

## Lack of Self-Averaging, Multiscaling, and $1/f$ Noise in the Kinetics of Domain Growth

Christopher Roland and Martin Grant

*Department of Physics, McGill University, Rutherford Building, 3600 University Street,  
Montréal, Québec, Canada H3A 2T8*

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The nature of non-self-averaging behavior in the kinetics of domain growth is studied by Monte Carlo simulations. Fluctuations in the scaling regime of the two-dimensional spin-flip Ising model are found to involve multiscaling, as is known from other problems, such as percolating resistor networks and diffusion-limited aggregation. The frequency-dependent fluctuations in the scaling regime are found to be  $(1/f)$ -like: The power spectrum obeys  $1/\omega^\phi$ , where  $\omega = 2\pi f$  is the frequency and  $\phi \approx 0.9$ . These results can be tested experimentally.

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During a first-order phase transition, when one quenches a system from a high temperature where the system is disordered to a low temperature, ordering takes place. A long-wavelength instability creates ordered domains which grow as time goes on.<sup>1</sup> When the domains' average size  $R(t)$ , as a function of time  $t$ , becomes large, it is often found that the growth follows  $R \sim t^n$ , where  $n$  is the growth exponent. For late times, the lengths scale to  $R(t)$ . For example, the order-parameter correlation function  $g(r, t)$  satisfies  $g(r, t) \approx G(r/t^n)$ . The exponent  $n$  is known for some universality classes. For model *A*, the universality class of the nonconserved Ising model, and model *B*, the conserved Ising model, the generally accepted results<sup>2,3</sup> are  $n = \frac{1}{2}$  and  $n = \frac{1}{3}$ , respectively.

In this Letter, we present a study of the fluctuations around this late-time growth. In particular, we show for the first time that those fluctuations, which are dominated by non-self-averaging behavior,<sup>4-11</sup> give rise to multiscaling phenomena and  $1/f$  flicker noise. These are familiar from the study of random systems such as percolating resistor networks and diffusion-limited aggregation,<sup>4-7</sup> and the self-organized critical phenomena of driven systems.<sup>8</sup> We believe that these phenomena are due to the interplay between different length and time scales (i.e., the polydispersity of the evolving system) during domain growth.

Lack of self-averaging during first-order transitions has previously been noted and studied by Sadiq and Binder,<sup>9</sup> Gawlinski *et al.*,<sup>10</sup> and recently in an elegant paper by Milchev, Binder, and Heermann.<sup>11</sup> A lack of self-averaging implies that the evolving system cannot be decomposed into many independent parts. Thus, provided edge effects are not important, the statistics from one sample of size  $N$  are a factor of 4 worse than the statistics from four systems of size  $N/4$ . This has serious implications, because self-averaging is the basis for the theory of thermodynamic fluctuations. The usual situation is that relative fluctuations of thermodynamic quantities are of order  $1/\sqrt{N}$ . In such cases the central-limit theorem can be invoked, the distributions of thermo-

dynamic quantities are Gaussians, and fluctuations vanish in the thermodynamic limit.<sup>12</sup> We study two consequences that the lack of self-averaging has for a system undergoing a first-order phase transition: Spatial fluctuations lead to multiscaling or a hierarchy of exponents, while temporal fluctuations lead to  $1/f$  noise.

The details of our Monte Carlo simulations are standard. We present results for the two-dimensional ferromagnetic Ising model with spin-flip dynamics (model *A*). Experimental systems which correspond to this include binary alloys and chemisorbed systems on substrates, undergoing order-disorder transitions. We have also studied the spin-exchange Ising model (model *B*); those results will be presented in a future paper.<sup>13</sup> The Ising Hamiltonian is  $\mathcal{H} = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j$ , where  $J$  is the interaction constant, the sum runs over distinct nearest-neighbor pairs on a square lattice, and the  $N$  spins can take on the values  $\sigma_i = \pm 1$ . The system is quenched from an infinite temperature to a low temperature,  $T = 0.5T_c$  ( $T = 0$  and  $T = 0.9T_c$  were also studied), where  $T_c$  is the critical temperature. The system was studied for up to 4000 Monte Carlo steps, and in at least 256 independent runs. Lattices of sizes  $64^2$ ,  $128^2$ , and  $256^2$  were considered. The simulations were performed using a multispin-cooling algorithm. The average domain size  $R(t)$  was estimated from one of the following measures: from the inverse perimeter density  $R_e(t) = 2/(2 + E/J)$ , where  $E$  is the energy per spin, and from the magnetization, where  $R_m^2(t) = NM^2$ , and  $M$  is the magnetization per spin.

To study the motion of the growth fronts, we have segregated the evolving system into sets of sites determined by the probability  $p$  of a spin flipping within one Monte Carlo step. In Fig. 1, typical configurations as well as "active" sites are shown. At an active site a spin changes sign within one Monte Carlo step. The probabilities  $p$  are generated by stopping a simulation at time  $t$ , and then generating a large number of independent configurations (250–1000, typically 500) at time  $t + 1$ . This was repeated at regular time intervals during the order-disorder transition. It is evident in Fig. 1 that in-

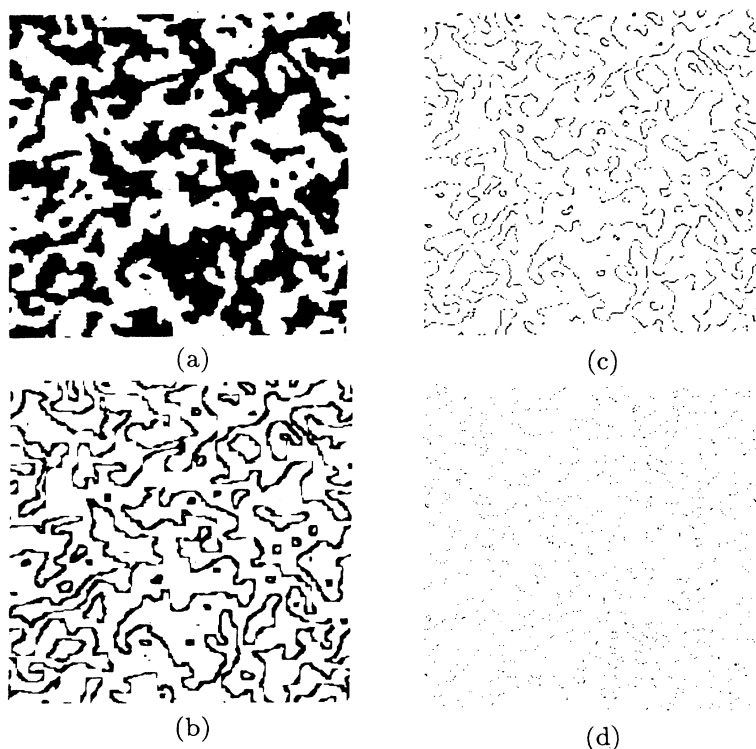


FIG. 1. (a) Typical configuration,  $N=256^2$ ,  $T=0.5T_c$ ,  $t=25$  Monte Carlo steps, and spins which are active with probability (b)  $p \geq 0.002$ , (c)  $p \geq 0.6$ , and (d)  $p \geq 0.9$ .

interface dynamics plays a key role in domain growth, as expected.<sup>2</sup> In particular, note that the description of the system in terms of  $p$ 's is both a convenient and physical way to analyze the distribution of domains. Domains with large curvatures have very active sites at their interfaces, while domains with small curvatures have relatively inactive sites. Thus, studying the  $p$ 's permits the study of the polydispersivity of the evolving system in the late-time scaling regime. By polydispersivity, we mean the system consists of domains of many different sizes.

We have estimated the site growth distribution function,  $\mathcal{N}(p, t)$ , by determining the number of sites active with the same  $p$ .<sup>14</sup> This distribution changes in time because, at early times, a large amount of interface is present and many sites are active, while at late times, few sites are active. We characterize these distributions by their moments,<sup>15</sup>  $M_k(t) = \sum_p \mathcal{N}(p, t) p^k$ , following standard treatments.<sup>4-7</sup> Since the time dependence is controlled by the domain size  $R(t)$ , we make the *Ansatz*

$$M_k(t) \sim R(t)^{-\tau(k)} \sim t^{-n\tau(k)},$$

where we make use of the known value of the growth exponent  $n = \frac{1}{2}$  in  $R \sim t^n$ . These exponents  $\tau(k)$  can be thought of as corrections to dynamical scaling found in first-order transitions. We have found that the *Ansatz* is reasonable, in that the values which we obtain for  $\tau$  are independent of time. Since there exists a large range of curvatures (which are probed by the  $p$ 's),  $\tau$  is a nonlinear

function of  $k$ , which is equivalent to multiscaling. The usual " $f(\alpha)$ " description is discussed below.

The multiscaling behavior can be contrasted with the scaling of the distribution  $\mathcal{P}(R, t)$  for the domain size itself,<sup>14</sup> shown in the inset of Fig. 2.  $\mathcal{P}(R, t)$  is defined over an ensemble of initial conditions. We have  $M'_k = \sum_R \mathcal{P}(R, t) R^k$ , with  $M'_k \sim t^{-n\tau'(k)}$ . In this case  $\tau'(k)$  is, to our accuracy, a linear function of  $k$ , which is called "gap scaling." Here, multiscaling is a consequence of the scale over which we probe our system:

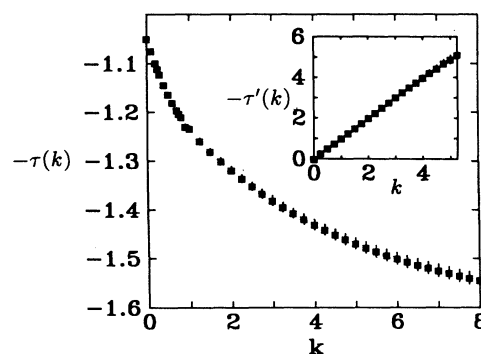


FIG. 2. Exponents  $\tau$  of  $k$ th moments of distribution of firing probabilities showing no gap scaling, i.e.,  $\tau$  is not a linear function of  $k$ . Inset: Gap scaling with  $R$  moments,  $\tau'$ . Error bars are estimated to be 3 standard deviations.

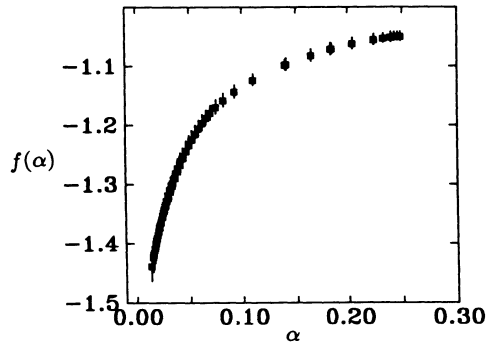


FIG. 3. Dimension  $f$  of set of active sites vs strength of singularity  $\alpha$ . Error bars are estimated to be 3 standard deviations.

$\mathcal{N}(p, t)$  contains explicit information about the short-wavelength scaling fields, while  $\mathcal{P}(R, t)$  probes only the long-wavelength behavior of an ensemble of systems. Gap scaling is associated with these long-wavelength relevant scaling fields, as discussed by Tremblay, Fourcade, and Breton,<sup>7</sup> where an explicit connection to critical phenomena is made.

While the behavior of  $\tau(k)$  determines the asymptotic properties of the moments  $M_k$ , it is also of interest to find the scaling behavior of  $\mathcal{N}(p)$  itself, where say  $\mathcal{N} \sim R^f$ . The analysis is standard.<sup>4-7</sup> The Legendre transformation of  $\tau(k)$  gives

$$f(\alpha) = k\alpha - \tau,$$

where  $\alpha = d\tau/dk$ . The form can be motivated as follows. Each moment  $M(k)$  is characterized by the most probable  $p = p^*(k)$  in the weighted sum over the distribution. Then, if  $p^* \sim R^{-\alpha}$ , and  $\mathcal{N}(p^*) \sim R^f$ , one obtains  $f = k\alpha - \tau$ , with a natural interpretation for the Legendre transformation.<sup>16</sup> The  $f$  vs  $\alpha$  curve is shown in Fig. 3. The exponent  $f$  can be interpreted as the dimension of the space of singularities of  $p$ 's with strengths  $\alpha$ . Loosely speaking, we have divided the system into many subsets characterized by  $\alpha \sim -\ln p^*/\ln R$ , which have different  $f$ 's. A constant value of  $f$  implies gap scaling, where  $\tau(k)$  is a linear function of  $k$ . Figure 3 shows that this is not the case here since  $f$  varies with  $\alpha$ . Note that we have defined  $\mathcal{N}$  in terms of *many* domains in a system of size  $N$ , while in other fields<sup>6</sup> only a *single* cluster is considered. This implies that  $f$  is negative here, since the number of domains decreases as time goes on. The maximum value for  $f$  is approximately  $-1$ , because active sites are at interfaces. Note that it is the large moments which probe small length scales with large curvatures.

It is possible that some of our results are affected by transients. Note that many studies<sup>1</sup> measure  $n$  and scaling, while here we are essentially measuring the corrections to scaling. Nevertheless, we expect multiscaling to be a common feature of the kinetics of first-order transitions. Indeed, we have seen the same qualitative trend in

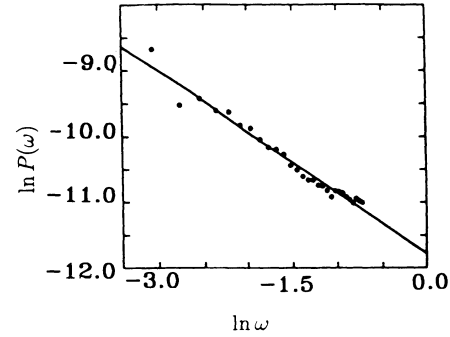


FIG. 4. Natural logarithm of power spectrum  $P(\omega)$  vs natural logarithm of frequency  $\omega$ . The line shows a fit by  $1/f$  behavior. Exponent is  $\phi \approx 0.9$ .

model  $B$ , the spin-exchange Ising ferromagnet.<sup>13</sup> It should be mentioned that these  $f$  vs  $\alpha$  curves can be obtained experimentally. The difficulty is that one requires a large number of independent experiments, since the curve is determined by *all* the probable motions of the interfacial fronts, rather than only by the most probable motion of those fronts.

As mentioned above, the hierarchy of exponents necessary to describe the corrections to scaling is due to the polydispersity of domain growth. Therefore, there are large spatial fluctuations between many nearby states of the dynamical system, which are almost equivalent. This is similar to the picture of Bak, Tang, and Wiesenfeld<sup>8</sup> for self-organized critical phenomena in simple driven systems. They find that scaling and self-organization are accompanied by temporal fluctuations which are  $(1/f)$ -like, where  $\omega = 2\pi f$  is the frequency. Thus, we have studied the temporal fluctuations in the scaling regime of the random variable  $\eta(t) = R(t)/t^{1/2} - \langle R(t)/t^{1/2} \rangle$ , where the angular brackets denote an average over initial conditions. Figure 4 shows that the power spectrum<sup>17</sup> of those fluctuations obeys

$$P(\omega) \sim 1/\omega^\phi,$$

for small  $\omega$ , where  $\phi = 0.9 \pm 0.05$ .

We speculate the  $(1/f)$ -like temporal correlations are a general feature of the kinetics of first-order transitions.<sup>18</sup> Note that, since  $\phi$  is due to fluctuations, it may be dependent upon the dimensionality of space, unlike the growth exponent  $n$ .<sup>19</sup> We expect  $\phi$  to be measurable through standard techniques. For example, inelastic neutron scattering from a phase-separating binary alloy, or a synchrotron-radiation study at sufficiently high resolution, should be able to obtain this quantity. We should also note that Tang and Bak<sup>8</sup> have suggested scaling relations for self-organized critical phenomena which may apply here. We have not been able to test those relations with our present data.

It is worth mentioning at this point why one would expect the physics of random systems to be analogous to

that of a pure system undergoing a first-order transition. Although this system is not random, it is polydisperse with a diverging size of correlated regions, due to the initial instability following the quench from the disordered state. The initial conditions are controlled by the weak-coupling infinite-temperature fixed point, while the subsequent dynamics are controlled by the strong-coupling  $T \rightarrow 0$  fixed point.<sup>20</sup> Both fixed points are attractive, as one would expect for self-organized critical phenomena. Also, note there are  $2^N$  microscopic replicas of the initial state, but only a few, say two, of the final one. The noise in the initial conditions is amplified by the instability, so that the replicas of the initial conditions give rise to many dynamical systems which are not "close," i.e., they cannot be reached by small thermal fluctuations. Indeed, as time goes on, thermal fluctuations are more and more ineffectual in averaging the structure of many domains. The lack of self-averaging, then, is because noise from the initial conditions is amplified by the instability to macroscopic length and time scales. One must average over the initial conditions (an ensemble average), rather than only the thermal fluctuations in different parts of one evolving replica, to control that noise.

We have found that the scaling regime of the kinetics of first-order transitions is dominated by non-self-averaging fluctuations. Those fluctuations give rise to multiscaling and  $1/f$  noise. In conclusion, we would like to reemphasize that this behavior is accessible by experiment. Such a study would, in our opinion, be of considerable value.

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<sup>1</sup>Reviews are given by 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; K. Binder, *Physica* (Amsterdam) **140A**, 35 (1986); see also, P. Fratzl, J. L. Lebowitz, J. Kalos, and J. Marro, *Acta Metall.* **31**, 1849 (1983), and references therein.

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<sup>10</sup>E. T. Gawłinski, M. Grant, J. D. Gunton, and K. Kaski, *Phys. Rev. B* **31**, 281 (1985).

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<sup>12</sup>We have estimated the distributions of  $R(t)/t^{1/2}$  for model A. For late times, the distribution appears to be independent of time and, as one would expect, non-Gaussian. To the accuracy of our study, the distribution is independent of system size and the ratio of the distribution's height to width is of order unity. This confirms and generalizes earlier studies (see Refs. 9–11).

<sup>13</sup>C. Roland and M. Grant (to be published).

<sup>14</sup>Following other authors (Ref. 6),  $\mathcal{N}(p, t)$  has not been normalized. The distribution  $\mathcal{P}(R, t)$  is normalized for convenience.

<sup>15</sup>It should be noted that negative moments do not exist for the distributions considered in this paper. Thus, only the left-hand side of the  $f$  vs  $\alpha$  curves exist.

<sup>16</sup>L. de Arcangelis, S. Redner, and A. Coniglio (Ref. 5) show how the Legendre transformation can be motivated by making saddle-point approximations to the sums for the moments.

<sup>17</sup>The power spectrum of a random variable  $\eta(t)$  is the Fourier transformation of  $\langle \eta(t) \eta(0) \rangle$ . There may be systematic errors in  $\phi$  comparable to the statistical errors quoted in the text. Standard methods have been used to filter and window the data. See, for example, W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge Univ. Press, Cambridge, 1986), Chap. 12.

<sup>18</sup>We have seen the same qualitative behavior in model B in two dimensions (Ref. 13), where we obtain  $\phi \approx 0.7$ .

<sup>19</sup>Growth exponents are simple fractions because first-order transitions are driven by thermodynamic forces (to minimize the surface free energy of all the domains) and not by fluctuations.

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