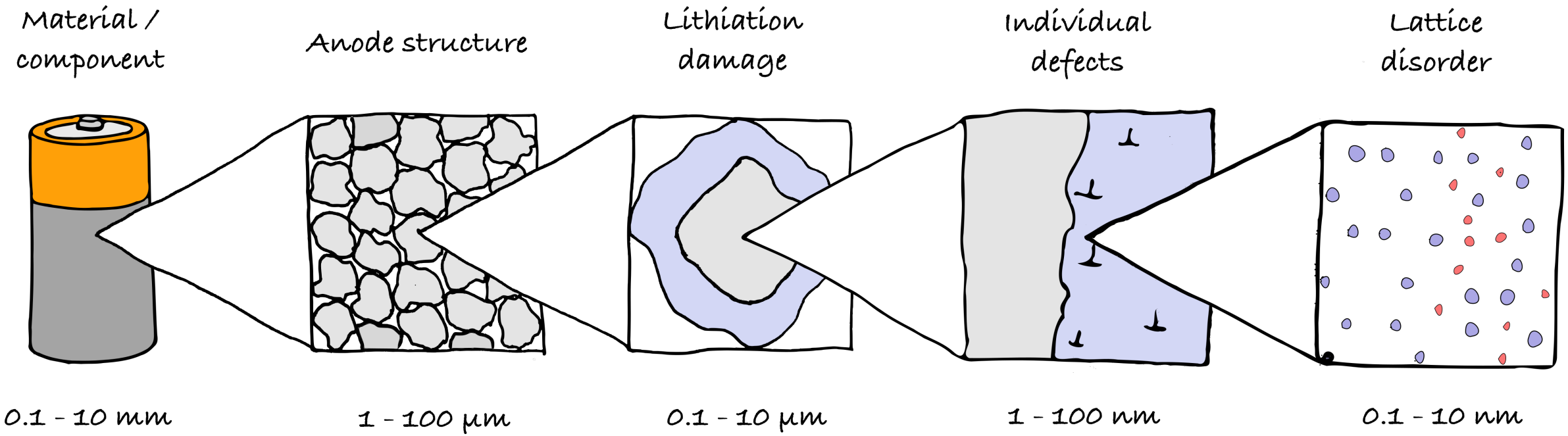
Hugh Simons, Technical University of Denmark

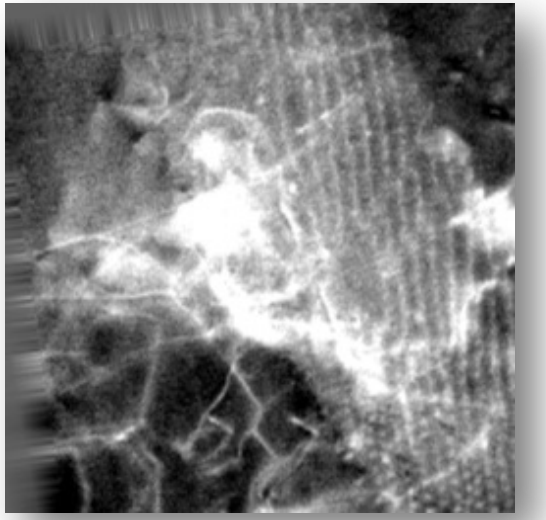
Dark-field x-ray microscopy

The challenge: Multi-scale characterisation

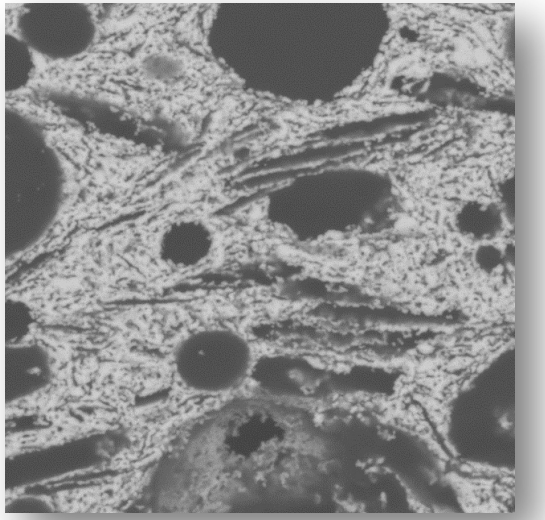


The challenge: Multi-scale characterisation

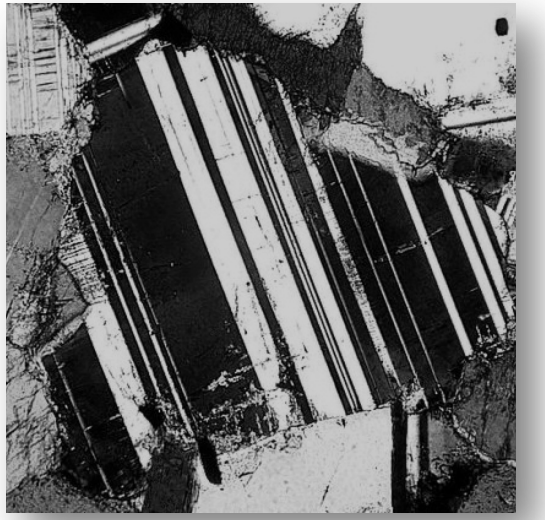
Metals



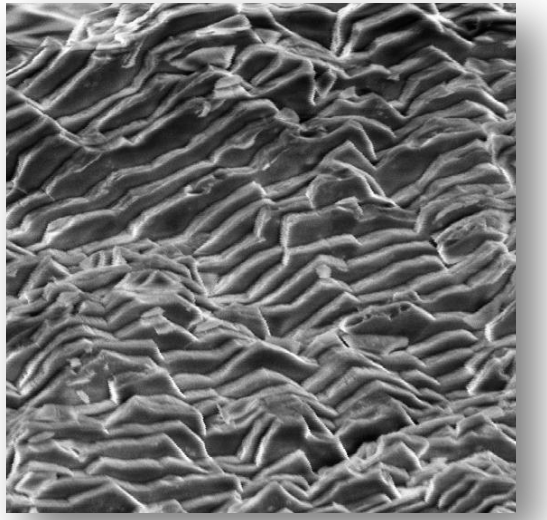
Ceramics



Minerals



Biomaterials



Dark-field x-ray microscopy:

Real space image from a finite volume in reciprocal space

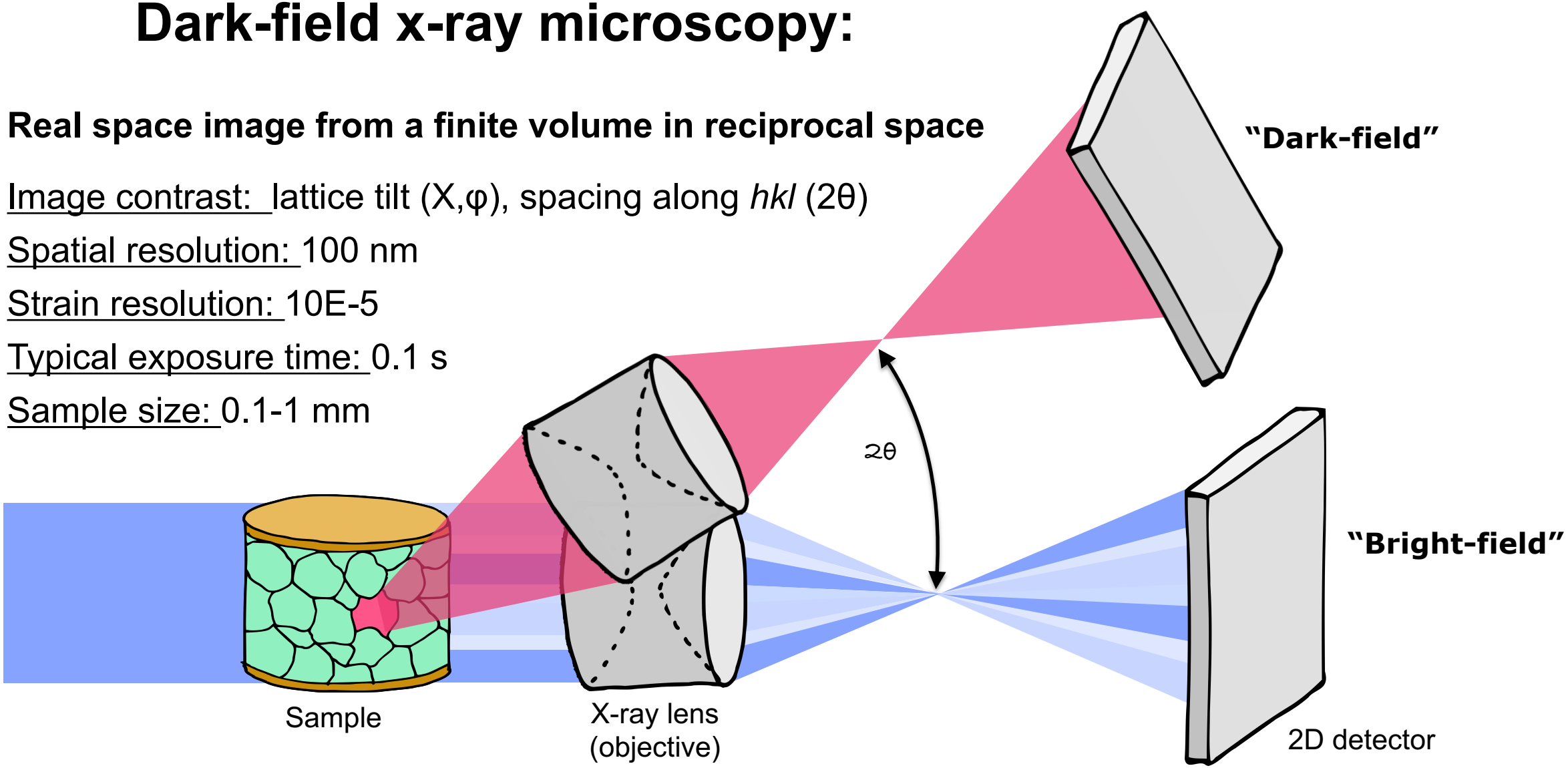
Image contrast: lattice tilt (X,φ), spacing along *hkl* (2θ)

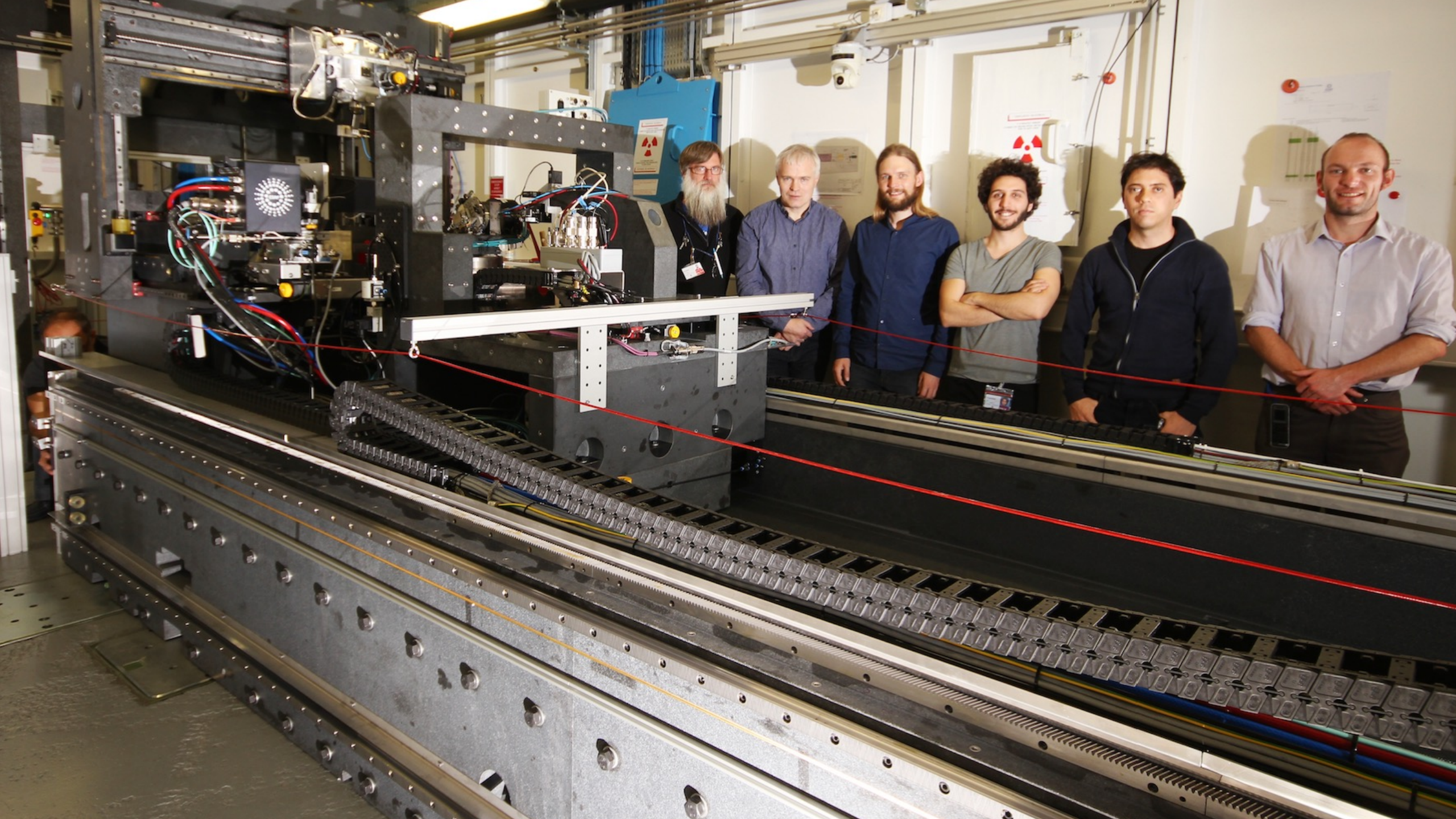
Spatial resolution: 100 nm

Strain resolution: 10E-5

Typical exposure time: 0.1 s

Sample size: 0.1-1 mm





ID06(→03): “Flagship” beamline for ESRF-upgrade

Focus on: Extremely Brilliant Source

Tilting the scales

EBSL2 will allow users to track structural dynamics across length scales in real time.

Most hard materials – from rocks to metals, to semiconductors and even bone – have complex internal structures, often comprising several layers of substructures interwoven across multiple length scales. What's more, these hierarchical structures define many of the properties of the material. Bone is a classic example: its unexpectedly high toughness stems from the way "hydroxyapatite" nanoparticles self-assemble into highly oriented fibres up to the macroscopic scale. Understanding the material means understanding the dynamics of how the individual sub-structures interact – but this requires direct access to every length scale, within the same sample and at the same time.

This is the idea behind the hard X-ray microscope: a single instrument offering comprehensive full-field and time-resolved imaging in two, three and four dimensions of the phases, grains, domains, stress fields and defects spanning length scales from 1 mm to 10 nm.

What makes hard X-ray microscopy unique is the broad range of problems it can tackle. Using high energy X-rays we can probe sub-structures within large samples or even full devices. By combining complementary imaging techniques with the versatility of X-ray optics, we can seamlessly switch between fast overviews of microscopic features and detailed images of individual defects. In this sense, the hard X-ray microscope is analogous to a transmission electron microscope, albeit one that leverages the penetrating power of X-rays to see orders of magnitude further into the samples.

Hard X-ray microscopy was developed jointly between the Technical University of Denmark (DTU) and the ESRF as part of an Advanced Grant from the European Research Council. Six years on, at the end of 2016, this collaboration resulted in a prototype instrument at ID06. First results have provided new insights into some longstanding and fundamental problems in metallurgy, energy and biological materials. In one case, we visualised the multi-scale structure of dislocation networks in diamond, an important industrial material and, more recently, an ideal sandbox for quantum-information technologies. Crucially, its photonic properties are largely at the mercy of these defects – their intermingling networks, and the long-range strains they create.

“We will capture real-time movies of structural dynamics.”

At the coarse scale, we used diffraction X-ray topography to map the location of every dislocation in the crystal. This allowed us to find a specific set of dislocation lines of interest, in this case a group of three containing kinks, including one with a clear stacking fault. Next, simply by inserting an X-ray objective lens into the beam of Bragg-diffracted X-rays, we created a magnified, full-field image of the individual dislocations within the crystal – a technique known as dark field microscopy. Coordinated movements of the sample and optics then enabled us to quantitatively map the local crystal strain and symmetry with a resolution of 100 nm. These maps showed how dislocations and stacking faults self-organise inside the material to create buried networks characterised by long-range strain fields and lattice distortions (see figure).

Now, however, this result is set to be just the tip of the iceberg. One of the ESRF–EBS upgrade beamlines, EBSL2, located on the ID03 port, will improve spatial resolution 10-fold, while making experiments hundreds or even thousand of times faster. This means we are no longer limited to seeing static structures, but can capture real-time movies of structural dynamics as they happen across multiple length scales. In the context of diamond, it could mean tracking the 3D motions of dislocations as they form networks under stress. More broadly, it allows us to pursue much more aggressive dynamics problems, such as nucleation and material failure.

The ability to extend 3D imaging into the temporal regime is a game-changer for materials science, providing the opportunity to truly guide and validate multi-scale materials models. Such models are essential to emerging approaches to the computational design of materials.

Henning Friis Poulsen and Hugh Simons, Technical University of Denmark, and Carsten Detlefs, ESRF

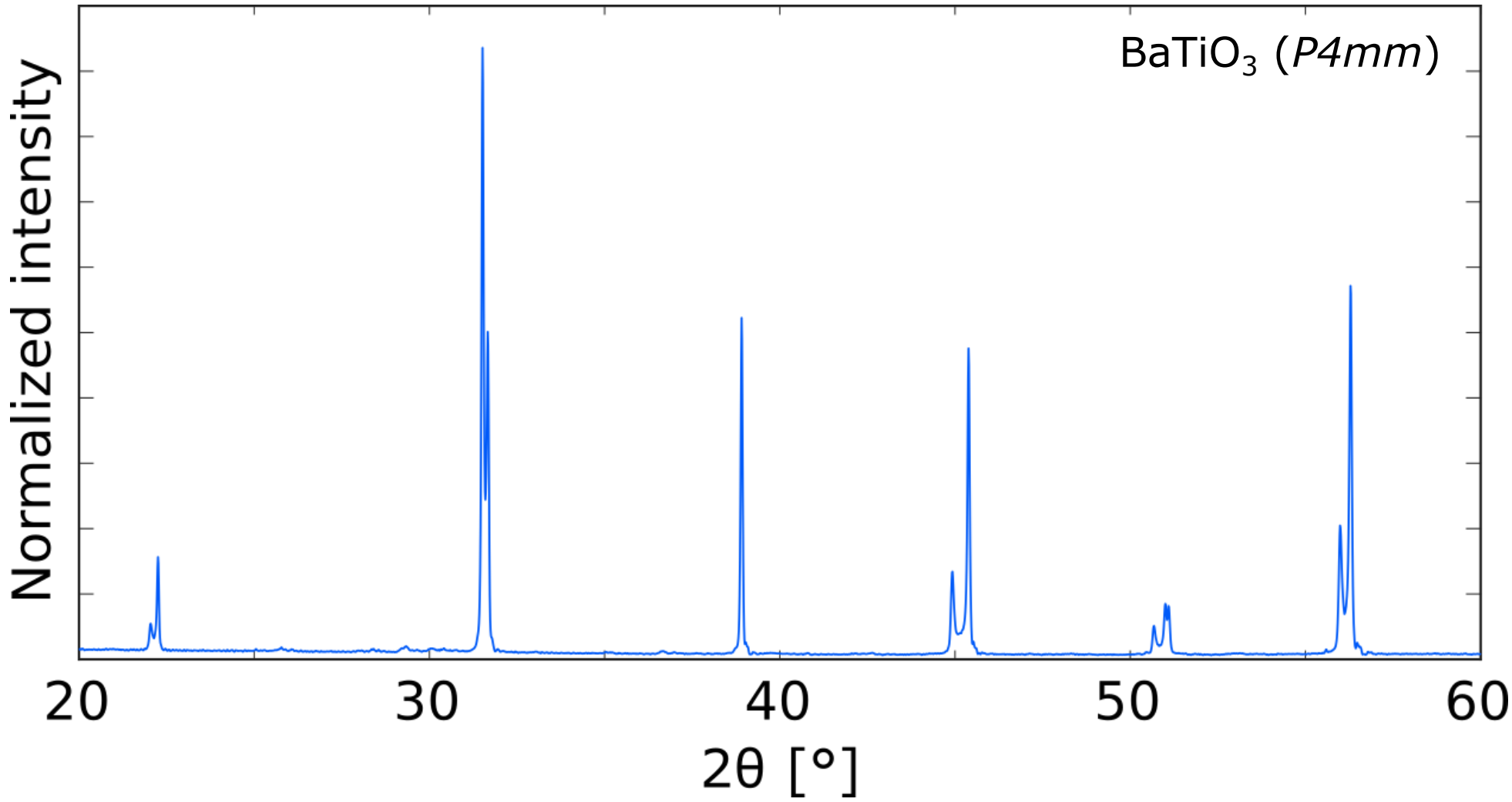
A multi-scale analysis of an artificial diamond shows the dislocation network, individual dislocation lines, and a single stacking fault embedded deep within the crystal. After the EBS upgrade, with the EBSL2 beamline, it may be possible to track the 3D motions of these features with sub-second resolution as the material is stressed.

Application example:

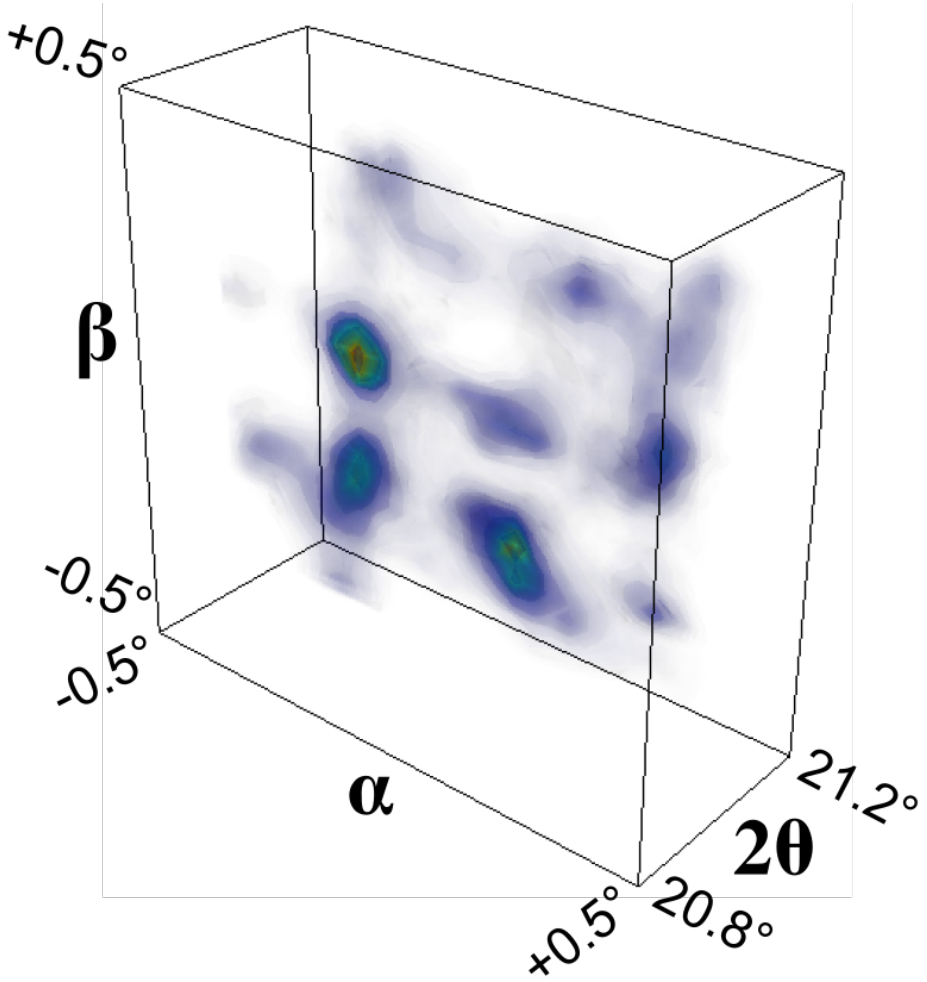
Strains in ferroelectric ceramics

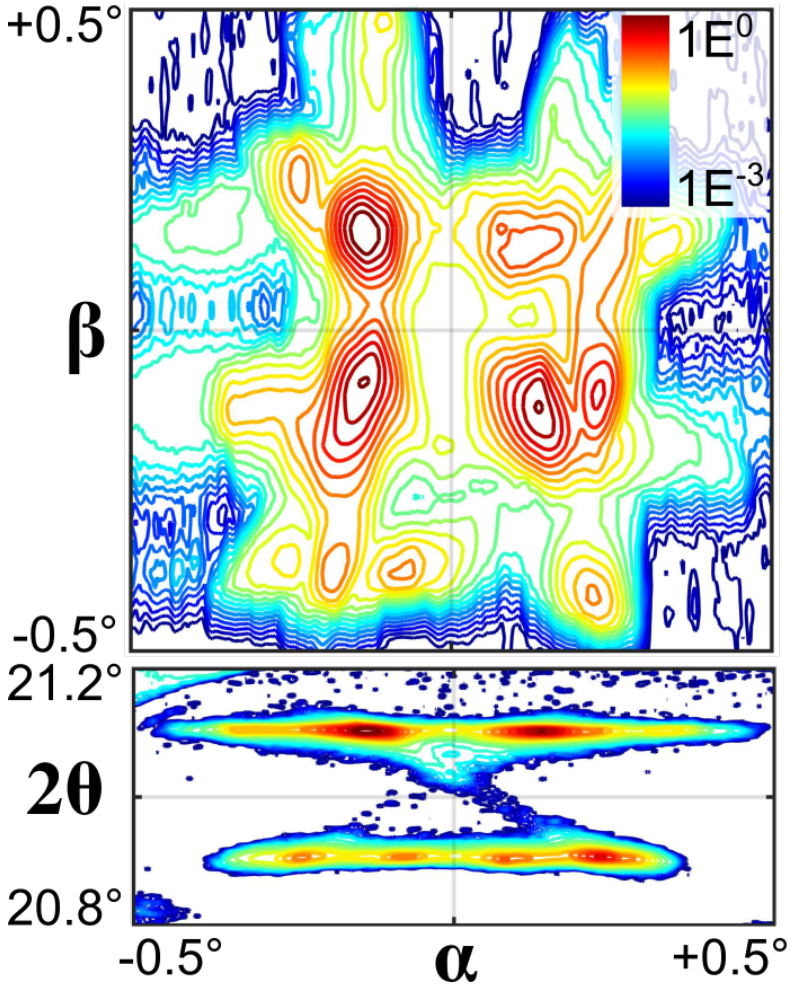
H. Simons, J. E. Daniels et al. *Nature materials*, *17*(9), (2018).

Polycrystalline $BaTiO_3$:

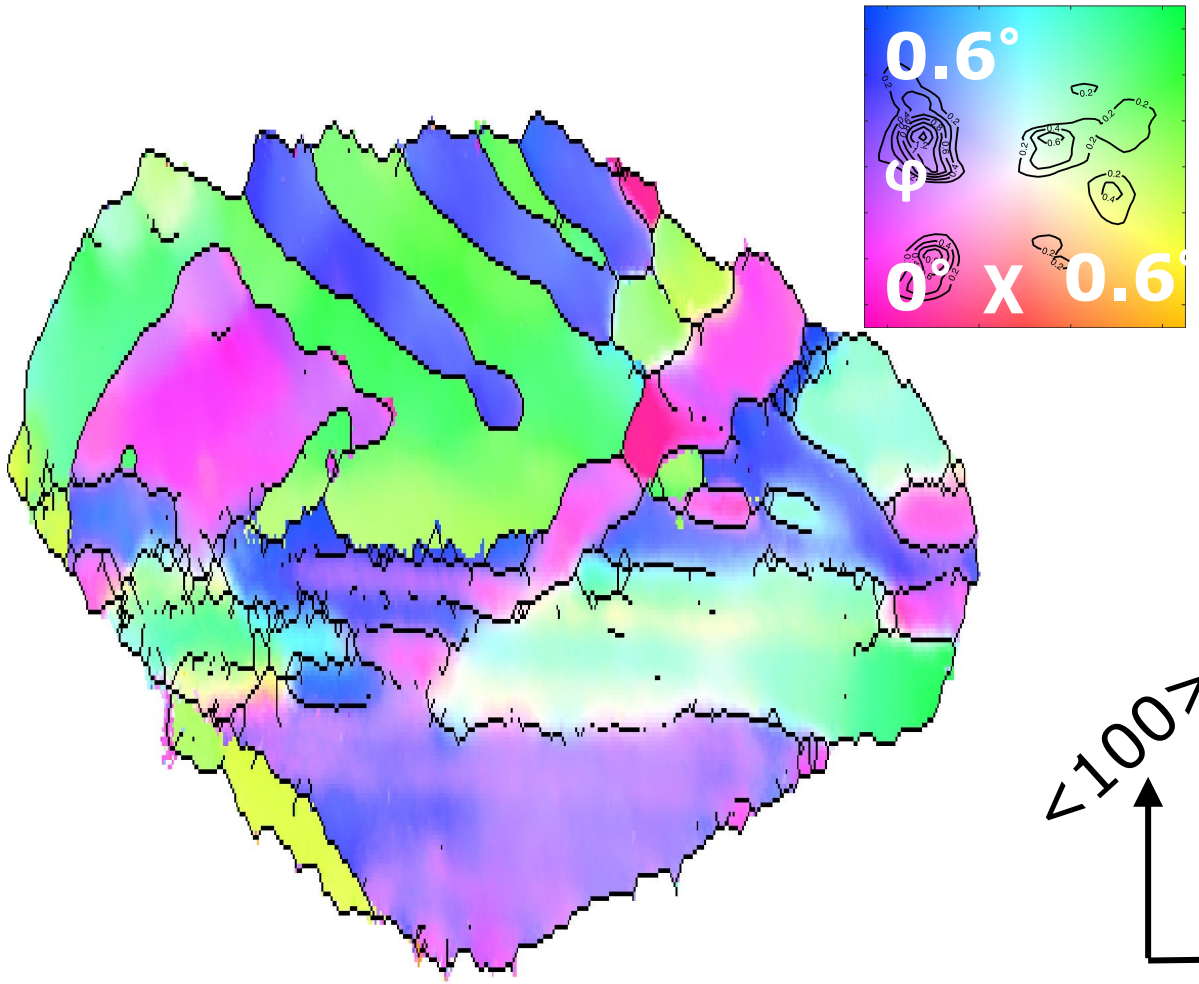


Grain-scale diffraction doesn't support this!

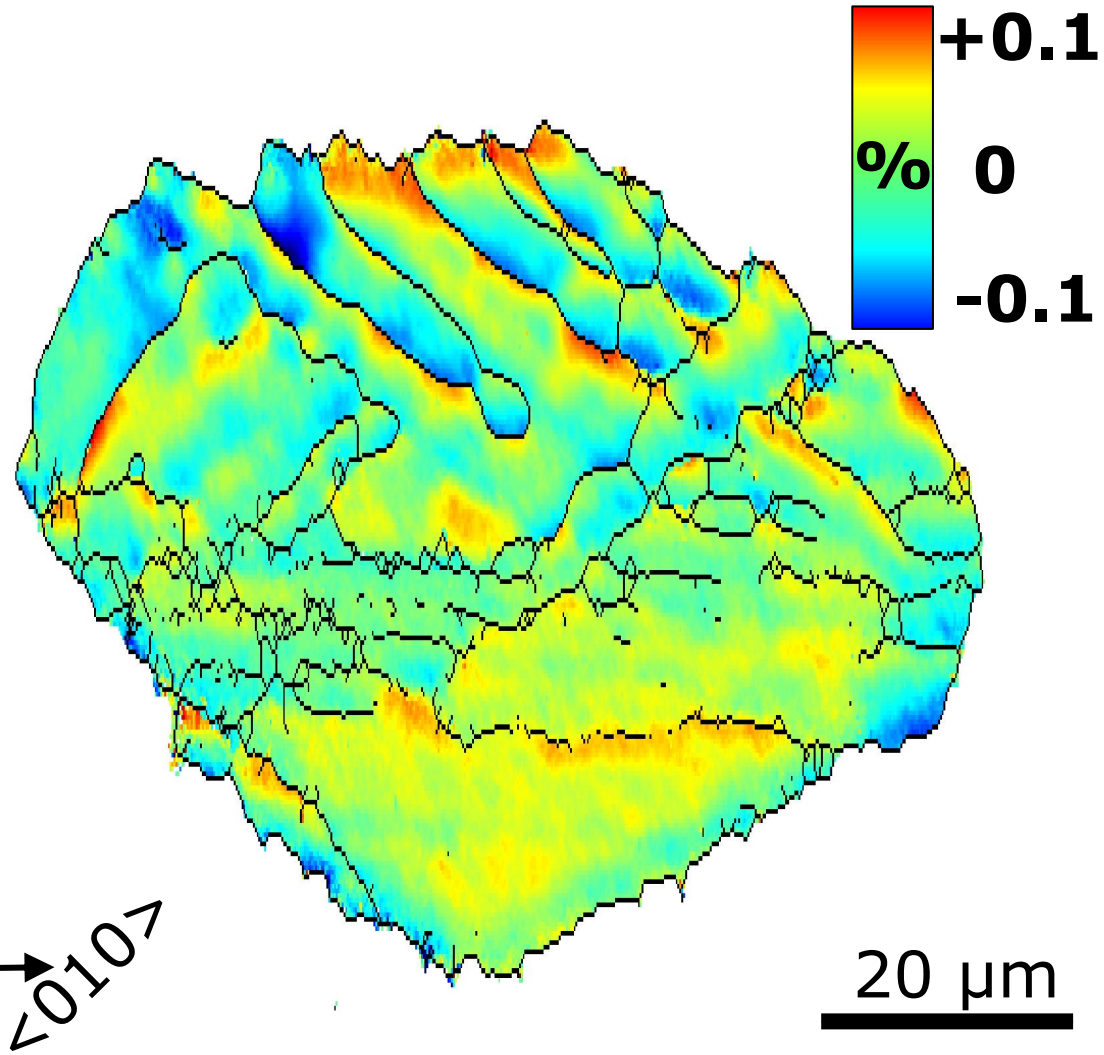




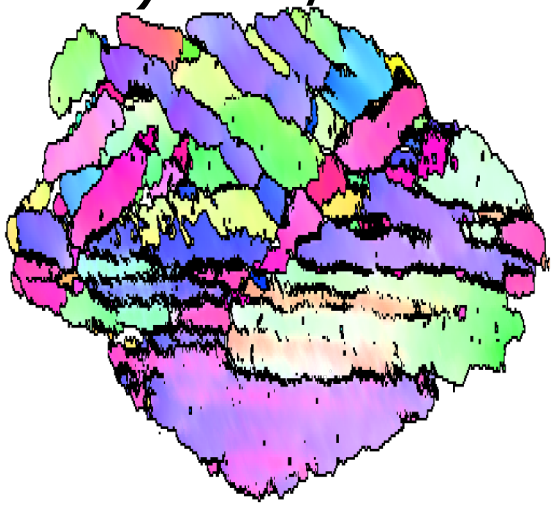
Lattice orientation (φ,χ):

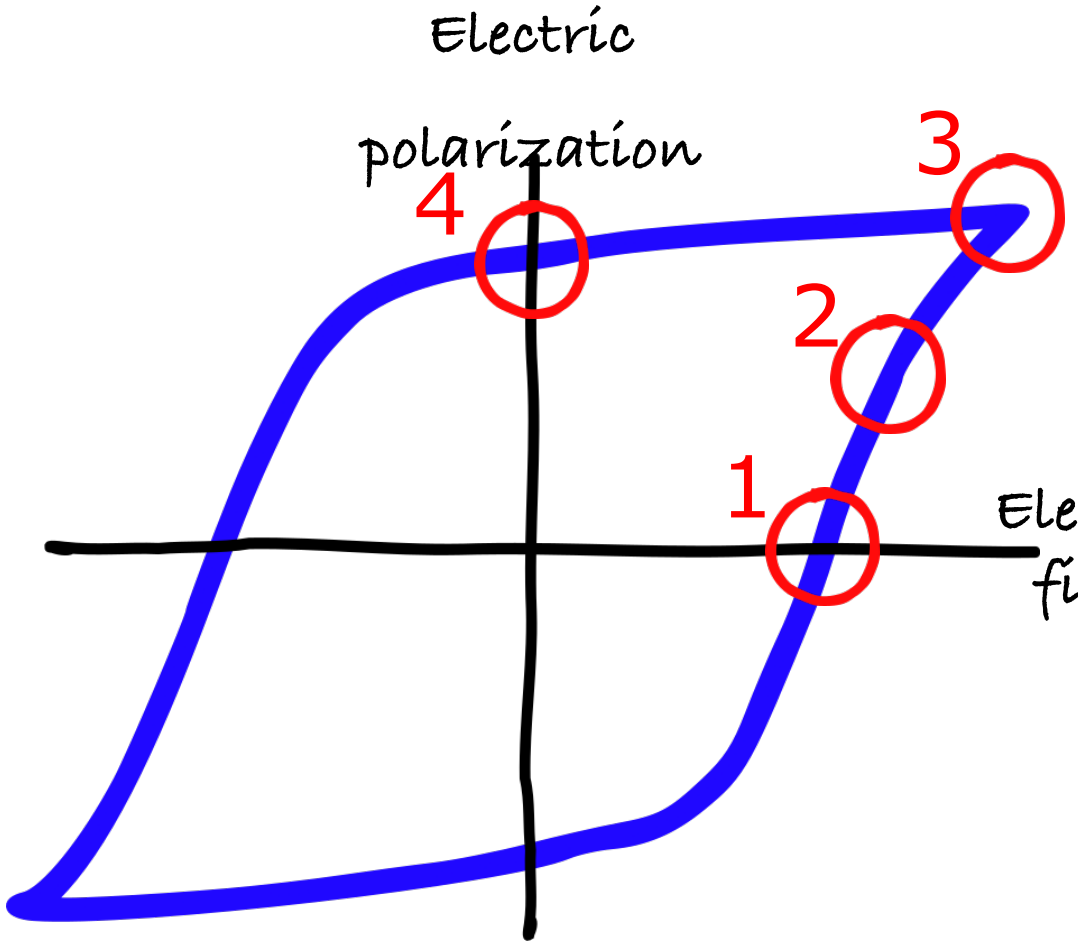


Lattice strain (ε_{33}):

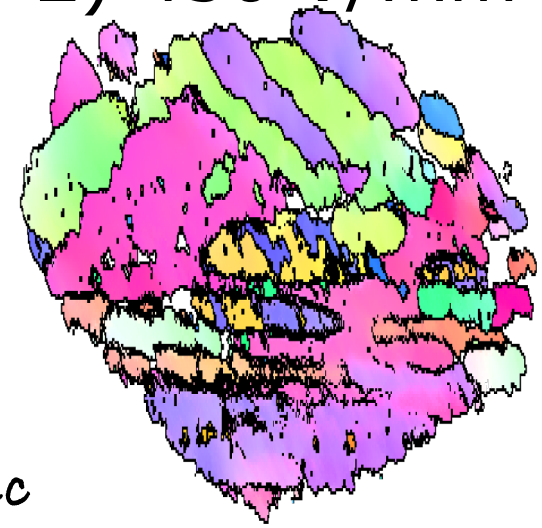


1) 0 V/mm

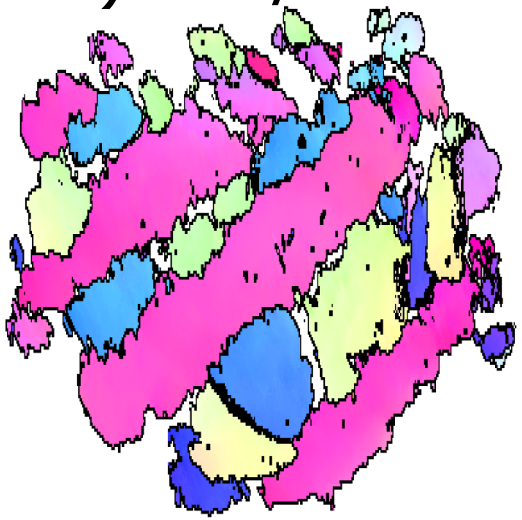




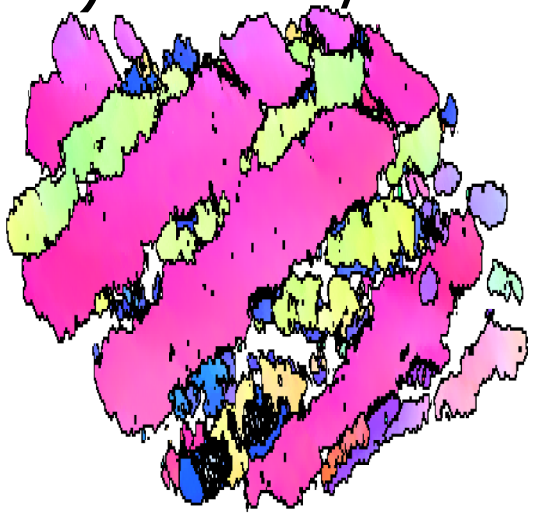
2) 450 V/mm



4) 0 V/mm

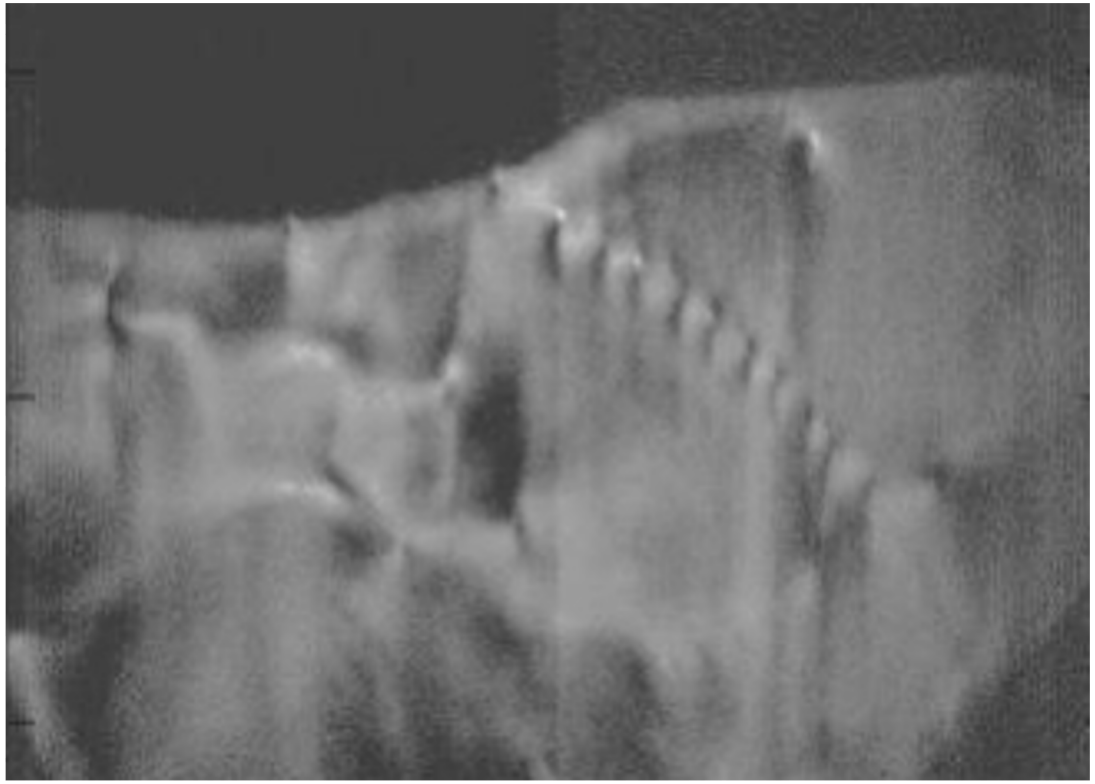


3) 900 V/mm



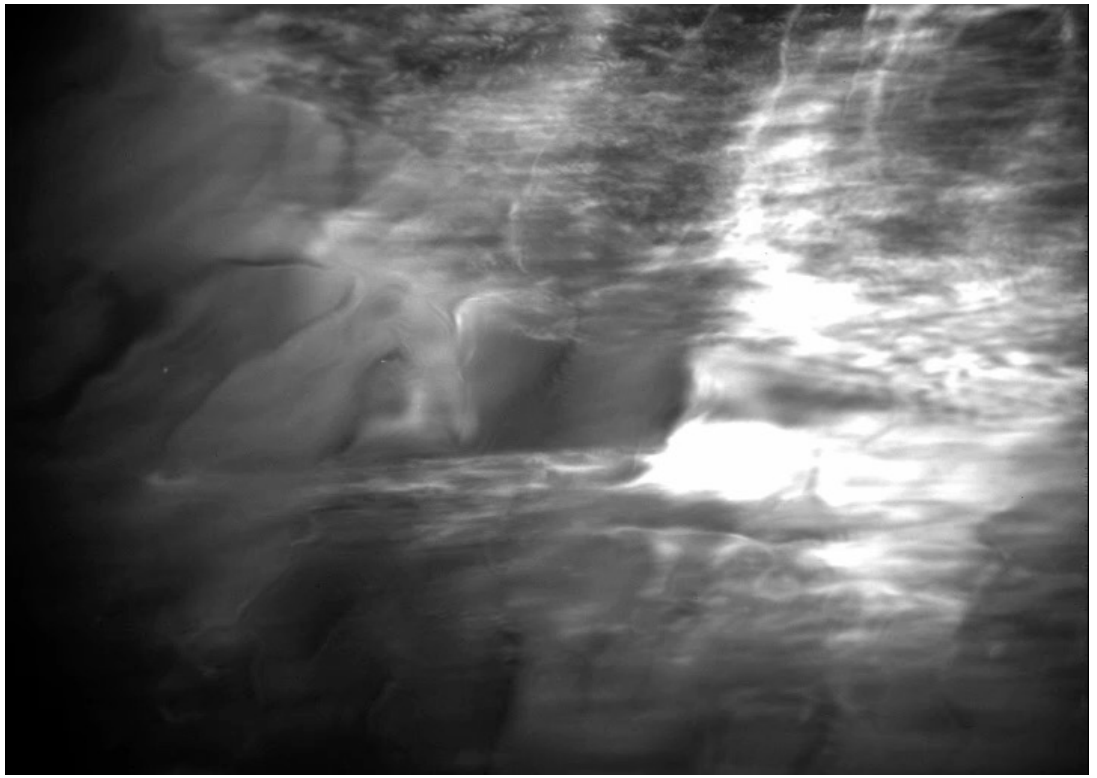
Other useful capabilities:

Imaging real-time dynamics:



L. Dresselhaus-Marais, H. Simons et al.,
Science Advances (2021)

Imaging defects in 3D:

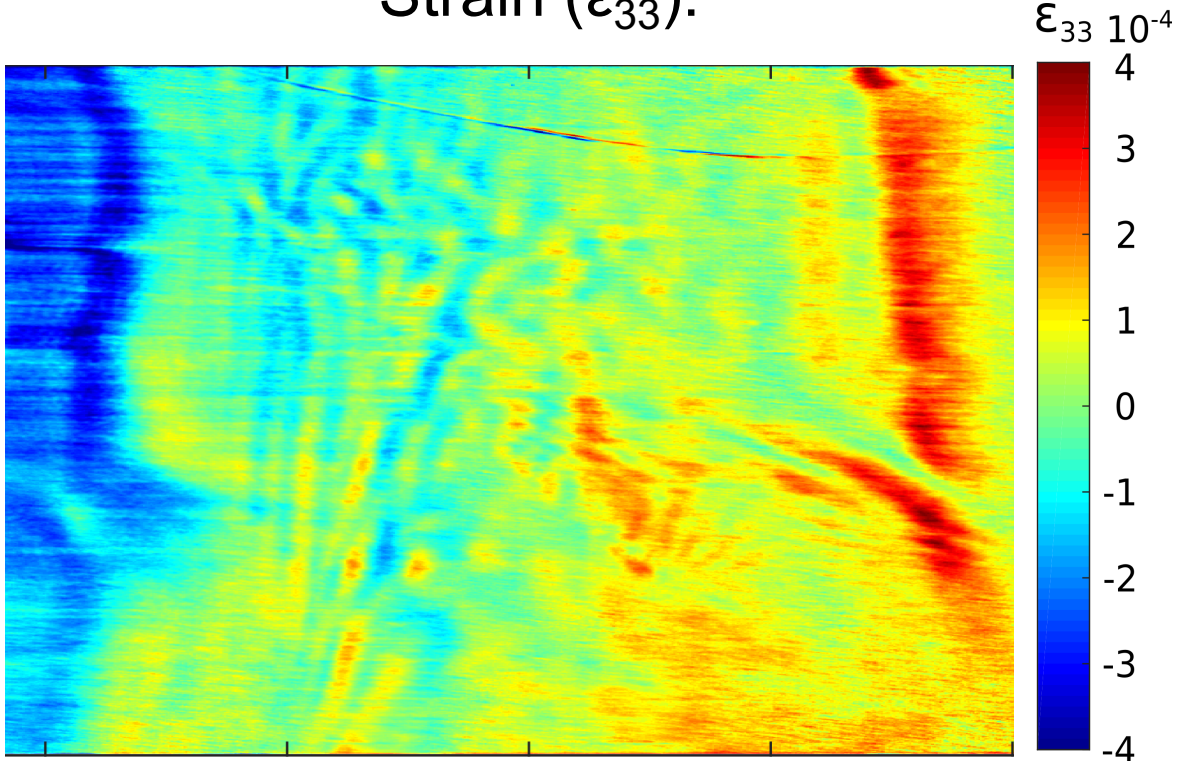


L. Porz, H. Simons et al., *ACS Nano (2021)*

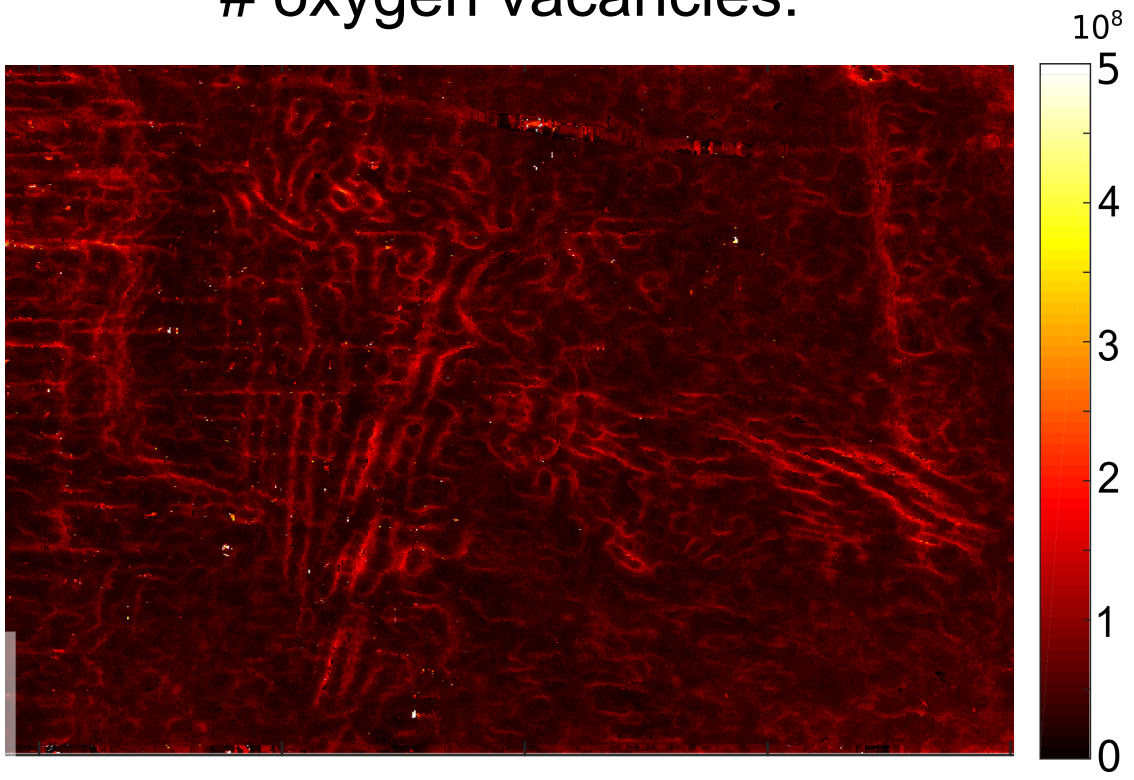
Ongoing developments…

Quantifying structures smaller than a pixel:

Strain (ε_{33}):



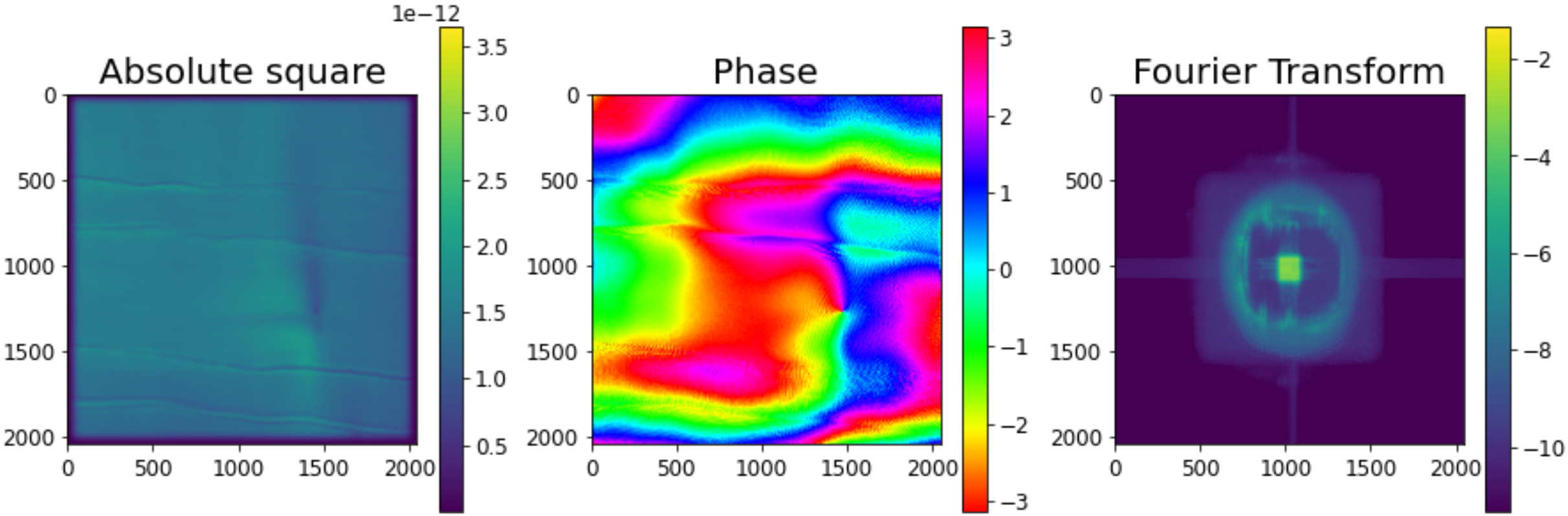
oxygen vacancies:



(Requires assumptions regarding defect type & distribution)

A. Gayoso, J. Ormstrup, H. Simons et al., In preparation (2021).

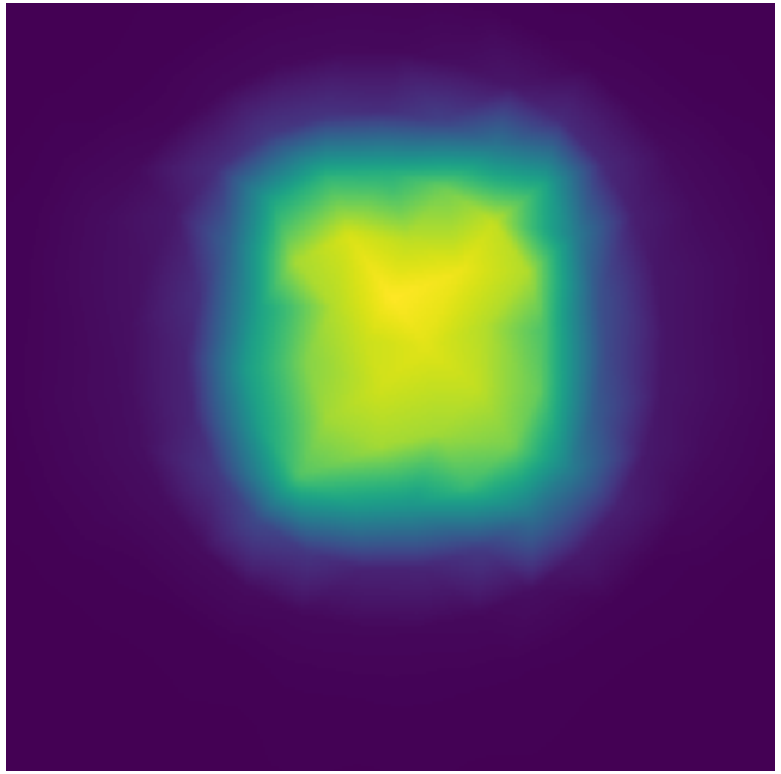
Utilizing coherent properties of upgraded source:



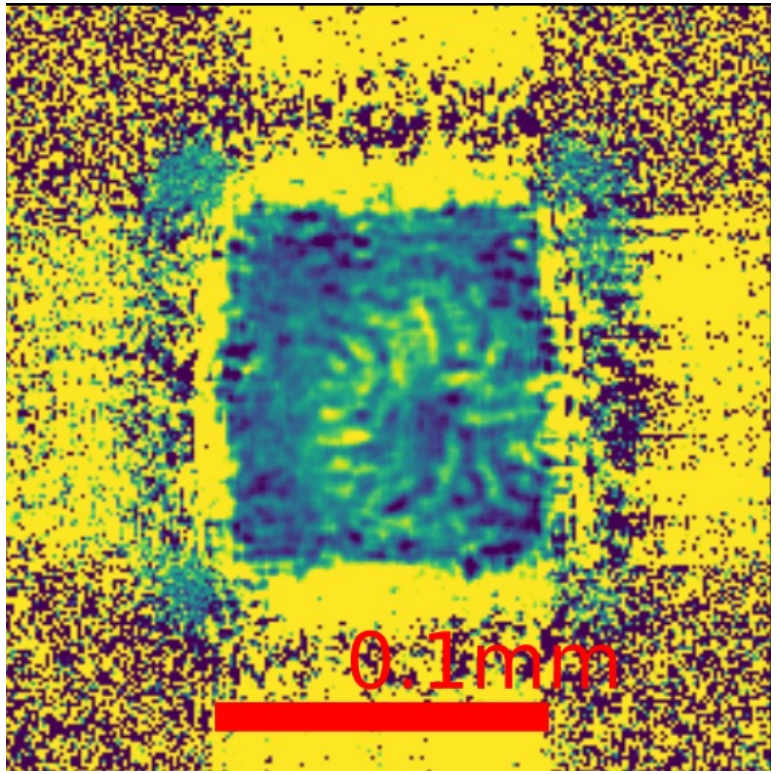
M.A. Carlsen, H. Simons et al., In preparation (2021)

Utilizing coherent properties of upgraded source:

Incoherent:

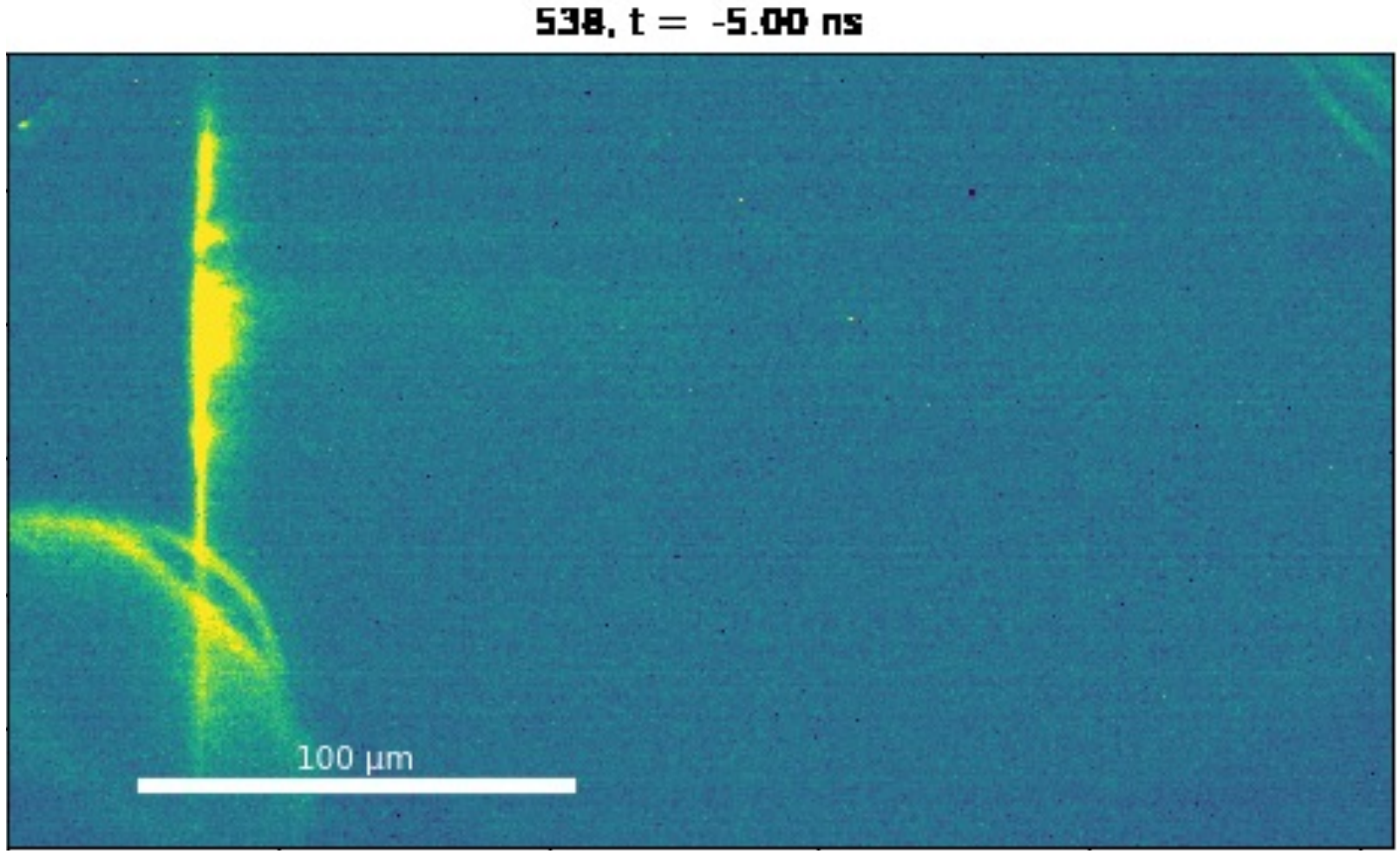


Coherent:



M.A. Carlsen, H. Simons et al., In preparation (2021)

Ultra-fast imaging (i.e. DFXM at X-FELs)



T.S. Holstad, T. Ræder, K. Haldrup, H. Simons et al., In preparation (2021)

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Leora Dresselhaus Marais