# Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF**.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

*https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do*

***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

## Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

## Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

| ESRF | **Experiment title:** Monitoring Laser-Induced Ice Nucleation | **Experiment number**: SC-4531 (Proposal: P53216) |
|---|---|---|
| **Beamline**: ID09 | **Date of experiment**: from: June 15, 2017 to: June 20, 2017 | **Date of report**: September 12, 2017 |
| **Shifts:** 15 | **Local contact(s):** Norman Kretzschmar and Matteo Levantino | *Received at ESRF:* September 12, 2017 |

**Names and affiliations of applicants** (* indicates experimentalists):

Iftach Nevo* – Dept. of Materials and Interfaces, Weizmann Institute of Science, Israel

Nir Naftali* – The Porter School of Environmental Studies, Tel-Aviv University, Israel

Leslie Leiserowitz* – Dept. of Materials and Interfaces, Weizmann Institute of Science, Israel

Jens Als-Nielsen* – Dept. of Condensed Matter Physics, University of Copenhagen, Denmark

**Report:** *Experimental setup* - included a cooling chamber, an X-ray detector, and an optical setup to guide the 532 nm ns-pulsed laser beam to impinge the water drop to induce ice nucleation. The following equipment and elements were tested successfully and used in the experiment: A home-made cooling chamber which allows (**a**) deposition of ultra-pure water drops on a hydrophobic glass slide in helium atmosphere (**b**) monitoring and illuminating the water drop with laser beams from two orthogonal directions (**c**) probing the growth of the laser-induced ice nuclei with a pulsed X-ray beam and X-ray detector. (**d**) a sleeve to fill the volume between the cooling chamber and the X-ray detctor with helium in order to increase the signal to noise ratio.

The optical setup was constructed to guide the beam to impinge the water drop either from above (vertical direction), the side (horizontal direction) or from both directions simultaneously. A further element in this setup was the ability to double the pulse duration from 5 ns to 10 ns. This was achieved by using two polarizing beam splitter cubes; first to split the pulse and then merge its split parts to be co-aligned after one part has traveled through a delay line of 3 meters that avoids a temporal overlap. In addition, to control the polarization status of the incoming linearly polarized laser beams, a quarter waveplate in each direction was installed before the corresponding entrance window. Each beam passed through a 0.5 mm apertures to reduce the angles of the refracted rays in the drop.

*Procedure* After deposition of water drop, we moved laterally the chamber with an X-Y-Z translational stage to overlap the drop with the intersection point of the laser beam/s with the X-ray beam. Once the point of incidence was set, the chamber was cooled to a certain temperature. Upon temperature stabilization (< 0.1 degree), the drop was exposed to laser pulses (at 10 Hz repetition rate) during periods of two minutes.

The probing pulsed X-ray beam was orthogonal to the laser beams, and was synchronized to probe diffraction 10 – 200 ns after the laser pulses. The overlap between the X-ray beam spot (100×60 $\mu m^2$) with the

spot of each laser beam (500 μm diameter) was verified with a fluorescent screen and a microscope equipped with a CCD camera.

<u>*Results*</u> The laser induced ice nucleation effect is shown in the series of snapshots in Fig.1a-d that illustrates the drop in different stages toward complete freezing. The first apperance of diffraction peaks after illumination with pulsed orthogonal laser beams (that were with linear and orthogonal polarizations) is evident from the X-ray detector image shown in Fig. 1e. The diffraction image of a fully frozen drop shown in Fig. 1f reveals a variety of diffraction peaks at different orientations. Finally, Fig. 1g shows the evolution of (scaled) intensity from a region of diffraction peaks (not shown) under different illumination configuration versus the time that is represented by the X-ray image number (shifted). The different transition times from a liquid state to a crystalline state for different illumination configurations is clearly evident from the figure. This transition time depends on the growing rate of the probed crystallites by the X-ray beam and the freezing dynamics of their surrounding. Thus, it hints about the nucleation mechanism.

The main problem in this type of experiment is to probe with the X-ray beam the initial location of nucleation. In case that the laser beam is directed to vertical incidence, we have evidences (CCD camera) for a preferred heterogeneous nucleation that started at the bottom interface with the hydrophobic glass (as expected). But, mistakenly the X-ray beam was directed to a higher level of 1 mm above this interface. Moreover, we had no programs to analyze such data in real time (already resolved). In principle, same problem holds for illumination from the side.

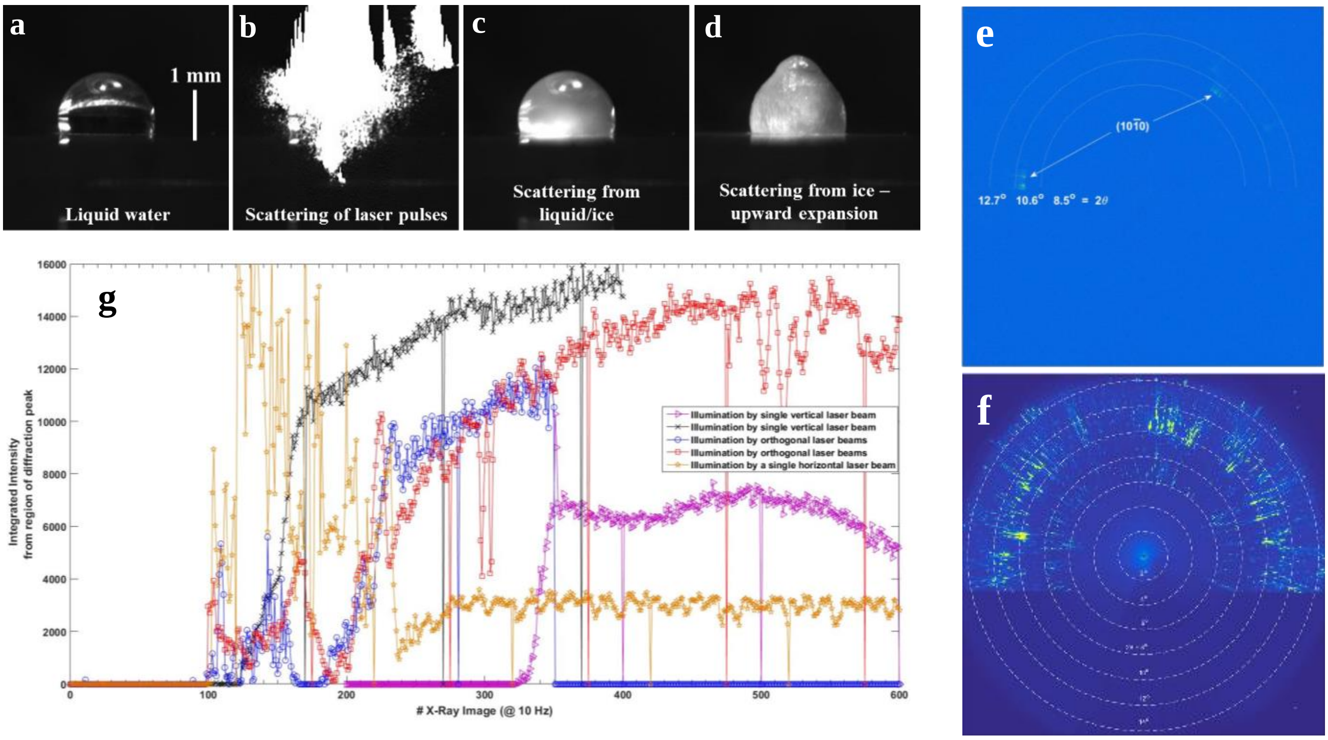


*Figure 1. (**a-d**) Snapshots illustrating the laser-induced freezing of a water drop on a hydrophobic glass slide. Freezing was achieved by illuminating the sample with two orthogonal pulsed laser beams with a time delay of 10 ns between the two. (**e**) Image from the X-ray detector based on a single X-ray pulse that probed the water drop 100 ns after a pair of laser pulses: the first appearance of diffraction peaks is clearly evident. (**f**) Integrated X-ray image of 1000 X-ray pulses from a frozen water drop under dark conditions. (**g**) The evolution of intensity in the region of different diffraction peaks from different runs; each X-ray image was acquired 15 – 100 ns after the laser pulse, and the time difference between successive images was 100 ms.*