Progress of Theoretical Physics, Vol. 75, No. 3, March 1986

Statistical Properties of Random Interface System

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(Received October 21, 1985)

Statistical properties of binary phase system with semi-macroscopic random interfaces are studied. Singularity and symmetry of the correlation function are discussed in the case of smooth interface. Characteristic length scales are defined by using the interface statistics. The existence of the equivalent sphere system is shown for the non-spherical irregular droplet. The irregularity of simple droplets is characterized by an inequality related to the topological invariant. It is shown that statistically self-complementary, smooth system is mutually percolating. As a statistical foundation of the random interface problem, the relations to the theory of excursion set of random field are discussed. The expression of area density of the boundary set is obtained for the Gaussian field in Euclidean space with arbitrary dimension d. The cross-over phenomena in the spinodal decomposition process is also discussed using the statistical analyses on the interface.

§ 1. Introduction

It has been shown that at the late stage of phase separation process, e.g., in the spinodal decomposition, the semi-macroscopic interface picture is useful, since the essential part of non-linearity is incorporated in the theory efficiently by assuming sharp interfaces.[1)~3)] This is the reason that the phenomenological droplet theory by Lifshitz and Slyozov[4)] is successful in explaining the problem with small volume fraction. Here the meaning of semi-macroscopic interface is that all characteristic length scales of it, e.g., the mean radius of curvature, are sufficiently larger than the thickness ξ of the interface which is of the same order as the correlation length of the order parameter fluctuations, but are not macroscopic yet. That is, we have a finite area density of interface defined by

$$A=\int da/V\,, \tag{1}$$

where da is the area element and V the volume of the system. Note that A has a dimension of inverse of length, and is a quantity proportional to the non-equilibrium, excess free energy density in our system. In this stage interfaces are randomly distributed in the system. Here 'random' is used in very loose meaning such as homogeneous, isotropic distributions. For example, it includes a randomly distributed sphere system.

The purpose of the present paper is to find some statistical properties of this random interface system apart from the dynamical natures of phase separation. Then the results are applicable to other problems of pattern formation. We restrict the problem only in the smooth interface case, i.e., assume that singularities such as corner, edge, intersection and contact do not exist in the system, or are negligible. The fractal surface[5)] is also excluded. That is, finite curvatures are uniquely defined at every point on the interface. This restriction does not narrow our problems. In the problem of phase separation, these singularities can be expected to disappear immediately by evaporation and coalescence.

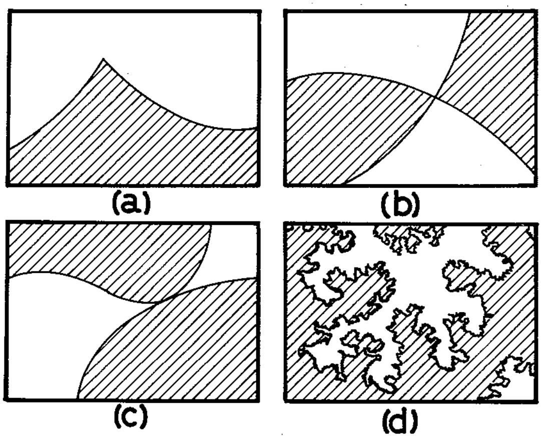

Fig. 1. Typical singularities of non-smooth interface. (a) corner (or edge), (b) intersection, (c) contact and (d) fractal surfaces.

The fractal surface with an infinite area density cannot be realized because the excess free energy should be finite in our system.

In the smooth system we find singularities only in the sharp profile of interface. Let us first investigate what kind of singularity and symmetry we may find in the correlation function of such system in § 2. Characteristic length scales are introduced and related to the statistical quantities of the interface in § 3. Three typical systems are discussed in § 4. A simple proposition on the percolation problem in the interface system, which has been revealed little compared with that in the lattice system, is given also in § 4. We need statistical foundations of the random interface to make more rigorous analyses on it. As an example the relation to the theory of excursion set of random field[6] is discussed in § 5. In § 6, the cross-over phenomena[7)~9)] in the spinodal decomposition is discussed.

§ 2. Singularity and symmetry of the correlation function

Let the order parameter field $\rho(\boldsymbol{r})$ be

$$\rho(\boldsymbol{r})=\begin{cases}1 & \text{in minority phase,}\\ 0 & \text{in majority phase,}\end{cases} \tag{2}$$

and define the volume fraction by

$$\phi=\frac{1}{V}\int\rho(\boldsymbol{r})\,d\boldsymbol{r}\equiv\langle\rho\rangle\,, \tag{3}$$

where V is the volume of the system and $d\boldsymbol{r}$ the d-dimensional volume element. A statistically important quantity is the correlation function of this field defined by

$$\begin{aligned} g(r)&=\frac{1}{V}\int d\boldsymbol{r}_1\rho(\boldsymbol{r}_1)\rho(\boldsymbol{r}_1+\boldsymbol{r})-\langle\rho\rangle^2\,,\\ &=\langle\rho(0)\rho(\boldsymbol{r})\rangle-\langle\rho\rangle^2\,, \end{aligned} \tag{4}$$

where homogeneous distribution of the interface is assumed. Here let us assume isotropy condition $g(\boldsymbol{r})=g(r)$ besides it.

For the purpose of getting an explicit form of $g(r)$ with the use of the interface statistics, it is advantageous to use

$$\begin{aligned} g_1(r)&=\langle\nabla\rho(0)\cdot\nabla\rho(\boldsymbol{r})\rangle\,,\\ &=-\nabla^2 g(r)\,, \end{aligned} \tag{5}$$

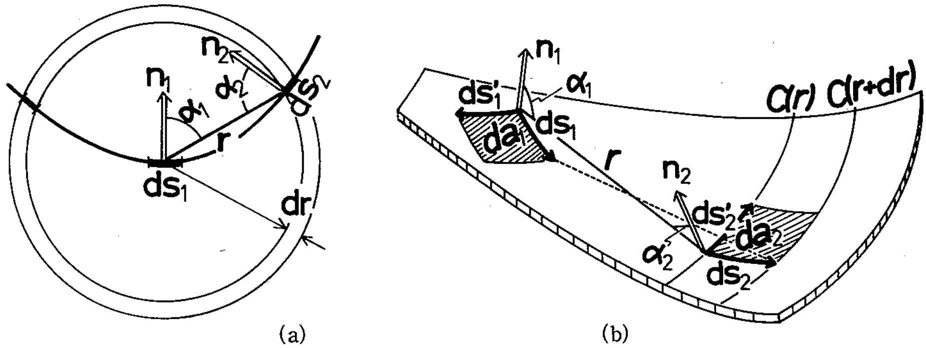

Fig. 2. Geometry of interface correlation function for (a) $d=2$ and (b) $d=3$.

instead of $g(r)$ itself, because $\nabla\rho$ has non-zero value only just on the interface in the present system: The new field variable $\nabla\rho(\boldsymbol{r})$ may be written as

$$\nabla\rho(\boldsymbol{r})=\boldsymbol{n}(\boldsymbol{r})\delta(u(\boldsymbol{r}))\,, \tag{6}$$

where $\boldsymbol{n}(\boldsymbol{r})$ is the normal unit vector at $\boldsymbol{r}$ on the interface and $u(\boldsymbol{r})$ a normal coordinate in this direction with conditions $u=0$ and $|\nabla u|=1$ at the interface.[10]

The explicit form of $g_1(r)$ for $d=2$ and 3 can be obtained by elementary geometry shown in Fig. 2: For $d=2$ one may find

$$2\pi r g_1(r)\,dr=\int\frac{ds_1}{V}\Sigma'\boldsymbol{n}_1\cdot\boldsymbol{n}_2 ds_2\,, \tag{7}$$

where V is the area of the system, ds_1 the integral line element of the interface, and $\boldsymbol{n}_1$, $\boldsymbol{n}_2$ the normal vectors at $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$ respectively. The summation Σ' is taken over all interface segments ds_2 between two circles with common center at $\boldsymbol{r}_1$ and radii r and $r+dr$. Here directional averaging is adopted instead of assuming isotropy. By using the angle α_2 between $\boldsymbol{r}$ $(=\boldsymbol{r}_2-\boldsymbol{r}_1)$ and $\boldsymbol{n}_2$ one may write the final form as

$$2\pi r g_1(r)=\int\frac{ds_1}{V}\Sigma'\frac{\boldsymbol{n}_1\cdot\boldsymbol{n}_2}{\sin\alpha_2}\,, \tag{8}$$

where the relation $|\boldsymbol{r}+\boldsymbol{ds}_2|=r+dr$, or $ds_2\sin\alpha_2=dr$ is used. Note that the right-hand side of Eq. (8) is *symmetric* with respect to suffixes 1 and 2. This can be shown by changing the integration element ds_1 to ds_2 with a condition $|\boldsymbol{r}+\boldsymbol{ds}_2-\boldsymbol{ds}_1|=r$, which results in the symmetric relation

$$ds_1\sin\alpha_1=ds_2\sin\alpha_2\,. \tag{9}$$

Analogously one may find for $d=3$

$$4\pi r^2 g_1(r)\,dr=\int\frac{da_1}{V}\int'\boldsymbol{n}_1\cdot\boldsymbol{n}_2\,da_2\,, \tag{10}$$

where da_1 and da_2 are area elements of the interface at $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$ and the integration $\int'$ is taken over the zone of the interface between two spheres with common center at $\boldsymbol{r}_1$ and radii r and $r+dr$. As is shown in Fig. 2 (b), let us take $\boldsymbol{ds}_2$ in the $(\boldsymbol{n}_2, \boldsymbol{r})$ plane, $\boldsymbol{ds}_2'$ in $\boldsymbol{n}_2\times\boldsymbol{r}$ direction so as to get $\boldsymbol{n}_2 da_2=\boldsymbol{ds}_2\times\boldsymbol{ds}_2'$. Then da_2 is given by $da_2=ds_2'\,dr/\sin\alpha_2$, where $ds_2\sin\alpha_2=dr$ is used. With the use of it the final form is written as

$$4\pi r^2 g_1(r)=\int\frac{da_1}{V}\oint_{C(r)}\frac{\boldsymbol{n}_1\cdot\boldsymbol{n}_2}{\sin\alpha_2}ds_2', \tag{11}$$

where $C(r)$ denotes the intersection curve between the interface and the sphere centered at $\boldsymbol{r}_1$ with radius r. Let us show the right-hand side of Eq. (11) is *anti-symmetric* with respect to suffixes 1 and 2: Take $d\boldsymbol{s}_1$ in the $(\boldsymbol{r}, \boldsymbol{n}_1)$ plane and $d\boldsymbol{s}_1'$ in $\boldsymbol{n}_1\times(-\boldsymbol{r})$ direction so as to get $\boldsymbol{n}_1 da_1=d\boldsymbol{s}_1'\times d\boldsymbol{s}_1$. One may find the same relation as Eq. (9) if one changes the integration variable from ds_1 to ds_2 with the condition $|\boldsymbol{r}+d\boldsymbol{s}_2-d\boldsymbol{s}_1|=r$. The anti-symmetric property is easily seen in the notation of exterior products of differentials[11] for the triple integral element as follows:

$$\frac{da_1 ds_2'}{\sin\alpha_2}=\frac{ds_1'\wedge ds_1\wedge ds_2'}{\sin\alpha_2}\underset{(1\leftrightarrow 2)}{\longrightarrow}\frac{ds_2'\wedge ds_2\wedge ds_1'}{\sin\alpha_1}$$

$$=\frac{ds_2'\wedge ds_1\wedge ds_1'}{\sin\alpha_2}=-\frac{ds_1'\wedge ds_1\wedge ds_2'}{\sin\alpha_2}.$$

Using the above considerations one may write

$$g_1(r)=\frac{(d-1)\gamma_d}{4}\frac{A}{r}G_1(r), \tag{12}$$

where $\gamma_d=4/\pi$ and 1 for $d=2$ and 3 respectively, and A is the interface area density defined in § 1. This is the typical form of $g_1(r)=-\nabla^2 g(r)$; $g_1(r)$ is proportional to the interface density A, and the singularity caused by the sharp profile of the interface results in the factor r^{-1}. The consequent quotient is the normalized, interface correlation function $G_1(r)$ which is a non-singular, even function of $\boldsymbol{r}$ and then of r. The expressions of $G_1(r)$ are given by

$$G_1(r)=\begin{cases}\dfrac{1}{A}\displaystyle\int\frac{ds_1}{V}\frac{1}{2}\sum{}'\frac{\boldsymbol{n}_1\cdot\boldsymbol{n}_2}{\sin\alpha_2}, & (d=2)\\[2ex] \dfrac{1}{A}\displaystyle\int\frac{da_1}{V}\oint_{C(r)}\frac{\boldsymbol{n}_1\cdot\boldsymbol{n}_2}{\sin\alpha_2}\frac{ds_2'}{2\pi r}, & (d=3)\end{cases} \tag{13}$$

where $G_1(0)=1$.

The original correlation function $g(r)$ can be given by integrating the equation $\nabla^2 g(r)=-g_1(r)$ and written in the same manner,

$$g(r)=\phi(1-\phi)-\frac{\gamma_d}{4}ArG(r), \tag{14}$$

where $G(r)$ is also an even function of r which satisfies $G(0)=1$ and $G'(0)=0$. The integration constant is determined by the condition

$$g(0)=\langle\rho^2\rangle-\langle\rho\rangle^2=\phi(1-\phi). \tag{15}$$

As a conclusion the singularity of the sharp profile of interface and the smoothness condition are simply expressed by an equation

$$\lim_{r\to 0}\left[\nabla^2 g(r)+\frac{(d-1)\gamma_d}{4}\frac{A}{r}\right]=0. \tag{16}$$

No other singularities in small r range can be expected so far as the smooth interface is assumed.

Define the structure function by

$$S(q)=\int d\boldsymbol{r}\, e^{i\boldsymbol{q}\cdot\boldsymbol{r}}g(r)\,, \tag{17}$$

which is normalized by the sum-rule

$$(2\pi)^{-d}\int d\boldsymbol{q}\; S(q)=\phi(1-\phi)\,. \tag{18}$$

Corresponding to the singularity in $g(r)$ there appears a kind of long tail, i.e., the Porod tail[12] in $S(q)$ given by

$$\lim_{q\to\infty} q^{d+1}S(q)=\beta_d A\,, \tag{19}$$

where $\beta_d=2$ and 2π for $d=2$ and 3. Whenever this limit exists, the smoothness condition (16) gives another sum-rule[13]

$$\int_0^\infty [q^{d+1}S(q)-\beta_d A]dq=0\,. \tag{20}$$

An exception is the mono-dispersive sphere system: As is shown in the next section, one finds an undumped oscillation in $q^{d+1}S(q)$ in this case, the right-hand side of Eq. (19) being merely the mean level of it.

§ 3. Characteristic length scales

The short range expansion of $g_1(r)$ and $g(r)$ has been obtained for arbitrary dimension d.[13] The results are summarized as

$$g_1(r)=\frac{(d-1)\gamma_d}{4}\frac{A}{r}\left\{1-\frac{d+1}{8}\frac{r^2}{R_m{}^2}+\cdots\right\}, \tag{21}$$

$$g(r)=\phi(1-\phi)-\frac{\gamma_d}{4}Ar\left\{1-\frac{d-1}{24}\frac{r^2}{R_m{}^2}+\cdots\right\},$$

where $\gamma_d=4\Gamma(d/2)/(d-1)\sqrt{\pi}\,\Gamma((d-1)/2)$ $(=4/\pi$ and 1 for $d=2, 3)$. R_m is related to a kind of mean curvature at each point on the interface as

$$R_m{}^{-2}=\int\bar{R}(a)^{-2}da\Big/\int da\,, \tag{22}$$

where $\bar{R}(a)^{-2}$ is the mean square of Euler's normal curvature averaged over the direction around the normal vector at each point. The explicit form of it with the use of the principal curvatures $\{R_i{}^{-1}\}$ is given by

$$\bar{R}(a)^{-2}=[2\sum_{i=1}^{d-1}R_i{}^{-2}+(\sum_{i=1}^{d-1}R_i{}^{-1})^2]/(d^2-1)\,. \tag{23}$$

Note that the lower order terms in the expansions should be determined only by the radius of curvature, which is the unique local scale in the smooth system. The first order term of it does not appear[12] because of the symmetry given in § 2. In other words, this is Babinet's reciprocity principle on the complementary system; when the values of the order parameter are reversed, i.e., from ρ to $1-\rho$, the correlation function $g(r)$ is not changed, while the sign of curvature is changed.

The asymptotic expansion of $S(q)$ is given by

$$S(q)=\frac{\beta_d A}{q^{d+1}}\left\{1+\frac{d^2-1}{8}\frac{1}{R_m{}^2 q^2}+\cdots\right\}, \tag{24}$$

where $\beta_d=2^{d-1}\pi^{(d-2)/2}\Gamma(d/2)$ $(=2, 2\pi$ for $d=2, 3)$.

Apparently R_m, a kind of mean radius of curvature, gives a characteristic length scale: Divided by $\sqrt{(d^2-1)/8}$, it may be used as a criterion for the Porod tail as is easily seen in Eq. (24).

One may define another scale which characterize the dominant part of $S(q)$ as follows: Define a wave-number where the sum-rule (18) is fulfilled upto some proportion of order unity, say upto 1/2. With the use of Eq. (24) the corresponding length scale is approximated by

$$D=\pi\phi(1-\phi)/2A\,. \tag{25}$$

The same definition has been used as a coherence range in the spin system.[14),15)] In the present system D gives a mean size of droplets (or particles) if the system is composed of separated droplets. Note that these two, i.e., R_m and D are assumed to be semi-macroscopic, i.e., sufficiently larger than the thickness of the interface ξ.

One more characteristic length in our system is the long-ranged correlation length L, the inverse of which characterizes the small q part of $S(q)$. It is impossible to get explicit form of L like those of R_m and D because L is non-local quantity, but L should be distinguished from D in general. For example, in a well-separated droplets system L is the droplet-droplet correlation length, while D is a mean size of droplets. On the contrary, both become undistinguishable and may be of the same order of magnitude in a ramified interface system. Note that the radius of gyration, i.e., the Guinier length defined by the small q expansion of $S(q)$ loses its original meaning if the interference between droplets or between parts of interface cannot be neglected.

§ 4. Typical systems

(A) Spherical droplet system

This is the typical case for $\phi\ll 1$. Using the indicator function defined by

$$\theta(R;\,\boldsymbol{r})=\begin{cases}1 & \text{for } |\boldsymbol{r}|\leqq R\,,\\ 0 & \text{for } |\boldsymbol{r}|>R\,,\end{cases} \tag{26}$$

the correlation function is written as

$$g(r)=\sum_R n(R)v(R)\gamma(R;\,r)-\sum_{R'}\sum_{R''}n(R')v(R')n(R'')v(R'')$$
$$\times\int d\boldsymbol{r}'\int d\boldsymbol{r}''\theta(R';\,\boldsymbol{r}')\theta(R'';\,\boldsymbol{r}'')[1-W^{(2)}(R', R'';\,\boldsymbol{r}+\boldsymbol{r}'-\boldsymbol{r}'')]\,, \tag{27}$$

where $v(R)$ and $n(R)$ are the volume and the number density of spheres with radius R, $\gamma(R;\,r)$ the single sphere correlation function defined by

$$\gamma(R;\, r)=\frac{1}{v(R)}\int d\boldsymbol{r}'\theta(R;\, \boldsymbol{r}')\theta(R;\, \boldsymbol{r}'+\boldsymbol{r})\,,$$

$$=\frac{d\gamma_d}{2}\int_{r/2R}^{1}(1-x^2)^{(d-1)/2}dx\,,\quad (r\leqq 2R) \tag{28}$$

with normalization $\gamma(R;0)=1$. $W^{(2)}(R', R''; \boldsymbol{r})$ denotes the pair distribution function of droplets with radii R' and R'' normalized as $W^{(2)}(\infty)=1$. Because of the coalescence effect $W^{(2)}$ can be expected to have an exclusive region in some extent around the absorptive core of radius $R'+R''$. Then the interference terms do not contribute the short range expansion of $g(r)$, and it is straightforward to get the expansion, if $n(R)$ is not a singular function. For $d=3$ we find

$$\phi=(4\pi/3)n\langle R^3\rangle\,,\quad A=4\pi n\langle R^2\rangle\,,\quad R_m{}^2=\langle R^2\rangle\,, \tag{29}$$

where

$$\langle R^k\rangle=\frac{1}{n}\sum_R R^k n(R)\,,\quad n=\sum_R n(R)$$

are used. Using $\phi\ll 1$, the characteristic scale D is given by

$$D=(\pi/6)\langle R^3\rangle/\langle R^2\rangle\,, \tag{30}$$

which is the Porod radius except for a numerical factor. Thus R_m and D are independent of ϕ and can be expected to be of the same order if $n(R)$ is an ordinary function, though we have an inequality $\langle R^3\rangle^2\geqq\langle R^2\rangle^3$. On the other hand, the last scale L depends on ϕ and $L\gg D$, R_m: An intuitive definition of L is the mean distance between spheres,[16] i.e.,

$$L\sim n^{-1/3}=(3\phi/4\pi\langle R^3\rangle)^{-1/3}\,. \tag{31}$$

Other results $L\sim\phi^{-1/2}$ have been obtained by introducing the screening effect in the theory.[3),17)]

The corresponding structure function $S(q)$ is given by using

$$\Psi(Q)=\frac{1}{v(1)}\int d\boldsymbol{r}\; e^{i\boldsymbol{Q}\cdot\boldsymbol{r}}\theta(1;\,\boldsymbol{r})$$

$$=\frac{\Gamma(d/2+1)}{(Q/2)^{d/2}}J_{d/2}(Q)\,, \tag{32}$$

where $J_\nu(x)$ is the Bessel function. Using the asymptotic form of it, the most dominant part of $S(q)$ for large q is given by

$$S(q)\sim\frac{2^d\pi^{d-1}}{q^{d+1}}\sum_R n(R)R^{d-1}\left\{1+\cos\left(2qR-\frac{d+1}{2}\pi\right)\right\}. \tag{33}$$

This results in the Porod law if the oscillating terms are smeared out by continuous spectrum $n(R)$. A simple exception is the mono-dispersive sphere system.[16]

(B) Irregular droplet system

As is shown in Appendix A, the spherical droplet picture becomes improper, at least above some critical fraction ϕ_c. Then the next type to be considered is the separated, non-spherical droplet system. Evidently, in this system the mean size D and the mean

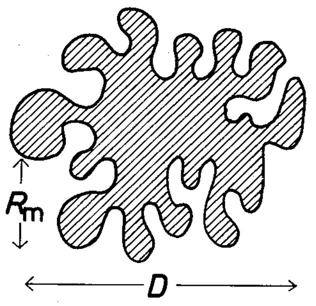

Fig. 3. Complicated droplet with $D \gg R_m$.

radius of curvature R_m are independent notions, though $L \gg D$, R_m is assumed here. The case $D \gg R_m$ can be expected for the droplet with complicated surface as is illustrated in Fig. 3.

The correlation function can be written in the same form as Eq. (27), if R is interpreted as the suffix to distinguish different shapes of droplets. In Appendix B it is shown that the normalized, single droplet correlation function $\gamma(r)$ averaged over all orientations is represented by an equivalent sphere system, i.e.,

$$\gamma(r) = \frac{1}{v}\sum_R n(R)v(R)\gamma(R;\, r)\,, \tag{34}^{*)}$$

where v is the volume of a droplet concerned. Equation (34) is expanded as

$$\gamma(r) = 1 - \frac{\gamma_d}{4}\frac{S}{v}r\left\{1 - \frac{d-1}{24}\frac{r^2}{R_m{}^2} + \cdots\right\}. \tag{35}$$

Here S is the surface area of the single droplet and $R_m{}^{-2}$ the same as Eq. (22) averaged over single surface S. These are given by $n(R)$ in the same manner as Eq. (29). It is interesting to note that the total number $n = \sum n(R)$ for $d=3$ is rewritten as

$$n = S/4\pi R_m{}^2 = \oint_S da/4\pi\bar{R}(a)^2\,,$$

where

$$\bar{R}(a)^{-2} = \frac{1}{R_1 R_2} + \frac{3}{8}\left(\frac{1}{R_1} - \frac{1}{R_2}\right)^2. \tag{36}$$

Then we find $n \geqq 1$, at least if S is an oriented, closed surface of simply connected droplet (not a simply connected surface), because we have a topological invariant

$$\oint_S da/R_1 R_2 = 4\pi \tag{37}$$

for such surface by Gauss and Bonnet's theorem. Here $n=1$ is satisfied only by spheres. It can be shown that n is divergent in the case of non-smooth surface like that of convex lens, though $R_1 = R_2$ everywhere is satisfied. The corresponding inequality for $d=2$ is written as

$$R_m{}^{-2} \geqq (2\pi/L_C)^2 \tag{38}$$

for an arbitrary simple closed curve C, i.e., the Jordan curve on $d=2$ plane, where L_C is the length of it and the invariant related to the curvature R^{-1}

$$\oint_C ds/R = 2\pi \tag{39}$$

is used. These inequalities can be used to characterize the irregularity of simple droplets.

*) The same expansion for the correlation function $g(r)$ of the total system is possible, but the meaning of $n(R)$ becomes rather ambiguous.

Such a universal inequality, however, has not been found for general surfaces (curves) with more complicated topology.

It can be said that the irregularity assures the rapid convergence to the Porod tail of $S(q)$, because the irregularity averaged over various types of droplets can be equivalent to a continuously poly-dispersive spectrum. However, this equivalence itself prevents us from finding a distinction between the irregular droplet system and poly-dispersive sphere system by any scattering experiment.

(C) Percolating system

At larger $\phi(\gtrsim\phi_c)$ droplets coalesce one another and become connected from infinity to infinity when $\phi\cong 1/2$. Here let such a state be called percolating. Evidently, it is impossible to distinguish the correlation length L and the mean size scale D in the system like this, while the mean radius of curvature R_m keeps its own meaning. Note that $\phi\cong 1/2$ is not necessary. Usually percolation is expected to occur at the smaller value, say at $\phi\sim 1/2d$, by analogy with that in the lattice system.[8] On the contrary, it is possible to fulfil the system up to the random packing limit[18] with separated, mono-dispersive spheres, and in principle, up to 99.99% with poly-dispersive spheres, at least if spheres are repulsive. Thus the percolation problem in the interface system has not been clarified yet.

An interesting system which seems tractable is one consisting of statistically equivalent binary phases, e.g., the 50%~50% symmetric alloy, or the Ising system without external field. Such system may be called a statistically self-complementary system. That is, the complementary system given by inverting the order parameter from ρ to $1-\rho$ is statistically equivalent to itself. For example, curvatures with opposite signs should be distributed with symmetric weight. Let us close this section by making the following propositions related to this system:

(I) At least one of the two phases, say the background phase, is percolating, whenever the interface is smooth everywhere.

(II) Then, the (statistically) self-complementary system with smooth interfaces is mutually percolating, i.e., both phases are percolating.

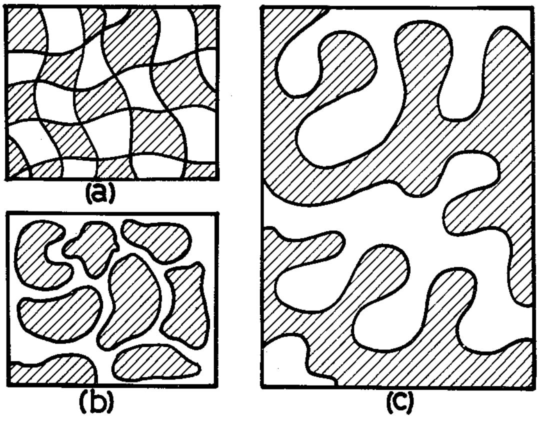

Fig. 4. (a) Self-complementary but not smooth, (b) smooth but not self-complementary, (c) smooth and self-complementary.

Here we need only the absence of intersection and contact among the smoothness conditions in § 1. In such system in $d=2$ the interface lines do not cross each other and are all Jordan's closed curves or open curves extended from infinity to infinity. If we have at least one open curve, both phases are percolating along it. Evidently, if all are Jordan's closed curves separated from each other, the exterior region of them, say the background phase is not disconnected, i.e., is percolating. Here, even if there is a closed curve which encloses other closed curve(s) in it, (e.g., hole(s), island(s) and so on), this statement is satisfied by taking into consideration only the outmost curve. In this context we should restrict the system within the bounded systems to avoid an infinite nesting of such structures. Note that the conditions of smoothness and self-complementarity are both important in the proposition as is shown in Fig. 4. For $d=3$ we may use the notion of oriented closed surface in the above statements instead of Jordan's curve. Now it is straightforward by definition to show (II).

It should be noted that (II) is merely a sufficient condition for percolation. It is always possible to transform a percolating, self-complementary system continuously to asymmetric one with $\phi<1/2$ (or $\phi>1/2$) conserving its topology of percolation.

§ 5. Statistical model for random interface

Let us discuss the statistical foundations of the random interface problem. We may use the theory of excursion set of random field[6] for this purpose. Let $X(\boldsymbol{r})$ be a scalar random field in d-dimensional Euclidean space, and define the excursion set by

$$V(u)=\{\boldsymbol{r}|X(\boldsymbol{r})\geqq u\}. \tag{40}$$

Then the boundary set, i.e., the level crossings defined by

$$\partial V(u)=\{\boldsymbol{r}|X(\boldsymbol{r})=u\}, \tag{41}$$

can be used as a statistically tractable model for random interface, if the statistics of $X(\boldsymbol{r})$ is defined. An important example is the Gaussian field, because it seems to include a wide variety of linearized or quasi-linearized theories[19)~21)] applied to the order parameter field problems. The definition of excursion set (40) with $u=0$ just corresponds to the non-linear transformation[3),19)]

$$s(\boldsymbol{r})=\mathrm{sign}(X(\boldsymbol{r})), \tag{42}$$

where $s=1-2\rho$ is used here.

Let $X(\boldsymbol{r})$ be a Gaussian random field with zero-mean, unit-variance and covariance $\{\sigma_{ij}=\langle X(\boldsymbol{r}_i)X(\boldsymbol{r}_j)\rangle\}$, where $\langle\cdots\rangle$ denotes the statistical expectation here, and define the indicator function for crossing level u by

$$s(\boldsymbol{r})=\mathrm{sign}(X(\boldsymbol{r})-u). \tag{43}$$

Note that this is completely equivalent to the problem of zero-level crossings in a Gaussian field with mean $-u$. The expectations $\langle s(\boldsymbol{r})\rangle$ and $\langle s(\boldsymbol{r}_1)s(\boldsymbol{r}_2)\rangle$ are given by

$$\langle s(\boldsymbol{r})\rangle=-\sqrt{\frac{2}{\pi}}\int_0^u e^{-t^2/2}dt \tag{44}$$

and

$$\langle s(\boldsymbol{r}_1)s(\boldsymbol{r}_2)\rangle=1-\frac{2}{\pi}\int_0^{\mathrm{Cos}^{-1}\sigma_{12}}\exp\left\{-\frac{u^2}{1+\cos\theta}\right\}d\theta, \tag{45}$$

where

$$\mathrm{sign}(x)=\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty}\frac{\lambda}{\lambda^2+\epsilon^2}e^{i\lambda x}\frac{d\lambda}{i\pi},$$

is used. The simple result for $u=0$ in Eq. (45),[22] i.e.,

$$\langle s(\boldsymbol{r}_1)s(\boldsymbol{r}_2)\rangle=(2/\pi)\text{Arcsin}\ \sigma_{12}\,, \tag{46}$$

has been used in the present problem by Kawasaki et al.[19] and by Ohta.[3]

For the homogeneous, isotropic system with

$$\sigma_{ij}=\sigma(|\boldsymbol{r}_i-\boldsymbol{r}_j|) \tag{47}$$

one may interpret the expectation $\langle\cdots\rangle$ as the spatial average. In this case the mean volume fraction of the excursion set $V(u)$ is given by

$$\phi=\frac{1}{\sqrt{2\pi}}\int_u^\infty e^{-t^2/2}dt\,. \tag{48}$$

The correlation function $g(r)$ defined by Eq.(4) is given by

$$g(r)=\phi(1-\phi)-\frac{1}{4}[\langle s(0)s(\boldsymbol{r})\rangle-1]\,. \tag{49}$$

Then expanding Eq. (45) with respect to r we find

$$A=\left[(d-1)\Gamma\left(\frac{d-1}{2}\right)/\,2\sqrt{\pi}\,\Gamma\left(\frac{d}{2}\right)\right]e^{-u^2/2}\sqrt{|\sigma''(0)|} \tag{50}$$

and

$$R_m{}^{-2}=\{\sigma^{(4)}(0)-(1-u^2)\sigma''(0)^2\}/\,(d-1)|\sigma''(0)|\,. \tag{51}$$

Here the covariance $\sigma(r)$ is assumed to be even function of r, which is the condition for the smooth interface in this problem. The same result for A with $u=0$ is obtained by Ohta.[3]

Global geometry of this system, however, has not been revealed except for a few properties, e.g., the boundary surfaces become convex everywhere with probability unity when $u\to\infty$ (or $\phi\to 0$).[6] That is, the oval droplet picture is proper in the system with small volume fraction.

§ 6. Discussion

It has been revealed theoretically[23),24] and experimentally[25] that a kind of scaling law exists at the late stage of spinodal decompositions. That is, the characteristic length scale $l(t)$ obeys a power law $l(t)\sim t^a$ and the structure function is well approximated by a scaled form as $S(q)=l(t)^d\,\mathcal{S}\,(l(t)q)$. The exponent a and the scaling function $\mathcal{S}\,(Q)$ are, however, not always universal, e.g., a cross-over phenomena is observed.[9]

Here let us discuss it: At small ϕ, the spherical droplet picture is proper and the relevant length scale is $D(\sim R_m)$. The dominant growth mechanism of droplets is Lifshitz and Slyozov's evaporation-condensation process, where $D\sim t^{1/3}$ is known.[4] The Brownian motion of droplet can be neglected in the solid mixture, since the diffusion constant is very small in this case. At larger ϕ where $L\sim D$, however, the Brownian coalescence is no more negligible. The earlier stage of droplet growth may be dominated by it, where $D\sim t^{1/6}$ is expected.[26),27] In this stage $D\sim R_m$ still holds and then $\mathcal{S}\,(Q)\sim Q^{-4}$. Next, percolation occurs, perhaps in a little while. After percolation the dominant growth process is smoothening of the complicated surface by evaporation-condensation process, where $R_m(t)\sim t^{1/3}$ is proposed by Kawasaki and Ohta.[1] In this stage it may be supposed that $D\gg R_m$, i.e., the Porod tail region goes far away from the dominant region, and then $\mathcal{S}(Q)$ becomes dominated by the next power Q^{-6}.

In this paper only a simple, sufficient condition is proposed with respect to the percolation problem in the semi-macroscopic random interface system. The significant property supposed in the proposition in § 4 is the symmetric distribution of curvature in the system. Though it is not the necessary condition, it is very suggestive for the future investigations on the statistical mechanics of random interface.

Acknowledgements

It is a great pleasure for the author to dedicate this paper to Professor K. Tomita on the occasion of his retirement from Kyoto University. He would like to thank Professor K. Tomita for his valuable discussions and continual encouragement during the past twenty years.

Appendix A
—— *Critical Volume Fraction for Absorptive Sphere System* ——

In the coalescing sphere system the sphere density should be decreased in some extent around the absorptive core, i.e., we may assume that

$$W^{(2)}(R, R'; r\leqq R+R')=0\,,$$

$$W^{(2)}(R, R'; r)\leqq W^{(2)}(R, R'; \infty)=1\,,$$

and then

$$\int d\boldsymbol{r}[1-W^{(2)}(R, R'; r)]\geqq v(R+R')\,. \tag{A·1}$$

This cannot be necessarily satisfied if we have a region where $W^{(2)}(r)>1$, as is usually seen in the equilibrium state of the hard core system. Compared to it, our system may be called absorptive sphere system, though the equilibrium state cannot exist. Here Eq. (A·1) is expected to be valid in the non-equilibrium state starting from an uncorrelated, uniform initial distribution.

On the other hand, the structure function should be positive definite for all q by definition, and at least at $q=0$ we have

$$\sum_R n(R)v(R)^2-\sum_{R,}\sum_{R'} n(R)v(R)n(R')v(R')\int[1-W^{(2)}(R, R'; r)]d\boldsymbol{r}\geqq 0\,. \tag{A·2}$$

Evidently, it is impossible to find a solution for $W^{(2)}(r)$ at least if

$$\sum_R n(R)v(R)^2-\sum_{R,}\sum_{R'} n(R)v(R)n(R')v(R')v(R+R')\geqq 0\,, \tag{A·3}$$

is not satisfied. This leads to a necessary condition $\phi\leqq\phi_c$ for the absorptive sphere model to be applicable, where

$$\phi_c=\begin{cases}1/2[1+\langle R^3\rangle^2/\langle R^4\rangle\langle R^2\rangle], & (d=2)\\ 1/2[1+3\langle R^5\rangle\langle R^4\rangle/\langle R^6\rangle\langle R^3\rangle]. & (d=3)\end{cases}$$

It can be shown that

$$2^{-d}\leqq\phi_c\leqq 1/2, \tag{A·4}$$

where the lower bound corresponds to the mono-dispersive system.

Appendix B

——*An Equivalent Sphere System for Irregular Droplets*——

Assume that the normalized, single droplet correlation function $\gamma(r)$ is written as

$$\gamma(r)=\frac{1}{v}\int_0^\infty dR\ n(R)v(R)\gamma(R;r), \tag{B·1}$$

where $n(R)$ is normalized by

$$\int_0^\infty dR\ n(R)v(R)=v.$$

Then let us show that it is always possible to find an explicit representation for $n(R)$ when $\gamma(r)$ is given. This can be attained by the Mellin transformation of Eq. (B·1) as follows: Using the definition of $\gamma(R;r)$, i.e., Eq.(28) in the text, one finds

$$\frac{1}{v}\int_0^\infty R\ n(R)v(R)R^{s-1}dR=\frac{s}{2^s}\frac{B\left(\frac{1}{2},\frac{d+1}{2}\right)}{B\left(\frac{s+1}{2},\frac{d+1}{2}\right)}\int_0^\infty\gamma(r)r^{s-1}dr, \tag{B·2}$$

where the Beta function representations

$$\int_0^1 x^s(1-x^2)^{(d-1)/2}dx=\frac{1}{2}B\left(\frac{s+1}{2},\frac{d+1}{2}\right)$$

and

$$\gamma_d=4/dB\left(\frac{1}{2},\frac{d+1}{2}\right),$$

are used. The coefficient on the right-hand side of Eq. (B·2) for $d=2, 3$ is rewritten as

$$\frac{sB\left(\frac{1}{2},\frac{d+1}{2}\right)}{B\left(\frac{s+1}{2},\frac{d+1}{2}\right)}=\begin{cases}\frac{1}{4}s^2(s+2)B\left(\frac{1}{2},\frac{s}{2}\right), & (d=2)\\ \frac{1}{3}s(s+1)(s+2), & (d=3)\end{cases} \tag{B·3}$$

respectively. By using the inverse transformation the final expressions are given by

$$n(R)\ v(R)/v=\begin{cases}-2R\int_{2R}^\infty\frac{1}{\sqrt{1-4R^2/x^2}}\frac{d^3\gamma(x)}{dx^3}dx, & (d=2)\\ -\frac{16}{3}R^3\left[\frac{d}{dx}\frac{1}{x}\frac{d^2\gamma(x)}{dx^2}\right]_{x=2R}. & (d=3)\end{cases}$$

Note that the result for $d=3$ can be directly obtained from Eq. (B·1) with the use of

$$\gamma(R;r)=\begin{cases} 1-\dfrac{3r}{4R}+\dfrac{r^3}{16R^3}, & r\leqq 2R, \\ 0, & r>2R. \end{cases}$$

As is easily seen in this result, $n(R)$ is singular at $R=0$ when the surface is not smooth, i.e., $\gamma''(0)\neq 0$.

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Note added in proof: Recently, Toyoki and Honda obtained the same formula as Eq. (50) for $u\neq 0$.[28] They also found the correlation function Eq. (45) in an expansion series.

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