The Effect of Static Disorder and Reactant Segregation on the A + B → ∅ Reaction

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We derive the long-time behavior of the A + B → ∅ reaction in two dimensions, finding a universal exponent and prefactor in the absence of disorder. Sufficiently singular disorder leads to a (sub)diffusion-limited reaction and a continuously variable decay exponent. Pattern matching between the reactant segregation and the disorder is not strong enough to affect the long-time decay.

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I. INTRODUCTION

The A + B → ∅ reaction shows quite different behavior from the A + A → ∅ reaction because of the distinguishability of A and B reactants. Ovchinnikov and Zeldovich, Toussaint and Wilczek, and Kang and Redner first noticed the diffusion-controlled kinetics that can arise from reactant segregation. Heuristic arguments predict that the concentration decays from an initially random configuration as \( c_A(t) \sim C_d t^{-d/4} \) in \( d \) dimensions for \( d \leq 4 \). This scaling arises essentially from the competition between diffusion and randomness in the initially random reactant densities. Simulation studies have confirmed these analytical results (see, for example, [3]). The results have been extended to the case of fractal media (see, for example, [3,4]). It turns out, however, that two dimensions is the upper critical dimension for this system, and so collective motions of the reactants become important for \( d \leq 2 \). By upper critical dimension, we mean the dimension below which perturbation theory diverges at long wavelengths. Standard arguments [3], then, suggest that mean field theories such as the standard reaction diffusion partial differential equation will fail in two dimensions and below. The most important consequence of these collective effects, and what causes the standard theories to fail, is renormalization of the effective, or apparent, reaction rate. We will derive this natural renormalization of the effective rate from renormalization group arguments. Disorder in the medium can also influence the reaction kinetics. The most important effect is renormalization of the effective diffusion constant. Anomalous diffusion can occur if the quenched defect potential is sufficiently long ranged.

The reaction A + B → ∅ with Poissonian random initial conditions has recently been analyzed via field theoretic techniques [5]. By placing bounds on the exact, stochastic partial differential equations (PDEs) for the concentration profiles, it was shown that \( c_A(t) \sim C_d t^{-d/4} \) for \( 2 < d \leq 4 \), and the coefficient \( C_d \) was determined. An approximate mean field theory was shown to apply for long times for \( d > 2 \). Rigorous bounds on the decay have been derived in all dimensions for the case of infinite reaction rate [10].

We here analyze the behavior of the A + B → ∅ reaction in two dimensions in the presence of correlated disorder. We describe the reaction and our motivation for this study in section II. We express the reaction in terms of a master equation. In section III we map the master equation onto a field theory using the coherent state representation [8]. In section IV we derive an exact, stochastic partial differential equation that can be used to analyze this reaction system. We state the most natural form of mean field theory and show that it applies for both short and long times. In section V we analyze the long-time behavior of the field theory using renormalization group theory, finding segregation of reactants into isolated domains. We match the RG flows to a result that is asymptotically exact in the diffusion-limited, matching limit. Through this procedure we are able to determine the universal exponent and prefactor, \( C_d \), for the concentration decay. We confirm our results with numerical solutions of the exact, stochastic PDEs for the concentration profile. We then turn to the effect of disorder on the segregation. For sufficiently singular disorder, the decay exponent changes. We calculate the new exponent, which comes solely from the fact that the reaction is (sub)diffusion limited. We conclude with a discussion of our results in section VI.

II. THE A + B → ∅ REACTION

The mathematical problem we consider is one where two distinct types of reactants, A and B, diffuse on a two-dimensional square lattice. When the reactants occupy the same lattice site, they react with reaction rate \( \lambda \). The reactants do not interact other than through the reaction. The reactants do, however, move in a potential field, \( v \), created by disorder. The energy of reactant A at position \( x \) is \( v(x) \), and the energy of reactant B at position \( x \) is \( -\nu(x) \). We take the correlators of the potential field to be long-ranged. Specifically, we consider the correlation function whose Fourier transform is given by \( \hat{\chi}_{vv}(k) = \int dx \exp(i k \cdot x) \chi_{vv}(x) = \gamma/k^2 \). It turns out than any disorder with a correlation function less singular than this at long wavelengths is technically irrelevant. Such disorder does not lead to any interesting, new scaling behavior.
Given this description, we can state the master equation that governs changes in the configuration of the lattice \[ \text{[12]} \]. The master equation relates how the probability \( P \) of a given configuration of particles on the lattice changes with time:

\[
\frac{\partial P(\{m_i\}, \{n_i\}, t)}{\partial t} = \frac{D_A}{\Delta r^2} \sum_{i,j} \left[ T^A_{ij}(m_j + 1)P(\ldots, m_i - 1, m_j + 1, \ldots, t) - T^A_{ij}m_iP \right] + \frac{D_B}{\Delta r^2} \sum_{i,j} \left[ T^B_{ij}(n_j + 1)P(\ldots, n_i - 1, n_j + 1, \ldots, t) - T^B_{ij}n_iP \right] + \frac{\lambda}{\Delta r^2} \sum_i \left[ (m_i + 1)(n_i + 1)P(\ldots, m_i + 1, \ldots, n_i + 1, \ldots, t) - m_in_iP \right] \tag{1}
\]

Here \( m_i \) is the number of A particles on site \( i \), and \( n_i \) is the number of B particles on site \( i \). The summation over \( i \) is over all sites on the lattice, and the summation over \( j \) is over the nearest neighbors of site \( i \). The lattice spacing is given by \( \Delta r \). The diffusive transition matrix for hopping from site \( i \) to a nearest neighbor site \( j \) is given by \( T^A_{ij} = [1 + \beta(v_i - v_j)/2] \) and \( T^B_{ij} = [1 - \beta(v_i - v_j)/2] \). The inverse temperature is given by \( \beta = 1/(k_BT) \).

The formal motivation for our study is the theoretical investigation of the effects of disorder on the \( A + B \to 0 \) reaction in two dimensions. Our model, however, could be considered roughly to represent a reaction between ions on the surface of a crystalline ionic lattice. The cubic substrate lattice has dislocation line pairs, which form line vacancies or line interstitials. These defects are static and generate a random, quenched electrostatic potential on the surface. Each line charge produces an electrostatic field on the surface of the crystal that grows logarithmically with distance from the charge. Charge neutrality, however, implies that the two-dimensional density-density correlation function of the defects should vanish as \( k^2 \) for small \( k \). Since the potential is given by the convolution of the defect density with Coulomb’s law, the appropriate form of the potential-potential correlation function on the surface of the crystal is that assumed above. Moreover, the statistics of the potential field are very likely Gaussian at small \( k \) \[13\]. On the surface of the crystal, the reaction \( A + B \to 0 \) occurs, where A and B are ions of opposite charge. The ions interact with a \( 1/r \) Coulomb potential, which turns out to be irrelevant in two dimensions. Technically, this means that the \( 1/r \) interaction will not affect any flow equations produced by renormalization group theory. This interaction can only affect the “matching limit.” In order to avoid any significant effects in the matching limit, and to facilitate segregation of the reactants, we may want to include a low density of counter ions. The density of counter ions should be lower than that of the defects, so as not to screen out the effects of the disorder as well. We will not consider these details further, however, and we will be content to analyze the mathematical problem defined by Eq. (1) of reactants A and B interacting with the random potential but not with each other.

III. MAPPING TO THE FIELD THEORY

We will write the reaction in terms of a field theory \[12\]. For initial conditions, we will assume that the A and B particles are placed at random on the lattice. The initial concentrations for A and B at any given site will, therefore, be Poissonian random numbers. So as to reach the most interesting scaling limit, we take the initial average densities to be the same, \( \langle c_A(x,0) \rangle = \langle c_B(x,0) \rangle = n_0 \). For simplicity, we assume \( D_A = D_B = D \). No adsorption or unimolecular desorption is allowed. The random potential is incorporated with the replica trick \[14\], using \( N \) replicas of the original problem. The concentrations averaged over initial conditions are given by

\[
\langle c_A(x,t) \rangle = \lim_{N \to 0} \langle a(x,t) \rangle \\
\langle c_B(x,t) \rangle = \lim_{N \to 0} \langle b(x,t) \rangle , \tag{2}
\]

where the average on the right hand side is taken with respect to \( \exp(-S) \). In the field-theoretic formulation, the random initial conditions have already been averaged over. The fields \( a, \tilde{a}, b, \tilde{b} \) remain to be integrated over in the field theory. Before averaging, they have no physical meaning and may assume complex values. The action is given by \( S = S_0 + S_1 + S_2 + S_3 \):

\[
S_0 = \int d^d x \int_0^{t_f} dt \tilde{a}_a(x,t) \left[ \partial_t - D \nabla^2 + \delta(t) \right] a_a(x,t) + \int d^d x \int_0^{t_f} dt \tilde{b}_a(x,t) \left[ \partial_t - D \nabla^2 + \delta(t) \right] b_a(x,t)
\]

\[
S_1 = \int d^d x \int_0^{t_f} dt \left[ \lambda_1 \tilde{a}_a(x,t)a_a(x,t)b_a(x,t) + \lambda_2 \tilde{b}_a(x,t)a_a(x,t)b_a(x,t) \right]
\]

\[
S_2 = -n_0 \int d^d x \left[ \tilde{a}_a(x,0) + \tilde{b}_a(x,0) \right]
\]

\[
S_3 = \frac{\beta^2 D^2}{2} \int dt_1 dt_2 \int d^d x_1 d^d x_2 \left( \delta(k_1 + k_2 + k_3 + k_4) \times \left[ \tilde{a}_{a_1}(k_1, t_1) \tilde{a}_{a_2}(k_2, t_2) - \tilde{b}_{a_4}(k_1, t_1) \tilde{b}_{a_2}(k_2, t_1) \right] \times \left[ \tilde{a}_{a_3}(k_3, t_2) \tilde{a}_{a_4}(k_4, t_2) - \tilde{b}_{a_4}(k_3, t_2) \tilde{b}_{a_4}(k_4, t_2) \right] \right)
\]
\[ \chi_{v}(k) = \frac{\gamma}{k^2} . \quad (4) \]

This correlation function leads to anomalous diffusion in two dimensions.

**IV. STOCHASTIC EQUATIONS OF MOTION**

An exact expression for the concentration profiles can be derived by performing a Hubbard-Stratonovich transformation on (3) and integrating out the fields \( \tilde{a} \) and \( \tilde{b} \):

\[ \partial_t a_{\eta v} = D \nabla^2 a_{\eta v} + \beta D \nabla \cdot (a_{\eta v} \nabla v) - \lambda_1 a_{\eta v} b_{\eta v} + (\eta_2 + i\eta_1) a_{\eta v} \]
\[ \partial_t b_{\eta v} = D \nabla^2 b_{\eta v} - \beta D \nabla \cdot (b_{\eta v} \nabla v) - \lambda_2 a_{\eta v} b_{\eta v} + (\eta_3 + i\eta_4) b_{\eta v} \]
\[ a_{\eta v}(x, 0) = b_{\eta v}(x, 0) = n_0 , \quad (5) \]

where the real, Gaussian, random field \( \eta \) has zero mean and variance

\[ \langle \eta_i(x, t) \eta_j(x', t') \rangle = \lambda_3 \delta_{ij} \delta(x - x') \delta(t - t') . \quad (6) \]

The potential field \( v(x) \) enters the equations in the expected fashion. The physical concentration is given by averaging the solution over the random fields \( \eta \). The solution before this averaging has no physical meaning—it is complex. Recall, also, that the random initial conditions have already been averaged over. It is for this reason that we have deterministic initial conditions in Eq. (3).

A natural approximation for the concentration profiles is the standard reaction diffusion PDE with Poissonian random initial conditions:

\[ \partial_t c_{A\gamma} = D \nabla^2 c_{A\gamma} + \beta D \nabla \cdot (c_{A\gamma} \nabla v) - \lambda c_{A\gamma} c_{B\gamma} \]
\[ \partial_t c_{B\gamma} = D \nabla^2 c_{B\gamma} - \beta D \nabla \cdot (c_{B\gamma} \nabla v) - \lambda c_{A\gamma} c_{B\gamma} . \quad (7) \]

Unlike the mean field theory that arises from a direct saddle point approximation to action (3), which has uniform initial conditions, these equations are valid for both short and long times. At intermediate times, they are incorrect, because they do not capture the renormalization of the effective rate constant. Our renormalization group treatment will predict how the effective reaction rate at length scale \( \Delta \) scales with \( l \). One should interpret this result as the effective, renormalized reaction rate observed at this length scale. With this interpretation, an approximate means of correcting Eq. (7) would be to use the running coupling \( \lambda(l) \) in place of the original rate constant \( \lambda \). As we will see, however, the reaction becomes diffusion limited at long times, and so the actual value of \( \lambda(l) \) does not matter.

**V. FLOW EQUATIONS AND THE MATCHING LIMIT**

We use renormalization group theory to analyze the long-time behavior of the action (3). The flow equations are similar to those for the \( A + A \rightarrow \emptyset \) reaction [4]. The flow equations in two dimensions, to one loop order, are

\[ \frac{d \ln \lambda_0}{dt} = 2 \]
\[ \frac{d \ln \lambda_3}{dt} = - \frac{\lambda_3}{4\pi} - \frac{\beta^2 \gamma}{4\pi} \]
\[ \frac{d \ln \lambda_3}{dt} = - \frac{\lambda_3}{4\pi} - \frac{\beta^2 \gamma}{4\pi} \]
\[ \frac{d \beta^2 \gamma}{dt} = 0 \ . \quad (8) \]

The dynamical exponent is given by

\[ z = 2 + \frac{\beta^2 \gamma}{4\pi} . \quad (9) \]

We note that if the sign of the attraction for the A and B reactants to the potential were the same, instead of opposite, the second term in the flow equations for \( \lambda_i \) would be \( +3\beta^2 \gamma/(4\pi) \).

The flow equations are integrated to a time such that

\[ t(t^*) = t e^{-\int_{t}^{t^*} z(t)dt} = t_0 . \quad (10) \]

The matching time, \( t_0 \), is chosen to be relatively small so that mean field theory can be applied. It must be non-zero, however, so as to remain within the scaling regime. We note that for large \( t^* \), the renormalized initial density is large, the renormalized reaction rate is small, and the renormalized disorder strength remains at the bare value. At these short times, the diffusion is normal with essentially the bare diffusivity.

We can derive the appropriate matching theory by considering the renormalized reaction within the original
master equation formulation. The Poissonian initial densities become asymptotically Gaussian since they are so large. We consider one lattice site and assume without loss of generality that \(c_A(x,0;l) > c_B(x,0;l)\). Because the renormalized density is large, the reaction terms will dominate. We see that \(c_B(x,t;l)\) will be driven to a value of order \(1/[k(l)c_A(x,t;l)]\). This will happen in a time of the order of \(1/[k(l)c_A(x,t;l)]\), before any significant diffusion can take place. For times \(t > t_1\), then, we will find that there is either a significant amount of A or B at each lattice site, but not both. The matching limit is, therefore, like the infinite reaction rate limit, as long as the renormalized reaction rate is not too small. The single-species lattice sites are the renormalized vestige of the long-time reactant segregation that occurs in the \(A + B \rightarrow \emptyset\) reaction. In this infinite reaction rate limit, the field \(\varphi(x,t) = c_A(x,t;l) - c_B(x,t;l)\) exactly satisfies the diffusion equation:

\[
\frac{\partial \varphi}{\partial t} = D\nabla^2 \varphi
\]

\[
\varphi(x,0) = \left(\frac{2n_0(l)}{D\Delta^2}\right)^{1/2} \hat{N}(x),
\]

where \(\hat{N}(x)\) is a unit Gaussian variable. The potential has been omitted from Eq. (11) because it does not affect the short time behavior. Furthermore, we can invert the equation and is Gaussian at \(x = 0\). The field \(\varphi\) is a Gaussian random variable, since it satisfies a linear equation and is Gaussian at \(t = 0\). The average concentration of A and B at the matching time is, then, given by

\[
\langle c_A(t_0;l) \rangle = \langle c_B(t_0;l) \rangle = \frac{1}{2} \langle |\varphi(t_0)| \rangle
\]

\[
= \left[ \frac{\langle \varphi^2(t_0) \rangle}{2\pi} \right]^{1/2} = \left[ \frac{2n_0(l)G(0,2t_0)}{2\pi} \right]^{1/2},
\]

where \(G(x,t) = \exp(-4Dt/\Delta^2)J_0^2(2Dt/\Delta^2)/\Delta^2\) is the two-dimensional lattice Green’s function evaluated at \(x = 0\). These average concentrations are independent of position, once we have averaged over the initial conditions in Eq. (11). For \(t_0 \gg \Delta^2/2D\), we find

\[
\langle c_A(t_0;l) \rangle = \langle c_B(t_0;l) \rangle = \left[ \frac{n_0(l)}{8\pi^2D t_0} \right]^{1/2}.
\]

To confirm this matching limit, we present in Figure 1 numerical solutions of the exact stochastic PDEs (3). The PDEs were integrated with an explicit Euler method [10] on square 256 \(\times\) 256 or 512 \(\times\) 512 grids. We clearly see three time regimes. During the first, \(0 < t < \Delta r/(k\sqrt{n_0})\), the minority reactant disappears at each lattice site. During the second, \(\Delta r/(k\sqrt{n_0}) < t < \Delta^2/D\), the reaction is diffusion limited, but lattice effects are present. During the third, \(t > \Delta^2/D\), the reaction is diffusion limited, and we have reached asymptotic scaling. A limited range of the asymptotic scaling regime is shown in Figure 1. Simulations run to longer time would clearly show this scaling. Note that the range of data shown in Figure 1 is enough to verify Eq. (12) in the short-time, matching limit, which is all we require. Furthermore, as shown below, the scaling of the concentration with \(\sqrt{n_0}\) for all but very short times, which is clearly seen in Figure 1, is enough to establish the exponent of the long-time decay.

Alternatively, we can do direct perturbation theory on Eq. (3). We define \(\psi = a - b, \phi = a + b\). We use the reference state \(a_0 = b_0 = 1/(n_0^{-1} + \lambda t)\) and perturb in the parameter \(\lambda_3\). The first order corrections are

\[
\langle \psi_1^2 \rangle = 2\lambda_3 \int_0^t dt' G(0,2(t-t'))
\]

\[
\langle \phi_1^2 \rangle = -\frac{2\lambda_3}{(n_0^{-1} + \lambda t)^2} \int_0^t dt' G(0,2(t-t'))
\times \langle n_0^{-1} + \lambda t \rangle^2.
\]

Furthermore, \(\psi\) is Gaussian to this order so we find \(\langle |\psi| \rangle = (2\langle \psi^2 \rangle/\pi)^{1/2}\). To second order we find

\[
\langle \phi_2 \rangle = \frac{\lambda_3}{2(n_0^{-1} + \lambda t)^2} \int_0^t dt' \left[ \langle \psi_1^2(t') \rangle - \langle \phi_1^2(t') \rangle \right]
\times \langle n_0^{-1} + \lambda t \rangle^2.
\]

In Figure 2 we plot the exact results as well as the perturbative results. We see that this perturbation theory for \(\langle |a| \rangle\) is accurate until the reaction becomes diffusion limited. In the long time regime, the perturbation theory fails. Interestingly, the first order result for \(\langle \psi^2 \rangle\) seems to be exact. Asymptotically we have \(\langle \psi_1^2 \rangle \sim n_0/(4\pi Dt)\), which we have seen is correct in the long-time, diffusion-limited regime. But \(\langle \psi_1^2 \rangle\) also agrees with the exact \(\langle \psi^2 \rangle\) for all times and for all initial densities that we have examined numerically. Note that the field \(\psi(x,t)\) has already been averaged over the initial conditions, and so it does not equal \(c_A(x,t) - c_B(x,t)\) for a given instance of the initial conditions. This identification does hold, however, when each lattice site contains only A or B.

Finally, we can also solve the standard reaction diffusion PDEs (3) with Poissonian initial conditions. Upon doing so, we would find that the reaction diffusion PDEs are accurate for short times. They are also accurate for long times, since they correctly capture the diffusion-limited nature of the reaction. Only for intermediate
times do we see a discrepancy. It is only during these times that the renormalization of the effective rate constant has an effect on the observed density. The discrepancy becomes smaller for larger values of $n_0$. It would not be noticeable for the parameters of Figure 1, for example.

We are now in a position to combine, with confidence, the RG flow equations and the matching theory. We take $t_0 \gg \Delta r^2/(2D)$ to be in the scaling regime. There will be no significant renormalization of $D$ if $t_0$ is not chosen too large ($t_0 = \Delta r^2/D$ is reasonable). Using Eqs. (8), (11), and (14) we find

$$\langle c_A(t) \rangle = \langle c_B(t) \rangle = \left( \frac{n_0}{8\pi^2 D t} \right)^{1/2} \frac{1}{(t/t_0)^{3/2}}, \quad (17)$$

with

$$\delta = \left( 1 + \frac{8\pi}{\beta^2 \gamma} \right)^{-1}. \quad (18)$$

VI. DISCUSSION

We have found the exponent and prefactor for the concentration decay of the $A + B \rightarrow \emptyset$ reaction at long times. These quantities are universal in the absence of disorder. Unlike the $A + A \rightarrow \emptyset$ reaction, we do not have a logarithmic correction in the upper critical dimension $d_c = 2$ in the absence of disorder. This is because $\lambda_i(l^*)$ has no effect on the matching limit. Like the $A + A \rightarrow \emptyset$ reaction, however, the correction to the decay exponent due to the disorder is exactly the same correction that occurs to the mean-square displacement exponent. This phenomenon is not accidental and occurs precisely because the reaction becomes diffusion limited in the long-time limit. For this reason, we do not expect higher loop calculations of the flow equation to produce terms that modify $\beta^2 \gamma$. We have checked that the two-loop flow equations do not modify $\beta^2 \gamma$. Higher order calculations may modify the flow equations for $\lambda_i$, but this will not affect the concentration decay at long times. In fact, if the flow equations for $\lambda_i$ were unchanged by higher orders of $\beta^2 \gamma$, the matching limit would be like the infinite reaction rate limit only for $\beta^2 \gamma < 4\pi$. Note that an extension of the RG argument gives $\langle c_A(t) \rangle \sim \sqrt{n_0}/[\sqrt{\pi}(8\pi D t)^{3/4}]$ for $1 \leq d < 4$ in the absence of disorder, extending the results of (4) to one and two dimensions.

The reactants A and B are attracted to regions of the potential that have opposite signs. One might expect that the segregation of the reactants will be biased by the presence of the disorder. Indeed, pattern matching between the reactant segregation and the potential does occur to some extent: when A and B have opposite attraction for the potential, the effective reaction rate is decreased, asymptotically reaching zero. If the attraction is the same, the effective reaction rate is increased, asymptotically reaching a non-zero fixed point value. The dominant mechanism for the long-time decay of the reactants, however, is diffusion-limited decay of initial density variations. These density variations are, in our model, not affected by the disorder.

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Figure 1. Presented are numerical solutions of the exact, stochastic PDEs (3) for $n_0 = 10^2, 10^4$, and $10^5$. For each solution we show $\langle a \rangle$ (solid), $\langle b \rangle$ (solid), $\langle a - b \rangle$ (short dashed), and $\sqrt{\langle \psi^2 \rangle}$ (long dashed). We have used $k = D = \Delta r = 1$, $\beta^2 \gamma = 0$ in this and all figures.

Figure 2. Presented are the perturbative results (13) and (14) as well as the exact results for $n_0 = 10$. We show $\langle a \rangle$ (solid), $\langle b \rangle$ (solid), $a_0 + \langle a_2 \rangle$ (long dashed), $\sqrt{\langle \psi^2 \rangle}$ (solid), and $\sqrt{\psi_1^2}$ (short dashed).
