Spectroscopic STEM imaging in 2D and 3D

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Atomic-resolution imaging with a spherical aberration-corrected scanning transmission electron microscope (STEM) is now widely used for the study of interesting, complex material systems. This is owed both to the flexibility in detecting the electrons scattered off from matter, but also to the improved efficiency in collecting spectroscopic signals. High-angle annular dark-field imaging is routine and has become a quantitative technique [1]. Developments like high-brightness guns and monochromators, improved electron energy-loss spectrometers (EELS) or X-ray (EDXS) detector concepts, have enabled elemental-specific imaging at that scale, and allow for a correlation of structural and chemical information [2,3,4]. The simultaneous collection of all these signals at various tilt angles even, defining the technique of STEM-EELS/EDXS tomography, has given unprecedented insight into the 2D and 3D structural and physico-chemical make-up of TEM specimens [5,6,7,8]. While the principal acquisition of large multi-modal data sets has become more straightforward, the data processing and the interpretation of the spectroscopic intensities now appear rather challenging.

For 2D, classical compositional analysis with core-loss EELS and EDXS signals, when executed accurately, still suffers from a multitude of ill-defined parameters and intensity conversions into concentrations for a particular quantification scheme often rely on “best-guess” quantities, such as sample densities, absolute thicknesses, theoretical ionization cross-sections, solid and take-off angles etc [9]. A recently described analysis concept tries connecting EDX and EELS signals in a common analysis framework helping to reduce the need for estimates [10, 11]. Secondly, the complex physics of scattering of the electron probe along aligned atomic columns produces a nonlinear relation between signal and composition and there is no longer a simple relationship between the observed analytical intensities from the projected atomic positions. Different approaches to recover the true concentrations from inelastic images, taken under strong channeling conditions, have been proposed [12,13,14]. Often image simulations and a calculation of the underlying scattering dynamics are required to “correct” the experiment. Alternatively, acquisition schemes to reduce the effects of channeling by tilting or precessing the beam [15] have been suggested.

When going to 3D tomography, linking two or more signals in the reconstruction is even more challenging. By introducing different variational norms and penalty terms for correlated reconstructions, our recently introduced total generalized variation (TGV) approach seems to deliver the most faithful 3D chemical phase reconstructions from unsharp interfaces [16]. Also for monochromated low-loss EELS tomography, some of the early issues in mapping and interpreting the evanescent fields of particle plasmons could be overcome [17,18,19,20,21], and recently, a novel tomography scheme lifted some of the restrictive conditions to compute the photonic local density of states, an important quantity in nano-optics [22,23,24].

Overall, the STEM represents a quantitative instrument which is capable of providing numerical data on the properties of thin specimens. The talk aims to give an overview to spectroscopic imaging in 2D and 3D and its challenges in the mentioned areas.

References

[1] J. M. LeBeau et al., Phys. Rev. Lett. 100 (2008) 206101
[2] D. A. Muller et al., Science 319 (2008) 1073
[3] H.S. von Harrach et al., J. of Physics: Conference Series 241 (2010) 012015
[4] A. Gubbens et al., Ultramicroscopy 110 (2010) 962
[5] K. Jarausch et al., Ultramicroscopy 109(4) (2009) 326
[6] S. Bals et al., MRS Bulletin 41(7) (2016) 525
[7] G. Haberfehlner et al., Nanoscale 6 (2014) 14563
[8] G. Haberfehlner et al., Nature Communications 6 (2015) 8779
[9] J. Kraxner et al., Ultramicroscopy 172 (2017) 30
[10] G. Kothleitner et al., Micro. Microanal., 20/3 (2014) 678
[11] L. Konrad et al., Micron 115 (2018) 18
[12] L. J. Allen et al., Ultramicroscopy 151 (2015) 11
[13] B. D. Forbes et al., Phys. Rev. B 82 (2010) 104103
[14] G. Kothleitner et al., Phys. Rev. Lett. 112 (2014) 085501
[15] N. R. Lugg et al., Ultramicroscopy 151 (2015) 150
[16] R. Huber et al., Nanoscale 11 (2019) 5617
[17] M. Bosman et al., Nanotechnology 18(16) (2007) 165505
[18] J. Nelayah et al., Nature Physics 3(5) (2007) 348
[19] B. Schaffer et al., Physical Review B 79(4) (2009) 041401
[20] D. Rossouw et al., Nano Lett. 11(4) (2011) 1499
[21] B. Ögüt et al., ACS Nano 5 (8) (2011) 6701
[22] F. J. García de Abajo et al., Phys. Rev. Lett. 100(10) (2008) 106804
[23] A. Hörl et al., ACS Photonics 2 (2015) 1429
[24] A. Hörl et al., Nature Communications 8 (2017) 37

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