Asymptotic expansion for reversible A + B ⇌ C reaction-diffusion process

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I. INTRODUCTION

Behavior of many physical, biological, and chemical systems is determined by evolution of a reaction front formed between initially separated reactants [1, 2, 3]. The simplest theoretical model of this phenomenon consists in assuming that initially two species A and B are uniformly distributed on opposite sides of an impenetrable barrier. The barrier is removed at time $t = 0$ and the two species start to mix and react, forming a dynamic reaction front. It is assumed that diffusion is the only transport mechanism and that the diffusion coefficients of each species are strictly constant, i.e., independent of spatial location, reactant concentrations, etc. The problem to be solved is to give a detailed description of spatiotemporal evolution of this reaction-diffusion system.

Simple as it is, this system exhibits many unexpected features, especially when the reaction is irreversible ($A + B \rightarrow C$). Using a scaling ansatz, Galfi and Rácz [3] showed that in this case the width of the reaction front grows asymptotically as $t^\alpha$ with surprisingly small value of the exponent $\alpha = 1/6$. Renormalization group technique [4, 5], dimensional analysis [6, 7], and extensive computer simulations [8] were then employed to demonstrate that $\alpha = 1/6$ only above the critical dimension $d_c = 2$ and for $d \leq d_c$ one must take into account fluctuations of reactant concentrations. It was also shown [9, 10] that the case where one of the reactants is immobile ($D_A = 0$ or $D_B = 0$) belongs to a separate universality class, with $\alpha = 1/2$. Short-time perturbation [10] and numerical [11] analysis revealed that the reaction front can move in a nonmonotonic manner. The quasistatic approximation [12, 13] was used to find a detailed description of concentrations of species A, B, and C outside the reaction zone [14, 15, 16, 17, 18] and the case of equal diffusion coefficient was treated rigorously [19]. These theoretical results are in full agreement with experiments [16, 17, 18, 19, 20] and were generalized to several more complex reactions [21, 22, 23, 24, 25, 26, 27].

In reality, however, most of chemical reactions are reversible. In spite of this, reversible $A + B \rightleftharpoons C$ reaction-diffusion processes did not attract so much attention. This should be probably attributed to the fact that reversible reaction-diffusion processes for a long time were not supposed to exhibit any “anomalous” properties, especially after Chopard et al. [28] showed that (a) the front width of a reversible reaction asymptotically scales with time as if the process was governed only by diffusion ($w(t) \propto t^{1/2}$) and (b) the fluctuations do not modify the scaling exponents even in one-dimensional systems. However, the problem of giving a detailed description of spatiotemporal evolution of reversible reaction-diffusion systems remained open.

This issue was recently considered by Sinder and Pelleg in a series of papers [29, 30, 31]. They restricted their analysis mainly to systems with very small backward reaction rate $g$ and found that in this case concentrations of species A, B, and C are practically the same as those observed in strictly irreversible reactions ($g = 0$) everywhere except in a very narrow reaction zone. They confirmed the result of Ref. [28] that there is a crossover between intermediate-time “irreversible” and long-time “reversible” regimes. They also showed that in contrast to irreversible systems, the effective asymptotic reaction rate $R$ can have two maxima and there can be even a region where it is negative. Moreover, they presented strong arguments in support of a conjecture that reversible reaction-diffusion processes belong to two distinct universality classes. One of them consists of systems with immobile reaction product C and asymptotically immobile reaction front, while all other systems belong the other universality class.

The aim of our paper is to work out a new tool for analysis of arbitrary reversible reaction-diffusion systems—expansion of concentrations as series in $1/t$ (where $t$ denotes time)—and to use it to find exact solutions of the problem at least for some particular combinations of control parameters. This kind of approach was already successfully applied to reaction fronts at very short times [32]. To our knowledge, however, such a technique has not been applied to the long-time regime.

The structure of the paper is as follows. In section II
we give the precise definition of the problem to be solved. In section III A we show how to apply the perturbation expansion to reversible reaction-diffusion systems. In section III C we employ this technique to study the case where diffusion coefficients of all species are the same. In particular, we present (a) explicit forms of the long-time reaction rate $R$ and the concentrations of species $A$, $B$, and $C$; (b) a recursive formula for any term of the expansions; (c) detailed analysis of the crossover to the long-time, reversible regime. In section III B we analyze two particular cases where some aspects of the system behaviour can be studied analytically. Finally, section IV is devoted to discussion of results.

II. MODEL

We shall consider the following system of nonlinear partial differential equations

$$\begin{align*}
\frac{\partial a(x,t)}{\partial t} &= D_A \frac{\partial^2 a(x,t)}{\partial x^2} - R(x,t) \quad (1) \\
\frac{\partial b(x,t)}{\partial t} &= D_B \frac{\partial^2 b(x,t)}{\partial x^2} - R(x,t) \quad (2) \\
\frac{\partial c(x,t)}{\partial t} &= D_C \frac{\partial^2 c(x,t)}{\partial x^2} + R(x,t) \quad (3)
\end{align*}$$

where

$$R(x,t) \equiv ka(x,t)b(x,t) - gc(x,t). \quad (4)$$

Here $a(x,t)$, $b(x,t)$, and $c(x,t)$ are concentrations of species $A$, $B$, and $C$, respectively, $D_A$, $D_B$, and $D_C$ are their respective diffusion coefficients, $R(x,t)$ denotes the effective reaction rate, while $k > 0$ and $g \geq 0$ are the forward and backward reaction rate constants, respectively. The initial conditions to (1) - (3) read

$$a(x,0) = a_0 H(x), \quad b(x,0) = b_0 H(-x), \quad c(x,0) = 0, \quad (5)$$

where $a_0$, $b_0$ are initial concentrations of species $A$ and $B$, respectively, and $H(x)$ is the Heaviside step function, which is 0 for $x < 0$ and 1 for $x > 0$. Note that in Refs [14, 28, 29] $R$ was called a “refined reaction rate” and denoted as $R_f$. Our main goal is to give a detailed description of the long-time solutions of (1) - (3).

As was pointed out in Ref. [28], by measuring length, time, and concentration in units of $\sqrt{D_A/a_0}$, $1/a_0$, and $a_0$, respectively, our original problem can be reduced to the one with

$$D_A = 1, \quad a_0 = 1, \quad k = 1. \quad (6)$$

We shall adopt these particular values in our further analysis, which will leave us with four independent control parameters: $g$, $b_0$, $D_B$, and $D_C$.

III. THE ASYMPTOTIC EXPANSION

A. Formalism

The analysis performed in Refs. [14, 28, 29] shows that in the asymptotic, long-time limit solutions to equations (1) - (3) take on a scaling form

$$\begin{align*}
 a(x,t) &\approx S_A(x/\sqrt{t}), \quad (7) \\
b(x,t) &\approx S_B(x/\sqrt{t}), \quad (8) \\
c(x,t) &\approx S_C(x/\sqrt{t}), \quad (9) \\
R(x,t) &\approx t^{-1} S_R(x/\sqrt{t}). \quad (10)
\end{align*}$$

We therefore assume that in the long-time limit all functions involved in (1) - (3) can be expanded as series in $\tau \equiv 1/t$, with coefficients being some functions of $\xi \equiv x/\sqrt{t}$. We thus write $a(x,t)$, $b(x,t)$, and $c(x,t)$ as

$$\begin{align*}
a(x,t) &= \sum_{n=0}^{\infty} \tau^n A_n(\xi), \quad (11) \\
b(x,t) &= \sum_{n=0}^{\infty} \tau^n B_n(\xi), \quad (12) \\
c(x,t) &= \sum_{n=0}^{\infty} \tau^n C_n(\xi). \quad (13)
\end{align*}$$

Using (10) we conclude that $R(x,t)$ can be expressed as

$$R(x,t) = \sum_{n=0}^{\infty} \tau^n R_n(\xi), \quad (14)$$

where

$$R_n(\xi) \equiv -gC_n(\xi) + \sum_{j=0}^{n} A_j(\xi)B_{n-j}(\xi). \quad (15)$$

Substituting (11) - (13) into (1) - (3) and collecting coefficients at $\tau^0$ we find that

$$R_0(\xi) \equiv A_0(\xi)B_0(\xi) - gC_0(\xi) = 0. \quad (16)$$

This expression expresses a fundamental property of the system: in the long-time limit it tends to a local chemical equilibrium (the forward and backward reaction rates become asymptotically equal) [28]. Collecting now coefficients at $\tau^{n+1}$, $n = 0, 1, \ldots$, we arrive at

$$\begin{align*}
 A_0''(\xi) + \frac{1}{2} \xi A_0'(\xi) + nA_n - R_{n+1}(\xi) &= 0 \quad (17) \\
 DB_n''(\xi) + \frac{1}{2} \xi B_n'(\xi) + nB_n - R_{n+1}(\xi) &= 0 \quad (18) \\
 DC_n''(\xi) + \frac{1}{2} \xi C_n'(\xi) + nC_n + R_{n+1}(\xi) &= 0 \quad (19)
\end{align*}$$
where we used a short-hand notation $f'(\xi) \equiv df/d\xi$, $f''(\xi) \equiv d^2f/d\xi^2$. In the lowest order ($n = 0$) we thus find

$$A_0''(\xi) + \frac{1}{2}\xi A_0'(\xi) = \mathcal{R}_1(\xi)$$

(20)

$$B_0''(\xi) + \frac{1}{2}\xi B_0'(\xi) = \mathcal{R}_1(\xi)$$

(21)

$$C_0''(\xi) + \frac{1}{2}\xi C_0'(\xi) = -\mathcal{R}_1(\xi).$$

(22)

Equations (16), (20) – (22) constitute a system of four equations for four unknown functions: $A_0(\xi)$, $B_0(\xi)$, $C_0(\xi)$, and $\mathcal{R}_1(\xi)$. These important functions control the asymptotic (long-time) properties of the system and can be readily identified with the scaling functions employed in relations (1) – (3). However, owing to a nonlinear form of the equation (16), the explicit form of these functions can be found only in a few particular cases discussed below.

B. The case of equal diffusion constants

1. Asymptotic solution

Following (1) we will now assume $D_A = D_B = D_C = 1$, $k = 1$ and $a_0 = 1$. The only free parameters of the problem are thus $b_0 = b$ and $g$. Upon adding (1) to (1) and (2) we arrive at two diffusion equations with well known solutions

$$a(x,t) + c(x,t) = \frac{1}{2}\text{erfc}(x/\sqrt{4t})$$

(23)

$$b(x,t) + c(x,t) = \frac{1}{2}b_0\text{erfc}(-x/\sqrt{4t}).$$

(24)

This immediately leads to

$$A_0(\xi) = \frac{\Phi(\xi) - g + \sqrt{\Delta(\xi)}}{2}$$

(25)

$$B_0(\xi) = \frac{-\Phi(\xi) - g + \sqrt{\Delta(\xi)}}{2}$$

(26)

$$C_0(\xi) = \frac{\text{erfc}(\xi/2) + b_0\text{erfc}(-\xi/2) + 2g - 2\sqrt{\Delta(\xi)}}{4}$$

(27)

$$\mathcal{R}_1(\xi) = \frac{g b_0(1 + g + b_0)\exp(-\xi^2/4)}{2\pi(\Delta(\xi))^{3/2}}$$

(28)

$$A_n(\xi) = B_n(\xi) = -C_n(\xi), \quad n \geq 1$$

(29)

where we denoted

$$\Phi(\xi) \equiv \frac{1}{2}a_0\text{erfc}(\xi/2) - b_0\text{erfc}(-\xi/2)$$

(30)

$$\Delta(\xi) \equiv [\Phi(\xi) - g]^2 + 2g\text{erfc}(\xi/2).$$

(31)

Note that $\mathcal{R}_1(\xi)$ can diverge to infinity. This can happen if and only if $\Delta(\xi) \to 0$ which, in turn, occurs only if $\Phi(\xi) \to 0$ and $g \to 0$. Since $\Phi(\xi)$ decreases monotonically from $a_0$ to $-b_0$, equation $\Phi(\xi) = 0$ has a unique solution, which will be denoted $\xi_f$.

In the limit of an irreversible reaction we find

$$\lim_{g \to 0} A_0(\xi) = \begin{cases} \Phi(\xi), & \xi < \xi_f \\ 0, & \xi \geq \xi_f \end{cases}$$

(32)

$$\lim_{g \to 0} B_0(\xi) = \begin{cases} 0, & \xi < \xi_f \\ -\Phi(\xi), & \xi \geq \xi_f \end{cases}$$

(33)

$$\lim_{g \to 0} C_0(\xi) = \begin{cases} \frac{b_0}{\sqrt{2}}\text{erfc}(\xi/2), & \xi < \xi_f \\ \frac{\xi}{2}\text{erfc}(\xi/2), & \xi \geq \xi_f \end{cases}$$

(34)

$$\lim_{g \to 0} \mathcal{R}_1(\xi) = \frac{b_0\exp(-\xi_f^2/4)}{\sqrt{\pi}\text{erfc}(\xi_f/2)}\delta(\xi - \xi_f)$$

(35)

where $\delta$ is the Dirac’s delta distribution. These relations are in full agreement with general formulas derived in Ref. [4] for the case of a strictly irreversible reaction, $g = 0$, and in Ref. [8] for $a_0 = b_0 = 0$ and $g \to 0$. Both species, A and B, become effectively segregated at $\xi_f$. We can thus identify $\xi_f$ as the position of the reaction front. Beyond this point the reaction rate tends to 0 and the concentrations $a(x,t)$, $b(x,t)$, and $c(x,t)$ satisfy diffusion equations (1) – (3) with $R(x,t) = 0$. However, the existence of a singularity in the above formulas does not mean that the reaction should be asymptotically restricted to a single point! Recalling that $\xi \equiv x/\sqrt{t}$ we conclude that this singularity indicates that the reaction must be restricted to a region much narrower than $\sqrt{t}$; it also suggests that for $g = 0$ and $x/\sqrt{t} \approx \xi_f$ we should try and take into account higher-order terms of expansions (1) – (3).

In the opposite limit of an infinitely large backward reaction rate $g$ we find

$$\lim_{g \to \infty} A_0(\xi) = \frac{1}{2}\text{erfc}(\xi/2)$$

(36)

$$\lim_{g \to \infty} B_0(\xi) = \frac{1}{2}b_0\text{erfc}(-\xi/2)$$

(37)

$$\lim_{g \to \infty} C_0(\xi) = \lim_{g \to \infty} \mathcal{R}_1(\xi) = 0.$$  

(38)

These equations express the fact that for large backward reaction rates $g$ if a particle C is being created as a result of a forward, A + B → C reaction, it is being immediately converted back into a pair A-B; consequently, the concentration of particles C tends to 0 and the concentrations of particles A and B evolve as if there was no reaction at all.

2. Recursive formula

To find the remaining terms of expansions (11) – (13) we employ relations (14), (15), (23), and (24), arriving at a recursive formula

$$C_{n+1}(\xi) = \frac{nC_n(\xi) + \frac{1}{2}\xi C'_n(\xi) + C''_n(\xi) + S_n(\xi)}{\sqrt{\Delta(\xi)}},$$

(39)
where \( S_n(\xi) \equiv \sum_{j=1}^n C_j(\xi) C_{n+1-j}(\xi) \) and \( n \geq 0 \). An important feature of this relation is that it allows to express the \((n+1)\)-th term of the expansion directly as a function of already determined, lower-order terms. Consequently, together with the explicit form of the zeroth order terms given in (27)–(28), equations (29) and (39) enable one to calculate (at least in principle) an arbitrary term of the expansions (11)–(13) analytically. In practice, however, the complexity of the appropriate formulas grows rapidly and even using computer-algebra systems it is very difficult to determine \( C_n(\xi) \) for more than a few smallest values of \( n \). Notice also that for \( \xi = \xi_f \) and \( g \to 0 \) the denominator of (39) goes to zero.

### 3. Crossover from “irreversible” to “reversible” reaction fronts

Let \( t^* \) denote the time when the system enters the asymptotic, long-time time regime. For times \( t \gg t^* \) each sum in expansions (11)–(13) should be dominated by its lowest-order nonvanishing term, while for \( t \lesssim t^* \) the system should behave as if the reaction was strictly irreversible \((g = 0)\). This cross-over time can be estimated from a relation \( A_0(\xi_f) \approx A_1(\xi_f)/t^* \). Using (22), (24), and (39) we find \( A_1(\xi) = -C_1(\xi) = R_1(\xi)/\sqrt{\Delta(\xi)} \).

In the limit \( g \to 0 \) there is thus \( A_0(\xi_f) \propto \sqrt{g} \) and \( A_1(\xi_f) \propto 1/g \). Therefore in this limit we have

\[
t^* \propto g^{-1/2}. \tag{40}
\]

Note that Chopard et al. [28] proposed a different relation, \( t^* \propto g^{-1} \). However, their conjecture was based on numerical analysis of a reaction-diffusion system with a very specific choice of system parameters: \( a_0 = b_0 \), \( D_A = D_B \), and \( D_C = 0 \). In other words, they investigated only symmetric systems with immobile particles C. Moreover, they assumed that the width of the reaction front can be identified as the width of the profile of particles C, \( w_C(t) = \int x^2 c(x, t) dx/\int c(x, t) dx \). Actually this is acceptable only when both particles C and the reaction front center are immobile \((D_C = 0, x_f(t) \sim 0)\), a condition implicitly satisfied in their simulations. For \( D_C \neq 0 \) or \( x_f(t) \neq 0 \) we expect that asymptotically \( w_C(t) \propto t^{1/2} \), while \( w(t) \propto t^{1/6} \) (at least for \( g = 0 \)), so \( w(t) \) cannot be identified with \( w(t) \). From this point of view the case studied numerically in Ref. [28] belongs to a separate universality class. To the same conclusion, though on different grounds, came Sinder and Pelleg [24], who investigated systems with immobile reaction product \((D_C = 0)\). They found that asymptotically \( w(t, g) \propto g^{1/2} t^{1/2} \) for systems with the moving reaction front \((x_f \neq 0)\) and \( w(t, g) \propto g^{1/3} t^{1/3} \) if \( x_f \sim 0 \). Therefore, on taking into account that for small \( g \) and intermediate times \( t \) one expects \( w(t, g) \propto g^{1/2} t^{1/2} \), we immediately arrive at the conclusion that for asymptotically immobile reaction fronts studied in Ref. [28] there should be \( t^* \propto g^{-1} \), while for systems with \( D_C = 0 \), \( g \ll 1 \), and a mobile reaction front \((x_f \neq 0)\), the crossover time is given by (40).

We verified the validity of relation (40) for systems studied in this section by computer-assisted analysis of \( A_0(\xi_f) \) for the fully symmetric case \( a_0 = b_0 = 1 \). It indicates that

\[
A_0(\xi_f) \propto (-1)^n g^{(1 - 3n)/2} \tag{41}
\]

for all \( n \leq 5 \). Most probably this relation continues to be true also for higher-order terms. This would mean that for \( t \gg t^* \), as expected, expansions (11)–(13) will be dominated by their first terms, while for \( t \ll t^* \) they are divergent at \( \xi_f \).

Using (40) we can estimate the width \( w \) and height \( h \) of the reaction front at the cross-over time. The former is defined as a square root of \( \int_{-\infty}^{\infty} (x - x_f)^2 R(x, t) dt/\int_{-\infty}^{\infty} R(x, t) dt \), while \( h = R(x_f, t) \) (here \( x_f \approx \sqrt{g} \xi_f \) is the exact location of the reaction front, see Ref. [3, 13]). Using (28) and (39) we find

\[
w(t^*) \propto \sqrt{g} t^*/(t^*)^{1/6}, \tag{42}
\]

\[
h(t^*) \propto (t^*)^{-1/2} \sqrt{\frac{g}{t^*}} \propto (t^*)^{-2/3}. \tag{43}
\]

These are the scaling relations derived by Galfi and Racz for the case of a strictly irreversible reaction, \( g = 0 \) and \( t \to \infty \).

### C. Other cases with rigorous solutions

#### 1. The limit \(|\xi| \to \infty\), or the tails of the distributions

Let us now consider the general case of arbitrary values of parameters \( b_0, D_B, D_C \) and \( g \) in the limit of \( t \to \infty \) and \( |\xi| \to \infty \) such that \( |\xi|/\sqrt{t} = |\xi| \to \infty \). For sufficiently large \( \xi \) we may expect that the concentration of particles B will be very close to its original value \( b_0 \). Substituting \( B_0(\xi) \approx b_0 \) in (20)–(22) we find that \( R_3(\xi) \approx 0 \) and

\[
C_0(\xi) \approx b_0 g^{-1} A_0(\xi) \approx \eta^+ \text{erfc}\left(\xi/\sqrt{4D_\text{eff}^+}\right) \tag{44}
\]

where

\[
D_\text{eff}^+ = \frac{g D_A + b_0 D_C}{g + b_0} \tag{45}
\]

and \( \eta^+ \) is an integration constant independent of \( \xi \). Note that, as might be expected on physical grounds, \( D_\text{eff}^+ \) lies between \( D_A \) and \( D_C \).

Similarly, in the opposite limit \( \xi \to -\infty \), we assume \( A_0(\xi) \approx a_0 = 1 \) and find

\[
C_0(\xi) \approx g^{-1} B_0(\xi) \approx \eta^- \text{erfc}\left(-\xi/\sqrt{4D_\text{eff}^-}\right) \tag{46}
\]

where

\[
D_\text{eff}^- = \frac{g D_B + D_C}{g + 1} \tag{47}
\]
and $\eta^-$ does not depend on $\xi$. We thus see that sufficiently far away from the reaction region all the species diffuse with an effective diffusion coefficient $D_{\text{eff}}^+(\xi)$ (for $\xi \to -\infty$) or $D_{\text{eff}}^-(\xi)$ (for $\xi \to \infty$). Note that $D_{\text{eff}}^+, D_{\text{eff}}^- \to D_0$ as $g \to 0$, in agreement with the findings of Ref. [14].

2. Reaction front at $\xi = 0$ for one or two vanishing diffusion constants

Equations (21) – (22) immediately imply that if $D_B$ or $D_A$ vanishes then

$$R_1(0) = 0.$$ (48)

Thus either $R_1(\xi)$ has a local minimum at $\xi = 0$ or it attains negative values in the vicinity of $\xi = 0$. Both possibilities have actually been observed in numerical simulations carried out in Ref. [29].

IV. CONCLUSIONS

We have applied perturbation analysis to reversible reaction-diffusion systems with arbitrary values of control parameters. Using this technique we obtained a system of four equations completely governing the long-time behaviour of the system. We then concentrated on several cases where analytical results can be derived.

In particular, we found the complete solution of the problem for the case where the diffusion coefficients of all species are the same. Its most interesting feature is the limit of vanishingly small backward reaction rate constant $g$. In this limit, as expected, the solutions become singular at a point which can be identified with the reaction-front center. Our method enables one to analyze explicitly how these singularities, especially the Dirac’s delta function in the expression for the local reaction rate $R_1$, appear in mathematical formulas. This holds out hope that it will be possible to construct a unified theory of reversible and irreversible reaction-diffusion systems. It should be also noticed that our asymptotic solution is also valid for $g = 0$ even though in this case all higher-order terms of the expansion diverge.

We also found a general recursive formula for any term of the expansion as a function of all lower-order terms. Unfortunately, complexity of expressions thus obtained grows very rapidly. Nevertheless we believe that this formula opens a new way of analyzing the evolution of reaction-diffusion systems at arbitrary times.

Using the information about the first correction to the asymptotic solution we showed that the anomalous properties of irreversible reaction-diffusion systems can be studied by taking a suitable limit in the formulas obtained for reversible systems. A potential advantage of this approach is that mathematical description of reversible reaction-diffusion systems is more regular, and hence more amenable to rigorous analysis.

We also studied the crossover time $t^*$ between intermediate-time (“irreversible reaction”) and long-time (“reversible reaction”) regimes. We proved that in the case of equal diffusion coefficients $t^*$ scales with $g^{-3/2}$. This conclusion agrees with results obtained by Sinder and Pelleg [21] and disagrees with those obtained by Cornell and Droz [3]. This discrepancy can be easily understood if one notices that Cornell and Droz studied only systems where the width $w_c$ of the concentration of species C grows in time as $t^{1/6}$, while our study was performed for a system where $w_c \propto t^{1/2}$.

The main message coming from our work may be summarized as follows: (a) reversible reaction-diffusion systems are more amenable to rigorous treatment than irreversible ones; (b) it is possible to investigate the more difficult irreversible reaction-diffusion systems by taking an appropriate limit in the reversible ones; (c) investigation of reversible reaction-diffusion systems is interesting not only per se, but constitutes an alternative technique of analyzing irreversible systems.

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