Relaxation of the intermediate decay structure of glasses and of solid solutions to a state of complete thermodynamic equilibrium

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We consider the relaxation of the intermediate decay structure of metallic glasses and solid solutions in the following stages after the spinodal decay has been completed. We show that in that stage one can characterize the quasistationary structures as a whole by some new (generally speaking, nonconserved) order parameters, such as the chemical potential, the internal energy, or the pressure. By means of this approach, which uses the Landau–Khalatnikov equation, we study the special case of one-dimensional symmetric relaxation when the mean density c_0 of the alloy is exactly equal to half the sum of the binodal densities. We obtain the time dependence of the characteristic period of the structure both in the initial binodal decay stage and in the coalescence stage. For the one-dimensional model we also establish the temperature dependence of the characteristic scales of the structure and of its relaxation time. We give an estimate of the width of the interphase boundary in the binodal relaxation stage.

1. INTRODUCTION

We consider, as before,[1,2] a two-component glass (or a solid solution) with a limited solubility of one of the components in the solid state. A typical state diagram of such a system in the $T-c$ variables (T is the temperature and c the atomic concentration of one of the components) is shown in Fig. 1. Homogeneous states are stable in region *1*, region *2* corresponds to metastability, and the states in region *3* are unstable against any deviation of the concentration from uniformity. For the unstable states one observes a spontaneous growth of small concentration perturbations in the whole of the volume of the sample; this is usually called spinodal decay. To observe spinodal decay in practice it is necessary to transfer the system as fast as possible from the region *1* into the region *3*, for instance, through quenching from the liquid state. If, however, the cooling is slow a complete or partial decay to equilibrium phases can proceed through the classical mechanism of nucleus formation even in the transfer process, i.e., in region *2*. On the other hand, in the case of fast cooling it turns out that in the systems considered only those degrees of freedom corresponding to long-range order of the interatomic distances manage to relax. The hydrodynamic modes, on the other hand, are found to be frozen in, as it were. This is because the viscosity "tracks" the temperature and it is just the viscosity which determines the rate of the relaxation of the hydrodynamic modes (in our case the large-scale fluctuations in the constitution). Estimates of the corresponding relaxation times are given, for instance, in Ref. 2.

We thus assume that the superfast cooling has already taken place, i.e., that the system turned out to be in a strongly nonequilibrium (labile) state. Our problem is to consider the subsequent evolution of the system under isothermal conditions. Such evolution does not require, at least not in the early stages, that energy barriers have to be surmounted through fluctuations. It is completely natural in this connection to consider the spinodal decay process on the basis of a differential equation (known as the generalized Cahn diffusion equation[3]). The basis of the distinctive feature of this equation is the negative diffusion coefficient which is responsible for upward diffusion. The use of such an approach in papers by Cahn,[4,5] Filipovich,[6,7] Khachaturyan and Suris,[8] Langer,[9] Mitlin,[10–13] and ourselves[1,2] made it possible to explain a number of the basic features of the spinodal decay. In particular, it was established that spinodal decay in solid solutions, polymers, or metallic glasses may lead to the appearance of intermediate quasistationary structures with mesoscopic scale inhomogeneities. Such structures are formed by periodically repeated fragments and in the general case are metastable. The metastability of these structures is connected with the slowing down of the kinetics, as was pointed out in Ref. 11. The free energy of the system in the kinetic stabilization stages then remains practically unchanged.

The intermediate decay structures which appear in the kinetic stabilization stage of the spinodal decay are quasistationary but they are still far from complete thermodynamic equilibrium. To study the relaxation of an intermediate decay structure to a state of complete thermodynamic equilibrium (on the level of a detailed description) it would be necessary to write down a nonlocal equation (e.g., an integro-differential equation) for the probability distribution $c(\mathbf{r})$ in function space. For a "crude" description, however, it is sufficient to bear in mind that in a phenomenological approach intermediate quasistationary structures can be characterized by a few nonequilibrium parameters such as, for instance, the chemical potential, the pressure, or the internal energy. These nonequilibrium order parameters characterize the system as a whole, rather than separate parts of it, and their relaxation to thermodynamic equilibrium can be studied by treating simple kinetic equations.

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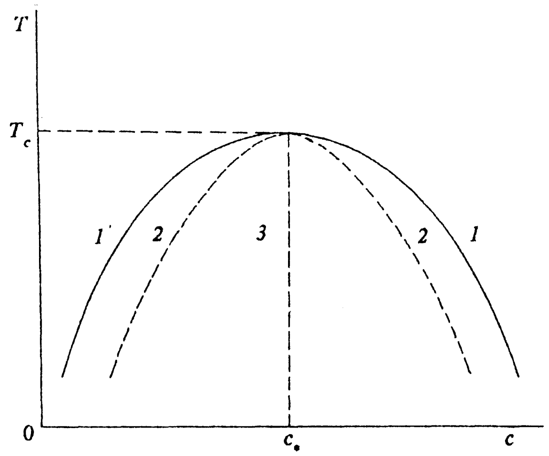

FIG. 1.

2. CHOICE OF VARIANT OF A THEORY OF THE RELAXATION OF AN INTERMEDIATE DECAY STRUCTURE

To describe the relatively slow evolution of large-scale concentration variations $c(\mathbf{r},t)$ of one of the components in a glass or a solid solution one starts from the relation connecting the particle flow density $\mathbf{j}$ and the gradient of the generalized chemical potential $\mu(\mathbf{r},t)\equiv\delta\mathcal{F}/\delta c(\mathbf{r},t)$:

$$\mathbf{j}(\mathbf{r},t)=-M\nabla\frac{\delta\mathcal{F}}{\delta c(\mathbf{r},t)}. \tag{1}$$

Here $\mathcal{F}$ is the free energy of the glass and M is the product of the volume concentration and the mobility of the atoms of the component considered. It is very important that the concentration satisfies the conservation law

$$\frac{\partial c}{\partial t}+\operatorname{div}\mathbf{j}=0. \tag{2}$$

The description of the relaxation based upon Eqs. (1) and (2) has a meaning up to the moment when the chemical potential μ (expressed as the functional derivative of the free energy with respect to the concentration) becomes spatially uniform in the whole of the system. According to (1) mass transfer in the system then stops and the stationary concentration distribution which was studied in detail in Ref. 8 is established. However, in actual fact, the relaxation processes in the system are not completed by this since the absolute minimum of the free energy is reached for a completely determined $\mu=\mu_{\min}$, whereas the value of μ established as a result of the spinodal decay is in no ways necessarily the same as $\mu_{\min}$. There is, apparently, a whole set of values of μ which correspond to local minima of the free energy functional $\mathcal{F}\{c(\mathbf{r})\}$ in the concentration configuration space. One can say that the intermediate decay structures correspond to states of partial equilibrium.

The transition from one partial equilibrium to another, the absolute minimum of the free energy, is reached, is no longer described by Eq. (1) since it neglects two principal points. Firstly, as was especially clearly shown in Ref. 14, the true relaxation equation must be nonlocal. Meanwhile, the local nature of (1) imposes nonphysical limitations on the cooperative behavior of the system and on the choice of the energetically favorable stationary structure. The present authors, for instance, have earlier in Ref. 2 obtained the way the spatial scale of an intermediate decay structure depends on the correlation radius of the initial fluctuations. The situation here is equivalent to the motion of a ball in a "mountain relief" filled with a viscous fluid when the falling of the ball into one or other local minimum of the relief depends to a large extent on the initial conditions.

Secondly, the true equation must contain random forces reflecting the influence of fluctuations caused by the finite temperature. Over long times, thanks to these forces, one can surmount the energy barriers which separate the kinetically stable intermediate structures described in Ref. 8 from the thermodynamically favorable ones.

In connection with the considerations presented above it is completely appropriate to give a palliative description of the relaxation of an intermediate decay structure on the basis of the phenomenological Landau–Khalatnikov equation:[15]

$$\frac{d\eta}{dt}=-\gamma\frac{\partial\mathcal{F}}{\partial\eta}, \tag{3}$$

where γ is a kinetic coefficient and η a suitably chosen order parameter which [in contrast to $c(\mathbf{r},t)$] characterizes the total system as a whole and which depends only on the time. The parameter η thus determines the evolution of the spatial concentration distribution.

We turn now to a specific choice of η and to finding explicitly the function $\mathcal{F}(\eta)$. Following Ref. 2 we shall consider only those intermediate decay structures which are described by a one-dimensional concentration distribution, $c(x,y,z)\equiv c(x)$. There is some basis for assuming[16] that a one-dimensional concentration stratification is energetically more favorable than a multidimensional stratification.

The free energy of the system is according to a popular model[8] described by the following functional:

$$\mathcal{F}\{c(x)\}=S\int_{-\infty}^{\infty}\left[f(c)+\frac{\beta}{2}\left(\frac{dc}{dx}\right)^2\right]dx. \tag{4}$$

Here S is the area of the cross-section of the sample; $f(c)$ is the specific free energy; β is a positive constant which is of the same order of magnitude as ρUZa^2, where U is the mixing energy, Z is the coordination number of the given alloy, a is the range of the interatomic interaction, and ρ is the total number of atoms per unit volume.

The stationary states of the system are determined by the extrema of the functional (4) under the additional condition that the total number of atoms of each of the components of the alloy is conserved:

$$S\rho\int_{-\infty}^{\infty}c(x)dx=N \tag{5}$$

with N being the total number of atoms of one of the components.

Finding the conditional extremum of the functional (4) under the additional condition (5) reduces to finding the unconditional extremum of the functional

$$\Omega\{c(x)\}=S\int_{-\infty}^{\infty}\left[f(c)+\frac{\beta}{2}\left(\frac{dc}{dx}\right)^2-\tilde{\mu}c\right]dx, \qquad (6)$$

where the Lagrangian multiplier $\tilde{\mu}$ in (6) is $\tilde{\mu}=\mu\rho$ with μ the chemical potential of the system. The standard variational procedure applied to the functional (6) leads to the following differential equation for the concentration $c(x)$:[16]

$$\beta\frac{d^2c}{dx^2}=\frac{df(c)}{dc}-\tilde{\mu}. \qquad (7)$$

One should emphasize that the chemical potential $\tilde{\mu}$ in Eqs. (6) and (7) is constant in the whole of the system, whereas according to Cahn's equation the decay process proceeds only because of the spatial inhomogeneity of the chemical potential. There is no paradox here since Eq. (7) describes equilibria when the equalization of the chemical potential has already been completed and it is fixed at some level. That level itself, though, remains undetermineed.

Mathematically this indeterminacy follows from the fact that Eq. (7) which has the same form as the equation of motion of a classical and, in general, nonlinear oscillator, has the integral of motion

$$E=f(c)-\tilde{\mu}c-\frac{\beta}{2}\left(\frac{dc}{dx}\right)^2. \qquad (8)$$

It is impossible to determine both constants E and $\tilde{\mu}$ from the single condition (5) and one of them (or some combination of them) remains undetermined.

Here one must choose this undetermined constant as the order parameter since the relaxation of the system, after some partial equilibrium has been reached by it, will proceed through changing from one partial equilibrium to another one which corresponds to a lower free energy until its absolute minimum has been reached.

The further discussion is based on a choice of a particular free energy density function in the form proposed by Landau.[17] At a temperature below the critical one (the temperature corresponding to the vertex of the spinodal cupola (Fig. 1) plays the role of the critical temperature T_c) we have

$$f(c)=\mathscr{E}-\alpha[(c_\beta-c_\alpha)^2(c-c_*)^2-2(c-c_*)^4]. \qquad (9)$$

Here c_α and c_β are the binodal concentrations for which $f(c)$ reaches a minimum, i.e., the concentrations in the two equilibrium phases which make up the system in a state of complete thermodynamic equilibrium, $c_*\equiv\frac{1}{2}(c_\alpha+c_\beta)$ is the concentration in the point where $f(c)$ is a maximum; the constants $\mathscr{E}$ and α have the dimensionality of an energy density and the constant $\mathscr{E}\equiv(\alpha/8)(c_\beta-c_\alpha)^4$ is chosen in such a way that $f(c)$ vanishes in the points where it is a minimum (see Fig. 2). In the model (9) the binodal and spinodal concentrations (c_{s1} and c_{s2}) are positioned symmetrically with respect to the concentration c_* and they are coupled to one another through the relation

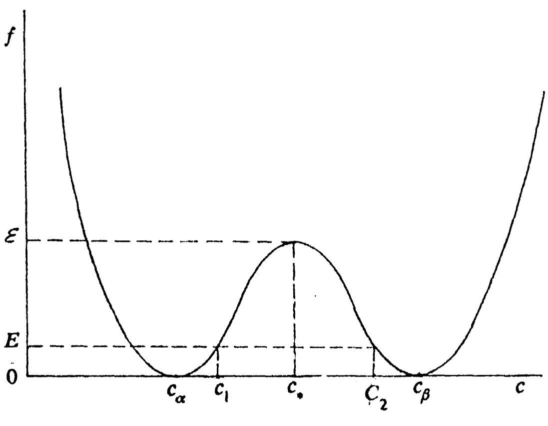

FIG. 2.

$$c_{s2}-c_{s1}=\frac{c_\beta-c_\alpha}{\sqrt{3}}\equiv\frac{c_{\beta\alpha}}{\sqrt{3}}.$$

The quantity E is similar to a pressure; the physical interpretation of this analogy is that in a supercooled gas, first of all the pressure of the gas and its chemical potential equalize in the whole of the volume, and after that they get established at the level corresponding to the Maxwell rule of equal areas.

In solid solutions and metallic glasses the quantities $\tilde{\mu}$ and E are interconnected through a relation following from (5), i.e., the actual form of the relation between $\tilde{\mu}$ and E is determined, apart from anything else, also by the average concentration c_0 of the alloy.

In the case of arbitrary values of c_0 the plan of the solution is the following: first of all we establish from Eq. (8) the form of the extremal $c(x,\tilde{\mu},E)$, then we use (5) to find the relation between $\tilde{\mu}$ and E, $\tilde{\mu}=\tilde{\mu}(E,c_0)$, then we find from (4) the function $\mathscr{F}(E)$ or $\mathscr{F}(\tilde{\mu})$, whichever is the more convenient, and, finally, identifying η, respectively, with E or $\tilde{\mu}$ we solve the Landau–Khalatnikov equation (3).

However, in the general case the realization of this plan encounters serious difficulties (of a rather technical nature), so that the general case need to be considered separately. The main features of the physical pattern of the relaxation of intermediate decay structures can, however, be exhibited by considering the special $c_0=c_*$ case which we may call the symmetric relaxation case. It is completely obvious that for $c_0=c_*$ the deviation of $c(x)$ from c_0 occurs with equal probability in both directions; the $c(x,t)$ curve is symmetric relative to the horizontal $c=c_0$ at any time and condition (5) is satisfied automatically while Eq. (8) is compatible with this symmetry only for $\tilde{\mu}\equiv0$. In this situation it is natural to choose as the order parameter the quantity E and to trace its relaxation in what follows.

3. SYMMETRIC RELAXATION

If we have $\tilde{\mu}=0$, it follows from (4) and (8) that

$$\mathscr{F}=S\int_{-\infty}^{\infty}[2f(c)-E]dx. \tag{10}$$

The analogy with the one-dimensional oscillator problem indicates[8] that for $0<E<\mathscr{E}$ the concentration changes periodically, "oscillating" between the values $c_1(E)$ and $c_2(E)$ (the turning points of the system), which are determined by the condition

$$f(c)=E. \tag{11}$$

From the four solutions of this equation we choose those two with $f(c)>E$ in the interval between them (see Fig. 2). The period of the structure is determined by an integral of the form

$$\lambda(E)=\sqrt{2\beta}\int_{c_1(E)}^{c_2(E)}\frac{dc}{\sqrt{f(c)-E}}. \tag{12}$$

Using the periodicity of the integrand in (10) with period λ in the coordinate x and replacing the integration over x by integration over c we are led to the formula:

$$\mathscr{F}(E)\equiv Vf(E)=\left\{\frac{\sqrt{2\beta}}{\lambda}\int_{c_1(E)}^{c_2(E)}\frac{2f(c)dc}{\sqrt{f(c)-E}}-E\right\}V, \tag{13}$$

where V is the volume of the sample.

One can evaluate the integrals (12) and (13) in two limiting cases: 1) when E is close to $\mathscr{E}$; 2) when E is close to zero. The corresponding criteria look as follows: in the first case we have $|E-\mathscr{E}|\ll\mathscr{E}$, and in the second case we have $E\ll\mathscr{E}$.

For the first case we restrict ourselves to writing down the results:

$$f(E)\equiv\frac{\mathscr{F}(E)}{V}=\mathscr{E}-\frac{(\mathscr{E}-E)^2}{4\alpha c_{\beta\alpha}^2}, \tag{14}$$

$$\lambda(E)=\sqrt{2}\pi\left(\frac{\beta}{\alpha}\right)^{1/2}\frac{1}{c_{\beta\alpha}}\left(1+\frac{\mathscr{E}-E}{2\alpha c_{\beta\alpha}^4}\right). \tag{15}$$

We now consider the second limiting situation when $E\to0$; this corresponds to a state of the alloy which is very close to complete thermodynamic equilibrium. Since in that case the concentrations $c_1(E)$ and $c_2(E)$ satisfy the inequalities

$$\frac{|c_1-c_\alpha|}{c_\alpha}\ll1,\quad\frac{|c_2-c_\beta|}{c_\beta}\ll1,$$

when evaluating the integrals (12) and (13) we can approximate the function $f(c)$ near $c_1(E)$ and $c_2(E)$ by the expressions $f_\alpha(c)\approx\xi(c-c_\alpha)^2$ and $f_\beta(c)\approx\xi(c-c_\beta)^2$, respectively, where we have $\xi=2\alpha c_{\beta\alpha}^2$. To find the integration limits in (12) and (13) we use the approximate expressions for $f(c)$ indicated above. Furthermore, in view of the symmetry of the integrals (12) and (13) each of them can be obtained by taking twice the integral taken with the limits from $c_1(E)$ to c_*.

The integral determining the period λ of the structure for $E\to0$ diverges logarithmically for $c\approx c_1\approx c_\alpha$. To evaluate this integral we can thus replace the function $f(c)$ with logarithmic accuracy by its limiting expression $\xi(c-c_\alpha)^2$, not only near c_α, but also in general in the whole integration range from $c_1(E)$ to c_*. The integral can then be evaluated and the result gives the dependence of the period λ of the structure on the parameter E

$$\lambda(E)\approx\left(\frac{\beta}{\alpha}\right)^{1/2}\frac{1}{c_{\beta\alpha}}\ln\frac{2\alpha c_{\beta\alpha}^4}{E}. \tag{16}$$

Logarithmic accuracy means that we cannot guarantee that the constant standing under the logarithmic sign in (16) is correct.

Similar considerations are valid also for evaluating the integral (13) which determines the free energy density:

$$f(E)\approx\frac{\alpha c_{\beta\alpha}^4}{\ln(2\alpha c_{\beta\alpha}^4/E)}. \tag{17}$$

We assume that the relaxation process consists in decreasing the order parameter E from some initial value E_0 to zero. If this is the case the free energy $\mathscr{F}(E)$ tends to zero in this process and the period of the structure increases, according to (15) and (16), to become infinite (in reality, of course, the period of the structure is bounded by the size of the sample).

The speed with which the order parameter E approaches its equilibrium value $E=0$ can be determined from the Landau-Khalatnikov equation (3). The initial value of the order parameter E_0 is determined by the characteristics of that intermediate decay structure the formation of which was completed in the spinodal stage of the decay. In particular, we shall assume that the initial value of the period of the structure is the same as the correlation radius R_c of the intermediate decay structure, which was determined in Ref. 2. Substituting the quantity R_c for λ in (15) or (16) we can determine the initial value of the order parameter $E_0=E(R_c)$.

The solution of Eq. (3) ($E\equiv\eta$) in the $|E-\mathscr{E}|/\mathscr{E}\ll1$ range has the form

$$E(t)=\mathscr{E}-(\mathscr{E}-E_0)e^{(t/\tau)}, \tag{18}$$

where we have $\tau=2\gamma^{-1}\alpha c_{\beta\alpha}^4$, i.e., in the initial stage the parameter E decreases exponentially fast. In order to describe the kinetics of the stratification of the alloy in the final stage (as $E\to0$) we must solve Eq. (3) using for $f(E)$ the value given by Eq. (17). We then get

$$E(t)=\sqrt{E_0^2-2\gamma\alpha c_{\beta\alpha}^4t}. \tag{19}$$

One sees easily from (19) that the stratification time turns out to be finite:

$$t_r=\frac{E_0^2}{2\gamma\alpha c_{\beta\alpha}^4}. \tag{20}$$

The completion of the relaxation of the nonequilibrium system in a finite time (20) is a reflection of the one-dimensional character of the problem and it apparently does not occur in real systems.

Knowing the function $E(t)$ makes it possible to determine the time dependence of the quantity which physically is of most interest—the period λ of the concentration inhomogeneities. From Eqs. (15) and (18) it follows that λ grows exponentially with time in the strong nonequilibrium stage ($|\mathscr{E}-E|\ll\mathscr{E}$).

At long times we find (as $E\to 0$) from (16) and (20)

$$\lambda(t)=\lambda_C \ln\left(\frac{4\alpha^2 c_{\beta\alpha}^8}{E_0^2-2\gamma\alpha c_{\beta\alpha}^4 t}\right), \tag{21}$$

where $\lambda_C\approx(\beta/\alpha)^{1/2}c_{\beta\alpha}\propto a/c_{\beta\alpha}^{-1}$ is the Cahn scale.[3] Equation (21) qualitatively reflects the fact that one observes in the coalescence stage a growth of the large-scale inhomogeneities due to the vanishing of the small size inhomogeneities. As far as a logarithmic, rather than a power-law (according to the law[18] $\lambda\propto t^{1/3}$) growth is concerned, this indicates here again the specific behavior of the one-dimensional problem.

The binodal decay time is to a large extent determined by that scale R_c of the intermediate decay structure from which this binodal decay starts. If we have $R_c\sim\lambda_C\propto(\beta/\alpha)^{1/2}c_{\beta\alpha}^{-1}$, the initial value of the parameter E is close to $\mathscr{E}$ and the decay time is

$$\tau\sim\frac{\alpha c_{\beta\alpha}^4}{\gamma}\propto\frac{\alpha}{\gamma}\left(\frac{T_c-T}{T_c}\right)^2, \tag{22}$$

i.e., this time decreases strongly as one approaches the critical point.

If, however, we have $R_c\gg\lambda_C$, we can easily ascertain from (16) and (20) (substituting in (16) the quantity R_c instead of λ) that the decay time is exponentially small:

$$t_r\propto\exp\left(-\frac{2R_c}{\lambda_C}\right). \tag{23}$$

The fact that the time t_r of the binodal stage is small can be explained by the fact that large-scale "frozen-in" concentration fluctuations were formed either already in the quenching stage, or they were formed in the spinodal decay stage.

CONCLUSION

Let us consider in more detail the analysis of the results obtained. We showed in Ref. 2 that as the result of the spinodal decay of a glass or a solid solution a kinetically frozen-in (but, in general, nonequilibrium) intermediate decay structure is formed which is characterized by some scale R_c which is either the same as the Cahn length λ_C or larger than it ($R_c>\lambda_C$). We established above that such a structure can also be described by means of some nonequilibrium order parameter. This parameter relaxes, beginning from some starting value which is determined as the result of the preceding spinodal decay. The relaxation of this order parameter describes the further decay of the system which can, in general, develop according to two scenarios.

The first of those is realized in the case when the initial value of the parameter E falls into the range of values $|\mathscr{E}-E|\ll\mathscr{E}$, i.e., when the system after the completion of the spinodal stage still remains strongly nonequilibrium. The concentration profile of the structure from which the subsequent relaxation starts off (in the symmetric case when we have $c_0=c_*$) is sinusoidal in this situation. In the strong nonequilibrium region the period λ of the sinusoidal concentration waves has the Cahn length λ_C as scale, where

$$\lambda_C\propto\left(\frac{\beta}{\alpha}\right)^{1/2}\frac{1}{c_{\beta\alpha}}\approx\frac{a}{c_{\beta\alpha}}, \tag{24}$$

with a the range of the interatomic interaction, and as relaxation takes place it changes very little [see (15)] in the region indicated above. The kinetics of the stratification is then basically characterized by the growth in the amplitude of the concentration waves, since the amplitude values of the concentrations $c_1(E)$ and $c_2(E)$ are still very far from their equilibrium values c_α and c_β, respectively.

We note that the period of the concentration waves depends, in general, also on the temperature. It follows from (15) that this dependence is mainly determined by the factor

$$c_{\beta\alpha}^{-1}\propto\left(1-\frac{T}{T_c}\right)^{-1/2}, \tag{25}$$

whence it follows that the spatial scale λ of the concentration fluctuations is considerably larger than the range a of the interaction ($\lambda\gg a$) only in the immediate vicinity of the critical point ($T\to T_c$). It is clear from (25) that this behavior has a scaling character with a critical index "$-1/2$".

To go on, for $E\ll\mathscr{E}$ the pattern of the relaxation changes completely. Firstly, the period λ of the structure corresponding to such a value of E is already larger than the Cahn scale ($\lambda\gg\lambda_C$).[2] Secondly, the concentration fluctuations themselves cease to be sinusoidal. The concentration profile $c(x)$ of the structure is now characterized by two spatial scales: one of them gives the average size of the "plateau" with concentrations close to the equilibrium ones (this is, in fact, the size of a "seed" of the new phase). The second scale corresponds to the width δ of the transition region between the almost equilibrium phases. The quantity δ can be estimated as follows:

$$\delta\approx\frac{\Delta c}{|dc/dx|}, \tag{26}$$

where we have $\Delta c=c_2(E)-c_1(E)$ while the magnitude of the concentration gradient can be found from (8). For $\tilde{\mu}=0$ we have

$$\left|\frac{dc}{dx}\right|=\sqrt{\frac{2}{\beta}\,[f(c)-E]}. \tag{27}$$

To estimate the value of δ as $E\to 0$ we must put $\Delta c\approx c_\beta-c_\alpha\equiv c_{\beta\alpha}$ and $f(c)\approx f(c_0)$ in (27), whence

$$\delta\approx c_{\beta\alpha}\left(\frac{\alpha c_{\beta\alpha}^4}{4\beta}\right)^{-1/2}\approx\frac{2a}{c_{\beta\alpha}}\sim\lambda_C, \tag{28}$$

i.e., the width of the transition region has the Cahn size. Moreover, it remains practically unchanged during the whole binodal relaxation time. Since the period of the structure in this stage decreases fast, according to (16) and (21), we may conclude that this growth is completely caused by the increase in the size of the "plateau" in the concentration profile $c(x)$, i.e., by the increase in the size of the one-dimensional "seeds" of a new phase.

This growth of the new phase region at the start of the binodal stage of the decay occurs at the expense of depletion of the surrounding matrix of the corresponding component. On the other hand, in the concluding stage of the binodal decay the nuclei grow so much that they begin to interact with one another, i.e., the coalescence stage begins when the large nuclei "devour" the small ones; this is described by Eqs. (16) and (21).

We shall not discuss in detail the second scenario of concentration stratification. We merely note that it, in fact, reduces to the concluding stage of the binodal decay (coalescence) which we analyzed above. This scenario is realized in the case when the system has already relaxed, as the result of the spinodal decay,[2] so much that the parameter E falls in the $E \ll \mathscr{E}$ range of values, i.e., when the period of the structure ($\lambda \equiv R_c$) from which the binodal decay starts is at once larger than the Cahn size ($\lambda > \lambda_C$).

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