Stochastic regime of thermodynamic, heterogeneous processes in a laser-radiation field

V. A. Bobyrev, F. V. Bunkin, N. A. Kirichenko, B. S. Luk'yanchuk, and
A. V. Simakin
P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted 2 October 1980)

Pis'ma Zh. Eksp. Teor. Fiz. **32**, No. 10, 608–611 (20 November 1980)

An experimental observation of a new type of quasi-steady-state regime in nonisothermal, heterogeneous, stochastic-combustion reactions due to the action of continuous laser radiation is reported. It is shown that combustion stochastization, which occurs as a result of sharp focusing of the radiation on the target surface, is caused by new degrees of freedom in the investigated nonlinear system, which are associated with the spatial inhomogeneity of the problem.

PACS numbers: 82.40.Py, 82.65.Jv, 82.60. – s

Thermochemical processes induced by laser radiation have been intensively studied in recent years. These processes include laser ignition and combustion of metals in an oxidizing medium.[1] In the laser ignition process the rate of the oxidizing reaction, its energy release, and the absorptivity of the metal surface with an increasing oxide layer vary in an interrelated manner. The complex thermal dynamics of such processes is governed by the positive feedback, which results in thermochemical instability,[2] and by the unique variation of the absorption characteristics of the surface.[2-4]

The results of studies of the laser combustion of metals show that the development process of thermochemical instability culminates in the establishment of a steady-state regime in the system (or self-oscillations). Such effects are also characteristic of several other nonequilibrium systems, in which chemical reactions occur (see, for example, Refs. 5 and 6).

We report in this paper the results of an experimentally observed new type of quasi-steady-state regime in heterogeneous reactions–stochastic laser combustion. In the theory of nonlinear oscillations such movement of a dynamic system in the phase plane corresponds to the so-called "strange attractor."[7]

We have experimentally investigated the heating dynamics of titanium targets by a CW CO_2 laser with a power $P = 18$ W. The thickness of the cylindrical targets varied from 0.5 to 1.5 mm, their diameter was in the range of 2 to 5 mm, and their mass in the range of 30 to 100 mg. The experiments were performed using the method described in Ref. 2. The target temperature $T(t)$ was measured with a chromel-alumel thermocouple, which was welded to the back side of the target. The value of $T(t)$ and its derivative dT/dt (at the output of a differentiating RC network with a time constant $\tau = 0.02$ sec) were recorded by means of an N-115 loop oscillograph.

Typical heating and combustion oscillograms for sufficiently uniform illumination of the target are shown in Fig. 1. The characteristics of titanium ignition and

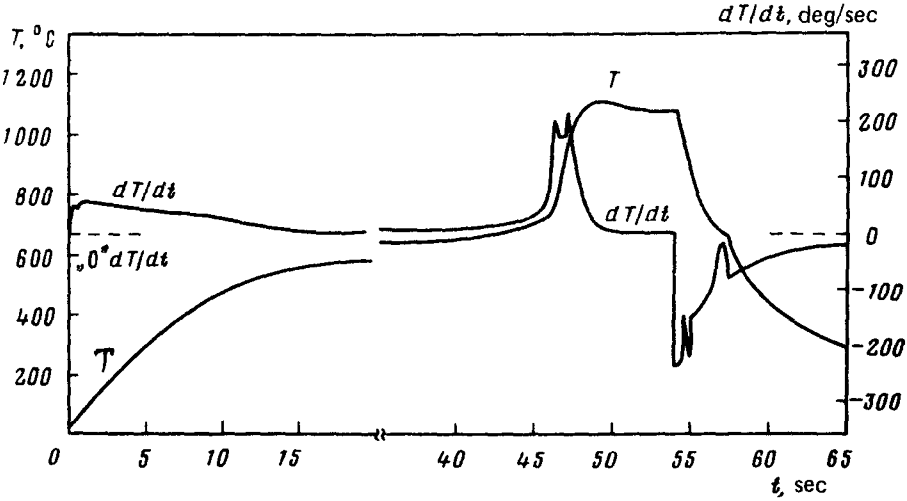

FIG. 1. Experimental oscillogram of $T(t)$ and dT/dt signals obtained during ignition of a titanium target with a diameter of 4 mm and mass $m = 43.1$ mg as a result of uniform illumination of the surface by laser radiation ($P = 18$ W). The laser was turned off at a time $t = 54$ sec.

combustion under such conditions were investigated in Refs. 3, 4, and 8. It is important that the steady-state regime in Fig. 1 and in all similar experiments was quickly established after the ignition (in Fig. 1 $dT/dt \approx 0$ for $t > 50$ sec). In addition, we established experimentally that the target absorptivity quickly reaches its asymptotic value, which is determined by the optical constants of the oxide, and that almost no evaporation of the oxide film occurs at these temperatures.

As the size of the focal spot, which was much smaller than the target radius, decreased, we observed a transition to a regime in which the heating rate varied randomly (the derivative dT/dt fluctuated randomly) (see Fig. 2). This heating regime was observed in a rather broad range of target parameters. A preliminary spectral and correlation analysis of the $dT(t)/dt$ dependence confirms its stochastic nature.

According to the theory of nonlinear oscillations,[7] a system must have more than two degrees of freedom in order for the stochasticity to occur. It follows from this that there should be no stochasticity when a thermally thin target is uniformly illuminated with a laser radiation, since the heating of such a target is accurately described by a system of two, nonlinear, differential equations

$$mc\frac{dT}{dt} = PA(x) - P_{\text{loss}}(T) + \rho s w \frac{dx}{dt} \tag{1}$$

$$\frac{dx}{dt} = \frac{d}{x}\exp\left(-\frac{T_d}{T}\right), \quad T\big|_{t=0} = T_{\text{init}}, \quad x\big|_{t=0} = x_{\text{init}}.$$

Here x is the thickness of the oxide film, $A(x)$ is the absorptivity of the target,[2] $P_{\text{loss}}(T)$ is the heat loss rate, m is the mass, ρ is the density, c is the specific heat, s is

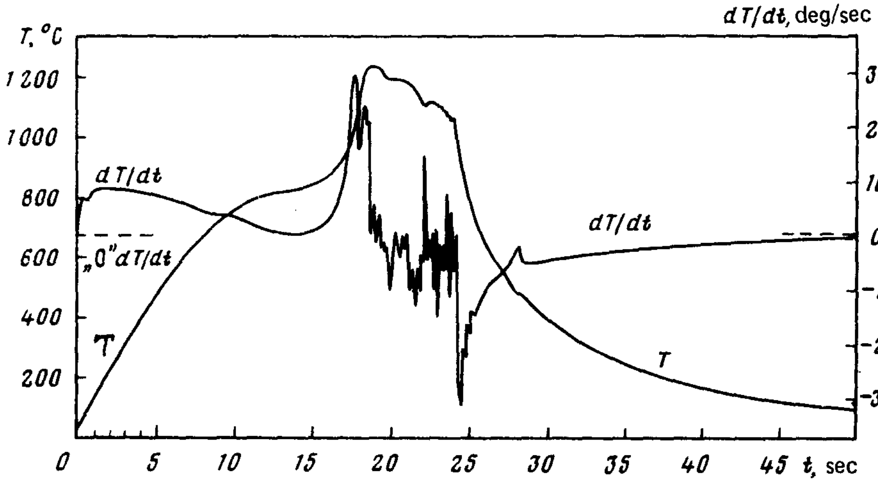

FIG. 2. Experimental oscillogram of $T(t)$ and dT/dt signals obtained by sharply focusing the continuous radiation ($P = 18$ W) on the target. The target parameters are: diameter 4 mm and mass $m = 50.5$ mg. The laser was turned off at $t = 24$ sec.

the total surface area of the traget, w is the specific heat of the reaction, and d and T_d are the oxidation-law constants. The radial heat conductivity plays an important role when the radiation is sharply focused. Direct measurements showed that the temperature difference between the center and the periphery of the target is $\Delta T = 50°\text{-}250°\text{C}$ in our experiments. Accordingly, when the radiation is sharply focused we must write the complete heat-conductivity equation instead of the heat-balance equation [the first equation in (1)]. The problem, therefore, becomes nonlocalized, which means that the system has more than two degrees of freedom. These degrees of freedom can be explicitly isolated by approximately reducing the problem to a finite number of ordinary differential equations. In the simplest case, the problem can be reduced to a system of equations for the temperatures and thicknesses of the oxide layers (or the energy release of the reaction) at the center and periphery of the target. Qualitatively, the stochasticity can be attributed to a strong, nonlinear, temperature dependence of the energy release and to the heat-conductivity delay. This delay $\tau_T \sim R^2/a$ (R is the radius and a is the thermal diffusivity coefficient of the target) is $\tau_T \sim 0.1\text{–}0.3$ sec, in good agreement with the oscillation period of dT/dt in our experiments. A more thorough, quantitative analysis of the experiments with a detailed numerical integration of the heating-dynamics equations will be carried out later. Here we note, however, that the problem discussed above gives an important example of a relatively simple physical system in which stochastic oscillations appear.

Thus, we have experimentally observed for the first time a stochastic regime of thermochemical reactions produced as a result of the action of laser radiation. Although this effect has been observed in heterogeneous processes, it is easy to see that such effects can also occur as a result of laser stimulation of homogeneous reactions.

The authors thank Yu. A. Kravtsov, A. M. Prokhorov, and M. I. Rabinovich for useful discussions.

1. M. I. Arzuov, A. I. Barchukov, F. V. Bunkin, V. I. Konov, and B. S. Luk'yanchuk, Kvantovaya Elektron. 6, 1339 (1979) [Sov. J. Quantum Electron. 9, 787 (1979)].
2. M. I. Arzuov, A. I. Barchukov, F. V. Bunkin, N. A. Kirichenko, V. I. Konov, and B. S. Luk'yanchuk, Kvantovaya Elektron. 6, 466 (1979) [Sov. J. Quantum Electron. 9, 281 (1979)].
3. A. M. Bonch-Bruevich, M. N. Libenson, V. S. Makin, S. D. Pudkov, I. I. Ivanova, and M. K. Kochengina, Pis'ma Zh. Tekh. Fiz. 4, 921 (1978) [Sov. Tech. Phys. Lett. 4, 371 (1978)].
4. V. I. Boĭko, F. V. Bunkin, N. A. Kirichenko, and B. S. Luk'yanchuk, Dokl. Akad. Nauk SSSR 250, 78 (1980) [Sov. Phys. Dokl. 23, 38 (1980)].
5. K. G. Shkadinskiĭ, B. I. Khaĭkin, and A. G. Merzhanov, Preprint IKhF Akad. Nauk SSSR, Moscow, 1970.
6. V. V. Vol'ter and I. E. Sal'nikov, *Ustoĭchivost' rezhimov raboty khimicheskikh reaktorov* (Stability of Chemical Reactor Operating Regimes), Khimiya, Moscow, 1972.
7. A. V. Gaponov-Grekhov and M. I. Rabinovich, Usp. Fiz. Nauk 128, 579 (1979) [Sov. Phys. Usp. 22, 590 (1979)].
8. I. N. Goncharov, A. A. Gorbunov, V. I. Konov, A. S. Silenok, Yu. A. Skvortsov, V. N. Tokarev, and N. I. Chapliev, Preprint FIAN, No. 76, Moscow, 1980.
9. V. A. Vasil'ev, Yu. M. Romanovskiĭ, and V. G. Yakhon, Usp. Fiz. Nauk 128, 625 (1979) [Sov. Phys. Usp. 22, 615 (1979)].

Translated by Eugene R. Heath
Edited by S. J. Amoretty