

# A Two-Particle Representation of Front Propagation in Diffusion-Reaction Systems

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A two-particle model is formulated which approximates the motion of the forwardmost particle in a lattice gas, which has recently been analyzed and numerically simulated. The lattice gas, which evolves on a linear chain, consists of particles which jump to each vacant nearest neighbor site with rate  $\gamma/2$  and also create new particles at these sites with rate  $1/2$ . This model is known to exhibit statistically steady propagation of the forwardmost particle, with mean propagation velocity converging to  $(2\gamma)^{1/2}$  for large  $\gamma$ . Here, a two-particle representation is used to estimate the propagation velocity for finite  $\gamma$ . The results are in good agreement with numerical simulations of the lattice gas. Implications concerning front propagation in diffusion-reaction systems are discussed.

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**KEY WORDS:** Lattice gas; interacting particle system; velocity selection; diffusion-reaction equation.

## 1. INTRODUCTION

A lattice-gas model which provides a simple paradigm for front propagation in diffusion-reaction systems has recently been introduced.<sup>(1-4)</sup> The model consists of sites, labeled by integers  $-\infty < i < +\infty$ , each of which is vacant [state  $\eta(i)=0$ ] or occupied by a single particle [state  $\eta(i)=1$ ]. A particle adjacent to a vacant site jumps to that site with rate  $\gamma/2$ , or creates a new particle at that site with rate  $1/2$ . Equivalently, the state vector  $\eta(i)$  evolves by independent, random exchanges of state between each pair of adjacent sites with rate  $\gamma/2$  per pair, and by random change of each vacant site  $i$  to occupied at a rate  $[\eta(i-1) + \eta(i+1)]/2$ .

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The model captures certain qualitative features of interacting particle systems involving diffusion and irreversible chemical reaction. In particular, the site occupancy  $u(i) = P[\eta(i) = 1]$  obeys the diffusion-reaction equation

$$u_t = \frac{1}{2}u_{xx} + u(1 - u) \quad (1)$$

where  $x = i/\sqrt{\gamma}$ , in the hydrodynamic limit  $\gamma \rightarrow \infty$ . Solutions of Eq. (1) are known<sup>(5,6)</sup> to relax from a step-function initial condition to a steadily propagating form  $u(x, t) = u(x - v_c t/\sqrt{\gamma})$ , where

$$v_c = (2\gamma)^{1/2} \quad (2)$$

( $c$  denotes the continuum limit.)

For the lattice-gas model, various equivalent definitions of the propagation velocity  $v$  can be formulated.<sup>(2,3)</sup> Here it is defined to be the velocity of the rightmost particle, based on an initial state in which there exists a rightmost particle.

It has recently been proven<sup>(2)</sup> that  $v_c$  as given by Eq. (2) is also the propagation velocity for the lattice-gas model in the limit  $\gamma \rightarrow \infty$ . This result demonstrates a microscopic selection principle consistent with the selection principle governing the continuum formulation.

It has been noted<sup>(3,4)</sup> that the  $\gamma$  dependence of the propagation velocity  $v$  away from the hydrodynamic limit is also physically and mathematically interesting. For small  $\gamma$ , it is easily shown<sup>(2,4)</sup> that

$$v = (1 + \gamma)/2 \quad (3)$$

to  $O(\gamma)$ . It is not obvious what functional form  $v(\gamma)$  assumes in the transition between the limiting forms (2) and (3). Computations<sup>(3,4)</sup> indicate that the large- $\gamma$  result (2) is approached slowly as  $\gamma$  increases. No explanation of this slow convergence has been offered.

This slow convergence and other features of  $v(\gamma)$  are of physical interest in the context of flame-front propagation through turbulent fuel-air mixtures.<sup>(4)</sup> Various functional dependences have been postulated<sup>(7)</sup> to relate measured flame speed to turbulence intensity. It is recognized that the respective limits of strong and weak turbulence intensity are qualitatively distinct, but no model has been proposed which captures both limits and the transition between them. If the exchange-rate parameter  $\gamma$  of the lattice-gas model is interpreted as the scaled turbulence intensity, then  $v(\gamma)$  can be interpreted as the turbulent flame speed. On this basis, numerical simulations of the lattice gas have been compared to measured flame speeds.<sup>(4)</sup>

No progress has been reported in solving exactly for  $v(\gamma)$  away from the respective limits. Here, an approximate solution is obtained for all  $\gamma$

by formulating a two-particle representation of front propagation. The two-particle model involves a self-consistent correction to compensate for neglected multiparticle effects, with a free parameter that is adjusted so that the model satisfies Eq. (2) for large  $\gamma$ . The model yields an algebraic equation involving  $v$  and  $\gamma$ . This equation gives numerical values of  $v$  which are within 10% of values obtained from the lattice-gas simulations, which span four orders of magnitude in  $\gamma$ . Curiously, the best agreement is obtained for  $\gamma$  of order unity, for which multiparticle effects are most influential. At large  $\gamma$ , the previously noted<sup>(3,4)</sup> slow convergence to the asymptotic  $\gamma$  dependence is reproduced. The latter result implies that the convergence is not necessarily slow in the analytical sense, although it is numerically slow. Namely, the two-particle model yields an algebraic correction to Eq. (2) rather than, say, a logarithmic correction.

The two-particle model is also compared to an *ad hoc* explicit expression for  $v$ . The *ad hoc* expression does not perform nearly as well. It is hoped that the present results will stimulate further efforts to determine  $v(\gamma)$  either exactly or by a controlled approximation.

Analysis of the two-particle model leads to a master equation which governs the transient evolution of the propagation velocity as well as its steady-state value. The transient evolution, which is of physical interest with regard to the early growth of a flame kernel, will be investigated in future work.

## 2. MOTIVATION FOR THE TWO-PARTICLE REPRESENTATION

The propagation velocity of the rightmost particle in the lattice gas may be expressed as

$$v = \frac{1}{2} + \frac{1}{2}\gamma P[\eta(i_r - 1) = 1] \quad (4)$$

where  $i_r = \max\{i: \eta(i) = 1\}$  is the location of the rightmost particle. The two-particle representation provides an estimate of  $P[\eta(i_r - 1) = 1]$  and thus of  $v$ .

Equation (4) is obtained as the sum of the respective contributions of creation and jump events to the propagation velocity. Since site  $i_r + 1$  is vacant by definition, particle creation occurs at that site at a constant rate  $1/2$ , accounting for the first term in Eq. (4). Jump events induce no net velocity of the rightmost particle if site  $i_r - 1$  is vacant, since the particle is equally likely to move in either direction. If site  $i_r - 1$  is occupied, only rightward jumps are allowed, inducing a net velocity  $\gamma/2$ . Multiplication by the probability of the latter condition yields the second term of Eq. (4).

The two-particle representation is motivated by the velocity-selection principle governing Eq. (1). Namely, if the source term of that equation is

generalized to  $f(u)$ , where  $f$  is positive and concave for  $0 < u < 1$ , then the propagation velocity, expressed in the coordinate  $i$ , is<sup>(5)</sup>  $v_c = [2\gamma f'(0)]^{1/2}$ . This result indicates that  $v_c$  is sensitive only to the dynamics governing the forwardmost development of the site-occupancy profile. Based on the correspondence between the lattice-gas model for large  $\gamma$  and Eq. (1), and the equality of their respective propagation velocities in this limit, this inference may also be valid for the large- $\gamma$  limit of the lattice gas.

These considerations motivate a truncated representation of the instantaneous state of the lattice gas, in which all but the two rightmost particles are discarded. Whenever a creation event adds a third particle, the leftmost of the three particles is discarded. Numbering particles from right to left, this representation differs from the dynamics of particles 1 and 2 of the lattice gas in that the effect of particle 3 on particle 2 is omitted. To compensate for this omission, a self-consistent correction to the dynamical rules governing particle 2 is proposed. (The correction is self-consistent in that it contains a quantity which governs the dynamics of particle 1.) The correction involves a free parameter that is adjusted so that the model satisfies Eq. (2) for large  $\gamma$ .

Consideration only of the two rightmost particles is intuitively sound also in the limit  $\gamma \rightarrow 0$ . In this limit, the lattice gas is rarely in a state such that the site to the left of particle 2 is vacant, so the interaction of particles 1 and 2 with the rest of the system may be approximated by assuming that particles 2 and 3 are always adjacent. The self-consistent correction incorporates this small- $\gamma$  behavior.

The two-particle representation is difficult to justify for  $\gamma$  of order unity, for which neither rationale is applicable. Curiously, the model which is proposed based on consideration of the limiting behaviors is found to be quantitatively most accurate at  $\gamma$  of order unity. It is so accurate, in fact, that it is difficult to dismiss the numerical agreement as fortuitous. The results thus raise an interesting issue for analytical investigation.

### 3. UNCORRECTED TWO-PARTICLE MODEL

In this section, a two-particle system is analyzed in which the particles obey the same dynamics as the lattice gas, except that the leftmost particle is discarded after each creation event. In Section 4, the dynamics is modified based on the consideration of limiting behaviors.

The state of the two-particle system is given by the respective locations  $i_1$  and  $i_2 < i_1$  of particles 1 and 2. Since the evolution of the two-particle system depends only on the relative separation of the particles, the system is sufficiently characterized by the number  $j = i_1 - i_2 - 1$  of vacant sites between the particles. A master equation<sup>(8)</sup> is formulated for the

probabilities  $\{p_j\}$  of states  $j=0, 1, \dots$ , where  $p_0$  is the two-particle analog of the quantity  $P[\eta(i_r - 1) = 1]$  in Eq. (4).

The master equation is obtained by determining the rates for all transitions to and from a given state  $j$ . For example, state  $j > 0$  can be reached from state  $j - 1$  by a rightward jump of particle 1 or by a leftward jump of particle 2, each of which occurs with rate  $\gamma/2$ . Thus, the  $j - 1 \rightarrow j$  transition rate is  $\gamma$ . State  $j > 0$  can be reached from state  $j + 1$  by analogous mechanisms, and also by a creation event at site  $i_2 + 1$ , which occurs with rate  $1/2$ . State 0 can be reached from any state  $j > 0$  by a creation event at site  $i_1 + 1$  or at site  $i_1 - 1$ . For  $j > 1$ , each of these events occurs at rate  $1/2$ . For  $j = 1$ , however, creation at site  $i_1 - 1 = i_2 + 1$  occurs at rate 1. Jump events with total rate  $\gamma$  also contribute to the  $1 \rightarrow 0$  transition rate.

Having thus specified all the nonzero transition rates, one can express the master equation for the uncorrected two-particle model as

$$\dot{p}_j = \gamma p_{j-1} + (\gamma + \frac{1}{2}) p_{j+1} - (2\gamma + \frac{3}{2}) p_j; \quad j > 0 \quad (5)$$

$$\dot{p}_0 = (\gamma + \frac{1}{2}) p_1 + 1 - (1 + \gamma) p_0 \quad (6)$$

[The normalization  $\sum_{j=0}^{\infty} p_j = 1$  has been used to simplify Eq. (6).]

Equations (5) and (6) govern the transient evolution as well as the steady-state equilibrium of the uncorrected two-particle model. The transient evolution may be obtained by solving Eqs. (5) and (6) for the initial condition  $p_0 = 1$ ,  $p_j = 0$  for  $j > 0$ . The equilibrium is obtained by setting all time derivatives equal to zero, yielding a set of linear recursion relations. The *ansatz*  $p_j = p_0(1 - p_0)^j$  for all  $j > 0$  satisfies Eqs. (5) and (6) and gives a quadratic equation for  $p_0$  whose solution satisfying  $0 \leq p_0 \leq 1$  for nonnegative  $\gamma$  is

$$p_0 = \frac{1}{4\gamma + 2} [(16\gamma + 9)^{1/2} - 1] \quad (7)$$

For  $\gamma = 0$ , Eq. (7) gives  $p_0 = 1$ , consistent with the behavior of the lattice-gas model as  $\gamma \rightarrow 0$ . To leading order in  $\gamma \gg 1$ , Eq. (7) gives  $p_0 = \gamma^{-1/2}$ . Substitution of this result into Eq. (4) gives  $v = (\gamma/4)^{1/2}$  to leading order, which is a factor of  $8^{1/2}$  smaller than the lattice-gas result, Eq. (2). This indicates that neglected multiparticle effects contribute to leading order in the hydrodynamic limit of the lattice gas.

#### 4. SELF-CONSISTENT CORRECTION

To compensate for neglected multiparticle effects, a self-consistent correction to the two-particle model is proposed. Particle 3 influences the

dynamics of particles 1 and 2 only when it is located at  $i_3 = i_2 - 1$ . Its influence under that circumstance is to prevent leftward jumps of particle 2. The assumption is adopted that the probability  $p_a$  that particle 3 is adjacent to particle 2 is independent of  $j$ . Therefore, wherever a rate  $\gamma/2$  associated with a leftward jump of particle 2 contributed to a transition rate computed in Section 3, that rate is now replaced by  $\frac{1}{2}\gamma(1 - p_a)$ . The corrected master equation may now be expressed as

$$\dot{p}_j = \gamma \left(1 - \frac{p_a}{2}\right) p_{j-1} + \left(\gamma + \frac{1}{2}\right) p_{j+1} - \left(2\gamma - \gamma \frac{p_a}{2} + \frac{3}{2}\right) p_j; \quad j > 0 \quad (8)$$

$$\dot{p}_0 = \left(\gamma + \frac{1}{2}\right) p_1 + 1 - \left(\gamma - \gamma \frac{p_a}{2} + 1\right) p_0 \quad (9)$$

To complete the formulation of the corrected model, an expression for the dependence of  $p_a$  on  $\gamma$  must be specified. Since the correction is motivated by the need to incorporate multiparticle effects at large  $\gamma$ , the behavior of  $p_a$  in that limit is first considered. A feature of the continuum formulation, Eq. (1), is the vanishing of the derivative of  $u$  with respect to the spatial coordinate  $i$  as  $\gamma \rightarrow \infty$ . This implies weak spatial variation of the particle number density in the lattice gas. Therefore,  $p_a$  should be of order  $p_0 \ll 1$  for large  $\gamma$ . This motivates the adoption of the expression

$$p_a = Bp_0 - (B - 1)p_0^2 \quad (10)$$

which specifies the  $\gamma$  dependence of  $p_a$  implicitly for all  $\gamma$  through the  $\gamma$  dependence of  $p_0$ . Equation (10) is of order  $p_0$  for large  $\gamma$ , and has an additional term which assures that  $p_a$  has the correct behavior at  $p_0 = 1$ , namely  $p_a = 1$ . The  $B$  is a free parameter which is evaluated shortly.

The correction procedure which has been adopted is self-consistent in that the correction term is dependent on the quantity whose solution is sought. In this respect, the procedure bears some analogy to self-consistent closures sometimes adopted in truncation procedures for kinetic-equation hierarchies. The present approximation procedure may be regarded as a simple paradigm of such truncations, particularly because the present procedure is amenable to systematic improvement by generalizing to  $n$ -particle representations (albeit with as yet unknown convergence properties for large  $n$ ).

The equilibrium solution of Eqs. (8) and (9) is obtained, as before, by adopting the *ansatz*  $p_j = p_0(1 - p_0)^j$  for all  $j \geq 0$ , which satisfies those equations provided that

$$(2\gamma + 1)p_0^2 - (\gamma p_a - 1)p_0 - 2 = 0 \quad (11)$$

Substitution of Eq. (10) for  $p_a$  gives

$$\gamma(B-1)p_0^3 + (2\gamma - \gamma B + 1)p_0^2 + p_0 - 2 = 0 \quad (12)$$

The parameter  $B$  is evaluated by requiring  $p_0 = (8/\gamma)^{1/2}$  to leading order in  $\gamma \gg 1$ , consistent with Eqs. (2) and (4). With this requirement, Eq. (12) gives  $B = 7/4$ . For this value of  $B$ , Eq. (10) has the desirable property that  $p_a$  is a strictly increasing function of  $0 \leq p_0 \leq 1$ .

Equation (12), a cubic equation for  $p_0$ , can be solved explicitly for  $p_0(\gamma)$ . Alternatively, Eq. (2) can be used to reexpress this equation as a cubic equation for  $v$ , again explicitly solvable. To obtain numerical results, it is more convenient to work directly with Eq. (12), rewritten as

$$\gamma = \frac{8 - 4p_0(p_0 + 1)}{p_0^2(3p_0 + 1)} \quad (13)$$

From this expression,  $\gamma$  is determined for any  $p_0$ , and  $v$  is then given by Eq. (4).

The corrected two-particle model thus predicts a  $\gamma$  dependence of  $v$  which is purely algebraic and therefore is devoid of terms like  $\ln \gamma$ . This observation is pertinent to the interpretation of the numerical results presented in the next section.

## 5. NUMERICAL RESULTS AND COMPARISONS

In Fig. 1, the  $\gamma$  dependence of  $v$  computed using Eq. (13) is displayed along with simulated results for the lattice gas, which were reported in ref. 3. As in that reference, the quantity  $v/v_c$  is shown in order to highlight the convergence to the hydrodynamic limit. For each  $\gamma$  value, the plotted simulation result is the average of the velocity estimates corresponding to the largest value of the relaxation-time parameter  $k$  of ref. 3.

In addition to the lattice-gas results of ref. 3, new simulation results are plotted, corresponding to an  $n$ -particle generalization of the uncorrected two-particle model. In these simulations, the leftmost particle is discarded after each creation event. Simulations were performed for  $n = 3, 5, \text{ and } 10$ , where  $n$  is the initial number of particles. The interface method described in ref. 3 was used. For each  $\gamma$  value,  $k$  was chosen to be as large as or larger than the value of  $k$  in the corresponding lattice-gas simulation. Each plotted point is the average of two velocity estimates, one based on Eq. (4) and one based on the velocity of the rightmost particle.

Also shown in Fig. 1 is an *ad hoc* functional dependence,

$$v = \frac{1}{2} + \frac{1}{2}\gamma\left(1 + \frac{1}{8}\gamma\right)^{-1/2} \quad (14)$$

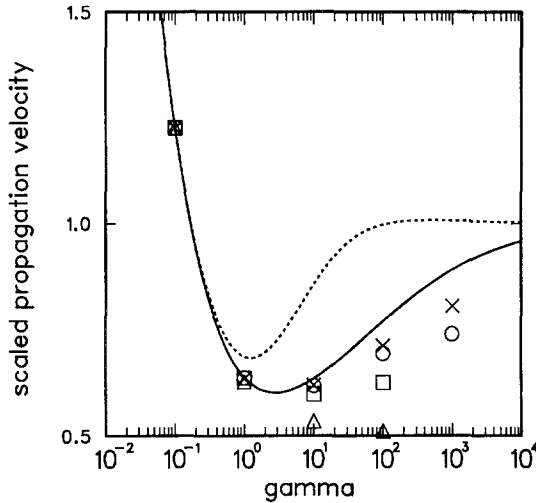


Fig. 1. Dependence of the scaled propagation velocity  $v/v_c$  [where  $v_c = (2\gamma)^{1/2}$ ] on the exchange-rate parameter  $\gamma$ . Simulation results are shown for the lattice gas<sup>(3)</sup> ( $\times$ ) and for the uncorrected  $n$ -particle model, where  $n=3$  ( $\Delta$ ), 5 ( $\square$ ), and 10 ( $\circ$ ). Also shown are the  $\gamma$  dependence of the corrected two-particle model (solid curve) and an *ad hoc* functional dependence (dashed curve). For all simulation results, the error bars are narrower than the width of the solid curve.

which is based on Eq. (4) and the assumption  $P[\eta(i_r - 1) = 1] = [1 + (\gamma/8)]^{-1/2}$ . The latter expression is adopted because it is a simple form which interpolates between known behaviors at large  $\gamma$  and at  $\gamma=0$ . Equation (14) is included in the comparisons in order to assess the utility of the mechanistic approximation of the previous section relative to straightforward interpolation.

As shown in the figure, values of  $v$  predicted by the corrected two-particle model are within 10% of values obtained from the lattice-gas simulations, which span four orders of magnitude in  $\gamma$ . As remarked earlier, the best agreement is obtained for  $\gamma$  of order unity, for which multiparticle effects are ostensibly most influential. At large  $\gamma$ , both the simulations and the two-particle representation exhibit slow convergence to the asymptotic  $\gamma$  dependence. This indicates that the convergence is not necessarily slow in the analytical sense (e.g., logarithmic), since the two-particle functional dependence is purely algebraic, as noted earlier. This question can be resolved conclusively only by analytic investigation.

Figure 1 also indicates the superior performance of the two-particle representation relative to the *ad hoc* postulated form, Eq. (14). This provides strong evidence of the mechanistic validity of the two-particle



representation, particularly at intermediate  $\gamma$  values for which it has no obvious justification.

The  $n$ -particle simulation results indicate that is not necessary to go to very large  $n$  to obtain results as good as or better than the corrected two-particle prediction over a wide range of  $\gamma$  values. It is evident, however, that convergence of the  $n$ -particle results to the lattice-gas results is not uniform in  $\gamma$ . It is not known whether a corrected  $n$ -particle representation can be formulated so as to obtain uniform convergence.

## 6. DISCUSSION

The salient features of the two-particle representation of front propagation in the lattice gas are as follows. First, it exploits the velocity-selection principle, which indicates that the forwardmost particles of the lattice gas are most influential in determining the propagation velocity in the hydrodynamic limit. Second, it incorporates a self-consistent correction which is formulated so as to match known results in the respective limits of large and small exchange rate. Third, numerical results are in good agreement with lattice-gas simulations, particularly at exchange rates of order unity, for which the approximate representation has no *a priori* justification. Fourth, the approach is subject to successive improvements by generalizing to an  $n$ -particle representation, although convergence, either pointwise or uniformly with respect to the exchange-rate parameter  $\gamma$ , has not been proven.

It has been noted that the lattice-gas model is a simple paradigm for diffusion-reaction systems, and that the two-particle representation is likewise a paradigm for closure approximations to kinetic-equation hierarchies. The former analogy was previously exploited in order to interpret turbulent flame propagation measurements.

Another potentially useful analogy concerns recent efforts to formulate approximate models of interface evolution.<sup>(9)</sup> Namely, phenomenological reasoning is used to obtain differential equations governing the propagation of an interface based only on local geometrical properties of the interface, in particular its curvature. In other work,<sup>(10)</sup> it has been shown that stochastic models of the microscopic dynamics can be useful for deriving properties of the differential equations obtained in the continuum limit. Here, an approximate procedure has been developed which, in effect, localizes the dynamics governing a stochastic interface propagation problem. The success of this approach suggests the following strategy for approximating other nonlocal interface propagation problems: local approximations to the underlying microscopic dynamics might approximate the nonlocal dynamics in the continuum limit.

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