Dendritic and cellular doublets: Morphologies of thin solid films growing along a substrate during the initial state of solidification of bulk melts

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We present detailed experimental investigations of thin solid films growing along a borosilicate substrate for different dilute alloys in the succinonitrile-argon system. These thin solid films revealed a dendritic growth pattern with close-spaced sidebranches of different morphologies. We found surface dendrites with doublet tips as well as three different classes of sidebranches: cellular sidebranches, dendritic sidebranches, and an unsteady pattern, which we term ''cauliflower structure.'' Although the cellular sidebranches grew in a highly unstable manner, we found that cellular doublets build a stable substructure at low concentrations. Dendritic sidebranches with double tips divided by a straight groove in the plane of symmetry build a stable substructure at high concentrations. In the intermediate concentration regime cellular and dendritic doublets grew unstably with curved grooves forming irregular and a highly unsteady growth pattern. Proceeding from these thin solid films, the remaining bulk melt solidifies perpendicular to the substrate. For dilute alloys the appearance of surface dendrites can effect the final microstructure of the cast part, at least in the outer regions, whereas for higher concentrated alloys, where cells or even dendrites appear, no influence is expected.
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I. INTRODUCTION

During cooling of a liquid in contact with a solid material by heat extraction through the solid material, the solidification starts at the melt-substrate interface if the heterogeneous nucleation on the solid material appears prior to the nucleation within the melt. With the exception of highly inoculated melts, this is usually the case. As soon as the temperature of the liquid at the substrate interface decreases to a critical temperature range, hemispherical grains start to form on the solid surface. If elsewhere on the interface the critical temperature range is not yet reached, already existing grains may grow along the substrate into these regions before further nuclei are formed. Thus growth of thin solid films on cooled solid materials can precede the subsequent solidification of the bulk melt. Especially during continuous casting (e.g., melt spinning) the solidification of the whole strip is conceivable with nucleation just at the beginning of the process and no further nucleation during the steady-state production phase.

During experimental work on the solidification of transparent organic alloys within long, fine capillary tubes with a square cross section, we observed thin solid films growing along the tube walls. These thin solid films revealed a dendritic morphology with doublet tips. For the sidebranches of the, as we call them, surface dendrites, even cellular morphologies with doublet and multiple tips were found.

In the literature a few recent reports of dendritic or cellular solidification patterns with doublet tips are given. Koo, Ananth, and Gill [1] grew ice crystals in slightly undercooled pure water. With undercooling below a certain limit, they found repeated tip splitting when viewed from the basal plane, although the crystal grew with constant velocity into an isothermal melt. For a short time after splitting, both of the leading tips grew simultaneously. Then one of these tips became dominant, while the other was retarded as a sidebranch. In contrast, the edge plane of the tip never splits.

Crystalline anisotropy arises from the orientation dependence of the surface energy and of the interfacial attachment kinetics that act simultaneously to encourage the growth of a dendrite along its main stem. The microscopic solvability theory [2–6] indicates that dendrites cannot have stable growth without anisotropy. Kessler, Koplik, and Levine [7] argued that minute temperature fluctuation in the melt, known as noise (also important for the formation of sidebranches), can induce tip splitting in materials that have a degree of anisotropy less than a certain critical value. Koo, Ananth, and Gill [1] estimated the degree of anisotropy for ice to yield $\epsilon_2 = 0.3$ for the edge plane and $\epsilon_6 \approx 0.002$ for the basal plane. However, they also pointed out that natural convection had to be considered in their tip-splitting experiments.

Jamgotchian, Trivedi, and Billia [8] performed thin film directional solidification experiment on the succinonitrile (SCN) -acetone system. They found a branch of cellular structure in which the interface pattern consists of a periodic array of coupled cells, called cellular doublets. Their dynamical studies confirmed the selection of the doublet interface as a stable solution of the cellular pattern formation beyond the threshold of planar interface stability at low solidification velocity.

Brener *et al.* analyzed the growth of two-dimensional crystals in a channel with and without anisotropy by a Green's function method [9]. The authors observed stable steady-state growth of nonsymmetrical fingers with doublet tips as the zero surface tension solution if the channel width exceeds some critical value. This observation was confirmed by Ben Amar and, Brener [10] who found parity-broken

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double fingers to appear with fully isotropic surface tension even in the range of small undercooling.

Dendrites with a doublet tip are recently predicted by Ihle and Müller-Krumbhaar [11]. They presented with a two-dimensional shape interface model, numerical studies, on the dependence of growth morphology on undercooling and surface tension anisotropy for the solidification of a pure melt. The basic predictions for the growth form compact and fractal dendrites for anisotropic surface tension and compact and fractal seaweed for vanishing anisotropy are confirmed. Within a range of medium anisotropy of the surface tension (and large undercooling) they found two stabile solutions dependent on the initial condition: fractal dendrites and dendrites with doublet tips, which they call a symmetry-broken (SB) double finger. If the anisotropy reaches a critical value, the dendrite with doublet tip becomes unstable.

To study the nature of the growth pattern in systems with a vanishing anisotropy, Akamastu, Faivre, and Ihle [12] investigated the morphologies within thin samples of CBr_4-C_2Cl_6 directionally solidified. If this system is forced to be two dimensional with the {111} crystalline plane parallel to the plane of the thin film, then the anisotropy of surface tension vanishes. Under these conditions they found a non-dendritic and unsteady growth pattern, similar to the theoretically predicted seaweed pattern. The building blocks of this pattern are pairs of SB fingers called doublons, whose lifetime is long but finite. With a ⟨100⟩ axis close to the pulling direction they found stable ''dendritic'' doublons or double dendrites above a critical growth velocity, which coexist with dendrites. These dendritic doublons are determined by the coupled growth of two ''half'' dendrites separated by an inner groove, more than 20 times smaller than the total width of a single doublon. Their tips retain the triangular shape of the symmetric dendrites.

In this paper we show that patterns occurring during growth of a thin solid film on a cold substrate can reveal dendritic or cellular doublet structure as well as a dynamically growth structure, which we term ''cauliflower structure.'' After describing the experimental procedure in Sec. II, details on experimental observations of so-called surface dendrites are given in Sec. III A, on the concentration dependence of the sidearm structures in Sec. III B, and on the influence of surface structures on bulk solidification in Sec. III C. Final conclusions are drawn in Sec. IV.

II. EXPERIMENTAL PROCEDURE

Borosilicate glass capillaries, 900 mm in length, with a square cross section, an inner opening of 200 μm, and a wall thickness of 100 μm were filled with different SCN-argon alloys. The SCN had to be distilled under vacuum and zone refined with more than 50 passes to achieve the desired purity. Maintaining thermal convection within the molten SCN, different argon-gas pressures onto the melt surface were applied to perform the alloying. After filling a capillary, the concentration of the alloy inside the tube was estimated by measuring the solid-liquid temperature interval ΔT_0 and referring to the SCN-Ar phase diagram from Chopra [13]. The measured temperature interval for the alloys were $\Delta T_0 = 235 \pm 40$ mK ($C_0 = 0.013$ wt. %), $\Delta T_0 = 450 \pm 50$ mK ($C_0 = 0.024$ wt. %), $\Delta T_0 = 570 \pm 60$ mK ($C_0 = 0.031$ wt. %), and $\Delta T_0 = 970 \pm 70$ mK ($C_0 = 0.052$ wt. %), respectively. (Due to the phase diagram of Chopra [13], a constant distribution coefficient of $k = 0.2$ and a constant liquidus slope of $m = -4.7$ wt. %/K was assumed to estimate C_0.) These temperature intervals were measured at different locations within the tubes. Details concerning the purification of the SCN are given in [14] and concerning the alloying, the filling procedure and the measurement of ΔT_0 are given in [15].

A segment of the capillary, 80 mm in length, was surrounded with an isothermal environment realized by placing the segment in a water jacket of about $80 \times 80 \times 28$ mm^3. The height of the water jacket was restricted to 28 mm because it had to fit under an optical microscope to observe the solidification. The temperature in the center of the jacket was measured with a quartz thermometer. The detector of this thermometer was cylindrical in shape, about 15 mm in length and 10 mm in diameter. The capillary tube was aligned parallel to the quartz thermometer with a spacing between them of 15 mm. The thermostat used to heat the water flowing though the jacket, as well as the thermometer, had a relative accuracy of ± 5 mK. The temperature within the jacket was found to vary by about ± 15 mK. Thus the relative accuracy of the whole system was estimated to be about ± 20 mK.

Starting with a molten alloy at constant temperature within the segment of the capillary, the heating of the thermostat was switched off. The temperature of the circulating water decreased with a cooling rate of $\dot{T} = 0.044 \pm 0.0035$ K/s. As soon as the temperature of the isothermal environment decreased below the liquidus temperature, the solid phase started to grow from outside the segment into the melt. Without moving the specimen, the solidification in the central part of the heated capillary segment was observed with an optical microscope to which a video recorder was attached. For the observation of the solidification within an edge of the square cross section capillary tube, they were turned by 16° with respect to the optical axis.

III. RESULTS AND DISCUSSION

A. Thin solid film of dendritic morphology with doublet tip

Figure 1 shows the growth morphology for the alloy with $\Delta T_0 = 970$ mK passing the central part of the segment. The growth direction was from right to left. The rapidly growing solid film was of dendritic form and is therefore referred to in this paper as a surface dendrite. The images were recorded in $\Delta t = 20$ ms time steps. The growth velocity of the tip was $V = 3.7$ mm/s. The actual temperature of the surrounding water, given in the inset, was $\Delta T = 980 \pm 50$ mK below liquidus ($T_L = 331.00$ K). Figure 2 shows secondary and tertiary branches of the same alloys on two successive images ($\Delta t = 20$ ms). Here the growth direction was opposite that of Fig. 1. The tip of the surface dendrite shown in Fig. 2 grew outside the observation window.

Due to the slow cooling rate, the temperature within the segment of the capillary located close to the quartz thermometer and the temperature of this detector were assumed to be equivalent. With the constant cooling rate of $\dot{T} = 0.044$ K/s, the temperature reduction from the liquidus to the temperature shown in the insets of Figs. 1(a)–1(d) took about Δt

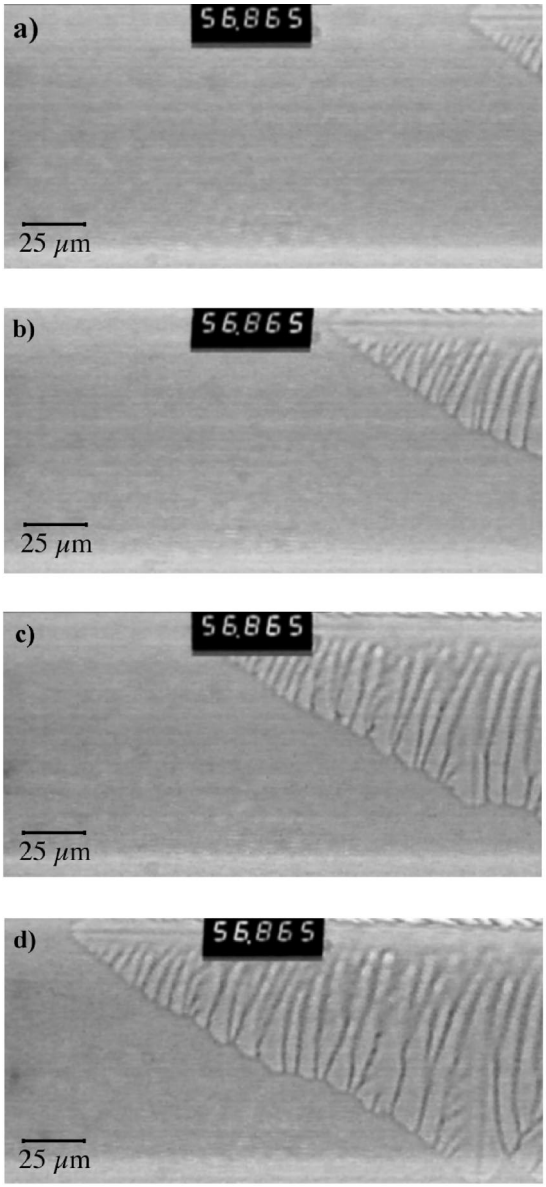

FIG. 1. Propagation of a surface dendrite in the alloy with $\Delta T_0 = 970$ mK. The tip of the surface dendrite grew beside an edge of the square cross section tube from right to left parallel to the tube axis. Note that the surface dendrite revealed a doublet tip. The growth direction of the sidebranches with respect to the growth direction of the surface dendrite changes from approximately 55° at the tip to an angle between 80° and 90° farther away from the tip. The interfaces between two neighboring sidebranches can be curved. Dendritic sidebranches also revealed doublet tips. The images were recorded in $\Delta t = 20$ ms time steps.

$= 22.3$ s. Assuming that the growth velocity increased from zero at the boundary to $V = 3.7$ mm/s at the central part of the molten segment (half length of 40 mm) linearly with time, the solid should have reached the central part after $\Delta t = 21.6$ s. Considering a short time interval for the solid-liquid interface, initially at rest, to start moving, this result confirms the assumption of constant acceleration. The acceleration was estimated to be $a = 0.165$ mm/s^2. From these values, an increase in growth velocity of 0.3% from Figs. 1(a) to 1(d) was calculated. Therefore, the morphology of the surface dendrites can be regarded as a quasi-steady-state.

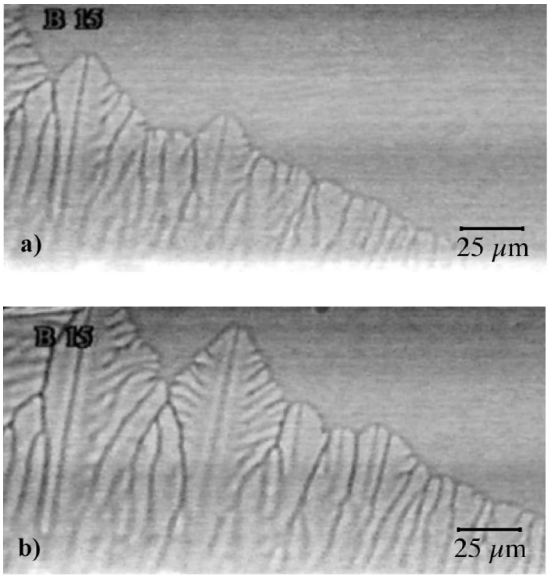

FIG. 2. Dendritic doublets growing as sidebranches shown for another experiment of the same alloy as in Fig. 1. Here the tip of the surface dendrite grew below the observation window from left to right ($\Delta t = 20$ ms). As for the main stem, the groove of the dendritic doublets is straight.

This result is also valid for other morphologies presented in this paper.

The following features concerning surface dendrites are found independent of concentration (within the investigated range). (i) The surface dendrites seem to grow parallel to the tube axis or at least with an inclination that could not be seen on the scale of the observation window. (ii) They are isolated objects, which were never observed to grow within a periodic array, as in common directional solidification experiments. In the majority of experiments only one surface dendrite was present. Only twice two surface dendrites growing on opposite tubes walls in the same direction were found. (iii) The surface dendrites grew always near or on the edges of the square cross section capillary tubes. Since the edges revealed a small radius of a few microns, it was, due to the optical effect, not possible to look right into it. Thus tips of surface dendrites growing in the edge were not clearly visible. Only for the alloy with $\Delta T_0 = 970$ mK did we find tips growing nearby but not in an edge (Fig. 1). (iv) The sidebranches grew in close contact with each other. They can be classified into three classes: *A*, cellular sidebranches; *B*, dendritic sidebranches; and *C*, an unsteady pattern that grew in a highly unstable manner and can adequately be described by the term cauliflower structure. Depending on the spacing, this structure has a certain similarity to the seaweed pattern presented by Akamastu, Faivre, and Ihle [12].

Especially for the alloy with $\Delta T_0 = 970$ mK the following observations were made. (i) The surface dendrites revealed a *doublet tip* with a thin groove between two ''half'' dendrites, which was about 100 μm in length [Fig. 1(b)]. (ii) The angle between the growth direction of the stem and the secondary arm front at the tip region is about 35°. This is caused by a smaller growth velocity of the secondary branches compared to the primary trunk of about 80%. (iii) The growth direction of the sidebranches with respect to the growth direction of the surface dendrite changes from approximately 55° at the

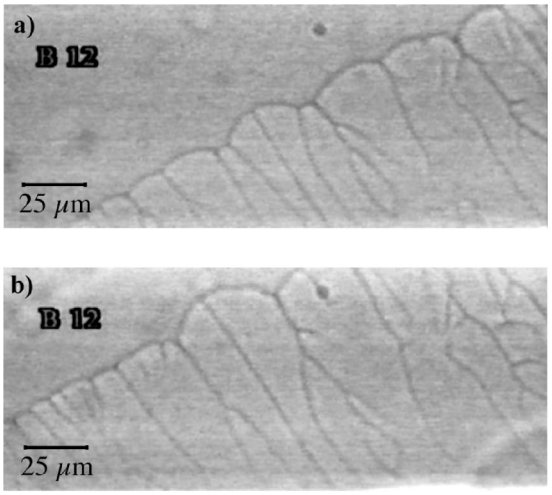

FIG. 3. Secondary branches of a surface dendrite for the alloy with $\Delta T_0 = 235$ mK. The tip of the surface dendrite grew below the observation window from right to left ($\Delta t = 20$ ms). The cellular area contained cellular doublets with straight grooves in the plane of symmetry that form a stable substructure within an unstable cellular pattern.

tip region (growth perpendicular to the secondary arm front) to an angle between 80° and 90° further away from the tip. The interfaces between two neighboring sidebranches can be curved [see Fig. 1(d)]. (iv) The dendritic sidebranches always revealed doublet tips. The probability of dendritic sidebranches increases the greater the distance from the tip region of the main stem. (v) The tertiary branches formed an angle of some 40° between the growth direction of the secondary branch and the tertiary arm front.

The fact that the growth direction of the surface dendrites is determined by the heat flow rather than the crystalline structure of the growing solid shows that the growth mechanism of dendrites and surface dendrites are different. This is confirmed by the value of the angle between the main stem and sidebranches at the tip and its change farther away from the tip (curved sidebranches). As pointed out in the Introduction, the appearance of stable doublet tips is accomplished with a low anisotropy of surface tension. Thus we suggest that the growth of a surface dendrite can be described by means of an ''effective'' surface tension, which takes into account that the substrate/solid/liquid three junction point reduces the effect of the anisotropy of the solid/liquid surface tension.

B. Concentration dependence of thin solid film morphologies

As mentioned above, the tip of a surface dendrite could only be observed directly for the alloy with $\Delta T_0 = 970$ mK. Here it revealed a doublet tip. For the other alloys, which had a lower concentration, the tip region was not visible and therefore it could not be decided whether or not the corresponding surface dendrites had a doublet tip.

The secondary branches for the alloys with $\Delta T_0 = 235$, 450, and 570 mK are shown in Figs. 3–5 in two successive images (a) and (b) for each alloy. Here the growth directions were from right to left. In Figs. 3 and 4 the main stem of the surface dendrites is below the observation window and in Fig. 5 above. The images were again recorded in $\Delta t = 20$ ms time steps.

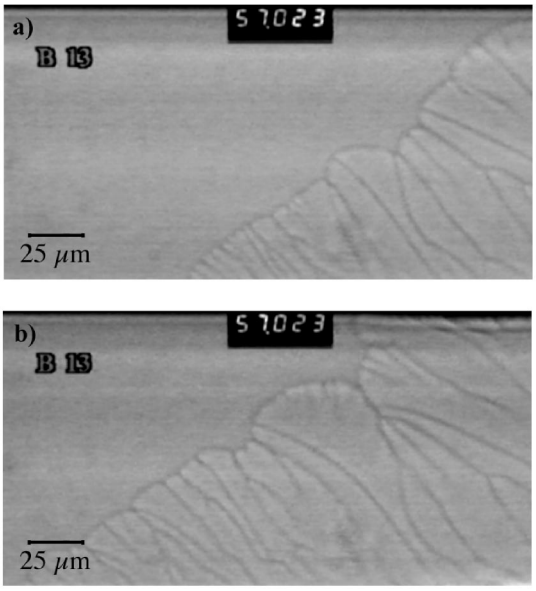

FIG. 4. Secondary branches of a surface dendrite for the alloy with $\Delta T_0 = 450$ mK. The tip of the surface dendrite grew below the observation window from right to left ($\Delta t = 20$ ms). Here the cellular doublets are no longer a stable substructure. They began to bend, resulting in an asymmetrical shape with a dominant and retarded part separated by a curved groove. From the dominate part a cauliflower structure was formed.

Comparing the morphology of sidebranches for the different alloys (Figs. 2–5), the following observations were made. (i) The spacing of the sidebranches increased with decreasing concentration. (ii) Sidebranches of alloys with $\Delta T_0 \leqslant 570$ mK grew in a highly unstable manner: Cell overgrowth appeared, as well as multiple tip splitting. (iii) For

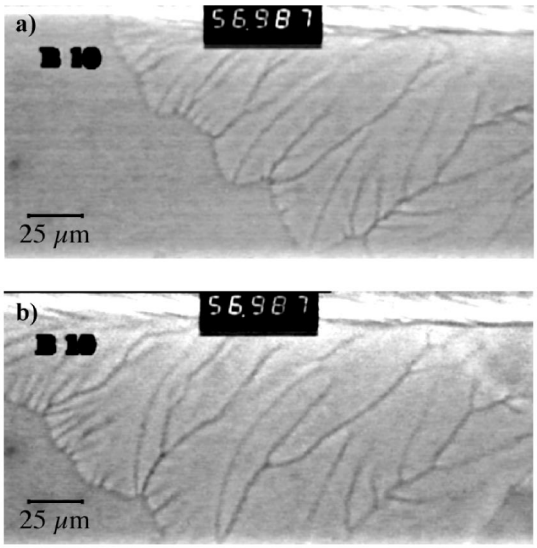

FIG. 5. Sidebranches of the surface dendrite for the alloy with $\Delta T_0 = 570$ mK revealed curved dendritic doublets that may continuously transform into a cauliflower structure. Here the tip of the surface dendrite grew above the observation window from right to left ($\Delta t = 20$ ms).

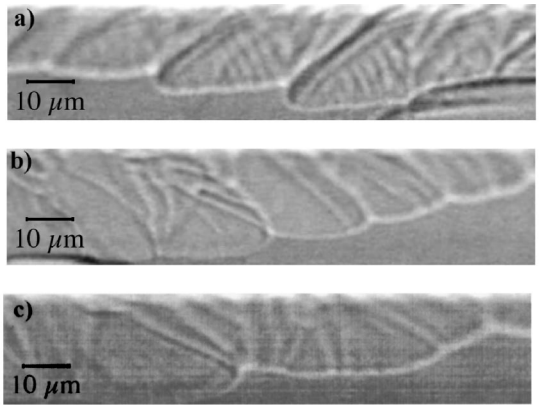

FIG. 6. Three-dimensional view of the three classes of sidebranches: (a) dendritic doublets (growing from the rear right towards the left fore), (b) cellular doublets (growing from left towards the right fore), and (c) an unsteady cauliflower structure [growing like (b)].

the alloy with $\Delta T_0 = 235$ mK only sidebranches of type *A*, namely, as deep cells, were found (Fig. 3). They revealed doublet tips, similar to the cellular doublets observed by Jamgotchian, Trivedi, and Billia [8]. Although the overall growth of the cellular pattern is not morphologically stable, the cellular doublets seems to build a stable substructure, with a straight groove in the plane of symmetry. (iv) With higher concentration the stability of the cellular pattern decreased further and sidebranches of type *C* appeared. Cellular doublets are no longer a stable substructure. They began to bend, resulting in an unsymmetrical shape with a dominant and retarded part separated by a curved groove. From the dominate part a cauliflower structure was formed as shown in Fig. 4 for the alloy with $\Delta T_0 = 450$ mK. (v) With a further increase in solute content ($\Delta T_0 = 570$ mK), the curved cellular doublets developed into curved dendritic doublets, which may be continuously transformed into a cauliflower structure as shown in Fig. 5. Due to the high dynamics of this sidebranch pattern, the cauliflower structure may also change back into curved dendritic doublets. (vi) The direction of the curved grooves within a cellular or a dendritic structure can be towards or away from the tip region of the main stem. (vii) As already mentioned in Sec. III A, the sidebranches of the alloy with $\Delta T_0 = 970$ mK contained dendritic doublets (Fig. 2). In contrast to the dendritic doublets of the alloy with $\Delta T_0 = 570$ mK (Fig. 5), they build a stable substructure with a straight groove in the plane of symmetry similar to the cellular doublets of the lowest concentrated alloy shown in Fig. 3.

Figure 6 shows a three-dimensional view of the three classes of sidebranches. Two parallel dendritic doublets are seen in Fig. 6(a), a cellular doublet growing between unsteady structures in Fig. 6(b), and an example of an unsteadily growing cauliflower structure in Fig. 6(c).

The transition from a dendritic to a cellular morphology by reducing the solute content is well known in solidification. Although not fully understood, this transition appears when the sidebranches of the dendrites vanish. Our observations show that with growing surface structures a similar transition occurs. In contrast to the common dendrite-to-cell transition, the transition of surface structures is accompanied by the appearance of unstable transition structures (curved dendritic doublets/cauliflower/curved cellular doublets). The transition starts with unstable bending of the surface dendrite rather than the disappearance of sidebranches. Thus we believe that the mechanism for the dendrite-to-cell transition in the case of surface structure is different from the mechanism that is active in common directional solidification experiments.

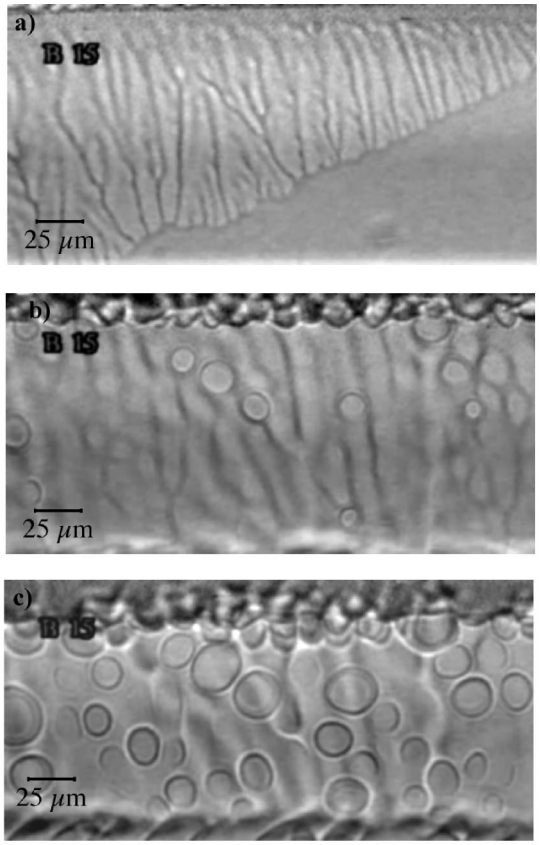

FIG. 7. Sequence of images taken after a surface dendrite of the alloy with $\Delta T_0 = 970$ mK had passed the observation window. The images were recorded in $\Delta t = 300$ ms time steps. Cells with a growth direction perpendicular to the tube walls started to grow from the sidebranches of the surface dendrite. The growth velocity perpendicular to the sidewall of the capillary turned out to be two orders of magnitude smaller than the growth velocity of the surface dendrite.

C. Influence of surface structures on bulk solidification

To see how the appearance of surface dendrites influences the solidification of the bulk melt, the further evolution of the surface dendrites was investigated. Figure 7 shows a sequence of images taken after a surface dendrite of the alloy with $\Delta T_0 = 970$ mK had passed the observation window. The images were recorded in $\Delta t = 300$ ms time steps. Figure 7(a) shows the sidebranches just behind the tip region. After having established a thin layer of solid on the tube wall, further solidification of the surface dendrite took place perpendicular to the sidewalls. For the alloy with $\Delta T_0 = 970$ mK the solidification perpendicular to the tube walls into the bulk melt proceeds by forming cellular structures (Fig. 7). From the side view of this cellular growth the velocity of the growth

perpendicular to the capillary was estimated to be of the order of $V_\perp \approx 50\ \mu$m/s. Thus $V_\perp$ is two orders of magnitude smaller than the growth rate of the tip or the sidebranches.

Under similar experimental conditions no cellular growth perpendicular to the tubes walls was found for the low concentrated alloy with $\Delta T_0 = 235$ mK. Here the fine and irregular spacing of the surface dendrite changed with further growth to form a configuration that appeared to be very similar to a polycrystalline structure. (Due to a very poor contrast of the images showing the further development of the thin solid layer for the alloy with $\Delta T_0 = 235$ mK, these images are not shown.) This has also been found by the authors under different experimental conditions [15]. The growth velocity perpendicular to the tube wall was found to be $V_\perp \approx 30\ \mu$m/s and is thus also for an alloy with $\Delta T_0 = 235$ mK two orders of magnitude smaller than the growth rate of the tip.

A surface dendrite can be regarded as a solid substrate from which an epitaxial growth into the bulk melt appears. For the alloy with $\Delta T_0 = 970$ mK the the solid layer formed by the surface dendrite turned out to be unstable with respect to the new growth direction and the reduced velocity; therefore the interface started to adapt to the new growth conditions by forming cells. The cellular growth in the bulk melt appeared to be independent of the initial shape of the surface dendrite. Because of the lack of cellular growth perpendicular to the tube walls for the lower concentrated alloy, it has to be assumed that $V_\perp$ is below the limit of constitutional undercooling for this alloy. Thus the fine undulations of the surface dendrite had to change into a planar front. In our experimental time scale this transition was not fully obtained. Although the interface changed from a fine scaled pattern (the surface dendrite with its sidebranches) to a morphology with coarser structures, the interface still revealed unevenness arising from the history of solidification.

IV. CONCLUSIONS

Thin solid films growing on a cold substrate reveal growth characteristics that are very different from the well known cellular and dendritic pattern studied in directional solidification or free growth experiments. It was observed that surface dendrites can show doublet tips. Their sidebranches are very closely packed and reveal, depending on the concentration present, either cellular or dendritic doublets or unstable transition structures. The observed growth behavior of these surface structures induces us to assume that the substrate/solid/liquid three junction point reduces the effect of the anisotropy of solid/liquid surface tension leading to a less anisotropic ''effective'' surface tension.

Due to the fact that the dendrite-to-cell transition in the case of surface structures is accompanied by the appearance of unstable transition structures (curved dendritic doublets/cauliflower/curved cellular doublets), we believe that the mechanism for the dendrite-to-cell transition in the case of surface structure is different from the mechanism that is active in common directional solidification experiments.

According to our observations, we conclude that the appearance of surface dendrites should not affect the microstructure of the final casting. The destabilization of the thin solid film covering the substrate is so fast that adaptation to the new growth condition (growth perpendicular to the substrate) appears instantaneously. However, for dilute alloys where the growth conditions favor a planar front growth perpendicular to the substrate, the system starts to coarsen during further growth and the undulations of the fine structured surface dendrite lead to boundaries that persist for a long time. Thus we believe that in this case the final microstructure will be affected by the presence of the surface dendrite in the outer regions.

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