# Statistical Analysis of Anomalous Transport Phenomena in Complex Media

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Transport phenomena on complex media are characterized by a subdiffusive regime called anomalous diffusion (Sahimi, 1983; Rammal, 1984). Anomalous diffusion has been observed in porous media (Katz and Thompson, 1985), capillarity networks (Adler, 1985) and percolation beds (Stauffer, 1985). In the last two years a great deal of research work has been carried out on this topic in the chemical engineering field (Sheintuch and Brandon, 1989; Tassopoulos et al., 1989; Sahimi et al., 1990; Siddiqui and Sahimi, 1990). The main features of anomalous diffusion are:

- Deviation from Gaussianity
- Anomalous scaling law of the mean square displacement $\langle r^2(t)\rangle$ vs. $t$.

$$\langle r^2(t)\rangle = \mathcal{K}t^{\beta},\ t\rightarrow\infty \tag{1}$$

with $\beta<1$ (subdiffusive regime). For regular diffusion we have $\beta=1$.

This relation can be explained in terms of the geometrical constraints imposed on the motion of the diffusing particles by the self-similar symmetry (fractality) of the medium. In fact, porous media and percolation clusters can be considered in a certain lengthscale range as fractal (Havlin, 1989). The concepts of fractal geometry can be therefore applied to transport phenomena on these structures. In this theoretical framework it is possible to show that the exponent $\beta$ is given by (Alexander and Orbach, 1982; Havlin, 1989):

$$\beta = D_s/D \tag{2}$$

where $D$ and $D_s$ are respectively the fractal dimension and the fracton (spectral) dimension of the medium.

While for a scaling theory of anomalous diffusion knowledge is sufficient of the three exponents (dimensions) $D$, $D_s$ and $d$, the dimension of the Euclidean space in which the medium is embedded. It is of great importance in chemical engineering science to obtain a more detailed picture of anomalous transport phenomena with a view to the formulation of a material balance equation. The great difficulty in writing down macroscopic Euclidean approximations for transport equations in complex fractal media lies in the intrinsic geometrical dishomogeneity of the medium, which is a consequence of its scaling structure (Sahimi et al., 1990). Moreover, for a detailed comparison between the solution of approximate macroscopic equations and experimental or computer simulation results, the scaling relation (Eq. 1) is not sufficient. A more general characterization is needed.

Keeping in mind that the solution of a diffusion process can be considered as a probability density function (Lasota and Mackey, 1987), we can make use of the classical methods of statistical analysis of random processes to obtain information on transport phenomena. This work presents an analysis based on the moment generating function of anomalous diffusion. This analysis is applied to some macroscopic approximations. It is shown how the spatio-temporal functional dependencies of the solution of the diffusion problem can be evaluated directly from the scaling of the moment generating function and of the generalized moments.

The practical application of the results obtained to computer simulation experiments is discussed, and the method is applied to some random walk simulations on fractals and percolation clusters.

## Moment Generating Function

To achieve a deeper knowledge of the spatio-temporal dependencies of the solution of a diffusion problem on a complex medium, it is necessary to introduce the moment-generating function (MGF) $C(q,t)$ (Seinfeld and Lapidus, 1974):

$$C(q,t) = \langle \exp(qx(t))\rangle = \lim_{N\rightarrow\infty}\frac{1}{N}\sum_{i=1}^{N}\exp\{q[x(t_i)-x(t_i-t)]\} \tag{3}$$

where $q$ is a real parameter. In Eq. 3 the brackets $\langle\ \rangle$ indicate the time average along the trajectories of the particles in the medium.

To this mechanico-statistical picture, which is well suited for computer simulation using Monte Carlo methods, there corresponds the macroscopic definition of $C(q,t)$. If $p(x,t)$ is the solution of the transport equation, then $C(q,t)$ is the restriction of the Laplace transform $\hat{p}(s,t)$ to the real line:

$$C(q,t) = \int e^{qx} p(x,t)dx = \hat{p}(-q,t). \quad (4)$$

The two definitions (Eqs. 3 and 4) are equivalent since time averages coincide with ensemble averages [with respect to the distribution $p(x,t)$].

Fujisaka used the MGF to study the deviation from Gaussian behavior in diffusional motion driven by chaotic deterministic maps (Fujisaka, 1984, 1985). For this purpose he introduced the generalized exponents $\lambda(q)$:

$$\lambda(q) = \lim_{t \to \infty} \frac{\log C(q,t)}{qt}. \quad (5)$$

This definition is well formulated if the limit in Eq. 5 exists and is not identical to zero for all the $q$. This is the case of regular (Gaussian) diffusion, where:

$$\lambda(q) = \mathfrak{D} q.$$

Definition of Eq. 5 is still useful for a great number of diffusional models driven by deterministic chaotic maps, for which the limit behavior ($t \to \infty$) of the mean square displacement is linear in time (Fujisaka, 1984; Geisel and Nierwetberg, 1982). Nevertheless, Eq. 5 fails to be valid in the case of anomalous diffusion. As an example, for fractional Brownian motion of index $\beta/2$ (Mandelbrot, 1982; Falconer, 1990), relation (1) holds with $0 < \beta < 2$ and:

$$C(q,t) \sim \exp(q^2 t^\beta).$$

The limit in Eq. 5, therefore, is identically equal to zero in the subdiffusive cases ($\beta < 1$) and cannot be defined for $\beta > 1$. For this reason we prefer to consider the logarithm of the MGF $L(q,t)$:

$$L(q,t) = \log C(q,t) \quad (6)$$

In the next section we show that $L(q,t)$ in anomalous diffusion scales, for $q \to \infty$, as:

$$\log L(q,t) = \gamma_q \log q + \gamma_t \log t + \log L_o. \quad (7)$$

In the same way, we can define the generalized moments $M(\bar{q},t)$:

$$M(\bar{q},t) = \langle x(t)^{\bar{q}} \rangle = \int x^{\bar{q}} p(x,t)dx \quad (8)$$

where $\bar{q}$ is a real parameter. For $\bar{q} = n =$ integer, $M(n,t)$ can be expressed in terms of the derivative of $C(q,t)$ (Seinfeld and Lapidus, 1974). The functional form of $M(\bar{q},t)$ in anomalous diffusion is given by:

$$\log M(\bar{q},t) = \mu \bar{q} \log t + \log M_o(\bar{q}). \quad (9)$$

As shown in the next section, the exponents $\gamma_q$, $\gamma_t$ and $\mu$ are related to the spatio-temporal scaling of $p(x,t)$. In fact, these exponents can be evaluated from computer simulation of transport phenomena.

## Statistical Analysis of Anomalous Diffusion

Relations 7 and 9 can be obtained from mean field approximation of the transport equation in the case of anomalous diffusion. This derivation will provide a set of equations relating the measurable quantities $\gamma_q$, $\gamma_t$, $\mu$ to the spatio-temporal functional dependencies of $p(x,t)$. As a result of the interest in chemical engineering in transport phenomena on radial symmetric structures (Sheintuch and Brandon, 1989) we consider here transport phenomena on statistical radial symmetric media (SRSM). An example of this kind of media is given by diffusion-limited aggregation clusters (Meakin, 1987). This entails replacing $x$ with the radial coordinate $r$. Furthermore, we replace $p(x,t)$ with $p(r,t)$, where $p(r,t)$ satisfies the condition:

$$\int_0^\infty p(r,t)dr = \text{constant}.$$

Up to now the more general way of writing macroscopic Euclidean approximations for transport phenomena in SRSM media has been given by (O'Shaugnessy and Procaccia, 1985):

$$p(r,t) = r^\kappa w(r,t) \quad (10)$$

$$\frac{\partial w(r,t)}{\partial t} = \frac{\mathfrak{D}_o}{r^\delta} \frac{\partial}{\partial r}\left( r^\nu \frac{\partial w(r,t)}{\partial r} \right) \quad (11)$$

where $\kappa$ is the space-scaling exponent and $w(r,t)$ is the mass concentration function.

This equation is a generalization of radial symmetric diffusion equation in Euclidean spaces (Cranck, 1975) and is derived from the fact that the diffusion coefficient $\mathfrak{D}$ in SRSM that have fractal properties is a function of $r$ (Orbach, 1986):

$$\mathfrak{D}(r) = \mathfrak{D}_o r^{-\theta} \quad (12)$$

where:

$$\theta = 2(D/D_s - 1). \quad (13)$$

The constant $\mathfrak{D}_o$ does not have the dimension of a diffusion coefficient and will be called the diffusion prefactor.

It should be pointed out that Eqs. 12 and 13 come from the usual definition of the diffusion coefficient $\langle r^2(t) \rangle = 2\mathfrak{D}t$, where $\mathfrak{D} = \mathfrak{D}(r)$ is defined in such a way that the fundamental scaling equation (Eq. 1) is fulfilled (Orbach, 1986). Therefore, the diffusion coefficient introduced by Eq. 12 has the meaning of an "equivalent diffusion coefficient" derived from scaling analysis in the case of anomalous media. According to this observation the diffusion prefactor $\mathfrak{D}_o$ does not possess, as in the regular Brownian case ($\theta = 0$), a direct microscopic meaning.

The exponents $\kappa$, $\delta$ and $\nu$ depend on the physical structure of the medium exhibiting anomalous behavior. If we assume Eq. 12 and insert it into the standard diffusion equation we have: $\kappa = d-1$, $\delta = d-1$, $\nu = d-1-\theta$. This corresponds to assuming that the fractal dimension of the medium is equal to the Euclidean dimension ($D = d$) but $D_s < D$. Otherwise, if we consider that the number of accessible sites of the medium $n(r)$ scales as $n(r) \sim r^D$ (fractal medium) and therefore the volume element of the medium is given by $dV(r) = \Lambda r^{D-1} dr$

(O'Shaugnessy and Procaccia, 1985; Vicsek, 1989) we have: $\kappa=D-1$, $\delta=D-1$, $\nu=D-1-\theta$.

The solution of Eq. 11, under the hypothesis that the medium can be considered infinitely extended $r\in[0,\infty)$, for an initial impulsive distribution ($p(r,0)=w_o\delta_D(r)$; $w(0,t)=0$ $t>0$) is given by:

$$w(r,t)=At^{-\eta}\exp(-r^{\xi}/Bt^{\zeta}) \tag{14}$$

where $A=w_o\xi/(\Lambda B^{(\kappa+1)/\xi}\Gamma((\kappa+1)/\xi))$, $\Gamma$ is the gamma function and:

$$\begin{aligned}\xi&=\delta-\nu+2=2+\theta\\ \eta&=(\nu+\xi-1)/\xi=D_s/2\\ \zeta&=1\\ B&=\mathfrak{D}_o\xi^2.\end{aligned} \tag{15}$$

The value of the exponents $\zeta$ in the solution of Eq. 11 is independent of the scaling exponents and identically equal to 1.

Nevertheless, some authors have found in the analysis of diffusion on percolation clusters and on deterministic fractals (Guyer, 1985; Sahimi, 1987) a functional form for $p(r,t)$ equal to Eqs. 10 and 14 with $\eta=D_s/2$, $\kappa=D-1$, but with the values of $\xi$ and $\zeta$ given by:

$$\xi=2/(2-\beta),\ \zeta=\beta/(2-\beta). \tag{16}$$

Note that assuming the expression 16 for $\xi$ and $\zeta$ the constant $B$ is somewhat related to the prefactor $\mathfrak{D}_o$ defined in Eq. 12 in a nonlinear way $B\sim\mathfrak{D}_o^{\beta/(2-\beta)}$.

To consider the two main models, expressed by Eqs. 15 and 16, we derive the scaling properties of the statistical functions related to the diffusion process from the general accepted expression 14 (Havlin and Ben-Avraham, 1987) in terms of the constants $\kappa,\eta,\xi$, and $\zeta$.

The MGF for SRSM is given by Eqs. 4 and 10:

$$C(q,t)=A\Lambda t^{-\eta}\int_0^{\infty}r^{\kappa}\exp(qr-r^{\xi}/Bt^{\zeta})dr. \tag{17}$$

Making use of the steepest descend method we obtain the following asymptotic expansion for $q\rightarrow\infty$:

$$C(q,t)\sim\exp\left[\frac{(\xi-1)}{B}\left(\frac{Bq}{\xi}\right)^{\xi/(\xi-1)}t^{\zeta/(\xi-1)}\right] \tag{18}$$

Hence,

$$\gamma_q=\xi/(\xi-1),\ \gamma_t=\zeta/(\xi-1). \tag{19}$$

From Eqs. 7 and 18, it is possible to obtain the value of the constant $B$ entering in Eq. 14:

$$\log\left[L_o/B^{1/(\xi-1)}\right]=\log(\xi-1)-\xi\log\xi/(\xi-1) \tag{20}$$

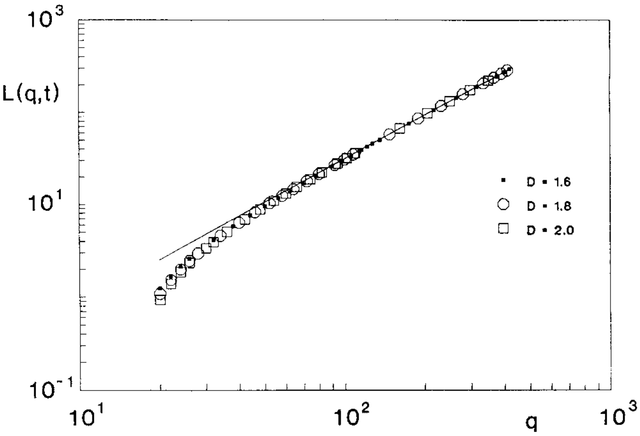


**Figure 1. Log-log plot of $L(q,t)$ vs. $q$ for fixed $t=0.1$.**

The values of transport equation exponents are chosen according to the O'Shaugnessy-Procaccia model: $\kappa=\delta=D-1$, $\nu=D-1.8$ for $D=1.6$, 1.8, 2. The straight line indicates the limit behavior according to (17). The slope is equal to $\gamma_q$ and the value at $q=1$ allows us to compute $L_o$

which, for the O'Shaugnessy-Procaccia model, Eq. 15, becomes

$$(\xi-1)\log\left[L_o/\mathfrak{D}_o^{1/(\xi-1)}\right]=(\xi-1)\log(\xi-1)-(\xi-2)\log\xi. \tag{21}$$

In both cases, Eqs. 20 and 21, the argument of the logarithm in the first term is adimensional. Therefore, the evaluation of $L_o$ makes it possible to estimate the constant $B$ or $\mathfrak{D}_o$ (if the O'Shaugnessy-Procaccia model is assumed as valid).

As an example of application of these results we have numerically computed $C(q,t)$ from Eq. 17, with the values of the constants $D=1.6$, 1.8, 2.0, $\xi=2.8$, $\zeta=1$, $\mathfrak{D}_o=0.01$, according to the O'Shaugnessy-Procaccia model. These values are physically admissible for a fractal structure embedded in a plane ($d=2$). The results for the scaling of $L(q,t)$ defined by Eq. 6 are shown in Figs. 1 and 2 from which one obtains Eq. 7, $\gamma_q=1.563$, $\gamma_t=0.556$, and $L_o=0.085$. As can be seen the values of the asymptotic scaling exponents $\gamma_q$, $\gamma_t$ are in-

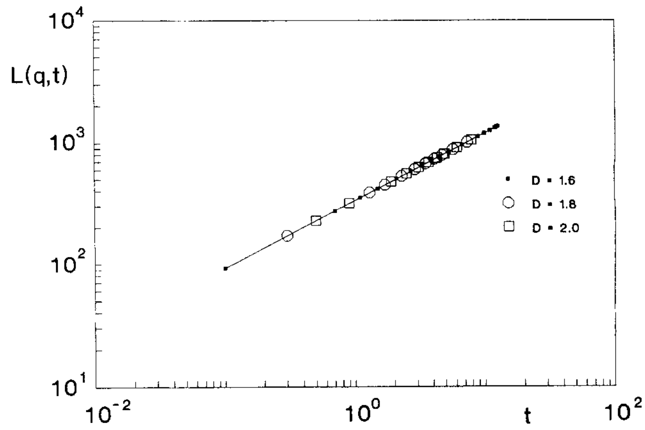


**Figure 2. Log-log plot of $L(q,t)$ vs. $t$ for fixed $q=200$.**

The values of the scaling exponents are the same as in Figure 1. The slope is equal to $\gamma_t$.

dependent of $D$ (Eq. 18). According to the asymptotic expression (18) the constitutive parameters $\xi$, $\zeta$, $\mathfrak{D}_o$ can be estimated from the asymptotic regime of $L(q,t)$. The application of Eqs. 19 and 20 to the numerical data gives $\xi = 2.78$, $\zeta = 0.99$, $\mathfrak{D}_o = 0.011$, which are in good agreement with the assumed values in the integration of Eq. 17. This result confirms the asymptotic expansion (Eq. 18). The information obtained from the asymptotic form of $C(q,t)$ can be completed with that derived from the analysis of $M(\bar{q},t)$, (Havlin and Ben-Avraham, 1987). From the definition, in the case of SRSM, we have:

$$\log M(\bar{q},t) = \left[\frac{\zeta(\bar{q}+1+\kappa)}{\xi} - \eta\right]\log t + \log M_o(\bar{q}). \quad (22)$$

From the conservation of mass (or equivalently, if we attribute a stochastic meaning to transport phenomena, from the conservation of probability) we obtain, putting $\bar{q}=0$, a relation linking $\kappa$ to $\eta$:

$$\zeta(1+\kappa)/\xi - \eta = 0$$

that can also be expressed in terms of $\beta$:

$$\beta(1+\kappa) - 2\eta = 0. \quad (23)$$

In both of the considered cases (Eqs. 15 and 16), condition 23 is obviously fulfilled. The exponent $\mu$ therefore reads:

$$\mu = \zeta/\xi \quad (24)$$

and is equal to $\mu = \beta/2$.

The analysis of $M(\bar{q},t)$ also furnishes a relation for the exponent $\kappa$ and the constant $B$: if we assume that the radial distribution function of the anomalous diffusion $p(r,t)$ is normalized to one, then $\kappa$ is related to the prefactor $\mathcal{K}$ via:

$$\mathcal{K} = \frac{\Gamma((\kappa+3)/\xi)}{\Gamma((\kappa+1)/\xi)} B^{2/\xi}. \quad (25)$$

It is to be noted that $\mathcal{K}$ in anomalous diffusion is not proportional to $B$ as in the Euclidean case ($2/\xi = 1$, $B \sim \mathfrak{D}$). This is not surprising since it is a consequence of the nonlinearity of $\langle r^2(t)\rangle$ with respect to time $t$, Eq. 1. Moreover, if we assume the O'Shaugnessy-Procaccia model, then $\mathcal{K} \sim \mathfrak{D}_o^{\beta}$.

We would like to direct a final comment to the practical use of the presented results. As noted by Sahimi, whenever the medium "has fractal structure continuum equations of diffusion and reaction do not hold" (Sahimi et al., 1990). We can, from an engineering point of view, look for approximate Euclidean equations.

The model (Eqs. 10 and 11) proposed by O'Shaugnessy and Procaccia and that of Guyer (Eqs. 14 and 16), are the basic examples of macroscopic models for diffusion in disordered and fractal media. The only possible ways to understand the validity of the various models are either to make use of a renormalization group approach, or to compare it with experimental and computer simulation data. Unfortunately the renormalization group approach can be successfully applied only to few simple cases and therefore computer simulation methods represent the standard and more appropriate way for a statistical analysis of transport phenomena in complex structures. The evaluation of the function $p(r,t)$ from computer experiments is quite cumbersome, while a numerical scaling analysis of $C(q,t)$ and $M(\bar{q},t)$ can be more easily performed.

## Simulation Results

The practical application of the developments presented in this work to random walk simulation experiments requires great care. The main difficulty in the direct application of Eqs. 18 and 19 lies in the extremely low convergence of $\langle e^{qr(t)}\rangle$. If for a fixed $\bar{t}$, $r_M$ is the maximal possible distance traveled by a random walking particle, the number $N$ of data points along particle trajectories necessary to obtain a discrete experimental convergence of $C(q,\bar{t})$ is about $N \sim 1/p(r_M,\bar{t}) \sim \exp(r_M^{\xi}/Bt^{\zeta})$. As an example in the Euclidean case for $t = 400$, ($r_M = 400$ lattice units) assuming $B = 1$ one obtains $N \simeq 5\ 10^{173}$. For this reason the usual way of obtaining information on the structure of $p(r,t)$ is to make use of exact enumeration techniques (Stauffer, 1985; Havlin and Ben-Avraham, 1987).

However, this raises the question of developing a method based on the analysis previously outlined which makes it possible to achieve a good evaluation of the scaling exponents entering into the expression of $p(r,t)$ directly from random walk experiments and processing a relatively small number of data points along particle trajectories, $N \simeq 10^4 \div 10^6$. A procedure for overcoming the numerical difficulties outlined above can be based on the following considerations. By considering two different media—a Euclidean space and a percolation cluster, let $L_1(q,t)$ and $L_2(q,t)$ be the logarithm of the MGF in the two cases. If we consider the ratio $R_{21}(q,t)$:

$$R_{21}(q,t) = L_2(q,t)/L_1(q,t) \quad (26)$$

where $L_1(q,t)$ *and* $L_2(q,t)$ are both evaluated from random walk simulations, from Eq. 7 we have:

$$\log R_{21}(q,t) = (\gamma_{q2}-\gamma_{q1})\log q + (\gamma_{t2}-\gamma_{t1})\log t + \log R_o \quad (27)$$

$\gamma_{qi}$, $\gamma_{ti}$ ($i = 1,2$) being the exponents of the two media entering into Eqs. 7 and 19. If for the first medium the scaling exponents are known from theoretical considerations ($\gamma_{q1} = 2$, $\gamma_{t1} = 1$, in the Euclidean case), the values for the scaling exponents of the second medium can be predicted directly from the scaling (Eq. 27) of $R_{21}(q,t)$. In general, the first medium can be chosen as the Euclidean space in which the disordered medium is embedded.

This technique has the advantage that $R_{21}(q,t)$ is a more robust function than $L_1(q,t)$, because the errors in the evaluation of $L_2(q,t)$ and $L_1(q,t)$ are of the same order of magnitude. It is expected that, taking the ratio, they will compensate each other.

Simulation experiments were performed on diffusion of percolation clusters in $400 \times 400$ lattice for a value of the percolation probability $p_p = 0.65$. In this case it was found that diffusion is anomalous with a value of the exponent $\beta = 0.85 \pm 0.01$, Fig. 3.

Denoting with $R_{21}(q,t)$ the ratio between $L_2(q,t)$ on the percolation cluster and $L_1(q,t)$ on a corresponding $400 \times 400$ Euclidean square lattice, we found by applying Eq. 27

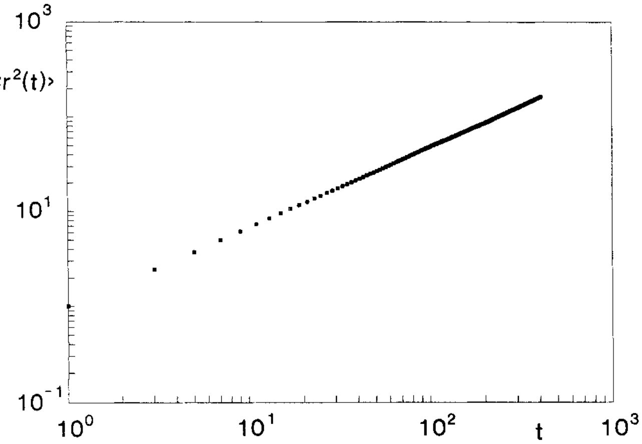


**Figure 3. $\langle r^2(t)\rangle$ vs. $t$ for a two-dimensional percolation cluster on a 400 × 400 square lattice for a percolation probability $p_p = 0.65$.**

The number of averaged data points were $N = 100{,}000$.

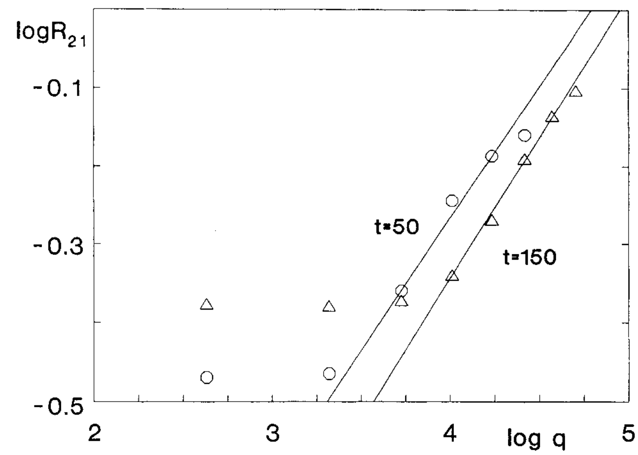


**Figure 4. Behavior of $R_{12}(q,t)$ vs. $q$ for different instants of time.**

The first medium is Euclidean 400 × 400 square lattice, the second is the percolation cluster from which the data of Figure 3 have been obtained. The number of data points used in the evaluation of $R_{21}(q,t)$ were $N = 500{,}000$.

$(\gamma_{q2} - \gamma_{q1}) = 0.3 \div 0.4$, (Fig. 4) and therefore $\xi_2 = \gamma_{q2}/(\gamma_{q2} - 1) = 1.71 \div 1.77$, which is in close agreement with the theoretical prediction (Eq. 16) compared with the value found for $\beta\ \xi_2 = 2/(2\beta - 1) = 1.72 \div 1.75$ (Guyer, 1985; Sahimi, 1987).

Moreover, by using the same techniques and off-lattice IFS-diffusion simulation (Giona and Giona, 1990) on deterministic fractal structures, we found for the two-dimensional Sierpinski gasket, for $N = 60{,}000$ averaged data points, a value of $\gamma_q = 2.10 \div 0.05$ which implies $\xi \simeq 1.90$. This result is in agreement with the result reported by Havlin and Ben-Avraham (1987).


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## Notation

$A, A'$ = constants
$B$ = parameter of the diffusion distribution function
$C$ = moment-generating function
$d$ = dimension of the Euclidean space
$D$ = fractal dimension
$D_s$ = fracton dimension
$\mathfrak{D}$ = diffusion coefficient
$\mathfrak{D}_o$ = diffusion prefactor
$dV$ = volume element
$\mathcal{K}$ = prefactor of the $\langle r^2\rangle$scaling law
$L$ = logarithm of the MGF
$L_o$ = prefactor of $L$
$M$ = generalized moments
$M_o$ = prefactor of $M$
$N$ = number of data points in the simulations
$p$ = distribution function associated with the diffusion
$\hat{p}$ = Laplace transform of $p$
$p_p$ = percolation probability
$q, \overline{q}$ = real parameters
$r$ = radial coordinate
$\langle r^2\rangle$ = mean square displacement
$r_M$ = maximal distance traveled by a random walker
$R_{12}$ = ratio of the logarithms of $C(q,t)$ for different media
$t$ = time
$\bar{t}, \bar{t}_i$ = instants of time
$w$ = mass concentration function
$w_o$ = constant
$x$ = spatial coordinate

### Greek letters

$\beta$ = scaling exponent of the mean square displacement
$\gamma_q, \gamma_t$ = scaling exponents of the logarithm of MGF
$\Gamma(x)$ = Gamma function of argument $x$
$\delta$ = exponent in the transport equation
$\delta_D$ = Dirac's delta-distribution
$\zeta$ = exponent in the diffusion distribution function
$\eta$ = exponent in the diffusion distribution function
$\theta$ = scaling exponent of the diffusion coefficient
$\kappa$ = space-scaling exponent
$\lambda$ = generalized exponents
$\Lambda$ = prefactor of the volume element
$\mu$ = scaling exponent of the generalized moments
$\nu$ = exponent in the transport equation
$\xi$ = exponent in the diffusion distribution function

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