Scale Invariance and Dynamic Phase Transitions in Diffusion-Limited Reactions

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Abstract. Many systems that can be described in terms of diffusion-limited 'chemical' reactions display non-equilibrium continuous transitions separating active from inactive, absorbing states, where stochastic fluctuations cease entirely. Their critical properties can be analyzed via a path-integral representation of the corresponding classical master equation, and the dynamical renormalization group. An overview over the ensuing universality classes in single-species processes is given, and generalizations to reactions with multiple particle species are discussed as well. The generic case is represented by the processes $A \rightleftharpoons A+A$, and $A \to \emptyset$, which map onto Reggeon field theory with the critical exponents of directed percolation (DP). For branching and annihilating random walks (BARW) $A \to (m+1)A$ and $A+A \to \emptyset$, the mean-field rate equation predicts an active state only. Yet BARW with odd m display a DP transition for $d \leq 2$. For even offspring number m, the particle number parity is conserved locally. Below $d_c' \approx 4/3$, this leads to the emergence of an inactive phase that is characterized by the power laws of the pair annihilation process. The critical exponents at the transition are those of the 'parity-conserving' (PC) universality class. For local processes without memory, competing pair or triplet annihilation and fission reactions $kA \to (k-l)A$, $kA \to (k+m)A$ with $k = 2, 3$ appear to yield the only other universality classes not described by mean-field theory. In these reactions, site occupation number restrictions play a crucial role.

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1 Introduction: Active to absorbing state transitions

The characterization of non-equilibrium steady states constitutes one of the prevalent goals in present statistical mechanics. Unfortunately, away from thermal equilibrium one cannot in general derive even stationary macroscopic properties from an effective free energy function. One might hope, however, that such a classification in terms of symmetries and interactions becomes feasible near continuous phase transitions separating different non-equilibrium steady states: Drawing on the analogy with equilibrium critical points, one would expect certain features of non-equilibrium phase transitions to be *universal* as well, i.e., independent of the detailed microscopic dynamical rules and even the initial conditions. The emerging power laws and scaling functions describing the long-wavelength, long-time limit should then hopefully be characterized by not too many distinct dynamic universality classes. Yet studies of a variety of non-equilibrium processes have taught us that critical phenomena as well as generic scale invariance far from thermal equilibrium, where restrictive detailed-balance constraints do not apply, are often considerably richer than their equilibrium counterparts. Indeed, intuitions inferred from the latter have frequently turned out to be quite deceptive.

But as in the investigation of equilibrium critical phenomena, field theory representations supplemented with the dynamical version of the renormalization group (RG) provide powerful methods to extract and systematically classify universal properties at continuous non-equilibrium phase transitions. Additional indispensable quantitative tools are of course Monte Carlo simulations, and other numerical approaches and exact solutions when available (the latter are usually restricted to specific one-dimensional systems). Analytical and numerical methods usually supplement each other: Simulation results often call for deeper understanding of the underlying processes, but also rely on some theoretical background as a basis for data analysis.

A prominent class of genuine non-equilibrium phase transitions separates *'active'* from *'inactive, absorbing'* stationary states where, owing to the absence of any agents, stochastic fluctuations cease entirely [1,2]. These occur in a variety of systems in nature; e.g., in chemical reactions which involve an inert state $\emptyset$ that does not release the reactants A anymore. Another example are models for stochastic population dynamics, combining, say, diffusive migration of a species A with asexual reproduction $A \to 2A$ (with rate σ), spontaneous death $A \to \emptyset$ (at rate μ), and lethal competition $2A \to A$ (with rate λ). In the inactive state, where no population members A are left, clearly all processes terminate. Similar effective dynamics may be used to model certain non-equilibrium physical systems. Consider for instance the domain wall kinetics in Ising chains with competing Glauber and Kawasaki dynamics [3]. Here, spin flips $\uparrow\uparrow\downarrow\downarrow \to \uparrow\uparrow\uparrow\downarrow$ and $\uparrow\uparrow\downarrow\uparrow \to \uparrow\uparrow\uparrow\uparrow$ may be viewed as domain wall (A) hopping and pair annihilation $2A \to \emptyset$, whereas spin exchange $\uparrow\uparrow\downarrow\downarrow \to \uparrow\downarrow\uparrow\downarrow$ represents a branching process $A \to 3A$. Notice that the para- and ferromagnetic phases respectively map onto the active and inactive 'particle' states, the latter rendered absorbing if the spin flip rates are computed at zero temperature, thus forbidding any energy increase.

The simplest mathematical description for such processes uses kinetic rate equations, which govern the time evolution of the mean 'particle' density $n(t)$. For example, the above population model leads to Fisher's rate equation

$$\partial_t \, n(t) = (\sigma - \mu) \, n(t) - \lambda \, n(t)^2 \ . \tag{1}$$

It yields both inactive and active phases: For $\sigma < \mu$ we have $n(t \to \infty) \to 0$, whereas for $\sigma > \mu$ the density eventually saturates at $n_s = (\sigma - \mu)/\lambda$. The explicit solution (with initial particle density n_0)

$$n(t) = n_0 \, n_s \Big/ \left[n_0 + (n_s - n_0) \, e^{(\mu - \sigma) t} \right] \tag{2}$$

shows that both stationary states are approached exponentially in time. The two phases are separated by a continuous non-equilibrium phase transition at

$\sigma = \mu$, where the temporal decay becomes algebraic, $n(t) = n_0/(1+n_0\lambda\, t) \to 1/(\lambda t)$ as $t \to \infty$, independent of the initial density. But Eq. (1) represents a mean-field approximation, as we have in fact replaced the joint probability of finding two particles at the same position with the square of the mean density. As in equilibrium, however, critical fluctuations are expected to invalidate simple mean-field theory in sufficiently low dimensions $d < d_c$, which defines the upper critical dimension. A more satisfactory treatment therefore necessitates a systematic incorporation of spatio-temporal fluctuations, including specifically the particle correlations as induced by the dynamics.

2 From the master equation to stochastic field theory

The renormalization group study of (near-)*equilibrium* dynamical critical phenomena relies on phenomenological Langevin equations for the order parameter and 'slow' hydrodynamic variables associated with conservation laws [4]. All other degrees of freedom are treated as Gaussian white noise, whose second moment is related to the relaxation coefficients through Einstein's fluctuation-dissipation theorem. As we shall see, however, such a description is not necessarily possible at all in reaction-diffusion systems. To the very least one would have to invoke fundamental conjectures on the noise correlators; but far from equilibrium these often crucially influence long-wavelength properties. It is therefore desirable to construct a long-wavelength effective theory for stochastic processes directly from their microscopic definition, without recourse to any serious additional assumptions or approximations.

Fortunately, there exists an established procedure to derive the Liouville time evolution operator for locally interacting particle systems immediately from the classical master equation, wherefrom a field theory representation is readily obtained [5]. The key point is that all possible configurations can be labeled by specifying the occupation numbers n_i of, say, the sites of a d-dimensional lattice. Let us for now assume that there are no site occupation restrictions, i.e., any $n_i = 0, 1, 2, \ldots$ is allowed (we shall address effects of particle exclusions in Sec. 7). The master equation governs the time evolution of the configurational probability $P(\{n_i\}; t)$. For example, the corresponding contribution from the binary coagulation process $2A \to A$ at site i reads

$$\partial_t P(n_i; t)|_\lambda = \lambda \Big[(n_i + 1) n_i P(n_i + 1; t) - n_i (n_i - 1) P(n_i; t) \Big] \ . \qquad (3)$$

This sole dependence on the integer variables $\{n_i\}$ calls for a representation in terms of bosonic ladder operators with the standard commutation relations $[a_i, a_j^\dagger] = \delta_{ij}$, and the empty state $|0\rangle$ such that $a_i|0\rangle = 0$. We next define the Fock states via $|\{n_i\}\rangle = \prod_i (a_i^\dagger)^{n_i} |0\rangle$ (notice the different normalization from standard quantum mechanics), and thence construct the formal *state vector*

$$|\Phi(t)\rangle = \sum_{\{n_i\}} P(\{n_i\}; t) \ |\{n_i\}\rangle \ . \qquad (4)$$

The linear time evolution imposed by the master equation can be cast into an 'imaginary-time Schrödinger' equation $\partial_t |\Phi(t)\rangle = -H\,|\Phi(t)\rangle$ with a generally non-Hermitian, local 'Hamiltonian' $H(\{a_i^\dagger\},\{a_i\})$. For instance, the on-site coagulation reaction is encoded in this formalism via $H_i^\lambda = -\lambda\,(1-a_i^\dagger)\,a_i^\dagger a_i^2$.

Our goal really is to evaluate time-dependent statistical averages for observables F, necessarily also just functions of the occupation numbers, $\langle F(t)\rangle = \sum_{\{n_i\}} F(\{n_i\})\,P(\{n_i\};t)$. Straightforward algebra using the identity $[e^a, a^\dagger] = e^a$ shows that this average can be written as a 'matrix element'

$$\langle F(t)\rangle = \langle \mathcal{P}|\,F(\{a_i\})\,|\Phi(t)\rangle = \langle \mathcal{P}|\,F(\{a_i\})\,e^{-Ht}\,|\Phi(0)\rangle \tag{5}$$

with the state vector $|\Phi(t)\rangle$ and the projector state $\langle\mathcal{P}| = \langle 0|\prod_i e^{a_i}$; notice that $\langle\mathcal{P}|0\rangle = 1$. Probability conservation implies $1 = \langle\mathcal{P}|\,e^{-Ht}\,|\Phi(0)\rangle$, i.e., for infinitesimal times $\langle\mathcal{P}|\Phi(0)\rangle = 1$ and $\langle\mathcal{P}|H = 0$, which is satisfied provided $H(\{a_i^\dagger \to 1\},\{a_i\}) = 0$. We remark that commuting the factors e^{a_i} to the right has the effect of shifting all $a_i^\dagger \to 1 + a_i^\dagger$. One may then emply coherent states, as familiar from quantum many-particle theory [6], to represent the matrix element (5) as a *functional integral* with statistical weight $\exp(-S)$. Omitting contributions from the initial state, the action becomes

$$S[\{\hat\psi_i\},\{\psi_i\}] = \int dt\left[\sum_i \hat\psi_i\,\partial_t\psi_i + H(\{\hat\psi_i\},\{\psi_i\})\right] . \tag{6}$$

Taking the continuum limit finally yields the desired field theory that faithfully encodes all stochastic fluctuations.

3 Reggeon field theory and directed percolation (DP)

Let us now return to our population dynamics example with random walkers A (with diffusion constant D in the continuum limit), subject to the Gribov reactions $A \rightleftharpoons A + A$ and $A \to \emptyset$. The corresponding field theory (6) reads

$$S = \int d^dx\,dt\Big[\hat\psi(\partial_t - D\nabla^2)\psi + \sigma(1-\hat\psi)\hat\psi\psi - \mu(1-\hat\psi)\psi - \lambda(1-\hat\psi)\hat\psi\psi^2\Big] . \tag{7}$$

The stationarity condition $\delta S/\delta\psi = 0$ is always solved by $\hat\psi = 1$; upon inserting this into $\delta S/\delta\hat\psi = 0$, and identifying $n(t) = \langle\psi(\boldsymbol{x},t)\rangle$, one recovers Fisher's mean-field rate equation (1). Higher moments of the field ψ, however, cannot be immediately connected with density correlation functions. In terms of an arbitrary momentum scale κ, we record the naive scaling dimensions $[\hat\psi] = \kappa^0$, $[\psi] = \kappa^d$, $[\sigma] = \kappa^2 = [\mu]$, and $[\lambda] = \kappa^{2-d}$. Hence the decay and branching rates constitute relevant operators in the RG sense, whereas the annihilation process becomes marginal at $d = 2$. Next we expand the action (7) about the stationary solution $\hat\psi = 1$, i.e., introduce $\tilde\psi(\boldsymbol{x},t) = \hat\psi(\boldsymbol{x},t) - 1$, whereupon we arrive at

$$S = \int d^dx\,dt\Big[\tilde\psi(\partial_t - D\nabla^2)\psi + (\mu-\sigma)\tilde\psi\psi - \sigma\,\tilde\psi^2\psi + \lambda\,\tilde\psi(1+\tilde\psi)\psi^2\Big] . \tag{8}$$

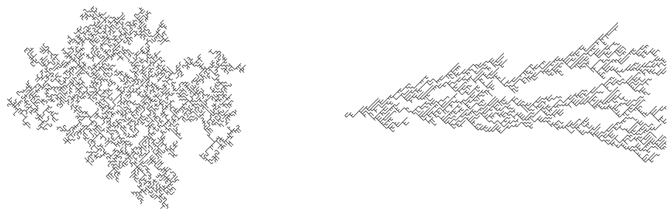

Fig. 1. Critical isotropic (a) and directed (b) percolation clusters (from Ref. [10])

Inspection of the one-loop fluctuation corrections shows that the effective coupling is in fact $u = \sqrt{\sigma\lambda}$, with scaling dimension $[u] = \kappa^{2-d/2}$, whence $d_c = 4$. At least in the vicinity of d_c, λ becomes irrelevant; certainly the ratio λ/u scales to zero under subsequent RG transformations. Simple rescaling $\tilde{\psi} = \tilde{\phi}\sqrt{\lambda/\sigma}$, $\psi = \phi\sqrt{\sigma/\lambda}$ then leaves us with the *effective* field theory

$$S_{\text{eff}}[\tilde{\phi}, \phi] = \int d^d x \, dt \left[\tilde{\phi} \left[\partial_t + D(r - \nabla^2) \right] \phi + u \left(\tilde{\phi} \, \phi^2 - \tilde{\phi}^2 \, \phi \right) \right] , \tag{9}$$

where $r = (\mu - \sigma)/D$. This should capture the critical behavior for the non-equilibrium phase transition at $r = 0$ in our population dynamics model.

The action (9) is known in particle physics as *'Reggeon' field theory* [7]. It is invariant with respect to 'rapidity inversion' $\phi(\boldsymbol{x}, t) \to -\tilde{\phi}(\boldsymbol{x}, -t)$, $\tilde{\phi}(\boldsymbol{x}, t) \to -\phi(\boldsymbol{x}, -t)$. Quite remarkably, the very same action is obtained for the threshold pair correlation function [8] in the geometric problem of *directed percolation* (DP) [9]. Fig. 1(b) depicts a critical directed percolation cluster, contrasted with the structure emerging at the threshold of ordinary isotropic percolation. At $d_c = 4$, the critical exponents as predicted by mean-field theory acquire logarithmic corrections, and are shifted to different values by the infrared-singular fluctuations in $d < 4$ dimensions. By means of the standard perturbational loop expansion in terms of the diffusion propagator and the vertices $\propto u$, and the application of the RG, the critical exponents can be computed systematically and in a controlled manner in a dimensional expansion with respect to $\epsilon = 4 - d$. The one-loop results, to first order in ϵ, as well as reliable values from Monte Carlo simulations in one and two dimensions [2] are listed in Table 1. Moreover, as a consequence of rapidity invariance there are only three independent scaling exponents, namely the anomalous field dimension η, the correlation length exponent ν, and the dynamic critical exponent z. All other exponents are then fixed by scaling relations, such as

$$\beta = \frac{\nu}{2} \left(z + d - 2 + \eta \right) = z \, \nu \, \alpha \tag{10}$$

for the order parameter and critical density decay exponents.

It is worthwhile noting that Reggeon field theory can be viewed as a dynamic response functional [11], and therefore is equivalent to an effective

Table 1. Critical exponents for the saturation density n_s, correlation length ξ, time scale t_c, and critical density decay $n_c(t)$ for the DP universality class

DP exponents	$d = 1$	$d = 2$	$d = 4 - \epsilon$
$n_s \sim \vert r\vert^\beta$	$\beta \approx 0.2765$	$\beta \approx 0.584$	$\beta = 1 - \epsilon/6 + O(\epsilon^2)$
$\xi \sim \vert r\vert^{-\nu}$	$\nu \approx 1.100$	$\nu \approx 0.735$	$\nu = 1/2 + \epsilon/16 + O(\epsilon^2)$
$t_c \sim \xi^z \sim \vert r\vert^{-z\nu}$	$z \approx 1.576$	$z \approx 1.73$	$z = 2 - \epsilon/12 + O(\epsilon^2)$
$n_c(t) \sim t^{-\alpha}$	$\alpha \approx 0.160$	$\alpha \approx 0.46$	$\alpha = 1 - \epsilon/4 + O(\epsilon^2)$

Langevin equation. To this end, we integrate out the field $\tilde{\phi}$, which yields the statistical weight $\exp(-G)$ with

$$G[\phi] = \int d^d x \, dt \left[\partial_t \phi + D(r - \nabla^2)\phi + u\,\phi^2\right]^2 \Big/ 4\,u\,\phi \ . \tag{11}$$

After rescaling, we may interpret (11) as the Onsager-Machlup functional associated with the Gaussian noise distribution for the stochastic process

$$\partial_t \, n(\boldsymbol{x}, t) = D(\nabla^2 - r)\, n(\boldsymbol{x}, t) - \lambda\, n(\boldsymbol{x}, t)^2 + \zeta(\boldsymbol{x}, t) \ , \tag{12a}$$

$$\langle \zeta(\boldsymbol{x}, t) \rangle = 0 \, , \ \langle \zeta(\boldsymbol{x}, t)\, \zeta(\boldsymbol{x}', t') \rangle = 2\,\sigma\, n(\boldsymbol{x}, t)\, \delta(\boldsymbol{x} - \boldsymbol{x}')\, \delta(t - t') \ . \tag{12b}$$

Here, the absorbing nature of the inactive state is reflected in the fact that the noise correlator is proportional to $n(\boldsymbol{x}, t)$. Of course, Eq. (12b) really means that the local density is to be factored in when the noise average is taken, c.f. Eq. (11). Alternatively, One may define $\zeta(\boldsymbol{x}, t) = \sqrt{n(\boldsymbol{x}, t)}\, \eta(\boldsymbol{x}, t)$, whereupon $\langle \eta(\boldsymbol{x}, t)\, \eta(\boldsymbol{x}', t') \rangle = 2\,\sigma\, \delta(\boldsymbol{x} - \boldsymbol{x}')\, \delta(t - t')$, at the cost of introducing 'square-root' multiplicative noise into the Langevin equation (12a). Within the Langevin framework, we can readily generalize to arbitrary reaction and noise functionals $r[n]$ and $c[n]$:

$$\partial_t \, n(\boldsymbol{x}, t) = D\, \nabla^2 \, n(\boldsymbol{x}, t) - r[n(\boldsymbol{x}, t)] + \zeta(\boldsymbol{x}, t) \ , \tag{13a}$$

$$\langle \zeta(\boldsymbol{x}, t) \rangle = 0 \, , \ \langle \zeta(\boldsymbol{x}, t)\, \zeta(\boldsymbol{x}', t') \rangle = c[n(\boldsymbol{x}, t)]\, \delta(\boldsymbol{x} - \boldsymbol{x}')\, \delta(t - t') \ . \tag{13b}$$

In the spirit of Landau theory, we may then expand the functionals $r[n]$ and $c[n]$ near the inactive phase ($n \ll 1$). In the absence of spontaneous particle production, both must vanish at $n = 0$, which is the condition for an absorbing state. Keeping only the lowest-order, relevant terms in the expansions with respect to n, we thereby infer that any active to absorbing state phase transition in a single-species system should *generically* be described by Eqs. (12a) and (12b), i.e., Reggeon field theory (9). Consequently, in the absence of any special symmetries, memory effects, and quenched disorder, we expect to find the critical exponents of the DP universality class [12].

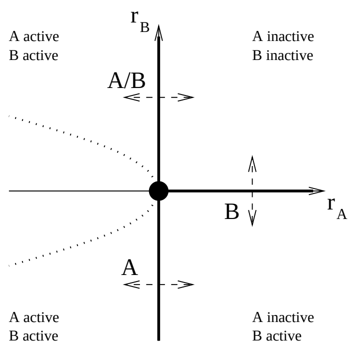

Fig. 2. Mean-field phase diagram for the two-stage unidirectionally coupled DP process. The arrows indicate active to absorbing transitions for the A and B species. The dotted parabola marks the boundary of the multi-critical regime [14]

4 Variants of directed percolation processes

In fact, DP-type processes with even arbitrarily many particle species have been fully classified. Consider the reactions $A \rightleftharpoons A + A$, $A \to \emptyset$, $B \rightleftharpoons B + B$, $B \to \emptyset$, etc., supplemented with bilinear couplings $A \to B + B$, $A + A \to B, \ldots$ Higher-order reactions then turn out to be irrelevant in the RG sense, and remarkably the critical behavior of the multi-species DP system is once again governed by the DP fixed point [13]. However, if we allow for unidirectional *linear* couplings through particle transmutations $A \to B$, $B \to C, \ldots$, with rates $\mu_{AB}, \mu_{BC}, \ldots$, *multi-critical* behavior may ensue [14]. This becomes already manifest on the level of the coupled rate equations

$$\partial_t \, n_A = D \left(\nabla^2 - r_A \right) n_A - \lambda_A \, n_A^2 \; , \tag{14a}$$

$$\partial_t \, n_B = D \left(\nabla^2 - r_B \right) n_B - \lambda_B \, n_B^2 + \mu_{AB} \, n_A \; , \tag{14b}$$

etc. For as long as the A species is in the active phase ($r_A < 0$), the B particle density will be non-zero as well. As depicted in Fig. 2, this effectively 'folds' half of the decoupled B transition line ($r_A < 0$, $r_B = 0$) over onto $r_A = 0$, $r_B > 0$. Along this half-line, the B particles become 'enslaved' by the A species; and so forth further down the hierarchy of particle species. As $r_A \to 0$ and $r_B \to 0$ simultaneously, one encounters a non-equilibrium multi-critical point. While the DP exponents ν and z governing the correlation length and critical slowing down remain unchanged, one finds successively reduced values for the order parameter exponents $\beta^{(j)}$ on the jth hierarchy level [14], as listed in Table 2. The crossover exponent associated with the multi-critical point is $\phi = 1$ to all orders in the ϵ expansion [13].

Another mechanism to induce a different universality class in a two-species system is to link diffusing agents A with a passive, spatially fixed, and initially homogeneously distributed species X through the reactions $X + A \to A + A$ and $A \to \emptyset$. One may then integrate out the X fluctuations; upon expanding

Table 2. Simulation and one-loop RG results for the saturation density critical exponents on the first three hierarchy levels in unidirectionally coupled DP processes

	$d=1$	$d=2$	$d=3$	$d=4-\epsilon$
$\beta^{(1)}$	0.280(5)	0.57(2)	0.80(4)	$1-\epsilon/6+O(\epsilon^2)$
$\beta^{(2)}$	0.132(15)	0.32(3)	0.40(3)	$1/2-\epsilon/8+O(\epsilon^2)$
$\beta^{(3)}$	0.045(10)	0.15(3)	0.17(2)	$1/4-O(\epsilon)$

about the mean-field solution, the resulting effective action becomes [15]

$$S_{\rm eff}[\tilde{\phi}, \phi] = \int d^d x \, dt \Big[\tilde{\phi}(\boldsymbol{x}, t) \left[\partial_t + D(r - \nabla^2) \right] \phi(\boldsymbol{x}, t) \\ + 2D \, u \, \tilde{\phi}(\boldsymbol{x}, t) \, \phi(\boldsymbol{x}, t) \int^t \phi(\boldsymbol{x}, t') \, dt' - u \, \tilde{\phi}(\boldsymbol{x}, t)^2 \, \phi(\boldsymbol{x}, t) \Big] \, . \quad (15)$$

The elimination of the passive particles X thus induces memory of all preceding times in the particle annihilation vertex. The DP rapidity inversion invariance is replaced by its non-local counterpart $\phi(\boldsymbol{x}, t) \to -D^{-1} \, \partial_t \tilde{\phi}(\boldsymbol{x}, -t)$, $\tilde{\phi}(\boldsymbol{x}, t) \to -D \int^{-t} \phi(\boldsymbol{x}, t') \, dt'$. The upper critical dimension for this *dynamic percolation* universality class is shifted to $d_c = 6$. Its static critical exponents are precisely the ones that characterize a critical *isotropic percolation* cluster [15], compare Fig. 1(a). To first order in $\epsilon = 6 - d$, one finds $\eta = -\epsilon/21$, $\nu = 1/2 + 5\epsilon/84$, $z = 2 - \epsilon/6$, and $\beta = \nu(d - 2 + \eta)/2 = 1 - \epsilon/7$. Multi-species generalizations proceed in the same way as for DP, with similar results: Whereas non-linear couplings to other particle species preserve the dynamic universality class, a multi-critical point emerges for unidirectional particle transmutations, with crossover exponent $\phi = 1$ [13].

5 Diffusion-limited annihilation processes

As a preparation for the following Sec. 6, let us now investigate the kth order *annihilation* reaction $kA \to \emptyset$. The associated rate equation reads $\partial_t \, n(t) = -\lambda \, n(t)^k$. For radioactive decay ($k = 1$), it is naturally solved by the familiar exponential $n(t) = n_0 \, e^{-\lambda t}$, whereas one obtains power laws for $k \geq 2$, namely

$$n(t) = \left[n_0^{1-k} + (k-1) \, \lambda \, t \right]^{-1/(k-1)} \, . \quad (16)$$

In order to consistently include fluctuations in the latter case, we again start out from the master equation, wherefrom we derive the action [16]

$$S[\hat{\psi}, \psi] = \int d^d x \, dt \left[\hat{\psi} \, (\partial_t - D \, \nabla^2) \, \psi - \lambda \, (1 - \hat{\psi}^k) \, \psi^k \right] \, . \quad (17)$$

After performing the shift $\hat{\psi}(\boldsymbol{x}, t) = 1 + \tilde{\psi}(\boldsymbol{x}, t)$, it becomes evident that this field theory does *not* have a simple Langevin representation. For in order to interpret $\tilde{\psi}$ as the corresponding noise auxiliary field, it should appear quadratically in the action only, and with negative prefactor. Thus even for the pair annihilation process, the Langevin equation derived from the action (17) would entail unphysical 'imaginary' noise with $c[n] = -2\,\lambda\, n^2$.

The scaling dimension of the annihilation vertex is $[\lambda_k] = \kappa^{2-(k-1)d}$, whence we infer the upper critical dimension $d_c(k) = 2/(k-1)$. This leaves the possibility of non-trivial scaling behavior in low physical dimensions only for the pair ($k = 2$) and triplet ($k = 3$) processes. Analyzing the field theory (17) further, we see that the diffusion propagator does not become renormalized at all. Consequently, $\eta = 0$ and $z = 2$ to all orders in the perturbation expansion. The simple structure of the action permits summing the entire vertex renormalization perturbation series by means of a Bethe-Salpeter equation; in Fourier space it reduces to a geometric series of the one-loop diagram [16].

For pair annihilation, this yields the following asymptotic behavior for the particle density: $n(t) \sim (\lambda\, t)^{-1}$, i.e., the reaction-limited power law of the rate equation for $d > 2$; but diffusion-limited decay $n(t) \sim (Dt)^{-d/2}$ for $d < 2$. At $d_c = 2$, one finds the logarithmic correction $n(t) \sim (Dt)^{-1} \ln Dt$. The slower decay for $d \leq 2$ originates in the fast mutual annihilation of any close-by reactants; after some time has elapsed, only well-separated particles are left. The annihilation dynamics thus produces *anti*-correlations, mimicking an effective repulsive interaction (which actually provides the interpretation for the negative correlator $c[n]$). In the ensuing *diffusion-limited* regime, the typical particle separation scales as $\ell(t) \sim (Dt)^{-1/2}$, whereupon indeed $n(t) \propto \ell(t)^{-d} \sim (Dt)^{-d/2}$. The same power laws hold for the pair coagulation process $2A \to A$, albeit with different amplitudes. Replacing ordinary diffusion with long-range Lévy flights with probability $\propto r^{-d-\rho}$ of hopping a distance r ($\rho < 2$) results in $n(t) \sim (Dt)^{-d/\rho}$ for $d < d_c = \rho$ [17]. For triplet annihilation, one can similarly show that the density decays as $n(t) \sim (\lambda t)^{-1/2}$ for $d > 1$, with mere logarithmic corrections $n(t) \sim \left[(Dt)^{-1} \ln Dt\right]^{1/2}$ at $d_c = 1$ [16].

Generalizations of the pair annihilation reaction to multiple particle types introduce interesting new physics. For the *two-species* case $A + B \to \emptyset$ (with no concurrent reactions of identical particles), the rate equations read $\partial_t n_{A/B} = -\lambda\, n_A\, n_B$. With equal initial densities $n_{A0} = n_{B0}$ they are again solved by $n_{A/B}(t) \sim (\lambda t)^{-1}$; however, with $n_{A0} > n_{B0}$, say, one obtains $n_B(t) \sim \exp[-(n_{A0} - n_{B0})\,\lambda\, t]$ for the minority species, while the majority density saturates at $n_{As} > 0$. In order to establish the effects of spatial fluctuations, it is crucial to notice that the density difference $n_A - n_B$ remains strictly *conserved* under the reactions; for $D_A = D_B$ it simply obeys the diffusion equation [18]. Consequently, regions with A or B particle excess become amplified in time. As a result, when $n_{A0} = n_{B0}$, one finds that for dimensions $d \leq 4$ species segregation into A/B rich domains occurs [19]. The annihilation processes are then confined to sharp reaction fronts, leading to a decelerated density decay $n_{A/B}(t) \sim (Dt)^{-d/4}$. For unbalanced initial conditions, stretched exponential relaxation ensues for $d < 2$: $\ln n_B(t) \sim -t^{d/2}$, whereas $\ln n_B(t) \sim -t/\ln t$ at $d_c = 2$ [20]. In one dimension, special initial configurations may change this picture: Consider, e.g., the alternating arrangement $\ldots ABABAB \ldots$ of particles that upon encounter react with probability one. Now there is no reason anymore to distinguish between A and B, and the system is in the $2A \to \emptyset$ universality class.

An obvious question is then what happens for diffusion-limited pair annihilation of $q > 2$ particle species, with equal initial densities as well as reaction and diffusion rates [21]. In contrast with the two-species case, there exists no local conservation law. Furthermore the renormalization of the reaction vertex proceeds exactly as for $2A \to \emptyset$. Consequently, at least for $d \geq 2$, where the initial state is not that crucial, the long-time limit should in fact *generically* be governed by the single-species pair annihilation universality class [22]. This is obvious for $q \to \infty$: In this limit, the probability of like particles ever meeting vanishes, which renders the distinction of different species meaningless. However, in one dimension, at least in the limit of large reaction rates (which should describe the asymptotic regime), particles of different types cannot pass each other. This topological constraint allows for species segregation to occur. Indeed, a simplified deterministic version of the q-species pair annihilation process yields [22]

$$n(t) \sim t^{-\alpha(q)} \; , \text{ with } \alpha(q) = (q-1)/2q \; , \tag{18}$$

which correctly reproduces $\alpha(2) = 1/4$ and $\alpha(\infty) = 1/2$ in $d = 1$. The asymptotic decay (18) along with the subleading correction $\sim t^{-1/2}$ of the pair annihilation process without segregation were recently confirmed in extensive simulations [23]. Yet again, special initial conditions such as $\ldots ABCDABCD \ldots$ may prevent segregation and instead lead to the $2A \to \emptyset$ decay law.

6 Branching and annihilating random walks (BARW)

In order to allow again for a genuine phase transition, we combine the annihilation $kA \to \emptyset$ ($k \geq 2$) with branching processes $A \to (m+1)\, A$. The associated rate equation for these *branching and annihilating random walks* (BARW) reads $\partial_t\, n(t) = -\lambda\, n(t)^k + \sigma\, n(t)$, with the solution

$$n(t) = n_s \Big/ \left(1 + \left[(n_s/n_0)^{k-1} - 1\right] e^{-(k-1)\,\sigma\, t}\right)^{1/(k-1)} \; . \tag{19}$$

Mean-field theory thus predicts the density to approach the saturation value $n_s = (\sigma/\lambda)^{1/(k-1)}$ as $t \to \infty$ for any positive branching rate σ. Above the critical dimension $d_c(k) = 2/(k-1)$ therefore, the system only has an *active* phase; $\sigma_c = 0$ represents a degenerate 'critical' point, with scaling exponents essentially determined by the pure annihilation model: $\alpha = 1/(k-1) = \beta$, $\nu = 1/2$, and $z = 2$. However, Monte Carlo simulations revealed a much richer picture, in low dimensions clearly distinguishing between the cases of *odd* and *even* number of offspring m [3,24]: For $k = 2$, $d \leq 2$, and m *odd*, a transition to an inactive, absorbing phase is found, characterized by the DP critical exponents. On the other hand, for *even* offspring number there emerges a phase transition in one dimension, described by a novel universality class with $\alpha \approx 0.27$, $\beta \approx 0.92$, $\nu \approx 1.6$, and $z \approx 1.75$.

The above mapping to a stochastic field theory, combined with RG methods, elucidates the physics behind those remarkable findings [25]. The action for the most interesting pair annihilation case becomes

$$S = \int d^d x \, dt \left[\hat{\psi} \, (\partial_t - D \, \nabla^2) \, \psi - \lambda \, (1 - \hat{\psi}^2) \, \psi^2 + \sigma \, (1 - \hat{\psi}^m) \, \hat{\psi} \, \psi \right] , \quad (20)$$

which in general allows no direct Langevin representation. Upon combining the reactions $A \to (m+1)A$ and $2A \to \emptyset$, one notices immediately that the loop diagrams generate the lower-order branching processes $A \to (m-1)A$, $A \to (m-3)A \ldots$ Moreover, the one-loop RG eigenvalue $y_\sigma = 2 - m(m+1)/2$ (computed at the annihilation fixed point) shows that the reactions with smallest m are the most relevant. For *odd* m, we see that the generic situation is represented by $m = 1$, i.e., $A \to 2A$, supplemented with the spontaneous decay $A \to 0$. After a first coarse-graining step, this latter process (with rate μ) must be included in the effective model, which hence becomes identical with the action (7). Thence we are led to Reggeon field theory (9) describing the DP universality class, *provided* the induced decay processes are sufficiently strong to render $\sigma_c > 0$. Yet for $d > 2$ the renormalized mass term $\sigma_R - \mu_R$ remains positive, which leaves us with merely the active phase. For $d \leq 2$, however, the involved fluctuation integrals are infrared-divergent, thus indeed allowing the induced decays to overcome the branching processes to produce a non-trivial phase transition. As function of dimension, the critical exponents display an unusual discontinuity at $d_c = 2$, as they jump from their DP to the mean-field values as a result of the vanishing critical branching rate [25].

It is now obvious why the case of *even* offspring number m is fundamentally different: Here, the most relevant branching process is $A \to 3A$, and spontaneous particle death with associated exponential decay is *not* generated, which in turn precludes the previous mechanism for producing an inactive phase with exponential decay. This important distinction from the odd-m case can be traced to a microscopic *local conservation law*, for the reactions $2A \to \emptyset$ and $A \to 3A$, $A \to 5A \ldots$ always destroy or produce an even number of reactants, preserving the particle number *parity*. Formally, this is reflected in the invariance of the action (20) under the combined inversions $\psi \to -\psi$, $\hat{\psi} \to -\hat{\psi}$. As we saw earlier, the branching rate σ certainly constitutes a relevant variable near $d_c = 2$. Therefore the phase transition can only occur at $\sigma_c = 0$, and for any $\sigma > 0$ there exists only an active phase, described by mean-field theory. In two dimensions one readily computes the following logarithmic corrections: $\xi(\sigma) \sim \sigma^{-1/2} \ln(1/\sigma)$, and $n(\sigma) \sim \sigma \, [\ln(1/\sigma)]^{-2}$ [25].

However, setting $m = 2$ in the one-loop value for the RG eigenvalue y_σ, we notice that fluctuations drive the branching vertex *irrelevant* in low

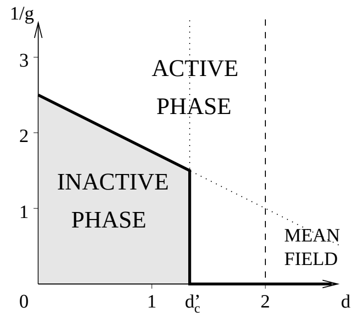

Fig. 3. Stationary states and unstable RG fixed point $1/g^*$ for BARW with even offspring number (PC universality class) as function of dimension d [25]

dimensions $d < d_c' \approx 4/3$. More information can be gained through a one-loop analysis at *fixed* dimension, albeit uncontrolled [25]. The ensuing RG flow equations for the renormalized branching rate $\sigma_R = \sigma / D\kappa^2$, and annihilation rate $\lambda_R = C_d \lambda / D\kappa^{2-d}$, with $C_d = \Gamma(2-d/2)/2^{d-1}\pi^{d/2}$ read (for $m = 2$):

$$\frac{d\sigma_R}{d\ell} = \sigma_R \left[2 - \frac{3\,\lambda_R}{(1+\sigma_R)^{2-d/2}} \right] , \quad \frac{d\lambda_R}{d\ell} = \lambda_R \left[2 - d - \frac{\lambda_R}{(1+\sigma_R)^{2-d/2}} \right] . \quad (21)$$

The effective coupling is then identified as $g = \lambda_R/(1+\sigma_R)^{2-d/2}$, which approaches the annihilation fixed point $g_a^* = 2 - d$ as $\sigma_R \to 0$, whereas for $\sigma_R \to \infty$ the flow tends towards the active state Gaussian fixed point $g_0^* = 0$. The separatrix between the two phases is given by the unstable RG fixed point $g_c^* = 4/(10-3d)$, which enters the physical regime below the borderline dimension $d_c' \approx 4/3$, as shown in Fig. 3. For $d < d_c'$, this describes a dynamic phase transition with $\sigma_c > 0$. The aforementioned fixed-dimension RG analysis yields the rather crude values $\nu \approx 3/(10-3d)$, $z \approx 2$, and $\beta \approx 4/(10-3d)$ for this *parity-conserving* (PC) universality class. The absence of any mean-field counterpart for this transition precludes a direct derivation of the 'hyperscaling' relations (10). Amazingly, in this non-equilibrium system fluctuations *generate* rather than destroy an ordered phase (translating back from the domain wall to the spin picture) in low dimensions. The inactive state is characterized by a vanishing branching rate, and consequently by the *algebraic* pair annihilation density decay. For particles undergoing Lévy flights, the existence of the power-law inactive phase is controlled by the anomalous diffusion exponent ρ, emerging for $\rho > \rho_c \approx 3/2$ in $d = 1$ [26].

Invoking similar arguments for the case of triplet annihilation $3A \to \emptyset$ combined with branching $A \to (m+1)A$ one would expect DP behavior with $\sigma_c > 0$ for $m \bmod 3 = 1, 2$, as then the processes $A \to \emptyset$, $A \to 2A$, and $2A \to A$ are dynamically generated. For $m = 3, 6, \ldots$, on the other hand, there can be different, novel scaling behavior, but because of $d_c = 1$ it will be limited to merely logarithmic corrections in one dimension [25]. It is also interesting to generalize the even-offspring BARW to q species, such that only equal particles can annihilate, $A_i + A_i \to \emptyset$, but both reactions $A_i \to 3A_i$ (with rate σ) and $A_i \to A_i + 2A_j$ (for $j \neq i$, and with rate σ') are possible. It turns out that the latter process always dominates, and in fact the ratio $\sigma/\sigma' \to 0$ under renormalization. Thus asymptotically one reaches the exactly analyzable $q \to \infty$ limit, with a mere degenerate phase transition at branching rate $\sigma'_c = 0$. Below $d_c = 2$, one finds the critical exponents $\alpha = d/2$, $\beta = 1$, $\nu = 1/d$, and $z = 2$ [25]. The situation for $q = 1$ is thus qualitatively different from all multi-component cases.

7 Annihilation–fission reactions

For single-species reactions without memory and disorder, the only remaining processes with potentially non-mean-field scaling behavior appear to be the combination of purely *binary* annihilation and fission reactions $2A \to A$ (with rate λ) and $2A \to (m+2)A$ (rate σ_m) with $d_c = 2$, and its *triplet* counterpart ($d_c = 1$). The former reactions subsequently generate $2A \to (m+1)A, mA, \ldots, 2(m+1)A, \ldots$, thus producing *infinitely* many couplings with identical scaling dimensions [27]. Upon including all these binary particle production reactions, the phase transition is readily seen to occur at $\lambda_c = \sum_m m\, \sigma_m$. The inactive, absorbing phase ($\lambda > \lambda_c$) is obviously characterized by the power laws of the pure coagulation model. Yet for $\lambda < \lambda_c$, the particle density diverges after a finite time, when no constraints on the site occupation numbers n_i are imposed. Thus the asymptotic density is finite only at the phase transition itself. These singular features of the 'bosonic' model with its highly discontinuous phase transition are overcome by restricting the site occupation numbers to $n_i = 0, 1$. Extensive Monte Carlo simulations have in fact revealed that this leads to a continuous transition, with critical exponents that seem to belong to a novel universality class (*pair contact process with diffusion*, PCPD) with critical dimension $d_c = 2$ [28]. However, owing to the difficulty of obtaining truly asymptotic properties in this system, where reactions become extremely rare at low densities, the precise nature of this critical point in purely binary reactions has remained elusive and rather controversial. This applies even to density matrix RG studies [29].

It is therefore fortunate that recent work has demonstrated how to consistently implement site occupation restrictions into the bosonic field theory [30]. For the above binary processes, the reaction part of the action becomes

$$S = \int d^d x\, dt \Big[\sigma_m \,(1-\hat{\psi}^m)\, \hat{\psi}^2\, \psi^2\, e^{-(m+2)v\,\hat{\psi}\,\psi} - \lambda\,(1-\hat{\psi})\,\hat{\psi}\,\psi^2\, e^{-2v\,\hat{\psi}\psi} \Big] . \quad (22)$$

Here the exponential terms capture the occupation number limitations, with $[v] = \kappa^{-d}$, which suggests that v represents a *dangerously irrelevant* coupling. Indeed, consider more generally the coupled reactions $kA \to (k-l)A$ with $0 < l \le k$ and $nA \to (n+m)A$ with $n, m > 0$, which display a continuous transition for $k \le n$. The mean-field equations obtained from the associated actions show that site occupation restrictions can be neglected at low densities, yielding the critical exponents $\beta = 1/(k-n)$, $\nu = (k-1)/2(k-n)$, $z = 2$, and $\alpha = 1/(k-1)$, except for the degenerate case $k = n$, where one finds $m\, n_s = \ln(m\sigma_m/l\lambda)$, whence $\beta = 1$, $\nu = k/2$, $z = 2$, and $\alpha = 1/k$ [31,32]. For $k = 1$, expanding the exponentials leads to the action (8), which establishes that the competing processes $A \to \emptyset$, $A \to 2A, \ldots$ with site exclusion yield a DP phase transition. The above field theory should also permit a systematic analysis of the fluctuation corrections for the purely binary and triplet reactions. For the latter, one expects mere logarithmic corrections at $d_c = 1$ to the mean-field scaling laws; yet current simulations are inconclusive [32].

8 Concluding remarks

In this overview, I have outlined how non-linear stochastic processes via their defining master equation can be represented by field theory actions, allowing for a thorough analysis and classification by means of the renormalization group. Systems with a single 'particle' species that display a non-equilibrium phase transition from an active to an inactive, absorbing state are generically captured by the directed percolation universality class. The second prominent example, sometimes applicable when additional symmetries (degenerate absorbing states) are present, is the parity-conserving universality class of even-offspring branching and annihilating random walks. The only other scenarios for non-trivial critical scaling behavior appear to be provided by the solely pair or triplet annihilation–fission reactions, where site occupation restrictions become relevant. The full classification of reaction-diffusion models with multiple particle species remains a formidable task. Even the difference in diffusivities may become a relevant control parameter [33]. Specifically in one dimension, exclusion constraints can play a crucial role [34].

Another obviously important open problem concerns the influence of quenched disorder in the reaction rates. For example, a field theory RG investigation for DP with random percolation threshold yields run-away flows [35], reflected in intriguing simulation results with not entirely clear interpretation [36]. A very recent strong disorder RG study, supplemented with numerical density matrix RG calculations, has revealed a novel disorder fixed point [37]. A better understanding of spatially varying reaction rates might also explain the conspicuous rarity of clear-cut experimental realizations even for the supposedly ubiquitous DP universality class [38]. In fact the single verification of DP scaling behavior appears to be its observation in spatio-temporal intermittency in ferrofluidic spikes [39]. Thus many intriguing issues are still open; but I expect that in addition to increasingly more extensive Monte Carlo simulations and sophisticated numerical techniques, field theory representations and subsequent analysis by means of the renormalization group will remain an invaluable tool for the further understanding of cooperative phenomena and scale-invariance in interacting non-equilibrium systems.

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