Available online at www.sciencedirect.com
SCIENCE DIRECT®
Surface Science 552 (2004) 1–7
SURFACE SCIENCE
www.elsevier.com/locate/susc

Monte Carlo study of order–disorder layering transitions in the Blume–Capel model

L. Bahmad, A. Benyoussef, H. Ez-Zahraouy *

Laboratoire de Magnétisme et de la Physique des Hautes Energies, Université Mohammed V, Avenue Ibn Batouta, B.P. 1014, Rabat 10000, Morocco

Received 29 October 2003; accepted for publication 26 January 2004

Abstract

The order–disorder layering transitions, of the spin-1 Blume–Capel model, are studied using the Monte Carlo (MC) simulations, in the presence of a variable crystal field. The system exhibits tri-critical behavior and reentrant phenomena. The reentrant phenomena is found for the first k_0 layers from the surface, where k_0 is exactly the number of layering transitions at the ground state. This is qualitatively in agreement with mean field results. Layer magnetizations, quadrupolar and magnetic susceptibilities are calculated. The critical exponents of magnetic susceptibility of each layer are computed for $N = 10$ and different sizes of the system. It is found that the critical exponent of a 2D layer in a film is different than the one of a free 2D layer. Indeed the critical exponent of a layer depends strongly on its position from the surface.
© 2004 Elsevier B.V. All rights reserved.

Keywords: Ising models; Monte Carlo simulations; Magnetic phenomena (cyclotron resonance, phase transitions, etc.); Magnetic films

1. Introduction

Since the theory of surface critical phenomena started developing, much attention has been devoted to the study of the Blume–Capel (BC) model over semi-infinite lattices, with modified surface couplings. Benyoussef et al. [1] have determined the phase diagram in the mean field approximation, reporting four possible topologies at fixed bulk/surface coupling ratios. An analogous analysis have also been done using a real space renormalization group transformation [2]. Some other works referring to particular regions of the phase space are, for example, those using: the mean field approximation [3], the effective field approximation [4] and the low temperature expansion [5]. The discussion presented in Ref. [6] shows that the strong interest in these models arises partly from the unusually rich phase transition behavior they display as their interaction parameters are varied, and partly from their many possible applications. The bilinear interaction considered in most of these cases is ferromagnetic. The spin-1 Ising systems are used to describe both the order–disorder transition and the crystallization of the binary allow, and it was solved for the anti-ferromagnetic case in the mean field approach [7]. The decomposition of a line of tri-critical points into a line of

* Corresponding author.
E-mail address: ezahamid@fsr.ac.ma (H. Ez-Zahraouy).

0039-6028/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.susc.2004.01.044

critical end points and one of double critical points is one of the most interesting features of the mean field phase diagram for the anti-ferromagnetic spin-1 Blume–Capel model in an external magnetic field [8]. The transfer-matrix and Monte Carlo finite-size-scaling methods [9], are also applied to study this model but such decomposition does not occur in the two dimensional case. The finite cluster approximation has been applied by Benyoussef et al. [10] in order to study the spin-1 Ising model with a random crystal field. On the other hand, the transverse field or crystal field effects of spin-1 Ising model has been studied by several authors [11–14].

The layer-by-layer transitions, in a film, are a sequence of layer ordering transitions from surface to deeper layers. The experimental measurements of layer-by-layer ordering phenomena have been established on free-standing liquid crystals films such as *nmOBC* (*n*-alkyl-4′-*n*-alkyloxybiphenyl-4-carboxylate) [15,16] and 54*COOBC* (*n*-pentyl-4′-*n*-pentanoyloxy-biphenyl-4-carboxylate) [17] for several molecular layers. More recently, Lin et al. [18] have used the three-level Potts model to show the existence of layer-by-layer ordering of ultra thin liquid crystal films of free-standing 54*COOBC* films, by adjusting the interlayer and intra layer couplings between nearest- neighboring molecules.

Depending on the value of the crystal field at the surface some systems undergo a phase transition from a completely disordered state, at high temperatures, to a state with the first k layers disordered and the rest of the layers ordered at low very temperatures. One can then observe a particuliar behaviour when the transition to the order state of the k^{th} layer occurs not only with the decrease but also with the increase of the temperature. Such phase transitions are called the reentrant phenomena.

The reentrant first-order layering transitions have been observed experimentally in multi-layer argon films on graphite by Youn and Hess [19]. In our previous work [20], interesting results were obtained within MFA such as order–disorder layering transitions and reentrant phenomena. But it is well known that the MFA approximation does not take into account the thermal fluctuations and neglects correlations between spins. This can lead to incorrect results. Our purpose, in one hand, is to improve the results obtained in [20] and to compute the critical exponents corresponding to the magnetic susceptibilities of each layer. It is found that the multi-critical points and reentrant phenomena still exist but within different critical behaviour. Indeed, we have shown that the critical exponent of a free 2D layer is different than the one of a single layer in a film. However, the critical exponent of each layer depends strongly on its position counted from the surface.

The paper is organized as follows. Section 2 describes the model and method. In Section 3 we present results and discussions.

2. Model and method

The system we are studying here is formed with N coupled ferromagnetic square layers in the presence of a crystal field. The Hamiltonian governing this system is given by

$$\mathscr{H} = -\sum_{\langle i,j\rangle} J_{ij} S_i S_j + \sum_i \Delta_i (S_i)^2, \tag{1}$$

where $S_l (l = i, j) = -1, 0, +1$ are the spin variables. The interactions between different spins are assumed to be constant so that $J_{ij} = J$. All sites i of a given layer k, $k = 1, k = 2, \ldots, k = N$ are subject to a crystal field amplitude $\Delta_i = \Delta_k$ so that $\Delta_1 > \Delta_2 > \cdots > \Delta_N$.

The model we are studying, in this paper, corresponds to a crystal field distributed according to the law

$$\Delta_k = \Delta_s / k^{\alpha}, \tag{2}$$

where $\Delta_s = \Delta_1$ is the crystal field acting on the surface (first layer $k = 1$) and α a positive constant. For a system with $\alpha = 0$, a constant crystal field is applied on each layer of the film. Whereas, for $\alpha \to \infty$ the crystal field is applied only on the first layer $k = 1$. In all the following we will give numerical results for $\alpha = 1.0$.

Experimental studies of a sequence of reentrant layering transitions of argon and krypton adsorbed on graphite substrate are given in Ref. [21]. This reference shows also the algebraically decaying of the applied field according to a law of type $(k^{-\alpha})$.

The quantities computed in the present work, using the Monte Carlo simulations for each layer k containing N_x spins in x-direction and N_y spins in the y-direction, are

- The layer average magnetizations $m_k = \langle M_k \rangle$, where the site average magnetization M_k of the layer k is given by

$$M_k = \left(\sum_{i \epsilon k} S_i \right) \Big/ (N_x N_y). \tag{3}$$

- The layer magnetic susceptibilities defined as

$$\chi_{m,k} = \beta N_x N_y \langle (M_k - m_k)^2 \rangle. \tag{4}$$

- The layer quadrupolar magnetic susceptibilities expressed as

$$\chi_{q,k} = \beta N_x N_y \langle (q_k - \langle q_k \rangle)^2 \rangle, \tag{5}$$

where

$$q_k = \left(\sum_{i \epsilon k} S_i^2 \right) \Big/ (N_x N_y). \tag{6}$$

- The layer critical exponents $\gamma_{m,k}$ related to the corresponding layer magnetic susceptibilities $\chi_{m,k}$, for a fixed layer k, giving by

$$\gamma_{m,k} = \frac{\partial \operatorname{Log}(\chi_{m,k})^{-1}}{\partial \operatorname{Log}|T_{c,k}/J - T/J|}, \tag{7}$$

where T/J stands for the reduced absolute temperature and $T_{c,k}/J$ is the reduced critical temperature of the layer k.

In the above equations $\beta = 1/(k_B T)$ with k_B being the Boltzmann constant.

The notation $D^k O^{N-k}$ will be used to denote that the first k layers from the surface are disordered while the remaining $N-k$ layers are ordered. In particular, O^N corresponds to an ordered film whereas D^N denotes a totally disordered film. The surface crystal field Δ_k, applied on each layer k is distributed according to the law given by Eq. (2).

3. Results and discussion

Monte Carlo simulations have been made on a system with N layers and $N_x = N_y$ spins for the x and y-directions of each layer. Runs of $500\,000/N$ Monte Carlo steps MCS were performed with the discard of the first $50\,000/N$ Monte Carlo steps.

We have performed a preliminary study and found that by changing the system sizes from $N = 10$ to $N = 15, 20$ layers and $N_x = N_y = 64$ to $N_x = N_y = 128$, the relevant measured quantities did not change appreciably. For this reason we give in all the following, numerical results for a film formed with $N = 10$ layers and $N_x = N_y = 64$ spins for the directions x and y.

We have established a detailed study of the ground state phase diagram of this model in Ref. [20]. In particular, we showed the existence of a critical order layer k_0 corresponding to the transition $D^{k_0} O^{N-k_0} \leftrightarrow D^N$, at a reduced surface crystal field given by

$$\Delta_s/J = (3(N-k_0)-1) \Big/ \left(\sum_{k=k_0+1}^{N} (1/k^{\alpha}) \right). \tag{8}$$

k_0 is exactly the number of layering transitions existing at $T/J = 0$. It is found that k_0 depends both on the parameter α and the film thickness N.

In order to outline the existence of the order–disorder layering transitions we plot in Fig. 1 the temperature-crystal field phase diagram for $\alpha = 1.0$. For nonzero but very low temperatures, the surface crystal field values found in this phase diagram are exactly those predicted by the ground state study established in Ref. [20], namely: $\Delta_s/J = 3(k)^{\alpha}$ for the first k_0 layers. Indeed, the first layer transition is found at $\Delta_s/J = 3.0$; the second transition at $\Delta_s/J = 6.0$; the third transition at $\Delta_s/J = 9.0$ and so on. Monte Carlo simulations show that the last layers $k = 8, 9$ and 10 transit simultaneously at the numerical value of the surface crystal field: $\Delta_s/J \approx 23.84$. This is in good agreement with the predicted value ($\approx$23.80) from the above Eq. (8) established on the basis of an analytical study.

The reentrant phenomena, shown in Fig. 1 for the first k_0 layers, is caused by the competition between thermal fluctuations, the applied crystal field and an effective magnetic field created by the deeper layers. Indeed, when these thermal fluctuations become sufficiently important, the magnetization of some spins, of deeper layers, becomes

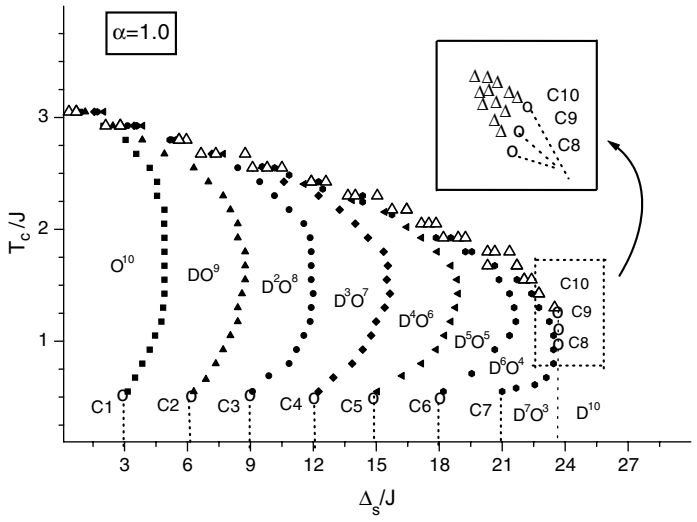


Fig. 1. The critical temperature behavior as a function of the surface crystal field Δ_s/J for $\alpha = 1.0$ and a film thickness $N = 10$ layers. For each layer k ($k = 1, 2, \ldots, N$), the first-order transition line (vertical dashed line) is connected to the second-order transition line (up-triangular points) by a tri-critical point C_k (open circle). The notations $D^k O^{N-k}$ are defined in the body text. In the inset, the tri-critical points C_8, C_9, and C_{10} are found at higher and different critical temperatures.

nonzero (+1 or −1). This leads to the appearance of an effective magnetic field. This magnetic field is responsible of the ordered phase seen for the layer k. This argument can also explain the absence of the reentrant phenomena for the last $N - k_0$ layers, once the value of the crystal field is not sufficient to overcome the effective magnetic field created by the remaining deeper layers.

It is worth to note that the reentrant phenomena is always present for the layers k, ($k \leqslant k_0$), and the corresponding tri-critical points C_i are located at a constant temperature.

In absence of the surface crystal field, $\Delta_s/J \to 0$, each layer of the film disorders at a fixed temperature $T_c/J = 3.15$. In order to compare this critical temperature value obtained for $N = 10$ layers, with the three-dimensional spin-1 critical temperature, we performed Monte Carlo simulations on films with a fixed surface $(N_x, N_y) = (128, 128)$ and increasing thicknesses: $N = 16; 32; 64$ and 128. It is found that the corresponding critical temperature increases very slowly with the number of layers once $N \geqslant 32$. For $N = 128$, the critical temperature we computed did not exceed the value $T_c/J = 3.19(5)$. This value corresponding to $N = 10$ layers, is smaller than that one computed for an infinite three-dimensional cubic lattice spin-1 Ising model, by the mean field approach, which is exactly $T_c/J = 4.0$.

On the other hand, as it is shown in Fig. 1, the first k_0 tri-critical temperature values, established by MC simulations are approximately: $T_{ci}/J \approx 0.50$. These temperature values are smaller than those established in Ref. [20] when using the mean field method: $T_{ci}/J \approx 0.78$. A detailed study is done for a temperature lower than T_{ci}/J, as it is shown in Fig. 2 for $T/J = 0.2$. On the other hand the layer quadrupolar magnetic susceptibilities $\chi_{q,k}$ defined by Eq. (5), for each layer k, present a strong peak at the surface crystal field values corresponding to each layer transition; as it is illustrated by Fig. 2b. The satellite of the peaks, present in this figure, means that the transitions are of first-order type for a very low temperature: $T/J = 0.2$. For a higher temperature, see Fig. 3 corresponding to $T/J = 1.0$, the layering transitions are of second-order type. Indeed, this is shown in Fig. 3a for the individual layering temperature behaviors, as a function of the surface crystal field. The corresponding magnetic susceptibilities $\chi_{m,k}$, defined by Eq. (4) for each layer k, present a strong peak at the corresponding surface

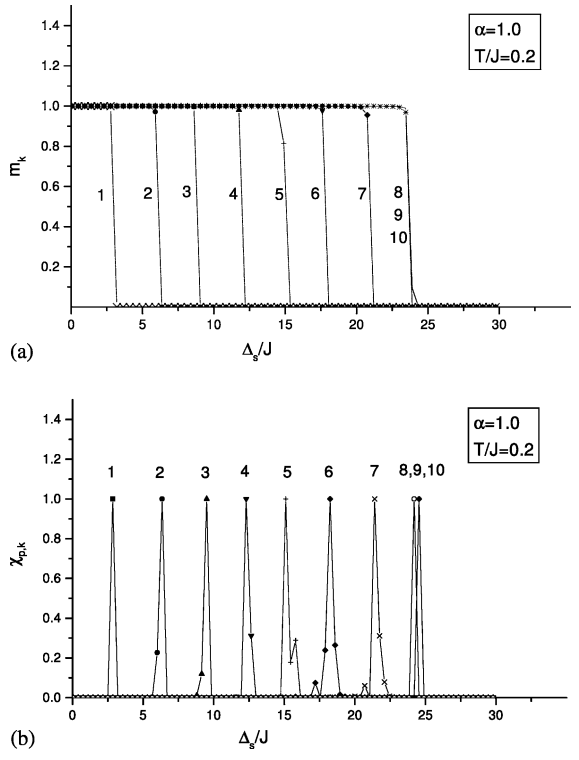


Fig. 2. The behavior of the reduced layer magnetizations m_k (a), and the layer quadrupolar magnetic susceptibilities $\chi_{q,k}$ (b), showing a first-order transition, as a function of the surface crystal field Δ_s/J, for a very low temperature: $T/J = 0.2$. The number accompanying each curve denotes the layer order from the surface to deeper layers.

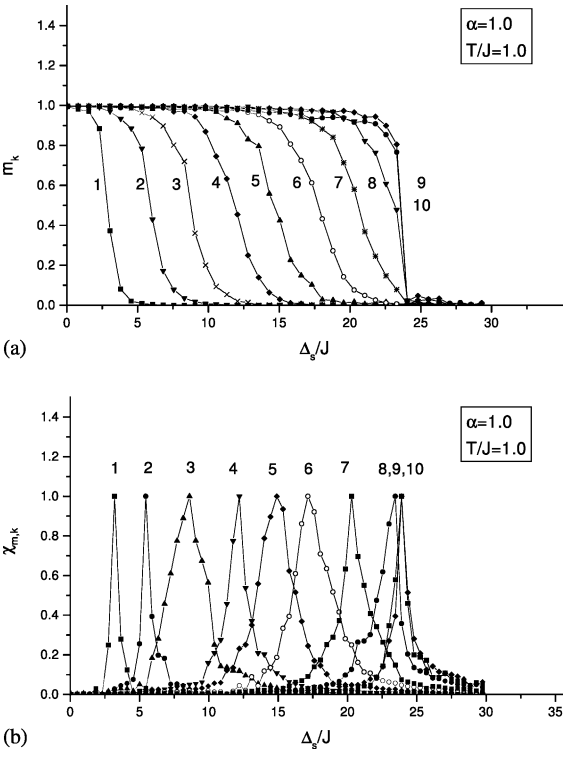


Fig. 3. For a higher temperature $T/J = 1.0$, the dependency of the layer magnetizations m_k (a), and the reduced layer magnetic susceptibilities $\chi_{m,k}$ (b), undergoes a second-order transition, as a function of the surface crystal field Δ_s/J.

crystal field. It is worth to note that Figs. 2b and 3b are plotted for reduced numerical values of the layer magnetic and the layer quadrupolar magnetic susceptibilities: $\chi_{q,k}$ and $\chi_{m,k}$, respectively.

To show the increasing temperature effect, for a fixed surface crystal field, on the order–disorder transitions; we illustrate in Fig. 4 the corresponding layering transitions for $\Delta_s/J = 2.0$ for each layer. It is found that these transitions are second-order type and are located at $T/J \approx 2.95$, see Fig. 4a for the layer magnetization behavior and Fig. 4b for the layer magnetic susceptibilities $\chi_{m,k}$. On the other hand the reentrant phenomenon, seen for the first k_0 layers is well illustrated in Fig. 5 for the layer $k = 7$ at a fixed surface crystal field value $\Delta/J = 22.0$. Indeed, the two transitions: from disorder to order and from order to disorder of the layer $k = 7$ are well seen in Fig. 5 for the layer magnetization m_7. The corresponding layer magnetic susceptibility $\chi_{m,7}$ exhibits two strong peaks at these transitions. In order to complete this study, we have investigated the layer critical exponents $\gamma_{m,k}$ for $N = 10$ layers and several system sizes $N_x \times N_y = 16 \times 16$, $N_x \times N_y = 32 \times 32$, $N_x \times N_y = 64 \times 64$, and $N_x \times N_y = 128 \times 128$; see Fig. 6. It is found that $\gamma_{m,k}$ decreases, for a fixed order k, with the system size $N_x \times N_y$ and stabilizes at certain value; and due to the free boundary conditions the critical exponents of the layers $k = 1$ and $k = N$ are found to be greater than those of the internal layers $2 \leqslant k \leqslant N - 1$.

4. Conclusion

The order–disorder layering transitions of the Blume–Capel Ising model have been studied under

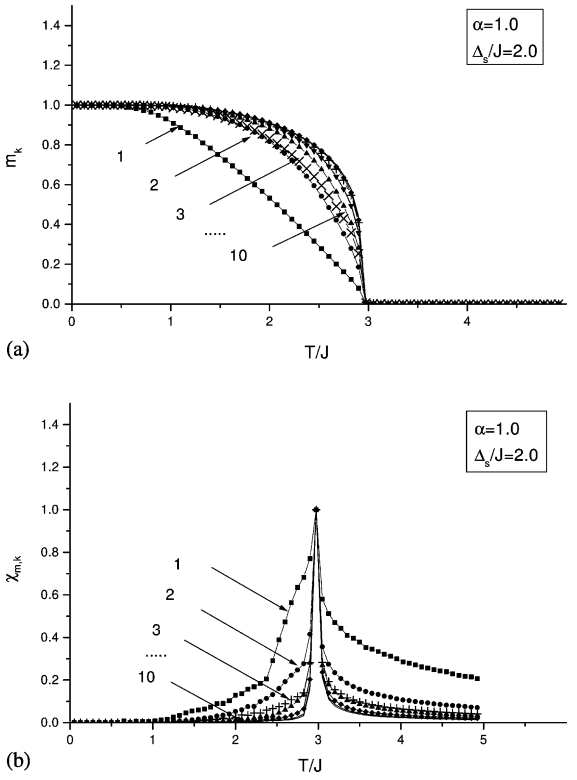


Fig. 4. For a fixed surface crystal field value $\Delta_s/J = 2.0$, the layer magnetizations m_k (a) and the reduced layer magnetic susceptibilities $\chi_{m,k}$ (b) show a second-order transition as a function of increasing temperature. For each layer k ($k = 1, 2, \ldots, N$), the transition is located at $T/J = 2.95$.

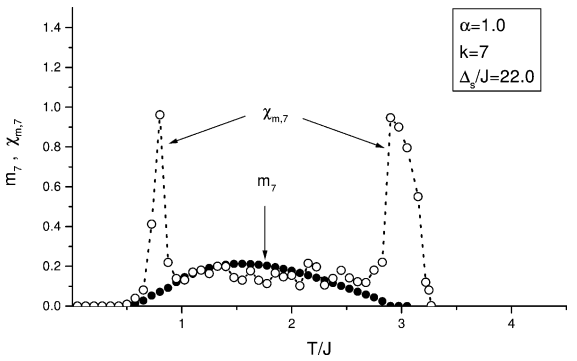


Fig. 5. Thermal behavior of the layer magnetization m_7 (line with solid circles) and the reduced layer magnetic susceptibility $\chi_{m,7}$ (line with open circles) for a deeper layer $k = 7$. The reentrant phenomena as well as the second-order transition are both outlined by the layer magnetization transition from D^7O^3 to D^6O^4 and from D^6O^4 to D^{10} corresponding to two peaks of the reduced layer magnetic susceptibility $\chi_{m,7}$ for $\Delta_s/J = 22.0$.

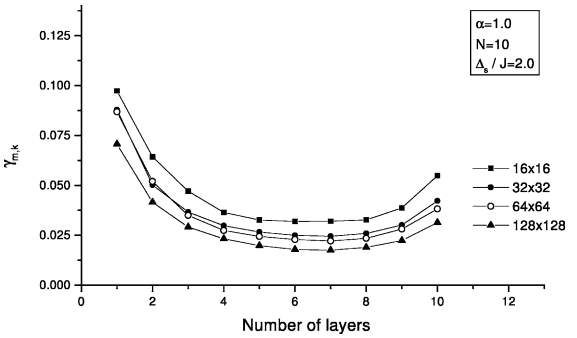


Fig. 6. Layer critical exponents $\gamma_{m,k}$ for $\Delta_s/J = 2.0$ as a function of the position k for a film thickness $N = 10$ layers, and several sizes: $N_x \times N_y = 16 \times 16$ (■); $N_x \times N_y = 32 \times 32$ (●); $N_x \times N_y = 64 \times 64$ (○); and $N_x \times N_y = 128 \times 128$ (▲).

the effect of a variable crystal field according to the law Eq. (2) and using Monte Carlo simulations. The reentrant phenomena, caused by a competition of thermal fluctuations and an inductor magnetic field created by the deeper layers, is found for the first k_0 layers counted from the surface; where k_0 is exactly the number of layering transitions allowed by the ground state study. We established the temperature-crystal field phase diagrams and found that the last $N - k_0$ layer tri-critical points are located at higher temperature values for a fixed exponent α and film thickness N. Although the model and the established equations are valid for any values of the parameters α and N, a numerical study is illustrated for $\alpha = 1.0$ and a film with $N = 10$ layers. For very low temperatures, the results are in good agreement with those predicted by the ground state study. The first-order and the second-order transition lines are connected by a tri-critical point, for each layer. On the other hand, the layer magnetizations m_k, the magnetic susceptibilities $\chi_{m,k}$, the quadrupolar magnetic susceptibilities $\chi_{q,k}$, and the critical exponents $\gamma_{m,k}$ for each layer k, have been investigated.

References

[1] A. Benyoussef, N. Boccara, M. Saber, J. Phys. C 19 (1986) 1983.
[2] A. Benyoussef, N. Boccara, M. Bouziani, Phys. Rev. B 34 (1986) 7775.
[3] X.P. Jiang, M.R. Giri, J. Phys. C 21 (1988) 995.

[4] I. Tamura, J. Phys. Soc. Jpn. 51 (1982) 3607.
[5] C. Buzano, A. Pelizzola, Physica A 195 (1993) 197.
[6] J.B. Collins, P.A. Rikvold, E.T. Gawlinski, Phys. Rev. B 38 (1988) 6741.
[7] Y. Saito, J. Chem. Phys. 74 (1981) 713.
[8] Y.L. Wang, K. Rauchwarger, Phys. Lett. A 59 (1976) 73.
[9] J.D. Kimel, P.A. Rikvold, Y.L. Wang, Phys. Rev. B 45 (1992) 7237.
[10] A. Benyoussef, H. Ez-Zahraouy, J. Phys.: Condens. Matter 6 (1994) 3411.
[11] R.B. Stinchcombe, J. Phys. C 6 (1973) 2459.
[12] J.L. Zhong, J. Liangli, C.Z. Yang, Phys. Status Solidi B 160 (1990) 329.
[13] U.V. Ulyanoc, O.B. Zalavskii, Phys. Rep. 216 (1992) 179.
[14] A. Benyoussef, H. Ez-Zahraouy, M. Saber, Physica A 198 (1993) 593.
[15] R. Geer, T. Stoebe, C.C. Huang, R. Pindak, J.W. Goodby, M. Cheng, J.T. Ho, S.W. Hui, Nature 355 (1992) 152.
[16] T. Stoebe, R. Geer, C.C. Huang, J.W. Goodby, Phys. Rev. Lett. 69 (1992) 2090.
[17] A.J. Jin, M. Veum, T. Stoebe, C.F. Chou, J.T. Ho, S.W. Hui, V. Surendranath, C.C. Huang, Phys. Rev. Lett. 74 (1995) 4863;
Phys. Rev. E 53 (1996) 3639.
[18] D.L. Lin, J.T. Ou, L.-P. Shi, X.R. Wang, A.J. Jin, Europhys. Lett. 50 (2000) 615.
[19] H.S. Youn, G.B. Hess, Phys. Rev. Lett. 64 (1990) 918.
[20] L. Bahmad, A. Benyoussef, H. Ez-Zahraouy, J. Magn. Magn. Mat. 251 (2002) 115.
[21] P.B. Weichmann, P. Day, D. Goodstein, Phys. Rev. Lett. 74 (1995) 918.