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Self-sustained oscillation processes observed on vanadium surfaces heated by laser light

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Abstract. Different laser-light induced dissipative structures arising under various irradiation conditions during oxidation of vanadium in air are discussed. The observed instabilities are explained in terms of hydrodynamics, since the oxidation process takes place in the liquid phase, taking into account the influence of surface tension forces and gravitational effects on liquids of different densities. The decisive role of positive feedback between thermal and hydrodynamic degrees of freedom in the process of instability development is emphasized.

1. Introduction

It is well known that processes of laser-light induced oxidation of metals are good examples for the study and modelling of different self-organization events, as well as dissipative structures, spiral waves, etc. [1–4]. The oxides formed at the temperatures achieved during our previous investigations were in the liquid phase (e.g. melting point of V_2O_5 is 680 °C), whereas the metal was solid.

We note that there are a number of investigations relating to the appearance of strongly regular (periodical) structure patterns in laser–solid interactions [5–10]. These works do not relate to our work because they concern the effect of coherent light interactions, whereas in our case the structures (thermal fields) do not have coherent sources.

In previous papers we examined the existence of Marangoni hydrodynamic instabilities in liquid V_2O_5. In these experiments the horizontally situated vanadium plates were illuminated by a CW CO_2 or a YAG laser light focused from above. Kelvin–Helmholtz instability was registered for a quasi-linear flow geometry and Taylor instability for a planar flow geometry [2–4].

The aim of this paper is to present experimental results and their theoretical interpretation concerning different structures arising in liquid vanadium oxide, due to different combinations of laser light illumination from below and above.

2. Experimental details

Two types of experiment were carried out using a metallic vanadium target measuring $40 \times 40 \times 0.9\ mm^3$, chemically cleaned and polished. The surfaces of the samples were very smooth and, depending on the quality of cleaning, their absorptivity (A_0) varied in

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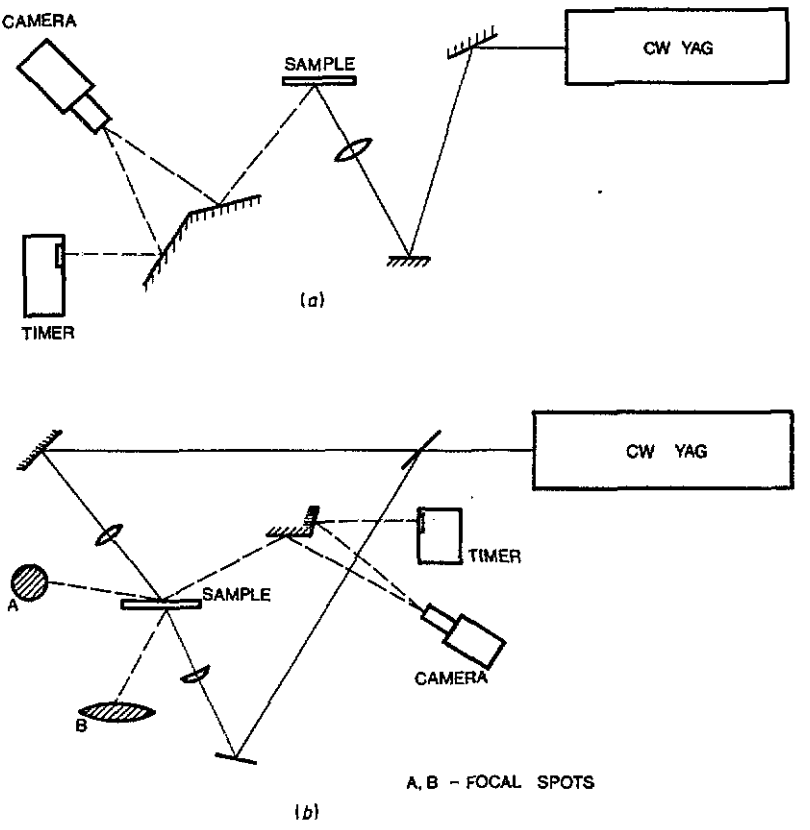

Figure 1. Scheme of experimental set-up used for irradiation of the sample from below (*a*) and from below and above (*b*).

the range 2–8% (which determines the time of activation during the oxidation process) [3].

The activation time of a heterogeneous reaction (here the oxidation of metal in air) is defined as the time during the process which gives rise to a positive feedback. As a result, a thermochemical instability develops in the form of a drastic increase of the absorptivity of the system.

In case (*a*), the target was irradiated on a highly expanded elliptical focal spot measuring 1 × 21 mm by a laser of power 80 W. In case (*b*), the radiation was divided into two equal parts for irradiation: a circular spot 15 or 25 mm in diameter on the upper surface, and an elliptical spot measuring 1 × 20 mm on the lower surface. In all experiments, the structures arising on the irradiated surfaces were monitored with a camera.

With the help of a specially mounted mirror system, a timer was photographed simultaneously with the same camera. For study of the temperature distribution in the structures, the photographic film was submitted to densitometric processing. In several experiments a set of thermocouples was embedded into the lower surface of the target in order to follow the temperature distribution independently of the densitometric method.

3. Results and discussion

3.1. Irradiation from below

Photographs taken at different times are presented in figure 2.

The full curve shows the trajectory of the brightest point ('hot point') in the phase space $\{x, t\}$. The broken curve shows the trajectory of points with a brightness level of $I = 0.1\, I_{max}$ of a unique spot (where I_{max} is the maximum brightness as observed at the centre of the target). Measurements of the thermal field with a series of thermocouples give results identical to those obtained with densitometry. It is clearly seen from figure 2 that the thermal field has a stationary external boundary and that the brightest point exhibits self-sustained oscillations with a period of 15 s.

The motion of this bright point could be explained on the basis of Rayleigh–Taylor instabilities. Such instabilities are generally observed in the fluid between concentric cylinders when the inner cylinder is rotated, and at certain speeds toroidal vortices spread from the ends until an array of vortices stacked upon each other covers the entire length of the flow domain. In the present work, what is relevant is the dynamical instability of the interface between heavy and light liquids. In such an instability the heavy liquid is displaced by the light liquid.

Many examples showing Rayleigh–Taylor instability are known, e.g. the instability of escape of cavitation bubbles [11–15].

Let us examine one of the simplest models of Rayleigh–Taylor instability: the instability in a gravitational field of the interface of two liquids with different densities. If the heavy liquid is below the light one, small perturbations of the interface represent waves satisfying the dispersion law $\omega = \sqrt{gk}$ (where g is the gravitational constant and k is the wavenumber). Otherwise, if the heavy liquid is above the light one, the interface

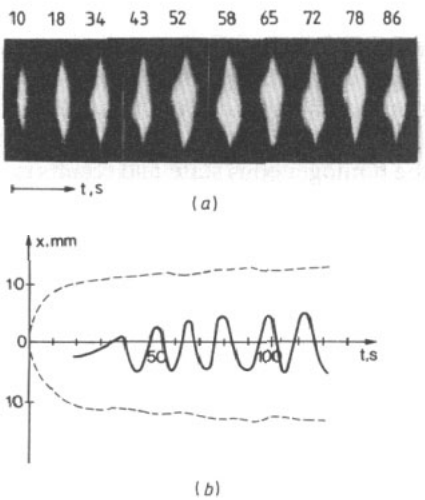

Figure 2. Thermal radiation from the heated surface. (*a*) Microphotographs made in different times. (*b*) Trajectory of the brightest spot in phase space $\{x, t\}$.

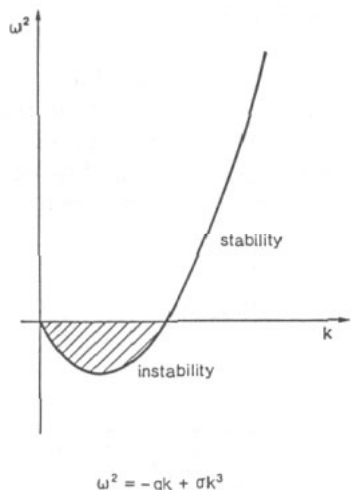

Figure 3. Function $\omega^2(k)$ showing areas for stable and unstable solutions.

becomes unstable, and small perturbations increase exponentially with an increment proportional to g. In this case, we have $\omega^2 = -gk$. Taking into account the influence of the surface tension (σ) on the spectrum of the surface waves, we have

$$\omega^2 = -gk + \sigma k^3. \tag{1}$$

It is clearly seen that the spectrum of instability modes will be cut at a certain wavenumber, namely $\omega^2 \leqslant 0$ in the range of wavenumbers 0–k_0 (figure 3).

In 'real' experiments using finite samples of size l, the oscillations with longest wavelength will be limited by boundary conditions. If $l < 1/k_0$, Rayleigh–Taylor instabilities will not develop.

If one examines the profile of a liquid oxide layer using a camera recording technique from the side view in our experiments, it may be concluded that the oxide surface oscillates with a wavelength (λ) comparable in magnitude to the length of the irradiated strip (figure 4). The inhomogeneity of the thermal field in the oxide layer, originating from the Gaussian laser beam on the sample, causes inhomogeneity in the surface tension coefficient, which can be described as

$$\sigma = \sigma_0 + (\mathrm{d}\sigma/\mathrm{d}T)(T - T_0)$$
$$\mathrm{d}\sigma/\mathrm{d}T = -B(\rho/\mu)^{2/3} \tag{2}$$

where ρ is the density, μ is the molar mass of the oxide, T_0 is room temperature, σ_0 is the surface tension coefficient at room temperature and $B = 2.1\ \mathrm{g\,cm^2\,K\,s^{-2}}$ is a proportionality constant peculiar to equation (2) [14].

The reason for the liquid layer stability loss can be explained in part by geometrical factors.

In the case of a beam with a homogeneous intensity distribution the temperature distribution in the steady state, at high enough temperature, is inversely proportional to the thickness of the layer.

Due to the surface tension variation (see equation (2)), surface currents from hot to cold appear. As a result, due to the positive feedback between the thermal and hydrodynamic degrees of freedom, the fluctuation in the thickness of the liquid layer can be increased. On irradiation of the sample from above, as in [1–4], this type of positive feedback causes a stability breakdown of the homogeneous state and results in the appearance of dissipative structures [1].

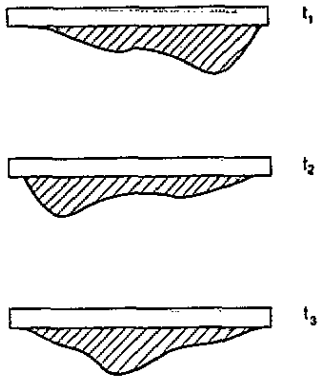

Figure 4. Cross sections of liquid oxide layers at different times.

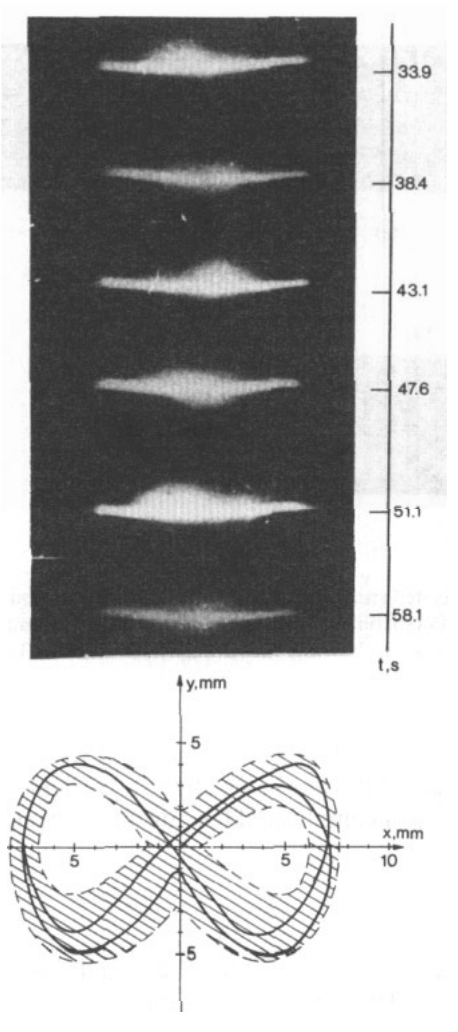

Figure 5. Microphotographs of area irradiated from below with trajectories in phase space $\{x, y\}$. The diameter of the beam reaching the target from above was 15 mm.

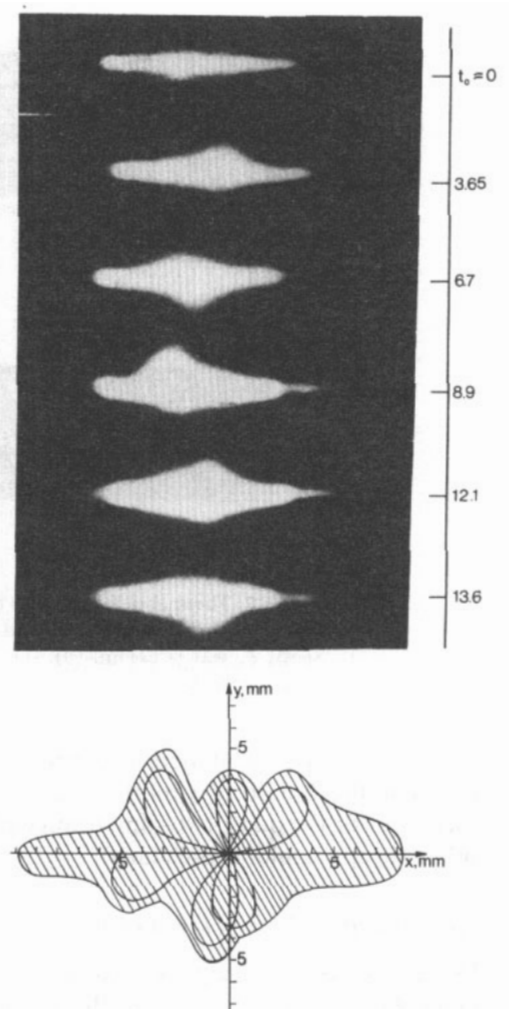

Figure 6. Micrographs of area irradiated from below with trajectories in phase space $\{x, y\}$. The diameter of the beam reaching the target from above was 25 mm.

On irradiation of the liquid layer from below, as in the present work, we can observe the competition of two effects:

(i) due to gravity forces, the thickness of the liquid layer should be a maximum in the centre, and

(ii) due to the Gaussian temperature distribution, the thickness of the liquid layer should decrease because of the influence of the outward directed surface currents arising from the surface tension variations.

As a result of the axial symmetry of the energy distribution in the liquid layer, losses and self-sustained oscillations appear which are supported by the laser-light energy absorbed in the system. Such self-sustained oscillations have been demonstrated experimentally as shown in figure 2.

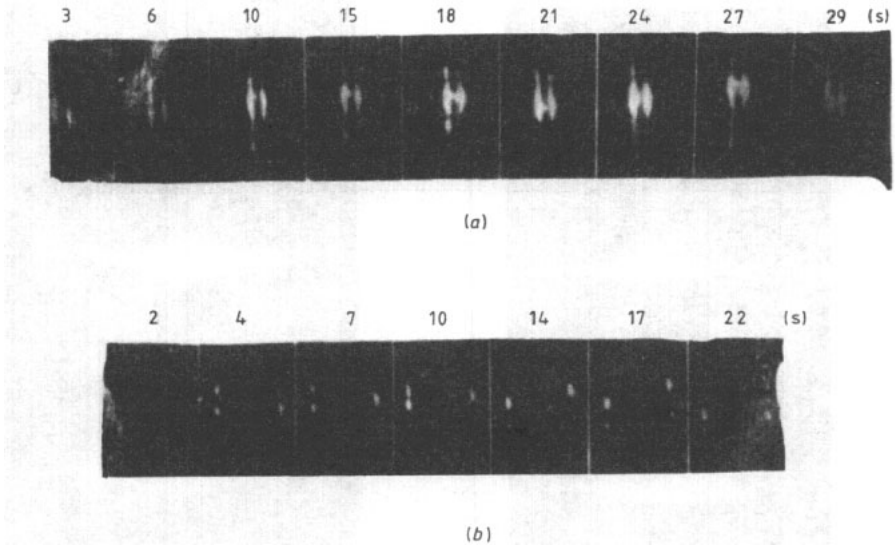

Figure 7. Thermal radiation from the heated surface by the radiation of two independent lasers. Pictures have been obtained in different times. (*a*) The distance between the elliptical spots, d, was ≈ 10 mm. (*b*) The distance, d, between the elliptical spots was ≈ 3 mm ($d_{crit} \approx 4$ mm).

For the quasi-one-dimensional problem discussed here, we had only one degree of freedom: the 'hot drop' exhibited oscillations along the line in which the laser light was focused. For the quasi-two-dimensional problem a complicated law for the movement of 'hot drops' could be presumed.

3.2. Irradiation from below and above

In order to check our suppositions, a special experiment was performed with the experimental set-up shown schematically in figure 1(*b*). The thermal fields arising in the lower part of the sample were monitored with a camera; the results of densitometric processing being presented in figures 5 and 6.

Table 1.

Irradiation	Form of the beam	Structures
From above	Stretched ellipse	Steady-state structures **(a lot of hot points)**
From above	Broadened spot (24 mm)	**Spiral waves**
	Medium spot (18 mm)	**Branch structures**
	Small spot (12 mm)	Reverberator
From below	Stretched ellipse	Autowaves of hot points
From below and from above	Above: circle	
	Below: ellipse	Lissajous figures
From above in two stripes	Stretched ellipses	**Synchronized** moving of hot points at $d < d_{crit}$ individual moving of hot points at $d > d_{crit}$

These figures present the phase space $\{x, y\}$ where the x and y coordinates correspond to the main axes of the elliptical focal spot (of figure 1(b)).

The area marked by the broken curves shows the limited $\{x, y\}$ space inside which the 'hot tongue' traces out different Lissajous trajectories. Such oscillations in the intensity of brightness demonstrate the existence of temperature gradients in the surface tension (as in the first experiment).

3.3. Irradiation from above

An experiment was performed to investigate the competition and synchronization of hot point motion in two spatially separated inhomogeneously heated strips. Two independent YAG laser sources, both with power 50 W, were focused from above in two stretched elliptical spots (15×1 mm^2) parallel to each other a distance d apart on the surface of vanadium. The distance between the two focal spots was varied from 10 to 3 mm. A correlation was observed experimentally for the hot point oscillations:

$$K = \frac{1}{L^2 T}\int_0^T (x_1 - x_2)^2 \, \mathrm{d}t \tag{3}$$

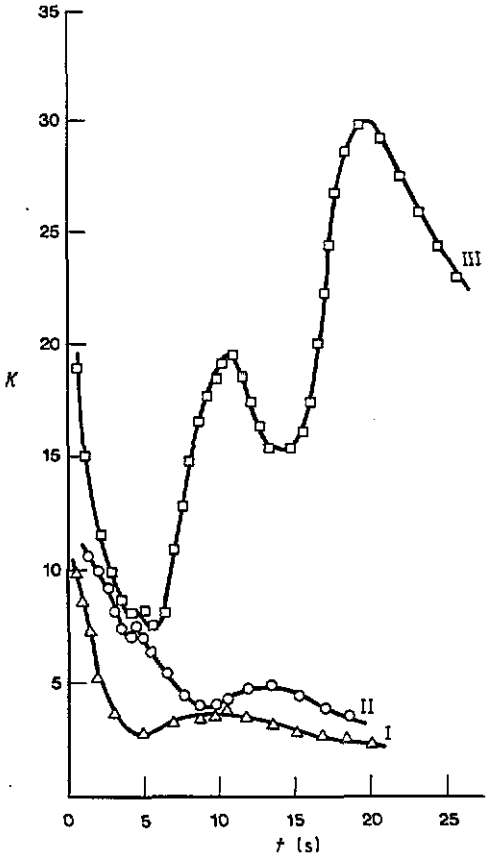

Figure 8. The correlation function constructed on the basis of equation (3). (Cases I and II with synchronization, case III without synchronization, i.e. I (d = 3 mm), II (d = 5 mm) and III (d = 10 mm)).

where L is the length of the irradiated spot (mm), T is time (s), and x_1 and x_2 are the coordinates of maximum brightness in the different spots. This function has values varying from 0 to 1.

If $K = 0$, we have full synchronization (the hot points move in phase), e.g. the spots with maximum brightness are oscillating in synchrony along the main axis of the elliptical focal spots.

Figure 7 demonstrates the situation for different moments of times at distances ($d = x_1 - x_2$) $d = 10$ mm and $d = 3$ mm.

Figure 8 presents the function $K = f(t)$ for experimental data at different d-values as obtained with (3).

It is clearly seen that for short distances there is good synchronization (K is decreasing), while for longer d the synchronization disappears, the oscillations becoming independent. We note that in our case there was a critical separation $d_{crit} \approx 4$ mm (see table 1).

4. Summary

The conclusions based on [1] and [4] and on the present work on laser-light induced space–time self-organization are summarized in table 1.

Acknowledgments

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