



Diffusion-limited reaction in one dimension

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Abstract

Reaction rates that are controlled by collisions between diffusing particles depend on the distribution of distances between particles as well as on the concentration of particles. Classical treatment of reaction rates thus produces a hierarchy of particle correlation functions. However, when collision between particles results in annihilation, it is possible to sidestep this hierarchy and find an exact solution for the mean number of particles per unit length as a function of time. This article is concerned with the case, in one space dimension, where new particles are born or “nucleated”, two at a time, at random times and positions, with a fixed distance b between the particles at birth. We review an exact method for calculating the mean lifetime of a particle and an exact rate equation in terms of the correlation function. In addition, the distribution of particle lifetimes is calculated under a “constant-killing-rate” approximation that compares favourably with the results of numerical experiments.

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1. Introduction

Rates of reaction are determined by two factors: the frequency with which reactants meet, and the time taken for the reaction to occur once the reactants have met [1]. In the “well-stirred” limit, the rate of reaction is determined entirely by the second factor. Here we examine the opposite case, where the reaction is instantaneous once two reactants meet.

We suppose the reactants are identical diffusing point particles in one dimension and the rate of reaction is the frequency of collisions between them. New particles are born or “nucleated”, two at a time, at random times and positions, with a fixed distance b between the particles at birth. Once born, particles follow Brownian paths until they meet another particle, whereupon both particles involved in the collision are annihilated. The three parameters of the model are

- (1) the nucleation rate Γ , the number of pairs created per unit length per unit time;
- (2) the separation b of pairs of particles at nucleation;
- (3) the diffusivity of a particle D .

We consider either infinite domains or finite-length domains with periodic boundaries, so that there is no preferred location.

A steady state density (mean number of particles per unit length) is attained at late times, resulting from the competition between nucleation of new particles and annihilation of existing ones. The dynamic balance between nucleation and annihilation is illustrated in Fig. 1.

We denote the mean number of particles per unit length, also called the density or concentration of particles, at time t by $\rho(t)$. (A numerical estimate of $\rho(t)$ is constructed simply by counting the number of particles in a long domain at time t and dividing by the length of the domain.) Because particles react in pairs, the standard chemical rate equation would read

$$\dot{\rho} = 2\Gamma - k\rho^2, \quad (1)$$

for some constant k . However, it is easy to see that this differential equation cannot correctly describe even the simple case $\Gamma = 0$ because there is no way to produce a constant k with the correct dimensions [2–5] from the one remaining parameter, D . Moreover, solution of (1) with $\Gamma = 0$ yields $\rho(t) \propto t^{-1}$ as $t \rightarrow \infty$, while numerical and analytical results show that $\rho(t) \propto (Dt)^{-1/2}$ as $t \rightarrow \infty$. For example, if the initial distribution of particles is random then [6]

$$\rho(t) = \rho(0) \exp(8Dt\rho^2(0)) \operatorname{erfc}(\rho(0)(8Dt)^{1/2}). \quad (2)$$

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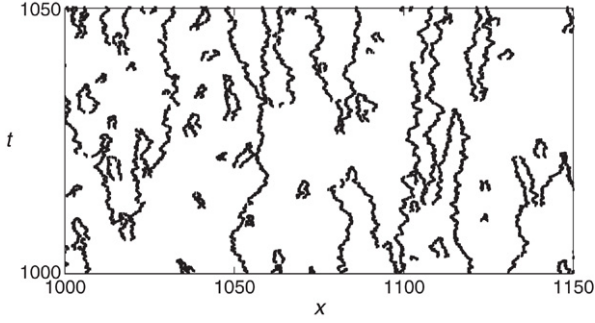


Fig. 1. A spacetime diagram of one part of a numerical solution. Time increases upwards and each dot indicates the position of a diffusing particle. $\Gamma = 0.01$, $b = 2$, $D = 0.5$.

In fact, no closed rate equation can capture the time evolution of the density because the correlations between particles must be taken into account.

In Section 2 we summarize an exact analytical calculation of the density of particles, introduced in [7] and [8]. Section 3 is concerned with the correlation function. In Section 4 we consider the *distribution* of particle lifetimes under a simplifying approximation.

2. Density of particles: Exact results

Let $\mathbf{N}_1(x_1, x_2, t)$ be the number of particles in the interval (x_1, x_2) at time t . For any choice of $x_1 \in \mathbb{R}$, $x_2 \in \mathbb{R}$ and $t \in \mathbb{R}^+$, with $x_1 \leq x_2$, this quantity is a random variable. We define the function $f_1(x, t)$ via

$$\langle \mathbf{N}_1(x_1, x_2, t) \rangle = \int_{x_1}^{x_2} f_1(x, t) dx, \quad (3)$$

where angled brackets indicate the mean over realizations. If $f_1(x, 0) = \rho(0)$, independent of x , then we say that the ensemble of initial conditions is translation invariant. Since the dynamics has no mechanism for breaking translation invariance, a consequence of this assumption is that $f_1(x, t) = \rho(t)$ for all $t \geq 0$, *i.e.* the density of particles is a function of time only. Any one realization will be spatially inhomogeneous, of course, but there are no preferred locations on average. Examples of translation-invariant ensembles of initial conditions are

- (i) no particles present at $t = 0$;
- (ii) a random initial distribution of particles.

Let the function $r(x, t)$ be defined for $x, t \geq 0$ by [7–9]

$$r(x, t) = \{\text{probability that the number of particles between } X \text{ and } X + x \text{ at time } t \text{ is even}\}. \quad (4)$$

Because of translational invariance, the choice of the reference location X is irrelevant. For any $t > 0$, as $x \rightarrow \infty$ an interval of length x is equally likely to contain an odd as an even number of particles. Thus $\lim_{x \rightarrow \infty} r(x, t) = \frac{1}{2}$.

If the distribution of particles is such that, as $\Delta x \rightarrow 0$,

$$r(\Delta x, t) = 1 - \rho(t)\Delta x + O(\Delta x^2), \quad (5)$$

then $\rho(t)$ is related to $r(x, t)$ by [7]

$$\rho(t) = - \lim_{x \rightarrow 0^+} \frac{\partial}{\partial x} r(x, t). \quad (6)$$

To calculate the evolution of $r(x, t)$, we need to consider diffusion of particles into, or out of, the interval $(X, X + x)$, and nucleation events that produce a single new particle in the interval. However, annihilation does not appear explicitly because two particles are destroyed in each annihilation event. The function $r(x, t)$ satisfies the partial differential equation [7]

$$\frac{\partial}{\partial t} r(x, t) = \begin{cases} 2D \frac{\partial^2}{\partial x^2} r(x, t) + 2x\Gamma(1 - 2r(x, t)) & x \leq b; \\ 2D \frac{\partial^2}{\partial x^2} r(x, t) + 2b\Gamma(1 - 2r(x, t)) & x > b, \end{cases} \quad (7)$$

with the boundary conditions

$$r(0, t) = 1 \quad \text{and} \quad \lim_{x \rightarrow \infty} r(x, t) = \frac{1}{2}, \quad t > 0. \quad (8)$$

The solution of (7), and hence an exact expression for $\rho(t)$ [7], can be written in terms of Airy functions Ai and Bi . The steady state solution of (7), satisfying

$$\frac{\partial}{\partial t} r(x, t) = 0,$$

is

$$r_e(x) = \begin{cases} \frac{1}{2} \left[c_1 \text{Ai} \left(\left(\frac{2\Gamma}{D} \right)^{\frac{1}{3}} x \right) + c_2 \text{Bi} \left(\left(\frac{2\Gamma}{D} \right)^{\frac{1}{3}} x \right) + 1 \right] & x \leq b; \\ \frac{1}{2} \left[c_3 \exp \left(- \left(\frac{2\Gamma b}{D} \right)^{\frac{1}{3}} x \right) + 1 \right] & x > b. \end{cases} \quad (9)$$

The constants c_1 , c_2 and c_3 are fixed by requiring $r_e(0) = 1$ and imposing continuity of $r_e(x)$ and $\frac{d}{dx} r_e(x)$ at $x = b$.

Let $\rho_e = \lim_{t \rightarrow \infty} \rho(t)$. Then¹

$$\begin{aligned} \rho_e &= -\frac{1}{2} \left(\frac{2\Gamma}{D} \right)^{\frac{1}{3}} (c_1 \text{Ai}'(0) + c_2 \text{Bi}'(0)) \\ &= \left(\frac{\Gamma}{4D} \right)^{\frac{1}{3}} \frac{|\text{Ai}'(0)|}{\text{Ai}(0)} \left(\frac{\text{Bi}'(\varepsilon) + \sqrt{3}\text{Ai}'(\varepsilon) + \varepsilon^{1/2}(\text{Bi}(\varepsilon) + \sqrt{3}\text{Ai}(\varepsilon))}{\text{Bi}'(\varepsilon) - \sqrt{3}\text{Ai}'(\varepsilon) + \varepsilon^{1/2}(\text{Bi}(\varepsilon) - \sqrt{3}\text{Ai}(\varepsilon))} \right), \end{aligned} \quad (10)$$

where the dimensionless quantity ε is defined by

$$\varepsilon = \left(\frac{2\Gamma}{D} \right)^{\frac{1}{3}} b. \quad (11)$$

In the limit $\varepsilon \rightarrow 0$, $\text{Bi}(\varepsilon) \rightarrow \sqrt{3}\text{Ai}(\varepsilon)$ and $\text{Bi}'(\varepsilon) \rightarrow -\sqrt{3}\text{Ai}'(\varepsilon)$, so

$$\begin{aligned} \rho_e &= \left(\frac{\Gamma}{4D} \right)^{\frac{1}{3}} \left(\varepsilon^{\frac{1}{2}} - \frac{1}{2}\varepsilon^2 + \dots \right) \\ &= \left(\frac{b\Gamma}{2D} \right)^{\frac{1}{2}} (1 + O(\varepsilon^{\frac{3}{2}})). \end{aligned} \quad (12)$$

¹ $\text{Ai}(0) = \text{Bi}(0)/\sqrt{3} = 3^{-2/3}\Gamma(2/3)$ and $\text{Ai}'(0) = \text{Bi}'(0)/\sqrt{3} = -3^{-1/3}\Gamma(1/3)$ [10].

This corresponds to the limit where the separation of newly nucleated pairs is much smaller than the typical distance between particles. In the opposite (large b) limit, $\varepsilon \rightarrow \infty$ and

$$\rho_e \rightarrow \frac{1}{2} \left(\frac{2\Gamma}{D} \right)^{\frac{1}{3}} \frac{|\text{Ai}'(0)|}{\text{Ai}(0)}. \quad (13)$$

Thus when the separation between nucleation pairs is large, the steady state density is proportional to the cube of the nucleation rate.

Use of the function $r(x, t)$ yields exact predictions that are useful for verification of numerical schemes, but gives no information about the distribution of distances between particles or about the distribution of particle lifetimes.

3. Correlations between particles

The distribution of distances between particles is measured by the correlation function $g(y, t)$. Under our assumption that particles diffuse and react instantaneously on contact, we derive an exact rate equation in terms of $g(y, t)$.

Consider two disjoint intervals (x_1, x_2) and (x_3, x_4) . The mean value of the product of the numbers of particles the intervals contain at time t can be written as

$$\langle \mathbf{N}(x_1, x_2, t) \mathbf{N}(x_3, x_4, t) \rangle = \int_{x_1}^{x_2} \int_{x_3}^{x_4} f_2(x, x', t) dx dx'. \quad (14)$$

With the usual assumption of translation invariance, the function $f_2(x, x + y, t)$ is independent of x and we define

$$g(y, t) = \rho(t)^{-2} f_2(x, x + y, t). \quad (15)$$

An equivalent definition is that, given that a particle is at x at time t ,

$$g(y, t) = \lim_{\Delta x \rightarrow 0} (\rho(t) \Delta x)^{-1} \text{P}[\text{another particle is in } (x + y, x + y + \Delta x) \text{ at time } t].$$

In numerical solutions, an estimate for $g(y, t)$ is constructed by selecting test particles and recording the number of particles as a function of distance from them.

Let $s(y, \Delta t)$ be the probability that two particles, diffusing independently with diffusivity D and initial separation y , collide before Δt . Then [11,12]

$$s(y, \Delta t) = \text{erfc} \left(\frac{y}{(8D\Delta t)^{\frac{1}{2}}} \right). \quad (16)$$

An exact expression for $g(y, t)$ is not available for any $t > 0$. Nevertheless, assuming that we may expand $g(y, t)$ as a Taylor series about $y = 0$ and using (16), the decrease in the density of particles between t and $t + \Delta t$ due to annihilation is given by

$$\begin{aligned} \int_0^\infty g(y, t) s(y, \Delta t) dy &= \int_0^\infty s(y, \Delta t) \left(g(0, t) + yg'(0^+, t) \right. \\ &\quad \left. + \frac{1}{2}y^2g''(0^+, t) + \dots \right) dy \\ &= \left(\frac{1}{\sqrt{\pi}} (8D\Delta t)^{\frac{1}{2}} g(0, t) + \frac{1}{4} (8D\Delta t) g'(0^+, t) + \dots \right). \end{aligned}$$

Let us now examine the chemical rate equation as typically written [13,7]:

$$\dot{\rho}(t) = 2\Gamma - 2\rho^2(t) \int_0^\infty g(y, t) K(y) dy, \quad (17)$$

where

$$K(y) = \lim_{\Delta t \rightarrow 0} \frac{1}{\Delta t} s(y, \Delta t).$$

The differential equation (17) is therefore well defined for all t such that

$$\lim_{\Delta t \rightarrow 0} \left(\Delta t^{-1} \int_0^\infty g(y, t) s(y, \Delta t) dy \right) = \text{finite}. \quad (18)$$

That is, consistency between (16) and (17) requires [7]

$$\lim_{y \rightarrow 0} g(y, t) = 0. \quad (19)$$

The exact rate equation is then

$$\dot{\rho} = 2\Gamma - 4D\rho^2 g'(0^+, t). \quad (20)$$

The “reaction kernel” $K(y)$ needs to be interpreted as a generalized function, similar to the derivative of $\delta(y)$, the Dirac delta function. Since $g(y)$ is only defined for $y \geq 0$, we use the following expression:

$$K(y) = 2D \lim_{a \rightarrow 0^+} \frac{\delta(y - a)}{a}. \quad (21)$$

The relation (19) implies an exclusion effect: particles are less likely to be found near any given particle than far away from it.

One important situation where (19) is not satisfied is a random distribution of particles, where $g(y, t) = 1$ for all y . Initial conditions of this type produce a $t = 0$ singularity, corresponding to rapid annihilation of particles that happen to be close to their neighbours; (19) then holds for $t > 0$.

Let $g_e(y) = \lim_{t \rightarrow \infty} g(y, t)$. Then, for systems such as ours where there are no infinite-range correlations,

$$\lim_{y \rightarrow \infty} g_e(y) = 1. \quad (22)$$

An exact solution for $g_e(y)$ for all y is not available. We have three pieces of information: (19), (22) and, from (20),

$$g_e'(0) = \frac{\Gamma}{2D} \rho_e^{-2}, \quad (23)$$

where ρ_e is the steady state density of particles. The following form is consistent with (22) and (23) and is a good approximation to the stationary correlation function observed numerically as $\varepsilon \rightarrow 0$ [7]:

$$g_e(y) = \begin{cases} y/b & y < b \\ 1 & y \geq b. \end{cases}$$

4. Distribution of lifetimes

The steady state density of particles is related to the mean lifetime of a particle in the steady state, τ , by

$$\rho_e = 2\Gamma\tau. \quad (24)$$

Because ρ_e is given by (10), τ is known exactly. The mean particle lifetime as $\varepsilon \rightarrow 0$ is thus

$$\tau = \frac{\rho_e}{2\Gamma} \rightarrow \left(\frac{b}{8D\Gamma}\right)^{1/2} = \frac{b}{4D\rho_e}. \quad (25)$$

An intuitive way to understand the scaling $\tau \propto \rho_e^{-1}$ was introduced by Büttiker and Christie [14]. Consider the fate of a Brownian particle that starts at $x = b$ in a domain with absorbing boundaries at $x = 0$ and $x = L$. The mean time to absorption is [15]

$$\tau_L = \frac{bL}{2D}. \quad (26)$$

If we choose $L = \rho_e^{-1}$ and recall that the distance between two independent Brownian particles is itself a Brownian motion with twice the diffusivity of a single particle, we regain (25).

Consideration of the various possible particle histories, from nucleation to annihilation, reveals that the distribution of particle lifetimes is not a simple function. Each particle is born with a unique partner, a distance b away at birth. The timeline of a particle ends when it collides with that of its partner (“recombination”) or with that of another particle (“non-recombinant annihilation”) [16]. Fig. 2 is a schematic diagram of the different types of annihilation processes, with recombination shown on the left.

In the steady state, the lifetime of a particle is a random variable \mathbf{t} with mean, $\langle \mathbf{t} \rangle$, given by τ . In order to understand the distribution of lifetimes, we can assign one realization of each of two random variables, \mathbf{t}_r and \mathbf{t}_μ , to each nucleated pair of particles, where

- \mathbf{t}_r is the time at which two particles performing Brownian motion with diffusivity D collide, given that their initial separation is b ;
- \mathbf{t}_μ is the first time that a path belonging to a member of the pair is hit by another particle.

The pair recombines if $\mathbf{t}_r < \mathbf{t}_\mu$. One member is annihilated in a collision with a different particle if $\mathbf{t}_\mu < \mathbf{t}_r$.

The distribution of \mathbf{t}_r can be calculated exactly by considering the diffusion of a nucleated pair, as if no other particles were present. The separation between the two particles performs Brownian motion with diffusivity $2D$ and initial condition b . Let the density of \mathbf{t}_r be denoted $R_{\mathbf{t}_r}(t) = \frac{d}{dt}P[\mathbf{t}_r < t]$. Then [11,17]

$$R_{\mathbf{t}_r}(t) = \frac{d}{dt} \operatorname{erf}\left(\frac{b}{8Dt}\right) = bt^{-\frac{3}{2}}(8\pi D)^{-\frac{1}{2}} \exp\left(-\frac{b^2}{8Dt}\right). \quad (27)$$

The mean lifetime of a particle, given that its timeline ends in recombinant annihilation, is denoted $\langle \mathbf{t}_r \mid \mathbf{t}_r < \mathbf{t}_\mu \rangle$.

Non-recombinant annihilation ($\mathbf{t}_r > \mathbf{t}_\mu$) leaves behind a “survivor” particle, one of the two paired particles before the collision, that lives for a (further) time \mathbf{t}_s . See Fig. 2. The mean lifetime of a particle, given that its timeline does not end in recombinant annihilation, is $\langle \mathbf{t}_\mu \mid \mathbf{t}_\mu < \mathbf{t}_r \rangle + \frac{1}{2} \langle \mathbf{t}_s \rangle$. The total mean lifetime of a particle is thus

$$\tau = P[\mathbf{t}_r < \mathbf{t}_\mu] \langle \mathbf{t}_r \mid \mathbf{t}_r < \mathbf{t}_\mu \rangle + (1 - P[\mathbf{t}_r < \mathbf{t}_\mu])$$

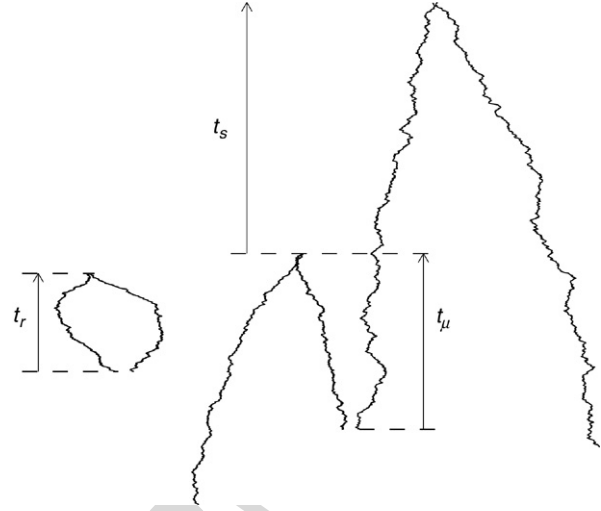


Fig. 2. Schematic spacetime diagram, with time increasing upwards, showing the times \mathbf{t}_r , \mathbf{t}_μ and \mathbf{t}_s . Recombination is shown on the left. The non-recombinant annihilation event shown in the centre of the diagram leaves behind a “survivor” particle that lives for a time \mathbf{t}_s longer than its partner.

$$\times \left(\langle \mathbf{t}_\mu \mid \mathbf{t}_\mu < \mathbf{t}_r \rangle + \frac{1}{2} \langle \mathbf{t}_s \rangle \right). \quad (28)$$

The distribution of particle lifetimes is given by

$$P[\mathbf{t} > t] = P[\mathbf{t}_r > t, \mathbf{t}_\mu > t] + \frac{1}{2} P[\mathbf{t}_\mu < \mathbf{t}_r, \mathbf{t}_\mu < t, \mathbf{t}_\mu + \mathbf{t}_s > t]. \quad (29)$$

4.1. The constant-killing-rate approximation

We shall construct an approximate expression for the density of particle lifetimes using the following approximation, first introduced in [16]. In the steady state, there is a constant probability per unit time, μ of a member of the pair being struck and “killed” by different particle. That is,

$$P[\mathbf{t}_\mu > t] = e^{-\mu t} \quad (30)$$

and \mathbf{t}_μ and \mathbf{t}_r are independent. We may expect this approximation to be a good one at least for long lifetimes, since the probability per unit time of encountering another particle can be expected to reach a steady state, after sufficient time has elapsed from nucleation.

Under the constant-killing-rate assumption,

$$P[\mathbf{t}_\mu > \mathbf{t}_r] = \int_0^\infty e^{-\mu t} R_{\mathbf{t}_r}(t) dt = e^{-\nu b}, \quad (31)$$

where

$$\nu^2 = \frac{\mu}{2D}. \quad (32)$$

The mean recombination time is then given by

$$\langle \mathbf{t}_r \mid \mathbf{t}_r < \mathbf{t}_\mu \rangle = e^{\nu b} \int_0^\infty t R_{\mathbf{t}_r}(t) dt = \frac{1}{2} \frac{b}{\sqrt{2\mu D}}. \quad (33)$$

(This quantity was printed incorrectly in [16].)

The mean time for non-recombinant annihilation events, that remove one member of a pair, is

$$\begin{aligned} \langle \mathbf{t}_\mu \mid \mathbf{t}_\mu < \mathbf{t}_r \rangle &= (1 - e^{-vb})^{-1} \int_0^\infty t \mu e^{-\mu t} \operatorname{erf} \left(\frac{b}{\sqrt{8Dt}} \right) dt \\ &= \frac{1}{\mu} \left(1 - \frac{vb}{2} \frac{1}{e^{vb} - 1} \right). \end{aligned} \quad (34)$$

The mean time between the birth of a pair of particles and the death of at least one member is

$$\tau_p = e^{-vb} \langle \mathbf{t}_r \mid \mathbf{t}_r < \mathbf{t}_\mu \rangle + (1 - e^{-vb}) \langle \mathbf{t}_\mu \mid \mathbf{t}_\mu < \mathbf{t}_r \rangle. \quad (35)$$

To complete the specification of the random times, we assume that a survivor particle has the same constant probability per unit time, μ , of being killed. That is $P[\mathbf{t}_s > t] = e^{-\mu t}$. The mean lifetime of a particle in the “constant-killing-rate” approximation is thus given by

$$\begin{aligned} \tau &= \tau_p + \frac{1}{2}(1 - e^{-vb}) \frac{1}{\mu} \\ &= \frac{1}{2} \frac{b}{\sqrt{2\mu D}} + (1 - e^{-vb}) \frac{1}{\mu} \left(\frac{3}{2} - \frac{vb}{2} \frac{1}{e^{vb} - 1} \right). \end{aligned} \quad (36)$$

The distribution of lifetimes, under the same approximation, is

$$\begin{aligned} P[\mathbf{t} > t] &= e^{-\mu t} P[\mathbf{t}_r > t] + \int_0^t \frac{1}{2} \mu e^{-\mu s} P[\mathbf{t}_r > s] e^{-\mu(t-s)} ds \\ &= e^{-\mu t} \operatorname{erf} \left(\frac{b}{\sqrt{8Dt}} \right) \\ &\quad + \frac{1}{2} \mu e^{-\mu t} \int_0^t \operatorname{erf} \left(\frac{b}{\sqrt{8Ds}} \right) ds. \end{aligned} \quad (37)$$

We now turn to the task of estimating the killing rate μ , which depends on D and ρ_e . From dimensional considerations, $\mu \propto \rho_e^2 D$. We write

$$\mu = \frac{1}{2} \alpha^2 \rho_e^2 D, \quad (38)$$

where α is a dimensionless constant. As $vb \rightarrow 0$, the mean lifetime (36) has the limit $\tau \rightarrow \frac{3}{2} \frac{b}{\alpha \rho_e D}$. To obtain agreement with (25), we therefore take $\alpha = 6$.

In Fig. 3 we plot a numerical histogram of lifetimes versus $R(t) = -\frac{d}{dt} P[\mathbf{t} > t]$, using the approximation (37) with $\alpha = 6$. The agreement is remarkably good, except for short lifetimes. The latter observation is consistent with the hypothesis that the probability per unit time that one of a pair encounters another particle is highest soon after nucleation. The constant-killing-rate assumption therefore underestimates the probability density at short lifetimes.

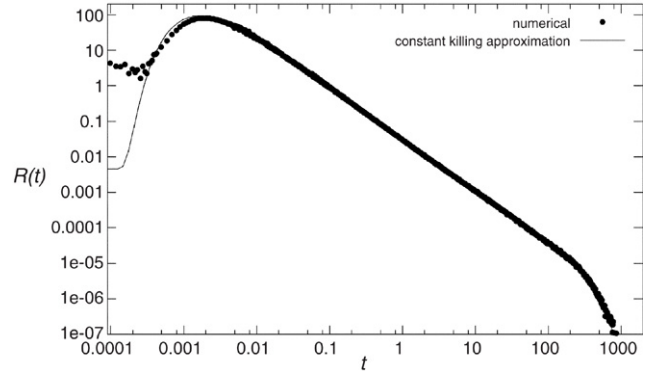


Fig. 3. Probability density of particle lifetimes (log–log scale). The dots are numerical results from direct simulation of the particle system. The line is obtained from (37), which uses the constant-killing-rate approximation. The parameters were $b = 0.1$, $D = 0.5$ and $\Gamma = 0.01$.

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