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Asymptotic properties of a reversible $A+B\leftrightarrow C$ (static) reaction-diffusion process with initially separated reactants

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The asymptotic properties of the reaction front formed in a reversible reaction-diffusion process $A+B\leftrightarrow C$ (static) with initially separated reactants are investigated. The case of arbitrary nonzero values of the diffusion constants D_A and D_B and initial concentrations a_0 and b_0 of the reactants A and B is considered. The system is studied in the limit of $t\rightarrow\infty$ and $g\rightarrow 0$, where t and g are the time and the backward reaction rate constant, respectively. The dynamics of the reaction front is described as a crossover between the ''irreversible'' regime at times $t\ll g^{-1}$ and the ''reversible'' regime at times $t\gg g^{-1}$. The general properties of the crossover are studied with the help of an extended scaling approach formulated in this work. On the basis of the mean-field equations the analytical solutions in the reversible regime $t\gg g^{-1}$ inside the reaction zone are discussed. It is shown that in the immobile reaction zone the reaction rate profile has two distinct maxima. This profile differs drastically from the usual single-maximum reaction rate profile inherent in the mobile reaction zone. The two-hump reaction zone profile is the result of the influence of C on the reaction rate in the reversible regime. Numerical computation of the mean-field kinetics equations supports the results of the asymptotic consideration.

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I. INTRODUCTION

The properties of the reaction front created by the irreversible reaction $A+B\rightarrow C$ with initially separated reactants have attracted great interest in the last decade [1–42]. The reaction front is a simple example of pattern formation [43,44], which has been considered from different points of view, including experimental [2,3,6,17,27,28–31].

The standard approach to the problem is to study kinetic equations for the local concentrations $\rho_A(x,t)$, $\rho_B(x,t)$, and $\rho_C(x,t)$, containing the macroscopic reaction rate $R(x,t)$ [1]. Various techniques have been used to study these equations at the asymptotic long-time limit for the cases where the space dimension $d>d_c=2$, when the mean-field expression $R\propto\rho_A\rho_B$ is valid, and for $d\leqslant d_c$, when the explicit form of R is unknown. These techniques are renormalization group analysis [18,19,24], the scaling ansatz [1,21], the quasistationary approximation [11,14], and the approach developed by Koza [25].

Using the scaling ansatz [1], the reaction-diffusion system inside the reaction zone may be represented in the form

$$\rho_A(x,t)=\eta_A t^{-\gamma_A} S_A\left(\frac{x-x_f(t)}{w(t)}\right), \tag{1}$$

$$\rho_B(x,t)=\eta_B t^{-\gamma_B} S_B\left(\frac{x-x_f(t)}{w(t)}\right), \tag{2}$$

$$\rho_C(x,t)=\eta_C t^{-\gamma_C} S_C\left(\frac{x-x_f(t)}{w(t)}\right), \tag{3}$$

$$R(x,t)=\eta_R t^{-\gamma_R} S_R\left(\frac{x-x_f(t)}{w(t)}\right), \tag{4}$$

where S_A, S_B, S_C, and S_R are some scaling functions, $x_f\propto t^{1/2}$ is the point where the reaction rate $R(x,t)$ has a maximum value, $w(t)\propto t^{\alpha}\ll t^{1/2}$ is the width of the reaction zone, η_A, η_B, η_C, and η_R are some parameters independent of x and t, and the exponents α, γ_A, γ_B, γ_C, and γ_R are some positive constants. The values of $\alpha=\frac{1}{6}$, $\gamma_A=\gamma_B=\frac{1}{3}$, $\gamma_C=0$, and $\gamma_R=\frac{2}{3}$ were obtained in the mean-field approximation for nonzero diffusion constants D_A, D_B, and D_C [1,22,42]. For nonzero values of D_A, D_B, and D_C and the initial concentrations a_0 and b_0, the magnitudes of α, γ_A, γ_B, γ_C, and γ_R, and the form of S_A, S_B, S_C, and S_R are independent of the above-mentioned constants η [1,4,18,19,25,42]. The situation is different if even one of the diffusion constants of A, B, or C equals zero and must be considered as a special class [12,26]. It should be noted that the dependence of the system properties on the diffusion constants D_A, D_B, and D_C is not symmetric. The reaction front properties such as w, x_f, $R(x_f,t)$, $R(t)$, $\rho_A(x,t)$, and $\rho_B(x,t)$ are independent of the diffusion constant of C for nonzero diffusion constants of A and B [$R(t)$ is the global reaction rate of C production, which is determined as $\int R_r(x,t)dx$]. These properties essentially change if one of the diffusion constants of A or B equals zero [12,22,26]. In the case of immobile C and nonzero diffusion constants of A and B, only the C profile is changed in comparison to the case when C is mobile [42], i.e., $\rho_C\sim t^{1/3}$ ($\gamma_C=-\frac{1}{3}$) for the immobile reaction zone, while $\rho_C\sim\text{const}$ ($\gamma_C=0$) for the mobile reaction zone.

In the quasistationary approach [11,14], two time scales are considered for the reaction front dynamics, namely, $\tau_J\propto[d(\ln J)/dt]^{-1}\propto t$ and $\tau_F\propto w^2/D\propto t^{2\alpha}$. D is one of the nonzero diffusion constants D_A or D_B. The time scale τ_J determines the change in the diffusive flows $J=J_A=J_B$ of the reactants A and B toward the reaction zone. The time scale τ_F is the equilibration time of the reaction front. From these definitions it follows that $\tau_F/\tau_J\rightarrow 0$ as $t\rightarrow\infty$, for $\alpha<\frac{1}{2}$. This

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means that the reaction in the reaction zone quickly converges to the flows of the reactants and the asymptotic expressions for ρ_A and ρ_B inside the reaction zone may be analyzed by simple quasistatic equations [11]. Time in these equations is included only as a parameter through the time-dependent boundary flows $J_A = J_B = J$. The above analysis can be performed only for nonzero values of D_A and D_B.

The asymptotic properties of the system outside the reaction zone were considered by Koza through the quasistationary approximation [25]. Several quantities were determined analytically as a function of the external parameters D_A, D_B, a_0, and b_0. In particular, the existence of long-time limits of $x_f(t)$, $J(t)$, and $R(t)$ was shown, i.e., as $t \to \infty$, $x_f(t)/\sqrt{t} \to C_f$, $J(t)\sqrt{t} \to C_J$, and $R(t)\sqrt{t} \to C_J$. The value of C_f can be computed from the equations

$$\Phi\left(\frac{-C_f}{2\sqrt{D_A}}\right) = \frac{a_0\sqrt{D_A}}{b_0\sqrt{D_B}}\,\Phi\left(\frac{C_f}{2\sqrt{D_B}}\right), \tag{5}$$

$$\Phi(x) \equiv [1 - \mathrm{erf}(x)]\exp(x^2), \tag{6}$$

where $\mathrm{erf}(x) \equiv 2\pi^{-1/2}\int_0^x \exp(-\eta^2)d\eta$ is the error function.

C_J can be calculated from the expressions

$$C_A = a_0/[\mathrm{erf}(C_f/2\sqrt{D_A}) + 1], \tag{7}$$

$$C_B = -b_0/[\mathrm{erf}(C_f/2\sqrt{D_B}) - 1], \tag{8}$$

$$\begin{aligned} C_J &= C_A\sqrt{D_A/\pi}\,\exp(-C_f^2/4D_A) \\ &= C_B\sqrt{D_B/\pi}\,\exp(-C_f^2/4D_B). \end{aligned} \tag{9}$$

The constants C_A and C_B control the forms of ρ_A and ρ_B outside the reaction zone. For $x \ll x_f - w$,

$$\rho_A(x,t) = a_0 - C_A[\mathrm{erf}(x/\sqrt{4D_A t}) + 1], \tag{10}$$

and for $x \gg x_f + w$,

$$\rho_B(x,t) = b_0 + C_B[\mathrm{erf}(x/\sqrt{4D_B t}) - 1]. \tag{11}$$

These properties change essentially if one of the diffusion constants of A or B equals zero [12,26]. In this case the quasisatic approximation is not valid and the basic approach for analysis is the scaling ansatz (1)–(4) in conjunction with heuristic assumptions related to the reactant profiles outside the reaction zone.

Many publications deal with the irreversible reaction $A + B \to C$; however, chemical reactions are reversible at sufficiently large time. The reversible reaction-diffusion system $A + B \leftrightarrow C$ with initially separated reactants was discussed in Refs. [13,41,42] for long times $t \to \infty$ and sufficiently small g, where g is the backward reaction rate constant. It was shown that the dynamics of the reaction front can be presented as a crossover between the ‘‘irreversible’’ regime at times $gt \ll 1$ and the ‘‘reversible’’ regime at times $gt \gg 1$ [13]. In the ‘‘irreversible’’ regime, the front dynamics is the same as in the irreversible reaction-diffusion system $A + B \to C$. In the ‘‘reversible’’ regime, a local equilibrium at the reaction front exists, and only the diffusion process governs the dynamics. It was shown that near the reaction zone the concentrations of A, B, and C may be expressed in the form of Eqs. (1)–(4) with the exponents γ_A, γ_B, γ_C, and γ_R equal to zero, whereas the reaction front width w, given by $w \sim t^{1/2}$, is independent of the space dimension [13].

In Ref. [41] the refined rate of C production $R_r(x,t)$, including forward and backward reactions, was studied on the basis of the mean-field equations. The study was done for the specific case of equal diffusion constants of A, B, and C and equal initial concentrations of the reactants. It was shown that the reversible regime ($gt \gg 1$) can be characterized by scaling the local rate of C production as $R_{r\mathrm{local}} \sim t^{-1}$, and by scaling the global rate of C production as $R_{r\mathrm{global}} \sim t^{-1/2}$. The general case of arbitrary nonzero diffusion constants and of arbitrary initial reactant concentrations was considered in Ref. [42]. The asymptotic properties of the crossover from the irreversible ($gt \ll 1$) to the reversible ($gt \gg 1$) regime were discussed for $t \to \infty$ and $g \to 0$. It was shown that through this crossover the macroscopic properties of the reaction front are unchanged. These properties are the global rate of C production $R_r(t)$, the motion of the reaction zone center $x_f(t)$, and the concentration profiles of the components outside the reaction front. The analytical expressions for these quantities are the same as in the irreversible regime and are presented in Eqs. (5)–(11) (replacing R by R_r). The profile of C is given by the analytical expressions

$$\rho_C(x,t) = \begin{cases} C_{C1}[\mathrm{erf}(x/\sqrt{4D_C t}) + 1], & x \ll x_f - w, \quad (12) \\ -C_{C2}[\mathrm{erf}(x/\sqrt{4D_C t}) - 1], & x \gg x_f + w. \quad (13) \end{cases}$$

The constants C_{C1} and C_{C2} are determined by

$$\begin{aligned} C_{C1} &= C_J\sqrt{\pi/4D_C}\,\Phi\left(\frac{C_f}{2\sqrt{D_C}}\right), \\ C_{C2} &= C_J\sqrt{\pi/4D_C}\,\Phi\left(\frac{-C_f}{2\sqrt{D_C}}\right). \end{aligned} \tag{14}$$

All these expressions are true if some of the natural assumptions are valid [42]. A most important assumption relates to the type of dependence $w = w(t,g)$. This dependence is assumed to be a monotonic link of two regions at $gt \sim 1$: $w \sim t^{\alpha}$ at times $gt \ll 1$ and $w \sim \sqrt{t}g^{\beta}$ at times $gt \gg 1$, where β is some positive constant. This assumption follows from the requirement that the ratio $\tau_F/\tau_J \sim w^2(t)/Dt \to 0$ as $t \to \infty$ and $g \to 0$ and it is essential in order that the quasistaic equations can be used for arbitrary values of gt. It should be emphasized that, as in Ref. [25], the assumptions do not restrict the form of R, and therefore the results are true also for the case of $d \leq d_c$, when the mean-field expression for R is not valid.

The results of the heuristic arguments were tested in Ref. [42] by computing the mean-field kinetic equations. In particular, the expressions for ρ_A, ρ_B, ρ_C, and R_r inside the reaction zone in the reversible regime $gt \gg 1$ for $g \to 0$ were obtained analytically. They have the form of Eqs. (1)–(4) with the exponents γ_A, γ_B, and γ_C equal to zero, $\gamma_{R_r} = 1$, $w \sim \sqrt{gt}$, $\eta_A \sim \eta_B \sim \eta_R \sim g^{1/2}$, and $\eta_C \sim g^0$.

The analysis in Ref. [42] was performed in the framework of the quasistatic approximation, which can be used only for nonzero diffusion constants of A, B, and C. When $D_C = 0$, the quasistatic approximation fails and the problem relates essentially to a different class, requiring a different technique for analysis. The scaling approach is an appropriate technique for this special case of $D_C=0$. The central theme of our work is the extension of this approach to the reversible regime and the crossover from the irreversible to the reversible regime. By this extended scaling approach the asymptotic properties of the reversible $A+B\leftrightarrow C$ (static) reaction-diffusion process with initially separated reactants are studied for $t\to\infty$ and $g\to 0$ assuming $D_C=0$, $D_A>0$, and $D_B>0$.

In Sec. II the change in the dynamics of the system with $D_C>0$ in the limit $D_C\to 0$ is presented. The general behavior of the system is studied in Sec. III through the extended scaling ansatz. Section IV is devoted to the analytical and numerical analysis of the mean-field kinetic equations. Section V summarizes the results of our work.

II. THE *C* PROFILE OUTSIDE THE REACTION ZONE IN THE LIMIT $D_C\to 0$

Consider macroscopic properties describing Eqs. (5)–(14) for some definite positive values of D_A, D_B, a_0, and b_0, and $D_C\to 0$. The values of $x_f(t)$, $R_r(t)$, $J(t)$, ρ_A, and ρ_B do not depend on D_C and therefore expressions (5)–(11) do not vary when the diffusion constant D_C goes to zero. This indicates that the macroscopic properties mentioned do not change through the crossover between the irreversible and the reversible regime, as in the case of nonzero D_C [42]. The numerical simulation of the mean-field equations in Sec. III confirms this conjecture.

D_C appears only in Eqs. (12)–(14) and therefore only the variation of ρ_C with $D_C\to 0$ will be considered. Assume that $C_f\neq 0$ or more specifically $C_f>0$, i.e., the reaction zone asymptotically moves to the right. Taking into account that $\lim\Phi(x)\to 2\exp(x^2)\to\infty$ for $x\to-\infty$ and $\lim\Phi(x)\to 1/(2x\sqrt{\pi})$ for $x\to+\infty$, we obtain for $x\gg x_f+w$ and $x<0$ $\rho_C(x,t)\to 0$ and for $0<x\ll x_f-w$ $\rho_C(x,t)\to c_0=2C_J/C_f$. At $C_f=0$, the reaction zone is asymptotically immobile. From Eq. (14) it follows that $C_{C1}=C_{C2}=C_J\sqrt{\pi/4D_C}$ and as $D_C\to 0$ $\rho_C(x,t)\to 0$ for $x\gg x_f+w$ and for $x\ll x_f-w$.

The expression $c_0C_f=2C_J$ is the same as that for b_0 for the case of $D_B\to 0$ ($b_0C_f=2C_J$) [26] and has the same physical interpretation. c_0 is the concentration level of C produced on the movable reaction front. The total amount of C produced by time t is asymptotically equal to $\int_0^t J_A(\tau)d\tau=\int_0^t J_B(\tau)d\tau\approx 2C_J\sqrt{t}$. This number may be estimated also as $\int_{-\infty}^{+\infty}\rho_C(x,t)dx\approx c_0x_f\approx c_0C_f\sqrt{t}$, which leads to $c_0C_f=2C_J$.

Thus it can be seen that the two cases of the mobile and immobile reaction zones correspond to essentially different C profiles outside the reaction zone. Consequently, these two cases belong to two different classes, which will be considered separately. Note that the above results coincide with the known reaction zone properties in the irreversible regime $gt\ll 1$ [12,22].

As indicated above, the C profile in the limit $D_C\to 0$ for a mobile reaction front has a steplike form with infinite gradient at the point $x=0$. This profile is a result of the fact that reaction in this region was ignored. This is true at $D_C>0$; however, for the case $D_C=0$ the reaction must be taken into account. In the irreversible regime $gt\ll 1$ at the early time period, when the reaction zone is close to the point $x=0$, C production is concentrated in the reaction zone with a finite width that includes the point $x=0$. From the movement of the reaction zone it follows that the level of ρ_C gently decreases from c_0 to zero. For the long-time limit $t\to\infty$ ($gt\ll 1$) the reaction rate becomes negligibly small and the C profile is independent of time in this region. In the reversible regime $gt\gg 1$ the situation is more complex. Because of the reversible reaction $A+B\leftrightarrow C$, continuous exchange between the mobile component B and the immobile component C occurs. In the region $x>0$ the C component disappears through the reaction $A+B\leftarrow C$ by producing B. Thus B diffuses into the region $x<0$, interacting with A by the reaction $A+B\to C$, and producing C. In this manner the component C is transferred from $x>0$ to $x<0$ and a continuous profile of C is produced. This process may be referred to as effective diffusion of the component C or as the distinctive reaction zone structure related to a first order reversible reaction. The analytical description of the reaction zone in the mean-field approximation for the reversible regime is discussed in the Appendix.

III. EXTENDED SCALING ANSATZ AND COUNTERPART OF QUASISTATIC EQUATIONS

The reversible reaction-diffusion system $A+B\leftrightarrow C$ (static) is described by the following equations [13]:

$$\frac{\partial\rho_A}{\partial t}=D_A\frac{\partial^2\rho_A}{\partial x^2}-R+g\rho_C,$$

$$\frac{\partial\rho_B}{\partial t}=D_B\frac{\partial^2\rho_B}{\partial x^2}-R+g\rho_C, \tag{15}$$

$$\frac{\partial\rho_C}{\partial t}=+R-g\rho_C,$$

with the initial state given by

$$\rho_A(x,t=0)=a_0H(-x),$$

$$\rho_B(x,t=0)=b_0H(+x),\quad \rho_C(x,t=0)=0, \tag{16}$$

where $H(x)$ is the Heaviside step function. Let us study the long-time behavior $t\to\infty$ of the system in the reaction zone region $|x-x_f|\leqslant\sqrt{4D_At},\sqrt{4D_Bt}$ assuming that $g\to 0$. The boundary conditions of the components will be formulated in accordance with their macroscopic distribution outside the reaction zone obtained in Sec. II.

The well-known scaling approach [1,26] can be applied in the irreversible regime $gt\ll 1$. In this regime the term $g\rho_C$ can be canceled in Eqs. (15). Assume that the scaling forms of Eqs. (1)–(4) are valid; then the following evaluations can be written: $\partial\rho_A/\partial t\propto t^{-\gamma_A-\alpha-1/2}(C_f\neq 0)$ or $\partial\rho_A/\partial t\propto t^{-\gamma_A-1}(C_f=0)$ and $D_A\partial^2\rho_A/\partial x^2\propto t^{-\gamma_A-2\alpha}$. Taking into account that $\alpha<\frac{1}{2}$, it can be seen that in the limit $t\to\infty$ the term $\partial\rho_A/\partial t$ is small compared to $D_A\partial^2\rho_A/\partial x^2$. The same procedure can be performed for ρ_B also, and therefore Eqs. (15) in $|x-x_f|\leqslant\sqrt{4D_At},\sqrt{4D_Bt}$ can be simplified to

$$0=D_A\frac{\partial^2\rho_A}{\partial x^2}-R,$$

$$0=D_B\frac{\partial^2\rho_B}{\partial x^2}-R, \tag{17}$$

$$\frac{\partial\rho_C}{\partial t}=+R,$$

with boundary conditions

$$D_A\partial\rho_A/\partial x\to -J_A(t),\quad \rho_B\to 0,$$

$$\rho_C\to c_0\ \ (C_f\neq 0)\quad \text{or}\quad \rho_C\to 0\ \ (C_f=0),\quad x\to-\infty, \tag{18a}$$

$$\rho_A\to 0,\quad D_B\partial\rho_B/\partial x\to J_B(t),\quad \rho_C\to 0,\quad x\to+\infty. \tag{18b}$$

Here $J_A(t)=J_B(t)=J(t)\propto t^{-1/2}$ [25].

Equations (17) with the boundary conditions (18) are the counterpart of the quasistatic approximation equations [42] for the case of nonzero D_C in the irreversible regime $gt\ll 1$. The following relations for the scaling exponents can be found from Eqs. (17) and (18):

$$\gamma_A=\gamma_B=\tfrac{1}{2}-\alpha,\quad \gamma_R=\tfrac{1}{2}+\alpha,\quad \gamma_C=0\ \ (C_f\neq 0),$$

$$\gamma_C=-\tfrac{1}{2}+\alpha\ \ (C_f=0). \tag{19}$$

Note that, if $D_C>0$, $\gamma_C=0$ [42], and the value of γ_C is the same as in the mobile reaction zone, but different from that in the immobile reaction zone $C_f=0$.

Consider now the reversible regime $t\to\infty$, $gt\gg 1$. In this regime the reaction rate $R_r\equiv R-g\rho_C\approx 0$ in the zero approximation [41] and the concentration profiles can be described by

$$0=-R+g\rho_C,$$

$$\frac{\partial\rho_A}{\partial t}+\frac{\partial\rho_C}{\partial t}=D_A\frac{\partial^2\rho_A}{\partial x^2}, \tag{20}$$

$$\frac{\partial\rho_B}{\partial t}+\frac{\partial\rho_C}{\partial t}=D_B\frac{\partial^2\rho_B}{\partial x^2}.$$

The reaction rate R_r can be calculated as a first approximation by substituting the solutions obtained from Eqs. (20) into any one of the original Eqs. (15) [41].

To extend the scaling approach to the reversible regime, assume that the scaling forms (1)–(4) obtained in Ref. [42] in the quasistatic approximation for the reversible regime $t\to\infty$, $g\to 0$, and $gt\gg 1$ are valid also in our case for the immobile reaction product *C*. Consequently, the following scaling relations may be assumed: $w\sim t^{0.5}g^{\beta}$, $\eta_A\sim g^{\phi_A}$, $\eta_B\sim g^{\phi_B}$, $\eta_C\sim g^{\phi_C}$, and $\eta_R\sim g^{\phi_R}$ $(\gamma_A=\gamma_B=\gamma_C=0,\ \gamma_{R_r}=1)$.

Evaluating $\partial^2\rho_A/\partial x^2$, $\partial\rho_A/\partial t$, etc. in the same manner as in the irreversible regime, the simplified equations in the region $|x-x_f|\leqslant\sqrt{4D_At},\sqrt{4D_Bt}$ are obtained as

$$0=-R+g\rho_C,$$

$$\frac{\partial\rho_C}{\partial t}=D_A\frac{\partial^2\rho_A}{\partial x^2}, \tag{21}$$

$$\frac{\partial\rho_C}{\partial t}=D_B\frac{\partial^2\rho_B}{\partial x^2},$$

with the boundary conditions of Eqs. (18). The relations between the scaling exponents may be obtained from Eqs. (21) and the boundary conditions (18) as

$$\phi_A=\beta,\quad \phi_B=\beta,\quad \phi_{R_r}=-\beta,$$

$$\phi_C=0\ \ (C_f\neq 0)\quad \text{or}\quad \phi_C=-\beta\ \ (C_f=0). \tag{22}$$

The difference between the case of $D_C>0$ and our case $D_C=0$ is related only to the asymptotically immobile reaction zone situation $C_f=0$, i.e., $\phi_C=-\beta$ if $D_C=0$ and $\phi_C=0$ if $D_C>0$. For a mobile reaction zone the scaling exponents are the same either for $D_C>0$ or for $D_C=0$, but the equations for these two cases are different.

The general physical reason for the formation of the well-defined thin reaction zone in both the irreversible and reversible regimes is the small value of the nondimensional ratio $w(g,t)/2\sqrt{Dt}$, where D is one of the nonzero diffusion constants D_A or D_B. With this general feature in mind, the scaling relations in the irreversible $gt\ll 1$ and the reversible $gt\gg 1$ regimes may be extended to the region of arbitrary $gt\sim 1$ $(t\to\infty,\ g\to 0)$ by scaling to the nondimensional ratio $p=w(g,t)/2\sqrt{Dt}\to 0$. Suppose that, as in the quasistationary case [42], the dependence $w=w(g,t)$ links two parts monotonically at $gt\sim 1$, namely, $w\sim t^{\alpha}$ at times $gt\ll 1$ and $w\sim\sqrt{t}g^{\beta}$ at times $gt\gg 1$. Thus the scaling relations may be written as

$$\rho_A(x,t)=\eta_Ap^{-\chi_A}S_A\left(\frac{x-x_f(t)}{w(g,t)}\right), \tag{23}$$

$$\rho_B(x,t)=\eta_Bp^{-\chi_B}S_B\left(\frac{x-x_f(t)}{w(g,t)}\right), \tag{24}$$

$$\rho_C(x,t)=\eta_Cp^{-\chi_C}S_C\left(\frac{x-x_f(t)}{w(g,t)}\right), \tag{25}$$

$$R_r=\eta_{R_r}t^{-1}p^{-\chi_{R_r}}S_{R_r}\left(\frac{x-x_f(t)}{w(g,t)}\right). \tag{26}$$

The form of Eq. (26) follows from the diffusionlike character of Eqs. (15). Indeed, bearing in mind that Eqs. (23)–(26) hold in the irreversible regime, the following thoughts are proposed. By taking into account Eq. (25) we have $R_r(x,t)=\partial\rho_C/\partial t\sim\partial(\ln w)/\partial t$, and consequently in the irreversible regime it can be shown that for $w\sim t^{\alpha}$ [1,8,40] and for $w\sim t^{\alpha}(\ln t)^{\delta}$ [23,24] (δ is a positive constant) the factor

$\sim t^{-1}$ appears. Note that in the case of the Sinai disorder model $w \sim (\ln t)^2$ [37] and $\sim (t \ln t)^{-1}$ replaces the factor $\sim t^{-1}$.

The simplified equations can be obtained by evaluating $\partial^2 \rho_A / \partial x^2, \partial \rho_A / \partial t$, etc. in the same way as in the irreversible and reversible regimes. The equations and boundary conditions obtained coincide with Eqs. (17) and only R has to be replaced by $R_r = R - \rho_C g$. The relations between the scaling exponents follow from these equations and are given by

$$\chi_A = 1, \quad \chi_B = 1, \quad \chi_{R_r} = -1,$$

$$\chi_C = 0 \ \ (C_f \neq 0) \quad \text{and} \ \ \chi_C = - \ \ (C_f = 0). \tag{27}$$

Taking into account the fact that generalized scaling is true either for $gt \ll 1$ or for $gt \gg 1$, the following summaries of Eqs. (19), (22), and Eqs. (27) can be obtained:

$$\gamma_A / (0.5 - \alpha) = -\phi_A / \beta = \chi_A = 1,$$

$$\gamma_B / (0.5 - \alpha) = -\phi_B / \beta = \chi_B = 1,$$

$$\gamma_C / (0.5 - \alpha) = -\phi_C / \beta = \chi_C = 0 \ \ (C_f \neq 0) \ \ \text{or} \ \ -1 (C_f = 0),$$

$$(\gamma_{R_r} - 1) / (0.5 - \alpha) = -\phi_{R_r} / \beta = \chi_{R_r} = -1. \tag{28}$$

These relations reflect the fact that the dependence of the reaction front width $w(g,t)$ on g and t controls the character of the complete reaction-diffusion process, i.e., the magnitudes of all the exponents. For the irreversible regime these values are determined by α and for the reversible regime by β. In the generalized scaling expression Eqs. (23)–(26) the dependence of $w(g,t)$ on g and t is not indicated, and because of this the exponents are determined uniquely.

Thus we can see that the condition $p = w(g,t)/2\sqrt{Dt} \rightarrow 0$ for $t \rightarrow \infty$ and $g \rightarrow 0$ is sufficient to obtain the simplified equations for the irreversible and reversible regimes and for the crossover between them [Eqs. (17) and (21) and modifications of Eq. (17)]. For the mobile components A and B with nonzero diffusion constants, these equations are the same as in the case of $D_C > 0$, i.e., the derivative time terms are neglected in the first and second of Eqs. (15). For the immobile component C the term $D_C \partial^2 \rho_C / \partial x^2$ is canceled and the term $\partial \rho_C / \partial t$ substitutes it. This term can be imagined to be different for the mobile and immobile reaction zones. As a consequence the scaling relations are different in these two cases. This can be seen directly by transforming to a coordinate system that is moving with the reaction zone, $x' = (x - x_f) - (dx_f/dt)t$, $t' = t$,

$$\frac{\partial \rho_C}{\partial t} = \frac{\partial \rho_C}{\partial t'} + \frac{dx_f}{dt} \frac{\partial \rho_C}{\partial x'}. \tag{29}$$

In the case of a moving reaction zone the second term is dominant, while for an immobile reaction zone $dx_f/dt = 0$ the first term becomes essential. Consequently, in the first case $C_f \neq 0$ and a quasistatic equation with the time-dependent parameter $dx_f/dt = v_f(t) \sim t^{-1/2}$ is obtained, i.e., $v_f(t) \partial \rho_C / \partial x = -R_r$. This equation, and therefore the full system, have quasistatic character in the moving reaction zone case. In the second case $C_f = 0$, the initial form of the equation does not change and it is given by $\partial \rho_C / \partial t = R_r$. Thus we can see that the simplified kinetic equations are indicative of the specific features of the asymptotically immobile reaction.

One can arrive at the same conclusion from the scaling exponents. Furthermore, taking into account the scaling exponents for the case $D_C > 0$, it may be concluded that only the case $C_f = 0$ and $D_C = 0$ relates to a different specific class. The three other cases ($C_f \neq 0$, $D_C = 0$; $C_f = 0$, $D_C > 0$; and $C_f \neq 0$, $D_C > 0$) relate to the same class.

IV. MEAN-FIELD APPROXIMATION: ASYMPTOTIC SOLUTIONS AND NUMERICAL SIMULATION

Assume that the mean-field approximation expression $R = k \rho_A \rho_B$ is true. From this relation the following values of the exponents may be calculated.

(a) In the irreversible regime, from $R = k \rho_A \rho_B$ we have $\gamma_A + \gamma_B = \gamma_R$ and taking into account Eqs. (19) the following values can be obtained: $\alpha = \frac{1}{6}$, $\gamma_A = \gamma_B = \frac{1}{3}$, $\gamma_C = 0$ ($C_f \neq 0$) or $\gamma_C = -\frac{1}{3}$ ($C_f = 0$), in accordance with Refs. [1,42].

(b) In the reversible regime, from the equation $g \rho_C = k \rho_A \rho_B$ it follows that $\phi_A + \phi_B = \phi_C + 1$ and with expressions (22), for $C_f \neq 0$ we obtain $\beta = \frac{1}{2}$, $\phi_A = \phi_B = \frac{1}{2}$, $\phi_{R_r} = -\frac{1}{2}$, $\phi_C = 0$. It $C_f = 0$ then $\beta = \frac{1}{3}$, $\phi_A = \phi_B = \frac{1}{3}$, $\phi_C = \phi_{R_r} = -\frac{1}{3}$.

On the basis of Eqs. (21) and (18) and appropriate scaling expressions given by Eqs. (1)–(4) the concentration profiles and reaction rate distribution can be calculated for $t \rightarrow \infty$ and $g \rightarrow 0$ in the reversible regime $gt \gg 1$. For the asymptotically mobile reaction zone $C_f \neq 0$ we have

$$\rho_A(x,t) = \sqrt{(2gc_0/k)(D_B/D_A)} f_A(\xi/w_1), \tag{30}$$

$$\rho_B(x,t) = \sqrt{(2gc_0/k)(D_A/D_B)} f_B(\xi/w_1), \tag{31}$$

$$\rho_C(x,t) = 2c_0 f_A(\xi/w_1) f_B(\xi/w_1) = c_0 f_C(\xi/w_1), \tag{32}$$

$$R_r(x,t) = (c_0/t) f_{R_r}(\xi/w_1). \tag{33}$$

Here $\xi \equiv x - x_f$, $w_1 = 2\sqrt{(gtD_A D_B)/(kC_J C_f)}$,

$$f_A(y) \equiv \exp(-y^2)/\sqrt{\pi}[1 + \text{erf}(y)],$$

$$f_B(y) \equiv y + \exp(-y^2)/\sqrt{\pi}[1 + \text{erf}(y)],$$

$$f_{R_r}(y) \equiv -y[f_A(y) + y f_A'(y) + 2\, f_A(y) f_A'(y)].$$

In the limit of large values of y, $f_A(y) \rightarrow \exp(-y^2)/2\sqrt{\pi}$, $f_B(y) \rightarrow y$, $f_{R_r}(y) \rightarrow y^3 \exp(-y^2)/\sqrt{\pi}$ as $y \rightarrow +\infty$; and $f_A(y) \rightarrow -y$, $f_B(y) \rightarrow -1/(2y)$, $f_{R_r}(y) \rightarrow 3/(2y^2)$ as $y \rightarrow -\infty$. Note that decay of $f_{R_r}(y)$ depends on direction: for $y \rightarrow +\infty$, $f_{R_r}(y) \sim \exp(-y^2)$, while for $y \rightarrow -\infty$, $f_{R_r}(y) \sim y^{-2}$. For the asymptotically immobile reaction zone $C_f = 0$ and we have

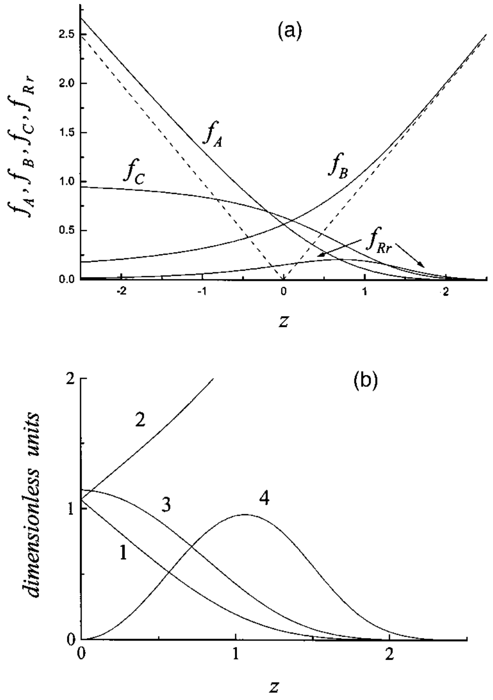

FIG. 1. The form of *A*, *B*, *C*, and R_r profiles calculated in the framework of the mean-field approximation in the reversible regime $gt \gg 1$. (a) Asymptotically mobile reaction zone $C_f > 0$ case described by Eqs. (30)–(33); and (b) asymptotically immobile reaction zone case $C_f = 0$ described by Eqs. (34)–(37). $1 - [f_u(z) - z] \sim f_A$, $2 - [f_u(z) + z] \sim f_B$, $3 - [{f_u}^2(z) - z^2] \sim f_C$, $4 - {f_u}''^{(z)} \sim f_{Rr}$. $f_u(z)$ was computed numerically. Functions and coordinate *z* are given in dimensionless units.

$$\rho_A(x,t) = (C_J/D_A)\sqrt[3]{(gD_AD_B)/(2C_Jk)}[f_u(x/w_2) - (x/w_2)], \tag{34}$$

$$\rho_B(x,t) = (C_J/D_B)\sqrt[3]{(gD_AD_B)/(2C_Jk)}[f_u(x/w_2) + (x/w_2)], \tag{35}$$

$$\rho_C(x,t) = (C_J/2)\sqrt[3]{(2C_Jk)/(gD_AD_B)}[f_u^2(x/w_2) - (x/w_2)^2], \tag{36}$$

$$R_r(x,t) = (C_J/2t)\sqrt[3]{(2C_Jk)/(gD_AD_B)}f_u''(x/w_2). \tag{37}$$

Here $w_2 = 2\sqrt{t}(gD_AD_B/2kC_J)^{1/3}$. $f_u(z)$ is the solution of the equation $f_u'' + z[f_u^2 - z^2)]' = 0$ with boundary conditions $f_u'(z=0) = 0$ and $f_u(z \to \infty) = z$. For $z < 0$ $f_u(z) \equiv f_u(-z)$. For $z \gg 1$ $f_u(z) \sim z + 1/(2z)$.

The solutions for the asymptotically mobile and asymptotically immobile reaction zones are shown in Figs. 1(a) and 1(b), respectively. The profiles of the *A*, *B*, and *C* components and the production rate R_r in the mobile reaction zone, shown in Fig. 1(a), clearly indicate that in the reversible regime the production of *C* is concentrated near the reaction front center, as in the irreversible regime. For the immobile reaction zone situation, shown in Fig. 1(b), the reaction rate profile has a characteristic two-maximum form. It is drastically different from the mobile reaction zone picture, and from the immobile reaction zone case in the irreversible regime. The *C* concentration in the center of the reaction zone is constant, because of the reversible character of the reaction, and this constant concentration level penetrates in two directions. The immobile *C* component concentrates near the reaction zone center and suppresses additional *C* production, namely, the production rate R_r equals zero in the reaction zone center. Far from the center the reaction rate is also small, because of the absence of the reactants *A* and *B*. In the irreversible regime [12,22] all reaction front characteristics, excluding the *C* profile, are the same, regardless if the mobile or immobile reaction zone is considered.

Note that the R_r profile described by Eqs. (37) is symmetrical and independent of the relation between D_A and D_B. This is a result of the even character of the function $f_u(z)$.

As mentioned in Sec. II, the distinctive reaction zone structure is formed near the $x = 0$ region in the case of a mobile reaction zone in the reversible regime. Near $x = 0$ the asymptotic solution can be obtained (see the Appendix) as shown below:

$$\rho_A(x,t) \approx \rho_A(0,t) = a_0 - C_A, \tag{38}$$

$$\rho_B(x,t) = [gc_0/(a_0 - C_A)]0.5[1 + \mathrm{erf}(x/w_3)], \tag{39}$$

$$\rho_C(x,t) = c_0 0.5[1 + \mathrm{erf}(x/w_3)], \tag{40}$$

$$R_r(x,t) = -c_0(1/t)(1/2\sqrt{\pi})(x/w)\exp(-x^2/{w_3}^2). \tag{41}$$

Here $w_3 = 2\sqrt{(gD_Bt)/[k(a_0 - C_A)]}$.

This solution describes two simultaneous processes, (a) the diffusion of *B* and (b) its exchange with *C*. Note that in Eqs. (38)–(41) $g \to 0$, $w_3 \sim \sqrt{g} \to 0$ are assumed, and the gradient of *A* near $x = 0$ is not taken into account.

To test the asymptotic properties of the reaction-diffusion process $A + B \leftrightarrow C$, Eqs. (15) and (16) in the mean-field approximation were solved numerically by an exact enumeration method [5,12,41,45,46]. A one-dimensional discrete lattice is considered. In the beginning the diffusion part is calculated and after this step the reaction is considered. The reaction step is calculated on the basis of Eqs. (15) without the diffusion terms. The local reaction rate is calculated from $R_{r\mathrm{local}}(j) \equiv R_r(j)$, where *j* is a discrete spatial point. The time step equals 1. The constants $k = 0.1$ and $g = 10^{-4}, 10^{-5}$ were used.

The results of the simulation confirm that the general properties of the crossover are the same as in the case $D_C > 0$, i.e., the macroscopic properties of the reaction front (such as the global rate of *C* production, the motion of the reaction zone center, and the concentration profiles of the components outside the reaction front) are unchanged [42]. In particular, there were no changes in the global rate $R_r(t) \sim n^{-1/2}$ and the coordinate of the reaction zone center $x_f(t) \sim n^{+1/2}$. The crossovers of the local rate of *C* production $R_r(x,t)$ and the width of the reaction zone *w* are associated with a change in the exponents, from $\sim n^{-2/3}$ to $\sim n^{-1}$ and from $\sim n^{+1/6}$ to $\sim n^{+1/2}$, respectively. The monotonic growth $w = w(g,n)$ supports the assumption $p = w(g,t)/2\sqrt{Dt} \to 0$, which is valid in the case of the mean-field approximation.

In Figs. 2 and 3 the results of the numerical calculation of the reaction rate profile $R_r(x,t)$ for $C_f > 0$ ($a_0 = 2b_0 = 1.0$, $D_A = 4D_B$) and for $C_f = 0$ ($a_0 = 2b_0 = 1.0$, $D_A = 0.25D$) are shown, respectively. For comparison the asymptotic so-

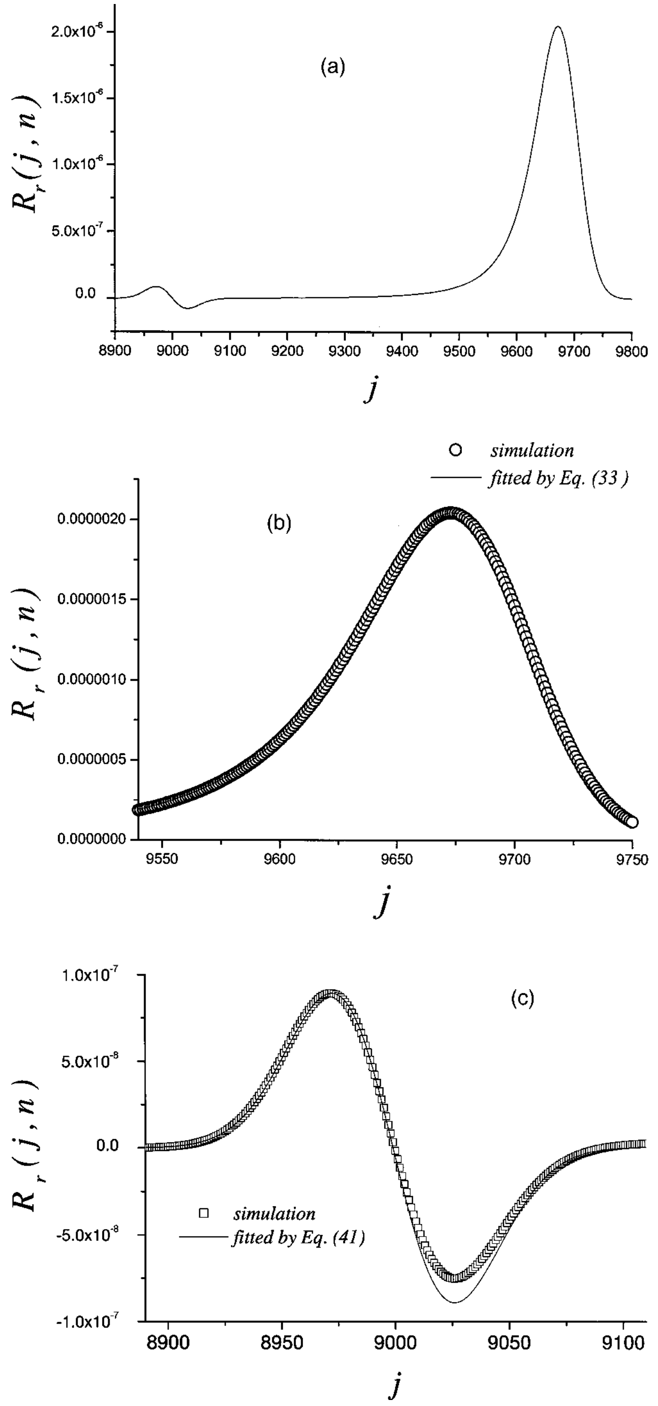

FIG. 2. Simulated profile of $R_r(j,n)$ for mobile reaction zone case $C_f>0$ at time $n=10^6$ ($k=0.1$, $g=10^{-4}$, $D_A=4D_B$, $a_0=2b_0$). Here j is a discrete space step number. General view of $R_r(j,n)$; (b) $R_r(j,n)$ profile inside reaction zone $x\sim x_f$ fitted by the asymptotic solution Eq. (33); (c) $R_r(j,n)$ profile near $x\sim 0$ fitted by Eq. (41).

lutions Eq. (33) and (41) (mobile reaction zone) and Eq. (37) (immobile reaction zone) are also presented.

The simulation data of Figs. 2(b) and 2(c) fit the curves obtained from Eq. (33) and Eq. (41) rather well. The data of Fig. 2(c), show some asymmetry. This asymmetry is related to the finite gradient of *A*, which was neglected in the calculations of Eqs. (38)–(41).

In Fig. 3 two maxima in the profile of R_r exist. The deviation in the two maxima is associated with the asymptotic character of the solution (37), which is true only for sufficiently small values of g and large values of t. Indeed, for $D_A\neq D_B$ the C distribution for short times $t\ll t_c\sim k^{-1}$, when the reaction may be neglected, is asymmetrical [12]. In the irreversible regime $t_c\sim k^{-1}\ll t\ll g^{-1}$, the reaction rate profile is symmetrical [1,22], independent of the diffusion constant ratio. The C profile in this regime may be considered as the sum of a short-time asymmetrical distribution part and the symmetrical C distribution that is produced in the irreversible regime. With the elapse of time, the profile becomes more and more symmetric. After the crossover to the reversible regime at times $gt\gg 1$, C start to influence the reaction rate $R_r(t)$, and the residual asymmetry in the C distribution leads to asymmetry in the $R_r(x,t)$ profile. The accurate symmetrical $R_r(x,t)$ profile may be obtained only in the limit $t\to\infty$ and $g\to 0$ or if $D_A=D_B$, when no short-time asymmetry exists. Simulations of different ratios of D_A/D_B and different values of g confirm our conclusions. In addition to this asymmetrical memory effect, it was also observed that $R_r(x,t)$ in the region near $x=0$ takes a negative value during the time of the crossover, and its value approaches zero from the negative side. Further investigation is required to determine the nature of this effect.

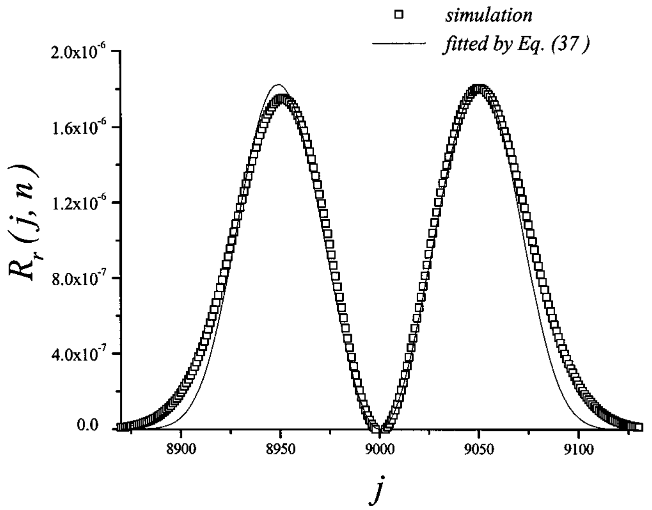

FIG. 3. Simulated profile $R_r(j,n)$ for immobile reaction zone case $C_f=0$ at time $n=10^6$ ($k=0.1$, $g=10^{-5}$, $D_A=0.25D_B$, $a_0=2b_0$). Here j is a discrete space step number. The profile inside the reaction zone fitted by the asymptotic solution Eq. (37), which is shown in Fig. 1(b).

Thus, the simulation indicates that in the case of the asymptotically immobile reaction zone the behavior of the reaction-diffusion system $A+B\leftrightarrow C$ in the reversible regime is influenced by the earlier states of the system. In particular, the C distribution obtained for short times, when the reaction may be neglected, and C distributions in the irreversible regime influence the reaction rate $R_r(x,t)$ profile in the reversible regime. Consequently, the solutions presented by Eqs. (37) are true only in the limits $t\to\infty$ and $g\to 0$. The investigation of the memory effect will be reported elsewhere.

V. SUMMARY

(1) The scaling approach extended to the case of the reversible regime ($gt\gg 1$) and to the crossover from the irre-

versible to the reversible regime (arbitrary values of gt) is formulated. The main assumption for its application is that the dependence $w=w(g,t)$ may be imagined as a monotonic link at $gt\sim 1$ between two parts, $w\sim t^{\alpha}$ at times $gt\ll 1$ and $w\sim\sqrt{t}g^{\beta}$ at times $gt\gg 1$. Some additional scaling exponents and the relations between them are introduced.

(2) In the framework of this extended scaling approach the diffusion-reaction system $A+B\leftrightarrow C$ (static) with initially separated reactants is investigated. Simplified kinetic equations, the counterpart of the quasistatic equations of the case $D_C>0$, are obtained. It is shown that for a mobile reaction zone the system is described by quasistatic equations that depend on time through the boundary currents and the velocity of the reaction zone center $v=v_f(t)$. For the immobile reaction zone the system of equations is nonquasistatic. A general analysis of these equations and the respective boundary conditions shows that the cases $D_C>0$ and $D_C=0$ fall into two different classes only for the case of the immoble reaction zone. For the mobile reaction zone both cases $D_C>0$ and $D_C=0$ belong to the same class.

(3) Asymptotic analysis of the equations in the mean-field approximation inside the reaction zone in the reversible regime confirms the results of the general consideration. For the mobile reaction zone the component and reaction rate profiles are qualitatively similar to those for the $D_C>0$ case. The reaction rate profile has a characteristic single maximum near the reaction zone center. For the asymptotically immobile zone a distinctive two-hump profile of $R_r(x,t)$ is obtained. This profile is associated with the influence of C on the reaction rate $R_r(x,t)$ in the reversible regime.

(4) It is shown that in the reversible regime, for the case of a mobile reaction zone, a second distinct reaction zone is formed near the point $x=0$. In this reaction zone through the exchange of C and B effective diffusion of the nonmobile C component occurs. This reaction zone structure is related to that of a first order reversible reaction.

(5) Numerical simulation of the complete system of mean-field equations confirms the results of the asymptotic analysis. At the same time, deviations from the symmetrical limiting solutions are observed. For the case of the immobile reaction zone, the deviations are related to the effect of the short-time asymmetric distribution of C (for $D_A\neq D_B$) on $R_r(x,t)$.

(6) Similar to the case $D_C>0$, the case $D_C=0$ is also characterized by conservation of the macroscopic properties through the crossover from the irreversible to the reversible regime. This occurs despite the fact that the cases $D_C=0$ and $D_C>0$ for the immobile reaction zone belong to different classes.

APPENDIX: CALCULATION OF THE REACTION ZONE STRUCTURE NEAR $x=0$ FOR $C_f>0$ IN THE REVERSIBLE REGIME

For the long-time regime $t\to\infty$, the concentration of A in the region $x\ll x_f-w$ considerably exceeds the concentration of B. Consequently, the influence of the reaction $A+B\leftrightarrow C$ on the A concentration in this region may be neglected and the A profile is described by Eq. (10). Near the point $x=0$, the A concentration is approximately constant and equals a_0-C_A. With this conjecture Eqs. (20) are reduced to a first order reversible reaction-diffusion problem, which is described by the following equations:

$$k\rho_A\rho_B=g\rho_C,$$

$$\rho_A=a_0-C_A, \tag{A1}$$

$$\frac{\partial\rho_C}{\partial t}+\frac{\partial\rho_B}{\partial t}=D_B\frac{\partial^2\rho_B}{x^2}.$$

The boundary conditions are

$$\rho_B\to 0,\quad \rho_C\to 0\quad \text{for } x\to-\infty, \tag{A2a}$$

$$\rho_B\to gc_0/k(a_0-C_A),\quad \rho_C\to c_0\quad \text{for } x\to+\infty. \tag{A2b}$$

Equations (A1) are equivalent to the standard diffusion equation with an effective diffusivity $D_{\mathrm{eff}}=D_B/[1+k(a_0-C_A)/g]\approx D_Bg/k(a_0-C_A)$. Its solution with the boundary conditions of Eq. (A2) is given by

$$\rho_B(x,t)=[gc_0/(a_0-C_A)]0.5[1+\mathrm{erf}(x/w_3)], \tag{A3}$$

$$\rho_C(x,t)=c_0 0.5[1+\mathrm{erf}(x/w_3)], \tag{A4}$$

where $w_3=2\sqrt{D_{\mathrm{eff}}t}=2\sqrt{(gD_Bt)/[k(a_0-C_A)]}$. The reaction rate is computed from

$$R_r(x,t)=\partial\rho_C/\partial t=-c_0(1/t)(1/2\sqrt{\pi})(x/w_3)\exp(-x^2/w_3{}^2). \tag{A5}$$

It should be noted that $R_r(x,t)<0$ for $x>0$ and $R_r(x,t)>0$ for $x<0$. This may be interpreted as the C component disappearing in the region $x>0$, producing B. This B crosses to the region $x<0$, where it undergoes an inverse transition to C.

Expressions (A3)–(A5) can also be obtained in the framework of the extended scaling approach described in Sec. II. The magnitudes of the scaling exponents are $\beta=\frac{1}{2}$, $\phi_A=\phi_C=0$, $\phi_B=1$, $\phi_{R_r}=-\frac{1}{2}$.

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

[2] Y. E. Koo, L. Li, and R. Kopelman, Mol. Cryst. Liq. Cryst. **183**, 187 (1990).

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

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

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

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

[7] B. Chopard and M. Droz, Europhys. Lett. **15**, 459 (1991).

[8] S. Cornell, M. Droz, and B. Chopard, Phys. Rev. A **44**, 4826 (1991).

[9] S. Cornell, M. Droz, and B. Chopard, Physica A **188**, 322 (1992).

[10] M. Araujo, S. Havlin, H. Larralde, and H. E. Stanley, Phys. Rev. Lett. **68**, 1791 (1992).

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

[12] H. Larralde, M. Araujo, S. Havlin, and H. E. Stanley, Phys. Rev. A **46**, 855 (1992); **46**, R6121 (1992).

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

[14] S. Cornell and M. Droz, Phys. Rev. Lett. **70**, 3824 (1993); S. Cornell, *ibid.* **75**, 2250 (1995).

[15] M. Araujo, H. Lerralde, S. Havlin, and H. E. Stanley, Phys. Rev. Lett. **71**, 3592 (1993); **75**, 2251 (1995).

[16] H. Taitelbaum, Physica A **200**, 155 (1993).

[17] Y. E. Koo, R. Kopelman, A. Yen, and A. Lin, in *Dynamics in Small Confining Systems*, edited by J. M. Drake *et al.*, Mater. Res. Soc. Symp. Proc. No. **290** (Materials Research Society, Pittsburgh, 1993), p. 273.

[18] B. P. Lee and J. Cardy, Phys. Rev. E **50**, R3287 (1994); J. Stat. Phys. **80**, 971 (1995).

[19] M. Howard and J. Cardy, J. Phys. A **28**, 3599 (1995).

[20] S. Cornell, Phys. Rev. E **51**, 4055 (1995).

[21] S. Cornell, Z. Koza, and M. Droz, Phys. Rev. E **52**, 3500 (1995).

[22] S. Havlin, M. Araujo, Y. Lereach, H. Larralde, A. Shehter, H. E. Stanley, P. Trunfio, and B. Vilensky, Physica A **221**, 1 (1995).

[23] P. L. Krapivsky, Phys. Rev. E **51**, 4774 (1995).

[24] G. T. Barkema, M. Howard, and J. Cardy, Phys. Rev. E **53**, R2017 (1996).

[25] Z. Koza, J. Stat. Phys. **85**, 179 (1996); Philos. Mag. B **77**, 1437 (1998).

[26] Z. Koza, Physica A **240**, 622 (1997).

[27] A. Yen, Y. E. Koo, and R. Kopelman, Phys. Rev. E **54**, 2447 (1996).

[28] H. Taitelbaum, R. Kopelman, A. Yen, S. Havlin, and G. H. Weiss, Phys. Rev. E **54**, 5942 (1996).

[29] H. Taitelbaum, B. Vilensky, A. Lin, A. Yen, Y. E. Koo, and R. Kopelman, Phys. Rev. Lett. **77**, 1640 (1996).

[30] A. Yen, A. Lin, Y. E. Koo, B. Vilensky, H. Taitelbaum, and R. Kopelman, J. Phys. Chem. A **101**, 2819 (1997).

[31] A. Yen and R. Kopelman, Phys. Rev. E **56**, 3694 (1997).

[32] A. Yen, Z. Y. Shi, and R. Kopelman, Phys. Rev. E **57**, 2438 (1998).

[33] V. Malyutin, S. Rabinovich, and S. Havlin, Phys. Rev. E **56**, 708 (1997).

[34] M. Rodrigez and H. S. Wio, Phys. Rev. E **56**, 1724 (1997).

[35] Z. Koza and H. Taitelbaum, Phys. Rev. E **54**, R1040 (1996).

[36] H. Taitelbaum and Z. Koza, Philos. Mag. B **77**, 1389 (1998).

[37] Z. Koza and H. Taitelbaum, Phys. Rev. E **56**, 6387 (1997).

[38] S. Cornell and M. Droz, Physica D **103**, 348 (1997).

[39] B. Chopard, M. Droz, J. Magnin, and Z. Rácz, Phys. Rev. E **56**, 5343 (1997).

[40] M. Araujio, Physica A **219**, 239 (1995).

[41] M. Sinder and J. Pelleg, Phys. Rev. E **60**, R6259 (1999).

[42] M. Sinder and J. Pelleg, Phys. Rev. E **61**, 4935 (2000). The relation $D_A\rho_A + D_C\rho_C \sim \mathrm{const} + [x - x_f(t)]/\sqrt{t}$ was obtained inside the reaction zone for the long-time limit $t \rightarrow \infty$. Taking into account Eq. (1) and the above relation the following expression may immediately be written: $\rho_C \sim \mathrm{const}_1 + \mathrm{const}_2 \times t^{-2/3} S_A[(x - x_f(t)/w(t)] + \mathrm{const}_3 \times [x - x_f(t)]/\sqrt{t}$. Reserving only the largest term, $\rho_C \sim \mathrm{const}_1$ is obtained, i.e., $\gamma_C = 0$.

[43] J. S. Langer, Rev. Mod. Phys. **52**, 1 (1980).

[44] G. T. Dee, Phys. Rev. Lett. **57**, 275 (1986).

[45] F. J. Muzzio and J. M. Ottino, Phys. Rev. A **40**, 7182 (1989).

[46] S. Havlin and D. ben-Avraham, Adv. Phys. **36**, 695 (1987).