Towards direct imaging of defects in carbon nanotubes with 4DSTEM

Dr. Antonin Louiset[1], Dr. Daniel FÖRSTER[2], Dr. Jean-Luc ROUVIERE[1], Dr. Vincent JOURDAIN[3], Dr. Christophe BICHARA[4], Dr. Hanako OKUNO[1]
[1]IRIG-MEM, CEA, Université Grenoble Alpes, Grenoble, France, [2]Laboratoire d'Etude des Microstructures, ONERA-CNRS, Université Paris-Saclay, Châtillon, France, [3]Laboratoire Charles Coulomb, CNRS, Université de Montpellier, Montpellier, France, [4]CINaM, CNRS, Université Aix-Marseille, Marseille, France

4-dimensional scanning transmission electron microscopy (4DSTEM) based imaging techniques, in particular multislice electron ptychography, demonstrated unprecedented lateral spatial resolution while also providing depth-resolved imaging of the sample [1]. It is therefore, one of the most promising technique for 3D imaging of TEM samples at the atomic scale. Carbon nanotubes (CNT) are an ideal test subject for multislice ptychographic reconstructions using electron beams. While CNT atomic scale imaging can be achieved by high-resolution HR(S)TEM imaging, resolving their complex atomic structure is still a challenge, mainly because of the overlapping signal coming from both side of the tubes [2]. It becomes even more difficult when one wants to solve the structure of atomic defects inside CNTs. Here, we investigate the efficiency of 4DSTEM experiments combined with multiple iterative phase retrieval methods to resolved topological defects in CNTs. In particular, our goal is to resolve separately both sides of nanotubes using multislice ptychographic reconstructions of a single projection.

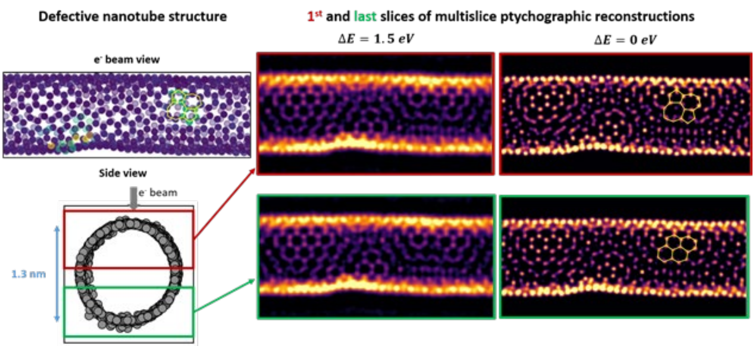
We used defective carbon nanotube structures, obtained by molecular dynamic calculations in a previous work [2], to simulate 4DSTEM datasets. Diffraction pattern simulations were computed using the open source abTEM package [3]. These simulated datasets serve two purposes: optimizing experimental conditions for high resolution imaging of nanotubes and evaluating the efficiency of phase retrieval algorithms. We performed phase images reconstruction using the open source py4DSTEM package [4], which features several iterative phase retrieval algorithms. We focused our work on the following methods: differential phase contrast, parallax, single and multislice ptychography [5]. 4DSTEM experiments were conducted on a double corrected TEM operating at 80kV and equipped with a Schottky field emission gun (X-FEG) and a direct electron detector. Experimental parameters were set as close to the optimum previously determined by simulations and phase images were reconstructed using the previously presented methods

In order to preserve tubes from contamination and irradiation damage, 4DSTEM experiments must be carried out in less than ideal conditions; i.e. low beam current (<20pA), short dwell time per pixel (<2ms), large real space step size (>1.5Å) and defocused STEM probe (tens of nm). In these conditions, the efficiency of the iterative ptychographic reconstructions strongly depends on the first guess given for the incident probe wave function. To optimize the determination of the initial probe parameters, i.e. defocus and residual aberrations, from experimental datasets, we computed differential phase contrast and parallax reconstructions [5], prior to the ptychographic reconstructions. We also acquired a vacuum probe reference in a separated dataset to constrain the incident wave function intensity. This method allowed the reconstructions to converge and strongly improved the resolution of CNTs images compared to high-resolution STEM images. However, phase images did not achieve the ultimate spatial resolution. Using simulated 4DSTEM datasets, we found out that both lateral and depth (in the case of multislice ptychography) resolutions were strongly degraded when dealing with partial temporal coherence, i.e. energy spread, of electron beams. The energy resolution of the microscope used in this study is limited to a FWHM of 1.5 eV. We showed that with a greater energy resolution, which is available on microscopes equipped with a cold FEG gun for example, it is possible to improve lateral resolution and even perform CNT depth sectioning using multislice electron ptychography to capture a direct image of topological defects.

4DSTEM based high-resolution imaging techniques, are very promising to obtain direct imaging of topological defects in carbon nanotubes. By conducting two parallel studies, one based on simulation and one based on real 4DSTEM experiments, we were able to improve the resolution of CNT STEM images by performing iterative phase retrieval reconstructions. We demonstrated that 3D imaging of carbon nanotubes is even possible using multislice electron ptychography when the electron beam's energy spread is reduced. We argue that this limitation can be overcome by state-of-the-art-microscopes and improved reconstruction algorithms.

 https://doi.org/10.1051/bioconf/202412907014

Graphic:



Keywords:

4DSTEM, electron ptychography, carbon nanotubes

Reference:

[1] Chen, Zhen, et al. "Electron ptychography achieves atomic-resolution limits set by lattice vibrations." Science 372.6544 (2021): 826-831.
[2] Förster, Georg Daniel, et al. "A deep learning approach for determining the chiral indices of carbon nanotubes from high-resolution transmission electron microscopy images." Carbon 169 (2020): 465-474.
[3] Madsen, Jacob, and Toma Susi. "The abTEM code: transmission electron microscopy from first principles." Open Research Europe 1 (2021).
[4] Savitzky, Benjamin H., et al. "py4DSTEM: A software package for four-dimensional scanning transmission electron microscopy data analysis." Microscopy and Microanalysis 27.4 (2021): 712-743.
[5] Varnavides, Georgios, et al. "Iterative Phase Retrieval Algorithms for Scanning Transmission Electron Microscopy." arXiv preprint arXiv:2309.05250 (2023).