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Momentum- and Energy-Resolved STEM at Atomic Resolution

Benedikt Haas[1]* and Christoph T. Koch[1]
[1.] Department of Physics & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany.
* Corresponding author: haas@physik.hu-berlin.de

Momentum-resolved STEM (4D-STEM) is an intensely researched area of electron microscopy with significant impact on the field as a whole [1]. We recently investigated the influence of zero-loss filtering on center-of-mass (COM) type analysis of atomic resolution 4D-STEM and found relatively little impact in accordance with theoretical arguments for the robustness of 4D-STEM in the presence of plasmon scattering [2].

Resolving maps of momentum transfer also in energy loss should allow for even more relevant data with multiple potential applications. To facilitate this, we acquired full detector frames with a high beam convergence angle in singly-focusing EELS mode, which offers in addition to the obvious energy-loss axis information about momentum transfer within the filter entrance aperture in the non-dispersive direction. As momentum information is only preserved along the non-dispersive direction of the spectrometer (the diffraction pattern is typically squeezed along the dispersive direction to obtain good energy resolution), we need additional data if we do not want to obtain momentum resolution along two dimensions.

In a simple first implementation, consecutive maps were acquired while changing the projector lenses in between so that a defined rotation of diffraction space by 90° is obtained, without any other changes to momentum space. Several 4D maps with the two different settings were acquired and the resulting 5D data was non-rigidly registered [3] so that the two different momentum directions can be attributed to the same, undistorted positions in real space. This allows to obtain different image moments (in the simplest case COM) of the energy-resolved data. In a more complex acquisition scheme, several rotations of the diffraction plane could be employed and using tomographic reconstruction methods, momentum space could then be reconstructed in 2D if necessary.

We collected data from a monolayer of hexagonal boron nitride using a Nion HERMES microscope operated at 60kV with a convergence angle of 36mrad and 72pA beam current. 8 maps of 128x128 scan points over a 4x4nm area were acquired for the two different projections each. 1028x130 detector pixels were recorded using a Dectris ELA with the 1028 pixels sampling energy loss at 0.5eV per pixel (so that the zero-loss peak and the B and N K-edges are covered) and the 130 pixels in momentum direction sampled about 72mrad in total (2 x the beam diameter of 36mrad). Here, we also applied template-matching afterwards in real space on the 4D-data to further increase the signal (corresponding to more than 10^{10}e/Å^2 dose in the averaged template), as momentum-resolved core-loss data is very faint (this endeavor would probably be futile using a detector with finite read-out noise).

The data can be seen in Fig. 1 with (a) showing an HAADF image of the sample after registration and (b) the template-matched average. One of the two momentum-resolved spectra at the position marked with a star in (b) is provided in (e). When mapping the integrated intensity of the two core-loss regions, trivial maps of the Boron and Nitrogen atoms are obtained (no background subtraction applied here), as seen in (c) and (d). To check the validity of the approach, we then calculated COM using the zero energy-loss region for the two projections and assembled that information into a vector for each scan position. This "zero-loss filtered" COM map is shown in (g) and its divergence in (h). The result clearly shows atomic resolution and looks similar to what one would expect (except for the difference in charge density for B and N, possibly due to the reduced angular range; currently under investigating). Finally, COM is calculated on B core-loss electrons, yielding the maps shown in (i) and (j) (a Gaussian filter was applied as the data is still quite noisy). This clearly demonstrates the experimental feasibility of energy- and momentum-resolved STEM.

Another example is shown in Fig. 2 of GaN in [11-20] orientation, acquired at 200kV using 26mrad convergence angle and 46pA current. 5 maps for each momentum projection of 128x128 scan points covering 8x8nm^2 were registered into a 5D dataset. 135mrad were sampled using 120 detector pixels and 20.6eV around the N K-edge using 50 detector pixels, exposing for 1ms for each scan point in each map (detector pixels later binned to 60x25).

Fig. 2 (a) shows the registered HAADF of the series and (b) again a template-matched region to enhance signal. A momentum-energy map of the position indicated with the pink star in (b) is shown in (c). By row-wise subtraction of the pre-edge signal, the background can be removed without altering the momentum-dependence of the core-loss signal, as depicted in (d). The two orthogonal momentum-projections of a single pixel are shown as the two momentum-energy images (e) and (f) of a position marked in (b) as a mint green star. The COM signal of the N K-edge region is shown in (g) and its divergence in (h).

This methodology should also be able to detect differences in momentum transfer of core-loss electrons from non-degenerate orbitals [4] and therefore allow to image atomic bondings to a certain degree. In addition, applying it in the meV regime offers interesting possibilities for mapping of vibrational signals. Different experiments, aspects of analysis, future applications and an improved implementation will be discussed [5].

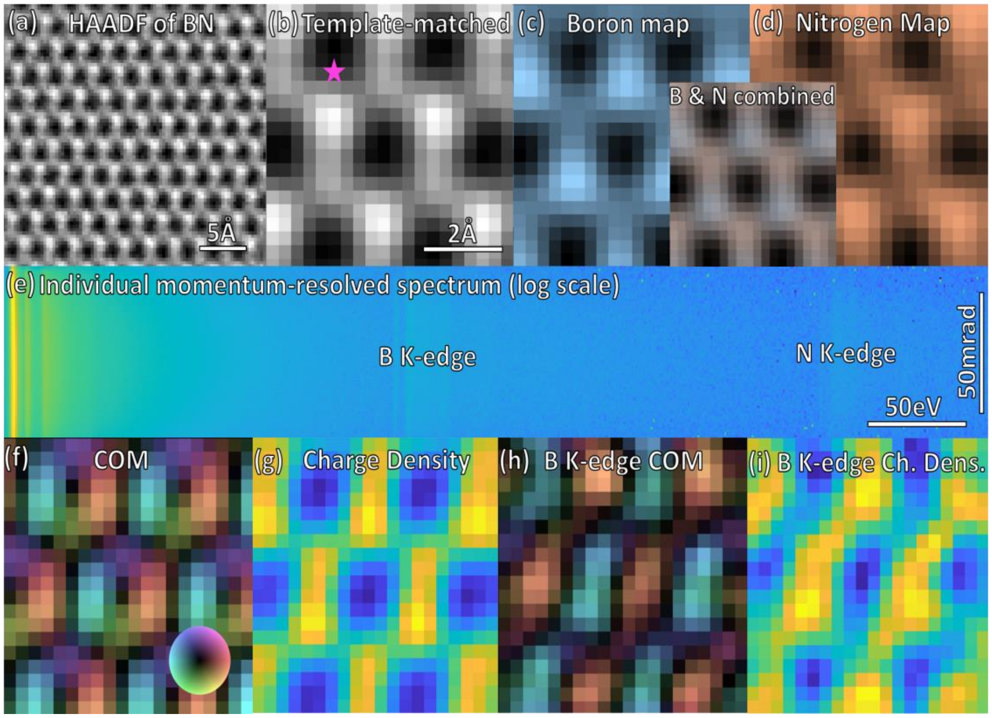

Figure 1. Analysis of 5-dimensional STEM data comprising an energy axis in addition to two spatial and two momentum axes. The data was acquired from an hBN monolayer at 60kV (details in text). An HAADF map of the material can be seen in (a) with the template-matched region in (b). A momentum-resolved spectrum from the position indicated by the star in (b) is given in (e). Another momentum-resolved spectrum exists from the same point with a 90° rotated momentum-axis, but is omitted (cf. Fig. 2). Maps of the integrated B signal and N signal (not background-subtracted) are shown in (c) and (d), respectively, and their combination in (f). The COM signal of the zero-loss peak is displayed in (g) and the charge density map in (h). More interestingly, the same can now also be done for core-loss electrons as shown in (i) and (j), demonstrating a new type of analysis.

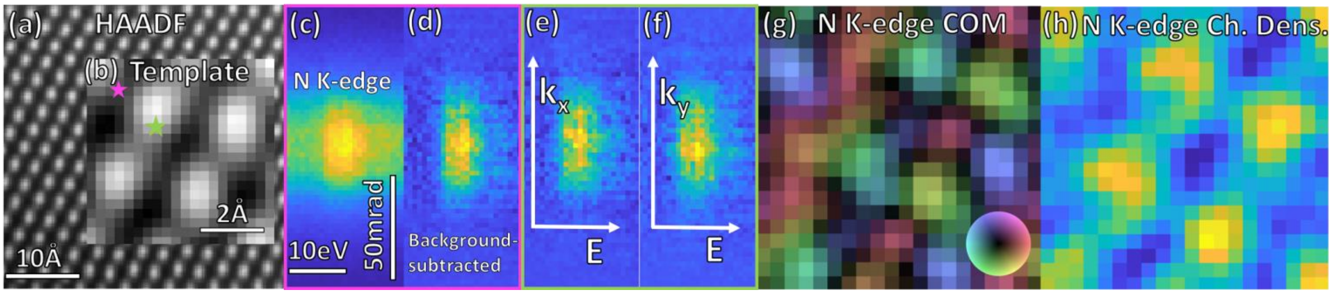

Figure 2. A 5-dimensional dataset from GaN in [11-20] orientation, acquired at 200kV. An HAADF is shown in (a) with the template-matched region given in (b) (these images show only the Ga columns, N is not resolved here). A momentum-energy image around the N K-edge from the region marked with a pink star in (b) is shown in (c) and the line-wise background-subtracted result in (d). Two background-subtracted momentum-energy images with perpendicular momentum projection from the same point marked as the mint green star in (b) are displayed in (e) and (f). The result of COM analysis and its divergence (projected charge density) of the core-loss electrons is shown in (g) and (h), respectively.

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