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Lamellar eutectic growth at large thermal gradient: I. Stationary patterns

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We study stationary front profiles of directionally solidified lamellar eutectics. We show that, in the limit of large thermal gradients, front shapes can be calculated analytically without resorting to the Jackson-Hunt ansatz [7] of equal average front undercooling of both phases. We find that the difference between them is comparable with the global average one \( \Delta T \), but that the variation of \( \Delta T \) with the lamellar period \( \lambda \) exhibits a minimum for a value of \( \lambda \) of the same order of magnitude as that predicted by J. H. We look for stationary tilted solutions. These can occur in general for isolated values of the tilt angle, but are absent in the large gradient limit, in agreement with the experimental observation of a velocity threshold for « tilt waves ».

1. Introduction.

Directional solidification, when applied to a binary metallic alloy with a concentration \( C_\infty \) close to that, \( C_E \), of its eutectic point, produces a solid organized, basically, as a periodic arrangement of the two stable solid phases \( \alpha, \beta \) (Fig. 1). Two types of such structures are

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observed in bulk samples: alternating lamellae and hexagonal rod patterns parallel to the common (Oz) direction of the pulling velocity $V$ and of the external thermal gradient $G$. These metallurgical studies [1] have been complemented with experiments on transparent eutectics [2] with microscopically rough solid-liquid interfaces (so that the growth kinetics be, as in metals, quasi-instantaneous). In these experiments, performed on thin samples in order to allow for continuous optical observations of the patterns — and, in particular, of their dynamics — the standard stationary growth mode appears to be lamellar. Most recently, interest has focused on the problem of wavelength selection in out-of-equilibrium periodically structured systems and on the nature of the dynamical mechanisms involved in this selection. The experiments of Faivre et al. [3] on the CBr$_4$-C$_2$Cl$_6$ eutectic have shown that among such dynamical defects, « solitary tilt waves » play an important role. These consist in domains in which the lamellae, which continue those of the « normal » patterns, are parallel but tilted at a well-defined finite angle $\phi$ with respect to the pulling direction. These domains are defects of the mesoscopic growth pattern, which are not correlated with defects of the underlying microscopic lattice. Their width is much larger than the wavelength $\lambda$ of the lamellar structure, and for given $V$ and $G$, domains of various widths are observed. Such dynamical defects have also been identified in two different experiments on systems with periodic front patterns, namely directional growth of a nematic liquid crystal [4] and directional viscous fingering [5].

Coullet et al. [6] have proposed a mathematical phenomenology describing these defects. This puts to the fore the fact that they are associated with the breaking of the reflection symmetry about the Oz axis, i.e. with the presence of a finite antisymmetric component of the front profile. This phenomenology predicts, that, above some velocity threshold, there should exist stationary solutions of the eutectic growth problem with periodic parallel tilted lamellae filling the whole sample.

The question thus arises of substantiating this description by directly solving the growth equations for specific systems, so as to determine their domain of existence in parameter space and their characteristics — e.g. their velocity of drift along the front.

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Fig. 1. — Phase diagram of a eutectic alloy with constant concentration gaps.
The basic theoretical description of stationary eutectic growth is due to Jackson and Hunt [7] (JH). The basic ingredients of their theory of untilted periodic lamellar (and rod) structures are the following:

(i) the solute diffusion field in the liquid is approximated by that of a planar front (with, of course, a periodic arrangement of \((\alpha)\) and \((\beta)\) regions);

(ii) the ansatz is then made that the average front undercoolings \(\Delta T_\alpha, \Delta T_\beta\) along \(\alpha\) and \(\beta\) lamellae are equal.

This enables them to predict a wavelength dependence of the average front undercooling \(\Delta T(\lambda)\) which exhibits a minimum for

\[
\tilde{\lambda} = \tilde{\lambda}_{\text{min}} \sim \sqrt{\tilde{d}_0 \ell}
\]

(where \(\tilde{d}_0\) and \(\ell\) are, respectively, the capillary and solute diffusion length), which is of the order of experimentally observed wavelengths.

In order to study tilted lamellar solutions, one is therefore naturally led to try and extend the JH approach. On can then immediately, for an arbitrary value of the tilt angle \(\phi\), calculate an average undercooling \(\Delta T(\lambda, \phi)\), which suggests the existence, for each value of \(\lambda\), of a continuum of stationary tilted solutions. This clearly contradicts the experimental observation [3] that \(\phi\) is sharply selected.

The source of this discrepancy can be traced back to the equal undercooling ansatz. Indeed, there are two steps in the JH theory: in the first one, averages \(\Delta T_\alpha, \Delta T_\beta\) are calculated with the help of the planar front approximation (i), by integrating along the front of each lamella the Gibbs-Thomson equation (i.e. of the condition of local thermodynamic equilibrium). This can be performed formally without calculating explicitly the front profile. JH then apply their ansatz and obtain \(\Delta T(\lambda)\). The front profile is later calculated without using the ansatz as a by product of this approach. It can be checked, as we will show, that the value of \(\Delta T(\lambda)\) which may then be deduced from the shape of this profile is not consistent with that resulting from the equal undercooling ansatz — although the predicted \(\lambda_{\text{min}}\) is only changed by a factor of order one, as can be expected from a qualitative evaluation of the order of magnitude of the capillary and diffusive contributions to \(\Delta T\).

This flaw, which has only a minor effect on the description of non-tilted patterns, becomes crucial when looking for tilted ones: indeed, we will see that, at finite \(\phi\), profiles calculated by solving the Gibbs-Thomson equation separately in the \((\alpha)\) and \((\beta)\) regions cannot, in general, be matched to fit the equilibrium conditions at the \(\alpha\)-\(\beta\)-liquid triple points. These remarks lead us to modify the JH approach and make it consistent in the following way: we first calculate a front profile satisfying the triple point conditions. This, of course, due to the non-linearities of the problem, we cannot perform exactly. We therefore retain the planar front approximation (i) in the calculation of the diffusion field to calculate the resulting front profile. We show that, as can be expected, this approximation is valid, in the experimental range \(\lambda \sim \lambda_{\text{min}}\), only in the large thermal gradient regime — more precisely, for \(l_T/\ell \ll 1\), where \(l_T\) is a thermal length \(\propto G^{-1}\), and \(\ell\) the diffusion length \(\propto V^{-1}\) (see § 2).

The validation of the planar front approximation in this regime was originally discussed by Brattkus and Davis [8] although error estimates appropriate for \(\tilde{\lambda} \sim \tilde{\lambda}_{\text{min}}\) were not derived. In this large gradient limit we calculate \(\Delta T(\lambda)\) and the precise value of \(\lambda_{\text{min}}\) for untilted lamellae.

We then investigate the existence of tilted solutions. We first show, by an exact counting argument, that they can exist only for discrete values of the tilt angle \(\phi\). We solve the front equation separately in the \((\alpha)\) and \((\beta)\) regions and obtain from the matching conditions at the triple points the eigenvalue equation satisfied by \(\phi\). We find that, in the large thermal
gradient limit where our approach is valid, this has no solution with $\phi \neq 0$, in agreement with
the experimental observation, at fixed $G$, of a lower velocity threshold $V_t$ for solitary tilt
waves. It appears in our calculation that the relevant parameter for the existence of tilted
lamellae is $l_T/l$. We can therefore reasonably infer from this that the threshold velocity
$V_t$ should be proportional to $G$.

2. The front equation.

The growing eutectic is described in the following simple model which preserves the essential
physical features of the system:

— We assume that the thermal gradient $G$ is constant and uniform. That is, we assume that
the thermal diffusivities are equal in the three ($\alpha$, $\beta$, $L$) phases, and that thermal transport
(evacuation of latent heat) is much faster than chemical diffusion.

— We assume that the kinetics of atomic attachment at the $\alpha/L$ and $\beta/L$ interfaces is
instantaneous on the time scale of the phenomena of interest, i.e. that the front is everywhere
at local thermodynamic equilibrium. This implies, in particular, that the interfaces are
microscopically rough.

— We neglect chemical diffusion in the solid phase (one-sided model): $D_{e_\alpha}, D_{e_\beta} \ll D$,
where $D$ is the solute diffusion coefficient in the liquid phase at near-eutectic composition.

We moreover make the simplifying (but unessential) assumption that, for liquid composi-
tions $C$ close to the eutectic one, $C_E$, the two concentration gaps on the phase diagram of
the binary system are constant (*). They will be denoted by $\Delta C_a = C_E - C_a$,
$\Delta C_\beta = C_\beta - C_E$ (see Fig. 1), and we set:

$$\delta = \frac{\Delta C_a}{\Delta C}.$$  \hfill (2)

(with $\Delta C = \Delta C_a + \Delta C_\beta$) and call $m_a$, $m_\beta$ the absolute values of the slopes ($dT/dC$) of the
liquidus lines at the eutectic point.

We define the dimensionless composition:

$$u = \frac{C - C_E}{\Delta C}$$  \hfill (3)

and express length and time in units of, respectively, the solute diffusion length and time:

$$\ell = D/V \quad \tau = D/V^2.$$  \hfill (4)

We are interested here only in lamellar structures, i.e. in 1-D front deformations. The
solidifying system is then described, in the laboratory frame — where the sample is pulled at
velocity $V$ along ($-Oz$) — by [7]:

a) In the liquid ($z > \zeta(x,t)$):

$$\frac{\partial u}{\partial t} = \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z^2} + \frac{\partial}{\partial z} \right) u$$  \hfill (5)

(* This happens to be a good quantitative approximation of the phase diagram of the
CBr_4-C_2Cl_6 mixture [9].)
b) On the front \( z = \zeta(x, t) \).

(i) Solute conservation:

\[- \hat{n} \cdot \nabla u = A(x, t)(1 + \dot{\zeta}) n_z \]  

where \( \hat{n} \) is the unit vector normal to the interface pointing into the liquid and:

\[ A(x, t) = \begin{cases} \delta & \text{for } x \text{ in } (\alpha/L) \text{ front regions} \\ 1 - \delta & \text{for } x \text{ in } (\beta/L) \text{ front regions} \end{cases} \]  

\[ (7) \]

(ii) local equilibrium:

At each triple \( (\alpha - \beta - L) \) point (see Fig. 2), it is expressed by the condition that the capillary forces balance. When the anisotropies of the three surface tensions \( \sigma_{\alpha L} \), \( \sigma_{\beta L} \), \( \sigma_{\alpha \beta} \) can be neglected — which we will from now on assume, this simply reduces to:

\[ \sigma_{\alpha L} + \sigma_{\beta L} + \sigma_{\alpha \beta} = 0 \]  

where each \( \sigma \) vector points out of the triple point and is tangent to the corresponding interface.

\[ (8) \]

Fig. 2. — Periodic lamellar pattern.

On all other points of the solid-liquid front local equilibrium is expressed by the Gibbs-Thomson equation for the relevant \( (\alpha - L \text{ or } \beta - L) \) interface, namely:

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

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

\[ (9.a) \]

\[ (9.b) \]

\[ \kappa (x, t) = - \frac{\zeta''}{(1 + \zeta'^2)^{3/2}} \]  

(with \( \zeta' = \frac{\partial \zeta}{\partial x} \)) is the front curvature and \( \zeta \) is measured from the
position of the $T = T_E$ isotherm. $d_i$ and $\ell_{\text{Ti}}(i = \alpha, \beta)$ are, respectively, the dimensionless capillary and thermal lengths associated with the (i) phase:

$$d_i = \frac{\tilde{d}_i}{\ell} = \frac{1}{\ell} \frac{\sigma_{iL} T_E}{m_i(\Delta C) L_i} \quad (10)$$

$$\ell_{\text{Ti}} = \frac{\ell_{\text{Ti}}}{\ell} = \frac{1}{\ell} \frac{m_i(\Delta C)}{G} \quad (11)$$

and the $L_i$ are the specific latent heats.

Equations (5) to (9), supplemented with the boundary condition:

$$\lim_{z \to \infty} u = u_\infty \quad (12)$$

provide a complete description, of the growth problem. As is well known it is possible in such a one-sided free boundary problem, to express exactly, with the help of equations (5), (6) and (12), the value of the diffusing field $u$ at the interface as a functional of the front shape. When this is done, equations (9), together with the triple point conditions (8), become a closed set of equations for the single unknown function $\zeta(x, t)$.

We will now perform this reduction, which will prove handy for qualifying our approximation and for the linear stability analysis performed in the companion paper [10].

As shown in reference [11] from equations (5) and (12), one can express exactly, in the 2-D one-sided model, the value of $u$ on the front as:

$$u(x, \zeta(x, t)) \equiv u(x', \zeta(x', t'), t') \quad (14.\text{a})$$

and

$$G(x, z, t \mid x', z', t') = \frac{1}{4 \pi(t-t')} \exp \left\{ - \frac{(x-x')^2 + (z-z' + t - t')^2}{4(t-t')} \right\} \quad (14.\text{b})$$

One can then eliminate $u(x, \zeta(x, t))$ and $(n \cdot \nabla u)_{x,\zeta}$, from equation (13) with the help of equation (6) and of the Gibbs-Thomson equations (9). This results in two integrodifferential equations for the front shape $\zeta(x, t)$, valid in the $(\alpha - L)$ and $(\beta - L)$ front regions respectively. These are to be solved together with the boundary conditions at the triple points, namely:

- continuity of $\zeta$
- equilibrium conditions (8).

These equations are — as usual in such free boundary problems — non linear in $\zeta$, which appears in particular within the diffusion propagator $G$.

However, an important simplification can be performed. Indeed, in lamellar eutectic solidification, the observed lamellar wavelengths $\lambda$ are typically, of the order of 10 microns, while diffusion lengths lie in the 500 $\mu$m range ($D \sim 10^{-5}$ cm$^2$/s, $V \sim$ a few $\mu$m/s). So

$$\lambda = \frac{\tilde{\lambda}}{\ell} \ll 1 \quad (15)$$

This is the so-called small Peclet number limit.
The dominant contribution to the integrals on the r.h.s. of equation (13) comes from the regions \(|x - x'| \leq 1, t - t' \leq 1\) corresponding to the diffusive range of our problem. On the other hand, slope variations of the front profile are restricted by the triple point equilibrium conditions. So, deformation amplitudes are at most of order \(\lambda \ll 1\), and one can legitimately linearize the diffusion propagator \(G\) in \(\langle \zeta - \zeta' \rangle\). Moreover, we assume the quasi-stationary approximation [12] to be valid. This amounts to setting \(\zeta(x', t') = \zeta(x', t)\) everywhere in the r.h.s. of equation (13). This approximation is valid, for time-dependent phenomena, when the relevant time scales are short compared with the time of propagation of the diffusive signal on the typical length scale of the front deformation, which will be the case for the phenomena we investigate.

Then, using equation (6) and performing the time integration, (13) reduces to:

\[
u(x, \zeta, t) = N(x, t) - \int_{-\infty}^{\infty} \frac{dx'}{2\pi} u(x', \zeta, x, t) K_0\left(\frac{|x - x'|}{2}\right)
\]

(16)

where \(K_0\) is the modified Bessel function [13] and:

\[
N(x, t) = 2u_\infty + \int_{-\infty}^{\infty} \frac{dx'}{2\pi} \{2(1 + \zeta(x', t)) - \zeta(x, t) + \zeta(x', t)\} \cdot \Delta(x', t) K_0\left(\frac{|x - x'|}{2}\right)
\]

(17)

\(\Delta(x, t)\) is the step-function defined by equation (7). For non-stationary front profiles, it depends on time via the positions of the triple points.

At this stage, one may of course simply replace \(u\) on both sides of equation (16) by expressions (9). However, note that the r.h.s. integration sweeps the whole \(x\)-axis, including triple points, where slope discontinuities give rise to \(\delta\)-function singularities of the curvature \(K\) which must be explicitly taken into account as an inhomogeneous driving term in the front equation [14].

This complication can be avoided in the following way: equation (16) can be rewritten formally as:

\[
\int dx' O(x - x') u(x', \zeta, t) = N(x, t)
\]

(18)

which can be inverted into:

\[
u(x, \zeta, t) = \int_{-\infty}^{\infty} dx' O^{-1}(x - x') N(x', t) = \int_{-\infty}^{\infty} \frac{dQ}{2\pi} e^{iQx}[1 - \Pi(Q)] N(Q, t).
\]

(19)

Using the expression [13] of the Fourier transform of \(K_0\):

\[
K_0(Q) = 2\pi(1 + 4Q^2)^{-1/2}
\]

(20)

one finds immediately:

\[
\Pi(Q) = [1 + (1 + 4Q^2)^{1/2}]^{-1}
\]

(21)

So finally, the front equations are obtained by replacing in the r.h.s. of equations (9), \(u\) by:

\[
u(x, \zeta, t) = u_\infty + \int_{-\infty}^{\infty} \frac{dQ}{2\pi} e^{iQx}\{\Pi(Q)[2\Delta(Q, t) + I_1(Q, t)] - (1 - \Pi(Q))I_2(Q, t)\}
\]

(22)
where:

\[
I_1(Q, t) = \int_{-\infty}^{\infty} \frac{dq}{2\pi} [2\zeta(q, t) + \zeta(q, t)] \Delta(Q - q, t) \tag{23.a}
\]

\[
I_2(Q, t) = \int_{-\infty}^{\infty} \frac{dq}{2\pi} \Delta(Q - q, t) \frac{K_0(Q - q)}{2\pi} \zeta(q, t). \tag{23.b}
\]

3. The standard stationary lamellar pattern.

We first study untilted stationary periodic lamellar patterns (Fig. 2). Let

\[x_{n1} = n\lambda, \quad x_{n2} = (\eta + n)\lambda\]

be the abscissae of, respectively, the \(\beta/\alpha\) and \(\alpha/\beta\) triple points, where the \(\alpha\)-phase fraction \(\eta\) is a priori unknown (except in the special case of vertical solidus lines [15], which we do not consider here, where \(\eta = 1 - C_\infty\)).

The values of the pinning angles \(\theta_\alpha, \theta_\beta\), are uniquely determined by equation (8), which reads in the present geometry:

\[
\begin{cases}
\sigma_{aL} \sin \theta_\alpha + \sigma_{bL} \sin \theta_\beta = \sigma_{\alpha\beta} \\
\sigma_{aL} \cos \theta_\alpha - \sigma_{bL} \cos \theta_\beta = 0.
\end{cases} \tag{24}
\]

So, the value of the profile slopes are constrained to be opposite at the two ends of a given lamella.

1. The large gradient limit. — The stationary front equations are obtained from the time independent version of equations (9), (22) together with the matching conditions

\[
\begin{align*}
\zeta_\alpha(x_{n1}) &= \zeta_\beta(x_{n1}) \quad \zeta_\alpha(x_{n2}) = \zeta_\beta(x_{n2}) \tag{25} \\
\zeta'_\alpha(x_{n1}) &= -\zeta'_\beta(x_{n2}) = \tan \theta_\alpha \\
\zeta'_\beta(x_{n1}) &= -\zeta'_\beta(x_{n2}) = -\tan \theta_\beta. \tag{26}
\end{align*}
\]

Although their diffusive term has been linearized, due to the non-homogeneity of the solid phase, their are highly non-trivial, and no exact solution is presently known.

However, the following remark can be made: the diffusion field ahead of the eutectic front exhibits two types of space-varying contributions: a diffusion layer of thickness \(\sim \ell\), associated with global solute rejection, and modulations due to the periodic structure of the solid front, which extend on a range of order \(\sim \lambda(\lambda \ll \ell)\). When the amplitude of front deformations is much smaller than this range, it is certainly reasonable to approximate the diffusion field by that of the average planar front (with alternating \(\alpha\) and \(\beta\) parts).

This is precisely the central approximation of the JH approach [7] as well as of the stability analysis of Datye and Langer [15].

However, the question of the range of validity, in parameter space, of these approximations, was left open. It is qualitatively reasonable to expect that, the larger the thermal gradient \(G\), the higher the energy cost of front excursions and thus the better this planar approximation.

That is, for small enough reduced thermal lengths \(\ell_T\), we assume that \(\zeta(x)\) in equations (22), (23) can be replaced by its average value \(\bar{\zeta}\) (unknown at this stage). The
Gibbs-Thomson equations are then easily checked to reduce to:

$$
\begin{align*}
&d_a \frac{\zeta'_a}{(1 + \zeta'^2_a)^{3/2}} - \frac{\zeta_a}{\zeta_{Ta}} = u_\infty + \eta + \delta - 1 + \phi(x); \\
&d_\beta \frac{\zeta'_\beta}{(1 + \zeta'^2_\beta)^{3/2}} - \frac{\zeta_\beta}{\zeta_{T\beta}} = -[u_\infty + \eta + \delta - 1 + \phi(x)]; \\
&\text{where use has been made in equation (22) of the relation:}
\end{align*}
$$

where $\phi(x)$ is the JH function given by:

$$
\phi(x) = \sum_{p = -\infty}^{\infty} \Delta_p^{(0)} \Pi(pK) e^{ipKx}(1 - \delta_{p0})
$$

where:

$$
K = 2\pi /\lambda
$$

$$
\Delta^{(0)}(x) = \begin{cases} 
\delta & n\lambda < x < (n + \eta)\lambda \\
- (1 - \delta) & (n + \eta)\lambda < x < (n + 1)\lambda .
\end{cases}
$$

In our small wavelength limit (15):

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

How small should $\ell_T$ be for this approximation to be valid will discussed below, where it will appear that equations (27) are the lowest order in an expansion in the parameter $\ell_T = \ell_T /\ell$.

2. STATIONARY FRONT PROFILE. — Note that, unlike the diffusion term, the capillary term in equations (27) cannot in general be linearized: the pinning angles — determined by the relative magnitudes of surface tensions — are in general finite, and, at least close to the triple points, $\zeta'$ cannot be neglected.

However, as argued above, a large thermal gradient counteracts the pinning effect by flattening the profile. As is apparent on equations (27), the meniscus shape resulting from this competition has a typical range of space variation:

$$
q_i^{-1} = (\ell_{Ti} d_i)^{1/2} .
$$

On the other hand, the r.h.s. diffusive terms vary on the scale of the lamellae thickness, of order $\lambda$. Thus, if the $\ell_T$'s are small enough for the condition:

$$
q_i \lambda \gg 1
$$

to be satisfied, it becomes possible to separate, within each lamella:

— edge regions where space variations of $\phi$ can be neglected while the non-linearity of the curvature is retained.

— a central part in which $|\zeta'| \ll 1$ and (27) can be fully linearized.

As mentioned in § 1, experimentally observed wavelengths are, typically, of the order of
the JH minimum undercooling one (Eq. (1)). We will from now on assume that \( \lambda \) lies in this range, so that, in order of magnitude:

\[
\lambda = 0(\sqrt{d}).
\]

So, condition (31) becomes:

\[
\ell_{Ti} \ll 1.
\]

Due to the symmetry of \( \phi(x) \) with respect to lamellae centers\( \left( x = \left(n + \frac{\eta}{2}\right) \lambda ; x = \left(n + \frac{1 + \eta}{2}\right) \lambda \right) \), and to that of the pinning conditions, the front profile of each lamella is itself symmetric.

Let us first build the solution (27) in a \( \alpha \) lamella \( (0 < x < \eta \lambda /2) \).

a) Edge region \( (0 \leq x \leq \eta \lambda /2) \)

In this region \( \phi(x) = \phi(0) \). Equation (27.a) then exactly reduces to that for the meniscus formed by a semi-infinite liquid submitted to gravity [16] with a slope at its edge \( (x = 0) \) fixed by condition (26):

\[
q_{\alpha} x = \ln \tan \frac{\theta_{\alpha}}{4} + 2 \cos \frac{\theta_{\alpha}}{2} + \cosh^{-1} \frac{2}{t_{\alpha}} - \sqrt{4 - t_{\alpha}^2} \quad (34.a)
\]

\[
t_{\alpha} = q_{\alpha} \left[ \zeta_{\alpha} + \ell_{Ta}(u_\infty + \eta + \delta - 1 + \phi(0)) \right]. \quad (34.b)
\]

b) Lamella center \( (q_{\alpha}^{-1} \ll x \ll \eta \lambda /2) \)

In this region, the capillary term in (27.a) reduces to \( d_{\alpha} \zeta_{\alpha}'' \). One obtains from the linearized version of equation (27.a)

\[
\zeta_{\alpha}(x) = -\ell_{Ta}(u_\infty + \eta + \delta - 1) + \mu_{\alpha} \cosh q_{\alpha} \left( x - \frac{\eta \lambda}{2} \right) + \nu_{\alpha} \sinh q_{\alpha} \left( x - \frac{\eta \lambda}{2} \right) + \left( q_{\alpha} d_{\alpha} \right)^{-1} \int_{\eta \lambda /2}^{x} dx' \sinh q_{\alpha}(x - x') \phi(x'). \quad (35.a)
\]

Taking into account the profile symmetry \( (\zeta_{\alpha}'(\eta \lambda /2) = 0) \) and the expression (29.c) of \( \phi(x) \):

\[
\zeta_{\alpha}(x) = -\ell_{Ta}(u_\infty + \eta + \delta - 1) + \left[ \mu_{\alpha} + \frac{\lambda}{d_{\alpha}} \sum_{p > 1} \frac{\sin p \pi \eta}{p^2(\frac{q_{\alpha}^2}{4} - p^2 K^2)} \right] \cosh q_{\alpha} \left( \frac{\eta \lambda}{2} - x \right) - \frac{\lambda}{d_{\alpha}} \sum_{p \neq 1} \frac{\sin p \pi \eta \cos p(\frac{\ell_{Ta}}{\eta \lambda} - \eta \pi)}{p^2(\frac{q_{\alpha}^2}{4} + p^2 K^2)} \quad (35.b)
\]

where \( \mu_{\alpha} \) is a (free) constant of integration.

c) Asymptotic matching

Such a matching is possible provided that the ranges of validity overlap, i.e., when condition (31) holds. The expansions of (34) and (35) in the common asymptotic region \( (q_{\alpha}^{-1} \ll x \ll \eta \lambda /2) \) both are of the form \( \zeta_{\alpha} \approx A + B \exp(-q_{\alpha} x) \). Matching the exponential terms determines \( \mu_{\alpha} \):

\[
\mu_{\alpha} = -\frac{8}{q_{\alpha}} \tan \frac{\theta_{\alpha}}{4} \exp - \left( \frac{q_{\alpha} \eta \lambda}{2} + 4 \sin^2 \frac{\theta_{\alpha}}{4} \right) - \frac{\lambda}{d_{\alpha}} \sum_{p \neq 1} \frac{\sin \pi \eta p}{p^2(\frac{q_{\alpha}^2}{4} + p^2 K^2)}. \quad (36)
\]
Compatibility between the two already known $A$ constants imposes that:

$$S = \phi (0)$$

$$S = \lambda \sum_{p \neq 1} \frac{q_{a}^{2}}{q_{a}^{2} + p^{2} k^{2}} \sin \frac{2 \eta \pi p}{2 p^{2} \pi^{2}}.$$  (37.a)

(37.b)

Using expression (29.b) for $\phi$, one easily checks that:

$$\left| \frac{S - \phi (0)}{\phi (0)} \right| \approx 0 ((q_{a} \lambda)^{-1})$$  (38)

that is, up to the order at which our calculation is performed, condition (37.a) is indeed satisfied.

The front equation (Eq. (9.b)) appropriate to $\beta$ lamellae is solved by the same procedure. We must impose the continuity condition (25.a) at the $\beta - \alpha$ triple point $x = 0$ (due to the reflexion symmetry of the profile with respect to each lamella center, this also ensures continuity at the $\alpha - \beta$ ones).

From expression (34):

$$\zeta_{\alpha} (0) = - \frac{2}{q_{a}} \sin \frac{\theta_{\alpha}}{2} - \ell_{T \alpha} [u_{\infty} + \eta + \delta - 1 + \phi (0)].$$  (39.a)

Similarly, from the solution in the $\beta$ lamella:

$$\zeta_{\beta} (0) = - \frac{2}{q_{\beta}} \sin \frac{\theta_{\beta}}{2} + \ell_{T \beta} [u_{\infty} + \eta + \delta - 1 + \phi (0)].$$  (39.b)

Continuity therefore results in the following compatibility condition, which determines the $\alpha$-phase fraction $\eta (\lambda)$ for a pattern of fixed wavelength $\lambda$:

$$u_{\infty} + \eta + \delta - 1 + \frac{\lambda}{2} P' (\eta) = (\ell_{T \alpha} + \ell_{T \beta})^{-1} \left[ \frac{2}{q_{\beta}} \sin \frac{\theta_{\beta}}{2} - \frac{2}{q_{a}} \sin \frac{\theta_{\alpha}}{2} \right]$$  (40)

where:

$$P' (\eta) = \frac{dP (\eta)}{d\eta}$$

$$P (\eta) = \sum_{p > 1} \frac{\sin^{2} \pi p \eta}{\pi^{3} p^{3}}.$$  (41)

The r.h.s. of equation (40) is of order $\left( \frac{\ell_{T} q}{\ell_{T}} \right)^{-1} \sim \left( \frac{d}{\ell_{T}} \right)^{1/2}$. Typically, capillary lengths $d \sim 50 \, \AA$. For CBr$_4$ - C$_2$ Cl$_6$, $d_{\alpha} = 26 \, \AA$, $d_{\beta} = 47 \, \AA$, while $m_{\alpha} \Delta C = 15 \, K$, $m_{\beta} \Delta C = 22 \, K$. So it is seen that, even in the large gradient regime (defined below), $\ell_{T} \sim 20 / G$, and $d < \ell_{T}$.

Since, moreover, $\lambda \ll 1$, equation (40) can be solved by iteration, and one finds, using conditions (32) and (33), which entail $\frac{\lambda}{(q \ell_{T})^{-1}} = 0 (\sqrt{\ell_{T}} \ll 1)$:

$$\eta = 1 - \delta - u_{\infty} + 0 \left( \sqrt{\frac{d}{\ell_{T}}} \right).$$  (42)
That is, the fractional amount of $\alpha$-phase is very close to what it would be for a system with a front at the eutectic temperature.

3. AVERAGE FRONT UNDERCOOLING. — Following JH, we define the average front undercooling as measured from the eutectic temperature $T_E$:

$$\Delta T = -G \bar{\xi} = -G \left[ \eta \bar{\xi}_\alpha + (1 - \eta) \bar{\xi}_\beta \right]$$

(43)

where, e.g.:

$$\bar{\xi}_\alpha = \frac{2}{\eta \lambda} \int_0^{\eta \lambda / 2} \, dx \, \xi_\alpha(x).$$

(44.a)

This we write as:

$$\bar{\xi}_\alpha = \frac{2}{\eta \lambda} \left[ \int_0^X \, dx \, \xi_\alpha(x) + \int_X^{\eta \lambda / 2} \, dx \, \xi_\alpha(x) \right]$$

(44.b)

where the cut-off $X$ lies in the region of asymptotic matching ($q_a^{-1} \ll X \ll \eta \lambda / 2$). We then obtain, with the help of equations (34), (35), (36):

$$\frac{\eta \lambda}{2} \bar{\xi}_\alpha = -\frac{\sin \theta_\alpha}{q_\alpha^2} - \frac{\lambda \ell_{T\alpha}}{2} \left[ \eta (u_\infty + \eta + \delta - 1) + \lambda P(\eta) \right].$$

(45.a)

Similarly:

$$\frac{(1 - \eta) \lambda}{2} \bar{\xi}_\beta = -\frac{\sin \theta_\beta}{q_\beta^2} + \frac{\lambda \ell_{T\beta}}{2} \left[ (1 - \eta)(u_\infty + \eta + \delta - 1) - \lambda P(\eta) \right].$$

(45.b)

So that:

$$\frac{\Delta T(\lambda)}{G} = \frac{2}{\lambda} \left( \frac{\sin \theta_\alpha}{q_\alpha^2} + \frac{\sin \theta_\beta}{q_\beta^2} \right) + \lambda \left( \ell_{T\alpha} + \ell_{T\beta} \right) P(\eta) +$$

$$+ (u_\infty + \eta + \delta - 1) \left[ \eta \ell_{T\alpha} - (1 - \eta) \ell_{T\beta} \right].$$

(46)

This expression, together with equation (40) which determines $\eta(\lambda)$ is to be compared with the JH results. Let us recall their procedure: they compute $\bar{\xi}_\alpha, \bar{\xi}_\beta$ by directly integrating equations (27) across each lamella, which, of course, precisely yields expressions (45). However, since they do not solve for $\xi(x)$, they miss the compatibility condition (40). In order to determine $\eta(\lambda)$, they make up for this with the complementary ansatz $\bar{\xi}_\alpha = \bar{\xi}_\beta$.

We are now in a position to check the value of this ansatz: from equations (45), we may calculate $\bar{\xi}_\alpha - \bar{\xi}_\beta$, which is immediately seen to be of the same order as the average undercooling itself. We should now compare the shape of the $\lambda$-dependence of $\Delta T$ as given by (46), (40) with the JH one, which exhibits a minimum for $\lambda = \lambda_{\text{min}}^{\text{JH}}$:

$$(\lambda_{\text{min}}^{\text{JH}})^2 = \frac{2}{P(\eta)} [(1 - \eta) d_\alpha \sin \theta_\alpha + \eta d_\beta \sin \theta_\beta].$$

(47)

Let us calculate:

$$\frac{d(\Delta T)}{d\lambda} = \left( \frac{\partial}{\partial \lambda} + \frac{d\eta}{d\lambda} \frac{\partial}{\partial \eta} \right) \Delta T.$$

(48)
where from equation (40):
\[
\frac{d\eta}{d\lambda} = - \frac{P'('\eta)}{2} \left[ 1 + \frac{\lambda P''('\eta)}{2} \right]^{-1} = - \frac{P'('\eta)}{2} .
\] (49)

Then, up to corrections of order \(\lambda\) and \((d/\ell_T)^{1/2}\), we find (using \(P('\eta) = P(1 - '\eta)\))
\[
G^{-1} \frac{d(\Delta T)}{d\lambda} \approx - \frac{2 \lambda^2}{\ell_T} \left( \frac{\sin \theta_\alpha}{q_\alpha^2} + \frac{\sin \theta_\beta}{q_\beta^2} \right) + \ell_T \lambda U('\eta_0) + \ell_T \eta U(1 - '\eta_0) .
\] (50)

where

\[
\eta_0 = 1 - \delta - u_\infty \quad (51.a)
\]
\[
U('\eta) = P('\eta) - \frac{\eta P'(\eta)}{2} \quad (51.b)
\]

\(U('\eta)\) is easily computed numerically and checked to be positive for all \(\eta\)'s \((0 < '\eta < 1)\). So, \(\Delta T(\lambda)\) also exhibits a minimum, at a wavelength \(\lambda_{\text{min}}\):
\[
\lambda_{\text{min}}^2 = \frac{2}{d_\alpha \ell_T \sin \theta_\alpha + d_\beta \ell_T \sin \theta_\beta} \left( \ell_T \lambda U('\eta_0) + \ell_T \eta U(1 - '\eta_0) \right) .
\]

So, our ansatz-free calculation yields a value of \(\lambda_{\text{min}}\) different from the JH one, but with the same order of magnitude \((\tilde{\lambda}_{\text{min}} \sim (d\ell)^{1/2})\). This results from the fact that, even though the equal undercooling ansatz is not correct, the error on \(\eta\) (of order \(\lambda\)) that is produces does not change the order of magnitude \((\ell_T \lambda)\) of the diffusive contribution to the undercooling.

4. RANGE OF VALIDITY OF THE LARGE GRADIENT APPROXIMATION. — We must now estimate the validity of our central approximation, i.e. of the replacement, in the front equation, of the true diffusion field \(u(x)\) by that of its planar average \(u_0(x) = u_\infty + \eta + \delta - 1 + \phi(x)\).

Setting
\[
u(x) = u_0(x) + \delta u(x)
\] (52)

we estimate \(\delta u(x)\) by replacing, in equation (22), the true \(\zeta\) by its large gradient approximation calculated above
\[
\delta u(x) = \sum_{n,p} \zeta(nK) \Delta(pK) e^{i(p + n)Kx} \left\{ \Pi((p + n + K)K) - \frac{K_0(pK)}{2\pi} [1 - \Pi((n + p + K)K)] \right\} .
\] (53)

Separating out the \(p = 0\) and \(n + p = 0\) terms, and neglecting everywhere \(p \neq 0\) terms such as \(\Pi(pk) \sim \frac{1}{|p| K}\) with respect to 1, we obtain:
\[
\delta u(x) \approx - (\delta + \eta - 1)(\zeta(x) - \bar{\zeta}) + \frac{1}{2\lambda} \int_0^\lambda dx \Delta(x)(\zeta(x) - \bar{\zeta}) + \frac{\lambda}{4\pi} \sum_{n+p \neq 0} \zeta(nK) \Delta(pK) \left( \frac{1}{|n+p|} - \frac{1}{|p|} \right) e^{i(n+p)Kx} .
\] (54)
The second term in the r.h.s. of equation (54) is of order, at most, \( \delta \zeta = |\vec{\xi}_a - \vec{\xi}_\beta| \); the first one is easily checked to be everywhere (including the triple point regions), of order \( |(\delta + \eta - 1) \delta \vec{\xi}| \ll |\delta \vec{\xi}| \). In order to estimate the third term, we replace \( \zeta(nK) \) by the Fourier coefficients of an approximate profile. The simplest choice is a crenel with heights \( \vec{\xi}_a, \vec{\xi}_\beta \) which yields a contribution \( O(\lambda \delta \vec{\xi}) \ll \delta \vec{\xi}. \) We have also tried a finer approximation where, in each lamella, \( \vec{\xi}_i \) is replaced, close to the triple points, by a straight line of slope, e.g., \( \tan \theta_a \), and checked that this does not change the above order of magnitude.

From equations (40), (42), and with the help of conditions (32), (33), \( u_0(x) = O(\sqrt{dT/\ell_T}) \).

On the other hand, from equation (45), \( \delta \vec{\xi} = O(\sqrt{d\ell_T}) \). So the condition of validity, \( \delta u \ll u_0 \), of our approximation on the diffusion term:

\[
\ell_T \ll \ell
\]

is precisely the same as that for the asymptotic matching used to calculate the front profile.

So, we can conclude that:

— The JH approximation for the diffusion field is valid only in the large gradient regime defined by equation (55);

— in this regime, their ansatz is not valid, since the difference between the front undercooling of the two phases is comparable with the global one, \( \Delta T \). However, it is unnecessary, since the large gradient problem can be solved completely, yielding the same qualitative behavior for \( \Delta T(\lambda) \) but a different value for \( \lambda_{\text{min}} \).

4. Search for tilted stationary patterns.

Let us now try to find whether it is possible to find modes of growth with parallel lamellae tilted at an angle \( \Phi \) with respect to the pulling direction \( Oz \) and with space period \( \lambda \) along the \( Ox \) average front direction (Fig. 3). Such a pattern corresponds to a front profile \( \zeta(x - vt) \) which drifts along \( Ox \) at velocity

\[
v = \tan \Phi.
\]

From the triple point equilibrium condition (8), the pinning angles (Fig. 3) measured from the \( x \)-direction are given by:

\[
\phi'_a - \phi = \phi_a + \phi = \theta_a
\]

\[
\phi'_\beta + \phi = \phi_\beta - \phi = \theta_\beta.
\]

So, in contradistinction with the untilted case, the pinning angles at the two ends of a given lamella now have different values. This immediately entails that the profile within a lamella is no longer symmetric with respect to its center: it necessarily develops an odd component, the amplitude of which can be identified with the order parameter of Coullet et al. [6].

One may be tempted, at least in order to get a qualitative description of such states, to resort to the JH approach. As seen above, this amounts to computing the concentration field of the average alternate planar front, i.e. setting in equation (22) \( \zeta(x - vt) = \bar{\zeta} \) (and \( \dot{\zeta}(x - vt) = 0 \), and \( \Delta(x, t) = \Delta(0)(x - vt) \). One immediately obtains (see Eq. (27))

\[
u(x, \bar{\zeta}, t) = u_{\infty} + \eta + \delta - 1 + \phi(x - vt)
\]

i.e., in the quasi-stationary approximation, the diffusion field in the moving frame is the same as that of the untilted problem with the same wavelength.
Then, averaging the Gibbs-Thomson equations over (α) and (β) lamellae, and using the equal undercooling ansatz, one immediately computes an average undercooling ΔΤ(λ, Φ) which only depends on Φ via the values of the pinning angles (57). This suggests that there should be, for a given λ, a continuum of tilted lamellar solutions, in contradiction with the observation of tilt waves with well-defined Φ's.

What has been missed in this simple-minded extension of the JH approach can be understood with the help of the following counting argument. Consider the Gibbs-Thomson equations (9) for our problem. In the drifting frame (x - vt → x) they become time-independent. u(x, ζ(x)) is a functional of the profile ζ(x), i.e., a function of its N(N → ∞) Fourier coefficients ζₙ. The solutions of the two (second-order differential) Gibbs-Thomson equations (9) therefore depend on N + 4 parameters, namely the N ζₙ's and 4 integration constants. Since the α fraction η is not fixed a priori, one thus has N + 5 unknowns. These must be determined by:

- 2 continuity conditions for ζ at the α - β and β - α triple points
- 4 pinning conditions on the profile slopes
- N self consistency conditions for ζ₁, ..., ζₙ, i.e., N + 6 equations.

Eliminating the N + 5 unknowns therefore yields a compatibility condition for the tilt angle Φ, which can only have discrete solutions.

It is now apparent that directly computing a ΔΤ is insufficient to ensure that it is possible to build an acceptable front profile.

That Φ = 0 is a trivial solution of this equation can be checked immediately: in this case, the pinning conditions are symmetric (φₐ,β = φₐ,β), the profile within each lamella is therefore symmetric, so that the two profile continuity conditions degenerate.
Note, however, that we have been considering here a system with isotropic interfacial tensions. When capillary anisotropies are taken into account, two cases must be distinguished:

— if the pulling direction is a direction of high symmetry for both the $\alpha$ and $\beta$ phases, the $\sigma_{\alpha\beta}$'s are symmetric, and the argument still works. There always is an untilted solution;

— if one is not pulling along a common principal axis, the symmetry argument no longer holds, $\Phi = 0$ is no longer a solution. That is, as is well known from experiments, in a misoriented grain lamellae are tilted. Let us however insist that, in general, the observed $\Phi$ has no reason to be strictly equal to the misorientation angle (how much they may differ in practice of course depends on the strength of the anisotropy of $\sigma_{\alpha\beta}$).

Let us now come back to the isotropic problem in the large gradient régime. We set:

$$u(x, \zeta(x)) = u_{\infty} + \eta + \delta - 1 + F(x)$$

where $x$ is now the horizontal coordinate in the frame drifting at velocity $v$, and $F(x)$ is defined by equations (22), (23) in which $\zeta(q, t) \Rightarrow -iq\nu\zeta(q)$. We then solve formally the Gibbs-Thomson equations with the same procedure as in § (III.2a-c). In the $x = 0$ edge region and in the center of the $\alpha$ lamella for example, $\zeta''(x)$ is given by equations (34), (35.a) where $\theta_{a} \rightarrow \phi_{a}$; $\phi(x) \rightarrow F(x)$. The asymptotic matchings at the two edges ($x = 0, x = \eta\lambda$) determine the integration constants $\mu_{a}, \nu_{a}$ (and analogously, $\mu_{\beta}, \nu_{\beta}$). We then must impose the continuity conditions (25.a). Using the analogues of expressions (39) for $\zeta_{i}(0)$, $\zeta_{i}(\eta\lambda)$, we get two coupled equations for $\eta$ and $\Phi$:

$$u_{\infty} + \eta + \delta - 1 + \frac{F(0) + F(\eta\lambda)}{2} = (\ell_{Ta} + \ell_{Tb})^{-1} \cos \frac{\Phi}{2} \left[ \frac{2}{q_{\beta}} \sin \theta_{\beta} - \frac{2}{q_{a}} \sin \theta_{a} \right]$$

$$4 \sin \frac{\Phi}{2} \left( \frac{1}{q_{a}} \cos \frac{\theta_{a}}{2} + \frac{1}{q_{\beta}} \cos \frac{\theta_{\beta}}{2} \right) = (\ell_{Ta} + \ell_{Tb})(F(0) - F(\eta\lambda))$$

In the $\Phi = 0$ limit, where lamellae profiles are symmetric and $v = 0$, $F(0) = F(\eta\lambda)$ and, while (60.a) determines $\eta$, equation (60.b) is trivially satisfied, as already seen from the counting argument.

When $\Phi \neq 0$, we must compute $F$ by plugging into equation (22) an approximate expression $\zeta_{ap}(x)$ for $\zeta(x)$ depending on a finite number of parameters, which must then be determined by self-consistency conditions.

The simplest approximation is, of course, as in the untilted case, the JH one: $\zeta_{ap}(x) = \tilde{\zeta}$. However since, as seen in § 3, the corresponding $F(x)$ is simply the symmetric JH field $\phi(x)$, equation (60.b) simply yields: $\sin \frac{\Phi}{2} = 0$, i.e. the planar front approximation produces no tilted solution.

We must then try to improve upon this by choosing a shape for $\zeta_{ap}(x)$ which describes more precisely the true profile, which, as discussed above, contains within each lamella, an odd component driven by the asymmetry of the pinning conditions. This we have done by choosing for $\zeta_{ap}(x)$ the shape shown in figure 4. The straight line edge profiles are the tangents to the meniscus ones at the triple points; the central straight lines:

$$\tilde{\zeta}_{ap}(x) = \zeta_{i} + p_{i}(x - X_{i})$$

depend on the four quantities:

$$\zeta_{i}, p_{i}.$$
A tedious but straightforward calculation analogous to that of § (3.4) yields, to lowest order in $\ell_T$, $\lambda$, the expression of $F(x)$ in terms of the $\zeta_i$, $p_i$. These are then made self consistent, from which the final expression of $F(0) - F(\eta \lambda)$ is obtained. We do not give here the corresponding detailed result since, as will now appear, its only important feature is its order of magnitude. This we find to be:

$$\Delta F = |F(0) - F(\eta \lambda)| = O(q^{-1})$$

which can be easily understood qualitatively. Since $\lambda \sim \lambda_{\text{min}} \sim \sqrt{d}$, $q^{-1} \sim \lambda \sqrt{\ell_T}$. So (61) simply expresses the fact the amplitudes of odd profile deformations (and therefore of odd concentration modulations) are squeezed by the large thermal gradient.

The r.h.s. of equation (60.b), $O(\ell_T/q)$, is of the order of terms which have been neglected in our approximation and must itself be neglected. So, in the large gradient limit, the only solution of equation (60.b) is the untilted one.

5. Conclusion.

The conclusion of our analysis are therefore somewhat disappointing: it is only in the large gradient limit that the JH approach, improved so as to become consistent, can be justified. In this limit, due to the fact that front deformations are restricted to small values, no tilted lamellar stationary state can exist.

From (Eq. (55)), together with (11), this regime corresponds to

$$G \gg \frac{V}{D} m(\Delta C).$$

In practice, pulling velocities in lamellar growth experiments are at least of the order of 1 $\mu$m/s, while $D \sim 10^{-5}$ cm$^2$/s. For CBr$_4$-C$_2$Cl$_6$ for example $m \Delta C \sim 20$ K. So, in practice, the large gradient regime corresponds to

$$G \gg 200 \text{ K/cm}.$$

So, it will unfortunately be very difficult for experiments to reach the large gradient regime. So if one wants for example to compare in detail measured front profiles or undercooling with calculated ones numerical solutions of the growth equations will be needed.
On the other hand, for values of $G$, $\sim 100$ K/cm, comparable to those in the experiments [3] where « tilt waves » have been observed, condition (62) implies:

$$V \ll 0.5 \, \mu\text{m/s}.$$  

So, our above negative conclusion is compatible with the experimental observation of a velocity threshold below which tilt waves do not exist.

It is clear that again more precise predictions about these patterns will demand numerical studies: indeed, as shown by the discussion of § 4, the relevant parameter which controls their possible existence is $\ell_T$. It is clear that it is only for $\ell_T \geq 1$ that the system can tolerate (odd) front deformations large enough for such solutions to become possible. At this stage, we can only infer from our analysis that, most likely, the threshold for a tilted growth mode to appear should be given by $\ell_T/\ell = \text{cst}$, i.e. the threshold velocity should then be proportional to the thermal gradient. Quantitative experimental studies of threshold velocities which are presently in progress [17] should permit to check on this conjecture.

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References