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Stochastic escape processes from a non-symmetric potential normal form III: extended explosive systems

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Abstract. The stochastic dynamics to reach the hot attractor of an explosive extended system is analytically studied by using a previously reported scheme: *The stochastic path perturbation approach* (Cáceres M O, Budde C E and Sibona G J 1995 *J. Phys. A: Math. Gen.* **28** 3877). A perturbation theory in the small noise parameter is introduced to analyse the random escape of the stochastic field from criticality. The anomalous fluctuations of the order parameter (the temperature profile) are calculated analytically using an instanton-like approximation. Emphasis is placed on a thermal non-homogeneous explosion in order to exemplify a system undergoing hysteresis in a first-order non-equilibrium phase transition. Concerning the stochastic propagation of the flame front we have carried out Monte Carlo simulations showing good agreement with our theoretical predictions.

1. Introduction

In order to investigate the existence of new physical solutions which emerge beyond the threshold of instability, nonlinear contributions—coming from the full equations of motion—must be taken into account [1]. As a matter of fact, all dynamical systems undergoing a bifurcation, can be described in a *reduced* subspace, the so-called *centre manifold*—to some dominant order—in the vicinity of the bifurcation by its *normal form* [2]. Among the several *universal* normal forms that can occur in nature, those breaking the reflection symmetry—in their associated potentials—are frequently encountered when two fixed points coalesce for a given value of the control parameter. This situation is typically what happens at the limit point in a hysteresis cycle.

In previous papers we were particularly interested in *limit point* bifurcations because in a well stirred chemical reactor, the Semenov model [1] reduces—in its centre manifold—to that class of *universal* normal form [3, 4]. A thermal explosion in a closed vessel in the limit of high activation energy [5], is an excellent example of a physicochemical system dealing with a process involving two timescales: a slow induction period followed by a slow saturation characteristic of the final approach toward the *stable* attractor. In the context of homogeneous thermochemical explosive systems, the order parameter is the temperature and the matching time between these two slow regimes is around the *random* ignition time [5–7]; similar situations can also occur in the analysis of rockets [8], and in reaction–diffusion processes [9].

The *normal form* analysis for explosive systems has also been made, in the past, and from different points of view: (a.1) in a deterministic approach (ignoring reactant consumption) for

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non-homogenous reactor tanks [10]; (a.2) for well stirred chemical reactors (homogeneous systems) [3]; (b) from a stochastic point of view and considering the coupling between the concentration and the thermal variable, in the homogenous [11] and in the non-homogeneous case [12]. In the latter, it has been shown that the coupling of these two-order parameters leads to the possibility of a transition towards a multiple steady state regime (cusp bifurcation). In particular, for the inhomogeneous case it has been proved that 'adiabatic elimination' reduces the problem to a 'critical' field characterized by a Landau–Ginzburg potential [12]. Nevertheless, Tirapegui and van den Broeck have emphasized that this 'reduction' is non-trivial and depends strongly on the dimensionality of the problem. The significance of the stochastic nature of the ignition process was first pointed out by Baras *et al* [5], and since then several works have been done in that field. In particular, for the homogeneous case, we have pointed out that the timescale characterizing the escape from the instability is the *lifetime* of the unstable state calculated as the mean first-passage time (MFPT) [4]. Using the stochastic path perturbation approach (SPPA), we have been able to develop a perturbation theory to calculate—analytically—the first-passage time distribution (FPTD) [13, 14]. One of the goals of this paper is to generalize that theory to spatially distributed systems. In particular, we will apply the SPPA to the analysis of the stochastic ignition in a non-homogeneous reactor.

Related theoretical studies—on the analysis of the stochastic ignition problem—for distributed systems without consumption, have also been done from the point of view of the Kramer–Langer thermal activation time [15]. We emphasize that Fedotov's approach is related to a generalized Kramer's activation from a given—extended—metastable state over a saddle point. This is not the purpose of our paper; here we study the lifetime from the critical unstable—extended—state, and we characterize its dynamics toward the stable extended attractor. Thus we assume that the system is started in a range of initial conditions and parameter values close to the limit-point bifurcation, i.e. at criticality. Therefore and because the only attractor available (the hot temperature profile) is far from the initial condition, a sudden jump will occur removing the system from criticality. Interestingly the SPPA gives us the possibility to study this evolution *analytically*, and then the anomalous fluctuations will be characterized and compared with Monte Carlo simulations.

In order to make the paper self-contained we have organized it in the following way. In section 2 we calculate the *normal form*—near the critical point—starting from the Frank–Kamenetskii thermochemical equation (*the equation of motion* for our single-order parameter: the temperature profile), to model a non-homogeneous reactor without consumption. In section 3 we develop—up to the dominant order in the small noise parameter—the SPPA for *extended systems* for a saddle-node normal form. This SPPA allows us to calculate, in the small noise approximation, the FPTD for each relevant Fourier mode of the critical field, i.e. the lifetime of the unstable Fourier components. The transient fluctuations of the critical field are also studied introducing an instanton-like approximation. In section 4 we present a general discussion and the conclusions concerning our future research program. Some detailed calculations of the steady state analysis of the Frank–Kamenetskii model are in appendix A. Appendix B is concerned with the FPTD involved in the characterization of the homogeneous mode. Appendix C deals with the analysis of space-averages, and finally appendix D with the Monte Carlo simulations.

2. The normal form analysis

2.1. Frank–Kamenetskii model for a thermochemical explosion

The Frank–Kamenetskii model results from considering a non-homogeneous chemical reactor, in mechanical equilibrium, closed to mass transfer but capable of exchanging energy with a thermal reservoir at constant temperature T_a. The chemical transformation taking place within the reactor is an irreversible process, which is assumed—in its simplest description—by a unimolecular exothermic decomposition: *fuel*+*oxygen* $\xrightarrow{k}$ *oxide*+*heat*, where the rate constant $k \equiv k(T)$ is an increasing function of the temperature [8]. If for simplicity one assumes that the concentration of the reactant varies on a scale that is much slower than heat transfer, this concentration can be taken as a constant c_0. Thus the relevant variable is the temperature profile T. From the energy balance, the temperature profile $T(\tilde{x}, \tilde{t})$ fulfils the dynamical equation [1] (in one dimension†)

$$\sigma c_v \frac{\partial}{\partial \tilde{t}} T(\tilde{x}, \tilde{t}) = Q c_0 k(T) + \kappa \frac{\partial^2}{\partial \tilde{x}^2} T(\tilde{x}, \tilde{t}) \tag{2.1}$$

where κ is the thermic conductivity of the reactant, σ is the mass density of the mixture, c_v the specific heat at constant volume, Q the heat of reaction, and $k(T)$ gives the temperature dependence of the velocity of reaction. This is the Frank–Kamenetskii equation which gives rise to a propagating flame front. If the reactor is well stirred, the diffusion term is replaced by a Newton's cooling law and the balance equation turns out to be the Semenov model, which we have studied before in the stochastic context [4, 13]. Equation (2.1) must be solved under the boundary conditions $T(\pm L, \tilde{t}) = T_a$ ($2L$ is the length of the one-dimensional reactor), where T_a is the temperature of the reservoir. Introducing the adimensional transformations

$$\theta = \frac{(T - T_a)U}{RT_a^2} \qquad \rho = \tilde{x}/L \qquad \tau = \frac{\kappa}{\sigma c_v L^2}\tilde{t} \tag{2.2}$$

where R is the gas constant and U is an activation energy (for example, if we use the Arrhenius rate model we have $k(T) = k_0 \mathrm{e}^{-U/RT}$), it is possible to rewrite (2.1) in a simpler form in terms of the adimensional temperature profile $\theta(\rho, \tau)$

$$\frac{\partial}{\partial \tau}\theta = \frac{\partial^2}{\partial \rho^2}\theta + \delta f(\theta) \qquad -1 \leqslant \rho \leqslant 1. \tag{2.3}$$

Thus the boundary conditions are now $\theta(\pm 1, \tau) = 0$. In (2.3) $f(\theta)$ is an arbitrary function representing the adimensional law for the rate constant $k(T)$, i.e. $f(\theta) = \mathrm{e}^{\theta}$ for the exponential model, $f(\theta) = \exp[\theta/(1 + E\theta)]$ for the Arrhenius model (where $E \equiv RT_a/U$), $f(\theta) = p + q\theta + r\theta^2$ for the quadratic approximation, etc. In (2.3) δ is the adimensional Kamenetskii control parameter

$$\delta = QUL^2 k_0 c_0 \frac{\exp(-U/RT_a)}{\kappa R T_a^2}. \tag{2.4}$$

From (2.3) and a typical $f(\theta)$ it is possible to see that depending on the Kamenetskii parameter δ there will coexist *non-homogeneous* stable and unstable stationary solutions. In particular there exists a critical value of the control parameter, $\delta = \delta_c$, where the phase space has a limit point. Thus the value of the control parameter δ leads to a bifurcation scenario in the phase space of the stationary field $\theta_{St}(\rho)$. In order to clarify this issue we have analysed the stationary solutions of (2.3) for the particular case when $f(\theta)$ is approximated by a *piecewise*

† In the general case the additional term $\frac{\kappa j}{\tilde{x}} \frac{\partial}{\partial \tilde{x}} T(\tilde{x}, \tilde{t})$, $j = 0, 1, 2$ for the infinite slab, infinite cylinder and sphere respectively, should be considered. Hence in this paper we only consider $j = 0$ which leads to a simpler analysis.

linear function $f(\theta)$, see appendix A. We note that the results of our paper are not based on any approximation concerning the nonlinear function $f(\theta)$. The stochastic evolution of the flame front will be studied on the basis of the normal form analysis, which of course is universal in the sense that its form is independent of the detailed structure of the underlying model $f(\theta)$. The structure of the function $f(\theta)$ only enters through renormalized coefficients in the *normal form* near the critical point (θ_c, δ_c).

2.2. The normal form near the critical point

In order to obtain the relevant dynamics in the neighbourhood of the point (θ_c, δ_c), we now introduce a multiple-scale transformation around this critical point (for a detailed analysis of the coalescence of the two stationary branches at the value δ_c see appendix A)

$$\frac{\partial}{\partial\rho} \to \frac{\partial}{\partial x} + \lambda^{1/4}\frac{\partial}{\partial x_1} \qquad \frac{\partial}{\partial\tau} \to \lambda^{1/2}\frac{\partial}{\partial t} \tag{2.5}$$

where $\lambda = \delta/\delta_c - 1 \geqslant 0$ measures the departure from the critical value δ_c. Thus, assuming an expansion in λ for the temperature profile

$$\theta = \theta_0 + \sqrt{\lambda}\theta_1 + \lambda\theta_2 + \cdots \tag{2.6}$$

where each term fulfils the boundary conditions $\theta_j(\pm 1, t) = 0$, $\forall j = 0, 1, 2, \ldots,$ and introducing an amplitude ϕ in the form

$$\theta_0 = \theta_c(x) \qquad \theta_1 = \phi(x_1, t)u_0(x) \qquad \text{etc} \tag{2.7}$$

we obtain an equation which can be analysed in increasing order in powers of λ. From (2.7) we see that $\theta_0(x)$ is a stationary solution, and $\theta_1(x, x_1, t)$ has been written as the product of an unknown profile $u_0(x)$ times the amplitude ϕ depending on the slow variables x_1 which grow in the slow time variable t. We remark that the present analysis is independent of the detailed structure of $f(\theta)$. Using (2.5)–(2.7) in (2.3) we obtain

$$\begin{aligned}\sqrt{\lambda}\left[\frac{\partial}{\partial t}\theta_0 + \sqrt{\lambda}\frac{\partial}{\partial t}\theta_1 + \lambda\frac{\partial}{\partial t}\theta_2 + \cdots\right] &= \frac{\partial^2}{\partial x^2}\theta_0 + \sqrt{\lambda}\frac{\partial^2}{\partial x^2}\theta_1 + \lambda\frac{\partial^2}{\partial x^2}\theta_2 + \cdots \\ &\quad + \sqrt{\lambda}\left[\frac{\partial^2}{\partial x_1^2}\theta_0 + \sqrt{\lambda}\frac{\partial^2}{\partial x_1^2}\theta_1 + \lambda\frac{\partial^2}{\partial x_1^2}\theta_2\right] + \cdots \\ &\quad + \delta_c(1+\lambda)[f_0 + f_1(\sqrt{\lambda}\theta_1 + \lambda\theta_2 + \cdots) + \tfrac{1}{2}f_2\lambda\theta_1^2 + f_2\theta_1\theta_2\lambda^{3/2} + \mathrm{O}(\lambda^2)]\end{aligned} \tag{2.8}$$

where we have used the notation

$$f_n = \left(\frac{\partial^n}{\partial\theta^n}f\right)_{\theta=\theta_c}. \tag{2.9}$$

Collecting terms of the same order in λ we get for $\mathcal{O}(\lambda^0)$

$$\frac{\partial^2}{\partial x^2}\theta_0 + \delta_c f_0 = 0. \tag{2.10}$$

For $\mathcal{O}(\sqrt{\lambda})$ we obtain

$$\frac{\partial}{\partial t}\theta_0 = \frac{\partial^2}{\partial x^2}\theta_1 + \frac{\partial^2}{\partial x_1^2}\theta_0 + \delta_c f_1\theta_1.$$

Nevertheless, using that

$$\frac{\partial}{\partial t}\theta_0 = \frac{\partial^2}{\partial x_1^2}\theta_0 = 0 \tag{2.11}$$

and that $\theta_1 = \phi(x_1, t)u_0(x)$, we find that $u_0(x)$ is given by the solution of

$$\frac{\mathrm{d}^2}{\mathrm{d}x^2}u_0 = -\delta_c f_1 u_0. \tag{2.12}$$

Finally, for $\mathcal{O}(\lambda)$ we get

$$\frac{\partial}{\partial t}\theta_1 = \frac{\partial^2}{\partial x^2}\theta_2 + \frac{\partial^2}{\partial x_1^2}\theta_1 + \delta_c\left(f_1\theta_2 + \frac{f_2\theta_1^2}{2} + f_0\right). \tag{2.13}$$

The solution of (2.10) is the stationary profile at the critical value δ_c. The solution of (2.12) with the boundary condition $u_0(\pm 1) = 0$ is the so called Kordylewski profile, which exists by virtue of the definition of criticality [16]. Multiplying (2.13) by $u_0(x)$ and integrating over the domain $\mathcal{D} \equiv [-1, 1]$, and due to the fact that

$$\int_{\mathcal{D}} u_0\left(\frac{\partial^2}{\partial x^2}\theta_2 + \delta_c f_1\theta_2\right)\mathrm{d}x = 0 \tag{2.14}$$

we get that the amplitude $\phi(x_1, t)$ satisfies a closed equation. Thus, going back to the old dimensional variables $(\tilde{x}, \tilde{t})$ (see (2.1)) the evolution of the amplitude ϕ is governed by

$$\frac{\partial}{\partial\tilde{t}}\phi(\tilde{x}, \tilde{t}) = D\frac{\partial^2}{\partial\tilde{x}^2}\phi(\tilde{x}, \tilde{t}) + a\phi(\tilde{x}, \tilde{t})^2 + b \qquad D > 0 \quad a > 0 \quad b > 0 \tag{2.15}$$

where $\tilde{x} \in [-L, L]$ and

$$D = \frac{\kappa}{\sigma c_v} \qquad a = \frac{\kappa\delta_c\sqrt{\lambda}}{2\sigma c_v L^2}\frac{\int_{\mathcal{D}} f_2 u_0^3\,\mathrm{d}x}{\int_{\mathcal{D}} u_0^2\,\mathrm{d}x} \qquad b = \frac{\kappa\delta_c\sqrt{\lambda}}{\sigma c_v L^2}\frac{\int_{\mathcal{D}} f_0 u_0\,\mathrm{d}x}{\int_{\mathcal{D}} u_0^2\,\mathrm{d}x}. \tag{2.16}$$

In what follows we will investigate the stochastic version of the amplitude equation (2.15).

The amplitude equation (2.15) represents the growth at criticality, this is so because if $\lambda \neq 0$ but small, the time-dependent profile (the flame front) is characterized by the contribution $\sim\sqrt{\lambda}\theta_1 = \sqrt{\lambda}\phi u_0$. Therefore the following question arises: due to the fact that fluctuations are always present in an explosive system (but they were not taken into account in the balance equation (2.1)) one may wonder what would be the *dynamics* of a stochastic flame front appearing at criticality? As a matter of fact, in the context of a zero-dimensional stochastic perturbation theory—in the small noise parameter—we have been able to characterize the *lifetime* (i.e. the MFPT) of the (homogeneous) unstable state that appears in the normal form associated with the Semenov model [4]. In [13] we have shown by using the SPPA that it is possible to obtain an analytical expression of the FPTD, which is useful to study the random explosive times in a well stirred reactor (see also [14] for the marginal case $b = 0$). Here we propose to generalize the SPPA for an extended system when its normal form at criticality is characterized by (2.15). Therefore in the next section we shall incorporate an additive noise $\xi(\tilde{x}, \tilde{t})$ in this normal form in order to consider random fluctuations in the amplitude equation.

3. Stochastic path perturbation approach for extended systems

In a zero-dimensional system we have shown that the SPPA is a powerful technique for calculating—analytically—the FPTD for different normal forms [13, 14, 17]. In appendix B we summarize these results for a particular non-symmetric potential normal form, i.e. we characterize the *lifetime* of the unstable state $X = 0$ from the stochastic differential equation $\dot{X} = aX^2 + b + \sqrt{\varepsilon}\xi(t)$, with $a > 0$, $b \geqslant 0$, and where $\xi(t)$ is a zero-mean Gaussian white-noise process.

Here we extend that approach to spatially distributed systems characterized by a limit point bifurcation. Therefore, let us study the universal *saddle-node* normal form (stochastic normal form)

$$\partial_{\tilde{t}}\phi(\tilde{x}, \tilde{t}) = D\partial_{\tilde{x}}^2\phi(\tilde{x}, \tilde{t}) + a\phi(\tilde{x}, \tilde{t})^2 + b + \sqrt{\varepsilon}\xi(\tilde{x}, \tilde{t}) \qquad \tilde{x} \in [-L, L] \tag{3.1}$$

where D is the diffusion constant, $a > 0$, $b \geqslant 0$, ε is the small-noise intensity and $\xi(\tilde{x}, \tilde{t})$ is a zero-mean Gaussian stochastic field characterized by the white correlation

$$\langle \xi(\tilde{x}, \tilde{t})\xi(\tilde{x}', \tilde{t}')\rangle = \delta(\tilde{x} - \tilde{x}')\delta(\tilde{t} - \tilde{t}'). \tag{3.2}$$

It has been pointed out, in view of the nonlinear and violent character of a chemical explosion that the effect of fluctuations should be taken into account; in a mesoscopic description the starting point to work out these fluctuations is a full master equation [5]. But in a more phenomenological way it can be shown that an augmented Semenov equation incorporating the effect of fluctuations has a Langevin-like structure [6] (additive noise). Hence our amplitude equation (3.1) can be considered in that framework [4]. In what follows we will study the lifetime of the free stochastic field $\phi(\tilde{x}, \tilde{t})$ from the unstable state $\phi = 0$ characterized by the evolution equation (3.1).

3.1. Fourier analysis of the escape processes

In order to study the stochastic dynamics (3.1) of the amplitude $\phi(\tilde{x}, \tilde{t})$, it is important to analyse the stochastic escape processes (lifetime) of each Fourier mode. This can be done by generalizing the SPPA [13, 14] to extended systems. Here we are going to use this generalized SPPA to solve the problem of an extended explosive system, but this approach can also be applied to other systems.

Remark 1. *At criticality, the relaxation process to $\theta_{hot}(\tilde{x})$ (the hot attractor of (2.3)) is triggered by the fluctuations; and the* dominant *equation under consideration is then (3.1), which in Fourier space reads*†

$$\dot{\phi}_k = a \sum_{n=-\infty}^{\infty} \phi_n \phi_{k-n} + b\delta_{0,k} - \left(\frac{k\pi}{L}\right)^2 D\phi_k + \sqrt{\varepsilon}\xi_k \qquad k = 0, \pm 1, \pm 2, \ldots \tag{3.3}$$

where the correlation (3.2) in Fourier representation reads

$$\langle \xi_k(\tilde{t})\xi_j(\tilde{t}')\rangle = \delta_{k,j}\delta(\tilde{t} - \tilde{t}'). \tag{3.4}$$

Remark 2. *The* lifetime *from the neighbourhood of the unstable state $\phi(\tilde{x}, \tilde{t}) = 0$ is characterized by the FPTD for each Fourier mode ϕ_k, to reach a macroscopic value $\phi_k \gg \sqrt{\varepsilon}$. These probability distributions can be obtained (analytically) by analysing the different stages of evolution of each Fourier mode ϕ_k. We will show that it is possible to separate several stages of evolution, from which the dynamics of $\phi(\tilde{x}, \tilde{t})$ toward the attractor $\theta_{hot}(\tilde{x})$ can be inferred.*

Basically the idea is to decompose the mode $\phi_0(t)$ into two parts, a process $H_0(t)$ associated with the very early stage of evolution and a process $Y_0(t)$, which takes into account the nonlinear terms of the dynamical equation. Up to this point the iterative scheme is similar (but not equal) to the zero-dimensional case [13]. Therefore the lifetime of the unstable homogeneous state is characterized by the escape of the mode ϕ_0, which can be studied in terms of the FPTD. The role of the non-homogeneous modes in the whole escape process will be discussed, in a similar way, in section 3.2. From now on we use the simpler notation $\phi_k(t)$ instead of $\phi_k(\tilde{t})$, etc.

In order to proceed with this programme, and inspired by the previous experience [13], we now introduce the nonlinear transformation ($\forall\ k = 0, \pm 1, \pm 2, \ldots$)

$$\phi_k(t) = \frac{H_k(t)}{Y_k(t)} \tag{3.5}$$

† The free fields $\phi(\tilde{x}, \tilde{t})$ and $\xi(\tilde{x}, \tilde{t})$ are expanded in Fourier modes using to the notation: $\phi(\tilde{x}, \tilde{t}) = \sum_{-\infty}^{\infty} \phi_k(t) \exp(\mathrm{i}k\pi\tilde{x}/L)$ and $\xi(\tilde{x}, \tilde{t}) = \sum_{-\infty}^{\infty} \xi_k(\tilde{t}) \exp(\mathrm{i}k\pi\tilde{x}/L)$, for periodic boundary condition on $\tilde{x} \in [-L, L]$.

and according to the initial condition $\phi(\tilde{x}, 0) = 0$ we use the Fourier initial conditions $\forall\, k$

$$H_k(0) = 0 \qquad Y_k(0) = 1. \tag{3.6}$$

Using the transformation (3.5) in (3.3) we get

$$\frac{\dot{H}_k}{Y_k} - \frac{H_k}{Y_k^2}\dot{Y}_k = a \sum_{n=-\infty}^{+\infty} \frac{H_n}{Y_n}\frac{H_{k-n}}{Y_{k-n}} - b\delta_{0,k} - D(\pi k/L)^2 \frac{H_k}{Y_k} + \sqrt{\varepsilon}\xi_k. \tag{3.7}$$

The SPPA consists of choosing a suitable nonlinear transformation†, such that from (3.7) two set of coupled equations can be found in order to be able to solve—in an iterative way—the evolution of the *initial stage*, in a small-noise approximation.

3.1.1. Mode $k = 0$. From (3.7) it is possible to write the following equivalent set of coupled equations:

$$\dot{H}_0 = \left[b + \sqrt{\varepsilon}\xi_0 + a \sum_{n=-\infty (n\neq 0)}^{\infty} \frac{H_n}{Y_n}\frac{H_{-n}}{Y_{-n}} \right] Y_0 \tag{3.8}$$

$$\dot{Y}_0 = -aH_0. \tag{3.9}$$

Note that if we can show that any term $\frac{H_n}{Y_n}\frac{H_{-n}}{Y_{-n}}$, $\forall\, n \neq 0$ is of $\mathcal{O}(\varepsilon)$, the iterative procedure up to $\mathcal{O}(\sqrt{\varepsilon})$ (if $b \neq 0$) will be analogous to the zero-dimensional case [13], i.e., the statistic of the escape time t_e of the homogeneous mode is governed by the statistic of the root of the random equation $Y_0(t_e) = 0$.

3.1.2. Modes with $k \neq 0$. From (3.7) we can write, for the non-homogeneous modes, the following equivalent set of coupled equations:

$$\dot{H}_k = \left[a \sum_{n=-\infty (n\neq k,0)}^{+\infty} \frac{H_n}{Y_n}\frac{H_{k-n}}{Y_{k-n}} + \sqrt{\varepsilon}\xi_k \right] Y_k \tag{3.10}$$

$$\dot{Y}_k = [-2a\phi_0 + D(\pi k/L)^2] Y_k \tag{3.11}$$

$\forall k \neq 0$. Therefore, from (3.11) and using (3.6) we write

$$Y_k(t) = \exp \int_0^t (-2a\phi_0(t') + \alpha_k)\, \mathrm{d}t' \tag{3.12}$$

where we define $\alpha_k \equiv D(\pi k/L)^2$. Approximating, at short times, $Y_k(t) \approx 1$ in the evolution equation for $\dot{H}_k$ we get

$$H_k(t) \approx \int_0^t \left[a \sum_{n=-\infty (n\neq k,0)}^{+\infty} \phi_n(t')\phi_{k-n}(t') + \sqrt{\varepsilon}\xi_k(t') \right] \mathrm{d}t'. \tag{3.13}$$

Considering that $\phi_k(t) = H_k(t)/Y_k(t)$ the small-noise iterative dominant contribution gives

$$\phi_k(t) \approx [\sqrt{\varepsilon}W_k(t) + \mathcal{O}(\varepsilon)] \exp \int_0^t (+2a\phi_0(t') - \alpha_k)\mathrm{d}t' \tag{3.14}$$

where $W_k(t)$ is the Wiener process:

$$W_k(t) = \int_0^t \xi_k(t')\mathrm{d}t' \qquad W_k(0) = 0. \tag{3.15}$$

† Note that for a different normal form the non-trivial transformation $\phi_k(t) = H_k(t)^\alpha / Y_k(t)^\beta$ could have different exponents α, β, see, for example, [17, 19].

From (3.14) we see that $\phi_k(t)$ will be bounded if $\phi_0(t)$ is of the $\mathcal{O}(\sqrt{\varepsilon})$. Hence at the initial stage we can take

$$\phi_k(t) \approx \mathcal{O}(\sqrt{\varepsilon}) \qquad \forall k \neq 0. \tag{3.16}$$

A better approximation will be given using (3.23), see section 3.2. The important point is that this approximation is enough to study, in a self-consistent way, the evolution of the initial stage of the homogeneous mode; i.e., using (3.16) in (3.8) we see that the dominant order, $\mathcal{O}(\sqrt{\varepsilon})$, in the evolution of $\phi_0(t)$ is the same as for the zero-dimensional case [13]. If $b = 0$ (marginal case) the role of the non-homogeneous modes is more important, this is so because in this case the iterative procedure must be performed up to $\mathcal{O}(\varepsilon)$ in order to improve the passage time statistics [14]. However, in the marginal case we expect that neglecting the term $aY_0 \sum_{n=-\infty(n\neq 0)}^{\infty} \frac{H_n}{Y_n} \frac{H_{-n}}{Y_{-n}}$ in (3.8) could lead to some discrepancy with the simulations. We show, in what follows, that this crude approximation reproduces quite well the Monte Carlo simulations, even for the marginal case, see section 3.3.

3.1.3. The stochastic paths. In the initial state the set of equations (3.8) and (3.9) can be solved iteratively in $\mathcal{O}(\sqrt{\varepsilon})$; higher corrections must be taken in the marginal case†, see appendix B. We remark that in the SPPA only the evolution of the *initial state* is necessary to be able to calculate the random escape times. Then the stochastic paths $\phi_0(t)$ can be written in the form

$$\phi_0(t) \approx \frac{bt + \sqrt{\varepsilon} W_0(t)}{1 - \frac{1}{2}abt^2 - a\sqrt{\varepsilon}\Omega_0(t)} \tag{3.17}$$

where $W_0(t)$ is a Wiener process, i.e.

$$W_0(t) = \int_0^t \xi_0(t')\,\mathrm{d}t' \qquad W_0(0) = 0 \tag{3.18}$$

and the Gaussian process $\Omega_0(t)$ is defined by

$$\Omega_0(t) = \int_0^t W_0(t')\,\mathrm{d}t'. \tag{3.19}$$

Scaling-out Wiener integrals: $W_0(t) = t^{1/2} W_0$ and $\Omega_0(t) = t^{3/2}\Omega_0$ where W_0 and Ω_0 are Gaussian random variables, the escape of the stochastic paths $\phi_0(t_e) = \infty$ (3.17) is characterized by the random algebraic equation $1 - \frac{1}{2}abt_e^2 - a\sqrt{\varepsilon}t_e^{3/2}\Omega_0 = 0$. Hence the lifetime of the unstable homogeneous state is characterized by the escape of the mode ϕ_0, which is given in equation (B2) in terms of the FPTD $P(t_e)$, see appendix B for the details.

Now let us deal with the $k \neq 0$ Fourier numbers, these modes can also be studied iteratively. First integrate $Y_k(t)$, from (3.11), then

$$Y_k(t)_{t<t_e} \approx \exp[\alpha_k t] \qquad \forall t < t_e \tag{3.20}$$

$$Y_k(t)_{t>t_e} \approx Y_k(t_e)_{t<t_e} \exp[(\alpha_k - 2a\mathcal{E})(t - t_e)] \qquad \forall t > t_e \tag{3.21}$$

where $\mathcal{E} \sim \mathcal{O}(1)$ is of the order of the *highest* temperature in the reactor (i.e., $\mathcal{E}$ is of the order of maximum temperature in the *hot* profile $\theta_{hot}(\tilde{x})$: see appendix A for a piecewise linear example). To get this expression we have used the instanton-like approximation for the temporal behaviour of $\phi_0(t)$, i.e. $\phi_0(t) = \mathcal{E}u(t - t_e)$. Here t_e is the random time characterizing the escape of the homogeneous mode (B2) ((B9) for the marginal case $b = 0$) and $u(t - t_e)$ is the Heaviside function. In order to approximate the processes $H_k(t)$ we first integrate to $\mathcal{O}(\sqrt{\varepsilon})$ using that at short times $Y_k(t \sim 0) \approx 1$ and $H_k(t \sim 0) \approx 0$; then

$$H_k(t) \approx \sqrt{\varepsilon} W_k(t) \tag{3.22}$$

† If $b = 0$, from (3.8) we approximate the paths to the dominant order $\mathcal{O}(\varepsilon)$, but in this case the influence of the non-homogeneous modes is of the same order.

where the Wiener processes $W_k(t)$ are statistically independent of $W_{k'}(t)$ for $k \neq k'$. Now, to improve these solutions we iterate $H_k(t)$ again using (3.20) and (3.22), then we get for $H_k(t)$ the improved solution (for $k \neq 0$)

$$H_k(t) \approx \int_0^t \mathrm{d}t'\, Y_k(t') \left[\sum_{n=-\infty(n,k-n\neq 0)}^{+\infty} a\varepsilon \frac{W_n(t')W_{k-n}(t')}{Y_n(t')Y_{k-n}(t')} + \sqrt{\varepsilon}\xi_k(t') \right]. \tag{3.23}$$

Remembering that $\phi_k(t) = \frac{H_k(t)}{Y_k(t)}$, it is possible to see the complicated coupling between the different Fourier modes. Nevertheless, from the expression (3.20) and (3.23) it is simple to see that the growth of the modes ϕ_k (for $k \neq 0$) is subordinated to the escape of the homogeneous mode ϕ_0. An important result that can also be inferred from this approximation is that only Fourier modes fulfilling the 'deterministic' selection rule

$$\alpha_k \equiv D(k\pi/L)^2 < 2a\mathcal{E} \tag{3.24}$$

will grow exponentially after the random time t_e. This condition connects the exploding Fourier indices $k \in [\pm 1, \pm 2 \ldots, \pm k^*]$ with the *deterministic* parameters of the system which appear in the bound index $k^* \sim$ integer$\left[\frac{L}{\pi}\sqrt{\frac{2a\mathcal{E}}{D}}\right]$, i.e., the diffusion coefficient D, the nonlinear parameter a, the size of the one-dimensional reactor (length L), and the highest temperature $\mathcal{E}$ of the profile $\theta_{hot}(\tilde{x})$.

Note that the 'escape' of the non-homogeneous mode $\phi_k(t)$ has its dominant random character through the process $Y_k(t)$, which (in its first iteration) is random by virtue of the random time t_e of the homogeneous mode $\phi_0(t)$. The distribution of these times t_e takes into account only the universal parameter $K = \frac{b^3}{a\varepsilon^2}$, which measure the departure from the marginal case $b = 0$ (delaying the explosion), and the deterministic time $\tau = \pi\sqrt{1/(4ab)}$ (if $b = 0$ the FPTD $P(t_e)$ only depends on the universal parameter $a\sqrt{\varepsilon}$, see appendix B). Another result that can also be seen from our SPPA is that the smaller the Fourier number k the faster its *exponential growth*.

We emphasize that our k^* characterizes a maximal 'effective' Fourier index, i.e., the maximal Fourier mode $\phi_k(t)$ which will grow exponentially up to the macroscopic important value $\mathcal{E}_k$ at random times. This is a result which can numerically be tested from (3.3). It should be pointed out that our estimation of this Fourier index is an approximation. Unfortunately we do not know the exact deterministic ($\varepsilon = 0$) solution of (3.3), hence we cannot introduce a renormalization procedure to improve our prediction of k^*. Note that a renormalization procedure was possible in the zero-dimensional case because the exact deterministic solution is known [13]. Figure 1 shows realizations of $\phi_k(t)$ for several values of $k(= 0, 1, 2, 3, 4)$, showing the occurrence of an effective maximal Fourier index, which in fact is well estimated by our approximation. This figure shows that the important exploding modes are in fact $\phi_0(t)$, $\phi_1(t)$, $\phi_2(t)$. Nevertheless modes $\phi_3(t)$, and $\phi_4(t)$ can also be seen to explode, but their respective amplitudes are much smaller and they are not important in the calculation of the space fluctuations of the flame front. For $D = 5$, $a = 5$, $\mathcal{E} = 43.9$, $L = 1$ and using the selection rule (3.24) the predicted maximal Fourier index gives $k^* =$ integer[2.98]. In several simulations (we run 10^2 Fourier modes) we could not see to explode beyond the $\phi_4(t)$ mode.

3.2. Passage times for the Fourier modes ϕ_k

In a similar way to how we characterized the *lifetime* of the unstable homogeneous state $\phi_0 = 0$ (i.e. the FPTD $P(t_e)$, see appendix B), the SPPA gives also the possibility to characterize the lifetime of each mode ϕ_k by calculating the joint probability distribution $\Pi(t_k; t_e)$. We will show that both random times are correlated, and the FPTD of the mode ϕ_k is given by the

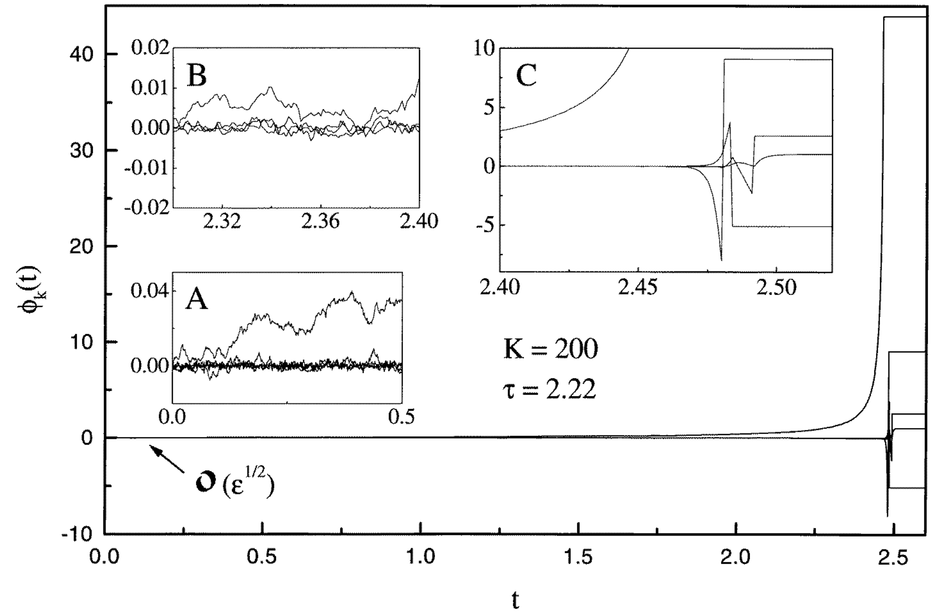

Figure 1. Realizations of $\phi_k(t)$ for several values of $k = (0, 1, 2, 3, 4)$ as function of time t for the universal parameter $K = 200$, deterministic time $\tau = 2.22$, length scale $L = 1$, and diffusion coefficient $D = 5$. The macroscopic highest temperature used to count the *escape* of the homogeneous mode was $\mathcal{E}_0 = 43.9$. The selection rule predicts an effective $k^* \simeq 3$. The Monte Carlo simulations were performed using 10^5 realizations and show the Fourier modes exploding at random times, after which their temporal behaviour is kept to their respective macroscopic values $\mathcal{E}_k$. Inset (*a*) shows that the fluctuations are of $\mathcal{O}(\varepsilon^{1/2})$; nevertheless, note that for this early interval of time $\phi_0(t)$ shows much higher bounded fluctuations. Inset (*b*) shows the realizations $\phi_1(t)$, $\phi_2(t)$, $\phi_3(t)$, and also $\phi_4(t)$ for a small time interval before the huge explosion of the homogeneous mode. Inset (*c*) shows a small time interval just after the explosion of $\phi_0(t)$; therein the realization of $\phi_4(t)$ can be seen to fluctuate but finally its explodes to a tiny value $\mathcal{O}(\mathcal{E}_4)$.

marginal distribution

$$\Pi(t_k) = \int_0^\infty \Pi(t_k; t_e)\,\mathrm{d}t_e \qquad \text{for} \quad \forall k \in [\pm 1, \pm 2 \cdots, \pm k^*].$$

Of course, the FPTD of the mode ϕ_0 is $P(t_e) = \int_0^\infty \Pi(t_k; t_e)\,\mathrm{d}t_k$.

Let us fix a value of the random time t_e, then from (3.20) and (3.23) the stochastic paths of the Fourier mode ϕ_k can be approximated by the dominant contribution

$$\phi_k(t) \simeq \frac{1}{Y_k(t)}\left[\sqrt{\varepsilon}\int_0^t Y_k(t')\xi_k(t')\,\mathrm{d}t' + \mathcal{O}(\varepsilon)\right] \qquad k = \pm 1, \pm 2, \ldots. \tag{3.25}$$

From this expression we see immediately that if $t < t_e$ the stochastic processes $\phi_k(t)$ are bounded, i.e. their variances are not increasing functions of time t (small fluctuations $\mathcal{O}(\varepsilon)$ can be seen taking the space average, see appendix C). Nevertheless, if $t > t_e$ and $\alpha_k < 2a\mathcal{E}$ the stochastic process $\phi_k(t)$ grows exponentially in time, in particular its variance diverges for $t \to \infty$.

Therefore let us focus on the stochastic process $\phi_k(t)$ for $t > t_e$, and for any k fulfilling the selection rule (3.24). From (3.25) and using the expressions of $Y_k(t)$, it follows (up to $\mathcal{O}(\sqrt{\varepsilon})$) that we can approximate

$$\phi_k(t)|_{t>t_e} \simeq \frac{\sqrt{\varepsilon}}{Y_k(t)}\left[\int_0^{t_e} Y_k(t')\xi_k(t')\,\mathrm{d}t' + \int_{t_e}^{t} Y_k(t')\xi_k(t')\,\mathrm{d}t'\right] \qquad \forall k \in [\pm 1, \pm 2 \ldots, \pm k^*].$$

(3.26)

From this expression it is simple to see that the dominant term characterizing the growth of the fluctuations, for times t of $\mathcal{O}(t_e)$, comes from the first integral. Thus we can approximate the growth of the Fourier mode ϕ_k (in its initial stage of evolution) by the stochastic paths (for $k \in [\pm 1, \pm 2 \ldots, \pm k^*]$)

$$\phi_k(t) \simeq \frac{\sqrt{\varepsilon}}{Y_k(t)}\left[\int_0^{t_e} Y_k(t')\xi_k(t')\,\mathrm{d}t'\right] = \Xi \exp(2a\mathcal{E} - \alpha_k)t \qquad t > t_e \tag{3.27}$$

where $\Xi \equiv \Xi(a, \alpha_k, \mathcal{E}, t_e)$ is a Gaussian random variable with mean value zero and variance

$$\langle \Xi^2 \rangle = \frac{\varepsilon}{2\alpha_k}[\exp(-(2a\mathcal{E} - \alpha_k)t_e) - \exp(-4a\mathcal{E}t_e)]. \tag{3.28}$$

We note that owing to the fact that $\alpha_k < 2a\mathcal{E}$ the dispersion (3.28) is bounded (in fact it is of $\mathcal{O}(\varepsilon)$).

Remark 3. *Equation (3.27) can be used to obtain the FPTD for any Fourier mode $k \neq 0$ fulfilling the rule (3.24). The random escape time t_k can be inferred from the random quantity $\phi_k(t_k)^2$ reaching a macroscopic threshold $\mathcal{E}_k^2 \gg \varepsilon$; then we write $\mathcal{E}_k^2 = \Xi^2 \exp 2(2a\mathcal{E} - \alpha_k)t_k$, which means*

$$t_k = \frac{1}{2(2a\mathcal{E} - \alpha_k)} \log \mathcal{E}_k^2/\Xi^2. \tag{3.29}$$

For $t_k \geqslant 0$ this equation can be used as a transformation mapping from the Gaussian random variable Ξ to the random variable t_k. Then applying the theorem of transformation we obtain

$$\Pi(t_k \mid t_e) = \langle \delta(t_k - t_k(\Xi)) \rangle_{P(\Xi)} = \frac{2(2a\mathcal{E} - \alpha_k)\mathcal{E}_k}{\sqrt{2\pi\langle \Xi^2 \rangle}} \left[\mathrm{erf}\left(\frac{\mathcal{E}_k}{\sqrt{2\langle \Xi^2 \rangle}}\right)\right]^{-1}$$
$$\times \exp\left(\frac{-\mathcal{E}_k^2 \exp(-2(2a\mathcal{E} - \alpha_k)t_k)}{2\langle \Xi^2 \rangle} - (2a\mathcal{E} - \alpha_k)t_k\right) \tag{3.30}$$

where all the non-trivial dependence on the random escape time t_e comes from $\langle \Xi^2 \rangle$, see (3.28). Here $\mathcal{E} \sim \mathcal{O}(1)$ is of the order of the highest hot temperature and the thresholds $\mathcal{E}_k$ are of the order of the Fourier weights of the hot attractor profile (thus we can take $\mathcal{E} \sim \mathcal{E}_0$ and, in general, $\mathcal{E}_k$ (for all k) from its Fourier expansion (3.39)).

The marginal probability distribution $\Pi(t_k)$ (for $k \in [\pm 1, \pm 2 \ldots, \pm k^*]$) is found from the integral

$$\Pi(t_k) = \int_0^\infty \mathrm{d}t_e\, \Pi(t_k \mid t_e) P(t_e) \tag{3.31}$$

where $P(t_e)$ can be read from appendix B. Note that here we have used the notation $\Pi(t_k \mid t_e)$ to emphasize the character of conditional FPTD. The strong correlation between t_k and t_e is evident because the joint probability distribution $\Pi(t_k; t_e) = \Pi(t_k \mid t_e)P(t_e)$ cannot be factorized.

Equation (3.29) shows an analogy with the Susuki scaling transformation appearing in the decay from an unstable homogeneous state [18]. In fact, considering the FPTD from a random variable transformation as in (3.29), and using the instanton approximation, it has been shown that Susuki's anomalous fluctuation may also be studied in the context of the SPPA [19]. This fact will be clarified and generalized to extended systems in the next sections.

3.2.1. Moments of the random escape time t_k. The integral (3.31) must be solved numerically due to the complicated structure of the function $P(t_e)$, see (B2), and (B9) for the marginal case. Nevertheless, one can introduce another method to get an analytical estimation of the MFPT: $\mathcal{T}_k = \int_0^\infty t_k \Pi(t_k) \mathrm{d}t_k$. This can be seen by considering the generating function

$$G(\omega) = \langle \exp(-\omega t_k) \rangle. \tag{3.32}$$

From (3.29) the function $t_k = t_k(\Xi)$ can be used; then the calculation of the conditional generating function is straightforward:

$$\begin{aligned} G(\omega|t_e) &= \int \frac{1}{\sqrt{2\pi\langle\Xi^2\rangle}} \exp\left(-\frac{-\Xi^2}{2\langle\Xi^2\rangle} - \omega t_k(\Xi)\right) \mathrm{d}\Xi \\ &\simeq \frac{\Gamma(\frac{\omega}{2(2a\mathcal{E}-\alpha_k)} + \frac{1}{2})}{\sqrt{\pi}} \left(\frac{2\langle\Xi^2\rangle}{\mathcal{E}_k^2}\right)^{\frac{\omega}{2(2a\mathcal{E}-\alpha_k)}} + \mathcal{O}\left(\mathrm{e}^{-\frac{\mathcal{E}_k^2}{2\langle\Xi^2\rangle}}\right) \end{aligned} \tag{3.33}$$

where $\Gamma(z)$ is the Gamma function, and the variance $\langle\Xi^2\rangle$ is characterized by the random escape time t_e see (3.28). All the conditional moments $\langle t_k^m \rangle_{t_e}$ can be calculated by differentiation of $G(\omega \mid t_e)$. In particular, the MFPT is the first moment

$$\begin{aligned} \langle t_k \rangle_{t_e} &= -\frac{\mathrm{d}}{\mathrm{d}\omega}[G(\omega|t_e)]_{\omega=0} = \frac{1}{2(2a\mathcal{E}-\alpha_k)} \\ &\quad\times \left\{\log\left(\frac{\mathcal{E}_k^2\alpha_k}{\varepsilon}\right) - \log[\exp(-2(2a\mathcal{E}-\alpha_k)t_e) - \exp(-4a\mathcal{E}t_e)] - \psi\left(\frac{1}{2}\right)\right\} \end{aligned} \tag{3.34}$$

where $\psi(\frac{1}{2})$ is the Digamma function. The expression for $\langle t_k \rangle_{t_e}$ should be averaged over the distribution of the random escape times t_e but this is difficult to do analytically, so we need to introduce a new approximation.

Because we are only interested in Fourier modes fulfilling selection rule (3.24), it is possible to approximate (for large values of α_k but smaller than $2a\mathcal{E}$) $\exp(-2(2a\mathcal{E}-\alpha_k)t_e) - \exp(-4a\mathcal{E}t_e) \approx \exp(-2(2a\mathcal{E}-\alpha_k)t_e)$ in (3.34), and then we obtain a simpler expression $\langle t_k \rangle_{t_e}$ which can now easily be averaged over $P(t_e)$

$$\begin{aligned} \mathcal{T}_k &= \int_0^\infty P(t_e)\langle t_k \rangle_{t_e} \mathrm{d}t_e \approx \langle t_e \rangle + \frac{1}{2(2a\mathcal{E}-\alpha_k)} \\ &\quad\times \left\{\log\left(\frac{\mathcal{E}_k^2\alpha_k}{\varepsilon}\right) + \gamma + \ln 4\right\} \qquad \text{for} \quad 2\alpha_k\langle t_e \rangle \gg 1. \end{aligned} \tag{3.35}$$

Here $\gamma = 0.577\,216$ is the Euler constant. This approximation is valid for $k \sim k^* = \text{integer}[\frac{L}{\pi}\sqrt{\frac{2a\mathcal{E}}{D}}]$ and gives an estimation of the largest timescale of the exploding Fourier mode. From (3.35) we see that the escape time of this non-homogeneous mode resembles Susuki's law. Nevertheless, in addition to the term $\sim\log(\frac{1}{\varepsilon})$ we have here a fine structure depending on the Fourier number and threshold $\mathcal{E}_k$. Of course, this formula shows that the MFPT $\mathcal{T}_k$ for each Fourier mode is larger than the homogeneous MFPT $\langle t_e \rangle$ because $\frac{\mathcal{E}_k^2\alpha_k}{\varepsilon} > 1$, as was expected.

Another interesting case is the limit of small α_k values; in this case (3.34) can also be analytically studied. Rewriting (3.34) the MFPT $\mathcal{T}_k$ can be put in the form

$$\begin{aligned} \mathcal{T}_k &= \frac{1}{2(2a\mathcal{E}-\alpha_k)} \int_0^\infty P(t_e) \\ &\quad\times \left[\log\left(\frac{\mathcal{E}_k^2}{\varepsilon}\right) - \psi\left(\frac{1}{2}\right) + \log\alpha_k + 4a\mathcal{E}t_e - \log(\exp(2\alpha_k t_e) - 1)\right] \mathrm{d}t_e. \end{aligned} \tag{3.36}$$

Then for a fixed time t_e and in the limit $2\alpha_k t_e \ll 1$ (small Fourier numbers) we get

$$\langle t_k\rangle_{t_e} \approx t_e + \frac{1}{4a\mathcal{E}}\left[\log\left(\frac{\mathcal{E}_k^2}{\varepsilon}\right) + \gamma + \ln 2 - \log t_e\right]. \tag{3.37}$$

We see, from (3.37), that $\langle t_k\rangle_{t_e}$ can be advanced relative to the case of large Fourier numbers $k \sim k^*$. The interesting point is to know the delayed time: $\mathcal{T}_k - \langle t_e\rangle$ as a function of the geometry (selection rule (3.24)) and the physical parameters of the system.

We emphasize that (3.35) and (3.37) are approximations which can, in principle, be improved by using (3.36). Therefore a crude approximation of the FPTD for the different Fourier random times t_k could be taken as

$$\Pi(t_k) \approx P\left(t_k + \frac{1}{2(2a\mathcal{E} - \alpha_k)}\log\frac{\mathcal{E}_k^2\alpha_k}{\varepsilon}\right) \tag{3.38}$$

$P(t)$ being the FPTD of the homogeneous mode.

In general, the conditional generating function $G(\omega \mid t_e)$ and the distribution $P(t_e)$ make it possible to calculate any moment of t_k.

3.3. Transient fluctuations of the flame front

The *saddle-node* normal form (3.1) describes the ignition; i.e., the dynamics from criticality toward the *hot* attractor $\theta_{hot}(\tilde{x})$. It is therefore important to have some *analytical* approximation for this time evolution, and, in particular, to study its space fluctuations in order to characterize the random explosion of a flame front.

A quantity which is representative of these fluctuations is the so-called anomalous fluctuation, which typically appears in any nonequilibrium phase transition [18]. In order to do this we first represent the hot attractor $\theta_{hot}(\tilde{x})$ by Fourier cosine analysis, then

$$\theta_{hot}(\tilde{x}) = \sum_{k=0,1,2,\ldots}\mathcal{E}_k\cos\left(\frac{k\pi\tilde{x}}{L}\right) \qquad x \in [-L, L] \tag{3.39}$$

where the constants $\mathcal{E}_k$ can be obtained easily. Therefore the dynamics of $\phi(\tilde{x},\tilde{t})$ (from the unstable state $\phi = 0$) can be approximated by the instanton-like stochastic field (neglecting the initial $\mathcal{O}(\sqrt{\varepsilon})$ space fluctuation around $\phi = 0$)

$$\phi(\tilde{x},\tilde{t}) = u(\tilde{t} - t_e)\mathcal{E}_0 + \sum_{k=1,2,\ldots} u(\tilde{t} - t_k)\mathcal{E}_k\cos\left(\frac{k\pi\tilde{x}}{L}\right). \tag{3.40}$$

This approximation describes quite well all the transient fluctuations, but also discards the final fluctuations $\mathcal{O}(\sqrt{\varepsilon})$ around the hot attractor. In (3.40) the random nature of the evolution profile is taken into account by the random character of the *escape* times t_e and t_k. In appendix B we have given the analytical expression for the FPTD $P(t_e)$; the non-homogeneous modes ($k \neq 0$) are characterized by the FPTD $\Pi(t_k)$, see section 3.2, equation (3.31) or approximation (3.38). Therefore, expression (3.40) allows us to explore the transient spatial fluctuations of the flame front. In particular, the anomalous fluctuations can be characterized by the function

$$\begin{aligned}\sigma_\phi(\tilde{t}) &= \frac{1}{\mathcal{D}_x}\int_{\mathcal{D}_x}(\langle\phi(\tilde{x},\tilde{t})^2\rangle - \langle\phi(\tilde{x},\tilde{t})\rangle^2)\,\mathrm{d}\tilde{x}\\ &= \frac{1}{\mathcal{D}_x}\int_{\mathcal{D}_x}\left[\sum_{k=0,1,2,\ldots}\sum_{k'=0,1,2,\ldots}\cos\left(\frac{k\pi\tilde{x}}{L}\right)\cos\left(\frac{k'\pi\tilde{x}}{L}\right)\mathcal{E}_k\mathcal{E}_{k'}\langle u(\tilde{t}-t_k)u(\tilde{t}-t_{k'})\rangle\right.\\ &\quad\left.-\sum_{k=0,1,2,\ldots}\left(\cos\left(\frac{k\pi\tilde{x}}{L}\right)\mathcal{E}_k\right)^2\langle u(\tilde{t}-t_k)\rangle^2\right]\mathrm{d}\tilde{x}.\end{aligned} \tag{3.41}$$

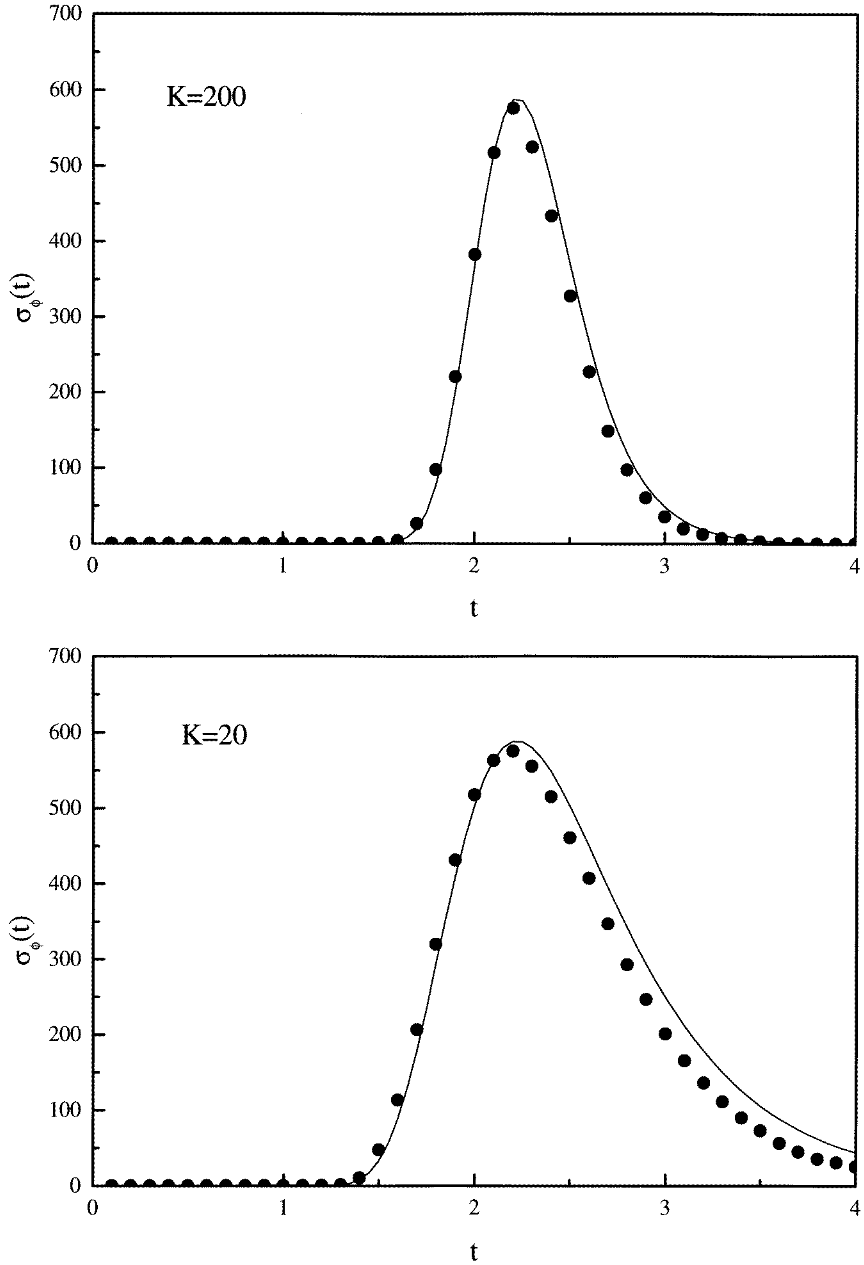

Figure 2. Anomalous fluctuations $\sigma_\phi(t)$ as function of time t for several values of the noise parameter ε. (*a*) $K = 200$ and $\tau = 2.22$; (*b*) $K = 20.22$ and $\tau = 2.22$; and (*c*) for the marginal case $b = 0$ with $a\varepsilon^{1/2} = 1$. Monte Carlo simulations (dots) were performed using 10^5 realizations.

Here $\mathcal{D}_x \equiv [-L, L]$ and the notations $t_0 \equiv t_e$, $\mathcal{E}_0 \equiv \mathcal{E}$ are understood. Using the orthogonal property of the Fourier basis, it follows that

$$\sigma_\phi(\tilde{t}) = \sum_{k=0,1,2,\ldots}^{k^*} \mathcal{E}_k^2 \left[\int_0^{\tilde{t}} \Pi(t_k)\, \mathrm{d}t_k - \left(\int_0^{\tilde{t}} \Pi(t_k)\, \mathrm{d}t_k \right)^2 \right]. \tag{3.42}$$

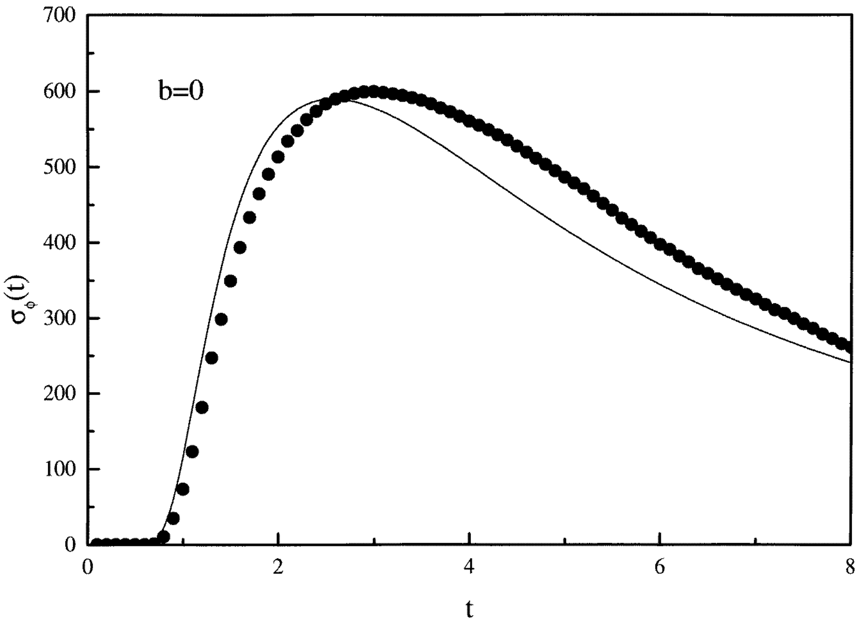

Figure 2. (Continued)

Here we have used the notation $\Pi(t_0) \equiv P(t_e)$. Note that k^* takes into account the selection rule (3.24) of the Fourier modes that 'effectively' grow in accord with the parameters of the system: $D, a, L, \mathcal{E}_0$.

Equation (3.42) is a closed expression given in terms of the FPTD which characterizes the explosions of each Fourier mode. In figures 2(*a*)–(*c*) the function $\sigma_\phi(t)$ has been compared with Monte Carlo simulations, showing thereby a good agreement with our theoretical predictions (see appendix D for the numerical details). If the system is far from the marginal case this function shows a narrow distribution near the deterministic time $\tau = \pi\sqrt{\frac{1}{4ab}}$, but this is not the case when $K \ll 1$. In the *marginal* situation the FPTD $P(t_e)$ has a long tail $\sim t^{-5/2}$; hence predicting a large dispersion. Note that for this case our paths (B6) are just the simplest approximation (neglecting some renormalization to the $\mathcal{O}(\varepsilon)$), which is why our theoretical prediction does not show a very good agreement with the Monte Carlo simulations of figure 2(*c*).

Formula (3.42) provides a good approach for characterizing the random ignition time of an extended system. Note that whatever the physical parameters are, our SPPA gives a good description (in the small-noise limit) of the space fluctuations. The marginal situation corresponds to the case when $b \to 0$ (and this, of course, depends on the geometry and the thermochemical parameters, see (2.16)); in this case $P(t_e)$ is given by (B9) which is only an approximation of the escape of the homogeneous mode, see appendix B. The influence of the escape of the non-homogeneous Fourier modes, is taken into account in our approach—in a non-trivial way—through the formula for $\Pi(t_k)$.

4. Conclusions

We have presented a stochastic perturbation theory—in the small-noise parameter—which allows us to study the anomalous fluctuations for extended systems near the critical point. The physical system was assumed to be at criticality, thereby the dynamics was represented by an universal normal form. In particular—in this paper—we have been concerned with a normal form having a limit point. This type of universal *evolution equation* appears when a system shows hysteresis in a first-order like nonequilibrium phase transition. This situation occurs, for example, in explosive non-homogeneous thermochemical reactors. We have used a multiple-scale transformation to obtain from the Frank–Kamenetskii unimolecular exothermic model its corresponding normal form (2.15). Then, from the stochastic version of this amplitude equation we have inferred, from criticality, the stochastic propagation of the flame front.

The present theoretical approach was carried out generalizing the stochastic path perturbation approach for non-homogeneous problems. Thereby the first-passage time distribution for each Fourier mode $\Pi(t_k)$ were characterized by analytical expressions. In particular, the mean first-passage time, i.e. the timescale characterizing the *lifetime* of each Fourier mode $\mathcal{T}_k$, was found as a function of all the physical parameters of the problem (3.36). Thus, using this *effective* theoretical approach we have succeeded in characterizing the spatial fluctuations $\sigma_\phi(t)$, and the random thermal ignition in non-homogenous physicochemical reactors without consumption. The interesting situation including reactant consumption is under investigation.

Let us finish with the quotation of a dark philosopher. 'All things are part of one primary substance, fire.' (Heraclitus, 540–475 BC.)

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Appendix A. Stationary states of the Frank–Kamenetskii model

In order to clarify the type of non-homogeneous stationary solutions that may appear in (2.3), let us introduce here a *piecewise linear* approximation of the function $f(\theta)$, i.e., considering the Arrhenius reaction model, for $f(\theta)$ we can approximate it by the nonlinear function

$$f(\theta) = Au(\theta - \theta_c) + 1 \tag{A1}$$

where $u(\theta - \theta_c)$ is the Heaviside function. Using this expression in $0 = \frac{\partial^2}{\partial\rho^2}\theta + \delta f(\theta)$ it is possible to see that there exist three stationary solutions θ_{St}, fulfilling $\theta_{St}(\pm 1) = 0$

$$\theta_{St} = \theta_{cold} \qquad \text{if} \quad \delta \leqslant 2\theta_c = \delta_c \tag{A2}$$

$$\theta_{St} = \theta_{hot} \qquad \text{if} \quad \delta \geqslant \theta_c \left[\frac{(A-1)^2}{4(A - \frac{1}{2})} + \frac{1}{2} \right]^{-1} = \delta_b \tag{A3}$$

$$\theta_{St} = \theta_{unst} \qquad \text{if} \quad \delta_b \leqslant \delta \leqslant 2\theta_c = \delta_c. \tag{A4}$$

Here the *hot* and the *unstable* profiles $\theta_{hot}, \theta_{unst}$ are even functions given by

$$\theta_{\substack{hot\\unst}}(\rho) = \begin{cases} -\dfrac{A\delta}{2}\rho^2 - (p_{\substack{h\\u}})^2 \delta \dfrac{(A-1)}{2} + \dfrac{\delta}{2} + (p_{\substack{h\\u}})\delta(A-1) & \forall \quad 0 \leqslant \rho \leqslant (p_{\substack{h\\u}}) \\ \dfrac{\delta}{2}(1-\rho^2) + (p_{\substack{h\\u}})\delta(A-1)(1-\rho) & \forall \quad (p_{\substack{h\\u}}) \leqslant \rho \leqslant 1 \end{cases} \tag{A5}$$

where

$$p_{\substack{h\\u}} = \frac{\delta(A-1)\pm\sqrt{\delta^2(A-1)^2 - 4\delta(A-\frac{1}{2})(\theta_c - \frac{\delta}{2})}}{2\delta(A-\frac{1}{2})}. \tag{A6}$$

The *cold* profile is characterized by the (even) function

$$\theta_{cold}(\rho) = -\frac{\delta}{2}(\rho-1)(\rho+1) \qquad \forall \quad \rho \in [0, 1]. \tag{A7}$$

We see that the analogy with the homogeneous Semenov model is entirely similar [4, 13, 14]. Due to the fact that the eingenvalue in the linear stability analysis of (2.3) is null in θ_{St}, it is not possible to perform a perturbation analysis in order to study—analytically—which of the solutions θ_{St} are asymptotically stable or unstable. Therefore, we have performed a numerical study of these solutions in order to classify which of them were stable or not, thus concluding that θ_{cold} and θ_{hot} are the stable solutions, and θ_{unst} is the unstable one. From this analysis we see that for $\delta \in [\delta_b, \delta_c]$ one finds two branches of coexisting stationary states having opposite stability properties, in particular at the point δ_c the unstable and the cold branch *collide* at θ_c and subsequently they are annihilated; for this reason this point can be called a *limit point*. We should remark that a similar conclusion can also be obtained for the more realistic Arrhenius model $f(\theta)$, but the present *piecewise linear* model allows us to get a simpler and analytical description of the non-homogeneous bifurcation scenario. We remark that in all the paper we will be interested in the temporal evolution of the *flame front* coming from the vicinity of the critical point (θ_c, δ_c), i.e. its normal form and not on the detailed structure of $f(\theta)$.

Appendix B. Escape times for the ϕ_0 Fourier mode

Here we summarize the results for the FPTD $P(t_e)$, which can be obtained from the analysis of the normal form associated to the (homogeneous) Semenov model. From the pole of $\phi_0(t_e) = \infty$ and scaling-out Wiener integrals (see (3.17)–(3.19)) the FPTD is [13]

$$P(t_e) = P(\Omega)\left|\frac{\mathrm{d}\Omega}{\mathrm{d}t_e}\right| \tag{B1}$$

$$P(t_e) = \sqrt{\frac{3}{2\pi}}\exp\left[-\frac{3}{2}\frac{\tau^3}{t_e^3}\sqrt{\frac{K}{8}}\left(1-\frac{t_e^2}{\tau^2}\right)^2\right]\left|\frac{\mathrm{d}\Omega}{\mathrm{d}t_e}\right| \tag{B2}$$

where

$$\left|\frac{\mathrm{d}\Omega}{\mathrm{d}t_e}\right| = \frac{1}{2\tau}\left(\frac{K}{8}\right)^{1/4}\left[\left(\frac{\tau}{t_e}\right)^{1/2} + 3\left(\frac{\tau}{t_e}\right)^{5/2}\right] \tag{B3}$$

and the deterministic escape time τ is (in the SPPA, factor $\frac{\pi}{\sqrt{8}}$ comes from a remormalization procedure [13])

$$\tau = \frac{\pi}{\sqrt{8}}\sqrt{\frac{2}{ab}} \tag{B4}$$

and the constant K measures the departure from the marginal case

$$K = \frac{b^3}{a\varepsilon^2}. \tag{B5}$$

The small-noise approximation invoked to get (B2) can be measured with the universal parameter $K \gg 1$.

The case when $b = 0$ (marginal) has to be worked out in a second-order perturbation theory in order to improve the statistics of the FPTD [14]. Nevertheless, the term $aY_0 \sum_{n=-\infty(n\neq 0)}^{\infty} \frac{H_n}{Y_n}\frac{H_{-n}}{Y_{-n}}$ in (3.8) is of the same order $\mathcal{O}(\varepsilon)$. So in order to be able to get a closed iterative procedure we have to neglected this contribution from the non-homogeneous modes. On the other hand, it is possible to see that this term will produce only a renormalization of $\mathcal{O}(\varepsilon)$; therefore we expect that our result (neglecting this term) will not be so wrong from the simulations. In fact our figure 2(*c*) shows that the predicted statistics are quite good. It is possible to understand heuristically why this term is important in the marginal case: this is so because only when $b = 0$ the *potential* is so flat that the diffusion term turns out to be crucial for the escape of the unstable state $\phi = 0$. Therefore, for the marginal case we go one step further and introduce the approximation of neglecting the term $aY_0 \sum_{n=-\infty(n\neq 0)}^{\infty} \frac{H_n}{Y_n}\frac{H_{-n}}{Y_{-n}}$ in (3.8). Hence the escape process is now approximated to $\mathcal{O}(\varepsilon)$ by the stochastic paths

$$\phi_0(t) \cong \frac{\sqrt{\varepsilon}W_0(t)}{1 - a\sqrt{\varepsilon}\Omega_0(t) + a^2\varepsilon\Theta_0(t)} \tag{B6}$$

where $W_0(t)$ and $\Omega_0(t)$ are as before (see (3.18) and (3.19)). Here the non-Gaussian process $\Theta_0(t)$ is characterized by

$$\Theta_0(t) = \int_0^t \eta_0(t')\,\mathrm{d}t' \tag{B7}$$

where the stochastic process $\eta_0(t)$ is defined by

$$\eta_0(t) = \int_0^t \Omega_0(t')\,\mathrm{d}W(t'). \tag{B8}$$

In this case the process $\Theta_0(t)$ scales like $t^3\Theta_0$, where Θ_0 is a non-Gaussian random variable [14]. Then from the pole of $\phi_0(t_e) = \infty$ and scaling-out all the integrals, see (B6)–(B8), the FPTD for the marginal case reads [14]

$$P(t_e) = \frac{3^{3/2}}{a\sqrt{2\pi\varepsilon}} \exp\left(\frac{-3}{2a^2\varepsilon t_e^3}\right) C(t_e) \tag{B9}$$

where $C(t_e)$ gives the second-order correction

$$C(t_e) = \frac{\varphi\sqrt{\chi/\pi}}{2(\varphi+\chi)} \exp\left(\frac{1}{\varphi+\chi}\right) \left\{ \frac{\sqrt{\pi}(2\varphi+\chi)}{\varphi\sqrt{\varphi+\chi}} \left[1 + \mathrm{erf}\left(\frac{1}{\sqrt{\varphi+\chi}}\right)\right] + \exp\left(\frac{-1}{\varphi+\chi}\right)\right\}$$

and

$$\varphi \equiv \varphi(t_e) = \frac{2a^2\varepsilon t_e^3}{3} \qquad \chi \equiv \frac{360}{9}.$$

In [13,14] we have shown the good agreement of (B2) and (B9) with Monte Carlo simulations.

Appendix C. Small fluctuations

Here we analyse the *space-average* fluctuations $\mathcal{O}(\varepsilon)$ of the processes $\phi_k(t)$, for the Fourier numbers $k \neq 0$ at earlier times $t < t_e$. In order to do this we note from the first stage of evolution, up to $\mathcal{O}(\sqrt{\varepsilon})$, that $H_k(t) \sim \mathcal{O}(\sqrt{\varepsilon})W_k(t)$ and $Y_k(t) \sim \mathcal{O}(1)$, i.e. $Y_k(t)$ is independent of the process $H_k(t)$. Using the fact that different Wiener processes $W_k(t)$ are statistically independent of each other, the *coupling* selection modes appearing in (3.23), and from the previously mentioned initial (first) stage of evolution, it follows that the average of each Fourier mode ϕ_k can be approximated by

$$\overline{\phi_k}(t)_{t<t_e} = \left\langle \frac{H_k(t)}{Y_k(t)} \right\rangle \cong Y_k(t)^{-1}\overline{H_k}(t). \tag{C1}$$

On the other hand, it is simple to see, using (3.23), that if k is odd $\overline{H_k}(t) = 0$. Then for any odd Fourier number it follows that the spatial average of the Fourier modes are null:

$$\overline{\phi_{2k+1}}(t)_{t<t_e} = 0. \tag{C2}$$

Nevertheless, for even Fourier modes using (3.23), (C1) and the fact that $\langle W_{k'}(t')^2\rangle = t'$ it is possible to see that

$$\overline{\phi_{2k}}(t)_{t<t_e} = \frac{4a\varepsilon \exp[-\alpha_{2k}t]}{\alpha_{2k}^2}\left\{\exp\left[\frac{\alpha_{2k}}{2}t\right]\left(\frac{\alpha_{2k}}{2}t-1\right)+1\right\} \tag{C3}$$

showing, on the average, a small increasing behaviour, of $\mathcal{O}(\varepsilon)$, which finally extinguishes for large t.

Even when there could be small random fluctuations located any where in the domain $\tilde{x} \in [-L, L]$, on average only even Fourier modes are non-zero. On the other hand, from the FPTD $\Pi(t_k)$ we know that the exponential *growth* of each Fourier mode $\phi_k(t)$ can only occur after the random escape time t_e and if $k \in [\pm 1, \ldots, \pm k^*]$. Interestingly, (C3) tells us that for any *even* k the average $\overline{\phi_k}(t)$ has a small increasing transient, $\mathcal{O}(\varepsilon)$, for times $t < t_e$, i.e. the signature of small fluctuations.

Appendix D. Monte Carlo simulations

In order to test our theoretical predictions we have carried out numerical simulations to calculate the anomalous fluctuations $\sigma_\phi(\tilde{t})$ of the stochastic field $\phi(\tilde{x}, \tilde{t})$. Note that our analytical expression (3.42) is given in terms of all the physical parameters of the system and in addition in terms of the set of numbers $\{\mathcal{E}_k\}$, which are none other than the Fourier weights characterizing the hot attractor $\theta_{hot}(\tilde{x})$ (see (3.39)).

A given model for the velocity rate $k(T)$ characterizes the unimolecular chemical reaction, so this rate characterizes the nonlinear function $f(\theta)$, which ultimately leads to the fine structure of the hot attractor $\theta_{hot}(\tilde{x})$. In appendix A we have shown that a *piecewise linear* approximation of the function $f(\theta)$ is enough to represent the behaviour of the non-homogeneous stationary states $\theta_{St}(\tilde{x})$. In this appendix we go one step further in the approximation procedure and we calculate the set $\{\mathcal{E}_k\}$ from the attractor solution of

$$0 = D\partial_{\tilde{x}}^2\phi_{at}(\tilde{x}) + a\phi_{at}(\tilde{x})^2 - c\phi_{at}(\tilde{x})^3 + b \qquad \tilde{x} \in [-L, L] \tag{D1}$$

where $\phi_{at}(\tilde{x})$ fulfils zero-boundary condition on $\mathcal{D}_x$, i.e. $\phi_{at}(\pm L) = 0$. This attractor can be Fourier-transformed:

$$\phi_{at}(\tilde{x}) = \sum_{k=0,1,2,\ldots} \mathcal{E}_k \cos\left(\frac{k\pi\tilde{x}}{L}\right) \qquad \tilde{x} \in [-L, L]. \tag{D2}$$

Then the set of numbers $\{\mathcal{E}_k\}$ depends strongly on the values of a, b, c, D, and L. For $D = 5$, $a = 5$, $b = 0.1$, $c = 0.1$, and $L = 1$ some of the values of the set $\{\mathcal{E}_k\}$ are

$$\mathcal{E} \equiv \mathcal{E}_0 = 43.9 \qquad \mathcal{E}_1 = 18.14 \qquad \mathcal{E}_2 = -10.22 \qquad \mathcal{E}_3 = 5.19$$
$$\mathcal{E}_4 = -2.76 \qquad \mathcal{E}_5 = 1.64 \qquad \mathcal{E}_6 = -1.09 \qquad \mathcal{E}_7 = 0.80 \qquad \text{etc}$$

so the effectives $\mathcal{E}_k$ can be considered from $\mathcal{E}_0$ up to $\mathcal{E}_3$.

Starting from the initial condition $\phi(\tilde{x}, 0) = 0$, the dynamics toward the attractor $\phi_{at}(\tilde{x})$ represents the stochastic evolution, or the *escape* of the amplitude ϕ to the hot attractor of our thermochemical explosive system. Following remarks 1 and 2 (section 2.1) the approach toward the attractor $\phi_{at}(\tilde{x})$ is triggered by the noise $\mathcal{O}(\sqrt{\varepsilon})$ and this dynamics is characterized by the FPTD $\Pi(t_k)$ given in appendix B for $P(t_e)$, and section 3.2 (for $k \neq 0$). The Monte Carlo simulations have been carried out considering (3.3), which in discrete-time representation are characterized by the set of equations

$$\Delta\phi_k = \left[a \sum_{n=-\infty}^{\infty} \phi_n\phi_{k-n} + b\delta_{0,k} - \left(\frac{k\pi}{L}\right)^2 D\phi_k\right]\Delta t + \Delta W_k \tag{D3}$$

$$\phi_k(t + \Delta t) = \phi_k(t) + \Delta\phi_k(t). \tag{D4}$$

Here ΔW_k is the random contribution given by $\Delta W_k = \sqrt{\varepsilon\Delta t}\eta_i$, where η_i are Gaussian random variables such that $\langle\eta_i\rangle = 0$ and $\langle\eta_i\eta_l\rangle = \delta_{il}$ (δ_{il} being the Kronecker delta). These random variables have been generated using the Box–Mueller algorithm.

As we said before, and in order to study the evolution away from the unstable state, we choose the initial condition $\phi_k(0) = 0$, and we consider the evolution (D4) for 10^2 Fourier modes reaching, if possible, to their respective saturation values $\{\mathcal{E}_k\}$, after which we leave these modes to keep only the fluctuations of the noise $\mathcal{O}(\sqrt{\varepsilon})$. All our simulations were performed using a time increment $\Delta t = 10^{-4}$ and running 10^5 Monte Carlo realizations. In figures 2(*a*) and (*b*) we have used the following set of parameters: $D = 5$, $a = 5$, $b = 0.1$, $c = 0.1$ and $\varepsilon(= 0.001; 0.003)$. Therefore, the universal parameter K and the deterministic time τ are characterized by

$$K = 200 \qquad \tau = 2.22 \qquad \text{if} \quad \varepsilon = 0.001$$
$$K = 20.22 \qquad \tau = 2.22 \qquad \text{if} \quad \varepsilon = 0.003.$$

In figure 2(*c*) we also show the marginal situation ($b = 0$) for this case we use $\varepsilon = 0.04$ which means $a\sqrt{\varepsilon} = 1$. In this case the set $\{\mathcal{E}_k\}$ has similar values as before, for example:

$$\mathcal{E} \equiv \mathcal{E}_0 = 44.01 \qquad \mathcal{E}_1 = 18.20 \qquad \mathcal{E}_2 = -10.25 \qquad \mathcal{E}_3 = 5.20 \qquad \text{etc.}$$

In all the situations our predictions are in good agreement with the simulations.

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