4D quantitative opto-acousto-optical imaging of transient material transformations

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Abstract: Picosecond acoustics is an experimental opto-acousto-optical technique based on ultrafast lasers, with high repetition rates, applied for the generation and detection of coherent acoustic pulses of nanometric spatial size and temporal size of the order of a few picoseconds. In optically transparent materials, these acoustic pulses can be detected along their propagation path inside a sample. The measured time signal thus contains, at a given time, information on the properties of the material at the position the acoustic pulse occupied in the thickness at the time considered. This technique is commonly called picosecond acoustic interferometry (PAI) [1] or time-domain Brillouin scattering (TDBS) [2]. Continuous measurement of coherent nano-acoustic pulse propagation (Fig. 1) provides an image of the inhomogeneity of the sample traversed by the acoustic pulse with nanometer axial resolution [3]. The lateral resolution is controlled by the focusing of the light. Recently, following our early work on thickness characterization [3,4] and the pioneering application of laser ultrasound to high-pressure experiments in a diamond anvil cell (DAC) by French researchers [5,6], the TDBS technique has been applied in our team to the three-dimensional imaging of textured polycrystalline materials with sub-micrometer axial resolution and micrometer lateral resolution [7,8] (Fig. 1). Very recently and for the first time, we have used acoustic shear waves at GHz frequencies for quantitative imaging of polycrystals at high pressures in a diamond anvil cell, waves that have allowed an undeniable gain in contrast and provided additional information for a more complete characterization [8].

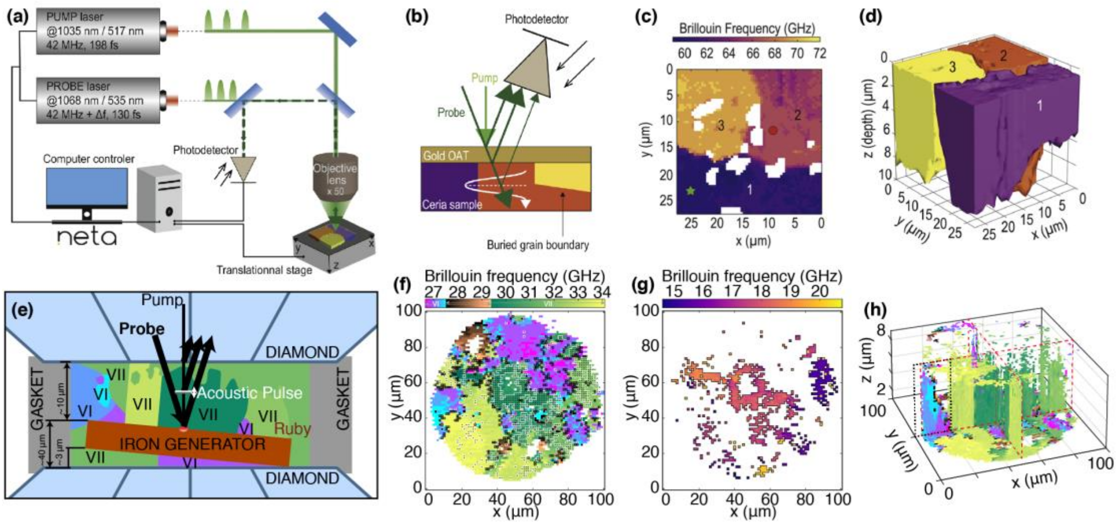

Figure 1. (a) Schematic of the experimental TDBS imaging system (JAX-M1). (b) Schematic of the probe beam reflections in the TDBS technique (reflection configuration): two stationary reflections occur at the interfaces of the optoacoustic transducer (OAT), while a third, weaker reflection occurs at different depths along the propagation path of the acoustic pulse. A tilted boundary between two grains is shown in the right-hand side of the polycrystal diagram [7]. Imaging results in ceria (c-d) and water ice (e-h) polycrystals: (c) 2D image of the free surface of the sample and (d) 3D reconstruction of 3 grains (1/purple, 2/orange and 3/yellow) with the longitudinal mode [7]. (e) Enlarged side view in cross section of the diamond anvil cell. The results (f-h) of the TDBS experiments [8] revealed the coexistence of two phases in the sample, ice VI (bluish) and ice VII (greenish). The disc-shaped iron opto-acoustic generator in the experimental chamber has a diameter of about 100 μm. It touches the lower diamond anvil at its right end. Although the trajectories of the pump and probe lasers are collinear in the experiment, the probe laser is shown tilted for a better visualization of its different reflections [8]. (f-g) Dominant frequency content attributed to the (f) longitudinal and (g) transverse modes of the first two nanoseconds of TDBS signals observed in the 100 x 100 μm^2 area of the ice sample. (h) 3D image representation of the entire probed volume [8].

The objective of the doctoral research project we propose here is to realize, using the TDBS technique, a very fast and four-dimensional imaging method: three-dimensional measurements in space and resolved in time (3+1 = 4), in order to quantitatively evaluate by this method (transient) processes in elastic media. The realization of this objective requires the use of asynchronous optical sampling (Fig. 1 (a)) [7-9] to generate and detect coherent picosecond acoustic pulses by lasers, in measurement volumes smaller than 10 μm^3. Asynchronous sampling allows to increase the acquisition rate by 10^6 times compared to a more classical configuration using a mechanical delay line to produce the delay between the arrival of the pump pulse and the probe pulse on the sample [1-5].

The fundamental phenomena of interest for such an imaging method are currently phase transitions, chemical reactions, grain boundary movement, polycrystallization, texture and flow of materials, and more generally any spatially inhomogeneous transient phenomenon induced, in volumes smaller than 10 μm^3, by external actions such as pressure or temperature increase, or the action of a UV laser or other types of radiation.

To achieve a quantitative 4D image with a micrometric to nanometric resolution, the application of efficient and fast signal processing methods appears to be essential, even more so than for 3D imaging, due to the huge volumes of data to be processed, obtained moreover on materials whose parameters are *a priori* poorly known or unknown, in particular concerning their variations with temperature, pressure or their exposure to UV radiation, for example. Recent developments in the field of artificial intelligence and neural networks, in particular by introducing a cost function integrating the physics of the inverse problems to be solved (neural network named PINN for physics-informed neural networks), seem very promising to obtain a quantitative 4D image of the evolution of the material parameters as a function of space (3D) and time, and this for two reasons: (i) the mathematical description of the physical phenomena involved in the generation and detection of acoustic waves in the TDBS technique, although highly multi-physical (behavior and interactions of charge carriers, electromagnetic, thermal and acoustic fields, in elastic, dielectric, semiconductor, ..., transparent or absorbing materials, ...), is well known [2, 10]; (ii) first results using PINN for imaging have demonstrated a strong potential of these approaches in terms of robustness to noise and good reconstruction under low sampling constraints [11-13], making these approaches attractive for imaging fast transient processes for which a strong averaging over a fine mapping would be limiting.

The main scientific tasks of this PhD research will be to perform spatio-temporal imaging and quantitative characterization, through the development and use of dedicated PINN arrays, for the following three transient processes: a) chemical reactions induced in materials such as epoxy and low dielectric nanoporous films by thermal curing and/or UV radiation; b) phase transitions induced in polymers by pressure increase and/or UV radiation exposure; c) relaxation of epitaxially grown thin films on substrates by pulsed laser deposition.

The knowledge of material structure and elasticity evolution under external actions (high pressures, high temperatures, high radiation doses) is of extreme importance for several branches of natural sciences, such as condensed matter physics or materials science and engineering. Our research will advance innovative experimental approaches for non-destructive evaluation and signal processing methods that could be useful for the application of the TDBS technique in other research areas dealing with nanometer-resolution imaging inside microscopic objects such as plant or animal cells [14] and nanostructures or microelectronic materials [2, 3, 15-17].

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