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Kinetic phase diagram for a binary system near the transition to diffusionless solidification

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Abstract – The rapid solidification of a binary mixture in the region of the interface velocities V close to the diffusion speed in the bulk of the liquid phase V_D is considered within the framework of the local nonequilibrium approach. In this high-speed region the derivation of the analytical expression for the "temperature-velocity" response function representing kinetic phase diagram is given without using the concept of the equilibrium phase diagram. The modes of movement of the interface both without and with the drag effect are analyzed. It is shown that the drag effect can be accompanied by a local interface temperature maximum at $V = V_D$.

Introduction. – Quantitative modeling of the process of nonequilibrium solidification of metal melts has been attracting attention for the past few decades and is an object of scientific interest at the present time [1,2]. This interest is primarily due to the fact that nonequilibrium solidification provides the potential opportunity for obtaining metastable materials and, in particular, supersaturated solid solutions [2].

At low speeds of the solidification front the composition and properties of the final phase can be predicted based on representations about local equilibrium of the interface and usage of the equilibrium phase diagram. However, at the sufficiently high front speeds, deviations from the local equilibrium at the interface can become significant. Rapid solidification experiments [3–7] show that on a fast-moving phase boundary the processes of segregation are suppressed, the solute distribution is no longer determined in accordance with the equilibrium phase diagram and the solute trapping by the solid phase takes place.

The effect of complete solute trapping arises when the solidification speed V reaches the value of the solute diffusion speed in the bulk of the liquid phase V_D [8]. In this case, the solute does not have time to diffuse into the bulk of the liquid and is trapped by the solid phase at a concentration equal to its initial concentration in the melt. The new phase is then formed under conditions of diffusionless solidification with the partition coefficient $k = 1$. A number of theoretical models proposed earlier for

describing solute trapping (see, for example, review [1]) actually assume that $V/V_D \ll 1$ and predict that complete solute trapping with $k = 1$ is only possible asymptotically for $V \to \infty$, that corresponds to an infinitely large value of V_D. However, available experimental data [4–7] clearly demonstrate that the transition to complete solute trapping giving rise to diffusionless solidification occurs at essentially finite values of V. Theoretically, the same conclusion follows from the locally nonequilibrium model (LNM) developed in the works [8–18]. According to LNM, the solidification speeds observed in modern experiments can reach or even exceed the diffusion speed V_D and, as a result, the deviation from local equilibrium can be significant not only at the interface but in the bulk of the liquid phase as well. The so-called local nonequilibrium approach has been successfully applied to numerical simulation of experimental data over a wide range of solidification rates and is consistent with MD modeling of rapid solidification of a number of binary systems [19–24].

The modeling of the solidification process depends to a large extent on the boundary conditions realized at the phase interface and, in particular, on the relationship between temperature, the interface velocity and the solute concentration (the so-called "temperature-velocity" response function). The response function together with the nonequilibrium partition coefficient $k(V)$ allows one to find the kinetic phase diagram generalizing the equilibrium phase diagram to nonequilibrium solidification process.

Within the frame of LNM derivation an analytical expression for the response function in the case of a dilute melt, sharp interface and linear approximation for equilibrium solidus and liquidus has been considered in [12,17]. In particular, it has been assumed that at the temperature at which the interface moves, the solid and liquid phases can be in equilibrium with each other with the solute concentrations determined by the equilibrium solidus and liquidus, *i.e.*, determined on the basis of the equilibrium phase diagram [25]. This approximation may be applicable if undercooling at the solidification front is relatively not large. However, at the large undercooling achievable in modern experiments it may turn out that moving at high speed the interface has a sufficiently low temperature at which an alloy in an equilibrium state in the liquid phase does not exist and usage of the above assumption is not correct. For example, during the solidification of a Si-9 at.% As alloy, often used as a test system, the interface temperature can reach a value of the order $T = 1373\,\mathrm{K}$ [6], below which a dilute alloy exists in equilibrium only in the form of a solid solution [26].

In this paper within the local nonequilibrium approach we consider the derivation of the "temperature-velocity" response function without using the concept of equilibrium phase diagram, *i.e.*, without using equilibrium properties systems, and without using the assumption about phase equilibrium at the temperature of the fast-moving interface. The problem is considered in the region of speeds V close to the critical speed V_D at which a transition from diffusion-controlled growth to diffusionless solidification with $k = 1$ occurs. The idea of considering the solidification process in this speed region has been previously proposed to find the boundary interface conditions and the nonequilibrium partition coefficient for the interface moving at a constant temperature [27]. Here, the approach developed in [27] is applied to take into account temperature changes at the interface and to obtain the response function for large supercooling in the case of a diluted melt. The response function (nonequilibrium liquidus) is then used to analyze the temperature behavior of the interface near the transition to diffusionless solidification and compare the modes of movement of the interface both with and without taking into account the drag effect.

The chemical potentials. – Consider a solidifying binary alloy consisting of two component A (solvent) and B (solute). Let the molar fractions of the solute on the liquid and the solid sides of the interface, respectively, be C_L and C_S. The liquid and solid phases are separated by a planar interface moving steady state with the velocity of V. When modeling the response function one usually proceeds from the kinetic equation relating the velocity V to the thermodynamic driving force of phase transformation ΔG

$$V = V_0(1 - e^{\Delta G/RT}), \qquad (1)$$

where T is the interface temperature, R is the gas constant and V_0 is the upper limit of the solidification rate [28].

In the model of solidification without solute drag effect, ΔG is represented as (see, for example, [29])

$$\Delta G = \Delta G_{DF} = C_S\Delta\mu_B + (1 - C_S)\Delta\mu_A, \qquad (2)$$

where ΔG_{DF} is the change of the Gibbs free energy when one mole of a substance solidifies in the composition C_S, $\Delta\mu_i = \mu_i^S - \mu_i^L$ and μ_i^{LS} are chemical potentials of the component i $(i = A, B)$ at the liquid (L) and the solid (S) sides of the interface. In the model with solute drag, ΔG is given by

$$\Delta G = \Delta G_C = \Delta G_{DF} - \Delta G_D = C_L\Delta\mu_B + (1 - C_L)\Delta\mu_A, \qquad (3)$$

where ΔG_C is the crystallization free energy and $\Delta G_D = (C_L - C_S)(\Delta\mu_A - \Delta\mu_B)$ is a part of the free energy spent on changing the composition of the solid phase with C_L on C_S when A and B atoms diffuse through the interface.

Using (2) (or (3)) eq. (1) can be written as

$$C_{S(L))}\Delta\mu_B + (1 - C_{S(L)})\Delta\mu_A = RT\ln(1 - V/V_0). \quad (4)$$

It should be noted that the driving force of the phase transformation ΔG at given changes $\Delta\mu_A$, $\Delta\mu_B$ is uniquely defined by eq. (2) (or (3) if the solute drag is taken into account). The nature of the nonequilibrium state in the bulk of phases is manifested only through the chemical potentials μ_i^{LS}. If the medium is in a local equilibrium state μ_i^{LS} are functions of the concentration C and temperature T. However, in the case of rapid motion of the interface, the local equilibrium in the diffusion field of the liquid phase may not have time to establish. In this case the local nonequilibrium approximation is more appropriate. In a local nonequilibrium state the entropy of the system S depends on not only the classical variables C and T but also the diffusion current J, which, like the concentration and temperature, is considered as an independent variable, *i.e.*, $S = S(C, T, J)$ [30]. It follows that in a local nonequilibrium state the chemical potential $\mu = -T\partial S/\partial C$ is a function of the same variables, that is $\mu_i^{LS} = \mu_i^{LS}(C, T, J)$ [27].

Now let the interface move stationary at a speed V close to the velocity V_D for which the diffusionless solidification takes place with $J_L = 0$ and the solute concentration in both phases equals the initial concentration in the melt c_0. Taking into account the above and neglecting diffusion in the solid phase, one can write down for chemical potentials in the region $V \sim V_D$ the following expansion [27]:

$$\mu_i^L(C_L, T, J_L) = \mu_{leq,i}^L(C_L, T) + \alpha_i\frac{RT}{\rho V_D}J_L + \cdots, \quad (5)$$

$$\mu_i^S(C_S, T, J_S) = \mu_{leq,i}^S(C_S, T), \qquad (6)$$

where ρ is the mass density of the medium [27]. The terms in (5) and (6), independent of J_L, represent the local equilibrium part of the chemical potential $\mu_{leq,i}^{LS}$. As follows from eq. (5) for $V_D \to \infty$, $\mu_i^L \to \mu_{leq,i}^L$. The coefficient at J_L has been chosen such that α_i is a dimensionless parameter of order one and its sign can be defined for the

following reasons. According to a well-known equality, for the entropy of the liquid phase one can write down, using eq. (5),

$$S = - \sum_i C_i \left(\frac{\partial \mu_i^L}{\partial T} \right)_{p,J} = S_{leq} - \frac{RJ_L}{\rho V_D} \sum_i \alpha_i C_i,$$

where p is the pressure. Since in the local equilibrium state the entropy of the system S_{leq} is greater than in the local nonequilibrium one[1], then $\Delta S = S - S_{leq} < 0$. The latter inequality is automatically executed when $\alpha_i > 0$ ($J_L > 0$).

For states close to diffusionless solidification the local equilibrium part of the chemical potential can be expanded into a series in powers of $C - c_0$ and $T - T^*$, where T^* is the temperature of the interface moving with a speed V_D. Then, restricting ourselves to the linear approximation, we write eqs. (5) and (6) in the following form:

$$\mu_i^L(C_L, T, J_L) = \mu_i^{*L} + \frac{\partial \mu_i^{*L}}{\partial c_0}(C_L - c_0)$$
$$+ \frac{\partial \mu_i^{*L}}{\partial T^*}(T - T^*) + \alpha_i \frac{RT}{\rho V_D} J_L, \qquad (7)$$

$$\mu_i^S(C_S, T, J_S) = \mu_i^{*S} + \frac{\partial \mu_i^{*S}}{\partial c_0}(C_S - c_0) + \frac{\partial \mu_i^{*S}}{\partial T^*}(T - T^*), \qquad (8)$$

where

$$\mu_i^{*LS} = \mu_i^{LS}(C_{LS} = c_0, T = T^*, J_{LS} = 0) = \mu_{leq, i}^{LS}(c_0, T^*). \qquad (9)$$

Further let us consider a dilute solution for which Raoult's and Henry's laws are valid. In this case local equilibrium chemical potentials can be represented as

$$\mu_{leq, A}^{LS}(C, T) = \mu_{0A}^{LS}(T) + RT \ln(1 - C), \qquad (10)$$
$$\mu_{leq, B}^{LS}(C, T) = \mu_{0B}^{LS}(T) + RT \ln C, \qquad (11)$$

where $\mu_{0A(B)}^{L(S)}(T)$ is the chemical potential of the pure component $A(B)$ at the temperature T in the liquid (solid) state. Calculating the derivatives of (10), (11) and substituting them in (7), (8), one obtains (at $J_S = 0$)

$$\mu_A^{LS} = \mu_A^{*LS} - \frac{RT^*}{1 - c_0}(C_{LS} - c_0)$$
$$+ [-S_{0A}^{*LS} + R \ln(1 - c_0)](T - T^*) + \alpha_A \frac{RT^*}{\rho V_D} J_{LS}, \quad (12)$$

$$\mu_B^{LS} = \mu_B^{*LS} - \frac{RT^*}{c_0}(C_{LS} - c_0)$$
$$+ [-S_{0B}^{*LS} + R \ln c_0](T - T^*) + \alpha_B \frac{RT^*}{\rho V_D} J_{LS}, \qquad (13)$$

---

[1] An isolated thermodynamic system, initially being in the local nonequilibrium state with entropy S, in the process of relaxation to an equilibrium state passes through an intermediate the local equilibrium state. Since its entropy can only increase during the irreversible process, $S < S_{leq}$ (see also discussion in [31]).

where $S_{0i}^{*LS} = -\partial \mu_{0i}^* / \partial T^*$ is the entropy pure component i in local equilibrium at temperature T^*. It follows from (12), (13) that the local nonequilibrium chemical potentials changes through the interface are

$$\Delta \mu_A = \Delta \mu_A^* + L_A^*(T - T^*)/T^*$$
$$+ RT^* \frac{C_L - C_S}{1 - c_0} - \alpha_A \frac{RT^*}{\rho V_D} J_L, \qquad (14)$$

$$\Delta \mu_B = \Delta \mu_B^* + L_B^*(T - T^*)/T^*$$
$$+ RT^* \frac{C_S - C_L}{c_0} - \alpha_B \frac{RT^*}{\rho V_D} J_L, \qquad (15)$$

where $L_i^* = -T^*(S_{0i}^{*S} - S_{0i}^{*L})$ is the latent heat solidification of the pure component i at the temperature T^* ($L_i^* > 0$), $\Delta \mu_i^* = \mu_i^{*S} - \mu_i^{*L} = \mu_{0i}^{*S} - \mu_{0i}^{*L} = \Delta \mu_{0i}^*$ and $\mu_{0i}^{*LS} = \mu_{0i}^{LS}(T^*)$.

The velocity-temperature response function. –

The interface movement without solute drag. Taking into account eqs. (14), (15), one writes down the equality (4) for the model without solute drag in the form

$$(1 - C_S) \Big[\Delta \mu_{0A}^* + L_A^*(T - T^*)/T^*$$
$$+ RT^* \frac{(C_L - C_S)}{1 - c_0} - \alpha_A \frac{RT^*}{\rho V_D} J_L \Big]$$
$$+ C_S \Big[\Delta \mu_{0B}^* + L_B^*(T - T^*)/T^* + \frac{RT^*}{c_0}(C_S - C_L)$$
$$- \alpha_B \frac{RT^*}{\rho V_D} J_L \Big] = RT \ln(1 - V/V_0). \qquad (16)$$

It follows from (16) that at $V = V_D$, $C_L = C_S = c_0$, $T = T^*$ and $J_L = 0$ there is the equality

$$(1 - c_0)\Delta \mu_{0A}(T^*) + c_0 \Delta \mu_{0B}(T^*) = RT^* \ln(1 - V_D/V_0), \qquad (17)$$

which can be considered as an equation for determining temperature T^* of the interface moving at the speed V_D at the initial concentration of the solute in the melt c_0. It can be shown that for a diluted melt and a small deviation $T^* - T_A$, where T_A is the melting temperature of the major component, the solution of this equation coincides with the interface temperature T^* found in the works [12,17].

Taking into account eq. (17) and the boundary condition $J_L = (C_L - C_S)\rho V$, one can rewrite eq. (16) in the form

$$(c_0 - C_S)(\Delta \mu_{0A}^* - \Delta \mu_{0B}^*) + L^*(C_S)(T - T^*)/T^*$$
$$+ \frac{RT^*(C_L - C_S)(c_0 - C_S)}{c_0(1 - c_0)}$$
$$- \alpha(C_S)RT^*(C_L - C_S)V/V_D =$$
$$RT \ln(1 - V/V_0) - RT^* \ln(1 - V_D/V_0), \qquad (18)$$

where the following notations have been introduced:

$$L^*(C_S) = (1 - C_S)L_A^* + C_S L_B^*,$$
$$\alpha(C_S) = (1 - C_S)\alpha_A + C_S \alpha_B.$$

In the absence of diffusion in the solid one can put $C_S = c_0$ and write eq. (18) in the linear approximation in $V - V_D$ and $T - T^*$ as

$$\frac{T - T^*}{T^*} = \frac{\alpha(1-k)C_L + (V_D - V)(V_0 - V_D)^{-1}}{L^*/RT^* - \ln(1 - V_D/V_0)}, \quad (19)$$

where $k = C_S/C_L = c_0/C_L$ is the nonequilibrium solute partition coefficient and L^*, α are taken at $C_S = c_0$. In the derivation of eq. (19) it has been taken into account that $1 - k \sim V_D - V$ at $V < V_D$. The equality (19) is the sought response function representing the kinetic phase diagram in the case of the interface movement with a speed close to V_D. At $V_D/V_0 \ll 1$ one can simplify the expression (19) and get the nonequilibrium liquidus equation in the form

$$T = T^* + \alpha\frac{RT^{*2}}{L^*}(1-k)C_L + \frac{RT^{*2}}{L^*}\frac{V_D - V}{V_0}. \quad (20)$$

In the linear approximation in $V - V_D$ the solidus and liquidus lines coincide since in this case $(1 - k)C_L = (1 - k)C_S/k \simeq (1 - K)C_S$.

In the region of the speeds V close to V_D the partition coefficient $k(V)$ can also be expanded into a series in powers of $V_D - V$. Since for the interface the only dimensionless quantity including V_D is the ratio V_{DI}/D_D, where V_{DI} is the interfacial diffusion speed, then for dimensional reasons the expansion of $k(V)$ in this region can be written as

$$k(V) = \begin{cases} 1 - f(V_{DI}/V_D)(1 - V/V_D) + \cdots, & V < V_D, \\ 1, & V \geq V_D, \end{cases} \quad (21)$$

where f is some dimensionless function, in the simplest case one takes $f(V_{DI}/V_D) = f_0 V_{DI}/V_D$ and $f_0 > 0$ is dimensionless coefficient of order one. It should be noted that eq. (21) represents the expansion of $k(V)$ near V_D in the most general form regardless of the details of the interface kinetics. The different representations of the velocity-dependent partition coefficient for some models of interface kinetics, taking into account complete solute trapping at $V \geq V_D$, can be found in refs. [15,16]. For example, for the partition coefficient from [15] $f(V_{DI}/V_D) = 2(1 - k_e)(1 - c_0)V_{DI}/V_D$, where k_e is the equilibrium partition coefficient.

Substitution of (21) into (20) gives in the linear approximation in $V - V_D$

$$\Delta T = (V_D/V_0)$$
$$\times \begin{cases} (1 + \alpha c_0 f_0 V_{DI} V_0/V_D^2)(1 - V/V_D), & V < V_D, \\ 1 - V/V_D, & V \geq V_D, \end{cases}$$
$$(22)$$

where $\Delta T = (T - T^*)L^*/RT^{*2}$. As can be seen from (22), at positive α, $\Delta T > 0$ for $V/V_D < 1$ and T decreases with the growth of V changing the slope at the point V_D (the behavior of T is qualitatively given by the curve 1 in fig. 1).
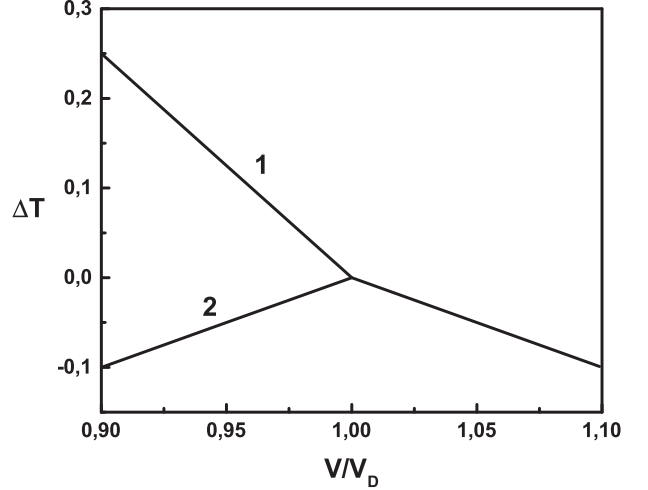

Fig. 1: The qualitative behavior of ΔT depending on V/V_D; the curve 1 corresponds to eq. (22) (model without solute drag); the curve 2 corresponds to eq. (28) (model with solute drag) at $|\Delta\mu_{0A}^*|V_0/10RT^*V_D > 1$.

Solute drag effect. In the model with drag effect one has

$$C_L\Delta\mu_B + (1 - C_L\Delta\mu_A) = RT\ln(1 - V/V_0). \quad (23)$$

Substituting now relations (14) and (15) in eq. (23) and performing transformations similar to those used in the derivation of eq. (19), one obtains in the linear approximation in $V - V_D$

$$\frac{T - T^*}{T^*} =$$
$$\frac{(\alpha + \Delta\mu_0^*/RT^*)(1-k)C_L + (V_D - V)(V_0 - V_D)^{-1}}{L^*/RT^* - \ln(1 - V_D/V_0)}, \quad (24)$$

where $\Delta\mu_0^* = \Delta\mu_{0A}^* - \Delta\mu_{0B}^*$. At $V_D/V_0 \ll 1$, eq. (24) yields the equation of the nonequilibrium liquidus in the form

$$T = T^* + \frac{RT^{*2}}{L^*}(\alpha + \Delta\mu_0^*/RT^*)(1-k)C_L + \frac{RT^{*2}}{L^*}\frac{V_D - V}{V_0}. \quad (25)$$

In the linear approximation in $V - V_D$ the solidus equation is also given by eq. (25) (see the note after eq. (20)). At $V \geq V_D$, eq. (25) and eq. (20) coincide. To analyze eq. (25) for $V < V_D$ we consider eq. (17) from which it follows that:

$$\frac{\Delta\mu_0^*}{RT^*} = \frac{\Delta\mu_{0A}^*}{c_0 RT^*} + \frac{V_D}{c_0 V_0}. \quad (26)$$

Taking into account eq. (26) one can rewrite eq. (25) as

$$\Delta T = \left(\alpha c_0 + \frac{\Delta\mu_{0A}^*}{RT^*} + V_D/V_0\right)\frac{1-k}{k} + \frac{V_D - V}{V_0}. \quad (27)$$

Bearing in mind the diluted melt ($c_0 \to 0$) and using (21), we obtain from (27) in the linear approximation in $V - V_D$

(by neglecting the second-order term $c_0(V - V_D)$)

$$\Delta T = \left[f_0 \frac{V_{DI}}{V_D} \left(\frac{\Delta\mu_{0A}^*}{RT^*} + \frac{V_D}{V_0} \right) + \frac{V_D}{V_0} \right](1 - V/V_D). \quad (28)$$

It is seen from (28) that for $f_0 \approx 1$ the inequality $\Delta T > 0$ holds[2] if

$$\frac{|\Delta\mu_{0A}^*|}{RT^*} \frac{V_0/V_D}{1 + V_D/V_{DI}} < 1. \quad (29)$$

Under the condition $V_0/V_D \gg 1$, the expression on the left-hand side of inequality (29) can be large enough (for example, for Si-9 at.% As $V_D/V_{DI} \sim 1$ [2]). Therefore, it is obvious that the condition (29) will not be automatically satisfied for all systems. This means that when the speed of the front increases towards the critical value of V_D in some cases the solute drag effect can be accompanied by an increase in the interface temperature, $\Delta T = T - T^* < 0$ (curve 2 in fig. 1). It is this behavior that was found in the numerical simulation of the rapid solidification of the Si-9 at.% As system [20].

Conclusion. – The most important consequence of the absence of local equilibrium in the bulk of the liquid phase in the processes of rapid solidification is transition to complete solute trapping at the finite velocity of the solidification front $V = V_D$. This circumstance makes it possible to analyze analytically the details of the solidification process in the region of high velocities close to V_D. For the indicated speed range the analytical expression for the temperature response function representing kinetic phase diagram and taking into account solute drag effect has been derived. In contrast to previous works, in the given approach the equilibrium properties of the alloy based on the equilibrium phase diagram of the system have not been used. At high speeds of the solidification front, the interface temperature may be quite low, so that in the equilibrium state at this temperature the alloy can only exist in the form of a solid solution and the use of equilibrium liquidus and solidus loses its meaning.

In the case of a dilute melt, as follows from the analysis of the response function (28), the solute drag can be accompanied by an increase of the temperature of the interface when its velocity approaches to the critical value of V_D. Numerical simulation of high-speed solidification of the Si-9 at.% As alloy shows also the existence of a local temperature maximum in the region of large V [20]. Growth of the interface temperature as V approaches to V_D is perhaps some sign of drag effect. However,

detailed research on this issue requires studying the solidification process on a large number of different binary systems.

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[2]At $T = T^* < T_A$, where T_A is the melting temperature of major component, the solid phase is more stable and $\Delta\mu_{0A}^* = \mu_{0A}^{*S} - \mu_{0A}^{*L} < 0$.