

## Temperature dependence of the dynamics of random interfaces

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We study the effects of thermal-noise-induced roughening on the dynamical evolution of random interfaces. This is done using the Allen-Cahn equation with noise, which is appropriate for a system with a nonconserved order parameter. The proportionality constant in the characteristic growth law of domains is renormalized by a temperature-dependent factor. Two different initial configurations are considered. Firstly, a random configuration of interfaces in  $d$  dimensions is analyzed using the linearization scheme of Ohta, Jasnow, and Kawasaki. The results suggest that the linearization breaks down at high temperatures; theoretical predictions for low and intermediate temperatures could be tested experimentally. Secondly, a two-dimensional circular domain is analyzed to first order in a low-temperature perturbation series. This is done in the manner of Safran, Sahni, and Grest. However, we obtain results which differ from those of Safran *et al.* Our results are consistent with those we obtain for the random configuration of interfaces.

### I. INTRODUCTION

There has been much interest recently in the time evolution of a one-phase (disordered) system quenched into the two-phase (ordered) region of a phase diagram.<sup>1</sup> This involves one of the fundamental problems in statistical physics: the behavior of systems far from equilibrium. In the intermediate time regime following the quench, convoluted random interfaces form. The evolution of these interfaces causes domain growth as the system progresses towards equilibrium. In this paper we will be concerned with the temperature dependence of the interfacial dynamics. We consider systems where the order parameter is not conserved.

It is now generally accepted that the characteristic length  $\bar{R}$  in such systems grows following a  $t^{1/2}$  growth law. Kawasaki, Yalabik, and Gunton<sup>2</sup> found this in their analysis of the time-dependent Ginzburg-Landau equation in a long-time, weak-coupling limit. Allen and Cahn<sup>3</sup> derived a deterministic equation of motion for the interfaces in a system with a nonconserved order parameter. Their equation of motion is given by

$$v = L'K, \quad (1.1)$$

where  $v$  is the velocity orthogonal to a point on the interface,  $K$  is the curvature at that point, and  $L'$  is a diffusion coefficient. Thus we see that the  $t^{1/2}$  growth law results because interfacial motion is diffusively driven by the curvature; the diffusion coefficient is dimensionally  $\text{cm}^2/\text{sec}$ , so any characteristic length constructed from  $L'$  will have the  $t^{1/2}$  time dependence, i.e.,

$$\bar{R} \sim \sqrt{L't}. \quad (1.2)$$

This growth law has been observed in many experimental studies.<sup>4-6</sup>

Recently, in an interesting paper, Ohta, Jasnow, and Kawasaki<sup>7</sup> have solved the Allen-Cahn equation using a linearization scheme based upon the isotropy of an initial-

ly random configuration of interfaces. They obtained an explicit solution for the area density in both two and three dimensions. Making the further assumption of a sharp, step-function-like order-parameter profile at the interface, they obtained the dynamic structure factor  $\mathcal{S}(k, t)$ , where  $k$  is the wave number. The structure factor was found to be *self-similar*, scaling with the characteristic length  $\sqrt{L't}$ . The theoretical expression gave a good, one-parameter fit to the computer experiments of Phani *et al.*<sup>5</sup> and Sahni *et al.*<sup>6</sup> The scaling function of Ohta *et al.* is closely related to that obtained earlier in the theory of Kawasaki *et al.*<sup>8,2</sup>

Of course, if the physical system described by Eq. (1.1) is to approach equilibrium as  $t \rightarrow \infty$ , the equation of motion must include a random force which obeys a fluctuation-dissipation relation. The form of the thermal fluctuating noise term has been independently given by Kawasaki and Ohta<sup>9</sup> and by Bausch, Dohm, Janssen, and Zia<sup>10</sup>; see Eqs. (2.18) and (2.19). Note that the fluctuation-dissipation relation involves the surface tension  $\sigma$ . This is because the equilibrium free energy is approximately given by the product of the surface tension  $\sigma$  and the surface area  $A$ . This is the dynamical-interface model which is discussed further in Sec. II.<sup>9</sup> The Allen-Cahn equation, together with thermal fluctuating noise, then comprises the dynamical-interface model.

Safran, Sahni, and Grest<sup>11</sup> (SSG) have analyzed the effect of thermal noise on the two-dimensional growth dynamics of circular domains. Their analysis *does not* begin with the Allen-Cahn equation with noise.<sup>12</sup> Their fundamental equation of motion can be obtained from the Allen-Cahn equation. Firstly, one prescribes that contributions due to tilting the interface are removed, regardless of the order of those contributions. Secondly, the surface tension in the fluctuation-dissipation relation is replaced by a rough approximation for the mean-field surface tension.<sup>13</sup> These replacements are, of course, not valid for the Allen-Cahn equation. The tilt terms are required for Euclidean invariance<sup>14</sup> of the Allen-Cahn equation.<sup>9,10</sup>

Further, the tilt terms and the surface tension are required for the system to approach equilibrium.<sup>9</sup> The SSG equation of motion may, however, have a justification independent of the Allen-Cahn equation with noise.<sup>12</sup> SSG obtain the temperature-dependent corrections to Eq. (1.2), i.e., they give

$$\bar{R}^2 \propto L' i \alpha, \quad (1.3)$$

with an explicit expression for  $\alpha = \alpha(k_B T/J)$ , where  $k_B$  is Boltzmann's constant,  $T$  is the temperature, and  $J$  is the interaction constant of, say, the Ising model. Their perturbative analysis gives  $\alpha$  to linear order in  $k_B T/J$ . Somewhat surprisingly, they find good agreement with computer experiment for  $k_B T/J > 1$ . The authors note that systematic problems with the computer simulations preclude a comparison of theory and experiment for  $k_B T/J < 1$ . We discuss their treatment in more detail in Secs. IV and V.

In this paper we will analyze the temperature dependence of the dynamics of random interfaces using the Allen-Cahn equation with thermal noise. We obtain the temperature-dependent roughening corrections to the growth law for interfaces with random initial configurations (using the linearization scheme of Ohta *et al.*<sup>7</sup>), and interfaces with circular initial configurations (using a perturbation treatment analogous to that of Safran *et al.*<sup>11</sup>).

Our results can be anticipated as follows. We note that domain growth occurs when the random convoluted interfaces "straighten out" by  $v = L'K$ . The thermal noise will roughen the interface and tend to make the interfaces stay random, thus effectively slowing down domain growth.<sup>15</sup> If we expand the characteristic length in a Taylor series about zero temperature we obtain linear corrections of order  $k_B T$ . Further, the temperature will be scaled to the (zero-temperature) surface tension  $\sigma$ , as in  $k_B T/\sigma a^{d-1}$ , where  $a$  is the lattice constant and  $d$  is the dimensionality. This is plausible physically: Since  $\sigma a^{d-1}$  is the effective energy of an interface it scales with the temperature. A large zero-temperature  $\sigma$  is equivalent to a low  $k_B T$ , and vice versa, as far as the roughening effects are concerned. Thus we expect

$$\bar{R} \rightarrow \bar{R} [1 - O(k_B T/\sigma a^{d-1}) + \dots].$$

We now outline the remainder of this paper. In Sec. II we review the derivation of the Allen-Cahn equation with noise.<sup>3,9</sup> This is done for a general curvilinear coordinate system.

In Sec. III we generalize the treatment of Ohta, Jasnow, and Kawasaki<sup>7</sup> to include the contribution of the thermal fluctuating noise. We calculate the area density and give the temperature-dependent corrections to all orders in  $k_B T$ , which are congruent with the linearization scheme of Ohta *et al.* Our results are given by Eqs. (3.28) and (3.29). It appears that the linearization scheme breaks down at high temperatures.

In Sec. IV we analyze the growth of circular domains, in the manner of Safran, Sahni, and Grest,<sup>11</sup> using the Allen-Cahn equation with noise. Contributions due to tilt terms (required to ensure the Euclidean invariance and the approach to equilibrium of the equation of motion<sup>9,10</sup>) and the surface tension (also required for the approach to

equilibrium<sup>9</sup>) in the Allen-Cahn equation, give temperature-dependent effects which differ from those predicted by Safran *et al.* Our results, given by Eq. (4.20), are, however, consistent with those of Sec. III. We discuss our results in Sec. V and draw conclusions from our study.

## II. THE ALLEN-CAHN EQUATION

Before obtaining the new results of this paper, given in the next two sections, we review the derivation of the Allen-Cahn equation. This will be our fundamental equation of motion. We follow the treatments of Allen and Cahn,<sup>3</sup> and Kawasaki and Ohta.<sup>9</sup>

Our starting point consists of the following set of equations. The time evolution of the nonconserved order parameter,  $\mathcal{M}(\vec{r}, t)$ , is determined by the Langevin equation,<sup>1</sup>

$$\frac{\partial \mathcal{M}(\vec{r}, t)}{\partial t} = -L'' \frac{\delta F[\mathcal{M}]}{\delta \mathcal{M}} + \zeta(\vec{r}, t), \quad (2.1a)$$

where  $F$  is the free-energy functional and  $L''$  is a kinetic coefficient. The random force  $\zeta$  obeys Gaussian-Markov statistics through the fluctuation-dissipation relation,

$$\langle \zeta(\vec{r}, t) \zeta(\vec{r}', t') \rangle = 2k_B T L'' \delta^d(\vec{r} - \vec{r}') \delta(t - t'), \quad (2.1b)$$

where  $d$  is the dimensionality. Equation (2.1) is constructed to drive the nonconserved order parameter to equilibrium ( $\partial \mathcal{M} / \partial t = 0$ ) through changes in the free energy  $F$ .

The free-energy functional is assumed to be of the form<sup>1</sup>

$$F = \int d^d r \left[ \frac{1}{2} C (\vec{\nabla} \mathcal{M})^2 + f(\mathcal{M}) \right], \quad (2.2)$$

where  $C$  is a constant and  $f(\mathcal{M})$  is the "bulk" free-energy density. All nonuniformity in  $\mathcal{M}$  is determined by the square-gradient ansatz in the integrand of Eq. (2.2).

In equilibrium, the order parameter  $\mathcal{M}$  will be equal to its time-independent equilibrium value which we shall denote by  $M$ . If equilibrium is characterized by a coexisting two-phase system separated by a flat diffuse interface located about, say,  $x = 0$ , then Eq. (2.2) gives

$$-C \frac{d^2 M(x)}{dx^2} + \frac{\partial f}{\partial M} = 0. \quad (2.3)$$

The equilibrium surface tension can also be obtained from Eq. (2.2),

$$\sigma = C \int dx \left[ \frac{dM(x)}{dx} \right]^2. \quad (2.4)$$

Far from equilibrium, in the intermediate-time regime following the quench, we will not have a flat interface. Instead the interface will be convoluted, and essentially random. It is useful to introduce a new curvilinear coordinate system,<sup>9</sup>  $\vec{u} = (u_1, \dots, u_d)$ . This coordinate system is constructed so that the interface is given by<sup>16</sup>

$$u_1(\vec{r}, t) \equiv 0. \quad (2.5)$$

We will require that the set of surfaces  $\vec{u}$  be mathematically smooth. This means that the interface must be "gently" curved. Explicitly, we must have  $w/R_c \ll 1$ , if

$w$  is the thickness of the interface and  $R_c$  is the radius of curvature at some point. As long as we are not too close to the critical temperature this condition is satisfied.

Our new coordinate system is given by<sup>17</sup>

$$\vec{u}(\vec{r}, t) = (u_1(\vec{r}, t), \dots, u_d(\vec{r}, t)). \quad (2.6a)$$

Unit vectors to the surfaces are given by

$$\hat{n}_i = h_i \vec{\nabla} u_i \quad (2.6b)$$

for  $i=1$  to  $d$ , where

$$h_i = |\vec{\nabla} u_i|^{-1} \quad (2.6c)$$

is the differential length in the  $u_i$  direction. The set of surfaces  $u_1(\vec{r}, t)$  is defined to be consistent with the *physical* surface  $u_1=0$ ; see Eq. (2.5). The other surfaces,  $u_2, \dots, u_d$ , are defined by the second-order differential equations,

$$\hat{n}_i \cdot \hat{n}_j = \delta_{ij}, \quad (2.6d)$$

for  $i, j=1$  to  $d$ . The curvature  $K$  of a given  $u_1$  surface is

$$K = -\vec{\nabla} \cdot \hat{n}_1. \quad (2.6e)$$

The differential element and Dirac  $\delta$  function are, respectively, given by

$$d^d r = \prod_{i=1}^d h_i du_i \equiv h_1 du_1 d^{d-1} S \quad (2.6f)$$

and

$$\delta^d(\vec{r}) = \prod_{i=1}^d \frac{\delta(u_i)}{h_i} \equiv \frac{\delta(u_1)}{h_1} \delta^{d-1}(\vec{S}).$$

$\vec{S}$  is the vector determining positions on the  $(d-1)$ -dimensional surface where  $u_1$  is constant. Again we note that, by construction,  $u_1=0$  is the only physical surface.

We now consider the Langevin equation, Eq. (2.1). We will assume that the order parameter  $\mathcal{M}$  is approximately given by its equilibrium value  $M$ , if  $M$  is evaluated at the convoluted surface  $u_1(\vec{r}, t)$  rather than the flat surface  $x$ , i.e.,<sup>3,9</sup>

$$\mathcal{M}(\vec{r}, t) \simeq M(u_1(\vec{r}, t)). \quad (2.7)$$

This is reasonable for a gently curved interface. An explicit demonstration of the negligible contributions of higher-order corrections to Eq. (2.7) is given by Kawasaki and Ohta.<sup>9</sup> Using Eq. (2.7) in Eqs. (2.1) and (2.2) we obtain

$$\frac{\partial u_1}{\partial t} \frac{dM(u_1)}{du_1} = -L' \left[ -C \nabla^2 M(u_1) + \frac{\partial f}{\partial M} \right] + \xi. \quad (2.8)$$

But from Eqs. (2.3) and (2.4),

$$\frac{\partial f}{\partial M(u_1)} = C \frac{d^2 M(u_1)}{(h_1 du_1)^2} \quad (2.9)$$

and

$$\sigma = C \int du_1 h_1 \left[ \frac{dM(u_1)}{h_1 du_1} \right]^2. \quad (2.10)$$

Thus in Eq. (2.8),

$$h_1 \frac{\partial u_1}{\partial t} \frac{dM(u_1)}{h_1 du_1} = -L' \left[ -\nabla^2 M(u_1) + \frac{d^2 M(u_1)}{(h_1 du_1)^2} \right] + \xi, \quad (2.11)$$

where

$$L' \equiv CL'' \quad (2.12)$$

is a diffusion coefficient. Now, from standard texts,<sup>17</sup> we obtain

$$h_1 \frac{\partial u_1}{\partial t} \frac{dM(u_1)}{h_1 du_1} = -L' \left[ K \frac{dM(u_1)}{h_1 du_1} \right] + \xi, \quad (2.13)$$

where  $K$  is the curvature defined in Eq. (2.6e).

The only physical realization of Eq. (2.13) is at the interface located about  $u_1=0$ . We can *project* the dynamics of Eq. (2.13) onto this physically relevant region through the operator  $\mathcal{P}$  defined by

$$\mathcal{P}g = \frac{1}{M(\infty) - M(-\infty)} \int du_1 h_1 \frac{dM}{h_1 du_1} g. \quad (2.14)$$

$K$  and  $h_1 \partial u_1 / \partial t$  are approximately constant over the gently curved interface. Thus, using the projection operator  $\mathcal{P}$  on Eq. (2.13), we obtain

$$-h_1 \frac{\partial u_1}{\partial t} = L'K + \eta, \quad (2.15)$$

where

$$\eta \equiv -\frac{C}{\sigma} \int du_1 h_1 \frac{dM}{h_1 du_1} \xi. \quad (2.16)$$

Since the velocity normal to the interface at  $u_1=0$ ,  $v$ , is given by

$$v = -h_1 \frac{\partial u_1}{\partial t} \quad (2.17)$$

(from continuity of flux at  $u_1=0$ ), we then have

$$v = L'K + \eta, \quad (2.18)$$

at  $u_1=0$ . The fluctuation-dissipation relation for  $\eta$  is found from Eqs. (2.1b), (2.10), and (2.16) to be

$$\langle \eta(\vec{S}, t) \eta(\vec{S}', t') \rangle = \frac{2k_B T L'}{\sigma} \delta^{d-1}(\vec{S} - \vec{S}') \delta(t - t'). \quad (2.19)$$

$\vec{S}$  is the vector determining positions on the  $u_1=0$  surface [from Eq. (2.6f)]. Equations (2.18) and (2.19) are the Allen-Cahn equation with thermal noise.<sup>3,9,10</sup>

We note the following about the above. The results are valid provided the thickness of the interface is much less than the radius of curvature of the interface. Note that interactions between interfaces are not considered. A free energy featuring higher-order gradient contributions than those given in Eq. (2.2) might lead to higher-order terms in the curvature  $K$  in Eq. (2.18). Note that it has not been necessary to specify a form of the free-energy density  $f(\mathcal{M})$  in Eq. (2.2).

Let us discuss the extent to which this continuum theory describes a physical system on a lattice. Any possi-

ble differences will be most important at low temperatures. Since the interfacial dynamics occur at  $u_1=0$ , it is implicit that we have averaged over the interface. Therefore, the smallest length in our treatment is the thickness of the interface  $w$ . This length, however, can be no less than the lattice constant  $a$ , because of the underlying lattice dynamics which the field theory herein approximately describes. Ultraviolet cutoffs act to incorporate the physics of the lattice, albeit in a simplified manner. In Kawasaki and Ohta's field-theoretic derivation<sup>9</sup> of the Allen-Cahn equation, they consider the limit of an infinitely deep potential well. This corresponds to an infinitely thin interface,  $w \rightarrow 0$ . In their notation  $\tau=1/w^2$ , so  $\tau \rightarrow \infty$ . However, in agreement with our remarks above, we will require that  $\tau_{\max}=1/a^2$  if we scale  $\tau$  to the lattice constant,  $\tau_{\max}=1$ . This means that  $\tau_{\max}$  (macroscopic length)<sup>2</sup>  $\gg 1$ . Strictly speaking then, there will be corrections to the Allen-Cahn equation of order  $1/\tau_{\max}$ . These could be incorporated through the formalism of Kawasaki and Ohta,<sup>9</sup> where a systematic procedure for incorporating large but finite  $\tau$  corrections is presented. It may be preferable, though, to obtain a discrete lattice-dynamical analog of the Allen-Cahn equation. We will not consider these possible extensions further, at this time.

The Allen-Cahn equation is rather subtle. Equations (2.18) and (2.19) determine the time-dependent behavior of a stochastically driven coordinate system, defined in Eq. (2.16). Thus, for example, the  $\delta$ -function correlation on the surface in Eq. (2.19) is an implicit function of  $u_1$  from Eq. (2.6d). The equation of motion is in a Euclidean invariant form<sup>14</sup>; this is a necessary physical condition. The tilt term  $h_1$  is required to ensure this.<sup>9,10</sup> Euclidean invariance is satisfied here through the orthogonality condition, Eq. (2.6d). Finally we note that the surface tension appearing in Eq. (2.14) is thermodynamically required if the system is to approach equilibrium.<sup>9</sup> This is more readily seen in the equivalent Fokker-Planck representation of the dynamics.

We transform the Langevin equation for the surface [Eqs. (2.18) and (2.19)] into a Fokker-Planck equation for the complete distribution function  $P$  by standard means.<sup>18</sup> Following Kawasaki and Ohta<sup>9</sup> we obtain

$$\frac{\partial}{\partial t} P(u_1, t) = L' \int d^{d-1}S \frac{\delta}{h_1 \delta u_1(\vec{S})} \left[ K + \frac{k_B T}{\sigma} \frac{\delta}{h_1 \delta u_1(\vec{S})} \right] \times P(u_1, t). \quad (2.20a)$$

The time-dependent average of a quantity is given by the functional integral,

$$\langle g(u_1) \rangle_t \equiv \int_-^- \int \mathcal{D}u_1 g(u_1) P(u_1, t). \quad (2.20b)$$

From Eq. (2.20a) it can be shown that the equilibrium distribution  $P_{\text{eq}}$ , where  $\partial P_{\text{eq}}/\partial t=0$  is given by

$$P_{\text{eq}} \propto \exp(-F_{\text{eq}}/k_B T),$$

where

$$F_{\text{eq}} = \sigma \int d^{d-1}S.$$

This is the interface free energy.<sup>9</sup> This model free energy neglects contributions from the bulk domains, recognizing that the crucial physics of the system is determined by the surface free energy. Both the surface tension and the tilt terms (which are implicit in the  $d^{d-1}S$  differential) are necessary if the equation of motion is to be consistent with thermal equilibrium. In the next two sections we will use the Allen-Cahn equation to analyze the temperature dependence of the kinetics of random interfaces.

### III. AN INITIALLY RANDOM CONFIGURATION OF INTERFACES

In the following section, we extend the work of Ohta, Jasnow, and Kawasaki<sup>7</sup> (OJK) to include the effects of thermal noise. OJK solved the deterministic Allen-Cahn equation assuming a random initial configuration of interfaces. If the interfaces are sufficiently convoluted, the physical system is essentially isotropic; the Allen-Cahn equation then becomes linear. One of our purposes is to investigate the validity of the OJK linearization as temperature effects become important. We find that the linearization is most valid at low temperatures and that it apparently breaks down at high temperatures.

The second moment of the Fokker-Planck equation, Eq. (2.20), gives

$$\begin{aligned} \frac{\partial}{\partial t} \langle u(\vec{S})u(\vec{S}') \rangle_t &= -L' \left\langle \frac{K(\vec{S})}{h_1(\vec{S})} u_1(\vec{S}') \right\rangle_t - L' \left\langle u_1(\vec{S}) \frac{K(\vec{S}')}{h_1(\vec{S}')} \right\rangle_t \\ &+ \frac{2k_B T}{\sigma} L' \left\langle \frac{1}{h_1^2(\vec{S})} \delta^{d-1}(\vec{S}-\vec{S}') \right\rangle_t. \end{aligned} \quad (3.1)$$

Recalling Eq. (2.6) we see that this is a very complicated nonlinear equation coupled to higher-order moments in the Fokker-Planck equation.

OJK have proposed a physically appealing approximation to this equation which results in great simplifications. Following Ref. 7 we will assume that the initial configuration of interfaces is random, i.e.,

$$\langle u_1(\vec{S})u_1(\vec{S}') \rangle_{t=0} = \gamma \delta^d(\vec{r}-\vec{r}'), \quad (3.2)$$

where  $\gamma$  is a constant. This is precisely the experimental situation following a quick quench of a one-phase, disordered phase into the two-phase, ordered region of a phase diagram. We also assume that, for all times of interest, the  $d$ -dimensional space is isotropic, i.e.,

$$\langle f(\vec{S})g(\vec{S}') \rangle_t = \langle fg \rangle_t(\vec{r}-\vec{r}'). \quad (3.3)$$

This means that the physical surface,  $u_1=0$ , is sufficiently convoluted that there is no unique symmetry in the system.

This is a strong assumption. It is reasonable in the intermediate-time regime following a quench. However, we note that this approximation cannot be true over long times as equilibrium is approached. Equilibrium will involve a breaking of symmetry and special directions will

exist. For example,<sup>1</sup> in equilibrium there is a symmetry-breaking soliton solution of the order parameter [from Eq. (2.3) with, say, an “ $M^4$ ” version of  $f(M)$ ]. This is no longer a solution given the isotropy assumption. Further, there can be no solitary wave solution of the dynamical Allen-Cahn equation given this assumption. See Chan’s discussion in Ref. 3.

If we also assume that all averages are Gaussian, and that  $\langle u_1^2 \rangle_t$  and  $\langle 1/h_1^2 \rangle_t$  are not zero, we obtain

$$\langle u_1(\vec{r}) \vec{\nabla} u_1(\vec{r}) \rangle_t = \langle \vec{\nabla} u_1(\vec{r}) \vec{\nabla} u_1(\vec{r}) \rangle_t = 0 \quad (3.4)$$

from Eq. (3.3). Now consider,

$$\begin{aligned} - \left\langle \frac{K(\vec{S})}{h_1(\vec{S})} u_1(\vec{S}') \right\rangle_t &= \langle \nabla^2 u_1(\vec{S}) u_1(\vec{S}') \rangle_t \\ &- \langle \hat{n}_1 \hat{n}_1 : \vec{\nabla} \vec{\nabla} u_1(\vec{S}) u_1(\vec{S}') \rangle_t. \end{aligned} \quad (3.5)$$

This is obtained from Eq. (2.6); note that  $\hat{n}_1 = \hat{n}_1(|\vec{\nabla} u_1|)$ . Since the average is assumed to be Gaussian, we have

$$\begin{aligned} - \left\langle \frac{K(\vec{S})}{h_1(\vec{S})} u_1(\vec{S}') \right\rangle_t &= \nabla^2 \langle u_1(\vec{S}) u_1(\vec{S}') \rangle_t \\ &- \langle \hat{n}_1 \hat{n}_1 : \vec{\nabla} \vec{\nabla} \langle u_1(\vec{S}) u_1(\vec{S}') \rangle_t \rangle_t. \end{aligned} \quad (3.6)$$

from Eq. (3.4). With the use of the isotropy approximation we have

$$\langle \hat{n}_1 \hat{n}_1 \rangle_t = \frac{1}{d} \vec{I}, \quad (3.7)$$

where  $\vec{I}$  is the unit, second-rank tensor. Thus we finally obtain<sup>19</sup>

$$- \left\langle \frac{K(\vec{S})}{h_1(\vec{S})} u_1(\vec{S}') \right\rangle_t \simeq \frac{d-1}{d} \nabla^2 \langle u_1(\vec{S}) u_1(\vec{S}') \rangle_t. \quad (3.8)$$

Recall from Eq. (2.6f) that

$$\delta^d(\vec{r} - \vec{r}') = \frac{\delta(u_1 - u_1')}{h_1} \delta^{d-1}(\vec{S} - \vec{S}'),$$

so that

$$\delta^d(\vec{r} - \vec{r}')|_{u_1=0} = \Delta \delta^{d-1}(\vec{S} - \vec{S}'), \quad (3.9)$$

where

$$\Delta \equiv \frac{\delta(u_1=0)}{h_1}. \quad (3.10)$$

But from isotropy,

$$\Delta = \frac{\delta(u_2=0)}{h_2} = \dots = \frac{\delta(u_d=0)}{h_d}. \quad (3.11)$$

Therefore, we have

$$\Delta = [\delta^d(\vec{r}=0)]^{1/d}. \quad (3.12)$$

This can be evaluated using a Fourier representation of the  $\delta$  function. Therefore, at  $u_1=0$ ,

$$\delta^{d-1}(\vec{S} - \vec{S}') \simeq \frac{1}{\Delta} \delta^d(\vec{r} - \vec{r}'), \quad (3.13)$$

with  $\Delta$  as given above.

With the use of Eqs. (3.8) and (3.13) in Eq. (3.1), we obtain the simplified equation

$$\begin{aligned} \frac{\partial}{\partial t} \langle u_1(\vec{r}) u_1(\vec{r}') \rangle_t &= L(\nabla^2 - \nabla'^2) \langle u_1(\vec{r}) u_1(\vec{r}') \rangle_t \\ &+ \frac{2k_B T d}{\sigma(d-1)\Delta} L \langle |\vec{\nabla} u_1|^2 \rangle_t \delta^d(\vec{r} - \vec{r}'), \end{aligned} \quad (3.14)$$

where

$$L \equiv \frac{d-1}{d} L'. \quad (3.15)$$

We will use this approximate equation of motion to evaluate the average area per unit volume, which is essentially the reciprocal of the characteristic length.

An expression for the area density is found as follows. Note the identity for the surface area  $A$ ,

$$A = \int d^{d-1} S = \int d^d r \frac{\delta(u_1)}{h_1}.$$

Thus the average area density  $\mathcal{A}$  is given by

$$\mathcal{A} = \left\langle \frac{\delta(u_1)}{h_1} \right\rangle. \quad (3.16)$$

This can be transformed into a more tractable form using the identity

$$h_1 \equiv |\vec{\nabla} u_1|^{-1} = \frac{1}{2\pi} \int d^2 q \frac{1}{q} \exp(i\vec{q} \cdot \vec{\nabla} u_1),$$

valid for  $d=2$  (a similar expression is valid for  $d=3$ ),<sup>7</sup> and a Gaussian representation for the  $\delta$  function. For a Gaussian average in Eq. (3.16) we then obtain

$$\mathcal{A} = \epsilon(d) \left[ \frac{\langle (\vec{\nabla} u_1)^2 \rangle_t}{\langle (u_1)^2 \rangle_t} \right]^{1/2}, \quad (3.17)$$

where<sup>20</sup>

$$\epsilon(d) = \begin{cases} \frac{1}{2\sqrt{2}}, & d=2 \\ \frac{2}{\pi\sqrt{3}}, & d=3 \end{cases}. \quad (3.18)$$

We now return to Eq. (3.14). We introduce the Fourier transformation

$$u_1(\vec{k}, t) = \int d^d r \exp(i\vec{k} \cdot \vec{r}) u_1(\vec{r}, t).$$

The formal solution of Eq. (3.14) is then given by

$$\begin{aligned} \langle u_1(\vec{k}) u_1^*(\vec{k}') \rangle_t &= (2\pi)^d \delta^d(\vec{k} - \vec{k}') \\ &\times \left[ \gamma e^{-2Lk^2 t} + \int_0^t D(t-t') e^{-2Lk^2 t'} dt' \right], \end{aligned} \quad (3.19)$$

where

$$D(t-t') \equiv \frac{2k_B T d}{\sigma(d-1)\Delta} L \langle |\vec{\nabla} u_1|^2 \rangle_{t-t'} . \quad (3.20)$$

[Recall from Eq. (3.3) that  $\langle |\vec{\nabla} u_1|^2 \rangle$  is not a function of spatial position.] If  $D(t)$  is slowly varying over the characteristic time,  $t_c = (2Lk^2)^{-1}$ , of the exponential in the integrand of Eq. (3.19), we have

$$\int_0^t dt' D(t-t') e^{-2Lk^2 t'} \simeq D(t) \int_0^t dt' e^{-2Lk^2 t'} . \quad (3.21)$$

The self-consistency of this approximation can be checked with the value of  $D(t)$  it implies. We find that we require

$$2Lk^2 t \gg 1 .$$

Thus we may calculate the area density to good approximation. The expression for  $\mathcal{A}$  given by Eq. (3.17) involves correlations at the same position. Unfortunately, this approximation precludes a quantitative discussion of the interesting small- $k$  behavior of the structure factor  $\mathcal{S}(k, t)$ .<sup>21</sup> We know of no way to improve the approximation of Eq. (3.21) to yield results for the small- $k$  regime. This is because, for small  $k$ , the integrand of Eq. (3.21) has a very sensitive dependence upon lattice cutoffs. We will make some qualitative statements about the temperature dependence of  $\mathcal{S}(k, t)$  in Sec. V.

With the use of Eq. (3.21), Eq. (3.19) becomes

$$\begin{aligned} \langle u_1(\vec{k}) u_1^*(\vec{k}') \rangle_t &= (2\pi)^d \delta^d(\vec{k} - \vec{k}') \\ &\times \left[ \gamma e^{-2Lk^2 t} + \frac{k_B T d}{\sigma(d-1)\Delta} \langle |\vec{\nabla} u_1|^2 \rangle_t \right. \\ &\quad \left. \times \frac{1 - e^{-2Lk^2 t}}{k^2} \right] . \quad (3.22) \end{aligned}$$

$$\mathcal{A} = \frac{\epsilon(d)\sqrt{d}}{\sqrt{4Lt}} \left[ \frac{1}{1 - \frac{k_B T}{\sigma} (k_{\max})^{d-1} \{ [(4\pi)^{d/2} (d/2)!]^{1/d-1} d/(d-1) \}} \right] , \quad (3.28)$$

where  $\epsilon(d)$  is given by Eq. (3.18). This is a new result. The temperature dependence here is determined to all orders. Note that the surface tension is, of course, temperature dependent. As we discuss below,  $k_{\max}$  is also dependent upon temperature.

This expression has the qualitative form expected from our discussion in Sec. I, since  $\mathcal{A} \sim 1/\bar{R}$ , where  $\bar{R}$  is the characteristic length. We see, however, that the treatment could break down at high temperatures where  $\mathcal{A}$  could become imaginary, which is unphysical. It is not surprising that the linearization scheme, based as it is on an assumption of isotropy, should break down at high temperatures. The higher the temperature, the more important the thermal-noise terms become. Since the crucial approximation has been to assume isotropy, the random forces no longer drive the system to (anisotropic) equilibrium. Instead the random forces are self-consistent with the isotropic, intermediate-time regime.

We would like to estimate the temperature at which Eq. (3.28) breaks down. Consider the average area of a

It only remains to Fourier-transform Eq. (3.22) back to real space. We obtain

$$\langle u_1^2 \rangle_t = 4Lt\lambda^2 \left[ 1 + \frac{\langle |\vec{\nabla} u_1|^2 \rangle_t}{4Lt\lambda^2} \frac{k_B T}{\sigma} g(d) \right] , \quad (3.23)$$

where

$$\lambda^2 \equiv \frac{\gamma}{(2\pi)^{d/2} (4Lt)^{d/2+1}} \quad (3.24)$$

and

$$g(d) = O(k_{\max}^{d-3}) , \quad (3.25)$$

where  $k_{\max}$  is the maximum wave number, which we will discuss below. Multiplying Eq. (3.22) by  $-\vec{k} \cdot \vec{k}'$  and Fourier-transforming we obtain

$$\langle |\vec{\nabla} u_1|^2 \rangle_t = \frac{\lambda^2 d}{1 - \frac{k_B T}{\sigma} [d/(d-1)] (\Delta^{d-1})} . \quad (3.26)$$

Recalling Eq. (3.12) we have

$$\Delta = \left[ \frac{\pi^{d/2} k_{\max}^d}{(d/2)!} \right]^{1/d} . \quad (3.27)$$

Using Eqs. (3.23), (3.26), and (3.27) in Eq. (3.17) we obtain the area density

domain for  $d=2$ . This will be proportional to  $\bar{R}^2 \sim \mathcal{A}^{-2}$ . Explicitly, from Eq. (3.28) we have

$$\bar{R}^2 \propto L' t \alpha_R , \quad (3.29)$$

with

$$\alpha_R = 1 - \frac{k_B T}{\sqrt{\pi\sigma w}} n$$

for  $d=2$ ,<sup>22</sup> where  $w$  is the thickness of the interface and

$$n \equiv k_{\max} w = O(1) . \quad (3.30)$$

There are no higher-order terms in  $k_B T$  in Eq. (3.29). The smallest length in the problem, consistent with our derivation of the Allen-Cahn equation, is the interfacial thickness. [See our remarks following Eq. (2.19) also.] At low temperatures we have  $w=a$ , the lattice constant. At high temperatures, as the critical temperature  $T_c$  is approached,  $w$  will have the same temperature dependence as

the bulk correlation length  $\xi$ . Note though, that our treatment will only be valid if  $w/\bar{R} \ll 1$ . We now evaluate Eq. (3.29) using two different expressions for  $\sigma$ .

The Onsager solution for the surface tension, in terms of the Ising interaction  $J$ , is<sup>23</sup>

$$\sigma a = 2J \left[ 1 - \frac{k_B T}{2J} \ln \coth \frac{J}{k_B T} \right]. \quad (3.31)$$

If the thickness of the interface is  $w = a$ , we find that  $\alpha_R$  vanishes at  $T/T_c \simeq 0.75$  if  $n \simeq 1$  (Fig. 1). Note that  $\alpha_R$  should not vanish until  $T = T_c$ . However, at high temperatures we should have  $w \propto \xi$  as discussed above. For the two-dimensional Ising model,<sup>24</sup>

$$\xi = k_B T / 2\sigma. \quad (3.32)$$

This would imply that  $\alpha_R$  is constant, close to  $T_c$ , where the temperature dependence of  $w$  is precisely that of  $\xi$ . We believe that this again signifies the breakdown of our treatment at high temperatures, at least for the Ising model in two dimensions. We make some further general remarks about the behavior of  $\alpha_R$ , close to  $T_c$ , at the end of this section.

Alternatively, we consider the mean-field surface tension. An  $M^4$  field theory of  $f(M)$  in Eq. (2.3) implies

$$M(x) = M_e \tanh(x/2w), \quad (3.33)$$

where  $\pm M_e$  is the magnetization in one of the two bulk phases. In the simplest mean field,  $M_e$  is determined by<sup>25</sup>

$$M_e = \tanh \left[ \frac{M_e}{T/T_c} \right]. \quad (3.34)$$

The mean-field surface tension, from Eqs. (3.33) and (2.4) with  $C = J$ , is given by

$$\sigma w = \frac{8}{3} J M_e^2. \quad (3.35)$$

We find that  $\alpha_R$  vanishes at  $T/T_c \simeq 0.85$  if  $n \simeq 1$  (Fig. 2).

Near  $T_c$  the temperature dependence of  $w$  will be given by the bulk correlation length  $\xi$ . We have argued that our

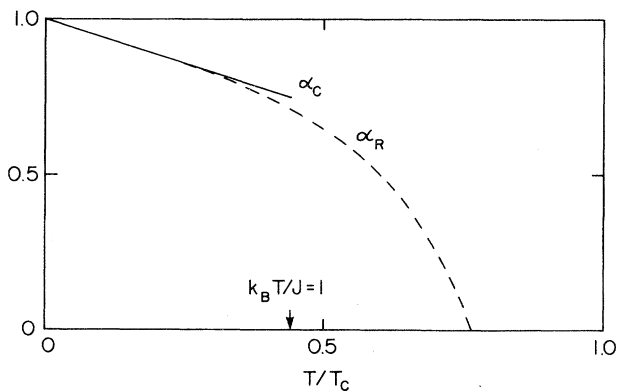


FIG. 1. Two theoretical expressions  $\alpha_R$  [from Eq. (5.2), dashed line] and  $\alpha_c$  [from Eq. (5.3), solid line], are plotted vs  $T/T_c$ . Parameter  $n$  in  $\alpha_R$  has been chosen to be  $\sqrt{\pi}/2 \simeq 0.9$ . Surface tension is the Onsager expression from Eq. (3.31).

treatment breaks down at high temperatures. Nevertheless, it may be of interest to note the temperature dependence of Eq. (3.28) as  $T \rightarrow T_c$  for  $d < 4$ . Hyperscaling requires that<sup>26</sup>

$$\sigma w^{d-1} \rightarrow \text{const} \quad (3.36)$$

as  $T \rightarrow T_c$ . Since  $\mathcal{A} \propto 1/\bar{R}$ , we can rewrite Eq. (3.28) as

$$\bar{R}^2 \propto L' t \alpha_R^{(d)}. \quad (3.37)$$

Thus at  $T \rightarrow T_c$ , we have

$$\alpha_R^{(d)} \sim (1 - bT/T_c), \quad (3.38)$$

for  $d < 4$ , where  $b$  is a constant close to  $T_c$ . If  $b$  is unity, higher-order terms in Eq. (3.36) may be important. We caution, though, that our analysis almost certainly breaks down close to  $T_c$ . (This is what we have seen above for the two-dimensional Ising model.) For example, our treatment could imply that the constant  $b$  in Eq. (3.38) was greater than one. This would make  $\alpha_R$  unphysically negative.

#### IV. AN INITIALLY CIRCULAR CONFIGURATION

In this section we will analyze the dynamics of two-dimensional circular domains—a region of up spins, say, in a sea of down spins. We will make use of the Allen-Cahn equation with thermal noise for our study.

As we noted in Sec. I, this problem has been considered by SSG (Ref. 11) making use of a somewhat different equation of motion. The SSG equation does not feature the tilt terms, required for Euclidean invariance, or the surface tension, required for consistency with thermal equilibrium. We believe, then, that it is worth carrying out their program using the Allen-Cahn equation with noise, since this equation of motion has these desirable features.

We will assume that there is a circular domain at  $t = 0$ . Let us define a class of surfaces by

$$u_1(\vec{r}, t) = r - R(\theta, t), \quad (4.1)$$

so that the physical surface is

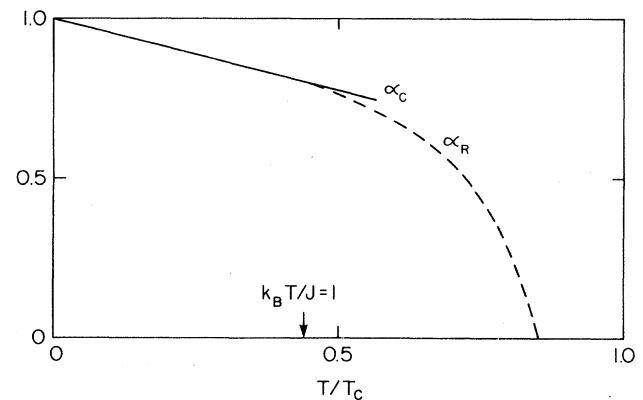


FIG. 2. Two theoretical expressions  $\alpha_R$  and  $\alpha_c$  are again plotted vs  $T/T_c$ . Parameter  $n$  in  $\alpha_R$  has been chosen to be  $\sqrt{\pi}/2 \simeq 0.9$ . Surface tension is the mean-field expression from Eq. (3.35).

$$u_1(\vec{r}, t) = 0. \quad (4.2)$$

From Eq. (2.6) the "tilt" term is

$$1/h_1 = \{1 + [(1/r)R_\theta]^2\}^{1/2}, \quad (4.3)$$

and the normal unit vector to the  $u_1$  surface is

$$\hat{n}_1 = \frac{\hat{r} - (1/r)R_\theta \hat{\theta}}{\{1 + [(1/r)R_\theta]^2\}^{1/2}}, \quad (4.4)$$

where

$$R_\theta \equiv \frac{\partial R}{\partial \theta}.$$

An expression for the curvature can be obtained by taking the gradient of this normal vector from Eq. (2.6e).

Recall the Langevin-equation version of the Allen-Cahn equation from Eqs. (2.18) and (2.19). It is straightforward to show that at  $u_1 = 0$

$$\begin{aligned} \frac{\partial R(\theta, t)}{\partial t} = & \frac{-L'}{R} + L' \frac{R_{\theta\theta}}{R^2} \left[ \frac{1}{1 + (R_\theta/R)^2} \right] \\ & - L' \frac{1}{R} \left[ \frac{(R_\theta/R)^2}{1 + (R_\theta/R)^2} \right] + \frac{\eta'}{R^{1/2}}, \end{aligned} \quad (4.5a)$$

where

$$\begin{aligned} \langle \eta'(\theta, t) \eta'(\theta', t') \rangle = & \frac{2k_B T L'}{\sigma} \delta(\theta - \theta') \delta(t - t') \\ & \times [1 + O(R_\theta/R)^2]. \end{aligned} \quad (4.5b)$$

This equation can be reduced to the SSG equation [Eqs. (4) and (5) of Ref. 11(b)] provided we put  $(R_\theta/R)^2 = 0$ , and replace  $\sigma$  by a rough approximation for its mean-field value.<sup>13</sup> These replacements are not justified for the Allen-Cahn equation, which we will consider here. A justification of the SSG equation of motion, independent of the Allen-Cahn equation, may be possible.<sup>12</sup>

Following the treatment of SSG, we expand  $R(\theta, t)$  in a nonanalytic series in (dimensionless) temperature, i.e.,

$$R(\theta, t) = R_0 + R_1 + R_2 + \dots, \quad (4.6)$$

where

$$\frac{\partial R_0}{\partial \theta} \equiv R_{0\theta} = 0.$$

The subscripts  $n$  refer to terms of order  $(k_B T)^{n/2}$ .<sup>27</sup> For simplicity we scale all lengths to the lattice constant and let  $L't \equiv t^*$ . We then obtain the following set of perturbation equations from Eqs. (4.5) and (4.6):

$$\frac{\partial R_0}{\partial t^*} = \frac{1}{R_0}, \quad (4.7a)$$

$$\frac{\partial R_1}{\partial t^*} = \frac{R_1}{R_0^2} + \frac{R_{1\theta\theta}}{R_0^2} + \frac{\eta'}{R_0^{1/2}}, \quad (4.7b)$$

and

$$\begin{aligned} \frac{\partial R_2}{\partial t^*} = & \frac{R_2}{R_0^2} - \frac{R_1^2}{R_0^3} + \frac{R_{2\theta\theta}}{R_0^2} - \frac{2R_1 R_{1\theta\theta}}{R_0^3} \\ & - \frac{R_{1\theta}^2}{R_0^3} - \frac{\eta' R_1}{2R_0^{3/2}}, \end{aligned} \quad (4.7c)$$

where

$$\langle \eta'(\theta, t^*) \eta'(\theta', t^{*'}) \rangle = \frac{2k_B T}{\sigma} \delta(\theta - \theta') \delta(t^* - t^{*'}). \quad (4.7d)$$

The next-to-last term on the right-hand side of Eq. (4.7c) gives the new deterministic contribution from the tilt terms in the Allen-Cahn equation, which is not present in the analysis of Safran *et al.* Except for this deterministic contribution and the factor of surface tension in Eq. (4.7d) (Ref. 13) this set of equations is identical to that given by SSG.<sup>11</sup>

We note immediately that if  $R_0(t=0) \equiv L_0$ , then

$$L_0^2 - R_0^2(t) = 2t^* \quad (4.8)$$

from Eq. (4.7a). For convenience, let us call

$$\bar{R}^2 |_{k_B T=0} \equiv L_0^2 - R_0^2(t) \propto t^*. \quad (4.9)$$

In the following, we calculate

$$\bar{R}^2 \propto t^* \alpha_c = L't \alpha_c, \quad (4.10)$$

where  $\alpha_c$  will determine the temperature-dependent effects. It corresponds to  $\alpha_R$  of Eq. (3.29) above. Following SSG, we define

$$\alpha_c \equiv \frac{\frac{\partial}{\partial t^*} \left[ \pi \int_0^{2\pi} \frac{d\theta}{2\pi} \langle R^2(\theta, t) \rangle \right]}{\frac{\partial}{\partial t^*} [\pi R_0^2(t)]}. \quad (4.11)$$

From Eq. (4.6) we obtain

$$\begin{aligned} \alpha_c = & 1 - \int_0^{2\pi} \frac{d\theta}{2\pi} \left[ \left\langle R_1 \frac{\partial R_1}{\partial t^*} \right\rangle + \frac{\partial R_0}{\partial t^*} \langle R_2 \rangle \right. \\ & \left. + R_0 \left\langle \frac{\partial R_2}{\partial t^*} \right\rangle \right] + \dots \end{aligned} \quad (4.12)$$

With the use of Eq. (4.7), this simplifies to

$$\alpha_c = 1 - \int_0^{2\pi} \frac{d\theta}{2\pi} \left[ \frac{\langle \eta R_1 \rangle}{2R_0^{1/2}} \right] + \dots \quad (4.13)$$

It is worth noting that the SSG theory gives another term in this expression which is of order  $\langle R_{1\theta}^2 \rangle$ ; see Eq. (7) of Ref. 11(b). In our treatment this contribution cancels with the new term self-consistently present in the Allen-Cahn equation.

We evaluate  $\alpha_c$  in Eq. (4.13) in the following way. Let

$$\eta'(\theta) = \sum_{n=-N}^{N-1} \eta'_n e^{in\theta} \quad (4.14)$$

and

$$R_1(\theta) = \sum_{n=-N}^{N-1} g_n e^{in\theta},$$



where  $N \simeq \pi R_0$ , so that the sums run over the dimensionless circumference of the circle. Equations (4.7b) and (4.7d) can then be written as

$$\frac{\partial g_n}{\partial t^*} = \frac{1-n^2}{R_0^2} g_n + \frac{1}{R_0^{1/2}} \eta'_n, \quad (4.15a)$$

where

$$\langle \eta'_n(t^*) \eta'_{n'}(t^{*'}) \rangle = \frac{k_B T}{\pi \sigma} \delta_{n+n',0} \delta(t^* - t^{*'}). \quad (4.15b)$$

This Langevin equation can be put into Fokker-Planck form for the probability distribution  $P$ ,

$$\frac{\partial}{\partial t^*} P(g, t^*) = \sum_n \frac{\partial}{\partial g_n} \left[ \frac{1-n^2}{R_0^2} g_n + \frac{k_B T}{2\pi R_0 \sigma} \frac{\partial}{\partial g_{-n}} \right] P(g, t^*). \quad (4.16)$$

Thus we obtain the second moment,

$$\frac{\partial}{\partial t^*} \langle g_n g_{-n} \rangle = 2 \frac{1-n^2}{R_0^2} \langle g_n g_{-n} \rangle + \frac{k_B T}{\pi R_0 \sigma}. \quad (4.17)$$

From Eqs. (4.15) and (4.17) we can straightforwardly obtain

$$\int_0^{2\pi} \frac{d\theta}{2\pi} \langle \eta'(\theta) R_1(\theta) \rangle = \sum_{n=-N}^{N-1} \langle \eta'_n g_{-n} \rangle = \frac{k_B T}{\sigma} R_0^{1/2} (N/\pi R_0). \quad (4.18)$$

Therefore in Eq. (4.13) we have

$$\alpha_c = 1 - \frac{k_B T}{2\sigma} \frac{N}{\pi R_0} + \dots \quad (4.19)$$

Now recalling that  $N \simeq \pi R_0$ , and that all lengths have been scaled to the lattice constant  $a$ , we have

$$\alpha_c = 1 - \frac{k_B T}{2\sigma a} + \dots \quad (4.20)$$

This is a new result. It is valid to first order in the temperature expansion; the surface tension is evaluated at its zero-temperature value. The form of  $\alpha_c$  is plotted in Fig. 1 using the Onsager surface tension [Eq. (3.31)]. In Fig. 2,  $\alpha_c$  is plotted using the mean-field surface tension from Eq. (3.35).

## V. DISCUSSION AND SUMMARY

Let us review our results. We find that the characteristic length  $\bar{R}$  obeys the growth law

$$\bar{R}^2 \propto L' t \alpha. \quad (5.1)$$

From our treatment of  $d=2$  random interfaces we obtain [Eq. (3.29)]

$$\alpha_R = 1 - \frac{k_B T}{\sqrt{\pi \sigma w}} n, \quad (5.2)$$

which is “summed” to all orders in  $k_B T$ , where  $n \equiv k_{\max} w \sim O(1)$ . At low temperatures we have chosen  $w = a$ , as we have discussed in Secs. II and III. [Results for arbitrary dimensionality are given in Eq. (3.28).] From our treatment of  $d=2$  circular interfaces we obtain [Eq. (4.20)]

$$\alpha_c = 1 - \frac{k_B T}{2\sigma a} + \dots \quad (5.3)$$

to leading order in the temperature expansion. The agreement between the two different treatments is gratifying, if somewhat surprising, considering the drastically different approaches of the last two sections. We note some important differences nevertheless.

Equation (5.3) is to linear order in the temperature. It will break down when higher-order terms in the expansion become important. An estimate of this can be made by examining the temperature dependence of the surface tension. From the Onsager solution for  $\sigma$ , the temperature dependence of  $\sigma$  becomes important for  $T/T_c \gtrsim 0.5$ , or  $k_B T/J \gtrsim 1$ , where  $J$  is the Ising interaction constant. Thus we expect Eq. (5.3) to be valid for  $k_B T/J < 1$ .

On the other hand, Eq. (5.2) is given to all orders in temperature. The expression appears to become unphysical at high temperatures. This signifies the breakdown of the linearization scheme,<sup>7</sup> as discussed at the end of Sec. III. Note though, that for  $n \simeq 1$ , this breakdown does not occur until  $T/T_c \gtrsim 0.75$ . It is possible then, that Eq. (5.2) [and Eq. (3.29)] gives the qualitative, or even quantitative, temperature dependence of the interfacial dynamics, provided we remain at low to intermediate temperatures. An experimental study would resolve this question.

We now discuss some implicit assumptions in our treatment. Note that we are analyzing the dynamical roughening in a nonequilibrium state. Roughening effects for a two-dimensional system in thermodynamic (infinite volume) equilibrium (infinite time) can lead to a diffuse interface of infinite thickness.<sup>28</sup> In our treatment we remain far from equilibrium.

We also note that our treatment has been in the continuum limit, notwithstanding the appearance of wave-number cutoffs in our results. We do not know if the differences between lattice and continuum models will complicate the comparison of our theoretical results with computer simulation. These differences will be most important at low temperatures. As discussed in Sec. II, it could be of interest to extend the treatment given here to take lattice effects into more explicit account. This could be done by either considering higher-order correction terms to the Allen-Cahn equation,<sup>9</sup> or by completely reformulating the Allen-Cahn equation as a lattice-dynamical theory.

Computer experiments on the growth rate of two-dimensional circular domains have been performed by Safran, Sahni, and Grest.<sup>11</sup> Their results were analyzed with their model equation of motion. This equation of motion is neither Euclidean invariant, nor consistent with equilibrium, as we have discussed in Secs. I and IV. A justification of this equation may, nevertheless, be possible.<sup>12</sup> Safran *et al.* obtained a quantity analogous to  $\alpha_c$  of

Fig. (5.3) above, to linear order in the temperature ( $k_B T/J$ ). Somewhat surprisingly, they found good agreement between theory and experiment for  $k_B T/J > 1$ . Neither of the expressions we have presented for  $d=2$ , Eqs. (5.2) and (5.3), are as good a representation of their data in this high-temperature regime.

We note the following about Safran *et al.*'s treatment. Their experimental results featured a latency period for  $k_B T/J < 1$  where no temperature-dependent effects were present. The authors attributed this to systematic problems arising in the simulation of circles on a square lattice. Unfortunately,  $k_B T/J < 1$  is the region where a sensitive test of a linear theory in  $k_B T/J$  to experiment can be made. It is possible that the good agreement for  $k_B T/J > 1$  is due to extensive cancellation of higher-order terms in the perturbation series. The origin of such cancellation is not obvious.<sup>29</sup>

One interesting effect found by Safran *et al.* was not discussed at length by the authors. They observed non-trivial differences between the temperature-dependent behavior of their Glauber and Kawasaki simulations of circular domains. Safran *et al.* scaled the Monte Carlo hopping time  $\tau_K$  of the Kawasaki dynamics to a temperature-dependent factor. This was done to obtain results in qualitative agreement with their Glauber dynamics simulation. The scaling of  $\tau_K$  would, we suppose, have its origin in the Master equation. A theoretical analysis could be of interest, since this temperature-dependent scaling of  $\tau_K$  is a stronger effect than the temperature dependence of domain growth reported by Safran *et al.*

Our approximations in Sec. III precluded a discussion of the temperature dependence of the structure factor  $\mathcal{S}(k,t)$ . We can make some qualitative comments about  $\mathcal{S}(k,t)$ , however. As we have seen, temperature-dependent roughening effectively slows down domain growth, i.e., reduces  $\bar{R}$ . However, the  $k^2$  moment of  $\mathcal{S}(k,t)$  is roughly proportional to  $1/\bar{R}^2$ , so if  $\bar{R}$  decreases, the second moment of  $\mathcal{S}(k,t)$  increases.  $\mathcal{S}(k,t)$  is roughly bell shaped and satisfies a normalization condition (zeroth sum rule). Thus, the structure factor will "flatten out" as the temperature is increased and so the second moment of  $\mathcal{S}(k,t)$  increases. This heuristic argument indicates that the flattening would be a monotonic function of temperature.

Note though, that the structure factor is relatively insensitive to the detailed dynamics of the interface. For example, the assumption of a sharp step-function-like

order-parameter profile at the interface (which is not a dynamical assumption) will result in a scaled structure factor which obeys  $1/(k^*)^{d+1}$  behavior,<sup>30</sup> for large  $k^* \equiv k\sqrt{4L't}$ . It is this asymptotic behavior which determines much of the structure factor reported by Ohta *et al.* in Figs. 1 and 2 of Ref. 7. An experimental study of the  $k^* \lesssim 1$  regime would be of interest. It seems clear, however, that experimental results for the area density would provide a more sensitive test of theory.

In conclusion, we have analyzed the temperature dependence of the kinetics of random interfaces. From the Allen-Cahn equation of motion, supplemented with thermal noise contributions, we obtained the temperature dependence of the characteristic growth rate of domains. The actual growth law,  $\bar{R} \sim t^{1/2}$ , was unchanged, but the proportionality constant was renormalized by a temperature-dependent coefficient. We analyzed two initial configurations: (i) a random configuration of interfaces in  $d$  dimensions, and (ii) a circular interface in two dimensions. Our results were given by Eqs. (3.29) and (5.1)–(5.3). The results of Sec. III indicated that the linearization scheme of Ohta *et al.*<sup>7</sup> breaks down at high temperatures, although it may be good at low to intermediate temperatures. In Sec. IV we obtained low-temperature results for circular domains which were consistent with the results of Sec. III. There appears to be a clear need for experiments to stringently test theory, particularly over low temperatures.

*Note added in proof.* A computer study of the temperature dependence of quenched systems has been performed recently by K. Kaski, C. Yalabik, J. D. Gunton, and P. S. Sahni [Phys. Rev. B **28**, 5263 (1983)].

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<sup>1</sup>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.  
<sup>2</sup>K. Kawasaki, M. C. Yalabik, and J. D. Gunton, Phys. Rev. A **17**, 455 (1978).  
<sup>3</sup>S. M. Allen and J. W. Cahn, Acta Metall. **27**, 1085 (1979); see also the complementary treatment of S.-K. Chan, J. Chem. Phys. **67**, 5755 (1977).  
<sup>4</sup>M. T. Collins and H. C. Teh, Phys. Rev. Lett. **30**, 781 (1973); T. Hashimoto, T. Miyoshi, and M. Ohtsuka, Phys. Rev. B **13**, 1119 (1976); T. Hashimoto, K. Nishimura, and Y. Takeuchi,

Phys. Lett. **65A**, 250 (1978); M. Sato and K. Hirakawa, J. Phys. Soc. Jpn. **42**, 433 (1977).  
<sup>5</sup>M. K. Phani, J. L. Lebowitz, M. H. Kalos, and O. Penrose, Phys. Rev. Lett. **45**, 366 (1980).  
<sup>6</sup>P. S. Sahni, G. Dee, J. D. Gunton, M. K. Phani, J. L. Lebowitz, and M. H. Kalos, Phys. Rev. B **24**, 410 (1981).  
<sup>7</sup>T. Ohta, D. Jasnow, and K. Kawasaki, Phys. Rev. Lett. **49**, 1223 (1982).  
<sup>8</sup>K. Kawasaki (private communication).  
<sup>9</sup>K. Kawasaki and T. Ohta, Prog. Theor. Phys. **67**, 147 (1982); **68**, 129 (1982); Physica **118A**, 175 (1983).

- <sup>10</sup>R. Bausch, V. Dohm, H. K. Janssen, and R. K. P. Zia, *Phys. Rev. Lett.* **47**, 1837 (1981).
- <sup>11</sup>(a) S. A. Safran, P. S. Sahni, and G. S. Grest, *Phys. Rev. B* **28**, 2693 (1983); (b) P. S. Sahni, G. S. Grest, and S. A. Safran, *Phys. Rev. Lett.* **50**, 60 (1983).
- <sup>12</sup>S. A. Safran (private communication).
- <sup>13</sup>Instead of  $\sigma$ , Safran *et al.* have  $4JM_e^2/w^2$ , where  $J$  is the interaction constant,  $M_e$  is the equilibrium value of the magnetization far from the interface, and  $w$  is the thickness of the interface. The mean-field surface tension is  $\frac{8}{3}JM_e^2/w$ .
- <sup>14</sup>By Euclidean invariance it is meant that the physical properties of the system are invariant under translations and rotations of the interface. Some analogies can be drawn between the tilt factors [see Eq. (4.3)] in Euclidean invariance, and the  $(1-v^2/c^2)^{1/2}$  factors in Lorentz invariance. See D. J. Wallace, in *Gauge Theories and Experiments at High Energies*, edited by K. C. Bowler and D. G. Sutherland (Scottish University Summer School Publications, Edinburgh, 1983). Of course the metric here is much simpler than the corresponding one in special relativity; dot products of vectors here do not involve the subtraction of any of the products of components of those vectors.
- <sup>15</sup>Similar arguments can be advanced for the shrinkage of circular domains. However, long thin strips evolve more rapidly with increasing temperature. See Ref. 11.
- <sup>16</sup>The actual surface is, of course, diffuse and is located *about*  $u_1=0$ . Its thickness  $w$  is given by the nonvanishing domain of the gradient of the order parameter.
- <sup>17</sup>G. Arfken, *Mathematical Methods for Physicists* (Academic, New York, 1970), Chap. 2.
- <sup>18</sup>See, for example, D. Bedeaux, *Phys. Lett.* **62A**, 10 (1977). This paper discusses a more general case.
- <sup>19</sup>If we evaluate the first moment of Eq. (2.20) in the same way, we find that  $\langle u_1 \rangle_t$  is constant in space and time. Thus  $\langle u_1 \rangle_t$  is always zero on recalling Eq. (2.5).
- <sup>20</sup>We obtain a factor of  $\sqrt{d}$  different from that given in Ref. 7.
- <sup>21</sup>The large  $k$  behavior of  $\mathcal{S}(k,t)$  gives the second moment,  $\int d^d k k^2 \mathcal{S}(k,t)$ , which is roughly proportional to  $\mathcal{A}^2$  (Ref. 6).
- <sup>22</sup>For  $d=3$ , Eq. (3.28) gives  $\bar{R}^2 \propto L't [1 - (3/2^5 \pi^4)^{1/3} (k_B T / \sigma w^2) n^2]$ , to all orders in  $k_B T$ .
- <sup>23</sup>L. Onsager, *Phys. Rev.* **65**, 117 (1944).
- <sup>24</sup>B. M. McCoy and T. T. Wu, *The Two Dimensional Ising Model* (Harvard University Press, Cambridge, Mass., 1973), Chap. XII.
- <sup>25</sup>R. K. Pathria, *Statistical Mechanics* (Pergamon, New York, 1972).
- <sup>26</sup>B. Widom, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, London, 1972), Vol. 2, p. 79.
- <sup>27</sup>Alternatively, this can be thought of as a power series in the random force  $\eta'$ . This is, of course, an unusual approach. We know of no first-principles justification for  $R(\theta,t)$  to be an analytic functional of  $\eta'$ .
- <sup>28</sup>This is a rigorous result for the two-dimensional Ising model: M. Aizenman, *Phys. Rev. Lett.* **43**, 407 (1979).
- <sup>29</sup>In Safran *et al.*'s theoretical analysis of their equation of motion the temperature dependence of  $M_e^2/w^2$  is neglected. See Refs. 11 and 13. This temperature dependence, which is not insignificant, would have to cancel with higher-order terms in the perturbation analysis.
- <sup>30</sup>This is discussed in Ref. 1 as well as in P. A. Rikvold and J. D. Gunton, *Phys. Rev. Lett.* **49**, 286 (1982). Note that this means that all wave-number moments of  $\mathcal{S}(k,t)$ , of order greater than one, will diverge unless there is an "ultraviolet" cutoff  $k_{\max}$ . However, if there is a lattice cutoff in the  $k$  integrals,  $\mathcal{S}(k,t)$  will have an additional weak time dependence beyond that implied by scaling.