Loci of limit cycles

Douglas Poland
Department of Chemistry, The Johns Hopkins University, Baltimore, Maryland 21218
(Received 5 August 1993)

A recent method of Delamotte [Phys. Rev. Lett. **70**, 3361 (1993)] for obtaining approximate analytic expressions for the loci of limit cycles in one variable is applied to coupled nonlinear first-order rate equations in several variables, the typical case for most models based on chemical kinetics. The first-order approximation works well near a bifurcation point, with higher-order terms being required the further the system is from the bifurcation point. The method complements linear stability analysis (which gives the limiting frequency of the limit cycle at the bifurcation point) by giving a simple method with which to construct an explicit formula that gives the evolution in space and time of a limit cycle near a bifurcation point.

PACS number(s): 05.45.+b, 82.40.Bj, 82.20.Wt, 02.60.Lj

I. INTRODUCTION

Limit cycles, stable orbits in phase space toward which a system moves spontaneously, are one of the more intriguing features that can arise from nonlinear chemical kinetics. Recently Delamotte [1] proposed a very simple method for determining approximate analytic forms for the loci of limit cycles. For the examples he treated he was able to obtain orders of magnitude improvement in the values of the period and amplitude of limit cycles compared with results obtained from very high-order perturbation theory. Delamotte considered second-order differential equations in a single variable. Here we cast his method in a form applicable to first-order differential equations in any number of variables, the common form for the equations of chemical kinetics.

We treat a system of coupled first-order differential equations that exhibit a limit cycle, for example

$$\frac{dx}{dt}=f(x,y)\ , \quad \frac{dy}{dt}=g(x,y)\ , \tag{1.1}$$

where f and g are in general nonlinear functions of x and y arising from mass action, or other kinetic mechanisms. The number of variables is arbitrary, but we need at least two variables to obtain a limit cycle. We will consider examples in two variables. We take ω as the characteristic frequency of the limit cycle. The essence of Delamotte's method is to write the locus of the limit cycle as a Fourier series

$$x(t)=a_x(0)+\sum_{n=1}^{N}[b_x(n)\sin(n\omega t)+c_x(n)\cos(n\omega t)]\ , \tag{1.2}$$

with a similar expression for $y(t)$. The approximation lies in the fact that we take a finite limit N on the sum; for a first-order approximation we take $N=1$, second order $N=2$, and so on. Using the Euler relations one can rewrite (1.2) in terms of the complex variable

$$\zeta=e^{i\omega t}\ , \tag{1.3}$$

obtaining

$$x(t)=a_x(0)+\sum_{n=1}^{N}\{a_x(n)\zeta^n+[a_x(n)]^*\zeta^{-n}\}\ , \tag{1.4}$$

where

$$\begin{aligned} a_x(n)&=\tfrac{1}{2}[c_x(n)-ib_x(n)]\ ,\\ [a_x(n)]^*&=\tfrac{1}{2}[c_x(n)+ib_x(n)]\ . \end{aligned} \tag{1.5}$$

One then substitutes the form of (1.4) into the kinetic equations, giving

$$\begin{aligned} F(\zeta)&=\frac{dx}{dt}-f(x,y)\\ &=A_x(0)+\sum_{n=1}^{M_x}\{A_x(n)\zeta^n+[A_x(n)]^*\zeta^{-n}\}\ ,\\ G(\zeta)&=\frac{dy}{dt}-g(x,y)\\ &=A_y(0)+\sum_{n=1}^{M_y}\{A_y(n)\zeta^n+[A_y(n)]^*\zeta^{-n}\}\ , \end{aligned} \tag{1.6}$$

where $[A_x(n)]^*$ and $[A_y(n)]^*$ are the complex conjugates of $A_x(n)$ and $A_y(n)$. The limits M_X and M_y (which are not necessarily the same) are generally larger than the limit N in (1.4) and depend on the exact nature of the nonlinear terms in f and g. Now if (1.4) is an exact solution of the locus of the limit cycle then one has

$$F(\zeta)=0\ , \quad G(\zeta)=0\ , \tag{1.7}$$

and the coefficient of each power of ζ in (1.6) will be zero. Delamotte's method consists of the requirement that the coefficients in (1.6) be zero for $n=0$ to N (thus it is an approximation since not all of the coefficients are required to be zero). One has explicitly [we note that $A_x(0)$ and $A_y(0)$ must be real]

$$A_x(0)=0\ ,\quad A_y(0)=0\ ,$$
$$\mathrm{Re}\,A_x(n)=0\ ,\quad \mathrm{Re}\,A_y(n)=0\ ,$$
$$\mathrm{Im}\,A_x(n)=0,\quad \mathrm{Im}\,A_y(n)=0,\quad (n=1\text{ to }N)\ . \tag{1.8}$$

Equations (1.8) are in general a set of coupled nonlinear equations for the coefficients in (1.4), i.e., $a_x(0)$, $b_x(n)$, $c_x(n)$, $a_y(0)$, $b_y(n)$, $c_y(n)$, and ω. For each order of approximation, e.g., $N=1$, 2, and so on, one gets a new set of equations and hence all the coefficients change at each order of approximation (hence it is not like a perturbation scheme where the lower-order terms remain fixed as one increases the number of terms). Delamotte does not prove that a solution of (1.8) for the coefficients in (1.4) in real numbers exists, nor that the solution must improve with increasing N. However, the numerical results he presents are very impressive.

The nature of the hierarchy of approximations is illustrated below (where we drop the subscripts x and y for simplicity). We illustrate the stages of approximation for the case $M=3$, i.e., the powers of ζ range from minus three to plus three. Each level of approximation (zeroth, first order, second order) is obtained by requiring all of the coefficients (real and imaginary parts) inside of the curly brackets to be zero; the coefficients outside of the curly brackets are not required to be zero (which is the approximation involved). The zeroth-order approximation gives the steady-state solution of the differential equations (a point).

$$\begin{aligned}
&[A(3)]^*\zeta^{-3}+[A(2)]^*\zeta^{-2}+[A(1)]^*\zeta^{-1}+\{A(0)\}+A(1)\zeta+A(2)\zeta^2+A(3)\zeta^3\ ,\quad \text{zeroth order}\ ,\\
&[A(3)]^*\zeta^{-3}+[A(2)]^*\zeta^{-2}+\{[A(1)]^*\zeta^{-1}+A(0)+A(1)\zeta\}+A(2)\zeta^2+A(3)\zeta^3\ ,\quad \text{first order}\ ,\\
&[A(3)]^*\zeta^{-3}+\{[A(2)]^*\zeta^{-2}+[A(1)]^*\zeta^{-1}+A(0)+A(1)\zeta+A(2)\zeta^2\}+A(3)\zeta^3\ ,\quad \text{second order}\ .
\end{aligned} \tag{1.9}$$

Delamotte explicitly treats second-order differential equations in a single variable

$$f\left[x,\frac{dx}{dt},\frac{d^2x}{dt^2}\right]=0\ . \tag{1.10}$$

He notes that (1.10) is equivalent to

$$\frac{d^2x}{dt^2}+\omega^2x=F(t)\ , \tag{1.11}$$

where $F(t)$ plays the role of an external force. Assuming the finite Fourier sum of (1.2) is the same as making an ansatz for $F(t)$. We now turn to examples.

II. VAN DER POL MODEL

One of the models that Delamotte treated is the Van der Pol limit-cycle model which can be written as two coupled first-order equations [2]

$$\begin{aligned}
\frac{dx}{dt}&=y\ ,\\
\frac{dy}{dt}&=-x-g(x^2-1)y\ ,
\end{aligned} \tag{2.1}$$

or as a second-order equation in x only,

$$\frac{d^2x}{dt^2}+g(x^2-1)\frac{dx}{dt}+x=0\ . \tag{2.2}$$

The model exhibits a limit cycle for arbitrary g. We illustrate the method for both sets of equations, first to review Delamotte's approach for equations in one variable and then to show that the same result can be obtained by explicitly treating the two-variable version (which is the form most commonly encountered in chemical kinetics).

We consider first the all-x version of (2.2). An important point is the consideration of the initial conditions. Since we are seeking the locus of a limit cycle we do not *a priori* know $x(t=0)$. However, since $x(t)$ is periodic we can choose an extremum in $x(t)$ as the starting point. Thus we take (the dot signifying the time derivative)

$$\dot{x}(0)=0\ . \tag{2.3}$$

In general the $N=1$ level of approximation for $x(t)$ would be

$$x=a+b\sin(\omega t)+c\cos(\omega t)\ . \tag{2.4}$$

The initial condition of (2.3) requires $b=0$ giving

$$x=a+c\cos(\omega t)=a+\tfrac{1}{2}c(\zeta+\zeta^{-1})\ . \tag{2.5}$$

Substituting (2.5) into (2.2) one obtains

$$\begin{aligned}
&A(0)=a=0\ ,\\
&\mathrm{Re}\,A(1)=\tfrac{1}{2}c(1-\omega^2)=0\ ,\\
&\mathrm{Im}\,A(1)=\tfrac{1}{2}c\omega-\tfrac{1}{2}a^2c\omega-\tfrac{1}{8}c^3\omega=0\ ,
\end{aligned} \tag{2.6}$$

which are satisfied by

$$a=0\ ,\quad c=2\ ,\quad \omega=1\ . \tag{2.7}$$

It is important to note that (2.6) do not contain the parameter g; thus for the Van der Pol model the parameters in (2.7) are independent of g. The locus of the limit cycle in first approximation is then

$$x=2\cos(t)\ . \tag{2.8}$$

The period, $T=2\pi/\omega$, is 6.283 185 and the maximum value of x is $x_{\max}=2$; these numbers compare well with the exact values (determined numerically [1] for the case of $g=1$) of $T=6.663\,287$ and $x_{\max}=2.008\,620$. Thus in

this case the lowest-order approximation is quite good. Using odd harmonics of the form $\sin(2k+1)\omega t$ and $\cos(2k+1)\omega t$ Delamotte notes that the results obtained for $k=8$ are five orders of magnitude better for the maximum amplitude and ten orders of magnitude better for the period than the values obtained using perturbation theory through g[163] (using Padé resummation; all results for the case of $g=1$). While this is remarkable, in this case the first-order approximation is already very close to the exact solution.

We now consider the set of first-order equations given in (2.1) and treat the same model explicitly as a two-variable system. Again we take $(dx/dt)_{t=0}=0$ (we can do this for only one variable since in general the extrema in the two variables will not occur at the same time). Thus the solution we assume for $N=1$ is

$$\begin{aligned} x &= a_x + c_x\cos(\omega t) = a_x + \tfrac{1}{2}c_x(\zeta+\zeta^{-1}) , \\ y &= a_y + b_y\sin(\omega t) + c_y\cos(\omega t) \\ &= a_y + \tfrac{1}{2}(c_y - ib_y)\zeta + \tfrac{1}{2}(c_y + ib_y)\zeta^{-1} . \end{aligned} \tag{2.9}$$

Substituting (2.9) into (2.1) gives

$$\begin{aligned} &A_x(0) = -a_y \, A_x(0) = -a_y , \\ &\mathrm{Re}\, A_x(1) = \tfrac{1}{2}c_y , \\ &\mathrm{Im}\, A_x(1) = \tfrac{1}{2}(b_y + c_x\omega) , \\ &A_y(0) = (a_x - a_y) + a_x^2 a_y + \tfrac{1}{2}a_y c_x^2 + a_x c_x c_y , \\ &\mathrm{Re}\, A_y(1) = \tfrac{1}{2}c_x + a_x a_y c_x - \tfrac{1}{2}c_y + \tfrac{1}{2}a_x^2 c_y + \tfrac{3}{8}c_x^2 c_y + \tfrac{1}{2}b_y\omega , \\ &\mathrm{Im}\, A_y(1) = \tfrac{1}{2}b_y - \tfrac{1}{2}a_x^2 b_y - \tfrac{1}{8}b_y c_x^2 + \tfrac{1}{2}c_y\omega , \end{aligned} \tag{2.10}$$

which are satisfied by

$$\begin{aligned} &a_x = 0 , \quad a_y = 0 , \\ &b_x = 0 , \quad b_y = -2 , \\ &c_x = 2 , \quad c_y = 0 , \\ &\omega = 1 , \end{aligned} \tag{2.11}$$

giving for the locus of the limit cycle

$$\begin{aligned} x &= 2\cos(t) , \\ y &= -2\sin(t) , \end{aligned} \tag{2.12}$$

which is of course equivalent to the results obtained from the x-only version. The importance of the two-variable approach is that in general we cannot eliminate y and produce a second-order equation in x only.

In Fig. 1 we compare the first-order approximation of (2.12) with the numerical solution of (2.1), both for the case of $g=1$. Curve a is the numerical solution while curve b is the first-order approximation. We have already pointed out that the first-order approximation gives a very good estimate of the maximum x amplitude and the frequency. What we see from Fig. 1 is that the first-order approximation does not reproduce the bulges along the y axis (which increase in magnitude with g). To see how the next order of approximation begins to incorporate this feature we follow Delamotte and use the next odd harmonic. The next-order approximation for x is then (using the x-only approach)

$$\begin{aligned} x = a &+ b(1)\sin(\omega t) + c(1)\cos(\omega t) \\ &+ b(3)\sin(3\omega t) + c(3)\cos(3\omega t) . \end{aligned} \tag{2.13}$$

The initial condition of (2.3) now gives

$$b(1) + 3b(3) = 0 . \tag{2.14}$$

The equations for the parameters are [for simplicity we set $b(1)=b$, $c(1)=c$, $b(3)=B$, and $c(3)=C$; we take $g=1$]

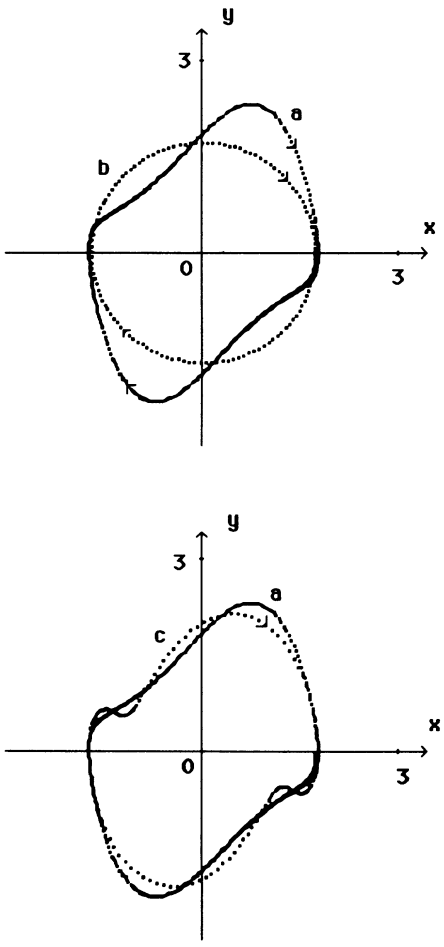

FIG. 1. Limit cycle for the Van der Pol model. Curve a is a numerical solution of (2.1) with $g=1$, curve b is the first-order approximation given by (2.12), and curve c is the third-order approximation given by (2.16). The arrows indicate the direction in which the limit cycle is traversed.

$$
\begin{aligned}
&A(0)=a=0\ ,\\
&\mathrm{Re}\,A(1)=\tfrac{1}{2}c+\tfrac{3}{2}B\omega-\tfrac{21}{4}B^3\omega-\tfrac{3}{4}BC^2\omega\\
&\qquad+\tfrac{3}{4}cBC\omega-\tfrac{1}{4}c^2B\omega-\tfrac{1}{2}c\omega^2\ ,\\
&\mathrm{Im}\,A(1)=\tfrac{3}{2}B-\tfrac{9}{8}B^2C\omega-\tfrac{1}{2}c\omega+\tfrac{5}{8}cB^2\omega+\tfrac{1}{4}cC^2\omega\\
&\qquad+\tfrac{1}{8}c^2C\omega+\tfrac{1}{8}c^3\omega-\tfrac{3}{2}B\omega^2\ ,\qquad(2.15)\\
&\mathrm{Re}\,A(3)=\tfrac{1}{2}C-\tfrac{3}{2}B\omega+\tfrac{21}{2}B^3\omega+\tfrac{3}{8}BC^2\omega-\tfrac{3}{8}c^2B\omega-\tfrac{9}{2}C\omega^2\ ,\\
&\mathrm{Im}\,A(3)=-\tfrac{1}{2}B-\tfrac{3}{2}C\omega+\tfrac{57}{8}B^2C\omega+\tfrac{3}{8}C^3\omega-\tfrac{27}{8}cB^2\omega\\
&\qquad+\tfrac{3}{4}c^2C\omega+\tfrac{1}{8}c^3\omega+\tfrac{9}{2}B\omega^2\ .
\end{aligned}
$$

The first of these equations gives $a=0$; the terms in $\mathrm{Re}\,A(2)$ and $\mathrm{Im}\,A(2)$, not shown, all contain the parameter a as a factor and thus these are identically zero. Using the parameters obtained from the solution of (2.15), Eq. (2.13) for $x(t)$ now becomes

$$
\begin{aligned}
x=&0.642\sin(\omega t)+1.912\cos(\omega t)-0.214\sin(3\omega t)\\
&+0.137\cos(3\omega t)\\
&(\omega=0.9425)\ .\qquad(2.16)
\end{aligned}
$$

Taking $y=dx/dt$ we obtain the third-order approximation for the limit cycle. This is shown in Fig. 1 as curve c. One sees that in this order of approximation the bulges along the y axis are now developing and that we have a fairly good fit to the actual limit cycle.

III. OTHER MODELS IN TWO VARIABLES

We now treat several models in two variables in lowest order, i.e., $N=1$. For all we have the form of (2.9) which incorporates the initial condition of (2.3). First we treat an exactly soluble textbook example of a two-variable model with a limit cycle. The model is given [2,3] by the equations

$$
\begin{aligned}
&\frac{dx}{dt}=y+x(1-x^2-y^2)\ ,\\
&\frac{dy}{dt}=-x+y(1-x^2-y^2)\ .
\end{aligned}\qquad(3.1)
$$

The locus of the limit cycle is the circle $x^2+y^2=1$. On the limit cycle the differential equations are

$$
\begin{aligned}
&\frac{dx}{dt}=y\ ,\\
&\frac{dy}{dt}=-x\ ,
\end{aligned}\qquad(3.2)
$$

which have the solution

$$
\begin{aligned}
&x=-\cos(t)\ ,\\
&y=\sin(t)\ .
\end{aligned}\qquad(3.3)
$$

For simplicity here we anticipate that $a_x=a_y=0$, i.e., the limit cycle is centered at the origin; we caution that the origin or a steady state need not determine the values of a_x and a_y. We substitute this simplified version of (2.9) into (3.1) and obtain the following relations:

$$
\begin{aligned}
&A_x(0)=0\ ,\\
&\mathrm{Re}\,A_x(1)=-\tfrac{1}{2}c_x+\tfrac{1}{8}b_y^2c_x+\tfrac{3}{8}c_x^3-\tfrac{1}{2}c_y+\tfrac{3}{8}c_xc_y^2\ ,\\
&\mathrm{Im}\,A_x(1)=\tfrac{1}{2}b_y-b_yc_xc_y+\tfrac{1}{2}c_x\omega\ ,\\
&A_y(0)=0\ ,\\
&\mathrm{Re}\,A_y(1)=\tfrac{1}{2}c_x-\tfrac{1}{2}c_y+\tfrac{3}{8}b_y^2c_y+\tfrac{3}{8}c_x^2c_y+\tfrac{3}{8}c_y^3+\tfrac{1}{2}b_y\omega\ ,\\
&\mathrm{Im}\,A_y(1)=\tfrac{1}{2}b_y-\tfrac{3}{8}b_y^3-\tfrac{1}{8}b_yc_x^2-\tfrac{3}{8}b_yc_y^2+\tfrac{1}{2}c_y\omega\ ,
\end{aligned}\qquad(3.4)
$$

which have the solution

$$
\begin{aligned}
&c_x=-0\ ,\quad c_y=0\ ,\\
&b_y=1\ ,\quad \omega=1\ ,
\end{aligned}\qquad(3.5)
$$

giving (3.3), the exact result.

Next we turn to the Lotka-Volterra model, the simplest two-variable system that exhibits oscillations. In this case the orbits are not true limit cycles in that there is no particular orbit toward which the system evolves. This feature will come out naturally in the present method. The model is [4]

$$
\begin{aligned}
&\frac{dx}{dt}=x-xy\ ,\\
&\frac{dy}{dt}=-y+xy\ .
\end{aligned}\qquad(3.6)
$$

There is a steady state at $x_s=y_s=1$ and the system exhibits closed orbits about this steady state with the amplitude determined by the initial conditions. The solution of (3.6) is not known but on taking the ratio of the differential equations one has

$$
\frac{dx}{dy}=\frac{x(y-1)}{y(1-x)}\ ,\qquad(3.7)
$$

which can be separated and solved to give the locus of the metastable orbit [4]

$$
\frac{1}{y}e^{y}=Cxe^{-x}\ .\qquad(3.8)
$$

Applying the present method we substitute (2.9) into (3.6) and obtain

$$
\begin{aligned}
&A_x(0)=a_x(1-a_y)-\tfrac{1}{2}c_xc_y\ ,\\
&\mathrm{Re}\,A_x(1)=\tfrac{1}{2}(c_x-a_yc_x-a_xc_y)\ ,\\
&\mathrm{Im}\,A_x(1)=+\tfrac{1}{2}(a_xb_y-c_x\omega)\ ,\\
&A_y(0)=-a_y(1-a_x)+\tfrac{1}{2}c_xc_y\ ,\\
&\mathrm{Re}\,A_y(1)=\tfrac{1}{2}(a_yc_x-c_y+a_xc_y-b_y\omega)\ ,\\
&\mathrm{Im}\,A_y(1)=\tfrac{1}{2}(b_y-a_xb_y-c_y\omega)\ ,
\end{aligned}\qquad(3.9)
$$

which has the solution

$$
\begin{aligned}
&a_x=1\ ,\quad a_y=1\ ,\\
&c_y=0\ ,\quad b_y=c_x\ ,\\
&\omega=1\ .
\end{aligned}\qquad(3.10)
$$

We note that in this case a_x and a_y are given by the steady-state values here. We are unable to specify c_x, which simply means that there is not a unique orbit. Thus we have

$$x=1+c_x\cos(t)\ ,$$
$$y=1+c_x\sin(t)\ . \tag{3.11}$$

This incidentally is the solution that one would obtain for the equations linearized about the steady state. It is not true that the $N=1$ level approximation is in general equivalent to linearizing the differential equations about the steady state as the next two examples will illustrate.

Our next example is a variant on the Lotka-Volterra model that does exhibit a proper limit cycle. This model was constructed as part of a study of the effects of cooperation, clustering of particles, and excluded volume, on the metastable orbits of the original Lotka-Volterra model [5]. The differential equations describing the model are

$$\frac{dx}{dt}=\alpha x^2(1-x-y)-xy\ ,$$
$$\frac{dy}{dt}=-\beta y+xy\ . \tag{3.12}$$

The system is constrained by the relation $(x+y)\le 1$. The steady state in the interior of the reaction simplex is given by

$$x_s=\beta,\quad \alpha=\frac{y_s}{x_s(1-x_s-y_s)}\ , \tag{3.13}$$

while the condition for a limit cycle is

$$x_s<(1-y_s)/2\ . \tag{3.14}$$

As an example of conditions under which the system exhibits a limit cycle we take $x_s=y_s=0.3$ with $\alpha=2.5$ and $\beta=0.3$.

Substituting (2.9) into (3.12) one obtains

$$\begin{aligned}
A_x(0)&=-\alpha a_x^2+\alpha a_x^3+a_xa_y+\alpha a_x^2a_y-\tfrac{1}{2}\alpha c_x^2+\tfrac{3}{2}\alpha a_xc_x^2\\
&\quad+\tfrac{1}{2}\alpha a_yc_x^2+\tfrac{1}{2}c_xc_y+\alpha a_xc_xc_y\ ,\\
\mathrm{Re}\,A_x(1)&=-\alpha a_xc_x+\tfrac{3}{2}\alpha a_x^2c_x+\tfrac{1}{2}a_yc_x+\alpha a_xa_yc_x+\tfrac{3}{8}\alpha c_x^3\\
&\quad+\tfrac{1}{2}a_xc_y+\tfrac{1}{2}\alpha a_x^2c_y+\tfrac{3}{8}\alpha c_x^2c_y\ ,\\
\mathrm{Im}\,A_x(1)&=-\tfrac{1}{2}a_xb_y-\tfrac{1}{2}\alpha a_x^2b_y-\tfrac{1}{8}\alpha b_yc_x^2+\tfrac{1}{2}c_x\omega\ ,\\
A_y(0)&=\beta a_y-a_xa_y-\tfrac{1}{2}c_xc_y\ ,\\
\mathrm{Re}\,A_y(1)&=\tfrac{1}{2}[-a_yc_x+\beta c_y-a_xc_y+b_y\omega]\ ,\\
\mathrm{Im}\,A_y(1)&=\tfrac{1}{2}[-\beta b_y+a_xb_y+c_y\omega]\ .
\end{aligned} \tag{3.15}$$

Taking $a_x=x_s=\beta$ and $c_y=0$ leads to a simple solution:

$$a_x=0.3,\quad a_y=0.266\,405\ ,$$
$$b_x=0,\quad b_y=0.197\,693\ ,$$
$$c_x=0.291\,194,\quad c_y=0\ ,$$
$$\omega=0.392\,404\ . \tag{3.16}$$

Note that while $a_x=x_s$ one has $a_y\neq y_s$. Our first-order approximation for the locus of the limit cycle is thus

$$x=0.3+0.291\cos(0.392t)\ ,$$
$$y=0.266+0.198\sin(0.392t)\ . \tag{3.17}$$

Figure 2 shows a numerical solution of (3.12) with $\alpha=2.5$ and $\beta=0.3$ which is compared with the first-order approximation of (3.17). The numerical solution of (3.12) gives $\omega=0.318$ which compares with $\omega=0.392$ in the first-order approximation of (3.16). One sees that the first-order approximation is a fair approximation to the actual limit cycle in this case.

A famous model from the Prigogine school is the Brusselator [6]:

$$\frac{dx}{dt}=A+x^2y-x(B+1)\ ,$$
$$\frac{dy}{dt}=Bx-x^2y\ . \tag{3.18}$$

The steady state is at

$$x_s=A,\quad y_s=B/A \tag{3.19}$$

and is unstable for

$$B>1+A^2\ , \tag{3.20}$$

in which case the system exhibits a limit cycle. There are no bounds on x and y in this model other than the requirement that they must lie in the positive real quadrant. We will take $A=1$ and keep B general; from (3.20) there is then a limit cycle for $B>2$.

Substituting (2.14) in (3.18) we obtain

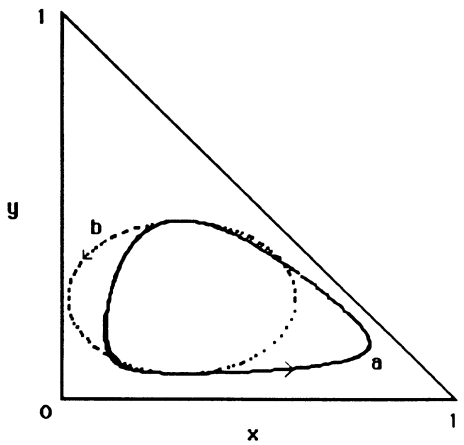

FIG. 2. Limit cycle for the modified Lotka-Volterra model. Curve a is a numerical solution of (3.12) with $\alpha=2.5$ and $\beta=0.3$, curve b is the first-order approximation given by (3.17). The system is constrained to lie in the reaction simplex shown. The arrows indicate the direction in which the limit cycle is traversed.

$$
\begin{aligned}
&A_x(0)=-1+a_x(1+B)-[+a_x^2a_y+\tfrac{1}{2}a_yc_x^2+a_xc_xc_y]\ ,\\
&\mathrm{Re}\,A_x(1)=\tfrac{1}{2}c_x(1+B)-[+a_xa_yc_x+\tfrac{1}{2}a_x^2c_y+\tfrac{3}{8}c_x^2c_y]\ ,\\
&\mathrm{Im}\,A_x(1)=[\tfrac{1}{2}a_x^2b_y+\tfrac{1}{8}b_yc_x^2]+\tfrac{1}{2}c_x\omega\ ,\\
&A_y(0)=-Ba_x+[+a_x^2a_y+\tfrac{1}{2}a_yc_x^2+a_xc_xc_y]\ ,\\
&\mathrm{Re}\,A_y(1)=-\tfrac{1}{2}Bc_x+\tfrac{1}{2}b_y\omega\\
&\qquad\qquad+[+a_xa_yc_x+\tfrac{1}{2}a_x^2c_y+\tfrac{3}{8}c_x^2c_y]\ ,\\
&\mathrm{Im}\,A_y(1)=\tfrac{1}{2}c_y\omega-[\tfrac{1}{2}a_x^2b_y+\tfrac{1}{8}b_yc_x^2]\ .
\end{aligned}
\tag{3.21}
$$

The solution of (3.21) is simplified by noting that on forming

$$
\begin{aligned}
&A_x(0)+A_y(0)=-1+a_x=0\ ,\\
&\mathrm{Re}[A_x(1)+A_y(1)]=\tfrac{1}{2}c_x+\tfrac{1}{2}b_y\omega=0\ ,\\
&\mathrm{Im}[A_x(2)+A_y(2)]=\tfrac{1}{2}c_x\omega+\tfrac{1}{2}c_y\omega=0\ ,
\end{aligned}
\tag{3.22}
$$

one has immediately (for general B)

$$
a_x=1,\quad c_x=-b_y\omega,\quad c_y=b_y\omega\ . \tag{3.23}
$$

We recall that we also have, in general, $b_x=0$ from the initial condition of (2.3). One then readily obtains the general solution of the first-order approximation for this model,

$$
\begin{aligned}
&a_y=\left|9+2B+\sqrt{-47+20B+4B^2}\,\right|/8\ ,\\
&(b_y\omega)^2=\tfrac{8}{3}(a_y-1-\tfrac{1}{2}B)\ ,\\
&\omega=\sqrt{1+\tfrac{1}{4}(b_y\omega)^2}\ .
\end{aligned}
\tag{3.24}
$$

For example, at $B=3$ one has

$$
\begin{aligned}
&a_x=1\quad a_y=2\tfrac{3}{4}\ ,\\
&b_x=0,\quad b_y=\sqrt{\tfrac{4}{7}}\ ,\\
&c_x=-\sqrt{\tfrac{2}{3}},\quad c_y=\sqrt{\tfrac{2}{3}}\ ,\\
&\omega=\sqrt{\tfrac{7}{6}}\ .
\end{aligned}
\tag{3.25}
$$

Note that $a_y\neq 3$, the steady-state value of y when $B=3$ and $A=1$. In Fig. 3 curve a gives the numerical solution of (3.18) for $A=1$ and $B=3$ while curve b gives the first-order approximation as determined by the parameters in (3.25). The numerical solution gives $\omega=0.879$ as compared with $\omega=1.080$ in the first-order approximation. In this case the first-order approximation is a very poor representation of the actual limit cycle. To understand more about the accuracy of this approximation we examine the behavior of the first-order approximation in more detail near the bifurcation point.

For $A=1$ the critical value (bifurcation point) is $B=2$. Introducing a variable that measures the distance from the critical point

$$
\epsilon=B-2 \tag{3.26}
$$

Eqs. (3.24) then yield the following expansions in ϵ about $\epsilon=0$:

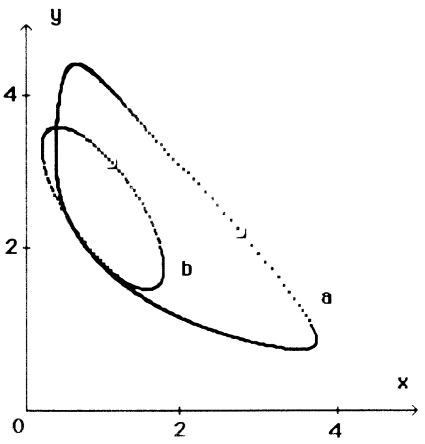

FIG. 3. Limit cycle for the Brusselator. Curve a is a numerical solution of (3.18) with $A=1$ and $B=3$ while curve b is the first-order approximation using the parameters of (3.26). The arrows indicate the direction in which the limit cycle is traversed.

$$
\begin{aligned}
&a_x=1\ ,\\
&a_y=2+\epsilon-\tfrac{2}{3}\epsilon^2+\cdots\ ,\\
&b_x=0\ ,\\
&b_y=\frac{2}{\sqrt{3}}\sqrt{\epsilon}(1-\tfrac{5}{6}\epsilon+\cdots)\ ,\\
&c_x=-\frac{2}{\sqrt{3}}\sqrt{\epsilon}(1-\tfrac{2}{3}\epsilon+\cdots)\ ,\\
&c_y=\frac{2}{\sqrt{3}}\sqrt{\epsilon}(1-\tfrac{2}{3}\epsilon+\cdots)\ ,\\
&\omega=1+\tfrac{1}{6}\epsilon-\tfrac{17}{72}\epsilon^2+\cdots\ .
\end{aligned}
\tag{3.27}
$$

We note that all of the amplitude terms increase as the square root of ϵ.

The extreme values of x [see (2.3)] are given by

$$
x_m=a_x\pm c_x\ . \tag{3.28}
$$

In Fig. 4 we plot $x_{\max}$ and $x_{\min}$, the extreme values of x on the limit cycle as a function of ϵ [or B; see (3.26)] as given by the first-order approximation and as determined by the numerical solution of (3.18). One sees that for B close to two ($\epsilon=0$) the first-order approximation estimates the amplitude of the limit cycle accurately but as B increases the approximate value levels off while the value obtained from the numerical solution keeps increasing. Figure 4 also shows ω, determined from the first-order approximation and from numerical solution of (3.18). As $B\to 2$, $\omega\to 1$; as B is increased above 2 the value of ω obtained from the first-order approximation becomes greater than one while the value obtained from the numerical solution shows the opposite trend. We also show an estimate of ω obtained from the kinetic equations linearized near the steady state. The dynamics near the steady state are given by $\exp(\lambda t)$ where the λ's are the eigenvalues obtained from the linearized equations which in this case are (for $A=1$ and $\epsilon=B-2$)

$$\lambda=\tfrac{1}{2}\epsilon\pm\sqrt{1-\tfrac{1}{4}\epsilon^2}\ . \tag{3.29}$$

Equation (3.29) shows that for $\epsilon>0$ ($B>2$) the system moves exponentially away from the steady state (the real part of λ is positive) with the quantity

$$\omega'=\sqrt{1-\tfrac{1}{4}\epsilon^2} \tag{3.30}$$

giving the rate of rotation as the system moves away from the steady state. One sees in Fig. 4 that ω' is a fair estimate of ω for the limit cycle as $\epsilon\to 0$; at $B=4$ one has $\omega'=0$ which simply means that the system does not initially oscillate as it moves away from the steady state toward the limit cycle.

Clearly we see from Fig. 4 that the first-order approximation is adequate only for values of B up to about $B=2.1$. To see how the next stage of the process improves the solution we go on to $N=2$ for the Brusselator. In this case one has

$$\begin{aligned} x&=a_x(0)+b_x(1)\sin(\omega t)+c_x(1)\cos(\omega t)\\ &\quad+b_x(2)\sin(2\omega t)+c_x(2)\cos(2\omega t)\ ,\\ y&=a_y(0)+b_y(1)\sin(\omega t)+c_y(1)\cos(\omega t)\\ &\quad+b_y(2)\sin(2\omega t)+c_y(2)\cos(2\omega t)\ . \end{aligned} \tag{3.31}$$

The condition $(dx/dt)_{t=0}=0$ now gives

$$b_x(1)+2b_x(2)=0\ . \tag{3.32}$$

We will not give the complete resulting equations in this case, but simply point out that the analogs of (3.22) for general B are

$$\begin{aligned} &a_x(0)=1\ ,\\ &c_x(1)=-\omega[b_x(1)+b_y(1)]\ ,\\ &b_x(1)=\omega[c_x(1)+c_y(1)]\ ,\\ &c_x(2)=-2\omega[b_x(2)+b_y(2)]\ ,\\ &b_x(2)=2\omega[c_x(2)+c_y(2)]\ . \end{aligned} \tag{3.33}$$

We note that (3.29) contains 11 parameters (including ω). Equations (3.32) and (3.33) allow us to eliminate six of the variables, giving five nonlinear equations in five unknowns to solve [the equations are $A_y(0)=0$, $\mathrm{Re}\,A_y(1)=0$, $\mathrm{Re}\,A_y(2)=0$, $\mathrm{Im}\,A_y(1)=0$, and $\mathrm{Im}\,A_y(2)=0$] or the corresponding A_x equations, but not both sets since the results of (3.33) are the consequence of adding the A_x and A_y equations [see (3.22)]. An iterative scheme whereby one starts by solving the $A_y(0)$ and $A_y(1)$ equations (assuming that the second-order parameters are zero), and then solving the $A_y(2)$ equations for the second-order parameters (keeping the first-order parameters constant), and so on, works well; the convergence to a solution is slow, but steady.

As an example we compare the results for the case where $B=2.1$ for $N=1$ and 2 which are shown below.

$N=1$:

$$\begin{aligned} x&=1-0.344\cos(\omega t)\ ,\\ y&=2.094+0.329\sin(\omega t)+0.344\cos(\omega t)\\ &\qquad(\omega=1.015)\ . \end{aligned} \tag{3.34}$$

$N=2$:

$$\begin{aligned} x&=1+0.108\sin(\omega t)-0.352\cos(\omega t)\\ &\quad-0.054\sin(2\omega t)+0.076\cos(2\omega t)\ ,\\ y&=2.102+0.245\sin(\omega t)+0.460\cos(\omega t)\\ &\quad+0.016\sin(2\omega t)-0.103\cos(2\omega t)\\ &\qquad(\omega=0.996)\ . \end{aligned} \tag{3.35}$$

We note that in the ($N=2$) approximation the frequency is now slightly less than one, and is in excellent agreement with that obtained from numerical solution ($\omega=0.995\pm 1$). In Fig. 5 we compare the numerical solu-

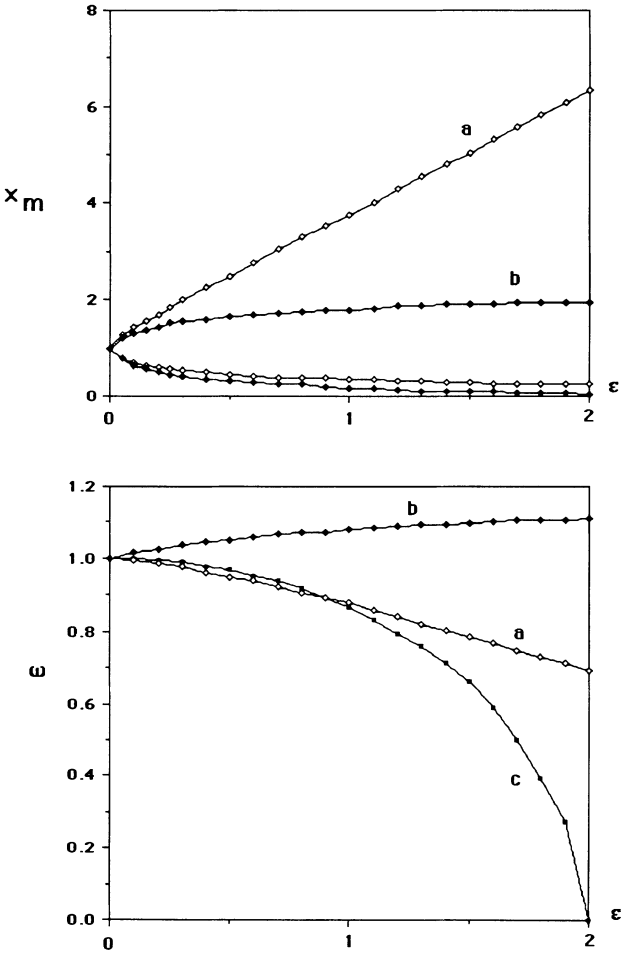

FIG. 4. Parameters for the Brusselator limit cycle as a function of the distance from the bifurcation point, $\epsilon=B-B_c$, where here $A=1$ and $B_c=2$. The upper graph shows the maximum and minimum amplitudes of x. The open squares (curve a gives $x_{\max}$) are obtained by numerical solution of (3.18), while the solid squares (curve b give $x_{\min}$) are the results of the first-order approximation given by (3.25). The lower graph gives the frequency as a function of ϵ: curve a is the numerical solution of (3.18); curve b is the first-order approximation of (3.25); and curve c uses (3.31) obtained from the linearized rate equations.

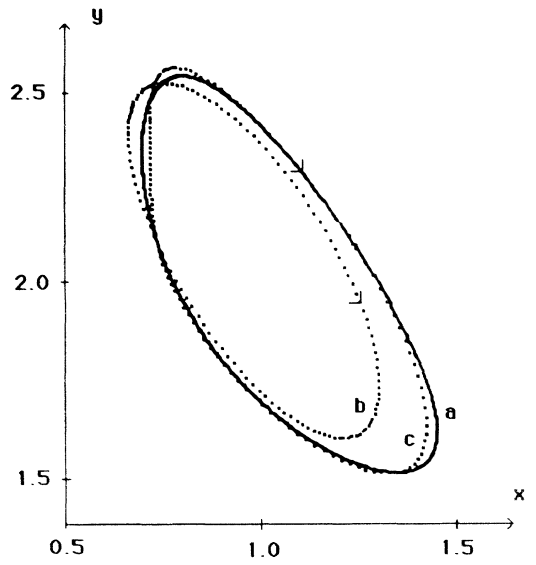

FIG. 5. Limit cycle for the Brusselator. Curve a is the numerical solution of (3.18) with $A=1$ and $B=2.1$ curve b is the first-order approximation of (3.35), and curve c is the second-order approximation of (3.36).

tion of (3.18) for $B=2.1$ (curve a) with the first-order (curve b) and second-order (curve c) approximations. Thus for $\epsilon=0.1$ the second-order approximation gives an excellent representation of the limit cycle (which contrasts with the poor results obtained in first order for $\epsilon=1$ as shown in Fig. 3).

IV. CONNECTION WITH LINEAR STABILITY ANALYSIS

In Fig. 4 we show that for the example treated the frequency of the limit cycle, both as determined by the numerical solution of the differential equations and by using the first-order approximation, approaches the value obtained from the linearized rate equations as the distance from the bifurcation point goes to zero. Here we show that this is so in general and hence that there is a connection between the present method and linear stability analysis.

We start with (1.1) and linearize the rate equations about a steady state (x_s,y_s) giving

$$\begin{aligned}\frac{d\Delta x}{dt}&=f_x\Delta x+f_y\Delta y\ ,\\ \frac{d\Delta y}{dt}&=g_x\Delta x+g_y\Delta y\ ,\end{aligned}\tag{4.1}$$

where

$$\Delta x=x-x_s,\quad \Delta y=y-y_s\ .\tag{4.2}$$

The matrix of derivatives (evaluated at the steady state)

$$\begin{bmatrix} f_x & f_y \\ g_x & g_y \end{bmatrix}\tag{4.3}$$

has the eigenvalues

$$\lambda=\tfrac{1}{2}\{(f_x+g_y)\pm\sqrt{(f_x+g_y)^2-4(f_xg_y-f_yg_x)}\}\ .\tag{4.4}$$

If the quantity in the square root is negative, then a Hopf bifurcation occurs when the real part of λ goes through zero and the nature of the steady state changes from stable to unstable (leading to a limit cycle). From (4.4) one sees that the condition for the bifurcation point is [which is equivalent to setting the trace of the matrix of (4.3) equal to zero]

$$f_x+g_y=0\ ,\tag{4.5}$$

in which case the eigenvalues are

$$\lambda=\pm i\sqrt{f_xg_y-f_yg_x}=i\omega_0\ .\tag{4.6}$$

Applying our first-order approximation to (4.1) we use (2.3) and (2.9),

$$\begin{aligned}x&=x_s+c_x\cos(\omega t)\ ,\\ y&=y_s+b_y\sin(\omega t)+c_y\cos(\omega t)\ ,\end{aligned}\tag{4.7}$$

which results in the relations

$$\begin{aligned}&\mathrm{Re}\,A_x(1)=c_xf_x+c_yf_y\ ,\\ &\mathrm{Im}\,A_x(1)=b_yf_y+c_x\omega\ ,\\ &\mathrm{Re}\,A_y(1)=c_xg_x+c_yg_y-b_y\omega\ ,\\ &\mathrm{Im}\,A_y(1)=b_yg_y+c_y\omega\ .\end{aligned}\tag{4.8}$$

Using the first two equations we can eliminate c_y and b_y,

$$\begin{aligned}c_y&=-c_x(f_x/f_y)\ ,\\ b_y&=-c_x\omega(1/f_y)\ .\end{aligned}\tag{4.9}$$

Using (4.5) and (4.9) the fourth equation becomes an identity and does not give any useful information. The third equation, however, yields the relation

$$\omega=\sqrt{f_xg_y-f_yg_x}=\omega_0\ ,\tag{4.10}$$

which is the result obtained from the eigenvalues for the linearized equations, (4.6). Near the bifurcation point the limit cycle has the form

$$\begin{aligned}x&=x_s+c_x\cos(\omega_0t)\ ,\\ y&=y_s-c_x\left|\frac{\omega_0}{f_y}\right|\sin(\omega_0t)-c_x\left|\frac{f_x}{f_y}\right|\cos(\omega_0t)\ .\end{aligned}\tag{4.11}$$

As with the case of the Lotka-Volterra model, we cannot obtain a unique value of the amplitudes (all depend on c_x) from (4.8) since in the neighborhood of the bifurcation point the orbits are metastable. What (4.11) tells us is that as one approaches the bifurcation point marking the onset of a limit cycle one has in general for a two-variable model

$$\begin{aligned}&a_x\to x_s,\quad a_y\to y_s,\quad \omega\to\omega_0\ ,\\ &\frac{b_y}{c_x}\to-\frac{\omega_0}{f_y},\quad \frac{c_y}{c_x}\to-\frac{f_x}{f_y}\ ,\\ &\frac{b_y}{c_x}\to-\frac{\omega_0}{f_y},\quad \frac{c_y}{c_x}\to-\frac{f_x}{f_y}\ .\end{aligned}\tag{4.12}$$

Thus in the neighborhood of the bifurcation point the magnitude of c_x is the only unknown quantity. To start a numerical search for the parameters the relations in (4.12) can be used to eliminate all of the variables except c_x. The full nonlinear form of the first-order approximation equations [the nonlinear analog of (4.8)] of course does yield values of all of the parameters including c_x. The present approach thus can be seen as a method by which one can go beyond the limits of linear stability analysis (which tells one that there is an instability leading to a limit cycle) and actually construct the limit cycle.

V. DISCUSSION

We have applied Delamotte's method for obtaining a hierarchy of approximations to the locus of a limit cycle for a second-order differential equation in one variable to the case of a set of coupled first-order differential equations in two or more variables. The method quickly leads to a set of coupled nonlinear equations in the unknown parameters required by the method. The solution of these equations is the main technical problem involved with the method. For our examples in two variables the first-order approximation requires the determination of six unknown parameters while the second-order approximation requires ten [this includes the frequency ω and the use of the initial condition (2.3)]. We have noted in general that the constant $a_x(0)$ in (1.4) cannot always be assumed to be identical with the steady state value of x (or y as the case may be); in all of Delamotte's examples [1] the limit cycle is assumed to be centered at $x=0$.

Delamotte makes the point that this method does not rely on the smallness of a critical parameter (such as is the case in perturbation expansions). While it is true that one can use the first order and higher orders of approximation for any values of the parameters involved (it is possible that no solution of the equations can be found in real numbers for certain extreme values of a parameter, but we have not found this to be the case for any of the models we have treated here) it is also clear from our analysis of the Brusselator that the first-order approximation works better the closer one is to the bifurcation point. In Fig. 4 one see that the x amplitude obtained from the first-order approximation levels off with ϵ (measuring the distance from the bifurcation point), while the actual amplitude keeps on increasing. Thus in a sense this is an asymptotic method, improving at low order as one approaches the onset of the limit cycle (bifurcation point). We note again that all of the coefficients change at each level of approximation, and that this is not an approximation that gives a fixed contribution at first order, and so on.

The method we have outlined here for first-order equations in several variables is a useful complement to linear stability analysis. Linear stability analysis can predict the onset of limit cycles and it can give the limiting value of their frequencies at the bifurcation point, but it cannot actually describe the limit cycle. The present method allows one to find a simple analytic expression for the limit cycle that is accurate near the bifurcation point.

Finally we point out that the loci of limit cycles (at least stable limit cycles) are easy to determine by numerical solution of the differential equations involved (the system is literally pulled toward the correct locus). Thus if one wants a mathematical expression that will represent the numerical results one can take a set of (x,y) points around the cycle together with the numerical value of the frequency and use (1.2) to fit the data. The a's, b's, and c's in (1.2) are then unknowns to be determined, but the numerical problem is one of solving simultaneous linear equations, which is easy to accomplish (and unique) no matter how many points are used. One could use this approach to see how many terms are required to represent the limit cycle to any required degree of accuracy. Of course the goal here is to obtain an approximate analytic form directly from the differential equations without resorting to numerical solutions.

[1] B. Delamotte, Phys. Rev. Lett. **70**, 3361 (1993).
[2] P. G. Drazin, *Nonlinear Systems* (Cambridge University Press, New York, 1992).
[3] D. A. Sanchez, *Ordinary Differential Equations and Stability Theory: An Introduction* (Dover, New York, 1979), p. 82.
[4] H. T. Davis, *Introduction to Nonlinear Differential and Integral Equations* (Dover, New York, 1962), p. 102.
[5] D. Poland, Physica D **35**, 148 (1989).
[6] G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley-Interscience, New York, 1977).