Dynamic scaling behavior of the Ziff-Gulari-Barshad model on regular fractal lattices: The influence of lacunarity

Zhuo Gao[2,*] and Z. R. Yang[1,2]
[1]*CCAST (World Laboratory), P.O. Box 8730, Beijing 100080, People's Republic of China*
[2]*Department of Physics and Institute of Theoretical Physics, Beijing Normal University, Beijing 100875, People's Republic of China*
(Received 27 October 1998)

A surface reaction model, the Ziff-Gulari-Barshad model, is studied on fractal lattices by the Monte Carlo method, and the influence of the lacunarity of the lattice on the dynamic scaling properties of the continuous transition in the model is investigated. The dynamic critical exponents are obtained on different lattices. We find that the transitions in the model on fractal lattices with different lacunarity do not belong to the same universality class. [S1063-651X(99)12403-6]

PACS number(s): 05.50.+q, 82.65.Jv, 64.60.Ht, 61.43.Hv

I. INTRODUCTION

The surface reaction models have attracted great interest in recent years. In 1986, Ziff, Gulari, and Barshad presented a model (the ZGB model) to demonstrate a chemical reaction process of O_2 and CO on the Pt surface [1]. According to this model, when an O_2 molecule (a dimer) contacts the surface, it requires two adjacent empty sites to deposit and dissociate immediately. In addition, a CO molecule (a monomer) requires only one empty site. The nearest-neighbor pairs of O and CO react and form a CO_2 molecule which desorbs immediately. The process can be demonstrated as follows:

$$\mathrm{CO}+*\rightarrow \mathrm{CO}* \quad \text{with probability } p, \tag{1a}$$

$$\mathrm{O_2}+2*\rightarrow 2\mathrm{O}* \quad \text{with probability } 1-p, \tag{1b}$$

$$\mathrm{O}*+\mathrm{CO}*\rightarrow \mathrm{CO_2}\uparrow+2* \quad \text{for nearest neighbors,} \tag{1c}$$

where * denotes an empty site of the surface, and O* and CO* denote the adsorbed O atom and CO molecule, respectively. p is the mole fraction of CO in the gas phase.

Obviously, the ZGB model is less realistic [2]. However, it is easy to study because of its simplicity and shows interesting phenomena in physics. According to the simulations [1], when p is lower than p_1, the system is finally saturated by O; if p is greater than p_2, the surface is eventually fully covered by CO; and if $p_1<p<p_2$, the system falls into a reactive phase. The transition at p_1 is continuous, while at p_2 it is first order. Both transitions have been extensively studied [3–5], and it is shown that the continuous transition at p_1 in ZGB model belongs to the directed percolation (DP) or Reggeon field theory class [4,5].

Motivated by the ZGB monomer-dimer model, other surface reaction models are introduced to investigate the nonequilibrium phase transitions. The simplest one is the monomer-monomer model [6], in which the two reactants are all monomers. It has been found that if desorptions of one species are allowed, the transition in this model is continuous and belongs to the DP universality class [7,8]. The influence of noise on the monomer-monomer model was investigated by Fichthorn, Gulari, and Ziff [9], and Clément, Leroux-Hugon, and Sander have shown that this model can be exactly solved on a Euclidean space of dimension d [10].

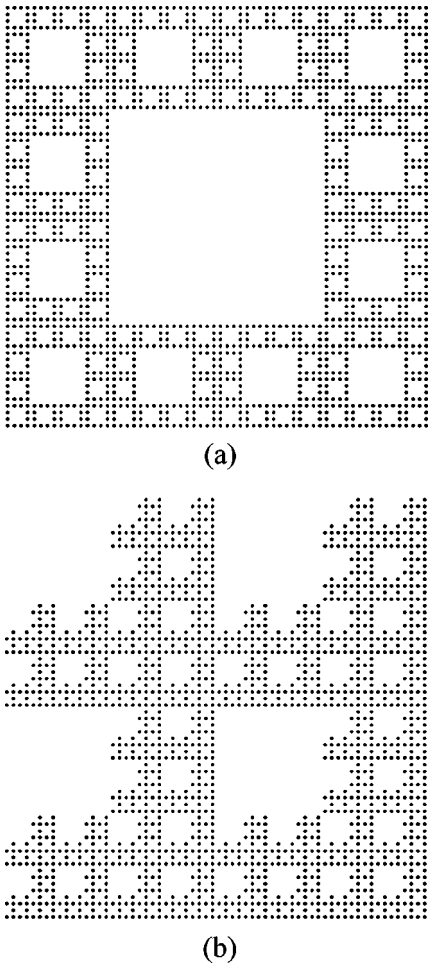

FIG. 1. The third stage of the two Sierpiński carpets with linear dimension of 64: (a) $\mathcal{L}_1$ lattice; (b) $\mathcal{L}_2$ lattice. The dots indicate the sites on the lattices. The bonds connecting the adjacent sites are not shown in the figures.

*Author to whom correspondence should be addressed. Address correspondence to Department of Physics, Beijing Normal University, Beijing 100875, P. R. China.

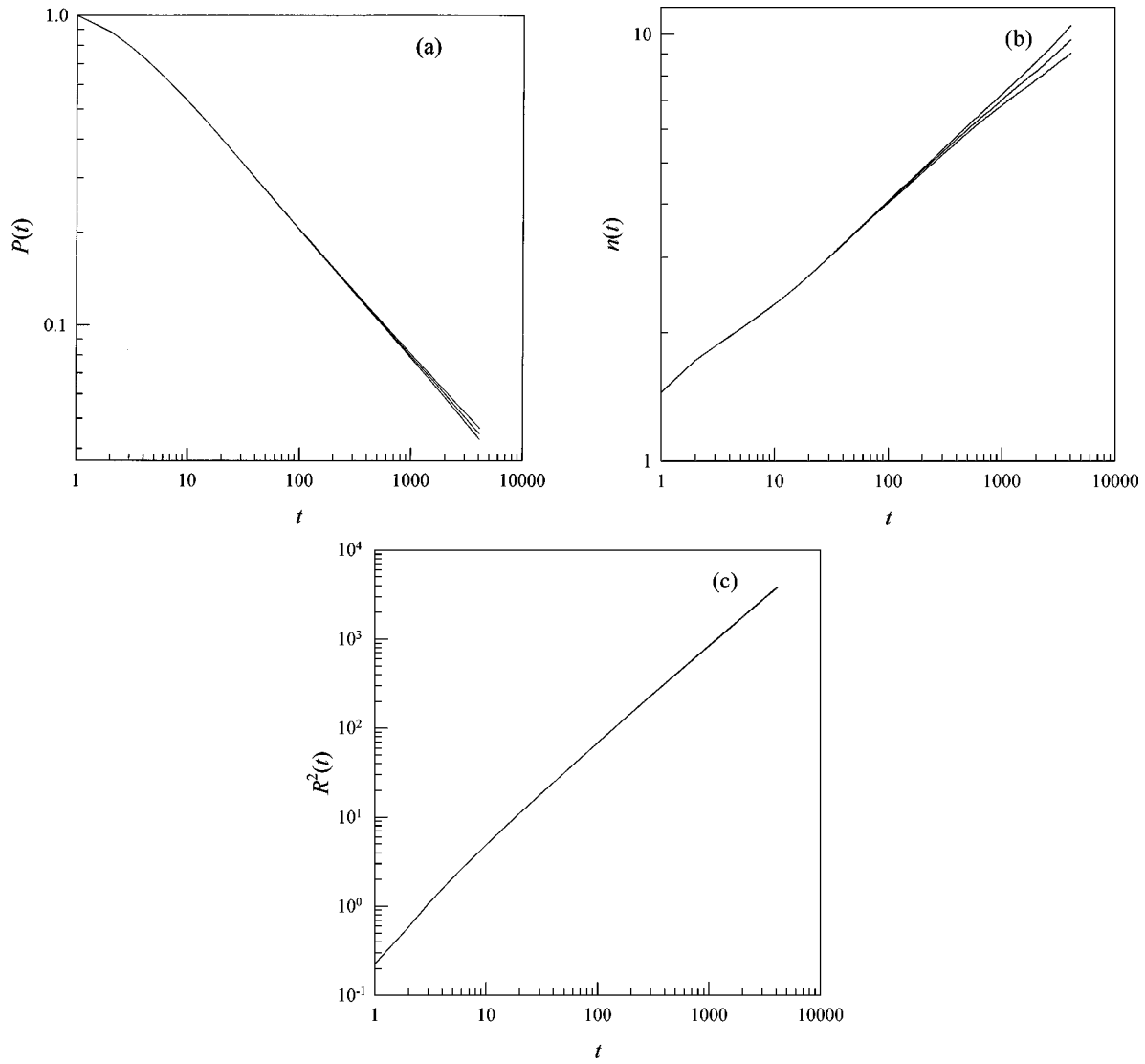

FIG. 2. The behavior of (a) $P(t)$, (b) $n(t)$, and (c) $R^2(t)$ for $\mathcal{L}_1$ with $p=0.4492,0.4491$, and 0.4490 (from top to bottom). In this and the following figures, the unit of time t is a Monte Carlo step (MCS).

Dimer-dimer models have also been studied [11–14]. In the monomer-monomer, monomer-dimer, and dimer-dimer models, only two reaction species are involved and interactions between adsorbing particles are ignored. Recently, a model involving three reaction species was studied by Zhang and Pan, and a complicated phase diagram was found [15]. Park, Kim, and Park studied a monomer-dimer model with repulsive interactions between the same species in one dimension, and found that the continuous transition in this model does not fall into the DP universality class [16,17]. However, it was found that when the symmetry between the adsorbing states is broken by introducing a symmetry-breaking field, the system goes back to the DP class [18].

In fact, real catalytic surfaces are not purely bidimensional and translationally invariant since they may contain defects. Therefore, the reactive substrate may have different geometrical structures. Then a question arises of whether and how the geometrical aspect influences the kinetics and the nonequilibrium transitions? In order to study this problem, it is convenient to treat the catalytic surfaces as fractal lattices. Recently, Clément, Leroux-Hugon, and Argyrakis studied both theoretically and numerically the monomer-monomer model on fractal lattices such as percolation clusters [19]. They found that the structural changes on the lattice strongly affect the reactivity of the catalysis. Before this work, monomer-dimer models have been studied also on percolation clusters [20,21] and Sierpiński-type fractals [22,23] by Monte Carlo simulations, and the influence of the fractal dimension of the lattices on critical points was discussed.

It is well known that the physical properties of fractal lattices are very different from those of translationally invariant lattices. There are several geometric parameters to describe a fractal, such as fractal dimension, lacunarity, and so on. The lacunarity measures the extent of the failure of a fractal to be translationally invariant [24]. It plays an important role in the study of equilibrium phase transitions on fractals [25–28].

In this paper, we study the dynamic scaling behavior of the ZGB model, which is defined as Eqs. (1), on fractal lattices, and investigate the influence of the lacunarity on the nonequilibrium continuous transitions. In order to do so, we use two kinds of Sierpiński carpets, with the same fractal dimension but different lacunarities, to represent the catalytic surfaces. It is found that the lacunarity of the fractal lattice

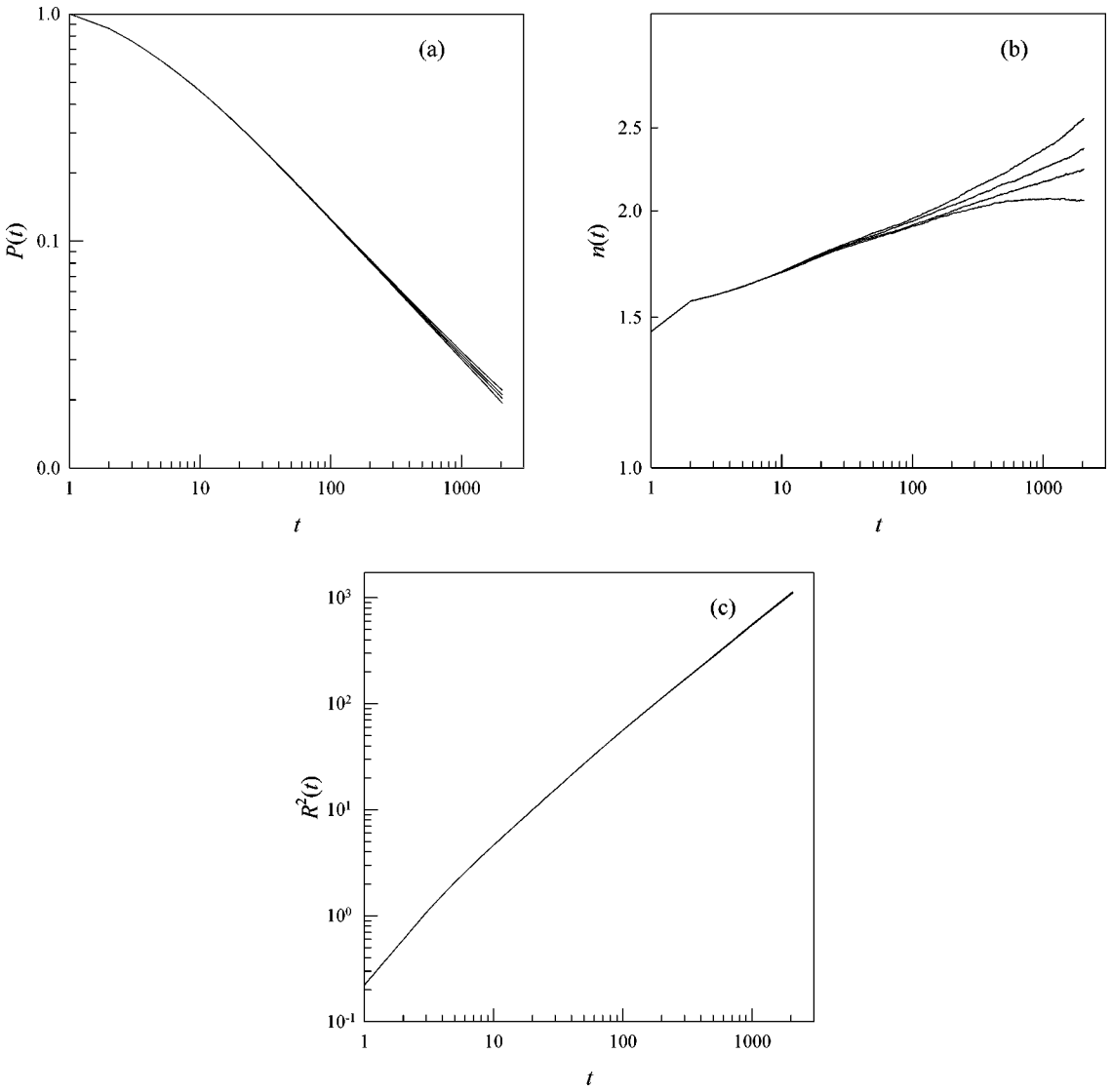

FIG. 3. The numerical results of (a) $P(t)$, (b) $n(t)$, and (c) $R^2(t)$ for $\mathcal{L}_2$ with $p=0.4448,0.4447,0.4446$, and 0.4445 (from top to bottom).

strongly affects the dynamic scaling properties of the continuous transition. According to our numerical results, the dynamical critical exponents of the transitions in the ZGB model are very different on lattices with different lacunarities. This indicates that they belong to different universality classes.

II. THE SIMULATIONS

The Sierpiński carpets are constructed in the following way [25]. Consider a square of unit area and subdivide it into b^2 subsquares, among which l^2 subsquares are cut out. Then one gets the first stage of the structure. By definition, a Sierpiński carpet has the fractal dimension $d_f=\ln(b^2-l^2)/\ln b$. The eliminated subsquares may be condensed (high lacunarity) or scattered (low lacunarity). In our simulations, we use the Sierpiński carpets to serve as the catalytic surfaces, and the lattice sites are considered to be located in the center of the subsquares.

Now we consider two Sierpiński carpets shown in Figs. 1(a) and 1(b), and in the following they are called $\mathcal{L}_1$ and $\mathcal{L}_2$, respectively. $\mathcal{L}_1$ and $\mathcal{L}_2$ have the same fractal dimension, $d_f=\ln 12/\ln 4\approx 1.7925$. However, obviously their lacunarities are different. According to the expression given by Lin and Yang [29], the lacunarities of $\mathcal{L}_1$ and $\mathcal{L}_2$ are 0.1374 and 0.0556, respectively [30]. This implies that $\mathcal{L}_1$ is less homogeneous than $\mathcal{L}_2$, which agrees with the direct observation.

Here we use the epidemic method to study the second-order transitions of the ZGB model on $\mathcal{L}_1$ and $\mathcal{L}_2$. The epidemic method has been used to study the contact process [31], the directed percolation problem in $2+1$ dimensions [32], and the ZGB model on the square lattice [4,5]. In Ref. [5], precise values of p_1 and dynamical critical exponents were obtained.

Initially, the system is completely covered by O atoms except for a single site, randomly chosen [33], which is set to be empty. The periodic boundary condition is considered. In order to make sure that the reactive region is much less than the whole lattice, a large system is necessary. Here the linear dimension of our lattices is taken to be $4^6=4096$. In other words, both fractal lattices are the sixth stage of the structure. (Figure 1 shows the third stage of the fractal structure.)

In the simulations, a site is randomly chosen. If the site is

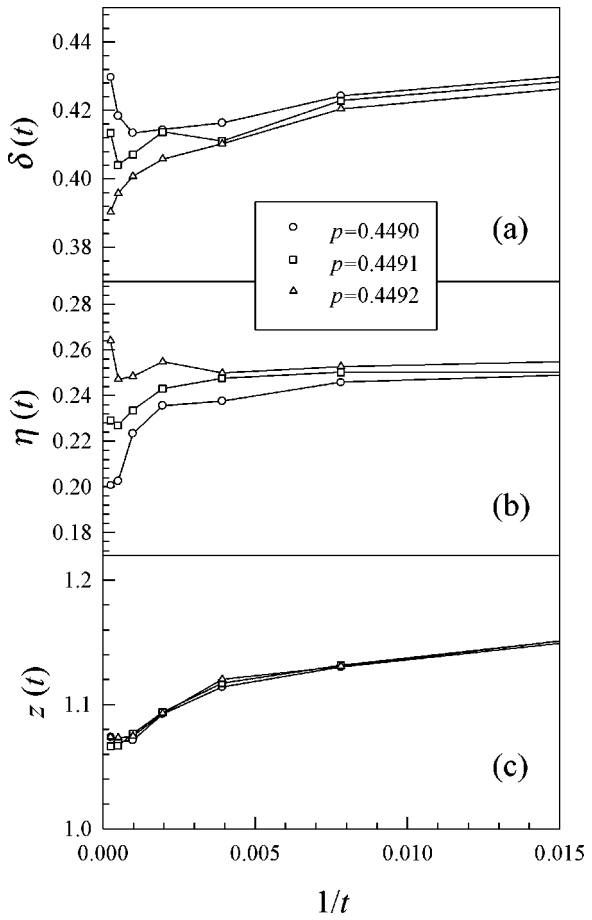

FIG. 4. The local slopes of (a) $\delta(t)$, (b) $\eta(t)$, and (c) $z(t)$ for $\mathcal{L}_1$.

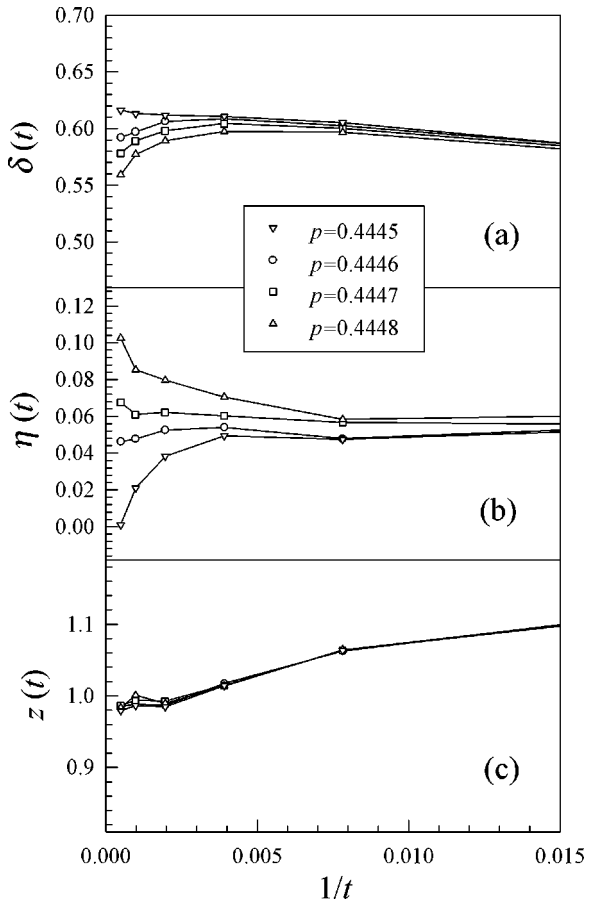

FIG. 5. The local slopes of (a) $\delta(t)$, (b) $\eta(t)$, and (c) $z(t)$ for $\mathcal{L}_2$.

empty, CO adsorbs on it with probability p, while with probability $1-p$, an O_2 molecule is considered. For the latter case, a neighboring site is randomly chosen, and if it is also empty, the O_2 molecule adsorbs and immediately dissociates into two O atoms which occupy the two sites. After a particle adsorbs on the lattice, it checks its neighbors in a random order. If a particle of the opposite species resides in one of its neighboring sites, they react and desorb immediately, leaving two empty sites. In this process, the number n and the distribution of vacant sites change from time to time. n may become greater due to the reactions and less due to the adsorptions.

Three quantities are measured during the simulations [5]: the survival probability $P(t)$, which is the chance that after t time steps there is still at least one vacant site, the average number of empty sites $n(t)$, and the average square radius of gyration of vacant sites $R^2(t)$. Notice that $n(t)$ is the average over all runs, while $R^2(t)$ is averaged over only surviving ones. In our simulations, for lattice $\mathcal{L}_1(\mathcal{L}_2)$, 1 000 000 (2 000 000) runs are performed up to 4096 (2048) time steps.

At the critical point, the three quantities are expected to follow the scaling laws [31]

$$P(t)\sim t^{-\delta}, \tag{2a}$$

$$n(t)\sim t^{\eta}, \tag{2b}$$

$$R^2(t)\sim t^{z}, \tag{2c}$$

where δ, η, and z are dynamical critical exponents. From these equations, the critical point p_1 can be determined since on the log-log plots these relations show a straight line at the critical point, while otherwise they deviate from the straight lines.

Results for the three quantities $P(t), n(t)$, and $R^2(t)$ for the $\mathcal{L}_1$ lattice are shown in Fig. 2, from which one can obtain $p_1^{(\mathcal{L}_1)}=0.4491\pm0.0001$. Similarly, the same quantities for the $\mathcal{L}_2$ lattice are shown in Fig. 3, and $p_1^{(\mathcal{L}_2)}=0.4447\pm0.0001$. Comparing between the two figures, one can see that the critical behavior of the ZGB model on $\mathcal{L}_1$ and $\mathcal{L}_2$ is distinctly different.

In order to estimate the dynamical critical exponents, the local slopes should be considered,

$$-\delta(t)=\frac{\ln[P(t)/P(t/d)]}{\ln d}, \tag{3}$$

and similarly for $\eta(t)$ and $z(t)$. These local slopes are shown in Fig. 4 and Fig. 5 with $d=2$ for $\mathcal{L}_1$ and $\mathcal{L}_2$, respectively. In general the local slopes can be expanded as [31,32]

$$\delta(t)=\delta+\frac{a}{t}+\frac{\delta' b}{t^{\delta'}}+\cdots, \tag{4}$$

and similarly for $\eta(t)$ and $z(t)$. From Figs. 4 and 5, extrapolating the local slopes to $t\to\infty$ and considering Eq. (4), we obtain

$$\delta^{(\mathcal{L}_1)}=0.41\pm0.02, \tag{5a}$$

$$\eta^{(\mathcal{L}_1)}=0.22\pm0.02, \tag{5b}$$

$$z^{(\mathcal{L}_1)}=1.05\pm0.02, \tag{5c}$$

and

$$\delta^{(\mathcal{L}_2)}=0.60\pm0.02, \tag{6a}$$

$$\eta^{(\mathcal{L}_2)}=0.05\pm0.02, \tag{6b}$$

$$z^{(\mathcal{L}_2)}=0.97\pm0.05. \tag{6c}$$

From Eqs. (5) and (6), $\delta^{(\mathcal{L}_2)}$ is greater than $\delta^{(\mathcal{L}_1)}$, and $\eta^{(\mathcal{L}_2)}$ and $z^{(\mathcal{L}_2)}$ are less than $\eta^{(\mathcal{L}_1)}$ and $z^{(\mathcal{L}_1)}$.

III. DISCUSSION

In the preceding section, the second-order transition in the ZGB model is studied on two Sierpiński carpets by the Monte Carlo method. These two lattices have the same fractal dimension but different lacunarities. The critical points and the dynamical critical exponents are obtained for the two cases. From the numerical results, one can see that both the critical points and the exponents of the two cases are clearly different. This implies that the transitions in the two cases, even though the lattices have the same fractal dimension, belong to different universality classes. Thus we can say that the lacunarity of the catalytic surface may have a great influence on the universality class of the continuous transition in the ZGB model.

In the problems of equilibrium phase transitions, it is clear that the transitions are dependent upon the detailed structures of lattices, and such dependence may be complicated (e.g., see [26–28]). According to our results, the geometric factors of the lattice may also play important roles in the nonequilibrium problems. How do the nonequilibrium transitions depend on these factors, and which parameters are crucial in determining the critical properties? These still need further investigations.

ACKNOWLEDGMENTS

One of the authors (Z.G.) would like to thank Professor J.-y. Zhu for his helpful discussions. This work is supported by the National Basic Research Project ''Nonlinear Science,'' the National Nature Science Foundation of China, and the State Education Department Grant for Doctoral Study.

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[30] For the concept of lacunarity, it was originally proposed by B. B. Mandelbrot for explaining the galaxy distributions in his famous book [B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982)]. Then lacunarity was introduced to describe the degree of inhomogeneity of a fractal in fractal geometry. The lacunarity of a fractal which is nearly translationally invariant should be small, and vice versa. So far, a general and exact expression of lacunarity has still not been found. However, for a kind of fractal, such as Sierpiński carpets, several approximate expressions have been suggested. These expressions may measure the extent of deviation of Sierpiński carpets of different structure from translationally invariant lattices. For a recent review on lacunarity, see D. A.

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