Non-deterministic approach to anisotropic growth patterns with continuously tunable morphology: the fractal properties of some real snowflakes

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LETTER TO THE EDITOR

Non-deterministic approach to anisotropic growth patterns with continuously tunable morphology: the fractal properties of some real snowflakes

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Abstract. We demonstrate that a tunable family of patterns resembling a range of experimentally observed snowflakes is obtained from a simple but somewhat *ad hoc* non-deterministic growth model—an extension of the Niemeyer-Pietronero-Wiesmann η model in which we (i) use 'noise reduction' to enable the asymptotic behaviour to become more readily apparent from simulations of reasonable size, (ii) replace the η model boundary conditions by the boundary conditions of the diffusion-limited aggregation (DLA) model and (iii) include a surface tension parameter. We measure the fractal dimension of some real snowflakes and obtain quantitative agreement with certain cases of the model. We conclude that the model, although somewhat *ad hoc*, gives patterns that are in both qualitative and quantitative agreement with a range of real snowflake patterns.

What are the physical principles underlying the formation of snowflake patterns? This is the classic question of considerable current interest. There is no answer to even the simplest of questions that one can pose about snowflake growth, such as why the six arms are roughly identical in length and why the overall pattern of each arm resembles the five others. In particular, the question of precisely what sort of physical mechanism gives rise to the irregular growth of long thin side branches has attracted many authors interested more generally in pattern formation in the presence of anistropy [1, 2]. Essentially all of these approaches have been deterministic in nature and the resulting patterns are perfectly symmetric! Thus these models have built into them—by deterministically requiring all six arms to be completely identical not only in length but in every other respect—the solution to the two puzzles: equal arm length and similar arm form. An experimental fact not widely known is that 'regular snowflakes are the exception, not the rule': *no two branches of the same snowflake are exactly alike* [3].

Is it possible that a different approach is called for, one based on the physics of random systems? Here we shall take a tentative step in this direction by exploring the consequences of an extremely simple *random* model which produces continuously tunable growth patterns that resemble a substantial fraction of experimental snowflake morphologies. Snowflake formation is thought to involve mainly the aggregation of water molecules and very tiny ice particles. Accordingly, we begin by recalling a family of models of the diffusion-limited aggregation (DLA) type; generally, these represent the solution of the Laplace equation in an isotropic medium with a 'sink' (the growth

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pattern) that is growing in time [4]. The Laplace equation describes the concentration of particles acted on by random forces and the DLA has been used to model a range of phenomena involving aggregating particles [4]. To the extent that snowflakes grow by accreting water molecules previously in the vapour or liquid phase, the growth rate is thought to be limited by the diffusion away from the growing snowflake of the latent heat released by these phase changes. Under conditions of small Peclet number, the diffusion equation describing the space and time dependence of the temperature field $T(\boldsymbol{r}, t)$ reduces to the Laplace equation [2]. Thus a reasonable starting point is DLA, independent of whether we wish to focus on particle aggregation, heat diffusion, or both. DLA reflects well the randomness inherent in a wide range of growth processes, including colloidal aggregation, but it fails to describe dendritic solidification: while the deterministic models of snowflakes produce patterns that are much too 'symmetric', the DLA approach suffers from the opposite problem: DLA patterns are too 'noisy'.

The approach we propose here retains the attractive features of DLA and, at the same time, produces patterns that resemble real (random) snowflakes. Firstly, we introduce a controllable level of noise reduction [5-7]: each perimeter site has a counter, which increments by one each time the perimeter site is chosen using the growth algorithm. The perimeter site becomes an actual cluster site only when the counter value reaches a threshold value s. It is believed [6, 7] that noise-reduced DLA ($s > 1$) is in the same universality class as ordinary DLA ($s = 1$), i.e. it has the same fractal dimension d_f, the only difference being an increase in the characteristic local length scale W_f†. However, one requires s random walkers for each new cluster site, so a more efficient method is called for. We therefore directly solve the Laplace equation for the growth probability p_i (the probability that the perimeter site i is the next to grow) using the same relaxation method used previously for the dielectric breakdown model (DBM) [8], which differs from DLA in the boundary conditions‡. We do not explicitly introduce anisotropy—the only anisotropy present is that arising from the underlying triangular lattice.

Figure 1(a) shows such a pattern for one value of the noise reduction parameter $s = 200$. We obtain the same general pattern for all values of s greater than about $s = 100$—the effect of increasing s seems to be that of increasing the width W_f of the fingers and side branches§. Note, however, that the fjords between the six main branches contain much empty space. Some snowflakes have such wide 'bays' but some do not. A better model would seem to require some tunable parameter that enables the complete range of snowflake morphologies to be generated. We have found one such parameter, η, that has the desired effect of reducing the difference in the ratio of the growth probabilities between the tips and fjords. Specifically, we relate by the non-linear rule $p_i \propto (\nabla\phi)^\eta$ the growth probability p_i to the driving potential ϕ (e.g. ϕ may be the temperature $T(\boldsymbol{r})$ at a point $\boldsymbol{r}$ or the probability that a tiny ice particle is at point $\boldsymbol{r}$). Our model is the analogue for DLA of the non-linear η model [8]. It

† For example, on a square lattice, a dendritic pattern was recently discovered by Meakin in 'noisy' DLA ($s = 1$) provided the mass is allowed to increase to roughly 4000 000 sites: we find a similar dendritic pattern in noise-reduced DLA already for a mass of only 4000 sites!

‡ For DLA a walker need only step on any perimeter site i for i to become a cluster site, while for DBM the random walker must actually *step on* the cluster in order that the last visited site becomes a cluster site.

§ Our finding that dendritic structures are present for all s differs from [7], where a transition to needle-like structures was reported for a large value of s, $s = 400$. The difference arises from the fact that we generate patterns of up to 4000 sites while the clusters of [7] are ten times smaller (as a test, we fixed $s = 400$, varied the cluster mass between 100 and 4000 and observed an apparent crossover from needles to dendrites).

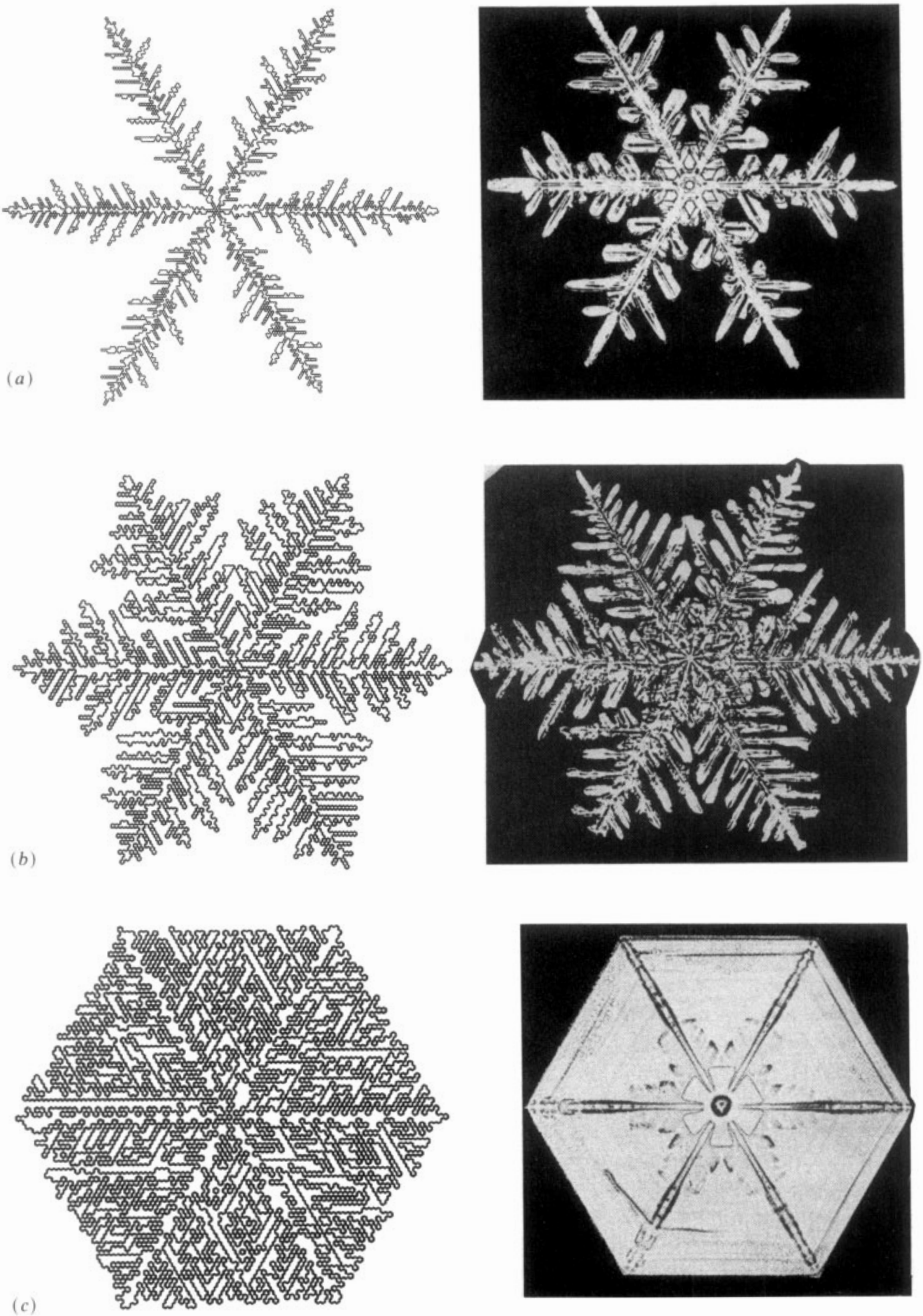

Figure 1. Typical growth patterns with 4000 particles and $s = 200$. (a) $\eta = 1$ ($d_f \simeq 1.5$), (b) $\eta \simeq 0.5$ ($d_f \simeq 1.85$), (c) $\eta = 0.05$ ($d_f \simeq 2$) and (d) $\eta = 1.50$ ($d_f \simeq 1.4$). The same values of d_f are found for the experimental patterns shown.

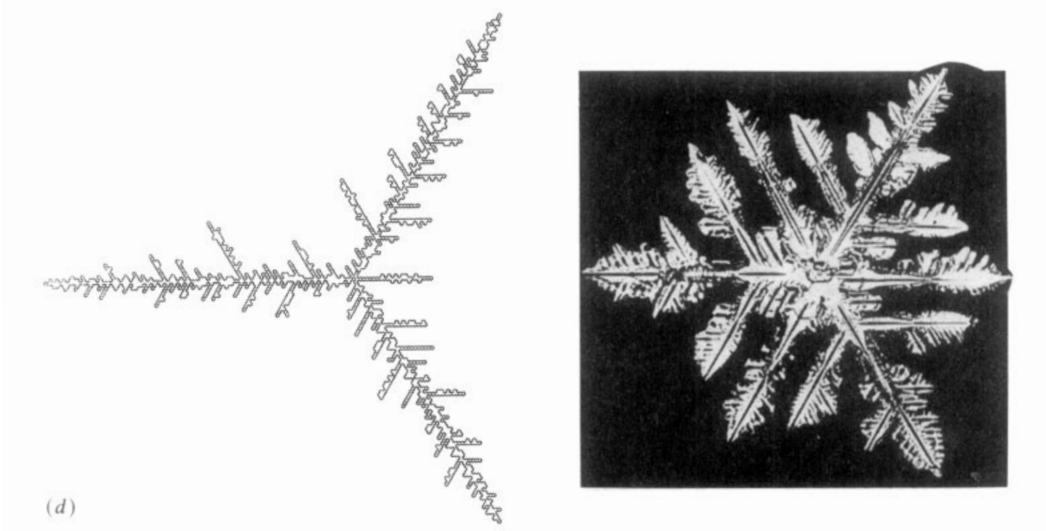

Figure 1. (continued).

must be emphasised that our model is somewhat *ad hoc* in the sense that there is no way of justifying a non-linear growth law, nor can we justify the non-conservation obtained when we set $\nabla^2\phi=0$, instead of $\nabla|\nabla\phi|^{\eta-1}\nabla\phi=0$.

The strength of the non-linearity parameter η tunes the balance between tip growth and fjord growth and by varying η we found growth patterns (figures 1(*b*) and (*c*)) resembling a wide range of experimentally observed snowflake morphologies [3–4]. Thus, while figure 1(*a*) resembles photo 3, p 182 of [3], figure 1(*b*) resembles photo 12, p 192 and figure 1(*c*) resembles photo 12, p 31. We have also calculated patterns with $\eta>1$ (e.g. the case $\eta=1.5$ is shown in figure 1(*d*)). Now the balance between tip and fjord growth is shifted in favour of tip growth. As a result, three of the arms grow so fast that they shield the other three. Experimentally, one also finds three-armed snowflakes (cf photo 10, p 197 [3]).

We found that the effect of tuning a surface tension parameter σ is to thicken the side branches, to round the sharper points of the pattern and to fill in the tiny holes that are present even for small values of η, since the potential ϕ_{IF} on the interface is not constant ($\phi=\phi_0$) but rather changes with the radius of curvature R_c: $\phi_{\mathrm{IF}}=\phi_0-\sigma/R_c$. We increase the growth probability for site i in proportion to the number of cluster sites inside a small box centred about site i; since we expect R_c to be of the order of a pixel length, we set $1/R_c=(4-\mathrm{NN})/3$, where NN denotes the number of occupied neighbours of the central cluster site. Of course, σ is best normalised with respect to the gradient at time zero.

To better understand the role of the tunable parameters σ and η in determining pattern morphology, we have constructed a phase diagram with one parameter on each axis. We systematically constructed 200 patterns for 20 different values of η and 10 different values of σ. Each pattern resembles some experimental snowflake pattern, e.g. figure 2(*a*) resembles photo 4, p 150 of [4], figure 2(*b*) photo 10, p 94 and figure 2(*c*) photo 7, p 145.

Are real snowflakes fractal objects? This intriguing question has been the object of considerable discussion in recent years. Our growth patterns *are* fractal, except in

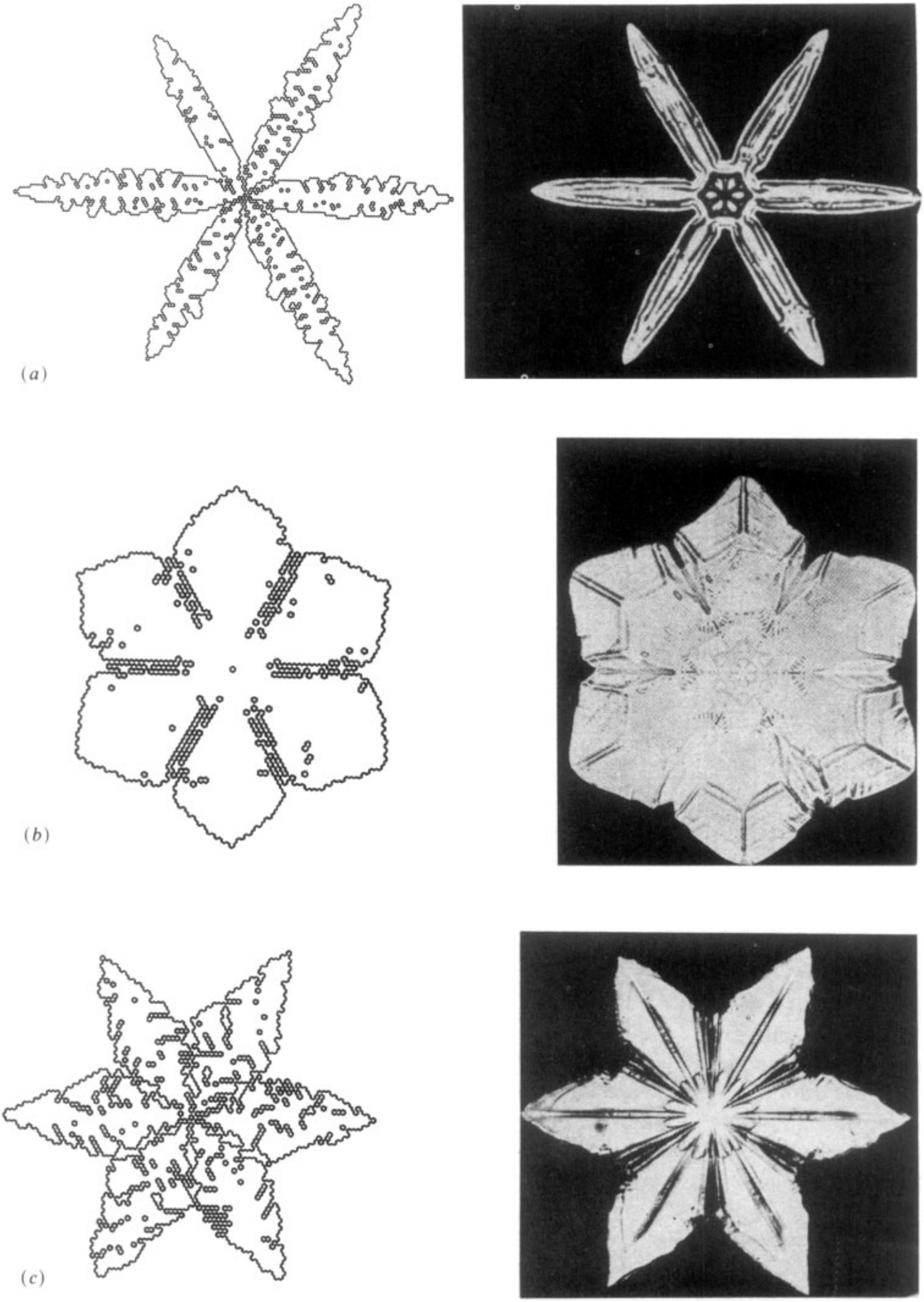

Figure 2. Typical growth patterns (also 4000 particles) with non-zero surface tension σ. Again, $s = 200$. (a) $\eta = 1.0$ and $\sigma = 0.1$, (b) $\eta = 0.5$ and $\sigma = 0.5$ and (c) $\eta = 0.1$ and $\sigma = 0.2$. The experimental patterns [3] are discussed in the text.

the limit $\eta \to 0$. We found that the fractal dimension d_f is independent of the value of the noise reduction parameter s (s seems to mainly renormalise the cluster mass), but d_f does depent on η. We found that our values for d_f agree remarkably well with values we obtain by digitising the corresponding photographs of experimentally observed snowflakes (figure 3).

Before concluding, we make the following remarks.

(i) We also found twelve-arm snowflakes (e.g. with $\sigma = 0.3$ for the quarter of growth and $\sigma = 0$ for the remainder). Such twelve-arm snowflakes are also commonly observed experimentally (cf [3], p 197, photos 2, 4, 6 and 7). This puzzle has been rationalised as arising from the perfectly symmetric 'fusion' of two six-arm snowflakes, but such an improbable symmetric event *need not occur* in order to obtain twelve-arm flakes.

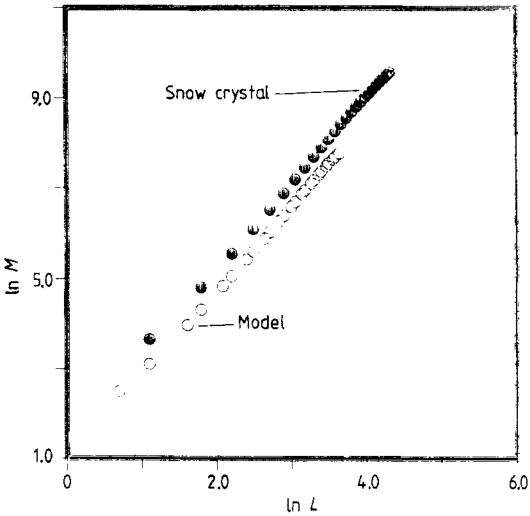

Figure 3. Typical log-log plot of the cluster mass M within a box of edge L as a function of L. Compared are the model and experimental pattern of figure 1(b). The same slope, $d_f = 1.85 \pm 0.06$, is found for both. The experimental data extend to larger values of L, since the digitiser used to analyse the experimental photograph has 20 000 pixels while the cluster has only 4000 sites.

(ii) Of course, no element in nature has a perfectly linear response. When η is a positive integer (say k), a perimeter site grows only if it is chosen k times in succession ($k = 1$ is pure DLA) [9]. Moreover, the response of a network of many linear elements each with a random distribution of threshold values *is* non-linear. Specifically, Roux and Herrmann [10] have recently found that if *linear* elements with a distribution of threshold values ε are combined in a network, then the response of the entire network is *non-linear*!

(iii) Many snowflakes possess relatively compact cores with ramified dressing on their surfaces, arising from different environments of assembly, and possibly also from melting and structural rearrangement taking place after formation. We can mimic the effect of the changing environments in which a given snowflake is actually assembled by varying parameters such as η or σ *during the course of the growth process of a given snowflake.* We have generated many patterns similar to real snowflakes by allowing for values of η and σ that change during the growth process, e.g. we might choose

$\eta \ll 1$ for an initial fraction f of the growth (thereby creating a hexagonal core) and $\eta = 1$ thereafter (thereby creating a ramified exterior portion).

(iv) No adequate explanation has yet been advanced for why, under certain conditions, a snowflake remains quasi-two-dimensional throughout its growth, despite the fact that the 'assembly plant' is three-dimensional. Our ideas on this subject stem from experience with critical phenomena and from recent theoretical and experimental work on pattern formation, where it was found that even minute amounts of anisotropy are sufficient to stabilise structures of lower effective dimension [2, 11, 12]. Of course, we introduce the anisotropy through a triangular lattice, while real snowflakes are made of molecules with an anisotropic shape. However [11] showed clearly that the same experimental patterns are obtained regardless of whether anisotropy arises from the internal structure of the constituents, or is externally imposed through a scratched cell (as in [12]).

(v) Diffusion of latent heat away from the growing aggregate is of paramount importance in dendritic growth of crystals from a liquid phase. An ideal model should encompass both the diffusion of heat away from the snowflake and the aggregation of particles toward the snowflake. Although both phenomena embody the physics of the Laplace equation, the timescales can be quite different. The two-timescale problem is an object of current investigation.

(vi) One obtains a DLA fractal even if the incoming random walkers have a sticking probability that is less than one. Hence we anticipate that DLA might describe structural rearrangement, and this possibility is being studied.

In summary, we have seen that a *non-deterministic* model produces growth patterns that resemble virtually the entire range of experimentally observed snowflake morphologies, from dendritic structures ($\eta \simeq 1$) to hexagonal plate structures ($\eta \ll 1$). It is hoped that this modest work might stimulate further investigation of the basic physics of random systems needed for the understanding of anisotropic growth patterns.

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