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STATISTICAL MODEL FOR ASSESSING THE OPTICAL DENSITY  
OF LASER-PRODUCED MICROPLASMA

**Key words:** Laser microspectral analysis, optical density, laser radiation absorption

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ABSTRACT

The space and time change of the optical density of laser-produced microplasma is studied. A statistical model is proposed which allows its utilization for assessing plasma optical density at a constant, as well as at a changing laser radiation intensity.

INTRODUCTION

It is very important to know the exact laser energy balance - absorbed and reflected by both the studied sample and the plasma torch - for the aims of laser microspectral analysis<sup>(1-3)</sup>. In the present study a statistical assessment of the optical density of the laser-produced torch is made. The sensitivity of laser microspectral analysis varies within broad limits

and is dependent on several conditions: apparatus used, excitation conditions etc. That is why the use of lasers in spectroscopy gives rise to many problems that involve the enhancement of element detection limits. This is in close connection with the optimization of the conditions under which the laser radiation energy is better utilized for evaporation and excitation of the material.

An improvement in the detection limits and concentration sensitivity of laser microspectral analysis is attained by inclining the sample in relation to the laser beam direction <sup>(4)</sup>. The laser radiation optical path is thus shortened, decreasing the ejected substance energy absorption.

#### EXPERIMENTAL AND METHOD

The experiments are carried out with a laser microanalyser LMA - 1 (Carl Zeiss - Jena) operating in a free-generation mode at a wavelength of 1060 nm. The pulse duration is 300  $\mu$ s and the initial energy of the laser was 0.5 - 1 J. The general diagram of the experiment set-up is presented in Fig. 1.

The probing of the LMA - 1 plasma torch is carried out by an attachment (2) for diverting part of the laser beam, placed between the laser source (1) and the microscope. Thus it is possible in the course of a single generated pulse to create a plasma torch (8) from the laser beam directly impinging on the sample (7). Simultaneously this same torch can be probed in a transverse direction.

The plasma absorption capacity is studied by means of the action of a focused laser beam on different samples of aluminium, carbon, brass, iron and galena. The plasma torch is probed at eleven points

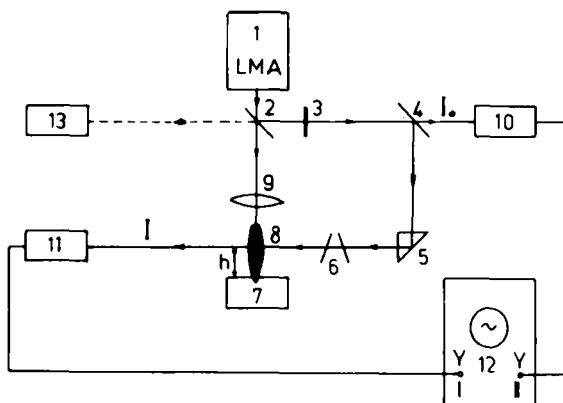


FIG. 1

**General diagram of the experiment set-up**

at every half a millimeter - from 0.5 mm to 5.5 mm above the sample surface. The plasma torch optical density was followed at a constant height of the probing beam for a duration of 300  $\mu$ s from the start of laser generation, at an interval of 25  $\mu$ s (13 points in all).

**STATISTICAL MODEL AND EXPERIMENTAL CHECK**

The plasma torch optical density is determined from the ratio of the intensities of the incident ( $I_o^m$ ) and the transmitted laser radiation through the torch ( $I^m$ ). It is obtained by comparing the amplitudes of the same generation peaks from the two channels of the dual-beam storage oscilloscope. The connection between the two intensities is expressed by the Bouguer - Lambert law:

$$I = I_o \exp(-\alpha l) \quad (1)$$

The  $I_o^m$  and  $I^m$  values are not the same in the case when there is no plasma torch. At fixed

intervals after the laser pulse starts the difference  $dI^m = I_o^m - I^m$  is registered for identical generation peaks.

The following statistical model may be treated. Let us denote the registered by the apparatus intensity values of the incident radiation with  $I_o^m$ , the transmitted through the torch laser radiation with  $I^m$ , and the difference between the two magnitudes in the absence of plasma at a torch optical density of  $k = 1$  ( $dI^m = I_o^m - I^m$ ).

The experimental conditions allow the hypothesis of noise additivity with a zero mathematical expectation to be accepted, i.e.

$$I_o^m = I_o + \tilde{I}_o = I_o^N ; E(\tilde{I}_o) = 0 \quad (2)$$

$$I^m = I + \tilde{I} - \Delta I = I^N - \Delta I ; E(\tilde{I}) = 0$$

$I^N$  and  $I_o^N$  are the corresponding intensities of the noise-corrupted laser radiation.

$$\Delta I = \Delta I_o e^{-\alpha l} = \Delta I_o k$$

$\Delta I_o$  corresponds to the laser beam losses occurring between the point of the  $I_o$  measurement and the plasma.  $\Delta I$  is the magnitude which would be measured if a laser beam corresponding to the losses, passes through the plasma.

Since

$$dI^m = (I_o^m - I^m)_{k=1} = \tilde{I}_o - \tilde{I} + \Delta I_o, \quad (3)$$

$$E(dI^m) = E(\Delta I_o).$$

At a constant generated laser radiation,  $I_o = \text{const}$ , the following assessment of the plasma torch density is obtained:

$$E(I^m) = E(kI_o) - E(k\Delta I_o).$$

Taking into account eq.(3) and since  $E(I_o^m) = I_o$ , it follows that:

$$k = \frac{E(I^m)}{E(I_o^m) - E(dI^m)} \quad (4)$$

In the cases when  $I_o = \text{const}$  can not be guaranteed, a statistical assessment of the plasma absorbing capacity  $k$  can be made. Let us denote the error of the measured and the determined by eq.(1) intensity of the transmitted through the torch laser radiation with  $\xi = I^N - k'I_o^N$ .

According to the criterion of minimum mathematical expectation of the square of the error  $E(\xi^2) = \min$ , for  $k'$  is obtained:

$$k' = \frac{E(I_o^N I_o^N)}{E((I_o^N)^2)}$$

The  $dI^m$  and  $I_o^m$  values are obtained from independent experiments and the hypothesis for the random magnitudes  $I_o^m$  and  $\Delta I_o$  independency can be accepted.

Taking into account eq.(2) the following is derived:

$$k' = \frac{E(I^m I_o^m) + E(I_o^m \Delta I)}{E((I_o^m)^2)} \quad (5)$$

Solving eq.(5) for  $k'$ , the following is obtained:

$$k' = \frac{E(I^m I_o^m)}{E((I_o^m)^2) - E(I_o^m)E(dI^m)} \quad (6)$$

In assessing  $k'$ , in the case when  $I_0 = \text{const}$ , the error can be determined by the formula (6). We accept the hypothesis of  $\tilde{I}_0$  and  $\tilde{I}$  independency (the errors of the two independent measurements channels). It follows:

$$k' I_0 = \frac{E[(I_0 + \tilde{I}_0) \cdot (I + \tilde{I})]}{E(I_0 + \tilde{I}_0)^2} I_0 = \frac{I}{1 + E(\tilde{I}_0^2)/I_0^2} \quad (7)$$

where:

$$\frac{E(\tilde{I}_0^2)}{I_0^2} = \frac{\text{VAR}(\tilde{I}_0)}{I_0^2} = V^2$$

The variation coefficient  $V$  ( $V \ll 1$ ) is a magnitude specific for the experimental conditions. Then  $k' \approx k$ , or at a known  $k'$

$$k = k'(1 + V^2) \quad (8)$$

The variation coefficient can be determined from the following equation

$$V^2 = \frac{E(\tilde{I}_0^2)}{I_0^2} = \frac{E(I_0^m - E(I_0^m))^2}{E(I_0^m)^2} \quad (9)$$

An assessment for the maximum variation coefficient due to a measurement error can be obtained:

$$V^2 = \frac{E(\Delta \cdot E(I_0^m))^2}{E(I_0^m)^2} \leq \frac{\Delta_{\max}^2 E(I_0^m)^2}{E(I_0^m)^2} \leq \Delta_{\max}^2 \quad (10)$$

where is the relative instrumental error.

$$\Delta = \frac{I_0^m - E(I_0^m)}{E(I_0^m)}$$

Thus, in addition to an assessment of the plasma optical density, a statistical criterion for the generated laser beam intensity constancy is obtained.

In the case when  $\Delta_{\max}^2 < V^2$ , the error due to the laser beam intensity inconstancy adds to the error due to the experiment setting and  $k$  is calculated from eq.(6). In the opposite case, even if a correction is made with eq.(8), the difference will be insignificant (Table 1), i.e. it is advisable eq.(6) to be employed in both cases. This equation yields reliable results in both cases ( $I_0 = \text{const}$ ,  $I_0 \neq \text{const}$ ). With its aid the plasma optical density can be assessed without controlling the laser radiation constancy.

TABLE 1  
Comparison of the optical density values calculated according to eq.(6) and eq.(8)

Sample	Brass		Aluminium		Galena	
Conditions	eq.(6)	eq.(8)	eq.(6)	eq.(8)	eq.(6)	eq.(8)
$h = 0.5 \text{ mm}$	0.364	0.367	1.478	1.490	1.754	1.784
$\tau = 50 \mu\text{s}$						
$h = 1.0 \text{ mm}$	0.703	0.707	1.057	1.064	1.069	1.111
$\tau = 100 \mu\text{s}$						
$h = 2.0 \text{ mm}$	0.669	0.717	0.660	0.666	0.709	0.711
$\tau = 150 \mu\text{s}$						
$h = 3.0 \text{ mm}$	0.265	0.286	0.370	0.381	0.583	0.596
$\tau = 200 \mu\text{s}$						

Fig. 2 presents the plasma torch optical density change with time for a brass sample followed at intervals of  $25 \mu\text{s}$  at fixed probing heights. Each point on the plot is obtained as an average of at least five measurement. The torch optical density varies between the values of 0.5 and 2. The results obtained by means of the described method are computer processed.

The presence of a maximum in the plasma torch optical density at the start of the generated pulse indicates a strong screening action of the plasma in the initial moments of laser generation. At power densities of about  $0.1 \times 10^6 \text{ W/cm}^2$  non-linear effects

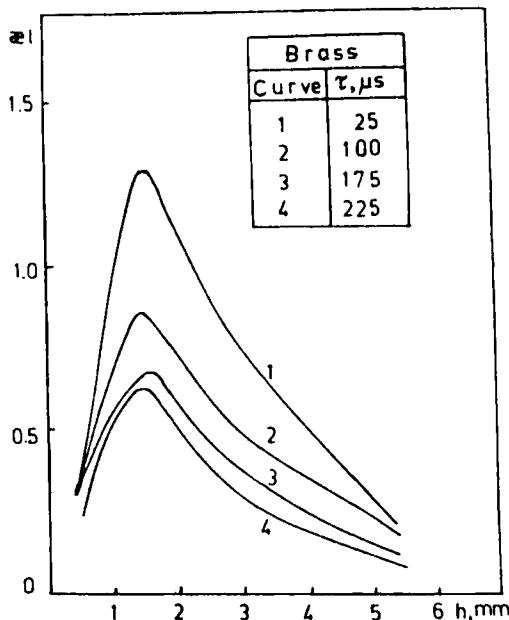


FIG. 2  
Torch optical density  
change with probing  
height at fixed moments  $\tau$ .

are not to be expected. The most likely explanation for the maximum values of the optical density during the initial moments of generation is the fractionation of the erosion products - a consecutive introduction of vapours, lighter particles and finally of heavier particles.

Fig. 3 shows the change in plasma optical density with the probing height at fixed moments after the beginning of the laser pulse. The maximum value for brass 1.57 is at a probing height of 1.5 mm. The assessments of the optical densities are guaranteed with a relative standard deviation of 0.06 - 0.35.

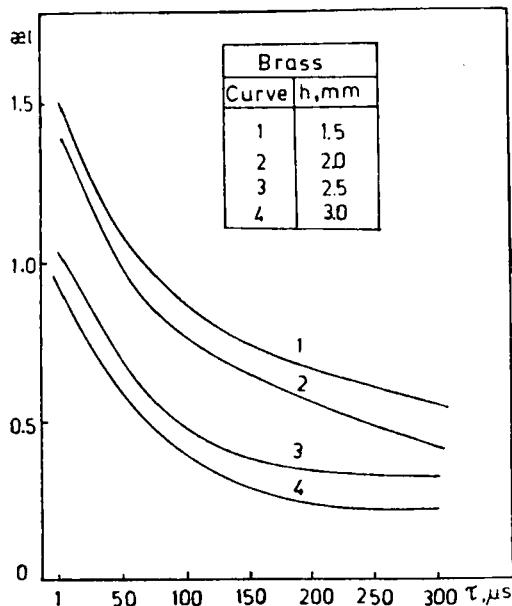


FIG. 3  
Torch optical density  
change with time at  
fixed probing heights  $h$ .

Although the data in Table 1 show a relatively satisfactory constancy of the laser energy, the relative standard deviation varies in a broad interval. This could be due to the significant space and time plasma heterogeneity.

### CONCLUSION

The statistical model allows the assessment of the generated laser beam intensity constancy. The derived formula (6) for the assessment of the plasma torch absorbing capacity is not sensitive towards the changes in the generated laser beam, i.e. it can be employed for assessing the optical density at a constant and changing laser beam intensity.

The change in the plasma torch optical density with the height of the studied zone above the sample surface follows a certain regularity. In the first moments of the generated pulse the optical density passes through a maximum, which is different for each material.

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