Reactive Particles in Random Flows

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We study the dynamics of chemically or biologically active particles advected by open flows of chaotic time dependence, which can be modeled by a random time dependence of the parameters on a stroboscopic map. We develop a general theory for reactions in such random flows, and derive the reaction equation for this case. We show that there is a singular enhancement of the reaction in random flows, and this enhancement is increased as compared to the nonrandom case. We verify our theory in a model flow generated by four point vortices moving chaotically.

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The study of chemical and biological activity of particles advected by fluid flows is of fundamental importance to the understanding of the dynamics of important environmental systems, such as the population dynamics of plankton in the sea [1], the depletion of the ozone layer [2], and the dynamics of pollutants in the atmosphere [3], to name only a few. In many situations, the flow in which activity occurs can be considered to be *open*, meaning that there is a net current flowing through the region of observation. An example is the flow in the wake of an obstacle. In most cases, open flows display *chaotic scattering* [4,5], where the dynamics of advected particles is dominated by a *chaotic saddle*, which is an infinite fractal set of nonescaping unstable orbits. For some simple flows, it has been found [6,7] that the presence of the chaotic saddle has important consequences for the reaction dynamics.

These previous results were obtained by assuming that the time dependence of the flow is periodic. In nature, however, very few periodic flows are found. Although it seems difficult to find a theory of reactions for completely turbulent flows, there is a class of flows to which a theory can be established, which is in between the regular and the fully turbulent cases. Many flows display well-defined *coherent structures*, that is, well-defined persisting space-time patterns, such as the large persistent vortices found in many real flows [8]. The theory we develop in this Letter applies to flows in which the coherent structures are the dominating feature. We assume that the number of these coherent structures is fixed, but their position and shape vary *chaotically*. It makes then sense to model the advection dynamics on snapshots taken with some period, as a map with randomly varying parameters. The advection dynamics is thus described by a *random map* [9], in which the random parameters are assumed to be taken from a stationary ensemble. From now on, we use the term *random flows* to designate such flows. In this Letter, we develop a general theory for the dynamics of active processes in random flows. We obtain the reaction equation, and show that there is a singular enhancement of the production. Hence, we get the important and far-reaching result that the enhancement of productivity is *not* restricted to periodic flows, but in fact holds for a far more general class of flows, which includes many important real-life systems, as already mentioned.

We consider, as an example of activity, an autocatalytic reaction $A + B \longrightarrow 2B$, where the stable phase B "invades" the unstable phase A. We consider a kinetic-type model, in which B is assumed to be made of particles advected by the flow, and A is assumed to be a uniform background. This means that we do not have to deal with reaction-diffusion equations directly. We consider a two-dimensional incompressible flow [10]. We assume that the border between the phases A and B is sharp, which means that a well-defined reaction front exists. This is true when diffusivity is small enough, and the Péclet number is large. The front moves from B towards A with constant velocity v_R. In an open flow, B particles are carried away due to outflow, and a nontrivial steady state configuration is reached. If the advection dynamics is chaotic, the B particles converge to the unstable manifold of the chaotic saddle, as the system evolves in time. This unstable manifold is made up of a Cantor set of one-dimensional curves [5]. In a random flow, both the chaotic saddle and its unstable manifold are fractal objects of well-defined dimensions which do not repeat their shapes in time [9]. The advection carries an initial distribution of B particles into thin stripes of typical width $\epsilon(t)$ surrounding the unstable manifold. If there were no reactions, escape would cause $\epsilon(t)$ to decrease to 0 as $t \longrightarrow \infty$, and there would be practically no B particles left in the system after a long enough time. However, the loss due to escape is counteracted by the production of new B particles in the autocatalytic reaction. The advection makes a B stripe of width ϵ contract at a rate given by $-\lambda\epsilon$, where λ is the local contraction rate. Hence, the total time

evolution of ϵ, with the reaction taken into account, is given by $\dot{\epsilon} = -\lambda\epsilon + c v_R$ [6], which can be conveniently rewritten as $d\ln\epsilon/dt = -\lambda + v_R c/\epsilon$. Here c is a geometric factor of order 1 which depends on the particular shape of the unstable manifold and on the overlapping of neighboring stripes. In random flows, λ and c are not constants: they are random variables. By averaging over the stationary distribution of the randomly varying parameters of the flow, we get [11]

$$\frac{d\overline{\ln\epsilon}}{dt} = -\overline{\lambda} + \overline{\left(\frac{c}{\epsilon}\right)} v_R. \tag{1}$$

There is no average on v_R, since it is an intrinsic property of the reaction.

In order to derive the reaction equation, we turn our attention to the area $B(t)$ occupied by the B particles at time t. A fundamental result in the theory of random maps is that there is a well-defined fractal dimension d *independent of time* [9]. For a particular realization of the flow parameters (that is, for one element of the ensemble of dynamical systems), we have the well-known relation $B = \mathcal{H}\epsilon^{2-d}$ for the area covering a fractal object with dimension d at a resolution ϵ [12]. $\mathcal{H}$ is another geometric factor. From this expression, we obtain

$$\frac{d}{dt}\ln B = (2-d)\frac{d}{dt}\ln\epsilon + \frac{d}{dt}\ln\mathcal{H}. \tag{2}$$

The above formula is valid for a particular element of the ensemble. As the fluctuations of the flow parameters and hence of $\mathcal{H}$ are taken from a stationary ensemble, the average of $\mathcal{H}$ is time independent. From (1) and (2) we find [11]

$$\frac{d}{dt}\overline{\ln B} = -\overline{\lambda}(2-d) + (2-d) v_R \overline{\left(\frac{c}{\epsilon}\right)}. \tag{3}$$

Since the generalization of the Kantz-Grassberger relation [13] is also valid for random maps [14], the dimension d of the chaotic saddle's unstable manifold can be expressed as $d = 2 - \kappa/\overline{\lambda}$. We therefore identify the first term on the right-hand side of (3) as $-\kappa$, where κ is the *escape rate* from the random chaotic saddle.

To get an equation for $\overline{B}$ in a closed form from Eq. (3), we need to derive a relation between $\overline{B}$ and $\overline{\epsilon}$. We do this by assuming weak relative fluctuations. We now write $\epsilon = \overline{\epsilon} + \delta\epsilon$, where $\delta\epsilon$ satisfies $\overline{\delta\epsilon} = 0$. We write a similar expression for the geometric factor: $c = \overline{c} + \delta c$, with $\overline{\delta c} = 0$. In view of the aforementioned weak relative fluctuations, the averaged terms in Eq. (1) are well approximated by

$$\overline{\ln\epsilon} = \ln\overline{\epsilon} - \frac{1}{2}\rho, \qquad \overline{\left(\frac{c}{\epsilon}\right)} = \left(\frac{\overline{c}}{\overline{\epsilon}}\right)(1+\rho), \tag{4}$$

where $\rho = \overline{(\delta\epsilon/\epsilon)^2} \ll 1$ is the strength of the randomness in the flow dynamics. Notice that the variance of c does not appear in these expressions, and we assumed that the cross correlation $\overline{\delta c\,\delta\epsilon}$ is zero [15].

Substituting Eq. (4) into Eq. (1), we get the equation for the time evolution of $\overline{\epsilon}$:

$$\frac{d\overline{\epsilon}}{dt} - \frac{1}{2}\overline{\epsilon}\frac{d\rho}{dt} = -\overline{\lambda}\,\overline{\epsilon} + v_R\overline{c}(1+\rho). \tag{5}$$

The effect of randomness on the average width is a multiplicative correction on the production term, which can be thought of as an increase in the effective reaction velocity v_R [16]. In other words, *randomness enhances productivity,* as compared to the nonrandom case.

Next, we average the expression $B = \mathcal{H}\epsilon^{2-d}$, and use the expansions $\mathcal{H} = \overline{\mathcal{H}} + \delta\mathcal{H}$ ($\overline{\delta\mathcal{H}} = 0$) and $\epsilon = \overline{\epsilon} + \delta\epsilon$. The result is

$$\overline{B} = \overline{\mathcal{H}}\,\overline{\epsilon}^{2-d}\left[1 + \frac{(2-d)(1-d)}{2}\rho\right], \tag{6}$$

where, for simplicity, we also assumed that the cross-correlation term $\overline{\delta\epsilon\,\delta\mathcal{H}}$ vanishes. Within this approximation $\overline{\ln B} = \ln\overline{B} - (2-d)^2\rho/2 - 1/2\overline{\delta\mathcal{H}^2}/\overline{\mathcal{H}}^2$ [17]. Substituting this result and (6) in Eq. (3), we get the final form for the time evolution equation for the area B of reacting particles:

$$\frac{d\overline{B}}{dt} - \frac{(2-d)^2}{2}\overline{B}\frac{d\rho}{dt} = -\kappa\overline{B} + v_R g\overline{B}^{-\beta}\left(1 + \frac{3-d}{2}\rho\right) \tag{7}$$

up to first order in ρ where $g = (2-d)\overline{c}\,\overline{\mathcal{H}}^{1/(2-d)}$ is an average overall geometric factor [16,18], and the power β is given by

$$\beta = \frac{d-1}{2-d} \geq 0. \tag{8}$$

Since $1 < d < 2$, the production term in Eq. (7) has a singular dependence on $\overline{B}$, and it diverges for $\overline{B} \to 0$. The relation (8) between β and d is formally the same as in the time-periodic case [6], but here d is the dimension of the unstable manifold *of the random flow.* Thus, we have obtained the crucial result that the singular enhancement of production is present *even in random flows*, and hence cannot be dismissed as an effect of artificial time-periodicity assumptions. It follows from (7) that the steady state B-content B^* scales as $B^* \sim v_R^{2-d}$ with the reaction velocity.

We did not mention diffusion in the above, because we have considered the case of flows with a very large Péclet number, which allows us to neglect diffusion effects. For finite diffusivity, there is a lower cutoff scale $\bar{\epsilon}$ below which the distribution is two dimensional, and therefore, density gradients cannot appear on arbitrarily fine scales [19].

Comparing Eq. (7) with the corresponding equation for time-periodic flows [6], the random nature of the flow is manifested in the fact that $d > d_0$, where d_0 is the

dimension of the nonrandom flow (in the limit $\rho \to 0$) from which $\beta > \beta_0$ follows [20]. This confirms the fact that productivity is enhanced by randomness.

In order to test our theory, we numerically simulate the autocatalytic reaction generated by four point vortices [20]. This is one of the simplest open 2D flows that has a chaotic time dependence [20]. Although this system is deterministic, its stroboscopic map can be modeled by a random map, due to the irregularity of the dynamics. The vortices of the model represent the coherent structures mentioned above. This chaotic vortex system can also be considered as a local model of 2D turbulence over a finite period of time [21]. The Hamiltonian equations of motion for an advected particle are written in terms of the stream function $\Psi = \Psi(x, y, t)$, given in our case by $\Psi = -\frac{1}{\pi}\sum_{i=1}^{4} \Gamma_i \ln r_i(t)$. Here Γ_i is the strength of the ith vortex, and r_i is the distance of the advected particle from the ith vortex. The positions (x_i, y_i) of the vortices evolve in time according to the equations $\Gamma_i \dot{x}_i = (\partial H/\partial y_i)$; $\Gamma_i \dot{y}_i = -(\partial H/\partial x_i)$, where the Hamiltonian H is $H = -\frac{1}{\pi}\sum_{i<j}\Gamma_i\Gamma_j \ln r_{ij}$, and r_{ij} is the distance between vortices i and j.

We numerically implement the autocatalytic reaction in the following way. We choose a rectangular region R such that initially all the four vortices are in R. In order to make the numerics feasible, we define a spatial grid in the flow. Our numerical procedure is a discretized (in both space and time) approximation to the continuous reaction used in our theory. We partition R into $n_x n_y$ rectangular grid cells corresponding to the division of the x and y axes into n_x and n_y segments, respectively. As the system evolves in time, the average position of the vortices undergoes an overall drift [20]. For this reason, we define R in the *comoving* coordinates x_c and y_c, defined by $x_c = x - \frac{1}{4}\sum_{i=1}^{4} x_i$ and $y_c = y - \frac{1}{4}\sum_{i=1}^{4} y_i$. In these new coordinates, the motion of the vortices is confined to a finite volume of the x-y plane if the total vorticity is zero, i.e., if $\sum_{i=1}^{4}\Gamma_i = 0$. Region R is chosen so that if a particle leaves R, it will necessarily escape towards infinity. A given particle in R is considered to be located in the center of the corresponding grid cell of R. Each particle evolves in time for a certain *reaction time lag* τ, and ends up within another cell. In this way, a cell-to-cell mapping is defined (in the comoving coordinates). If the mapping takes a particle outside R, the particle is discarded. After advection, the particles undergo the autocatalytic reaction: if a given cell contains a B particle before the reaction, all eight surrounding cells will also contain B particles after the reaction. If a cell already has a particle, it remains unaltered. The complete dynamics of the system is thus composed of an advection phase followed by a reaction phase, and this is repeated indefinitely. Denoting the grid size by σ, this discrete approximation goes over into the continuous theory in the limit $\tau \to 0$ and $\sigma \to 0$, with the ratio $\sigma/\tau = v_R$ remaining finite. For fixed τ we therefore expect $B^* \sim v_R^{2-d} \sim \sigma^{2-d}$. Therefore, all the results of our discretized numerical model can be rewritten in terms of variables of the continuous system.

We use vortex strengths $\Gamma_1 = \Gamma_2 = \Gamma_3 = 1$, $\Gamma_4 = -3$, and choose $R = [-1.3, 1.3] \times [-0.2, 1.4]$. The initial vortex positions are $x_1 = x_2 = x_3 = x_4 = 0$, and $y_1 = 0.1$, $y_2 = 0.6$, $y_3 = 1$, $y_4 = 0.4$. The reaction time is kept constant, at the value of $\tau = 2$. We start with a uniform initial distribution of B particles on the grid. After a few reaction steps, the system reaches a state of dynamical equilibrium, which is independent of the initial distribution. Because of the flow's aperiodicity, the number of B particles does not stay constant in time, but fluctuates in a randomlike fashion; see the right inset in Fig. 1. Our theory predicts that after the transient period is over, the B particles should be distributed in a filamentary fashion, shadowing the unstable manifold of the advection dynamics. In other words, the B distribution is a fractal for scales larger than the width ϵ. In Figs. 1 and 2, the B distribution is shown at different times when the transient phase is over. It is clear that the B distribution is very intricate and filamentary. In order to test our theory quantitatively, we compute the equilibrium distribution for several values of the grid size σ, keeping all the other parameters constant. Let R_0 be a subregion of the rectangle R. If $N(\sigma)$ is the number of B particles in the steady state distribution within R_0 [22], it should scale as $N(\sigma) \sim \sigma^{-d_B}$ [12], where d_B is the fractal dimension of the B particle distribution [note that $N(\sigma)\sigma^2 = B^*$]. The insets of Figs. 1 and 2 show the numerical result for $N(\sigma)$ in R_0 at different times. It is seen that $N(\sigma)$ does indeed follow a power law, from which we find $d_B = 1.96 \pm 0.01$ and $d_B = 1.94 \pm 0.01$, respectively. Comparing this with the value $d = 1.95$ of the unstable manifold dimension of

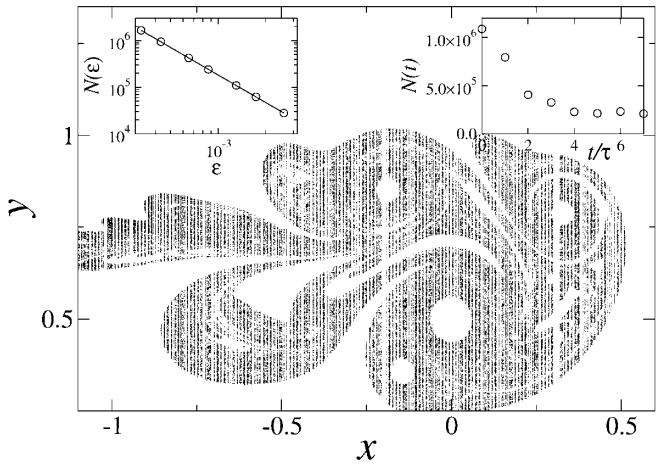

FIG. 1. Distribution of B particles at time $t = 14$, right after the 7th reaction step. The upper left inset illustrates the scaling of $N(\sigma)$ for the region given by $R_0 = [-1.5, -0.26] \times [0.2, 1.2]$. The result is a well-defined power law, with a fractal dimension of 1.96. The upper right inset shows the time evolution of the number of B particles towards the steady state, where the time t is measured in units of the reaction time τ.

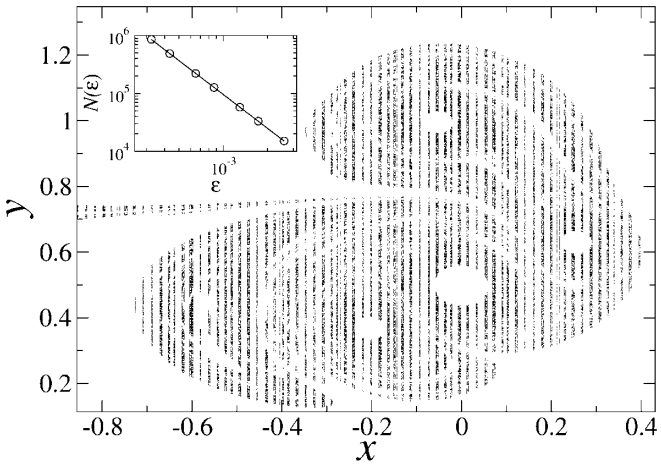

FIG. 2. Distribution of B particles at time $t = 16$, right after the 8th reaction steps in the counting region R_0. The inset shows the scaling of $N(\sigma)$ for $R_0 = [-0.8, -0.28] \times [0.1, 0.8]$. The measured fractal dimension is 1.94, which is within statistical errors equal to the result of Fig. 1.

the advection dynamics, calculated in Ref. [20], we see that they agree within numerical uncertainties, as predicted by our theory.

We have tested the theory in other systems, including the alternating sinusoidal shear flow [23] and the baker map. In all cases, the scaling of $N(\sigma)$ obtained from the simulations were in accordance with the theory.

In conclusion, we have studied the autocatalytic reaction dynamics in randomly varying 2D open flows. Other reactions, such as, e.g., $A + B \to C$ [6], or a competitive population dynamics [24], are described by different rate equations, but they all contain singular terms of power $\beta_0 = (d_0 - 1)/(2 - d_0)$. Because this is a result due solely to the presence of a chaotic saddle in the advective dynamics, our theory implies that this structure for the rate equation remains unchanged when the flow is random. This implies that the singular enhancement of the reaction (dynamical catalysis) must be a feature of several flows found in Nature.

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[15] Otherwise $\overline{c/\epsilon} = \overline{c}/\overline{\epsilon}(1 + \rho - \rho')$ where $\rho' = \overline{\delta\epsilon\delta c}/(\overline{\epsilon}\,\overline{c})$, and $|\rho'| \ll 1$.

[16] Since ρ is a relative variance, it is natural to assume that it goes to a constant, $d\rho/dt = 0$, much before the reaction reaches a steady state.

[17] If cross correlation is present, $\overline{B} = \overline{\mathcal{H}}\,\overline{\epsilon}^{2-d}[1 + (2 - d) \times (1 - d)\rho/2 + (2 - d)\rho'']$ where $\rho'' = \overline{\delta\epsilon\delta\mathcal{H}}/(\overline{\epsilon}\,\overline{\mathcal{H}})]$, and $|\rho''| \ll 1$. At the same time $\overline{\ln B} = \ln\overline{B} - \overline{\delta\mathcal{H}^2}/(2\overline{\mathcal{H}^2}) - (2 - d)\rho'' - (2 - d)^2\rho/2$.

[18] In the presence of cross correlations an additional term $-(2 - d)\overline{B}d\rho''/dt$ appears on the right-hand side, and $\rho'' - \rho'$ is to be added to the parentheses on the right-hand side.

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