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Combined impact features for laser plasma generation

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Abstract. Laser-induced plasma has been considered for multiple applications by the moment, and its characteristics strongly depend on laser radiation parameters. Reaching demanded values for the latter might be rather costly, but, in certain cases, similar or even better results could be reached in case of additional impact (optical, electric, magnetic, corpuscular, mechanical etc.). Combined impact effects are mainly based on target properties or interaction mechanism change, and found to decrease plasma generation thresholds by orders of magnitude, improve energy efficiency significantly, and also broaden the range of plasma parameters. Application area, efficiency and optimal regimes for laser plasma generation at such combined impact have been considered. Analysis based on published data and own experiments was performed for both target material and induced plasma flows. Criterial parameters have been suggested to characterize both combined impact and response to it. The data on plasma generation thresholds, controlled parameters, working media supply systems and recovery rate of droplets are very important for technology setups, including those for material modification.

1. Introduction

Control of plasma parameters is crucial for surface modification technology. Competitive plasma setups should provide high performance, be energy efficient and consist of reliable and inexpensive components. The analysis of data published on combined generation and control of laser plasma properties reveals several ways for performance improvement: modification of material properties for the most efficient laser radiation absorption and energy conversion; reduction of losses by proper energy transfer form and means; plume collimation and acceleration without energy consumption.

Laser impact efficiency is usually characterized by ablation threshold W_a (minimum radiation fluence enough for macroscopic material removal), ablation rate h (crater depth increase after pulse), specific mass flow rate $\Delta m/E$. Laser plasma generation efficiency is determined by specific impulse $I_{sp}\approx\langle v\rangle/g$ ($\langle v\rangle$ – is mass averaged rate), momentum coupling coefficient $C_m\approx\Delta m\langle v\rangle/E$, laser to kinetic energy efficiency $\eta\approx\Delta m\langle v\rangle^2/2E$ and some other parameters [1, 2], and can be improved by additional impact on the material or plume. For example, the following additional impacts led to increased laser plasma generation efficiency: material heating to fusion or glass transition temperature; material UV irradiation leading to excitation of chromophores and breaking of bonds; material surface covering with ~1 mm gel layer; multiple irradiation of the same spot until deep narrow crater formation (or preliminary manufacturing of those, e.g., wide blind pores); attachment of a cylindrical nozzle with a diameter ~2.4 times bigger than laser focal spot; attachment of a plate capacitor with divergent electrodes narrowing towards the ends, and irradiation with the focus closer to the cathode; application of a constant magnetic field (>0.15 T) normal to the irradiated surface; radiation focusing at two closely localized points under two-pulse impact.

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More promising ways for the improvement of combined impact efficiency are the following: local modification of material optical properties at the irradiated area; laser impact on ferrofluids at magnetic field and liquids confined in pored matrices; combined impact of laser and charged particle beams; radial confinement of laser plasma plume. Investigation of these ways of laser-induced plasma generation could provide new data on laser-matter interaction. Additional impact on working medium can also improve the performance of storage, transportation and dosing subsystems of plasma technology setups.

2. Review of previous works

Material heating can be performed electrically, optically or chemically. For some materials optimal temperature rates have been discovered (depending on efficiency parameters), those coming from the change in material properties, intensity of physical and chemical processes (e.g., hardness change or oxide films formation). One of convenient ways to control temperature is the change of interpulse delay between heating and acting irradiation. It has been shown experimentally that energy deposited per amount of removed material decreases. Since only surface layer needs to be heated, inductors and microwave heaters seem to be very promising.

Under double pulse UV impact on PMMA (with resonance absorption at 308 nm) a very pronounced synergistic effect has been discovered caused by excitation of chromophore groups and breaking of intermolecular bonds (followed by formation of oligomers) [3]. The most pronounced effect was observed under simultaneous impact that is considered as the evidence of photo processes. Except oligomerization, UV radiation can induce condensation, crystallization and polymerization (the latter can be also induced under electron beam impact and heating). Transient modification of material properties occurs at ultrashort prepulse irradiation [4], which makes quartz opaque to visible radiation due to excitation of electrons until relaxation or local damage. The structure of some materials can be also sensitive to magnetic or electric field due to orientation of dipoles.

Solid or liquid confining layers are also used (both at front and back side of the target) for the combined impact induced reduction of energy losses, plasma compression and heating at initial stages (also due to efficient absorption of laser radiation), increase of the duration of plasma-target interaction. Laser impact energy can be reduced for such layered targets, and the momentum coupling coefficient is multiplied significantly due to detonation [5]. If two solid surfaces are divided by a liquid layer, the effect becomes even more pronounced at high radiation intensities because of matter density increase at the interface. Confining layer optimal characteristics can not be evaluated theoretically (tried to though [6]), and can be obtained only experimentally for specific target, cover and laser parameters.

Efficiency of different nozzles (cylinder, wedge, cone, hemisphere, paraboloid) in laser plasma injectors has been studied extensively [7]. The results demonstrate, that momentum coupling coefficient reaches its maximum at wedge [8] or cone angles less than 15° [9], slightly converging nozzle can be also effective [10]. Inputs of shockwave and ablation plume are found analytically to be about equal in a plain nozzle sealed from one side [11]. An optimum for nozzle throat to laser focal spot ratio was found to be 2.2 for cone and 2.4 for cylinder [12]. For deep ablation craters, as a kind of micro-nozzles, it was demonstrated that at reduced radiative losses, plasma temperature is proportional to the crater depth, and it decreases slower. The same relates to particle number density [13], and therefore, to the pressure. These effects become less pronounced at crater diameter increase [14] and depth-to-diameter ratio decrease [15]. Filling micro cavities with a liquid leads to a significant increase of the momentum coupling coefficient proportionally to a liquid layer thickness, but specific impulse decreases due to formation of droplets [16]. Nozzles can be used not only for reactive force increase, but as a reflective optics as well. This is possible with some difficulties though, which are mainly related to wall cooling under radiative flux much stronger than a convective one and to manufacturing of micro-nozzles of demanded quality. Annular focusing could be helpful for resolving these problems, since it leads to cylindrical plume formation [17].

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In most cases, ablation plume is ionized significantly, so electric field could be obviously used for its acceleration to reach mass averaged velocity of 10s km/s. Combined laser-electric configurations have been investigated with 3 ways of acceleration: electrostatic, electromagnetic, and electro-thermal. Control of plasma characteristics with laser radiation makes it possible (with corresponding shape of electrodes) to implement all of these types in a single device by setting parameters depending on demanded performance.

Plasma generation efficiency at laser ignition in a plate capacitor with ablating dielectric increased significantly at focal spot positioning closer to a cathode [18]: initial plume speed was the highest at the anode. Moreover, the magnetic field of up to 0.25 T appears in such capacitor [19], and current oscillations lead to the formation of at least 3 ionization waves. Combined geometric (narrow cylindrical channel) and electrostatic impact under negative potential application increased electron number density by side-compression of the plume and slowed down the recombination at late stages (>100 ns); positive potential application led to plasma radiation output at initial stages (<100 ns).

Magnetic impact has been considered for flow axis control [20], acceleration [21], compression [22, 23] and collimation [24]. Those are most pronounced for the high-energy fraction of the plume [25] and lead to the stratification and internal electric field generation [26]. Axial symmetry of the plume (this is characteristic for pure laser impact) disappears in transverse magnetic field [23]. This also leads to flow deceleration [22], since plasma tends to move along the field lines [27], and does not improve gas dynamic performance (but these effects are less pronounced for light particles [28]). Axial magnetic field starts affecting the plume structure from values of ~0.04–0.16 T [29]. For axial field of 4.5 T recoil momentum was found to double [30], most likely, due to plume compression, so the field input is about equal to laser ablation one [31]. Preliminary collimation of the flow (due to a concave surface) under multiple irradiation of the same spot [27] can be significantly improved by 1 T magnetic field. These effects can be obtained using permanent magnets, since no external energy is spent for those both absolute and specific performance can be improved.

A significant plume velocity and ionization rate increase takes place at double-pulse irradiation, but temperature increases slightly as compared to a single impact [32]. If the target surface is covered by a liquid layer, double-pulse impact has a negative effect (as compared to the uncovered target) due to the increased density of the upper layer [33]. There are several time segments characteristic for different physical processes, for which local optima can be obtained at different impact conditions.

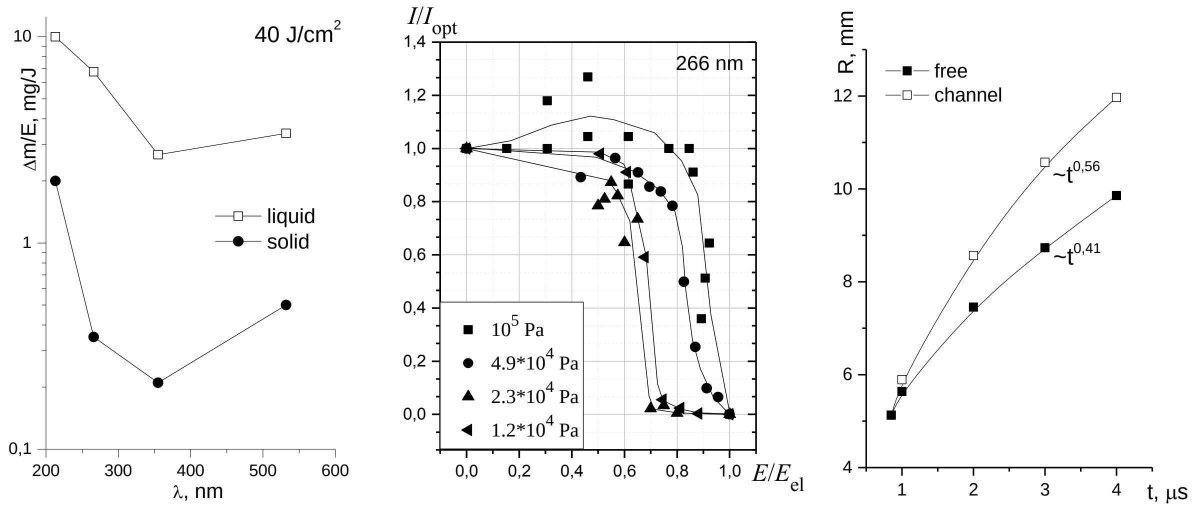
The previous paragraph considered collinear double pulse impact. In the orthogonal scheme (second pulse radiation is focused in the plume in parallel to the target surface [34]), plasma flow velocity depends on this focal spot distance from the target surface and decreases due to the interaction of flows induced by separated sources, but leads to highly effective collisional excitation and ionization [35]. In the case of the pre-pulse parallel to the target surface (with the energy which is sub-threshold for the ablation), crater volume increased only slightly and depended on the first pulse energy at 0.2–1000 μs delays (with the maximum effect reached at ~10–30 μs) [36]. Parallel scheme has been also investigated [37]. Unlike pre-pulse orthogonal one with no ablation at the first stage, the shockwave energy in the parallel scheme is higher due to spatial confinement. And unlike the collinear scheme, the second pulse goes through the region free of nanoparticles and clusters, formed by the pre-pulse, which increases the fraction of radiation absorbed by the target. Combination of these factors leads to a significant ablation rate increase; craters do not have so obvious rims as in the pre-pulse orthogonal scheme due to more effective melt drainage.

3. Experimental results

We have investigated experimentally several new ways of combined impact for laser plasma generation: irradiation of ferrofluids in axial magnetic field of 0.05–0.5 T; modification of optical properties of the liquids containing photoactivators and absorbing additives; symmetric radial confinement of the plume; laser-electric breakdown of gases. Detailed descriptions of experimental setups were published elsewhere [38, 39]. All the results were evaluated in terms of plasma generation threshold, ablation mass flow rate, recoil momentum, radiation spectral output, and the specific parameters calculated (specific impulse, momentum coupling coefficient, energy efficiency, quantum yield) [40]. Examples of combined impact benefits are presented in Figure 1.

Laser ablation rate of the ferrofluid (averaged for 10 pulses) was 0.21 g/J, in magnetic field at 140 mT it was 0.16 g/J (decrease was proportional to the viscosity increase); in glass cylindrical nozzle at 480 mT it decreased by 3 orders of magnitude down to 0.2 mg/J, and was about equal to that for solid polymer targets [41]. The nozzle had no such effect at zero magnetic field, since the droplets remained on the walls. With the field applied, the droplets moved back towards the magnet, so only evaporated matter took off. Magnetic field moving along nozzle walls could accelerate retrieval of the droplets. When ferromagnetic (steel) nozzle was used, droplets moved towards its end, since magnetic flux at the edges had the highest value. The increase of the momentum coupling coefficient under the impact on droplets in the magnetic field was ca. 30% (up to 4 mN*s/J), and energy efficiency increased by half (up to 3%), staying at low level that is characteristic for liquids. Droplets stopping is important not only for material saving and mass averaged velocity increase, but also in technology, e.g. for thin films deposition.

The efficiency of combined laser-electric impact on gases has been also investigated (more detailed results are published in this volume). Specific regimes of combined impact were discovered, leading either to the decrease or the increase of breakdown thresholds. Combined impact efficiency rate has been suggested as a relation of optical and electric breakdown threshold values at combined and pure impact. Optimal pressures for combined impact were found to be in the range of 10^2–$3*10^5$ Pa, and the efficiency gradual increase was inversely proportional to the radiation wavelength of 213–1064 nm. These results were predictable theoretically. The comparatively long-living (several microseconds) dense radiating cord was a less expected experimental finding. It was, formed at certain ratios of laser and electric arc induced shockwaves energies [42, 43]. This should increase output of short wavelength radiation, used for surface modification.



a b c

Figure 1. Examples of combined impact benefits (a – reduction of mass flow rate of photopolymer, b – reduction of breakdown threshold components under laser-electric impact at different pressures of Kr, c – reduction of shock wave deceleration in a square channel).

Spatial confinement effects were investigated using dual-wavelength interferometry and shadowgraphy of ablation plumes for 6 configurations (axial, radial, combined, static and dynamic) at ambient conditions and vacuum, linear, single- and double point focusing [44]. Generation regimes, structure and lifetimes have been evaluated for stagnating, vortex, compressed and radiating areas. Analysis of shockwaves dynamics in relatively narrow (<3 focal spot diameters) transparent channels has revealed the change of shock front dimensionality from 2D ellipsoid to a quasi-1D flat. The shockwave energy remains concentrated at a small surface, unlike fast growing hemispherical front, so the efficiency of material shock treatment can be improved.

4. Conclusion

The analysis of different combined impacts on target materials and laser plasma flows has been performed in terms of efficient energy absorption and reduction of losses, plume acceleration and collimation. Several specific methods for the improvement of laser plasma generation efficiency have been found out from previously published results. Also, several promising ways have been suggested and evaluated. The reduction of mass loss by three orders of magnitude due to droplet phase stop and retrieval has been demonstrated for laser ablation of ferrofluids in axial magnetic field and confined by paramagnetic nozzle. This finding is extremely important for surface modification plasma setups.

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