

Evaluation of laser-induced thin-layer removal by using shadowgraphy and laser-induced breakdown spectroscopy

M. S. Rabasović¹ · D. Šević¹ · N. Lukač² · M. Jezeršek² · J. Možina² · P. Gregorčič²

Received: 10 September 2015/Accepted: 4 February 2016 © Springer-Verlag Berlin Heidelberg 2016

Abstract Shadow photography and laser-induced breakdown spectroscopy (LIBS) are studied as methods for monitoring the selective removal of thin (i.e., under 100 µm) layers by laser ablation. We used a laser pulse of 5 ns and 16 mJ at 1064 nm to ablate an 18-µm-thin copper layer from the fiberglass substrate. On the basis of shadowgraphs of the laser-induced shock waves, we measured the optodynamic energy-conversion efficiency, defined as the ratio between the mechanical energy of the shock wave and the excitation-pulse energy. Our results show that this efficiency is significantly higher for the laser pulse-copper interaction than for the interaction between the excitation pulse and the substrate. LIBS was simultaneously employed in our experimental setup. The optical emission from the plasma plume was collected by using a spectrograph and recorded with a streak camera. We show that advancing of laser ablation through the copper layer and reaching of the substrate can be estimated by tracking the spectral region between 370 and 500 nm. Therefore, the presented results confirm that LIBS method enables an on-line monitoring needed for selective removal of thin layers by laser.

1 Introduction

Selective laser ablation of thin layers means subliming or vaporizing a thin (i.e., under 100 µm) layer without damaging the substrate or the intermediate underlying layer. This is especially important in several applications, such as rapid prototyping of printed circuit boards. In this case, it is essential that the copper is removed from an engraved track without damaging the composite substrate under the copper, as the damaged substrate may carbonize and therefore cause the unwanted altering of its electrical properties [1]. Since the laser-engraved track depth significantly varies with different parameters, such as copper layer thickness, surface conditions, and processing-laser parameters, an adaptation of the laser-processing parameters is necessary to achieve optimal results. However, the current laser-engraving systems lack an on-line monitoring method that would enable selective layer removal of changing surface properties during the laser processing.

Several methods of detecting the acoustic waves and plasma spectrum during the thin-layer removal have been investigated [1-3]. Here, the acoustic waves have been measured by a microphone and a laser-beam-deflection probe [2, 4-6]. The main idea behind these methods is to detect the time of flight of the shock wave, which is generated during the laser ablation, since the shock wave's velocity varies with the optodynamic (OD) energy-conversion efficiency, defined as the ratio between the mechanical energy of the macroscopic motion and the excitation-pulse energy [7]. This variation results in different time of flights at a constant distance between the probe or microphone and the laser-material interaction site. The OD energy-conversion efficiency changes when the ablated material changes, since a thin layer is already removed and the ablation of the substrate takes place. The

P. Gregorčič peter.gregorcic@fs.uni-lj.si

¹ Institute of Physics, University of Belgrade, Pregrevica 118, 11080 Zemun, Serbia

² Faculty of Mechanical Engineering, University of Ljubljana, Aškerčeva 6, 1000 Ljubljana, Slovenia

second type of the method for monitoring the removal of thin layers is laser-induced breakdown spectroscopy (LIBS) [3, 8-10] that enables the spectrum analysis of the plasma emission generated during the laser-material interaction. In such a way, the identification of the elemental composition of the material being ablated is possible [11].

The main aim of this paper is to simultaneously use two monitoring methods, shadowgraphy [7] and LIBS [12], and to evaluate both techniques for the analysis and monitoring of laser ablation of copper from the printed circuit boards. LIBS has been chosen, since in the last decade it has been extensively used for selective thin-layer removal [13–19]. Therefore, we employed it as a comparative and/or reference technique in our experimentation. On the other hand, the main goal of using shadowgraphy is to measure the OD energy-conversion efficiency of the laser ablation. In such a way, it is possible to gain better insight into the shockwave dynamics. This could lead to improvements in measurements using other optodynamic methods [2], such as the laser-beam-deflection probe and microphone measurements enabling an on-line monitoring of selective thinlayer removal.

2 Experimental setup

Our experimental setup is sketched in Fig. 1. The optical breakdown on a printed circuit board was induced by a Q-switched Nd:YAG laser ($\lambda = 1064$ nm, Quantel, France, Brilliant) with pulse duration of 5 ns, pulse energy up to 270 mJ, and repetition rate of 10 Hz. We used an attenuator to reduce the pulse energy to 17 mJ. The excitation pulse was focused on a target by using a quartz lens with the focal length of 100 mm (L1). As a target, we used



Fig. 1 Experimental setup for shadowgraphic and LIBS analysis of laser ablation

a printed circuit board having an 18- μ m-thick copper layer on a fiberglass substrate. The target was mounted on three motorized stages that enabled its positioning in all three directions with a resolution of 1 μ m.

For monitoring the optodynamic phenomena, we simultaneously employed two measuring methods: shadowgraphy [7] and LIBS [12]. Shock-wave dynamics was measured by shadowgraphy. For this purpose, we used an illumination flashlamp with duration of 8 ns (Nanolite, Germany, KL-K). The spark of the flashlamp was placed in the focus of the lens L3 having a focal length of 20 mm applied as a collimator. When the rays of the illumination lamp reach the shock-wave front, they refracts due to the refractive-index gradient. This casts a shadow that can be visible on the screen [20]. Instead of the screen, we used a DSLR camera (Nikon, Japan, D90) with a macrolens. The excitation laser, streak camera, illumination pulse, and DSLR camera were synchronized with a signal generator (Tektronix, US, AFG 3102). The experiment was controlled by custom-made software written in MATLAB, running on a personal computer.

For LIBS, the plasma emission was collected by L1 and then reflected by a beam splitter (BS) to another quartz lens (L2) with focal length of 100 mm. This lens collects the plasma emission and focuses it on the slit of the spectrograph (Princeton Instruments, USA, SpectraPro 2300i). A streak camera (Hamamatsu, Japan, C4334) was used for time-resolved monitoring of the ionic and neutral emission lines. The fundamental advantage of the streak scope is its two-dimensional nature, which is especially important in measuring time-resolved LIBS spectra. By using a streak camera, it is possible to acquire the time-resolved data from a single shot. This avoids the problems of assuring the repeatability that is required in other techniques, such as LIBS analysis by time-gated CCD. The used camera has the spectral range from 200 to 850 nm. The data were acquired and analyzed by using High Performance Digital Temporal Analyzer (HPD-TA) software, provided by Hamamatsu. Our time-resolved measurements can be accomplished in the time ranges from 1 ns to 10 ms. The time range of the streak images presented in results is 2 µs.

The shadowgraphs and the spectrographs, obtained by the LIBS method, were collected during the experimentation and processed offline after the measurements.

3 Results and discussion

3.1 Visualization of shock-wave dynamics

Typical shadowgraphs of plasma and shock wave, induced by the interaction between the excitation pulse and the printed circuit board, are shown in Fig. 2. The time after



15.5 µ

the excitation-pulse initiation is shown on the bottom-righthand side of each image. The shock wave expands spherically into a half-space with gradually decreasing velocity.

From acquired shadowgraphical images, we measured the shock-wave radius as a function of time. The radius was measured by fitting a circle to the shock-wave front. The obtained quantitative results are presented in Fig. 3. The



Fig. 3 Time-resolved shock-wave radius for the excitation pulse interaction with the copper surface (the *blue squares*) and on the clear substrate (the *violet circles*). The fitted Jones model [Eq. (1)] is shown by the *solid curves*

blue squares show the time evolution of the shock-wave radius that is induced by the interaction between the excitation pulse and the copper surface. The violet circles correspond to the measured results, when the excitation pulse interacts with the clean fiberglass substrate.

The OD energy-conversion efficiency $\eta = E_0/E_p$, defined as the ratio between the shock-wave energy E_0 and the pulse energy E_p [7], can be obtained by fitting the Jones model to the measured results [21]. Therefore, the solid curves in Fig. 3 show the fit of the Jones model [22]:

$$t = \frac{R_{\rm C}}{c_0} \left(\frac{2}{5}\right)^{\frac{2}{3}} \left[\left(1 + \left(\frac{5}{2}\right)^{\frac{5}{3}} \left(\frac{R}{R_{\rm C}}\right)^{\frac{5}{2}}\right)^{\frac{5}{2}} - 1 \right]$$
(1)

to the measured data. In Eq. (1), *t* is time, $c_0 = 346$ m/s is the speed of sound in air for ambient conditions (T = 25 °C, $p_0 = 10^5$ Pa), and R_C is the characteristic radius, defined as [22]:

$$R_{\rm C} = \left(\lambda_{\rm S}^5 \frac{E_0}{\gamma p_0}\right)^{\frac{1}{3}} \tag{2}$$

In Eq. (2), fitting parameter E_0 stands for the shockwave energy, $\gamma = 1.4$ is the adiabatic index of the gas, and $p_0 = 10^5$ Pa is the ambient pressure. The dimensionless geometry-dependent parameter for half-space expansion [23] equals $\lambda_5^5 = 2.35$.

15.0 μ

From the fit of Eq. (1) to the experimental data, using the least squares method, we obtained the energy of shock waves. In such a way, we measured the energy of the shock wave to be $E_{0C} = 3.7$ mJ for the pulse-copper interaction and $E_{0S} = 1.8$ mJ, when the interaction takes place on a clear substrate. From these results, we can conclude that the OD energy-conversion efficiency for the copper coating $(\eta_{\rm C} = 22 \%)$ is approximately two times larger than for the clear substrate ($\eta_{\rm S} = 11$ %). It should be noted here that the OD energy-conversion efficiency does not change significantly during the copper layer removal (i.e., before the substrate is reached), since this layer is thinner than the spot size. Therefore, monitoring of the shock-wave dynamics during the pulse-surface interaction allows determining when the layer is removed. However, the obtained results just confirm the proof of the concept. Since the data are processed offline, the shadowgraphic method does not allow us an online monitoring. For doing this, an on-line method, such as a laser-beam-deflection probe [5], should be employed.

3.2 The temporal evolution of the ionic and neutral emission lines

Compared to the results by Tong et al. [3], our streak camera system enables time-resolved LIBS analysis, allowing us to determine the time frame where spectra discrimination is best for a given laser excitation (regarding wavelength and energy). Real applications should be based on these results, using much cheaper spectroscopic equipment. Our LIBS analysis relies on identifying Cu (copper layer) and Si, Ca, Al, Na, O (fiberglass, printed circuit substrate).

Streak images of LIBS of copper conductor and printed circuit board substrate are shown in Fig. 4. Here, the vertical axes show time after the pulse excitation, while the wavelength is shown in the horizontal axes. The integrated spectrum of the range, defined by a dashed rectangle, is presented by the red line on the bottom of each image.

To distinguish various sample materials in real time, several spectra processing and comparison algorithms were studied by Tong et al. [3]. After initial trials, we decided to use linear correlation method with threshold treatment of acquired data, as an optimal solution for our experimental data.

Our calculations are based on modified correlation formula [3]. For a two-point data series (x_j, y_j) , where $j = 1 \dots N$, the linear correlation coefficient between the two series *COR* is defined as:

$$COR = \frac{\sum_{j} x_{j} y_{j}}{\left(\sum_{j} x_{j}^{2}\right)^{1/2} \left(\sum_{j} y_{j}^{2}\right)^{1/2}}$$
(3)

Here, *j* denotes the row index of streak image corresponding to the certain wavelength (defined by spectrometer calibration), and x_j and y_j represent the intensities of integrated

spectra at that wavelength. A correlation value close to unity means that the resemblance between the two spectra is high, and a value near zero implies little resemblance. It can be seen that the copper (conductive layer) and silicon and calcium (fiberglass) spectral lines are grouped together in the region between 510 and 530 nm, so this part of spectra is not very helpful for discrimination between the copper layer and the substrate of printed circuit board. To determine the correlation coefficients of measured spectra, we decided to use the spectral region between 370 and 500 nm. We used a normalized threshold of 0.1. The correlation coefficients were calculated by using the C code written for this purpose.

LIBS streak images of printed circuit board at the start, after 50 laser shots, 200 laser shots, 500 laser shots, and when the substrate is fully exposed, are shown in Fig. 5.

We tried various time frames for integrating the line profiles used in calculations. Typical evolutions of calculated correlation coefficients as a function of the excitationpulse number for three representative time frames are shown in Fig. 6. From these results, it is clearly visible that the steepest decrease in the correlation coefficient between the whole spectrum and the spectrum of the copper layer appears for the integrating time frame between 0.7 and 1.4 µs. Therefore, we selected this time frame as the most appropriate for discriminating the printed board layers for the parameters used in our experimentation. The correlation coefficient should decrease with increasing number of laser shots. If the plasma continuum is included in integration time, the decrease in correlation coefficient is slowed. If the correlation coefficient is calculated when the signal is weaker, the reliability of calculations decreases, as expected.

Figure 7 shows typical spots on the copper layer. From these images, it can be concluded that the breakthrough of the copper layer is achieved between 20 and 100 shots. From the results, presented in Fig. 6b, it can be seen that the increase in the correlation coefficient of spectra with the pure substrate spectrum starts at about 50 shots (i.e., cor. coeff. with copper of about 0.9) and that correlation coefficients intersect at about 120 shots (i.e., cor. coeff. with copper of about 0.55). The middle value of these two numbers of shots (i.e., 85) corresponds to the coefficient of 0.8. However, in real application, the initial breakthrough of the copper layer will be the time when the laser beam should be moved across the target, or vice versa, i.e., printed circuit board prototype should be moved using 3D motorized stages. In our measurements, with a large number of laser expositions, we have actually obtained mixed spectra, corresponding to the exposed part of the substrate and the still not ablated copper perimeter of the drilled hole. As the number of laser shots increases, more and more of the printed circuit board substrate is exposed and the correlation coefficient decreases. As explained, our estimate is that the laser beam should be moved across the



Fig. 4 LIBS streak images of **a** copper conductor and **b** a printed circuit board substrate

target after the correlation coefficient, calculated in real time by using Eq. (3), decreases below 0.8.

4 Conclusion

We have simultaneously used two monitoring methods: shadowgraphy and LIBS, for the evaluation of laser ablation of copper from printed circuits boards. The shadowgraphic technique was used for measuring the shock-wave dynamics. We have found that the OD energyconversion efficiency for the excitation pulse–substrate interaction is around $\eta_{\rm S} = 11$ %, and it is significantly lower than the OD energy-conversion coefficient, when the ablation takes place on the copper layer ($\eta_{\rm C} = 22$ %). These results prove the concept that the shock-wave dynamics can also be used for monitoring the selective layer removal by laser ablation. Nevertheless, for a real-



Fig. 5 LIBS streak images of printed circuit board **a** at the beginning, **b** after 50 laser shots, **c** after 200 laser shots, **f** after 500 laser shots, and **d** when the substrate is fully exposed. The integrated (as denoted by *dashed lines*, between 0.7 and 1.4 μ s) line profiles are provided in **e**



Fig. 6 The evolution of calculated correlation coefficients as a function of number of laser shots for the following time frames: **a** 0.5–1.2 μ s; **b** 0.7–1.4 μ s; and **c** 1.0–1.7 μ s



Fig. 7 Typical spots on the copper layer. The number of shots is given in the *top-left*-hand side of each image, while the scale is shown on the *bottom-right*-hand side of the first image

time application, the presented results should be used in conjunction with an on-line optodynamic method, e.g., a laser-beam-deflection probe.

The acquired time-resolved LIBS spectra were analyzed by using the linear correlation method with threshold treatment to determine when the laser beam should be moved regarding the ablated sample. We have tried various time frames for integrating the line profiles used in calculations. These evaluations reveal that the steepest decrease in the correlation coefficient between the whole spectrum and the spectrum of the removing layer should be defined as the most appropriate time frame. In our case, it equals to the interval between 0.7 and 1.4 μ s.

Our streak camera system enables time-resolved LIBS analysis, allowing us to determine the time frame where spectra discrimination is best for a given laser excitation (regarding wavelength and energy). Real applications should be based on these results, using much cheaper spectroscopic equipment that is still able to track the correlation coefficient. According to our results, the substrate under the copper layer is reached when this coefficient decreases below 0.8.

References

- A. Gorkič, D. Kovačič, J. Diaci, Int. J. Adv. Manuf. Technol. 42, 138 (2009)
- 2. J. Diaci, J. Možina, Appl. Phys. A 55, 352 (1992)
- 3. T. Tong, J.G. Li, J.P. Longtin, Appl. Opt. 43, 1971 (2004)
- C. Sanchez-Ake, H. Sobral, M.V. Muniz, L. Escobar-Alarcon, E. Camps, Opt. Lasers Eng. 39, 581 (2003)

- R. Petkovšek, P. Gregorčič, J. Možina, Meas. Sci. Technol. 18, 2972 (2007)
- G. Bosiger, T. Perhavec, J. Diaci, Strojniški Vestnik-J. Mech. Eng. 60, 172 (2014)
- P. Gregorčič, J. Zadravec, J. Možina, M. Jezeršek, Appl. Phys. A 117, 353 (2014)
- M. Castillejo, M. Martin, M. Oujja, E. Rebollar, C. Domingo, J.V. Garcia-Ramos, S. Sanchez-Cortes, J. Cult. Herit. 4, 243 (2003)
- A. Ragusich, G. Taillon, M. Meunier, L. Martinu, J.E. Klemberg-Sapieha, Surf. Coat. Technol. 232, 758 (2013)
- F. Bredice, P.P. Martinez, C. Sanchez-Ake, M. Villagran-Muniz, Spectrochim. Acta, Part B 107, 25 (2015)
- T.J. Li, Q.H. Lou, Y.R. Wei, F. Huang, J.X. Dong, J.R. Liu, Appl. Surf. Sci. 181, 225 (2001)
- M.S. Rabasović, B.P. Marinković, D. Šević, IEEE Trans. Plasma Sci. 42, 2588 (2014)
- H. Balzer, M. Hoehne, V. Sturm, R. Noll, Spectrochim. Acta, Part B 60, 1172 (2005)
- P. Pouli, K. Melessanaki, A. Giakoumaki, V. Argyropoulos, D. Anglos, Spectrochim. Acta, Part B 60, 1163 (2005)
- K. Novotny, T. Vaculovic, M. Galiova, V. Otruba, V. Kanicky, J. Kaiser, M. Liska, O. Samek, R. Malina, K. Palenikova, Appl. Surf. Sci. 253, 3834 (2007)
- N.F.C. Mendes, I. Osticioli, J. Striova, A. Sansonetti, M. Becucci, E. Castellucci, J. Mol. Struct. 924, 420 (2009)
- M. Abdelhamid, S. Grassini, E. Angelini, G.M. Ingo, M.A. Harith, Spectrochim. Acta, Part B 65, 695 (2010)
- H.A. Ardakani, S.H. Tavassoli, Spectrochim. Acta, Part B 65, 210 (2010)
- M.P. Mateo, V. Pinon, G. Nicolas, Surf. Coat. Technol. 211, 89 (2012)
- G.S. Settles, Schlieren and Shadowgraph Techniques (Springer, Berlin, 2001)
- 21. P. Gregorčič, J. Diaci, J. Možina, Appl. Phys. A 112, 49 (2013)
- 22. D.L. Jones, Phys. Fluids 11, 1664 (1968)
- 23. G. Taylor, P. Roy, Soc. Lond. A Mat. 201, 175 (1950)