Microscopic analysis of the coarsening of an interface in the spinodal decomposition of a binary fluid

V. Kumaran
Department of Chemical Engineering, Indian Institute of Science, Bangalore 560 012, India

(Received 13 February 1998; accepted 29 April 1998)

The coarsening of a random interface in a fluid of surface tension γ and viscosity μ is analyzed using a curvature distribution function $A(K_m,K_g,t)$ which gives the distribution of the mean curvature K_m and Gaussian curvature K_g on the interface. There is a variation in the area distribution function due to the rate of change of K_m, K_g and the compression of the interface due to tangential motion. The rates of change of mean and Gaussian curvature at a point are related to the rate of change of the normal velocity in the tangential directions along the interface. The fluid velocity is governed by the Stokes equation for a viscous flow, and the velocity field at a point is determined as an integral of the product of the Oseen tensor and the normal force at other points on the interface. Using a general form for this integral, it is shown that there is a characteristic variable $K_*=K_g/(K_m^2-4K_g)^{1/2}$ which is independent of time even as K_m and K_g decrease proportional to t^{-1} and t^{-2}, respectively. In the late stages, analytical forms for the distribution function are determined in the limit $K_m \ll K_*$ using a similarity variable $\eta=(\gamma K_m t/\mu)$. Two reasonable approximations are used for the characteristic length for the correlation of the curvature and normal along the interface, and the results for these two approximations are quadratic polynomials in $|\eta|$ which are nonzero for a finite interval about $\eta=0$. It is expected that the actual distribution function is in between these two limiting cases. © *1998 American Institute of Physics.*
[S0021-9606(98)50130-4]

I. INTRODUCTION

The late stage of spinodal decomposition in a binary fluid is characterized by the presence of sharp interfaces between the two phases. The structure formed during coarsening depends on the relative concentrations of the two species in the mixture. A bicontinuous structure, where the two phases are separated by a random interface, is observed for a nearly symmetric mixture. In the case of an asymmetric mixture, decomposition proceeds by the growth of droplets of the minority phase in a supersaturated matrix of the majority phase. A microscopic analysis of the growth of a random interface in a fluid is the subject of the present study.

The late stage decomposition in binary alloys and magnetic systems has been studied in some detail. The Lifshitz–Slyozov theory[1] for the late stage coarsening of droplets in an alloy predicts that the average droplet radius increases proportional to $t^{1/3}$, and this growth law has been confirmed by experiments. The theories for the coarsening of a random interface in solid systems are less well developed. Ohta, Jasnow, and Kawasaki[2] used a constitutive relation for the motion of the interface as a function of the curvature and surface tension at a point, and predicted that the characteristic length in the system increases proportional to $t^{1/2}$ for a nonconserved order parameter system. In a conserved order parameter system, the characteristic length increases proportional to $t^{1/3}$ due to the additional constraint of conservation of magnetization. Both of these scaling laws were found to be in agreement with simulations.[3] Though the scaling of the characteristic length has been obtained, a more quantitative description of, for example, the distribution of mean and Gaussian curvatures along the interface, is still lacking.

The late stage coarsening of interfaces in a fluid has received relatively less attention, possibly due to the complexity of the dynamical equations. In particular, the hydrodynamic interactions in a fluid are long range, and the velocity at a point on an interface depends on the forces exerted on the fluid by other sections of the interface. Consequently, the interface velocity is given by an integral equation, which contains the correlations in the curvature at different locations. This causes difficulties in the rigorous derivation of averaged equations for the interface dynamics, and the calculation of the distribution of curvatures along the interface. The earliest prediction of the coarsening of an interface by Siggia[4] was based on a simple dimensional analysis. If the only parameters which determine the interface dynamics are the surface tension γ (which causes the normal force) and the fluid viscosity μ, then the characteristic length of the interface l has to scale as $l=(\gamma t/\mu)$. Furukawa[5] also put forth similar arguments for the scaling of the characteristic length of the interface, and this has been verified in experiments.[6,7]

A more detailed approach was pursued by the author,[8] where a characteristic curvature $K=(K_1^2+K_2^2)^{1/2}$ was used to describe the dynamics of the interface. A curvature distribution function $A(K,t)$ was defined such that $A(K,t)dK$ is the area per unit volume with curvature in the interval dK about K at time t. There is a change in the curvature distribution due to the change in curvature, and due to the tangential compression of the interface. Using the simple assumption that the only length scale which determines the dynamics at a point is the curvature K itself, simple constitutive relations

were obtained for the rate of change of curvature and the tangential compression of the interface. Using these, an expression for the curvature distribution function was derived,

$$A(\eta)=(1-\eta)^a, \tag{1}$$

where $\eta=(a_1K\gamma/\mu)$ is a similarity variable, and a is a phenomenological constant in the constitutive equations for the rate of change of curvature and the tangential compression of the interface.

The analysis leading to Eq. (1) has the disadvantage that it is based on phenomenological relations for the rate of change of curvature and the tangential compression of the interface. Consequently, the constant a in Eq. (1) is as yet unknown. The objective of the present analysis is to analyze the dynamics starting from a microscopic description of the motion of the interface. The present analysis is more exact, and the dynamics of the interface is expressed using an area distribution function $A(K_m,K_g,t)$ which is a function of the mean curvature K_m and Gaussian curvature K_g. The fluid velocity due to the force exerted by the interface is determined from the solution of the Stokes equation, and the rates of change of curvatures and area are determined as functions of the variations in the normal velocity along the interface. To obtain averaged equations for the rates of change of curvature, the microscopic equations for the rates of change of curvature are averaged over the distributions interface configurations in the vicinity of a point where the curvature and unit normal are known. These equations indicate that there is a variable $K_*=K_g/(K_m^2-4K_g)^{1/2}$ which is invariant during the decomposition process, while K_m and K_g decrease proportional to t^{-1} and t^{-2}. The conservation equation for the area distribution function is difficult to solve in general, but analytical solutions are obtained in the late stages where $K_*\gg K_m$ using some approximations regarding the interaction between different sections of the interface.

II. ANALYSIS

The two-dimensional surface in three dimensions is completely defined by the mean curvature $K_m=K_\alpha+K_\beta$ and the Gaussian curvature $K_g=K_\alpha K_\beta$, where K_α and K_β are the principle curvatures, which are the extrema of the curvatures at a point. The dynamics of the surface is described using a ''curvature distribution function'' $A(t,K_m,K_g)$ such that $A(t,K_m,K_g)dK_mdK_g$ is the area per unit volume which has mean curvature in the interval dK_m about K_m and Gaussian curvature in the interval dK_g about K_g. A conservation equation for the curvature distribution function can be written as

$$\frac{\partial A}{\partial t}+\frac{\partial A}{\partial K_m}\frac{dK_m}{dt}+\frac{\partial A}{\partial K_g}\frac{dK_g}{dt}=\frac{\partial_c A}{\partial t}. \tag{2}$$

The left-hand side of Eq. (2) is the change in the curvature distribution function in a ''Lagrangian'' reference frame in the K_m-K_g plane, and the right-hand side represents the change in the area due to contraction of the surface caused by the tangential motion of fluid along the interface. The conservation equation can be solved if expressions for the rates of change of curvature, (dK_m/dt) and (dK_g/dt), and the rate of change of area due to contraction $(\partial_c A/\partial t)$, are known. These are determined using a microscopic model for the fluid velocity induced by the surface tension forces at the interface.

The fluid velocity field is obtained by solving the Navier–Stokes equations for a simple fluid with density ρ and viscosity μ. The equations are simplified by assuming that inertial effects are negligible, the validity of this assumption was examined earlier.[8] With this approximation, the velocity field at a point $\mathbf{x}$ in the fluid is given by

$$v_i(\mathbf{x})=\gamma\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}'), \tag{3}$$

where $\mathbf{x}'$ is a point on the interface, dS' is a differential surface area of the interface, the Oseen tensor J_{ij} is

$$J_{ij}(\mathbf{x})=\frac{1}{8\pi\mu}\left(\frac{\delta_{ij}}{|\mathbf{x}|}+\frac{x_ix_j}{|\mathbf{x}|^3}\right), \tag{4}$$

and the pressure on the fluid at the point $\mathbf{x}'$ is given by $\gamma n_j'(\mathbf{x}')K_m'(\mathbf{x}')$, where γ is the surface tension, $\mathbf{n}'(\mathbf{x}')$ is the unit normal to the interface at this point, and the integral in Eq. (3) is carried out over the surface of the interface.

The change in curvature and the contraction of the interface at a point $\mathbf{x}$ caused by the fluid motion are examined next. Consider an orthogonal coordinate system at a point $\mathbf{x}$ on the interface, where $\mathbf{n}$ is the unit normal and α and β are tangents to the interface along the directions of extremal curvature x_α and x_β. The principal curvatures K_α and K_β are the curvatures of the interface along the x_α and x_β directions, and the curvatures are considered positive if the center of curvature is on the side of the interface into which the unit normal is directed. The normal velocity of the interface v_n at the point $\mathbf{x}$ is

$$v_n=\gamma n_i(\mathbf{x})\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}'). \tag{5}$$

The rate of change of curvature (dK_α/dt) is the second derivative of the normal velocity along the direction tangential to the surface $(\partial_\alpha^2 v_n)$. The following identities are necessary for determining the gradients along the surface:

$$\begin{aligned}
&\partial_\alpha n_i=-K_\alpha\alpha_i,\\
&\partial_\alpha\alpha_i=K_\alpha n_i\\
&\partial_\alpha\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j(\mathbf{x}')K_m(\mathbf{x}')\\
&\quad=\alpha_k\int dS'\partial_k J_{ij}(\mathbf{x}-\mathbf{x}')n_j(\mathbf{x}')K_m(\mathbf{x}'),
\end{aligned} \tag{6}$$

where $\partial_\alpha\equiv(\partial/\partial x_\alpha)$ is the gradient along the interface, and $\partial_i\equiv(\partial/\partial x_i)$ is the gradient in three dimensions. Using the above relations, the rate of change of the principal curvature K_α is

$$\frac{dK_\alpha}{dt}=\partial_\alpha^2 v_n$$
$$=\gamma\Big[(-(\partial_\alpha K_\alpha)\alpha_i-K_\alpha^2 n_i)\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')\times n_j'(\mathbf{x}')K_m'(\mathbf{x}')+K_\alpha(n_i n_k-2\alpha_i\alpha_k)\times\int dS'\partial_k J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')+n_i\alpha_k\alpha_l\int dS'\partial_k\partial_l J_{ij}n_j'(\mathbf{x}')K_m'(\mathbf{x}')\Big], \quad (7)$$

where $\partial_\alpha K_\alpha$ represents the rate of change of curvature along the coordinate α on the surface. A similar expression can be derived for the rate of change of curvature in the β direction, (dK_β/dt), and using these, the rate of change of the mean curvature and the Gaussian curvature are determined,

$$\frac{dK_m}{dt}=\gamma\Big[(-(\partial_\alpha K_\alpha)\alpha_i-(\partial_\beta K_\beta)\beta_i-(K_m^2-2K_g)n_i)\times\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')+(K_m n_i n_k-2K_\alpha\alpha_i\alpha_k-2K_\beta\beta_i\beta_k)\times\int dS'\partial_k J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')+n_i(\alpha_k\alpha_l+\beta_k\beta_l)\times\int dS'\partial_k\partial_l J_{ij}n_j'(\mathbf{x}')K_m'(\mathbf{x}')\Big]. \quad (8)$$

A similar expression can be derived for the Gaussian curvature by adding $K_\beta(dK_\alpha/dt)+K_\alpha(dK_\beta/dt)$:

$$\frac{dK_g}{dt}=\gamma\Big[(-K_\beta(\partial_\alpha K_\alpha)\alpha_i-K_\alpha(\partial_\beta K_\beta)\beta_i-K_m K_g n_i)\times\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')+2K_g(n_i n_k-\alpha_i\alpha_k-\beta_i\beta_k)\int dS'\partial_k J_{ij}(\mathbf{x}-\mathbf{x}')\times n_j'(\mathbf{x}')K_m'(\mathbf{x}')+K_m n_i(\alpha_k\alpha_l+\beta_k\beta_l)\times\int dS'\partial_k\partial_l J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')\Big]. \quad (9)$$

The change in area due to the contraction of the interface is determined from the relative tangential velocity of points on the interface. The normal velocity of a point on the interface is $n_i v_n$, and the rate of change of this velocity along the coordinate direction α is $\partial_\alpha(n_i v_n)$. The rate of change of the interfacial velocity along the direction x_α tangential to the interface is $\alpha_i\partial_\alpha(n_i v_n)$. A similar expression can be derived for the rate of change of the relative velocity in the x_β direction along the interface. The rate of change of area due to interfacial compression is the product of the interfacial area and the sum of the rates of change of velocity in the tangential directions,

$$\frac{1}{A}\frac{\partial_c A}{\partial t}=\gamma(\alpha_i\partial_\alpha+\beta_i\partial_\beta)\times\Big[n_i n_j\int dS' J_{jk}(\mathbf{x}-\mathbf{x}')n_k'(\mathbf{x}')K_m'(\mathbf{x}')\Big]. \quad (10)$$

Using Eq. (6), the rate of change of surface area due to contraction of the surface is

$$\frac{1}{A}\frac{\partial_c A}{\partial t}=\gamma\Big[-K_m n_j\int dS' J_{jk}(\mathbf{x}-\mathbf{x}')n_k'(\mathbf{x}')K_m'(\mathbf{x}')\Big]. \quad (11)$$

The average rate of change of curvature is determined by averaging over the distribution of microscopic configurations of the interface at the point $\mathbf{x}'$ in Eqs. (8) and (9),

$$\Big\langle\int dS' J_{ij}(\mathbf{x}-\mathbf{x}')n_j'(\mathbf{x}')K_m'(\mathbf{x}')\Big\rangle=\int dV' J_{ij}(\mathbf{x}-\mathbf{x}'1)\Big[\int d\mathbf{n}'\int dK_g'\times\int dK_m' A_2(t,\mathbf{n}',K_m',K_g',\mathbf{x}'|\mathbf{n},K_m,K_g,x)n_j' K_m'\Big], \quad (12)$$

where $A_2(\mathbf{n}',K_m',K_g',\mathbf{x}'|\mathbf{n},K_m,K_g,\mathbf{x})dV'd\mathbf{n}'dK_m'dK_g'$ is the area of the interface in the volume dV' about $\mathbf{x}'$ with curvatures between K_m' and $K_m'+dK_m'$, K_g' and $K_g'+dK_g'$ and unit normal in the interval $\mathbf{n}'$ and $\mathbf{n}'+d\mathbf{n}'$ given that there is a surface element with curvature K_m, K_g and unit normal $\mathbf{n}$ at the position $\mathbf{x}$. In order to proceed further, it is necessary to specify a form for the function within the square brackets in Eq. (12). For a homogeneous and isotropic system, this function should be linear in the unit normal $\mathbf{n}_i$ and dependent only on the distance $|\mathbf{x}-\mathbf{x}'|$. Further, it is expected that the function A_2 is close to $\delta(K_m-K_m')\delta(K_g-K_g')\delta(\mathbf{n}-\mathbf{n}')$ if $(\mathbf{x}-\mathbf{x}')$ is small compared to the radius of curvature of the surface. When $(\mathbf{x}-\mathbf{x}')$ becomes large compared to the radius of curvature, the function A_2 should tend to zero. Further, in the absence of any intrinsic length scale in the system, this is only a function of the curvatures K_m and K_g at the point $\mathbf{x}$. Consistent with these expectations, the most general form for the term in square brackets in Eq. (12) is

$$\Big[\int d\mathbf{n}'\int dK_g'\int dK_m'\times A_2(t,\mathbf{n}',K_m',K_g',\mathbf{x}'|\mathbf{n},K_m,K_g,\mathbf{x})n_j' K_m'\Big]=n_j(\mathbf{x})K_m(\mathbf{x})g(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t)+t_j h(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t), \quad (13)$$

where $\mathbf{t}$ is the perpendicular to the unit normal $\mathbf{n}$ in the plane containing $\mathbf{n}$ and $(\mathbf{x}-\mathbf{x}')$, $g(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t)$, and $h(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t)$ are functions of the mean and Gaussian curvature which is presently unspecified. When the integral of Eq. (13) is carried out over the coordinate $\mathbf{x}'$ on the interface, the resulting expression has no dependence on the di-

rection **t** due to symmetry considerations, and the resulting expressions for the average values of the integrals in Eqs. (8), (9), and (11) are

$$\left\langle \int dS' J_{ij}(\mathbf{x}-\mathbf{x}') n_j'(\mathbf{x}') K_m'(\mathbf{x}') \right\rangle = \frac{n_i(\mathbf{x}) K_m(\mathbf{x}) f(K_m(\mathbf{x}),K_g(\mathbf{x}),t)}{\mu}, \tag{14}$$

$$\left\langle \int dS' \partial_k J_{ij}(\mathbf{x}-\mathbf{x}') n_j'(\mathbf{x}') K_m'(\mathbf{x}') \right\rangle = 0, \tag{15}$$

$$\left\langle n_i(\mathbf{x})(\alpha_k\alpha_l+\beta_k\beta_l) \times \int dS' \partial_k\partial_l J_{ij}(\mathbf{x}-\mathbf{x}') n_j'(\mathbf{x}') K_m'(\mathbf{x}') \right\rangle = 0, \tag{16}$$

where

$$f(K_m,K_g,t) = \frac{\mu}{3} \int dV' J_{ii}(\mathbf{x}-\mathbf{x}') g(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t). \tag{17}$$

Note that the function $f(K_m,K_g,t)$ has the dimensions of length, and physically represents a length scale over which the curvature of the interface is correlated. The forms of Eqs. (14) and (15) are evident from symmetry, but Eq. (16) requires further justification. Using the general form

$$\int dS' \partial_k\partial_l J_{ij}(\mathbf{x}-\mathbf{x}') g(K_m,K_g,|\mathbf{x}-\mathbf{x}'|,t) = A(K_m,K_g,t)\delta_{ij}\delta_{kl} + B(K_m,K_g,t)(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) \tag{18}$$

it can easily be inferred, by contracting both sides with $\delta_{ik}\delta_{jl}$ that $A=-4B$. Contracting both sides of Eq. (18) with $\delta_{ij}\delta_{kl}$, we get

$$-30B = \int dS' \partial_k^2 J_{ii}(\mathbf{x}-\mathbf{x}') g(K_m,K_g,|\mathbf{x}-\mathbf{x}'),t). \tag{19}$$

It can easily be verified that $\partial_k^2 J_{ii}(\mathbf{x}-\mathbf{x}')$ for $|\mathbf{x}-\mathbf{x}'|>0$, resulting in Eq. (16). There is some subtlety involved in obtaining $\partial_k^2 J_{ii}(\mathbf{x}-\mathbf{x}')$ for $|\mathbf{x}-\mathbf{x}'|=0$, since the function tends to a delta function at this point. However, since the force exerted on the fluid by the interface is confined to a surface, the normal velocity at the interface can be evaluated as the limit of the velocity in the fluid as the interface is approached along the direction normal to the interface. This limiting value is zero due to Eq. (19), resulting in the identity (16).

Using the relations (14)–(16), the equations for the rate of change of curvature and interfacial area are

$$\left\langle \frac{dK_m}{dt} \right\rangle = \frac{-\gamma(K_m^2-2K_g)K_m f(K_m,K_g,t)}{\mu}, \tag{20}$$

$$\left\langle \frac{dK_g}{dt} \right\rangle = \frac{-\gamma K_g K_m^2 f(K_m,K_g,t)}{\mu}, \tag{21}$$

$$\left\langle \frac{1}{A}\frac{\partial_c A}{\partial t} \right\rangle = \frac{-\gamma K_m^2 f(K_m,K_g,t)}{\mu}. \tag{22}$$

In Eqs. (20)–(22), the *average* $\langle\cdots\rangle$ *is an average over the distribution of the unit normal and curvature adjacent to a point where the curvature and unit normal are specified*, and not an ensemble average over the curvatures and unit normals at a point. To simplify the notation in the following analysis, the angular brackets are not shown explicitly, it being understood that (dK_m/dt), (dK_g/dt), and $(1/A)\times(\partial_c A/\partial t)$ are averaged in the sense noted above.

Though the conservation equation (2) cannot be solved using relations (20)–(22), it is possible to find a characteristic direction for the variables K_m and K_g. The independent variables are transformed from (K_m,K_g) to a new set of variables (K_*,K_m), where the new variable $K_*(K_m,K_g)$ is defined such that

$$\frac{dK_*}{dt} = \frac{\partial K_*}{\partial K_g}\frac{dK_g}{dt} + \frac{\partial K_*}{\partial K_m}\frac{dK_m}{dt} = 0. \tag{23}$$

This implies that the new variable K_* remains a constant at a point on the interface (in the averaged sense noted above). Equation (23) can be easily solved to get the characteristic variable K_*,

$$K_* = \frac{K_g}{\sqrt{K_m^2-4K_g}}. \tag{24}$$

Note that K_* is always real, since $K_m^2-4K_g=(K_\alpha-K_\beta)^2$ is positive. With the introduction of this similarity variable, the area distribution function can be expressed using (t,K_m,K_*) as the three independent variables, and the conservation equation is

$$\frac{dA}{dt} = -\frac{\partial A}{\partial K_m}\frac{dK_m}{dt} - \frac{\gamma K_m^2 A f(K_m,K_*,t)}{\mu}. \tag{25}$$

The conservation equation (25) is still difficult to solve analytically, but an analytical solution can be derived in the limit of large t. In this limit, it is expected that the magnitude of the mean curvature of the interface decreases proportional to $(\mu/\gamma t)$, and that of the Gaussian curvature proportional to $(\mu/\gamma t)^2$. However, the magnitude of the variable K_* remains a constant, since this variable remains unchanged at all points on the interface. This implies that $K_m \ll K_*$ in the late stages of interface coarsening. From Eq. (24) for K_*, it can be inferred that $K_m^2=4K_g$ in the leading approximation in the limit $K_m \ll K_*$, and $K_m^2-4K_g$ decreases as t^{-4} in the late stages (in contrast to the t^{-2} decrease of K_m^2 or K_g). In this case, the conservation equation for the area distribution function reduces to

$$\frac{\partial A}{\partial t} = \left[\frac{K_m^3}{2}\frac{\partial A}{\partial K_m} - K_m^2 A\right]\frac{\gamma f'(K_m,t)}{\mu}. \tag{26}$$

In Eq. (26), $f(K_m,K_*,t)$ has been set equal to $f'(K_m,t)$ for the late stages of coarsening, since it is expected that there is no dependence on K_* in this limit. The conservation equation (26) for the area distribution function can be solved using the similarity variable $\eta=(\gamma K_m t/\mu)$,

$$\frac{\partial A}{\partial \eta} = \left[\frac{\eta^2}{2}\frac{\partial A}{\partial \eta} - \eta A\right]\frac{\mu f'(K_m,t)}{\gamma t}. \tag{27}$$

In Eq. (27), the function $f'(K_m,t)$ is still unknown, and represents a measure of the length over which the correlations in the curvature of the interface are correlated. In order to proceed, it is necessary to make one further approximation regarding the function $f'(K_m,t)$. The function $f'(K_m,t)$ at a point could depend on the local curvature at the point, K_m, as well as the mean curvature of the interface at time t which determines the area distribution about the point under consideration. The exact form has to be determined by solving Eq. (8) self-consistently for the area distribution function. However, one could consider two limiting cases for the function $f'(K_m,t)$.

(1) If we assume that the function $f'(K_m,t)$ at a point depends only on the local value of K_m at that point, then the permitted functional form for $f'(K_m,t)$ is

$$f'(K_m,t)=\frac{f_*}{|K_m|}, \tag{28}$$

where f_* is a constant. In this case, the conservation equation (27) can be solved to obtain

$$A(\eta)=\begin{cases} A_0(2-f_*|\eta|)^2 & \text{for } f_*|\eta|\leqslant 2 \\ 0 & \text{for } f_*|\eta|>2 \end{cases} \tag{29}$$

where A_0 is determined from the normalization condition.

(2) If the function $f'(K_m,t)$ at a point depends only on the time t and not on the curvature at the point, then the only permitted functional form for f is

$$f'(K_m,t)=\frac{\gamma t f_{**}}{\mu}, \tag{30}$$

where f_{**} is a constant. In this case, the distribution function is

$$A(\eta)=\begin{cases} A_0[2-(f_{**}\eta)^2] & \text{for } f_{**}|\eta|\leqslant\sqrt{2} \\ 0 & \text{for } f_{**}|\eta|>\sqrt{2} \end{cases}. \tag{31}$$

This provides two possible functional forms for the area distribution function. These two functions have different forms—one of these, Eq. (30), has negative slope and positive curvature at $\eta=0$, and the other Eq. (31)—has zero slope and a negative curvature at the origin. However, both of these functions are quadratic polynomials in $|\eta|$, and are nonzero only in a finite domain about $\eta=0$. Since these represent extreme cases of dependence of $f'(K_m,t)$ on K_m and t, it is expected that the actual distribution is likely to be in between these two distributions.

III. CONCLUSIONS

The coarsening of a random interface in a fluid is more complex than that in a solid alloy or a magnetic system due to the nonlocal interaction between different sections of the interface, as evident from Eq. (3). While previous studies, have used dimensional analysis to determine the time variation of the characteristic length of the interface, the present analysis is an attempt to obtain a more detailed description using a curvature distribution function, which gives the distribution of the mean and Gaussian curvature at different points on the interface. A phenomenological model which considered only one characteristic curvature was the subject of an earlier publication.[8] In the present paper, a microscopic model for the interface coarsening (3) was used as the starting point of the analysis. Two significant results for the dynamics of interface coarsening have been derived using varying degrees of approximation, one of which applies to the entire coarsening process and the other to the late stages.

The first result is that there is a function $K_*=K_g/(K_m^2-4K_g)^{1/2}$ which is invariant as the interface coarsens. This was derived using the general functional form (13) for the distribution of unit normals and curvatures about a point where the unit normal and curvature are fixed, and this is expected to apply for the entire coarsening process. This implies that at the late stages when the mean and Gaussian curvature decay proportional to t^{-1} and t^{-2}, respectively, $K_m^2-4K_g \ll K_m^2$ decays proportional to t^{-4}. Consequently, on average, the difference in the principal curvatures in the orthogonal directions is small compared to the magnitude of the principal curvatures.

Though an invariant characteristic direction was obtained for the curvature of the interface, the area distribution function could not be obtained in general. However, a similarity solution for the distribution function could be obtained in the late stages, when $K_m \ll K_*$ using the leading order approximation $K_m^2=4K_g$ in this limit. An approximation for the functional form of $f'(K_m,t)$, which represents the characteristic length over which the curvature and unit normal of the interface are correlated, was required. Two limiting forms of this characteristic length gave different similarity solution for the distribution function (29) and (31). Though these distribution functions are different, they are both polynomials in the characteristic variable $|\eta|=|\gamma K_m t/\mu|$, and are nonzero only in a finite interval along the η coordinate. It is expected that the actual area distribution function is in between these two forms. The distribution function (29) is also of the form anticipated in an earlier publication,[8] though the exponent a, which was a phenomenological constant in that analysis, is fixed by the microscopic model in the present case.

[1] J. D. Gunton and G. Droz, *Introduction to the Theory of Metastable and Unstable States*, Lecture Notes in Physics Vol. **183** (Springer, Berlin, 1983).
[2] T. Ohta, D. Jasnow, and K. Kawasaki, Phys. Rev. Lett. **49**, 1223 (1082).
[3] A. J. Bray, in *Phase Transition and Relaxation in Systems with Competing Energy Scales*, edited by T. Riste and D. Sherrington (Kluwer, Dordrecht, 1993).
[4] E. D. Siggia, Phys. Rev. A **20**, 595 (1979).
[5] H. Furukawa, Adv. Phys. **34**, 703 (1985).
[6] H. Tanaka, J. Chem. Phys. **105**, 10099 (1996).
[7] N. C. Wong and C. M. Knobler, Phys. Rev. A **24**, 3205 (1981).
[8] V. Kumaran, J. Chem. Phys. **108**, 3038 (1998).