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No limit cycle in two species second order kinetics

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Abstract

We give a new and direct proof of the nonexistence of limit cycle in a bimolecular system and the characterization of the unique bimolecular oscillator. The proof is an application of classification theorems on vector fields with homogeneous second degree polynomial perturbations. © 2003 Éditions scientifiques et médicales Elsevier SAS. All rights reserved.

Résumé

On donne une nouvelle démonstration de la non existence de cycle limite dans un système bimoleculaire et la caractérisation de l'unique oscillateur bimoléculaire. La preuve est une application directe des théorèmes de classification des champs de vecteurs polynomiaux avec une perturbation homogène quadratique.

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1. Introduction

One of the fundamental statements on the oscillatory behavior of two species second order reactions is the nonexistence of limit cycles. (As we only consider mass action type

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kinetics the expressions *second order* and *bimolecular* are thought to be synonymous in the present paper.) This statement has been formulated and argued for first by Hanusse [7]. Next Tyson and Light [15] gave a proof neglecting some less probable and less important cases. Finally, Póta [11] gave a full proof of the statement using a theorem of Dulac (different from the one we are going to use here).

Another, related problem, of which we shall not present the whole history is to find the smallest or simplest chemical oscillator. One possible approach to this problem was shown by Tóth and Hárs [14] who has shown that it is again the Lotka–Volterra model which gives the answer: if the linear part is given then no other model than this can have as few species and reaction steps as this. Here, we prove indeed that the smallest chemical oscillator is the unique one.

The structure of our paper is as follows. First we define the class of polynomial differential equations for which there exist classification theorems in the most succint form: in terms of complex variables. Then we summarize the theorems by Dulac [4], Bautin [1,18], Loud [9] and Chicone [2]. Next we rewrite the equation in real form, and consider it as one obtained by a translation of the stationary point to the origin. Thus, in order to return to the original kinetic equation we have to translate the equation back to the stationary point generally taken to be an arbitrary point in the first orthant. Then we have the most general form of polynomial equations which can be classified. At this point we present the system of inequalities expressing the fact that the polynomial system is kinetic (i.e. it contains no negative cross effect). Then we check whether individual conditions of the classification theorems can be fulfilled or not. We shall almost always find a negative answer, except in the single case of the Lotka–Volterra reaction corresponding to the result we wanted to reproduce.

In the last section we shall formulate a few problems (both mathematical and kinetic) which hopefully can be attacked by the present methods.

2. The class of equations and some mathematical preliminaries

Below we shall use several times classification theorems related to the class of polynomial differential equations which can be written in terms of complex variables as follows

$$\dot{z} = (i+\lambda)z + a_{20}z^2 + a_{11}z\bar{z} + a_{02}\bar{z}^2,\tag{1}$$

where λ is a real parameter (in some it equals to zero), and the coefficients a_{20} , a_{11} , a_{02} are arbitrary complex numbers. The unknown complex valued function z is in general defined on some finite or infinite interval of the real line.

Together with the conditions we also give their names in parentheses which may also be a formula, as e.g. in the second case of Loud's theorem below.

Definition 2.1. The origin is said to be a *center* to Eq. (1) if all the orbits in a small neigbourhood of it are closed curves surrounding the origin.

Theorem 2.2 [4,18]. The origin is a center for the polynomial differential equation (1) iff $\lambda = 0$ and at least one of the four conditions is satisfied:

1. (H.C.) $a_{20} = -\frac{1}{2}\bar{a}_{11}$ (Hamiltonian center); 2. (D.C.) $a_{20} = 2\bar{a}_{11}$, $|a_{11}|^2 = |a_{02}|^2$ (Darboux center); 3. (L.V.) $a_{11} = 0$ (Lotka–Volterra); 4. (S.C.) $\Im(a_{20}a_{11}) = \Im(a_{20}^3a_{02}) = 0$ (symmetric center).

Definition 2.3. A center is said to be *isochronous* if all the periodic orbits in a neighbourhood of the origin have the same period.

Theorem 2.4 [9]. The origin is an isochronous center for the polynomial differential equation (1) iff $\lambda = 0$ and at least one of the five conditions is satisfied:

1. (h.I.C) $a_{11} = a_{02} = 0$ (holomorphic center); 2. $(\dot{\theta} = 1) a_{20} = \bar{a}_{11}, a_{02} = 0;$ 3. (I.C.1) $a_{20} = \frac{5}{2}\bar{a}_{11}, |a_{11}|^2 = \frac{4}{9}|a_{02}|^2;$ 4. (I.C.2) $a_{20} = \frac{7}{6}\bar{a}_{11}, |a_{11}|^2 = 4|a_{02}|^2;$ 5. (H.I.C.) $a_{20} = a_{11} = a_{02} = 0$ (Hamiltonian isochronous center).

The conditions of this theorem imply those of the Dulac theorem (as they should) as follows:

 $1. \Rightarrow 3.$ $1. \Rightarrow 4.$ $2. \Rightarrow 4.$ $3. \Rightarrow 4.$ $4. \Rightarrow 4.$ $5. \Rightarrow 1., 2., 3., 4.$

of which only the fourth and fifth implications need some calculation.

It is an important and general problem to find the stratification of the center variety by isochronous centers; e.g. in the Hamiltonian case [13] it has been shown that (H.C.) implies (H.I.C.) for homogeneous polynomial perturbations of any degree in \mathbb{R}^2 .

Definition 2.5. The trivial equilibrium point (the origin) of system (1) is said to have a *cyclicity k* with respect to the space of all quadratic systems if any quadratic perturbation of system (1) has at most k limit cycles in a neighbourhood of the origin, and k is the smallest number with this property.

Theorem 2.6 [1]. The cyclicity of the trivial equilibrium point z = 0 of system (1) with respect to the space of all quadratic systems is less than or equal to 3.

We recall the structure of the first return map given by Bautin (see for example [18]):

Theorem 2.7. If L(r) denotes the first intersection of the positive semitrajectory with the x-axis starting from $r \ge 0$ (i.e. L is the first return map), and if it is of the form $L(r) = r + \sum_{k\ge 1} L_{2k+1}r^{2k+1}$, then its coefficients are generated by the following three polynomials (called focal values):

$$v_{3} = -2\pi \Im(a_{20}a_{11}), \qquad v_{5} = (-2\pi/3)\Im((2a_{20} + \bar{a}_{11})(a_{20} - 2\bar{a}_{11})\bar{a}_{11}a_{02}),$$

$$v_{7} = (-5\pi/4)\Im((|a_{11}|^{2} - |a_{02}|^{2})(2a_{20} + \bar{a}_{11})\bar{a}_{11}^{2}a_{02}).$$

A consequence of this theorem formulated by Żołądek follows.

Theorem 2.8. The cyclicity of the trivial equilibrium point is

- 0, if $\lambda \neq 0$;
- 1, *if* $\lambda = 0$, $\Im(a_{20}a_{11}) \neq 0$;
- 2, if $\lambda = \Im(a_{20}a_{11}) = 0$, $a_{20} 2\bar{a}_{11} \neq 0$;
- 3, if $\lambda = a_{20} 2\bar{a}_{11} = 0$.

We recall the cyclicity of the equilibrium point in the isochronous case:

Theorem 2.9 [2]. Let $a_{20} \neq 0$. Then, the number of limit cycles of Eq. (1) which can bifurcat *e* from an isochronous center with respect to the space of all quadratic systems is at most

- 0, if $\lambda \neq 0$;
- 1, if $\lambda = 0$, and Eq. (1) is in the class (h.I.C);
- 2 if $\lambda = 0$, and Eq. (1) is in one of the classes ($\dot{\theta} = 1$), (I.C.1), (I.C.2).

Remark. The number of limit cycles which can bifurcate from a Hamiltonian isochronous center (H.I.C.) is 3 as a corollary of the Bautin's theorem.

3. The system of two real equations translated to the stationary point

We should like to consider Eq. (1) as if it were obtained by a translation of the stationary point to the origin and now we make the inverse transformation. Before that let us rewrite the equation in the form of two real variables as this form will have an immediate chemical meaning.

If we introduce the following notations

$$z =: x + iy, \qquad a_{20} =: \alpha_{20} + i\beta_{20}, a_{11} =: \alpha_{11} + i\beta_{11}, \qquad a_{02} =: \alpha_{02} + i\beta_{02},$$
(2)

and assume that

$$X := x + \xi, \quad Y := y + \eta, \quad \text{with } \xi, \eta > 0,$$
 (3)

then Eq. (1) will have the following form

$$\begin{split} \dot{X} &= \eta - \lambda \xi + \xi^2 (\alpha_{20} + \alpha_{11} + \alpha_{02}) - 2\xi \eta (\beta_{20} - \beta_{02}) + \eta^2 (-\alpha_{20} + \alpha_{11} - \alpha_{02}) \\ &+ X (\lambda - 2\xi (\alpha_{20} + \alpha_{11} + \alpha_{02}) + 2\eta (\beta_{20} - \beta_{02})) \\ &+ Y (-1 + 2\xi (\beta_{20} - \beta_{02}) - 2\eta (-\alpha_{20} + \alpha_{11} - \alpha_{02})) \\ &+ X^2 (\alpha_{20} + \alpha_{11} + \alpha_{02}) - 2XY (\beta_{20} - \beta_{02}) + Y^2 (-\alpha_{20} + \alpha_{11} - \alpha_{02}), \quad (4) \\ \dot{Y} &= -\xi - \lambda \eta + \xi^2 (\beta_{20} + \beta_{11} + \beta_{02}) + 2\xi \eta (\alpha_{20} - \alpha_{02}) + \eta^2 (-\beta_{20} + \beta_{11} - \beta_{02}) \\ &+ X (1 - 2\xi (\beta_{20} + \beta_{11} + \beta_{02}) - 2\eta (\alpha_{20} - \alpha_{02})) \\ &+ Y (\lambda - 2\xi (\alpha_{20} - \alpha_{02}) - 2\eta (-\beta_{20} + \beta_{11} - \beta_{02})) \\ &+ X^2 (\beta_{20} + \beta_{11} + \beta_{02}) + 2XY (\alpha_{20} - \alpha_{02}) + Y^2 (-\beta_{20} + \beta_{11} - \beta_{02}). \quad (5) \end{split}$$

The system of equations (4)–(5) is a kinetic equation iff no negative cross effects (see e.g. [14] and the references therein) are present, i.e. the following inequalities hold:

$$\eta - \lambda \xi + \xi^2 (\alpha_{20} + \alpha_{11} + \alpha_{02}) - 2\xi \eta (\beta_{20} - \beta_{02}) - \eta^2 (\alpha_{20} - \alpha_{11} + \alpha_{02}) \ge 0, \quad (6)$$

$$-1 + 2\xi(\beta_{20} - \beta_{02}) + 2\eta(\alpha_{20} - \alpha_{11} + \alpha_{02}) \ge 0, \tag{7}$$

$$-\alpha_{20} + \alpha_{11} - \alpha_{02} \geqslant 0,\tag{8}$$

$$-\xi - \lambda \eta + \xi^2 (\beta_{20} + \beta_{11} + \beta_{02}) + 2\xi \eta (\alpha_{20} - \alpha_{02}) - \eta^2 (\beta_{20} - \beta_{11} + \beta_{02}) \ge 0,$$
(9)

$$1 - 2\xi(\beta_{20} + \beta_{11} + \beta_{02}) - 2\eta(\alpha_{20} - \alpha_{02}) \ge 0, \tag{10}$$

$$\beta_{20} + \beta_{11} + \beta_{02} \ge 0. \tag{11}$$

In the next section, we show that the unique kinetic bimolecular system with periodic orbits is the Lotka–Volterra system.

4. On the existence of a center

Theorem 4.1. *The system of Eqs.* (4)–(5) *can only have a center if condition* (L.V.) *of Dulac's theorem holds. It cannot have an isochronous center.*

In order to have either a center or an isochronous center, $\lambda = 0$ should hold.

We consider the case of Theorems 2.2, and 2.4 with conditions $a_{20} = \varepsilon \bar{a}_{11}$, $\varepsilon \in \{1, 2, \frac{5}{2}, \frac{7}{6}\}$, which correspond to the cases $(\dot{\theta} = 1)$, (D.C.), (I.C.1), (I.C.2). Since, $\alpha_{20} = \varepsilon \alpha_{11}$ and $\beta_{20} = -\varepsilon \beta_{11}$, we obtain from Eqs. (6)–(11)

$$\eta + \xi^{2} ((\varepsilon + 1)\alpha_{11} + \alpha_{02}) - 2\xi \eta (-\varepsilon \beta_{11} - \beta_{02}) + \eta^{2} ((-\varepsilon + 1)\alpha_{11} - \alpha_{02}) \ge 0,$$
(12)

$$-1 + 2\xi(-\varepsilon\beta_{11} - \beta_{02}) - 2\eta((-\varepsilon + 1)\alpha_{11} - \alpha_{02}) \ge 0,$$
(13)

$$(-\varepsilon+1)\alpha_{11} - \alpha_{02} \ge 0, \tag{14}$$

$$-\xi + \xi^{2} ((-\varepsilon + 1)\beta_{11} + \beta_{02}) + 2\xi \eta (\varepsilon \alpha_{11} - \alpha_{02}) + \eta^{2} ((\varepsilon + 1)\beta_{11} - \beta_{02}) \ge 0,$$
(15)

$$1 - 2\xi \left((-\varepsilon + 1)\beta_{11} + \beta_{02} \right) - 2\eta (\varepsilon \alpha_{11} - \alpha_{02}) \ge 0, \tag{16}$$

$$(-\varepsilon+1)\beta_{11}+\beta_{02} \ge 0. \tag{17}$$

Eqs. (13) and (14) imply

$$-\varepsilon\beta_{11} - \beta_{02} \ge 0,\tag{18}$$

Eq. (15) + ξ Eq. (16) is equivalent to $-\xi^2((-\varepsilon+1)\beta_{11}+\beta_{02})+\eta^2((\varepsilon+1)\beta_{11}-\beta_{02}) \ge 0$, and by Eq. (17), we obtain

$$(\varepsilon+1)\beta_{11} - \beta_{02} \ge 0,\tag{19}$$

Eq. (12) + η Eq. (13) is equivalent to $-\eta^2((-\varepsilon+1)\alpha_{11}-\alpha_{02})+\xi^2((\varepsilon+1)\alpha_{11}+\alpha_{02}) \ge 0$, and by Eq. (14), we obtain

$$(\varepsilon + 1)\alpha_{11} + \alpha_{02} \ge 0. \tag{20}$$

Eq. (14) + Eq. (20) is equivalent to $\alpha_{11} \ge 0$, Eq. (17) + Eq. (19) is equivalent to $\beta_{11} \ge 0$, Eq. (17) + Eq. (18) is equivalent to $(-2\varepsilon + 1)\beta_{11} > 0$. Because for all the values of ε we have $(-2\varepsilon + 1) < 0$, but we have just learned that $\beta_{11} \ge 0$, we have a contradiction.

(H.I.C.) case: Hamiltonian isochronous center. $\alpha_{20} = \alpha_{11} = \alpha_{02} = 0$, $\beta_{20} = \beta_{11} = \beta_{02} = 0$. It is impossible by Eq. (9) as it would mean $\xi \leq 0$.

(h.I.C.) case: holomorphic isochronous center. $\alpha_{11} = \beta_{11} = \alpha_{02} = \beta_{02} = 0$.

By the transformation $z \mapsto \xi . z, \xi \in \mathbb{C}^* := \mathbb{C} \setminus \{0\}$, we can choose $\beta_{20} = 0$, i.e. $a_{20} \in \mathbb{R}$. By Eq. (8) $\alpha_{20} \leq 0$, and Eq. (7) is equivalent to $-1 + 2\eta\alpha_{20} \ge 0$, which is impossible.

(H.C.) case: Hamiltonian center. $\alpha_{20} = -\frac{1}{2}\alpha_{11}, \beta_{20} = \frac{1}{2}\beta_{11}$. Eq. (19) is equivalent to $\frac{1}{2}\beta_{11} - \beta_{02} \ge 0 \Leftrightarrow \beta_{02} \le \frac{1}{2}\beta_{11}$,

Eq. (17) + Eq. (19) is equivalent to $\beta_{11} \ge 0$,

Eq. (17) is equivalent to $\frac{3}{2}\beta_{11} + \beta_{02} \ge 0$, and with Eq. (19) we obtain $\frac{1}{2}\beta_{11} \ge \beta_{02} \ge -\frac{3}{2}\beta_{11}$.

After the action of $z \mapsto \xi . z, \xi \in \mathbb{C}^*$, on the system (1), we can choose $\beta_{11} = 0$. Hence, $\beta_{02} = 0$.

So, Eq. (13) is equivalent to $-1 - 2\eta(\frac{3}{2}\alpha_{11} - \alpha_{02}) \ge 0$, a contradiction to $\frac{3}{2}\alpha_{11} - \alpha_{02} \ge 0$ obtained from Eq. (14).

(S.C.) case: Symmetric center. $\Im(a_{20}a_{11}) = \Im(a_{20}^3a_{02}) = 0.$

By the transformation $z \mapsto \xi . z$, $\xi \in \mathbb{C}^*$, we can choose that $a_{02} = \rho_{02} \in \mathbb{R}^+$. So, the algebraic conditions (S.C.) give $\Im(a_{20}a_{11}) = 0$, and $\Im(a_{20}^3) = 0$. Hence,

 $a_{20} = \rho_{20} \mathrm{e}^{\frac{ki\pi}{3}}, \quad k \in \mathbb{Z}, \ \rho_{20} \in \mathbb{R}^*.$

If k = 1, Eq. (1) is equivalent to

$$\dot{z} = iz + \rho_{20} e^{\frac{i\pi}{3}} z^2 + a_{11} z \bar{z} + \rho_{02} \bar{z}^2.$$

By $\Im(e^{\frac{i\pi}{3}}a_{11}) = 0$ and $a_{11} = \rho_{11}e^{i\theta_{11}}$, we obtain $\theta_{11} + \frac{\pi}{3} = k'\pi$, $k' \in \mathbb{Z}$. For the value

$$k'=1, \qquad \theta_{11}=\frac{2\pi}{3},$$

and Eq. (1) $\Leftrightarrow \dot{z} = iz + \rho_{20}e^{\frac{i\pi}{3}}z^2 + \rho_{11}e^{\frac{i2\pi}{3}}z\overline{z} + \rho_{02}\overline{z}^2$. Now, by the transformation $z \mapsto e^{\frac{-i\pi}{3}}z$, Eq. (1) is equivalent to

$$\dot{z} = iz + \rho_{20} e^{\frac{i\pi}{3}} e^{\frac{-i\pi}{3}} z^2 + \rho_{11} e^{\frac{i2\pi}{3}} e^{\frac{i\pi}{3}} z\bar{z} + \rho_{02} e^{i\pi} \bar{z}^2$$

$$\Leftrightarrow \quad \dot{z} = iz + \rho_{20} z^2 + \rho_{11} z\bar{z} - \rho_{02} \bar{z}^2.$$

This procedure allows us to reduce the algebraic condition to the case $\Im(a_{20}) = \Im(a_{11}) = \Im(a_{02}) = 0$, i.e. we obtain that $\beta_{20} = \beta_{11} = \beta_{02} = 0$ (for example, in the case $a_{20} = \rho_{20}e^{\frac{2i\pi}{3}}$, we use $z \mapsto e^{\frac{\pm i\pi}{3}}z$, if we choose $\theta_{11} = \frac{-2\pi}{3}$, k' = 0. We use the same procedure to reduce, in all cases, the coefficients to real ones).

From inequality (8), $(-\alpha_{20} + \alpha_{11} - \alpha_{02}) \ge 0$, we obtain that the kinetic condition Eq. (7), $-1 - 2\eta(-\alpha_{20} + \alpha_{11} - \alpha_{02}) \ge 0$ is impossible.

(L.V.) case: Lotka–Volterra center. $\alpha_{11} = \beta_{11} = 0$. Now Eq. (6) + η Eq. (7) implies $(\xi^2 + \eta^2)(\alpha_{20} + \alpha_{02}) \ge 0$, but because of Eq. (8)

$$\alpha_{20} + \alpha_{02} = 0 \tag{21}$$

should hold. Similarly, Eq. (9) + ξ Eq. (10) implies $-(\xi^2 + \eta^2)(\beta_{20} + \beta_{02}) \ge 0$, but because of Eq. (11) we have

$$\beta_{20} + \beta_{02} = 0. \tag{22}$$

Eqs. (21) and (22) together imply that the system of Eqs. (4)–(5) now reduces to

$$\dot{X} = (\eta + 4\xi\eta\beta_{02}) - 4X\eta\beta_{02} - Y(1 + 4\xi\beta_{02}) + 4XY\beta_{02},$$
(23)

$$\dot{Y} = -(\xi + 4\xi\eta\alpha_{02}) + 4Y\xi\alpha_{02} + X(1 + 4\eta\alpha_{02}) - 4XY\alpha_{02}.$$
(24)

As the above system should still be a kinetic equation, we have to eliminate negative cross effects, which here means that

$$1 + 4\xi\beta_{02} \ge 0, \qquad -(1 + 4\xi\beta_{02}) \ge 0,$$
 (25)

$$1 + 4\eta \alpha_{02} \ge 0, \qquad -(1 + 4\eta \alpha_{02}) \ge 0,$$
 (26)

should simultaneously hold. Therefore,

$$1 + 4\xi\beta_{02} = 0, \qquad 1 + 4\eta\alpha_{02} = 0 \tag{27}$$

which then give for Eqs. (4)–(5) in that case

$$\dot{X} = X\left(\frac{\eta}{\xi} - Y\right), \qquad \dot{Y} = Y\left(X - \frac{\xi}{\eta}\right),$$
(28)

which is the well known Lotka–Volterra system with periodic orbits around the equilibrium point $(\frac{\xi}{n}, \frac{\eta}{\xi})$.

We now recall the definition of the smallest chemical system [17]:

Definition 4.2. The *smallest bimolecular system* can be characterized by the following four features:

- lowest number of reactants,
- lowest number of quadractic terms,
- minimal number of parameters, i.e. minimal number of reactions,
- minimal number of bimolecular reactions.

Remark 4.3. As the obtained model is the smallest one in the sense of Definition 4.2, we reproduced also the result of the paper [14].

5. On the nonexistence of a limit cycle

We now state the result concerning the nonexistence of a limit cycle in a two species system with second order kinetics [7].

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Theorem 5.1. *The system of Eqs.* (4)–(5) *cannot have a limit cycle.*

For the proof, it is sufficient to check the conditions that ensure the bifurcation of limit cycles from a center, since there is no isochronous center two species system with second order kinetics.

We study the cases where the cyclicity of the equilibrium point is:

3. It is the case if $a_{20} - 2\bar{a}_{11} = 0$. It turned out from the proof of Theorem 4.1, in the Darboux case (D.C.) that it is impossible to obtain a kinetic system which fulfils this algebraic condition.

2. $\Im(a_{20}a_{11}) = 0$, $a_{20} - 2\bar{a}_{11} \neq 0$. If Eq. (1) is defined on the (H.C.) or (S.C.), there is no kinetic system. In the (D.C.) an immediate contradiction is obtained. In the case of (L.V.), all the focal values are equal to 0 (existence of a center by Theorem 2.7), and no limit cycle can appear.

1. $\Im(a_{20}a_{11}) \neq 0$. This condition implies that it is impossible for Eq. (1) to be the case (S.C.) or (L.V.). As in the case (H.C.) we have $a_{20}a_{11} = -\frac{1}{2}|a_{11}|^2$, therefore $a_{20}a_{11}$ is a real number, thus we cannot have $\Im(a_{20}a_{11}) \neq 0$. Similarly, in the (D.C.) case $a_{20}a_{11} = 2|a_{11}|^2$, therefore again the imaginary part of a real number cannot be different from zero.

6. Discussion and perspectives

It has been shown in [8] that a three component system with only bimolecular reactions can show limit-cycle oscillations (see also [10,12]). In [17], the model for the minimal oscillating chemical reaction with Hopf bifurcation is given. In [5] (and in a certain sense also in [12]) the problem of the classification of chemical oscillators is presented. Finding the simplest chemical models with given behavior in an exact or approximate way has always been an interesting question [3,6,14,16]. Our aim is to give a unified approach to these kinds of problems.

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