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Three-dimensional modes of unsteady solid-flame combustion

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Mathematical simulation is being widely used in modeling wave propagation in various physical, chemical, and biological processes, such as gasless combustion, frontal polymerization, etc. Numerical simulation is especially important in three-dimensional (3D) modeling of the spinning and chaotic waves propagating in nontransparent solids. In this paper, we analyze the system of equations that describes the propagation of a self-sustained wave through a cylindrical sample of combustible mixture. In this case, sample composition, sample radius, and heat loss from the sample surface will be used as variable parameters. We will describe (i) the combustion modes that give rise to periodic screw motion of one or several hot spots, (ii) inner wave structure, (iii) effect of parameters on the wave structure, (iv) some modes that have not yet been observed experimentally, (v) a loss of periodicity that leads to chaotic propagation of a 3D self-sustained wave. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1540772]

In this paper, we will consider the propagation of an unsteady three-dimensional combustion front during self-propagating high-temperature synthesis, a well-known method for production of advanced materials. Earlier publications in the field largely treated the steady propagation of a planar front. In some cases, the planar front was found to twist and give rise to formation of one (or several) hot spots that propagated alongside the screw trajectory over the sample surface. Using numerical simulation, we determined the inner structure of the unsteady waves as well as described different modes of their propagation and accompanying phenomena leading to chaos.

I. INTRODUCTION

The propagation of a combustion wave during the so-called self-propagating high-temperature synthesis (SHS, also synthesis by combustion, or combustion synthesis) is known[1] to be a self-sustained process supported by the heat released in an exothermic reaction. Once initiated at the top of a cylindrical sample (Fig. 1), the combustion wave propagates downward over the entire sample. Here we will confine ourselves only to the so-called solid-flame combustion[2] when all starting reactants and reaction products are present in their solid state. This implies that the processes of gas evolution, melting, crystallization, etc., will be neglected. Under this assumption, we may concentrate on the most essential features and diminish the number of system parameters. We will take into account only heat-related factors, such as heat release, internal heat transfer, and heat loss through the sample surface.

In the case of a planar front, the problem is one-dimensional (1D). Unsteady wave propagation was first predicted upon solution of the 1D problem:[3] the planar combustion front was found to propagate at a changing velocity and temperature (pulsating propagation). When the front temperature is relatively high (well above the adiabatic temperature of the front), the velocity of front propagation is high. This period was termed "splash." The splash is followed by the period of "depression" when the temperature and propagation velocity become low. The extinction of combustion was found to happen during the period of depression. The range of system parameters within which the front loses its stability (for both 1D and multi-D cases) was found[3] to obey

$$\alpha_{\mathrm{st}}=9.1\frac{cRT_b^2}{E_aQ}-2.5\frac{RT_b}{E_a}<1,$$

where α_{st} is some combination of reaction parameters: heat capacity c, universal gas constant R, combustion (burning) temperature T_b, apparent activation energy of reaction E_a, and heat release in reaction per unit volume Q.

The rise of instability can be explained as follows. During steady propagation of a planar wave, there exist some balance between the heat release in reaction and heat transfer toward starting material. But when E_a is sufficiently high (or T_b is low), this balance becomes upset. During the depression period, the evolved heat is rapidly abstracted from the reaction zone to the zone of starting material. This diminishes both the front temperature and velocity of its motion. As the unreacted reagents are warmed up, the heat abstraction from the reaction zone decelerates until a moment when the heat loss stops to suppress the reaction. This gives rise to a splash during which the front temperature exceeds the adiabatic one. The high-temperature wave begins to propagate over a preheated reactive mixture. But gradually the reaction front enters the area of less preheated mixture, which diminishes the front temperature and, accordingly, the rate of its propagation. At last, the reaction zone comes to the area where the heat transfer toward unreacted mixture becomes prevailing, that is, we again come to the period of depression.

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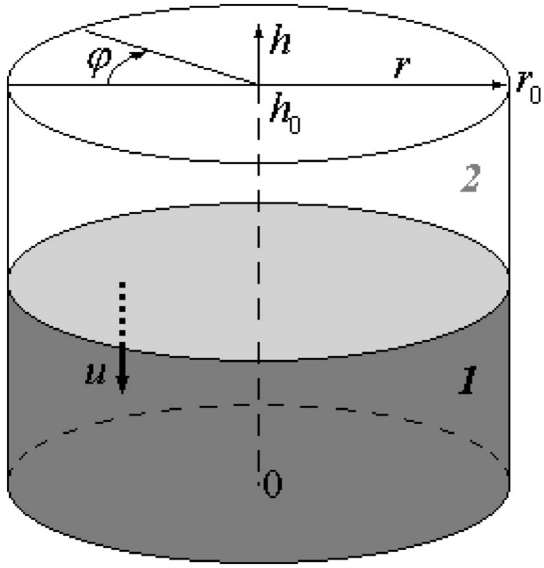

FIG. 1. Sample geometry ***1***—starting mixture; ***2***—product.

The spinning combustion—screw motion of one or several bright hot spots alongside a screw trajectory—was first experimentally observed[4] for combustion of Hf in an N_2-Ar mixture. The reaction was found to proceed in a thin near-surface layer. This observation made a basis for modeling combustion of a thin-wall cylinder,[5] that is, for two-dimensional modeling. Subsequent studies in the field were carried out by Bayliss and Matkowsky.[6]

The effect of heat losses was also investigated for other modes of combustion. Given that the distribution of T and η is independent of φ, these quantities may be assumed to be identical for any cross section of the sample. This provides grounds to analyze the 2D problem[7] with account of only r and h (upon neglect of φ).

However, the experimentally observed unsteady 3D waves[8] can be modeled only in terms of the 3D problem. Note that the structure of single-spot wave observed for SHS[8] is essentially the same as that obtained in case of frontal polymerization.[9] Other combustion modes—periodic multispot and aperiodic ones—have also been observed experimentally[10–16] but no reasonable explanation has been provided by the authors. Explicit analytical solution to such a complicated system of differential equations is impossible. Meanwhile, the approximate analytical solution[17,18] gave only a rough and strongly distorted pattern of the phenomenon under investigation. Various unsteady processes have been reviewed by Barzykin and Merzhanov.[19]

Our preliminary data[20] on solid-flame combustion under adiabatic conditions show the possibility of the following combustion modes: (i) one-spot combustion (steady, and unsteady with pulsation of T alongside the sample axis), (ii) two-spot combustion, (iii) three-spot combustion (symmetric and asymmetric), and (iv) multispot combustion. We also determined the range of stability for these combustion modes and demonstrated the nonuniqueness of solution (several modes are possible for the same set of system parameters).

Our numerical calculations also illustrate the important role of heat loss in the occurrence of that or another combustion mode. Moreover, some modes were found to exist only in the presence of heat losses. In this paper, we will describe the above-mentioned combustion modes in more detail, their dependence on system parameters, and the phenomena leading to chaos.

II. FORMULATION OF THE PROBLEM

Let us consider the following simple model that takes into account only heat transfer processes in the sample, macrokinetics of reaction, and heat loss to the environment. During combustion, all reagents and products are assumed to remain in their solid state while thermophysical parameters are independent of temperature and degree of conversion. In this case, we may use a widely adopted system of equations written in the cylindrical coordinates:

$$c\rho_0\frac{\partial T}{\partial t}=\lambda\left(\frac{\partial^2 T}{\partial r^2}+\frac{1}{r}\frac{\partial T}{\partial r}+\frac{1}{r^2}\frac{\partial^2 T}{\partial\varphi^2}+\frac{\partial^2 T}{\partial h^2}\right)+\rho_0 Q\frac{\partial\eta}{\partial t},$$

$$\frac{\partial\eta}{\partial t}=\begin{cases} k_0(1-\eta)\exp\left(-\dfrac{E_a}{RT}\right) & \text{for}\quad \eta<1,\\ 0\quad \text{for}\quad \eta\geqslant 1,\end{cases}$$

with the following boundary conditions:

$$t=0:\quad T=T_0,\quad \eta=0,$$

$$t>0,\quad r=r_0:\quad \frac{\partial T}{\partial r}=-\alpha_N(T-T_0),$$

$$h=0:\quad \lambda\frac{\partial T}{\partial h}=0,$$

$$h=h_0:\quad \begin{cases} T=T_{\mathrm{ign}} & \text{for}\quad t\leqslant t_{\mathrm{ign}},\\ \lambda\dfrac{\partial T}{\partial h}=0 & \text{for}\quad t>t_{\mathrm{ign}}.\end{cases}$$

Here T is temperature, η is the extent (degree) of conversion for a deficient component, ρ_0 is the mean density of sample material (assumed to remain unchanged), T_0 is the initial sample temperature, t is time, r, φ, h are the cylindrical coordinates (Fig. 1); r_0, h_0 are the radius and height of cylinder; λ is the thermal conductivity; k_0 is the pre-exponential factor; T_{ign} is the ignition temperature, t_{ign} is the duration of igniting pulse, and α_N is the Newton coefficient of heat transfer from the sample surface into environment.

The above-mentioned system of equations was solved in the following dimensionless variables:

$$x=\frac{r}{h_*},\quad z=\frac{h}{h_*},\quad \tau=\frac{t}{t_*},\quad \tau_{\mathrm{ign}}=\frac{t_{\mathrm{ign}}}{t_*},$$

$$R_0=\frac{r_0}{h_*},\quad \theta=\frac{(T-T_b)E_a}{RT_b^2},\quad \theta_{\mathrm{ign}}=\frac{(T_{\mathrm{ign}}-T_b)E_a}{RT_b^2}$$

$$\alpha=\frac{\alpha_N h_*}{\lambda},\quad \mathrm{Ar}=\frac{RT_b}{E_a},\quad \mathrm{Td}=\frac{cRT_b^2}{E_aQ},\quad \theta_0=-\frac{1}{\mathrm{Td}}.$$

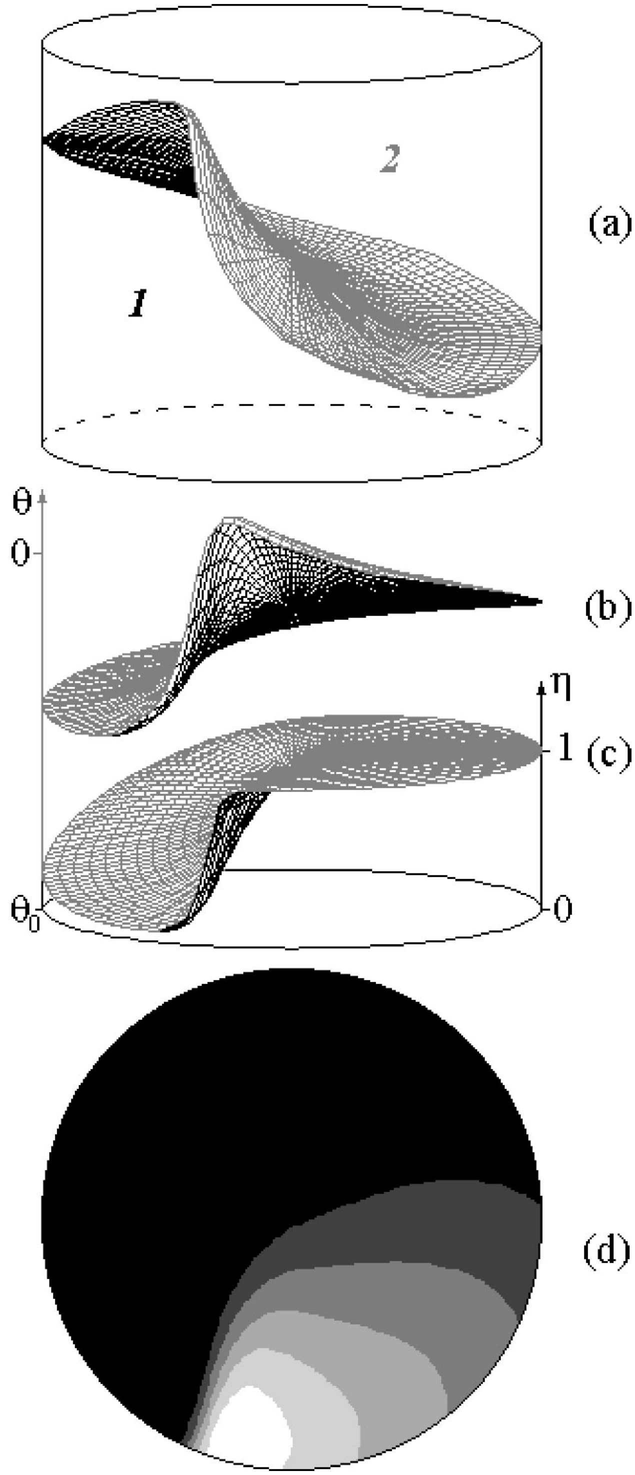

FIG. 2. Steady one-spot spinning wave: $\alpha_{st}\approx 0.97$, Td=0.14, Ar=0.12, $R_0=23$, $\alpha=0.1$. Wave front structure (a). Spatial distribution of (b) θ and (c) η in the cross section of θ_{max}. The distribution of θ in the form of isotherms (d). Here and hereinafter dark areas correspond to $\theta<-1$. Neighboring isotherms have been obtained at a pitch of $\theta=0.5$.

Here $T_b=T_0+Q/c$ is the adiabatic temperature of steady burning, $h_*=\sqrt{\lambda t_*/(c\rho)}$ is the characteristic width of the reaction zone,

$$t_*=\frac{cRT_b^2}{k_0E_aQ}\exp\left(\frac{E_a}{RT_b}\right)$$

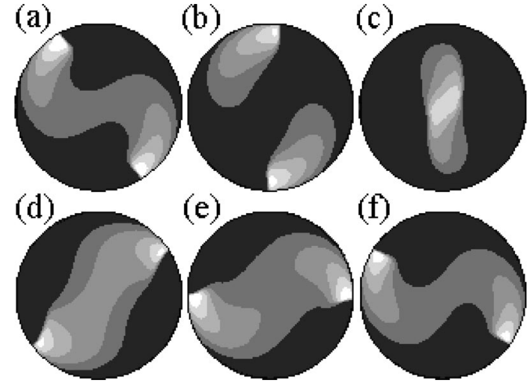

FIG. 3. Unsteady two-spot symmetric spinning wave: $\alpha_{st}\approx 1.1$, Td=0.16, Ar=0.14, $R_0=68$, $\alpha=1.5$. Still frames (a)–(f) illustrate the motion of spots. Near the surface, the spots move clockwise.

is the characteristic reaction time, Ar and Td are the Arrhenius and Todes numbers.[21] The problem was solved (in the dimensionless form) by finite difference method by using a nonuniform adaptable spatial grid (with an unfixed number of nodes).

The following quantities were used as problem parameters: dimensionless sample radius R_0 (ratio of real radius to the characteristic length of the reaction zone), the extent of penetration into the range of instability α_{st}, and the coefficient of heat loss α. Under adiabatic conditions, T_b corresponds to $\theta=0$.

We will consider the narrow zones of reaction, so that the reaction front can be regarded as a manifold of points with $\eta=0.5$. A planar front (as in Fig. 1) is observed for $\alpha_{st}>1$ and $\alpha=0$. Reaction is initiated at the top, so that it then propagates downward.

III. STRUCTURE OF STEADY SPINNING WAVE

Since a single-spot spinning wave propagating alongside a screw trajectory is most frequently observed in experiments, let us first consider the structure of such a wave.

The structure of the combustion front is given in Fig. 2(a). This overall view can be compared with the front structure suggested in experimental studies.[8,9] The distribution of θ in Fig. 2(b) and η in Fig. 2(c) is given for the same moment of time in the cross section bearing a point with θ_{max}. This cross section is front-bound and moves downward as the sample burns out. The distribution of θ is nonuniform: it exhibits a high-temperature area (hot spot) where $T>T_b$, that is, $\theta>0$. The spot moves clockwise. In this case, θ is sufficiently high just ahead of the hot spot where the mixture remains to be virtually unreacted. Here the heat flux from the hot spot becomes sufficient for ignition, so that the hot spot gradually shifts into this zone. This can be explained by the ignition of unreacted but already warmed-up reagents, which gives rise to clockwise rotation of the hot spot. Since all this cross section also moves downward, we obtain a resultant motion alongside the screw trajectory. The distribution of θ, Fig. 2(d), in the form of isotherms is given for the same moment of time and in the same cross section. Here and in the following the brighter the area, the higher the temperature.

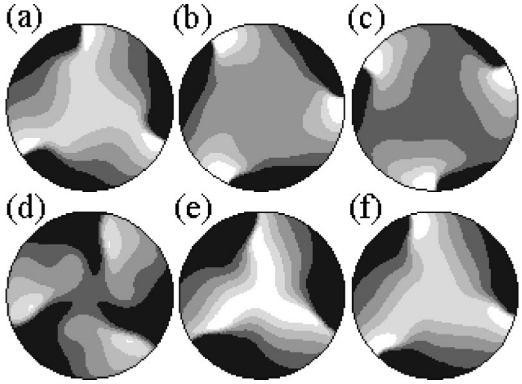

FIG. 4. Unsteady three-spot symmetric spinning wave: $\alpha_{st}\approx 0.97$, Td=0.13, Ar=0.087, $R_0=100$, $\alpha=0.4$. Still frames (a)–(f) illustrate the motion of spots. Near the surface, the spots move counterclockwise.

Note that the front configuration as well as the distribution of θ and η remain unchanged but only rotate when moving downward. Such a wave may be termed as "steady single-spot spinning wave." There also exists a "steady two-spot spinning wave." In this case, two hot spots may appear on the sample surface. The spots (at the ends of the diameter) move alongside the screw trajectory (bifilar helix). The front structure remains unchanged but rotates and propagates downward at a constant velocity. Such waves can be expected to happen for relatively low R_0.

IV. PERIODIC SYMMETRIC SPINNING WAVES

With increasing R_0, the waves tend to become unsteady, periodic, or aperiodic. The symmetric periodic two- or three-spot spinning waves move alongside the sample axis in a pulsating mode, while near the surface the spots move in a spinning mode.

Figure 3 shows two spots that move clockwise. As they move downward, the temperature of the central part goes down, cf. Figs. 3(a) and 3(b). At some moment, a splash appears somewhere around the sample center (c). In this case, a hot spot near the axis becomes ahead of the near-surface ones, so that in Fig. 3(c) θ_{max} at the axis is lower than that for the two near-surface spots. For this reason, we see only one central spot in the cross section of θ_{max} while the two near-surface spots are out of this plane (invisible). Now the temperature in the central zone gradually diminishes due to the heat transfer from the central spot to surrounding material. As θ_{max} at the central spot becomes lower than θ_{max} in the outer spots, the cross section of observation changes its position in space (by definition). The cross section corresponding to (d) is above the cross section corresponding to (c) (see Fig. 1). Upon further motion of spots, we come to picture (e) and (f). Picture (f) is much similar to starting picture (a), that is, we come to the beginning of a new cycle. Because of strong coupling between pulsating motion at the center and spinning motion near the surface, both motions must have an identical mean propagation velocity.

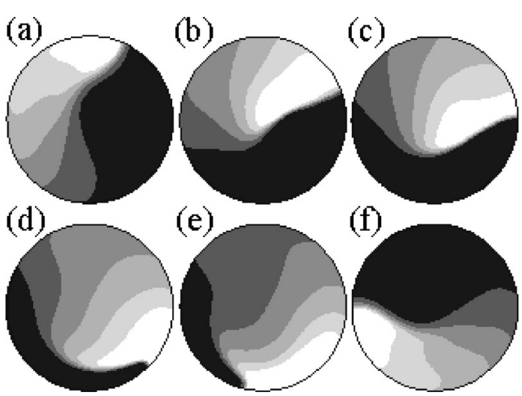

FIG. 5. Unsteady periodic one-spot spinning wave: $\alpha_{st}\approx 0.9$, Td=0.13, Ar=0.1117, $R_0=65$, $\alpha=0.1$. Still frames (a)–(f) illustrate the motion of spots. On the surface, the spot is seen as moving clockwise.

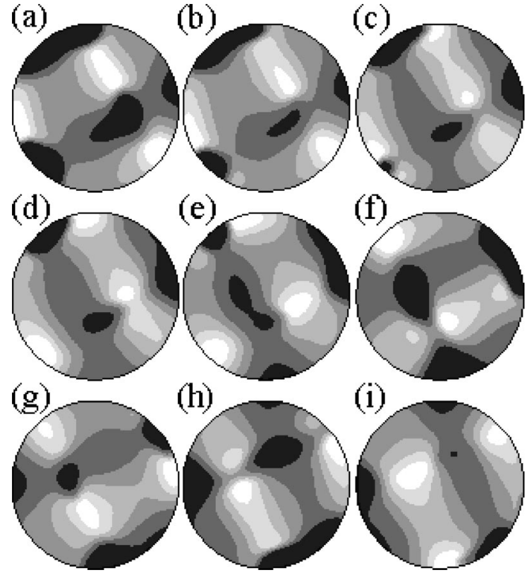

FIG. 6. Unsteady periodic asymmetric three-spot spinning wave: $\alpha_{st}\approx 1.1$, Td=0.16, Ar=0.14, $R_0=80$, $\alpha=1.5$. In this case, the motion of spots on the surface is complicated. The main motion is counterclockwise. But this is also accompanied by clockwise motion of spots and their merging together on the surface.

The same manner of propagation for three spots is presented in Fig. 4(a)–4(f), but here the spots move counterclockwise, and the splash is shown in picture (e).

In both of the above-mentioned cases θ_{max} oscillates in the near-surface layers around some mean value. For this reason, we may talk about "flickering" hot spots that here flicker simultaneously.

V. PERIODIC ASYMMETRIC SPINNING WAVES

With increasing R_0, the one-spot wave undergoes some transformations during a period. The propagation of spin wave was experimentally found[11] to proceed in an unsteady mode. In this case, the inner areas burn out in a pulsation mode, while a flickering spot moves in the near-surface layers. But due to asymmetry, the spot in the near-surface layers markedly varies in its shape and size, see Fig. 5. In frame (a), one can see one small spot near the surface. Then the spot spreads in the radial direction (b) and covers the central zone. At this moment, the splash in the central area can be observed in experiment. The moving central front consumes the reagents (c),(d), so that after this a narrow spot comes to

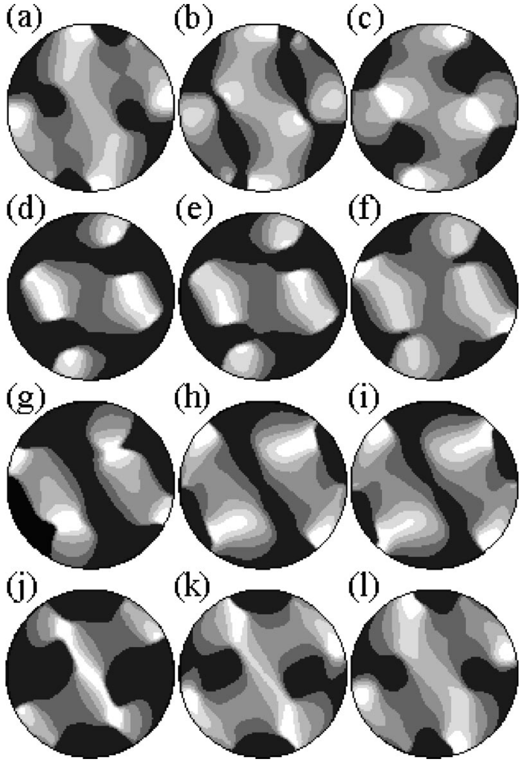

FIG. 7. Unsteady periodic multispot spinning wave: $\alpha_{st}\approx 0.9$, Td=0.13, Ar=0.112, $R_0=100$, $\alpha=0.8$. Still frames (a)–(l) illustrate the motion of spots.

occupy a prolonged circular area (e). Later, the spot again diminishes in its size (because the inner area has already burned out), i.e., a new period of wave motion gets started (f).

Figure 6 shows the motion of spots in the asymmetric three-spot spinning wave. The spots are seen to dive in the depth of the sample, merger together, separate, appear on the surface, etc. Just as in symmetrical cases, the spots near the surface are flickering, but in this case they flicker by turn. They detach from the surface and go inside the cylinder by turns. When the hot spot is at the surface of cylinder, we see bright flash there. Otherwise, the brightness of the spot at the surface decreases.

Note that the merger of two spots (f) and (g) gives birth to two new spots (h) and (i) that move in opposite directions normal to the direction of their collision.

VI. QUASIPERIODIC SPINNING WAVES

Upon further increase in R_0, the wave structure becomes still more complicated. The motion of the spots whose number changes during one period (multispot waves) is illustrated in Fig. 7. According to Ref. 20, several modes of spinning wave may correspond to the same set of system parameters. Besides periodic modes, we also predicted the occurrence of various quasiperiodic modes one of which is shown in Fig. 8. At first glance, this mode of propagation looks like a periodic one. In reality, the neighboring periods are somewhat different due to some fluctuations, so that these were termed ‘‘quasiperiods.’’

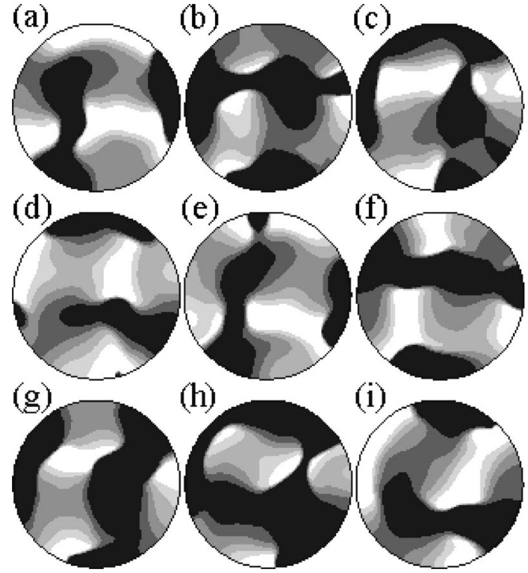

FIG. 8. Quasiperiodic mode: $\alpha_{st}\approx 0.85$, Td=0.115, Ar=0.08, $R_0=105$, $\alpha=0.0$.

VII. APERIODIC SPINNING WAVES

With increasing R_0 and decreasing α_{st}, the number of coexisting propagation modes and fluctuations also increases. Realization of that or another mode depends on the conditions of initiation, this dependence becoming stronger with increasing R_0. By definition,[22] chaotic modes are strongly dependent on the initial conditions. Therefore, we come here to a chaotic (aperiodic) propagation of combustion wave.

Among chaotic modes, of particular interest is the so-called limiting combustion mode[10] that was observed upon addition of increasing amounts of inert diluent (normally end product) into a starting mixture. At the combustion limit, one or several hot spots were found to spread over the surface until formation of a glowing ring. Somewhat later, new spots that appear below the ring were found to form a new ring, etc.

Figure 9 illustrates what is going on in the sample during formation of the ring. A spot formed inside the cylinder (a) is seen to propagate across the sample (b), (c) and increase in

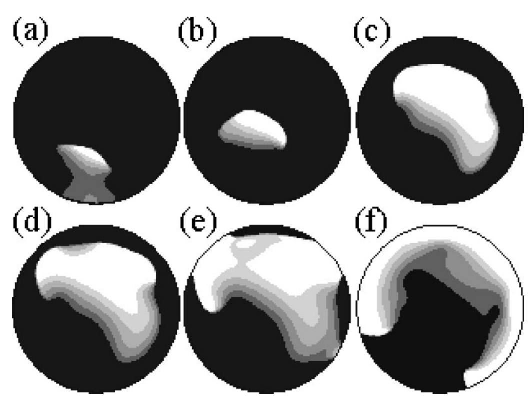

FIG. 9. Transverse motion of spot and formation of ‘‘belt:’’ $\alpha_{st}\approx 0.6$, Td =0.086, Ar=0.072, $R_0=100$, $\alpha=0$.

its size (d). The spot appears at the surface (e) in three areas (although the appearance at the surface may be assumed to proceed otherwise). As a result, a prolonged spot may embrace the cylinder as a ring (f). The next spot is formed inside in the vicinity of meeting point. This mode is not periodic. The size and number of spots may vary with time. The spots may also appear in different cross sections simultaneously. All this allows us to regard this combustion mode as a chaotic one. It is amazing that the mean (over cross section) value of θ and mean longitudinal velocity u are nearly identical to hose calculated in terms of the 1D model.

VIII. EFFECT OF HEAT LOSSES

A solution to the two-dimensional problem[7] implies that, with increasing heat losses, the combustion front becomes crooked, so that the near-surface portion of the combustion front turns out left behind the inner one. In case of three-dimensional problem (when α_{st} and R_0 are sufficiently high), the front shape becomes paraboloid-like. For α_{st} markedly above unity, a steady mode may exist even in case of infinitely large heat losses, that is when $\theta_{surf} = \theta_0$.

With increasing heat losses, the above-given modes retain their basic features, except for stronger bending of front surface. For $\alpha_{st} < 1$, the extinction of combustion takes place at relatively low heat losses. Marked front bending is observed for sufficiently high α_{st} and R_0. Upon an increase in heat losses, the established combustion mode may rearrange into another mode with a lower number of hot spots.

In conditions of strong heat loss, the material on the sample surface between the routes of spots' motion may remain unburned.

IX. STEADY QUASIPARABOLIC WAVES WITH SPIN PERTURBATION OF THE SURFACE OF HEAT LOSS

Under the action of heat losses, the steady modes (when α_{st} is only slightly over unity) may lose its stability. Of particular interest are the modes that take place only in the presence of heat losses. Let us consider such a mode in more detail.

All pictures in Fig. 10 correspond to one and the same moment of time. As follows from Fig. 10(a), the inner portion of the front (core) has a shape close to paraboloid, while in the vicinity of the surface the front is bent. The front core moves alongside the axis at a constant velocity. Figure 10(b) shows the position of spots in the cross section of θ_{max}. In this case, seven spots move counterclockwise. The effect of heat loss manifests itself stronger in the near-surface layers, which results in unsteady propagation of combustion front in these areas. Therefore, the spots are localized in the near-surface layers and do not enter the central zone (a)–(d).

This mode is intermediate between conventional steady modes and spinning modes of front propagation of solid flame and may be termed as the propagation of quasi-paraboloid wave with spin perturbation of the surface of heat loss. The front core is not a strict paraboloid since each spot perturbs the temperature profile, thus giving rise to some bending of the peripheral zones of the core.

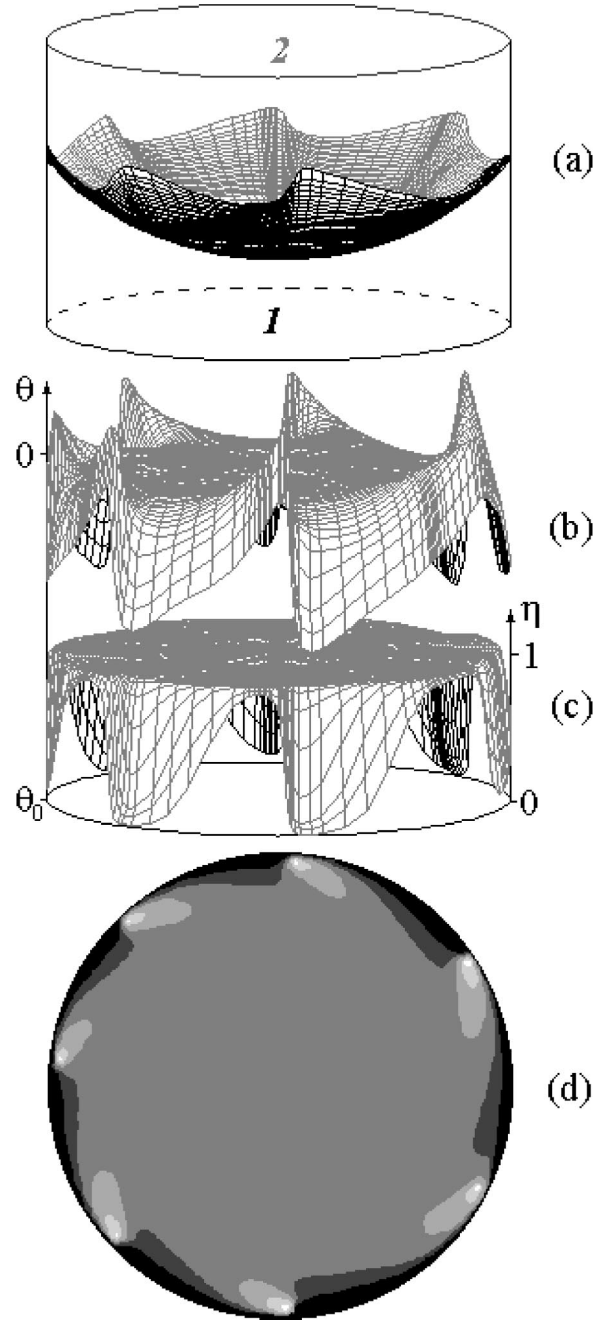

FIG. 10. Steady quasiparabolic spinning wave with spin perturbation of the surface of heat loss: $\alpha_{st} \approx 1.1$, Td=0.16, Ar=0.14, $R_0 = 200$, $\alpha = 6.5$. Wave front structure (a). Spatial distribution of (b) θ and (c) η in the cross section of θ_{max}. The distribution of θ (d) in the form of isotherms.

The existence of such a wave is due to the presence of heat losses. As already mentioned, the planar steady wave loses its stability at $\alpha_{st} = 1$. In our case, $\alpha_{st} > 1$ but the conditions are nonadiabatic. With increasing heat losses, θ_b diminishes and, accordingly, α_{st} goes down. The effect is strongest in the near-surface layers. In the case of low heat losses (when $\alpha_{st} > 1$), the front is crooked but remains to be steady. In case of strong heat loss, θ_b may decrease to such an extent that the front propagation becomes unsteady even alongside the sample axis (i.e., $\alpha_{st} < 1$ at the sample axis). In between these cases, intermediate modes are possible for which $\alpha_{st} > 1$ at the axis but $\alpha_{st} < 1$ in the near-surface areas.

It is important that the axial propagation velocity u be identical to the mean velocity of front motion alongside the surface. An increase in R_0 (at the heat loss that ensures an increase in θ at the sample center) may promote a change in the combustion mode, namely, front stabilization in the near-surface layers. Indeed, the velocity of axial front propagation is defined by the value of $\theta \approx 0$. If the near-surface layers (affected by intense heat loss to the environment) cannot burn in the spinning mode at the same velocity, the reaction rate in these layers will be defined by heat flux from the inner areas, which ensures a constant front propagation velocity in the near-surface layers. Conversely, a decrease in R_0 may result in the extension of instability inward, which may result in establishing nearly adiabatic modes.[20] For given R_0 and α_{st}, a decrease in heat losses manifests itself just as a increase in R_0, and *vice versa*.

In addition, our data also imply the following.

(1) Figure 10 shows a combustion mode with seven hot spots moving counter-clockwise. However, their number and direction of moving may be different, depending on system parameters and conditions of ignition.

(2) The spots in the near-surface layers may occur at different height (although within a narrow disk) and flicker by turns. As a result, the period may become very long.

(3) Just as in adiabatic conditions,[20] several combustion modes at the same set of system parameters (nonuniqueness of solution) may also be expected to exist in the presence of heat loss from the sample surface.

X. CONCLUSIONS

Besides a planar (adiabatic conditions) or axisymmetric (in the presence of heat loss from the surface) mode, a combustion wave in a cylindrical sample may also propagate in a spinning mode. With increasing radius, the spatiotemporal structure of the spinning combustion wave becomes more and more complicated. Transition to chaos proceeds due to increasing influence of initial conditions and appearance of the quasi-periodic modes.

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