## RANDOM ANISOTROPY MAGNETS\*

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A review of theoretical, experimental and computational work on Random Anisotropy Magnetic systems is given. Monte Carlo simulations of two dimensional spins on a two dimensional lattice in the presence of random anisotropy fields have been carried out. In the absence of randomness this is the familiar XY model in two dimensions, which has a low temperature phase with algebraic order; this is the well-known Kosterlitz-Thouless phase. In the presence of random anisotropy evidence is presented that there are three phases: a low temperature orientational glass phase, an intermediate temperature Kosterlitz-Thouless phase, and a high temperature disordered paramagnetic phase.

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#### 1. Introduction

In these talks I shall be concerned with the magnetic properties of amorphous alloys containing rare earth metals. A wide class of such materials have been observed to have spin glass-like properties at low temperatures, and it is naturally of interest to understand what general features of these materials lead to such interesting behaviour. In this symposium much attention is being given to transitions between integrable and chaotic dynamical behaviour; a crucial feature of this transition concerns the disappearance of predictibility. Glass transitions have some features in common with this. At high temperatures the statistical mechanics of most materials can be predicted using classical methods — the Boltzmann statistics, partition functons and so on. The initial conditions of the system are essentially uninteresting. Beyond a glass transition, however, the initial conditions become all-important. Insurmountable energy barriers (in the sense that they

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grow with the size of the system) crop up. The behaviour of the system becomes less predictable, and from a statistical point of view, becomes non-ergodic; that is, the dynamical path of the system no longer has uniform probability over the Boltzmann surface in phase space.

I shall organise these talks in the following way. Following this brief introduction, I shall give a brief experimental overview in §2. Then, in §3, I shall discuss in greater detail the theoretical framework which is in wide use in order to understand the experiments, and also discuss the theoretical progress which has been made over the last fifteen years or so. In §4 I shall discuss computational results on model systems obtained by other workers. In §5 I shall present the main features of our own computational results on model of two dimensional random anisotropy magnets. Finally in §6 I shall make some concluding remarks.

## 2. Experimental systems

The subject of interest of this paper is the magnetic behaviour of amorphous alloys of rare earth metals [1]. Among systems which have been studied are the alloys Dy-Cu, Dy-Al, Tb-Fe<sub>2</sub>, Ho-Fe, Er-Co, Dy-Ni, Gd-Co-R, Tb-Co, Dy-Fe-B, Tb<sub>2</sub>Fe<sub>1-x</sub>Ni<sub>x</sub> (where R is a rare earth metal). These systems have magnetic properties with a number of common features. Among these are:

- a) A history dependent magnetisation. More specifically the magnetisation in a magnetic field H at low temperatures depends on whether the system has been cooled in that field (field cooled), or in its absence (zero-field cooled). Typically the field cooled samples show a large magnetisation, whereas a zero-field cooled samples show only a relatively small magnetisation. The two curves diverge at a temperature  $T_g(H)$  which one can identify with the onset of a glassy phase. We show in Fig. 1 a typical curve [2] of the magnetisation M as a function of temperature in the two cases. This result encourages one to believe that the low temperature phase of these materials is a spin glass phase, of the type much discussed in the literature in the case of dilute magnetic alloys such as Gd-Al or Cu-Mn [3].
- b) The glass temperature  $T_g(H)$  is field dependent; the higher the field, the lower the temperature has to be before the magnetic behaviour becomes history-dependent [4]. This behaviour is shown schematically in Fig. 2. This result is reminiscent of the De Almeida-Thouless line [5] predicted to exist in classical spin glasses, which separates regions in the phase diagram where the Boltzmann statistics describe the Statistical Mechanics from non-ergodic regions.

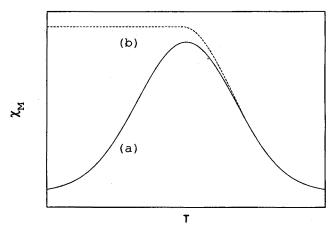


Fig. 1. Random Magnetic Anisotropy magnetisation (a) — field cooled and (b) — non-field cooled.

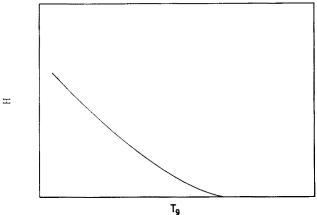


Fig. 2. Behaviour of glass transition temperature  $T_g(H)$  as a function of magnetic field.

- c) In practice, experiments to determine this dependence are carried out in AC fields. Classical spin glass phenomena appear to be time dependent, dynamic phenomena [6], in which the onset of glassy behaviour depends on the experimental time scale. One might expect these spin glasses to behave analogously, in which case the glass transition temperature would depend on the frequency of the AC magnetic field. Such frequency dependence is not, however, observed [4].
- d) From Fig. 1 it can be seen that there is a magnetic susceptibility maximum at  $T_g(0)$ . There are hints that this may even be a cusp, but there is no divergence of the type usually associated with a thermodynamic phase

transition. Occasionally there are even two peaks in the susceptibility  $\chi$  [7]. However, there do not seem to be the usual thermodynamic singularities in quantities such as the specific heat.

- e) The existence of a non-zero magnetisation in field-cooled samples (even when the field is removed) is consistent with the experimental observation of hysteresis curves, which are usually associated with the existence of bona fide magnetism. These systems exibit remanent magnetisation and coercive field, as do other more ortodox magnets, However, the hysteresis curves are bumpy, poorly reproducible, and dependent on the time-scale of the experiment.
- f) There is less consistency about the form of the low temperature correlations, which are usually measured in scattering experiments. Different authors have fitted their results for the magnetic component of the structure factor S(k) to a power law form [8]:  $S(k) \sim k^{-2.4}$ , giving rise to (in three dimensions) an expected form for the magnetic correlations:

$$\langle m(r)m(0)\rangle \sim r^{-0.6}. \tag{1}$$

Boucher et al. [9], on the other hand observe that their measurements of S(k) in RFe<sub>2</sub> compounds may be fitted to a  $k^{-\gamma}$  form at low k, and for higher k it can be fitted to a  $k^{-3}$  form. Other workers [10] find that S(k) can be fitted to a "Lorentzian plus a Lorentzian squared" form:

$$S(k) = \frac{A}{k^2 + \kappa^2} + \frac{B}{(k^2 + \kappa^2)^2}, \qquad (2)$$

where the Lorentzian squared term becomes preponderant at low temperatures.

#### 3. Theoretical models

#### 3.1. The HPZ model

The clue to a theoretical understanding of these materials seems to lie in the orbital angular momentum due to the electrons in the partly filled 4f shell; this feature is common to all the rare earth metals. In crystalline rare earth metals, this angular momentum couples with the crystal field due to the nearest neighbour atoms, causing a magnetocrystalline anisotropy with the symmetry of the underlying crystal lattice. Once the lattice is amorphous, however, and this may be true in pure materials, although it is much more common in alloys, the leading order effect of the anisotropy at each site is to create a term in the Hamiltonian which couples with  $\mathcal{J}_z^2$  (with  $\mathcal{J}$  the angular momentum operator), but with a special direction z

which now changes, randomly, from lattice site to lattice site. Thus the idea of a Random Anisotropy Magnet (RAM) came into being.

The insight of Harris, Plischke and Zuckermann [11], was that the magnetic properties of these systems depend crucially on the random magnetic anisotropy at each site, but that the precise topology of the lattice is unimportant. This strategy for dealing with the effects of disorder seems to have been successful in the past; Anderson, for instance, used it in order to successfully model the localisation of electrons in a desordered metal [12], and later in the classic Edwards-Anderson [13] spin glass model.

The HPZ model for RAMs is a lattice model with classical (i.e. not quantum) spins on each lattice site, and with energies dictated by local coupling between neighbouring spins and by coupling between spins and a magnetic field. The Hamiltonian is:

$$\mathcal{H} = -J \sum_{\{i,j\}} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_{l} (\mathbf{S}_i \cdot \mathbf{n}_i)^2 - D_0 \sum_{i} (\mathbf{S}_i \cdot \mathbf{n}_0)^2 - \mathbf{H} \cdot \sum_{i} \mathbf{S}_i, (3)$$

where the sums over pairs  $\{i,j\}$  are taken over nearest neighbours on a regular (usually cubic) lattice and all sums over sites i are taken over all sites of the lattice. The vector spins  $S_i$  are characterised by m components; normally we would expect m=3 (the so-called Heisenberg model), but the analysis of the model is sometimes simpler for m=2 (the XY model), without, one hopes, changing the physics dramatically.

There are four terms in the Hamiltonian (3). The first term is the magnetic exchange term, and is present in all magnetic models. The quantity J sets the energy and temperature scale for all magnetic processes in the material. The second term is the random anisotropy term. A crucial quantity in understanding the phase behaviour will be the quantity  $\lambda = D/J$ . The third term is due to coherent anisotropy (CA). This will occur, for instance, as a result of magnetoelastic interaction between a uniaxial stress field and the local magnetic moments. The final term is the interaction between an external magnetic field and the local spins. We shall be interested, first of all, in the phase diagrams in the parameter space of  $\tilde{T} = T/J$  and  $\lambda = D/J$ . We shall then discuss the effect of an external magnetic field, and finally possible complications due to the coherent anisotropy. We note also that this Hamiltonian does not take account of the long-range magnetic dipolar forces responsible for magnetic domain structure in permanent magnets.

At this stage we observe that although we have a well-posed mathematical problem, we are far from having solved it. As with many such problems in Statistical Mechanics, the regimes in which the problem can be solved exactly lie far from physical reality. We note three parameters which can be changed in this problem, and which will change the nature of the phase diagram (or at least the critical behaviour) as a function of  $\tilde{T}$ 

and  $\lambda$ . These are the <u>dimension</u> d, the <u>number of spin components</u> m and the <u>number of equivalent easy directions</u> p. The classical RAM model has p=2; the random <u>field</u> problem [14] has p=1. We shall now explore some solutions of the HPZ model in some easy limits. Except where we indicate it explicitly, we shall consider the H=0,  $D_0=0$  limits.

### 3.2. The Ising limit

A particular simple limit of the HPZ model occurs when  $D \to \infty$ . The spins on each site are then constrained to lie in one of two directions, parallel or antiparallel to the local easy axis. At each site we may arbitrarily assign a + direction; the value of each spin is then  $\pm 1$ , and the Hamiltonian reduces to:

$$\mathcal{H} = -\sum J_{ij} S_i S_j , \qquad (4)$$

where  $J_{ij} = J(\mathbf{n}_i \cdot \mathbf{n}_j)$ . This Hamiltonian is reminiscent of the Ising spin glass Hamiltonian [13]; formally, indeed, it has the same form. The difference lies in the method of generating the  $J_{ij}$  from an underlying distribution. In the Edwards-Anderson (EA) spin glass the interactions are thought of as being generated by the oscillating RKKY interaction, the spins representing dilute magnetic impurities. Thus the probabilities  $p(J_{ij}=1)=p(J_{ij}=-1)=1/2$ . By contrast in the  $D\to\infty$  limit of the HPZ model, presumably  $\mathbf{n}_i\cdot\mathbf{n}_j$  is a random quantity. However, one would not really expect this to have a dramatic effect on the phase behaviour. Both Hamiltonians incorporate the crucial element of <u>frustration</u>; it is impossible to arrange the spins in such a way as to minimize all the interaction energies at the same time. Another element in common is the existence of a gauge invariance; one can redefine the spins and interactions consistently in such a way as to leave the Hamiltonian invariant.

# 3.3. Spin glasses

At this stage it is worth giving a (very!) brief review of what is meant in the literature by a spin glass. For a much more comprehensive review we refer the reader to Ref. [3]. The dilute magnetic alloys now known as spin glasses were observed to have a cusp in the magnetic susceptibility at a temperature  $T_g$  at which it was hypothesised that local magnetic order became frozen in, although there is no global magnetic order. Edwards and Anderson [13] hypothesised Eq. (4) as a suitable Hamiltonian for such systems. An order parameter which describes such frozen-in magnetic order is:

$$q = \frac{1}{N} \sum_{i} |\langle \mathbf{S}_i \rangle^2|. \tag{5}$$

Careful computational and analytical studies [6, 15] of the Statistical Mechanics of the Hamiltonian (4) in two and three dimensions show that the low temperature phase does not have a strictly non-zero value, but the relaxation times become increasingly large; the glassiness is a <u>dynamic</u> phenomenon. The progress toward magnetic relaxation can be measured using a dynamic spin glass order parameter:

$$q(t) = \frac{1}{N} \sum_{i} \langle \mathbf{S}_{i}(\tau) \cdot \mathbf{S}_{i}(t+\tau) \rangle; \qquad (6)$$

the initial time  $\tau$  is taken in such a way that the system has already equilibrated. It is evident that  $q = q(t \to \infty)$ .

Only in four dimensions does it seem that there is a true spin glass phase. For two and three dimensions the crucial features of the low temperature spin glass phase seem to be:

- a) many <u>inequivalent</u> ground states, whose number goes up with system size;
- b) energy barriers between these states whose height goes up with system size:
- c) non-ergodic behaviour;
- d) an effective glass temperature whose exact value depends on the time scale of the experiment, the size of the system and the imposed magnetic field.

The question that evidently poses itself is: to what extent are any spin glass phases in RAMs analogous to these "classical" spin glass phases?

# 3.4. Basic theory

Over the last fifteen years a variety of methods have been used in order to study the HPZ Hamiltonian. These methods include mean field theory [16–18], the replica trick [19, 20], the Migdal–Kadanoff (M–K) bond-moving procedure [21] and the renormalisation group in  $4-\epsilon$  dimensions [22–24] and two dimensions [25]. There have also been a number of exact calculations in particular limits [26–28], and a large number of calculations based on implementations of the Imry–Ma [14] procedure by Chudnovsky and colleagues [29–33]. We defer till the next section a discussion of the Monte Carlo simulations of the HPZ model.

Exact calculations on the HPZ model in the Ising limit are particularly instructive. The calculation of Derrida and Vannimenus [25] of a similar problem in the limit of infinite range interactions (or, equivalently, infinite dimension) shows that in this limit, at least, the low temperature ferromagnetic phase is preserved. On the other hand, the calculation of Thomas [26]

shows that in one dimension the ground state is strictly non-magnetic. Intuitively this can be seen as being caused by a drift of the preferred direction through phase space as one goes along a chain. One can conclude that there is a lower critical dimension for the retention of the ferromagnetic phase. Indeed, an M-K calculation by Jayaprakash and Kirkpatrick [34] indicates a critical dimension of between 2 and 3. Presumably in this regime one should expect a spin glass low temperature phase, but the M-K method is unclear on this feature because there is no spin glass phase explicitly described by the model.

One can make estimates of critical temperatures using conventional Curie-Weiss mean field theory. This give rise to a critical temperature for the onset of ferromagnetism of  $T_c = zJ/2$  (in three dimensions) or  $T_c = 2zJ/\pi$  (in two dimensions), where z is the number of nearest neighbours on the lattice. By contrast, the same procedure predicts a spin glass transition temperature of  $J\sqrt{z/d}$ . By this criterion ferromagnetism should always win, because the transition to the spin glass phase occurs at a lower temperature. However, one can use methods first introduced by Imry and Ma [14] to check on the stability of such a ferromagnetic phase. We shall find that the ferromagnetic phase is unstable with respect to break up into domains of dimension  $\xi$ , where, for dimension d < d,  $\xi$  only diverges in the strictly nonrandom, d > 0, limit. These ideas have been more extensively developed by Chudnovsky and collaborators [29–33], although in what follows we shall merely follow the spirit of these ideas.

#### 3.5. Domain structure

# 3.5.1. The Imry-Ma-Chudnovsky argument

The crucial idea which we emphasize in this section is that the low temperature phase of the HPZ model involves a balance between the magnet trying to align with its neighbours, and trying to align along a local favourable direction. If the neighbours win, presumably the system will be ferromagnetic, but it will have to pay a price for not aligning along the easy axis. Conversely if the local axis wins, there is an energy price to pay for lack of local alignment. In practice, we shall find that there is (as so often in such problems) a compromise, in which there is ferromagnetic-like alignment over a coherent length scale  $\ell_m$ .

Let us calculate the order of magnitude of  $\xi_m$ . We suppose that the whole sample is broken up into domains of dimension  $\xi_m$ , as shown in Fig. 3. Within each domain the spins are (more or less) aligned along the average easy axis. The energy  $E_{\rm anis}$  associated with rotating the spins from a favourable to an unfavourable configuration is approximately  $DN_d^{1/2}$ , where  $N_d$  is the number of spins in the cluster. This follows from the central

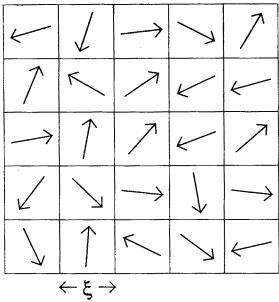


Fig. 3. Schematic break-up of a sample into magnetic domains of size  $\xi_M$ .

limit theorem; one is adding  $N_d$  random anisotropies. Now,  $N_d$  is related to  $\xi_m$  by the relation:

 $N_d \sim (\xi_m/a)^d \,, \tag{7}$ 

where a is a microscopic length scale of the material.

This energy must be balanced against the exchange energy  $E_{\rm exch}$ . This energy can be calculated by multiplying the energy of a *surface* between two domains by the total surface area caused by the existence of the domains. The surface area S of each domain is (to within numerical factors which are not important in this essentially dimensional argument):

$$S \sim \xi_m^{d-1} \,. \tag{8}$$

The surface free energy (per site)  $\gamma$  is governed by the fact that continuous spins can shift direction slowly, rather than abruptly. To shift direction by an angle  $\Delta\theta\sim o(1)$  over the distance  $\xi_m$  (which is all that is available between domains) involves a shift of angle  $\delta\theta\sim a/\xi_m$  between neighbouring spins. The energy price (relative to a purely ferromagnetic state), for each set of neighbours whose orientations are separated by  $\delta\theta$ , is given by:

$$\epsilon = J(1 - \cos \delta\theta) \sim \frac{1}{2}J(\delta\theta)^2 \sim J(a/\xi_m)^2$$
 (9)

Hence the surface energy per site is given by multiplying  $\epsilon$  by the number of sites contributing to the directional shift, which is approximately  $(\xi/a)$ , and hence:

$$\gamma \sim \epsilon(\xi_m/a) \sim J(a/\xi_m). \tag{10}$$

Finally the surface energy of each domain is given by:

$$E_{\text{dom}} \sim (\gamma/a^{d-1})S \sim J(\xi_m/a)^{d-2}. \tag{11}$$

We can now calculate the total energy of the sample:

$$A_{\text{tot}} \sim N_{\text{cell}}(E_{\text{each}} + E_{\text{anis}}),$$
 (12)

where  $N_{\text{cell}} \sim (L/\xi_m)^d$  is the total number of domains in a sample of dimension L. Substituting from Eqs (7)–(12), we obtain:

$$A_{\text{tot}} \sim (L/\xi_m)^d \left\{ -D(\xi_m/a)^{d/2} + J(\xi_m/a)^{d-2} \right\};$$
 (13)

yielding:

$$A_{\text{tot}} \sim (L/a)^d \left\{ -D(\xi_m/a)^{-d/2} + J(\xi_m/a)^{-2} \right\}.$$
 (14)

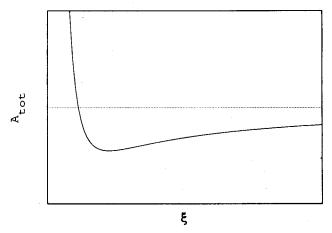


Fig. 4. Domain structure energy per site  $vs \ \xi_M$ , showing competition between exchange and random anisotropy terms.

Eq. (14) is the final result. The functional form of  $A_{\text{tot}}(\xi_m)$  is plotted in Fig. 4. For dimension d>4 this is minimised at  $\xi_m=\infty$ ; domain size diverges and the system is ferromagnetic. For d=4, this approach suggests two alternatives; either D< J (weak randomness), in which case  $\xi_m\to\infty$ , and ferromagnetism wins, or D>J (strong randomness), in which case  $\xi_m\to a$  (its minimum possible value), which corresponds to a spin glass phase. Finally for d<4, the anisotropy term dominates at high  $\xi_m$ , the exchange term dominates at low  $\xi_m$ , and in between there is a minimum, with the magnetic coherence length given by:

$$\xi_m \sim a(D/J)^{-2/(4-d)} \sim a\lambda^{-2/(4-d)}$$
. (15)

Thus  $\xi_m$  diverges in the limits of

- a) randomness disappearing (as is expected), and
- b) system dimension approaching four.

The lower critical dimension is four; we should not really expect our qualitative considerations to be too helpful in that régime.

It is of some interest to consider the physical specific cases of d=2 and d=3. For d=3, Eq. (15) yields  $\xi_m \sim \lambda^{-2}$ , whereas for d=2 one obtains  $\xi_m \sim \lambda^{-1}$ . In practice, of course, there are further complications for d=2 because even in the non-random case the low temperature phases are not ferromagnetic.

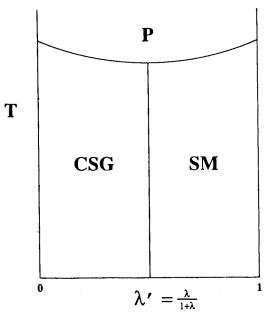


Fig. 5. Schematic phase diagram showing cluster spin glass spectromagnet and paramagnetic phases.

Qualitatively speaking, for the three dimensional case, we can identify two distinct régimes. For  $\lambda \lesssim 1$ , the domain dimension  $\xi_m$  will be larger than the intermolecular dimension, and a domain will consist of more than one spin. This régime has come to be known as a <u>cluster spin glass</u> (CSG) [29-33] (or occasionally as an asperomagnet [35]). By contrast, if  $\lambda \geq 1$ , the domains will only consist of one spin, the spins will essentially be uncorrelated with their neighbours, and the only effect of the neighbours will be to choose between the two local easy directions. This is the <u>speromagnetic</u> (SM) régime [35]. There is now experimental evidence concerning the existence of both of these phases [4,7,8,35]. We show in Fig. 5 a schematic phase diagram.

### 3.5.2. Locally correlated anisotropy

The argument of the previous section can be extended to take account of the related case of a polycrystalline sample. In such a case the anisotropy directions will be correlated over the dimensions  $\xi_c$  of the crystallites. The free energy per spin can be calculated by analogy with the methods of Eqs (7)–(14). The contribution from the exchange terms is unchanged. The direct contribution from the anisotropy to a domain energy is now given by the number of crystallites per domain; thus

$$E_{\rm anis} \sim -DN_{\rm cr}(N_{\rm d}/N_{\rm cr})^{1/2}; \qquad (16)$$

where  $N_{\rm cr}$  is the number of spins per crystallite and  $N_{\rm d}$  is, as before, the number of spins per domain. Now  $N_{\rm cr} \sim \xi_c^d$ , and combining Eqs (7), (14) and (16) yields:

$$A_{\text{tot}} \sim (L/a)^d \left\{ -D(\xi_m/\xi_c)^{-d/2} + J(\xi_m/a)^{-2} \right\}.$$
 (17)

Minimizing this for d < 4 yields:

$$\xi_m \sim \xi_c \{ (\xi_c/a)^2 \lambda \}^{-2/(4-d)}$$
 (18)

The smallest size of a magnetic domain must now be  $\xi_c$ ; the existence of  $CSG(\xi_m > \xi_c)$  or SM phases is now governed by the parameter  $\Lambda = \lambda(\xi_c/a)^2$ .

# 3.5.3. Magnetic susceptibility

From here on we return to systems in which the magnetic easy axes are not correlated from site to site. In order to calculate the magnetic susceptibility  $\chi_m$  we need to take account of the energy of interaction between the spins and an imposed magnetic field **H**. The total free energy change per site will be:

$$\Delta \epsilon = \Delta \epsilon_{\text{field}} + \Delta \epsilon_{\text{lattice}}, \tag{19}$$

where  $\Delta \epsilon_{\text{lattice}}$  comes from the change in energy of the terms due to exchange and anisotropy, and  $\Delta \epsilon_{\text{field}}$  comes from the magnetic field. Now, suppose that the effect of the magnetic field is to shift the mean direction at each site i by an angle  $\delta \theta_i$ . The magnetisation in the direction of the field at site i is  $m_i$ , and the shift as a response to the field H is  $\delta m_i$ . Now  $\delta m_i = \delta(\cos \theta_i) \sim \delta \theta_i$ , and hence:

$$\Delta \epsilon_{\text{field}} = -\langle (m_i + \delta m_i)H \rangle \sim -\delta m_i H \sim -H \delta \theta_i. \tag{20}$$

On the other hand, the shift in the lattice energy is necessarily positive and quadratic in  $\delta\theta_i$ , because in the absence of the field the energy is minimal.

Indeed the order of magnitude of the shift will be given by the energy in the absence of the field, and that has been implicitly calculated in Eqs (14) and (15). Thus:

$$\Delta \epsilon_{\text{lattice}} \sim \epsilon_{\text{lattice}} (\delta \theta_i)^2 \sim D \lambda^{d/(4-d)}$$
. (21)

Equilibrium will occur when  $\Delta \epsilon_{\text{field}}$  and  $\Delta \epsilon_{\text{lattice}}$  are of the same order of magnitude; this is equivalent to minimising the expression (19). Comparing Eqs (20) and (21), we obtain that:

$$\delta m_i \sim \delta \theta_i \sim H/H_c$$
, (22)

with the characteristic field  $H_c = D\lambda^{d/(4-d)}$ . Thus the magnetic susceptibility is given by:

$$\chi_m \sim H_c^{-1} \sim D^{-1} (J/D)^{d/(4-d)} \sim D^{-4/(4-d)}$$
. (23)

This result is particularly interesting as it predicts a finite magnetic susceptibility in the low temperature phase, in contrast to the prediction of Aharony and Pytte [22], who predicted an infinite susceptibility phase. The field  $H_c$  has the following significance: it is that field at which the approximation that the angular shift at each site be small breaks down. For  $H < H_c$  the original domain structure is maintained, but each spin is somewhat perturbed in order to appease the magnetic field. Beyond  $H_c$  a new situation obtains, in which the spins are primarily aligned in the field direction, and one must consider perturbations from that primary direction. We discuss this situation in the next section.

## 3.5.4. Ferromagnet with wandering axes

In the high magnetic field limit the spins primarily align themselves in the direction of the field. However, there are deviations from this direction which occur in the attempt to keep the random anisotropy and the exchange happy. As in the last section we can write the total energy per spin as a sum of the three contributions:

$$\epsilon = \epsilon_{\text{exch}} + \epsilon_{\text{field}}. \tag{24}$$

Let us now suppose that a typical deviation of the spin from the field direction is  $\theta$ , and that such deviations are typically correlated over distances  $\xi_H$ . The system is now broken up into clusters of size  $\xi_H$  each pointing more or less uniformly in a direction departing from the mean direction by angle of order  $\theta$ , giving rise to the idea of a ferromagnet with wandering axes. The separate contributions can now be evaluated. Following the ideas of Eq. (9),

 $\epsilon_{\rm exch} \sim J(a\theta/\xi_H)^2$ . The magnetic field contribution, measured from a zero in which all spins are aligned, is  $\epsilon_{\rm field} \sim mH \sim H(1-\cos\theta) \sim H\theta^2$ . The effect of the anisotropy is also measured with respect to a zero in the fully aligned state. The mean alignment energy available per spin, on the assumption that each domain were completely aligned with the average local axis, is now, following the discussion accompanying Eq. (7),  $-D(a/\xi_H)^{d/2}$ . However, the mean canting toward the direction  $(\Phi, \text{say})$  is only  $\theta$ , and so the energy shift is:

$$\epsilon_{\rm anis} \sim D(a/\xi_H)^{d/2} \left\{ \cos^2(\Phi - \theta) - \cos^2 \Phi \right\} \sim -D(a/\xi_H)^{d/2} \theta$$
. (25)

In equilibrium the three contributions will have equal order of magnitude, yielding equations for  $\theta$  and  $\xi_H$ :

$$\xi_H \sim a(J/H)^{1/2};$$
 (26)

and

$$\theta \sim (H_c/H)^{1/4} \,, \tag{27}$$

where the characteristic field  $H_c$  has been defined in Eq. (23) above. The crucial experimental quantity is now the departure  $\delta m$  of the magnetisation from the saturated value; this is:

$$\delta m \sim \theta^2 \sim (H_c/H)^{1/2} \,. \tag{28}$$

This argument will work so long as the cluster dimension size  $\xi_H$  can be regarded as variable. This will *not* be the case for a SM (in which case  $\xi_H \sim a$  always) or for a CSG in the high field régime, for which  $\xi_H \sim a$ , or equivalently  $H \gg H_{c2} \sim J$ . In either of these situations the exchange energy can be ignored, and now the condition that  $\epsilon_{\rm field} \sim \epsilon_{\rm anis}$  yields:

$$D\theta \sim H\theta^2$$
, (29)

or

$$\theta \sim D/H$$
, (30)

yielding

$$\delta m \sim (D/H)^{-2} \,. \tag{31}$$

## 3.5.5. Coherent anisotropy

In the final section of domains we discuss the effect of reintroducing a non-random anisotropy (CA) to the system. This is the third term in the Hamiltonian (3). As discussed above this might be caused by externally imposed stress and a magnetoelastic effect. A non-random anisotropy by

itself favours the presence of a ferromagnetic ground state. The obvious question to pose is whether the same effect is induced in the presence of the RAM. The answer to this question seems likely to be yes, so long as the CA is large enough. The following Imry-Ma-Chudnovsky argument provides an order of magnitude estimate.

We first calculate the thickness of the Bloch wall between two domains of opposite polarity in the presence of CA of magnitude  $D_0$ . The anisotropy energy associated with a region of thickness  $\delta_B$  in which the spins are turning is approximately  $D_0(\delta_B/a)$ . The angle between spins in this region is of order  $a/\delta_B$ , and hence the exchange energy price to be paid per spin is approximately  $J(a/\delta_B)^2$ . The total exchange energy price comes from multiplying this by the number of spins,  $\sim \delta_B/a$ , participating in the wall region; this is  $\sim J(a/\delta_B)$ . The thickness  $\delta_B$  can be calculated by requiring that the exchange and anisotropy energies are of the same order of magnitude:

$$J(a/\delta_B) \sim D_0(\delta_N/a);$$
 (32)

OF

$$\delta_B \sim a(D_0/J)^{-1/2}$$
 (33)

The Bloch wall thickness  $\delta_B$  must be compared with the characteristic domain dimension  $\xi_m$  in the presence of the random anisotropy. The length  $\delta_B$  is the minimum length scale over which the system can respond to the presence of a non-zero  $D_0$ . However, due to the random anisotropy term the system is in any case changing its magnetism on length scales of order  $\xi_m$ . Thus if  $\delta_B > \xi_m$  the CA will simply be ignored. However, if the reverse is the case, the system can respond to the CA, and so magnetism will be restored. The condition for magnetism can be reexpressed as:

$$(D_0/J) > (D/J)^{4/(4-d)}$$
. (34)

Interesting problems which have not so far been addressed in the literature, but which can surely be discussed using this approach, include the behaviour of the critical temperature and the low temperature magnetic susceptibility in the presence of CA.

### 3.6. The two dimensional XY model

The system with smallest m and d for which there are non-trivial magnetic effects is the two dimensional XY model, for which m=2 and d=2. From a computational point of view is important; one always starts with the system which will run in the shortest amount of time and use the smallest amount of memory! Unfortunately, as is by now well-known, the two dimensional XY model in fact exhibits highly non trivial behaviour.

We first recall the salient features of the non-random, D = 0, two dimensional XY model. The high temperature phase is paramagnetic, with only local magnetic correlations, which decay exponentially, with  $\langle \mathbf{m}(0) \cdot \mathbf{m}(\mathbf{r}) \rangle \sim$  $e^{-r/\xi}$ , where the magnetic correlation length increases with decreasing temperature. On the other hand, relatively simple spin wave arguments show that there is no low temperature ferromagnetic phase. Nevertheless, the magnetic correlations only die off algebraically, with  $\langle \mathbf{m}(0) \cdot \mathbf{m}(\mathbf{r}) \rangle \sim r^{-\eta}$ . The exponent n(T) increases from 0 at T=0, where the system is perfectly ordered, to 1/4 at a phase transition temperature  $T_{KT}$ , beyond which the system is paramagnetic. The first detailed analysis of the low temperature properties of this somewhat anomalous system was made about twenty years ago by Kosterlitz and Thouless [36, 37] emphasized the importance of vortices as important excitations whose interactions must be described accurately in any theory. Thus for this case the low temperature phase can truthfully be said to be almost magnetic. The question is to what extent does any added random anisotropy disrupt this almost magnetism. It is interesting to note that this model is also applicable to the study of liquid helium absorbed on rough surfaces.

The problem has been attacked theoretically by Dotsenko and Feigelman [38], Houghton, Kenway and Ying [39], and Cardy and Ostlund [25]. These authors use the replica trick and derive renormalisation group equations which generalise the classic studies of the XY model by Kosterlitz [37] and Young [40]. The general consensus is that, for a given degree of randomness, the Kosterlitz-Thouless algebraically ordered phase does survive, but that at lower temperatures still, there will be, for sufficiently large number p of axes of anisotropy, some kind of glassy phase. In the study of Cardy and Ostlund the critical value of p is 3; our case (p=2) is really outside the range of validity of that theory. We shall see below, however, that even the p=2 case exhibits this general property.

# 4. Review of previous computational work

The variety of theoretical predictions for the phase behaviour of the HPZ model have inspired attempts to determine this behaviour experimentally. In this case the experiments are Monte Carlo simulations of the statistical mechanics, using the Metropolis algorithm [41] and variations thereof. In practice, however, the application of the Metropolis algorithm in random systems is beset with difficulties and dangers, some of which we have encountered ourselves, and shall describe below in greater detail.

Early studies by Chi and Alben [42] working with the HPZ model on a random *lattice*, indicated that, even in the presence of randomness, the ground state of the HPZ model was magnetic. Subsequent work by Chi

and Egami [43], who used, not a Monte Carlo algorithm, but a molecular dynamical relaxation algorithm, showed that this was merely a finite size effect. These studies showed a qualitative resemblence between the finite field properties and real experimental behaviour in RAMs. However, they were hampered by simple problems of the computational power available at that time; both these studies used only 10<sup>3</sup> sites. Further work was carried out by Harris and Sung [44,45], who faced similar problems in determining the nature of the low temperature phase with even fewer lattice sites.

Jayaprakash and Kirkpatrick [34] studied in great detail the Ising limit discussed in §3.2 above. Their systems were larger; up to  $20^3$  sites, and their simulations ran for up to  $4 \times 10^5$  Monte Carlo cycles (1 cycle = 1 Monte Carlo move for each particle). Their conclusion was that although there was evidence for a stable low temperature phase, but there is no long range ferromagnetic order in this phase, and no obvious critical behaviour delineating the crossover from the low temperature to the high temperature régime. Nevertheless there was a relatively narrow peak in the specific heat  $C_V$  which presumably marked the onset of the low temperature phase, and, more interestingly, measurements of hysteresis curves gave signatures which might have been thought to be consistent with magnetic phases. In particular, a remanent magnetisation was observed, and a coercive field which increases with decreasing temperature.

Chakrabarti [46] took one aspect of this work further and calculated spin glass order parameters, concluding that the low temperature phase was characterised by long-time local spin correlations, as would be expected in a spin glass. These systems were smaller, however: only 402 in two dimensions, and 123 in three dimensions. This work has in any case been criticized by Fisch [47], who pointed out that, for any dynamical studies, the classical Monte Carlo algorithm (in which each spin is moved independently of its neighbours) can give rise to extremely long relaxation, thus calling into question spin glass order parameter data. Fisch's paper made a very careful stude of the ground state of a HPZ XY system (in the "Ising" limit in the sense of §2.2) with 163 sites, using a sophisticated Monte Carlo algorithm which allowed for small clusters to rotate, as well as single spins. He found a well-defined ground state, with algebraic correlations. In further work [48]. he has extended this to finite temperatures (and large lattices), showing that a well-defined "freezing" temperature exists in which there are algebraic spin correlations which seem to be consistent with some experimental results.

Finally some mention should be made of some work by Serota and Lee [49] who investigated the ground state of a one dimensional RAM model, and by Dieny and Barbara [50], who investigated the behaviour of a two dimensional HPZ XY model in a finite field. This work discovered that the

magnetic relaxation is heavily affected by vortex dynamics and in particular by vortex-antivortex annihilation.

## 5. Monte Carlo study of 2D XY random anisotropy magnet

### 5.1. Preliminaries and computational details

We have made a detailed study of the properties of the two dimensional XY HPZ model given by Eq. (3). We have not considered the effect of coherent anisotropy, but we have considered the effect of changing temperature  $\tilde{T}$ , randomness  $\lambda = D/J$  and magnetic field H. We have looked at system sizes between  $8^2$  and  $128^2$ , using the traditional Monte Carlo algorithm in which each spin is moved independently of all others, and have examined results of simulations with variable run times between 500 and  $10^6$  cycles. Among physical variables monitored were hysteresis curves with associated coercive fields and remanent magnetisations, magnetic susceptibility, specific heat and spin-spin correlations as a function of both space and Monte Carlo time. A preliminary report of some of these results has been presented elsewhere [51], and a more detailed presentation will also be made.

The simulations were run on a Meiko system with a parallel architecture, with 32 T-800 transputers contributing to the simulation. Each transputer carries 256 kbytes of memory and contributes roughly 1 Mflop ( $10^6$  floating point operations per second). Most of our production runs were made on systems of  $64^2$  lattice sites, though since one of the transputers developed a hardware fault this has been reduced to  $62^2$ . A schematic diagram of the computational system is shown in Fig. 6.

#### 5.2. Results

A common criterion for the existence of magnetism in a given material is the presence of hysteresis loops in the M vs H characteristic and wew have seen that the hysteresis loops of amorphous rare earth alloys show bumpy and irreproducible features. In Fig. 7, we show a typical hysteresis loop observed in one of our computer experiments. (Irreproducible) steps in the M vs H characteristic, which bear considerable resemblance to features observed experimentally, can be seen. Similar curves were seen by Dieny and Barbara [51] in their zero temperature simulations of precisely the same model. We can identify two crucial magnetic fields in this run;

- a) the coercive field  $H_{co}$ , required to (just) reverse the magnetisation direction, and
- b) the critical reversible field  $H_{rev}$ , required that the same magnetisation be no matter what the history of the sample.

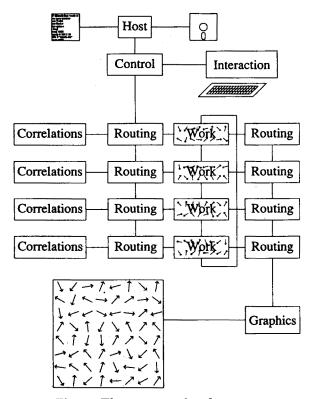


Fig. 6. The computational set-up.

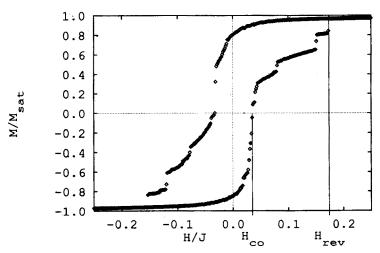


Fig. 7. A typical computer hysteresis curve at T/J = 0.1, D/J = 0.4, showing irreproducible jumps.

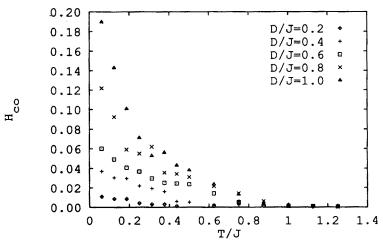


Fig. 8. Coercive field  $H_{co}(T)$  for various values of D.

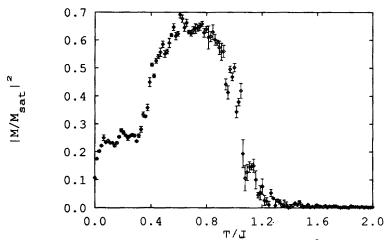


Fig. 9. Magnetisation as a function of temperature for a  $64^2$  lattice quenched to T=0, and subsequently warmed.

It is helpful to collect together the hysteresis data from many different simulations. The curve  $H_{\rm rev}(T)$  for a given D may be thought of as analogous to the De Almeida-Thouless line [5] separating ergodic and non-ergodic regions in the spin-glass phase diagram. We find that it is easier computationally to plot  $H_{\rm co}(T)$ ; the ratio of  $H_{\rm co}$  and  $H_{\rm rev}$  is more or less constant in any given situation. A series of such plots, for various values od D, is shown in Fig. 8. It will be seen that all hysteresis ceases above  $T_{\rm c} \simeq 1.0$ , which we identify with the Kosterlitz-Thouless ordering temperature, but that for low values of D hysteresis appears to cease at lower temperatures. There is, however, only very weak dependence of these curves on the magnetic field

sweep rate dH/dt; these simulations typically involve 5000 cycles per field point.

We have also studied the behaviour of quenched systems. In Fig. 9 we show the behaviour of M(T) at zero magnetic field for a system quenched from a random configuration to T=0 and slowly heated up. It will be seen that the magnetisation reaches a maximum at  $T \simeq 0.4$ , suggesting a glassification transition at about this temperature. Further studies over very long relaxation times confirm this point of view. These studies start with a perfectly aligned state and run for 106 cycles. From these long runs the following picture emerges. At very low temperatures the system is glassified and the final (stable) configuration remembers the initial configuration. At higher temperatures the initial configuration is forgotten and the system on average lies along the easy anisotropy direction closest to the initial direction. At higher temperatures still, even this memory is forgotten, and the system is in a Kosterlitz-Thouless state; apparently in this regime the randomness is irrelevant. Finally above the Kosterlitz-Thouless temperature the spins are completely disordered. This seems to be more or less independent of D in the systems we have tested, and finite size scaling studies identify reasonably unambiguously the crossover between the last two behaviours.

The intersting régime is the Kosterlitz-Thouless region. Here there is a characteristic "flip-time" over which the system reverses its direction from one easy axis to the other. This flip time increases with D and system size, and decreases as a function of temperature. We have not, however, yet been able to make a definitive prediction about how it extrapolates to infinite system size. More detailed studies of this relaxation and of the relaxation of the spin glass order parameter suggest that a general property of the Kosterlitz-Thouless phase is that relaxation times increase with a power law dependence on system size, although we have not been able to determine that power law. This would be consistent with the idea that this phase is everywhere critical. Thus the distinction that we draw above between flips and one which does not seems to depend on the time scale of the experiment; for sufficiently long times at any given system size the system will flip. Conversely, for any given time scale, a sufficiently large system will not flip.

Other measurements reinforce this view of three separate phases, which, incidentally, is consistent with the theoretical picture which emerged in Refs [25,38,39]. Measurements of  $|M|^2$  as a function of T, for different values of D, are shown in Fig. 10. In these studies the systems were cooled slowly. Whilst this quantity is surely *not* a thermodynamic quantity (it is dependent on system size L), the differences between the curves do indicate at what point the randomness causes the system *not* to be Kosterlitz—

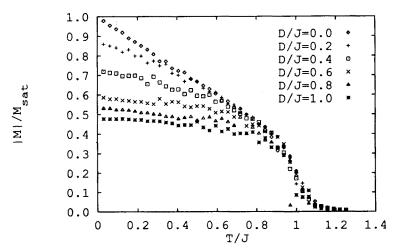


Fig. 10.  $|M^2(T)|$  for a 64<sup>2</sup> lattice for systems slowly cooled for various values of D, showing different behaviour for different degrees of randomness.

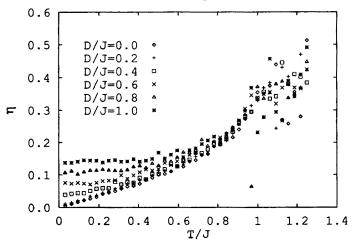


Fig. 11. The spin correlation decay exponent  $\eta(T)$  for different values of D in slowly cooled samples. Note that for  $T > T_{\rm KT}$ , the fits give  $\eta > 1/4$ , but are not consistent between different size systems, and where effective value of  $\eta$  depart from Kosterlitz-Thouless value in low temperature régime these fits are also not reliable or consistent.

Thouless-like, and hence, presumably, delineates a phase boundary. We have also examined the spin correlations, both for cool and for quenched samples. The spin correlations have been fitted to the form:

$$g(r) \sim r^{-\eta} \exp -(r/\xi). \tag{35}$$

The results for  $\eta(T)$  for a number of different values of D in the case of the cooled samples are shown in Fig. 11. They show a dramatic difference

between the intermediate temperature régime, in which the spin correlations are algebraic and exactly as predicted in the Kosterlitz-Thouless theory, and a low temperature régime. In this region the spin correlations fall off more quickly, the form of g(r) can not be fitted well to the hypothetical form of Eq. (35), and, interestingly, the form of g(r) itself seems to depend on the history of the sample. In the high temperature régime the correlations fall off exponentially.

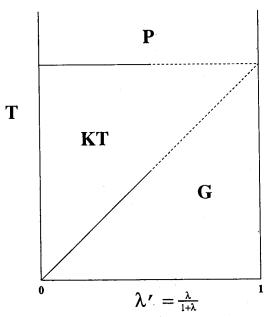


Fig. 12. Phase diagram resulting from computational study in parameter space of  $\tilde{T} = T/J$  and  $\lambda = D/J$ , showing paramagnetic (P), Kosterlitz-Thouless (KT), and glassy (G) phases.

Finally putting together these studies, we are able to draw a tentative phase diagram, shown in Fig. 12, which delineates the various régimes.

### 6. Conclusions

In this article we have given a review of recent work on random anisotropy magnets, concentrating on theoretical and computational studies. We have presented some of our own recent results on the XY model in two dimensions with random anisotropy. We find, as do other authors, strong evidence of irreversible behaviour and a glassy phase in the low temperature régime, but at this stage it is not yet clear whether this is true spin-glass-like order. Further studies will enable one to do more detailed comparison

between simulation and both experiment and theory; space has precluded us giving a more detailed description of the evidence we have gathered so far.

I should like to thank the organisers of this symposium for inviting me to give this talks in such a convenial environment. I should also like to thank my other hosts in Poland, Dr Andrzej Poniewierski, Dr Alina Ciach and Professor Jan Stecki in Warsaw, and Dr Lech Longa in Cracow, for the effort that they have gone to in order to make my stay in Poland as pleasent as it has been. In addition my collaborators and I thank the Southampton University Research Fund and SERC for providing support for D.R.D. We are grateful to Brian Carpenter, John Chalker, and in particular, Peter de Groot, for useful comments and conversations during the course of this work.

#### REFERENCES

- [1] A comprehensive early review was given by R.W. Cochrane, R. Harris, M.J. Zuckermann, *Phys. Rep.* 48, 1 (1978).
- [2] B. Dieny, B. Barbara, Phys. Rev. Lett. 57, 1169 (1986).
- [3] See e.g.: (a) J. Souletie, Ann. Phys. Paris 10, 69 (1983); (b) M. Mézard, G. Parisi, M.A. Virasoro, Spin Glass Theory and Beyond, World Scientific, Singapore 1987, and references therein.
- [4] D.J. Sellmyer, S. Nafis, Phys. Rev. Lett. 57, 1173 (1986).
- [5] J.R.L. de Almeida, D.J. Thouless, J. Phys. A 11, 983 (1978).
- [6] See e.g. H. Sompolinsky, A. Zippelius, Phys. Rev. B25, 6860 (1982).
- [7] K.M. Lee, M.J. O'Shea, J. Appl. Phys. 63, 3740 (1988).
- [8] S.J. Pickart, H.A. Alperin, J.J. Rhyne, Phys. Lett. 64A, 377 (1977).
- [9] B. Boucher, A. Lienard, J.P. Rebouillat, J. Schweizer, J. Phys. F 9, 1433 (1979).
- [10] P.A.J. de Groot, B.D. Rainford, S.H. Kilcoyne, R. Cywinski, L. Cussen, E. Lindley, *Physica* B156-7, 208 (1989).
- [11] R. Harris, M. Plischke, M.J. Zuckermann, Phys. Rev. Lett. 31, 160 (1973).
- [12] P.W. Anderson, Phys. Rev. 109, 1492 (1958).
- [13] S.F. Edwards, P.W. Anderson, J. Phys. F 5, 965 (1975).
- [14] Y. Imry, S.K. Ma, Phys. Rev. Lett. 35, 1399 (1975).
- [15] N.D. McKensie, A.P. Young, Phys. Rev. Lett. 49, 301 (1982).
- [16] R. Harris, D. Zobin, J. Phys. F 7, 337 (1977).
- [17] E. Callen, Y.J. Liu, J.R. Cullen, Phys. Rev. B16, 263 (1977).
- [18] J.D. Petterson, G.R. Gruzalski, D.J. Sellmyer, Phys. Rev. B18, 1377 (1978).
- [19] J.H. Chen, T.C. Lubensky, Phys. Rev. Lett. B16, 2106 (1977).
- [20] V.S. Dotsenko, M.V. Feigelman, J. Phys. C 16, L803 (1983).
- [21] R. Fisch, Phys. Rev. B41, 11705 (1990).

- [22] A. Aharony, E. Pytte, Phys. Rev. Lett. 45, 1583 (1980).
- [23] D. Mukamel, G. Grinstein, Phys. Rev. B25, 381 (1982).
- [24] R.A. Pelcovitz, E. Pytte, J. Rudnick, Phys. Rev. Lett. 40, 476 (1978).
- [25] J.L. Cardy, S. Ostlund, Phys. Rev. B25, 6899 (1982).
- [26] B. Derrida, J. Vannimenus, J. Phys. C 13, 3261 (1980).
- [27] H. Thomas, in Ordering in Strongly Fluctuating Condensed Matter Systems, ed. T. Riste, Plenum, New York 1980.
- [28] A.J. Bray, M.A. Moore, J. Phys. C 18, L139 (1982).
- [29] E.M. Chudnovsky, R.A. Serota, J. Phys. 16, 4181 (1983).
- [30] E.M. Chudnovsky, R.A. Serota, Phys. Rev. B26, 2697 (1982).
- [31] E.M. Chudnovsky, W.M. Saslow, R.A. Serota, Phys. Rev. B33, 251 (1986).
- [32] E.M. Chudnovsky, J. Appl. Phys. 64, 5770 (1988).
- [33] E.M. Chudnovsky, J. Mag. Mag. Mat. 79, 127 (1989).
- [34] C. Jayaprakash, S. Kirkpatrick, Phys. Rev. B21, 4072 (1980).
- [35] J.M.D. Coey, J. Appl. Phys. 49, 1646 (1978).
- [36] J.M. Kosterlitz, D.J. Thouless, J. Phys. C 6, 1181 (1972).
- [37] J.M. Kosterlitz, J. Phys. C 7, 1046 (1974).
- [38] V.S. Dotsenko, M.V. Feigelman, J. Phys. C 14, L823 (1981).
- [39] A. Houghton, R.D. Kenway, S.C. Ying, Phys. Rev. B23, 298 (1981).
- [40] A.P. Young, Phys. Rev. B19, 1855 (1979).
- [41] N. Metropolis, A.W. Rosenbluth, M.N. Rosenbluth, A.N. Teller, E. Teller, J. Chem. Phys. 21, 1087 (1953).
- [42] M.C. Chi, R. Alben, J. Appl. Phys. 48, 2987 (1977).
- [43] M.C. Chi, T. Egami, J. Appl. Phys. 50, 1651 (1979).
- [44] R. Harris, S.H. Sung, J. Phys. F 8, L299 (1978).
- [45] R. Harris, J. Phys. F 10, 2545 (1980).
- [46] A. Chakrabarti, J. Appl. Phys. 63, 3735 (1988).
- [47] R. Fisch, Phys. Rev. B39, 873 (1989).
- [48] R. Fisch, Phys. Rev. Lett. 66, 204 (1991).
- [49] R.A. Serota, P.A. Lee, Phys. Rev. B34, 1806 (1986).
- [50] B. Dieny, B. Barbara, Phys. Rev. B41, 11549 (1990).
- [51] D.R. Denholm, B.D. Rainford, T.J. Sluckin, J. Mag. Mag. Mat. (in press).