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Ab initio calculation of Heisenberg parameters in thin magnetic films

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ABSTRACT

We present first principles calculations of the Heisenberg exchange parameters (J_{ij}) in thin films on top of a disordered local moment description of the paramagnetic state and also from the magnetically ordered ground state. For Co and Ni films on a Cu(001) substrate ferromagnetic nearest-neighbour (NN) and next-nearest-neighbour (NNN) interactions play throughout a dominating role. Furthermore, J_{ij}'s display significant dependence on the layer positions, giving a typical maximum for layers near the surface of the film as well as the interface with the substrate. As presented for the case of the Fe_5/Cu(001) overlayer system, the long-ranged behaviour of the exchange pair interactions has to be ultimately taken into account in order to recover the antiferromagnetic ground states of thin Fe films on Cu(001) obtained by total energy calculations. Theoretical Curie temperatures (T_C) calculated within a simple statistical mean-field approach follow qualitatively well the trends against the film thickness observed in the experiments.

§1. INTRODUCTION

Various types of magnetic phase transitions in thin films have attracted much experimental and theoretical effort during the recent decade. One particularly important phenomenon is the dependence of the Curie temperature with respect to several parameters, such as the film thickness, the geometrical structure or the composition of the film. While methods with different sophistication of the statistical mechanics, based on Ising or Heisenberg type Hamiltonians, were able to elucidate, e.g. the general features of T_C as a function of the film thickness (Jensen *et al.* 1992, Strandburg *et al.* 1992), the need for obtaining realistic, material specific parameters which enter these models is obvious. Recent studies of Spišák and Hafner (1997a, b) in terms of a Green function technique showed indeed a broad hierarchy of the exchange interactions at Fe surfaces, at the interface of a Fe substrate with antiferromagnetic Mn overlayers, as well as in thin face-centred cubic (fcc) Fe films on Cu(001), which remarkably can influence the predictions of the simple statistical models.

In the present contribution we employ two techniques to calculate exchange interactions in fcc Co, Fe and Ni films on Cu(001), namely, as effective pair interactions within a disordered local moment (DLM) description of the paramagnetic state (Pindor *et al.* 1983, Györffy *et al.* 1985) and also, similar to Spisák and Hafner (1997a, b), in terms of infinitesimal rotations based on the electronic structure of the ordered (ferromagnetic or antiferromagnetic) ground state. First, self-consistent calculations were carried out for each of the overlayer systems in terms of the scalar-relativistic spin-polarized screened Korringa–Kohn–Rostoker method (Szunyogh *et al.* 1994). For the determination of the DLM state, a random alloy comprising in fact

50%:50% of components with opposite (say 'up' and 'down') spin alignments, the coherent-potential approximation (CPA) as implemented to layered systems was used (Weinberger *et al.* 1996). Subsequently, the energy of the spin system has to be mapped into pair-wise contributions,

$$H = \frac{1}{2}\sum_{pq}{\sum_{ij}}' J_{pi,qj}\,\mathbf{S}_{pi}\,\mathbf{S}_{qj}\ , \tag{1}$$

where $\mathbf{S}_{pi}$ denotes a classical vector-spin of unit length located at the ith site of layer p, $J_{pi,qj}$ is the exchange interaction between two particular sites (pi) and (qj), the factor $\frac{1}{2}$ corrects for double-counting, while the exclusion of the on-site terms is indicated by the prime in equation (1). Within a DLM picture, multiple scattering theory enables one to derive the following expression

$$J_{pi,qj} = -\frac{1}{2\pi}\mathrm{Im}\int^{E_\mathrm{F}} \mathrm{Tr}\left(\left[X^{p}_{\uparrow}(\varepsilon) - X^{p}_{\downarrow}(\varepsilon)\right]\tau_c^{pi,qj}(\varepsilon)\left[X^{q}_{\uparrow}(\varepsilon) - X^{q}_{\downarrow}(\varepsilon)\right]\tau_c^{qj,pi}(\varepsilon)\right)\mathrm{d}\varepsilon, \tag{2}$$

where E_F is the Fermi energy, $X^{p}_{\uparrow\downarrow}(\varepsilon)$ denote layer- and spin-dependent excess scattering matrices and $\tau_c^{pi,qj}(\varepsilon)$ is the respective off-diagonal CPA scattering path matrix (Weinberger *et al.* 1996). Starting from the magnetically ordered state as reference we followed the method of infinitesimal rotations by Liechtenstein *et al.* (1987) leading to

$$J_{pi,qj} = -\frac{1}{2\pi}\mathrm{Im}\int^{E_\mathrm{F}} \mathrm{Tr}\left(\left[m^{p}_{\uparrow}(\varepsilon) - m^{p}_{\downarrow}(\varepsilon)\right]\tau_{\uparrow}^{pi,qj}(\varepsilon)\left[m^{q}_{\uparrow}(\varepsilon) - m^{q}_{\downarrow}(\varepsilon)\right]\tau_{\downarrow}^{qj,pi}(\varepsilon)\right)\mathrm{d}\varepsilon. \tag{3}$$

In (3) $m^{p}_{\uparrow(\downarrow)}(\varepsilon)$ stands for the inverse of the single-site t-matrix. Note that both in equations (2) and (3) the trace is taken in angular momentum space and two-dimensional translation invariance for the layered system was assumed.

§2. Results and discussion

Figure 1 illustrates typical exchange pair interactions in the Co_5/Cu(001) and Ni_5/Cu(001) overlayers as derived by using equation (3). It should be noted that the DLM calculations for Ni overlayers resulted into much smaller magnetic moments than in the ferromagnetic state. In consequence, we obtained unrealistically small exchange interactions for these systems. Conversely, for Co overlayers the magnetic moments and also the exchange pair interactions obtained from the DLM and ferromagnetic calculations were very similar. With the exception of the monolayer case, however, possibly due to an extra effective exchange field represented by the ordered spin-state, the nearest-neighbour interactions (NNI) obtained from the ferromagnetic state were systematically larger in magnitude than those calculated from the DLM state.

In figure 1 the Heisenberg parameters are sorted with respect to pairs of layers and plotted against the distance of the pairs, $\left|\mathbf{R}_{pi} - \mathbf{R}_{qj}\right|$. Although in both overlayer systems the ferromagnetic (negative) NNIs dominate, especially for the first and second neighbour layers ($\left|q - p\right| = 1$ and 2), one can notice also sizeable NNN interactions. Clearly, NNIs near the surface (5–5 and 4–5) are largest in magnitude, a fact which apparently correlates with the well-established trend to enhanced ferromagnetism at many metallic surfaces. At the interface of the Co film and Cu substrate a similar trend applies, however, NNIs at the Ni–Cu interface (1–1 and 1–2) are obviously smaller in magnitude than those even in the interior of the Ni film (3–3

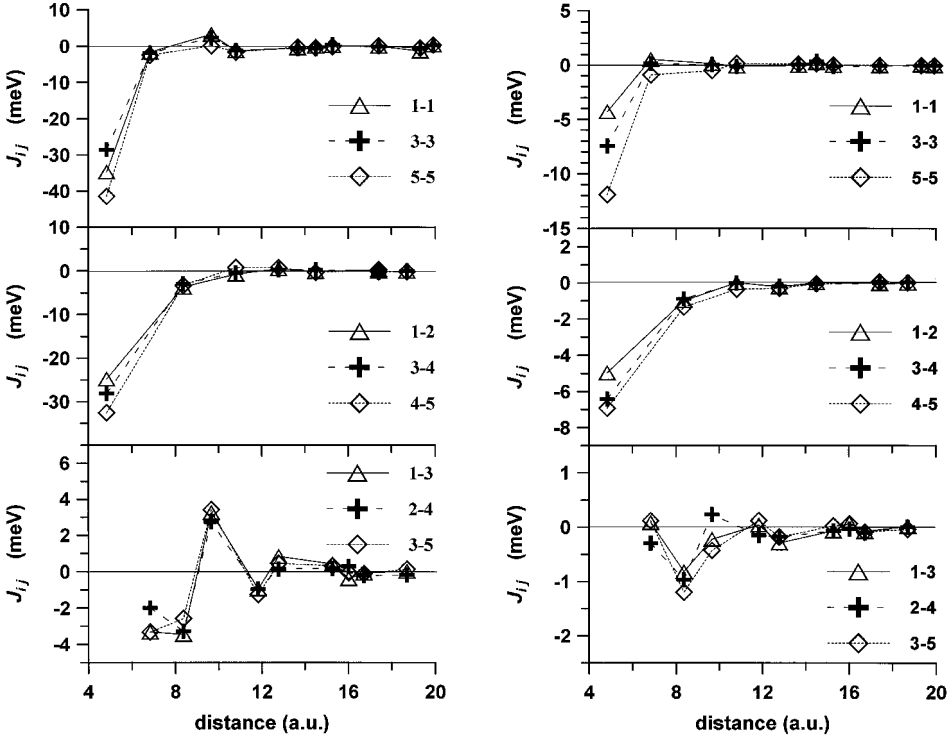

Figure 1. Heisenberg parameters as calculated by using equation (3) for (*a*) Co_5/Cu(001) and (*b*) Ni_5/Cu(001) overlayers. In the legends the pair of numbers *p*–*q* label the corresponding pair of layers. The labelling of the layers increases from the substrate toward the surface.

and 3–4). This might be a consequence that hybridization effects that can lower the trend to ferromagnetism are more pronounced between Ni and Cu states than between Co and Cu states.

As can be seen from figure 2, the hierarchy of J_{ij}'s is much more complicated in Fe_5/Cu(001) than in the previous two cases. Again, pronounced ferromagnetic NN but also NNN interactions characterize the surface region. Remarkably, the 'intra-surface' (5–5) NNI calculated from the (antiferromagnetic) ground state (right panel) is of double magnitude than that derived from the DLM state (left panel), while the corresponding surface–subsurface (4–5) NNIs are nearly equal. Conversely, the NN and NNN interactions in the Fe layer at the interface (1–1) obtained from the DLM state are clearly larger in magnitude than the corresponding interactions calculated from the ground state. It seems to be, however, that in the ground state a ferromagnetic ordering within surface (5), subsurface (4) and the interface (1) layers, as well as between the surface and subsurface layers directly arises from the interactions characterized above.

Spišák and Hafner (1997b) argue that the ↓↑↓↑↑ ground state configuration of the Fe_5 film is stabilized by a strong AFM-NNNI between the surface and the third layer (3–5), a FM-NNNI between layers 2 and 4 as well as from weak AFM interactions between the layer at the interface and that next to the interface (1–2). In

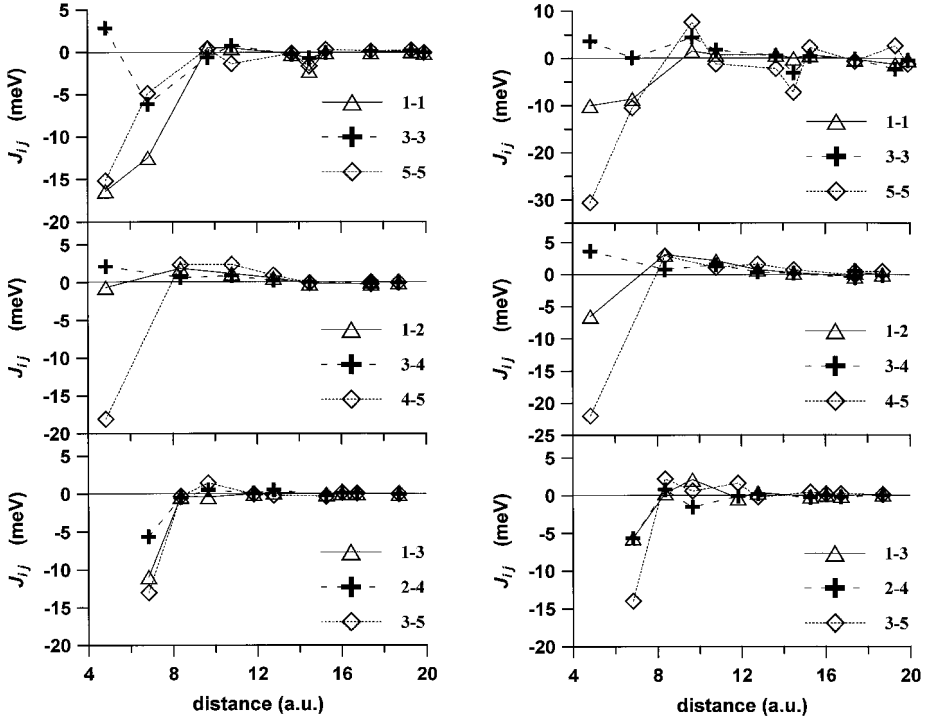

Figure 2. Heisenberg parameters for the $Fe_5/Cu(001)$ overlayer system as calculated (*a*) by using equation (2) from the DLM state and (*b*) by using equation (3) from the antiferromagnetic ground state. For the meaning of the legends see figure 1.

contrast, as can be seen from figure 2, we obtained large ferromagnetic NNNI between layers 3 and 5. One site in layer 5 has, however, only one such neighbour in layer 3, therefore, the role of more extended neighbours is increased. The relevant *effective layer–layer interactions*, $J_{pq} = \sum_i J_{p0,qi}$, are as follows: $J_{35} = -19.7$ or 15.1 meV and $J_{34} = 38.7$ or 53.7 meV as based on the DLM or the AFM ground state, respectively. Thus, in our scenario it is mainly the effective AFM coupling between the subsurface and the third layer that leads to the antiparallel alignment of spins in the third layer with respect to the two layers at the surface. Furthermore, the values $J_{23} = 36.1$ meV, $J_{12} = 42.0$ meV and $J_{13} = -20.9$ meV for the DLM calculations, but also $J_{24} = -19.6$ meV and $J_{12} = 27.3$ meV for the ordered ground state calculations clearly stabilize the ground state configuration obtained by total energy LDA calculations. Reassuringly, for all the layer thicknesses under consideration, $n = 2$–7, both the DLM and ground state exchange interactions were consistent with the ground state as reported by Szunyogh *et al.* (1997).

Finally, the Curie temperatures obtained within a mean-field approach are plotted in figure 3 together with available experimental data. Since thermal fluctuations are expected to be more and more important when the dimension of the system is reduced, our results for T_C should be regarded only as crude estimates. This is most pronounced for the monolyers and bilayers. Taking into account these general arguments, for the ferromagnetic Co and Ni films the monotonic increase of T_C with the film thickness is well reproduced. Since, as stated above, the exchange interac-

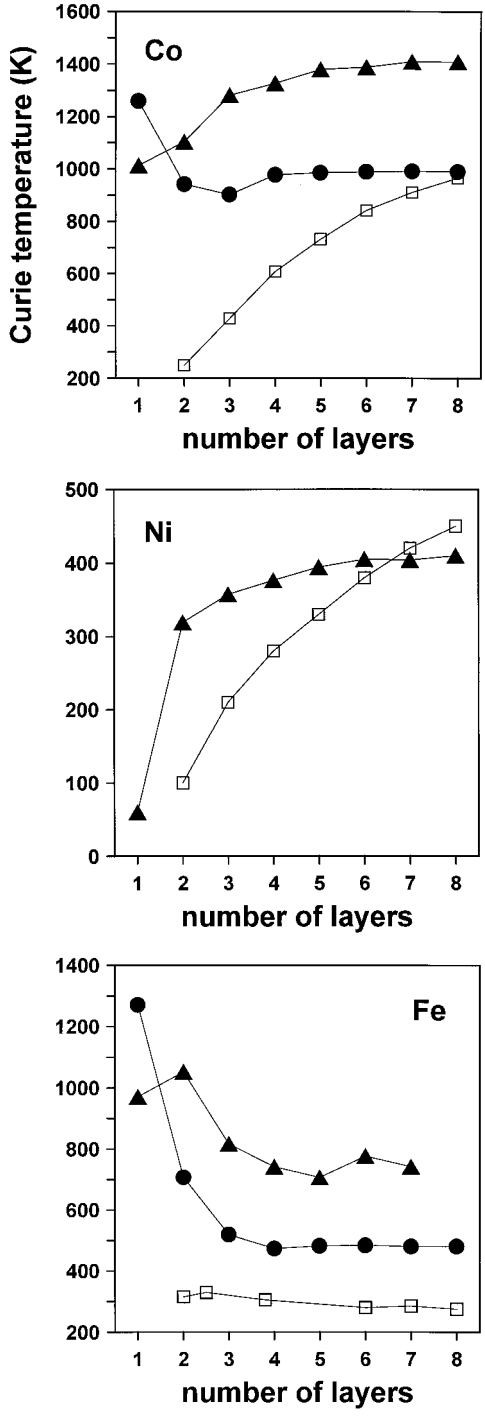

Figure 3. Curie temperatures for Co, Ni and Fe overlayers on Cu(001). Open squares depict experimental data (for Co and Ni by Wu *et al.* (1996)); for Fe by Detzel *et al.* (1996), while all other symbols display results of the mean-field approach by using Heisenberg parameters as calculated from the DLM state (filled circles) and from the ordered ground state (filled triangles). Solid lines serve as a guide for the eyes.

tions derived from the ordered state are larger in magnitude than those calculated relying on the DLM state, for Co films a similar relation between the corresponding T_C's can be seen. For Fe films on Cu(001) a maximum of T_C at a thickness of slightly above 2 ML was reported by Detzel *et al.* (1996). The theoretical T_C's calculated with parameters from the ordered state, in agreement with the results of Spišák and Hafner (1997b), clearly reproduce this observation. In our calculations, this effect can be associated with an increasing weight of antiferromagnetic exchange interactions for $n \geq 3$. It is, however, puzzling why (especially for thicker Co and Fe films) the T_C's calculated with parameters from the disordered state fit better in magnitude to the measured values than those derived from the ordered state. Therefore, the present work cannot establish preference to any of the two methods to calculate exchange pair interactions. Possibly, application of more sophisticated methods of the statistical physics and also calculations of further physical quantities (magnetic susceptibility, excitation energies) can do that.

Acknowledgements

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