Unveiling the significance of spallation layer redeposition during ultrashort pulse ablation in liquid

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Ultrashort laser ablation is governed by two main ablation mechanisms, namely spallation and phase explosion. The former of the two only occurs when the stress confinement condition is met, which states that the time for heating of the material, controlled by either the pulse duration or electron-phonon coupling time, must be shorter than the acoustic relaxation time. While spallation significantly contributes to the ablation efficiency during laser ablation in air, simulations predict that a large amount of material ejected via spallation during laser ablation in liquid (LAL) is redeposited to the target surface [1]. As this mechanism would reduce the ablation volume during LAL, a profound understanding of the significance of material redeposition is crucial to understand LAL efficiency and further upscale the process.

In this study we aim to experimentally verify spallation layer redeposition and quantify the magnitude of redeposited material for Au and $Fe_{0.5}Ni_{0.5}$. For this pump-probe microscopy (PPM) [2] and ablation efficiency measurements are carried out in air and water at different pulse durations. By varying the pulse duration with a pulse-stretcher and electron-phonon coupling time with the choice of material, stress confinement and thus the contribution of spallation can be controlled.

The time-resolved PPM experiments clearly demonstrate spallation layer redeposition, which up until now was only shown by computational methods. Furthermore, the control of stress confinement allows us to determine the significance of spallation layer redeposition, amounting to more than 80 % of the ablated material. Our results clearly show that spallation layer redeposition during LAL is the main mechanism limiting ablation efficiency. Based on this fundamental finding, strategies beyond utilizing stress confinement, such as increasing fluence or employing double pulses may be formulated to reduce redeposition, increase ablation efficiency, and further scale up the LAL process.

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References: [1] C. Chen, L. Zhigilei, Appl. Phys. A 129, 288 (2023), [2] M. Spellauge, C. Doñate-Buendía, S. Barcikowski, B. Gökce, H. Huber, Light Sci. Appl. 11, 68 (2022)