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LETTER TO THE EDITOR

Random space-filling-tiling: fractal properties and kinetics

N V Brilliantov†§, P L Krapivsky†|| and Yu A Andrienko†

† Department of Physics, Moscow State University, Moscow 119899, Russia

‡ Center for Polymer Studies and Department of Physics, Boston University, Boston, MA 02215, USA

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Abstract. A kinetic version of random Apollonian packing model is introduced. In this model, droplets nucleate spontaneously, grow at a uniform rate and stop growing upon collisions. The fractal dimension, D_f of the pore space is found to be equal to $D_f = d(1 - \exp[2 - (2^{d+2} - 2)/(d + 2)])$, a result which is confirmed exactly in 1D and numerically in 2D.

Pattern formation processes are numerous and include, e.g., diffusion-limited aggregation, dendritic growth, dielectric breakdown, solidification of supercooled liquids, growing and coalescing droplets [1–3]. These moving-boundary problems are easy to formulate but extremely difficult to solve. Given the importance and complexity of non-equilibrium pattern formation processes, it is interesting to investigate models which are amenable to theoretical analysis but nevertheless may mimic some features of pattern formation phenomena occurring in nature. In the present study, we describe one such model which is exactly soluble in one dimension and tractable on a mean-field level in arbitrary dimension d . Based on the kinetic description, we then deduce geometric properties of arising patterns including the fractal dimension of the pore space.

There are a number of pattern formation models in which the space-filling patterns are formed by placing some non-overlapping units of smaller and smaller sizes. The oldest construction of this kind, the Apollonian packing (AP) introduced by Apollonius of Perga around 200 BC (see [1]), describes the filling of the space between three mutually touching discs by placing the disc that touches all three discs and then repeating this procedure iteratively [1, 4]. The AP construction gives probably the first scientific example of fractals. The exact value of its fractal dimension is still unknown although rigorous bounds [5] have been found and extremely precise numerical estimates [6, 7] have been recently reported. There are several generalizations of the AP [8–12], including the random AP also known under the name of space-filling-bearing, in which the centres of added discs are also chosen randomly. It has been speculated that (random) AP models may mimic a variety of natural phenomena ranging from the motion of matter in seismic gaps up to some aspects of turbulence (see [9, 12]).

The growth velocity in the random AP model may be considered as infinitely large. Indeed, since the added disc touches the closest disc one can consider it as growing with an infinite rate before collision. Thus the AP model is non-trivial only if a typical size of

§ Also at Max-Planck AG 'Nichtlineare Dynamik' Potsdam University, D-14415, Potsdam, Germany.

|| E-mail address: paulk@buphyk.bu.edu

the system is finite (e.g., if the process takes place in a hole between three touching discs or in a strip).

In this letter we investigate the kinetic version of random AP without these drawbacks. In this model, the space filling is generated by spherical droplets which start to grow at a uniform rate from nucleation sites and stop growing upon collisions. The model may be called the touch-and-stop model or the kinetic AP. Although the condition of non-overlapping resembles the random sequential adsorption model [13] while other features of our model are similar to the Kolmogorov–Johnson–Mehl–Avrami nucleation and growth model [14] both the kinetics and the resulting patterns in the present model are very different. The most interesting property of the kinetic AP is an intimate relation between the fractal properties of arising patterns and the pattern formation kinetics. Namely, we will show that the fractal dimension can be expressed in terms of the exponent describing the long-time asymptotic behaviour of the fraction of uncovered volume.

In general, the process of droplet formation proceeds either by spontaneous nucleation or by growth from initial centres of nucleation. In the latter case, the pattern formation continues until a jamming configuration where further growth is impossible [15]. We will study the more interesting former model which leads to formation of fractal patterns. Note also that only the former model provides an adequate kinetic version of the AP model.

Consider first the kinetic AP model in one dimension. We assume that seeds are nucleated with a constant rate Γ per unit length and then grow with constant velocity V . Following a procedure applied in [16, 17] to nucleation and growth processes, let us first investigate an auxiliary ‘one-sided’ problem in which nuclei are scattered to the right and no nuclei are placed to the left of the origin. Denote by $\phi(t)$ the probability that the origin is uncovered at time t in a one-sided problem. Thus, $(-d\phi/dt)dt$ is the probability that the origin is covered during the time interval $(t, t + dt)$ by some droplet. Such a droplet could have been nucleated at any point x in the spatial interval $(0, Vt)$ between times $t - x/V$ and $t + dt - x/V$. Hence

$$\frac{d\phi}{dt} dt = - \int_0^{Vt} \Gamma dx dt \exp[-2\Gamma x(t - x/2V)] \phi(t). \quad (1)$$

Here $\Gamma dx dt$ is the probability of nucleation of a droplet in the spatial interval $(x, x + dx)$ during the time interval $(t - x/V, t + dt - x/V)$. The exponential factor in the right-hand side of (1) is equal to the product of two factors $\exp[-2\Gamma x(t - x/V)] \exp[-2\Gamma x^2/V]$, where the former factor is the probability that no nucleations have occurred in the interval $(0, 2x)$ during the time interval $(0, t - x/V)$ while the latter factor is the probability of the same during the time interval $(t - x/V, t)$ in the uncovered part of the spatial interval $(0, 2x)$. Finally, the last factor in the right-hand side of (1) ensures that droplets outside the interval $(0, 2x)$ do not stop a droplet covering the origin before time t .

Computing the integral in the right-hand side of (1) and then solving the resulting equation yields

$$\phi(t) = \exp \left[-\Gamma V \int_0^t ds_1 e^{-\Gamma V s_1^2} \int_0^{s_1} ds_2 e^{\Gamma V s_2^2} \right]. \quad (2)$$

Notice that in the derivation of (1) and (2) we have tacitly assumed that the system is initially uncovered and therefore $\phi(0) = 1$.

Let us now return to the original ‘two-sided’ problem. Denote by $\Phi(t)$ the probability that an arbitrary point, say the origin, is uncovered at time t . Since $\Phi(t)$ is just the

probability for a point to be uncovered both from the right and from the left, $\Phi(t)$ is related with $\phi(t)$ by $\Phi(t) = \phi(t)^2$. Using this relation one can find

$$\Phi(t) = \exp \left[- \int_0^T d\omega e^{-\omega} \sinh^{-1} \sqrt{\frac{T}{\omega} - 1} \right]. \quad (3)$$

In deriving (3) from (2) we used the variable $\omega = \Gamma V (s_1^2 - s_2^2)$ and performed the integration over s_2 . In (3) we denoted by T the square of dimensionless time, $T = \Gamma V t^2$. An asymptotic analysis of (3) shows that in the long-time limit, $T \gg 1$, $\Phi(t)$ decays as

$$\Phi(t) \simeq \frac{e^{-\gamma/2}}{\sqrt{4\Gamma V}} t^{-1} \quad (4)$$

where γ is the Euler's constant, $\gamma = 0.577215\dots$

One can obtain an exact expression for more complex correlation functions such as the probability that a droplet nucleated at time t_0 continues growing at time t , $t > t_0$, $\Psi(t, t_0)$. It is not difficult to find that

$$\Psi(t, t_0) = \exp[-2\Gamma V t_0(t - t_0)] \exp \left[-2\Gamma \int_{t_0}^t d\tau V(t - \tau) \right] \phi(t)^2. \quad (5)$$

Here the former exponential factor gives the probability that no particles have arisen in the interval of length $2V(t - t_0)$ during the time interval $(0, t_0)$ while the latter factor is the probability of the same during the time interval (t_0, t) in the uncovered part of the interval $2V(t - t_0)$. The factor $\phi(t)^2$ gives the probability that the droplet which has been nucleated at time t_0 at the centre of the interval $2V(t - t_0)$ had not been stopped up to time t by collisions, neither from the right nor from the left. Thus we arrive at

$$\Psi(t, t_0) = \exp[-\Gamma V (t^2 - t_0^2)] \Phi(t) \quad (6)$$

with $\Phi(t)$ given by (3). Using function $\Psi(t, t_0)$ one can further find the density, $G(L, t)$, of droplets of length L which stopped their growth before time t :

$$G(L, t) = \frac{\Gamma}{2V} \int_0^{t-L/2V} dt_2 \left[-\frac{\partial \Psi(t_1, t_2)}{\partial t_1} \right]. \quad (7)$$

Here t_2 is the time at which the droplet was nucleated, t_1 is the time at which the droplet reached the length L and stopped its growth, $t_1 = t_2 + L/2V < t$. Substituting (6) into (7) gives

$$G(L, t) = \Gamma^2 \exp(\Gamma L^2/4V) \int_{L/2V}^t dt_1 \Phi(t_1) e^{-\Gamma L t_1} \left[t_1 + e^{-\Gamma V t_1^2} \int_0^{t_1} dt_0 e^{\Gamma V t_0^2} \right]. \quad (8)$$

By inserting the asymptotic result (4) and (8) one can find that for sufficiently small dimensionless length, $L\sqrt{\Gamma/V} \ll 1$, the final density of intervals of length L , $G_\infty(L)$, behaves as

$$G_\infty(L) \simeq \frac{\Gamma e^{-\gamma/2}}{\sqrt{4\Gamma V}} L^{-1}. \quad (9)$$

Equation (9) will be used below to calculate the fractal dimension of one-dimensional patterns.

In a recent study [15], we developed a mean-field theory of the kinetic AP model in arbitrary dimension d . We now sketch some of these results which will be used below. Notice that the definition of functions $\Phi(t)$ and $\Psi(t, t_0)$ implies

$$\Psi(t, t) = \Phi(t). \quad (10)$$

Since the growing droplets are spherical the functions $\Phi(t)$ and $\Psi(t, t_0)$ are related by equation

$$\frac{d\Phi}{dt} = -\beta \int_0^t \Psi(t, t_0)(t - t_0)^{d-1} dt_0. \quad (11)$$

Here we have introduced the short-hand notation $\beta = \Gamma\Omega_d V^d$, where $\Omega_d = 2\pi^{d/2}/\Gamma(d/2)$ is the surface area of a d -dimensional unit sphere with $\Gamma(d/2)$ being the Euler gamma function.

Equations (10) and (11) are exact. In [15], we completed the system (10)–(11) by *approximate* equation

$$\Psi(t, t_0) = A\Phi(t) \exp \left[-(\beta/d)(t - t_0)^d \left(t - \frac{t - t_0}{d+1} \right) + B\Phi(t) \right] \quad (12)$$

with $B = (2^{d+2} - 2)/(d+2) - 2$ and $A = \exp(-B)$. A derivation of this equation is similar to the derivation of corresponding *exact* result (5); in one dimension, (12) coincides with (5). In higher dimensions, (12) ignores multi-droplet correlations and thus provides only a mean-field description. Note also that (12) guarantees that the initial condition (10) holds manifestly.

Substituting (12) into (11) we arrive at the closed-form equation for the single variable $\Phi(t)$:

$$\frac{d\Phi}{dt} = \beta A \Phi \int_0^t dt_0 (t - t_0)^{d-1} \exp \left[-(\beta/d)(t - t_0)^d \left(t - \frac{t - t_0}{d+1} \right) + B\Phi(t) \right]. \quad (13)$$

Analysing (13) one can find that in the long-time limit the main contribution to the integral accumulates near the upper limit, $t_0 \rightarrow t$. Computing the integral asymptotically one finds

$$t \frac{d\Phi}{dt} = -A\Phi \exp[B\Phi]. \quad (14)$$

The solution to this equation decays as a power-law. Writing the final result in manifestly dimensionless form we obtain

$$\Phi(t) \simeq C[(\Gamma V^d)^{1/(d+1)} t]^{-A} \quad A = \exp \left[2 - \frac{2^{d+2} - 2}{d+2} \right]. \quad (15)$$

A dimensionless constant $C = C(d)$ should be determined from complete analysis of (13); in one dimension, (4) shows that $C(1) = \frac{1}{2} \exp(\gamma/2) = 0.374\,653\dots$

Making use of function $\Psi(t, t_0)$ one can compute the final density, $G_\infty(r)$, of droplets of radius r :

$$G_\infty(r) = \frac{\Gamma}{V} \int_0^\infty dt_0 \left[-\frac{\partial \Psi(t, t_0)}{\partial t} \right] \quad t = t_0 + r/V. \tag{16}$$

Here t_0 is the time at which the droplet was nucleated, t is the time at which the droplet reached the radius r and stopped its growth.

If one inserts the expression for $\Psi(t, t_0)$, given by (12), into (16) one finds a cumbersome final result for the density $G_\infty(r)$. However, for our primary goal, i.e., for obtaining the fractal dimension, we must know the behaviour of $G_\infty(r)$ only for small radii. This most important part of the distribution is accumulated in the long-time limit. Using the asymptotic behaviour (15) and keeping only the most significant contribution in the integral in (16) we obtain

$$G_\infty(r) \simeq dCA\Omega_d^{A-1} \Gamma(2-A)(\Gamma/V)^{Ad/(d+1)} r^{-1-d(1-A)}. \tag{17}$$

We can now determine the fractal dimension, D_f , of the pore space by introducing a (dimensionless) cut-off size, ϵ , and calculating the number of droplets per unit volume, $N(\epsilon)$, with dimensionless radii greater than ϵ . (The unit of length in d dimensions is equal to $(V/\Gamma)^{1/(d+1)}$.) When this number behaves as a power law at the small size limit

$$N(\epsilon) = \int_\epsilon^\infty G_\infty(r) dr \sim \epsilon^{-D_f} \tag{18}$$

we conclude that the fractal dimension is D_f . Notice that the porosity $S(\epsilon)$, e.g., the empty volume, behaves as

$$S(\epsilon) = 1 - \frac{\Omega_d}{d} \int_\epsilon^\infty r^d G_\infty(r) dr \sim \epsilon^{d-D_f}. \tag{19}$$

By comparing (17) and (18) or (19) one can derive the final result for the fractal dimension:

$$D_f = d(1-A) = d \left(1 - \exp \left[2 - \frac{2^{d+2} - 2}{d+2} \right] \right). \tag{20}$$

For $d = 1$ we have $D_f = 0$; $N(\epsilon)$ in one dimension diverges logarithmically, $N(\epsilon) \sim \ln(\epsilon)$. This asymptotic result might be derived by using the complete analytical solution (8) for the density $G_\infty(L)$. For other dimensions (20) provides a mean-field answer. In particular, for $d = 2$ (20) gives $D_f = 2(1 - e^{-3/2}) = 1.55374 \dots$

We performed numerical simulations in two dimensions with periodic boundary conditions. The maximum dimensionless time in simulations was equal to 1600. (The units of time and length in 2D are $(\Gamma V^2)^{-1/3}$ and $(V/\Gamma)^{1/3}$, respectively.) We found that the fraction of uncovered area, $\Phi(t)$, decays as a power-law with the exponent $A = 0.22 \pm 0.01$ very close to the mean-field result, $A = e^{-3/2} = 0.223130 \dots$ (see (15)). From the plot of $\ln(N(\epsilon))$ versus $\ln(\epsilon)$ (see figure 1) we determined the fractal dimension of final pattern, $D_f \simeq 1.528$, and again obtained an unexpectedly good agreement with the mean-field prediction.

In summary, we have introduced the kinetic AP model and obtained the intimate relationship between the fractal dimension D_f of the arising patterns and the kinetic exponent

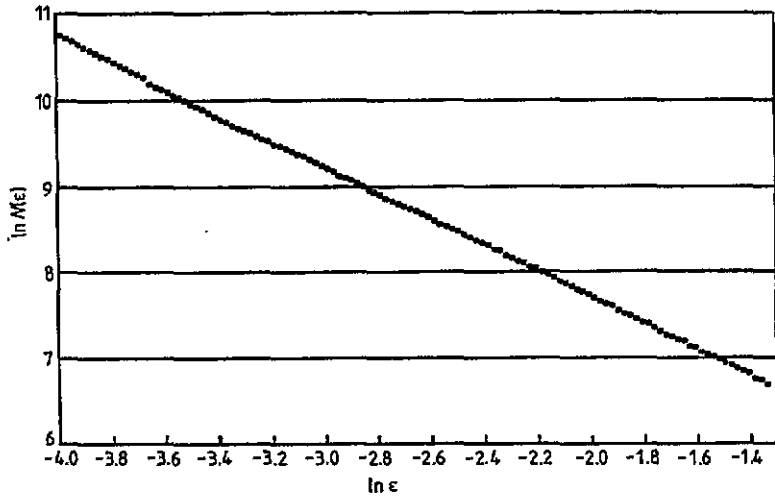


Figure 1. The plot of $\ln[N(\epsilon)]$ versus $\ln(\epsilon)$ where $N(\epsilon)$ is the number of discs per unit area with radii greater than ϵ . The small-scale slope indicates that the fractal dimension of the pore space is 1.528.

A describing the pattern formation kinetics. Although our results are exact only in one dimension, a surprisingly good agreement between theory and simulations in two dimensions suggests that our main findings are valid in arbitrary dimension.

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