No pattern yet for snowflakes

Attempts to model the growth of highly symmetrical crystal structures are most conspicuous for their failure. The urgent need is for truly cooperative models.

THE problem of calculating the shapes of snowflakes persists. That is the simplest conclusion to be drawn from the best effort so far to apply to the growth of a crystal with intrinsic symmetry the theory of the growth of solid phases by the random aggregation of particles (atoms, say) in circumstances in which growth is limited by the speed of diffusion. For the numerical experiments carried out by Tamas Vicsek from the Institute of Technical Physics at Budapest, Hungary, but working last year at the physics department of Emory University at Atlanta, Georgia, suggest that even in the simplest circumstances (the growth of a two-dimensional crystal on a square lattice), the underlying symmetry is only crudely represented in the shape of the crystal *(Phys. Rev Lett.* **53**, 2281;1984). But Vicsek's calculations do at least suggest what might next be tried.

During the past two years, simulations of diffusion-limited aggregation have become extremely fashionable, partly because they are a natural way of generating fractal structures, partly because of the practical importance of the process. The now standard way of carrying them out is conceptually to take a finite piece of some lattice (for example, a piece of a square lattice a few hundred lattice intervals in each direction), to suppose there is a nucleation somewhere in the lattice, to suppose that particles capable of joining a growing aggregate can enter the stage at random points on the periphery and then migrate through the lattice by means of a random walk from one point on the lattice to a nearest neighbour and then, finally, to suppose that such a particle will stay put once it reaches a lattice point adjacent either to the nucleation centre or to a point occupied by the growing aggregate based upon it. The model is simply if tediously calculated (but computer time mounts up to such an extent that nobody seems to have attempted a three-dimensional lattice). The simplest results are those most simply expected. Newly arriving particles stick at the places where they first encounter neighbours. The result is a randomly constructed network of particles which are less densely packed at the periphery than towards the centre, which is another way of describing a structure with fractal properties — the total mass (or number of particles in the aggregate) increases less quickly than the square of the linear dimension of the growing structure. The greater the distance from the nucleation centre, the fuller the whole structure is of nothing.

The upshot of this kind of stimulation has so far been what might, on reflection, have been expected. The larger the central structure, the much greater the chance that it will intercept the random walk of a newly arriving particle. By the time the aggregation has grown to fill a substantial proportion of the space set aside for it at the outset, the chances that the next random walk will yield a particle that sticks on the side it enters must be high. This is another way of saying that the simulatory system is less a good model of diffusion-limited aggregation as the aggregated structures grow. Perhaps practitioners in the field should scale their models to take account of this source of bias.

The problem of geometrical symmetry is much more difficult, if only because it must involve physical considerations. Vicsek seeks to solve the problem by changing the rules of aggregation on a simulated piece of lattice. First, he says, a randomly-walking particle must not stick whenever it comes next to an occupied site, but the probability that it will stick must be a function of some representation of the local curvature, most simply measured (by computers) as the ratio of occupied to unoccupied sites within some given distance. Second, the argument goes, it makes sense to allow newly arriving particles to relax to nearest-neighbour sites if the potential energy will thereby be decreased (or the number of nearest neighbours increased).

The two modifications of the sticking rule that Vicsek introduces are not as simple-minded as they may seem. In the real world of solids and fluids, there is every reason why an aggregating particle's chance of sticking to a growing surface should depend on the most immediately local curvature, for which purpose a simple count of the immediately neighbouring sites which are occupied should be a good approximation to the Gibbs–Thomson macroscopic rule relating aggregation probability to surface tension. That newly aggregated particles should relax to neighbouring lattice sites on a growing aggregate is also commonsensical; at the least, this may be a good simulation of what happens in a reality, the permanent sticking of new particles that reach energetically favoured sites, and the rapid solution (volatization) of particles that first stick elsewhere.

The snag, in the sequence of four simulations of growth on a two-dimensional square lattice reproduced with Vicsek's paper, is that the outcome is far from being a structure with fourfold symmetry that would be expected. To be sure, there are signs of predominant growth in each of four axial directions, largely as a consequence of the positive correlation between sticking and curvature, but there is no detailed simulation of the exquisite detailed rotational symmetry embodied in every other ice crystal (where the axis of ratation is, of course sixfold and not fourfold).

Vicsek says in his conclusion that the outcome of his simulations is only qualitative, which is fair enough. Nobody would pretend otherwise of a simulation of a real physical system in which only the interactions between nearest neighbours are counted as significant. The obvious difficulty, of course, is that there can be very little hope of reaching better conclusions by taking account of next-nearest neighbours, or of further refinements of that kind. And a little reflection will show why no other conclusion is possible. The aggregation of particles onto a growing surface will be determined exclusively by local properties, among which surface tension and the opportunities for energetically advantageous migration will be important. But the symmetry of a whole crystal, represented by the exquisite sixfold symmetry of the standard snowflake, must be the consequence of some cooperative phenomenon involving the growing crystal as a whole. What can that be? What can tell one growing face of a crystal (in three dimensions this time) what the shape of the opposite face is like? Only the lattice vibrations which are exquisitely sensitive to the shape of the structure in which they occur (but which are almost incalculable if the shapes are not simply regular).

What Vicsek seems therefore to have accomplished is to have demonstrated the limitations of computer simulation in the representation of high crystal symmetry. The simple sticking rules of the model for diffusion-limited aggregation may serve well enough for dealing with the growth of amorphous particles (soot in the atmosphere, for example) but they are unlikely to throw much light on the reasons why so many simple crystal-growing operations yield such symmetrical structures. Back now in Budapest, Vicsek may feel inclined to think of adding the complexity of lattice vibrations to the problem of simple Ising lattice calculations. But if not him, then somebody. Please. **John Maddox**