Comparison of Laser-Induced Ablation Thresholds of Tantalum in a Wide Range of Pulse Durations

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Short intense laser pulses are proposed to be used for in-situ first-wall diagnostics of upcoming fusion plasma devices as described in Ref. [1]. In particular for laser-induced breakdown spectroscopy (LIBS) pulse durations of ns to ps duration are used today. While shorter laser pulses allow a higher depth-resolution of fuel retention the use of sub 10 fs can be an option. The characteristics of the interaction of these few cycle laser pulses with solid target material at high intensities differs explicitly from nanosecond or picosecond lasers and even from pulse durations down to 100 fs [2]. It is the goal of our work to identify the basic processes governing ablation and plasma formation with sub 10 fs laser pulses on high-Z materials. Here we give an overview and comparison of the different pulse duration regimes by presenting data on the ablation thresholds of pure tantalum ($Z = 73$) and the plasma expansion characteristics. Craters depth and diameter caused by the laser pulses are analyzed via confocal microscopy as a function of fluence and intensity. For the measurements a nanosecond laser of 280 mJ pulse energy from Dalian University of Technology and a picosecond laser of 46 mJ pulse energy from Forschungszentrum Jülich are used. These long-pulse results are compared to a mode-locked Ti:Sa laser providing 8 fs pulses with a pulse energy of 1 mJ from the Institute of Laser and Plasma Physics in Düsseldorf. The laser provides intensities of up to $10^{18}\,\mathrm{W/cm^2}$ which can be tuned over four orders of magnitude using a reflective attenuator. In addition the plasma expansion behavior observed by a gated camera for different pulse durations helps us to obtain a deeper understanding of the basic processes governing ablation and plasma formation in the sub 10 fs regime in comparison with even longer pulses.

Ablation Threshold

The ablation experiments are executed in vacuum chambers (10^{-5} to 10^{-6} mbar) on a tantalum samples of 99.9 % purity and relatively high roughness of $S_a = (0.7 \pm 0.3)\,\mu\mathrm{m}$ to ensure comparable properties to the fusion device first-wall. The laser pulses are focussed by lenses (ns- and ps- laser) and an Off-Axis Parabola (fs-laser) to the surface. As presented in Ref. [3] the ablation

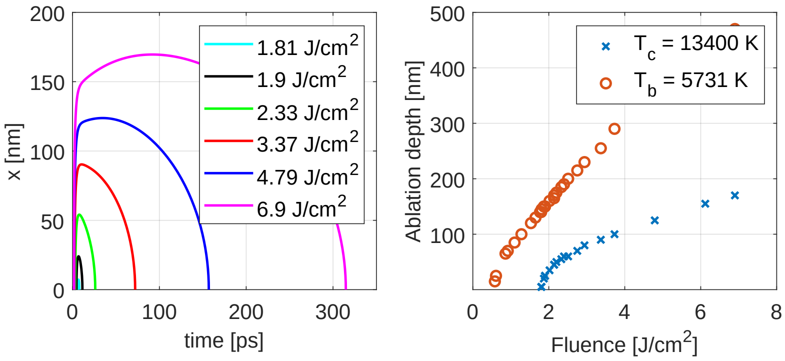

Figure 1: *(left) Contour plot of the critical temperature T_c along the sample depth x with respect to simulation time for different laser fluences. (right) Ablation depth calculated by the depth of critical temperature and boiling temperature T_b*

threshold laser fluence F_{th} decreases and the threshold intensity I_{th} increases for shorter pulse duration. In the experiment an ablation threshold fluence of $F_{th} = 0.17\,\mathrm{J/cm^2}$ is found for the fs-laser. Moreover it appears that the craters produced by 200 ultrashort laser pulses are of high accuracy exhibit steep edges compared to the nanosecond craters. It can be predicted that the heat-affected zone (HAZ) that is caused by the ns-laser limits the depth resolution of material composition analysis in different target layers. To overcome this issue in impurity investigation of the plasma-facing surface of ITER (International Thermonuclear Experimental Reactor) or EAST (Experimental Advanced Superconducting Tokamak) shorter laser pulses should be used.

To be able to give a prediction of the ablated volume for varying pulse energy of single ultrashort laser pulses a two-temperature model is used as a numerical approach given amongst others by Ref. [4]. In our one-dimensional model the thermophysical properties of the material are updated with rising electron and lattice temperature as proposed in Ref. [5] due to the electron density of state function. The model is executed for different laser fluences with a pulse duration of 8 fs and 790 nm central wavelength. Here a x-grid size of 1 µm with increments of $\mathrm{d}x = 5\,\mathrm{nm}$ and $\mathrm{d}t = 0.1\,\mathrm{fs}$ are used. It can be supposed that the material will be removed or ablated from the ambient sample, when the lattice temperature exceeds the temperature at the thermodynamic critical point ($T_{c,\mathrm{Ta}} \approx 13400\,\mathrm{K}$, Ref. [6]). At this point the high pressure in the ablation region is released by mechanical expansion and adiabatic cooling Ref. [7]. In Fig. 1 the ablation yield can be predicted using the depth where T_c is reached. In the contour plot (left) the time and fluence dependent critical temperature limit is shown which rises with higher laser fluence. The right plot shows the maximum depth of boiling temperature (red) and critical tem-

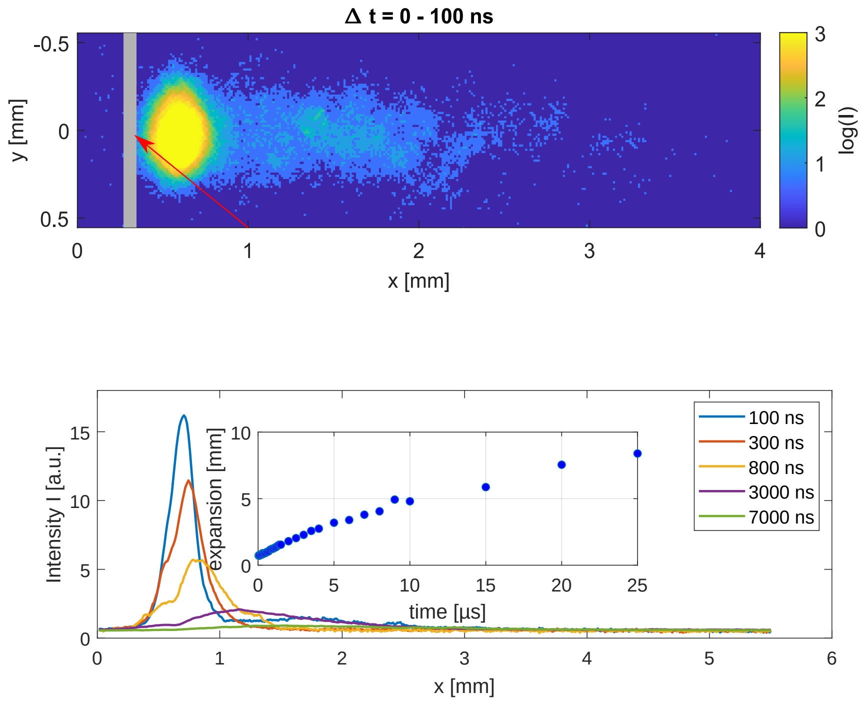

Figure 2: *(top) Image of the expanding plasma in the time frame of* Δt *after the interaction with a* 8 fs *laser pulse of pulse energy* $E = 0.61\,\mathrm{mJ}$ *and beam diameter* $D_0 = 0.2\,\mathrm{mm}$. *The incoming laser pulse is indicated in red and the Ta-sample position in grey. (bottom) Accumulated lineouts of the expanding plasma at five selected times and the plasma front expansion distance with respect to time in the inset.*

perature (blue) with respect to the laser fluence. The threshold fluence can be approximated as $F_{th} = 1.8\,\mathrm{J/cm^2}$ following the blue data points, which is distinctly higher than the experimentally observed parameter for the rough sample. Note that the simulation is executed with perfect reflection conditions due to Drude-Lorentz model. In principle this code is able to predict the ablation volume for given laser parameters and will be used for further studies on the ablation process.

Plasma Plume Expansion

Using a gated optical system consisting of a CCD camera (*Allied Vision*) attached to a micro-channel plate (MCP) image intensifier (type II18G, *Lambert Instruments*) equipped with a photo-lens, the plasma plume is observed tangential to the sample surface. The MCP gate delay of 40 ns to 30 µs are used to observe the temporal evolution of 420.5876 nm and 420.6404 nm Ta I spectral lines and continuum emission of nanopartincles, which were selected by an optical band-pass filter having 420.6 nm central wavelength and FWHM of 1.5 nm. In the top part of Fig. 2 the plasma plume is shown for a time frame $\Delta t = 100\,\mathrm{ns}$ after the laser surface interaction with a logarithmic color scale. In this time frame we observe a fast plume expanding into vacuum while the main part is following with a lower velocity. To get a better overview on this phenomena we show lineouts for five different time frames in the second part of the figure. The accumulated lineouts are generated along the expansion direction within the first 7 µs. The intensity values are divided by the used MCP gate width to be able to compare the plots. To find the expansion velocity of the plume the plasma front is exhibit by an exponential fit to the falling edge. In the inset of this figure the positions where the intensity decreased to 37 % are plotted with respect to the corresponding time. It follows that the plasma plume is propagating with an average velocity of 350 m/s. We can compare this finding to the work of Harilal et al. [8], who observed the expansion of tungsten plasma induced by femtosecond pulses. The measured velocity indicates that we observed the continuum radiation of expanding nanoparticles in this second phase after 100 ns. In the first expansion phase most likely the spectral lines of pure tantalum can be observed. To prove this claim an experiment with a gated spectrometer is needed.

Conclusion and Summary

The presented experiments and simulations give a wide overview over well described processes in the context of pulsed laser ablation on the high-Z material tantalum. All these findings are necessary to establish the application of short or ultrashort laser pulses to analyze the plasma-facing surface of a fusion reactor in future experiments. The comparison of the ablation yield per pulse for pulsed lasers of different pulse duration shows that picosecond or femtosecond lasers should be used in this context, because of the high accuracy of the ablated volume. A possible LIBS experiment for impurity scanning on the plasma-facing surface will bring a high depth resolution with shorter laser pulses.

To be able to apply a LIBS setup to a sub 10 fs laser it is essential to know the plasma expansion behavior after the interaction. With the gated plasma image experiment we are able to predict a time frame where it is possible to observe characteristic spectral lines of the expanding tantalum plasma to avoid the dominant continuum radiation caused by the emerging nanoparticles.

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