Mean field equation for the curvature-driven motion of bicontinuous, random interfaces

Hiroyuki Tomita
Department of Physics and Beckman Institute, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801
*and Department of Fundamental Sciences, Faculty of Integrated Human Studies, Kyoto University, Sakyo, Kyoto 606-01, Japan**
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A mean field theoretical expression for the interface velocity in the phase separating system of symmetric binary mixtures is derived with use of the Green function method. It is assumed that the ramified random bicontinuous interface is almost a minimal surface in the limit of a linear theory with respect to the mean curvature. The formula shows a surface diffusion as well as the usual evaporation-condensation process in the long range limit. Both processes depend on the characteristic scaling length $\lambda(t)$ which is used as a cutoff or a screening length of the bulk diffusion, and both obey the $t^{1/3}$ evolution law. On the contrary, in the short range limit, i.e., in the range smaller than $\lambda(t)$ and greater than the interface thickness ξ, a cutoff-independent motion compatible with the q^3 dispersion relation is obtained. The gradient dynamics related to the present dynamics is discussed qualitatively with use of a localized dissipation function as the definition of the inner product.

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I. INTRODUCTION

The problem of the phase ordering process associated with the first order phase transition has a long history [1,2], but is a still challenging nonlinear problem. Especially, the interface dynamics has become interesting not only to physicists but also to mathematicians [3]. However, though many remarkable phenomena, e.g., the time-dependent scaling law, have been investigated in detail, the interface evolution equation itself of the order parameter conserving system has not been well established in a sense, as is discussed below.

Let us consider the late stage of the ordering process of quenched, symmetric binary mixtures described by the Cahn-Hilliard equation [4],

$$\frac{\partial}{\partial t}s(\mathbf{r},t)=L\nabla^2\mu(\mathbf{r},t), \tag{1}$$

where

$$\mu(\mathbf{r},t)=-\tfrac{1}{2}s(\mathbf{r},t)[1-s(\mathbf{r},t)^2]-\xi^2\nabla^2 s(\mathbf{r},t) \tag{2}$$

is the chemical potential and ξ is a smallness parameter which is to be related with the interface thickness soon. Here we consider the case of a constant transport coefficient L. Then the late stage evolution is described by a bulk diffusion limited interface motion, i.e., the evaporation-condensation process [5]: Let δs be the deviation of the order parameter s from the so-called kink solution,

$$s_K(u)=\tanh\left(\frac{u}{2\xi}\right), \tag{3}$$

which is the steady solution of Eqs. (1) and (2) in the case of a planar interface where u is the normal coordinate perpendicular to it and thus ξ corresponds to the interface thickness. When the geometrical characteristic length λ, e.g., the representative radius of curvature of the interface S, satisfies $\lambda \gg \xi$, the deviation δs can be assumed to be small and by linearizing Eqs. (1) and (2) we obtain a diffusion equation,

$$\frac{\partial}{\partial t}\delta s(\mathbf{r},t)=L\nabla^2\delta s(\mathbf{r},t), \tag{4}$$

with the Gibbs-Thomson boundary condition

$$\delta s|_S=\tfrac{1}{3}\xi H, \tag{5}$$

where H is the mean curvature of S defined by $H=-\nabla\cdot\mathbf{n}$ with the unit normal vector $\mathbf{n}=\nabla s_K/|\nabla s_K|$ at the interface $(u=0)$. Note that in the present notations the surface tension is given by $\gamma=2\xi/3$. The interface normal velocity is given by

$$v_n=-\frac{L}{2}[\mathbf{n}\cdot\nabla\delta s]_S, \tag{6}$$

where $[\ldots]_S$ denotes the flux gap across the interface S. Thus the late stage of the evolution is described as a Stefan problem consisting of Eqs. (4)–(6). In the present case, however, the interface velocity $v_n\sim L\delta s/\lambda$ is very small compared with the representative diffusion velocity $v_D\sim L(\delta s/\lambda)/\delta s\sim(\lambda/\xi)v_n$, and a quasistatic approximation

$$\nabla^2\delta s=0 \tag{7}$$

is applicable. It should be noted here that in this stage of evolution the order parameter conservation is expressed as the volume conservation, i.e.,

$$\oint_S v_n(a)\,da=0, \tag{8}$$

*Permanent address.
Electronic address: tomita@phys.h.kyotou.ac.jp

where da is the surface element of the interface S, and S is regarded as a closed surface in the meaning as will be remarked in Sec. III. One more conserved quantity proper to this stage is a "dipole" moment [6] which is a measure of uniformity of the geometric pattern defined by

$$\mathbf{p}=\int \mathbf{r}[s(\mathbf{r})-\bar{s}]dV, \tag{9}$$

where $\bar{s}$ is the bulk average of the order parameter s. With use of Eqs. (6) and (7) it can be shown that

$$\frac{d\mathbf{p}}{dt}=2s_0 \oint_S \mathbf{r}v_n(a)da=\mathbf{0}, \tag{10}$$

where $2s_0$ is the order parameter gap ($s_0=1$ here). This fact shows that, though the characteristic length $\lambda(t)$ grows as $\lambda(t)\sim t^{1/3}$ in this stage as is generally shown using a kind of dimension analysis [7], the dipole moment defined by Eq. (9) remains constant and is not scaled. That is, the uniformity of the system is preserved in the scaling stage whenever the initial state is uniform [6].

This simplified scheme was used by Mullins and Sekerka [8] to investigate the instability in solidification. Kawasaki and Ohta [7] derived an integral equation form which is just the same as the equation for the equivalent simple layer for the above Dirichlet problem in the potential theory. A systematic derivation with an asymptotic expansion was given by Pego [9] and recently by Oono [10] with use of the renormalization group method. It should be noted that Kawasaki and Ohta's derivation is essentially based on the Onsager variational principle [11] by rewriting the free energy and the dissipation function for the diffusion process in the interfacial forms. Thus, speaking in the currently familiar language, the evaporation-condensation dynamics is the gradient dynamics [3] with use of the interface version of the positive definite dissipation function as the definition of the corresponding inner product,

$$\Phi(v,w)=\frac{(2s_0)^2}{2L}\oint_S \oint_S G_0(\mathbf{x}(a),\mathbf{x}(a'))\times v(a)w(a')da\,da', \tag{11}$$

where $G_0(\mathbf{x},\mathbf{x}')$ is the Green function for the three dimensional Laplacian ∇^2 defined by

$$G_0(\mathbf{x},\mathbf{x}')=\frac{1}{4\pi|\mathbf{x}-\mathbf{x}'|}. \tag{12}$$

If the Green function for the surface Laplacian ∇_S^2 was used instead, a surface diffusion motion would be obtained [3].

Although the above scheme has been well established and investigated exhaustively, we have not found yet an explicit formula for the interface velocity v_n itself, like the Allen-Cahn formula [12] for the order parameter nonconserving system. We have no general solution for the Dirichlet problem in an explicit form in the potential theory. That is, none of explicit solutions can be found unless the actual geometry of the surface is incorporated into the theory anyway, e.g., like the image method in electrostatics. Successful examples are seen in the early works by Todes [13], Lifshitz and Slyozov [14], and Wagner [15] for the spherical droplet system of very small volume fraction $\phi\ll 1$ and by Mullins and Sekerka [8] for the interface of simple geometry. The difficulty in the system prepared with the nearly critical quench ($\phi\sim 0.5$) is the geometrical complexity of its spatial pattern, i.e., a ramified bicontinuous phase or a random sponge structure observed by simulations [16–18]. Recently a direct picture of this three dimensional structure was obtained experimentally by using the position-sensitive atomic probe (PoSAP) on Fe-Cr alloy [19].

The author took advantage of the following properties of this structure into a phenomenological theory [20]: (i) The spatial structure of it is mesoscopically uniform because of the conservation law, Eq. (10), and has a well defined characteristic length scale $\lambda(t)$, which obeys the $t^{1/3}$ growth law. (ii) The ramified structure causes localization or screening of the diffusion process like the electrostatic shielding phenomena. The screening length must be of order of $\lambda(t)$. (iii) The mean curvature of the interface S is small and slowly varying everywhere.

Using the method of the Green function,

$$\nabla^2 G(\mathbf{x},\mathbf{x}_0)=\delta(\mathbf{x}-\mathbf{x}_0) \quad \text{and} \quad G=0 \quad \text{on } S, \tag{13}$$

a cutoff length λ was introduced according to the assumption (ii) to

$$\sigma_{\text{ind}}(\mathbf{X};\mathbf{x}_0)=-\mathbf{n}\cdot\nabla G(\mathbf{X},\mathbf{x}_0), \tag{14}$$

which is the surface charge induced on the grounded conductor S by a negative unit charge -1 located at $\mathbf{x}_0$. Hereafter the uppercase $\mathbf{X}$ is used to denote the points on S and the lowercase $\mathbf{x}$ those in the bulk. Based on the above assumptions an explicit expression for v_n was derived in an expansion form with respect to the surface gradient operator ∇_S on the mean curvature H up to the second order. However, this expansion is incomplete, because the correlation length of the curvature H must be comparable to the representative geometrical length λ. Besides, there exists ambiguity in the range of validity of the adopted form of the charge density σ_{ind}, which was estimated from the planar conductor problem. The purpose of the present paper is to improve this phenomenology by replacing the assumption (iii) by the following: (iii′) The mean curvature is small everywhere and the interface S can be regarded as an almost minimal surface when $\phi=0.5$.

This means that

$$H^2\ll|K|\sim\lambda^2, \tag{15}$$

everywhere. Here K is the Gauss curvature which is negative almost everywhere. Since the mean curvature H is included already in the boundary condition, Eq. (5), in our problem, we can regard the interface as a minimal surface ($H=0$) within the linear theory in H.

In Sec. II the potential theory for the minimal surface is surveyed and a good foundation for σ_{ind} used in the previous papers [20] is obtained. With use of this σ_{ind} a mean field theoretical expression for the interface velocity v_n and the corresponding u field [21,22] or the contour equation [23] are derived in Sec. III. The relation with the gradient dynamics is discussed in Sec. IV. Prescaling behavior, which as-

sures the geometrical assumptions enumerated above in the scaling stage and a geometrical instability which is likely a growing mechanism of the pattern, are also discussed there.

II. GREEN FUNCTION FOR MINIMAL SURFACES

Let us survey the potential problem for the minimal surface in this section. We have the Laplace equation

$$\nabla^2\psi(\mathbf{x})=0, \tag{16}$$

with a Dirichlet-type boundary condition,

$$\psi=\psi(\mathbf{X}) \quad \text{on S.} \tag{17}$$

In terms of the induced charge density defined by Eq. (14) a formal solution is given by

$$\psi(\mathbf{x})=\oint \psi(\mathbf{X})\sigma_{\text{ind}}(\mathbf{X};\mathbf{x})da. \tag{18}$$

Note that in the present problem we need only the gradient of the potential at the surface S in Eq. (6). [Of course the Dirichlet problem would be completely solved, if $\mathbf{n}\cdot\nabla\psi(\mathbf{X})$ on S would be found.] This means that we need the Green function G defined by Eq. (13) only in the very vicinity of S, i.e., in the limit $z\to 0$ when we set $\mathbf{x}=\mathbf{X}_0+z\mathbf{n}$.

Let (u_1,u_2,u_3) be an orthogonal curvilinear system and define the linear metric by

$$g_i=|\partial\mathbf{x}/\partial u_i|=1/|\nabla u_i|. \tag{19}$$

Let u_3 be the normal coordinate perpendicular to S and $g_3=1$, and (u_1,u_2) be the orthogonal local coordinates of S which is not necessarily a minimal surface for the time being. In this coordinate system the Laplacian is written as

$$\nabla^2=\frac{1}{\epsilon}\left(\frac{\partial}{\partial u_1}\frac{g_2}{g_1}\frac{\partial}{\partial u_1}+\frac{\partial}{\partial u_2}\frac{g_1}{g_2}\frac{\partial}{\partial u_2}\right)+\frac{\partial^2}{\partial u_3^2}-H\frac{\partial}{\partial u_3}, \tag{20}$$

where $\epsilon=g_1g_2$, and the mean curvature H is defined by

$$H=-\nabla\cdot\mathbf{n}=-\frac{1}{\epsilon}\frac{\partial\epsilon}{\partial u_3}. \tag{21}$$

It is always possible to find a conformal map for a local vicinity of any smooth surface into that of a plane because we can set $g_1=g_2=1$ locally. The range of its validity is given by

$$|\nabla G|/G\sim 1/r\gg|H|. \tag{22}$$

However, this condition on the transverse distance r imposes no restriction on the minimal surface where $H=0$ everywhere: The condition (22) becomes

$$1/r\gg|u_3K|, \tag{23}$$

because the mean curvature of a surface $u_3=u$ (a parallel surface of S [24]) is given by

$$H(u)=\frac{H(0)-2uK(0)}{1-uH(0)+u^2K(0)}, \tag{24}$$

where $H(0)=0$. As is mentioned above, we need the limit $u_3\to 0$. Therefore, we have practically no restriction on r. Using Eqs. (21) and (24) one finds

$$\epsilon(u)=\frac{\epsilon(0)}{1-uH(0)+u^2K(0)}. \tag{25}$$

Then we can assume

$$H(u_1,u_2,u_3)\simeq 0 \quad \text{and} \quad \epsilon(u_1,u_2,u_3)\simeq\epsilon(u_1,u_2,0) \tag{26}$$

in the very vicinity of the minimal surface so long as $|u_3|\ll 1/\sqrt{|K(0)|}$. Further, we can define an isometric orthogonal system $g_1=g_2$ (to a certain extent) globally on the minimal surface by choosing the pair of parameters of lines of curvature as (u_1,u_2) [24]. This is approximately valid in the vicinity of the minimal surface again. Thus Eq. (13) for the Green function G for the minimal surface S is well approximated by

$$\left(\frac{\partial^2}{\partial u_1^2}+\frac{\partial^2}{\partial u_2^2}+\epsilon(u_1,u_2)\frac{\partial^2}{\partial u_3^2}\right)G=\delta(u_1)\delta(u_2)\delta(u_3-z), \tag{27}$$

where the source strength is not changed by choosing a frame $\epsilon(0,0)=1$, i.e., a $3d$-isometric system at the source point $(0,0,z)$. This is the Poisson equation for an anisotropic medium with a diagonal dielectric tensor. In this medium the refraction of flux lines occurs only in u_3 direction. If we introduce a new distance by

$$r'(r,\varphi)=\int_0^r\epsilon(\rho\cos\varphi,\rho\sin\varphi)d\rho, \tag{28}$$

where (r,φ) are the polar coordinates in the (u_1,u_2) plane, all flux lines diverging from $(0,0,z)$ are mapped into straight lines. It should be noted, however, that this dilation in the (u_1,u_2) plane is *not* conformal. The new plane (u_1',u_2') is solely a reference frame for the geometrical computation of the solid angle spreading from the source point $(0,0,z)$. The number of flux lines (i.e., the solid angle) which start from $(0,0,z)$ where $\epsilon=1$ and are *directed* to a surface element $du_1'du_2'$ located by distance r' from $(0,0)$ is given by

$$d\Omega=\frac{z\,du_1'du_2'}{(r'^2+z^2)^{3/2}}, \tag{29}$$

Then multiplying by a factor of 2 due to the corresponding image charge we get

$$\sigma_{\text{ind}}da=-\frac{\partial G}{\partial u_3}da=\frac{z}{2\pi(r'^2+z^2)^{3/2}}du_1'du_2', \tag{30}$$

where $du_1'du_2'=\epsilon^2du_1du_2=\epsilon\,da$ is used. Thus the form of the induced charge density assumed in the previous work [20] is established for a new plane. The range of validity of it is not restricted within the neighborhood of the point $(0,0)$ so long as the parametric plane (u_1,u_2) originated at $(0,0)$ covers the whole $2d$ plane R^2. However, this is not always true. Especially in such a ramified bicontinuous phase as considered here, the mapping may cover only a restricted

region of R^2 depending on the reference point (0,0). This restriction will be used as a cutoff in the next section. Another difficulty is that there remain unmapped regions on S also and the induced charge on these remaining parts induces additional charges and deforms Eq. (30). This indirect effect is assumed to be very small from a viewpoint of the electrostatic shielding and will be neglected.

Although the form of the induced charge density Eq. (30) is derived strictly based on the assumption of a minimal surface when $\phi=0.5$, the result itself may be approximately applicable so long as the inequality Eq. (15) is valid. Henceforth let us extend it to nearly symmetric systems ($\phi\sim 0.5$). Of course, if we restrict the range of its application within a sufficiently narrow neighborhood of the reference point (0,0), it must be always a good approximation. The important condition we need here, however, is that the range of its validity should be comparable to the representative length λ in order to apply the mean field approximation to the outer range ($>\lambda$) in the next section.

III. MEAN FIELD THEORY OF INTERFACE MOTION

First, note that we should be careful in considering the potential problem with an infinitely extended boundary S such as the bicontinuous phase assumed in the present problem and also the outer region of an infinitely distributed sphere system. Precisely speaking on physically uniform systems only, we have a compatibility condition on the surface average of the boundary value,

$$\overline{\psi_S}=0, \tag{31}$$

instead of the usual boundary condition $\lim_{\mathbf{X}\to\infty}\psi(\mathbf{X})=0$ in order to use Green's theorem. The infinitely extended interface S can be treated as a closed surface in this sense. Note that all of the useful formulas for the potential problem, e.g., the induced charge method used in Sec. II, the equivalent simple layer or double layer methods and so on, are derived from Green's theorem. Therefore, if we wish to employ a potential theoretical approach to the present problem, the boundary condition (5) should be modified to

$$\psi(\mathbf{X})=\xi[H(\mathbf{X})-\bar{H}]/3, \tag{32}$$

where

$$\bar{H}=\oint_S H(\mathbf{X})da \Big/ \oint_S da. \tag{33}$$

Let us apply the result obtained in Sec. II to the formula

$$\psi(\mathbf{X}+z\mathbf{n})=\oint_S \psi(\mathbf{X}+\mathbf{X}_1)\sigma_{\text{ind}}(\mathbf{X}+\mathbf{X}_1;\mathbf{X}+z\mathbf{n})da_1, \tag{34}$$

to calculate

$$\mathbf{n}\cdot\nabla\psi(\mathbf{X})=\lim_{z\to 0}\frac{\psi(\mathbf{X}+z\mathbf{n})-\psi(\mathbf{X})}{z}. \tag{35}$$

As is assumed in the previous work [20], in the present random sponge phase the electrostatic induction by a point charge -1 must be well localized within the effective cage of radius $\sim\lambda$ due to the electrostatic shielding because of the ramified shape of the conductor surface S. In fact, the induction must be negligibly small at least in the geometrically shaded region. The range of the parametric mapping remarked in Sec. II, if exists, may be the same order as it. As we have no definite definition for this characteristic length yet, let us introduce a cutoff length λ which is uniform measured in the (u_1',u_2') plane, and divide the integral in Eq. (34) into two parts as

$$\oint_S=\int_{<\lambda}(\text{local part})+\int_{>\lambda}(\text{nonlocal part}). \tag{36}$$

As is examined afterward the nonlocal part does not affect the interface velocity because of the symmetry after random averaging. In the local part let us use the expression (30) for σ_{ind}, that is, let us neglect the indirect induction from the outer region ($>\lambda$) because the total charge induced there is supposed to be very small [$\sim O(z/\lambda)$]. Then the Taylor expansion for $\psi(\mathbf{X}+\mathbf{X}_1)$ in the (u_1',u_2') plane gives

$$\psi(\mathbf{X}+z\mathbf{n})\simeq\left(1-\frac{z}{\lambda}V(-\lambda^2\nabla_S^2)\right)\psi(\mathbf{X}), \tag{37}$$

where V is a function defined in terms of the Bessel functions by

$$V(Q^2)=-\sum_{n=0}^{\infty}\frac{1}{(2n-1)2^{2n}(n!)^2}(-Q^2)^n$$
$$=J_0(Q)-QJ_1(Q)+Q\int_0^Q J_0(q)dq. \tag{38}$$

Note that in the Taylor expansion the operator $(\partial/\partial u_1')^2+(\partial/\partial u_2')^2$ is replaced by the surface Laplacian ∇_S^2 because we used the frame $\epsilon(0,0)=1$ in Sec. II. With the aid of Eqs. (6), (32), and (35), Eq. (37) yields the final result,

$$v_n(\mathbf{X})=-\frac{L\xi}{3\lambda(t)}V(-\lambda(t)^2\nabla_S^2)[H(\mathbf{X})-\bar{H}], \tag{39}$$

where the cutoff length λ is regarded as a time-dependent parameter which should be determined self-consistently by Eq. (39) itself. Alternatively, we may set $\lambda(t)=ct^{1/3}$ with an adjustable constant c conveniently in so far as we know that it obeys the $t^{1/3}$ evolution law.

The function $V(Q^2)$ is expanded for small Q as

$$V(Q^2)\simeq 1+\frac{Q^2}{4}+\cdots, \tag{40}$$

which was obtained by the author in Ref. [20]. The first term denotes directly the evaporation-condensation process ($\sim[H-\bar{H}]$) due to the fluctuation of the mean curvature around its average value. The second term may be interpreted to be a type of surface diffusion ($\sim-\nabla_S^2H$) due to the spatial variation of the curvature. Both processes depend on the parameter $\lambda(t)$ and the interface motions caused by them obey the $t^{1/3}$ law. Then this surface diffusion differs from that

obtained by inhibiting the bulk diffusion [3]. The interface motion associated with the latter obeys the $t^{1/4}$ law.

In contrast for large Q we have

$$V(Q^2)\simeq Q, \tag{41}$$

which is a good approximation practically for $Q>2$. Owing to $\lambda(t)$ in the denominator in Eq. (39) the interface velocity $v_n\simeq(L\xi/3)|\nabla_S|H$ is independent of the cutoff length $\lambda(t)$. This form was first used by Ohta and Nozaki [25] and derived by Hayakawa and Koga [26] as a limit of more general results for long range interactions assuming an almost planar interface. Since $H\sim q^2$, where q is the wave number corresponding to Q/λ, this behavior is compatible with the q^3 dispersion relation of the surface mode derived by Jasnow and Zia [27] and by Shinozaki and Oono [28]. Of course, q should be sufficiently smaller than ξ^{-1} as is limited in the present theory.

Now let us consider the nonlocal part. The induced charge density obeys the following integral equation by definition ($G=0$ on S):

$$\tfrac{1}{2}\sigma_{\rm ind}(\mathbf{X}_1;\mathbf{X}+z\mathbf{n})=-(\mathbf{n}_1\cdot\nabla_1)\Big(-G_0(\mathbf{X}_1,\mathbf{X}+z\mathbf{n})+\oint_S G_0(\mathbf{X}_1,\mathbf{X}+\mathbf{X}')\times\sigma_{\rm ind}(\mathbf{X}+\mathbf{X}';\mathbf{X}+z\mathbf{n})da'\Big), \tag{42}$$

where ∇_1 denotes $\partial/\partial\,\mathbf{X}_1$. Let $\mathbf{X}_1$ be a point in the outer region ($>\lambda$) and divide the integral in the right-hand side into the inner part $\int_{<\lambda}$ and the outer part $\int_{>\lambda}$. Using the direct induction form, Eq. (30), as $\sigma_{\rm ind}$ in $\int_{<\lambda}$, one obtains

$$\begin{aligned}\tfrac{1}{2}\sigma_{\rm ind}(\mathbf{X}_1;\mathbf{X}+z\mathbf{n})&=-(\mathbf{n}_1\cdot\nabla_1)\Big[-G_0(\mathbf{X}_1,\mathbf{X}+z\mathbf{n})+\Big(1-\frac{z}{\lambda}V(-\lambda^2\nabla_S^2)\Big)G_0(\mathbf{X}_1,\mathbf{X})+\int_{>\lambda}\cdots\Big]\\&=(\mathbf{n}_1\cdot\nabla_1)\Big[z\Big((\mathbf{n}\cdot\nabla)+\frac{1}{\lambda}V(-\lambda^2\nabla_S^2)\Big)G_0(\mathbf{X}_1,\mathbf{X})-\int_{>\lambda}G_0(\mathbf{X}_1,\mathbf{X}+\mathbf{X}')\sigma_{\rm ind}(\mathbf{X}+\mathbf{X}';\mathbf{X}+z\mathbf{n})da'\Big].\end{aligned} \tag{43}$$

Define a unit dipole potential at $\mathbf{x}'$ caused by a dipole $\mathbf{n}$ located at $\mathbf{X}$ by

$$F_0(\mathbf{x}';\mathbf{X})=\frac{\mathbf{n}\cdot(\mathbf{x}'-\mathbf{X})}{4\pi|\mathbf{x}'-\mathbf{X}|^3}=\mathbf{n}\cdot\nabla G_0(\mathbf{X},\mathbf{x}'). \tag{44}$$

Then by substituting $\sigma_{\rm ind}$ in Eq. (43) iteratively, the flux gap including the effect of the outer region is finally given by

$$[\mathbf{n}\cdot\nabla\psi]_S=-\frac{2}{\lambda}V(-\lambda^2\nabla_S^2)\Big(\psi(\mathbf{X})-\frac{\Psi_\lambda^+(\mathbf{X})+\Psi_\lambda^-(\mathbf{X})}{2}\Big)+\mathbf{n}\cdot\nabla[\Psi_\lambda^+(\mathbf{X})-\Psi_\lambda^-(\mathbf{X})], \tag{45}$$

where

$$\Psi_\lambda^\pm(\mathbf{x})=\pm\int_{>\lambda}F_0(\mathbf{x};\mathbf{X}_1)\tau_\lambda^\pm(\mathbf{X}_1)da_1 \tag{46}$$

are double layer potentials and their sources τ_λ^+ and τ_λ^- are given by

$$\tfrac{1}{2}\tau_\lambda^\pm(\mathbf{X})=\psi(\mathbf{X})\mp\int_{>\lambda}F_0(\mathbf{X};\mathbf{X}_1)\tau_\lambda^\pm(\mathbf{X}_1)da_1. \tag{47}$$

Note that this integral equation is just the same as that for the equivalent double layer for the Dirichlet problem in the potential theory, if the restriction ($>\lambda$) in the integral is replaced by a principal value integral excluding the point $\mathbf{X}_1=\mathbf{X}$. Since this definition of $\tau_\lambda^\pm$ is in a sense an averaging over the random variable $\psi(\mathbf{X})\sim[H(\mathbf{X})-\bar{H}]$ in the outer region, the symmetry consideration allows us to assume the double layer potentials Ψ_λ^+ and Ψ_λ^- to be

$$\Psi_\lambda^\pm(\mathbf{x})\simeq\pm\tfrac{1}{2}\langle\tau_\lambda\rangle\Big(1\pm\frac{z}{\sqrt{\lambda^2+z^2}}\Big)\simeq\pm\tfrac{1}{2}\langle\tau_\lambda\rangle\Big(1\pm\frac{z}{\lambda}\Big), \tag{48}$$

where we put $\mathbf{x}=\mathbf{X}+z\mathbf{n}$ and $\langle\tau_\lambda^+\rangle\sim\langle\tau_\lambda^-\rangle\sim\langle\tau_\lambda\rangle$, and the approximations are based on the calculation of the solid angle of the "window" part ($<\lambda$) and are solely guidelines to estimate z and λ dependence. Hence the outer part has no effect on the interface equation at $z=0$, but it must be taken into account in considering the deviation of the interface ($z\neq0$). That is, if we introduce the so-called u-field [21,22] defined by

$$s(\mathbf{r},t)=\mathrm{sgn}(u(\mathbf{r},t)), \tag{49}$$

its evolution equation, or the contour equation [23], is given by

$$\frac{1}{|\nabla u|}\frac{\partial u}{\partial t}=\frac{L\xi}{3\lambda(t)}V(-\lambda(t)^2\nabla_S^2)\frac{1}{|\nabla u|}\Big(\nabla_S\cdot\nabla+\frac{\kappa^2}{\lambda(t)^2}\Big)u, \tag{50}$$

where the definition of the mean curvature,

$$H=-\nabla\cdot\mathbf{n}=-\frac{1}{|\nabla u|}\nabla_S\cdot\nabla u, \tag{51}$$

is used. The dimensionless constant κ is defined by putting

$$\langle\tau_\lambda\rangle=-\frac{\xi\kappa^2}{3\lambda(t)}\quad(<0), \tag{52}$$

i.e., by assuming an effectively concave mean field environment and the scaling property $H\sim\lambda(t)^{-1}$. Thus the long range coupling by the diffusion process yields the time-dependent unstable range of small wave numbers, which is analogous to the Mullins-Sekerka instability in solidification [8] and is a well-known remarkable feature of the spinodal decomposition. This instability was attributed to ∇_S^2 in $V(-\lambda^2\nabla_S^2)$ by the present author [20], but the argument is incorrect. The mean field theoretical treatment on it employed here is just equivalent to those used by Ohta and Nozaki [25] and Hayakawa and Koga [26] for nearly planar interfaces.

If we assume a Gaussian random field for $\{u(\mathbf{r})\}$, we can calculate the correlation function $g(\mathbf{r},t)=\langle s(0)s(\mathbf{r})\rangle_t$ and the corresponding structure function $S(\mathbf{q},t)$ together with a kind of random phase approximation for ∇_S^2 [21]. The structure function has been calculated by Ohta and Nozaki for $V(Q^2)\simeq Q$ [25] and by the present author for $V(Q^2)\simeq 1+Q^2/4$ [20]. An evolution equation similar to the latter is obtained by Yeung, Oono, and Shinozaki [29] based on the scaling hypothesis within the Gaussian approximation. However, as is clearly demonstrated by them, the Gaussian approximation itself includes a severe conflict with the order parameter conservation, i.e., in the small wave number limit $\lim_{\mathbf{q}\to 0}S(\mathbf{q},t)$. A post-Gaussian correction on this fault is proposed by Mazenko [30]. Further, in the finite wave number region an apparent discrepancy of the position of the second peak in the Porod plot for the structure function is found between the theory [20,25] and the simulation [18,31]. Even if we use the present evolution equation (50), little improvement is found with respect to this fault. Thus a breakthrough beyond the Gaussian closure is desired to reproduce the observables $g(\mathbf{r},t)$ and $S(\mathbf{q},t)$ more faithfully for finite wave numbers $\mathbf{q}$ as well as the limit $q\to 0$.

IV. DISCUSSION

The mean field theoretical expression for the interface evolution equation in the random bicontinuous phase is derived by assuming the screening effect of the diffusion process on the analogy of the electrostatic shielding. It is analogous to the screening length for the droplet-droplet correlation introduced by Tokuyama and Kawasaki [32] to extend the Lifshitz-Slyozov theory to the system of finite volume fraction ϕ. So far a simple cutoff procedure employed here seems to be the best to incorporate the screening effect for the random sponge structure of $\phi\sim 0.5$, because other phenomenological functions, e.g., the Debye-Hückel-type exponential screening adopted in the droplet system [33,34], never yield the behavior $V(Q^2)\simeq Q$ which coincides with the q^3 dispersion relation.

The random sponge structure observed in the simulations [16,18] resembles the periodic minimal surface [35] so far as we are looking at it locally. Then it is assumed here that the interface is almost minimal surface within the linear theory with respect to the mean curvature. However, this nature ($K<0$ almost everywhere) itself cannot be deduced from the evolution equation at this stage. The equation obtained here is merely compatible with it. The origin of it should be attributed to the initial phase, or more exactly, to the prescaling behavior. In this context it should be noted that, when the characteristic length λ is not so much larger than the interface thickness ξ, the interface free energy ΔF has a correction term of order ξ^2, i.e.,

$$\Delta F\simeq\gamma\left(\oint_S da+\beta\xi^2\oint_S K(a)da\right), \tag{53}$$

for $d=3$, [36] where γ is the surface tension and β is a positive constant dependent on the interface profile, e.g., $\beta=1.289\ldots$ for Eq. (3). Then it is concluded that the sponge structure ($K<0$ almost everywhere) must be preferred in the prescaling stage. Thus the prescaling dynamics seems to be inevitable in order to reveal the scaling structure.

Another problem associated with this structure is how it grows. The characteristic length $\lambda(t)$ cannot grow (as $t^{1/3}$) unless any topological jump occurs. Let us discuss here one likely mechanism, that is, the catenoid instability. As is well known, catenoid and circular film are only two types of revolving minimal surface [35]. The stability of the catenoid depends on its aspect ratio. When the ratio [height]/[minimal radius] is greater than $1.199\ldots$, it becomes unstable (*not* minimal though $H=0$). With use of the present dynamics it can be shown that the dynamical stability coincides with this geometrical stability within a linear analysis. Suppose a catenoidlike part in the sponge structure happens to be drawn along its axial direction by some environmental deformation. It suddenly begins to diminish when its aspect ratio reaches the above critical value, and disappears in a short time, i.e., is transformed into another minimal surface, a pair of circular films. Then there occurs a topological jump.

Lastly, let us discuss the possibility of the gradient dynamics [3] related to the present dynamics. As is surveyed in Sec. I, the late stage dynamics of the Cahn-Hilliard equation with a constant transport coefficient L, i.e., the evaporation-condensation dynamics, is interpreted as a gradient dynamics with use of the interface version of the Onsager dissipation function as the definition of the inner product. With use of Eq. (11) the interface velocity is given by

$$v_n(\mathbf{X})=\frac{\gamma L}{(2s_0)^2}\oint_S\Gamma_0(\mathbf{X},\mathbf{X}')[H(\mathbf{X})-\bar{H}]da', \tag{54}$$

where $\gamma=2\xi/3$ and $\Gamma_0(\mathbf{X},\mathbf{X}')$ is the inverse of the Green function $G_0(\mathbf{X},\mathbf{X}')$ on the interface S and is *not* the $3d$ Laplacian [7]. The $\bar{H}$ term in this case results from the Lagrange multiplier term for the order parameter conservation constraint Eq. (8). However, this equation is merely formal and physically useless, unless the actual geometry of the given problem is inevitably taken into account. If it is re-

membered that the inner product, Eq. (11), is the Coulomb interaction energy of the surface charges in terms of electrostatic language, we may approximate it by a localized form with use of some kind of an effective screened potential $\tilde{G}(\mathbf{X},\mathbf{X}')$ corresponding to the approximation of the present work. Then its inverse $\tilde{\Gamma}(\mathbf{X},\mathbf{X}')$ also becomes a similarly localized one, though it may not have such a sharp cutoff as introduced in Sec. III. Thus it is expected that a more reasonable derivation of the interface evolution equation for the random sponge phase will be obtained in this course.

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[1] J. D. Gunton and M. Droz, *Introduction to the Theory of Metastable and Unstable States* (Springer, Berlin, 1983).
[2] J. S. Langer, *Models of Pattern Formation in First Order Phase Transitions* (World Scientific, Singapore, 1986).
[3] J. W. Cahn and J. E. Taylor, Acta Metall. Mater. **42**, 1045 (1994).
[4] J. W. Cahn and J. E. Hilliard, J. Chem. Phys. **28**, 258 (1958).
[5] The following survey is quite in accordance with A. Onuki, *Dynamics of Phase Transition* (Iwanami, Tokyo, 1992) (in Japanese).
[6] H. Tomita, Prog. Theor. Phys. **85**, 47 (1991).
[7] K. Kawasaki and T. Ohta, Prog. Theor. Phys. **67**, 147 (1982); **68**, 129 (1982); Physica **118A**, 175 (1983).
[8] W. W. Mullins and R. F. Sekerka, J. Appl. Phys. **34**, 323 (1963).
[9] R. L. Pego, Proc. R. Soc. London Ser. A **422**, 261 (1989).
[10] Y. Oono (unpublished).
[11] L. Onsager, Phys. Rev. **37**, 405 (1931); **38**, 2265 (1931); L. Onsager and S. Machlup, *ibid.* **91**, 1505 (1953).
[12] S. M. Allen and J. W. Cahn, Acta Metall. **27**, 1085 (1979).
[13] O. M. Todes, J. Phys. Soc. (Sov. Union) **20**, 629 (1946).
[14] I. M. Lifshitz and V. V. Slyozov, J. Phys. Chem. Solids **19**, 35 (1961).
[15] C. Wagner, Z. Elektrochem. **65**, 581 (1961).
[16] A. Chakrabarti, R. Toral, and J. D. Gunton, Phys. Rev. B **39**, 4386 (1989).
[17] Y. Oono, IEICE Trans. E **74**, 1379 (1991).
[18] A. Shinozaki and Y. Oono, Phys. Rev. E **48**, 2622 (1993).
[19] A. Cerezo, J. M. Hyde, R. P. Setna, and G. D. W. Smith (unpublished).
[20] H. Tomita, Prog. Theor. Phys. **90**, 521 (1993); Physica A **204**, 693 (1994). Note that an opposite direction of $\mathbf{n}$ and a definition $H=1/2\nabla\cdot\mathbf{n}$ are used here.
[21] T. Ohta, D. Jasnow, and K. Kawasaki, Phys. Rev. Lett. **49**, 1223 (1982).
[22] T. Ohta, Ann. Phys. (N. Y.) **163**, 31 (1984).
[23] Y. Giga and S. Goto, J. Math. Soc. Jpn. **44**, 99 (1992).
[24] C. E. Weatherburn, *Differential Geometry* (Cambridge University Press, Cambridge, 1930).
[25] T. Ohta and H. Nozaki, in *Space-Time Organization in Macromolecular Fluids*, edited by F. Tanaka *et al.* (Springer, Berlin, 1989).
[26] H. Hayakawa and T. Koga, J. Phys. Soc. Jpn. **59**, 3542 (1990).
[27] D. Jasnow and R. K. P. Zia, Phys. Rev. A **36**, 2243 (1987).
[28] A. Shinozaki and Y. Oono, Phys. Rev. Lett. **47**, 804 (1993).
[29] C. Yeung, Y. Oono, and A. Shinozaki, Phys. Rev. E **49**, 2693 (1994).
[30] G. F. Mazenko, Phys. Rev. E **49**, 3717 (1994).
[31] A. Shinozaki and Y. Oono, Phys. Rev. Lett. **66**, 173 (1991).
[32] M. Tokuyama and K. Kawasaki, Physica A **123**, 386 (1984).
[33] J. A. Marqusee and J. Ross, J. Chem. Phys. **80**, 536 (1984).
[34] H. Tomita, Prog. Theor. Phys. **71**, 1405 (1984).
[35] U. Dierkes, S. Hildebrandt, A. Küster, and O. Wohlrab, *Minimal Surfaces* (Springer-Verlag, Berlin, 1991).
[36] C. Murakami (private communication).