Annihilation reactions in two-dimensional percolation clusters: Effects of short-range interactions

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We report Monte Carlo simulations of annihilation reaction $A+A\rightarrow 0$ in two-dimensional percolation clusters. In the model, the particles diffuse coupled by nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions. The following cases have been studied: (i) repulsive NN interaction, (ii) repulsive NN and NNN interactions, and (iii) repulsive NN and attractive NNN interactions. In the intermediate-time regime, for cases (i) and (ii), we found that the density of particles ρ approximately decays as $\rho\sim t^{-\gamma}(\frac{2}{3}<\gamma<1)$, where γ depends on specific values of interactions but is independent of the initial density $\rho(t=0)$. For case (iii) there is no power-law decay. For the short- and intermediate-time regimes, a scaling ansatz for cases (i) and (ii) is found.

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I. INTRODUCTION

In the last few years, much effort has been dedicated to the study of the annihilation reaction (see, e.g., [1,2] and references cited therein)

$$A+A\rightarrow 0\ . \tag{1}$$

In these works the particles diffuse on a specific (Euclidean [3,4], fractal [5–7], multifractal [8], or anisotropic [9]) substratum and disappear when two of them collide. The reaction itself is considered to be instantaneous and irreversible. When there is no input of particles, as in the present work, the density $\rho(t)$ decays as a function of t. In most of these models the interactions between diffusive particles were not taken into account. In this so-called "diffusion-controlled reaction" only one characteristic time t_1 appears [4]. In the long-time regime ($t\gg t_1$) it was found that [4,6,7,10,11]

$$\rho(t)\sim\begin{cases} t^{-d_s/2} & \text{if } d_s<2 \\ t^{-1} & \text{otherwise} \end{cases}\ , \tag{2}$$

d_s being the spectral dimension of the substratum where the particles diffuse [12]. Due to the anomalous behavior of diffusion for $d_s<2$ [10], the theoretical study in one dimension and fractal substrata has attracted special interest. Equation (2) has also been experimentally verified [1].

In real systems, the interactions are present (see, e.g., [13]). In an effort to understand their influence, we have made a careful examination of $\rho(t)$ for a simple annihilation reaction model, with short-range interactions in the fractal two-dimensional percolation cluster ($d_s=\frac{4}{3}$).

II. THE MODEL AND THE MONTE CARLO SIMULATION

We have considered the reaction (1) between particles diffusing on a two-dimensional percolation cluster. In the simulation, the standard percolation model (see, e.g., [14]) was used on a 350×350 square lattice. Only clusters which percolate in both directions of the square lattice, at the critical probability $\rho_c=0.593$ (see, e.g., [15]), were selected.

In our model, we consider nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions between particles with energies U' and W' respectively. We will concentrate our attention on the cases of $U'>0$ (repulsive) with either $W'>0$ (repulsive), $W'=0$ (null), or $W'<0$ (attractive) interactions.

The energy H of a given configuration of particles can be written as

$$H/k_BT=\sum_{\text{NN } i,j} Un_in_j+\sum_{\text{NNN } i,k} Wn_in_k\ , \tag{3}$$

where n_i, equal to 0 or 1, is the occupation number of site i in the lattice. k_B is the Boltzmann constant and T the temperature. U $(=U'/k_BT)$ and W $(=W'/k_BT)$ are the dimensionless NN and NNN interactions between particles. NN i,j and NNN i,k denote the summation over the NN and NNN pairs.

The reaction takes place between NN occupied sites. We arrange the initial configuration of particles so that there are no pairs of NN occupied sites. In order to do this, we take a set of all NNN sites that belong to the percolation cluster and we occupy each of these sites with equal probability $2\rho_0$. In this way, the actual initial density, defined as the number of particles per cluster site, is ρ_0 (then $\rho_0=0.5$ is the maximum possible coverage). After that, the diffusion starts.

The particles perform a random walk between NN sites of the percolation cluster. As is usually done (see, e.g., [7] and Meakin and Stanley in [6]), we use periodic boundary conditions in order to avoid edge effects. The lattice is large enough to avoid the exponential decay of ρ at large times caused by the artificial visitations in finite periodic systems [7].

In the simulations, at *each jump attempt,* one of the $N(t)$ particles present in the system at time t, randomly

chosen, attempts to jump to any of the four NN sites of the square lattice with equal probability $\frac{1}{4}$. The following situations may appear.

(1) The chosen site does not belong to the cluster and the jump is not performed.

(2) The chosen site belongs to the cluster, then the selected particle attempts to jump with probability P given by (Metropolis method, see, e.g., [16])

$$P=\min[1,\exp(-\Delta H/k_BT)] , \tag{4}$$

where ΔH is the energy change in the movement of the selected particle to the chosen site.

If the jump is performed and the chosen site is NN of a second particle, both particles react and the number $N(t)$ is reduced to $N(t)-2$. If the particle jumps and the chosen site is NN of more than one particle, the reaction is performed with one of these particles with equal probability.

In the simulation, a time interval equal to 1 is defined as the time needed for the $N(t)$ particles to have, on average, one chance to jump. Specifically, after each jump attempt, the time t is increased by $1/N(t)$.

The density $\rho(t)$ is obtained by averaging the number of particles per cluster site at time t over many (typically 20–40) samples. Every five samples, a new percolation cluster was generated.

III. RESULTS AND DISCUSSION

In the present paper we have worked with the following cases of short-range interactions: (a) Null interaction ($U=W=0$); (b) repulsive NN interaction ($U>0$, $W=0$); (c) repulsive NN and NNN interactions with the condition $U\geq W>0$; and (d) repulsive NN interaction and attractive NNN interaction with the condition $U>-W>0$. Figure 1 shows the time dependence of the density (in the log-log scale) for different values of the interactions. Specifically we have plotted the cases $U=W=0$; $U=4$, $W=0$; $U=W=4$, and $U=4$, $W=-3$.

A. Case (a) and general comments

Let us start discussing the general conclusions which can be obtained from Fig. 1. After that, we will analyze specifically each case of interaction. The case $U=W=0$ is shown for comparison with other cases. For a very long-time regime, it is known that

$$\rho(t)\sim t^{-\gamma_a} \quad (t\to\infty) , \tag{5}$$

where, for the percolation cluster, the value of the *asymptotic exponent* γ_a is [see Eq. (2)]

$$\gamma_a=d_s/2=\tfrac{2}{3} . \tag{6}$$

The full straight line of Fig. 1 has a slope $-\frac{2}{3}$. In the interval $4<\log_{10}t<6$ our results can be fitted by a straight line of slope 0.63 ± 0.01. This value (less than γ_a) is in agreement with other Monte Carlo simulation results of d_s [17] and γ_a [7]. The small curvature suggests that the asymptotic behavior will hold for longer times not reached in our simulation.

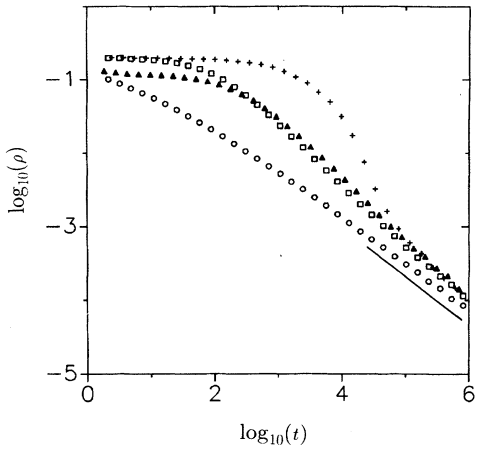

FIG. 1. Density ρ vs time t in $\log_{10}$-$\log_{10}$ scales for annihilation reactions on percolation clusters ($p_c=0.593$) on square lattice of linear size $L=350$ and for different short-range interactions [see Eq. (3)]: ○, $U=W=0$; □, $U=4$, $W=0$; ▲, $U=W=4$; and +, $U=4$, $W=-3$. The straight line is drawn as a guide and has a slope $-\frac{2}{3}$.

For very long times, independently of the interactions, the density ρ is very small, therefore, on average, the interparticle distance is very large. For this reason, one expects that the rate of reaction (1) will be controlled by diffusion processes. Then it is expected that after a second characteristic time t_2, Eqs. (5) and (6) will hold independently of the short-range interactions. This asymptotic regime was not reached in our simulation. However, the approach of all curves to the case $U=W=0$, observed in Fig. 1 at $t\sim10^6$, is an indication that the asymptotic behavior will be valid for longer times. The asymptotic regime has also been suggested in other related models [18,19].

Since repulsive interaction diminishes the reaction probability, $\log_{10}\rho$ will remain almost constant for a longer-time regime $0<t\ll t_1$ compared with the case of no interaction. The plateau is clearly seen in Fig. 1 where it is also seen that t_1 depends on the specific values of U and W. For intermediate times $t_1\ll t\ll t_2$, the behavior of $\rho(t)$ strongly depends on the interactions.

B. Case (b)

In Fig. 2 we plot $\log_{10}\rho(t)$ versus $\log_{10}t$ for $U=4$, $W=0$ and $U=6$, $W=0$; for $\rho_0=0.2$ and 0.05. From this figure one can see that in the intermediate-time regime ($t_1\ll t\ll t_2$) the density approximately behaves as

$$\rho(t)\sim t^{-\gamma} . \tag{7}$$

When $U\gg0$, the value of the *intermediate exponent* γ depends on the value of U, but is independent of the initial density ρ_0, suggesting some degree of universality. Strictly speaking, the exponent γ slightly depends on ρ_0 for small values of U. This dependence decreases as U increases. For example, for $U=4$ we found that the straight lines have slopes $\gamma=0.80\pm0.01$ and 0.85 ± 0.01 for $\rho_0=0.05$ and 0.2, respectively. For $U=6$, the

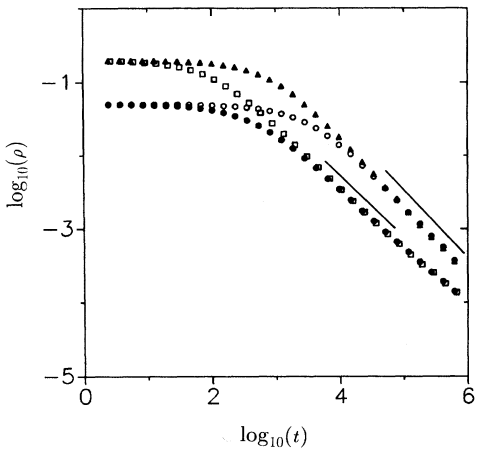

FIG. 2. Density ρ vs time t in $\log_{10}$-$\log_{10}$ scales for potentials $U>0$, $W=0$. The curves correspond to the following cases: □, $U=4$, $\rho_0=0.2$; ●, $U=4$, $\rho_0=0.05$; ▲, $U=6$, $\rho_0=0.2$; and ○, $U=6$, $\rho_0=0.05$. The straight lines have slopes -0.93 and -0.85.

straight lines have slopes $\gamma=0.90\pm0.01$ and 0.93 ± 0.01 for the same values of ρ_0. It is expected that in the limit $U\to\infty$ this dependence disappears for all $\rho_0>0$. Nevertheless this limit cannot really be reached because also $t_1\to\infty$ when $U\to\infty$ (the reaction never occurs). Let us remark that the difference in the values of γ obtained for finite values of U (of about 6% for $U=4$ and 3% for $U=6$, when ρ_0 changes by a factor 4, from $\rho_0=0.05$ to 0.2) is small and it is of the same order as the error in the exponents for finite systems. For example, as was mentioned, for $U=W=0$ we obtain $\gamma_a=0.63\pm0.01$ in place of the exact value $\gamma_a=\frac{2}{3}$ (the error is 6%).

Let us note that the reaction delay produced at short times is recovered at intermediate times. That is, $\gamma>\gamma_a$ (see also Fig. 1). Notice also that the value of γ increases as U increases. The length of the intermediate-time regime, where Eq. (7) holds, also increases as U increases [see Figs. 3(a) and 3(b)]. Let us stress that this length is of about two decades for $U=6$.

The same kind of behavior as those shown in Fig. 2 was obtained in [18] for the annihilation reaction (1) in a one-dimensional lattice. In the model of Ref. [18] the interactions are not present, but the reaction (1) occurs with a probability p when two particles attempt to occupy the same lattice site. The probability $p<1$ there plays a role similar to that of the interaction $U>0$ in our present work. From Ref. [18] we propose the following scaling ansatz for the short- and intermediate-time regimes $0<t\ll t_2$ and for a fixed value of U:

$$\rho(t)=\rho_0 f(t/t_1)\ , \tag{8}$$

where

$$f(x)\sim\begin{cases}1 & \text{for } x\ll 1\\ x^{-\gamma} & \text{for } x\gg 1 \text{ and } t\ll t_2\end{cases}\ . \tag{9}$$

From Eq. (7) one has that $\rho(t)\sim\rho_0(t/t_1)^{-\gamma}$, and using the fact that $\rho(t)$ is independent of ρ_0 when $t\gg t_1$ one obtains

$$t_1\sim\rho_0^{-1/\gamma}\ . \tag{10}$$

In order to check this scaling ansatz we plot, in Fig. 3, $\log_{10}(\rho t^{\gamma})$ versus $\log_{10}(t\rho_0^{1/\gamma})$ for the cases $U=4$, $W=0$, and $U=6$, $W=0$, using two different values of ρ_0. As is expected, the data collapse improves when U increases [see Figs. 3(a) and 3(b)]. Other similar data collapses (not shown here) have been obtained for different values of $U>0$, $W=0$, and ρ_0. All these results suggest that the proposed scaling ansatz is a very good representation of our numerical data for large values of U.

From Ref. [18] and the results obtained in the present work (the value of γ increases, as U increases, see Fig. 2), it is expected that when $U\to\infty$, $\gamma\to1$. The case $\gamma=1$ is described by the mean-field equation $-d\rho/dt\sim\rho^2$, where diffusion effects are irrelevant. For one-dimensional sys-

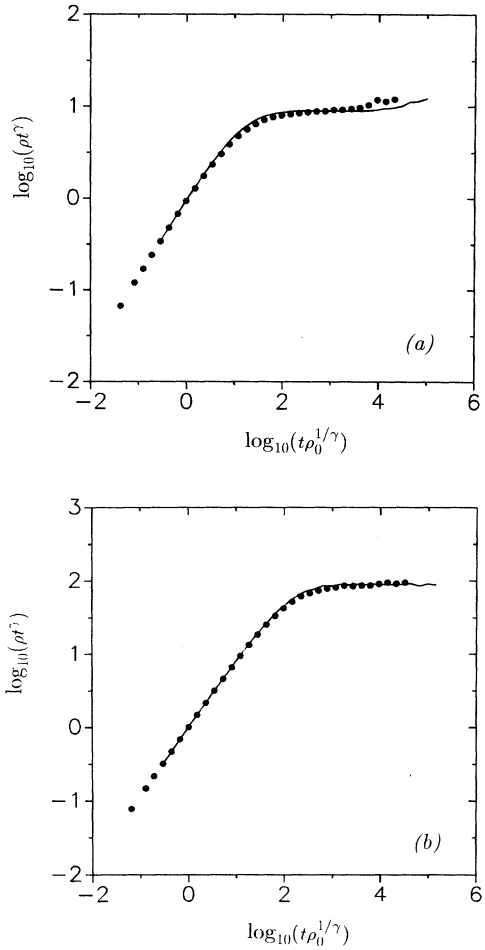

FIG. 3. Plot of ρt^{γ} vs $t\rho_0^{1/\gamma}$ in $\log_{10}$-$\log_{10}$ scales for potentials $U>0$, $W=0$. The election of the axes is more appropriate here than in Fig. 2, in order to carefully analyze the validity of the power-law decay of Eq. (7) (which here corresponds to a straight line of slope zero). (a) corresponds to $U=4$, $\gamma=0.85$; curved line, $\rho_=0.2$; ●, $\rho_0=0.05$. (b) corresponds to $U=6$, $\gamma=0.93$; curved line, $\rho_0=0.2$; ●, $\rho_0=0.05$.

tems the behavior $\rho(t)\sim t^{-1}$ is also suggested in Refs. [19,20].

The deviation of the plateau behavior (i.e., the breakdown of the scaling ansatz) at long times, shown in Fig. 3(a), is an indication of the change towards the asymptotic regime behavior [Eqs. (5) and (6)] expected for $t \gg t_2$ ($\gamma_a = \frac{2}{3} < \gamma = 0.85$). It is also expected that the same deviation should appear for the case $U=6$, $W=0$ for longer times than those shown in Fig. 3(b).

C. Case (c)

In order to study the influence of the repulsive NNN interaction ($W>0$), we will start discussing the limiting case $U=W>0$ (the case $W>U$ seems to be unrealistic, and for this reason it is not studied). In Fig. 4 we plot $\log_{10}[\rho(t)]$ versus $\log_{10}t$ for $U=W=4$ and $\rho_0=0.2$ and 0.05. Special initial configurations without pairs of NNN occupied sites were used. We found that the power-law behavior of Eq. (7) for intermediate times holds also for this case. Comparing the data of Figs. 4 and 2, one concludes that the scaling ansatz of Eqs. (8)–(10) will also be valid. From a detailed analysis (not shown here) we find that the collapse improves when the value of U increases, for cases $U=W$ and $U>W\geq 0$ with W fixed.

If one starts with initial configurations defined in Sec. II, the initial pairs of NNN occupied sites can easily annihilate at very short times [see Eqs. (3) and (4) and remember that the reaction takes place between NN occupied sites]. This effect is shown in Fig. 4 (see circles and triangles there). In general, for cases with and without initial NNN occupied sites, with the same values of $U\geq W>0$, the behavior of $\rho(t)$ in the intermediate- and long-time regimes is the same. Then the power-law decay holds for both cases with the same value of γ.

D. Case (d)

Let us finally consider case (d) where attractive NNN interaction ($W<0$) is present. Due to this interaction, the most energetically favorable configuration of two particles [see Eq. (3)], is when they form a pair of NNN occupied sites (see Fig. 5).

A way of characterizing this structure formation is to measure the number of particle pairs at NNN sites. We define $\Theta(t)$ as the number of pairs of NNN occupied sites per particle present on the percolation cluster at time t.

In Fig. 6 $\log_{10}\rho(t)$ and $\Theta(t)$ are shown as a function of $\log_{10}t$. The interaction used was $U=4$ and $W=-3$. In these figures, Θ grows at short times ($0\leq t<10^3$ and $0\leq t<10^{3,4}$, for $\rho_0=0.05$ and 0.2, respectively), indicating structure formation (also see Fig. 5). In intermediate times the structures disintegrate. An abrupt decay of $\Theta(t)$ and $\log_{10}\rho(t)$ takes place ($3.4<\log_{10}t<4.6$ and $4<\log_{10}t<4.8$, for $\rho_0=0.05$ and 0.2). The decay of $\Theta(t)$ and $\log_{10}\rho(t)$ is more abrupt for larger values of ρ_0. Let us remark that the maximum of $\Theta(t)$ takes place at time $t_{\max}\simeq t_1$ ($t_{\max}\sim 10^3$, $10^{3,4}$ for $\rho_0=0.05$ and 0.2, respectively). In the intermediate regime the behavior of $\log_{10}\rho(t)$ strongly depends on ρ_0 and for this reason it is not possible to find a simple scaling ansatz of the type of Eqs. (8)–(10). At long times, the structures disappear ($\Theta\rightarrow 0$).

In Fig. 6(b) values of $\Theta(t)$ for the rest of the cases are shown as a function of $\log_{10}t$. Let us note that only for case (d) ($W<0$) does $\Theta(t)$ increase at short times. For all the other cases $\Theta(t)$ decreases for all times. Strictly speaking, for the case with no initial NNN occupied sites (not shown in the figure), there is a very small increase of $\Theta(t)$ at short times, but this increase is irrelevant because we *impose* that $\Theta=0$ at $t=0$.

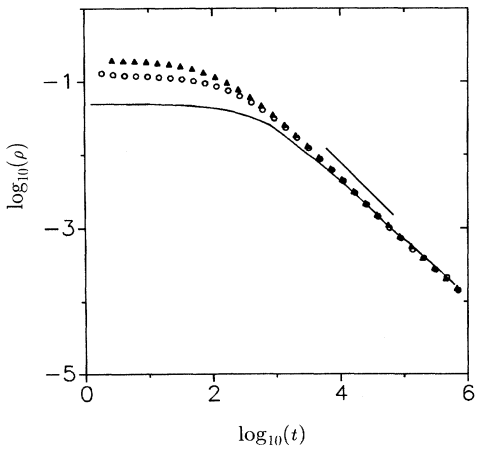

FIG. 4. Density ρ vs time t in $\log_{10}$-$\log_{10}$ scales for the interaction $U=W=4$. ▲, $\rho_0=0.2$ and initial configuration without NNN occupied sites. ○, $\rho_0=0.2$ and initial configuration with NNN sites; note the decay at very short times with respect to the previous case. Curved line, $\rho_0=0.05$ without NNN occupied sites. The straight line has a slope −0.87.

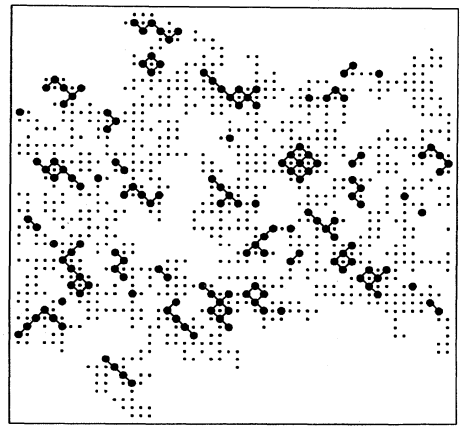

FIG. 5. The NNN structures of particles. A 50×50 section of the percolation cluster on a 350×350 lattice is shown. The empty and occupied cluster sites are denoted by small and big circles, respectively. The snapshot picture corresponds to $t=500$ with interaction $U=4$, $W=-3$, and initial density $\rho_0=0.2$. The particles separated by NNN distance have been joined by lines in order to guide the eyes. One clearly sees that most of the particles belong to NNN structures of particles.

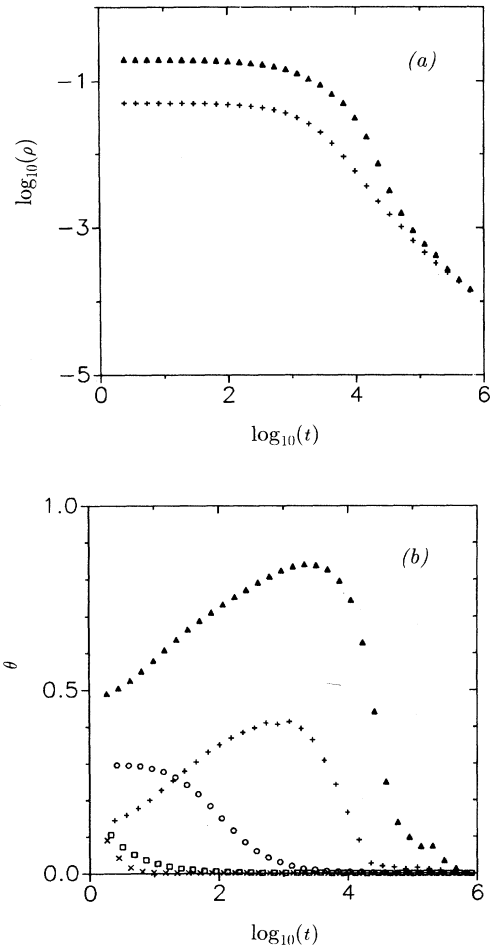

FIG. 6. (a) $\log_{10}\rho$ vs $\log_{10}t$ and (b) Θ vs $\log_{10}t$ for the interaction $U=4$, $W=-3$, and for the following initial densities: ▲, $\rho_0=0.2$ and +, $\rho_0=0.05$. Both graphics have the same time scale. In (b) there are also plotted the following interactions for $\rho_0=0.2$: □, $U=W=0$; ○, $U=4$, $W=0$; and ×, $U=W=4$ with initial NNN occupied sites. Note that the case with $W=3$ is the only one in which the value of Θ increases in the short-time regime.

IV. CONCLUSION

In summary, we studied the annihilation reaction $A+A\rightarrow 0$ in the two-dimensional percolation cluster. After a random deposition of particles on the substratum, they diffuse and interact with three kinds of short-range interactions [cases (b), (c), and (d), see Sec. II]. The reaction occurs between NN occupied sites. The main conclusions follow.

(1) In general the repulsive NN interaction ($U>0$) present in the cases (b)–(d) produce, at short times ($t \ll t_1$), a delay in the reaction comparing with the case of no interaction (see Fig. 1). At intermediate times ($t_1 \ll t \ll t_2$) the behavior of the density of particles $\rho(t)$ strongly depends on the kind of interaction. At long times ($t\rightarrow\infty$) one expects that the behavior of $\rho(t)$ is given by Eqs. (5) and (6) regardless of the interactions.

(2) At intermediate times, the power-law behavior given by Eq. (7) holds for cases (b) and (c) for large values of U (see Figs. 2 and 4). For given values of the short-range interaction, the value of the exponent γ is *independent* of initial density of particles ρ_0, which means some degree of universality. The length of the time interval where the power-law behavior is fulfilled increases when U increases. Nevertheless, due to the lack of exact theoretical result for $\rho(t)$, the power-law decay must be considered as a way, the simplest one, to approximate our numerical data. For this reason the exponent γ must be considered as an *effective* exponent.

(3) In the short and intermediate regimes the scaling ansatz given by Eqs. (8)–(10) (see Fig. 3) is verified for large values of U and for cases (b), and (c) without initial NNN occupied sites.

(4) For case (d) it is interesting to remark the formation of groups of particles connected by NNN distances at short times (see Figs. 5 and 6) and the abrupt decay of $\log_{10}\rho$ and $\Theta(t)$ at intermediate times, which means the sudden extinction of NNN structures.

Let us finally comment that, even though our Monte Carlo simulations are restricted to two-dimensional percolation clusters, we expect similar conclusions for all substrata with the spectral dimension $d_s<2$ (for these cases $d_2/2<\gamma<1$). On the other hand, in the intermediate-time regime, the density is not so low as in the asymptotic regime, where one expects the universal behavior of Eq. (2). Therefore, from the experimental point of view, the short and intermediate regimes acquire importance. We hope that our work will encourage new experiments on annihilation reactions between adsorbed particles on disordered media with $d_s<2$. Experimentally, the random initial deposition can be thought of as the condensation of incident particles onto a cooled substratum. After that, with a sudden increase in temperature, the diffusion starts. Although we do not attempt to predict any specific experiment, it is interesting to comment that, for example, an adimensional interaction $U=4$ at $T=200$ K corresponds to an interaction energy U' of about 6.7 kJ/mol, which is on the same order as that of the adsorbate-adsorbate lateral interaction observed in real systems.

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