EUROPEAN SYNCHROTRON RADIATION FACILITY
ESRF User Office
CS 40220, F-38043 GRENOBLE Cedex 9, France
Delivery address: 71 avenue des Martyrs, 38000 GRENOBLE, France
Tel: +33 (0)4 7688 2552; fax: +33 (0)4 7688 2020; email: useroff@esrf.fr; web: http://www.esrf.fr

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Application for beam time at ESRF – Project Description

Proposal Summary: Diffraction limited storage ring (DLSR) synchrotrons such as the ESRF-EBS, APS - Upgrade and others increase the coherent hard x-ray flux density by 2-3 orders of magnitude compared to previous sources. We propose a novel coherent diffraction imaging technique to capitalize on this revolutionary flux and enable near-atomic to atomic resolution Bragg coherent diffraction imaging (AR-BCDI). This method is based on a direct method approach incorporating molecular dynamic simulations and maximum likelihood estimation methods. We request beamtime at the upgraded ESRF-EBS beamline ID01 to gather the necessary multi-reflection Bragg peak data and further refine our algorithm. ID01 is one of a few possible facilities currently world-wide that has sufficient coherent flux density to test this algorithm. However, a complication from such high flux densities has been sample damage and movement due to radiation pressure. We plan to overcome these challenges with a novel dewetted gold nanocrystal sample on strontium titanate substrates that is robust to radiation pressure and presents many advantages over previous samples for collecting AR-BCDI data at ID01.

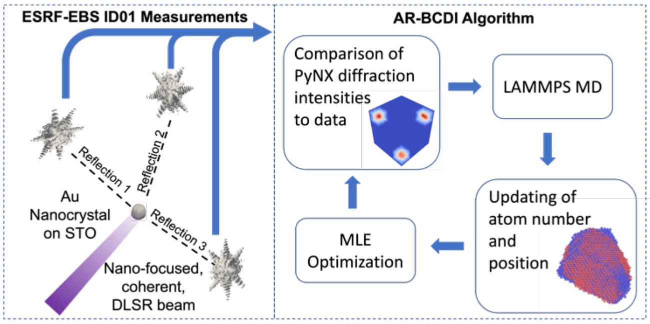

Fig. 1: Proposed measurement (left) at ID01 and algorithm for AR-BCDI (right). A nano-focused 10 keV beam a dewetted gold nanocrystal and multiple Bragg reflections are measured. These hkl values are fed into the AR-BCDI algorithm that initializes with a set of random atomic positions in LAMMPS molecular dynamics. LAMMPS and maximum likelihood estimation techniques then refine the atomic positions until the error converges with less than 1 Angstrom average error.

Scientific background: Coherent diffraction imaging (CDI) has always been the potential for wavelength limited resolution, however, the achieved resolution has always been many times larger than this for hard x-ray wavelengths. One of the most successful variants of CDI is Bragg CDI where oversampled diffraction patterns are measured by measuring a rocking curve and has provided time resolved nanoscale resolution in single crystal [1,2] and polycrystalline samples [3]. In BCDI a coherent x-ray beam illuminates a crystalline sample, and a diffraction pattern is collected on a pixelated detector at an allowed Bragg peak reflecting from the sample. The sample is then rotated and the detector measures 2D slices of the 3D Bragg peak intensity. A computer phase retrieval algorithm reconstructs a high-resolution image from the recorded diffraction patterns. When multiple Bragg peaks are measured, 3D strain maps to a few nanometer resolution have been demonstrated [4–6].

However, several challenges have prevented atomic resolution BCDI to be achieved: (1) available hard x-ray coherent flux, (2) large regions of reciprocal space being measured, (3) prohibitively large data sets and computational cost, and (4) samples that are not robust to the needed flux densities. In this beamtime, we plan to overcome all these limitation with a novel AR-BCDI algorithm and dewetted gold sample. First, we have developed a novel direct method for solving the phase problem based on atomic positions defined and refined through subsequent molecular dynamics simulations in the LAMMPS package (see Fig. 1). These positions are forward modeled with the PyNX package and simulated diffraction intensities are compared with the measured Bragg reflections. Atoms are added or removed from the sample by testing their energy via LAMMPS relaxations and positions are then refined through maximum likelihood estimation (MLE) optimization methods (*J. Meziere et al, in preparation 2023*). This method has robustly reconstructed strained nanocrystals in 2D and 3D with three simulated Bragg peaks. It has the following advantages to overcome challenges (2) and (3) above. It does not need to have continuous measurements of reciprocal space in order to converge and therefore has much smaller datasets compared with previous proposals for more traditional AR-BCDI [7]. We have been able to reconstruct on the order of 5000 gold atoms in simulation in less than 10 hours on a single GPU node with Poisson noise and only on the order of 1 million measured photons between the three Bragg peaks. We have been able to reconstruct grains with vacancies and screw dislocations with similar flux simulated measured in Bragg peaks from 34ID-C at APS (~10^8 photons per Bragg peak).

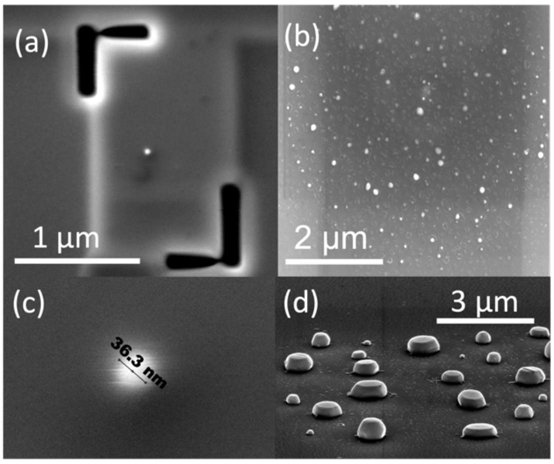

Fig. 2: SEM images Au on STO samples: (a) isolated <40 nm Au nanocrystal with fiducials; (b) dense patch of <100 nm diameter crystals; (c) close up of <40 nm crystal shown in (a); (d) larger nanocrystals from a different sample prepared in a similar way to show high quality faceted crystals.

The second innovation that should enable AR-BCDI at ID01 is the development of dewetted Au nanocrystals on strontium titanate substrates. These samples have produced low strain, high quality nanocyrstals from 20 – 5000 nm in diameter as shown in Fig. 2. We can control the size of these crystals by reducing the Au layer thickness during thermal evaporation prior to annealing and the density with optical lithography. We have routinely illuminated them with intense pink beam at APS 34ID-C during Laue grain mapping and have found that they do not move or rotate as previous dewetted nanoparticles on silicon [8]. During a very recent ID01 ESRF experiment (MA 5371 – PI: Hruszkewycz), we illuminated these samples under the full focused beam and did not see sample rotation or damage. Thus, they appear to be ideally suited for testing AR-BCDI samples. The samples have a preferred 111 orientation out of plane and we can add fiducials for greater ease in finding them at ID01. Previous measurements 111 reflections from a <50 nm grain at APS 34ID-C yielded ~1000 photons per second per pixel in the Bragg peak. Therefore, the enhanced flux at ID01 will be necessary for these measurements.

Experimental plan: We have precharacterized a series of Au on STO samples with SEM/EBSD and at APS 34ID-C during beamtime in April 2023 and orientated a ~40 nm crystal with Laue diffraction microscopy. We were then able to find Bragg peaks from this crystal. This is a major accomplishment and greatly reduces the risk to the experiment and will enable rapid finding of Au nanocrystal Bragg peaks at ID01 without the need for lengthy searches. We will focus on the isolated, fiducial located nanocrystals initially, but if these samples do not work for some reason we will move onto the dense patch of sub-100 nm nanocrystals with 111 out of plane orientation where we have easily found peaks at 34ID-C. If neither of these samples work for some reason, we will try the Pt nanocrystals embedded in sapphire prepared by co-proposer Dr. Marie-Ingrid Richard at the beamline.

Beamline(s) and beamtime requested with justification: We request 6 days of beam time (18 shifts) at Beamline ID01 at ESRF-EBS, which has some of the highest coherent hard x-ray flux in the world (6.4×10^{12} photons s^{-1} μm^{-2} in a zone plate focus of ~100 nm). This is approximately 300 times higher than the flux we have available at 34ID-C at the Advanced Photon Source currently. At our recent beamtime, we are aware of the setup with the Maxipix and Eiger and their limitations. Also, even with this increased flux, we estimate 6-8 hours of measurement per peak, making possibly as much as 24 hours for one nanocrystal (minimum of 3 peaks per nanocrystal). With sample finding, aligning, and testing multiple grains, we expect a minimum of 6 days will be necessary.

Results expected and impact - analysis strategy and significance of the results: We plan to obtain the experimental data needed to validate the MD based AR-BCDI algorithm we have developed. This would be a first for coherent x-ray imaging and especially BCDI and would be applicable at new DLSR world-wide. It would also enable revolutionary new studies of materials phenomenon such as catalysis and battery science and damage and failure in metals via visualization of strain and disclocation dynamics at the atomic scale.

References

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