

# Numerical study of $A + A \rightarrow 0$ and $A + B \rightarrow 0$ reactions with inertia

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(Dated: July 9, 2007)

Using numerical methods we study the annihilation reactions  $A + A \rightarrow 0$  and  $A + B \rightarrow 0$  in one and two dimensions in the presence of inertial contributions to the motion of the particles. The particles move freely following Langevin dynamics at a fixed temperature. Our focus is on the role of friction.

PACS numbers: 05.40.-a, 82.20.Fd, 82.20.-w, 82.20.Uv

## I. INTRODUCTION

The simplest irreversible bimolecular reactions in low dimensions and in other constrained geometries exhibit “anomalous” behavior in the sense that they do not obey the macroscopic laws of mass action. The bimolecular reaction  $A + B \rightarrow 0$  has been studied in great detail in the diffusion-limited regime on a large number of geometries and with different initial distributions. This reaction has been analyzed theoretically, numerically, and/or experimentally in regular and in fractal geometries, on ordered and disordered systems, in configurations in which the reactants are initially mixed and also in initially separated configurations leading to chemical fronts. The results offer a vast and interesting collection of different temporal and spatial behaviors [1–13]. It is noteworthy that almost all results that can be found about this reaction are restricted to the diffusion-limited regime, a limit in which inertial effects play no role.

The  $A + A \rightarrow 0$  and  $A + A \rightarrow A$  reactions have also been extensively studied [2, 14–23]. In the diffusion-limited regime these reactions in one dimension have provided a wealth of information because they can be solved exactly for the concentrations as a function of time. In addition, for the  $A + A \rightarrow A$  it is also possible to solve for the spatial distribution of reactants as a function of time in one dimension [17–21]. Again, these time dependences and distributions deviate from those underlying the macroscopic laws of mass action. The exact results have provided a wealth of information for testing numerical methods and analytic approximations [15, 22].

The  $A + A \rightarrow 0$  reaction has also been studied in one dimension in the ballistic limit, that is, the limit in which the particles move ballistically until they encounter and annihilate one another [23–33]. Here one must not only deal with spatial distributions of reactants but as well with their velocity distribution, which also directly affects the reaction kinetics. In any case, in this regime the reaction again does not follow the macroscopic laws

of mass action.

Numerical simulation results for these reactions in general involve extensive molecular dynamics or Monte Carlo simulations [34]. These simulations are extremely valuable because of the dearth of exact analytic results, and because the analytic results are limited to particular regimes, for example, diffusion-limited or ballistic. Recently we introduced a simple Langevin picture according to which the reactants move in space according to the classical laws of motion in a thermal environment and react upon encounter [35]. In the simplest representation one can imagine the reactants to be hard sphere particles that only interact (and react) upon encounter, so that between encounters the equation of motion of each reactant particle in, say, one dimension might be the simple Langevin equation

$$m\ddot{x} = -\gamma\dot{x} + \xi(t). \quad (1)$$

Here  $x$  is the position of the particle,  $m$  is the mass of the particle (which we set to unity),  $\gamma$  is the coefficient of friction, and  $\xi(t)$  is a white thermal noise that obeys the fluctuation-dissipation relation at temperature  $T$ ,

$$\langle \xi(t)\xi(t') \rangle = 2\gamma k_B T \delta(t - t'). \quad (2)$$

It is of course straightforward to extend this to higher dimensions and to include a force  $-V'(x)$  on the right hand side of the equation. The advantage of this approach is that it is numerically far less intensive than other simulation methods and that it allows a straightforward study of inertial effects (and the effects of an external potential  $V(x)$ , which we will not pursue in this paper).

Our purpose here is to investigate the effects of friction, and therefore  $\gamma$  will be our only free parameter. Our previous study [35] focused on the reactions  $A + A \rightarrow 0$  and  $A + A \rightarrow A$  in one dimension, but only in the limits in which there are analytic results available for comparison. In particular, we associated our single model with both limiting scenarios by assuming (and confirming via our

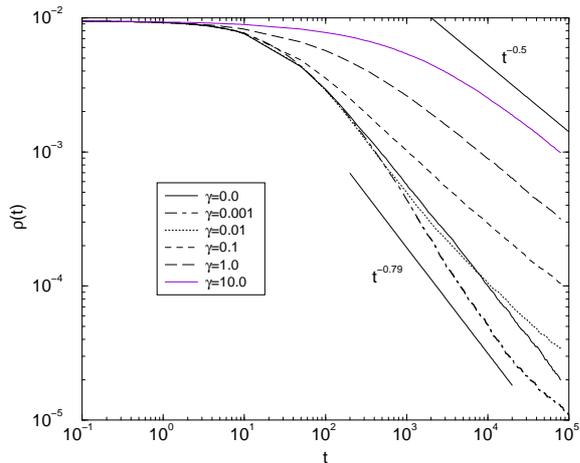


FIG. 1: Dynamical evolution of the density of  $A$  particles in the  $A + A \rightarrow 0$  reaction in one dimension for different values of the friction. Initially there are 100,000 particles in an interval of length  $L = 2,097,152$  so that the initial density is  $\rho(0) = 0.00954$ . The slopes for the overdamped limit  $\gamma \rightarrow \infty$  and the ballistic limit  $\gamma = 0$  are indicated for comparison.

simulation results) that the diffusion-limited regime is recovered when  $\gamma$  is large, and that the ballistic behavior is recovered in the limit  $\gamma = 0$ . In those simulations as well as here we need to specify a number of other parameter choices. In particular one must choose the temperature (we set  $k_B T = 0.2$ ), the radius  $R$  of the reactants (we set  $R = 0.1$ , see the appendix for a rationale), and the probability  $p$  of a reaction upon collision (we set  $p = 1$ ). The particles in  $d$ -dimensions move in a system of size  $L^d$ , and initially  $N$  particles are placed in the system, so that the initial (dimensionless) concentration  $\rho(t = 0)$  is  $N/(L/2R)^d$ . One must specify the initial position (random) and velocity (Maxwellian) distributions of the reactants. Other details of the simulation are presented in the appendix.

In Sec. II we present our results for the  $A + A$  reaction in one and two dimensions, and in Sec. III those for  $A + B$ . We end with a brief recap in Sec. IV.

## II. $A + A$ REACTION

### A. Dimension $d = 1$

We start with the  $A + A \rightarrow 0$  reaction in one dimension by showing the ubiquitous measure of reaction kinetics, that is, the time dependence of reactant concentrations,  $\rho(t)$ . In Fig. 1 we see the results for the density of  $A$  particles in the  $A + A \rightarrow 0$  reaction for different values of the friction. This figure makes evident the interplay of a number of different time scales. First, at early times  $\rho(t)$  remains essentially constant because for the reaction to start the reactants must meet. One can estimate this

time by comparing the root mean square displacement, easily calculated from Eq. (1), with the inverse of the initial density since this is a measure of the distance that must be covered by a reactant for an encounter. For small friction the motion is initially essentially ballistic,  $\langle x^2 \rangle \sim k_B T t^2$ , where we have set the root mean square velocity  $\langle v^2 \rangle = k_B T$ . The time  $t_0$  at which  $\langle x^2 \rangle^{1/2}/2R \sim 1/\rho(0)$  for the parameters used here is of  $O(10^1)$ . This is consistent with the low  $\gamma$  curves in the figure. For large friction the motion is diffusive,  $\langle x^2 \rangle \sim k_B T t/\gamma$ , so that  $t_0 \sim O(10^3)\gamma$ . Again, this is consistent with the high  $\gamma$  curves in the figure.

The predicted slope for the long-time decay of the particle density in the diffusion-limited regime is  $d/2 = 0.5$  [2, 14]. For the ballistic reaction several values have been predicted in the literature including 0.666 [25, 26, 29], 0.769 [30], and 0.805 [23]. Our  $\gamma = 0$  simulations lead to the value 0.79. For  $\gamma = 10$  the diffusion-limited slope is reached very quickly once the concentration starts to decay. For lower values of the friction we observe transient behavior akin to the ballistic regime, which lasts until  $t \gg O(1/\gamma)$ . Thereafter, the kinetics are dominated by diffusive behavior. Thus, for  $\gamma = 0.001$  we see the slope beginning to bend over at times  $t > 10^3$ , while for  $\gamma = 0.01$  this behavior is already evident at times  $t > 10^2$ . We thus conclude that the density remains essentially constant until reactants first meet at time  $\sim t_0$ , which is longer for larger friction; that there is an intermediate regime between  $t_0$  and a time greater than  $1/\gamma$  where the behavior is essentially that of a zero-friction system, after which the system behaves diffusively. The intermediate transient is longer for lower friction. Note that there is yet another time scale in the problem, of course, and that is the time for the reactants to be consumed altogether. This depends on the initial concentration, and it may thus happen that for low friction the diffusive behavior never becomes apparent.

In a diffusion-limited  $A + A \rightarrow 0$  reaction one does not need to keep track of particle velocities. On the other hand, in the ballistic case the distribution of velocities can change as the reaction proceeds. If the initial velocity distribution is simply dichotomous, e.g., half of the particles start out with velocity  $v$  and the other half with  $-v$  (a case that has been studied analytically in some detail), then the distribution does not change in time since pairs of particles with opposing velocities annihilate with each reaction event [23–32]. On the other hand, if the particles have a continuous velocity distribution such as Maxwell-Boltzmann, the system cools down, the distribution become distorted as the particles with higher velocities react first, and the distribution approaches a  $\delta$  function at zero velocity [23, 25, 26, 29–33]. The question then is, what happens at intermediate values of friction?

In Fig. 2 we show the evolution of the mean square velocity of the surviving reactants as a function of time for different values of the friction. The mean square velocity is a measure of the system temperature. As expected, when the friction is very high (higher than the largest

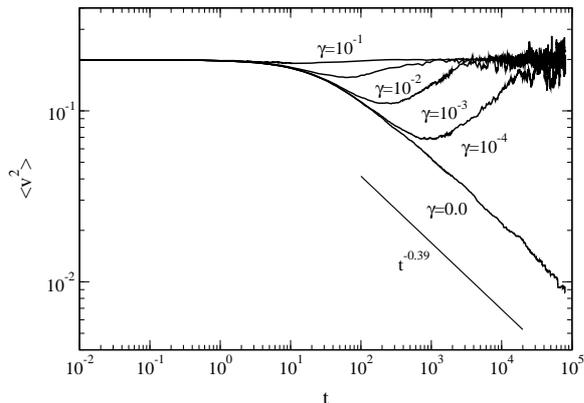


FIG. 2: Evolution of  $\langle v^2 \rangle$ , proportional to the system temperature, for the reaction  $A + A \rightarrow 0$  for different values of the friction parameter. At infinite friction this value remains constant, and at zero friction it decays according to a power law. The predicted power law is indicated in the figure.

value shown here) the mean square velocity remains constant. At zero friction, the system progressively cools down as the fastest particles react, and the mean square velocity exhibits a power law decay. Theoretical predictions for the sum rule  $\xi + \alpha = 1$  involving the exponent  $\xi$  for the decay of  $\rho(t)$  and the exponent  $\alpha$  for that of  $\langle v^2(t) \rangle$  are satisfied if we set the exponent here equal to 0.39. The interesting behavior occurs at low but non-zero friction. There is of course no cooling until the particles can meet and react; the flat portion of the curve goes up to time  $\sim t_0$ . The system then begins to cool down, but since it is in a thermal environment the energy of the particles is replenished and eventually thermal equilibrium is re-established. The transient cooling period (followed by warming) is longer for smaller friction, and the time scale for this behavior is again the frictional time scale  $1/\gamma$ . As before, it is of course possible (depending on initial concentrations) that the reactants are consumed altogether before re-equilibration can occur.

A quantity that is frequently studied to understand the source of the deviations from classical behavior is the spatial distribution of reactants, in particular, the probability distribution for the separation between neighboring particles. In our earlier work [35] we addressed the differences between the diffusion-limited and ballistic behaviors in the evolution of this distribution function. In the former, it is well known that a gap develops between neighboring reactants as pairs of proximate  $A$ 's react and diffusion is slow to bring new pairs together. An initial Poisson distribution has a peak near the origin, which indicates many adjacent  $A - A$  pairs. As the gap develops, the peak moves away from the origin, indicating a growing finite distance between nearest reactants. The result is a rather structured lattice-like configuration of reactants. This same behavior is apparent in the Langevin model at high friction, as shown in the upper

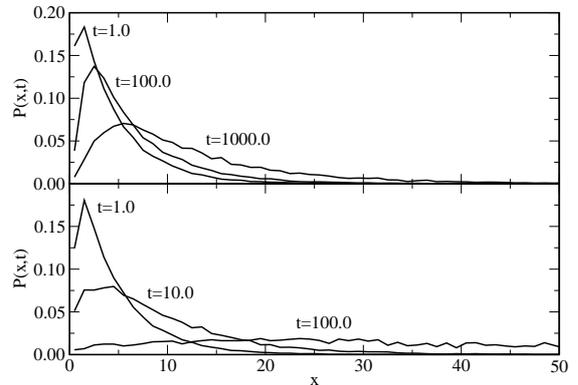


FIG. 3: Interparticle distribution functions for the reaction  $A + A \rightarrow 0$  for  $\gamma = 50.0$  (upper panel) and  $\gamma = 0.1$  (lower panel).

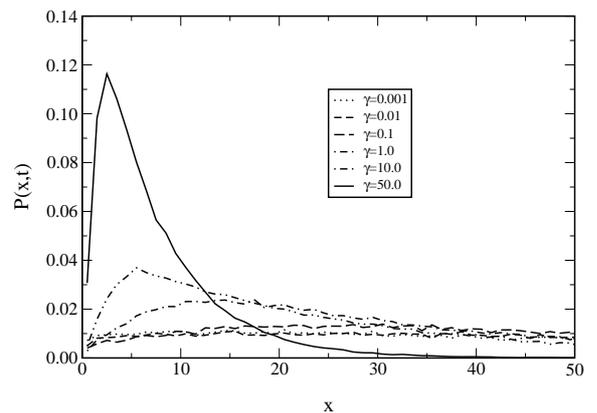


FIG. 4: Interparticle distribution function for  $A + A \rightarrow 0$  for several values of the friction parameter at time  $t = 100$ .

panel of Fig. 3. On the other hand, in the ballistic limit the distribution becomes homogeneous (rather than random), that is, one in which all interparticle distances become equally probable. This behavior is observed in the Langevin model at low friction, as seen in the lower panel of Fig. 3. Figure 4 shows the interparticle distribution function for different values of the friction at the same fixed time, thus confirming the more structured configuration at high friction and the essentially homogeneous distribution at low friction.

## B. Dimension $d = 2$

For the  $A + A \rightarrow 0$  reaction,  $d = 2$  is the critical dimension for classical asymptotic behavior in the diffusion-limited case as well as in the ballistic case. This means that the asymptotic decay of the density in both lim-

its should go as  $t^{-1}$ . Numerical results for this scenario are shown in Fig. 5. At early times the same arguments used earlier apply, so that the initially unchanging density is due to the time it takes for particles to first meet. The low friction curves all start decreasing at the same time because the time for first encounter is dominated by the inertial contribution that is independent of friction. The curves for higher friction remain constant for a longer time since the time for first encounters grows as friction increases. Note that while the decay of the density is essentially classical no matter the friction, the velocity distribution does reflect the effects of friction, as shown in Fig. 6. We clearly see that at high friction the second moment of the velocity distribution remains constant, indicating that the system velocities are in thermal equilibrium at all times. At lower friction we observe the cooling and re-equilibrating behavior, and again the time scale for this behavior is  $1/\gamma$ . At zero friction there is of course no re-equilibration.

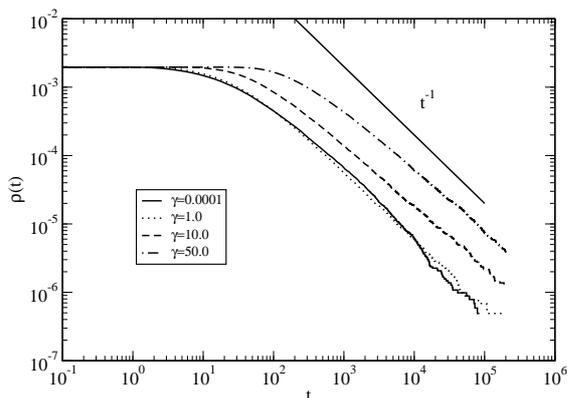


FIG. 5: Evolution of the density of  $A$  particles for the  $A + A \rightarrow 0$  reaction in two dimensions for different values of the friction. Up to  $\gamma \sim 1$  the curves for different  $\gamma$  are essentially indistinguishable. The initial density is  $\rho(0) = 1.96 \times 10^{-3}$ .

### III. $A + B$ REACTION

Let us now turn to the  $A + B \rightarrow 0$  reaction. While the detailed dynamics and the source of the deviations from macroscopic mean field behavior are different in this reaction than in the  $A + A$  system, one would expect that the effects of friction would be similar. No time dependences of any kind should set in until the reactants can reach each other, and the arguments giving the appropriate friction-dependent time scale  $t_0$  here are the same as in the  $A + A$  reaction. Furthermore, at zero friction the ballistic motion determines the behavior of the system at all times, and the asymptotic behavior at finite friction should be the same as in the diffusion-limited regime. These asymptotic behaviors are different than those of the  $A + A$  reaction. Between the early and late

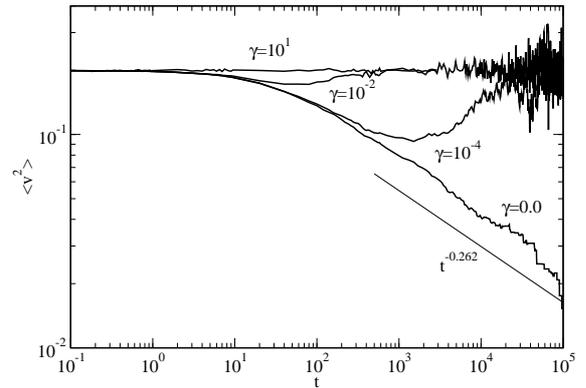


FIG. 6: Evolution of  $\langle v^2 \rangle$  for the reaction  $A + A \rightarrow 0$  in two dimensions for different values of the friction parameter. The power 0.262 is a numerical fit to the  $\gamma = 0$  data.

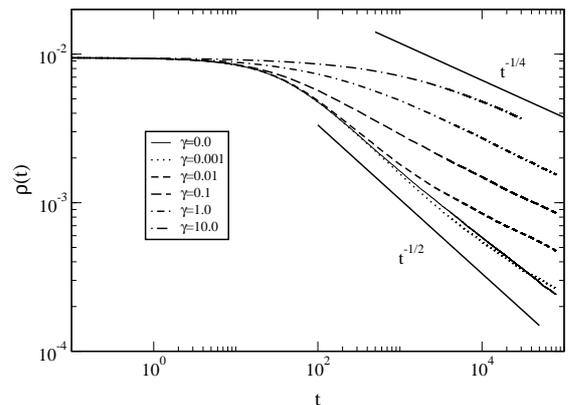


FIG. 7: Dynamical evolution of the density of  $A$  particles in the  $A + B \rightarrow 0$  reaction in one dimension for different values of the friction. Initially there are 100,000 particles, half of each species, in an interval of length  $L = 2,097,152$  so that the initial density of each species is  $\rho(0) = 0.00477$ . The predicted behaviors  $t^{-1/4}$  for the overdamped limit  $\gamma \rightarrow \infty$  and  $t^{-1/2}$  for the ballistic limit  $\gamma = 0$  are also indicated.

time behaviors we expect a transient regime starting at  $t_0$  and ending at  $t > 1/\gamma$  where the behavior is essentially ballistic.

Before discussing the simulation results, we note that contrary to the  $A + A \rightarrow 0$  case, we are aware of no analytic theory to predict the  $A + B \rightarrow 0$  reaction kinetics in the ballistic limit (some predictions can be found for these reactions occurring under Lévy mixing [36]). Nevertheless one can provide a simple scaling argument along the lines of the Toussaint and Wilczek argument [5] that leads to the correct exponent in the diffusion-limited regime. An initially random distribution of particles of density  $\rho$  will exhibit local fluctuations so that in a region of size  $l^d$  ( $l$  is a distance scaled by the particle diameter) one has on the order of  $\rho^{1/2}l^{d/2}$  more particles of one

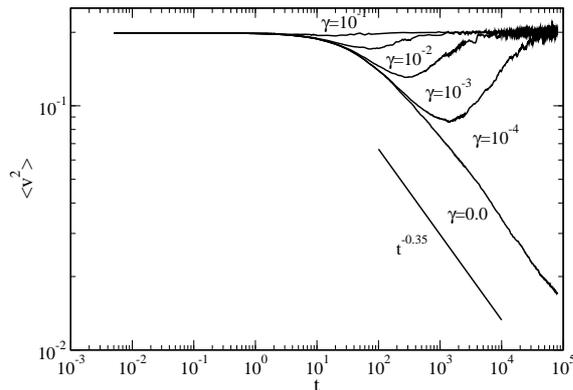


FIG. 8: Evolution of  $\langle v^2 \rangle$  for the reaction  $A + B \rightarrow 0$  for different values of the friction parameter. The value associated with the power law decay at zero friction is a numerical fit.

species than the other. The majority particles will eliminate the minority in a time that it takes to move over the distance  $l$ . In the diffusive case this time is  $t \sim l^2$  and so this corresponds to a remaining density  $\rho \sim t^{-d/4}$ . For ballistic motion the distance  $l$  is covered in a time  $t \sim l$ , which leads to a density decay that goes as  $\rho \sim t^{-d/2}$ . For  $d = 1$  these decays thus go as  $t^{-1/4}$  and  $t^{-1/2}$  respectively.

#### A. Dimension $d = 1$

Figure 7 shows the evolution of the mean density of the reactant  $A$  in the  $A + B \rightarrow 0$  reaction, and exhibits all the behaviors and time scales anticipated above. In these simulations we have set the parameters  $m$  and  $\gamma$  of both species to be equal, and have chosen equal initial densities. In agreement with the above speculations, the density  $\rho(t)$  of each reactant is constant up to time  $\sim t_0$ , decays as  $\sim t^{-1/2}$  up to times around  $1/\gamma$ , and decays as  $t^{-1/4}$  thereafter. We again expect the most rapid particles to react first, and the system to cool down until the velocity distribution can become equilibrated after the transient time of  $O(\gamma^{-1})$ . The mean of the square velocity confirming this behavior is shown in Fig. 8. There is no theoretical prediction for the cooling exponent in the zero friction case. The value shown in the figure is a numerical fit, and might be the same as that of this regime for the  $A + A$  reaction.

#### B. Dimension $d = 2$

For the  $A + A$  reaction,  $d = 2$  is the critical dimension for normal classical law of mass action behavior, but for the  $A + B$  reaction it is  $d = 4$ , so it is interesting to check whether this model captures the features discussed above but for two dimensions. Figure 9 shows the evolution of

the density for the  $A + B$  reaction in this case. The predicted diffusion-limited and ballistic slopes are also indicated. Note that while we would expect diffusive behavior at long times in all cases, the reaction is essentially completed in the low friction cases before the change in slope from ballistic,  $t^{-1}$ , to diffusive,  $t^{-1/2}$ , can be observed. Associated mean square velocity simulations are shown in Fig. 10.

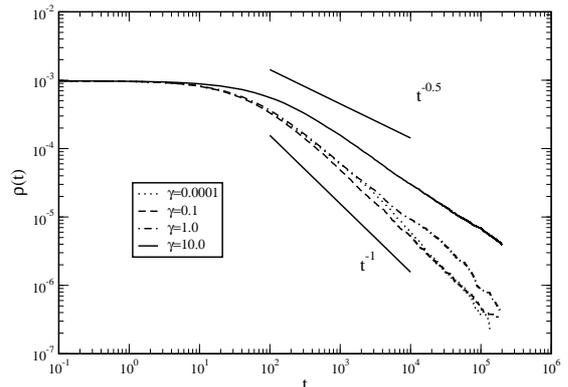


FIG. 9: Density of  $A$  particles in the  $A + B \rightarrow 0$  reaction in two dimensions for different values of the friction. The theoretical overdamped and infradamped limits are also plotted for comparison.

## IV. RECAP

We have presented a numerical study of the  $A + A \rightarrow 0$  and  $A + B \rightarrow 0$  annihilation reactions in dimensions  $d = 1, 2$  based on a Langevin approach that can subsume known results in appropriate limits but that allows us to explore regimes not previously investigated. The Langevin equation includes inertial terms and frictional

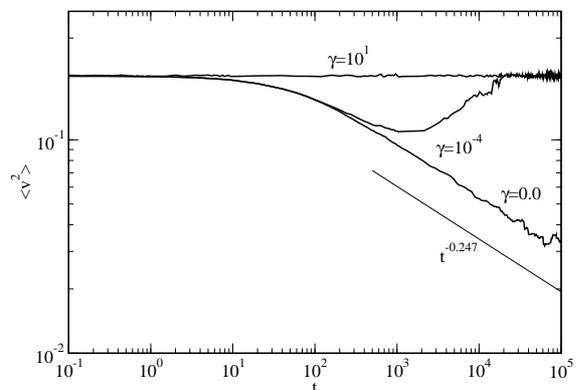


FIG. 10: Evolution of  $\langle v^2 \rangle$  for the reaction  $A + B \rightarrow 0$  in two dimensions for different values of the friction parameter. The power 0.247 is a numerical fit to the  $\gamma = 0$  data.

$t^{-\beta}$	$\rho(t)$		$\langle v^2(t) \rangle$	
	Diff-lim	Ballistic	Diff-lim	Ballistic
$A + A$				
$d = 1$	1/2	0.79	0	0.39
$d = 2$	1	1	0	0.26*
$A + B$				
$d = 1$	1/4	1/2	0	0.35*
$d = 2$	1/2	1	0	0.25*

TABLE I: Exponents  $\beta$  for the asymptotic behavior of  $\rho(t) \sim t^{-\beta}$  or  $\langle v^2(t) \rangle \sim t^{-\beta}$ . Values indicated as integers or whole fractions are associated with well-grounded theories or scaling arguments. Decimal values are our numerical fits; those without an asterisk are consistent with existing theories. The asterisk denotes cases for which we are not aware of any theory in the literature.

contributions with friction coefficient  $\gamma$ . The particles are initially randomly positioned in the system, with a Boltzmann velocity distribution. In the high friction limit we recover well-known diffusion-limited behavior, and in the zero friction limit the results of ballistic kinetics are observed. These latter results are known for the  $A + A \rightarrow 0$  reaction but are new for the  $A + B \rightarrow 0$  case. A summary of these limiting behaviors is collected in Table I. The intermediate friction results are all new.

The decay of the particle density in the  $A + A \rightarrow 0$  reaction in the high friction limit thus goes as  $\rho(t) \sim t^{-d/2}$ , and the interparticle distribution function in one dimension exhibits the well-known lattice-like structure as a consequence of the evolution of interparticle gaps. The velocity distribution and associated particle temperature remain fixed throughout the reaction process, that is, the system is thermalized at all times.

At zero friction for the  $A + A \rightarrow 0$  reaction we recover the decay law  $\rho(t) \sim t^{-0.79}$ , which is an exponent within the range of values found by other methods that deal directly with ballistic kinetics. At nonzero friction there is a transient regime for  $t < \gamma^{-1}$  that exhibits this decay, and beyond this regime the decay becomes diffusive (as long as there are any particles left in the system). The interparticle distribution function becomes uniform at long times, indicating that all interparticle distances become equally probable. The velocity distribution exhibits interesting behavior. At zero friction the system cools down as the reaction proceeds because the fastest particles encounter other particles first and are thus eliminated from the system, and there is no mechanism for input of thermal energy. The temperature of the system decays as  $t^{-0.39}$ , again consistent with known results. At nonzero friction we find that there is a cooling down period where the temperature decays in this way. This period is of the order of  $t \sim \gamma^{-1}$ , after which thermalization brings the system back to the original temperature.

For the  $A + B \rightarrow 0$  reaction in the high friction limit the density decays as  $\rho(t) \sim t^{-d/4}$ , a well-known result in

the diffusion-limited regime. At zero friction the decay goes as  $\rho(t) \sim t^{-d/2}$ , a result for which we offer a scaling argument. For nonzero but small friction the decay follows the ballistic behavior for times below  $\gamma^{-1}$  and then goes over to the high-friction behavior provided there are any particles remaining. The temperature behaves as in the  $A + A$  reaction: it remains fixed at sufficiently high friction, the system cools down at zero friction, and at intermediate friction the system first cools down, and at times  $t > \gamma^{-1}$  it is able to rethermalize to the original temperature.

Our Langevin equation formulation of the problem allows us to easily include a substrate potential. This, along with the inclusion of reverse steps and of reactions described by more complex mechanisms, is among our plans for future work.

### Acknowledgments

This work was partially supported by the National Science Foundation under Grant No. PHY-0354937 (K.L.), by the UC Institute for Mexico and the United States (UC MEXUS) (A.H.R. and K.L.), CONACYT-Mexico Project No. 48783-F (A.H.R.), and by the MCyT (Spain) under Project No. FIS2006-11452 (J.M.S. and A.M.L.).

### Appendix

In this appendix we provide some of the details of our numerical solution of the set of Langevin equations (1) (or their two-dimensional generalization), one for each particle, and the implementation of the reaction scheme between particles.

In general, to test for system size effects (and to avoid such effects), in some cases we have increased both  $L$  and  $N$  so as to preserve the initial concentration and ascertained that there are no changes in the results. We implement periodic boundary conditions. A reaction is defined to occur if the distance between the centers of two particles falls below one particle diameter (since integration updates take place at discrete times, particles may interpenetrate during one integration step). A comment on our particular choice of  $R$  ( $R = 0.1$ ) is in order. It must be sufficiently large so that in one integration step of time  $\Delta t$  the Langevin dynamics do not move the particles so far that they are likely to “cross” paths through each other, i.e., we must choose  $R$  so that  $\langle v^2 \rangle^{1/2} \Delta t \ll 2R$ . Here  $\langle v^2 \rangle^{1/2}$  is the root mean square velocity of the ensemble, used as a measure of the “typical” velocity. On the other hand, for comparison with diffusion and kinetic theories  $R$  must be sufficiently small so that the spatial distribution is minimally distorted by finite particle size effects when compared to continuum particle densities. We have ascertained that the chosen  $R$  meets these requirements, and that changing it within

reasonable bounds does not materially affect our results. Upon encounter, the reacting partners simply disappear.

Initially the particles are deposited randomly aside from the fact that we do not allow particle overlap. If we did allow overlap, the initially overlapping pairs would react immediately and a considerable number of particles would disappear, thus effectively reducing the initial concentration. Not allowing an overlap causes our distribution to differ from a continuous Poisson form only at very small interparticle distances, which affects the evolution of the distribution only for very short times. The particles have an initial Maxwell-Boltzmann velocity distribution  $P(v, 0)$  whose second moment  $\langle v^2 \rangle$  is associated with the temperature  $T$ .

Once the particles are deposited, we numerically integrate the Langevin equation for each particle using the Heun method, which is an extension of a second order

Runge-Kutta algorithm for stochastic differential equations [37]. The time step is  $\Delta t = 0.005$ , which we ascertain to be sufficiently small for the desired accuracy of our results. After each integration step we check for all pairs of particles that are sufficiently close (overlapping) and remove them from the system. To improve the speed of this procedure, we have implemented the “method of the neighboring list,” which assigns particles to pre-defined boxes. Only particles within the same box and in neighboring boxes are considered when checking for reactive configurations. The neighboring list is updated every 10 iterations.

As the integration and reaction steps proceed, we keep track of all particle positions and velocities and associated histograms, and continue until the number of particles becomes smaller than a predetermined number or until a predetermined maximum time is reached.

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