Theory of Ballistic Aggregation

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We give a mean-field, continuum treatment of ballistic aggregation on a seed and a line. The treatment is deterministic, except for one statistical assumption, the so-called tangent rule which determines the mean direction of growth. Our treatment represents progress toward the explanation of the columnar microstructure.

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In recent years much interest has focused on nonequilibrium aggregation processes, that is, the formation of structures by the irreversible addition of subunits from outside. An example of such a process is diffusion-limited aggregation[1] (DLA) where fractals are formed. A simpler problem than the diffusion-limited case (where the aggregating particles perform random walks) is ballistic aggregation. In this process particles moving in straight lines are added to a structure whenever they touch a previously added particle. Early work on this problem seemed to show that fractals were produced, but it is now believed both on the basis of more detailed numerical studies[2] and from analytical results[3] that ballistic aggregates are amorphous solids of fixed density. Nevertheless, the patterns formed in this simple problem are both intriguing theoretically and technologically interesting. In Fig. 1 we show two types of ballistic aggregates: one (a "fan") formed by attachment to a seed[4] and another by attachment to a plane of a beam of nonnormal incidence.[5] In both cases the particles all move in parallel straight lines, as shown, from random launching points. The similarities of the patterns are striking. The peculiar long open streaks are the unexpected feature.

For the case of attachment to a plane these patterns are known as the columnar microstructure. The columns form both in computer simulations (as shown in Fig. 1) and in the real world in vapor-deposited thin films of both metals and insulators. For example, aluminum films deposited on cold substrates often show this morphology. An understanding of this structure is of particular technological interest as the surface properties, notably electrical and optical, are substantially modified from the bulk properties of the material by the surface microstructure. See Ref. 5 for more detail and actual experimental results. Even in the presence of short-range attractive forces among the atoms, which curve the straight-line trajectories, columns still form in numerical simulations.[5] The streaks and columns have not heretofore been explained.

At first glance, it seems that any such explanation would be very complicated because the voids clearly arise from shadowing of one part of the structure by another. In fact, this is an interesting feature of the system; precisely the same sort of nonlocal shadowing produces the fractals of the DLA problem. However, we will show here that many features of the structures can be explained in a remarkably simple way.

In the next section we propose a kind of mean-field treatment for aggregation both on a point and on a line. We will always consider a situation in which the particles move in two dimensions. We will consider coarse-grained statistical fluctuations and try to specify only the average behavior of a portion of the interface. In the last section we summarize our results.

A remarkable feature of the columnar microstructure was pointed out in Ref. 5: Though the columns fluctuate in direction, on the average they are *not* parallel with the incident beam. In fact, an empirical relation between α, the angle between the incident beam and the normal to the plane, and β, the column angle, is

$$\tan\beta = \tfrac{1}{2}\tan\alpha. \tag{1}$$

This relation is obeyed with remarkable accuracy over a wide range of angles. It is easy to see, in a general way, where this comes from: Particles passing the "high" side of an existing column can be caught and cause it to tilt towards the normal, so that β should be less than α.

A rough argument for Eq. (1) might go as follows[6]:

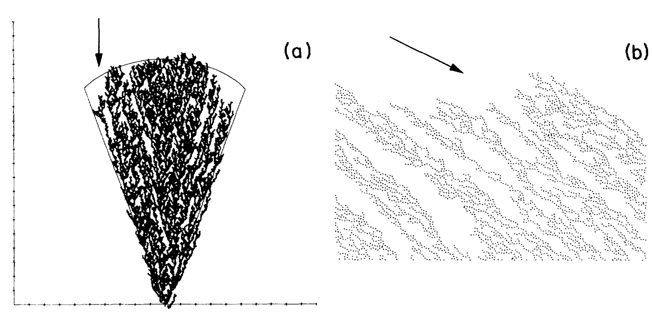

FIG. 1. (a) Ballistic aggregation on a point seed. The solid lines are the prediction from Eq. (4). The particles rain vertically. (b) Ballistic aggregation on a line. The direction of the incident beam is shown.

Replace the particles being deposited by vertically oriented rods of length equal to the diameter of a particle, d. Now rain the rods at random onto a half-line [see Fig. 2(a)] from a direction α to the vertical. No rod can land in the shadow of another already deposited. Since the length of a shadow is $d \tan\alpha$, the location of the stick closest to the end of the half-line must be within a distance $d \tan\alpha$ from the end. Its mean position is just $(d \tan\alpha)/2$. The direction of growth of the edge of the line is given by the angle β, defined by the direction from the edge of the line to the top of the rod closest to it, yielding Eq. (1) as an average relationship. This argument has obvious weaknesses. Whether the reader chooses to regard Eq. (1) as a result of numerical simulation is a matter of taste. In any case, in what follows we will adopt Eq. (1) as an axiom. We will regard it as the rule giving the average direction of the velocity of a portion of the growing interface of the aggregate which will be described in coarse-grained continuum language. Equation (1) is the only statistical feature of our treatment. The rest of the development is in terms of deterministic growth equations. Now, since $\theta = \alpha - \beta$ is the angle between the incident beam and the growth direction (see Fig. 2), and

$$\theta = \tan^{-1}[\tan\alpha/(2+\tan^2\alpha)] \tag{2}$$

has a maximum at 19.5°, we would never expect to see a fan (as in Fig. 1) with an opening angle larger than 19.5° away from the incident direction. In fact, as we will see below, this limit is precisely attained in off-lattice simulations of ballistic aggregation on a seed.

We will first use Eq. (1) to formulate the asymptotic shape of a ballistic fan structure grown on a point seed. We can assume in the long-time limit that each element of interface moves in a straight line. That this must be so is clear: Otherwise at some point the fan would show a buildup of density beyond the solid limit.

Consider a situation in which particles moving in the $-y$ direction rain onto a seed at the origin. Then (see Fig. 2)

$$\partial y/\partial x = -\tan\alpha, \tag{3a}$$

$$y/x = \tan(\alpha-\beta). \tag{3b}$$

These, together with Eq. (1), completely specify the problem. Converting to polar coordinates we see that

$$\partial r/\partial\theta = r\tan\beta. \tag{3c}$$

The polar angle, θ, is measured from the y axis, and is specified in Eq. (2).

It is not difficult to solve this equation. The relevant branch of the solution is

$$r = r_0 f(\theta), \tag{4a}$$

$$f(\theta) = \frac{1}{\sqrt{2}|\sin\theta|}\,\frac{(n+1)^{1/4}(n-1)^{1/2}}{(3n-1)^{3/4}}, \tag{4b}$$

$$n = (1-8\tan^2\theta)^{-1/2}. \tag{4c}$$

Here r_0 is a (time-dependent) constant. It can be shown that f can be written in a more compact form:

$$f = (\cos\alpha)^{1/2}/\cos\beta. \tag{5}$$

There are several interesting features of the solution. The limiting fan angle appears naturally here: When $\tan\theta = 1/\sqrt{8}$, n diverges. This corresponds to $\theta = 19.5°$, as we mentioned above. The significance of the limiting angle is easy to understand: If we start with a surface at a larger angle it will move into the shadow of a less tilted surface and stop growing. In Fig. 1(a) we have superimposed the asymptotic profile, Eq. (4), on an off-lattice numerical simulation. The agreement for the opening angle is essentially perfect. We have grown and examined ten such fans of 12 000 particles each. Their opening angles are $18.3 \pm 1.50°$, in agreement with our prediction. The growing front is less well represented by the prediction. We will see the reasons for this discrepancy below.

Implicit in our postulate of stable asymptotic growth is the time dependence of r_0: We must have constant asymptotic velocity of growth, $r_0 = v_0 t$. The basic assumption of the model is that a constant flux of particles, $\mathbf{J}$, rains on unit area of fan per unit time. In length dl of interface we collect

$$dN = J\cos\alpha\, dl\, dt \tag{6a}$$

particles. The area swept out in this time is

$$dA = f v_0\, dl\, dt \tag{6b}$$

Thus the density is given by

$$\rho(\theta)/\rho(0) = \cos\alpha/f. \tag{7}$$

This relation is plotted in Fig. 3, and compared to simulation results. Since the density depends only on the direction of growth relative to $\hat{\mathbf{J}}$, when

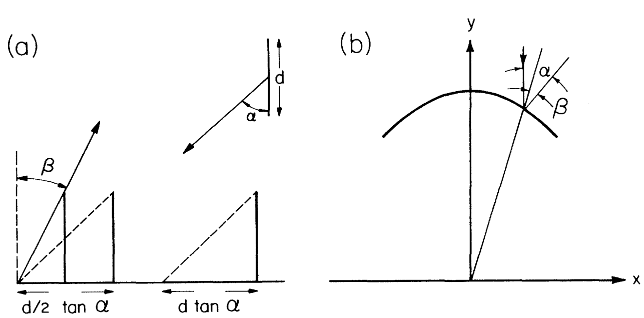

FIG. 2. (a) Geometry depicting average direction of growth at the edge of half-line. (b) Geometry of the growing fan.

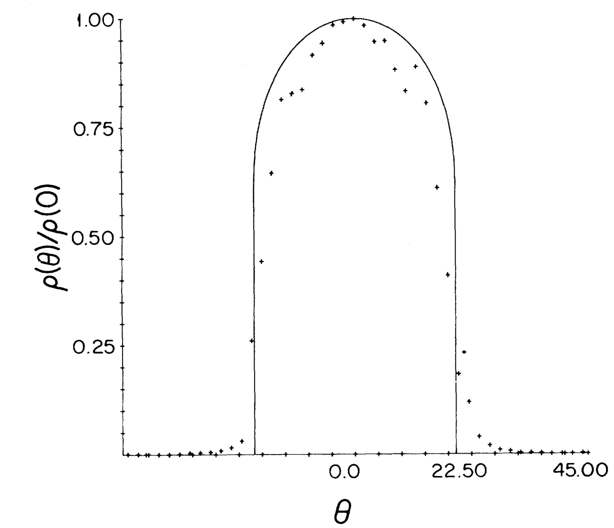

FIG. 3. Density of particles in a fan. The solid line is the prediction of Eq. (7) and the crosses are simulation results.

$\alpha > \tan^{-1}\sqrt{2}$, it must be replaced by α' in Eq. (7) where $\tan\alpha \tan\alpha' = 2$.

We now turn to the formulation of an equation of motion for the growing front. We specify the velocity of each element of interface whose normal is $\hat{\mathbf{n}}$:

$$\mathbf{v} = v\hat{\mathbf{m}}, \tag{8}$$

where $\hat{\mathbf{m}}$ is a unit vector in a direction between the incident beam and the normal, as given by the tangent rule, Eq. (1). To be consistent with the asymptotic results we take

$$v = -\hat{\mathbf{n}}\cdot\mathbf{J}/\rho = v_0 f, \tag{9}$$

where f is given by Eq. (5). We must adjoin to Eq. (9) the condition that if a surface is in a geometrical shadow, e.g., if $\cos\alpha < 0$, there is no growth.

The growth rate of the surface in any direction, $\hat{\mathbf{e}}$, is clearly

$$R = \hat{\mathbf{n}}\cdot\mathbf{v}/\hat{\mathbf{n}}\cdot\hat{\mathbf{e}}. \tag{10}$$

For the fan problem we return to polar coordinates and take $\hat{\mathbf{e}} = \hat{\mathbf{r}}$. From Eqs. (8)–(10) and some algebra we find

$$\partial r(\theta,t)/\partial t = v_0[(1+Q^2)\cos\alpha]^{1/2}, \tag{11a}$$

$$Q = \partial r/r\partial\theta, \tag{11b}$$

$$\cos\alpha = (\cos\theta - Q\sin\theta)/(1+Q^2)^{1/2}. \tag{11c}$$

Note that Eq. (4) is an exact solution to the partial differential equation (11), with the initial condition of growth from a point. In general, of course, an exact solution is not simple to give. We can examine the first approximation of Eq. (11) in powers of Q.

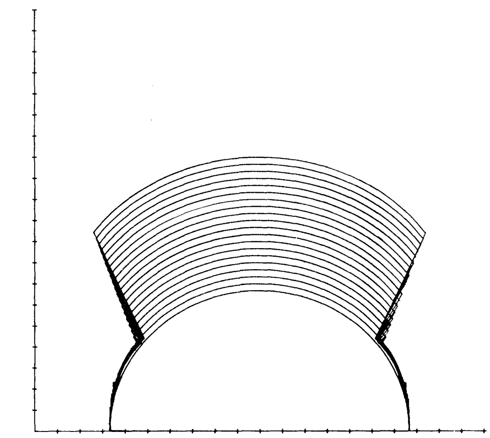

FIG. 4. Numerical solution of Eq. (11) for growth on a semicircle.

Neglecting Q altogether we have

$$r(\theta,t) = v_0 t \cos^{1/2}\theta, \tag{12}$$

which reproduces the profile of Eq. (4) to within 4%. Note that in the entire treatment so far we have not had to consider nonlocal shadowing. This must be put in explicitly when one solves Eq. (11) numerically.

We now turn our attention to the stability of the envelope function $f(\theta)$. We slightly deform the profile and perform a linear stability analysis. Let us denote the solution given in Eq. (4) by $\bar{r}(\theta,t)$. Consider a deformation of the form:

$$r = \bar{r} + \delta(\theta,t). \tag{13}$$

It is not difficult to show that $\partial\delta/\partial t = 0$ to first order in δ. Thus the profile is marginally stable. Of course, the fractional perturbation $\delta/\bar{r}$ decreases with time.

Many initial conditions which are smooth enough develop into the fan shape given in Eq. (4), as we can show by a direct numerical solution of Eq. (11). In Fig. 4 we show growth of a fan starting from a semicircle.

Let us return now to the question of the "streaks" in Fig. 1, and consider how they might arise from a random initial distribution of particles. We represent this crudely by a condition of the form

$$\delta(\theta,0) = \delta_0\cos(m\theta). \tag{14}$$

If δ_0 is small, the marginal stability mentioned above holds. As soon as δ_0 is large enough, a geometric shadow can form. Clearly this happens first near the edge of the fan. Once an area is shadowed it cannot grow further and a channel opens up. In Fig. 5 we show the result of a direct numerical solution of Eq. (11) with $\delta_0/\bar{r}(0) = 0.05$, and $m = 44$. The streaks and ragged edges of Fig. 1(a) probably arise in this way.

We now return to the column structure and apply

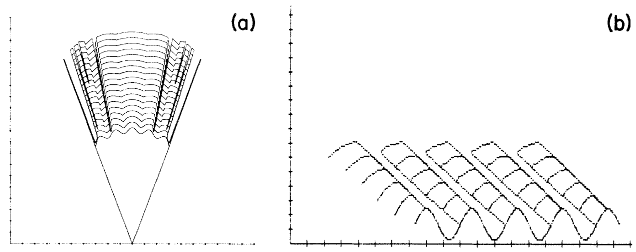

FIG. 5. (a) Numerical solution of Eq. (11) for a perturbed initial condition (growth on a seed). (b) Numerical solution of Eq. (11) for a perturbed initial condition (growth on a line).

the same line of reasoning. A flat surface grows uniformly on the average according to Eqs. (8)–(10). It is, furthermore, marginally stable.[5] An analysis of an initial condition like Eq. (4) (with θ replaced by x) gives the result shown in Fig. 5(b), which recovers the column structure in our mean-field continuum approximation.

The analysis given in this paper is exceedingly simple and attempts only to define certain overall features of the geometry of ballistic aggregates. No serious attempt was made to account for fluctuations about the average behavior other than to introduce asymmetric initial conditions. For example, it seems likely that the column structure should coarsen with time as a result of fluctuations. A description of how this happens could presumably be given starting from what we have done.

Nevertheless, as it stands, the work is not without interest. The columnar microstructure is, as we have mentioned, a ubiquitous phenomena which occurs for thin films of a large number of materials. The columnar instability has important properties. We regard this work as a possible first step in a description of these quantities.

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[6]We are indebted to A. Kerstein for suggesting this to us.