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INFLUENCE OF SURFACE AMORPHIZATION ON PHASE DIAGRAMS OF THIN FERROMAGNETIC FILMS[1]

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The phase diagrams of thin ferromagnetic films with the surface amorphization are studied using the Kirkwood approximation that partly takes into acount thermal fluctuations. A discrete Handrich–Kaneyoshi distribution for the surface exchange integrals is assumed. The phase diagrams for the film of the f.c.c. lattice with the (100) face and various surface magnetic moments are presented.

1. Introduction

The determination of the magnetic proporties of ferromagnetic thin films, sandwiches, and superlattices with a defined thickness d has become a very active field recently [1] – [3]. In general the critical temperature is determined by local quantities like the exchange coupling J and the magnetic moments μ as well as by the global topology of the film. In the ordered ferromagnetic thin films Curie temperature $T_C(d)$ may reach the bulk Curie temperature T_C^b within a few atomic layers, or may even exceed T_C^b exhibiting a maximum at small film thicknesses [4], [5]. $T_C(d)$ is not only determined by the reduced coordination number, but depends also strongly on the surface/interface magnetic moments μ_S which may deviate from the bulk value μ_b as well as on the global topology of the film. A number of theoretical studies obtained the possibility of an enhancement of μ_S by 20%–30% relative to μ_b for Fe free surfaces and interfaces and for Ni free surfaces [1],[6]. Due to the roughness or intermixing effects μ_S may even vanish ("dead layer").

Recently amorphous films described by the so–called lattice model with a bond disorder [7],[8], have been examined. The purpose of this paper is to present the phase diagrams of thin ferromagnetic films, consisting of d atomic layers, with surface amorphization, taken into account: the surface exchange integral fluctuates obeying a discrete Handrich–Kaneyoshi (H–K) distribution [9].

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2. Method

In this paper, we will use a generalized mean–field theory in which thermal magnetic fluctuations are partly taken into account by so–called Kirkwood approximation [10]. Quite generaly, if one writes the total Hamiltonian as

$$H = H_0 + V \tag{1}$$

where H_0 is the free Hamiltonian and V is the interaction term, then the exact partition function Q can be written in terms of the reference partition function Q_0 and an average in the reference system,

$$\begin{aligned} Q &= \mathrm{Tr} e^{-\beta(H_0+V)} = \mathrm{Tr} e^{-\beta H_0} \frac{\mathrm{Tr} e^{-\beta H_0} e^{-\beta V}}{\mathrm{Tr} e^{-\beta H_0}}, \\ Q &= Q_0 \left\langle e^{-\beta V} \right\rangle_0 \end{aligned}$$

which sets up the free energy perfectly to be expanded in cumulants. Defining the relevant cumulants

$$\ln \left\langle e^{-\beta V} \right\rangle_0 = \ln \left[\sum_{n=0}^{\infty} \frac{1}{n!} (-\beta)^n \left\langle V^n \right\rangle_0 \right] = \sum_{n=1}^{\infty} \frac{1}{n!} (-\beta)^n C_n(V) \tag{2}$$

we are quickly enable to write the free energy as desired

$$-\beta F = \ln Q = -\beta F_0 + \sum_{n=1}^{\infty} \frac{1}{n!} (-\beta)^n C_n(V). \tag{3}$$

The Kirkwood's method used the formalism of semi–invariants to effect an expansion in powers $1/kT$, valid at high temperature. In practice, the phrase "mean–field theory" usually means to take this expansion and truncate it at the $n = 1$ level. However, whith the exeption of the first cumulant, which is simply the average interaction energy,

$$C_1(V) = \langle V \rangle_0 , \tag{4}$$

the cumulnats represent the fluctuations

$$C_2(V) = \left\langle (V - \langle V \rangle_0)^2 \right\rangle_0 = \langle V^2 \rangle_0 - \langle V \rangle_0^2 , \tag{5}$$

$$C_3(V) = \left\langle (V - \langle V \rangle_0)^3 \right\rangle_0 , \quad \text{etc.}, \tag{6}$$

so that one has to go to an order beyoned $n = 1$ to build any of the effects of fluctuations into the theory, besides those due to just noninteracting reference. In this language, what Kirkwood did, essentialy, was to truncate Eq. (3) at $n = 2$. Though his procedure still led to a mean–field theory in the sense of giving classical critical exponents (as would truncating at any finite order) [11], the inclusion of nontrivial fluctuation corrections turned out to give a significante quantitative improvement of the mean–field theory.

This approximation was applied to non–uniformly ordered phases (the axial next–nearest–neighbor Ising (ANNNI) model [10] and crystalline ferromagnetic thin films [12]). It simply requires us to replace single order parameters by a set of order parameters and to compute the necessary cumulants in terms of those new order parameters. This procedure gives us the desired set of equations for the layer magnetization.

The effective exchange coupling J between two magnetic moments μ_i and μ_j is approximately given by $\mu_i \mu_j J$. We assume that only the surface exchange interactions $\tilde{\mathrm{J}}_s$ fluctuate and are described by a discrete distribution function

$$P(\tilde{\mathrm{J}}_s) = \frac{1}{2} \left\{ \delta[\tilde{\mathrm{J}}_s - \mathrm{J}_s - \Delta \mathrm{J}_s)] + \delta[\tilde{\mathrm{J}}_s - \mathrm{J}_s + \Delta \mathrm{J}_s)] \right\}, \tag{7}$$

where $\Delta \mathrm{J}_s$ denotes the fluctuation of the surface exchange coupling about some mean value J_s. Inside the film, the middle planes are crystalline. The different lattice types and crystal faces are considered by the different coordination numbers z_0 and z_1 in the same layer and between adjacent layers, respectively. The bulk coordination number $z = z_0 + 2z_1$.

For Ising–1/2 spins we obtain a set of d nonlinear equations describing the layer magnetization of our layered system

$$m_1 = \tanh[KR(Rz_0 m_1 + z_1 m_2) - \Delta h_1] \tag{8}$$

$$m_2 = \tanh[K(z_0 m_2 + z_1 R m_1 + z_1 m_3) - \Delta h_2] \tag{9}$$

$$m_k = \tanh[K(z_0 m_k + z_1 m_{k+1} + z_1 m_{k-1}) - \Delta h_k] \tag{10}$$

$$m_{d-1} = \tanh[K(z_0 m_{d-1} + z_1 R m_d + z_1 m_{d-2} - \Delta h_{d-1}] \tag{11}$$

$$m_d = \tanh[KR(Rz_0 m_d + z_1 m_{d-1}) - \Delta h_d] \tag{12}$$

where

$$\begin{aligned} \Delta h_1 &= (KR)^2 m_1 [z_0 R^2 (1 + D_S^2)(1 - m_1^2) + z_1 (1 - m_2^2)] \\ \Delta h_2 &= K^2 m_2 [z_0 (1 - m_2^2) + z_1 R^2 (1 - m_1^2) + z_1 (1 - m_3^2)] \\ \Delta h_k &= K^2 m_k [z_0 (1 - m_k^2) + z_1 (1 - m_{k+1}^2) + z_1 (1 - m_{k-1}^2)] \\ \Delta h_{d-1} &= K^2 m_{d-1} [z_0 (1 - m_{d-1}^2) + z_1 R^2 (1 - m_d^2) + z_1 (1 - m_{d-2}^2)] \\ \Delta h_d &= (KR)^2 m_d [z_0 R^2 (1 + D_S^2)(1 - m_d^2) + z_1 (1 - m_{d-1}^2)] \end{aligned}$$

and $K = \beta \mathrm{J}, R = \mu_S / \mu_b$. The mean value of the surface interatomic exchange interaction J_s is assumed to be equal to the bulk one J and $D_S = \Delta K_s / K$ ($K_s = \beta \mathrm{J}_s$). Δh_n, is the correction due to the KA which is also known as the Onsager reaction field [13].

The condition for the Curie temperature results from the linearisation of eqs. (8) – (12). As a result of the linearization we obtain a set of linear equations:

$$\begin{aligned} [z_0 KR^2 &- (KR)^2 (z_0 R^2 (1 + D_S^2) + z_1) - 1] m_1 + z_1 KR m_2 = 0, \\ z_1 K m_{k-1} &+ [z_0 K - K^2 (z_0 + 2z_1) - 1] m_k + z_1 K m_{k+1} = 0, \\ z_1 KR m_{d-1} &+ [z_0 KR^2 - (KR)^2 (z_0 R^2 (1 + D_S^2) + z_1) - 1] m_d = 0. \end{aligned} \tag{13}$$

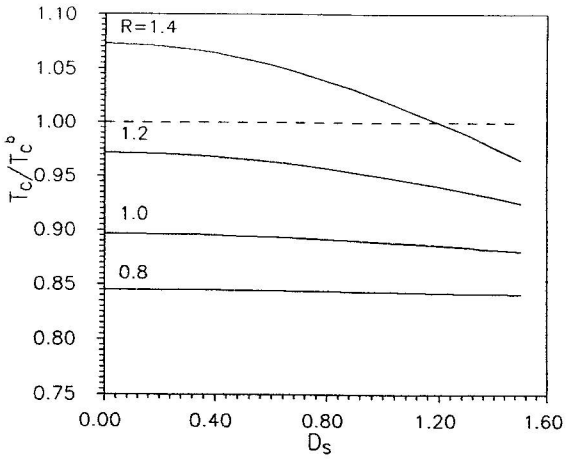

Fig. 1. The Curie temperature of the film reduced to the bulk (T_C/T_C^b) versus the surface amorphization parameter D_S for the film with thickness $d = 5$ and for the surface magnetic moments $R = \mu_s/\mu_b = 1.4; 1.2; 1.0$ and 0.8.

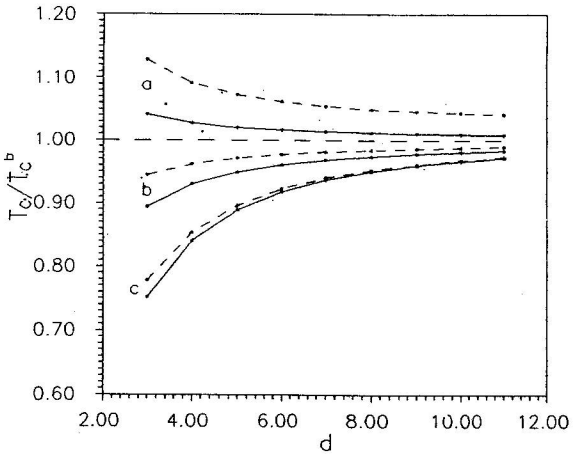

Fig. 2. The Curie temperature reduced to the bulk (T_C/T_C^b) for films with various thickness. The curves are ploted for surface magnetic moments $\mu_s = 1.4\mu_b; \mu_s = 1.2\mu_b$ and $\mu_s = 1.0\mu_b$. Dashed lines for crystaline films (surface amorphization parameter $D_S = 0.0$) and for films with the amorphous surfaces ($D_S = 1.0$).

The determinant of this set of equations should be equal to zero and this condition yields d different solutions for the temperature. The largest solution can be interpreted as the Curie temperature of the thin film [14].

3. Results and discussion

In fig. 1 the Curie temperature $T_C(d)$ of the film with thickness $d = 5$ in terms of the bulk Curie temperature $T_C^b = 10.8989$ for a face central cubic lattice (f.c.c.) with

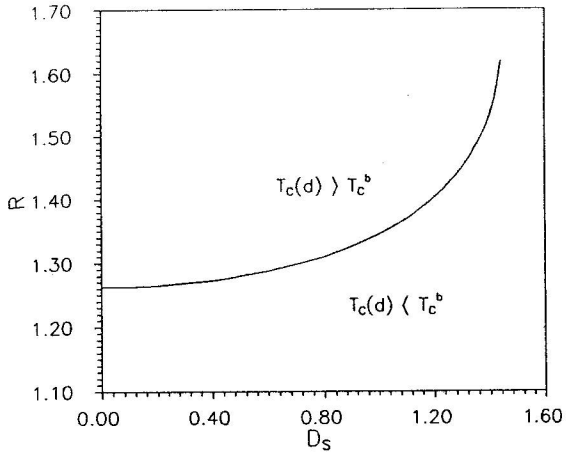

Fig. 3. The surface magnetic moment μ_s reduced to the bulk magnetic moment μ_b versus the surface amorphization D_S.

$z_0 = z_1 = 4$ and face (100) is presented as a function of the surface amorphization parameter D_S. The curves refer to different surface magnetic moment ($R = \mu_S/\mu_b$) as indicated in the figure. From this figure it can be concluded that for very small amorphization parameter the curves for $R = 1.4, 1.2, 1.0$ and 0.8 are tending to the Curie temperatures of the crystalline films. With increase of surface amorphization the Curie temperature decreases slowly to a limiting value that is lower than the Curie temperature of a crystalline lattice for all values of the surface magnetic moment.

In fig. 2 the Curie temperature reduced to the bulk are presented for films with various thicknesses d. Curves a, b and c correspond to the surface magnetic moment $\mu_s/\mu_b = 1.4, 1.2$ and 1.0, respectively. Full lines: surface amorphization parameter $D_S = 1.0$, dashed lines: crystalline lattice. A different behavior of $T_C(d)$ is seen due to variation of μ_S. For $\mu_S/\mu_b = 1.0$ and 1.2 the Curie temeperature increases when the film thickness increases. On the other hand, when $\mu_S/\mu_b = 1.4$, we note just the opposite tendency. $T_C(d)$ exceeds T_C^b despite the reduced number of nearest neighbors. Regarding the crystalline films this behavior has been obtained in [3].

It is obvious that when $\mu_S/\mu_b = 1.2$, 1.0, the Curie temperature is lower than the Curie temperature of the crystalline bulk, T_C^b. However, this lowering can be compensated by the appropriate enhancement of the surafce magnetic moment. In fig. 3 we present the reduced surafce magnetcic moment $R = \mu_S/\mu_b$, which is required for the compensation of the Curie temperature in order to obtain the bulk value. We note that this compensative enhancement depends on the suraface amorphization parameter D_S. The curve presented in fig. 3 (the critical isotherm) separates two regions: below, where the Curie temperature of a film is lower than that of the bulk, and above, where the Curie temperature is higher than T_C^b. The curve from fig. 3 is universal for any film thickness.

4. Conclusion

The purpose of this paper was to study the influence of surface amorphization on the Curie temperature of thin ferromagnetic films taking into account thermal magnetic fluctuations and different surface magnetic moments. We have obtained the new phase diagrams for the crystalline films with amorphous surfaces and with different surface magnetic moments.

The formalism presented here will also bevalid for the cases when a) the surafces and the middle planes are uniformly amorphous, b) the surfaces are crystalline and films remain amorphous inside (re–crystallisation).

References

[1] J.P.Reanard, P.Beauvillain: *Phys.Scr.* **T19** (1987) 405.

[2] H.C.Siegman: *J.Phys.: Condens. Matter* **4** (1992) 8395.

[3] P.J.Jensen, H.Dreysse, K.H.Bennemann: *Europhys. Lett.* **18** (1992) 463.

[4] M.Stampanoni, A.Vaterlaus, M.aeschlimann, F. Meier: *Phys. Rev. Lett.* **59** (1987)2483.

[5] M.Stampanoni: *Appl. Phys.* **A49** (1989) 449.

[6] S. Blügel, B.Drittler, R. Zeller, P.H. Dederichs: *Appl. Phys.* **A49** (1989) 547.

[7] T. Balcerzak, J. Mielnicki, A. Urbaniak-Kucharczyk, G.Wiatrowski: *Phys. Stat. Sol (b)* **164** (1991) 291.

[8] T. Balcerzak: *JMMM* **129** (1994) 279.

[9] T. Kaneyoshi: *Introduction to Surface Magnetism*. Chemical Rubber Company, Boca Raton, 1991.

[10] T.De Simone, R.M.Stratt: *Phys.Rev.* **B32** (1985) 1537.

[11] J. DeBoer: *Phys. Norv.* **5** (1971) 271.

[12] P.J.Jensen, H.Dreysse, K.H.Bennemann: *Surface Science* **269/270** (1992) 627.

[13] R. Brout, H. Thomas: *Physics* **3** (1967) 317.

[14] A.R. Ferchmin, S. Krompiewski: *J.Phys.* **C8** (1975) 1901.