Nonequilibrium Dynamics of a Diffusion-Limited Reaction Driven by a Cluster-Memory Mechanism

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The diffusion-limited reaction $A + A \leftrightarrow A + B$ is studied in general dimension. The asymptotic decay of the system is found to depend nontrivially upon the initial concentration of A particles for certain ranges of the diffusion constant, backward reaction rate, and total concentration of particles. This non-equilibrium behavior is due to the formation of clusters centered about the initial A particles. A perturbative analysis in $d = 1$ shows that the transition to the nonequilibrium dynamics is sharp and is quite similar to another previously studied reaction $A + A \leftrightarrow A$. For $d > 1$, a scaling argument is presented which describes the dependence of the asymptotic decay on the initial concentration of A particles and the equilibrium concentration for large backward reaction rates. Monte Carlo data are shown which confirm the analytic work in $d = 1, 2$, and 3.

1. INTRODUCTION

When a chemical system is close to equilibrium, one generally expects the time scales involved to be independent of the initial conditions. This is especially the case if the system evolves from a state in which the constituents are uncorrelated. Typically, such a system will quickly "forget" its initial configuration and the long-time decay modes will be the same as those in a system homogeneously perturbed from its equilibrium state (see, e.g., ref. 1).

There are, however, certain simple reactive systems in which this rule does not hold. These are so-called diffusion-limited systems, [2-5] characterized by time scales dominated by diffusive transport of the constituents,

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879

rather than the reaction time for particles in close proximity. Some of these systems can remember their initial configuration as they approach infinitesimally close to equilibrium. This inherently nonequilibrium behavior is due to the potential of the system to develop interesting spatial features which govern the dynamics.

One such reactive system is the reversible coagulation model $A + A \leftrightarrow A$. In a recent paper, Burschka et al.[6] considered the case where the reaction time in the forward direction is neglected, i.e., the model is purely diffusion-limited. They investigated the evolution of the system from an initial configuration characterized solely by the initial concentration of particles (c_0). With these restrictions, they were able to obtain an exact solution of the model in one dimension. Quite remarkably, they also discovered a "phase transition" in the dynamics, at which the long-time decay mode suddenly changes its dependence upon c_0. Above a certain value of initial concentration, the decay to equilibrium has the usual exponential form $e^{-\lambda t}$, where λ is a constant with respect to c_0. Below this value the decay has the same form, but λ depends nontrivially upon c_0.

The anomalous dependence of the long-time decay of the system $A + A \leftrightarrow A$ upon c_0 is due to the development of distinct regions having a relatively high density of A particles (i.e., clusters). When c_0 is much lower than the equilibrium concentration c_{eq}, the particles in the system at $t = 0$ become centers for these growing clusters as t increases. The analytic solution interestingly shows that the formation of these clusters governs the dynamics asymptotically and that there is a well-defined value of c_0 at which the transition to this new dynamics occurs. However, if c_0 is too close to c_{eq}, only a small number of A particles per unit volume can be added before the system reaches equilibrium. There is then almost no chance that clusters will be formed.

It is natural to ask if the cluster-memory mechanism persists in dimensions other than $d = 1$. At first glance it would appear so, but upon closer examination one finds that this is not necessarily the case. If diffusion is neglected so that the only process taking place is the addition of new particles into the system via the backward reaction, the problem simplifies to one studied some 50 years ago. This is the so-called "rust spot" problem[7,8] where clusters form by spreading from centers randomly placed in the system at $t = 0$. The probability that any point remains uncovered by the spreading rust decays as $\exp(-at^d)$. In $d = 1$ this is in agreement with the analytic result for $A + A \leftrightarrow A$. However, for $d > 1$, the decay due to spreading of clusters is functionally different from the exponential decay one always expects asymptotically when diffusion is present. This would suggest that the long-time decay of the system is not governed by the clusters when $d > 1$.

Nevertheless, some other diffusion-limited systems may preserve the cluster-memory mechanism in higher dimensions as indeed the reaction studied in this paper appears to do. The new model, $A + A \leftrightarrow A + B$, is similar enough to $A + A \leftrightarrow A$ that the same analytic techniques developed by Burshka et al.[6] may be used to solve it. The analysis then shows that an almost identical phase transition occurs in the dynamics of both models for $d = 1$.

In higher dimensions, however, $A + A \leftrightarrow A + B$ appears to be strikingly different. Monte Carlo simulations of the reaction in $d = 2$ and $d = 3$ demonstrate the existence of the cluster-memory mechanism for a large range of initial concentrations. Furthermore, a scaling argument based on a hypothetical structure for the clusters near equilibrium gives the correct scaling as found from the Monte Carlo data.

2. DESCRIPTION OF THE MODEL

The model reaction studied in this paper is $A + A \leftrightarrow A + B$, where the forward reaction $A + A \rightarrow A + B$ is diffusion-limited but the backward reaction $A + A \leftarrow A + B$ is reaction-limited. Physically, this means that the B particles are much more mobile than the A particles and so are always distributed randomly throughout the system. The model is defined on a d-dimensional hypercubic lattice where each lattice site is either occupied by a single A particle or empty: multiple occupation by A particles is not allowed. The B particles are also present on the lattice but because of their high mobility it is only necessary to consider the number of these rather than their specific locations. In effect, this implies that more than one B can occupy a given lattice site even if an A particle is also present; which is entirely consistent with the nature of the backward reaction.

The particles may undergo the following processes:

1. *Diffusion:* An A particle may hop away with a rate $D/(\Delta x)^2$; it then arrives at a randomly selected nearest neighbor. Here, D is the diffusion constant with units of $[L]^2/[T]$.

2. *Forward Reaction:* If an A particle hops via the diffusion process onto a lattice site already occupied by another A particle, the forward reaction immediately occurs, so that only one A particle is left at that site and a B particle is produced.

3. *Backward Reaction:* An A particle at a given site may react with a B particle to produce a new A particle at any one of the nearest-neighbor sites with uniform probability. If another A particle already resides at the chosen site, the forward reaction at that site occurs immediately.

The rate at which the backward reaction occurs is $\gamma c_B(t)/(\Delta x)^{2-d}$, where c_B is the concentration of B particles and γ is the backward reaction constant. The units of γ are $[L]^2/[T]$ and have been chosen so that the model has a valid continuum limit while γ remains finite. Physically, one can understand this choice of units by noting how the backward and forward reactions balance as one approaches the continuum limit. In $d = 1$, the backward reaction rate must diverge because otherwise the diffusive hopping rate, which also diverges, will always consume the new A particles as soon as they are produced. In contrast, for $d > 2$, the diffusive hopping rate does not diverge fast enough to compensate for the production of new A particles, so that the backward reaction rate must vanish with the appropriate power of Δx.

At equilibrium, the system will be in a state of maximal entropy where both the A and the B particles are distributed randomly. The equilibrium concentration of A particles c_A can be calculated in d dimensions by solving for the stationary occupation probability at an arbitrary lattice site s. Denoting the occupation probability by $p(t)$, we write the rate of change of p as

$$\frac{dp}{dt} = \frac{D}{(\Delta x)^2}(1-p)p - \frac{D}{(\Delta x)^2}p + \gamma c_B(\Delta x)^{d-2}(1-p)p \qquad (1)$$

Although Eq. (1) describes a *homogeneous* rather than a diffusion-limited decay, the equilibrium concentration is nevertheless the same.

The first term on the right-hand side is due to particles hopping in from neighboring sites. The second term comes from A particles hopping out of s, while the third term comes from the backward reaction occurring at neighboring sites with the added constraint that the newly created A particle occupies s. The concentration of B particles c_B in the third term may be replaced by $(q - c_A)$, where $q = c_A + c_B$ is the total concentration of particles.

The equilibrium value of p, p_{eq}, can be found by setting dp/dt to zero. The equilibrium concentration $c_{eq} = p_{eq}/(\Delta x)^d$ is then implicitly given by

$$c_{eq} = \frac{\gamma(q - c_{eq})}{D + \gamma(q - c_{eq})(\Delta x)^d} \qquad (2)$$

In the limit $\Delta x \to 0$, one obtains the continuum result

$$c_{eq} \to \frac{q\gamma}{\gamma + D} \qquad (3)$$

whereas the limit $\Delta x \to 1$ leads to the lattice value

$$
c_{eq} \to \frac{(D/\gamma + 1 + q)}{2} - \frac{[(D/\gamma + 1 + q)^2 - 4q]^{1/2}}{2} \tag{4}
$$

3. ANALYTIC ANALYSIS IN $d = 1$

The model can be solved perturbatively near equilibrium, in $d = 1$, by using the formalism developed by ben-Avraham, Burshka, and Doering[6,9] for the diffusion-limited reaction $A + A \leftrightarrow A$. The reader is referred to the above authors for a more detailed account. The model is initially defined on a lattice and then one proceeds to take the continuum limit.

The basic idea is to recast the problem in terms of the empty interval probability $E_n(t)$, defined to be the probability that n successive lattice spacings are devoid of A particles between the times t and $t + \Delta t$. The observation that $E_n - E_{n+1}$ is the probability that n successive sites are empty, with an A particle occupying the $(n + 1)$th site (or, symmetrically, the zeroth site) leads to the following expression for the forward rate of change of E_n:

$$
\left(\frac{dE_n}{dt} \right)_{FR} = \frac{D}{(\Delta x)^2} (E_{n+1} - 2E_n + E_{n-1}) \tag{5}
$$

The production of new A particles via the backward reaction will decrease E_n at the rate

$$
\left(\frac{dE_n}{dt} \right)_{BR} = -\frac{\gamma}{\Delta x} \left(q + \frac{E_1 - 1}{\Delta x} \right) (E_n - E_{n+1}) \tag{6}
$$

In Eq. (6), c_A has been put in the lattice form $(1 - E_1)/\Delta x$. This follows from noting that E_1 is the probability that no A particles are present at an arbitrary lattice site.

In the continuum limit, $E_n(t) \to E(x, t)$ with $x = n \Delta x$. The boundary condition $E(0, t) = 1$ is implemented because the probability of finding an empty interval of length zero must always be unity. The rate equation then becomes

$$
\frac{\partial E}{\partial t} = D \frac{\partial^2 E}{\partial x^2} + \gamma \left(q + \frac{\partial E}{\partial x} \bigg|_0 \right) \frac{\partial E}{\partial x} \tag{7}
$$

The above equation is closed, so that, in principle, one can solve for $E(x, t)$ and from it calculate relevant quantities such as the concentration. The information deriving from higher moments of the density distribution has

been folded into the empty interval probability $E(x, t)$, so that the usual hierarchy of moment equations does not occur. Nevertheless, the scheme is limited in that complete information pertaining to the two-point correlation function (and higher correlations) is unavailable. To obtain the two-point information, for example, one would have to solve an equation for $E(x_1, x_2, y_1, y_2, t)$, the probability of the simultaneous existence at time t of two empty intervals (x_2, x_1) and (y_2, y_1).[10]

The nonlinearity of Eq. (7) makes it difficult to solve exactly. However, the equilibrium empty interval probability $E_s(x)$ is easily obtained, and one can perturb around this function to analyze the long-time decay. Setting the left-hand side of Eq. (7) equal to zero and solving for $E_s(x)$ gives

$$E_s(x) = e^{-[\gamma q/(\gamma + D)] x} \tag{8}$$

This result represents the random distribution of A particles expected at equilibrium and the concentration is given by $\gamma q/(\gamma + D)$. Linearizing Eq. (7) about E_s gives the following equation for the first perturbation of E, E_1:

$$\frac{\partial E_1}{\partial t} = D \frac{\partial^2 E_1}{\partial x^2} + \gamma \left(q + \frac{dE_s}{dx}\bigg|_0 \right) \frac{\partial E_1}{\partial x} + \gamma \left(\frac{\partial E_1}{\partial x}\bigg|_0 \right) \frac{dE_s}{dx} \tag{9}$$

A modal analysis may now be performed by writing E_1 in the form

$$E_1 = \sum_\lambda a_\lambda E_\lambda(x) e^{-\lambda t} \tag{10}$$

This procedure leads to an inhomogeneous ODE in x for each mode:

$$D \frac{d^2 E_\lambda}{dx^2} + \tilde{\gamma} \frac{dE_\lambda}{dx} + \lambda E_\lambda = -\gamma \left(\frac{dE_\lambda}{dx}\bigg|_0 \right) \frac{dE_s}{dx} \tag{11}$$

where $\tilde{\gamma} = D\gamma q/(\gamma + D)$ and q is the total concentration of particles.

Denoting $\gamma (dE_\lambda/dx)|_0$ by r (a number which must be solved for self-consistently) and making the substitution $E_\lambda(x) = [\exp -(\tilde{\gamma}/2D)] F_\lambda(x)$ yields the equation

$$\frac{d^2 F_\lambda}{dx^2} - \left[\left(\frac{\tilde{\gamma}}{2D} \right)^2 - \frac{\lambda}{D} \right] F_\lambda = r \frac{\tilde{\gamma}}{D^2} \exp\left(-\frac{\tilde{\gamma} x}{D} \right) \tag{12}$$

which is an inhomogeneous simple harmonic oscillator equation. It is then straightforward to solve for $F_\lambda(x)$ using, for example, the method of Green's functions:

$$F_{\text{inh}}(x) = \int_0^x ae^{-bx'} G_2(x \mid x')\, dx' + \int_x^\infty ae^{-bx'} G_1(x \mid x')\, dx' \tag{13}$$

where $a = r\tilde{\gamma}/D^2$ and $b = \tilde{\gamma}/2D$.

In the usual manner, the sign of the bracketed factor in Eq. (12) determines whether the solution represents the overdamped or underdamped case. The final results are

$$E_\lambda(x) = Ae^{-bx} \sinh(\alpha x) + \frac{ae^{-2bx}}{b^2 - \alpha^2} - \frac{ae^{-(b+\alpha)x}}{b^2 - \alpha^2}, \qquad \alpha = \left(\frac{\lambda}{D} - b^2\right)^{1/2}; \ \lambda > \frac{\tilde{\gamma}^2}{4D}$$

$$= Ae^{-bx} \sin(\alpha x) e^{-\tilde{\gamma}x/D} + \frac{ae^{-2bx}}{b^2 + \alpha^2} - \frac{a\cos(\alpha x) e^{-bx}}{b^2 + \alpha^2}, \tag{14}$$

$$\alpha = \left(b^2 - \frac{\lambda}{D}\right)^{1/2}; \ \lambda < \frac{\tilde{\gamma}^2}{4D}$$

Although these expressions are slightly more cumbersome than those obtained by ben-Avraham *et al.*,[9] it is evident that they possess the same basic structure. For initial concentrations considerably lower than the equilibrium concentration, small modes are excited in the system. These modes are given by the hyperbolic sine term in the overdamped solution. The smallest mode, and hence the mode generating the longest decay in the asymptotics, is given implicitly by

$$c_0 = b - (b^2 - \lambda/D)^{1/2} \tag{15}$$

Here, c_0 is the initial concentration of A particles.

Solving for λ, one obtains

$$\lambda = Dc_0(c_{eq} - c_0) \tag{16}$$

which is the same expression obtained for $A + A \leftrightarrow A$. The modal analysis strongly suggests that the two models behave in the same manner in $d = 1$. However, because one cannot solve Eq. (7) exactly, it is not possible to prove the existence of the dynamical phase transition for $A + A \leftrightarrow A + B$ as in the case of $A + A \leftrightarrow A$. Nevertheless, the similar structure of the two models near equilibrium, together with the Monte Carlo data discussed in the next section, strongly indicates that the phase transition exists in $A + A \leftrightarrow A + B$.

4. SCALING ARGUMENT FOR $d > 1$

One of the most interesting questions concerns the existence of the dynamic phase transition in higher dimensions. Unfortunately, there is no known analytical formalism for the calculation of relevant quantities in the model $A + A \leftrightarrow A + B$ for $d > 1$. Nonetheless, the model is a good candidate for a reaction having a memory mechanism based on the forma-

tion of clusters. This is particularly so in the large γ/D limit, where discrete clusters form rapidly, seeded by the initial A particles. The new A particles are added to the clusters via the backward reaction much more quickly than the diffusion processes can dissipate the clusters. Consequently, one expects to see well-defined clusters even in the late-stage growth. Whether or not the cluster structure influences the system as t increases without bound is not clear from these qualitative considerations.

If the clusters do indeed drive the long-term behavior of the system, one can develop a phenomenological scaling argument, the predictions of which can be compared with Monte Carlo data. The argument is based on the following assumptions concerning the structure of the clusters near equilibrium:

1. The interior each cluster is homogeneous and in a state of quasi-equilibrium.

2. Each cluster has a well-defined surface having a width ΔR of the same order as the typical distance between particles in the interior.

Under the above assumptions, the growth of clusters will be dominated by the addition of A particles to their peripheries. This process slowly depletes the number of particles in their interiors. The quasi-equilibrium condition implies that one can define an internal concentration $c_i(t)$ for the interior of the clusters. This internal concentration will be somewhat higher than the equilibrium value c_{eq}, whereas the entire system will have a concentration $c_A(t)$ slightly less than c_{eq}. In such a scenario, the slow decay of c_i is given by the equation

$$\dot{c}_i = -\gamma(q - c_A)\, c_i \frac{\Delta R}{R} \tag{17}$$

The above equation is the standard rate equation for the backward reaction applied to the homogeneous distribution of particles assumed to make up the clusters. The backward reaction rate is multiplied by an extra factor $\Delta R/R$ representing the probability that a B particle reacts with an A particle on the outer edge of the clusters.

The radius R is obtained through noting from dimensional analysis that the number of particles in a typical cluster is c_A/c_0. The volume of the cluster must be the number of particles multiplied by the volume per particle c_i^{-1}:

$$R^d = \frac{c_A}{c_0 c_i} \tag{18}$$

The thickness of the outer edge of the typical cluster ΔR must be of the order of $c_i^{-1/d}$. The result is then the equation

$$\dot{c}_i = -\gamma(q - c_A)\, c_i^{1/d} c_0^{1/d} c_A^{-1/d} \tag{19}$$

The quasiequilibrium assumption also allows one to write c_i in terms of c_A in the following manner. For short time intervals, c_i is nearly constant. In contrast to the slow time scales of Eq. (19), the forward and backward reactions almost balance for the short time scales:

$$\dot{c}_i \sim 0 \sim \gamma(q - c_A)\, c_i - D c_i^2 \tag{20}$$

Defining $c_A = c_{eq} - \delta c$ and solving Eq. (20) gives

$$c_i = \frac{\gamma}{D}\,(q - c_{eq} + \delta c) \tag{21}$$

Substituting for c_A and c_i into Eq. (19) and expanding in powers of $\varepsilon = (\delta c)/c_{eq}$ yields the result

$$\dot{\varepsilon} = -D c_{eq}^{-1/d} c_0^{1/d} \left[\frac{1}{d}(q - c_{eq}) + 2 c_{eq} \right] \varepsilon \tag{22}$$

Here, terms of order higher than ε have been neglected. Constant terms have also been dropped, because physically they must be zero so that the system will decay to equilibrium.

When γ/D is large, $q - c_{eq}$ is much smaller than c_{eq}. The first term in the brackets on the right-hand side of Eq. (22) can therefore be neglected. The solution to Eq. (22) is then $\varepsilon = e^{-\lambda t}$, where

$$\lambda = 2(D c_{eq}^{2/d})\, c_{eq}^{1 - 3/d} c_0^{1/d} \tag{23}$$

The factor in parentheses is the natural time step in d dimensions. Measured in these natural time steps, Eq. (23) predicts that the long-time decay mode scales with c_0 by the power $1/d$ and with c_{eq} by the power $1 - 3/d$.

5. MONTE CARLO RESULTS

The analytic results of the two previous sections have been checked directly by comparing them to Monte Carlo simulations. The system used in the simulations consists of a d-dimensional square lattice where A particles are allowed to hop to their nearest-neighbor sites with a uniform probability. In the event that an A particle hops onto a site where another

A particle resides, one of the A particles remains at the site and the number of B particles is incremented by one. The hopping rate is determined by the external parameter D. This is how the diffusion-limited forward reaction $A + A \rightarrow A + B$ is realized.

Each A particle can also react with any one of the B particles to produce another A particle at one of its nearest-neighbor sites. This is the reaction-limited backward reaction $A + A \leftarrow A + B$. The probability of the backward reaction is proportional to the concentration of B particles, with the constant of proportionality γ being the external parameter controlling the reaction rate. The B particles are assumed to be well-mixed in the system at all times, which means that in the simulations one need only keep track of their number, and not their specific positions.

All of the analytic work in the two preceding sections concerns the continuum limit of the lattice model. The simulation results are therefore expected to agree only when the concentration of A particles is low so that the lattice is sparsely occupied. In addition to this limitation, normal finite-system-size effects will cause deviations from the calculations (which assume an infinite system).

Figure 1 shows the results of the $d = 1$ simulation runs. The concentration of A particles was recorded as a function of time, and the long-time tail was fit to an exponential dependence $e^{-\lambda t}$. The value of λ determined from the data has been plotted as a function of the initial concentration of A particles c_0. The solid line in the same figure shows the theoretical
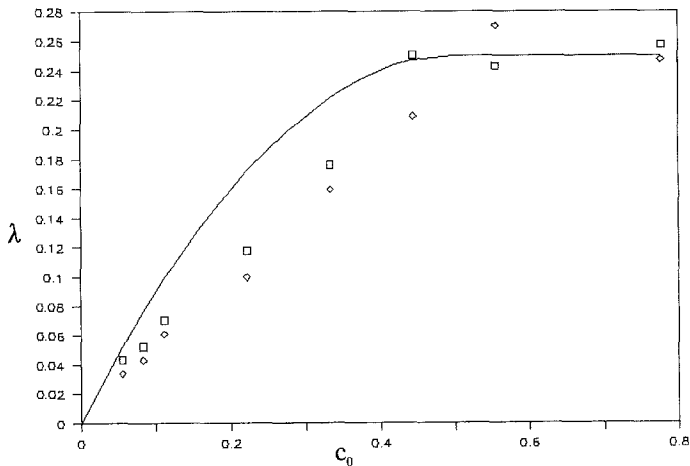

Fig. 1. Asymptotic decay constant λ determined from Monte Carlo concentration data versus initial concentration c_0. The units are Dc_{eq}^2 and c_{eq}, respectively. ($\diamond$) $c_{eq} = 0.1$; ($\square$) $c_{eq} = 0.01$. The solid line indicates the theoretical dependence.

dependence of λ on c_0 as calculated in Section 3. The ratio of γ/D was set to 10. Two equilibrium concentrations were used, as indicated by the two sets of symbols in the figure. Although the experimental data do not fall precisely along the theoretical curve, the behavior is quite suggestive that in the continuum limit the simulations will agree with the theory. The data from runs having a lower equilibrium concentration are noticeably closer to the theoretical curve than the data with a higher c_{eq}. The trend therefore would be for the data to move toward the theoretical curve as c_{eq} is decreased so as to lessen the lattice effects. Data were not taken for lower values of c_{eq} because of computational time constraints.

In order to test the scaling result of Section 4, simulations were run in two and three dimensions with γ/D typically set to 1000. As in the case of the $d=1$ simulations, the large-t dependence of the concentration of A particles as a function of time was fit to an exponential form $e^{-\lambda t}$.

Figure 2 shows the dependence of λ as a function of c_0 with c_{eq} held constant for $d=2$. The data are plotted logarithmically on both scales in the anticipation of a power-law dependence. There are two regimes to the graph. For large c_0, the values of λ appear to be nearly constant, while for smaller c_0 the data fall in a straight line with slope $-1/2$. This value for the slope agrees with the theoretical one obtained from Eq. (23).

The dependence of λ on the c_{eq} for a constant value of c_0 is tested in Fig. 3 for $d=2$. Although there is some curvature in the data for large c_{eq},
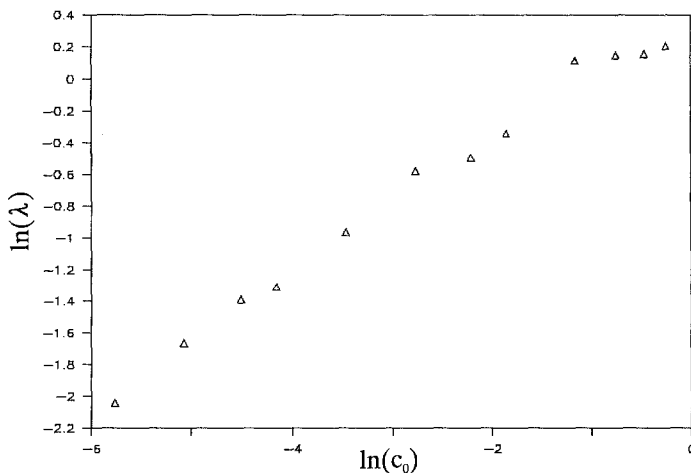

Fig. 2. Asymptotic decay constant determined from Monte Carlo concentration data versus initial concentration c_0 in $d=2$. The units are Dc_{eq} and c_{eq}, respectively, and the slope of the graph is measured to be 0.47 ± 0.03.
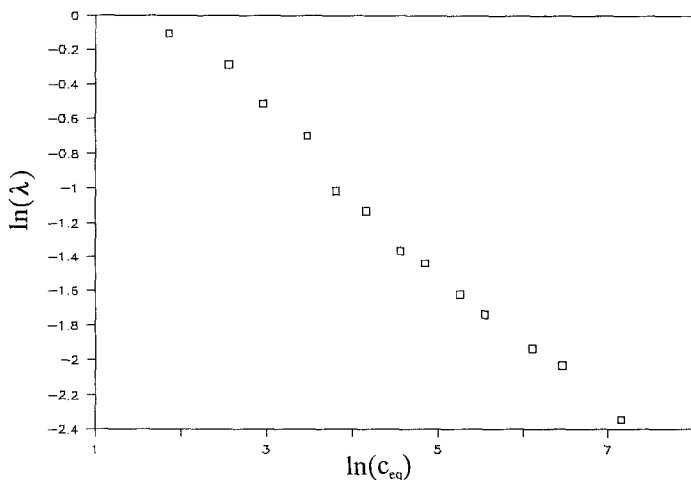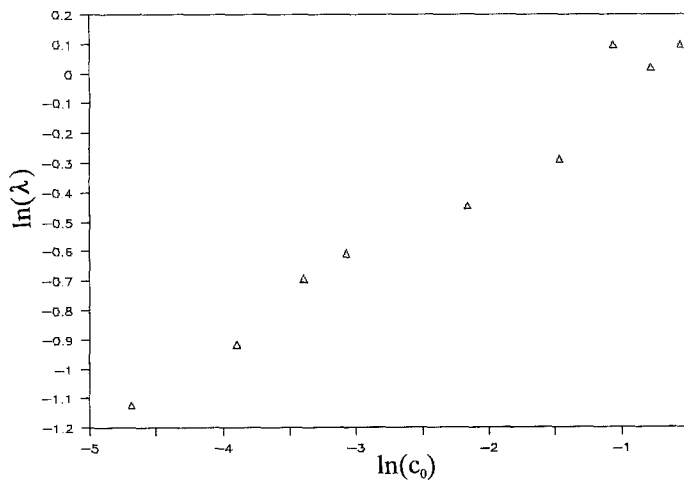
Fig. 3. Asymptotic decay constant determined from Monte Carlo concentration data versus equilibrium concentration c_{eq} in $d = 2$. The units are Dc_{eq} and c_0, respectively. c_0 was set to 1.56×10^{-4} particles per unit volume and the asymptotic slope of the graph is measured to be -0.45 ± 0.06.

Fig. 4. Asymptotic decay constant determined from Monte Carlo concentration data versus initial concentration c_0 in $d = 3$. The units are $Dc_{eq}^{2/3}$ and c_{eq}, respectively, and the slope of the graph is measured to be 0.32 ± 0.02.
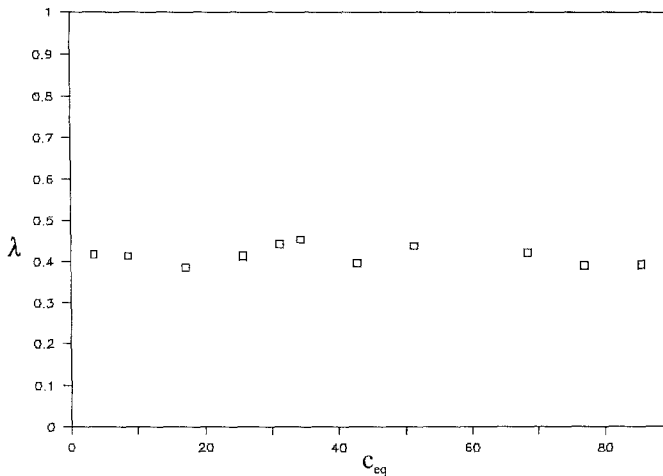
Fig. 5. Asymptotic decay constant determined from Monte Carlo concentration data versus equilibrium concentration c_{eq} in $d = 3$. The units are $Dc_{eq}^{2/3}$ and c_0, respectively. c_0 was set to 2.89×10^{-4} particles per unit volume and the slope of the graph is zero to the accuracy of our computations.

a linear fit to the double-logarithmic plot in the low-c_{eq} range gives a slope quite close to the expected value from Eq. (23) of $+1/2$.

Figures 4 and 5 show the data for simulations in three dimensions. The dependence of λ as a function of c_0 with c_{eq} held constant as shown in Fig. 4 is close to the theoretical prediction of $1/3$ from Eq. (23). Figure 5 gives the dependence of λ on c_{eq} for fixed c_0 and seems to be constant. This is also corroborated by Eq. (23).

6. CONCLUDING REMARKS

The existence of a cluster-memory mechanism in the model $A + A \leftrightarrow A + B$ has been demonstrated for $d = 1, 2$, and 3. For the appropriate range of c_0/c_{eq} the system surprisingly exhibits nonequilibrium behavior in that it always remembers its initial concentration for finite t. More precisely, the smallest mode which determines the asymptotic decay of the system to equilibrium depends upon the initial concentration. Although the cluster-memory mechanism holds, it is not clear whether the phase transition in the dynamics exists in higher dimensions. The transition to the cluster-driven dynamics may not be sharp as it is in $d = 1$.

The scaling argument for $d > 1$ seems to be general enough to give the correct behavior as $d \to \infty$. The geometrical nature of the phenomenological assumptions suggests that an upper critical dimension does not exist.

The assumptions, however, do not hold in $d = 1$. Although there is a cluster memory in all dimensions, the details of its mechanism differ between $d = 1$ and $d > 1$. The models $A + A \leftrightarrow A + B$ and $A + A \leftrightarrow A$ exhibit identical asymptotic behavior in $d = 1$, but not for higher dimensions.

The Monte Carlo data have been taken only when γ/D is large. An open question remains concerning the behavior of the system at small and intermediate values of γ/D. It may be interesting to investigate the complete phase diagram for this model, which may be somewhat richer than the results of the present work.

ACKNOWLEDGMENTS

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