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Modeling of heat explosion with convection

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The work is devoted to numerical simulations of the interaction of heat explosion with natural convection. The model consists of the heat equation with a nonlinear source term describing heat production due to an exothermic chemical reaction coupled with the Navier–Stokes equations under the Boussinesq approximation. We show how complex regimes appear through successive bifurcations leading from a stable stationary temperature distribution without convection to a stationary symmetric convective solution, stationary asymmetric convection, periodic in time oscillations, and finally aperiodic oscillations. A simplified model problem is suggested. It describes the main features of solutions of the complete problem. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1695211]

Natural convection can change critical conditions of heat explosion and can also lead to complex oscillations and oscillating heat explosion. We observe two routes to complex oscillations. One of them is due to a sequence of period-doubling bifurcations and Sharkovskii sequences. Different oscillating regimes can coexist for the same values of parameters, in particular, chaotic and periodic oscillations. Another type of aperiodic solutions is related to quasiperiodic oscillations where several vortices oscillate with different frequencies without their synchronization. In this case, solutions are not structurally stable and can essentially change under a small change of parameters. We have developed a simplified model problem. This is a system of ordinary differential equations for an average temperature and stream function. The model problem shows the main features of the complete problem: simple and oscillating heat explosion, simple and complex oscillations. An important property of the model problem is the presence of homoclinic orbits and bifurcation of periodic solutions from them. Structurally unstable oscillations are observed for it also.

I. FORMULATION OF THE PROBLEM

The modern theory of heat explosion begins with the works due to Semenov,[1,2] who introduced the simplest model

$$\frac{d\theta}{dt}=e^{\theta}-\alpha\theta, \tag{1}$$

based on the assumption that the temperature θ is everywhere the same inside the reactor, and that the consumption of the reactants can be neglected. The first term in the right-hand side of Eq. (1) represents heat production by an exothermal chemical reaction, and the second term heat loss through the reactor walls. The next stage in the development of the theory of heat explosion is related to the works by Frank-Kamenetskii.[3] He removed the assumption that the temperature was homogeneous in space and studied the equation

$$\frac{\partial\theta}{\partial t}=\Delta\theta+ke^{\theta}, \tag{2}$$

with the boundary condition $\theta=0$ for the dimensionless temperature at the boundary of the domain. This model was studied in a number of physical and mathematical works (see Refs. 4–6). One of the limitations of the model is that it neglects reactant depletion. This approximation is justified if the characteristic time of reactant depletion is much longer than the characteristic time to reach steady state. It is applicable if $(T_b-T_0)/T_0\gg R_0T_0/E$, where T_0 is the initial temperature, T_b the adiabatic temperature, E the activation energy, and R_0 the universal gas constant (see, e.g., Ref. 6).

One of the later developments of the theory was related to the influence of natural convection on heat explosion. Merzhanov and Stessel[7] were first to study the model

$$\frac{\partial\theta}{\partial t}+u\frac{\partial\theta}{\partial x}+v\frac{\partial\theta}{\partial y}=\Delta\theta+ke^{\theta}, \tag{3}$$

$$\frac{\partial u}{\partial t}+u\frac{\partial u}{\partial x}+v\frac{\partial u}{\partial y}=-\frac{\partial p}{\partial x}+P\left(\frac{\partial^2u}{\partial x^2}+\frac{\partial^2u}{\partial y^2}\right), \tag{4}$$

$$\frac{\partial v}{\partial t}+u\frac{\partial v}{\partial x}+v\frac{\partial v}{\partial y}=-\frac{\partial p}{\partial y}+P\left(\frac{\partial^2v}{\partial x^2}+\frac{\partial^2v}{\partial y^2}\right)+R\theta, \tag{5}$$

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}=0, \tag{6}$$

where the reaction-diffusion equation was coupled with the Navier–Stokes equations in the Boussinesq approximation applicable if the density dependence on temperature and pressure is sufficiently weak. They showed that the critical condition of heat explosion could be changed by convection. The same model was studied in Refs. 8–11, where it was shown that the interaction of heat explosion and convection could lead to stable or unstable periodic oscillations and to oscillating heat explosion. It is characterized by oscillations with growing amplitude at the first stage of the development of the process, and by monotonically growing temperature

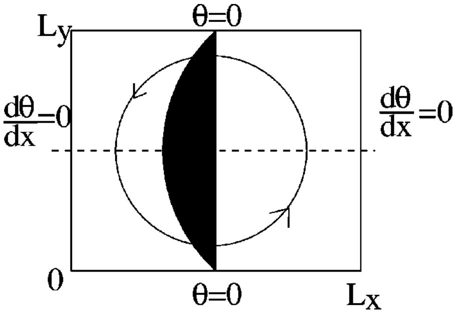

FIG. 1. Schematic representation of the stationary solution θ_S and a convective vortex.

during the explosion itself where the temperature becomes unbounded. Influence of natural convection on reaction fronts in a chemical reactor is studied in Ref. 12 with reactant depletion taken into account, and a variety of spatiotemporal structures is described.

In this work we continue to study heat explosion with convection. We will describe complex dynamics of solutions including chaotic oscillations and chaotic heat explosion. We specify that u in Eqs. (3)–(6) is the horizontal component of the velocity, v the vertical component, P is the Prandtl number, and R is the Rayleigh number. This system of equations is considered in the rectangular domain $0 \leqslant x \leqslant L_x, 0 \leqslant y \leqslant L_y$, with the boundary conditions

$$x=0,L_x: \quad \frac{\partial \theta}{\partial x}=0, u=0, \frac{\partial v}{\partial x}=0;$$

$$y=0,L_y: \quad \theta=0, v=0, \frac{\partial u}{\partial y}=0, \qquad (7)$$

or

$$x=0,L_x: \quad \theta=0, u=0, v=0; \quad y=0,L_y: \quad \theta=0, v=0, u=0. \qquad (8)$$

The boundary condition for the temperature means that the lateral walls of the reactor are adiabatic in (7) and with a constant temperature in (8), the lower and the upper walls are kept at a constant temperature. The boundary condition (7) provides the existence of a one-dimensional stationary solution $\theta(y)$ if the parameter k is less than a critical value k_c. If $u=v=0$, the system corresponds to Eq. (2). For $L_y=2$, $k_c \approx 0.88$ (see Ref. 3). In the case of boundary conditions (8), stationary solutions without convection do not exist. The boundary condition for the velocity is the so-called free-surface boundary condition in (7) and no-slip boundary conditions in (8).

II. COMPLEX DYNAMICS

A. Boundary conditions (7)

We begin with boundary conditions (7), where there exists a stationary solution $\theta_s(y)$ that depends only on the vertical space variable. For small Rayleigh numbers it is stable, and for large Rayleigh numbers it loses its stability and convective regimes appear. The stationary solution and a convective vortex are shown schematically in Fig. 1. Linear stability analysis of the stationary solution and numerical simulations of problems (3)–(7) are carried out in Ref. 10 in the case of a square domain. The stability analysis is done for a simplified problem where the exponential in (3) is replaced by a linear function. This approximation is justified and gives the onset of convection close to that observed numerically if the maximal temperature for the stationary solution is not very close to the critical temperature $\theta_c \approx 1.2$ where the explosion occurs.

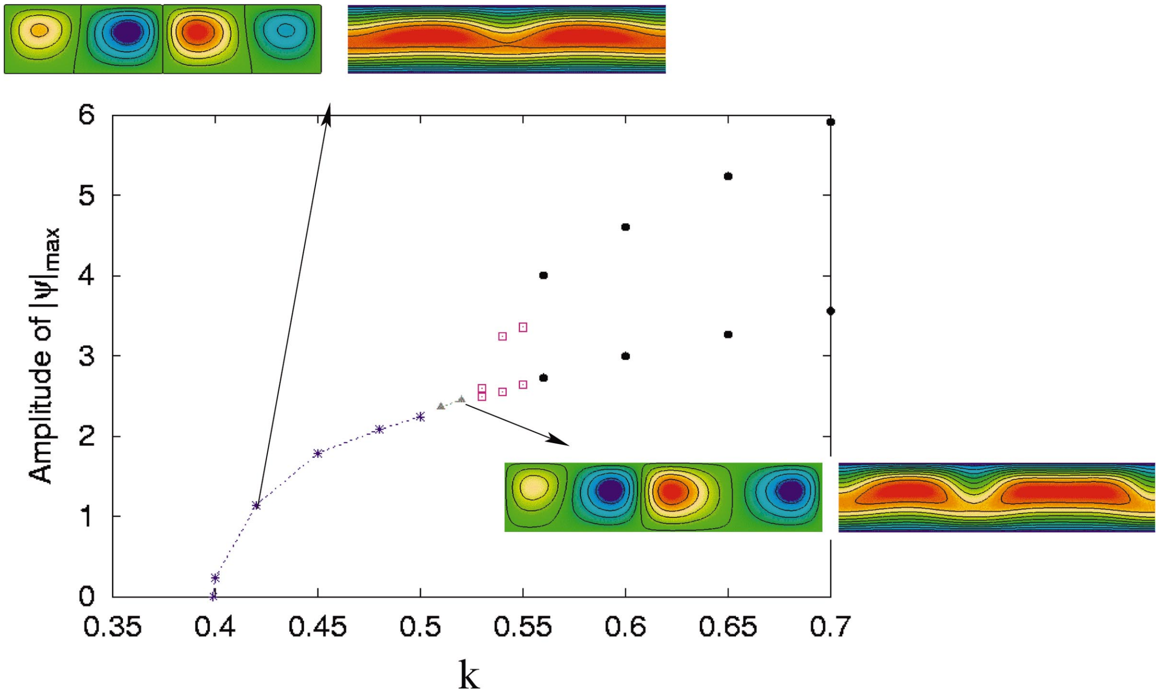

FIG. 2. (Color) Bifurcation diagram for $R=1000$ and $L_x=8$.

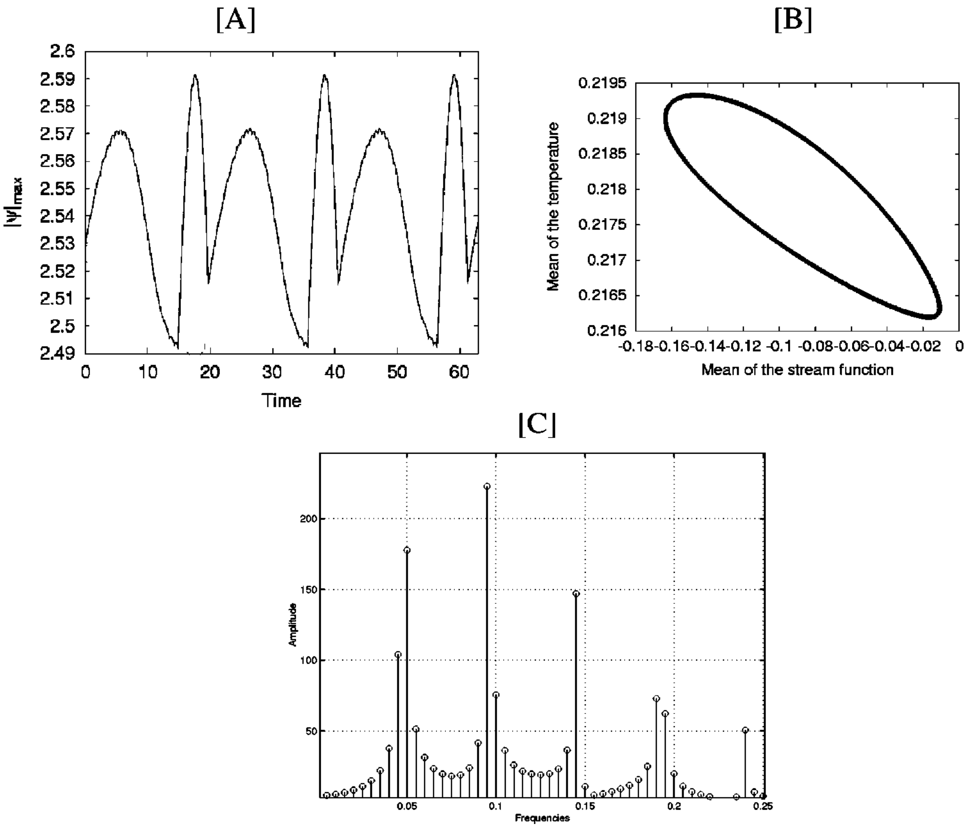

FIG. 3. Oscillations analysis for $L=8$ and $k=0.53$. (A) Evolution in time of $|\Psi|_{\max}$; (B) phase space trajectory of the mean of the temperature as a function of the mean of the stream function; (C) spectral analysis.

When an eigenvalue of the linearized problem crosses the origin, a branch of convective solutions bifurcates from the stationary nonconvective solution. Two branches of convective solutions can intersect, resulting in stability exchange and secondary bifurcations.

We present here the results of numerical simulations in the case of the rectangular domain where the behavior of solutions is essentially different compared with the square domain and where complex oscillations can be observed. Figure 2 represents the bifurcation diagram for $L_x=8$ with k as the bifurcation parameter. Stationary solutions are shown with a unique point denoting the maximum of the stream function. Periodic in time solutions are shown with two points on the same vertical line, representing the amplitude of oscillations of the maximum of the stream function.

For k sufficiently small, the one-dimensional stationary regime without convection is stable. A convective solution bifurcates for $k \approx 0.4$. It has four vortices and it is symmetric with respect to the central line of the domain. The secondary bifurcation occurs for $k \approx 0.51$, where an asymmetric four-

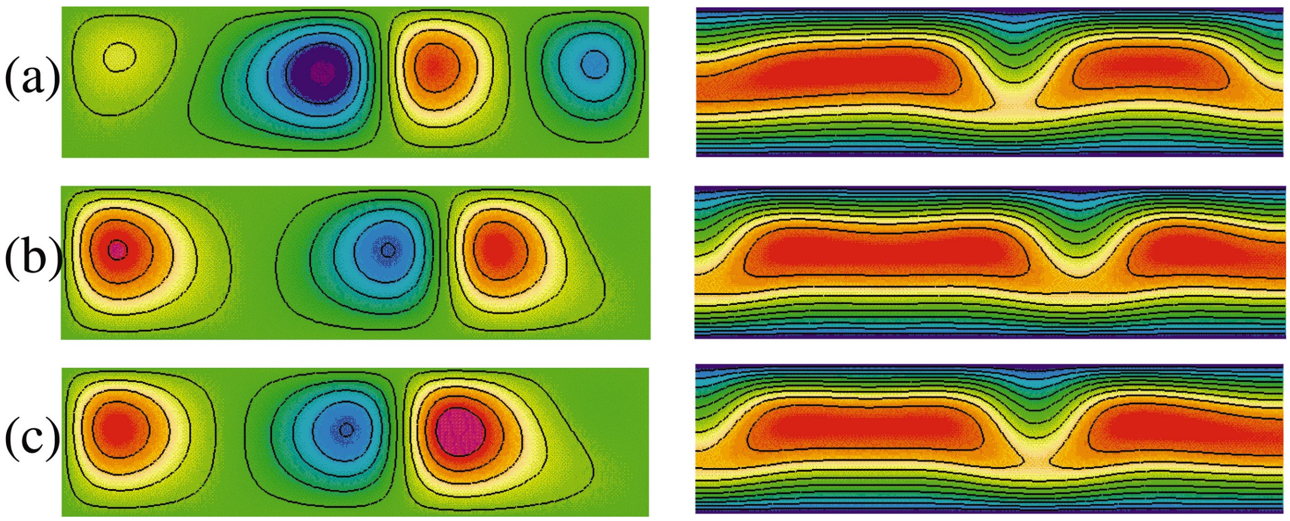

FIG. 4. (Color) Streamlines and isotherms for the case $L_x=8$, $R=1000$, and $k=0.54$: Periodic solution; (a), (b), (c) correspond to moments of time shown by the same letters in Fig. 5(A).

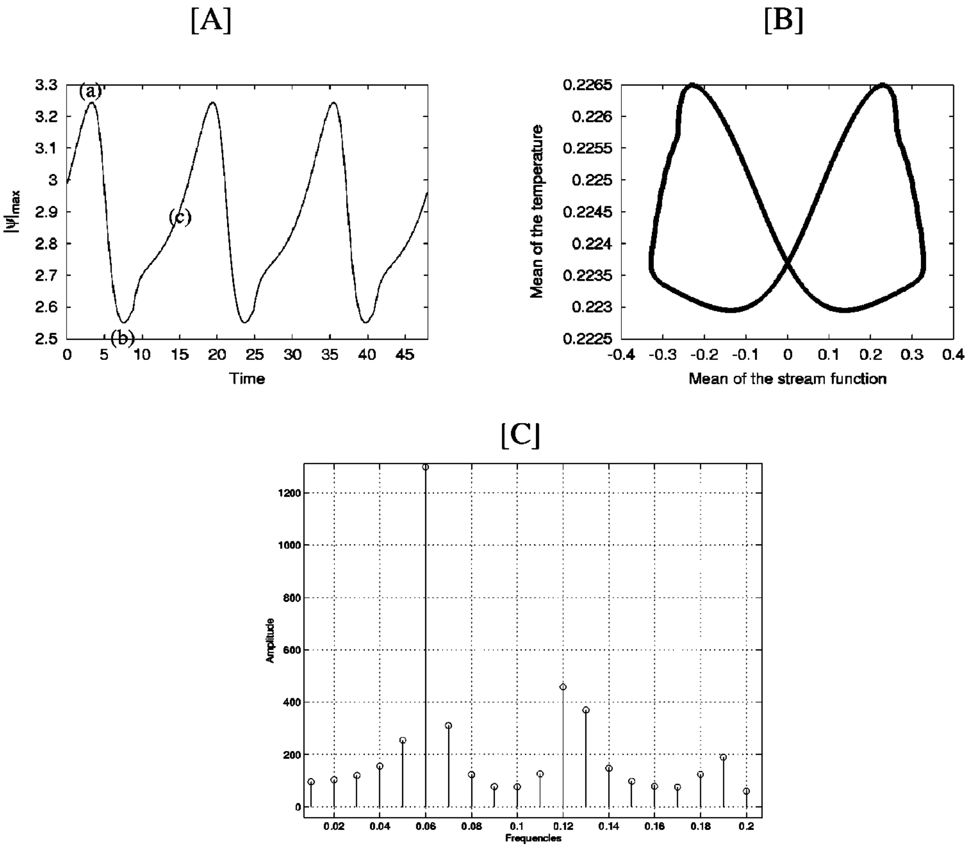

FIG. 5. Oscillations analysis for $L=8$ and $k=0.54$. (A) Evolution in time of $|\Psi|_{\max}$; (B) phase space trajectory of the mean of the temperature as a function of the mean of the stream function; (C) spectral analysis.

vortex stationary convective regime appears. The corresponding temperature and stream function distributions are shown in Fig. 2.

Further increase of k leads to appearance of periodic in time solutions. The first one is observed for $k=0.53$. It has a small amplitude and it is close to the asymmetric stationary four-vortex regime. This situation allows us to assume that it is a supercritical Hopf bifurcation. The maximum of the stream function for this solution is shown in Fig. 3(A). This curve is periodic, with two peaks in each period. In fact, the

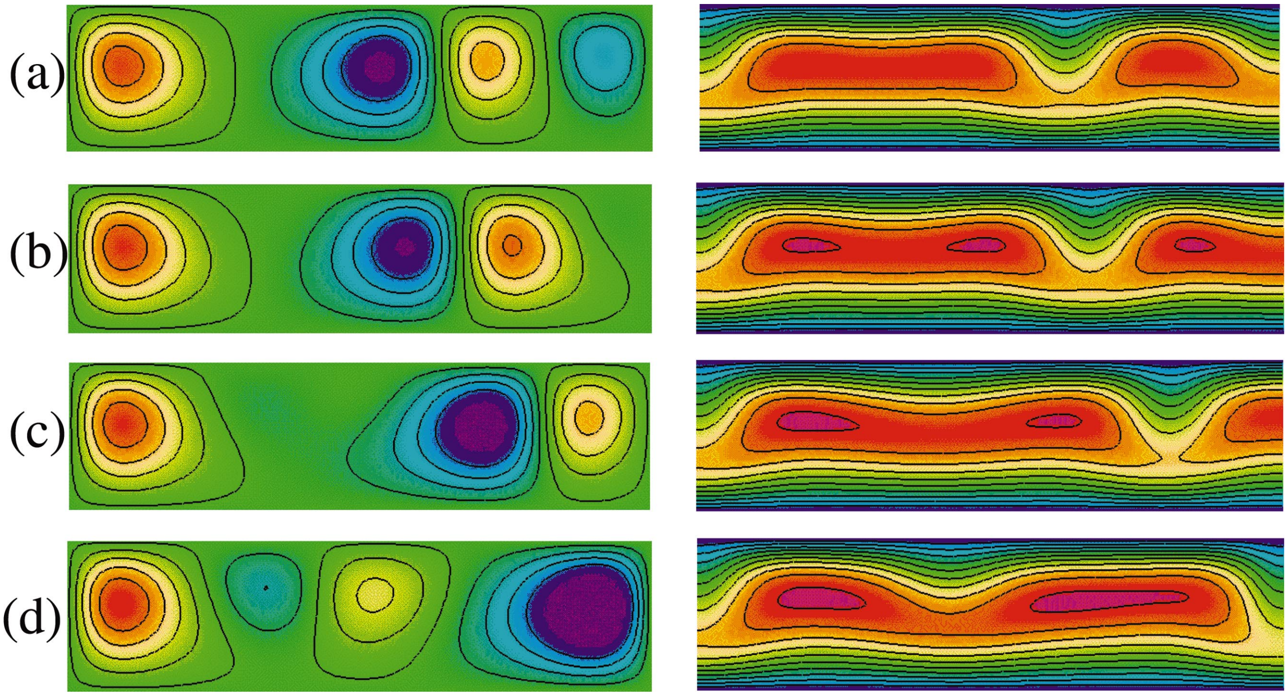

FIG. 6. (Color) Streamlines and isotherms for the case $L_x=8$, $R=1000$, and $k=0.56$: Periodic solution; (a), (b), (c), (d) correspond to moments of time shown by the same letters in Fig. 7(A).

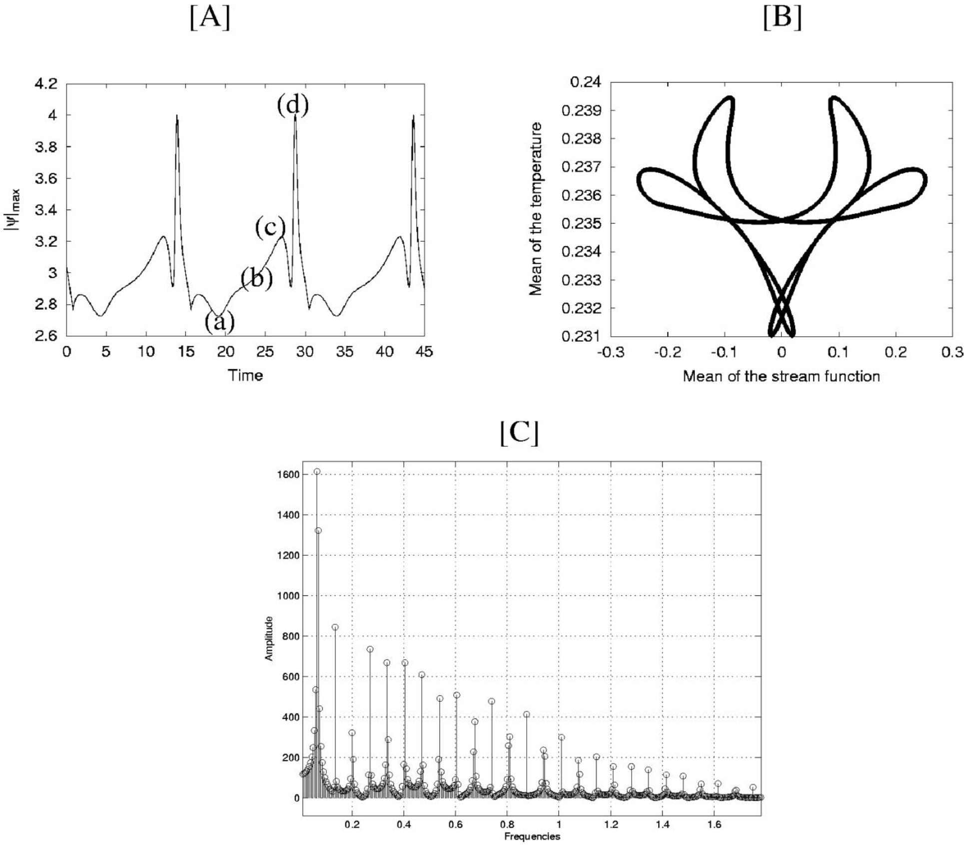

FIG. 7. Oscillations analysis for $L=8$ and $k=0.56$. (A) Evolution in time of $|\Psi|_{\max}$; (B) phase space trajectory of the mean of the temperature as a function of the mean of the stream function; (C) spectral analysis.

maximum of the stream function is not reached all the time at the same vortex. It changes its place periodically from one of the central vortices to another one. If we measure the maximum of the stream function only at the left central vortex, we would have a periodic sinus-like curve, the same with the right central vortex. In Fig. 3(A) these two curves are superposed and the maximum of them is taken.

Figure 3(B) represents the solution on the plane "mean value of the stream function—mean value of the temperature." We will call it the $(\Psi-\theta)$ plane. The solution is represented by a simple elliptic curve.

Figure 3(C) shows the Fourier modes of the function in Fig. 3(A). The main two frequencies, 0.05 and 0.095, correspond to oscillations of both central vortices. The interaction of the vortices produces all frequencies with a multiple of 0.005.

Periodic in time solutions are observed also for greater values of k, though the structure of oscillations is different. The solution for $k=0.54$ is shown in Fig. 4. The number of vortices changes here from three to four. The maximum of the stream function is a sinus-like curve [see Fig. 5(A)] with the period about 16.7. The spectral analysis gives the main frequency about 0.06, which corresponds approximately to the period indicated above [see Fig. 5(C)].

It is interesting to note that the form of the solution on the $(\Psi-\theta)$ plane differs from that in the previous case. Now, it is a curve symmetric with respect to zero of the stream function. This means that on the first and second half-periods the solution is exactly the same except for the sign of the stream function.

Further increase of k leads once again to another type of periodic oscillation. A specific example is shown in Fig. 6 for

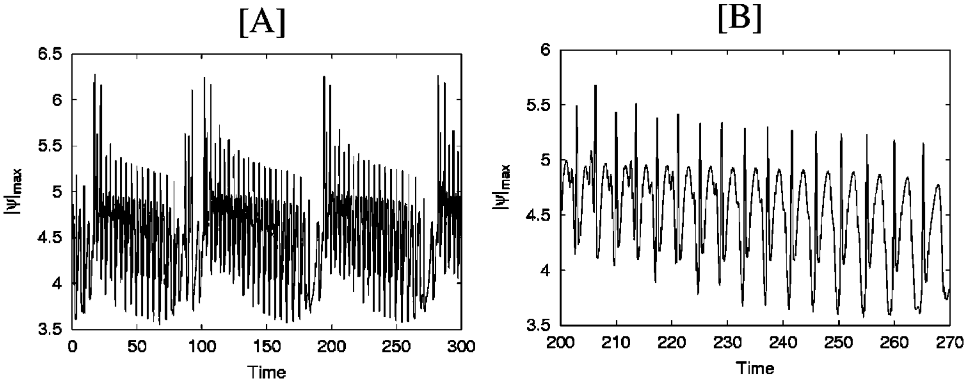

FIG. 8. Evolution in time of $|\Psi|_{\max}$ for $R=1000$, $k=0.7$, and $L_x=9.4$. (A) $0<t<300$; (B) $200<t<270$.

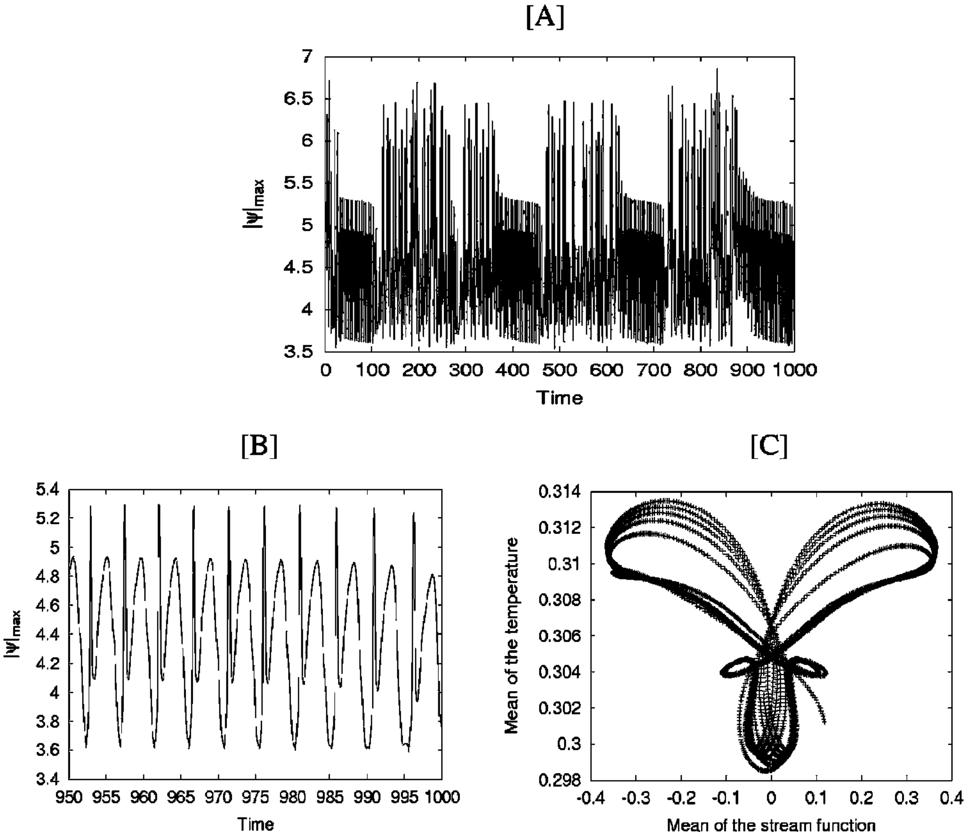

FIG. 9. Oscillations for $R=1000$, $L=9.5$, and $k=0.7$. (A) Evolution in time of $|\Psi|_{max}$; (B) zoom on the pattern; (C) phase space trajectory of the mean of the temperature as a function of the mean of the stream function.

$k=0.56$. Once again, three to four vortices are observed during the period. The curve on the $(\Psi-\theta)$ plane is symmetric [see Fig. 7(B)].

The Fourier modes of the function Ψ_{max} fill the real axis in a rather dense way [see Fig. 7(C)]. It allows us to propose a possible scenario of transition to this branch of solutions: it can be a secondary Hopf bifurcation, where a second pair of complex conjugate eigenvalues passes the imaginary axis. The frequencies of the corresponding eigenfunctions are in a rational relation with the frequencies corresponding to the first pair of eigenvalues because the resulting regime is still periodic. However, they should be close to each other. Then, the nonlinear interaction of these oscillations will produce all other frequencies shown in Fig. 7(C).

Other interesting regimes appears for larger values of L_x. Figure 8 shows an example of modulated oscillations for $L_x=9.4$. Then, more complex oscillations (with chaotic windows, Fig. 9) are observed for $L_x=9.5$. Figure 10 shows the stream function and the temperature for several consecutive moments of time. The number and the structure of vortices change here aperiodically. Finally, new periodic in time regimes appears for $L_x=9.6$ (with an unusual phase space trajectory, Fig. 11) and $L_x=9.7$ (Fig. 12). The continuation method, where we gradually changed the length and moved along the branches of solutions, did not allow us to determine the character of the transition between these regimes: neither supercritical nor subcritical bifurcations were found (see the discussion in Sec. IV).

B. Boundary conditions (8)

The principal difference of this case with respect to the previous one is that there are no stationary solutions without convection. Complex dynamics of oscillating solutions appears to be also quite different. For fixed space dimensions and R we vary k. We observe several different branches of solutions (Fig. 13). For each of them the specific behavior is characterized by transition from simple oscillations to chaos, then back to periodic regimes, and then again to chaos.

We recall briefly that such sequences of biburcations, sometimes called Sharkovskii sequences, were discovered in 1964[13] and are well studied for mappings $x_{n+1}=F(x_n)$ (see, e.g., Ref. 14). The sequence of period-doubling bifurcations is followed by chaotic oscillations. Beyond the first chaos, there are sequences of periodic oscillations with periods $\cdots 7\cdot 2^n, 5\cdot 2^n, 3\cdot 2^n \cdots$ alternating with chaotic oscillations. The sequence of bifurcations is ended by oscillations with periods 7,5,3. Oscillations with a period different from 2^n indicate the presence of chaos.

Similar sequences of bifurcations are observed for other problems where the order of bifurcations is not necessarily the same (see Ref. 15). Sequences of period-doubling bifurcations are found numerically in problems of flame propagation.[16,17]

In our case, first two branches of solutions begin with period-doubling bifurcations [Figs. 14(a)–14(c)] and end with period three oscillations [Figs. 14(e)–14(f)] in agreement with Sharkovskii sequences. We have also observed period six oscillations on the first branch. Next, two branches end with chaotic oscillations (Fig. 15). Period five and ten oscillations are found on the fourth branch. On the last branch simple periodic oscillations are directly followed by chaotic oscillations, and then by transition from chaotic oscillations to heat explosion (Fig. 16). In some cases the behavior of solutions is extremely sensitive to parameters. So,

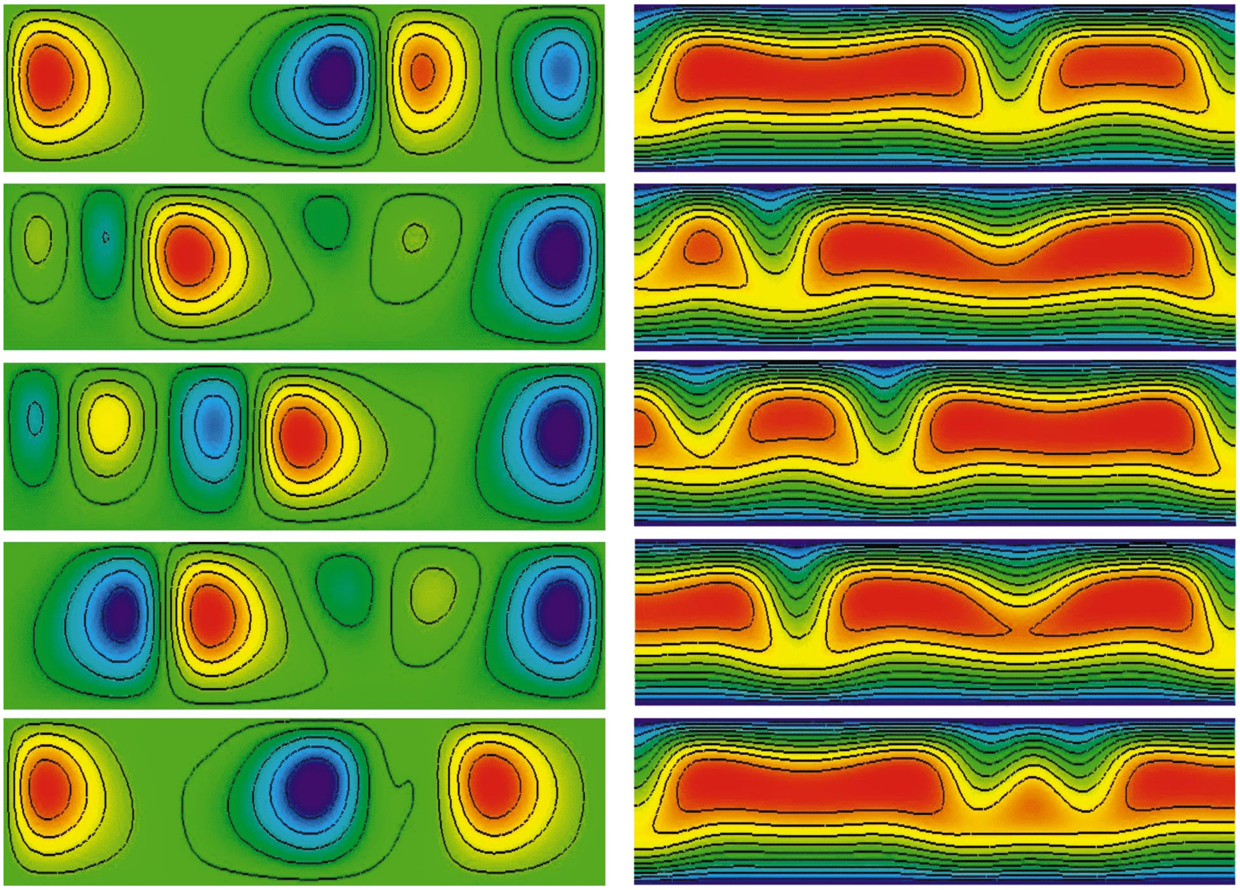

FIG. 10. (Color) Streamlines and isotherms for the case $L_x = 9.5$, $R = 1000$, and $k = 0.7$.

oscillations with other periods may also exist but are not found.

We note finally that different branches can coexist for the same value of k. In particular, chaotic oscillations and periodic oscillations can be observed for the same values of parameters.

III. MODEL PROBLEM

A. System of two equations

In this section we show how Semenov's model of heat explosion can be adapted to the case with convection. In this case the coefficient of heat loss α should be proportional to the intensity of convection. If we measure this intensity by an average stream function ψ, then $\alpha = \alpha(\psi)$. We will consider for simplicity a linear dependence, $\alpha(\psi) = \alpha_0 + k\psi$, where α_0 and k are positive constants. On the other hand, numerical simulations of problem (3)–(6) with the boundary condition (8) show that the intensity of convection is well approximated as a linear function of an average temperature. Thus, the evolution of ψ can be described by the equation

$$\frac{d\psi}{dt} = a\theta - b\psi. \tag{9}$$

We obtain a closed system (1), (9) with respect to the variables (θ,ψ). Models of this type were first studied in Ref. 10, where instead of (9) an equation for a supercritical bifurcation describing the appearance of convection in problem (3)–

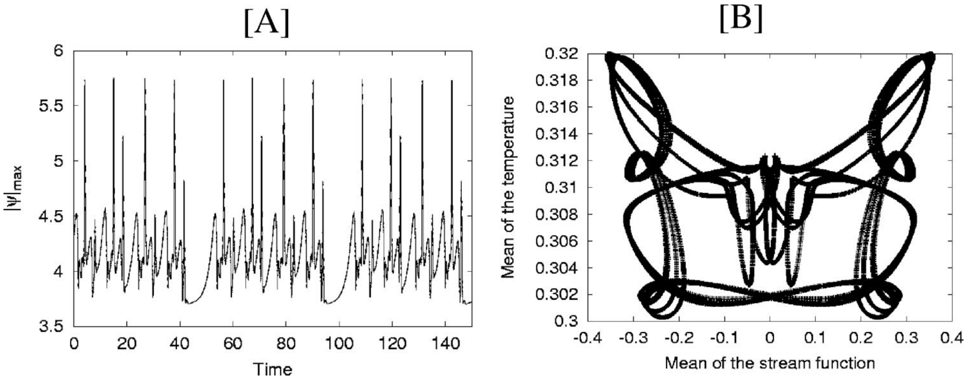

FIG. 11. Length $L_x = 9.6$. (A) Evolution in time of $|\Psi|_{\max}$; (B) phase space trajectory of the mean of the temperature as a function of the mean of the stream function.

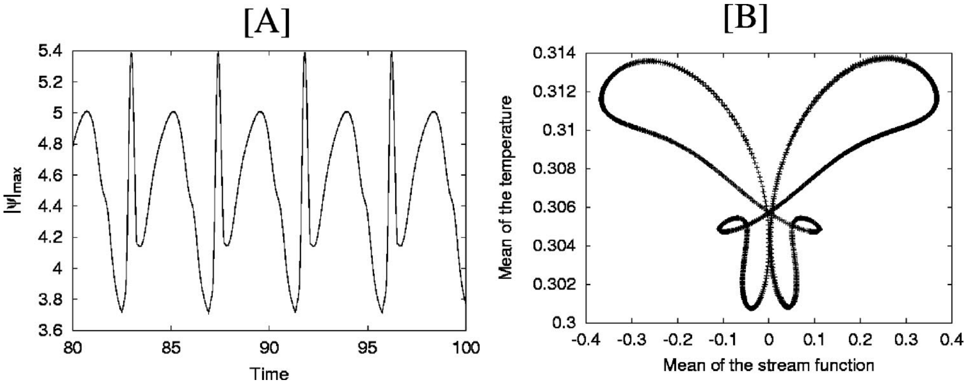

FIG. 12. Length $L_x = 9.7$. (A) Evolution in time of $|\Psi|_{\max}$; (B) phase space trajectory of the mean of the temperature as a function of the mean of the stream function.

(7) was considered. For problem (3)–(6), (8) convection does not appear as a result of a bifurcation. It exists for all temperature distributions being proportional to the average temperature.

This model shows three basic regimes observed for the complete model: stable stationary solutions, oscillations, and explosion. We discuss here the most interesting case where system (1), (9) has two stationary points, denoted by (θ^1, ψ^1) and (θ^2, ψ^2), and explain briefly its qualitative behavior.

The point (θ^2, ψ^2) with larger values of θ and ψ is a saddle, while another one can change its type depending on the parameters. The key property of this model is that it can have a loop of separatrix of the saddle point. A small change of the system leads generically to disappearance of the homoclinic orbit and to a bifurcation of a limit cycle from it.[18] The cycle is stable if $\exp(\theta^2) - \alpha(\psi^2) < b$, and unstable if the inequality is opposite. If the cycle is stable, the system exhibits stable periodic oscillations. If it is unstable, the solution either converges to the stable stationary point (θ^1, ψ^1) (Fig. 17, orbit A) or goes to infinity, which corresponds to heat explosion (Fig. 17, orbit B).

Further change of parameters can lead to the retraction of the unstable limit cycle to the point (θ^1, ψ^1) and to its disappearance due to a subcritical Hopf bifurcation. In this case, the point (θ^1, ψ^1) becomes an unstable focus. For an initial condition in a neighborhood of this point, the solution shows oscillations with growing amplitude that, finally, result in explosion (see Fig. 18).

B. Coupled oscillators

Thus, the model problem allows us to explain the new phenomenon, oscillating heat explosion, observed also for the complete problem. The system of two equations cannot describe oscillations more complex than the periodic ones. However, the presence of a homoclinic solution suggests that, for a more complete model, complex oscillations can also be observed.

If numerical simulations of the complete system show the presence of two vortices, then for each of them we can introduce the average temperature and stream function. Therefore, we will have a system of four equations with two subsystems of the same type as above. The coupling between them corresponds to heat exchange between the vortices or to momentum exchange due to viscosity

$$\frac{d\theta_1}{dt} = e^{\theta_1} - \alpha_1(\psi_1)\theta_1 - \sigma_\theta(\theta_1 - \theta_2), \tag{10}$$

$$\frac{d\psi_1}{dt} = a \quad \theta_1 - b \quad \psi_1 - \sigma_\psi(\psi_1 - \psi_2), \tag{11}$$

$$\frac{d\theta_2}{dt} = e^{\theta_2} - \alpha_2(\psi_2)\theta_2 - \sigma_\theta(\theta_2 - \theta_1), \tag{12}$$

$$\frac{d\psi_2}{dt} = a \quad \theta_2 - b \quad \psi_2 - \sigma_\psi(\psi_2 - \psi_1), \tag{13}$$

where

$$\alpha_1(\psi_1) = \alpha_0 + k_1\psi_1, \quad \alpha_2(\psi_2) = \alpha_0 + k_2\psi_2.$$

In the case of symmetric vortices, $k_1 = k_2$. We should note that their contribution to the heat loss is determined not only by the average of the stream function but also by their size and geometry. Therefore, if the vortices are different, we may have $k_1 \neq k_2$.

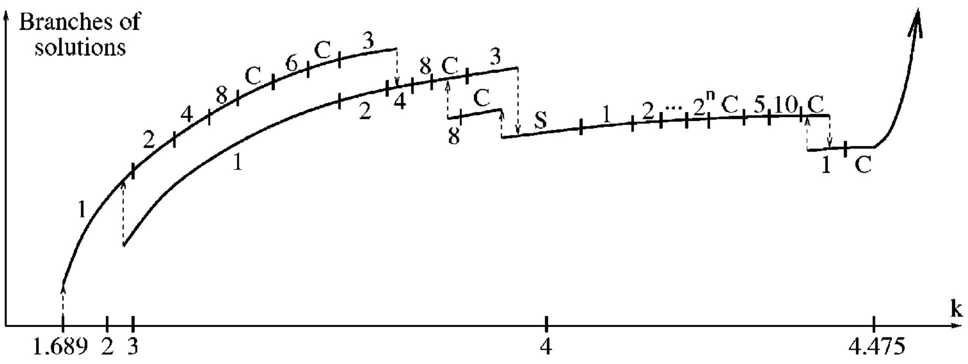

FIG. 13. Schematic representation of branches of solutions; S—stationary oscillations; C—chaotic oscillations; 1—simple periodic oscillations; 2—period-two oscillations; m—period-m oscillations; the dashed lines show transitions between the branches and the arrow shows the transition to explosion.

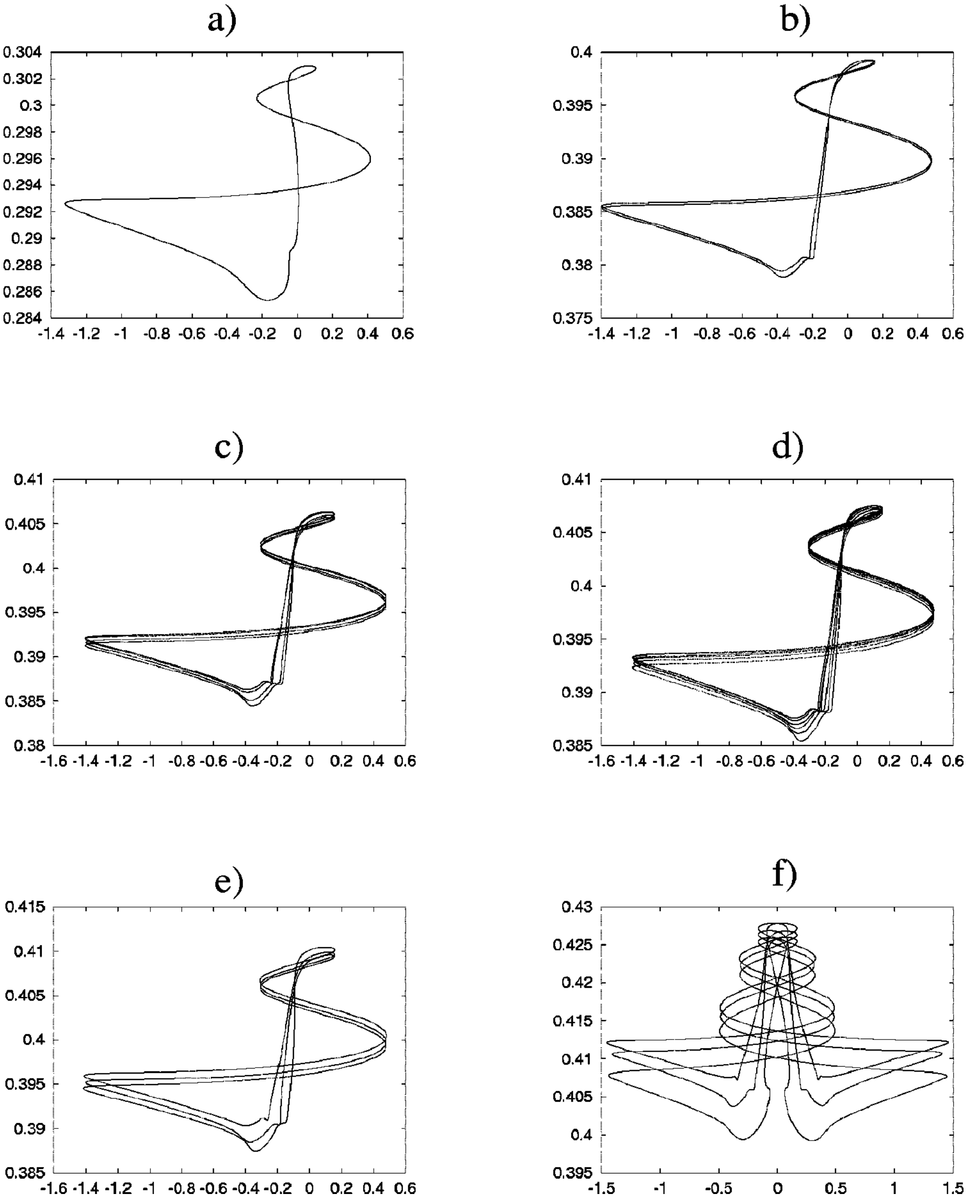

FIG. 14. (a) 1-periodic, $k=2.4$; (b) 2-periodic, $k=3.0$; (c) 4-periodic, $k=3.038$; (d) 6-periodic, $k=3.0448$; (e), (f) 3-periodic solutions ($k=3.06$, $k=3.15$).

In the case $k_1=k_2$, and if the initial conditions are such that $\theta_1^0=\theta_2^0$, $\psi_1^0=\psi_2^0$, then the behavior of the solution is exactly the same as for the system of two equations. As above, there exists a homoclinic orbit and periodic solutions. The homoclinic orbit persists under a small perturbation of the system.[19] However, if $k_1\neq k_2$, then it may be not a limit cycle but a more complex structure that bifurcates from it. Figure 19 shows an example of such trajectory in the case where $\sigma_\psi=0$. The behavior is qualitatively the same if $\sigma_\psi\neq 0$.

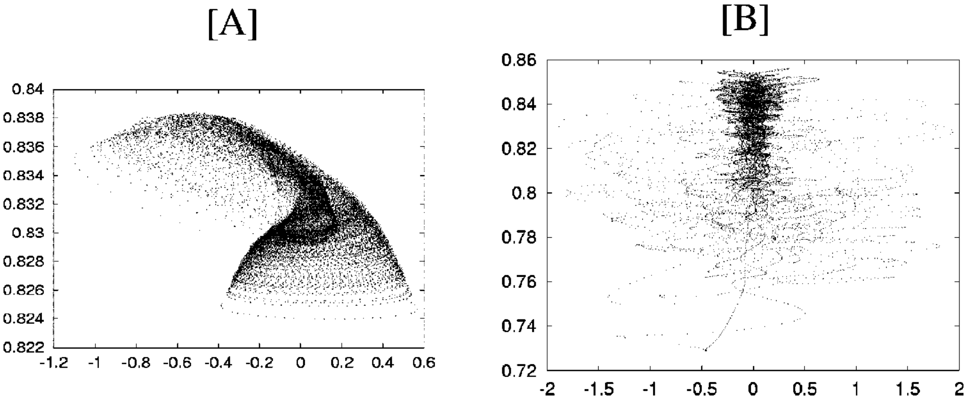

FIG. 15. Chaotic oscillations. (A) $k=4.364$; (B) $k=4.38$.

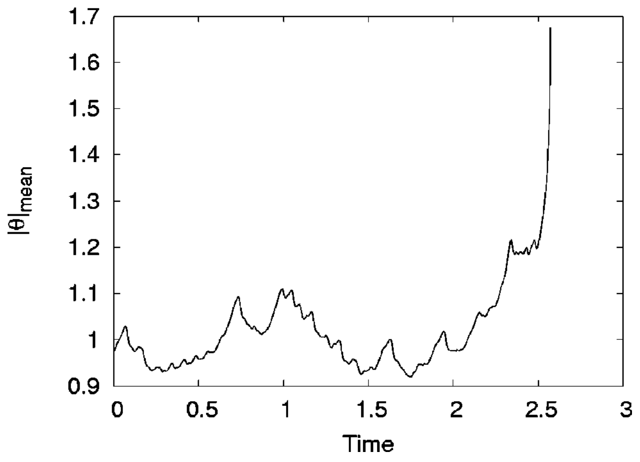

FIG. 16. Chaotic heat explosion: evolution of the average temperature in time ($k=4.475$).

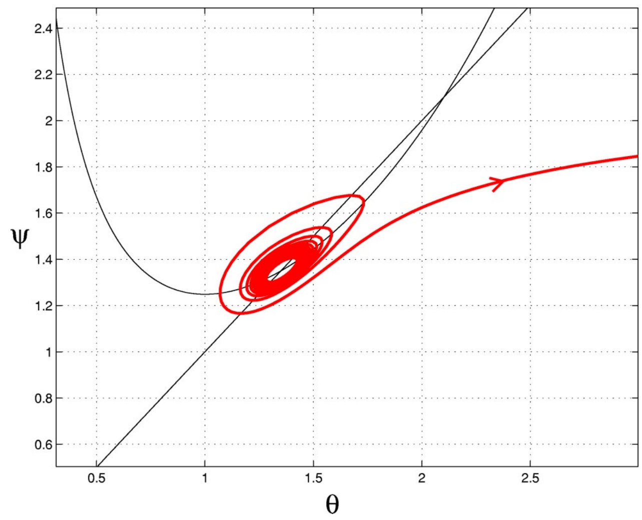

FIG. 18. (Color online) Oscillating heat explosion.

IV. CONCLUSIONS

Natural convection can have an essential influence on heat explosion including the critical conditions of the explosion, the routes to it, and the structure of the regimes in the case where the explosion does not occur, that is, the temperature remains bounded. In this work, we study complex nonlinear dynamics in the problem of heat explosion with convection. We show how successive bifurcations can lead to complex oscillations, either through a sequence of period-doubling bifurcations and Sharkovskii sequences, or through quasiperiodic oscillations. We have found a chaotic heat explosion where the temperature oscillates aperiodically before exploding.

Different solutions can coexist for the same values of parameters. For example, there can be chaotic and periodic oscillations.

In the case where the length of the rectangle is considered as a bifurcation parameter (Sec. II A), we observe a transition from periodic to aperiodic oscillations, and vice versa. We were interested in how to construct the branches of solutions and determine the types of bifurcations underlying these transitions. However, these transitions do not seem to occur through bifurcations, and these are not continuous branches of solutions. This is probably a single but discontinuous (!) branch of solutions: quasiperiodic oscillations may not be structurally stable and can essentially change under a small perturbation of the problem.

A simplified model problem based on the Semenov's model of heat explosion with an additional equation for the averaged stream function gives a good description of the main features of the problem: existence or nonexistence of stationary solutions, simple or oscillating heat explosion, periodic oscillations.

An important property of the model system, that explains some aspects of heat explosion with convection, is the presence of homoclinic orbits with stable or unstable periodic solutions bifurcating from them. Oscillating heat explosion can be observed when the limit cycle is unstable.

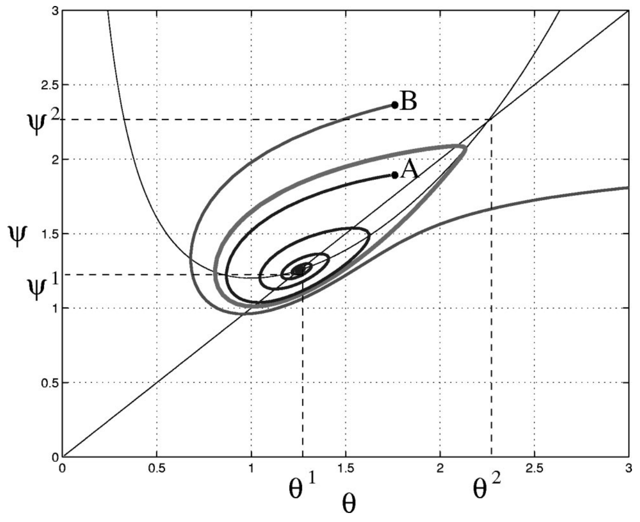

FIG. 17. Trajectories of system (1), (9): unstable limit cycle, trajectory converging to the stable focus (A), trajectory going to infinity (B).

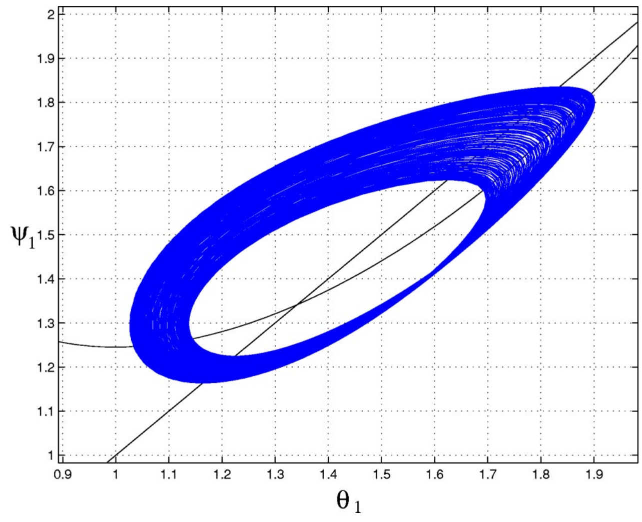

FIG. 19. (Color online) Complex oscillations for the model system (10)–(13).

If we take into account a possible existence of several vortices, and consider an average temperature and stream function for each of them, then the model problem consists not of two equations but, in the case of two vortices, of four equations. In this case, the system can exhibit more complex dynamics.

We have studied structural stability of solutions for the model system (10)–(13). If $k_1 = k_2$, we observe simple periodic orbits. If $k_1 \neq k_2$, and the difference of the two values is very small, the trajectory behaves as shown in Fig. 19. It does not converge to the simple periodic orbit as k_1 approaches k_2.

This can be explained with the following example. Consider the parametrically given curve

$$x = \tfrac{1}{2}(\cos t + \cos(1+\epsilon)t), \quad y = \tfrac{1}{2}(\sin t + \sin(1+\epsilon)t).$$

If $\epsilon = 0$, it is a circle. For any irrational ϵ this curve fills densely the interior of the circle. Therefore, there is no continuous dependence of the curve on the parameter.

Numerical simulations of both the complete and the simplified problem allow us to conclude that vortices can oscillate without synchronization of their frequencies, and the solutions may not be structurally stable.

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