

Front propagation in $A+B\rightarrow 2A$ reaction under subdiffusion

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We consider an irreversible autocatalytic conversion reaction $A+B\rightarrow 2A$ under subdiffusion described by continuous-time random walks. The reactants' transformations take place independently of their motion and are described by constant rates. The analog of this reaction in the case of normal diffusion is described by the Fisher-Kolmogorov-Petrovskii-Piskunov equation leading to the existence of a nonzero minimal front propagation velocity, which is really attained by the front in its stable motion. We show that for subdiffusion, this minimal propagation velocity is zero, which suggests propagation failure.

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I. INTRODUCTION

The theory of reactions under subdiffusion recently attracted considerable attention both because of theoretical and mathematical challenges posed by such problems, and also due to their growing practical relevance for description of phenomena taking place in porous media (as exemplified by geophysical structures) and in the crowded interior of living cells (e.g., Refs. [1–3]). Several recent works were dedicated to the theoretical description of Turing patterns and of fronts in such systems for different reaction schemes and different conditions [4–8]. Thus Ref. [6] concentrates on the front behavior in the system that in the case of normal diffusion would be described by the Fisher-Kolmogorov-Petrovskii-Piskunov (FKPP) equation, and shows that under the description adopted, there exists a (minimal) stable propagation velocity of such a front, just as it is under normal diffusion. As we proceed to show, this is not always the case. Considering the fully irreversible analog of the reaction discussed in Ref. [6] under conserved overall concentration, we show that the minimal propagation velocity is zero, which corresponds essentially to propagation failure. The system we consider here corresponds to the $A+B\rightarrow 2A$ irreversible reaction, which in the case of normal diffusion is also described by the FKPP equation.

The FKPP equation [9], proposed by Fisher in 1937 [10] as a model for propagation of a favorable gene in a population, corresponds to a mathematical description of the (irreversible or reversible) reaction whose main stage is a bimolecular autocatalytic conversion $A+B\rightarrow 2A$. Initially, the whole system consists of particles (individuals) of type B. The introduction of the A-individuals into some bounded spacial domain (which is described by an initial condition sharply concentrated in vicinity of the origin of coordinates) leads to a propagation of a front of A into the B-domain. Physically this corresponds to a front propagating into the unstable state. Under normal diffusion, the process is described by a partial differential equation

$$\frac{\partial A}{\partial t} = D\Delta A + kAB$$

for an irreversible reaction, where the initial concentration of B is assumed to be homogeneous and equal to $B(x,0)=B_0$ everywhere except for the vicinity of the origin. In this case, the overall concentration is conserved due to the local stoichiometry of the reaction, which does not change the number of particles. Here and in what follows, we denote the nature of particles in reaction equations by roman letters, while the corresponding concentrations or particle numbers are written in *italic*.

Using the conservation law, the corresponding reaction-diffusion equation can be rewritten as

$$\frac{\partial A}{\partial t} = D\Delta A + kAB_0 - kA^2. \quad (1)$$

The equation for the reversible reaction has the same form but different coefficients in front of the two last terms on the right-hand side (e.g., [11]). The FKPP equation is the simplest model of front propagation into an unstable state and serves as a paradigmatic model for many related phenomena.

The front velocity in the genuine FKPP equation is determined by that of its leading edge, i.e., by the behavior of concentrations for $x\rightarrow\infty$. Since the concentration of converted particles in this leading edge is very small, the equation can be linearized, and the possible velocity of the front is given by the analysis of the *linear* propagation problem [12,13]. For the FKPP equation, Eq. (1) leads for $x\rightarrow\infty$ (and $B\rightarrow B_0$) to

$$\frac{\partial A}{\partial t} = D\Delta A + kB_0A \quad (2)$$

(or by the corresponding equation for $\delta B=B_0-B=A$) having the exponential propagating solution $A\approx\exp(-x+vt)$ for all $v\geq 2\sqrt{DkB_0}$ where the condition on v is imposed by the natural boundary condition $A=0$ for $x\rightarrow\infty$ and by forbidding oscillatory behavior. Further analysis shows that the minimal velocity $v=2\sqrt{DkB_0}$ is exactly the one attained, which fact is

known as the marginal stability principle. The front in the FKPP system is an example of the so-called ‘‘pulled front,’’ as it is ‘‘pulled’’ into the unstable state by its leading edge, and its propagation velocity does not depend on what happens in the interior of the front where the conversion of the most particles takes place.

Modeling reaction phenomena in subdiffusive media requires some preliminary assumptions on the nature of the transport process. Parallel to Refs. [14,15], we assume that the subdiffusive motion on a mesoscopic scale is a consequence of trapping of particles on a mesoscopic scale, due to, e.g., the bottlenecks connecting the voids in a porous system, while on a microscopic scale within the pores the $A+B \rightarrow 2A$ reaction takes place in a homogeneous solution and follows the mass action law. The local conservation of particle concentration is an inherent property of such systems, which also holds on the mesoscopic scale. The model adopted corresponds, therefore, to the systems that at smaller scales consist of compartments in which the reaction follows the usual kinetic laws, while the subdiffusive transport between the compartments is described by continuous-time random walks with a probability density function (pdf) of sojourn times that may lack the first moment. As we proceed to show, due to the coupling between the reaction and transport term inherent for subdiffusion [14,15], the behavior of the reaction front under subdiffusion and under the conditions discussed above is vastly different from that under normal diffusion. The minimal propagation velocity of the front is zero, which corresponds to propagation failure. The preliminary results of our numerical simulations confirm that the front’s velocity decays with time. These numerical results will be discussed in detail elsewhere.

II. THE $A+B \rightarrow 2A$ REACTION UNDER SUBDIFFUSION

A. General considerations

Following the same procedure as in [8,14], one obtains the equations for the particle concentrations as a consequence of the conservation laws. Discretizing the system into compartments (sites) numbered by the index i , we get for the mean numbers of B-particles at site i

$$\frac{\partial B_i(t)}{\partial t} = \frac{1}{2}j_{i-1}^-(t) + \frac{1}{2}j_{i+1}^-(t) - j_i^-(t) - kA_i(t)B_i(t). \quad (3)$$

Equation (3) is a local balance equation for the number of B-particles at site i with $j_i^-(t)$ being loss fluxes of particles B from the site i at time t given by

$$j_i^-(t) = \psi(t)P_B(t,0)B_i(0) + \int_0^t \psi(t-t')P_B(t,t') \times \left[\frac{\partial B_i(t')}{\partial t'} + j_i^-(t') + kA_i(t')B_i(t') \right] dt' \quad (4)$$

$$P_B(t,t') = \exp \left[-k \int_{t'}^t A_i(t'') dt'' \right], \quad (5)$$

where $\psi(t)$ is the waiting time pdf. In what follows, two different forms of $\psi(t)$ will be considered: the exponential one, $\psi(t) = \frac{1}{\tau} \exp(-\frac{t}{\tau})$, corresponding to normal Markovian diffusion, and a power-law asymptotic form $\psi(t) \propto t^{-1-\alpha}$ with $0 < \alpha < 1$, corresponding to anomalous diffusion. The first term on the right-hand side of Eq. (4) for the corresponding flux gives the contribution of those B-particles that were at i from the very beginning and survived until t . The second term describes the particles that arrived at i at a time t' and neither reacted nor performed a jump until t . Equation (5) gives the survival probability of B-particles. The A-concentration is then given by

$$\frac{\partial A_i(t)}{\partial t} = \frac{1}{2}f_{i-1}^-(t) + \frac{1}{2}f_{i+1}^-(t) - f_i^-(t) + kA_i(t)B_i(t), \quad (6)$$

$$f_i^-(t) = \psi(t)P_A(t,0)B_i(0) + \psi(t)A_i(0) + \int_0^t \psi(t-t') \left[\frac{\partial A_i(t')}{\partial t'} + f_i^-(t') - kA_i(t')B_i(t') \right] dt' + \int_0^t \psi(t-t')P_A(t,t') \times \left[\frac{B_i(t')}{\partial t'} + j_i^-(t') + kA_i(t')B_i(t') \right] dt', \quad (7)$$

$$P_A(t,t') = 1 - P_B(t,t'). \quad (8)$$

The loss fluxes for A-particles are denoted through $f_i(t)$. The first term on the right-hand side of Eq. (7) corresponds to the particles that were at i from the beginning and converted from B to A until t . The second term represents the A-particles that were at i from the very beginning. The third and fourth term describe the particles that arrived at i at a time t' as A-particles, or arrived as B-particles and reacted until t . The probability P_A to gain new A-particles arises from the conservation of the total number of particles and the probability P_B that the B-particles react. The A-concentration depends on the B-concentrations at all previous times.

Equations (3)–(8) are consistent with the conservation of the total number of particles in the reaction $A+B \rightarrow 2A$. Let $C_i = A_i + B_i$ be the total mean particle number at i and $g_i^-(t) = f_i^-(t) + j_i^-(t)$ the total loss flux of particles at i . From Eqs. (3) and (6), we obtain the balance equation,

$$\frac{\partial C_i(t)}{\partial t} = g_i^+(t) - g_i^-(t) = \frac{1}{2}g_{i-1}^-(t) + \frac{1}{2}g_{i+1}^-(t) - g_i^-(t). \quad (9)$$

From Eqs. (4) and (7), it follows that

$$\begin{aligned}
g_i^-(t) &= \psi(t)[B_i(0) + A_i(0)] + \int_0^t \psi(t-t') \\
&\quad \times \left[\frac{\partial B_i(t')}{\partial t'} + \frac{\partial A_i(t')}{\partial t'} + f_i^-(t') + j_i^-(t') \right] dt' \\
&= \psi(t)C_i(0) + \int_0^t \psi(t-t') \left[\frac{\partial C_i(t')}{\partial t'} + g_i^-(t') \right] dt'.
\end{aligned} \tag{10}$$

This equation can be solved by means of the Laplace transform,

$$\tilde{g}_i^-(u) = \frac{u\tilde{\psi}(u)}{1-\tilde{\psi}(u)}\tilde{C}_i(u), \tag{11}$$

which yields a diffusion equation for $C_i(t)$ in the Markovian case $\psi(t) = \frac{1}{\tau} \exp[-\frac{t}{\tau}]$, and a subdiffusion equation in the non-Markovian case $\psi(t) \propto t^{-1-\alpha}$, i.e., shows that the behavior of the total particle concentration is diffusive or subdiffusive, respectively. Moreover, if we choose the initial condition in a way that $C_i(t) = \text{const}$, the total number of particles is also locally conserved, $C_i(t) = C_i(0) = C_0$. This reduces the overall problem to the one for only one species. Passing to the continuous variables $x=ai$, we obtain

$$\frac{\partial B(x,t)}{\partial t} = \frac{a^2}{2}\Delta j^-(x,t) - k[B_0 - B(x,t)]B(x,t),$$

$$\begin{aligned}
j^-(x,t) &= \psi(t)P_B(x,t,0)B(x,0) + \int_0^t \psi(t-t')P_B(x,t,t') \\
&\quad \times \left[\frac{\partial B(x,t')}{\partial t'} + j^-(x,t') + k[B_0 - B(x,t')]B(x,t') \right] dt', \\
P_B(x,t,t') &= \exp \left[-k \int_{t'}^t [B_0 - B(x,t'')] dt'' \right],
\end{aligned} \tag{12}$$

where the concentration of B is given by $B(x,t) = B_i(t)/a^3$, and the corresponding dimensional constant is absorbed into a new reaction rate $k = \kappa a^3$. The concentration B_0 is set to unity in what follows. Equation (12) can be rewritten in a

form following from the equations of Ref. [16] for the irreversible case,

$$\begin{aligned}
\frac{\partial B(x,t)}{\partial t} &= -k[1 - B(x,t)]B(x,t) + \frac{a^2}{2}\Delta \int_0^t M(t-t') \\
&\quad \times B(x,t') \exp \left[- \int_{t'}^t k[1 - B(x,t'')] dt'' \right] dt'
\end{aligned} \tag{13}$$

with $\tilde{M}(u) = u\tilde{\psi}(u)/[1-\tilde{\psi}(u)]$.

B. Leading edge linearization

To analyze the behavior in the leading edge, we note that $A(x,t) = 1 - B(x,t)$ becomes small for $x \rightarrow \infty$ and hence

$$\begin{aligned}
\frac{\partial B}{\partial t} &= \frac{a^2}{2} \int_0^t \Delta \left\{ M(t-t')B(x,t') \right. \\
&\quad \left. \times \exp \left[-k \int_{t'}^t [1 - B(x,t'')] dt'' \right] \right\} dt' - k[1 - B(x,t)],
\end{aligned} \tag{14}$$

where we have interchanged the sequence of differentiation over x and temporal integration. For $x \rightarrow \infty$, we have $\exp\{-k \int_{t'}^t [1 - B(x,t'')] dt''\} \rightarrow 1$ so that the integrand can be put into the form

$$\begin{aligned}
M(t-t') &\left[\Delta B(x,t') + 2 \nabla B(x,t') \int_{t'}^t k \nabla B(x,t'') dt'' \right. \\
&\quad \left. + B(x,t') \int_{t'}^t k \Delta B(x,t'') dt'' \right. \\
&\quad \left. + B(x,t') \left(\int_{t'}^t k \nabla B(x,t'') dt'' \right)^2 \right].
\end{aligned}$$

We now assume $B(x,t)$ to be $1 - A_0 \exp[-\lambda(x-vt)]$ for large x , i.e., consider a wavefront with an exponentially decaying leading edge moving at a constant velocity v . Inserting this into Eq. (14) and retaining only the terms of the first order in A_0 , we get

$$\begin{aligned}
-\frac{\partial}{\partial t} \{A_0 \exp[-\lambda(x-vt)]\} &= -kA_0 \exp[-\lambda(x-vt)] + \frac{a^2}{2} \int_0^t M(t-t') \left(-\lambda^2 A_0 \exp[-\lambda(x-vt')] \right. \\
&\quad \left. + \frac{k\lambda}{v} A_0 \exp[-\lambda(x-vt')] - \frac{k\lambda}{v} A_0 \exp[-\lambda(x-vt)] \right) dt' \\
&= -kA_0 \exp[-\lambda(x-vt)] + \frac{a^2}{2} A_0 \left[-\lambda^2 + \frac{k\lambda}{v} \right] \int_0^t M(t-t') \exp[-\lambda(x-vt')] dt' \\
&\quad - \frac{a^2 k\lambda}{2v} A_0 \exp[-\lambda(x-vt)] \int_0^t M(t-t') dt',
\end{aligned} \tag{15}$$

with the memory kernel $\tilde{M}(u) = u\tilde{\psi}(u)/[1 - \tilde{\psi}(u)]$. We note that the traditional way to proceed is first to linearize the equation and then to insert the exponential solution, however, since the solution of the linearized equation is an exponential, we can also proceed the other way around, saving on tedious calculations.

1. Markovian case

Let us first show that for the Markovian case, the standard expression for the minimal velocity of the stable propagation is reproduced. Taking $\psi(t) = \frac{1}{\tau} \exp[-\frac{t}{\tau}]$, one obtains $M(t-t') = \frac{1}{\tau} \delta(t-t')$ and

$$\begin{aligned} 0 &= \frac{\partial}{\partial t} \{A_0 \exp[-\lambda(x-vt)]\} - kA_0 \exp[-\lambda(x-vt)] \\ &\quad + \frac{a^2}{2\tau} A_0 \exp[-\lambda(x-vt)] \left[-\lambda^2 + \frac{k\lambda}{v} - \frac{k\lambda}{v} \right] \\ &= \lambda v \exp[-\lambda(x-vt)] - kA_0 \exp[-\lambda(x-vt)] \\ &\quad - \frac{a^2}{2\tau} \lambda^2 A_0 \exp[-\lambda(x-vt)], \end{aligned} \tag{16}$$

which for $z = x - vt$, $z \rightarrow \infty$ leads us to the following equation:

$$\frac{a^2}{2\tau} \lambda^2 - v\lambda + k = 0. \tag{17}$$

This is the standard dispersion relation for the FKPP front, showing that real values of λ (corresponding to physically sound solutions with non-negative concentrations) are only possible for $v \geq v_{\min} = 2\sqrt{a^2 k / 2\tau} \equiv 2\sqrt{Dk}$ with $D = a^2 / 2\tau$ being the diffusion coefficient (note that B_0 is set to unity). Thus the existence of the minimal propagation velocity is connected to the fact that the dispersion relation (17) ceases to have real roots below some value of $v = v_{\min}$. The marginal stability principle then states that the solution propagating with $v = v_{\min}$ is the one really attained under a broad class of initial conditions. Note that the corresponding result emerges due to the cancellation of two terms of different nature in Eq. (16), which, as we proceed to show, does not take place in the non-Markovian case.

2. Non-Markovian case

We now assume the waiting time pdf to decay as a power law $\psi(t) \propto t^{-1-\alpha}$, $0 < \alpha < 1$ for large t . In this case, the integral operator with kernel M is the Riemann-Liouville fractional derivative [17]. With $\hat{t} = t - t'$, we get

$$\begin{aligned} \int_0^t M(t-t') \exp[\lambda vt'] dt' &= \exp[\lambda vt] \int_0^t M(\hat{t}) \exp[-\lambda v \hat{t}] d\hat{t} \\ &= \exp[\lambda vt] \tilde{M}(\lambda v) \end{aligned}$$

and moreover that

$$\int_0^t M(t-t') dt' = \frac{\text{const}}{\tau^\alpha} t^{\alpha-1}, \tag{18}$$

so that the last term in Eq. (15) vanishes for large t . Altogether we have then

$$\begin{aligned} &\frac{\partial}{\partial t} \{1 - A_0 \exp[-\lambda(x-vt)]\} \\ &= -kA_0 \exp[-\lambda(x-vt)] + \frac{a^2}{2} A_0 \exp[-\lambda(x-vt)] \\ &\quad \times \left[\left(-\lambda^2 + \frac{k\lambda}{v} \right) \tilde{M}(\lambda v) \right], \end{aligned}$$

and with $z = x - vt$

$$\begin{aligned} &- \lambda v A_0 \exp[-\lambda z] \\ &= -kA_0 \exp[-\lambda z] + \frac{a^2}{2} A_0 \exp[-\lambda z] \\ &\quad \times \left[\left(-\lambda^2 + \frac{k\lambda}{v} \right) \tilde{M}(\lambda v) \right], \\ &0 = -\lambda v + k + \frac{a^2}{2} \left(\lambda^2 - \frac{k\lambda}{v} \right) \tilde{M}(\lambda v). \end{aligned} \tag{19}$$

For example, taking $\psi(t)$ to be given by a Mittag-Leffler function $\psi(t) = E_\alpha[-(t/\tau)^\alpha]$, we have exactly $\tilde{\psi}(u) = [1 + (u\tau)^\alpha]^{-1}$ and $\tilde{M}(u) = \tau^{-\alpha} u^{1-\alpha}$, so that

$$0 = -\lambda v + k + \frac{a^2}{2\tau^\alpha} \left(\lambda^2 - \frac{k\lambda}{v} \right) (\lambda v)^{1-\alpha}.$$

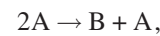
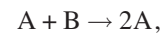
This equation is equivalent to

$$(v\lambda - k) \left(\frac{a^2}{2\tau^\alpha} \lambda^{2-\alpha} v^{-\alpha} - 1 \right) = 0 \tag{20}$$

and possesses two non-negative roots for any $v \geq 0$, at variance with the Markovian case, where such roots exist only for $v > v_{\min}$. This finding means that the minimal propagation velocity in this case is zero, which we interpret as if the asymptotic front will not propagate at all. The propagation failure corresponds essentially to a continuum approaching of $v_{\min} \propto \sqrt{D}$ to zero for the case in which $D \rightarrow 0$, as is the case in subdiffusion. We note that the Markovian case corresponding to $\alpha = 1$ is somewhat special. In this case, we have to retain the subleading term given by Eq. (18), which for $\alpha = 1$ turns out to be time-independent, i.e., of the same order of magnitude as the leading terms in Eq. (19).

III. COMPARISON WITH A RELATED MODEL OF REF. [6]

Our result here differs from the one of Yadav *et al.* [6], where the stable front propagation with the constant velocity was found. The authors of Ref. [6] consider a situation in which the A-particles undergo a reversible reaction corresponding to the branching-coalescence scheme,



which for the Markovian case is also described by the FKPP equation. The equation used in Ref. [6] for the non-Markovian case reads

$$\frac{\partial A(x,t)}{\partial t} = kA(x,t) - k_1[A(x,t)]^2 + a^2 \Delta \left\{ \int_0^t M[t-t'] A(x,t') \right. \\ \left. \times \exp \left[- \int_{t'}^t kA(x,t'') dt'' \right] dt' \right\}. \quad (21)$$

The velocity of the front was then obtained by means of the Hamilton-Jacobi approach via a hyperbolic scaling (a more elegant method known to lead to the same results as leading edge linearization in the Markovian case) and reads in our notation

$$v_{\min} = \frac{k(\alpha-3)}{\alpha-2} \sqrt{\frac{a^2}{\tau} \left[\frac{k(\alpha-3)}{\alpha-2} \frac{1-\alpha}{\tau} \right]^{\frac{1-\alpha}{2-\alpha}} \frac{2-\alpha}{k}} \\ = \sqrt{k^{2-\alpha} K_\alpha \left(\frac{\alpha-3}{\alpha-2} \right)^{3-\alpha} (2-\alpha)}, \quad (22)$$

with a generalized diffusion constant $K_\alpha = a^2/\tau^\alpha$.

Since the term quadratic in A does not contribute to the linearized solution determining the pulled front properties, the only difference between our approach and that of the authors of Ref. [6] is that Eq. (21) is written for the concentration of the A -particles that are *created* during the reaction, while our equation Eq. (13) is put down for the B -particles that *irreversibly disappear*. Taking Eq. (21) for granted, we first checked that the method of the leading-edge linearization used in the previous sections yields the same result for the minimal front velocity as in Ref. [6]. Therefore, the differences between the results are not connected to the method applied, but to the equation itself.

The difference in the results is explained by the fact that the equations for particles that are created during the reaction under the conditions discussed look differently from Eq. (21): Since the particle making its step as A might have entered the corresponding site both as A and as B (and hav-

ing undergone a transformation), the equation for the product of the reaction (in our case A) has to contain the Laplacians of both A and B concentrations, as discussed in [14]. Moreover, considering reversible reactions put additional problems due to possible multiple $A \rightleftharpoons B$ transformations during the waiting time at one site [15]. Although leading to the classical FKPP results in the Markovian case, the equations proposed in [16] are not well suited for the description of particles created during the reaction and for reversible reactions under conditions discussed in the beginning of the paper.

IV. CONCLUSION

Let us summarize our findings. We considered the reaction-subdiffusion problem for the irreversible autocatalytic conversion reaction $A+B \rightarrow 2A$, which, in the case of normal diffusion, is described by the FKPP equation. We show that in the case of subdiffusion, under the assumption of the reaction taking place irrespectively of the particles' mesoscopic motion, the minimal front's propagation velocity is zero, suggesting the propagation failure. This regime can be considered as a result of a continuous transition to $D \rightarrow 0$ in the normal FKPP situation. The result obtained is in contrast to the treatment in [6] where the finite propagation velocity of a traveling wave was found. The differences in the assumptions of the two approaches (corresponding essentially to two *very different situations*) are discussed.

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