Directional solidification of a faceted crystal. II. Phase dynamics of crenellated front patterns
B. Caroli, C. Caroli, B. Roulet

To cite this version:
B. Caroli, C. Caroli, B. Roulet. Directional solidification of a faceted crystal. II. Phase dynamics of crenellated front patterns. Journal de Physique, 1989, 50 (20), pp.3075-3087. <10.1051/jphys:0198900500200307500>. <jpa-00211126>

HAL Id: jpa-00211126
https://hal.archives-ouvertes.fr/jpa-00211126
Submitted on 1 Jan 1989

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
**Directional solidification of a faceted crystal.**

**II. Phase dynamics of crenellated front patterns**

B. Caroli (*), C. Caroli and B. Roulet

Groupe de Physique des Solides de l'Ecole Normale Supérieure, associé au C.N.R.S., Tour 23, Université Paris VII, 2 place Jussieu, 75251 Paris Cedex 05, France

(Reçu le 4 avril 1989, accepté le 19 juin 1989)

Résumé. — Nous faisons une analyse de stabilité linéaire des fronts périodiques crénelés qui définissent un seuil d'amplitude pour l'instabilité cellulaire d'un front de solidification directionnelle faceté [1]. Nous étudions la limite où les facettes sont beaucoup plus longues que les parties courbes du front (contremarches). Le front crénelé est stable vis-à-vis de la diffusion de phase. Alors que les variations d'amplitude sont essentiellement contrôlées par la cinétique de facette, la mobilité latérale des contremarches est limitée par la dynamique du champ de diffusion solutale.

Abstract. — We perform a linear stability analysis of the periodic crenellated front profiles which provide an amplitude threshold for the cellular instability of a faceted directional solidification front [1]. This analysis assumes that facets are much longer than curved parts of the front (risers). We show that, while the crenellated front is stable as regards phase diffusion, it always has a branch of unstable modes which are a mixture of amplitude and phase fluctuations. While amplitude variations are primarily controlled by facet kinetics, the lateral riser mobility is limited by the solute diffusion field dynamics.

1. Introduction.

In a recent article [1] hereafter referred to as (I), the stability of a planar directional solidification front corresponding to a facet orientation of the growing solid has been studied. It has been shown that, when the pulling velocity $V$ is larger than the standard Mullins-Sekerka (MS) instability threshold, there exists, for each $V$, a continuum of stationary non-planar periodic « crenellated » solutions for the front profile. Each such profile consists of an alternation of hot and cold facets, connected by curved parts which, following Chernov and Nishigana [2], we will call « risers ».

These results are obtained under three assumptions:

(i) the equilibrium shape of the crystal is only partly faceted, and there are no missing orientations between the relevant facet and the contiguous rough regions;
(ii) the wavelength $\lambda$ of the crenellated profile is much smaller than the diffusion length $\ell = 2 D / V$, where $D$ is the chemical diffusion coefficient in the (binary) liquid phase;
(iii) the deformation amplitude of the profile is small enough for linearization to be legitimate.

The $x$-extension, $c$ (see Fig. 1) of each curved part is, for a given $V$, independent of $\lambda$. It is simply equal to half the wavelength, $\pi / q$, of the neutral MS mode with largest wavevector. The value of $\lambda$ determines the lengths and heights of the faceted parts.

It was also shown that, when the facets are much longer than the risers, these stationary profiles are unstable against amplitude fluctuations, which entails that they define an amplitude threshold for the cellular instability.

Fig. 1. — Full line: stationary periodic crenellated front profile. Dashed line: displaced crenellated profile.

However, this stability analysis was performed under the restrictive assumption that phase modulations are not coupled with amplitude fluctuations: namely, it was assumed that a small change in the relative height of hot and cold facets does not modify the lengths of the facets.

Our aim here is to perform a full linear stability analysis for the crenellated fronts, allowing not only for amplitude, but also for phase fluctuations, i.e., for lateral motion of the risers. This will be done in the same limit as in (I), namely $\lambda \gg c$ (facets much longer than risers), where the diffusion field associated with the stationary profile can be approximated by that of a perfect crenel ($c = 0$). The aim of this analysis is twofold:
— determine under which precise conditions the approximation used in (I) of decoupling between relative facet height and phase fluctuations is justified and, if necessary, improve upon it;
— study the dynamics of phase modes. These can be qualitatively classified into « optical » and « acoustical » one depending on whether, in these modes, two neighboring risers move with equal or opposite phases. It is clear that the scenario of evolution of a crenellated front will be very different depending on whether, for example, the optical phase modes are growing or relaxing. The question also arises of whether the Eckhaus instability (instability with respect to a slow modulation of the wavelength of the basic structure, i.e., of acoustical phase modes) survives in the presence of facets.
The main results which will emerge from this analysis are the following:
- the crenellated front has, at all values of the dissipation $F$, one branch of unstable modes, the « breathing » ones, which always involve a fluctuation of the relative heights of hot and cold facets. However, contrary to what was postulated in (I), this fluctuation is coupled with optical phase fluctuations and global front motion in the pulling direction. It is only in the absence of facet interface dissipation (i.e. of kinetic undercooling) that these motions decouple and that the growth rate of the unstable mode reduces to that calculated in (I);
- long wavelength acoustical phase modes do not couple to amplitude fluctuations, and are always stable, i.e. as can be qualitatively expected, long facets suppress the phase diffusion instability. Their relaxation rate is governed by the dynamics of the solute diffusion field only.

From these results, one may define qualitatively a diffusion-limited lateral mobility for a steep riser, which should be relevant to the question of macrostep motion. It is proportional to $[\ln \lambda/c]^{-1}$, and thus characteristic of the geometry of the riser.

2. Linearized equations of motion.

We describe the 2-D two phase system by the same model as in (I): one-sided chemical diffusion, equal thermal diffusivities, instantaneous thermal diffusion. We moreover take for the solute segregation coefficient $K = 1$ (constant concentration gap $(\Delta C)$ — a minor but simplifying assumption. As in (I) we only consider small amplitude crenellated profiles, so that the front integral equation can be linearized, and anticipate the quasi-static approximation [3] to be valid, as can be checked on the results.

The dynamical front equation for the front profile $Z(x, t)$ then reads (see Eq. (33) of (I)):

$$- \left\{ \gamma + \frac{d^2\gamma}{d\theta^2} \right\} \mathcal{K}(x, t) + (2 - \mu) Z(x, t) + F(V) - \mathcal{F} \left[ V \left\{ 1 + \frac{1}{2} \frac{\partial}{\partial \theta} \right\}, \theta(x, t) \right] = \mathcal{K}\{Z\}$$ (1)

with:

$$\mathcal{K}\{Z\} = \int_{-\infty}^{\infty} dx' P(x-x') \left[ Z(x', t) + \frac{1}{4} \hat{Z}(x', t) \right]$$ (2)

$Z$ is measured, along the $z$ axis parallel to the pulling direction, from the position of the infinite planar front. Lengths and times are measured in units of $\ell = 2 D/V$, $\tau = 4 D/V^2$. All notations are those of (I). Let us simply recall that the surface dissipation $\mathcal{F}$ is assumed to be temperature-independent and to be zero for all non-facet orientation ($\theta \neq 0$).

The diffusion kernel $P(x)$ appropriate to our one-sided model in the r.h.s. of equation (2) is the Fourier transform of:

$$P(k) = \frac{4}{1 + \sqrt{1 + k^2}}.$$ (3)

We now set:

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

where $\zeta(x)$ is that stationary crenellated solution of equation (1) which has space period $\lambda = 2 \pi/k$ ($\lambda \ll 1$ in our reduced units). The lengths and heights of its hot and cold facets are respectively $2a$, $\zeta_+$ and $2b$, $\zeta_-$. Its shape as well as the parameters defining the fluctuation $\delta \zeta(x, t)$ are depicted in figure 1.
As in (I), in order to make the linear stability problem amenable to analytical treatment, we limit ourselves to the case where facet lengths $2a, 2b$ are much larger than the $x$-extension $c = \pi /q$ of the risers. We can then safely neglect (up to corrections of order $c/\lambda \ll 1$) the width of the risers when computing the diffusion term $\tilde{\mathcal{K}}(\mathcal{Z})$. That is, in $\tilde{\mathcal{K}}(\mathcal{Z})$, we take:

$$
\zeta(x) = \sum_n \{ \zeta_+ \theta (x - n\lambda + a) \theta (n\lambda + a - x) + \zeta_- \theta (x - n\lambda - a) \theta (n\lambda + \lambda - a - x) \} \quad (5)
$$

where $\theta(x)$ is the step function. Analogously:

$$
\delta \zeta(x, t) = \sum_n \{ \delta \zeta_+(t) \theta (x - n\lambda + a) \theta (n\lambda + a - x) + 
\quad + \delta \zeta_-(t) \theta (x - n\lambda - a) \theta (n\lambda + \lambda - a - x)
\quad + \frac{1}{2} (\delta a_{n,1}(t) + \delta b_{n,1}(t))(\zeta_+ - \zeta_-) \delta (x - n\lambda - a)
\quad + \frac{1}{2} (\delta b_{n,2}(t) + \delta a_{n,2}(t))(\zeta_- - \zeta_+) \delta (x - n\lambda - \lambda + a) \} \quad (6)
$$

In equations (5), (6), since we neglect $c$ as compared with $a, b$ we have set $\lambda = 2(a + b)$. In equations (5), (6), $\delta a_{ni}, \delta b_{ni}$ do not appear separately, but only via the displacement $(\delta a_{ni} + \delta b_{ni})/2$ of the riser center. That is, naturally, our long facet approximation does not permit to calculate the modes associated with modulations $(\delta a_{ni} - \delta b_{ni})$ of rider widths. However, because we are precisely looking at the case where risers are well separated and « locked » by the facets, we expect these modes to be rapidly relaxing as compared with those studied here.

Taking advantage of Floquet's theorem, we now look for solutions of equation (1) of the form:

$$
\delta \zeta_+(t) = \delta \zeta_+ e^{ipn\lambda} e^{\omega t} = e^{ipn\lambda} \delta \zeta_+ (t) \quad (7a)
$$

$$
\delta \zeta_-(t) = \delta \zeta_- e^{ip(a + 1/2)\lambda} e^{\omega t} \quad (7b)
$$

$$
\begin{pmatrix}
\delta a_{n1}(t) \\
\delta b_{n1}(t)
\end{pmatrix} = 
\begin{pmatrix}
\alpha \\
\beta
\end{pmatrix} e^{ip(n\lambda + a)} e^{\omega t} \quad (7c)
$$

$$
\begin{pmatrix}
\delta a_{n2}(t) \\
\delta b_{n2}(t)
\end{pmatrix} = 
\begin{pmatrix}
\gamma \\
\delta
\end{pmatrix} e^{ip(n\lambda + \lambda - a)} e^{\omega t}. \quad (7d)
$$

We consider first the case where $x$ belongs to one of the (shifted) risers. We plug expressions (4) to (7) into equations (1), (2), linearize $\mathcal{Y}$ in $\delta \phi$ and subtract out the stationary front equations. The resulting $x$-integral in $\tilde{\mathcal{K}}$ decomposes into a sum of contributions coming from riser center lateral shifts and from hot and cold facet vertical displacements. Let us consider for example the hot facet contribution. It reads:

$$
\tilde{\mathcal{K}}_{h,t} = (4 \delta \zeta_+(t) + \delta \phi_+(t)) \left[ J_+ (x) + \frac{\sin (pa)}{2p\lambda} P(p) e^{ipx} \right] \quad (8)
$$

where :

$$
J_+ (x) = \frac{1}{2} \sum_{m=-\infty}^{\infty} (1 - \delta_{m0}) \frac{\sin (p + mk) a}{p + mk} P(p + mk) e^{(p + mk)x} \quad (9)
$$

The stationary solution $\zeta(x)$ was calculated in (I) in the highly non-local limit $\lambda \ll 1$ where the diffusion field ahead of the crenellated front can be approximated by that of its planar
average. In order to remain coherent with this calculation we must neglect here in equation (8) the contribution of all \( m \not= 0 \) Fourier components in the \( \delta \xi_+ \) term. It can be checked directly that this is negligible as compared with l.h.s. terms as soon as \( (2 - \mu) \gg \Gamma_0 \) (where \( \Gamma_0 = (\gamma + d^2 \gamma/d\theta^2)_{\theta = 0+} \)) which was precisely the condition of validity of the stationary calculation. Note that, on the contrary, this argument does not hold for \( \delta \xi_- \) terms (for example, in the absence of kinetics, the l.h.s. of Eq. (1) is independent of \( \delta \xi_- \)), in which we therefore retain all Fourier components.

Performing the same approximation on the cold facet and riser contributions, we find that, in riser regions, \( \delta \xi_+(x) = \delta \xi_+(x, t) e^{-\omega t} \) obeys the equation:

\[
\Gamma_0 \frac{d^2}{dx^2} \delta \xi_+(x) + (2 - \mu) \delta \xi_+(x) = \omega H(x) + \left(1 + \frac{\omega}{4}\right) U e^{ipx}
\]

with:

\[
U = \frac{P(p)}{\lambda} \left[ \frac{2 \sin pa}{p} \delta \xi_+ + \frac{2 \sin pb}{p} \delta \xi_- + \frac{(\xi_+ - \xi_-)}{2} (\alpha + \beta - \gamma - \delta) \right]
\]

and

\[
H(x) = \delta \xi_+ J_+(x) + \delta \xi_- J_-(x) + \frac{(\xi_+ - \xi_-)}{2} \left[ \frac{\alpha + \beta}{2} J_1(x) - \frac{\gamma + \delta}{2} J_2(x) \right]
\]

\( J_+(x) \) is defined in equation (9)

\[
J_-(x) = \frac{1}{2\lambda} \sum_{m=-\infty}^{\infty} (1 - \delta_{m0}) (-\gamma^m \sin (p + mk) b P(p + mk) e^{i(p + mk)x})
\]

\[
J_r(x) = \frac{1}{4\lambda} \sum_{m=-\infty}^{\infty} (1 - \delta_{m0}) \exp \left[-(\gamma^m \sin (p + mk) b P(p + mk) e^{i(p + mk)x} \right] \quad (r = 1, 2).
\]

As in (I), due to the \( \delta \)-function singularity in the surface stiffness \( (\gamma + d^2 \gamma/d\theta^2) \) associated with the facet-induced cusp on the Wulff plot of \( \gamma \), along the shifted facets this local equation must be replaced by global ones obtained by integrating equation (1) along each faceted region of the front separately [4]. These integrated equations express the fact that equilibrium on a facet is a global, and not a local, constraint (see (I)). Linearizing, again, about the corresponding stationary facet equation, we obtain the following two equations:

a) hot facets:

\[
a \delta \xi_+ \left[2(2 - \mu) - \omega VF' \right] + [(2 - \mu) \xi_+ - 2 \xi_0] (\alpha e^{ipa} - \delta e^{-ipa}) = \frac{2 \sin pa}{p} U + \omega \left[ \nu_a (a) \delta \xi_+ + \nu_b (a) \delta \xi_- + \frac{(\xi_+ - \xi_-)}{2} \{ \nu_1 (\alpha + \beta) - \nu_2 (\gamma + \delta) \} \right]
\]

b) cold facets:

\[
b \delta \xi_- \left[2(2 - \mu) - \omega VF' \right] + [(2 - \mu) \xi_- - 2 \xi_0] (\gamma e^{ipb} - \beta e^{-ipb}) = \frac{2 \sin pb}{p} U + \omega \left[ \nu_a \delta \xi_+ + \nu_b (b) \delta \xi_- + \frac{(\xi_+ - \xi_-)}{2} \{ \nu_3 (\alpha + \beta) - \nu_4 (\gamma + \delta) \} \right]
\]
where:

\[ F' = \frac{\partial F(V, 0)}{\partial V} \]  \hspace{1cm} (16)

\[ \nu_+ (a) = \frac{1}{\lambda} \sum_{m=-\infty}^{\infty} P(p + mk) \left[ \frac{\sin (p + mk) a}{p + mk} \right]^2 \]  \hspace{1cm} (17a)

\[ \nu_- = \frac{1}{\lambda} \sum_{m=-\infty}^{\infty} (-)^m P(p + mk) \frac{\sin (p + mk) a \sin (p + mk) b}{(p + mk)^2} \]  \hspace{1cm} (17b)

\[ \nu_1 = \nu_2^* = \frac{1}{2\lambda} \sum_{m=-\infty}^{\infty} e^{-im\alpha} P(p + mk) \frac{\sin (p + mk) a}{p + mk} \]  \hspace{1cm} (17c)

\[ \nu_1 = \nu_4^* = \frac{1}{2\lambda} \sum_{m=-\infty}^{\infty} (-)^m e^{-im\alpha} P(p + mk) \frac{\sin (p + mk) b}{p + mk} \]  \hspace{1cm} (17d)

\( \xi_0 \) is the average position of the stationary crenellated front.

Finally, equations (10), (14), (15) must be supplemented with matching conditions expressing that the deformed front is continuous and smooth (no missing orientations at the facet edges). Setting:

\[ \delta \zeta (x) = \delta \zeta_1 (x - n\lambda) e^{ip\lambda} \]  \hspace{1cm} (18a)

in the \( n \)-th downward-riser \( (n\lambda + a + \delta a_{n1} < x < n\lambda + a + c + \delta b_{n1}) \), and

\[ \delta \zeta (x) = \delta \zeta_2 (x - \lambda) e^{ip\lambda} \]  \hspace{1cm} (18b)

in the \( n \)-th upward one \( (n\lambda + \frac{\lambda}{2} + b + \delta b_{n2} < x < n\lambda + \frac{\lambda}{2} + b + c + \delta a_{n2}) \), to first order in the deformation parameters, these matching conditions read:

\[ \delta \zeta_1 (a) = \delta \zeta_2 \left( \frac{\lambda}{2} + b + c \right) e^{-ip\lambda} = \delta \zeta_+ \]  \hspace{1cm} (19a)

\[ \delta \zeta_1 (a + c) = \delta \zeta_2 \left( \frac{\lambda}{2} + b \right) = \delta \zeta_- e^{ip\lambda/2} \]  \hspace{1cm} (19b)

\[ \delta \zeta_1 '(a) - \alpha e^{ipa} \frac{(\zeta_+ - \zeta_-) q^2}{2} = 0 \]  \hspace{1cm} (20a)

\[ \delta \zeta_1 '(a + c) + \beta e^{ipa} \frac{(\zeta_+ - \zeta_-) q^2}{2} = 0 \]  \hspace{1cm} (20b)

\[ \delta \zeta_2 \left( \frac{\lambda}{2} + b \right) + \gamma e^{ip(a - \lambda)} \frac{(\zeta_+ - \zeta_-) q^2}{2} = 0 \]  \hspace{1cm} (20c)

\[ \delta \zeta_2 \left( \frac{\lambda}{2} + b + c \right) - \delta e^{ip(a - \lambda)} \frac{(\zeta_+ - \zeta_-) q^2}{2} = 0 \]  \hspace{1cm} (20d)

where we have made use of the fact that:

\[ \zeta'' (a) = \zeta'' \left( \frac{\lambda}{2} + b + c \right) = - \zeta'' (a + c) = - \zeta'' \left( \frac{\lambda}{2} + b \right) = - (\zeta_+ - \zeta_-) \frac{q^2}{2}. \]  \hspace{1cm} (21)

Note that, contrary to what was done above when computing \( \bar{\zeta} \), the riser width \( c \) cannot be neglected here since, at this stage, we are calculating (in an iterative fashion), the whole shape of the deformed front, including that of the risers themselves.
The solution of equation (10) in the downward riser can be written as:

$$\delta \xi_1(x) = A_1 \sin \left[ q \left( x - a - \frac{c}{2} \right) + \varphi_1 \right] + Y_1(x)$$  \hspace{1cm} (22)

where:

$$Y_1(x) = \left( 1 + \frac{\omega}{4} \right) \frac{U}{\Gamma_0 (q^2 - p^2)} e^{ipx} + \frac{\omega}{\Gamma_0 q} \int_x^{x'} dx' \sin \left[ q(x - x') \right] H(x')$$  \hspace{1cm} (23)

and we have used the relation (see (I)):

$$2 - \mu = \Gamma_0 q^2.$$  \hspace{1cm} (24)

Plugging expressions (22), (23) into the matching conditions (19, 20), and eliminating $A_1$, $\varphi_1$ one obtains:

$$Y_1(a) + Y_1(a + c) = \delta \xi_+ + \delta \xi_- e^{ip\lambda / 2}$$  \hspace{1cm} (25a)

$$Y'_1(a) + Y'_1(a + c) = \frac{\xi_+ - \xi_-}{2} q^2 (\alpha - \beta) e^{ip\alpha}.$$  \hspace{1cm} (25b)

Analogously, solving for the upward riser produces the two relations:

$$Y_2(\lambda - a) + Y_2(\lambda - a - c) = \delta \xi_+ + \delta \xi_- e^{ip\lambda / 2}$$  \hspace{1cm} (26a)

$$Y'_2(\lambda - a) + Y'_2(\lambda - a - c) = \left( \xi_+ - \xi_- \right) \frac{q^2}{2} (\delta - \gamma) e^{ip(\lambda - a)}.$$  \hspace{1cm} (26b)

where:

$$Y_2(x) = \left( 1 + \frac{\omega}{4} \right) \frac{U}{\Gamma_0 (q^2 - p^2)} e^{ipx} + \frac{\omega}{\Gamma_0 q} \int_x^{x'} dx' \sin q(x - x') H(x').$$  \hspace{1cm} (27)

That is, our eigenmode problem is determined by the closed set of six equations (14), (15), (25a, b), (25a, b) relating the six unknowns, $\delta \xi_\pm$, $\alpha$, $\beta$, $\gamma$, $\delta$.

However, as discussed above, our long facet approximation does not permit to treat consistently modulations of riser widths. So, in order to be coherent with our evaluation of the diffusion term $\mathcal{H}(x)$, i.e. of the r.h.s. of equations (14), (15) and $Y_1$, $Y_2$, we must discard equations (25b), (26b) and approximate $\alpha$, $\beta$ (resp. $\gamma$, $\delta$) by $(\alpha + \beta) / 2$ (resp. $(\gamma + \delta) / 2$) in the l.h.s. of equations (14), (15).

Note however that solving the complete sixfold system, although inconsistent, provides an estimate of the validity of the long facet approximation. We have checked that the corresponding shifts of the relaxation rates of the amplitude and phase modes which we will now calculate are, as expected, $O(c/\lambda)$.

We are therefore left with a $4 \times 4$ problem specified by equations (25a), (25b) and (14), (15) simplified as defined above, for the four unknowns $\delta \xi_\pm$, $\alpha + \beta$, $\gamma + \delta$.

3. Long wavelength eigenmodes.

3.1 Acoustical phase mode. — We are now interested in studying the long wavelength (small $p$) limit of the fluctuation spectrum. Among the four (amplitude and phase) modes which we calculate here one, which we term the acoustical phase mode, must be singled out.
since, for $p \to 0$, its tends towards pure lateral translation and thus becomes neutral. This means that the determinant $\Delta(\omega, p)$ built from equations (14, 15, 25a, 26a) can be written:

$$\Delta(\omega, p) = \sum_{r=0}^{4} \omega^r a_r(p)$$

(28)

with:

$$a_0(0) = 0.$$  

(29)

So, while the relaxation rate of the other three (optical phase and amplitude) modes can be obtained by simply computing $\Delta(\omega, p = 0)$, the acoustical phase mode one is given, at long wavelength, by:

$$\omega_{ap}(p) = -a_0(p)/a_1(0)$$

(30)

where $a_0(p)$ is to be evaluated to lowest order in $p$.

We are thus left simply with calculating the quantities $a_0(p) = \Delta(0, p)$ and $\Delta(\omega, 0)[a_1(0) = (d\Delta(\omega, 0)/d\omega)_{\omega=0}]$.

Taking the $\omega = 0$ limit of equations (14, 15, 25a, 26a) one immediately finds that:

$$a_0(p) = \mu(2 - \mu) ab(\xi_+ - \xi_-)^2 p^2$$

(31)

where we have made use of equation (24) and of the relation (see (1)):

$$(2 - \mu)(a\xi_+ + b\xi_-) = 2(a + b)\xi_0 = \lambda \xi_0.$$  

(32)

On the other hand, multiplying equations (25a) (resp. (26a)) by $e^{-ip\alpha}$, (resp. $e^{ip(a - \beta)}$) and adding up, one finds that $\Delta(\omega, 0)$ can be written as:

$$\Delta(\omega, 0) = \omega C \Delta^{(3)}(\omega)$$

(33)

with:

$$C = \frac{1}{\omega} \left\{ [Y_1(a) + Y_1(a + c)] e^{-ip\alpha} + [Y_2(\lambda - a) + Y_2(\lambda - a - c)] e^{ip(a - \beta)} \right\}_{p = 0} =$$

$$= \frac{1}{I_0} \int_0^{c/\lambda} du \cos qu \sum_{m=1}^{\infty} P(mk) \cos(mku)(1 - \cos 2mka)$$

(34)

and:

$$I_0 = \frac{1}{2\lambda} \int_0^{c/\lambda} du \cos qu \sum_{m=1}^{\infty} P(mk) \cos(mku)(1 - \cos 2mka)$$

(35)

where we have taken advantage of $P(-mk) = P(mk)$.

The detailed expression of the $3 \times 3$ determinant $\Delta^{(3)}(\omega)$ is given in the appendix, as well as the value of $I_0$ — to lowest order in $c/\lambda$ —.

From equation (33):

$$a_1(0) = C \Delta^{(3)}(0).$$

(36)

Using expression (A1), one easily finds:

$$\Delta^{(3)}(0) = \frac{4}{\lambda} \mu(2 - \mu)(a^2 + b^2)(\xi_+ - \xi_-)$$

(37)

so that, with the help of equations (A12) and (31), the relaxation rate of the acoustical phase mode is given by:

$$\omega_{ap}(p) = -Dp^2$$

(38)
where the phase diffusion coefficient $\mathcal{D}$ is:

$$
\mathcal{D} = \frac{\pi \lambda}{4} \left( 2 - \mu \right) \frac{ab}{(a^2 + b^2) \ln \left( \frac{\lambda}{c} \right)}.
$$

Using the explicit expressions of $a$, $b$ (Eqs. (30) of (I)) in the limit $q \lambda \gg 1$, this can be rewritten:

$$
\mathcal{D} = \frac{\pi \lambda}{8 \ln \left( \frac{\lambda}{c} \right)} \left\{ 1 + \left( \frac{\lambda F}{\Delta \gamma'} \right)^2 \right\}^{-1/2}.
$$

So, it is seen that a crenellated front with long facets is always Eckhaus-stable. Since the « local wavevector » $q$ which characterizes the risers lies on the neutral MS curve, if facets were absent, such a structure would be Eckhaus unstable. It is the presence of the long facets which stabilizes the front against phase diffusion.

An important feature of the above result is the presence, in expression (40) for $\mathcal{D}$, of the $(\ln (\lambda / c))^{-1}$ factor. This is clearly the signature of the diffusion field of the steep riser — in the strongly non local limit, where the diffusion equation reduces to Laplace's equation. As will appear again in the following, this factor can be understood as characterizing the lateral « mobility » of the risers.

Finally, the last factor on the r.h.s. of equation (40) is essentially a geometrical one. It accounts for the fact that, the larger the kinetic dissipative term $F$, the longer the hot facet for a given $\lambda$, i.e. the weaker the coupling between successive crenels.

### 3.2 Amplitude and Optical Phase Modes.

Their relaxation rates at long wavelength are obtained by solving the equation:

$$
\Delta^{(3)}(\omega) = 0.
$$

It can be shown, using the same combination of equations (24a), (25b) which led to expression (33), that all these mode have $a + \beta = - (\gamma + \delta)$. That is, they can only involve riser motions of the « optical » type.

In order to be able to classify the modes, let us first consider the simple limit of zero dissipation. In this limit, $F = F' = 0$, $a = b = \lambda / 4$, $I_1 = \Psi = 0$ (Eqs. (A8, 9, 13)). $\Delta^{(3)}$ then reduces to:

$$
\Delta^{(3)}_0(\omega) = (\zeta_+ - \zeta_-) \frac{\lambda^2}{16} (2 \mu + \omega) \left( 2 \Gamma_0 q^2 - \omega \frac{2 \lambda \Phi_0}{\pi^2} \right) \left( \frac{\omega}{\pi \Gamma_0 q^2 \ln \left( \frac{\lambda}{c} \right)} + \frac{2}{\lambda} \right)
$$

where:

$$
\Phi_0 = \frac{4}{\pi} \sum_{m=1}^{\infty} \frac{\sin^2 m \kappa}{m^3}.
$$

The three modes separate immediately into:

a) « climbing » amplitude mode: it has:

$$
\omega_{cl} = - 2 \mu
$$

and corresponds to a global translation of the front along the temperature gradient ($\delta \zeta_+ = \delta \zeta_-, \alpha + \beta = \gamma + \delta = 0$). It is of course relaxing. Note that equation (44) is exact.
(independent of small $\lambda$ and long facet approximations), as can be shown by direct integration of equation (1) along the whole front;

b) \textit{breathing amplitude mode} : this mode is the unstable one calculated in (I), its growth rate is given by:

$$\omega_{br} = \frac{\pi^2 \Gamma_0 q^2}{\lambda \Phi_0}$$

which is the dissipation-free limit of the expression obtained before ((I), Eq. (43)).

It corresponds to opposite motions of hot and cold facets ($\delta \xi_+ = - \delta \xi_- )$ without riser motion ($\alpha + \beta = \gamma + \delta = 0$);

c) \textit{Optical phase mode} : it is relaxing, with relaxation rate:

$$\omega_{op} = - \frac{2 \pi \Gamma_0 q^2}{\lambda \ln (\lambda / \lc)}.$$

The associated front motion is described by: $\gamma + \delta = -(\alpha + \beta)$, i.e. two neighboring risers move with opposite phases. This lateral motion drags a facet rise such that the average front position remains fixed: $\alpha + \beta = -(\lambda / 2 \xi_+) \delta \xi_+$. The expressions of $\omega_{cl}$, $\omega_{br}$ and $\omega_{op}$ confirm that the accessory \cite{3BB/c} factor is indeed, as stated above, characteristic of the riser mobility: it is present in $\omega_{op}$, and absent from $\omega_{cl}$ and $\omega_{br}$ which are associated with pure amplitude fluctuations.

Let us now come back to the finite dissipation case. Since all three modes found at zero $F$ involve finite $\delta \xi$'s, all three of them are affected by facet dissipation. Two remarks can be made about the complete characteristic equation (41):

(i) one can show by writing explicitly the $\omega$ expansion of $\Delta^{(3)}(\omega)$ that terms involving $\Psi_0$, i.e. $I_1$ and $\Psi$, are always $O(\lambda)$ or $O\left((\ln \lambda / \lc)^{-1}\right)$ with respect to dominant contributions, i.e. they can be dropped from expression (A1);

(ii) in contradiction with what was postulated in (I), the breathing mode couples with optical riser motion. Indeed, decoupling of breathing and optical phase motions would imply that the $[3, 3]$ element of $\Delta^{(3)}$ be zero. Would this be the case, the breather growth rate would be given by $[\Delta^{(3)}]_{32} = 0$, i.e. precisely by the expression found in (I).

The fact that $[\Delta^{(3)}]_{33}, [\Delta^{(3)}]_{12}$ are non-zero at finite dissipation indicates that what was overlooked in (I) is the fact that a change in the crenel height imposes an adjustment of relative facet lengths. The cubic characteristic equation has, in general, no simple solution, and writing the explicit expressions of its three roots would be of little interest in the absence of any experimental result, in view of the large number of parameters involved — among which bot $F$ (via $a, b$) and $F' = dF / dV$.

The most important question is to decide whether the breathing instability identified at $F = 0$ persists for any value of the dissipation and whether the modes evolving from the climbing and optical phase ones remain stable. That is, can dissipation induce an exchange of stability? This question can be answered by considering the expressions, given in the Appendix, of the coefficients of the polynomial expansion of $\Delta^{(3)}$:

$$\Delta^{(3)}(\omega) = \sum_{r=0}^{3} c_r \omega^r.$$  

It is seen on equations (A15, A19) that $c_0 > 0$, $c_3 < 0$. (Remember that $F'$ is a positive quantity). So, the product of the relaxation rates is always strictly positive: no real root of equation (41) can change sign.
We must also examine the possibility that two conjugate imaginary roots change the sign of their real part. This can happen only when \( c_0/c_2 = c_1/c_3 > 0 \). Since (see Eq. (A16)) \( c_1 > 0, \ c_3 < 0 \), this possibility also is excluded. There is always one unstable and two stable modes.

One may check easily that, to lowest order in \( F, F' \) the three roots (44-46) are shifted by terms proportional to \( VF' \) only, and in that limit the result of (I) therefore remains valid. As expected, a finite \( F' \) slows down the growth or relaxation rates of the three modes.

In the opposite limit of very large \( F, F' \), it can be shown that two of the relaxation rates are of order \((VF')^{-1}\), while one is of order 1. This one can be identified as corresponding to a pure optical phase mode without facet climbing; it is relaxing.

We can therefore conclude that, although the assumption made in (I) that amplitude and phase motion decouple is not justified at finite dissipation, its qualitative result about crenellated front stability remains right: such a front profile is always unstable with respect to a mode involving facet displacement along the growth direction. How much optical riser motion is involved in this mode depends on the importance of facet dissipation \( F \) as well as on the growth mechanism (via \( F' = dF/dV \) which is different for, e.g., dislocation and 2D nucleation kinetics).

Crenellated fronts are stable with respect to phase diffusion. From the expressions of the phase diffusion coefficient and of the zero-\( F \) relaxation rates of the other modes, it appears that diffusion slows down the lateral motion of a steep riser by a factor \((\ln \lambda/c)^{-1}\), where \( c \) is the riser width. We believe that this slowing down factor will appear in all situations involving motion of the so-called « macrosteps » observed on faceted fronts [2]. This is for example the case of periodic vicinal fronts in directional solidification [5].

### Appendix

The \( 3 \times 3 \) determinant \( \Delta^{(3)} \) defined by equation (33) can be written, after some tedious but straightforward manipulations, as:

\[
\Delta^{(3)}(\omega) = (\xi_+ - \xi_-) D
\]

with:

\[
D = \begin{vmatrix}
\frac{4 \mu + 2 \omega}{\Gamma_0 q^2} & \frac{4(a-b)}{\lambda} + \frac{\omega}{\Gamma_0 q} I_1 & \frac{4 + \omega}{\Gamma_0 q^2 \lambda} + \frac{\omega}{\Gamma_0 q} I_2 \\
\mu + \omega & 0 & \frac{a}{\lambda} \left( \mu + \frac{\omega}{2} \right) \\
0 & 2b\Gamma_0 q^2 - \omega \left( bVF' + \frac{\lambda \Phi}{2a} \right) - \frac{\Gamma_0 q^2(a-b)}{2a} - \frac{\omega \lambda}{4a} \Psi \\
\end{vmatrix}.
\]

This expression of \( \Delta^{(3)} \) is obtained by using independent variables proportional respectively to \((\alpha \delta \xi_+ + b \delta \xi_-), (\delta \xi_+ - \delta \xi_-), (\alpha + \beta - \gamma - \delta)\) where:

\[
I_1 = \frac{1}{\pi} \int_{\phi=0}^{\phi=c/2} d\phi \cos \phi \sum_{m=-1}^{\infty} \frac{P(mk)}{m} \sin(2mka) \cos(mku) \tag{A2}
\]

\[
I_2 = \frac{1}{2\lambda} \int_{\phi=0}^{\phi=c/2} d\phi \cos \phi \sum_{m=1}^{\infty} P(mk) \cos(mku) [1 + \cos 2mka] \tag{A3}
\]
Since we are dealing with the highly non-local limit, \( k > 1 \) so that, in \( I_1, I_2, \Phi, \Psi \), as well as the integral \( I_0 \) defined by equation (35), we can use:

\[
\Phi = \frac{\lambda}{2 \pi^2} \sum_{m=1}^{\infty} P(mk) \frac{\sin^2 mk}{m^2} \tag{A4}
\]

\[
\Psi = \frac{1}{4 \pi} \sum_{m=1}^{\infty} P(mk) \frac{\sin 2 mk}{m} \tag{A5}
\]

\( P(mk) \equiv \frac{4}{k |m|} \) \hspace{1cm} (A6)

\( \Phi \) and \( \Psi \) can be written as:

\[
\Phi = \frac{1}{k^2} \Phi_0 \tag{A7}
\]

\[
\Psi = \frac{1}{k} \Psi_0 \tag{A8}
\]

where:

\[
\Phi_0 = \frac{4}{\pi} \sum_{m=1}^{\infty} \frac{\sin^2 mk}{m^3} \tag{A9}
\]

\[
\Psi_0 = \frac{1}{\pi} \sum_{m=1}^{\infty} \frac{\sin 2 mk}{m^2} \tag{A10}
\]

Note that \( \Phi_0 \) and \( \Psi_0 \) are, for all values of \( (ka) \), numbers of order 1.

The \( m \)-sums in \( I_0 \) and \( I_2 \) are easily performed, while that in \( I_1 \) can be expressed as:

\[
-\frac{\lambda}{\pi} \int_0^u dt \ln \left| \frac{\sin k(a+t/2)}{\sin k(a-t/2)} \right| \tag{A11}
\]

The \( t \)-integral can then be calculated approximately by taking advantage of the fact that \( c \ll \lambda \) (long facet approximation). We then find, to lowest order in \( (c/\lambda) \):

\[
I_0 \equiv I_2 \equiv \frac{1}{\pi q} \ln \left( \frac{\lambda}{c} \right) \tag{A12}
\]

\[
I_1 \equiv \frac{2 \lambda}{\pi} \frac{\Psi_0}{q} \tag{A13}
\]

Finally, let us write down the coefficients of the \( \omega \)-expansion of \( \Delta^{(3)} \):

\[
\Delta^{(3)}(\omega) = \sum_{r=0}^{3} c_r \omega^r.
\]

Setting:

\[
c_r = d_r (\xi_+ - \xi_-) ab \tag{A14}
\]

one finds from (A1):

\[
d_0 = \frac{4 \mu \Gamma_0 q^2 a^2 + b^2}{\lambda \mu ab} \tag{A15}
\]

\[
d_1 = (1 + VF') \frac{2 \Gamma_0 q^2 a^2 + b^2}{\lambda ab} + \frac{4 \mu \ln \left( \frac{\lambda}{c} \right)}{abk^2} - \frac{2 \mu \Phi_0}{abk} + \frac{6 \Psi_0 \mu (a-b)}{abk} \tag{A16}
\]
It is easily seen that the third and fourth terms are both of order $\lambda$ with respect to the first one in the r.h.s. of (A16), and therefore negligible, so that:

$$d_1 \approx (1 + VF') \frac{2 \Gamma_0 q^2 a^2 + b^2}{\lambda} \frac{4 \mu}{ab} + \frac{4 \mu}{\pi} \ln \left( \frac{\lambda}{c} \right). \quad (A17)$$

Neglecting, analogously, terms of order $\lambda$ or $\left( \ln \left( \frac{\lambda}{c} \right) \right)^{-1}$ with respect to one in $d_2$ and $d_3$ leads to:

$$d_2 = \frac{2 VF'}{\lambda} + \frac{2}{\pi} (1 + VF') \ln \left( \frac{\lambda}{c} \right) - \frac{2}{\Gamma_0 q^2 \lambda} \left( 2 VF' + \Gamma_0 q^2 + \frac{\mu \lambda}{\pi} \ln \left( \frac{\lambda}{c} \right) \right) \left( VF' + \Phi_0 \frac{\lambda}{2 abk^2} \right) \quad (A18)$$

$$d_3 = -\frac{1}{\Gamma_0 q^2} \left( VF' + \Phi_0 \frac{\lambda}{2 abk^2} \right) \left( VF' + \frac{1}{\pi} \ln \left( \frac{\lambda}{c} \right) \right). \quad (A19)$$

References


