Scaling and Multiscaling in Models of Fragmentation

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We introduce a simple geometric model which describes the kinetics of fragmentation of d-dimensional objects. In one dimension, our model coincides with the random scission model and show a simple scaling behavior in the long-time limit. For $d>1$, the volume of the fragments is characterized by a single scale $1/t$, while other geometric properties such as the length are characterized by an infinite number of length scales and thus exhibit multiscaling.

I. INTRODUCTION

The phenomenon of fragmentation which occurs in numerous physical, chemical, and geological processes, has attracted a considerable recent interest. Fragmentation can be exemplified by polymer degradation, grinding of minerals, atomic collisions cascades, energy cascades in turbulence, multivalley structure of the phase space of disorder systems, *etc*. [1–7]. In general, fragmentation is a kinetic process with scattering, breaking, or splitting of particular material into smaller fragments. With such wide-ranging applications it is natural to try to abstract the essential features of fragmentation and to model them as simple as possible. One characteristic feature of these cascade processes is that fragments continue splitting independently. This allows one to describe the evolution by linear rate equations. Another restriction which is used in almost all studies of fragmentation is the implicit assumption that fragments may be described by a single variable, say their mass or size. The simplest model satisfying these restrictions is the so-called random scission model [5,8,9]. In this model, the distribution of sizes is described by the integro-differential equation,

$$\frac{\partial P(x,t)}{\partial t} = -xP(x,t) + 2\int_x^\infty dy P(y,t), \tag{1}$$

where $P(x,t)$ is the concentration of fragments of mass (size) x, x-mers, at time t. The loss term on the right-hand side represents the decrease of x-mers due to binary breakups. The probability of breaking at every point is assumed to be constant in the random scission model and hence the overall rate at which an x-mer breaks is equal to x. The gain term in Eq. (1) represents the increase of x-mers due to breakups of longer fragments. The general solution [5,8,9] to Eq. (1) is

$$P(x,t) = e^{-xt}P_0(x) + e^{-xt}\int_x^\infty dy P_0(y)\left[2t + t^2(y-x)\right]. \tag{2}$$

In the long-time limit, this exact solution approaches the scaling form

$$P(x,t) = Ct^2 e^{-xt}, \quad C = \int_0^\infty dy\, y P_0(y), \tag{3}$$

if we keep xt finite while taking the limit $t\to\infty$ and $x\to 0$.

The random scission model is a representative example of "one-dimensional" fragmentation processes in which fragments are described by a single variable. The kinetics of such fragmentation processes is now well understood and numerous explicit and scaling solutions have been found [4,5,8–14].

The geometry of fragments clearly influences the fragmentation processes. However, it was ignored in so far studied models. In this paper, we introduce simple kinetic models describing the splitting of two-dimensional and more generally d-dimensional objects. We find that multiscaling appears for dimensions larger than one. In section II, we present the two-dimensional model and analyze the behavior of the moments of the size distribution of the fragments. Furthermore, we investigate the area distribution of the fragments an show that it exhibits ordinary scaling. In Section III, we generalize the asymptotic results to arbitrary dimensions and show that multiscaling occurs in higher dimensions as well. In Section IV, we introduce a two-dimensional isotropic fragmentation process. Numerical study of this process suggests that it belongs to a different universality class.

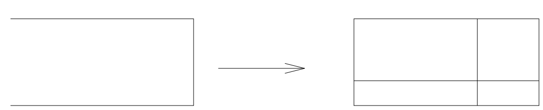

FIG. 1. The fragmentation process.

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II. FRAGMENTATION IN TWO DIMENSIONS

In close analogy with the one-dimensional fragmentation process we investigate the following process in two dimensions. A fragmentation event takes place at an arbitrary internal point of the rectangle and gives birth to four smaller rectangles as illustrated in Figure 1. The distribution function $P(x_1, x_2; t)$ describing rectangles of size $x_1 \times x_2$, is governed by the following kinetic equation:

$$\frac{\partial P(x_1,x_2;t)}{\partial t} = -x_1 x_2 P(x,t) + 4\int_{x_1}^{\infty}\int_{x_2}^{\infty} dy_1 dy_2 P(y_1,y_2;t). \tag{4}$$

Note that Eq. (4) implies the conservation of the total area,

$$\int_0^{\infty}\int_0^{\infty} dx_1 dx_2 x_1 x_2 P(x_1,x_2;t) = const. \tag{5}$$

To analyze Eq. (4) we introduce the double Mellin transform of the distribution function $P(x_1, x_2; t)$,

$$M(s_1,s_2;t) = \int_0^{\infty}\int_0^{\infty} dx_1 dx_2 x_1^{s_1-1} x_2^{s_2-1} P(x_1,x_2;t). \tag{6}$$

The functions $M(s_1, s_2; t)$ at fixed s_1 and s_2 will be called the moments. By combining Eqs. (4) and (6) we arrive at the equation

$$\frac{\partial M(s_1,s_2;t)}{\partial t} = \left(\frac{4}{s_1 s_2} - 1\right) M(s_1+1,s_2+1;t). \tag{7}$$

A surprising feature of Eq. (7) is that it implies the existence of an infinite number of conservation laws. The moments $M(s_1, s_2; t)$ with s_1 and s_2 satisfying the relation $s_1 s_2 = 4$ are independent of time. Thus in addition to the conservation of the total area there is an infinite amount of hidden conserved integrals. These integrals are in fact responsible for the absence of scaling solutions to Eq. (4). Indeed, the scaling solution $P(x_1, x_2; t) = t^w Q(t^z x_1, t^z x_2)$, implies an infinite amount of scaling relations, $w = z(s_1 + s_2)$ at $s_1 s_2 = 4$, which cannot be satisfied by the scaling exponents, w and z.

We will solve Eq. (7) by Charlesby's method [8] (for more recent applications of this method see, *e. g.*, [10,15]). For the random scission of the unit square, $P(x_1, x_2; 0) = \delta(x_1 - 1)\delta(x_2 - 1)$, or equivalently $M(s_1, s_2; 0) = 1$. By iterating Eq. (7) one can compute all derivatives of $M(s_1, s_2; t)$ at $t = 0$ and then find $M(s_1, s_2; t)$ from the Taylor's series, $M(s_1, s_2; t) = M(0) + tM'(0) + t^2 M''(0)/2! + t^3 M'''(0)/3! + \ldots$. This gives a solution in terms of a generalized hypergeometric function [16],

$$M(s_1,s_2;t) = {}_2F_2(a_+, a_-; s_1, s_2; -t), \tag{8}$$

with

$$a_\pm \equiv a_\pm(s_1,s_2) = \frac{s_1+s_2}{2} \pm \sqrt{\left(\frac{s_1-s_2}{2}\right)^2 + 4}. \tag{9}$$

Computation of first few moments gives $M(1, 1; t) \equiv N(t) = 1 + 3t$ for the total number of fragments, $N(t)$; $M(2, 2; t) \equiv 1$ for the total area; and $M(3, 3; t) = \frac{1}{t} + \frac{1}{3t^2} + e^{-t}\left(\frac{1}{6} - \frac{2}{3t} + \frac{1}{3t^2}\right)$ for the next diagonal moment. The first moment can be easily understood. The rate of creation of rectangles is equal to 3 since every fragmentation event introduces 3 additional rectangles, and hence, the total number of rectangles is $1 + 3t$. These results suggest the following power-law asymptotic behavior of the moments $M(s_1, s_2; t)$:

$$M(s_1,s_2;t) \simeq A(s_1,s_2) t^{-\alpha(s_1,s_2)}. \tag{10}$$

Substituting this asymptotic form into Eq. (7) yields the difference equations for the exponent $\alpha(s_1, s_2)$, and for the prefactor $A(s_1, s_2)$,

$$\begin{aligned} \alpha(s_1,s_2) + 1 &= \alpha(s_1+1,s_2+1), \\ \alpha(s_1,s_2) A(s_1,s_2) &= \left(1 - \frac{4}{s_1 s_2}\right) A(s_1+1,s_2+1). \end{aligned} \tag{11}$$

With the boundary conditions, $\alpha(s_1, s_2) = 0$ and $A(s_1, s_2) = 1$ at $s_1 s_2 = 4$, Eqs. (11) are readily solved to give

$$\begin{aligned} \alpha(s_1,s_2) &= a_-(s_1,s_2) = \frac{s_1+s_2}{2} - \sqrt{\left(\frac{s_1-s_2}{2}\right)^2 + 4}, \\ A(s_1,s_2) &= \frac{\Gamma(s_1)\Gamma(s_2)\Gamma(a_+ - a_-)}{\Gamma(a_+ - s_1)\Gamma(a_+ - s_2)\Gamma(a_+)}. \end{aligned} \tag{12}$$

The preceding formulas, Eqs. (10) and (12), may be established rigorously from the asymptotic behavior of the generalized hypergeometric functions.

For ordinary scaling distributions the exponent $\alpha(s_1, s_2)$, describing the asymptotic decay of the moments is linear in the variable $s_1 + s_2$. However, for two-dimensional fragmentation this exponent depends also on the variable $s_1 - s_2$. This manifests the non-trivial scaling properties of the two-dimensional fragmentation process. One can also compare the average value of $x_1^{n_1} x_2^{n_2}$, $\langle x_1^{n_1} x_2^{n_2}\rangle$, defined by

$$\langle x_1^{n_1} x_2^{n_2}\rangle = \frac{\int_0^{\infty}\int_0^{\infty} dx_1 dx_2 x_1^{n_1} x_2^{n_2} P(x_1,x_2;t)}{\int_0^{\infty}\int_0^{\infty} dx_1 dx_2 P(x_1,x_2;t)} \equiv \frac{M(n_1+1,n_2+1;t)}{M(1,1;t)}, \tag{13}$$

with the product $\langle x_1^{n_1}\rangle\langle x_2^{n_2}\rangle$. It turns out that the ratio of these quantities *depends* asymptotically on time t, while for any scaling distribution $P(x_1,x_2;t)$ such a ratio would be a constant. In particular,

$$\frac{\langle (x_1x_2)^n\rangle}{\langle x_1^n\rangle\langle x_2^n\rangle}\sim t^{-\left(\sqrt{n^2+16}-4\right)}. \tag{14}$$

Only in the limit $n\to 0$ this ratio reaches a constant, while for every positive n the ratio decays in time. By considering the case $n=1$ one sees that the average length, $\langle x_1\rangle\sim t^{-(5-\sqrt{17})/2}\sim t^{-.438}$, decays slower than the square root of the average area, $\sqrt{\langle x_1x_2\rangle}\sim t^{-1/2}$. This again confirms that the fragment distribution function $P(x_1,x_2;t)$ in the two-dimensional random scission model does not approach a scaling form in the long-time limit. However, since all the moments still show a power-law behavior we conclude that the model exhibits a multiscaling asymptotic behavior.

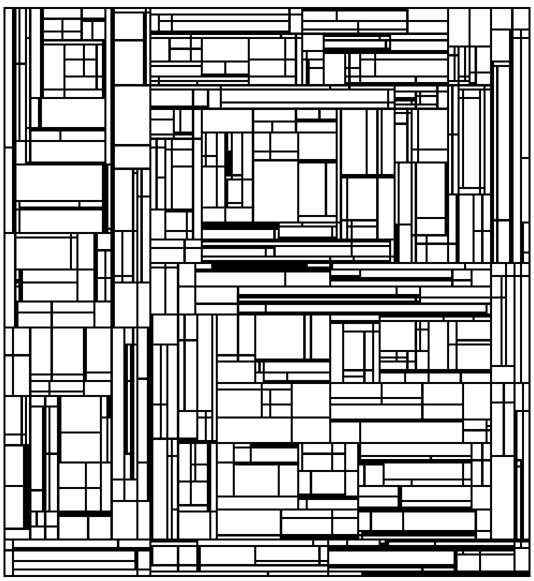

FIG. 2. Realization of the fragmentation process on a unit square at time $t=1000$.

The moments provide an almost complete analytical description of the fragmentation process. However, a snapshot of the system at the later stages remains intriguing (see Fig. 2). This unexpectedly rich pattern arising in such a simple process can be viewed as a consequence of the fact that the process is not fully self-similar. Instead, the pattern is formed of sets of different scales which are spatially interwoven. Fig. 2 also shows that a number of rectangles have large aspect ratio. This qualitative observation is in agreement with the asymptotic behavior of the $n^{\rm th}$ moment of aspect ratio,

$$\langle (x_1/x_2)^n\rangle \to B\; t^{\sqrt{n^2+4}-2} \tag{15}$$
$$B=\frac{\Gamma(1+n)\Gamma(1-n)\Gamma(2\sqrt{n^2+4})}{\Gamma(\sqrt{n^2+4}+n)\Gamma(\sqrt{n^2+4}-n)\Gamma(1+\sqrt{n^2+4})},$$

which is valid for $|n|<1$. [For $|n|\geq 1$, these moments do not exist as follows from a general feature of the d-dimensional random scission model - the moments $M(s_1,\ldots,s_d;t)$ do exist only if $s_j>0$ for all j]. The aspect ratio appears to be growing in time, in other words, perfect squares have a great tendency of breaking into long and thin rectangles.

Let us consider the *area* distribution function, $P(A,t)$,

$$P(A,t)=\int_0^\infty\int_0^\infty dx_1dx_2\delta(x_1x_2-A)P(x_1,x_2;t), \tag{16}$$

which provides a partial description of our system. We will show that $P(A,t)$ approaches a scaling form similar to those found for a number of other one-dimensional fragmentation systems [9,10,12]. Indeed, the diagonal moments scale in time according to

$$M(s,s;t)\simeq\frac{6\Gamma(s)}{s(s+1)}t^{2-s}, \tag{17}$$

or in other words, the normalized $n^{\rm th}$ moments of the area $\langle A^n\rangle^{1/n}$ are all proportional to t^{-1}. Hence, the area distribution function follows the scaling form

$$P(A,t)\simeq t^2\Phi_2(At), \tag{18}$$

where the scaling function $\Phi_2(z)$ satisfies

$$\int_0^\infty dz z^{s-1}\Phi_2(z)=\frac{6\Gamma(s)}{s(s+1)}. \tag{19}$$

Performing the inverse Mellin transforms yields the explicit expression for the scaling function [17],

$$\Phi_2(z)=6\int_0^1 d\zeta\left(\frac{1}{\zeta}-1\right)e^{-z/\zeta}, \tag{20}$$

with the limiting behavior

$$\Phi_2(z)\to\begin{cases}6z^{-2}e^{-z}, & \text{if } z\gg 1,\\ 6\ln(1/z), & \text{if } z\ll 1.\end{cases} \tag{21}$$

Notice that the scaling solution of Eq. (18) is characterized by the same exponents as the scaling solution (3) for the one-dimensional random scission model, $P(x,t)\sim t^2\Phi_1(xt)$. However, the scaling functions are different: $\Phi_1(z)=e^{-z}$ (see Eq. (3)) is regular everywhere while $\Phi_2(z)$ diverges logarithmically near the origin.

One can consider variations of this model for describing the kinetics of fragmentation of multidimensional objects. For example, one can change the governing rule of the fragmentation events (see Fig. 1) and keep only two rectangles, say the rectangle in the bottom left corner and in the upper right one. This rule implies that the total length is conserved while the total area decays to zero. Interestingly, this case has been partially studied in connection with the problem of random sequential adsorption of needles [18]. This model can be treated by applying our approach. One should just change in Eq. (4) the factor 4, corresponding to creation of four rectangles, by factor 2. Many results like Eqs. (8), (10), and (12) remain the same, with

$$a_{\pm} = \frac{s_1 + s_2}{2} \pm \sqrt{\left(\frac{s_1 - s_2}{2}\right)^2 + 2}, \tag{22}$$

instead of Eq. (9). All the qualitative conclusions also do not change: the model exhibits a multiscaling asymptotic behavior and, *e. g.*,

$$\frac{\langle (x_1 x_2)^n \rangle}{\langle x_1^n \rangle \langle x_2^n \rangle} \sim t^{-\left(\sqrt{n^2+8}-\sqrt{8}\right)}. \tag{23}$$

The area distribution function again scales according to

$$P(A,t) \simeq t^{\sqrt{2}} \Phi_2(At). \tag{24}$$

Here the scaling function $\Phi_2(z)$ is given by

$$\Phi_2(z) = C \int_0^1 \frac{d\zeta}{\zeta} (1-\zeta)^{\sqrt{2}-1} e^{-z/\zeta}, \tag{25}$$

where $C = \Gamma(2\sqrt{2})/\Gamma^2(\sqrt{2}) \cong 2.18482$.

In the limits of large and small area one has

$$P(A,t) \to \begin{cases} C A^{-2} e^{-At}, & \text{if } At \gg 1, \\ D t^{\sqrt{2}} \ln(1/At), & \text{if } At \ll 1, \end{cases} \tag{26}$$

with $D = \Gamma(2\sqrt{2})/\Gamma^3(\sqrt{2}) \cong 2.46432$. Therefore the area distribution function again diverges logarithmically in the small-A limit.

III. GENERALIZATION TO HIGHER DIMENSIONS

We turn now to the general d-dimensional random scission model. The asymptotic method presented for the two-dimensional case can be generalized by using a simple geometric construction. We are interested in obtaining the moments $M(\mathbf{s};t)$, where we have used the notation $\mathbf{s} \equiv (s_1, \ldots, s_n)$. In analogy with the two-dimensional case, we will assume the power-law behavior: $M(\mathbf{s};t) \sim t^{-\alpha(\mathbf{s})}$. The exponents α satisfy the following difference equation

$$\alpha(\mathbf{s}) + 1 = \alpha(\mathbf{s} + \mathbf{1}), \tag{27}$$

with the notation $\mathbf{1} = (1, \ldots, 1)$. Meanwhile, the exponents should also reflect the hidden conserved integrals, *i. e.*, $\alpha(\mathbf{s}^*) = 0$ on the hypersurface which is formed of points $\mathbf{s}^*$ satisfying the relation $\Pi_j s_j^* = 2^d$. The solution to Eq. (27) with these boundary conditions is given by the formal expression

$$\alpha(\mathbf{s}) = \alpha(\mathbf{s}^* + k\mathbf{1}) = k. \tag{28}$$

This solution clearly satisfies the boundary condition as well as Eq. (27). Hence the problem is reduced to finding roots of the algebraic equation $\Pi_j(s_j - k) = 2^d$. Since this equation is of degree d, a solution is feasible only for $d \le 4$. An alternative way of viewing the solution is geometric. In Eq. (28), $\mathbf{s}^* + k\mathbf{1}$ represents a line along the $(1, \ldots, 1)$ direction originating at $\mathbf{s}^*$ and ending at $\mathbf{s} = \mathbf{s}^* + k\mathbf{1}$. The exponent $\alpha(\mathbf{s})$ equals the projection of this line on an arbitrary axis, *e. g.* , on the s_1 axis. Figure 3 illustrates this construction.

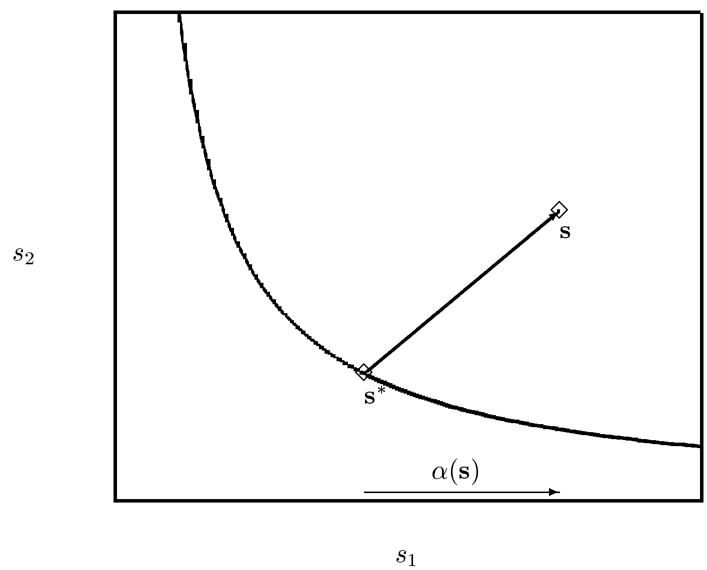

Fig. 3 The Geometric Solution. The hypersurface $\mathbf{s}^*$ satisfies $\Pi_j s_j = 2^d$.

The main features found for the two-dimensional case such as multiscaling occur for higher dimensions as well. As a manifestation of the existence of multiple length scales in the system let us consider the ratio of the average volume $\langle V \rangle$ to the d^{th} power of the average length, $\langle l \rangle$. We define the exponent β_d by

$$\frac{\langle V \rangle}{\langle l \rangle^d} \sim t^{-\beta_d}, \tag{29}$$

or equivalently, $\beta_d = 1 - d\,[\alpha(2, 1 \ldots, 1) + 1]$. Using the construction of Eq. (28), we find $\beta_d = 0, 0.1231, 0.1486$ for $d = 1, 2, 3$ respectively, while in the limit $d \to \infty$ this exponent saturates at $1 - 2\log(3/2) \cong 0.1891$. Note that β_d measures the deviation between the asymptotic behavior of the length and the volume. As the dimension is increased, this discrepancy becomes more pronounced, and hence multiscaling is stronger in higher dimensions. Another consequence of the same phenomenon is the nonuniversal behavior of the various moments of the length distribution. We find that the n^{th} moment decays asymptotically according to

$$\langle l^n \rangle \sim t^{-2\ln(1+n/2)/d}, \tag{30}$$

indicating the presence of an infinite number of length scales.

One can also show that different directions behave independently to a certain degree in the limit of infinite dimensions. Specifically, one can show that

$$\left\langle \Pi_j x_j^{n_j} \right\rangle = \Pi_j \left\langle x_j^{n_j} \right\rangle, \tag{31}$$

if $n_j = 0$ for all indices j except for a finite number. The average over a finite number of variables decouples into a product over single variable averages, while the average over an infinite number does not decouple.

For completeness, we present the general dimension results for the diagonal moments, $M(s, \ldots, s; t)$, which will be shortly denoted by $M(s,t)$. The governing equation for these moments reads

$$\frac{\partial M(s,t)}{\partial t} = \left[(2/s)^d - 1\right] M(s+1,t). \tag{32}$$

We substitute the power-law asymptotic behavior, $M(s,t) \simeq A(s) t^{-\alpha(s)}$, into Eq. (32) and take into account the boundary conditions $\alpha(s=2) = 0$ and $A(s=2) = 1$. By solving the resulting difference equations we find

$$\alpha(s) = s - 2, \quad A(s) = \Gamma^d(s) \prod_{j=1}^{d-1} \frac{\Gamma(2 - 2\cdot\zeta^j)}{\Gamma(s - 2\cdot\zeta^j)}, \tag{33}$$

with $\zeta = \exp(2\pi i/d)$.

In the long-time limit the volume distribution function, $P(V,t)$, approaches the scaling form

$$P(V,t) \simeq t^2 \Phi_d(Vt), \tag{34}$$

with $\Phi_d(z)$ being the inverse Mellin transform of $A(s)$. After a lengthy calculation one can find the asymptotic behavior of the volume distribution function:

$$P(V,t) \to \begin{cases} C_d V^{-2} e^{-Vt}, & \text{if } Vt \gg 1, \\ D_d t^2 \ln^{d-1}(1/Vt), & \text{if } Vt \ll 1, \end{cases} \tag{35}$$

where $C_d = \prod_{1\leq j \leq d-1} \Gamma(2 - 2\cdot\zeta^j)$ and $D_d = 2^{d-1}(2^d - 1)/\Gamma(d)$. Thus for all $d > 1$ the volume distribution function diverges logarithmically in the small-volume limit.

To summarize, in the d-dimensional random scission model, the volume is characterized by only one scale, $V \sim t^{-1}$. However, other geometrical characteristics such as the average length and the surface area decay nonuniversally in time because of the existence of an infinite amount of length scales, namely multiscaling.

One can also consider a varying fragmentation rate and study the case where the overall rate depends on the volume as a power-law, *i. e.*, as V^λ (the case $\lambda = 1$ corresponds to the random scission model). When λ is positive, this generalization also results in multiscaling of the fragments distribution. The total number of fragments $N(t) \equiv M(\mathbf{1}; t)$ grows algebraically in time, $N(t) \sim t^{1/\lambda}$. Hence, the case $\lambda = 0$ is a critical one and the number of fragments grows exponentially in time. Finally, for $\lambda < 0$ the shattering transition takes place: the total volume decreases monotonically and the total number of fragments reaches infinity within an infinitesimally small time interval. Moreover, a finite fraction of the volume breaks into zero-volume rectangles. This phenomenon is well known in the context of one-dimensional fragmentation [10,11] and has been examined in the context of two-dimensional fragmentation with length conservation in a very recent study [19].

IV. ISOTROPIC FRAGMENTATION

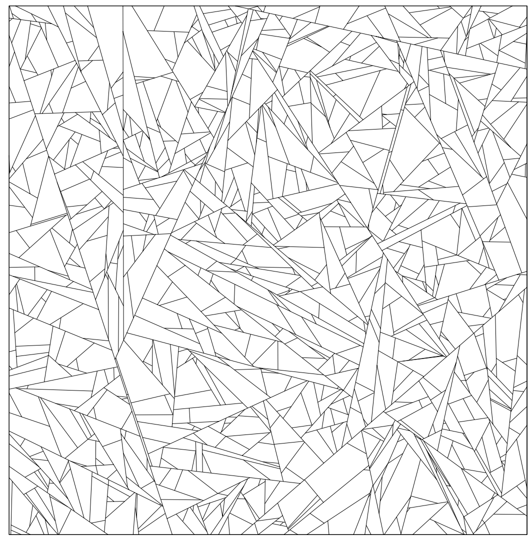

FIG. 3. Realization of the random orientation fragmentation on a unit square at time $t = 1000$.

Intrigued by the rich kinetics of the rectangular fragmentation problem, we also investigated numerically an isotropic fragmentation process. In situations such as shattering of a thin glass plate or in membrane crumpling, the fragments are polygons with a varying number of sides. Hence, we introduce a process where a randomly oriented crack appears with a uniform rate at a random point of the surface and propagates with an infinite speed until it meets an existing crack. The original model can be viewed as deposition of such perfectly oriented "cross" shaped cracks. The overall fragmentation rates in both processes are equal the volume of the fragment. For randomly oriented fragmentation, each fragmentation event creates an additional polygon and hence the total number of polygons grows linearly in time according to $N(t) = M(1,1;t) = 1 + t$. The average volume thus scales as $1/N(t)$ or $A \sim t^{-1}$.

A Monte-Carlo simulation study of isotropic fragmentation process on a unit square with hard boundary conditions have been performed. Numerical results suggest that unlike oriented fragmentation, only one length scale exists in the isotropic problem. The average length of a polygon side is plotted in Fig. 4 and appears to decay as $t^{-1/2}$. Therefore, the length follows the same asymptotic behavior as does the square root of of the average area. A snapshot of a realization of the system at time $t = 1000$ is shown in Fig. 5. This picture suggests that it may prove insightful to investigate various structure properties of the system such as the area distribution function and the side number probabilities of the polygons.

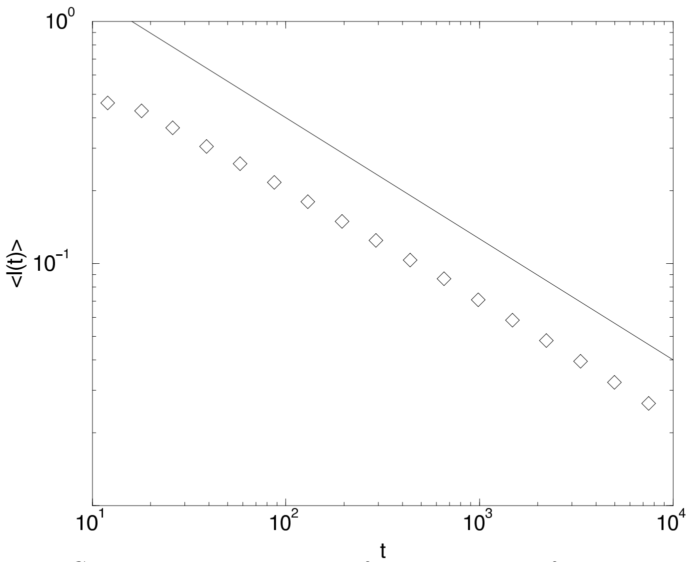

FIG. 4. The average length of a polygon side, for the random orientation fragmentation process. Shown are $\langle l(t)\rangle$ *vs.* t (diamonds) and a line of slope $-1/2$ (solid) for reference.

In conclusion, we have studied two fragmentation processes in spatial dimensions larger than one. For oriented fragmentation, where the fragments are always rectangular, multiscaling is found in the long-time limit. Specifically, the length distribution function has moments that scale algebraically in time with an infinite number of independent length scales, while the area distribution function is characterized by a single length scale. The area distribution function also exhibits a weak logarithmic singularity near the origin. Multiscaling appears to depend strongly on the geometric nature of the process. For isotropic fragmentation, a single length scale describes the decay of the length as well as the area.

After completing of this work we became aware of a closely related study by D. Boyer, G. Tarjus and P. Viot; we thank them for informing us of their results. We also want to thank S. Cornell, E. Kramer, S. Redner, and T. Witten for interesting discussions. We gratefully acknowledge ARO grant #DAAH04-93-G-0021, NSF grant #DMR-9219845, and the Donors of The Petroleum Research Fund, administered by the American Chemical Society, for partial support of this research.

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