Journal of Statistical Physics, Vol. 54, Nos. 5/6, 1989

Pattern Formation in Diffusion-Limited Reactions

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Received March 30, 1988; revision received April 15, 1988

The conditions for macroscopic segregation of A and B in a steady-state $A + B \rightarrow 0$ reaction are studied in infinite systems. Segregation occurs in one and two dimensions and is marginal for $d = 3$. We note the dependence of these results on the precise experimental conditions assumed in the theory. We also note the difference between these results and our earlier ones for finite systems where the critical dimension is $d = 2$.

KEY WORDS: Diffusion-limited reactions; macroscopic segregation; infinite systems; transients; critical dimension.

1. INTRODUCTION

Diffusion-limited reactions in low-dimensional systems exhibit behavior very different from classical kinetics.$^{(1-10)}$ The reaction $A + A \rightarrow$ *products*, of relevance in a number of physical systems,$^{(7,10,11)}$ exhibits nonclassical behavior for dimension $d \leqslant 2$ (including fractal dimensions).$^{(1-10)}$ The reaction $A + B \rightarrow$ *products*, of interest in chemical kinetics and in solid-state reactions, exhibits long-time anomalous kinetics and reactant segregation below dimension $d = 4$.$^{(1,3,5)}$ These results are for reactions of the "big-bang" type in which particle creation occurs only at the origin of time.

Recent simulations have shown that segregation of chemical species over macroscopic distances occurs for *steady-state* $A + B$ reactions.$^{(8)}$ The simulations start from zero density and thereafter A and B particles are

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0022-4715/89/0300-1429$06.00/0

added to the system at a steady rate. The two species are added at random locations and times but always in equal numbers so that there are no fluctuations in the global density difference $\rho_A^G - \rho_B^G = 0$. The simulations appear to indicate an upper critical dimension $d = 2$ for macroscopic segregation.(8,12,13)

Recently we analyzed a set of reaction-diffusion equations for the $A + B$ reaction in integer dimensions in the steady state and showed that in *finite* systems the critical dimensions is $d = 2$.(14) We found that macroscopic segregation occurs in one dimension and does not occur in three dimensions; in two dimensions the degree of segregation depends on the detailed parameter values. Since the simulations are also carried out in finite systems, the analytic results are in agreement with the numerical ones.(8)

Herein we analyze the same set of reaction-diffusion equations in *infinite* systems. The behavior we find is quite different from that of the finite systems in two important respects. First, we find that there is no steady state in one and two dimensions (a steady state in two dimensions is recovered if one introduces a finite correlation distance in the deposition of A–B pairs(15)). Second, the critical dimension for macroscopic segregation is $d = 3$ rather than $d = 2$ (unless the above finite correlation distance is introduced(15)). Thus, segregation occurs in one and in two dimensions, but in three dimensions the degree of segregation depends on detailed parameter values.

In Section 2 we present the reaction-diffusion equations and the constraints that we impose on the deposition of A and B molecules. In Section 3 we consider the equation for the difference in the local densities of the two species. We point out that the difference variable yields necessary but not sufficient conditions for segregation: the latter requires information about the total local density. In Section 4 we introduce a measure of segregation(14,16) that includes the latter information and discuss the presence or absence of segregation in one, two, and three dimensions. Further conclusions are presented in Section 5.

2. EQUATIONS AND CONSTRAINTS

Let $\rho_A(\mathbf{r}, t)$ and $\rho_B(\mathbf{r}, t)$ denote the local concentrations of species A and B at time t. Unlike species annihilate one another upon (suitably defined) contact,(17)

$$A + B \rightarrow 0 \tag{2.1}$$

Both species diffuse with coefficient σ and are created at random locations and times. Earlier we used the following set of reaction-diffusion equations to describe this process(14):

$$\dot{\rho}_A(\mathbf{r}, t) = \sigma\nabla^2\rho_A(\mathbf{r}, t) - F(\rho_A, \rho_B) + \eta_A(\mathbf{r}, t) \tag{2.2a}$$

$$\dot{\rho}_B(\mathbf{r}, t) = \sigma\nabla^2\rho_B(\mathbf{r}, t) - F(\rho_B, \rho_A) + \eta_B(\mathbf{r}, t) \tag{2.2b}$$

where η_A and η_B are random source terms chosen from an appropriate ensemble. The local reaction is described by the symmetric function $F(\rho_A, \rho_B) = F(\rho_B, \rho_A)$. The average number of A and B molecules is kept equal at all times, i.e.,

$$\int d\mathbf{r}\ \rho_A(\mathbf{r}, 0) = \int d\mathbf{r}\ \rho_B(\mathbf{r}, 0) \tag{2.3}$$

$$\int d\mathbf{r}\ \langle\eta_A(\mathbf{r}, t)\rangle = \int d\mathbf{r}\ \langle\eta_B(\mathbf{r}, t)\rangle \tag{2.4}$$

where the brackets denote an ensemble average. The constraint (2.4) is weaker than that used earlier, where the instantaneous total numbers of A's and B's were required to be equal in each member of the ensemble.

The symmetry of the problem allows us to simplify the rate equations by introducing the sum and difference variables

$$\gamma(\mathbf{r}, t) = \tfrac{1}{2}[\rho_A(\mathbf{r}, t) - \rho_B(\mathbf{r}, t)] \tag{2.5}$$

$$\rho(\mathbf{r}, t) = \tfrac{1}{2}[\rho_A(\mathbf{r}, t) + \rho_B(\mathbf{r}, t)] \tag{2.6}$$

and similarly for $\eta_\gamma(\mathbf{r}, t)$ and $\eta_\rho(\mathbf{r}, t)$. The difference variable then obeys the linear diffusion equation

$$\dot{\gamma}(\mathbf{r}, t) = \sigma\nabla^2\gamma(\mathbf{r}, t) + \eta_\gamma(\mathbf{r}, t) \tag{2.7}$$

from which the reaction term is absent. The sum variable satisfies the nonlinear reaction-diffusion equation

$$\dot{\rho}(\mathbf{r}, t) = \sigma\nabla^2\rho(\mathbf{r}, t) - F(\rho + \gamma, \rho - \gamma) + \eta_\rho(\mathbf{r}, t) \tag{2.8}$$

The constraints (2.3) and (2.4) imply that

$$\int d\mathbf{r}\ \langle\gamma(\mathbf{r}, t)\rangle = \int d\mathbf{r}\ \langle\eta_\gamma(\mathbf{r}, t)\rangle = 0 \tag{2.9}$$

at all times.

Our intent is to establish whether macroscopic segregation of the chemical species can arise in this system. Such segregation might arise as a consequence of the detailed interplay of the three counteracting mechanisms implicit in (2.2). Diffusion tends to mix the species. Reaction annihilates the thoroughly mixed regions, thus favoring spatially separated regions. The sources tend to create spatially nonuniform patches on a scale determined by the density and distribution of sources (this latter scale is in general much smaller than the macroscopic scales of interest for segregation). For sufficiently high dimension one expects diffusion to dominate and the spatial distribution of species to become uniform at long times. The interesting question is whether there exists a critical dimension below which macroscopic segregation occurs.

Elsewhere(14) we have examined the question of segregation for systems of finite size (L^d). We found that the random deposition of A's and B's at uncorrelated locations (but subject to the equal number constraint) leads to a steady state in all dimensions. A steady state manifesting *macroscopic segregation* is obtained in one dimension ($d=1$), marginal segregation is found for $d=2$, and no segregation occurs for $d \geqslant 3$. Note that these results require the equal-number constraint: if one allows instantaneous fluctuations in the relative numbers to be created, then each system of the ensemble will ultimately become all of one species. Furthermore, the results also change if one correlates the locations of the particles being created(15): in the limit of geminate creation there is no segregation in any dimension.(8)

Herein we reexamine this problem when $L \to \infty$. In this case we find that no steady state is established for $d \leqslant 2$, and hence the results for the infinite system cannot be obtained from those for the finite system by taking the limit $L \to \infty$.

3. DIFFERENCE VARIABLE

There is, of course, no unique measure of the degree of segregation of the chemical system. The measures we choose are determined in part by the requirement of analytic tractability. A measure of the spatial variations in the difference variable that we introduced earlier is the correlation function(14)

$$C_\gamma(\mathbf{r}-\mathbf{r}', t) \equiv l^{2d} \langle \gamma(\mathbf{r}, t)\, \gamma(\mathbf{r}', t) \rangle \tag{3.1}$$

where l^d is the volume of a unit cell in the corresponding d-dimensional discrete problem.(17) Note that this definition leads to the same units for C_γ in all dimensions. Our first step is to evaluate this correlation function.

To evaluate C_γ it is necessary to specify the first and second moments of the source term η_γ. These moments are a consequence of the spatially and temporally random deposition of A's and B's subject to the constraint (2.4) or equivalently (2.9). For the first moment, (2.9) and translational invariance clearly imply that

$$\langle \eta_\gamma(\mathbf{r}, t) \rangle = 0 \tag{3.2}$$

For the second moment in the infinite system one finds$^{(17)}$

$$\langle \eta_\gamma(\mathbf{r}, t)\, \eta_\gamma(\mathbf{r}', t') \rangle = (Rn/l^d)\, \delta(t-t')\, \delta(\mathbf{r}-\mathbf{r}') \tag{3.3}$$

R is the total deposition rate of A and B molecules per unit cell l^d, n is the average number of A and B molecules deposited in a unit cell in the time interval τ, and A and B molecules are deposited at spatially random and uncorrelated locations that change every τ units of time. The limit $\tau \to 0$ in relation to other time scales has been taken in writing (3.3).

The spatial Fourier transform of (2.7) according to the definition

$$\hat{f}(\mathbf{k}) = \frac{1}{(2\pi)^{d/2}} \int d\mathbf{r}\, f(\mathbf{r})\, e^{(i\mathbf{k}\cdot\mathbf{r})} \tag{3.4}$$

leads to an ordinary linear differential equation for $\hat{\gamma}(\mathbf{k}, t)$ whose solution is

$$\hat{\gamma}(\mathbf{k}, t) = \int_0^t dt'\, e^{-\sigma k^2(t-t')} \hat{\eta}_\gamma(\mathbf{k}, t') \tag{3.5}$$

The initial densities of A and B have been set to zero. To evaluate the correlation function (3.1), we first construct the correlation function $\hat{C}_\gamma(\mathbf{k}, \mathbf{k}', t)$ of the Fourier amplitudes,

$$\hat{C}_\gamma(\mathbf{k}, \mathbf{k}', t) = l^{2d} \langle \hat{\gamma}(\mathbf{k}, t)\, \hat{\gamma}(\mathbf{k}', t) \rangle \tag{3.6}$$

which is easily evaluated using the double Fourier transform of (3.3):

$$\langle \hat{\eta}_\gamma(\mathbf{k}, t)\, \hat{\eta}_\gamma(\mathbf{k}', t') \rangle = (Rn/l^d)\, \delta(t-t')\, \delta(\mathbf{k}+\mathbf{k}') \tag{3.7}$$

Inverse transformation of $\hat{C}_\gamma(\mathbf{k}, \mathbf{k}', t)$ according to

$$f(\mathbf{r}) = \frac{1}{(2\pi)^{d/2}} \int d\mathbf{k}\, \hat{f}(\mathbf{k}) \exp(-\mathbf{k}\cdot\mathbf{r}) \tag{3.8}$$

then yields

$$C_\gamma(\mathbf{r}-\mathbf{r}', t) = \frac{Rnl^d}{(2\pi)^d} \int d\mathbf{k} \int_0^t dt' \exp[-\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')] \exp(-2\sigma k^2 t') \tag{3.9}$$

The **k** integration can be carried out explicitly in each dimension, while the time integration can be done in closed form in $d=1$ and $d=3$ but not in $d=2$.

In one dimension one finds

$$C_{\gamma}^{(1)}(x-x',t)=\frac{Rnl}{(2\pi\sigma)^{1/2}}\left[t^{1/2}\exp\left(\frac{-(x-x')^2}{8\sigma t}\right)\right.$$
$$\left.-|x-x'|\left(\frac{\pi}{8\sigma}\right)^{1/2}\operatorname{erfc}\left(\frac{|x-x'|}{(8\sigma t)^{1/2}}\right)\right] \tag{3.10}$$

where erfc(z) is the complementary error function. For $(x-x')^2 \gg 8\sigma t$, Eq. (3.10) decays as

$$C_{\gamma}^{(1)}(x-x',t)\to\frac{Rnl}{(2\pi\sigma)^{1/2}}\frac{4\sigma t^{3/2}}{(x-x')^2}\exp\left(\frac{-(x-x')^2}{8\sigma t}\right) \tag{3.11}$$

The mean square value of the difference variable is independent of x and is given by

$$C_{\gamma}^{(1)}(t)\equiv C_{\gamma}^{(1)}(0,t)=\frac{Rnlt^{1/2}}{(2\pi\sigma)^{1/2}} \tag{3.12}$$

This boundless growth in time shows that, unlike in a finite system, a steady state is not achievable in one dimension. The behavior of (3.12) indicates a growth in the variability of the difference variable which is an indicator that segregation is possible, but in itself is insufficient t establish segregation.

In two dimensions the result of doing the **k** integration in (2.9) along with a suitable change of variables yields

$$C_{\gamma}^{(2)}(\mathbf{r}-\mathbf{r}',t)=\frac{Rnl^2}{4\pi\sigma}\int_{t^{-1/2}}^{\infty}dy\,\frac{\exp(-|\mathbf{r}-\mathbf{r}'|^2\,y^2/8\sigma)}{y} \tag{3.13}$$

For $(\mathbf{r}-\mathbf{r}')^2 \gg 8\sigma t$ one obtains

$$C_{\gamma}^{(2)}(\mathbf{r}-\mathbf{r}',t)\to\frac{Rnl^2 t}{\pi\,|\mathbf{r}-\mathbf{r}'|^2}\exp\left(\frac{-|\mathbf{r}-\mathbf{r}'|^2}{8\sigma t}\right) \tag{3.14}$$

and for the mean square of the difference variable we obtain

$$C_{\gamma}^{(2)}(t)=\frac{Rnl^2}{8\pi\sigma}\ln t \tag{3.15}$$

Again we find that no steady state is achieved, and that segregation is in principle possible due to the (slow) growth of the variability of the difference variable with time. Note that *any* correlation in the deposition of A and B particles *over a finite distance* inhibits the divergence obtained in (3.15) and causes $C_\gamma^{(2)}(t)$ to achieve a steady state.(15)

In three dimensions performing the two integrations in (3.9) leads to

$$C_\gamma^{(3)}(\mathbf{r}-\mathbf{r}', t)=\frac{Rnl^3}{8\pi\sigma}\frac{\operatorname{erfc}[|\mathbf{r}-\mathbf{r}'|/(8\sigma t)^{1/2}]}{|\mathbf{r}-\mathbf{r}'|} \tag{3.16}$$

The two limiting forms of this expression are

$$C_\gamma^{(3)}(\mathbf{r}-\mathbf{r}', t)\rightarrow\frac{Rnl^3}{8\pi\sigma}\frac{(8\sigma t/\pi)^{1/2}}{|\mathbf{r}-\mathbf{r}'|^2}\exp\left(\frac{-|\mathbf{r}-\mathbf{r}'|^2}{8\sigma t}\right) \tag{3.17}$$

for $|\mathbf{r}-\mathbf{r}'|\gg 8\sigma t$, and

$$C_\gamma^{(3)}(\mathbf{r}-\mathbf{r}', t)\rightarrow\frac{Rnl^3}{8\pi\sigma\,|\mathbf{r}-\mathbf{r}'|} \tag{3.18}$$

for $|\mathbf{r}-\mathbf{r}'|^2\ll 8\sigma t$. The divergence in (3.18) is specious: if we average $\mathbf{r}'$ over a unit cell surrounding $\mathbf{r}$, we obtain[5]

$$C_\gamma^{(3)}(t)\equiv\frac{1}{l^3}\int_{l^3}d\mathbf{r}'\,C_\gamma^{(3)}(\mathbf{r}-\mathbf{r}', t)=\frac{Rnl^2}{4\sigma} \tag{3.19}$$

Note that a steady state is thus achieved in three dimensions.

4. A MEASURE OF SEGREGATION

Earlier we introduced a measure $S(t)$ of the degree of macroscopic segregation as follows(14,16):

$$S(t)=\langle\gamma^2(\mathbf{r}, t)\rangle/\langle\rho^2(\mathbf{r}, t)\rangle \tag{4.1}$$

The maximum value of $S(t)$ is unity, and $S(t)\rightarrow 1$ when essentially only one of the chemical species is present at $\mathbf{r}$. On the other hand, $S(t)\rightarrow 0$ indicates that the relative excess of one species over the other is vanishingly small.

To find the value of $S(t)$ as $t\rightarrow\infty$ we again select for the reaction term in (2.2) the form(14)

$$F(\rho_A, \rho_B)=K\rho_A^X\rho_B^X \tag{4.2}$$

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[5] For convenience we integrate over a spherical volume. This choice affects the coefficient of (3.19) in a minor way.

where K is a rate constant. The order X is positive and is usually chosen to be unity, but we need not as yet make this choice. Equation (2.8) then is

$$\dot{\rho}(\mathbf{r}, t) = \sigma \nabla^2 \rho(\mathbf{r}, t) - K[\rho^2(\mathbf{r}, t) - \gamma^2(\mathbf{r}, t)]^X + \eta_\rho(\mathbf{r}, t) \tag{4.3}$$

To determine the mean square value $\langle \rho^2(\mathbf{r}, t) \rangle$ in (4.1), we find it expedient to make the replacement

$$\langle [\rho^2(\mathbf{r}, t) - \gamma^2(\mathbf{r}, t)]^X \rangle = [\langle \rho^2(\mathbf{r}, t) - \gamma^2(\mathbf{r}, t) \rangle]^X \tag{4.4}$$

When $X = 1$ this is of course a tautology; for other values of X this replacement introduces unknown errors that are unlikely to modify the asymptotic time dependence of this term. We furthermore note that

$$\langle \eta_\rho(\mathbf{r}, t) \rangle = R/l^d \tag{4.5}$$

The ensemble average of (4.3) with (4.4) and (4.5) then is

$$\langle \dot{\rho}(\mathbf{r}, t) \rangle = \sigma \nabla^2 \langle \rho(\mathbf{r}, t) \rangle - K[\langle \rho^2(\mathbf{r}, t) \rangle - \langle \gamma^2(\mathbf{r}, t) \rangle]^X + \frac{R}{l^d} \tag{4.6}$$

Consider the time dependences of the various terms in this equation. In all dimensions the source term is of $O(t^0)$. In one dimension $\langle \gamma^2 \rangle \sim O(t^{1/2})$. Suppose $\langle \rho(\mathbf{r}, t) \rangle \sim O(t^\alpha)$ with α to be determined. Then $\langle \dot{\rho}(\mathbf{r}, t) \rangle \sim O(t^{\alpha - 1})$ and $\langle \rho^2(\mathbf{r}, t) \rangle \sim O(t^{2\alpha})$, where again factorization is assumed not to change the asymptotic time dependence. Unless there are pathologically large spatial gradients (not observed in any simulations[(8)]), $\nabla^2 \langle \rho(\mathbf{r}, t) \rangle$ is at most of $O(t^\alpha)$. Balancing the powers of time that appear in (4.6) and noting that $\langle \rho^2(\mathbf{r}, t) \rangle \geqslant \langle \gamma^2(\mathbf{r}, t) \rangle$ leads, with $X > \frac{1}{2}$, to the unique choice[6]

$$\alpha = 1/4 \tag{4.7}$$

resulting in

$$\langle \rho(\mathbf{r}, t) \rangle \sim t^{1/4} \tag{4.8}$$

and the even stronger condition

$$S(t) \xrightarrow[t \to \infty]{} 1 \tag{4.9}$$

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[6] If $\nabla^2 \langle \rho \rangle = O(t^\alpha)$, then the above analysis requires $X > \frac{1}{2}$. If $\nabla^2 \langle \rho \rangle \leqslant O(t^\beta)$ with $\beta \leqslant \alpha - \frac{1}{4}$, then X is unrestricted (albeit positive). If $\nabla^2 \langle \rho \rangle = O(t^\alpha)$ and $X \leqslant \frac{1}{2}$, the analysis is more complicated and the segregation index may not approach unity. This case seems not to have been observed in practice and we do not pursue it further here.

This indicates that there is asymptotic segregation in one dimension. Equations (3.10) and (3.11) show that the size of a region occupied by a large excess of one chemical species is diffusion dominated, i.e., of $O[(8\sigma t)^{1/2}]$. The other possible distance scale in the problem, the distance $nl/R\tau$ defined by the random distribution of source terms η_A and η_B, only affects the overall number of molecules present at any time, i.e., it appears only in the overall coefficient in (3.10).

In two dimensions with (3.15), the leading terms in (4.6) with $X > \frac{1}{2}$ can again only be balanced if

$$\langle \rho(\mathbf{r}, t) \rangle \sim \ln^{1/2} t \tag{4.10}$$

and one obtains asymptotic segregation, i.e.,

$$S(t) \xrightarrow[t \to \infty]{} 1 \tag{4.11}$$

The size of the segregated regions is again diffusion dominated. We contrast this two-dimensional result with the one obtained in a finite system: there we found a segregation index whose value depends on the detailed parameters of the problem and in particular on the relative rates of diffusion and reaction. We also mention that the result (4.11) is a consequence of our assumption of spatially random deposition of A's and B's with no correlation in the A–B distances. If one introduces a correlation in the A–B distances which remains finite as the volume of the system goes to infinity, then (4.11) does not hold and one recovers parameter-dependent results of the sort obtained for finite systems.(15)

In three dimensions the mean square of the difference variable does not grow in time [cf. (3.18)] and the reaction-diffusion equations can be balanced with $\langle \rho(\mathbf{r}, t) \rangle \to$ constant in time as $t \to \infty$. A steady state is thus achieved. In this limit one expects that the spatial gradient term at any one position vanishes as $t \to \infty$ (i.e., $\sigma \nabla^2 \langle \rho \rangle \sim \langle \rho \rangle / t$), so that

$$\lim_{t \to \infty} \langle \rho^2(\mathbf{r}, t) \rangle \approx \lim_{t \to \infty} \langle \gamma^2(\mathbf{r}, t) \rangle + (R/l^3 K)^{1/X} \tag{4.12}$$

and consequently

$$S(t) \xrightarrow[t \to \infty]{} \left[1 + R^{(1-X)/X} \frac{4\sigma l^{4-3/X}}{n K^{1/X}} \right]^{-1} \tag{4.13}$$

Whether or not one observes segregation in three dimensions therefore depends on the detailed parameter values. In the simplest case ($n = 1$, $X = 1$) the segregation index is determined by the ratio $\sigma l/K$, i.e., by the relative rates of diffusion and reaction.

5. CONCLUSION

We have explored the effect of dimensionality in infinite systems on the spontaneous segregation of chemical species A and B that react ($A + B \to 0$), diffuse, and are replenished at random temporal and spatial points. When the deposition of A's and B's is totally uncorrelated, then we find that in one and two dimensions there is no steady state and spontaneous segregation occurs over regions whose size is diffusion limited. In three dimensions a steady state is established and it may be possible to observe marginal segregation if the rate of reaction is sufficiently high compared with the rate of diffusion. These results should be contrasted with those for a finite system, where the "marginal" dimension is 2, with definite segregation in one dimension but none in three dimensions.

We make a final comment about the subtle interplay of diffusion, reaction, and deposition in the results obtained here and in our study of finite systems. It would be tempting to conclude that diffusion inhibits segregation by promoting mixing, and that one therefore obtains macroscopic segregation in lower dimensions but not in higher dimensions. This is not completely correct; diffusion (but not too much of it) is actually necessary to obtain macroscopic segregation: If there were none, then the spatial A–B correlations would simply be those of the source terms, in our case $C_\gamma(\mathbf{r} - \mathbf{r}', t) \propto \delta(\mathbf{r} - \mathbf{r}')$ as seen in (3.9) if $\sigma \to 0$. One would thus not observe segregation in any dimension in the absence of diffusion. It is interesting to speculate what (perhaps fractal) dimension actually leads to maximal segregation.

ACKNOWLEDGMENTS

We thank C. R. Doering for a number of interesting discussions. This research is supported in part by NSF grant DMR 83-03919 (R.K.), NSF grant DMR 86-19650-A1 (K.L.), NIH Special Fellowship PHS NS 08256-01 (R.K.), and DOE grant DE-FG03-86ER13606 (B.J.W. and K.L.). One of us (R.K.) acknowledges the hospitality and support of the Institute for Nonlinear Science and the Department of Chemistry at UCSD.

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