Feedback control of unstable cellular solidification fronts

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(Dated: October 16, 2018)

We present a numerical and experimental study of feedback control of unstable cellular patterns in directional solidification (DS). The sample, a dilute binary alloy, solidifies in a 2D geometry under a control scheme which applies local heating close to the cell tips which protrude ahead of the other. For the experiments, we use a real-time image processing algorithm to track cell tips, coupled with a movable laser spot array device, to heat locally. We show, numerically and experimentally, that spacings well below the threshold for a period-doubling instability can be stabilized. As predicted by the numerical calculations, cellular arrays become stable, and the spacing becomes uniform through feedback control which is maintained with minimal heating.

PACS numbers: 64.70.Dv, 81.10.Aj, 81.30.Fb

Keywords: directional solidification, control, feedback

The control of cellular microstructures in directional solidification (DS) of dilute binary alloys is a subject of both industrial and fundamental interest [1, 2]. DS is produced in the presence of a thermal gradient, G, which moves at velocity \( V_p \). Cellular microstructures arise from the morphological instability of a planar front when the velocity of the thermal gradient is above some threshold, \( V_c \), that depends on the gradient \( G \) and the alloy concentration. Once the planar front becomes unstable, it reestablishes (ideally) into a periodic array of solid fingers or “cells”. A dynamic competition between solute diffusion, in the liquid, and capillary effects, at the moving solid-liquid interface, determines the typical cell size, but the local wavelength or cell spacing, \( \Lambda \), admits a wide range of stable values. When the average cell spacing, \( \Lambda_0 \), is above some spacing threshold, \( \Lambda_c \), the array is stable and achieves a steady configuration. When \( \Lambda_0 < \Lambda_c \), some cells, generally those of larger local wavelength, grow faster than their neighbors. This leads to the amplification of some modes and, eventually, to the elimination of, approximately, one cell out of two (period-doubling instability). During the initial evolution of the cell array, the elimination process is repeated, increasing progressively \( \Lambda_0 \), until a stable configuration with \( \Lambda_0 > \Lambda_c \) is reached.

In a previous experimental work, Lee and Losert [5] (see also [6]) have shown, using the so-called “combing method”, that it is possible to select a uniform cell spacing within the stable range. The combing method consists of using strong local temperature perturbations in the vicinity of the front during the initial transient evolution. The thermal perturbation sets the periodicity to the cell array. However, a permanent control of cellular patterns outside the stability domain had not yet been achieved. In this letter, we propose a new scheme to achieve this control and demonstrate its feasibility in both phase-field simulations and experiments. Our scheme works, essentially, by slowing down the growth of any cell which overgrows the average position of other cells in the direction of growth \( y \). For this purpose, we apply local heating close to the protruding cell tips. A key feature of this scheme is that the magnitude of feedback perturbations (i.e., the magnitude of heating) essentially vanishes in the controlled state.

We have performed numerical calculations using a modified version of a recently proposed quantitative phase-field model of binary alloy solidification [7, 8, 9, 10]. For the experiments, we use a model transparent alloy, namely, succinonitrile (SCN)-coumarin 152 (C152), in thin sample (see, e.g., [2] and refs. therein).

In the numerical calculations, we use a feedback control scheme which is a simple step function, i.e. the amount of heat injected in a spot-like region is constant regardless of how far the targeted cell grows beyond the average cell position \( \bar{y} \). We consider an array of \( N \) cells in a rigid box (with no-flux boundary conditions). For a given cell \( q \), local heating at the tip is applied when the distance from the average cell position, \( y_q - \bar{y} \), is larger than a pre-defined cut-off \( \delta > 0 \). The temperature field \( T(\vec{x}, t) \), neglecting the production of latent heat (“frozen temperature approximation”), can be expressed as follows:

\[
T(\vec{x}, t) = T_0 + G(y - V_p t) + p(\vec{x}, t),
\]
TABLE I: Parameters for impure succinonitrile used in phase-field simulations [1]. See also [12].

| Parameter                          | Value          |
|------------------------------------|----------------|
| $m|c_m$ (shift in melting temperature) | 2 K            |
| $D$ (diffusion coefficient)         | $10^{-9} \, m^2/s$ |
| $\Gamma$ (Gibbs-Thompson coefficient) | $6.48 \times 10^{-8} \, Km$ |
| $V_p$ (pulling speed)               | 32 $\mu m/s$   |
| $G$ (thermal gradient)              | 143.587 $K/cm$  |
| $d_0$ (capillary length)            | $1.3 \times 10^{-2} \, \mu m$ |
| $k$ (partition coefficient)         | 0.3            |
| $\epsilon_4$ (0.7 % anisotropy)    | 0.007          |

$p(\vec{x}, t) = \sum_q g \lambda_0 H[q_y(t) - \bar{y}(t) - \delta] \exp \left( \frac{-(\vec{x} - \vec{x}_q(t))^2}{\xi^2} \right)$

where $\vec{x}$ is the two-dimensional vector position, $p(\vec{x}, t)$ is the imposed thermal perturbation, $H[...]$ is the Heaviside step function, and the sum extends to the $N$ cells. These equations enter as a modification of Eqs. (132-133) in Ref. [10]. They approximate the fact that each heating spot in the experiments results in the build up of a Gaussian bump in the thermal field of constant width, $\xi$, and amplitude, $g \lambda_0$. A precise determination of the phase diagram for the SCN-C152 alloy used in this study is presently lacking. Therefore, for sake of realism, we carry out the phase-field simulations for physical parameters (Table I) estimated in Ref. [11] for a SCN-X alloy (where $X$ stems for an unknown impurity).

The feasibility of our control scheme is best illustrated by comparing the dynamic evolution of a strongly unstable cellular array with and without control. The wavelength of the array $\lambda_0 = 11.3 \, \mu m$ is well below the stability threshold $\Lambda_c \approx 50 \, \mu m$ for cell elimination [12]. Without control, the known spatial period-doubling instability that leads to cell elimination is observed. In contrast, with control on, this instability is suppressed. This is illustrated in Figure 1 where we show the evolution in time of the front position, $y_q$, at specific $x$ positions that initially correspond to cell tips, when control is on. It is observed that $\bar{y}$ decreases at the same time that $y_q$ dispersion decreases. Later, when $y_q$ dispersion is reduced, the average position advances arriving to a steady state value. In Figure 2 we show some snapshots of the front evolution. Due to the large initial value of $|y_q - \bar{y}|$ along the pattern, a relatively strong (or frequent) heating is necessary for melting protruding cells backwards (decreasing $\bar{y}$). This entails a transitory disorder visible in Fig. 2. However, this large-perturbation stage ceases as soon as cell tips reach an almost equal undercooling (Fig. 2c). Eventually, a uniform pattern is stabilized (Fig. 2d). We will see below that a uniform $\Lambda(x)$ distribution, obtained numerically by construction of the array, will also be obtained in the experiments. Interestingly, the heating frequency (not shown) decreases during the whole process, and reaches a very small value when control has been achieved. Note also that the system could not be controlled by setting $\delta = 0$ ($\delta/\Lambda = 0.5\%$ in Fig. 2). The reason for this is that it is practically impossible to maintain all tips at exactly the average fixed position. Therefore, our feedback algorithm with $\delta = 0$ heats continuously some of the tips and leads to a completely melted out of cells, destroying the periodicity of the array. Naturally, feedback control also fails for $\delta$ values larger than, say, 0.1 $\Lambda$. Note finally that a similar cut-off (about one pixel) was also introduced in our experimental feedback program.

In the experiments, we solidified a binary SCN-0.1wt% C152 alloy in a gradient $G = 10 \, Kcm^{-1}$, and with $V_p = 2 - 20 \, \mu ms^{-1}$ (same device as in Ref. 3). The alloy, which crystallizes in a $bcc$ cubic crystal, is confined in a 100 $\mu m$ thick, 2 mm wide, 150 mm long, glass-wall microtube. Prior to DS experiments, a single crystal with its [100] axis almost parallel, within less than 2°, to the solidification axis was selected by an empirical procedure and grown in such a way that it fills the container; it served as a permanent seed for further experiments. Experiments justify the use of a 0.7% anisotropy for the surface tension [13]. The local heating system is a holographic laser tweezer system (BioRyx200 from Arryx Inc) which consists of a 2-W NdYAG ($\lambda=532$ nm) laser that is focused onto a spatial light modulator. The single laser spot is then split into multiple diffraction spot pattern, which is projected into the imaged region through a (4x) objective of an inverted microscope. The system software allows independent positioning of hundreds of spots into controlled arrangements. Heating in the liquid is due to partial absorption of light by the fluorescent dye [14]. In a first approximation, a laser spot (about 10 $\mu m$ in diameter) acts as a point-like source of heat which diffuses rapidly in a quasi-bulk medium, including the thick glass walls. A Gaussian bump superimposed to the linear gradient (see Eq. 2g) is, thus, a realistic representation of the modified thermal field. From a rough calibration, we estimate that for short-exposure times (of order 1 s) and
low (about 0.2 \( W \)) laser power, the material is heated by less than 0.1 Kelvin in a region which extends over less than 40 \( \mu \)m. The numerical simulation heating power corresponds to \( g\Lambda_0 \approx 0.4 \) Kelvin and extends in \( \xi \approx 10 \) \( \mu \)m. There is no measurable overlapping between two neighboring laser spots, which are much smaller than \( \Lambda_0 \) (typically 100 \( \mu \)m).

We image the solidification front with a (1024x1280 pixel\(^2\)) digital camera. After some image processing, the front line is detected and smoothed with a moving central average of 5 pixels and the list of tips is detected. They are subsequently sent to laser controller after calculating the deviation from the average tip position. We use a feedback control program based on the numerical simulations to automatically place laser spots in the liquid region ahead of the protruding tips. By restricting the number of controlled spots to eight (\( N = 8 \)), we are able to update the laser spots at approximately one Hz.

A sequence of images during a typical feedback control experiment is shown in Figure 3. A simplified spatio-temporal diagram of the experiment is shown in Fig. 4. First, we perform a fast partial melting of the sample to obtain a planar solid-liquid interface at rest (not shown). Then, we start pulling at relatively high velocity (\( V_1 \approx 18 \mu m s^{-1} \)) which allows us to obtain a cellular array with small spacing by using the combing method (Fig. 3a). Then, we switched off the combing laser array, turned on control, and decreased the velocity stepwise down to a final value \( V_4 = 4.6 \mu m s^{-1} \) (\( \approx V_1/4 \)) at which the pattern is unstable. After a relatively short transient, during which the pattern is strongly perturbed due to frequent illumination (Fig. 3b), a well controlled, small-spacing pattern is eventually obtained (Fig. 3c). Note that the area accessible to the laser spot array is limited to about 1 mm, thus, only one half of the cellular pattern may be controlled. The other half was imaged but grew freely without control. As it can be seen in the spatio-temporal diagram of Fig. 4, many cells are eliminated outside the controlled window, while the number of controlled cells remains constant in the controlled area.

We then switched intentionally the laser light off (after about 10 minutes of successful control) during a few minutes, and observed the onset of the period-doubling instability (Fig. 3d). By turning feedback control on again, we were able to prevent cell elimination and to restabilize a small-spacing cell pattern similar to the initial one (Fig. 3e). We stress the striking resemblance between numerical (Fig. 2) and experimental (Figs. 3d to 3f) runs – including the transient stage after the laser is turned on (Fig. 2b and Fig. 3e). Finally, we switched feedback control off again and let the instability fully develop, and, as expected, approximately one cell out of two is eliminated in the previously controlled area (Figs. 3g and 3h). Let us make two important remarks.
In numerical as well as in experimental runs, the spacing distribution in a well controlled pattern is remarkably uniform (it is not so outside the controlled area) as seen, for example, in Figure 3. In addition, the controlled small-spacing cells sit slightly behind, thus have a larger undercooling than the large uncontrolled ones, as expected. This is not due to the added heat from the feedback control, but due to interactions between cells.

The second remark concerns the heating power applied to the cell pattern, or, equivalently, the laser spot exposure frequency \( f \) ahead of cells. Our main observation is that \( f \) gradually decreases as control goes on. In an experiment performed at \( V_p = 2.9 \, \mu m s^{-1} \) (not shown), we measured \( f \approx 0.5 \, s^{-1} \) at the beginning of the run, and, \( f \approx 0.05 \, s^{-1} \), 10 minutes later. As expected, the final characteristic time \( 1/f \approx 20 \, s \) is smaller than the characteristic amplification time of the period-doubling instability \( \tau \approx 80 \, s \) measured in situ in an uncontrolled, unstable pattern. Furthermore, we always performed feedback control sequences much longer (more than 10 minutes) than \( \tau \). It is interesting to note, also, that the final \( 1/f \) is comparable to the so-called diffusion time \( \tau_d = D/V^2 = 10 - 100 \, s \) (\( D \) falls in the \( 10^{-10} - 10^{-9} \, m^2 s^{-1} \) range), which signals a quasi steady regime. Therefore, control can be maintained as long as wanted.

In conclusion, we have shown experimentally and numerically that highly unstable cellular solidification arrays can be stabilized using feedback control. Experiment and simulation showed similar dynamical evolution as the array was controlled. Unlike feedback stabilization schemes for planar fronts slightly above \( V_c \), proposed by Savina et al., our approach stabilizes efficiently highly unstable states and can be implemented experimentally with only a discrete number of controllable heating points. An alternate control scheme which includes a tunable heating power for each spot has been implemented numerically; an experimental realization is currently in progress. Although a quantitative comparison between numerical and experimental results is not yet possible because of the lack of detailed knowledge of the alloy phase diagram, the observed qualitative behaviors are identical.

The use of other localized physical perturbations generated e.g. by x-rays or ultrasound could potentially make it possible to extend this control scheme to metallic alloys where the growth of much finer array structures with superior mechanical properties is of considerable practical interest. In addition, the ability to access experimentally unstable steady-state patterns provides an important new tool to enrich our fundamental understanding of pattern formation in directional solidification.

We thank B. Echebarria for helpful discussions. This research was supported by NASA grant NNM04AA15G and by Ministerio de Educación y Ciencia under the grant number EX2005-0085, and by a Research Corporation Research Innovation Award.

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