Disorder trapping by rapidly moving phase interface in an undercooled liquid

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Abstract. Non-equilibrium phenomena such as the disappearance of solute drag, the origin of solute trapping and evolution of disorder trapping occur during fast transformations with originating metastable phases [D.M. Herlach, P.K. Galenko, D. Holland-Moritz, *Metastable solids from undrrercooled melts* (Elsevier, Amsterdam, 2007)]. In the present work, a theoretical investigation of disorder trapping by a rapidly moving phase interface is presented. Using a model of fast phase transformations, a system of governing equations for the diffusion of atoms, and the evolution of both long-range order parameter and phase field variable is formulated. First numerical solutions are carried out for a congruently melting binary alloy system.

1 Introduction

Disorder trapping is a known non-equilibrium phenomenon occurring during transformations of congruently melting ordered intermetallic compounds [1, 2]. This phenomenon occurs especially during rapid crystallization of undercooled melts of intermetallics with a superlattice crystalline structure [3–5]. In such systems, the phase interface propagation is sluggish at small undercoolings. The attachment of atoms from the liquid to the phase interface needs short-range atomic diffusion, since atoms have to move to the proper lattice site in the superlattice structure. If the undercooling increases, the non-equilibrium effect of disorder trapping leads to the formation of a metastable disordered structure. Experimental evidence of disorder trapping has been demonstrated by *in situ* diffraction studies using synchrotron radiation on levitation-processed samples, in which a transition from ordered to disordered growth at a critical undercooling was unambiguously shown [6]. The phenomenon of order–disorder transition with distinct change of the order parameter is also important in structural transformations with non-monotonic relaxation processes of melts [8] and in liquid–liquid phase separation of undercooled metallic alloys [9, 10].

The disorder trapping was investigated from theoretic perspective by sharp interface models [2, 3, 6], a diffuse interface model [11] and methods of atomistic simulation [1, 12]. In the present work, using an existing approach for fast phase transformations [13], a diffuse

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interface model results in a system of hyperbolic equations to describe (i) the rapid interface motion and (ii) the transition from ordered to disordered structures formed from undercooled liquids. The pioneering atomistic model of Chernov [1] and thermodynamic model of Boettinger and Aziz [2] consider a process of ordering at the interface only. The present diffuse interface model introduces spatial variation of order parameter such that the order-disorder transition and relaxation of order parameter exist in the diffuse zone between phases as well as in a solid phase far behind the solid-liquid interface. In addition to previous works [1–3, 6, 11], the present model takes into account highly rapid interface propagation at which locally non-equilibrium states in chemical composition and in atomic ordering are freezing in solid phase with an appearance of metastable states.

In the numerical solutions of the model, we test the existence of pronounced disorder trapping by direct exchange of dissimilar atoms between sublattices at high interface velocities moving in an undercooled binary liquid. The main focus of the present work is to analyze numeric results and to formulate conditions for disorder trapping in rapidly transforming congruently melting alloy undercooled in the liquid state.

2 Definitions

2.1 Main functions

We consider a binary mixture consisting of A-atoms and B-atoms. For a given temperature T, the main functions of the model, which should be obtained by solutions, are defined as follows. *The first function* is φ the phase field with $\varphi = 1$ in the S-phase (solid) and $\varphi = 0$ in the L-phase (liquid). *The second function* is x_B the overall continuous concentration of B-atoms (which can be defined through the L-phase and S-phase concentrations). Concentrations in the S-phase are given by $x_i^j = n_i^j/(n_A^j + n_B^j)$, where n_i^j is the number of moles of atom i (i=A,B) on the sublattice j ($j = \alpha, \beta$). Finally, *the third function* is the long-range order parameter η, which is defined by the concentrations $x_i^j = (x_A^\alpha, x_A^\beta, x_A^\alpha, x_B^\beta)$ of the $\alpha-$ and $\beta-$sublattices as

$$\eta = x_A^\alpha - x_A^\beta = x_B^\beta - x_B^\alpha. \tag{1}$$

A completely disordered state in the S-phase occurs with $\eta = 0$. Atoms exhibit full order on sublattices if $\eta = 1$. The concentrations in the S-phase on sublattices are

$$x_A^\alpha = 1 - x_B - \frac{\eta}{2}, \quad x_B^\alpha = x_B - \frac{\eta}{2}, \tag{2}$$

$$x_A^\beta = 1 - x_B + \frac{\eta}{2}, \quad x_B^\beta = x_B + \frac{\eta}{2}. \tag{3}$$

2.2 Gibbs free energy

2.2.1 The entire system

The Gibbs free energy for the entire binary system is given by

$$G(\varphi, \nabla\varphi, \eta, \nabla\eta, x_B) = G_S(x_B, \eta)p(\varphi) + G_L(x_B)p(1 - \varphi)$$
$$+ \frac{\varepsilon_\varphi}{2}(\nabla\varphi)^2 + \frac{\varepsilon_\eta}{2}(\nabla\eta)^2 + W_\varphi g_\varphi(\varphi) + W_\eta g_\eta(\eta). \tag{4}$$

The double-well functions in Eq. (4) for the phase field g_φ and for the long-range order parameter g_η are given by

$$g_\varphi(\varphi) = \varphi^2(1 - \varphi)^2, \quad g_\eta(\eta) = \eta^2(1 - \eta)^2. \tag{5}$$

The Gibbs free energy is interpolated between the phases using the following function

$$p(\varphi) = \varphi^2(3 - 2\varphi). \tag{6}$$

2.2.2 The phases

In the *L*-phase we assume the following approximation

$$G_L(T, x_B) = (1 - x_B)G_L^A(T) + x_B G_L^B(T), \tag{7}$$

$$G_L^A(T, x_B) = G_L^{0A}(T) + RT\ln(1 - x_B) + x_B\Omega_L, \tag{8}$$

$$G_L^B(T, x_B) = G_L^{0B}(T) + RT\ln(x_B) + (1 - x_B)\Omega_L, \tag{9}$$

which can be considered as contribution (7) from elements *A* and *B*, reference terms G_L^{0i}, ideal mixture terms $RT\ln x_i$, and excess terms $x_i\Omega_L$ (where again $i = A, B$).

In the *S*-phase, the Gibbs free energy has additional terms related to the disorder parameter

$$G_S(T, x_B, \eta) = G_S^{ref}(T, x_B) + G_S^{id}(T, x_B, \eta) + G_S^{ex}(x_B, \eta). \tag{10}$$

Assuming that the *L*-phase transforms into the body centered cubic modification of the *S*-phase, the following contributions into the free energy (10) are taken into account:
- reference contribution

$$G_S^{ref}(T, x_B) = (1 - x_B)G_A^{bcc}(T) + x_B G_B^{bcc}(T), \tag{11}$$

- ideal mixture contribution

$$G_S^{id}(T, x_B, \eta) = \frac{1}{2}RT\Bigg((x_B - \frac{\eta}{2})\ln(x_B - \frac{\eta}{2}) + (x_B + \frac{\eta}{2})\ln(x_B + \frac{\eta}{2})$$
$$+ (1 - x_B - \frac{\eta}{2})\ln(1 - x_B - \frac{\eta}{2}) + (1 - x_B + \frac{\eta}{2})\ln(1 - x_B + \frac{\eta}{2})\Bigg), \tag{12}$$

- excess free energy

$$G_S^{ex}(x_B, \eta) = \Omega_2\left(x_B(1 - x_B) + \frac{\eta^2}{4}\right) + \Omega_3\eta^3 + \Omega_4\eta^4. \tag{13}$$

3 Governing equations

Following the model of fast phase transformations [13], one can derive governing equations represented by hyperbolic equations for non-conserved and conserved field variables. They were obtained by the condition of non-decreasing entropy in time that, for isothermal systems, is equivalent to the condition of non-increasing Helmholtz free energy in time [14]. Following these approaches [13, 14], one can derive the system of governing equations using the Gibbs free energies from Section 2.2 as thermodynamic potentials accessible from thermodynamic databases. Then, one can find governing equations for
- the phase field

$$\tau_\varphi\frac{\partial^2\varphi}{\partial t^2} + \frac{\partial\varphi}{\partial t} = -M_\varphi\frac{\delta G}{\delta\varphi}, \tag{14}$$

- the long-range order parameter

$$\tau_\eta\frac{\partial^2\eta}{\partial t^2} + \frac{\partial\eta}{\partial t} = -M_\eta\frac{\delta G}{\delta\eta}, \tag{15}$$

- the concentration and chemical potential

$$\tau_D \frac{\partial^2 x_B}{\partial t^2} + \frac{\partial x_B}{\partial t} = \nabla \cdot (M_x \nabla \mu_B), \quad \mu_B = \frac{\delta G}{\delta x_B}. \tag{16}$$

The hyperbolic equations (14) and (15) are damped wave equations, which extend known parabolic equation for the non-conserved order parameter derived first by Mandel'shtam–Leontovich [15, 16] and known in literature as the time–dependent Ginzburg-Landau equation [16, 17] or Allen-Cahn equation [18]. The hyperbolic equation (16) generalizes the known parabolic equation for the conserved order parameter derived and analyzed first by Khachaturyan [19] and Cahn and Hilliard [20]. Equations (14)-(16) were applied to non-monotonic relaxation [7, 8], phase segregation [9] and were analyzed in the context of fast dynamics during transitions from un(meta)stable to (meta)stable states [21]. The validity of hyperbolic type models in fast phase transition theory has been verified by comparison with experimental data [22], in molecular dynamics simulations of the solute trapping effect by rapidly moving interfaces [23] and by coarse graining derivations of phase field equations [24].

3.1 Phase-field

In its explicit form, the governing equation for the phase field comes from Eqs. (4)-(14). As a result one finds

$$\begin{aligned}
\frac{\tau_\varphi}{M_\varphi} \frac{\partial^2 \varphi}{\partial t^2} + \frac{1}{M_\varphi} \frac{\partial \varphi}{\partial t} &= \varepsilon_\varphi \nabla^2 \varphi - W_\varphi \frac{dg_\varphi(\varphi)}{d\varphi} - \Bigg[(1 - x_B) G_A^{bcc} + x G_B^{bcc} \\
&\quad + \frac{RT}{2} \Big((x_B - \eta/2) \ln(x_B - \eta/2) + (x_B + \eta/2) \ln(x_B + \eta/2) \\
&\quad + (1 - x_B - \eta/2) \ln(1 - x_B - \eta/2) + (1 - x_B + \eta/2) \ln(1 - x_B + \eta/2) \Big) \\
&\quad + \Omega_2(x_B(1 - x_B) + (1/4)\eta^2) + \Omega_3 \eta^3 + \Omega_4 \eta^4 \Bigg] \frac{dp(\varphi)}{d\varphi} \\
&\quad - \Big((1 - x_B)\big(G_L^{0A} + RT \ln(1 - x_B) + x_B \Omega_L\big) + x_B\big(G_L^{0B} + RT \ln(x_B) + (1 - x_B)\Omega_L\big) \Big) \frac{dp(\varphi)}{d\varphi}
\end{aligned} \tag{17}$$

Without the ordering process, i.e. if $\eta = 0$, Eq. (17) transforms into the one derived in Refs. [13, 14].

3.2 Order parameter

Atomic ordering in α- and β-sublattices is described by Eq. (15) together with the free energy (4)-(13) by the following equation:

$$\begin{aligned}
\frac{\tau_\eta}{M_\eta} \frac{\partial^2 \eta}{\partial t^2} + \frac{1}{M_\eta} \frac{\partial \eta}{\partial t} &= \varepsilon_\eta \nabla^2 \eta - W_\eta \frac{dg_\eta(\eta)}{d\eta} - \Bigg[\frac{RT}{2} \Big(-\frac{1}{2} \ln(x_B - \eta/2) + \frac{1}{2} \ln(x_B + \eta/2) \\
&\quad - \frac{1}{2} \ln(1 - x_B - \eta/2) + \frac{1}{2} \ln(1 - x_B + \eta/2) \Big) \\
&\quad + \frac{1}{2} \Omega_2 \eta + 3\Omega_3 \eta^2 + 4\Omega_4 \eta^3 \Bigg] p(\varphi)
\end{aligned} \tag{18}$$

From Eq. (18) it follows that the process of atomic ordering is coupled with the phase field by the interpolation function $p(\varphi)$.

3.3 Chemical diffusion

The chemical potential from Eq. (16) is given by the variational derivative

$$\mu_B = \frac{\delta G}{\delta x_B} =$$

$$\left[G_B^{bcc} - G_A^{bcc} + \frac{RT}{2}\left(\ln(x_B - \frac{\eta}{2}) + \ln(x_B + \frac{\eta}{2}) - \ln(1 - x_B - \frac{\eta}{2}) - \ln(1 - x_B + \frac{\eta}{2}) \right) + \Omega_2(1 - 2x_B) \right] p(\varphi)$$

$$+ \left[G_L^{0B} - G_L^{0A} + RT\{\ln(x_B) - \ln(1 - x_B)\} + 2(1 - 2x_B)\Omega_L \right] p(1 - \varphi). \quad (19)$$

The mobility M_x in the diffusion equation Eq. (16) is an interpolation between the bulk mobilities in the liquid M_L and in the solid M_S:

$$M_x = M_L\left(1 - p(\varphi)\right) + M_S(\eta)p(\varphi) = M_L\left(1 - p(\varphi)\right) + \left(M_S^{disorder}(1 - \eta) + M_S^{order}\eta \right) p(\varphi). \quad (20)$$

As follows from Eq. (20), the bulk mobility $M_S(\eta)$ in the solid is interpolated by atomic mobilities M_S^{order} and $M_S^{disorder}$ in the ordered state and disordered state, respectively.

Table 1. Physical parameters of the $A_{50}B_{50}$ alloy used for phase field modeling

Parameter	Value	Reference
Concentration of A(B), x_B	0.5 mole fraction	present work
η-rate relaxation time, τ_η	$4 \cdot 10^{-8}$ s	present work
φ-rate relaxation time, τ_φ	$4 \cdot 10^{-11}$ s	present work
x_B-rate relaxation time, τ_D	$4 \cdot 10^{-10}$ s	present work
Mobility of the η-field, M_η	$8 \cdot 10^4\ mole \cdot J^{-1} \cdot s^{-1}$	present work
Mobility of the φ-field, M_φ	$4 \cdot 10^5\ mole \cdot J^{-1} \cdot s^{-1}$	present work
Mobility of B-atoms in the L-phase, M_L	$2 \cdot 10^{-8}\ mole \cdot m^2 \cdot J^{-1} \cdot s^{-1}$	present work
Mobility of B-atoms in the disordered state, $M_S^{disorder}$	$7 \cdot 10^{-11}\ mole \cdot m^2 \cdot J^{-1} \cdot s^{-1}$	present work
Mobility of B-atoms in the ordered state, M_S^{order}	$9 \cdot 10^{-12}\ mole \cdot m^2 \cdot J^{-1} \cdot s^{-1}$	present work
Gradient factor for the η-field, ε_η	$1.4 \cdot 10^{-13}\ J \cdot m^2 \cdot mole^{-1}$	present work
Gradient factor for the φ-field, ε_φ	$5.5 \cdot 10^{-12}\ J \cdot m^2 \cdot mole^{-1}$	present work
Energy barrier between states in the η-field, W_η	$2.5 \cdot 10^3\ J \cdot mole^{-1}$	present work
Energy barrier between states in the φ-phase, W_φ	$4.5 \cdot 10^3\ J \cdot mole^{-1}$	present work
1st Thermodynamic parameter, Ω_2	$-2 \cdot 10^3 R$ J/mole	[2]
2nd Thermodynamic parameter, Ω_3	$-1.225 \cdot 10^6 R$ J/mole	present work
3rd Thermodynamic parameter, Ω_4	$-5.662 \cdot 10^2 R$ J/mole	[2]
4th Thermodynamic parameter, Ω_L	$-1.72 \cdot 10^4$ J/mole	[2]

4 Material parameters and modeling

The spatially inhomogeneous evolution of ordered and disordered states has been modeled by the numerical solution of the equations for diffusion (16), (19) and (20), phase field motion (17) and ordering (18). Material parameters have been chosen for a binary congruently melting $A_{50}B_{50}$ alloy. Functions $G_L^{0A}(T)$, $G_L^{0A}(T)$, $G_A^{bcc}(T)$, and $G_B^{bcc}(T)$ from Eqs. (7)-(9) and
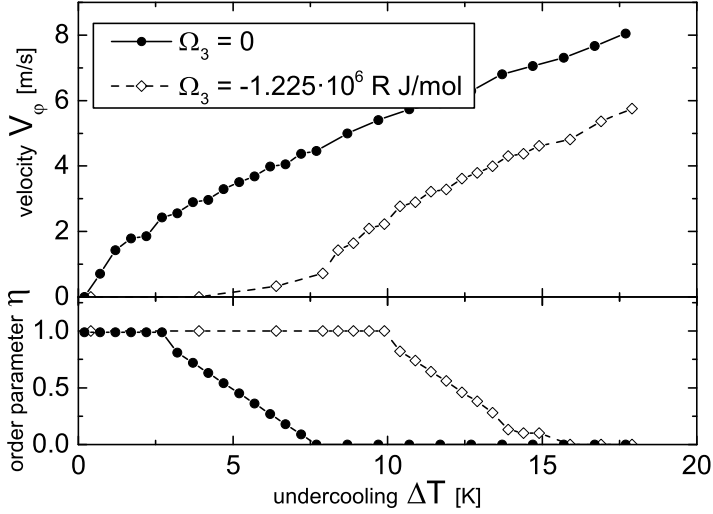
Figure 1. Behavior of the phase field interface velocity V_φ and long-range order parameter η as functions of liquid undercooling $\Delta T = T_L - T$, where T_L is the liquidus temperature (calculated as the temperature for "liquid–ordered phase" co-existence). Two cases show the change of the transformation kinetics depending on the thermodynamic parameter Ω_3 which regulates the barrier height for atomic diffusion jumps between sublattices α and β, see Eq. (13).

(11) are taken from the thermodynamical data base of elements (CALPHAD) for a $Ni_{50}Al_{50}$ alloy.

The numerical scheme for the solution of the model consists of a finite difference (FD) scheme of second order with explicit time stepping. The initial concentration distribution was taken as homogeneous, $x_B = 0.5$, while both phase field φ and order parameter η were initialized to unity in the solid and zero in the liquid, connected by a smooth transition at the interface, which was described by a Gaussian error function with a width of 1 micron, corresponding to 10 FD nodes. Boundary conditions were Dirichlet type conditions on the solidifying side and Neumann type (i.e., zero flux) conditions in the liquid far away from the interface.

Numerical solutions of Eqs. (16), (19), (20), 17, and (18) were achieved in a one-dimensional space. To obtain steady state results, average values of the interface velocity V_φ for the phase field and the long-range order parameter η where determined as follows. The phase-field has a step-like shape with $\varphi = 1$ in the solid and $\varphi = 0$ in the liquid. Because the solid-liquid interface is a diffuse interface, the conventional definition of the interface position is the point z_f where $\varphi(z_f) = 0.5$. In this work we adopt another definition of the interface position. $z_f = \int_{-\infty}^{+\infty} \varphi(z')dz'$ is exact for the case of a symmetric interface profile and allows us to find an exact interface velocity in case of a steady state profile. During the simulation run, we collect the values of the interface position z_f^n at times t^n after a fixed number (e.g. 2000) of time steps. The current interface velocity V_φ is obtained as $V_\varphi^n = (z_f^n - z_f^{n-1})/(t^n - t^{n-1})$. After the system reaches a steady state regime, we calculate *the steady state interface velocity* $V_\varphi^{(avg)}$ as the average of the last ten values V_φ^n. *The average value of the order parameter η in the solid phase right beyond the interface is determined as the average of 11 FD nodes*
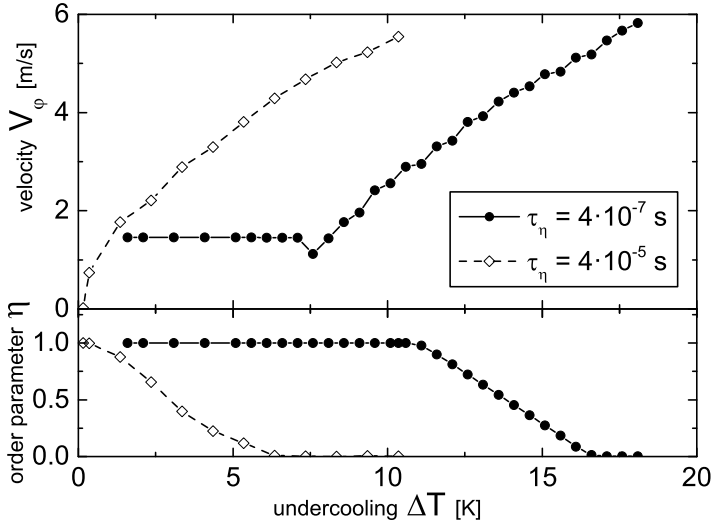
Figure 2. Behavior of the phase field interface velocity V_φ and long-range order parameter η as functions of liquid undercooling $\Delta T = T_L - T$, where T_L is the liquidus temperature (calculated as the temperature for "liquid–ordered phase" co-existence). Two cases show the change of the transformation kinetics depending of the time τ_η for relaxation of the ordering rate $\partial\eta/\partial t$ to change the regime from inertial (ballistic) to dissipative (diffusive) [21]. The η-gradient factor has been chosen as $\varepsilon_\eta = 1.4 \cdot 10^{-11} \; (J \cdot m^2 \cdot mole^{-1})$.

at a distance of 10 FD spacings behind the interface, i.e. the FD nodes 10 to 20 behind the calculated interface position.

5 Results

Figures 1 and 2 show the influence of various parameters for the ordering process on both the transition from L-phase to S-phase and on the ordering process itself. Namely, Fig. 1 presents the kinetics change due to different values of Ω_3. It can be seen that with a non-zero value of the barrier Ω_3 [see Eq. (13)] the kinetics of transformation qualitatively changes with a decrease in V_φ and a shift in the disorder transformation velocity V_{DT} to its large values. Figure 2 illustrates the influence of gradient factor and relaxation time τ_η on the transformation kinetics. The time τ_η characterizes the relaxation to local equilibrium in the η-field, i.e., the relaxation of the rate $\partial\eta/\partial t$. An increase in the relaxation time τ_η moves the initiation of disorder trapping to smaller undercoolings.

6 Kinetics and temporal conditions for trapping of disorder

Finally, on the basis of our numerical results (see Section 5), we formulate a condition of disorder trapping by a moving diffuse interface. During fast transformations the disorder trapping effect occurs when a particular atom has no time to find its equilibrium position within the diffuse interface and remains in a higher energetic position. Instead of a phase with complete atomic order, a disordered metastable phase is appearing behind the diffuse interface. This effect becomes important at phase interface velocities V_φ larger than a characteristic
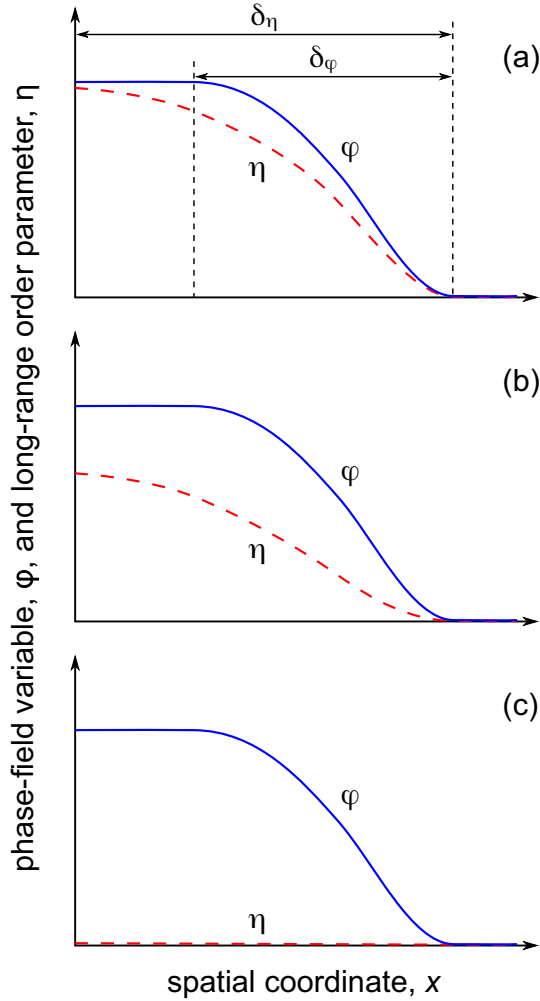
Figure 3. Profiles of the phase field ϕ and long-range order parameter η with their thickness δ_φ and δ_η, respectively. (a) Fully ordered state, $V_\varphi \ll V_{DT}$. (b) Partial disorder trapping, $V_\varphi \approx V_{DT}$. (c) Complete disorder trapping, $V_\varphi > V_{DT}$.

value of the order of $V_{DT} \propto D_\eta/\delta_\eta$ (with $D_\eta = M_\eta \varepsilon_\eta$ being the characteristic diffusion coefficient in solid phase for the ordering process, and δ_η the thickness of the interface for ordering). Indeed, when $V_\varphi > V_{DT}$, the characteristic time δ_φ/V_φ of the diffuse interface of thickness δ_φ becomes smaller than the typical time δ_η^2/D_η required for complete ordering within the diffuse interface. As a result, disorder trapping by the diffuse interface exists under the velocity condition $V > D_\eta/\delta_\eta$, or under the time scale condition $\delta_\varphi/V_\varphi < \delta_\eta^2/D_\eta = \delta_\eta/V_{DT}$. From these conditions it follows that the critical velocity for the disorder trapping, V_{DT}, becomes smaller as the ratio between the thickness of the ordering zone δ_η and the diffuse interface thickness δ_φ increases:

$$V_{DT} < \frac{\delta_\eta}{\delta_\varphi} V_\varphi. \tag{21}$$

As follows from the inequality (21), the disorder trapping becomes more pronounced with the increase of the diffuse interface velocity V_φ and the decrease of the diffuse interface thickness δ_φ. Furthermore, with a "sharper" ordering zone, i.e., with decreasing δ_η, the beginning of disorder trapping is shifted to larger phase interface velocities. These situations are illustrated in Fig. 3.

7 Conclusions

1. Using a phase field method in the theory of fast phase transformations, a new model for disorder trapping by a rapidly moving interface is formulated. The model consists of a system of hyperbolic equations for mass transport by atomic diffusion, dynamics of the phase field, and dynamics of atomic ordering.

2. The system of model equations has been solved numerically to reach steady-state from initially non-stationary states. For these steady-states, the interface velocity and long-range order parameter are analyzed depending on the type of free energy and relaxation time τ_η for the rate of long-range order parameter $\partial\eta/\partial t$.

3. It has been shown quantitatively that there are critical undercoolings at which the trapping of disorder begins and finishes with the formation of a completely disordered phase. Using the results of our computations, a criterion for disorder trapping by rapid diffuse interface has been formulated.

Acknowledgments

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