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Author(s)	Chen, Jim Jui-Kai; Yuyama, Ken-ichi; Sugiyama, Teruki et al.
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Supplementary data

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

Jim Jui-Kai Chen[1], Ken-ichi Yuyama[1,2]*, Teruki Sugiyama[1,3,4]*, and Hiroshi Masuhara[1,3]*

[1]*Department of Applied Chemistry, National Chiao Tung University, Hsinchu 30010, Taiwan*

[2]*Research Institute for Electronic Science, Hokkaido University, Sapporo, Hokkaido 001-0020, Japan*

[3]*Center for Emergent Functional Matter Science, National Chiao Tung University, Hsinchu 30010, Taiwan*

[4]*Division of Materials Science, Graduate School of Science and Technology, Nara Institute of Science and Technology, Ikoma, Nara 630-0192, Japan*

E-mail: yuyama@es.hokudai.ac.jp, sugiyama@g2.nctu.edu.tw, masuhara@masuhara.jp

Experimental procedure

We prepared pure H_2O and L-Phe H_2O unsaturated solution as the sample. The L-Phe solution was prepared by mixing L-Phe powder of 17.5 mg in H_2O of 1.0 g, and its saturation degree was estimated at 0.58. The mixture was vigorously shacked for several hours and then gradually cooled down to room temperature (25 °C). This sample preparation was carried out automatically with a programmable shacking machine (TAITEC, Bioshaker BR-21UM). Pure H_2O 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 Nd^{3+}:YVO_4 laser (Spectra-Physics, BL-106C,

λ = 1064 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 × 60 μm^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 × 35 μm^2 was sequentially collected at every 6 msec after turning off the laser.

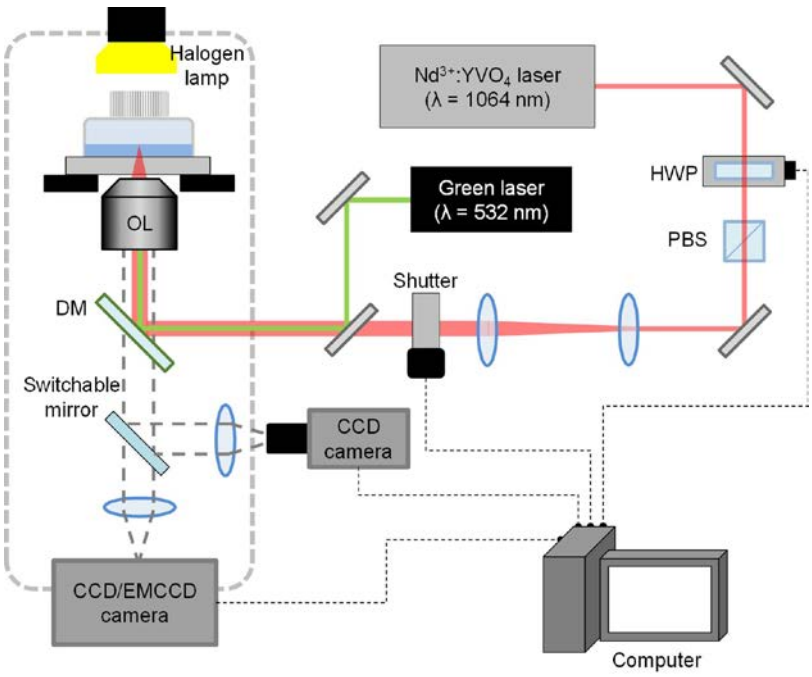

Fig. S1. A schematic illustration of optical system. HWP; half-wave plate, PBS; polarizing

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