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IT-5-IN-1534 Can orbitals be mapped in the TEM?

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The energy, position, and momentum distributions of electrons inside a material are decisive for most of the material's properties, ranging from optical, electrical and magnetic properties to hardness, durability, or the melting point. Therefore, the electron distribution is a key quantity in many fields of research. Unfortunately, it is also elusive and directly imaging electronic orbitals and bonds in the bulk has not been possible so far.

In the last few years, several authors reported measurements of orbital properties using electron energy loss spectrometry (EELS) [1-3]. While these are great advances, it would be even better to actually "see" the orbitals in an image. Recently, the possibility to record maps of transition probabilities – from which orbitals can be deduced – was predicted theoretically [4]. In this work, we investigate the requirements and the feasibility to realize that prediction in an actual experiment.

On the one hand, the point group symmetry of the sample atoms plays a crucial role. Intuitively, this is readily understandable. Taking an isolated atom, for example, one is faced with a spherically symmetric problem. Clearly, its solutions must produce rotationally symmetric images. Hence, the symmetry of the system has an important influence on orbital maps. Here, we investigate the requirements on the crystal structure in order to be able to see a directional dependence of transition probabilities, orbitals, and bonds (see Fig. 1).

On the other hand, experimental parameters such as the signal to noise ratio (where the signal is the difference from the average; see Fig. 2), as well as the stability of the specimen and of the microscope are vital for successfully recording high-resolution maps. Based on experimental energy filtered images recorded with very high spatial resolution, we evaluate the requirements on both the sample and the microscope to obtain reproducible and directly interpretable maps. This nurtures the hope that orbital mapping will become a reality in the near future and will become an invaluable tool for many fields of research.

[1] Löffler et al., Ultramicroscopy 111 (2011) 1163
[2] Neish et al., PRB 88 (2013) 115120
[3] Hetaba et al., Micron, in print
[4] Löffler et al., Ultramicroscopy 131 (2013) 39

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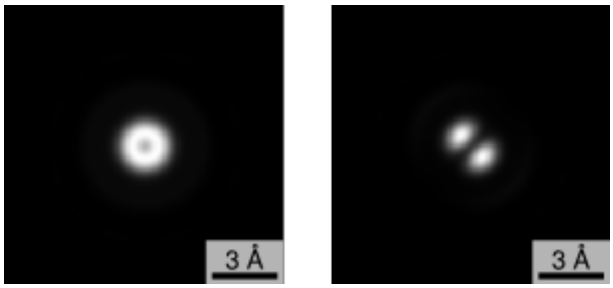

Fig. 1: Comparison of the K-edge maps for an Oxygen atom with full O(3) point-group symmetry (left) and with C_{2v} symmetry (right). For the simulations, an acceleration voltage of 80 kV, a collection semi-angle of 24 mrad and ideal imaging conditions (C_s=0, C_c=0, df=0) were assumed.

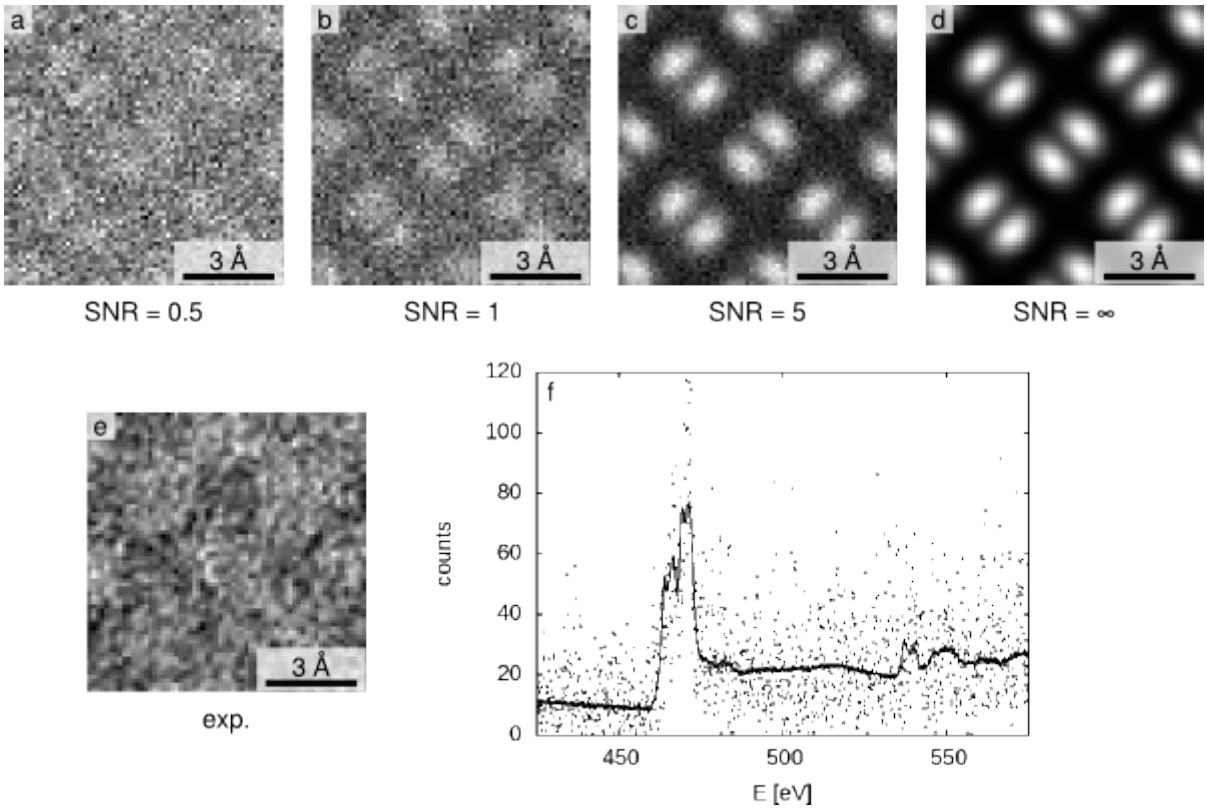

Fig. 2: Predicted maps of the Ti L edge for a thin Rutile sample in [001] zone axis for different signal-to-noise ratios (SNR) as indicated (a-d). Preliminary experimental map as acquired (e). EELS signal of 1 px (dots) and averaged over 14000 px (line) (f). An acceleration voltage of 80 kV and an energy-window of 4 eV on the L_2 edge were used.