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Abstract. We present a physically realizable random field model in the form of dilute crystals undergoing structural phase transitions, with the impurities generating a random strain field. The mode softening that occurs at the transition is anisotropic. We show how this anisotropic mode softening reduces the upper critical dimension from 6 to 4. We perform an ϵ-expansion at three dimensions about the upper critical dimension, from which we obtain static critical exponents which, to $O(\epsilon)$, are equal to those corresponding to the pure three-dimensional Ising model. The crossover behaviour is described. Elastic long-range forces reduce the upper critical roughening dimension from 5 to 3. We find that even at the upper critical roughening dimension of 3, domain wall roughness is not eliminated. This is due to anisotropic shape effects in the domain walls, induced by the anisotropic long-range forces. This is an instance of how the concept of the upper critical roughening dimension is not very useful for understanding domain wall roughness and dynamics for models with anisotropic interactions. Given the connection between domain wall roughness and dynamics in random field systems, for our model at three dimensions, metastability and long relaxation times associated with random field models remain, though reduced.

1. Introduction

Random field models have long posed theoretical difficulties with both their static and dynamic critical properties [1–5]. So far attention has mostly been focused on the random field models with short-range interactions. One exception is the random field Ising model with long-range dipolar interactions, studied by authors like Nattermann [6]. This work studies structural phase transitions with random strains as an example of a random field model with long-range forces. Most experiments on random field models have been done on dilute antiferromagnets in a uniform field (DAFF). Relatively few have been done on our model, i.e. impure crystals.

Structural phase transitions with random strains are a realization of a random field model, arising from different physical effects, e.g. electronic band structure, ferroelectric fluctuations, ferroelastic mode softening and the Jahn–Teller effect. The order parameter ϕ is the strain tensor. In some special cases, the symmetry is such that ϕ collapses into a one-component vector, and only quadratic ϕ^2 and quartic ϕ^4 terms are allowed in the Hamiltonian of the pure system. We then get an Ising model with a second-order phase transition. The important difference is that structural phase transitions undergo mode softening along restricted directions in momentum k-space dictated by the above-mentioned elastic constant and lattice symmetries. This translates into anisotropic direction-dependent quadratic terms in k-space in the

Hamiltonian, whereas the random field model with short-range nearest-neighbour interactions (short range RFIM) contains only isotropic terms.

Nattermann [6] investigated the role of long-range dipolar forces in random field systems, its effects on the static equilibrium properties and the growth of metastable domains. For the random field Ising model (RFIM), dipolar forces lower the upper critical dimension by 1 but leave the lower critical dimension unaffected. The static critical exponents are altered. These changes can be viewed in terms of the bare propagator of a pure Ising model with dipolar interactions $G(q) = [T - T_0 + q^2 + g(q_z/q)^2]^{-1}$. Dipolar forces induce the anisotropic $g(q_z/q)^2$ term which alters the integrals in a perturbation treatment of static and interface properties. Following the approach of Nattermann [6], we study an RFIM with a bare propagator of the corresponding pure model with another type of anisotropy.

Random fields move the system to a disordered universality class, while long-range forces move the system towards the pure mean-field class. In our model, how do the competing tendencies of random fields and long-range forces interact? We will address this question in the static and dynamical aspects. Is our model more like a random field model or a pure model? One motivation for our work was to investigate to what extent long-range forces reduce the problems that have made random field models intractable to analysis and a source of controversy. On the static front, we investigated how the long-range forces modified the upper critical dimension, the critical exponents and the crossover behaviour. On the dynamic front, we investigated how the long-range forces modified the upper critical roughening dimension, the domain wall roughness and the dynamics. A major difference between random field models and pure models lies in the dynamical behaviour. Random field models exhibit much longer critical slowing down times than pure models. This is due to metastable effects absent in pure models. One major motivation for this work is to investigate to what extent our long-range forces reduce the slowing down times and metastability.

One aim of this paper is to show how anisotropic mode softening tends to lower the upper critical dimension of the system. Random fields raise the upper critical dimension [9, 10], while anisotropy lowers it [7]. As will be shown later in this paper, we rigorously demonstrate that the upper critical dimension is 4, which is lower by 2 than the short-range RFIM upper critical dimension, and equal to the pure short-range Ising model upper critical dimension. Expanding about this upper critical dimension, using renormalization group (RG) techniques, this paper will extract static critical exponents for the dimension of physical interest ($d = 3$) n spite of uncertainties as to the validity of the ϵ-expansion in RF systems. The case for and against the ϵ-expansion in RF systems will be elaborated later in this paper.

Short-range RFIM and dipolar RFIM have such high upper critical dimensions (6 and 5 respectively) that no rigorous analytic derivations of static critical exponents have been performed at dimensions of physical interest ($d = 2, 3$) and also no experimental studies of behaviour close to d_c is possible. Monte Carlo simulations have been done of short-range RFIM at high dimensions close to d_c, i.e. at $d = 4, 5$, by Houghton *et al* [12]. Numerical results of exponents at three dimensions for the short-range RFIM were given by Ogielski [8b]. Here we give a physically realizable case of a random field model which at three dimensions is just one dimension below d_c. Capacitance response and ultrasonic measurements have been done on one such system, $Dy(As_xV_{1-x})O_4$, by Graham, Taylor and co-workers [13].

2. Hamiltonian

In the structural phase transitions this paper is investigating, the primary order parameter ϕ is a homogenous deformation of the crystal structure. The phase transition occurs through the 'softening' of a linear combination of elastic constants c_{ij} as $T \to T_c$, e.g.

$$\lim_{T\to T_c}(c_{11} - c_{12}) = 0\,.$$

The physical origins of this elastic softening is varied. They could be ferroelastic fluctuations, Jahn–Teller effects or electronic band structure.

Cowley [7] gives the required crystal symmetries and properties that will produce the pure part of our Hamiltonian. Below is a list of the lattice symmetries of the high-temperature phase of the *pure* parent crystal before the addition of impurities. This list is gleaned from Cowley [7].

(i) Tetragonal $4mm$, $\bar{4}2m$, 422, $4/mmm$, with irreducible representation B_1 or B_2 (e.g. $DyVO_4$).

(ii) Orthorhombic classes with irreducible representations B_1, B_2, B_3.

In addition there is a group of ferroelectric crystals which will map onto the Ising model and undergo a second-order transition. The reader is referred to Cowley [7].

The addition of impurities generates random strain fields, which when coupled to the order parameter ϕ creates the random field contribution to the Hamiltonian.

For crystals with the symmetry for $T > T_c$ given above, with the addition of impurities, the critical behaviour can be described by the Hamiltonian

$$H = -\int [a(T-T_c) + v\cos^2\varphi\sin^2\varphi\sin^2\theta + g\cos^2\theta + eq^2]\phi(q)\phi(-q) - \int h(-q)\phi(q) - u_0\int\int\int \phi(q_1)\phi(q_2)\phi(q_3)\phi(-q_1-q_2-q_3) \tag{1}$$

$$\cos\theta = \frac{q_z}{q} \tag{2}$$

$$\sin\theta\cos\varphi\sin\varphi = \frac{q_x q_y}{q q_\perp} \tag{3}$$

$$q^2 = q_z^2 + q_\perp^2 \tag{4}$$

$$\overline{h(q)} = 0 \tag{5}$$

$$\overline{h(q_1)h(q_2)} = \Delta\delta(q_1 + q_2) \tag{6}$$

where $\bar{x}$ means the ensemble average of x. The origin of the φ angle will depend on the representation and symmetry class. $\phi(q)$ is the order parameter, and $h(-q)$ is the random field. We are in momentum q-space.

Thus we have the Hamiltonian for a random field model with one major difference: in addition to short-range isotropic nearest-neighbour interactions we get angular terms that depend on θ and φ in momentum space. In real space such terms are long-range forces. For instance, $g\cos^2\theta$ is the dipolar interaction. Nattermann [14] has dealt with this dipolar contribution in his paper.

3. Upper critical dimension

We could either adopt the anisotropic scaling as presented by Birgeneau [15a] or the isotropic scaling which Aharony [15b] applied to the pure dipolar problem. Both are mutually and mathematically consistent. We adopt anisotropic scaling. Figures 1(a) and 1(b) illustrate the concepts behind anisotropic scaling. The crystals to which our work applies have either tetragonal or orthorhombic symmetry in the *pure* phase above T_c. Note how the mode softening in figures 1(a) and 1(b) reflects this symmetry. We have chosen without loss of generality for the mode softening to occur along [110] and [1$\bar{1}$0]. This anisotropy of domain shape has been observed by experimentalists [6]. Thus, anisotropic scaling appeals to physical intuition.

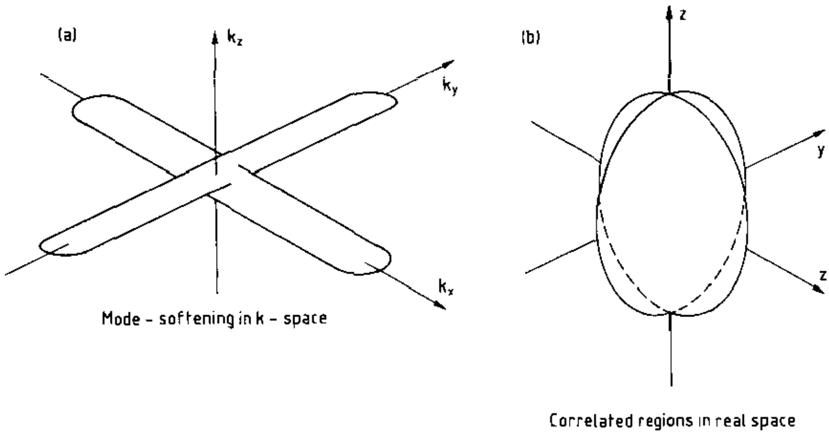

Figure 1. (a) Mode softening in muomentum space. (b) Correlated regions in real space.

Under anisotropic scaling, we require g and v to be constant.

$$g' = g \tag{7}$$

$$v' = v \,. \tag{8}$$

It is important to note that there are no graphical perturbation corrections to the above RG iterations for g and v. This is because g and v are coefficients of angular terms $(q_z/q)^2$, $q_x^2 q_y^2/[q^2(q_x^2 + q_y^2)]$. In any graphical term in the perturbation expansion, the outer shell of the Brillouin zone is integrated over as the momentum gets rescaled under RG iteration. In integrating over the outer shell, the angular terms will yield a value dominated by the magnitude of the external momentum vector, but independent of its direction. Thus, no graphs will contain any external angular terms and the RG iteration for angular terms do not contain graphs. The lack of graphical contribution is also found and explained in Aharony's [15b] treatment of the pure dipolar problem. In our notation $\boldsymbol{q} = (\boldsymbol{q}_1, q_x, q_y, q_z)$; $\boldsymbol{q}_1$ is a $(d-3)$-dimensional vector.

Looking at figure 1(a), we see that there is a choice of q_x or q_y in the anisotropic scaling. This reflects the fourfold symmetry about z. For non-zero v, a breaking of symmetry between the x and y directions is inevitable whichever choice of scaling is taken. Without loss of generality, we choose $\boldsymbol{q}_1$, q_x to scale as ξ^{-1}, q_y, q_z as

$\xi^{-\zeta}$, $\zeta > 1$:

$$q_1 = q_1' b^{-1} \tag{9}$$

$$q_x = q_x' b^{-1} \tag{10}$$

$$q_y = q_y' b^{-\zeta} \tag{11}$$

$$q_z = q_z' b^{-\zeta} . \tag{12}$$

To retain the fluctuation term $\int q^2 \phi^2 \mathrm{d}q$, we require e to be constant under scaling. Using the above scaling rules for q, $\mathrm{d}q$, e, the requirement that g, v be constant implies

$$\zeta = 2 - \frac{\eta}{2} . \tag{13}$$

The inclusion of η in the anisotropic mode softening means that thermal fluctuations, if relevant, will affect the shape and volume of the domains and mode softening. In our notation $\overline{x}$ means the ensemble average of x, $\langle x \rangle$ means the thermal average of x.

We now introduce random field scaling. We adopt the scaling rules of Bray [10]. Like Bray, we have three independent critical exponents, including a new exponent $\overline{\eta}$. $\overline{\eta}$ is a new independent exponent that takes into account random field (RF) fluctuations. Fisher [11] (1986) and Villain [20] have also used three independent exponents in their scaling approach to the random field problem. By contrast, pure problems have only two. Figures 2 and 3 illustrate the concepts of random field fixed point, universality class and scaling.

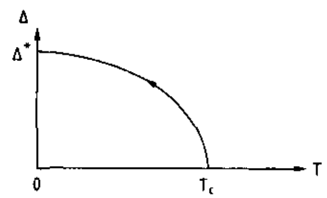

Figure 2. RG flow and phase diagram for random field models.

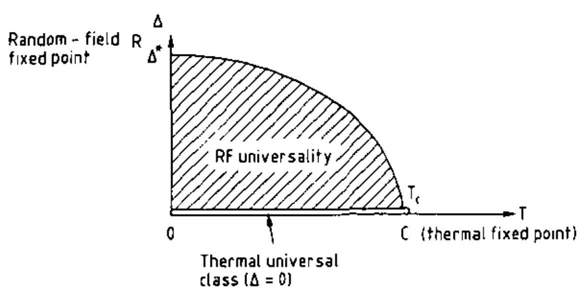

Figure 3. Random field and thermal universality classes.

Following Bray [10], we obtain the scaling relation for the mean square RF strength Δ:

$$\Delta = b^{2\eta-\overline{\eta}}\Delta'[1 + \text{graphs}] \tag{14}$$

$$\phi = c\phi' . \tag{15}$$

From all the scaling relations stated above and scale invariance,

$$c = b^{d/2+3-\eta/2-\overline{\eta}/2}c' \tag{16}$$

$$T = b^{2+\eta-\overline{\eta}}T' . \tag{17}$$

By comparison, $c = b^{d/2+1}$ for pure short-range systems, $c = b^{d/2+2-\overline{\eta}/2}$ for the short-range RFIM (cf Bray [10]).

T goes to 0 under scaling and we have a *zero temperature* random field fixed point (see figure 3). $1/T$ appears in the Boltzmann weight $\exp(H/T)$, so T is a *dangerously irrelevant* parameter. In our scaling formalism, we must always take into account $1/T$ and T, wherever they appear.

At the upper critical dimension d_c, all fluctuations are irrelevant, in which case $\eta = \overline{\eta} = 0$. For $d \geqslant d_c$, using the scaling rules for c and T,

$$\frac{u_0}{T}\int \phi^4 \mathrm{d}q = u_0\left[\frac{1}{T'}\int \phi'^4 \mathrm{d}q'\right] b^{4-d} . \tag{18}$$

We see that u_0 is irrelevant above $d = 4$. Hence the upper critical dimension

$$d_c = 4 . \tag{19}$$

Cowley [7] showed that anisotropy for *pure* systems reduces $d_c = 4$ to $d_c = 2$. Random fields increase $d_c = 4$ to $d_c = 6$ for the *short-range* RFIM [10]. Using scaling arguments, we have rigorously shown that for d_c, dimensional reduction strictly applies, i.e. $d_c = 4 + 2 - 2 = 4$.

4. Critical Exponents

In this section we proceed in three stages. First, we state the theoretical uncertainties concerning the applicability of the RG to RF models. Next, we proceed to perform an RG ϵ-expansion in spite of such uncertainties. Finally, we state the reasons for the plausibility and motivations of an RG calculation applied to our model at three dimensions.

According to Fisher [11] and Villain [20], random fields induce a large number of local minima, and there are problems assigning the correct Boltzmann weights to them. Conventional RG techniques assume only one minimum for the free energy. Fisher [11] conjectures that this problem of infinitely many minima will render the RG invalid even at high dimensions close to d_c.

In spite of this lack of certainty, we will proceed to set up RG equations for our model and perform an ϵ-expansion to $\mathrm{O}(\epsilon)$ at $\epsilon = 1$, where $d = 3$. We use three independent exponents, adopting the approach of Bray [10]. For pure systems, we need determine only two independent exponents. We now set up the RG perturbation expansions for the various parameters, using all the previous scaling relations listed in section 3. We define

$$r_0 \equiv a(T - T_c)\,. \tag{20}$$

The RG equations for our random field model are as follows:

$$r_0' = b^{2-\eta}[r_0 + $$  $$ + \cdots] \tag{21}$$

$$g' = g \tag{22}$$

$$v' = v \tag{23}$$

$$e' = b^{-\eta}[e - $$  $$\ln b + \cdots] \tag{24}$$

$$\Delta' = b^{\overline{\eta}-2\eta}[\Delta + $$  $$ + \cdots] \tag{25}$$

$$u_0' = b^{4-d-\overline{\eta}}[u_0 + $$  $$ + \cdots]\,. \tag{26}$$

Figures 4 and 5 illustrate the Feynman graphs involved in our RG calculations. We now perform a one-loop ϵ-expansion to $O(\epsilon)$. At the one-loop level

$$\Delta' = \Delta \tag{27}$$

$$e' = e \tag{28}$$

$$\overline{\eta} = 2\eta = 0\,. \tag{29}$$

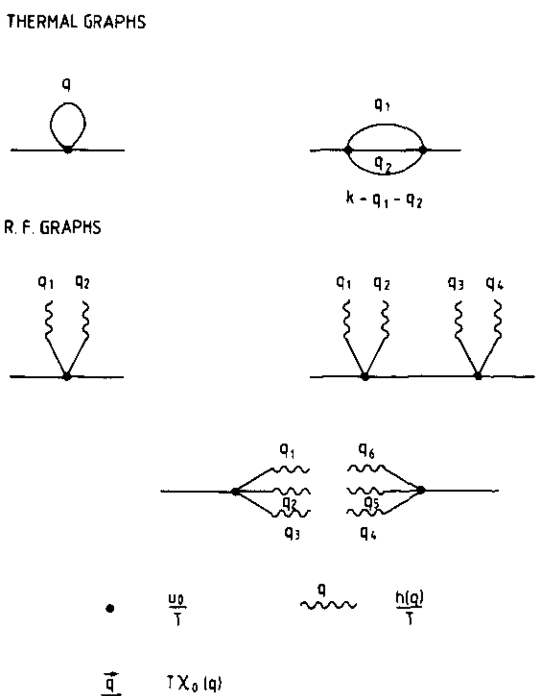

Figure 4. Thermal graphs and random field graphs. We have included T because $T \to 0$ and so $1/T$ is relevant.

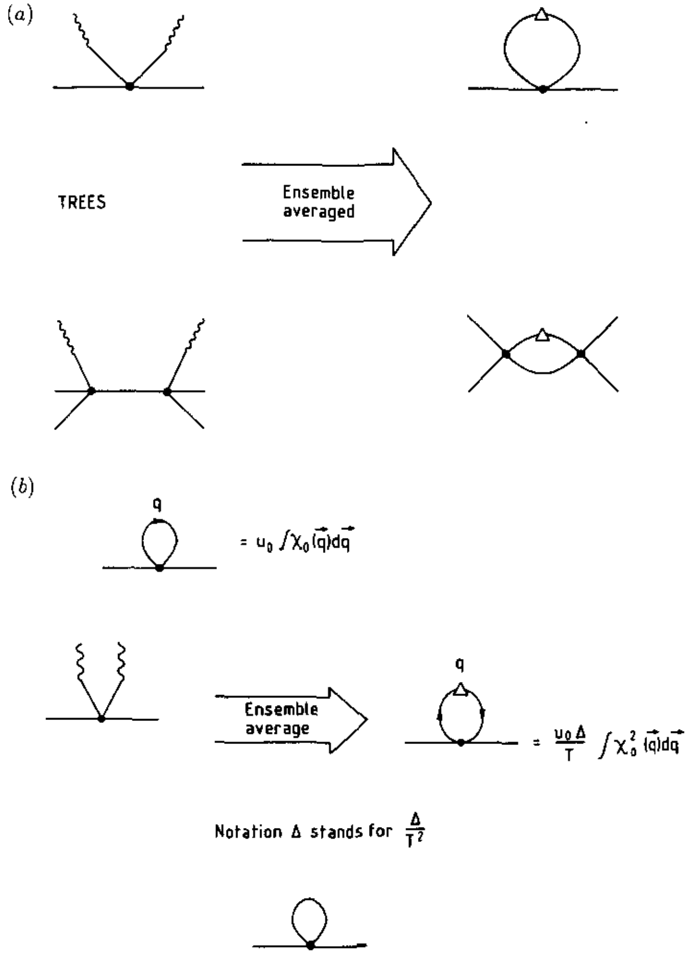

Figure 5. (*a*) Random field tree graphs are ensemble-averaged to form graphs with random field fluctuations. In this diagram Δ stands for Δ/T^2. (*b*) Mathematical expressions for one-loop thermal and random field ensemble-averaged graphs.

At three dimensions, i.e. one dimension below d_c, two-loop graphs are numerically smaller than one-loop graphs, so $\overline{\eta}, \eta$ should be very small at $d = 3$, and can be neglected without serious error. We linearize near the fixed point. At $3 = d_c - 1$ dimensions, we obtain

$$\nu = 0.6 \tag{30}$$

$$\gamma \approx 2\nu = 1.2\,. \tag{31}$$

We have predicted a value of 1.2 for γ, at the one-loop approximation. To O (ϵ), the critical exponents for our model are equal to those of the pure short-range Ising model, with both models at three dimensions. Thus we call our fixed point the quasi-Ising fixed point. Of course, by inspecting the graphs, it can be seen that to higher orders in ϵ, this equivalence of the two universality classes should break down, due to the presence of the extra exponent $\overline{\eta}$ which is non-zero at O (ϵ^2). For d well below

d_c, η, $\overline{\eta}$ can be very large, as testified by numerical work [8a, b]. For the short-range RFIM, Ogielski's numerical work [8b] predicts a value of 1.0 for ν.

We now explain why we consider the results of our ϵ-expansion plausible, even though not yet rigorously justified.

(i) Following Bray [10], our calculations account for RF fluctuations by including an extra independent exponent $\overline{\eta}$.

(ii) We are working close to the upper critical dimension. At three dimensions, only RF graphs will be relevant (see figures 5(a) and 5(b)). Aharony [9] and Villain [20] also advanced similar plausibility arguments.

(iii) There is numerical evidence in support of our conjecture that an ϵ-expansion for random field models would be 'safe' at high enough dimensions. Monte Carlo simulations of the short-range RFIM [12] at $d = 5$, which is one dimension below the upper critical dimension of 6, have given static exponents numerically similar to that predicted by RG techniques. It is easy to show that applying Bray's calculation [10] to the short-range RFIM will yield critical exponents equal to ours to $O(\epsilon)$.

(iv) For the short-range RFIM, at any dimension below d_c, the static critical exponents must satisfy certain inequalities [21]. These inequalities must also be satisfied by our model, with an important modification: the dimension d is replaced by $(d+2-\eta)$, because the short-range RFIM has a correlated domain volume of ξ^d whereas ours has a domain volume of $\xi^{d+2-\eta}$. Villain [20] has also developed a group of inequalities to bound the critical exponents with. The set of inequalities [21], in our notation, is as follows:

$$\gamma \geqslant 0 \tag{32}$$

$$\frac{\beta}{\nu} \geqslant \overline{\eta} - \eta - 2 + \frac{d+2-\eta}{d} \qquad \text{(Schwartz–Soffer inequality)} \tag{33}$$

$$1 \geqslant \eta - \overline{\eta} - d - 2 + \eta\,. \tag{34}$$

For our model, at three dimensions, in the one-loop approximation, $\eta = \overline{\eta} = 0$, $\gamma = 1.2$, $\nu = 0.6$, $\beta = 0.3$. We can see that with these values the above inequalities are indeed satisfied.

We compare our results to those obtained experimentally by Graham, Taylor and co-workers [13]. They studied the mixed Jahn–Teller system $Dy(As_xV_{1-x})O_4$. We treat $DyVO_4$ as the pure parent and As as the impurity. Then according to Cowley [7], $DyVO_4$, with symmetry $4/mmm$, B_1 mode and acoustic modes along [110] or [1$\overline{1}$0], will have mode softening given by

$$\frac{1}{2}(c_{11}-c_{12}) + \left(c_{44} - \frac{1}{2}c_{11} + \frac{1}{2}c_{12}\right)\left[\left(\frac{q_z}{q}\right)^2 + \frac{c_{11}+c_{12}}{c_{12}+c_{44}}\sin^2\theta\sin^2\varphi\cos^2\varphi\right].$$

Here the azimuthal angle φ is defined as zero along the [110] direction. This, with As supplying the random field, will give the required Hamiltonian. Graham *et al* [13] measured the γ to be 1.25 crossing over to 1.6 as $(T - T_c)$ tends to zero. In a later work [13] (1991) they measured $\gamma = 1.79 \pm 0.07$. This is much higher than our predicted value, which they cite. Graham *et al* [13] (1991) also measured the pure parent crystal $DyVO_4$. They found γ to have a non-critical value of 1.15, higher than the mean-field value of 1. In their paper [13] (1991), they state that $DyVO_4$ is not a good mean-field system. Cowley [7] predicted for $DyVO_4$ that the long-range forces would be strong enough to induce a crossover to the mean-field regime, with an upper critical dimension of 2. A crucial assumption underlying Cowley's theoretical treatment [7] and our work (which is derived from Cowley [7]) is that the long-range forces are strong enough to induce a crossover that can be experimentally observed. Perhaps $Dy(As_xV_{1-x})O_4$ is not a good candidate for our theoretical model, even though it fits our symmetry requirements.

We do not assert that our predicted values of critical exponents are correct, or that those of Graham *et al* [13] are wrong. The motivation for stating the results of our ϵ-expansion at three dimensions, in spite of its lack of rigorous theoretical justification, is as follows. Ours is a random field model with an upper critical dimension of 4. At the experimentally interesting dimension of 3, it is easy to perform an ϵ-expansion. The short-range RFIM and the dipolar RFIM of Nattermann [6] have upper critical dimensions of 6 and 5 respectively, which makes an ϵ-expansion at three dimensions problematic. We wish to see if experiments and computer simulations can verify or contradict our RG predictions. If careful computer simulations and experiments can confirm our results, then the validity of an ϵ-expansion for a random field model will be made even more plausible. If they prove otherwise, then the RG will be conclusively shown to be invalid for random field models. Either way, a definite advance in our understanding of random field systems will have been made.

5. Crossover behaviour

The short-range RFIM has its crossover behaviour described by two fixed points—pure Ising and short-range RFIM. The crossover behaviour of our random field model involves more fixed points and is correspondingly more complicated. The crossover behaviour will consist of competition amongst the relative magnitudes of g, v and the mean-square random field strength. g and v are determined by elastic constants of the pure system [7]. The random field strength will be determined by the concentration of impurities and the strain coupled to the order parameter. For $T < T_c$, when the random field strength and g and v are negligible compared to $(T_c - T)$, mean-field behaviour will dominate because both thermal and RF fluctuations are negligible. If the mean-square random field strength is much weaker than g and v, g and v will suppress thermal fluctuations and mean-field theory will still hold up to temperatures close to T_c. Very close to T_c, however, random fields will induce a crossover to the quasi-Ising fixed point, where the static exponents will be very close to those of the three-dimensional short-range pure Ising model.

If the mean-square random field strength is much larger than g and v, as the temperature approaches T_c, there will first be a crossover to 3D short-range RFIM behaviour. When $(T_c - T)$ is small enough so that the effects of the anisotropy terms g and v are being felt, there will be competition between g and v. Eventually, the static exponents close to those of the 3D pure short-range Ising model should be measured. The following elaboration should make this clear.

Any thermodynamic quantity W can be written as

$$W = t^{\omega}\Omega\left(\frac{g}{t^{\phi}}, \frac{v}{t^{\psi}}\right) \tag{35}$$

where ω is the short-range 3D RFIM exponent of W, and $t = T - T_c$. ϕ, ψ are the crossover exponents of g, v respectively. When $t \leqslant g^{1/\phi}$ we get a crossover to a new universality class involving g. When $t \leqslant v^{1/\psi}$ we get a crossover to a new universality class involving v. Section 3 shows that g, v both scale like $\xi^{2-\eta}$ under *isotropic* scaling. Thus we can also write

$$W = t^{\omega}\Omega(\xi^{2-\eta}g, \xi^{2-\eta}v) . \tag{36}$$

In momentum space, the thermodynamic quantity will have the scaling form

$$\begin{aligned} C(q_1) = q_1^{\omega/\nu} F\left(q_1\xi, q_x\xi, \frac{q_z^2}{q_1^{4-\eta}}, \frac{q_y^2}{q_1^{4-\eta}}, g\left(\frac{q_1}{q_z}\right)^2, v\left(\frac{q_1}{q_y}\right)^2\right) \\ + q_1^{\omega/\nu} F\left(q_1\xi, q_y\xi, \frac{q_z^2}{q_1^{4-\eta}}, \frac{q_x^2}{q_1^{4-\eta}}, g\left(\frac{q_1}{q_z}\right)^2, v\left(\frac{q_1}{q_x}\right)^2\right) . \end{aligned} \tag{37}$$

$C(q_1)$ reflects the invariance under the interchange of q_x and q_y, given the tetragonal symmetry between the x and y directions (see figures 1(a) and 1(b)).

There are three possible cases:

(i) $g \sim v$: there is a direct crossover from the short-range three-dimensional RFIM to the quasi-Ising fixed point, i.e.

$$\phi = \psi = (2 - \eta)\nu_s \tag{38}$$

where ν_s is the critical exponent for the correlation length ξ for the three-dimensional short-range RFIM.

(ii) $g \gg v$: the effects of g are felt first, before v. There is first an intermediate crossover to the 3D RFIM with dipolar interactions with the upper critical dimension of 5. This model has been studied by Nattermann [14]. In this case

$$\phi = (2 - \eta)\nu_s . \tag{39}$$

Then there is the crossover to the quasi-Ising fixed point where v at last becomes noticeable. In this case

$$\psi = (2 - \eta_d)\nu_d . \tag{40}$$

η_d, ν_d are the critical exponents of the dipolar RFIM. ν_d is the critical exponent of the correlation length in particular. Now $\phi \neq \psi$!

(iii) $v \gg g$: similiar to case (ii), except that g and v interchange.

Monte Carlo simulations of the three-dimensional dilute antiferromagnet in a uniform field [8], which maps on to the three-dimensional short-range RFIM, have found

$$\begin{aligned} &\eta = 0.5 \pm 0.1 \qquad \nu_s = 1.3 \pm 0.3 \\ &\Rightarrow (2 - \eta)\nu_s = 1.95 \pm 0.30 . \end{aligned}$$

Numerical results by Ogielski [8a] have found for the three-dimensional short-range RFIM

$$\overline{\eta} = -0.9 \qquad \nu_s = 1.0 \qquad \beta = 0.05 .$$

We have found no available data for the dipolar RFIM exponents.

Using the above Monte Carlo and numerical values, we can determine the crossover exponents for case (i).

For case (i), the crossover exponents $\phi = \psi = 1.95 \pm 0.30$. For case (ii), we can only determine one exponent $\phi = 1.95 \pm 0.30$. ψ depends on dipolar exponents which we have not been able to find, so ψ is undecided for case (ii). For case (iii), since g and v interchange, ϕ and ψ correspondingly interchange and so $\psi = 1.95 \pm 0.30$ and ϕ is undecided.

For the crossover described above to be observable, the elastic forces represented by g, v, must be strong compared to the random field represented by Δ.

6. Dynamics

In this section, we focus on the dynamic properties of structural phase transitions with random strains. As this paper will show, there is an intimate connection amongst the domain wall roughness, upper critical roughening dimension, slowing down time and relaxation mechanism of our model. Random field models have long posed theoretical difficulties with their dynamic properties [1–5]. A major difference between random field (RF) models and pure models lies in the dynamical behaviour. RF models experience much longer critical slowing down times than the corresponding pure models. This is due to the presence of metastable domains in RF models, absent in pure systems. Several authors [1–5] have postulated that random fields roughen the domain walls and pin them, hence slowing down drastically the growth and shrinkages of domains.

Nattermann [6] investigated the role of long-range dipolar forces in random field systems and their effects on the growth of non-equilibrium metastable domains. The interfacial roughness exponent of the domain walls is altered. Similar to Nattermann [6], we will show that long-range forces reduce the upper critical roughening dimension. Beyond that, we will also investigate the shape of our domains caused by the anisotropy of our forces, and will find that shape effects have an important role in the dynamics of our model.

6.1. Interfacial roughening

For a random field model with domains, whether the domain walls are smooth or rough depends on the competition between the domain wall 'surface tension' and the energy to be gained from random fields [2, 20]. Random fields encourage roughening of domain walls [20]. It has been postulated that the roughness of domain walls are responsible for the very long relaxation time involved in the critical slowing down [1–5]. The upper critical roughening dimension d_u is defined as follows. For $d > d_u$, the wall is smooth on the length scale of the model, i.e. the roughness divided by the length scale of the model goes to zero as this length scale tends to infinity. For $d < d_u$, a roughening transition of domain walls occurs at some temperature $T_R < T_c$. T_R is defined as the roughening temperature.

To obtain d_u we perform a calculation similar to Nattermann [6], to which the reader is referred for the details. However, we will subsequently show that d_u is not sufficient to determine whether the domain walls of our model are rough or smooth. To determine this requires an understanding of anisotropic shape effects, as we will show.

6.2. Upper critical roughening dimension

The bulk Hamiltonian in real space is

$$H = \int \mathrm{d}\boldsymbol{r} \int \mathrm{d}\boldsymbol{r}' S(\boldsymbol{r}) D(\boldsymbol{r} - \boldsymbol{r}') S(\boldsymbol{r}') + \int \mathrm{d}\boldsymbol{r}\, h(\boldsymbol{r}) S(\boldsymbol{r}) . \qquad (41)$$

$S(\boldsymbol{r})$ is the spin, $h(\boldsymbol{r})$ the random field, and $D(\boldsymbol{r} - \boldsymbol{r}')$ the long- and short-range forces.

We divide the space into the single direction perpendicular to the wall z, and the subspace parallel to the wall ρ:

$$\boldsymbol{r} = (\boldsymbol{\rho}, z) \qquad \boldsymbol{r}' = (\boldsymbol{\rho}', z') . \qquad (42)$$

Like Nattermann [6], we define a fluctuation f, and assume $f = f(\rho)$ to vary in the z-direction perpendicular to the wall, and assume an interface of zero curvature.

$$S(\boldsymbol{r}) = S(z - f(\boldsymbol{\rho})) \qquad (43)$$

$$z > f(\rho) \Rightarrow S = 1 \qquad z < f(\rho) \Rightarrow S = -1 . \qquad (44)$$

$S(\boldsymbol{r})$ behaves like a step function with a boundary $f(\boldsymbol{\rho})$. We make the assumption that with no roughening, i.e. $f \equiv 0$, we have an interface of zero curvature. Figure 1 makes this clearer.

We examine the random field term in (41). It is extremely difficult to derive an exact expression for the random field term in terms of the roughness f, so we resort to an approximation. We define a *random rod field* as a random field that fluctuates along the domain wall, but does not change in the direction perpendicular to the wall. In the *random rod field* case, since the field is uniform perpendicular to the wall, it is energetically more advantageous for the wall to deviate than for the *random field* case. Thus, if it can be shown that the interface is smooth in the presence of *random rod fields*, it can be concluded that the interface is smooth in the presence of *random fields*.

We define the mean-squared interfacial fluctuation, averaged over random fields and thermally, to be $\overline{\langle f^2 \rangle}$. Following Nattermann [6] and using the random rod field approximation, the following inequality is obtained:

$$\frac{\overline{\langle f^2 \rangle^{1/2}}}{L} \leqslant \log L$$

where L is the linear dimension of the wall.

At three dimensions, we can expect the roughness of the domain walls to diverge at most logarithmically with the length scale of the system. We thus arrive at the main result of this subsection, $d_{\mathrm{u}} \leqslant 3$, for structural phase transitions with random strains.

6.3. Anisotropic shape effects

In the previous subsection, we showed that the upper critical roughening dimension $d_{\mathrm{u}} \leqslant 3$. In this subsection, we will show that the upper critical roughening dimension alone is not enough to determine that the domain walls will smooth, even at three dimensions greater than or equal to d_{u}. We now investigate the role of the shapes

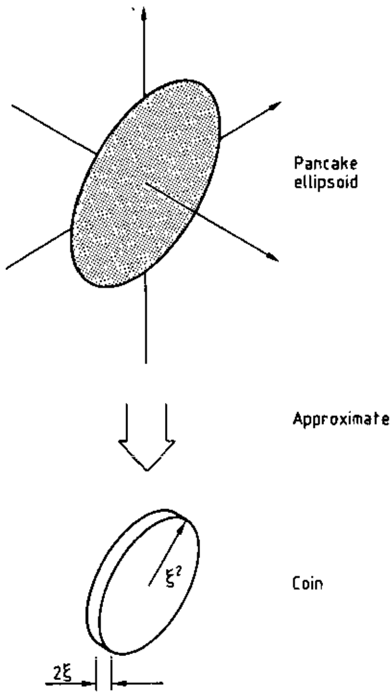

Figure 6. Flat interface approximation of domain wall. The ellipsoidal domain is approximated by a disc. The face of the disc is assumed flat.

of domains. The anisotropic nature of our long-range forces cause the domains to have an ellipsoidal shape before roughening (see figures 1 and 6). The previous approximation, where a flat interface was assumed and the edge of the domain neglected, works well for extremely large domains (see figure 6). In the regime of small domains, the edge can no longer be neglected and curvature effects have to be taken into account. We will show that this small domain regime is intimately tied up with the dynamics and metastability of the system, indeed is crucial in determining the wall roughness and hence the metastability and dynamics.

We divide the domain wall into two aspects—the 'broad' and the 'narrow' (see figure 7). We make the approximation that the broad aspect has a single radius of curvature R_1 (see figure 8), and that the narrow aspect has two radii of curvature—R_2 and R_2^2 (see figure 8).

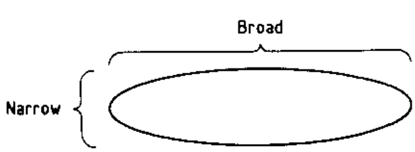

Figure 7. Side cross section of domain.

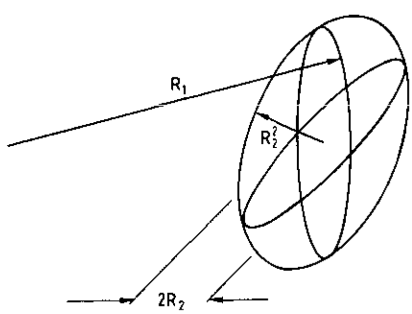

Figure 8. Domain with cross sections and radii of curvature.

When a random field system is quickly quenched from above T_c to low temperatures, domains of various length scales L form. Some will shrink to nothing, others may take a much longer (possibly infinite) time to shrink. In the latter case the domain is said to be metastable. The rate at which the domains shrink is determined by the energetics of the domain walls. Energy considerations cause the domain wall to roughen, creating bumps $w(L) \equiv \sqrt{\langle f^2\rangle(L)}$ of various length scales L. A bump $w(L)$ is created to take advantage of a local region where a majority of the random fields are aligned with the order parameter. The random field energy gained from such a bump is $[\Delta w(L)A(L)]^{1/2}$; statistically it scales as the square root of the volume $w(L)A(L)$ created by the bump. $A(L)$ is defined as the $(d-1)$-dimensional area over which the bump occurs. For a description of how domains form bumps to minimize their energy in random fields, see [2, 20].

These bumps create an energy barrier $E[w(L), L]$ (see figures 10(*a*) and 10(*b*)). The very presence of these bumps $w(L)$ is responsible for the metastable energy barriers $E[w(L), L]$ which in turn is responsible for the very long relaxation times of random field systems. If $w(L) = 0$ for all length scales L, then $E \equiv 0$ and there would be no energy barriers. When there are no bumps $w(L)$ on any length scale L, there is no longer any random field energy $[\Delta w(L)A(L)]^{1/2}$ to be gained, and then random fields become irrelevant, and the dynamics is essentially that of the pure system.

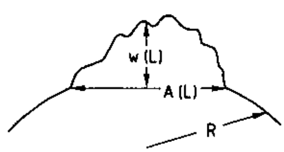

Figure 9. Bump on domain wall with radius of curvature R.

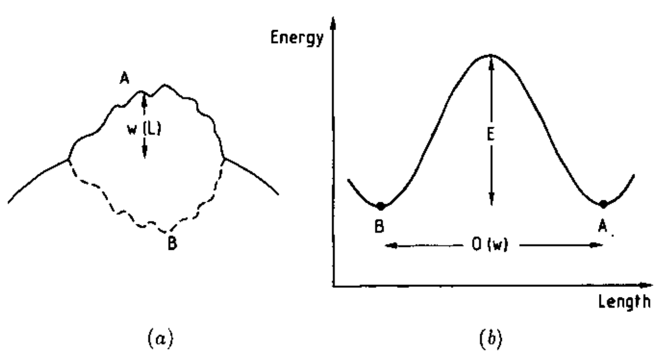

Figure 10. (*a*) Two metastable bump configurations, A and B. (*b*) Energy barrier separating bump configurations A and B.

Of the various treatments of the dynamics of the random field problem [1–5], the most complete are those of Andelman and Joanny [2], and Nattermann and Vilfan [3]. Both are essentially equivalent. We choose to adopt the approach of the former.

We have the equation of the energy barrier [2]

$$E[w(L), L] = -\sigma A(L)\left[\left(\frac{w}{L}\right)^2 + \frac{w}{R}\right] + [\Delta A(L)w]^{1/2} \qquad (46)$$

σ is the surface tension, $A(L)$ the $(d-1)$-dimensional area over which the bump occurs, L the length scale, and R the radius of curvature. $L \leqslant R$. The first term on the right-hand side of this equation is the interfacial energy for the increase in domain wall surface area due to the bump w. The second term is the Laplace contribution for a curved interface of radius R. The third term is the random field energy term.

There is a minimal radius of curvature R_c such that for domains with $R < R_c$, $w(L) = 0$ for all $L \leqslant R$. In that case $E = 0$, and surface tension causes the domain to collapse. R_c depends on the random field strength and is independent of temperature. This is the dynamical picture presented by Bruinsma and Aeppli [1].

At higher temperatures but below T_c, the thermal energy $\sim O(k_B T)$ enables the domain to overcome the metastable barriers $E[w(L), L]$ whenever $E < k_B T$. Thus the domain shrinks by thermal hopping even if the surface tension is insufficient to overcome energy barriers. R_T is the 'thermal' minimal radius which depends on temperature and grows logarithmically:

$$R_T(t) = R_0 \ln \frac{t}{\tau}. \qquad (47)$$

This is the dynamical picture presented by Villain [4].

Andelman and Joanny [2] incorporated the two effects, and defined a minimal radius R_{min}

$$R_{min}(t) = \max[R_c, R_T(t)]. \qquad (48)$$

At any time t, all domains have radii of curvature that are no smaller than the minimal radius $R_{min}(t)$. We have two radii—a 'surface tension' radius R_c and a thermal radius $R_T(t)$. We proceed to calculate R_c, R_T for our system. Since we are seeking to calculate minimal radii, we assume that R is small; $w/R \gg (w/L)^2$. We will study the two aspects—'broad' and 'narrow'—of the domain walls separately, beginning with the broad (see figure 7). Andelman and Joanny [2] were dealing with an isotropic system; as such for their work the area $A(L) = L^{d-1}$. We are dealing with a system that scales anisotropically. For the broad aspect of our domain wall, $A_{broad}(L) = L^{2(d-1)} = L^4$ for $d = 3$, and $R = R_1$.

We calculate the 'surface tension' radius R_{1c} for the broad aspect, following the method presented in Andelman and Joanny [2]. We found that $R_{1c} = \infty$ at $d = 3 \leqslant d_u$. Hence we have shown that even taking curvature effects into account, the broad aspect is smooth and will not contribute to the metastability at three dimensions.

We now examine the narrow aspect of the domain wall. The anisotropic scaling in this part of the domain wall is not unlike the dipolar problem (cf [6]). The narrow aspect involves a short radius R_2 and a long radius R_2^2; the dipolar RFIM domain is an ellipsoid with a short principal radius R and a long principal $R^{3/2}$ [6]. In both the narrow aspect of our problem and the dipolar RFIM, there are 2 different length scales involved namely L and L^2; $A(L) = A_{narrow}(L) = L^2 L = L^3$. To take into account the different length scales and radii, we have to modify the methods of Andelman and Joanny [2]. One must keep in mind that Andelman and Joanny [2] applied their calculations to an isotropic system, and we are calculating an anisotropic system.

To find R_{2c}, we go through the same method as for the broad aspect, except that the radii and length scales are altered. We found for the 'surface tension' radius of the narrow aspect

$$R_{2c} = \frac{1}{2}\left[a_l^5\left(\frac{(2\sigma)^4}{\Delta^2}\right) + \sqrt{a_l^{10}\left(\frac{(2\sigma)^8}{\Delta^4}\right) + 4a_l^5\left(\frac{(2g)^4}{\Delta^2}\right)}\right]. \tag{49}$$

Thus we do find metastability for the narrow aspect even though there is none for the broad aspect. As such it is crucial not to neglect the narrow aspect in studying the metastability and dynamics of our model: it is the minimal radius of the narrow aspect R_{2c} that determines the minimal volume of the domain. Inspecting (46), we see that the smallest radius R gives the largest energy E.

The minimal volume of the domain is $V_c \sim R_{2c}^5$. By comparison, for a three-dimensional short-range RFIM, Andelman and Joanny [2] predicted the minimal radius $R_c = a_l^2[(2\sigma)^2/\Delta]$ and the minimal volume $V_c = R_c^3$. For our model, even though anisotropy does not remove metastability, the minimal volume of the domains is increased.

We now analyse the thermal minimal radius $R_{2T}(t)$ for the narrow aspect. The thermal minimal radius $R_{2T}(t)$ grows logarithmically in time: $R_{2T}(t) = R_0 \ln(t/\tau)$. We proceed to calculate R_0. We maximize $E[w, L]$ by differentiating E with respect to w:

$$\frac{\partial E}{\partial w} = 0\,.$$

The maximum energy obtained is $E_m = \Delta R_0^2/[4\sigma(R+1)]$. For small domains, $E_m[w, L] < k_B T$ for all length scales L, and thermal fluctuations will overcome the energy barriers to collapse the domain. We define the minimal thermal radius coefficient R_0 to be such that

$$E_m = \frac{\Delta R_0^2}{4\sigma(R_0+1)} = k_B T \tag{50}$$

from which we obtain

$$R_0(T, \Delta) = \frac{2\sigma k_B T}{\Delta} + 2\sqrt{\left(\frac{\sigma k_B T}{\Delta}\right)^2 + \frac{\sigma k_B T}{\Delta}}\,. \tag{51}$$

Note that R_0 depends on temperature and random field strength. By comparison, $R_0 = 4\sigma k_B T/\Delta$ for the short-range RFIM [2]. For our model, the thermal volume $V_T(t) \sim R_T^5(t)$, whereas for the short-range RFIM, $V(t) \sim R_T^3(t)$. Not only is the thermal minimal volume enlarged, but the rate of growth, although still logarithmic, has its power increased by 2.

To sum up the dynamical picture, anisotropic mode softening does not eliminate metastability or logarithmic relaxation times, but increases the minimal volume of the metastable domains and their growth rate. This is owing to the anisotropic domain shape. For short-range RFIM, the volume grows in time as $(\ln t)^3$, but for our model it grows as $(\ln t)^5$. In three dimensions, the broad aspect of the domain wall is always smooth, but the narrow aspect is locally rough, and it is the roughness of the narrow aspect that determines the metastable and dynamical behaviour of our model.

7. Conclusion

We have studied a random field model with long-range anisotropic forces. For the corresponding pure model, the effects of these long-range forces have been studied [7]. The short-range RFIM, the dipolar RFIM and our random field model represent three distinct random field models, each with its distinct universality class, fixed points, upper critical dimensions and crossover behaviour.

Upper critical dimension d_c. Short-range RFIM = 6; dipolar RFIM = 5; our random strain model = 4; pure short-range Ising model = 4.

Upper critical roughening dimension d_u. Short-range RFIM = 5; dipolar RFIM = 4; our random strain model = 3; pure short-range Ising model = 3.

Crossover behaviour. Our model exhibited the most complex crossover behaviour of the three random field models, since it incorporates the most additional long-range forces. The fixed points encountered in the crossover behaviour of our model include the short-range RFIM and dipolar RFIM fixed points.

Domain wall roughness. In isotropic models, if the bulk dimension is less than or equal to the upper critical roughening dimension, the domain wall will be smooth on the length scale of the domain size. In our anisotropic model, this is not the case. Predicting the upper critical roughening dimension does not guarantee smoothness in the domain walls of anisotropic models. We show how the domain wall is still rough in the narrow aspect even at three dimensions which is the upper critical roughening dimension of our model. The persistence of domain wall roughness in our model accounts for the persistence of metastable effects and long slowing down times in our random field model. The fact that our random field model and the short-range random field model exhibit the same qualitative dynamical behaviour should not obscure the different physics underlying the two models. The short-range RFIM is an isotropic system; if the dimension is high enough the domain walls will be smooth, likewise for the pure Ising model. However, ours is an anisotropic system; dimensionality alone is not enough to guarantee smoothness—anisotropic shape effects must be accounted for. Although anisotropic long-range forces do not eliminate metastability and long slowing down times in the dynamics of our model, one minor effect is to increase the minimum metastable domain size and the power of the logarithmic slowing down rate.

Critical exponents. We come to the one uncertain result of this work. In spite of the lack of rigorous theoretical justification, we have applied the renormalization group to extract static critical exponents from our system at three dimensions. From our ϵ-expansion, we have obtained critical exponents which to $O(\epsilon)$, are the same as for a pure Ising model, with both models at three dimensions. The pure 3D Ising model has $\gamma = 1.25$, and we predict for our model $\gamma = 1.2$. We state these results of our RG calculations in spite of the theoretical uncertainty still surrounding the validity of the RG for random field models. We do so because we regard these results as plausible, and also because ours is a random field model on which experimentalists can test the validity of the RG at three dimensions. This is not so for the short-range RFIM or the dipolar RFIM of Nattermann [6]. We encourage experimentalists and computer hacks to come up with accurate results that will either confirm or contradict our results. Either way, new light would have been shed on random field models.

To recap the question in the introduction, is our model more like a random field model or a pure model? We may answer that it is more like a random field model, although the truth is more subtle and less straightforward.

Ours is a random field model which at three dimensions is just one dimension below the upper critical dimension, whereas most other random field models, at three dimensions, are well below their upper critical dimensions. Hence ours is a random field model on which experiments can be done to study the behaviour of random field models close to the upper critical dimension. Since this model has experimental applicability, we have included in our paper details of experimental interest.

Using Cowley [7] as a guide, one physical example of our model is $Dy(As_xV_{1-x})O_4$, on which Graham, Taylor and co-workers [13] performed experiments. They measured the critical exponent γ to be $1.6 \sim 1.8$, which is much higher than 1.2 predicted by us. We offer no rigorous proof that our value is the correct one, and no rigorous explanation for the discrepancy. One possible explanation is that perhaps in $Dy(As_xV_{1-x}O_4)$, the random fields are much stronger than the elastic forces, so the crossover to the pseudo-Ising fixed point is not discernible. Graham *et al* [13] (1991) stated that the pure parent crystal $DyVO_4$ is not a good mean-field system. If that is so, the predictions of Cowley [7] and our work would not be applicable to this substance and its impure counterpart. We advise experimentalists to find a crystal whose elastic forces is strong enough to induce a crossover to the pseudo-Ising fixed point even in the presence of random fields. Experiments can be performed on physical examples of our model to measure domain wall roughness, and to analyse the dynamical behaviour. If possible it is hoped that experiments can correlate domain wall smoothness/roughness with the dynamical behaviour. One such experiment has been performed by Taylor *et al* [13] on $Dy(As_xV_{1-x})O_4$, a crystal which is a physical realization of the random field model studied in this paper. They observed the existence of metastable domains.

To our knowledge, ours is the first theoretical treatment of structural phase transitions of mixed crystals that goes beyond mean-field theory. Mayer and Cowley [22] used mean-field theory to analyse and predict the critical behaviour of $(KCN)_x(KBr)_{1-x}$, which by symmetrical considerations does not correspond to our model. Kasten et al [23] used a modified mean-field theory to describe $(Tm_xTb_{1-x})AsO_4$ and $(Tm_xTb_{1-x})VO_4$. Gehring [24] used molecular field theory to describe $(Tm_xLu_{1-x})VO_4$. The Hamiltonians describing these substances are not the same as our Hamiltonian, although they have a random field coupling to the order parameter. Both Kasten *et al* [23] and Gehring *et al* [24] were able to account for the experimental data of the specific heat fairly well with their mean-field analysis.

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References

[1] Bruinsma R and Aeppli G 1984 *Phys. Rev. Lett.* **52** 1547

[2] Andelman D and Joanny J 1985 *Phys. Rev.* B **32** 4818

[3] Nattermann T and Vilfan I 1988 *Phys. Rev. Lett.* **61** 223

[4] Villain J 1984 *Phys. Rev. Lett.* **52** 1543

[5] Grinstein G and Fernandez J 1984 *Phys. Rev.* B **29** 6389

[6] Nattermann T 1983 *J. Phys. C: Solid State Phys.* **16** 4125

[7] Cowley R 1976 *Phys. Rev.* B **13** 4877

[8a] Ogielski A and Huse D 1986 *Phys. Rev. Lett.* **56** 1298

[8b] Ogielski A 1986 *Phys. Rev. Lett.* **57** 1251

[9] Aharony A 1978 *Phys. Rev.* B **18** 3318

[10] Bray A 1986 *J. Phys. C: Solid State Phys.* **19** 6225
Bray A and Moore M 1985 *J. Phys. C: Solid State Phys.* **18** L927

[11] Fisher D S 1985 *Phys. Rev.* B **31** 7233; 1986 *Phys. Rev. Lett.* **56** 416

[12] Houghton A, Khurana A and Seco F 1986 *Phys. Rev.* B **34** 1700

[13] Graham J T, Maliepaard, Page J H, Smith S R P and Taylor D R 1987 *Phys. Rev.* B **35** 2098
Taylor D R, Graham J T, Matthews T P, Noakes D R and Buyers W J L 1988 *Int. Conf. Magnetism (Paris)* p 1239
Graham J T, Page J H and Taylor D R 1991 *Phys. Rev.* B **44** 4127

[14] Nattermann T 1988 *J. Phys. A: Math. Gen.* **21** L645

[15a] Als-Nielsen J and Birgeneau R 1977 *Am. J. Mod. Phys.* **45** 554

[15b] Aharony A 1973 *Phys. Rev.* B **8** 3363

[16] Berker N 1984 *Phys. Rev.* B **29** 5243

[17] Imry Y and Ma S 1975 *Phys. Rev. Lett.* **35** 1399

[18] Schwartz M 1988 *J. Phys. C: Solid State Phys.* **21** 753

[19] Parisi G and Sourlas N 1979 *Phys. Rev. Lett.* **43** 744

[20] Villain J 1985 *Scaling Phenomena in Disordered Systems* ed R Pynn and A Skjeltorp (New York: Plenum); 1985 *J. Physique* **46** 1843

[21] Nattermann T 1989 *7th Int. Meeting on Ferroelectricity (Saarbrücken)*

[22] Mayer A and Cowley R 1988 *J. Phys. C: Solid State Phys.* **21** 4835

[23] Kasten A, Kahle H G, Klöfer P and Shäfer-Siebert D 1987 *Phys. Status Solidi* b **144** 423

[24] Gehring G, Swithenby S and Wells M 1976 *Solid State Commun.* **18** 423