INTERFACE INSTABILITY IN THE PHASE TRANSFORMATION PROCESS

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To the best of our knowledge, the stability problem for a plane interface between two phases in the process of phase transformation was initially formulated in Sekerka's work [1] on the solidification of one component of a binary alloy. In the present study we are concerned mainly with this specific problem, but our results can, with certain reservations, be applied to the process of crystallization from a supercooled liquid if the process is described by the Stefan problem in the isotropic approximation [2].

Sekerka's work [1] suffers from a significant shortcoming insofar as the temporal decay of the velocity of the front is disregarded and, accordingly, a conclusive answer is not obtained as to the stability of a plane interface. Here we determine the pattern of evolution of the distortions of a plane interface with time in the linear approximation. We also give previously obtained results [3] of an investigation of the stability of spherical growth of a nucleus of the new phase. Although [3] referred to the stability of electron–hole droplets in semiconductors, its results are of a general nature and can be used to describe the above-mentioned phase transformations.

1. Model of Solidification of a Binary Alloy. We assume that a matrix of one material contains particles of another substance (solution), forming an alloy. If the heat transfer between both substances is sufficiently effective, it can be assumed that the temperature is equal to the temperature of the matrix T. It is postulated that for a certain concentration of the solution particles they form another modification by a first-order phase transition. It is assumed that the nature of these two phases is inconsequential, since the problem is treated in the isotropic approximation, i.e., the solution is actually regarded as a supersaturated vapor, and the new, denser phase as a condensed liquid. Normally the degree of supersaturation $\delta n = n - n_T$ (where n_T is the saturated vapor density and n is the vapor density) is always small in comparison with the density of the condensed phase N, and indeed we consider this to be true below. Following the stage of formation of a critical nucleus, disregarded here, the phase transformation process entails the diffusion of solution particles toward the surface of the condensed phase with subsequent condensation on that surface. Thus, the equation for the diffusion of solution particles through the matrix must hold in the vapor:

$$\partial n/\partial t = D\Delta n, \quad (1.1)$$

where D is the diffusion coefficient.

The following condition must hold at the phase interface:

$$n|_S = n_T(1 + 2\sigma/NRT), \quad (1.2)$$

where σ is the coefficient of surface tension and R is the radius of curvature of the surface [4]. We thus assume that the condensation is a slow process and the conditions are close to thermodynamic equilibrium (this problem is analyzed in more detail in [3]). Another condition follows from the mass conservation principle:

$$D\partial n/\partial \mathbf{v}|_S = v_\nu N, \quad (1.3)$$

where v_ν is the normal velocity of the condensation front as specified by the surface S(t). At large distances from the front

$$n|_{r\to\infty} = n_\infty > n_T.$$

The case of formation of a solid phase from a supercooled liquid differs from the one discussed above insofar as during transition the pressure (rather than the matrix temperature) is assumed to be constant, whereas the temperature varies from a certain value T_∞ lower than the melting point T_M in the liquid to the melting point at the surface:

$$T|_S = T_M\,(1 - 2\sigma/Nq_TR), \quad (1.2a)$$

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where q_T is the heat of phase transformation. The heat flux admitted to the interface sets it in motion, in which case

$$-c\chi\partial T/\partial\nu|_S = v_\nu q_{T\cdot} \tag{1.3a}$$

where c is the heat capacity and χ is the thermal diffusivity of the liquid. The heat-conduction equation must hold in the liquid:

$$\partial T/\partial t = \chi\Delta T. \tag{1.1a}$$

It is seen at once that Eqs. (1.1a)-(1.3a) differ from (1.1)-(1.3) only by a change of notation and correspond to the Stefan problem [2]. We abide by the notation of (1.1)-(1.3). The results of our investigation can be applied to the condensation of a vapor in air if the latter is regarded as the matrix, which maintains a constant temperature.

We consider the motion of a plane condensation or crystallization front. Normally the effort to sustain such a plane front stems from the desire to obtain a good homogeneous crystal. Inasmuch as $\delta n/N \ll 1$, diffusion is incapable of transporting the required quantity of solute to ensure motion of a plane front with a constant velocity (unlike the assumption made in [1]). However, a self-similar solution $n_0 = n_0(z/2\sqrt{Dt})$ exists, where

$$n_0(u) = n_\infty + A\int_\infty^u e^{-x^2}dx, \quad u = \frac{z}{2\sqrt{Dt}}. \tag{1.4}$$

The corresponding interface is given by the equation $z_0 = 2u_0\sqrt{Dt}$, where the constants A and u_0 are evaluated from the boundary conditions (1.2) and (1.3). In the limit of small $\delta n/N$ we obtain ($\delta n = n_\infty - n_T$)

$$u_0 \simeq \delta n/\sqrt{\pi}N, \; A \simeq 2\delta n/\sqrt{\pi}. \tag{1.5}$$

We now study the behavior of small perturbations of the solution (1.4), for the time being considering one Fourier component, and putting

$$n = n_0(u) + f_k(z, t)\,e^{i\mathbf{k}\boldsymbol{\rho}}, \tag{1.6}$$

where $f_k(z, t)$ is a small quantity associated with the small variation of the interface surface

$$z_0' = 2u_0\sqrt{Dt} + \zeta_k(t)\,e^{i\mathbf{k}\boldsymbol{\rho}}, \tag{1.7}$$

and the vectors $\mathbf{k}$ and $\boldsymbol{\rho}$ are situated in the xy plane, which is parallel to the unperturbed interface. The function $f_k(z, t)$ obeys the equation

$$\partial f_k/\partial t = D(\partial^2 f_k/\partial z^2 - k^2 f_k). \tag{1.8}$$

After linearization, the boundary conditions (1.2) and (1.3) assume the form

$$f_k|_{z_0} + \zeta_k\frac{\partial n_0}{\partial z}\Big|_{z_0} = n_T\gamma k^2\zeta_k = \frac{N}{D}v_k\zeta_k; \tag{1.9}$$

$$N\frac{d\zeta_k}{dt} = \frac{N}{D}v_0 v_k\zeta_k + D\zeta_k\frac{\partial^2 n_0}{\partial z^2}\Big|_{z_0} + D\frac{\partial f_k}{\partial z}\Big|_{z_0}, \tag{1.10}$$

where $v_0 = dz_0/dt = u_0\sqrt{D/t}$, $v_k = (n_T/N)\gamma Dk^2$, $\gamma = \sigma/NT$.

From Eqs. (1.8)-(1.10) we can obtain an integral equation relating f_k to its value f_{k_0} at the boundary $z = z_0(t)$ and the quantity $\zeta_k(t)$, provided that we use the Fourier transform with respect to the variable z:

$$f_k(z, t) = \frac{1}{2\sqrt{\pi D}}\int_0^t \frac{e^{-Dk^2(t-\tau)}}{\sqrt{t-\tau}}\exp\left[-\frac{(z - z_0(\tau))^2}{4D(t-\tau)}\right]$$
$$\times\left[f_{k0}(\tau)\frac{z - z_0(\tau)}{2(t-\tau)} - N\frac{d\zeta_k(\tau)}{d\tau}\right]d\tau + \int_{-\infty}^{\infty}\exp\left[-D(k^2+q^2)t + iqz\right]f_q|_{t=0}\,dq. \tag{1.11}$$

The last term can be dropped, since it tends rapidly to zero and is not associated with the existence of a moving boundary.

Equation (1.11) can be transformed into an integral equation for the displacement of the front ζ_k by passing to the limit $z \to z_0(t)$ therein. It is necessary to exercise the usual caution here, because the denominator of the first term in the brackets decays to zero as $(t-\tau)^{3/2}$ and convergence is attributable to the factor $\exp[(-z-z_0(\tau))^2/(4D(t-\tau))]$. For small differentials $\delta z = z - z_0(t)$ this consideration will be important only near $t = \tau$, and so the singular part of this term can be written approximately as

$$\frac{f_{k0}(t)}{4\sqrt{\pi D}} \lim_{\delta z \to 0} \delta z \int_0^t \frac{\exp\left[-\frac{(\delta z)^2}{4D(t-\tau)}\right]}{(t-\tau)^{3/2}} d\tau \simeq \frac{f_{k0}(t)}{2}.$$

From this result we finally obtain for $z = z_0(t)$

$$f_{k0}(t) = \frac{1}{\sqrt{\pi D}} \int_0^t \frac{d\tau}{\sqrt{t-\tau}} \exp\left[-Dk^2(t-\tau) - \frac{(z_0(t) - z_0(\tau))^2}{4D(t-\tau)}\right]\left[f_{k0}(\tau)\frac{z_0(t) - z_0(\tau)}{2(t-\tau)} - N\frac{d\zeta_k(\tau)}{d\tau}\right]. \tag{1.12}$$

Expressing $f_{k0}(t)$ in terms of $\zeta_k(t)$ according to (1.9), we thus arrive at an integral equation for $\zeta_k(t)$. However, a certain simplification is possible in connection with the smallness of $\delta n/N$ and, accordingly, u_0 and z_0. In this regard, we note that

$$\frac{[z_0(t) - z_0(\tau)]^2}{4D(t-\tau)} = u_0^2 \frac{\sqrt{t} - \sqrt{\tau}}{\sqrt{t} + \sqrt{\tau}} \leqslant u_0^2 \ll 1.$$

We can therefore omit the corresponding exponential. Invoking the boundary condition (1.9) as well, we finally obtain

$$[v_k - v_0(t)]\,\zeta_k(t) = \sqrt{\frac{D}{\pi}} \int_0^t \frac{e^{-Dk^2(t-\tau)}}{\sqrt{t-\tau}} d\tau \left\{[v_k - v_0(\tau)]\frac{u_0\zeta_k(\tau)}{\sqrt{D}(\sqrt{t} + \sqrt{\tau})} - \frac{d\zeta_k(\tau)}{d\tau}\right\}. \tag{1.13}$$

We analyze only the behavior of $\zeta_k(t)$ for large times, without any concern for how the arbitrary constant entering into this asymptotic representation is related to the initial data [including $f_k(t = 0)$].

The integral of (1.13) contains the factor $e^{-Dk^2(t-\tau)}$, which shows that values of $t - \tau \sim 1/Dk^2$ are essential. Neglecting the quantity ζ_k on the right-hand side of (1.13) and taking $d\zeta_k/d\tau$ outside the integral sign for the value $\tau = t$, we find that the asymptotic behavior of $\zeta_k(t \to \infty)$ is given by the equation

$$[v_0(t) - v_k]\,\zeta_k(t) = \frac{d\zeta_k(t)}{dt}\sqrt{\frac{D}{\pi}} \int_0^t \frac{e^{-Dk^2(t-\tau)}}{\sqrt{t-\tau}} d\tau,$$

and, considering the rapid convergence of the integral, we can set the upper limit of the integral equal to infinity. As a result, we obtain

$$\zeta_k(t) = B_k \exp\left[\int_0^t k(v_0(\tau) - v_k)\,d\tau\right]. \tag{1.14}$$

It is seen that the individual Fourier component is always stable; surface tension stabilizes the growth of the perturbation, since for large times $v_0 \sim 1/\sqrt{t} \ll v_k$. However, for a fixed time t the argument of the exponential function in (1.14) is positive for

$$2u_0\sqrt{Dt} > \frac{n_T}{N}\gamma k^2 Dt, \tag{1.15}$$

i.e., for

$$k^2 < \frac{2u_0 N}{n_T \gamma \sqrt{Dt}} = k_c^2(t). \tag{1.16}$$

Thus, as $t \to \infty$ a risk is presented by arbitrarily small values of k. We now test the validity of the foregoing approximations. First, in connection with the integration in (1.13)

$$\frac{t-\tau}{t} \sim \frac{1}{k_c^2 Dt} \sim \frac{n_T\gamma}{Nu_0\sqrt{Dt}} \to 0 \quad \text{as} \quad t \to \infty.$$

It is also evident that, since $t - \tau \ll t$ in the characteristic domain of integration, the ζ_k term can indeed by neglected on the right-hand side of (1.13), and the $d\zeta_k/d\tau$ term taken outside the integral sign.

We note here that the expression of $\zeta_k(t)$ in the form (1.14) can be deduced from the following elementary considerations. Because of the smallness of $\delta n/N$ the motion of the boundary is slow, and the quasisteady-state approximation is valid:

$$\partial f_k/\partial t \ll k^2 D f_k, \tag{1.17}$$

i.e., in place of Eq. (1.8) we have

$$\partial^2 f_k/\partial z^2 = k^2 f_k.$$

Clearly, for a solution decaying with z

$$\partial f_k/\partial z = -k f_k\,,$$

and on the basis of (1.9) condition (1.10) goes over to

$$\frac{d\zeta_k(t)}{dt} = k(v_0 - v_k)\zeta_k - \frac{v_0}{D}(v_0 - v_k)\zeta_k. \tag{1.18}$$

Next, we take into consideration the fact that for $k \sim k_c$ the first term on the right-hand side of (1.18) $\sim t^{-3/4}$, while the second is of the order $(\delta n)^2/Nt$, so that once again we obtain $\zeta_k(t)$ in the form (1.14) as $t \to \infty$. Using this result, we can test the quasisteady-state condition (1.17).

Thus, we finally obtain the following expression for the displacement of the interface:

$$\zeta(\rho, t) = \int B_k e^{ik\rho} \exp\left[\int_0^t k(v_0(\tau) - v_k)\, d\tau\right] \frac{d^2k}{(2\pi)^2}, \tag{1.19}$$

where ρ is a vector parallel to the phase interface. Inasmuch as small values of k are essential for large times, for a value of B_k that does not have singularities in this k domain we obtain

$$\zeta(\rho, t) \sim B_0 \int_0^\infty J_0(k\rho)\, e^{\varphi(k,t)} k\, dk, \tag{1.20}$$

where $\varphi(k, t) = k[2u_0\sqrt{Dt} - (n_T/N)\gamma k^2 Dt]$. Computing the integral with respect to k for large values of t by the method of steepest descent, we finally obtain

$$\zeta(\rho, t) \sim \frac{B_0}{\gamma^{3/4}(Dt)^{5/8}} \left(\frac{N^2 \delta n}{n_T^3}\right)^{1/4} \exp\left[\alpha\left(\frac{Dt}{\gamma^2}\right)^{1/4}\right] J_0\left(\frac{\beta\rho}{(\gamma^2 Dt)^{1/4}}\right), \tag{1.21}$$

where

$$\alpha = \frac{1}{\pi}\left[\frac{32(\delta n)^3}{27\sqrt{\pi} n_T N^2}\right]^{1/2}; \quad \beta = \left(\frac{2\delta n}{3\sqrt{\pi} n_T}\right)^{1/2};$$

and $J_0(y)$ is the Bessel function of zero order. Of course, the choice of origin in the basal plane is arbitrary. In the general case the surface configuration must be made up of randomly distributed perturbations of the type indicated.

Thus, the plane interface turns out to be unstable, and for large times the perturbation does not grow by a simple exponential law, but rather as an exponential function of $t^{1/4}$. We note that the space scale of the perturbation in the plane of the interface at a time when the perturbation becomes appreciable is given by the expression $\rho \sim r_{cr} n_T/\delta n$, where $r_{cr} = 2\sigma/T\delta n$ is the so-called critical radius of the nucleus [4].

2. Instability of Spherical Growth of Nuclei. This problem, which has been discussed in [3], is somewhat simpler than the preceding one, and its solution is also based on the smallness of $\delta n/N$, i.e., relatively slow motion of the interface in comparison with the diffusion process. The spherical growth of a nucleus is described by the diffusion equation

$$\partial n/\partial t = D\Delta n, \tag{2.1}$$

in which the slow motion of the interface permits $\partial n/\partial t$ to be neglected, so that ($\delta n = n_\infty - n_T$)

$$n_0 = n_\infty - \frac{\delta n}{r} R(t), \tag{2.2}$$

where R(t) is the radius of the nucleus and the boundary condition $n|_s = n_\infty - \delta n$ is used. Making use of the boundary conditions, we obtain

$$dR(t)/dt = D\delta n/NR. \tag{2.3}$$

Since the diffusion rate at a distance of order R is $v_D \sim D/R$, in reality the ratio

$$\frac{1}{v_D}\frac{dR}{dt} \sim \frac{\delta n}{N} \ll 1.$$

If the shape of the nucleus deviates from spherical, we assume that the vapor density and droplet radius are given by the relations

$$n = n_0(r) + f(\Omega, t, r), \; R' = R(t) + \zeta(\Omega, t).$$

Neglecting the time derivative of f in the diffusion equation (2.1), we have

$$f = \sum_{l,m} A_{lm} Y_{lm}(\Omega) r^{-l-1}, \tag{2.4}$$

where $Y_{lm}(\Omega)$ denotes spherical harmonics. Linearizing the boundary conditions with respect to ζ, we obtain

$$\zeta_{lm}\frac{\partial n_0}{\partial r}\Big|_R + A_{lm} = \frac{n_T \gamma (l-1)(l+2)}{R^2(t)}\zeta_{lm}; \tag{2.5}$$

$$N\frac{d\zeta_{lm}}{dt} = D\zeta_{lm}\frac{\partial^2 n_0}{\partial r^2}\Big|_R - D\frac{(l+1)}{R^{l+2}}A_{lm}, \tag{2.6}$$

where

$$\zeta(\Omega) = \sum_{l,m} \zeta_{lm}(t) Y_{lm}(\Omega).$$

We finally have

$$\frac{1}{\zeta_{lm}}\frac{d\zeta_{lm}}{dt} = \frac{(l-1)}{R^2} D\frac{\delta n}{N} - \frac{n_T \gamma D (l^2-1)(l+2)}{NR^3}. \tag{2.7}$$

To assess the instability it is necessary to compare the growth of the deviation with the growth of the droplet itself R(t) as described by Eq. (2.3). Therefore, the growth-rate factor characterizing instability is

$$\lambda_l = \frac{d}{dt}\ln\frac{\zeta(t)}{R(t)} = \frac{D(l-2)\delta n}{R^2 N} - \frac{n_T \gamma D (l^2-1)(l+2)}{NR^3}. \tag{2.8}$$

It is evident from this expression that with growth of the nucleus the perturbations lose stability with ever-greater l, because for large radii the surface tension becomes ineffectual. However, instability first sets in for $R = R_c$ when $l = 3$, where

$$R_c = 40\gamma n_T/\delta n. \tag{2.9}$$

It can be shown that

$$\zeta \sim \zeta_0 [R(t)/R_c]^{l-1}, \tag{2.10}$$

where ζ_0 is of the order of the initial perturbation, which is unknown. Since the exponent grows rapidly with l and perturbations disrupting the spherical shape always occur in reality, we can assume that the characteristic space scale of the resulting droplets must be $R \sim R_c$ (see also [5]).

We note in conclusion that the principal mechanism of the above-analyzed instability is such that the forward-advancing parts of the front acquire a large diffusion flow and therefore grow more rapidly.

We have thus shown that both a plane new-phase front and the spherical growth of a single nucleus are unstable processes. In the case of a plane portion of the front of finite dimensions stability can be attained and, accordingly, a homogeneous new phase precipitated if those dimensions are made sufficiently small for the given gradient (cutting off small values of k).

The given statement of the problem is very general and can be applied to a fairly broad class of processes associated with the formation of a new phase (neglecting anisotropy) in first-order phase transitions, which include, for example, crystal growth and liquid—vapor transitions. In particular, the investigated mechanism can be used to explain the small sizes of electron—hole droplets formed in semiconductors [3].

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STRUCTURAL CHARACTERISTICS OF SHOCK WAVES FROM UNDERWATER EXPLOSIONS OF HELICAL CHARGES

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Explosive sound sources have intrigued researchers for many years as a principal component of various kinds of sonar devices designed for the transmission of directional quasicontinuous-wave signals of long duration and large acoustic power. The category of such sources is broad and includes spark-discharge generators [1], condensed liquid [2] and solid high explosives [3-5], explosive gas mixtures [6-8], and the shock-generating effects of collapsing cavities (implosions) [9, 10]. The total energy parameters of the signals from certain explosive sound sources in water have been compared [11], and the spectral characteristics have been studied experimentally [12-16].

Naturally, explosive sources have considerable power, and their transmission is recorded at large distances. These attributes, however, prove inadequate for a broad class of problems in geophysical research, sonic navigation, and scientific investigations of the processes of shock wave propagation in the ocean. Typical problems are the directivity and relatively long duration of the signal, which are not a trivial matter to realize within the framework of explosive sources as predominantly "point" sources. An important problem is the tonal "coloring" of the signal to protect it against reverberation noise. It is not too surprising, therefore, that some of the solutions obtained to date have been based on the familiar notions of classical acoustics in regard to directional transmission from specially distributed sources. The latter represent: an explosive-cord line charge, which ensures shock wave propagation predominantly in a plane perpendicular to its axis [5]; a vertical line array of concentrated charges detonated with a definite frequency [3] and thus generating a prescribed sequence of shock waves, i.e., to a certain extent solving the problem of the duration and "coloring" of the transmitted signal as a result of its directionality. The coherent jetting effect has been utilized in the generation of directional signals by the detonation of a charge in a specially profiled conical liner [4].

There has been definite interest lately in sources in the form of spatial helix configurations of a high-explosive cord (HEC) charge, the transmission from which has a number of specific advantages: directivity both in the vicinity of the axis (typical of an annular source, owing to the high detonation rate) [17] and in the perpendicular plane (certain model of a line source in the case of a long helix); long duration [18]; highly controllable frequency of succession of shock waves for one given total length of the cord charge [19]. The characteristics of the evolution of the wave field from the underwater detonation of such charges are of unquestionable interest. Below, we discuss the fundamental results of their

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