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Scaling of reaction zones in the $A+B\rightarrow 0$ diffusion-limited reaction

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We study reaction zones in three different versions of the $A+B\rightarrow 0$ system. For a steady state formed by opposing currents of A and B particles we derive scaling behavior via renormalization group analysis. By use of a previously developed analogy, these results are extended to the time-dependent case of an initially segregated system. We also consider an initially mixed system, which forms reaction zones for dimension $d<4$. In this case an extension of the steady-state analogy gives scaling results characterized by new exponents.

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The reaction-diffusion process $A+B\rightarrow 0$ has received much attention since the work of Toussaint and Wilczek [1], in which it was demonstrated that density fluctuations are important when the dimensionality $d<4$. Attempts to incorporate the fluctuation effects, including the application of renormalization group (RG) methods, have proven successful for one-species reactions [2–4]. However, the physics of the two-species reaction is somewhat different, as the number difference of A and B particles is locally conserved. Nonetheless, the main results of the RG analysis may be extended to this case [5,6].

In the present work we are concerned with situations in which the A and B particles are well segregated, so that reactions are then confined to reaction zones, on the boundaries between the A- and B-dominated regions. The segregation may be a consequence of the initial conditions, or arise asymptotically from a mixed initial state when $d<4$. Both of these cases may be compared to that of a steady state formed by directing steady, uniform currents of A and B particles towards each other. In all three cases the structures of the reaction zones are very similar.

Reaction zones were studied by Gálfi and Rácz [7] in the context of a system with segregated initial conditions. Their analysis is essentially a mean-field result, as we will show, which holds asymptotically for $d>2$. More recently, Ben-Naim and Redner have derived the mean-field behavior for the steady-state problem [8], and Cornell and Droz extended this to $d<2$ via RG motivated scaling arguments [9]. Reaction zones in initially homogeneous systems have been studied by Leyvraz and Redner [10,11]. In this paper we use exact RG methods both to confirm previous results and to derive new exponents which characterize the scaling behavior of the reaction zone.

The model we shall use for the $A+B\rightarrow 0$ reaction is one of particles undergoing continuous-time random walks on a hypercubic lattice. We consider general, nonzero values of the diffusion constants D_A and D_B. If an A and a B particle are together on a lattice site, then they annihilate with some characteristic reaction rate λ. In the field-theoretic approach it is convenient to allow multiple occupancy of lattice sites, regardless of particle type, but this is not expected to alter the universal properties.

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TABLE I. The exponents, as defined in the text, for the scaling behavior in the cases of segregated and homogeneous initial conditions.

		α	β	γ
Segregated	$d\leq 2$	$\frac{1}{2(d+1)}$	$\frac{d+2}{2(d+1)}$	$\frac{1}{2(d+1)}$
	$d>2$	$\frac{1}{6}$	$\frac{2}{3}$	$\frac{1}{3d}$
Homogeneous	$d\leq 2$	$\frac{d+2}{4(d+1)}$	$\frac{(d+2)^2}{4(d+1)}$	$\frac{d+2}{4(d+1)}$
	$2<d<4$	$\frac{d+2}{12}$	$\frac{d+2}{3}$	$\frac{d+2}{6d}$

First we consider the steady state reached when equal currents J of A and B particles are directed towards each other. The average densities then vary only in the direction of the currents. In this case the width of the reaction zone goes as a power of J, as $J\to 0$,

$$w\sim\begin{cases} J^{-1/(d+1)}, & d\leq 2 \\ J^{-1/3}, & d>2. \end{cases} \tag{1}$$

The typical nearest-neighbor distance in the reaction zone ℓ_{rz} scales as

$$\ell_{rz}\sim\begin{cases} J^{-1/(d+1)}, & d\leq 2 \\ J^{-2/3d}, & d>2. \end{cases} \tag{2}$$

The second example we consider is that of initially segregated particles. Consider a system prepared with only A particles in the region $x<0$ and only B particles for $x>0$. The behavior in the reaction zone again scales, now with respect to time, with $w\sim t^{\alpha}$ and $\ell_{rz}\sim t^{\gamma}$. The densities in the reaction zone, where $|x-x_c|\lesssim w$, have the scaling form

$$\langle a\rangle,\langle b\rangle\sim t^{-\gamma d}F_{a,b}\left(\frac{x-x_c}{t^{\alpha}}\right), \tag{3}$$

where $x_c\propto t^{1/2}$ is the reaction zone center. The angular brackets will be defined precisely below. The reaction rate R also has the scaling form

$$R(x,t)\sim t^{-\beta}G\left(\frac{x-x_c}{t^{\alpha}}\right). \tag{4}$$

The values of the exponents are given in Table I.

The final example is that of random, homogeneous initial conditions, with equal densities n_0 of A and B particles. It has been shown in the mathematical treatment of Bramson and Lebowitz [12] that this system exhibits asymptotic segregation for $d<4$. We present a simple derivation of this result from an effective field theory, which is valid for $2<d<4$. We argue that reaction zones which result from the segregation can be studied in the same fashion as in the previous problems. As a result, we predict the same scaling forms as given by Eqs. (3) and (4), with new values for the exponents, which are listed in rows 3 and 4 of Table I.

The characteristic nearest-neighbor distance in the reaction zone ℓ_{rz} is found from the scaling form of the densities in the reaction zone (3). Consider the total number of particles N in the volume of the reaction zone for a given interfacial surface area S_{d-1},

$$N\sim S_{d-1}\int_{x_c-w}^{x_c+w}dx\langle a(x)+b(x)\rangle\sim S_{d-1}t^{\alpha-\gamma d}. \tag{5}$$

Since the volume of this region is $S_{d-1}t^{\alpha}$, then the average volume per particle goes as $t^{\gamma d}$. If one assumes that there is just one length scale describing the typical nearest-neighbor distance, both in the direction of the interface and perpendicular to it, and also for both like and unlike particles, then this length is given by $\ell_{rz}\sim t^{\gamma}$. A convenient definition of ℓ_{rz}, which follows from the derivation above, is $\ell_{rz}\equiv\langle a(x_c)\rangle^{-1/d}$.

The model for $A+B\to 0$ can be mapped to a field theory, which is useful for the application of RG methods [5,13]. We summarize here the results of a study of this field theory which pertain to the problem at hand. A more detailed account will appear in [6]. One feature, which is general to all irreversible reaction-diffusion systems, is that there are no diagrams which dress the propagator. This means that there is no wave function renormalization, and consequently no anomalous dimension for the fields. The significance of this will be noted below.

There is only one coupling constant λ_0 in the field theory (in the notation of [5]), which is given in terms of the original microscopic parameters as $\lambda_0=\lambda h^d/\bar{D}$, where $\bar{D}=(D_A+D_B)/2$ and h is the lattice size. The coupling λ_0 is found to be irrelevant, in the RG sense, for $d>2$, marginal for $d=2$, and relevant for $d<2$.

In general, one can derive from the field theory the equations of motion

$$\partial_t\langle a\rangle=D_A\nabla^2\langle a\rangle-R,\quad \partial_t\langle b\rangle=D_B\nabla^2\langle b\rangle-R, \tag{6}$$

where $R=\lambda_0\bar{D}\langle ab\rangle$ is the reaction rate. The angular brackets refer to averages over the stochastic processes of diffusion and reaction, but not over the initial conditions. Averages over the initial conditions will be denoted with a bar: $\overline{\langle a\rangle}$. Note that the quantity $\langle a-b\rangle$ obeys a simple diffusion equation when $D_A=D_B$.

When $d>2$ the irrelevance of the coupling leads to the asymptotic result that $R\sim\Gamma\langle a\rangle\langle b\rangle$. Then the equations of motion (6) are simply differential equations with rate constant Γ. This is the starting point in the analysis of Refs. [7,8], consequently their results are applicable for $d>2$. When $d\leq 2$ one finds that the coupling λ_0 can be renormalized exactly, allowing us to derive the scaling behavior of the reaction zones.

Consider the steady state reached by imposing currents $\mathbf{J}_a=J\hat{\mathbf{x}}$ at $x=-L$ and $\mathbf{J}_b=-J\hat{\mathbf{x}}$ at $x=L$, where $L\gg w$. From the analysis in [7,8] one finds for $d>2$ that

$$R\sim\langle a\rangle\langle b\rangle\sim J^{4/3}f(xJ^{1/3}), \tag{7}$$

from which the exponents for $d>2$ given in Eqs. (1) and (2) follow directly.

For $d\leq 2$ one must consider the effects of renormalization. We work in units of time where the average dif-

fusion constant $\tilde{D}=1$, and define the parameter $\delta=(D_A-D_B)/(D_A+D_B)$. One can show that δ is not renormalized, for the same reason that there is no wave-function renormalization. In these units the dimension of the current is $[J]=(\text{length})^{-1-d}$. The width w can depend in the steady state on λ_0, J, and δ only.

In the field-theoretic RG, the λ_0 dependence is traded for a dependence on a renormalized coupling λ_R, defined at an arbitrary length scale $J_0^{-1/(d+1)}$. We then consider w as depending on J, δ, J_0, and the the dimensionless renormalized coupling $g_R\equiv\lambda_R J_0^{(d-2)/(d+1)}$. This then satisfies an RG equation whose solution is

$$w(J,g_r,J_0,\delta)=\left(\frac{J}{J_0}\right)^{-1/(d+1)} w(J_0,\tilde{g}(J/J_0,g_R),\delta)\ , \quad (8)$$

where, as $J\rightarrow 0$, the running coupling $\tilde{g}\rightarrow g^*=O(2-d)$, *independently* of the initial value of λ_0. When $d=2$ the running coupling goes as $\tilde{g}\sim B/\ln|J|$ for small J. The simple form of the overall scale factor in (8) is a result of the absence of wave-function renormalization.

The significance of Eq. (8) is that, in the limit of small J, the only J dependence on the right-hand side comes from the overall scale factor. Therefore any length will scale asymptotically as $w\sim\ell_{rz}\sim J^{-1/(d+1)}$, giving the results stated in Eqs. (1) and (2) for $d\leq 2$. When $d=2$ the running coupling $\tilde{g}$ is J dependent. However, it can be shown that the right-hand side is finite in the limit $\tilde{g}\rightarrow 0$ [6]. Therefore the leading term for small J is unchanged, and there are no logarithmic corrections. It should be noted that while this result holds for general $|\delta|<1$, the limit of $|\delta|\rightarrow 1$ does not necessarily commute with the asymptotic limit in our analysis, so our results are only for the case when both species are mobile.

The asymptotic limit of any dimensionful quantity can be found by the same technique, using only dimensional analysis. Therefore the densities in the reaction zone have the scaling form

$$\langle a(x)\rangle,\langle b(x)\rangle\sim J^{d/(d+1)}F_{a,b}(xJ^{1/(d+1)}) \quad (9)$$

and the reaction rate

$$R(x)\sim J^{(d+2)/(d+1)}G(xJ^{1/(d+1)}). \quad (10)$$

The scaling functions $F_{a,b}$ and G are expected to be universal, up to metric factors, and should be calculable within a $(2-d)$ expansion. Note that $R\propto\langle ab\rangle\sim J^{(2-d)/(d+1)}\langle a\rangle\langle b\rangle$ for small J. This result can be demonstrated explicitly within the original field theory [6].

An equivalent and less formal way to state these results is that there are only two input parameters with dimension: J, and λ_0. However, for $d\leq 2$, in the asymptotic limit the theory is independent of the initial value of λ_0, so the remaining J dependence can be determined on dimensional grounds.

These results were found by Cornell and Droz [9], by use of an RG-motivated scaling analysis. Our work provides a quantitative confirmation of the lack of anomalous dimension and asymptotic lack of λ_0 dependence. They performed simulations for $d=1,2,3$ which are in good agreement with the predictions.

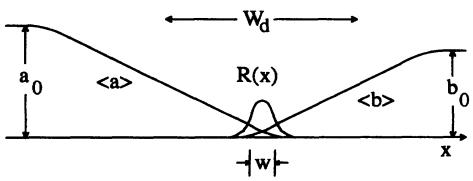

FIG. 1. The profile of a reaction zone.

We now turn to the problem of segregated initial conditions. At $t=0$ the boundary is at $x=0$, and the A and B particles are randomly distributed within their regions, with initial densities a_0 and b_0, respectively. A profile of the densities at a later time t is sketched in Fig. 1. The densities are depleted out to a range $W_d\sim t^{1/2}$, which is the length over which particles will have had a chance to diffuse into reaction range. Provided that $\alpha<1/2$, which will be verified self-consistently, then asymptotically $w\ll W_d$. Consequently one finds in the depletion regions $w\ll|x-x_c|\ll W_d$ that $R\approx 0$ and Eq. (6) reduces to the diffusion equation. As a result, it follows that the density profiles in this depletion region are linear in $x-x_c$. The slope can be determined by observing that the A particle density goes from a_0 to zero in a range W_d, so the slope $-a_0/W_d\sim -t^{-1/2}$.

In Ref. [8] Ben-Naim and Redner show that this linear depletion regime provides the same boundary conditions for the reaction zone as was found in the steady-state system. In that case the slope in the linear region $(w\ll|x|<L)$ is given by $\partial_x\langle a\rangle=-J$. Therefore we can apply the results obtained from the steady state to the time-dependent reaction zone by making the scaling substitution $J\sim t^{-1/2}$. The result is the scaling forms given by Eqs. (3) and (4) with the exponents shown in Table I. The exponent γ has not been calculated previously.

The exponents α and β, which describe w and R, were derived in the analysis of Cornell and Droz [9]. They find reasonable agreement with these predictions in $d=1$ simulations, although the asymptotic region is difficult to obtain. Other simulations in $d=1$ find evidence for multiscaling [14]. Since the parameters in these simulations are at the opposite extreme from the weak coupling expansion implicit in the field-theoretic approach, it is conceivable that they might fall into a separate universality class. However, more extensive simulations seem to indicate that ordinary scaling is recovered asymptotically [15].

The final case we consider is the system with homogeneous initial conditions. The A and B particles are randomly distributed with equal initial densities n_0. Starting from Eq. (6) one can show that the average over initial conditions yields a density $\overline{\langle a\rangle}\sim t^{-d/4}$ for $d<4$ [6]. As argued by Bramson and Lebowitz, the A and B particles segregate asymptotically, which may be shown directly from Eq. (6) for $2<d<4$ as follows. We use the notation $\langle a\rangle\rightarrow a$, since $R=\Gamma ab$ for $d>2$.

Given that $\bar{f}^2\leq\overline{f^2}$, where f is a real quantity, then

$$\overline{\min(a,b)}^2\leq\overline{\frac{1}{2}(a^2+b^2-(a+b)|a-b|)}. \quad (11)$$

Since a and b correspond to the physical density for a par-

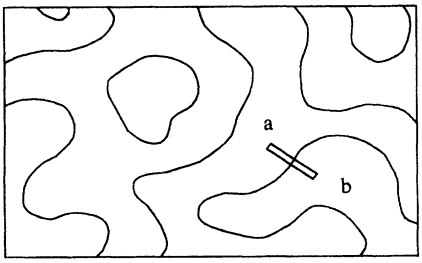

FIG. 2. A sketch of the percolated domain configuration in $d=2$. The domains have a characteristic length scale which grows in time as $t^{1/2}$.

ticular initial condition, they are everywhere non-negative. Therefore $a+b \geq |a-b|$ at every point $(\mathbf{x},t)$, which implies that

$$\overline{\min(a,b)}^2 \leq \frac{1}{2}\overline{(a^2+b^2-(a-b)^2)} = \overline{ab}. \tag{12}$$

From Eq. (6) it follows that

$$\overline{ab} = \frac{1}{\Gamma}\,\partial_t \bar{a} = O(t^{-1-d/4}), \tag{13}$$

so one has the result

$$\overline{\min(a,b)} \leq O(t^{-1/2-d/8}) \ll t^{-d/4} \tag{14}$$

when $d<4$. Therefore the local minority density goes to zero relative to that of the majority species. A sketch of the segregated domains in $d=2$ is shown in Fig. 2.

In this case we may make a similar analogy to the steady-state case, except now the height at the edge of the depletion region is time dependent. The rectangle in Fig. 2 gives the cross section of a reaction zone, which will have the same form as Fig. 1. The time dependence of the height is in general complicated, but we argue that in most regions it will scale with the bulk density. The picture one has is that the densities in the bulk will be fairly uniform since there the particles diffuse without reacting. This uniform density must scale as $t^{-d/4}$, so that averages over the reaction zones of the whole system will yield exponents which can be derived simply by taking $a_0, b_0 \to t^{-d/4}$. The resulting steady-state analog is given by $J \sim t^{-(d+2)/4}$. This gives the scaling results in Eqs. (3) and (4), with the exponents listed in Table I for the homogeneous case.

The values for the exponents α and β have not been derived previously. We note that $\alpha \to 1/2$ from below as $d \to 4$. This is consistent with the view that segregation is breaking down, since the reaction zones are then on the same scale as the domains. The exponent γ was derived by Leyvraz and Redner [10,11] by a different method. They were interested in the quantity ℓ_{AB}, which is the characteristic nearest-neighbor distance when the nearest neighbor of a particle is of the opposite species. This quantity can be measured directly in simulations. Since the particles are segregated, the contributing particles to the distribution for ℓ_{AB} must be within the reaction zone, and therefore $\ell_{AB} \sim \ell_{rz} \sim t^{\gamma}$. Leyvraz and Redner found from both scaling arguments and simulations values of γ which agree with our predictions for $d=1,2$. However, they argue for $d>2$ that $\gamma = 1/4$, and show the results of $d=3$ simulations. Our prediction for $d=3$ is $\gamma = 5/18$, which is very close to $t^{1/4}$. It appears from the data of [11] that either value of γ is an equally good fit. A numerical value for a fit to the data is not provided. The significance of this discrepancy is that the nearest-neighbor distance in the bulk $\overline{\langle a \rangle}^{-1/d} \sim t^{1/4}$ for $d<4$. We claim that ℓ_{rz} scales with a different exponent than the bulk nearest-neighbor distance for all $d<4$.

A check on the above results is to calculate the characteristic domain size. Integrating the equations of motion (6) over the entire system of size V gives

$$\int d^d x\, \partial_t \langle a \rangle \sim -\int d^d x \langle ab \rangle \tag{15}$$

or equivalently

$$V t^{-1-d/4} \sim A \int dx_\perp R(x_\perp) \sim A t^{-\beta+\alpha}, \tag{16}$$

where A is the interfacial area of the domain boundaries. This first result follows from the fact that the only significant contribution to the reaction rate comes from the reaction zones, the second from Eq. (4). This leads to the characteristic domain size

$$\ell = V/A \sim t^{-\beta+\alpha+1+d/4} \sim t^{1/2} \tag{17}$$

for all $d<4$. This result, while often assumed, follows for $d<2$ only because $R \neq \langle a \rangle \langle b \rangle$.

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