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Optimization of orbital mapping: the quest for the perfect image

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Electronic states are responsible for most of the electrical, magnetic and optical properties of materials. Thus, a complete understanding of the electronic states is crucial for improving and developing new (opto-)electronic constituents, especially when interfaces and heterostructures are considered. However, a complete understanding can only be gained by imaging the relevant states in real space.

In the last years, it has repeatedly been shown that mapping transitions to individual electronic states for bulk samples is indeed possible using STEM-EELS [1,2]. Probe electrons that inelastically interact with sample electrons can trigger a transition from an initial core state to the conduction band. Information about the transition to a specific final electronic state is contained in the EELS fine structure and can, thus, be mapped when a suitably small energy window is chosen. However, this so-called orbital mapping pushes even modern transmission electron microscopes to their limits. The demanding needs for high spatial resolution and stability, together with the tiny energy windows of the spectrometer, result in a low signal-to-noise ratio and strongly limit the everyday use of this method. Thus, careful planning of experiments is of utmost importance to obtain usable data.

We have performed extensive simulations of a transition metal system (rutile), a system comprised of a light element (graphite), as well as a perovskitic interface (STO-LMO). The simulations are based on a multi-slice algorithm for the elastic propagation combined with the mixed dynamic form factor formalism with density functional theory (DFT) data [3]. With the use of an image difference metric, specially adapted to TEM images [4], we strive to fully investigate the chosen systems. By comparing the images to a best-case reference image, we find ranges of parameters (high voltage, sample thickness, image dose, ...) that would result in the optimal experimental images. However, this method is only as good as the reference image. Thus, it is an ongoing work in progress to continually improve the ab-initio simulations. The next step is to incorporate excitonic effects into the underlying DFT data [5] which would further improve the simulated reference image. In the long run this will bring us ever closer to the goal of an easier and broader applicability of orbital mapping. [6]

1 Löffler et al., Ultramicroscopy, 26 (2017), p. 177.
2 Bugnet et al., Phys. Rev. Lett. 128 (2022), p. 116401.
3 Löffler et al., Ultramicroscopy, 131 (2013), p. 39.
4 Ederer et al., Ultramicroscopy, 240 (2022), p. 112578.
5 Laskowski et al., Phys. Rev. B, 82 (2010), p. 205104.
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Figure 1. (a), (b) Simulated STEM-EELS maps of rutile at 539 eV energy loss for a sample thickness of 0.3 nm and 20 nm, respectively. Circles mark the positions of titanium (green) and oxygen (orange) atoms. (c) Image difference heat map of rutile orbital maps as a function of the sample thickness and incident dose. Image (a) serves as the reference image.