

# Transmittance in a thin aluminum layer at nanosecond pulsed laser ablation

Y.V. SENATSKY,<sup>1</sup> N.E. BYKOVSKY,<sup>1</sup> S.M. PERSHIN,<sup>2</sup> AND A.A. SAMOKHIN<sup>2</sup>

<sup>1</sup>P.N. Lebedev Physics Institute of the Russian Academy of Sciences, Leninsky prospect, 53, 119991 Moscow, Russia

<sup>2</sup>A.M. Prokhorov General Physics Institute of the Russian Academy of Sciences, Vavilova Street, 38, 119991 Moscow, Russia

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## Abstract

Since the beginning of the 1960s and up to the present time, laser ablation from metal surfaces under the action of short light pulses has been the topic of many researches. One of the first objects in the early ablation experiments presented thin metal films evaporated by radiation, which were used in lasers with nanosecond pulses as optical gates for  $Q$ -switching in resonators or for decoupling in amplifiers. Bleaching of the gates based on metal layers with a time constant  $\tau_t \approx 10^{-8}$  s observed in a number of experiments, was usually considered as a result of a simple evaporation of matter. We analyze the data of the experiments with a mylar tape [ $\approx 0.05$   $\mu\text{m}$  aluminum (Al) layer on  $\approx 20$   $\mu\text{m}$  lavsan substrate] used as a gate for optical isolation in one of the first Nd: glass laser facilities with a power of  $\approx 1$  GW. That gate was irradiated by pulses of  $Q$ -switched oscillators: pulse duration  $10^{-7}$ – $10^{-8}$  s, intensity  $10^7$ – $10^8$  W/cm<sup>2</sup>. A jump in the transmittance of the expanding Al layer was registered (from  $\approx 0.1$  to 50% at  $\tau_t \approx 10^{-9}$  s). The present study shows that one should consider the metal–insulator phase transition in a superheated liquid metal layer as the mechanism of the fast (up to  $10^{-10}$ – $10^{-11}$  s) increase in the transmittance of the Al film gate at nanosecond-pulsed laser ablation.

**Keywords:** Neodymium laser; Optical gate; Aluminum film ablation; Metal–insulator transition

## 1. INTRODUCTION

One of the first available optical gates used in the 1960–1970s to  $Q$ -switch in the laser cavity, to decouple cascades in amplifiers or the whole laser system from a target and in some other experiments was a thin metal layer evaporated by laser radiation (Grant, 1963; Basov *et al.*, 1966; Asmus, 1969; Senat sky, 1970; Vanyukov *et al.*, 1971; Askar'yan & Tarasova, 1973). Despite the advantages of such a gate (simple technology, low cost), it was a single-action device, which could not regain the initial low transmittance after actuation. Hence, this gate was soon replaced in laser setups by electro-optic shutters and bleachable dyes. At the same time, the analysis of data on the metal layer gate functioning (Senat sky, 1970; Vanyukov *et al.*, 1971; Zuev & Senat sky, 2015; Bykovsky *et al.*, 2016) is of interest in the context of studies of mechanisms of laser radiation interaction with metals, which have been performed since the 1960s up to the present time. Here we analyze the results of the early experiments on irradiation of a mylar tape

[aluminum (Al) layer on a lavsan substrate] by nanosecond ( $10^{-7}$ – $10^{-8}$  s) neodymium laser pulses, where an increase in the transmittance of the ablating Al layer by two to three orders of magnitude for a time  $\tau_t \approx 1$  ns was revealed (Basov *et al.*, 1966; Senat sky, 1970). The mechanism of the fast (up to  $10^{-10}$ – $10^{-11}$  s) increase in the transparency of the mylar with allowance for the overheating of a liquid Al layer to the metal–insulator transition temperature is discussed.

Zeldovich and Landau (1944) published the paper “On the relation between the liquid and the gaseous states of metals”, where they pointed to the possibility of the metal–insulator phase transition for an expanded metal at subcritical temperatures. Later that suggestion was considered when discussing a sharp (by several orders of magnitude) decrease in the electrical conductivity of mercury upon stationary heating (Kikoin & Senchenkov, 1967). Mott (1974) pointed out that localized electron states should arise when the metal density becomes sufficiently low. After the development of lasers capable to evaporate metals and produce plasma on their surfaces, one could expect the metal–insulator transition induced by laser radiation to be observed. Bonch-Bruevich *et al.* (1968) observed a change in the reflectance from

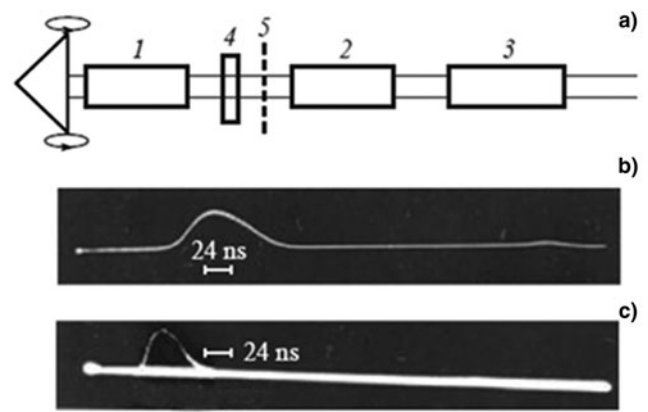
Address correspondence and reprint requests to: Y.V. Senat sky, P.N. Lebedev Physics Institute of the Russian Academy of Sciences, Leninsky prospect, 53, 119991 Moscow, Russia. E-mail: [senatsky@sci.lebedev.ru](mailto:senatsky@sci.lebedev.ru)

metals during the laser pulse when focusing a Nd:glass laser onto the surfaces of samples (Cu, Al, etc.). However, the mechanism of the metal–insulator transition was not discussed in that publication. Batanov *et al.* (1973) showed that, when a bulk metal target was heated by a laser with an intensity of  $10^7$ – $10^8$  W/cm<sup>2</sup>, the metal–insulator transition occurred, and it was accompanied by a wave of transparency in the melt. In accordance with that, the laser energy transmitted through the melt was spent on heating the solid metal under the melt layer at the bottom of the crater and on its deepening. Several other experiments and theoretical studies on the interaction of nanosecond laser pulses with metal targets provided data indicative of the metal–insulator transition and the formation of a transparency wave in the radiation–target interaction zone (Andreev *et al.*, 2003; Fishburn *et al.*, 2004; Pershin *et al.*, 2006; Porneala & Willis, 2006).

In the most publications of the 1960–1970s, an increase in transparency of thin metal films gates subjected to pulsed laser radiation was considered to be the result of a simple evaporation of a metal (Grant, 1963; Asmus, 1969; Vanyukov *et al.*, 1971; Askar'yan & Tarasova, 1973). We discuss the results of the early experiments on Al films ablation by nanosecond laser pulses and show that the transmittance jump in the Al layer observed in the experiments with a mylar tape (Basov *et al.*, 1966; Senatsky, 1970) was apparently the first direct indication on the metal–insulator transition induced by laser irradiation. That mechanism was responsible for the switching in the Al film gates.

## 2. EXPERIMENTS

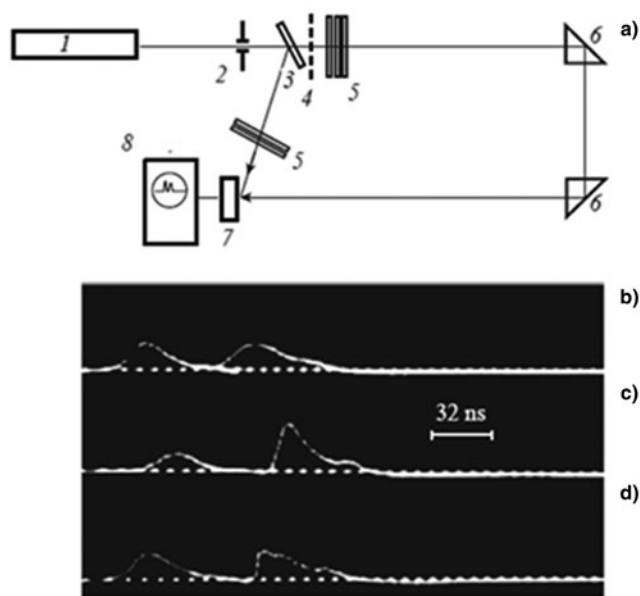
Within the research program aimed at developing a high-power laser (Basov *et al.*, 1966) a thin Al layer was used for the first time as an optical gate in an Nd: glass oscillator–amplifier laser system to decouple a rotating-prism *Q*-switched master oscillator from amplifier cascades and to sharpen the leading edge of the amplified pulse (Fig. 1). Rods of  $\text{Ø}30 \times 600$  mm, made of KGSS-7 glass in GOS-300 laser heads (LOMO, Leningrad) were applied both in the oscillator and the two-cascade amplifier. The rod end faces were cut at a small angle ( $1.5^\circ$ – $2^\circ$ ) to the axis. The film gate was placed outside the oscillator before the amplifier and consisted of a  $\approx 0.05$ - $\mu\text{m}$ -thick Al layer deposited on a  $\approx 20$ - $\mu\text{m}$  lamsan tape (mylar). The initial transmittance,  $K_0$ , of the Al film for a weak signal at the 1.06  $\mu\text{m}$  laser wavelength did not exceed 0.1%; hence, the oscillator was decoupled from the amplifier. Pulses at the outputs of the oscillator and amplifier were recorded by a FEK-09 photocell connected to a C1–14 oscilloscope. In the absence of the mylar, the oscillator and amplifier rods were optically coupled and formed in fact a unified oscillator, which produced several pulses with widths of 150–200 ns and an energy up to 80 J. With the mylar gate installed a single  $\approx 80$  ns pulse with energy up to 10 J was formed in the oscillator (Fig. 1b). A  $\text{Ø}30$ -mm beam from the oscillator



**Fig. 1.** (a) Schematics of the oscillator with a rotating prism *Q*-switch and a two-cascade amplifier based on KGSS-7 Nd:glass rods  $\text{Ø}30 \times 600$  mm (1–3) with a mylar gate (5), 4 – output coupler; (b, c) oscillograms of laser pulses: at the output of the oscillator (b) and at the output of the oscillator–amplifier system with a mylar gate (c).

was directed (without focusing) to the mylar and then to the amplifier. After the Al film absorbed the initial part of the pulse, the Al layer was evaporated, and the rest of the pulse arrived at the amplifier. Under these conditions, one would expect sharpening of the leading edge and a decrease in the pulse width by the time spent on heating the Al layer to evaporation. A  $\approx 50$  ns pulse (leading edge  $\tau_t \approx 10$  ns) with the energy  $\approx 60$  J and a power above 1 GW (beam divergence  $\approx 3 \times 10^{-3}$  rad) was produced at the amplifier output (Fig. 1c) (Basov *et al.*, 1966). This set of parameters was an achievement for Nd: glass lasers at that time.

An additional experiment was carried out in order to estimate the operating speed of the mylar gate (Fig. 2) (Basov *et al.*, 1965; Senatsky, 1970). A  $\text{Ø}10$  mm laser beam from a Kerr-cell *Q*-switched oscillator ( $\approx 20$  ns pulse, energy  $\approx 1$  J) was directed (without focusing) to the same mylar tape placed outside the resonator. It was found that at a pulse energy density  $\varepsilon < \varepsilon_{\text{th}} \approx 0.4$  J/cm<sup>2</sup>, the Al layer was not evaporated (the  $\varepsilon_{\text{th}}$  corresponded to the evaporation threshold). At  $\varepsilon < \varepsilon_{\text{th}}$ , the entire pulse passing through the Al film was attenuated by a factor of  $\approx 10^3$ . To eliminate only the weak initial portion of an oscillator pulse and select its main part, one must deal with  $\varepsilon > \varepsilon_{\text{th}}$ . Under these conditions, if the energy density in the initial part of the pulse does not reach  $\varepsilon_{\text{th}}$ , the film blocks this part. With an increase in the pulse intensity, the inequality  $\varepsilon > \varepsilon_{\text{th}}$  was satisfied, the gate became transparent, the main part of the pulse passed through the gate. The Al layer was removed from the tape in the irradiated area. The lamsan base transmittance  $K_{\text{lav}}$  for laser beams with intensity up to  $10^{10}$  W/cm<sup>2</sup> was  $K_{\text{lav}} \approx 90\%$ . The integral transmittance of the mylar gate (for the entire duration of the laser pulse) was  $K_{\text{int}} \approx 50\%$ . In a series of experiments with  $\varepsilon > \varepsilon_{\text{th}}$ , the Al film was vaporized both by the total  $\text{Ø}10$  mm oscillator beam (without a limiting diaphragm) and by the central part of the beam, cut by the diaphragm installed outside the oscillator. The pulses at the input and the output of the gate (after



**Fig. 2.** (a) Schematics of the experiment on shaping laser pulses of a  $Q$ -switched oscillator (1) by an external mylar film gate (4): diaphragm (2), glass plate (3), filters (5), prisms (6), photocell (7), oscilloscope (8). Oscillograms before (left) and after the gate and the optical delay line (right): ablation of the Al film by beams of  $\varnothing 10$  mm (b);  $\varnothing 5$  mm (c);  $\varnothing 1.5$  mm (d).

$\approx 23$  m optical delay line) were recorded using a FEK-09 photocell and a C1-14 oscilloscope (Fig. 2). A comparison of the oscillograms before and after the gate shows that a significant decrease in the width of the pulse-leading edge is observed but only for beams cut by a diaphragm (Fig. 2c, 2d). Irradiation of the film without a diaphragm barely changed the pulse after the gate (Fig. 2b). At the same time, the use of the  $\varnothing 5$  mm diaphragm reduced the leading edge of the transmitted pulse to  $\tau_t \approx 5$  ns (Fig. 2c), whereas in the case of the  $\varnothing 1.5$  mm diaphragm the leading edge was  $\tau_t \approx 1$  ns (Fig. 2d). The jump in the transmittance of the mylar gate was  $K_{\text{int}}/K_0 = 10^2\text{--}10^3$ . A more detailed description of the experiments according to Figures 1 and 2 is given by Zuev and Senatsky (2015) and Bykovsky *et al.* (2016).

### 3. DISCUSSION

The results of shaping pulses by an Al film gate in the experiments in Figures 1 and 2 can be explained by the spatial and temporal structure of laser beams, which was due to the non-uniform (over the active element cross section) development of lasing in oscillators. In the experiment where the mylar was irradiated by a whole  $\varnothing 10$  mm laser beam without a diaphragm (Fig. 2b), the spatial and temporal structures of the laser pulse led to a spread in transparency formation instants over the beam cross-section. Under these conditions, the pulse-leading edge did not shorten. On the contrary, when the film was exposed to radiation coming from only a small region of the active medium (i.e., when an external diaphragm was used), the spread in the transparency formation

instants over the beam cross-section decreased, and one could observe leading edge sharpening and pulse shortening (Fig. 2c, 2d). The leading edge width  $\tau_t \approx 1$  ns for the  $\varnothing 1.5$  mm diaphragm corresponded to the time resolution of the detection channel, which gives grounds to expect even shorter pulse-leading edge. These data suggest a conclusion that specifically the non-uniform spatial and temporal distribution of intensity in our laser system based on  $\varnothing 30 \times 600$ -mm active elements (Fig. 1) did not make it possible to observe high bleaching rates of the gate. Apparently, by the same reason, a fast increase in Al films transparency was not observed in experiments by Vanyukov *et al.* (1971) and Askar'yan and Tarasova (1973).

Thus, an optical gate composed of a  $\approx 0.05$   $\mu\text{m}$  Al layer on a lamsan tape (mylar) provided decoupling in an oscillator–amplifier laser system and sharpening of the leading edge of pulses transmitted through the gate (Basov *et al.*, 1966; Senatsky, 1970). The result obtained revealed also that the metal layer becomes transparent at a high rate (likely, for a time shorter than 1 ns). When discussing the observed transmittance jump in an Al film, it was indicated (Senatsky, 1970) that a “possible mechanism of fast increase in transparency is the ‘de-collectivization’ of conduction electrons in the expanding heated metal layer, that is, metal–insulator transition”.

A theoretical analysis of the stationary evaporation from the surface of a bulk metal target for radiation intensities up to  $10^8$   $\text{W}/\text{cm}^2$  in the presence of the metal–insulator transition (Andreev *et al.*, 2003) showed that this regime could be implemented only in a relatively narrow intensity range. An increase in laser intensity over these values may lead to explosive boiling of the superheated metastable phase and formation of plasma, as it was confirmed by experiments and calculations for metal bulk targets and films (Mazhukin *et al.*, 2014). Al films irradiated by laser pulses in the early experiments by Basov *et al.* (1966); Asmus (1969); Senatsky (1970); Vanyukov *et al.* (1971); Askar'yan and Tarasova (1973) became transparent in the limited intensity range  $10^7\text{--}10^8$   $\text{W}/\text{cm}^2$ . The increase in the transparency up to  $K_{\text{int}}(\epsilon) = 40\text{--}80\%$  of Al films with initial transmittances ranging from  $K_0 = 0.05$  to 2%, irradiated by  $\approx 30$ -ns Nd:glass laser pulses, was observed by Vanyukov *et al.* (1971) at  $\epsilon = 0.1\text{--}1.2$   $\text{J}/\text{cm}^2$  (Fig. 3). Apparently, this increase should be related to the metal–insulator transition in the expanding metal layer as well, although this mechanism was not considered in their work. The range of energy densities for transparency formation was limited:  $K_{\text{int}}(\epsilon)$  first increased with an increase in  $\epsilon$  and then decreased (Fig. 3). When passing the values  $\epsilon > 1.5$   $\text{J}/\text{cm}^2$ , an increase in radiation intensity causes ionization of Al atoms, due to which the transmittance of the formed plasma layer decreases (Fig. 3).

In order the metal–insulator transition was accomplished, it is necessary that the energy acquired by metal atoms was higher than their binding energy in the crystal lattice. The distance between atoms should be so increased that their short-range order was violated and there occurred

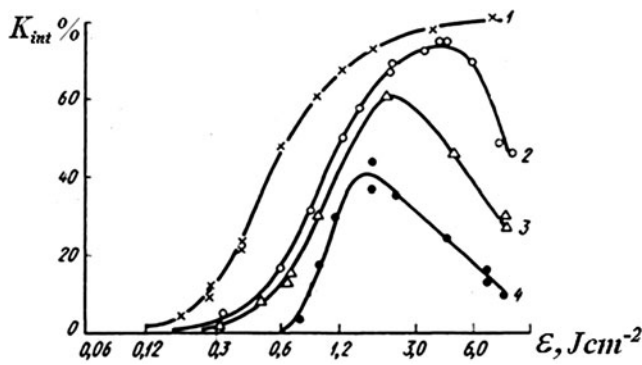


Fig. 3. The dependence of the transmittance  $K_{int}$  of Al films of different thicknesses on the energy density incident on the Al layer at different values of the film initial transmission: 1–2.2; 2–1.0; 3–0.5; 4–0.05% (Vanyukov et al., 1971).

localization of electrons on atoms (Mott, 1974). For a pulsed laser heating these processes occur as a result of explosive expansion of the region where the laser energy was deposited. A possibility to achieve the metal–insulator phase transition in an Al layer in conditions of experiments by Basov et al. (1966) and Senatsky (1970) may be shown using the following estimations. With the data on thermal diffusivity of Al,  $k \approx 0.1 \text{ cm}^2/\text{s}$ , one can estimate the film heating time. For the film with the thickness  $h \approx 0.05 \text{ }\mu\text{m}$  this time is  $h^2/k \approx 250 \text{ ps}$ . Thus, the heating of the Al film in our experiments can be considered as occurring simultaneously throughout the entire film volume during 20–80 ns laser pulses. Let us estimate for a film fragment  $\varnothing 5 \text{ mm}$  and  $h \approx 0.05 \text{ }\mu\text{m}$  (area  $S \approx 0.2 \text{ cm}^2$  and volume  $V \approx 10^{-6} \text{ cm}^3$ ) the temperature to which this fragment can be heated when absorbing the radiation with  $\varepsilon = 0.2 \cdot \varepsilon_{th} \approx 0.08 \text{ J/cm}^2$  (80% reflection is taken into account). The mass  $m$  of the fragment (Al density  $\rho = 2.7 \text{ g/cm}^3$ ) is  $\approx 2.7 \times 10^{-6} \text{ g}$ . For the Al-specific heat  $c = 940 \text{ J/kg/K}$  (Porneala & Willis, 2006), we find the temperature to which the film is heated by radiation,  $T_h \approx 6500 \text{ K}$ . The critical temperature for Al is  $T_c \approx 7400 \text{ K}$  (Khomkin & Shumikhin, 2015) and the metal–insulator transition temperature is  $T_{mi} \approx 0.8T_c$  (Porneala & Willis, 2006). Thus, the estimate shows that the Al film with  $h \approx 0.05 \text{ }\mu\text{m}$ , irradiated by a laser pulse with the absorbed energy density  $\varepsilon \approx 0.08 \text{ J/cm}^2$  in our experiments could be heated to the metal–insulator transition temperature. The metal density decreases upon heating, and the metal–insulator transition may occur yet in the condensed phase (in the superheated liquid metal at the subcritical temperature range  $T_{mi} < T_c$ ), a circumstance that was pointed by Zel'dovich and Landau (1944).

The characteristic time  $\tau_t$  of the transmittance jump, observed in an Al film with  $h \approx 0.05 \text{ }\mu\text{m}$ , can be estimated on the assumption that the front of the transparency caused by the metal–insulator transition propagates in the film jointly with its expansion at a velocity  $v$ . The film-free expansion velocity  $v$  in the conditions of our experiments should not exceed the speed of sound in metallic Al ( $v < v_{Al} \approx 5 \times$

$10^5 \text{ cm/s}$ ). Therefore, the transmittance jump time can be estimated as  $\tau_t = 10^{-10}–10^{-11} \text{ s}$ . The fast transparency increase made it possible to form a leading edge,  $\tau_t \approx 1 \text{ ns}$  for a laser pulse transmitted through a mylar (Fig. 2d).

The observed stepwise increase in the Al film transparency can be compared with the dynamics of transparency formation, in case if that dynamics was a result of metal evaporation from the surface (layer-by-layer) with the successive decrease in the film thickness. The time necessary to completely evaporate and thus to make transparent an Al film with  $h \approx 0.05 \text{ }\mu\text{m}$  in this regime can be estimated as follows. Let us assume that a film fragment  $\varnothing 5 \text{ mm}$  is already heated by the radiation of the laser pulse-leading edge to the boiling temperature and continues to absorb radiation with a power density of  $\approx 2 \times 10^7 \text{ W/cm}^2$ . So, the energy of 4 mJ is supplied to a fragment of  $S = 0.2 \text{ cm}^2$  for  $\approx 1 \text{ ns}$ . With allowance for the film reflection, the absorbed energy is 0.8 mJ. The latent evaporation heat of Al is  $L_b \approx 10^4 \text{ J/g}$  (Porneala & Willis, 2006) and the energy necessary to evaporate Al from the film fragment is  $Q_b = mL_b \approx 27 \text{ mJ}$ . To accumulate it, the film must be irradiated for a time of  $\approx 30 \text{ ns}$ . Therefore, the time necessary to evaporate (layer-by-layer) an Al film of  $h \approx 0.05 \text{ }\mu\text{m}$  at the laser intensity of  $2 \times 10^7 \text{ W/cm}^2$  exceeds the experimentally found time of transparency formation in the film by a factor of more than 30. This estimation suggests that the observed stepwise increase in the film transparency is rather due to the metal–insulator transition in the expanding Al layer than its successive evaporation. Experimental data indicate also that an Al film, after absorbing the energy of the leading edge of a 20–80 ns laser pulse and transforming its properties to the new phase (metal–insulator transition), retains high transmittance and a rather good optical quality for several tens of nanoseconds. Indeed, the film gate used in our oscillator–amplifier system (Fig. 1) could support formation of collimated laser beams. This indicates to the absence of high scattering loss in the film for laser pulses with a width up to 50 ns passing through it.

#### 4. CONCLUSION

The experimental data on the ablation of thin Al films by nanosecond laser pulses with intensity of  $10^7–10^8 \text{ W/cm}^2$  and their analysis suggest that the fast increase in transparency of these films (used as optical gates in laser setups in 1960–1970s) should be related to the metal–insulator phase transition in the superheated liquid metal in the subcritical temperature range. It should be noted that the physical mechanisms of the interaction of the radiation of the moderate intensity with metals, with allowance for the metal–insulator transition, have been still studied insufficiently. To gain a deeper insight into the dynamics of the observed stepwise ( $\tau_t \approx 1 \text{ ns}$ ) increase in the transparency of a metal film under laser irradiation at  $10^7–10^8 \text{ W/cm}^2$ , it seems to be expedient to use ultrashort pulsed laser diagnostics of transient states of matter in an ablating medium during transformation

of its optical and physical parameters as it was done at first in the study of Sokolowski-Tinten *et al.* (1998).

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