# Three-dimensional growth morphologies in diffusion-controlled channel growth

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The growth of a supercritical nucleus from a diffusion field is studied numerically using a phase-field model. This represents growth of a crystal with small or vanishing anisotropy. We find three-dimensional cooperating multiplets instead of dendrites, in analogy to the doublons found previously in two dimensions. Triplet structures in particular appear to be the building blocks of the resulting morphology under conditions of free growth in three dimensions. [S1063-651X(97)02506-3]

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Growth phenomena often occur in conjunction with a first order phase transition. There a critical nucleus is formed by some thermodynamical fluctuation. This nucleus afterwards grows in a deterministic way to an in principle arbitrarily large size. The growth of a crystal from the melt or from a solution is a typical example of such a process [1,2].

This type of phase change usually requires the transport of at least one conserved quantity, the solute material or the latent heat of solidification, which is transported via diffusion. While in two-dimensional geometry substantial progress has been made during recent years, it is still rather unclear what happens in three dimensions, where the interface between the growing solid and the nourishing fluid is an at least two-dimensional object. Particular interest is devoted to the behavior on a long time scale, where one can expect universal features to characterize the system dynamics.

It has been known for about three decades [3] that such a growing nucleus becomes unstable as its radius becomes larger than a few times the critical radius. If the surface tension is anisotropic, for example due to crystalline anisotropy, it is generally accepted that the nucleus finally deforms into a dendritic pattern like a snow flake ([1,2]). The limit of vanishing anisotropy, however, is much less clear.

There has been a recent attempt to formulate a theory [4,5] for the fundamental morphologies and the most relevant parameters controlling their appearance. This was based on scaling relations together with asymptotic matching requirements. The resulting morphology diagram and its recent modification [5] uses *supercooling vs crystalline anisotropy* as the principal axes, and discriminates between seaweed and dendrites as the basic patterns, where the dendritic patterns are characteristic for anisotropic growth conditions.

Some basic predictions of this theory have been recently confirmed in the two-dimensional case. For typical experimental situations [6,7] the compact dendritic growth morphology then is the most likely one to occur, and the theory for the growth of single dendrites [1,8,9] appears to describe the situation quite well. A summary can be found in [10].

TABLE I. Parameters and results of the simulations. $LZ$ is the cell-length in the growth direction (discretized in 200 grid-points), $LX$ and $LY$ the lateral size, Ltot is the total length grown after the system has settled to essentially steady behavior, and $\ell_D = 2D/v$ is the diffusion length observed as measure for the inverse growth rate. The other model parameters were $\tau=1$, $\xi=1.5$, $V_0=1$, $\mu_0=9$, $D=1$, and $\Delta=0.8$, all lengths in arbitrary units.

| Fig. No. | $LX$ | $LY$ | $LZ$ | Ltot | $\ell_D$ |
|---|---|---|---|---|---|
| 1 | 7.5 | 50 | 100 | 7000 | 1.70 |
| 2 | 15 | 100 | 100 | 800 | 1.57 |
| 3 | 30 | 30 | 100 | 2200 | 1.61 |
| 4, 5 | 60 | 60 | 100 | 8600 | 1.62 |

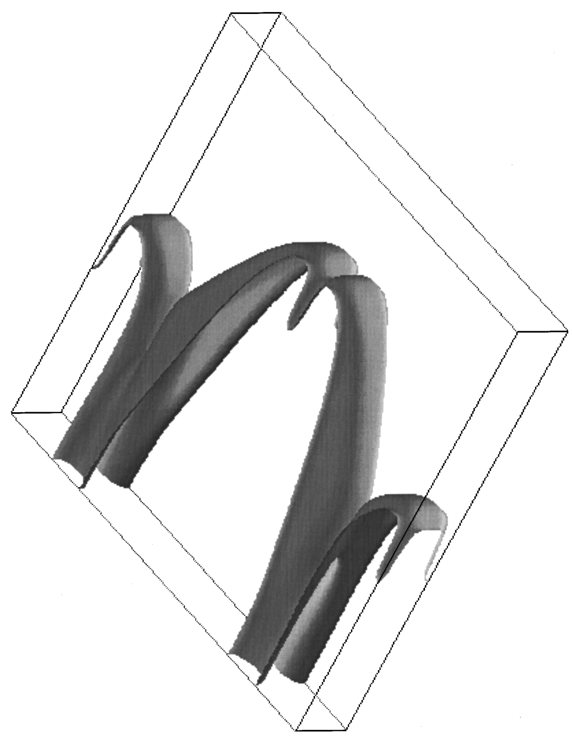

FIG. 1. Symmetry-broken finger growing in a flat cell with reflecting walls. For convenience, only the surface of the growing object is shown in all figures. The small satellites seen on the sides are dying away very slowly during growth. When the cell thickness is further reduced, a two-dimensional doublon is finally obtained. Note, however, that only one-fourth of this cell was computed; the whole cell shown here was reconstructed by mirror symmetry. See Table I for parameters used in the computation of all figures.

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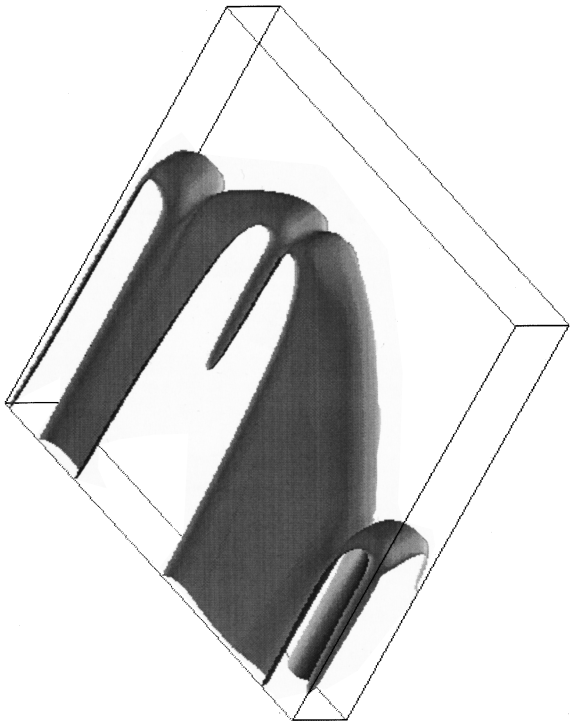

FIG. 2. Three-dimensional doublon. Same configuration as in Fig. 1, but here the whole cell was used in the computation, no twofold or fourfold mirror symmetry was exploited. The sidewalls of the cell again have mirror conditions. Just like the doublons obtained in two dimensions (Refs. [11] and [12]) the three-dimensional doublon here consists of two symmetry-broken fingers which appear to stabilize each other during growth. This pattern resembles experimental results (Refs. [28] and [29]) for directional solidification.

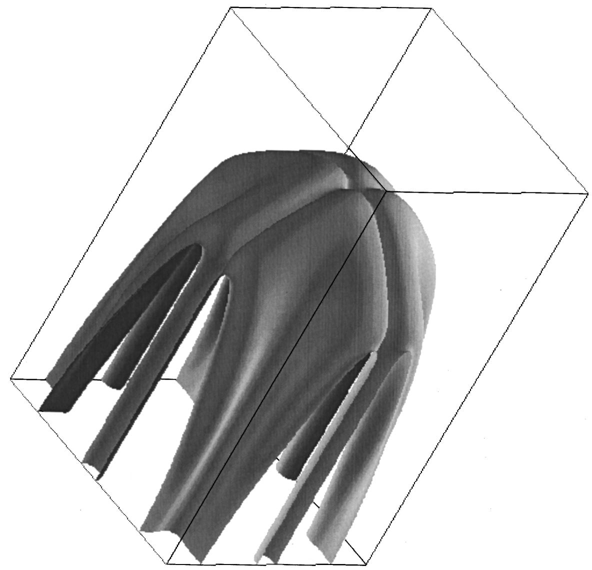

FIG. 3. The channel has now been made quadratic in cross section, in contrast to the rather flat channels shown in Figs. 1 and 2. A quadruplet of symmetry-broken fingers is growing steadily in the center. This fourfold symmetry in the center is imposed by mirror symmetry, so that we see four identical images around the center. This figure shows that asymmetrical fingers may be the basic ingredients of the growth structure, but does not yet allow conclusions about their cooperation (see following figures).

The limit of vanishing crystalline anisotropy has been recently investigated in detail [11,12], and leads to a different growth pattern. This seaweedlike pattern consists of substructures which are called *doublons*. They resemble dendritic patterns, but consist of cooperating symmetry-broken double fingers [11–13].

In principle, a morphology diagram as in Ref. [5] could also have been proposed for the threedimensional case, but on a much weaker basis. What seems to have been settled quite recently is the growth of single dendrites in three dimensions. After a description of the three-dimensional dendrite tip [14], an analytical solution for the tail-region was also obtained [15]. This was further elaborated to account for sidebranches under the influence of noise [16]. The theory gives detailed scaling relations for the various geometrical aspects of the three-dimensional dendrite structure, which is quite far from the almost parabolic shape obtained in the two-dimensional case. Recent experiments [7] gave striking quantitative agreement with some crucial predictions of that theory. Thus we may assume that the basic mechanism for the formation and growth of three-dimensional dendrites is understood.

It is still an open problem what happens in the limit of very small or vanishing anisotropy. One may expect that patterns similar to the *doublons* observed in two dimensions [11] may also exist in three dimensions, in some analogy with the dendrites, but no details are known so far. It is the purpose of this paper to shed some light on this region in parameter space where one must expect growth patterns which are quite different from the dendritic patterns observed for larger crystalline anisotropies.

We used a *phase-field model* [13,17–24] to simulate crystal growth under control of a diffusion field in a channel over long times. The standard version of a phase-field model describes the time evolution of an order-parameter field $\phi$ which is not conserved, but which is coupled to at least one second field $u$ which obeys a continuity equation. This field $u$ describes a conserved variable like energy or a chemical component. The equations of motion can be derived from a functional for the appropriate thermodynamic potential of the system. They can be written as a set of coupled partial differential equations in time and space [13,17–25]:

$$\tau\frac{\partial}{\partial t}\phi(x,t)=\xi^2\nabla^2\phi-V_0(\phi^3-\phi)+\mu_0\frac{d\Gamma}{d\phi}(u+\Gamma), \tag{1}$$

$$\frac{\partial}{\partial t}u(x,t)=D\nabla^2u-\frac{\partial}{\partial t}\Gamma$$

where $\Gamma(\phi)=\phi/(1+\phi^2)$ is a function which switches between $\pm 1/2$ as $\phi$ varies between $-1$ and 1 from solid to liquid. The solid-liquid coexistence corresponds to $u=0$, and we supercool the distant liquid by $u=-\Delta$. Furthermore, $\tau$ is a time scale, $\xi$ a correlation length, and $V_0$ characterizes a potential barrier between the pure states $\phi=\pm 1$. The model is rather similar to the one used by Kobayashi [24], where $\phi=\pm 1$ in the pure solid or liquid phase, also under nonequilibrium conditions. Details are not important here, and will be discussed in a different context [25].

The model was tested quantitatively in detail in one, two and three dimensions for symmetrical geometries, and also in

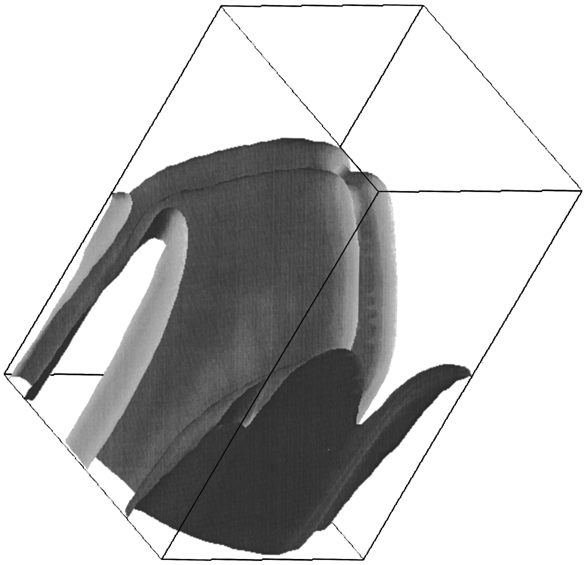

FIG. 4. This is one of the central results. In contrast to the previous figures, this computation was done using periodic boundary conditions at the channel sidewalls. The crucial point here is that this *self-organized triplet structure* is not imposed by symmetry, and that it consists of three *cooperating symmetry-broken fingertips*. The pattern has grown over 100 times the width of the cell and over 1000 times a typical tip radius of the fingers without any appreciable change. The initial condition was a flat interface with a central bump. The initial condition disappeared during a rather chaotic looking dynamically evolving pattern, until it reached this essentially steadily growing pattern.

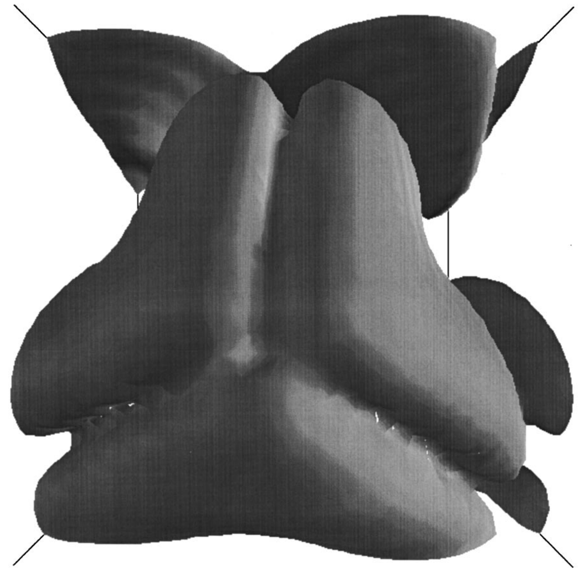

FIG. 5. Same triplet as shown in Fig. 4, but now seen from the top. Note that the apparent axis of the triplet does not exactly coincide with the orientation of the computational grid, which is clear proof of the self-organizing abilities of these patterns. This picture can be continued in all four directions by periodic continuation.

comparison with results on dendritic growth in two dimensions obtained previously by a sharp-interface calculation [11,26]. In particular, we have shown that the numerical anisotropy due to the discretization is not significant. For the present purpose this will be sufficient, since all our basic results can be presented in scaled form. These results are given in the figures, and discussed in the corresponding captions for convenience. All simulations were performed at the same physical parameters but for different cell geometries; see Table I.

Results for growth in a channel with reflecting walls are shown in Figs. 1–3, and are discussed in the captions. The most prominent feature is that multiplet structures of the interface are found, while the environmental conditions apart from the channel geometry are isotropic, so that dendritic structures should not exist. While the symmetry of the multiplets in these narrow channels is determined largely by the symmetry of the channel walls, we also found some multiplets not imposed by the wall symmetry as our most important results. This is most pronounced in Figs. 4 and 5, now for *periodic boundary conditions* on the sidewalls as a better approximation to free growth conditions than the reflection condition.

In summary, the results of Figs. 2, 4, and 5 prove here within numerical possibilities that the observed multiplet structures of *cooperating symmetry-broken fingers* are also dynamically stable objects in three dimensions, as generalizations of the two-dimensional doublons. They dominate the morphology for low enough anisotropy, where dendrites cease to exist. A hexagonal or triplet structure should be expected to occur under free growth conditions from basic symmetry considerations [27], since these growth problems do not have reflection symmetry about some average interface position. The resulting multiplets, consisting of symmetry-broken cooperating fingers of the growing phase, finally seem to be the basic building blocks for the compact seaweed morphology; in particular, the triplet structure appears to be characteristic of free growth. Finally we hope to extend our previous studies of the morphology diagram with the help of these results.

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