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Time-domain Brillouin scattering for the evaluation of materials interface inclination: Application to imaging of crystal degradation upon non-hydrostatic compression

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Laser ultrasonics is a very handy technique to characterize, evaluate and image materials remotely under extreme (harmful) conditions, such as high pressures. Time-domain Brillouin scattering (TDBS) is such a technique that uses ultrafast laser pulses to generate and detect coherent acoustic pulses (CAPs) as they propagate through a transparent material for the probe laser wavelength [1-2]. In TDBS, the probe light scattered by the propagating CAPs interferes with the probe light reflected on a stationary interface, which leads to a signal with Brillouin oscillations (BOs). If a CAP is reflected from and transmitted through a material interface, the TDBS signal can also feature an echo. Both BOs and echo features can be used for profilometry of (buried) interfaces [3-5] by using information from measurements performed at different lateral positions while scanning co-focused pump and probe beams. A recently developed theory of this type of non-collinear sound-light interaction [6] also provides the opportunity of interface profilometry without the drawback of lateral averaging, i.e., from a local estimation of the angle using a single measurement. The TDBS technique was here applied to monitor the degradation of a single crystal made of lithium niobate (LiNbO_3 – LN) upon non-hydrostatic compression in a diamond anvil cell (DAC). The plate-like sample made of a single crystal of LN was prepared from a nominally 20 μm -thick single-crystal disk (x-cut). The nominal geometrical characteristics of the surfaces of the disk provided by the supplier allowed us to estimate the maximum surface inclination to be 0.6° at sub- μm scale and averaged down to 0.02° at μm -scale. One side of this crystal was then coated with a 70 nm-thick Ti layer which served mostly as the optoacoustic generator in our TDBS measurements. The crystal was then loaded in a DAC with water as pressure-transmitting medium and compressed up to 10.4 GPa, where the liquid water transforms to the phase VII of water ice. The 3D TDBS imaging experiments conducted in the DAC reveal modifications of the thickness profile of the crystal with initially plane-parallel surfaces, as well as of one of its face profiles, caused by the non-hydrostatic compression. The map of the echo arrival times reveals at least two orders increase in the inclination angles. Using our developed analytical theory [6], the inclination angle of the Ti-coated surface of the LN relative to the diamond anvil surface has been estimated by fitting the experimental signals. The evaluation performed from each point of measurement allows to locally assess the inclination, which can be revealed either through the modifications of the BOs in the absence of the detectable CAPs, or through the detection of the CAP echoes arrivals at the interface in the absence of BOs, or simultaneously through both, which further complement the usual use of BOs amplitude and frequency variation. The zoology of TDBS signals gathered in this sample is very rich and will necessitate further theoretical investigations to fully explain all the waveforms by taking into account the optical and elastic anisotropy of LN and water ice, for example. Performing TDBS experiments by analyzing probe polarization would also complement our current analysis.

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