Fluctuation-dominated $A+B\rightarrow 0$ kinetics under short-ranged interparticle interactions

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In the present paper we analyze the kinetics of irreversible $A+B\rightarrow 0$ reactions in situations appropriate to electrolyte solutions. We consider diffusing species which experience short-range repulsive (attractive) interactions between like (unlike) particles and we highlight the role of fluctuations in the particles' spatial distributions. We focus mainly on one-dimensional systems and devise a many-particle description of the reaction kinetics. In terms of our analytical approach we show that at intermediate times the interplay between fluctuations and short-ranged interactions gives rise to unusual behaviors, characterized by novel dynamical exponents of the particles' mean concentrations. For batch reactions at longer times, when the concentrations drop off significantly, the usual fluctuation-induced behavior is recovered. We also analyze systems with external, steady sources of particles and we show that in such situations the effects of the interactions become decisive at long times. Our analytical findings are in good agreement with the results of numerical simulations, which we also present. © 1996 American Institute of Physics. [S0021-9606(96)50636-7]

I. INTRODUCTION

The subject of reaction kinetics under diffusion limited conditions is an old topic,[1,2] which has recently experienced increased interest following the remarkable discovery of important many-particle effects.[3–10] Such effects are due to the fluctuations in the spatial distributions of reactants and show essential departures from conventionally expected behaviors. The most pronounced deviations from the predictions of the standard approaches[1,2] were found for the irreversible recombination reaction

$$A+B\rightarrow 0. \qquad (1)$$

Reaction (1) involves particles A and B which diffuse and react on contact. It was recognized that the usual Gaussian fluctuations in the initial spatial distribution of the reactants have a strong impact on the long-time kinetics and that they slow down the decay. For equal mean initial concentrations $C_A(0)=C_B(0)$ the reaction follows at long times the law[3–10]

$$C_A(t)=C_B(t)\propto C^{1/2}(0)(Dt)^{-d/4} \quad \text{for } d\leq 4, \qquad (2)$$

an expression which can be contrasted to the conventional Smoluchowski-type forms[1,2]

$$C_A(t)=C_B(t)\propto \frac{1}{K_{\text{eff}}t} \qquad \text{for } d=3, \qquad (3a)$$

$$C_A(t)=C_B(t)\propto \frac{\log(Dt)}{Dt} \qquad \text{for } d=2, \qquad (3b)$$

and

$$C_A(t)=C_B(t)\propto \frac{1}{\sqrt{Dt}} \qquad \text{for } d=1. \qquad (3c)$$

In Eqs. (2)–(3c) d is the spatial dimension of the reactive system, D denotes the diffusion coefficient of the A and the B particles (for simplicity taken to be the same), and K_{eff} is an effective rate constant,[2] depending both on D and on the reaction probability.

Remarkably, the behavior of Eq. (2) is due mainly to statistical aspects. We note that as time progresses, diffusion smoothes out inhomogeneities on scales which grow with time as $l_D \propto \sqrt{Dt}$. The number of unreacted particles at time t can be estimated as the excess in the number of particles in regions of size l_D assuming that all other particles in the volume l_D^d reacted pairwise. For Gaussian distributions this yields as mean concentration $C(t) \propto \sqrt{C(0)l_D^d}/l_D^d$, i.e., Eq. (2).[3–10] These illustrative arguments are also confirmed by a more rigorous approach, based on correlation functions.[3] The long-time behavior of $C(t)$ is controlled by the decay of the correlation function $G_{AB}(t)$ (*vide infra*, Sec. II)[3]

$$C(t) = \sqrt{-G_{AB}(t)}. \tag{4}$$

On the other hand, the evolution of $G_{AB}(t)$ depends only on the difference in local concentrations, which changes only due to diffusion. Under our reaction conditions it follows that[3]

$$G_{AB}(t) \propto -\frac{C(0)}{(Dt)^{d/2}}, \tag{5}$$

from which again Eq. (2) follows.

The remarkable result of Eq. (2) was first obtained in Refs. 3–6, which was reproduced by means of elaborated analytical and numerical techniques in Refs. 7–13 and was proven rigorously in Refs. 14 and 15. Subsequent works focused on more general situations, by including stoichiometric and nonstoichiometric, or geminate initial conditions,[16–21] the possibility of the backward reaction[17,22–24] and side reactions,[25,26] different diffusion coefficients for different species,[27–29] and the influence of external stirring,[30–32] they also examined the recombination kinetics taking place in non-Euclidean systems (fractals).[8,10,16,33–39] Considerable progress was also gained in the understanding of the spatial aspects, involving correlation functions, cluster sizes, and cluster boundaries,[9,40–43] which are also amenable to experimental analysis.[44] More details are given in recent review papers.[45–47]

In the last few years the interest shifted to systems in which the reaction is accompanied by a steady inflow of reactants. The salient feature here is that under certain conditions the reaction never reaches a steady state, in contrast to the classical descriptions,[2] which predict an exponential approach to a steady state concentration

$$C_A(t=\infty) = C_B(t=\infty) \propto \sqrt{\frac{I}{K}}. \tag{6}$$

Here I is the average intensity of the source of reactants and K is the reaction rate. In particular, it was shown[48–52] (see also Refs. 33, 34 and 53, 54 for more details) that in low-dimensional systems the concentrations increase in time following

$$C_A(t) = C_B(t) \propto t^{1/4} \quad \text{for} \quad d = 1, \tag{7}$$

and

$$C_A(t) = C_B(t) \propto \sqrt{\log(t)} \quad \text{for} \quad d = 2, \tag{8}$$

when there is an uncorrelated, steady flow of particles into the system. The case $d = 3$ is marginal and $C_A(t) = C_B(t)$ ultimately approach a steady state. However, even for $d = 3$ the situation is peculiar: First, the relaxation to the steady state obeys a power-law dependence,[17,34,54] in place of the exponential law predicted by the classical theories[2] and, second, the steady state concentration also differs from the classical prediction. Moreover, we note that the approach of the concentrations to zero after switching off the source is also peculiar and shows unusual memory effects.[55] We note finally that Eqs. (7) and (8) can be derived from correlation

functions and related quantities; the same holds also for Eq. (4) in the presence of an external source, when one takes into account the time dependence of $G_{AB}(t)$.

Therefore, in systems with a steady inflow, an ongoing accumulation of particles takes place in low-dimensional systems. However, the accumulation is very sensitive to the way in which the A and B particles are injected into the system; if they are introduced in a correlated manner, say as geminate pairs, Eqs. (4) and (5) no longer hold.[17,23,34,55]

The results reported so far pertain to an idealized situation in which all interactions between the particles (apart from the reaction act) are neglected. Now in typical realizations of the reaction scheme of Eq. (1), such as, for instance, for electron–hole and for radical recombination the A and the B particles interact with each other. There are several types of interactions,[56] the most pronounced being the long-ranged ones of Coulombic origin. Other examples are multipolar interactions and also dispersion (London's) interactions, which are important for particles which carry no net charge. We note that in solutions the long-ranged interactions tend to be screened; the effective forces are then short ranged. Such short-ranged interactions influence significantly the reaction kinetics. In the presence of an inflow of particles the growth of concentrations also differs from Eqs. (7) and (8) even at long times.[57–59]

In the present paper we devise a systematic many-particle description of the kinetics of recombination reactions involving diffusing particles with short-ranged repulsive (attractive) interactions between like (unlike) species. We start with a discrete-space picture and formulate the master equations for the time evolution of the local particle concentrations. The short-ranged interactions between particles are included into the transition rates, while the reaction is accounted for through a nonlinear term. Under the assumption that the spatial fluctuations are Gaussian, we derive closed equations for the concentrations and pair-correlation functions.

Here we focus on equal A and B concentrations and mainly (because of numerical ease) on reaction kinetics in $1d$. As usual, we consider two different ways by which the particles enter the system:

(a) All reactants are introduced into the system at time $t = 0$ at random positions.

(b) The particles are introduced steadily, at all times, into the system.

Furthermore we also address the question of reaction kinetics in higher dimensions. We confirm our analytical results by numerical simulations, which we also discuss. This paper is structured as follows: In Sec. II we display the model and the general equations for the concentrations and for the pair-correlation functions. In Sec. III we derive the solutions of these equations for case (a) (no steady source). In Sec. IV we address case (b), reaction kinetics in the presence of a steady source of particles. Section V presents the results of our numerical simulations. Finally, in Sec. VI we conclude with a summary and discussion.

II. THE MODEL AND THE BASIC EQUATIONS

We formulate the model on a one-dimensional chain with lattice spacing a. Particles of types A and B are introduced at $t=0$ at random positions, so that their respective concentrations are $C(0)$. Both types of particles migrate on the lattice. As soon as two particles of different kind meet at a lattice site they can react with rate constant K, in which case the reaction product is immediately removed from the system. The reaction constant K depends on the chemical properties of the A and the B molecules. In the following we suppose that K is known and that it is large (i.e., there is a low probability that meeting particles do not react); this allows us to focus on the statistical aspects, rather than on the chemical features of the model.

We denote the local concentrations at site x by $C_A(x)$ and $C_B(x)$. Their evolution may be described in the continuous-time limit by the following (approximate) master equations:

$$\dot{C}_A(x) = -KC_A(x)C_B(x) - C_A(x)[F_A(x,x+a) + F_A(x,x-a)] + C_A(x-a)F_A(x-a,x) \times C_A(x+a)F_A(x+a,x) \tag{9}$$

and

$$\dot{C}_B(x) = -KC_A(x)C_B(x) - C_B(x)[F_B(x,x+a) + F_B(x,x-a)] + C_B(x-a)F_B(x-a,x) + C_B(x+a)F_B(x+a,x), \tag{10}$$

where the overdot denotes the time derivative. Here the first terms on the right-hand side of Eqs. (9) and (10) describe the loss of A and B particles due to the reaction. The remaining terms describe the random motion; the functions $F_{A(B)}(x,y)$ denote the rate at which an $A(B)$ particle jumps from site x to the neighboring site y. For the $F_{A(B)}(x,y)$ we adopt the following forms:

$$F_A(x,y) = \frac{1}{2\tau} + \alpha\{[C_A(x) - C_A(y)] + [C_B(y) - C_B(x)]\} \tag{11}$$

and

$$F_B(x,y) = \frac{1}{2\tau} + \alpha\{[C_B(x) - C_B(y)] + [C_A(y) - C_A(x)]\}, \tag{12}$$

where τ is the mean time spent by a particle at a given lattice site and α is an interaction parameter, say the charge. Eqs. (11) and (12) mimic a situation appropriate for electrolyte solutions: all particles are subject to thermal forces [the first term in Eq. (11) or Eq. (12)]. Furthermore the particles are subjected to short-ranged electrostatic interactions: like (unlike) particles repel (attract) each other. The corresponding forces result from the charges present at the neighboring sites and at the site occupied by the particle. Now it is convenient to introduce the following finite-difference operators:

$$\nabla_+ C_A(x) = C_A(x+a) - C_A(x), \tag{13}$$

$$\nabla_- C_A(x) = C_A(x) - C_A(x-a), \tag{14}$$

and

$$\Delta C_A(x) = C_A(x+a) + C_A(x-a) - 2C_A(x), \tag{15}$$

and to rewrite Eqs. (9) and (10) in terms of the operators

$$\dot{C}_A(x) = \frac{1}{2\tau}\Delta C_A(x) + \alpha C_A(x)[\Delta C_A(x) - \Delta C_B(x)] + \alpha C_A(x-a)[-\nabla_- C_A(x) + \nabla_- C_B(x)] + \alpha C_A(x+a)[\nabla_+ C_A(x) - \nabla_+ C_B(x)] - KC_A(x)C_B(x) \tag{16}$$

and

$$\dot{C}_B(x) = \frac{1}{2\tau}\Delta C_B(x) + \alpha C_B(x)[\Delta C_B(x) - \Delta C_A(x)] + \alpha C_B(x-a)[-\nabla_- C_B(x) + \nabla_- C_A(x)] + \alpha C_B(x+a)[\nabla_+ C_B(x) - \nabla_+ C_A(x)] - KC_A(x)C_B(x). \tag{17}$$

Going now to the continuum we expand the local concentrations and their gradients to second order in the lattice spacing a,

$$C(x\pm a) = C(x) \pm a\frac{\partial C(x)}{\partial x} + \frac{a^2}{2}\frac{\partial^2 C(x)}{\partial x^2} + \cdots, \tag{18}$$

$$\nabla_+ C(x) = a\frac{\partial C(x)}{\partial x} + \frac{a^2}{2}\frac{\partial^2 C(x)}{\partial x^2} + \cdots, \tag{19}$$

$$\nabla_- C(x) = a\frac{\partial C(x)}{\partial x} - \frac{a^2}{2}\frac{\partial^2 C(x)}{\partial x^2} + \cdots, \tag{20}$$

and

$$\Delta C(x) = a^2\frac{\partial^2 C(x)}{\partial x^2} + \cdots. \tag{21}$$

Then Eqs. (16) and (17) take the form

$$\dot{C}_A(x) = D\frac{\partial^2 C_A(x)}{\partial x^2} + \mu\frac{\partial}{\partial x}\left[C_A(x)\frac{\partial}{\partial x}\{C_A(x) - C_B(x)\}\right] - KC_A(x)C_B(x) \tag{22}$$

and

$$\dot{C}_B(x) = D\frac{\partial^2 C_B(x)}{\partial x^2} + \mu\frac{\partial}{\partial x}\left[C_B(x)\frac{\partial}{\partial x}\{C_B(x) - C_A(x)\}\right] - KC_A(x)C_B(x), \tag{23}$$

where we have introduced $D=a^2/2\tau$ as the ''bare'' diffusion coefficient of a single particle in the absence of interactions and $\mu=2a^2\alpha$, which characterizes the interaction strength.

Apart from the reaction terms $KC_A(x)C_B(x)$, Eqs. (22) and (23) have the standard Fokker–Planck form under a concentration-dependent potential energy. The potential energy term follows, for example, from the expansion of the chemical potential to first order in the particles' concentrations.[58,59] The product form $KC_A(x)C_B(x)$ for the reaction [which follows here in a straightforward fashion from Eqs. (9) and (10)] is, on the other hand, in the theory of chemical reactions only an approximation, see Refs. 14, 38, 60 for a detailed discussion.

From Eqs. (22) and (23) we derive the temporal evolution of the concentrations and correlation functions. For this we first write:

$$C_A(x,t)=C(t)+\delta C_A(x,t) \tag{24}$$

and

$$C_B(x,t)=C(t)+\delta C_B(x,t), \tag{25}$$

where $C(t)$ stands for the volume average (denoted by brackets):

$$C(t)=\langle C_A(x,t)\rangle=\langle C_B(x,t)\rangle \tag{26}$$

and $\delta C_{A,B}(x,t)$ are the fluctuations.

Furthermore, we introduce the pair-correlation functions

$$G_{AA}(\lambda,t)=\langle \delta C_A(x,t)\delta C_A(x+\lambda,t)\rangle, \tag{27}$$

$$G_{BB}(\lambda,t)=\langle \delta C_B(x,t)\delta C_B(x+\lambda,t)\rangle, \tag{28}$$

and

$$G_{AB}(\lambda,t)=\langle \delta C_A(x,t)\delta C_B(x+\lambda,t)\rangle. \tag{29}$$

Taking the volume average of Eqs. (22) and (23) we find that the terms relevant to the motion of the particles cancel each other; so that the time evolution of $C(t)$ is given by a Riccati equation:

$$\dot{C}(t)=-K[C^2(t)+G_{AB}(\lambda=0,t)]. \tag{30}$$

We proceed by analyzing the time dependence of G_{AB}. For this, we follow Ref. 3, in which the system of equations for $\dot{C}_{AB}(t)$ were linearized. This corresponds to taking into account terms up to $(\delta C)^2$ in the equations for $G_{ij}(\lambda,t)$, (i and j are either A or B) which makes the equations for $G_{ij}(\lambda,t)$ closed. Such a seemingly simple approach leads to the correct fluctuation-induced forms, i.e., in the absence of interactions between particles to $C(t)\propto t^{-d/4}$. In Ref. 17 it was shown that such a linearization is, in fact, equivalent to the assumption that the $\delta C_{A,B}(x,t)$ are random Gaussian functions at all times and correspond to decoupling the fourth-order correlation functions into a product of pair correlations,

$$\langle \delta C_A(x,t)\delta C_B(x+a,t)\delta C_i(x+\lambda,t)\delta C_j(x,t)\rangle$$

$$=G_{AB}(a,t)G_{ij}(\lambda,t). \tag{31}$$

In fact, such an assumption is physically reasonable, since the system under study starts from a Gaussian initial state

and the diffusion can only smooth out inhomogeneities and does not create singularities. Linearization in this spirit leads to

$$\delta\dot{C}_A(x,t)=D\frac{\partial^2}{\partial x^2}\delta C_A(x,t)+\mu C(t)\frac{\partial^2}{\partial x^2}[\delta C_A(x,t)$$
$$-\delta C_B(x,t)]-KC(t)[\delta C_A(x,t)$$
$$+\delta C_B(x,t)] \tag{32}$$

and to

$$\delta\dot{C}_B(x,t)=D\frac{\partial^2}{\partial x^2}\delta C_B(x)+\mu C(t)\frac{\partial^2}{\partial x^2}[\delta C_B(x,t)$$
$$-\delta C_A(x,t)]-KC(t)[\delta C_A(x,t)$$
$$+\delta C_B(x,t)]. \tag{33}$$

Under the same assumptions as above the pair correlation functions obey the following (closed) system of linear equations:

$$\dot{G}_{AB}(\lambda,t)=\langle \delta\dot{C}_A(x,t)\delta C_B(x+\lambda,t)\rangle$$
$$+\langle \delta C_A(x,t)\delta\dot{C}_B(x+\lambda,t)\rangle$$
$$=2D\frac{\partial^2}{\partial\lambda^2}G_{AB}(\lambda,t)+\mu C(t)\frac{\partial^2}{\partial\lambda^2}$$
$$\times[2G_{AB}(\lambda,t)-G_{AA}(\lambda,t)-G_{BB}(\lambda,t)]$$
$$-KC(t)[2G_{AB}(\lambda,t)+G_{AA}(\lambda,t)$$
$$+G_{BB}(\lambda,t)], \tag{34}$$

$$\dot{G}_{AA}(\lambda,t)=\langle \delta\dot{C}_A(x,t)\delta C_A(x+\lambda,t)\rangle$$
$$+\langle \delta C_A(x,t)\delta\dot{C}_A(x+\lambda,t)\rangle$$
$$=2D\frac{\partial^2}{\partial\lambda^2}G_{AA}(\lambda,t)+2\mu C(t)\frac{\partial^2}{\partial\lambda^2}$$
$$\times[G_{AA}(\lambda,t)-G_{AB}(\lambda,t)]$$
$$-KC(t)[G_{AB}(\lambda,t)+G_{AA}(\lambda,t)], \tag{35}$$

and

$$\dot{G}_{BB}(\lambda,t)=\langle \delta\dot{C}_B(x,t)\delta C_B(x+\lambda,t)\rangle$$
$$+\langle \delta C_B(x,t)\delta\dot{C}_B(x+\lambda,t)\rangle$$
$$=2D\frac{\partial^2}{\partial\lambda^2}G_{BB}(\lambda,t)+2\mu C(t)\frac{\partial^2}{\partial\lambda^2}$$
$$\times[G_{BB}(\lambda,t)-G_{AB}(\lambda,t)]$$
$$-KC(t)[G_{AB}(\lambda,t)+G_{BB}(\lambda,t)]. \tag{36}$$

The system of equations for the mean concentrations and for the pair-correlation functions is now complete. From it we can derive the time evolution of these functions, provided that the initial conditions both for the mean concentrations and for the pair-correlation functions are given. In Sec. III

we consider the special case of uncorrelated initial distributions of particles, i.e., the ''batch'' situation in the terminology introduced by Kopelman.[39] We note that the important case, when reactants are in (geminate) $A-B$ pairs[17,33,34,55] can also be studied in terms of our formalism, although we will not consider it here. Finally, in Sec. IV we extend our consideration to the case of reactions in the presence of a steady inflow (source) of reactants.

III. INSTANTANEOUS GENERATION OF UNCORRELATED PARTICLES: BATCH REACTIONS

We consider now the solution to Eqs. (30) and (34)–(36) for systems in which all particles are generated at $t=0$. We take the particle distribution to be Gaussian and delta correlated with mean $C(0)$, so that[3]

$$G_{AA}(\lambda,t=0)=G_{BB}(\lambda,t=0)=C(0)\delta(\lambda) \tag{37}$$

and

$$G_{AB}(\lambda,t=0)=0, \tag{38}$$

where we took the reactions at $t=0$ into account.

To solve Eqs. (34)–(36) subject to the initial conditions Eqs. (37) and (38) we notice first that, due to symmetry

$$G_{AA}(\lambda,t)=G_{BB}(\lambda,t) \tag{39}$$

and thus Eqs. (34)–(36) reduce to

$$\dot{G}_{AB}(\lambda,t)=2D\,\frac{\partial^2}{\partial\lambda^2}\,G_{AB}(\lambda,t)$$
$$+2\mu C(t)\,\frac{\partial^2}{\partial\lambda^2}\,[G_{AB}(\lambda,t)-G_{BB}(\lambda,t)]$$
$$-2KC(t)[G_{AB}(\lambda,t)+G_{BB}(\lambda,t)] \tag{40}$$

and

$$\dot{G}_{BB}(\lambda,t)=2D\,\frac{\partial^2}{\partial\lambda^2}\,G_{BB}(\lambda,t)$$
$$+2\mu C(t)\,\frac{\partial^2}{\partial\lambda^2}\,[G_{BB}(\lambda,t)-G_{AB}(\lambda,t)]$$
$$-2KC(t)[G_{AB}(\lambda,t)+G_{BB}(\lambda,t)]. \tag{41}$$

This system of equations can be further simplified by noticing that it possesses two integrable combinations. The first is

$$S_-(\lambda,t)=G_{AB}(\lambda,t)-G_{BB}(\lambda,t), \tag{42}$$

which mirrors the fact that even locally the difference in concentrations is not affected by the reaction. The second combination is

$$S_+(\lambda,t)=G_{AB}(\lambda,t)+G_{BB}(\lambda,t). \tag{43}$$

These combinations obey

$$\dot{S}_-(\lambda,t)=(2D+4\mu C(t))\,\frac{\partial^2}{\partial\lambda^2}\,S_-(\lambda,t) \tag{44}$$

with the initial condition

$$S_-(\lambda,t=0)=-C(0)\delta(\lambda), \tag{45}$$

and

$$\dot{S}_+(\lambda,t)=2D\,\frac{\partial^2}{\partial\lambda^2}\,S_+(\lambda,t)-4KC(t)S_+(\lambda,t) \tag{46}$$

with the initial condition

$$S_+(\lambda,t=0)=C(0)\delta(\lambda). \tag{47}$$

The solution to Eqs. (44) and (45) can be found readily by noticing that the change of the time variable

$$T=2Dt+4\mu\int_0^t C(\tau)d\tau \tag{48}$$

reduces Eqs. (44) and (45) to

$$\frac{\partial}{\partial T}\,S_-(\lambda,T)=\frac{\partial^2}{\partial\lambda^2}\,S_-(\lambda,T) \tag{49}$$

with

$$S_-(\lambda,T=0)=-C(0)\delta(\lambda). \tag{50}$$

Equation (49) is a standard diffusion equation with a diffusion coefficient equal to 1. The new time variable T will be repeatedly used in what follows.

The solution of Eq. (49) thus reads

$$S_-(\lambda,T)=-\frac{C(0)}{\sqrt{2\pi T}}\exp\left(-\frac{\lambda^2}{4T}\right). \tag{51}$$

Now, the solution for $S_+(\lambda,t)$ can be obtained directly and reads as

$$S_+(\lambda,t)=\frac{C(0)}{2\sqrt{\pi Dt}}\exp\left(-4K\int_0^t C(\tau)d\tau-\frac{\lambda^2}{8Dt}\right). \tag{52}$$

Consequently, the expression $G_{AB}(\lambda=0,t)$ needed for Eq. (30) is

$$G_{AB}(\lambda=0,t)=\frac{S_-(\lambda=0,t)+S_+(\lambda=0,t)}{2}$$
$$=-\frac{C(0)}{4\sqrt{\pi(Dt+2\mu\int_0^t C(\tau)d\tau)}}+\frac{C(0)}{4\sqrt{\pi Dt}}$$
$$\times\exp\left(-4K\int_0^t C(\tau)d\tau\right). \tag{53}$$

From standard chemical kinetics for the batch reaction it is known that the decrease of $C(t)$ with time is not faster than $1/t$. Hence the integral $\int_0^t C(\tau)d\tau$ in the exponent of Eq. (52) is monotonous and diverges (sublinearly) for $t\to\infty$. It follows that the second term in Eq. (53) vanishes faster than the first term. Consequently at longer times:

$$G_{AB}(\lambda=0,t)\approx-\frac{C(0)}{4\sqrt{\pi(Dt+2\mu\int_0^t C(\tau)d\tau)}}. \tag{54}$$

Substituting Eq. (54) into Eq. (30), we thus have for the concentration C

$$\dot{C}(t) = -K\left[C^2(t) - \frac{C(0)}{4\sqrt{\pi(Dt + 2\mu\int_0^t C(\tau)d\tau)}}\right]. \quad (55)$$

The asymptotic behavior of $C(t)$ at long times depends on the decay form of $G_{AB}(\lambda=0,t)$: should $G_{AB}(\lambda=0,t)$ decay faster than t^{-1} then the second term in brackets would become irrelevant at longer times and the solution would tend to the classical kinetic form, $C(t)=1/kt$. Here, however, $G_{AB}(\lambda=0,t)$ decays slower than t^{-1}, as can be seen from Eq. (54), so that at long times, from Eq. (55) $C(t) \cong \sqrt{-G_{AB}(\lambda=0,t)}$ follows. This is exactly the form given by Eq. (4). The situation is somewhat similar to the standard case of fluctuation-dominated kinetics in low dimensions. However, here the pair-correlation function has a more complex form, due to the presence of interparticle interactions.

We note now that Eq. (54) displays two distinct regimes, corresponding to the prevalence of either the first or the second term in Eq. (54). The first regime is observed when

$$Dt \ll 2\mu\int_0^t C(\tau)d\tau. \quad (56)$$

Then $G_{AB}(\lambda=0,t)$ decreases as

$$G_{AB}(\lambda=0,t) \approx -\frac{C(0)}{4\sqrt{2\pi\mu\int_0^t C(\tau)d\tau}}, \quad (57)$$

and thus as above

$$C^2(t) = -G_{AB}(\lambda=0,t) = \frac{C(0)}{4\sqrt{2\pi\mu\int_0^t C(\tau)d\tau}}. \quad (58)$$

We now look for a solution of Eq. (58) of the form

$$C(t) = \frac{A}{t^\gamma}, \quad (59)$$

where the amplitude A of the decay function and the exponent γ, characterizing the decay, have to be determined from Eq. (58). Inserting Eq. (59) into Eq. (58) we have

$$\frac{A^2}{t^{2\gamma}} = \frac{C(0)}{4}\sqrt{\frac{1-\gamma}{2\pi\mu A t^{1-\gamma}}}, \quad (60)$$

which yields $\gamma=1/5$ and $A=C^{2/5}(0)/(40\pi\mu)^{1/5}$; for the decay we obtain accordingly

$$C(t) = \left(\frac{C(0)}{2\sqrt{10\pi\mu}}\right)^{2/5} t^{-1/5}. \quad (61)$$

From Eqs. (61) and (56) we can estimate the range of validity of this decay law; substituting Eq. (61) into the inequality in Eq. (56) we find that the decay in Eq. (61) holds in the intermediate time regime, such that

$$t \ll t_{\text{cross}} \approx 0.8\frac{C^2(0)\mu^4}{D^5}. \quad (62)$$

Now, let us turn to the large-t limit, such that the following inequality holds:

$$Dt \gg 2\mu\int_0^t C(\tau)d\tau. \quad (63)$$

In this time regime the interparticle interactions become irrelevant, the dynamics is diffusive, and the decay of the $A-B$ correlations follows the law

$$G_{AB}(\lambda=0,t) = -\frac{C(0)}{4\sqrt{\pi Dt}}. \quad (64)$$

Then Eq. (4) implies

$$C^2(t) = \frac{C(0)}{4\sqrt{\pi Dt}}, \quad (65)$$

which yields

$$C(t) = \frac{\sqrt{C(0)}}{2}(\pi Dt)^{-1/4}, \quad (66)$$

i.e., the standard fluctuation-induced decay in $d=1$. Equation (66) is not, of course, an unexpected physical result, since in this time regime the particle concentrations get very small and thus short-ranged interactions become irrelevant.

To close this section we mention that the analysis presented here can be extended to higher dimensions. Generalizing the transition rates in Eqs. (11) and (12) to d dimensions we find that the main difference is the behavior of the correlation functions. In d dimensions one has

$$G_{AB}(\lambda=0,t) \approx -\frac{C(0)}{(16\pi(Dt+2\mu\int_0^t C(\tau)d\tau))^{d/2}}. \quad (67)$$

Hence the intermediate time behavior of $C(t)$ in d-dimensions is characterized by the exponent $\gamma=d/(d+4)$, while the asymptotic, long-time behavior obeys the standard form with $\gamma=d/4$. A detailed analytical and numerical analysis of the reaction kinetics of charged particles in d-dimensional systems will be presented in a forthcoming paper.

IV. REACTION KINETICS UNDER A STEADY INFLOW OF PARTICLES

We study here the kinetics of the $A+B\rightarrow0$ reaction involving interacting particles in the presence of a steady inflow of reactants. We suppose that particles enter continuously at uncorrelated, random places the system and that the currents for A and B have an equal average intensity I. In a continuous-space and continuous-time picture the evolution of the local concentrations is given by

$$\dot{C}_A(x) = D\frac{\partial^2 C_A(x)}{\partial x^2} + \mu\frac{\partial}{\partial x}\left[C_A(x)\frac{\partial}{\partial x}\{C_A(x) - C_B(x)\}\right] - KC_A(x)C_B(x) + I_A(x,t) \quad (68)$$

and

$$\dot{C}_B(x) = D \frac{\partial^2 C_B(x)}{\partial x^2} + \mu \frac{\partial}{\partial x} \left[C_B(x) \frac{\partial}{\partial x} \{C_B(x) \right.$$

$$\left. - C_A(x)\} \right] - K C_A(x) C_B(x) + I_B(x,t), \quad (69)$$

where $I_{A,B}(x,t)$ describe the local production rates of A and B particles.

From Eqs. (68) and (69) we have for the mean concentration

$$\dot{C}(t) = -K[C^2(t) + G_{AB}(\lambda=0,t)] + I, \quad (70)$$

where I denotes the intensity of the source,

$$\langle I_A(x,t) \rangle = \langle I_B(x,t) \rangle = I. \quad (71)$$

As in Refs. 12 and 17, the linearized form of the equations for the pair correlations is

$$\dot{G}_{AB}(\lambda,t) = 2D \frac{\partial^2}{\partial \lambda^2} G_{AB}(\lambda,t)$$

$$+ 2\mu C(t) \frac{\partial^2}{\partial \lambda^2} [G_{AB}(\lambda,t) - G_{BB}(\lambda,t)]$$

$$- 2KC(t)[G_{AB}(\lambda,t) + G_{BB}(\lambda,t)] \quad (72)$$

and

$$\dot{G}_{BB}(\lambda,t) = 2D \frac{\partial^2}{\partial \lambda^2} G_{BB}(\lambda,t)$$

$$+ 2\mu C(t) \frac{\partial^2}{\partial \lambda^2} [G_{BB}(\lambda,t) - G_{AB}(\lambda,t)]$$

$$- 2KC(t)[G_{AB}(\lambda,t) + G_{BB}(\lambda,t)] + I\delta(\lambda). \quad (73)$$

Now, the combinations of Eqs. (72) and (73) obey the following relations. First,

$$\dot{S}_-(\lambda,t) = (2D + 4\mu C(t)) \frac{\partial^2}{\partial \lambda^2} S_-(\lambda,t) - I\delta(\lambda), \quad (74)$$

which has, by setting $A(x,0) \equiv 0 \equiv B(x,0)$, the initial condition

$$S_-(\lambda,t=0) = 0. \quad (75)$$

Second,

$$\dot{S}_+(\lambda,t) = 2D \frac{\partial^2}{\partial \lambda^2} S_+(\lambda,t) - 4KC(t)S_+(\lambda,t) + I\delta(\lambda) \quad (76)$$

with the initial condition

$$S_+(\lambda,t=0) = 0. \quad (77)$$

The solutions of Eqs. (74) and (75) are readily obtained through a Fourier transformation. We introduce

$$S_\pm(p,t) = \int_0^\infty S_\pm(\lambda,t)\cos(p\lambda)d\lambda \quad (78)$$

so that the inverse transform is

$$S_\pm(\lambda,t) = \frac{1}{2\pi} \int_0^\infty S_\pm(p,t)\cos(p\lambda)dp. \quad (79)$$

These functions obey

$$\dot{S}_-(p,t) = -p^2 \dot{T}(t) S_-(p,t) - I, \quad (80)$$

where $T(t)$ is as defined in Eq. (48), and with

$$S_-(p,t=0) = 0 \quad (81)$$

and furthermore

$$\dot{S}_+(p,t) = -[2Dp^2 + 4KC(t)]S_+(p,t) + I \quad (82)$$

with

$$S_+(p,t=0) = 0. \quad (83)$$

The solution to Eq. (80) is

$$S_-(p,t) = -I \int_0^t d\tau \exp\left(-p^2 \int_\tau^t d\tau' \dot{T}(\tau') \right) \quad (84)$$

and the solution to Eq. (82) reads

$$S_+(p,t) = I \int_0^t d\tau \exp\left(-2D(t-\tau)p^2 \right.$$

$$\left. -4K \int_\tau^t d\tau' C(\tau') \right). \quad (85)$$

Now taking the inverse Fourier transformation we obtain in the λ domain

$$S_-(\lambda,t) = -\frac{I}{4\sqrt{\pi}} \int_0^t d\tau \left(\int_\tau^t d\tau' \dot{T}(\tau') \right)^{-1/2}$$

$$\times \exp\left(-\frac{\lambda^2}{4\int_\tau^t d\tau' \dot{T}(\tau')} \right) \quad (86)$$

and

$$S_+(\lambda,t) = \frac{I}{4\sqrt{2\pi D}} \int_0^t \frac{d\tau}{\sqrt{t-\tau}} \exp\left(-\frac{\lambda^2}{8D(t-\tau)} \right.$$

$$\left. -4K \int_\tau^t d\tau' C(\tau') \right). \quad (87)$$

The desired quantity, $G_{AB}(\lambda=0,t)$, then follows:

$$G_{AB}(\lambda=0,t) = -\frac{I}{8\sqrt{\pi}} \int_0^t d\tau \left[\int_\tau^t d\tau' \dot{T}(\tau') \right]^{-1/2}$$

$$+ \frac{I}{8\sqrt{2\pi D}} \int_0^t \frac{d\tau}{\sqrt{t-\tau}}$$

$$\times \exp\left(-4K \int_\tau^t d\tau' C(\tau') \right). \quad (88)$$

Let us note now estimate the time behavior of the first and second terms in Eq. (88). To do this we first note that the mean concentration $C(t)$ in the presence of the source is evidently a nonvanishing function of time. Anticipating that in the limit $t \rightarrow \infty$ it is described by the dependence

$$C(t) = Bt^\beta \quad \text{with} \quad 0 \le \beta \le 1 \tag{89}$$

and substituting Eq. (89) into the first term of Eq. (88) we have

$$-\frac{I}{8\sqrt{\pi}} \int_0^t d\tau \left[\int_0^t d\tau' \dot{T}(\tau') \right]^{-1/2}$$

$$= -\frac{I}{8\sqrt{\pi}}$$

$$\times \int_0^t \frac{d\tau}{\sqrt{2D(t-\tau) + 4\mu B(t^{1+\beta} - \tau^{1+\beta})/(1+\beta)}}$$

$$\approx -\frac{I\sqrt{1+\beta}}{16\sqrt{\pi\mu B}} \int_0^t \frac{d\tau}{\sqrt{(t^{1+\beta} - \tau^{1+\beta})}}$$

$$= -\frac{I}{16\sqrt{\pi\mu B(1+\beta)}} B\left(\frac{1}{1+\beta}, \frac{1}{2}\right) t^{(1-\beta)/2}, \tag{90}$$

where B(x,y) denotes the beta function. Here the change from the first to the second line corresponds to neglecting the first term in Eq. (48) compared to the second one under the assumption that $\beta > 0$. The integration is then performed using Eq. (3.249.7) of Ref. 61. Equation (90) thus shows that the first term in Eq. (88) grows in time as $t^{(1-\beta)/2}$.

We turn now to the behavior of the second term in Eq. (88) and proceed by showing that it is irrelevant at long times and thus may be neglected. To show this we will merely bound it from above by a function which grows in time at a slower rate than $t^{(1-\beta)/2}$. Employing a classic Hölder inequality for integrals, i.e.,

$$\int_a^b |f(\tau)g(\tau)| d\tau \le \left[\int_a^b |f(\tau)|^p d\tau \right]^{1/p} \left[\int_a^b |g(\tau)|^q d\tau \right]^{1/q}, \tag{91}$$

in which p and q are arbitrary numbers such that $p > 1$, $q > 1$, and $1/p + 1/q = 1$, and choosing $f(\tau) = 1/\sqrt{t-\tau}$ and $g(\tau) = \exp(4KB\tau^{1+\beta}/(1+\beta))$, we majorize the second term in Eq. (88) by

$$\frac{I}{8\sqrt{2\pi D}} \int_0^t \frac{d\tau}{\sqrt{t-\tau}} \exp\left(-4K \int_\tau^t d\tau' C(\tau') \right)$$

$$\le \frac{I}{8\sqrt{2\pi D}} \exp\left(-\frac{4KBt^{1+\beta}}{1+\beta} \right)$$

$$\times \left[\int_0^t \frac{d\tau}{(t-\tau)^{p/2}} \right]^{1/p} \left[\int_0^t d\tau \exp\left(\frac{4KBq\tau^{1+\beta}}{1+\beta} \right) \right]^{1/q}. \tag{92}$$

The first integral on the right-hand side of the inequality in Eq. (92) converges only for $p < 2$ and thus p must be some number from the open interval]1,2[. We choose $p = 4/3$ and $q = 4$. Then the first expression in square brackets in Eq. (92) grows in time as $t^{1/4}$, while the second expression in square brackets goes as $\exp(4KBt^{1+\beta}/(1+\beta))t^{-\beta/4}$. Accordingly,

the second term in Eq. (88) is majorized by a function which grows in time at most as $t^{(1-\beta)/4}$, i.e., at a slower rate than the first term in Eq. (88).

Therefore, in the large-t limit the leading term in the pair-correlation function follows

$$G_{AB}(\lambda=0,t) \approx -F(\beta) \frac{I}{\sqrt{\mu B}} t^{(1-\beta)/2} \tag{93}$$

with the coefficient $F(\beta)$ being

$$F(\beta) = \frac{1}{16\sqrt{\pi(1+\beta)}} B\left(\frac{1}{1+\beta}, \frac{1}{2}\right).$$

Now we have to determine the parameters B and β. Substituting Eq. (93) into Eq. (70) we have

$$B\beta t^{\beta-1} = -K\left[B^2 t^{2\beta} - \frac{IF(\beta)}{\sqrt{\mu B}} t^{(1-\beta)/2} \right] + I. \tag{94}$$

We notice first that at short times the concentration of particles is expected to be small and thus neither the interactions nor the reaction between particles are significant. In this regime the concentration follows an unrestricted growth, determined merely by the production rate [the last term on the right hand side of Eq. (94)] and consequently $C(t) \approx It$. At longer times, however, the correlation function in Eq.(93) gets large. In this regime the growth of $C(t)$ obeys Eq. (4), which means that the terms in brackets in Eq. (94) compensate each other, i.e.,

$$B^2 t^{2\beta} = \frac{IF(\beta)}{\sqrt{\mu B}} t^{(1-\beta)/2}. \tag{95}$$

Equation (95) yields

$$\beta = 1/5 \tag{96}$$

and

$$B = \frac{1}{4} \left(\frac{2\Gamma(5/6)}{\Gamma(4/3)} \right)^{2/5} \left(\frac{5I^2}{6\mu} \right)^{1/5}, \tag{97}$$

where we used the explicit form for $F(\beta)$ and expressed the B function through the Γ functions. Thus we find explicitly

$$C(t) \approx \frac{1}{4} \left(\frac{2\Gamma(5/6)}{\Gamma(4/3)} \right)^{2/5} \left(\frac{5I^2 t}{6\mu} \right)^{1/5}. \tag{98}$$

Finally, we note that Eq. (98) can be extended to d-dimensional systems essentially along the same lines as in the absence of the source. Here the difference between the general, d-dimensional case and the one-dimensional situation lies in the behavior of the correlation functions. In d dimensions the leading term in $G_{AB}(t)$ grows as

$$G_{AB}(\lambda=0,t) \approx \frac{I(1+\beta)^{d/2}}{(\pi\mu B)^{d/2}} \int_0^t \frac{d\tau}{(t^{1+\beta} - \tau^{1+\beta})^{d/2}}$$

$$\approx It^{1-d(1+\beta)/2}, \tag{99}$$

which gives, from Eq. (4),

$$\beta = \frac{2-d}{d+4}. \qquad (100)$$

Thus one infers from Eq. (100) that also in the presence of short-ranged interactions the marginal dimension is $d=2$, see Eq. (8). Above $d=2$ a steady state exists, but is not a mean-field-like function of the pertinent parameters. An interesting open question is the way in which this steady state is approached at long times.

V. NUMERICAL SIMULATIONS

Let us first summarize our analytical findings. For the kinetics of recombination between interacting, diffusing particles we have predicted for the batch case the appearance of two different regimes, corresponding to an intermediate and to an asymptotic behavior. In the first regime, i.e., for times t less than a certain crossover time t_{cross}, $C(t)$ follows a $t^{-1/5}$ law, which stems from the interplay between the smoothing of the spatial fluctuations and the short-ranged interactions. At even longer times, when the concentrations get so low that the interparticle interactions become irrelevant, $C(t)$ crosses over to the standard form Eq. (2), typical for fluctuation-dominated kinetics in one-dimensional systems.

Under a steady source of particles, $C(t)$ grows algebraically in time. This growth is also characterized by several regimes. Starting from an empty system at very short times, i.e., for very small $C(t)$, one finds that $C(t)$ first grows linearly with time, since most of the particles neither interact, nor react with each other. At very long times, when the particles' density gets to be high, the interactions between particles become dominant; this leads to the growth pattern $C(t) \propto t^{1/5}$.

Now we display the results of direct numerical simulations of reactions between interacting particles in $1d$. The simulation procedure is described in detail in Refs. 58 and 59. We consider a one-dimensional lattice of length L, where L extends normally over 10^4–10^5 sites for the case of reactions under particle generation and over 10^6 sites for the case of batch reactions. In order to avoid finite-size effects the actual size of the system was taken to be $L \gg \Lambda(t_{\max})$, where $t_{\max}$ is the longest simulation time. For the batch reaction all particles are generated simultaneously and independently of each other. Under steady particle input the A and B particles are generated independently in space, so that K particles of kind A and K particles of kind B are created for each time step (hence $I=K/L$). Parallel to our analytical model, we model the diffusion of a particle in the field of the others by jumps to one of its neighboring sites. The hopping probabilities for moving from the site x to the right ($+$) and to the left ($-$) nearest-neighbor site are:

$$p_{\pm}(x) = 1/2 \pm f(x), \qquad (101)$$

where the additional term $f(x)$ depends on the configuration of the surrounding particles. In the case of a continuous input, Ref. 59, we found that all short-ranged interactions used led to the same patterns of behavior. In the present work we restricted ourselves to a truncated exponential interaction, taking
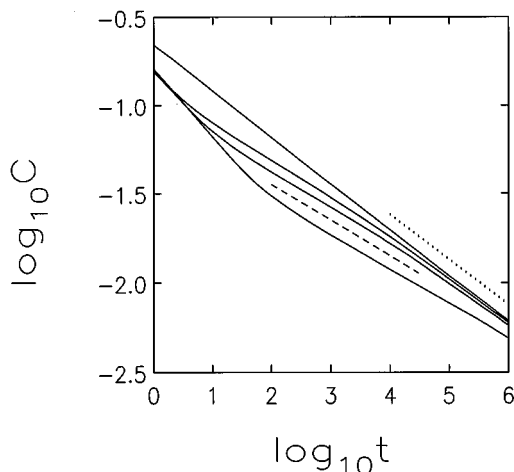

FIG. 1. The concentration decay for the case of a batch reaction in a one-dimensional system, see the text for details. The upper curve corresponds to $f_0=0$ (no interparticle interactions), the other ones to $f_0=1$ and to $\kappa=0.5$, 0.3, and 0.1, from top to bottom. Note the double-logarithmic scales. The dashed line has the slope $-1/5$, and the dotted line the slope $-1/4$.

$$f(x_i) = f_0 e_i [e_{i-1} \exp(-\kappa(x_i - x_{i-1})) - e_{i+1}$$
$$\times \exp(-\kappa(x_{i+1} - x_i))], \qquad (102)$$

where the e_i denotes the charge of the ith particle; the charges equal to $+1$ for the A and -1 for the B particles, and f_0 and κ are parameters which describe the strength and range of the interaction. Equations (101) and (102) correspond to a system which obeys Eqs. (24) and (25) with a diffusion constant of $D=1/2$ and with a tunable interaction parameter μ which depends on f_0 and κ.

This parametrization allows us to tune the simulations in such a way that the interesting asymptotic forms of behavior can be readily displayed. In the case of strong interactions and small distances Eq. (16) can lead to values of $|f|$ that
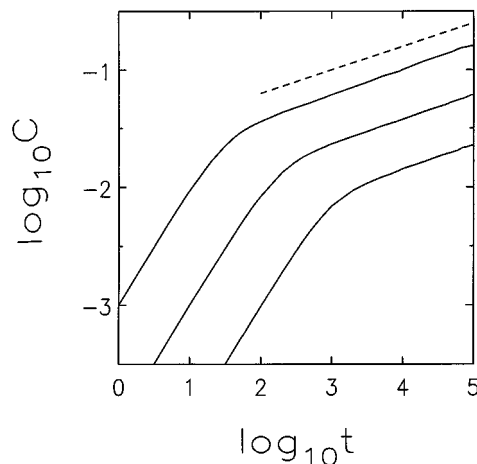

FIG. 2. The time dependence of the concentration under steady particle flow in a system with $\kappa=0.5$. The generation rates are $I=10^{-3}$, 10^{-4}, and 10^{-5}, from top to bottom. Note the double-logarithmic scales. The dashed line has the slope $1/5$.
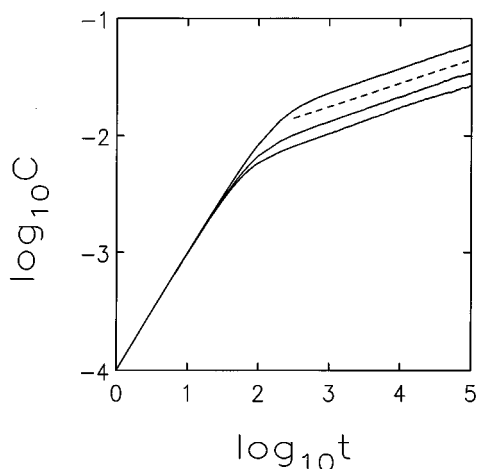
FIG. 3. Same as in Fig. 2, but now for a fixed value of $I=10^{-4}$ and different interaction parameters, $\kappa=0.5$, 0.1, and 0.05 (from top to bottom).

exceed 1/2. In this case we simply put $f(x)=\pm 1/2$: the particle moves then in the direction of the acting force with probability 1.

The numerical results for the case of batch reactions are shown in Fig. 1. The upper line corresponds to the case of noninteracting particles [where we simply set $f_0=0$ in Eq. (102)], the three lower ones correspond to interacting particles with $f_0=1$ and $\kappa=0.5$, 0.3, and 0.1. In the double-logarithmic plot of Fig. 1 the upper curve has for $t>10^4$ a slope which is very close to the theoretical prediction of $-1/4$; a least-square fit of the data gives a slope of -0.254 ± 0.005. The other three curves show, as theoretically predicted, well-developed intermediate regions described by a power-law $C(t)\propto t^{\alpha}$ with $\alpha=-0.2$. For longer times these curves approach the noninteracting curve, showing the expected crossover behavior. This crossover can be readily seen for $\kappa=0.5$ and for $\kappa=0.3$. Furthermore if the initial concentrations are low the crossover region is rather short.

The behavior for the case of the steady generation of particles was discussed in detail in Ref. 59. Here we show $C(t)$ for different generation rates and different interaction parameters. The results in Fig. 2 correspond to $\kappa=0.5$ and to the generation rates $I=10^{-3}$, 10^{-4}, and 10^{-5}. In Fig. 3 we fixed $I=10^{-4}$ and took the interaction parameter κ to be $\kappa=0.5$, 0.1, and 0.05. The initial linear regime and the long-time asymptotic behavior $C(t)\propto t^{1/5}$ can be clearly seen in Figs. 2 and 3, which therefore confirm the results of our theoretical approach. Moreover, the I dependence of the concentration C for fixed t conforms with Eq. (98) which predicts $C\propto I^{2/5}$, see Ref. 59 for more details.

VI. CONCLUSIONS

We have considered analytically the kinetics of the recombination reaction $A+B\rightarrow 0$ with A and B particles which diffuse and interact via short-ranged forces. In the present work we considered both the case of batch reactions and the case of a steady input of particles. We have shown analytically that the interplay between the spatial fluctuations and

the interactions between the particles gives rise to a large variety of temporal patterns. For the case of batch reactions in $1d$ the mean concentration follows first the dependence $C(t)\propto t^{-1/5}$, which then crosses over to the asymptotic pattern $C(t)\propto t^{-1/4}$. Under a steady input $C(t)$ grows as $t^{1/5}$ for long times. All these regimes were also found in our numerical simulation of systems in which the particles diffuse, interact via truncated exponential forces and react on contact.

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