Spectroscopic BCDI: combined chemical and structural 3D mapping of bimetallic nanocatalysts

Thomas Sarrazin[1], Marie-Ingird Richard[2], Vincent Favre-Nicolin[1]
[1] *ESRF, France*
[2] *CEA/ESRF, France*

Presentation type: Oral **Presentation time:** 09:45-10:00

Strain, composition, and defects are difficult to disentangle in heterogeneous catalysts. Bragg coherent diffraction imaging (BCDI) [1] retrieves three-dimensional displacement fields and morphology with ~10 nm resolution from a single nanocrystal, but composition and strain produce overlapping contrast in the reconstructed phase. The ambiguity is acute in chemically heterogeneous bimetallics such as Ni-Fe and Pt-Pd, alloys used in CO hydrogenation catalysis, where composition and structure rearrange together under reaction conditions. We are developing nano-focused 3D spectroscopic Bragg coherent imaging (spectro-BCDI) at the ID01-EBS nanoprobe of the European Synchrotron (ESRF). The method combines BCDI with multiwavelength anomalous diffraction (MAD) [2].

Scanning the incident energy across an absorption edge modulates f and f of one element. This adds chemical contrast (composition, oxidation state, local bonding) to the same Bragg reflection that already reports lattice strain, displacement, and defects. Simultaneous nano-focused fluorescence provides element-specific concentration and edge-step calibration. Reconstruction proceeds in two stages. We first fit the diffraction intensities across the energy scan at each point in reciprocal space, using the known f and f , to separate the anomalous and non-anomalous structure factors. Each is then phase-retrieved separately, and the voxel-wise ratio of the two reconstructed densities gives the local composition.

We validate the method on simulated diffraction data with realistic photon statistics and recover composition maps across the particle volume. We demonstrate the technique on two systems: Ni Fe nanocrystals at the Ni K-edge (8.333 keV) and PtPd nanocrystals at the Pt L -edge (11.564 keV). The two cases differ in morphology, fluorescence background and anomalous contrast. We aim to extend the method to in-situ CO hydrogenation, where we can follow how composition and strain evolve together in single nanocrystals during reaction and relate structure to catalytic selectivity.

References

[1] I. Robinson and R. Harder, Coherent X-ray diffraction imaging of strain at the nanoscale, Nat. Mater. 8, 291 (2009).
[2] V. Favre-Nicolin, M. G. Proietti, C. Leclere, N. A. Katcho, M.-I. Richard, and H. Renevier, MAD and DAFS for studying Semiconductor Nanostructures, Eur. Phys. J. Spec. Top. 208, 189 (2012).