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Topological and geometrical properties of random fractals†

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Abstract. The underlying structure of random fractals is described in terms of two exponents, ζ and ν. The exponent ζ is sensitive only to the topology or 'connectedness' of the fractal, whereas ν depends only on the geometry of the fractal. The topological exponent arises from the scaling form for the distribution of paths, n_l, on a finite fractal, where a path is defined as the shortest walk from one site to another. This path length distribution function is then used to derive expressions for the radius of gyration, pair correlation function, static structure factor and intensity of radiation scattered from percolation clusters at the gel point. From these calculations the fractal dimension is shown to be ζ/ν. Finally, recent numerical results for percolation clusters, percolation backbones, and lattice animals are discussed.

Random fractals, such as percolation clusters and diffusion-limited growth aggregates, are currently of interest as models of equilibrium and kinetic growth. To date, however, much of the numerical work on fractals has focused on evaluating the fractal dimension of the mass alone. Unfortunately, it has already been observed that strikingly different objects can have the same fractal dimension; for example, in three dimensions both lattice animals (Isaacson and Lubensky 1980) and linear polymers (Flory 1953) (at the theta temperature) have a fractal dimension of 2. This indicates that the fractal dimension is only a very partial description of the structure of a random object.

In this letter it is shown that random fractals can be more fully described in terms of a topological exponent, ζ, and a geometrical exponent, ν. The topological exponent describes the connectivity of the fractal in terms of a set of minimal walks, and the geometrical exponent relates the contour length of a minimal walk to its root-mean-square extension. Random fractals are shown to emerge in a natural way from a combination of these 'orthogonal' properties, and it is shown that the fractal dimension is just ζ/ν. Also, corrections to scaling for the pair correlation and scattering function are shown to be attributable to a surface term in a topological distribution, n_l, which describes the density of minimal paths on the fractal. Comparison with numerical data for two-dimensional lattice animals, percolation clusters and percolation backbones, all of which have markedly different fractal dimensions, reveals an interesting feature: the variation in fractal dimension is one of topology alone, the geometrical exponent being nearly conserved in these systems. Finally, it is shown that at the critical dimension the geometrical exponent is $\frac{1}{2}$ for these structures, and this is proposed as a universal feature of the critical dimension.

The fractal dimension (Mandelbrot 1977), D, of a particle may be equivalently defined through the relation of the radius of gyration, R, to the mass, M; the dependence

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of the pair correlation function, $g(r)$, on r; or the dependence of the static structure factor, $S(q)$, on the momentum transfer, q. These relations are

$$R^D \sim M, \qquad g(r) \sim 1/r^{d-D} \quad (r \ll R), \qquad S(q) \sim 1/q^D \quad (qR \gg 1),$$

where d is the dimensionality of space. In lattice simulations the radius of gyration and the pair correlation function are the most convenient methods of determining D, but in the laboratory the relation for the structure factor is finding greater use (Schaefer *et al* 1984a, b, c).

The topological exponent, defined by Hong *et al* (1984) can be understood by considering a fractal of N sites on a lattice. Define a 'minimal' path on this lattice fractal to be the shortest topological walk (smallest contour length) from one site to another, where in degenerate cases an arbitrary choice is made for this path. There are then just $N(N-1)$ directed minimal paths (hereafter these are simply called paths) on the fractal. The path length distribution function, n_l, is defined as the number of these paths of length l, and the topological length of the longest path on the fractal (the topological diameter), L, is defined through the normalisation

$$\sum_{1}^{L} n_l = N(N-1). \tag{1}$$

The form of n_l can be surmised by imagining that our finite fractal of N sites (labelled from 1 to N) is embedded in a very large 'embedding' fractal. For example, in a diffusion-limited growth model the N sites could be an early time image of the embedding cluster. Suppose an enumeration of all paths of length l emanating from the ith site, including those which might terminate on the embedding fractal (neglect of 'surface' effects), gives p_i paths. The principal conjecture made here is that the mean number of walks per site, $w = \langle p_i \rangle$, is proportional to $l^{\zeta-1}$, where ζ is called the 'topological' exponent and the average is carried over all sites. In fact, such power-law behaviour has already been demonstrated for percolation clusters, percolation backbones, and lattice animals (see discussion below). Multiplying w by N, the number of sites, gives the first-order approximation, $n_l = ANl^{\zeta-1}$, for the path length distribution function.

This first-order form of n_l neglects surface effects and therefore overcounts paths. It is expected that overcounting becomes important when l becomes comparable to the topological diameter, L. To account for the surface we subtract from the first-order n_l those paths which walk off the cluster. This gives

$$n_l = ANl^{\zeta-1}[1 - b(l/L)] \tag{2}$$

where the function $b(x)$ has obvious limits $b(0) = 0$ and $b(1) = 1$. Scaling arguments presented below demonstrate the correctness of choosing l/L as the argument of $b(x)$.

The geometrical exponent (Pike and Stanley 1981) relates the contour length of a path to the root-mean-square separation of its end points. That is, suppose the ith and jth sites are topologically separated by a path of length l, and geometrically separated by a distance $r_{ij} = r_i - r_j$. The ensemble of sites separated by paths of length l defines the distribution $P_l(r)$, and the mean square length $\langle r_l^2 \rangle = \int r^2 P_l(r)\, \mathrm{d}^d r / \int P_l(r)\, \mathrm{d}^d r$. The geometrical exponent, ν, is then defined by

$$\langle r_l^2 \rangle^{1/2} = a l^{\nu} \tag{3}$$

where a is a constant. Equation (3) does not represent an additional conjecture but, as shown below, is a direct result of the assumption of power-law behaviour in n_l and

the stated purpose of describing fractals. We are now ready to use (1)–(3) to develop expressions for the radius of gyration, the pair correlation function and the scattering function for random fractals.

Lagrange's theorem (Flory 1969) gives the radius of gyration in terms of the site-site vectors r_{ij}:

$$R^2 = (1/2N^2) \sum_i \sum_j \langle r_{ij}^2 \rangle$$

where the averages take into account any finite flexibility of the fractal. Passing to an integral, and making use of the path length distribution function, gives

$$R^2 = (1/2N^2) a^2 \int_0^L n_l l^{2\nu} \, \mathrm{d}l. \tag{4}$$

Converting sums to integrals extends the range of self-similarity to arbitrarily small length scales, and in this way the lattice nature of our approach vanishes.

The pair correlation function can be calculated from a scaling form of the distribution $P_l(r)$. Normalised to unity, this is

$$P_l(r) = P(r/al^\nu)/(al^\nu)^d P_0 \tag{5}$$

where $P_0 = \int P(s) \, \mathrm{d}^d s$. Constraining the moments to be finite requires that $P(x)$ decay faster than a power law. Using $g(r) = N^{-2} \Sigma_i \Sigma_j P_{ij}(r)$ and converting to an integral gives

$$g(r) \sim N^{-2} \int_0^L n_l l^{-\nu d} P(r/al^\nu) \, \mathrm{d}l. \tag{6}$$

The structure factor can be written in terms of $\langle \exp(\mathrm{i}q \cdot r_{ij}) \rangle$, the characteristic function of the distribution $P_{ij}(r)$:

$$S(q) = N^{-2} \sum_i \sum_j \langle \exp(\mathrm{i}q \cdot r_{ij}) \rangle.$$

Taking the Fourier transform of (5) gives

$$S(q) = N^{-2} \int_0^L n_l f(qal^\nu) \, \mathrm{d}l \tag{7}$$

where it is readily shown that f, the characteristic function of $P_l(r)$, depends on qal^ν alone.

Fractals produced by gelation processes are complicated by the large degree of polydispersity in such systems. To interpret scattering data from such polydisperse solutions requires that the scattered intensity of a single cluster be averaged over the distribution of cluster sizes. The scattered intensity from a single particle is just $BN^2 S_N(q)$ where B is a constant which depends on the instrumental geometry, the solvent/fractal contrast factor and the type of radiation scattered. Let $V(N)$ be the number of particles of N sites in the scattering volume. Then $\Sigma \, NV(N) = cv$, where c is the concentration in sites (mass) per unit volume and v is the scattering volume. Define the number distribution $P(N) = V(N)/cv$, normalised by $\Sigma \, NP(N) = 1$. In terms of the weight distribution $W(N) = NP(N)$, the intensity is (Berne and Pecora 1976)

$$I(q) = Bcv \sum_N NW(N) S_N(q).$$

Using (7) gives

$$I(q) = Bcv \int_0^\infty \langle n_l \rangle f(qal^\nu)\, \mathrm{d}l \tag{8}$$

where $\langle n_l \rangle$ is the polydispersity averaged path length distribution function:

$$\langle n_l \rangle = \int_{a' l^\zeta}^\infty n_l(N) P(N)\, \mathrm{d}N \tag{9}$$

and a' is a constant.

For percolation clusters near the gel point, Stauffer (1979) has introduced a scaling form for the number distribution:

$$P(N) = N^{-\tau} h(\varepsilon N^\sigma). \tag{10}$$

Here $\varepsilon = p - p_c$, p is the conversion (fraction of filled sites), p_c is the conversion at the gel point, and σ and τ are exponents which can be related to the standard critical exponents. In this paper, interest is restricted to the gel point, where $P(N) \sim N^{-\tau}$.

It is instructive to derive expressions for the radius, correlation function, and structure factor while neglecting the surface term in (2). From (1) it is observed that L, the longest path length on the cluster, scales with cluster mass like

$$L \sim N^{1/\zeta}. \tag{11}$$

The radius can be evaluated from (4), with the result

$$R^{\zeta/\nu} \sim N. \tag{12}$$

This demonstrates the fractal nature of this formulation and identifies ζ/ν as the fractal dimension, D. The correlation function is obtained for $r \ll R$ by setting the upper limit of (6) to infinity. The result is

$$g(r) \sim 1/Nr^{d-D} \tag{13}$$

where, consistent with (12), $D = \zeta/\nu$.

The structure factor is conveniently divided into two universal regimes: the 'Guinier' regime ($qR \ll 1$) and the 'Porod' regime ($qR \gg 1$). In the Guinier regime the d-dimensional expansion of the structure factor is $S(q) = 1 - q^2R^2/d + \ldots$, independent of the fractal dimension. Furthermore, the intensity per unit concentration at small q is $I/c \sim N(1 - q^2R^2d + \ldots)$ and so is proportional to the particle mass. The form of the structure factor in the Porod regime is more interesting since here it depends directly on the fractal dimension. This can be seen by setting the upper limit of integration in (7) to infinity (the characteristic function decays quickly). This gives

$$S(q) \sim 1/u^D, \qquad I/c \sim 1/q^D, \tag{14a, b}$$

where $u = qR$. That I/c is independent of N, the particle mass, is a simple consequence of mass conservation. But from a scaling point of view it is a consequence of $n_l \sim N$. One can also easily demonstrate that only with $n_l \sim N$ will the scattering exponent be equal to the exponent for the radius—a necessary condition for fractals.

For percolation clusters at the critical point, the intensity is calculated from (8) and (9). At the gel point, the polydispersity averaged distribution of path lengths is

$$\langle n_l \rangle \sim l^{(3-\tau)\zeta - 1} \tag{15}$$

and the intensity is

$$I/c \sim 1/q^{\mu} \tag{16}$$

where $\mu = D(3-\tau)$. Using the exponent relations $D = 1/\sigma\nu$, $3-\tau = \sigma\gamma$ and $\gamma/\nu = 2-\eta$ gives $\mu = 2-\eta$, in agreement with previous results for percolation clusters at the critical point (Stanley 1977, Martin and Akerson 1985).

From these results it can be concluded that an entirely consistent picture of fractals can be developed from a separate consideration of the topological and geometric parts of the problem. Further, the first-order form of n_l is shown to be sufficient to obtain these results. We now demonstrate that the effect of the surface term is to provide corrections to scaling for the structure factor and pair correlation function, while leaving unaltered the essential fractal relations.

The surface term, $-ANl^{\zeta-1}b(l/L)$, is easily understood when applied to topologically one-dimensional random fractals (linear polymers). Without approximation, $n_l = 2N(1-l/L)$ in this case. The first term counts the number of paths of length l which emanate from the N sites of a polymer embedded in an infinite chain. The term $-2Nl/L$ subtracts from this number those paths which do not terminate on the N sites. In agreement with (11), $L = N$ for these fractals. In the more general case where $\zeta > 1$, the functional form of the surface term is not easily computed. The purpose here is twofold: to demonstrate that the function $b(x)$ must depend on l/L alone, and to explore the consequences of this surface term.

With the surface term the left-hand side of (1) becomes

$$NL^{\zeta}\left(\zeta^{-1} - \int_0^1 t^{\zeta-1}b(t)\,\mathrm{d}t\right).$$

The surface term modifies the proportionality constant in $L \sim N^{1/\zeta}$, but does not change the exponent. The effect on the radius is similar: the proportionality constant in $R^{\zeta/\nu} \sim N$ is modified by the integral $\int_0^1 t^{2\nu+\zeta-1}b(t)\,\mathrm{d}t$. Since these prefactors are difficult to measure and interpret, the effect of the surface term on (11) and (12) is inconsequential.

The correlation function and the structure factor are more substantially affected by the surface term. Even without explicit calculation, the dilation symmetry of fractals implies that the surface corrected correlation function must be of the form $g(r) \sim (1/Nr^{d-D})[1-C(r/R)]$, where $C(r/R)$ is a function with the limit $C(0) = 0$ (a more usual, but equivalent, scaling form is $(1/Nr^{d-D})C(r/R)$ where $C(0) = 1$). Indeed, this result can be shown directly from (2) and (6):

$$\begin{aligned} g(r) &\sim (1/Nr^{d-D})[1-C(r/R)], \\ C(x) &\sim \int t^{\zeta-\nu d-1}P(t^{-\nu})b(tx^{1/\nu})\,\mathrm{d}t. \end{aligned} \tag{17}$$

That $C(x)$ is a function of r/R alone is a consequence of choosing l/L for the argument of $b(x)$. This demonstrates the correctness of the scaling form of n_l.

Similar arguments apply to the structure factor. Here the expected scaling form is $S(q) \sim u^{-D}[1-C'(u)]$, where again $C'(0) = 0$. From (2) and (7)

$$\begin{aligned} S(q) &\sim u^{-D}[1-C'(u)], \\ C'(u) &\sim \int t^{\zeta-1}f(t^{\nu})b(t/u^{1/\nu})\,\mathrm{d}t. \end{aligned} \tag{18}$$

A 'correction to scaling' for the correlation function and the structure factor can be computed from the small-l behaviour of the surface term. Define the exponent λ by the small-x limiting behaviour $b(x) \sim x^{\lambda}$. Since $f(t)$ vanishes quickly for large t, the small-r (large-u) form of the correlation function (structure factor) is

$$g(r) \sim (1/Nr^{d-D})[1-(r/R)^{\lambda/\nu}+\ldots], \qquad S(q) \sim u^{-D}(1-u^{-\lambda/\nu}+\ldots). \tag{19}$$

So the correction to scaling is related to the geometrical exponent and a new exponent which characterises the small-l behaviour of the surface term.

Finally, consider the effect of the surface term in polydisperse systems at the gel point. At the gel point the correlation range becomes infinite and a very general scaling analysis shows that $I/Bcv = \text{constant} \times q^{-\mu}$ exactly (no correction to scaling). But from (18) it is clear that the surface term does contribute to the scattering from any given constant-N fraction of clusters. We must conclude that if our formulation is correct, a polydispersity average over the surface term must contribute $O(q^{-\mu})$ to I/C.

From (9) it can be seen that

$$\langle n_l \rangle = l^{(3-\tau)\zeta-1}\left(\text{constant}' - \text{constant}'' \int t^{1-\tau} b(1/t)\, dt\right) \sim l^{(3-\tau)\zeta-1}.$$

This leads directly to the physically necessary result $I/Bcv = \text{constant} \times q^{-\mu}$ exactly. The effect of polydispersity on the surface term is thus to transform the correction to scaling $(-q^{-D}C(qR))$ into $-q^{-\mu}$. Again, this can be traced to the requirement that $b(x)$ be a function of l/L alone.

We have now established that it is possible to develop a theory of the underlying structure of random fractals based on the assumption of separable topological and geometrical exponents. Although it is not clear that all fractal systems need conform to this decomposition, Stanley and coworkers (Hong *et al* 1984, Majid *et al* 1984, Havlin *et al* 1984) have collected substantial numerical data that demonstrate this behaviour for lattice animals ($d = 1, 2, 3, 4, 8$), percolation clusters ($d = 2$) and percolation backbones ($d = 2$). But before discussing the available data, it is well to consider the range of the exponents. The geometrical exponent has the obvious d-independent upper bound $\nu = 1$. The lower bound is given by a dense packing constraint (ball of string) and gives $\nu = 1/d$. The lower bound on ζ is 1 (obtained for topologically one-dimensional structures), and the upper bound is d (d-dimensional balls). In summary,

$$1 < \zeta < d \qquad 1 < d\nu < d.$$

A few trivial examples are the d-dimensional ball ($\nu = 1, \zeta = d$) and linear polymers ($\zeta = 1$, and $\nu \simeq 3/(d+2)$ or $2/(d+1)$ for good or theta solvents).

What about more interesting fractals? In what cases can power law behaviour be found for the distribution of path lengths? In a paper on random walks on $D = 2$ percolation backbones, Hong *et al* (1984) described an exponent that measures the number of sites within a 'chemical' distance l. This exponent, which appears in a natural way in considering transport on fractals, is just the exponent ζ defined here. Using an exact enumeration algorithm for the walks, Hong *et al* found $\zeta = 1.44$ and $D = 1.66$. This gives $\nu = 0.87$, which satisfies the requirement $0.5 < \nu < 1$.

In another paper using the exact enumeration method, this time on $d = 2$ percolation clusters at criticality, Majid *et al* (1984) found $\zeta = 1.63$. Using $D = 1.89$ gives $\nu = 0.86$, essentially the result for percolation backbones.

Havlin *et al* (1984) studied random walks on lattice animals and found $\zeta = 1$, 1.33, 1.47, 1.61, 2 and $D = 1$, 1.56, 2, 2.4, 4 in $d = 1$, 2, 3, 4, 8. From these data ν is seen to decrease with dimension. In order of increasing d, $\nu = 1$, 0.85, 0.74, 0.67, $\frac{1}{2}$. Again, in $d = 2$, ν is very similar to the percolation results. The difference between percolation clusters, percolation backbones and lattice animals seems to be primarily one of topological structure.

Flory theory can be used to obtain some insight into the large dimensionality behaviour of the topological exponent ζ. In Flory theory the free energy is expressed as $F/kT = (R/R_0)^2 + vN^2R^{-d} + wN^3R^{-2d} + \ldots$ where v and w are the binary and ternary cluster integrals, and R_0 is the unperturbed radius. If $v > 0$, minimisation of F gives $D = (d_c/2)(d+2)/(d_c+2)$, where d_c is the critical dimension. Minimisation for $v = 0$ gives $D = (2d_c/3)(d+1)/(d_c+1)$. At the critical dimension, excluded volume effects vanish, and the geometrical exponent assumes its random walk value of $\frac{1}{2}$. For $v > 0$ this gives $\zeta = d_c/4$ for the asymptotic value of the topological exponent. For example, the critical dimension of linear polymers in good solvents is 4, giving the obvious result $\zeta = 1$. For branched polymers (Isaacson and Lubensky (1980) applied Flory analysis to this problem) the critical dimension is 8 and $\zeta = 2$ for $d \geqslant 8$. In the 'theta' case where $v = 0$, $\zeta = d_c/3$. Applied to theta polymers ($d_c = 3$), this gives $\zeta = 1$. For branched polymers in theta solvents $d_c = 6$, giving $\zeta = 2$.

From this simple analysis it is clear that the exponents ζ and ν provide greater insight into fractal geometry than the fractal dimension alone. As a final example of this, Witten-Sanders (1981) diffusion-limited growth aggregates (DLAS in $d = 3$ are known to have $D = 2.5$, which is the result for percolation clusters at the critical point. Does this mean that DLAS have the same statistical geometry as percolation clusters? Probably not. DLAS have the very obvious feature that they are essentially devoid of 'lakes' (vacant sites bounded by the fractal); percolation clusters, on the other hand, exhibit a plethora of lakes. This difference may be reflected in the exponents ν and ζ.

We have shown how a theory of fractals can be developed by a consideration of exponents and distributions associated with the 'orthogonal' topological and geometrical properties of random fractals. In terms of these exponents, respectively ζ and ν, the fractal dimension is shown to be ζ/ν. Results for the radius of gyration, pair correlation function, static structure factor and intensity at the gel point are shown to be in agreement with general scaling results for fractals. Finally, numerical data for percolation clusters ($d = 2$), percolation backbones ($d = 2$) and lattice animals ($d = 1, 2, 3, 4, 8$) amply demonstrate power-law behaviour for the topological property and verify the treatment given here.

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