
Appendices

A Phase and amplitude variables

In Chapter 3, chemical turbulence in oscillatory systems is analyzed based on a phase and amplitude representation of the experimental results. Spatial distributions of phase and amplitude variables are computed from the experimental data employing a variant of the analytic signal approach [228, 229]. Using this method, a second variable is generated from the experimental time series by means of a Hilbert transform. The dynamics is then projected into the plane spanned by the experimental signal and its Hilbert transform.

The transformation is carried out as follows. For the experimental signal $s(\mathbf{x}, t)$, its Hilbert transform is obtained at each point \mathbf{x} in space,

$$\tilde{s}(\mathbf{x}, t) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{s(\mathbf{x}, t')}{t - t'} dt'. \quad (\text{A.1})$$

Here, $s(\mathbf{x}, t)$ is the intensity of the two-dimensional PEEM image, or the one-dimensional distribution of the double layer potential, respectively. The Hilbert transform can be easily computed by determining the Fourier transform of s , shifting each complex Fourier coefficient by $-\pi/2$, and performing the reverse Fourier transform [229]. Using $s(\mathbf{x}, t)$ and its Hilbert transform $\tilde{s}(\mathbf{x}, t)$, a complex variable can be defined,

$$\zeta(\mathbf{x}, t) = s(\mathbf{x}, t) + i\tilde{s}(\mathbf{x}, t), \quad (\text{A.2})$$

that is generally referred to as the *analytic signal* [228]. The time-dependent spatial distributions of the phase $\phi(\mathbf{x}, t)$ and amplitude $R(\mathbf{x}, t)$ are determined directly from the analytic signal (A.2). The phase is computed as $\phi = \arg \zeta$, representing the polar angle in the plane spanned by the variables s and \tilde{s} . The amplitude is defined as

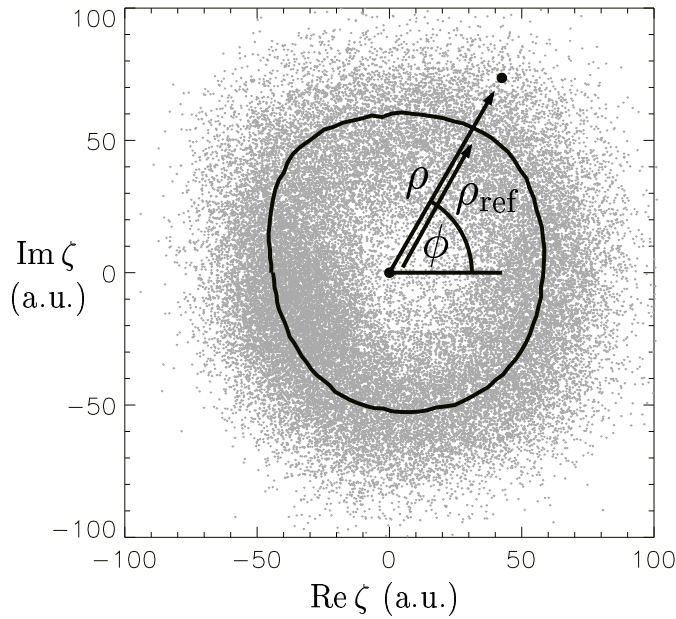


Figure A.1: Transformation to phase $\phi = \arg \zeta$ and amplitude $R = \rho/\rho_{\text{ref}}(\phi)$ variables based on the analytic signal ζ . The reference orbit $\rho = \rho_{\text{ref}}(\phi)$ is indicated by the closed line [214].

$R = \rho/\rho_{\text{ref}}(\phi)$ with $\rho = |\zeta|$ the modulus of the complex number ζ . Normalization of the amplitude by $\rho_{\text{ref}}(\phi)$ compensates for deviations of oscillations in the experiment from harmonic oscillations. The reference amplitude $\rho_{\text{ref}}(\phi)$ is obtained in the following way. The time series of $\zeta(\mathbf{x}, t)$ is plotted into the complex plane (for all locations \mathbf{x} in the case of the one-dimensional electrochemical data and for 50×50 equidistant sample points in the case of the two-dimensional PEEM data). This is illustrated in Fig. A.1 for a set of turbulent data from catalytic CO oxidation. The reference amplitude $\rho_{\text{ref}}(\phi)$ is determined as the arithmetic mean of $\rho = |\zeta|$ inside each of 100 equidistant intervals of the polar angle ϕ . The closed curve $\rho = \rho_{\text{ref}}(\phi)$ in the complex plane can be considered as a reference orbit of the system deduced from the experimental data.

B PDF of topological defects

In the following, a detailed derivation of the probability distribution function (PDF) of the number of topological defects is presented. Based on the gain and loss rates of defects, $k_+(n)$ and $k_-(n)$, the master equation for the probability $p(n, t)$ reads

$$\begin{aligned} \partial_t p(n, t) = & k_+(n-1) p(n-1, t) + k_-(n+1) p(n+1, t) \\ & - k_+(n) p(n, t) - k_-(n) p(n, t), \end{aligned} \quad (\text{B.1})$$

where n is the number of positive or negative defects, $n \equiv n_{\pm}$ (in first approximation, it is assumed that there are equal numbers of defects with positive and negative topological charge, $n_+ \approx n_-$). In the asymptotic regime, $\partial_t p(n, t) = 0$, Eq. (B.1) transforms into a recursive relation for the probability $p(n)$,

$$p(n) = \frac{k_+(n-1)}{k_-(n)} p(n-1). \quad (\text{B.2})$$

Based on simple assumptions (see Section 3.2.3) the gain and loss rates are approximated by the following expressions,

$$k_+(n) = c + d n \quad (\text{B.3})$$

$$k_-(n) = a n^2, \quad (\text{B.4})$$

where c denotes the constant contribution to the rate of creation, $d n$ takes into account replication of defects, and $a n^2$ is the rate of annihilation. Inserting Eqs. (B.3) and (B.4) into the recursion relation (B.2) yields

$$p(n) = \frac{c + d(n-1)}{a n^2} p(n-1), \quad (\text{B.5})$$

and can be expanded further to

$$p(n) = p(0) \prod_{k=1}^n \frac{c + d(k-1)}{a k^2} = \frac{p(0)}{(n!)^2} \frac{d^n}{a^n} \prod_{k=0}^{n-1} \left(\frac{c}{d} + k \right). \quad (\text{B.6})$$

Using the general relation $\prod_{k=0}^{n-1} (x+k) = \frac{\Gamma(x+n)}{\Gamma(x)} \equiv (x)_n$,

Eq. (B.6) can be simplified to

$$p(n) = p(0) \frac{\gamma^n}{(n!)^2} (\nu)_n = p(0) \frac{\gamma^n}{(n!)^2} \frac{\Gamma(\nu+n)}{\Gamma(\nu)} \quad (\text{B.7})$$

with $\gamma = \frac{d}{a}$ and $\nu = \frac{c}{d}$. Since $p(n)$ is a probability, normalization is required,

$$\sum_{n=0}^{\infty} p(n) = 1. \quad (\text{B.8})$$

Substituting $p(n)$ from Eq. (B.7) into Eq. (B.8) leads to

$$p(0) \sum_{n=0}^{\infty} \frac{\gamma^n}{(n!)^2} (\nu)_n = p(0) \sum_{n=0}^{\infty} \frac{\gamma^n}{n!} \frac{(\nu)_n}{(1)_n} = 1, \quad (\text{B.9})$$

and thus

$$p(0) = \frac{1}{\Phi(\nu, 1; \gamma)}, \quad (\text{B.10})$$

where Φ denotes the degenerate hypergeometric function defined as [230]

$$\Phi(\alpha, \beta; z) = 1 + \frac{\alpha}{\beta} z + \frac{\alpha(\alpha+1)}{\beta(\beta+1)} \frac{z^2}{2!} + \dots = \sum_{n=0}^{\infty} \frac{(\alpha)_n}{(\beta)_n} \frac{z^n}{n!}. \quad (\text{B.11})$$

Taking into account normalization, the probability distribution finally reads

$$\boxed{p(n) = Q \frac{\gamma^n \Gamma(\nu+n)}{(n!)^2}} \quad \text{with} \quad Q = \frac{1}{\Phi(\nu, 1; \gamma) \Gamma(\nu)}. \quad (\text{B.12})$$

From the distribution (B.12) the mean value can be determined,

$$\begin{aligned}
 \langle n \rangle &= \sum_{n=0}^{\infty} n p(n) \\
 &= Q \sum_{n=0}^{\infty} n \frac{\gamma^n}{(n!)^2} \Gamma(\nu + n) \\
 &= Q \sum_{n=0}^{\infty} (n+1) \frac{\gamma^{n+1}}{((n+1)!)^2} \Gamma(\nu + n + 1) \\
 &= Q \sum_{n=0}^{\infty} \frac{\gamma^n \gamma}{n!(n+1)!} \Gamma(\nu + 1 + n) \\
 &= \frac{\gamma \Gamma(\nu + 1)}{\Phi(\nu, 1; \gamma) \Gamma(\nu)} \sum_{n=0}^{\infty} \frac{\gamma^n}{n!} \frac{(\nu + 1)_n}{(2)_n}
 \end{aligned}$$

$$\boxed{\langle n \rangle = \frac{\gamma^\nu}{\Phi(\nu, 1; \gamma)} \Phi(\nu + 1, 2; \gamma)}. \quad (\text{B.13})$$

By a similar derivation, the second moment, $\langle n^2 \rangle = \sum_{n=0}^{\infty} n^2 p(n)$, is obtained,

$$\boxed{\langle n^2 \rangle = \frac{\gamma^\nu}{\Phi(\nu, 1; \gamma)} \Phi(\nu + 1, 1; \gamma)}. \quad (\text{B.14})$$

C Phase equation in the presence of TDAS

Close to the onset of oscillations, the system of ordinary differential equations that describes a specific oscillator, may be reduced to a simple universal equation for the complex oscillation amplitude $\eta(t)$, which is known as the Stuart-Landau equation (see also Section 2.4.1). Here, the phase dynamics equation for a single oscillator in the presence of time-delay autosynchronization (TDAS) will be derived starting from the general Stuart-Landau model. The Stuart-Landau equation with an additional TDAS reads

$$\dot{\eta} = (1 - i\omega_0)\eta - (1 + i\beta)|\eta|^2\eta + F(t), \quad (\text{C.1})$$

where $F(t) = \mu e^{i\chi}[\eta(t) - \eta(t - \tau)]$. Here, ω_0 is the frequency of small-amplitude oscillations, the coefficient β determines the nonlinear frequency shift, and the parameter χ accounts for a possible phase shift in the application of the control force. The feedback intensity is specified by the coefficient μ . It is assumed that the feedback is weak, $\mu \ll 1$. The Stuart-Landau equation (C.1) is written in a dimensionless form, by choosing as a time unit the relaxation time which is proportional to the distance from the bifurcation point. Since this relaxation time diverges at the Hopf bifurcation, whereas the oscillation period remains finite, it is $\omega_0 \gg 1$ in the neighborhood of the Hopf bifurcation.

Through the ansatz $\eta(t) = \rho(t)e^{-i\phi(t)}$ for the complex oscillation amplitude η , the oscillation phase ϕ is introduced and the feedback term can be written as

$$F(t) = \mu e^{i\chi}[\rho(t)e^{-i\phi(t)} - \rho(t - \tau)e^{-i\phi(t - \tau)}]. \quad (\text{C.2})$$

Substituting (C.2) into Eq. (C.1) and separating real and imaginary parts, the following equations for the real amplitude $\rho(t)$ and for the phase $\phi(t)$ are obtained,

$$\dot{\rho} = \rho(1 - \rho^2) + \mu[\rho(t)\cos\chi - \rho(t - \tau)\cos(\chi + \Delta\phi)], \quad (\text{C.3})$$

$$\dot{\phi} = \omega_0 + \beta\rho^2 - \mu\left[\sin\chi - \frac{\rho(t - \tau)}{\rho(t)}\sin(\chi + \Delta\phi)\right], \quad (\text{C.4})$$

where $\Delta\phi(t) = \phi(t) - \phi(t - \tau)$. Since the feedback is weak ($\mu \ll 1$), it leads to only

small variations of the real amplitude ρ , so that $\rho(t) = 1 + \delta\rho(t)$ where $\delta\rho$ is of order μ . Therefore, Eq. (C.3) approximately yields

$$\dot{\delta\rho} = -2\delta\rho + \mu[\cos\chi - \cos(\chi + \Delta\phi)]. \quad (\text{C.5})$$

Thus, small perturbations of ρ adjust to the variation of $\Delta\phi$ within the relaxation time of order unity. On the other hand, the variations of $\Delta\phi$ are slow and are characterized by the time scales of order $1/\mu$ which are much larger than unity. Hence, adiabatic approximation can be used leading to

$$\delta\rho = \frac{\mu}{2}[\cos\chi - \cos(\chi + \Delta\phi)]. \quad (\text{C.6})$$

Substituting this result into Eq. (C.4) and again retaining only the terms linear in μ , the following approximate phase dynamics equation is derived

$$\dot{\phi} = \omega + \mu f(\Delta\phi). \quad (\text{C.7})$$

The function $f(\Delta\phi)$ in this equation is given by

$$f(\Delta\phi) = a \sin(\Delta\phi) + b \cos(\Delta\phi) - b, \quad (\text{C.8})$$

where the notations $\omega = \omega_0 + \beta$, $a = \cos\chi + \beta \sin\chi$, and $b = \sin\chi - \beta \cos\chi$ are introduced. It can be easily checked that the variations of $\Delta\phi$ are indeed slow. The slow phase variable $\varphi(t) = \phi(t) - \omega t$ can be defined. Note that in this notation $\Delta\phi = \Delta\varphi + \omega\tau$. Now, Eq. (C.7) can be written as

$$\dot{\varphi} = \mu f(\Delta\varphi + \omega\tau). \quad (\text{C.9})$$

Thus, the rate of variation of φ is proportional to the feedback intensity which is small, $\mu \ll 1$. Because $\Delta\phi$ differs from $\Delta\varphi$ only by a constant, the same is true for $\Delta\phi$.

Although the phase dynamics approximation (C.7) was derived from the Stuart-Landau equation, a similar equation can be found for any limit-cycle oscillator under the action of weak TDAS. In general, the 2π -periodic function $f(\Delta\phi)$ then takes different forms satisfying the condition $f(0) = 0$.

D Numerical methods

Two different programming tools were employed to carry out numerical simulations. On the one hand, a number of small integrators was written in FORTRAN 77. They were used to study the phase dynamics equation, the complex Ginzburg-Landau equation, and the realistic model of catalytic CO oxidation under uniform global feedback in Chapter 5. On the other hand, simulations of the CO oxidation model under nonuniform global feedback (Chapter 6) were performed using PV-WAVE¹, an array based programming language with a large number of built-in functions for signal and image processing.

In all cases, model equations were integrated in time by an explicit Euler method. Simulations in Section 5.1 did not involve any spatial degrees of freedom. In Section 5.2 and Chapter 6, all simulations were carried out on a one-dimensional domain (except for one series of two-dimensional simulations indicated in the text). An equidistant grid and periodic boundary conditions were used throughout. Second order finite-differences were employed to discretize the Laplacian operator. The numerical parameters such as the separation between grid points or the size of the time step are specified in the text, as they are different for the various models.

¹ a product of Visual Numerics, Inc.