

A diluted mixed spin-2 and spin-5/2 ferrimagnetic Ising system; a study of a molecular-based magnet

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Abstract. The magnetic properties of a diluted spin-2 and spin-5/2 ferrimagnetic Ising system are investigated on the basis of the effective-field theory with correlations. In particular, the effect of a positive single-ion anisotropy D on the compensation temperature in a pure system with D only on spin-5/2 atoms is investigated, in order to clarify the characteristic feature of the temperature dependence of the total magnetization M observed in a molecular-based magnetic material, $N(n-C_4H_9)_4Fe^{II}Fe^{III}(C_2O_4)_3$. The influences of D and the concentrations of magnetic atoms on the properties of the system on a honeycomb lattice are examined. The results show that several (two or three) compensation points are possible in the diluted system with special values of D and concentrations of magnetic atoms.

1. Introduction

A variety of mixed spin Ising systems consisting of two kinds of magnetic atom, A and B atoms with spins S_A and S_B , have been studied theoretically by means of different theoretical methods [1–5]. Most of these studies treat the system with $S_A = 1/2$, $S_B = S$ ($S > 1/2$) and a single-ion anisotropy constant D for the B atom. The Hamiltonian of the system is given by

$$H = J \sum_{ij} S_{iA}^z S_{jB}^z - D \sum_j (S_{jB}^z)^2 \quad (1)$$

where the first term runs over only the nearest-neighbour pairs of atoms and J is the exchange interaction. In these ferrimagnetic systems with $J > 0$, particular attention has been paid recently to the possibility of many compensation points [3, 6]. The exact solutions for the possibility of two compensation points have been found in [4, 5].

A number of experimental studies have accumulated recently in the area of molecular-based magnetic materials, and the magnetic properties—that is, the molecular magnetism—have become an important focus of scientific interest. Among these materials, many bimetallic molecular-based magnetic materials in which two kinds of magnetic atom, A and B, alternate regularly have exhibited ferrimagnetic properties and seem to be rather well interpreted by the use of the mixed spin (Heisenberg or Ising) model [7]. Most of these ferrimagnetic materials have not exhibited a compensation point. But, in the study of the compounds $AM^{II}Fe^{III}(C_2O_4)_3$ ($A = N(n-C_3H_7)_4$, $M^{II} = Mn, Fe$), whose structures are two-dimensional honeycomb networks [8], a remarkable behaviour, namely the occurrence of apparently negative magnetization at low temperature, has been observed for the $Fe^{II}Fe^{III}$ compounds and this phenomenon has been analysed by assuming the so-called Néel model

[9] to be appropriate: $S_A = 5/2$ for Fe^{III} and $S_B = 2$ for Fe^{II} . The magnetization of the Fe^{III} sublattice follows the Brillouin curve in the molecular-field approximation (MFA), and the Fe^{II} sublattice magnetization increases more steeply below the transition temperature T_C than that of Fe^{III} because of the positive single-ion anisotropy ($D > 0$) of the Fe^{II} ion due to spin-orbit coupling. According to the proposed model, the system may be described by the Hamiltonian (1) with $S_A = 5/2$ for Fe^{II} , $S_B = 2$ for Fe^{III} and $D > 0$. As far as we are aware, however, whether a compensation point exists or not for such a system has not been discussed.

On the other hand, a diluted mixed spin-1/2 and spin-1 ferrimagnetic Ising system [10], a diluted mixed spin-1/2 and spin-3/2 ferrimagnetic Ising system [11] and a diluted mixed spin-1 and spin-3/2 ferrimagnetic Ising system [12] have been discussed on the basis of the effective-field theory with correlations (EFT) introduced by one of the present authors (TK) [13]. The existence of many (two, three, four) compensation points for these systems has also been proposed.

The aim of this work is to study the magnetic properties of a diluted mixed spin-5/2 and spin-2 ferrimagnetic Ising system on a honeycomb lattice within the framework of the EFT, in order to clarify the characteristic behaviour of the molecular-based magnet, $\text{N}(n\text{-C}_4\text{H}_9)_4\text{Fe}^{\text{II}}\text{Fe}^{\text{III}}(\text{C}_2\text{O}_4)_3$. By extending the Hamiltonian (1) to the diluted system, we get

$$H = J \sum_{ij} S_{iA}^z S_{jB}^z \xi_{iA} \xi_{jB} - D \sum_j (S_{jB}^z)^2 \xi_{jB} \quad (2)$$

where S_{iA}^z takes the values $\pm 5/2, \pm 3/2, \pm 1/2$, S_{jB}^z can be $\pm 2, \pm 1, 0$, and ξ_{iA} (or ξ_{jB}) is the site occupancy number which takes the value unity or zero, depending on whether the site i (or j) is occupied by a magnetic atom of type A (or B) or not.

In section 2, we present the formulation of the system in the EFT. In section 3, the effect of a positive single-ion anisotropy D on the compensation temperature for the pure (nondiluted) system is investigated, in order to clarify the characteristic behaviour of the temperature dependence of the total magnetization M observed for $\text{N}(n\text{-C}_4\text{H}_9)_4\text{Fe}^{\text{II}}\text{Fe}^{\text{III}}(\text{C}_2\text{O}_4)_3$. In section 4, the possibility of many compensation points for the diluted system is also examined.

2. Formulation

We consider a diluted two-sublattice ferrimagnetic Ising system on a honeycomb lattice described by the Hamiltonian (2). The total magnetization M of the system is

$$M = \frac{N}{2}(pm_A + cm_B) \quad (3)$$

where N is the total number of lattice points and p (or c) is the concentration of magnetic A (or B) atoms defined by $p = \langle \xi_{iA} \rangle_r$ (or $c = \langle \xi_{iB} \rangle_r$). The sublattice magnetizations m_A and m_B are defined by

$$m_A = \frac{\langle \langle \xi_{iA} S_{iA}^z \rangle \rangle_r}{\langle \xi_{iA} \rangle_r} \quad m_B = \frac{\langle \langle \xi_{iB} S_{iB}^z \rangle \rangle_r}{\langle \xi_{iB} \rangle_r} \quad (4)$$

where $\langle \dots \rangle$ and $\langle \dots \rangle_r$ denote the thermal and random averages, respectively.

Within the EFT (see the review article [13] for the EFT, or the appendix), the sublattice magnetizations m_A and m_B are given by

$$\begin{aligned} m_A &= \left[1 - c + c \left\{ \cosh(J\eta_B \nabla) - \frac{m_B}{\eta_B} \sinh(J\eta_B \nabla) \right\} \right]^3 F_A(x) \Big|_{x=0} \\ m_B &= \left[1 - p + p \left\{ \cosh(J\eta_A \nabla) - \frac{m_A}{\eta_A} \sinh(J\eta_A \nabla) \right\} \right]^3 F_B(x) \Big|_{x=0} \end{aligned} \quad (5)$$

where $\nabla = \partial/\partial x$ is a differential operator. The parameters η_A and η_B are given by

$$\begin{aligned} (\eta_A)^2 &= \left[1 - c + c \left\{ \cosh(J\eta_B \nabla) - \frac{m_B}{\eta_B} \sinh(J\eta_B \nabla) \right\} \right]^3 G_A(x) \Big|_{x=0} \\ (\eta_B)^2 &= \left[1 - p + p \left\{ \cosh(J\eta_A \nabla) - \frac{m_A}{\eta_A} \sinh(J\eta_A \nabla) \right\} \right]^3 G_B(x) \Big|_{x=0} \end{aligned} \quad (6)$$

where the functions $F_\alpha(x)$ and $G_\alpha(x)$ ($\alpha = A$ or B) are respectively given by

$$\begin{aligned} F_A(x) &= \frac{1}{2} \frac{5 \sinh(2.5\beta x) + 3 \sinh(1.5\beta x) + \sinh(0.5\beta x)}{\cosh(2.5\beta x) + \cosh(1.5\beta x) + \cosh(0.5\beta x)} \\ F_B(x) &= \frac{4 \sinh(2\beta x) + 2 \exp(-3\beta D) \sinh(\beta x)}{2 \cosh(2\beta x) + 2 \exp(-3\beta D) \cosh(\beta x) + \exp(-4\beta D)} \end{aligned} \quad (7a)$$

$$\begin{aligned} G_A(x) &= \frac{1}{4} \frac{25 \cosh(2.5\beta x) + 9 \cosh(1.5\beta x) + \cosh(0.5\beta x)}{\cosh(2.5\beta x) + \cosh(1.5\beta x) + \cosh(0.5\beta x)} \\ G_B(x) &= \frac{8 \cosh(2\beta x) + 2 \exp(-3\beta D) \cosh(\beta x)}{2 \cosh(2\beta x) + 2 \exp(-3\beta D) \cosh(\beta x) + \exp(-4\beta D)} \end{aligned} \quad (7b)$$

with $\beta = 1/k_B T$.

Here, the transition temperature T_C of the system can be obtained by the standard method (the linearization of sublattice magnetizations in (5) and (6)); by solving the relation

$$1 = 9pc \frac{I_A I_B}{\eta_A^0 \eta_B^0} \quad (8)$$

with

$$\begin{aligned} I_A &= \sinh(J\eta_B^0 \nabla) [1 - c + c \cosh(J\eta_B^0 \nabla)]^2 F_A(x) \Big|_{x=0} \\ I_B &= \sinh(J\eta_A^0 \nabla) [1 - p + p \cosh(J\eta_A^0 \nabla)]^2 F_B(x) \Big|_{x=0} \end{aligned} \quad (9)$$

numerically, we can get T_C , where the parameters η_A^0 and η_B^0 are determined from

$$\begin{aligned} (\eta_A^0)^2 &= [1 - c + c \cosh(J\eta_B^0 \nabla)]^3 G_A(x) \Big|_{x=0} \\ (\eta_B^0)^2 &= [1 - p + p \cosh(J\eta_A^0 \nabla)]^3 G_B(x) \Big|_{x=0}. \end{aligned} \quad (10)$$

The compensation temperature T_κ , if it exists for the system, can be determined from the relation

$$\frac{M}{N} = 0 \quad (11)$$

below T_C .

3. A molecular-based magnet

In this section, let us study the characteristic behaviour of a molecular-based magnet, $N(n-C_4H_9)_4Fe^{II}Fe^{III}(C_2O_4)_3$, on the basis of the formulation (EFT) in section 2. Since the system is not diluted by nonmagnetic atoms, T_C and T_K can be determined from (8) and (11) by substituting $p = 1$ and $c = 1$ into them.

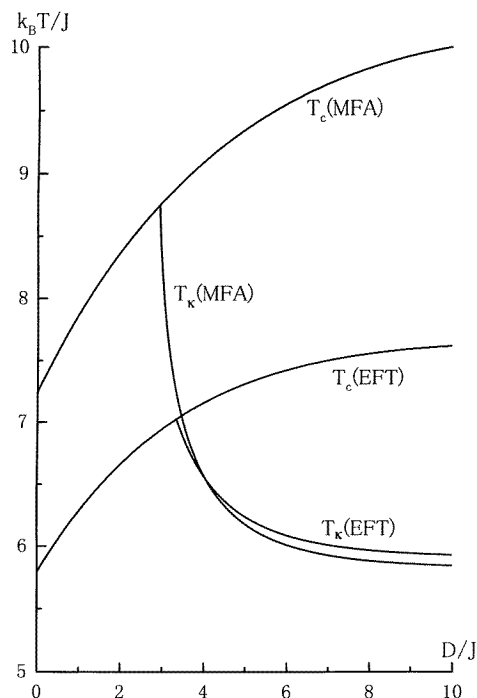


Figure 1. The variations of the transition temperature T_C and the compensation temperature T_K with the change of D/J in the pure ($p = 1.0$ and $c = 1.0$) mixed spin-5/2 and spin-2 ferrimagnetic Ising system on a honeycomb lattice, when the two theoretical frameworks are used, namely the effective-field theory with correlations (EFT) and the mean-field approximation (MFA).

The variations of T_C and T_K versus the single-ion anisotropy constant D of B atoms ($D/J \geq 0$) in the ferrimagnetic honeycomb lattice are depicted in figure 1. In the figure, the results of using the MFA are also depicted for comparison, where, instead of (5), the sublattice magnetizations in the MFA are given by

$$m_A = -F_A(3Jm_B) \quad m_B = -F_B(3Jm_A). \quad (12)$$

As is seen from the figure, the dependences of T_K on the value of D/J are very similar in the two theories. The value of T_C is revised in a reasonable way when the EFT is used instead of the MFA. As a result, the critical value D_C of D above which the compensation point may appear is increased from $D_C/J = 2.934$ for the MFA to $D_C/J = 3.349$ for the EFT. This implies that the critical D_C/J takes a value larger than $D_C/J = 3.349$ for the EFT, when a better approximation is applied. Thus, the mixed spin-5/2 and spin-2 ferrimagnetic Ising system on a honeycomb lattice may exhibit a compensation point below the transition temperature when the value of D/J is larger than a critical value D_C/J .

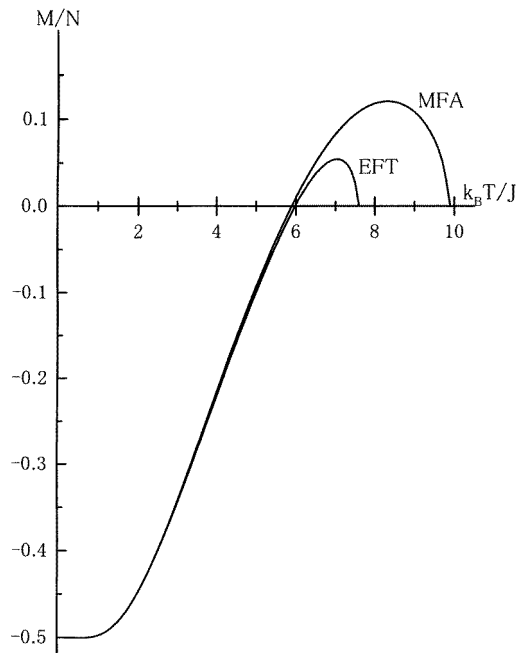


Figure 2. The temperature dependences of M in the mixed spin ferrimagnetic Ising honeycomb lattice with $S_A = 5/2$, $S_B = 2$ and $D/J = 8.0$, when the two theoretical frameworks (EFT and MFA) are used.

In figure 2, the temperature dependences of M in the pure system with $p = 1$ and $c = 1$ for the EFT as well as the MFA are shown; they were obtained by selecting the value $D/J = 8.0$ ($D/J > D_C/J$) and solving the coupled equations (5) and (12) numerically. The curves are similar to that predicted in [8] for the compound $N(n\text{-C}_4\text{H}_9)_4\text{Fe}^{\text{II}}\text{Fe}^{\text{III}}(\text{C}_2\text{O}_4)_3$ with $S_A = 5/2$ (Fe^{III}) and $S_B = 2$ (Fe^{II}).

Now, it is important to note the following facts. As shown in figure 1, the value of T_κ seems to be rather insensitive to the choice of theoretical approximation, if it exists for the system. When the value of S_B for the system with $S_A = 5/2$ and $S_B = 2$ is replaced by $S_B = 1$ or $3/2$, no compensation point could be found in the region of $D/J \geq 0$ within the framework of the MFA. The same situation is also observed for the mixed spin-1 and spin-3/2 ferrimagnetic honeycomb lattice with $D/J \geq 0$ as well as a mixed spin-1/2 and spin- S ($S > 1/2$) ferrimagnetic system with $D/J \geq 0$. In other words, the mixed spin-5/2 and spin-2 ferrimagnetic Ising system described by (1) with $D/J \geq 0$ is a special system in the class of mixed spin ferrimagnetic systems, since a compensation point can be observed when $D/J > D_C/J > 0$. Thus, the single-ion anisotropy constant D/J for the compound $N(n\text{-C}_4\text{H}_9)_4\text{Fe}^{\text{II}}\text{Fe}^{\text{III}}(\text{C}_2\text{O}_4)_3$ must be larger than the critical value D_C/J when the model proposed in [8] is valid for the material. This fact also indicates that the Ising model described by (1) may be a good starting Hamiltonian for discussions of the compound.

4. A diluted system

In this section, let us study the magnetic properties (indicated by T_C , T_κ and M) of the mixed spin-5/2 and spin-2 ferrimagnetic Ising system on a honeycomb lattice, when the

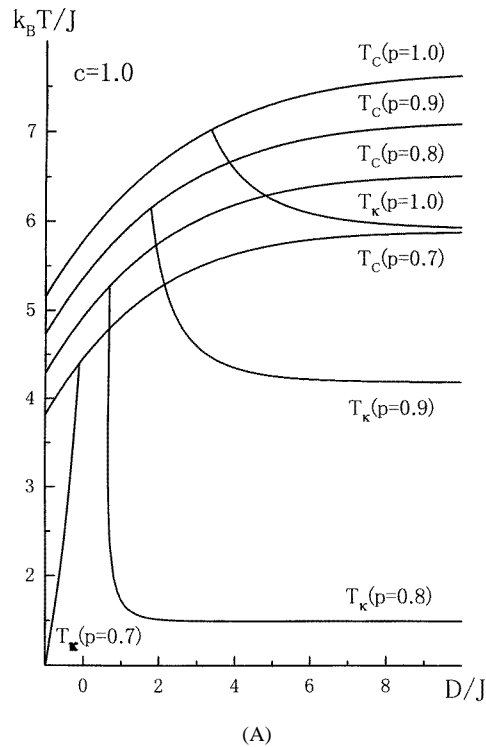


Figure 3. Phase diagrams of the diluted mixed spin ferrimagnetic system on a honeycomb lattice in the T - D plane: (A) when $c = 1$ and the value of p is changed from $p = 1$ to $p = 0.7$; (B) when $c = 1$ and $p = 0.8$; and (C) when $c = 0.8$ and four values of p are selected: $p = 0.8, 0.7, 0.6$ and 0.5 .

system is diluted by the two types of nonmagnetic atom ($p < 1.0$ and $c < 1.0$). The numerical results can be obtained by solving the relations of the EFT given in section 2.

In figure 3, we show how the dilution affects the phase diagram of figure 1, selecting some typical cases; figure 3(A) is plotted for the system with positive D -values where c is fixed at $c = 1$ and p is changed from $p = 1$ to $p = 0.7$. With the decrease of p , the T_K -curve may be changed dramatically, although the T_C -curve takes the same form as that for $p = 1$. In particular, the T_K -curve labelled $p = 0.8$ exhibits the possibility of two compensation points. In order to show this clearly, the T_C - and T_K -curves for the system with $c = 1$ and $p = 0.8$ are also plotted in figure 3(B) on expanded scales. The results indicate that the system with a value of D near $D/J = 0.65$ can exhibit two compensation points below T_C . Next, the features of the T_C - and T_K -curves are depicted in figure 3(C) for the system with a fixed value of c of 0.8, for four selected values of p . The T_K -curve labelled $p = 0.5$ in the figure also indicates that two compensation points are also possible for the system with a small negative value of D .

In figure 4, the behaviour of the T_C - and T_K -curves in T - p space is depicted for the diluted ferrimagnetic system with a fixed value ($c = 0.9$) of the concentration, for selected typical values of D/J . The five values of D/J that are selected for figure 4(A) are $D/J = 1.0, 0.75, 0.5, 0.25$ and 0.0 . We can see that at a definite D , the more the sublattice is diluted, the lower the transition temperature T_C is, and there is a critical concentration p_C at which T_C reduces to zero. In the vicinity of p_C , furthermore, the systems with

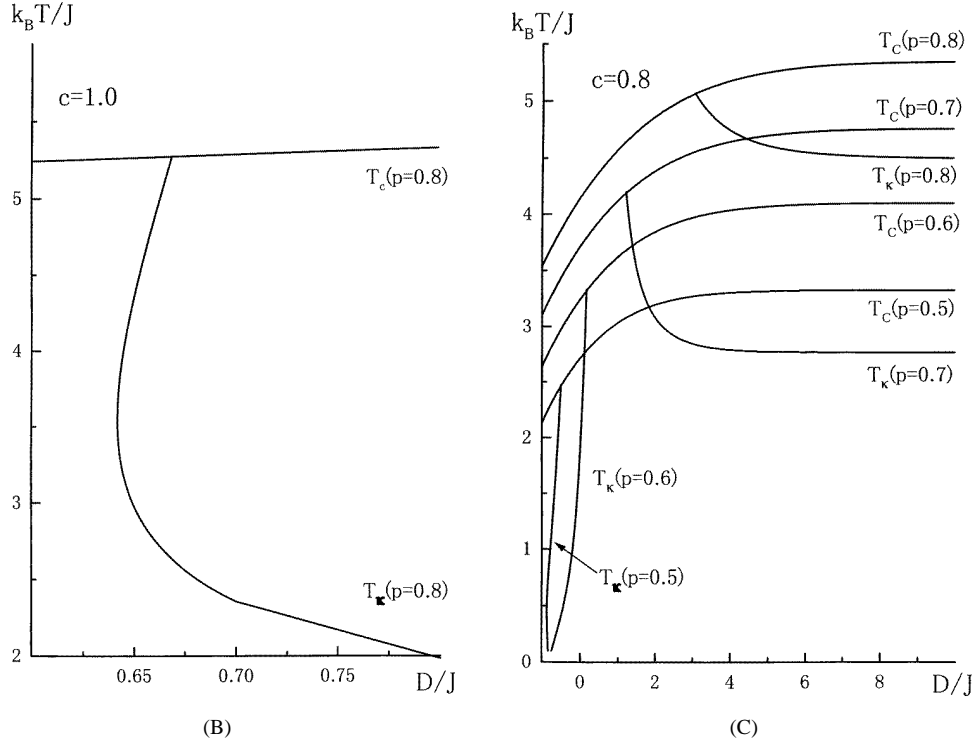


Figure 3. (Continued)

$D/J = 0.75, 0.5$ and 0.25 may show re-entrance for T_C . These features of T_C as well as T_K in the figure have been obtained in the phase diagrams of the diluted mixed spin-1/2 and spin-1 ferrimagnetic Ising honeycomb system with a negative value of D [10]. Here, one should notice that the results of figure 4(A) are obtained for positive values of D . In particular, we can also find some characteristic features for the T_K -curve in the system when the figure is given with an expanded scale; the results are shown in figure 4(B). For the T_K -curve labelled $D/J = 0.5$, we can also see that two compensation points are possible. By using an expanded scale for plotting the T_K -curve labelled $D/J = 0.25$, we can see that three compensation points are possible, as depicted in figure 4(C).

In figure 4(C), the possibility of three compensation points is apparent. In order to confirm whether three compensation points can be found in the temperature dependence of the total magnetization for the system with $c = 0.9$, $D/J = 0.25$ and $p = 0.705$ (for example, in figure 4(C)), the plot is given in figure 5. The (M/N) -curve clearly exhibits three compensation points below T_C , which is consistent with the prediction of figure 4(C).

5. Conclusions

In this work, we have studied the magnetic properties of a diluted mixed spin ferrimagnetic Ising system consisting of two kinds of magnetic atom, A and B, with spins $S_A = 5/2$ and $S_B = 2$. They have been discussed within the framework of the EFT. Numerical results are obtained in section 3 for the pure case ($p = 1.0$ and $c = 1.0$) and in section 4 for the diluted case ($p < 1.0$ and $c < 1.0$). In particular, we have examined the effect of

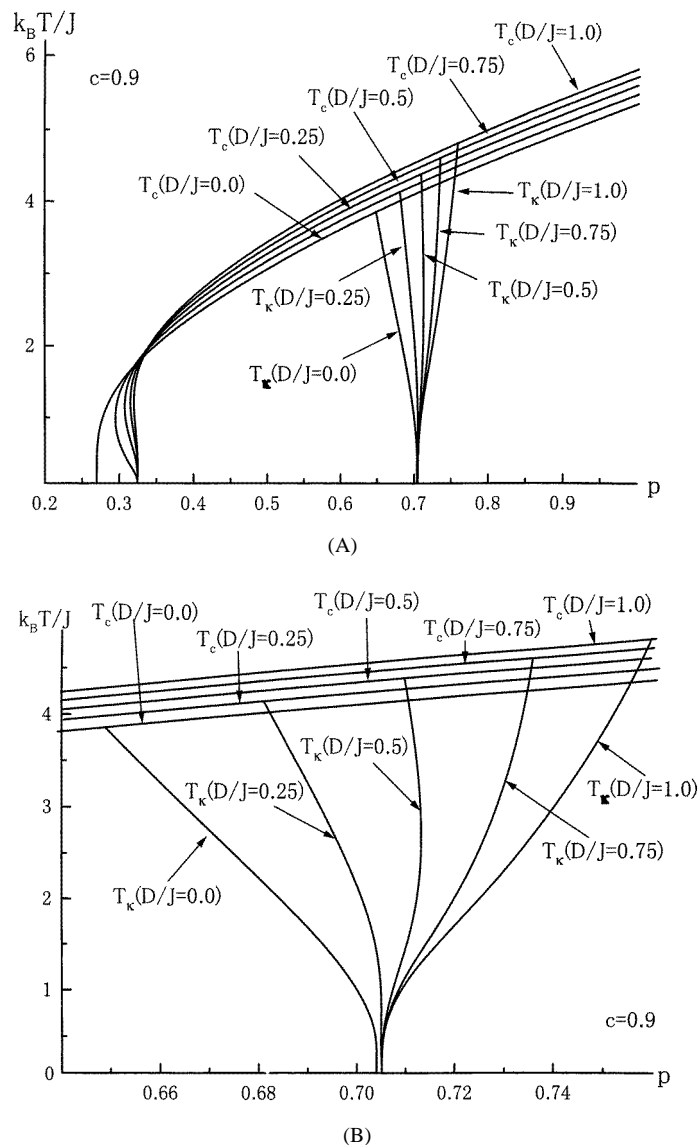


Figure 4. Phase diagrams of the diluted mixed spin ferrimagnetic honeycomb lattice with a fixed value of c of 0.9 in the T - p plane, for different values of D . (A) The T_C - and T_K -curves are plotted for five values of D/J , namely $D/J = 1.0, 0.75, 0.5, 0.25$ and 0.0 . (B) The T_C - and T_K -curves of (A) are plotted using an expanded scale, in order to display the characteristic features of T_K . (C) The T_K -curve for the system with $c = 0.9$ and $D/J = 0.25$ in the vicinity of $p = 0.705$.

a positive single-ion anisotropy D on the compensation temperature in the pure system on the basis of the EFT as well as the MFA, in order to clarify the characteristic feature of the temperature dependence of M observed for a molecular-based magnetic material, $N(n-C_4H_9)_4Fe^{II}Fe^{III}(C_2O_4)_3$ with $S_A = 5/2$ (Fe^{III}) and $S_B = 2$ (Fe^{II}). We find that there is a critical value of D above which a compensation point can be found, and hence that the behaviour of M for this compound can be explained by the Néel model, as has been

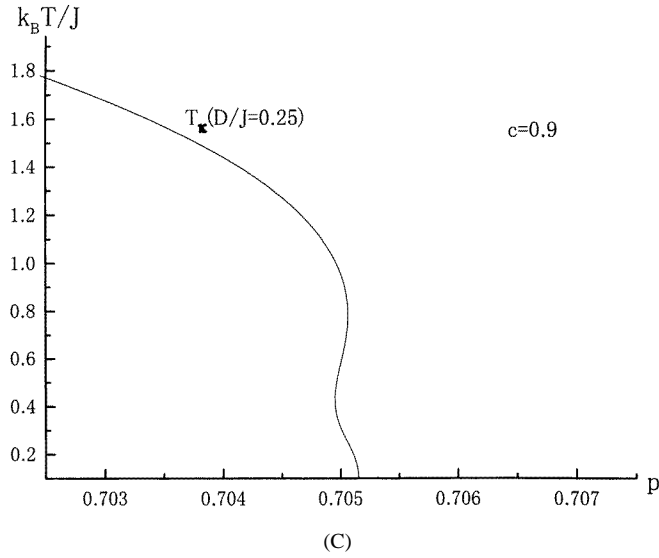


Figure 4. (Continued)

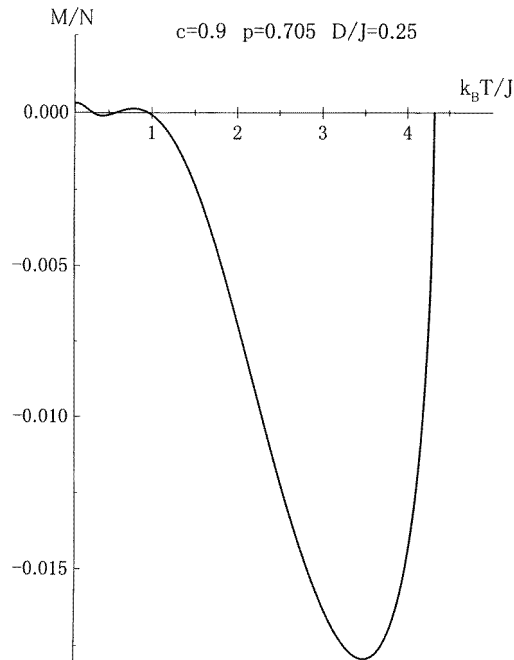


Figure 5. The temperature dependence of M in the diluted mixed ferrimagnetic system with $c = 0.9$, $D/J = 0.25$ and $p = 0.705$. The three compensation points are obtained below T_C , which is consistent with the prediction of figure 4(C).

proposed in [8].

In section 4, the effects of dilution on the phase diagrams as well as the temperature dependence of the total magnetization in the ferrimagnetic honeycomb system have been

examined numerically within the framework of the EFT. As shown in figures 3–5, we can see that several (two or three) compensation points are possible in the diluted mixed spin-5/2 and spin-2 ferrimagnetic Ising system on a honeycomb lattice, even when the value of D is positive. This is clearly at variance with the results of previous work [4–5, 10–12] where such points were only deemed possible for $D/J \leq 0.0$. Of course, for the present system, for $D/J \leq 0.0$ the possibility of many compensation points will be indicated, for example by the curve labelled $p = 0.5$ in figure 3(C).

Finally, the results of section 4 indicate that from the behaviour observed on diluting the molecular-based magnet $N(n\text{-C}_4\text{H}_9)_4\text{Fe}^{\text{II}}\text{Fe}^{\text{III}}(\text{C}_2\text{O}_4)_3$, some characteristic features of the magnetic properties may be observed. We hope that such a problem will be investigated experimentally.

Appendix

The sublattice magnetizations (5) can be derived by using the Ising spin identities and the differential operator technique [13–17]. The sublattice magnetization m_α ($\alpha = \text{A}$ or B) is given exactly by

$$m_\alpha = \frac{\langle \xi_{i\alpha} \langle S_{i\alpha}^z \rangle \rangle_r}{\langle \xi_{i\alpha} \rangle_r} = \frac{1}{\langle \xi_{i\alpha} \rangle_r} \langle \xi_{i\alpha} \langle F_\alpha(E_\alpha) \rangle \rangle_r = \frac{1}{\langle \xi_{i\alpha} \rangle_r} \langle \xi_{i\alpha} \langle \exp(E_\alpha \nabla) \rangle \rangle_r F_\alpha(x) \Big|_{x=0} \quad (\text{A1})$$

with

$$E_\alpha = -J \sum_{\delta} S_{i+\delta\alpha'}^z \xi_{i+\delta\alpha'} \quad (\text{A2})$$

where $\nabla = \partial/\partial x$ is the differential operator and the mathematical relation $\exp(a\nabla)f(x) = f(x+a)$ is used.

In order to treat (A1) further, we can use the identity introduced in [14], namely

$$\exp(aS_{i\alpha}^z) = \cosh(a\eta_\alpha) - \frac{S_{i\alpha}^z}{\eta_\alpha} \sinh(a\eta_\alpha) \quad (\text{A3})$$

where the parameter η_α is defined by

$$\eta_\alpha = \frac{1}{\langle \xi_{i\alpha} \rangle_r} \langle \xi_{i\alpha} \langle (S_{i\alpha}^z)^2 \rangle \rangle_r \quad (\text{A4})$$

and the relation $(\xi_{i\alpha})^2 = \xi_{i\alpha}$ holds. By the use of these relations, equation (A1) (as well as equation (A4)) can be written in the form

$$m_\alpha = \left\langle \left\langle \prod_{\delta} \left[1 - \xi_{i+\delta\alpha'} + \xi_{i+\delta\alpha'} \left\{ \cosh(a\eta_{\alpha'}) - \frac{S_{i+\delta\alpha'}^z}{\eta_{\alpha'}} \sinh(a\eta_{\alpha'}) \right\} \right] \right\rangle \right\rangle_r F_\alpha(x) \Big|_{x=0} \quad (\text{A5})$$

with $a = J\nabla$, where δ represents the number of nearest-neighbour atoms.

In order to derive the sublattice magnetizations (5), the decoupling approximation was introduced into (A5) for treating the multispin correlation functions:

$$\langle \langle \xi_{i\alpha} S_{i\alpha}^z \xi_{j\beta} S_{j\beta}^z \cdots \xi_{k\gamma} S_{k\gamma}^z \rangle \rangle_r \simeq \langle \langle \xi_{i\alpha} S_{i\alpha}^z \rangle \rangle_r \langle \langle \xi_{j\beta} S_{j\beta}^z \rangle \rangle_r \cdots \langle \langle \xi_{k\gamma} S_{k\gamma}^z \rangle \rangle_r \quad (\text{A6})$$

for $i \neq j \neq \cdots \neq k$. As discussed in [13–17], the statistical accuracy (of (A6)) corresponds to the Zernike approximation [18] of a spin-1/2 Ising model. As shown in figure 1, the transition temperature shows a reasonable improvement over the MFA result.

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