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Supplementary data

Bubble generation and molecular crystallization at solution surface by intense continuous-wave laser irradiation

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Experimental procedure

We prepared pure H₂O and L-Phe H₂O unsaturated solution as the sample. The L-Phe solution was prepared by mixing L-Phe powder of 17.5 mg in H₂O of 1.0 g, and its saturation degree was estimated at 0.58. The mixture was vigorously shaken for several hours and then gradually cooled down to room temperature (25 °C). This sample preparation was carried out automatically with a programmable shaking machine (TAITEC, Bioshaker BR-21UM). Pure H₂O or the sample solution was poured into a hand-made glass container of which side wall and bottom are highly hydrophilic, and a solution thin film with about 150 μm thickness was prepared. The glass container was closed by a plastic cap and set on the sample stage of an inverted microscope (Olympus, IX71).

Figure S1 shows a schematic illustration of the optical setup used in this study. A linearly polarized near-infrared cw laser beam from an Nd³⁺:YVO₄ laser (Spectra-Physics, BL-106C,

$\lambda = 1064 \text{ nm}$) was used as a trapping light source. The laser beam was introduced to the inverted microscope and focused into a sample solution through an objective lens (60 magnification, numerical aperture; 0.90). The laser power throughout the objective lens was tuned at 0.5–1.1 W by rotating a half-wave plate positioned in front of a polarizing beam splitter. The half-wave plate was set in a motorized holder, so that it can be rotated with a computer. A green laser (Altechna, LD-WL206) was also introduced to the microscope in the optical pass same as the 1064-nm laser in order to adjust the focal position before switching on the trapping laser. The green laser was focused at the air/solution interface. Then the green laser was turned off, and the 1064-nm laser was switched on.

The bubble generation behavior was observed with a charge-coupled device (CCD) video camera (WATEC, WAT-231S2) under halogen lamp illumination. The field of view of this camera was $80 \times 60 \mu\text{m}^2$. In order to observe a wider area, we used another CCD camera (The Imaging Source, DFK41AU02) coupled with a lens tube (Micronet, NY-CZ71461). A transmission image was sequentially captured at a video rate. We used an electron multiplying CCD (EMCCD) camera for investigating the bubble dissolution dynamics after switching off the laser. The camera was synchronized with a shutter (Lambda, IQ25-566) of which response time is 8 msec. A transmission image of $35 \times 35 \mu\text{m}^2$ was sequentially collected at every 6 msec after turning off the laser.

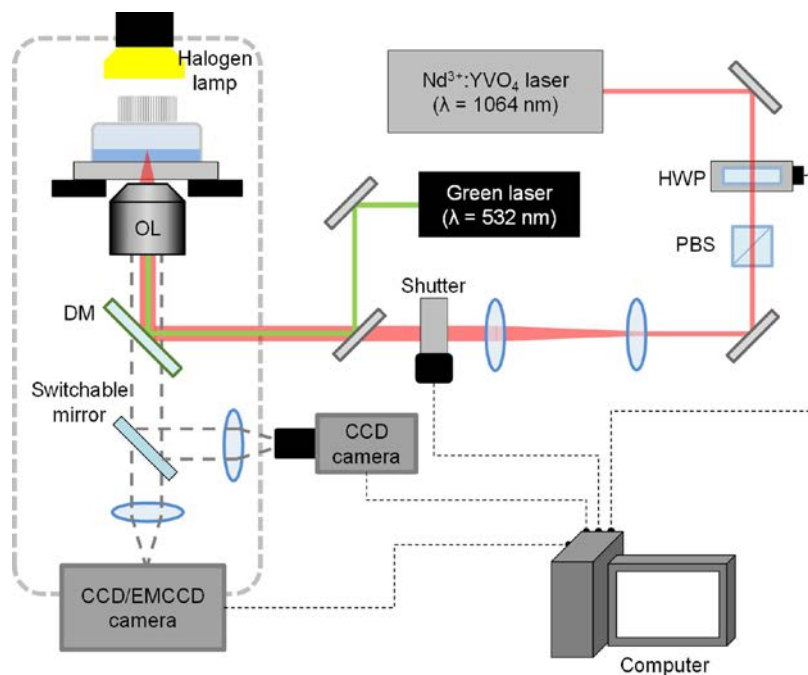


Fig. S1. A schematic illustration of optical system. HWP; half-wave plate, PBS; polarizing

beam splitter, DM; dichroic mirror, OL; objective lens.