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Phase diagrams of two inhomogeneous Ising models on the triangular lattice

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Abstract. Phase diagrams of two inhomogeneous annealed Ising models on the triangular lattice are constructed in an exact way. The interimpurity exchange constants (bonds), of the same but arbitrary value and sign, can be located along the horizontal edges of the triangles. In the first model the remaining non-random couplings are ferromagnetic and in the second they are antiferromagnetic. The dependence of the correlation functions and magnetisation on the temperature is calculated and discussed. It is shown that, for certain values of the impurity bond couplings and a range of their concentrations, there is no net magnetic moment at $T = 0$ and low temperatures. The magnetisation appears at some finite temperature and then vanishes at a higher one. This increase of the magnetisation with the temperature is connected with the role of entropy in determining the minimum of the free energy.

1. Introduction

Ising models on the triangular lattice have been investigated by many authors. References to earlier works can be found in a series of papers by Stephenson (1964, 1970, a, b), where the most complete discussion of the correlation functions for the case of isotropic and anisotropic interactions is also given. The main difference between phase diagrams on the triangular lattice and those on other two-dimensional lattices is that, when all the interactions are equal and antiferromagnetic, each triangle is frustrated and as a result the net magnetic moment is zero, even at $T = 0$. If one of the interactions is weaker, the spins along that direction order ferromagnetically, despite antiferromagnetic coupling between them, frustration is lifted and non-zero magnetisation appears. In the exactly solved Ising model on the square lattice with additional interactions along one of the diagonals to next-nearest neighbours (NNN) it was found (Vaks *et al* 1965) that for a certain ratio of the NN to NNN interactions, there are two transition temperatures and the ground state is magnetically not ordered. The same effect has also been found on the square lattice. This unusual behaviour—increasing of the magnetisation with the temperature—is called re-entrance.

Syozi (1972) used the decorated bonds technique for a model with unequal ferro- and antiferromagnetic couplings. Re-entrance appeared in the (T_c, x) phase diagram, where x was the concentration of one kind of bond at the side of the weaker exchange coupling. Thorpe and Beeman (1976) found the re-entrance in an annealed Ising model for rectangular or Gaussian distribution of the coupling constants, provided the width of the distribution was large enough to contain positive and negative couplings. Kitatani *et al* (1985), where an extensive list of references can be found,

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considered a system with three kinds of interactions—two ferro- and one antiferromagnetic. The phase diagram was constructed through investigations of spin correlations in a finite lattice.

Explanation of this phenomenon has been given by Oguchi and Ueno (1975), Derrida *et al* (1978) and Ueno (1986) as the effect of minimising the free energy by increasing the entropy in a frustrated system.

In this paper we shall present two Ising models on the triangular lattice. In both cases the disorder will be restricted to one direction only. Although the annealed approach will be used, for certain values of the parameters frustration will be present in both models. On the other hand, the models will show the re-entrance phenomenon even when the frustration is absent. Exact equations to determine temperature dependence of the magnetisation, three types of correlation functions and construction of the phase diagrams will be derived.

2. The models

Two models, A and B, are considered. In model A two interactions within each triangle are ferromagnetic, while the third one (horizontal) can be exchanged for an impurity bond. Its coupling constant, the same for all impurities, can have arbitrary value and sign. In model B, the two constant interactions are equal and antiferromagnetic, while the third one (also horizontal) has random sign and strength as in model A. Both cases will be solved exactly in the annealed approach. Let us begin with model A. Denoting the spin at site 1 by s_1 ($S=\frac{1}{2}$) and the random coupling constant by $J_{i,l}$, the Hamiltonian is

$$\beta\mathcal{H}_{\mathrm{A}} = -\sum_{\{I\}} j_{i,l}s_i s_l - j\sum_{\{II\}} s_i s_l \tag{1}$$

where $\{I\}$ means summation over random (horizontal) bonds and $\{II\}$ over pairs of non-random ones (see figure 1). The lower case j denotes the temperature normalised coupling constant $j = J/kT$. Its inverse, $kT/J = t$, is our reduced temperature. Introducing $q_{i,l}$ equal to 1 if the bond between the sites i and l is J_2 and 0 if it is equal to J, we have

$$J_{i,l} = q_{i,l}J_2 + (1-q_{i,l})J. \tag{2}$$

Since we are using the annealed approach, the grand canonical partition function is

$$Z_{\mathrm{G}}^{\mathrm{A}} = \sum_s \exp\left(j\sum_{\{II\}} s_i s_l\right)\sum_{\{p\}}\exp\left(j_2\sum_{\{I\}} q_{i,l}s_i s_l + j\sum_{\{I\}}(1-q_{i,l})s_i s_l + \mu\sum_{\{I\}} q_{i,l}\right). \tag{3}$$

Here μ is the chemical potential of the impurity bonds. We may map our disordered system into a regular anisotropic Ising model on the triangular lattice by demanding that, for a single bond,

$$\sum_p \exp[j_2 q s_0 s_1 + j(1-q)s_0 s_1 + \mu q] = \exp(k s_0 s_1 + C) \tag{4}$$

where k and C are to be determined. In the regular model the horizontal coupling is k and the other two are, as in the original disordered model, j. Hence

$$Z_{\mathrm{G}}^{\mathrm{A}} = \exp(CN)Z(k) \tag{5}$$

where N is the number of horizontal bonds and $Z(k)$ is the partition function of the regular anisotropic model. From (4) we get two equations to determine k and C:

$$\exp(2k) = [\exp(j) + \exp(j_2 + \mu)][\exp(-j) + \exp(-j_2 + \mu)]^{-1} \tag{6}$$

$$\exp(2C) = [\exp(j) + \exp(j_2 + \mu)][\exp(-j) + \exp(-j_2 + \mu)]. \tag{7}$$

The concentration, p_A, of the impurity bonds can be found by differentiating the free energy derived from (5) with respect to the chemical potential:

$$p_A = \frac{\partial C}{\partial \mu} + \varepsilon_1 \frac{\partial k}{\partial \mu} \tag{8}$$

where $\varepsilon_1 = \langle s_0 s_1 \rangle$ is the NN correlation function along the horizontal line for the regular model. The chemical potential, μ, was chosen to be temperature dependent, so as to render p_A independent of the temperature. Eliminating μ with the help of (6) and (7) we arrive at

$$p_A = \frac{\sinh(k-j)}{\sinh(j_2-j)} [\cosh(j_2-k) + \varepsilon_1 \sinh(j_2-k)]. \tag{9}$$

To get p_A as a function of the two original couplings, J and J_2, we have to express K by them. This can be done (Syozi 1972), but only on the critical line, where

$$\exp(-2k) = \sinh(2j). \tag{10}$$

Putting it into (9) we get finally

$$p_A = \frac{\exp(4j) - 3}{4[\exp(2j) - \exp(-2j_2)]} \left(\exp(2j_2)(1 + \varepsilon_1) + \frac{1 - \varepsilon_1}{\sinh(2J)} \right). \tag{11}$$

To calculate ε_1 we use the expression derived by Stephenson (1964), which in our case is

$$\varepsilon_1 = \frac{1}{\pi} \int_0^{\pi} \mathscr{D}\mathscr{A} \, d\omega \tag{12}$$

where

$$\mathscr{D} = (\mathscr{A} + \mathscr{B})^{-1/2} \tag{13}$$

with

$$\begin{aligned} \mathscr{A} &= 2v_k(1+v^2)^2 + 4v^2(1+v_k^2) - (1-v^2)^2(1+2v_k)\cos\omega \\ \mathscr{B} &= (1-v^2)^2(1-2v_k)\sin\omega \end{aligned} \tag{14}$$

and

$$v = \tanh(2j) \qquad v_k = \tanh(2k). \tag{15}$$

Having the values of v and v_k, we may once again take a formula from Stephenson (1964), this time for magnetisation:

$$m = (1 - \varkappa^2)^{1/4} \tag{16}$$

where

$$\varkappa^2 = \frac{(1-v^2)^4(1-v_k^2)^2}{16(1+v^2v_k)(1+v_k)^2v^2(v_k+v^2)}. \tag{17}$$

It is of interest to also calculate other correlation functions: $\varepsilon_2 = \langle s_0 s_2 \rangle$, $\varepsilon_3 = \langle s_0 s_3 \rangle$ for the second the third neighbours on the horizontal line, as well as those along one

of the remaining edges ('diagonal'): $\varepsilon d_1 = \langle s_0 s_4 \rangle$, $\varepsilon d_2 = \langle s_0 s_5 \rangle$ and $\varepsilon d_3 = \langle s_0 s_6 \rangle$ (see figure 1). Following the calculations presented by Stephenson (1964) we get

$$\langle s_0 s_2 \rangle = \varepsilon_1^2 - a_1 a_{-1} \tag{18}$$

$$\langle s_0 s_3 \rangle = \varepsilon_1^3 + a_2^2 a_{-1}^2 + a_1^2 a_{-2} - \varepsilon_1 (a_2 a_{-2} + 2 a_1 a_{-1}) \tag{19}$$

where

$$a_{\pm 1} = \frac{1}{\pi} \int_0^{\pi} (\mathscr{A} \cos \omega \mp \mathscr{B} \sin \omega)\, \mathrm{d}\omega \tag{20a}$$

$$a_{\pm 2} = \frac{1}{\pi} \int_0^{\pi} (\mathscr{A} \cos 2\omega \mp \mathscr{B} \sin 2\omega)\, \mathrm{d}\omega \tag{20b}$$

with $\mathscr{A}$ and $\mathscr{B}$ given by (14). To obtain εd_1, εd_2, εd_3, the formulae (20) can be used with

$$\mathscr{A} = 2v(1+v^2)(1+v_k)^2 - (1-v^4)(1+v_k^2) \cos \omega$$
$$\mathscr{B} = (1-v^2)^2 (1-v_k^2) \sin \omega. \tag{21}$$

To get the temperature dependence of the magnetisation and correlation functions we find k from (9) for given values of j and j_2, then calculate v and v_k and insert it into (17). The results are shown in figures 2 and 3, respectively.

In model B, the two constant interactions J are antiferromagnetic and the third, horizontal, J_{ij} is random. Hence the Hamiltonian is ($J < 0$)

$$\beta \mathscr{H}_{\mathrm{B}} = -\sum_{\{I\}} j_{il} s_i s_l - j \sum_{\{II\}} s_i s_l \tag{22}$$

which leads to the grand partition function, as given by (3). Mapping on a regular antiferromagnetic triangular lattice, we obtain expressions analogous to (6) and (7). Differentiating the logarithm of the partition function with respect to the chemical potential, we get the concentration, p_{B}, of the impurity bonds

$$p_{\mathrm{B}} = \frac{\sinh(k+j)}{\sinh(j_2+j)} [\cosh(j_2-k) + \varepsilon_1 \sinh(j_2-k)]. \tag{23}$$

Using the expression for the critical line

$$\exp(2k) = -\sinh(2j) \tag{24}$$

we express p_{B} by the original couplings j and j_2:

$$p_{\mathrm{B}} = \frac{1+\exp(4j)}{4[\exp(2j_2-2j)-1]} \left(\exp(2j_2-2j)(1+\varepsilon_1) + \frac{1-\varepsilon_1}{1-\exp(4j)} \right). \tag{25}$$

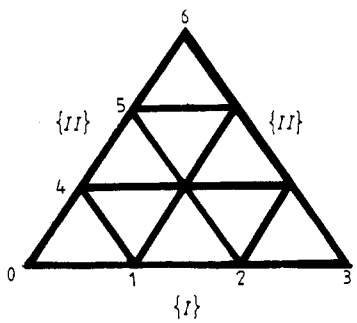

Figure 1. Basic triangle. Impurity bonds may be located only on the horizontal edges of the triangles.

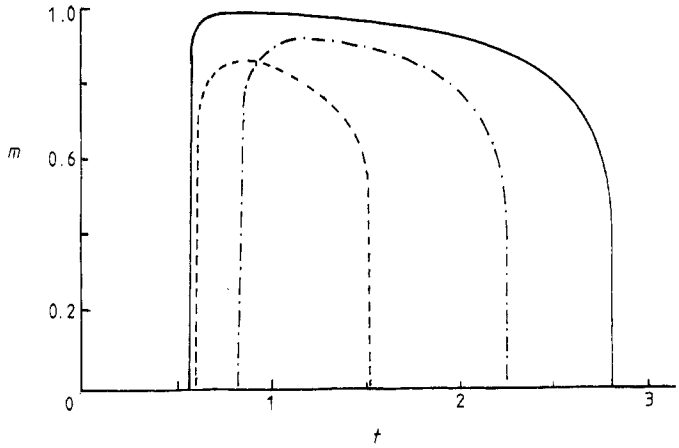

Figure 2. Magnetisation plotted against reduced temperature, $t = kT/J$. ——: model A, $p = 0.2$, $J_2 = -1.3$; — · —: model A, $p = 0.3$, $J_2 = -1.3$; - - -: model B, $p = 0.48$, $J_2 = 1$.

Here ε_1 is the NN correlation function along the horizontal line. To calculate the magnetisation and the correlation functions, (16) and (18)-(21), together with (23), should be used. The resulting curves are shown in figures 2 and 4.

3. Results

3.1. Model A

Using (11) we construct phase diagrams (t_c, p_A) for different values of the impurity couplings J_2 (figure 5). For $0 < J_2 < J = 1$, t_c decreases with p. For $J_2 = 0$, which corresponds to dilution, we arrive, at $p = 1$, to the Ising model on the square lattice and the Onsager value for t_c is recovered. For $-1 < J_2 < 0$ the ferromagnetic bonds are stronger and hence the antiferromagntic bonds are broken, i.e. the antiferromagnetically coupled spins order ferromagnetically. The cost in the energy paid for breaking the bonds shows in diminishing t_c, which however remains finite. For $J_2 = -1$ the strength of the host and impurity couplings is the same and frustration appears in the system. The number of frustrated plaquettes depends on the arrangement of the impurities. For $p > \frac{1}{2}$ the net magnetic moment is absent, even at $t = 0$. A re-entrant behaviour is found for a small range of concentrations exceeding $p = \frac{1}{2}$. For $J_2 < -1$ and a sufficiently small concentration of the impurities, there are two transition temperatures, t_{c1} and t_{c2}, where $t_{c2} > t_{c1}$. As shown in figure 2, the system is ferromagnetic between t_{c1} and t_{c2} and corresponds to a ferromagnetic region bounded by a paramagnetic region below and above this region. Because the interactions are not equal, there is no frustration, although some bonds have to be broken to realise a state with minimum energy.

To explain this unusual behaviour for $J_2 < -1$, let us start with the ground state. If an antiferromagnetic impurity bond is isolated, then it is energetically more favourable to break this single bond rather than the five neighbouring ferromagnetic ones (see figure 6(a)). The energy loss, per impurity bond, is

$$E_1 = |J_2|.$$

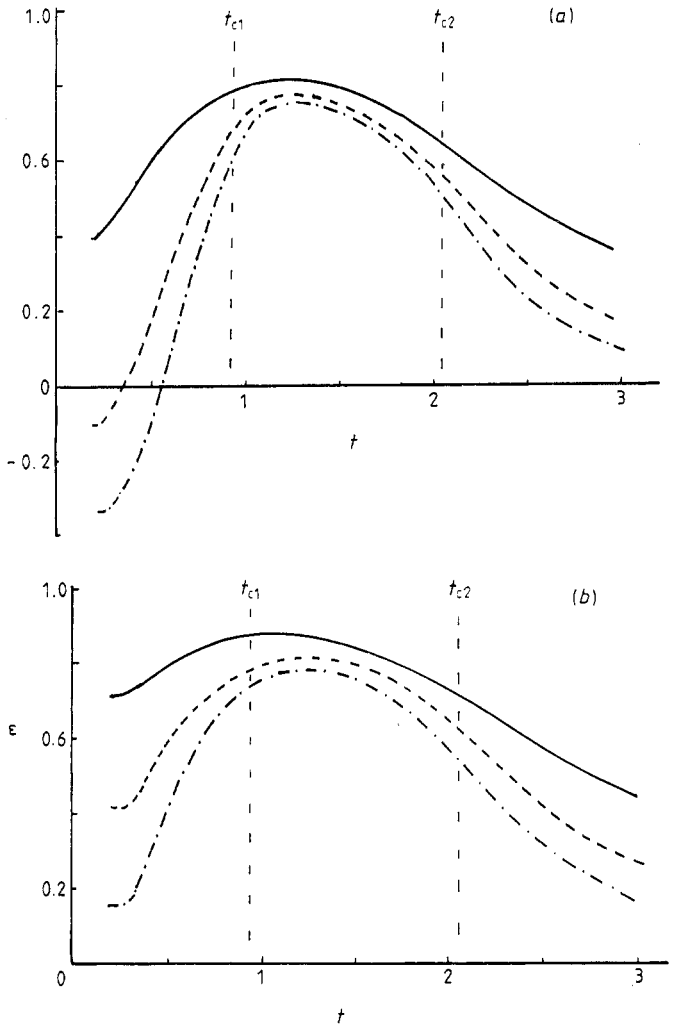

Figure 3. Model A. Correlation functions plotted against reduced temperature. $p = 0.33$, $J_2 = -1.3$. (*a*) Horizontal correlations. ——: $-\varepsilon_1$; - - -: $-\varepsilon_2$, — · —: $-\varepsilon_3$. (*b*) Diagonal correlations. ——: $+\varepsilon d_1$; - - -: $-\varepsilon d_2$; — · —: $-\varepsilon d_3$.

If, however, the impurities are arranged in stripes, as shown in figure 6(*b*), the ferromagnetic bonds get broken and that costs

$$E_2 = J.$$

The system splits into domains of opposite magnetisation, separated by 'domain walls' along the impurities and broken ferromagnetic bonds. The net magnetic moment is zero. The difference of the two energies is

$$\Delta E = E_1 - E_2 = |J_2| - J. \tag{26}$$

Hence, if $|J_2| < J = 1$, the first situation of isolated impurities, is more favourable and $m > 0$, while if $|J_2| > J$, the domain-like pattern is chosen, yielding $m = 0$. In this case, for p close to 1, the 'domains' are quite small and they become larger with decreasing p. To create a wall, in a $N \times N$ system, approximately N impurity bonds are needed.

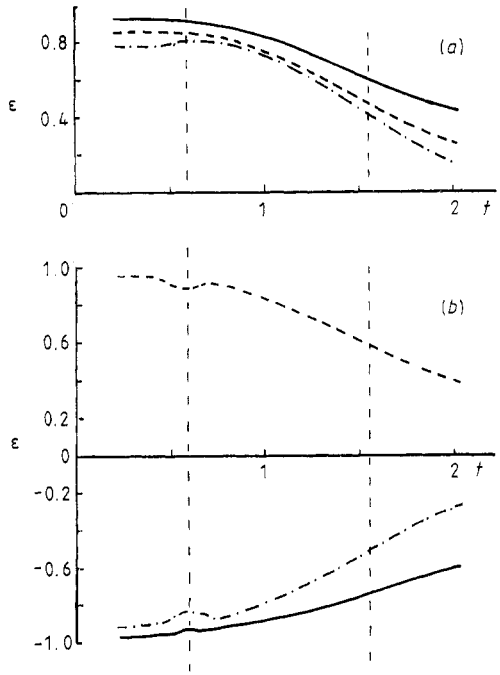

Figure 4. Model B. Correlation functions plotted against reduced temperature. $p = 0.48$, $J_2 = 1$. (*a*) Horizontal correlations. ——: $-\varepsilon_1$; - - -: $-\varepsilon_2$; — · —: $-\varepsilon_3$. (*b*) Diagonal correlations. ——: $-\varepsilon d_1$; - - -: $-\varepsilon d_2$; — · —; $-\varepsilon d_3$.

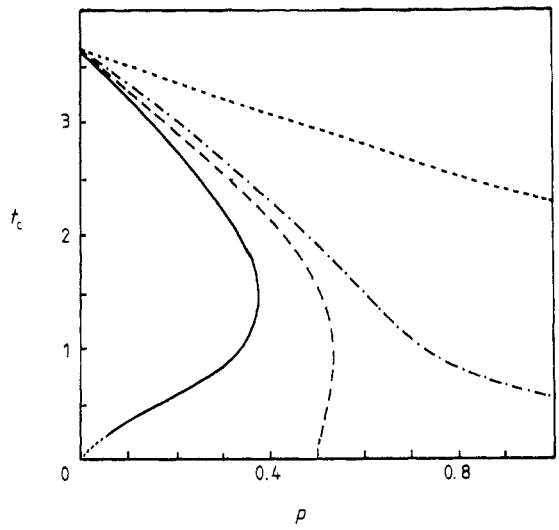

Figure 5. Model A. (t_c, p_A) phase diagram. ——: $J_2 = -1.3$; - - -: $J_2 = -1$; — · —: $J_2 = -0.8$; - - - -: $J_2 = 0$.

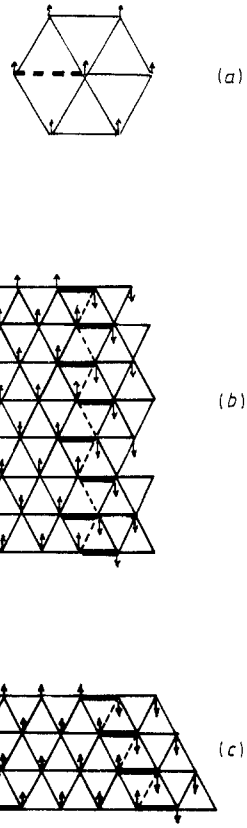

Figure 6. Model A. Impurity bond arrangements. Thick lines correspond to the impurity bonds, broken ones to unsatisfied (broken) bonds. (*a*) Isolated impurity, (*b*), (*c*) two domain-like patterns.

Therefore domain walls may exist even for $p \sim 1/N$. There are several ways of forming a wall, all corresponding to the same energy E_2. In a macroscopic system it is, however, irrelevant which one is realised.

To explain the effect of the temperature, we shall follow the reasoning of Oguchi and Ueno (1975), Derrida *et al* (1978) and Ueno (1986). Suppose that $J_2 < -1$. Then at $T = 0$ the domain-like state is energetically preferred. The pattern shown in figure 6(*b*) is not unique. There are, however, far more possibilities for the impurity bonds to be arranged randomly, inside a domain, than in the state of isolated impurities. As long as $T = 0$, the minimum of the free energy is equivalent to the minimum of the internal energy E. With increasing temperature the entropy, S, comes into play and the state the system is in is determined by

$$\min F = \min(E - TS).$$

Therefore the gain by the second (negative) term can exceed the loss through the first one connected with passing from the topologically ordered (but with $m = 0$) domain-like state into the state with scattered impurity bonds and $m > 0$. The transition is therefore driven by the entropy. The temperature destroys the topological order of the ground state by moving the impurity bonds out of the domain walls and into the domains. If

the domains are large, as is the case for small p (few domain walls), there are more available bond positions. This means the entropy is larger than it is for greater p, when the domains are narrower. Larger entropy means, in turn, that a lower temperature is needed to compensate for the loss of the internal energy, hence the entropy-driven transition into the ferromagnetic state occurs at lower temperatures than for larger p. With increasing p the upper critical temperature decreases as well, since there are more antiferromagnetic bonds in the system. Finally, at a threshold concentration, p_t, depending on J_2, the system has $m=0$ at all temperatures. For $T>0$, the impurity bonds are scattered but the ferromagnetic bonds are too weak, as compared with J_2, to sustain long-range order.

As seen from the comparison of the two magnetisation curves in figure 2, the entropy-driven transition is getting sharper at lower concentrations. This might be explained by the low temperatures at which it takes place and by weak thermal fluctuations of the spins.

The behaviour of the correlation functions is typical for a system with two transition temperatures (see, e.g., Kitatani *et al* 1985). For low temperatures all correlation functions reach constant values, different from those attained at high temperatures. This suggests the existence of magnetic ordering, even if the net magnetic moment is zero. Higher values of the diagonal correlations than the horizontal ones, as well as the values taken by both, indicate that the ground state may well have the form of a domain-like structure shown in figure 6(b), with a possible admixture of other types of domains, like the one shown in figure 6(c). The domains are, for $p_A=0.33$, two ferromagnetic couplings wide, which gives $\varepsilon_1>0$ but ε_2, $\varepsilon_3<0$. Decreasing the concentration to, say, $p_A=0.2$, leads to $\varepsilon_2>0$ and ε_3 only slightly negative. The diagonal correlations act predominantly within the domains and are therefore positive.

3.2. Model B

For $p_B=0$, which corresponds to the pure, completely frustrated, antiferromagnetic system, $t_c=0$ and there is no long-range order in the model. Therefore its (t_c, p_B) phase diagram (shown in figure 7) differs from that for model A. Irrespective of the value of the impurity coupling, the system cannot order ferromagnetically if $p_B<0.5$. If the impurities are antiferromagnetic and stronger than the host ones, there is no ordering for any concentration, in agreement with the earlier observation by Stephenson (1964). If the impurities are antiferromagnetic but weaker, ferromagnetic ordering along the horizontal lines appears for $p>0.5$. This is the reason that the re-entrance here manifests itself only for p slightly larger than $\frac{1}{2}$. The critical temperature grows as the impurity coupling strength approaches zero and becomes ferromagnetic. At that point the re-entrance appears and the region of concentrations over which it can be observed increases with increasing strength of the (ferromagnetic) impurity coupling. The region is, however, bounded. No matter how strong the impurity coupling, there cannot be a state with ferromagnetic ordering below $p_B=0.4$. The mechanism of the re-entrance is the same here as in model A. However, since the concentration of impurities is much higher, the possibility of deplacement is more resstricted, and consequently the role of the entropy is weaker. The ground state, shown in figure 8, has the form of ferromagnetic stripes coupled antiferromagnetically.

Hence εd_1 and εd_3 are always negative, while εd_2 and all ε are always positive. As in model A, this pattern is not the only one possible. When $p\neq 0.5$, coupling within the layers is not homogeneous. Because of the nearly antiferromagnetic ordering (for

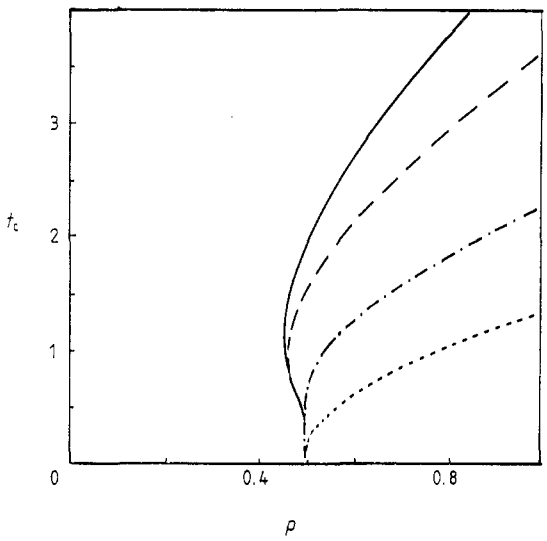

Figure 7. Model B. (t_c, p_B) phase diagram. ——: $J_2 = 2$; - - -: $J_2 = 1$; — · —: $J_2 = 0$; · · · ·: $J_2 = -0.5$.

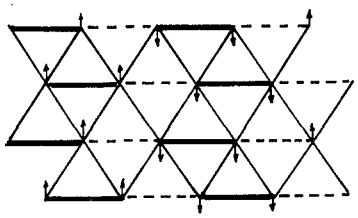

Figure 8. Model B. Bond arrangements in the ground state for $p_B = \frac{1}{2}$ and $J_2 = 1$. Thick lines: impurity bonds; broken ones: unsatisfied couplings.

$p \neq 0.5$) in the ground state, the correlation functions have larger values than in model A.

4. Conclusions

We have presented two Ising models on the triangular lattice with impurities which may affect only the horizontal bonds. In model A, the host system is ferromagnetic and in model B it is antiferromagnetic. The impurity bond in both models may have arbitrary value and sign. The models have been solved within the annealed approach and they both show, for certain concentrations of impurities and values of their coupling constants, the re-entrance phenomenon. In the ground state the system has zero magnetic moments which persist up to the lower critical temperature, t_{c1}. Further increase in the temperature produces a non-zero magnetisation, which vanishes again at the upper critical temperature, t_{c2}. This kind of behaviour can be explained by

assuming that, in the ground state, the system is split into domains of opposite magnetisation, separated by domain walls composed of impurities and broken bonds. For $J_2 \neq -J$ (model A) and $J_2 \neq J$ (model B), there is no frustration, but there are plaquettes with bonds which are 'broken' in the sense that the arrangements of the spins is determined not by bond coupling but by the free energy minimum. The temperature behaviour of the correlation functions, from which the ground-state pattern is deduced, shows that, for model A and $p_A = 1$, an antiferromagnetic state with tilted planes of spins of the same orientation is realised at $t = 0$. A decrease in p_A produces domains. Such a situation corresponds to the minimum of the energy, which is also the free energy minimum at $t = 0$. With $t > 0$ the entropy term becomes important, favouring random arrangement of the impurities. They move away from the domain walls, which are deformed. The domain pattern is destroyed and a state with non-zero magnetisation is created. Since $J_2 \neq J$, there is no frustration.

Models A and B exhibit several differences. For $p_B < 0.5$ there is no net magnetic moment in the ground state of model B, which evidently arises from complete frustration of the pure system. In contrast to model A, impurities in model B reduce the amount of frustrated plaquettes or broken bonds. At $t = 0$ and $p_B \leqslant 0.5$, the antiferromagnetic ordering is destroyed by thermal fluctuations of the spins and by displacements of the impurities, as a consequence of the annealed approach. The result is a continuous transition, at t_{c1}, from antiferromagnetic to ferromagnetic ordering. It is marked on the correlation functions diagrams by weakening of the correlations around t_{c1}, which increases with the range of the correlations (see figure 4).

Qualitative comparison of the results obtained in this paper with those already known shows that the phase diagram for model B with the common point, at $t = 0$, for all T_c curves is similar to that obtained by Syozi (1972). Also the existence of a threshold concentration below (or above, depending on the model) which in the ground state there is a net magnetic moment is known (see, e.g., Thorpe and Beeman 1976, Kitatani *et al* 1985). We have this situation for model B, irrespective of the coupling constants ratio, and in model A when the ferro- and antiferromagnetic couplings have the same strength. The appearance of the 'domain' structure is novel.

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