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This project was devoted to a study of the kinetics of diffusion-controlled and surface catalytic reactions. For catalytic reactions, the kinetics of the saturation process was elucidated. The role of interactions between adsorbates on the surface reaction was examined. For diffusion-controlled reaction, a comprehensive theoretical framework to describe the spatial organization of reactants was developed. Particular attention was devoted to understanding the reaction dynamics at an interface which separates two reactive species. Geometrical features of this interface and the time dependence of the reaction rate were determined.

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KINETICS AND DYNAMICS OF REACTING SYSTEMS

FINAL REPORT

Sidney Redner

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1. Foreword

There were two major results in this project. First, we elucidated the kinetics of surface reaction models which serve as idealization of catalytic reactions. We determined the kinetics of the saturation process, whereby the surface becomes filled by a single species and no further reaction is possible. We also examined the role of interactions between adsorbates on the reaction kinetics. Second, we developed a comprehensive theoretical framework to describe the spatial organization of reactants in two-species annihilation, $A + B \rightarrow 0$, and in related reactions. Additionally, we investigated the reaction dynamics at an interface which separates a population of A's and B's. We determined geometrical features of this interface and the time dependence of the reaction rate.

These topics are treated in more detail in the following report. In Sec. 2, we describe the scaling behavior and non-equilibrium phase transitions of surface reaction processes, and in Sec. 3, we discuss the wide variety of interesting features associated with the spatial organization of reactants in two-species annihilation.

2. Kinetics of Catalytic Surface Reaction Models

(a) Kinetics of saturation in the monomer-monomer model

In the monomer-monomer model,[1-3] two reactive species, A and B, adsorb irreversibly onto single sites of a catalytic substrate and nearest-neighbor AB pairs can bond to form a reaction product which desorbs from the substrate. This bimolecular surface reaction of the two adsorbed monomeric species can be represented by the following steps:

$$\begin{aligned} \mathrm{A} + \mathrm{S} &\xrightarrow{k_\mathrm{A}} \mathrm{A}_s \\ \mathrm{B} + \mathrm{S} &\xrightarrow{k_\mathrm{B}} \mathrm{B}_s \\ \mathrm{A}_s + \mathrm{B}_s &\xrightarrow{k_r} \mathrm{AB} \uparrow + 2\mathrm{S}. \end{aligned} \tag{1}$$

Here A and B denote the two species in the gas phase, S denotes a free surface site, and the subscript s denotes the adsorbed species. If the adsorption rates, k_A and k_B, are much greater than the surface reaction rate, k_r, the process is reaction-limited, while the process is adsorption-limited in the opposite case. In the adsorption step, an A is chosen with probability $p = k_\mathrm{A}/(k_\mathrm{A} + k_\mathrm{B})$, or a B is chosen with probability $q = 1 - p$, and an attempt is made to adsorb the chosen molecule onto an empty lattice site. If adsorption occurs, then if $\mathrm{A}_s\mathrm{B}_s$ nearest-neighbor pairs are created, one such pair bonds to form an AB molecule which desorbs from the surface at a rate governed by k_r. We first consider this reaction in the irreversible limit where there is no desorption of unreacted A's or B's.

While this idealized model does not precisely correspond to an actual catalytic reaction, the model incorporates the basic steps of adsorption and surface reaction that are characteristic of most heterogeneous catalysis reactions.[4] Additionally, the simplicity of the monomer-monomer model renders it amenable to detailed theoretical analysis. Thus the investigation of the monomer-monomer model provides useful insights into more realistic catalytic processes. We first investigated the kinetics of the monomer-monomer model in a mean-field approximation,[3] in which the substrate is considered to be an N-site complete graph. We calculated the probability density that the surface has a given density difference by analyzing the Fokker-Planck equation[5] that arises from continuum limit of the underlying master equation for the probability distribution of the particle numbers, $P(n_A, n_B)$.

To write this equation, consider the reaction-limited process on an N-site complete graph in which each pair of sites is connected. In the reaction-limited case, when an AB pair reacts, it is replaced by AA, BB, AB, or BA, with respectively probabilities p^2, q^2, pq, and pq, respectively. Thus $P(n_A, n_B)$ evolves according to a stochastic process with the corresponding hopping probabilities

$$W(n_A, n_B \rightarrow n_A \pm 1, n_B \mp 1) = 2 \left\{ \begin{matrix} p^2 \\ q^2 \end{matrix} \left(\frac{n_A}{N}\right) \left(1 - \frac{n_A}{N}\right), \right. \tag{2}$$

with the rate for the null process $W(n_A, n_B \rightarrow n_A, n_B) = 1 - W(n_A, n_B \rightarrow n_A + 1, n_B - 1) - W(n_A, n_B \rightarrow n_A - 1, n_B + 1)$. From the master equation for $P(n_A, n_B)$, the corresponding Fokker-Planck equation for $P(x)$ in the continuum limit, when $p = q$, is

$$\frac{\partial P(x,t)}{\partial t} = \frac{1}{2N} \frac{\partial^2}{\partial x^2} \left(x(1-x)P(x,t)\right). \tag{3}$$

Here x denotes the concentration of the A's, $x = n_A/N$. The state-dependent diffusion coefficient $D(x) = x(1-x)$ reflects the fact that the probability of a reaction event is proportional to the concentration of AB pairs, $x(1-x)$. Thus the evolution of the surface concentration is analogous to diffusion in a medium that is increasingly "sticky" near the extremities of a finite absorbing interval, $x \rightarrow \pm 1$.

The full solution to the probability distribution is an eigenfunction expansion over Gegenbauer polynomials of order 3/2 (while in the the adsorption-limited case, the expansion is over integer-order Bessel functions).[3] From these solutions, we determined that discrete fluctuations ultimately cause the surface to become "saturated", *i. e.*, completely covered by only one species, so that the reaction ultimately stops. As the reaction proceeds, the probability that the catalyst has not saturated by time t decays exponentially in time, and the mean time until saturation, $\langle t \rangle$, is proportional to the number of reactive surface sites. Numerical simulations indicate that spatial dimension $d = 2$ is a critical value, above which the kinetics of the surface reaction is accurately described by the mean-field behavior. In one dimension, it is also obtained an exact solution in the reaction-controlled limit,[6] in which $\langle t \rangle$ is proportional to the square of the length of the system; this agrees with our earlier numerical studies.

When desorption of reactants is also allowed to occur in the monomer-monomer model, a bistability transition occurs as a function of the desorption and reaction rates.[1] In the master equation for the evolution of the concentration difference on the complete graph, desorption leads to both a diffusion and a drift term which drives the system toward equal concentration of A's and B's. This term counteracts the influence of the inhomogeneous diffusive term due to the surface reaction which tends to drive the system toward saturation. The corresponding Fokker-Planck equation is[7]

$$\frac{\partial P(x,t)}{\partial t} = \frac{p_D}{2} \frac{\partial}{\partial x} \left((2x-1)P(x,t)\right) + \frac{p_D}{4N} \frac{\partial^2}{\partial x^2} \left(P(x,t)\right) + \frac{(1-p_D)}{2} \frac{\partial^2}{\partial x^2} \left(x(1-x)P(x,t)\right). \tag{4}$$

The stationary solution to this equation is $P(y) \propto (1-(y/y_0)^2)^{-\alpha}$, where y_0 and α depend on the desorption rate, and $y = (2x-1)$ is the density difference between A's and B's. For large desorption rates, $P(y)$ is peaked at $y = 0$, (monostable), while for small rates, $P(y)$ is peaked at $y = \pm 1$ (bistable). Our prediction[7] of a transition between monostability and bistability has been observed in numerical simulations.[1]

(b) Kinetics of interacting catalysis models

We have investigated the kinetics of a monomer-monomer catalysis model in which there is a variable excluded volume interaction among particles of the same species (the A's for example).[8,9] For no interaction, this system reduces to the monomer-monomer model for which there is a first-order transition between A- and B-saturated phases as a function of the relative adsorption rates of the two species. For infinitely strong excluded-volume interactions, one can view the process as involving a large A particle whose diameter is larger than a lattice spacing (the "dollar"), so that two dollar particles cannot be nearest-neighbors, while the smaller ("dime") particles have a sufficiently small radius that they do not exhibit a nearest-neighbor exclusion. This geometric constraint on the dollar particles has far-reaching effects on the kinetics. Instead of a first-order transition between *dollar*- and *dime*-saturated phases, we find that there is now a second-order kinetic phase transition between a dime-saturated phase and a reactive steady state as a function of the relative deposition rates of the two species. Through numerical simulations, we concluded that this transition is in the universality class of Reggeon Field theory,[10] a class which encompasses nearly all single component reaction schemes with a unique absorbing state.

We also considered the general case where the magnitude of the excluded volume self-interaction is variable.[9] According to such an interaction, the probability that a particle of the interacting species can adsorb onto an empty site which is adjacent to a site that is already occupied by a member of the same species is reduced by an amount which depends on the interaction strength. As a function of this interaction, the first-order transition line for weak interaction terminates at a tricritical point where two second-order transitions meet. These transitions, which also appear to be in the Reggeon Field Theory universality class, separate the dollar-saturated, reactive, and dime-saturated phases. To determine the location of the transition and the corresponding scaling exponents, we have performed Monte Carlo simulations, series expansions, and cluster mean-field approximations.

From Monte Carlo simulations of the steady-state and cluster mean-field approximations, we find that the concentration of vacancies and dollars both vanish as the dime saturated phase is approached as $c_v \sim (p-p_c)^{\beta_v}$ and $c_D \sim (p-p_c)^{\beta_D}$, respectively. Here p denotes the relative input rate of dollars, and p_c is the value of this parameter at the phase transition. We thereby estimate the order-parameter exponents for vacancies and dollar particles to be $\beta_v \cong \beta_D = 0.28 \pm 0.05$. From subcritical series expansions and time-dependent Monte Carlo simulations, we also estimate the dynamical exponents for the survival probability and the order parameters to be $\delta = 0.16 \pm 0.01$ and $\eta_v \simeq \eta_D = 0.315 \pm 0.005$. Here the survival probability exponent δ is defined in terms of the survival probability, *i. e.*, the probability that the system has not yet entered a saturated state at time t. This quantity is expected to vary at the critical point as $P(t) \sim t^{-\delta}$. Similarly the exponents η_v and η_D are defined in terms of the time dependence of the number of vacancies or dollars, respectively, when the system is initially prepared with all dimes and a single vacancy. All of our exponent estimates are very close to those of the

Reggeon field theory in one dimension, suggesting that the two systems are in the same universality class.

3. Spatial Organization in Two-Species Annihilation

(a) Closest particle to an absorbing trap

We used a quasi-static approximation and extreme value statistics to obtain the typical value of the distance from an absorbing trap to the nearest particle, x_{min}, in a Brownian particle system.[11] The quasi-static approximation is an extremely powerful yet simple theoretical tool to analyze the spatial distributions of particles in reacting systems that are governed by diffusive dynamics.[12] In this method, the diffusion equation is replaced by the steady-state Laplace equation, and the boundary between the growing depletion layer due to trapping and the static large-distance profile is accounted for by a moving boundary condition. This leads to a greatly simplified form for the probability distribution function, but one which still retains all of the correct asymptotic behavior. We have then applied extreme value statistics[13] to this probability distribution to determine the location of the particle which is closest to the trap.

In one dimension, we find that the typical value of the distance from the trap to the closest particle grows with time as $x_{\mathrm{min}} \sim t^{1/4}$, and that the probability distribution for x_{min} is a Gaussian times a power law. This reproduces the results from the exact solution, which is much more complicated, technically.[14] In two dimensions, the exact solution is extremely involved and physically unilluminating,[15] while the quasi-static approximation provides a simple and physically-appealing method of solution. We find that $\rho \equiv x_{\mathrm{min}}/(\text{trap radius})$ grows as $\sqrt{\ln t}$, and the controlling factor in the distribution for ρ behaves as $\exp(-\text{const.} \times \rho^2 \ln \rho/\sqrt{\ln t})$. Finally, in three and higher dimensions, we find that $\rho \to$ const. as $t \to \infty$.

(b) Spatial distribution of reactants in two-species annihilation

In diffusion-limited two-species annihilation, $A + B \to 0$, particles organize into continuously growing single-species domains of linear dimension proportional to $t^{1/2}$, while the typical interparticle spacing grows as $t^{1/4}$, when $d < 4$.[16] This spatial organization is the mechanism responsible for the well-known slow kinetics, as reactions take place only on domain boundaries rather than homogeneously throughout the system. Rather unexpectedly, we found that a new length scale,[10] which grows as $t^{3/8}$, is needed to describe the "gaps" between domains in $d = 1$.[17] This interdomain gap size can be derived in terms of the known decay of the density $c(t)$. If one postulates that the gaps are of length $\ell_{AB} \sim t^{\zeta}$, then there will typically be a reaction in each gap in a time interval $\Delta t \propto \ell_{AB}^2$. This leads to a change in the concentration which is proportional to the inverse domain size, since one pair typically reacts per domain. As a result, $\frac{\Delta c}{\Delta t} \cong \frac{dc}{dt} \propto t^{-1/2-2\zeta}$. Thus in order that $c(t)$ is proportional to $t^{-1/4}$, it is necessary that $\zeta = 3/8$.

Concomitantly, we found that the average density profile within a domain is spatially inhomogeneous. Just as in the case of diffusing particle near a static trap, the density of one species is depleted as the boundary that defines the opposite species region in approached. Particles near the center of the domain are relatively closely spaced, with a typical spacing which grows as $t^{1/4}$, while particles are systematically more distantly spaced as the edge of a domain is approached. Within an adiabatic approximation, which is suitable for slowing growing domains, the shape of the density profile within a single domain has a sinusoidal shape, with the density vanishing linearly as the domain edge is approached. From this inhomogeneous profile, we found an unusual behavior for the time dependence of moments of the distance distribution between adjacent same species particles: $M_n \equiv \langle \ell_{AA}^n \rangle^{1/n}$. Namely,

$$M_n \sim \begin{cases} t^{1/4} & \text{for } n < 1, \\ t^{1/4} \ln t, & \text{for } n = 1, \\ t^{(3n-1)/8n} & \text{for } n > 1. \end{cases} \tag{5}$$

The logarithmic factor in the ratio between the average and typical spacing between neighboring like particles stems directly from the linear decrease in the density near the domain edge. This same mechanism also leads to a power-law diverging factor for the higher moments. As $n \rightarrow \infty$, the reduced moment is dominated by the sparsely populated region near the gap where the particles are separated by a distance which grows as $t^{3/8}$.

Even richer, and unanticipated, spatial organization effects occur when the diffusion coefficients of the two species, D_A and D_B, are unequal. For the extreme case where $D_B = 0$, the spatial correlations of the mobile and immobile species are dramatically different. The probability of finding a gap of length x between two nearest-neighbors of the same species decays exponentially in x for the mobile (A) species, and as $x^{-3/2}$ for the static species (B). The power law decay arises because the spatial correlation of the B's is altered only by the diffusion of A's into B domains. This kinetics can be described in terms of the first-passage probability for one-dimensional random walks.[18] Thus B-rich regions exhibit a very slow decay of correlations and this is ultimately responsible for the power-law behavior of the gap lengths. Similarly, the distribution of domain lengths L for the A's also decays exponentially in L, while the distribution decays as $L^{-1/2}$ for the B's.

(c) Dynamics at a reaction interface

In homogeneous two-species annihilation, domains form spontaneously from a random initial reactant distribution, and the kinetics is governed by the reaction rate at the interfaces between domains. This motivates our study of a system with a single well-defined reactive interface, so that the microscopic aspects of the reaction can be probed.[19] We therefore investigated steady-state geometrical properties of the reaction interface in two-species annihilation, $A + B \rightarrow 0$, when a flux j of A and B particles is injected at opposite extremities of a finite domain.

The kinetics of this process is described by the reaction-diffusion equations

$$\begin{aligned} \frac{\partial c_A}{\partial t} &= D_A \nabla^2 c_A - k c_A c_B, \\ \frac{\partial c_B}{\partial t} &= D_B \nabla^2 c_B - k c_A c_B, \end{aligned} \tag{6}$$

where D_i is the diffusion coefficient of species i and k is the reaction constant. For equal fluxes of A's and B's injected at opposite ends of the system, the resulting steady state is described by setting the time derivative to zero and postulating the boundary conditions

$$Dc'_A\big|_{x=L} = j, \quad Dc'_B\big|_{x=L} = 0 \qquad \text{and} \qquad Dc'_A\big|_{x=-L} = 0, \quad Dc'_B\big|_{x=-L} = -j. \tag{7}$$

These boundary conditions account for the constant and opposing particle fluxes and the confinement of reactants within $[-L, L]$.

Within this description, we determined the extent of the reaction zone and the spatial distribution of the two species both by an asymptotic solution to the above equations and from simple heuristic arguments. For example, for a large input flux, the concentration profile is nearly linear near the domain boundaries with the magnitude of the slope proportional to j/D. Since the reaction zone is the region for which the concentrations of both species are non-negligible, then the concentration in the reaction zone should be of the order jw/D, where w is the reaction zone width. Consequently, the number of annihilation events per unit time is of order kc_Ac_Bw, which is obtained by integrating the reaction term over the reaction zone. This quantity should equal the flux of particles j entering the domain. Therefore, balancing these two rates gives

$$kc_Ac_Bw \sim k\left(\frac{wj}{D}\right)^2 w \sim j, \tag{8}$$

or the reaction zone width $w \sim \left(\frac{D^2}{jk}\right)^{1/3}$. Similarly, the typical concentration in the reaction zone is

$$c_A, c_B \sim jw/D \sim \left(\frac{j^2}{Dk}\right)^{1/3}. \tag{9}$$

In the low flux limit, the concentration is almost independent of position and is proportional to $\sqrt{j}$. In the latter case, the local reaction rate reaches a maximum at the edges of the system rather than at the midpoint. When the two species approach at a finite velocity, there exists a critical velocity, above which the reactants essentially pass through each other. Results similar to those in one dimension were found in two- and three-dimensional radial geometries. Finally, we applied the quasistatic approximation to our steady-state solution to recover the known time dependence for the reaction zone width for the case of initially separated components with no external input. Our approaches are also applicable to other stoichiometries.

(d) Kinetics and spatial structure in a competing population biology model

We ascertained the role of spatial inhomogeneities[20] in the kinetics of competing biological species by investigating the kinetics in a prototypical population dynamics model[21] which involves two competing species. Each diffusing species undergoes logistic growth, *i. e.*, a growth rate proportional to the density, and a self-regulation, which is proportional to the density squared. In addition, there is a mutual competition where different species tend to annihilate when they meet. These processes are embodied by the reaction-diffusion equations,

$$\begin{aligned}\frac{\partial A}{\partial t} &= D_A\nabla^2 A - k_A A(1-A) - k_{AB}AB,\\ \frac{\partial B}{\partial t} &= D_B\nabla^2 A - k_B B(1-B) - k_{BA}AB.\end{aligned} \tag{10}$$

In the mean-field limit, where the species are assumed to be homogeneously distributed, there is coexistence for sufficiently weak competition, $k_{AB}, k_{BA} \ll 1$, while only one species survives when the competition is strong.

For diffusing species in one dimension, we have found a qualitatively different behavior. In the strong competition limit in one dimension, the populations organize into single-species domains whose characteristic length grows logarithmically in time. We have determined the detailed process by which a small domain of B's (for example) is overrun by the larger surrounding domains of A's. The smaller domain shrinks rather slowly until a critical size is reached, beyond which the smaller domain disappears almost immediately afterward. This qualitative behavior is also found to occur in simulation studies in two dimensions. In this case, domains grow as a power law in time, and the spatial pattern of the domains strongly resembles those observed in spinodal decomposition.[22]

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