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Diffusion on two-dimensional percolation clusters with multifractal jump probabilities

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By means of Monte Carlo simulations we studied the properties of diffusion limited recombination reactions (DLRR's) and random walks on two dimensional incipient percolation clusters with multifractal jump probabilities. We claim that, for these kind of geometric and energetic heterogeneous substrata, the long time behavior of the particle density in a DLRR is determined by a random walk exponent. It is also suggested that the exploration of a random walk is compact. It is considered a general case of intersection in d euclidean dimension of a random fractal of dimension D_F and a multifractal distribution of probabilities of dimensions D_q (q real), where the two dimensional incipient percolation clusters with multifractal jump probabilities are particular examples. We argue that the object formed by this intersection is a multifractal of dimensions $D'_q = D_q + D_F - d$, for a finite interval of q.

I. Introduction

Recently, fractals and multifractals [1a] which appear in many physical applications have attracted a growing interest (Refs. 1–5 and references cited therein). For example, it is known [6 and references therein] that the surface of most solids at the molecular scale is fractal. Consequently, many physico-chemical properties and processes related to such systems have to be carefully examinated. In this context, diffusion and reaction studies on disordered systems have recently been reported [7–12]. Major interest arises because a random walk on fractal media exhibits anomalous long-time behavior which implies that the reaction order of a diffusion limited recombination reaction (henceforth DLRR) is also anomalous in a low concentration regime [9–12] and this fact has been experimentally verified [9, 11].

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On the other hand, the study of the properties of random walks on square lattices with multifractal distribution of jumping probabilities [13], which constitutes an interesting open problem, has recently been initiated.

The purpose of this work is to study, by means of the Monte Carlo simulations, the behavior of random walkers and DLRR's on two dimensional percolation clusters with multifractal jump probabilities. These substrata can be considered as the "intersection" of two dimensional incipient percolation clusters (i.e. geometric heterogeneous objects) and planar multifractal distributions of jumping probabilities (i.e. energetic heterogeneous objects), where the concept of "intersection" has to be interpreted in a restricted manner as it is discussed in Sect. II. As it is shown in Sect. III, these substrata are a new kind of multifractals, called percolation multifractals (PM), which have a non trivial spatial fractal dimension. The PM's combine both geometric and energetic heterogeneities which are strongly complex and to the best of our knowledge, this is the first model introduced in order to study the diffusion on substrata with these properties. Explicitly, our study is mainly focused on the computation of random walk exponents and the exponent related to the time behaviour of the particle density in a DLRR and to analyse the relation between them. Our hope is that this investigation could help in the comprehension of complex recombination reactions which occur on fractal catalysts with geometric and energetic heterogeneities.

It is also presented a conjecture about the set of dimensions of the objects formed by the intersection of a random geometric fractal and a multifractal distribution of probabilities.

The paper is organized as follows: In Sect. II it is defined the PM model. In Sect. III the theoretical arguments to obtain the conjecture about the intersection of fractals and multifractals are discussed. In Sect. IV it is presented the theory about random walkers and DLRR's on multifractal structures. In Sect. V the Monte Carlo simulation is described. Finally in Sect. VI the discussion of the results and the conclusion are stated.

II. The PM model

Let us start with the definition of planar multifractals which have recently been proposed by Meakin [1e, 13] and will be used in this work. The multifractal distribution of probabilities on square lattices of size $L \times L$, with $L = 2^n$, are constructed as follows. Four normalized probabilities P_i ($i = 1, \ldots, 4$), are selected. In the first step, these probabilities are randomly assigned to the four quadrants of linear size $l_1 = L/2$, of the lattice (see Fig. 1). In the second step, each quadrant is divided into four smaller quadrants of linear size $l_2 = L/2^2$ (in the m-th step, $l_m = L/2^m$), and the probability associated to each quadrant prior to the division is multiplied by P_1, P_2, P_3 and P_4 in random order. This procedure is continued and after n-generations each lattice site B is associated to a probability μ_B of the form

$$\mu_B = P_1^{S_1} P_2^{S_2} P_3^{S_3} P_4^{S_4}, \tag{1}$$

with $S_1 + S_2 + S_3 + S_4 = n$. In the limit $n \to \infty$ a multifractal distribution of probabilities on the two dimensional space is obtained (see below). Also, one can assume that

$$\mu_B = \exp(-E_B/kT), \tag{2}$$

where E_B is the activation energy of diffusion at the site B, k the Boltzmann constant and T the temperature. Equation (2) is consistent with the jumping probability of a random walk defined in Sect. IV (see (29)).

A multifractal is characterized by an infinite set of dimensions D_q (q real) [3, 5]. For the planar multifractals used in this work, these dimensions can be defined by

$$\sum_{S=1}^{2^{2m}} \mu_S^q \sim (lm/L)^{(q-1)D_q}, \tag{3}$$

where the sum runs over all the quadrants S of linear size lm, and μ_S is the probability (or measure) associated to the S^{th} quadrant. This measure is given by $\mu_S = \sum_B \mu_B$, where the sum runs over all the sites of the S^{th} quadrant. This implies that in the limit $lm/L \to 0$, the number N_α of measures behaving as (for more details see [5])

$$\mu_\alpha \sim (lm/L)^\alpha, \tag{4}$$

scales as

$$N_\alpha \sim (lm/L)^{-f(\alpha)}, \tag{5}$$

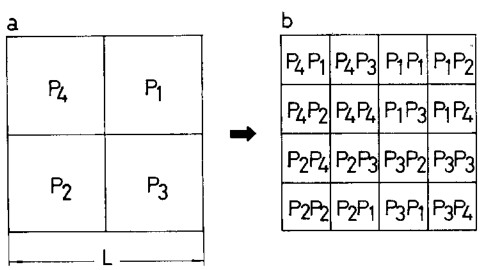

Fig. 1a, b. An example of construction of a planar multifractal. **a** first step; **b** second step

where $\alpha = d[(q-1)D_q]/dq$ and $f(\alpha) = q\alpha - (q-1)D_q$. This means that the system consists of pieces with fractal dimension $f(\alpha)$ and power law singularities α. Explicitly, in our case, from relation (3) one obtains

$$D_q = \frac{\ln(P_1^q + P_2^q + P_3^q + P_4^q)}{(1-q)\ln 2}. \tag{6}$$

The percolation model has been extensively studied (see for example [14–16] and references therein) in the field of geometric phase transitions, and its definition and properties will neither be presented nor discussed here. But let us recall that the incipient percolation cluster in two dimensions is a geometric fractal structure with fractal dimension $D_F = 91/48$ [17]. Let us stress that, in general, for a geometric fractal of dimension D_F one can assume that $\mu_C = \text{const}$ if the site C belongs to a fractal, and $\mu_C = 0$ otherwise. Then using the appropriate version of relation (3) one obtains $D_q = D_F$ for all q. In this sense a geometric fractal is a "trivial" multifractal.

Let us finally define the PM model as the intersection of an incipient percolation cluster on a square lattice with a planar multifractal of probabilities μ_B defined above. Note that both, the percolation clusters and the multifractals are confined in $2D$ planes and the intersection considered here is restricted to the case where the angle δ between these planes is zero (cases with $\delta \neq 0$ which cause the formation of $1D$ objects are not considered). So, in the PM model the probability μ'_B associated to a site B is given by

$$\mu'_B = \begin{cases} C\mu_B, & \text{if the site } B \text{ belongs to the incipient percolation cluster} \\ 0, & \text{otherwise} \end{cases} \tag{7}$$

where C is the normalization constant obtained by demanding that the following equation holds

$$\sum_B \mu'_B = 1, \tag{8}$$

where the sum runs over all B sites of the square lattice (note that $\sum_B \mu_B = 1$).

Let us stress that the explicit value of C does not affect at all the jumping probability of random walkers (see (29)), but we impose the condition (8) in order to deal with normalized probability distributions as used in multifractals (as it will be shown in Sect. III, the PM is a multifractal in the limit of very large square lattices).

Due to the intersection, the PM is *a structure which combines both, the geometric* (fractal) *heterogeneity* of the percolation cluster *and the energetic heterogeneity* (see (2)) of the planar multifractal. It can also be thought of as a dilute planar multifractal.

III. The intersection of fractals and multifractals

Let us consider a general case (in which the PM is a special one) of intersection, in a space of d euclidean dimensions, of a geometric random fractal structure of dimension D_F with a multifractal (normalized) probability distribution, both with overall linear size R and a short cut off r_0, where the restricted interpretation of the concept of intersection has to be remembered.

At the scale r, with $r_0 \leqq r \ll R$ (strictly speaking in the limit $r/R \to 0$), the fractal is composed by $(r/R)^{-D_F}$ boxes of linear size r and the multifractal has

$$N_\alpha \sim (r/R)^{-f(\alpha)} \tag{9}$$

boxes of measure

$$\mu_\alpha \sim (r/R)^\alpha. \tag{10}$$

The intersection of two fractal objects of dimensions D_F and f respectively, gives a new fractal of dimension f', where [18, 19]

$$f' = \begin{cases} f + D_F - d, & \text{if } f + D_F - d > 0 \\ 0, & \text{otherwise} \end{cases} \tag{11}$$

Before the intersection, the measures are normalized

$$\sum_\alpha N_\alpha \mu_\alpha = 1. \tag{12}$$

The intersection is defined following the same procedure as in the PM. That is, at the short cut off r_0 the measure μ_B becomes $\mu'_B = C_0 \mu_B$ if the box B belongs to the fractal; and $\mu'_B = 0$, otherwise, where C_0 is the normalization constant. Consequently, at the scale $r > r_0$ one has

$$\mu'_{\alpha'} = C \mu_\alpha, \tag{13}$$

with the same C value for all α', and the measures μ' remain normalized

$$\sum_{\alpha'} N'_{\alpha'} \mu'_{\alpha'} = 1. \tag{14}$$

Assuming $f + D_F - d > 0$, one obtains from (9) and (11)

$$N'_{\alpha'} \sim (r/R)^{-f'} \sim N_\alpha (r/R)^{d - D_F}, \tag{15}$$

and then (see (13))

$$N'_{\alpha'} \mu'_{\alpha'} \sim C (r/R)^{d - D_F} N_\alpha \mu_\alpha. \tag{16}$$

But from (12) and (14) one has

$$C \sim (r/R)^{D_F - d}, \tag{17}$$

and finally (see (10) and (13))

$$\mu'_{\alpha'} \sim (r/R)^{\alpha'}, \quad \alpha' = \alpha + D_F - d. \tag{18}$$

Let us note that a more detailed analysis (see [5]) shows that in the limit $r/R \to 0$, only the term with $\alpha = \hat{\alpha}$, where $\hat{\alpha}$ is obtained from the equality $f(\hat{\alpha}) = \hat{\alpha}$, contributes to the sum of (12). But the arguments to obtain (18) remain valid.
In summary, from (11) and (18) one has

$$f'(\alpha + D_F - d) = f(\alpha) + D_F - d, \tag{19}$$

and from this relation one can obtain the dimensions $D'_{q'}$ of the measures μ'. In fact [5], as $(q-1) D_q = q\alpha - f(\alpha)$, where $q = df/d\alpha$, and analogously for the measure μ', one has (note that $q' = q$)

$$D'_q = D_q + D_F - d. \tag{20}$$

In the above discussion, it is assumed that $f + D_F - d > 0$. As usual, small values of f correspond to large values of $|q|$, so (20) is only valid for $q_{min} \leqq q \leqq q_{max}$, where the values q_{min} and q_{max} depend on each particular intersection. In summary, *we claim that in d euclidean dimensions the equation* (20) *holds for the intersection of a random geometric fractal of dimension D_F with a multifractal probability distribution of dimensions D_q, at least for a finite interval of q.* In this sense, the intersection gives a new multifractal structure.

Let us note that for both, the case $q = 0$ (D_0 is the fractal dimension of the support of the measures) and the case of a trivial multifractal (that is $D_q = D$, for all q), the Eq. (20) is verified because it corresponds to the intersection of two fractal objects [18, 19].

For the special case of PM's, D'_q can be defined through (3) but replacing μ_S by μ'_S ($\mu'_S = \sum_B \mu'_B$, where the sum runs over all sites of the S^{th} quadrant). Then

$$\sum_{S=1}^{2^{2m}} \mu'^q_S \sim (lm/L)^{(q-1) D'_q}, \tag{21}$$

but now this relation is only valid in the limit $lm/L \to 0$ (i.e. in the case of very large lattices, $L \to \infty$; and for $m \to \infty$).
The relation (21) can be written as

$$\sum_{S=1}^{2^{2m}} \mu'_S \exp[(q-1) \ln \mu'_S] \sim \exp[(q-1) D'_q \ln(lm/L)]. \tag{22}$$

Expanding this relation in the limit $q \to 1$, one obtains (remember that due to the normalization $\sum \mu'_S = 1$)

$$\sum_{S=1}^{2^{2m}} \mu'_S \ln \mu'_S \sim \sigma' \ln(lm/L), \tag{23}$$

where

$$\sigma' \equiv \lim_{q \to 1} D'_q, \tag{24}$$

is the so-called information dimension [3]. On the other hand, for a planar multifractal one has (see (6))

$$D_q = \frac{1}{(1-q) \ln 2} \ln \sum_{i=1}^{4} P_i \exp[(q-1) \ln P_i]. \tag{25}$$

Then

$$\sigma \equiv \lim_{q \to 1} D_q = -\frac{1}{\ln 2} \sum_{i=1}^{4} P_i \ln P_i. \tag{26}$$

Finally, Eq. (20) implies

$$\sigma' = \sigma + \tfrac{91}{48} - 2, \tag{27}$$

where σ' and σ are obtained from (23) and (26) respectively. For $q=2$ one has

$$D'_2 = D_2 + \tfrac{91}{48} - 2, \tag{28}$$

where D'_2 and D_2 are obtained from (21) and (6) with $q=2$, respectively. As we will see below (in Sect. VI) the Monte Carlo results suggest that (27) and (28) are fulfilled.

IV. Random walkers and recombination reactions on multifractal structures

Meakin [13] has studied the properties of random walkers on planar multifractals. According to this work, we have assumed that the probability P_{BC} of a random walk at the site B with measure μ'_B to jump into a randomly chosen nearest-neighbour site C with measure μ'_C is

$$P_{BC} = \begin{cases} \mu'_C/\mu'_B, & \text{if } \mu'_C < \mu'_B \\ 1, & \text{otherwise} \end{cases} \tag{29}$$

In the computation of the time t only the jumping attempts to sites with no null measures are considered (see (7)). The average number S_N of distinct sites visited by a random walker after N steps is expected to behave, for large N, as

$$S_N \sim N^{\eta}, \tag{30}$$

where $\eta (0 \leqq \eta \leqq 1)$ is the random walk exponent η (for geometric fractals, $\eta = \tilde{d}/2$, for $\tilde{d} < 2$, where $\tilde{d}$ is the spectral dimension related to the density of states for scalar harmonic excitations of the fractal [20, 21]).

The visitative efficiency ε of a random walker is defined through the derivative of S_N with respect to time

$$\varepsilon \equiv \mathrm{d}S_N/\mathrm{d}t. \tag{31}$$

For DLRR's between A particles;

$$A + A \to \text{products}, \tag{32}$$

where the products are removed from the substratum; the reaction rate may be written as

$$-\mathrm{d}\rho/\mathrm{d}t \sim \varepsilon \rho^2, \tag{33}$$

where ρ is the density of A particles. This relation is only rigorous for the two body approximations (i.e. using the concept of relative diffusion between two walkers [9–11]) and for the substrata used in this work, the validity of the above mentioned relation is the first hypothesis. Note that t is proportional to the number of jumping attempts whereas N is the number of successful jumping events. Then, one *could* assume (the second hypothesis) that in average

$$N \sim t, \tag{34}$$

for large N. Let us stress that in a multifractal the jumping probability strongly depends on the spatial region of the substratum, then the relation (34) is not trivial at all (note that for some substrats, $N \sim t^{\beta}$, with $\beta \neq 1$ [22]). From (30), (31), (33) and (34) one obtains in the low density regime (i.e. in the limit $t \to \infty$)

$$\rho \sim t^{-\eta}. \tag{35}$$

In summary, *the conjecture is that in multifractals the diffusion limited recombination reaction exponent* η (35) *is the same as the random walk exponent* η (30).

On the other hand, the behaviour for large N of the average square distance R_N^2 from the origin of the walk is characterized by the random walk exponent ν. That is

$$R_N^2 \sim N^{2\nu}. \tag{36}$$

Furthermore, as D_0 is the fractal dimension of the support of the measure, one can see that the number of points within a region of radius R_N behaves, for large N, as $R_N^{D_0}$. Then from (36) one gets

$$\Sigma_N \sim N^{\nu D_0}, \tag{37}$$

where Σ_N is the number of accessible sites for a random walker after N steps. Now assuming that the random walk explores all the accessible space [21, 23] (i.e. $S_N \sim \Sigma_N$), one obtains from (30) and (37) that

$$\eta = \begin{cases} \nu D_0, & \text{if } \nu D_0 < 1 \\ 1, & \text{otherwise} \end{cases} \tag{38}$$

where the case $\eta = 1$ means that the number S_N of distinct sites visited by a random walker cannot exceed the number of N steps. This *compact exploration* assumption holds in fractals [21, 23] and it will be analysed for the multifractals used in this work.

V. The Monte Carlo simulation

Monte Carlo simulations are performed using $L \times L$ quare lattices with $L = 2^8 = 256$. The planar multifractals are generated by using

$$P_i = Q^{i-1} \left(\sum_{j=1}^{4} Q^{j-1} \right)^{-1}; \quad i = 1, \dots, 4, \tag{39}$$

where $Q (0 < Q \leqq 1)$ is a free parameter [13]. For obtaining PM's only clusters which percolate in both directions of the square lattice (i.e. they have both their width and their length equal to L) at the critical probability $p_C = 0.5927$ (see for example [24, 25]) are selected. Free (periodic) boundary conditions for the random walkers (DLRR's, respectively) are used.

In order to verify the relations (27) and (28), we computed $\sum \mu'_S \ln \mu'_S$ and $\sum \mu'^2_S$ for PM's (see (40) and (41) below and Fig. 2).

For a simple random walk (using the jumping probability (29), S_N and R_N (see (30) and (36)) are computed

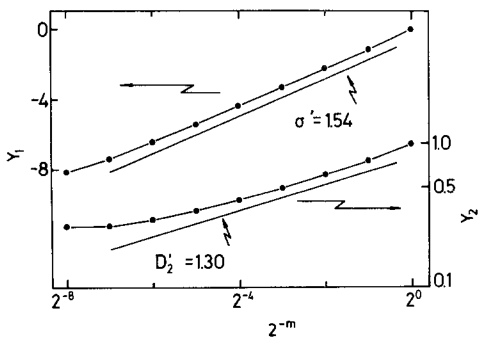

Fig. 2. Plot of y_1 and y_2 (ln scale) as a function of $lm/L=2^{-m}$ (ln scale, see (40) and (41)). The data, averaged over 400 samples, were obtained for PM's with $Q=0.5$ ($L=2^8=256$)

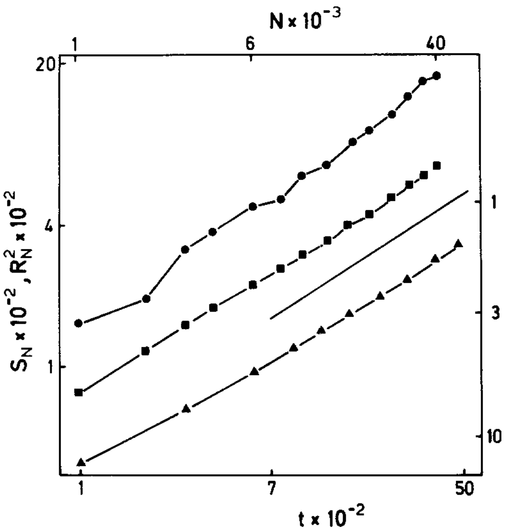

Fig. 3. ln-ln plot of R_N^2 (●), S_N (■) and ρ (▲) versus N and t, respectively; for PM substrata with $Q=0.75$. The results of R_N and S_N (of ρ) were obtained averaging over 200 walks (75 simulations), a new substratum was generated after 50 walks (15 simulations, respectively) and each of these walks starts from different points. The straight full line corresponds to $\eta=0.61$

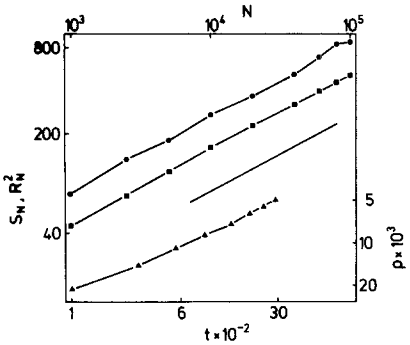

Fig. 4. The same as Fig. 3 but now for $Q=0.50$. The results of R_N and S_N (of ρ) were obtained averaging over 210 walks (40 simulations) and a new substratum was generated for every 30 walks (10 simulations, respectively). The straight full line corresponds to $\eta=0.52$

as a function of N and these results are averaged over a large number of walks (see Figs. 3 and 4).

To simulate the DLRR's the sites with no null measures are covered by A particles at random. After that the diffusion starts. The probability of a randomly selected A particle to jump into a randomly chosen nearest-neighbour site with a no null measure is defined as for random walkers (see (29)). When two A particles are at the same substratum site as a consequence of the jumps, both particles are removed from the substratum (successful recombination event). The time t is defined as $t=0.05\,M$, where M is the number of jumping attempts per particle (i.e. the number of successful jumping events plus the failed ones). The density ρ of A particles against t (see (35)) is obtained and averaged over many simulation reactions (see Figs. 3 and 4).

VI. Discussion of the results and conclusion

VI.1. The σ' and D'_2 for PM's

Let us now obtain $D'_1 \equiv \sigma'$ and D'_2 for PM's. As $lm/L = 2^{-m}$, from (23) ($q=1$) one has

$$y_1 \equiv \sum_{S=1}^{2^{2m}} \mu'_S \ln \mu'_S \sim \sigma' \ln 2^{-m}, \tag{40}$$

and from (21), (with $q=2$) it follows

$$y_2 \equiv 2^m \sum_{S=1}^{2^{2m}} \mu'^2_S \sim 2^{-m(D'_2-1)}. \tag{41}$$

In Fig. 2 we have a plot of y_1 versus $\ln 2^{-m}$ and ln-ln plot of y_2 versus 2^{-m} for PM's with $Q=0.5$. For this case, and using (6) and (26)–(28) one obtains

$$\sigma' \simeq 1.536, \qquad D'_2 = 1.300. \tag{42}$$

Comparing both y_1 and y_2 with the two straight lines of slopes 1.54 and D'_2-1 (with $D'_2=1.30$) respectively, in Fig. 2, one can see that the Monte Carlo results are in agreement with (27) and (28). Then the relation (20) is verified for PM's in the limit $q \to 1$ and for $q=2$.

VI.2. The Exponent η

Figures 3 and 4 show plots of S_N versus N and ρ versus t obtained for PM's with $Q=0.75$ and $Q=0.50$, respectively. The slopes of these curves strongly suggest that the random walk and the DLRR exponents η (see (30) and (35)) are the same. This conclusion agrees with the results obtained working with other PM's as well as with planar multifractals (see Table 1).

Based on all these evidences, *we claim that for the multifractals studied in this work, the random walk exponent* η (30) *and the diffusion limited recombination reaction exponent* η (35) *are the same.* Then, the simulation of DLRR's is an alternative method to obtain the random walk exponent η. Furthermore, in DLRR's η is related to the reaction order $X=1+1/\eta$ [9–12, 26], which can be experimentally determined.

Table 1. The exponents η and ν. The error bars are of about $\pm 5\%$ (a), $\pm 10\%$ (b), and they do not take into account any possible corrections due to finite size effects. For PM substrata, the η values were obtained considering both, random walk and DLRR simulation results. For planar multifractals the η values correspond to DLRR simulations only [26]. The square $L\times L$ lattice size is $L=256$. ξ and ξ' are the values previously published by Meakin [13] for random walks, assuming that S_N behaves as N^{ξ} or $(N/\ln N)^{\xi'}$, respectively; and on square lattices of size $L=1024$. Note the remarkable agreement with the results obtained from DLRR's even when the used lattice sizes were different

Q	PM		Planar Multifractal		
	η	2ν	η	ξ	ξ'
0.75	0.61[a]	0.70[b]	0.96[a]	0.885	0.985
0.50	0.52[a]	0.52[a]	0.79[b]	0.769	0.856
0.25	0.28[b]	0.26[b]	0.56[b]	0.541	0.603

VI.3. The compact exploration

Figures 3 and 4 also show plots of R_N^2 versus N. The ν exponents obtained from these figures and for the case $Q=0.25$ are presented in Table 1. Let us note that: *i)* on an incipient percolation cluster the visitation is compact [21, 23]; *ii)* the fractal dimension D_0 of PM substrata is the same as that of the incipient percolation cluster; (D_F) and *iii)* the obtained exponents (see (Table 1) for MP's (with $Q<1$) are smaller than the ν value ($\nu=0.352$) for the percolation cluster. This implies that for the number N of steps, the distance R_N travelled by the random walk on a PM structure is smaller than that over a percolation cluster. Therefore, one expects that the exploration would also be compact on PM substrata. Moreover, the results shown in Table 1 agree,, within the error bars, with $\eta=\nu D_0$, and Meakin [13] has shown that the visitation is compact on planar multifractals.

Summing up, from these evidences it seems that the compact exploration assumption holds for the multifractals studied in this work.

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