PHYSICAL REVIEW E VOLUME 56, NUMBER 2 AUGUST 1997

Behavior of the reaction front for $A+B\rightarrow C$ diffusion-reaction systems: An analytic approach

Miguel A. Rodriguez
Instituto de Física de Cantabria, CSIC–Universidad de Cantabria, Santander, Spain

Horacio S. Wio*
International Centre for Theoretical Physics, P.O. Box 586, 34100-Trieste, Italy

(Received 11 December 1996)

The time behavior of the reaction front between initially separated reactants of the diffusion-limited reaction $A+B\rightarrow C$ in a one-dimensional system is studied by means of a stochastic approach. The asymptotic results of this scheme give a rather good agreement with simulations regarding the time power indexes of scaling for the height and width of the distribution of C particles. [S1063-651X(97)07308-X]

PACS number(s): 05.20.Dd

I. INTRODUCTION

The recent interest raised by the kinetics of the recombination process $A+B\rightarrow C$ (C inert and immobile) is due to the fact that segregation of like particles occurs, leading to anomalous reaction rate laws [1,2]. There is a remarkable sensitivity of segregation to changes on initial conditions, presence of sources, disorder, external forces, etc. Most of the recent literature is devoted to the analysis of these phenomena under the assumption that some kind of rate equations are valid [1–10].

A particular aspect that has been studied through experiments, theory, and simulations is the situation in which the reactants are initially separated in space [11–22]. In such a case the system develops a reaction front at the interface separating the reactants, which is marked by the concentration of the C particles. As was discussed, the behavior of such a reaction front should be of relevance for a variety of biological, chemical, and physical systems [23].

Usually, the theoretical analysis of the behavior of this front is done in terms of diffusion-reaction equations where the reaction terms are modeled according to chemical kinetics [11–19]. Recently, renormalization group techniques have also been used, starting from a master equation describing the process and transforming it into a second quantized version [22]. In this paper we will use a different, more accesible, framework, whose starting point is a stochastic equation for the density of a species of particles in which the other species acts as an absorber with a given absorption shape function. It is a generalization of the Galanin methods used in nuclear reactor theory for modeling neutron absorption [24]. This equation can be averaged exactly. Then, by using a diagrammatic technique it is possible to select and take into account the leading contributions for short and long times. The result is an equation for the density of particles in which the reaction term is given by a convolution in space and time of a kernel with the density. The method has been used initially for analyzing the evolution of the $A+B\rightarrow B$ homogeneous reaction [25] and afterwards extended to the case $A+B\rightarrow C$ [26]. As was pointed out there the method has several advantages. On one hand, it is able to describe with good precision the system evolution for any time, while usually only asymptotic results are obtained. Within this approach we have the possibility of varying the reaction length by controlling the probability of reaction. Then, it is possible to study the cases of perfect and imperfect reactions. On the other hand, the resulting equation is the continuous limit of a master equation directly related to simulations. For this reason we have always obtained a good agreement between simulations and solutions of such an equation [25,26].

In Sec. II we will briefly review the formalism we are going to use. After that, we will apply the scheme to the reaction front problem described above and we will show how the exponents indicating the time behavior of the front width and height that result from the present analysis are in rather good agreement with those known from simulations [13,18,20]

II. FORMULATION OF THE PROBLEM AND AVERAGED EVOLUTION EQUATIONS

Here we will make a brief review of our scheme. The complete details of the method can be found in Refs. [25,26].

We consider two kinds of particles, A and B, with independent motion and with the possibility of annihilation when they are in contact. We assume that the motion of an individual particle is a Markovian process with conditional probability $G_A(r,t|r',t')$ for the A particles and $G_B(r,t|r',t')$ for the B ones. These probabilities fulfill the evolution equations:

$$\dot{G}_A = L_A G_A , \tag{1a}$$

$$\dot{G}_B = L_B G_B , \tag{1b}$$

with $G_A(r,t_0|r',t_0) = G_B(r,t_0|r',t_0) = \delta(r-r')$ as initial conditions, and L_A, L_B being linear operators. The Markovian character, which is essential in this formulation, makes it possible to factorize the one particle joint probability in the usual way.

---

*Permanent address: Centro Atómico Bariloche (CNEA) and Instituto Balseiro (UNC), 8400 San Carlos de Bariloche, Argentina.

Each particle moves independently, but its survival depends on the evolution of the other particles. We identify each particle with a numerical index. Since the number of particles is not conserved, the set of indices corresponding to the existing particles varies with time, $\{i(t)\}$. The density of particles of one kind at time t is given by

$$n_{A,B}(r,t)=\left\langle \sum_{\{i_{A,B}(t)\}} \delta(r-r_{i_{A,B}}(t)) \right\rangle, \tag{2}$$

where the brackets $\langle\ \rangle$ indicate an average over the motion process and over all possible annihilation processes. Also, the density functions defined by

$$n_{A,B}(r,t'|t)=\left\langle \sum_{\{i_{A,B}(t)\}} \delta(r-r_{i_{A,B}}(t')) \right\rangle. \tag{3}$$

correspond to the density of particles at time t' that survive until time t. Both densities are related through

$$n_{A,B}(r,t)=\int G_{A,B}(r,t|r',t')n_{A,B}(r',t'|t)dr'. \tag{4}$$

To invert these equations in order to obtain $n_{A,B}(r',t'|t)$ as a functional of $n_{A,B}(r,t)$ is quite difficult, and it is possible only when the evolution is essentially diffusive or the density is rather uniform.

In order to consider the reaction process, we assume that when two particles of different species are close enough to each other they can annihilate. The rate of probability of such a process when the relative position between the particles is r, is given by a function $\gamma_\epsilon H_\epsilon(r)$, where ϵ is a mean interaction length, H_ϵ is a normalized function that tends to $\delta(r)$ when $\epsilon\rightarrow 0$, and γ_ϵ is a rate of probability per unit volume of interaction of one particle. The effective interaction time is given by $\epsilon^{-d}\gamma_\epsilon^{-1}$, and the reaction will be either perfect (fast) or imperfect (slow) if this time is shorter or longer than the collision time [17].

To formulate the evolution equations, we define $n_{A,B}(r,t|\{B,A\})$ as the density of the A or B particles conditioned to the occurrence of a process $\{r_{i_B}(t)\}$ or $\{r_{i_A}(t)\}$ in the evolution of the A and B particles. Hence, the evolution of these densities is governed by

$$\begin{aligned}\dot{n}_{A,B}(r,t\{B,A\}) &= L_{A,B}n_{A,B}(r,t|\{B,A\})\\ &\quad -\gamma_\epsilon \sum_{\{i_{B,A}(t)\}} H_\epsilon(r-r_{i_{B,A}}(t))\\ &\quad \times n_{A,B}(r,t|\{B,A\}).\end{aligned} \tag{5}$$

This equation generalizes the Galanin approach for extended absorbers [24]. In the limit $\epsilon\rightarrow 0$ we recover the original form of point absorbers. This limit is strictly correct in dimension one, but in higher dimensions we need to take into account the size of the particle. The equations are coupled in the sense that the target particles act like an external absorber. The rate of absorption for each particle is then the same only in average. In other words, the annihilation of one particle does not necessarily imply the annihilation of the other. This is the main approximation of the method that, as the simulations indicate [25,26], is excellent even when the reaction is imperfect.

The most naive approximation for the average of Eq. (5) consists of the simple factorization of the reaction term: $\langle \Sigma_{\{i(t)\}} H_\epsilon(r-r_{i_{B,A}}(t))n_B(z|\{A\})\rangle \approx n_A^H(z)n_B(z)$, where the superscript H means a space convolution with the normalized function H. This corresponds to a mean field approximation which is only valid at short time intervals. An exact method for the averaging of this type of equations is given in Ref. [25]. With such a method it is possible to find a systematic expansion of products and convolutions of conditional probabilities and mean densities. Here we use this method with the same notation used therein and in Ref. [26].

For the sake of simplicity we only consider one of Eqs. (5). The other equation requires an identical treatment. First we take the integral form of Eq. (5), iterate it and multiply by $\Sigma_{\{i(t)\}}H_\epsilon(r-r_{i_B}(t))$. Averaging this equation, taking into account the factorization properties of the joint probability, we obtain

$$\frac{\partial n_A}{\partial t}=L_A n_A-\gamma \mathcal{A}_A, \tag{6}$$

where the *absorption function*

$$\mathcal{A}_A(z)\equiv\langle \Sigma_{\{i\}}\Delta_i(z)n_A(z|\{B\})\rangle$$

is given by the expansion

$$\begin{aligned}\mathcal{A}_A(z) &= G_{Az,\underline{z'}}n_B^H(z'|t)\,\delta(t')n_A(z')-\gamma_\epsilon G^H_{Bz,\underline{z_1}}\\ &\quad \times G_{Az,z_1}G^H_{Bz_1,\underline{z'}}G_{Az_1,z'}n_B^H(z'|t)\,\delta(t')n_A(z')\\ &\quad -\gamma_\epsilon G^H_{Bz,\underline{z_1}}G_{Az,z_1}\\ &\quad \times n_B^H(z_1|t)G_{Az_1,\underline{z'}}n_B^H(z')\,\delta(t')n_A(z')+\cdots\\ &\quad +(-\gamma_\epsilon)^n G^H_{Bz,\underline{z_1}}G_{Az,z_1}G_{Az_1,\underline{z_2}}\cdots\\ &\quad \times G_{Az_n,\underline{z'}}\,\delta(t')n_A(z')\\ &\quad \times[n_B^H(z_1|t)n_B^H(z_2)\cdots n_B^H(z')\\ &\quad +G^H_{Bz_1,z_2}\cdots G^H_{Bz_n,z'}n_B^H(z'|t)]+\cdots\end{aligned} \tag{7}$$

Here we have used the following shorthand notation: we denote by z_i the pair (r_i,t_i), $\underline{z_i}$ indicates integration over the variables r_i,t_i for all functions containing them, $G_{z,z'}$ is $G(r,t|r',t')$, and $\Delta_i(z)=H_\epsilon(r-r_i(t))$. A more complete picture of this expansion is given by means of a diagrammatic representation as shown in the Appendix of Ref. [26].

Since the density is a monotonically decreasing function of time, the long-time behavior will be dominated by the lowest orders in $n_B^H(t)$, that can be summed giving

$$\mathcal{A}_A(z)\approx T(z,\underline{z_1})n_B^H(z_1|t)G_{Az,\underline{z'}}\,\delta(t')n_A(z'), \tag{8}$$

with

$$T(z,z')=\delta(z-z')-\gamma_\epsilon G_{Az,z'}G^H_{Bz,z'} + \gamma_\epsilon^2 G_{Az,\underline{z_1}}G^H_{Bz,z_1}G_{Az_1,z'}G^H_{Az_1,z'}+\cdots \tag{9}$$

The remaining terms can also be expressed in terms of these functions. Also, in order to get a good approximation for short times we must keep the contribution of the highest orders in n_B^H. Hence, neglecting the crossed terms of mixed products of G_B and n_B^H, $\mathcal{A}_A(z)$ will be the solution of the following integral equation:

$$\mathcal{A}_A(z)=T(z,\underline{z_1})n_B^H(z_1|t)G_{Az_1,\underline{z'}}\delta(t')n_A(z') + T(z,\underline{z_1'})n_B^H(z_1'|z)G_{Az_1',\underline{z_1}}\mathcal{A}_A(z_1). \tag{10}$$

After some more replacements and rearrangements we finally obtain an equation for $n_A(z)$

$$\frac{\partial n_A}{\partial t}=L_A n_A-\gamma_\epsilon T(z,\underline{z_1})n_B^H(z_1|t)n_A(z_1) \tag{11a}$$

valid for short and long times. A completely similar equation is obtained for the averaged density of B particles

$$\frac{\partial n_B}{\partial t}=L_B n_B-\gamma_\epsilon T(z,\underline{z_1})n_A^H(z_1|t)n_B(z_1)\cdot \tag{11b}$$

Equations (11a) and (11b) are the main result obtained in Ref. [26] and will be our starting point in the study of the problem of initially separated reactants. As pointed out earlier, the derivation of $n^H(z_1|t)$ as a function of $n(z)$ is not possible in general making it necessary to resort to some kind of approximation. This will depend on each particular problem, but, as a first approach, two alternatives are possible, either $n(z_1|t)\sim n(z_1)$ or $n(z_1|t)\sim n(z)$. The first (a kind of sudden approximation) seems to be better adapted to problems in which diffusion is the main phenomenon. The second is an adiabatic approximation valid when the shape of the density varies slowly.

III. BEHAVIOR OF THE REACTION FRONT

In order to apply our scheme to the problem of the reaction front we will consider the case of a set of particles A and B with initially separated uniform densities: $n_A(x,t=0)=n_B(-x,t=0)=n_0\theta(x)$, and with the same diffusion coefficient D. L_A and L_B are diffusion operators $L_A=L_B=\Delta$ and the corresponding Green function has the usual form $G_A=G_B=(4\pi Dt)^{-d/2}\exp(-r^2/4Dt)$. Since we restrict our analysis to a one-dimensional situation, we can use the point absorber approach (that is, the limit $\epsilon\to 0$). In such a case, the convoluted functions $n_A^H(z_1|t)$ and G_b^H become ordinary functions $n(x,t)$ and $G_B(x,t|x',t')$. In order to proceed with our calculation, we will partially follow the approach of Ref. [18a]. We define the function $F(x,t)=n_A(x,t)-n_B(x,t)$, that fulfills the equation

$$\frac{\partial F}{\partial t}=D\frac{\partial^2 F}{\partial x^2}, \tag{12}$$

which, due to the indicated initial conditions, has the solution

$$F(x,t)=n_0\mathrm{Erf}\left(\frac{x}{\sqrt{4Dt}}\right). \tag{13}$$

We now rewrite the concentrations of A and B particles as

$$n_A(x,t)=G_1(x,t)+\delta n(x,t), \tag{14a}$$

$$n_B(x,t)=G_2(x,t)+\delta n(x,t), \tag{14b}$$

where

$$G_1(x,t)=\begin{cases} n_0\mathrm{Erf}\left[\dfrac{x}{\sqrt{4Dt}}\right] & x>0\\ 0 & x<0,\end{cases} \tag{15}$$

and also $G_2(x,t)=G_1(-x,t)$. In terms of these functions we can write $F(x,t)$ and $F(|x|,t)$ as

$$F(x,t)=G_1(x,t)-G_2(x,t),$$

$$F(|x|,t)=G_1(x,t)+G_2(x,t).$$

The function defined by $S(x,t)=n_A(x,t)+n_B(x,t)$ must be a continous one given by

$$S(x,t)=F(|x|,t)+2\,\delta n(x,t).$$

Substitution of the forms of $n_A(x,t)$ and $n_B(-x,t)$ into Eqs. (11a) and (11b), reduces this set of equations to the following equation for $\delta n(x,t)$ alone:

$$\frac{\partial\delta n}{\partial t}=D\frac{\partial^2\delta n}{\partial x^2}-\gamma_\epsilon T(z,\underline{z_1})F(|x_1|,t_1)\,\delta n(z_1|t) - \gamma_\epsilon T(z,\underline{z_1})\,\delta n(z_1)\,\delta n(z_1|t), \tag{16}$$

with the boundary condition

$$\delta n_x(0^{\pm},t)=\mp\frac{n_0}{4\sqrt{Dt}},$$

which comes from the continuity of the function $S(x,t)$ at the origin.

We have indicated in Ref. [21] that it is the *absorption function*, i.e., the reaction term of Eq. (16), that is the relevant function for determining the evolution of the concentration of the product C. Here, as we have done there, we also need to resort to Fourier-Laplace transforming this term. Hence, instead of dealing with T through the series in Eq. (9) we consider its Fourier-Laplace transform $T(k,s)$. In the present case it reduces to

$$T(k,s)=\frac{\dfrac{1}{\sqrt{2\pi}}}{1+\dfrac{\gamma_\epsilon\pi}{\sqrt{(Dk)^2+2sD}}}. \tag{17}$$

At this point it is possible to show that the *sudden* approximation described above [i.e., $\delta n(z_1|t)=\delta n(z_1)$] is equivalent to, and yields the same results as, a *mean field* approach. However, guessing that the shape of the δn varies subdiffusively (that is, the typical time scale is slower than $t^{1/2}$), we must resort to the *adiabatic* approximation [i.e., $\delta n(z_1|t) = \delta n(x,t)$]. Hence, we can follow the line of work indicated in Ref. [21], resorting to the Fourier-Laplace transform (Φ_{FL}) of $T(z,\underline{z_1})F(|x_1|,t_1)$, yielding the result

$$\Phi_{\mathrm{FL}}\{T(z,\underline{z_1})F(|x_1|,t_1)\} = n_0 T(k,s)\frac{1}{s}\left[\sqrt{2\pi}\,\delta(k) - \frac{\sqrt{(2s/\pi D)}}{\frac{s}{D}+k^2}\right], \tag{18}$$

where $\Phi_{\mathrm{FL}}\{\ \}$ means the Fourier-Laplace transformation. We then find that the inverse Fourier transform gives

$$L\{T(z,\underline{z_1})F(|x_1|,t_1)\} = \frac{n_0}{s}\big[T(k=0,s) - T(k=i\sqrt{s/D},s) \times \exp(-\sqrt{s/D}|x|)\big], \tag{19}$$

which asymptotically $(s\rightarrow 0)$, and in the approximation $x \ll \sqrt{4Dt}$, reads

$$L\{T(z,\underline{z_1})F(|x_1|,t_1)\} \sim \pi^{-3/2}\left(1-\frac{1}{\sqrt{2}}\right)\frac{n_0}{\gamma}(D/s)^{1/2}. \tag{20}$$

Then, Eq. (16) can be asymptotically written as

$$\frac{\partial \delta n}{\partial t} = D\frac{\partial^2 \delta n}{\partial x^2} - \frac{a}{\sqrt{t}}\delta n - \gamma_\epsilon T(z,\underline{z_1})\,\delta n(z_1)\,\delta n(x,t), \tag{21}$$

with $a = (1-[1/\sqrt{2}])n_0\sqrt{D}$.

An approximate solution of this equation can be obtained using a scaling method. Let us assume the scaling function $\delta n(x,t) \sim t^{-\gamma} g(x/t^\alpha)$. The asymptotic behavior of $T(z,\underline{z_1})\,\delta n(z_1)$ can be obtained as before using the Fourier-Laplace transformation

$$\Phi_{\mathrm{FL}}\{T(z,\underline{z_1})\,\delta n(z_1)\} \simeq T(k,s)\,s^{\gamma-1}k^{-1}G(k/s^\alpha). \tag{22}$$

Substituting Eq. (17) here, taking the asymptotic limit $(s\rightarrow 0)$ and taking the inverse transformation, we obtain

$$T(z,\underline{z_1})\,\delta n(z_1) \simeq t^{-\gamma-\alpha} g\left(\frac{x}{t^\alpha}\right). \tag{23}$$

After substituting the scaling function into Eq.(21) and taking into account the last equation we find that a solution of Eq. (21) exists with the following values for the scaling exponents: $\alpha = \gamma = \frac{1}{4}$. Moreover, the scaling function fulfills the equation $(u = x/t^\alpha)$:

$$D\frac{d^2}{du^2}g - ag - vg^2 = 0, \tag{24}$$

with the boundary condition $g_u(0^\pm) = \mp n_0/(4\sqrt{D})$. The constant v could be determined after solving Eq. (24).

The knowledge of the asymptotic behavior of δn allows us to determine all the relevant quantities. In particular, we can determine the so called reaction rate $R(t)$ [13,18,20] that is the most common quantity determined in simulations. In the present case

$$R \simeq \frac{a}{t^{1/2}}\delta n \simeq t^{-3/4} g(xt^{-1/4}). \tag{25}$$

Concerning the scaling exponents, this result is in complete agreement with the most recent and refined simulations [20]. However, the asymptotic form of the scaling function $[g \sim \exp(-\sqrt{a/Du})]$ resulting from Eq. (24) does not reproduce the Gaussian form found in the indicated simulations. This fact is not unexpected, on one hand because the neglected (crossed) terms in Eq. (7), that are clearly not relevant in the homogeneous case, could play a non-negligible role in the present case; while in the other hand we have the nonuniform convergence of the scaling limit to the true solution [27].

IV. CONCLUSIONS

The recently introduced Galanin-like scheme to describe the reaction $A+B\rightarrow B$ [25], extended to the case of two species annihilation reaction $A+B\rightarrow C$ [26], has been applied to the problem of initially separated reactants in the reaction $A+B\rightarrow C$. The system we have considered is one-dimensional and we have assumed that the evolution between collisions is diffusive (with equal diffusion coefficients for both reactants), and that there is only a finite probability of annihilation in any collision of unlike particles. This method allows us to obtain, within certain approximations, a closed equation for the quantity δn (16) that can be treated through scaling procedures. In such a framework, we have obtained scaling exponents that agree with those obtained in the most recent simulations.

The adequacy of the present method to treat reaction kinetic processes is apparent from the present results as well as previous ones for the case of homogeneous systems and other results corresponding to extensions of the Galanin-like approach for: traps executing a non-Markovian motion, the inclusion of sources, competition between coupled reactions, etc, [28].

However, the analysis of the form of the the scaling function will require further investigation in order to provide a more complete picture of nonhomogeneous situations within the present model. For instance, the analysis of the related problem of a pointlike source injected into a homogeneous substrate [29] is under way.

ACKNOWLEDGMENTS

The authors want to thank to G. Abramson, P. Alemany, and D. Zanette for discussions, S.J. Cornell for providing information related to the present work prior to publication, and V. Grunfeld for the revision of the manuscript. Financial support from Ministerio de Educación y Ciencia, Spain (Project DGICyT No. PB93-0054-C02-02) and from CONICET, Argentina (Project No. PIP-4953/96) are greatly acknowledged. One of us (H.S.W.) thanks the ICTP for the kind hospitality extended to him during his stay at Trieste.

[1] See the recent review by S. Redner and F. Leyvraz, in *Fractals in Science*, edited by A. Bunde and S. Havlin (Springer-Verlag, Berlin, 1994), p. 197, and the references included therein. See also several articles on this subject in the Raoul Kopelman Festschrift issue of J. Phys. Chem. **98** (1994).

[2] S. Havlin and D. Ben-Avraham, Adv. Phys. **36**, 695 (1987); Y.B. Zeldovich and A.S. Mikhailov, Usp. Fiz. Nauk. **153**, 469 (1987) [Sov. Phys. Usp. **30**, 977 (1988)]; K. Lindenberg, B.J. West, and R. Kopelman, in *Noise and Chaos in Nonlinear Dynamical Systems*, edited by F. Moss, L. Lugiato, and W. Schleich (Cambridge University Press, Cambridge, England, 1990), p. 142; D. Ben-Avraham, M.A. Burschka and C.R. Doering, J. Stat. Phys. **60**, 695 (1990).

[3] D. Ben-Avraham and H.L. Frisch, Phys. Rev. A **40**, 505 (1989).

[4] L.W. Anacker and R. Kopelman, Phys. Rev. Lett. **58**, 289 (1987).

[5] S. Kanno Prog. Theor. Phys. **79**, 721 (1988).

[6] E. Clement, L.M. Sander, and R. Kopelman, Phys. Rev. A **39**, 6455 (1989); **39**, 6466 (1989).

[7] F. Leyvraz and S. Redner, Phys. Rev. A **46**, 3132 (1992).

[8] M. Bramson and J.L. Lebowitz, Physica A **168**, 88 (1990).

[9] K. Lindenberg, B.J. West, and R. Kopelman, Phys. Rev. Lett. **60**, 1777 (1988); B.J. West, R. Kopelman, and K. Lindenberg, J. Stat. Phys. **54**, 1429 (1989); K. Lindenberg, B.J. West, and R. Kopelman, Phys. Rev. A **42**, 890 (1990).

[10] H.S. Wio, M.A. Rodriguez, C.B. Briozzo, and L. Pesquera, Phys. Rev. A **44**, R813 (1991); H.S. Wio, in *Instabilities and Nonequilibrium Structures IV*, edited by E. Tirapegui and W. Zeller (Kluwer, Dordrecht, 1993); P.A. Alemany, D.H. Zanette, and H.S. Wio, Phys. Rev. E **50**, 3646 (1994).

[11] L. Galfi and Z. Racz, Phys. Rev. A **38**, 3151 (1988).

[12] Z. Jiang and C. Ebner, Phys. Rev. A **42**, 7483 (1990).

[13] B. Chopard and M. Droz, Europhys. Lett. **15**, 459 (1991); S. Cornell, M. Droz, and B. Chopard, Phys. Rev. A **44**, 4826 (1991); S. Cornell, M. Droz, and B. Chopard, Physica A **188**, 322 (1992).

[14] Y.-E. Lee Koo and R. Kopelman, J. Stat. Phys. **65**, 893 (1991).

[15] H. Taitelbaum, S. Havlin, J. Kiefer, B. Trus, and G.H. Weiss, J. Stat. Phys. **65**, 873 (1991).

[16] H. Taitelbaum, Y.-E. Lee Koo, S. Havlin, R. Kopelman, and G.H. Weiss, Phys. Rev. A **46**, 2151 (1992).

[17] E. Ben-Naim and S. Redner, J. Phys. A **25**, L575 (1992).

[18] (a) H. Larralde, M. Araujo, S. Havlin, and H.E. Stanley, Phys. Rev. A **46**, 855 (1993); (b) **46**, R6121 (1992); (c) M. Araujo, S. Havlin, H. Larralde, and H.E. Stanley, Phys. Rev. Lett. **68**, 1791 (1992).

[19] B. Chopard M. Droz, T. Karapiperis, and Z. Racz, Phys. Rev. E **47**, R40 (1993).

[20] S.J. Cornell, Phys. Rev. Lett. **75**, 2250 (1995); S.J. Cornell, Phys. Rev. E **51**, 4055 (1995).

[21] F.G. Nicolini, M.A. Rodriguez, and H.S. Wio, International Centre for Theoretical Physics Report No. IC/94/162, 1994 (unpublished); M.A. Rodriguez and H.S. Wio, International Centre for Theoretical Physics Report No. IC/94/161, 1994 (unpublished).

[22] B.P. Lee and J. Cardy, Phys. Rev. E **50**, R3287 (1994); B.P. Lee and J. Cardy, J. Stat. Phys. **80**, 971 (1995); M. Howard and J. Cardy, J. Phys. A **28**, 3599 (1995).

[23] D. Avnir and Kagan, Nature (London) **307**, 717 (1984); G.T. Dee, Phys. Rev. Lett. **57**, 275 (1986); R.E. Liesegang, Naturwiss. Wochensch. **11**, 353 (1896); T.A. Witten and L.M. Sander, Phys. Rev. Lett. **47**, 1400 (1981); K.F. Mueller, Science **225**, 1021 (1984).

[24] A.D. Galanin, *Thermal Reactor Theory*, 2nd ed. (Pergamon, New York, 1960); M.M.R. Williams, *Random Processes in Nuclear Reactors* (Pergamon, Oxford, 1974); I. Martinez and M.A. Rodriguez, Ann. Nucl. Energy **12**, 113 (1985).

[25] M.A. Rodriguez, G. Abramson, H.S. Wio, and A. Bru Espino, Phys. Rev. E **48**, 829 (1993); H.S. Wio, G. Abramson, M.A. Rodriguez, and A. Bru Espino, Chaos Solitons Fractals **6**, 575 (1995); G. Abramson and H.S. Wio, *ibid.* **6**, 1 (1995).

[26] G. Abramson, A. Bru Espino, M.A. Rodriguez, and H.S. Wio, Phys. Rev. E **50**, 4319 (1994); G. Abramson, Ph.D. thesis, Instituto Balseiro, Universidad Nacional de Cuyo, Argentina, 1995; A. Bru, Ph.D. thesis, Universidad Complutense de Madrid, Spain, 1995.

[27] A. Schenkel, P. Wittwer, and J. Stubbe, Physica D **69**, 135 (1993).

[28] G. Abramson and H.S. Wio, Phys. Rev. E **53**, 2265 (1996); A.D. Sánchez and H.S. Wio, Physica A (to be published); A.D. Sánchez, E.M. Nicola, and H.S. Wio, Phys. Rev. Lett. **78**, 2244 (1997); S. Bouzat and H.S. Wio (unpublished).

[29] H. Larralde, Y. Lereah, P. Trunfio, J. Dror, S. Havlin, R. Rosenbaum, and H.E. Stanley, Phys. Rev. Lett. **70**, 1461 (1993).