Anisotropic Diffusion-Limited
Reactions with Coagulation and Annihilation

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ABSTRACT

One-dimensional reaction-diffusion models $A + A \rightarrow \emptyset$, $A + A \rightarrow A$, and $A + B \rightarrow \emptyset$, where in the latter case like particles coagulate on encounters and move as clusters, are solved exactly with anisotropic hopping rates and assuming synchronous dynamics. Asymptotic large-time results for particle densities are derived and discussed in the framework of universality.

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Diffusion-limited reactions involving aggregation and annihilation processes are important in many physical, chemical and biological phenomena [1] such as star formation, polymerization, recombination of charge carriers in semiconductors, soliton and anti-soliton annihilation, biologically competing species, etc. In this work we study by exact solution effects of anisotropy in 1D, for single-species reactions $A + A \rightarrow A$ or $\emptyset$, and a two-species annihilation model $A + B \rightarrow \emptyset$ in which like particles coagulate irreversibly. Scaling approaches [1-2] suggest that in 1D these reactions are fluctuation-dominated, and we cannot expect the rate equation approach to be valid. Indeed the mean-field rate equation approximation ignores effects of inhomogeneous fluctuations. Exact solutions and asymptotic arguments, in 1D, have been used [3] to check general scaling and universality expectations. The 1D reactions have also found some experimental applications [4]. These studies have assumed isotropic hopping (diffusion).

For the reaction $A + B \rightarrow \emptyset$, numerical results and phenomenological considerations suggest [5] that making the hopping fully directed would change the universality class in 1D. Specifically, the large-time particle concentrations (assuming equal densities of both species) would scale according to $c(t) \sim t^{-1/3}$ instead of the isotropic-hopping power law $t^{-1/4}$. A few exact and numerical results available in the literature on anisotropic reactions involving only one species [6] indicate that the power law is not changed. The model of [5] assumed that like particles interact via hard-core repulsion; this seems to be an essential ingredient for observing the changeover in the universality class.

In this work we report the exact solution for two-particle annihilation with anisotropic hopping. However, in order to achieve exact solvability we took “sticky-particle” rather than hard-core interactions: the like particles coagulate on encounters and diffuse as groups. Our exact calculations yield the $t^{-1/4}$ power law, found earlier for “sticky-particles” with different dynamics and isotropic hopping [7]. For unequal initial concentrations, the large-time behavior changes; the crossover between the two regimes is de-
rived analytically. We also obtain exact results for \( A + A \rightarrow A \) or \( A \) with anisotropy. The universality class of dynamics of these reactions is not affected by hopping anisotropy. An extended version of this work will be reported elsewhere [8].

In lattice models the particles hop randomly, to the extent allowed by their interactions, to their nearest neighbor sites. Two like particles can annihilate on encounters, \( A + A \rightarrow \emptyset \), or aggregate, \( A + A \rightarrow A \). The 1D kinetics of these reactions is non-mean-field, with the typical large time diffusional behavior of the concentration (density per site), \( c(t) \sim t^{-1/2} \). For the two-species model, to be termed the \( AB \) model, unlike particles annihilate, \( A + B \rightarrow \emptyset \). When like species meet, some interaction must be assumed. The simplest interaction is hard-core. Assuming equal \( A \)- and \( B \)-concentrations and random initial conditions, particle concentrations in the isotropic case scale according to \( c(t) \sim t^{-1/4} \) in 1D. A surprising recent result [5] is the new exponent \( \approx 1/3 \), replacing \( 1/4 \), for anisotropic hard-core particle hopping.

In order to obtain a solvable model in 1D, we consider the \( AB \) annihilation model with the “sticky particle” interaction. Thus, like particles coagulate irreversibly on encounters, e.g., \( nA + mA \rightarrow (n + m)A \), and diffuse as clusters. When unlike clusters meet at a lattice site, the outcome of the reaction is \( nA + mB \rightarrow (n - m)A \) if \( n > m \), \( \emptyset \) if \( n = m \), and \( (m - n)B \) if \( n < m \). Recent numerical results and scaling considerations for these reactions [9] in \( D = 1, 2, 3 \) indicate that they are mean-field in \( D = 2, 3 \). However, in 1D the power-law exponent for the density is \( 1/4 \) [7,9], with a faster power-law decay \( \sim t^{-3/2} \) of the minority species in case of unequal densities of \( A \) and \( B \).

Following [10], we first consider diffusion of nonnegative charges on the 1D lattice. Initially, at \( t = 0 \), we place positive unit charge at each site with probability \( p \) or zero charge with probability \( 1 - p \). Furthermore, we consider synchronous dynamics, i.e., charges at all lattice sites hop simultaneously in each time step \( t \rightarrow t + 1 \), where the probabilities of hopping to the right, \( r \), and to the left, \( \ell = 1 - r \), are not necessarily
equal. This dynamics decouples the even-odd and odd-even space-time sublattices; it suffices to consider only those charges which are at the lattice sites \( j = 0, \pm 2, \pm 4, \ldots \) at times \( t = 0, 2, 4, \ldots \), and lattice sites \( j = \pm 1, \pm 3, \pm 5, \ldots \) at times \( t = 1, 3, 5, \ldots \). The “interaction” between the charges is defined by the rule that all charge accumulated at site \( j \) at time \( t \) coagulates. There can be 0, 1 or 2 such charges arriving at \( j \), depending on the random decisions regarding the directions of hopping from sites \( j \pm 1 \).

This model can also be viewed as diffusion-coagulation of unit-charge “particles" \( C \), i.e., \( nC + mC \rightarrow (n + m)C \). Such reactions, without the limitation of positive or integer charges, and with an added process of feeding-in charge at each time step, have been considered as models of self-organized criticality and coagulation [11-12], assuming isotropic hopping, \( r = \ell = \frac{1}{2} \). This coagulation reaction can be mapped [7,10] onto both our single-species and “sticky” \( AB \) models. However, before discussing this mapping, let us present the exact solution of the model of coagulating charges with anisotropic hopping, following the ideas of [10-11].

We define stochastic variables, \( \tau_j(t) = 1 \) or 0, with probabilities \( r \) and \( \ell \), respectively. The stochastic equation of motion for the charges \( q_j(t) \), equal to the number of \( C \) particles at site \( j \) at time \( t \), is

\[
q_n(t + 1) = \tau_{n-1}(t)q_{n-1}(t) + [1 - \tau_{n+1}(t)]q_{n+1}(t) .
\]

(1)

The total number of \( C \)-particles, or the total charge, in an interval of \( k \) consecutive proper-parity-sublattice sites, starting at site \( j \) at time \( t \), is given by

\[
S_{k,j}(t) = \sum_{i=0}^{k-1} q_{j+2i}(t) = q_j(t) + q_{j+2}(t) + \cdots + q_{j+2k-2}(t) .
\]

(2)

Due to conservation of charge, the equations of motion (1) yield the following relation,

\[
S_{k,n}(t+1) = \tau_{n-1}(t)q_{n-1}(t) + q_{n+1}(t) + \cdots + q_{n+2k-3}(t) + [1 - \tau_{n+2k-1}(t)]q_{n+2k-1}(t) .
\]

(3)
Thus, only the two random decisions at the end points are involved in the dynamics of charges in consecutive-site intervals. The exact solvability of coagulating-charge models is based on this property [11].

Let us introduce the function $I(s, m) = \delta_{s,m}$, and averages, $f_{k,m}(t) = \langle I(S_{k,j}(t), m) \rangle$. The averaging is over the stochastic dynamics, i.e., over $\tau_i(t)$, as well as over the initial conditions. Since the latter are uniform, $f_{k,m}(t)$ do not depend on $j$. Other choices for $I(s, m)$ have been used [10-12]. In our case $f_{k,m}(t)$ correspond to the probability to find $m$ charge units in an interval of $k$ sites, so that $f_{1,m}(t)$ is the density (fraction) of sites with charge $m$.

The variables $\tau_i(t)$ and $S_{k,n}(t)$ are statistically independent because the latter depend only on “decision making” at earlier times. As a result, one obtains, by (3), the discrete diffusion-like equation,

$$f_{k,m}(t+1) = r\ell \left[ f_{k+1,m}(t) + f_{k-1,m}(t) \right] + (r^2 + \ell^2) f_{k,m}(t) . \tag{4}$$

The $m$-dependence only enters via the initial conditions: $f_{k,m}(0) = p^m (1 - p)^{k-m} \binom{k}{m}$, provided $0 \leq m \leq k$, and 0 for $m > k$. We also define the boundary conditions $f_{0,m}(t) = I(0, m) = \delta_{0,m}$ in order to extend (4) to all $t = 0, 1, 2, \ldots$.

In order to solve (4) we introduce the double generating function, $g_k(u, w) = \sum_{t=0}^{\infty} \sum_{m=0}^{\infty} f_{k,m}(t) u^t w^m$. It is also convenient to introduce the variable $a = r - \ell$ directly measuring the hopping anisotropy, $r = (1 + a)/2$ and $\ell = (1 - a)/2$. One can then derive the following equations,

$$g_{k+1}(u, w) + 2\frac{(1 + a^2) u - 2}{(1 - a^2) u} g_k(u, w) + g_{k-1}(u, w) = -\frac{4}{(1 - a^2) u} (wp + 1 - p)^k , \tag{5}$$

with the initial and boundary conditions $g_k(0, w) = (wp + 1 - p)^k$ and $g_0(u, w) = 1/(1 - u)$.

The solution of (5) is obtained as a linear combination of the special solution $\Omega(wp+1-p)^k$ proportional to the right-hand side, and that solution of the homogeneous
equation which is regular at \( u = 0 \). The coefficient \( \Omega \) is obtained by substitution,

\[
\Omega = -\frac{4(wp + 1 - p)}{(1 - a^2)u(wp + 1 - p - \Lambda_+) (wp + 1 - p - \Lambda_-)} ,
\]

where \( \Lambda_{\pm} \) are the roots of the characteristic equation,

\[
\Lambda_{\pm} = 2 - \left(1 + a^2\right)u \pm 2\sqrt{(1 - u)(1 - a^2)u} .
\]

The root \( \Lambda_- \), which is nonsingular as \( u \to 0 \), gives the homogeneous solution proportional to \( \Lambda_-^k \), where the proportionality constant is determined by the boundary conditions. In summary, the solution takes the form

\[
g_k(u, w) = \left(\frac{1}{1 - u} - \Omega\right) \Lambda_-^k + \Omega(wp + 1 - p)^k .
\]

Densities of reactants at lattice sites derive from \( f_{k=1,m}(t) \). The \( m \)-dependence here follows by expanding (8) in powers of \( w \). The resulting \( u \)-dependence is complicated. Therefore we will keep the time-dependence in the generating-function form. Our explicit time-dependent expressions will be derived as asymptotic results valid for large times. The power series in \( u \) are then controlled by the singularity at \( u = 1 \), and analytical results can be derived by appropriate expansions. We consider the time-generating function for the quantities \( f_{1,m}(t) \) which represent the probability to find charge \( m \) at a lattice site at time \( t \),

\[
G_m(u) = \sum_{t=0}^{\infty} f_{1,m}(t) u^t = \delta_{m,0} \left[ \frac{\Lambda_-}{1 - u} - \frac{4}{(1 - a^2)u} \right] - \frac{4\Lambda_+(-p)^m}{(1 - a^2)u(1 - p - \Lambda_+)^{m+1}} .
\]

We now turn to the single-species reactions introduced earlier. Our approach follows recent work [10] and related ideas, e.g., [13]. Consider first the reaction \( A + A \to A \). In the coagulating-charge model we now regard each “charged” site as occupied by an \( A \)-particle, and each “uncharged” site as empty of \( A \)-particles. The dynamics of the coagulating charges then maps onto the dynamics of the reaction \( A + A \to A \). The
quantity $f_{1,0}(t)$ gives the density of empty sites in both models. Therefore, the particle density (per lattice site), $c(t)$, in the aggregation model, is given by $c(t) = 1 - f_{1,0}(t)$, where $c(0) = p$. The generating function follows from (9),

$$E(u) = \sum_{t=0}^{\infty} c(t)u^t = \frac{1}{1 - u} - G_0(u) = \frac{1 - \Lambda_-}{1 - u} + \frac{4(1 - p)}{(1 - a^2)u(1 - p - \Lambda_+)} . \quad (10)$$

The function $E(u)$ is regular at $u = 0$; the Taylor series is controlled by the singularity at $u = 1$,

$$E(u) = \frac{2}{\sqrt{1 - a^2}} \left[ \frac{1}{\sqrt{1 - u}} + \mathcal{O}(1) \right] . \quad (11)$$

This yields the leading-order large-time behavior,

$$c(t) \approx \frac{2}{\sqrt{(1 - a^2) \pi t}} . \quad (12)$$

We are not aware of other exact solutions for this model with anisotropic hopping. However, the leading-order large time behavior is expected to be universal in that it does not depend on the initial density $p$. Furthermore, the particle diffusion constant $D(a) = (1 - a^2)D(0)$ decreases proportional to $1 - a^2$ when the anisotropy is introduced. Therefore, as a function of $D(a)t$, the result (12) does not depend on the anisotropy and in fact it is the same as expressions found for other $A + A \rightarrow A$ models, with different detailed dynamical rules [3].

For the reaction $A + A \rightarrow \emptyset$, the appropriate mapping is to identify odd charges with particles $A$ and even charges with empty sites [10]. The generating function is obtained as follows,

$$E(u) = \sum_{j=0}^{\infty} G_{2j+1}(u) = \frac{4\Lambda_+ p}{(1 - a^2)u[(1 - p - \Lambda_+)^2 - p^2]} . \quad (13)$$

The large-time behavior is similar to the aggregation reaction, with the universal expression which is less than (12) by a factor of 2.
The finite-time results for both models do depend on details of the dynamical rules. For our particular choice of synchronous dynamics, there exists an exact relation [10] which holds also for the anisotropic case, checked by comparing the generating functions,

$$2c_\emptyset(t; p) = c_A(t; 2p).$$  \hspace{1cm} (14)

Here the subscripts denote the outcome of the reaction while the added argument stands for the initial density.

For the $AB$ model, we assume that initially particles are placed with density $p$, but now a fraction $\alpha$ of them are type $A$, and a fraction $\beta = 1 - \alpha$ are type $B$. The concentration difference is constant during the reaction; it remains $(\alpha - \beta)p$. At large times, this is also the limiting value of the density of the majority species, while the density of the minority species vanishes. In what follows we assume $\alpha \geq \beta$ without loss of generality; $c(t)$ will refer to the density of the majority species $A$.

The dynamics of the $AB$ model can be related to that of the coagulating-charge model by adapting the ideas of [7]. The dynamics of the “sticky” $A + B \rightarrow \emptyset$ model can be viewed as coagulation. Thus, we consider the $AB$ particles as new charges, +1 for $A$, and $-1$ for $B$. If the net charge of a coagulated cluster is positive than we regard it as a group of $A$ particles (equal in their number to the charge value). If the charge is negative, we consider the cluster $B$-particle, while if the charge is 0, we regard this cluster as nonexistent ($\emptyset$).

The probability of having an $m$-particle cluster in the original positive-charge-only model was given by $f_{1,m}(t)$. Each such cluster can have charge $n = -m, -m+2, \ldots, m-2, m$, where we now refer to the new, $\pm$ charge definition. The key observation is that having a “species” label assigned to a particle at time $t = 0$ is statistically independent of its motion and coagulation as part of clusters at later times. The density (per site)
of $m$-particle clusters with exactly $n$ units of charge can be calculated as follows,

$$\Psi_{m,n}(t) = \alpha^{m+n} \beta^{m-n} \frac{m!}{(m+n)! (m-n)!} f_{1,m}(t). \quad (15)$$

The concentration of $A$-particles, i.e., the density per site of the $+$ charge, can be written as

$$c(t) = \sum_{n=1}^{\infty} n \left[ \sum_{m=n,n+2, \ldots} \Psi_{m,n}(t) \right]. \quad \text{After some algebra, we get the generating function,}$$

$$E(u) = \frac{4\Lambda_+}{(1-a^2)u(p+\Lambda_+ - 1)} \left( x \frac{\partial}{\partial x} - y \frac{\partial}{\partial y} \right) S(x, y). \quad (16)$$

Here we introduced the function

$$S(x, y) = \sum_{n=1}^{\infty} \sum_{j=0}^{\infty} x^{n+j} y^j \binom{n+2j}{j} = \frac{2x}{\sqrt{1-4xy} \left( 1 - 2x + \sqrt{1-4xy} \right)}, \quad (17)$$

and the variables $x = p\alpha / (p + \Lambda_+ - 1)$, $y = p\beta / (p + \Lambda_+ - 1)$. The evaluation of the double-sum is quite nontrivial; see [8] for details.

It is useful to introduce the parameter $b = \alpha - \beta \geq 0$ which measures the excess of $A$ at time $t = 0$,

$$\alpha = (1+b)/2 \quad \text{and} \quad \beta = (1-b)/2. \quad (18)$$

For the equal concentration case, $b = 0$, the large-time behavior is governed by the singularity at $u = 1$,

$$E(u) = \frac{1}{(1-u)^{3/4}} \left[ \frac{\sqrt{p}}{2 (1-a^2)^{1/4}} - \frac{1-p}{4 \sqrt{p} (1-a^2)^{3/4}}(1-u)^{1/2} + O(1-u) \right]. \quad (19)$$

The leading-order behavior of the density follows from the first term,

$$c(t) \approx \frac{\sqrt{p}}{2\Gamma(3/4) (1-a^2)^{1/4} t^{1/4}}. \quad (20)$$

The most significant feature of this result is that, similar to the single-species reactions, the anisotropy, $a$, dependence can be fully absorbed in the diffusion constant, in terms of $D(a)t = (1-a^2) D(0)t$. The exponent $1/4$ was derived in [7] for different (isotropic) dynamical rules.
An expansion for fixed \( b > 0 \) yields
\[
E(u) = \frac{bp}{1-u} + \frac{1-b^2}{(1-a^2)b^3p} - \frac{2}{(1-a^2)^{3/2}} \frac{(1-b^2)(2-b^2p)}{b^5p^2} \sqrt{1-u} + \mathcal{O}(1-u) .
\] (21)
The leading term corresponds to the constant contribution \( c(t) = bp + \ldots \) which is expected for the majority species. In fact, expansions near \( u = 1 \) are nonuniform in the limits \( b \to 0^+ \) and \( b \to 0^- \); here we used for the first time the fact that the majority species is \( A \). The approach to the constant asymptotic density is given by the third term,
\[
c(t) - bp \approx \frac{(1-b^2)(2-b^2p)}{\sqrt{\pi} b^5p^2 (1-a^2)^{3/2} t^{3/2}} .
\] (22)
This difference is just the density of the minority species \( B \). As before, the anisotropy dependence of this leading-order power-law correction is fully absorbed in the diffusion rate, while the exponent is consistent with the results of [7].

It is of interest to explore the nonuniform behavior near \( b = 0 \) within the crossover scaling formulation. The appropriate scaling combination turns out to be proportional to \( b/(1-u)^{1/4} \), as determined by inspection of various limiting expressions. It proves convenient to absorb certain constants into the precise definition of the scaling combination \( \sigma \),
\[
\sigma = \sqrt{p} (1-a^2)^{1/4} b/(1-u)^{1/4} .
\] (23)
In the double-limit \( b \to 0 \) and \( u \to 1^- \), taken with fixed values of \( \sigma \), we obtain the scaling relation
\[
E(u) \approx p^{-1} (1-a^2)^{-1} b^{-3} R(\sigma) ,
\] (24)
where the scaling function \( R \), analytic at \( \sigma = 0 \), can be derived exactly,
\[
R(\sigma) = \frac{\sigma^3 (\sigma + \sqrt{4 + \sigma^2})^2}{4\sqrt{4 + \sigma^2}} .
\] (25)
For \( \sigma \ll 1 \) the following expansion applies, \( R(\sigma) = \frac{1}{2} \sigma^3 + \frac{1}{2} \sigma^4 + \mathcal{O}(\sigma^5) \). The leading term here reproduces the first term in (19). The latter was the limiting form for \( u \to 1 \)
at $b = 0$. The second term in (19), however, is not of the form $\sim b^{-3}\sigma^4$. Corrections to the leading scaling behavior correspond to this term in the $b = 0$ expansion (19).

In the opposite limit, $\sigma \to +\infty$, we get the expansion $R(\sigma) = \sigma^4 + 1 - 4\sigma^{-2} + \mathcal{O}(\sigma^{-4})$. The leading term here reproduces the first term in (21); the limit $\sigma \to +\infty$ corresponds to $u \to 1$ at fixed small positive $b$. Interestingly, the next two terms in (21) are also reproduced in their small-$b$ form by the next two terms here. The second term yields $1/\left[(1 - a^2) b^3 p \right]$ in $E(u)$. The third term in (21) is reproduced with the numerator 4 which is the small-$b$ limiting value.

The scaling description provides a uniform limiting approximation in the double-limit $b \to 0$ and $u \to 1$. Specifically, the region of nonuniform behavior near $b = 0$ is exploded by the large factor $\sim (1 - u)^{-1/4}$. In terms of $\sigma$, the behavior is smooth and well defined. For instance, the result (25) applies equally well for $\sigma < 0$ which corresponds to $A$ becoming the minority species. The limit of $u \to 1^-$ at small fixed $b < 0$ is described by the limit $\sigma \to -\infty$. The appropriate expansion takes the form $R(\sigma) = -1 + 4\sigma^{-2} + \mathcal{O}(\sigma^{-4})$, similar in structure to the $\sigma \to +\infty$ expansion but without the constant-density first term.

In summary, our exact results for the leading-order large-time particle densities of reaction-diffusion models in 1D show expected universal power-law behaviors. Anisotropy of hopping has no effect on the universality class of the models studied; it can be absorbed in the diffusion constant. Finite-time results are more sensitive [6] to the value of the anisotropy parameter $a$; they are cumbersome to derive and of less interest than the leading-order expressions. One interesting exception is the relation (14) which holds in our synchronous-dynamics models.
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