STRUCTURES IN IR-LASER GAS-MIXTURE HEATING

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INTRODUCTION

Here we consider some aspects of laser gas-mixture heating. The system exchanges energy with the environment and is thus dissipative. Various types of self-organization phenomenon occur in a nonlinear dissipative system, where stationary and nonstationary structures arise [1,2]. The structures have fairly extensive attraction regions (in terms of the initial conditions) and may be considered as asymptotically stable proper states (autostructures).

IR laser kinetics [3] can provide scope for simulating these phenomena, since one can vary the beam parameters to control the type of nonlinearity, which is a feature usually representing some difficulty for other types of system, and which substantially restricts the types of structure that can be observed.

Here we consider two types of structure occurring on IR irradiation: self-oscillatory and soliton-type ones. These correspond to nontrivial self-organization types and clearly demonstrate the wide scope for controlling system dynamics.

1. GAS-MIXTURE OSCILLATIONS

Experiments have been reported [4,5] on the transmission of a continuous-wave CO_2-laser beam through a mixture of active and inert gases (SF_6 + NH_3, SF_6 + air, and so on). There is an incident-beam parameter range where one gets oscillatory power levels, where the characteristic oscillation times (10-100 sec) [4,5] indicate transport by diffusion, not by gas-dynamic processes. These oscillations have been ascribed [4-7] to thermal diffusion in the inhomogeneously heated gas, but there are alternative explanations, in particular due to local density change produced by temperature gradients.

Consider the heating in an inert-gas mixture in a cell fairly thin along the beam axis. Let the beam be absorbed by one component. The heating in the beam region reduces the density by thermal expansion and thus reduces the amount of absorbing component, so the gas cools and the density rises again; the process then repeats. By analogy with [6,7], one can show that oscillations require a fairly high absorptivity temperature dependence, which is characteristic of many molecular gases in the IR range [8,9], and sometimes one has

$$A(n, T) = \beta n N \exp(-T_*/T), \quad \beta, T_* = \text{const}, \tag{1}$$

where N is density and n absorbing-component concentration. Let the radiation pass along the Y axis to a long cell (along the $X \perp Y$ axis) that is thin along the Y axis and contains an inert two-component mixture. The temperature T and concentration n are described by

$$\frac{1}{a}\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} + \frac{1}{\varkappa h}[A(n,T)l(x) - \eta(T - T_*)], \quad T|_{t=0} = T_*,$$
$$\frac{1}{D}\frac{\partial (nN)}{\partial t} = \frac{\partial}{\partial x}\left(N\frac{\partial n}{\partial x}\right), \quad n|_{t=0} = n_*; \quad -R < x < R. \tag{2}$$

Here a, κ, and D are the thermal diffusivity, thermal conductivity, and diffusion coefficient, while h is the cell thickness. The $[-\eta(T-T_*)]$ term describes the heat exchange

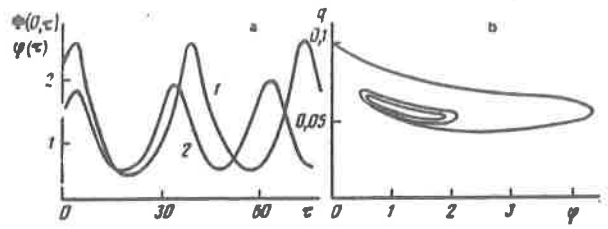

Fig. 1. a) Time dependence of gas temperature at the beam axis derived from the exact system (curve 1) and the approximate one (curve 2); b) phase pattern for approximate system (8) for the case of Fig. 1a. Parameter values $\nu=3$; $b=1$; $j=2{,}805$; $n_0=0{,}1$; $\gamma=4{,}95$; $R/x_0=5$.

with the environment, which has temperature T_e. The intensity distribution I(x) is

$$I(x)=I_0\exp(-x^2/x_0^2). \tag{3}$$

The local N after fast gas-dynamic relaxation is related to the local temperature by

$$N=N_h\frac{T_e}{T},\quad \frac{1}{T_e}=\frac{1}{2R}\int_{-R}^{R}\frac{dx}{T} \tag{4}$$

(N_h is the gas density in the homogeneous state). This follows from P = NkT, in which one takes P as constant throughout the cell.

(2) neglects thermal diffusion and the diffusion-based thermal effect, which is applicable for many gas mixtures [10], although the converse situation can occur [11].

We introduce the symbols

$$\Phi=(T-T_e)/T_e,\quad Q=nN/N_h,\quad \tau=t/t_0,\quad \xi=x/x_0,\quad \gamma=T_d/T_e,$$
$$b=\eta x_0^2/\varkappa h,\quad \nu=a/D,\quad j=(N_e\beta I_d/\eta T_e)e^{-\gamma},\quad t_0=x_0^2/a. \tag{5}$$

Then (1)-(4) becomes

$$\frac{\partial\Phi}{\partial\tau}=\frac{\partial^2\Phi}{\partial\xi^2}+b\left[j\exp(-\xi^2)\exp\left(\frac{\gamma\Phi}{\Phi+1}\right)Q-\Phi\right],\quad \Phi|_{\tau=0}=0,$$
$$\nu\frac{\partial Q}{\partial\tau}=\frac{\partial^2 Q}{\partial\xi^2}+\frac{\partial}{\partial\xi}\left(\frac{Q}{\Phi+1}\frac{\partial\Phi}{\partial\xi}\right),\quad Q|_{\tau=0}=n_0. \tag{6}$$

We supplement (6) with the boundary conditions corresponding to heat and mass insulation at the cell boundaries: $\Phi_\xi=Q_\xi=0$ at $\xi=\pm R/x_0$. These conditions provide for conservation of the total amount of gas in the cell. In what follows, we are interested in the limit $R\to\infty$.

There is a notable feature. The above implies that the spatial inhomogeneity in the radiation field is important for oscillations to arise. It can be shown by analogy with [6] that homogeneous stationary states are stable and that oscillations will not be excited for $x_0\to\infty$, i.e., for a homogeneous intensity distribution throughout the cell length.

It is convenient to reduce (6) to a system of ordinary differential equations for analytic examination. We use a form of the Bubnov-Galerkin method [12] as employed in [6]: we introduce the Galerkin coordinates $\phi(\tau)$ and $q(\tau)$ from

$$\Phi(\xi,\tau)\approx\varphi(\tau)\exp(-\xi^2),$$
$$Q(\xi,\tau)=n_0+[q(\tau)-n_0]\exp(\xi^2). \tag{7}$$

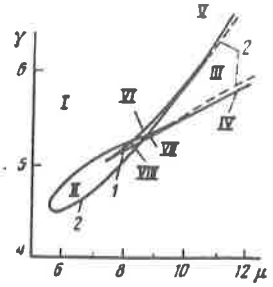

Fig. 2

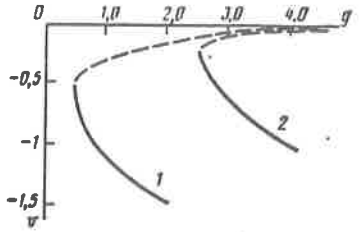

Fig. 3

Fig. 2. Division of the {μ, γ} parameter plane by curves representing the boundaries for the saddle points (1) and the stability boundaries for points not of saddle type (2). The dashed lines represent continuation of curve 2 on the saddle point surface, ν(b + 1) = 10.

Fig. 3. Soliton velocity v as a function of radiation power g derived from (25) for α of 1 (1) and 0.1 (2). The solid lines correspond to the stable solution and the dashed ones to the unstable one.

The coordinates {ϕ, q} describe the gas temperature and the absorbing-component density at the axis (ξ = 0). One can simplify (7) further by introducing more complete sets $\{\phi_i, q_i\}$, e.g., as coefficients in the expansion of Φ and Q in terms of Hermitian polynomials:

$$\Phi(\xi,\tau)=\exp(-\xi^2)\sum_i \varphi_i(\tau)H_i(\xi),$$
$$Q(\xi,\tau)=n_0+\exp(-\xi^2)\sum_i [q_i(\tau)-n_0]H_i(\xi). \qquad (7')$$

To obtain the required system, we restrict ourselves to the (7) approximation. We substitute (7) into (6) and multiply both equations in (6) term by term by $\exp(-\xi^2)$ and integrate with respect to ξ with limits $-\infty$ to $+\infty$ to get

$$\frac{d\varphi}{d\tau}=-(b+1)\varphi+bjq\exp\left(\frac{\gamma\varphi}{\varphi+1}\right), \quad \nu\frac{dq}{d\tau}=\frac{2+\varphi}{2(1+\varphi)}n_0-\frac{2+3\varphi}{2(1+\varphi)}q. \qquad (8)$$

When the solutions to the exact system (6) and the approximate one (8) are compared, it is found that the simplified model gives a good qualitative description.

Figure 1a shows ϕ(τ) and Φ(0, τ) for identical parameter values derived by numerical solution from (8) and (6) correspondingly, and Fig. 1b shows the (8) phase pattern for the same parameter values.

We examined (8) by standard methods from nonlinear-oscillation theory [13,14]; Fig. 2 shows a typical subdivision of the {μ, γ} parameter plane into subregions differing in the number and stability of the singular points. The existence boundary for the saddle points (curve 1) is given in parametric form by

$$\mu=\frac{b+1}{bjn_0}=\frac{1-z}{z}\frac{2-z}{2+z}\exp(\gamma z), \quad \gamma=\left(1+\frac{4z(1-z)}{4-z^2}\right)\frac{1}{z(1-z)}, \qquad (9)$$

while the stability limits for points not of saddle type (curve 2) are given by

$$\mu=\frac{1-z}{z}\frac{2-z}{2+z}\exp(\gamma z), \quad \gamma=\left(1+\frac{2+z}{2\nu(b+1)}\right)\frac{1}{z(1-z)} \qquad (10)$$

(z = ϕ/(η + 1) is a parameter). Table 1 classifies the singular points corresponding to the various subregions in Fig. 2.

Table 1

Region number in Fig. 2	Singular points		
	lower	saddle	upper
I	Coincides with upper	Absent	Stable
II	»	»	Unstable
III	Stable	$p_1+p_2>0$	Stable
IV	»	$p_1+p_2<0$	»
V	»	$p_1+p_2<0$	»
VI	Unstable	$p_1+p_2>0$	»
VII	Stable	$p_1+p_2>0$	Unstable
VIII	Unstable	$p_1+p_2>0$	»

Note. 1. The upper and lower singular points denote the high-temperature and low-temperature stationary solutions to (8). 2. For a saddle point (which is always unstable), we give the sum of the characteristic parameters p_1 and p_2, which are defined as follows: if $\varphi^{(0)}$ and $q^{(0)}$ represent a a certain stationary solution, one can linearize (8) with respect to the small deviations $\Delta\varphi = \varphi - \varphi^{(0)}$, $\Delta q = q - q^{(0)}$ and put $(\Delta\varphi, \Delta q) \sim \exp(pr)$, to get a characteristic equation from which p_1 and p_2 are derived.

Oscillation requires $\gamma > 4$ and $\nu(b+1) > \nu_* \approx 4.8$. Then as ν and b increase (i.e., the beam radius increases, since $b \sim x_0^2$), the oscillation region expands.

Finally, without entering into details, we estimate the oscillation period in various limiting cases:

$$\tau \approx 5{,}5\nu \quad \text{or} \quad t \approx 5{,}5x_0^2/D. \tag{11}$$

Typical values are $D \simeq 0.1$ cm$^2 \cdot$sec^{-1} and $x_0 \simeq 0.5$ cm, which give $t \simeq 15$ sec.

2. CHEMICAL SOLITON

Another type of dissipative structure is a traveling pulse, which we consider for IR heating in a mixture showing a reversible reaction. Such a gas on laser heating at a suitable wavelength can show bistability [15,16], i.e., in a certain power range ($W_1 < W < W_2$), there are two stable equilibrium states that differ in temperature and component concentration. The phenomenon has been examined in detail for an ideal-mixing reactor, for the case where the temperature and concentration gradients are negligible and the radiation attenuation on passage through the gas can be neglected.

Numerical simulation has been used [17] to examine the behavior of such a mixture on the basis of Bouguer attenuation but without allowance for heat and mass transfer. If the bistability conditions are met, the reaction is effectively localized in a bounded region $0 \leq x \leq x_k$ ($x = 0$ is the radiation entry point). Here we consider a similar system with allowance for thermal conduction and Bouguer absorption.

The reaction $B_1 \rightleftharpoons B_2$ occurs in a long thin cell illuminated from the end with power $W \in (W_1, W_2)$. The concentration n of substance B_2 is defined by a kinetic equation that incorporates diffusion:

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial x^2} + k_1(1-n)\exp\left(-\frac{T_1}{T}\right) - k_2 n \exp\left(-\frac{T_2}{T}\right). \tag{12}$$

We assume that $T_1 > T_2$. In the homogeneous stationary state ($n_t = n_{xx} = 0$), n is

$$n = \left[1 + \frac{k_2}{k_1}\exp\left(\frac{T_1 - T_2}{T}\right)\right]^{-1} = n(T), \tag{13}$$

where n(T) increases from 0 to $(1 + k_2/k_1)^{-1}$ as T increases. The radiation is absorbed only by B_2. Then the temperature is given by the conduction equation

$$\frac{1}{a}\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} - \frac{1}{\varkappa}\frac{\partial I}{\partial x} - \frac{\eta_1}{\varkappa}(T - T_*), \tag{14}$$

where η_1 is the heat-loss constant per unit cell length. The intensity diminishes in accordance with Bouguer's law:

$$\frac{\partial I}{\partial x} = -\beta n I, \quad \beta = \text{const.} \tag{15}$$

The phenomenon is as follows. At the start, the system is in the lower concentration and temperature state. If the medium is transferred to the upper state in some finite region, the absorption there increases and the gas begins to heat up. Adjacent regions also begin to be heated by thermal conduction and absorption. The darkening zone will enlarge until the radiation power after it becomes less than W_1, the minimum necessary for bistability. The zone begins to move in the opposite direction to the beam, since conduction heats the region before it, which increases the absorption, while the region after it cools by heat transfer to the environment and thermal conduction. One thus gets a chemical soliton, namely a localized region in which the medium is in the upper (excited) state.

This effect occurs because of energy exchange with the environment (i.e., in a dissipative system) and thus differs from a soliton in a nonlinear conservative system; the parameters such as speed, width, and amplitude are independent of the initial conditions to a considerable extent. On the existing terminology [18], such a phenomenon is called a traveling pulse.

To simplify the analysis, we now take subsequently that $T_e = 0$, with the cell taken as long enough, where we introduce the symbols

$$\tau = \frac{\eta_1 a}{\varkappa} t, \quad \xi = \sqrt{\frac{\eta_1}{\varkappa}}\, x, \quad \Phi = \frac{T}{T_1 - T_2}, \quad j = \frac{I}{(T_1 - T_2)/l\varkappa\eta_1},$$
$$\nu = \frac{a}{D}, \quad \alpha = \sqrt{\frac{\varkappa}{\eta_1}}\,\beta, \quad k = \frac{k_2}{k_1}. \tag{16}$$

Then the initial equations become

$$\frac{\partial \Phi}{\partial \tau} = \frac{\partial^2 \Phi}{\partial \xi^2} - \frac{\partial j}{\partial \xi} - \Phi, \quad \frac{\partial j}{\partial \xi} = -\alpha n j,$$
$$\frac{\partial n}{\partial \tau} = \frac{1}{\nu}\frac{\partial^2 n}{\partial \xi^2} + K(\Phi)[n(\Phi) - n]; \quad n(\Phi) = \left[1 + k\exp\left(\frac{1}{\Phi}\right)\right]^{-1}, \tag{17}$$
$$K(\Phi) = \frac{\varkappa k_1}{\eta_1 a}\left[\exp\left(-\frac{T_1}{T_1 - T_2}\frac{1}{\Phi}\right) + k\exp\left(-\frac{T_2}{T_1 - T_2}\frac{1}{\Phi}\right)\right].$$

Thermal conduction and heat loss are the principal factors resulting in the soliton. Diffusion merely smears the soliton, while an inadequate reaction rate reduces the speed and restricts the parameter existence range. Therefore, we assume that the diffusion is sufficiently slow ($\nu \gg 1$) and that the reactions are so fast that $n = n(\Phi)$ at any instant. Then

$$\frac{\partial \Phi}{\partial \tau} = \frac{\partial^2 \Phi}{\partial \xi^2} - \frac{\partial j}{\partial \xi} - \Phi,$$
$$\frac{\partial j}{\partial \xi} = -\alpha n(\Phi) j, \quad n(\Phi) = [1 + k\exp(1/\Phi)]^{-1}. \tag{18}$$

We transfer to a frame of reference moving uniformly with velocity v, i.e., we introduce a coordinate $y = \xi - v\tau$, and consider localized solutions stationary in that system, which are defined by the boundary-value problem

$$\Phi_{yy} + v\Phi_y - \Phi - j_y = 0,$$
$$j_y = -\alpha n(\Phi) j; \quad \begin{matrix} \text{for } y \to \pm\infty & \Phi_y \to 0, \\ \text{for } y \to -\infty & j \to j_0, \end{matrix} \tag{19}$$

where j_0 is the dimensionless radiation power at the input.

Two comments must be made before we calculate the soliton parameters.

1. The homogeneous state having $\Phi = 0$ is stable under small perturbations. One linearizes (18) near $\Phi = 0$ and puts $\Phi(\xi, \tau)=\Phi_1 \exp(p\tau+is\xi)$ to get $p = -s^2 - 1 < 0$, i.e., small perturbations are always damped, and soliton generation requires a large initial perturbation. One can obtain a constraint on such excitation by integrating the first equation in (18) with respect to ξ and using the second:

$$F_\tau=[j_0-j(+\infty)]-F, \quad j(+\infty)=j_0\exp(-\alpha N);$$
$$F(\tau)=\int_{-\infty}^{+\infty}\Phi(\xi,\tau)d\xi, \quad N(\tau)=\int_{-\infty}^{+\infty}n(\xi,\tau)d\xi. \tag{20}$$

Then if initially $F(0)<j_0[1-\exp(-\alpha N(0))]$, $F_\tau > 0$ and the perturbations increase.

2. There is a radiation-power soliton-generation threshold, which we estimate by integrating (19) with respect to y on the basis that $\Phi \to 0$ for $y \to \pm\infty$. Then $j_0-j(+\infty)=F \equiv \int_{-\infty}^{+\infty}\Phi(y)dy$, where $j(+\infty)$ is defined by a formula analogous to (20), and

$$j_0=F[1-\exp(-\alpha N)]^{-1}. \tag{21}$$

The bound for integral N is

$$N=\int_{-\infty}^{+\infty}n(y)dy=\int_{-\infty}^{+\infty}\frac{n(y)}{\Phi(y)}\Phi(y)dy\leqslant\left(\frac{n}{\Phi}\right)_{\max}F,$$

which gives

$$j_0>F\left[1-\exp\left(-\alpha F\left(\frac{n}{\Phi}\right)_{\max}\right)\right]^{-1}. \tag{22}$$

The right-hand side in (22) is a monotone function of F that attains its minimum value $\left[\alpha\left(\frac{n}{\Phi}\right)_{\max}\right]^{-1}$ for $F = 0$, so (22) becomes

$$j_0\alpha\left(\frac{n}{\Phi}\right)_{\max}>1. \tag{23}$$

That condition has a simple meaning: when (23) is obeyed, the heat-loss curve $w_l = \Phi$ intersects the heat-input curve $w_{abs}=\alpha n(\Phi)j_0$ at three points, i.e., bistability occurs for $j = j_0$. We consider the maximum in $\Phi(y)$ ($\Phi_y=0$, $\Phi_{yy}<0$) to get a similar condition from (19): $(\alpha j n(\Phi)/\Phi)|_{\Phi=\Phi_m} > 1$.

We now derive the soliton speed and characteristic width, where we approximate $n(\Phi)$ as a step function:

$$n(\Phi)=\begin{cases}0, & \Phi<\Phi_n\\ 1, & \Phi>\Phi_n\end{cases}$$

where Φ_n is the point of inflection. This applies for $k \ll 1$. Then if $\Phi > \Phi_n$ for $0 \leq y < L$, instead of (19) we have

$$\Phi_{yy}+v\Phi_y-\Phi=0, \quad y<0 \text{ and } y>L,$$
$$\Phi_{yy}+v\Phi_y-\Phi=-\alpha j_0\exp(-\alpha y), \quad 0<y<L, \tag{24}$$

whose solutions are linked up from the conditions for continuity in the temperature Φ and heat flux Φ_y at the points $y = 0$ and $y = 1$ (at which $\Phi = \Phi_n$). Also, we select solutions such that $\Phi \to 0$ for $y \to \pm\infty$, which gives us a transcendental equation for the soliton's speed:

$$M^n(1-M^r)=(1-M)r,$$

in which

$$M=1-\frac{(p_+ +\alpha)(p_+ -p_-)}{\alpha g},\quad r=\frac{p_- +\alpha}{p_- +\alpha},\quad g=\frac{j_0}{\Phi_n},$$
$$m=-\frac{p_-}{p_+ +\alpha},\quad p_\pm=\frac{-v\pm\sqrt{v^2+4}}{2}. \qquad (25)$$

Figure 3 shows v(g) derived from (25) for two characteristic values of α. Out of the two branches of v(g) (for each α), one selects the physical one (solid lines), which corresponds to the stable state. The second solution (dashed lines) is unstable: any deviation from it leads to establishment either of the homogeneous state or a soliton. It is evident that v is always negative, so the soliton moves in the opposite sense to the laser beam. The solutions exist only for radiation power levels exceeding a critical value (which is dependent in particular on α). (25) readily gives the asymptotic v(g): $v\approx-\sqrt{\alpha g}$ for $g\to\infty$, or in dimensional form

$$V\approx-a\sqrt{\beta l_\varphi \varkappa T_n}. \qquad (26)$$

Finally, from (24) one gets an expression for the soliton's width L: $L=(\ln M)/(p_+ +\alpha)$, or for large g

$$L\approx-v\ln(-v/g)\approx\sqrt{\alpha g}\ln\sqrt{g/\alpha}. \qquad (27)$$

The soliton corresponding to the low-speed branch (dashed lines in Fig. 3) has speed v_1 and width L_1 as follows:

$$v_1\approx-2/3g,\quad L_1\approx 2/g\alpha,$$

i.e., the slow soliton is also narrow, which determines the sensitivity to various fluctuations and hence the instability.

Numerical estimates are as follows for typical values of the constants: $T_n \approx 400$ K, $\eta_1 \approx 2\cdot10^{-3}$ W·cm^{-3}·K^{-1}, $a^* \approx 1$ cm^2·sec^{-1}, $\varkappa \approx 2\cdot10^{-4}$ W·cm^{-1}·K^{-1}, and $\varkappa \approx 1$ cm^{-1}, where (23) with n = 1 and $\Phi = \Phi_n$ gives $I \gtrsim 0.2$ W·cm^{-2}. With $I \approx 10$ W·cm^{-2}, (26) gives $V \approx 10$ cm·sec^{-1}, while (27) after conversion to dimensional quantities gives $L \approx 7$ cm.

We have considered the situation where bistability occurs because of a reversible reaction, but analogous results are obtained for a chemically inert system whose absorptivity varies rapidly with temperature. Similar equations apply also to an ionization wave produced by a laser beam (see reviews [19,20]), for which also there are characteristic states of optical-discharge propagation of slow combustion type.

CONCLUSIONS

These results show that IR radiation can produce various nontrivial structures in a gas: oscillatory, soliton-type, and so on. An important common point here is that there is spatial inhomogeneity in the laser beam. In the first example, the inhomogeneity is due to the finite diameter, and in the second to the attenuation on passage through the medium. The inhomogeneity leads to qualitatively new structures lacking with homogeneously distributed intensity. From the formal viewpoint, when we introduce external inhomogeneity, we deprive the system of certain symmetries and thus life the degeneracy, which extends the possible types of solution, although the number of phase variables is unaltered.

In nonlinear dissipative-system analysis, one frequently distinguishes [2] major variables (order parameters) and minor ones (subordinate). The first-group variables determine the main behavior. One can thus say that introducing inhomogeneity increases the number of order parameters (major degrees of freedom).

As spatial inhomogeneity is characteristic of many systems, research on this class is very important to elucidating nonlinear-system self-organization.

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