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Anomalous kinetics of attractive $A+B\rightarrow 0$ reactions

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We investigate the kinetics of the $A+B\rightarrow 0$ reaction with the attractive interaction between opposite species in one spatial dimension. The attractive interaction leads to isotropic diffusions inside segregated single species domains, and accelerates the reactions of opposite species at the domain boundaries. At equal initial densities of A and B, we analytically and numerically show that the density of particles (ρ), the size of domains (ℓ), the distance between the closest neighbor of same species (ℓ_{AA}), and the distance between adjacent opposite species (ℓ_{AB}) scale in time as $\rho\sim t^{-1/3}$, $\ell_{AA}\sim t^{1/3}$, and $\ell\sim\ell_{AB}\sim t^{2/3}$, respectively. These dynamical exponents define critical behavior distinguished from the class of uniformly driven systems of hard-core particles.

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The irreversible two-species annihilation reaction $A+B\rightarrow 0$ has been intensively and widely investigated as a basic model of various phenomena in physics [1,2], chemistry [3], and biology [4]. The reaction starts from the configuration in which two species are distributed randomly in space. The reaction instantaneously takes place with a rate k when two particles of opposite species encounter on the same site (generally within a reaction radius) during the motion of particles. The reaction forms the third inert species, which is then disregarded thereafter. For the same initial density of A and B, $\rho_A(0)=\rho_B(0)$, the mean-field equations predict that ρ_A and ρ_B decay linearly in time as $(kt)^{-1}$. However, it turned out that the random fluctuation of the number of initial particles results in segregation into an A-rich or B-rich area [1,5–7]. The fluctuation and segregation develop in time so that the reactions take place only at the boundaries of two adjacent segregated domains. As a result, in sufficiently low dimensions, the effect of fluctuation leads to the anomalous kinetics. Therefore the evolution of the density of particles strongly depends on fluctuations, and cannot be derived from mean-field rate equations.

The density decay has been known to depend on the motion and the mutual statistics of particles. For isotropic diffusions, the particle density $\rho(t)$ scales in time t as $\rho(t)\sim t^{-d/4}$ in d dimensions ($d\leqslant d_c=4$) [5–10]. Here d_c is the upper critical dimension and thus in $d>d_c\rho(t)$ follows the mean-field result as $\rho(t)\sim t^{-1}$. With the global relative drift of one species, $\rho(t)$ scales as $\rho(t)\sim t^{-(d+1)/4}$ for $d\leqslant 3$ [6]. The hard-core (HC) interaction (constraint) between identical particles is irrelevant to the case of the isotropic diffusion and the relative drift [6]. However, when both species are uniformly driven to the same direction, the HC constraint completely changes the asymptotic scaling as $\rho\sim t^{-(d+1)/6}$ for $d\leqslant 2$, $t^{-d/4}$ for $2<d\leqslant d_c(=4)$ [11,12]. Without the HC constraint, $\rho(t)$ decays as $\rho\sim t^{-d/4}$ as in the isotropic diffusion due to Galilean invariance. Recent studies on the reaction under Lévy mixing [13] and on scale-free networks [14] showed that some mixing mechanism that homogenizes reactants can suppress the role of the fluctuations.

In reality where the recombination of oppositely charged particles into inert particles is the key physical process, the attractive interaction between opposite species should be much more important than the global uniform bias. Real examples for the attractive interactions are electron-hole recombination or photoluminescence in irradiated semiconductor structures [9] and particle-antiparticle reactions in the early universe [1]. Recently there has been enormous progress in manufacturing and understanding the low-dimensional semiconducting systems such as quantum wells, wires, and dots [15,16]. In such quasi-one-dimensional (quasi-two-dimensional) systems or quantum wires (wells) the electron-hole recombination processes have also been extensively studied [15,17]. We investigate the kinetics of $A+B\rightarrow 0$ reaction with the attractive interaction between opposite species, which may be of interest in the recombination processes in various dimensions. In this paper the scaling behavior of the reaction is mainly studied in one spatial dimension. However, we also briefly discuss higher dimensional conjecture for the reaction with a numerical confirmation.

If two opposite species particles surround a particle as AAB in one dimension, the central particle A is ballistically driven to the opposite species B. As a result, the attractive bias depends on the local configurations of adjacent particles, and accelerates the reactions of opposite species at the boundaries of segregated domains. However, inside segregated domains each particle has the same neighboring species and effectively performs isotropic random walks, even for the case with the repulsion between the particles of the same species. Due to the isotropic diffusion inside domains, the HC interaction should be irrelevant in our model. Our model should thus be physically a good model for the electron-hole recombination in a one-dimensional structure (quantum wire), because the long-ranged Coulomb interactions among particles become increasingly more important for a decreased dimensionality of the semiconductor structures [16]. With the attractive bias in one spatial dimension, we analytically and numerically show, regardless of the existence of the hard core (HC) constraint, that the density ρ of particles, the distance ℓ_{AA} between the closest neighbor of the same species, the size ℓ of domains, and the distance ℓ_{AB} between adjacent opposite species scale in time as $\rho\sim t^{-1/3}$, $\ell_{AA}\sim t^{1/3}$, and $\ell\sim\ell_{AB}\sim t^{2/3}$, respectively. These dynamical exponents define completely new critical behavior distinguished from the class of uniformly driven systems of hard-

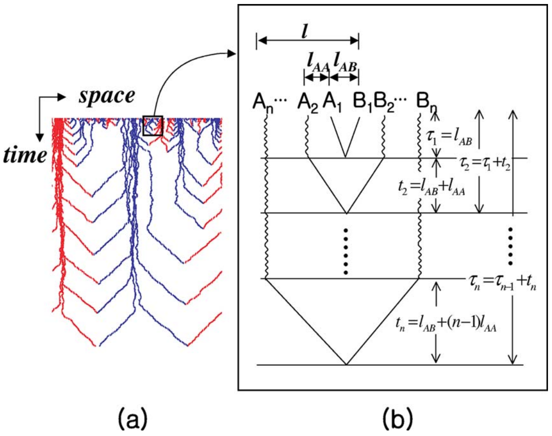

FIG. 1. (Color online) (a) Snapshot of space-time trajectories of $A+B \to 0$ with the attractive bias between opposite species. (b) The magnified schematic trajectories of adjacent opposite species domains. Subscripts {1,2,…,n} indicate the order of the positions of particles from a given domain boundary.

core particles [11,12], where the exponents for ℓ_{AA}, ℓ, and ℓ_{AB} are still controversial. The two features of ballistic and diffusive motions result in pentagonal space-time trajectories of bulk particles [Fig. 1(a)], which allow us to derive the asymptotic scaling analytically.

We consider a configuration in which A and B species are randomly distributed on a one-dimensional lattice with an equal initial density, $\rho_A(0) = \rho_B(0)$. A randomly chosen particle performs either isotropic or biased random walks depending on the configurations of neighbors. When the chosen particle is surrounded by two same species neighbors such as BAB, the chosen particle A performs isotropic random walks. If two opposite species particles surround a particle such as AAB, the chosen particle A is constantly driven to its opposite species B.

In the region of a length ℓ, the number of A species is initially $N_A = \rho_A(0)\ell \pm \sqrt{\rho_A(0)\ell}$ and the same for N_B. After a time $t \sim \ell^z$, particles travel throughout the whole of the region, and annihilate in pairs. The residual particle number is the number fluctuation in the region so we have the relation $N_A \sim \sqrt{\ell}$ or $\rho_A \sim 1/\sqrt{\ell}$ for a given length ℓ [5,6]. As the processes evolve, the system becomes a homogeneous collection of alternating A-rich and B-rich domains. To characterize the structure of segregated domains, we introduce three length scales as in Ref. [9]. The length ℓ of the domain is defined as the distance between the first particles of adjacent opposite species domains [9]. The length ℓ_{AB} is defined as the distance between two adjacent particles of opposite species, while $\ell_{AA}(\ell_{BB})$ is the distance between adjacent $A(B)$ particles in a $A(B)$ domain. These length scales asymptotically increase in time as

$$\ell \sim t^{1/z}, \quad \ell_{AA} \sim t^{1/z_{AA}}, \quad \ell_{AB} \sim t^{1/z_{AB}}. \tag{1}$$

Due to the attractive bias, a bulk particle inside single species domains diffuses isotropically until it becomes a boundary particle, so its space-time trajectory is pentagonal as shown in Fig. 1(a). These pentagonal trajectories should form self-similar (self-affine) fractal structures, because they should have the scaling symmetry due to the power-law behavior of (1). A typical base unit of the self-similar pentagonal trajectories of adjacent opposite domains is schematically depicted in Fig. 1(b). This base unit allows us to calculate a time τ_ℓ needed to remove the unit of size ℓ surrounded by one-scale larger ones. Then the size of the larger unit increases by ℓ during τ_ℓ so we have

$$d\ell/dt \sim \ell/\tau_\ell, \tag{2}$$

which gives the dynamic exponent z.

As only boundary particles of each domain have opposite-species neighbors, the boundary particles ballistically annihilate due to the attractive bias. It takes a time $\tau_1 = \ell_{AB}$ for two boundary particles to annihilate. The second particle from the boundary isotropically diffuses during time τ_1 until the boundary particles annihilate. After the time τ_1, the second particle becomes a boundary particle, and constantly moves to its counterpart during $t_2 = \ell_{AB} + \ell_{AA}$. So it takes time $\tau_2 = \tau_1 + t_2$ in total for the second particle to annihilate. Similarly, the nth particle from the initial boundary will annihilate after $\tau_n = \tau_{n-1} + t_n$, where $t_n = \ell_{AB} + (n-1)\ell_{AA}$ for $n \geqslant 2$. From the recurrence relation of τ_n, we find

$$\tau_n = n\ell_{AB} + n(n-1)\ell_{AA}/2. \tag{3}$$

As the number of particles in a domain of size ℓ is $N_\ell \sim \sqrt{\ell}$, the time τ_ℓ needed to annihilate the domain in the base unit is given by

$$\tau_\ell \sim N_\ell \ell_{AB} + N_\ell^2 \ell_{AA}, \tag{4}$$

for $N_\ell \gg 1$. In the above calculations, we consider the mean positions of bulk particles to effectively be fixed, and neglect the increase of $\ell_{AA}(t)$ by diffusions during the annihilation of the base unit. After a smaller unit is completely annihilated, the remainder of particles redistribute over the larger unit increased by the size of the annihilated unit. Hence we approximate $\ell_{AA}(t) = \cdots = \ell_{AA}(t+\tau_n) = \cdots = \ell_{AA}(t+\tau_\ell)$ during the annihilation of a smaller unit.

The scaling of ℓ_{AA} is simply $\ell_{AA} \sim \sqrt{\ell}$ from the relation $\ell_{AA}(t) \sim 1/\rho(t)$. Hence ℓ_{AA} scales as $\ell_{AA} \sim t^{1/2z}$ with $z_{AA} = 2z$. On the other hand, the change of ℓ_{AB} during τ_ℓ should be in the order of $N_\ell \ell_{AA}$ because of N_ℓ successive annihilations of two opposite particles at boundaries. So we get $d\ell_{AB}/dt \sim \Delta\ell_{AB}/\tau_\ell \sim \ell/t$. With the scaling of $t \sim \ell^z$, we find that ℓ_{AB} follows the same scaling as ℓ, i.e., $\ell_{AB} \sim t^{1/z}$ with $z_{AB} = z$. Finally using the relations $\ell_{AA} \sim \sqrt{\ell}$, $N_\ell \sim \sqrt{\ell}$, and $\ell_{AB} \sim \ell$, we find τ_ℓ from Eq. (4) as $\tau_\ell \sim \ell^{3/2}$. Substituting $\tau_\ell \sim \ell^{3/2}$ into Eq. (2) and integrating the resultant equation, we finally arrive at the following scaling relation:

$$t \sim \ell^{3/2}. \tag{5}$$

As a result, we find $z = z_{AB} = 3/2$ with the attractive bias between opposite species.

From the scaling of ℓ, ℓ_{AB}, and ℓ_{AA}, asymptotic decays of various densities can be extracted. The densities of total particles ($\rho = 2\rho_A$), adjacent pairs of same species ($\rho_{AA} = \rho_{BB}$) and adjacent pairs of opposite species (ρ_{AB}) scale, respectively, as

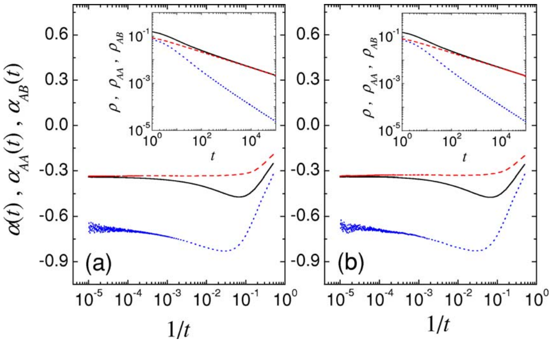

FIG. 2. (Color online) Densities (inset) and their effective exponents of HC particles (a) and bosonic particle (b). From top to bottom, each line corresponds to α_{AA}, α, and α_{AB}. In the inset, the order is ρ, ρ_{AA}, and ρ_{AB}, respectively.

$$\rho \sim t^{-\alpha}, \quad \rho_{AA} \sim t^{-\alpha_{AA}}, \quad \rho_{AB} \sim t^{-\alpha_{AB}}. \tag{6}$$

As ρ is in the order of $\rho \sim 1/\sqrt{\ell}$, we have $\rho \sim t^{-1/2z}$ with $\alpha = 1/2z = 1/3$. ρ_{AA} follows the same scaling of ρ due to $\rho_{AA} \sim 1/\ell_{AA} \sim 1/\sqrt{\ell}$ so $\rho_{AA} \sim t^{-\alpha}$ with $\alpha_{AA} = \alpha = 1/3$. Finally ρ_{AB} is $\rho_{AB} \sim 1/\ell$, which leads to $\rho_{AB} \sim t^{-1/z}$ with $\alpha_{AB} = 1/z = 2/3$. Using self-similar structures of space-time trajectories and scaling arguments for fluctuations of [5,6,9], we find the following exponents for the reactions $A+B\to 0$ with the attractive bias between opposite species:

$$\alpha = \alpha_{AA} = 1/3, \quad \alpha_{AB} = 2/3,$$

$$z = z_{AB} = 3/2, \quad z_{AA} = 3. \tag{7}$$

Intriguingly and incidentally $\rho(t)$ decays with the same exponent $1/3$ as that of the uniformly driven hard-core particles [11,12], in which the driven motion of a single species domain was argued to be described by the noisy Burgers equation [11]. However, for the present model the $1/3$ decay of $\rho(t)$ comes from the interplay of isotropic diffusions inside domains and ballistic annihilations at boundaries. For scalings of interdomain distances and others, our results (7) are completely different from those of [12], where $\ell \sim t^{7/12}$, $\ell_{AB} \sim t^{3/8}$, and $\ell_{AA} \sim t^{1/3}$. Hence we conclude that the attractive bias between opposite species leads to new critical behavior of irreversible $A+B\to 0$ reactions.

To confirm our analytic predictions, we now discuss the simulation results for the model with the attractive bias. With $\rho_A(0) = \rho_B(0)$, A and B particles distribute randomly on a lattice. In the simulations we consider both HC particles and the particles without the HC constraints, which we call the bosonic particles. In the model with HC particles there can be at most one particle of a given species on a site. In the bosonic model there can be many particles of the same species on a site. As we shall see, the simulation results are independent of the HC constraints.

All the simulations are done on one-dimensional chains with the size up to $L = 3 \times 10^6$ and the initial density always set as $\rho_A(0) = \rho_B(0) = 0.1$. We average $\rho(t)$, $\rho_{AA}(t)$, and $\rho_{AB}(t)$ up to 10^5 time steps over 7 200 independent runs. In Fig. 2, we plot the densities and their effective exponents defined as $\alpha(t) = -\ln[\rho(t)/\rho(t/2)]/\ln 2$ and similarly for others. As shown in Fig. 2, the data for HC particles [Fig. 2(a)] are almost identical to those for bosonic particles [Fig. 2(b)]. While α_{AB} still shows larger fluctuations for both HC and bosonic particles, α and α_{AA} nicely converge to the same value. We estimate $\alpha = 0.33(1)$, $\alpha_{AA} = 0.33(1)$, and $\alpha_{AB} = 0.68(2)$ for both HC and bosonic models, which agrees well with the prediction (7).

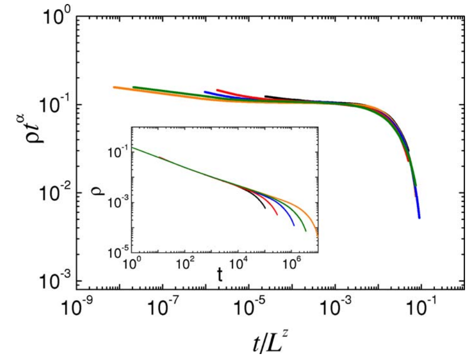

FIG. 3. (Color online) Scaling collapse for $\rho(t)$ of hard-core particles with $\alpha = 1/3$ and $z = 3/2$. The inset shows the raw data for $\rho(t)$. Used particles are hard-core particles. The plot for bosonic particles is not shown because it is almost identical to the main plot.

To estimate the dynamic exponent z, we measure $\rho(t,L)$ for various L from 2^{14} up to 2^{18}. With the scaling assumption $\rho(t,L) \sim t^{-\alpha} f(t/L^z)$, and $\alpha = 1/3$, we observe the best data collapse at $z = 1.50(2)$ which also agree well with the prediction (7) as shown in Fig. 3. For the time dependence of average distances defined in Eq. (1), we measure ℓ, ℓ_{AA}, and ℓ_{AB} under the same measurement conditions as those of densities. The effective exponents of the distances are defined similarly as for $\alpha(t)$ except the minus sign. The results for the average distances are shown in Fig. 4. Here we can also see that the data for HC particles [Fig. 4(a)] are almost identical to those for bosonic particles [Fig. 4(b)]. From Fig. 4, we estimate $1/z = 0.683(3)$, $1/z_{AA} = 0.338(3)$, and $1/z_{AB} = 0.66(1)$ for both HC and bosonic particles.

All the simulation results in Figs. 2–4 numerically confirm the prediction (7). They also confirm the irrelevance of the HC constraints in our model unlike in the uniformly driven systems [6,11]. The isotropic diffusion inside segre-

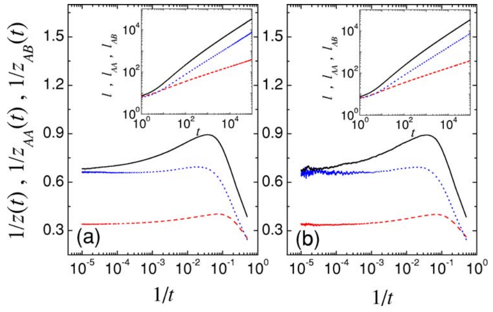

FIG. 4. (Color online) Average distances (inset) and their effective exponents in the model with HC particles (a) and with bosonic particles (b). From top to bottom, each line corresponds to $1/z$, $1/z_{AB}$, and $1/z_{AA}$, respectively. In the inset the order of lines is the same as in the main plot.

gated domains leads to Galilean invariance of domains so the scaling behavior is not affected by HC constraint.

In higher dimensions, the length ℓ of a typical domain at time t is still expected to satisfy $\ell \sim t^{2/3}$ due to the diffusive and ballistic motions. We thus predict $\rho \sim 1/\sqrt{V} = 1/\sqrt{\ell^d} \sim t^{-d/3}$ with the upper critical dimension $d_c = 3$. We also numerically confirm the scaling $\rho \sim t^{-d/3}$ in two dimensions [18]. Furthermore we confirm continuously varying α's and z's depending on δ when the attractive interaction between a pair of opposite species varies as $r^{-\delta}$ where r is the distance between the pair [18].

To summarize, we investigate the kinetics of $A+B \rightarrow 0$ reactions with the attractive bias between opposite species. As reactions proceed, A-rich and B-rich domains appear alternatively, and annihilations of opposite species take place only at the boundaries of closest neighboring domains [1,5,6]. However the reactions at domain boundaries are accelerated by the attractive bias, while particles inside domains perform isotropic random walks. The interplay of isotropic diffusions and ballistic annihilations of boundary particles leads to pentagonal self-similar trajectories, which allow us to derive the analytic prediction (7). The anomalous density decay $t^{-1/3}$ appears to be the same scaling behavior as uniformly driven systems of HC particles, which was argued to be described by noisy Burgers equations [11]. However, our system does not show the same scaling behavior as the uniformly driven system because scaling behaviors of basic distances are different [12]. The difference can be inferred from the underlying mechanisms. In our model, isotropic diffusions inside domains lead to the Galilean invariance, so HC constraint has no effects on the kinetics. Furthermore, there is no global bias to one direction, which changes the kinetics of HC particles. Only boundary particles feel bias to opposite species, which is the essential physical factor to distinguish our model from the models in Refs. [1,5,6]. We conclude that the attractive bias between opposite species is the key feature of the new critical behavior characterized by Eq. (7) and another path to the anomalous density decay of $t^{-1/3}$ in one dimension.

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