

with the low temperature spectra shows that there are two lines remaining constant at different water contents, at 1534 and 1633 cm^{-1} . In between one finds a broadband increasing in intensity and splitting into two lines at 1575 and 1591 cm^{-1} at higher water content.

(vi) At room temperature two broad lines arise at 3300 (assigned to ν NH, γ NH, and τ CN¹¹) and 3103 cm^{-1} (assigned to ν NH, τ CN¹¹).

The question concerning the nature of these lines is not easy to answer. For such low concentrations of water (in NMA) one could first consider the vibrational lines of the isolated molecule, but not all possible lines (1595, 3151, and 3652 cm^{-1})¹⁶ fit to our observed ones. The problem seems to be more complicated as the water molecule will be bound in some way to the hydrogen bonds of the NMA crystal. One might even speculate that there are at least two different sites that are accessible to different numbers of water molecules. This could cause the different sensitivities of the two lines in the amide I/II region at room temperature. Further investigations on deuteration effects may lead to the necessary new assignments although the two factors of deuterating the amide group in NMA and water can only be switched at the same time because of the fast H/D exchange between NMA and the added water.

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Fractal behavior of correlated random walk on percolating clusters

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Correlation in diffusional motion has been shown in the past to be a necessary idea for the explanation of experimental data ranging from the diffusion of hydrogen in metals¹ and models of diffusion in concentrated lattice gases,² to the relaxation mechanism of low-lying excited states of organic molecules at low temperatures³ as studied by the use of random walk hopping models.⁴ Recently a new model was introduced^{6,7} that incorporates the effects of correlation in the usual⁵ random walk models, first on perfect lattices,⁶ and then on mixed binary lattices.⁷ Also of interest⁸ has been the application of the simple stochastic random walk on the fractal structures of percolating clusters around the critical threshold point. In the present paper we study the behavior of correlated random walks on such fractal structures, we test the possible universality of these phenomena, and also their crossover to the classical limit of a perfect crystal.

Percolating clusters are generated by a Monte-Carlo simulation method, using the cluster growth technique.⁹ The only prescribed parameter is the concentration (relative fraction) of the open (allowed) lattice sites C , ranging from the critical value $C_c = 0.593$, to the limit of perfect lattice $C = 1.00$. Correlation is the retention of the directional memory over a certain number of lattice spacings. This is quantitatively described by the fraction p_f , which is the probability of a forward jump, and it is in the range: $a^{-1} < p_f < 1.00$, where a is the lattice coordination number.

The well-known relation⁸ connecting S_N , the number of distinct sites visited in an N -step walk, with N (time) is

$$S_N \sim N^{d_s/2}$$

which has been shown⁹ to hold true for a variety of lattices for stochastic random walks with a d_s value: $d_s^e = 1.30$, for

a random walk restricted to the largest cluster only, while $d'_s = 1.23$, for the random walk originating on *any* cluster, small or large,¹⁰ all for the square two-dimensional lattice. This is exhibited by the curve marked $p_f = 0.25$ in Fig. 1. In this case of the square lattice all four directions carry the same 0.25 probability of scattering, so that this is the limit of a totally uncorrelated walk. All calculations shown in Fig. 1 are performed using random walks that may start on any size clusters, so that exactly at the critical point ($C_c = 0.593$) the (modified^{10,11}) spectral dimension $d'_s = 1.23$. However, for $C = 0.60$, we observe that $d'_s = 1.33$, showing that we are already in the region of "crossover" to Euclidean behavior. As has been recently shown,⁹ this crossover is very sudden close to the critical point, so that the observed difference (1.23 to 1.33) is not surprising, but rather in agreement with these recent results. We observe that the standard deviations for all the above values are about ± 0.02 . We further observe that the crossover to Euclidean behavior eventually ($C = 1.00$) results in the value of $d'_s = 1.8$. (Here d'_s is an *effective* spectral dimension; as is well known, the difference from the $d_s = 2$ classical value is due to the logarithmic correction terms that the two-dimensional random walk exhibits.⁵)

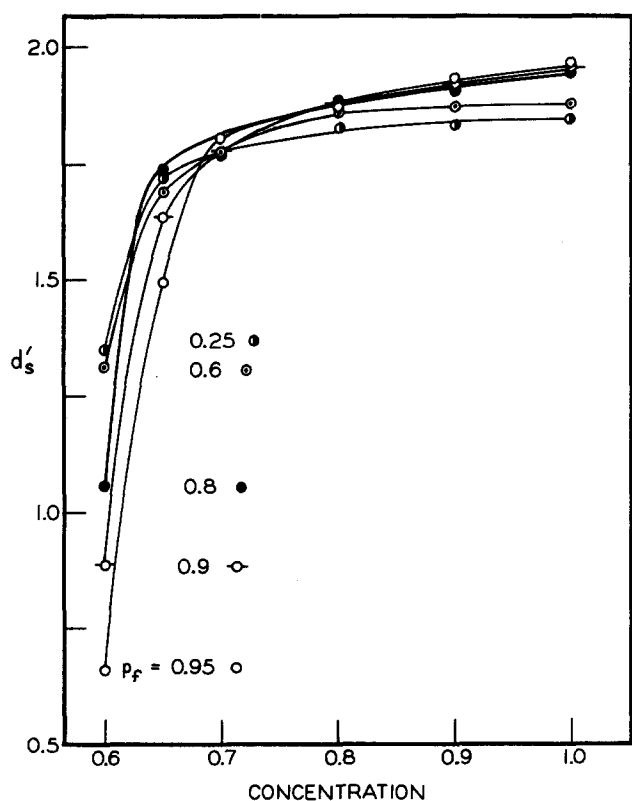


FIG. 1. The spectral dimension d'_s vs the concentration C , as a function of the forward correlation parameter p_f . These are results of random walks on lattices of 2000×2000 , averaged over 500 realizations, and the spectral dimensions are calculated in the long time limit, $N = 2 \times 10^5$ steps, from walks that may originate on any size cluster.

Of special interest here are the other curves in Fig. 1, which refer to higher p_f ($p_f > 0.25$) values. In the fractal limit ($C = 0.60$) we see that d'_s sharply decreases as p_f increases. For $p_f = 0.95$ (at $C = 0.60$) we see that $d'_s = 0.66$ only. This sharp decrease accompanies the fact that at the critical percolation threshold the correlated walks have a much smaller S_N value than the uncorrelated walks, because, as was originally shown in the past,⁴ the particle indulges for long times in revisiting the same row of sites over and over again. We also used this idea³ to interpret experimental data on mixed naphthalene alloys at 2 K. The trend in this decrease is not simple, however. Actually for small p_f , in the region $0.25 < p_f < 0.50$, we observe an increase in the d'_s value compared to $p_f = 0.25$, coming from a corresponding increase in the S_N respective values. Only for high p_f , i.e., $p_f > 0.60$, do we observe a clear decrease in the spectral dimension, resulting from a drastic decrease in S_N . This shows that the problem is considerably more complex than Fig. 1 shows, but its details are not to be addressed here. Thus, our preliminary conclusion is that correlated walks do *not* belong to the universality class of stochastic walks. Factors that have to be taken into account include: the exact value of C (remembering that $C = 0.60$ is not the critical point but $C_c = 0.593$), the length of time, contributions from runs on small finite clusters, the use of the "blind" or "myopic" ant model,¹² etc.

As C increases, one observes for each p_f value the corresponding crossover to the classical behavior, since now, above $C = 0.80$, correlated walks are much more efficient than uncorrelated ones. The region $0.70 < C < 0.80$, as seen from Fig. 1, is the "crossover region" between the different p_f values.

In summary, we have presented a preliminary study for correlated random walk motion on fractal structures, and we investigated the two types of crossover that occur, i.e., the crossover from fractal to Euclidean behavior for any type of walk, and the crossover from uncorrelated to correlated walk at any given concentration.

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