Self-oscillations in a certain Belousov–Zhabotinsky model

Liudmila Kondratieva[1,*] and *Aleksandr* Romanov[2]

[1]Moscow Aviation Institute (National Research University), 4, Volokolamskoe shosse, Moscow, 125993, Russia
[2]School of Applied Mathematics, HSE University, 34, Tallinskaya st., Moscow, 123458, Russia

Abstract. We consider the dynamic properties of a system of three differential equations known as the oreganator model. This model depends on four external parameters and describes one of the periodic Belousov–Zhabotinsky reactions. We obtain broad conditions for the parameters that ensure the existence of non-stationary steady-state regimes in oregonator model. With classical values of the parameters, the localization of the limit (at a long time) dynamics in the phase space has been improved. In fact, using numerical analysis, we significantly narrow the bounded region of the phase space containing the trajectories of the system. An iterative procedure is proposed for the approximate localization of closed trajectories (cycles) of the system on algebraic surfaces in R^3. A promising problem of theoretical substantiation of the numerical convergence of this procedure is posed.

1 Introduction

The problem of the existence of periodic regimes in problems of mechanics, chemistry, biology, economics, etc. is highly relevant. Let us consider in this connection one of the models of the Belousov–Zhabotinsky reaction [1–4], known in the literature as the "oregonator". This model corresponds to the system of ODEs

$$\begin{aligned} \frac{dx}{dt} &= s(y - xy + x - qx^2), \\ \frac{dy}{dt} &= s^{-1}(-y - xy + sfz), \\ \frac{dz}{dt} &= wx - wz \end{aligned} \tag{1}$$

with positive constants s, w, f, q. According to [1–3], system (1) describe oscillations in a real chemical reaction at $s = 77.27$, $q = 8.375 \cdot 10^{-6}$ and appropriate constraints on w, f. Various aspects of this model were considered in [5–7]. An interesting work [8] demonstrates the possibility of applying the oregonator model to problems not related to chemistry.

For $q < 1$, system (1) admits [2] a bounded positively invariant domain $D \subset R^3_+$ containing a single stationary point $\bar{p}$. We find broad (different from the well-known [2]) conditions on the parameters s, w, q and $f \leq 1$, under which a given point is unstable. Under these conditions, almost all solutions of system (1) go to nonstationary regimes when $t \to +\infty$. An

*e-mail: liudmila.kondratieva@inbox.ru

unstable invariant manifold $M^u(\bar{p})$ of a unstable point $\bar{p}$ is two-dimensional, and a stable manifold $M^s(\bar{p})$ is one-dimensional. System (1) generates in D a smooth phase semiflow $\{\Phi_t\}_{t\geq 0}$ and for each point $p \in D\backslash M^s(\bar{p})$ the limit set $\omega(p)$ does not contain $\bar{p}$, and therefore has a nontrivial geometric structure. Acting as in [2], the existence of a closed trajectory (cycle) $\Gamma \subset D$ of system (1) can be established using the so-called torus principle [9, § 18].

We analyze the instability of the system at $q < 1,\ f \leq 1$. It is convenient to consider these two parameters as control ones. In principle, reducing (1) to a two-dimensional manifold $M^u(\bar{p})$ and applying the Poincaré–Bendixson theory can provide asymptotic orbital stability of the cycle Γ. A similar technique was applied in [10–12] to the problem of satellite motion, as well as to one biochemical model of dynamics in cellular processes.

In the case $s = 77.27$, $q = 8.375\cdot 10^{-6}$, $w = 41.86$, $f = 1$, we significantly narrow the positively invariant domain of system (1). The new domain still contains a stationary point $\bar{p}$and an unstable manifold $M^u(\bar{p})$. Thus, we improve the localization of the limit (as $t \to +\infty$) phase dynamics of system (1). Numerical analysis using the Maple package demonstrates that the system enters the self-oscillation mode ($M^u(\bar{p})$ contains a cycle Γ) and this cycle "well placed" on some algebraic surface of the fifth order in R^3.

2 Stationary point instability

As seen from (1), the positive octant R^3_+ is positively invariant. It is easy to show (see [2]) that the parallelepiped

$$D = D(f, q)\colon \left\{1 < x < \frac{1}{q},\ \frac{fq}{1+q} < y < \frac{f}{2q},\ 1 < z < \frac{1}{q}\right\}$$

has the same property.

We find the only stationary point $\bar{p} = (\bar{x}, \bar{y}, \bar{z}) \in R^3_+$, $\bar{p} = \bar{p}(f, q)$, of system (1) from the relations

$$y - xy + x - qx^2 = 0, \quad -y - xy + fz = 0, \quad x - z = 0.$$

We have $q\bar{x}^2 + (q + f - 1)\bar{x} - 1 - f = 0$ and

$$\bar{x} = \bar{z} = \frac{1}{2q}\left(\sqrt{(q + f - 1)^2 + 4q(1 + f)} - (q + f - 1)\right),$$

$$\bar{y} = \frac{f\bar{x}}{1 + \bar{x}} = \frac{1}{2}(1 + f - q\bar{x}), \quad \bar{y} < f.$$

The convex bounded positively invariant domain $D \subset R^3_+$ contains at least one stationary point; therefore, $\bar{p} \in D$. The identity $q\bar{x}(1 + \bar{x}) = 2f + (1 - f)(1 + \bar{x})$ implies the estimates:

$$1 + \bar{x} > \sqrt{\frac{2}{q}}, \quad f = 1; \quad 1 + \bar{x} > \frac{1 - f}{q}, \quad f < 1. \tag{2}$$

Consider the Jacobi matrix

$$J'(\bar{p}) = \begin{pmatrix} s(1 - \bar{y} - 2q\bar{x}) & s(1 - \bar{x}) & 0 \\ -s^{-1}\bar{y} & -s^{-1}(1 + \bar{x}) & s^{-1}f \\ w & 0 & -w \end{pmatrix}$$

and its characteristic polynomial $\det(\lambda E - J'(\bar{p})) = \lambda^3 + \alpha\lambda^2 + \beta\lambda + \gamma$, where E is the unit matrix. Calculate:

$$\gamma = -\det J'(\bar{p}) = w(1 + q\bar{x} + 2\bar{y}) > 0$$

at all $q, f, s, w > 0$. Further,

$$\alpha = \frac{3}{2} sq\bar{x} + s^{-1}(1+\bar{x}) + w + \frac{s}{2}(f-1) > 0$$

at $f = 1$. The estimate $\alpha > 0$ is also valid [2, p. 681] for all $0 < f < 1$.

Since $\gamma > 0$, $\alpha > 0$, then according to the Routh–Hurwitz criterion the instability of a stationary point provides a condition $\gamma > \alpha\beta$, which therefore the condition $\beta \le 0$. The only stationary point $\bar{p} \in D$ is non-degenerate, its Poincare index is 1, and the Jacobi matrix $J'(\bar{p})$ with all permissible parameter values has in $\mathrm{Re}\,\lambda > 0$ either two (with multiplicity) of eigenvalues or none. So, with $\beta \le 0$ an unstable manifold of the point $\bar{p}$ two-dimensional.

The value β is the sum of the main 2×2 minors of the matrix $J'(\bar{p})$ and

$$\beta = \bar{y}(2+sw) + (2q\bar{x}-1)(1+\bar{x}+sw) + s^{-1}w(1+\bar{x}).$$

Put $1 + \bar{x} = A$, then $\bar{y} = \dfrac{f\bar{x}}{1+\bar{x}} = f - \dfrac{f}{A}$ and $2q\bar{x} - 1 = 1 + 2f - 4\bar{y} = 1 - 2f + \dfrac{4f}{A}$. As $\bar{y} < f$,

$$\beta < 6f + sw(1-f) + (1-2f)A + \frac{3swf}{A} + \frac{w}{s}A.$$

In particular, the condition $\beta \le 0$ is fair if

$$\left(2f - 1 - \frac{w}{s}\right)A^2 - 2\theta A - 3swf > 0$$

at $\theta = 3f + \dfrac{sw}{2}(1-f)$. Taking into account estimates (2), we obtain sufficient conditions for the instability of the stationary point $\bar{p} \in D$ as

$$f = 1, \quad w < s, \quad \sqrt{\frac{2}{q}} > \frac{3 + \sqrt{9 + 3w(s-w)}}{1 - s^{-1}w} = \mu_0, \tag{3.1}$$

$$\frac{1}{2}\left(1 + \frac{w}{s}\right) < f < 1, \quad \frac{1-f}{q} > \frac{\theta + \sqrt{\theta^2 + 3sw(2f - 1 - s^{-1}w)}}{2f - 1 - s^{-1}w} = \mu_1. \tag{3.2}$$

It is convenient to record estimates (3.1), (3.2) in the form of $q < \dfrac{2}{\mu_0^2}$ at $f = 1$ and $q < \dfrac{1-f}{\mu_1}$ at $f < 1$. Under conditions (3), the stationary point $\bar{p}$ is a saddle with a one-dimensional stable and two-dimensional unstable manifold.

3 Localization and visualization of limit dynamics

After normalization of variables $x \to s^{-1}x$, $y \to sy$, $z \to s^{-1}z$, the system (1) acquires the form

$$\begin{aligned} \frac{dx}{dt} &= s^{-1}y - xy + sx - s^2qx^2, \\ \frac{dy}{dt} &= -s^{-1}y - xy + sfz, \\ \frac{dz}{dt} &= wx - wz \end{aligned} \tag{4}$$

with a positively invariant domain

$$\Omega = \Omega(f, q, s)\colon \left\{ s^{-1} < x < \frac{s^{-1}}{q},\ \frac{sfq}{1+q} < y < \frac{sf}{2q},\ s^{-1} < z < \frac{s^{-1}}{q} \right\}.$$

We preserve the designations of the coordinates x, y, z and the stationary point $\bar{p}$ when moving from the system (1) to the system (4). We assume $f = 1$ and try to reduce the domain Ω. Let us show that on the surface

$$S_1: \quad z = g_1(x, y) \doteq \frac{s}{4} + \frac{1}{s}(x + y)^2$$

the vector field $\bar{F}$ of system (4) is directed to the domain. Indeed, if we take the field of normals on S_1 in the form $\bar{n} = (2s^{-1}(x + y), 2s^{-1}(x + y), -1)$, then the scalar product

$$\bar{F} \cdot \bar{n}\Big|_{S_1} = 2s^{-1}(x + y)(s^{-1}y - xy + sx + s^2qx) + 2s^{-1}(x + y)(-s^{-1}y - xy + sz) + w(z - x) =$$
$$= 2(x + y)(z - 2s^{-1}xy) + 2x(x + y)(1 - sqx) + w(z - x) > 0$$

since all expressions in parentheses are positive. Since $s^{-1} < s/4 + s^{-2}$ for all $s > 0$, the lower edge of the parallelepiped Ω is contained in the domain Ω_1.

We further assume that $w = 41.86$ and (as in [1–3]) $s = 77.27$, $q = 8.375 \cdot 10^{-6}$. Consider the surface

$$S_2: \quad z = g_2(x, y) \doteq -s^{-1}(x + 48)(y - 263) + 333$$

and take the field of normals on on the surface in the form $\bar{n} = (s^{-1}(y - 263), s^{-1}(x + 48), 1)$. Numerical analysis shows that on the surface S_2 the vector field of system (4) is directed to the domain $\Omega_2 : z < g_2(x, y)$, i.e. $\bar{F} \cdot \bar{n}|_{S_2} < 0$.

The plane $z = z_0$, $z_0 > s/4$, intersects the surfaces S_1 and S_2 in the straight line $x + y = \sqrt{s(z_0 - s/4)}$ and in the hyperbola $(x + 48)(y - 263) = s(333 - z_0)$, respectively. We accept $z_0 = 251$, then (also, numerically) it can be shown that on a part of the plane $z = z_0$, $(x, y, z_0) \in \Omega_1 \cap \Omega_2$, the vector field $\bar{F}$ is directed to the domain $\Omega_3 : z < z_0$, i.e. $\bar{F} \cdot \bar{n} = w(x - z) < 0$ for the normal $\bar{n} = (0, 0, 1)$.

As we can see, the domain

$$\Omega_0 = \Omega \cap \Omega_1 \cap \Omega_2 \cap \Omega_3$$

is positively invariant for system (4) and homeomorphic to a ball. The domain Ω_0 contains a single stationary point $\bar{p}$, its two-dimensional unstable manifold $M^u(\bar{p})$, and a stable (as numerical analysis shows) limit cycle $\Gamma \subset M^u(\bar{p})$. At the same time, domain Ω_0 is significantly less than Ω. Let the y^+, y_0 and z^+, z_0 be the maximum coordinate values of y and z in the domains Ω and Ω_0, respectively. Then we have $y^+ \approx 4.6 \cdot 10^6$, $y_0 \approx 789.9$ and $z^+ \approx 1543.3$, $z_0 = 251$.

Let us pose the problem of approximate localization of the cycle Γ on some algebraic set (algebraic surface). For an arbitrary polynomial $Q \neq \text{const}$ in R^3, we put

$$S(Q) = \{(x, y, z) \in R^3 : Q(x, y, z) = 0\}.$$

If $S(Q) \supset \Gamma$ and gradient $\nabla Q \neq 0$ on Γ then the vector $\bar{F}$ lies in the tangent plane of the surface $S(Q)$ for all points of the cycle, i.e. $\nabla Q \cdot \bar{F} = 0$ on Γ. Let us introduce a linear differential operator $L: Q \to \nabla Q \cdot \bar{F}$. We have

$$(LQ)(x(t), y(t), z(t)) = \frac{d}{dt} Q(x(t), y(t), z(t))$$

for an arbitrary solution $(x(t), y(t), z(t))$ of system (4) in Ω_0, i.e. LQ is the derivative of the function $Q(x, y, z)$ by virtue of (4). For the polynomial Q_0, we set $Q_1 = LQ_0$ and, in general, $Q_k = LQ_{k-1}$ for $k \geq 1$. At each iteration, the degrees of the polynomials are increased by one. The sets $S(Q_k)$, $k \geq 1$, contain stationary points of system (4) in R^3.

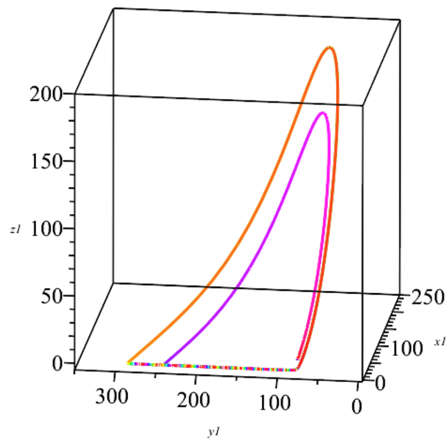

Figure 1. Limit cycle image

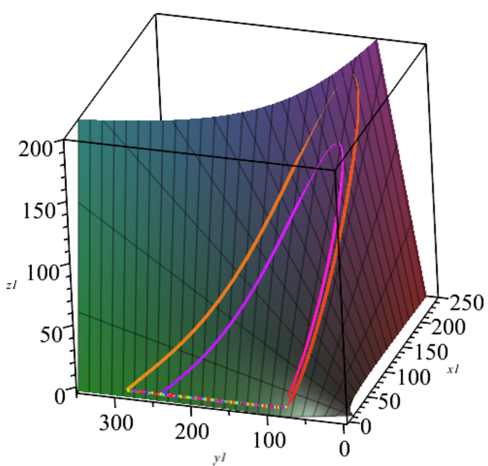

Figure 2. The hyperbolic paraboloid $S(Q_0)$ and limit cycle Γ

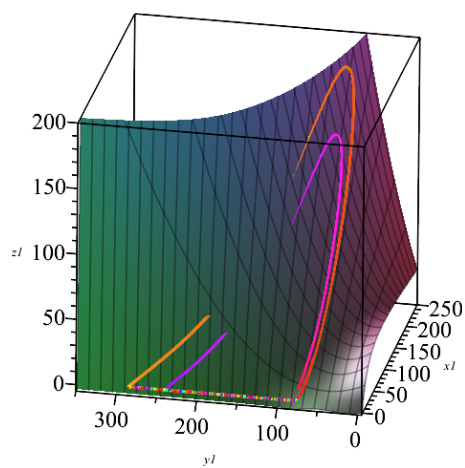

Figure 3. The 3rd order surface $S(Q_1)$ and limit cycle Γ

Let us try to choose an initial approximation Q_0 such that, with increasing k, the distances (deviations) from the cycle Γ to $S(Q_k)$ decrease. Figure 1 shows the limit cycle Γ and the trajectory approaching it.

Using computer visualization, we select Q_0 such that the surface $S(Q_0)$ repeats the outline Γ. For example, put $Q_0(x, y, z) = xy - sz$, i.e. $S_0 = S(Q_0)$ is the hyperbolic paraboloid $z = s^{-1}xy$. At the first iteration, we find the polynomial $Q_1 = LQ_0 = \nabla Q_0 \cdot \bar{F}$ of degree 3 and the corresponding surface

$$S(Q_1)\colon z(w + x)s^2 = syx^2(qs^2 + 1) + x(ws^2 - ys^2 + sy^2 + y) - y^2.$$

Figures 2 and 3 show the localization of the limit cycle Γ near surfaces $S(Q_0)$ and $S(Q_1)$, respectively.

At the second iteration, we find the polynomial $Q_2 = LQ_1 = \nabla Q_1 \cdot \bar{F}$ of degree 4 and the surface $S(Q_2)$. At the third iteration, we obtain the polynomial $Q_3 = LQ_2 = \nabla Q_2 \cdot \bar{F}$ of

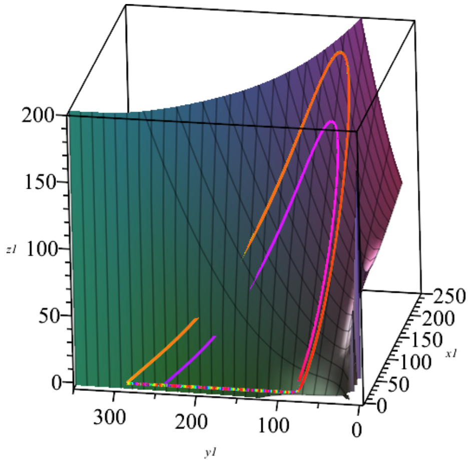

Figure 4. The 4th order surface $S(Q_2)$ and limit cycle Γ

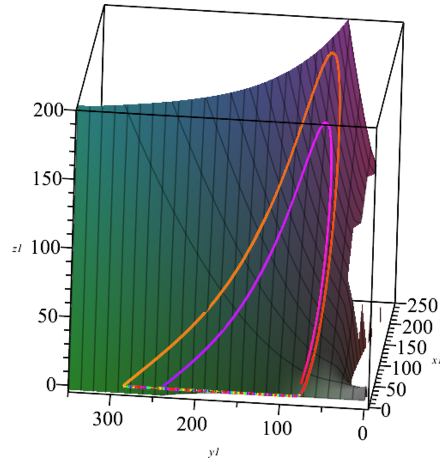

Figure 5. The 5th order surface $S(Q_3)$ and limit cycle Γ

degree 5 and the surface $S(Q_3)$. Figures 4 and 5 show the localization of the limit cycle Γ near surfaces $S(Q_2)$ and $S(Q_3)$, respectively.

One can see (visually) that the deviations $\rho_0, \rho_1, \rho_2, \rho_3$ are sequentially decrease and the cycle Γ "well placed" on the surface of the fifth order $S(Q_3) \subset R^3$. It is interesting to find out under what conditions for the considered iterative procedure the deviations $\rho_k \to 0$ at $k \to \infty$. This task looks very promising and may become the subject of further research.

4 Conclusion

The area of variation of parameters $s, w, f, q > 0$ is described, at which non-stationary limiting modes arise in system (1) and in its equivalent system (4). In the case $s = 77.27$, $q = 8.375 \cdot 10^{-6}$, $w = 41.86$, $f = 1$, the known positively invariant domain is substantially reduced to one containing a single stationary point and a stable limit cycle Γ. An iterative procedure for the approximate localization of the cycle Γ on a suitable two-dimensional algebraic surface in three-dimensional phase space is described.

References

[1] R.J. Field, R.M. Noyes, J. Chem. Phys., **60**, 1877 (1974)
[2] S.P. Hastings, J.D. Murray, SIAM J. Appl. Math., **28**, 678 (1975)
[3] G. Nicolis, I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley, New York, 1977)
[4] I.R. Epstein, J.A. Pojman, *An introduction to nonlinear chemical dynamics* (Oxford University Press, New York, 1998)
[5] A.K. Dutt, AIP Advances, **1**, 042147 (2011)
[6] R.D. Ikramov, S.A. Mustafina, International Research Journal (Phys. and Math.), **9-2**, 124 (2016)
[7] A. Adamatzky, Int. J. Bifurc. Chaos, **27(3)**, 1750041 (2017)
[8] A. Adamatzky, N. Phillips, R. Weerasekera, M.A. Tsompanas, G.Ch. Sirakoulis, arXiv:1803.01632 (2018)

[9] V.A. Pliss, *Nonlocal Problems of the Theory of Oscillations* (Academic Press, New York, 1966)

[10] L.A. Kondratieva, Aerospace MAI Journal, **19**, 75 (2012)

[11] L. Kondratieva, *Computational model for satellite periodic motion*, AIP Conf. Proc., **2181**, 020002 (2019)

[12] L.A. Kondratieva, A.V. Romanov, Elect. J. Qual. Th. Differ. Eq., **96**, 1 (2019)