Time resolved studies reveal the origin of the unparalleled high efficiency of one nanosecond laser ablation in liquids

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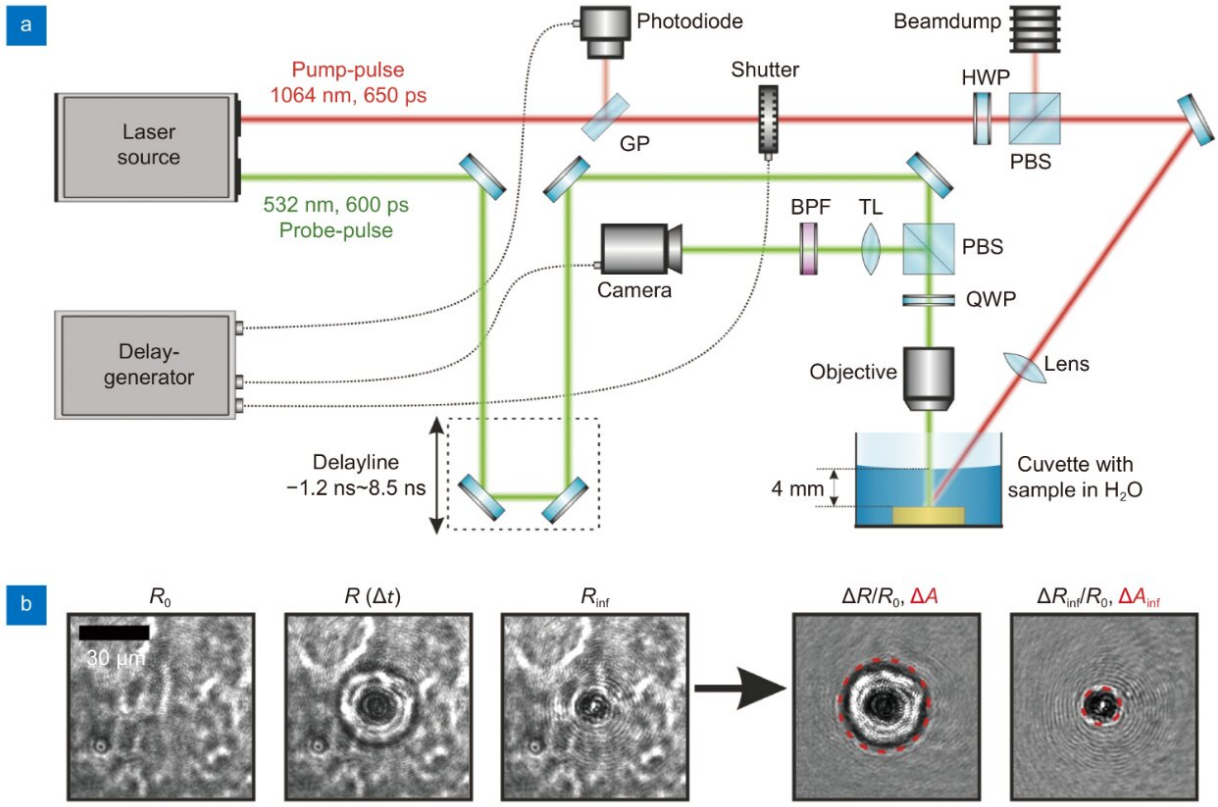

(a) Pump-probe microscopy setup for the ablation in water. For the analysis of the ablation process in air, no cuvette is used, otherwise the setup is the same. (b) Image post processing of the three recorded images. The images of the pristine surface (R0), during the ablation process (R(Δt)) and after the ablation process has finished (Rinf) were used to calculate the transient (ΔR/R0) and final state (ΔRinf/R0) relative reflectivity change. The red dashed circles mark the

transient laser-modified area ΔA and the final laser-modified area ΔAinf. Credit: Compuscript Ltd

Laser ablation in liquid is a scalable nanoparticle production method in areas like catalysis and biomedicine. Different energy dissipation channels, such as absorption by the liquid and scattering at the ablation plume and cavitation bubble, lead to reduced laser energy available for nanoparticle production. Ultrashort pulse durations cause unwanted effects. A study, recently published in in *Opto-Electronic Advances*, explored pico- to nanosecond pulse duration regimes and found that pulse durations around 1–2 ns enable the most efficient laser ablation in liquid.

Laser ablation in liquid (LAL) is a versatile method for synthesizing nanoparticles (NPs), enabling the production of ligand-free colloids. Compared to chemically synthesized colloids, particles generated by LAL exhibit a higher signal-to-noise ratio. They are suitable as reference materials for nano-toxicological assays, enable the decoration of microparticles for 3D printing, and have high potential in heterogeneous catalysis. Furthermore, with LAL, it is possible to produce molar fraction series of alloy NPs from pressed micro powder targets, even if an element miscibility gap exists.

Although laser-generated particles show advantages over chemically synthesized NPs, only a few commercial distributors offer laser-generated NPs. One reason might be that LAL only becomes economically more feasible than chemical synthesis for nanoparticle productivities exceeding 550 mg/h (for gold). High LAL productivities can only be achieved with high-power picosecond laser systems (>200 W). The high-power laser systems must operate at high repetition rates of several MHz. Fast polygon scanners are required to leverage the

available laser power effectively. The process complexity and high investment costs explain why only a few commercial distributors offer LAL-generated NPs.

Besides scaling up the productivity by increasing the laser power, careful tuning of the laser pulse duration represents another route to optimize the LAL process. Nanosecond LAL (ns-LAL) has achieved similar power-specific ablation rates as picosecond LAL (ps-LAL). However, mostly ~ 10 ns lasers are employed, while only a few publications investigated the lower pulse duration limit (~ 0.6 ns) that cavity-length limited, Q-switched lasers deliver.

In contrast to ablation in air, additional energy loss mechanisms occur during LAL. These mechanisms can be divided into two categories. One category represents losses due to interactions between the laser pulse with the water layer. The other category shows losses from interactions with induced ablation dynamics. The extent of these loss mechanisms' influence on LAL productivity strongly depends on the laser pulse duration.

Optical breakdown within liquid occurs when the laser pulse generates a plasma with a certain critical electron density through multiphoton ionization followed by cascade ionization. The emerging plasma absorbs a large portion of the pulse energy (~ 50%). Only the trailing pulse edge experiences plasma absorption. Thus, a larger portion of the pulse energy (~ 80%) is transmitted through the optical breakdown volume.

NPs present in the liquid can significantly lower the threshold fluence for the optical breakdown of ns pulses. LAL performed with ultrashort laser pulses is often accompanied by non-linear effects. When the losses induced by non-linear effects and optical breakdown are combined, up to 70% of the pulse energy may be extinct before reaching the target surface. Furthermore, there are attempts to account for NP

concentration-dependent shielding effect.

It was found that the ablation efficiency varies strongly depending on the experimental conditions. The efficiency is maximal for laser systems operating at a pulse duration of one nanosecond compared to a few ps or >5 ns. Neither a mechanistic explanation has been given, nor single-pulse conditions were applied. It should be emphasized that the optimal pulse duration of ~1 ns is readily available by Q-switching of short-cavity laser resonators. Optimal LAL processing may be possible by utilizing low-cost Q-switched lasers, avoiding the high investment cost of mode-locked femto- and picosecond lasers.

Laser ablation synthesis of colloids in liquids is a promising nanomaterial fabrication method. Short pulsed LAL at around ten ns pulse duration is similarly efficient to ultrashort pulsed LAL. In both cases, intra-pulse attenuation caused by the liquid or plume, vapor and cavitation bubble limits efficient energy deposition. The researchers determined characteristic shielding times, translating into optimal laser pulse durations. The reduction of shielding effects during the laser pulse irradiation increases the ablation efficiency observed for 1 ns LAL of Au. Cavity-length limited, Q-switched nanosecond lasers may advance LAL to even higher efficiencies.

More information: Sarah Dittrich et al, Time resolved studies reveal the origin of the unparalleled high efficiency of one nanosecond laser ablation in liquids, *Opto-Electronic Advances* (2022). DOI: 10.29026/oea.2022.210053

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