Kinetic equations for reaction-subdiffusion systems: Derivation and stability analysis

A. Yadav and Werner Horsthemke
Department of Chemistry, Southern Methodist University, Dallas, Texas 75275-0314, USA
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We derive general kinetic equations for reacting and subdiffusing entities based on a nonlinear continuous time random walk formalism proposed by Vlad and Ross [Phys. Rev. E **66**, 061908 (2002)]. Reaction and diffusion processes are separable in a typical reaction-diffusion system, and their combined influence on the evolution of the density of a species is a simple sum. Our derivation shows that this is no longer true for subdiffusive entities undergoing reactions. The strong memory effects in the transport process, i.e., the non-Markovian nature of subdiffusion, results in a nontrivial combination of reactions and spatial dispersal, which we discuss in detail. We carry out a linear stability analysis of the derived reaction-subdiffusion system to understand the effects of memory on pattern formation. We find that the Turing instability persists in the subdiffusive system. However, the memory modifies the Turing threshold and the characteristics of the band of unstable modes close to this threshold.

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I. INTRODUCTION

Systems of particles or individuals, such as molecules or organisms, that spread spatially and interact with each other play an important role in physics, chemistry, biology, and other sciences and are often modeled by reaction-diffusion equations, if fluctuations can be ignored [1]. The interaction of entities is captured by the kinetic rate terms, and spatial dispersal is described by the diffusion terms. Reaction-diffusion models have advanced the understanding of many important facets of complex nonequilibrium systems, most notably the various phenomena of pattern formation [2,3].

In the classical reaction-diffusion picture, the effects of reaction and diffusion are separable and combine additively to influence the total spatiotemporal evolution of the concentration field of a given species, $\rho(x,t)$, a direct consequence of the Markovian nature of diffusion (Brownian motion) and reaction kinetics [1]. Brownian motion is a memoryless transport process, and diffusion appears as a local operator in a standard reaction-diffusion equation,

$$\frac{\partial\rho(x,t)}{\partial t} = D\nabla^2\rho(x,t) + f(\rho(x,t)). \tag{1}$$

The combined effect of reactions and transport with memory on the dynamics of a complex nonequilibrium system is still not well understood. An important class of non-Markovian transport processes is subdiffusion, with ample motivating experimental contexts ranging from proteins in cell membranes [4] to transport in media with obstacles or binding sites [5–7]. Subdiffusion is often modeled by a continuous time random walk (CTRW) [8] characterized by a waiting time probability density function (PDF) $\phi(t)$ and a spatial jump length PDF $\lambda(r)$. In the absence of reactions, the large-scale, long-time limit of the CTRW is a normal diffusion process, if the waiting time distribution has a finite mean and the spatial jump distribution has a finite variance. The density $\rho(x,t)$ obeys the classical diffusion equation (Fick's law), and $\langle x^2(t)\rangle \propto t$, where $\langle x^2(t)\rangle$ is the mean squared spatial displacement [9]. A waiting time distribution with a long tail, $\phi(t) \sim t^{-1-\alpha}$ $(0<\alpha<1)$, no longer has a finite mean waiting time, and the resultant large-scale, long-time limit corresponds to subdiffusion. The evolution of the density $\rho(x,t)$ may be viewed as governed by a fractional diffusion equation [10,11], and $\langle x^2(t)\rangle \propto t^{\alpha}$ [9]. A fractional time derivative is inherently a nonlocal operator involving memory, and subdiffusion is a non-Markovian process. There is good experimental evidence for the occurrence of long-tailed waiting time PDFs in a variety of natural and technological systems; for a review see, e.g., Ref. [12]. Theoretical analyses of disordered systems with deep traps, e.g., amorphous solids, show that such systems display waiting time PDFs with a long tail; see, e.g., [13,14].

While the classical diffusion equation has a fractional diffusion equation as its analog in the subdiffusive case, it is unclear how to properly take account of the combined effects of subdiffusion and reactions. The crucial question is if the effects of subdiffusion and reaction are separable as in the classical reaction-diffusion system, or does memory prevent such a separation? A second question concerns the consequences of the strong memory effects in the transport process for spatial pattern formation.

Various schemes have been advanced previously in an attempt to combine reactions and subdiffusion in the activation-controlled regime. Reaction terms are simply added to a fractional diffusion equation in Refs. [15–17], assuming at the outset that subdiffusion and reactions are separable. In some cases, a fractional time derivative is applied to both the diffusion and reaction terms [18], again assuming that the effects of subdiffusion and reactions are simply additive. Hornung, Berkowitz, and Barkai have introduced a modified CTRW framework to model subdiffusing morphogens with degradation [19]. They assume that subdiffusive jumps and reactions (degradation) are mutually exclusive. It may be shown that this framework is equivalent to a fractional diffusion equation with a time fractional derivative acting on the linear degradation term.

The aim of this paper is twofold. First, starting from the nonlinear CTRW formalism proposed by Vlad and Ross [20,21], we derive a general reaction-subdiffusion equation and show that the contributions of subdiffusion and reactions to the evolution of the density of a species are not separable. In the context of chemical systems, the type of evolution equations we study in this paper describe reactions in the activation-controlled limit. We do not consider the opposite regime of subdiffusion-controlled chemical reactions [18,22–27].

Second, we perform a linear stability analysis of the derived reaction-subdiffusion equations with the goal to understand the influence of the strong memory in the transport on pattern formation, specifically the Turing instability [28]. We show that memory effects associated with subdiffusion shift the Turing instability threshold in parameter space. Also, the characteristic size and the location of upper and lower cutoffs to the band of unstable modes change as a direct consequence of memory.

This paper is organized as follows. In Sec. II we briefly summarize the CTRW description introduced in Ref. [20] and derive a proper reaction-subdiffusion equation in the large-scale, long-time limit. We carry out a linear stability analysis of the reaction-subdiffusion system in Sec. III, derive the condition for a Turing instability to occur, and determine the band of unstable modes.

II. REACTION-SUBDIFFUSION EQUATIONS

To account for the memory effects of the transport, Vlad and Ross take the age structure of the system explicitly into account [20,21]. Let $\xi_i(x,t,\tau)$ be the density of particles of type i $(i=1,2,\ldots,n)$ whose waiting time (age) at position x and time t lies in the range $(\tau,\tau+d\tau)$. The concentration of species i, $\rho_i(x,t)$, at position x and time t, is then given by

$$\rho_i(x,t)=\int_0^\infty \xi_i(x,t,\tau)d\tau. \tag{2}$$

The entities undergo reactions, birth-and-death processes, with a birth rate $R_i^+(\rho(x,t))\geqslant 0$ and a death rate $R_i^-(\rho(x,t))\geqslant 0$, where $\rho(x,t)=(\rho_1(x,t),\rho_2(x,t),\ldots,\rho_n(x,t))$. It is a basic principle of kinetics that the rate of removal or death of particles of a given type must go to zero as the density of the particles goes to zero, $R_i^-(\rho)\to 0$ as $\rho_i\to 0$. Otherwise, the concentration ρ_i of those particles can become negative, which is unphysical. To ensure the nonnegativity of the age-dependent densities $\xi_i(x,t,\tau)$, it is sufficient to require that $R_i^-(\rho)/\rho_i$ remains bounded from above as $\rho_i\to 0$. Define $W_i(x'\to x,\tau')$ to be the rate at which an individual of the species i with an age between τ and $\tau+d\tau$ moves from position x' to x. The evolution of $\xi_i(x,t,\tau)$ is governed by the balance equation [20]

$$\left(\frac{\partial}{\partial t}+\frac{\partial}{\partial \tau}\right)\xi_i(x,t,\tau)=-\xi_i(x,t,\tau)\int_{x'}W_i(x\to x',\tau)dx' - \frac{\xi_i(x,t,\tau)}{\rho_i(x,t)}R_i^-(\rho(x,t)), \tag{3}$$

with the boundary condition

$$\xi_i(x,t,\tau=0)=R_i^+(\rho(x,t))+\int_{x'}\int_{\tau'}\xi_i(x',t,\tau') \times W_i(x'\to x,\tau')dx'd\tau'. \tag{4}$$

This boundary condition implies that entities with zero age at a particular position are either created there with a rate $R_i^+(\rho(x,t))$, or arrive there from other positions.

We denote the joint PDF of jumps and waiting times of the CTRW of species i by $\psi_i(x\to x',\tau)$ and introduce the survival probability of a particle of type i at position x:

$$l_i(x,\tau)=\int_{x'}\int_{\tau'=\tau}^\infty \psi_i(x\to x',\tau')dx'd\tau'. \tag{5}$$

The connection between $W_i(x\to x',\tau')$ and $\psi_i(x\to x',\tau)$ is given by the following relation [21]:

$$\psi_i(x\to x',\tau)=l_i(x,\tau)W_i(x\to x',\tau). \tag{6}$$

We differentiate Eq. (5) with respect to τ and use Eq. (6) to obtain

$$\frac{\partial l_i(x,\tau)}{\partial \tau}=-l_i(x,\tau)\int_{x'}W_i(x\to x',\tau)dx', \tag{7}$$

or

$$l_i(x,\tau)=\exp\left(-\int_{x'}\int_0^\tau W_i(x\to x',\tau')dx'd\tau'\right). \tag{8}$$

The solution to Eq. (3) with boundary condition Eq. (4) reads [20]

$$\begin{aligned}Z_i(x,t)=&R_i^+(\rho(x,t))+\int_0^t\int_{x'}Z_i(x',t-\tau')\psi_i(x'\to x,\tau')\\&\times\exp\left(-\int_{t-\tau'}^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx'd\tau'\\&+\int_t^\infty\int_{x'}\xi_i(x',t=0,\tau'-t)\frac{\psi_i(x'\to x,\tau')}{l_i(x',\tau'-t)}\\&\times\exp\left(-\int_0^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx'd\tau',\end{aligned} \tag{9}$$

$$\begin{aligned}\rho_i(x,t)=&\int_0^t l_i(x,\tau)Z_i(x,t-\tau)\exp\left(-\int_{t-\tau}^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)d\tau\\&+\int_t^\infty\xi_i(x,t=0,\tau-t)\frac{l_i(x,\tau)}{l_i(x,\tau-t)}\\&\times\exp\left(-\int_0^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)d\tau,\end{aligned} \tag{10}$$

where $Z_i(x,t)$ is defined to be the zero-age density $Z_i(x,t)\equiv\xi_i(x,t,\tau=0)$. Equations (9) and (10) extend the usual linear CTRW formalism to include general nonlinear birth and death processes. We use these equations as our starting point to derive reaction-subdiffusion equations with arbitrary nonlinear kinetic rate terms.

In the following we consider the usual case of a spatially homogeneous CTRW with independent jump and waiting time PDFs, i.e., $\psi_i(x\to x',\tau)=\psi_i(x'-x,\tau)=\lambda_i(x'-x)\phi_i(\tau)$. The survival probability then does not depend on position, $l_i(x,\tau)=l_i(\tau)$. Without loss of generality, we choose the initial condition as $\xi_i(x,t=0,\tau)=\rho_i(x,0)\delta(\tau)$, i.e., at time zero all individuals are at the beginning of a waiting period. With the transformations $t-\tau'=t'$, $t-\tau=t'$ in Eqs. (9) and (10), respectively, we obtain,

$$Z_i(x,t)=R_i^+(\rho(x,t))+\int_0^t\int_{x'}Z_i(x',t')\psi_i(x-x',t-t')\times\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx'dt'+\int\rho_i(x',0)\psi_i(x-x',t)\times\exp\left(-\int_0^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx' \tag{11}$$

and

$$\rho_i(x,t)=\int_0^t l_i(t-t')Z_i(x,t')\times\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)dt'+\rho_i(x,0)l_i(t)\times\exp\left(-\int_0^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right). \tag{12}$$

In the absence of reaction terms, Eqs. (11) and (12) reduce to the usual linear CTRW formalism, e.g. (see [9]),

$$Z_i(x,t)=\int_0^t\int_{x'}Z_i(x',t')\psi_i(x-x',t-t')dx'dt'+\int\rho_i(x',0)\psi_i(x-x',t)dx', \tag{13}$$

$$\rho_i(x,t)=\int_0^t l_i(t-t')Z_i(x,t')dt'+\rho_i(x,0)l_i(t). \tag{14}$$

Taking the Laplace and Fourier transforms of Eqs. (13) and (14), we obtain

$$\rho_i(k,u)=\frac{1-\phi_i(u)}{u}\frac{\rho_i(k,t=0)}{1-\lambda_i(k)\phi_i(u)}. \tag{15}$$

Here, and in what follows, the Laplace transform of a function $P(x,t)$ is denoted either as $L[P(x,t)]$ or as $P(x,u)$, where u is the conjugate variable in Laplace space. Similarly, the Fourier transform of a function $P(x,t)$ is denoted as $P(k,t)$, where k is the wave number. The combined Laplace-Fourier transform of $P(x,t)$ is $P(k,u)$. Since our goal is to derive reaction-subdiffusion equations, we consider in the following CTRWs with short-range jump length PDFs $\lambda(r)$, e.g., a Gaussian with variance $\sigma^2/2$, and long-tailed waiting time PDFs, e.g., a PDF derived from a Mittag-Leffler function for the survival probability, $l(t)=E_\alpha(-t^\alpha)$ with $0<\alpha<1$ [29]. The asymptotic behavior of the waiting time PDF is given by $\phi(t)\sim t^{-(1+\alpha)}$ as $t\to\infty$. To take the long-time limit, we consider the scaled waiting time PDF [29]:

$$\phi_i(t)\to\frac{\phi_i(t/\eta_i)}{\eta_i},\quad \eta_i>0, \tag{16}$$

which results in

$$\phi_i(u)\to 1-(u\eta_i)^\alpha+o(\eta_i^\alpha). \tag{17}$$

Consequently, the long-time limit corresponds to setting $\phi_i(u)=1-(u\eta_i)^\alpha$ with $\eta_i\to 0$. Similarly, the large-scale limit corresponds to setting $\lambda_i(k)=1-\sigma_i^2k^2$ with $\sigma_i\to 0$. Taking the inverse Laplace and Fourier transforms of Eq. (15), we recover a normal diffusion equation if $\alpha=1$ and a fractional diffusion equation for $0<\alpha<1$.

We now proceed to derive the long-time and large-spatial scale behavior of the general system of Eqs. (11) and (12). We differentiate Eq. (12) with respect to t and obtain

$$\frac{\partial\rho_i(x,t)}{\partial t}=-\int_0^t\phi_i(t-t')Z_i(x,t')\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)dt'-\frac{R_i^-(\rho(x,t))}{\rho_i(x,t)}\int_0^t l_i(t-t')Z_i(x,t')\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)dt'+Z_i(x,t)-\rho_i(x,0)\phi_i(t)\exp\left(-\int_0^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)-\frac{R_i^-(\rho(x,t))}{\rho_i(x,t)}\rho_i(x,0)l_i(t)\exp\left(-\int_0^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right). \tag{18}$$

Substituting Eqs. (11) and (12) into Eq. (18), we get

$$\frac{\partial\rho_i(x,t)}{\partial t}=R_i^+(\rho(x,t))-R_i^-(\rho(x,t))+\int_0^t\int_{x'}\psi_i(x-x',t-t')Z_i(x',t')\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx'dt'-\int_0^t\phi_i(t-t')Z_i(x,t')\exp\left(-\int_{t'}^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right)dt'+\int\rho_i(x',0)\psi_i(x-x',t)\exp\left(-\int_0^t\frac{R_i^-(\rho(x',t''))}{\rho_i(x',t'')}dt''\right)dx'-\rho_i(x,0)\phi_i(t)\exp\left(-\int_0^t\frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')}dt''\right). \tag{19}$$

In the large-spatial scale limit, $\lambda_i(k)=1-\sigma_i^2 k^2$, Eq. (19) simplifies to the integro-differential equation

$$\begin{aligned}\frac{\partial \rho_i(x,t)}{\partial t} &= R_i^+(\rho(x,t)) - R_i^-(\rho(x,t)) \\ &\quad + \sigma_i^2 \nabla^2 \left[\int_0^t \phi_i(t-t') Z_i(x,t') \right. \\ &\quad \left. \times \exp\left(- \int_{t'}^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' \right] \\ &\quad + \sigma_i^2 \nabla^2 \left[\phi_i(t)\rho_i(x,0) \exp\left(- \int_0^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) \right].\end{aligned} \tag{20}$$

Terms representing the influence of the initial conditions become negligible in the long-time limit and we find

$$\begin{aligned}\frac{\partial \rho_i(x,t)}{\partial t} &= R_i^+(\rho(x,t)) - R_i^-(\rho(x,t)) \\ &\quad + \sigma_i^2 \nabla^2 \left[\int_0^t \phi_i(t-t') Z_i(x,t') \right. \\ &\quad \left. \times \exp\left(- \int_{t'}^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' \right],\end{aligned} \tag{21}$$

$$\rho_i(x,t) = \int_0^t l_i(t-t') Z_i(x,t') \exp\left(- \int_{t'}^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' . \tag{22}$$

We now eliminate $Z_i(x,t)$ from the system of Eqs. (21) and (22). Rewriting Eq. (22) as

$$\begin{aligned}&\rho_i(x,t) \exp\left(\int_0^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) \\ &= \int_0^t l_i(t-t') Z_i(x,t') \exp\left(\int_0^{t'} \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt'\end{aligned} \tag{23}$$

and Laplace transforming, we obtain

$$\begin{aligned}&\frac{u\phi_i(u)}{1-\phi_i(u)} L\left[\rho_i(x,t) \exp\left(\int_0^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) \right] \\ &= \phi_i(u) L\left[Z_i(x,t') \times \exp\left(\int_0^{t'} \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) \right].\end{aligned} \tag{24}$$

Inverse Laplace transforming Eq. (24) leads to

$$\begin{aligned}&\int_0^t \Theta_i(t-t') \rho_i(x,t') \exp\left(\int_0^{t'} \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' \\ &= \int_0^t \phi_i(t-t') Z_i(x,t') \exp\left(\int_0^{t'} \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' ,\end{aligned} \tag{25}$$

where we define $\Theta_i(u) \equiv u\phi_i(u)/[1-\phi_i(u)]$. Equation (21) may be rewritten as

$$\begin{aligned}\frac{\partial \rho_i(x,t)}{\partial t} &= R_i^+(\rho(x,t)) - R_i^-(\rho(x,t)) + \sigma_i^2 \nabla^2 \\ &\quad \times \left[\exp\left(- \int_0^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) \int_0^t \phi_i(t-t') Z_i(x,t') \right. \\ &\quad \left. \times \exp\left(\int_0^{t'} \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' \right].\end{aligned} \tag{26}$$

Substituting Eq. (25) into Eq. (26) provides

$$\begin{aligned}\frac{\partial \rho_i(x,t)}{\partial t} &= R_i^+(\rho(x,t)) - R_i^-(\rho(x,t)) + \sigma_i^2 \nabla^2 \left[\int_0^t \Theta_i(t-t') \right. \\ &\quad \left. \times \rho_i(x,t') \exp\left(- \int_{t'}^t \frac{R_i^-(\rho(x,t''))}{\rho_i(x,t'')} dt'' \right) dt' \right],\end{aligned} \tag{27}$$

which is the generalized reaction-diffusion equation that we sought. The reaction terms and the Laplacian operator in Eq. (27) are reminiscent of a standard reaction-diffusion equation. However, unlike in a standard reaction-diffusion equation, the Laplacian acts on a nonlocal memory term captured by a time integral. The presence of both the kernel $\Theta_i(t-t')$, related to the waiting time PDF of the CTRW, and the death rate $R_i^-(\rho(x,t))$ in the memory term indicates that the effects of reaction and subdiffusion are, indeed, not separable.

To illustrate this point further, we chose $R_i^+(\rho(x,t))=0$ and $R_i^-(\rho(x,t))=\epsilon\rho_i(x,t)$, i.e., a linear death process. With this choice of reaction terms, Laplace transforming Eq. (27) and taking the long-time limit $\phi_i(u)=1-(u\eta_i)^\alpha$, we obtain an evolution equation for the density $\rho_i(x,t)$ in the long-time and large-spatial scale limit, which reads

$$u\rho_i(x,u) = \rho_i(x,t=0) + \frac{(u+\epsilon)^{1-\alpha}}{\eta_i^\alpha} \sigma_i^2 \nabla^2 \rho_i(x,u) - \epsilon\rho_i(x,u) \tag{28}$$

in Laplace space. If $\epsilon=0$, Eq. (28) reduces to the usual fractional diffusion equation [9]. In the presence of reactions (nonzero ϵ), the reaction and subdiffusive motion is coupled, as seen by the presence of the term $[(u+\epsilon)^{1-\alpha}/\eta_i^\alpha]\sigma_i^2\nabla^2\rho_i(x,u)$. For $\alpha=1$, a standard reaction-diffusion equation results. In recent works [30,31], Sokolov *et al.* also arrive at Eq. (28). However, their treatment is limited to linear kinetics.

In deriving Eq. (28), we make certain implicit assumptions about subdiffusion and reaction time scales that we now elaborate upon. In the absence of reactions, subdiffusive behavior results from the accumulation of many jumps of the CTRW on a time scale much larger than η_i. Mathematically, one obtains this limit by setting $\phi_i(u) \sim 1-(u\eta_i)^\alpha$, neglecting higher powers of $u\eta_i$ since the long-time scale of subdiffusion $1/u$ is large compared to η_i. In the presence of reactions, since one is interested in the combined influence of reactions and subdiffusion, it must be true that reaction and subdiffusion processes occur on similar time scales. Thus $1/u \sim 1/\epsilon$, and the long-time limit of a combined reaction subdiffusion process $\phi_i^\epsilon(t)=\phi_i(t)\exp(-\epsilon t)$ is given by $\phi_i^\epsilon(u)=1-[(u+\epsilon)\eta_i]^\alpha$. Furthermore, for this reason, terms of size $O(k^2[(u+\epsilon)\eta_i])$ are neglected, whereas terms of size $O(k^2[(u+\epsilon)\eta_i]^\alpha)$ and $O(k^2[(u+\epsilon)\eta_i]^{1-\alpha})$ are retained while deriving Eq. (28). It is easy to see that Eq. (27) simplifies to a classical reaction-diffusion system if the CTRW is Markovian, i.e., the waiting times are exponentially distributed, $\phi_i(t)=(1/\eta_i)e^{-t/\eta_i}$. In this case $\Theta_i(u)=1/\eta_i$, and therefore $\Theta_i(t)=\delta(t)/\eta_i$.

III. LINEAR STABILITY OF REACTION-SUBDIFFUSION EQUATIONS

Having derived a reaction-subdiffusion equation, Eq. (27), we seek to analyze its linear stability properties, especially the appearance of a Turing instability. In a classical reaction-diffusion system, Turing patterns form if the homogeneous steady state undergoes a stationary instability with respect to perturbations with finite wavelengths caused by coupling between diffusion and the nonlinear kinetics. Beyond the Turing instability threshold, a band of modes with an upper and lower cutoff wavelength are unstable. The final pattern arises from a competition between these growing (unstable) modes.

We show that the Turing instability persists in the presence of subdiffusion. The memory effects associated with subdiffusion shift the Turing instability threshold. Also, as we shall see, the characteristic size of a pattern and the upper and lower cutoffs in the band of unstable modes change as a direct consequence of memory.

We examine the linear stability of a system consisting of two species $i=1,2$, with total density fields $\rho_1(x,t)$, $\rho_2(x,t)$ obeying Eq. (27). The density fields for different entities are coupled by the reaction terms $R_1^+(\rho(x,t))$, $R_1^-(\rho(x,t))$, $R_2^+(\rho(x,t))$, and $R_2^-(\rho(x,t))$, where $\rho(x,t)=(\rho_1(x,t),\rho_2(x,t))$.

The homogeneous steady state solution of Eq. (27) is given by the solution of $R_i^+(\rho(x,t))-R_i^-(\rho(x,t))=0$, since the contributions of the time derivative and the operation of the Laplacian vanish. We assume in the following that, as usual, the homogeneous steady state is nontrivial, i.e., the steady state densities are nonzero. We denote the steady state fields by ρ_i^0.

Introducing the perturbations $\delta\rho_1(x,t)$ and $\delta\rho_2(x,t)$ about the steady state fields ρ_1^0 and ρ_2^0 in Eq. (27) for $i=1,2$, results in

$$\begin{aligned}\frac{\partial \delta\rho_1(x,t)}{\partial t} &= \sigma_1^2\nabla^2\left[\rho_1^0\int_0^t \Theta_1(t-t')e^{-p(t-t')}\left(-A_1\int_{t'}^t \delta\rho_1(x,t'')\right.\right.\\ &\quad \left.\left.\times dt'' - A_2\int_{t'}^t \delta\rho_2(x,t'')dt''\right)dt'\right]\\ &\quad + \sigma_1^2\nabla^2\int_0^t \delta\rho_1(x,t')\Theta_1(t-t')e^{-p(t-t')}dt'\\ &\quad + R_{11}^+(\rho^0)\delta\rho_1(x,t) + R_{12}^+(\rho^0)\delta\rho_2(x,t)\\ &\quad - R_{11}^-(\rho^0)\delta\rho_1(x,t) - R_{12}^-(\rho^0)\delta\rho_2(x,t), \end{aligned} \tag{29}$$

where the second index in the subscript denotes the derivative with respect to that indexed field,

$$A_1 = \frac{\partial}{\partial\rho_1}\left[\frac{R_1^-(\rho(x,t))}{\rho_1(x,t)}\right]_{(\rho_1^0,\rho_2^0)},$$

$$A_2 = \frac{\partial}{\partial\rho_2}\left[\frac{R_1^-(\rho(x,t))}{\rho_1(x,t)}\right]_{(\rho_1^0,\rho_2^0)}, \tag{30}$$

and $p=R_1^-(\rho^0)/\rho_1^0>0$. Here ρ^0 is the vector of steady state values of all pertinent fields.

We focus on the case of subdiffusion and take the long-time limit $\phi_1(u)=1-(u\eta_1)^\alpha$. For rational values of α, we can Laplace and Fourier transform Eq. (29) and arrive at (see the Appendix),

$$\begin{aligned} u\delta\rho_1(k,u) &= \delta\rho_1(k,t=0) - \frac{\sigma_1^2k^2\rho_1^0(p\eta_1)^{1-\alpha}h_\alpha(u,p)}{u\eta_1}\\ &\quad \times[A_1\delta\rho_1(k,u)+A_2\delta\rho_2(k,u)]\\ &\quad - \frac{\sigma_1^2k^2(u+p)^{1-\alpha}}{\eta_1^\alpha}\delta\rho_1(k,u) + R_{11}^+(\rho^0)\delta\rho_1(k,u)\\ &\quad + R_{12}^+(\rho^0)\delta\rho_2(k,u) - R_{11}^-(\rho^0)\delta\rho_1(k,u)\\ &\quad - R_{12}^-(\rho^0)\delta\rho_2(k,u), \end{aligned} \tag{31}$$

where $h_\alpha(u,p)=1-p^{\alpha-1}(u+p)^{1-\alpha}$. Proceeding in the same way, we obtain for the second species

$$\begin{aligned} u\delta\rho_2(k,u) &= \delta\rho_2(k,t=0) - \frac{\sigma_2^2k^2\rho_2^0(q\eta_2)^{1-\beta}h_\beta(u,q)}{u\eta_2}\\ &\quad \times[B_1\delta\rho_1(k,u)+B_2\delta\rho_2(k,u)]\\ &\quad - \frac{\sigma_2^2k^2(u+q)^{1-\beta}}{\eta_2^\beta}\delta\rho_2(k,u) + R_{22}^+(\rho^0)\delta\rho_2(k,u)\\ &\quad + R_{21}^+(\rho^0)\delta\rho_1(k,u) - R_{22}^-(\rho^0)\delta\rho_2(k,u)\\ &\quad - R_{21}^-(\rho^0)\delta\rho_1(k,u), \end{aligned} \tag{32}$$

where

$$B_1 = \frac{\partial}{\partial\rho_1}\left[\frac{R_2^-(\rho(x,t))}{\rho_2(x,t)}\right]_{(\rho_1^0,\rho_2^0)},$$

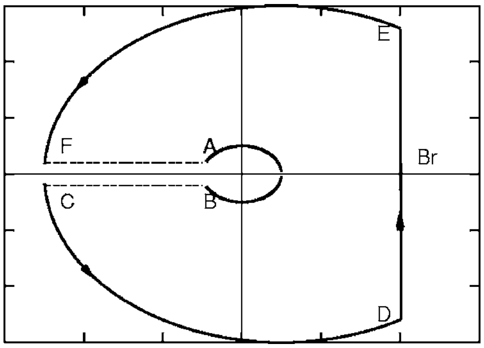

FIG. 1. Contour in the u plane.

$$B_2 = \frac{\partial}{\partial \rho_2}\left[\frac{R_2^-(\rho(x,t))}{\rho_2(x,t)}\right]_{(\rho_1^0,\rho_2^0)}, \tag{33}$$

$q = R_2^-(\rho^0)/\rho_2^0 > 0$, and $\phi_2(u) = 1-(u\eta_2)^\beta$. The stability properties of the homogeneous steady state and the conditions for a Turing instability may now be obtained by solving for $\delta\rho_1(k,u)$ and $\delta\rho_2(k,u)$ using Eqs. (31) and (32), followed by taking an inverse Laplace transform. For simplicity we assume that $\beta=1$, i.e., the second entity undergoes normal diffusion. Solving Eqs. (31) and (32), we obtain

$$\delta\rho_1(k,u) = \frac{\nu(u)}{\Delta(u)}, \tag{34}$$

where

$$\nu(u) = (u - g_2 + \sigma_2^2 k^2/\eta_2)\delta\rho_1(k,t=0) + \left(f_2 - \sigma_1^2 k^2 \rho_1^0 (p\eta_1)^{1-\alpha}\frac{h_\alpha(u,p)}{u\eta_1}\right)\delta\rho_2(k,t=0) \tag{35}$$

and

$$\Delta(u) = [u - f_1 + \sigma_1^2 k^2 (u+p)^{1-\alpha}/\eta_1^\alpha][u - g_2 + \sigma_2^2 k^2/\eta_2] - g_1 f_2 + \sigma_1^2 k^2 \rho_1 (p\eta_1)^{1-\alpha}\frac{h_\alpha(u,p)}{u\eta_1}(A_2 g_1 + A_1 u - A_1 g_2 + A_1 \sigma_2^2 k^2/\eta_2). \tag{36}$$

The combined reaction terms are defined as $f \equiv R_1^+(\rho(x,t)) - R_1^-(\rho(x,t))$ and $g \equiv R_2^+(\rho(x,t)) - R_2^-(\rho(x,t))$, and $f_i \equiv (\partial f/\partial \rho_i)|_{\rho^0}$ and $g_i \equiv (\partial g/\partial \rho_i)|_{\rho^0}$. A similar expression for $\delta\rho_2(k,u)$ may be written down with the same denominator as in Eq. (34).

We now show that the long-time asymptotic behavior of the perturbation $\delta\rho_1(k,u)$ is controlled only by the zeros of the denominator in Eq. (34). The inverse Laplace transform of Eq. (34) is given by the integral

$$S_{Br} = \frac{1}{2\pi i}\int_{Br} \delta\rho_1(k,u)e^{ut}du. \tag{37}$$

The contour Br, depicted in Fig. 1, lies to the right of all the poles of $\delta\rho_1(k,u)$ in the complex u plane. The point $u=-p$ is a branch point, and the branch cuts lie in the left half plane as shown. Consider the integral

$$S = \frac{1}{2\pi i}\oint \delta\rho_1(k,u)e^{ut}du, \tag{38}$$

along the closed contour $ABCDEF$. The arcs EF and CD, forming a semicircle in the left half plane, are represented in polar coordinates by $u+p = R\exp(i\theta)$. In the limit $R\to\infty$, we have $\delta\rho_1(k,Re^{i\theta})\to 0$. Therefore, by Jordan's lemma [32], the integral along these arcs vanishes as $R\to\infty$. The circle obtained by completing the arc AB is given by $u+p = r\exp(i\theta)$ in polar coordinates. Thus

$$S_{AB} = \frac{e^{-pt}}{2\pi i}\int \delta\rho_1(k,re^{i\theta})ire^{i\theta}d\theta \tag{39}$$

vanishes in the limit $r\to 0$, since in this limit $\delta\rho_1(k,re^{i\theta})$ is a constant.

In the double limit $r\to 0$ and $R\to\infty$, $u+p = y\exp(i\pi) = -y$ along the segment FA, where y is a real variable. The integral along the segment FA is written as

$$S_{FA} = -\frac{e^{-pt}}{2\pi i}\int_0^\infty \delta\rho_1(k,-p-y)e^{-yt}dy. \tag{40}$$

It is easily verified by examining Eq. (34) that $y=0$, and hence $u=-p$, is not a pole of $\delta\rho_1(k,u)$ for arbitrary values of the reaction rates f, g, and constants σ_i and η_i. Hence, $\delta\rho_1(k,-p-y)$ may be Taylor expanded about $y=0$ to provide a form of the integral S_{FA} suitable for the application of Watson's lemma [33]. The lemma states that

$$\int_0^\infty e^{-yt}H(y)dy \sim \sum_n^\infty \left.\frac{d^n H(y)}{dy^n}\right|_{y=0}\frac{\Gamma(n+1)}{n!t^{n+1}}, \tag{41}$$

as $t\to\infty$ and for a general $H(y)$ with a Taylor series expansion about $y=0$. Therefore, we have

$$S_{FA} \sim -\frac{e^{-pt}}{2\pi i}\sum_n^\infty \left.\frac{d^n\delta\rho_1(k,-p-y)}{dy^n}\right|_{y=0}\frac{\Gamma(n+1)}{n!t^{n+1}}, \tag{42}$$

a vanishing contribution in long-time limit. Similarly one may show that the contribution S_{BC}, along the segment BC vanishes as $t\to\infty$.

The integral along the closed contour $ABCDEF$ equals the sum of residues inside the contour. However, as we have shown, the contributions along all curves except the Bromwich contour Br vanish. Therefore, S_{Br} equals the sum of residues:

$$S_{Br} = \delta\rho_1(k,t) = \sum \mathrm{Res}[\delta\rho_1(k,u)] = \sum_i \chi(u_i)e^{u_i t}. \tag{43}$$

Here u_i are the zeros of the denominator $\Delta(u_i)$ on the right hand side of Eq. (34):

$$\Delta(u_i) = [u_i - f_1 + \sigma_1^2 k^2 (u_i+p)^{1-\alpha}/\eta_1^\alpha](u_i - g_2 + \sigma_2^2 k^2/\eta_2) - g_1 f_2 + \sigma_1^2 k^2 \rho_1^0 (p\eta_1)^{1-\alpha}\frac{h_\alpha(u_i,p)}{u_i\eta_1} \times (A_2 g_1 + A_1 u_i - A_1 g_2 + A_1\sigma_2^2 k^2/\eta_2) = 0, \tag{44}$$

while $\chi(u_i)$ are time-independent coefficients of the exponential time-dependent terms in Eq. (43). The perturbation $\delta\rho_1(k,t)$ grows if at least one of the zeros u_i has a positive real part, else it decays. This holds true for $\delta\rho_2(k,t)$ as well, since the long-time asymptotics are controlled by the zeros of the same polynomial $\Delta(u)$.

An instability occurs if all u_j have negative real parts except for u_{j_c} whose real part changes from negative to positive at the instability. In practice, this instability threshold is crossed as a control parameter of the system is varied; see, e.g., Ref. [34]. If the imaginary part of u_{j_c} is not zero, then the instability is oscillatory. A Turing instability is a stationary instability and u_{j_c} is real. Therefore, the Turing instability threshold corresponds to $\Delta(u_{j_c}\to 0)=0$. Equation (44) provides the Turing condition,

$$[(-f_1+\sigma_1^2k^2p^{1-\alpha}/\eta_1^\alpha)(-g_2+\sigma_2^2k^2/\eta_2)-g_1f_2] + \sigma_1^2k^2\rho_1^0p^{-\alpha}\eta_1^{-\alpha}(\alpha-1)(A_2g_1-A_1g_2+A_1\sigma_2^2k^2/\eta_2)=0. \tag{45}$$

To ensure that the homogeneous steady state is stable against homogeneous perturbations, i.e., the instability occurs indeed with nonzero wavelength, the following conditions must be satisfied as in the case of a standard reaction-diffusion system:

$$f_1+g_2<0, \tag{46}$$

$$f_1g_2-f_2g_1>0. \tag{47}$$

Introducing the generalized diffusion coefficient $K_{\alpha;1}\equiv\sigma_1^2/\eta_1^\alpha$ (see [9]) and the regular diffusion coefficient $D_2\equiv\sigma_2^2/\eta_2$, we write the Turing condition as

$$[(-f_1+K_{\alpha;1}k^2p^{1-\alpha})(-g_2+D_2k^2)-g_1f_2] + [K_{\alpha;1}k^2\rho_1^0p^{-\alpha}(\alpha-1)](A_2g_1-A_1g_2+A_1D_2k^2)=0. \tag{48}$$

Equation (48) is the general condition for the occurrence of a Turing instability in a two-component system when one of the components subdiffuses. The classical Turing condition, for the case when both entities undergo normal diffusion,

$$(-f_1+D_1k^2)(-g_2+D_2k^2)-g_1f_2=0, \tag{49}$$

where $D_1=K_{1;1}=\sigma_1^2/\eta_1$, is obtained by choosing $\alpha=1$ in Eq. (48). (For a derivation of the classical Turing condition for standard reaction-diffusion equations see, e.g., Refs. [34,35].) To be able to compare directly a reaction-subdiffuson system with a standard reaction-diffusion system, we introduce the effective diffusion constant

$$\hat{D}_{\alpha;1}=K_{\alpha;1}p^{1-\alpha}=D_1(p\eta_1)^{1-\alpha} \tag{50}$$

for the subdiffusing species. Then the Turing condition (48) reads

$$[(-f_1+\hat{D}_{\alpha;1}k^2)(-g_2+D_2k^2)-g_1f_2]+[\hat{D}_{\alpha;1}k^2\rho_1^0p^{-1}(\alpha-1)] \times(A_2g_1-A_1g_2+A_1D_2k^2)=0. \tag{51}$$

When the subdiffusion exponent α is close to unity, Eq. (51) reduces to the condition

$$(-f_1+\hat{D}_{\alpha;1}k^2)(-g_2+D_2k^2)-g_1f_2=0. \tag{52}$$

This is formally identical to the classical Turing condition with the effective diffusion coefficient of the subdiffusing species, $\hat{D}_{\alpha;1}$, in place of the original diffusion constant D_1 of the purely diffusive case. Thus for small deviations from purely diffusive behavior into the subdiffusive regime, the memory of the transport shifts the Turing instability threshold and the characteristic wave number of the unstable mode at the threshold. This translates into a change in the characteristic size of the resulting pattern. Also, the upper and lower cutoffs of the band of unstable modes are shifted as seen through the direct solution of Eq. (52).

Equation (52) holds for all values of α if the death rate of the subdiffusing species 1 is of the form $R_1^-(\rho)=p\rho_1$, since in this case $A_1=A_2=0$. The Brusselator [1], $f(\rho_1,\rho_2)=a-(b+1)\rho_1+\rho_1^2\rho_2$, the Gierer-Meinhardt model [36], $f(\rho_1,\rho_2)=1-\rho_1+a\rho_1^2/\rho_2$, and the Schnakenberg model [37], $f(\rho_1,\rho_2)=a-\rho_1+\rho_1^2\rho_2$, belong to this class. Models with a nonlinear death rate for the subdiffusing species will be considered elsewhere [38]. For models with linear death rates it follows from Eq. (52) that the critical value of $d=D_2/\hat{D}_{\alpha;1}$ for a Turing instability to occur is given by

$$d_c=\theta_c. \tag{53}$$

Here θ_c is the critical value of the ratio of diffusion coefficients for a Turing instability in a standard reaction-diffusion system:

$$\theta_c=\left(\frac{1}{f_1}(\sqrt{f_1g_2-f_2g_1}+\sqrt{-f_2g_1})\right)^2. \tag{54}$$

Therefore the diffusion coefficient of the normally diffusing inhibitor must be larger than

$$D_{2,c}=\theta_c\hat{D}_{\alpha;1}=\theta_cD_1(p\eta_1)^{1-\alpha} \tag{55}$$

for a Turing instability to occur if the activator displays subdiffusion with exponent α. Since $p\eta_1$ is a small quantity, subdiffusion of the activator lowers the critical value of the inhibitor diffusion coefficient. The Schnakenberg model with a subdiffusing activator and a normally diffusing inhibitor has been investigated numerically by Weiss [39]. The shifting of the diffusion constant to a new effective value is consistent with his results. Weiss observes the stabilization of Turing patterns in an activator-inhibitor system due to subdiffusion and attributes it to the subdiffusion mimicking the effects of a lower effective diffusion constant. A quantitative comparison between the Turing threshold obtained by Weiss's simulations and our results is not feasible. The simulations are stochastic in nature, and particle number fluctuations have a strong effect. They shift the Turing threshold from the mean field value obtained for the deterministic evolution equations. Our result is valid in the mean field limit and cannot be compared quantitatively to Weiss's simulations.

Rewriting Eq. (48) in the form

$$k^4 + c_2 k^2 + c_4 = 0, \tag{56}$$

with

$$c_2 = \frac{-D_2 f_1 p^\alpha + (\alpha-1)A_2 g_1 K_{\alpha;1}\rho_1^0 - g_2 K_{\alpha;1}[p + (\alpha-1)A_1\rho_1^0]}{D_2 K_{\alpha;1}[p + (\alpha-1)A_1\rho_1^0]} \tag{57}$$

and

$$c_4 = \frac{(f_1 g_2 - f_2 g_1)p^\alpha}{D_2 K_{\alpha;1}[p + (\alpha-1)A_1\rho_1^0]}, \tag{58}$$

furnishes the upper and lower cutoffs of the band of unstable modes,

$$\frac{(-c_2 - \sqrt{c_2^2 - 4c_4})}{2} < k^2 < \frac{(-c_2 + \sqrt{c_2^2 - 4c_4})}{2}, \tag{59}$$

provided $c_2 < 0$, and $c_2^2 - 4c_4 > 0$. At the Turing threshold, $c_2^2 - 4c_4 = 0$, and the critical wave number is $k_c^2 = -c_2/2$.

Our findings regarding the Turing instability in a reaction-subdiffusion system differ considerably from those obtained for a fractional diffusion equation with additive reaction terms as proposed by Henry and Wearne [16,17]. The Turing condition for our reaction-subdiffusion system has a form that closely resembles the classical Turing condition and approaches the latter in a smooth way as $\alpha \to 1$. A second difference is the existence of a lower and an upper cutoff for the band of unstable modes for all values of α, whereas in a system with additive subdiffusion and reaction terms no upper cutoff exists if $0 < \alpha < 1$.

IV. CONCLUSIONS

We have presented a derivation of general reaction-subdiffusion equations with a nonlinear continuous time random walk as the starting point. Unlike previous attempts at combining reaction and subdiffusion processes, where it is assumed at the outset that reactions and subdiffusion are separable, our detailed analysis reveals that memory associated with subdiffusion results in a nonseparable combination of reaction and subdiffusion processes in the kinetic equations governing the evolution of the density of a species. We study Eq. (27) in the context of subdiffusion associated with long tails in $\phi_i(t)$. However, in the derivation of that equation we do not explicitly refer to the particular form of the waiting time PDF. Equation (27) is valid for arbitrary waiting time PDFs $\phi_i(t)$ and has much wider applicability than subdiffusive transport. Potential applications include traveling fronts in reactions-transport systems with memory [40], virus infection fronts [41–43], biological invasions [44], and human population dynamics [45–47]. Note that Vlad and Ross [20] already applied their formalism to the Neolithic transition.

We have presented a linear stability analysis of the homogeneous steady state of the reaction-subdiffusion equations that we derive. The Turing instability is shown to persist in this system. However, we see that the effects of memory not only shift the Turing threshold, but also the upper and lower cutoffs in the band of unstable modes close to the Turing threshold. In contrast to the nonexistence of an upper cutoff in models with separable reaction and subdiffusion [16,17], we do find an upper cutoff of the band of unstable modes. We have shown that subdiffusion of the activator facilitates the formation of Turing patterns if the rate of degradation of the activator is linear. If the activator subdiffuses, then a Turing instability can occur for a smaller critical value of the inhibitor diffusion coefficient. As Weiss [39] already remarked, this fact is likely to be of importance for structure formation in cell biology where examples of subdiffusive transport are abundant.

We emphasize that the Turing condition in the subdiffusive regime may be expressed purely in terms of a generalized diffusion constant, as shown in Eq. (48). Although the generalized diffusion constant $K_{\alpha;1}$ is related to the parameters σ_1 and η_1 of the underlying random walk, it has a broader physical meaning, independent of the microscopic details of the random walk used in its derivation. In practice, one may directly measure the generalized diffusion coefficient [48] and utilize it in Eq. (48) to study the Turing instability in the subdiffusive regime.

Our kinetic equations are easily extended to higher dimensions. Further studies, including detailed numerical simulations of our kinetic equations should provide new insights into effects of memory on other types of patterns: oscillatory instabilities, coherent structures like solitons, fronts, pulses, and spatiotemporal patterns in higher dimensions.

Finally, we have assumed throughout this paper that the spatial jump PDF $\lambda(k)$ is of the form $\lambda(k) = 1 - \sigma^2 k^2$. It is straightforward to extend our derivation to the case of long-range jumps or Lévy flights by choosing $\lambda(k) = 1 - |\sigma k|^\gamma$, $1 < \gamma < 2$.

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APPENDIX: LINEAR STABILITY

Taking the Laplace-Fourier transform of Eq. (29) is straightforward except for terms containing double integrals of the form

$$J = \int_0^t \Theta_1(t-t')e^{-p(t-t')}\left(\int_{t'}^{t} \delta\rho_1(x,t'')dt''\right)dt'. \tag{A1}$$

We rewrite the integral J as

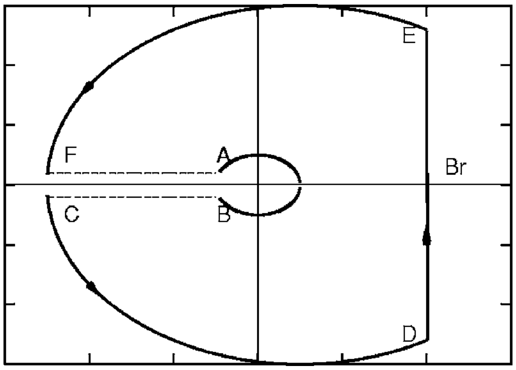

FIG. 2. Contour in the z plane

$$J=\int_0^t\int_0^t \Theta_1(t-t')e^{-p(t-t')}\delta\rho_1(x,t'')dt''dt' - \int_0^t\int_0^{t'}\Theta_1(t-t')e^{-p(t-t')}\delta\rho_1(x,t'')dt''dt' = \left(\int_0^t \Theta_1(t-t')e^{-p(t-t')}dt'\right)\left(\int_0^t \delta\rho_1(x,t'')dt''\right) - \int_0^t \Theta_1(t-t')e^{-p(t-t')}\left(\int_0^{t'}\delta\rho_1(x,t'')dt''\right)dt' \quad \text{(A2)}$$

to obtain a product of two integrals and a convolution on the right hand side of Eq. (A2). The Laplace-Fourier transform of the convolution is easily handled. We concentrate now on evaluating the Laplace transform of the product

$$J_1=\left(\int_0^t \Theta_1(t-t')e^{-p(t-t')}dt'\right)\left(\int_0^t \delta\rho_1(x,t'')dt''\right). \quad \text{(A3)}$$

The Laplace transform of the product of two functions may be evaluated as the convolution of their respective Laplace transforms,

$$L[J_1]=\int_{Br}\frac{\Theta_1(z+p)\delta\rho_1(x,u-z)}{z(u-z)}dz = \int_{Br}\frac{(z+p)\{1-[(z+p)\eta_1]^\alpha\}\delta\rho_1(x,u-z)}{\{[(z+p)\eta_1]^\alpha\}z(u-z)}dz \quad \text{(A4)}$$

where the contour Br lies to the right of $z=0$ in the z plane as shown in Fig. 2. To evaluate the Br line integral, we have constructed a closed contour with a branch point at $z=-p$, and branch cuts in the left half z plane depicted by dashed lines. Such a contour structure is valid for all rational values of α. The poles of $\delta\rho_1(x,u-z)/(u-z)$ lie to the right of the Br contour (see, for example, [32]). It is easy to show that the contributions to the closed contour integral along the arcs EF and CD vanish. Also, the contribution along the circle formed by closing the arc AB vanishes in the limit of an infinitesimal radius. Therefore, the contour integral along Br equals the contributions from the segments FA and BC and the sum of all residues. Thus,

$$L[J_1]=\frac{\delta\rho_1(x,u)[1-(p\eta_1)^\alpha]p}{u[p\eta_1]^\alpha} - \frac{1}{\pi}\int_0^\infty \frac{\eta_1^{-\alpha}y^{1-\alpha}\sin(\pi\alpha)\delta\rho_1(x,u+p+y)}{(p+y)(u+p+y)}dy. \quad \text{(A5)}$$

The first term on the right hand side of Eq. (A5) is the contribution from the residue at $z=0$, while the second term is the combined contribution from the segments FA and BC. One may use Eq. (A5) as it is in obtaining the linear stability properties of the perturbation $\delta\rho_1(x,t)$. However, further simplifications arise by making the following considerations. Inverse Laplace transforming Eq. (A5) provides

$$J_1=\frac{p[1-(p\eta_1)^\alpha]}{[p\eta_1]^\alpha}\int_0^t \delta\rho_1(x,t'')dt'' - \left(\int_0^t \delta\rho_1(x,t'')dt''\right)\times\left(\int_0^\infty \frac{\eta_1^{-\alpha}y^{1-\alpha}\sin(\pi\alpha)e^{-(y+p)t}}{\pi(p+y)}dy\right). \quad \text{(A6)}$$

Clearly, for $0<\alpha<1$,

$$\int_0^\infty \frac{y^{1-\alpha}\sin(\pi\alpha)e^{-(y+p)t}}{\pi(p+y)}dy < \int_0^\infty \frac{y^{1-\alpha}\sin(\pi\alpha)e^{-(y+p)t}}{\pi y}dy = e^{-pt}t^{\alpha-1}\Gamma(1-\alpha)\sin(\pi\alpha)/\pi, \quad \text{(A7)}$$

since $p>0$, and

$$\int_0^\infty \frac{y^{1-\alpha}\sin(\pi\alpha)e^{-(y+p)t}}{\pi(p+y)}dy = e^{-pt}t^{-1}E(-pt)\sin(\pi\alpha)/\pi, \quad \text{(A8)}$$

when $\alpha=1$, where $E(-pt)$ is the exponential integral function. Thus, the first term on the right hand side of Eq. (A5) is the leading term in the long-time asymptotics of J_1. The second term decays faster than the first term in the long-time limit, if $\delta\rho_1(x,t)$ is a decaying function. It grows much slower than the first term, if $\delta\rho_1(x,t)$ grows with increasing time. In light of this discussion, we may neglect the second term in Eq. (A5). Therefore,

$$L[J]=\frac{\delta\rho_1(x,u)[1-(p\eta_1)^\alpha]p}{u[p\eta_1]^\alpha} - \frac{\phi_1(u+p)\delta\rho_1(x,u)}{u} = \frac{\delta\rho_1(x,u)[1-(p\eta_1)^\alpha]p}{u[p\eta_1]^\alpha} - \frac{(u+p)\{1-[(u+p)\eta_1]^\alpha\}\delta\rho_1(x,u)}{u[(u+p)\eta_1]^\alpha}. \quad \text{(A9)}$$

Proceeding with the Fourier-Laplace transform of Eqs. (29), where terms of size $O(k^2[(u+p)\eta_i])$ and greater are neglected as before, we obtain Eq. (31).

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