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# THREE-DIMENSIONAL IMAGING OF CRYSTALLINE STRUCTURE IN WATER ICE AT HIGH PRESSURE BY TIME-DOMAIN BRILLOUIN SCATTERING

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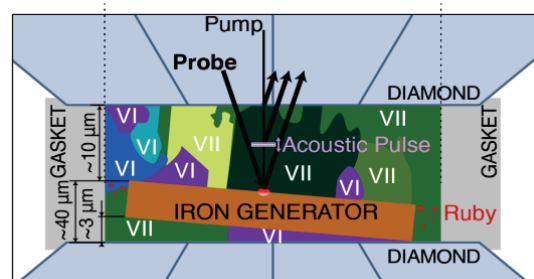
## ABSTRACT

Time-domain Brillouin scattering uses ultrashort laser pulses to generate coherent acoustic pulses of picoseconds duration in a solid sample and to follow their propagation in order to image material inhomogeneities with sub-optical depth resolution. The width of the acoustic pulse limits the spatial resolution of the technique along the direction of the pulse propagation to less than several tens of nanometres. Thus, the time-domain Brillouin scattering outperforms axial resolution of the classical frequency-domain Brillouin scattering microscopy, which uses continuous lasers and thermal phonons and which spatial resolution is controlled by light focusing. The technique benefits from the application of the coherent acoustic phonons, and its application has exciting perspectives for the nanoscale imaging in biomedical and material sciences. In this study, we report on the application of the time-domain Brillouin scattering to the 3D imaging of a polycrystal of water ice containing two high-pressure phases. The imaging, accomplished via a simultaneous detection of quasi-longitudinal and quasi-shear waves, provided the opportunity to identify the phase for individual grains and evaluate their crystallographic orientation. Monitoring the propagation of the acoustic waves in two neighbouring grains simultaneously provided an additional mean for the localisation of the grain boundaries.

## 1. INTRODUCTION

Time-domain Brillouin scattering (TDBS) is a non-destructive opto-acousto-optic pump-probe technique [1] which allows study of a variety of transparent materials [2]. We report here extension of the TDBS technique to 3D imaging of elastic inhomogeneities in water ice compressed in a diamond anvil cell (DAC) to 2.15 GPa. To make the 3D imaging in a reasonable time, we applied the technique based on an asynchronous optical sampling (ASOPS). As shown in Fig. 1, an iron plate of about 40  $\mu\text{m}$ -thick and 100  $\mu\text{m}$  in diameter was used as an optoacoustic generator. Pump laser pulse (515 nm wavelength, 150 fs duration) was absorbed by the generator which emitted picosecond acoustic pulses into the water ice sample [3]. Propagation of the acoustic pulses

in ice was monitored using a probe laser pulse (532 nm wavelength, 150 fs duration) delayed with respect to the probe pulses. In 2 hours, it is possible to obtain 3D images of the Brillouin frequency in  $40 \times 40 \times 10 \mu\text{m}^3$  volume with a lateral resolution of 2  $\mu\text{m}$ . We report here on a longer scan with more averages and a  $100 \times 100 \mu\text{m}^2$  scanned area with 1.25  $\mu\text{m}$  step to map the entire sample. 3D maps were obtained both with longitudinal and shear acoustic pulses. We have revealed that domains of the highest and the lowest Brillouin frequencies correspond to ice VII and ice VI, respectively, while the intermediate frequencies could be due to the presence of both ice VII and ice VI, which coexist at this pressure.

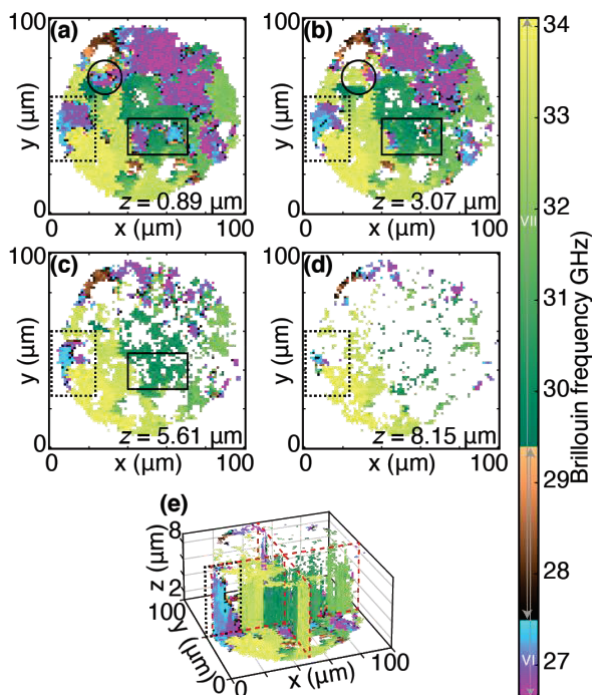


**Figure 1.** Vertically expanded cross section of the  $\text{H}_2\text{O}$  sample in a DAC, optical and acoustical paths. The disk-shape iron opto-acoustic generator has the diameter of about 110  $\mu\text{m}$ . It touches the lower diamond anvil at its right end. Even though the pump and probe laser paths are collinear in the experiment, the probe one is shown inclined for a better visualisation of its reflections at different depths.

## 2. EXPERIMENTAL RESULTS

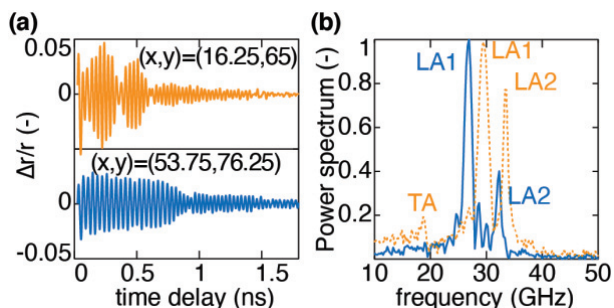
In Fig. 2, the dominant frequency related to the quasi-longitudinal acoustic (LA) mode are estimated at different depths by calculating the FFT of the acoustic signal sliced with a Hann window of 0.23 ns (about 7 oscillations of the LA mode), which gives a depth resolution of about 1.2  $\mu\text{m}$ . Using the temporal indication of the centre of the sliding window and the measured local velocities at the previous instants for the corresponding acoustic mode, the time axis can be transformed in the depth axis; the smaller the time, the closer the probed zone to the iron optoacoustic transducer. Note that this change of coordinates implies knowledge of the local refractive index. For the sake of simplicity, we have chosen here to attribute only one

refractive index to a given voxel, even though it contains two phases. The volume zones pointed out by circle and rectangles are of interest because of the presence of multiple grains.



**Figure 2.** 3D TDBS imaging of the polycrystalline water ice sample. (a)-(d) The slices are shown at particular positions along the  $z$ -axis shown in the right-bottom corner of each slice. (e) 3D representation of the complete probed volume with the first (a) and last (not shown) slices ( $xy$  planes), and the middle slices ( $yz$  and  $xz$  planes) at  $x = 50 \mu\text{m}$  and  $y = 50 \mu\text{m}$ , respectively (red dashed rectangles).

We also report the TDBS monitoring of two quasi-longitudinal coherent acoustic pulses propagating in parallel and in the same direction in two differently oriented ice grains along their mutual interface (orange lines in Fig. 3). All TDBS signals as the one shown by orange colour were detected in the vicinity of grain boundaries and never in grain volumes. Accordingly, detection of such signals could be fruitful for locating grain boundaries.



**Figure 3.** Experimental acoustic contributions to transient reflectivity signals as a function of time delay (a) and their Fourier spectrum density (b), showing two LA modes propagating in two different grains either “in parallel” (orange lines) or “in sequence” (blue lines).

### 3. CONCLUSIONS AND PERSPECTIVES

We reported on advances in applications of the TDBS technique in high-pressure experiments in a DAC that followed the recent achievements of the 3D TDBS imaging at ambient conditions. The first high-pressure 3D TDBS imaging and the first observation in a DAC of the TDBS with quasi-shear coherent acoustic pulses were reported. We demonstrated some examples of the fruitful application of the TDBS imaging with several acoustic modes simultaneously. We also revealed the possibility to localise positions of grain boundaries in polycrystal by the identification of the specific TDBS signals that are due to the simultaneous propagation of an acoustic pulse in two adjacent grains. Overall our reported results are a big step towards the perspective of the full 3D characterisation of sample texture at extreme conditions (high pressures and/or temperatures) and its evolution on further compression or temperature change. Such characterisation is of special interest for different branches of research at extreme conditions. It will make possible an examination of the texture of minerals present in the deep Earth and its evolution upon nonhydrostatic compression with the detailedness presently not accessible by other techniques. Such information will permit a conclusion about the nature of seismic anisotropies observed in the Earth’s mantle.

### 4. ACKNOWLEDGEMENTS

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