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Exact Solution of Two-Species Ballistic Annihilation with General Pair-Reaction Probability

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The reaction process $A + B \to \varnothing$ is modeled for ballistic reactants on an infinite line with particle velocities $v_A = c$ and $v_B = -c$ and initially segregated conditions, i.e., all A particles to the left and all B particles to the right of the origin. Previous models of ballistic annihilation have particles that always react on contact, i.e., pair-reaction probability $p = 1$. The evolutions of such systems are wholly determined by the initial distributions of particles and therefore do not have a stochastic dynamics. However, in this paper the generalization is made to $p \leqslant 1$, allowing particles to pass through each other without necessarily reacting. In this way, the A and B particle domains overlap to form a fluctuating, finite-sized reaction zone where the product $\varnothing$ is created. Fluctuations are also included in the currents of A and B particles entering the overlap region, thereby inducing a stochastic motion of the reaction zone as a whole. These two types of fluctuations, in the reactions and particle currents, are characterised by the *intrinsic reaction rate*, seen in a single system, and the *extrinsic reaction rate*, seen in an average over many systems. The intrinsic and extrinsic behaviors are examined and compared to the case of isotropically diffusing reactants.

KEY WORDS: Ballistic annihilation; reaction process; reaction zone; non-equilibrium statistical mechanics; exactly solved model.

I. INTRODUCTION

A detailed understanding of reaction systems is an essential ingredient for the study of a broad range of problems.[1] In systems of many interacting reactants, it often happens that knowledge of the precise physical mechanism whereby reactions occur is irrelevant in determining the macroscopic behaviour. The most important factors are the number of different reacting species that combine in a single reaction, the type of motion that

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each of these species performs and the initial/boundary conditions in the vessel containing the reactants. Thus, simple theoretical models that mimic these elements can be used to study real systems as diverse as exciton dynamics in polymer chains,[2] monopole annihilation in the early universe[3] as well as the more conventional chemical processes.[4] However, even such highly simplified models can prove difficult to analyse. This is especially true if the reactions are not fully reversible as the methods of equilibrium statistical mechanics cannot be used, due to the lack of detailed balance.

Many different, idealised reaction systems have been studied analytically over the last few decades[1–22] usually for the case of fully irreversible reactions. These include same species ($nA \to \varnothing$) and multi-species ($\sum_k n_k A_k \to \varnothing$) processes, where n_k of each distinct A_k species combine to form a single inert product $\varnothing$. The multi-species reaction systems exhibit particularly rich behaviour because reactions can only occur at places where all the necessary constituents are present—so called *reaction zones*. Often, due to the initial/boundary conditions or spontaneous symmetry breaking[13, 14] each species of reactant is largely confined to its own domain. The places where these domains overlap usually takes up only a small part of the whole system, often causing the net reaction rate to differ drastically from that predicted by the mean-field-like rate equation. Such behaviour can give rise to complicated structures, and is of particular interest in the context of pattern formation and growth determination in organisms.[23] The physics of multi-species reaction processes is, therefore, largely determined by activity in and around the reaction zones. The important factors being the way reactants flow from their respective domains into the overlap region as well as the behaviour of the reaction mix inside the reaction zone.

In this paper a model of the two-species system $A + B \to \varnothing$ is introduced for the case of ballistically moving reactants and a pair-reaction probability less than one. The model is then solved exactly, allowing an analytic study of the dynamic reaction zone formed at the overlap of the A and B domains. However, before describing the model in detail two important statistical quantities associated with the reaction zone are defined.

A. The Intrinsic and Extrinsic Reaction Rates

A comprehensive analysis in the RG framework for the two-species $A + B \to \varnothing$ reaction system with *diffusive* reactants[12] identified two distinct sources of fluctuations affecting the behaviour of the reaction zone,

first inside the reaction zone itself and second in the currents of particles entering the reaction zone.

The shape and size of the reaction zone is determined by the typical lifetimes and motions of reactants in the overlap region. These lifetimes in turn depend on the fluctuating local densities of reactants, as well as the reaction probability for an interacting pair of A and B particles. As the density fluctuations will be caused (in part) by reactions that have already occurred, the reaction rate can become highly correlated both spatially and temporally. This behaviour in the overlap region leads to the following definition for the *intrinsic reaction rate.*

• The *intrinsic reaction rate* $\mathscr{R}_I(x_r, t)$ measured at a distance x_r from the centre of the overlap region at time t, is the typical spatial reaction rate, i.e., production rate of $\varnothing$ particles per unit time per unit space, seen in a single realization of the system's evolution. The intrinsic reaction rate characterizes the *intrinsic reaction zone*—the instantaneous reaction zone formed where the A and B domains overlap.

A second source of fluctuations can come from the A and B currents that flow into this (intrinsic) reaction zone. Even if these currents are on average equal in magnitude, any fluctuations about the average will contribute a noisy component to the reaction zone's motion, i.e., it will move stochastically about its expected position. Hence, given some initial conditions only a probabilistic statement can be made about the reaction rate at some later place and time (x, t). This suggests the definition of a second quantity, the *extrinsic reaction rate.*

• The *extrinsic reaction rate* $\mathscr{R}_E(x, t)$ is defined as the probability density for reactions to occur at a time t and position x given some initial distribution function for the particles' positions. Therefore, $\mathscr{R}_E$ is the expected reaction rate found by averaging over all allowed realizations of the system's evolution.

Models with ballistically moving reactants have been extensively studied in the context of the reaction kinetics in an ideal gas, where the mean free path of the reacting particles is similar to their separation[6–8] In particular the two-species case, with particles A and B having velocities $v_A = +c$ and $v_B = -c$ respectively, has been studied with *homogeneous* initial conditions, i.e., each species initially randomly placed throughout the infinite line. More recently, the initial conditions of *segregated* particles was studied[9, 10] with the As initially to the left and the Bs to the right of the origin, again with reactants that always annihilate on contact. The initial positions for each species of particle (confined to their own domains on either side of the origin) were chosen to be random, thereby introducing

fluctuations into the particle currents flowing into the reaction zone. By averaging over all initial distributions the form of the extrinsic reaction rate was derived and shown to be Gaussian. However, because there can be no reaction fluctuations in this model (the particles always annihilate on contact) the A and B domains never overlap and the instantaneous reaction zone has effective width zero. Also, as the reactants move ballistically the system does not have a genuinely stochastic evolution, i.e., once the initial conditions (the particle positions) are fixed so is the system's future.

This model has a generalisation to the case of arbitrary reaction probability; this is the general model introduced below. The fluctuations in the particle streams can be retained and combined with the fluctuations in the (now) finite-sized overlap region. This then allows the intrinsic and extrinsic behaviour of the $A + B \rightarrow \varnothing$ reaction system to be studied analytically.

B. The Definition of the Model

The model consists of a one-dimensional continuous space in which two species of reactants, A and B particles, move with fixed velocities $v_A = +c$ and $v_B = -c$, see Fig. 1. Initially, the reactants are separated, i.e., at time $t = 0$ A particles are distributed in the interval $[-\infty : 0]$ at positions $(y_1, y_2 \cdots)$ and the B particles are in the interval $[0 : \infty]$ at positions

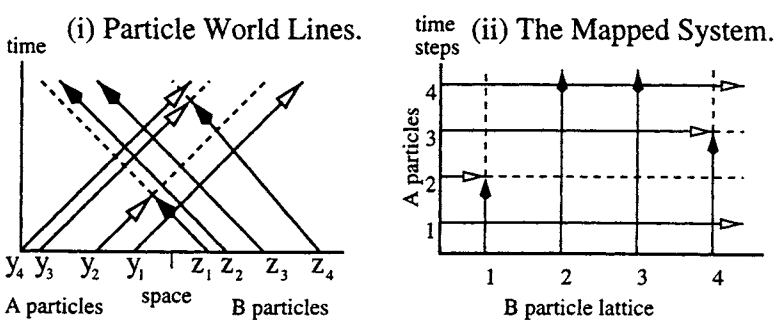

Fig. 1. (i) A realisation of the model described in Section 1. The solid lines are paths of particles in space and time, with the dotted lines the trajectories annihilated particles would have taken. The A and B particles start at positions y_m and z_n respectively and move with fixed velocities $v_A = c$ and $v_B = -c$. An intersecting pair of particles either annihilates (probability p) or continues unaffected (probability $q = 1 - p$). (ii) The simplified, mapped system described in Section 2. At unit time steps A particles are "shot" through the B lattice, passing through each occupied site (with the same microscopic reaction probabilities as above). A reaction occurring at a site produces a vacancy, and it is the statistics of these vacancies that is used to derive $P(m, n)$, the pair-reaction probability.

$(z_1, z_2 \cdots)$. The subscripts on y_m and z_n refer to the relative *initial* order of the particles counted from the origin. Because of the ballistic motion of the reactants, the trajectories of the particles retain their initial ordering for all time.

Two different distributions for $\{y\}$ and $\{z\}$ will be considered, first equally-spaced reactants (Section 3) and second, random positions of the reactants (Section 4). For both cases the average density is chosen to be ϱ, leading to average particle currents of $\pm c\varrho$. However, the initial conditions studied in Section 4 will be shown to introduce Gaussian fluctuations about these average values.

The position x and time t that the mth A and nth B particles' trajectories intersect are

$$x=(y_m+z_n)/2 \qquad t=(z_n-y_m)/2c \tag{1}$$

When such an (m, n) pair of reactants' trajectories meet, there are three distinct events that can occur.

- If both reactants still occupy their trajectories a reaction can occur with probability p. If a reaction occurs both particles are removed from the system and a $\varnothing$ product particle is considered to have been deposited at the point of annihilation. The $\varnothing$ merely serves as a marker and plays no further role in the evolution of the system.

- If both reactants are still travelling along their trajectories then with probability $q=(1-p)$ no reaction occurs and the particles continue unaffected.

- If one of the particles has previously been annihilated, i.e., one trajectory is unoccupied, the other particle continues unaffected with probability 1. (Of course, if neither particle is present no change occurs when the empty trajectories cross.)

Thus, two distinct sources of fluctuations are included in this model, in the reactions (if $p<1$) and in the currents of particles flowing into the reaction zone (if $\{y\}$ and $\{z\}$ have random elements). Hence, the system models a finite-sized stochastically moving reaction zone, with measurable intrinsic and extrinsic behaviour. However, it is clear that this model represents a special case, in that the two sources of noise are uncoupled. The fluctuations in the trajectories are quenched at $t=0$, and the probability that an (m, n) pair annihilates depends only on the total number of trajectories crossed by each particle, and not the trajectories' exact positions. This uncoupling of fluctuations allows the probability density

$\mathscr{D}$ for an (m, n) pair to react at (x, t) to be written as the product of two independent distributions

$$\mathscr{D}(m, n, x, t) = P(m, n)\, \mathscr{G}_{mn}(x, t)$$

where $P(m, n)$ is the probability for an (m, n) pair to mutually annihilate and $\mathscr{G}_{mn}(x, t)$ is the probability density for the trajectory intersections (1). The calculation of the total annihilation probability can therefore be decomposed into, (i) a counting problem for the integer variables (m, n), and (ii) the derivation of the distribution functions for the continuous random numbers $\{y\}$ and $\{z\}$.

The remainder of the paper is structured as follows. In Section 2 the probability of pair annihilation $P(m, n)$ is derived (12) by mapping the model onto a simple system of target particles on a one-dimensional lattice. The behaviour of the lattice system is then briefly examined in the context of radiation damage of crystals. In Section 3 the form of the (intrinsic) reaction rate for the simple case of equally-spaced reactants is studied. In particular, the steady-state reaction-rate (16) and particle densities (17), and the time-dependence of the reaction rate (19) are derived. Finally, in Section 4 fluctuations in the initial particle positions are treated. The distribution function $\mathscr{G}_{mn}(x, t)$ is found and used to derive the forms of the intrinsic and extrinsic reaction rates, (24) and (25) respectively. The appendix shows how the calculation in Section 2 may be translated into the second-quantisation formalism and relates the mapped system to the algebra $SU_q(2)$.

II. THE PAIR-REACTION PROBABILITY

The main result of this section is the derivation of the (m, n) pair annihilation probability $P(m, n)$, Eq. (12). In Section 1, it was noted that this quantity is independent of the initial positions of the particles, depending only on their relative order. Making use of this fact, a simpler system can be treated, that still preserves the order that the A and B particles pass through each other.

Consider now a one-dimensional, semi-infinite lattice (with sites $n = 1, 2 \cdots$) with a B particle initially occupying each site. At discrete time steps ($m = 1, 2 \cdots$) the mth A particle is "shot" through the B array, passing sequentially through each site until its eventual annihilation, see Fig. 1. An A particle either moves through a site with a B present with probability q, or reacts with a B at that site with probability p. Once an A and B pair have reacted, the site that the B occupied becomes vacant and any subsequent A that passes through the vacant site does so with

probability 1. This whole process, of a single A passing through the lattice and eventually reacting, is considered to happen in a negligible amount of time. This mapped system is merely a deformation of the coordinates used in the original model, so both systems share the same pair annihilation probability $P(m, n)$.

It is useful to consider the statistics of the positions of the m vacancies that exist in the lattice immediately after the mth time step. The positions of these vacancies are labelled $(n_1 \cdots n_m)$, where it is important to note that the subscript used refers to the relative positions of the sites, i.e., $n_1 < n_2 < \cdots < n_m$, and not which A particle caused the vacancy at that position.

The rest of this section is devoted to the calculation of $P(m, n)$, the probability that the mth A reacts at site n. First, the probability of a particular distribution $\Psi_m(n_1 \cdots n_m)$ for the positions of the m vacancies is derived, Eq. (3). These probabilities can be used as a convenient basis, in the sense that all other probabilistic quantities may be expressed as linear combinations of the $\{\Psi_m\}$. This basis is then used to find the expected vacancy density $V_m(n)$ at site n, after m A particles have passed, Eq. (9). As an aside, a parallel is drawn between this simplified model and a crystal that has been damaged by radiation. In particular, it is shown that the damaged region described by $V_m(n)$ propagates like a soliton through the B array, Eq. (10). Finally, the discrete gradient of $V_m(n)$ is then used to calculate the required quantity $P(m, n)$, Eq. (12). The method described below translates into the second-quantisation formalism and shows the system to be described by the algebra $SU_q(2)$. The techniques used in this formalism are briefly reviewed in the appendix.

A. The Basis for the mth Time Step Ψ_m

The probability $\Psi_m(n_1 \cdots n_m)$ that after m A particles have passed through the B lattice vacancies exist at sites $n_1 < n_2 < \cdots < n_m$ is now derived. Consider first the simplest case $m = 1$, i.e., just after the first time step. The probability that the A particle has annihilated with a B on site n_1 producing a vacancy there is

$$\Psi_1(n_1) = pq^{n_1 - 1} = (q^{-1} - 1)\, q^{n_1}$$

where p is the probability a single reaction could occur, and $q = (1 - p)$.

Now consider the state of the system after the second time step, i.e., after a total of two A particles have passed through the B lattice. Vacancies now exist at sites n_1 and n_2 (where relabelling may be necessary to ensure that $n_1 < n_2$). There are two histories that contribute to this configuration,

each with different probabilities. Either a vacancy first appeared at site n_2 and then the second one at site n_1, or the first vacancy at site n_1 and the second at site n_2. The probability of the first history is just the product $\Psi_1(n_1)\,\Psi_1(n_2)$ as the second A particle does not pass through the vacancy produced by the first A particle. However, in the second history described, the second A particle does pass through the vacancy at n_1, increasing its chance of reacting with any site $n > n_1$ by a factor of q^{-1}. Therefore,

$$\begin{aligned}
\Psi_2(n_1, n_2) &= \Psi_1(n_1)\,\Psi_1(n_2) + q^{-1}\Psi_1(n_1)\,\Psi_1(n_2) \\
\Psi_2(n_1, n_2) &= (1 + q^{-1})\,\Psi_1(n_1)\,\Psi_1(n_2) \qquad (2) \\
\Psi_2(n_1, n_2) &= (q^{-2} - 1)(q^{-1} - 1)\,q^{n_1 + n_2}
\end{aligned}$$

It is simple to generalise to the m-vacancy basis $\Psi_m(n_1 \cdots n_m)$. The states at the $(m-1)$th time step that can contribute to an mth time step configuration are those with vacancies at $(n_2 \cdots n_m)$, $(n_1, n_3 \cdots n_m)$, ..., $(n_1 \cdots n_{m-1})$. In each of these cases, to produce the final state $(n_1 \cdots n_m)$ the mth A particle must react with sites $n_1, n_2 \cdots n_m$ respectively. Therefore, taking account of how many vacancies the mth A particle must pass through in each case, a relation between the bases that describe the system after time steps $(m-1)$ and m can be written

$$\begin{aligned}
\Psi_m(n_1 \cdots n_m) = {} & \Psi_1(n_1)\,\Psi_{m-1}(n_2 \cdots n_m) \\
& + q^{-1}\Psi_1(n_2)\,\Psi_{m-1}(n_1, n_3 \cdots n_m) + \cdots \\
& + q^{-(m-1)}\Psi_1(n_m)\,\Psi_{m-1}(n_1, n_2 \cdots n_{m-1})
\end{aligned}$$

Assuming that it is possible to write $\Psi_{m-1} = C_{m-1} \prod_{j=1}^{m-1} \Psi_1(n_j)$ and using the result (2), the form for general m follows by induction

$$\Psi_m(n_1 \cdots n_m) = (1 + q^{-1} + \cdots q^{-(m-1)})\, C_{m-1} \prod_{j=1}^{m} \Psi(n_j) = C_m \prod_{j=1}^{m} \Psi(n_j) \qquad (3)$$

$$\Psi_m(n_1 \cdots n_m) = \prod_{j=1}^{m} \left[\frac{q^{-j} - 1}{q^{-1} - 1}\, \Psi_1(n_j) \right] = \prod_{j=1}^{m} \left[(q^{-j} - 1)\, q^{n_j} \right]$$

Hence, the distribution of the m vacancies is given by a product of exponentials in the site labels, with care being taken to preserve the order $n_1 < n_2 < \cdots < n_m$ in any sums that they appear.

B. The Vacancy Density $V_m(n)$

The probability that a vacancy is found at site n after m A particles have passed through the lattice, $V_m(n)$, is now derived. This density is given by the sum of all the Ψ_m that include a vacancy at the site n,

$$V_m(n) = \sum_{k=1}^{m} \tilde{\Psi}_m(n_1 \cdots, n_k = n, \cdots n_m)$$

where the notation $\tilde{\Psi}$ is used to denote an internal sum over all the unfixed variables, i.e., any n_j with $j \neq k$, that respects the order $n_1 < n_2 < \cdots < n_m$. Hence,

$$\tilde{\Psi}_m(n_1 \cdots, n_k = n, \cdots n_m) = \sum_{n_1=1}^{n-(k-1)} \cdots \sum_{n_{k-1}=n_{k-2}+1}^{n-1} \sum_{n_{k+1}=n+1}^{\infty} \cdots \sum_{n_m=n_{m-1}+1}^{\infty} \Psi_m(n_1 \cdots, n_k = n, \cdots n_m) \tag{4}$$

For the case $m = 1$, the vacancy density is simply $V_1(n) = \Psi_1(n)$. However, after the second A particle passes through the B array a sum must be made over the unfixed variables

$$V_2(n) = \tilde{\Psi}_2(n_1 = n, n_2) + \tilde{\Psi}_2(n_1, n_2 = n)$$

$$V_2(n) = \sum_{n_2=n+1}^{\infty} \Psi_2(n_1 = n, n_2) + \sum_{n_1=1}^{n-1} \Psi_2(n_1, n_2 = n)$$

$$V_2(n) = (1 + q^{-1})\, \Psi_1(n) \left(\sum_{j=n+1}^{\infty} \Psi_1(j) + \sum_{j=1}^{n-1} \Psi_1(j) \right)$$

$$V_2(n) = q^n(q^{-2} - 1) \left(\sum_{j=1}^{\infty} \Psi_1(j) - \Psi_1(n) \right)$$

$$V_2(n) = q^n(q^{-2} - 1)(1 - V_1(n))$$

The density for $m = 2$ is therefore related to the $m = 1$ case, by making use of the product form of Ψ_2. It is possible to generalise this result and obtain a recursion relation. The following two results will prove useful. First the product form of Ψ_m is used to relate Ψ_m to Ψ_{m-1}

$$\Psi_m(n_1 \cdots, n_k = n, \cdots n_m) \equiv q^n(q^{-m} - 1)\, \Psi_{m-1}(n_1 \cdots n_{k-1}, n_{k+1} \cdots n_m) \tag{5}$$

Second, a slightly less trivial result, that nevertheless has a simple interpretation

$$\sum_{k=1}^{m} \tilde{\Psi}_{m-1}(n_1 \cdots n_{k-1}, n_{k+1} \cdots n_m)$$

$$\equiv 1 - \sum_{k=1}^{m-1} \tilde{\Psi}_{m-1}(n_1 \cdots, n_k = n, \cdots n_{m-1}) \equiv 1 - V_{m-1}(n) \qquad (6)$$

The LHS of this equation is the sum of all the Ψ_{m-1} that do not include a vacancy on site n. This is simply the sum of all possible Ψ_{m-1} ($\equiv 1$ by normalisation) less those that include the site n; the statement on the RHS of (6). The $\tilde{\Psi}$s in this equation are identical to those in Eq. (4) in as much as they involve an ordered sum over all unfixed variables. However, it should be noted that, though on the LHS the term $n_k = n$ has been factored out by using (5), the order restriction still holds, i.e., $n_{k-1} < n$ and $n_{k+1} > n$.

Both these results generalise the method used already to find $V_2(n)$. Therefore, for the case of m vacancies

$$V_m(n) = \sum_{k=1}^{m} \tilde{\Psi}_m(n_1 \cdots, n_k = n, \cdots n_m)$$

$$V_m(n) = q^n(q^{-m} - 1) \sum_{k=1}^{m} \tilde{\Psi}_{m-1}(n_1 \cdots n_{k-1}, n_{k+1} \cdots n_m) \qquad (7)$$

$$V_m(n) = q^n(q^{-m} - 1)(1 - V_{m-1}(n)) \qquad (8)$$

The recursion relation (8) can be solved, with the initial condition $V_1(n) = \Psi_1(n)$, to give the vacancy density at site n after mA particles have passed through the B array

$$V_m(n) = -\sum_{j=1}^{m} \left[\prod_{k=j}^{m} (q^n - q^{n-k}) \right]$$

$$V_m(n) = -q^{(n-m)} \sum_{j=0}^{m-1} \left[q^{(n-m)j} \prod_{k=0}^{j} (q^m - q^k) \right] \qquad (9)$$

Interestingly, in the limit of large m (in particular $q^m \ll p$) the vacancy profile depends purely on the difference $s = (n - m)$. Interpreting the system as a crystal (the B array) that is being damaged by incoming radiation

(the A particles), this implies that the interface between the damaged and undamaged regions reaches a steady-state moving profile

$$\lim_{m\to\infty} V_m(n) = V(s) = q^s \sum_{j=0}^{\infty} (-q^s)^j q^{j(j+1)/2} \tag{10}$$

$$\sim \tfrac{1}{2}(1-\tanh(ps/2)) + O(p) \tag{11}$$

where the second equation becomes valid in the case of low reaction probability $p \ll 1$ and is analytically continued for $s < 0$. Therefore, the damaged region propagates like a soliton through the B array.

C. The Pair-Reaction Probability $P(m, n)$

The difference between the vacancy density at site n just after the mth and $(m-1)$th time steps is the probability that at the mth time step an annihilation occurs at site n. This is the required pair reaction probability $P(m, n)$.

$$\begin{aligned} &P(m,n) = V_m(n) - V_{m-1}(n) \\ &P(m,n) = q^{(n-m)} \sum_{j=0}^{m-1} \left[q^{(n-m)j}(1-q^{j+1}) \sum_{k=1}^{j} (q^m - q^k) \right] \end{aligned} \tag{12}$$

which is the main and final result of this section. For the case studied in ref. 9, i.e., $q = 0$, this result reduces to a delta function as expected, i.e., $P(m, n) = \delta(m, n)$. In the next section $P(m, n)$ is used to study the original model described in Section 1, with equal spacing between neighbouring reactants at $t = 0$.

III. THE REACTION ZONE WITHOUT CURRENT FLUCTUATIONS

Reverting back to the original model described in the first section, $P(m, n)$ is reinterpreted as the probability that an (m, n) pair of particles mutually annihilates in a system of ballistically moving reactants. In this section, the model is studied with the initial conditions $y_m = -m\varrho^{-1}$ and $z_n = n\varrho^{-1}$, i.e., with no fluctuations in the particle currents. The point of intersection of an (m, n) pair of trajectories is therefore

$$x = x_r = (n-m)/2\varrho \qquad t = (m+n)/2c\varrho \tag{13}$$

i.e., reactions occur only at discrete positions and times. The lack of fluctuations in the currents negates the need to discuss the extrinsic behaviour,

as the intrinsic reaction zone will not wander stochastically. In fact as $\mathscr{R}_I = \mathscr{R}_E$ for these initial conditions, the zero current-fluctuation reaction zone will be denoted by $\mathscr{R}_0$ to avoid confusion with Section 4. The steady currents also mean that the centre of the overlap region is always at the origin, and therefore $x_r = x$ in this case. As will be shown in the next section, even when current fluctuations are taken into account, the form of the intrinsic reaction rate remains the same as for this case, as long as x_r is then measured from the centre of the stochastically moving overlap centre seen in Section 4.

In the rest of this section several aspects of Eq. (12) are studied. First, the steady-state reaction rate (16) and particle densities are derived (17). Second, the dynamics of $P(m, n)$ in the $O(p)$ approximation is studied and the system is shown to decay to the steady state in finite time (19). Finally, the correlations in the reaction zone are briefly discussed, with reference to the mean-field and $O(p)$ approximations.

A. The Steady-State Limit and $O(p)$ Time Dependence

After a sufficient length of time, characterized by $q^m \ll p$, Eq. (12) becomes a function of $(n-m) = s$ only. This is in much the same way as for Eq. (10) and can also be interpreted as a late-time or steady-state limit. Therefore, for large m (or n by symmetry) any $(m+l, n+l)$ pair will have the same probability of reacting, regardless of the value l takes,

$$\lim_{m \to \infty} P(m, n) = P(s) = q^s \sum_{j=0}^{\infty} [(-q^s)^j q^{j(j+1)/2}(1-q^{j+1})] \tag{14}$$

$$P(s) = \frac{p}{4\cosh^2(ps/2)} + O(p^2) \tag{15}$$

The production rate of $\varnothing$ particles at a position $x_r = s/2\varrho$ can now be found, namely $c\varrho P(2x_r\varrho)$. However, the nature of the initial conditions means that reactions can occur only at discrete positions ($s = 0, \pm 1 \cdots$). So, to derive the reaction rate $\mathscr{R}_0(x_r)$ which is the production rate *per unit space*, a coarse-graining is made over a length scale ϱ^{-1}

$$\mathscr{R}_0(x_r) = 2c\varrho^2 P(2x_r\varrho) \tag{16}$$

This reaction rate (characterizing the intrinsic reaction zone) is shown graphically in Fig. 2 for $p = q = 1/2$ with comparison made between the $O(p)$ and mean-field approximations (to be described below). The particle densities can also be found, given that $[1 - V(s)]$ is the probability a B

particle still exists just after a reaction occurs at site s (and similarly for the A particles). Therefore, the density profiles $\varrho_A(x_r)$ and $\varrho_B(x_r)$ in the steady-state are

$$\varrho_A(x_r) = \varrho[1 - V(-2\varrho x_r)] \qquad \text{and} \qquad \varrho_B(x_r) = \varrho[1 - V(2\varrho x_r)] \quad (17)$$

These profiles are also shown, for the same parameters $q = p = 1/2$, in Fig. 2, with comparison made to the $O(p)$ approximation.

To examine the passage to the steady state, it is illuminating to derive the time dependence of (12) to first order in p, i.e., take the limit $p \ll 1$ in Eq. (12) but still keep m finite.

$$P(m, n) = p(2\cosh(p(n-m)/2) - \exp^{-p(m+n)/2})^{-2} + O(p^2) \quad (18)$$

$$\mathscr{R}_0(x_r, t) = \frac{pc\varrho^2\theta(c\varrho t - 1 - \varrho|x_r|)}{2(\cosh(p\varrho x_r) - \exp^{-pc\varrho t})^2} + O(p^2) \quad (19)$$

where $\theta(u) = 1$ or 0 for $u \geqslant 0$ or $u < 0$, respectively. It is clear that the system reaches a steady state exponentially quickly (a characteristic of systems with short-range correlations) with a decay time $(pc\varrho)^{-1}$. To leading order in t, this exponential decay time will also be a feature of the model with randomly distributed particles to be examined in Section 4.

B. Correlations, the $O(p)$ and Mean-Field Approximations

The first order in p approximations (11), (15), (18) and (19) valid for $p \ll 1$ represent the case where each particle passes through very many of its opposite kind before finally annihilating. In this $p \ll 1$ limit it could therefore be expected that a *mean-field* approach becomes exact. For the reaction model the mean-field approximation is one that neglects density correlations in the reaction zone. It assumes that the probability of a reaction occurring depends on the product of the probabilities that an A and B particle are present, i.e., that the A and B densities are independent statistical quantities. Therefore, given that $[1 - V_{m-1}(n)]$ is the probability that the nth B has not annihilated before meeting the mth A particle (and similarly for the mth A particle by symmetry), the mean-field approximation is

$$\begin{aligned} P_{mf}(m, n) &= p[1 - V_{n-1}(m)][1 - V_{m-1}(n)] \\ P_{mf}(s) &= p[1 - V(s-1)][1 - V(s+1)] \end{aligned} \quad (20)$$

where the second equation is the steady-state limit. From these equations it can be seen by substitution that the $O(p)$ approximations (valid only in

the limit $p \to 0$) are indeed mean-field like. Though, this is not to say that the $O(p)$ and mean-field approximations are equivalent outside the small p limit, as can been seen in Fig. 2 where comparison can be made for $p = 0.5$.

For the $p \ll 1$ case, the reaction zone will be much larger than the interparticle spacing, and any structure will slowly vary with the variables m or n. Therefore, the $O(p)$ approximations can also be captured from the *continuum* mean-field equations

$$\frac{\partial \varrho_A}{\partial t} = -c\frac{\partial \varrho_A}{\partial x} - r\varrho_A\varrho_B \qquad \frac{\partial \varrho_B}{\partial t} = +c\frac{\partial \varrho_B}{\partial x} - r\varrho_A\varrho_B$$

where r is the reaction parameter. However, the full results (9), (10), (12) and (14) cannot be obtained by such a mean-field approach, as can be seen in Fig. 2 where $P_{mf}(s)$ and $P(s)$ are compared. The figure shows that the correlations are strongest in the centre of the reaction zone and decay quickly towards the edge of the overlap region.

Interestingly, a similar cross-over to mean-field behaviour is also seen in the case of diffusive reactants[12] in the limit of high diffusivity and low reaction rate. The interpretation, of many interactions between particles before final annihilation occurs, is the same.

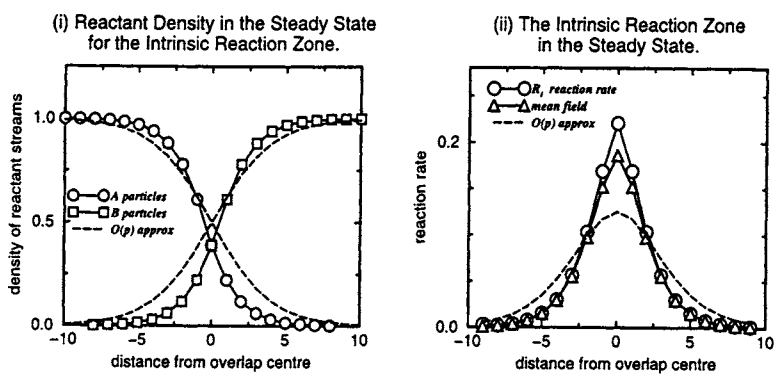

Fig. 2. (i) The steady-state density profiles of the A and B reactants at $x_r = s/\varrho$ for the case $p = q = 1/2$. The densities given in Eq. (17) involve the vacancy densities (10) for the exact case (circles and squares) and (11) for the $O(p)$ approximation (dashed lines). (ii)The steady-state reaction rate at position $x_r = s/\varrho$, again for $p = q = 1/2$. The the exact result (circles) is given in Eq. (16) with x_r measured from the origin, or in Eq. (24) with x_r measured from the centre of the fluctuating overlap region. Also plotted are the mean-field (triangles) and the $O(p)$ approximations (dashes) given in Eqs. (20) and (19) respectively. Comparison between the exact and mean-field results show the reaction rate to be most correlated at the centre of the overlap region.

IV. REACTION ZONES

In this section the case of fluctuations in the initial conditions is treated (though still with each species initially segregated to either side of the origin). This allows the intrinsic and extrinsic reaction rates for the system with fluctuations in both reactions and particle currents to be found, Eqs. (24) and (25).

The initial positions of each species of particle are chosen to be uncorrelated with an average spacing ϱ^{-1} and are described by Poissonian distributions with the variances of the interparticle spacing being ϱ^{-2}. Hence, both y_m and z_n, the initial positions of the mth A and nth B particles, can be written as sums of independent random numbers

$$y_m = (y_m - y_{m-1}) + (y_{m-1} - y_{m-2}) + \cdots + (y_2 - y_1) + (y_1 - 0)$$
$$z_n = (z_n - z_{n-1}) + (z_{n-1} - z_{n-2}) + \cdots + (z_2 - z_1) + (z_1 - 0)$$

The central limit theorem can now be used, stating that for large m and n such sums of many independent random numbers, drawn from the same distributions, become the Gaussians

$$\mathscr{Y}(y_m) \propto \exp\left(-\frac{(\varrho y_m + m)^2}{2m}\right) \qquad \mathscr{Z}(z_n) \propto \exp\left(-\frac{(\varrho z_n - n)^2}{2n}\right) \tag{21}$$

The probability density for the mth A and nth B trajectories to intersect at position x and time t (the function $\mathscr{G}_{mn}(x, t)$ defined in Section 1) is now easily calculated. As both x and $2ct$ are linear combinations of independent Gaussian variables, for $s \ll m$ their distribution functions are also Gaussians with means and variances linear combinations of those in Eq. (21)

$$\mathscr{G}_{mn}(x, t) = \frac{2c\varrho^2}{\pi(n+m)} \exp\left(-\left[\frac{(2\varrho x - (n-m))^2}{2(m+n)} + \frac{(2c\varrho t - (m+n))^2}{2(m+n)}\right]\right) \tag{22}$$

Before calculating the intrinsic and extrinsic reaction rates for $p < 1$, the deterministic case $p = 1$ examined in ref. 9 is first reviewed. Because in this case, the reaction probability $P(m, n) = \delta_{mn}$ the two domains of A and B particles never overlap and only an extrinsic reaction rate can be meaningfully defined. The probability density for reactions to occur, i.e., for (m, m) pairs to intersect, at (x, t) is given by

$$\begin{aligned} \mathscr{R}_{p=1} &\simeq \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \delta_{mn} \mathscr{G}_{(m,n)}(x) \\ \mathscr{R}_{p=1} &\simeq \frac{c\varrho^2}{\sqrt{\pi c \varrho t}} \exp\left(-\frac{(\varrho x)^2}{c\varrho t}\right) \end{aligned} \tag{23}$$

in the limit of large m. This is the extrinsic reaction rate for the $p=1$ model and it implies the reaction front is a Gaussian random walker, covering a typical distance $\sim(ct/\varrho)^{1/2}$ in a time t.

For the general case $p \leqslant 1$ the points of intersection of these (m, m) pairs will be used as a convenient definition for the centre of the stochastically moving reaction zone. The forms of the intrinsic and extrinsic reaction rates can now be calculated.

A. The Intrinsic Reaction Rate

The intrinsic reaction rate as defined in Section 1, is the reaction-rate profile seen in a single realisation of the system's evolution (measured relative to the centre of the A and B domains' overlap region). In this section it will be argued that, if reactions are measured relative to the position of the (m, m) pair intersections, the intrinsic reaction rate is on average equal to $\mathscr{R}_0$, the reaction rate for the zero current-fluctuations case examined in Section 3.

Consider the set of intersecting particle pairs that can be written $(m-k, m+k)$ with m fixed and k varying. These pairs all share the same average time of intersection. Therefore, it is convenient to define the relative coordinates of intersection (x_r, t_r) of these pairs to the central (m, m) pair

$$2x_r = (y_{m-k} + z_{m+k}) - (y_m + z_m) \qquad 2ct_r = (z_{m+k} - y_{m-k}) - (z_m - y_m)$$

$$x_r = k\varrho^{-1} + O(k^{1/2}\varrho^{-1}) \qquad t_r = 0 + O(k^{1/2}(2c\varrho)^{-1})$$

where the $O(k^{1/2})$ deviations are those expected from the Gaussian fluctuations (21). As the deviations are not functions of m, the noise in the particles' initial positions does not produce any time-dependent dispersion, i.e., the statistics of the pair intersections in the overlap region reach a steady-state. The relative time and positions are on average the same for the equally-spaced case seen in Section 3, though the Gaussian fluctuations about these average values introduce $a \sim k^{1/2}\varrho^{-1}$ uncertainty in the position of the reactions. However, this broadening is not sufficiently strong to disrupt the $\mathscr{R}_0$ profile if the inequality $p^{1/2} \ll 1$ is satisfied, i.e., if the width of the reaction zone is much greater than the broadening. Rather, the fluctuations act to smooth the discontinuous nature of the equally-spaced reactant currents, for which a coarse-graining was necessary in

Section 3. Therefore, from Eq. (19) the steady-state intrinsic reaction rate is

$$\begin{aligned} \mathscr{R}_I(x_r) &= 2c\varrho^2 P(2\varrho x_r) \\ \mathscr{R}_I(x_r) &= \frac{pc\varrho^2}{2\cosh^2(p\varrho x_r)} + O(p^2) \end{aligned} \tag{24}$$

It also follows that the particle streams have the coarse-grained density distribution seen in the ordered case. Hence, Fig. 2 also represents the profiles seen in the case with Gaussian fluctuations in the particle currents.

The width of the intrinsic reaction zone is $\sim(p\varrho)^{-1}$ and from (23) in a time ΔT it will typically move (as a whole) a distance $\sim(c\Delta T/\varrho)^{1/2}$. Hence, any measurement made of the $\varnothing$ production rate must have $\Delta T \ll (p^2 c\varrho)^{-1}$. Such a restriction can indeed be satisfied, and the $\varnothing$ production rate that (24) predicts can be clearly seen in a Monte Carlo simulation of a single system, i.e., the system is self-averaging.

In the above analysis the steady state was assumed for the intrinsic reaction rate calculation. However, the arguments also follow through if the time-dependent intrinsic rate (19) is used, for $p \ll 1$. The width of the intrinsic reaction zone W_I therefore varies as

$$\begin{aligned} W_I &\propto (ct) \qquad \text{for} \quad t \ll t_I \\ W_I &\propto (p\varrho)^{-1} \qquad \text{for} \quad t \gg t_I \end{aligned}$$

where $t_I = (pc\varrho)^{-1}$ is the relaxation time for the intrinsic rate given in the previous section. Hence, at early times the width increases linearly, until finally saturating at a finite time-independent width.

It is unlikely that the present model studied is in the same universality class as the case of diffusive reactants. However, it is interesting to note that the asymptotics of the intrinsic reaction rates derived for the diffusive case and in the present case of ballistic reactants are both of the same form $\sim e^{-a|x|}$, in the steady state.

B. The Extrinsic Reaction Rate

The extrinsic reaction rate, defined as the reaction rate at (x, t) averaged over all possible evolutions, implies an average over all the appropriately weighted initial particle positions $\{y\}$ and $\{z\}$. It is therefore equivalent to the sum of probability densities for any (m, n) pair to annihilate at (x, t). After the intrinsic reaction rate has relaxed to its

steady-state limit, i.e., for times $t \gg t_I$, the extrinsic reaction rate can be expressed purely as a function of $s=(n-m)$

$$\mathscr{R}_E(x,t)=\sum_m \sum_n P(m,n)\,\mathscr{G}_{mn}(x,t)$$

$$\lim_{t\to\infty} \mathscr{R}_E(x,t)=\sum_s P(s)\,\mathscr{G}_s(x,t)$$

The function $\mathscr{G}_s(x,t)$ is the probability density that an $(m, m+s)$ pair meet at (x,t) for any value of m with s fixed, in the limit $t \gg (cQ)^{-1}$

$$\mathscr{G}_s(x,t)=\lim_{t\to\infty} \sum_{m=1}^{\infty} \frac{cQ^2}{\pi m} \exp\left(\frac{-1}{4m}((2Qx-s)^2+(2cQt-2m)^2)\right)$$

$$\mathscr{G}_s(x,t)=\frac{cQ^2}{\sqrt{\pi cQt}} \exp\left(\frac{-(2Qx-s)^2}{4cQt}\right)$$

Therefore, the extrinsic reaction rate can be written as a convolution of the intrinsic rate over all the allowed paths the intrinsic reaction zone can take (each being weighted by a Gaussian).

$$\mathscr{R}_E(x,t)=\frac{cQ^2}{\sqrt{\pi cQt}} \sum_{s=-\infty}^{\infty} P(s) \exp\left(\frac{-(2Qx-s)^2}{4cQt}\right)$$

$$\mathscr{R}_E(x,t)=\sqrt{\frac{p^2cQ^3}{16\pi t}} \sum_{s=-\infty}^{\infty} \frac{\exp\left(\dfrac{-(2Qx-s)^2}{4cQt}\right)}{\cosh^2(ps/2)}+O(p^2) \qquad (25)$$

A second characteristic time $t_E=(p^2cQ)^{-1}$ is now introduced into the system by the Gaussian noise. This is the timescale on which the uncertainty in the position of the intrinsic reaction zone becomes equal to its steady-state width, i.e., the standard deviation of the position of the reaction zone's centre is of the order $(pQ)^{-1}$. For $t \ll t_E$ the extrinsic and intrinsic rates are effectively the same, as there is little broadening. However, for times $t \gg t_E$ the noise in the particle currents dominates, the extrinsic reaction rate becomes a Gaussian and the result[9] is recovered. This is not surprising as for these times the structure of the intrinsic zone is unimportant and can be considered a point-like random walker. The width W_E of the extrinsic reaction rate as a function of time is therefore

$$W_E \propto (ct) \qquad \text{for} \quad t \ll t_I$$
$$W_E \propto (p\varrho)^{-1} \qquad \text{for} \quad t_I \ll t \ll t_E$$
$$W_E \propto (ct/\varrho)^{1/2} \qquad \text{for} \quad t \gg t_E$$

where $t_I = (pc\varrho)^{-1}$ and $t_E = (p^2 c\varrho)^{-1}$. The asymptotic behaviour (large x values) of the extrinsic reaction rate also has distinct forms as a function of time

$$\mathscr{R}_E \sim \theta(ct - \varrho\,|x|) \qquad \text{for} \quad t \ll t_I$$
$$\mathscr{R}_E \sim e^{-a|x|} \qquad \text{for} \quad t_I \ll t \ll t_E$$
$$\mathscr{R}_E \sim e^{-bx^2/t} \qquad \text{for} \quad t \gg t_E$$

Hence, there is a cross over from exponential to Gaussian behaviour at late times.

V. DISCUSSION

In the present work, a new model for the reaction system $A + B \to \varnothing$ with ballistic reactants has been introduced and solved exactly. The model includes two types of noise, in the reactions (due to the reaction probability being less than one) and in the currents of particles (due to disorder in the particles' initial positions). These fluctuations allow some of the characteristics of real systems to be exhibited, including a fluctuating A and B overlap region where the reactions occur, and a reaction zone that moves stochastically throughout the system. Comparison was made between the present model of ballistically moving reactants and that of isotropically diffusing reactants[12] studied under the RG framework. In particular, it was noted that the intrinsic reaction rates (the dynamic reaction region formed between the fluctuating A and B domains) have the same asymptotic form, though it is unclear if the two systems share the same universality class. An important physical difference between these two cases is that, in the ballistic model, particle order is preserved and interactions between a given pair of particles can occur only once. It would therefore be interesting to study the case of reactants that perform biased diffusion, because such a system would interpolate between the cases of isotropic diffusion and ballistic motion of reactants. As shown in the appendix, the method outlined in Section 2 can be translated into the second-quantisation formalism with the vacancy dynamics described by the $SU_q(2)$ algebra. Using the tools available in this formalism it may be possible to introduce interaction terms in the evolution operator that break the order of the particles,

thereby introducing a bias-diffusive component into the otherwise deterministic motion.

Another case for further study would be to examine more closely the relation between the present model and the partially-asymmetric exclusion process (PASEP) with reflecting boundaries—also described by $SU_q(2)$.[24] The present model displays short-range correlations, as does the PASEP with reflecting boundaries. However, the PASEP with open boundaries is known to have three phases,[25-29] one of which has long-range power-law behaviour. It would therefore be interesting to see if the open system also translates into a system of reacting particles.

APPENDIX. "SECOND-QUANTISATION" FORMULATION

As stated in Section 2, the method used to calculate $P(m, n)$ can be viewed in terms of quantum-mechanical interacting particles. Here this formulation is briefly reviewed and the recipe for calculating quantities of interest is described.

The system studied in Section 2 consists of a one-dimensional lattice with sites numbered 1, 2..., etc. A lattice site k, described by the binary variable η_k, can either be occupied by a particle or a vacancy. If a vacancy is present at site k then $\eta_k = 1$, otherwise $\eta_k = 0$. Hence, the state of a single system is described by the set of variables $\eta \equiv \{\eta_k\}$ and can be represented as a vector in a Fock space, i.e.,

$$|\eta\rangle = |\eta_1, \eta_2, \cdots \eta_k \cdots\rangle$$

$$\langle \eta' | \eta \rangle = \prod_{k=1}^{\infty} \delta(\eta'_k, \eta_k) = \delta(\eta', \eta)$$

Initially the system is full of B particles, represented by the state $|0\rangle$. The evolution of the system involves an A particle being "shot" through the B array at time steps $m = 1, 2...$, etc. Each A particle passes through a B with probability q, or annihilates with the B with probability p. If no B is present at a site the A particle passes through to the next site with probability one, see Fig. 1. After each time step m, the system will have some probability $P_m(\eta)$ of being in the state η. The *vacancy* creation and annihilation operators are now introduced

$$C_k^+ | \cdots 0_k \cdots \rangle = \left[p \prod_{i=1}^{k-1} q^{(1-\eta_i)} \right] | \cdots 1_k \cdots \rangle$$

$$C_k | \cdots 1_k \cdots \rangle = \left[p \prod_{i=1}^{k-1} q^{(1-\eta_i)} \right]^{-1} | \cdots 0_k \cdots \rangle$$

with the auxiliary relations $C_k^+ \mid \cdots 1_k \cdots \rangle = C_k \mid \cdots 0_k \cdots \rangle = 0$. Defining the q-commutator as $[A, B]_q = AB - qBA$, the following commutations relations hold for $n_1 < n_2$

$$\begin{gathered}[C_{n_1}^+, C_{n_2}^+]_q = [C_{n_2}, C_{n_1}^+]_q = [C_{n_1}, C_{n_2}^+]_q = [C_{n_2}, C_{n_1}]_q = 0 \\ [C_n, C_n^+]_+ = 1\end{gathered} \tag{26}$$

where the final anticommutator is for same-site operators. These commutation relations provide a representation of $SU_q(2)$, i.e., "q-deformed" spin-half particles.[28] The evolution equation for the system can now be written in terms of these vacancy creation and annihilation operators

$$|P_m\rangle = \sum_\eta P_m(\eta)\, |\eta\rangle = \hat{T}^m\, |0\rangle$$

$$\hat{T} = \sum_{k=1}^{\infty} C_k^+$$

where $\hat{T}$ is the evolution operator. Introducing the left state $\langle S| = \sum_{\eta'} \langle \eta'|$ any observable $A(P_m)$ represented by the operator $\hat{A}$ can be written in the following way

$$\langle S|\, \hat{A}\hat{T}^m\, |0\rangle = \sum_{\eta'} \sum_{\eta} \langle \eta' | \eta \rangle\, A(\eta)\, P_m(\eta)$$

$$\langle S|\, \hat{A}\hat{T}^m\, |0\rangle = \sum_{\eta} A(\eta)\, P_m(\eta)$$

In evaluating such observables it is useful to use a factorisation characteristic of the creation operators. If a string of creation operators is arranged, such that each operator acts in descending order (with respect to its subscript) on the right empty state, then the following property can be used

$$\langle S|\, C_{n_1}^+ C_{n_2}^+ \cdots C_{n_m}^+\, |0\rangle = \langle S|\, C_{n_1}^+\, |0\rangle \langle S|\, C_{n_2}^+\, |0\rangle \cdots \langle S|\, C_{n_m}^+\, |0\rangle \tag{27}$$

where the single-vacancy expectations are simply $\langle S|\, C_k^+\, |0\rangle = pq^{k-1}$.

In the formalism described above, all quantities expressible in terms of operators can be evaluated. Therefore

$$\Psi(n_1, \ldots, n_m) = \langle S|\, \sum_{perms} C_{n_1}^+ C_{n_2}^+ \ldots C_{n_m}^+\, |0\rangle$$

$$V_m(n) = \langle S|\, C_n^+ C_n \hat{T}^m\, |0\rangle$$

$$P(m, n) = \langle S|\, C_n^+ \hat{T}^{m-1}\, |0\rangle$$

are the forms for each of the objects calculated in Section 2. These can be evaluated by using the commutation relations (26) in the following way. Firstly for a given string, commute all annihilation operators to the right of the string. These objects when acting on the left state $\langle S|$ will leave strings containing only creation operators. These strings can then be rearranged by using the appropriate relation in (26) so that they act on the zero state in descending order of subscript. Finally the factorisation property (27) can be used to calculate the expectation value.

Interestingly, the algebraic structure outlined here is identical to that used in the description of the partially-asymmetric exclusion process (PASEP) with reflecting boundary conditions, therefore corresponding to a noisy Burgers equation with zero *average* current.[24] Microscopically, the PASEP describes systems of particles that hop in a preferred direction with a repulsive interaction. Assuming that the particles hop with a leftwards bias, the density at site n seen when m such particles are confined to a semi-infinite lattice ($[1:\infty]$), is identical to the vacancy density $V_m(n)$ given by Eq. (10).

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