Momentum-resolved STEM measurement of atomic electric fields, charge densities, polarisations and chemical composition

Müller-Caspary, K.[1,2,3], Rösner, M.[3], Duchamp, M.[4], Béché, A.[5], Winkler, F.[1], Krause, F.[3], Lobato, I.[5], Schowalter, M.[3], Migunov, V.[1], Simson, M.[6], Soltau, H.[6], Grieb, T.[3], Löffler, S.[7], Wehling, T.[3], Dunin-Borkowski, R.E.[1], Van Aert, S.[5], Verbeeck, J.[5], Schattschneider, P.[7], Zweck, J.[8] and Rosenauer, A.[3]

[1] Forschungszentrum Jülich, Germany, [2] University of Antwerp, Germany, [3] University of Bremen, Germany, [4] Nanyang Technical University, Singapore, [5] University of Antwerp, Belgium, [6] PN Detector GmbH, Germany, [7] Vienna University of Technology, Austria, [8] University of Regensburg, Germany

Momentum-resolved Scanning Transmission Electron Microscopy (STEM) combines the established bright-/dark field and differential phase contrast (DPC) STEM approaches in one single acquisition while enabling conceptually new insights into the physical properties of specimen. In particular, both high spatial and momentum resolution are obtained by scanning a focused probe over the specimen, and recording the detailed intensity distribution in diffraction space for each scan point. The capability to measure electric fields and charge densities down to the subatomic scale, and to perform angular multi-range analyses constitute the rising interest in momentum-resolved STEM, especially due to the recent enhancement of ultrafast cameras.

We first outline the theoretical framework for the measurement of electric fields from first moments calculated in diffraction space [1,2]. Using basic quantum mechanics, it is shown that the first moment is a robust measure of the average momentum transferred to the STEM probe, irrespective of the specimen thickness. Via Ehrenfest's theorem, the expectation values for the momentum transfer and the electric field in the specimen can be formally related. However, the direct quantitative interpretation of momentum transfers in terms of the electric field projected along electron beam direction is limited to specimen thicknesses below 5 nm due to multiple scattering and propagation.

This predestines momentum-resolved STEM to investigate 2D materials. In particular, we present a comprehensive experimental and simulation study for the atomic-scale electric field and charge density mapping across a mono-/bilayer boundary of 2D-MoS_2. In methodological respect, quantitative agreement between experiment, dynamical simulations taking partial coherence into account, and the direct interpretation is found. At present, conclusions on specimen tilt, stacking sequence and edge termination can be drawn from the charge density map whereas a mapping of Bader charges remains challenging due to noise limitations and is hence outlined in a simulation study based on density functional theory.

To extend the applicability of momentum-resolved STEM to thicker specimen containing built-in polarisation fields, we report different averaging schemes to separate the contribution of atomic electric fields from polarisation-induced ones that are orders of magnitude weaker. For spontaneous and piezoelectric fields in AlN/GaN, we first employ nano-beam electron diffraction where the position of the undiffracted beam and the relative distance between Bragg reflections yield electric field and strain simultaneously. Second, unit cell averaging is used in experimental atomically and momentum-resolved STEM data to eliminate atomic contributions. Systematic errors arising from dynamical scattering and mean inner potential gradients are quantified by simulations. For an AlN/GaN heterostructure, we find electric fields of 5.3±1 MV/cm.

From a broader perspective beyond electrical characterisation, we elucidate the potential of momentum-resolved STEM for the simultaneous compositional, thickness and strain mapping via an angular multi-range analysis [3] of GaNAs/GaAs heterostructures. A detailed investigation of the angular dependence of scattered intensity is given by comparing experiments with frozen phonon multislice simulations. Significant discrepancies are found in the low-angle regime whose origins are discussed in terms of inelastic scattering.

[1] Nature Communications 5, 56531 (2014).

[2] Ultramicroscopy 178, 62 (2018).

[3] Scientific Reports 6, 37146 (2016).

For the data that has been recorded on the direct electron detector at EMAT (University of Antwerp), support by the Hercules fund from the Flemish Government is gratefully acknowledged.
K. M.-C. acknowledges funding from the Initiative and Network Fund of the Helmholtz Association in the framework of the Helmholtz Young Investigator Group *more*STEM at Forschungszentrum Jülich, Germany.

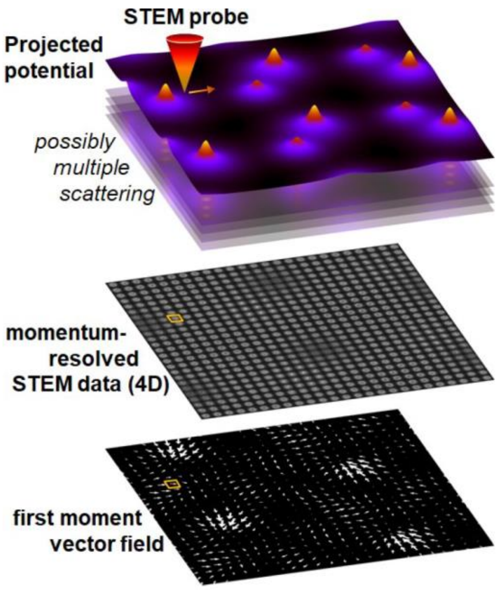

Fig. 1 Concept of momentum-resolved STEM. A focused probe is scanned over the specimen, and a diffraction pattern is recorded at each scan point

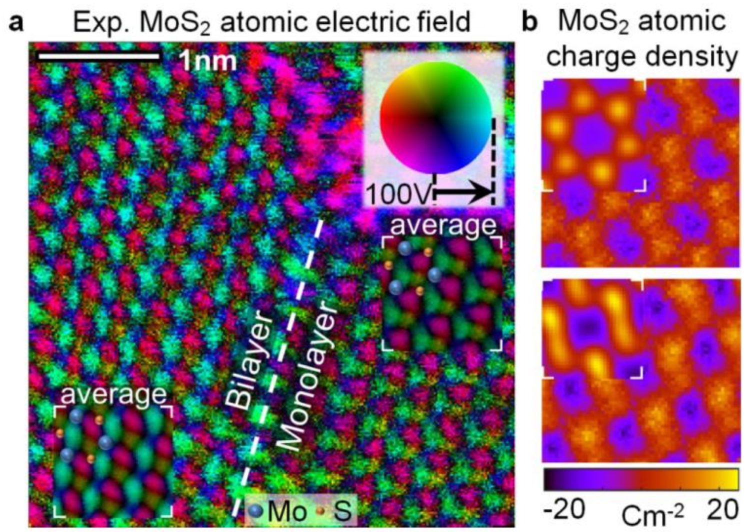

Fig. 2 In thin specimen, the momentum transfer is proportional to the projected electric field, convolved with the probe intensity. This example shows (a) the electric field in an MoS_2 mono-/bilayer region, (b) unit cell averages and simulation of the charge density calculated from (a).

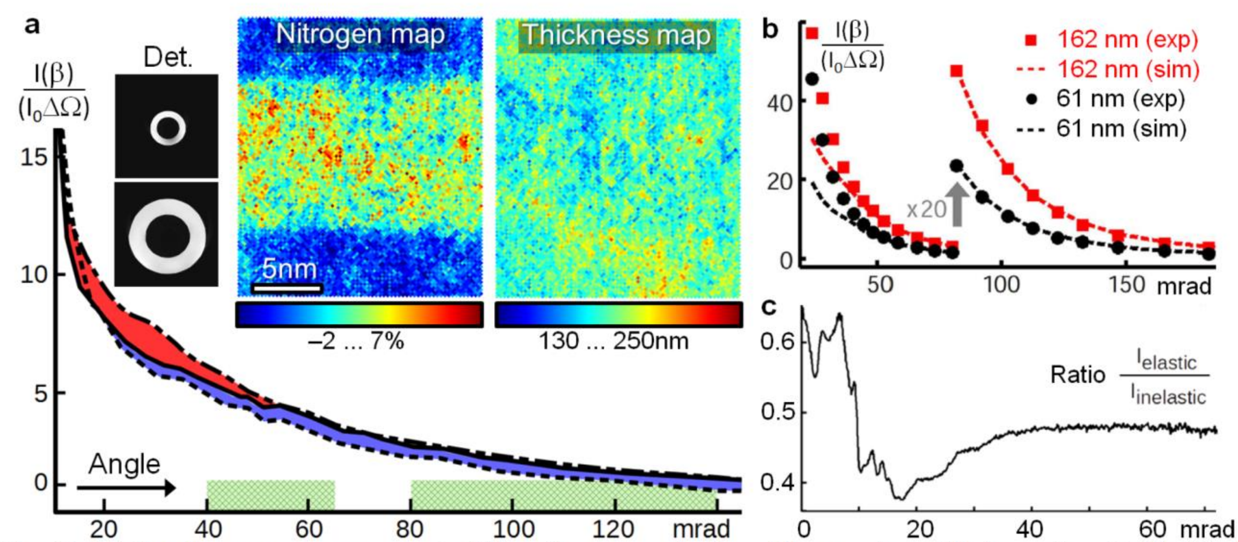

Fig. 3 (a) Simultaneous measurement of the nitrogen content and the specimen thickness in a GaNAs/GaAs heterostructure using 2 STEM images with dedicated detector angles, sensitive to composition (red shading) and thickness (blue shading). (b) Comparison of the angle-resolved scattered intensity in experiment (dots) and simulation (dashed) for Si, showing a mismatch at low angles. (c) Measured ratio of the angle-resolved scattered intensity using zero-loss-filtering and the inelastic scattering.