LOCALISATION TRANSITION OF A DYNAMIC REACTION FRONT

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Abstract: We study the reaction-diffusion process $A + B \rightarrow \emptyset$ with injection of each species at opposite boundaries of a one-dimensional lattice and bulk driving of each species in opposing directions with a hardcore interaction. The system shows the novel feature of phase transitions between localised and delocalised reaction zones as the injection rate or reaction rate is varied. An approximate analytical form for the phase diagram is derived by relating both the domain of reactants $A$ and the domain of reactants $B$ to asymmetric exclusion processes with open boundaries, a system for which the phase diagram is known exactly, giving rise to three phases. The reaction zone width $w$ is described by a finite size scaling form relating the early time growth, relaxation time and saturation width exponents. In each phase the exponents are distinct from the previously studied case where the reactants diffuse isotropically.

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1. Introduction

The behaviour of the two-species reaction-diffusion system $A + B \rightarrow \emptyset$ has been widely studied over the last decade[1]. For a given realisation of this process, choices must be made for the type of motion allowed for the $A$ and $B$ reactants and the environment in which the reactions take place. For the case of purely diffusive reactants, situations of particular interest have been i) the evolution of a system of initially segregated reactants on an infinite lattice [2] and ii) the steady-state behaviour on a finite lattice with fixed currents of the $A$ species at the left boundary and the $B$ species at the right boundary [3]. In both cases a reaction zone (a region where reactions occur with non-negligible rate) is formed between the two domains of reactants. The scaling of the reaction zone has been studied within the mean-field approximation [2, 3] giving a good account of the behaviour in dimension two and above [4]. However, in one dimension the confined geometry makes diffusive mixing of reactants inefficient. This in turn leads to significant density fluctuations which invalidate the mean-field approach: distinct non-mean-field scaling behaviour is seen [5]–[13]. It is also known [14, 15] that in the case of homogeneous initial conditions (both species are distributed randomly) the model with hard core exclusion and biased diffusion of reactants is in a different universality class to that of the reaction-diffusion system where the reactants diffuse isotropically.

In the present work we show that on the one-dimensional finite system the introduction of hard-core exclusion between reactants together with opposed bulk driving, i.e. $A$ particles move stochastically to the right and $B$ particles to the left, produces the novel phenomena of phase transitions when the reaction rate and boundary injection rate are varied. In particular we find transitions between a phase where the reaction zone is localised (occupying a vanishingly small fraction of the system in the thermodynamic limit) to phases where it is delocalised (filling the whole system).

Let us first discuss the situation where no bulk drive is imposed: the reactants are not driven but just diffuse. On the infinite system with initially segregated reactants, the reaction zone width $w$ is expected to grow (in the scaling regime) as $t^\alpha$. Mean-field theory predicts $\alpha = 1/6$ [2] whereas one-dimensional simulations and scaling arguments point to $\alpha = 1/4$ [4, 8, 11, 12, 13]. In fact an RG calculation [13] suggests logarithmic corrections $w \sim t^{1/4}(\log t)^{1/2}$ which may explain the slow convergence to $\alpha = 1/4$ found in several simulations [8, 11, 12].

In the complementary situation of a finite lattice of size $L$ where reactant $A$ is injected at the left boundary (site 1) and reactant $B$ is injected at the right (site $L$), the system attains a steady state as $t \rightarrow \infty$. The scaling of the reaction zone is predicted in mean-field theory to depend on the injected flux $j$, reaction rate $\lambda$ and diffusion constant of reactants $D$ through $w \sim (D^2/(j\lambda))^{1/3}$ [3]. This is distinct from the one-dimensional case where scaling
arguments \[8, 12\] and an RG calculation \[10\] suggest \( w \sim (D/j)^{1/2} \). Recently it has been shown that when the wandering as well as the intrinsic contribution to the reaction zone width is taken into account, RG and Monte Carlo simulations predict \( w \sim (D/j)^{1/2}(\log L)^{1/2} \) \[13\].

We now consider the effects of hardcore exclusion (though still with no drift) with reactions occurring between particles on neighbouring sites. On the infinite system the exclusion is not expected to affect the value of \( \alpha \). However, one effect of exclusion on the finite lattice is that that current can no longer be fully controlled from outside the system, since a reactant can only enter if a boundary site is empty. If one attempts to inject reactants at a finite rate the resultant current is in fact \( \mathcal{O}(1/L) \). To see this note that in the bulk the current is \( D \frac{d\rho}{dx} \) where \( \rho \) is the concentration. Now, due to exclusion the maximum value \( \rho \) can take is one, therefore, if we assume a reaction zone \( w \ll L \) the concentration gradient, and hence the current, is of order \( 1/L \). From this we deduce \( w \sim 1/j^{1/2} \sim L^{1/2} \) confirming that the reaction zone is localised i.e. \( w/L \to 0 \) as \( L \to \infty \).

In order to relate the behaviour on the finite system to the infinite system with initially segregated reactants a finite size scaling form appears natural. The early time growth of the reaction zone on the finite system should be the same as that on the infinite system since the effect of the boundaries will not have been felt: the boundaries will only influence the dynamics after a time \( t \sim L^2 \). Therefore, ignoring any logarithmic corrections, we propose a finite size scaling form

\[
w \sim t^\alpha f(t/L^z) \tag{1}
\]

From the \( t \to \infty \) limit discussed in the previous paragraph we have the scaling relation \( 1/2 = \alpha z \). When \( \alpha \) takes its expected value of \( 1/4 \) this implies \( z = 2 \), consistent with a diffusive motion of reactants. We ran simulations (with hardcore exclusion) and found agreement with the scaling form but with \( \alpha \approx 0.29 \). This is consistent with other simulations where a slow approach to \( \alpha = 0.25 \) has been found.

In the following we consider the effects of opposed bulk driving together with exclusion and with boundary injection on the reaction zone behaviour. We shall show \( i \) that phase transitions occur as the reaction rate and injection rate are varied, and \( ii \) in all phases finite size scaling forms \([1]\) hold but have different \( \alpha, z \) to the isotropic diffusion case. In particular two phases are predicted where the reaction zone width varies as \( w \sim \mathcal{O}(L) \) and is thus delocalised.

### 2. Definition of the Model

The model consists of a one-dimensional lattice of \( L \) sites, each of which can be occupied by a single particle, either of type \( A \) or \( B \), or unoccupied (which we denote by 0). Type \( A \)
particles are injected at the left edge of the system and type $B$ particles are injected at the right, both with rate $\delta$. Once in the lattice, the $A$s always hop to the right and the $B$s always hop to the left. If ever an $A$ particle and a $B$ particle are on adjacent sites they react with rate $\lambda$ and are both removed from the lattice. Since particles cannot occupy the same site, we see that once an $A\ B$ pair are adjacent they simply wait a random time, exponentially distributed with mean $\lambda^{-1}$, before annihilating. An inert product of the reaction is then deposited on the sites previously occupied by the particles. However, this product plays no further role in the evolution of the system.

The dynamics can be written in terms of microscopic rules for a bond of two adjacent sites $i, i+1$ in the bulk of the system

\begin{align*}
A_i 0_{i+1} &\to 0_i A_{i+1} \quad \text{and} \quad 0_i B_{i+1} \to B_i 0_{i+1} \quad \text{both with rate 1} \\
A_i B_{i+1} &\to 0_i 0_{i+1} \quad \text{with rate } \lambda
\end{align*}

where these rules are valid for $1 \leq i \leq (L-1)$. At the boundaries we have injection of particles

\begin{align*}
0_1 &\to A_1 \quad \text{and} \quad 0_L \to B_L \quad \text{both with rate } \delta, \text{ at the lhs and rhs respectively.} \quad (3)
\end{align*}

(If either an $A$ or a $B$ particle ever reaches the opposite end of the lattice it is removed with unit rate. This seldom occurs and has a negligible effect on the behaviour of the system.)

The rules thus specified allow one to simulate the model numerically by Monte Carlo techniques. We define $a_i$ and $b_i$ as the binary variables denoting occupation of site $i$ by an $A$ or $B$ particle respectively (i.e. if site $i$ has an $A$ particle on it $a_i = 1$ otherwise $a_i = 0$). We also define $\langle X(t) \rangle$ as the quantity $X$ at time $t$ starting from some fixed initial condition and averaged over the stochastic dynamics up to time $t$. In a Monte Carlo simulation this is conveniently performed by averaging over many systems, each one having been evolved from the same initial state at $t = 0$. Quantities of interest can now be considered such as the particle densities $\langle a_i \rangle, \langle b_i \rangle$.

As the dynamics does not allow for particles to pass through each other, the system is segregated into two domains: a domain comprising $A$s and $0$s and a domain comprising $B$s and $0$s. The reaction front is defined as being at the instantaneous position of the interface of the two domains (strictly only defined when an $A$ and $B$ particle are on adjacent sites). Due to the fluctuating particle currents flowing into the interface, the reaction front will move stochastically through the lattice. The probability of finding the reaction front at bond $i, i+1$ at time $t$ is $\langle a_i(t)b_{i+1}(t) \rangle$ giving the rate of production of the inert product at site $i$, $R_i(t)$, as $\lambda(\langle a_{i-1}(t)b_i(t) \rangle + \langle a_i(t)b_{i+1}(t) \rangle)$. The distribution $R(t)$ will be used to define
the reaction zone, the region where reactions can occur with non-zero rate, which in turn characterises the phase structure of the model.

3. The Phase Diagram

The parameter space of the model is spanned by $\delta$ (the injection rate) and $\lambda$ (the reaction rate). Using Monte Carlo simulation with initial conditions of segregated reactants, we examined the density profiles $\langle a_i \rangle, \langle b_i \rangle$ over a time-scale less than the typical time for large fluctuations of the reaction front to occur (short time average) and in the $t \to \infty$ limit (long time average). The normalised distribution of the inert product (proportional to the production rate) was also measured. Considering $0 \leq \delta \leq 1$ and $0 \leq \lambda < \infty$ we determined three distinct phases, existing in the following approximate regions (see figure 1):

**Phase I.** $0 < \delta < 0.5$ and $\delta << \lambda$. The short-time average shows flat profiles of particle density $\simeq \delta$ away from the reaction zone. The long-time average shows linearly decaying profiles with a broad distribution of inert product. This implies the reaction front is likely to visit all sites of the system: the reaction front is delocalised and the reaction zone fills the system. (The structures near the boundaries are of finite lateral extent and are therefore unimportant in the large $L$ limit).

**Phase II.** $0 < \lambda < 1$ and $\lambda << \delta$. Again, the short-time average shows flat particle profiles away from the reaction zone, though in this phase the density is a function of $\lambda$. As in phase I, the long-time average shows clear evidence for a delocalised reaction front and a reaction zone that fills the system.

**Phase III.** $\delta > 0.5$ and $\lambda > 1$. The short and long time averages show power law decays of the density profiles away from the boundaries and reaction front (i.e. long range structure). The long-time average shows a peaked distribution of inert product near the centre of the system. The reaction front is localised and the width of the reaction zone is much less than the system size.

An approximate analytical form for the phase diagram can be easily derived from a knowledge of the Totally Asymmetric Simple Exclusion Process (TASEP). The TASEP comprises a system of particles moving stochastically in a preferred direction with hardcore exclusion interactions on a lattice of $N$ sites. In the case of open boundary conditions the microscopic rules are: $A_i 0_{i+1} \to 0_i A_{i+1}$ with rate 1 across all sites $i, i+1$ where $i = 1 \to (N-1)$ and at the boundaries there is injection of particles $0_1 \to A_1$ with rate $\delta$ at the lhs and ejection of particles $A_N \to 0_N$ with rate $\beta$ at the rhs. The TASEP has 3 phases: a low density phase (density $\rho = \delta$) for $\delta < 0.5$ and $\beta > \delta$, a high density phase
\( \rho = 1 - \beta \) for \( \beta < 0.5 \) and \( \delta > \beta \) and a maximal current phase \( \rho = 0.5 \) for \( \delta > 0.5 \) and \( \beta > 0.5 \).

A simple approximation to the reaction diffusion model can be made by considering each domain of reactants as a TASEP (hopping to the right for the \( A \) particles and to the left for the \( B \) particles). The underlying assumption is a separation of time-scales i.e. that the domains equilibrate to TASEP profiles faster than the time taken for large fluctuations in the position of the reaction front to occur. Therefore, an effective ejection rate for the \( A \) particles can be defined as \( \beta_{\text{eff}} \approx \lambda \rho_B \) where \( \rho_B \) is the density of \( B \) particles at the reaction front. As we are assuming the reaction front is stationary on the time-scale of the TASEP \( \rho_B \approx \rho_A \). In the TASEP \( \beta \) can be thought of as the density of a reservoir of holes just outside the rhs of the lattice, implying \( \beta_{\text{eff}} \approx (1 - \rho_A) \). Hence, a self-consistent relation can be written for \( \beta_{\text{eff}} \) in terms of \( \lambda \):

\[
\lambda (1 - \beta_{\text{eff}}) = \beta_{\text{eff}}. \tag{4}
\]

With this relation the phases of the TASEP can be reinterpreted in terms of the reaction-diffusion system. Defining \( \rho \) as the (short-time average) density of reactants far away from the reaction front, we find the following phase boundaries and densities within this approximation:

- **Phase I** in the region \( 0 < \delta < 0.5 \) and \( \delta < \frac{\lambda}{1+\lambda}, \rho = \delta \).
- **Phase II** in the region \( 0 < \lambda < 1 \) and \( \frac{\lambda}{1+\lambda} < \delta \), \( \rho = (1 + \lambda)^{-1} \).
- **Phase III** in the region \( \delta > 0.5 \) and \( \lambda > 1 \), \( \rho = 1/2 \).

In phase I, the TASEP approximation for the density of reactants is in excellent agreement with simulation. However, in phase II \( \rho \) is slightly less than \( (1 + \lambda)^{-1} \), the TASEP prediction. It also appears from simulations, that the phase II/phase III phase boundary may be displaced from its predicted position (although the usual difficulties of locating a second order phase transition make it hard to be sure). These observations suggest that the above approximation for \( \beta_{\text{eff}} \) might be improved: the approximation clearly neglects fluctuations in the position of the reaction front as well as correlations in time for the occurrence of reactions.

### 4. Localisation Measurements

A quantity of physical relevance - the total amount of inert product deposited onto a site up to time \( t \), \( \mathcal{R}_i(t) = \int_0^t R_i(t')dt' \), can be used to study the extent to which the reaction front
is localised in each of the phases. Therefore, the time-dependent width of the distribution of deposited inert product, which we take to define the reaction zone, is given by:

$$w^2(t) = \frac{\sum_{k=1}^{L} (k - k_f)^2 R_k(t)}{\sum_{k=1}^{L} R_k(t)}$$

where $k_f = \frac{\sum_{k=1}^{L} k R_k(t)}{\sum_{k=1}^{L} R_k(t)}$ (5)

Steady State Width. For the reaction front to be considered localised, the steady state width, $w_\infty = \lim_{t \to \infty} w(t)$, must scale as $L^\gamma$ with $\gamma < 1$. By Monte Carlo simulation, the exponent $\gamma$ was measured for various $L$ for the three phases, figure 1. The exponents measured were: phase I $\gamma \simeq 1.01 \pm 0.02$, phase II $\gamma \simeq 1.04 \pm 0.05$ and phase III $\gamma \simeq 0.75 \pm 0.01$, suggestive of $\gamma = 1, 1, 3/4$ respectively. In phase I and II the distribution of the inert product remains constant in the limit $L \to \infty$. However, in phase III the fraction of the system containing the inert product vanishes as $\sim L^{-1/4}$ i.e. the reaction front is localised in the middle of the system.

Relaxation Time. In order to investigate the dynamical properties of the model we considered the time taken for the system to relax to steady state behaviour (expected to vary as $\tau \sim L^z$). We measured the exponent $z$ by evolving the system from an initial configuration with the reaction front at a boundary, i.e. the system full of $A$ particles, and examining the $L$ dependence of $\tau$ the time taken for the reaction front to reach the centre of the system, figure 2. Monte Carlo simulations averaged over $\sim 500$ systems gave: phase I $z \simeq 1.93 \pm 0.08$, phase II $z \simeq 2.02 \pm 0.05$ and phase III $z \simeq 2.25 \pm 0.02$, suggestive of $2, 2, 9/4$ respectively. In phase I and II simulations suggest that the reaction front behaves as an unbiased random walker: after waiting a time $\sim L^2$ the reaction front will have explored the whole system. In the TASEP approximation this is expected from the flat profiles and the lack of long-range correlations in these phases. Indeed, in the limit $\lambda \to 0$ (in phase II) the system will be devoid of zeros except when occasional reactions happen and it is easy to show that the dynamics of the front is exactly that of a random walk. In phase III the time taken for the front to reach the centre of the system varies as $\sim L^{9/4}$, i.e. the motion is subdiffusive. However, when the time taken for the front to travel from the centre of the system to a boundary was measured it was found to increase exponentially with $L$. This implies that in phase III the reaction front is stable over short periods of time, but the net motion is biased towards the centre of the system.

Finite Size Scaling Forms. A commonly studied situation in reaction-diffusion systems is the evolution of the system from an initial configuration (at $t = 0$) of segregated reactants. In this case, initially half the lattice ($i = 1 \to L/2$) contains $A$ particles with density $1/2$ and the other half of the lattice ($i = L/2 \to L$) contains $B$ particles, also of density $1/2$. We
expect the growth of $w(t)$ from the initial condition $w(0) = 0$ to follow a finite size scaling form (1). As $w(\infty) = L^\gamma$ we have the scaling relation $\gamma = z\alpha$ giving, in conjunction with our above results, $\alpha = 1/2$ for phases I and II and for the localised phase III $\alpha = 1/3$. The function $w(t)$ was measured for various $L$ for phases I, II and III. The data collapse (figure 3) supports the following scaling forms where $g_{I,II}, \tilde{g}_{I,II}, h, \tilde{h}$ are scaling functions:

For phases I and II
$$w(t) = t^{1/2} g_{I,II} \left( \frac{t}{L^2} \right) \text{ or } w(t) = L g_{I,II} \left( \frac{t}{L^2} \right).$$

(6)

For phase III
$$w(t) = t^{1/3} h \left( \frac{t}{L^{9/4}} \right) \text{ or } w(t) = L^{3/4} \tilde{h} \left( \frac{t}{L^{9/4}} \right).$$

(7)

For phase II and III we believe the data collapses are convincing evidence of the scaling form. However, for phase I larger simulations are really required to confirm the early time growth region ($w \sim t^{1/2}$).

5. Discussion

We have introduced a variant of the $A + B \rightarrow \emptyset$ reaction-diffusion system which shows transitions between phases with delocalised and localised reaction zones as the injection rate $\delta$ and reaction rate $\lambda$ are varied. By treating a single domain of a reactant as a Totally Asymmetric Exclusion Process (TASEP), an approximate relationship (1) for an effective ‘ejection rate’, $\beta_{eff}$, can be found in terms of $\lambda$, allowing an approximate phase diagram to be derived. A particularly interesting transition is that from phase I (delocalised reaction zone) to phase III (localised reaction zone), which occurs (for $\delta < 1/2$) by simply increasing the reaction rate.

In each of the phases the exponents $\{\alpha, z, \gamma\}$ (early time growth of the reaction zone $w \sim t^\alpha$; relaxation time $t \sim L^z$; reaction zone saturation width $w \sim L^\gamma$) obey the finite size scaling relation $\alpha z = \gamma$. In each case, $\{1/2, 2, 1\}$ for the delocalised phases and $\{1/3, 9/4, 3/4\}$ for the localised phase, these exponents are distinct from those of the model discussed in the introduction where the reactants have no bias ($\{1/4, 2, 1/2\}$) suggesting a different universality class for the present model. We believe that the new exponents are related to the behaviour of current fluctuations in the TASEP [18] which in turn are related to KPZ exponents, although we could not find a clean argument.

It would be interesting to examine the robustness of the model to (i) relaxation of the bias to allow for a small backward hopping rate of reactants, and (ii) relaxation of the hardcore exclusion between the different species. If the TASEP approximation is valid the effect of relaxing the bias should preserve the phase structure of the model: partially asymmetric exclusion and asymmetric exclusion belong to the same universality class. However, the relaxation of hardcore exclusion between $A$ and $B$ reactants may strongly affect phase II where the reaction rate is low.
Lastly it would be illuminating to make precise the relation of our TASEP approximation of the reaction diffusion system to a conventional mean field theory which would be expected to hold in dimension $d \geq 2$. One could then explore the feasibility of the RG approach which has been successful in the case where the reactants diffuse isotropically \[13\]

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Figure Captions

Figure 1.
Short and long time averages for each phase from Monte-Carlo simulation. The filled squares plot the density of $A$; the empty squares plot the density of $B$; the line plots the distribution of inert product, normalised so that maximum value is $1/2$. Long time averages were carried out over $10^8$ MCS. The short time averages were over 300 MCS for phase I and 500 MCS for phases II and III all averaged over 5000 systems. The values of $(\delta, \lambda)$ are Phase I $(0.2, 9)$; Phase II $(0.7, 0.3)$; Phase III $(0.9, 10)$.

Figure 2.
(a) Log-log plot of the relaxation time $\tau$ (see main text) versus system size $L$. The measured gradients predict $\tau \sim L^z$ where $z = 1.93 \pm 0.08$ in phase I; $z = 2.02 \pm 0.05$ in phase II; $z = 2.25 \pm 0.02$ in phase III.
(b) Log-log plot of the reaction zone width $w$ defined in (5) versus $L$. The measured gradients predict $w \sim L^\gamma$ where $\gamma = 1.01 \pm 0.02$ in phase I; $\gamma = 1.04 \pm 0.05$ in phase II; $\gamma = 0.75 \pm 0.01$ in phase III.
In both (a) and (b) the values of $(\delta, \lambda)$ are phase I $(0.2, \infty)$; phase II $(1, 0.2)$; phase III $(1, \infty)$.

Figure 3.
Finite size scaling collapses for various values of $L$ between 200 and 1000. $w/L^\gamma$ versus $t/L^z$ with
(a) $\gamma = 3/4$, $z = 9/4$ (see Eq. [7]) for phase III
(b) $\gamma = 1$, $z = 2$ (see Eq. [8]) for phase I (empty symbols) and phase II (filled symbols).
Figure 1:
(a) Measurement of $z$ exponent.

(b) Measurement of $\gamma$ exponent.

Figure 2:
(a) Phase III finite size scaling collapse.  (b) Phase I and II finite size scaling collapse

Figure 3: