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LETTER TO THE EDITOR

Deterministic growth model of pattern formation in dendritic solidification†

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Abstract. A deterministic growth process, based on the solution of the lattice Laplace equation for the temperature field, is introduced for modelling pattern formation in dendritic solidfication. By a simple parameterisation of the physical and environmental conditions during crystal growth a wide variety of dendritic patterns, similar to regular snowflakes, are produced. The fractal scaling of the dendritic patterns is investigated.

The growth of dendritic crystals is a profound example among a wide range of pattern-forming phenomena in physics, chemistry, biology and engineering, where simple dynamic systems spontaneously generate complex structured patterns [1, 2]. The formation of snowflakes [3, 4] is perhaps the most fascinating and puzzling example of these processes. Although subject to intensive efforts, previous attempts have not produced such regular and intricate dendritic structures as those found in nature. Theoretical treatments of the mechanisms leading to these extremely complex patterns have also left a number of questions unanswered. Therefore, a simple approach founded on the basic physics of the problem *and* which produces realistic dendritic patterns, including snowflakes, would be valuable for understanding dendritic crystal growth.

In this letter we introduce a deterministic growth process for modelling pattern formation in two-dimensional dendritic solidification. The essential physics of the problem [1]—heat diffusion, the interfacial boundary conditions and growth anisotropy—are accounted for by solving the Laplace equation with the proper boundary conditions on regular lattices using a relaxation technique. The interface grows deterministically at a rate depending on the local gradient by adding particles to the growing cluster in a way similar to various aggregation models [5, 6]. By varying the parameters of the model to reflect changes in the physical and the environmental conditions during solidification, a great variety of qualitatively different patterns is found, many of which are strikingly similar to real snowflakes—perhaps a result achieved for the first time. Measurements of the radius of gyration exponent indicate the fractal [7] character of these structures. Finally, we measure the exponents describing the scaling of the length and the width of the individual stems emanating from the centre [8] with the mass of the clusters and compare them with recent theoretical predictions. [9]

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The rate-limiting step in the growth of a solid from its supercooled vapour is the diffusion of latent heat away from the moving interface to a cold bath at a large distance R [1]. In the quasi-stationary approximation the reduced temperature field T satisfies the heat-diffusion equation

$$\nabla^2 T = 0. \tag{1}$$

The growth velocity is given by

$$v_n = -k\nabla T \tag{2}$$

where v_n is the velocity normal to the interface, the parameter k is a constant and the thermal conductivity of the solid is assumed to be much larger than that of the liquid or vapour phase. The dependence of the interface temperature on the surface curvature κ is determined by the Gibbs–Thomson condition

$$T_{\text{int}} = 1 - d_0\kappa \tag{3}$$

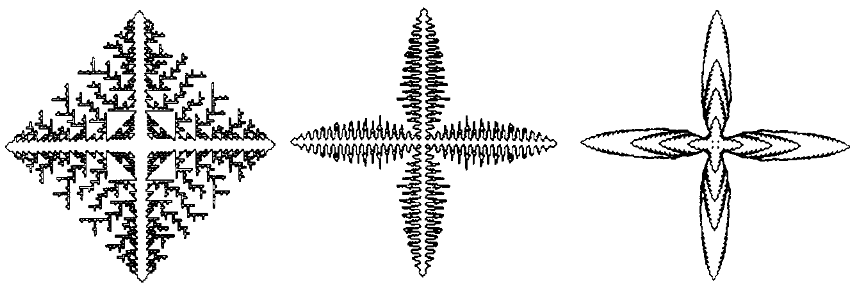
with the capillary length $d_0 = \gamma/L$ depending on the surface tension γ and the latent heat L. Numerical procedures for solving equations (1)–(3) include standard discretisation methods [10] and the boundary integral method [11], which has been shown to be effective for studying the role of crystalline anisotropy [11].

The process we propose is designed to provide the best possible cluster growth method for solving equations (1)–(3) on a lattice and is a descendant of various aggregation models [5, 6], which are particularly suited for simulating diffusion-limited pattern formation [12–18]. The structures generated by the diffusion-limited aggregation (DLA) model of Witten and Sander [5] and the related dielectric breakdown model of Niemeyer *et al* [6] are random fractal patterns, which have been observed experimentally [19, 20] in cases where the surface tension is very small and has no anisotropy.

It has recently been recognised [21, 22] that surface tension anisotropy leads to regular dendritic patterns. This was first discovered in studies of the boundary layer [21] and the geometrical [22] models and seems to be true for more realistic non-local theories [23]. Strong support for this idea was provided by numerical solutions of the Laplace equation [11] and by experiments on viscous fingering [24, 25] which showed that with anisotropy even hydrodynamic systems can produce regular dendritic patterns. Noise inherent in aggregation-type simulations, however, prevents the clusters from having a regular and symmetric shape like real dendritic patterns [17, 18]. Noise reduction methods [17, 18] have been shown to be capable of producing many features of dendritic growth except when the patterns are perfectly symmetric. This suggests the utility of deterministic cluster growth algorithms [15] in which, when necessary, noise can be introduced in a controlled manner. Garik *et al* [15] constructed a deterministic model of fractal growth and obtained regular clusters having some properties of DLA clusters [5] and structures with stable tips.

In our method the growth starts from a seed particle placed on a lattice. After each time step, the temperature field T is calculated by solving the lattice Laplace equation (1) with boundary condition (3) at the surface, and $T = 0$ on a circle of radius R, much larger than the size of the cluster. After the gradients at the surface sites are determined, we normalise them by dividing by the largest gradient. All of these perimeter sites are examined [26] and only those having a normalised gradient larger than a parameter p are filled. The effects of the anisotropic surface tension are induced by the underlying lattice. This approach is completely deterministic, consistent with equations (1)–(3).

In the limit $p = 0$, all the perimeter sites are filled and the resulting structure is a dense polygon with m tips reflecting the m-fold symmetry of the underlying lattice. For finite p the patterns are regular fractals with a fractal dimension which varies from 2 to 1 as p is increased from 0 to 1. This non-trivial result is demonstrated in figure 1(a) which shows the pattern generated on a square lattice for $p = 0.35$. When p is a constant, v_n is constant at surface sites having a normalised gradient larger than p and zero otherwise. In contrast, dendritic growth is governed by equation (2) and the interface growth velocity must be proportional to the local gradient. This implies that, within a time interval Δt, p must vary linearly with time so that sites having the maximum gradient are always filled, while those with smaller gradients are filled less frequently, depending on the local gradient.



(*a*) (*b*) (*c*)

Figure 1. Patterns generated by the deterministic growth model on a square lattice for three sets of parameters. (a) In the absence of surface tension, and for constant p, a regular fractal carpet is generated. The cluster shown was obtained with $p = 0.35$ and has a fractal dimension of ~ 1.61. (b) A parametrisation of p according to (4) with $a = 0.1$, $b = 0.2$, $c = 5$ and zero surface tension produces regular patterns similar to dendritic crystals. (c) The time evolution of a pattern grown with $d_0 = 0.4$ shows how a stable parabolic tip appears as a result of anisotropy and a finite surface tension.

As a discretised approximation, we assume that, during a characteristic time interval $\Delta t = c$, p has the following form:

$$p = a + b(t \bmod c) \tag{4}$$

which is a piecewise function having c steps and approximating a straight line of slope b. Since in every time step a particle is always added to the tips, this characteristic time interval introduces a length scale equal to c lattice spacings. The effect of this length scale is most pronounced for small size clusters when the surface tension is zero and when the lattice induces a high degree of anisotropy in the growth. This can be clearly seen in figure 1(b) where, because of the small size of the arms (~ 100 lattice spacings), the periodic modulations on the surface of the four needle arms is equal to c. On the other hand, in figure 2(a) the effect of c is non-trivially mixed with the lattice spacing on a triangular lattice and the characteristic distances in the modulations are not equal to c. In addition, if the capillary length d_0 is finite, as in figures 1(c), 2(b) and 2(c), the effect of c is not observable.

The calculations were carried out on triangular and square lattices. We used a recently introduced method [12] of estimating the local surface curvature κ by counting the number of filled sites n_r within a circle of radius r ($r = 3$ in the present simulations) centred at the given perimeter site and using the expression $\kappa = (n_r - n_0)/n$, where n_0

is the number of lattice sites corresponding to a flat interface and n is the number of sites in the circle. The values of T on the surface are then calculated from (3). The new values of T at each lattice point are then determined by using the Gauss–Seidel over-relaxation method. With a tolerance of 10^{-6} it took several hundred iterations for the field to relax. We could also take advantage of the symmetry and relax only over a part of the lattice. By keeping the radius R much larger than the cluster size, the shape of the patterns was not affected by this choice. After the gradients at the surface sites have been determined and normalised, sites having gradients larger than p are occupied. The above process is repeated until a large cluster (8–10 000 particles) is generated.

The above method can produce practically all types of observed two-dimensional dendritic patterns by changing the surface tension in (3) and the parameters a and b in (4). The various limiting cases include faceted growth, needle crystals and regular fractal structures, while for intermediate values of the parameters, combinations of these patterns are obtained. In addition, the parameter a in (4) can be varied stochastically during growth resulting in a behaviour which simulates the effect of changing the temperature near the melting temperature. The effectiveness of our method is best demonstrated by the great variety of possible morphologies it generates. Here, we show only a few selected examples of dendritic patterns with fourfold symmetry in figure 1, and in figure 2 we present some examples of sixfold symmetric snowflakes generated by varying the environmental conditions during growth. Figures 1(b) and 2(a) were generated by approximating the linear growth rule (p varying linearly with t) by the stepwise function (4), with $c = 5$. The best approximation to the straight line is obtained by choosing $a = 1/2c$ and $b = 1/c$, i.e. $a = 0.1$ and $b = 0.2$ for $c = 5$. Figure 1(c) shows the time evolution of a pattern grown with finite surface tension ($d_0 = 0.4$). Figures 2(b) and (c) were generated by varying a ($\sim$50 times) to simulate different environmental conditions during the growth of a snowflake. To obtain a faceted near-equilibrium interface pattern, a was made negative ($0 > a > -0.1$), and to obtain a boundary with sharp dendritic shape, a was made greater than zero ($0.5 > a > 0$). As a was changed, b was adjusted to account for the fact that, at the end of each time interval, the straight line must pass through the point $p = 1$.

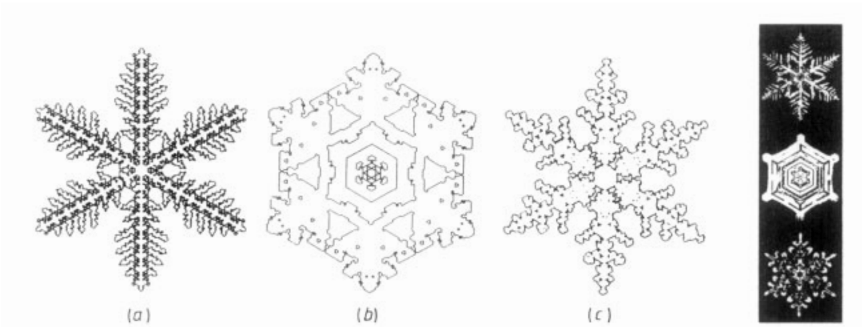


Figure 2. Three examples of patterns generated by the deterministic growth model on a triangular lattice for different values of the parameters, reflecting a variety of growth conditions. (a) Linear growth rule (similar to figure 1(a)) with $a = 0.1$, $b = 0.2$. In (b) and (c) a was changed a number of times during the growth to simulate different environmental conditions. The time evolution of cluster (b), shown by distinct layers, indicates that the intricate internal patterns of real snowflakes are likely to correspond to various stages of the growth of a snow crystal. The insert shows a few typical snowflakes reproduced from [4]. In (a)–(c) $c = 5$.

The question of dimensionality arises naturally and can be investigated by calculating the exponent β, defined by $R_g \sim N^{\beta}$, where N is the number of particles in a cluster with radius of gyration R_g [27]. We have determined β from the slope of log-log plots of R_g against N for a few selected values of the parameters a and b and the results are shown in table 1. Our data indicate that all dendritic patterns generated in the simulations with time-independent values of a and b are fractals [7] on a length scale depending on the parameters with a non-universal fractal dimension.

Table 1. The radius of gyration exponent β of the dendritic patterns generated by the deterministic growth model for selected values of the parameters a and b in the expression $p = a + b(t \text{ mode } c)$, for $c = 5$ and $d_0 = 0$. Although values of β appear to be different statistically, the possibility of a slow crossover cannot be ruled out.

Lattice	a	b	β
Square	0.3	0.0	0.58 ± 0.01
	0.4	0.0	0.67 ± 0.01
	0.5	0.0	0.71 ± 0.01
	0.6	0.0	0.79 ± 0.01
	0.7	0.0	0.89 ± 0.01
	0.1	0.15	0.59 ± 0.01
	0.1	0.20	0.62 ± 0.01
	0.3	0.15	0.79 ± 0.01
Triangular	0.1	0.20	0.58 ± 0.01
	0.2	0.18	0.61 ± 0.01
	0.4	0.13	0.67 ± 0.01
	0.5	0.08	0.70 ± 0.01

The individual arms growing from the origin of the clusters can be characterised by two lengths [8]: the width w and the length l. The way w and l scale with N is a question of great current interest [9]. As a typical example, the dependences of $\ln(w)$ and $\ln(l)$ or $\ln(N)$, for $a = 0.1$ and $b = 0.2$, on a square lattice are shown in figure 3. From the slopes of least-squares fits of the data to straight lines we find that $l \sim N^{\nu_{\parallel}}$, with $\nu_{\parallel} = 0.66$, and $w \sim N^{\nu_{\perp}}$, with $\nu_{\perp} = 0.5$. Recent theoretical considerations [9] have shown that, if the arms approach needle-like shapes asymptotically, then $\nu_{\parallel} = \frac{2}{3}$ independent of the lattice and $\nu_{\perp}^{-1} = 3(D-1)$, where D is the fractal dimension of the cluster. The value of $\nu_{\parallel}$ agrees with this prediction. From the radius of gyration exponent β we estimate $D = 1/\beta \sim 1.6$. This implies that $\nu_{\perp} \sim 0.56$, again consistent with the data in figure 3. From considerations [9] of the critical size needed to observe the needle-like structure it appears that $\nu_{\parallel}$ will be less than $\frac{2}{3}$ for small clusters and for structures with m-fold symmetry where $m > 4$. The reason is that the asymptotic needle-like structures assumed in the theoretical calculations are never reached in reality. In fact, for the triangular lattice we find $\nu_{\parallel} = 0.62$, consistent with this observation.

Simulations of the model introduced in this letter suggest the following conclusions: (i) $\nabla^2 T = 0$ with boundary conditions $v_n = \text{constant}$ for $\nabla T > p$ and $v_n = 0$ for $\nabla T < p$ produces Laplace fractal carpets, (ii) some of the puzzling features of real dendritic structures can be elucidated by solving the Laplace equation on a lattice with appropriate boundary conditions, (iii) the great variety of patterns emerging from the same solidification process is demonstrated to be due to the environmental conditions changing in time. Since $p \leq 0$ corresponds to faceted growth, boundary conditions (3)

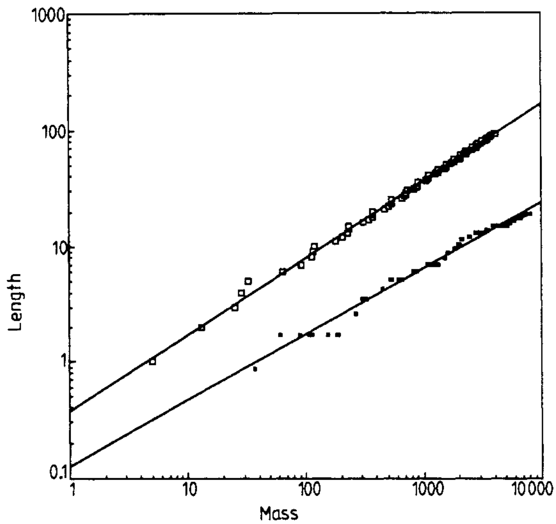


Figure 3. The scaling of the maximal length l (□) and the caliper width w (■) of the arms with the cluster mass N for a pattern generated with $a = 0.1$, $b = 0.2$ and $c = 5$ on a square lattice is demonstrated by the straight lines showing a least-squares fit to the data.

and (4) are capable of simultaneously simulating various growth mechanisms such as surface diffusion. Figures 2(a)-(c) indicate that such complex structures as snowflakes are produced by the interplay of such factors varying in time.

The approach presented here appears to be more effective in producing complex dendritic patterns than the previous methods based on numerical solutions of the solidification equations and can be a useful tool for sorting out various long-standing and puzzling aspects of dendritic pattern formation. In particular, such questions as noise-driven sidebranching [28], and the role of non-lattice-induced anisotropy can be studied using this approach. The deterministic growth model can easily be extended for the investigation of three-dimensional solidification; a fundamental, but much less understood process than two-dimensional dendritic growth.

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