

# Spatiotemporal Control of Ice Crystallization in Supercooled Water by Focused Irradiation with Laser Pulses

Hozumi Takahashi<sup>1\*</sup>, Tatsuya Kono<sup>2</sup>, Kosuke Sawada<sup>2</sup>, Satoru Kumano<sup>2</sup>, Yuka Tsuru<sup>2</sup>, Mihoko Maruyama<sup>1,3</sup>, Masashi Yoshimura<sup>4</sup>, Daisuke Takahashi<sup>5,6</sup>, Yukio Kawamura<sup>6</sup>, Matsuo Uemura<sup>6</sup>, Seiichiro Nakabayashi<sup>5</sup>, Yusuke Mori<sup>1</sup>, Yoichiro Hosokawa<sup>2</sup>, Hiroshi Y. Yoshikawa<sup>1</sup>.

\*Hozumi Takahashi: takahashi@mp.ap.eng.osaka-u.ac.jp

1 Graduate School of Engineering, Osaka University, Japan.

2 Graduate School of Science and Technology, Nara Institute of Science and Technology, Japan.

3 Graduate School of Life and Environmental Science, Kyoto Prefectural University, Japan.

4 Institute of Laser Engineering (ILE), Osaka University, Japan.

5 Graduate School of Science and Engineering, Saitama University.

6 Faculty of Agriculture, Iwate University, Japan.

Ice crystallization is a significantly important phenomenon in various scientific and industrial fields such as geology, cryobiology, and food processing. To investigate the mechanism of ice crystallization, a number of researchers have tried to observe the crystallization dynamics with controlling the ice nucleation in a spatial and temporal manner by utilizing external stimuli such as electric field [1] and ultrasound [2] so far. As a different approach, our group have demonstrated the precise spatiotemporal control of nucleation of various materials by utilizing impulses such as shockwave and cavitation bubble generation that are triggered by focused irradiation with laser pulses into liquids [3-5]. The laser-induced impulses were well-localized when the ultrashort laser pulses were used due to the multiphoton excitation, thus precise spatiotemporal control of crystallization can be realized. Herein, we applied this laser method to spatiotemporal control of ice crystallization in supercooled water system and carried out the detailed observation of ice crystallization dynamics with a resolution of micrometers and microseconds.

To trigger the ice nucleation, ultrashort laser pulse (pulse duration: 150 fs – 5 ps) obtained from a regeneratively amplified laser system were irradiated to the supercooled water system through an objective lens (10 $\times$ , NA = 0.25 or 0.4). Soon after the focused irradiation with a single laser pulse, a cavitation bubble was formed at the laser focus. It expanded and reached its maximum size (diameter:  $\sim$  400  $\mu$ m) at  $t = 24$   $\mu$ s. Then, the cavitation bubble started to shrink and finally collapsed. At the same time, long-lasting bubbles appeared and remained for least several hundreds of microseconds. Then generation of ice crystals were typically observed on these long-lasting bubbles. This optical approach can be used in any aqueous system if the container is optically transparent, thus we foresee that this method will be a practical tool for investigating the mechanism of ice crystallization under various environmental conditions (e.g., temperature, additives).

## References

- [1] Yang F et al. Ice nucleation at the contact line triggered by transient electrowetting fields. *Appl. Phys. Lett.* 2015;107:264101 (5 pages).
- [2] Chow R et al. The importance of acoustic cavitation in the sonocrystallisation of ice - high speed observations of a single acoustic bubble. *IEEE Ultrasonics Symposium*. 2003;1447-1450.
- [3] Yoshikawa HY et al. Laser ablation for protein crystal nucleation and seeding. *Chem. Soc. Rev.* 2014;43:2147-2158.
- [4] Takahashi H et al. Crystallization from glacial acetic acid melt via laser ablation. *Appl. Phys. Express* 2021;14:045503 (4 pages).
- [5] Tsuru Y, Yoshikawa HY et al., Effects of pulse duration on laser-induced crystallization of urea from 300 fs to 1200 fs: impact of cavitation bubbles on crystal nucleation. *Appl. Phys. A* 2022;128:803.