Term Paper - PHYS 7224

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Abstract

This term paper is a report on some results obtained in the paper "Hamilton-Jacobi method for molecular distribution function in a chemical oscillator" by Nakanishi, Sakaue, and Wakou [1]. None of the material presented here is new work by the author of this term-paper.

1 Introduction

The basic model is a Markov process for the evolution of species in a chemical reaction. Our state variable is $\{X_i(t)\}$, which represents the number of molecules of the $i-$th chemical species, where $i = 1, ...d$. $\mathbf{X}(t) = \{X_i(t)\}$ evolves in time as a stochastic process due to inflow, outflow, and reaction processes. We can represent them all by "reactions" indexed as $\rho = 1, 2, ...r$. Let the system volume be denoted by Ω. In general, the reaction rate of the $\rho-$th reaction $W_\rho = W_\rho(\mathbf{X})$ depends on the concentration of chemicals $\mathbf{X}$ and Ω.

The master equation for the probability distribution $\bar{P}(\mathbf{X}, t)$ at time t can be written as follows:

$$\frac{d\bar{P}(\mathbf{X}, t)}{dt} = \sum_{\rho=1}^{r}[W_\rho(\mathbf{X} - \Delta\mathbf{X}_\rho)\bar{P}(\mathbf{X} - \Delta\mathbf{X}_\rho) - W_\rho(\mathbf{X})\bar{P}(\mathbf{X})] \tag{1}$$

where $\Delta\mathbf{X}_\rho$ is the change in $\mathbf{X}$ due to the reaction ρ. The first and second terms in the $\rho-$th sum, can be interpreted as the gain and loss terms, respectively. Notice that the above equation preserves the normalization $\sum_{\{X_i\}} \bar{P}(\mathbf{X}, t) = 1$.

2 The Model

Let's start with the master equation (1). Define the intensive parameter $\mathbf{x} = \frac{\mathbf{X}}{\Omega}$. The next step is to obtain an evolution equation for the probability distribution $P(\mathbf{x}, t) := \Omega^d \bar{P}(\mathbf{X}, t)$ in the large Ω regime or "weak noise" regime. In this regime, the jumps in the Markov process become relatively small, and hence we can approximate the function $W_\rho(\mathbf{X} - \Delta\mathbf{X}_\rho)\bar{P}(\mathbf{X} - \Delta\mathbf{X}_\rho)$ through a truncated Taylor expansion. We get the generalized Fokker-Planck equation

$$\frac{1}{\Omega}\frac{\partial P(\mathbf{x}, t)}{\partial t} = -\frac{1}{\Omega}\sum_i \frac{\partial}{\partial x_i}\{[F_i(\mathbf{x}) + \frac{1}{\Omega}G_i(\mathbf{x})]P(\mathbf{x}, t)\} + \frac{1}{\Omega^2}\sum_{i,j}\frac{\partial^2}{\partial x_i \partial x_j}\{Q_{ij}(\mathbf{x})P(\mathbf{x}, t)\} \tag{2}$$

where

$$F_i(\mathbf{x}) = \lim_{\Omega\to\infty}\frac{1}{\Omega}\sum_{\rho=1}^{r}\Delta X_\rho^i W_\rho(\Omega\mathbf{x})$$

$$Q_{ij}(\mathbf{x}) = \lim_{\Omega\to\infty}\frac{1}{2\Omega}\sum_{\rho=1}^{r}\Delta X_\rho^i \Delta X_\rho^j W_\rho(\Omega\mathbf{x})$$

$$G_i(\mathbf{x}) = \lim_{\Omega\to\infty}[\sum_{\rho=1}^{r}\Delta X_\rho^i W_\rho(\Omega\mathbf{x}) - \Omega F_i(\mathbf{x})]$$

This evolution preserves the probability $\int P(\mathbf{x}, t)d\mathbf{x} = 1$. Note that the deterministic macroscopic time evolution is governed by the rate equation

$$\frac{d\mathbf{x}}{dt} = \mathbf{F}(\mathbf{x}) \tag{3}$$

In the following, we use the Hamilton-Jacobi approach to find approximate solutions of the Fokker-Planck equation (2), which signify the evolution of the noise about the rate equation orbit.

3 The Hamilton-Jacobi Approach

Assume a solution to (2) of the form $P(\mathbf{x},t) = e^{\Omega\phi(\mathbf{x},t)}$. Substituting this in (2) and only retaining the leading term, we end up with having to solve the following Hamilton-Jacobi equation:

$$\frac{\partial \phi}{\partial t} + H(\mathbf{x}, \frac{\partial \phi}{\partial \mathbf{x}}) = 0 \tag{4}$$

where $H(\mathbf{x},\mathbf{p}) = \sum p_i F_i(\mathbf{x}) + \sum p_i p_j Q_{ij}(\mathbf{x})$. Let us assume that the initial distribution is given by $\phi(\mathbf{x},0) = \phi_0(\mathbf{x})$. To solve (4), we study the flow in the $\mathbf{x}-\mathbf{p}$ space starting from the point $(\mathbf{x}_0,\mathbf{p}_0)$, where $p_i(0) = \frac{\partial \phi}{\partial x_i}|_{\mathbf{x}_0}$. The flow is given through the Hamiltonian H as follows:

$$\frac{d\mathbf{x}}{ds} = \frac{\partial H}{\partial \mathbf{p}} = F_i(\mathbf{x}) - \sum_j 2Q_{ij}(\mathbf{x})p_j$$

$$\frac{d\mathbf{p}}{ds} = -\frac{\partial H}{\partial \mathbf{x}} = -\sum_j \frac{\partial F_j(\mathbf{x})}{\partial x_i} p_j + \sum_{j,k} \frac{\partial Q_{jk}(\mathbf{x})}{\partial x_i} p_j p_k$$

Through this flow, we can express the function $\phi(\mathbf{x},t)$ as follows:

$$\phi(\mathbf{x},t) = \int_0^t ds(\sum p_i \dot{x_i} - H(\mathbf{x}(s),\mathbf{p}(s))) + \phi_0(\mathbf{x}) \tag{5}$$

To summarize, given $\phi(\mathbf{x},0)$, do the following to find $\phi(\mathbf{x},t)$:

1. Generate the initial conditions $\mathbf{x}_0$ and $\mathbf{p}_0 = \frac{\partial \phi}{\partial \mathbf{x}}|_{\mathbf{x}_0}$ such that $\mathbf{x}(t) = \mathbf{x}$.
2. Evolve along the Hamiltonian flow in the $\mathbf{x}-\mathbf{p}$ space until time t.
3. Plug in the result of the above into (5).

The relationship $\mathbf{p} = \frac{\partial \phi}{\partial \mathbf{x}}$ will be preserved through the evolution.

Remark 3.1. The case $\mathbf{p} = 0$ would mean that $\frac{\partial \phi(\mathbf{x})}{\partial \mathbf{x}} = 0$ and the Hamiltonian evolution lies on the invariant subspace $\mathbf{p} = 0$ and the evolution for $\mathbf{x}$ reduces to the rate equation (3). Hence, for instance, if this critical point is a maximum for ϕ at value $\mathbf{x}^*$, the motion of this peak in t is given by the rate equation flow.

We now apply this formalism to study the evolution of the initial distribution given by the Gaussian $\phi_0(\mathbf{x}) = -\frac{1}{2\sigma_0^2}(\mathbf{x}-\mathbf{x}_0)^2$ peaked at $\mathbf{x}_0^*$. The plan will be to take the standard deviation $\sigma_0 \to 0$ to understand the evolution of the Dirac delta at $\mathbf{x}_0^*$.

First, we find the solution $\mathbf{x}^*(t)$ of the rate equation (3) with initial condition $\mathbf{x}^*(0) = \mathbf{x}_0^*$. From the previous remark, we know that this will give us the peak of the distribution $\phi(\mathbf{x},t)$ at time t. We can now compute $\phi(\mathbf{x},t)$ in the so-called "Gaussian approximation" by Taylor-expanding it about its peak.

$$\phi(\mathbf{x},t) = \phi(\mathbf{x}^*(t),t) + (\mathbf{x}-\mathbf{x}^*(t))^T \frac{\partial \phi}{\partial \mathbf{x}}|_* + \frac{1}{2}\sum_{i,j} \frac{\partial^2 \phi}{\partial x_i \partial x_j}|_*(x_i - x_i^*(t))(x_j - x_j^*(t)) + ...$$

The linear term vanishes because $\mathbf{x}^*(t)$ is a local maximum. Also, note that the zeroth order term vanishes, which can be seen as follows: To reach $\mathbf{x}^*(t)$ we can simply follow the rate equation evolution which entails $\mathbf{p} = 0$. Using this in equation (5), the claim is proved.

We hence obtain:

$$\phi(\mathbf{x},t) \approx -\frac{1}{2}\sum_{i,j} \hat{M}_{ij}^{-1}(t)(x_i - x_i^*(t))(x_j - x_j^*(t)) \tag{6}$$

in the truncated expansion, where the matrix M is defined using the above. Thus, the distribution $P(\mathbf{x},t)$ given $\phi_0(\mathbf{x})$ is:

$$P(\mathbf{x},t) = Z \exp[-\frac{\Omega}{2}(\mathbf{x}-\mathbf{x}^*(t))^T \hat{M}^{-1}(t)(\mathbf{x}-\mathbf{x}^*(t))] \tag{7}$$

where Z is the normalization. It remains to see how the M can be estimated numerically:

From the Taylor expansion, we see that

$$-\hat{M}_{ij}^{-1}(t) = \frac{\partial^2 \phi(\mathbf{x}, t)}{\partial x_i \partial x_j}|_* = \frac{\partial p_j}{\partial x_i}|_*$$

The second inequality follows from the relationship $\mathbf{p} = \frac{\partial \phi}{\partial \mathbf{x}}$. To be more precise, the p_j in the above is $p_j(\mathbf{x}_0, t)$, where $\mathbf{x}_0$ is that initial condition which drives the evolution to the required point $\mathbf{x}$ at time t.

Hence, we need to study the object $\frac{\delta p_j}{\delta x_i}$ near the rate equation orbit $\mathbf{x}^*(t)$. To understand its behavior, we study the Hamiltonian flow linearized about $\mathbf{x}^*(t)$. Set $\delta\mathbf{x}(t) = \mathbf{x}(t) - \mathbf{x}^*(t)$ and $\delta\mathbf{p}(t) = \mathbf{p}(t)$ so that we get:

$$\delta\dot{\mathbf{x}}(t) = \hat{L}(t)\delta\mathbf{x}(t) - 2\hat{Q}_L(t)\delta\mathbf{p}(t) \tag{8}$$

$$\delta\mathbf{p}(t) = -\hat{L}^T(t)\delta\mathbf{p}(t) \tag{9}$$

Define the time evolution operator $\hat{U}^L(t, t_0)$ for the evolution of $\delta\mathbf{x}(t)$ within the $\mathbf{p} = 0$ subspace. Since $\hat{L}$ is just the Jacobian of vector field of the rate equation (3), $\hat{U}_L$ corresponds to the evolution of the deviation specified by the rate equation alone.

Then, a solution of (8) and (9) can be written as:

$$\delta\mathbf{x}(t) = \hat{U}_L(t, 0)[\delta\mathbf{x}(0) - 2\hat{Q}_L(t)\delta\mathbf{p}(0)] \tag{10}$$

$$\delta\mathbf{p}(t) = \hat{U}_L^T(0, t)\delta\mathbf{p}(0) \tag{11}$$

where $\hat{Q}_L(t) = \int_0^t du \hat{U}_L(0, u)\hat{Q}(u)\hat{U}_L^T(0, u)$.

We will now show how to compute the matrix $\frac{\delta\mathbf{p}(t)}{\delta\mathbf{x}(t)}$ from equations (10) and (11), in the limit $\sigma_0 \to 0$. For starters, what is this matrix at time 0?

From the relationship $\mathbf{p} = \frac{\partial \phi}{\partial \mathbf{x}}$, and using $\phi_0 = -\frac{1}{\sigma_0^2}(\mathbf{x} - \mathbf{x}_0)^2$, we get that $\delta\mathbf{p}(0) = -\frac{1}{2\sigma_0^2}\delta\mathbf{x}(0)$. Substituting this in equations (10) and (11), we end up with

$$\delta\mathbf{p}(t) = \hat{U}_L^T(0, t)(\frac{-1}{\sigma_0^2})[1 + \frac{2}{\sigma_0^2}\hat{Q}_L(t)]^{-1}\hat{U}_L(0, t)\delta\mathbf{x}(t)$$

Expanding the matrix inverse term in the bracket and taking the limit of small σ_0, finally, we get

$$\hat{M}(t) = \hat{U}_L(t, 0)2\hat{Q}_L(t)\hat{U}_L^T(t, 0)$$

How does this help us compute the matrix elements of M? Notice that the operators $\hat{U}_L$ and $\hat{Q}_L$ govern the evolution in equations (8) and (9). So all we need to do is solve those equations with appropriate initial conditions. More precisely,

$$\begin{aligned} \langle i|\hat{M}|j\rangle &= \langle i|\hat{U}_L(t, 0)2\hat{Q}_L(t)\hat{U}_L^T(t, 0)|j\rangle \\ &= \sum_\alpha \langle i|\hat{U}_L(t, 0)2\hat{Q}_L(t)|\alpha\rangle\langle\alpha|\hat{U}_L^T(t, 0)|j\rangle \end{aligned}$$

where $|\alpha\rangle$ are elements of an orthonormal basis of $\mathbb{R}^d$.

Now, notice from (10) that $-\hat{U}_L(t, 0)2\hat{Q}_L(t)|\alpha\rangle$ is the solution of (8), (9) with initial condition $\delta\mathbf{x} = 0$, $\delta\mathbf{p} = |\alpha\rangle$.

Similarly, from (11), we see that $\langle\alpha|\hat{U}_L^T(t, 0)$ is the transpose of the solution to (8), (9) with initial condition $\delta\mathbf{x} = |\alpha\rangle$, $\delta\mathbf{p} = 0$.

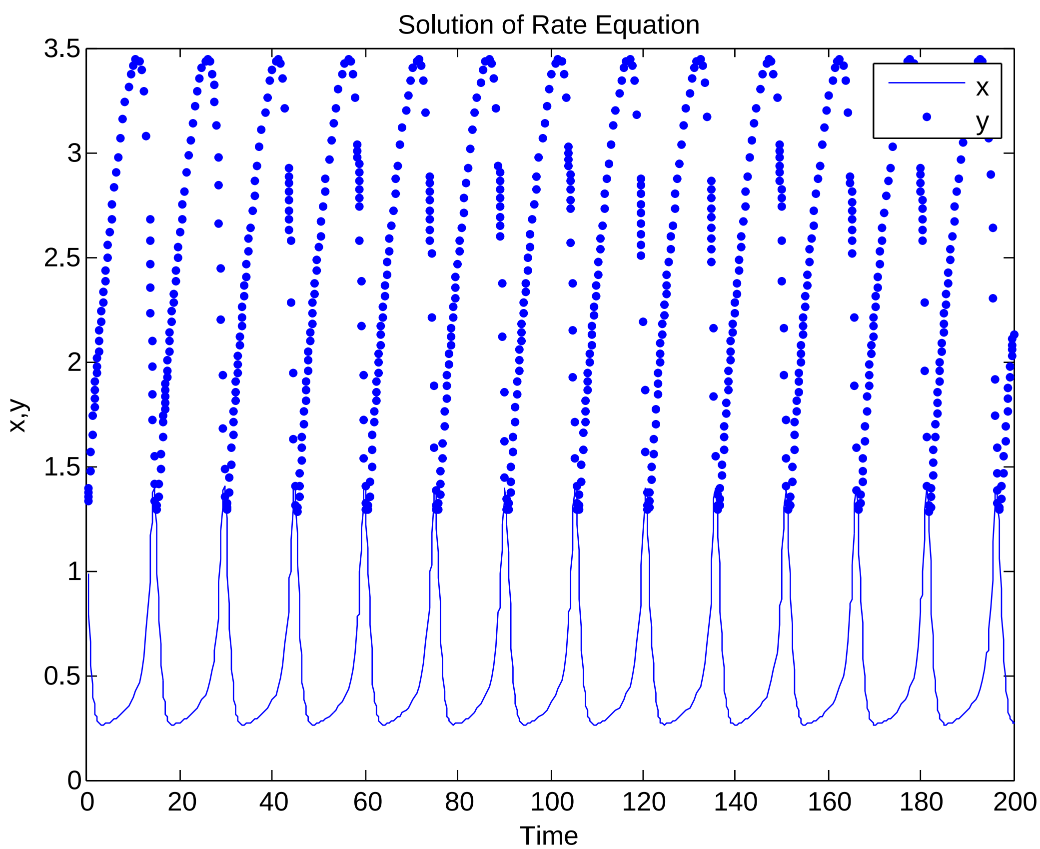
The conclusion is that we can obtain $\hat{M}$ by solving (8), (9) with appropriate initial conditions. In the next section, we apply this formalism to the Brusselator model.

4 Brusselator

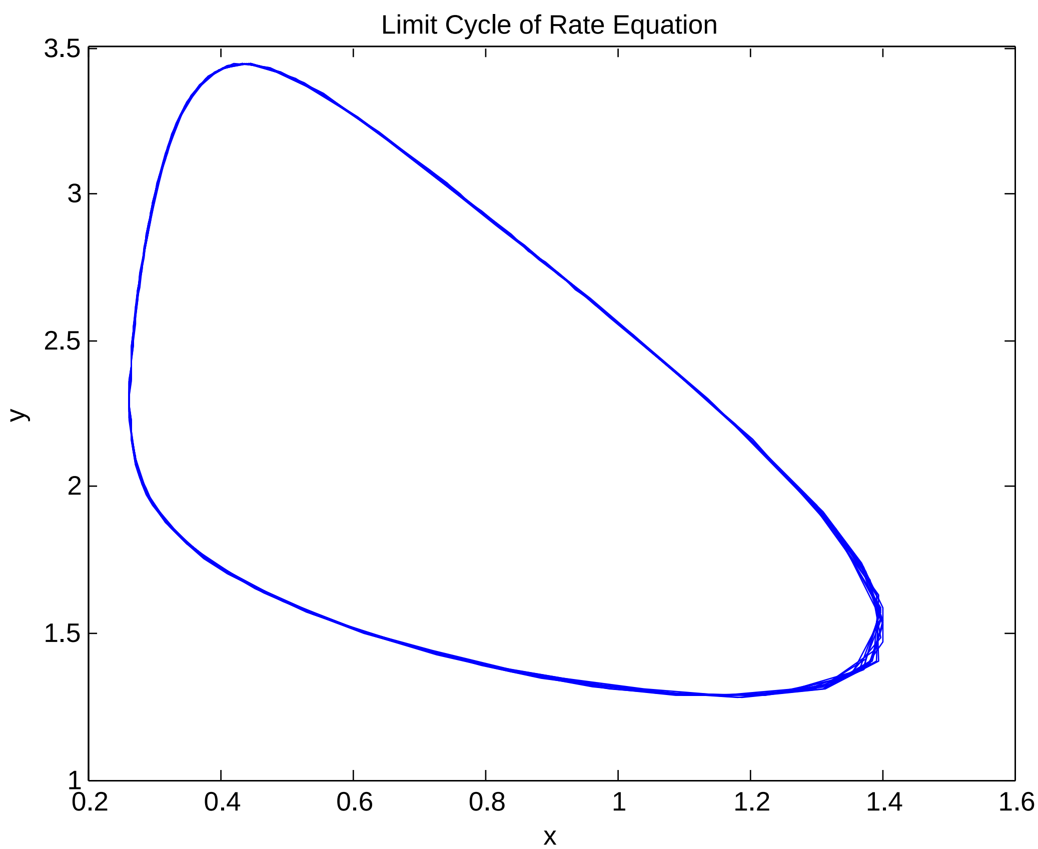
The Brusselator models the interaction of two species, whose concentrations are given by x and y, respectively. The macroscopic behaviour is governed by the rate equations (for a particular set of parameter values):

$$\frac{dx}{dt} = \frac{1}{2} - \frac{3}{2}x + x^2 y - x$$
$$\frac{dy}{dt} = \frac{3}{2}x - x^2 y$$

This system admits a stable periodic orbit $\mathbf{x}^*(t) = (x^*(t), y^*(t))$. See attached picture.



Solution of Rate Equation
x,y
Time
x
y
0
0.5
1
1.5
2
2.5
3
3.5
0
20
40
60
80
100
120
140
160
180
200


Limit Cycle of Rate Equation
3.5
3
2.5
2
1.5
1
y
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
x

It is expected the the fluctuations to this trajectory in the Gaussian approximation can be understood through the solutions of the corresponding generalized Fokker-Planck equation via the Hamilton-Jacobi approach. For this model:

$$Q_{xx} = \frac{1}{2}(\frac{1}{2} + \frac{3}{2}x + x^2y + x)$$

$$Q_{xy} = Q_{yx} = -\frac{1}{2}(\frac{3}{2}x + x^2y)$$

$$Q_{yy} = \frac{1}{2}(\frac{3}{2}x + x^2y)$$

The components of the matrix L, where $L_{ij}(t) = \frac{\partial F_i}{\partial x_j}|_{\mathbf{x}^*(t)}$ are:

$$L_{xx}(t) = -\frac{3}{2} + 2x^*(t)y^*(t) - 1$$

$$L_{xy}(t) = x^*(t)^2$$

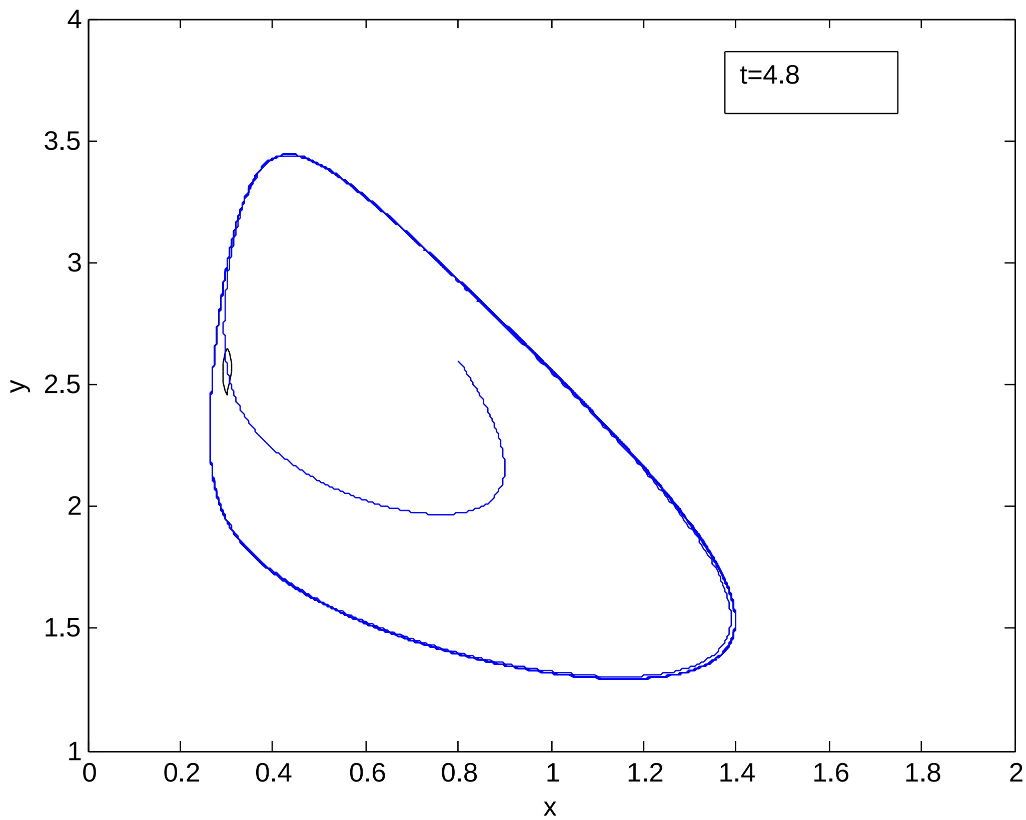
$$L_{yx}(t) = \frac{3}{2} - 2x^*(t)y^*(t)$$

$$L_{yy}(t) = -x^*(t)^2$$

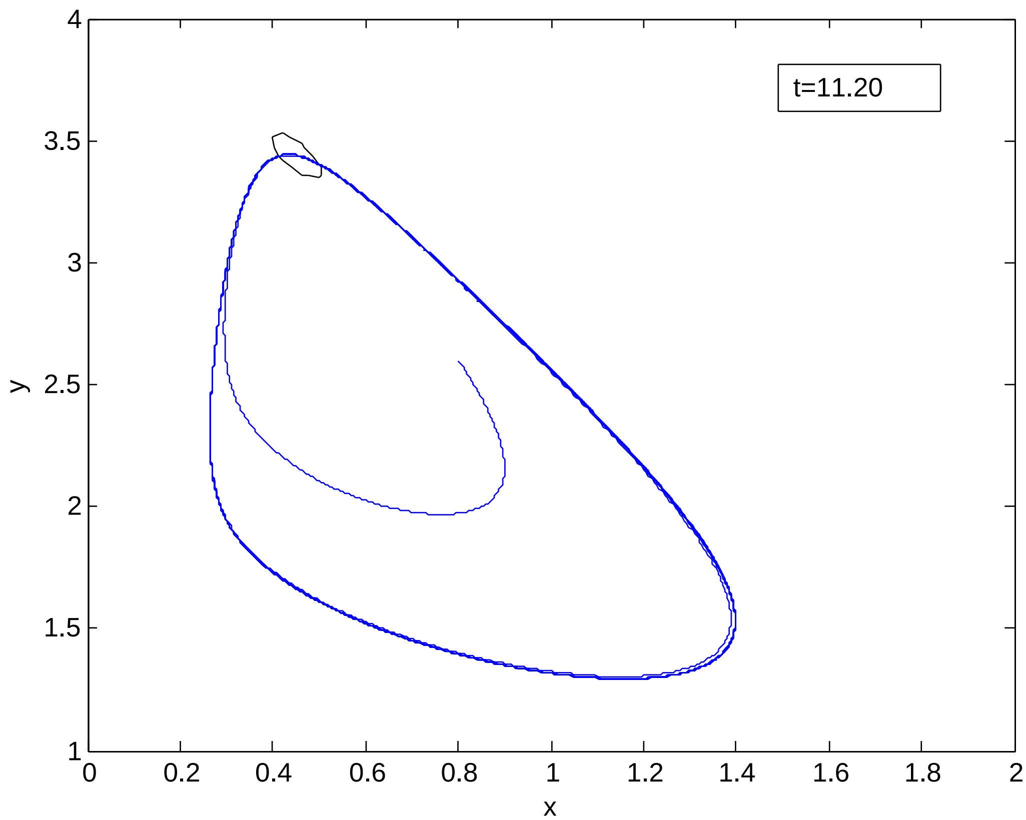
4.1 Numerical Results for the Brusselator

We simply re-generate some of the results in [1] using the ideas in [1]. Starting from an initial point $(0.8, 2.6)$, we obtain the covariance matrix $M(t)$ at different times using the method described in the previous section. They are depicted pictorially on a series of snapshots at selected times. The blue lines represent the rate equation orbit, which converges to the limit cycle. The black ellipses are graphs of the equation $\mu_{xx}(x - x^*(t))^2 + 2\mu_{xy}(x - x^*(t))(y - y^*(t)) + \mu_{yy}(y - y^*(t))^2 = \frac{4}{\Omega}$, where μ_{ij} are the elements of $\hat{M}^{-1}$. We choose, as in the paper, $\Omega = 10^4$.

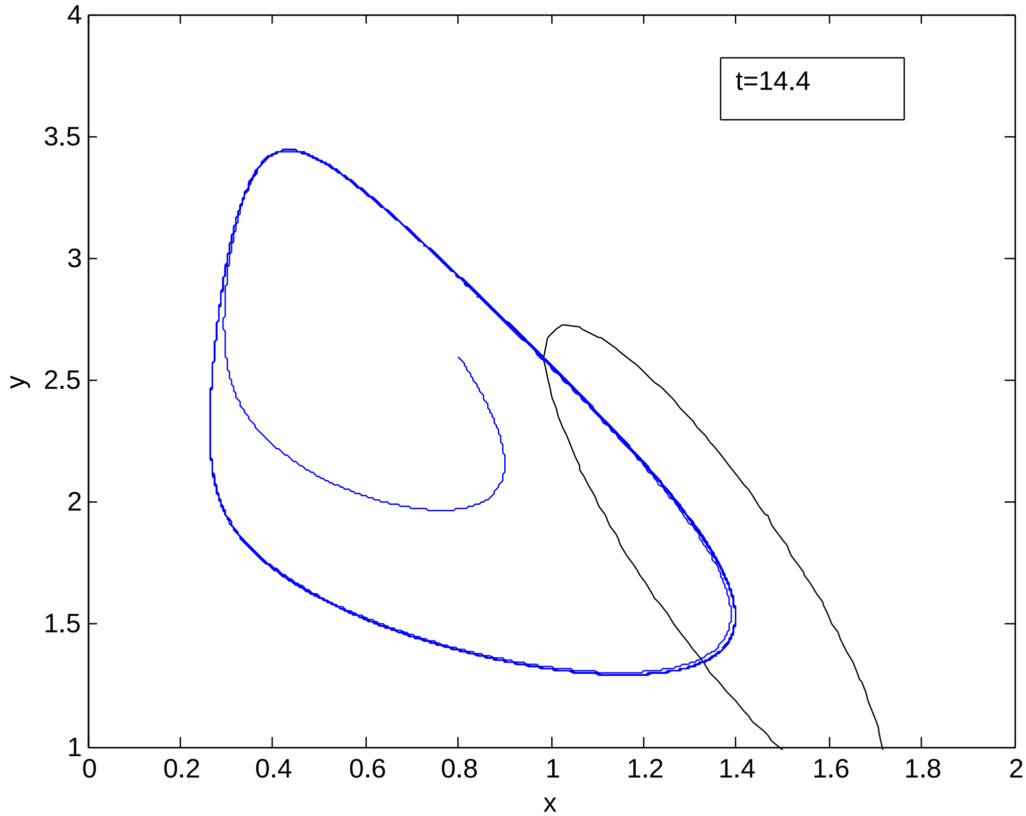
The times are indicated on the graphs.



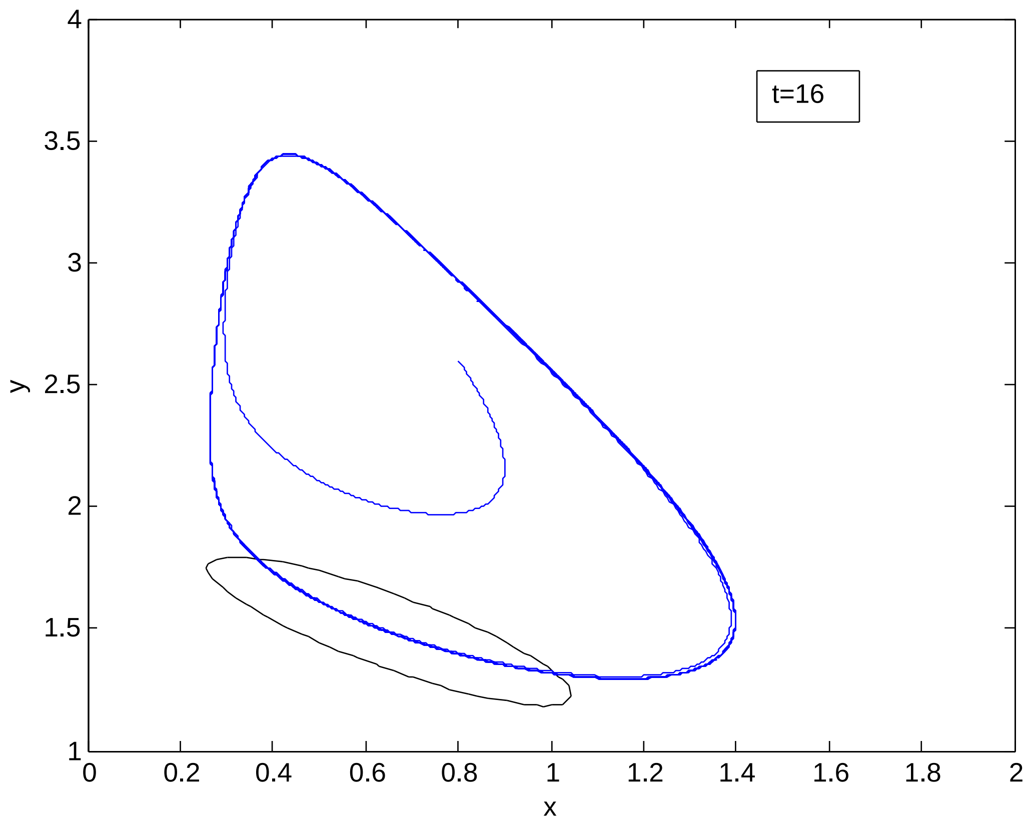
t=4.8
4
3.5
3
2.5
2
1.5
1
y
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
x

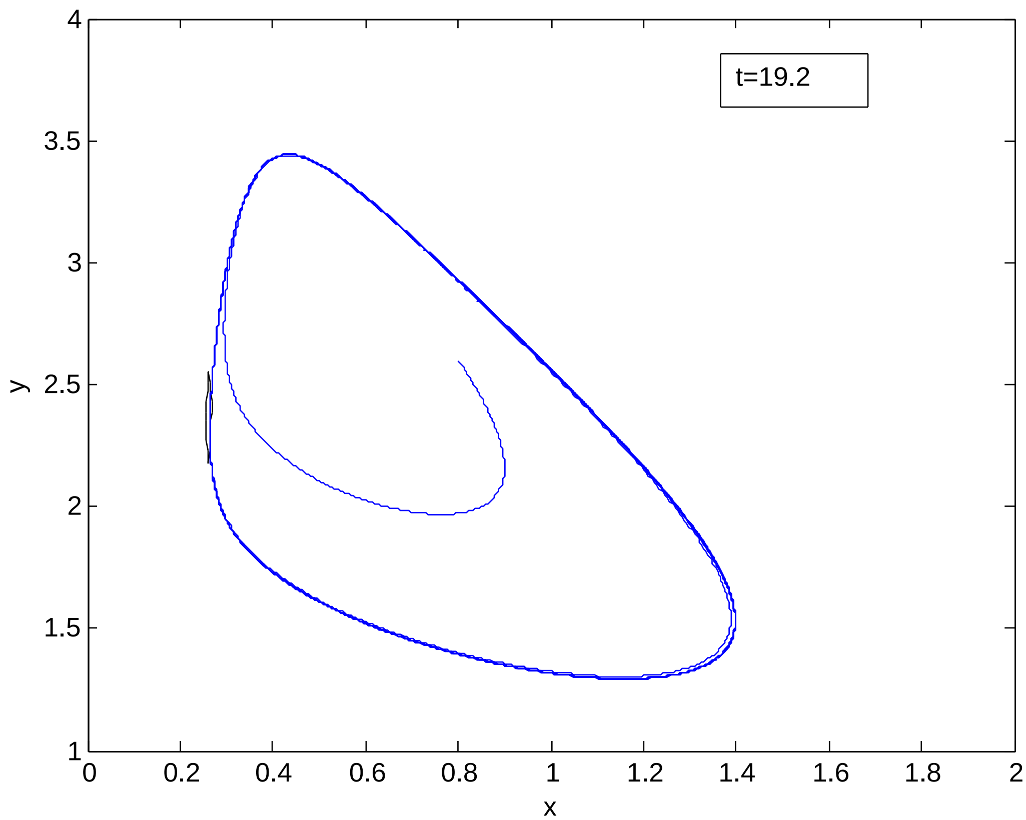


t=11.20
y
x
4
3.5
3
2.5
2
1.5
1
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2



t=14.4
y
x
4
3.5
3
2.5
2
1.5
1
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2


t=16
4
3.5
3
2.5
2
1.5
1
y
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
x



t=19.2
4
3.5
3
2.5
2
1.5
1
y
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
x

Before reaching the limit cycle, the distributions seem to extend perpendicular to the orbit, but it expands along the orbit after reaching the limit cycle. We observe that the distributions indicated by the ellipses narrow down even after deviating widely from the limit cycle orbit at intermediate times.

In [1], the relaxation time to the steady state distribution is also computed, the description of which we leave out in this term-paper.

5 Conclusion

We report one of the main results in [1], that is the computation of the covariance matrix of the Gaussian approximation of a distribution evolving through a chemical Fokker-Planck equation, under some assumptions. We also obtain similar numerical estimates as in the paper for a Brusselator model.

References

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