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LIMIT OF ABSOLUTE STABILITY FOR CRYSTAL GROWTH INTO UNDERCOOLED ALLOY MELTS

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Abstract—The limit of absolute stability for crystal growth of binary alloys into an undercooled melt is discussed in respect of a positive temperature gradient in the solid and different thermal properties of liquid and solid. It is shown that the positive temperature gradient and the better heat conductivity and diffusivity of the solid phase reduce the velocity corresponding to the limit of absolute stability. Thus although the solid grows into an undercooled melt a planar interface may be stable with a quite lower growth rate than the absolute thermal velocity.

Résumé—La limite de stabilité absolue de la croissance de cristaux d'alliages binaires dans le liquid sousrefroidi a été discuté en tenant compte d'un gradient de température positif dans le solide et des propriétés thermiques différentes du liquide et du solide. Il a été montré qu'un gradient de température positif et une conductivité resp. diffusivité thermique supérieure du solide diminuent la vitesse de stabilité absolue. Dans ces conditions lors de la croissance du solide dans le liquide sousrefroidi par une interface plane est stable á des vitesses de croissance inférieures á la 'vitesse thermique absolue'.

Zusammenfassung—Unter Berücksichtigung eines positiven Temperaturgradienten in der festen Phase und unterschiedlicher thermischer Eigenschaften von Schmelze und Festkörper wird die Grenze für die absolute Stabilität beim Kristallwachstum von binären Legierungen in unterkühlte Schmelzen diskutiert. Es wird gezeigt, daß aufgrund eines positiven Temperaturgradienten und einer besseren thermischen Leitfähigkeit bzw. einer höheren Temperaturleitzahl des entstehenden Kristalls, die Geschwindigkeit bei der absolute Stabilität auftritt, erniedrigt ist. Daher kann eine ebene Erstarrungsfront mit weitaus geringerer Geschwindigkeit als der 'absolute thermal velocity' stabil in eine unterkühlte Schmelze wachsen.

1. INTRODUCTION

Based on the linear perturbation theory of Mullins and Sekerka [1] for the stability of a planar interface, Trivedi and Kurz [2] have extended this stability analysis for the conditions of conventional and rapid solidification. There results show that the absolute stability of a planar interface at large growth rate exists not only under constrained growth condition but also for growing into undercooled pure and alloy melts. With the assumption that the thermal conductivities K_l and K_s and the thermal diffusivity a_l and a_s in liquid and solid are equal, they found that the absolute velocity under constrained growth condition, above which a planar interface in the presence of solute is stable is

$$(V_{abs})_C = \frac{D\Delta T_0}{\Gamma k} \qquad (1)$$

where D is the solute diffusion coefficient, ΔT_0 is the temperature difference between liquidus and solidus at the initial solute content C_0, Γ is the Gibb's Thomson coefficient and k is the equilibrium distribution coefficient. $(V_{abs})_C$ is called the solute absolute velocity. This absolute stability criterion is the same as that obtained by Mullins and Sekerka.

For growing into undercooled alloy melts assuming $G_s = 0$, $K_s = K_l$ and $a_s = a_l$ they obtain

$$V_{abs} = \frac{D\Delta T_0}{\Gamma k} + \frac{a_l \Delta H}{\Gamma C_p}$$

or

$$V_{abs} = (V_{abs})_C + (V_{abs})_T \qquad (2)$$

where ΔH is the latent heat of fusion per unit volume and C_p is the specific heat. $(V_{abs})_T$ is called the thermal absolute velocity. Thus in this case the limit of absolute stability consists of two contributions the solute and the thermal absolute velocity. Generally the thermal absolute velocity is larger than the solute absolute velocity by orders of magnitude.

In the case of an undercooled melt solidifying on a cold substrate a large positive temperature gradient in the solid may be present at the dendrite tip. Such conditions are realized during the splat cooling of levitated and undercooled droplets or at the bottom of a drop tower, provided that the droplets have not been solidified while reaching the bottom. Furthermore this situation may occur in rapid solidification techniques like melt spinning or splat cooling where experimental observations on microstructures can be explained if high undercooling is assumed [3].

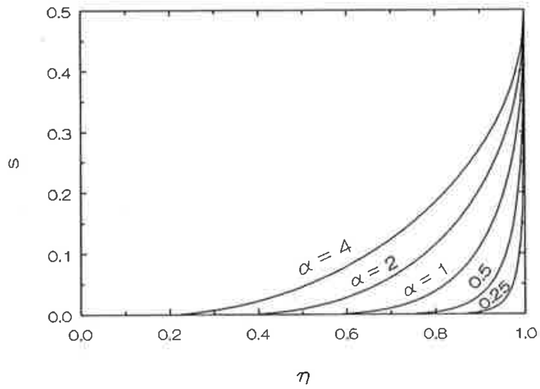

Fig. 1. The maximum s of the stability condition (12) is plotted over η for different values of $\alpha(=\beta)$.

Inserting the expressions for γ and χ into equation (12) the condition transforms to $V \geqslant V_{abs}$, where

$$V_{abs} = \frac{D\Delta T_0}{\Gamma k} + 2s\frac{a_l \Delta H}{\Gamma C_p}$$

or

$$V_{abs} = (V_{abs})_C + 2s(V_{abs})_T \quad (14)$$

for the stable growth of a planar interface.

3. DISCUSSION

As it is shown in Figs 1 and 2 s has a maximum of 0.5 at $\eta = 1$ (i.e. at $G_s = 0$) independent of the values α and β leading to the same condition of absolute stability as the one prescribed by equation (2).

Assuming that 10% of the heat of fusion is transported into the solid, $s \approx 0.1$ reducing V_{abs} to about 20% of the value corresponding to $G_s = 0$. Increasing further the portion of the heat of fusion transported into the solid it has been found that the contribution of the thermal absolute velocity becomes negligible (at 50% and above). Thus an efficient cooling of the solid may compensate the destabilizing effect of the negative gradient in the melt.

An additional stabilising effect may occur if $G_s > 0$ and the thermal conductivity and diffusivity of the solid are considerably better than those of the liquid (Fig. 2; $\alpha < 1$). Let's assume that $\eta = 0.9$ and that the thermal properties of the solid are twice as good as of the liquid (i.e. $\alpha = 0.5$, which is very common) then we get $s \approx 0.04$ in contrast to $s \approx 0.1$ for the case with equal thermal properties; that is a reducing of another 60%. On the other hand the interface may be destabilized again if the liquid has better thermal properties (Fig. 2, $\alpha > 1$). But even when the thermal properties of the liquid are four times better than those of the solid (which is very uncommon), the stabilizing effect of a positive G_s predominates.

To understand the stabilising influence of G_s and the different thermal properties more deeply, the simple approach shown in Fig. 3 is considered, in which the velocities of the highest and the lowest points at a slightly perturbed interface are compared. For a stable growth of the interface $\Delta V = V_A - V_B$ must be negative, where V_A and V_B are the velocities at points A and B, respectively. Using $\Delta G_s = G_s|_A - G_s|_B$ and $\Delta G_l = G_l|_A - G_l|_B$ results in

$$\Delta V = (K_s \Delta G_s - K_l \Delta G_l)/\Delta H. \quad (15)$$

As shown in the paper of Trivedi and Kurz the temperature field ahead of the perturbed interface is given by

$$T_l - T_0 = (G_l a_l / V)(1 - \exp(-V/a_l \cdot Z)) + \delta(-\Gamma\omega^2 - G_l)\sin(\omega X)\exp(-\omega_l Z) \quad (16)$$

where ω_l is defined in equation (20b). Let's denote the perturbed interface with Φ, it is obtained by derivating T_l with respect to Z and regarding that the perturbation is infinitesimal that

$$G_l|_\Phi = G_l + [-G_l(V/a_l) - \omega_l(-\Gamma\omega^2 - G_l)]\,\delta\,\sin(\omega X) \quad (17)$$

and for $\Delta G_l = G_l|_A - G_l|_B$

$$\Delta G_l = 2\delta\omega_l[\Gamma\omega^2 + G_l(1 - (V/a_l\,\omega_l))]. \quad (18)$$

With a similar derivation for the difference in the solid gradients at A and B it is obtained that

$$\Delta G_s = 2\delta\omega_s[-\Gamma\omega^2 + G_s(-1 - (V/a_s\omega_s))] \quad (19)$$

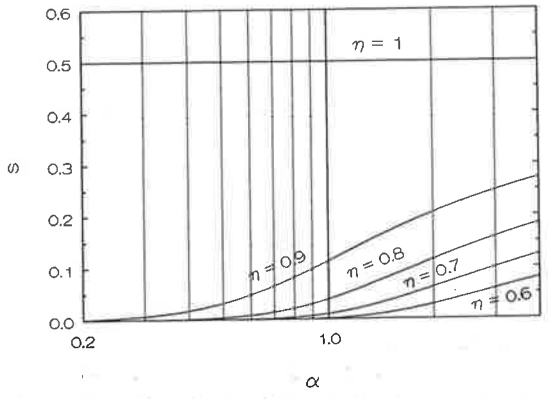

Fig. 2. The maximum s of the stability condition (12) is plotted over $\alpha(=\beta)$ for different values of η.

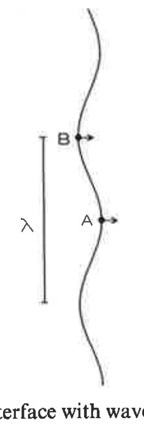

Fig. 3. A perturbed interface with wavelength λ. A and B are the highest and the lowest points on the interface.