Bimolecular annihilation reactions with immobile reactants: Unequal reactant concentrations

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We study the bimolecular annihilation reaction $A + B \to 0$ with immobile reactants interacting via exchange. We extend our previous analyses to the case of unequal concentrations of A and B particles. A comparison between the direct simulation of the reaction process and an analytical approach, which is based on Kirkwood's superposition approximation, shows reasonable agreement, although deviations are larger than in the case of equal A and B concentrations. Interestingly, the asymptotic decay forms for the particle concentrations (when formulated in terms of a suitable reaction length) parallel those encountered in diffusion-limited reactions. Here the diffusion length $l_D = (Dt)^{1/2}$ gets replaced by the reaction radius $\xi = \ln t$. We discuss implications of this finding for a qualitative (but quite accurate) picture of the temporal evolution behavior.

I. INTRODUCTION

Bimolecular reactions of the type $A + B \to 0$ between unlike particles (A and B) have led to a series of recent investigations.[1-8] This type of reaction plays an important role in different kinds of experiments,[4] such as certain chemical reactions, energy-transfer processes (exciton migration), recombination of defects in solids, electron–hole recombination, or electron scavenging. Thus, for example, the capture of an electron by a scavenger may be considered as a reaction in which one free electron and one active scavenger disappear. Experiments for this type of reaction were reported by Miller,[9,10] who studied the annihilation of trapped electrons (e_{tr}^-) in a matrix of 2-methyltetrahydrofurane from the reaction with naphthalene (Nh):

$$e_{tr}^- + \text{Nh} \to \text{Nh}^-.$$

In a similar experiment the annihilation of trapped electrons in an alkaline water matrix from the reaction with O^- anion radicals was observed at low temperatures.[11,12]

Another example for $A + B \to 0$ type reactions is the recombination of electrons and holes in amorphous hydrogenated silicon.[13] In samples of α-Si:H with a low density of broken bonds the basic reaction channel is tunneling radiative recombination. At low temperature $T < 20$–30 K it is assumed that diffusion of the localized photocarriers (thermally activated or tunneling) can be neglected during the short-time thermalization processes. Then the light-generated electrons and holes get trapped in localized states of the band tail and recombine via tunneling.[13] Further systems, in which electron–hole recombination leading to luminescence occurs, are chalcogenide glass semiconductors.[14] For injected charge carriers one has, in general, different initial numbers n_A^0 and n_B^0 of electrons and holes in the system, an important feature for the present work.

Most of the recent analytical work centered on diffusion-limited reactions, where particles move diffusively and react on contact. For a spatially homogeneous distribution of particles at all times one has for the concentrations $n_A(t)$ and $n_B(t)$ the following equation:[4]

$$\frac{\partial n_A(t)}{\partial t} = \frac{\partial n_B(t)}{\partial t} = -kn_A(t)n_B(t). \tag{1}$$

In Eq. (1) the constant k is the reaction rate; furthermore, we denote the initial concentrations by n_A^0 and n_B^0. Now, the solution of Eq. (1) for $n_A^0 = n_B^0$ is

$$n_A(t) = \frac{n_A^0}{1 + n_A^0 kt}, \tag{2}$$

which for large t follows a t^{-1} law. On the other hand, for $n_A^0 < n_B^0$ one obtains with $\Delta = n_B^0 - n_A^0$:

$$n_A(t) = \frac{\Delta}{(n_B^0/n_A^0)\exp(\Delta kt) - 1}, \tag{3}$$

i.e., for large t the concentration of the minority species decays exponentially in time, $n_A(t) \sim \exp(-\Delta kt)$.

This behavior, however, changes drastically, when, due to the discrete nature of the particles, spatial fluctuations in the initial distributions occur. Then in the course of the reaction clusters of like particles (i.e., regions where only one kind of particles is present) may develop, a fact which leads to a slowing down of the overall reaction, as only particles at the border of such clusters are likely to react. In this case the decay law depends on the specific conditions of the reaction and on the restrictions on the space in which the reaction takes place. Thus, in the diffusion-limited case with $n_A^0 = n_B^0$ one finds $n_A(t) \sim t^{-d/4}$ in dimensions $d < 4$.[1-3] The appearance of a marginal dimension, $d = 4$, is due to the tendency of diffusion to mix the clusters, a fact which gets enhanced in higher dimensions, so that for $d > 4$ the system recovers its well-stirred t^{-1} behavior. In order to avoid this mixing effect of diffusion, we recently[15-17] studied a model with immobile A and B particles which react via exchange,

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$w(\mathbf{r}) = w_0 \exp(-r/r_0)$, where w is the probability rate that one A and one B molecule situated at a distance $\mathbf{r}$ apart react with each other and r_0 is a measure of the range of the interaction. Note that this exponential form is typical for exchange-dominated reactions in solids, like scavenging or recombination of electrons and holes via tunneling, or static tunnel recombination of defects in solids created by irradiation.

For exchange-type reactions and for equal initial concentrations $n_A^0 = n_B^0$ we found from direct computer simulations of the $A + B \to 0$ reaction, in which the annihilation process for random distributions of A and B particles was simulated using a minimal process method,[18] that the particle concentrations decay as[15,16]

$$n_{A,B}(t) \sim \xi^{-d/2} \sim (\ln w_0 t)^{-d/2}, \tag{4}$$

where $\xi = r_0 \ln(w_0 t)$ is the (time-dependent) reaction radius. This result remains also valid for fractal lattices, where instead of d the fractal dimension $\bar{d}$ enters. We remark that Eq. (4) was already suggested for this type of reaction by Burlatski and Ovchinnikov[6] and was recently shown to be asymptotically exact.[19] We note that logarithmic-type time dependencies were employed by Zamaraev et al.[11] in explaining the effect of tunneling annihilation for trapped electrons. They studied the kinetics of tunneling annihilation at low temperatures for trapped electrons in an alkaline water matrix from the reaction with O^- anion radicals which are formed during radiolysis.

Another analytical approach was developed by Kuzovkov and Kotomin[8] and is based on Kirkwood's superposition approximation. In the case of equal initial concentrations $n_A^0 = n_B^0$ this method performs very well and (as shown by us) provides both the correct long-time behavior of the concentrations as well as the correct AB-particle segregation patterns on one- and two-dimensional lattices[15,16] and on Sierpinski gaskets.[17]

In this article we extend our analyses and simulations to the case of *different* initial concentrations, $n_A^0 < n_B^0$. In the following section we will summarize the basic ideas of the approach of Kuzovkov and Kotomin[8] and will concentrate on the set of differential equations which have to be evaluated numerically. In Sec. III we compare the solutions of this set of equations to the results of direct simulations of the annihilation process. In Sec. IV we point out some parallels to diffusion-limited reactions and suggest an asymptotic, analytical expression for the concentration decay. We summarize our findings in Sec. V.

II. THE ANALYTICAL APPROACH

A complete derivation of the analytical equations is given elsewhere.[8,16] Here we only summarize the ideas of the approach and present the final set of equations.

The starting point of the analysis is an infinite set of differential equations for the many-point densities $\rho_{m,m'}$,

$$\rho_{m,m'} = \left\langle \prod_{i=1}^{m} n_A(\mathbf{r}_i, t) \prod_{j=1}^{m'} n_B(\mathbf{r}_j', t) \right\rangle, \tag{5}$$

representing products of reactant concentrations, averaged over all initial distributions of particles, which fulfill the same initial condition, and over all realizations of the annihilation process. m and m' denote the number of A and B particles involved in the product. In order to decouple the infinite set of equations, one expresses three-point densities in terms of two-point densities according to

$$\langle n_1 n_2 n_3 \rangle = \frac{\langle n_1 n_2 \rangle \langle n_2 n_3 \rangle \langle n_3 n_1 \rangle}{\langle n_1 \rangle \langle n_2 \rangle \langle n_3 \rangle}. \tag{6}$$

This approximation is due to Kirkwood and is often called the superposition approximation. Its range of validity, however, cannot be well assessed by analytical methods, but has to be checked by comparison to experiments or simulations. For the one- and two-dimensional cases we have already demonstrated that this approximation works very well for the bimolecular annihilation process with equal concentrations of A and B particles.[15,16]

The final set of coupled differential equations is

$$\frac{\partial n_A(t)}{\partial t} = \frac{\partial n_B(t)}{\partial t} = -n_A(t) n_B(t) \int w(\mathbf{r}') Y(\mathbf{r}', t) d^d r', \tag{7}$$

$$\frac{\partial \ln Y(r, t)}{\partial t} = -w(\mathbf{r}) - \int w(\mathbf{r}') Y(\mathbf{r}', t) \{ n_A(t)$$
$$\times [X_A(\mathbf{r} - \mathbf{r}', t) - 1] + n_B(t)$$
$$\times [X_B(\mathbf{r} - \mathbf{r}', t) - 1] \} d^d r', \tag{8}$$

$$\frac{\partial \ln X_{A,B}(\mathbf{r}, t)}{\partial t} = -2 n_{B,A}(t) \int w(\mathbf{r}') Y(\mathbf{r}', t)$$
$$\times [Y(\mathbf{r} - \mathbf{r}', t) - 1] d^d r', \tag{9}$$

where $n_{A,B}(t)$ is the concentration of $A(B)$ particles and $Y(\mathbf{r}, t)$ is the correlation function for unlike particles, i.e., $n_A(t) n_B(t) Y(\mathbf{r}, t)$ is the probability for finding an AB pair with distance $\mathbf{r}$. $X_{A,B}(\mathbf{r}, t)$ are the corresponding correlation functions for AA and BB pairs, respectively.

A simple way to visualize, say Eq. (8), is to observe that a given AB pair may disappear $[\partial(n_A n_B Y)/\partial t]$ either through direct annihilation ($-w n_A n_B Y$) or by having one of the partners annihilate with a third particle, whose position distribution is given in the Kirkwood approximation by a $n_A^2 n_B YYX_A$- or $n_A n_B^2 YYX_B$-type product. The -1 in the square brackets stems from the $\partial n_{A,B}/\partial t$ terms in the $\partial(n_A n_B Y)/\partial t$ expression, where use was made of Eq. (7).

The starting point both for the simulations and also for the analytical development according to Eqs. (7)–(9) is an uncorrelated distribution of A and B particles with initial densities $n_{A,B}(0) = n_{A,B}^0$ and $Y(\mathbf{r}, 0) \equiv X_{A,B}(\mathbf{r}, 0) \equiv 1$. Evidently in the course of the reaction one has at all times

$$\lim_{r \to \infty} X(\mathbf{r}, t) = \lim_{r \to \infty} Y(\mathbf{r}, t) = 1. \tag{10}$$

In the following we use dimensionless variables by measuring distances in units of r_0, concentrations in units of r_0^{-d}, and time in units of w_0^{-1}. Thus the reaction rate is now of the form $w(\mathbf{r}) = \exp(-r)$.

III. COMPARISON BETWEEN SIMULATIONS AND ANALYTICAL APPROACH

Here we present the results of numerical investigations of the annihilation process $A + B \rightarrow 0$ on one- and two-dimensional lattices. The details of the simulation are as given in Ref. 16. As in our previous works we let the parameter r_0 equal $5a$, where a is the nearest-neighbor distance, this choice being representative for findings in several molecular crystals. For equal numbers of A and B particles we have already demonstrated that the agreement between the simulations and the numerical evaluation of Eqs. (7)–(9) is very good, both in as far as the concentrations and also the correlation functions are concerned. In this paper we focus on the case of unequal initial concentrations, $n_A^0 < n_B^0$.

In Figs. 1 and 2 we compare the simulation results to the outcomes of Eqs. (7)–(9) in one and two dimensions. Every simulation started from fixed initial numbers of A and B particles, randomly distributed in the available space. This was either a linear chain with 10^5 sites or a 1000×1000 square lattice, both endowed with periodic boundary conditions. The initial number of A particles was 10^4, whereas the numbers chosen for the B particles were 10^4, 1.5×10^4, and 2×10^4 in $d = 1$ and 10^4, 1.25×10^4, and 1.5×10^4 in $d = 2$. The solid curves a–c in Figs. 1 and 2 are the results of the simulations, each curve being averaged over 10 realizations of the annihilation process. The curves are given as a function of the different initial concentrations of the B particles. In the figures the dotted lines indicate the standard deviation of the simulation data, whereas the dashed lines are the corresponding numerical solutions of Eqs. (7)–(9).

For comparison with earlier results,[15,16] we also present in Figs. 1 and 2 the case of equal initial concentrations, $n_A^0 = n_B^0$, curves a; for these the agreement between simula-

FIG. 2. Same as Fig. 1 but for $d = 2$. Here the initial concentrations are $n_A^0 = 0.25$ (curves a–d) and $n_B^0 = 0.25$ (curve a), 0.3125 (curve b) and 0.375 (curves c and d).

tion and numerical evaluation of Eqs. (7)–(9) is very good and both sets of data follow asymptotically, Eq. (4).

For the case $n_A^0 < n_B^0$ (curves b and c) Eqs. (7)–(9) lead to forms for which the agreement is less impressive. Increasing the difference in the A and B concentrations leads to more marked deviations for the results of the two methods. As a cautionary note, one should, however, remark that in both Figures the ordinate axis is logarithmic; this choice of axes is meant to pictorially enhance any discrepancy. As a matter of fact, for the two sets of data only the relative error increases with decreasing concentration, whereas the absolute error decreases too.

Nonetheless, the agreement between simulation and the analytical approach, based on the superposition approximation, is still very impressive, when one takes into consideration other possible approximate schemes. Thus one may envisage a simpler approximation than Eqs. (7)–(9), in which correlations between like particles are neglected; this corresponds to $X_{A,B}(\mathbf{r},t) \equiv 1$. In this case one has from Eq. (8):

$$Y(\mathbf{r},t) = \exp[- w(\mathbf{r})t]. \qquad (11)$$

Now only Eq. (7), where $Y(\mathbf{r},t)$ is given by Eq. (11), has to be evaluated numerically. The outcome of this lower-level approximation is indicated in Figs. 1 and 2; the results are the curves indicated by d, which were calculated for the same parameters as curves c. Compared to this simpler scheme, the results of the superposition approximation fit the simulation data much better.

On the other hand, this positive result raises the question of possible extensions to our system of Eqs. (7)–(9) by including higher-order terms. From our experience we view this as being a hard task, which also would be very expensive in terms of computer time. Further improvements to the approach presented here, obtained by extending the analytical scheme to include further, higher-order correlation functions seem to us at this stage prohibitive.
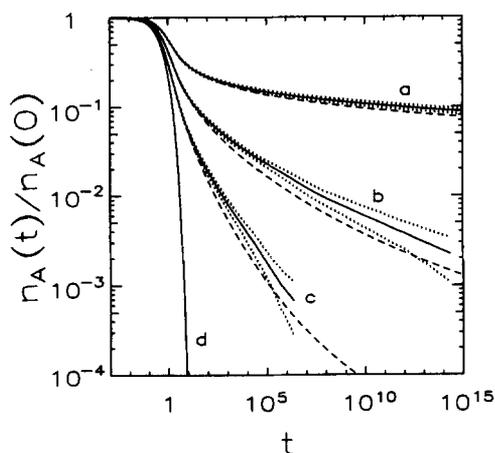

FIG. 1. Decay of the concentration $n_A(t)$ in $d = 1$ resulting from direct simulations of the reaction (solid curves a–c), the corresponding numerical evaluation of Eqs. (7)–(9) (dashed lines), or an even simpler approximation, neglecting correlations between like particles (curve d). The dotted lines indicate the standard deviation of the simulated curves, each of which being the average over 10 realizations of the reaction process. The initial particle concentrations are $n_A^0 = 0.5$ (curves a–d) and $n_B^0 = 0.5$ (curve a), 0.75 (curve b) and 1 (curves c and d).
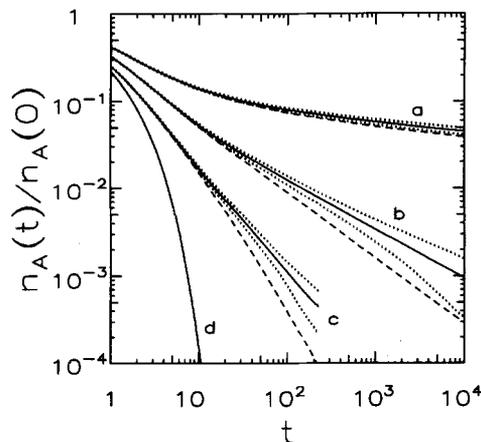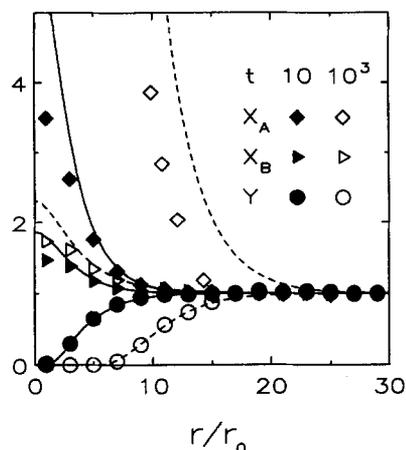
FIG. 3. Correlation functions for like (X_A, X_B) and unlike particles (Y) in $d = 2$, calculated numerically from Eqs. (7)–(9) (solid or dashed curves) or obtained from direct simulations of the reaction (symbols), each value averaged over 10 realizations and over a distance of $2r_0$ ($n_A^0 = 0.25$, $n_B^0 = 0.3125$, $r_0 = 5$). Solid lines and filled symbols belong to $t = 10$, whereas dashed lines and open symbols belong to $t = 10^3$.

Why are in the case $n_A^0 < n_B^0$ the deviations between simulation and analytical approach larger than those for $n_A^0 = n_B^0$? Some insight into this problem may be obtained by analyzing the correlation functions. Thus we have plotted in Fig. 3 the correlation functions which correspond to curve b of Fig. 2. Here the solid and dashed curves are the solutions of Eqs. (7)–(9), whereas the symbols indicate the corresponding simulation results. Although the correlation functions $Y(\mathbf{r}, t)$ for pairs of unlike particles agree very well, there are significant deviations for the correlations between like particles, $X_A(\mathbf{r}, t)$ and $X_B(\mathbf{r}, t)$. This implies that at a given instant of time the predicted size of clusters is greater than what the simulations show, i.e., the functions $X_{A,B}(\mathbf{r}, t)$ of the analysis overestimate the formation of clusters of like particles. This becomes most obvious in the case of $X_A(\mathbf{r}, t)$, the correlation function for the minority species. Whereas for $n_A^0 = n_B^0$, X_A and X_B are identical and grow moderately with increasing time,[15,16] here X_A grows very fast, possibly overstretching the limits of validity of the superposition approximation.

IV. PARALLELS TO DIFFUSION-LIMITED REACTIONS

Let us now make some remarks on the connection to diffusion-limited reactions. As we have already pointed out,[16] for $n_A^0 = n_B^0$ there exists an analogy between the diffusion length $l_D = (Dt)^{1/2}$ and the reaction radius $\xi = \ln t$. In either case the decay law for the concentration may be written as $n \sim l_D^{-d/2}$ or $n \sim \xi^{-d/2}$, respectively. It would be natural to assume that the same analogy holds also in the case $n_A^0 < n_B^0$. For diffusion-limited reactions Kang and Redner suggested the decay law[3]

$$n_A(t) \sim \exp\left\{ - \left[(n_B^0)^{1/2} - (n_A^0)^{1/2} \right] l_D^{d/2} \right\} \qquad (12)$$

and confirmed its validity by computer simulations in one and two dimensions. The idea behind Eq. (12) is the following: In order to obtain the correct decay behavior $n \sim l_D^{-d/2}$ for $n_A^0 = n_B^0$ from a kinetic scheme one can modify Eq. (1) by replacing the reaction constant k by a time-dependent reaction rate $k(t) \sim \partial l_D^{d/2}/\partial t$. Now, assuming that the reaction rate does not change with the initial concentrations and solving the kinetic scheme, Eq. (1), for $n_A^0 < n_B^0$, one arrives at Eq. (12).

As pointed out by Kang and Redner,[3] for diffusion-limited reactions in $d > 2$ Eq. (12) is problematic, since it predicts a decay which is faster than that of the pseudounimolecular trapping reaction $A + B \rightarrow B$, a clearcut contradiction.

Let us now follow the Kang and Redner procedure for the situation discussed here, i.e., immobile reactants. Rewriting Eq. (12) with l_D being replaced by $\gamma \xi = \gamma \ln t$, where γ is constant, we are led to the following approximate decay law for the $A + B \rightarrow 0$ reaction:

$$n_A(t) \sim \exp\left\{ - \left[(n_B^0)^{1/2} - (n_A^0)^{1/2} \right] \times (\gamma \ln t)^{d/2} \right\}. \qquad (13)$$

Interestingly, in our case we do not get any problems by comparison to the $A + B \rightarrow B$ reaction in any dimension. For the $A + B \rightarrow B$ reaction with immobile reactants and exchange-type interaction one has namely the following decay behavior:[4]

$$n_A(t) \sim \exp\left[- \beta (\ln t)^d \right], \qquad (14)$$

where the constant β depends on the concentration of B particles. Thus the decay described by Eq. (14) is faster than that of Eq. (13) in all dimensions, and therefore we do not encounter here the contradiction found by Kang and Redner for diffusion-limited reactions.

Reanalyzing our data in terms of Eq. (13), we found very good agreement with the predicted asymptotic behavior. This may readily be seen in the two-dimensional case (Fig. 2) where (since $d = 2$) one would expect from Eq. (13) a power-law dependence for $n_A(t)$, which is really fulfilled. Moreover, rescaling the time according to

$$\ln t' = \left[(n_B^0)^{1/2} - (n_A^0)^{1/2} \right]^{2/d} \times \ln t,$$

curves b and c almost coincide. With these replacements Eq. (13) results in

$$n_A(t) \sim \exp\left[- (\gamma \ln t')^{d/2} \right]. \qquad (15)$$

In Fig. 4 we replotted curves b and c of Fig. 1 ($d = 1$) with properly rescaled axes. Despite a constant prefactor (which is a constant shift on a logarithmic scale) the two curves are identical in the long-time regime, thus indicating that both curves follow Eq. (15) with the same scaling factor γ.

It thus appears that the relaxation patterns underlying both diffusion-limited reactions and reactions with immobile reactants are related, the connection being provided by quantities which measure the spatial extension of the course of the reactions; here these quantities are l_D or ξ, respectively. A more detailed study of this aspect will be part of our future investigations.
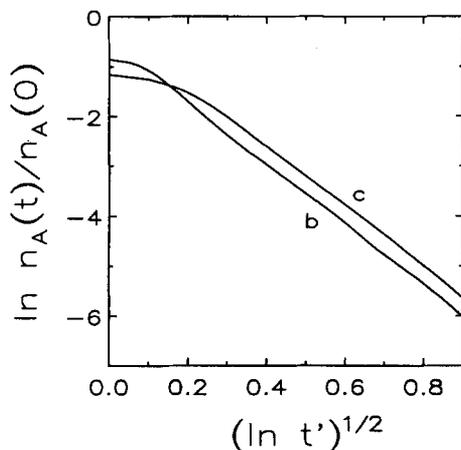
FIG. 4. Simulated curves b and c of Fig. 1, replotted in another presentation, which stresses the connections to diffusion-limited reactions; see text.

V. CONCLUSIONS

In this and the preceding articles[15-17] we studied the kinetics of the bimolecular annihilation reaction $A + B \to 0$ with immobile reactants interacting via exchange, which is typical for solid-state reactions at low temperatures, such as electron scavenging,[9-12] electron–hole recombination,[13,14] or annihilation of radiation-induced defects. An analytical approach to this type of reaction, which was developed by Kuzovkov and Kotomin[8] uses Kirkwood's superposition approximation to decouple the infinite hierarchy of differential equations for the many-point densities $\rho_{m,m'}$. Comparing the outcomes of this analytical approach to the results of direct simulations of the annihilation process, we have shown that very good agreement is obtained for equal initial concentrations $n_A^0 = n_B^0$ of A and B particles. Here the concentration decays as $n_A(t) \sim (\ln t)^{-d/2} \sim \xi^{-d/2}$, with the reaction radius ξ being a consequence of the assumed exchange interaction. In the case of unequal initial concentrations, $n_A^0 < n_B^0$, deviations between analytical approach and simulations are more pronounced, especially in the long-time regime. An investigation of the reasons behind this behavior revealed that the correlation functions $X_A(r,t)$ of the minority species are overemphasized in the analytical approach, a fact which may overstretch the validity of the superposition approximation at long times.

Nonetheless the analytical approach presented here yields very good results in the short and medium time domains, the main range accessible to experiments. Furthermore, the approach is superior to all simpler approximations, which correspond to decoupling at an earlier stage. Further improvements can only be obtained by taking into account higher-order correlation functions, which would drastically increase the numerical effort for solving the set of differential equations, and which may well be beyond practical utility.

A comparison to the results of Kang and Redner[3] for diffusion-limited reactions shows many parallels, if one replaces the diffusion length l_D by the reaction radius ξ. Expressed in these characteristic lengths the decay of concentration has the same form in both types of reactions.

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