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Lamellar eutectic growth at large thermal gradient. II. Linear stability

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Résumé. — Nous analysons la stabilité linéaire des structures eutectiques lamellaires périodiques stationnaires étudiées dans l'article précédent [I] vis-à-vis des perturbations de grande longueur d'onde. Nous montrons que, dans la limite des grands gradients thermiques considérée dans [I], et pour les structures de longueur d'onde de l'ordre de celle, $\lambda_{\text{min}}$, correspondant au surrefroidissement minimum : 1) il apparaît une instabilité associée à une variation quasi uniforme de la proportion de chacune des deux phases solides. Elle apparaît seulement pour les mélanges dont l'écart $u_\infty$, à la composition eutectique est du côté du diagramme de phase qui présente le plus grand « gap » de température. Dans ce cas, les structures stationnaires avec $\lambda < \lambda_0$, où $\lambda_0 \propto u_\infty$, sont instables ; 2) toutes les structures avec $\lambda / \lambda_{\text{min}} = O (1)$ sont stables vis-à-vis du mécanisme d'Eckhaus. Ceci entraîne que l'hypothèse dynamique utilisée dans des travaux antérieurs, selon laquelle les interfaces solide-solide seraient localement normales au front, n'est pas compatible avec l'approximation usuelle des petites déformations de front.

Abstract. — We analyze the linear stability of the periodic eutectic lamellar patterns studied in the preceding article [I] with respect to long wavelength perturbations. We show that, in the large thermal gradient limit considered in [I], and for patterns with wavelengths comparable with the minimum undercooling one, $\lambda_{\text{min}}$: 1) an instability with respect to a quasi-uniform variation of the phase fraction occurs only for mixtures with a departure from eutectic composition $u_\infty$ on the side of the phase diagram with the larger liquid-solid temperature gap. In this case, stationary patterns with $\lambda < \lambda_0$, where $\lambda_0 \propto u_\infty$, are unstable ; 2) all patterns with $\lambda / \lambda_{\text{min}} = O (1)$ are Eckhaus stable. This entails that the dynamical ansatz, used in previous works, that solid-solid interfaces are locally normal to the front, is not compatible with the usual small front deformation approximation.

1. Introduction.

In the preceding paper [1], hereafter referred to as [I], we have calculated analytically, in the limit of large thermal gradients, the front profiles associated with stationary lamellar eutectic directional solidification patterns. Although this analysis disproves the equal undercooling

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ansatz of Jackson-Hunt (JH) [2], our results agree qualitatively with their predictions: one such solution, with lamellae perpendicular to the average front, exists for each value of the space period $\tilde{\lambda}$ of the pattern (with $\tilde{\lambda} \ll \ell$ where $\ell$ is the chemical diffusion length). The average undercooling $\Delta T(\tilde{\lambda})$ exhibits a minimum for $\tilde{\lambda} = \hat{\lambda}_{\text{min}}$, where $\hat{\lambda}_{\text{min}}$ is of order $(d_1)^{1/2}$, where $d$ is a capillary length associated with liquid-solid surface tension(s). Since $\ell = D/V$ (where $D$ is the chemical diffusion coefficient in the liquid, $V$ the pulling velocity), $\hat{\lambda}_{\text{min}} \propto V^{-1/2}$.

It is found experimentally that, in a given experiment, lamellar spacing distributions [3] are rather narrow, and centered about an average value $\tilde{\lambda}$ of the order of magnitude of $\hat{\lambda}_{\text{min}}$. This « selected » wavelength $\tilde{\lambda} \propto V^{-\alpha}$ with, for many material [4], $\alpha = \frac{1}{2}$.

These observations raise the question of wavelength selection within the continuum of stationary patterns, and therefore, as a first step, the problem of their local stability. Several approaches to this problem have been proposed. The first ones [5, 6] did not, in particular, allow for variations of lamellar spacings. This a priori excludes, for example, studying a possible Eckhaus instability. Later approaches [7, 8] included the possibility of such motions, but assumed a flat front profile. More recently, Datye and Langer performed an extensive stability analysis of the Jackson-Hunt stationary [9] solutions, involving:

(i) an approximation of small front deformations and
(ii) a dynamical ansatz, first suggested by Cahn and JH [2]. This assumes that the solid-solid interfaces (lamella boundaries) always remain perpendicular to the local average solid-liquid front orientation.

They find that the lamellar system is Eckhaus unstable when its wavelength $\tilde{\lambda} < \hat{\lambda}_{\text{min}}$, in agreement with the following simple qualitative argument [2]: a convex front deformation (bump) increases the local $\tilde{\lambda}$, if $\tilde{\lambda} \ll \hat{\lambda}_{\text{min}}$ (resp. $\tilde{\lambda} \gg \hat{\lambda}_{\text{min}}$) the local undercooling decreases (resp. increases), thus amplifying (resp. reducing) the deformation. They also predict an oscillatory instability at twice the basic wavelength for sufficiently off-eutectic systems in a small thermal gradient.

We have shown in [1] that the small front deformation approximation, used to compute the diffusion field, is justified only in the large gradient limit, i.e. for $\ell_T \ll \ell$, where the thermal length $\ell_T$ (Eq. (1.11)) is inversely proportional to the thermal gradient $G$. Now, since it is the thermal field which primarily controls front deformations, it can be suspected that, the larger $G$ (the more valid (i)), the more difficult front orientational adjustment, and thus the more doubtful ansatz (ii).

In the order clear up this point, in this paper we study the local stability of the large gradient stationary solutions calculated in [1] with respect to deformations of wavelength $2\pi/k$ much larger than the basic one $\tilde{\lambda}$ $(k\tilde{\lambda} \ll 1)$ assumed to be of the same order of magnitude as $\hat{\lambda}_{\text{min}}$. This ansatz-free calculation is performed with the help of an approximation which extends to the non-stationary system that justified for the stationary one. Namely, the diffusion field is approximated by that of the « locally-averaged » front (i.e. only profile modulations on the scale of $k^{-1}$ are retained).

In this approximation, three branches of modes appear, one of which corresponds to phase diffusion. We find that lamellar patterns at large $G$ are Eckhaus-stable in the whole range of $\tilde{\lambda} (O(\hat{\lambda}_{\text{min}}))$ covered by our calculation, in contradiction with the prediction based on the orientational adjustment ansatz.
The other two modes are, at small $\hat{\ell}$, mixtures of optical phase motions (i.e. variations of the local phase fraction) and average front profile modulations. One is always stable, while the second one may be unstable, at small enough values of the basic pattern wavelength $\hat{\lambda}$, if the system is sufficiently off-eutectic. This only occurs for a departure from eutectic composition of a given sign, defined by the asymmetry of the phase diagram of the mixture.

2. Formulation of the dynamical problem.

The equations describing the dynamics of the front profile of a growing lamellar eutectic have been written in [I]. We have shown that, since front deformation amplitudes $(\zeta - \bar{\zeta})$, are small (due to the small value of $\lambda/\ell$) it is legitimate to linearize the expression of the concentration field in $(\zeta - \bar{\zeta})$. These equations read:

$$- d_\alpha \kappa (x, t) - \frac{1}{\ell_{\beta \alpha}} \zeta (x, t) = u [x, \zeta (x, t), t] \quad (\alpha-L \text{ regions}) \quad (1a)$$

$$- d_\beta \kappa (x, t) - \frac{1}{\ell_{\beta \beta}} \zeta (x, t) = - u [x, \zeta (x, t), t] \quad (\beta-L \text{ regions}) \quad (1b)$$

where $u$ is the dimensionless departure from eutectic concentration, and:

$$u(x, \zeta (x, t), t) = u_\infty + \int_{-\infty}^{\infty} \frac{d\xi}{2\pi} e^{i\xi x} \{ \Pi(Q) [2 \Delta(Q, t) +$$

$$+ \int_{-\infty}^{\infty} \frac{d\xi}{2\pi} [2 \zeta(q, t) + \xi(q, t)] \Delta(Q - q, t) \} - [1 - \Pi(Q)] \int_{-\infty}^{\infty} \frac{d\xi}{2\pi} \frac{K_0(q)}{2\pi} \zeta(Q - q, t) \Delta(q, t) \}.$$  (2)

All the notations are those defined in [I]. In particular lengths are measured in units if the diffusion length.

Equations (1) and (2) must be supplemented with boundary conditions at the triple points of abscissae $x_{n1}(t)$ ($\beta \rightarrow \alpha$ contact) and $x_{n2}(t)$ ($\alpha \rightarrow \beta$ contact) (see Fig. 1):

- continuity of $\zeta$
- front equilibrium:

$$\sigma_{\alpha L} + \sigma_{\beta L} + \sigma_{\alpha \beta} = 0 \quad (3)$$

where the surface tension forces point out of the triple point.

We expand the front profile about the stationary one, $\zeta_0(x)$, as:

$$\zeta (x, t) = \zeta_0(x) + Z(x, t) \quad (4)$$

$$x_{n1}(t) = n\lambda + X_{n1}(t) \quad (5a)$$

$$X_{n2}(t) = (n + \eta) \lambda + X_{n2}(t) \quad (5b)$$

From the Floquet-Bloch theorem, we know that the eigenmodes that we are looking for are of form:

$$Z_k(x, t) = e^{\omega t} e^{ikx} z_k(x) \quad (6)$$
where \( z_k(x) \) is a periodic function with the period, \( \lambda \), of the basic stationary pattern. This implies that the corresponding lateral displacements of the triple points have the form:

\[
X_{n1}(t) = X_1 e^{ikx} e^{\omega t} \\
X_{n2}(t) = X_2 e^{ik(n + \eta)\lambda} e^{\omega t}.
\]

(7a) \hspace{1cm} (7b)

The geometrical factor \( \Delta(x, t) \) of which \( \Delta(\Phi, t) \) in equation (2) is the Fourier transform is given by:

\[
\Delta(x, t) = \sum_{n = -\infty}^{\infty} \left[ \delta \left( x - x_{n1}(t) \right) \theta \left( x_{n2}(t) - x \right) - (1 - \delta) \theta \left( x - x_{n2}(t) \right) \theta \left( x_{n+1,1}(t) - x \right) \right] \tag{8}
\]

where \( \theta \) is the usual step function and \( \delta = \Delta C_a/\Delta C \) (see Eq. (2) of [II]). So to first order in the deviations from the stationary pattern:

\[
\Delta(x, t) - \Delta_0(x) = \Delta^{(1)}(x, t) = \sum_{n = -\infty}^{\infty} \left[ - \delta \left( x - n\lambda \right) X_{n1}(t) + \delta \left( x - (n + 1)\lambda \right) X_{n2}(t) \right] \tag{9}
\]

where \( \Delta_0(x) \) corresponds to the basic periodic pattern.

We have checked in [I] that, in the large gradient limit, up to corrections of order \( \ell_T = \ell_T/\ell \), the concentration on the front can be approximated by that corresponding to the planar average \( \bar{\zeta} \) of the true profile. The profile deformation (6) has Fourier components at \( q = k \) and \( k + nK \) \( (n \neq 0) \), where \( K = 2\pi/\lambda \gg 1 \). Since we only consider here long wavelength modes with \( k \ll 1 \) and in order to be consistent with the stationary case, we only retain, when calculating the first order variation of \( u(x, \zeta(x, t), t) \), the \( q = k \) component of the front deformation. That is, we set, in equation (2)

\[
\bar{\zeta}(x, t) \approx \bar{\zeta} + Z e^{ikx + \omega t}. \tag{10}
\]

Linearizing equation (2) we find, with the help of (9), (10), for the concentration variation:

\[
u^{(1)}(x) = \left[ u(x, \zeta(x, t), t) - u_0(x) \right] e^{-\omega t} \\
= e^{ikx} \left[ - X_1 A_1(x) + X_2 A_2(x) + Z(\omega F(x) + G(x)) \right]. \tag{11}
\]
Expressions (11-12) must then be plugged into the Gibbs-Thomson equations (1). In the large-$G$ limit that we are considering, these could then be solved by the asymptotic matching procedure used in [I]. However, in order to simplify somewhat the rather heavy forthcoming algebra, we assume that the solid-liquid surface tensions $\sigma_{SL}$, $\sigma_{S}$ are much larger than the solid-solid one, $\sigma_{S}$, so that contact angles at the triple points (and thus profile slopes everywhere) are small enough for the curvatures $\kappa_{a}$ to be linearized:

$$\kappa(x, t) = -\xi_0^a(x) - Z^a(x, t).$$

That this is a physically unessential assumption can be inferred from the solution of the stationary problem.

The solution of the fully linearized version of equations (1) can be written, in the front region, $0 < x < \lambda$:

--- \alpha lamella ($0 < x < \eta \lambda$):

$$z_{a}(x) = e^{-ikx} \left[ \mu_{a} \left( q_{a} \left( x - \frac{\eta \lambda}{2} \right) \right) + \nu_{a} \left( q_{a} \left( x - \frac{\eta \lambda}{2} \right) \right) \right] + Y_{a}(x)$$

with:

$$Y_{a}(x) = (q_{a} d_{a})^{-1} \int_{\eta \lambda / 2}^{x} dx' \left[ q_{a} \left( x - x' \right) \right] u^{(1)}(x')$$

--- \beta lamella ($\eta \lambda < x < \lambda$):

$$z_{\beta}(x) = e^{-ikx} \left[ \mu_{\beta} \left( q_{\beta} \left( x - \frac{(1 + \eta) \lambda}{2} \right) \right) + \nu_{\beta} \left( q_{\beta} \left( x - \frac{(1 + \eta) \lambda}{2} \right) \right) \right] + Y_{\beta}(x)$$

$$Y_{\beta}(x) = \left( q_{\beta} d_{\beta} \right)^{-1} \int_{(1 + \eta) \Lambda / 2}^{x} dx' \left[ q_{\beta} \left( x - x' \right) \right] u^{(1)}(x')$$
where:

\[ q_i^{-1} = (\ell_i^2/d_i)^{1/2} \quad (i = \alpha, \beta) \]  

(16)

The deformed front profile \( \zeta(x, t) \) must, moreover, satisfy the boundary conditions at the triple points which we also linearize in \( Z(x, t), X_{n1}(t), X_{n2}(t) \).

2.1 CONTINUITY OF THE PROFILE. — It reads, for example, at the \( \alpha \rightarrow \beta \) triple point \( x_{02}(t) \)

\[ \zeta_\alpha(\eta \lambda + X_{02}(t), t) = \zeta_\beta(\eta \lambda + X_{02}(t), t) \]  

(17)

Linearizing equation (17) and using \( \zeta_{0\alpha}' - \theta_{\alpha} = -\tan \theta_{\alpha}, \zeta_{0\beta}' = \theta_{\beta} \), this becomes:

\[ z_{k\alpha}(\eta \lambda) - z_{k\beta}(\eta \lambda) = (\theta_{\alpha} + \theta_{\beta}) X_2 \]  

(18a)

Similarly, continuity at the \( \beta \rightarrow \alpha \) triple point yields:

\[ z_{k\alpha}(0) - z_{k\beta}(0) = - (\theta_{\alpha} + \theta_{\beta}) X_1 \]  

(18b)

\( \theta_{\alpha}, \theta_{\beta} \) are the contact angles for the stationary front (see Fig. 2 of [1]).

2.2 EQUILIBRIUM CONDITIONS. — Since the triple points abscissae \( x_{ni} \) are time-dependent, the solid-solid contact lines make an angle \( \phi_{ni}(t) \) with the pulling direction \( Oz \) (see Fig. 1), given by:

\[ \tan \phi_{ni} \equiv \phi_{ni} = \frac{dx_{ni}}{dt} = \omega X_{ni}(t) \]  

(19)

The equilibrium condition (3) imposes that the angles between the three interfaces at each triple point are fixed, so that the variations of the contact angles \( \delta \theta_{ni} \), measured from the \( x \)-axis (Fig. 1), satisfy:

\[ \delta \theta_{n1}^{(\alpha)} = - \delta \theta_{n1}^{(\beta)} = - \omega X_{n1} \]  

\[ \delta \theta_{n2}^{(\alpha)} = - \delta \theta_{n2}^{(\beta)} = - \omega X_{n2} \]  

(20)

The slope condition on, for example, the \( \alpha \) side of the \( x_{01} \) triple point, which reads:

\[ \frac{\partial \zeta_\alpha}{\partial X} \bigg|_{x_{01}(t), t} = \theta_{01}^{(\alpha)}(t) \]  

(21)

is easily linearized into:

\[ [\omega + \zeta_{0\alpha}'(0)] X_1 + [ikz_{k\alpha}(0) + z_{k\alpha}'(0)] = 0 \]  

(22a)

Similarly, we get from the other three slope conditions:

\[ [\omega + \zeta_{0\beta}'(\lambda)] X_1 + [ikz_{k\beta}(\lambda) + z_{k\beta}'(\lambda)] = 0 \]  

(22b)

\[ [\omega + \zeta_{0\alpha}'(\eta \lambda)] X_2 + [ikz_{k\alpha}(\eta \lambda) + z_{k\alpha}'(\eta \lambda)] = 0 \]  

(22c)

\[ [\omega + \zeta_{0\beta}'(\eta \lambda)] X_2 + [ikz_{k\beta}(\eta \lambda) + z_{k\beta}'(\eta \lambda)] = 0 \]  

(22d)

The primes indicate space derivatives.
Finally, the $k$-Fourier coefficient of the front profile (14a), (15a) must satisfy the self-consistency condition:

$$\lambda Z = \int_{0}^{\eta/\lambda} z_{ka}(x) \, dx + \int_{\eta/\lambda}^{\Lambda} z_{k\beta}(x) \, dx.$$  \hspace{1cm} (23)

We then solve equations (22a-d) for the four integration constants $\mu_i$, $\nu_i$, and plug the resulting values into the continuity conditions (18) and the self-consistency equation (23). This results into three equations which are homogeneous and linear in the three dynamical variables $X_1$, $X_2$, $Z$. Each of these equations is linear in $\omega$. The solvability condition of this system yields the dispersion equation of the lamellar pattern. The fact that this spectrum contains only three branches of modes results from the approximation we use to calculate the diffusion field: the only degrees of freedom retained in this small-$k$ approximation, are the positions of the triple points and the local average (on a basic period) of the profile height. That is, we cannot describe modes which would involve profile deformations on a scale $\ll \tilde{\Lambda}$. These are likely to be strongly relaxing in the large gradient limit which we study here.

Writing down the three above-mentioned equations is totally straightforward, but their final detailed form is exceedingly heavy, due to the large number of material parameters ($q_\alpha$, $q_\beta$, $\ell_T \alpha$, $\ell_T \beta$). In order to simplify somewhat the resulting algebra, we assume from now on that our eutectic system is what we call « pseudo-symmetric ». Namely we set:

$$\sigma_{aL} = \sigma_{bL}; \quad m_a = m_\beta$$  \hspace{1cm} (24a)

or, equivalently:

$$\ell_T \alpha = \ell_T \beta = \ell_T; \quad q_a = q_\beta = q = (\ell_T d)^{1/2}; \quad \theta_a = \theta_\beta = \theta$$  \hspace{1cm} (24b)

$d$ is the (common) capillary length.

The only remaining asymmetry is that of the concentration gaps, reflected in the a priori finite parameter $(\delta - 1/2)$.

Taking advantage of the fact that in the large gradient limit, and for $\lambda$ values of the same order of magnitude, $d^{1/2}$, as $\lambda_{min}$, $q\lambda \gg 1$, we neglect terms $0[\exp - \eta q\lambda]$ and $0[\exp - (1 - \eta) q\lambda]$. The zeroth-order curvatures $\xi_0^\alpha$, $\xi_0^\beta$ are readily obtained from the fully linearized version of the results of (1) (e.g. Eqs. (1.9) where $K = -\zeta_0^\alpha$). For example:

$$\xi_0^\alpha(0) = -q \theta \coth \left( \frac{\eta \lambda q}{2} \right) - \left[ d \sinh \left( \frac{\eta \lambda q}{2} \right) \right]^{-1} \int_0^{\pi \lambda/2} dx' \sinh q \left( \frac{\eta \lambda}{2} - x' \right) \frac{d\phi(x')}{dx'}$$  \hspace{1cm} (25a)

where:

$$\phi(x) = \lambda \sum_{p=1}^{\infty} \frac{\sin (\eta p \pi) \cos (Kx - \eta \pi)}{\pi^2 p^2}.$$  \hspace{1cm} (25b)

We then obtain the $3 \times 3$ system:

$$\begin{pmatrix} \alpha_{11} & \alpha_{12} & \alpha_{13} \\ \alpha_{21} & \alpha_{22} & \alpha_{23} \\ \alpha_{31} & \alpha_{32} & \alpha_{33} \end{pmatrix} \begin{pmatrix} X_{k+} \\ X_{k-} \\ Z \end{pmatrix} = 0$$  \hspace{1cm} (26)

where:

$$X_{kz} = X_1 \pm X_2 e^{ik\eta\lambda}.$$  \hspace{1cm} (27)
The first two equations of (26) are obtained by respectively adding and substracting equations (18a) and (18b). Then:

\[
\alpha_{11}(k, \omega) = 2 \omega + \frac{8}{\lambda d} \sum_{m = -\infty}^{\infty} \left[ \frac{\Pi(k + mK)}{q^2 + (k + mK)^2} \sin^2 \frac{\eta \lambda (k + mK)}{2} - \frac{\Pi(mK)}{q^2 + m^2 K^2} \sin^2 \left( \eta \pi m \right) \right]
\]

(28)

\[
\alpha_{12}(k, \omega) = -\alpha_{21}(k, \omega) = -\frac{4i}{\lambda d} \sum_{m = -\infty}^{\infty} \frac{\Pi(k + mK)}{q^2 + (k + mK)^2} \sin \left[ (k + mK) \eta \lambda \right]
\]

(29)

\[
\alpha_{13}(k, \omega) = 4i \sum_{m = -\infty}^{\infty} \left[ S_m(k) - S_m(0) \right]
\]

(30a)

\[
S_m(k) = \frac{\Pi(k + mK)}{q^2 + (k + mK)^2} \sin \left( \frac{(k + mK) \eta \lambda}{2} \right) e^{ik(k + mK)/2} \Delta_m \left[ 2 \omega + 1 - \frac{K_0(mK)}{K_0(k + mK)} \right]
\]

(30b)

\[
\alpha_{31}(k, \omega) = \frac{\omega + \zeta''(0)}{q^2 + k^2} \left[ e^{-ik(q^2 + k^2)} \left( 1 + \frac{ik}{q} \right) \right] - 1 + \frac{ik}{q} + \frac{\omega + \zeta''(0)}{q^2 + k^2} \left[ 1 + \frac{ik}{q} + e^{-ik(q^2 + k^2)} \left( -1 + \frac{ik}{q} \right) \right] + \frac{2i}{d} \left( 1 - 2 \eta \right) \frac{\Pi(k)}{q^2 + k^2} e^{-ik(q^2 + k^2) / 2} \sin \left( k \eta \lambda / 2 \right)
\]

\[
- \frac{8i e^{-ik(q^2 + k^2) / 2}}{\lambda d(q^2 + k^2)} \sum_{m = -\infty}^{\infty} \left[ \frac{1}{mK} - \frac{k}{q^2 + (k + mK)^2} \right] \sin \eta \pi m \sin \left( \frac{(k + mK) \eta \lambda}{2} \right) \frac{\eta \lambda}{\Pi(k + mK)}
\]

(31)

We will see below that we do not need, when studying the long wavelength spectrum, the full expressions of the four other \( \alpha \)'s, but only their \( k = 0 \) limit, which reads:

\[
\alpha_{22}(\omega, 0) = 2 \omega + \frac{4}{dq} \left[ 1 + \frac{2}{dq} P''(\eta) \right]
\]

(32)

\[
\alpha_{23}(\omega, 0) = -\frac{2\omega}{dq} \left[ 2(\eta + \delta - 1) + \lambda P'(\eta) \right]
\]

(33)

\[
\alpha_{32}(\omega, 0) = -2 \ell_T \left[ \eta - \frac{1}{2} + \frac{\lambda}{2} P'(\eta) \right]
\]

(34)

\[
\alpha_{33}(\omega, 0) = \lambda \left[ 1 + 2 \omega \ell_T \left\{ \left( \eta - \frac{1}{2} \right) (\eta + \delta - 1) + \lambda P(\eta) \right\} \right]
\]

(35)

where:

\[
P(\eta) = \sum_{m = 1}^{\infty} \frac{\sin^2 \left( \eta \pi m \right)}{\pi^3 m^3}.
\]

(36)

3. Discussion.

As discussed above, our approximation (Eq. (10)) for the diffusion field restricts our analysis to long wavelength modes \( (k \lambda \ll 1) \). So, we will first investigate the \( k = 0 \) limit of the spectrum.
1) \( k = 0 \) modes. — Noticing that \( \Pi \) (Eq. (12f)) is an even function, it is immediately seen on equations (28)-(31) that:

\[
\begin{align*}
\alpha_{11}(0, \omega) &= 2\omega \\
\alpha_{12}(0, \omega) &= \alpha_{13}(0, \omega) = \alpha_{21}(0, \omega) = \alpha_{31}(0, \omega) = 0.
\end{align*}
\]

(37)

The \( k = 0 \) spectrum therefore decomposes, as expected, into:

(i) a neutral mode with \( X_1 = X_2, Z = 0 \), i.e. uniform translation of the basic pattern along the \( x \)-direction;

(ii) two modes with \( X_1 = -X_2 \) and frequencies given by:

\[
(\alpha_{22} - \alpha_{33} - \alpha_{23} \alpha_{32})_{0, \omega} = 0.
\]

(38)

These modes therefore mix front profile modulations and « optical » phase motions in which two consecutive triple points move out of phase, i.e. variations of the phase fraction \( \eta \).

Equation (38) can be written, with the help of equations (32)-(35):

\[
a \omega^2 + b \omega + c = 0
\]

(39)

where:

\[
\begin{align*}
a &= \lambda \left[ P(\eta) + \frac{P'(\eta)}{2} \left( \frac{1}{2} - \eta \right) \right] - u_\infty \left( \eta - \frac{1}{2} \right) \\
b &= \lambda \left[ \frac{qd}{2T} + 2P(\eta) + \left( \frac{1}{2} - \eta \right) P'(\eta) + O(u_\infty) + O(\lambda) \right] \\
c &= \frac{1}{T} \left[ 1 + O(\lambda) \right]
\end{align*}
\]

(40a, 40b, 40c)

since \( |u_\infty|, \lambda \ll 1, c \gg 0 \).

We have shown in [1] that \( U(\eta) = P(\eta) - \frac{\eta}{2} P'(\eta) \) is positive whatever \( \eta \) \((0 < \eta < 1)\). Since \( P(\eta) = P(1-\eta) \), this entails that:

\[
2P(\eta) + \left( \frac{1}{2} - \eta \right) P'(\eta) \equiv U(\eta) + U(1-\eta) > 0
\]

(41)

and that \( b > 0 \).

Finally, the compatibility condition (Eq. (40)) for the stationary pattern reads, for our pseudo-symmetric model:

\[
\eta - \frac{1}{2} = \frac{1}{2} - \delta - u_\infty - \frac{\lambda}{2} P'(\eta).
\]

(42a)

For a finite asymmetry of the eutectic concentration gaps \( \Delta C_{\alpha,\beta} \) (finite \( \left( \frac{1}{2} - \delta \right) \)) this reduces to:

\[
\eta - \frac{1}{2} \approx \frac{1}{2} - \delta
\]

(42b)

and it is seen on equation (40a) that two cases may arise:

— if \( u_\infty \left( \delta - \frac{1}{2} \right) > 0 \)
a is always positive. Then the two roots of equation (39) have negative real parts. Both the
non-neutral modes are relaxing (with or without oscillations, depending on the values of \( \lambda \),
\( u_{\infty}, \delta, ... \))

\[
u_{\infty}\left(\delta - \frac{1}{2}\right) < 0
\]

\( a \) is negative for stationary patterns with wavelengths:

\[
\lambda < \lambda_0 = 2u_{\infty}\left(\frac{1}{2} - \delta\right)[U(\delta) + U(1 - \delta)]^{-1}.
\]  (43)

In this case, one of the roots of (39) is positive, the stationary pattern is unstable.

So we find that the near-eutectic pseudo-symmetric system exhibits an asymmetric
behavior, depending on its concentration.

If its representative point in the phase diagram (see Fig. 2) is on the side of the eutectic
point corresponding to the smaller concentration gap, it is stable, whatever \( \lambda \), with respect to
uniform variations of \( \eta \) and of the average undercooling.

If, on the contrary it is on the larger gap side, only those stationary patterns with
\( \lambda > \lambda_0 \) are stable with respect to these deformations.

The question then naturally arises of whether and how such an asymmetric behavior is
modified when one relaxes the pseudo-symmetric approximation and deals with the more
realistic general asymmetric model \((\sigma_\alpha \neq \sigma_\beta, m_\alpha \neq m_\beta)\). In the \( k = 0 \) limit, this
generalization, although heavy, is relatively easy to perform. We find that the instability
condition (43) extends into:

\[
\lambda < \lambda_0 = u_{\infty}[\ell_{T\beta}(1 - \delta) - \ell_{Ta}\delta][\ell_{Ta} U(1 - \eta) + \ell_{T\beta} U(\eta)]^{-1}.
\]  (44)

So, the asymmetry parameter which decides on the existence of a stability threshold
\( \lambda_0 \) for the basic pattern is now:

\[
u_{\infty}[m_\beta \Delta C_\beta - m_\alpha \Delta C_\alpha] = u_{\infty}(\Delta T_\beta - \Delta T_\alpha)
\]

where \( \Delta T_\alpha, \beta \) are defined in figure 2. So the \( k = 0 \) instability only occurs when the
representative point A (Fig. 2) is on the side of \( C_E \) with the larger \( \Delta T \).

Of course, since the two modes, studied here, have finite relaxation rates for
\( k = 0 \), it is clear that these stability results extend to long but finite wavelengths.

Fig. 2. — Phase diagram of eutectic alloy. The representative point A corresponds to the imposed
concentration \( C_\alpha \).
2) Phase diffusion coefficient. — We are left with studying, for the pseudo-symmetric model, the behavior of the acoustical phase mode which continues at small but finite \( k \) the uniform neutral one (i). At finite \( k \), the full dispersion equation is given by:

\[
\text{Det} (k, \omega) = \sum_{r=0}^{3} \omega^r a_r(k) = 0
\]

where \( \text{Det} \) is the determinant of the \( 3 \times 3 \{a_{ij}\} \) matrix and, as seen above, \( a_0(0) = 0 \).

The relaxation rate \( \omega_{\text{a.p.}}(k) \) of the acoustical phase mode is therefore given, to lowest order in \( k \), by:

\[
\omega_{\text{a.p.}}(k) = - \frac{a_0(k)}{a_1(0)} = - \frac{\text{Det} (k, 0)}{\left[ \frac{d}{d\omega} \text{Det} (0, \omega) \right]_{\omega = 0}}
\]

where \( \text{Det} (k, 0) \) itself must be calculated to lowest order in \( k \).

It is immediately found that:

\[
\left[ \frac{d}{d\omega} \text{Det} (0, \omega) \right]_{\omega = 0} = 2[\alpha_{22} 33 - \alpha_{23} \alpha_{32}]_{0, 0} = \frac{8}{qd} \left[ 1 + \frac{\lambda}{2} P''(\eta) \right].
\]

Expanding in powers of \( k \) the expressions (28)-(31) for the \( \alpha_{ij}(k, 0) \) and \( \alpha_{1i}(k, 0) \), and neglecting corrections \( O(q\lambda) = O(k^{1/2}) \) we find:

\[
\begin{align*}
\alpha_{11}(k, 0) &\approx k^2 \ell_T q \lambda \left\{ \eta^2 + \frac{\lambda}{2} \left[ \eta^2 P''(\eta) - 2\eta P'(\eta) + 2P(\eta) \right] \right\} \\
\alpha_{12} &\approx -\alpha_{21}(k, 0) = -ikq\ell_T \{2 \eta + \lambda [\eta P''(\eta) - P'(\eta)] \} \\
\alpha_{13}(k, 0) &\approx -ikq\ell_T \lambda^2 P(\eta) \\
\alpha_{31}(k, 0) &\approx -ik\ell_T \left\{ \lambda \eta \left( \eta - \frac{1}{2} \right) + 2d\theta - \lambda^2 \left[ P(\eta) - \eta P'(\eta) \right] \right\}.
\end{align*}
\]

So, \( \text{Det} (k, 0) = O(k^2) \), and, to this order, one must use the \( k = 0 \) values (Eqs. (32)-(35)) of the remaining \( \alpha_{ij} \)'s.

Using expression (52) of [1] for the minimum undercooling wavelength, namely:

\[
\lambda_{\text{min}}^2 = 4d\theta \left[ 2P(\eta) + \left( \frac{1}{2} - \eta \right) P'(\eta) \right]^{-1}
\]

we then find that the relaxation rate of the acoustical phase mode is given by:

\[
\omega_{\text{a.p.}}(k) = -D(\lambda) k^2
\]

where the phase diffusion coefficient:

\[
D(\lambda) = \frac{\lambda^2}{2} \sqrt{\frac{\ell_T}{d}} P(\eta) \left[ 1 + \frac{2\theta}{\lambda q^2} \left( 1 - \frac{\lambda^2}{\lambda_{\text{min}}^2} \right) \right].
\]

Since we consider basic patterns such that \( \lambda / \lambda_{\text{min}} = O(1) \), the second term in the square bracket in equation (51) is of order \( (\lambda q^2)^{-1} \sim d^{1/2} \ell_T \ll 1 \), so that:

\[
D(\lambda) = \frac{\lambda^2}{2} \sqrt{\frac{\ell_T}{d}} P(\eta) > 0.
\]
That is, in the range of wavelengths of physical interest, all periodic lamellar patterns are Eckhaus-stable at large thermal gradients. The only possible long wavelength instability, in this regime, is the « optical phase » one discussed above.

Our result about the phase diffusion coefficient contradicts the Datye-Langer [9] prediction that patterns with \( \lambda < \lambda_{\text{min}} \) should be Eckhaus unstable. This clearly means that, in the only parameter region — namely large thermal gradients — where the diffusion field can be calculated in the average planar front approximation, Cahn’s dynamical ansatz about lamellar orientation is not valid.

As we discussed in [1], unfortunately, the large gradient limit in which the above analytic treatment is justified is restricted to values of the gradient \( G \) which are difficult to reach experimentally. So, there is an obvious need for an extension of our calculations to lower \( G \)’s.

It is reasonable to expect that, the smaller \( G \) (i.e. the less confined front deformations), the better the dynamical ansatz should become. However, it is clear that, at the same time, the less justified it is to use the planar front approximation for the diffusion field. In this perspective numerical studies of both the stationary front profile and of its stability are clearly necessary.

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References