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Phase diagrams of a diluted Ising ferromagnetic film with spin 1 in a transverse field

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Abstract. The phase transition properties of a diluted Ising ferromagnetic film with $s = 1$ in a transverse field are investigated and calculated within the framework of the effective-field theory. We discuss mainly the phase diagrams as well as the dependence of the critical concentration of magnetic atoms on the surface coupling constant and thickness of film. Some interesting properties are found.

1. Introduction

From both the experimental and the theoretical points of view the Ising magnetic thin film is very important. It can be taken as a model used to investigate the magnetic size effects and can be regarded as a quasi-two-dimensional system when it is very thin. The magnetic properties of the film will approach those of the corresponding semi-infinite system when it is very thick. The semi-infinite system, as we know, is an example by which one can investigate magnetic surface properties, and some especially interesting results were obtained by using different methods [1–4]. Magnetic ordering, for instance, can first appear on the surface as long as the coupling constants between magnetic atoms on the surface are strong enough. For a very thin Ising film, as has been discussed [5, 6], its properties are obviously different from those of the corresponding bulk and semi-infinite systems. On the other hand, there is a class of magnetic systems which are diluted magnetic alloys. The magnetic atoms on some lattice sites probably are randomly replaced by non-magnetic atoms. Generally speaking, the magnetic properties of the diluted magnetic systems are comparable with those of the corresponding pure systems. It is known that some new physical phenomena can appear in these magnetic systems. A semi-infinite Ising system with surface dilution was investigated in [2] by means of the effective-field theory. In [7] and in [8] a diluted semi-infinite Ising system was studied using the mean-field theory and renormalization-group method, respectively, and, in [9–11], calculations were made for diluted Ising thin films. All the studies mentioned above are concerned with Ising systems with $s = \frac{1}{2}$. For $s = 1$, in [12], in [13] and in [14] the properties of bulk Ising magnetic systems, semi-infinite magnetic systems and Ising magnetic thin films, respectively, were discussed. However, it is difficult to find studies of a diluted transverse Ising film or semi-infinite system with a higher spin.

In this paper, we investigate the phase transition properties of a diluted Ising thin film with $s = 1$. In recent years, various transverse Ising models have attracted the attention

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of theoretical physicists, and these models have been used to describe the properties of many practical systems [15–18]; therefore we also place this system in a transverse field to examine the influence of the transverse field on the phase transition properties. We use the effective-field method, which contains partly correlations between spins and is more accurate than the mean-field method [20] when it is used to calculate the critical temperature. In our system, the transverse fields on all magnetic atoms are the same, the concentration of magnetic atoms in the film is homogeneous, but the nearest-neighbour coupling constant J_s between surface atoms is different from that between internal atoms.

2. Method

We take a simple-cubic lattice with two free surfaces (001) as our model system. When some of its lattice sites are occupied by non-magnetic atoms, these sites are empty. We apply the Ising model with $s = 1$ to describe our diluted magnetic system and place it in a transverse field. When the transverse field is equal to zero, the system becomes an ordinary diluted Ising magnetic film with $s = 1$. The Hamiltonian of the system can be written as

$$H = -\sum_{ij} J_{ij} S_i^z S_j^z \xi_i \xi_j - Q \sum_i S_i^x \xi_i \tag{1}$$

where J_{ij} is the exchange interaction between two nearest-neighbour magnetic atoms. S^z and S^x are the components of a spin operator S. Q represents the transverse field. ξ_i is the occupation number on lattice site i; $\xi_i = 1$ if the lattice site is occupied by a magnetic atom and $\xi_i = 0$ if the lattice site is occupied by a non-magnetic atom. The nearest-neighbour coupling constant J_{ij} is equal to J_s if sites i and j are on the surface and to J otherwise.

From equation (1), according to the basic framework of the effective-field theory, we can easily obtain

$$\langle S_i^z \rangle = \left\langle \prod_j \xi_j [(S_j^z)^2 \cosh(DJ_{ij}) + S_j^z \sinh(DJ_{ij}) + 1 - (S_j^z)^2] + 1 - \xi_j \right\rangle F(x) \Big|_{x=0} \tag{2}$$

where $D = \partial/\partial x$ is a differential operator and the expression for $F(x)$ is as follows:

$$F(x) = \frac{x}{(x^2 + Q^2)^{1/2}} \frac{2\sinh[\beta(x^2 + Q^2)^{1/2}]}{1 + 2\cosh[\beta(x^2 + Q^2)^{1/2}]} \tag{3}$$

where $\beta = 1/kT$. From (2), one can see that the formula is similar to equation (2) in [14] except that ξ_j and $1 - \xi_j$ appear in the formula. If we ignore the correlations between spin operators on different lattice sites, we can obtain

$$\langle S_i^z \rangle = \left(\prod_j \xi_j [\langle (S_j^z)^2 \rangle \cosh(DJ_{ij}) + \langle S_j^z \rangle \sinh(DJ_{ij}) + 1 - \langle (S_j^z)^2 \rangle] + 1 - \xi_j \right) F(x) \Big|_{x=0}. \tag{4}$$

Different from the Ising system with $s = \frac{1}{2}$, here there exists a new order parameter $\langle (S_i^z)^2 \rangle$. its expectation can be obtained in the same way as the derivation of $\langle (S_i^z) \rangle$:

$$\begin{aligned} \langle (S_i^z)^2 \rangle &= \left\langle \prod_j \xi_j [(S_j^z)^2 \cosh(DJ_{ij}) + S_j^z \sinh(DJ_{ij}) + 1 - (S_j^z)^2] + 1 - \xi_j \right\rangle G(x) \Big|_{x=0} \\ &= \left(\prod_j \xi_j [\langle (S_j^z)^2 \rangle \cosh(DJ_{ij}) + \langle S_j^z \rangle \sinh(DJ_{ij}) \right. \\ &\quad \left. + 1 - \langle (S_j^z)^2 \rangle] + 1 - \xi_j \right) G(x) \Big|_{x=0} \end{aligned} \tag{5}$$

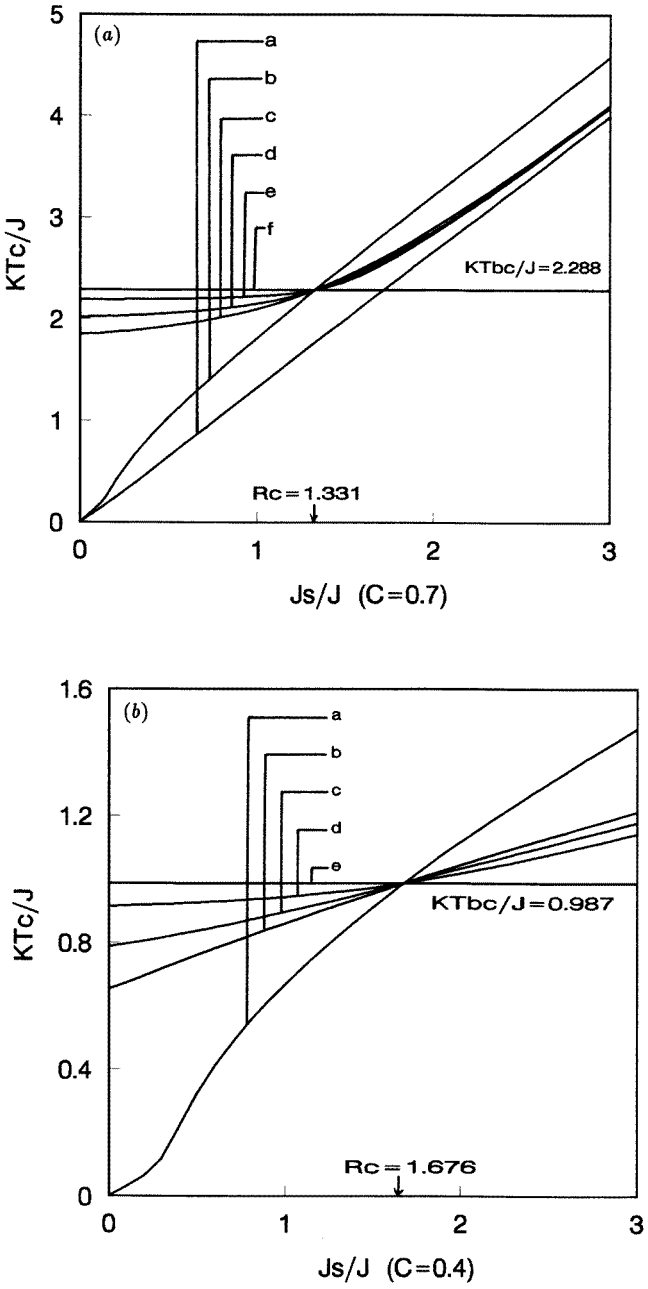

Figure 1. Critical temperature as a function of surface coupling constant for a given concentration c of magnetic atoms: (a) for $C = 0.7$ and the transverse field $Q = 0$ (curve a, $L = 1$; curve b, $L = 2$; curve c, $L = 4$; curve d, $L = 5$; curve e, $L = 8$; curve f, $L = 80$); (b) for $C = 0.4$ and $Q = 0$ (curve a, $L = 2$; curve b, $L = 4$; curve c, $L = 5$; curve d, $L = 8$; curve e, $L = 80$); (c) for $C = 0.33$ and $L = 5$ (curve a, $Q = 0$; curve b, $Q = 0.1$; curve c, $Q = 0.2$; curve d, $Q = 0.3$; curve e, $Q = 0.4$; curve f, $Q = 0.5$). R_c is the critical value of J_s/J and L is the number of layers in the film.

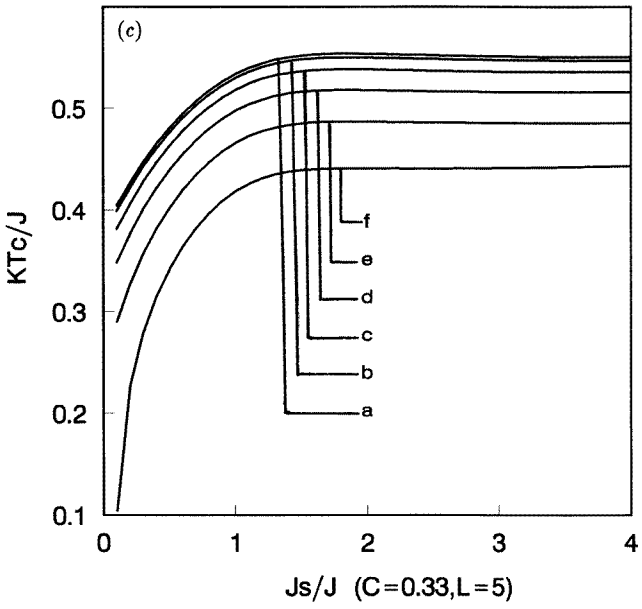

Figure 1. (Continued)

where the expression for $G(x)$ is given by

$$G(x) = \frac{Q^2 + (Q^2 + 2x^2)\cosh[\beta(x^2 + Q^2)^{1/2}]}{(Q^2 + x^2)\{1 + 2\cosh[\beta(x^2 + Q^2)^{1/2}]\}}. \tag{6}$$

This expression for $G(x)$ is different from the corresponding expression in [14], where the expression is incorrect. The correct derivation of (6) has been presented by Kaneyoshi *et al* [19].

Since magnetic atoms are randomly distributed on lattice sites of the system, we should, further, consider the configurational average. The atomic average spin M_i and the order parameter q_i, respectively, can thus be written as

$$M_i = \langle \xi_i \langle S_i^z \rangle \rangle_r = \langle \xi_i \rangle_r \langle\langle S_i^z \rangle\rangle_r = c \langle\langle S_i^z \rangle\rangle_r \tag{7}$$

and

$$q_i = \langle \xi_i \langle (S_i^z)^2 \rangle \rangle_r = \langle \xi_i \rangle_r \langle\langle (S_i^z)^2 \rangle\rangle_r = c \langle\langle (S_i^z)^2 \rangle\rangle_r. \tag{8}$$

Then we have

$$M_i = C \prod_j [q_j \cosh(DJ_{ij}) + M_j \sinh(DJ_{ij}) + 1 - q_j] F(x)|_{x=0} \tag{9}$$

$$q_i = C \prod_j [q_j \cosh(DJ_{ij}) + M_j \sinh(DJ_{ij}) + 1 - q_j] G(x)|_{x=0}. \tag{10}$$

Since the average magnetizations M or the order parameter q on lattice sites in the same layer should be equal, the subscript i can be considered as the layer index.

In this paper, we investigate only the properties of the phase transition and do not calculate the layered magnetizations M_n. Near the critical point, the atomic magnetization of each layer is very small so that we can linearize equation (9), or only linear terms in the atomic magnetizations are retained when M_i is expanded. In principle, one can solve

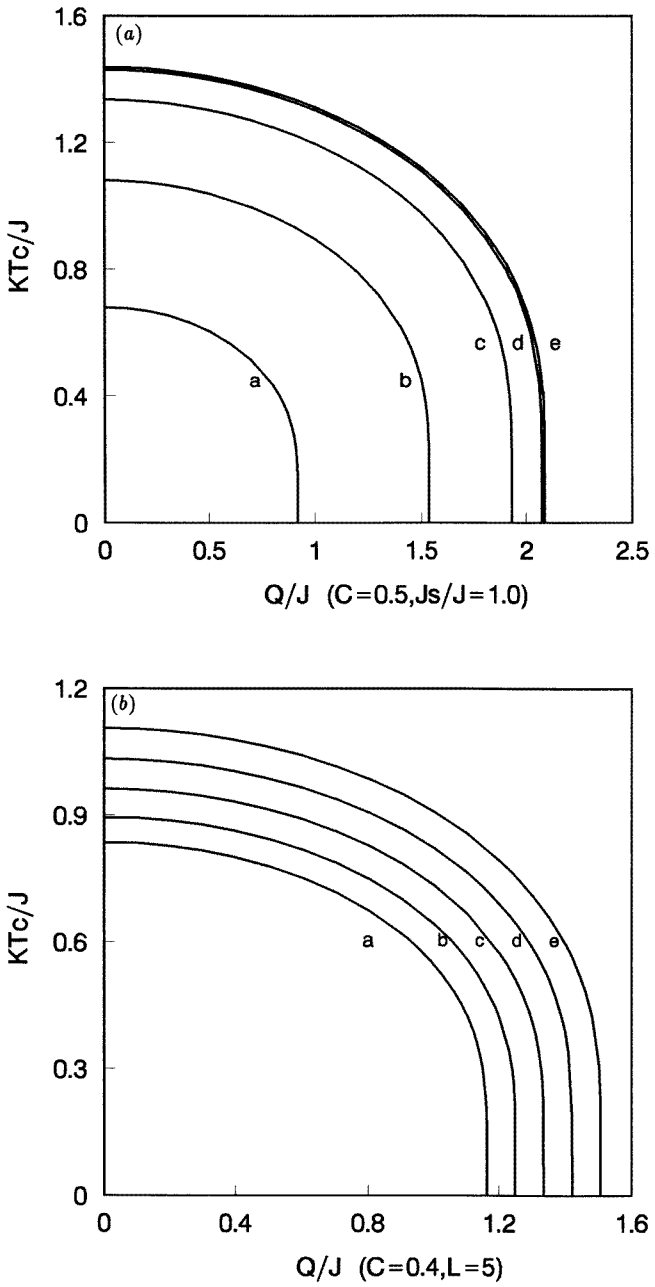

Figure 2. Critical temperature versus transverse field: (a) for $C = 0.5$ and $J_s/J = 1.0$ (curve a, $L = 1$; curve b, $L = 2$; curve c, $L = 5$; curve d, $L = 10$; curve e, $L = 80$); (b) for $C = 0.4$ and $L = 5$ (curve a, $J_s/J = 0.5$; curve b, $J_s/J = 1.0$; curve c, $J_s/J = 1.5$; curve d, $J_s/J = 2.0$; curve e, $J_s/J = 2.5$).

the critical temperature through this procedure, but for a large number L this is laborious and also requires a long time for the computation. A simpler method is to assume that the

q_n $(n = 1, 2, 3, \ldots, L)$ remain unaltered after a certain layer, and here we take $q_n = q$ $(3 \leqslant n \leqslant L-2)$ [14, 23], which allows us to obtain a briefer equation which the critical temperature satisfies. As a result of the symmetry of the film, we should note that $q_1 = q_L$ and $q_2 = q_{L-1}$. If the number L of atomic layers in the film is six or lower, the method becomes accurate. The differently limited layer approximations have been used in many Ising systems and better computation results have been presented [20].

After linearizing the simultaneous equations (9), we have

$$\begin{pmatrix} \frac{1-E_1}{E_2}, -1 & & & & & \\ -1, \frac{1-E_3}{E_4}, -1 & & & & & \\ \cdots & & & & & \\ -1, \frac{1-E_3}{E_4}, -1 & & & & & \\ \cdots & & & & & \\ -1, \frac{1-E_3}{E_4}, -1 & & & & & \\ -1, \frac{1-E_1}{E_2} & & & & & \end{pmatrix} \begin{pmatrix} M_1 \\ M_2 \\ \vdots \\ M_n \\ \vdots \\ M_{L-1} \\ M_L \end{pmatrix} = 0. \tag{11}$$

Here the coefficients E_1, E_2, E_3 and E_4 in the simultaneous equations can be determined from

$$E_1 = 4C[q_1 \cosh(DJ_s) + 1 - q_1]^3[q_2 \cosh(DJ) + 1 - q_2] \sinh(DJ_s) F(x)|_{x=0} \tag{12a}$$

$$E_2 = C[q_1 \cosh(DJ_s) + 1 - q_1]^4 \sinh(DJ) F(x)|_{x=0} \tag{12b}$$

$$E_4 = E_3/4 = C[q \cosh(DJ) + 1 - q]^5 \sinh(DJ) F(x)|_{x=0}. \tag{12c}$$

From equation (10), we obtain the expressions for q_1, q_2 and q at the critical temperature:

$$q_1 = C[q_1 \cosh(DJ_s) + 1 - q_1]^4[q_2 \cosh(DJ) + 1 - q_2] G(x)|_{x=0} \tag{13a}$$

$$q_2 = C[q_2 \cosh(DJ) + 1 - q_2]^4[q_1 \cosh(DJ) + 1 - q_1][q \cosh(DJ) + 1 - q] G(x)|_{x=0} \tag{13b}$$

$$q = C[q \cosh(DJ) + 1 - q]^6 G(x)|_{x=0}. \tag{13c}$$

The critical behaviour of the system can be determined by the condition $\det A = 0$; here $\det A$ is the determinant of the coefficient matrix of equation (11). According to [14], let $x = (1/E_4 - 4)$, $x - a = (1 - E_1)/E_2$, and $x = 2\cos k$; the condition $\det A = 0$ for determining the critical temperature is reduced to

$$\tan(kL) = (r_0 \sin k)/(\cos k - t) \tag{14}$$

where

$$r_0 = (a^2 - 1)/(a^2 + 1) \tag{15}$$

$$t = 2a/(a^2 + 1). \tag{16}$$

Equation (14) is the final equation. Applying this equation we can determine the dependences of the critical temperature on the thickness, transverse field, concentration of magnetic atoms and surface coupling constant.

3. Results and discussion

In this section, we give numerical results by solving (14). As discussed in section 1, our system is an ordinary diluted Ising system with $s = 1$ when the transverse field is equal to zero; it is a diluted transverse Ising ferromagnetic film when the transverse field is not equal to zero.

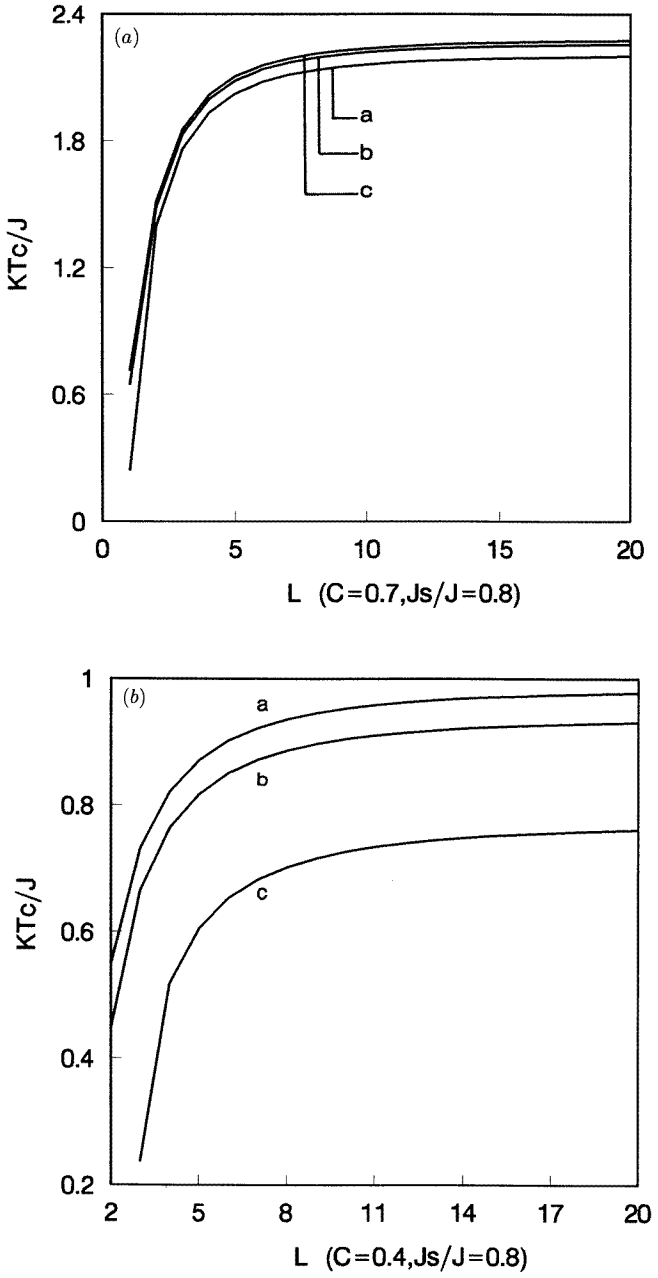

Figure 3. Critical temperature versus film thickness for $J_s/J = 0.8$: (a) for $C = 0.7$ (curve a, $Q = 1.0$; curve b, $Q = 0.5$; curve c, $Q = 0$); (b) for $C = 0.4$ (curve a, $Q = 0$; curve b, $Q = 0.5$; curve c, $Q = 1.0$).

It is known that the exchange interaction between atoms on the surface may be larger than that between two spins in the inside, and also may be smaller than that. For this reason,

we first calculate the critical temperature as a function of the surface coupling constant, and the results can be seen in figure 1. For the transverse field $Q = 0$, figure 1(a) and figure 1(b) correspond to a high concentration ($C = 0.7$) and a low concentration of magnetic atoms ($C = 0.4$), respectively.

First, we discuss figure 1(a). For $L = 1$ (L is the number of layers in the film), our system is a two-dimensional Ising lattice. It is seen from figure 1(a) that the system always has the lowest critical temperature for the same condition, and the critical temperature is proportional to J_s/J. For $L \geqslant 2$, all the curves intersect at the same point ($kT_c/J = 2.288$, $J_s/J = 1.331$), where $R_c = 1.331$ is the critical value of J_s/J. When $J_s/J < R_c$, an increase in the thickness of the film causes the critical temperature to rise. When $J_s/J = R_c$, the critical temperature is independent of the thickness and equal to the value for the corresponding bulk. For $J_s/J > R_c$, the critical temperature decreases as L is increased. Second, we discuss figure 1(b) related to a low concentration of magnetic atoms, i.e. $C = 0.4$. Here the curve for $L = 1$ cannot exist because the concentration is lower than the critical value of the two-dimensional system. In addition, on comparison of figure 1(b) with figure 1(a), R_c is obviously increased but kT_{bc}/J is greatly reduced and the increase in critical temperature with increasing surface coupling constant becomes slower. For a given value of J_s/J and $J_s/J \geqslant R_c$, it is interesting that the critical temperature decreases or is unchanged as the film thickness is increased. In general, the critical temperature increases with increasing film thickness [10, 14, 21, 22]. We think that, because of the stronger surface coupling constant, the influence of the surface layers on the phase transition temperature becomes more important, and for a thicker film the two surface layers possess a lower ratio in the film so that the critical temperature is lower. When we reduce the concentration further to $C = 0.33$, the cross behaviour disappears and we do not give the figure corresponding to this property but present figure 1(c) related to $L = 5$ where the transverse fields have different values. The clear differences between figures 1(b) and 1(a) are as follows: for $J_s/J < 1$, the critical temperature increases rapidly with increasing J_s/J and, for $J_s > 1.5$, it almost does not change with increasing J_s/J and even decreases slightly for smaller transverse field values. Similar features were found by us for the diluted Ising ferromagnetic film with $s = \frac{1}{2}$ [10]. Although the distinction between the concentrations used in figures 1(b) and 1(c) is very small, the features of the curves are obviously different.

Figure 2 shows the dependence of critical temperature on the transverse field. Figure 2(a) displays a set of curves corresponding to different film thicknesses. For a given surface interaction J_s/J and concentration of magnetic atoms, figure 2(b) gives the critical temperature as a function of transverse field for various values of surface interaction. From these figures, one can see that the results are similar qualitatively to those of the pure Ising ferromagnetic film with $s = 1$ in a transverse field [14].

For a given concentration of magnetic atoms and surface coupling constant, the critical temperature as a function of film thickness in different transverse fields is presented in figure 3. Figure 3(a) is related to a high concentration of magnetic atoms and figure 3(b) to a low concentration of magnetic atoms. We find that the influence of the transverse field on the critical temperature is obvious for a low concentration of magnetic atoms and the common feature of the two figures is that, for $L < 5$, the critical temperature increases rapidly as L increases; however, for $L > 8$, it rises slowly as the film thickness increases.

Using equation (14) and letting $kT_c/J = 0$, one can determine the critical concentration of magnetic atoms as a function of other parameters, as shown in figure 4. Figure 4(a) describes the dependence of the critical concentration C_c on the surface coupling constant J_s/J for different transverse fields for $L = 5$. It is very interesting that for the transverse field $Q = 0.0$, or for an ordinary diluted Ising magnetic film with $s = 1$ and $L = 5$, the

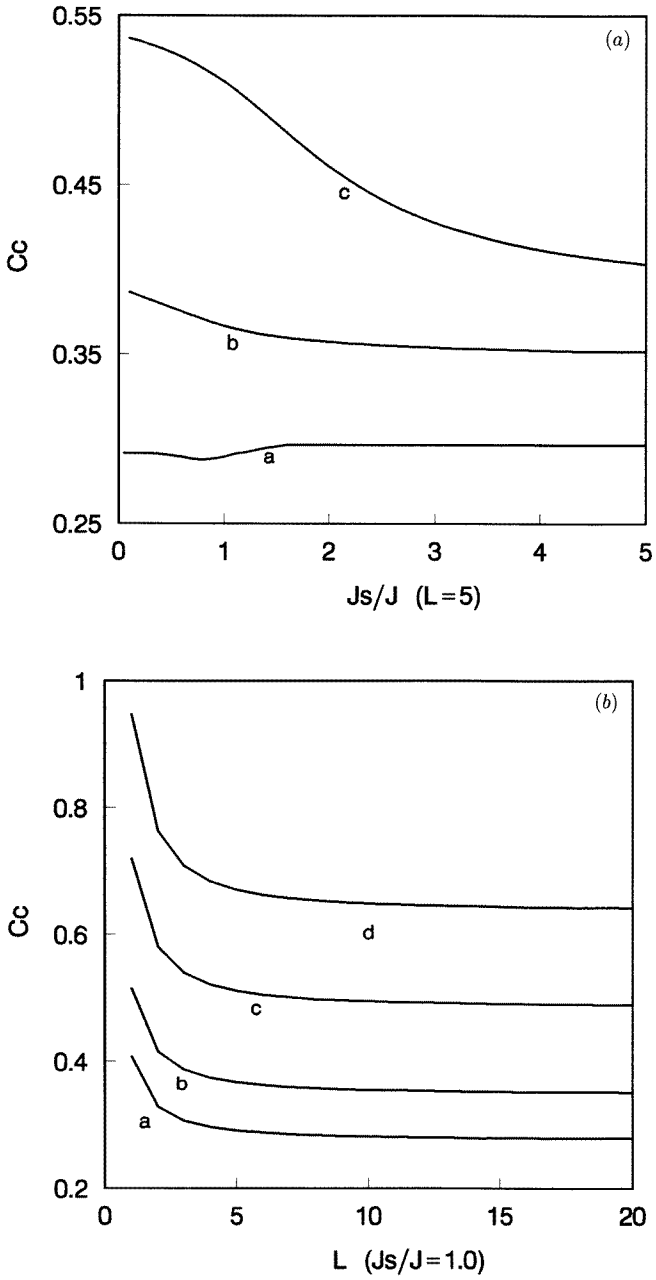

Figure 4. Dependence of critical concentration of magnetic atoms on the surface coupling constant or the film thickness L: (a) for $L = 5$ and different transverse fields (curve a, $Q = 0$; curve b, $Q = 1.0$; curve c, $Q = 2.0$); (b) for $J_s/J = 1$ and different transverse fields (curve a, $Q = 0$; curve b, $Q = 1.0$; curve c, $Q = 2.0$; curve d, $Q = 3.0$).

critical concentration has the lowest value at about $J_s/J = 0.8$ and does not change in the range $J_s/J > 1.5$. The other two curves show that C_c decreases monotonically as J_s/J is

increased. Figure 4(b) shows C_c versus L for $J_s/J = 1$ and for different transverse fields. As L is increased, C_c decreased and this property also exists for a diluted Ising film with $s = \frac{1}{2}$ [12]. Figures 4(a) and 4(b) both show that the critical concentration increases with increasing transverse field.

4. Summary

We have investigated a diluted Ising magnetic film with $s = 1$ in a transverse field and some interesting properties have been found. They are as follows.

(i) There is a critical value R_c of the surface coupling constant J_s/J. When $J_s/J < R_c$, as the film thickness increases, the critical temperature kT_c/J increases. When $J_s/J > R_c$, as the film thickness increases, kT_c/J decreases. However, for $J_s/J = R_c$, kT_c/J does not change with increasing film thickness.

(ii) When the concentration is low enough but higher than the critical concentration, in the range $J_s/J > 1.5$, kT_c/J does not increase with increasing J_s/J.

(iii) The critical concentration as a function of surface coupling constant reaches a minimum for the transverse field $Q = 0.0$. We have also calculated the critical temperature and concentration as functions of other parameters.

The Ising model in a transverse field can be applied to describe ferromagnetic systems with strong uniaxial anisotropy and hydrogen-bonded ferroelectrics [18], but there are few systematic experimental data on corresponding materials, especially diluted magnetic films, for comparing our calculation with the data. Although we use the approximation $q_n = q$ $(3 \leqslant n \leqslant L - 2)$, it is effective only for $L \geqslant 7$. In addition, we see from figure 3 that this approximation does not cause any deviation which can be distinguished.

Acknowledgments

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