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Amplitude equations for a sub-diffusive reaction–diffusion system

Y Nec and A A Nepomnyashchy

Department of Mathematics, Technion, Israel Institute of Technology, Haifa, Israel

E-mail: flyby@techunix.technion.ac.il

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Abstract
A sub-diffusive reaction–diffusion system with a positive definite memory operator and a nonlinear reaction term is analysed. Amplitude equations (Ginzburg–Landau type) are derived for short wave (Turing) and long wave (Hopf) bifurcation points.

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1. Introduction

The notion of anomalous sub-diffusion generalizes the property of mean square displacement proportionality to time, typical of Brownian motion, to the power law $\langle r^2 \rangle \sim t^\gamma$ with an exponent $0 < \gamma < 1$. Sub-diffusion characterizes systems where various physical factors impede the free random walk of particles. Basic properties of such systems and their mathematical modelling can be found in [1, 2].

Since the discovery of anomalous sub-diffusion in many biological systems and especially living cells, it has become of importance in analysis of realistic diffusive processes. Anomaly exponents, the measure of diffusion hindrance, have been used to quantify molecular properties such as cytoplasm crowding [3], particle mobility over lattices [4] and within nuclei [5], and even to change prevailing concepts regarding molecular motion [6]. The use of diffusion models accounting for anomaly has proved successful in numerous comparisons with experimental data [7, 8].

One of the pertinent utilities for the description of anomalous diffusion is the continuous time random walk model [1, 2]. According to that model, the molecules are trapped so that the probability of performing a jump depends on the time elapsing since the last jump, i.e. the *age* of the molecule. Actually, the molecule is released from its trap due to some changes in its environment rather than changes in the molecule itself. Introduction of a waiting time distribution of this type resulted in integro-differential memory operators, known as fractional derivatives.

The motion of molecules within a living cell naturally involves chemical reactions: proteins and lipids within cell or organelle membranes bind to receptors, molecules within a cytoplasm bind to enzymes, etc. However, a straightforward attempt to generalize a reaction–diffusion system by the application of a fractional derivative has entailed a not necessarily positive definite evolution operator, i.e. for a positive given initial condition the system might develop an unphysical negative molecule density [9]. Thus, more complicated integro-differential operators are required for proper modelling of reactive systems with anomalous diffusion.

In [10, 11] a model for a linear monomolecular conversion $A \leftrightharpoons B$ subject to diffusion with memory was discussed. Stability analysis of a general linear system of two reagents was given in [12]. A nonlinear system was posed in [14] based on the approach developed in [13] and its linear stability was analysed. An important and often tacit assumption of any reaction model touches on the aging of molecules created due to a reaction event. When a chemical equilibrium is attained, the underlying reactions occur at the rate dictated by such external factors as temperature, etc. yet there are no global changes in the species concentration. The models of [13, 14] assign a zero age to each newborn molecule, thus suppressing diffusion even under conditions of a chemical equilibrium. A different postulate of age ascription in the course of reaction—a proportional distribution of age between newly created molecules—is used in [10–12] with linear kinetics. In the current work the latter approach is adopted for a model involving a positive definite operator and describing a general nonlinear reaction kinetics with sub-diffusion.

The sub-diffusive reaction–diffusion systems, akin to the normal ones, are subject to short wave monotonous (Turing) and long wave oscillatory (Hopf) instability. So far only a linear stability analysis has been performed. Here a weakly nonlinear theory is set for patterns developing due to the aforementioned instabilities. For the first time amplitude equations are derived for both instability types.

2. Mathematical model

A general reaction–diffusion system of n species is considered. It is assumed that a molecule of species i that arrives to the point $\mathbf{r}'$ at the time instant t', performs its next jump to $\mathbf{r}$ at t with the probability density $\psi_i(\mathbf{r} - \mathbf{r}', t - t') = \mathrm{m}_i(\mathbf{r} - \mathbf{r}')W(t - t')$. The jump length distribution $\mathrm{m}_i(\mathbf{r} - \mathbf{r}')$ is species dependent, whereas the waiting time distribution $W(t - t')$ depends solely on the age $\tau = t - t'$ of the molecule performing the jump. It is assumed that the system possesses a certain spatially homogeneous chemical equilibrium state, described by a density vector $\mathbf{N}^0(t - t')$ characterizing the equilibrium distribution of molecules age. The deviations from the equilibrium state are determined by the density vector $\mathbf{n}(\mathbf{r}, t, t')$, the deviation of molecules distribution arriving to $\mathbf{r}$ at the instant t' and remaining there till t. The time evolution of $\mathbf{n}(\mathbf{r}, t, t')$ is affected by two factors: total decrease due to jumps and chemical composition change due to the reaction. Therefore, it is governed by

$$\frac{\partial}{\partial t}\mathbf{n}(\mathbf{r}, t, t') = (-W(t - t')\mathrm{I} + \mathrm{M}(\rho))\mathbf{n}(\mathbf{r}, t, t'), \qquad \mathbf{r} \in \Omega, \quad t > 0, \quad 0 < t' < t \qquad (1a)$$

with vectors denoted by a bold font, matrices—by upright type and scalar quantities—by usual math type. The domain is the whole space or has a rectangular shape $\Omega \subset \mathbb{R}^p$, $p \in \{1, 2, 3\}$. M is a nonlinear kinetics matrix dependent on the total species density deviation (molecules of all ages)

$$\rho(\mathbf{r}, t) = \int_0^t \mathbf{n}(\mathbf{r}, t, t')\,\mathrm{d}t'. \qquad (1b)$$

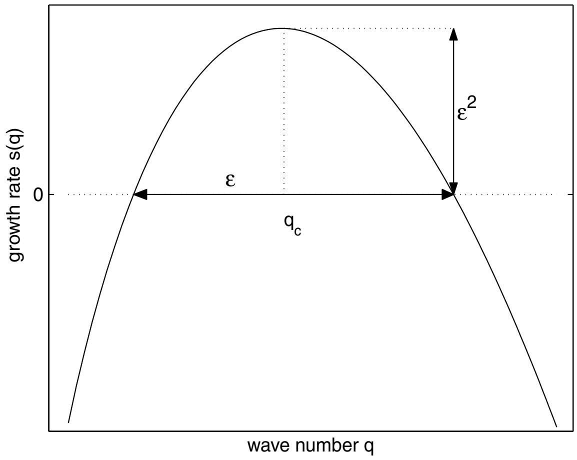

Figure 1. Linear growth rate curve.

Equation (1*a*) describes a system, where uniformly distributed ages are ascribed to molecules produced by a reaction, contrary to the approach taken in [14], where all newborn molecules are of age zero. An initial condition complementing (1*a*) is

$$\mathbf{n}(\mathbf{r}, t, t) = \int_\Omega \mathrm{m}(\mathbf{r} - \mathbf{r}') \int_0^t W(t - t')\mathbf{n}(\mathbf{r}', t, t')\,\mathrm{d}t'\,\mathrm{d}\mathbf{r}', \qquad (1c)$$

where m is a diagonal matrix of a jump length distribution, usually dependent on the distance $\mathbf{r}-\mathbf{r}'$ only and defined by its Fourier transform [15] (Fourier transformed quantities are denoted by a hat and $\mathbf{q}$ is the wave vector)

$$\hat{\mathrm{m}}(q) \sim \mathrm{I} - q^2\sigma \mathrm{D} + o(q^2), \qquad q = |\mathbf{q}|, \qquad (2)$$

with I being an identity matrix, D a diagonal matrix of diffusion coefficients and $\sigma > 0$ a second moment of the jump length distribution (as no asymmetry in particle scattering prevails, the first moment vanishes and σ is the primary characteristic of the jump length distribution function). For $W(t-t')$ a sub-diffusive behaviour is defined in Laplace space (see appendix A):

$$\mathcal{L}\left[W(\tau)\,\mathrm{e}^{-\int_0^\tau W(\tau')\,\mathrm{d}\tau'}\right](s) \sim 1 - \Gamma(1-\gamma)\tau_0^\gamma s^\gamma + o(|s|^\gamma), \qquad 0 < \gamma < 1. \qquad (3)$$

3. Amplitude equations

The linear theory results are reviewed briefly for further clarity of the subsequent nonlinear derivations. A linear problem similar to (1*a*) was analysed in [12], and the dispersion relation derived there was of the form

$$\det(\Gamma(1-\gamma)\tau_0^\gamma(\mathrm{I}s - \mathrm{M}(\mathbf{0}))^\gamma + q^2\sigma\mathrm{D}) = 0 \qquad (4)$$

with $\mathrm{M}(\mathbf{0})$ a constant matrix. In a two species systems

$$\mathrm{D}(d) = \begin{pmatrix} 1 & 0 \\ 0 & d \end{pmatrix}, \qquad (5)$$

the growth rate curve $s(q; d)$ corresponding to (4) was bell shaped (figure 1), and a neutral curve $d = d_u(q)$ existed with $q \sim O(1)$, $d_u > 1$, corresponding to the instability of the Turing type with $s > 0$. Furthermore, pure imaginary roots were proved to ensue if and only if $q = 0$ and $\operatorname{tr} \mathrm{M} = 0$, corresponding to Hopf instability. As is shown hereinafter, system (1) entails an identical dispersion relation, and thus bifurcations of these two types are expected.

3.1. Turing instability: Landau equation

Defining $\boldsymbol{\eta}(\mathbf{r}, t, \tau) = \mathbf{n}(\mathbf{r}, t, t')$, $\tau = t - t'$, (1) becomes

$$\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial \tau}\right) \boldsymbol{\eta}(\mathbf{r}, t, \tau) = (-W(\tau)\mathrm{I} + \mathrm{M}(\boldsymbol{\rho}))\boldsymbol{\eta}(\mathbf{r}, t, \tau), \qquad \mathbf{r} \in \Omega, \quad t > 0, \quad 0 < \tau < t, \tag{6a}$$

$$\boldsymbol{\rho}(\mathbf{r}, t) = \int_0^t \boldsymbol{\eta}(\mathbf{r}, t, \tau)\, \mathrm{d}\tau, \tag{6b}$$

$$\boldsymbol{\eta}(\mathbf{r}, t, 0) = \int_\Omega \mathrm{m}(\mathbf{r} - \mathbf{r}') \int_0^t W(\tau)\boldsymbol{\eta}(\mathbf{r}', t, \tau)\, \mathrm{d}\tau\, \mathrm{d}\mathbf{r}'. \tag{6c}$$

Consider a two species system and choose a wave number $q = q_u$. Take d in the vicinity of the neutral curve $d = d_u(q_u) + \epsilon^2 d_2$, $\epsilon \ll 1$, $d_2 \sim O(1)$ and $s(q_u, d_u) \sim O(\epsilon^2)$. Then

$$\hat{\mathrm{m}} \sim \hat{\mathrm{m}}^{(0)} + \epsilon^2 \hat{\mathrm{m}}^{(2)} + o(\epsilon^2), \qquad \hat{\mathrm{m}}^{(0)} = \mathrm{I} - q^2\sigma \mathrm{D}(d_u), \qquad \hat{\mathrm{m}}^{(2)} = -q^2\sigma \begin{pmatrix} 0 & 0 \\ 0 & d_2 \end{pmatrix}. \tag{7}$$

Near a Turing instability point the characteristic evolution time t will be a slow scale, while the aging process scale τ will remain of the order of unity. The bifurcation parameter thereby defines a slow time scale $t_2 = \epsilon^2 t$ with $\tau \sim O(1)$ and asymptotic expansions

$$\boldsymbol{\eta}(\mathbf{r}, t, \tau) \sim \sum_{j=1}^{\infty} \epsilon^j \boldsymbol{\eta}^{(j)}(\mathbf{r}, t_2, \tau), \qquad \boldsymbol{\eta}^{(j)} \sim O(1) \tag{8a}$$

$$\boldsymbol{\rho}(\mathbf{r}, t_2) \sim \sum_{j=1}^{\infty} \epsilon^j \boldsymbol{\rho}^{(j)}(\mathbf{r}, t_2), \qquad \boldsymbol{\rho}^{(j)} = \int_0^\infty \boldsymbol{\eta}^{(j)}(\mathbf{r}, t_2, \tau)\, \mathrm{d}\tau, \tag{8b}$$

where the upper integration limit t_2/ϵ^2 was replaced by infinity, and bearing in mind that deviations from the equilibrium state are considered,

$$\mathrm{M}_{ij} \sim \mathrm{M}_{ij}|_{\mathbf{0}} + \sum_{k=1}^{n} \left.\frac{\partial \mathrm{M}_{ij}}{\partial \rho_k}\right|_{\mathbf{0}} \rho_k + \frac{1}{2}\sum_{k=1}^{n}\sum_{\ell=1}^{n} \left.\frac{\partial^2 \mathrm{M}_{ij}}{\partial \rho_k \partial \rho_\ell}\right|_{\mathbf{0}} \rho_k \rho_\ell + \cdots \qquad 1 \leqslant i, j \leqslant n \tag{8c}$$

with ρ_k being the kth component of $\boldsymbol{\rho}$. Substituting expansions (8) into (6a),

$$\left(\epsilon^2 \frac{\partial}{\partial t_2} + \frac{\partial}{\partial \tau} + W(\tau)\right) \sum_{j=1}^{\infty} \epsilon^j \boldsymbol{\eta}^{(j)} = \sum_{j=0}^{\infty} \epsilon^j \mathrm{M}^{(j)} \sum_{j=1}^{\infty} \epsilon^j \boldsymbol{\eta}^{(j)}, \tag{9}$$

where by (8b), (8c)

$$\mathrm{M}^{(0)} = \mathrm{M}|_{\mathbf{0}}, \tag{10a}$$

$$\mathrm{M}_{ij}^{(1)} = \sum_{k=1}^{n} \left.\frac{\partial \mathrm{M}_{ij}}{\partial \rho_k}\right|_{\mathbf{0}} \rho_k^{(1)}, \tag{10b}$$

$$\mathrm{M}_{ij}^{(2)} = \sum_{k=1}^{n} \left.\frac{\partial \mathrm{M}_{ij}}{\partial \rho_k}\right|_{\mathbf{0}} \rho_k^{(2)} + \frac{1}{2}\sum_{k=1}^{n}\sum_{\ell=1}^{n} \left.\frac{\partial^2 \mathrm{M}_{ij}}{\partial \rho_k \partial \rho_\ell}\right|_{\mathbf{0}} \rho_k^{(1)}\rho_\ell^{(1)}. \tag{10c}$$

Expanding also the initial condition (6*c*),

$$\sum_{j=1}^{\infty}\epsilon^{j}\boldsymbol{\eta}^{(j)}(\mathbf{r},t_2,0)=\int_{\Omega}(\mathrm{m}^{(0)}(\mathbf{r}-\mathbf{r}')+\epsilon^2\mathrm{m}^{(2)}(\mathbf{r}-\mathbf{r}')+\cdots)$$
$$\times\int_0^{\infty}W(\tau)\sum_{j=1}^{\infty}\epsilon^{j}\boldsymbol{\eta}^{(j)}(\mathbf{r}',t_2,\tau)\,\mathrm{d}\tau\,\mathrm{d}\mathbf{r}'. \tag{11}$$

Extracting the problem of order $O(\epsilon)$ from (9) yields a homogeneous equation

$$(\mathcal{H}\mathrm{I}-\mathrm{M}^{(0)})\boldsymbol{\eta}^{(1)}=\mathbf{0},\qquad \mathcal{H}\stackrel{def}{=}\frac{\partial}{\partial\tau}+W(\tau) \tag{12a}$$

whose solution is

$$\boldsymbol{\eta}^{(1)}(\mathbf{r},t_2,\tau)=\mathrm{e}^{\mathrm{H}(\tau)}\boldsymbol{\eta}^{(1)}(\mathbf{r},t_2,0),\qquad \mathrm{H}(\tau)=-\int_0^{\tau}W(\tau')\,\mathrm{d}\tau'+\mathrm{M}^{(0)}\tau, \tag{12b}$$

proving the positive definiteness of the homogeneous operator. The corresponding initial condition is

$$\boldsymbol{\eta}^{(1)}(\mathbf{r},t_2,0)=\int_{\Omega}\mathrm{m}^{(0)}(\mathbf{r}-\mathbf{r}')\int_0^{\infty}W(\tau)\boldsymbol{\eta}^{(1)}(\mathbf{r}',t_2,\tau)\,\mathrm{d}\tau\,\mathrm{d}\mathbf{r}'. \tag{12c}$$

Substituting (12*b*) into (12*c*) and passing back to $\mathbf{n}(\mathbf{r},t,t')$ with the aid of

$$\mathbf{n}(\mathbf{r},t,t')\sim\sum_{j=1}^{\infty}\epsilon^{j}\mathbf{n}^{(j)}(\mathbf{r},t_2,t_2'),\qquad \mathbf{n}^{(j)}\sim O(1) \tag{13}$$

yields for $t_2>0$

$$\mathbf{n}^{(1)}(\mathbf{r},t_2,t_2)=\int_{\Omega}\mathrm{m}^{(0)}(\mathbf{r}-\mathbf{r}')\int_0^{\infty}W(t_2-t_2')\,\mathrm{e}^{\mathrm{H}(t_2-t_2')}\mathbf{n}^{(1)}(\mathbf{r},t_2',t_2')\,\mathrm{d}t_2'\,\mathrm{d}\mathbf{r}', \tag{14}$$

where the infinite upper integration limit is retained, and $W(t_2-t_2')$ is defined to vanish for any negative argument. Then assuming that $\mathrm{M}^{(0)}$ is diagonalizable, $\exp(\mathrm{M}^{(0)}\tau)=\mathrm{V}\exp(\Lambda\tau)\mathrm{V}^{-1}$ with a constant matrix of the eigenvectors V and diagonal matrix of eigenvalues Λ, combined Fourier and Laplace transform of (14) with respect to the third argument of $\mathbf{n}^{(1)}(\mathbf{r},t_2,t_2)$ yields to leading order in s and q (tilde stands for Laplace transformed quantities)

$$\big(\mathrm{I}-\hat{\mathrm{m}}^{(0)}(q)\big(\mathrm{I}-\Gamma(1-\gamma)\tau_0^{\gamma}(\mathrm{I}s-\mathrm{M}^{(0)})^{\gamma}\big)\big)\hat{\tilde{\mathbf{n}}}^{(1)}(q,t_2,s)=\mathbf{0}. \tag{15}$$

In [12] equation (15) possessed a non-homogeneous right-hand side coming from an additional initial condition for $\mathbf{n}^{(1)}(\mathbf{r},0,0)$. Using (7), the dispersion relation to leading order in s is given by (4). For simplicity the derivation is reduced to one spatial dimension. Thus, for $n=2$ the solution for $\boldsymbol{\eta}^{(1)}$ is

$$\boldsymbol{\eta}^{(1)}(x,t_2,0)=A_1(t_2)\,\mathrm{e}^{\mathrm{i}q_ux}\mathbf{v}+A_1^*(t_2)\,\mathrm{e}^{-\mathrm{i}q_ux}\mathbf{v}^*, \tag{16a}$$

where the asterisk stands for complex conjugation, q_u is the selected wave number and $\mathbf{v}$ is a constant vector. The density vector $\boldsymbol{\rho}^{(1)}$ is of the same form as $\boldsymbol{\eta}^{(1)}$, only with another constant vector

$$\boldsymbol{\rho}^{(1)}(x,t_2)=\int_0^{\infty}\boldsymbol{\eta}^{(1)}(x,t_2,\tau)\,\mathrm{d}\tau=A_1\,\mathrm{e}^{\mathrm{i}q_ux}\mathbf{r}^{(1)}+A_1^*\,\mathrm{e}^{-\mathrm{i}q_ux}\mathbf{r}^{(1)*}. \tag{16b}$$

At order $O(\epsilon^2)$ equation (9) yields a non-homogeneous problem

$$(\mathcal{H}\mathrm{I}-\mathrm{M}^{(0)})\boldsymbol{\eta}^{(2)}=\mathrm{M}^{(1)}\boldsymbol{\eta}^{(1)}=\sum_{\alpha}A_{\alpha}^2\,\mathrm{e}^{\alpha iq_ux}\mathbf{p}_{\alpha}(\tau),\qquad \mathbf{p}_{-\alpha}=\mathbf{p}_{\alpha}^*, \tag{17a}$$

where the right-hand side functional form ensues by combination of entries of the type $\exp(\pm iq_u x)$ in $\mathrm{M}^{(1)}$ with the same terms from $\boldsymbol{\eta}^{(1)}$, i.e. $\alpha = \{2, 0, -2\}$ and respectively $A_\alpha = \{A_1, |A_1|, A_1^*\}$. Then

$$\boldsymbol{\eta}^{(2)}(x, t_2, \tau) = \mathrm{e}^{\mathrm{H}(\tau)} \left(\boldsymbol{\eta}^{(2)}(x, t_2, 0) + \int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau')} \sum_\alpha A_\alpha^2\, \mathrm{e}^{\alpha i q_u x} \mathbf{p}_\alpha(\tau')\, \mathrm{d}\tau' \right), \tag{17b}$$

$$\boldsymbol{\eta}^{(2)}(x, t_2, 0) = \int_\Omega \mathrm{m}^{(0)}(x - x') \int_0^\infty W(\tau) \boldsymbol{\eta}^{(2)}(x', t_2, \tau)\, \mathrm{d}\tau\, \mathrm{d}x'. \tag{17c}$$

Substituting (17b) into (17c), it is seen that the homogeneous part is identical to that of (12c). The problem linearity implies a particular solution of superimposed terms

$$\boldsymbol{\eta}_\alpha^{(2)} = a_\alpha(t_2)\, \mathrm{e}^{\alpha i q_u x} \mathbf{q}_\alpha, \qquad \mathbf{q}_\alpha = \text{const}, \tag{18}$$

upon substitution into (17c) and application of the Fourier transform satisfying

$$a_\alpha \mathbf{q}_\alpha = \hat{\mathrm{m}}^{(0)}(q_u) \int_0^\infty W(\tau)\, \mathrm{e}^{\mathrm{H}(\tau)} \left(a_\alpha \mathbf{q}_\alpha + \int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau)} A_\alpha^2 \mathbf{p}_\alpha(\tau')\, \mathrm{d}\tau' \right) \mathrm{d}\tau. \tag{19}$$

Note that $\mathcal{F}[\mathrm{e}^{\alpha i q_u x}] = \delta(q - \alpha q_u)$, cancelling throughout. A unique solution for $\mathbf{q}_\alpha$ exists if and only if $\alpha \neq 1$. Without loss of generality $a_\alpha = \left\{A_1^2, |A_1|^2, A_1^{*2}\right\}$. Therefore,

$$\boldsymbol{\eta}^{(2)}(x, t_2, 0) = A_2(t_2)\, \mathrm{e}^{\mathrm{i}q_u x} \mathbf{v} + A_2^*(\tau)\, \mathrm{e}^{-\mathrm{i}q_u x} \mathbf{v}^* + \sum_\alpha A_\alpha^2\, \mathrm{e}^{\alpha i q_u x} \mathbf{q}_\alpha, \tag{20a}$$

$$\boldsymbol{\rho}^{(2)}(x, t_2) = A_2\, \mathrm{e}^{\mathrm{i}q_u x} \mathbf{r}^{(2)} + A_2^*(\tau)\, \mathrm{e}^{-\mathrm{i}q_u x} \mathbf{r}^{(2)*} + \sum_\alpha A_\alpha^2\, \mathrm{e}^{\alpha i q_u x} \mathbf{s}_\alpha \tag{20b}$$

with $\mathbf{r}^{(2)}$, $\mathbf{s}_\alpha$ constant vectors. At order $O(\epsilon^3)$ equation (9) gives

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(3)} = -\frac{\partial}{\partial t_2}\boldsymbol{\eta}^{(1)} + \mathrm{M}^{(1)}\boldsymbol{\eta}^{(2)} + \mathrm{M}^{(2)}\boldsymbol{\eta}^{(1)} = \sum_\alpha \mathrm{e}^{\alpha i q_u x} \mathbf{p}_\alpha(\tau), \qquad \mathbf{p}_{-\alpha} = \mathbf{p}_\alpha^* \tag{21a}$$

with $-3 \leqslant \alpha \leqslant 3$ and vectors $\mathbf{p}_\alpha$ different from those in (17a). Then

$$\boldsymbol{\eta}^{(3)}(x, t_2, \tau) = \mathrm{e}^{\mathrm{H}(\tau)} \left(\boldsymbol{\eta}^{(3)}(x, t_2, 0) + \int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau')} \sum_\alpha \mathrm{e}^{\alpha i q_u x} \mathbf{p}_\alpha(\tau')\, \mathrm{d}\tau' \right) \tag{21b}$$

with the initial condition now involving the matrix $\mathrm{m}^{(2)}$

$$\begin{aligned}\boldsymbol{\eta}^{(3)}(x, t_2, 0) = &\int_\Omega \mathrm{m}^{(0)}(x - x') \int_0^\infty W(\tau) \boldsymbol{\eta}^{(3)}(x', t_2, \tau)\, \mathrm{d}\tau\, \mathrm{d}x' \\ &+ \int_\Omega \mathrm{m}^{(2)}(x - x') \int_0^\infty W(\tau) \boldsymbol{\eta}^{(1)}(x', t_2, \tau)\, \mathrm{d}\tau\, \mathrm{d}x'.\end{aligned} \tag{21c}$$

Substitution of (21b) into (21c) and application of the Fourier transform reveal a secular term due to $\mathbf{p}_1$, whose amplitude depends solely on A_1

$$\mathbf{p}_1 = -\frac{\partial A_1}{\partial t_2}\mathbf{c}_\ell(\tau) + A_1|A_1|^2 \mathbf{c}_{n\ell}(\tau), \tag{22}$$

as well as due to the new term in (21c)

$$\mathbf{s} = \hat{\mathrm{m}}^{(0)}(q_u) \left(-\frac{\partial A_1}{\partial t_2}\mathbf{w}_0 + A_1|A_1|^2 \mathbf{c}_0 \right) + \hat{\mathrm{m}}^{(2)}(q_u)(A_1 \mathbf{w}_2). \tag{23}$$

The constant vectors $\mathbf{w}_0, \mathbf{c}_0, \mathbf{w}_2$ should belong to the subspace spanned by the basis of the homogeneous part of (21c) $\{\mathbf{v}\ \ \mathbf{v}^*\}$. Hence $\det([\mathbf{v}\ \ \mathbf{s}]) = 0$ gives the solvability condition in the form of a Landau equation

$$\frac{\partial}{\partial t_2} A_1(t_2) = C_\ell\left(q_u^2\right) A_1 - C_{n\ell}\left(q_u^2\right) A_1 |A_1|^2. \tag{24}$$

The signs of all terms were chosen arbitrarily and $C_\ell, C_{n\ell}$ might bear any sign. The value of C_ℓ is related to $s(q; d)$, the root of (4), as

$$C_\ell = \left(\frac{\partial}{\partial d} s(q; d)\right)_{(q_u, d_u)}. \tag{25}$$

3.2. Turing instability: the Ginzburg–Landau equation

In the previous section the solution spatial periodicity limited the generality in some sense. This limitation can be abated by an arbitrary modulation of the solution wave. The bifurcation pair (q_c, d_c) should correspond to the smallest ratio of diffusion coefficients involving instability, i.e. the curve $s(q; d)$ (root of (4)) must satisfy

$$s(q_c; d_c) = \left(\frac{\partial}{\partial q} s(q; d)\right)_{(q_c, d_c)} = 0. \tag{26}$$

Then a slow spatial scale $x_1 = \epsilon x$ emerges as q_c is a unique wave with zero growth rate at $d = d_c$, and all other waves decay (figure 1). Dependence on x and x_1 in physical domain corresponds to $q = q_c + \epsilon q_1$ in Fourier space. Inclusion of the slow spatial scale affects the initial condition only, so the Fourier transform of (11) is replaced by

$$\sum_{j=1}^{\infty} \epsilon^j \hat{\boldsymbol{\eta}}^{(j)}(q_1, t_2, 0) = \sum_{j=0}^{\infty} \epsilon^j \hat{\mathrm{m}}^{(j)} \int_0^\infty W(\tau) \sum_{j=1}^{\infty} \epsilon^j \hat{\boldsymbol{\eta}}^{(j)}(q_1, t_2, \tau)\, \mathrm{d}\tau \tag{27a}$$

with

$$\hat{\mathrm{m}}^{(0)} = \mathrm{I} - q_c^2 \sigma \mathrm{D}_c, \qquad \hat{\mathrm{m}}^{(1)} = -2 q_c q_1 \sigma \mathrm{D}_c,$$
$$\hat{\mathrm{m}}^{(2)} = -q_1^2 \sigma \mathrm{D}_c - q_c^2 \sigma \begin{pmatrix} 0 & 0 \\ 0 & d_2 \end{pmatrix}, \qquad \mathrm{D}_c = \begin{pmatrix} 1 & 0 \\ 0 & d_c \end{pmatrix}. \tag{27b}$$

At order $O(\epsilon)$ the solution is

$$\boldsymbol{\eta}^{(1)}(x, x_1, t_2, 0) = A_1(x_1, t_2)\, \mathrm{e}^{\mathrm{i} q_c x} \mathbf{v} + A_1^*(x_1, t_2)\, \mathrm{e}^{-\mathrm{i} q_c x} \mathbf{v}^*. \tag{28}$$

At order $O(\epsilon^2)$

$$\boldsymbol{\eta}^{(2)}(x, x_1, t_2, \tau) = \mathrm{e}^{\mathrm{H}(\tau)} \left(\boldsymbol{\eta}^{(2)}(x, x_1, t_2, 0) + \int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau')} \sum_\alpha A_\alpha^2\, \mathrm{e}^{\alpha i q_c x} \mathbf{p}_\alpha(\tau')\, \mathrm{d}\tau'\right) \tag{29a}$$

with the initial condition

$$\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, 0) = \hat{\mathrm{m}}^{(0)} \int_0^\infty W(\tau) \hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, \tau)\, \mathrm{d}\tau + \hat{\mathrm{m}}^{(1)} \int_0^\infty W(\tau) \hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, \tau)\, \mathrm{d}\tau. \tag{29b}$$

The last term in (29b) is not truly secular because the critical point corresponds to the extremum of the growth rate curve (26). For a formal derivation see appendix B. Hence the term $\hat{\mathrm{m}}^{(1)}\boldsymbol{\eta}^{(1)}$ at order $O(\epsilon^2)$ and $\hat{\mathrm{m}}^{(1)}\boldsymbol{\eta}^{(2)}$ at $O(\epsilon^3)$ can be ignored. Now condition (29b) will yield a result identical to (20) with $A_2 = A_2(x_1, t_2)$, $A_\alpha = A_\alpha(x_1, t_2)$.

At order $O(\epsilon^3)$ the equation

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(3)} = -\frac{\partial}{\partial t_2}\boldsymbol{\eta}^{(1)} + \mathrm{M}^{(1)}\boldsymbol{\eta}^{(2)} + \mathrm{M}^{(2)}\boldsymbol{\eta}^{(1)} \tag{30a}$$

is accompanied by the initial condition

$$\hat{\boldsymbol{\eta}}^{(3)}(q_1, t_2, 0) = \hat{\mathrm{m}}^{(0)} \int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(3)}(q_1, t_2, \tau)\,\mathrm{d}\tau + \hat{\mathrm{m}}^{(1)} \int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, \tau)\,\mathrm{d}\tau + \hat{\mathrm{m}}^{(2)} \int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, \tau)\,\mathrm{d}\tau. \qquad (30b)$$

The truly secular terms come from the first harmonic particular solution for $\hat{\boldsymbol{\eta}}^{(3)}$, identical to the Landau equation, and from the last term $\hat{\mathrm{m}}^{(2)}\hat{\boldsymbol{\eta}}^{(1)}$

$$\mathbf{s} = \hat{\mathrm{m}}^{(0)}\left(-\frac{\partial A_1}{\partial t_2}\mathbf{w}_0 + A_1|A_1|^2\mathbf{c}_0\right) + \hat{\mathrm{m}}^{(2)}(A_1\mathbf{w}_2), \qquad (31)$$

only this time $\hat{\mathrm{m}}^{(2)}$ contains terms proportional to q_c^2 as well as q_1^2. Hence $\det([\mathbf{v}\ \ \mathbf{s}])$ yields after the inverse Fourier transform the solvability condition in the form of the Ginzburg–Landau equation

$$\frac{\partial A_1}{\partial t_2} = C_\ell\left(q_c^2\right)A_1 + \tilde{C}_\ell\left(q_c^2\right)\frac{\partial^2 A_1}{\partial x_1^2} - C_{n\ell}\left(q_c^2\right)A_1|A_1|^2. \qquad (32)$$

The constant $\tilde{C}_\ell$ is related to $s(q; d)$, the root of (4), as

$$\tilde{C}_\ell = \frac{1}{2}\left(\frac{\partial^2}{\partial q^2}s(q; d)\right)_{(q_c,d_c)}. \qquad (33)$$

Note that the memory mechanism does not influence the dynamics near the threshold, as the memory scale is $O(1)$, whereas the characteristic time scale is $O(\epsilon^{-2})$.

3.3. Hopf point: the Ginzburg–Landau equation

As mentioned in the preamble of section 3, a different type of bifurcation results if $q_c = 0$ and $\operatorname{tr}\mathrm{M}^{(0)} = 0$, independently of the memory presence. Then the linear growth rate $s(q_c; d_c)$ is pure imaginary rather than zero. Taking the bifurcation parameter $0 < \mu \sim O(1)$ as

$$\mathrm{M}(\boldsymbol{\rho})|_{\rho_0} = \mathrm{M}^{(0)} + \epsilon^2\mathrm{M}_b, \qquad \operatorname{tr}\mathrm{M}^{(0)} = 0, \qquad \mathrm{M}_b = \begin{pmatrix} 0 & 0 \\ 0 & \mu \end{pmatrix}, \qquad (34)$$

the time scales of the problem become $t_0 = t$ and $t_2 = \epsilon^2 t$, corresponding to the linear and modulating waves, and τ for the aging process scale. $x_1 = \epsilon x$ is the slow spatial scale, corresponding to q_1 in Fourier space. Defining

$$\mathbf{n}(x, t, t') = \boldsymbol{\eta}(x_1, t_2, t_0, \tau) = \sum_{j=1}^{\infty} \epsilon^j \boldsymbol{\eta}^{(j)}(x_1, t_2, t_0, \tau), \qquad (35)$$

equation (9) is replaced by

$$\left(\frac{\partial}{\partial t_0} + \frac{\partial}{\partial \tau} + W(\tau) + \epsilon^2\frac{\partial}{\partial t_2}\right)\sum_{j=1}^{\infty}\epsilon^j\boldsymbol{\eta}^{(j)} = \sum_{j=0}^{\infty}\epsilon^j\mathrm{M}_h^{(j)}\sum_{j=1}^{\infty}\epsilon^j\boldsymbol{\eta}^{(j)}, \qquad (36a)$$

where the expressions for $\mathrm{M}_h^{(j)}$ are as in (10*a*)–(10*c*) with t_0 incorporated as an additional variable in

$$\boldsymbol{\rho}(x_1, t_2, t_0) = \int_0^\infty \boldsymbol{\eta}(x_1, t_2, t_0, \tau)\,\mathrm{d}\tau, \qquad (37)$$

and $\mathrm{M}_h^{(2)} = \mathrm{M}^{(2)} + \mathrm{M}_b$. The initial condition is as in (11) with the dependence on t_0 included and bearing in mind the type of bifurcation, $\hat{\mathrm{m}}^{(0)} = \mathrm{I}$, $\hat{\mathrm{m}}^{(2)} = -q_1^2\sigma \mathrm{D}_c$. At order $O(\epsilon)$ the problem is homogeneous

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(1)} = \mathbf{0}, \qquad \mathcal{H} \overset{def}{=} \frac{\partial}{\partial t_0} + \frac{\partial}{\partial \tau} + W(\tau) \tag{38a}$$

Using the transformation of variables $\theta = t_0 - \tau$, $\vartheta = \tau$,

$$(\mathcal{H}_\vartheta \mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(1)}(x_1, t_2, \theta, \vartheta) = \mathbf{0}, \qquad \mathcal{H}_\vartheta \overset{def}{=} \frac{\partial}{\partial \vartheta} + W(\vartheta), \tag{38b}$$

solved as

$$\boldsymbol{\eta}^{(1)}(x_1, t_2, \theta, \vartheta) = \mathrm{e}^{\mathrm{H}(\vartheta)}\boldsymbol{\eta}^{(1)}(x_1, t_2, \theta, 0). \tag{38c}$$

Passing back to (t_0, τ) and substituting into the initial condition,

$$\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, 0) = \hat{\mathrm{m}}^{(0)} \int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, \tau)\,\mathrm{d}\tau. \tag{39}$$

Changing the integration variable to $\zeta = t_0 - \tau$ and noting that $\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, \zeta, 0)$ vanishes for any negative value of ζ,

$$\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, 0) = \hat{\mathrm{m}}^{(0)} \int_0^{t_0} W(t_0 - \zeta)\,\mathrm{e}^{\mathrm{H}(t_0-\zeta)}\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, \zeta, 0)\,\mathrm{d}\zeta. \tag{40}$$

The Laplace transform with respect to t_0 and minor manipulation results in

$$\big(\mathrm{I} - \hat{\mathrm{m}}^{(0)}\big(\mathrm{I} - \Gamma(1-\gamma)\tau_0^\gamma(\mathrm{I}s - \mathrm{M}^{(0)})^\gamma\big)\big)\hat{\tilde{\boldsymbol{\eta}}}^{(1)}(q_1, t_2, s, 0) = \mathbf{0}. \tag{41}$$

Since $\hat{\mathrm{m}}^{(0)} = \mathrm{I}$, the resulting dispersion relation is $\det(\mathrm{I}s - \mathrm{M}^{(0)}) = 0$, along with $\operatorname{tr}\mathrm{M}^{(0)} = 0$ giving $s = \pm\mathrm{i}\omega$, $\omega^2 = \det \mathrm{M}^{(0)}$. Thus

$$\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, 0) = A_1(q_1, t_2)\,\mathrm{e}^{\mathrm{i}\omega t_0}\mathbf{v} + A_1^*\,\mathrm{e}^{-\mathrm{i}\omega t_0}\mathbf{v}^*, \tag{42a}$$

$$\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, \tau) = \mathrm{e}^{\mathrm{H}(\tau)}(A_1\,\mathrm{e}^{\mathrm{i}\omega(t_0-\tau)}\mathbf{v} + A_1^*\,\mathrm{e}^{-\mathrm{i}\omega(t_0-\tau)}\mathbf{v}^*), \tag{42b}$$

$$\hat{\boldsymbol{\rho}}^{(1)}(q_1, t_2, t_0) = \int_0^\infty \hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, \tau)\,\mathrm{d}\tau = A_1\,\mathrm{e}^{\mathrm{i}\omega t_0}\mathbf{r}^{(1)} + A_1^*\,\mathrm{e}^{-\mathrm{i}\omega t_0}\mathbf{r}^{(1)*} \tag{42c}$$

with $\mathbf{v}$, $\mathbf{r}^{(1)}$ constant vectors. Extracting the problem of order $O(\epsilon^2)$,

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(2)} = \mathrm{M}^{(1)}\boldsymbol{\eta}^{(1)} = \sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega t_0}\mathbf{p}_\alpha(\tau), \qquad \mathbf{p}_{-\alpha} = \mathbf{p}_\alpha^*, \tag{43a}$$

where the right-hand side functional form ensues by combination of entries of $\mathrm{M}^{(1)}$ of the type $\exp(\pm\mathrm{i}\omega t_0)$ with the same terms from $\boldsymbol{\eta}^{(1)}$, i.e. $\alpha = \{2, 0, -2\}$ and respectively $A_\alpha = \{A_1, |A_1|, A_1^*\}$. The solution is

$$\boldsymbol{\eta}^{(2)}(x_1, t_2, \theta, \vartheta) = \mathrm{e}^{\mathrm{H}(\vartheta)}\left(\boldsymbol{\eta}^{(2)}(x_1, t_2, \theta, 0) + \int_0^\vartheta \mathrm{e}^{-\mathrm{H}(\vartheta')}\sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega(\theta+\vartheta')}\mathbf{p}_\alpha(\vartheta')\,\mathrm{d}\vartheta'\right). \tag{43b}$$

Returning to the plane (t_0, τ) and substituting (43b) into the initial condition

$$\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, t_0, 0) = \hat{\mathrm{m}}^{(0)} \int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, t_0, \tau)\,\mathrm{d}\tau, \tag{44}$$

yields a homogeneous part identical to the previous order and a non-homogeneous part, solvable by a superposition of

$$\boldsymbol{\eta}_\alpha^{(2)}(q_1, t_2, t_0) = a_\alpha(q_1, t_2)\,\mathrm{e}^{\alpha\mathrm{i}\omega t_0}\mathbf{q}_\alpha, \qquad \mathbf{q}_\alpha = \mathrm{const}, \qquad a_\alpha = A_\alpha^2. \tag{45}$$

Hence

$$\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, t_0, 0) = A_2(q_1, t_2)\,\mathrm{e}^{\mathrm{i}\omega t_0}\mathbf{v} + A_2^*\,\mathrm{e}^{-\mathrm{i}\omega t_0}\mathbf{v}^* + \sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega t_0}\mathbf{q}_\alpha \tag{46a}$$

$$\hat{\boldsymbol{\eta}}^{(2)}(q_1, t_2, t_0, \tau) = \mathrm{e}^{\mathrm{H}(\tau)}\Bigg(A_2\,\mathrm{e}^{\mathrm{i}\omega(t_0-\tau)}\mathbf{v} + A_2^*\,\mathrm{e}^{-\mathrm{i}\omega(t_0-\tau)}\mathbf{v}^* + \sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega(t_0-\tau)}\mathbf{q}_\alpha + \int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau')}\sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega(t_0-\tau+\tau')}\mathbf{p}_\alpha(\tau')\,\mathrm{d}\tau'\Bigg), \tag{46b}$$

$$\hat{\boldsymbol{\rho}}^{(2)}(q_1, t_2, t_0) = A_2\,\mathrm{e}^{\mathrm{i}\omega t_0}\mathbf{r}^{(2)} + A_2^*\,\mathrm{e}^{-\mathrm{i}\omega t_0}\mathbf{r}^{(2)*} + \sum_\alpha A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}\omega t_0}\mathbf{s}_\alpha \tag{46c}$$

with $\mathbf{r}^{(2)}$, $\mathbf{s}_\alpha$ constant vectors. At order $O(\epsilon^3)$

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(3)} = -\frac{\partial}{\partial t_2}\boldsymbol{\eta}^{(1)} + \mathrm{M}^{(1)}\boldsymbol{\eta}^{(2)} + (\mathrm{M}^{(2)} + \mathrm{M}_b)\boldsymbol{\eta}^{(1)} = \sum_\alpha \mathrm{e}^{\alpha\mathrm{i}\omega t_0}\mathbf{p}_\alpha, \qquad \mathbf{p}_{-\alpha} = \mathbf{p}_\alpha^*, \tag{47a}$$

with $-3 \leqslant \alpha \leqslant 3$, solved as

$$\boldsymbol{\eta}^{(3)}(x_1, t_2, \theta, \vartheta) = \mathrm{e}^{\mathrm{H}(\vartheta)}\Bigg(\boldsymbol{\eta}^{(3)}(x_1, t_2, \theta, 0) + \int_0^\vartheta \mathrm{e}^{-\mathrm{H}(\vartheta')}\sum_\alpha \mathrm{e}^{\alpha\mathrm{i}\omega(\theta+\vartheta')}\mathbf{p}_\alpha(\vartheta')\,\mathrm{d}\vartheta'\Bigg). \tag{47b}$$

Returning to the plane (t_0, τ) and substituting (47*b*) into the initial condition

$$\hat{\boldsymbol{\eta}}^{(3)}(q_1, t_2, t_0, 0) = \hat{\mathrm{m}}^{(0)}\int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(3)}(q_1, t_2, t_0, \tau)\,\mathrm{d}\tau + \hat{\mathrm{m}}^{(2)}\int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_2, t_0, \tau)\,\mathrm{d}\tau, \tag{47c}$$

reveals a secular term due to

$$\mathbf{p}_1 = -\frac{\partial A_1}{\partial t_2}\mathbf{c}_{\ell 2}(\tau) + A_1\mathbf{c}_{\ell 1}(\tau) + A_1|A_1|^2\mathbf{c}_{n\ell}(\tau) \tag{48}$$

and the new term in (47*c*)

$$\mathbf{s} = -\frac{\partial A_1}{\partial t_2}\mathbf{w}_0 + A_1\mathbf{w}_1 + A|A_1|^2\mathbf{c}_0 + \hat{\mathrm{m}}^{(2)}(A_1\mathbf{w}_2). \tag{49}$$

As $\hat{\mathrm{m}}^{(2)}$ is proportional to q_1^2, $\det([\mathbf{v}\ \ \mathbf{s}])$ yields, similarly to the monotonous critical point, a solvability condition in the form of the Ginzburg–Landau equation

$$\frac{\partial A_1}{\partial t_2} = C_\ell A_1 + \tilde{C}_\ell\frac{\partial^2 A_1}{\partial x_1^2} - C_{n\ell}A_1|A_1|^2. \tag{50}$$

4. Conclusion

A multiple scales analysis was used to derive amplitude equations for a reaction–sub-diffusion system with general nonlinear kinetics and uniform age distribution between molecules created in the reaction process. The sub-diffusion property was introduced via a positive definite memory operator, ensuing from a basic molecular decay law in the absence of chemical reactions. This approach encompasses a wide variety of systems with an equilibrium characterized by ongoing reactions, yet globally constant species concentrations.

Two types of bifurcation points were considered: a point with short wave monotonous (Turing) and a long wave oscillatory (Hopf) instability. For a Turing point two amplitude equations were derived: one of Landau type for any unstable short wave and one of Ginzburg–Landau type for a unique wave number, corresponding to the smallest ratio of diffusion coefficients entailing instability. For a Hopf point an equation of Ginzburg–Landau type was derived. The presence of anomaly neither entailed qualitative changes in the equation form nor introduced any memory effects into the system dynamics near the threshold due to a temporal scale separation between the $O(1)$ memory effect and $O(\epsilon^{-2})$ perturbation evolution. Quantitative changes in both linear and nonlinear equation coefficients are expected.

An intriguing phenomenon reported recently is an increase of noise during measurements at a threshold and consequently a broader parameter estimation uncertainty: near the percolation limit [7] and due to a hierarchy of binding sites within a nucleus [8]. Such results imply an approach of an instability threshold, rendering the weakly nonlinear analysis of special interest. With future advance in experimental visualization techniques the amplitude equations may be applied to explain and predict pattern formation in reaction–sub-diffusion systems. The normal form of the equations implies that the sub-diffusion by itself does not change the universal behaviour of the system near a bifurcation point.

A system where the memory and slow characteristic time scales are of the same order of magnitude remains a topic for future research. There the presence of memory should affect the form of the amplitude equations.

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Appendix A. Functional form of time waiting distribution

In order to determine correctly the functional dependence of the waiting time distribution $W(t-t')$, equation (1*a*) in the absence of reaction

$$\frac{\partial}{\partial t} n(x,t,t') = -W(t-t')n(x,t,t'), \tag{A.1a}$$

will be compared with the more common postulate of decay

$$\frac{\partial}{\partial t} n(x,t,t') = -w(t-t')n(x,t',t'). \tag{A.1b}$$

Equivalence of the two equations yields

$$w(t-t') = W(t-t')\,\mathrm{e}^{-\int_{t'}^{t} W(y-t')\,\mathrm{d}y} \tag{A.2a}$$

and

$$w(t-t') = W(t-t')\left(1 - \int_{t'}^{t} w(y-t')\,\mathrm{d}y\right). \tag{A.2b}$$

Defining $\tau = t - t'$,

$$w(\tau)/W(\tau) = 1 - \int_0^{\tau} w(y)\,\mathrm{d}y. \tag{A.3}$$

The Laplace transform gives

$$\mathcal{L}[w/W](s) = (1-\tilde{w})/s. \tag{A.4}$$

The decay of w in the case of sub-diffusion is known

$$\tilde{w} \sim 1 - \Gamma(1-\gamma)\tau_0^{\gamma} s^{\gamma} + o(|s|^{\gamma}), \tag{A.5}$$

where Γ denotes the gamma function and τ_0 is a characteristic decay time (see also [15]). Hence

$$\mathcal{L}[w/W](s) \sim \Gamma(1-\gamma)\tau_0^{\gamma} s^{\gamma-1} + o(|s|^{\gamma-1}). \tag{A.6}$$

By the identity

$$\int_0^\infty \mathrm{e}^{-s\tau}\tau^{\alpha-1}\,\mathrm{d}\tau = s^{-\alpha}\Gamma(\alpha), \qquad \alpha > 0 \tag{A.7}$$

with $\alpha = 1-\gamma$

$$W(\tau) \sim w(\tau)\left(\frac{\tau}{\tau_0}\right)^{\gamma}, \qquad \tau \gg 1. \tag{A.8}$$

Substituting into (A.3), cancelling $w(\tau)$ (that never vanishes) and differentiating gives the decay law of both functions

$$w(\tau) \sim \frac{\gamma\tau_0^{\gamma}}{\tau^{\gamma+1}}, \qquad W(\tau) \sim \frac{\gamma}{\tau}, \qquad \tau \gg 1. \tag{A.9}$$

Appendix B. Ostensible secularity in the Ginzburg–Landau equation

In order to show formally that certain terms of secular nature, appearing in the course of the derivation of the Ginzburg–Landau equation in the vicinity of a Turing bifurcation point, are in fact superfluous, an intermediate time scale $t_1 = \epsilon t$ is introduced in addition to the usual slow scale $t_2 = \epsilon^2 t$. The governing equation (9) then becomes

$$\left(\epsilon\frac{\partial}{\partial t_1} + \epsilon^2\frac{\partial}{\partial t_2} + \frac{\partial}{\partial\tau} + W(\tau)\right)\sum_{j=1}^{\infty}\epsilon^j\boldsymbol{\eta}^{(j)} = \sum_{j=0}^{\infty}\epsilon^j\mathrm{M}^{(j)}\sum_{j=1}^{\infty}\epsilon^j\boldsymbol{\eta}^{(j)},$$

$$\boldsymbol{\eta}^{(j)} = \boldsymbol{\eta}^{(j)}(x, x_1, t_1, t_2, \tau), \tag{B.1a}$$

complemented by the initial condition

$$\sum_{j=1}^{\infty}\epsilon^j\hat{\boldsymbol{\eta}}^{(j)}(q_1, t_1, t_2, 0) = \sum_{j=0}^{\infty}\epsilon^j\hat{\mathrm{m}}^{(j)}\int_0^\infty W(\tau)\sum_{j=1}^{\infty}\epsilon^j\hat{\boldsymbol{\eta}}^{(j)}(q_1, t_1, t_2, \tau)\,\mathrm{d}\tau. \tag{B.1b}$$

At order $O(\epsilon)$ a homogeneous equation ensues

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(1)} = \mathbf{0}, \tag{B.2a}$$

$$\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_1, t_2, 0) = \hat{\mathrm{m}}^{(0)}\int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(1)}(q_1, t_1, t_2, \tau)\,\mathrm{d}\tau, \tag{B.2b}$$

entailing results analogous to (12) and (16), i.e.

$$\boldsymbol{\eta}^{(1)}(x, x_1, t_1, t_2, \tau) = \mathrm{e}^{\mathrm{H}(\tau)}\boldsymbol{\eta}^{(1)}(x, x_1, t_1, t_2, 0), \tag{B.3a}$$

$$\boldsymbol{\eta}^{(1)}(x, x_1, t_1, t_2, 0) = A_1(x_1, t_1, t_2)\,\mathrm{e}^{\mathrm{i}q_c x}\mathbf{v} + A_1 * (x_1, t_1, t_2)\,\mathrm{e}^{-\mathrm{i}q_c x}\mathbf{v}^*. \tag{B.3b}$$

At order $O(\epsilon^2)$ a non-homogeneous equation is obtained

$$(\mathcal{H}\mathrm{I} - \mathrm{M}^{(0)})\boldsymbol{\eta}^{(1)} = -\frac{\partial A_1}{\partial t_1}\,\mathrm{e}^{\mathrm{i}q_c x}\,\mathrm{e}^{\mathrm{H}(\tau)}\mathbf{v} - \frac{\partial A_1^*}{\partial t_1}\,\mathrm{e}^{-\mathrm{i}q_c x}\,\mathrm{e}^{\mathrm{H}(\tau)}\mathbf{v}^* + \sum_{\alpha} A_\alpha^2\,\mathrm{e}^{\alpha\mathrm{i}q_c x}\mathbf{p}_\alpha(\tau), \tag{B.4a}$$

$$\hat{\boldsymbol{\eta}}^{(2)}(q_1,t_1,t_2,0)=\hat{\mathrm{m}}^{(0)}\int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(2)}(q_1,t_1,t_2,\tau)\,\mathrm{d}\tau+\hat{\mathrm{m}}^{(1)}\int_0^\infty W(\tau)\hat{\boldsymbol{\eta}}^{(1)}(q_1,t_1,t_2,\tau)\,\mathrm{d}\tau, \tag{B.4b}$$

with $\mathbf{p}_\alpha$ and A_α identical to (17*a*). The solution of (B.4*a*) is given by

$$\boldsymbol{\eta}^{(2)}(x,x_1,t_1,t_2,\tau)=\mathrm{e}^{\mathrm{H}(\tau)}\left\{\boldsymbol{\eta}^{(2)}(x,x_1,t_1,t_2,0)+\int_0^\tau \mathrm{e}^{-\mathrm{H}(\tau')}\left(\sum_\alpha A_\alpha^2\mathrm{e}^{\alpha i q_c x}\mathbf{p}_\alpha(\tau')\right.\right.$$
$$\left.\left.-\frac{\partial A_1}{\partial t_1}\mathrm{e}^{\mathrm{i}q_c x}\,\mathrm{e}^{\mathrm{H}(\tau')}\mathbf{v}-\frac{\partial A_1^*}{\partial t_1}\mathrm{e}^{-\mathrm{i}q_c x}\,\mathrm{e}^{\mathrm{H}(\tau')}\mathbf{v}^*\right)\mathrm{d}\tau'\right\}, \tag{B.5}$$

therefore the secular term in (B.4*b*) is

$$\mathbf{s}=-\hat{\mathrm{m}}^{(0)}\frac{\partial A_1}{\partial t_1}\int_0^\infty \tau W(\tau)\mathbf{v}\,\mathrm{d}\tau+\hat{\mathrm{m}}^{(1)}A_1\int_0^\infty W(\tau)\,\mathrm{e}^{\mathrm{H}(\tau)}\mathbf{v}\,\mathrm{d}\tau. \tag{B.6}$$

As $\mathrm{m}^{(1)}$ is proportional to q_1, the solvability condition $\det([\mathbf{v}\quad\mathbf{s}])=0$ leads to

$$\frac{\partial A_1}{\partial t_1}=\mathcal{C}_\ell(q_c^2)q_1A_1 \tag{B.7}$$

or after inverse Fourier transform

$$\frac{\partial A_1}{\partial t_1}=\mathcal{C}_\ell(q_c^2)\frac{\partial A_1}{\partial x_1}. \tag{B.8}$$

The coefficient $\mathcal{C}_\ell$ is related to $s(q;d)$, the root of (4), as

$$\mathcal{C}_\ell\propto\left(\frac{\partial}{\partial q}\operatorname{Re}s(q;d)\right)_{(q_c,d_c)}=0. \tag{B.9}$$

Thus all terms containing the matrix $\mathrm{m}^{(1)}$ in the derivation without the scale t_1 (section 3.2) entail no true secularity, but imply that A_1 does not depend on t_1.

References

[1] Klafter J, Zumofen G and Shlesinger M F 1997 *The Physics of Complex Systems* ed F Mallamace and H E Stanley (Amsterdam: IOS Press)

[2] Metzler R and Klafter J 2000 The random walk's guide to anomalous diffusion: a fractional dynamics approach *Phys. Rep.* **339** 1–77

[3] Weiss M, Elsner M, Kartberg F and Nilsson T 2004 Anomalous subdiffusion is a measure for cytoplasmic crowding in living cells *Biophys. J.* **87** 3518–24

[4] Wong I Y, Gardel M L, Reichman D T, Weeks E R, Valentine M T, Bausch A R and Weitz D A 2004 Anomalous diffusion probes microstructure dynamics of entangled F-actin networks *Phys. Rev. Lett.* **92** 178101

[5] Wachsmuth M, Waldeck W and Langowski J 2000 Anomalous diffusion of fluorescent probes inside living cell nuclei investigated by spatially resolved fluorescence *J. Mol. Biol.* **298** 677–89

[6] Feder T J, Brust-Mascher I, Slattery J P, Baird B and Webb W W 1996 Constrained diffusion or immobile fraction on cell surfaces: a new interpretation *Biophys. J.* **70** 2767–73

[7] Saxton M J 2001 Anomalous subdiffusion in fluorescence photobleaching recovery: a Monte Carlo study *Biophys. J.* **81** 2226–40

[8] Seksek O, Biwersi J and Verkman A S 1997 Translational diffusion in macromolecule-sized solutes in cytoplasm and nucleus *J. Cell. Biol.* **138** 131–42

[9] Henry B I, Langlands T A M and Wearne S L 2006 Anomalous diffusion with linear reaction dynamics: from continuous time random walks to fractional reaction-diffusion equations *Phys. Rev.* E **74** 031116

[10] Sokolov I M, Schmidt M G W and Sagués F 2006 Reaction-subdiffusion equations *Phys. Rev.* E **73** 031102

[11] Schmidt M G W, Sagués F and Sokolov I M 2007 Mesoscopic description of reactions for anomalous diffusion: a case study *J. Phys.: Condens. Matter* **19** 065118

[12] Nec Y and Nepomnyashchy A A 2007 Turing instability in sub-diffusive reaction–diffusion systems *J. Phys. A: Math. Theor.* **40** 14687–702

[13] Vlad M O and Ross J 2002 Systematic derivation of reaction-diffusion equations with distributed delays and relations to fractional reaction-diffusion equations and hyperbolic transport equations: application to the theory of neolithic transition *Phys. Rev.* E **66** 061908

[14] Yadav A and Horsthemke W 2006 Kinetic equations for reaction-subdiffusion systems: derivation and stability analysis *Phys. Rev.* E **74** 066118

[15] Henry B I and Wearne S L 2000 Fractional reaction—diffusion *Physica* A **276** 448