High-Speed Polarization Imaging of Laser Ablation-Induced Crystallization of Ice in Supercooled Water

Hozumi Takahashi¹, Norifumi Fuji¹, Tomohiko Tateshima², Ryutaro Shimada², Takashi Onuma2, Yuka Tsuri3, Yusuke Mori1, Mihoko Maruyama1,4, Yoichiroh Hosokawa3, and Hiroshi Y. Yoshikawa1,*

 Osaka University, 2-1, Yamada-oka, Suita, Osaka, Japan Photonic Lattice, Inc., 6-6-3, Minami-Yoshinari, Aoba-ku, Sendai, Miyagi, Japan Nara Institute of Science and Technology, 8916-5, Takayama-cho, Ikoma, Nara, Japan Kyoto Prefectural University, 1-5, Hangi-cho, Shimogamo, Sakyo-ku, Japan * Corresponding author email: hiroshi@ap.eng.osaka-u.ac.jp

Laser ablation of supersaturated/supercooled liquid has shown promise for controlling crystal nucleation of various materials in a spatiotemporal manner [1-3]. As for the mechanism, previous studies suggest that macroscopic morphological changes (e.g., generation of shockwaves and cavitation bubbles) via laser ablation of liquid plays a crucial role in enforcing crystal nucleation. In particular, laser ablation induced by the focused irradiation with an ultrashort laser pulse can act as a well localized, photomechanical stimulus, which allows for the fine monitoring of crystallization dynamics from targeted places [1-3]. In fact, we recently succeeded in motoring microsecond and micrometer-scale dynamics of ice crystallization that was triggered by the focused irradiation with a single ultrashort laser pulse (e.g., $\Delta t = 5$ ps) into supercooled water, which can provide fundamental insights into the primary process of ice crystal nucleation of which understanding important in various scientific and industrial fields (e.g., cryobiology, food processing). These results clearly suggest that the spatiotemporal controllability of the laser ablation-induced crystallization is promising for the detailed investigation of ice crystallization mechanism.

To further clarify the laser ablation-induced crystallization dynamics, here we developed a new experimental system with an ultrashort laser (λ = 800 nm, Δ*t* = 100 fs – 10 ns) and a high-speed polarization camera (Figure 1a), which enables us to visualize the orientation structure of crystals. In the experiment, laser pulses were focused into supercooled water through an objective lens ($NA = 0.40$) from the bottom side of a glass container. The polarized images were captured from the side or bottom of the container. Notably, the high-speed polarization camera could visualize the growth and orientation of individual ice single crystals even when several crystals were formed at the same time, which is crucially challenging in conventional bright-field imaging. We foresee that our laser methods will offer new insights into the crystallization mechanism of ice.

Figure 1: Optical setup for high-speed polarization imaging of laser-ablationindued crystallization dynamics.

References: [1] H. Y. Yoshikawa et al., *Chem. Soc. Rev.* **43**, 2147-2158 (2014); [2] H. Takahashi, H. Y. Yoshikawa et al., *Appl. Phys. Exp.* **14** 045503 (2021); [3] H. Takahashi, Y. Hosokawa, and H. Y. Yoshikawa et al., *J. Phys. Chem. Lett.* **14**, 4394-4402 (2023).