

Extending 4D-STEM to Defect and Short-Range Ordering Analysis: Principles, Methodology and Applications

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Defects in crystals, such as dislocations, stacking faults, short-range order, and aggregates of point defects, disrupt coherent Bragg scattering. Under suitable diffraction conditions, the disruption is large enough to give rise to “diffraction contrast” in TEM images formed from a single Bragg beam, which maps out the strain field around a defect [1-3]. At high resolution, using many beams, the atomic arrangement of defects can be observed directly. Together, diffraction contrast imaging and HREM had contributed much of our knowledge of defects in real materials [4]. However, recent developments in advanced materials, such as multi-principal element alloys or high entropy alloys (HEAs), mixed phases battery materials and three-dimensional nanodevices, have posed significant challenges for electron imaging [5]. Additionally, characterization of highly defective materials has always been a challenge for electron microscopy.

With recent developments in fast electron detectors and efficient computer algorithms, it now becomes possible to collect unprecedentedly large datasets of diffraction patterns (DPs) [6, 7]. Data mining of DPs has provided a rich field to explore new electron imaging techniques. Especially, extracting crystallographic information to form images or tomograms based on crystal structural properties is a powerful approach for quantitative analysis of materials microstructures.

This talk focuses on the principles of 4D-STEM based electron nanodiffraction techniques for defect, strain and short-range ordering analysis using electron diffuse scattering [8, 9]. We review recent progress made in scanning electron nanodiffraction (SEND) data collection, new algorithms based on cepstral analysis [8], and machine learning based electron DP analysis [10]. These progresses will be highlighted using defect detection, and short-range ordering analysis as application examples. The materials of the study are the medium entropy alloy, CrCoNi, which has exceptional low-temperature mechanical strength and ductility [11]. We will show how SEND helps our understanding of non-random chemical mixing in a CrCoNi alloy, resulting from short-range ordering (Fig. 1), behind the mechanical strength in CrCoNi and how these developments provide general opportunities for an atomistic-structure study in advanced alloys.

Future opportunities based on smart sampling and machine learning of 4D-STEM datasets will also be discussed. The example here is the metrology of 3D transistor devices. We will highlight the benefits of combined defect, strain and orientation analyses.

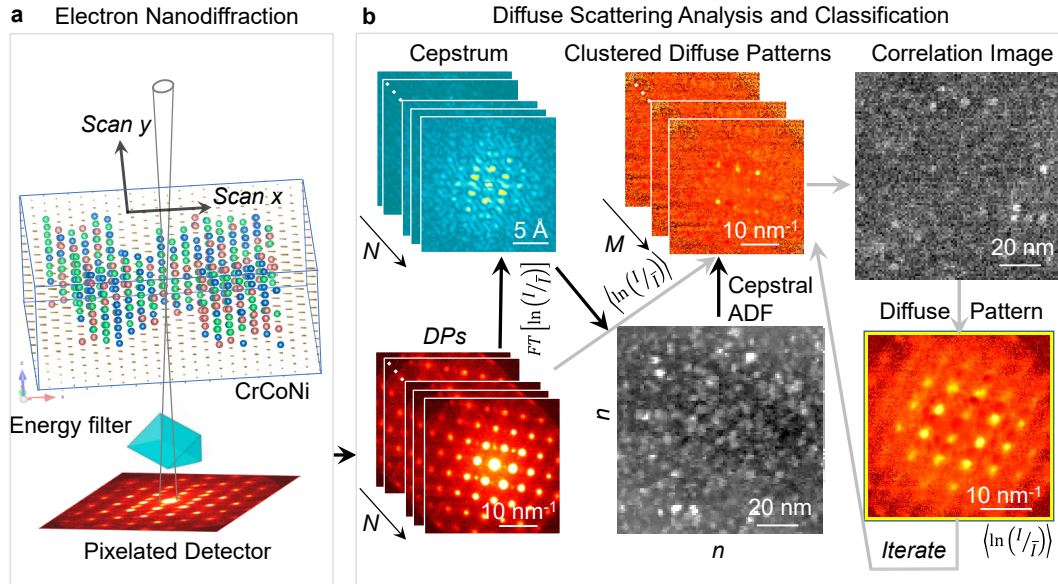


Figure 1 Data mining of SEND datasets for short range ordering analysis [5].

References:

- [1] P. Hirsch, A. Howie, R.B. Nicolson, D.W. Pashley, M.J. Whelan, *Electron Microscopy of Thin Crystals*, Robert E. Krieger Publishing Company, Malaba, Florida, 1977.
- [2] D.B. Williams, B.C. Carter, *Transmission Electron Microscopy, A Textbook for Materials Science*, 2nd Edition ed., Springer, New York, 2009.
- [3] J.M. Zuo, J.C.H. Spence, *Advanced Transmission Electron Microscopy, Imaging and Diffraction in Nanoscience*, Springer, New York, 2017.
- [4] J.C.H. Spence, *High resolution electron microscopy*, 4th Edition ed., Oxford University Press, Oxford, UK, 2013.
- [5] H.-W. Hsiao, R. Feng, H. Ni, K. An, J.D. Poplawsky, P.K. Liaw, J.-M. Zuo, Data-driven electron-diffraction approach reveals local short-range ordering in CrCoNi with ordering effects, *Nature Communications*, 13 (2022) 6651.
- [6] C. Ophus, Four-dimensional scanning transmission electron microscopy (4D-STEM): From scanning nanodiffraction to ptychography and beyond, *Microscopy and Microanalysis*, 25 (2019) 563-582.
- [7] J.-M. Zuo, R. Yuan, Y.-T. Shao, H.-W. Hsiao, S. Pidaparthi, Y. Hu, Q. Yang, J. Zhang, Data-driven electron microscopy: electron diffraction imaging of materials structural properties, *Microscopy*, (2022) In print.
- [8] E. Padgett, M.E. Holtz, P. Cueva, Y.-T. Shao, E. Langenberg, D.G. Schlom, D.A. Muller, The exit-wave power-cepstrum transform for scanning nanobeam electron diffraction: robust strain mapping at subnanometer resolution and subpicometer precision, *Ultramicroscopy*, 214 (2020) 112994.
- [9] Y.-T. Shao, R. Yuan, H.-W. Hsiao, Q. Yang, Y. Hu, J.-M. Zuo, Cepstral scanning transmission electron microscopy imaging of severe lattice distortions, *Ultramicroscopy*, 231 (2021) 113252.
- [10] R. Yuan, J. Zhang, L. He, J.-M. Zuo, Training artificial neural networks for precision orientation and strain mapping using 4D electron diffraction datasets, *Ultramicroscopy*, (2021) 113256.
- [11] B. Gludovatz, et. al., Exceptional damage-tolerance of a medium-entropy alloy CrCoNi at cryogenic temperatures, *Nature Communications*, 7 (2016) 10602.
- [12] This work is funded by NSF DMR MMN (DMR-2226495), Under Program Director, Dr. Jonathan Madison