Effects of biased diffusions on dynamical surface structures for the $A+B\rightarrow 0$ reaction

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The dynamical scaling property of surface eroded by a chemical reaction $A+B\rightarrow 0$ is studied by the simulation. To consider the effect of interactions between an A particle and the material which consists of B particles, the A particle is assumed to undergo a drifted-diffusive motion or a biased random walk before it touches the material. The surface of the material is eroded by the chemical reaction with a B particle which the A particle first encounters. In dimension $d=2$, we found three regimes in the dynamical surface structure. When there is attractive bias to the material, the dynamical scaling property belongs to the universality class with the dynamic exponent $z=2$. When there is no bias or relatively small repulsive bias, the scaling property belongs to the class with $z=1$. The surface roughening behavior disappears when repulsive bias becomes quite large. We also discuss the properties of the crossover between the existing regimes.

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I. INTRODUCTION

It is very important to physically understand the dynamical evolution of the surface that is eroded by chemical or physical reactions between external particles (A particles) and host particles (B particles) of a given material, because such evolution can be the key process for the electrolytic polishing, corrosion, etching, stable fluid invasion [1–4], chemical processes mediated by a catalytic particle [5–7], etc. Until now there have been two kinds of suggested theoretical models for such erosion phenomena. In the first kind of models [5,6], the particle A is assumed to be a catalytic particle or a random walker that can have a strong annihilation potency to be capable of annihilating many B particles before it disappears. In this kind of model, the B particles inside the material as well as those on the surface can be annihilated and the main interest is focused on the structure of the eroded cluster. In the second kind of models [2–4], each time an A particle encounters a B particle, both particles simultaneously disappear through a reaction like $A+B\rightarrow 0$. Then, the reaction in the second kind of models occurs almost entirely at the surface. The incoming particle A is then effectively screened out from the interior of the material, which should remain compact. So in this kind of model the surface or interface of the compact material can be defined rather keenly, and the dynamical evolution of surface should become the main interest. The dynamical scaling theory for kinetic roughening of surface [8] can then be useful for the analysis of the surface evolution in the second kind of models.

Among the second kind of models, one of the most interesting models is diffusion-limited erosion (DLE) [2,4], which is a time-reversed process of diffusion-limited deposition (aggregates) [9]. This model is theoretically as well as experimentally important, because DLE was found to be a good model to explain a stable Laplacian front [2,3]. DLE is also the only significant stochastic discrete model which has been proved to follow the linear growth equation with dynamic exponent $z=1$,

$$\frac{\partial h_q(t)}{\partial t}=-\nu|\mathbf{q}|^z h_q(t)+\eta_q(t)(z=1). \tag{1}$$

Here $h_q(t)$ is the Fourier component of the surface height $h(\mathbf{x},t)$, and $\eta_q(t)$ is the Fourier component of Gaussian white noise with $\langle \eta_q(t)\rangle=0$ and $\langle \eta_q(t)\,\eta_{q'}(t)\rangle=D\,\delta_{qq'}\delta(t-t')$. Other linear growth equations with $z=2$ or $z=4$, and discrete growth models related to such growth equations have been extensively studied throughout the last decade [8], so that the study on Eq. (1) with $z=2$ and $z=4$ has almost been completed. In contrast, few studies on Eq. (1) with $z=1$ have been done yet. The surface width (or the root-mean-square fluctuation of the surface heights) W described by Eq. (1) in the substrate dimension $d_s=1$ satisfies the relation [2,3]

$$W^2=\frac{D}{2\pi\nu}\{\ln(L/a)+\ln[1-\exp(-4\pi\nu t/L)]\}$$
$$=W_\infty^2+f(t/L), \tag{2}$$

instead of the usual dynamical scaling $W(L,t)=L^\alpha f(t/L^z)$ $(z=\alpha/\beta)$ [8], since the marginal dimension d_s^c of DLE is $d_s^c=1$. Of course the roughness exponent $\alpha=0$ and the growth exponent $\beta=0$ in $d_s=1$.

Even though DLE is a good model for erosion phenomena, the motion of an A particle is constrained to an unbiased random walk or a simple diffusion. But in reality as in electrolytic polishing, the A particle can be driven to an electrode [10] or be repelled from the material [6]. So we need some theoretical models to describe the possible bias effects for the erosion phenomena, but there have been a few models to consider such effects. In the first kind of models, the effect of a sort of repulsive biases on the structure of the eroding cluster was considered [6]. For the second kind of models, there have been no significant works on the effect of bias or drift except for the theoretical analysis of the effect on the

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Laplacian fronts [2]. For the repulsive bias [2], the logarithmic roughness as in Eq. (2) was shown to be limited to a length scale $l<l_u$ [2], where $l_u=D/|u|$ and u is the drift velocity. For the attractive bias, the local diffusion [or Edwards-Wilkinson (EW) [11,12]] term $\nabla^2 h$ was argued to be the main term of the continuum equation for the Laplacian front in the length scale $l<l_u$ [2]. However, in the large length scale Kardar-Parisi-Zhang (KPZ) [8,13] equation was argued to be the continuum equation for the Laplacian fronts with the attractive bias [2]. Even though there existed the analysis on the effect of the bias on the Laplacian front, the analysis has not been seriously compared to the results of relevant simulations. Furthermore even though the Laplacian front was proved to be related to DLE, the Laplacian front itself cannot directly be a model of the second kind. The systematic studies for the effect of the interactions between the A particle and the material on the scaling property of the second kind of models themselves have not been done yet. The first motivation of this study is thus to investigate the effect of both attractive and repulsive biases on the dynamical scaling properties of the second kind of models themselves. These studies will be done through the systematic establishment of simple stochastic discrete models which take the interactions between the A particle and the material into consideration. We also hope that some of the bias effects predicted by the theoretical analysis [2,3] on the Laplacian fronts can be seen through our model. As explained when introducing DLE model, the studies on the physical properties of Eq. (1) with $z=1$ have been very rare until now. It is thus our second motivation to understand the physics of Eq. (1) with $z=1$ to the more profound level through the established models.

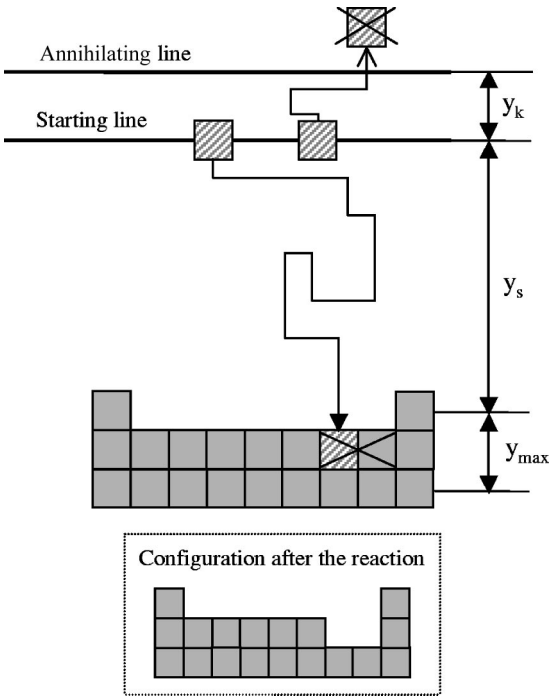

FIG. 1. Schematic diagram for the model in $d_s=1$. It is shown that an A particle (the slashed square) starts from a site on the starting line and undergoes the biased random walk with the hopping probability assignment in Eqs. (3) and (4). The A particle arrives at a simultaneous nearest neighbor site to three B particles (gray squares). A randomly chosen B particle among three and the A particle disappear through the reaction $A+B\rightarrow 0$. It is also shown that an A particle which arrives at the annihilating line is abandoned.

II. MODEL

We now explain the details of our model, which will be investigated for the motivations of the present study. Figure 1 presents the schematic description of our model in $d_s=1$. An A particle starts from a random site on a starting line, which is y_s units higher than the highest surface site occupied by a B particle. The A particle undergoes a biased random walk on a square lattice under the following hopping probability assignment. The hopping probabilities of the particle at a site (x,y) to the nearest neighbor sites $(x,y-1)$, $(x,y+1)$, $(x-1,y)$, and $(x+1,y)$ are assigned as

$$P(x,y-1)=P_b \quad (0\leqslant P_b\leqslant 1), \tag{3}$$

$$P(x-1,y)=P(x+1,y)=P(x,y+1)=(1-P_b)/3. \tag{4}$$

When the A particle reaches a nearest neighbor site to a B particle, then both A and B particles disappear by the reaction $A+B\rightarrow 0$. If the particle reaches a site which is a simultaneous nearest neighbor site to more than one B particle, then one of the B particles is chosen randomly for the reaction. After each reaction process, the newly formed surface sites are identified. Then another A particle is launched from a site on the starting line. For the acceleration of the processes, the A particle which reaches the annihilation line is abandoned and a new particle starts from the given starting line. The annihilation line is y_k units higher than the starting line. The periodic boundary conditions are used in the lateral direction throughout the simulation. When $P_b>1/4$, the A particle experiences the attractive bias to the material. When $P_b<1/4$, the A particle then experiences the repulsive bias from the material. When $P_b=1/4$, the A particle remains unbiased and the model becomes nearly the same one as DLE except for one subtle difference. In the DLE model [4] the reaction $A+B\rightarrow 0$ occurs if the A particle hops onto a site occupied by a B particle, but in our model the reaction occurs when the A particle reaches a nearest neighbor site to one occupied by a B particle. However, we have seen that this subtle difference does not make any difference physically. (See also Sec. III.) When $P_b=1$, then the particle does the ballistic motion and the model reproduces the ballistic erosion, which is the time-reversed process of the ballistic deposition [14]. Recent studies on the ballistic erosion [6,15] showed that the scaling property of the ballistic erosion belongs to the EW universality class [8,11,12].

Instead of the probability assignment in Eqs. (3) and (4), one can take another assignment as

$$P(x,y-1)=P_b, \quad P(x,y+1)=1/2-P_b \quad (0\leqslant P_b\leqslant 1/2), \tag{5}$$

$$P(x-1,y)=P(x+1,y)=1/4. \tag{6}$$

The assignment in Eqs. (5) and (6) is more conventional, because the steps along the x (lateral) direction are random but steps along the y (vertical) direction to surface are biased. If the assignment in Eqs. (5) and (6) is taken, the parameter P_b can vary in the range $0 \leqslant P_b \leqslant 1/2$. In contrast with the assignment in Eqs. (3) and (4), P_b can vary in $0 \leqslant P_b \leqslant 1$, and then our model can cover the ballistic erosion by setting $P_b = 1$. That is why we take the assignment in Eqs. (3) and (4). However, we have confirmed that in the common range $0 \leqslant P_b \leqslant 1/2$, the simulation results for the assignment in Eqs. (5) and (6) coincide with those for the assignment in Eqs. (3) and (4) within one standard error limit. (See Fig. 4.)

III. SIMULATION RESULTS

We have performed the simulations for the models with various P_b. The simulations are started from the flat surface with $y = y_i$, where the sites with $y \leqslant y_i$ are all occupied by B particles and those with $y > y_i$ are vacant. In the simulation y_s is taken as $y_s \geqslant L/4$, where L is the lateral size of the material, and y_k is set as $y_k = 2y_s$. In DLE [4], y_s was taken a few lattice units, because the A particle undergoes an unbiased random walk. In contrast, we take much larger y_s in our simulations, so that the A particle can have enough time to see the bias effect before reaching a site for the reaction. All the numerical data in this paper are taken after averaging over more than 100 independent runs.

We now explain the results of the simulations. For the comparison to DLE model [2,3], we first want to explain the results for the model with $P_b = 1/4$ briefly. W for $P_b = 1/4$ satisfies Eq. (2) or the scaling behavior with $z = 1$ very well as in DLE. (See the upper inset of Fig. 5.) So the subtle difference between our model with $P_b = 1/4$ and DLE [4], which was pointed out when defining the model, makes no difference in the dynamical scaling property.

Next, let us explain the results for the models with $P_b > 1/4$ (the attractive bias). Figure 2 shows the results for $P_b = 0.26$, i.e., for the model with a very small attractive bias. In Fig. 2, the early time behavior of $W(t)$ [or the data for $W(t \ll L^z)$] for the lateral size of the material $L = 1024$ is shown. $W(t)$ satisfies the usual power-law behavior $W(t) \simeq t^{\beta}$ [8] with $\beta = 0.100(2)$ instead of the scaling relation (2). In the inset of Fig. 2, W's in the saturation regime or $W(L, t \to \infty)$'s for $L = 32$, 64, 128, 256 are displayed. The data for $W(L, \infty)$ also satisfy the usual kinetic roughening behavior [8], $W(t \gg L^z) \simeq L^{\alpha}$ with $\alpha = 0.18(1)$. Estimated dynamic exponent $z(= \alpha/\beta)$ from these results is equal to 1.8(1), which is much larger than $z = 1$ from Eq. (2) [2,3]. The result $z \simeq 1.8$ for $P_b = 0.26$ is more close to $z = 2$ of EW value and considerably larger than $z = 3/2$ of KPZ value [8,13]. Next to show the effect of the relatively large attractive bias, the results for $P_b = 0.73$ are displayed in Fig. 3. The data of $W(t \ll L^z)$ satisfy $W \simeq t^{\beta}[\beta = 0.262(3)]$. The result $\beta \simeq 0.26$ is very close to $\beta = 1/4$ which is the exact value from EW equation. Furthermore as is displayed in the inset of Fig. 3, $W(t \gg L^z)$ satisfies $W \simeq L^{\alpha}[\alpha = 0.51(1)]$. This result $\alpha \simeq 0.51$ is very close to EW value ($\alpha = 1/2$). The estimated numerical values for the exponents α, β, and z for various $P_b \geqslant 0.26$ are displayed in Fig. 4. All the estimated

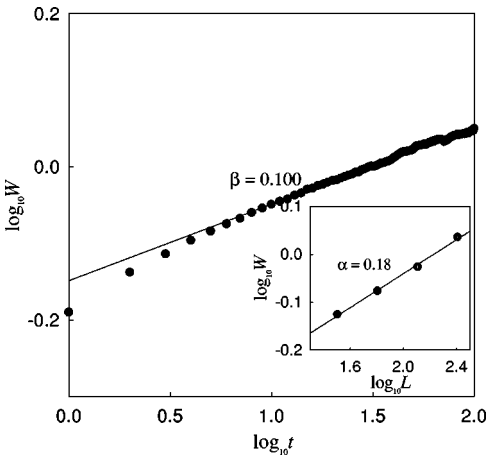

FIG. 2. Log-log plot of W of the model with $P_b = 0.26$ against t in the early time regime ($t \ll L^z$). Used lateral size of the material is $L = 1024$. The solid line denoted by $\beta = 0.100$ shows that W satisfies the power law $W \simeq t^{\beta}$ with $\beta = 0.100(2)$, even for the small attractive bias. The inset shows the log-log plot of W of the same model against L in the saturation regime ($t \gg L^z$). The solid line denoted by $\alpha = 0.18$ represents the relation $W(t \gg L^z) \simeq L^{\alpha}$ with $\alpha = 0.18(1)$. Used lateral sizes are $L = 32$, 64, 128, 256. The estimated value of the dynamic exponent z from α/β is 1.8(1), which is much larger than $z = 1$, the value for the unbiased model.

values of z for $P_b \geqslant 0.27$ are nearly equal to 2. The estimated z is rapidly increasing and approaching $z = 2$ as P_b increases from 0.25. Even for the very small attractive bias as $P_b = 0.26$, z is very much larger than $z = 1$ for the unbiased model or $P_b = 0.25$. Furthermore for the models with $P_b > 0.3$, all the estimated α's and β's satisfy $\alpha > 0.45$ and $\beta > 0.22$. The estimated α and β are also rapidly increasing and approaching the EW values ($\alpha = 0.5$ and $\beta = 0.25$) as P_b increases from $P_b = 0.25$. From the results in Figs. 2–4, it is concluded that the models with the attractive bias belong to EW universality class. Even for the models with small attractive biases ($P_b = 0.26$ or 0.27), the scaling behavior is physically quite different from the DLE model. Taking the finite-size effects into consideration, we can conclude that the crossover behavior from the universality class with $z = 1$ to that with $z = 2$ is very sudden if P_b is increased from 1/4. The KPZ behavior cannot be found in our model with the attractive bias.

To see the effect of the repulsive bias, the numerical results for the models with $P_b < 1/4$ are now explained. In Fig. 5, we display the numerical data of $W(L, t)$ for the models with $P_b = 0.2$. The lower inset shows the raw data for W^2. The main figure shows that W^2 for various L collapses quite well onto the single curve which represents the scaling function (2) with $z = 1$. This behavior of the scaling collapses to Eq. (2) for $P_b = 0.2$ is the same as that for the unbiased model with $P_b = 0.25$. (See the upper inset of Fig. 5.) The scaling behavior for $P_b = 0.25$ is the same as that of DLE [2,3], which we have already explained. We confirmed that the models for $0.18 \leqslant P_b \leqslant 0.25$ show nearly the same scaling behavior as the model with $P_b = 0.2$ and $P_b = 0.25$. In contrast, we found that the models with $P_b \leqslant 0.16$ do not show any surface-roughening behavior. This result means that the

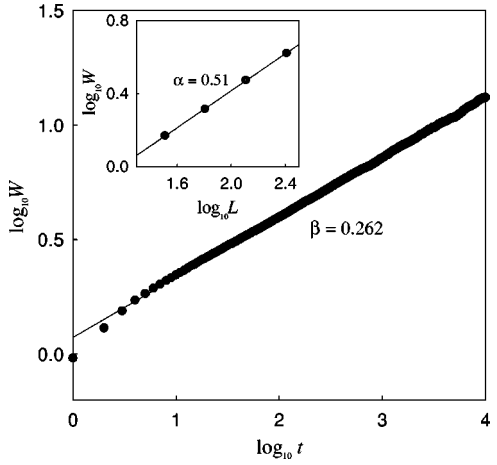

FIG. 3. Log-log plot of W of the model with $P_b=0.73$ against t in the early time regime ($t \ll L^z$). Used lateral size of the material is $L=1024$. The solid line denoted by $\beta=0.262$ shows that W satisfies the power law $W \simeq t^\beta$ with $\beta=0.262(3)$. The inset shows the log-log plot of W of the same model against L in the saturation regime ($t \gg L^z$). The solid line denoted by $\alpha=0.51$ represents the relation $W(t \gg L^z) \simeq L^\alpha$ with $\alpha=0.51(1)$. Used lateral sizes are the same as those in Fig. 2.

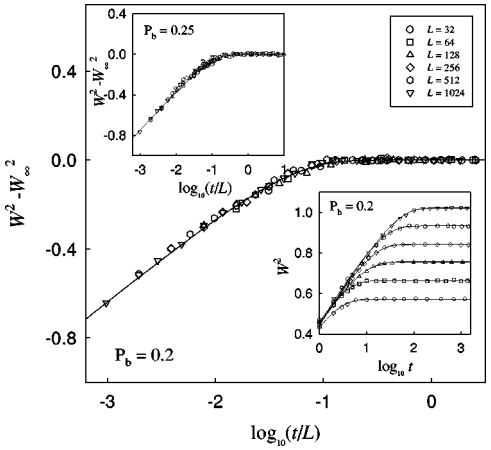

FIG. 5. Scaling plot showing that the data for $W^2-W_\infty^2$ plotted against $\log_{10}(t/L)$ for various L collapse to a single curve supporting the scaling function (2). The data used are taken by using the model with $P_b=0.2$. The used raw data for W^2 are displayed in the lower inset. The upper inset shows the data for the unbiased model with $P_b=0.25$ also collapse to nearly the same single curve. The results in this figure show that the scaling behavior of the models with a considerable range of the repulsive bias is that with $z=1$.

models for the considerable range of the repulsive bias show the scaling behavior (2) with $z=1$ or follow the linear equation (1) until the repulsive bias becomes large to make the surface unroughened. The crossover from $z=1$ behavior to that of the smooth surface occurs around $P_b=0.17$. The crossover range around $P_b=0.17$ was found to be broad.

IV. SUMMARY AND DISCUSSION

In this paper we have introduced a model for the dynamical evolution of the surface eroded by the reaction $A+B \rightarrow 0$ between an A particle, which undergoes a biased random walk and approaches the material from outside, and a B particle on the surface of the material. The scaling properties

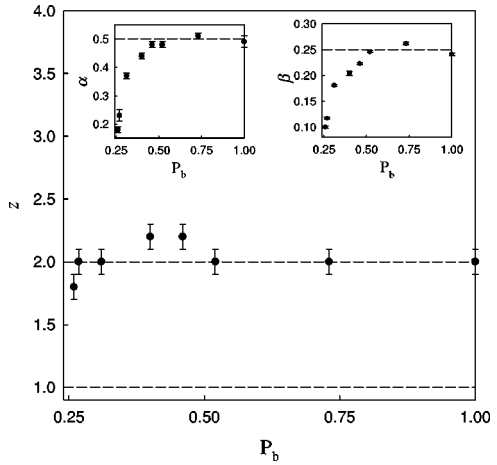

FIG. 4. Estimated values of the exponents z, α, β for various $P_b>1/4$. The rapid increase of the exponents to the values of EW behavior means that the sudden crossover from $z=1$ behavior to EW behavior at $P_b=1/4$.

of the model which have been investigated by the simulations are summarized as what follows. The models with $P_b>1/4$ (or with attractive bias) belong to EW universality class with $z=2$, regardless of the magnitude of the bias factor $|P_b-1/4|$. In contrast, the models with $0.18 \leqslant P_b \leqslant 0.25$ (or with relatively small repulsive bias) still belong to the universality class with $z=1$. The models with large repulsive bias (or $P_b \leqslant 0.16$) do not show any roughening behavior. Thus the models show three different scaling behaviors, smooth (or no roughening) regime for large repulsive bias, the regime with $z=1$ for the relatively small repulsive bias or no bias, and the EW regime with $z=2$ for attractive bias. The crossover from the regime with $z=1$ to that with $z=2$ was found to be sharp at $P_b=1/4$, but the crossover range from the regime with $z=1$ to the smooth phase which occurs around $P_b=0.17$ was found to be broad.

As was discussed earlier, Krug and co-worker [2,3] predicted that the roughness of the Laplacian front with repulsive bias shows the scaling behavior (2) in the length scale $l<l_u=D/|u|$. This prediction physically means that the scaling behavior (2) can be seen in a certain range of the repulsive bias. This prediction is thus consistent with our simulation result that the models with $0.18 \leqslant P_b \leqslant 0.25$ show the scaling behavior (2). In our model the drift velocity u satisfies $|u| \propto |P_b-1/4|$ and l_u $(=D/|u|)$ decreases as P_b decreases from 1/4. If the analysis of the Laplacian front can be applied to our model, then a critical lateral length scale L_c for a given $P_b<1/4$ is expected to exist, so that the scaling behavior (2) cannot be seen for the surface with $L>L_c$. Then the crossover range of P_b from the regime with $z=1$ to the smooth phase should be that in which L_c satisfies $L_{min}<L_c<L_{max}$, where $L_{min}(L_{max})$ is the minimal (maximal) lateral length among L's considered in the simulations. When $L_{min}<L_c<L_{max}$ for a certain P_b, then the scaling behavior (2) can be seen only on the surface with $L<L_c$. This could

be the reason that the crossover range from the regime with $z=1$ to the smooth phase is broad. We thus conclude that the physics related to the repulsive bias to the Laplacian front is qualitatively the same as that for our model with the repulsive bias.

For the attractive bias in the analysis of Laplacian front, the EW behavior was predicted for the relatively small length scale, but the KPZ-type nonlinear equation was argued to be the continuum equation for the front in the large length scale regime [2]. In contrast, we have only seen the EW behavior for the models with the attractive bias. In DLE, the $\mathcal{A}$ particle does the unbiased random walk and thus the probability that the $\mathcal{A}$ particle hits the protruded part of the surface is higher than the flat part. This global nature of noises in DLE make the surface relatively flat and show $z=1$ behavior. In contrast, the $\mathcal{A}$ particle with the attractive bias does the motion with ballistic character and the range along the lateral (or x) direction which the $\mathcal{A}$ particle visits before hitting the surface is less broad. This local nature of the attractively biased model can make the model behave like ballistic erosion model [6,15], which shows EW behavior. This localness from the attractive bias is the physical origin of the EW behavior. In the renormalization group (RG) sense it can be said that the attractive bias is a relevant parameter, which makes the RG flow away from the unbiased (DLE) fixed point to the ballistic erosion fixed point. The local nature of the model with the attractive bias might be due to the simplicity of our model, because discrete models like ours cannot correctly convey the physical factors like hydrodynamic continuum effects, which the theory for the Laplacian front is based on. For the more realistic model which considers the possible nonlinear term in the second kind of models, we recently restudied the erosion phenomena as in DLE by the use of the time-reversed process to that in the dielectric breakdown model [16] and found the KPZ term [17].

ACKNOWLEDGMENTS

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