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LETTER TO THE EDITOR

Fluctuations and overlap distributions in the kinetics of first-order phase transitions

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Abstract. We introduce analogies between large fluctuations in the scaling regime of the kinetics of first-order transitions, and equilibrium fluctuations in systems with quenched disorder such as spin glasses. The sensitive dependence on initial conditions in the former problem is treated by us in the same manner as quenched disorder in the latter case. We calculate time-dependent overlap functions, which we find are directly related to the order parameter for the transition, and suggest methods to test our results experimentally.

In a first-order phase transition, the macroscopic order parameter changes discontinuously from an initial to a final equilibrium value. The dynamical process by which this occurs has been extensively studied recently [1]. Herein, we shall be concerned with the kinetics of a system prepared in an unstable state by rapid cooling from a high-temperature disordered state to a low temperature T where it is ordered in equilibrium. Small domains of the ordered phase form and then grow to macroscopic size as time increases. The size R of these domains grows in time t following $R \sim t^n$, for late times. For systems whose dynamics is non-conserved, such as binary alloys undergoing an order–disorder transition, $n = \frac{1}{2}$, while for systems whose dynamics is controlled by a conserved mode, such as a binary alloy undergoing phase separation by spinodal decomposition, $n = \frac{1}{3}$. These correspond to the dynamical universality classes of models A and B, respectively [1].

The existence of this diverging length implies scaling in the problem: the system is approximately invariant on changing length and time scales in an appropriate way determined by the growth law. This is shown in figure 1 for configurations from a simulation of a spin-flip kinetic Ising model (model A). Thus, for example, the correlation function of the local order parameter $\psi(x, t)$ obeys $\langle \psi(x, t)\psi(x', t)\rangle \sim G(|x - x'|/t^n)$, for late times. A convenient definition of the amount of order $|\psi|$ in the system is $|\psi| = \langle m^2 \rangle^{1/2}$, where $m \equiv \int dx\, \psi(x)/L^d$, L^d is the volume of the d-dimensional system, and the angular brackets denote an ensemble average (over thermal noise and initial conditions). It is evident from the scaling relation for the correlation function that $|\psi| \sim t^{nd/2}/L^{d/2}$. Thus scaling and the growth exponent probe the manner in which $|\psi|$ changes from 0 to a number of order 1. Furthermore, the hydrodynamic limit, $L \to \infty$, $t \to \infty$, must be taken with care since $0 < |\psi| < 1$, i.e. $|\psi|$ is of order unity, in the scaling regime.

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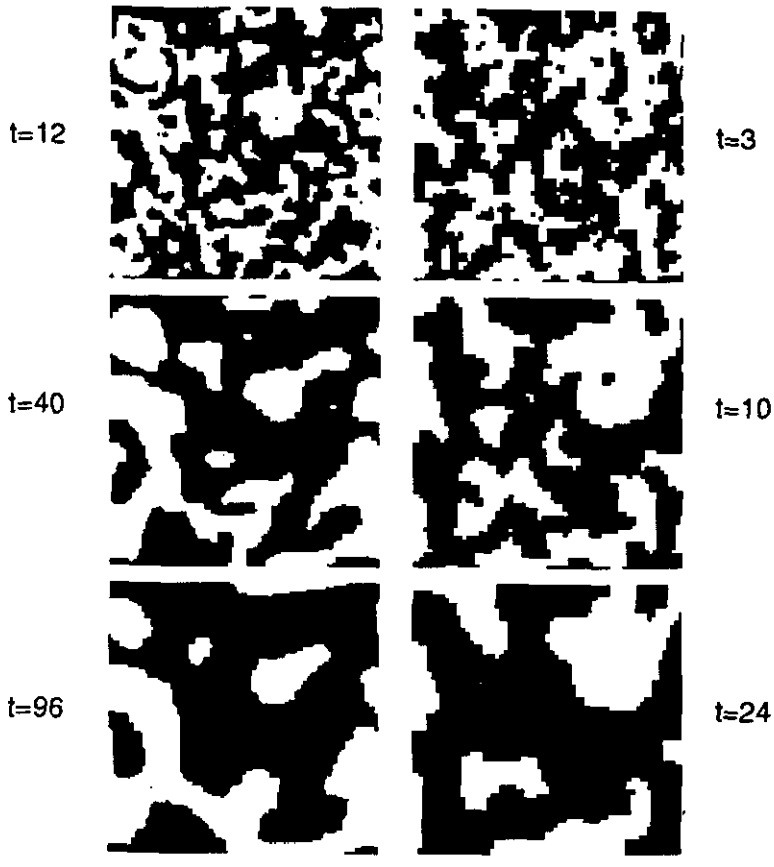

Figure 1. Configurations from a simulation of a 128^2 kinetic Ising model with non-conserved order parameter at $T=0$. Left column: the full system at the times indicated (*in* MCS). Right column: magnification by a factor of 2 of the central 64^2 part of the system at the times indicated, which are a factor of 4 smaller than in the left column. For this choice of time and length scaling factors, the qualitative similarity between left and right columns is evident.

In this letter, we discuss the fluctuations *around* this late-time growth. These fluctuations are evident even in the configurations of figure 1. Although similar, the original and rescaled configurations differ in the shape and location of the domains, so that they are the same only in an average sense. Similar differences appear between configurations evolving from different initial conditions. We will show that these large fluctuations can be usefully described using the language developed to study systems with quenched disorder, such as spin glasses, and other systems with broken ergodicity. We further suggest ways to experimentally probe these phenomena.

That fluctuations are not small [2, 3] can be seen by invoking the law of large numbers: consider a system of volume L^d, which can be partitioned into a large number N of independent subsystems. If X is an extensive quantity, then although $\langle X\rangle = \mathcal{O}(N)$, the quantity $\delta X = X - \langle X\rangle$ has different properties: $\langle(\delta X)^2\rangle^{1/2}/\langle X\rangle = \mathcal{O}(1/N)^{1/2}$. The number of independent parts in the growing system is $N = (L/R(t))^d$, so we find that the relative fluctuations obey

$$\langle(\delta X)^2\rangle^{1/2}/\langle X\rangle = \mathcal{O}(t^n/L)^{d/2} \sim |\psi|. \tag{1}$$

The natural interpretation of this result is as a Ginzburg criterion for the scaling regime [4]: fluctuations are not small in this regime, where $0<|\psi|<1$, and there is no upper critical dimension in which those fluctuations become small.

Noise from the initial conditions is amplified by the long-wavelength instability causing domain growth, unlike the thermal noise in the quenched state which will be irrelevant as soon as domains have grown much larger than the thermal correlation length. At that time the zero-temperature fixed point [5] begins to control the dynamics, the initial stages having been controlled by the infinite-temperature fixed point. The state of the system then acts as an effective initial condition determining the subsequent, essentially deterministic, evolution. This argument can be formalized by considering a Ginzburg–Landau model of the dynamics of phase transitions [1]. At early times, when the state of the system is still close to the high-temperature initial condition, we can safely linearize the equation of motion around $\psi(x, t)=0$. The resulting equation, written in Fourier space $\{k\}$, reads:

$$\dot{\psi}(k, t)=\gamma_k\psi(k, t)+\sqrt{\varepsilon}\ \eta(k, t) \tag{2}$$

where $\eta(k, t)$ is a normalized white noise giving the thermal fluctuations in the quenched state. A typical expression for γ_k is $(k^2)^\alpha(q_c^2-k^2)$, where $\alpha=0$ (or 1) for a non-conserved (or conserved) order parameter. The solution of equation (2) can be written as

$$\psi(k, t)=e^{\gamma_k(t-t_0)}\left(\psi(k, t_0)+\sqrt{\varepsilon}\int_{t_0}^{t}dt'\,e^{\gamma_k(t_0-t')}\eta(k, t')\right) \tag{3}$$

where an *effective* initial condition $\psi(k, t_0)$ has been introduced in terms of the initial condition $\psi(k, t=0)$:

$$\psi(k, t_0)\equiv e^{\gamma_k t_0}\left(\psi(k, 0)+\sqrt{\varepsilon}\int_0^{t_0}dt'\,e^{-\gamma_k t'}\eta(k, t')\right). \tag{4}$$

The validity of these linear expressions breaks down when the nonlinear terms in the original Ginzburg–Landau equation are no longer negligible. This happens [4, 6] at $t_{NL}\approx\ln\varepsilon^{-1}/2\gamma_{k_0}$ where γ_{k_0} is the maximum of γ_k. It is clear from equation (3) that choosing $\gamma_k^{-1}\ll t_0<t_{NL}$, with k in the range of interest, the thermal-noise term will have no effect on the deterministic evolution of $\psi(k, t_0)$. By the range of interest of k we mean the modes sufficiently developed at t_{NL}, that is, those with a positive and not too small γ_k. They will dominate the late linear and the early nonlinear regime. In the fully nonlinear regime, the influence of thermal noise should be still smaller. For the models above, values of t_0 satisfying these inequalities can be found for small noise if q_c^2 is not too small, that is, if the thermal correlation length $\xi\sim 1/q_c$ at the final temperature is much smaller than the system size. The picture which emerges is that thermal noise modifies the initial condition $\psi(k, 0)$ during a time of order γ_k^{-1}, building up the effective initial condition $\psi(k, t_0)$. This effective initial condition is not further modified by noise in a significant way, but is amplified by the instability.

The implication of this is that thermal noise becomes less important as time goes on, whereas noise in the initial conditions is amplified by the instability. Thus, different realizations of the initial conditions give rise to many macroscopic dynamical states which are not 'close', i.e. they are not mixed by small thermal fluctuations. One can give a simple estimate of the number of such dynamical states for $t>t_0$. Say there are two stable phases and N^+ and N^- are the number of domains of each one at time t when domains have grown to a size $R=R(t)$. The total number of domains is

$N \sim \mathcal{O}(L/R)^d$. Using $N \sim \langle m^2 \rangle^{-1}$ we find that $N^{\pm} \sim (N/2)(1 \pm N^{-1/2})$. Thus the number of states at time t is approximately,

$$\mathcal{N} \sim \binom{N}{N^+} \sim 2^{(L/t^n)^d} \sim \exp(\mathcal{O}(1/|\psi|^2)) \tag{5}$$

which is initially large, becoming smaller during the evolution. The existence of a large number of states which cannot be reached by thermal fluctuations is reminiscent of glassy systems. This suggests it would be useful to analyse the fluctuations in the scaling regime using the formalism developed to study systems such as spin glasses and other systems with broken ergodicity. It is worth noting that our analysis is greatly facilitated by the fact that dynamical scaling here is controlled by a zero-temperature fixed point. Thus, for the most part, we need only establish our results at $T = 0$, since finite-temperature corrections involve the non-diverging thermal correlation length ξ.

In broken-ergodicity systems [7], any possible equilibrium probability measure ρ can be decomposed into a convex sum of *pure* states ρ_a:

$$\rho = \sum_a p_a \rho_a \qquad \sum_a p_a = 1 \qquad 0 \leq p_a \leq 1. \tag{6}$$

One of the most important properties characterizing the pure states is the cluster property. A way of formulating it is through the vanishing of all the *connected* correlation functions at long distances. The explicit condition for the two-point connected function $C_a \equiv \langle m^2 \rangle_a - \langle m \rangle_a^2$ in a finite system is $C_a \approx 0$. The notation $\langle \ldots \rangle_a$ means a thermal average with measure ρ_a. The analogy with the dynamics of phase transitions follows by identifying ρ_a with the probability measure of the state of the system $\psi(x, t)$ at time t for a given initial condition (labelled by a). This measure describes the effect of thermal fluctuations on a given initial condition. By identifying p_a in equation (6) with the probability of a particular initial condition a, equation (6) is simply the statement that the complete ensemble average is the combination of thermal and initial-condition average.

The arguments after equation (3) imply that a cluster property must be valid for times larger than t_0, so that at any $t > t_0$, the system is in one of the disjoint *pure* states corresponding to a particular *effective* initial condition in equation (4). In the present context, the cluster property is obviously true for $T = 0$, and we have explicitly checked its validity at $T > 0$ by computer simulation of the two-dimensional spin-flip kinetic Ising model (model A). An initial condition was generated from an infinite temperature ensemble, and evolved at $T = 0.5 T_c$ until a time t_0. Then a number N_a of independent thermal evolutions were calculated from this effective initial condition to obtain C_a. The process was repeated N_0 times to estimate the average over initial conditions $\overline{C_a(t)}$ †. Results for a 128^2 square lattice are shown in figure 2. A small field was applied to the spins at the boundaries to avoid the possibility of spontaneous transitions between the two final equilibrium states. This possibility disappears in an infinite system but can introduce spurious contributions to C_a in finite systems. As shown in figure 2, the maximum value of $\overline{C_a(t)}$ is only a few percent of the final value of the magnetization, thus confirming the cluster property. This result persisted for all the system sizes and times t_0 considered (t_0 from a fraction to a few Monte Carlo steps (MCS) and systems of size from 32^2 to 128^2 sites). The late-time tail of $\overline{C_a(t)}$ vanished with system size as L^{-2}, as expected for two-dimensional equilibrium connected

† The average over initial conditions will be denoted by an overbar.

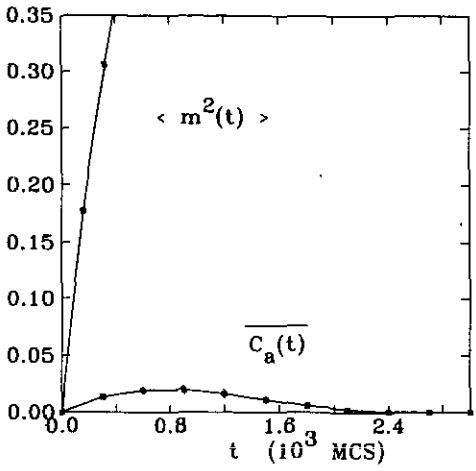

Figure 2. Average connected correlation function $\overline{C_a(t)}$ for a 128^2 Ising system. $T = 0.5T_c$, $t_0 = 1$ MCS, $N_a = 32$, and $N_0 = 10$. The average (over thermal histories and initial conditions) squared magnetization $\langle m(t)^2 \rangle$ is also shown for comparison.

functions, but the zone around the maximum showed a much weaker dependence on system size. Quantification of this dependence will be presented elsewhere.

After showing the splitting of the set of possible states of the system into separate components, we now proceed to the characterization of such splitting. A fundamental quantity which has proven useful in the study of spin glasses [8] and other systems with broken ergodicity [9] is the *overlap* between different states. The natural definition of overlap in the present context is the following

$$q^{ab}(t) = \frac{1}{L^d} \int dx \langle \psi(x, t) \rangle_a \langle \psi(x, t) \rangle_b. \tag{7}$$

It is a random variable depending on the particular pair (a, b) of initial conditions considered.

The overlap distribution, $P(q, t) \equiv \overline{\delta(q - q^{ab}(t))}$, describes the nature of the ensemble of pure states. The average is over all the pairs of independent initial conditions. The form of $P(q, t)$ gives the following information [8, 9]: when it consists of a delta function at zero value of the overlap, no particular decomposition of phase space occurs and the system is disordered, while when it consists of delta functions at values of the overlap related by a symmetry of the system, it describes a situation of broken symmetry. However, if it gives weight to a continuum of values of the overlap, it represents a *glassy* decomposition of phase space, implying the existence of a very large number of macroscopically different states unrelated by symmetry.

As is the case for spin glasses, the definition equation (7) is inconvenient for computational purposes. A preferable expression would be

$$q^{ab}(t) = \frac{1}{L^d} \int dx \, \psi^a(x, t) \psi^b(x, t). \tag{8}$$

In this second expression, $\psi^a(x, t)$ and $\psi^b(x, t)$ are the values of the order parameter at time t, starting from the initial condition a and b, respectively. so that q^{ab} is a random variable depending on the particular pair of initial conditions and their

respective thermal histories. The question of whenever the statistical properties of equation (7) are the same as those of equation (8) is trivial at $T=0$. As mentioned above, this should be sufficient since the dynamics is controlled by a zero-temperature fixed point. In any case, for other temperatures we can follow Parisi's demonstration [10] of the equivalence of both quantities. What is needed for such a demonstration is precisely the cluster property, checked explicitly in figure 2 for the two-point case.

Histograms estimating the distribution $P(q, t)$ from our computer simulations are shown in figure 3. It evolves from a delta function on $q=0$ for $t=0$, corresponding to the disordered initial ensemble, to the final form consisting of delta functions at the values of the equilibrium square magnetization (for our boundary conditions, only one such value exists). Between this initial and final state, $P(q, t)$ is non-trivial, its width describing the diversity of non-equivalent possible evolutions. In the initial stages of the broadening, the system consists of a large number of independent domains, so that we can apply the law of large numbers to (8), showing that $P(q, t)$ is a Gaussian distribution. This Gaussian form is lost when the number of domains in the system begins to significantly decrease. We note that the moments of q^{ab} can be expressed in terms of m-point order parameter correlation functions in the following way:

$$\langle (q^{ab}(t))^m \rangle = \frac{1}{L^{md}} \int \mathrm{d}x_1 \ldots \mathrm{d}x_m \langle \psi(x_1, t) \ldots \psi(x_m, t) \rangle^2. \tag{9}$$

The averages are over thermal noise and initial conditions. In the scaling regime, before boundary conditions become important, we can use the scaling behaviour of the correlation function to show that $\langle (q^{ab}(t))^2 \rangle \approx t^{nd}/L^d \sim |\psi|^2$. A more general result is $\langle (q^{ab}(t))^m \rangle \sim |\psi|^{2(m-1)}$. Thus we find that the overlap function is directly and naturally related to the order parameter for the first-order phase transition.

We have studied other boundary conditions numerically and analytically†, confirming that the results presented here are independent of those boundary conditions. These results will be reported elsewhere. What is important for our conclusions is that the overlap distribution is always broad, as in the case of spin glasses. For

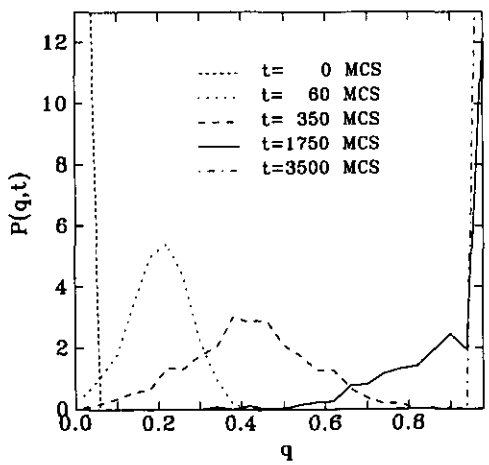

Figure 3. Overlap distribution functions obtained from 640 pairs of independent quenches to $T=0.5T_c$ of a 128^2 Ising system.

† The analytic calculations have been performed using the methods of [11].

periodic boundary conditions, there are additional contributions to the width of the overlap distribution from a fraction of runs which end up in slab states, where one phase forms a stripe stretching across the entire system [12]. We have avoided those states here by applying an infinitesimal field at the boundaries, and plan to study them in a future publication.

An experimental test of our predictions can be made using a novel scattering method of Sutton *et al* [13] using coherent hard x-rays. They observed speckle patterns from the random interconnected structure of Cu_3–Au during the kinetics of an order–disorder transition. We propose the following. By irradiating the material with two coherent x-ray beams, the interference between radiation coming from the two illuminated regions can be analysed. If those regions are separated by a distance much larger than the domain size, one can show that the probability distribution of the quantity $\int d\boldsymbol{k}(I_{ab} - I_a - I_b)/2$ is precisely $P(q, t)$. $I_a(\boldsymbol{k}, t)$, $I_b(\boldsymbol{k}, t)$, and $I_{ab}(\boldsymbol{k}, t)$ are the scattered intensities at angle $\boldsymbol{k}$ when only region a, only region b, or both regions, respectively, are illuminated. Such an experimental measurement of $P(q, t)$ would be of a great deal of interest.

Finally, while our investigation has focused on the kinetics of first-order transitions, we speculate that our results have more general applicability: we expect that three critieria, random initial conditions, a long-wavelength instability, and scaling controlled by a zero-temperature fixed point, imply glassy behaviour in related problems, such as crystal growth [14]. It is also worth mentioning that since scaling and fluctuations here are controlled by attractive fixed points, and that those fluctuations are analogous to those of systems with quenched disorder, the kinetics of first-order transitions provides an example of some of the ideas concerning self-organized criticality [15]. Unlike self-organized criticality, however, scaling in domain-growth kinetics is thought to involve no upper critical dimension, with exponents determined by engineering dimensions.

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References

[1] Gunton J D, San Miguel M and Sahni P S 1983 *Phase Transitions and Critical Phenomena* vol 8, ed C Domb and J L Lebowitz (London: Academic)
Binder K 1986 *Physica* **140A** 35
Fratzl P, Lebowitz J L, Kalos J and Marro J 1983 *Acta Metall.* **31** 1849 and references quoted therein
[2] Roland C and Grant M 1989 *Phys. Rev. Lett.* **63** 551
Yeung C and Jasnow D 1990 *Phys. Rev.* B **42** 10523 and references quoted therein
[3] Sadiq A and Binder K 1984 *J. Stat. Phys.* **35** 617
Gawlinski E T, Grant M, Gunton J D and Kaski K 1985 *Phys. Rev.* B **31** 281
Milchev A, Binder K and Heermann D W 1986 *Z. Phys.* B **63** 521
Coniglio A and Zannetti M 1989 *Europhys. Lett.* **10** 575
Newman T, Bray A J and Moore M A 1990 *Phys. Rev.* B **42** 4514
Klein W 1990 *Phys. Rev. Lett.* **65** 1462
[4] Binder K 1984 *Phys. Rev.* A **29** 341

[5] Mazenko G F and Valls O T 1984 *Phys. Rev.* B **27** 6811
Bray A J 1990 *Phys. Rev. Lett.* **62** 2841
[6] Haake F, Haus J W and Glauber R 1981 *Phys. Rev.* A **23** 3255
[7] Ruelle D 1969 *Statistical Mechanics* (New York: Benjamin)
Parisi G 1988 *Statistical Field Theory* (Reading, MA: Addison-Wesley)
Glim J and Jaffe A 1987 *Quantum Physics* (Berlin: Springer) 2nd edn
Palmer R G 1982 *Adv. Phys.* **31** 669
[8] Mézard M, Parisi G and Virasoro M A 1987 *Spin Glass Theory and Beyond* (Singapore: World Scientific)
[9] Goldbart P and Goldenfeld N 1989 *Phys. Rev.* A **39** 1402, 1412
[10] Parisi G 1983 *Phys. Rev. Lett.* **50** 1946
[11] Ohta, T, Jasnow D and Kawasaki K 1982 *Phys. Rev. Lett.* **17** 1223
Guo H, Zheng Q and Gunton J D 1988 *Phys. Rev.* A **38** 11547
[12] Viñals J and Grant M 1987 *Phys. Rev.* B **36** 7036
[13] Sutton M, Mochrie S G J, Greytack T, Nagler S E, Berman L E, Held G A and Stephenson G B 1991 *Nature* **352** 608
[14] Harris R and Grant M 1990 *J. Phys. A: Math. Gen.* **23** L567
Jasnow D and Viñals J 1989 *Phys. Rev. A* **40** 3864; 1990 *Phys. Rev. A* **41** 6910
Liu F and Goldenfeld N 1990 *Phys. Rev.* A **42** 895
[15] Bak P, Tang C and Wiesenfeld K 1987 *Phys. Rev. Lett.* **59** 381