Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for

Clear observation of the formation of nanoparticles inside the ablation bubble through a laser-induced flat transparent window by laser scattering

Kota Ando[a] and Takashi Nakajima[a*]

[a]*Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan*

*Corresponding author: nakajima@iae.kyoto-u.ac.jp

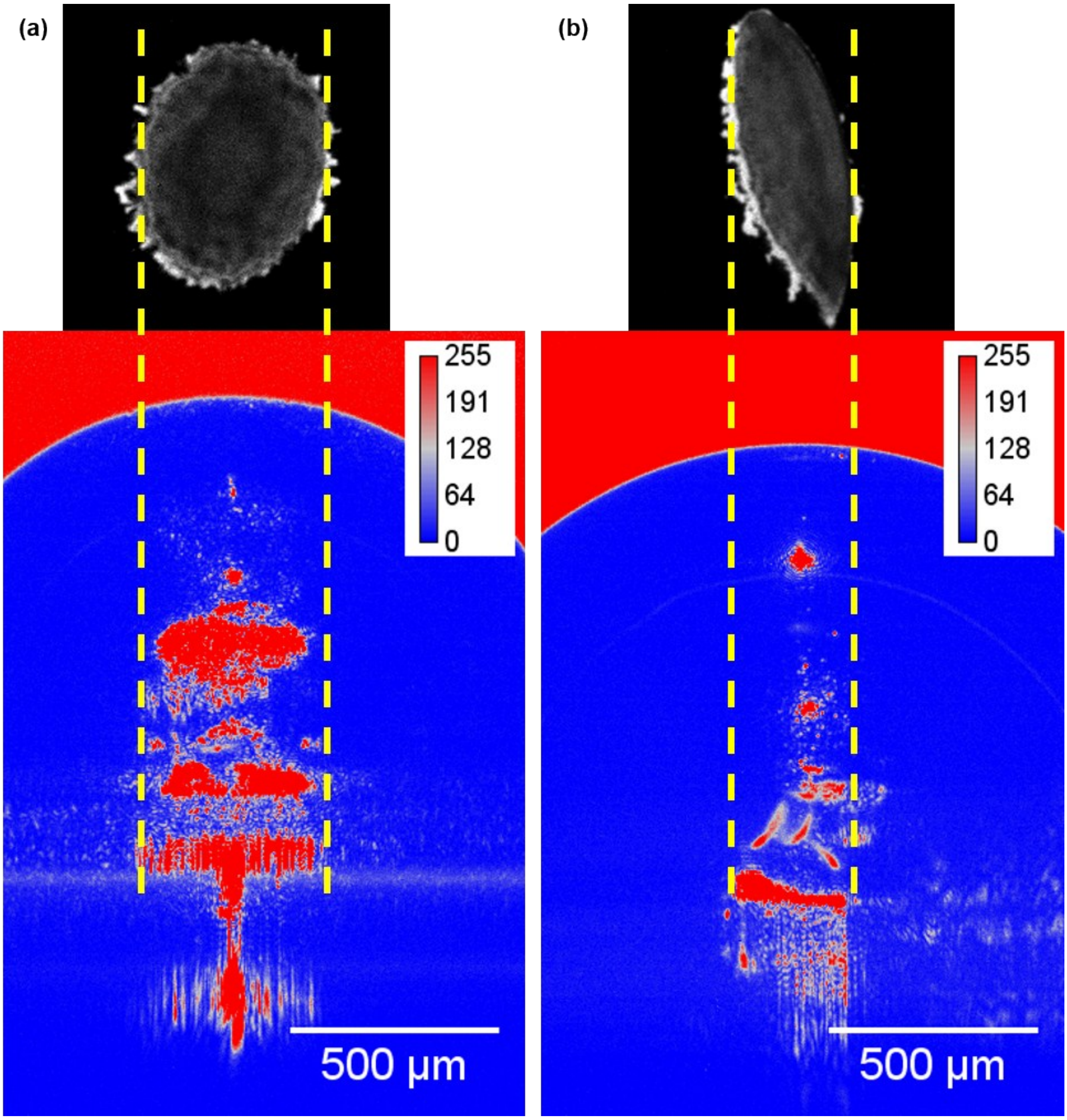

Figure S1. Correlation of the shape of ablation hole and laser scattering image for the cases of (a) round and (b) intentionally elongated ablation holes under the same pump pulse fluence. Both scattering images are processed by ImageJ for better visualization. As the vertical yellow dashed lines suggest the probe pulse illumination is more homogeneous through the round ablation hole, while the influence of the film rims which may have been created around the hole upon ablation is not visible.

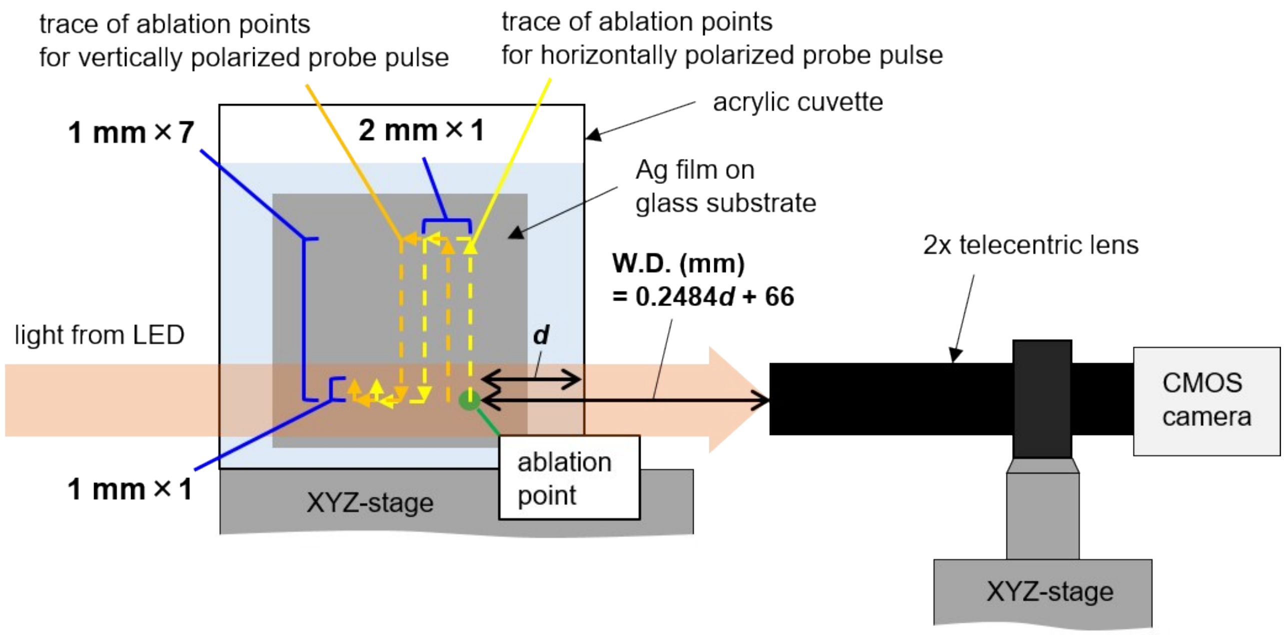

Figure S2. Side view of the acrylic cuvette (50 mm × 50 mm × 50 mm) on a motorized XYZ-stage where the pump pulse is horizontally (i.e., vertically-to-paper) incident onto the Ag film. After every measurement with a set of pump pulse and time-delayed probe pulse together with the CMOS camera exposure, the cuvette and hence the Ag film is translated by 1 mm with a stepping motor, as shown by the dashed yellow arrow. Then, water around the next ablation point is flushed away by small amount of pure water in a micropipette. Since the total amount of water in the cuvette is so much for the total number of pump pulses employed for the scattering experiments the above procedure works well to wash out the fragments as well as nanoparticles produced by the previous pump pulse from the new ablation point. The position of the CMOS camera is moved by 0.5 mm horizontally only when the acrylic cuvette is moved horizontally by 2 mm to correct the focal position of the objective lens.

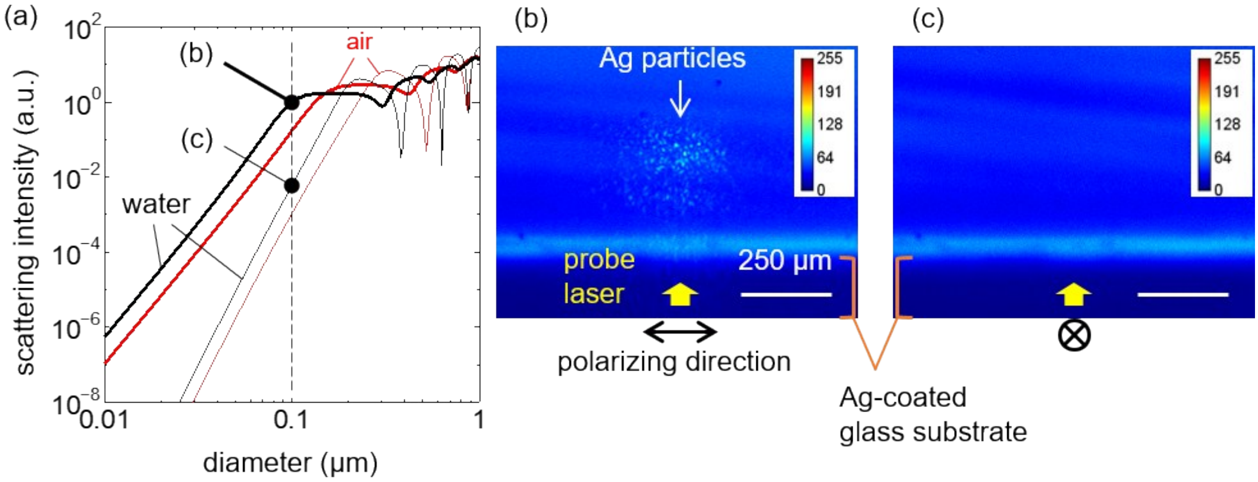

Figure S3. (a) Scattering intensities of Ag particles in water (black lines) and in air (red lines), respectively, as a function of Ag particle diameter at the scattering angle of 90 degrees. The polarization direction of the probe laser is assumed to be horizontal (thick lines) or vertical-to-paper (thin lines). All the intensities are normalized by that of Ag particles with a diameter of 100 nm in water for the horizontally polarized probe laser. (b) and (c) Laser scattering images of Ag particles with a diameter of 100 nm (Sigma Aldrich, lot number: 730777, 0.02 mg/mL) in water. The brightness of the two images is doubled for better visibility. The 10 μL colloidal solution of Ag particles is ejected just above a laser-created transparent window on an Ag film on a glass substrate which immersed in water purified by a syringe filter (mean pore size 0.22 μm, Membrane Solutions). Upon making the local number density of the colloidal solution sufficiently high only above the transparent window, we send the linearly polarized probe laser through the transparent window. The polarization direction of the probe laser is indicated by the arrow and cross marks in panels (b) and (c).

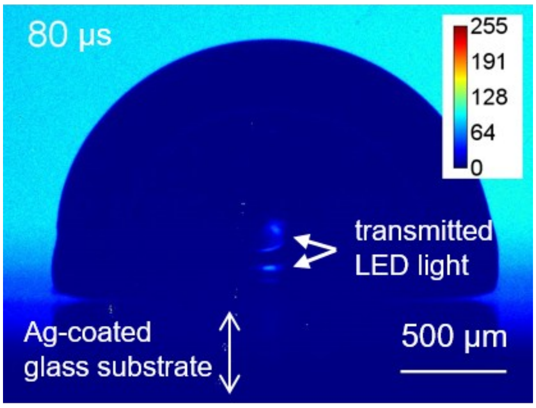

Figure S4. Shadowgraph image of the ablation bubble at 80 µs delay after the pump pulse where the bubble size is *maximum* and hence the curvature of the bubble surface is smallest. Note that the probe laser is off for this image. The dim blurred spots around the centre of the bubble originate from the LED light transmitted through the bubble. Note that they appear at the height where the normal of the bubble surface is parallel to the optical axis from the LED light to the camera.

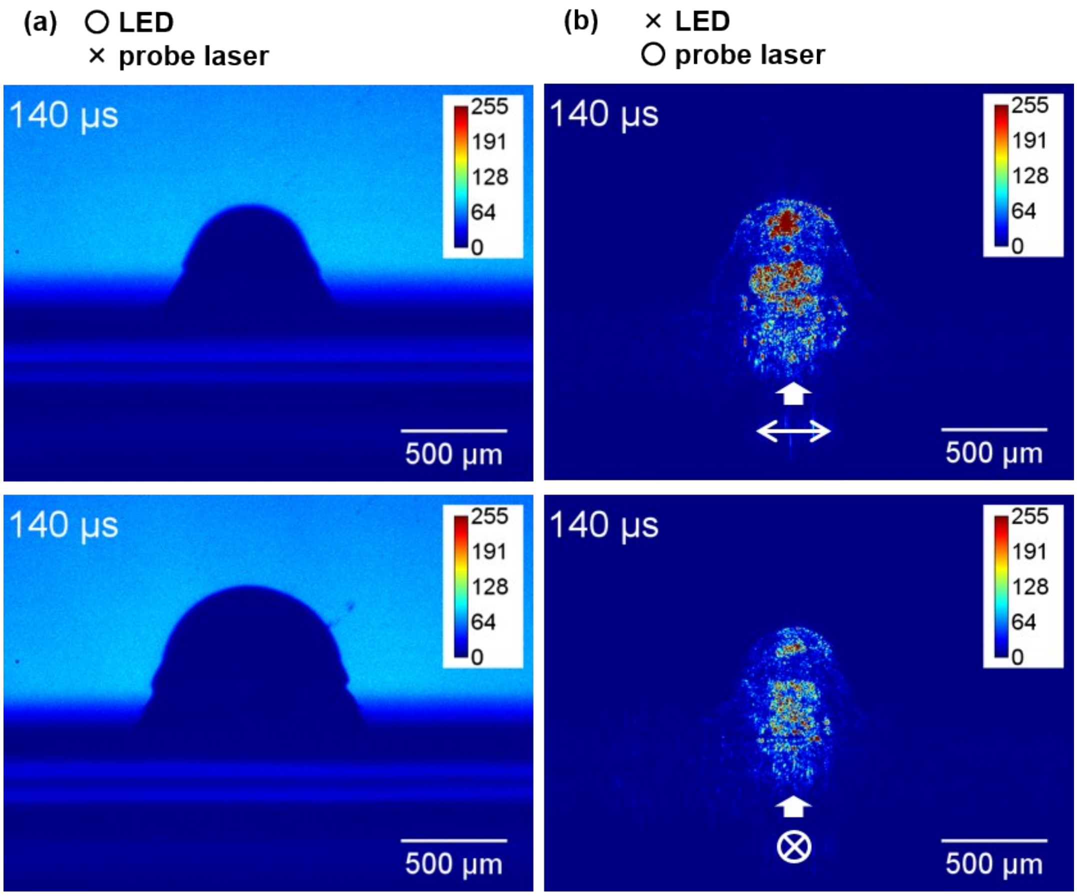

Figure S5. (a) Shadowgraph and (b) laser scattering images of the *shrinking bubble* taken at 140 μs delay with the LED light and probe pulse only, respectively, to clarify the influence of LED light illumination on the laser scattering image. The dim and blurred spots which appear around the centre of the bubble at 80 μs delay (Fig. S3) are not seen anymore because the diffraction of the LED light at the higher curvature of the shrinking bubble surface severely attenuates the transmission of the LED light toward the camera.