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Magnetic properties of ferrimagnetic binary-alloy Ising thin films

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Magnetic properties (phase diagrams, compensation temperatures, and magnetizations) of ferrimagnetic disordered binary alloy Ising films are investigated within the framework of standard mean-field theory. The obtained results show some outstanding features, such as the decrease of the critical temperature with increasing number of layers or the existence of multicompensation points in the system. [S0163-1829(96)03037-8]

I. INTRODUCTION

A number of theoretical and experimental works in the area of magnetic thin films have been stimulated by recent technological progresses. In particular, modern high-vacuum techniques, such as the epitaxial growth techniques, allow one to fabricate very thin magnetic films of controllable thickness[1–3] that may show a number of interesting phenomena not observed in the bulk materials. From the theoretical point of view, the Ising model has been frequently and successfully adopted for the description and understanding of many characteristic features of thin magnetic films. During the last decade various types of Ising thin films have been often investigated by using the standard mean-field theory[4–6] and the effective-field theory[7–10] that correctly account for self-spin correlations. A more elaborated method (so-called third-order Matsudaira approximation) has also been applied for the case of a diluted ferromagnetic Ising thin film.[11] Most of the above-mentioned works have been devoted to the study of thickness dependences of the critical temperature and magnetization. Very recently, some attention has been directed to understanding of the transverse field[12–15] and crystal field[16,17] effects as well as the first-order phase transition[18] of the thin ferromagnetic films. A more complicated model of the ferromagnetic thin film with binary-alloy-type free surface has been studied as well.[19]

Here, one should emphasize that the above-mentioned theoretical works deal exclusively with the ferromagnetic thin films, although experimental data of ferrimagnetically ordered thin films have been recently reported in some experimental works.[20] As far as we know, however, no attempts have been made to study thin films ordered ferrimagnetically. In this work, the standard mean-field approximation is applied to study a ferrimagnetic Ising thin film with the s.c. symmetry. The film consists of n binary-alloy-type layers composed of two kinds of atoms with different spins that are randomly distributed in each layer. In fact, recent theoretical studies of ferrimagnetic disordered binary alloys A_pB_{1-p} have revealed a number of unexpected and interesting results,[21–24] such us the existence of multicompensation points or new types of temperature dependences of magnetization. Therefore it is also interesting to investigate whether similar behaviors may also be observed in binary alloy thin films.

The outline of this paper is as follows. In Sec. II, the general mean-field theory for the description of binary alloy thin films consisting of two kinds (A and B) of atoms with arbitrary spins (S_A and S_B) is developed. In Sec. III, the numerical results for the case of $S_A=1/2$ and $S_B=1$ are presented and discussed. Concluding remarks are given in Sec. IV.

II. FORMULATION

We consider an Ising binary-alloy-type n-layer thin film with the simple cubic (s.c.) symmetry, as depicted in Fig. 1. The system consists of two kinds of magnetic atoms (denoted A and B) with arbitrary spins S_A and S_B that are randomly mixed in each layer. The composition of the system is assumed to be A_pB_{1-p}, where p is the concentration of the A atoms and $(1-p)$ is the concentration of the B atoms. The Hamiltonian of the system is given by

$$\mathcal{H}=-\sum_{i<j} J_{ij}[(\xi_{jA}S^z_{jA}+\xi_{jB}S^z_{jB})\xi_{iA}S^z_{iA} + (\xi_{jB}S^z_{jB}+\xi_{jA}S^z_{jA})\xi_{iB}S^z_{iB}], \tag{1}$$

where the summation is over all nearest-neighbor pairs, and the spin variables S^z_{iA} and S^z_{iB} take $2S_A+1$ and $2S_B+1$ values that are allowed for A and B atoms, respectively. The

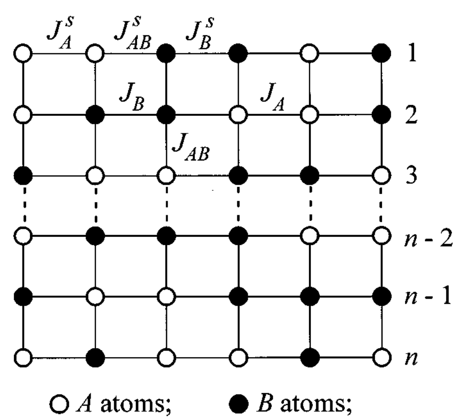

FIG. 1. Part of two-dimensional cross section through the magnetic binary alloy thin film consisting of two kinds of atoms A and B. The exchange parameters are labeled as J_A^s, J_B^s, J_{AB}^s, J_A, J_B, and J_{AB} depending on the position and type of atoms.

exchange interaction parameters J_{ij} depend on the type of bond and the position of atoms, and they can take the following values:

$$J_{ij}=\begin{cases} J_A^s>0, & \text{for the } A\text{–}A \text{ bond in the surface layer;}\\ J_A>0, & \text{for the } A-A \text{ bond otherwise;}\\ J_B^s>0, & \text{for the } B\text{–}B \text{ bond in the surface layer;}\\ J_B>0, & \text{for the } B\text{–}B \text{ bond otherwise;}\\ J_{AB}^s<0, & \text{for the } A\text{–}B \text{ bond in the surface layer;}\\ J_{AB}<0, & \text{for the } A\text{–}B \text{ bond otherwise.} \end{cases}$$

The occupation numbers $\xi_{i\alpha}=0,1$ $(\alpha=A,B)$ depending whether the ith site is occupied by an atom of type α or not, and they satisfy the relations

$$\xi_{iA}+\xi_{iB}=1 \quad \text{and} \quad \langle \xi_{iA}\rangle_c+\langle \xi_{iB}\rangle_c=1, \tag{2}$$

where $\langle \cdots \rangle_c$ denotes the random configurational average. Thus, the A and B atoms are randomly distributed in the thin film with the concentration $\langle \xi_{iA}\rangle_c=p$ and $\langle \xi_{iB}\rangle_c=1-p$, respectively.

Now, let us define the magnetization per site in the kth layer as

$$m_k=pm_{Ak}+(1-p)m_{Bk}, \tag{3}$$

where $m_{ak}=\langle\!\langle \xi_{i\alpha}S_{i\alpha}^z\rangle\!\rangle_c/\langle \xi_{i\alpha}\rangle_c$, and $\langle \cdots \rangle$ means the thermal average. When we use the standard mean-field approximation, then for the ferrimagnetic thin film consisting of n layers one obtains the following set of equations for the magnetizations $m_{\alpha k}$:

$$\begin{aligned} &m_{A1}=S_A\mathcal{F}_A(\beta E_{A1}S_A), \quad m_{B1}=S_B\mathcal{F}_B(\beta E_{B1}S_B),\\ &m_{A2}=S_A\mathcal{F}_A(\beta E_{A2}S_A), \quad m_{B2}=S_B\mathcal{F}_B(\beta E_{B2}S_B),\\ &\qquad\vdots\\ &m_{AK}=S_A\mathcal{F}_A(\beta E_{Ak}S_A), \quad m_{Bk}=S_B\mathcal{F}_B(\beta E_{Bk}S_B),\\ &\qquad\vdots\\ &m_{An}=S_A\mathcal{F}_A(\beta E_{An}S_A), \quad m_{Bn}=S_B\mathcal{F}_B(\beta E_{Bn}S_B). \end{aligned} \tag{4}$$

Here, $\beta=1/(k_BT)$, $\mathcal{F}_\alpha$ denotes the familiar Brillouin function

$$\mathcal{F}_\alpha(x)=\frac{2S_\alpha+1}{2S_\alpha}\coth\left(\frac{2S_\alpha+1}{2S_\alpha}x\right)-\frac{1}{2S_\alpha}\coth\left(\frac{1}{2S_\alpha}x\right),$$

$$\alpha=A \quad \text{or} \quad B, \tag{5}$$

and the mean effective fields $E_{\alpha i}$ are given by

$$\begin{aligned} E_{A1}&=p(4J_A^sm_{A1}+J_Am_{A2})+(1-p)(4J_{AB}^sm_{B1}+J_{AB}m_{B2}),\\ E_{B1}&=p(4J_{AB}^sm_{A1}+J_{AB}m_{A2})\\ &\quad+(1-p)(4J_B^sm_{B1}+J_Bm_{B2}),\\ E_{A2}&=pJ_A(m_{A1}+4m_{A2}+m_{A3})\\ &\quad+(1-p)J_{AB}(m_{B1}+4m_{B2}+m_{B3}),\\ E_{B2}&=pJ_{AB}(m_{A1}+4m_{A2}+m_{A3})\\ &\quad+(1-p)J_B(m_{B1}+4m_{B2}+m_{B3}),\\ &\vdots\\ E_{Ak}&=pJ_A(m_{Ak-1}+4m_{Ak}+m_{Ak+1})\\ &\quad+(1-p)J_{AB}(m_{Bk-1}+4m_{Bk}+m_{Bk+1}),\\ E_{Bk}&=pJ_{AB}(m_{Ak-1}+4m_{Ak}+m_{Ak+1})\\ &\quad+(1-p)J_B(m_{Bk-1}+4m_{Bk}+m_{Bk+1}),\\ &\vdots\\ E_{An}&=p(4J_A^sm_{An}+J_Am_{An-1})\\ &\quad+(1-p)(4J_{AB}^sm_{Bn}+J_{AB}m_{Bn-1}),\\ E_{Bn}&=p(4J_{AB}^sm_{An}+J_{AB}m_{An-1})\\ &\quad+(1-p)(4J_B^sm_{Bn}+J_Bm_{Bn-1}). \end{aligned} \tag{6}$$

In order to obtain the transition temperature T_c, it is necessary to solve the coupled equations (4). For this purpose, we expand the right-hand sides of these equations and consider only terms linear in $m_{\alpha k}$. Then, for the n-layer thin film we obtain the following matrix equation:

$$\mathcal{A}\begin{pmatrix} m_{A1}\\ m_{B1}\\ \vdots\\ m_{An}\\ m_{Bn} \end{pmatrix}=0, \tag{7}$$

where the form of the matrix $\mathcal{A}$ depends only on the thickness of the film and can be easily found from the linearized Eqs. (4). The critical temperature of the system is then determined from the condition

$$\det\mathcal{A}=0. \tag{8}$$

The compensation temperature T_k is a temperature at which the resultant (or total) magnetization vanishes below the critical point. For our system, the total magnetization is given by

$$M=\sum_{k=1}^{n} m_k, \tag{9}$$

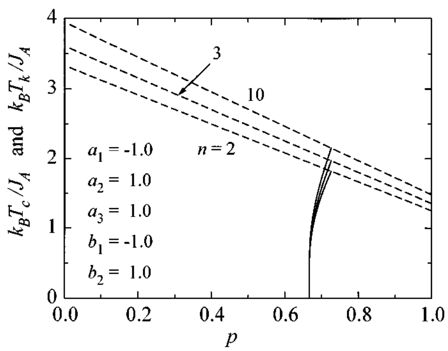

FIG. 2. Concentration dependences of the critical temperature T_c (dashed curves) and the compensation temperature T_k (solid curves) for ferrimagnetic binary alloy thin films of different thicknesses ($n=2,3,10$), when $|a_1|=a_2=a_3=|b_1|=b_2=1.0$.

where m_k means the magnetization in the kth layer defined by (3). Consequently, the compensation temperature of our system is determined from the equation

$$p\sum_{k=1}^{n} m_{Ak}+(1-p)\sum_{k=1}^{n} m_{Bk}=0, \tag{10}$$

in which the magnetizations $m_{\alpha k}$ are solutions of (4). Furthermore, it is useful to note the possibility of the compensation at $T=0$. In the limit of $T\rightarrow 0$ we obtain from Eqs. (4) solutions $m_{Ak}=-S_A$ and $m_{Bk}=S_B,\forall k$ (or $m_{Ak}=S_A$ and $m_{Bk}=-S_B,\forall k$). Substituting these solutions into Eq. (10), one finds that for $T=0$ and nonzero exchange interactions, the compensation always appears in our system at a certain concentration, which is exactly given by

$$p_0=\frac{S_B}{S_A+S_B}. \tag{11}$$

Finally, one should note that the formulation presented in this section is very easily extendable to more complicated systems. For example, if the crystal-field term is included into Hamiltonian (1) all equations in our formulation remain unchanged, but the Brillouin function must be replaced by another function $\mathcal{F}_\alpha(x)$ defined as follows:

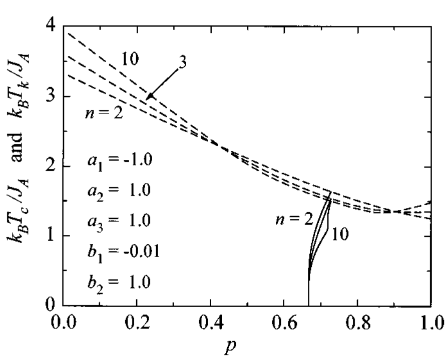

FIG. 3. The same as in Fig. 2 but for $|a_1|=a_2=a_3=b_2=1.0$ and $b_1=-0.01$.

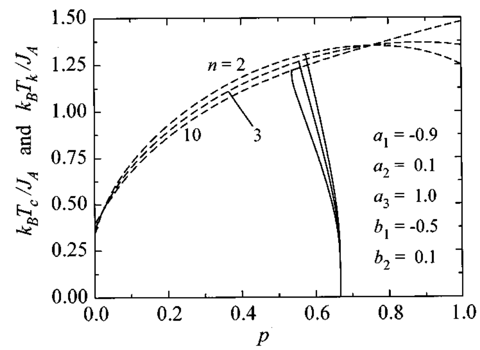

FIG. 4. Concentration dependences of the critical temperature T_c (dashed curves) and the compensation temperature T_k (solid curves) for ferrimagnetic binary alloy thin films of different thicknesses ($n=2,3,10$), when $a_1=-0.9$, $a_2=0.1$, $a_3=1.0$, $b_1=-0.5$, and $b_2=0.1$.

$$\mathcal{F}_\alpha(x)=\frac{\Sigma_{l=-S_\alpha}^{S_\alpha} l\exp(\beta D_\alpha l^2)\sinh(l\beta x)}{\Sigma_{l=-S_\alpha}^{S_\alpha}\exp(\beta D_\alpha l^2)\cosh(l\beta x)}, \tag{12}$$

where D_α denotes the crystal-field parameter.

III. NUMERICAL RESULTS

In this section, we present numerical results for the phase diagrams (T_c), compensation temperatures (T_k), and magnetizations of the n-layer ferrimagnetic thin film selecting $S_A=1/2$ and $S_B=1$. For this aim, it is convenient to introduce the following dimensionless parameters:

$$a_1=J_{AB}^s/J_A,\quad a_2=J_B^s/J_A,\quad a_3=J_A^s/J_A,$$
$$b_1=J_{AB}/J_A,\quad b_2=J_B/J_A. \tag{13}$$

Of course, the behavior of our thin-film system will depend on the values of a_i, b_i as well as the thickness of the film. In fact, many numerical calculations have been done selecting representative sets of the parameters a_i and b_i, however, it is clearly impossible to present all the results. Therefore, most of the results that are qualitatively similar as those of bulk A_pB_{1-p} binary alloy[25] have been excluded from the present analysis. Before discussing the results, it is also worth noticing that in the real rare-earth (RE)/transition metal (TM) ferrimagnetic alloys the exchange interactions J_A, J_{AB}, and J_B correspond to TM-TM, RE-TM, and RE-RE interactions, respectively. The magnitudes of interaction usually satisfy the relation $J_B<|J_{AB}|<J_A$. Accordingly, in the following, let us study the systems satisfying the inequality and examine whether some interesting phenomena can be observed in them.

At first, Figs. 2 and 3 show the concentration dependences of critical and compensation temperatures for thin films of different thicknesses ($n=2$, 3, and 10) for two sets of parameters. In Fig. 2, we have selected $|a_1|=a_2=a_3=|b_1|=b_2=1.0$. In the figure, both the critical and compensation temperature exhibit behaviors very similar to those of usual A_pB_{1-p} disordered binary alloys.[25] In particular, one should notice that the compensation temperature T_k may increase with the increase of the thickness in the thin film, when the concentration is fixed, for instance, at $p=0.7$. Such

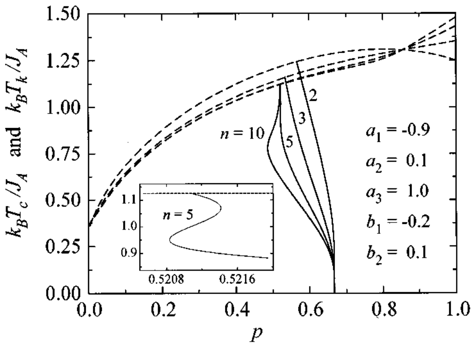

FIG. 5. Concentration dependences of the critical temperature T_c (dashed curves) and the compensation temperature T_k (solid curves) for ferrimagnetic binary alloy thin films of different thicknesses ($n=2,3,5,10$), when $a_1=-0.9$, $a_2=0.1$, $a_3=1.0$, $b_1=-0.2$, and $b_2=0.1$. The inset shows in detail the dependence of the compensation temperature for the five-layer thin-film system ($n=5$) near the critical boundary line.

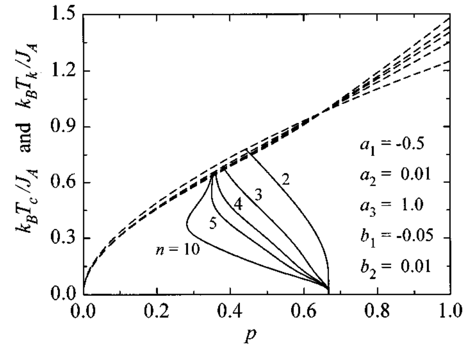

FIG. 7. The same as in Fig. 6 but for $a_1=-0.5$, $a_2=0.01$, $a_3=1.0$, $b_1=-0.05$, and $b_2=0.01$.

a behavior may be observed experimentally in a ferrimagnetic thin film when the film thickness increases. On the other hand, some characteristic results are found by selecting the small values of the parameter b_1, as illustrated in Fig. 3 for the system with $b_1=-0.01$. Namely, in a certain range of concentration (roughly estimated $0.45<p<0.92$) the critical temperature decreases with increasing thickness of the thin film. The behavior may appear in the system when one of the exchange parameters J_{AB} or J_B becomes weak (i.e., $|b_1|\ll 1$ or $b_2\ll 1$) and the region in which this phenomenon can be observed depends on the values of other exchange parameters. In Fig. 3, an interesting behavior is observed for the concentration variations of the compensation temperature: the possibility of two compensation points appears in the system when the thickness of the film becomes thick enough. As far as we know, these phenomena have not been reported in the literature of thin magnetic films.

Let us now investigate whether such a characteristic behavior can be observed in the system which satisfies the inequality $J_B<|J_{AB}|<J_A$. The case is illustrated in Figs. 4–6 for the system with the values of exchange parameters fixed as $a_1=-0.9$, $a_2=b_2=0.1$, $a_3=1.0$, when some typical values of the parameter b_1 are selected. From the figures, one can see that the critical temperature decreases with increasing thickness of the thin film in a wide concentration region because both J_{AB} and J_B are weak. In particular, two or three compensation points can be observed in the system when the parameter b_1 takes a small value, such as in Fig. 5. The inset of Fig. 5 shows in detail the case when three compensation points are possible. Generally speaking, one can find the following important fact: As illustrated in Figs. 6 and 7, the multicompensation points can be rather easily found for $n\geqslant 4$, when the exchange parameter J_{AB} takes very small values. Then, notice that the condition $|J_{AB}^s|\geqslant|J_{AB}|$ seems to be of principal importance for the appearance of the multicompensation points in ferrimagnetic thin films. Although it is impossible to prove this condition analytically, we could not find multicompensation points when this condition has not been valid. Thus, the results presented in this section indicate that multicompensation points can be easily found in the ferrimagnetic films of rather great thickness (for example, $n=10$). From this fact, one can expect that similar behaviors may be found for the bulk disordered A_pB_{1-p} alloy with a free surface. Further, one can see from the figures that the compensation temperature reduces independently of n to zero at $p_0=2/3$ in agreement with Eq. (11).

Finally, to prove whether the predictions of the phase diagrams obtained in Figs. 2–7 are correct or not, it is necessary to study the temperature dependence of the total magnetization (9). The temperature variations of the total and layer

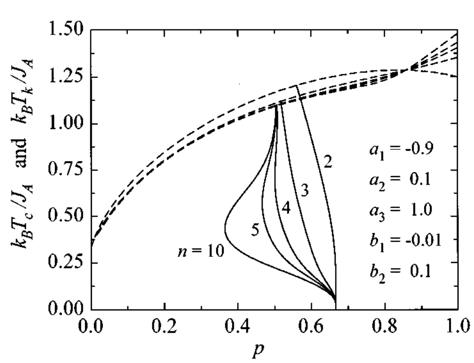

FIG. 6. Concentration dependences of the critical temperature T_c (dashed curves) and the compensation temperature T_k (solid curves) for ferrimagnetic binary alloy thin films of different thicknesses ($n=2,3,4,5,10$), when $a_1=-0.9$, $a_2=0.1$, $a_3=1.0$, $b_1=-0.01$, and $b_2=0.1$.

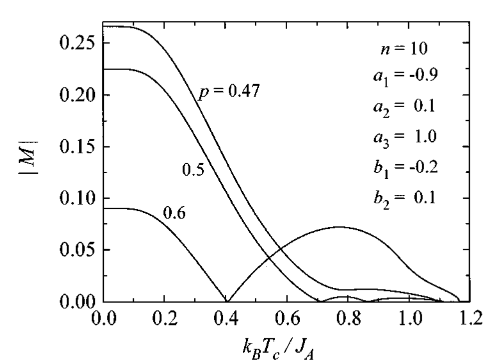

FIG. 8. Temperature variations of the absolute value of the total magnetization for the n-layer ferrimagnetic binary alloy thin film, when $a_1=-0.9$, $a_2=0.1$, $a_3=1.0$, $b_1=-0.2$, $b_2=0.1$, and the concentration p is changed.

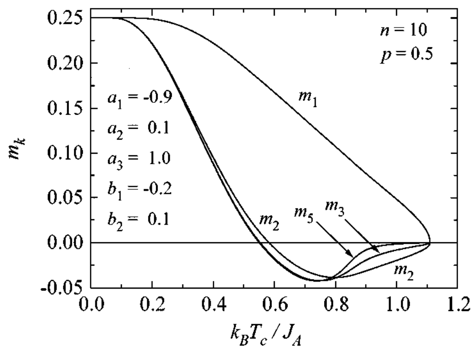

FIG. 9. Temperature variations of the layer magnetizations corresponding to the system of Fig. 8 labeled as $p=0.5$.

magnetizations are presented in Figs. 8 and 9 for the system that exhibits two compensation points. The results are equivalent to the predictions of T_k and T_c obtained from the phase diagram (or Fig. 5). In particular, the magnetization curves labeled $p=0.49$ and 0.6 exhibit some characteristic features. Thus, the results of Fig. 8 represent the types of magnetization that have not been predicted in the classical Néel theory.[26] However, notice that they are qualitatively very similar as those reported in Refs. 21–24 for other systems. In Fig. 9, the corresponding layer magnetizations are plotted. The layer magnetizations exhibit also their own compensation points that are, of course, not directly related to the compensation point of the total magnetization.

IV. CONCLUSION

In this work, we have studied the ferrimagnetic binary alloys thin films consisting of two kinds of Ising-type atoms. In Sec. II, we have developed the general mean-field theory which can be applied for studying various effects in the systems under consideration. The detailed numerical analysis has been restricted to the systems with fixed spin values, namely, $S_A=1/2$ and $S_B=1$, and the most interesting results have been presented in Sec. III. Our investigation has revealed some results that have not been predicted for the thin magnetic films. In particular, we have found that in some concentration regions the critical temperature can decrease with increasing thickness of the thin film, when the exchange interaction J_{AB} or J_B takes a small value. Of course, the critical temperature of the system under investigation is independent of the signs of J^s_{AB} and J_{AB}, and hence our results for the phase boundaries are also valid for ferromagnetic binary alloy thin films. From the investigation of the compensation temperature, on the other hand, we can conclude that multicompensation points can be found in the ferrimagnetic binary alloy thin films when the condition $|J^s_{AB}|\geqslant|J_{AB}|$ is fulfilled. As shown in Figs. 8 and 9, the new types of temperature dependences of the total magnetization can be observed in the system. At this place, one should notice that the investigation of compensation points becomes important in connection with possible technological applications, since some of ferrimagnetic materials are used as recording media. We hope that systems similar to those discussed in this paper may be prepared experimentally, so that the experimental search of the multicompensation points will be possible in the future.

Finally, the formulation presented in this paper can be also extended to include the crystal- or transverse-field effects that may lead to some interesting results. Some of these problems will be studied in the near future.

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