Damage spreading in the Ziff-Gulari-Barshad model

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The spreading of initial damage globally distributed on the system is studied in a dimer-monomer irreversible reaction process (i.e., the ZGB model [Ziff, Gulari, and Barshad, Phys. Rev. Lett. **56**, 2553 (1986)]) in two dimensions. It is found that the damage heals within the poisoned states but spreads within the reactive regime. Both the frozen-chaotic and reactive-poisoned irreversible transitions occur at the same critical points and are of the same order. However, the order parameter critical exponents at the second-order transition are different, suggesting that damage spreading introduces a new dynamic critical behavior. A variant of the ZGB model (e.g., the ZGBER model), which is obtained by the addition of an Eley-Rideal reaction step, is also studied. In two dimensions, damage heals within the poisoned state. However, in contrast to the ZGB model, within the reactive regime, a frozen-chaotic transition is found to occur at a different critical point than the poisoning-reactive transition. At the frozen-chaotic critical point the damage heals according to a power-law behavior, $D(t)\propto t^{-\delta}$, with $\delta\cong 0.65$. The order parameter critical exponent is also determined and the fact that damage spreading introduces a new kind of dynamic critical behavior is established. Damage healing is observed in one dimension for the ZGBER model.

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I. INTRODUCTION

The understanding of irreversible dynamical many-particle systems is relevant in many fields of physics, chemistry, biology, ecology, etc. However, studying these systems one frequently deals with complex mechanisms consisting of a vast number of elementary processes. Since the handling of such a large amount of mechanisms imposes severe problems, the usual procedure is to rationalize the study by analyzing such elementary steps separately. Within this context the study of simple models for irreversible reaction processes has received much attention. For example, the dimer-monomer reaction model, as proposed by Ziff, Gulari, and Barshad (i.e., the ZGB model) [1], has attracted growing attention [2–22]. The ZGB model mimics the catalytic oxidation of carbon monoxide (e.g, $A\equiv CO$, $B_2\equiv O_2$, and $AB\equiv CO_2$) [23], which proceeds according to the Langmuir-Hinshelwood mechanism [23]:

$$A(\mathrm{g})+V\rightarrow A(\mathrm{ad}) , \quad (1)$$

$$B_2(\mathrm{g})+2V\rightarrow 2B(\mathrm{ad}) , \quad (2)$$

$$B(\mathrm{ad})+A(\mathrm{ad})\rightarrow 2V+AB(\mathrm{g}) . \quad (3)$$

Here V represents a vacant site on the catalyst surface and (g) and (ad) denote the gas and adsorbed phases, respectively.

The ZGB model uses a square lattice to represent the catalytic surface. A and B_2 molecules are selected randomly with relative probabilities Y and $(1-Y)$, respectively, and an attempt is made to add the selected species to the surface. If the selected species is A, one surface site is selected at random, and if that site is vacant, A is absorbed on it [Eq. (1)]. Otherwise, if that site is occupied, the trial ends and a new molecule is selected. If the selected species is B_2, a pair of nearest neighbor (NN) sites are selected at random and the molecule is added to them only if they are both vacant [Eq. (2)]. After each adsorption event, the NN sites of the added molecule are examined in order to account for the reaction given by Eq. (3). If more than one $\{B(\mathrm{ad}),A(\mathrm{ad})\}$ pair is identified, a single one is selected at random and removed from the surface.

Interest in the ZGB model arises due to rich and complex irreversible critical behavior [1–22]. In fact, in two dimensions and for the asymptotic regime ($t\rightarrow\infty$), the system reaches a stationary state whose nature solely depends on the parameter Y. For $Y\leq Y_{1c}\cong 0.3907$ ($Y\geq Y_{2c}\cong 0.525$) the surface becomes irreversibly poisoned by B (A) species, while for $Y_{1c}<Y<Y_{2c}$ a steady state with sustained production of AB is observed. So, just at Y_{1c} and Y_{2c} the model exhibits irreversible or kinetic phase transitions (IPT's) between the reactive regime and poisoned states, which are of second and first order, respectively.

Since the ZGB model is a simplified description of the actual catalytic reaction, a number of studies have been performed in order to investigate the influence of relevant additional parameters [3–7,10,11,13,16–22]. Among others, the addition of an Eley-Rideal (namely, the ZGBER model) reaction step,

$$B(\mathrm{ad})+A(\mathrm{g})\rightarrow V+AB(\mathrm{g}) , \quad (4)$$

has been considered by various authors [9,12].

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On the other hand, very recently, I have shown that damage spreading introduces a different kind of dynamic critical behavior in some irreversible reaction models [24]. The damage spreading problem consists first in taking a steady state configuration of the system $\{\sigma^A\}$ and to create at $t=0$, an initial damage $D(0)$ in that configuration (which gives a second configuration $\{\sigma^B\}$). Then, one investigates the time evolution of both configurations, using the same dynamics, calculating their Haming distance or damage, defined by

$$D(t)=\frac{1}{N}\sum_{i=1}^{N}|\sigma_i^A(t)-\sigma_i^B(t)| , \quad (5)$$

where N is the number of sites of the system. Physically, $D(t)$ just measures the fraction of sites for which both configurations are different. Starting from a small $D(0)$ value, $D(t)$ will go asymptotically to zero in the so called frozen phase, whereas it will tend to a finite value different from zero in the so called chaotic phase ([25–30]; for a review see [31]).

While the study of damage spreading in systems exhibiting reversible phase transitions has received much attention [25–31], similar studies in systems undergoing irreversible transitions are still in their infancy. So, the aim of this work is to investigate the spreading of damage in both the ZGB and the ZGBER models in order to study frozen-chaotic transitions in irreversible reaction systems.

II. SIMULATION METHOD AND THEORETICAL BACKGROUND

The Monte Carlo simulation procedure for both the ZGB and the ZGBER models has been outlined in the Introduction. For further details see, for example, Refs. [1–22]. The Monte Carlo time unit is defined such that each site of the lattice is visited once, on the average. Starting steady state configurations are obtained after $t=2\times10^3$. Then the damage is created and its spreading is monitored following the dynamics of both configurations simultaneously. For this purpose, the crucial idea is to apply, on the configurations $\{\sigma^Y\}$, the same sequence of random numbers in the algorithm in order to produce the same dynamics. This procedure requires special care because for each adsorption-reaction trial and due to the damage, one frequently needs to use a different amount of random numbers to follow the dynamics of the configurations $\{\sigma^Y\}$. So, in order to keep the synchronism, a set of random numbers $\{r_i\}$, $i=1,\ldots,M$, with M just enough to account for all possible situations, is generated before starting the trial. Then, during the trial, the random numbers are used sequentially for the same purpose, e.g., to choose the incoming species, to select a neighboring site, etc. Of course, some random numbers may not be necessary, so they are disregarded before starting a new trial.

According to the discussion above, one would then call the dynamic behavior of the systems chaotic if $D(t)$ takes a finite value for large times if $D(0)\rightarrow0$ [25–31]. On the contrary, the damage is healed in the frozen phase, i.e., $D(\tau)=0$ for $\tau\rightarrow\infty$; this means that during their respective time evolution both configurations become identical.

Note that in Eq. (5), D is defined in terms of σ, i.e., a spin variable which usually takes two values. Nevertheless, for the models studied in this work one has that the sites of the lattice may be V, A, and B when they are vacant, and occupied with A and B species, respectively. So, the three contributions to D given by V-A, V-B, and A-B are taken to be equal to unity in the evaluation of Eq. (5).

Since one has to work with finite, although small $D(0)$ values, it is necessary to take the limit $D(0)\rightarrow0$ in order to obtain reliable results. This tedious work can be avoided if three configurations $\{\sigma^A\}$, $\{\sigma^B\}$, and $\{\sigma^C\}$, such as $D_{AB}(0)=D_{BC}(0)=(\frac{1}{2})D_{AC}(0)=s$, are considered [31]. Then,

$$D(t)=D_{AB}(t)+D_{BC}(t)-D_{AC}(t) \quad (6)$$

is a very good extrapolation to $D(0)\rightarrow0$. We are interested in the study of an initial damage globally distributed on the system, in contrast to another approach which considers an initially localized damage. Therefore, in most cases considered in this work, the initial damage is created changing a fraction $s=0.1$ of the sites at random. However, in some particular cases clearly identified in the text, we study the dependence of the damage on s for smaller s values.

Simulations are performed in lattices of size $L=100$ ($L=10\,000$) in two (one) dimensions, respectively. Periodic boundary conditions are assumed and results are averaged over 100 different samples.

III. RESULTS AND DISCUSSION

A. ZGB model in two dimensions

In this model and for $Y<Y_{2c}$, the initial damage can only be created removing at random a fraction s of B species, since the concentration of A species within the reactive regime is almost negligible [1,2]. It is found that for $Y\leq Y_{1c}$ and $Y\geq Y_{2c}$ the damage becomes quickly healed, indicating that, as expected, the B- and A-poisoned phases, respectively, are also frozen phases. This behavior characterizes systems having unique poisoned states. However, if the poisoned state is nonunique one may observe damage spreading also within this state as, e.g., in the case of the dimer-dimer reaction process [32]. Within the reactive regime of the ZGB model, i.e, for $Y_{1c}\leq Y\leq Y_{2c}$, one observes that the damage spreads and after a short transient period reaches a stationary value, as is shown in Fig. 1. Here the time evolution of the damage has been monitored for different s values and also for two Y values close to the critical points. Figure 1 also shows that the final damage is independent of the initial distance between the starting configurations, pointing out the consistency of Eq. (6). Figure 2 shows the dependence of the stationary value of the damage on Y, within the reactive regime and taking $s=0.1$. From Fig. 2, it follows that the critical points at which the onset of damage spreading is found are the same as those characteristics of the reactive-poisoning

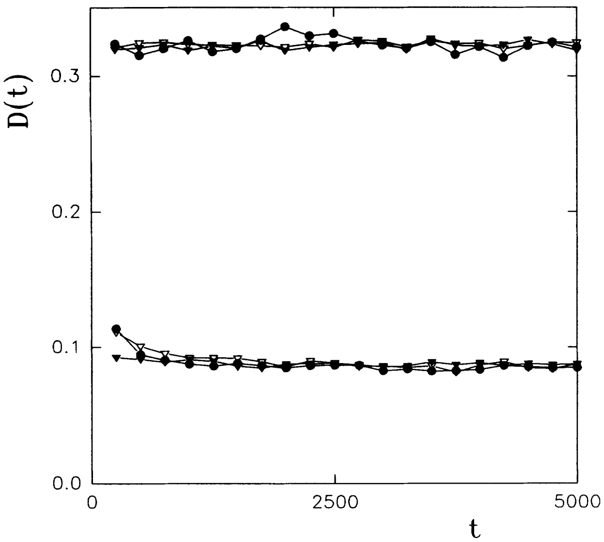

FIG. 1. Plots of $D(t)$ vs t or the ZGB model obtained for two different Y values and various s values: $Y=0.52$, upper curves and $Y=0.3917$, lower curves. ●, $s=0.10$; ∇, $s=0.05$; and ▼, $s=0.01$.

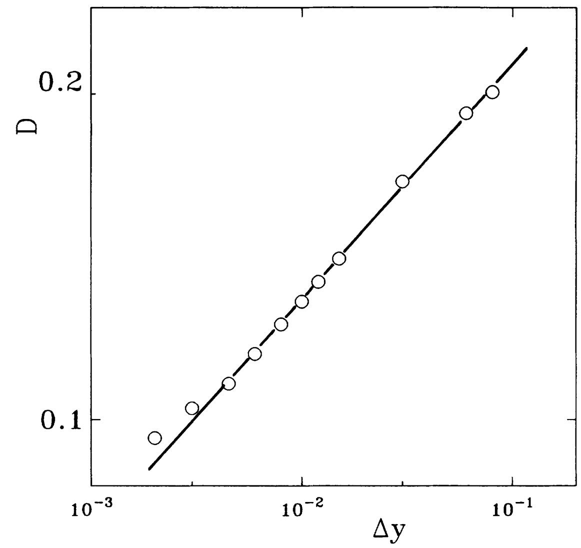

FIG. 3. Log-log plot of $D(t\rightarrow\infty)$ vs ΔY [see Eq. (7)]. Results are obtained using lattices of side $L=100$. Each point is averaged within the interval $2\times10^3 \leq t \leq 5\times10^3$ and over 100 different samples. The straight line with slope $\beta=0.33\pm0.01$ corresponds to the best fit of the data.

transitions. Also, the frozen-chaotic transition at Y_{1c} (Y_{2c}) is continuous (discontinuous), respectively. In spite of the fact that the plot of D versus Y shown in Fig. 2 resembles the dependence of the rate of AB production on Y [1], we have not found any direct relationship between both quantities.

Close to the continuous frozen-chaotic transient the natural order parameter is the damage itself, which is expected to behave as

$$D(t\rightarrow\infty)\propto(Y-Y_{1c})^{\beta}\ , \tag{7}$$

where β is the order parameter critical exponent. Figure 3 shows a log-log plot of D versus $\Delta Y = Y - Y_{1c}$ and

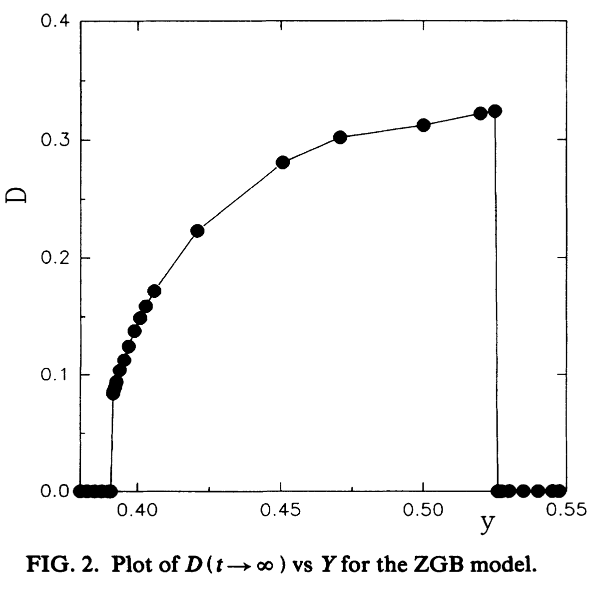

FIG. 2. Plot of $D(t\rightarrow\infty)$ vs Y for the ZGB model.

from the slope very close to Y_{1c}, one gets $\beta \cong 0.12\pm0.02$. However, most points shown in Fig. 3 lie on a straight line with slope $\beta \cong 0.33\pm0.01$, where in both cases the error bars merely reflect the statistical error. Since a small error of measuring Y_{1c} causes quite a large error in β, we have employed the best available value of the critical probability given by $Y_{1c} \cong 0.390(65)\pm0.000(10)$ [8]. In spite of the considerable uncertainties involved in the determination of β, the obtained exponents considerably differ from that of the well know irreversible poisoning transition at Y_{1c} given by $\beta^* \cong 0.58$, i.e., the value corresponding to the Reggeon field theory (RFT) universality class [8]. This finding can be understood considering the contributions to D given by V-A, V-B, and A-B, as discussed above. Since close to Y_{1c} the coverage with A species is almost negligible [1], D is dominated by the V-B term with critical exponent β. On the other hand, for the poisoning transition and close to Y_{1c} the natural order parameter is the concentration of minority species θ_A, which behaves as

$$\theta_A \propto (Y-Y_{1c})^{\beta^*}\ . \tag{8}$$

So, the contribution to the damage given by the term V-A ($\Delta\theta_A$) should be proportional to θ_A and, consequently, it may be dominated by the same exponent [33]. To check this argument we have recorded the term V-A separately (see Fig. 4), which is found to be governed by the exponent $\beta^* \cong 0.578\pm0.010$ [33], in excellent agreement with the RFT value [8].

The fact that both transitions at the same critical point, i.e., the frozen-chaotic and the poisoning-reactive transitions, have different order parameter critical exponents strongly suggests that in the ZGB model, dam-

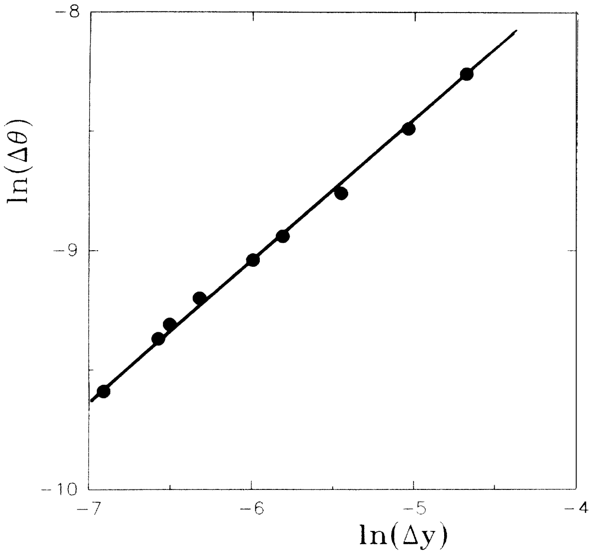

FIG. 4. Plot of $\ln(\Delta\theta_A)$ vs $\ln(\Delta Y)$ [see Eq. (8)]. Results are obtained using lattices of side $L=150$. Each point is averaged within the interval $2\times10^3 \le t \le 5\times10^3$ and over 100 different samples. The straight line with slope $\beta=0.578\pm0.010$ corresponds to the best fit of the data.

age spreading introduces a new dynamic critical phenomenon.

B. ZGBER model in two dimensions

Meakin [9] has reported simulations of the ZGB model with the addition of the Eley-Rideal reaction step, given by Eq. (4). The poisoned state with B species is not observed ($Y_{1c}\to 0$), and the first-order irreversible transition between the reactive regime and the poisoned state with A species becomes shifted to $Y_{2c}\cong 0.4972$.

It is found that for $Y>Y_{2c}$ the damage is quickly healed. So, as expected, the poisoned state belongs to the frozen phase. For $0.05<Y<Y_{2c}$ the damage spreads and after roughly $t\cong 5\times10^3$ reaches a stationary value. However, surprisingly, for $Y=0.01$ the damage becomes healed. So, let us define the spreading critical point Y_s to be the Y value at which the frozen-chaotic transition take place. Close to Y_s we assume the following Ansatz:

$$D(t)=t^{-\delta}F(\Delta Y t^{1/\nu}) , \tag{9}$$

where $\Delta Y = Y - Y_s$. For large t the scaling function should behave as $F(x)\propto x^{\hat\beta}$; δ, ν, and $\hat\beta$ are critical exponents. For $\Delta Y>0$ and $t\to\infty$ one has that $D(t)$ take finite values independent of t, so $D(t)\propto \Delta Y^{\hat\beta}$, and $\hat\beta=\nu\delta$. Note that D is the appropriate order parameter and $\hat\beta$ the associated critical exponent [16]. For $\Delta Y=0$ and $t\to\infty$ the damage should be healed according to a single power-law decay and consequently a log-log plot of $D(t)$ versus t should give a straight line. On the other hand, for $\Delta Y<0$ ($\Delta Y>0$) the curves should veer downward and upward, respectively. This property will allow us to determine both Y_s and δ quite accurately. In fact, Fig. 5 shows log-log plots of $D(t)$ versus t obtained using

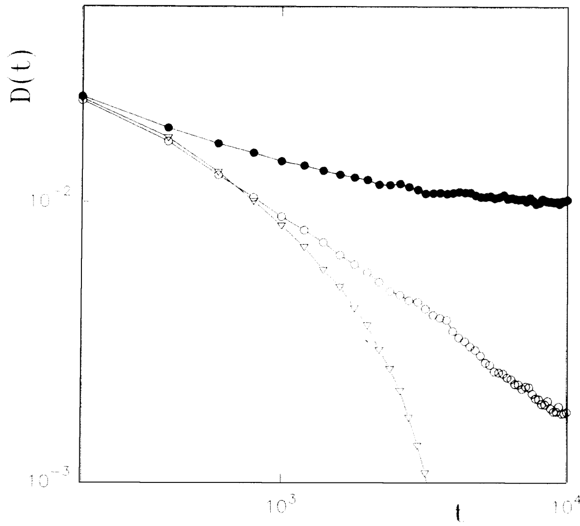

FIG. 5. Log-log plot of $D(t)$ vs t for the ZGBER model in two dimensions. ●, $Y=0.05$; ○, $Y=0.02$; and ∇, $Y=0.01$.

different Y values. Taking $Y=0.05$ one has that the damage becomes almost stable for $t\to10^4$, while taking $Y=0.01$ the damage is healed. However, at $Y=0.02$ the damage decrease according to a single power law, strongly suggesting the validity of Eq. (9) with $Y_s\cong 0.02$. We have checked that the asymptotic behavior of $D(t)$ just at Y_s is independent of the initial distance between the starting configurations, as is shown in Fig. 6. A least squares fit of the data within the asymptotic regime ($t>10^3$) gives $\delta\cong 0.65\pm0.02$, where the error bars are evaluated considering the different δ values obtained for different s values. The fact that the reactive-poisoning transition and the frozen-chaotic transition occur at different criti-

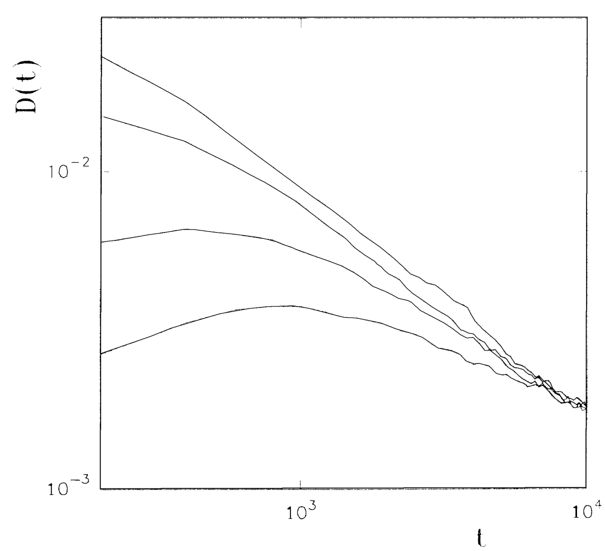

FIG. 6. Log-log plots of $D(t)$ vs t for the ZGBER model in two dimensions obtained at $Y=Y_s=0.02$ and using different s values; from top to bottom, $s=0.10$, $s=0.05$, $s=0.02$, and $s=0.01$.

cal values ($Y_s \neq Y_{2c}$) indicates that damage spreading the ZGBER model also introduces a new dynamic critical phenomenon. It should be noted that the monomer-monomer irreversible reaction process (MMP) with one species desorption exhibits a similar behavior [24]. In fact, the latter has a poisoning-reactive critical point at $Y_c \cong 0.5099$ [34], while the frozen-chaotic transition takes place close to $Y_s \cong 0.745$ [24].

Once Y_s has been determined by evaluating the damage healing kinetics, one can analyze the behavior of the damage within the chaotic phase. A log-log plot of $D(t \to \infty)$ versus ΔY (Fig. 7) confirms the validity of Eq. (9) and a least squares fit of the data gives $\hat{\beta} \cong 1.18 \pm 0.03$ for the order parameter critical exponent. It should be noted that for the MMP in two dimensions one has a quite different exponent, e.g., $\hat{\beta} \cong 0.492 \pm 0.007$ [24].

Also, for the sake of comparison, let us mention that in the case of thermally driven reversible phase transitions it has been reported that the spreading temperature of the Ising model in three dimensions may be shifted about 3% below the critical temperature [31], i.e., a small shift when compared with those observed in both the ZGBER model and the MMP.

In the limit $Y=0$ the ZGBER (and also the ZGB) model corresponds to the well known random dimer filling problem (RDFP) in two dimensions (for a review see [35]). Under this condition a full coverage with B species cannot be achieved and the final state of the surface is jammed, the jamming coverage being $\theta_J \cong 0.907$ [35]. It is clear that any initial damage created in the jammed state cannot be healed and, consequently, for this limiting value of Y the final damage will adopt a nonzero value. So, we have also investigated the process of damage healing for $Y < 0.01$, i.e., the lowest Y value shown in Fig. 5. Figure 8 shows plots of $D(t)$ versus t, taking

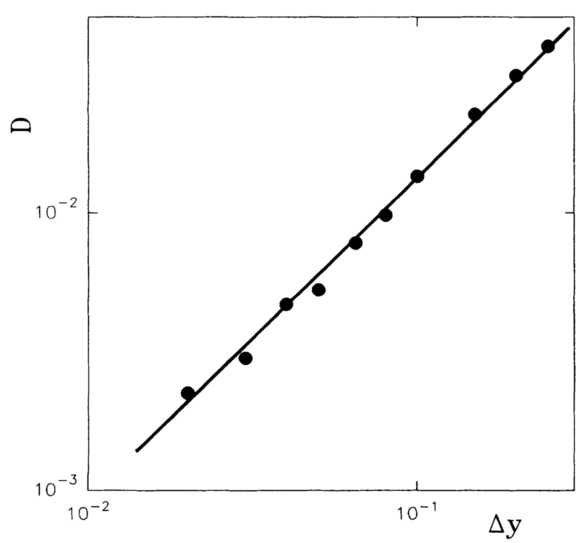

FIG. 7. Log-log plot of $D(t \to \infty)$ vs ΔY [see Eq. (9)]. Results obtained using lattices of side $L=100$. Each point is averaged within the interval $5 \times 10^3 \leq t \leq 10^4$ and over 100 different samples. The straight line with slope $\beta = 1.18 \pm 0.03$ corresponds to the best fit of the data.

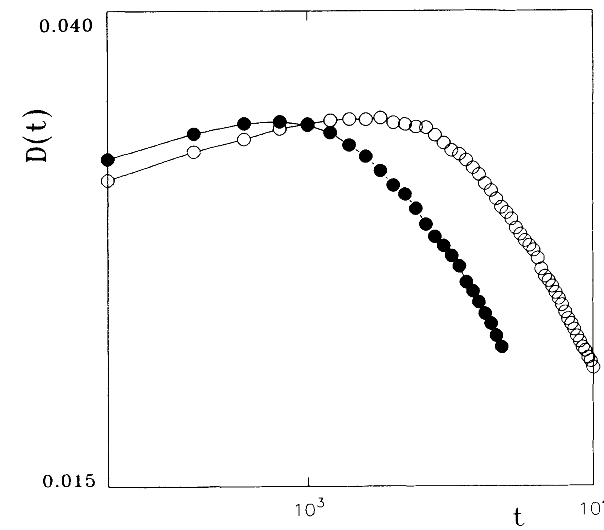

FIG. 8. Log-log plot of $D(t)$ vs t for the ZGBER model in two dimensions. ●, $Y=0.001$; and ○, $Y=0.0005$.

$Y=0.001$ and $Y=0.0005$. On the one hand, one observes that now the damage healing process is slower for the smaller Y value. On the other hand, it becomes evident that also the kinetics of damage healing is much slower than for $Y=0.01$ (Fig. 5). So, the observed behavior is consistent with the fact that for $Y=0$ one has nonzero damage.

C. ZGBER model in one dimension

According to Meakin [9] the addition of the Eley-Rideal reaction step, given by Eq. (4), to the ZGB model in one dimension causes the occurrence of a finite-width reaction window. In fact, the poisoned state with B species is not observed ($Y_{1c} \to 0$) and a continuous ir-

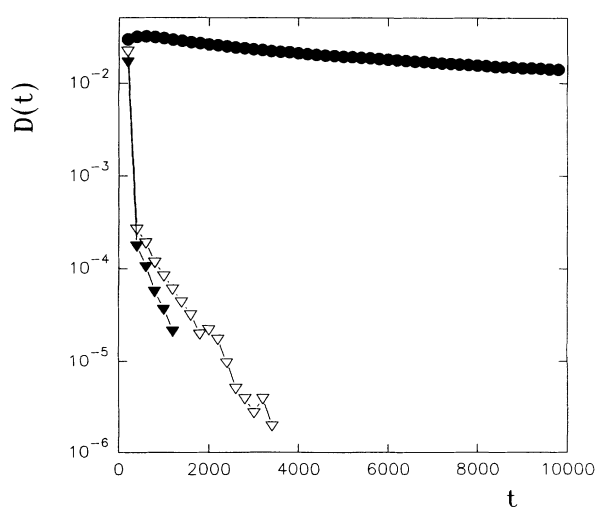

FIG. 9. Plots of $\log\{D(t)\}$ vs t for the ZGBER model in one dimension. ▼, $Y=0.30$; ▽, $Y=0.10$; and ●, $Y=0.01$.

reversible transition between the reactive regime and the poisoned state with A species is found close to $Y_{2c} \cong 0.3058$ [9].

It is found that the damage heals for $Y > 0$, as is shown for same typical Y values in Fig. 9. However, damage healing requires more time when approach $Y \rightarrow 0$. In fact, for $Y = 0.30$ and 0.10 one has damage healing after roughly $t = 1200$ and 3400, respectively, while for $Y = 0.01$ the healing process still continues after $t = 10^4$ (Fig. 9). This behavior can be understood in terms of the same idea discussed for the ZGDER model in two dimensions. That is, for $Y = 0$ one has the RDFP in one dimension and the sample is jammed, the jamming coverage with B species being $\theta_J = 1 - e^{-2}$ [35]. Therefore, also in this case, one has nonzero damage for $Y = 0$.

It is interesting to note that damage healing has also been observed within the reactive regime of the monomer-monomer reaction process with one species desorption in one dimension [24]. This result, together with the above discussed behavior of the ZGBER model, may suggest that damage spreading cannot take place within the reactive regime of irreversible reaction system in one dimension. Preliminary results for the A model (BK model), as defined, e.g., in Refs. [36] and [37], respectively, are in agreement with the above conjecture.

IV. CONCLUSIONS

The spreading of damage is studied in the ZGB and the ZGBER models in one and two dimensions. For the ZGB model in two dimensions, damage spreading (healing) is observed within the reactive regime and poisoned states, respectively. Frozen-chaotic transitions occur at the same critical points as transitions between the reactive regime and the poisoned states. However, the order parameter critical exponents of the second-order transitions are different, pointing out the occurrence of different critical behavior. For the ZGBER model in two dimensions, the frozen-chaotic transition occurs well inside the reactive regime at $Y_s \cong 0.02 < Y_{2c} \cong 0.4972$. Just at Y_s the damage heals according to a single power-law behavior with exponent $\delta \cong 0.65 \pm 0.02$. The order parameter critical exponent is found to be $\hat{\beta} \cong 1.18 \pm 0.03$. The absence of damage spreading in the ZGBER model in one dimension, as well as in other reaction processes in one dimension, leads us to conjecture that this is a common feature characteristic of their reactive regime in one dimension. Further studies aimed at determining the upper critical dimension for damage spreading in single and multicomponent reaction system are in progress.

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