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The role of the exchange and dipolar interactions in the determination of the magnetic ordering of a two-dimensional lattice with random vacancies

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Abstract
The phase diagram of a system of interacting spins on a square lattice with a small percentage of random vacancies is determined from Monte Carlo simulations. The spins are confined to rotate in the plane and interact through the exchange and dipolar interactions. Despite the anisotropic nature of the dipolar interaction, for the parameter set considered in the present work, the ground states of the system are continuously degenerate in the absence of vacancies. In the case $J = 0$, it has been shown that the disorder arising from both vacancies and thermal fluctuations lifts the degeneracy of the ground state and generates long-range antiferromagnetic order. This phenomenon is referred to as 'order from disorder'. Because of the competing nature between the disorder created by vacancies and thermal fluctuations, for $J = 0$ such a system has been shown to undergo a transition from a collinear to microvortex phase with decreasing temperature. In this paper we extend the results from these earlier simulations to include a finite exchange interaction. A number of new and interesting results emerge from these simulations. For $J/g = -0.3$ and -2.5 (where J and g denote the exchange and dipolar interaction constants respectively), we obtain results that are qualitatively similar to those obtained for $J = 0$, with a transition from the collinear to the microvortex phase with decreasing temperature. However, for $J/g = -1.5$ our simulations show that the sequence is reversed and we instead observe a transition from the microvortex phase to the collinear phase with decreasing temperature. Furthermore, preliminary results for larger values of $|J|/g$ indicate that the competition between the disorder created by the vacancies and the thermal fluctuations extends to both the simple antiferromagnetic case ($J/g = -4.0$)

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and the ferromagnetic case ($J/g = 1.5$), giving rise to an analogous in-plane reorientation transition. In all the cases we consider, the temperature at which the transition occurs increases with increasing vacancy concentration.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The dipolar interaction can play an important role in determining the properties of two-dimensional magnetic systems [1]. This arises in large part because of the long-range character of the dipolar interaction and the fact that it is anisotropic. Despite the anisotropic nature of the dipolar interaction, for $J = 0$, there exist several two-dimensional lattice structures for which the ground state is continuously degenerate, in the absence of vacancies [2, 3]. In the case of highly frustrated systems such as these, the effect of the dipolar interaction can lead to a very subtle and complex phase behaviour. An important illustration of this occurs in the case of a system of spins on a square lattice. For this system it has been shown that both thermal fluctuations and randomly distributed vacancies can lift the degeneracy of the ground state, giving rise to long-range magnetic order [3–5]. The competition between the inherent frustration of the ground state and the effect of disorder has been studied in a wide range of systems [6, 7], and has been referred to in the context of antiferromagnetic systems as 'order from disorder'.

Recent studies have shown that, in the absence of vacancies, the precise nature of the magnetic ordering in this system is extremely sensitive to the nature of the disorder. For example, in the case in which the spins interact only through the dipolar interaction ($J = 0$), the thermal fluctuations give rise to an antiferromagnetic state in which the spins are aligned ferromagnetically along one of the axes of the lattice, with the ferromagnetic rows ordered antiferromagnetically along the other axis. This is referred to as the collinear phase (AF1), and is shown in figure 1(a). However, the introduction of an antiferromagnetic exchange interaction appears to change the character of the thermal fluctuations in such a way that over a certain range of exchange constant the spins align at an angle of 45° to the lattice axis [8–10]. This state is referred to as the microvortex state (AF2), and is shown in figure 1(b).

For $J = 0$ the introduction of vacancies can also be shown to induce the microvortex phase, at sufficiently low temperature. Simulations show that the competition between the vacancies and the thermal fluctuations can give rise to a reorientation transition from the microvortex state to the collinear phase with increasing temperature [5]. The transition appears to be first order, and the transition temperature depends on the concentration of vacancies. For low concentrations of vacancies the phase boundary separating the microvortex phase and the collinear phase is consistent with the phase diagram postulated by Henley and Prakash [3]. However, at higher concentrations of vacancies, our earlier results [5] indicate that there exist a number of non-equivalent nearly degenerate low-energy states, and this gives rise to metastability effects. In this paper we consider only low concentrations of vacancies where the effect of the exchange interaction on the low-temperature state can be more readily discerned.

We present the results of Monte Carlo simulations which examine the combined effect of an exchange interaction and randomly distributed vacancies on the magnetic phase behaviour of a system of spins located on a square lattice that interact through the dipolar interaction. In particular, we show that in the region $-3.2 < J/g < 0$ the sequence of transitions as a function of temperature depends on the precise value of the exchange interaction in a somewhat unusual manner. In addition, preliminary results indicate that the reorientation transition from the microvortex to the collinear phase has a close analogue in both the 'pure' antiferromagnetic,

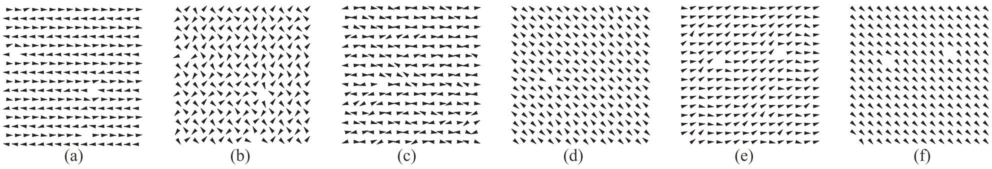

Figure 1. Sections of spin configurations from simulations on a 32×32 lattice. The phases from left to right are denoted as AF1 (collinear), AF2 (microvortex), AA1, AA2, F1 and F2.

in which each spin is aligned antiferromagnetically with each of its nearest neighbours (AA1 and AA2 in figure 1), and in the ferromagnetic phase (F1 and F2 in figure 1).

We consider the case of a system of planar spins with Hamiltonian given by

$$H = -J \sum_{\langle i,j \rangle} \vec{\sigma}_i \cdot \vec{\sigma}_j + g \sum_{i \neq j} \left(\frac{\vec{\sigma}_i \cdot \vec{\sigma}_j}{r_{ij}^3} - 3 \frac{(\vec{\sigma}_i \cdot \vec{r}_{ij})(\vec{\sigma}_j \cdot \vec{r}_{ij})}{r_{ij}^5} \right). \quad (1)$$

The spins $\vec{\sigma}_i$ are treated as classical vectors of fixed length located at the sites of the lattice $\{i\}$, positioned at $\{\vec{r}_i\}$ and confined to the plane of the lattice.

Results were calculated for lattices sizes $N = 32 \times 32$ and 64×64. Periodic boundary conditions were imposed on the spin configurations by constructing an infinite plane from replicas of the finite system, including the vacancies and the dipolar interaction calculated using the Ewald summation method [1]. The sizes of the lattices are sufficiently large that the vacancies can be considered to be randomly distributed. The simulations were carried out using the standard Metropolis algorithm. For the results reported in the current work an equilibration time of 10^4 Monte Carlo steps per site (MCS/site) were used for each simulation. The number of samples used to calculate the averages for the larger lattices was 5×10^4 MCS/site, using samples taken every 10 MCS/site. For the smaller lattice size the respective numbers were 10×10^4 MCS/site and 10 MCS/site.

2. Collinear and microvortex phases

For $J = 0$, in the absence of vacancies, the ground-state spin configurations form a continuously degenerate manifold of antiferromagnetic spin configurations characterized by a microvortex angle $\phi_{\rm mv}$ [4]. Prakash and Henley [3] have shown that, in a model with a dipolar-like interaction that includes only the nearest-neighbour terms, the thermal fluctuations remove this degeneracy, and hence the free energy is dependent on the microvortex angle at finite temperature. The dependence of the free energy on the microvortex angle reflects the fourfold symmetry of the underlying lattice. Monte Carlo simulations also show that for $J = 0$, the system manifests long-range antiferromagnetic order at low temperature, with a microvortex angle $\phi_{\rm mv} = n\pi/2$ [4], corresponding to the collinear phase (AF1) shown in figure 1(a). While the introduction of an antiferromagnetic exchange interaction does not remove the degeneracy of the dipolar ground state, it nevertheless does change the character of the thermal fluctuations. As discussed in the introduction, this subtle change in the character of the thermal fluctuations can change the equilibrium magnetic phase to one with $\phi_{\rm mv} = \pi(2n + 1)/4$, referred to as the microvortex phase (AF2), shown in figure 1(b). The phase diagram for such a system is reported in [9, 10].

Patchedjiev *et al* have also investigated the effect of low concentrations of vacancies in the magnetic lattice on the phase behaviour for a system of spins that interact only through

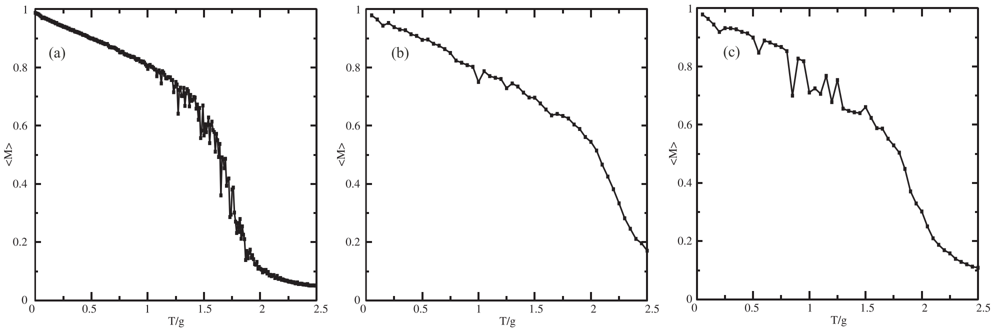

Figure 2. The average sublattice magnetization for (a) $J/g = -0.3$, (b) $J/g = -1.5$ and (c) $J/g = -2.5$.

the dipolar interaction ($J = 0$) [5]. The results obtained show that the competition between the microvortex phase, favoured by the vacancies, and the collinear phase, favoured by the thermal fluctuations, gives rise to a reorientation transition from the microvortex phase at low temperature to the collinear phase at high temperature, with a transition temperature that increases with increased vacancy concentration. The data are consistent with the phase diagram conjectured by Prakash and Henley [3].

In these earlier studies the collinear phase and the microvortex phase were distinguished through the conjugate field P [4],

$$P(T) = \frac{1}{N}\left\langle \sum_{\vec{R}} \left(\sigma_x^4 + \sigma_y^4\right) \right\rangle. \tag{2}$$

A random orientation of the spins yields a value $P = 0.75$, while orientations along the x or y directions, as in the collinear phase, give $P = 1.0$. When the spins are oriented at 45° to the x and y axes, as in the microvortex phase, $P = 0.5$.

We first look at the phase behaviour for $J/g = -0.3$. In the absence of vacancies, the stable phase at low temperature corresponds to the collinear phase [9, 10]. Figure 2(a) plots the order parameter, defined in [9] as a function of temperature for a concentration $c = 1\%$. The data show the appearance of long-range magnetic order at low temperature. To distinguish between the collinear and microvortex phases we plot the conjugate field P as a function of decreasing temperature in figure 3, for $N = 64 \times 64$ and vacancy concentrations $c = 1\%$ and 2%. The results here are similar to those obtained for $N = 32 \times 32$, and the subsequent plots are shown only for the smaller lattice size. At high temperature the spins in the system are randomly oriented in the plane, which yields a value of $P = 0.75$. As the temperature is lowered below the Néel temperature, $T/g \sim 1.5$, the value of P begins to increase, indicating that the spins are starting to align along one of the lattice axes to form the collinear phase. The formation of the collinear phase is also shown in figure 3(b), which plots the average orientation of the spins in the four-site magnetic unit cell as a function of temperature.

As the temperature is lowered even more, figure 3(a) shows that the conjugate field P begin to decrease at around $T/g \approx 0.1$ and 0.2 for $c = 1\%$ and 2% respectively. This is the signature of the in-plane reorientation transition at which the system switches from the collinear phase to the microvortex phase. This is also indicated by the change in the orientation of the average spin angles in the magnetic unit cell to $\pi/4$, $3\pi/4$, $5\pi/4$ and $7\pi/4$ shown in figure 3(b). We denote by T_{R} the temperature at which the reorientation transition from the collinear phase to the microvortex phase takes place. The increase in T_{R} with increasing

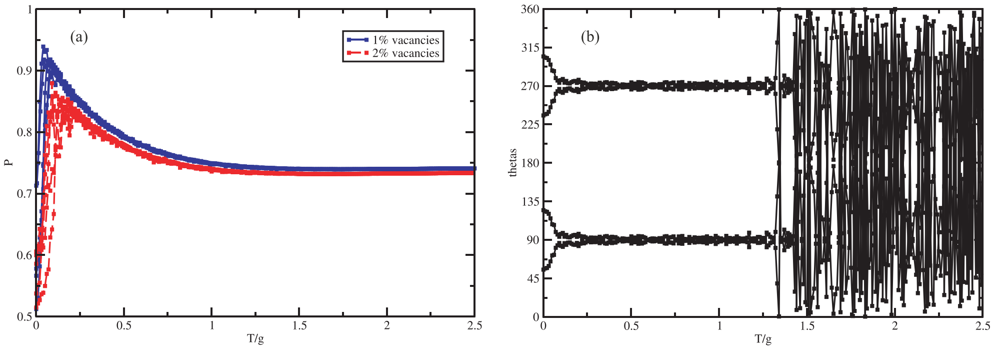

Figure 3. (a) The conjugate field P plotted as a function of temperature for $J/g = -0.3$, for five separate Monte Carlo runs for a vacancy concentrations of $c = 1\%$ (solid line) and $c = 2\%$ (dashed lines). (b) The average angle for the four sublattice magnetizations plotted as a function of temperature for $c = 1\%$. The size of the lattice is 64×64.

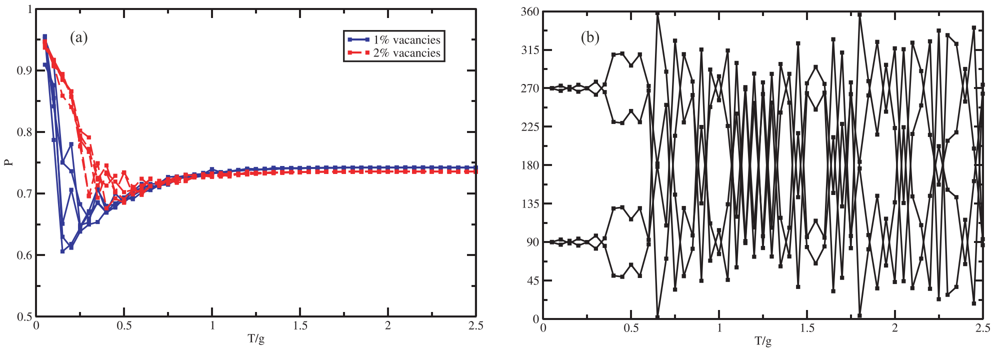

Figure 4. (a) The conjugate field P plotted as a function of temperature for $J/g = -1.5$, for five separate Monte Carlo runs for a vacancy concentrations of $c = 1\%$ (solid line) and $c = 2\%$, (dashed lines). (b) The average angle for the four sublattice magnetizations plotted as a function of temperature for $c = 1\%$. The size of the lattice is 32×32.

concentration is qualitatively similar to that reported for $J = 0$ [5], although the temperatures are slightly higher.

The corresponding data for $J/g = -1.5$ are presented in figures 2(b) and 4. For $J/g = -1.5$, the equilibrium phase at low temperature in the absence of vacancies is the microvortex phase [9, 10]. The plot of the dipolar order parameter shown in figure 2(b) shows the appearance of long-range magnetic order as the temperature is lowered below the Néel temperature. In figure 4 we plot the conjugate field P and the angles for the sublattice magnetizations for $N = 32 \times 32$, vacancy concentrations of $c = 1\%$ and 2%, and $J/g = -1.5$. As the temperature is reduced the conjugate field P decreases, indicating the formation of the microvortex phase. Evidence for the appearance of the microvortex state is also seen in figure 4(b), which plots the average orientation of the spins in the four-site magnetic unit cell as a function of temperature.

As the temperature is lowered further, we see, at $T/g \approx 0.1$ for $c = 1\%$ and $T/g \approx 0.2$ for $c = 2\%$, the conjugate order parameter begin to rise and the average orientation of the spins on the four-site magnetic unit cell switches from $\pi/4$, $3\pi/4$, $5\pi/4$ and $7\pi/4$ to $\pi/2$ and $3\pi/2$.

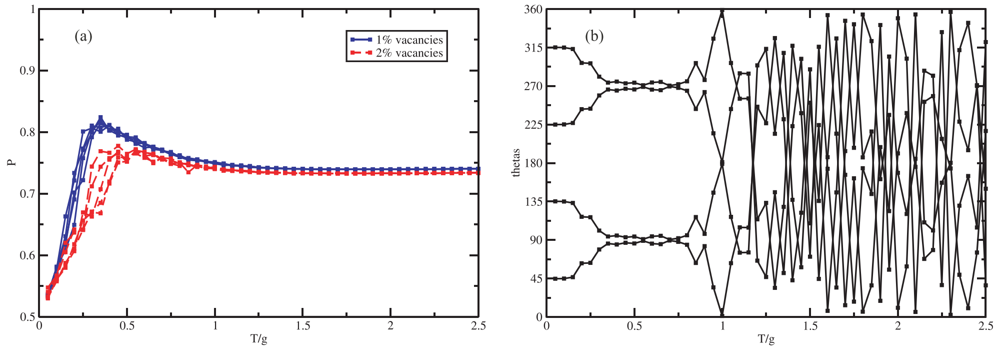

Figure 5. (a) The conjugate field P plotted as a function of temperature for $J/g = -2.5$, for five separate Monte Carlo runs for a vacancy concentrations of $c = 1\%$ (solid line) and $c = 2\%$ (dashed lines). (b) The average angle for the four sublattice magnetizations plotted as a function of temperature for $c = 1\%$. The size of the lattice is 32×32.

This is the signature of a reorientation transition from the microvortex phase to the collinear phase.

This result implies that for $J/g = -1.5$ the effect of the exchange interaction is to reverse the role of the vacancies and the thermal fluctuations in determining the orientation of the equilibrium antiferromagnetic order observed for the case $J/g = -0.3$. This rather unexpected result illustrates the very subtle role that the exchange interaction plays in modifying the spin wave excitation spectra and the sensitivity of the order parameter to the nature of the thermal fluctuations.

The conjugate field P and the angle of the sublattice magnetization are plotted as a function of temperature for $J/g = -2.5$ in figures 5(a) and (b) respectively for a lattice size of $N = 32 \times 32$ for $c = 1\%$ and 2%. In the absence of vacancies the collinear phase is the equilibrium phase at low temperature [9, 10]. The data shown in figures 5(a) and (b) show the formation of the collinear phase as the temperature is lowered below the Néel temperature. However, for $J/g = -2.5$, the effect of the vacancies is to induce a reorientation transition from the collinear phase to the microvortex phase as the temperature is lowered. The reorientation transition temperatures are $T/g \approx 0.2$ and 0.25 for $c = 1\%$ and $c = 2\%$, respectively.

The results from a series of Monte Carlo simulations similar to these discussed above, for J values in the range $-3.2 < J/g < 0$, yield the phase diagram for $c = 1\%$ and 2%, shown in figure 6. The exact location of the reorientation transition temperature is difficult to determine due to domain formation and the dependence of the transition on specific distribution of the vacancies on the lattice. The important feature to note, however, is the fact that at low temperature the sequence of equilibrium phases with increasing exchange interaction is given by microvortex → collinear → microvortex, while at higher temperature the sequence is given by collinear → microvortex → collinear. A tentative phase diagram is also presented in figure 7, comparing the results obtained for $c = 1\%$ with those obtained in [9] for $c = 0$.

This change in the sequence of stable phases with temperature, for $-3.2 < J/g < 0$, may be described, at least qualitatively, by a difference in free energy between the microvortex phase and the collinear phase given by

$$\begin{aligned}\Delta F(J, T) &= F(\text{collinear}) - F(\text{microvortex}) \\ &= -\alpha(T - mc)(J - J_1)(J - J_2). \qquad (3)\end{aligned}$$

The surface representing the free-energy difference between the microvortex phase and the

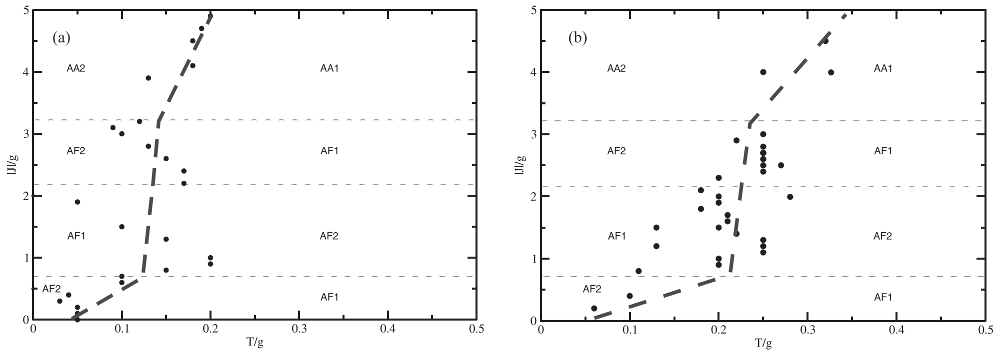

Figure 6. A plot showing the reorientation transition temperature T_R for several values of J/g for (a) $c = 1\%$ and (b) $c = 2\%$. The lines denoting the (tentative) phase boundaries are also shown.

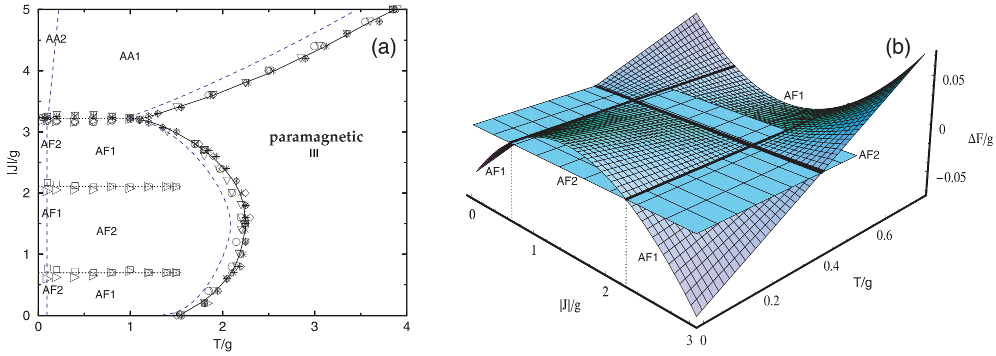

Figure 7. (a) The phase diagram for $c = 1\%$ and $c = 0$. (b) The free-energy surface described by equation (3).

collinear phase, as determined by equation (3), is shown schematically in figure 7(b) as a function of the exchange interaction and temperature. The parameters used are $J_1/g = 0.5$ and $J_2/g = 2.2$, which define the boundaries between the collinear and microvortex phases, $m = 0.5$, which defines the slope of the coexistence line for $J = 0$, and $\alpha = 0.1$. The derivation of the phenomenological free energy given by equation (2), either by spin wave theory or using Monte Carlo techniques, would shed important light on the complex interplay between the exchange interaction and the disorder induced by the vacancies and the thermal fluctuations.

3. The antiferromagnetic and ferromagnetic phases

In the previous section we considered the region $-3.2 < J/g < 0$ in which the equilibrium spin configurations consist of the collinear (AF1) phase and the microvortex (AF2) phase. For $J/g < -3.2$, the ground state is given by an 'simple' antiferromagnetic spin configuration in which each spin is aligned antiferromagnetically with its four nearest neighbours. This state is also continuously degenerate, despite the anisotropic nature of the dipolar interaction. In the absence of vacancies at low temperature the spins align along one of the lattice axes, as shown in figure 1(c). We refer to this as the AA1 phase. Figure 8(a) shows the behaviour of the conjugate field P for $J/g = -4$ and for $c = 1\%$ and 2%. The data show that, as the

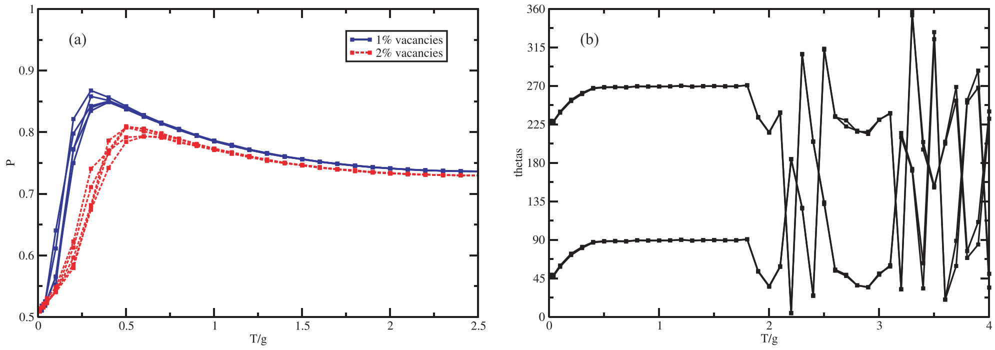

Figure 8. (a) The conjugate field P plotted as a function of temperature for $J/g = -4.0$, for five separate Monte Carlo runs for vacancy concentrations of $c = 1\%$ (solid line) and $c = 2\%$, (dashed lines). (b) The average angle for the sublattice magnetizations are plotted as a function of temperature for $c = 1\%$. The size of the lattice is 32×32.

temperature is lowered below the Néel temperature, the value of P begins to increase as the spins align along the y-axis ($\phi = \pi/2$). This is reflected in figure 8(b), in which the angle of the sublattice magnetizations is plotted as a function of temperature and which shows the spins on one sublattice aligned along the positive y-axis ($\phi = \pi/2$) and those on the other sublattice aligned in the opposite direction ($\phi = 3\pi/2$). As the temperature is reduced further, P begins to decrease, indicating that the spins reorient to $\phi = \pi/4$. Again this is reflected in the plot of the angle of the sublattice magnetizations as a function of temperature, which shows the spins on one sublattice aligned at 45° to the positive x-axis ($\phi = \pi/4$) and those on the other sublattice aligned in the opposite direction ($\phi = 5\pi/4$), as shown in figure 1(c). We refer to this as the AA2 phase. This signals the appearance of an in-plane reorientation transition in which the magnetization axis rotates 45° as a consequence of the competition between the thermal fluctuations and the structural disorder from the random vacancies, in close analogy with the previous example. We note that the reorientation temperature increases with increasing vacancy concentration.

Data from similar Monte Carlo simulations, with other values of $J/g < -3.2$, show a similar behaviour. However, while the simulations indicate that the reorientation transition temperature T_R increases with increasing $|J|$, as in the case of the collinear/microvortex phase transition the exact determination of the coexistence line is complicated by the effects of hysteresis and domain formation. The data for $c = 1\%$ and 2% are shown in the phase diagrams shown in figures 6(a) and (b) respectively.

The above result suggests that the reorientation transition, in which the magnetization axis switches as the temperature is lowered, is not limited to the collinear/microvortex phase. We may wonder, therefore, as to the effect of the impurities on the planar ferromagnetic phase, as the energy of the ferromagnetic ground state is also independent of the orientation of the magnetization.

In figure 9(a) we plot the conjugate field P as a function of temperature, for $J/g = 1.5$ for $c = 1\%$ and 2%. As in the case of the antiferromagnetic exchange discussed in the previous section, the data were obtained from a series of Monte Carlo simulations. The data show the conjugate field P increasing with decreasing temperature as the spins order along the y-axis, as shown in figure 1(e). We refer to this as the F1 phase. However, at $T/g \approx 0.25$ and $T \approx 0.5$ the conjugate field drops rapidly for $c = 1\%$ and 2% respectively. This is signature of a reorientation transition in which the magnetization axis undergoes a rotation from $\phi = 90°$ to

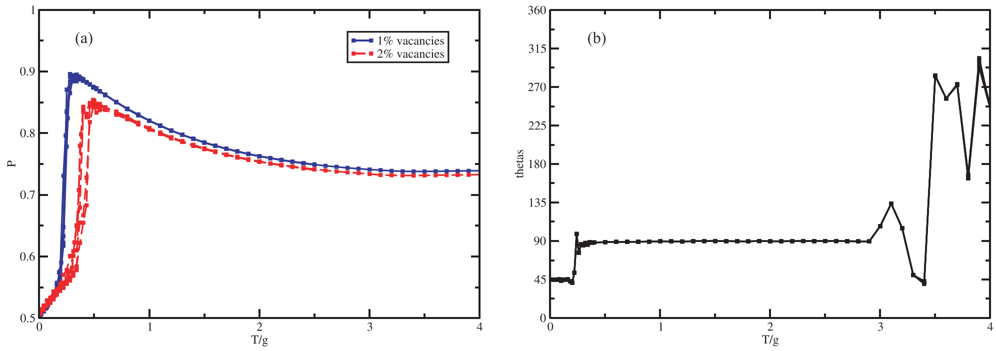

Figure 9. (a) The conjugate field P plotted as a function of temperature for $J/g = 1.5$, for five separate Monte Carlo runs for vacancy concentrations of $c = 1\%$ (solid line) and $c = 2\%$ (dashed lines). (b) The average angle for magnetization is plotted as a function of temperature for $c = 1\%$. The size of the lattice is 32×32.

45°. Below the reorientation temperature the conjugate field continues to decrease as the spins order at 45° to the x-axis, reaching a value $P \approx 0.5$ at $T = 0$, as shown in figure 1(f). We refer to this as the F2 phase. This is consistent with the plot of the magnetization angle as a function of temperature presented in figure 9(b). The results from these simulations appear to confirm that the competition between thermal fluctuations and structural disorder of the vacancies can also give rise to an in-plane reorientation in the ferromagnetic case.

4. Summary

Results are presented from a series of Monte Carlo simulations on a system of planar spins arranged on a square lattice with randomly distributed vacancies. The interaction between the spins consists of the exchange and dipolar interactions. We consider both ferromagnetic and antiferromagnetic exchange. In all the cases we have considered, the ground state is continuously degenerate in the absence of vacancies. This degeneracy is lifted by the structural disorder of the vacancies and the thermal fluctuations, resulting in long-range magnetic order. The magnetization axis is determined by the competition between the thermal and structural disorder, in a manner that is entirely analogous to that observed for $J = 0$ [5], and gives rise to an in-plane reorientation transition as the magnetization axis switches with increasing or decreasing temperature. The temperature at which the reorientation transition occurs, T_R, increases with increasing vacancy concentration, reflecting the nature of the competition between the structural and thermal disorder. In the regime $-3.2 < J/g < 0$ the ground state and the sequence of phases at the reorientation transition depends on the magnitude of the exchange constant J. In particular, we find that the equilibrium spin configuration corresponds to the microvortex phase at low temperature, for both $J/g = -0.3$ and -2.5, and to the collinear phase at higher temperatures. However, for $J/g = -1.5$ this sequence is reversed, and we find that at low temperature the equilibrium spin configuration corresponds to the collinear phase, and at higher temperatures to the microvortex phase. Thus, for $J/g = -1.5$, the effect of the exchange interaction on the spin wave excitations is to reverse the role of the thermal and structural disorder in determining the alignment of the magnetization axis.

For larger values of $|J|/g$, our simulations also show an in-plane rotation of the magnetization axis as a function of temperature, for both the simple antiferromagnetic and ferromagnetic phases. This is analogous to the microvortex/collinear phase transition and arises from the competition between the disorder created by the vacancies and that created by the thermal fluctuations. However, we should emphasize that we have completed simulations only for a limited number of data sets and hence both the results obtained and the conclusions we draw from them, while interesting, should be seen as preliminary.

Acknowledgments

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