Emission and expansion features of ns and fs laser ablation plumes in an ambient environment

S. S. Harilal[1], N. Farid[1], A. Miloshevsky[1,2], P. K. Diwakar[1], and A. Hassanein[1]
[1]*Center for Materials Under eXtreme Environment, School of Nuclear Engineering, Purdue University, West Lafayette, Indiana 47907 USA*
[2]Department of Physics, *Purdue University, West Lafayette, Indiana 47907 USA*
Author e-mail address: hari@purdue.edu

Abstract: We investigated the emission and hydrodynamic expansion dynamics of ns and fs laser ablated metal plasmas in the presence of an ambient with pressures ranging from vacuum to atmospheric levels. The role of background pressure on spectral emission features, absolute line intensities, signal to background ratios, plume hydrodynamics, and ablation craters were studied. The structure and dynamics of both ns and fs plumes obtained from optical diagnostic tools were compared to continuum hydrodynamic model (CFD) results.

OCIS codes: (280.4788) Optical Sensing; (300.2140) Emission Spectroscopy, (280.5395) Plasma diagnostics

I. Introduction:

Nanosecond and femtosecond laser ablation (LA) have been demonstrated as a powerful tool for numerous applications. Some of the well-known applications are laser-induced breakdown spectroscopy (LIBS), laser-ablation inductively coupled plasma spectroscopy (LA-ICP-MS), precise micromachining, etc.[1,2] Most of the LA applications are performed in the presence of an ambient gas at various pressure levels. So it is important to understand the fundamental physics of ns and fs laser–material interaction in the presence of an ambient considering its ever growing and attractive applications in various fields. Numerous studies have been available in the literature showing the interaction between highly transient ns laser plasmas and background gas, however, limited studies have been carried out for developing clear-cut understanding of fs LA plume interaction with an ambient. The LA plumes expand freely and adiabatically in vacuum. However, the entire hydrodynamics of plume expansion becomes much more complex in the presence of an ambient gas causing effects such as plume splitting, sharpening, confinement, and the formation of shock waves and internal plume structures. In the presence of moderate to high ambient pressures, multiple shock waves will be formed, propagating both in the forward and backward directions. Moreover, the plasma plumes becomes more collisional in the presence of an ambient leading to further excitation of plume species as well as enhancing excited molecular species formation mainly through three-body recombination.[3] In this work, we made a comprehensive comparison of ns and fs LA plume emission and expansion dynamics in the presence of an ambient under varying pressure levels using fast gated photography, time and space resolved emission spectroscopy and shadowgraphy. The internal structures and plume hydrodynamics expansion features obtained from the experiments were compared with a continuum hydrodynamic model.

II. Experimental and modeling approach:

Laser ablation on a Cu target was performed using either pulses from a Nd:YAG laser (6 ns, 1064 nm) or a Ti-Sapphire amplifier laser system (40 fs, ~ 800 nm). Due to a pulse duration difference of approximately five orders of magnitude between typical ns and fs laser systems, comparative laser ablation studies using similar laser power densities were not possible. However for analytical applications, the power density used for fs LA is typically in the ~10^{13}-10^{14} W/cm^2 range, while for ns lasers this is often in the range ~10^9-10^{10} W/cm^2. So for the present studies we used laser intensities of 5×10^9 W/cm^2 (100 mJ/pulse) and 3 $\times10^{14}$ W/cm^2 (4.5 mJ/pulse) for ns and fs LA, respectively. The diagnostic tools used in this study include space and time resolved optical emission spectroscopy (OES), fast gated imaging, focused shadowgraphy and crater morphology analysis. The modeling of the expansion of LA plume in the presence of ambient gas is carried out using computational fluid dynamics (CFD) simulations.[4] The shape of the LA plume, and position of shock front obtained from the modeling results were compared with experimental results obtained from fast gated imaging and shadowgraphy.

III. Results and Discussion:

We used fast gated imaging employing intensified CCD for capturing hydrodynamic expansion features of ns and fs LA plumes at various pressure levels. Typical intensity normalized images obtained at 500 ns after the onset of plasma formation for fs and ns LA is given in Fig. 1 at various pressure levels. All the images are spectrally integrated in the visible region. Instead of spherical expansion noticed in ns LPP, fs LPP plumes are found to expand with a much stronger forward bias in directions normal to the target surface. Though both ns and fs LA plumes expand freely and rapidly in the forward direction, the persistence of the ns LA emission is found to be significantly higher for ns LA plumes compared to fs LA plumes. This could be related to increased laser-plasma coupling rather than laser-target coupling. The use of higher laser energy for generating ns LA plume (100 mJ/pulse) may also contribute higher persistence since a major part of the ns laser energy will be utilized for plasma reheating. Both ns and fs LA plumes expand freely and adiabatically in vacuum. The images given for moderate to higher ambient pressure levels clearly show the collisional effects indicated by enhanced emission from the plume and confinement. Enhanced emission can be explained by increased collisions between plume species with ambient gas species resulting in the excitation of plume species. In this pressure range both ns and fs LA plumes showed spherical geometry. The difference is plume geometry and internal structures are more distinct with increasing pressure, however, plume deceleration is apparent in both cases. For fs LA, the plumes attain a spherical geometry at moderate pressure levels (0.1-1 Torr) and at 10 Torr the plume changed into a torpedo-like shape. However, the ns LA maintained approximately spherical morphology regardless of the ambient pressure levels. At still higher pressures, both the plasmas are confined and effective length is reduced to a few mm.

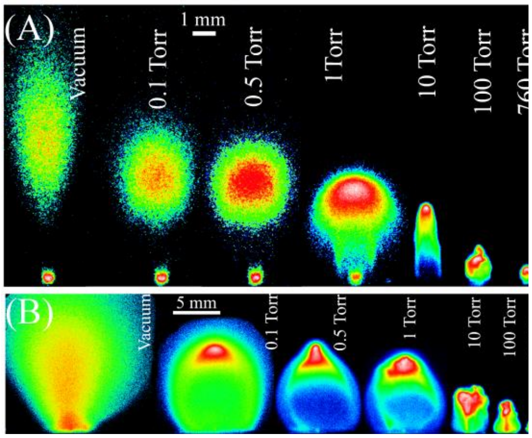

Figure 1: Fast gated images of LA plume generated by (A) fs and (B) ns lasers. There images were captured at 500 ns after the onset of plasma generation at various pressure levels.

We also evaluated the visible spectral features of Cu plasmas at various pressure levels. Though the spectral studies showed significant enhancement in signal for all emission lines and optimum conditions for maximum emission are found to be at ~ 300 Torr and ~ 600 Torr, respectively, for fs and ns LA, the best S/B was observed at ~ 20-50 Torr levels for both plumes because of disproportional continuum emission at higher pressure levels. The emission spectra content showed significant differences for ns LA with increasing pressure by the presence of additional ionic and ambient gas lines, while for fs LA only incremental increase in spectral intensity and continuum were seen. This can be understood considering large differences in excitation temperature with pressure for ns LA in comparison with fs LA. Comparing the crater morphologies, the ns LA craters showed drastic differences in images obtained in vacuum and atmosphere conditions. The common features at atmosphere and in vacuum are the re-solidification of molten material. In the presence of background gas, plasma shielding becomes dominant, which not only reduces the mass ablation, but also absorbs the energy from the incoming laser, resulting in higher temperature plasma in front of the target surface. However, the crater morphologies for fs LA under vacuum and ambient gas cases are found to be similar.

We used shadowgraphy to investigate the LA shockwave formation at high ambient pressure levels. The structure and dynamics of the shockwave obtained from the optical diagnostic tool were compared to numerical simulation employing CFD model. Typical shadowgram and contour map of the pressure field obtained using CFD model at 500 ns after the onset of plasma formation when the Cu plume expanded into 1 atm Ar is given in Fig. 2. The results clearly show that the main features of plume expansion in background argon observed in the experiments can be reproduced by the CFD model. The shape of plasma plume, the position of shock front are found in good agreement with the experimental results.

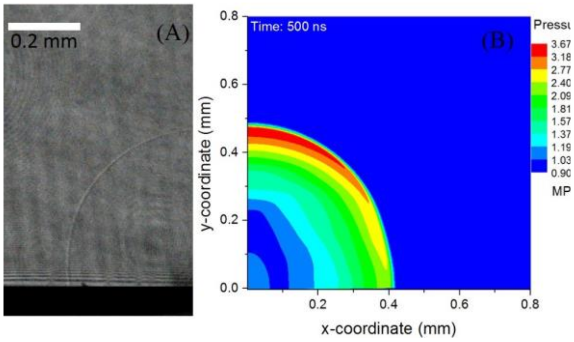

Fig. 2: (A) Shadowgram recorded at 500 ns after fs LA at 1 atm Ar. The contour map of the pressure field obtained using CFD modeling is given in (B)

Acknowledgments: This work is supported by the US DOE NNSA under award number DE-NA0001174 and NSF PIRE program.

References: [1]N. L. LaHaye *et al.*, J. Appl. Phys. **114**, 023103 (2013); [2]P. K. Diwakar *et al*., Spectrochim. Acta B **87**, 43 (2013); [3]N. Farid *et al.*, Appl. Phys. Lett. **103**, 191112 (2013); [4]S. S. Harilal *et al.*, Phys. of Plasmas **19**, 083504 (2012).