

# Three-dimensional imaging of inhomogeneities in transparent solids compressed in a DAC by time-domain Brillouin scattering

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The time-domain Brillouin scattering (TDBS) is a non-destructive opto-acousto-optic pump-probe technique [1] which allows the study of a variety of transparent materials [2]. In this technique, a femtosecond pump laser pulse, absorbed by an optoacoustic transducer, emits a picosecond acoustic pulse into the sample. The width of this pulse is in nanometric spatial scale. The acoustic pulse reflects a time-delayed probe laser pulse generated by the same or another femtosecond laser. The detected transient reflectivity signal is a result of the interference at the photodetector of probe light reflected by stationary surfaces/interfaces of the sample and of probe light reflected by acoustic pulse. It contains the information on the properties of the material in a time-dependent spatially localized position of the acoustic pulse. Therefore, the technique allows imaging of materials along the acoustic pulse propagation path with a spatial resolution better than optical one. Two dimensional TDBS imaging has been earlier applied for revealing the texture of solid H<sub>2</sub>O [3] and Ar [4], the phase transitions [5] and the pressure dependences of single crystal elastic moduli  $C_{ij}(P)$  in water ice up to 82 GPa [6].

We report here the extension of the TDBS technique to the 3D imaging of transparent materials compressed in a DAC. To accelerate the data acquisition and thus to make 3D imaging possible in a reasonable time, we have applied an ultrafast laser technique based on an asynchronous optical sampling (ASOPS). In the ASOPS technique, the time delay between the pump and the probe pulses is controlled electronically by an offset of the repetition rate frequency of two lasers without the use of a mechanical optical delay line. The TDBS experiments described here have been performed using an ASOPS-based picosecond acoustic microscope (JAX-M1, NETA, France).

First, TDBS experiments have been realised on water ice compressed in a DAC to 2.1 GPa. An iron plate of ~40  $\mu\text{m}$  thickness and ~110  $\mu\text{m}$  in diameter has been used as an optoacoustic generator. The diameter of the pump and probe laser beams at the surface of the generator was ~1.4  $\mu\text{m}$ . Irradiated by pump laser pulse at 515 nm wavelength, the optoacoustic generator emits picosecond acoustic pulses into the 14.5  $\mu\text{m}$  layer of water ice [5]. The obtained 3D distribution of the Brillouin frequencies in the sample volume with dimensions of 40×40×10  $\mu\text{m}^3$  is represented in Figure 1 in form of its 3 cross sectional slices. The slices

correspond to the average delay times of 0.11 ns, 0.36 ns and 0.74 ns, corresponding to the average depths around

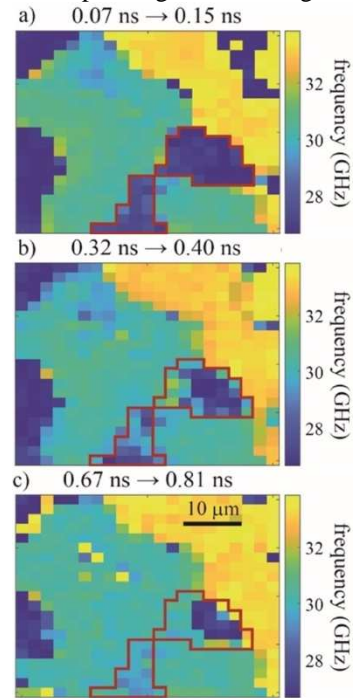


Figure 1. 2D maps of the Brillouin frequencies distribution in the 40×40  $\mu\text{m}^2$  cross sections of the tested ice volume at 2.1 GPa for three shifting time slices. The red line highlights one of the lateral regions with important variations of the Brillouin frequency as a function of the axial distance from the generator, which increases with time delay.

0.6  $\mu\text{m}$ , 2  $\mu\text{m}$ , and 4  $\mu\text{m}$ , respectively. The time durations of the presented cross sectional slices are 0.08 ns, 0.08 ns and 0.14 ns, corresponding to spatial thicknesses of approximately 400 nm, 400 nm, and 750 nm, respectively. The lateral resolution within each cross-section was 2  $\mu\text{m}$ . The total time, needed for the collection of this 3D distribution, was of about 2 hours. The maps of the Brillouin frequencies have been obtained using a time-frequency analysis based on the synchronous detection principle [6]. It is worth noting that while each cross sectional time slice has a constant duration for any lateral coordinate, its spatial thickness and the average axial depth are different in different lateral points because acoustic waves propagate with different velocities in different lateral points. The estimations demonstrate that the highest and the lowest detected Brillouin frequencies correspond to the ice VII,

while the intermediate frequencies in the lower half of the frequency spectrum could be due to the presence of ice VII and ice VI, which could coexist at 2.1 GPa at room temperatures. This is due to the ranges of the sound velocity variations related to the elastic anisotropy in ice VI (5100 m/s – 5500 m/s) and in ice VII (4900 m/s – 5900 m/s) and, secondly, because of an overlap of the related Brillouin frequency ranges.

Second, the ASOPS technique applied to 3D imaging of compressed 2-(hydroxyethyl)methacrylate (HEMA) could significantly extend our knowledge about the earlier recognised two different states of this amorphous material above and below 6.5 GPa [7], which cannot be distinguished by any other technique. Moreover, it should allow following in time the 3D spatial spreading of the polymerization reaction starting from the nucleation site [8]. Since polymerized HEMA is also amorphous, the in-situ information on its density changes on compression is currently absent.

In this work we have demonstrated that ASOPS-based 3D TDDBS imaging of the transparent materials in a DAC should provide information, which cannot be obtained by other methods applied earlier. This 3D imaging allows us to visualize shapes of crystallites formed in sample volume and their transformation with increasing load. In perspective, the 3D imaging of the

transient processes at high pressures with nanometers depth resolution could be possible.

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