3D imaging of water ice under high-pressure non-hydrostatic load by time-domain Brillouin scattering

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Background – Time-domain Brillouin scattering (TDBS) technique is an opto-acousto-optic pump-probe technique [1] that uses ultrashort laser pulses to generate coherent acoustic pulses (CAPs) of picoseconds duration in a solid sample and follow their propagation in order to image inhomogeneities of acoustic, optical and/or photo-elastic parameters of materials transparent to the probe light wavelength [2]. This technique presents an axial resolution deeply sub-optical (to nm-scale), controlled by the CAPs width, and a lateral one down to the optical diffraction limit, controlled by lateral focusing of used laser beams. Detection of propagating CAPs is possible because they scatter time-delayed probe laser pulses heterodyned by the probe-pulse reflections from stationary sample surfaces, giving rise to an oscillating component in a transient reflectivity signal: the so-called Brillouin oscillation (BO).

The instantaneous frequency of the BOs is proportional to the product of local optical refractive index (n) and sound velocity along the beam path (v). Hence, in polycrystalline transparent samples made of anisotropic materials where n and v change from grain to grain, the Brillouin frequency changes with the probe delay give access to the depth arrangement of grains. Two-dimensional lateral scanning of the sample hence allows the TDBS technique to provide 3D-imaging of sample texture. In anisotropic grains, up to three bulk acoustical modes, one longitudinal and two shear, might be monitored. The time-frequency analyzes of frequency contents of the TDBS signals provide then access to shapes, relative coordinates of all grains (if resolved), as well as crystallographic orientation of the identified grains with respect to a common coordinate system [3].

The non-contact feature of the TDBS technique further permits to examine samples located in devices used to reproduce extreme conditions such as diamond anvil cells (DACs) for ultrahigh pressure. We report here on the 3D imaging of water ice under high-pressure non-hydrostatic load by TDBS. The 3D characterization of individual grains of two coexisting high-pressure water ice phases is reported, as well as imaging of a monocrystal fracture induced by non-hydrostatic compression allowing to follow the polycristallization process occurring upon load increase in a DAC.

Methods – The TDBS experimental set-up (Fig. 1(a)) was based on asynchronous optical sampling, where accumulating time delay between green pump (515 nm, 198 fs duration) and green probe (535 nm, 130 fs duration) laser pulses is due to difference in the repetition rates of these lasers, allowing fast data acquisition. It was hence possible to obtain 3D images of the Brillouin frequency distribution (of any detected acoustical modes) in a 100×100×10 μm^3 volume with a lateral resolution of 2.5 μm limited by the overlap of pump and probe laser beams. The data collection rate depended on the chosen pump and probe laser powers and the number of averages needed to reach acceptable signal-to-noise ratio.

As depicted in Fig. 1(b), the generation of CAPs propagating in the transparent water ice compressed in the DAC occurred thanks to an absorbing metal layer serving as an optoacoustic transducer (OAT). Pressure inside the DAC was measured using fluorescence spectra of (embedded) ruby grains which R1-line position is calibrated vs. pressure. The optical path for pressure monitoring was included in the set-up (Fig. 1(a)) to allow measurement without removing the DAC from the sample stage, hence facilitating the comparison of 3D images at different pressures and visualizing evolution of crystallite shapes with compression.

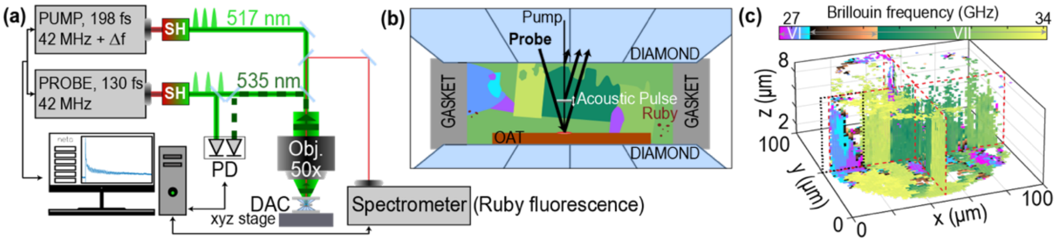

Fig. 1. (a) Experimental set-up and (b) TDBS experiment scheme, where the pump absorbed in OAT launched a CAP in the polycrystalline water ice where it scatters the probe. (c) 3D visualization of polycrystalline water ice microstructure.

Results summary and conclusive remarks – The used TDBS set-up provided a comprehensive, reliable high-resolution *in-situ* 3D visualization of microstructure of a transparent polycrystalline sample of water ice compressed in a DAC to 2.15 GPa where two phases, ice VI and ice VII, coexist (Fig. 1(c)). We observed, for the first time at high pressures in a DAC, TDBS signals containing contribution of quasi-shear CAPs, fruitfully used for grains characterization. Grain boundaries were also localized by identifying specific TDBS signals, caused by CAPs simultaneously propagating in two adjacent grains. Last but not least, the monocrystal fracture induced by non-hydrostatic loading was followed in 3D, further extending the horizons of investigation of solids and their evolution at (changing) extreme conditions.

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