

# Morphological stability of a planar interface subject to rapid quenching

A. Ludwig\*, K. Greven, P.R. Sahn

*Giesserei-Institut der RWTH Aachen, 52056 Aachen, Germany*

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## Abstract

Rapid quenching of a solid–liquid interface is a common method of investigating solidification morphologies and the distribution of solute during crystallisation. As the destabilisation of the interface needs a certain but up to now unknown time interval, the quantities finally observed might change during the quenching process. In this paper the destabilisation of a planar interface with respect to a suddenly acting cooling rate is investigated. A linear stability analysis is performed considering the time-dependent concentration profile in front of an infinitesimal perturbed planar front, as superposition of the base state and a perturbation. Both profiles are estimated numerically. The time interval necessary for destabilisation is calculated as a function of wavelength. A comparison with the predictions of the analysis of Sekerka and Coriell (and Mullins and Sekerka) using the instantaneous value of the concentration gradient and solidification velocity is performed. © 1998 Elsevier Science B.V. All rights reserved.

*Keywords:* Rapid quenching; Morphological stability; Linear-stability analysis

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## 1. Introduction

Rapid quenching is a widely used method of investigating solidification morphologies and solute distributions during crystallisation [1–4]. As the destabilisation of the solidification front takes a finite time interval, the observed quantities, such as the fraction of solid, might have changed during the quenching process. The aim of this paper is to

estimate this time interval and the distance which the solidification front grows until destabilisation takes place.

We used a simple model to calculate the destabilisation of a planar solidification front during the quenching processes. The time-dependent solidification velocity,  $V$ , the interfacial solute concentration gradient,  $G_C$ , and the perturbation growth rate,  $\delta$ , are calculated numerically. The time interval until instability occurs is calculated as a function of perturbation wavelength  $\lambda$ . The results are compared with the analysis of Sekerka and Coriell (SC analysis) [5, 6], which is a slightly

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\* Corresponding author. E-mail: ludwig@gi.rwth-aachen.de.

and the change of the base-field concentration due to the variation of the position from  $z = 0$  to  $z = \delta$ .

To estimate the terms in Eq. (3) the concentration profile of the base state and the perturbation are solved numerically. The boundary condition at the interface for  $C^0$  is determined by applying a constant cooling rate,  $\dot{T}$ , and considering thermodynamic equilibrium

$$C^0|_{z=0} = C_\infty + \frac{\dot{T}}{m} t, \quad (4)$$

where  $C_\infty$  is the initial solute concentration in the melt and  $m$  the liquidus slope. The boundary condition at the end of the numerical domain is chosen to be satisfied by the steady-state solute concentration.

The boundary condition for  $C^1$  at the interface results from the assumption that the unperturbed and perturbed interfaces are at the same temperature, the capillary condition and the approximations mentioned above:

$$m(C^1|_{z=0} + \delta G_C) - \Gamma \omega^2 \delta = 0, \quad (5)$$

where  $\Gamma$  is the Gibbs–Thomson coefficient. At the end of the numerical domain  $C^1$  is considered to vanish.

### 3. Numerical solution

To formulate the numerical algorithm, we introduce a change of coordinates so that the unperturbed solid–liquid interface is at a fixed position in the new coordinate system. The resulting differential equation is spatially discretised with second-order accuracy on a nonuniform mesh in the spatial variable  $z$ . The grid spacing  $\Delta z_i$  is calculated recursively by  $\Delta z_i = a \Delta z_{i-1}$  with  $a = 1.005$ .  $\Delta z_0$  is defined by using  $n = 1500$  mesh points on a domain of length  $l = 500 \mu\text{m}$  which ensures that the concentration fields are not affected by the domain boundary. A time step of  $\Delta t = 2 \times 10^{-6}$  s is used. It was found that the result of the calculations is independent of the time step and the number of mesh points as long as  $\Delta t$  is not increased by at least more than a factor of 2 and  $n$  not reduced by at least more than 50%. The differential equations

were solved using the implicit tridiagonal matrix algorithm (TDMA) technique [10].

To increase the numerical efficiency, the base state is calculated alone from  $t = 0$  to  $t = t_1$ , and then a finite perturbation amplitude  $\delta$  is introduced and the perturbation field is estimated. The solidification velocity of the unperturbed interface  $V$  is calculated from the corresponding solute-flux balance. The growth rate  $\dot{\delta}$  is evaluated by using Eq. (3). Neither the perturbation profiles nor the time dependence of  $\dot{\delta}$  was found to be influenced by the choice of  $\delta$  or  $t_1$ .

### 4. Results and discussion

Prior to quenching, a flat interface at rest with a constant concentration in the liquid,  $C_\infty$ , and a constant concentration in the solid,  $kC_\infty$ , is considered, where  $k$  is the distribution coefficient. The temperature is assumed to be uniform at  $T_\infty = T_f + mC_\infty$ , where  $T_f$  is the melting point of the pure liquid and  $m$  the liquidus slope. During quenching, the temperature of the whole system is decreased with a constant cooling rate  $\dot{T}$ . Therefore, the assumption of thermodynamic equilibrium at the interface leads to Eq. (4). A cooling rate of  $\dot{T} = 100$  K/s is considered.

Fig. 1 shows the evolution of the solidification velocity of the unperturbed interface  $V$  and the corresponding interfacial solute gradient  $G_C$  as a function of time. Note that  $V$  is directly coupled with  $G_C$  through the flux balance. While the liquid concentration at the interface increases linearly with time (Eq. (4)) the increase of  $G_C$  is more complex. The time evolution of the concentration at a certain distance from the interface is characterised by the competition between the convective and the diffusive terms in the differential Eq. (1). While the diffusive term predominates initially the convective term becomes dominant with increasing time. Together with the monotonous increase of interfacial liquid concentration, this causes the boundary layer thickness to decrease and  $G_C$  to increase nonlinearly with time. Of course, at very high  $V$ , the assumption of a spatial uniform temperature is no longer valid and the growth velocity is limited by the appearing thermal gradients at the interface.

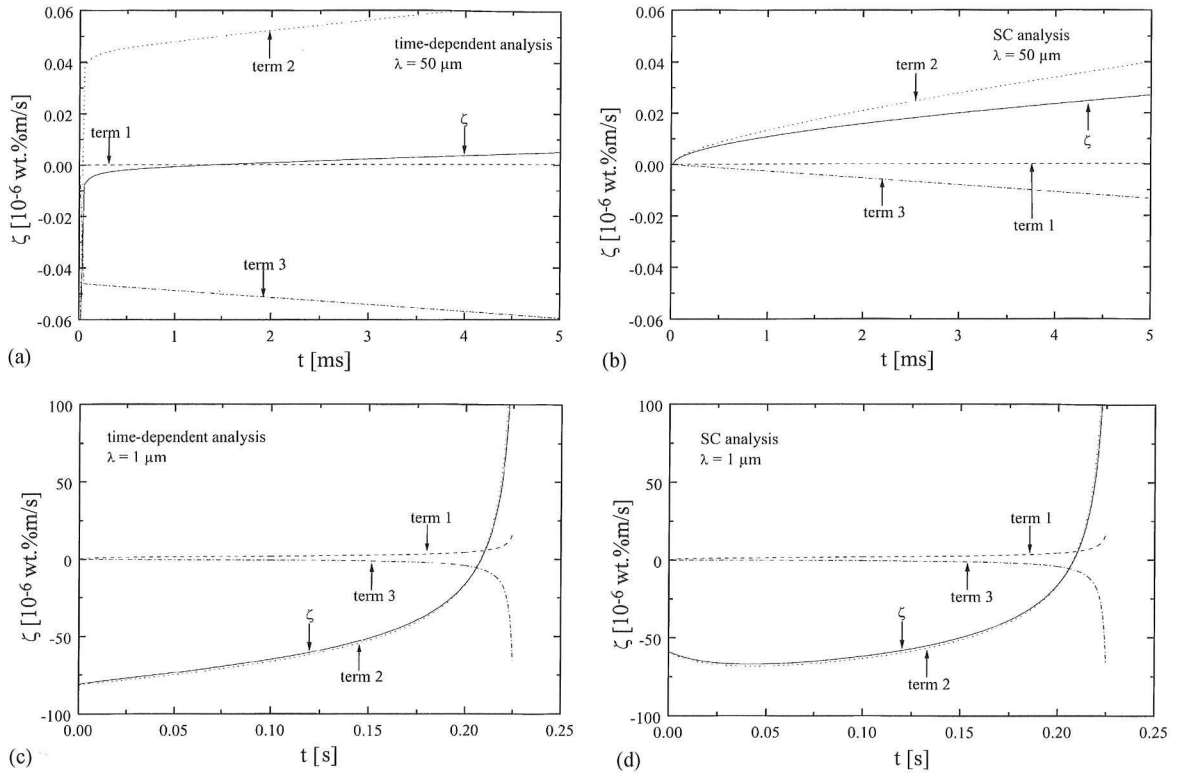


Fig. 3. The quantity  $\zeta$  from Eq. (6) as a function of time  $t$  applying the fully time-dependent analysis for two different wavelengths ((a) and (c)) and the corresponding curves for the SC analysis (Eq. (7)) calculated with the time-dependent values of  $V$  and  $G_C$  for the same wavelengths ((b) and (d)). Terms 1–3 refer to the different terms in Eqs. (6) and (7). For both wavelengths the corresponding MS analysis is almost equivalent to the SC analysis for term 2 and  $\zeta$  and identical for term 1 and 3. For  $\lambda = 50 \mu\text{m}$  the curves of term 2 and  $\zeta$  of the MS analysis lie slightly below the corresponding SC curves. The deviations increase with time. For  $\lambda = 1 \mu\text{m}$  the SC analysis reveals a negative slope of term 2 and  $\zeta$  at the beginning of the quenching process, whereas in the MS analysis the slope of term 2 and  $\zeta$  is always positive. In this case the deviations between both analyses vanish with increasing time.

with  $\zeta = \delta \dot{C}^0|_{z=0}(1 - k)$ . Since  $\zeta$  and  $\delta$  have the same sign  $\zeta \leq 0$  is equivalent to  $\delta \leq 0$ . Inserting the assumptions of Sekerka and Coriell [5, 6] the corresponding expression for the SC analysis is

$$\zeta_{\text{SC}} = -V \underset{\text{term 1}}{[C^1]_{z=0} + \delta G_C}(1 - k) - \left[ \underset{\text{term 2}}{-D\omega_C C^1|_{z=0}} - \underset{\text{term 3}}{V\delta G_C} \right], \quad (7)$$

with  $\omega_C = V/2D + \sqrt{(V/2D)^2 + \omega^2 + \sigma/D}$  and  $\sigma = \dot{\delta}/\delta$ . Like Mullins and Sekerka, they considered an exponential profile with boundary layer thickness  $D/V$  for  $C^0$  and a second exponential profile

with a time-dependent boundary layer thickness  $(1/\omega_C)$  for  $C^1$ .

In Fig. 3,  $\zeta$  is given as a function of time calculated with the fully time-dependent analysis and the SC-analysis for a short wavelength ( $\lambda = 1 \mu\text{m}$ , Fig. 3a and Fig. 3b) and a long wavelength ( $\lambda = 50 \mu\text{m}$ , Fig. 3c and Fig. 3d). To investigate the differences between both analyses, the three terms defining  $\zeta$  (see Eqs. (6) and (7)) are also plotted. A comparison of Eqs. (6) and (7) shows that term 1 is equal for both analyses. Thus, the difference between the fully time dependent and the SC analysis can be reduced to the different magnitudes of terms 2 and 3, which turn out to be the

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