PHYSICAL REVIEW B VOLUME 55, NUMBER 10 1 MARCH 1997-II

Nonlinear relaxation dynamics in decomposing alloys: One-dimensional Cahn-Hilliard model

A. A. Fraerman,* A. S. Mel'nikov, I. M. Nefedov, I. A. Shereshevskii, and A. V. Shpiro
Institute for Physics of Microstructures, Russian Academy of Sciences 603600, Nizhny Novgorod, GSP-105, Russia

(Received 8 July 1996)

The coarsening process in decomposing alloys is studied within the one-dimensional (1D) Cahn-Hilliard model using both the analytical and numerical methods. We have developed the analytical approach based on the solitonlike description of interacting phase boundaries and obtained the equations of their motion. For a single 1D nucleus in the infinite medium the equation of the interface motion appears to be essentially nonlocal in time. The peculiarities of the dynamic behavior of this system are studied for an arbitrary initial nucleus size a_0. For $a_0 < a_c \sim \ln(1/h)$ the characteristic time scale of the nucleus dissolution is found as a function of a_0 and the supersaturation h in the system. In the limit $a_0 \gg a_c$ we obtained the well-known square-root dependence of the nucleus size $a \sim h\sqrt{t}$. The late stage of the spinodal decomposition in multilayered structures has been investigated. The specific features of the period doubling process are studied. [S0163-1829(97)00105-7]

I. INTRODUCTION

A number of important physical aspects of relaxation kinetics in decomposing alloys may be described within the phenomenological Cahn-Hilliard model (see, e.g., Refs. 1,2). The intensive study of these phenomena was stimulated, in part, by various applications of a wide class of materials consisting of the components with the limited solubility, including artificial layered compounds Fe/Cr,[3] Co/Cu,[4,5] and Me(Fe,Ni,Pt, ...)/C.[6,7] In this paper we develop a solitonlike description of the coarsening process in one-dimensional (1D) decomposing alloys. The 1D case is most relevant to the artificial layered systems. The typical phase diagram of a decomposing binary (AB) alloy is schematized in Fig. 1. The solid line is the equilibrium two-phase boundary, and the dashed line is the spinodal curve. According to the Cahn-Hilliard theory[8] the states lying between these two curves are metastable, while the states lying below the spinodal curve are unstable to weak long-wavelength fluctuations. The growth of these fluctuations leads to formation of the A and B phase domains. We use the notations $C^{(1)}$ and $C^{(2)}$ for the equilibrium concentrations of these phases. When the inhomogeneity scales are much greater than a lattice spacing, one may use a continuum approximation. In this case the free-energy functional has the form:

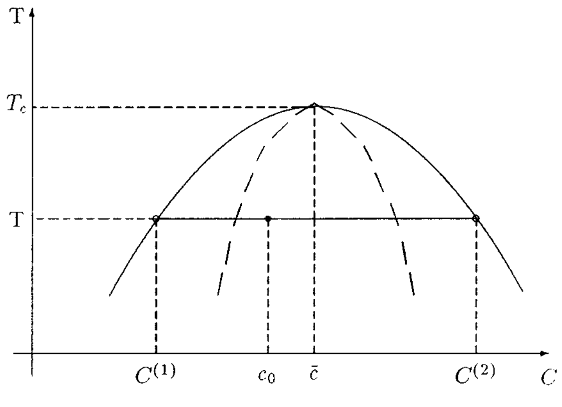

FIG. 1. The phase diagram of a decomposing binary alloy. Solid line is the equilibrium two-phase boundary, the dashed line is the spinodal curve, T_c is the critical temperature, $C^{(1,2)}$ are the equilibrium phase concentrations and c_0 is the average alloy concentration.

$$F=\int\left[\frac{1}{2}\gamma(\nabla C)^2-\frac{1}{2}\alpha(C-\overline{c})^2+\frac{1}{4}\beta(C-\overline{c})^4\right]d\mathbf{R}, \quad (1)$$

where $\alpha=\overline{\alpha}(T_c-T)$ and positive coefficients $\overline{\alpha}$, β, γ depend on the pressure, temperature, and crystal structure. These coefficients can be defined from the microscopic theory, as well as the $\overline{c}$ value. In the case of a binary mixture there is an additional conservation condition for the atoms of each sort:

$$\frac{1}{v}\int c\,d\mathbf{R}=c_0,$$

$$c=(C-\overline{c})\sqrt{\frac{\beta}{\alpha}} \quad (2)$$

where v is the volume of the system, c_0 is the average concentration. As a consequence the dynamics of the order parameter may be described using the continuity equation which reads[8]

$$\frac{\partial C}{\partial t}+D\Delta\frac{\delta F}{\delta C}=0, \quad (3)$$

where D is the kinetic coefficient. Introducing the dimensionless variables $c=(C-\overline{c})\sqrt{\beta/\alpha}$, $t\equiv t\sqrt{D\alpha^2/(4\gamma)}$, $\mathbf{r}=\mathbf{R}/R_0$ ($R_0=\sqrt{2\gamma/\alpha}$), one obtains the Cahn-Hilliard equation:

$$\frac{\partial c}{\partial t}=-\Delta[\Delta c+2(c-c^3)]. \quad (4)$$

We have omitted here the random noise term which expresses the statistical fluctuations and contributes to the right-hand side of Eq. (4).[9] Equation (4) takes account of both the finite interface thickness R_0 and the diffusion type of the dynamic behavior far away from the phase boundaries.

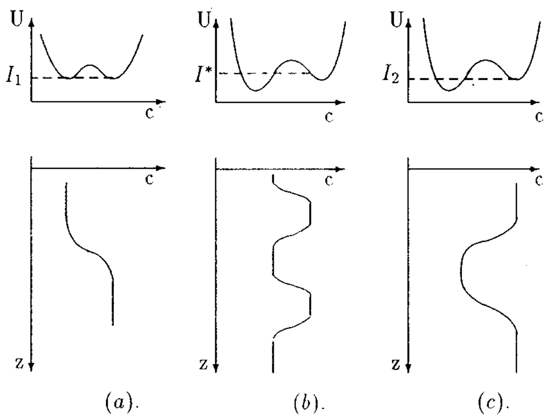

FIG. 2. The 1D stationary solutions of Eq. (4). (a) The stable equilibrium state which corresponds to the decomposition. (b) Periodic structure. (c) The critical new phase nucleus.

We start our treatment of the 1D decomposition dynamics with three remarks that are perhaps obvious.

First, the finite interfacial thickness leads to the exponentially small interaction of the phase boundaries. This interaction accounts for the existence of specific 1D stationary solutions of Eq. (4) (see Fig. 2 and Appendix): (i) periodic structures consisting of domains of different phases; (ii) the unstable critical nucleus of the new phase. Each phase boundary has the form of a kink (or antikink). The critical nucleus size a_c depends logarithmically on the supersaturation degree h which is proportional to the energy difference per one atom in different phases. The h value defines the degree of metastability in the alloy. An arbitrary 1D concentration distribution at the late decomposition stage (and small h) may be considered as a set of moving kinks and antikinks. The 1D coarsening process is caused by their exponential interaction. Note that for 2D and 3D cases this process can be described without taking account of the finite interface thickness.[10]

Second, the solutions considered above appear to be unstable within the Cahn-Hilliard model. A single nucleus dissolves or grows depending on the ratio a/a_c, where a is the initial nucleus size. A periodic concentration distribution is unstable with respect to the period doubling process.[11]

Third, far away from the interfaces the concentration disturbance produced by the kink motion satisfies the ordinary diffusion equation (see also Sec. II). This diffusion field is responsible for a force acting on a phase boundary. For the 1D system the characteristic time scale of the diffusion field relaxation is $t_d \sim d_m^2$, where d_m is the maximum distance between the neighboring kinks (for a single nucleus in the infinite medium $d_m \to \infty$). As a consequence, for $t < t_d$ the time dispersion in our system plays an essential role and the equation of kink motion should be nonlocal in time.

In Sec. II we develop the analytical method for description of the late nonlinear stage of the decomposition in the 1D case. Contrary to the treatment in Refs. 11–13, our method is based on the solitonlike description of the migration of interfaces. We employ the approach analogous to the one developed previously for the solution of a large number of nonlinear nonstationary problems,[14] including description of the dynamics of new phase nuclei in 2D and 3D cases.[15–17] We obtain the system of equations which describe the dynamics of the interfacial fronts in the 1D case. In Sec. III we study the dynamics of the 1D new phase nucleus and show that the equation which describes the layer thickness evolution is essentially nonlocal in time. The interaction of the phase boundaries is shown to result in the nucleus dissolution even for positive values of the supersaturation degree. In Sec. IV we investigate the late stage of the spinodal decomposition in finite-size multilayer structures. We define the characteristic time scale of the period doubling and compare the analytical results with the ones obtained numerically.

II. BASIC EQUATIONS

In this section we discuss the relaxation dynamics in systems where the initial $c(z)$ distribution does not correspond to the stationary one. At the early stage of decomposition (when the concentration is far from equilibrium) Eq. (4) may be linearized in c and solved analytically.[8] This solution predicts a limitless growth of concentration fluctuations. The stage of formation of domains with concentrations close to the equilibrium ones cannot be described within the linear theory. Hereafter we restrict ourselves to the late stage, when the domains and interfacial layers have been already formed. We also will not take into account a random noise term, which, in principle, may be included in Eq. (4).

Now we formulate a method of solution of the Cahn-Hilliard equation, which enables us to describe analytically the late stage of the dynamics of the 1D decomposing alloy. In the 1D case the Cahn-Hilliard equation (4) takes the form

$$\frac{\partial c}{\partial t} = -\frac{\partial^2}{\partial z^2}\left(\frac{\partial^2 c}{\partial z^2} + 2(c - c^3)\right). \tag{5}$$

This equation should be supplemented by the boundary conditions. The order-parameter conservation condition (2) yields the first pair of boundary conditions:

$$j = \frac{\partial}{\partial z}\left(\frac{\partial^2 c}{\partial z^2} + 2(c - c^3)\right)\bigg|_{0,L} = 0, \tag{6}$$

where L is the system size. The second pair of boundary conditions is determined by the segregation effects[18] and supersaturation degree (see Secs. III, IV). Let us take the initial concentration distribution

$$c(z, t=0) = c_0 + \sum_{n=1}^{N} (-1)^n \tanh[z - z_n(t=0)], \tag{7}$$

where z_n are the coordinates of interfaces, c_0 is defined by the average concentration of the alloy. Note, that each term in the sum (7) is the stationary solution of Eq. (5). The following treatment is based on the assumption that the form of the interfacial fronts is slightly changing during their migration. We search for the solution in the form

$$c(z,t) = u(z,t) + c_0 + \sum_{n=1}^{N} (-1)^n \tanh[z - z_n(t)]. \tag{8}$$

Assuming the velocities $V_n=\partial z_n/\partial t$ and the correction $u(z,t)$ to be small, one has

$$\frac{\partial u}{\partial t}-y(z,t)=-\frac{\partial^2}{\partial z^2}\left(\frac{\partial^2 u}{\partial z^2}-4u+6ug(z,t)+f(z,t)\right), \tag{9}$$

where

$$g(z,t)=\sum_{n=1}^{N}\frac{1}{\cosh^2(z-z_n)},$$

$$y(z,t)=\sum_{n=1}^{N}(-1)^n\frac{V_n}{\cosh^2(z-z_n)},$$

$$f(z,t)=2\left[c_0+\sum_{n=1}^{N}(-1)^n\tanh^3[z-z_n(t)]-\left(c_0+\sum_{n=1}^{N}(-1)^n\tanh[z-z_n(t)]\right)^3\right].$$

The condition of the concentration conservation reads

$$\frac{\partial}{\partial t}\int_0^L u\,dz=2\sum_{n=1}^{N}(-1)^n V_n, \tag{10}$$

i.e., the concentration changes caused by the interfacial fronts migration should be exactly compensated by the change of the integral value of the correction u. It also follows from Eq. (9) that far from interfacial fronts the equation for u takes the form of the ordinary diffusion equation. That is why it is convenient to search for the solution of Eq. (9) in the form

$$u(z,t)=\varphi(z,t)+\psi(z,t), \tag{11}$$

where ψ is the diffusion field produced by the interfacial fronts migration, φ is the correction to the stationary form of the fronts. The functions ψ and φ satisfy the following equations:

$$\frac{\partial\psi}{\partial t}=4\frac{\partial^2\psi}{\partial z^2}+y(z,t), \tag{12}$$

$$\frac{\partial\varphi}{\partial t}=-\frac{\partial^2}{\partial z^2}\left(\frac{\partial^2\varphi}{\partial z^2}-4\varphi+6g\varphi+f+\frac{\partial^2\psi}{\partial z^2}+6g\psi\right). \tag{13}$$

Note, that layer thickness changes are caused by both the interfacial interaction and the diffusion field. Let us consider the operator

$$\hat{L}=-\frac{\partial^2}{\partial z^2}\left(-\frac{\partial^2}{\partial z^2}-6g+4\right).$$

The expansion of the function φ in terms of the eigenfunctions of the operator $\hat{L}$ is

$$\varphi=\sum_n b_n\varphi_n,$$

where $\hat{L}\varphi_n=\lambda_n\varphi_n$. Let us define a conjugate set of functions:

$$\varphi_n=\frac{\partial^2\chi_n}{\partial z^2},\qquad \int_0^L\chi_n\varphi_m dz=0\quad\text{when } n\neq m. \tag{14}$$

After multiplying Eq. (13) by χ_n and integrating over z from 0 to L one has

$$\frac{\partial b_n}{\partial t}=-\lambda_n b_n-\Phi(t),$$

$$\Phi(t)=\frac{\int_0^L\varphi_n(f+\partial^2\psi/\partial z^2+6g\psi)dz}{\int_0^L\varphi_n\chi_n dz}. \tag{15}$$

$$b_n(t)=e^{-\lambda_n t}\int_0^t e^{\lambda_n\tau}\Phi(\tau)d\tau. \tag{16}$$

If the interaction between the interfacial fronts is absent (i.e., they are isolated from each other), the eigenvalue $\lambda=0$ corresponds to the eigenfunctions $\varphi_n=\cosh^{-2}(z-z_n)$. If the distances between the fronts are large but finite, then the N-fold generated eigenvalue $\lambda=0$ splits into N close to zero values. For $\lambda_n<0$ one has a limitless growth of b_n when $t\rightarrow\infty$. For small $\lambda_n>0$ [assuming $\Phi(t)$ to change slowly in time] one has $b_n\leqslant 1$ when $t\leqslant\lambda^{-1}$. That is why the condition of the u value smallness makes us set $\Phi=0$ for each eigenfunction φ_n, which corresponds to the small eigenvalue λ_n. Since the multiplier $\Psi=f+\partial^2\psi/\partial z^2+6g\psi$ is exponentially small, it is enough to require orthogonality of the quantity Ψ to each eigenfunction of the problem with isolated interfacial fronts

$$\int_0^L\frac{1}{\cosh^2(z-z_n)}\left(f+\frac{\partial^2\psi}{\partial z^2}+6g\psi\right)dz=0. \tag{17}$$

These equations contain the diffusion field ψ which is determined by the velocities of kinks [see Eq. (12)] and boundary conditions. In fact, Eq. (17) defines the interfaces migration velocities in the implicit form. The process of diffusion relaxation depends essentially on the distances between kinks. Obviously, the characteristic time of establishment of the quasistationary concentration distribution is $t_d\sim d_m^2$, where d_m is a maximum from the layer thicknesses. Below we discuss two cases: (i) the dynamics of a single pair kink-antikink in an infinite medium ($d_m\rightarrow\infty$, $t_d\rightarrow\infty$), (ii) the dynamics of kinks in a finite-size multilayer structure.

III. DYNAMICS OF A SINGLE 1D NUCLEUS

We now continue with the derivation of the dynamic equation describing the evolution of a single layer in the infinite medium with the average concentration $c=1-h/4$. The initial concentration distribution is

$$c(z,t=0)=\tanh(z-a_0)-\tanh(z+a_0)+1-\frac{h}{4}. \tag{18}$$

Boundary conditions for ψ and φ take the form:

$$\psi|_{\pm\infty}=-\frac{h}{4},$$

$$\varphi|_{\pm\infty}=0. \tag{19}$$

Solving the diffusion equation (12) with the boundary conditions (19) and taking into account only the linear in V terms one obtains the diffusion field values at the points $z=\pm a(t)$. On rearrangement, Eq. (17) can be written as follows:

$$\int_0^t \frac{V(\tau)}{\sqrt{t-\tau}}\,d\tau=\frac{\sqrt{\pi}}{4}[h-16\exp(-4a)]. \tag{20}$$

We consider here the interval $t \gg a^2$, when the uniform ψ distribution inside the layer has been formed. The diffusion field is essentially time dependent, which leads to a strong time dispersion in the equation of motion.

The interaction of the interfacial fronts results in the layer dissolution even for positive values of the supersaturation. In the case when the initial layer size is equal to the critical one $[a_0=a_c=(1/4)\ln(16/h)]$, the velocity of the interfacial fronts migration is equal to zero. The layer thickness grows for $a_0>a_c$. Introducing new variables

$$\xi=4[a(t)-a_0],$$

$$\theta=2^8\exp(-8a_0)t/\pi,$$

$$H=\frac{h}{16}\exp(4a_0), \tag{21}$$

we may rewrite Eq. (20) as

$$\int_0^\theta \frac{\dot{\xi}(s)}{\sqrt{\theta-s}}\,ds=\pi[H-\exp(-\xi)], \tag{22}$$

where $\dot{\xi}=\partial\xi/\partial s$. The value $H=1$ corresponds to the critical layer thickness. After multiplying Eq. (22) by $(y-\theta)^{-0.5}$ and integrating it over θ from 0 to y one has

$$\xi=\int_0^y \frac{[H-\exp(-\xi)]}{\sqrt{y-\theta}}\,d\theta. \tag{23}$$

Thus, the layer thickness grows when $H>1$ and the ξ value can be estimated as follows:

$$2(H-1)\sqrt{y}<\xi(y)<2H\sqrt{y}. \tag{24}$$

When $H \gg 1$, the size of function $\xi(y)$ follows the well-known square-root dependence.[19] The results of the numerical analysis of Eq. (22) are shown in Fig. 3.

As follows from the formal solution of Eq. (23), $\xi \to -\infty$ at the finite time y^*. For y close to y^* we search for the solution of Eq. (23) in the form

$$\exp(-\xi)=\frac{1}{(y^*-y)^{\omega}}\sum_{j=0}^{\infty} c_j(y^*-y)^j. \tag{25}$$

To obtain the logarithmic divergence of the integral in the right-hand side of Eq. (23) (when $y \to y^*$) one should take $\omega=0.5$. Restricting ourselves to the first term in the sum (25) we have

$$c_0=\frac{1}{2}, \quad 2\ln 2=2H\sqrt{y^*}-\ln\sqrt{y^*}. \tag{26}$$

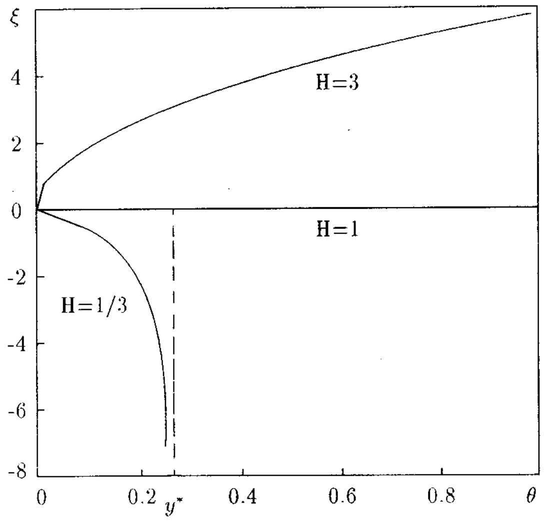

FIG. 3. The results of numerical simulation of Eq. (22). Time dependence of the dimensionless layer thickness ξ. The case $H=1$ corresponds to the critical nucleus.

These arguments are proved by the results of the numerical solution of Eq. (22) for $H<1$. It is obvious that the solution makes sense only for $\xi>-4a$ (the ξ value $\xi \sim -4a$ corresponds to the complete dissolution of the nucleus). Nevertheless, it is possible to use the expression

$$T=\frac{\pi y^*}{2^8}\exp(8a_0) \tag{27}$$

to estimate the dissolution time for $a \gg 1$. Thus, the dissolution time depends exponentially on the initial layer thickness. The dynamic behavior discussed above is peculiar to infinite systems. For a finite-size system the layer dynamics depends essentially on the boundary conditions. For the particular case of the flux absence at the sample edges the stationary distribution of the diffusion field establishes at a characteristic time scale $t \sim L^2$ and the layer size will stop changing.

IV. DECOMPOSITION IN MULTILAYER STRUCTURES

Let us discuss an evolution of the initial concentration distribution, which corresponds to a multilayered system:

$$c(z,t=0)=\frac{(-1)^N+1}{2}+\sum_{n=1}^{N}(-1)^n\tanh[z-z_n(t=0)],$$

N is the number of phase boundaries in a multilayer structure. Instead of Eq. (19) we take the second pair of boundary conditions in the form:

$$\left.\frac{\partial c}{\partial z}\right|_{0,L}=0, \tag{28}$$

which corresponds to the absence of the surface segregation effect.[18] To obtain the equations of motion we should first solve the diffusion equation (12). At the stage $t \gg d_m^2$ we may neglect the time derivative in the left-hand side of this equation if the alternating function $y(z,t)$ averaged over the interval $\Delta z \sim \sqrt{t}$ vanishes. The solution of the resulting equations for V_n given below [as well as the direct numerical solution of Eq. (4)] shows that the latter condition is fulfilled for finite-size systems. The solution of the stationary equation (12) is

$$\psi=-\frac{1}{4}\sum_{n=1}^{N}(-1)^n V_n \ln\cosh[z-z_n(t)]+Az+B, \tag{29}$$

$$A=\pm\frac{1}{4}\sum_{n=1}^{N}(-1)^n V_n=0. \tag{30}$$

Note that the condition (30) corresponds to the conservation of the average concentration. From Eqs. (17) and (29) one has the system of $N+1$ equations for z_m $(m=1\text{–}N)$ and B:

$$\begin{gathered}
\sum_{n=1}^{N}(-1)^n V_n\frac{z_m-z_n}{\tanh(z_m-z_n)}+(-1)^m 16[\exp(-2l_{m+1})\\
-\exp(-2l_m)]+B=0,\\
m=2,3\ldots N-1,\\
\sum_{n=1}^{N}(-1)^n V_n\frac{z_1-z_n}{\tanh(z_1-z_n)}-16[\exp(-2l_2)]+B=0,\\
\sum_{n=1}^{N}(-1)^n V_n\frac{z_N-z_n}{\tanh(z_N-z_n)}-(-1)^N 16\\
\times[\exp(-2l_N)]+B=0,\\
\sum_{n=1}^{N}(-1)^n V_n=0,
\end{gathered} \tag{31}$$

where $l_m=z_m-z_{m-1}$ is the thickness of the layer with the number m. As follows from the system (31), the fronts velocities are small if the layer thickness is large compared to the interfacial thickness. The assumption of $u(z,t)$ smallness is also valid under this condition. Deriving the system (31) we ignored the terms of the order of $\exp[-2(l_m+l_{m+1})]$. Nevertheless, the m front migration depends on the coordinates of the $(m-2)$th and the $(m+2)$th fronts. This results from the interfacial interaction through the diffusion field of the dissolved atoms. This type of interaction dominates in 2D and 3D structures and can be described within the framework of the Lifshitz-Slyosov model.[10] Note that the system (31) differs essentially from the one obtained previously within the Landau model with the nonconserved order-parameter field (see Ref. 12 and references therein).

Let us investigate the stability of a periodic multilayered system with a period $2d$. We will search for the solution of the system (31) in the form

$$z_n(t)=nd+\delta_n(t), \quad |\delta_n|\ll 1. \tag{32}$$

For δ_n one has

$$\begin{gathered}
\frac{d\delta_1}{dt}=\frac{8\exp(-2d)}{d}(1-2\delta_3+2\delta_2),\\
\frac{d\delta_2}{dt}=\frac{8\exp(-2d)}{d}(1+4\delta_2-2\delta_1-2\delta_4),\\
\frac{d\delta_n}{dt}=-\frac{16\exp(-2d)}{d}(\delta_{n+2}-2\delta_n+\delta_{n-2}),\\
\frac{d\delta_{N-1}}{dt}=\frac{8\exp(-2d)}{d}(-1-4\delta_{N-1}+2\delta_N+2\delta_{N-3}),\\
\frac{d\delta_N}{dt}=\frac{8\exp(-2d)}{d}(-1+2\delta_{N-2}-2\delta_{N-1}).
\end{gathered} \tag{33}$$

As follows from Eq. (33), an infinite multilayered system is absolutely unstable to the fluctuations of the layer thickness. The wave-number dependence of the fluctuation increment

$$\lambda(k)=\frac{64\exp(-2d)}{d}\sin^2(kd), \quad 0\leqslant k\leqslant\frac{\pi}{d} \tag{34}$$

has a maximum at $k=\pi/2d$, which results in the period doubling. The expression for the increment (34) coincides with that obtained in Ref. 11.

The system (31) provides us with a useful tool for the analysis of the interface dynamics in various multilayer systems for arbitrary initial layer thicknesses. We restrict ourselves to the structures consisting of films of the identical initial thicknesses d, which are of particular interest in the context of applications to the study of thermostability of real multilayers. In finite-size systems the layer thickness changes may be caused by the boundary effects. We have studied the influence of these effects on the doubling process in multilayer structures for various d values and layer number N using both equations (31) and the direct numerical simulation of Eq. (5).

To solve Eq. (5) numerically let us write the discrete analog of the equation on the grid $z_i=(i+1/2)l$, $i=0,1,\ldots,n-1;l=L/n$ for the vector function $\mathbf{c}(t)=\{c_i(t)\}_{i=0}^{n-1},c_i(t)=c(z_i,t)$:

$$\frac{\partial\mathbf{c}}{\partial t}=-\hat{P}\mathbf{c}+\hat{Q}(\mathbf{c}). \tag{35}$$

Here $\hat{P}$ and $\hat{Q}$ denote the following difference operators:

$$\begin{gathered}
\hat{P}=\hat{D}^2+2\hat{D},\\
(\hat{D}\mathbf{c})_i=(c_{i-1}-2c_i+c_{i+1})/l^2, \quad i=0,1,\ldots,n-1,\\
(\hat{Q}(\mathbf{c}))_i=2(c_{i-1}^3-2c_i^3+c_{i+1}^3)/l^2,\\
i=0,1,\ldots,n-1.
\end{gathered}$$

The values $c_{-2},c_{-1},c_n,c_{n+1}$ are determined by the boundary conditions (6),(28):

$$c_{-2}=c_1,c_{-1}=c_0,c_n=c_{n-1},c_{n+1}=c_{n-2}. \tag{36}$$

As we will see below, one needs to solve the problems (35), (36) for a long-time interval. From this point of view the applied numerical method has to be stable. The known explicit Euler scheme is stable only for a very small time step, and application of the stable implicit schemes are hampered due to high order of the spatial derivatives in Eq. (35).

So we employ the following scheme. First, we use a split-step method[20] separating linear and nonlinear parts of the equation:

$$\mathbf{c}(t+\Delta t)=\exp(-\Delta t\hat{P})[\mathbf{c}(t)+\Delta t\hat{Q}(\mathbf{c})]+O(\Delta t^2). \qquad (37)$$

This scheme includes the explicit Euler step for the nonlinear operator $\hat{Q}$. However, it may be shown that a stability domain of scheme (37) is wider than the stability domain of the explicit Euler method for equation (35).

To calculate $\exp(-\Delta t\hat{P})$ in Eq. (37) we express the operator $\hat{P}$ as a sum of two operators:[21]

$$\hat{P}=\hat{P}_0+\hat{G}, \quad \hat{G}=\hat{P}-\hat{P}_0,$$

where the operator $\hat{P}_0$ is similar to $\hat{P}$ but corresponds to the periodic boundary conditions, and the operator $\hat{G}$ has the form:

$$(\hat{G}\mathbf{c})_i=\begin{cases}\tau c_0-c_1+c_{n-2}-\tau c_{n-1}, & i=0\\ -c_0+c_{n-1}, & i=1\\ 0, i=2,3,\dots, & n-3\\ c_0-c_{n-1}, & i=n-2\\ -\tau c_0+c_1-c_{n-2}+\tau c_{n-1}, & i=n-1\end{cases},$$

where $\tau=2(2-l^2)/l^4$.

Using the Trotter-Kato formula for the exponential function of the sum of two noncommutative operators[22] in Eq. (37) we obtain

$$\mathbf{c}(t+\Delta t)=\exp(-\Delta t\hat{P}_0)\exp(-\Delta t\hat{G})[\mathbf{c}(t)+\Delta t\hat{Q}(\mathbf{c})] +O(\Delta t^2).$$

The exponential function $\exp(-\Delta t\hat{P})$ is easy to find by the discrete Fourier transformation, while the operator $\exp(-\Delta t\hat{G})$ may be calculated analytically. This scheme allows us to obtain a solution with a good accuracy for a long-time interval of interest.

Let us start with the simple case $N=3$, when the system (31) can be solved analytically. It follows from the symmetry arguments that

$$V_2=V_1+V_3=0, \qquad (38)$$

and the system (31) reduces to the equation

$$-\dot{l}(2l-1)=16e^{-2l}, \qquad (39)$$

where $l=z_3-z_2=z_2-z_1$. The time dependence of the front coordinates in this case is shown in Fig. 4. The thicknesses of the outer layers are growing, while the thicknesses of the inner layers are decreasing. From now on we introduce the notation T for the characteristic dissolution time (see Fig. 4) determined by the condition $l(T)=0$. The $T(d)$ dependence takes the form

$$T=\frac{d-1}{16}\exp(2d). \qquad (40)$$

This analytical dependence agrees well with the one obtained from the numerical solution of Eq. (5) (see Fig. 5).

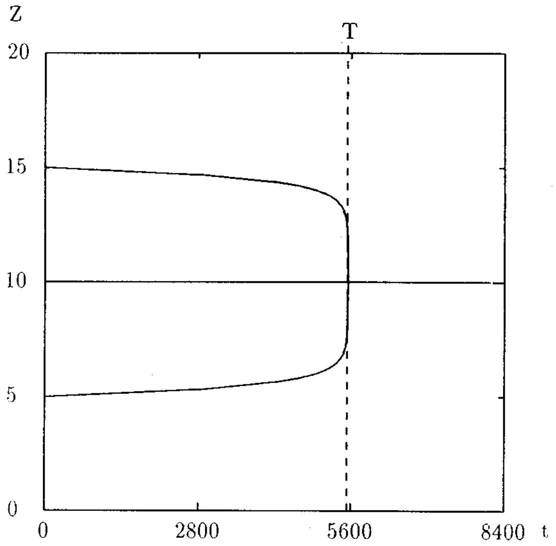

FIG. 4. Time dependence of the coordinates of the phase boundaries ($N=3$, $d=5$).

We have carried out the analogous calculations for $N=5-15$ and $d=3-6$. For the particular case $N=15$ and $d=4$ the time dependence of the interface coordinates is shown in Fig. 6. The doubling process consists of two stages. At the first one ($t<T$) the dissolution of the 3rd and 14th films occurs. At the end of the second stage ($t\sim 2T$) we obtain the structure with film thicknesses $\sim 2d$. The duration T of the first stage appears to depend exponentially on d (see Fig. 7). We have also obtained the dependence $T(N)$ (Fig. 8) which proves the essential influence of boundaries on the doubling scenario discussed above. The quantitative agreement between the analytical and numerical results confirms the validity of the solitonlike description developed in this paper.

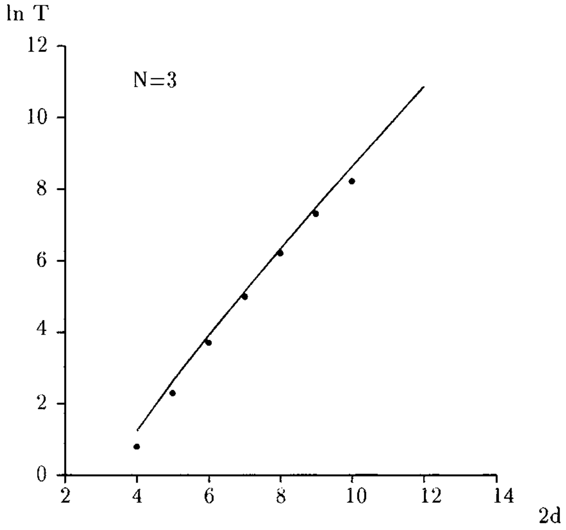

FIG. 5. Dependence of the dissolution time T on the initial film thickness: solid line indicates the calculation based on Eq. (40), ● indicates the results of the numerical simulation of Eq. (5).

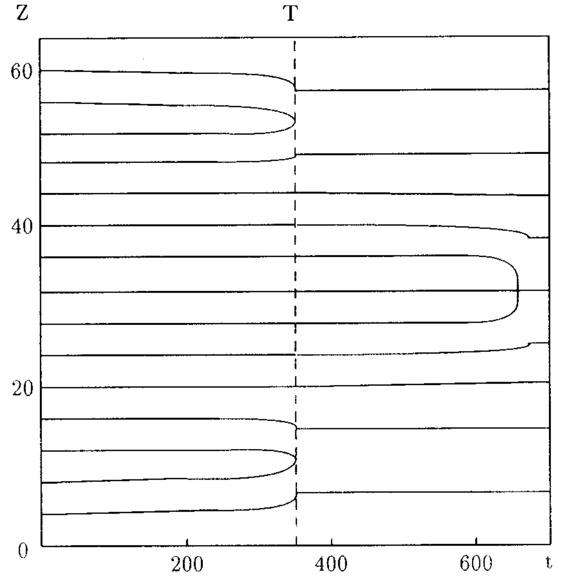

FIG. 6. Time dependence of the coordinates of the phase boundaries ($N=15$, $d=4$).

V. CONCLUSIONS

To summarize, we have investigated the nonlinear relaxation dynamics of layered decomposing alloys. The results obtained above are valid for systems with layer thicknesses much greater than the interfacial thickness. The approximate solution of the Cahn-Hilliard equation is based on the assumption about a slight change of the form of moving interfaces. We have studied the features in the dynamic behavior, peculiar for 1D systems. The equation of the interface motion, which describes the growth or dissolution of a single layer has been obtained. This equation appears to be essentially nonlocal in time. Within this approach we estimated the layer dissolution time and obtained the velocity of a new phase layer growth. In the limit $a_0 \gg a_c$ our results are in good agreement with the ones obtained in Ref. 19. The dissolution time grows exponentially with an increase in the initial layer thickness. We have obtained the equations for interface velocities (31), which describe the decomposition process in a wide class of multilayers and provide a starting point for the analysis of thermostability of artificial layered structures. The diffusion relaxation in these systems occurs via the period doubling. The concrete scenario of this process depends essentially on total number of the layers in the system. The velocity of the analyzed processes decreases exponentially with a growing layer thickness. Nevertheless, this velocity is noticeable for ultrathin (~ 1 nm) layers, which may provide a possibility of the experimental observation of the discussed effects.

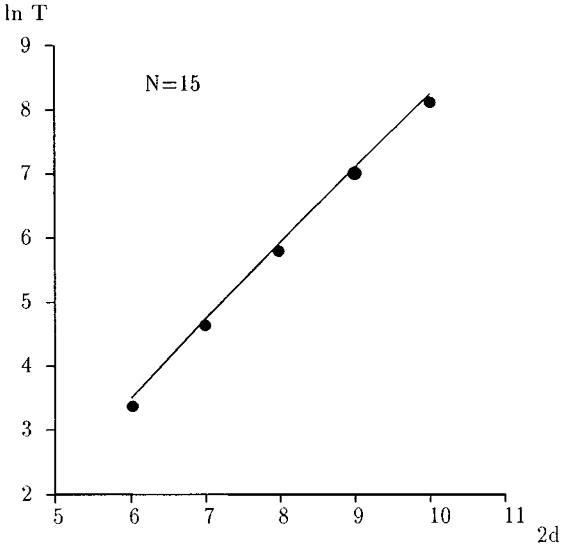

FIG. 7. Dependence of the dissolution time T on the initial film thickness: solid line represents the results of the solution of the system (31), ● shows the results of the numerical simulation of Eq. (5).

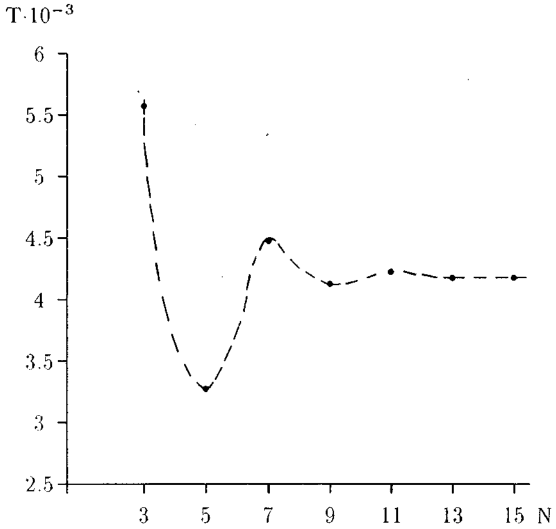

FIG. 8. Dependence of the dissolution time T on the total number N of interfaces in the structure.

ACKNOWLEDGMENTS

We thank Professor K.A. Gorshkov for some valuable comments. This work was supported by the Russian Foundation for Fundamental Research (Grants No. 95-02-05388a, 95-01-00573a, 95-02-04994a). A.S.M. acknowledges the financial support of the International Center for Fundamental Physics in Moscow (INTAS Grant No. 93-2492-ext).

APPENDIX

Let us briefly discuss the 1D stationary solutions of Eq. (4). Taking into account condition (2) one obtains

$$\frac{\partial^2 c}{\partial z^2}+2(c-c^3)=h. \tag{A1}$$

The interfacial thickness (the width of the transition layer between the phases) is of the order of unity in dimensionless variables. The first integral of Eq. (A1) is

$$\frac{1}{2}\left(\frac{\partial c}{\partial z}\right)^2+U(c)=I,$$

$$U(c)=c^2-\frac{1}{2}c^4-hc,\quad h>0, \tag{A2}$$

The form of the stationary solutions depends on the I quantity. In the case $h=0$ there exists an equilibrium state ($I=I_1$) which corresponds to the decomposition[23] [see Fig. 2(a)]. The concentration distribution like this is usually called a ''kink'' (''antikink''). For $h\neq 0$ there is a solution with $I=I^*$, which corresponds to a critical new-phase nucleus [see Fig. 2(c)]. This solution may be considered as two interacting kinks with the asymptotic behavior $c(z)\to c_0$ when $z\to\pm\infty$. The case $I=I_2$ corresponds to the periodic concentration distribution, i.e., to the set of kinks and antikinks [see Fig. 2(b)]. Let us find the size of the critical new-phase nucleus. Equation (A2) with $I=U(c_0)$ results in the following expression:

$$c=c_0+\frac{1}{2}\sqrt{(c_0-c_1)(c_0-c_2)}\left[\tanh\left(\frac{x-2a}{2}\right)\right.$$
$$\left.-\tanh\left(\frac{x+2a}{2}\right)\right],$$

$$x=z\sqrt{(c_0-c_1)(c_0-c_2)},$$

$$\cosh(2a)=\frac{2c_0-(c_1+c_2)}{c_1-c_2}, \tag{A3}$$

where $c_0>c_1>c_2$ are the solutions of the equation $U(c)=U(c_0)$. When $h\ll 1$ (i.e., the supersaturation degree is small) one has

$$c=1-\frac{h}{4}+\tanh(z-a)-\tanh(z+a),\quad a=\frac{1}{4}\ln\frac{16}{h},$$

$$c_0=1-\frac{h}{4},c_{1,2}=-1\pm\sqrt{h}. \tag{A4}$$

Thus we obtained the logarithmic dependence of the nucleus size on the supersaturation degree. Note that in 2D and 3D cases such a dependence is inversely proportional (the critical nucleus exists due to different dependences of the surface and bulk energy on the nucleus size[24,25]).

---

*Electronic address: andr@ipm.sci.nnov.ru

[1] H. Furukawa, Adv. Phys. **34**, 703 (1985).

[2] V.P. Skripov and A.V. Skripov, Sov. Phys. Usp. **22**, 389 (1979).

[3] M.N. Baibich, J.M. Broto, A. Fert, F. Nguyenn van Dan, F. Petroff, P. Etienne, G. Greuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[4] S.S.P. Parkin, S.J. Li, and D.J. Smith, Appl. Phys. Lett. **58**, 2710 (1991).

[5] A.E. Berkowitz, J.R. Mitchell, M.J. Careu, A.P. Yang, S. Zhang, F.E. Spada, F.T. Parker, A. Hutten, and J. Tomas, Phys. Rev. Lett. **68**, 3745 (1992).

[6] A.D. Akhsakhalyan, A.A. Fraerman, Yu.Ya. Platonov, N.I. Polushkin, and N.N. Salaschenko, Thin Solid Films **203**, 317 (1991).

[7] Y. Blyakhman, A.D. Akhsakhalyan, N.I. Polushkin, N.N. Salaschenko, S.A. Gusev, and V.G. Semenov, Phys. Rev. B **52**, 10 303 (1995).

[8] J.W. Cahn and J.E. Hilliard, J. Chem. Phys. **28**, 258 (1958).

[9] *Dynamics of Ordering Processes in Condensed Matter*, edited by S. Komura and H. Furukawa (Plenum, New York, 1988).

[10] I.M. Lifshitz and V.V. Slyozov, J. Phys. Chem. Solids **19**, 35 (1961).

[11] J.S. Langer, Ann. Phys. **65**, 53 (1971).

[12] S.N. Majumdar and D.A. Huse, Phys. Rev. E **52**, 270 (1995).

[13] J.S. Langer and R.F. Sekerka, Acta Metall. **23**, 1225 (1975).

[14] Y.S. Kivshar and B.A. Malomed, Rev. Mod. Phys. **61**, 763 (1989).

[15] Yu. N. Devyatko, S.V. Rogozhkin, and B.A. Fedotov, Phys. Rev. B **50**, 5857 (1994).

[16] A.Z. Patashinsky and B.I. Shumilo, Sov. Phys. JETP **50**, 712 (1979).

[17] R.L. Pego, Proc. R. Soc. London Ser. A **422**, 241 (1989).

[18] S. Puri and K. Binder, Phys. Rev. E **49**, 5359 (1994).

[19] J.W. Christian, *The Theory of Transitions in Metals and Alloys* (Pergamon, New York, 1975), Pt. 1.

[20] R.H. Hardin and F.D. Tappert, SIAM Rev. **15**, 423 (1973).

[21] I.M. Nefedov and I.A. Shereshevskii, Mat. Model. **7**, 88 (1995).

[22] T. Kato, *Perturbation Theory for Linear Operators* (Springer-Verlag, Berlin, 1966).

[23] A.G. Khachaturyan and R.A. Suris, Sov. Phys. Crystallogr. **13**, 63 (1968).

[24] Ya.B. Zeldovich, Zh. Eksp. Teor. Fiz. **12**, 525 (1942).

[25] K. Binder and D. Stauffer, Adv. Phys. **25**, 343 (1976).