Simulation of meteors by TC-LIBS: Advantages, limits and challenges

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Important features in meteor spectra are usually interpreted by synthetic convolution of lines extracted from databases. We developed new approach in this field: experimental techniques based on ablation of real meteorite samples using a wide range of laser sources. We performed experiments in order to provide spectra of several molecular, radical and molecular ion species that are likely to appear in meteor or cometary impact plasma. The spectra are recorded by high-resolution laboratory Echelle spectrograph and simultaneously using a high-resolution Meteor Spectral Camera for real meteor observation. In this manner, we show that instead of theoretical spectra simulation, laboratory experiments can be applied at least for qualitative evaluation of the observational data and assignment of important spectral features in meteor emission spectra. We also provide examination of ablation spots for future estimation of parameters of a high-power laser, which is used for remote LIBS applied in astronautics.

1 Introduction

The most remarkable advantage of comparative measurements using LIBS lies in the applicability of this method to the real-time in-situ analysis of any sample of any real meteorite without any preceding treatment, preparation or isolation. Plasma is generated in the laboratory under strictly defined conditions, and the elements are evaporated together with the whole matrix, mimicking the meteoroid or asteroid descent (Ferus et al. 2018a, 2019). These attributes put LIBS in line for a wide range of laboratory studies revealing physical and chemical consequences of atmospheric entry of an extra-terrestrial body as well as its impact on a planetary surface (Ferus et al. 2015b).

More than 15 years ago, our laboratory indulged in research mainly focused on the exploration of chemical consequences of early planetary bombardment during the post-accretion period after the dissolution of a protoplanetary disk. During the formation of the Sun, a disk of material which consisted mainly of gas and small dust particles orbited the new-born star. Over a few tens of millions of years, particles from the disc collapsed and formed what today are the planets and asteroids of our Solar system. This process eventually led to frequent and massive impacts on the proto-Earth (Canup and Asphaug 2001). The impact frequency slowly decreased over time, only to rise again during the Late heavy bombardment (LHB) about 4-3.85 Gyr ago (Koeberl 2006). The LHB phenomenon has been explained by gravitational resonance interactions of Jupiter with Neptune and other gas giants (Tsiganis et al. 2005; Nesvorny and Morbidelli 2012). This transient instability of the resonance ratios led to a change in Jupiter’s orbit as well as the ejection of asteroids and comets from their previously stable orbits. The would-be observed effect on Earth was a 10-fold increase in the frequency of extraterrestrial body impacts relative to the frequency immediately before LHB (Morbidelli et al. 2012; Geiss and Rossi 2013). This in effect meant 10^9 tons of impacting material per year (Koeberl 2006). Typical impact velocities are estimated to have increased from 9 $km.s^{-1}$ to 21 $km.s^{-1}$. Also the gravitational cross-sections ratio of the Moon and the Earth is approximately 1:17, which means 17 impact basins should have been formed on Earth per one lunar basin (Bottke et al. 2012). These lunar craters are still visible nowadays and their age and origin have been successfully ascribed to the LHB. The impact frequency decreased again after the LHB period until 3.8 Gyr ago and since then has not fluctuated much. Our explorations show that extraterrestrial body impacts were a major source of minerals, volatiles (e.g. water) and energy for chemical transformations. This raises a question of the influence on the early Earth’s environment, more specifically the decomposition or synthesis of prebiotically relevant compounds and early chemical evolution of our planet. To model such conditions as precisely as possible, it is best to use impact plasma, which can be generated e.g. by a high-power laser source.

During the last two decades, our team demonstrated several experiments mostly focused on chemical consequences of hypervelocity impacts on atmosphere or interaction with solid or liquid surfaces (Babankova et al. 2006). Most studies have been focused on impact-induced synthesis of biomolecules (Šponer et al. 2016) such as canonical nucleobases (Ferus et al. 2012, 2014, 2015a, b, 2017c) sugars (Civis et al. 2016b) and amino acids (Civis et al. 2004); or transformation atmospheric molecules on early terrestrial planets such (Civis et al. 2008) as formation or decay of prebiotic substances such as formamide (Ferus et al. 2011), isocyanic acid,(Ferus et al. 2018c) transformations of hydrogen cyanide (Ferus et al. 2017b), acetylene (Civis et al. 2016a), methane, (Civiš et al. 2017) or carbon monoxide (Civis et al. 2008; Ferus et al. 2009).

Apart from the chemical consequences of impacts, another important topic is the study of physics and the spectroscopic parameters of atmospheric entry and impact (Madiedo et al. 2013)(Jenniskens 2007). The chemical composition of distant objects and events in the universe can be determined using only spectroscopic techniques. To understand the observed spectra, laboratory experiments must be performed to compare the spectral features of individual systems under controlled physical and chemical conditions (individual species concentration, temperature, pressure, and electron density), (Ferus et al. 2017a, 2018a, 2019).

Understanding meteor spectra is extremely important. Meteoroid source bodies, asteroids, are remnants from the materials that first formed the planetesimals and planets. Meteorites are pieces of asteroids on Earth that allow us to measure many of the properties of their parent bodies in detail. However, a fundamental problem exists in linking specific meteorites to their parent bodies (primary matter, asteroids, and comet nuclei). Moreover, most bodies are evaporated and disintegrated completely during their descent and their emission spectra measured using spectrographs are the only record of their chemical composition. In specific cases the body can be found as a meteorite and recorded spectra together with this real sample allow detail spectroscopic study leading to full understanding to meteor spectra, composition of parent body, its origin, chemistry etc.

The detail description and understanding of their behaviour in the atmosphere is therefore a challenging scientific problem worth studying. When a body enters the Earth’s atmosphere, it is immediately surrounded by a meteor plasma and interacts with the highest layers of the Earth's atmosphere at very high speeds (up to tens of kilometres per second). Descent of a meteoroid through the atmosphere leads to rapid heating, surface ablation and parent body disintegration. The initial height of a meteoroid ablation (early stage of the light – visible – part of atmospheric trajectory) depends on the geocentric entry speed and the initial mass. For known major meteor showers (with retrograde orbit, e.g. Leonids, Perseids, Orionids), and sporadic meteors with high geocentric velocity, the initial height of ordinary meteors varies between 120 km and 100 km. On the contrary, for meteor showers and sporadic meteors with low geocentric velocities (e.g. Phoenicids, Arietids, Drakonids), the initial height ranges between 90 and 80 km. In the case of showers with high geocentric speeds, e.g. Leonids or Orionids, some meteors occur at a very high initial height, which in rare cases can reach up to 150 km. Lowest point of meteoroid visible atmospheric trajectory (end of ablation process) depends besides geocentric speed and initial mass also on the zenithal angle of entry into the atmosphere. Meteors with low geocentric velocity and the mass in order of kg may have the visible path end height between 30−50 km above the Earth's surface. Meteoroids called Earth−grazers have a special trajectory. They enter the Earth's atmosphere at a very small angle and, in case of a suitable combination of low geocentric speed and initial mass in order of kilograms, the body fails to fully vaporize and continues along new heliocentric orbit with significantly different elements.

In previous studies dealing with the LIBS method (Ferus et al. 2018a, 2019), we demonstrated that the emission intensity of a particular spectral line of a studied analyte depends not only on the physical parameters of the line and the quantity of the emitting element but also on the matrix where it is embedded. This matrix dependence leads to the necessity for calibration curves or matrix matched standards, which in some practical situations, including samples of meteorites, are simply unavailable. Ciucci et al. proposed a novel Calibration Free Laser Induced Breakdown Spectroscopy (CF-LIBS) procedure to overcome the mentioned matrix effect (Ciucci et al. 1999). The CF-LIBS method is based on direct analysis of emission lines of an analyte together with a matrix, instead of looking at the matrix as an independent problem. We analysed, using this method, a wide range of chondrite meteorites and developed all the subsequent steps of data processing using the Calibration Free method, suitable for meteor analysis, and we used this method for the interpretation of bright bolide Perseid and Leonid meteor spectra (Ferus et al. 2018a, 2019). Moreover, as mentioned above, we used the reference experimental spectra of meteorite ablation, laser induced breakdown (LIDB) in the air and glow discharge in the air.

In our previous studies (Ferus et al. 2018a, 2019), we demonstrated calibration free analysis of a meteor spectrum and we created a catalogue of main meteor spectral features based on ablation experiments using terawatt-class laser (TC-LIBS). Regarding the works of other authors proposing laser-based laboratory studies related to astrophysics and astrochemistry, we should also note publications by Hawkes (Hawkes et al. 2008) proposing that laboratory based laser ablation techniques can be used not only to study the size of the meteor luminous region, but also to predict spectral features, estimate the luminous efficiency factor, and assess the role of chemically differentiated thermal ablation of meteoroids. Further, Milley (Milley et al. 2007), simulated meteor luminosity through laser ablation of meteorites. In 2017, Ebert (Ebert et al. 2017) simulated the virtually instantaneous melting of target rocks during meteorite impacts. Alongside with these studies, the simulated space weathering together with the deflection of asteroids by lasers remain the most studied research topic connected with application of lasers in this field. Deflection of asteroids and comets by lasers was proposed by Park and Mazanek (Park and Mazanek 2003), and recently, Aristova et al. (Aristova et al. 2018) studied impact physics of nuclear explosion on hazardous asteroids. Moroz et al. (Moroz et al. 1996) simulated the optical effects of impact melting and repeated crystallization on asteroidal surfaces using laser ablation and Kurahashi (Kurahashi et al. 2002) conducted laser ablation laboratory simulation of space weathering focused on the source of difference between reflectance spectra of ordinary chondrites and their parent bodies: S-type asteroids. Loeffler et al. (Loeffler et al. 2008) studied the effect of redeposition of impact-ejecta on mineral surfaces using laser ablation.

2 Experimental Part

Complex spectra within the entire UV / VIS – range have been recorded by High Resolution Echelle Spectra Analyzer ESA 4000 (LLA Instruments GmbH, Germany). The optical analyser unit enables spatial and temporal resolved image of lowest spectral intensities. The resolution is a few pm in the range 200 – 780 nm with resolution of 0.005 nm (200 nm) to 0.019 nm (780 nm). Simultaneously, the spectra have been recorded for direct calibration by common astronomical spectroscopic camera directly employed in observation of real meteors for comparative measurements. We have used high-resolution camera PointGrey Grasshoper3 GS3-U3-32S4M-C with high quantum efficiency (QE= 76%, 525 nm), dynamical range (71,34 dB), with CMOS chip Sony Pregius 2048×1536 px, grating 1000 lines/mm, which allows resolution 0,48 nm/px. The spectral intensity recorded by the instrument is calibrated using approaches applied by our team: Calibration to standard sources (Deuterium lamp, Tungsten source and calibration using standard spectra of Venus). Wavelength calibration is achieved using high resolution data and standard wavelengths of calibration sources (deuterium lamp). For spectroscopic studies of meteors, we also use older system based on spectrographic cameras equipped with QHY5LII-M hardware, 1280x960 px CMOS chip and an objective Tamron (f/1,00; F/3-8 mm) using 1000 lines/mm grating, which allows a resolution up to 0.97 nm/px. In our measurements, for every sample of meteorite, the Echelle spectrograph was set to trigger 1 µs after the laser pulse with the gate open for 2 µs for a total accumulation of 1 signal after 1 large laser shot. Laser PALS repeated laser pulse after every 30 minutes necessary for cooling down the system. Low resolution observational spectrograph opened the gate for 1 s being triggered by the laser system.

The plasma shock wave induced by the impact of a meteoroid was simulated using the high−power laser PALS (Prague Asterix Laser System). The laser beam was focused on the sample placed in the interaction chamber by a plano-convex lens, with a diameter of 20 cm and a focal length of 50 cm. During the ablation of the meteorite surface initiated with a laser pulse of energy of 600 J (time interval ≈400 ps, wavelength of 1.315 µm) all manifestations connected with a high−energy density take place: shock rises in temperature to several thousand K, the formation of a shock wave plasma ejected from the surface, and the generation of secondary hard radiation (UV−VUV, XUV, X−Ray). All the experiments have been performed under pressure of 2 mbar corresponding to altitudes typical for the beginning of the meteoroid body ablation (above 110 km).

Meteoroid plasma was simulated in our laboratory via the laser ablation of meteorite samples using a Lambda Physik (ArF) excimer laser. The laser emits ~10-ns pulses with a wavelength of 193 nm and an energy of 200 mJ. The laser beam was focused using a calcium fluoride lens (focal length of 50 mm) onto a solid target (sample of meteorite) attached to an XYZ rotation stage. In a similar manner, the irradiation was also performed using Nd:YAG laser (6 ns, 1064 nm, 450 mJ) and femtosecond Ti:SAF laser (50 fs, 810 nm, 1 mJ). The rotation system with the holder of particular meteorite sample was placed in a vacuum interaction chamber equipped with a collimator connected similarly to arrangement of terawatt laser PALS directly with a high-resolution Echelle Spectrograph (ESA 4000, LLA Instruments GmbH, Germany). The low-resolution spectrum was simultaneously measured using an astronomical spectrograph.

3 Results and Discussion

In order to validate this approach, there are several points to be considered. First, a significant parameter for studying of atmospheric entry plasma is its temperature. Meteor plasma temperature is more or less a rigid parameter and reaches about 5000 K. The temperature exhibits only a slight dependence on the impact velocity or mass of the in falling object. The temperature range is usually 4000 K – 5800 K). The laser impact plasma has a different time-evolution than the in falling object plasma. Straight after the sub-nanosecond pulse of the laser, its energy is transformed into simulated atmospheric entry or impact plasma. A tiny spot at the impact site with large energy density produces extremely charged ions and temperatures of 7 eV, i.e. up to 100 000 K. This ‘ignition stage’ is very limited in time scale and we do not expect any chemistry. Also, this stage of the plasma development is not spectroscopically observable due to continuum emission from the system. Two microseconds after the laser pulse, characteristic emission of atomic lines with temperature estimated in previous works of Babanková to be about 4500 K arises in the case of gas phase experiments. According to our chemical models, this is related to ignition of radical reactions and decomposition of compounds. (Civis et al. 2016a)

Under collision conditions, these excited states usually decay faster than as is predicted by transition dipole moments. Conversely, in collisional excitations and energy transfer, several short living atomic states can exhibit longer emission. During the afterglow, it is assumed that the Boltzmann distribution among excited states is maintained. Finally, according to chemical models, also some background chemistry and physics can occur among reactive species on milliseconds timescales. Apart from the ignition state, therefore, the laser impact plasma is a valid model of the impact plasma of an extra-terrestrial body in the sense of temperature. Other parameters, such as the species density (concentration), are parameters, which are easily adjustable during the experimental procedure.

Surprisingly, a different behaviour is exhibited by plasma formed after ablation of solid targets. Our measurements reveal that laser induced breakdown on meteorite surfaces exhibit temperatures about 10 000 K for TC-LISB, but about 6 000 K for excimer lasers. This is closely related to the behaviour of frontal shockwave created in front of descending meteoroid body. Based on our measurements, typical values of electron densities estimated for laser induced plasma on meteorite surfaces are in order of 10^{16} cm^{-3} decreasing during microseconds after the laser initiation. Meteor trails exhibit electron densities ranging from 10^{12} to 10^{14} cm^{-3} (Ferus et al. 2018b) and references therein).

In our recent paper (Ferus et al. 2019), we have highlighted a major advantage of laser experiments under laboratory-controlled conditions for the production of precise tables of main spectral features observed in meteor spectra. Meteor plasma is formed after an evaporation of a very complex matrix – rocks and minerals embedded in the meteoroid or asteroid body. Only precise experiments can show the major coincidences of spectral lines, positions of spectral features formed as result of particular lines and convolution the observed spectrum. The peak maximum of an individual spectral feature can be different for a particular composition of the matrix and also temperature. Therefore, the common procedure of spectral lines (or rather spectral features) assignment in meteor spectra and the calibration of meteor camera cannot be precise if only theoretical tables are taken into account. Instead, we strongly propose assignment of spectra lines based on the data provided for ablation experiments of real meteorite samples in our recent paper.

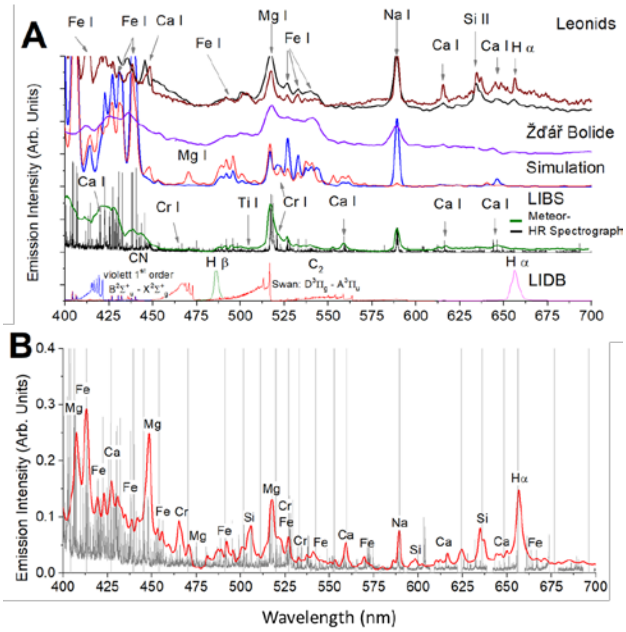

Figure 1 – A comparison of several meteor spectra, simulated and experimental LIBS (Panel A) with TC-LIBS spectrum recorded by high resolution echelle spectrograph and meteor camera (Panel B). The specific line list for spectral features recorded using high power laser is supplied in Ref. (Ferus et al. 2019).

The example meteor spectra compared with experimental TC-LIBS and standard laboratory LIBS spectrum recorded for a chondrite sample are depicted in Figure 1. Panel A shows spectrum of two examples of Leonid meteor shower (2016), famous very bright bolide Žďár (16^{th} of December 2014), simulation of ablations spectra together with real ablation spectra of a chondrite specimen provided by Excimer laser and also spectra of gas phase LIDB of several gas mixtures (methane, hydrogen, nitrogen) recorded at terawatt laser PALS. Panel B shows spectrum of TC-LIBS recorded at PALS infrastructure. The red envelope represents low resolution spectrum recorded simultaneously by astronomical spectrograph and the black lines indicate spectrum recorded by high resolution Echelle instrument.

We expect that precise measurement under laboratory conditions can also provide important parameters for the design of future space missions intended for direct survey of elemental compositions of bodies in the asteroid belt using remote high-resolution spectroscopy of laser induced ablation plasma over their surfaces. State of art laboratory laser sources are able to ablate mm^2 size spots while emission is measured very close to the target (e.g. Nd:YAG or Excimer providing energy in order of 10^{-2} J in sub-nanosecond pulses) . TC-LIBS performed for instance by high-power facility PALS is able to evaporate about 1 cm^2 (energy of 600 J). Our survey of craters performed by Optical Microscopy and Scanning Electron Microscopy (SEM) shows that high-power lasers create in one laser shot typically ablation spot of 10^2 μm in depth and about 1 cm (power of 600 J) in diameter while Nd:YAG laboratory laser source creates a spot with 10^1 μm in depth and diameter of 1 mm (power of 600 mJ). Only a series of several hundreds of pulses can create an ablated area comparable in depths and diameter with a single shot of large terawatt laser. The corresponding example is provided in Figure 2. Panel A shows ablation spot of PALS laser examined with electron microscopy, panel B shows a detail of this area. Panel C shows an example of microscopic mapping of a crater created by about 1000 pulses of Nd:YAG laser on rotating sample of a chondritic meteorite.

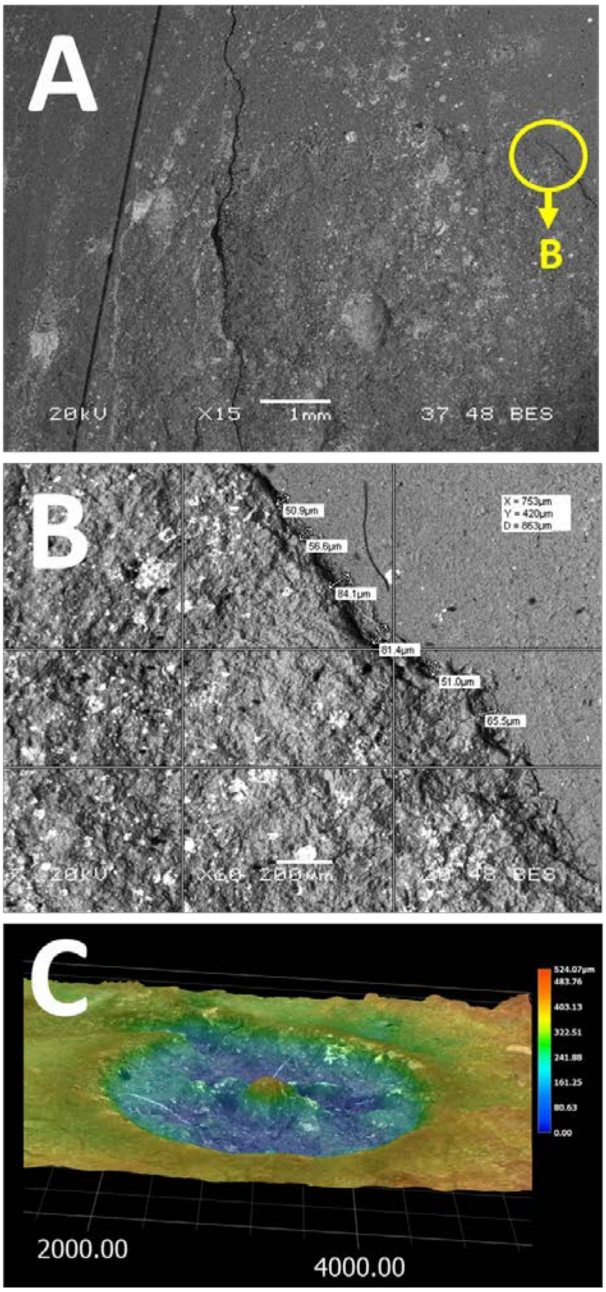

Figure 2 – An example of laser ablation spot survey performed my electron microscope for an ablation spot after a single pulse of terawatt laser facility PALS (panel A and detailed in panel B). Relief measurement of ablation spot after about 1000 pulses of standard Nd:YAG laser (Panel C).

The concluding comparison of laser ablation experiments with meteor plasma is provided in Table 1. First of all, it should be noted that ablation experiments represent the only state of the art method for the simulation of meteor plasma. Other methods (flames, electric discharges or induced plasma sources) require dissolution or further reprocessing of the material. Hypervelocity guns can serve as a very good model for target experiments. However, the projectile must be manufactured of copper or steel. Original sample of meteorite (or any other surface, e.g. dust or a plate of material subjected to an interaction test) cannot be fired for the gun.

Table 1 – Parameters of ablation plasma induced by several laser sources: ArF excimer laser, Nd:YAG laser, Ti:SAF laser and terawatt laser PALS. T is temperature expressed in K, n_e is electron density expressed in number of electrons per cm^3.

	ArF Excimer	Nd:YAG	Ti:SAF	PALS
T [K]	6 965	8 169	8 343	10 274
n_e [cm^{-3}]	$3.97x10^{16}$	$5.83\ x10^{16}$	$6.27x10^{16}$	$5.34\ x10^{16}$

The This means that the target as well as plasma formed after interaction is contaminated by the projectile material. Moreover, the typical speed of projectile reaches 10 $km.s^{-1}$, which is actually the lowest possible speed of a meteoroid. Also, any observable plasma is created only after the collision with the target and therefore, such kind of experiments cannot simulate the airglow around the meteoroid body. In comparison, laser ablation technique performed particularly by the terawatt-class laser (TC-LIBS) provides large volume of plasma (around 1 L) and evaporates significantly large surface of a meteorite specimen (about 1 cm^2). Although the physical initiation differs from meteoroid ablation, this technique has ambitions to provide real simulation of meteor chemistry and physics. However, it is necessary to test different laser energies, time scales after laser initiation, laser pulse lengths, buffer air speed and pressure etc. in order to approach all the parameters of extraterrestrial body atmospheric entry and impact.

4 Conclusion

Our explorations of advantages and limits of high-power laser application for laboratory simulation of chemical and physical consequences of an extraterrestrial body atmospheric entry as well as the extrapolation of technical parameters of high-power laser systems for space applications show that first of all, all the spectral features can be studied on laboratory level for any complex matrix and laser sources can be used for LIBS in astronautics. Physical parameters of high-power laser sources for deflection or destruction of potentially hazardous objects or for remote LIBS exploration of elemental composition of objects in the asteroid belt can be defined based on laboratory experiments. The main advantage of high-power laser application in laboratory astrochemistry and astrophysics is that they represent clear sources of energy directly delivered to vacuum sealed vessels or interaction chambers for chemical and physical experiments. The system is not contaminated by any other material. All the phases (buffer gas atmosphere, solid or liquid interaction target) represent neat materials with well-defined chemical composition. In such a set-up, we can study all the manifestations of high-power plasma in a well-controlled system without the influence of material introduced from outside (projectile, electrodes). Especially very harsh conditions (temperatures over 4000 K or manifestation of a shock wave) cannot be simulated by other techniques.

The starting conditions such as pressure, temperature, chemical composition, can be easily tuned and kept. If all the parameters cannot be simulated even with high-power lasers, this technique still represents better starting system for peak parameters extrapolation that engagement of small-scale laboratory systems such as small lasers. Serious disadvantage of this kind of experiments lies in the different kind of high energy plasma initiation (e.g. real high-speed atmospheric entry and ablation of the solid material by collisions with gas phase air molecules vs. simulation by melting of the solid target surface by laser light and its subsequent evaporation to the gas phase). High-power laser facility PALS also provides daily only very small number of pulses (1 every 30 minutes). This seriously limits the range of experimental conditions which can be explored.

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