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To cite this version:

Nicolas Pajusco, Samuel Raetz, Nikolay Chigarev, Sathyan Sandeep, Mathieu E Edely, et al.. 4D imaging of water ice under high-pressure nonhydrostatic load by time-domain Brillouin scattering. The 22nd International Conference on Photoacoustic and Photothermal Phenomena (ICPPP22), Carlos Serpa, Aug 2024, Coimbra, Portugal. pp.206-208. hal-04750941

HAL Id: hal-04750941 <https://hal.science/hal-04750941v1>

Submitted on 24 Oct 2024

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4D imaging of water ice under high-pressure nonhydrostatic load by time-domain Brillouin scattering

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Time-domain Brillouin scattering (TDBS) is a technique that involves generating a coherent acoustic pulse (CAP) at GHz frequencies using a picosecond/femtosecond pump laser pulse. This method operates by detecting the interaction between a time-delayed probe laser pulse and the CAP [1-2]. The resulting signal comprises Brillouin oscillations, whose time-varying frequency is directly related to the local optical and elastic properties of a transparent material. In an anisotropic elastic solid, three types of bulk waves can propagate: one quasi-longitudinal and two quasi-shear. Depending on the polarization of each wave, given by the propagation direction in the crystalline axis, and the probe laser polarization up to three distinct Brillouin frequencies can be detected. The detected signal is in terms of time-delay between the CAP generation and the probe laser allowing to follow the CAP. The instantaneous frequency allows to extract the product of the refractive index at the probe wavelength and the acoustic wave velocity at a given time delay, representing the properties at the CAP position. By knowing the refractive index, it is possible to extract the acoustic velocity as a function of depth allowing depth profiling along z direction. Then by doing a 2D lateral scan in the x-y plane, it is possible to reconstruct the 3D map of bulk velocity. This method has been demonstrated for imaging coexisting grains of water ice VI and VII [3]. Varying the non-hydrostatic pressures and repeating the 3D extraction leads to 4D imaging, i.e., space $(3D)$ + pressure $(1D)$, which allows to follow the pressure-induced movement of interfaces.

To control the non-hydrostatic pressure, a membrane diamond anvil cell (DAC) is employed. It is based on two diamond anvils with working surface of 0.3 mm diameter, which allow to apply pressure up to 150 GPa. A 20 nm-thick Au layer is deposited on the bottom diamond acting as an opto-acoustic transducer (OAT). The water ice is sandwiched between the OAT and the top diamond. Figures 1a-b show the optical images of water ice VII at 2.3 and 4.5 GPa, respectively. The experimental setup employs asynchronous optical sampling (ASOPS) with a 535 nm probe laser, focused through the top diamond at the OAT/water ice VII interface to a beam diameter at $1/e^2$ in intensity of 2.6 μ m by a 50x, 0.45 NA microscope objective. The pump laser has 515 nm wavelength and is focused from the opposite side at the bottom diamond/OAT interface to a beam diameter at $1/e^2$ in intensity of 5.7 μ m by a 20x, 0.4 NA microscope objective. At each pressure step, a 2D scan is performed with 93x93 points and a 1.5 μ m lateral step leading to 4D imaging. The thermal background is filtered out using a third-

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order bandpass Butterworth filter from 5 to 80 GHz. Subsequently, the frequency of the spectral maximum quasi quasi-Summer Supering Superingtherm

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Shear CAP. bandpass Butterworth filter from 5 to 80 GHz. Subsequently, num is extracted in the range of $25 - 50$ GHz for quasi-longitudishear CAP. -50 GHz for quasi-longitudinal CAP and $5 - 25$ GHz for

Fig. 1. Experimental data on water ice VII in a DAC at 2.3 GPa (first row) and 4.5 GPa (second row). (a-b) Optical image amplitude in the range of: (c-d) [25-50] GHz, corresponding to the quasi-longitudinal CAP, and (e-f) [5-25] GHz corresponding to a quasi-shear CAP. In (c-e), colormaps are depicted on top of the optical image shown in (a). imental data on water ice VII in a DAC at 2.3 GPa (first row) and
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obtical images. (c-e) Colormaps depicting the spatial distribution of the frequency with the maximum

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Figures 1c-d show the Brillouin frequency map of quasi-longitudinal CAP on top of the optical image at 2.3 and 4.5 GPa, respectively. At 2.3 GPa, there are five distinct Brillouin frequencies in the range of 25-Figures 1c-d show the Brillouin frequency map of quasi-longitudinal CAP on top of the optical image at
2.3 and 4.5 GPa, respectively. At 2.3 GPa, there are five distinct Brillouin frequencies in the range of 25-
50 GHz, in 50 GHz, indicating five different crystal orientations of water ice VII. Quasi-shear Brillouin frequencies
are also visible in four regions and absent in one, as shown in Fig.1e. At 4.5 GPa, the number of Figures 1c-d show the Brillouin frequency map of quasi-longitudinal CAP on top of the optical image at 2.3 and 4.5 GPa, respectively. At 2.3 GPa, there are five distinct Brillouin frequencies in the range of 25-50 GHz, ind shown in Fig function of time, leading to depth imaging. shown in Figs. 1d and 1f. Through time-frequency analysis, the Brillouin frequencies are extracted as a
function of time, leading to depth imaging.
Water ice VII, having cubic crystalline structure is optically isotropic, Brillouin frequency regions increases to 7, depicting grain' separation and shown in Figs. 1d and 1f. Through time-frequency analysis, the Brillouin f function of time, leading to depth imaging.
Water ice VII, having cubic 2.3 and 4.5 GPa, respectively. At 2.3 GPa, there are five 50 GHz, indicating five different crystal orientations of are also visible in four regions and absent in one, as Brillouin frequency regions increases to 7, depicti is. (c-e) Colormaps depicting the spatial distribution of the ge of: (c-d) [25-50] GHz, corresponding to the quasi-longitudial dasi-shear CAP. In (c-e), colormaps are depicted on top of the the Brillouin frequency map of q Through time-frequency analysis, the Brillouin frequencies are extracted as a ion GHz, corresponding to the quasi
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