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Spatial Structure in Diffusion-Limited Two-Particle Reactions

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We analyze the limiting behavior of the densities $\rho_A(t)$ and $\rho_B(t)$, and the random spatial structure $\xi(t) = (\xi_A(t), \xi_B(t))$, for the diffusion-controlled chemical reaction $A + B \to$ inert. For equal initial densities $\rho_A(0) = \rho_B(0)$ there is a change in behavior from $d \leqslant 4$, where $\rho_A(t) = \rho_B(t) \approx C/t^{d/4}$, to $d \geqslant 4$, where $\rho_A(t) = \rho_B(t) \approx C/t$ as $t \to \infty$; the term C depends on the initial densities and changes with d. There is a corresponding change in the spatial structure. In $d < 4$, the particle types separate with only one type present locally, and ξ, after suitable rescaling, tends to a random Gaussian process. In $d > 4$, both particle types are, after large times, present locally in concentrations not depending on type or location. In $d = 4$, both particle types are present locally, but with random concentrations, and the process tends to a limit.

KEY WORDS: Diffusion-limited reaction; annihilating random walks; asymptotic densities; spatial structure; exact results.

1. INTRODUCTION

Consider a system of particles of two types on $\mathbb{Z}^d$, A and B, which execute simple random walks in continuous time. That is, the motion of different particles is independent and a particle at site x will jump to a given one of its $2d$ nearest neighbors at rate $1/2d$. Particles are assumed not to interact with their own type—multiple A particles or multiple B particles can occupy a given site. However, when a particle meets a particle of the opposite type, both disappear. (When a particle simultaneously meets more than one particle of the opposite type, it will cause only one of these particles to disappear.)

To study the time evolution of this system, one needs to specify an

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initial measure for the process. We consider here the case in which one independently throws down A and B particles according to the homogeneous Poisson random measures with densities r_A and r_B; if there are initially both A and B particles at x, they immediately cancel each other out as much as possible. We denote by $\xi(t) = (\xi_A(t), \xi_B(t))$ the random state of the system at time t; $\xi(G; t)$ denotes the number of particles of each type in the set $G \subset \mathbb{Z}^d$.

This process can serve as a model for the irreversible chemical reaction $A + B \rightarrow$ inert, where both particle types A and B are mobile. A and B can also represent matter and antimatter. There has been much interest in this model over the last several years following papers by Ovchinikov and Zeldovich[1] and Toussaint and Wilczek[2]; see Bramson and Lebowitz[3,4] and refs. 5–8 for a more complete set of references. The main concern has been with the behavior of the densities in spatially homogeneous systems, i.e., with the expected number of A and B particles per site, $\rho_A(t)$ and $\rho_B(t)$, as $t \rightarrow \infty$. (The density of course does not depend on the site x.) The two basic cases are when (a) $0 < \rho_A(0) = \rho_B(0)$ (equal densities) and (b) $0 < \rho_A(0) < \rho_B(0)$ (unequal densities). Note that (a) corresponds to $0 < r_A = r_B$ and (b) to $0 < r_A < r_B$. Since $\rho_B(t) - \rho_A(t)$ must clearly remain constant for all t, one has $\rho_A(t) = \rho_B(t)$ in (a), and $\lim_{t \rightarrow \infty} \rho_B(t) = \rho_B(0) - \rho_A(0) > 0$ in (b). The rate at which $\rho_A(t) \rightarrow 0$ is analyzed in ref. 3. We summarize the results for both equal and unequal densities here, but will be primarily interested in the former. In this case, the asymptotic density of $\rho_A(t)$ changes from $t^{-d/4}$ in $d \leqslant 4$ to t^{-1} in $d \geqslant 4$.

In addition to the long-term behavior of the density, there has also been interest in the limiting spatial structure of the model (see, e.g., ref. 2). Simulations indicate that in $d \leqslant 3$, the particle types separate as time increases, so that most regions are dominated by one type. This behavior is in accordance with the above asymptotics for $\rho_A(t)$. One does not expect this separation in $d \geqslant 4$, again by reference to $\rho_A(t)$. We rigorously analyze this asymptotic behavior. We show that after suitable rescaling, $\xi(t)$ converges to a limiting process. Its local densities are given by a parabolic equation with random initial data, and the process itself is locally a Poisson random field. The results are qualitatively different in $d < 4$ and $d > 4$, in conformity with expectations. The behavior in $d = 4$ is a hybrid of the two other cases. We present a summary here, with a detailed treatment being given in ref. 4.

2. ASYMPTOTIC DECAY OF DENSITIES

For $\rho_A(0) = \rho_B(0)$, one can reason that $\rho_A(t)$ should decrease like $1/t^{d/4}$ for $d \leqslant 4$ and like $1/t$ for $d \geqslant 4$. The standard logic is that if one "neglects" the random fluctuations in the number of the two types of particles present in a local region, as can be achieved physically by vigorous stirring, one can treat the positions of particles for the two types as being independent. The rate at which A particles meet B particles is then proportional to the density of each type present. This gives the "law of mass action" or mean-field behavior

$$\frac{d\rho_A(t)}{dt} = -k\rho_A(t)\,\rho_B(t) \tag{1}$$

for appropriate $k > 0$. Since $\rho_A(t) = \rho_B(t)$, we have for the solution of (1)

$$\rho_A(t) \approx 1/kt \qquad \text{for large } t \tag{2}$$

Here, by $a(t) \approx b(t)$ we mean that $a(t)/b(t) \to 1$ as $t \to \infty$.

On the other hand, if one examines the effect of local fluctuations of the initial state, one notes that the difference of the number of A particles and B particles in a cube of length M has variance of order M^d in d dimensions. The difference in the number of A and B particles is hence typically of order $M^{d/2}$. This gives a particle density of at least $M^{-d/2}$. Since it takes particles not initially close to the boundary of the order of time M^2 to enter or leave the cube, one might expect this difference to continue up through times of this order. Plugging in $t = M^2$, one obtains a lower bound of $t^{-d/4}$ for $\rho_A(t)$ and $\rho_B(t)$.

One needs to reconcile these bounds with (2). The standard heuristics are that the effect of local fluctuations dominates in $d < 4$, whereas the mean-field limit in (2) is accurate for $d \geqslant 4$. The densities $\rho_A(t)$ and $\rho_B(t)$ should therefore decay asymptotically like $t^{-d/4}$ for $d \leqslant 4$ and t^{-1} for $d \geqslant 4$. The following result, an improvement of Theorem 1 in ref. 3, verifies this behavior.

Theorem 1. Assume that the initial measure is Poisson with $r_A = r_B > 0$. There exist positive constants $C_d > 0$ such that

$$\begin{aligned} \rho_A(t) = \rho_B(t) &\approx C_d \sqrt{r_A}/t^{d/4}, && d < 4 \\ &\approx C_4(\sqrt{r_A} \vee 1)/t, && d = 4 \\ &\approx C_d/t, && d > 4 \end{aligned} \tag{3}$$

[$\alpha \vee \beta$ (resp. $\alpha \wedge \beta$) denotes the larger (resp. smaller) of α and β.]

For $\rho_A(0) < \rho_B(0)$, the asymptotic behavior of $\rho_A(t)$ is quite different. Since $\lim_{t \to \infty} \rho_B(t) = \rho_B(0) - \rho_A(0) = r_B - r_A \equiv b > 0$, there is always at least density b of type B particles in the population. The density $\rho_A(t)$ must therefore decrease much more rapidly than if $\rho_A(0) = \rho_B(0)$. From (1), one obtains

$$\frac{d\rho_A(t)}{dt} = -k[b + o(1)]\,\rho_A(t) \tag{4}$$

Consequently, one might expect that

$$\rho_A(t) = \rho_A(0)\,e^{-k[b+o(1)]t} \tag{5}$$

On the other hand, as in the case $\rho_A(0) = \rho_B(0)$, local fluctuations might alter the relative proportions of type A and type B particles locally, and cause a different rate of decay. Presumably, as before, this change would be associated with lower dimensions. The following result was proved in ref. 3.

Theorem 2. Assume that the initial measure is Poisson with $0 < r_A < r_B$. There exist positive constants Λ_d and λ_d so that

$$\exp[-\Lambda_d \phi_d g_d(t)] \leqslant \rho_A(t) \leqslant \exp[-\lambda_d \phi_d g_d(t)] \tag{6}$$

for large enough t, where

$$g_d(t) = \begin{cases} \sqrt{t}, & d = 1 \\ t/\log t, & d = 2 \\ t, & d \geqslant 3 \end{cases} \tag{7}$$

and

$$\phi_d(t) = \begin{cases} (r_B - r_A)^2/r_B, & d = 1 \\ r_B - r_A, & d \geqslant 2 \end{cases} \tag{8}$$

The mean-field limit is thus valid in $d \geqslant 3$, but not in $d = 1, 2$. The dependence on initial densities is different in $d = 1$ than that in $d > 1$, which corresponds to (5). (The reason is the presence of greater fluctuations in $d = 1$.) Theorem 2 will also provide some insight in understanding the local spatial structure for $r_A = r_B$ in $d < 4$.

3. SPATIAL STRUCTURE—MACROSCOPIC BEHAVIOR

We wish to analyze the asymptotic behavior of the process $\xi(t) = (\xi_A(t), \xi_B(t))$ as $t \to \infty$. As would be expected, we rescale both space and time to pass to a limiting process. Individual particles execute random walks (until annihilation). Since rescaling time by t and space by $\sqrt{t}$

produces Brownian motion in the limit for such particles, it makes sense to undertake the same rescaling here. We therefore set

$$\xi^t(G; s) = \xi(\sqrt{t}\, G; ts) \tag{9}$$

where $G \subset \mathbb{R}^d$, $\sqrt{t}\, G$ is the set G multiplied by $\sqrt{t}$ in each direction, and $s, t \geqslant 0$. After rescaling by $\sqrt{t}$, we need to compensate for the increase in particle density in this new scale. Applying Theorem 1, we see that on this scale one has density of magnitude $t^{-d/4} \cdot t^{d/2} = t^{d/4}$ for $d \leqslant 4$ and $t^{-1} \cdot t^{d/2} = t^{d/2-1}$ for $d \geqslant 4$ as $t \to \infty$. In either case, there are hopefully enough particles present so that the local behavior is determined by some law of large numbers, and the evolution of the process is, on this macroscopic, or hydrodynamic, scale, asymptotically deterministic. (See Lebowitz *et al.*[9] for a general reference on hydrodynamic scaling.)

A limiting process η attained in such a manner will depend on its initial measure and a rule prescribing its evolution. Since $\xi(0)$ is defined by Poisson random measures which are independent at different sites, rescaling $\xi(0)$ gives white noise, which we denote by Φ. [That is, Φ is the generalized Gaussian random field with covariance structure

$$E[\Phi(\varphi)\, \Phi(\psi)] = \int_{\mathbb{R}^d} \varphi(x)\, \psi(x)\, dx \tag{10}$$

where φ, ψ are test functions. Alternatively, one can substitute indicator functions for φ and ψ, or write $E[\Phi(x)\, \Phi(y)] = \delta(x-y)$. In $d=1$, Φ is just the "derivative" of Brownian motion.] One might hope that the evolution of η is connected with normal distributions. Here N_s denotes a normal distribution in d dimensions with mean 0 and variance s.

We now state Theorem 3, which gives the behavior of η in $d<4$. Here the asterisk denotes convolution and $[x]^- = -(x \wedge 0)$, $[x]^+ = x \vee 0$.

Theorem 3. For $d<4$,

$$t^{-d/4}\xi^t \xrightarrow{\text{w}} \eta \qquad \text{as} \quad t \to \infty \tag{11}$$

where $\eta = (\eta_{\text{A}}, \eta_{\text{B}})$ and

$$\begin{aligned} \eta_{\text{A}}(G; s) &= [(N_s * \Phi)(G)]^- \\ \eta_{\text{B}}(G; s) &= [(N_s * \Phi)(G)]^+ \end{aligned} \tag{12}$$

In (11), $\xrightarrow{\text{w}}$ denotes weak convergence with a slight abuse of notation, since the convergence is not uniform in s close to 0.

Theorem 3 states that for large t, $t^{-d/4}\xi^t$ is approximated by η, whose components are specified by $\Psi_s = N_s * \Phi$. This convolution is a smooth function of x in $\mathbb{R}^d$, on account of N_s. Equivalently, Ψ_s is the Gaussian random field with mean 0 and covariance

$$E[\Psi_s(x)\,\Psi_s(y)] = \frac{1}{(4\pi s)^{d/2}} \cdot e^{-|y-x|^2/4s}$$

Note that whereas Φ is random, N_s is not. So the evolution of the system on this macroscopic scale is specified (in the limit) completely by its initial data. As indicated by (12), there is only one type of particle locally, in accordance with the non-mean-field rate of decay $t^{-d/4}$.

Some motivation for the behavior in (11)–(12) is provided by the following observation. Set

$$\begin{aligned} W(t) &= E[\xi_{\mathrm{B}}(t) - \xi_{\mathrm{A}}(t) \mid \xi(0)] \\ W^t(G; s) &= W(\sqrt{t}\, G; ts) \end{aligned} \tag{13}$$

That is, W is the mean signed measure of ξ conditioned on knowledge of the initial configuration, and W^t is the corresponding rescaled measure. The analog of (11)–(12) is then

$$t^{-d/4} W^t \xrightarrow{\mathrm{w}} \eta_{\mathrm{B}} - \eta_{\mathrm{A}} = N * \Phi \tag{14}$$

It is easy to see that the density $\bar{u}(x, s)$ of $N * \Phi$ is given by

$$\frac{\partial \bar{u}}{\partial s} = \frac{1}{2} \Delta \bar{u}, \qquad \bar{u}(\cdot, 0) = \Phi \tag{15}$$

So, one might expect that (14) is obtainable in some simpler manner.

This is in fact the case: replace the system $\mathrm{A} + \mathrm{B} \to$ inert by the corresponding system of independent random walks. That is, particles of each type move as before, but there is no interaction. If one starts with a Poisson random measure and scales space and time as before, then on account of the central limit theorem, random walks are replaced by Brownian motion as $t \to \infty$ and one obtains the density $\bar{u}$ in the limit. One can verify, on the other hand, that the expectations for the interactive and noninteractive systems are the same. (When an A and B pair meet in the interactive system, they can be thought of as sticking together and forming a combined particle of "neutral" type.) Therefore, (14) must also hold for the system $\mathrm{A} + \mathrm{B} \to$ inert (for all d). The point in Theorem 3 is that the system η in $d < 4$ in fact evolves deterministically for a prescribed initial configuration, with only one type of particle ever present locally. The result is proved in ref. 4.

The asymptotic behavior of ξ^t for the case $d > 4$ is simpler. Since, according to Theorem 1, mean-field reasoning gives the right result in this case, the limiting process η should not have any spatial structure. So it should also have no dependence on the particular initial configuration, since the initial measure is ergodic. It is therefore not surprising that one obtains the following result.

Theorem 4. For $d > 4$,

$$t^{1-d/2}\xi^t \xrightarrow{\mathrm{w}} \eta \qquad \text{as} \quad t \to \infty \tag{16}$$

where $\eta = (\eta_A, \eta_B)$ is nonrandom and has densities C_d/s, $C_d > 0$.

Note that, unlike the case $d < 4$, both types of particles are always present locally, in fact in the same concentration.

The asymptotic behavior of ξ^t for the case $d = 4$ is the most difficult to work with. This is the critical case: annihilation as given by both local fluctuations and mean-field reasoning plays a role. The result of this is that both types of particles are present locally, but in different concentrations. So one can neither interpret the limit η in terms of differences of densities as in $d < 4$ or as a scalar as in $d > 4$. Instead, η has a density which satisfies the system of equations given below.

Theorem 5. For $d = 4$,

$$t^{-1}\xi^t \xrightarrow{\mathrm{w}} \eta \qquad \text{as} \quad t \to \infty \tag{17}$$

where η has density $u(x, s) = (u_A(x, s), u_B(x, s))$ given by

$$\begin{aligned} \frac{\partial u_A}{\partial s} &= \frac{1}{2} \Delta u_A - \Gamma u_A u_B \\ \frac{\partial u_B}{\partial s} &= \frac{1}{2} \Delta u_B - \Gamma u_A u_B \end{aligned} \tag{18}$$

$\Gamma > 0$, with

$$u_A(\cdot, 0) = \Phi^-, \qquad u_B(\cdot, 0) = \Phi^+ \tag{19}$$

In (19), Φ^- and Φ^+ denote the negative and positive parts of white noise. Since Φ has infinite variation everywhere, one really needs to be more precise about what one means by these quantities. With some effort, one can define them as limits in terms of η_s as $s \downarrow 0$. The blowup that occurs here due to rescaling as $s \downarrow 0$ is consistent with the increasingly rapid annihilation of particles as given by (18) when u_A and u_B both increase proportionally. This is explained in ref. 4.

4. SPATIAL STRUCTURE—MICROSCOPIC BEHAVIOR

We wish to investigate the asymptotic behavior of ξ more carefully. Theorems 3–5 give its behavior when space is rescaled by $\sqrt{t}$, which is the proper scaling to follow the evolution of the process in terms of the local densities of its particles. One may instead wish to view the process on the microscopic level, so that the evolution of ξ maintains its random character. The natural scaling to use in this case is t^β, where $\beta = 1/4$ for $d \leqslant 4$ and $\beta = 1/d$ for $d \geqslant 4$. Referring to Theorem 1, we see that this is what one needs to compensate for the decay in densities of the A and B particles. We therefore set

$$\hat{\xi}(G; x, t) = \xi(t^\beta G + x; t), \qquad E \subset \mathbb{R}^d \tag{20}$$

One can consider, for instance, cubes G which are centered at 0. Then $t^\beta G + x$ provides a "window" around x which is large enough to view the local behavior of ξ. Note that since $\beta < 1/2$ in all cases, the densities calculated in Theorems 3–5 are all asymptotically constant on this scale.

We make a number of basic observations which will make the behavior of $\hat{\xi}$ clearer. We note that for the system of particles A + B → inert and large t:

1. Not much annihilation of particles occurs over $[t, t(1+\delta)]$ for $0 < \delta \ll 1$. This is not surprising, since, according to Theorem 1 and Theorems 3–5, no sudden jumps occur in ρ or the distribution of ξ.
2. Consequently different particles move (more or less) independently over this time period.
3. This motion produces a random field at time $t(1+\delta)$ which is locally (more or less) homogeneous and Poisson in the two types of particles when conditioned on the configuration at time t. So the process is locally a convex combination of Poisson random fields.
4. On account of Theorems 3–5, the following must be true:

   a. For $d < 4$, the Poisson random fields have only one type of particle locally.

   b. For $d > 4$, both types are always present with equal densities depending only on t.

   c. For $d = 4$, both types are present locally, but with varying densities.

To phrase the conclusions in (3) and (4) more precisely, we observe that the limiting random measure η in Theorems 3–5 always has a random density $u(x, s) = (u_A(x, s), u_B(x, s))$, with $x \in \mathbb{R}^d$, $s > 0$. Since u is homogeneous in space and scales in time, we restrict ourselves to $x=0$, $s=1$, and denote by

$$F(a, b) = P[u_A(0, 1) \leqslant a, u_B(0, 1) \leqslant b] \tag{21}$$

the corresponding distribution function. Note that in $d<4$, all of the mass is located at $a=0$ or $b=0$, and in $d>4$, everything is at one point. Also, let $\mathscr{P}_{a,b}$ denote the homogeneous Poisson random field with means a and b for the A- and B-type particles, and set

$$\mathscr{P}_F = \int \mathscr{P}_{a,b}\, dF(a, b) \tag{22}$$

that is, $\mathscr{P}_F$ is the convex combination of these fields weighted according to F.

The conclusion in observations 3 and 4 can be stated as follows.

Theorem 6. For all d,

$$\hat{\xi}(x, t) \xrightarrow{\text{w}} \mathscr{P}_F \qquad \text{as} \quad t \to \infty \tag{23}$$

It is clear by translation invariance that the convergence is uniform in x. The above statement can of course also be phrased in terms of convergence over a range of times instead of just over space (more in analogy with the phrasing in Theorems 3–5). Letting time range over $[t, t+t^{2\beta}]$, for instance, the limit is then a system of independent random walks with invariant distribution $\mathscr{P}_F$.

Of substantial interest is the question of what one can say about the behavior of ξ over space scales that are intermediate to $\sqrt{t}$, which was employed in the macroscopic scaling, and t^{β}, which has been employed here. One can, for instance, inquire as to how quickly the density of a particle type, which is in the local minority in some intermediate-sized region, decreases in $d<4$. In analogy with Theorem 2, one would expect that once one type of particle becomes dominant locally, then it drives down the local density of the other type "exponentially" fast. (Since the density of the dominant type is also tending to 0 at rate $t^{d/4}$, the time scale needs to be attenuated by a corresponding factor.) We need to balance against this the influx of particles from the boundaries of other regions where the dominance relationship is reversed. For regions which are not too close to such boundaries, the rate of such influx should also be "exponentially" small. In $d=4$, on the other hand, the density of the locally dominant type is just of magnitude t^{-1}. So annihilation of the minority type only occurs at this rate; this is slow enough so that particles from within distances of magnitude $\sqrt{t}$ (including where the other type is dominant) meanwhile have sufficient time to diffuse in. In $d>4$, neither particle type achieves local dominance. Although these questions have not yet been treated rigorously, the basic geometric picture is fairly clear.

Given that one has enough confidence to accept (in $d<4$) such a nearly complete local absence of minority-type particles, one can apply this reasoning to related questions, for instance, the distance between clusters of different particle types. As explained by Redner and Leyvraz[10], one has reason to expect an intercluster distance of magnitude $t^{3/8}$ in $d=1$. This conclusion also results from our setting, the idea being as follows. After scaling space by $t^{1/2}$ at time t and the concentration by $t^{1/4}$, the behavior of η_A and η_B is given by $N_1 * \Phi$ (Theorem 3). The crossover from type A to type B particles occurs where the value of the curve is 0. There, the curve will have nonzero slope. Measuring from this point, the integral of $N_1 * \Phi$ over an interval of length $x \ll 1$ is of order x^2. Returning to the original scale, one should see on the order of $t^{1/4}x^2$ particles on the corresponding interval of length $t^{1/2}x$. The intercluster distance should be on the scale obtained by setting $t^{1/4}x^2 = 1$; then the expected number of particles in such an interval will be of order 1. This substitution $x = t^{-1/8}$ gives $t^{1/2}x = t^{3/8}$ as the order of the anticipated length of the intercluster distance in $d=1$. One can also look at related questions in $d=2, 3$, although there the geometry of the clusters of particle types will of course be more complicated, and one has to put more thought into how to measure the intercluster distance. As before, one can analyze the local behavior of $N_1 * \Phi$.

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