# Causality bound on the density of aggregates

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(Received 2 January 1984)

The irreversible accretion of diffusing particles onto a large cluster results in a tenuous aggregate characterized by a fractal dimension $D_0$ smaller than that of space. The rate of aggregation onto the fastest-growing sites in such a process must not increase indefinitely as the cluster grows. This fact sets a lower limit on the fractal dimension, viz., the dimension $d$ of space minus 1. For aggregation of ballistically moving particles, this bound implies that the aggregate must be compact: The fractal dimension must equal that of space. In general, if the aggregating particles follow trajectories of fractal dimension $D_1$, the bound implies $D_0 \geq d - D_1 + 1$.

The spatial structure of the random clusters formed by the irreversible aggregation of particles has been the subject of several recent studies.[1-3] Many of these aggregates appear to have spatial correlations of power-law form, so that their average density decreases indefinitely as their size increases. The recent Comment of Bensimon, Domany, and Aharony[4] described clusters formed from "ballistic" particles, moving in random straight-line trajectories in two dimensions. They found that the mass $M$ of such a cluster increases as its linear size $R$ according to $M \sim R^{D_0}$ with "fractal dimension"[5] $D_0 = 1.93$. This means that the density decreases as $R^{-0.07}$.

This Rapid Communication describes a bound on the decrease of density of an aggregating object, based on the observation that the growth rate of the outer radius is limited by microscopic considerations: The boundary of the aggregated region can only move at a limited speed. This may be viewed as a sort of causality limit. For diffusing particles in $d$-dimensional space (diffusion-limited aggregation) this bound implies $D_0 \geq d-1$, in agreement with simulation data.[6] For ballistic particles it implies that $D_0 \geq d$, in disagreement with the results of Bensimon *et al.*

The bound describes aggregation from a dilute gas of moving particles of some microscopic radius $a$. Initially the aggregate consists of a seed particle at the origin. Whenever a moving particle touches the aggregate, it is adsorbed, thus increasing the mass and size of the aggregate. Because of this adsorption, the outer radius $R$ of the aggregate grows in time. Initially, the growth speed $dR/dt$ has some value $v_0$ proportional to the density $u$ of moving particles. Later, the growth speed is proportional to the flux of particles onto the outer tips of the aggregate. This flux is partly screened by the rest of the aggregate, and is thus smaller than it was initially. Thus the growth speed can only decrease in time: $dR/dt < v_0 \sim u$. Other characteristic radii, such as the radius of gyration, are necessarily smaller than the outer radius, and thus can grow no faster than this $dR/dt$.

The flux of particles onto an existing aggregate is related in a simple way to its radius. This may be seen for a variety of types of motion using a geometric picture. We imagine the trajectories of all the moving particles as they would have been in the absence of the aggregate. We suppose that each particle takes one step per unit time. For ballistic motion the steps are in a straight line; for diffusive motion the steps are in random directions. The steps may also follow a Levy flight,[5] with a power-law distribution of jumps. The density of steps after time $t$ is simply $ut$, and the average number of contacts with the aggregate is $Mut$. The trajectories which produce these contacts evidently enter the aggregate region, within distance $R$ of the origin. The average number of contacts of such a trajectory with the aggregate depends on the spatial correlations of each. The average number of contacts at a given point is the product of the aggregate density and the step density on that trajectory. The trajectories considered here have power-law density correlations like the aggregate itself. The average number of steps within radius $R$ of a step thus grows as a power $D_1$ of the radius, with $D_1 < d$. The average number of contacts of one trajectory with the aggregate thus goes as $R^{D_0+D_1-d}$. If this power is positive, a typical trajectory within the cluster region intersects the aggregate many times. The number $C$ of *first* contacts between trajectories and the aggregate is the total number divided by the number per trajectory:

$$C \sim MutR^{d-D_0-D_1} \sim utR^{d-D_1} \ .$$

The number is thus independent of $D_0$; since a particle entering the aggregate region has a probability of contact approaching one, the number of first contacts is the same as for a solid adsorbing sphere with radius of order $R$. The aggregate is "opaque" to the particles, and the actual density within the aggregate is not important.

Each time $C$ increases by one, a trajectory touches the aggregate and is adsorbed. Thus the flux onto the aggregate is $dC/dt$. This adsorbed flux increases the mass $M$ of the aggregate: $dM/dt = dC/dt$. This, in turn, can be related to the growth speed $dR/dt$: $dM/dt = (dM/dR)(dR/dt)$. Using our expression for the flux $dM/dt$, and the power-law relation $M \sim R^{D_0}$, this gives

$$uR^{d-D_1-D_0+1} \sim dR/dt \ .$$

This relation for $dR/dt$ was derived for diffusing particles by Deutch and Meakin[7] using a more conventional approach. Since $dR/dt$ is bounded by $v_0 \sim u$, the exponent of $R$ must be nonnegative—even in the dilute limit $u \to 0$. Thus

$$D_0 \geq d + D_1 - 1 \quad .$$

This is the causality limit on $D_0$.

The above argument assumed that $D_0 + D_1$ was greater than $d$, so that the aggregate was opaque to the moving particles. In the opposite limit, the average number of contacts per trajectory in the aggregate region goes to zero. Then the aggregate is transparent to the particles, and growth occurs nearly equally over the entire aggregate. Such growth can only increase the average density until the aggregate is no longer transparent.[8] Thus asymptotically, the cluster cannot be transparent, and $D_0$ values in this range need not be considered.

We are grateful to Philip Pincus for his suggestions on the manuscript.

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