

Editorial

Special Issue: Electron Diffraction and Structural Imaging—Volume II

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Following the success of the first edition of our Special Issue “Electron Diffraction and Structural Imaging”, we present Volume II, featuring new and innovative contributions that further expand the scope and depth of this rapidly evolving field.

Driven by improved TEM hardware (e.g., Cs correctors, direct detector cameras) and novel techniques such as beam precession, 4D-STEM, and 3DED, electron diffraction (ED) and structural imaging enable atomic-scale analysis of a wide range of materials, including nanocrystals and beam-sensitive compounds. ED’s low dosage and nanometric spatial resolution are ideal while X-ray methods fall short. Combining these methods with in situ sample holders, researchers can now study materials in real-time conditions, uncovering structural changes and symmetry evolution at the atomic level [1–10].

Volume II of this Special Issue features nine original contributions that showcase the versatility, depth, and continued innovation in the sphere of electron diffraction and structural imaging.

The first article, by Andrusenko et al., presents two new organic co-crystals formed via a simple solution growth method using paracetamol or metacetamol and 7,7,8,8-tetracyanoquinodimethane (TCNQ). These compounds belong to a class of organic charge-transfer complexes, with the acetamidophenol molecule acting as an electron donor and TCNQ as an acceptor. Due to the crystals’ sub-micron sizes, precession-assisted 3D electron diffraction was employed for structural characterization. While the paracetamol–TCNQ structure was solved using standard direct methods, the metacetamol–TCNQ system required merging two datasets and applying simulated annealing due to its lower symmetry and data resolution. Both co-crystals have 1:1 stoichiometry, mixed-stack configuration, and non-centric P1 symmetry. Interestingly, they lack the strong hydrogen bonding seen in related orthocetamol-based co-crystals [11].

The second paper, by Hernández-Robles et al., investigates the relative orientations of rotated graphene bilayers (RGBs) grown via chemical vapor deposition. Using selected-area electron diffraction, the authors identified spontaneously preferred orientations that minimize lattice complexity based on possible Burgers vectors. They introduce the concept of secondary singular interfaces—non-singular orientations that still fulfill criteria of singularity based on their angular proximity to true singular states. These notable interfaces exhibit simpler displacement fields and reduced strain, suggesting that RGBs may reorient spontaneously toward such configurations. The study also provides a new explanation for the emergence of high- Σ interfaces, previously overlooked due to limitations in defining singularity [12].



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The third paper, by Jandl et al. from Eldico Scientific, reports the absolute structural determination of anhydrous zinc (II) tartrate metal–organic frameworks (MOFs) with the chiral ligands [Zn (L-TAR)] and [Zn (D-TAR)] using electron diffraction on sub-micrometric crystals. The structures crystallize in the rare I222 space group, confirmed by dynamical refinement, which showed a clear distinction between enantiomorphs based on R-factor differences. These MOFs form dense 3D networks dominated by octahedral coordination, and similar structures were shown to be synthesizable using other divalent metals (Mg, Mn, Co, Ni, Cu). The chiral frameworks are described by Δ and Λ configurations, highlighting the potential of ED for the absolute structural determination of small crystals of chiral MOFs [13].

The fourth paper, by Chou et al., examines how precession angle, energy filtering, and sample thickness affect the structural analysis of amorphous SiO₂ thin films using electron reduced density functions. The authors find that while peak positions remain largely stable across conditions, peak intensities—and thus the derived coordination numbers—are significantly influenced by both precession and energy filtering. Notably, using small precession angles ($\leq 2^\circ$) and energy filtering yields coordination numbers for Si and O that better match the expected values of 4 and 2, particularly in thicker samples [14].

The fifth paper, by Passuti et al., introduces scanning precession electron tomography (SPET) as an advanced method for the accurate structural analysis of epitaxial perovskite thin films, particularly when the region of interest (ROI) is very small. By combining precession-assisted electron diffraction tomography (PEDT) with scanning over a defined area, the authors extracted spatially resolved intensities for precise structural refinement. A 35 nm PrVO₃ film on SrTiO₃ was used as a test case, revealing subtle structural variations across film thicknesses. It was demonstrated that SPET is a powerful alternative to traditional PEDT, capable of providing accurate structural data in ROIs as small as 10 nm [15].

The sixth paper, by Truong et al., shares practical insights from Rigaku's extensive experience with 3D electron diffraction (3D ED/MicroED) for solving structures of sub-micron single crystals. The authors highlight three key conclusions: (1) Cryo-transfer significantly improves data quality for hydrated compounds via preventing dehydration, as shown with trehalose dihydrate; (2) a streamlined workflow for dynamical diffraction enables reliable absolute structural determination for both small and large organic molecules (e.g., tyrosine and clarithromycin); (3) the crystal-to-detector distance must be optimized (even for small molecules such as cystine, longer distances can yield better results). These findings help define best practices across diverse sample types [16].

The seventh paper, by Snopiński et al., explores a novel grain boundary engineering (GBE) strategy for additively manufactured AlSi10Mg alloys using the KoBo extrusion method, which enables thermo-mechanical processing in a single step. Through EBSD and TEM analysis, the authors observed not only significant grain refinement but also an increased fraction of coincidence-site lattice (CSL) boundaries—especially low- Σ twin boundaries—indicating enhanced grain boundary character. The study links CSL formation to dynamic recrystallization, suggesting that KoBo extrusion offers a promising route to engineer tailored grain boundary networks in aluminum-based AM components. These results pave the way for next-generation high-performance alloys [17].

The eighth paper, by Gallegos-Moncayo et al., investigates cathode electrolyte interphase (CEI) formation in NMC 811 (LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂), a key challenge in high-capacity lithium-ion batteries. Due to the proximity of the material's LUMO level to the HOMO of liquid electrolytes, oxidation reactions lead to surface degradation during charging. Using precession-assisted 4D-STEM-ACOM/ASTAR and STEM-EDX, the authors examined CEI composition at 4.3 V and 4.5 V, revealing a fluorine-rich layer that varied with voltage:

it consisted of LiF alone at 4.5 V and LiF + LiOH at 4.3 V. Despite LiF's reputation as a stable layer, it fails to prevent degradation in NMC 811. The study emphasizes the need for protective components, such as tailored additives or coatings, to enhance stability [18].

The ninth paper, by Örs et al., presents the first complete structural determination of zeolite ECR-1, an aluminosilicate with EON topology, previously inaccessible due to its submicron crystal size and stacking faults. Using a single nano-crystal with minimal defects, the authors performed precession electron diffraction (PED) at 103 K, achieving dynamical refinement with $R_{obs} = 0.097$. The structure comprised 8.16 Na⁺ ions across six crystallographic sites and ~four water molecules per unit cell. The material was hydrothermally synthesized with trioxane as a structure-directing agent, and a Monte Carlo simulation further validated the experimental cation and water distributions, marking significant progress in the structural study of faulted zeolites [19].

Conflicts of Interest: The authors declare no conflicts of interest.

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