Making the most of your multidimensional EELS data

F. de la Peña[1,*], T. Ostasevicius[1], R. Arenal[2], R.K. Leary[1], O. Nicoletti[1], P.A. Midgley[1], C. Ducati[1]

[1] Department of Materials Science & Metallurgy - University of Cambridge
Cambridge - United Kingdom.
[2] Instituto de Nanociencia de Aragon (INA) – Universidad de Zaragoza
Zaragoza - Spain
[*] fjd29@cam.ac.uk

Recent technological breakthroughs, such as fully computer-controlled electron microscopes with the ability to acquire multiple signals simultaneously, automatic acquisition of tilt series, aberration correctors and monochromators, have dramatically increased the level, rate and precision at which we can acquire electron energy-loss spectra in an electron microscope. Therefore, as in many other fields, the TEM-EELS community is facing the challenge of how to better analyse large quantities of data extending over multiple dimensions.
In some cases, analytical procedures developed for single-spectrum analysis can easily be extended to multi-dimensional datasets by performing the analysis individually on each signal in the set. However, this is not always possible as such procedures frequently require input parameters that are different for each element of the set. Moreover, for optimal usage of this usually highly redundant information, data analysis methods from the machine learning domain are becoming increasingly useful in the solution of problems that would be highly challenging, if not impossible, with more conventional methods [1].
For example, the curve fitting method for elemental quantification of EELS spectra [2, 3] can be extended to multi-dimensional datasets by performing parameter optimization at each coordinate. Indeed, with the advent of EELS spectrometers that can quasi-simultaneously acquire low-loss and core-loss spectra [4, 5] this method should become increasingly popular. However, non-linear regression methods are very sensitive to the initial parameters and, therefore, a simplistic extension to multi-dimensional datasets usually requires setting starting parameters for the whole dataset [6], which is not only inconvenient, but also challenging. Here we show that it is possible to turn the burden to our advantage by designing strategies to automatically generate starting parameters. This method, in conjunction with parallelization of the fitting routine, can considerably ease the task of analysing very large datasets e.g. tomographic EELS tilt-series.
Alternatively, one could use machine learning methods to learn the model from the dataset. These methods are especially useful when an accurate model of the signal is not available. For example, we have recently demonstrated an application of non-negative matrix factorization (NMF) [7] to the decomposition of a low-loss tilt-series in components related to surface plasmon modes of a silver nanocube [8]. In principle, a similar methodology could be applied to bonding analysis. However, while available methods can be useful to analyse very thin samples, in general, for thicker samples, they fail because multiple scattering renders the problem non-linear [1]. Here we show how a combination of curve fitting, compressed sensing [9] and machine learning methods can ease the path towards bonding tomography.
In the spirit of reproducible research, we make all of our methods available by contributing them to the open-source software HyperSpy [11].

Références

[1] F. de la Peña, M.-H. Berger, J.-F. Hochepied, F. Dynys, O. Stephan, and M. Walls, Ultramicroscopy, **111** (2011), p.169.
[2] T. Manoubi, M. Tence, M. G. Walls, and C. Colliex, Microsc. Microanal. Microstruct., **1** (1990), 23.
[3] J. Verbeeck and S. V. Aert, Ultramicroscopy, **101** (2004), 207.
[4] H. Pinna and M. Tence, "Device for Obtaining the Image and/or Spectra of Electron Energy Loss," FR2874124A1.
[5] A. Gubbens, M. Barfels, C. Trevor, R. Twesten, P. Mooney, P. Thomas, N. Menon, B. Kraus, C. Mao, and B. McGinn, Ultramicroscopy, **110.8** (2010).
[6] F. de la Peña, N. Barrett, L. F. Zagonel, M. Walls, and O. Renault, Surf. Sci., **604** (2010), 1628.
[7] D. D. Lee and H. S. Seung, Nature, **401** (1999), 788.
[8] O. Nicoletti, F. de la Peña, R. K. Leary, D. J. Holland, C. Ducati, and P. A. Midgley, Nature, **502**, (2013), 80.
[9] R. Leary, Z. Saghi, P.A. Midgley, D.J. Holland, Ultramicroscopy **131** (2013), 70.
[11] http://hyperspy.org