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Oscillatory Curie temperature of 2D-ferromagnets

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Abstract

The effective exchange interactions of the magnetic overlayer Fe/Cu(0 0 1) 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 spin-wave stiffness constants and overlayer Curie temperatures within the random-phase approximation. Oscillatory behavior of the overlayer Curie temperature, spin-wave stiffness, and magnetic moment as a function of the cap-layer thickness was found and explained.

Keywords: Curie temperature, Heisenberg model, Spin waves—two dimensional, Multilayers—metallic

In recent experimental studies [1,2] it was shown that (i) the Curie temperature of FCC(0 0 1)-based magnetic ultrathin films on a Cu(0 0 1) substrate is considerably modified upon coverage by a Cu-cap layer, and (ii) it varies in a non-monotonous manner as a function of the Cu cap layer thickness indicating an oscillatory variation. Such a behavior 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 last 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 and one has to rely upon some approximation schemes. A simple and yet accurate approach consists in a mapping of the complicated itinerant electron system onto an effective Heisenberg model (EHM), $H = -\sum_{i \neq j} J_{ij}\ \mathbf{e}_i \cdot \mathbf{e}_j$, where $\mathbf{e}_i$ and $\mathbf{e}_j$ are the unit vectors of the magnetic moments at sites i and j, and the effective exchange interactions (EEIs) J_{ij} between any pair of magnetic moments are determined from first principles. This approach proposed by Liechtenstein [3] can be also generalized to low-dimensional systems which are of interest in this paper. The thermodynamic properties of the ferromagnet including determination of the Curie temperature can be then calculated from the EHM by using statistical mechanical methods. The success of this two-step approach relies upon the fact that it provides an almost exact description of low-lying magnetic excitations (spin-waves) which give the largest contribution to the Curie temperature. On the other hand this approach completely disregards longitudinal fluctuations of magnetic moments such as the Stoner excitations and therefore it is not suitable to describe ferromagnets with small exchange splitting such as, e.g., FCC-Ni, in which Stoner excitations with a rather low energy exist. We have recently applied this approach to bulk BCC-Fe, FCC-Co, and FCC-Ni and obtained a reasonable agreement with experimental Curie temperatures and spin-wave stiffness constants for Fe and Co, but not for Ni [4] similarly as in recent calculations based on the adiabatic spin-wave theory [5,6] or an alternative first-principles theory of spin fluctuations based on the linear-response theory [7].

In the present paper we wish to construct the two-dimensional EHM corresponding to monolayers of Fe and Co on FCC-Cu(0 0 1) and on its basis to investigate the influence of the capping layer on magnetic moments,

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spin-wave stiffness constants, and the Curie temperatures. The electronic structure of the system was determined in the framework of the first principles scalar-relativistic tight-binding linear muffin-tin orbital method (TB-LMTO) generalized to surfaces [8,9]. The expression for the EEIs between two sites i and j anywhere in the system is a direct generalization of the bulk counterpart [3]

$$J_{ij}^{(n)} = \frac{1}{4\pi}\,\mathrm{Im}\int_C \mathrm{tr}_L\left\{\delta_i^{(n)}(z)\,g_{ij}^{(n)\uparrow}(z)\,\delta_j^{(n)}(z)\,g_{ji}^{(n)\downarrow}(z)\right\}\mathrm{d}z\,. \quad (1)$$

Here tr_L denotes the trace over the angular momentum $L=(\ell m)$, $\delta_i^{(n)}(z)=P_i^{(n)\uparrow}(z)-P_i^{(n)\downarrow}(z)$ where $P_i^{(n)\sigma}(z)$ are L-diagonal matrices of potential functions of the TB-LMTO method ($\sigma=\uparrow,\downarrow$), 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 $g_{ij}^{(n)\sigma}\times(z)$ are the site off-diagonal blocks of the system Green function corresponding to a given geometry. It should be noted that all quantities in Eq. (1) depend on the number of cap-layers n (marked by the superscript).

We have calculated the EEI pairs $J_{ij}^{(n)}$ up to 101 shells of the FCC(0 0 1) surface for each cap-layer thickness $n\in(0,15)$ (i.e., up to the distance of about $10\,a$, 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 it is also known for the bulk case [6,4]. The spin-wave spectrum $E^{(n)}(\mathbf{q}_\parallel)$, the spin-wave stiffness constant $D^{(n)}$, and the Curie temperature $T_c^{(n)}$ evaluated in the framework of the RPA are expressed, respectively, in terms of the EEIs as follows

$$E^{(n)}(\mathbf{q}_\parallel)=\Delta^{(n)}+\frac{4\mu_B}{M^{(n)}}\sum_{i\neq 0}J_{0i}^{(n)}\left(1-\exp(i\mathbf{q}_\parallel\cdot\mathbf{R}_i)\right),$$

$$D^{(n)}=\frac{\mu_B}{M^{(n)}}\sum_{i\neq 0}J_{0i}^{(n)}R_{0i}^2,$$

$$\frac{1}{k_B T_c^{(n)}}=\frac{6\mu_B}{M^{(n)}}\frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\frac{1}{E^{(n)}(\mathbf{q}_\parallel)}. \quad (2)$$

Here, the $\mathbf{q}_\parallel$-sum extends over the FCC(0 0 1) surface Brillouin zone (SBZ), $N_\parallel$ is the number of sites in the SBZ, μ_B is the Bohr magneton, $R_{0i}=|\mathbf{R}_0-\mathbf{R}_i|$ is the interatomic distance, $M^{(n)}$ is the layer magnetic moment per atom, and $\Delta^{(n)}$ is the magnetic anisotropy energy. The expression for $T_c^{(n)}$ is a generalization of the bulk counterpart [10,11] to the case of magnetic monolayers covered by varying thickness of the non-magnetic cap: a vanishing $T_c^{(n)}$ is obtained for $\Delta^{(n)}=0$ in an agreement with the Mermin–Wagner theorem [12,13] and small relativistic effects have to be considered in order to obtain a non-vanishing value of $T_c^{(n)}$. The anisotropy energy $\Delta^{(n)}$ is taken here as an adjustable parameter although it could be determined also from first principles. The RPA Curie temperature has only a weak logarithmic dependence upon Δ [13]. It is thus sufficient to know the order of magnitude of Δ which is typically of the order of the dipolar energy $2\pi[M^{(n)}]^2/V$, where V is the atomic volume (for Fe- and Co-monolayer is Δ of the order of 0.14 and 0.05 mRy, respectively). The sum for the evaluation of the spin-wave stiffness constant is non-convergent due to the RKKY character of magnetic interactions in metallic systems and to overcome this difficulty we have calculated it by a regularization procedure [4].

The EEIs depend strongly upon the presence of a substrate and a capping layer [14]. In particular, the EEIs are significantly enhanced as compared to their bulk counterparts due to the reduced atomic coordination. For example, the leading first nearest-neighbor values of the EEIs for the BCC-Fe bulk, Fe(0 0 1) monolayer embedded in FCC Cu-bulk, and for uncovered FCC-Fe/Cu(0 0 1) overlayer are 1.43, 2.62, and 2.69 mRy, respectively. Related values for the FCC-Co bulk, Co(0 0 1) monolayer embedded in FCC-Cu host, and for uncovered FCC-Co/Cu(0 0 1) overlayer are 1.09, 2.01 and 2.34 mRy, respectively. More important for the present study is, however, the oscillatory behavior of the EEIs with the thickness of the capping layer (Fig. 1b). The reason for such behavior is the fact that the coupling is not only mediated through the magnetic layer itself but also via the substrate and the capping layer. This oscillatory behavior of the EEIs is a precursor of similar oscillatory behavior of spin-stiffness constants and Curie temperatures as it is obvious from

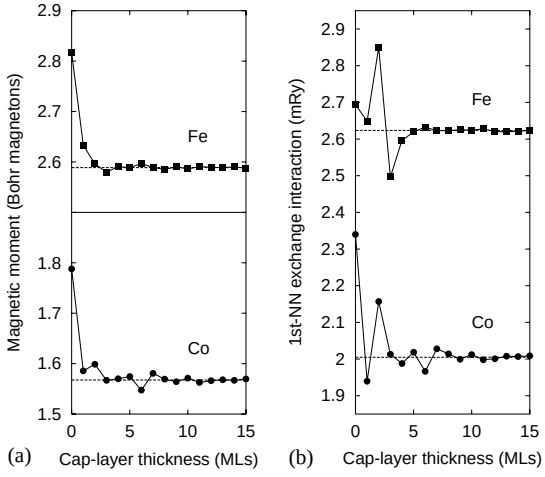

Fig. 1. The magnetic moment (a) and the first nearest-neighbor exchange interactions (b) of the Fe- and Co-monolayer on FCC-Cu(0 0 1) as a function of the cap-layer thickness. The dashed lines represent the embedded layer limit (infinite cap thickness) while the limit of zero cap thickness corresponds to the uncovered overlayer case.

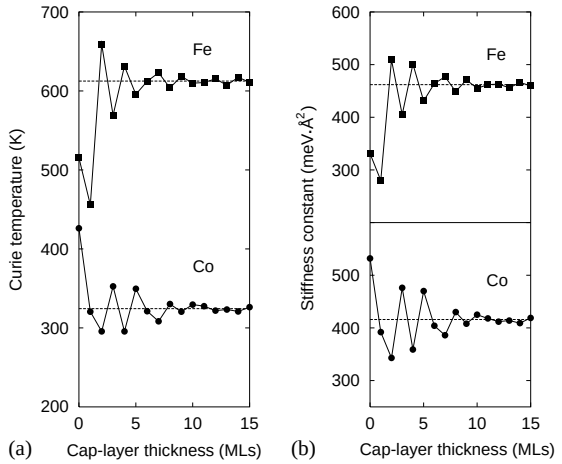

Fig. 2. The Curie temperatures (a) and the spin-stiffness constants (b) of the Fe- and Co-monolayer on FCC-Cu(0 0 1) as a function of the cap-layer thickness. The dashed lines represent the embedded layer limit (infinite cap thickness) while the limit of zero cap thickness corresponds to the uncovered overlayer case.

Eq. (2). This is illustrated in Fig. 2a and b where we plot Curie temperatures and spin-stiffness constants as a function of the cap layer thickness. It should be noted that the RPA Curie temperatures are strongly reduced as compared to the mean-field approximation (MFA) Curie temperatures thereby improving comparison with the experiment (the MFA values are of order of 1000 K). 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. Nevertheless, the RPA Curie temperatures are still too large as compared to typical observed Curie temperatures of ferromagnetic monolayers which are of order 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 experiments. On the contrary, such important experimental facts as the strong influence of the metallic coverage on the Curie temperature as well as the oscillatory character of T_c^{RPA} around the value corresponding to an infinite cap, i.e., to the limit of the embedded layer, are well explained by our theory as illustrated in Fig. 2a. The fluctuations of the Curie temperature are of order of 50–70 K again in a reasonable agreement with experiment [1,2]. Present calculations also predict oscillations of spin-wave stiffness constants as a function of the cap-layer thickness, Fig. 2a. A similarity between oscillatory behavior of Curie temperatures and spin-stiffness constants is obvious and it can be again related to an oscillatory behavior in the EEIs as a function of the Cu-cap thickness, Fig. 1b. The quantum-mechanical reason of the oscillatory behavior of the EEIs is the formation of quantum-well states in the cap layer: the barrier is formed on one side by the vacuum and on the other side by the effective barrier due to very different electronic structure of the minority Fe- or Co-bands as compared to Cu-bands of the cap-layer. The position of these quantum states varies with the cap thickness which in turn causes similar changes in the electronic structure and of the EEIs. Finally, in Fig. 1a we illustrate an oscillatory behavior of monolayer magnetic moments as a function of the cap-layer thickness. The physical origin of these oscillations is the same, namely the quantum-size effect in the cap layer.

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References

[1] R. Vollmer, S. van Dijken, M. Schleberger, J. Kirschner, Phys. Rev. B 61 (2000) 1303.
[2] A. Ney, F. Wilhelm, M. Farle, P. Poulopoulos, . Srivastava, K. Baberschke, Phys. Rev. B 59 (1999) R3928.
[3] A.I. Liechtenstein, M.I. Katsnelson, V.P. Antropov, V.A. Gubanov, J. Magn. Magn. Mater. 67 (1987) 65.
[4] M. Pajda, J. Kudrnovský, V. Drchal, I. Turek, P. Bruno, Phys. Rev. B, in press and arXiv: cond-mat/0007441.
[5] S.V. Halilov, H. Eschrig, A.Y. Perlov, P.M. Oppeneer, Phys. Rev. B 58 (1998) 293.
[6] V.P. Antropov, B.N. Harmon, A.N. Smirnov, J. Magn. Magn. Mater. 200 (1999) 148.
[7] J.B. Staunton, B.L. Gyorffy, Phys. Rev. Lett. 69 (1992) 371.
[8] I. Turek, V. Drchal, J. Kudrnovský, M. Šob, P. Weinberger, Electronic Structure of Disordered Alloys, Surfaces and Interfaces, Kluwer, Boston, 1997.
[9] I. Turek, J. Kudrnovský, V. Drchal, Electronic Structure and Physical Properties of Solids, in: H. Dreyssé (Ed.), Lecture Notes in Physics, Vol. 535, Springer, Berlin, 2000, p. 349.
[10] S.V. Tyablikov, Methods of Quantum Theory of Magnetism, Plenum Press, New York, 1967.
[11] C.S. Wang, R.E. Prange, V. Korenman, Phys. Rev. B 25 (1982) 5766.
[12] N.D. Mermin. H. Wagner, Phys. Rev. Lett. 17 (1966) 1133.
[13] P. Bruno, Mater. Res. Soc. Symp. Proc. 321 (1992) 299.
[14] M. Pajda, J. Kudrnovský, V. Drchal, I. Turek, P. Bruno, Phys. Rev. Lett. 85 (2000) 5424.