Oscillatory Curie Temperature of Two-Dimensional Ferromagnets

M. Pajda,1 J. Kudrnovský,1,2 I. Turek,3 V. Drchal,2 and P. Bruno1

1Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany
2Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, CZ-18221 Prague 8, Czech Republic
3Institute of Physics of Materials, Academy of Sciences of the Czech Republic, Žižkova 22, CZ-61662 Brno, Czech Republic

(Received 27 July 2000)

The effective exchange interactions of magnetic overlayers Fe/Cu(001) and Co/Cu(001) covered by a Cu-cap layer of varying thickness were calculated in real space from first principles. The effective two-dimensional Heisenberg Hamiltonian was constructed and used to estimate magnon dispersion laws, spin-wave stiffness constants, and overlayer Curie temperatures within the mean-field and random-phase approximations. Overlayer Curie temperature oscillates as a function of the cap-layer thickness in a qualitative agreement with a recent experiment.

PACS numbers: 75.10.–b, 71.15.–m, 75.30.Ds, 75.70.Ak

The Curie temperature is one of the most important characteristics of ferromagnets. In particular, the Curie temperature of low-dimensional systems such as ultrathin films is of considerable interest. In a recent experimental study, Vollmer et al. [1] have shown that (i) the Curie temperature of fcc(001)-Fe ultrathin films on a Cu(001) substrate is considerably modified upon coverage by a Cu-cap layer, and (ii) that it varies in a nonmonotonous manner as a function of the Cu-cap-layer thickness, which indicates an oscillatory variation. An oscillatory behavior of the Curie temperature as a function of the spacer thickness was also found for fcc(001)-Co/Cu/Ni trilayers [2]. Such a behavior clearly cannot be explained within a localized picture of magnetism and calls for a first-principles theory of the Curie temperature in itinerant ferromagnets. In spite of considerable efforts in the past decades a first-principles calculation of the Curie temperature in the framework of itinerant magnetism, in particular, for low-dimensional systems, remains a very serious challenge.

One therefore has to rely upon some approximation schemes in order to calculate the Curie temperature of itinerant ferromagnets. A particularly simple and yet accurate approach consists of a mapping of the complicated itinerant ferromagnets. A particularly simple and yet accurate approximation. Overlayer Curie temperature oscillates as a function of the cap-layer thickness in a qualitative agreement with a recent experiment.

The effective exchange interactions of magnetic overlayers Fe/Cu(001) and Co/Cu(001) covered by a Cu-cap layer of varying thickness were calculated in real space from first principles. The effective two-dimensional Heisenberg Hamiltonian was constructed and used to estimate magnon dispersion laws, spin-wave stiffness constants, and overlayer Curie temperatures within the mean-field and random-phase approximations. Overlayer Curie temperature oscillates as a function of the cap-layer thickness in a qualitative agreement with a recent experiment.

PACS numbers: 75.10.–b, 71.15.–m, 75.30.Ds, 75.70.Ak

The electronic structure of the system was determined in the framework of the first-principles tight-binding linear muffin-tin orbital method (TB-LMTO) generalized to surfaces [12]. In the framework of the magnetic force theorem [3,13], the expression for the EEIs between two sites i and j anywhere in the system is [3,14]

\[ J_{ij} = \frac{1}{4\pi} \int_{\mathcal{C}} \text{Im} \, \text{tr}_L \{ \delta_i(z) g_{ij}(z) g_{ji}(z) \} \, dz \, . \]  

Here \( \text{tr}_L \) denotes the trace over the angular momentum \( L = (\ell m) \), \( \delta_i(z) = P_{ij}^L(z) - P_{ji}^L(z) \), and \( P_{ij}^L(z) \) are...
L-diagonal matrices of potential functions of the TB-LMTO method \((\sigma = 1,1)\), energy integration is performed in the upper half of the complex energy plane over a contour \(C\) starting below the bottom of the valence band and ending at the Fermi energy, and \(g_{ij}^s(z)\) are the site off-diagonal blocks of the system Green function corresponding to a given geometry. Possible lattice and/or layer relaxations at the overlayer are neglected. The intersite Green functions \(g_{ij}^s(z)\) can be evaluated either in the real space by using the cluster approach [3], the recursion method [8,15], or, as is done in the present paper and in Ref. [7], by the Bloch transformation which employs the two-dimensional translational symmetry of a given layer (for more details concerning the computational method, see Ref. [12]). We have calculated the EEI pairs \(J_{ij}\) up to 101 shells of the fcc(001) surface (i.e., up to the distance of about 10\,\text{a}, where \(a\) is the lattice constant of the fcc lattice). Such a large number of the EEIs is needed, in particular, for an accurate estimate of the spin-wave stiffness constant in a real space, as is also known for the bulk case [9,14]. In actual calculations the sites \(i,j\) were limited to the magnetic layer, which is a good approximation in view of the smallness of the moments induced in the Cu. The spin-wave spectrum \(E(q)\), the spin-wave stiffness constant \(D\), and the Curie temperatures \(T_{c}^{\text{MFA}}\) and \(T_{c}^{\text{RPA}}\) are expressed, respectively, in terms of the EEIs as follows:

\[
E(q) = \frac{4\mu_B}{M} \sum_{i \neq 0} J_{0i} [1 - \exp(iq \cdot R_i)] + \Delta,
\]

\[
D = \frac{\mu_B}{M} \sum_{i \neq 0} J_{0i} R_{0i}^2, \quad k_B T_{c}^{\text{MFA}} = \frac{2}{3} \sum_{i \neq 0} J_{0i},
\]

\[
\frac{1}{k_B T_{c}^{\text{RPA}}} = \frac{6\mu_B}{M} \frac{1}{N_{||}} \sum_{q_i} \frac{1}{E(q_i)}.
\]

Here, \(N_{||}\) is the number of sites per layer, the \(q_{||}\) sum extends over the fcc(001) surface Brillouin zone, \(\mu_B\) is the Bohr magneton, \(R_{0i} = |R_0 - R_i|\) is the interatomic distance, \(M\) is the magnetic moment per atom, and \(\Delta\) is the magnetic anisotropy energy. It should be noted that \(T_{c}^{\text{MFA}}\) can be evaluated directly by using the one-site rotation term \(J\) (expressed in terms of the site-diagonal element of the magnetic layer Green function similarly as its bulk counterpart [3,9]). The expression for \(T_{c}^{\text{RPA}}\) is a generalization of the bulk counterpart [10] to the case of magnetic layers: a vanishing \(T_{c}^{\text{RPA}}\) is obtained for \(\Delta = 0\) [16] in agreement with the Mermin-Wagner theorem [17] and small relativistic effects have to be considered in order to obtain a nonvanishing value of \(T_{c}^{\text{RPA}}\). The anisotropy energy \(\Delta\) is taken here as an adjustable parameter. This is not a serious problem as the RPA Curie temperature has only a weak logarithmic dependence upon \(\Delta\) [16], and it is thus sufficient to know the order of magnitude of \(\Delta\). The latter is typically of the order of the dipolar energy \(2\pi M^2/V\), where \(V\) is the atomic volume. In calculations we used \(\Delta_{\text{Co}} = 0.052\,\text{mRy}\) and \(\Delta_{\text{Fe}} = 0.140\,\text{mRy}\).

The evaluation of \(T_{c}^{\text{RPA}}\) is facilitated by the observation that it is proportional to the real part of the magnon Green function \(G_m(z) = \sum_{q_i} [z - E(q_i)]^{-1}\) corresponding to a dispersion law \(E(q_i)\) and evaluated at \(z = 0\). The corresponding \(q_{||}\) summation is performed for complex energies from which the value at \(z = 0\) is obtained by an analytic continuation technique [18]. The sum for the evaluation of the spin-wave stiffness constant is nonconvergent due to the RKKY character of magnetic interactions in metallic systems, and to overcome this difficulty we have calculated it by a regularization procedure [19] described in detail in Ref. [9].

The calculated EEIs for the first ten shells, magnetic moments, spin-wave stiffness constants, and the RPA and MFA Curie temperatures are summarized in Tables I and II for three limiting cases of magnetic Fe and Co layers, namely, the freestanding fcc(001) layer, the overlayer on the fcc-Cu(001) substrate, and the fcc(001) layer embedded in the fcc-Cu host.

| Table I. Calculated values of effective exchange interactions for the first ten shells of fcc(001) Fe and Co magnetic layers: a freestanding (fs) layer, an overlayer (ov) on fcc(001)-Cu, and an embedded (em) layer in fcc-Cu. Numbers of atoms in a given shell and the corresponding shell radii (in units of lattice constants) are given in parentheses and square brackets, respectively. Corresponding values of magnetic moments \(M\) are also given. |
|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| Shell       | fs     | \(J_{(Co)}\) [mRy] | ov     | em     | fs     | \(J_{(Fe)}\) [mRy] | ov     | em     | M [\(\mu_B\)] |
| 1 (4) [0.71]| 2.85   | 2.34   | 2.01   | 3.40   | 2.69   | 2.62   |
| 2 (4) [1.00]| 0.24   | 0.14   | -0.12  | 0.12   | 0.22   | 0.39   |
| 3 (4) [1.41]| -0.02  | -0.06  | -0.01  | -0.39  | -0.37  | -0.30  |
| 4 (8) [1.58]| -0.03  | 0.05   | 0.03   | -0.23  | 0.15   | 0.05   |
| 5 (4) [2.00]| 0.03   | 0.01   | -0.01  | 0.05   | 0.03   | 0.04   |
| 6 (4) [2.12]| -0.01  | -0.07  | -0.07  | -0.01  | 0.15   | 0.20   |
| 7 (8) [2.24]| -0.02  | 0.00   | 0.00   | 0.09   | -0.03  | -0.07  |
| 8 (8) [2.55]| 0.00   | 0.00   | 0.00   | 0.01   | 0.04   | 0.03   |
| 9 (4) [2.83]| 0.00   | 0.04   | 0.05   | -0.03  | -0.03  | -0.01  |
| 10 (8) [2.92]| 0.02  | -0.02  | -0.03  | 0.01   | 0.07   | 0.04   |
| \(M[\mu_B]\)| 2.22   | 1.79   | 1.57   | 3.06   | 2.82   | 2.59   |
Concerning the EEIs, the following general conclusions can be drawn: (i) A pronounced dependence of magnetic moments on the coordination number is found, namely, their decrease with increasing number of nearest neighbors, the effect being stronger for the Fe layer; (ii) the EEIs are significantly enhanced (typically by a factor of 2 or more) as compared to their bulk counterparts; (iii) the EEIs depend strongly upon the presence of a substrate and a capping layer. The latter dependence is due to the RKKY character of the EEIs in metals: the coupling is not only mediated through the magnetic layer itself but also via the substrate and capping layer. This behavior is also clearly visible on the spin-wave spectra shown in Fig. 1 for Fe and on the exchange stiffness constant (Table II). We also present corresponding magnon densities of states (DOS) determined from the magnon Green function $G_m(z)$. A characteristic step of the height proportional to $1/D$ at the bottom of the magnon DOS accompanied by a pronounced van Hove singularity in the middle of the band are typical features of the two-dimensional bands with the nearest-neighbor interactions which are here only slightly modified by nonvanishing interactions in next shells. Interestingly, the spin-wave stiffness constants and Curie temperatures behave differently as a function of the atomic coordination for Co and Fe layers, i.e., for cases of the freestanding layer, the overlayer, and the embedded layer. This behavior can be related to the values of leading EEIs in both cases, in particular, to large antiferromagnetic couplings of 3rd and 4th nearest neighbors of Fe-based layers which effectively reduce the value of the spin-wave stiffness constant [see Eq. (2)], in particular, for the freestanding layer. On the contrary, the Co-based EEIs have the prevailing ferromagnetic character giving thus increasing spin-wave stiffness constants due to increasing values of the EEIs with reduced atomic coordination. The antiferromagnetic character of the EEIs for fcc-based Fe layers is strongly enhanced as compared to the bcc-Fe case [9] while the prevailing ferromagnetic character of the EEIs for bulk fcc-Co [9] and for fcc-Co layers remains unchanged.

The MFA Curie temperatures are typically of the same order magnitude as the corresponding bulk ones due to the fact that the reduced coordination is approximately compensated by the increase of the EEIs. This observation is in a strong disagreement with experimental data: this failure is due to the fact that the MFA violates the Mermin-Wagner theorem due to the neglect of collective transverse fluctuations (spin waves) and it is thus inappropriate for two-dimensional systems.

The RPA Curie temperatures as a function of the anisotropy energy $\Delta$ are shown in Fig. 2 for cases of the Co overlayer on fcc-Cu (001) and fcc-Co(001) layers embedded in Cu. The weak logarithmic dependence of $T_c^{\text{RPA}}$ on $\Delta$ [16] is obvious: $T_c^{\text{RPA}}$ varies by about 25% as $\Delta$ varies by an order of magnitude so that the results are not significantly influenced by our semiempirical choice of $\Delta$. The RPA Curie temperatures are strongly reduced as compared to the corresponding bulk values thereby improving on the MFA results. Nevertheless, they are still too large as compared to observed Curie

![FIG. 1. Magnon dispersion laws (left frame) and corresponding densities of states [in states/(meV \cdot atom)] for the Fe layer embedded in fcc-Cu (full line), Fe overlayer on fcc-Cu(001) (dashed line), and the freestanding Fe layer (dash-dotted line). We have set here $\Delta = 0$.](image1)

![FIG. 2. RPA Curie temperatures of the Co overlayer on fcc-Cu(001) (full squares) and of the fcc(001)-Co layer embedded in fcc-Cu (full circles) as a function of the magnetic anisotropy energy $\Delta$. Note the logarithmic scale on the abscissa.](image2)
temperatures of ferromagnetic monolayers (being of the order of 150–200 K). It is unclear whether this is due to some inaccuracy of the theory or to some imperfections of the samples used in the experiments. On the contrary, such important experimental facts as the strong influence of the metallic coverage on the Curie temperature [1] are well explained by our theory as illustrated in Fig. 3. The oscillatory character of $T^\text{RPA}_c$ around the value corresponding to an infinite cap, i.e., to the limit of the embedded layer, is clearly visible and is in a qualitative agreement with the recent experiment of Vollmer et al. [1]. The origin of these oscillations can be traced back to the oscillatory behavior of the EEIs and it has the same origin as related oscillations of the interlayer exchange interactions for the Co/Cu-Co(001) trilayer with a varying Cu-cap-layer thickness [20]. These oscillations are due to quantum-well states in the Cu-cap layer formed between the vacuum and the magnetic layer which, in turn, influence properties of the magnetic layer. We have verified that amplitudes of oscillations of the EEIs decay with the thickness $d$ of the cap layer approximately as $d^{-2}$. The same thickness dependence was also found for the related case of the interlayer exchange interactions for the Co/Cu-Co trilayer with the varying thickness of the Cu-cap layer [21]. A similar behavior was also verified for the oscillatory dependences of $T^\text{RPA}_c$ and $T^\text{MFA}_c$ which, in turn, are derived from the EEIs. It should be noted that amplitudes and phases of oscillations can be influenced by the thickness of the magnetic layer and/or the presence of the disorder in the system.

In conclusion, in view of the interpretation proposed here, the oscillatory behavior of the Curie temperature of Fe films as a function of the Cu-cap thickness as reported by Vollmer et al. [1] would constitute the first direct experimental evidence of the oscillatory RKKY character of exchange interactions in itinerant ferromagnets.

J.K., I.T., and V.D. acknowledge the financial support provided by the Grant Agency of the Czech Republic (No. 202/00/0122), the Grant Agency of the Academy of Sciences of the Czech Republic (No. A1010829), and the Czech Ministry of Education, Youth and Sports (No. OC P3.40 and No. OC P3.70).

[19] The procedure consists of introducing an exponential damping factor $\exp(-\eta R_0/\alpha)$ which makes the sum absolutely convergent and subsequently taking the limit $\eta \rightarrow 0$. The error bars for $D$ in Table II correspond to the uncertainty due to this latter step.