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Review

Shock Waves in Laser-Induced Plasmas

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Abstract: The production of a plasma by a pulsed laser beam in solids, liquids or gas is often associated with the generation of a strong shock wave, which can be studied and interpreted in the framework of the theory of strong explosion. In this review, we will briefly present a theoretical interpretation of the physical mechanisms of laser-generated shock waves. After that, we will discuss how the study of the dynamics of the laser-induced shock wave can be used for obtaining useful information about the laser–target interaction (for example, the energy delivered by the laser on the target material) or on the physical properties of the target itself (hardness). Finally, we will focus the discussion on how the laser-induced shock wave can be exploited in analytical applications of Laser-Induced Plasmas as, for example, in Double-Pulse Laser-Induced Breakdown Spectroscopy experiments.

Keywords: shock wave; Laser-Induced Plasmas; strong explosion; LIBS; Double-Pulse LIBS

1. Introduction

The sudden release of energy delivered by a pulsed laser beam on a material (solid, liquid or gaseous) may produce a small (but strong) explosion, which is associated, together with other effects (ablation of material, plasma formation and excitation), with a violent displacement of the surrounding material and the production of a shock wave.

The time evolution of a laser-induced shock wave and its geometry are dominated by the energy of the laser and the shape of the plasma generated. If the energy of the laser is delivered in a small spot, we may have spherical or hemispherical shock waves, while in case of elongated plasma (in liquids, for example) the shock wave may assume a cylindrical shape. Intermediate situations can be more complex (see Figure 1).

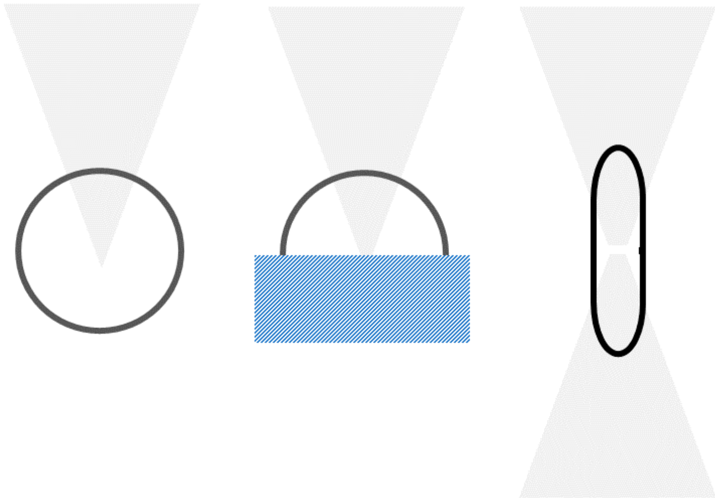

Figure 1. Spherical, hemispherical and cylindrical (with hemispherical caps) shock waves induced by a laser in a gas, solid surface or inside a liquid.

The theoretical basis for the study of shock wave generation and propagation were laid by studies of the blast wave generated in nuclear explosions. The studies performed at Los Alamos by Bethe et al. in 1941 [1] (published in a 1947 in a report then declassified in 1958) and in UK by the British physicist G. I. Taylor [2] (published in 1950) evidenced the main characteristics of a nuclear explosion versus an ordinary explosion, i.e., the fact that the energy is released in a nuclear explosion in a very limited volume. The theory describing the blast wave produced by a nuclear explosion is thus the theory of a point strong explosion. In Reference [1], the theoretical treatment of the point strong explosion was developed by John von Neumann. While realizing the difficulties of an exact treatment of the shock wave propagation, von Neumann, taking into account the dimensions of the explosion source, demonstrated that a great simplification of the model can be obtained by considering the source of energy as a point.

In this approximation, a self-similar solution to the propagation of the shock waves can be searched for, assuming a sudden release of an energy E_0 in a negligible time and volume.

Assuming an adiabatic expansion of the (spherical) shock wave, a purely dimensional reasoning leads to the determination of the expansion law of the shock wave as:

$$R(t) = \left(\frac{E_0}{K(\gamma)\rho_0}\right)^{1/5} t^{2/5}, \tag{1}$$

where $R(t)$ is the radius of the shock wave at time t, ρ_0 is the unperturbed density of the gas and $K(\gamma)$ is a constant which depends on the adiabatic coefficient g of the gas. Similar studies were developed in the same period in the Soviet Union by L.I. Sedov.

Sedov extended the self-similar treatment to strong explosions produced by linear (cylindrical waves) or planar (plane waves) sources. An immediate generalization of Equation (1) to these shock wave geometries leads to the expression:

$$R(t) = \left(\frac{E_0}{K(\gamma)\rho_0}\right)^{1/(2+\alpha)} t^{2/(2+\alpha)}, \tag{2}$$

with α = 3 for spherical shock waves, α = 2 for cylindrical and α = 1 for plane shock waves.

The work of Sedov, originally published in a Russian journal [3], later became available in the Western world (in 1959) with the publication of the famous book *Similarity and Dimensional Methods in Mechanics* [4]. In fact, the description of the shock wave propagation is often referred to the Sedov–Taylor point strong explosion theory.

An interesting discussion on the limits of the Taylor approach, which obtained some popular success after the successful determination of the (classified) energy of the first atomic bomb tested in 1945 in the Nevada desert (see Figure 2), is reported by Michael A.B. Deakin in Reference [5]. According to Deakin, Taylor's calculation of the Trinity bomb energy [6] (which, in the word of his biographer G.K. Batchelor, caused him to be "mildly admonished by the US Army for publishing his deductions from their (unclassified) photographs" [7]) was indeed overestimated, although by a mere 3% with respect to the calculations that could have been derived by the exact treatment of von Neumann or Sedov.

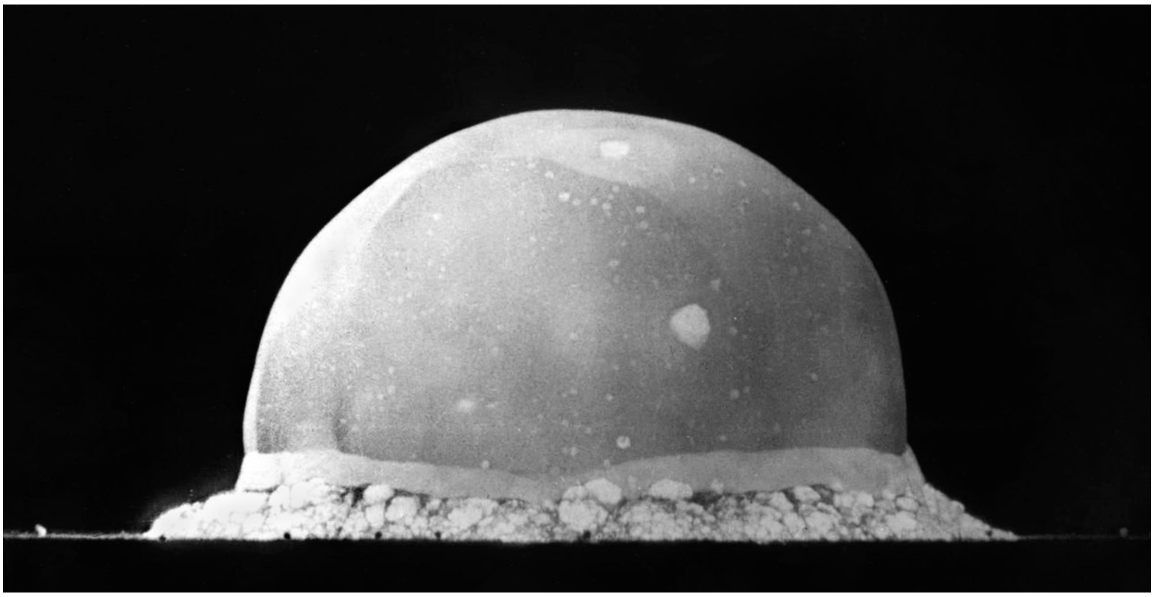

Figure 2. The blast wave produced by the first nuclear test, 16 ms after the explosion. The radius of the wave is about 200 m. (Trinity test, New Mexico 1945).

The critical point of the discussion is the evaluation of the K constant in Equation (1), which is a function of the adiabatic coefficient γ of the gas in which the shock wave propagates. While the treatment of von Neumann and Sedov is exact, the determination of the K constant using Taylor's approach is approximated. One should consider, in any case, that Equation (1) always underestimates the total energy released in the explosion, since the thermal losses—which can be very important in a nuclear explosion—are neglected.

Considering the parameter E_0 in Equation (1) as a kind of effective energy carried out by the shock wave, however, one can describe quite precisely the evolution of a laser-induced spherical shock wave in different gases, as reported by Gatti et al. in 1988 [8], using a value of $K(\gamma)$ comprised between 0.6 and 0.8, depending on the gas (see Figure 3).

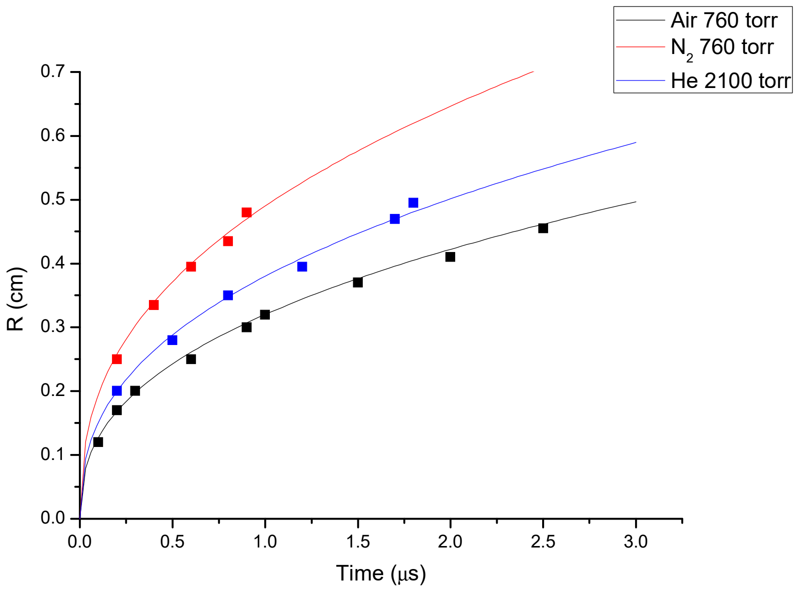

Figure 3. Expansion of the spherical shock wave produced by a Nd:YAG laser in gas (black points: Air at pressure of 760 torr; red points: Nitrogen at 760 torr; and blue points: He at 2100 torr). The continuous line represents the predictions of the point strong explosion theory (Equation (1)). The data are from Reference [8].

What is interesting is the fact that, although the point strong explosion theory should hold only for very strong explosions (i.e., very fast expanding shock waves), the agreement between Equation (1) and experimental results remains very good up to times where the shock wave expansion velocity, given by

$$v(t) = \frac{2}{5}\left(\frac{E_0}{K(\gamma)\rho_0}\right)^{1/5} t^{-3/5}, \tag{3}$$

arrives at Mach numbers $M \approx 2$.

The reliability of the point strong explosion theory in describing the expansion of laser-induced shock waves was further demonstrated by Harith et al. [9], in a wide range of absorbed laser energies (from $E_0 = 0.05$ J up to $E_0 = 2$ J) in different gases at different pressures. While reducing the energy of the shock wave, however, a decrease of the density jump at the shock front was observed.

In fact, in the ideal conditions of strong point explosion, the density jump at the shock front is given by the relation:

$$\frac{\rho}{\rho_0} = \frac{\gamma+1}{\gamma-1}. \tag{4}$$

Equation (4) corresponds to the limit for $M >> 1$ of the expression

$$\frac{\rho}{\rho_0} = \frac{(\gamma+1)M^2}{(\gamma-1)M^2+2}, \tag{5}$$

which can be derived taking into account the counter-pressure of the ambient gas, which is not negligible in the later stages of the shock wave expansion [10]:

$$\frac{p}{p_0} = \frac{2\gamma M^2-(\gamma-1)}{(\gamma+1)}. \tag{6}$$

Accurate numerical simulations of the process of shock wave propagation in gases were performed by Dors and Parigger [11] and successfully compared with high-speed, high-resolution shadowgraph techniques. Other important computational and experimental studies on laser-induced shock waves were performed by Harilal and coworkers [12–14], in the framework of extensive and articulated research devoted to the study of the expansion of the laser plume in different ambient gases and experimental configurations [15–20]. In a recent paper, Hermann et al. [21] studied the evolution of the pressure in a steel plasma, acquiring its emission spectrum and successfully comparing the experimental data with the results of computer simulations.

2. Shock Wave and Plasmas

Laser-induced shock waves may have an important role in the early stages of plasma formation in solid targets. In fact, if the energy of the shock wave generated by the laser is enough for ionizing the surrounding gas, a so-called laser-supported detonation (LSD) [22,23] might be activated.

The onset of the laser-supported detonation depends critically on the laser power released on the sample. At laser irradiances around 7–8 × 10^8 W/cm^2, the shock wave ionizes the surrounding gas, which may absorb the energy of the laser pulse tail, thus generating a hot plasma which expands outward with the shock wave. The laser-supported detonation mechanism increases the concentration of the elements of the ambient gas in the plasma. While the concentration of these elements is higher in the outer region of the expanding plasma, the plasma turbulences may bring the ambient gas elements into the plasma core, where they can become dominant with respect to the elements of the target.

The mechanism of laser-supported detonation was recently studied in detail by Cristoforetti et al. [24] and Ma et al. [25] for the generation of plasmas in air and argon, respectively. Cristoforetti et al. found that for an aluminum plasma generated in air at high laser irradiances, a significant mixing of the sample material with the ambient gas occurs, with the concentration of nitrogen and oxygen being predominant on Al even in the core of the plasma.

At lower laser irradiances, on the other hand, the “piston effect” of the shock wave generated by the laser could be able to displace the outer gas before the arrival of the hot core plasma, thus effectively preventing the mixing between ambient gas and target plasma. In this regime, on Al samples the inhibition of the formation of AlO molecules in the plume has been observed, due to the “barrier effect” of the shock wave, which prevents the mixing of Al ions and oxygen molecules from ambient air [26,27].

More complex phenomena occur in the interaction of two laser-induced plumes (and related shock waves). These effects have been studied in a number of papers, for example, by Hough and coworkers [28–31].

From a technological point of view, the study of the interaction between the laser-induced shock wave and the plasma plume is interesting for its implications in the generation of metallic nanoparticles by laser in gas [32–34].

3. Diagnostic Applications

The dependence of shock wave speed on the energy delivered by the laser has attracted some interest for the diagnostics of laser-induced plasmas in solids. Israel Schechter and coworkers [11] exploited Equation (1) for evaluating the laser energy released on the sample on sand/soil mixtures [35]. The authors found an excellent correlation between shock wave velocity and laser energy and was able to explain the origin of the matrix effect in sands of different densities in terms of the different energy released on the sample. The Schechter idea was revived in the following years by several authors without substantial improvements. Tsuyuki et al., for example, in 2006 measured the hardness of concrete samples through the measurement of the laser-induced shock wave speed [36]. They also observed a correlation between the concrete hardness and the CaII/CaI line intensity ratio. Lahna et al. in 2017 measured the shock wave speed for evaluating the hardness of organic samples [37].

Chen and Yeung [38], in 1988, found a correlation between the acoustic wave intensity and the line emission intensity in laser-induced plasmas. Similarly, Pang et al. [39] in 1991 used the acoustic wave intensity as a diagnostic tool of the laser ablation process in LA-ICP-MS applications. The acoustic wave intensity is a feature of the shock wave which is indubitably easier to measure than its propagation speed, and the authors of [39] reported an improvement of a factor of 2 in the reproducibility of their measurements using this parameter for normalizing the intensity of the signal obtained to the ablated mass. Conesa et al. [40], a few years after the publication of the Pang work, tested the idea on Laser-Induced Breakdown Spectroscopy (LIBS) plasmas and successfully linked the acoustic properties of the laser-generated shock wave to the plasma dynamics.

After more than 30 years the idea of using the acoustic wave intensity as a measure of the ablated mass was revived by Murdoch et al. [41] and Chide et al. [42], with the aim of improving the performances of the SuperCam LIBS instrument which is supposed to be delivered to Mars in 2020 [43].

The propagation of acoustic waves on Mars may be, on the other hand, quite problematic (the tests reported in [42] were performed in terrestrial conditions, at normal atmospheric pressure), and the entity of the interference of the Martian wind has yet to be quantified.

Although some improvement might surely be obtained, especially in the rejection of outlier spectra, it should be however considered that the perceived intensity of the acoustic wave can be affected by a number of phenomena not directly associated with the energy released by the laser on the target or, at least, not easily explained by the point strong explosion theory, which is valid only for strong (i.e., supersonic) shock waves. The time dependence of shock wave radius on the energy released on the sample is quite weak ($E_0^{1/5}$) and its intensity (proportional to the pressure jump at the shock wave front, expressed by Equation (5)) is even less sensitive on the energy of the shock wave, which is in turn assumed to be proportional to the ablated mass, when the Mach number M of the shock waves approaches 1.

Other proposed applications of the acoustic wave intensity as proxy of some physical parameter of the target involve the study of the temperature of ethylene-air premixed flames [44]. The new technique proposed, named LIBT (Laser-Induced Breakdown Thermometry) by the authors, exploits the variation in acoustic wave intensity associated with the variations of gas density produced by temperature changes. The temperature/density effect is dominant on the shock wave intensity with respect to the compositional variations of the flame. The authors are forced to use heavy statistical analysis to eliminate the effect of anomalous results but, in the end, they seem to be confident that the analysis of the acoustic signal produced by the shock wave could be used for estimating the flame temperature and its spatial distribution inside the flame.

4. Reflection and Interaction of Shock Waves

The shock waves generated by laser can be reflected by obstacles and interact with other shock waves, as occurs for the ordinary acoustic waves. The laws which regulate the behavior of the shock waves are, indeed, more complex than the ones valid for the acoustic waves. The phenomena associated are treated extensively in the Landau and Lifshitz book devoted to *Fluid Mechanics* [45], the sixth book in the series of the *Course of Theoretical Physics*.

One of the most interesting effects which arises during the (oblique) reflection of shock waves from solid surfaces, is the generation of a third wave, called a Mach wave, which propagates parallel to the surface (see Figure 4).

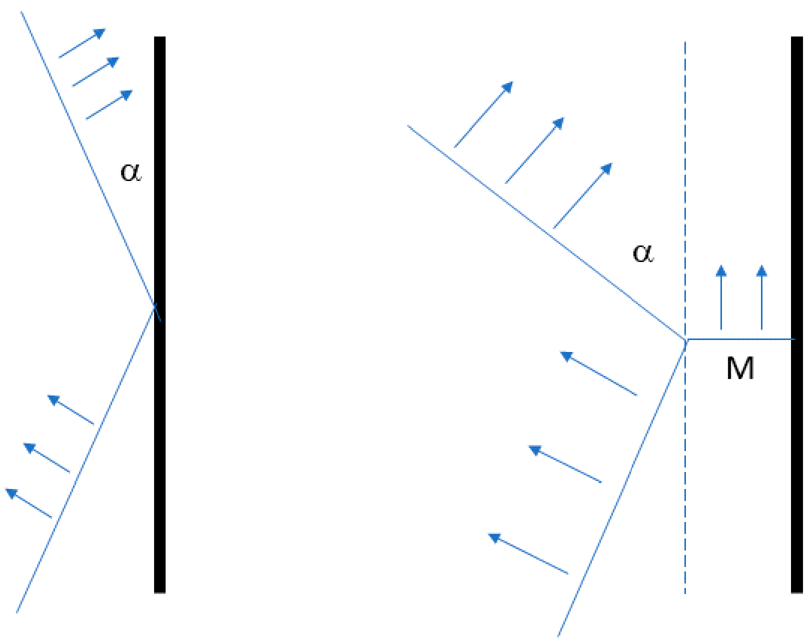

Figure 4. Normal reflection of a shock wave (left) vs. Mach wave generation (right). The critical angle α for the onset of Mach reflection depends on the gas and on the Mach number of the shock wave.

There is a critical angle, depending on the shock wave Mach number, for the onset of the Mach wave. De Rosa et al. [46] calculated this critical angle in 1992, obtaining values in excellent agreement with the experimental results obtained in the interaction of identical laser-generated spherical shock waves in gas [47,48].

The generation of Mach waves in the interaction of spherical shock waves contributes to the homogenization of annular shock waves of spherical converging shock waves produced by individual point electrical discharges, as discussed by Harith et al. in [49].

Another important effect related to the generation of Mach waves is the propagation of a spherical shock wave confined inside a crater. Corsi et al. [50] showed that the spherical shock front transforms into a planar front when exiting the crater, because of the Mach reflection (see Figure 5).

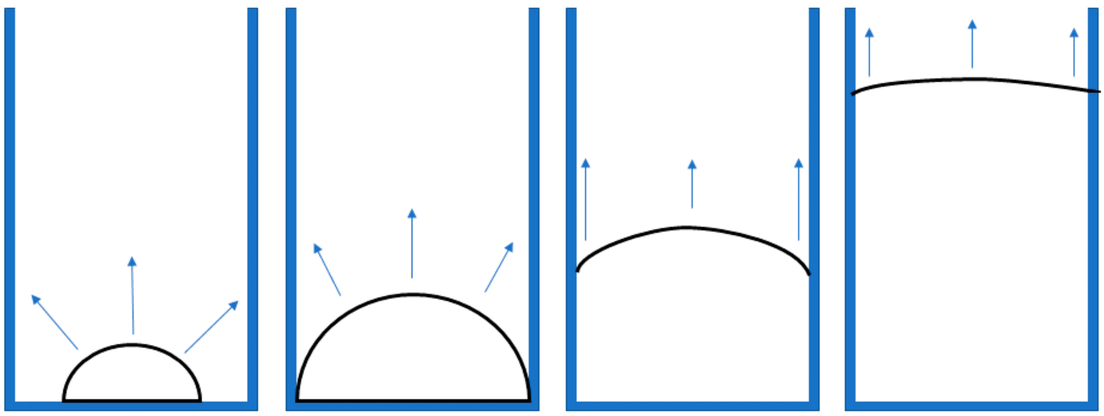

Figure 5. Schematic representation of the transformation of a spherical shock wave in a planar one, due to the Mach reflection effect on the walls of a crater. The Mach wave is faster than the initial spherical wave.

The neat effect observed in [50] was an enhancement of the spectral signal emitted by the plasma with respect to the signal generated on the surface of the sample. This topic will be discussed in detail in the next section.

5. Enhancement Effects of Laser-Induced Plasma Optical Emission

The shock wave produced by the laser pulse coexists and interacts with the plasma created by the same laser or by other means. Several papers have been published in the early stages of development of the Laser-Induced Breakdown Spectroscopy technique dealing with the confinement of the plasma and the emission enhancement effects produced through the interaction with the shock wave. Giulietti et al. in 1990 [51] studied the reflection of a laser-induced spherical shock wave produced in hollow glass balls, and the interaction of this wave with the plasma produced at the center of the ball was discussed theoretically and studied experimentally by Singh et al. [52–55].

As mentioned in the previous section, the influence of the shock wave in the enhancement of the LIBS signal in stratigraphic analysis was also discussed in 2005 by Corsi et al. [50], in relation to the confinement effect of the plasma by the (planar) shock wave produced at the bottom of the laser-produced crater.

As often occurs in the fast-growing community of LIBS research, these old ideas have been developed and perfected in the first decade of this century (Popov et al. [56,57] used spatial confinement of the plasma to enhance the LIBS signal in soils) and then recently "re-discovered" (see for example Yin et al. [58] in 2015, Li et al. [59] in 2016 and Ahmed et al. [60] in 2019).

These applications of shock wave confinement are probably now superseded by the advent of double-pulse Laser-Induced Breakdown Spectroscopy, proposed by Uebbing et al. in 1991 [61] and studied extensively by St-Onge et al. [62,63]. The fundamental role of the shock waves in double-pulse LIBS was not immediately clear. Sergey Pershin [64] in 1989 was probably the first to realize that the signal enhancement in double-pulse LIBS was not due to the simple reheating of the first plasma but to the evolution of the second pulse in the modified environment produced by the first. Unfortunately, his intuition was more or less ignored by the LIBS community, until the mechanism of double-pulse LIBS signal enhancement was independently interpreted in 2004 by Corsi et al. [65], considering the fundamental effect of the shock wave produced by the first laser pulse.

An account of the theoretical analysis which outlines the essential role of the laser-induced shock wave in double pulse LIBS is given in several reviews, notably Cristoforetti and Palleschi [66] in 2011 and Tognoni and Cristoforetti [67] in 2014, which basically attribute the signal enhancement in collinear, parallel and pre-ablation orthogonal double-pulse LIBS (see Figure 6) to the optimal balance between ablated mass and plasma temperature, obtained by reducing the plasma shielding in a low-pressure environment.

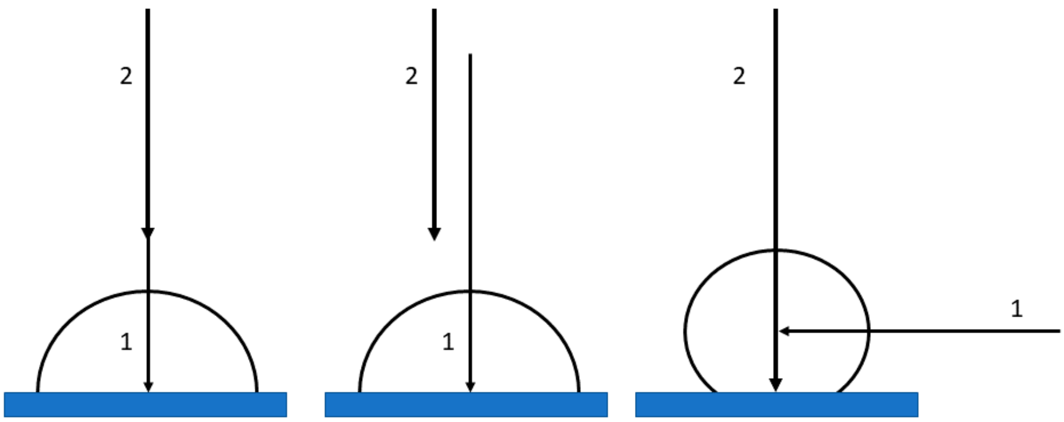

Figure 6. Schematic representation of the different double-pulse LIBS schemes where the laser-induced shock wave plays a fundamental role. From the left: Collinear double pulse, parallel double pulse and orthogonal double pulse in pre-ablation configuration.

Many experimental work confirmed this simple, yet complete model of the double-pulse effect (Cristoforetti et al. [68–72]). The comprehension of the role of the laser-induced shock wave in double pulse LIBS allows the optimization of the experimental parameters; that is, the energy of the first laser pulse, for example, may be considerably lower than the energy of the second pulse as soon as a strong shock wave is generated.

6. Shock Waves in Liquids

The generation of shock waves in liquids by laser is often made difficult by the need of using long focal lenses to access the bulk of the liquid, a fact which may lead to the creation of elongated, quasi-cylindrical shock waves [73] as the one depicted schematically in Figure 1. When the laser energy can be delivered successfully in a single spot, spherical shock waves can be created that behaves as micro-bubbles in the liquid.

The formation of bubbles in liquids and their interaction have been studied extensively in the past century. The process of cavitation (collapse) of the bubbles formed under the effect of blades and turbines was the main cause of damage of these objects in the past [74].

A spherical shock wave produced by a laser in a liquid can undergo a sequence of expansion, cavitation and re-expansion, until its energy is finally dissipated into the medium because of friction [75,76].

The size and dynamics of the laser-induced shock wave in liquids depend on several parameters, and in particular on the energy and time duration of the laser pulse [77]. The typical shock waves produced by a nanosecond Nd:YAG laser in water may last several hundreds of microseconds, expanding up to a few millimeters in diameter and then collapsing during cavitation.

There are many interesting aspects in the generation, propagation and interaction of laser-induced shock waves in water. Some work was done in the last decade of the last century by Harith et al. [78] and Famà et al. [79] on the study of reflection and transmission properties of planar shock waves in water. At present, the attention of the LIBS community is drawn toward the possibility of exploiting the characteristics of laser-induced shock wave in water for the analysis of submersed objects.

De Giacomo et al. [80] in 2007 discussed the advantage of underwater double-pulse LIBS versus single-pulse LIBS on submersed solid targets. While conventional, single-pulse LIBS is characterized by high breakdown thresholds, short lifetime of the plasma and a large continuum emission, whereas the double pulse strategy allows to create a bubble on the target where the second pulse can propagate, as we have already seen in conventional double-pulse LIBS analysis in gas. The delay time between the first laser pulse, which creates the shock wave/bubble, and the second which creates the plasma on the target, is crucial for determining the plasma parameters. Different levels of confinements can be obtained depending on the relative timing of the two pulses (see also Lazic et al. [81,82]).

In a recent paper, Cristoforetti et al. [83] made a careful calculation of the optimum conditions for underwater double-pulse LIBS analysis for optimizing the reproducibility of the cavitation bubble generation on submersed targets.

7. Conclusions

The interest in the generation and propagation of shock waves in gases was sparked at the end of WWII, by the effort in developing the nuclear bombs that tragically ended the conflict. After slightly less than 80 years from that time, the geo-political situation has changed, and the attention of the researchers has shifted towards the many possible applications which have been opened by the possibility of generating a strong point explosion using a laser, instead of an atomic bomb. In this short review, we have underlined the possibility of exploiting the physics of laser-induced shock waves for the diagnostics of the laser ablation process, for extraterrestrial exploration, for measuring the temperature of a flame or remotely analyzing the composition of objects on the ground or underwater. Many other interesting topics have been treated only in passing, such as the interaction of laser plumes with obstacles or between them, or the generation of nanoparticles on metallic targets by laser in the presence of ambient gas.

The experimental strategies for studying the generation and propagation of laser-induced shock waves, such as shadowgraphy, interferometry, holography, etc., have not been discussed in detail, since they alone should have deserved a dedicated review. Moreover, most of them are not specific to the analysis of laser-induced shock waves (for a recent discussion of the main methods used for shock wave detection and diagnostics, see for example [84]).

The number of applications discussed in recently published papers demonstrate the fact that, despite their long history, interest in laser-induced shock waves is not lost in the scientific community. Indeed, they are still the subject of research and vivid discussion.

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