where B_{ijk} is $3C$ if A_{ij}, A_{jk}, and A_{ik} are all $-\frac{1}{3}$, and is $-C$ otherwise. These three-axis functions clearly have properties (b) and (c), and a little arithmetic confirms property (a).

[10]When $C = 0$ a full set of four-axis functions with properties (a)–(c) can be found. This follows from Appendix A of Ref. 8, where it is shown that the Clauser-Horne inequalities do imply the existence of four-axis functions in the spin-$\frac{1}{2}$ case whenever the distributions p_{ik} have inversion symmetry. Counterexamples to Fine's conjecture in the spin-$\frac{1}{2}$ inversion-symmetric case can, however, be found at the level of five-axis functions. A simple example is given by Eq. (3) with $C = 0$, four values $(0,1,2,3)$ for i and k, and the A_{ik} given by 1 when $ik = 00$, 11, or 22, and given by $-\frac{1}{3}$ otherwise. The Clauser-Horne inequalities continue to hold, as do all the properties of the four-axis functions derived above. By considering the various four-axis functions that can be obtained as marginals of $F_{013,23}(m_1m_2m_3, +-)$ one concludes that this function must vanish for *any* values of m_1, m_2, and m_3. But this is incompatible with any of the distributions $p_{ik}(m,m')$ derivable as marginals from $F_{013,23}$ having nonzero values for all their arguments, as they do.

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Universal Scaling in the Motion of Random Interfaces

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(Received 30 July 1982)

The dynamics of interfaces where the normal component of an interface velocity is proportional to the curvature is studied. The dynamic structure function due to the motion of random interfaces is shown to satisfy a scaling law. The results are compared with Monte Carlo simulations of the kinetics of the order-disorder transition in a quenched system.

PACS numbers: 05.70.Fh, 68.10.Jy

The purpose of this Letter is to study the motion of random interfaces and to apply it to the ordering process in a system quenched below the order-disorder transition temperature. Computer simulations of the kinetics of an order-disorder transition[1,2] and of spinodal decomposition[3] have shown that the time evolution of order-parameter fluctuations from the thermodynamically unstable to the final equilibrium in the ordered phase satisfies a scaling law. Experiments on spinodal decomposition in critical binary fluids[4] have also supported approximately this scaling behavior. We are here concerned with the kinetics of the order-disorder transition where the order parameter is not conserved since in that case the evidence for scaling behavior appears to be strongest.

The authors of Refs. 1 and 2 have analyzed their experimental data by the theory of the motion of domain walls (interfaces) developed by Lifshitz[5] and by Allen and Cahn.[6] Recently Kawasaki[7] has also proposed an interfacial approach to study the scaling law of the spinodal decomposition. One of the main results of the Lifshitz-Allen-Cahn theory is the $t^{1/2}$ prediction of the characteristic length associated with the growth in time t of the domains. This result is meant to apply in the time regime where the value of the order parameter in each domain separated by an interface is assumed to be very close to the equilibrium value. Kawasaki, Yalabik, and Gunton[8] have also obtained the $t^{1/2}$ behavior in the time-dependent Ginzburg-Landau (TDGL) equation utilizing a weak coupling, long-time approximation. However, no theory has been available to obtain the scaling function. In this Letter we wish to derive approximately the explicit form of the scaling function of the correlation of the order-parameter fluctuations as well as the $t^{1/2}$ power law.

We start with the equation of motion of an interface

$$v(\vec{r}(a,t)) = L' K(\vec{r}(a,t)), \tag{1}$$

where $v(\vec{r}(a,t))$ is the normal component of the interface velocity at the position $\vec{r}(a,t)$. K is the total curvature. The position vector $\vec{r}(a,t)$ is defined at each point a on the interface at time t. The proportionality constant L' is given by the bulk Onsager coefficient. Equation (1) was first derived by Allen and Cahn.[6] Kawasaki and Ohta[9] and Bausch, Dohm, Janssen, and Zia[10] have independently obtained Eq. (1) with the fluctuating random force starting from the TDGL equation with a nonconserved order parameter. We have omitted the random force in Eq. (1) since it is not necessary for the present purpose. Note also that Eq. (1) ignores the interaction between interfaces.

A convenient representation is necessary for progress in analyzing Eq. (1). The evolution of the system is given by Eq. (1) along with a geometrical statement specifying the configuration of interfaces. Suppose $z = u(\vec{r}, t)$ specifies a smooth surface with $\vec{r}$ a d-dimensional vector. The intersection of the surface with the plane $z = 0$ determines the interfaces or boundaries. The normal unit vector $\vec{n}(\vec{r}, t)$ on the interface is then given by $\vec{n}(\vec{r}, t) = \nabla u / |\nabla u|$. Note that $du(\vec{r}, t)/dt = 0$ in the frame moving with the interface. Thus one obtains[11] $v(\vec{r}, t) = -(1/|\nabla u|)\partial u(\vec{r}, t)/\partial t$. The formula[12] $K = -\nabla \cdot \vec{n}$ then transforms Eq. (1) as

$$\frac{\partial}{\partial t} u(r,t) = L' \sum_{\alpha,\beta} n^{\alpha}(\vec{r},t) n^{\beta}(\vec{r},t)(\delta_{\alpha\beta}\nabla^2 - \nabla^{\alpha}\nabla^{\beta}) u(\vec{r},t), \tag{2}$$

where $\alpha, \beta = 1, 2, \ldots, d$ the bulk spatial dimensionality. The configuration of interfaces is specified by the contour surfaces with $u(\vec{r}, t) = 0$ of the solution of Eq. (2).

Various quantities which are accessible experimentally can be represented in terms of $u(\vec{r}, t)$. The area density of the interfaces is given by

$$A(t) \equiv \int da \langle \delta(\vec{r} - \vec{r}(a)) \rangle = \langle |\nabla u| \, \delta(u(\vec{r}, t)) \rangle, \tag{3}$$

where the average $\langle\ \rangle$ is taken over the initial configuration of the interface. The integral over a is understood to run over all interfaces. The dynamic scattering function $S(\vec{q}, t)$ of the order-parameter fluctuations due to the motion of interfaces may be written as[13]

$$S(q,t) = (\Delta M/q)^2 \int d^d r_1 \sum_{\alpha} \langle [\nabla^{\alpha} u(\vec{r}_1, t)][\nabla^{\alpha} u(\vec{r}_2, t)] \delta(u(\vec{r}_1, t)) \delta(u(\vec{r}_2, t)) \rangle \exp[i\vec{q}(\vec{r}_1 - \vec{r}_2)], \tag{4}$$

where ΔM is the gap of the order-parameter profile at an interface.

Here we wish to solve Eq. (2) in an approximate manner. That is, we replace $n^{\alpha}(\vec{r}, t) n^{\beta}(\vec{r}, t)$ by the average $\langle n^{\alpha}(\vec{r}, t) n^{\beta}(\vec{r}, t) \rangle$ so that the resulting equation is linear in u. The difference $n^{\alpha}n^{\beta} - \langle n^{\alpha}n^{\beta} \rangle$ can be treated by a perturbation expansion. Thus one obtains up to the lowest order

$$\partial u(\vec{r}, t)/\partial t = (L'/d)(d-1)\nabla^2 u(\vec{r}, t), \tag{5}$$

where we have assumed that the system is isotropic, i.e., $\langle n^{\alpha}n^{\beta} \rangle = (1/d)\delta_{\alpha\beta}$.

In order to evaluate the averages in (3) and (4) one needs the initial probability distribution of the field $u(\vec{r}, t)$. We shall explore the consequences of an assumption that the distribution is of Gaussian form with the average $\langle u \rangle \equiv U$, and that the fluctuations of $u(\vec{r}, t)$ are spatially uncorrelated:

$$G_q(t=0) \equiv \int d^d r_1 \langle \delta u(\vec{r}_1) \delta u(\vec{r}_2) \rangle \exp[i\vec{q}(\vec{r}_1 - \vec{r}_2)] = C, \tag{6}$$

where $\delta u(\vec{r}) = u(\vec{r}) - U$. Equation (5) with (6) enables us to calculate the time evolution of $G_q(t)$ or its inverse Fourier transform $G(\vec{r}, t)$ as

$$G(\vec{r}, t) = C(8\pi L t)^{-d/2} \exp[-r^2/8Lt], \tag{7}$$

where $L = L'(d-1)/d$. Note that since $\langle n^{\alpha}(\vec{r}, t) \times \nabla^{\alpha}\nabla^{\beta} u(\vec{r}, t) \rangle = 0$, Eq. (5) turns out to be correct to first order in the expansion.

The area density (3) in $d = 3$ is calculated as follows:

$$A(t) = \left\langle \frac{(\nabla u)^2}{|\nabla u|} \delta(u(\vec{r}, t)) \right\rangle = 4\pi \int \frac{d^d q}{(2\pi)^3} \frac{1}{q^2} \left\langle (\nabla u)^2 \delta(u(\vec{r}, t)) e^{i\vec{q}\cdot\vec{\nabla} u} \right\rangle. \tag{8}$$

After taking the Gaussian average over δu one obtains

$$A(t) = \frac{2\sqrt{3}}{\pi} (4Lt)^{-1/2} \exp[-(U^2/2C)(8\pi L t)^{3/2}]. \tag{9}$$

In two dimensions the coefficient $2\sqrt{3}/\pi$ is replaced by $1/\sqrt{2}$ and $(8\pi Lt)^{3/2}$ by $(8\pi Lt)^1$. The $t^{-1/2}$ dependence, which is independent of d, has been derived by a dimensional argument.[6] In the kinetics of the order-disorder transition the only characteristic length of the problem (apart from the width of the interface) is $(4Lt)^{1/2}$ which represents the growth of the ordered domains. Therefore (see below) we restrict ourselves to the case $U=0$. At $t=0$, $A(t)=\infty$ which implies that the initial interfaces take a random configuration with infinite folding.

The scattering function (4) is also calculated in a similar way. With use of the correlation function (6) some algebraic manipulations yield

$$\bar{S}(q,t)=(4Lt)^{d/2}I(q(4Lt)^{1/2}), \tag{10}$$

where

$$I(x)=\frac{2}{\pi}\frac{(2\pi)^{d/2}}{x}\int_0^\infty dR\,R^d[\exp(R^2)-1]^{-1/2}(xR)^{1-d/2}J_{d/2}(xR) \tag{11}$$

with J_ν the Bessel function of the first kind. Note that C defined in (6) has disappeared in (10). The scattering function $\bar{S}(q,t)$, which is normalized such that $\int d^dq/(2\pi)^d\bar{S}(q,t)=1$, is of the scaling form observed by computer simulations.[1,2] The scaling function $I(x)$ was evaluated numerically. Figures 1 and 2 display the results in three and two dimensions, respectively. The new variable z of the function $\hat{I}(z)=I(x)$ has been introduced in such a way that $\hat{I}(z)=\hat{I}(0)[1-z^2/d+O(z^4)]$, where $\hat{I}(0)\simeq 10.47$ for $d=3$ and $\hat{I}(0)=2\pi\ln 2\simeq 4.355$ for $d=2$. The proportionality constant c with $z=cx$ is given by $c\simeq 1.206$ in three dimensions and $c=\{[(\ln 2)^2+\pi^2/12]/2\ln 2\}^{1/2}\simeq 0.9695$ in two dimensions. The values of $\hat{I}(0)$ and c for $d=3$ were obtained numerically.

For large values of z, $\hat{I}(z)$ is found from (11) to exhibit algebraic decay,

$$\hat{I}(z)\simeq z^{-\omega}, \tag{12}$$

with $\omega=d+1$. On the other hand, a rough estimation of the exponent from the experiments[1,2] gives $\omega\simeq 2.6$ and 3.5 in two and three dimensions, respectively. The d dependence is consistent with (12) although each value is somewhat smaller.

The complete comparison of $\hat{I}(z)$ itself without any adjustable parameters seems impossible at present. The finite-size effect in simulations makes it difficult to estimate the scaled variable z from the data. Note also that the second moment $[x^2]\equiv\int d^dx\,x^2I(x)/\int d^dx\,I(x)$, which has been used to scale $kt^{1/2}$ in Ref. 2, is not well defined here without introduction of an ultraviolet cutoff because of the asymptotic behavior of (12). Nevertheless if one adjusts the unknown proportionality constant between z and $kt^{1/2}$ as $z/kt^{1/2}\simeq 0.1$ and 0.24 in two and three dimensions, respectively, $\hat{I}(z)$ in Figs. 1 and 2 show good agreement with the experiments. In the plot the vertical axis was not adjusted since the values of $\hat{I}(0)$ coincide roughly with those of the experiments.

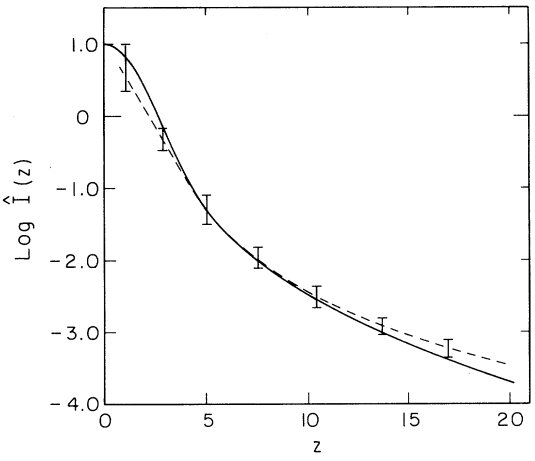

FIG. 1. The scaling function in three dimensions. The full line exhibits the function $\hat{I}(z)$. The data in Fig. 3 of Ref. 1 are shown by the broken line with the bars indicating typical scatter.

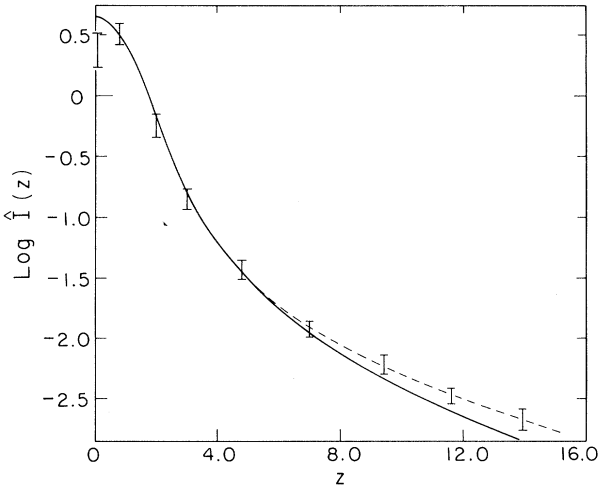

FIG. 2. The scaling function in two dimensions. The experimental data were taken from Fig. 6(a) of Ref. 2. The meanings of the lines are same as those of Fig. 1.

The present results depend on the choice of the initial distribution of $u(\vec{r}, t)$. As is shown in (9) if $U \neq 0$, the $t^{1/2}$ power law of the area density breaks down in the time regime $U^2(8\pi Lt)^{3/2}/2C \gtrsim 1$.[14] Other choices of the correlation function (6) would also alter the results. However, note from (10) that $q \sim O((Lt)^{-1/2})$ and $u(\vec{r}) \sim O((Lt)^{-d/4})$. Therefore a deviation from the Gaussian distribution of the field $u(\vec{r})$ and the q dependence of $G_q(t=0)$ vanish asymptotically. Thus as long as $G_q(t=0)$ for $q=0$ is finite the result (14) with (15) remains unchanged. In some sense the situation seems similar to the determination of the critical surface on the initial parameter space in the renormalization-group theory of phase transitions. Although the agreement of the scaling form with the simulations is encouraging, further study is necessary to clarify this point.

In summary we have studied the statistical properties of interface motion with a random initial configuration. As an example of phenomena which are governed by random interfaces we have discussed the time evolution of fluctuations in the order-disorder transition of a quenched system. The universal scaling form of the scattering function has been obtained and compared to the experiments.

One of us (T.O.) is grateful to Bharat B. Pant for assistance with the numerical computations. This work was completed during a visit by one of us (T.O.) to the Department of Physics and Astronomy, University of Pittsburgh, which he thanks for partial support. The support of the National Science Foundation through the Division of Materials Research is also gratefully acknowledged.

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[14]However, note the following invariance. An observable quantity must be independent of choice of the contour surfaces $u(\vec{r}, t) = u_0 = \text{const}$. This requires that $U = u_0$. In the present case $u_0 = 0$ so that $U = 0$.