Self-consistent model of an annihilation-diffusion reaction with long-range interactions

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We introduce coarse-grained hydrodynamic equations of motion for a diffusion-annihilation system with a power-law long-range interaction. By taking into account fluctuations of the conserved order parameter — charge density — we derive an analytically solvable approximation for the nonconserved order parameter — total particle density. Asymptotic solutions are obtained for the case of random Gaussian initial conditions and for system dimensionality $d \geqslant 2$. Large-t, intermediate-t, and small-t asymptotics were calculated and compared with existing scaling theories, exact results, and simulation data. [S1063-651X(97)02901-2]

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I. INTRODUCTION

In recent years, the annihilation-diffusion problem has generated significant interest, both theoretical and experimental. The annihilation-diffusion problem usually corresponds to that of the kinetics of particle density decay in the annihilation reaction $A+A \rightarrow \varnothing$ (one-species annihilation) and $A+B \rightarrow \varnothing$ (two-species annihilation). In the latter case, in most physical systems that one is interested in, in addition to the thermal diffusion and kinetic annihilation, particles A and B are charged and interact via a power-law long-range interaction (LRI), usually of Coulomb type, although here we will consider a more general LRI. Such physically important interaction clearly can strongly influence the annihilation dynamics by introducing an additional time scale in the annihilation process and can lead to a new mechanism for slow dynamics. The two-species annihilation reaction with the LRI can be studied in a variety of experiments. A thermal quench of a freely suspended liquid-crystal film from the smectic-A to the smectic-C phase [1,2] is one experimental system where this annihilation process has been studied in great detail. In such experiments, immediately after quench, the singularities of the smectic director (two-dimensional vector) field appear as positive and negative vortices interacting (due to elastic forces) via a logarithmic potential. As time elapses, vortices of opposite sign slowly annihilate, exhibiting complex dynamics that is clearly and strongly influenced by temperature, an initial particle distribution, and a LRI that leads to an attraction between annihilating partners. Similar annihilation problems also appear in turbulent flow, superconductivity, spinodal decomposition, and many other condensed-matter systems. The annihilation process is also relevant to coarsening of topological defects produced by a symmetry-breaking field in particle physics models, after an early temperature quench due to the fast initial universe expansion, a process that is thought in part to determine the large-scale structure of today's universe. [3]

It is well known that in classical chemical kinetics, density decay for both one-species and two-species annihilation is described by the kinetic rate equation (see, e.g., [4])

$$\frac{d\rho}{dt} = -\mathcal{K}\rho^2, \tag{1}$$

with the large-t asymptotics given by

$$\rho(t) \simeq (\mathcal{K}t)^{-1}, \tag{2}$$

where $\mathcal{K}$ is a reaction constant. Equation (1) completely neglects all spatial fluctuations and correlations of the particle density. Although the dynamics described by Eqs. (1) and (2) is correct above an upper critical space dimension d_{UC}, for $d \leqslant d_{UC}$, both diffusion and fluctuations play an important role, substantially slowing down the density decay. It was indeed shown [5] that for the one-species annihilation without a LRI the $d_{UC}=2$. That is, for systems of dimensionality less than 2, the density decay is given by

$$\rho(t) \simeq \rho_0 (D\rho_0^{(2/d)} t)^{-d/2}, \tag{3}$$

where D is diffusion constant and ρ_0 is the initial particle density. Equation (3) can be understood either by invoking a single length scaling argument [5] or by using a more elaborate Smoluchowski approach [6,7]. For the two-species case, it was shown (Refs. [5,8–11]) that the upper critical dimension $d_{UC}=4$, and that for $d<4$, the large-t asymptotics is

$$\rho(t) \simeq (\rho_0)^{1/2} (Dt)^{-d/4}. \tag{4}$$

The decay law (4) was confirmed in several numerical simulations of one-, two-, and three-dimensional systems [5,12,13]. It is important to emphasize that in order to observe such power-law decay, it is necessary initially to have an equal number of positive and negative charges, distributed at random. If initial numbers of positive and negative charges are different, one should observe an exponential density decay to the nonvacuum equilibrium (see, e.g., [14]). If the system is stirred well, i.e., long-wavelength fluctuations of charge density are suppressed, the decay law would also be different. We will not consider such cases in this paper.

To better study the role of correlations in reactions without a LRI, a description in terms of secondary quantization operators of creation and annihilation was proposed [15,16]. Further developing this approach, Peliti [17] proposed an ''exact'' (valid to all orders in perturbation expansion) renormalization-group theory for the one-species annihilation and Lee and Cardy [18] suggested a similar approach to the two-species annihilation, both based on the rigorous master equation converted into a field-theoretic formulation. In the latter case, it was shown that the decay law (4) is an asymptotic one for large t and small ρ_0. It was also rigorously proven that the assumptions used in the derivation of Eq. (4) are correct only for $d \geqslant 2$, while for $d < 2$, a more elaborate renormalization-group procedure that includes an effective noise with nontrivial correlations should be carried out.

The addition of the LRI further complicates the picture. Equations of motion become strongly coupled via the nonlocal interaction and only approximate solutions of these equations can be found. Up to date, only Coulombic [19] systems in two dimensions ($d=2$, $n=1$, the case corresponding to particles interacting via a logarithmic potential on a two-dimensional substrate or film) were studied numerically [13,20–22] and results appear to be inconclusive. In these simulations, the particle density exhibited a power-law decay $\rho(t) \sim t^{-\nu}$ with the exponent ν varying from 0.79 ± 0.04 [22] to 0.85 ± 0.05 [13]. Ginzburg *et al.* proposed a scaling theory [13,23] suggesting that for a Coulombic ($n=d-1$) two-dimensional diffusion-annihilation system, annihilation exponent is equal to 0.85, which is close to but less than the mean-field exponent $\nu=1$. Oshanin *et al.* [24] suggested that the exponent should be exactly 1; Ispolatov and Krapivsky [25] came to the same conclusion using an ''inpenetrable domain'' scaling theory. In their theoretical and computational study of the defect annihilation in two-dimensional XY model, Yurke *et al.* [26] argued that the annihilation exponent is 1, with logarithmic correction due to the logarithmic dependence of the mobility on the defect size. Thus the problem of annihilation behavior for the Coulombic system in two dimensions has not been completely resolved, although it seems rather plausible that the final asymptotics is governed by a classical exponent 1, with possible logarithmic correction. It seems to be certain, however, that annihilation in the three-dimensional Coulombic system has a mean-field-type final asymptotics with exponent 1.

Finally, we should point out that until recently, there has been no discussion of the LRI other than Coulombic. Recently, several scaling theories have been suggested to analyze the arbitrary power-law interaction (more short ranged than Coulombic) [23,25,27]. Such problems may arise, e.g., in describing interactions between vacancies and interstitials in a two- or three-dimensional crystal.

In this paper, we propose a self-consistent theory based on coarse-grained hydrodynamic equations of motion for particle number density and charge density fields. This theory allows us to systematically calculate the dependence of the density decay law on initial conditions and to investigate the role of the LRI. It can be shown [31] that the self-consistent approximation that we employ here is equivalent to a resummation of an infinite class of Feynman graphs, which take into account the conserved charge density fluctuations but ignore the less important nonconserved number density fluctuations. This approximation can be further systematically improved by the use of perturbation theory and the renormalization group analysis that is planned to be the subject of future work [31]. In the limit of weak long-range interactions, this approach agrees well with the known theoretical results (see Refs. [5,18,28–30]) for the two-species annihilation $A+B \to \varnothing$, thereby further clarifying the underlying assumptions that led to these results.

The paper is organized as follows. In Sec. II we define all variables and present the equations of motion on which our analysis and results will be based. In Sec. III the self-consistent approximation is described and the asymptotic solutions are obtained and analyzed. Finally, in Sec. IV we analyze the resulting phase diagram and discuss it in the context of the previously obtained results for various diffusion-annihilation systems, which are selected points on our phase diagram.

II. EQUATIONS OF MOTION

Let us consider a system consisting of two kinds of particles, A and B, with A having a positive charge $+q$ and B having a negative charge $-q$. We label their time- and position-dependent concentrations as $n_1(\mathbf{r},t)$ and $n_2(\mathbf{r},t)$, respectively, and impose the condition that $\langle n_1(\mathbf{r},t=0)\rangle = \langle n_2(\mathbf{r},t=0)\rangle = n_0$. The equation of motion for the densities is based on the generalized law of mass conservation, violated by the annihilation process

$$\frac{\partial n_i(\mathbf{r},t)}{\partial t} + \nabla \cdot \mathbf{J}_i = -\mathcal{K} n_1(\mathbf{r},t) n_2(\mathbf{r},t), \tag{5}$$

where the mass current is given by

$$\mathbf{J}_i = -D \nabla n_i(\mathbf{r},t) - \mu q_i n_i(\mathbf{r},t) \nabla V(\mathbf{r},t) \tag{6}$$

and

$$V(\mathbf{r},t) = q \int d^d \mathbf{r}' \frac{n_1(\mathbf{r}',t) - n_2(\mathbf{r}',t)}{|\mathbf{r}' - \mathbf{r}|^{n-1}} \tag{7}$$

is the electrostatic long-range potential at a point $\mathbf{r}$ at time t due to local charge fluctuations away from neutrality; n is the power exponent of the long-range force, μ is particle mobility taken to be a constant, and q is particle charge. Equations (5)–(7) should be solved in conjunction with initial conditions for $n_1(\mathbf{r},t=0)$ and $n_2(\mathbf{r},t=0)$. Since here we are interested in statistical averages, rather than in a dynamic solution for a given system with specific initial conditions, we will focus on the density correlation functions, with averages over random $t=0$ initial conditions.

Equations (5) and (6) represent the coarse-grained continuum limit for the ''real'' equations of motion; all variables in these equations are averaged over ''elementary volume'' n_0^{-1}. Thus only long-wavelength modes are actually described by these equations and therefore only intermediate-time and large-time regimes can be analyzed.

It is more natural and convenient to describe the system in terms of the particle number and charge densities. We denote the former one as $\rho(\mathbf{r},t)$ and the latter one as $f(\mathbf{r},t)$ and relate them to densities n_1 and n_2 as

$$\rho(\mathbf{r},t) = \frac{1}{2}[n_1(\mathbf{r},t) + n_2(\mathbf{r},t)], \tag{8}$$

$$f(\mathbf{r},t) = \frac{1}{2}[n_1(\mathbf{r},t) - n_2(\mathbf{r},t)]. \tag{9}$$

Rewriting Eqs. (5)–(7) using densities f and ρ, we obtain

$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} - D\nabla^2 \rho(\mathbf{r},t) = -\mathcal{K}[\rho^2(\mathbf{r},t) - f^2(\mathbf{r},t)] - Q\nabla\left(f(\mathbf{r},t)\nabla \int d^d\mathbf{r}' \frac{f(\mathbf{r}',t)}{|\mathbf{r}'-\mathbf{r}|^{n-1}} \right), \tag{10}$$

$$\frac{\partial f(\mathbf{r},t)}{\partial t} - D\nabla^2 f(\mathbf{r},t) = -Q\nabla\left(\rho(\mathbf{r},t)\nabla \int d^d\mathbf{r}' \frac{f(\mathbf{r}',t)}{|\mathbf{r}'-\mathbf{r}|^{n-1}} \right). \tag{11}$$

where $Q = \frac{1}{2}\mu q^2$.

In the absence of a LRI, Eqs. (10) and (11) are those analyzed in Refs. [5,11,18,30]. However, the presence of the additional long-range interactions makes the analysis of their asymptotic solutions rather nontrivial and, as we will show below, leads to different dynamic regimes.

It is important to note that Eqs. (10) and (11) do not contain noise terms on their right-hand sides. It has been rigorously shown that such noise terms represent important correlations and in some cases may even become predominant in determining the asymptotic decay rate. In equations of motion describing a near-equilibrium dynamics, a powerful fluctuation-dissipation theorem determines the form of noise correlations. In contrast, in systems far from equilibrium, such as a system of annihilating particles, it can be shown [18,31] that the effective hydrodynamic equations of motion derived from the fundametal master equations contain noise terms with very nontrivial correlations, of a form that could not be easily guessed *a priori*. Lee and Cardy [18] proved that in a two-species reaction without a LRI, such noise leads only to the renormalization of the reaction rate $\mathcal{K}$, but not to the change of the scaling exponents, provided that space dimensionality $d>2$. We have shown, in a similar fashion [31], that for systems with a LRI, the noise has no effect on the asymptotic dynamics for $d>2$, if the renormalization of both $\mathcal{K}$ and Q is implied. In addition, since Eqs. (10) and (11) provide a coarse-grained description on the length scale larger than the interparticle spacing $\rho_0^{-1/d}$, the kinetic coefficients, e.g., $\mathcal{K}$, are effective coefficients that incorporate finite renormalization due to the correlations on short length scales.

In order to simplify further analysis, we divide each of the equations (10) and (11) by ρ_0 and transform everything to dimensionless variables as

$$\rho \to \rho/\rho_0, \quad f \to f/\rho_0, \quad D \to D(\rho_0)^{2/d},$$

$$\mathcal{K} \to \mathcal{K}\rho_0, \quad r \to r(\rho_0)^{2/d}, \quad Q \to Q(\rho_0)^{(n+1)/d}.$$

It is important to notice that Eq. (11) is linear with respect to f, while Eq. (10) is quadratic with respect to f (this points to the system's invariance with respect to the simultaneous charge sign reversal for all particles). In the next section, we will describe the self-consistent approximation and its solutions.

III. SELF-CONSISTENT APPROXIMATION AND SOLUTIONS OF THE EQUATIONS OF MOTION

A. Self-consistent approximation

We now make an important approximation in order to further simplify the analytical treatment of Eqs. (10) and (11), namely, we choose to ignore the fluctuations of the particle density ρ and concentrate on the fluctuations of the conserved charge density f. This assumption is somewhat similar in spirit to the approach of Glotzer and Coniglio [32] for the problem of spinodal decomposition and to the spherical approximation for the Ising model in the limit of $N\to\infty$. Unlike the "classical" mean-field approach, however, the proposed approximation does take into account exactly the charge density fluctuations, and is expected, therefore, to describe at least some of the features of the fluctuation-dominated kinetics.

The justification of the proposed assumption lies in a simple observation that, while the average particle density at any time is nonzero, so that $\langle(\rho-\langle\rho\rangle)^2\rangle/\langle\rho\rangle^2$ is finite and likely to be small, the average charge density is always zero and, therefore, in comparison the charge fluctuations $\langle(f-\langle f\rangle)^2\rangle$ are large. Thus we expect the former fluctuations to be less important that the latter, and we can approximate the particle number density by its average (time-dependent) value in the equations of motion without losing their important features. In a sense, our approximation is a generalization of an argument used by Toussaint and Wilczek [5], in which they based their scaling decay law on a suggestion that $\langle\rho\rangle \approx \sqrt{\langle\rho^2\rangle}$. It seems clear that the approximation of ignoring the number density fluctuations must break down at least below some upper-critical dimension d_{UC} since asymptotically ρ vanishes. In this case our approximation will be valid for $d>d_{UC}$ for all times and in systems below d_{UC} it will be a good approximation up to a crossover time beyond which the asymptotics will be modified by the number density fluctuations. Systematically taking into account these additional fluctuations will be a subject of future work [31].

Taking into account the above approximation, we rewrite Eq. (11) in Fourier representation, taking $\rho(t)$ as a spatially independent but time-dependent function

$$\left(\frac{\partial}{\partial t} + Dk^2 + Q\rho(t)k^{2-\sigma}\right) f(\mathbf{k},t) = 0, \tag{12}$$

where $\sigma = d+1-n$. Equation (10) in this self-consistent approximation is rewritten as

$$\frac{d\rho}{dt} + \mathcal{K}\rho^2 = \mathcal{K}\int \frac{d^dk}{(2\pi)^d} \langle f(\mathbf{k},t) f(-\mathbf{k},t)\rangle, \tag{13}$$

where $\langle\,\rangle$ denotes averaging over initial conditions.

These equations of motion have to be supplemented with initial conditions. It is well known that the initial density distribution plays an important role in determining the scaling decay law. Although the self-consistent approximation employed here is well suited for a comprehensive study of the influence of initial conditions on the dynamics, here we limit our study to a single type of initial condition. Through-out this paper we will focus on the dynamics initiated with a random Gaussian particle distribution, completely characterized by

$$\langle f(\mathbf{k},0)\rangle = 0\,, \tag{14}$$

$$\langle f(\mathbf{k}_1,0)f(\mathbf{k}_2,0)\rangle = \Delta(2\pi)^d\delta^{(d)}(\mathbf{k}_1+\mathbf{k}_2), \tag{15}$$

which constrains the system to charge neutrality at all times, and for simplicity we take the variance $\Delta=1$ ($=\rho_0^2$ in physical units). For most charged experimental systems more relevant initial conditions incorporate the suppression of long-wavelength fluctuations in charge density, which can be modeled (within, e.g., the Debye-Hückel approximation) by $\Delta(k)=\Delta_0 k^2/(k^2+k_s^2)$ in Eq. (15).

The diffusion-only (DO) case ($Q=0$) and Coulombic case ($n=d-1$) are the simplest systems with a relatively clear, yet interesting asymptotic behavior. All the intermediate interactions (arbitrary n and d) lead to a more complicated scaling behavior, with several regimes and crossovers. We will devote a subsection to each of these three cases.

B. Systems without long-range interactions

There are two ways of approaching the limit of ''no long-range interactions'': by decreasing the force constant Q to 0 or by increasing the power exponent n to infinity (interaction with an effectively vanishing range). Obviously, these limits should give the same answer. For simplicity, we will set $Q=0$ and show that our self-consistent approximation yields the well-known results [5–11]

$$\rho(t)\simeq\begin{cases}(\mathcal{K}t)^{-1} & \text{for } d>4 \qquad (16)\\ (Dt)^{-d/4} & \text{for } d<4\,. \end{cases} \tag{17}$$

For $Q=0$, the kinetic equation for f reduces to a simple diffusion equation, with the solution

$$f(\mathbf{k},t)=f(\mathbf{k},0)e^{-Dk^2t}. \tag{18}$$

Substituting this solution (18) for $f(\mathbf{k},t)$ into Eq. (13) and taking into account the initial conditions (14) and (15), we obtain

$$\frac{d\rho}{dt}+\mathcal{K}\rho^2=\frac{\mathcal{K}}{(1+2Dt)^{d/2}}. \tag{19}$$

The exact solution expressible in terms of confluent hypergeometric functions is possible [18]. It can also be easily shown that Eq. (17) describes the asymptotic solution of Eq. (19). This is expected since, as we argued above, the approximations made by Toussaint and Wilczek [5] are very similar to our self-consistent model. The case of $n\to\infty$ will be analyzed in Sec. III D, where it will be shown that for all $n>1+d/2$, the decay law is asymptotically the same as for DO systems.

C. Coulombic systems

In Coulombic systems, the long-range interaction is the strongest possible that one can achieve without making the system thermodynamically unstable (systems with interactions stronger than Coulombic have infinite pressure and chemical potential even if their total charge is zero). Because of this, one would expect the particle density decay for Coulombic systems to be very close or equal to the mean-field law $\rho(t)\approx(\mathcal{K}t)^{-1}$ [33]. As we describe below, the self-consistent approximation predicts the decay exponent $\nu=1$ consistent with this expectation and with some simulations reported in the literature [26]. However, it can be shown [31] that a correction to this mean-field-like decay law can arise from the number density fluctuations and noise for $d\leqslant 2$, both of which have been neglected in the self-consistent theory presented here.

The equation for the evolution of charge density f for the Coulombic systems in the self-consistent approximation can be exactly solved to yield

$$f(\mathbf{k},t)=f(\mathbf{k},0)\exp\left(-Dk^2t-Q\int_0^t\rho(\tau)d\tau\right). \tag{20}$$

Using this solution (20) and the initial condition (14) and (15), $\rho(t)$ can be easily shown to satisfy the differential equation

$$\frac{d\rho}{dt}+\mathcal{K}\rho^2=\mathcal{K}\exp\left(-2Q\int_0^t\rho(\tau)d\tau\right)\int\frac{d^dk}{(2\pi)^d}\times\exp(-2Dk^2t), \tag{21}$$

In order to find the asymptotic solutions, we introduce a variable

$$\Theta=\int_0^t\rho(\tau)d\tau. \tag{22}$$

Equation (21) then transforms to

$$\frac{d^2\Theta}{dt^2}+\mathcal{K}\left(\frac{d\Theta}{dt}\right)^2=\exp(-2Q\Theta)\frac{\mathcal{K}}{(1+2Dt)^{d/2}}. \tag{23}$$

Let us find the ''critical'' dimension d_{UC}, above which the mean-field behavior is manifested. The mean-field solution for Θ is given by

$$\Theta(t)=\frac{1}{\mathcal{K}}\ln(1+\mathcal{K}t)+\cdots, \tag{24}$$

where the ellipsis corresponds to subdominant constant terms and terms decreasing with time. By counting powers of t on the right-hand side (RHS) and the left-hand side (LHS) of Eq. (23), we find that the power of the LHS is -2 and the power of the RHS is $-d/2-2(Q/\mathcal{K})$. Obviously, for the mean-field solution to be valid asymptotically, the power of the LHS should be larger than the power of the RHS, which happens for systems with dimensionality larger than the critical dimension,

$$d>d_{\mathrm{UC}}=4\left(1-\frac{Q}{\mathcal{K}}\right). \tag{25}$$

If $Q\geqslant\mathcal{K}$, the asymptotic kinetics is determined by the slower process, which is the annihilation, with possibly the interaction renormalized $\mathcal{K}$ (implicitly assumed here), and the Coulomb interaction and diffusion are asymptotically irrelevant. For $Q<\mathcal{K}$, the Coulomb interaction has an interesting effect of continuously lowering the upper-critical dimension from 4 (for $Q/\mathcal{K}=0$, in which case for $d<4$ the diffusion dominates giving $\nu=d/4$) down to d_{UC} given above.

In order to analyze the kinetics when space dimensionality d is below d_{UC} we employ the ''steady-state'' approximation, which suggests that at long times the time derivative on the LHS of Eq. (21) is the smallest of the three terms. In this case, the equation of motion can be written as

$$\frac{d\Theta}{dt}=\exp(-Q\Theta)(1+2Dt)^{-d/4}, \tag{26}$$

$$\Theta(0)=0\,. \tag{27}$$

An exact solution of this equation is

$$\Theta=\frac{1}{Q}\ln\left(1+\frac{Q}{2D(1-d/4)}(\{1+2Dt\}^{1-d/4}-1)\right) \tag{28}$$

and

$$\rho=\dot{\Theta}. \tag{29}$$

It can be easily shown that for large t, the asymptotic solution for the particle density decay ($d<d_{\mathrm{UC}}\leqslant 4$) is

$$\rho\simeq\frac{1-d/4}{Qt}, \tag{30}$$

predicting the asymptotic decay exponent $\nu=1$ for Coulomb systems, as in the mean-field regime, although with a Q-rather than $\mathcal{K}$-determined amplitude. This large-t limit is achieved when

$$t>t_L=\frac{1}{2D}\left[\frac{2D(1-d/4)}{Q}\right]^{4/(4-d)}, \tag{31}$$

and it can be easily seen that in the limit of $Q\rightarrow 0$ (vanishing interactions) the transition time t_L to this region becomes infinite, i.e., this time is never reached.

If diffusion is faster than the deterministic Coulomb interaction-driven relaxation, i.e., $D>Q$, then for times less than t_L, the annihilation is governed by the intermediate asymptotics

$$\Theta=\frac{1}{(1-d/4)D}(2Dt)^{1-d/4}, \tag{32}$$

so the particle density is described by the Toussaint-Wilczek solution up to the crossover time t_L:

$$\rho=\dot{\Theta}\simeq(Dt)^{-d/4}. \tag{33}$$

This intermediate asymptotics, which exists only for $d<4$, reflects early times diffusion-dominated decay, with the slower deterministic Coulomb interaction-driven classical t^{-1} decay appearing only at times later than t_L. In contrast, for $D<Q$ or if $d>4$, there is no extended intermediate regime and one should see a quick transition to a classical decay law. Although within the self-consistent approximation the asymptotic $1/t$ decay is not affected by the choice of reasonable initial conditions, the intermediate diffusion-dominated decay is certainly affected by our choice of random Gaussian uncorrelated initial conditions given in Eq. (15). For instance, if the screened Debye-Hückel initial conditions are used with $\Delta(k)=\Delta_0 k^2/(k^2+k_s^2)$, then this $\Delta(k)$ will appear as a multiplicative kernel under the k integral in Eq. (21). It will then modify the intermediate decay exponent from $d/4$ to $\nu=(d+2)/4$ (and, for $d>2$, eliminating this intermediate region altogether), without modifying the asymptotic decay of Eq. (30). To sum up, we find that within the self-consistent approximation, Coulombic systems ($n=d-1$) asymptotically exhibit the t^{-1} density decay, consistent with several scaling arguments and simulations [24–26,33].

D. Intermediate systems

Let us now consider the general case of long-range interactions with a power-law $d-1<n<\infty$ that is of shorter range (weaker) than the Coulomb interaction considered in the preceding subsection. Equations (12) and (13) can be solved to yield

$$f(\mathbf{k},t)=f(\mathbf{k},0)\exp\left(-Dk^2t-Qk^{2-\sigma}\int_0^t\rho(\tau)d\tau\right), \tag{34}$$

$$\frac{d\rho}{dt}+\mathcal{K}\rho^2=\mathcal{K}\int\frac{d^dk}{(2\pi)^d}\exp\left(-2Dk^2t-2Qk^{2-\sigma}\int_0^t\rho(\tau)d\tau\right), \tag{35}$$

where $\sigma=d-n+1$.

Equation (35) is significantly more complicated than its analogs for either Coulombic or noninteracting cases. Nevertheless, it is possible to find its power-law asymptotic solutions. Using an asymptotic analysis analogous to that described in Sec. III C, we find several kinetic regimes depending on the values of d and n. These regimes depend crucially on the charge density relaxation mechanism, i.e., whether the LRI or diffusion determines the relaxation rate of $f(\mathbf{k},t)$ at late times. In order to analyze the asymptotic behavior of the system, we again introduce the integrated density $\Theta(t)$ as defined in Eq. (22). We also assume a power law for the density and, for $d<4$, neglect the term $d\rho/dt$. In this case, the equations of motion are

$$\frac{d\Theta}{dt}=\sqrt{\int\frac{d^dk}{(2\pi)^d}\exp[-2Dk^2t-2Qk^{2-\sigma}\Theta(t)]}, \tag{36}$$

$$\rho(t)=\frac{d\Theta}{dt}. \tag{37}$$

Depending on σ and d, either the first or the second term in the exponential in Eq. (36) dominates for large t, corresponding to either diffusive or superdiffusive relaxation. We first assume that diffusive relaxation is prevalent and determine the conditions when it is true. In the case of diffusive relaxation mechanism and at large t, Eq. (36) can be simplified to yield

$$\frac{d\Theta}{dt}=\sqrt{k_d(Dt)^{-d/2}\int_0^\infty x^{d/2-1}dx\exp\left(-x-\frac{2Q\Theta(t)}{(2Dt)^{1-\sigma/2}}x^{1-\sigma/2}\right)},\tag{38}$$

where k_d is a dimensionless constant absorbing integration over angular variables and the integral becomes time independent for large t if the second term in the exponential vanishes with time. If we assume the power-law dependence for ρ and Θ,

$$\rho(t)\propto t^{-\nu},\tag{39}$$

$$\Theta(t)\propto t^{1-\nu},\tag{40}$$

then it follows from Eq. (38) that for the predominantly diffusive system $\nu=d/4$, as expected. Thus, in order for this solution to be self-consistent, we must require that

$$1-\nu<1-\sigma/2,\tag{41}$$

$$\sigma>d/2,\tag{42}$$

and, from the definition of σ, we determine the region where the relaxation and density decay are diffusion limited:

$$n>1+d/2,\tag{43}$$

$$d<4\ .\tag{44}$$

This region is marked FD (fluctuation-dominated) in Fig. 1. In it, the LRIs are irrelevant at large t, although they may influence the density decay kinetics for intermediate t. The asymptotic decay law in the FD region is given by

$$\rho\simeq(Dt)^{-d/4}.\tag{45}$$

In the region referred to as the IR (intermediate region), which lies below FD in Fig. 1 ($d-1\leqslant n\leqslant 1+d/2$), the LRIs are strong and dominate the diffusion at large t. To investigate the asymptotics of the decay in this region, we rewrite Eq. (36) as

$$A(1-\nu)t^{-\nu}=\sqrt{k_d(QAt^{1-\nu})^{-d/(2-\sigma)}\int_0^\infty x^{[d/(2-\sigma)]-1}dx\exp\left(-x-\frac{2Dt}{(2QAt^{1-\nu})^{2/(2-\sigma)}}x^{2/(2-\sigma)}\right)},\tag{46}$$

using $\Theta=At^{1-\nu}$. By solving Eq. (46) approximately we find two asymptotics in this region:

$$\rho(t)\simeq(Dt)^{-d/4},\tag{47}$$

valid at intermediate times, and

$$\rho(t)\simeq(Qt)^{-\nu},\tag{48}$$

where

$$\nu=\frac{d}{4+d-2\sigma}=\frac{d}{2-d+2n}\tag{49}$$

for asymptotically large times.

The crossover time t_c from the diffusion-dominated decay to the LRI-dominated decay is

$$t_c\approx D^{(2-d+2n)/(2+d-n)}Q^{-2/(1+d/2-n)}.\tag{50}$$

Thus, in this region (marked IR in Fig. 1) the LRI accelerates the relaxation of the initial density fluctuations and thereby speed up the annihilation. If $Q<D$, the diffusive relaxation and the $d/4$ law may be observed for the intermediate t before the transition to the superdiffusive relaxation and faster decay takes place for $t>t_c$.

If $d>4$ [the mean-field (MF) region in Fig. 1], spatial fluctuations become irrelevant and the classical kinetic-rate equation becomes asymptotically correct, so the decay law in this region is given by

$$\rho(t)\simeq(\mathcal{K}t)^{-1},\tag{51}$$

as previously discussed.

IV. SUMMARY AND CONCLUSIONS

In the present work we derive approximate kinetic equations for the annihilation-diffusion process with long-range forces. To analyze the asymptotic decay law for systems with $d>2$, we proposed a self-consistent method of calculating the average particle density as a function of time. Since the total particle density is a nonconserved order parameter with positive average at all times, we argued that its fluctuations are less important in determining dynamics of annihilation than that of a conserved order parameter — the charge density. This approximation self-consistently decouples two kinetic equations and makes it possible to find the asymptotic solutions. In the limit of weak long-range interaction (via taking either $n\to\infty$ or $Q\to 0$), self-consistent equations of motion are reduced to those of Toussaint and Wilczek [5].

For Coulombic systems in more than two dimensions, our model yields the mean-field exponent $\nu=1$, yet the role of segregation (i.e., charge density fluctuations) is important

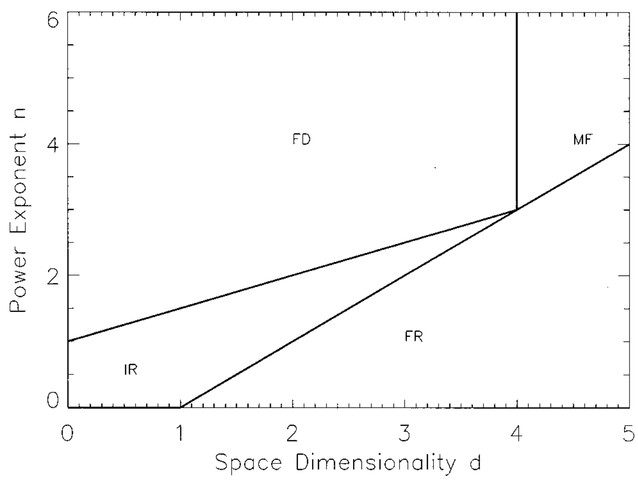

FIG. 1. Phase diagram of the annihilation-diffusion reaction with long-range forces. FR, forbidden region (below the Coulombic line); IR, intermediate region, where the large-*t* asymptotics is determined by the LRI, FD, fluctuation-dominated region, where the large-*t* asymptotics is determined by diffusion and initial fluctuations; MF, mean-field region, where the large-*t* asymptotics is determined by the kinetic-rate equation.

and cannot be simply left out. Ispolatov and Krapivsky [25] proposed the unpenetrable domain scaling concept, which in the Coulombic case results in decay exponent 1 independently of space dimensionality. Both their model and our self-consistent approximation neglect possible fluctuation modes due to the spatial variation of particle density, as well as noise, which can lead to some slowing down of the reaction kinetics, as indicated by simulations. Elucidation of such modes and their role should require a detailed account of noise and possibly use of a renormalization-group analysis when $d=2$, since it is the critical dimension for the annihilation-diffusion problem. Since neglecting noise in this problem appears to be justified for long-time asymptotics for systems with $d>2$ [31], it is possible that the exponent $\nu=1$ for $d>2$ Coulombic systems is exact, even though there is no experimental evidence to support this conclusion.

The analysis of the self-consistent equations of motion (12) and (13) suggests that there is a region in the (n,d) phase diagram (the region labeled IR in Fig. 1) in which the large-t asymptotics of the density decay is determined by long-range forces. The annihilation initally depletes the positively charged region of negative particles and vice versa, and then the decay rate is determined by the speed of particle drift from such regions. Again, our large-*t* asymptotics here agrees with the unpenetrable domain theory of Ispolatov and Krapivsky, although their model predicts different boundaries of the IR region in the (n,d) phase space ($n=1+d/2$ is the upper boundary of the IR region in the self-consistent model and $n=d$ is the upper boundary of the IR region in the unpenetrable domain model; the lower boundary in both theories is the Coulombic line $n=d-1$). The self-consistent theory also predicts a crossover from diffusion-dominated decay to the LRI-dominated decay at large times for the systems in this region.

Because the self-consistent model is a semi-mean-field approximation (it completely neglects particle density fluctuations and takes into account only the concerved charge density fluctuations), it should be considered only as a first step. A systematic perturbative analysis of Eqs. (10) and (11) around our self-consistent solution is needed to assess the role of neglected number density fluctuations and noise [31].

The proposed self-consistent model, its somewhat uncontrolled approximations notwithstanding, represents an important tool in the qualitative analysis of dynamic processes in two-component systems with one conserved and one nonconserved variable. It predicts different annihilation behavior (IR regime) and different crossovers between diffusion-driven and LRI-driven decay regions, reproduces all known results for the annihilation problem in special limits, and can be used to systematically study the role of initial conditions in such processes.

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