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1992 J. Phys.: Condens. Matter 4 7997

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Ferromagnetic phase transition in ultra-thin films and surfaces

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Received 26 November 1991, in final form 12 May 1992

Abstract. The ferromagnetic phase transition in ultra-thin films and surfaces is studied by the Green function method based upon the Heisenberg model ($S \geq 1$) with the out-of-plane spin orientation of the atoms in the surfaces. A model calculation shows that the order parameter (spontaneous magnetization) changes with the atomic planes in the ferromagnetic films but the Curie temperatures of the atomic planes are all the same; the Curie temperature of ultra-thin films may be higher or lower than the bulk Curie temperature which depends on the fact that the spin is pinned to some degree at the surfaces. It is impossible for an ordered surface phase to coexist with a disordered bulk phase and it is also impossible for a disordered surface phase to coexist with an ordered bulk phase in homogeneous films or a bulk magnet.

1. Introduction

Recent studies have shown that the ferromagnetic phase transition in ultra-thin films and surfaces may differ markedly from that in the bulk. Experiments [1] have shown that in the general case the Curie temperature of ultra-thin films is lower than the bulk Curie temperature and, as the thickness of an ultra-thin film decreases, its Curie temperature also decreases while the value obtained by Curie temperature measurement strongly depends on the conditions of the actual film preparation (i.e. the surface situation). In some special cases the Curie temperature of ultra-thin films is higher than the bulk Curie temperature; this remarkable phenomenon was observed for Gd [2] and Tb [3]. For sufficient enhancement of the interaction between the surface spins, an ordered surface phase is expected to coexist with a disordered bulk phase [4].

In this work, we investigate the ferromagnetic phase transition in ultra-thin films and surfaces based on the Heisenberg model with an out-of-plane spin orientation of the atoms in the surfaces. We introduce a pinning spin model to describe the surface situation of the actual ultra-thin film, in order to interpret the magnetic behaviour of ultra-thin films. To study the magnetic theory of ferromagnetic films, the basic difficulty is that loss of translation invariance in one direction prevents decoupling of the motion of the spin into translational and relative coordinates. In [5], the spontaneous magnetizations of the atomic planes were assumed to be approximately equal and the analytical solution of coupling Green functions was obtained, but this is invalid just below the Curie temperature. In this paper, using the numerical calculation method of coupling Green equations [6] we remove the approximate

condition in [5] and obtain reasonable results for the physics. The expressions for the Curie temperature and order parameter (spontaneous magnetization) of atomic planes which are parallel to the surface of ultra-thin films have been derived. A model calculation shows that the order parameter changes with the position of the atomic planes in the ferromagnetic films but the Curie temperatures of the atomic planes are equal; the Curie temperature and the order parameter strongly depend on the fact that the spin is pinned to some degree at the surfaces and the thickness of the ultra-thin films. The experimental results could be explained by this model calculation.

2. Pinning spin model

Gay and Richter [7] calculated the spin anisotropy of monolayers of Fe, Ni, V and Co by incorporating the spin-orbit interaction into the self-consistent local-orbital method. They found that the easy direction of magnetization is perpendicular to the plane of the monolayer for Fe and V, but in the plane of the monolayer for Ni and Co. In terms of energy per atom, the monolayer anisotropies are large. For example, the anisotropy of the Fe monolayer (about 0.4 meV/atom) is 100 times the anisotropy of bulk Fe (4 μ eV/atom). This is a consequence of the reduced symmetry of the monolayers which includes the anisotropy as a second-order term. To explain the magnetic behaviour of ultra-thin films, the spin anisotropy of the surface must be considered [5]. We assume that the surfaces of n -monolayer (n -ML) films of a simple cubic crystal are parallel to (001) planes and perpendicular to the Z axis. The n -ML film has a finite amount of n atoms or atomic planes in the Z [001] direction and an infinite amount of N atoms ($N \rightarrow \infty$) in both the X [100] and the Y [010] directions. The spin Hamiltonian contains two parts: the Heisenberg exchange term and an anisotropic term due to the surfaces. To consider incorporating the spin-orbit interaction into the local orbit [7], an anisotropic term can be deduced by the method of second quantization [8]. The spin Hamiltonian is given by

$$\mathcal{H} = -\frac{1}{2} \sum_{\alpha f_1 f_2} J_{f_1 f_2} S_{f_1}^{\alpha} S_{f_2}^{\alpha} - \frac{1}{2} \sum_f D_f (S_f^z)^2$$

where the lattice site f stands for the position vector j in the X - Y plane and the coordinate ν in the Z direction: $f = (j, \nu)$. ν marks the atomic planes of the n -ML films ($\nu = 1, 2, 3, \dots, n$); α denotes X , Y and Z . For simplicity, the exchange interaction of only nearest-neighbour coupling ($J_{f_1 f_2} = J$) is taken into account. We assume that the n -ML films have a symmetrical surface and the spin orientation of the atoms in the surfaces is out of plane, i.e. the surface anisotropic constant (SAC) D_f takes the values $D_f = D_{\nu} = 0$ (for $\nu = 2, 3, \dots, n-1$) and $D_1 = D_n = D > 0$, which measures the fact that the spin is perpendicularly pinned to some degree at the surface. In order that this Hamiltonian may give the surface anisotropic term, it is necessary that $S \geq 1$, where S denotes the quantum number of the spin operator.

3. Curie temperature T_C and spontaneous magnetization σ_{ν} of each atomic plane

This section examines the relative magnetization σ_f at site f , $\langle S_f^z \rangle = \sigma_f S$, in which $\langle \rangle$ denotes the statistical average. We introduce the double-time Green function [9]

$$G_{f_1 f_2}(t-t') = \langle \langle S_{f_1}^+(t) | (S_{f_2}^z(t'))^n S_{f_2}^-(t') \rangle \rangle = \int_{-\infty}^{\infty} G_{f_1 f_2}(\omega) \exp[-i\omega(t-t')] d\omega.$$

To take into account the translation invariance along the plane (X, Y), a two-dimensional Fourier transform is introduced:

$$G_{f_1, f_2}(\omega) = \frac{1}{N^2} \sum_h \exp[i(j_1 - j_2, h)] G_{\nu_1, \nu_2}(\omega, h)$$

$$h_x = 2\pi n_x / N \quad h_y = 2\pi n_y / N$$

where the allowed values of n_x and n_y are $0, \pm 1, \pm 2, \dots, \pm N/2$ ($N \rightarrow \infty$); ν_1 and ν_2 take the values $1, 2, 3, \dots, n$. $G_{\nu_1, \nu_2}(\omega, h)$ is the energy-momentum coordinate representation of the Green function $G_{f_1, f_2}^2(t - t')$. Because of translation symmetry, σ_f and $\langle (S_f^z)^n S_f^- S_f^+ \rangle$ will depend only on the position ν of the atomic plane involved, so that $\sigma_f = \sigma_\nu$ and $\langle (S_f^z)^n S_f^- S_f^+ \rangle = \langle (S_\nu^z)^n S_\nu^- S_\nu^+ \rangle$. From the relation between the correlation function and the Green function, we now get [9]

$$\begin{aligned} \langle (S_\nu^z)^n S_\nu^- S_\nu^+ \rangle &= \frac{1}{N^2} \sum_h i \int_{-\infty}^{\infty} \left(\frac{1}{\exp(\omega/k_B T) - 1} \right. \\ &\quad \left. \times [G_{\nu\nu}(\omega + i\epsilon, h) - G_{\nu\nu}(\omega - i\epsilon, h)] \right) d\omega \end{aligned} \tag{1}$$

where k_B is Boltzmann's constant and T the absolute temperature.

From the equation of motion and the Tyablikov decoupling approximation for the Green function [9], we obtain the following coupled equations:

$$\sum_{\mu} (\omega \delta_{\nu_1\mu} - P_{\nu_1\mu}) G_{\mu\nu}(\omega, h) = \frac{\delta_{\nu_1\nu}}{2\pi} \langle [S_\nu^+, (S_\nu^z)^n S_\nu^-] \rangle \tag{2}$$

where

$$P_{\nu_1\mu} = [2JS\sigma_{\nu_1}(2 - \cos h_x - \cos h_y) + JS(\sigma_{\nu_1+1} + \sigma_{\nu_1-1})$$

$$+ D_{\nu_1} S\sigma_{\nu_1}] \delta_{\nu_1\mu} - S\sigma_{\nu_1} J_{\nu_1\mu}$$

$$J_{\nu_1\mu} = \begin{cases} J & \mu = \nu_1 \pm 1 \\ 0 & \mu \neq \nu_1 \pm 1 \end{cases}$$

$$D_{\nu_1} = \begin{cases} D & \nu_1 = 1 \text{ or } n \\ 0 & \nu_1 = 2, 3, \dots, n-1 \end{cases}$$

$$\sigma_{n+1} = \sigma_0 = 0.$$

We find from (2) that

$$G_{\nu\nu}(\omega, h) = \frac{\langle [S_\nu^+, (S_\nu^z)^n S_\nu^-] \rangle}{2\pi} \sum_k \frac{U_{\nu k}^2}{\omega - \omega_{hk}} \tag{3}$$

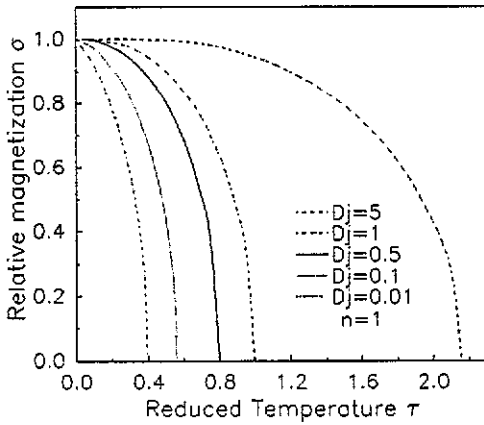


Figure 1. The curves of relative magnetization σ versus reduced temperature τ for various Dj in 1-ML films.

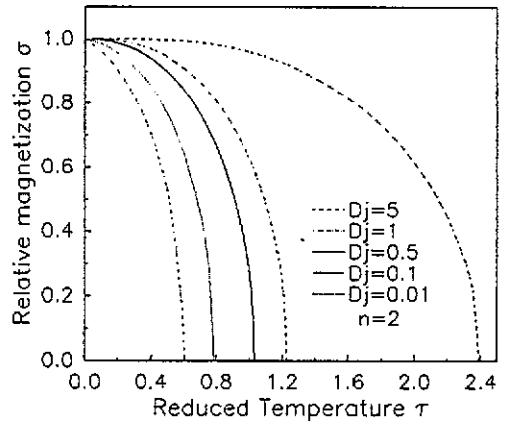


Figure 2. The curves of relative magnetization σ versus reduced temperature τ for various Dj in 2-ML films.

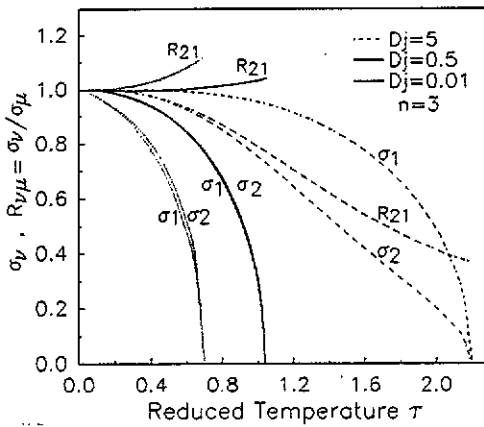


Figure 3. The curves of the relative magnetization σ_ν of each atomic plane and the ratio $R_{\mu\nu} = \sigma_\mu/\sigma_\nu$ as functions of reduced temperature τ for various Dj in 3-ML films.

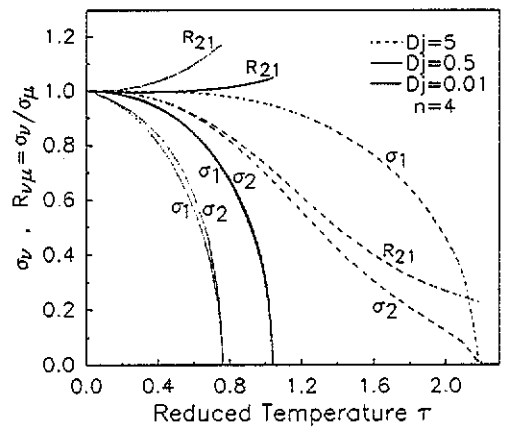


Figure 4. The curves of the relative magnetization σ_ν of each atomic plane and the ratio $R_{\mu\nu} = \sigma_\mu/\sigma_\nu$ as functions of reduced temperature τ for various Dj in 4-ML films.

in figures 1–6. For inequivalent atomic planes of n -ML films its σ_ν are different but its $T_{C\nu}$ are equal.

From (6) and (7) and the high-temperature approximation [9], we deduce the spontaneous magnetization σ_ν just below the Curie temperature $T_{C\nu}$:

$$\sigma_\nu = [\Gamma_\nu(1 - T/T_{C\nu})]^{1/2}$$

$$T_{C\nu} = [2S(S + 1)/C_\nu](J/k_B) \tag{8}$$

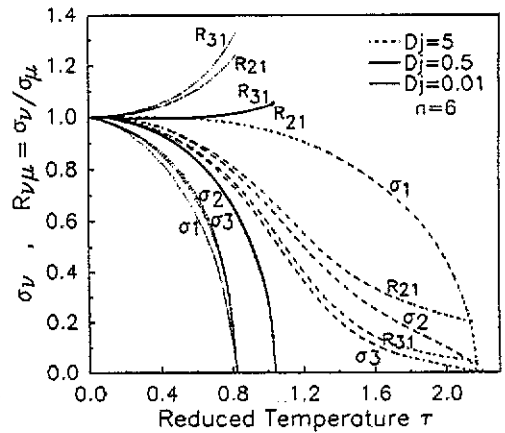
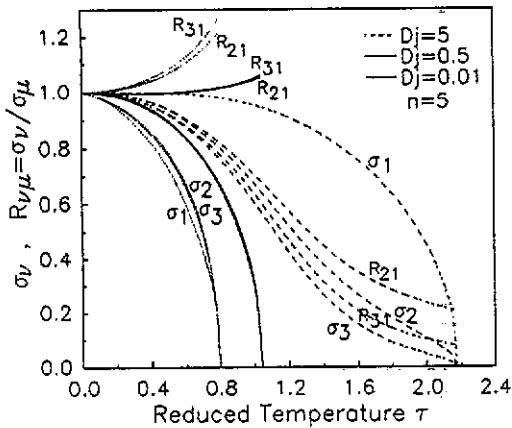


Figure 5. The curves of the relative magnetization σ_ν of each atomic plane and the ratio $R_{\mu\nu} = \sigma_\mu/\sigma_\nu$ as functions of reduced temperature τ for various D_j in 5-ML films.

Figure 6. The curves of the relative magnetization σ_ν of each atomic plane and the ratio $R_{\mu\nu} = \sigma_\mu/\sigma_\nu$ as functions of reduced temperature τ for various D_j in 6-ML films.

where

$$C_\nu = \frac{1}{(2\pi)^2} \int \int_{-\pi}^{\pi} dh_x dh_y \sum_{k=1}^n \frac{6S\sigma_\nu JU_{\nu k}^2}{\omega_{hk}} \tag{9}$$

$$D_\nu = \frac{1}{(2\pi)^2} \int \int_{-\pi}^{\pi} dh_x dh_y \sum_{k=1}^n \frac{\omega_{kh} U_{\nu k}^2}{6JS\sigma_\nu}$$

$$\Gamma_\nu = 20(S + 1)^2/3(4S^2 + 4S + 5C_\nu D_\nu - 3)$$

$$C_\nu = C_{n+1-\nu} \quad D_\nu = D_{n+1-\nu} \quad T_{C\nu} = T_{C_{n+1-\nu}} \quad (\nu = 1, 2, \dots, n).$$

Because ω_{hk}/σ_ν depends on σ_μ/σ_ν (see equation (4) and appendix), the convergent condition of integrals (9) requires that the values of σ_ν/σ_μ are finite and non-zero or σ_ν ($\nu = 1, 2, 3, \dots, n$) simultaneously approach zero at the Curie temperature T_C , i.e. the Curie temperatures $T_{C\nu}$ of atomic planes in n -ML films are equal:

$$T_C = T_{C1} = T_{C\nu} \text{ or } C_1 = C_\nu (\nu = 2, 3, \dots, n/2 \text{ or } (n + 1)/2). \tag{10}$$

To satisfy the condition (10) the Curie temperature T_C and the ratio of σ_1 to σ_ν ($\nu = 2, 3, \dots, n$) have been determined simultaneously by the use of (8) and (9), as shown in figures 7 and 8.

When the above-discussed different methods for determining the Curie temperature are compared, their results are consistent, as shown in figures 1-8.

4. Numerical calculation

For the bulk magnet of a single cubic lattice (corresponding to the n -ML films with $n \rightarrow \infty$) the Curie temperature T_{Cb} is given by

$$T_{Cb} = [2S(S + 1)/C](J/k_B) \tag{11}$$

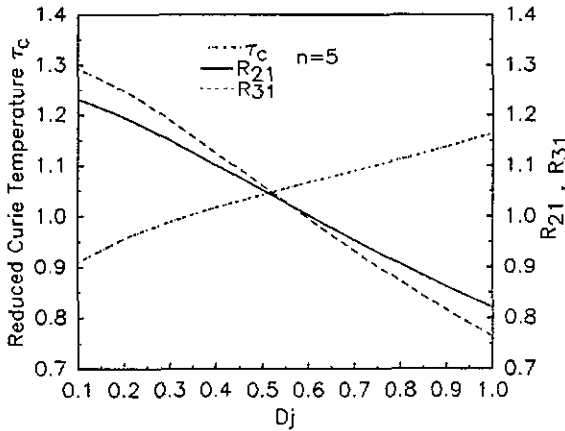


Figure 7. The curves of the reduced Curie temperature τ_C and the ratio $R_{\nu 1} = \sigma_\nu / \sigma_1$ as functions of D_j in 5-ML films.

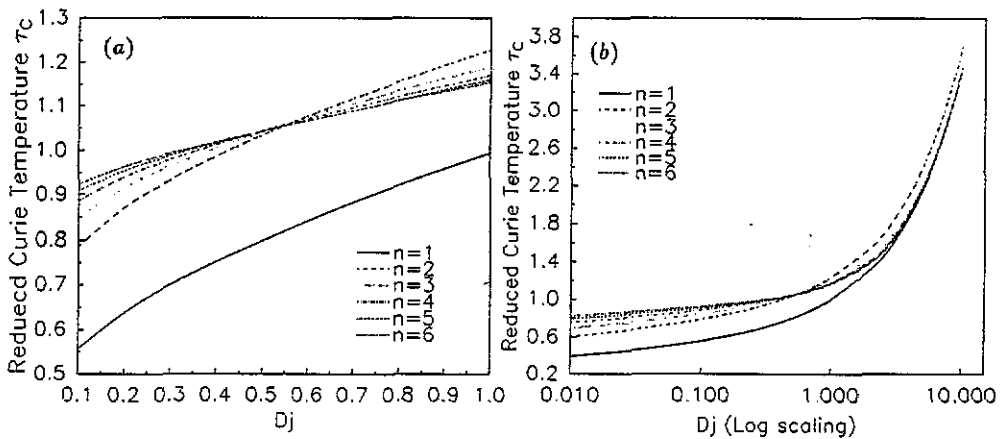


Figure 8. The D/J -dependence of the reduced Curie temperature τ_C for various thicknesses n in n -ML films.

where J is an exchange integral, k_B is Boltzmann's constant and C is a constant (equal to 1.516386).

Using a computer to perform the numerical calculation of the temperature dependence of the relative magnetization σ_ν and the Curie temperature, we let $S = 1$ and design the first computer program as follows. Because $\sigma_\nu = 1$ ($\nu = 1, \dots, n$) at 0 K, if the increment in temperature t and the initial values $\sigma_\nu = 1$ ($\nu = 1, \dots, n$) are substituted into (4), (6) and (7), the final values of $\sigma_\nu = \sigma_\nu(t)$ at the temperature t are given by iterative solution. Similarly, if the increment in temperature $2t$ and the initial values $\sigma_\nu = \sigma_\nu(t)$ ($\nu = 1, \dots, n$) are substituted into (4), (6) and (7), the final values of $\sigma_\nu = \sigma_\nu(2t)$ at the temperature $2t$ are given by iterative solution. We repeat the calculation at the temperatures $3t, 4t, \dots$ step by step in the above way until $\sigma_\nu = 0$ ($\nu = 1, 2, \dots, n$) at the Curie temperature. The relative magnetization σ_ν of each atomic plane and the ratio $R_{\mu\nu}$ as functions of the reduced temperature τ and the reduced Curie temperature τ_C are illustrated schematically in figures 1-6,

where $Dj = D/J$, $\tau = T/T_{Cb}$, $\tau_C = T_C/T_{Cb}$ and $R_{\mu\nu} = \sigma_\mu/\sigma_\nu$.

The second program to determine the Curie temperature is as follows. We substitute the initial values of σ_ν/σ_1 ($\nu = 2, 3, \dots, n$) and $\sigma_\nu/\sigma_\mu = (\sigma_\nu/\sigma_1)/(\sigma_\mu/\sigma_1)$ into equation (9). By iterative solution, we obtain the final values of σ_ν/σ_1 and the corresponding value of C_1 , which satisfy the condition (10). From equation (8) the Curie temperature is determined. τ_C , R_{21} and R_{31} versus D/J for 5-ML films are calculated from (8), (9) and (10); these curves are illustrated in figure 7 and are consistent with the results calculated from (6) and (7), shown in figure 5.

5. Discussion

The variation in τ_C with respect to D/J for various thicknesses of the n -ML film is illustrated schematically in figure 8; we see that the Curie temperature of a 1-ML film ($n = 1$) is lower than that of 2-ML or more films ($n \geq 2$) with an identical value of Dj . The results are easy to see from the dimensional effects. The exchange interaction of atoms for three-dimensional magnets is more than that for two-dimensional magnets owing to a decrease in the number of nearest-neighbour atoms. From figure 8, we find that the surface anisotropy may be divided into two cases according to the value of Dj : one is strong surface anisotropy (roughly $Dj > 0.5$) and the other is weak surface anisotropy (roughly $Dj < 0.5$). In the case of weak surface anisotropy, as the thickness of the n -ML films is decreased, the Curie temperature decreases and the Curie temperature of n -ML films is lower than that of the bulk magnet, which agrees with the experimental results reported in [1]. In the case of strong surface anisotropy, as the thickness of films of 2-ML or more is decreased, the Curie temperature rises and the Curie temperature of films of 2-ML or more is higher than that of the bulk magnet; by employing this theory we may interpret the experimental results reported in [2, 3]. Experiments [1] have shown that there are quite a few discrepancies in the values of Curie temperature measurement. We introduce the SAC to describe the surface situation of the film and suppose that the SAC is extremely sensitive to the lattice geometry, the type of substrate and coating layers, absorbates, lattice imperfections, and the stress and strain due to the growth conditions. Because of the strong dependence of Dj (or the SAC) on the actual film preparation, different experimental results have been obtained for the same system [1].

The temperature dependences of σ_ν for various D/J for 1-ML, 2-ML, 3-ML, 4-ML, 5-ML and 6-ML films are shown in figures 1, 2, 3, 4, 5 and 6, respectively. Generally, it has been proved that the order parameter changes with the position of the atomic planes, which are parallel to the surfaces of the n -ML films, but the Curie temperatures of atomic planes are all the same. In this paper we have discussed the special case when the SACS on both surfaces are equal ($D_1 = D_n$). It has been shown in figures 2–6 that the order parameters of inequivalent atomic planes are different. These figures show that for the case of weak surface anisotropy the order parameter of the surface atomic plane is less than those of the other atomic planes and for the case of strong surface anisotropy the order parameter of the surface atomic plane is more than those of the other atomic planes. For the case of very strong surface anisotropy (for example $Dj > 5$), the order parameter of the surface atomic plane are very different from those of the other atomic planes; their differences are roughly an order or several orders of magnitude just below the Curie temperature and this phenomenon becomes more marked as the thickness of the n -ML films or the value of D/J increases.

6. Conclusion

The variation in the Curie temperature with respect to the thickness and SAC of ultra-thin films has been demonstrated. The dependence of the Curie temperature on the thickness of ultra-thin films results from the surface anisotropy existing in these films. The Curie temperature of films of 2-ML or more is lower or higher than the bulk Curie temperature depending on whether the ratio D/J of the SAC to the exchange integral is less or more than Djc (for the SC lattice $Djc \simeq 0.5$, for the BCC lattice $Djc \simeq 2$ and for the FCC lattice $Djc \simeq 5$). A model calculation shows that it is impossible for an ordered surface phase to coexist with a disordered bulk phase and that it is also impossible for a disordered surface phase to coexist with an ordered bulk phase in homogeneous films or bulk magnet.

Acknowledgment

We are grateful for financial support from the National Natural Science Foundation of China under grant 1880767.

Appendix

For the case $n = 1$ (ν and $k = 1$)

$$\omega_{h1} = E_1$$

$$U_{11}^2 = 1.$$

For the case $n = 2$ (ν and $k = 1, 2$; $\sigma_2 = \sigma_1$),

$$\omega_{h1} = E_1 + J_1 \quad \omega_{h2} = E_1 - J_1$$

$$U_{11}^2 = U_{12}^2 = \frac{1}{2}.$$

For the case $n = 3$ (ν and $k = 1, 2, 3$; $\sigma_1 = \sigma_3$),

$$\omega_{h1} = E_1 \quad \omega_{h2} = \frac{1}{2}(A + B) \quad \omega_{h3} = \frac{1}{2}(A - B)$$

$$U_{11}^2 = \frac{1}{2} \quad U_{12}^2 = \frac{1}{4}[1 + (E_1 - E_2)/B] \quad U_{13}^2 = \frac{1}{4}[1 - (E_1 - E_2)/B]$$

$$U_{21}^2 = 0 \quad U_{22}^2 = \frac{1}{2}[1 - (E_1 - E_2)/B] \quad U_{23}^2 = \frac{1}{2}[1 + (E_1 - E_2)/B]$$

where

$$A = E_1 + E_2 \quad B = [(E_1 - E_2)^2 + 8J_1J_2]^{1/2}.$$

For the case $n = 4$ (ν and $k = 1, 2, 3, 4$; $\sigma_1 = \sigma_4$; $\sigma_2 = \sigma_3$),

$$\omega_{h1} = \frac{1}{2}(A + E) \quad \omega_{h2} = \frac{1}{2}(A - E) \quad \omega_{h3} = \frac{1}{2}(B + F) \quad \omega_{h4} = \frac{1}{2}(B - F)$$

$$U_{11}^2 = J_1J_2/E(E - D) \quad U_{12}^2 = J_1J_2/E(E + D) \quad U_{13}^2 = J_1J_2/F(F - C)$$

$$U_{14}^2 = J_1J_2/F(F + C) \quad U_{21}^2 = \frac{1}{4}(1 - D/E) \quad U_{22}^2 = \frac{1}{4}(1 + D/E)$$

$$U_{23}^2 = \frac{1}{4}(1 - C/F) \quad U_{24}^2 = \frac{1}{4}(1 + C/F)$$

where

$$A = E_1 + E_2 - J_2 \quad B = E_1 + E_2 + J_2 \quad C = E_1 - E_2 - J_2$$

$$D = E_1 - E_2 + J_2 \quad E = [(E_1 - E_2 + J_2)^2 + 4J_1J_2]^{1/2}$$

$$F = [(E_1 - E_2 - J_2)^2 + 4J_1J_2]^{1/2}.$$

For the case $n = 5$ (ν and $k = 1, 2, 3, 4, 5$; $\sigma_1 = \sigma_5$; $\sigma_2 = \sigma_4$) from equation (4) factorization, we obtain the characteristic equations

$$(\omega_{hk} - E_1)(\omega_{hk} - E_2) - J_1J_2 = 0 \quad (\text{A1})$$

$$(\omega_{hk} - E_1)(\omega_{hk} - E_2)(\omega_{hk} - E_3) - J_1J_2(\omega_{hk} - E_3) - 2J_2J_3(\omega_{hk} - E_1) = 0. \quad (\text{A2})$$

After determining ω_{h1} and ω_{h2} from equation (A1) and ω_{h3} , ω_{h4} and ω_{h5} from equation (A2), we can simplify $U_{\nu k}^2$ as follows:

$$\omega_{h1} = (E_1 + E_2 + Q)/2 \quad \omega_{h2} = (E_1 + E_2 - Q)/2 \quad \omega_{h3} = R \cos(\theta/3) - a$$

$$\omega_{h4} = R \cos[(\theta + 2\pi)/3] - a \quad \omega_{h5} = R \cos[(\theta - 2\pi)/3] - a$$

where

$$Q = [(E_1 - E_2)^2 + 4J_1J_2]^{1/2} \quad a = -(E_1 + E_2 + E_3)/3$$

$$b = E_1(E_2 + E_3) + E_2E_3 - J_2(J_1 + 2J_3) \quad c = E_3(J_1J_2 - E_1E_2) + 2E_1J_2J_3$$

$$U = -c/2 - a(a^2 - b/2) \quad P = b/3 - a^2 \quad V = (|U^2 + P^3|)^{1/2}$$

$$(\text{because } U^2 + P^3 < 0) \quad \theta = \tan^{-1}(V/U) \quad R = 2(U^2 + V^2)^{1/6}$$

$$U_{11}^2 = (\omega_{h1} - E_2)/2(\omega_{h1} - \omega_{h2}) \quad U_{12}^2 = (\omega_{h2} - E_2)/2(\omega_{h2} - \omega_{h1})$$

$$U_{13}^2 = [(\omega_{h3} - E_2)(\omega_{h3} - E_3) - 2J_2J_3]/2(\omega_{h3} - \omega_{h4})(\omega_{h3} - \omega_{h5})$$

$$U_{14}^2 = [(\omega_{h4} - E_2)(\omega_{h4} - E_3) - 2J_2J_3]/2(\omega_{h4} - \omega_{h3})(\omega_{h4} - \omega_{h5})$$

$$U_{15}^2 = [(\omega_{h5} - E_2)(\omega_{h5} - E_3) - 2J_2J_3]/2(\omega_{h5} - \omega_{h3})(\omega_{h5} - \omega_{h4})$$

$$U_{21}^2 = (\omega_{h1} - E_1)/2(\omega_{h1} - \omega_{h2}) \quad U_{22}^2 = (\omega_{h2} - E_1)/2(\omega_{h2} - \omega_{h1})$$

$$U_{23}^2 = (\omega_{h3} - E_1)(\omega_{h3} - E_3)/2(\omega_{h3} - \omega_{h4})(\omega_{h3} - \omega_{h5})$$

$$U_{24}^2 = (\omega_{h4} - E_1)(\omega_{h4} - E_3)/2(\omega_{h4} - \omega_{h3})(\omega_{h4} - \omega_{h5})$$

$$U_{25}^2 = (\omega_{h5} - E_1)(\omega_{h5} - E_3)/2(\omega_{h5} - \omega_{h3})(\omega_{h5} - \omega_{h4}) \quad U_{31}^2 = 0 \quad U_{32}^2 = 0$$

$$U_{33}^2 = [(\omega_{h3} - E_1)(\omega_{h3} - E_2) - J_1J_2]/(\omega_{h3} - \omega_{h4})(\omega_{h3} - \omega_{h5})$$

$$U_{34}^2 = [(\omega_{h4} - E_1)(\omega_{h4} - E_2) - J_1J_2]/(\omega_{h4} - \omega_{h3})(\omega_{h4} - \omega_{h5})$$

$$U_{35}^2 = [(\omega_{h5} - E_1)(\omega_{h5} - E_2) - J_1J_2]/(\omega_{h5} - \omega_{h3})(\omega_{h5} - \omega_{h4}).$$

For the case $n = 6$ (ν and $k = 1, 2, 3, 4, 5, 6$; $\sigma_1 = \sigma_6$; $\sigma_2 = \sigma_5$; $\sigma_3 = \sigma_4$), from equation (4) factorization, we obtain the characteristic equations

$$(\omega_{hk} - E_1)(\omega_{hk} - E_2)(\omega_{hk} - E_3) - J_1 J_2 (\omega_{hk} - E_3) - J_2 J_3 (\omega_{hk} - E_1) + J_3 [(\omega_{hk} - E_1)(\omega_{hk} - E_2) - J_1 J_2] = 0 \tag{A3}$$

$$(\omega_{hk} - E_1)(\omega_{hk} - E_2)(\omega_{hk} - E_3) - J_1 J_2 (\omega_{hk} - E_3) - J_2 J_3 (\omega_{hk} - E_1) - J_3 [(\omega_{hk} - E_1)(\omega_{hk} - E_2) - J_1 J_2] = 0. \tag{A4}$$

After determining ω_{h1} , ω_{h2} and ω_{h3} from equation (A3) and ω_{h4} , ω_{h5} and ω_{h6} from equation (A4), we can simplify $U_{\nu k}^2$ as follows:

$$\begin{aligned} \omega_{h1} &= R_1 \cos(\theta_1/3) - a_1 & \omega_{h2} &= R_1 \cos[(\theta_1 + 2\pi)/3] - a_1 \\ \omega_{h3} &= R_1 \cos[(\theta_1 - 2\pi)/3] - a_1 & \omega_{h4} &= R_2 \cos(\theta_2/3) - a_2 \\ \omega_{h5} &= R_2 \cos[(\theta_2 + 2\pi)/3] - a_2 & \omega_{h6} &= R_2 \cos[(\theta_2 - 2\pi)/3] - a_2 \end{aligned}$$

where

$$a_1 = -(E_1 + E_2 + E_3 - J_3)/3$$

$$b_1 = E_1(E_2 + E_3) + E_2 E_3 - J_2(J_1 + J_3) - J_3(E_1 + E_2)$$

$$c_1 = E_3(J_1 J_2 - E_1 E_2) + E_1 J_2 J_3 + (E_1 E_2 - J_1 J_2) J_3$$

$$U_1 = -c_1/2 - a_1(a_1^2 - b_1/2)$$

$$P_1 = b_1/3 - a_1^2 \quad V_1 = (|U_1^2 + P_1^3|)^{1/2} \quad (\text{because } U_1^2 + P_1^3 < 0)$$

$$\theta_1 = \tan^{-1}(V_1/U_1) \quad R_1 = 2(U_1^2 + V_1^2)^{1/6}$$

$$a_2 = -(E_1 + E_2 + E_3 + J_3)/3$$

$$b_2 = E_1(E_2 + E_3) + E_2 E_3 - J_2(J_1 + J_3) + J_3(E_1 + E_2)$$

$$c_2 = E_3(J_1 J_2 - E_1 E_2) + E_1 J_2 J_3 - (E_1 E_2 - J_1 J_2) J_3$$

$$U_2 = -c_2/2 - a_2(a_2^2 - b_2/2)$$

$$P_2 = b_2/3 - a_2^2 \quad V_2 = (|U_2^2 + P_2^3|)^{1/2} \quad (\text{because } U_2^2 + P_2^3 < 0)$$

$$\theta_2 = \tan^{-1}(V_2/U_2) \quad R_2 = 2(U_2^2 + V_2^2)^{1/6}$$

$$U_{11}^2 = [(\omega_{h1} - E_2)(\omega_{h1} - E_3 + J_3) - J_2 J_3]/2(\omega_{h1} - \omega_{h2})(\omega_{h1} - \omega_{h3})$$

$$U_{12}^2 = [(\omega_{h2} - E_2)(\omega_{h2} - E_3 + J_3) - J_2 J_3]/2(\omega_{h2} - \omega_{h3})(\omega_{h2} - \omega_{h1})$$

$$U_{13}^2 = [(\omega_{h3} - E_2)(\omega_{h3} - E_3 + J_3) - J_2 J_3]/2(\omega_{h3} - \omega_{h1})(\omega_{h3} - \omega_{h2})$$

$$U_{14}^2 = [(\omega_{h4} - E_2)(\omega_{h4} - E_3 - J_3) - J_2 J_3]/2(\omega_{h4} - \omega_{h5})(\omega_{h4} - \omega_{h6})$$

$$U_{15}^2 = [(\omega_{h5} - E_2)(\omega_{h5} - E_3 - J_3) - J_2 J_3]/2(\omega_{h5} - \omega_{h6})(\omega_{h5} - \omega_{h4})$$

$$\begin{aligned}
U_{16}^2 &= [(\omega_{h6} - E_2)(\omega_{h6} - E_3 - J_3) - J_2 J_3]/2(\omega_{h6} - \omega_{h4})(\omega_{h6} - \omega_{h5}) \\
U_{21}^2 &= (\omega_{h1} - E_1)(\omega_{h1} - E_3 + J_3)/2(\omega_{h1} - \omega_{h2})(\omega_{h1} - \omega_{h3}) \\
U_{22}^2 &= (\omega_{h2} - E_1)(\omega_{h2} - E_3 + J_3)/2(\omega_{h2} - \omega_{h3})(\omega_{h2} - \omega_{h1}) \\
U_{23}^2 &= (\omega_{h3} - E_1)(\omega_{h3} - E_3 + J_3)/2(\omega_{h3} - \omega_{h1})(\omega_{h3} - \omega_{h2}) \\
U_{24}^2 &= (\omega_{h4} - E_1)(\omega_{h4} - E_3 - J_3)/2(\omega_{h4} - \omega_{h5})(\omega_{h4} - \omega_{h6}) \\
U_{25}^2 &= (\omega_{h5} - E_1)(\omega_{h5} - E_3 - J_3)/2(\omega_{h5} - \omega_{h6})(\omega_{h5} - \omega_{h4}) \\
U_{26}^2 &= (\omega_{h6} - E_1)(\omega_{h6} - E_3 - J_3)/2(\omega_{h6} - \omega_{h4})(\omega_{h6} - \omega_{h5}) \\
U_{31}^2 &= [(\omega_{h1} - E_1)(\omega_{h1} - E_2) - J_1 J_2]/2(\omega_{h1} - \omega_{h2})(\omega_{h1} - \omega_{h3}) \\
U_{32}^2 &= [(\omega_{h2} - E_1)(\omega_{h2} - E_2) - J_1 J_2]/2(\omega_{h2} - \omega_{h3})(\omega_{h2} - \omega_{h1}) \\
U_{33}^2 &= [(\omega_{h3} - E_1)(\omega_{h3} - E_2) - J_1 J_2]/2(\omega_{h3} - \omega_{h1})(\omega_{h3} - \omega_{h2}) \\
U_{34}^2 &= [(\omega_{h4} - E_1)(\omega_{h4} - E_2) - J_1 J_2]/2(\omega_{h4} - \omega_{h5})(\omega_{h4} - \omega_{h6}) \\
U_{35}^2 &= [(\omega_{h5} - E_1)(\omega_{h5} - E_2) - J_1 J_2]/2(\omega_{h5} - \omega_{h6})(\omega_{h5} - \omega_{h4}) \\
U_{36}^2 &= [(\omega_{h6} - E_1)(\omega_{h6} - E_2) - J_1 J_2]/2(\omega_{h6} - \omega_{h4})(\omega_{h6} - \omega_{h5}).
\end{aligned}$$

References

- [1] Gradmann U 1974 *Appl. Phys.* **3** 161
 Koon N C, Jonker B T, Volkening F A, Krebs J J and Prinz G A 1987 *Phys. Rev. Lett.* **59** 2463
 Stampanoni M, Vatterlaus A, Aeschlimann M and Meier F 1987 *Phys. Rev. Lett.* **59** 2483
 Liu C, Mong E R and Bader S D 1988 *Phys. Rev. Lett.* **60** 2422
 Durr W, Taborelli M, Paul O, Germar R, Gudat W, Pescia D and Landolt M 1989 *Phys. Rev. Lett.* **62** 206
 Liu C and Bader S D 1990 *J. Appl. Phys.* **67** 5758
- [2] Weller D, Alvarado S F, Gudat W, Schroder K and Campagna M 1985 *Phys. Rev. Lett.* **54** 1555
- [3] Rau C and Robert M 1987 *Phys. Rev. Lett.* **58** 2714
 Rau C, Jin C and Robert M 1988 *J. Appl. Phys.* **63** 3667
- [4] Mills D L 1971 *Phys. Rev. B* **3** 3887
- [5] Shi L P 1987 *J. Appl. Phys.* **61** 4267
- [6] Shi L P, Ji Z R and Hu L 1991 *J. Appl. Phys.* **69** 5313
- [7] Gay J G and Richter R 1987 *J. Appl. Phys.* **61** 3362; 1986 *Phys. Rev. Lett.* **56** 2728
- [8] Potapkov N A 1962 *Dokl. Akad. Nauk. SSSR* **144** 297
- [9] Tyablikov S V 1967 *Methods in the Quantum Theory of Magnetism*. (New York: Plenum) section 35
 Praveczi E 1963 *Phys. Lett.* **6** 147
 Callen H B 1963 *Phys. Rev.* **130** 890