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Reaction-diffusion with a time-dependent reaction rate: the single-species diffusion-annihilation process

L Turban

Laboratoire de Physique des Matériaux, Université Henri Poincaré (Nancy 1), BP 239, F-54506 Vandœuvre lès Nancy Cedex, France

E-mail: turban@lpm.u-nancy.fr

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Abstract

We study the single-species diffusion–annihilation process with a time-dependent reaction rate, $\lambda(t) = \lambda_0 \, t^{-\omega}$. Scaling arguments show that there is a critical value of the decay exponent $\omega_{\rm c}(d)$ separating a reaction-limited regime for $\omega > \omega_{\rm c}$ from a diffusion-limited regime for $\omega < \omega_{\rm c}$. The particle density displays a mean-field, ω -dependent, decay when the process is reaction limited whereas it behaves as for a constant reaction rate when the process is diffusion limited. These results are confirmed by Monte Carlo simulations. They allow us to discuss the scaling behaviour of coupled diffusion–annihilation processes in terms of effective time-dependent reaction rates.

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1. Introduction

Time-dependent effective reaction rates can be introduced into mean-field kinetic equations in order to simulate the effect of concentration fluctuations below the upper critical dimension d_c where deviations from the standard mean-field behaviour are observed [1]. In this work, we take the opposite point of view and look how far the fluctuations in a reaction–diffusion process can be affected by a time-dependent reaction rate.

We study the single-species diffusion-annihilation process

$$A\varnothing \stackrel{1}{\longleftrightarrow} \varnothing A$$
 (diffusion)
$$AA \stackrel{\lambda(t)}{\longrightarrow} \varnothing \varnothing$$
 (annihilation), (1)

where nearest-neighbour particles annihilate with a reaction rate

$$\lambda(t) = \frac{\lambda_0}{t^{\omega}},\tag{2}$$

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decaying as a power of the time. The diffusion rate associated with the exchange of a particle (A) with a vacancy (\emptyset) is equal to 1, which fixes the time scale.

When the reaction rate is constant ($\omega=0$), the kinetic equation $\dot{\rho}=-\lambda_0\rho^2$ leads to the mean-field asymptotic behaviour $\rho(t)\simeq(\lambda_0t)^{-1}$. Simple scaling arguments [2, 3] indicate that, due to the concentration fluctuations, the decay is slower in low dimensions. At time t, for $d\leqslant 2$ where the exploration is dense, a surviving particle has swept out a region with a linear size given by the diffusion length \sqrt{Dt} . Therefore, the volume per particle grows as $(Dt)^{d/2}$ and the particle density decays algebraically as

$$\rho(t) = Ct^{-\alpha}, \qquad \alpha = \frac{d}{2}, \qquad d < d_{c} = 2.$$
(3)

Thus mean-field theory gives the correct scaling behaviour above the upper critical dimension $d_c = 2$.

This picture is confirmed by exact results in one dimension (1d) [4–6], rigorous bounds [7] and renormalization group results [8, 9] showing that the amplitude C is universal. As usual, logarithmic corrections occur at d_c where the particle density decays as [9]

$$\rho(t) = \frac{1}{8\pi} \frac{\ln t}{Dt}, \qquad d = d_{\rm c} = 2,$$
(4)

where D is the diffusion constant.

We consider the influence of the decay exponent ω , the amplitude λ_0 and the dimension d of the system on the scaling behaviour of the particle density $\rho(t)$. In section 2, we give scaling arguments showing that the mean-field behaviour should be recovered when the decay exponent is greater than a critical value $\omega_c(d)$ when $d \leq d_c$ and calculate the time evolution of the particle density in mean-field theory. In section 3 these results are confronted with Monte Carlo simulation data in dimension d=1 to 3. In section 4 we show how time-dependent reaction rates are effectively realized in the case of coupled reactions. Our results are summarized in section 5.

2. Scaling considerations and mean-field theory

In a continuum description in d dimensions, the spacetime evolution of the particle density field, $\rho(\mathbf{r}, t)$, is governed by a Langevin equation containing diffusion, reaction and noise terms [10–12]

$$\partial_t \rho(\mathbf{r}, t) = -\frac{\lambda_0}{t^{\omega}} \rho^2(\mathbf{r}, t) + D\nabla^2 \rho(\mathbf{r}, t) + \zeta(\mathbf{r}, t). \tag{5}$$

The noise term $\zeta(\mathbf{r}, t)$ accounts for the fluctuations of the particle density at position \mathbf{r} at time t. Let us consider the behaviour under rescaling of the reaction term

$$\partial_t \rho(\mathbf{r}, t)|_{\text{reaction}} = -\frac{\lambda_0}{t^{\omega}} \rho^2(\mathbf{r}, t)$$
 (6)

at the stable fixed point of the system with a constant reaction rate, when the fluctuations are relevant, i.e., below the upper critical dimension d_c .

Under a change of the length scale L' = L/b, the particle density field and its space average $\rho(t)$ scale with the same dimension x_{ρ} and the scaling dimension of t is the dynamical exponent z = 2, so that

$$[\rho(\mathbf{r},t)]' = b^{x_{\rho}}\rho(\mathbf{r},t), \qquad t' = \frac{t}{b^{z}}.$$
 (7)

For the average particle density, one obtains

$$[\rho(t)]' = \rho\left(\frac{t}{b^z}\right) = b^{x_\rho}\rho(t). \tag{8}$$

Taking $b = t^{1/z}$ leads to the power-law decay

$$\rho(t) = \rho(1)t^{-\alpha}, \qquad \alpha = \frac{x_{\rho}}{z}.$$
 (9)

Using the transformations (7) in (6), we obtain

$$\partial_{t'}[\rho(\mathbf{r},t)]'|_{\text{reaction}} = b^{x_{\rho}+z} \partial_{t} \rho(\mathbf{r},t)|_{\text{reaction}}$$

$$= -\frac{\lambda'_{0}}{t'^{\omega}} [\rho^{2}(\mathbf{r},t)]'$$

$$= -b^{z\omega+2x_{\rho}} \frac{\lambda'_{0}}{t^{\omega}} \rho^{2}(\mathbf{r},t)$$
(10)

so that the reaction-rate amplitude transforms as

$$\lambda_0' = b^{-z(\omega - 1 + x_\rho/z)} \lambda_0 = b^{-z(\omega - 1 + \alpha)} \lambda_0, \tag{11}$$

where the last expression follows from (9).

When ω is smaller than the critical value ω_c , given by

$$\omega_{\rm c} = 1 - \alpha = 1 - \frac{d}{2}, \qquad d \leqslant d_{\rm c}, \tag{12}$$

according to (3), λ_0 increases under rescaling, i.e., the process is diffusion limited. The concentration fluctuations are relevant and the critical behaviour is governed by the same fixed point as for a constant reaction rate, for which $\alpha = d/2$.

When $\omega > \omega_c$, the reaction-rate amplitude decreases and the process is reaction limited. The concentration fluctuations are suppressed by diffusion and the system should display a mean-field behaviour governed by a fixed line, parametrized by ω .

When $\omega = \omega_c$, λ_0 is a marginal variable and logarithmic corrections to the mean-field behaviour are expected.

The behaviour of $\rho(t)$ follows from the mean-field rate equation

$$\dot{\rho}(t) = -\lambda(t)\rho^2(t). \tag{13}$$

With the initial condition $\rho(t_0) = \rho_0$, the average density reads

$$\frac{1}{\rho(t)} = \frac{1}{\rho_0} + \lambda_0 \frac{\left(t^{1-\omega} - t_0^{1-\omega}\right)}{1-\omega} \qquad (\omega \neq 1)$$

$$\frac{1}{\rho(t)} = \frac{1}{\rho_0} + \lambda_0 \ln\left(\frac{t}{t_0}\right) \qquad (\omega = 1).$$
(14)

In the asymptotic regime, $t \gg t_0$, one obtains

$$\rho(t) \simeq \frac{1 - \omega}{\lambda_0} t^{-(1 - \omega)} \qquad (\omega < 1)$$

$$\rho(t) \simeq \frac{1}{\lambda_0 \ln(t)} \qquad (\omega = 1)$$
(15a)

$$\rho(t) \simeq \frac{1}{\lambda_0 \ln(t)} \qquad (\omega = 1) \tag{15b}$$

$$\rho(t) \simeq \rho_{\infty} + \rho_{\infty}^2 \lambda_0 \frac{t^{-(\omega - 1)}}{\omega - 1} \qquad (\omega > 1).$$

$$(15c)$$

When $\omega > 1$, the particle density decays algebraically to a non-vanishing asymptotic value, ρ_{∞} , behaving as

$$\frac{1}{\rho_{\infty}} = \frac{1}{\rho_0} + \lambda_0 \frac{t_0^{-(\omega - 1)}}{\omega - 1} \qquad (\omega > 1)$$

$$\rho_{\infty} \simeq \frac{\omega - 1}{\lambda_0} \qquad (\omega \to 1 +).$$
(16)

When $d \geqslant d_c$, $\omega_c = 0$ according to (12). For positive values of ω , the annihilation process is reaction limited and the mean-field behaviour in (15*a*)–(15*c*) and (16) applies. For negative values of ω the reaction rate, which increases under rescaling, no longer controls the annihilation process. Thus one expect a scaling behaviour independent of ω , the same as for $\omega = 0$ with $\alpha = 1$ and logarithmic corrections given by (4) at d_c when $\omega < 0$.

3. Monte Carlo simulations

Numerical simulations of the diffusion–annihilation process have been performed in order to check the results of the last section for the asymptotic behaviour of the particle density $\rho(t)$ in dimensions d=1 to 3.

3.1. Algorithm

We work on hypercubic lattices with $L^d = 10^6$ sites and periodic boundary conditions in all directions. In order to avoid finite-size effects the simulations are stopped when the diffusion length reaches some fraction (1/4 to 1/10) of the size L of the system. The particle density $\rho(t)$ is averaged over 5 to 20 samples.

At $t_0 = 1$ the L^d sites are independently occupied by a particle with probability ρ_0 . At time t, one of the N(t) surviving particles is randomly selected and a jump towards one of the 2d neighbouring sites is attempted, with the same probability for all the sites. When the target site is empty, the jump is accepted. When it is occupied, either the two particles annihilate with probability $\lambda(t)$ or they keep their original location with probability $1 - \lambda(t)$, i.e., multiple occupancy of a site is not allowed. Finally, the time is incremented by 1/N(t) in all the cases and the process is repeated.

When $\omega < 0$, the algorithm has to be modified since the reaction rate may increase beyond 1. In this case, we let two particles annihilate with probability $\lambda(t)\tau(t)=1$. We take $\lambda_0=1$ such that $\lambda(t)\geqslant 1$ and $\tau(t)\leqslant 1$. The diffusion jumps are attempted with probability $\tau(t)\leqslant 1$ and the time is incremented by $\tau(t)/N(t)$ at each Monte Carlo step.

3.2. Numerical results in 1d

The influence of ω on the scaling behaviour of $\rho(t)$ is shown in figure 1. The asymptotic slope, $-\alpha$, is the same as for a constant reaction rate (-1/2, indicated by a dashed line) as long as $\omega \leq \omega_c = 1/2$. The amplitude of $\rho(t)$ is independent of ω below ω_c and it is universal as for $\omega = 0$ [9]: it depends neither on the initial density ρ_0 nor on the reaction-rate amplitude λ_0 , the process being diffusion limited.

The form of the logarithmic correction to the algebraic decay, expected at ω_c in relation to the marginality of λ_0 , could not be extracted from our Monte Carlo data.

Above ω_c , α decreases continuously to 0 when ω increases to 1. As shown in figure 2 the amplitude of $\rho(t)$ is no longer universal: it remains independent of ρ_0 but, the process being now reaction limited, it depends on λ_0 . The inset shows that the amplitude actually varies as $1/\lambda_0$, in agreement with the mean-field result in (15*a*).

The scaling behaviour of $\rho(t)$ at $\omega = 1$ is illustrated in figure 3. The product $\rho \ln(t)$ tends to a constant value at long times, in agreement with mean-field theory in equation (15b).

The variation of α with ω is illustrated in figure 4. The exponents were deduced from the extrapolation of two-point approximants for the slope of $\ln(\rho)$ versus $\ln(t)$ using the Burlisch–Stoer (BS) algorithm [13, 14]. These data were obtained with $\rho_0 = 0.3$ in order to accelerate the approach of the asymptotic regime (see figure 2 for a comparison with $\rho_0 = 1$ when

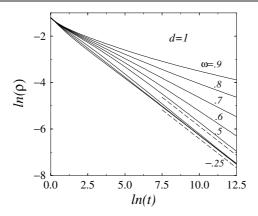


Figure 1. Time dependence of the particle density in 1d for different values of the decay exponent ω . The initial density is $\rho=0.3$ and the reaction-rate amplitude $\lambda_0=1$. The asymptotic slope, $-\alpha$, varies with ω in the reaction-limited mean-field regime, above the critical value $\omega_c=1/2$. Below ω_c , in the diffusion-limited regime, it remains constant and is equal to -1/2 (dashed lines). The amplitude is universal below ω_c , the data for $\omega=0.25$, 0 and -0.25 collapsing asymptotically on a single line.

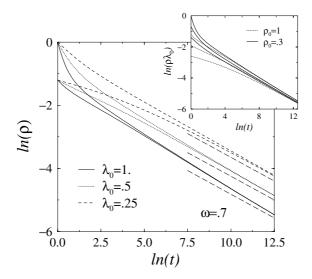


Figure 2. Influence of ρ_0 and λ_0 on the amplitude of the particle density $\rho(t)$ in 1d. At $\omega = 0.7$, i.e., in the reaction-limited regime, the amplitude does not depend on the initial density ρ_0 and it varies as $1/\lambda_0$ as shown in the inset where an asymptotical collapse is obtained for $\rho\lambda_0$.

 $\lambda_0=1$). The value $\lambda_0=1$ was selected to spare computer time although smaller values can lead to better estimates of the exponent as illustrated for $\omega=0.7$ where the circles correspond to values of $\lambda_0=1,0.4,0.2$ and 0.1 from top to bottom. The results are in good agreement with the expected behaviour.

3.3. Numerical results in 2d and 3d

The log-log plots for the time evolution of the particle density for different values of ω are shown in figure 5 for d=2 and figure 6 for d=3. As above the data were obtained with an

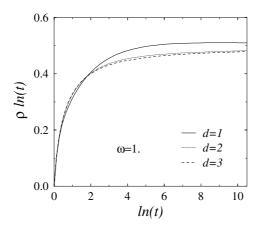


Figure 3. Logarithmic scaling behaviour at $\omega=1$ for d=1 to 3. In the mean-field reaction-limited regime the particle density decays asymptotically as $1/\ln(\rho)$ when $\omega=1$. The data were obtained with $\rho_0=1$ and $\lambda_0=1$.

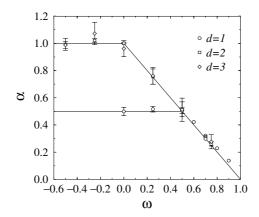


Figure 4. Variation of the particle density exponent α with the reaction-rate exponent ω for d=1,2 and 3. The data, obtained with $\rho=0.3$ and $\lambda_0=1$, are in overall agreement with the expected behaviour (solid lines). Below $\omega_{\rm c}$ (1/2 when d=1,0 when d=2,3) the process is diffusion limited and α is a constant. Above $\omega_{\rm c}$ the process is reaction limited and α decays as $1-\omega$ in agreement with mean-field theory.

initial density $\rho_0=0.3$ and a reaction-rate amplitude $\lambda_0=1$. In both cases the critical value of the reaction-rate exponent is $\omega_c=0$. α varies with ω in the reaction-limited regime, $\omega>\omega_c$, and remains constant in the diffusion-limited regime, $\omega\leqslant\omega_c$.

In 2d there is a systematic deviation from the slope -1 (indicated by a dashed line in figure 5) when $\omega \leqslant 0$. This may be traced to the logarithmic correction in equation (4), occurring at the upper critical dimension $d_c=2$ in the diffusion-limited regime. When ρ is divided by $\ln(t)$ (dotted lines) the slopes are asymptotically close to the expected value -1 corresponding to the mean-field exponent $\alpha=1$. The amplitude is probably universal, the same for all values of $\omega \leqslant 0$, although the evolution to the asymptotic behaviour becomes quite slow when approaching $\omega_c=0$.

In $d=3>d_{\rm c}$ the mean-field result of equation (15a) with $\omega=0$, $\rho(t)\sim t^{-1}$, applies in the whole diffusion-limited regime $\omega\leqslant 0$ as shown in figure 6 where the slopes are close to the expected value, indicated by the dashed lines, without any correction. Here too we expect

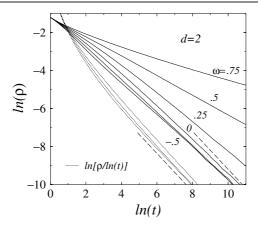


Figure 5. Time dependence of the particle density in 2d with $\rho_0=0.3$ and $\lambda_0=1$. The asymptotic slope depends on ω in the reaction-limited regime $\omega>\omega_c=0$ and remains constant in the diffusion-limited regime ($\omega=0,-0.25,-0.5$). There is a deviation from the expected slope, -1 (dashed lines), due to the logarithmic correction occurring in the diffusion-limited regime at $d_c=2$. When ρ is divided by $\ln(t)$ (dotted lines) the correct slope is recovered.

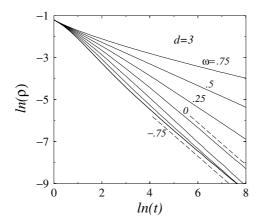


Figure 6. Time dependence of the particle density in 3d with $\rho_0=0.3$ and $\lambda_0=1$. The critical value of the reaction-rate exponent is $\omega_c=0$, the same as in 2d. Since $d>d_c$, there are no logarithmic corrections and the asymptotic slopes are close to -1 (dashed lines) in the diffusion-limited regime ($\omega=0,-0.25,-0.5,-0.75$).

the amplitude to be universal when $\omega \leq 0$ but the evolution to the true asymptotic behaviour is even slower than in 2d when ω_c is approached.

The asymptotic logarithmic decay of the particle density at $\omega=1$ (see equation (15b)) is illustrated in figure 3. The variation of α with ω , shown in figure 4, is still in agreement with the expected behaviour. The exponents were obtained through a non-linear least-square fit, taking into account an effective correction-to-scaling exponent, since the statistical fluctuations were too high to use the BS algorithm.

4. Time-dependent effective reaction rates

Time-dependent effective reaction rates with a power-law decay can be realized in the case of coupled reactions. Let us first consider a diffusion–annihilation process in 2d where the

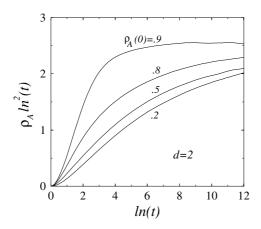


Figure 7. Scaling behaviour of an asymmetric diffusion–annihilation process in 2d where the annihilation of *A* particles is catalysed by *B* particles whereas the annihilation of *B* particles is free. The product $\rho_A \ln^2(t)$ tends to a constant value at long time.

annihilation of a pair of A particles is catalysed by B particles whereas B particles annihilate without any further condition:

$$ABA \xrightarrow{\lambda_A} \varnothing B\varnothing$$
 (catalysed annihilation)
$$BB \xrightarrow{\lambda_B} \varnothing \varnothing$$
 (simple annihilation). (17)

The effective reaction rate for AA annihilation is governed by the mean density of B particles, $\rho_B(t)$, if the concentration fluctuations are negligible, which is true in 2d. Thus $\omega_A = \alpha_B = 1$ for the single-species annihilation in 2d. The AA process being reaction limited with $\omega_A = 1$, mean-field theory applies and a logarithmic decay is expected. Actually, one has to take into account the logarithmic correction in equation (4) so that the effective reaction rate is $\lambda_A(t) = \lambda_{A0} \ln(t)/t$. The mean-field rate equation (13) leads to the asymptotic behaviour

$$\rho_A(t) \simeq \frac{2}{\lambda_{A0}} \frac{1}{\ln^2(t)}.\tag{18}$$

This process has been simulated on the square lattice with $\lambda_A = \lambda_B = 1$ and $\rho_A(0) + \rho_B(0) = 1$. The size of the system, the boundary conditions and the number of samples are the same as before. When a particle jump is attempted towards an occupied site, the two particles annihilate in the case of a *B* pair. For a pair of *A* particles, the annihilation occurs only when a *B* particle is first neighbour of one of the *A* particles and second neighbour of the second. When the two particles are different they just keep their positions.

The Monte Carlo results, shown in figure 7, confirm this scaling behaviour although the convergence is quite slow for small or intermediate initial densities of *A* particles.

The second example concerns a system of two symmetrically coupled single-species diffusion–annihilation processes in 2d with

$$ABA \xrightarrow{\lambda_A} \varnothing B\varnothing$$
 (catalysed annihilation)
$$BAB \xrightarrow{\lambda_B} \varnothing A\varnothing$$
 (catalysed annihilation). (19)

The density of one species controls the reaction rate of the other so that $\omega_A = \alpha_B$. Now, assuming that the system is in the reaction-limited regime, equation (15a) leads to $\alpha_A = 1 - \omega_A = 1 - \alpha_B$ and

$$\alpha_A + \alpha_B = 1. \tag{20}$$

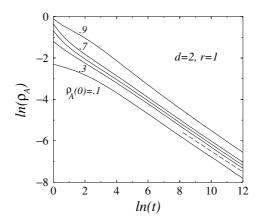


Figure 8. Scaling behaviour of the particle density for a 2d symmetric diffusion–annihilation process where the annihilation of two particles of one species is catalysed by a particle of the other species. The reaction rates are equal, $r = \lambda_A/\lambda_B = 1$, and both densities decay as $t^{-1/2}$. The density of B particles with initial density $\rho_B(0)$ is equal to the density of A particles with initial density $\rho_A(0) = 1 - \rho_B(0)$. The asymptotic slope, -1/2, is indicated by a dashed line.

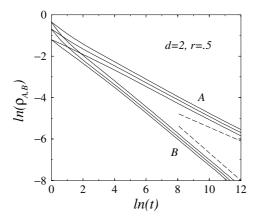


Figure 9. Scaling behaviour of the particle density for a 2d symmetric diffusion–annihilation process where the annihilation of two particles of one species is catalysed by a particle of the other species. Here the reaction rates are different with $r = \lambda_A/\lambda_B = 1/2$. The initial densities are such that $\rho_A(0) + \rho_B(0) = 1$ and equal to 0.7, 0.5, 0.3, from top to bottom for A and B particles. The expected asymptotic slopes, -1/3 for A particles and -2/3 for B particles, are indicated by dashed lines. The convergence is slow for the majority species.

When the process is fully symmetric, i.e., when $\lambda_A = \lambda_B$, both species decay with the same exponent $\alpha_A = \alpha_B = 1/2$, a value consistent with the assumed reaction-limited mean-field behaviour since $\omega_c = 0$ in 2d. Actually, the fully symmetric process reduces to the $3A \rightarrow A$ process which is known to decay as $t^{-1/2}$ above its upper critical dimension $d_c = 1$ [15].

The Monte Carlo simulations on the square lattice with $r = \lambda_A/\lambda_B = 1$, $\rho_A(0) + \rho_B(0) = 1$ and the same rules as above for the catalysed annihilation of the two species are shown in figure 8. The scaling behaviour is in complete agreement with a decay exponent equal to 1/2.

A deviation of the reaction-rate ratio r from 1 is sufficient to break the symmetry between the two species, leading to different decay exponents. This follows from the solution of the

system of coupled mean-field rate equations

$$\dot{\rho}_A(t) = -\lambda_A \rho_A^2(t) \rho_B(t), \qquad \dot{\rho}_B(t) = -\lambda_B \rho_B^2(t) \rho_A(t). \tag{21}$$

Multiplying the first equation by ρ_B , the second by ρ_A and adding, one obtains a differential equation for $\rho_A \rho_B$ leading to the asymptotic behaviour

$$\rho_A(t)\rho_B(t) \sim t^{-1} \tag{22}$$

in agreement with (20). Dividing the first equation by ρ_A and using (22), one finally obtains

$$\rho_A(t) \sim t^{-r/(1+r)}, \qquad \rho_B(t) \sim t^{-1/(1+r)}, \qquad r = \lambda_A/\lambda_B.$$
(23)

The decay exponents continuously vary with the ratio r of the reaction rates but their sum remains equal to 1 as expected from the effective reaction-rate argument leading to (20). Actually, it is easy to verify that λ_A and λ_B are marginal variables when $\alpha_A + \alpha_B = 1$.

The Monte Carlo results are shown in figure 9 for d=2 and r=1/2. They confirm this behaviour, although the convergence to the asymptotic slope is rather slow for the majority species.

5. Conclusion

We have shown, through scaling considerations and Monte Carlo simulations, that when the reaction rate of the single-species diffusion–annihilation process decays as $t^{-\omega}$, there is a critical value $\omega_{\rm c}=1-d/2$ for $d\leqslant d_{\rm c}$ separating reaction-limited behaviour for $\omega>\omega_{\rm c}$ from diffusion-limited behaviour for $\omega<\omega_{\rm c}$. In the reaction-limited regime, mean-field theory is always valid and leads to an ω -dependent decay of the particle density whereas in the diffusion-limited regime, due to concentration fluctuations, the particle density decays as $t^{-d/2}$ when $d\leqslant d_{\rm c}=2$ as for a constant reaction rate.

In the case of coupled reactions where the annihilation of one type of particles is catalysed by the other, one obtains a time-dependent effective reaction rate which is easy to identify when the process is reaction limited, i.e., when mean-field theory applies.

The main aim of decreasing the time-dependent reaction rates is to extend the validity of mean-field theory below the usual upper critical dimension and to lead to unusual mean-field behaviour.

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