is clear that the DM interaction shifts the energy of the systems with different chirality to the opposite directions.

So far the self-consistent potential has been determined for the normal to plane ferromagnetic configuration (V_{ferro}). In order to find a potential consistent with the ground states self-consistent calculations have been performed for the $\kappa_z = 1$ system ($V_{\text{in-plane}}$) and the simulations

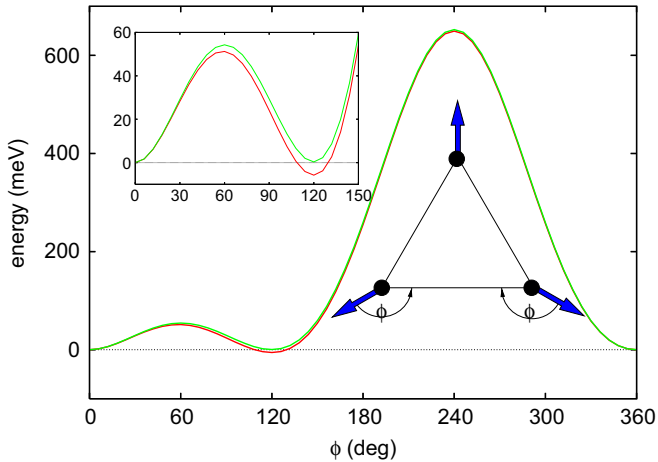


Fig. 2. Energy of the trimer as a function of the canting angle ϕ . The red and green curves represent the results obtained by including and excluding the DM interactions, respectively. In both cases the energy of state (a) (see Fig. 1) has been chosen to be the zero level.

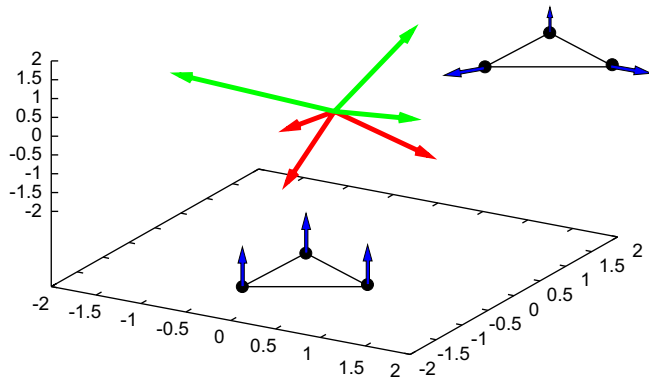


Fig. 3. DM vectors provided by the two different self consistent potentials. The red arrows belong to the potential with ferromagnetic configuration and the green arrows belong to the potential determined for the in-plane configuration. The triangles with arrows denote the two different conformations the potential has been calculated for.

have been repeated using the new set of parameters. The isotropic exchange is slightly enhanced and the biquadratic terms are reduced but the most striking difference caused by the new potential is the change of the sign of the z component of the DM vectors as is depicted on Fig. 3.

According to Eq. (4) the shift due to the DM interaction has just the opposite sign as it was in the previous case, consequently, the configurations denoted by (a) in Fig. 1 becomes the new ground state. The magnetic anisotropy energy (MAE) defined as the difference between the energy of the normal to plane and the energy of the in-plane ferromagnetic system turned out to be an order of magnitude smaller (Table 1). The reduced anisotropy energy cannot keep the magnetic moments in the plane of the triangle and the state corresponding Fig. 1(b) is no longer a local minimum of the system.

In order to confirm our results a self-consistent spin dynamic calculation has been performed applying a

Table 1

Magnetic anisotropy energy corresponding to the potentials determined for the ferromagnetic and in-plane magnetic configuration

$$E_{\text{ferro}} = 1.7 \text{ meV}$$

$$E_{\text{in-plane}} = 0.03 \text{ meV}$$

method which was developed by Stocks et al. [12,13] based on the concept of the constrained density functional theory. In this procedure a local (transverse) constraining field—that can be determined self-consistently—ensures the stability, within DFT, of the nonequilibrium orientational state demanded by the equation of motion. The constraining field SD simulations provided the same ground state as we got using the self-consistent potential obtained for in-plane magnetization. During the simulations there were no sign of any metastable state in agreement with results of the simulations based on the model Hamiltonian in Eq. (2).

4. Summary

A fully relativistic Green's function embedding method has been applied to determine the electronic structure of a Cr trimer on Au(111) surface. The band energy of several magnetic configurations of the trimer has been calculated and the results have been fitted by a model Hamiltonian containing the essential magnetic interactions. Following the time evolution of the magnetization described by the LLG equation two stationary states have been found. The self-consistent potentials have been recalculated for both states and new set of exchange and anisotropy parameters have been determined. This procedure turned out to be very effective in finding the magnetic ground state of the system. Furthermore, it gives an insight into the role of different magnetic interactions on the properties of small clusters. We have found that the DM interaction lifted the degeneracy of the states with different chirality.

Acknowledgment

The present work is supported by the Center for Computational Materials Science (Contract Nos. FWF W004 and GZ 45.547) and the Hungarian National Scientific Research Foundation (OTKA T037856 and T046267).

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