IM 7: Phase-related techniques

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Mapping of atomic electric fields and charge densities by momentum-resolved STEM

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Aberration-corrected STEM currently achieves a *spatial* resolution down to 50 pm. In contrast, recording STEM diffraction patterns with established circular, annular or segmented detectors provides no or very coarse *momentum* resolution due to the integration of diffracted intensities over large angular domains. Attaining both high spatial and momentum resolution is now feasible by employing ultrafast cameras in STEM. By recording 2D diffraction patterns on a 2D scan raster, a 4D data set is obtained as the central quantity in momentum-resolved STEM. Here we show in simulations and experiments how this can be used to measure atomic electric fields and charge densities directly.
In quantum mechanics, the first moment of a diffraction pattern is related to the expectation value of the momentum transfer and quantitatively yields the angular deflection of the STEM probe [1]. This overcomes ambiguities in conventional differential phase contrast STEM where segmented detectors record portions of the diffraction pattern [2]. Our concept is explained in Fig. (a), showing a focused STEM probe at (1) a nearly field-free region and (2) close to an atom. Whereas the propagation direction is preserved in case (1), the interaction with the electric field for case (2) causes both a distorted wave front and a deflection to the right. For a Ga column in a GaN crystal with 1.3 nm thickness, we simulated the Ronchigrams on the right and determined the first moments as indicated. In this way, the complexity of the Ronchigram condenses to a single vector with fundamental physical meaning: the average deflection of the STEM probe.
Due to Ehrenfests theorem, is proportional to the expectation value of the electric field. For sufficiently thin specimens, is also proportional to the projected electric field , convolved with the STEM probe intensity. Furthermore, its divergence directly yields the projected charge density, convolved with the probe intensity [1,3]. An early 4D STEM experiment for $SrTiO_3$ is shown in Fig. (b), where a slow-scan CCD camera was employed to raster a unit cell with 20^2 STEM pixels. The redistributing Ronchigram intensity in the vicinity of the atomic columns is clearly seen (left). From the first moments, the atomically resolved electric field is obtained (right). As expected from the screened nuclear charge, atoms are sources of the electric field, their magnitude being determined by the atomic number.
We then present 4D-STEM results on 2D sheets of MoS_2 employing the ultrafast pnCCD camera [4] with 4 kHz frame rate. Fig. (c) depicts the projected charge density measured at a mono-/bilayer (ML/BL) edge (left) with respective unit cell averages (right). By comparison with DFT and 4D STEM simulations we show that the data agrees with theory quantitatively. Additionally, a comprehensive analysis of partial coherence effects is given. Finally, we find an AA stacking of the BL and a Mo-terminated ML/BL edge.
With its ability to map electric fields directly without structural input, momentum-resolved STEM can shed light on the electrical configuration of vacancies, dopant atoms or polarisation fields in future, which we exemplarily discuss using simulations [3].

References:
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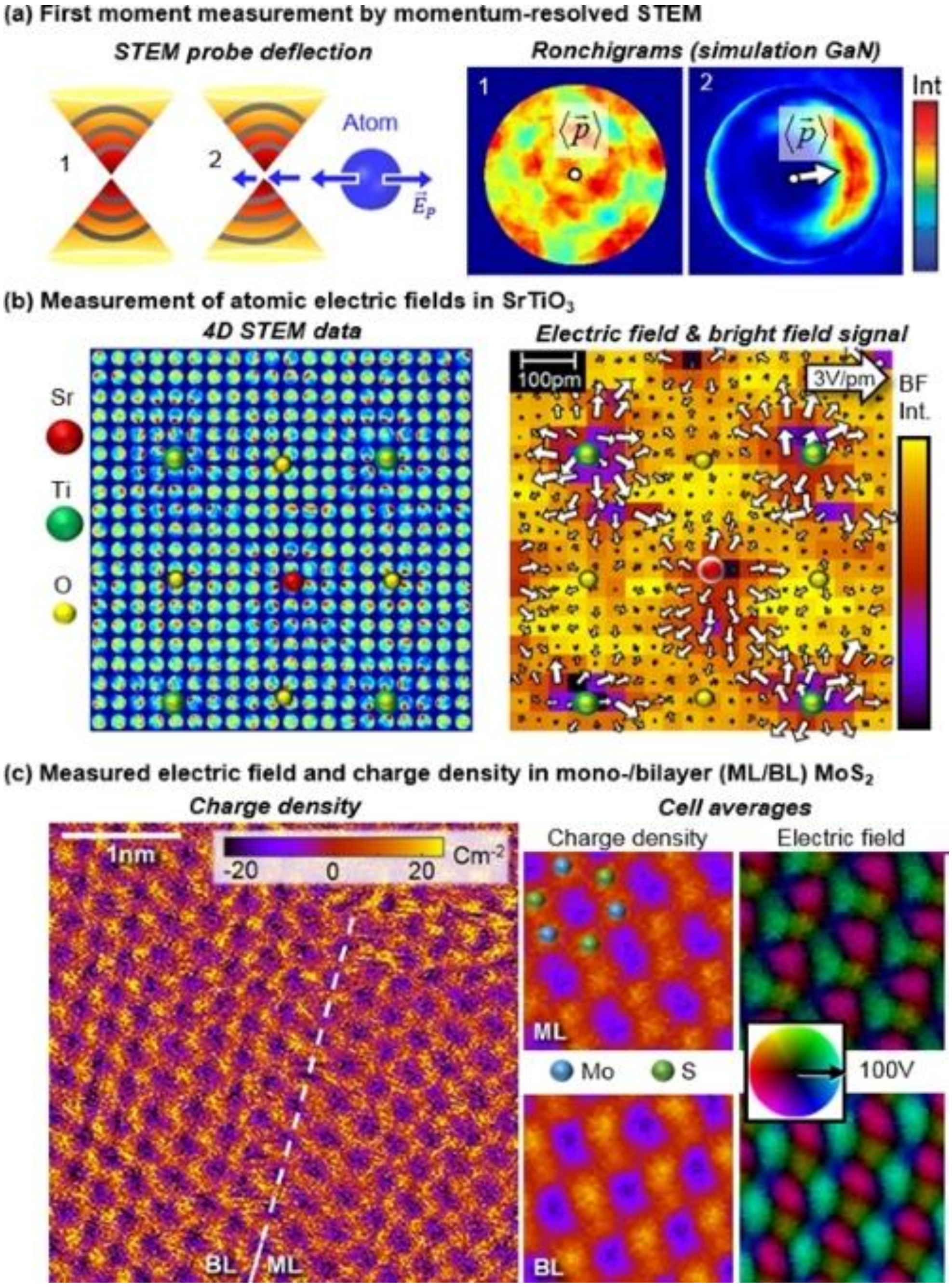

Figure 1. Demonstrating the principle and applications of momentum-resolved STEM: (a) The first moment (centre of mass) is an exact measure of the average angular deflection of the beam. (b) Using a slow-scan CCD, the Ronchigrams of $SrTiO_3$ (left) were acquired on a 20^2 STEM raster. The electric field (right) was calculated from the first moments employing Ehrenfests theorem. (c) Application to 2D-MoS_2 using the ultrafast pnCCD camera at 4kHz frame rate. The STEM raster was 256^2 pixels. Average monolayer (ML) and bilayer (BL) charge densities and electric fields are shown on the right.