Peculiar effects in transient plasmas generated by high-fluence laser ablation: from ns to ps to fs regimes

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We present an overview of studies performed in our group over the last decade on the characterization of transient plasma plumes generated by laser ablation in various temporal regimes, from nanosecond to femtosecond. Optical (fast gate intensified CCD camera imaging and space- and time-resolved emission spectroscopy) and electrical (mainly Langmuir probe) methods have been applied to experimentally investigate the dynamics of the plasma plume and its constituents. Peculiar effects as plume splitting and sharpening or oscillations onset have been evidenced at high laser fluence. New theoretical approaches have been developed to account for the experimental observations. Aside these fundamental studies, we will present two examples of applications, in plasma space propulsion and growth of thin films by pulsed laser deposition.

1. Introduction

Understanding the complex processes triggered by the pulsed laser interaction with the condensed matter is key for the development and optimization of a huge number of applications in various fields, from nuclear fusion [1] to precision micromachining [2] and nanoscale synthesis of high technological potential materials [3], to analytical sciences [4] or medicine [5]. The difficulty in unveiling this complexity arises (at least partially) from the multi-physics nature of the ablation process, coupling optics (absorption of light by the target material), thermodynamics (heating, phase transitions, cooling), gas dynamics (expansion of ablation plume into vacuum or background gas), plasma physics (collisions, electric interactions, if the plume reaches high-enough ionization) and laser – plume interaction (plasma heating by absorption of laser photons), some of these evolving on very short time scales, which can make it challenging to find the adequate resolving probe. Moreover, the specific details of all stages listed above are highly dependent on the target material and the laser parameters (e.g. laser-plume interaction is absent for femtosecond ablation pulses), making arduous (if not impossible) the task of drawing a unified scenario, at both experimental and theoretical levels [6].

In this frame, our groups have performed systematic experimental studies on the characterization of plasma plumes generated by laser ablation in various temporal regimes (ns, ps, fs) on materials ranging from simple metals (Cu, Al, …) to more complex compounds (ceramics, chalcogenide glasses, ferrites) [7-23]. Some new theoretical approaches [10, 13-15, 17, 19, 23] have been proposed to explain the experimental findings. An overview of these works will be presented.

2. Experimental and theoretical methods

The experimental set-up used for most of the studies at the University of Lille, France, has been described in detail in our previous papers [7-23]. Briefly, the solid targets were placed in high vacuum or controlled atmosphere and irradiated by ns, ps or fs lasers at various wavelengths (mostly 532 nm Nd:YAG and 800 nm Ti:Sa). We used laser fluences spanning the 10^{-1} – 10^{3} J/cm^2 range, corresponding to irradiances in the 10^{6} – 10^{14} W/cm^2 limits. As electric diagnostics, we used mainly Langmuir probes (individual or bunches) to measure the ionic and electronic currents at various positions in the plasma. Transient voltages on the ablation target and deposition substrate (in PLD configuration) were also recorded. Fast gate ICCD imaging was used to monitor the evolution of the plasma plume structures, while space- and time-resolved optical emission spectroscopy provided information on the individual excited components in the plume. Some experiments were performed on the national ground facility PIVOINE 2G at ICARE Institute (CNRS, Orléans, France), in the frame of the French National Network "Plasma Propulsion in Space".

The new theoretical method [10] proposed to describe the expansion of the plasma plume is based on a fractal hydrodynamic model mainly developed at the Technical University "Gh. Asachi" of Iasi, Romania. More “classical” approaches (plasma ion frequency, electron-ion collision damping [15]) were also proposed to tentatively explain the oscillatory behaviour experimentally observed.

3. Results

The electrical and optical investigations performed revealed a splitting of the ablation plume in (at least) two structures above a material-dependent laser fluence threshold. This behaviour had been already reported both experimentally [24, 25] and theoretically [25], and had been attributed to the interaction between the plasma plume and the background gas. Note however that in our case this occurs even in high vacuum. The splitting behaviour is well reproduced by the new fractal hydrodynamic model [10]. Center-of-mass velocities measured from fast-gate ICCD snapshots were in the range of 10^4 m/s for the first (fast) structure and 10^3 m/s for the second (slow) one.

In order to get insight on the dynamics of individual species present in the plume, we performed a space- and time-resolved optical emission spectroscopy study. In most cases, this study revealed a significant difference between the velocities of ions and neutrals, with the former being concentrated in the fast structure and the latter in the slow one [11]. This led us to the conclusion that the ions would be ejected on a very short timescale through a Coulomb process in the very intense field left by electron laser excitation and detachment, while the neutrals would come from a subsequent thermal process (e.g. phase explosion [26], which needs more time to establish [27]).

Excitation temperatures have been derived from the spectral lines intensities using the well-known Boltzmann plot method, in the assumption of local thermodynamic equilibrium. Electron density has been calculated using the Saha-Eggert equation in the same conditions. These studies revealed the presence of two populations (“hot” and “cold” electrons). Space- and time-resolved Langmuir probe I-V characteristics returned temperature and number density values which corroborated the optical observations.

The oscillations initially observed in the Langmuir probe current (and further confirmed by target and substrate transient potential and optical emission measurements) appeared as highly surprising to the community. However, similar behaviour had been observed already in the late ‘80s by Borowitz et al. [28] using an 100 J Nd:YAG laser (I ~ 10^{15} W/cm^2). They explained the occurrence of the oscillations by the formation of a double layer at the target surface. Using similar irradiance values (ns iodine laser of 45 J), Laska et al. [29] recorded multi-peak structures in the time-of-flight current profile of a plasma generated from Au target, but they attributed this to multiply charged ions. Oscillations were also observed at lower irradiance (10^8 W/cm^2), but they seem to have been overlooked in favour of prompt electrons emission [30]. Gilton et al. [31] evidenced oscillations in the photoemission current at irradiances as low as 10^5 W/cm^2, and also explain them through the occurrence of a space charge layer in the very vicinity (a few μm) of the target. More recently, two works [32, 33] reported the observation of oscillations in multi-component plasmas generated by high-fluence laser ablation (18 - 40 J/cm^2, I ~ 10^9 W/cm^2). In the first study [32] a simple Langmuir probe was used, and the authors interpreted the multiple peaks in the ionic current temporal profile in terms of mass-dependent expansion velocities of different elements present in the plume. However, when using a triple probe to simultaneously measure the ionic current and the floating potential in the second study, their conclusion was that “… analysis confirmed that the observed oscillations are the genuine propagating plume fluctuations” [33]. Finally, Singh et al. [34] reported ion flux enhancements and oscillations in spatially confined laser-produced aluminum plasmas and revisited our Josephson effect [10] and collisional [15] arguments to explain the occurrence of oscillations and to account for their frequency. We therefore consider that there is significant experimental and theoretical evidence for the existence of the oscillatory behavior in laser-produced plasmas.

A systematic study has been performed on pure metal targets in order to investigate the influence of various experimental parameters (laser fluence, pulse duration, wavelength, background pressure, target atomic mass, target-probe distance, target bias etc.) on the plasma plume oscillations. Preliminary results of this extensive study will be presented at the conference.

4. References

[1] R. K. Kirkwood, J. D. Moody, J. Kline, E. Dewald, S. Glenzer, L. Divol, P. Michel, D. Hinkel, R. Berger, E. Williams, J. Milovich, L. Yin, H. Rose, B. MacGowan, O. Landen, M. Rosen, J. Lindl, *Plasma Phys. Control. Fusion* **55** (2013) 103001.

[2] K. Sugioka, M. Meunier, A. Piqué (*eds.)*, *Laser Precision Microfabrication*, Springer 2010.

[3] C. Phipps *(ed.)*, *Laser Ablation and its Applications*, Springer 2007.

[4] R. E. Russo, X. Mao, J. J. Gonzalez, V. Zorba, J. Yoo, *Anal. Chem.* **85** (2013) 6162.

[5] D. Bäuerle, *Laser Processing and Chemistry*, 4th ed., Springer 2011, pp. 727-733.

[6] S. I. Anisimov, B. S. Luk'yanchuk, *Phys. – Usp.* **45** (2002) 293.

[7] S. Gurlui, M. Sanduloviciu, M. Strat, G. Strat, C. Mihesan, M. Ziskind, C. Focsa, *J. Optoelectron. Adv. Mater.* **8** (2006)148.

[8] S. Gurlui, M. Sanduloviciu, C. Mihesan, M. Ziskind, C. Focsa, *AIP Conf. Proc.* **812** (2006) 279.

[9] C. Focsa, M. Ziskind, C. Ursu, S. Gurlui, D. Pagnon, S. Pellerin, N. Pellerin, M. Dudeck, *J. Optoelectron. Adv. Mater.* **10** (2008) 2380.

[10] S. Gurlui, M. Agop, P. Nica, M. Ziskind, C. Focsa, *Phys. Rev. E* **78** (2008) 026405.

[11] C. Ursu, S. Gurlui, C. Focsa, G. Popa, *Nucl. Instrum. Meth. B* **267** (2009) 446.

[12] C. Focsa, P. Nemec, M. Ziskind, C. Ursu, S. Gurlui, V. Nazabal, *Appl. Surf. Sci.* **255**, (2009) 5307.

[13] P. Nica, P. Vizureanu, M. Agop, S. Gurlui, C. Focsa, N. Forna, P. Ioannou, Z. Borsos, *Jpn. J. Appl. Phys.* **48** (2009) 066001.

[14] M. Agop, P. Nica, S. Gurlui, C. Focsa, *AIP Conf. Proc.* **1278** (2010) 612.

[15] P. Nica, M. Agop, S. Gurlui, C. Focsa, *Eur. Phys. Lett.* **89** (2010) 65001.

[16] C. Ursu, O. G. Pompilian, S. Gurlui, P. Nica, M. Agop, M. Dudeck, C. Focsa, *Appl. Phys. A* **101** (2010) 153.

[17] M. Agop, P.E. Nica, S. Gurlui, C. Focsa, V. P. Paun, M. Colotin, *Eur. Phys. J. D* **56**, (2010) 405.

[18] S. Gurlui, C. Focsa, *IEEE T. Plasma Sci.* **39** (2011) 2820.

[19] P. Nica, M. Agop, S. Gurlui, C. Bejinariu, C. Focsa, *Jpn. J. Appl. Phys.* **51** (2012) 106102.

[20] L. Balika, C. Focsa, S. Gurlui, S. Pellerin, N. Pellerin, D. Pagnon, M. Dudeck, *Spectrochim. Acta B* **74-75** (2012) 184.

[21] O. G. Pompilian, S. Gurlui, P. Nemec, V. Nazabal, M. Ziskind, C. Focsa, *Appl. Surf. Sci.* **278** (2013) 352.

[22] L. Balika, C. Focsa, S. Gurlui, S. Pellerin, N. Pellerin, D. Pagnon, M. Dudeck, *Appl. Phys. A* **112** (2013) 123.

[23] S. Irimiciuc, M. Agop, P. Nica, S. Gurlui, D. Mihaileanu, S. Toma, C. Focsa, *Jpn. J. Appl. Phys.* **53** (2014) 116202.

[24] S. S. Harilal, C. V. Bindhu, M. S. Tillack, F. Najmabadi, A. C. Gaeris, *J. Appl. Phys.* **93** (2003) 2380

[25] A. V. Bulgakov, N. M. Bulgakova, *J. Phys. D* **31** (1998) 693.

[26] A. Peterlongo, A. Miotello, R. Kelly, *Phys. Rev. E* **50** (1994) 4716.

[27] J. H. Yoo, S. H. Jeong, R. Greif, R. E. Russo, *J. Appl. Phys.* **88** (2000) 1638.

[28] J. L. Borowitz, S. Eliezer, Y. Gazit, M. Givon, S. Jacket, A. Ludmirsky, D. Salzmann, E. Yarkoni, A. Zigler, B. Arad, *J. Phys. D: Appl. Phys.* **20** (1987) 210.

[29] L. Laska, J. Krasa, K. Masek, M. Pfeifer, K. Rohlena, B. Kralikova, J. Skala, E. Woryna, P. Parys, J. Wolowski, W. Mroz, B. Sharkov, H. Haseroth, *Rev. Sci. Inst.* **71** (2000) 927.

[30] S. Amoruso, M. Arenante, R. Bruzzese, N. Spinelli, R. Velotta, X. Wang, *Appl. Phys. Lett.* **75** (1999) 7.

[31] T. L. Gilton J. P. Cowin, G. D. Kubiak, A. V. Hamza, *J. Appl. Phys.* **68** (1990) 4802.

[32] S. Sunil, A. Kumar, R. K. Singh, K. P. Subramanian, *J. Phys. D: Appl. Phys.* **41** (2008) 085211.

[33] A. Kumar, R. K. Singh, J. Thomas, S. Sunil, *J. Appl. Phys.* **106** (2009) 043306.

[34] S. C. Singh, C. Fallon, P. Hayden, M. Mujawar, P. Yeates, J. T. Costello, *Phys. Plasmas* **21** (2014) 093113.