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## Domain growth kinetics in strongly disordered Ising magnets

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**Abstract.** — We investigate the law of domain growth in *strongly* disordered Ising magnets in two dimensions by Monte Carlo simulation. The average linear domain size is found to grow with time  $t$  as  $R(t) \sim (\ln t)^x$ . For all the concentrations of the impurities used in our simulations the numerical value of the exponent  $x$  is found to be smaller than the corresponding theoretical prediction thereby indicating the possibility that the asymptotic regime, where the theory *may* hold, lies beyond the time scale of our observation. Nevertheless, in contrast to the recent laboratory experiment of Ikeda *et al.*, the *effective* exponents are found to exhibit at least a kind of « quasi-universality » (temperature-independence) provided the Monte Carlo data for the domain size at a given impurity concentration are fitted to the form  $(\ln t)^x$  over the same time interval consistently at all temperatures.

### 1. Introduction.

The phenomenon of spinodal decomposition in simple non-random systems is now quite well understood (see Refs. [1-4] for reviews). However, much less attention has been paid so far to the effects of disorder on the kinetics of ordering although impurities are known to be ubiquitous. Few efforts in this direction have been made over the last few years, mostly using computer simulation. One of the prototype models studied in these works is the Ising model in the presence of *quenched* or *annealed* randomness; the precise nature of the randomness depends on the physical situation the model represents. These models not only represent a class of magnetic materials but also several other systems, e.g., a particular type of chemisorbed layers [5]. In these investigations the  $d$ -dimensional spin system is quenched from the high temperature paramagnetic phase to a temperature well below the coexistence curve and the size of the domains is monitored as a function of time  $t$ . Disorder can enter the Ising model in several different ways, e.g., through random field [6] or through random exchange [7]. Domain growth in the random-field Ising model (RFIM) has been studied theoretically [8-12] and by experimental investigation of the growth of superlattices of chemisorbed layers [13] as well as by computer simulation [14-16]. In this communication,

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however, we are concerned with domain growth in the *random-exchange* Ising models [7]. In one of the versions of the random-exchange Ising model the randomness enters through the random occupation of the lattice sites by the Ising spins; a fraction  $p$  of the lattice sites is occupied randomly by the spins and the fraction  $1 - p$  is occupied by non-magnetic impurities (or vacancies). Disorder in this model is said to be annealed if the impurities are mobile. The phenomenon of domain growth in the Ising model with annealed disorder has also received some attention in the recent literature [17, 18]. Very recently, the role of mobile vacancies in the process of phase separation in binary alloys has also been investigated [19]. In this communication we shall, however, focus attention on the Ising model with *quenched* (i.e., immobile) impurities, called the dilute Ising model (DIM).

In a non-random system the growth of the domains in the late stages is driven by the interfacial curvature, i.e., the local velocity  $dR/dt$  of the interface is proportional to its local curvature  $1/R$ . Consequently, in these systems with non-conserved order parameter the linear domain size  $R(t)$  follows the Lifshitz-Allen-Cahn (LAC) growth law [1]  $R^2 \sim t$ . The domain growth in the DIM, on the other hand, proceeds through thermally activated motion of the interfaces. The main aim of this communication is to determine the law of domain growth in the *strongly* diluted Ising model by Monte Carlo (MC) simulation. It has been suggested earlier [20], albeit based on heuristic arguments, that during the late stages the *typical* (linear-) domain size in the DIM should grow as  $R(t) \sim (\ln t)^x$  where  $x$  is a universal quantity that depends on the dimensionality  $d$  of the system but is independent of the other details, e.g., temperature. Specifically,  $x = 4$  in  $d = 2$ . All the earlier simulations [21-23] were performed in the weak disorder regime. Although a crossover from power-law growth to a slower growth was observed during the late stages in these simulations, neither the value of  $x$  nor its possible temperature-dependence was investigated. In this communication we not only present evidence in support of the form  $R(t) \sim (\ln t)^x$  in the strong disorder regime but also investigate the temperature dependence of  $x$ . We also discuss here some possible reasons for the apparent inconsistency between a recent experiment [24] and the theoretical prediction [20]. A short comment on the interpretation of the data from this experiment has been made earlier in the light of the preliminary observations in our MC simulations [25].

Recently Ikeda *et al.* [24] have studied domain growth in the two-dimensional Ising system  $\text{Rb}_2\text{Co}_p\text{Mg}_{1-p}\text{F}_4$ . In contrast to the earlier MC simulations [21-23], Ikeda *et al.* [24] focussed attention on the strong disorder regime (spin concentration  $p$  only slightly above the percolation threshold  $p_c$ ). A crossover from power-law growth to a slower growth was observed at the late stages in this experiment. This crossover is qualitatively similar to that observed in the earlier MC simulations. However, by fitting the experimental data to the form  $R(t) \sim (\ln t)^x$  in the long-time regime they found that the exponent  $x$  is a temperature-dependent non-universal quantity. The experimental observation of Ikeda *et al.* is of fundamental importance because it apparently contradicts the earlier theoretical prediction [20]. As a first step towards a better understanding of this kinetic growth phenomenon the existing gap between laboratory experiment and computer experiment must be bridged by performing MC simulation in the strong dilution regime (preferably in the same ranges of parameters as those in the experiment of Ikeda *et al.* [24]). In this communication we report the results of such simulations and compare these with the theoretical predictions and the experimental observations.

## 2. Monte Carlo simulation.

We begin by mentioning the differences between the experimental approach of Ikeda *et al.* [24] and our method of computing  $R(t)$ . Since  $\text{Rb}_2\text{Co}_p\text{Mg}_{1-p}\text{F}_4$  is an *antiferromagnet* Ikeda *et al.* followed an indirect method to measure  $R(t)$ .  $\text{Rb}_2\text{Co}_p\text{Mg}_{1-p}\text{F}_4$  is known to be a physical

realization of the two-dimensional Ising antiferromagnets. For example, long range Neel order sets in  $\text{Rb}_2\text{Co}_{0.6}\text{Mg}_{0.4}\text{F}_4$  at  $T_N = 20$  K. In a uniform magnetic field these disordered Ising antiferromagnets are known to be experimental realizations of the RFIM. It is now also well established [6] that RFIM does not exhibit long range order (LRO) in two dimension. Therefore, LRO in  $\text{Rb}_2\text{Co}_{0.6}\text{Mg}_{0.4}\text{F}_4$  can be destroyed by applying an external magnetic field. The system, however, recovers LRO slowly after the field is switched off and the system behaves as a simple dilute Ising system with antiferromagnetic exchange interaction. Monitoring the decay of magnetization during this recovery of LRO, Ikeda *et al.* extracted the growth law for the *average* domain size by relating magnetization  $M(t)$  to domain size through the relation  $R(t) \sim [M(t)]^{-1}$ . We have, however, computed the *average* domain size *directly* for the *ferromagnetic* DIM from the Sadiq-Binder definition of  $R(t)$  [26] used earlier in the literature [22]. According to this prescription

$$R^2(t) = [pN \langle \Sigma (S_i/pN)^2 \rangle],$$

in a lattice consisting of  $N$  sites, i.e.,  $pN$  spins, where the symbol  $\langle \cdot \rangle$  denotes average over a large number of the quenches of the system whereas  $[\cdot]$  denotes averaging over a large number of impurity configurations.

We have carried out the simulation on a square lattice where a fraction  $p$  of the sites is occupied by the Ising spins; nearest-neighbour spins are assumed to interact with each other through exchange interaction of constant strength  $J$ . All the data have been generated for  $100 \times 100$  systems to reduce finite-size effects. The systems with  $p = 0.75$  were simulated using a CONVEX C210 computer at ICTP and those for  $p = 0.65$  were simulated using a Kontron Intel 860 computer at HLRZ. The latter was about two times faster than the CONVEX computer and about as fast as an IBM 3090. We updated the spins randomly rather than sequentially to avoid any spurious kinematic effect of the algorithm. We quenched the system from very high temperatures, where each of the spins is equally likely to be up or down, to a temperature  $T$  well below the corresponding transition temperature  $T_c(p)$ . We have monitored  $R^2(t)$  as a function of time using the standard Metropolis algorithm as the system evolved with time following the single-spin-flip Glauber kinetics. At each temperature  $T$  we averaged the data for  $R^2(t)$  over 100 impurity configurations at  $p = 0.75$  and over 1 000 impurity configurations at  $p = 0.65$ . Since the quantity of interest is known to be a non-self-averaging one [27] averaging over such a large number of configurations is absolutely essential. Unfortunately, we could not carry out the simulation for  $p < 0.65$  because of prohibitively large CPU time required to produce data of equally high quality. Moreover, the closer is  $p$  to  $p_c$  the lower is  $T_c(p)$  and the narrower is the range of temperature over which the system exhibits long-range order. Therefore, in order to study the  $T$ -dependence of  $x$  over a reasonably wide range of temperature with affordable computer time we chose the two values  $p = 0.65$  and  $p = 0.75$ . However, as we shall show below, the data of our simulation at these concentrations have some features similar to those of experimental data and, therefore, are adequate to explore the possible reasons of disagreement between theory and experiment.

### 3. Analysis of the Monte Carlo data.

Let us now analyze our MC data for  $R^2(t)$ . First of all, the crossover from the power-law to logarithmic growth regime is gradual. Therefore, while fitting the data to the  $(\ln t)^x$  form over the interval  $t_{\min} \leq t \leq t_{\max}$  one must choose sufficiently large  $t_{\min}$  so that the power-law regime is excluded. Throughout our analysis we have chosen  $10^3$  as  $t_{\min}$  because the slower growth was observed at all temperatures already around that time. So far as the choice of  $t_{\max}$  is concerned one has to be careful. The growth of the domains effectively stops after

attaining a temperature-dependent maximum average size  $R_{\max}$ ; the higher is the temperature the larger is the  $R_{\max}$ . This is a consequence of the fact that the thermal energy available at the given temperature is inadequate to move the interface over length scales longer than  $R_{\max}$  within the observation time in our simulation. Similar effects have been observed in all the earlier works in the weak disorder regime [21-23]. Note that  $R_{\max}$  is usually much smaller than the system length. Therefore, one must choose  $t_{\max}$  such that  $R(t_{\max}) < R_{\max}$ .

From the theory [20] one would expect that  $R^2 \sim (\ln t)^{2x}$  with  $x = 4$  in  $d = 2$ . However, we get a straight line fit to the data at  $T = 0.15 T_c(1)$  for  $p = 0.75$  by plotting  $R^2(t)$  against  $(\ln t)^{0.5}$  with  $x = 0.25$  over the time interval  $10^3 \leq t \leq 3.5 \times 10^4$  (see Fig. 1). At first sight this apparently contradicts the theoretical prediction. A similar analysis (see Fig. 2) at  $T = 0.25 T_c(1)$  for the same concentration  $p = 0.75$  over the interval  $10^3 \leq t \leq 5 \times 10^4$  yields an *effective* exponent  $x = 1.5$ . The opposite curvatures in the plots of  $R^2$  against

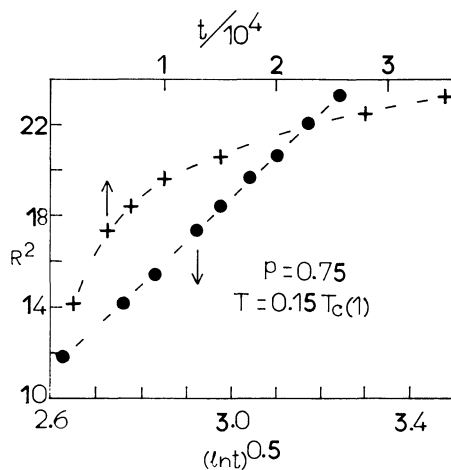


Fig. 1. —  $R^2$  at  $T = 0.15 T_c(1)$  for  $p = 0.75$  plotted against  $t$  and  $(\ln t)^{0.5}$ .

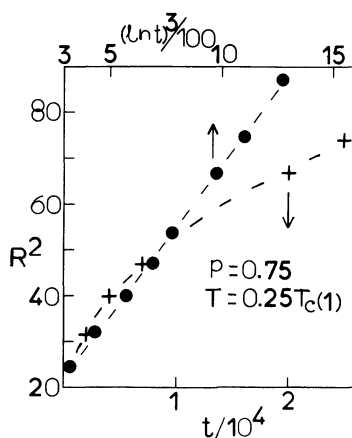


Fig. 2. —  $R^2$  at  $T = 0.25 T_c(1)$  for  $p = 0.75$  plotted against  $t$  and  $(\ln t)^3$ .

$(\ln t)^2$  and  $(\ln t)^5$  in figure 3 not only demonstrates the high accuracy of our data but also convincingly shows that the effective exponent  $x$  observed over this time interval at this temperature must lie between 2 and 5. From these observations at  $T = 0.15 T_c(1)$  and  $T = 0.25 T_c(1)$  for  $p = 0.75$  it is tempting to conclude that our MC simulation is consistent with a temperature-dependent exponent  $x$ , thereby violating the theoretical prediction. But such a conclusion would be too premature. Fitting the  $R^2$  data for  $p = 0.75$  against

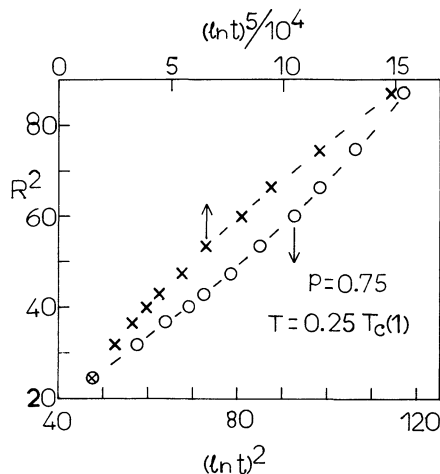


Fig. 3. —  $R^2$  at  $T = 0.25 T_c(1)$  plotted against  $(\ln t)^2$  and  $(\ln t)^5$ .

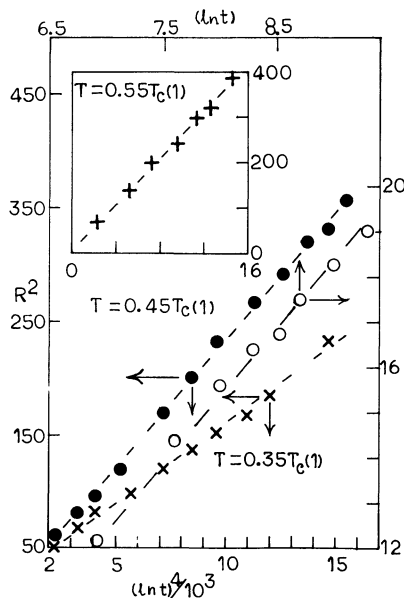


Fig. 4. —  $R^2$  at  $T = 0.35 T_c(1)$  ( $\times$ ),  $T = 0.45 T_c(1)$  ( $\bullet$ ) and  $T = 0.55 T_c(1)$  ( $+$ ) for  $p = 0.75$  plotted against  $(\ln t)^4$ .  $R^2$  at  $T = 0.4 T_c(1)$  for  $p = 0.65$  ( $\circ$ ) is plotted against  $(\ln t)$  (see the different scales for different set of parameters).

$(\ln t)^{2x}$  over the time interval  $10^3 \leq t \leq 10^5$  at higher temperatures  $T = 0.35 T_c(1)$ ,  $T = 0.45 T_c(1)$  and  $T = 0.55 T_c(1)$  we get the same effective exponent  $x = 2$  at all these three temperatures (see Fig. 4). The data at lower temperatures  $T = 0.15 T_c(1)$  and  $T = 0.25 T_c(1)$  could not be fitted over the latter interval because the average domain size at these lower temperatures saturate to  $R_{\max}$  much before  $t = 10^5$ . However, the data at the higher temperatures can always be fitted over a smaller time interval. Carrying such an analysis we observed that the  $R^2$  data at  $T = 0.55 T_c(1)$  as well as that at  $T = 0.25 T_c(1)$  can be fitted to  $(\ln t)^{2x}$  with the same exponent  $x = 1.5$  provided we fit the data over the same interval  $10^3 \leq t \leq 5 \times 10^4$ ; the curvature at  $T = 0.55 T_c(1)$  becomes visible only after the data at longer times are also included in the fitting. For  $p = 0.65$  we found  $x = 0.5$  by fitting  $R^2(t)$  at  $T = 0.4 T_c(1)$  over the interval  $10^3 \leq t \leq 2 \times 10^4$  (see Fig. 4).

We have computed the *effective* exponent  $x(t)$  at successive time intervals from  $t = 10$  to  $t = 10^5$  from the slope of the  $\ln R^2$  versus  $\ln(\ln t)$  curve and observed the rate of increase of  $x$  to decrease with increasing  $t$ . Therefore, it is difficult to extrapolate  $x(t)$  to  $t \rightarrow \infty$ . Nevertheless, although the *effective* exponent  $x$  turns out to be smaller than the true asymptotic exponent, our analysis demonstrates that a kind of «quasi-universality» (temperature-independence) is exhibited by the *effective* exponents provided, of course, the data at all  $T$  is consistently fitted to the form  $(\ln t)^{2x}$  over the same time interval.

#### 4. Conclusion.

Now we speculate on the possible reasons of disagreement between the theoretical prediction and the experimental observation. It is well known from the literature on static critical phenomena that the effective exponents extracted from the data outside the true critical regime need not exhibit «universality». It is quite possible that the time scale where the theoretical prediction is expected to hold is much beyond the time scale of observation in the experiment of Ikeda *et al.* If this is true, then the apparent disagreement between theory and experiment is just a consequence of short time scale of experimental observation.

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#### Note added in proof :

After the submission of this work we obtained a new growth law by incorporating into the Huse-Henley theory the dynamical effects of the self-similarity of the percolating structure near  $p_c$ . We have compared this new growth law with our MC data (details are given in Ref. [25]). We now feel that the true asymptotic regime might have been probed in the experiment of Ikeda *et al.* ; but their data must *not* be compared with the growth law proposed originally in reference [20] because the latter is not expected to hold for  $p$  close to  $p_c$ .

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