The Role of Density Gradient Energy in Alloy Phase Segregation*

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Diffusion behavior of composition variation peaks in supersaturated solid solutions has been discussed using Fourier transform of Cahn-Hilliard's diffusion equation. For the cases where the peak of positive compositional deviation at the location \(x=0\) is a maximum of composition variation, it is shown that the condition for a rise of the peak is given by \(\psi - D_x=0 > 0\). Here, \(D_x=0\) is the interdiffusion coefficient of the peak composition and \(\psi = -2K \beta^2 \frac{\Sigma q_n \cdot \Sigma \alpha_n}{\Sigma q_n \cdot \alpha_n} h^2 Q(h)\). \(h\beta\) and \(Q(h)\) are the wavenumber of a Fourier wave and its component, respectively, and \(K\) is (gradient energy coefficient) \(\times\) (mobility of atoms). In high solute alloys, the peak rises even in the range where \(-D_x=0 < 0\) by formation of the Fourier spectra for which \(\psi > 0\), and \(\psi\) becomes equal to \(D_x=0\) when the composition peak reaches the equilibrium composition. In very low solute alloys the peak sinks even in the range where \(-D_x=0 > 0\) by generation of the spectra for which \(\psi < 0\).

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I. Introduction

Cahn and Hilliard built up the concept of composition or density gradient energy which is abbreviated to gradient energy in the present paper, and applied this concept to estimate composition profiles of critical nuclei and of equilibrium states\(^{(1)(2)}\). In the spinodal decomposition theory formulated by Cahn, the gradient energy has been utilized as a factor by which the amplification of high frequency Fourier waves is inhibited\(^{(3)}\). Recently, Hilliard and Tsakalakos pointed out that the motive force for composition variations to amplify beyond the spinodal line is the gradient energy\(^{(4)}\).

In the present paper the diffusion behavior of composition variations in supersaturated solid solutions has been analyzed. The results have confirmed that the idea of Hilliard and Tsakalakos holds not only for spinodal decomposition but also for nucleation-growth, and also that although the gradient energy contributes to the evolution of composition variations in high solute alloys, it serves to shrink the composition variations in very low solute alloys.

II. Theory

1. Conditions for a rise of composition peak and diffusion equation

When the gradient energy is taken into consideration, the diffusion equation is given by

\[
\frac{\partial c}{\partial t} = \frac{\partial}{\partial x} \left[ D \frac{\partial c}{\partial x} \right] - 2K \left( \frac{\partial^4 c}{\partial x^4} \right). \tag{1}
\]

The interdiffusion coefficient, \(D\), is negative for compositions inside the spinodal and positive for compositions outside the spinodal. When \(M\) represents the mobility of atoms, \(K/M\) is the gradient energy coefficient. When we express a composition deviation from an average composition \(c_0\) at the location \(x\) and time \(t\) as \(q(x,t) = c(x,t) - c_0\), eq. (1) is written as

\[
\frac{\partial q}{\partial t} = \frac{\partial}{\partial x} \left[ D \frac{\partial q}{\partial x} \right] - 2K \left( \frac{\partial^4 q}{\partial x^4} \right). \tag{2}
\]

In this paper only the cases where the maximum of composition variation is located at \(x=0\) are treated. The composition peak such as

\[
\begin{align*}
q_{x=0} &> 0 \\
(\frac{\partial q}{\partial x})_{x=0} &> 0 \\
(\frac{\partial^2 q}{\partial x^2})_{x=0} &< 0,
\end{align*}
\]

is called the composition peak for brevity and its diffusion behavior is discussed. Similar discussion may be applied for the peak such as

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and the obtained results are essentially the same.

We define $\xi$ and $\psi$ as

$$\xi = -(\partial^2 q/\partial x^2)_{x=0} > 0,$$  \hspace{1cm} (4)

and

$$\psi = 2K(\partial^4 q/\partial x^4)_{x=0} / (\partial^2 q/\partial x^2)_{x=0}.$$  \hspace{1cm} (5)

From eq. (2) the behavior of the composition peak is given by eqs. (6) and (7).

$$(\partial q/\partial t)_{x=0} = \bar{D}_{x=0} (\partial^2 q/\partial x^2)_{x=0}$$

$$- 2K(\partial^4 q/\partial x^4)_{x=0} = \xi \cdot (\psi - \bar{D}_{x=0}),$$

where $\bar{D}_{x=0}$ is the interdiffusion coefficient for the composition at $x=0$, $c_{x=0}$.

Neglecting the second term of the right-hand side in eq. (6), we may consider that the composition peak rises when

$$(\partial q/\partial t)_{x=0} = \bar{D}_{x=0} (\partial^2 q/\partial x^2)_{x=0} > 0.$$  \hspace{1cm} (6)

Evidently, the condition for the composition peak to rise is given by

$$- \bar{D}_{x=0} > 0,$$

and the condition to stop rising is given by

$$- \bar{D}_{x=0} = 0.$$  \hspace{1cm} (7)

This means that the composition peak rises when its composition is inside the spinodal, and it stops rising when its composition attains the spinodal line. We denote the spinodal compositions of low and high solute contents as $c_{s1}$ and $c_{s2}$, respectively.

The diffusion equation in which the gradient energy term is not included possesses three fatal defects. One is that the composition peak does not rise beyond $c_{s2}$ even on spinodal decomposition. This means that decomposition is not completed only by the diffusion process. More generally, the second defect is that precipitates of the equilibrium composition are always resolved by diffusion, until their composition reaches $c_{s2}$. The last one is that the composition peak within the spinodal region always rises up to $c_{s2}$, even if the solid solution is not supersaturated.

Hilliard and Tsakalakos pointed out the first defect of the conventional diffusion equation. As the mechanism that on spinodal decomposition composition variations amplify until the equilibrium compositions, they suggested that the second term of eq. (6) becomes positive in the region where the first term is negative. This important proposition means that not only the gradient energy inhibits development of high frequency Fourier waves, but also it plays an important role in respect to the propriety of the attainment of the equilibrium state.

When the gradient energy is taken into account, the condition for a rise of the composition peak is given by

$$\psi - \bar{D}_{x=0} > 0,$$  \hspace{1cm} (8)

and the condition for the composition peak to stop rising is given by

$$\psi - \bar{D}_{x=0} = 0.$$  \hspace{1cm} (9)

In order that the composition peak may rise in the region where $c_{x=0} > c_{s2}$, it is required at least that $\psi > 0$, i.e., $(\partial^4 q/\partial x^4)_{x=0} < 0$. Equation (8) also implies that the composition peak is able to sink for a large value of $(\partial^4 q/\partial x^4)_{x=0}$, even if $c_{x=0}$ is inside the spinodal.

2. Conditions for a rise of composition peak and Fourier spectra

We expand $q(x, t)$ into the Fourier series as

$$q(x, t) = \sum_{h \neq 0} Q(h) \exp(ih\beta x).$$  \hspace{1cm} (10)

$h\beta$ and $Q(h)$ are the wavenumber of a Fourier wave and its component, respectively. $h$ is an integer excluding a zero and $\beta = 2\pi/L$. $L$ is the size of a region where eq. (1) is applied. We may regard $Q(h)$ as representing a Fourier spectrum corresponding to a composition variation.

From eq. (10) the composition deviation at the peak, $q_{x=0}$, is given by

$$q_{x=0} = \sum_{h} Q(h).$$  \hspace{1cm} (11)

Putting eq. (10) into eqs. (4) and (5), we obtain

$$\xi = \sum_{h} h^2 Q(h),$$  \hspace{1cm} (12)
and
\[ \psi = -2K\beta^2 \left[ \sum_h h^4 Q(h) \right] \] (13)

As \( \xi > 0 \), \( \psi \) is positive when
\[ \sum_h h^4 Q(h) < 0. \] (14)

In effect, the formation of Fourier spectra satisfying eq. (14) is required at least for the composition peak to rise in the region where \( D_x = 0 > 0 \).

It was shown in a previous paper(5) that the interdiffusion coefficients, \( \bar{D}(x, t) \), in each part of a composition variation are written as
\[ D(x, t) = D_0 - D_1 \cdot q(x, t) + D_2 \cdot q^2(x, t), \] (15)
in the alloy system whose free energy, \( f \), is given by
\[ f = \gamma \left[ (0.5 - c_e)^2 - (0.5 - c)^2 \right]^2. \] (16)
Here,
\[ \begin{align*}
D_0 &= 12\gamma M \left[ (0.5 - c_0)^2 - (0.5 - c_e)^2 \right] \\
D_1 &= 24\gamma M (0.5 - c_0) \\
D_2 &= 12\gamma M.
\end{align*} \] (17)
\( c_e \) is the equilibrium composition and \( \gamma \) is a positive constant.

It was also shown that a change of composition variation with diffusion is expressed as a change of the Fourier spectrum. The derived equation which is the Fourier transform of eq. (2) is written as
\[ \frac{\partial Q(h)}{\partial t} = -K\beta^2 \left[ (D_0 + 2h^2K\beta^2)Q(h) - D_1 \cdot R(h)/2 + D_2 \cdot S(h)/3 \right]. \] (18)
Here, \( R(h) \) and \( S(h) \) are the functions obtained by convoluting \( Q(h) \) once and twice, respectively. \( \sum_h \left[ \partial Q(h)/\partial t \right] \) coincides with eq. (6), that is, eq. (18) represents resolution of eq. (6) into the behavior of an individual Fourier wave.

The behavior of \( Q(h) \) is controlled by the first term of eq. (18) when the amplitude of composition variations is small. With an increase in amplitude of composition variations the controlling term changes from the first term to the second term, and then to the third term(6). The transfer of the controlling term may be considered to occur with an increase of \( q_x = 0 \) when the maximum of composition varia-

tions is located at \( x = 0 \). The function of the first term is to increase or decrease the absolute value of a Fourier component which already exists. The second and the third term represent the interaction of Fourier waves and yield Fourier waves of new wavenumber. The Fourier spectra yielded by the second term are entirely different from those by the third term. The second term yields spectra with components of the same sign, while the third term yields spectra with components of periodically alternating signs. We denote the former spectra by \( Q_d(h) \) and the latter spectra by \( Q_a(h) \).

In the cases treated in this paper, \( Q(h) > 0 \) in \( Q_d(h) \) and \( Q(h) > 0 \) for the lowest value of \( |h| \) in \( Q_a(h) \).

It is obvious that eq. (14) is impossible to be realized in \( Q_d(h) \). In \( Q_a(h) \) there is the possibility that eq. (14) is fulfilled and, in fact, this condition is satisfied by developed \( Q_a(h) \) as will be described in III: Generally speaking, we may conceive that \( \psi \) decreases by the development of \( Q_d(h) \) and increases by the development of \( Q_a(h) \).

3. Alloy composition and behavior of the composition peak

Assuming \( c_e = 0.50 \pm 0.45 \) in eq. (16), we have \( c_s = 0.50 \pm 0.26 \) and the free energy vs composition curve shown in Fig. 1. \( D_0/D_2 \) and \( D_1/D_2 \) for this alloy system are shown in Fig. 2. \( D_0 = \bar{D}_e = c_0 \) and \( D_2 \) is independent of \( c_0 \). \( D_0 + 2h^2K\beta^2 \) prescribes the work of the first term in the right-hand side of eq. (18). \( D_1/D_2 \)

![Fig. 1 Free energy vs composition curve represented by eq. (16). Assumed \( c_e = 0.50 \pm 0.45 \).](image-url)
prescribes the ratio of the activity between the second term and the third term. A large value of $D_1/D_2$ implies that $Q_a(h)$ develops conspicuously and the spectrum change from $Q_d(h)$ to $Q_a(h)$ is delayed. Thus, when compared to an equal value of $q_x=0$, $\phi$ in low solute alloys is low.

When the composition variation amplifies until the third term of eq. (18) works thoroughly, the decomposition is completed and the equilibrium state is realized. Decomposition processes differ according to alloy composition. In high solute alloys for which $D_0<0$ and $D_1>0$, the values of $|Q(h)|$ for low $|h|$ increase by the action of the first term and then $Q_a(h)$ is formed by the third term. This process corresponds to spinodal decomposition, because the composition variation amplifies over the whole range of $L$. The formation of $Q_a(h)$ at the later stage of aging causes the composition variation to be squared. In low solute alloys for which $D_0>0$ and $D_1>0$, $Q_d(h)$ is formed by the action of the second term while the initial values of $|Q(h)|$ decrease by the first term. $Q_d(h)$ produces a uniform matrix and a sharp peak of composition variation at $x=0$. When the third term is possible to come into action after the operation of the second term, $Q_a(h)$ is produced. This causes that the composition peak is squared and rises up to $c_{e2}$. The composition peak beyond $c_{e2}$ is considered to correspond to what is called a nucleus. The size of the nucleus increases by subsequent diffusion, which is attributed to a complex effect of the first term and the third term. Thus we regard this decomposition process as nucleation-growth.

In high solute alloys $Q_a(h)$ is generated with evolution of composition variation without any delay. So eq. (8) always holds in the range where $\tilde{D}_{x=0}>0$, and the composition peak necessarily rises up to the equilibrium composition. In low solute alloys the progress of decomposition depends on whether eq. (8) holds or not. Roughly speaking, in alloys of $D_1/D_2<0.7$ the spectrum change from $Q_d(h)$ to $Q_a(h)$ is made before the composition peak attain $c_{e2}$, and the composition peak rises up to $c_{e2}$ by that $\psi$ continues to have a larger value than $\tilde{D}_{x=0}$. In alloys of lower solute content, the spectrum change from $Q_d(h)$ to $Q_a(h)$ is delayed and this makes the rise of the composition peak beyond $c_{e2}$ impossible. In further low solute alloys eq. (8) does not hold even inside the spinodal owing to the remarkable development of $Q_d(h)$.

### III. Computer Simulation

#### 1. Experimental method

For supersaturated solid solutions whose $D_0/D_2$ and $D_1/D_2$ are given in Fig. 2, the changes of $Q(h)$ with aging time were computed using eq. (18). $q_x=0$, $D_{x=0}$, $\psi$, $\xi$, and $\phi$ were estimated from $Q(h)$, where $\phi$ is given by

$$\phi = (\beta/2\pi) \int_{-\pi/\beta}^{\pi/\beta} (c-c_0)^2 dx = \sum_k Q_k^2(h).$$

As the cases where a large composition peak exists only at $x=0$ are treated in this paper, $q_x=0$ is equal to the maximum value of a composition variation. $-\xi$ is a curvature of
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The profiles of Fourier spectra depend on the activity of each term in eq. (18): the values of \((D_0 + 2h^2K\beta^2)/D_2\) and \(D_1/D_2\), and the amplitude of composition variation. As \(D_0/D_2\) and \(D_1/D_2\) are settled by alloy composition, we can choose only the ratio of \(D_2\) to \(K\beta^2\) and the initial values of \(Q(h)\). In the experiment it is assumed that \(D_2 = 10^{-20}\) m²/s⁴ and \(K = 1.0125 \times 10^{-41}\) m⁴/s⁶, and various values were chosen for \(\beta^2\). The spectrum range on the calculation was chosen so as to include all Fourier waves for which \(|g(h)| < 10^{-6}\). When the difference between \(\psi\) and \(\Delta x = 0\) is small as in Fig. 4, Fourier waves of much higher frequency were included in the calculation in order to obtain a strictly smooth \(\psi\) curve.

2. Experimental results

\(D_0/D_2 = -0.0676\) and \(D_1/D_2 = 0\) in the alloy of \(c_0 = 0.50\). Figure 3 shows the changes of the parameters with aging in the case where the initial composition variation, \(q_i\), is given by \(q_i = 0.01 \cos \beta x\) along with \(\beta^2 = 5 \times 10^{19}/9\) m⁻². \(\psi\) is negative at the initial stage of diffusion, but \(\Delta x = 0\) increases because \(\psi > \Delta x = 0\). The increase of \(\Delta x = 0\) gives rise to the generation of \(Q(h)\), which changes \(\psi\) to a positive value. Although \(\Delta x = 0\) becomes positive when the composition peak exceeds the spinodal line, \(\psi\) becomes positive before \(\Delta x = 0\) attains zero. The rising rate of the composition peak is given by \((\psi - \Delta x = 0) \cdot \xi\). This value is high at \(t = 1000 \sim 1200\) s. Figure 3 shows that \(\phi\) is saturated at the short period of aging through the process of the large increasing rate in \(\phi\). This means that decomposition progresses over the whole range of diffusion area and is completed in the short period of diffusion. Equation (9) is satisfied when the composition peak reaches a near-equilibrium composition.

\(D_0/D_2 = 0.0224\) and \(D_1/D_2 = 0.6\) in the alloy of \(c_0 = 0.20\) which is outside the spinodal. Figure 4 shows the changes of the parameters during aging in the case where \(q_i = 0.1 \cos \beta x\) and \(\beta^2 = 10^{18}/9\) m⁻². After the long incubation period, the composition peak rises rapidly at \(t = 5500 \sim 7000\) s and reaches the vicinity of

![Fig. 3 Changes of \(q_x = 0\), \(\phi\), \(\Delta x = 0\), \(\psi\) and \(\xi\) with diffusion. \(c_0 = 0.50, q_i = 0.01 \cos \beta x\) and \(\beta^2 = 5 \times 10^{19}/9\) m⁻².](image1)

![Fig. 4 Changes of \(q_x = 0\), \(\phi\), \(\Delta x = 0\), \(\psi\) and \(\xi\) with diffusion. \(c_0 = 0.20, q_i = 0.1 \cos \beta x\) and \(\beta^2 = 10^{18}/9\) m⁻².](image2)
This rapid rise corresponds to nucleation. When the composition peak rises up to near \(c_x = c_{x_2}\), the rising rate of the composition peak becomes slow by the decrease in \(\psi - \tilde{D}_{x=0}\) and \(\xi\). \(\phi\) decreases at the initial stage of aging. This means that the smooth composition variation over the whole range of diffusion area changes to the uniform matrix and the sharp peak at \(x=0\). \(\phi\) turns to increase when the rapid rise of the composition peak begins, and \(\phi\) continues to increase even after the increase of \(q_x = 0\) has almost saturated. The consecutive increase of \(\phi\) results from the fact that the size of the nucleus increases after the composition of the nucleus has attained near \(c_x = c_{x_2}\). In other words, the decomposition progresses by the growth of the nucleus at the later stage of aging. \(\tilde{D}_{x=0}\) attains the lowest value at \(c_x = 0.14\) and then increases. \(\psi\) decreases remarkably once by the development of \(Q_d(h)\) and then changes to a positive value by the formation of \(Q_a(h)\). The figure shows that \(\psi - \tilde{D}_{x=0}\) continues to be narrowly positive. This is the reason why the composition peak can rise beyond \(c_x = c_{x_2}\) and the decomposition progresses up to its completion.

If the increase in \(\psi\) occurred slightly later in Fig. 4, the \(\psi\) and the \(\tilde{D}_{x=0}\) curve would intersect and the composition peak would turn from rising to sinking. Such an example is shown in Fig. 5. In the alloy of \(c_0 = 0.14\) for which \(D_0/D_2 = 0.062\) and \(D_1/D_2 = 0.72\), it is assumed that \(q_i = 0.05 \sum_{h=1}^{3} \cos h\beta x\) along with \(\beta^2 = 10^{18}/9\) m\(^{-2}\). Unless eq. (8) holds consecutively until the completion of decomposition, the composition variation disappears ultimately and the uniform solid solution is formed as in this case. When the same values as in Fig. 5 are given for \(q_i\) and \(\beta^2\) in the alloy of \(c_0 = 0.15\), the \(\psi\) and the \(\tilde{D}_{x=0}\) curve do not intersect and the decomposition progresses up to the completion. The alloy composition of \(c_0 = 0.14\) is considered to be nearly a limit for decomposition by the process that a composition variation of small amplitude evolves only by diffusion.

Figure 6 shows the behavior of composition peak in the alloy of \(c_0 = 0.10\) with \(D_0/D_2 = 0.924\) and \(D_1/D_2 = 0.8\). The initial condition which represents a narrow and high composition peak is given by \(q_i = 0.02 \sum_{h=1}^{3} \cos h\beta x\) along with \(\beta^2 = 10^{18}/9\) m\(^{-2}\). Although high frequency waves of negative components are generated at the early stage of diffusion and \(\psi\) increases rapidly, \(Q_d(h)\) does not develop so sufficiently that the \(\tilde{D}_{x=0}\) and the \(\psi\) curve do not intersect. Thus \(q_x = 0\) decreases continuously, which causes the change from \(Q_a(h)\) to \(Q_d(h)\). Consequently \(\psi\) decreases, which yields the minimum in the \(\psi\) curve at \(t = 5\sim 6\) s. The increase of \(\psi\) at the low rate subsequent to the minimum is due to shrinkage of the spectrum, and \(\tilde{D}_{x=0} > \psi\) even
for the lowest value of $\bar{D}_x=0$ at $c_0+q_{x=0}=0.5$.
Evidently, the disappearance of the composition variation is attributed to the undevelopment of $Q_a(h)$ and the well-development of $Q_d(h)$ due to the large value of $D_1/D_2$. This means that the initial composition variation should consist at least of $Q_a(h)$ for which $\psi > \bar{D}_{x=0}$, in order that the decomposition in very low solute alloys progresses starting from a local composition variation with large $(q_i)_{x=0}$. Although the required composition profiles for decomposition starting from a local composition variation are under investigation, such the composition variation is considered to have a broad and high composition peak which is accompanied by a solute depleted zone over a wide range.

**IV. Discussion**

The composition peak rises when $-\bar{D}_{x=0} > 0$ in the treatment that the gradient energy is neglected. The composition peak rises when $\psi - \bar{D}_{x=0} > 0$ in the treatment that the gradient energy is taken into consideration. The role of the gradient energy is to promote the increase of $q_{x=0}$ when $\psi$ is positive, while it is to impede the increase when $\psi$ is negative.

The composition peak rises even in the range where $-\bar{D}_{x=0} < 0$ by virtue that $\psi$ is positive in Figs. 3 and 4. While the composition peak sink even in the range where $-\bar{D}_{x=0} > 0$ by virtue that $\psi$ is negative in Figs. 5 and 6. This means that the gradient energy has a vital effect on the behavior of composition peaks in terms of $\psi$. The values of $\psi$ depend on the kinds of Fourier spectra: the development of $Q_d(h)$ decreases $\psi$, while the development of $Q_a(h)$ increases $\psi$. The degree of development of $Q_d(h)$ or $Q_a(h)$ depends upon the values of $D_1/D_2$ which are determined by alloy composition, e.g., $Q_d(h)$ develops well and $Q_a(h)$ does not develop sufficiently in low solute alloys with large values of $D_1/D_2$. By this sequence of causes and effects, composition variations in low solute alloys decay out by virtue of the gradient energy, while those in high solute alloys evolve to the completion of decomposition.

The diffusion behavior of composition variations in low solute alloys is influenced strongly by initial conditions. For example, when the initial composition variations in the alloy of $c_0=0.20$ are given by $q_i=(0.06/n) \sum_{k=1}^{n} \cos \beta kx$ along with $\beta^2=10^{18}/9 \text{ m}^{-2}$, the composition variations behave in the same manner as Fig. 4 for $n=1$, as Fig. 5 for $n=2\sim8$ and as Fig. 6 for $n>9$. The difference of the behavior is caused obviously by the fact that $Q_d(h)$ well develops for large values of $n$.

In the classical theory the formation of nuclei by thermal fluctuation is a requisite for phase separation. Borelius considered that a composition peak can rise spontaneously when the peak composition exceeds $c_2$ in Fig. 1, and he regarded such the composition peak as the nucleus(8). However, this idea does not fit the present treatments. For instance, although the initial composition of the peak in Fig. 4 is below $c_2$, the peak rises up to the equilibrium composition by diffusion. We obtain another example of the same situation when the $q_i$ given in Fig. 5 is adopted for the alloy of $c_0=0.15$. On the other hand, the composition peak in Fig. 6 sinks by diffusion, though the initial peak composition exceeds $c_2$. The inconsistency is attributed partly to the neglect of the gradient energy or the interface energy in Borelius's theory. The other cause is that in the classical theory only local composition variations which arise from thermal fluctuation are argued, under the assumption that uniform solid solutions are formed on quenching. However, it is improbable that quenched materials are perfectly uniform(9). As described in the previous paper, a composition variation over a wide region is concentrated at its composition peak through diffusion by the work of the second term in eq. (18)(6).

As solid solutions of $c_0=0.10$ are unstable, we should expect the progress of decomposition for large initial composition variations. However, the foregoing result gives a strong restriction on the profiles of composition variations which are able to grow by subsequent diffusion. It seems improbable that a wide and high composition peak enclosed by a solute depleted zone over a extensive range is formed by thermal fluctuation. The author would rather insist that the limit of homogeneous precipitation is equal to the composition limit.
of $c_0=0.14$ or so, beyond which composition variations of a small amplitude over a wide range amplify up to the completion of decomposition only by diffusion.

REFERENCES