Similarity Laws in Eutectic Growth

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(Received 12 October 1990)

We show that the full boundary integral equation reduces, for small Péclet numbers, to a nonlinear similarity equation which contains only two dimensionless parameters, $\sigma = d_0 l/\lambda^2$ and $\chi = l/l_T$, where d_0, l, and l_T are the capillary, diffusion, and thermal lengths and λ is the wavelength of the pattern. All physical quantities should depend on σ and χ only. This is confirmed by numerically integrating the original equations. The selected wavelength should scale as $\lambda \sim V^{-1/2} f(l/l_T)$ (V is the velocity). Our results suggest that $\lambda \sim V^{-\alpha}$, where $\alpha \simeq \frac{1}{2}$ for large V and $\alpha < \frac{1}{2}$ for small V. Our results agree with experiments and we propose further experimental tests.

PACS numbers: 61.50.Cj, 05.70.Fh, 68.70.+w, 81.30.Fb

When eutectics are submitted to directional solidification, the liquid-solid interface forms parallel lamellae of the two coexisting solid phases.[1] Under some conditions the interface may undergo a parity-breaking transition:[2] Tilted lamellae appear. We have shown recently[3] that the fully isotropic model of eutectic growth supports tilted solutions. We found that, for not too small growth velocities V ($l_T/l > 4$, l and l_T being the diffusion and thermal lengths), the tilt angle ϕ remains approximately constant along the line $\lambda^2 V=$const, where λ is the periodicity. A further prediction was that parity breaking occurs at a critical velocity V_c which approximately scales as $V_c \sim \lambda^{-2}$. On the other hand, various experiments reported that the selected wavelength, or the average spacing, scales within experimental precision as $\lambda \sim V^{-1/2}$. The same scaling holds at the minimum undercooling point[4] in the Jackson and Hunt[4] approximation. This point has often been suspected to be the operating point. It is therefore natural to ask whether the scalings encountered in different contexts can be understood on the basis of general considerations. The primary goal of this Letter is to deal with this question. Indeed, we show that in the experimental range of parameters where the Péclet number, $P = \lambda/l$, is much smaller than unity, the full integral equation reduces to a similarity equation containing only two dimensionless parameters $\sigma = d_0 l/\lambda^2$ and $\chi = l/l_T$, where d_0 is the capillary length. The similarity is confirmed by numerically integrating the full integral equation. The dependence on χ is smooth only for large enough velocities, thus indicating that the relevant parameter in this regime is σ ($\simeq \lambda^2 V$). We will see below that for small enough V, however, the presence of the thermal gradient causes a departure from the relation $\lambda^2 V=$const. Such an effect was observed in many experiments.[5] Indeed, for small enough V power laws $\lambda \sim V^{-\alpha}$, with $\frac{1}{4} < \alpha < \frac{1}{3}$, were found. Surprisingly, in most of the literature, essentially in the 1980's, such results are not mentioned. An important result to emerge from our considerations is that the selected wavelength λ should scale as $\lambda \sim V^{-1/2} f(l/l_T)$. We compute f for the *minimum undercooling* point for symmetric solutions. This point is suspected to be the operating point. If such is the case then our results suggest that the selected wavelength scales as $\lambda \sim V^{-\alpha}$, with $\alpha \simeq \frac{1}{2}$ at large V and α decreasing with V at small V. Our results agree well with experiments and inspire alternative tests. We will also comment on the great progress offered by the similarity properties in numerical investigations.

For ease of presentation, our *analytical discussion* will be restricted to axisymmetric growth. The extension to asymmetric lamellae is straightforward. Furthermore, we assume that $d_0^\alpha = d_0^\beta$, $l_T^\alpha = l_T^\beta$. This allows our formulas to be compact. Since d_0 and l_T differ in both phases by multiplicative constants only, this assumption does not alter at all the scaling laws.

Our starting point is the integral equation that governs one-dimensional deformations of the solidification front. For the one-sided model it can be written as[6]

$$\frac{\epsilon[\sigma\kappa(x)+\chi\zeta(x)]}{2} = \frac{u_\infty}{P} - \frac{P}{2\pi}\int_{-\infty}^{\infty} dx' \left[\epsilon(x')(2k_v-1)[\sigma\kappa(x')+\chi\zeta(x')] - \frac{2\Upsilon(x')}{P}\right] e^{-P\Delta\zeta} K_0(P\rho)$$
$$+ \frac{P}{2\pi}\int_{-\infty}^{\infty} dx' \frac{\Delta\zeta - \Delta x \zeta_{x'}}{\rho}\epsilon(x')[\sigma\kappa(x')+\chi\zeta(x')] e^{-P\Delta\zeta} K_1(P\rho)\,. \quad (1)$$

Lengths are scaled by the wavelength λ. $\epsilon = 1,-1$ and $\Upsilon = \delta, \delta-1$ for the α,β phase, respectively, where $\delta = (c_e - c_\alpha)/(c_\beta - c_\alpha)$, c_e, c_α, c_β being the equilibrium concentrations at the eutectic temperature of the coexisting liquid, the α, and the β phase, respectively. Note that the β phase is the one with the higher concentration. $u_\infty = (c_\infty - c_e)/(c_\beta - c_\alpha)$ measures the departure from the eutectic concentration and k_v is the partition coefficient for the coexistence of the liquid and the v solid ($v=\alpha,\beta$). The K_i's are the modified Bessel functions, $\Delta x = x - x'$, $\Delta\zeta = \zeta(x) - \zeta(x')$, and $\rho = (\Delta x^2 + \Delta\zeta^2)^{1/2}$.

Pattern formation occurs in standard experiments with wavelengths λ that are much smaller than the diffusion length $l=2D/V$. Typically $P=\lambda/l \sim 10^{-2}$. We therefore concentrate on this situation. Inspection of Eq. (1) shows that there are three types of terms. We consider them in the order of increasing difficulty. Expressions of the first type vanish in the small-P limit as $P\ln(P)$. Such contributions originate from the terms proportional to $(\sigma\kappa+\chi\zeta)K_0$. The next sort stems from the terms in the second integral on the right-hand side of Eq. (1). Using the small-argument expansion of K_1 we obtain order-1 contributions. Finally, the most subtle contribution comes from $\int_{-\infty}^{\infty} dx' \Upsilon(x') K_0(P\rho) e^{-P\Delta\zeta}$. The integrand of this term obviously diverges logarithmically in the $P \to 0$ limit. We circumvent this difficulty as follows. We add to and subtract from this term the quantity $\int_{-\infty}^{\infty} \Upsilon(x') K_0(|P\Delta x|)$. The subtraction serves to cancel the logarithmic singularity. The added part can be evaluated exactly by first expanding the periodic function $\Upsilon(x')$ in a Fourier series. The resulting integrals of the form $\int_{-\infty}^{\infty} dx' e^{2i\pi n x'} K_0(|P\Delta x|)$ are standard and can be evaluated analytically. Taking now the $P \to 0$ limit we obtain the similarity equation

$$\epsilon(\sigma\kappa+\chi\zeta) = h(\sigma,\chi) - \frac{1}{\pi}\int dx' H(x') \ln\left[\frac{(x-x')^2+(\zeta-\zeta')^2}{(x-x')^2}\right] + \sum_{n=1}^{\infty}\frac{\sin(\pi n\eta)\cos(2\pi n x)}{\pi^2 n^2} - \frac{1}{2\pi}\int dx' \frac{\zeta-\zeta'+(x-x')\zeta_{x'}}{(x-x')^2+(\zeta-\zeta')^2}(\sigma\kappa+\chi\zeta)\Upsilon, \tag{2}$$

where η is the volume fraction of the α phase,

$$H(x) = \sum_{n=1}^{\infty}\frac{\sin(\pi n\eta)\cos(2\pi n x)}{\pi n} + \eta + \delta - 1,$$

and $h(\sigma,\chi) = (u_\infty + \eta + \delta - 1)/P$. The latter quantity which *appears* to be of order $1/P$ is in fact of order unity. Indeed we can show from the mass conservation law on the global scale that

$$u_\infty + \eta + \delta - 1 = P\int_0^1 dx\, k_v(x)\epsilon(x)[\sigma\kappa(x) + \chi\zeta(x)]. \tag{3}$$

This equation is obtained by integrating the local mass conservation equation over a surface (this is a volume per unit length in the y direction since we consider only one-dimensional deformations) bounded by the following contour: a segment parallel to the x axis lying at $z=\infty$ and having a length λ, and a boundary of extent λ along the interface on the solid side. To close the contour the ends of these two boundaries are joined by two segments parallel to the z axis. Note that to leading order in an expansion in the Péclet number we can set, in the integration boundaries, $\eta = 1 - u_\infty - \delta$, so that $(u_\infty + \eta + \delta - 1)/P \equiv h(\sigma,\chi)$, which is a function of σ and χ only. We should mention that in deriving Eq. (2) we have used the fact that since interface excursions are limited to one wavelength at most, the quantity $\Delta\zeta \simeq 1$ (in units of λ).

Equation (2) constitutes the similarity equation which contains only two dimensionless parameters $\sigma = d_0 l/\lambda^2 = 2Dd_0/\lambda^2 V$ and $\chi = l/l_T = 2DG/Vm(c_\beta - c_\alpha)$, where G is the applied thermal gradient and m is the absolute value of the liquidus slope. The similarity equation implies that the physical quantities, in particular the pattern, are self-similar under a simultaneous stretching (or shrinkage) of λ and of (V,G) by α and α^{-2}, respectively, where α is an arbitrary real number.

We have tested our results numerically. The numerical procedure was presented in recent work.[3] For definiteness, we have used the material parameters for the CBr_4-C_2Cl_6 material which are detailed in Table I of Ref. 3. Figure 1 shows the dependence of the position in the z direction of the triple point along the line $(\sigma,\chi)=$const, compared to that obtained when only σ is kept constant (this means $\lambda \sim V^{-1/2}$). The same has been done for the tilt angle of asymmetric solutions[3] (Fig. 2). It is clear that these physical quantities are functions of σ and χ only. Already from Fig. 2 we observe that the hitherto suspected scaling, according to which $\lambda \sim V^{-1/2}$, is accurate only at sufficiently large V (when l_T is appreciably larger than l). When V is small a deviation from the exponent $\frac{1}{2}$ is noticed. We can even go farther just by looking at Fig. 2: At small V the exponent α (with the proviso that a simple algebraic law holds) should be smaller than one-half. This idea is motivated by first noticing that ϕ increases with growing V at small V when only the product $\lambda^2 V$ is kept constant,

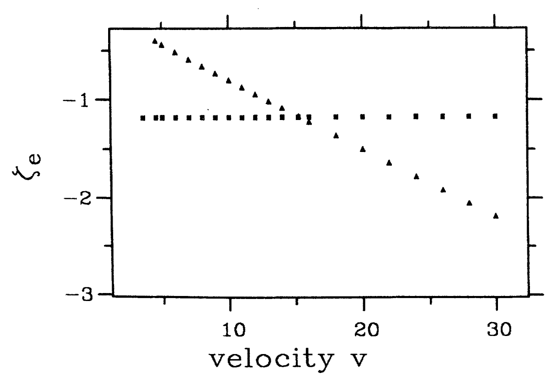

FIG. 1. The triple-point position as a function of the growth velocity. Squares: at fixed (σ,χ). Triangles: at fixed (σ,l_T) (see text).

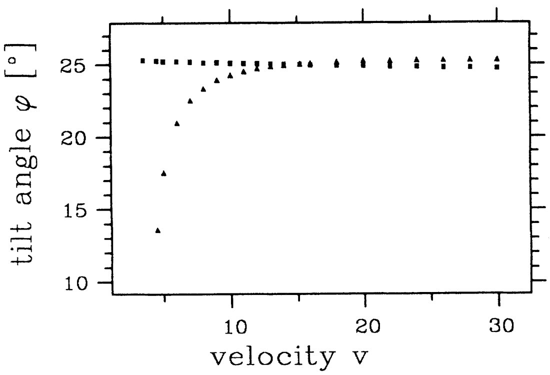

FIG. 2. The tilt angle as a function of the growth velocity. Squares: at fixed (σ,χ). Triangles: at fixed (σ,l_T) (see text).

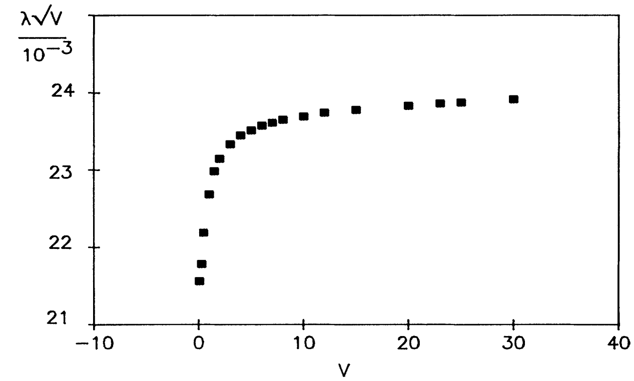

FIG. 3. The dependence of $\lambda V^{+1/2}$ on the growth velocity.

and second, that ϕ is expected to decrease when the thermal gradient increases. The last point can be understood by noting that an increase in ϕ implies an increasing distortion of the interface of (mainly) the thinner lamella. This implies excursions of the interface towards the hot thermal contact. The thermal gradient always acts against such a tendency. To complete our argument we mention that thanks to the similarity property, and when σ is kept constant, a decrease in V implies a variation of the parameter χ which is equivalent to an increase by the same amount in the thermal gradient. More results regarding the similarity properties will be given in a forthcoming publication.

An important consequence of the similarity equation (2) is that the wavelength of the pattern λ should scale as

$$\lambda \sim V^{-1/2} f(l/l_T). \tag{4}$$

It does not matter *how* the wavelength selection mechanism operates. Our result follows from dimensional considerations only. At this stage Eq. (4) does not tell us what the behavior of the selected wavelength (if any) is. It simply gives the form of the scaling law. We need additional information to determine f. We have computed f at the minimum undercooling point. This point has conventionally been assumed in the metallurgical literature to locate the operating point; and the resulting predictions seem to be in good agreement with experiments.

From our calculation we conclude that f is an increasing function of V and saturates at large V. Our results suggest that the selected wavelength follows this scaling. Using the computed f we obtain the dependence of the wavelength as a function of the velocity. Figure 3 shows this behavior. More precisely Fig. 3 shows the dependence of $\lambda^2 V$ as a function of V. At large V this quantity is approximately constant. However, as V decreases a deviation from the conventional law ($\lambda \sim V^{-1/2}$) is noticed. At small V the wavelength is definitely smaller than what is predicted from the Jackson-Hunt theory. Or, in other words, we can say that λ in this regime can be represented by $\lambda \sim V^{-\alpha(V)}$, where $\alpha(V) < \frac{1}{2}$. It is understood that algebraic laws of this type constitute only reasonable fits within a precision (about 1%–5%) beyond experimental resolution.

Our results are in good agreement with different experimental findings. Indeed careful experimental investigations[5] have shown that the law $\lambda \sim V^{-1/2}$ holds only at sufficiently large velocities. At small velocities, however, λ is observed to scale as $V^{-\alpha}$ with $\alpha \simeq 0.3$–0.35. We propose an additional experimental test: In those materials[5] where it has been possible to explore the small-V regime it would be interesting to investigate the wavelength selection (or the average lamella spacing) by keeping the ratio of the pulling speed to the thermal gradient constant. Indeed our scaling law [Eq. (4)] implies that in that case $\lambda \sim V^{-\alpha}$ with $\alpha = 0.5$ instead of 0.3 in the whole range of velocities since f is constant. This is a rather large difference which can, in our opinion, easily be detected.

Before concluding we would like to point out the great progress offered by the similarity properties in numerical investigations. As the velocity decreases (the diffusion length increases) the range of interaction between lamellae becomes longer and longer. This means that the effective size of the interface increases, thereby increasing the computing time by the square of the same factor. However, thanks to the similarity equation, decreasing V by a certain factor is equivalent to a simultaneous multiplication of the capillary length and the thermal length by the same factor. As these lengths enter the local dynamics only, they do not affect the effective size of the interface. This procedure constitutes a considerable savings of computing time.

In summary, we have shown using the boundary integral formulation that the interface equation reduces to a similarity equation in the small-Péclet-number limit. This limit is relevant to experiments. The similarity properties are confirmed numerically. We have given a general form for the scaling of the wavelength of the pattern. We have found that only for large enough velocities is the law $\lambda \sim V^{-1/2}$ valid. For small velocities we found $\lambda \sim V^{-\alpha(V)}$, where $\alpha < \frac{1}{2}$. This agrees with experiments on metallic eutectics.[5] The general analysis presented here indicates that if the ratio of the thermal gradient G to the pulling velocity V is kept constant in an experiment, the wavelength should scale as $\lambda \sim V^{-1/2}$. Therefore, performing experiments with a fixed G/V for those materials for which[5] an exponent of about 0.3–0.35 has been observed at a given G would constitute an important experimental test of our results. This work is also a call for careful experiments on transparent eutectics in the small-velocity regime.

One of us (C.M.) is grateful to C. Caroli and A. Ghazali for stimulating discussions. During this work C.M. visited the Forschungszentrum of Jülich for a short period. He acknowledges the hospitality and the financial support. Groupe de Physique des Solides, Université Paris VII, is associated with CNRS.

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