Equilibration of random-field Ising systems

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The equilibration of Ising systems in random magnetic fields at low temperatures (T) following a quench from high T is studied in the framework of a simple solid-on-solid model. The rate at which ordered domains in the model grow with time in any dimension (d) is estimated as a function of the random-field strength, the exchange strength, and T, on the basis of an approximate analogy to the problem of a one-dimensional random walk in a random medium. Domains are argued to grow logarithmically with time for all d. This result has a simple interpretation in terms of energy barriers which must be surmounted in the equilibration process.

Despite intense theoretical study over almost a decade, the behavior of the Ising model in a random magnetic field (RFIM) remains imperfectly understood.¹ There is now widespread acceptance of the simple physical arguments which suggest that the lower critical dimension d_c (the dimension below which no long-range-ordered state can occur in the RFIM) is 2. That acceptance remains cautious, however, given the lack of a definitive refutation of the field theoretic analyses predicting $d_c = 3$, and ambiguities in the interpretation of experiments on random antiferromagnets in uniform magnetic fields-the most convenient physical realizations² of the RFIM. The source of the experimental ambiguities, namely, long equilibration times and history-dependent behavior, are striking in and of themselves.³ A skeletal outline of the salient experimental facts for three-dimensional (3D) samples follows.

When cooled in a nonzero random (i.e., applied) field the random antiferromagnets typically do not exhibit long-range order down to the lowest temperatures studied. This is true even for comparatively modest fields, though for small enough fields the short-range order can extend out to very long distances⁴-thousands of angstroms-before it dies off. On the other hand, the antiferromagnetic long-range order established by cooling in zero field typically persists under the application of even quite large fields. Thus at a given point in the random field-temperature plane the system can either exhibit long-range order or not, depending on how that point has been reached. It is not at all clear which (if either) of these two cooling procedures produces the system's true equilibrium state upon which so much theoretical attention has been lavished. Depending on one's prejudice one can argue that static equilibium is achieved either by finite or zero-field cooling⁵ and hence that d_c is, respectively, 3 or 2.

One approach to this dilemma is to adopt one of these competing explanations and examine it more critically. One might ask, say, whether the simple arguments predicting $d_c = 2$ (and hence a long-range-ordered equilibrium state in 3D) also predict the existence of metastable states which persist over experimental time scales and possess only short-range order. Can one account for the correlation length of that order and understand how it diverges to produce long-range order in the long time limit? Our goal here

is to take a step in this direction by constructing a model to study the decay of an isolated droplet of spins of one sign, immersed in a sea of spins of the other, in the RFIM at temperatures (T) well below the transition temperature T_c . (In 2D, where $T_c = 0$, we consider T's well below the mean-field critical temperature, say. We will beg the important question of how such a droplet is formed on cooling slowly from above T_c . Rather, we imagine quenching the system from a temperature above T_c to some T significantly less than T_c . We therefore take as given the existence of droplets out of equilibrium and estimate, as a function of their linear size and the other parameters in the problem, the time required for them to evolve to their equilibrium size.)

Our model, a discrete-lattice solid-on-solid (SOS)⁶ representation similar to those used⁷ to argue for $d_c = 2$ in static equilibrium, is such that the droplet (taken initially as a hypercube of side L in d dimensions) typically finds it energetically favorable to shrink to zero or to a small fraction of its original size.^{7,8}

Our central result is that the time t_L required for this decay in any dimension ≥ 2 has the form

$$t_L \sim \exp(Lh^2/2JT), \quad L > L^* \quad , \tag{1a}$$

$$t_L \sim \exp(2L^{1/2}h/T), \ L < L^*$$
; (1b)

here, $L^* \equiv (4J/h)^2$ is a crossover length whose significance will emerge shortly; J and h are the exchange and average random-field strengths, respectively, and t_L and L are, respectively, measured in units of some characteristic microscopic spin-flip time, say 10^{-14} or 10^{-13} sec, and the lattice spacing. The inverse of (1) gives the linear size L(t) of the smallest droplets which cannot equilibrate in time t. That is, starting at t=0 and low T with a collection of (independent) droplets of different sizes, one would find, roughly speaking, only droplets of size L > L(t) left in the system after time t, where

$$L(t) \sim L^*T/T^*, T > T^*$$
, (2a)

$$L(t) \sim L^* (T/T^*)^2, \quad T < T^*$$
, (2b)

and $T^* \equiv 8J/(\ln t)$. Taking a characteristic time, say a minute or an hour, for t (i.e., $t \sim 10^{16}$) yields $T^* \sim J/5$, a

small temperature. At all but the lowest temperatures, therefore, (2a) is the appropriate formula. For sufficiently small h result (2) naturally crosses over to the far more rapid equilibration appropriate to the pure system:⁹ $L(t) \sim t^{1/2}$. The fact that the random fields produce energy barriers which must be climbed in the equilibration process accounts, as we shall see, for the striking difference between this algebraic result in pure systems and the exponentially long times of Eqs. (1).

Typical scattering experiments on the RFIM in fixed field are performed by cooling slowly whereas our arguments apply to a quench to temperatures somewhat lower than T_c . It is, moreover, likely that random exchange (i.e., dilution) of the antiferromagnets studied experimentally contributes significantly to the equilibration characteristics of those materials.^{1,2,10} This dilution is not in our model. Furthermore, the relevance of our calculations for isolated SOS droplets to the more complex equilibration of the full RFIM and its physical realizations is far from clear. We, therefore, do not attempt a detailed comparison of (2) with experiment.¹¹ However, it is worth considering the consequences of speculating that L(t) provides a reasonable measure of the characteristic linear size $\xi(t)$ of correlated domains¹² in the RFIM and its experimental realizations at time t following a rapid quench of the system at t=0 to some $T < T_c$. The rough consistency of (2) with two qualitative features of neutron measurements of ξ make one hopeful that this speculation is not too unrealistic.

(i) Taking $T^* \sim J/5$ and JT/h^2 between 1 and 100 say (e.g., both T/h and J/h between 1 and 10, corresponding roughly to the experimental range) one obtains L's between 4 and 7000 lattice constants. Thus one gets rough orderof-magnitude agreement with measurements¹³ of ξ . [The fact that L(t) decreases with decreasing T in (2) seems in flagrant contradiction of the experiments, where, in fixed field, ξ increases and eventually saturates as T is lowered. However, lowering T in (2) does *not* represent the slow cooling of a typical experiment but rather a series of quenches to progressively lower temperatures T. Once the system is quenched down to some $T < T_c$ in our model, L is determined according to (2) and ought to remain frozen at that value over any reasonable experimental time scale if the temperature is lowered further.]

(ii) The fact that (2) holds independent of dimension (d) for all $d \ge 2$ is also consistent¹¹ with the qualitative similarity of scattering results^{3,13} on the RFIM in 2D and 3D. [Interpreting (2) in 2D involves some ambiguity because the 2D RFIM cannot have long-range order;⁷ therefore, the equilibrium ξ , ξ_{eq} , is probably finite even at T=0. The form $\xi_{eq} \sim \exp[(J/h)^2]$, characteristic of a lower critical dimension,¹⁴ is often hypothesized,⁷ and numerical evidence supporting this expression has been obtained.¹⁵ If it is correct, then, at least for small enough h/J, the $\xi(t)$ inferred from Eq. (2) is $\langle \xi_{eq}$ for all reasonable t, in which case $\xi(t)$ will be observed experimentally and the finiteness of ξ_{eq} is presumably not terribly relevant. The qualitative similarity of the 2D and 3D measurements is at least consistent with this scenario.]

As we shall see, the independence of Eqs. (1) and (2) on dimension reflects the fact that the limiting slow step in the decay of a droplet in d dimensions (d an integer) is, for all $d \ge 3$, the removal of the outermost (d-1)-dimensional layer. Thus for all $d \ge 2$ the decay time is identical to the 2D decay time. This is a manifestation of d_c being 2 in the SOS model employed.⁷

To derive (1), let us start in 2D. Consider a square droplet of side L of down spins surrounded by up spins in the RFIM on a square lattice with both T and h small compared to J. To simplify the description, we adopt the SOS representation;⁶ i.e., let the droplet's shape at every moment be specified by the instantaneous (integer) heights of the Lcolumns of down spins constituting it (Fig. 1). The standard Metropolis algorithm is a convenient one for describing the temporal evolution of the column heights.

In equilibrium at low T, a 2D SOS droplet of width L in the RFIM has height^{7,16} $w \sim (h/J)^2 L$. Thus, for weak random fields, $w \ll L$. An initially square droplet will, therefore, decrease considerably in height, from L to roughly w, at low T in the long time limit. To estimate the decay time, first consider the limit $J \rightarrow \infty$, where the droplet disappears completely at long times. Any change of the column heights which costs exchange energy (i.e., which increases the surface area of the droplet) then requires infinite energy and so is forbidden. It follows that the droplet's shape remains perfectly convex, admitting no "inlets" (Fig. 1); its surface area is exactly twice the sum of its width (i.e., the fixed constant L) and its height (i.e., the height of its tallest column). Whenever the height decreases by one, an infinite energy is gained making the decrease irreversible; the height can never increase. For the height to decrease from L to L-1 obviously requires the elimination of the droplet's entire top layer. Since "inlets" are prohibitively costly this row can only be eroded systematically from its two ends: taking bites out of its middle is forbidden. Thus, the top row disappears when its two vertical boundaries, diffusing inward from the right and left, meet. The motion of each of these boundaries is like that of a particle walking randomly through a disordered medium, the disorder being supplied by the random fields. For example, when the boundary moving in from the left tries to move past a site



FIG. 1. Droplet (shaded) of down spins immersed in sea of up spins. Droplet consists of L columns (labeled by n) and has height w. Dotted lines indicate the initial (square) configuration of the droplet. Region A is an "inlet" of up spins.

where the random field points *down* (recall the droplet consists of *down* spins) it encounters an energy barrier of 2h which it crosses only with probability $e^{-2h/T}$. If there are several such adverse random fields close to one another creating a large barrier, the boundary may well get stalled and move back to the left, surmounting the barrier only on some subsequent attempt. On the other hand, the boundary progresses easily through sites with favorable (i.e., up) fields.

Sinai¹⁷ has proven exactly that a particle walking randomly on a 1D lattice wherein the probability of hopping to the right or left is a random function of its position covers distance $L \sim (\ln t)^2$ in time t (for large L and t). The physical content of this result¹⁸ is that, statistically, in traversing a large distance L, the particle will encounter regions where sites past which it has a low probability of moving outnumber sites with high transmission probability by $O(L^{1/2})$. A time $\sim \exp(L^{1/2})$ is required to cross this "barrier." In our case, regions where adverse fields outnumber favorable ones by $O(L^{1/2})$ produce barriers of $2hL^{1/2}$. We therefore estimate that the top row is annihilated in a time $t_t^{1D} \sim e^{2L^{1/2}h/T}$. One's first guess is that a total time Lt_t^{1D} is required to wipe out all L rows of the droplet, but since subsequent rows begin to erode long before the top one disappears, the droplet has probably lost a fraction of O(1)[not merely of O(1/L)] of its area by the time the top row disappears. Thus t_L^{1D} should be a good estimate for the decay time of the whole droplet.19

How is this picture altered by finite J (still in 2D)? First, the droplet decays only until the height reaches $O((h/J)^2L)$, but this is insignificant. Second, provided $t_l^{1D} > e^{2J/T}$ [i.e., $L \ge L^* \equiv (4J/h)^2$] the droplet will decay in a much less organized fashon. Its surface area will sometimes increase, a process costing at least 2J of energy and so requiring time $e^{2J/T}$. "Inlets" will now occur and, more importantly, the height can now increase: having decimated a row one is no longer assured that it will not regenerate. Indeed, since the energy to be overcome in wiping out an unfavorable row is $\sim 2L^{1/2}h$ while the exchange energy gained is only $\sim 2J$, one will, even for small h/J, actually lose energy in the elimination provided $L \ge L^*$. If, e.g., several unfavorable rows occur in succession, even if the first of them is destroyed, it may very well rebuild itself before the next one is eliminated.

Let us then model the situation as follows. Imagine that the droplet grows or shrinks by having whole rows either added to or removed from its top. The L column heights are thus constrained to move together; the droplet is always a perfect rectangle. Assume further that each horizontal row has a net random field of either $hL^{1/2}$ or $-hL^{1/2}$, each with probability $\frac{1}{2}$. Since moving the boundary down (up) decreases (increases) the droplet's surface area by 2 and hence gains (costs) 2J in exchange, the possible energy changes ΔE for downward (upward) moves are $-2J \pm 2hL^{1/2}$ ($2J \pm 2hL^{1/2}$). Thus one arrives at the following approximate picture of the droplet's progress: at discrete, regular time intervals (the natural length of which should be t_L^{1D} , the typical time required to build or destroy a row) one tries randomly to move the upper boundary either up or down. Depending on ΔE for the prospective move, it is either made or not, according to the Metropolis algorithm.

The droplet's evolution has thus been reduced to the

motion of a particle (viz., the droplet's upper boundary) in a 1D random medium. The particle feels a constant force 2J downward due to the exchange energy but encounters barriers-unfavorable rows of random fields-which for $L > L^*$ are bigger than 2J. In their exact treatment of a similar 1D random walk in a random medium, Derrida and Pomeau²⁰ showed that the presence of sufficiently strong barriers will reduce the distance R that the particle travels in time t from the linear dependence, $R \sim t$, that obtains, without the barriers, to a weaker power law, $R \sim t^a$ with a < 1. One's first thought, therefore, is that our droplet's height should reach its equilibrium value in a time algebraically dependent on L. However, the present situation differs from that of Ref. 20 in that the barrier heights grow (as $hL^{1/2}$) with the distance [of O(L)] that the particle must fall. A simple generalization of the calculations of Ref. 20 to such a situation shows that the particle requires a much longer time, viz., $\tilde{t}_L \sim e^{h^2 L/2JT}$, for the fall. The physical interpretation of this result is clear: the big-

gest barrier the particle must scale limits its overall speed. To estimate this barrier, suppose that the particle falls through *n* sites in an unfavorable region. For n >> 1 this means that $n^{1/2}$ more unfavorable rows than favorable ones have been eliminated, so the particle has crossed a randomfield barrier $\sim 2n^{1/2}L^{1/2}h$. It has simultaneously gained 2Jnin exchange energy. The difference of these two energies is maximized when $n = (h/2J)^2L$, yielding a maximal energy barrier of $Lh^2/2J$ or a climbing time of $e^{Lh^2/2JT}$. This argument suggests that the result ought to be independent of our approximating the interface by a straight line. Even when the interface looks very jagged, the exchange energy of the system is given, crudely speaking, by 2J times the overall height of the droplet. The system, therefore, on average, gains 2J of exchange for each unit drop in height, just as in the flat-interface picture. The rough estimate of $2n^{1/2}L^{1/2}h$ for the field-energy barrier encountered during a height change of *n* likewise does not require a flat interface.

In our flat-interface (1D) approximation of droplet evolution the time \tilde{t}_L should be multiplied by the step size $t_L^{1D} \sim e^{2L^{1/2}h/T}$ to give the total time for the decay: $t_L^{2D} \sim e^{2L^{1/2}h/T + Lh^{2/2JT}}$. This extra factor t_L^{1D} may be an artifact of the flat interface. Since the actual droplet decays with different speeds in different columns, many rows erode or build simultaneously. It may be an overestimate to assign a time t_L^{1D} to each overall height change of the system. We therefore keep only the leading (linear) term in L in the exponential for t_L^{2D} , i.e., we write $t_L^{2D} \sim \tilde{t}_L$. This agrees with the simple physical estimate based on the maximum energy barrier. On the other hand, it is clear that \tilde{t}_L alone does not adequately describe the decay for all L since, e.g., in the limit $L < L^*$, where the exchange energy gain from annihilating any row dominates the field-energy cost, the decay time for the droplet should approach the $J \rightarrow \infty$ (i.e., the 1D) limit t_L^{ID} . Exactly how the crossover at $L \sim L^*$ between \tilde{t}_L and t_L^{ID} occurs is unclear. We will not speculate on this; our estimates for t_L^{2D} in the two limits $L > L^*$ and $L < L^*$ are summarized in Eq. (1).

To generalize these results to three and higher dimensions, imagine constructing a 3D SOS model droplet, consisting of a 2D array of columns. Take the droplet to be a cube of side L initially. Removing one 2D layer then gains 4JL in exchange energy and either gains or loses $\sim 2hL$ in field energy. Therefore, if 2J > h (a condition we assume satisfied) the exchange inevitably dominates. One might think, then, that the boundary falls linearly with time and that the droplet decays in a time of O(L). This is incorrect. Each 2D layer takes time t_L^{2D} to erode. Presumably by the time the first layer of the droplet has completely disappeared a volume fraction of O(1) [not O(1/L)] of the droplet has eroded; the total time for the decay of the 3D droplet is, in consequence, something like t_L^{2D} . A similar argument applies to all dimensions higher than 3: in any dimension d > 2 the peeling off of the first (d-1)-dimensional layer is the slowest step and dominates the decay time in all higher integral dimensions.

Subsequent to the completion of this research we received a report of work prior to publication of Villain,²¹ who has

- ¹See, e.g., Y. Imry, J. Stat. Phys. (to be published); G. Grinstein, J. Appl. Phys. <u>55</u>, 2371 (1984), for brief reviews and detailed bibliographies.
- ²S. Fishman and A. Aharony, J. Phys. C <u>12</u>, L729 (1979).
- ³See, e.g., R. J. Birgeneau, R. A. Cowley, G. Shirane, and H. Yoshizawa, J. Stat. Phys. <u>34</u>, 817 (1984); P.-z. Wong, J. W. Cable, and P. Dimon, J. Appl. Phys. <u>55</u>, 2377 (1984), for brief reviews.
- ⁴Consistent with this is the fact that specific heats in Fe_xZn_{1-x}F₂, measured in fixed field, look so sharp. See, D. P. Belanger, A. R. King, V. Jaccarino, and J. Cardy, Phys. Rev. B <u>28</u>, 2522 (1983).
- ⁵P.-z. Wong and J. W. Cable (unpublished) have observed a very slow increase with time of the correlation length obtained by finite field cooling in $Fe_xCo_{1-x}Cl_2$.
- ⁶See, e.g., J. D. Weeks, in Ordering in Strongly Fluctuating Condensed Matter Systems, edited by T. Riste (Plenum, New York, 1980).
- ⁷G. Grinstein and S.-k. Ma, Phys. Rev. Lett. <u>49</u>, 685 (1982); Phys. Rev. B <u>28</u>, 2588 (1983); J. Villain, J. Phys. (Paris) Lett. <u>43</u>, L551 (1982); J. Villain, B. Semeria, F. Lancon, and L. Billard, J. Phys. C <u>16</u>, 6153 (1983); K. Binder, Z. Phys. <u>50</u>, 343 (1983); T. Nattermann, J. Phys. C <u>16</u>, 6407 (1983); D. S. Fisher, J. Frohlich, and T. Spencer, J. Stat. Phys. (to be published).
- ⁸At reasonably low temperatures it is highly unlikely (though possible) that the droplet remains large after long times, since large droplets represent a substantial exchange energy cost.
- ⁹I. M. Lifshitz, Zh. Eksp. Theor. Fiz. <u>42</u>, 1354 (1962) [Sov. Phys. JETP <u>15</u>, 939 (1962)]; J. W. Cahn and S. M. Allen, Acta Metall. <u>27</u>, 1085 (1979). See, also, e.g., J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1983), Vol. 8; M. K. Phani, J. L. Lebowitz, M. H. Kalos, and O. Penrose, Phys. Rev. Lett. <u>45</u>, 366 (1980); P. Fratzl, J. L. Lebowitz, J. Marro, and M. H. Kalos, Acta. Metall. <u>31</u>, 1849 (1983); P. S. Sahni, G. S. Grest, S. A. Safran, Phys. Rev. Lett. <u>50</u>, 60 (1983); G. S. Grest, S. A. Safran, and P. S. Sahni, J. Appl. Phys. <u>55</u>, 2432 (1984); A. Sadiq and K. Binder, Phys. Rev. Lett. <u>51</u>, 674 (1983), and references therein.

obtained a result identical (aside from a factor of 2) to (2a) independently and by arguments somewhat different from ours. (He uses, e.g., a continuum model rather than our discrete one.) He has also obtained a generalization of (2) applicable for T's up to and above T_c . In so doing he has argued that, at least for dimensions d < 4, cooling through T_c indeed results in the nonequilibrium configurations we have assumed. Bruinsma and Aeppli²² and Shapir²³ have also studied the equilibration of the RFIM, obtaining results similar in spirit but different in detail from ours.²⁴

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(1983).

- ¹¹R. J. Birgeneau (private communication) has undertaken such a comparison.
- ¹²It is easy to argue (see Ref. 19) that this hypothesis is correct for the pure Ising model at low T. That is, a single SOS droplet of linear size L in any dimension decays away in time $t_L \sim L^2$, in agreement with the result $\xi(t) \sim t^{1/2}$ of Ref. 9. (See Sahni *et al.*, Ref. 9, for detailed discussion.)
- ¹³M. Hagen, R. A. Cowley, S. K. Satija, H. Yoshizawa, G. Shirane, R. J. Birgeneau, and H. J. Guggenheim, Phys. Rev. B <u>28</u>, 2602 (1983); R. J. Birgeneau, H. Yoshizawa, R. A. Cowley, G. Shirane, and H. Ikeda, *ibid.* <u>28</u>, 1438 (1983).
- ¹⁴E. Pytte, Y. Imry, and D. Mukamel, Phys. Rev. Lett. <u>46</u>, 1173 (1981) and H. S. Kogon and D. J. Wallace, J. Phys. A <u>14</u>, L527 (1981) derive $\xi \sim \exp[(J/h)^2]$ in 3D for interface representations which predict $d_c = 3$ for the RFIM; see, also, A. Aharony and E. Pytte, Phys. Rev. B <u>27</u>, 5872 (1983).
- ¹⁵J. F. Fernandez and E. Pytte (private communication).
- ¹⁶J. F. Fernandez, G. Grinstein, Y. Imry, and S. Kirkpatrick, Phys. Rev. Lett. <u>51</u>, 203 (1983).
- ¹⁷Ya. G. Sinai, in Proceedings of the Berlin Conference on Mathematical Problems in Theoretical Physics, edited by R. Schrader, R. Seiler, and D. A. Uhlenbrock (Springer, Berlin, 1982), p. 12.
- ¹⁸E. Marinari, G. Parisi, D. Ruelle, and P. Windey, Phys. Rev. Lett. <u>50</u>, 1223 (1983).
- ¹⁹Note that since $t \sim L^2$ is the time required for a particle in 1D to walk randomly through distance L in a *pure* system (h=0), this argument correctly predicts (Ref. 9) $\xi \sim t^{1/2}$ at low T in the pure 2D Ising model. A similar argument predicts $\xi \sim t^{1/2}$ for all d.
- ²⁰B. Derrida and Y. Pomeau, Phys. Rev. Lett. <u>48</u>, 627 (1982).
- ²¹J. Villain, Phys. Rev. Lett. <u>52</u>, 1543 (1984).
- ²²R. Bruinsma and G. Aeppli, Phys. Rev. Lett. <u>52</u>, 1547 (1984).
- ²³Y. Shapir (unpublished).
- ²⁴M. Grant and J. D. Gunton, Phys. Rev. B <u>29</u>, 1521 (1984) and (unpublished), have studied the initial stages of domain formation and growth in the RFIM.

¹⁰See, e.g., P.-z. Wong and J. W. Cable, Phys. Rev. B 28, 5361