Inter-Particle Distribution Functions for One-Species

Diffusion-Limited Annihilation, $A + A \rightarrow 0$

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**ABSTRACT:** Diffusion-limited annihilation, $A + A \rightarrow 0$, and coalescence, $A + A \rightarrow A$, may both be exactly analyzed in one dimension. While the concentrations of $A$ particles in the two processes bear a simple relation, the inter-particle distribution functions (IPDF) exhibit remarkable differences. However, the IPDF is known exactly only for the coalescence process. We obtain the IPDF for the annihilation process, based on the Glauber spin approach and assuming that the IPDF’s of nearest-particle pairs are statistically independent. This assumption is supported by computer simulations. Our analysis sheds further light on the relationship between the annihilation and the coalescence models.

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1. Introduction

One-species annihilation, $A + A \rightarrow 0$, and coalescence, $A + A \rightarrow A$, have been extensively studied as basic prototypes of diffusion-limited reactions [1-21]. In one dimension, these simple models yield themselves to exact analysis: exact results for coalescence have been obtained, for example, with the method of inter-particle distribution functions (IPDF) [12-18], and annihilation has been analyzed exactly through a Glauber spin formalism [19-21] (and by several other techniques).

The distribution of distances between nearest particles, i.e., the IPDF, plays a fundamental role in the Smoluchowski theory of diffusion-limited reactions and it has been studied extensively for various reaction models [21-30]. In the case of one-species coalescence in one dimension the IPDF is obtained exactly, as a byproduct of the IPDF method [15]. For one-species annihilation in one dimension, several aspects of the kinetics are known exactly, including the time dependence of the concentration of particles, but not the IPDF.

In this paper, we compute the IPDF for the annihilation process from the (exactly known) spin-spin correlation function of the Glauber spin formalism. Our derivation is based on the assumption that the IPDF’s of different particle pairs are statistically independent. Although this may seem a strong assumption, comparison to computer simulations data suggests that it may be exact. At the very least, our approach yields a remarkably good analytic approximation for the IPDF of the one-species annihilation model.

The rest of this paper is organized as follows. In the next section we briefly summarize the exact results known for the annihilation and the coalescence processes. We also discuss the similarities between the IPDF method and the Glauber spin formalism. In Section 3, we develop a relation between the spin-spin correlation function in the Glauber method and the IPDF of the annihilation process. In Section 4, we apply this technique to the computation of the IPDF for annihilation, and for annihilation with input, and compare to results from numerical simulations. In the discussion, in Section 5, we reexamine the
conjecture of statistical independence. We demonstrate the need for proof by showing that the conjecture is wrong in the case of the closely related model of coalescence.

2. Coalescence and Annihilation

Our models are defined on a one-dimensional lattice with lattice spacing $\Delta x$. Each lattice site can be either empty, or occupied with exactly one particle. Particles hop to the nearest site to their right or left at rate $D/(\Delta x)^2$. On long length and time scales this yields normal diffusion with diffusion coefficient $D$. When a particle hops onto a site which is already occupied, a reaction takes place: in the case of coalescence the impinging particle is removed, modeling the reaction $A + A \rightarrow A$, and in the case of annihilation both particles (the hopping particle and the target) are removed, modeling $A + A \rightarrow 0$.

Coalescence has been analyzed exactly by the IPDF method (also known as the method of empty intervals) [see ref. 15 for a review]. In this approach, one defines $E_n(t)$ as the probability that an arbitrary sequence of $n$ consecutive sites is empty at time $t$. From the $E_n$ one can compute various quantities of interest. For example, the concentration of particles is given by

$$c(t) = \frac{1 - E_1(t)}{\Delta x}. \quad (1)$$

The $E_n$ satisfy a simple, closed master equation which can be solved exactly not only for the basic coalescence model, but also for a number of variations, including the back reaction, $A \rightarrow A + A$, input of $A$ particles, inhomogeneous initial distributions, and finite-size lattices [11-18].

Although the analysis can be carried out in discrete form, it is much simpler (and more transparent) to consider the continuum limit. This is done by defining $x = n\Delta x$ and then letting $\Delta x \rightarrow 0$. The empty interval probabilities are replaced by the two-variable functions, $E(x, t)$, and the concentration, for example, becomes

$$c(t) = -\frac{\partial E(x, t)}{\partial x} \bigg|_{x=0}. \quad (2)$$
The IPDF $p(x,t)$, \textit{i.e.}, the probability that the nearest particle to an arbitrary particle is a distance $x$ away at time $t$, can also be obtained from $E(x,t)$:

$$c(t)p(x,t) = \frac{\partial^2 E(x,t)}{\partial x^2}. \quad (3)$$

Throughout the remainder of this paper we will confine ourselves to the continuum limit.

In the case of simple coalescence the equation of motion for the empty interval probability is

$$\frac{\partial E}{\partial t} = 2D \frac{\partial^2 E}{\partial x^2}, \quad (4)$$

with boundary conditions

$$E(0,t) = 1 \quad \text{and} \quad E(\infty,t) = 0. \quad (5)$$

For generic homogeneous initial conditions (excluding exotic fractal distributions of the particles [11]) the system arrives at a universal long time asymptotic regime, where

$$c(t) \to \frac{1}{\sqrt{2\pi Dt}}, \quad \text{as} \quad t \to \infty, \quad (6)$$

and

$$p(x,t) \to \frac{x}{4Dt} \exp(-\frac{x^2}{8Dt}), \quad \text{as} \quad t \to \infty. \quad (7)$$

The IPDF can be put in scaling form in terms of the dimensionless interparticle distance $\xi = c(t)x$:

$$p(\xi,t) = c(t)p(x,t) \to \frac{\pi}{2} \xi \exp(-\frac{\pi \xi^2}{2}), \quad \text{as} \quad t \to \infty. \quad (8)$$

Notice that in the long time asymptotic limit it becomes stationary.

The annihilation model is most easily analyzed in terms of its dual Glauber spin process (in the zero temperature limit) [19,21]. In the latter, each lattice site is occupied by a ‘spin’ variable which assumes one of two states, $\sigma = \pm 1$. Any spin adjacent to a spin of opposite state may change its state, at rate $D/(\Delta x)^2$. If we associate a particle with each pair of anti-parallel spins \textit{i.e.,} the particles are at the boundaries between alternating
domains of parallel spins, Fig. 1), these particles are seen to diffuse and annihilate upon encounter, exactly as in the annihilation model.

Consider the spin-spin correlation function, $G(x', x'', t) = \langle \sigma(x', t)\sigma(x'', t) \rangle$, where the angular brackets denote an average over different realizations of the process. For our purpose it is sufficient to limit the discussion to homogeneous initial distributions of ‘particles’, in which case $G(x', x'', t)$ is a function of $x = x'' - x'$. The correlation function, $G(x, t)$, satisfies the diffusion equation [19,21]

$$ \frac{\partial G}{\partial t} = 2D \frac{\partial^2 G}{\partial x^2}, \tag{9} $$

with boundary conditions

$$ G(0, t) = 1 \quad \text{and} \quad G(\infty, t) = 0. \tag{10} $$

The concentration of particles is obtained from the correlation function, as

$$ c(t) = -\frac{1}{2} \frac{\partial G(x, t)}{\partial x} \bigg|_{x=0}. \tag{11} $$

Notice that $G$ satisfies the same equation as $E$, and for generic initial conditions it attains the same asymptotic limit as $E$. It follows that the concentration of particles for annihilation is exactly one half of the concentration in the coalescence process (in the long time asymptotic limit). In fact, the two processes maintain a similar simple relation at all times [3].

Another approach to the annihilation model relies on $P_e(x, t)$, the probability that there is an even number of particles in an arbitrary interval of length $x$ at time $t$ [20]. $P_e$ bears a simple relation to the spin-spin correlation function: $G = 2P_e - 1$, and hence it too satisfies Eq. (9), but with $P_e(\infty, t) = 1/2$. The advantage of this approach is in that it deals with the particles directly and it does not require the dual process with spins, but it is essentially the same.

In spite of these exact analyses, there is no obvious way to determine the IPDF for the annihilation process. Computer simulations reveal that the long distance tail of
the IPDF falls off exponentially, as $e^{-x}$ [15,21]. This is also supported by non-rigorous theoretical arguments [15]. In this respect the annihilation process is radically different from coalescence (compare Eq. 8). In the following, we develop a method for calculating the IPDF in the annihilation process.

3. Relation between IPDF and Correlation Functions

For the annihilation process, the IPDF may be related to the spin-spin correlation function by assuming that at any particular time the particles (or the spin flips) constitute a renewal process on the line [31]. The IPDF is the renewal probability density. For simplicity, we shall omit the time variable, since it plays no role in the spatial renewal process of the particles.

Let $p_\parallel(x)$ be the probability that two spins separated by a distance $x$ are parallel. Then, the spin-spin correlation function is simply

$$G(x) = p_\parallel - (1 - p_\parallel) = 2p_\parallel(x) - 1. \tag{12}$$

Two spins would be parallel if there are no spin flips in between them, or if there is an even number of spin flips in between them. (Indeed, $p_\parallel$ is identical to $P_e$.) Let $p_0(x)$ be the probability that the first flip from a given spin occurs at a distance $x$ from it (the forward waiting distribution function). $p_0$ is related to the renewal probability distribution (the IPDF, $p$) through [32]:

$$p_0(x) = \int_x^\infty c x' p(x') \frac{dx'}{x'} = c \int_x^\infty p(x') dx', \tag{13}$$

where $c = 1/ \int_0^\infty xp(x) dx$ is a normalization constant which happens to be equal to the concentration of particles. The probability that there are no spin flips up to a distance $x$ from a given spin is

$$p_0'(x) = \int_x^\infty p_0(x') dx'. \tag{14}$$
(Notice that $p_0'$ is identical to the empty interval probability $E$, defined for the coalescence process.) Finally, let $p_1(x)$ be the probability that there are no spin flips up to a distance $x$ from a given spin flip:

$$p_1(x) = \int_x^\infty p(x') \, dx' = \frac{p_0(x)}{c}. \tag{15}$$

We can now express $p_\parallel$ as the sum of $p_0'$, and $p_0$ convoluted with $p$ an odd number of times (this yields an even number of spin flips) and convoluted with $p_1$. Working with the Laplace transform of the distribution functions (which we shall denote by a tilde), the convolutions assume the simpler form of products:

$$\tilde{p}_\parallel = \tilde{p}_0' + \tilde{p}_0 (\tilde{p} + \tilde{p}^3 + \cdots) \tilde{p}_1 = \frac{1}{s} - \frac{c}{s^2} \frac{1 - \tilde{p}}{1 + \tilde{p}}. \tag{16}$$

Here $s$ is the Laplace transform variable (conjugate to $x$), and the last equality has been obtained using Eqs. (13)-(15) and standard properties of the Laplace transform. From Eq. (12), we finally obtain the relation between the spin-spin correlation function and the IPDF:

$$\tilde{p}(s) = \frac{1 - h(s)}{1 + h(s)}; \quad h(s) \equiv \frac{s}{2c} (1 - s\tilde{G}(s)). \tag{17}$$

Assuming statistical independence of consecutive IPDF’s one can also derive an expression for the density-density correlation function $c^{(2)}(x,t)$, i.e., the joint probability of finding any two particles separated by a distance $x$ at time $t$. Because there can be any integer number of particles between two arbitrary particles, the density-density correlation function is given by a sum over convolutions of the IPDF. In terms of the Laplace transform:

$$\frac{1}{c} \tilde{c}^{(2)} = \tilde{p} + \tilde{p}^2 + \tilde{p}^3 + \cdots = \frac{\tilde{p}}{1 - \tilde{p}} = \frac{1 - h(s)}{2h(s)}. \tag{18}$$

4. IPDF’s for the Annihilation Process

Let us now apply the above formalism to annihilation in the long time asymptotic limit. In this case, for generic initial conditions the correlation function approaches a universal
form:

\[ G(x, t) = \text{erfc} \left( \frac{x}{2\sqrt{2Dt}} \right) \quad \text{as} \; t \to \infty, \tag{19} \]

and the concentration reaches the asymptotic limit

\[ c(t) = \frac{1}{2\sqrt{2\pi Dt}}, \quad \text{as} \; t \to \infty. \tag{20} \]

With these expressions, we get from Eq. (17),

\[ \tilde{p} = \frac{1 - \sqrt{\pi} ks e^{k^2 s^2} \text{erfc}(ks)}{1 + \sqrt{\pi} ks e^{k^2 s^2} \text{erfc}(ks)}, \tag{21} \]

where \( k = \sqrt{2Dt} \). This can be inverted either numerically or through series expansions.

For small \( x \) we find (in scaling form, with \( \xi = cx \))

\[ \frac{1}{c} p = \pi \xi - \frac{5}{6} \pi^2 \xi^3 + \frac{49}{120} \pi^3 \xi^5 + \cdots \quad (\xi \ll 1). \tag{22} \]

The linear term has been derived exactly by Amar and Family [21]. For large \( x \) we get the asymptotic form (by contour integration)

\[ \frac{1}{c} p \sim \frac{4\sqrt{\pi} a}{4a^2 + 1} e^{-2\sqrt{\pi}a\xi}, \quad (\xi \gg 1), \tag{23} \]

where \( a = -z_0 \approx 0.357835 \) is the absolute value of the smallest negative root of \( 1 + \sqrt{\pi} z \exp(z^2) \text{erfc}(z) = 0 \). Thus, we do confirm the large distance exponential decay of the IPDF.

We have performed a similar analysis for the transient regime at early times. Here, a discrete formulation is more appropriate, but that is derived in a straightforward, analogous way to the continuum limit of Section 3 [31]. Some typical results are shown in Figs. 2(a) and (b). In Fig. 2(c) we compare simulation results at a very late time, well after the long time asymptotic limit sets in, to the analytical IPDF obtained from Eq. (21). There is perfect agreement between theory and simulations, to the level of the statistical noise. Notice, however, that it is difficult to obtain good statistics in the long time asymptotic limit, due to the small number of surviving particles.
We next analyze annihilation with a steady homogeneous input of particles at rate $R$ per unit time per unit length. This model has the advantage of a stationary state, making it possible to obtain exceptionally good statistics for the IPDF from simulations. In this case, the spin-spin correlation function satisfies the equation \[ \frac{\partial G}{\partial t} = 2D \frac{\partial^2 G}{\partial x^2} - 2RG, \] with the stationary solution \[ G(x) = \frac{\text{Ai}(r^{1/3}x)}{\text{Ai}(0)}, \] \[ (r \equiv R/D). \] The stationary concentration is \[ c = \frac{|\text{Ai}'(0)|}{2\text{Ai}(0)} r^{1/3}, \] where the prime denotes differentiation of the Airy function.

In Fig. 3 we compare simulation results to the analytic IPDF obtained from Eqs. (17), (25) and (26). Again, the agreement is excellent. The slight discrepancy at the peak of the curve may be a result of discreteness. Indeed, this discrepancy becomes smaller for smaller steady state concentrations. For short distances, we get \( (1/c)p(\xi) = 4(\text{Ai}(0)/|\text{Ai}'(0)|)^3\xi. \). For long distances, we find an exponential decay of the IPDF tail, similar to pure annihilation. This is interesting, in view of the fact that the spin-spin correlation function decays as $\exp(-x^2)$ for pure annihilation, but as $\exp(-x^{3/2})$ in the case of input.

5. Discussion

Our analytical approach would be exact if the IPDF’s for different particle pairs in the annihilation process were statistically independent. Only then could the convolutions leading to Eqs. (17) and (18) be justified. Therefore, the excellent agreement between simulations and the analytical computations leads us to the following conjecture:

*The distribution functions of the inter-particle distances of different particle pairs in the annihilation process are statistically independent from each other.*
We assume, of course, that at time \( t = 0 \) the initial distribution of particles is such that IPDF’s are statistically independent. If such is the case, our results suggest that the statistical independence would be maintained at all times.

It is not obvious why this conjecture should be true. In fact, we now show that in the coalescence process the IPDF’s of different particle pairs are statistically dependent. The density-density correlation function for the coalescence process has been obtained exactly [33, 34]:

\[
c^{(2)}(\xi) = c^2 \left[ 1 - e^{-\frac{\pi}{2} \xi^2} + \frac{\pi}{2} e^{-\frac{\pi}{4} \xi^2} \text{erfc}(\frac{\sqrt{\pi}}{2} \xi) \right].
\]  

(27)

On the other hand, we can compute \( c^{(2)} \) assuming statistical independence, from Eqs. (8) and (18). In spite of a rough qualitative agreement, there are important discrepancies: for example, the exact result of Eq. (27) shows that \( c^{(2)} \) approaches the long distance limit as \( c^2 - c^{(2)} \sim \exp(-\frac{\pi}{2} \xi^2) \), monotonously, while statistical independence predicts an exponential approach with oscillations, \( c^2 - c^{(2)} \sim \exp(-a \xi) \cos(b \xi + \phi) \), \( (a, b, and \phi \) are known constants [34]).

This poses the following puzzle. Consider diffusion-limited polymerization of \( n \)-mers, \( A_n \). The polymers diffuse on the line, with a diffusion constant independent of their size, and polymerize upon encounter: \( A_i + A_j \rightarrow A_{i+j} \). Initially, all particles are monomers, \( A_1 \). This process codes for coalescence and for annihilation simultaneously [10]: If we disregard size, and consider all polymers as equivalent particles, the process is analogous to coalescence, \( A + A \rightarrow A \). On the other hand, if we focus only on polymers of an odd number of monomers, the process is equivalent to annihilation, \( A + A \rightarrow 0 \). Imagine now that we start this “master-process” and we look at a snapshot of the system at some later time. There are polymers of odd and even size, roughly in equal amounts, and apparently well mixed (there is no segregation into clusters of odd and even sized polymers). From our foregoing discussion we conclude that the distances between any pair of nearest polymers are statistically dependent (we are looking at coalescence). But if our conjecture is true, the distances between nearest polymers of odd size (annihilation)—a finite subset of all
polymers—are statistically independent! There is of course no contradiction, however, the spatial distribution of the polymers in the master-process must then be rather peculiar.

In conclusion, we have presented strong numerical evidence that the distribution functions of distances between nearest particle pairs in the annihilation process, and in the annihilation process with input, are statistically independent. If we accept this as true, then the IPDF for the annihilation model has been computed exactly. Our computations reproduce the exact short distance limit of Amar and Family [21]. We also confirm the exponential decay of the IPDF tail. The IPDF for the case of annihilation with input is similar to that of pure annihilation, but the depletion zone near the origin is narrower. This is the result of the input, which tends to bring the IPDF closer to a pure exponential, because of its random nature.

Our work suggests a most significant difference between annihilation and coalescence: the IPDF’s are statistically independent for the former, but not for the latter. An interesting result is that the density-density correlation function exhibits oscillations in the case of annihilation, but not for coalescence. Such oscillations are typical of fluids with hard core repulsion interactions, and are expected because of the effective repulsion between particles due to the reactions. Their absence in the case of coalescence is a striking consequence of the correlations between IPDF’s.

The excellent agreement between our analytical derivations and simulations suggests that our approach is exact. We have not yet been able to prove the necessary conjecture of statistical independence (for the annihilation model). This remains an open problem.

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Figure 1: Relation between the Glauber spin model and the annihilation process. The $A$ particles (bottom) correspond to the spin flips, or domain boundaries, in the spin process (top).

Figure 2: Comparison of simulated IPDF’s to analytical derivations for the annihilation process, $A + A \rightarrow 0$. Simulation results are shown in histogram form for (a) $t=1$, (b) $t = 10$, and (c) $t = 1000$. All simulations began with a homogeneous density of particles, $1/2$.

Figure 3: Comparison of the simulated IPDF (circles) to the analytical derivation (solid curve) for the annihilation process with input. The input rate ($R = 0.0001$) is low enough so that the continuum limit applies ($c \approx 0.0213 \ll 1$). Simulation results represent an average over $16 \times 10^6$ time units (performed after the steady state was reached).