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Quantitative comparison of momentum-resolved STEM and off-axis electron holography at atomic resolution

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Off-axis electron holography (OAEH) is an established technique that can be used to record an electron wavefunction in a transmission electron microscope (TEM) with high spatial resolution. For a very thin specimen, the phase of the wavefunction can be related directly to the electrostatic potential of the specimen (1). However, OAEH puts high demand on the stability of microscope and specimen in order to achieve high spatial and signal resolution. Another promising technique, which is referred to as momentum-resolved scanning TEM (MR-STEM), is increasingly used to measure electrostatic fields in materials at atomic resolution. The latter technique involves recording diffraction patterns at each scan position and relating the first moment of the intensity distribution to the projected electrostatic field, according to Ehrenfest"s theorem (2). Both techniques are sensitive to electrostatic potentials or fields respectively and offer prospects for the quantitative measurement of fundamental material properties with atomic spatial resolution. The aim of the present work is to investigate experimental conditions of the two techniques producing similar or even equivalent results. This may enable us to benefit from particular advantages offered by a technique in aspects such as better dose efficiency, reduced irradiation damage, relaxed experimental limitations, or lower requirements for instrumental quality and stability. We present a case study for a thin $SrTiO_3$ crystal, where the signal can be related directly to the electrostatic potential or field. The different illumination setups that are required for the two techniques, i.e. parallel illumination in OAEH and convergent illumination for MR-STEM, are found to lead to different responses of the recorded signal to dynamical diffraction. We study, under which conditions, a quantitative measurement of these quantities is achievable with the two techniques and how experimental parameters, such as defocus, can be optimized for a straightforward interpretation of the recorded signal (c.f. Fig. 1). We further show that thermal diffuse scattering (TDS) is negligible even for thicker specimens. Finally, we discuss practical aspects of the applicability of each technique.

Fig.1: Reconstructed electrostatic potentials (divided by specimen thickness) obtained from MR-STEM and OAEH simulations for $SrTiO_3$. Each of the first 3 columns corresponds to a different specimen thickness. The defocus is labelled in the top right corner of each image. The signal is observed to change significantly with specimen thickness for each setup. The scale bar is 0.1 nm. The right hand column shows reference electrostatic potential distributions calculated from neutral atomic scattering factors for both approaches, including the influence of Debye-Waller factors and probe-forming (imaging) apertures.

References:

(1) S. Borghardt et al., *Physical Review Letters* 118 (8), 086101

(2) K. Müller et al., *Nature communications* 5, 5653

Fig. 1