BULETINUL INSTITUTULUI POLITEHNIC DIN IAŞI
Publicat de
Universitatea Tehnică „Gheorghe Asachi” din Iaşi
Volumul 63 (67), Numărul 4, 2017
Secţia
MATEMATICĂ. MECANICĂ TEORETICĂ. FIZICĂ

LASER ABLATION: STATE OF THE ART AND PERSPECTIVES

BY

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Received: November 20, 2017
Accepted for publication: December 12, 2017

Abstract. Laser ablation community has seen a strong development in the past 20 years. This is related with the technological development of shorter laser beams and more flexible diagnostics systems. In this paper we focus on the fundamental mechanisms of laser ablation and state of the art in the matter of diagnostics methods and the complete picture that we now have over the laser ablation process and laser produced plasma dynamics.

Keywords: laser produced plasmas; fundamental ablation mechanism; Coulomb explosion.

1. Short History of Laser Ablation

Laser ablation is defining a series of processes that are the results of a laser beam impinge onto a solid surface. The effects of “photonic ablation” (or the interaction of photons with the matter) have been tentatively known for centuries. Some example can be found even in the Greek literature, when in 303 B.C. are presented the properties of a globe filled with water that can light fire, regardless of the environmental conditions. Also, the concept of “photonic ablation” is even mentioned by Archimedes, who proposed in 203 B.C. to reflect and focus the sunlight on the Phoenicians attacking the city using an array of mirrors (Miller, 1994).

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The “modern” history of the laser ablation started with a series of conference papers and talks from the 1960’s. The results reported in those papers covered a series of fundamental aspects that are considered as the pillars of laser ablation and laser-produced plasmas which led to the development of an entirely new research direction. The first recorded “regular” paper was a theoretical study of Askar'yan and Moroz (1963), where they made some calculations regarding the recoil pressure during the laser ablation of a solid target and discussed the acceleration of small particles or droplets in the framework of an "one-sided evaporation" model. They also predicted the presence of ultrasonic and hypersonic oscillations produced by modulated laser ablation. Following, in a similar pioneering experimental drive, Honig and Woolston (1963) reported some results from the investigation of laser ablation of various targets (metals, semiconductors and dielectrics). They reported for the first time a quantitative measurement of the ejected particles (3 x 10^{16} electrons and 10^8 positive ions per m^3). The published paper presented the first detailed study of the electron emission and its temporal profile. They analyzed the mass distribution with a modified commercial, double-focusing mass spectrometer, thus demonstrating the first use of the ion microprobe analysis. This study will further be the basis for ion mass spectrometry and paved the way for electrical investigations of LPPs. In later papers Lichtman and Ready (1963), using a simple assumption of thermionic emission, derived the temperature of the surface during laser-target interaction, finding values of about 3300 K for a ruby laser interaction with a tungsten target. Ready (1963) proved for the first time the implementation of high-speed photography as a viable method to study the temporal and spatial profiles of the plume of ejected material. The paper reported on a carbon laser ablation plasma. One of the main results presented in that paper was that the emitted light from the plasma reached its maximum at about 120 ns after the start of the laser pulse and had an estimated life-time of a few microseconds. From here, the expansion velocity of the plume was estimated as being of 20 km/s. Follow-up studies on carbon-based targets were performed by Howe (1963), who reported on the energy of the ejected particles by means of the vibrational (0.86 - 1.72 eV) and rotational (0.38 eV) temperatures extracted from fluorescence spectra of CN and C2. This represents one of the first mentioning of possible non-equilibrium conditions that were attributed to the cooling of the ejected particles during an adiabatic expansion. This subject was further investigated by Berkowitz and Chupka (1964), who observed, after post-ionization of the ablated plume, cluster ions of carbon (n ~ 14), boron (n ~ 5) and manganese (n ~ 2). Exploring the production of large structures during laser ablation, there have been reports (Neuman, 1964) of large “blobs of molten material” and “fragments of material” suggested by the first momentum transfer measurements. This short period of time is characterized by a fast expansion of laser-matter interaction and related topics, during which the first reported papers concentrated on the study of various properties of the ejected particles (electrons, ions, neutrals, clusters and emitted photons). This, coupled with the first estimations of plasma temperatures, velocities and densities, led to the formation of a coherent image of the complex processes involved in the laser-matter interaction.

In the following years there was a "boom" of articles focused on fundamental investigations of laser produced plasmas performed over a wide range of laser characteristics (beam power, pulse width, repetition rate etc.). The development of the laser technology and the measurement techniques led to more sophisticated experiments and more comprehensive theoretical models. Exciting results began to arise due to the new means of study, such as visible, ultra-violet and X-ray emission measurements (Ehler and Weissler, 1966; Benavides *et al*., 2016), coupled with the findings of multiply-charged ions (Archbold and Hughes, 1964) and two- and three-photon emission (Sonnenberg *et al*., 1964). All these achievements and findings led to the development of new applications that were proposed as alternatives to the already existing ones.

In 1964, Berkowitz and Chupka (1964) proposed for the first time the laser ablation technique as an alternative to fusion, thus arose the idea of laser confinement fusion. Another spectacular application that was born was the Pulsed Laser Deposition (PLD), as a response to the already existing sputtering techniques. Smith and Turner (1965) reported the first representative experiment of PLD. Although the authors experimented on a variety of materials using a ruby laser, the quality of the resulted thin films was secondary to the ones produced by sputtering. Not until the 80's was laser film growth able to compete with the other well established deposition techniques, when Dijkkamp *et al*. (1987) deposited a high quality thin film of YBa2Cu3O7. Since then, the PLD technique has been used to successfully produce thin films with a wide variety of properties, amongst which a series of thin films with high crystallinity (ceramic oxides, nitrides, metallic multilayers) (Eason, 2007; Craciun *et al*., 1994, 2005; Perriere *et al*., 2002). The main advantages of PLD are the relatively low costs, with respect to molecular beam epitaxy (MBE) or metal-organic chemical vapor deposition (MOCVD), and the better control over stoichiometry and phase composition, which is very beneficial regarding the growth of complex materials, including high-quality nanomaterials that are impossible to synthesize otherwise. Some of the main successes of the technique can be summed up by the type of complex target resulted (nanowires of Si and Ge (Morales and Lieber, 1998), binary (In_2O_3 (Li *et al*., 2003), SnO_2 (Liu *et al*., 2003), ZnO (Yang *et al*., 2006)) and ternary systems ($GaAs_{0.6}P_{0.4}$, $InAs_{0.5}P_{0.5}$, CdS_xSe_{1-x}, indium tin oxide (Savu and Joanni, 2006)), and more complex materials (Eisenhawer *et al*., 2011).

As the thin-film deposition technology flourished, the growing reliability and stability of commercial lasers, particularly Q-switched YAG lasers, improved the uniformity of film growth and the reproducibility of microprobe measurements. Significant progress was made, simultaneously, on the fundamentals aspects of the deposition process. This was achieved through plume diagnostics and the development of theoretical models. The pulsed laser deposition process is a complex one. This complexity comes also from the correlations between a series of variables like: target composition, laser characteristics as fluence, wavelength or pulse width, background gas species, substrate's physical properties, overall PLD geometry etc. Changing one parameter often shifts the ideal settings for others. The effects of changing a single variable can be identified by keeping all other variables constant, and variables are generally kept constant for simplicity. Due to this network of inter-relationships, the control of the deposition process becomes complicated, as well as the overall understanding of the LPP dynamics and how the properties of the plume can influence the final product. This image can be somehow simplified. Let us observe the deposition process from three different perspectives based on the possible influencing factors. One perspective covers the interactions between the laser beam and the target, governed by the physical properties of the target (reflectivity, thermal/electrical conductivity, heat of vaporization, etc.) and those of the laser beam (wavelength, pulse width, shape, etc.) (Zavestovskaya *et al*., 2008; Benavides *et al*., 2016). A second perspective will describe the relationships between the physical properties of the target and the properties of the laser produced plasmas (Williams *et al*., 2008; Hermann *et al*., 2012) and the third one the influence of the ejected particles on the properties of the resulted thin film. In order to achieve some knowledge on any of these dependences it is imperative to use well-established investigation techniques (OES, ICCD fast camera imaging, Langmuir probe method, mass spectrometry, etc.) in order to find a unification relationship between all these "variables".

The benefits of the proper use of the investigation techniques further led to the discovery of other spectacular results. Splitting of the plume is one of them and it was first reported by Geohegan's group (Geohegan and Puretzky, 1995, 1996), when investigating the dynamics of LPP in an ambient gas. This group also proposed a theoretical description based on multiple-scattering and hydrodynamic approaches (Leboeuf *et al*., 1996). The plume splitting has been further confirmed and studied by other groups (Harilal *et al*., 2002, 2003; Wu *et al*., 2013). All these results were obtained in typical PLD experimental conditions, *i.e*. fluence in the range of 1 J/cm^2 and background gas pressure of 1-100 mbar. We emphasize that similar results were also reported for laser ablation in vacuum (background pressure < 10-5 mbar) and at fluences typically higher than 10 J/cm^2 (Gurlui *et al*., 2006, 2008, 2009; Ursu *et al*., 2009). From a theoretical perspective the plume splitting is seen at the results of two distinct mechanisms for the particle ejection (Kelly and Miotello, 1997 Nica *et al*., 2009, 2010; Pompilian *et al*., 2014; Yoo *et al*., 2000; Ursu *et al*., 2009): the ions would be ejected on a very short time scale through a Coulomb process in the very intense field left by the electrons laser excitation and detachment, while the neutrals would come from a subsequent thermal process (phase explosion (Kelly and Miotello, 1998)) which needs more time to establish (Yoo *et al*., 2000).

Besides the overall dynamics of the plume, looking closely to the individual dynamics of the ejected charged particles, an oscillatory behavior was observed. The first reports of plasma oscillations were published in the 1980's. Borowitz *et al*. (1987) recorded a fast oscillation structure on the target current of about 100 ps period when irradiating with an 100 J, ns laser beam (fluence up to 10^5 J/cm^2). The first attempts for the comprehension of this "peculiar" behavior were based on the formation of single or multiple double-layers in the very vicinity of the target. This picture was the main focus to a long series of papers reporting on charge separation in laser-produced plasma, mainly from the 1970 – 1980 (Pearlman and Dahlbacka, 1977; Ludmirsky *et al*., 1984, 1985). Eliezer (1989) gathered in a very comprehensive manner the state of the art regarding the double and multiple layers in laser-produced plasmas. One of the remarkable results reported are experimental proof with double-layer electric fields of 10^5 – 10^6 V/cm and widths of 10-100 Debye lengths (Eliezer and Ludmirsky, 1983).

In this short introduction, it was attempted to review a few "firsts". All the results are building blocks as the techniques implemented in the 60's led to the development of new theoretical models and new aspects of the laser-produced plasmas never seen before. The history of laser ablation is full of "firsts": the first optical emission spectroscopy measuring led to the development of the Laser-Induced Breakdown Spectroscopy (LIBS) technique, the first measurements of the ionic energy distribution led to the further development of mass spectrometry or the first picture of the ejected material foreshadowing the development of the ICCD fast camera imagining method. These are pillars on which was built the image that we have today of laser ablation process as a whole. Now we can see the effects of all these great moments in laser ablation history, the rise of fast camera photography paved the way for the plume splitting effects while the probing of the charged particles led to the observation of plasma oscillations.

2. Laser Ablation in Various Temporal Regimes

With the aim of reaching the enthusiastic objectives presented above, we have to understand what can connect the data provided by the typical LPP measurements with the physical properties of the targets. All the available techniques study the ejected particle during expansion (Geohegan, 1992; Lunney *et al*., 2007; Ursu *et al*., 2010), thus all the investigations will be performed post laser - target interaction. Henceforth, our main goal becomes to look at the plasma during its expansion and then relate that to the initio properties of the electrons and ions in the target. This aspect becomes essential, as in order to understand the connection between the target and the LPP it is imperative to assume that the ejected particles and the information that they carry contain the “memory” embedded with the properties of the target. The memory of the ejected particles is mediated through the ablation mechanisms involved with respect to each ablation regime. Since the thesis is focused on the study of laser-produced plasmas in various ablation regimes (ns, ps, fs), we will attempt in the following to present the main mechanisms that manifest at various temporal regimes. In Fig. 1 (Rethfeld *et al*., 2004) there are presented as examples different processes that take place after the laser energy is absorbed by the lattice of the target. We notice some considerable differences between the ablation regimes used in this thesis. For the fs regime (fs ~ 1 ps) there are mainly non-thermal processes involved which end with the Coulomb explosion as the main ejection mechanism (Shirk and Molian, 1998). For the ps regime, if the pulse width is higher than ~ 10 ps, the thermal mechanisms are becoming predominant, starting with the thermal damage of the lattice (homogeneous melting), if the pulse is shorter, there is defined a transition area between pure non-thermal and thermal effect where Coulomb explosion (Bulgakova *et al*., 2005) is still the main ablation mechanism and the thermal effects are reduced. In the ns regime, the longer pulse width leads to strong thermal effects followed by the subsequent laser beam absorption by the ejected particles (Mao *et al*., 2013). Here, mechanisms like Coulomb explosion are secondary to the thermal ones.

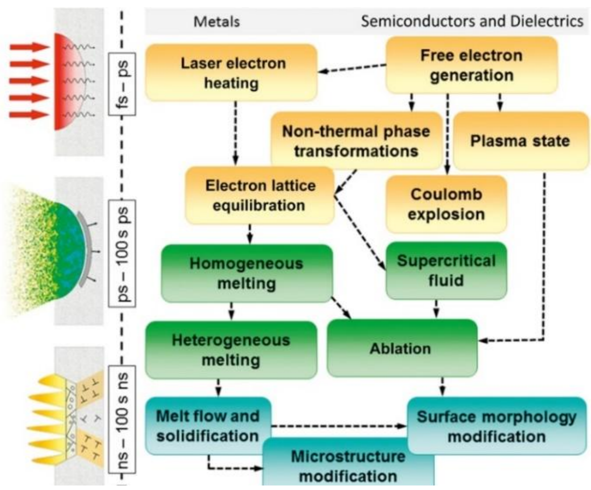

Fig. 1 – Mechanisms involved in various laser ablation regimes (Rethfeld *et al*., 2004).

The simplest way to describe the ablation process is to divide it into four stages. In the first stage are included all the laser-target interactions such as laser absorption by the target electrons or target heating (Fig. 1). Here we can see the manifestation of the ablation mechanisms. In the second stage, the particles are ejected from the target and the plume forms. During this stage, in the ns ablation regime and for the ps regime (if the pulse width is higher than 10 ps) the plume absorbs a part of the laser energy (the absorption of the laser radiation is done through single-photon processes (Tokarev *et al*., 1995), mainly by Inverse Bremsstrahlung (IB) effect (Mao *et al*., 1996)). Also, at this stage, the laser-plasma interactions are dominant. For the fs and ps (1 - 10 ps) regimes this stage does not exist because the plume expansion occurs after the laser pulse has ended. The third stage occurs after the laser pulse has ended. At this stage, in vacuum conditions, the plume is expanding adiabatically (Doggett and Lunney, 2011). The expansion of the plume differs with respect to the background gas conditions, which leads to the fourth stage that describes the plume dynamics if the ablation takes place in a background gas. After the development of the three stages the plume expansion is dictated by the interactions between the plume particles (ions, atoms, electrons, clusters) and the background gas particles.

Understanding the main processes involved in laser-matter interaction is important for their fundamental relevance and also for comprehending the capabilities and limitations of laser-based applications and technologies. The last ablation stages are used for the diagnosis purposes and where the main "bulk" of information is extracted from the plasma, but before we can delve into studies related to the dynamics of the ejected particles it is essential to emphasize some fundamental aspects of the first stage. Generally, when a laser beam impinges on the material, laser energy is first absorbed by free electrons (Mao *et al*., 2013). The absorbed energy then propagates through the electron system and is transferred to the lattice (Mao *et al*., 2013). In literature are identified three characteristic time scales: Te – the electron cooling time, which is on the order of 1 ps; Ti – the lattice heating time (~ 10 ps); and Tl – the duration of laser pulse.

Let us first consider a general case covering a range of pulse widths from continuous wave (cw) laser to ms pulsed laser, Tl (~ ms) >> Ti >> Te. The typical time scale is much larger than the electron-lattice energy coupling time, and thus the main processes involved will be the melting and the subsequent ejection of the molten material assisted by the particle gas (Phipps, 2007). This ablation regime is completely described by the classical heat transfer laws which are often used for the modeling of the laser ablation process in the ms regime. Due to the particularities of the ablation mechanism, this regime is often used in applications like laser cutting, which covers a wide range of materials (steel, nonferrous metals, and nonmetals). For shorter ablation regimes (~ ns) a second case arises: Tl (~ ns) >> Ti >> Te. In this case, electron absorbed laser energy has enough time to be transferred to the lattice, electrons and lattice can reach thermal equilibrium, and the main energy loss is the heat conduction into the solid target. Therefore, the target is melted, followed by evaporation occurring from the liquid state (Kelly and Miotello, 1998). Usually, the heat affected zone is smaller than that of the cw laser processing. These properties make ns-laser ablation a powerful tool for technological applications like laser drilling (Lawrence, 2010), grooving, marking, or scribing. Nevertheless, the presence of a melted layer makes precise material removal rather difficult. In this time scale, the typical lasers used are Q-switched solid state lasers, such as the Nd:YAG laser (1024 nm - 266 nm). Another case corresponds to the ultra-fast laser ablation, Tl << Te << Ti, where Tl is on the femtosecond scale, and laser pulse duration is shorter than the electron cooling time. The electrons in the surface layer undergo cooling by heat diffusion and by heat transfer to the lattice ions. This stage continues for several picoseconds. The picture changes in the case of a semiconductor target that is heated by an ultrashort pulse. The laser energy is deposited into the solid by creating a "bath" of hot electrons and holes (Shirk and Molian, 1998). Hot carriers subsequently transfer energy to the lattice by creating optical and acoustic phonons. In the case of both metals and semiconductors, the thermalization of laser energy in the hot carrier bath takes place within a few femtoseconds ($\approx$ 10 fs), while the typical time-scale for lattice heating falls within the 1 – 10 ps range, where thermal conduction is totally negligible (Leitz *et al*., 2011).

So far, in the literature the majority of the experimental investigations have been carried by Ti: Sapphire laser systems with variable pulse widths. The same systems are often used for the case of ps laser ablation. There, although the pulse width is much shorter than the typical thermal conduction time (hundreds of ns), the laser pulse duration is of the same order as the hot carriers-lattice relaxation time (few ps).

If we analyze in depth the particularities of the short and ultra-short laser ablation we can differentiate between several ablation regimes. Considering the strong differences between the various temporal regimes (ns, ps, fs), it is of the utmost interest to investigate the laser-produced plasmas in those regimes and try to comprehend how the fundamental mechanism affects the properties of the ablation plasmas. Moreover, the comparative study amongst different targets with different physical properties could allow us to understand the connections that can be made in each specific ablation regime and try to correlate with the physical processes involved in the material removal.

3. Laser Ablation Mechanisms

In the literature (Miller, 1994; Phipps, 2007; Lawrence, 2010; Stafe *et al.*, 2014) the mechanisms are generally divided into two main categories. There are the primary mechanisms that are involved only in the removal of the target material (*Normal Vaporization*, *Normal Boiling*, *Phase Explosion* and *Coulomb Explosion*) and the secondary mechanisms meant to describe the behavior of the expelled particles after the interaction between the laser beam and the target surface took place (*Knudsen-Layer Processes*, *Effusion-Like Release* and *Normal Outflow*). A better way we can differentiate between the ablation mechanisms is by the type of processes involved. Using these criteria, we can attribute mechanisms for each ablation regime. Thus, we find thermal (Normal Vaporization, Normal Boiling and Phase Explosion) and electrical (Electronic Processes, Coulomb Explosion) mechanisms. For the nanosecond or longer pulse lasers the thermal mechanisms are dominant and for the picosecond or shorter the electrical ones are dominant. Also, for the ultrafast lasers the interaction time between the laser beam and the target is shorter and as a result the mechanisms involved in the removal of the target particles can be different from the ones involved in the nanosecond laser ablation.

3.1. Normal Vaporization and Normal Boiling

Normal vaporization is a term which describes a group of processes having in common a thermal origin. This mechanism does not have a dependence on the laser fluence or pulse length, thus the main dependence is on the properties of the target. The term “thermal” is not accurate enough to describe this process, as it was reported by (Mele *et al.*, 1997), due to the fact that the temperature (which is usually a measure of a system at equilibrium) is transient, they suggested that a more accurate term would be “thermal spike”. As a result of laser-matter interactions, the atoms, electron, ions, etc. are ejected from the outer layer of the surface. Due to the short interaction time vapor bubbles do not form at the surface, nor from layer beneath the surface. In this scenario, the particle flux emitted can be described by the Hertz-Knudsen equation:

$$Particle\ flux = \alpha(p_{sv} - p_v)(2\pi m k_b T)^{-\frac{1}{2}}(particles/s\ cm^2) \tag{1}$$

where α is the vaporization coefficient (Anisimov *et al.*, 1974), p_{sv} – the saturated vapor pressure, p_v – the vapor pressure, m – the particle mass, T – the surface temperature and k_B – the Boltzmann constant. By multiplying Eq. (1) by m / ρ ($\equiv \lambda^3$), where ρ is the mass density and λ is the length of the atomic bond, then it will define the velocity of the surface recession in a 1-D situation:

$$\left.\frac{\partial x}{\partial t}\right|_{x=0} = \alpha(p_{sv} - p_v)(2\pi m k_B T)^{-\frac{1}{2}}\lambda^3\ (cm/s) \tag{2}$$

$$= \alpha\left[p_b e^{\frac{\Delta H_v m}{k_B}\left(\frac{1}{T_b}-\frac{1}{T}\right)} - p_v\right](2\pi m k_B T)^{-1/2}\lambda^3 \tag{3}$$

where ΔH_v is the heat of vaporization, assuming there is no re-condensation (Yoo *et al.*, 2000), p_b is the boiling pressure and T_b the boiling temperature. A second type of thermal spike often reported in the literature requires a longer pulse length, long enough for the heterogeneous nucleation of the vapor bubble (Dell'Aglio *et al.*, 2015) to occur. If p_v is higher than p_b the "normal boiling" will occur. However, the density of nucleation sites is rather low (Yoo *et al.*, 2000), this means that although the necessary conditions are met, the main ablation mechanism still remains normal vaporization.

3.2. Phase Explosion

In order for the phase explosion to manifest itself, it requires high laser fluences and relatively short pulse widths (ns, ps, fs) (Schittenhelm *et al.*, 1996). In literature are reported some specific thresholds for this ablation mechanism, related to the laser beam properties and the physical properties of the target (mainly the laser wavelength and the binding energy of the target lattice ions) (Russo *et al.*, 2000; Yoo *et al.*, 2000). The main result that can be quantified using phase explosion as an ablation mechanism is the overall quantity of the ablated mass per pulse. From an experimental point of view the quantity of ablated mass can be also correlated to the depth of the crater made by the laser beam (Fig. 2).

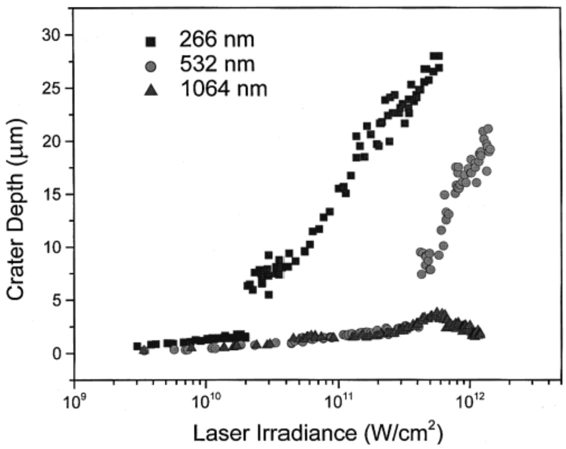

Fig. 2 – Dependence of the crater depth on the incident laser wavelength and irradiance (Schittenhelm *et al.*, 1996).

If the laser beam fluence is high enough, above the threshold, the surface will reach a temperature of ~ $0.9 \cdot T_c$ (T_c is the thermodynamic critical temperature). Close to the thermodynamic critical temperature the vapor nucleation rate raises (Martynyuk, 1977), because the necessity of the nuclei formation is no longer a kinetic obstacle. It was shown that the formation rate of nuclei has a big variation from 10^{-25} cm^{-3}s^{-1} to 10^{25} cm^{-3}s^{-1} when the temperature increases from 0.88 T/T_c to 0.92 T/T_c (Yoo *et al.*, 2000). Also, due to the high temperature, the nucleation of the homogenous vapor bubbles occurs and these vapor bubbles reach a critical size. The size of the bubbles is characterized by the critical radius, with the bubbles having a lower radius most probably collapsing. Generally speaking, the target changes its state from superheated liquid to mixture of liquid droplets and vapors. In the end, the bubbles explode and the particles and the clusters are ejected. The presence of the phase explosion is followed by an increase in the quantity of mass removed, as it was reported by Yoo *et al*. (2000). The increase in the mass removed will lead to an increase of the crater (Yoo *et al.*, 2000) created by the laser pulse as it is shown in Fig. 3.

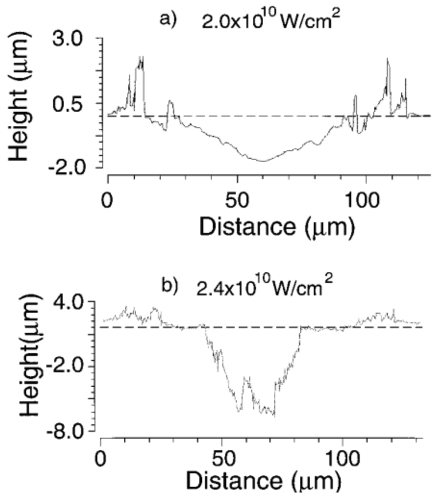

Fig. 3 – Cross-sectional images of the crater for: *a* – laser irradiance slightly below the phase explosion threshold (20 GW/cm^2) and *b* – laser irradiance slightly above the threshold (24 GW/cm^2) (Schittenhelm *et al.*, 1996).

Further studies have shown also that the ejected vapors and the liquid droplets are separated in time. Therefore, due to the difference in their mass, the vapors are reported to be detectable at t < 500 ns, while the droplets at t > 25 μs (Schittenhelm *et al.*, 1996; Kelly and Miotello, 1997; Yoo *et al.*, 2000). These conclusions where further used to understand results related to the structure and overall dynamics of the ejected cloud obtained by ICCD fast camera imaging and Langmuir probe measurements (Harilal *et al.*, 2002; Ursu *et al.*, 2009; Irimiciuc *et al.*, 2014, 2017; Focșa *et al.*, 2017).

3.3. Coulomb Explosion

The *Coulomb explosion* is one of the electrostatic mechanisms of the laser ablation (Bulgakova *et al.*, 2005). This mechanism has been discussed in many papers over the last years (Jiang and Tsai, 2003; Dachraoui *et al.*, 2006; Werner and Hashimoto, 2011; Lin *et al.*, 2012; Focşa *et al.*, 2017). Coulomb explosion plays an important role in different applications such as surface nanostructuring (Rapp *et al.*, 2016) or nanoparticle formation (De Giacomo *et al.*, 2013). Coulomb explosion has been observed first on dielectric materials, while for semiconductors and metals the subject remained controversial (Gamaly *et al.*, 2002). It was however proven that for higher fluences of the laser beam, the generated electric field can be high enough for the disintegration of the surface even for semiconductors and metals (Bulgakova *et al.*, 2004).

One of the important results was the reporting of the energetic ions of several species having the same momenta but different energies (Stoian *et al.*, 2000). Other reports (Ursu *et al.*, 2009, 2010; Pompilian *et al.*, 2013; Irimiciuc *et al.*, 2017; Wellershoff *et al*., 1999) based on investigation of the overall dynamics of the plume and individual kinetics of the ejected particles through optical methods revealed that doubly-charged ions had the velocities almost twice as high as the single-charged ions. The difference indicates that the ions are accelerated in the same electric field. The electric field is generated by intensive electron photoemission and by the separation between the fast escaping electrons and the ions left on the surface (or in the plume, but behind the electrons). Usually the repulsive force between ions is higher than the binding energy, which results in the disintegration of the surface (Fig. 4).

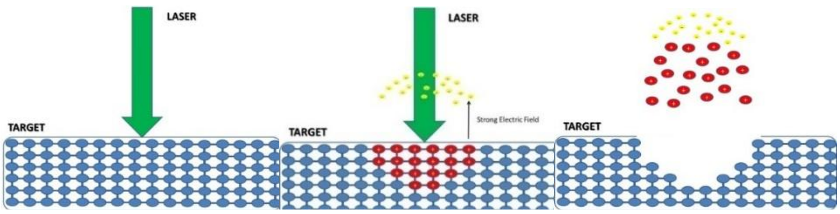

Fig. 4 – The stages of the Coulomb Explosion.

If the electric field, generated by photoemission, is higher than the atomic bonding energy, the density of the electrostatic energy per atom has to exceed the value of the sublimation energy per atom. For an ultra-short laser (fs, ps), where the thermal mechanisms do not play an important part, the threshold electric field can be approximated as:

$$E_{C_f}\Big|_{x=0} = \sqrt{\frac{2\Lambda n_0}{\varepsilon\varepsilon_0}} \tag{4}$$

where Λ (kJ/mol) is the sublimation energy per atom, n_0 (cm^{-3}) is the lattice density, and ε is the dielectric permittivity. For longer pulses (ns, ps) it has to be taken into account the heating of the lattice. Due to the heating, the vibrational energy of the atoms increases and the probability of the atoms to escape from the target due to thermal effects increases. That being said, Eq. (4) further becomes:

$$E_{C_n}\Big|_{x=0} = \sqrt{\frac{2(\Lambda - 3k_B T_s)n_0}{\varepsilon\varepsilon_0}} \tag{5}$$

4. Conclusions and Perspective

The complexity of all the processes involved lead to the implementation of complicated investigation techniques that often only capture a particular facet of the laser produced plasma. The optical diagnostics only captures information about the excited states present in the plasma, the electrical ones follow the dynamics of charged particles, while techniques involving mass spectrometry offer information about the molecules and clusters formed during the ablation process. The solution to have a more complete image of the laser produced plasmas, is the use of complimentary method in a simultaneous manner.

Also, a good alternative to the experimental approach is the use of appropriate theoretical model that would ideally, cover all the fundamental processes and ablation mechanisms. Since Coulomb Explosion and Phase Explosion do manifest themselves at different temporal scale, preferably the theoretical model needs incorporate be a multi-scale, multi-physics type approach. All the requirements are covered by the fractal hydrodynamic model proposed by our group about ten years ago, that recently showed how the differences in the fundamental mechanism involved in various ablation regimes reflect the fractalization of the system. Such a mathematical approach can be used to study the dynamic of similar systems that can be assimilated to a fractal fluid like discharge plasmas, polymers, blood or other complex polymers (Agop *et al*., 2009, 2010; Anisimov and Rakhamatulina, 1973).

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ABLAȚIA LASER: STADIU ACTUAL ȘI PERSPECTIVE

(Rezumat)

În această lucrare se prezintă o trecere în revistă a principalelor descoperiri și dezvoltări ce au avut loc în ultimii zeci de ani pe domneiul ablației laser. Accentul este pus pe tehnologiile dezvoltate pentru a evidenția diversele comportamente ale plasmei și principalele rezultate raportate în literatură. Obervațiile experimentale cât și abordările teoretice sunt discutate în raport cu procesele fundamentale ce duc la îndepărtarea materialului țintei prin ablație laser. Mecanismele fundamentale sunt discutate amplu pentru diverse regimuri temporale de ablație și se prezintă cele două mari categorii de mecanisme: electrostatice (explozia Coulomb) și termice (explozia de fază). Înțelegerea acestor aspecte fundamentale duce la posibile dezvoltări tehnologice cât și a modelelor matematice ce iau în considerare gradul de complexitate ridicat al acestui proces.