Reaction-Controlled Diffusion

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The dynamics of a coupled two-component nonequilibrium system is examined by means of continuum field theory representing the corresponding master equation. Particles of species A may perform hopping processes only when particles of different type B are present in their environment. Species B is subject to diffusion-limited reactions. If the density of B particles attains a finite asymptotic value (active state), the A species displays normal diffusion. On the other hand, if the B density decays algebraically $\propto t^{-\alpha}$ at long times (inactive state), the effective attractive A-B interaction is weakened. The combination of B decay and activated A hopping processes gives rise to anomalous diffusion, with mean-square displacement $\langle \vec{x}_A(t)^2 \rangle \propto t^{1-\alpha}$ for $\alpha < 1$. Such algebraic subdiffusive behavior ensues for n-th order B annihilation reactions ($nB \to \emptyset$) with $n \geq 3$, and $n = 2$ for $d < 2$. The mean-square displacement of the A particles grows only logarithmically with time in the case of B pair annihilation ($n = 2$) and $d \geq 2$ dimensions. For radioactive B decay ($n = 1$), the A particles remain localized. If the A particles may hop spontaneously as well, or if additional random forces are present, the A-B coupling becomes irrelevant, and conventional diffusion is recovered in the long-time limit.

I. INTRODUCTION

There has been considerable effort to elucidate the properties and conditions of anomalous diffusive behavior. A simple physical realization is given by diffusion on a fractal lattice [1], where due to the increasing number of paths within the lattice, the time for a diffusion process will be prolonged. Also diffusion in random media with quenched disorder may be anomalous. Depending on the distribution of barrier heights (or depths of traps) one may observe normal diffusive or subdiffusive behavior, respectively, if the available number of diffusive paths is reduced by the presence of obstacles [2,3]. Here we discuss a quite different situation in which diffusion is activated by the presence of particles or excitations which also propagate diffusively, but in the course of time decay. As a result the activated diffusion is rendered anomalous because the number of available paths decreases with time. However, the resulting structure of diffusive paths is not static, but evolves temporally. One may call this phenomenon dynamical fractality or dynamical disorder, depending on how the spatial distribution of excitations evolves in time.

We model this scenario by starting from a two-component system consisting of distinct particle species A and B, with local time-dependent densities $\rho_A(\vec{x}, t)$ and $\rho_B(\vec{x}, t)$. An A particle is allowed to perform hopping processes between adjacent neighboring sites on a lattice, provided there are one or more B particles present in its vicinity. To be more specific, an A particle hops from a site j to a neighboring point i subject to the condition that this site i is already occupied with a particle of species B, and with a rate proportional to the local B particle density. Obviously, such an effective attractive interaction strongly influences the diffusive mobility of the A species: Their mean-square displacement $\langle \vec{x}_A(t)^2 \rangle$ will depend on the time evolution of the local B density $\rho_B(\vec{x}, t)$.

A non-trivial temporal behavior for $\rho_B(\vec{x}, t)$ will result if we submit the B species locally to diffusion-limited reactions such as n-th order annihilation $nB \to \emptyset$ (at the same or adjacent lattice points) or combined annihilation ($n \geq 2$) and spontaneous offspring production $B \to (m+1)B$ (the B particles then perform branching and annihilating random walks, BARW). Once the time-dependence of $\rho_B(t)$ has been determined, we shall see that the A kinetics is in the long-time limit to good approximation described on the basis of the associated mean-field rate equation. When the B species is in an active state, i.e., $\rho_B(\vec{x}, t \to \infty) = \rho_B^\infty > 0$, with a basically homogeneous distribution in space, the A particles will display normal diffusive behavior, with a diffusion constant $D_A \propto \rho_B^\infty$. In such a situation one has dynamical disorder, but there is always a finite fraction of sites available for hopping. However, an inactive phase, or the BARW critical point, are described either by an exponential decay $\rho_B(\vec{x}, t \to \infty) \propto e^{-\lambda t}$, in which case the A particles remain localized, or by a power-law decrease $\rho_B(\vec{x}, t \to \infty) \propto t^{-\alpha}$ with a characteristic exponent $\alpha > 0$. The diminishing density of B particles reduces the induced mobility of the A species, and these competing effects lead to subdiffusive behavior $\langle \vec{x}_A(t)^2 \rangle \propto t^{1-\alpha}$ for $\alpha < 1$. In the borderline case $\alpha = 1$ one has merely logarithmic growth $\langle \vec{x}_A(t)^2 \rangle \propto \ln t$.

Intuitively some of this behavior can be easily understood in the limit of vanishing diffusion of the B-particles.

We allow for multiple occupation, i.e., the site occupation number can be any integer between zero and infinity. When the occupation number of a B particles at a certain lattice point is nonzero, that site is available for the A species. In an inactive state the number of B particles will be permanently reduced by reactions leading to a decreasing density of available sites for the A species. This procedure can be viewed as an effective "thinning out" of lattice sites that leads to subdiffusive behavior for the A particles reminiscent of (but distinct to) the mathematically considerably more complex phenomenon of diffusion on a fractal lattice.

II. THE MODEL

Here we present a more precise definition of our model in terms of a master equation which we formulate in the standard Fock-space formulation [4–6], sometimes called "quantum Hamiltonian formalism" [7,8], particularly for particles with hard-core repulsion. Physically, this interaction is usually insignificant unless exclusion between different particle species or external driving forces need to be taken into account. This is intuitively clear for annihilating processes where the particle density tends to zero at late times [9–12], but remains true also in the absence of particle reactions [13] even in one dimension. In some models, however, e.g., the annihilation-fission model [14] or pair contact process with diffusion [15,16], site occupation number constraints do play a crucial role. Hence, usually the specific choice of model is prescribed by the mathematical treatment used to analyze it. In the present context where we shall employ mean-field techniques and renormalization group arguments it is more advantageous to consider particles without site exclusion (for recent reviews, see e.g. Refs. [7,17]).

We consider a system consisting of two different types of particles denoted as A and B. The time evolution can be represented through an evolution operator L [4–6]. The corresponding annihilation and creation operators are written as a_i (b_i) and $a_i^\dagger$ ($b_i^\dagger$), where the index indicates a lattice point in d space dimensions. For example, the normal hopping process of species B from a site j to its neighbor i is described by the evolution operator $D\,(b_i^\dagger b_j - b_j^\dagger b_j)$, and for the entire lattice therefore

$$L_B = D \sum_{(ij)} \left(b_i^\dagger - b_j^\dagger \right) (b_j - b_i) \ , \qquad (2.1)$$

where D is the hopping rate or diffusion constant.

An analogous expression would describe free diffusion of the A particles. Here, however, we examine the situation that such a process is only allowed if there is at least one B particle present at site i. If no representative of species B is available at that site, an A particle cannot move there. The time evolution operator for that process is proportional to $(a_i^\dagger a_j - a_j^\dagger a_j) b_i^\dagger b_i$. The corresponding

hopping process will occur provided an A particle is in fact present at site j and at least one B particle occupies site i. Moreover, its rate is actually proportional to the number of B particles present at site i. For the full system we obtain

$$L_A = \widetilde{D} \sum_{(i,j)} \left(a_i^\dagger - a_j^\dagger \right) \left(a_j\, b_i^\dagger\, b_i - a_i\, b_j^\dagger\, b_j \right) \ . \qquad (2.2)$$

Here, $\widetilde{D}$ denotes the induced transition rate for the dynamical process of species A.

In contrast to species A, the B particles are subject to local reactions. A decreasing number of B particles will lead to a slowing down for the motion of A's through the lattice. For n-th order annihilation reactions $nB \to \emptyset$, the nonequilibrium evolution operator reads [18]

$$L_R = \lambda_n \sum_i \left(1 - b_i^{\dagger\,n} \right) b_i^n \ . \qquad (2.3)$$

Obviously, this operator describes the annihilation of n particles of type B at a lattice site i provided such particles are available; λ_n denotes the corresponding rate. Similarly, spontaneous branching processes $B \to (m+1)B$ with rate σ_m are described by [19]

$$L_P = \sigma_m \sum_i \left(b_i^{\dagger\,m} - 1 \right) b_i^\dagger b_i \ . \qquad (2.4)$$

Together with Eq. (2.1), L_R and L_P represent the time evolution operator for branching annihilating random walks (BARW).

The complete dynamics is determined by $L = L_A + L_B + L_R(+L_P)$, and may be encoded into a time-dependent "state vector" [4]

$$|F(t)\rangle = \sum_{n_i} P(\vec{n}, t) \, |\vec{n}\rangle \ . \qquad (2.5)$$

Here $P(\vec{n}, t)$ is the evolving probability distribution for the unrestricted site occupation numbers $\vec{n} = \{n_i\}$ for both A and B particles, and $|\vec{n}\rangle$ a basic vector containing all possible entries $n_i = 0, 1, 2, ...\infty$, i.e., the eigenvalues of the second-quantized bosonic particle number operators $a_i^\dagger a_i$ and $b_i^\dagger b_i$, respectively. The state $|0\rangle$ represents the vacuum with no particles present, $a_i |0\rangle = 0 = b_i |0\rangle$. The state vector obeys the equation of motion

$$\partial_t |F(t)\rangle = L \, |F(t)\rangle \ , \qquad (2.6)$$

or formally $|F(t)\rangle = e^{Lt} |F(0)\rangle$.

The nonequilibrium operator L corresponds to, and is obtained from the evolution operator L' of the classical master equation that can generally be written as

$$\partial_t \, P(\vec{n}, t) = L' \, P(\vec{n}, t) \ , \qquad (2.7)$$

and the matrix elements of L and L' are uniquely related to each other. The time-dependent average of an arbitrary physical quantity $G(\vec{n})$ with the probability distribution $P(\vec{n}, t)$ can be cast into a "matrix element" form for the corresponding second-quantized operator $G(t)$

$$\langle G(t)\rangle = \sum_{n_i} P(\vec{n},t)\, G(\vec{n}) = \langle\Psi|\, G\, |F(t)\rangle \ , \qquad (2.8)$$

with the projection state $\langle\Psi| = \langle 0|\, \exp\sum_i(a_i+b_i)$. Using the relation $\langle\Psi|\, L = 0$, the evolution equation for an arbitrary operator G becomes

$$\partial_t\,\langle G\rangle = \langle\Psi|\,[G,L]\,|F(t)\rangle \ . \qquad (2.9)$$

All the dynamical equations governing the classical problem are thus determined by the commutation rules of the underlying operators and the structure of the evolution operator L. In our case the dynamics of the model is given by induced hopping processes for the A particles and diffusion-limited reactions for the B species, which we shall assume to be distributed randomly at the initial time $t=0$.

As a final step, we employ coherent basis states to represent the matrix element (2.8) by means of a path integral [6,17], and take the continuum limit. Absorbing factors containing the lattice constant into the diffusion and reaction rates, we may compute averages with a dynamical weight $\exp\big(-\mathcal{A}[\hat{a},a,\hat{b},b]\big)$ consisting of contributions to the bosonic field action $\mathcal{A}$ describing the ordinary B diffusion

$$\mathcal{A}_B[\hat{b},b] = \int d^dx \int dt\ \hat{b}\,\big(\partial_t b - D\,\nabla^2 b\big) \ , \qquad (2.10)$$

the pure n-th order annihilation reactions

$$\mathcal{A}_R[\hat{b},b] = -\lambda_n \int d^dx \int dt\ \big(1-\hat{b}^n\big)\, b^n \ , \qquad (2.11)$$

or offspring production processes,

$$\mathcal{A}_P[\hat{b},b] = \sigma_m \int d^dx \int dt\ \big(1-\hat{b}^m\big)\, \hat{b}\,b \ , \qquad (2.12)$$

respectively. Finally the A diffusion as induced by the coupling to the B species,

$$\mathcal{A}_A[\hat{a},a,\hat{b},b] = \int d^dx \int dt\ \hat{a}\,\Big[\partial_t a - \widetilde{D}\,(\nabla^2 a)\,\hat{b}\,b$$
$$+\widetilde{D}\,a\,\nabla^2(\hat{b}\,b)\Big] \ . \qquad (2.13)$$

Notice that $\hat{b}(\vec{x},t)\, b(\vec{x},t)$ represents the local density $\rho_B(\vec{x},t)$ (when appropriate ensemble averages are taken); the A diffusion is thus mediated by the presence of B particles. We remark that apart from the continuum limit, the mapping of the master equation onto the above field theory is exact and involves no further approximations. (We have omitted the boundary contributions stemming from the initial conditions and the projection state here.)

III. GENERAL CONSIDERATIONS

Clearly, the dynamic process for the A particles as defined above is induced by the coupling to the reactive B species only. When there are no B particles present, $\rho_B(x,t)=0$, the A dynamics obviously ceases. Indeed, it turns out that there appears no noise in the dynamic equation governing the A kinetics, which would formally appear as a contribution $\propto \hat{a}\,\hat{a}$ (or higher powers of $\hat{a}$) in the dynamic functional. In fact, any stochasticity emerges as a result of spatio-temporal fluctuations for the B species (essentially reaction noise here). In order to further elucidate this point, we may derive effective Langevin-type equations for the local densities ρ_A and ρ_B. To this end, we need to perform the shifts $\hat{a}=1+\widetilde{a}$, $\hat{b}=1+\widetilde{b}$, which take care of the annihilation operators appearing in the projection state $\langle\Psi|$, see Ref. [17]. To be specific, let us consider the case of B pair annihilation reactions. Omitting temporal boundary terms describing the initial configuration, the new action becomes

$$\mathcal{A}[\hat{a},a,\hat{b},b] = \int d^dx \int dt\ \Big[\widetilde{a}\Big(\partial_t a - D'\,\nabla^2 a - \widetilde{D}\,(\nabla^2 a)\,b$$
$$+\widetilde{D}\,a\,(\nabla^2 b)\Big) - \widetilde{D}\,\widetilde{a}\,(\nabla^2 a)\,\widetilde{b}\,b + \widetilde{D}\,\widetilde{a}\,a\,\nabla^2(\widetilde{b}\,b)$$
$$+\widetilde{b}\,\big(\partial_t b - D\nabla^2 b + 2\lambda\,b^2\big) + \lambda\,\widetilde{b}^2 b^2\Big] \ . \qquad (3.1)$$

Here, we have allowed for additional ordinary A diffusion processes with rate D'. This dynamic action is equivalent to the following set of coupled Langevin equations,

$$\partial_t a = D'\,\nabla^2 a + \widetilde{D}\,(\nabla^2 a)\,b - \widetilde{D}\,a\,(\nabla^2 b) + \zeta \ , \qquad (3.2)$$
$$\partial_t b = D\,\nabla^2 b - 2\lambda\,b^2 + \eta \ , \qquad (3.3)$$

where the fluctuating forces with zero mean are characterized by the noise correlations

$$\langle\zeta(\vec{x},t)\,\zeta(\vec{x}',t')\rangle = 0 \ ,$$
$$\langle\zeta(\vec{x},t)\,\eta(\vec{x}',t')\rangle = \widetilde{D}\,[\nabla^2 a(\vec{x},t)]\,b(\vec{x},t)\,\delta(\vec{x}-\vec{x}')\,\delta(t-t')$$
$$-\widetilde{D}\,a(\vec{x},t)\,\nabla^2[b(\vec{x},t)\,\delta(\vec{x}-\vec{x}')\,\delta(t-t')] \ ,$$
$$\langle\eta(\vec{x},t)\,\eta(\vec{x}',t')\rangle = -2\,\lambda\,b(\vec{x},t)^2\,\delta(\vec{x}-\vec{x}')\,\delta(t-t') \ . \qquad (3.4)$$

Taking averages, we may then identify $\rho_A(t) = \langle a(\vec{x},t)\rangle$ and $\rho_B(t) = \langle b(\vec{x},t)\rangle$, as Eqs. (3.2) and (3.3) obviously generalize the mean-field rate equations for the local particle densities. The reaction noise for the B species displays the characteristic negative correlations ("imaginary noise"), which reflect the particle *anti*correlations induced by the annihilation reaction [18,20,17,14]. When there are no B particles left $[b(\vec{x},t)=0]$, the fluctuations cease, characteristic of an absorbing inactive state. As anticipated, no noise contributions exist for the pure A dynamics, but there appear A-B noise cross-correlations. (Notice that pure diffusion noise does not appear explicitly here.)

Next, let us study what happens when the A particles are subject to an additional random force that leads to ordinary diffusion, i.e., the term $\propto D'$ in the action (3.1). Obviously, one should expect that the induced diffusion

$\propto \widetilde{D}$ is suppressed in this situation, and in the long-time limit standard diffusion prevails. This becomes indeed clear through simple power counting, introducing a momentum scale κ, i.e., $[x] = \kappa^{-1}$, and measuring time scales as $[t] = \kappa^{-2}$, as appropriate for diffusive dynamics. Then $[D] = [D'] = \kappa^0$ become dimensionless, and we infer the field scaling dimensions $[\hat{a}] = [\hat{b}] = [\widetilde{a}] = [\widetilde{b}] = \kappa^0$ and $[a] = [b] = \kappa^d$, as to be expected for d-dimensional particle densities. The remaining couplings (reaction rates) acquire the scaling dimensions

$$[\sigma_m] = \kappa^2 \ , \ [\lambda_n] = \kappa^{2-(n-1)\,d} \ , \ [\widetilde{D}] = \kappa^{-d} \ . \quad (3.5)$$

A positive scaling dimension means that the corresponding parameter is relevant in the renormalization-group (RG) sense. E.g., the branching rate σ_m carries the dimensions of a "mass" term, and indeed represents the decisive control parameter for BARW: In mean-field theory, the critical point must be at $\sigma_m = 0$, and is therefore described by the pure annihilation model, while for any positive σ_m there will be only an active phase characterized by exponential correlations. The annihilation rate is relevant for $d < 2/(n-1)$ dimensions, and irrelevant for $d > 2/(n-1)$ [19]. Hence we identify the upper critical dimension, below which fluctuations in fact dominate the asymptotic behavior, as $d_c(n) = 2/(n-1)$ for n-th order annihilation processes [21,18]. Thus, for $n > 3$ fluctuations are not too important in any physical dimension $d \geq 1$.

Furthermore we notice that the coupling $\widetilde{D}$ is *irrelevant*, i.e., compared to the other parameters in the theory its influence should become negligible in the asymptotic long-time, long-wavelength limit. Evidently, $\rho_B(\vec{x}, t)$ either vanishes (inactive phase) or approaches a constant ρ_B^∞ (active phase) as $t \to \infty$. In the former case, normal B diffusion, if present ($D' > 0$), will dominate; in the latter situation, the combined quantity $\widetilde{D}\rho_B^\infty$ will effectively act as an ordinary diffusion constant, numerically renormalizing D'. In any case, we see that the ordinary A diffusion process is not qualitatively affected by the induced hopping through attractive coupling to the B density, and the associated noise cross-correlations. Also when $D' = 0$, as in our original model, and in a system with an initially *finite* number of B particles, *asymptotically* the A particles either remain localized or display standard diffusion. In this respect, in numerical simulations the induced anomalous diffusion in which we are interested here would appear as a *crossover* feature in the long-time kinetics and correspond to corrections to scaling to the leading asymptotic time dependence. In an infinite system, however, with initially finite B *density*, the anomalous diffusion regime will persist indefinitely.

A corollary of these observations is that the rate $\widetilde{D}$ does not acquire any non-trivial frequency or time dependence in the infrared. In the field theory language, we note that neither diffusive propagator for the A or B species can be renormalized by the $(\hat{a}\hat{b}ba)$ four-point vertex in the unshifted action (2.13), or equivalently, the

three- and four-point vertices in the shifted action (3.1). Consequently, the renormalization for the vertex functions $\Gamma_{\hat{a}\hat{b}ba}$ or $\Gamma_{\widetilde{a}\widetilde{b}ba}$ and $\Gamma_{\widetilde{a}ba}$, respectively, can be determined to *all* orders in the perturbation expansion (with respect to $\widetilde{D}$) by means of a Bethe–Salpeter equation, or equivalently, a geometric series of loops containing just the A and B propagator. This leads to the renormalized wavevector- and frequency-dependent coupling

$$\widetilde{D}_R(\vec{q}, \omega) = \widetilde{D}\left[q^2 + \widetilde{D}\int \frac{d^d p}{(2\pi)^d}\, \frac{p^2\,[(\vec{q}-\vec{p})^2 - p^2]}{-i\omega + D'p^2 + Dp^2}\right]^{-1} , \quad (3.6)$$

where $\vec{q}$ and ω denote the momentum and frequency transfer between the A and B particles. We may now set $D' = 0$ again, and investigate the long-wavelength limit $\vec{q} \to 0$,

$$\left.\frac{\partial}{\partial q^2}\widetilde{D}_R(\vec{q}, \omega)\right|_{q=0} = \widetilde{D}\left[1 + \frac{\widetilde{D}}{D}\left(\frac{\omega}{D}\right)^{\frac{d}{2}}\int \frac{d^d k}{(2\pi)^d}\frac{k^2}{-i + k^2}\right]^{-1} , \quad (3.7)$$

where $k^2 = Dp^2/\omega$. Thus, as $\omega \to 0$, the fluctuation corrections vanish (provided the integral is regularized in the ultraviolet with an appropriate cutoff), and the renormalized coefficient $\widetilde{D}_R$ in Eq. (3.7) approaches the original "bare" constant $\widetilde{D}$.

This is to be contrasted with the infrared-singular behavior of e.g. the B pair annihilation rate, for which an analogous procedure yields [18]

$$\lambda_R(\vec{q}, \omega) = \lambda\left[1 + \frac{\lambda}{D}\int \frac{d^d p}{(2\pi)^d}\, \frac{1}{-i\omega/D + q^2/4 + p^2}\right]^{-1} ,$$

$$\lambda_R(0, \omega) = \lambda\left[1 + \frac{\lambda}{D}\left(\frac{\omega}{D}\right)^{\frac{d-2}{2}}\int \frac{d^d k}{(2\pi)^d}\frac{1}{-i + k^2}\right]^{-1} . \quad (3.8)$$

For $d > d_c(2) = 2$, again $\lambda_R(0,0) = \lambda$ is just the original rate constant, resulting in the mean-field power law $\rho_B(t) \propto t^{-1}$. However, for $d < d_c(2) = 2$, $\lambda_R(0,\omega) \propto \omega^{1-d/2}$ vanishes for low frequencies. Inserting the corresponding effective time-dependent rate $\lambda_R(t) \propto t^{-1+d/2}$ into Eq. (3.3) leads to the correct slower algebraic decay $\rho_B(t) \propto t^{-d/2}$.

In summary, the B process itself is, per definition of our model, not influenced by the A dynamics. In the renormalization group treatment, this is reflected by the fact that the coupling $\widetilde{D}$ is irrelevant, and thus does not affect the long-time behavior. Yet the induced hopping rate $\widetilde{D}$ is of course crucial for the A species kinetics, and must be kept even in the mean-field approximation. We may thus solve for the B kinetics first, and then explore its influence on the induced A diffusion. Henceforth, we shall again set $D' = 0$, as otherwise simple ordinary A

diffusion would ensue, with $\widetilde{D}$ then irrelevant also for the A kinetics, and the entire coupling of the A and B processes would disappear asymptotically. In the following, we shall study the A kinetics assuming a spatially homogeneous, but time-dependent distribution of B particles, which leads us to a mean-field description.

IV. MEAN-FIELD EVOLUTION EQUATIONS

A. Annihilation kinetics

Let us assume we can ignore spatial fluctuations for the B species entirely, and ignore the reaction noise. For the n-th order annihilation processes, we saw that this as at least a qualitatively correct description for $d > d_c(n) = 2/(n-1)$, i.e., for $d > 2$ in the case of pair annihilations, $d > 1$ for the third-order process $3B \to \emptyset$, and in any physical dimension for $n > 3$. The evolution equation can either be obtained directly from the non-equilibrium operator L_R in Eq. (2.3) and the equation of motion (2.9), or from solving for the stationarity condition $\delta\mathcal{A}/\delta\hat{b} = 0$ for the action $\mathcal{A} = \mathcal{A}_B + \mathcal{A}_R$, Eqs. (2.10) and (2.11), setting $D = 0$. (Notice that $\delta\mathcal{A}/\delta b = 0$ is always solved by $\hat{b} = 1$.) Either procedure results in the obvious mean-field rate equation

$$\partial_t \rho_B(t) = -n\lambda_n \rho_B(t)^n , \qquad (4.1)$$

which is readily integrated for $n > 1$,

$$\rho_B(t) = \frac{\rho_B(0)}{(1 + t/\tau)^{1/(n-1)}} , \quad \tau = \frac{\rho_B(0)^{1-n}}{n(n-1)\lambda_n} , \quad (4.2)$$

i.e., for $t \gg \tau$ the B density decays algebraically $\propto t^{-1/(n-1)}$ in this approximation, while of course for $n = 1$

$$\rho_B(t) = \rho_B(0)\, e^{-\lambda_1 t} . \qquad (4.3)$$

In the same manner, we may obtain the evolution equation for the A species, or just consider Eq. (3.2) for $D' = 0$ and vanishing noise. In the spirit of mean-field theory, we assume a homogeneous B density, and obtain

$$\partial_t \rho_A(\vec{x}, t) = \widetilde{D}\,\rho_B(t)\,\nabla^2 \rho_A(\vec{x}, t) . \qquad (4.4)$$

Again, this equation can be solved exactly, considering a delta-like density distribution for the A species at the initial time $t = 0$. As in this mean-field approach the $\rho_B(t)$ is assumed to be spatially uniform, the A species will be Gaussian distributed in space, just like in ordinary diffusion,

$$\rho_A(\vec{x}, t) = \left(\frac{1}{2\pi \langle \vec{x}_A^2(t)\rangle}\right)^{d/2} \exp\left(-\frac{\vec{x}^2}{2\langle \vec{x}_A^2(t)\rangle}\right) . \quad (4.5)$$

However, the B decay (or lattice depletion) will be reflected in the anomalous time-dependence of the width

(mean-square displacement). A straightforward brief calculation yields

$$\langle \vec{x}_A^2(t)\rangle = 2\widetilde{D} \int_0^t \rho_B(t')\, dt' . \qquad (4.6)$$

For $n = 1$, i.e., the simple exponential decay (4.3), the result is

$$\langle \vec{x}_A^2(t)\rangle = \frac{2\widetilde{D}\,\rho_B(0)}{\lambda_1}\left(1 - e^{-\lambda_1 t}\right) . \qquad (4.7)$$

Initially ($\lambda_1 t \ll 1$) one finds normal diffusion with effective diffusion constant $\bar{D} = \widetilde{D}\,\rho_B(0)$, but at long times the mean-square displacement approaches a constant, and the A particles remain localized in a region of volume $\propto \langle \vec{x}_A^2(t \to \infty)\rangle^{d/2} = (2\widetilde{D}\,\rho_B(0)/\lambda_1)^{d/2}$. Given that this simple process is characterized by short-range correlations in space and time only, we do not expect any considerable modification through fluctuation effects.

In the pair annihilation case, $n = 2$, one finds

$$\langle \vec{x}_A^2(t)\rangle = 2\widetilde{D}\,\rho_B(0)\, \ln\left(1 + \frac{t}{\tau}\right) , \qquad (4.8)$$

while the mean-field result for $n > 2$ reads

$$\langle \vec{x}_A^2(t)\rangle = 2\widetilde{D}\,\rho_B(0)\,\frac{n-1}{n-2}\,\tau\left[\left(1 + \frac{t}{\tau}\right)^{\frac{n-2}{n-1}} - 1\right] . \quad (4.9)$$

In the asymptotic regime $t \gg \tau$, this implies anomalous diffusion according to

$$\langle \vec{x}_A^2(t)\rangle \propto t^{2/(2+\Theta)} \qquad (4.10)$$

with a positive exponent $\Theta = 2/(n-2)$ indicating *sub-diffusive* behavior. In the limit $n \to \infty$ we have $\Theta \to 0$, and conventional diffusion is recovered. The reason is of course that for large n the depleting reactions become very unlikely, as n particles are required to meet at the same lattice site. Thus, low-order B species reactions are much more effective in slowing down the A diffusion. The time scale for the crossover to the pure algebraic decay of the B particle density and subsequently for the anomalous A diffusion is given by $\tau \propto \rho_B(0)^{1-n}/\lambda_n$. The crossover to the asymptotic slow dynamics is fast for large initial densities and reaction rates.

The above analysis should be qualitatively correct for $n > 3$, as the corresponding critical dimension $d_c(n) < 1$. For $n = 2$, i.e., B pair annihilation processes in $d \leq 2$ dimensions, we know that at long times *anti*correlations develop [20,18,17]: Initially close-by particles disappear quickly, and only widely separated ones survive. This effective "repulsion" should result in a roughly uniform spatial B distribution even for a clustered initial configuration. Given that the coupling coefficient $\widetilde{D}$ itself does not renormalize, we therefore expect that our decoupling assumption leading to Eq. (4.4) should represent a fair approximation, provided the correct time dependence of

the B density is inserted. For $d < 2$, the asymptotic result is

$$\rho_B(t) \propto t^{-d/2} \ , \tag{4.11}$$

see Ref. [18] and also Sec. III following Eq. (3.8), whence

$$\langle \vec{x}_A^2(t) \rangle = 2\bar{D}\,t^{1-d/2} \tag{4.12}$$

with an appropriate effective rate $\bar{D} \propto \widetilde{D}/(1-d/2)$. In low dimensions, this algebraic subdiffusive behavior with $\Theta = 2d/(2-d)$ replaces the logarithmic law (4.8). At the critical dimensions $d_c(2) = 2$, one finds the typical logarithmic corrections [18]

$$\rho_B(t) \propto t^{-1} \ln t \ , \tag{4.13}$$

implying

$$\langle \vec{x}_A^2(t) \rangle \propto \widetilde{D}\,(\ln t)^2 \ , \tag{4.14}$$

which also describes slower kinetics than given by the mean-field result (4.8). For the case of $n = 3$ at its critical dimension $d_c(3) = 1$,

$$\rho_B(t) \propto (t^{-1}\,\ln t)^{1/2} \ , \tag{4.15}$$

and one would therefore expect the leading time dependence

$$\langle \vec{x}_A^2(t) \rangle \propto \widetilde{D}\,(t\,\ln t)^{1/2} \ , \tag{4.16}$$

i.e., essentially a square-root power law with logarithmic corrections.

B. BARW kinetics

We now extend the B dynamics and include branching processes of the form $B \to (m+1)\,B$ with rate σ_m, described by Eqs. (2.4) or (2.12). The mean-field rate equation (4.1), with $n \geq 2$, is then replaced by

$$\partial_t \rho_B(t) = -n\lambda_n\,\rho_B(t)^n + m\sigma_m\,\rho_B(t) \ , \tag{4.17}$$

which has two stationary solutions $\rho_B = 0$ (inactive phase) and

$$\rho_B^\infty = \left(\frac{m\sigma_m}{n\lambda_n} \right)^{1/(n-1)} \tag{4.18}$$

(active phase). For any $\sigma_m > 0$, the latter turns out to be stable, i.e., BARW are always in the active phase in the mean-field approximation. The explicit solution of Eq. (4.18) furthermore shows that the asymptotic density ρ_B^∞ is exponentially approached,

$$\rho_B(t) = \frac{\rho_B^\infty}{\left[1 + C\,e^{-(n-1)m\sigma_m t} \right]^{1/(n-1)}} \ , \tag{4.19}$$

where $C = [\rho_B^\infty/\rho_B(0)]^{n-1} - 1$. Again, Eq. (4.4) is solved by the Gaussian distribution (4.5) with mean-square displacement (4.6). The ensuing integral is readily calculated for some special cases, e.g., for $n = 2$

$$\langle \vec{x}_A^2(t) \rangle = 2\widetilde{D}\,\rho_B^\infty \left[t + \frac{1}{m\sigma_m}\ln\left(1 + C\,e^{-m\sigma_m t}\right) \right] \ , \tag{4.20}$$

whereas for $n = 3$

$$\langle \vec{x}_A^2(t) \rangle = \frac{\widetilde{D}\rho_B^\infty}{m\sigma_m}\,\ln\left(\frac{\sqrt{1 + C\,e^{-2m\sigma_m t}} + 1}{\sqrt{1 + C\,e^{-2m\sigma_m t}} - 1} \right) \ . \tag{4.21}$$

In general, asymptotically normal diffusion with effective diffusion coefficient $\widetilde{D}\,\rho_B^\infty$ is recovered in the active state,

$$\langle \vec{x}_A^2(t) \rangle = 2\widetilde{D}\,\rho_B^\infty\,t \ . \tag{4.22}$$

The properties of the active phase with an asymptotically homogeneous B density are not much influenced by fluctuations, and hence Eq. (4.22) should aptly describe the ensuing A kinetics even beyond mean-field theory.

For the possible existence of an inactive phase, and the characterization of the ensuing critical behavior, fluctuation effects are however of utmost importance for $n = 2$, and it turns out that the cases of odd and even offspring number m need to be distinguished. For odd m, aside from all lower-order branchings, first-order decay processes $B \to \emptyset$ are generated, and become sufficiently efficient to shift the critical point to $\sigma_c > 0$ for $d \leq 2$ dimensions. The emerging transition at $\sigma_c > 0$ can be shown to be in the generic directed-percolation (DP) universality class [19]. The inactive phase is then governed by exponential B density decay, whereupon the A species will become localized according to Eq. (4.7). At the critical point itself, the B species density decays according to a power law $\rho_B(\vec{x},t) \sim t^{-\alpha}$, with $\alpha = \beta/z\nu_\perp$ given by DP critical exponents in $d = 1$ and $d = 2$, respectively. This would suggest $\langle \vec{x}_A^2(t) \rangle \propto t^{1-\alpha}$; yet the B density is far from uniform at the critical point, and is instead characterized by the appearance of *fractal* density clusters. While we would still expect subdiffusive behavior for the A species with $\Theta > 0$, this exponent will likely be influenced by the power-law correlations in the critical B density. In the case of even m, for which the B particle number parity is locally conserved under the reactions, a non-trivial transition with $\sigma_c > 0$ is possible only for $d \leq d_c' \approx 4/3$ dimensions. The inactive phase is then given by the pure pair annihilation theory, and consequently Eq. (4.12) should provide a fair description for the ensuing anomalous A diffusion. The critical behavior is governed by a different parity-conserving universality class, with $\alpha < 1/2$. In this instance, we again expect the above mean-field description to be rather inaccurate.

V. CONCLUSIONS

We have studied a novel mechanism to induce anomalous diffusion. Whenever an active particle of the A species performs a random walk on a lattice, it may visit a certain lattice site only provided this site is already occupied by at least one B particle. The random walk is prolonged when the B particles react with each other in such a manner that the B species density is decreasing. If that decay is exponential (first-order reaction), then after a short time interval (given by the inverse decay rate) the B species has disappeared and a further visit of an A particle at that site is impossible. As a consequence the A species, after some initial mixing, remains localized. When the B species undergo reactions of higher order, requiring at least two B particles to meet at a lattice site, an algebraic decay ensues that allows hopping processes for the A species to occur for a much longer period. However, the random walk process is slowed down considerably as the B density diminishes, resulting in a much shorter mean-square displacement of A particles as compared with conventional diffusion. The emerging anomalous diffusion is governed by power laws or logarithmic behavior that can (approximately) be related to the asymptotic time behavior of the reacting B particle density. In this instance one may view this process as resembling diffusion on a dynamical fractal. Only when at long times the B density remains finite and nearly homogeneous, conventional A diffusion is recovered. This situation corresponds to diffusion with dynamical disorder, where in the long-time limit the B particles, with largely decayed fluctuations, merely resemble a quasi-static inhomogeneous background for the A kinetics. The consistent mathematical treatment of diffusion on a static fractal, as well as induced diffusion processes on critical (isotropic or directed) percolation clusters or near BARW critical points remains an open problem that requires more sophisticated analysis beyond the largely mean-field approach presented here.

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