

Magnetic moments of a pair Cr_4 atomic clusters embedded in bulk Fe in terms of the spatial orientation

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The magnetic properties of a pair Cr_4 atomic clusters embedded in bulk Fe are determined by using a realistic *spd*-band Hubbard-like model. The spin density distribution is calculated self-consistently in the unrestricted Hartree-Fock approximation. The local magnetic moments μ_i are obtained at various atoms i of the cluster and of the surrounding Fe matrix. We consider two different orientations between the clusters; the axis of symmetry of the clusters are collinear (C) and non-collinear (NC), for all the cases studied we compare the results with the results to just one Cr_4 atomic cluster embedded in bulk Fe.

Keywords: Local magnetic moments; Hybridizations; Magnetic interface coupling; Spatial orientation

1. Introduction

Advances in preparation techniques now allow the production of ferromagnetic and/or nonmagnetic nanostructures, this enables the very interesting possibility of inducing magnetic moments in materials not normally magnetic by placing them in close proximity to a ferromagnetic structure. These interlayer magnetic moments are ultimately responsible for the oscillatory exchange coupling which is observed in these systems [1]. This occurs as follows: hybridization at the ferromagnetic and/or nonmagnetic interface induces a magnetic moment in the electrons belonging to the interlayer atoms. Indirect exchange coupling is generally expected to be orientation dependent, since (in an RKKY model) it depends on the magnitude of the Fermi wave vector parallel to the growth axis [2]. This has been verified specially in Fe/Cr through first-principles calculations of multilayers in various orientations; when calculated directly, the interlayer moments in Fe/Cr multilayers are predicted to depend on crystallographic orientation. Additionally, their detailed behavior also depends on the nature of the Fe/Cr interfaces and the Fe moment configuration [3], that is, whether the Fe layers are arranged ferromagnetically, or otherwise. The measurement of interlayer moments is challenging because the ferromagnetic layer moments overwhelm the very small interlayer moments [3]. A much less work has focused on the magnetic moments of embedded atomic clusters [4] and on the magnetic moments of a set of interacting embedded clusters in a ferromagnetic matrix. In this paper we study the specific problem about the magnetic moments of a pair of Cr_4 atomic clusters embedded in bulk Fe in terms of the orientation between them due to the competition between the antiferromagnetism of Cr and the ferromagnetism of the Fe matrix offers particularly interesting physical situations for studying the interplay between the magnetic properties of

interacting clusters and those of the environment, the study of these systems stresses the local aspects of the magnetic interactions between ferromagnetic and antiferromagnetic materials. Therefore, from the perspective of interface magnetism, this work provides useful complementary information to investigations of the Fe/Cr systems.

2. Model

In order to study this problem we consider a realistic model Hamiltonian for the valence *s*, *p* and *d* electrons, which includes intra-atomic Coulomb interactions in the unrestricted Hartree-Fock approximation. In the usual notation, the Hamiltonian is given by

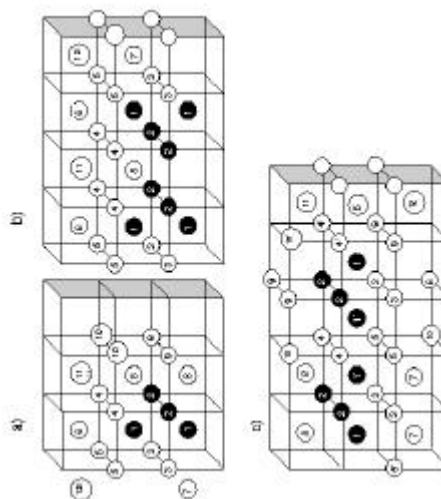


Figure 1. Illustration of the lattice of Cr_4 clusters embedded as substitutional impurities in bulk Fe (bcc structure). The Cr atoms are represented by filled circles and the Fe atoms by open circles. The numbers label the nonequivalent atomic sites. The a) represent the structure of one Cr_4 atomic cluster, b) the (NC) Arrangement and c) represent the (C) arrangement.

$$H = \sum_{ias} \mathbf{e}_{ias} n_{ias} + \sum_{abs} \sum_{i \neq j} t_{ij}^{ab} c_{ias} c_{jbs} \quad (1)$$

where i and j refer to the atomic sites, \mathbf{a} and \mathbf{b} the orbitals (s, p, g) and σ to the spin. The hopping integrals are obtained from the canonical two-center approximation [5] and the environment dependent energy levels by \mathbf{e}_{ias} are given by:

$$\mathbf{e}_{ias} = \mathbf{e}_{ia}^0 + \sum_b (U_{ab} \Delta \mathbf{n}_{ib} - \mathbf{S} J_{ab} \mathbf{m}_{ib}) + \zeta_i \Omega_a$$

where \mathbf{e}_{ias}^0 stands for the energy level of the orbital α of atom i in the paramagnetic solution of the bulk (e.g., Cr, V, or Fe). The second term in eq. (2) takes into account the level shifts due to the redistribution of the spin polarized density and to the resulting intra-atomic Coulomb interactions. $\Delta \mathbf{n}_{ib} = \mathbf{n}_{ib} - \mathbf{n}_{ib}^0$, where $\mathbf{n}_{ib} = \langle \hat{\mathbf{n}}_{ib} \rangle + n_{ib} \hat{\mathbf{n}}$ is the average electronic occupation of the orbital ib and \mathbf{n}_{ib}^0 the corresponding average occupation in the bulk $\mu_{ib} = \langle n_{ib} \rangle - n_{ib}$ refers to the spin polarization of the orbital ib . The intra-atomic direct and exchange Coulomb integrals are denoted by U_{ab} and J_{ab} respectively. Finally, the last term in eq. (2) takes into account the environment-dependent energy-level shifts due to nonorthogonality effect [5] and to the crystal-field potential of the neighboring atoms [6], \mathbf{n}_{ia} and \mathbf{m}_{ib} at site i are determined self-consistently by requiring

$$\langle n_{ias} \rangle = \int_{-\infty}^{\epsilon_F} \mathbf{r}_{ias}(\mathbf{e}) d(\mathbf{e}) \quad (2)$$

where $\mathbf{r}_{ias}(\epsilon) = (-I/p) \text{Im} G_{ias,ias}(\epsilon)$ refers to the local density of states (DOS) at the spin-orbital ias . In the case of finite embedded clusters, the Fermi energy ϵ_F is given by the matrix (in the present case Fe). The local Green's functions are calculated from Eqs. (1) and (2) by using the recursion method [7]. The number of levels M of the continued fraction expansion of $G_{ias,ias}$ is chosen large enough so that the calculated orbital occupations and magnetic moments are independent of M . In all the cases here studied local charge neutrality is imposed at each site i

3. Results and discussion

The lattice structure is illustrated in Fig. 1. Here the numbers label the different atomic sites at each case; Fig. 1a shows the structure of one embedded atomic cluster, Figs. 1b and 1c show the structure for a pair of atomic clusters when the axis of symmetry are non-collinear (NC) and collinear (C) respectively. In the Table 1, we show the connectivity between the atomic sites in the all the cases studied, because some sites do not appear in Fig. 1, in the Table 2 we show the values of the local magnetic moments for all the cases studied here. In the case of a single Cr_4 atomic cluster, the local magnetic moments are remarkably enhanced as compared to the sublattice magnetization $\mu_b = |0.66| \mu_B$ of bulk Cr, the magnetic moments $\mu(1)$ and $\mu(2)$ are treated as independent variables. Thus, the result $\mu(1) = \mu(2)$ is not obvious a priori, particularly since $i = 1$ and $i = 2$

Table 1. The connectivity between the different atoms around the Cr_4 clusters is shown, because in the figure 1 does not exist all the atoms in the structures; the upper panel represents the connectivity in the (NC) configuration figure 1b) and the lower panel represents the connectivity in the (C) configuration figure 1c). The numbers refer to the Fe matrix.

Connectivity between the different atoms in one Cr_4				
1→13	2→14	3→14	4→16	5→17
6→18	7→19	8→20	9→21	10→22
Connectivity between the different atoms in configuration (NC). The site type 9 and 10 do not exist in this case				
1→13	2→14	3→15	4→16	5→17
6→13	4→19	8→20		
Connectivity between the different atoms in configuration (C)				
1→13	1'→13'	2→14	3→15	4→16
4'→16'	5→17	5'→17'	6→18	7→19
7'→19'	8→20	8'→20'	9→21	10→22
10'→22'				

are first NN and would thus belong to different magnetic sublattices in Cr, the Cr moments point in the same direction, this is, the magnetic order within the cluster is ferromagnetic-like and the magnetic coupling at the interface cluster-matrix is antiferromagnetic, this behavior is a common property among Fe/Cr interfaces. This behavior can be qualitatively understood in terms of the hybridizations between Fe and Cr d orbitals. In the case of Cr, the d orbital energy levels $\epsilon_{d\sigma}$ (Cr) are closer to the Fermi energy ϵ_F and to the spin-down d levels of Fe than to spin-up Fe d levels that are well below ϵ_F . Therefore, the down states of Cr hybridize more strongly with the Fe matrix than the Cr up states. Consequently, Cr down states are preferentially occupied yielding antiferromagnetic interface coupling [8]. The larger exchange splitting at the Fe atoms and the fact that the almost all the NN of the Cr atoms are Fe fields large $\mu(i)$ within the cluster, the magnetic order in the matrix is not modified by the presence of the Cr atoms, but some $\mu(\text{Fe})$ are reduced about 7%, because they have 2 Cr NN. This same behavior in the magnetic order is found in the cases (NC) and (C), into the clusters the magnetic order is ferromagnetic-like, and at the interface clusters-matrix the magnetic coupling is antiferromagnetic, see Table 2, the presence of other atomic cluster, with the same symmetry and same number of atoms modifies the value of the different $\mu(i)$ of the system and in the case (C) the number of non-equivalent atomic sites is increased, see Tables 1 and 2. For the case (NC), in the Cr atoms ($i = 1$ and $i = 2$), both atomic sites are reduced but $\mu(2)$ is reduced stronger than $\mu(1)$, see Table 2 due to the proximity of the other Cr cluster, see Fig. 1, this structure modifies the local environment of some Fe sites, the atomic site $i = 8$ now it has 4 Cr NN and 4 Fe NN atoms, therefore $\mu(8)$ is reduced about 14%, in the other Fe sites the $\mu(i)$ does not change significantly, see Table 2;

Table 2. Local magnetic moments $\mu(i)$, in units of μ_B , for the structures of Cr_4 clusters embedded in bulk Fe (results closed by parenthesis), the other numbers represent the nonequivalent sites, the sites in boldface represent the Cr atoms and the other numbers un roman represent the Fe sites.

One Cr_4										
1	2	3	4	5	6	7	8	9	10	11
(-1.34)	(-1.34)	(1.97)	(2.12)	(2.18)	(2.23)	(2.24)	(1.97)	(2.24)	(2.29)	(2.26)
12	13	14	15	16	17	18	19	20	21	22
(2.27)	(2.12)	(2.23)	(2.26)	(2.27)	(2.28)	(2.27)	(20.29)	(2.18)	(2.27)	(2.28)
(NC) Arrangement										
1	2	3	4	5	6	7	8	9	10	11
(-1.32)	(-1.13)	(1.99)	(2.15)	(2.18)	(2.23)	(2.24)	(1.73)			(2.26)
12	13	14	15	16	17	18	19	20	21	22
(2.27)	(2.13)	(2.24)	(2.26)	(2.28)	(2.28)	(2.27)	(2.29)	(2.26)		
(C) Arrangement										
1	1'	2	3	4	4'	5	5'	6	7	7'
(-1.34)	(-1.16)	(-1.32)	(1.98)	(2.01)	(2.01)	(2.18)	(2.09)	(2.23)	(2.24)	(2.25)
8	8'	9	10	10'	11	12	13	13'	14	15
(1.98)	(1.98)	(2.24)	(2.31)	(2.29)	(2.26)	(2.27)	(2.12)	(2.13)	(2.23)	(2.26)
16	16'	17	17'	18	19	19'	20	20'	21	22
(2.28)	(2.27)	(2.28)	(2.29)	(2.27)	(2.29)	(2.30)	(2.20)	(2.18)	(2.28)	(2.29)

the other geometrical arrangement is the (C) case, where the axis of symmetry are collinears, see Fig. 1c, in this case into the cluster, the change most significantly is $\mu(1')$, this magnetic moment changes about 13%, this site is closer to the other cluster than the sites 1 and 2 and interacts stronger with the other cluster, while the site 1 is screened by the sites 1' and 2 therefore $\mu(1)$ is not changed by the presence of the other cluster, in the Fe matrix only the site type 4 the local environment is changed and $\mu(4)$ is reduced about 5% keeping the antiparallel alignment with the Cr atoms in the interface and the same time the parallel alignment with the matrix. An interesting magnetic behavior is expected for clusters with different symmetry, the changes in the local environment in the atoms is stronger than in the case of symmetrical clusters.

4. Conclusions

The magnetic properties of Cr_4 atomic clusters have been determined by solving a realistic spd band model Hamiltonian in the unrestricted Hartree-Fock approximation. The main conclusions are summarized as follows.

- The number of nonequivalent sites depends of the orientation between the Cr_4 clusters, in the case (C) exist most nonequivalent sites, see Table 1, therefore in this arrangement the problem is more difficult to solve from a computational point of view.
- For one Cr_4 , the magnetic order is ferromagnetic-like and the magnetic moments, $\mu(1)$ and $\mu(2)$ are enhanced with respect to Cr bulk magnetization, see Table 2.
- For the cases (C) and (NC) this magnetic order is not broken, the changes are in the modulus of the $\mu(i)$, in both cases the local magnetic moments in Cr atoms are enhanced too, see Table 2 I

- In all the cases studied, the magnetic coupling at the interface Cr-Fe is antiferromagnetic, keeping the typical magnetic coupling in the Fe/Cr systems, see Table 2.
- The magnetic order in the matrix is not changed due the presence of the Cr clusters, the only changes are in the modulus of the $\mu(i)$ in the Fe atoms closer to the Cr atoms, see Table 2.

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