THE FINAL STATUS OF A METAL SURFACE AFTER MULTIPULSE LASER IRRADIATION IN AN AMBIENT GAS

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1. INTRODUCTION AND EXPERIMENTAL

The breakdown plasma is usually initiated in front of solid (metallic) samples at significantly lower incident laser intensities than in the free gas [1-5]. This is the result of a two stages process [6,7]. A certain, generally small amount of substance is expelled in a first stage from the surface of the sample. The avalanche ionization develops starting from these initial centers and the plasma rapidly develops in the mixture of vapors and gas. The ignited plasma keeps in contact during an important part of its life with the solid sample. Accordingly, the plasma-sample interaction leaves a characteristic "touch" on the surface of the sample.

As recently pointed out, the study of the zone on the surface of the sample which was in contact with the plasma is of interest for:

1. the design of the metallic optical elements which are able to stand high intensity radiation which become now available with the last generation technological laser sources;

2. the application of breakdown laser plasmas in front of solid (mainly metallic, but also semiconductor) samples for the synthesis of thin compound layers with utilization in "hot" technological fields like VLSI microelectronics, chemistry, metallurgy a.s.o. [8-10].

3. the efficient laser processing of metallic parts (as e.g. welding, surface hardening, implantation and alloying) [11].

We have therefore approached with this paper the effects caused by laser and plasma actions upon a typical metal target submitted to high intensity multipulse laser treatment in an inert ambient gas at a slightly superatmospheric pressure. There is an unsignificant substance ablation in this regime, an essential requirement for the application of some processing operations (surface hardening, surface doping and alloying, synthesis of surface compound layers with special properties). We choose a pulsed CO2 laser source because at this laser wavelength in mid IR ($\lambda = 10.6 \mu m$), the decrease of plasma initiation threshold in front of a solid sample as against in free gas is among the largest one observed (of hundred times).

A general scheme of the experimental arrangement is represented in Fig. 1. The laser source is operated at a 10 Hz frequency repetition rate. In order to obtain a homogeneous spot, the laser beam is diaphragmed. A part of the laser beam is directed to a fast photon-drag detector. The pulse energy is monitored with a precise joule meter. The typical temporal shape of the pulses (Fig. 2) - which consists of a prominent first peak followed by a long lasting tail of lower intensity- is varied by changing the CO2 / N2 / He active gas mixture. Three different types of laser pulse temporal shapes are selected. They are reproduced in Fig. 2. The maximum value of incident intensity is of $I = 400 \text{ MW/cm}^2$ and is reached with A type (Fig. 2) temporal laser pulse. The irradiation are performed in a stainless steel chamber. The pumping system
ensures the chamber evacuation down to a residual pressure of \(10^{-4}\) Pa. The buffer gas He, \(\text{N}_2\) or \(\text{Ar}\) is introduced into the chamber at a given controlled pressure between 100 Pa and 10 kPa. The samples, of pure (99.9 \%) polycrystalline Ti, have dimensions of 10x20x1 mm\(^3\). Before irradiation, their surface is carefully cleaned.

The samples surface are first examined with an optical stereo microscope and then carefully investigated by Scanning Electron Microscopy (SEM) with the aid of a JEOL 100 CX apparatus.

2. RESULTS

The Laser Treated Zone (LTZ) exhibits a completely differing look - in either optical or electron microscopy - as compared to untreated (virgin) zones. One important remark is that the geometrical shape of LTZ almost entirely coincides with the laser irradiation spot on the surface, even though (as observed visually and by high speed photography) the plasma width considerably exceeds the spot dimension. LTZ has an elliptical shape, characteristic of the laser beam profile. At a low pressure, \(p < 200\) Pa, the central LTZ is encircled by a distinct, well-contrasting "Hallo-Ring" (HR). The HR width increases and the contrast between LTZ and HR improves when \(p\) diminishes and/or the incident laser fluence (\(E_0\)) and the number of subsequent laser pulses applied to the same irradiation site (\(N\)) are increased.

SEM investigation allows a detailed study of the areas which have been in close contact with the laser plasma.

2.1 Laser irradiation performed in helium

A net transition is observed from the regular polycrystalline structure of the virgin surface to a single - crystalline one in the center of LTZ. This evolution is clearly visible in Figs. 3 a, b, where SEM micrographs of areas at the LZT border which were submitted to prolonged (\(N = 4000\)) multipulse laser treatment, are reproduced. They show the same general features even though the two respective multipulse irradiation series were performed in rather different conditions - with \(A\) or \(C\) type laser pulses at atmospheric pressure or at \(p_0 = 5\) kPa.

We thus notice the formation of a smooth surface within LTZ which is sometimes however exhibiting evidence of damages in form of pores or crackings. The HR is particularly distinct at \(p_0 = 5\) kPa (Fig. 3 b) while spherical droplets are much more abundant after the action of the \(A\) type laser pulses in He at atmospheric pressure. At a closer examination, the HR appears to be a mixture of fine polycrystallites and many spherical droplets, sometimes overlapping (Fig. 3).

Pores, with typical diameters in the mm range are a general evidence (Fig. 4 a). Quite extended, very smooth, crystalline area (Fig. 4 b) with dimensions of several tens to several hundreds of micrometers, have been often identified. The largest crystalline areas form as a result of prolonged irradiation (\(N = 2000\)) with C-type laser pulses of low energy values (\(E_0 = 70\) mJ). The further increase of pulse energy causes the extension of fracturing visible from Fig. 4 b.

In case of a prolonged heating of the sample surface - like in He and/or under the action of longer (C type) laser pulses - the formation is favored by extended single-crystalline zones. This evolution which can be exploited e.g. for obtaining a high quality welding operation.

2.2 Laser irradiation performed in \(\text{Ar}\) or \(\text{N}_2\) ambient atmosphere

To the difference of multipulse laser irradiation performed in He, we obtain in \(\text{N}_2\) a transition from the regular (hexagonal) structure of the virgin Ti surface towards an irregular but very well-packed polycrystalline structure within LTZ (Fig. 5). Such a structure is for sure related to a large microhardness value and an excellent adherence of the surface film to the bulk.

The crystallites within LTZ become larger and their geometrical shape is more complicated when multipulse laser irradiation is prolonged in long series.

The increase of the laser pulse energy results in a further complication of the geometrical shape of the polycrystallites within LTZ and in the increase of the spherical droplets concentration.

As a general evidence, LTZ looks much more perturbed after the action of \(A\)-type laser pulses than after the action of \(C\)-type laser pulses, under otherwise identical conditions. Particularly, a significant enhancement of the concentration of spherical droplets is noted.

The most perturbed LTZ's are observed after the action of \(A\) type laser pulses in \(\text{Ar}\) at large pressure (close to the atmosphere). Indeed, as visible from the micrographs reproduced in Fig. 6 a, b, plenty of spherical droplets are observed in this case, spreaded among very fine polycrystallites within LTZ.

LTZ's look changes to one similar to that observed in the case of laser irradiation in \(\text{N}_2\) when the pressure of \(\text{Ar}\) is diminished to 10 - 20 kPa. However, when the pressure of \(\text{Ar}\) or \(\text{N}_2\) is further diminished to 5 kPa or less, a single - crystallization tendency is sometimes observed in these gases, too, even though at much lower extent as compared to He. This behavior is important with the \(C\) type laser pulses.

As a last remark, we note that the HR evidence is hardly detectable after laser multipulse irradiation in \(\text{Ar}\) or \(\text{N}_2\), even at quite modest pressures (5 - 10 kPa).
3. DISCUSSIONS

We first point out the good coincidence between the geometrical shape of the laser irradiation spot on the surface and the aspect of the forming LTZ. This way, the advantage is preserved of an accurate spatial control which stays among the best performances of laser surface treatment.

Next, we observe that HR formation is a result of unidirectional half spherical expulsion of metal vapor and plasma, a situation which is particularly characteristic of irradiation performed in ambient He at pressures below 5 kPa and which stronger manifests under the action of more energetic (C type) laser pulses. It is less encountered with the other two gases, Ar and N₂, where it was observed for very low pressure only.

One important conclusion of our microscopic investigations is that the samples appear to have been always melted in a surface layer. In these conditions, the plasma is always in contact with a liquid layer whose thickness is determined by both, laser irradiation parameters as well as by gas and metal characteristics. This is first proved by the omnipresence of spherical droplets (Figs. 3 a, b) within LTZ or HR, as well as far outside the laser irradiation spot. These droplets are expelled in a liquid phase from the laser irradiation area, then froze during their trip through the ambient gas and, finally fall upon the sample surface during surface cooling.

Secondly, we observe that the formation of rather extended single-crystalline zones (Fig. 4 b) is precisely a result of the liquid phase annealing process. This process appears to be particularly enhanced in He and under the action of C type laser pulses - i.e. under conditions in which the sample surface layer is more intensively heated and submitted to a minimum perturbation by recoil action of the gas breakdown plasma. These conditions are met also at very low pressures of Ar and N₂ - as confirmed by the self-crystallization tendency of LTZ. We notice that the possibility to obtain high-quality and very smooth surfaces by multipulse laser irradiation of metal samples in an inert gas atmosphere opens a way towards new performant processing technologies.

Due to the fast quenching of the surface layer, the large density of pores which are sometimes observed within LTZ (Fig. 4) provides a direct evidence for the intense and permanent gas circulation through the liquid layer covering the sample surface during laser pulse action. The gas is explosively pushed into the liquid by the strong plasma recoil action backwards upon the irradiated area. In some conditions, the pores are not observed as they disappear during the surface layer relaxation by cooling.

A special remark is to be made concerning the very well-packed polycrystalline structures (Fig. 5) forming within LTZ after A type multipulse laser irradiation in high pressure N₂. This points to the efficient synthesis of a compact, hard and adherent nitrided layer.

Investigations of the chemical content within LTZ are in progress.

4. CONCLUSIONS

Different plasma evolution regimes leave different touches backwards upon the irradiated site. The explosive detachment of the breakdown plasma from the irradiated surface - like in Ar - causes an enhanced perturbation of the surface. A "smoother" breakdown plasma-sample interaction can contribute to the efficient synthesis of compound layers, very adherent to the substrate - as in N₂. Finally, in case of a prolonged heating of the sample surface - like in He and/or under the action of longer laser pulses - the irradiated area gets a smooth aspect with extended single-crystalline areas, an evolution which can be exploited to the benefit of a laser processing of good quality.

All experimental evidences which have been pointed out are appropriately accounted for in terms of a model based upon the laser plasma interaction with a melted surface layer covering the sample within the laser irradiated area. A detailed quantitative of this model is in progress.

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REFERENCES


FIGURE CAPTIONS

Fig. 1 Experimental setup
Fig. 2 Three laser pulse temporal shapes used in the experiments
Fig. 3 The border between LTZ and the virgin zones on sample surface after prolonged multipulse irradiation in He. Irradiation conditions:
a. A type pulse shape, N = 4000, Eo = 67 mJ, po = 10^5 Pa
b. C type pulse shape, N = 4000, Eo = 67 mJ, po = 5 kPa
Fig. 4
a. Micrograph illustrating the tendency towards single - crystallization within LTZ and the presence of fine pores. Irradiation conditions:
A type pulse shape, N = 4000, Eo = 40 mJ, po = 20 kPa
b. Single - crystalline area with extended fractures. Irradiation conditions:
C type pulse shape, N = 4000, Eo = 67 mJ, po = 10^5 Pa
Fig. 5 The border between LTZ and the virgin zones on the surface of a sample submitted to prolonged multipulse irradiation in N2. Irradiation conditions:
A type pulse shape, N = 4000, Eo = 67 mJ, po = 10^5 Pa
Fig. 6 The border between LTZ and the virgin zones on sample surface (a) and detail within LTZ (b). Irradiation conditions:
A type pulse shape, N = 4000, Eo = 67 mJ, po = 10^5 Pa of Ar