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IT-5-P-6028 Addressing challenges in Electron Energy Loss Spectroscopy on individual atoms

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The latest generation of STEM microscopes based on many instrumental developments (Cs corrector, lower primary voltages, EELS and EDX detector improvements...) offers the ability to track in a spectrum-image mode several signals generated simultaneously by individual atoms [1,2,3] and to rekindle the STEM-EELS spectro-microscopy of single atoms.

A Nion UltraSTEM microscope equipped with a Cs corrector and with a home-made fast EELS detector has been used to record a few typical cases illustrating the present situation in individual atom spectroscopy. With the new spectroscopic hardware, we can acquire EELS spectrum images of typically 100x100 pixels and covering a range of 1600 channels at an acquisition rate of 2300 spectra/s. Furthermore, the representation, exploitation and analysis of such data require some specific algorithms. The most widely used technique is the Principal Component Analysis (PCA) [4][5] and, as a filtering technique, offers an improvement of the signal to noise ratio. However, for high noise levels, a bias is introduced by PCA as signal bearing components are discarded with the removal of components considered as noise [6]. We have tested some algorithms based on non-local methods for denoising by exploiting the natural redundancy of patterns inside an image.

The first case is the determination of the position of Sm interstitial/substitutional dopants in ceria nanoparticles together with their valence changes in accordance with the variation of the ferromagnetic properties measured as a function of the nominal doping level [7]. The spectrum image has a high noise level and Sm doping could not be identified with usual PCA denoising. We have therefore tested Non-Local Sparse PCA [8] which produces interesting results: the filtered spectra display fine structures of edges and both spatial and spectral resolutions are preserved. The second example addresses the challenge of identifying the characteristic EELS signals from heavy (Tb, Th) atoms in rapid motion on a thin carbon layer which imposes a compromise between time acquisition and detection limit (see Figure).

This contribution emphasizes the possibilities currently offered by a tiny electron probe, a suitable efficient detector strategy and a well chosen signal analysis tool for single atom spectroscopy.

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[7] S.-Y. Chen et al. Phys.Chem.Chem.Phys. 16 (2014) 3274.

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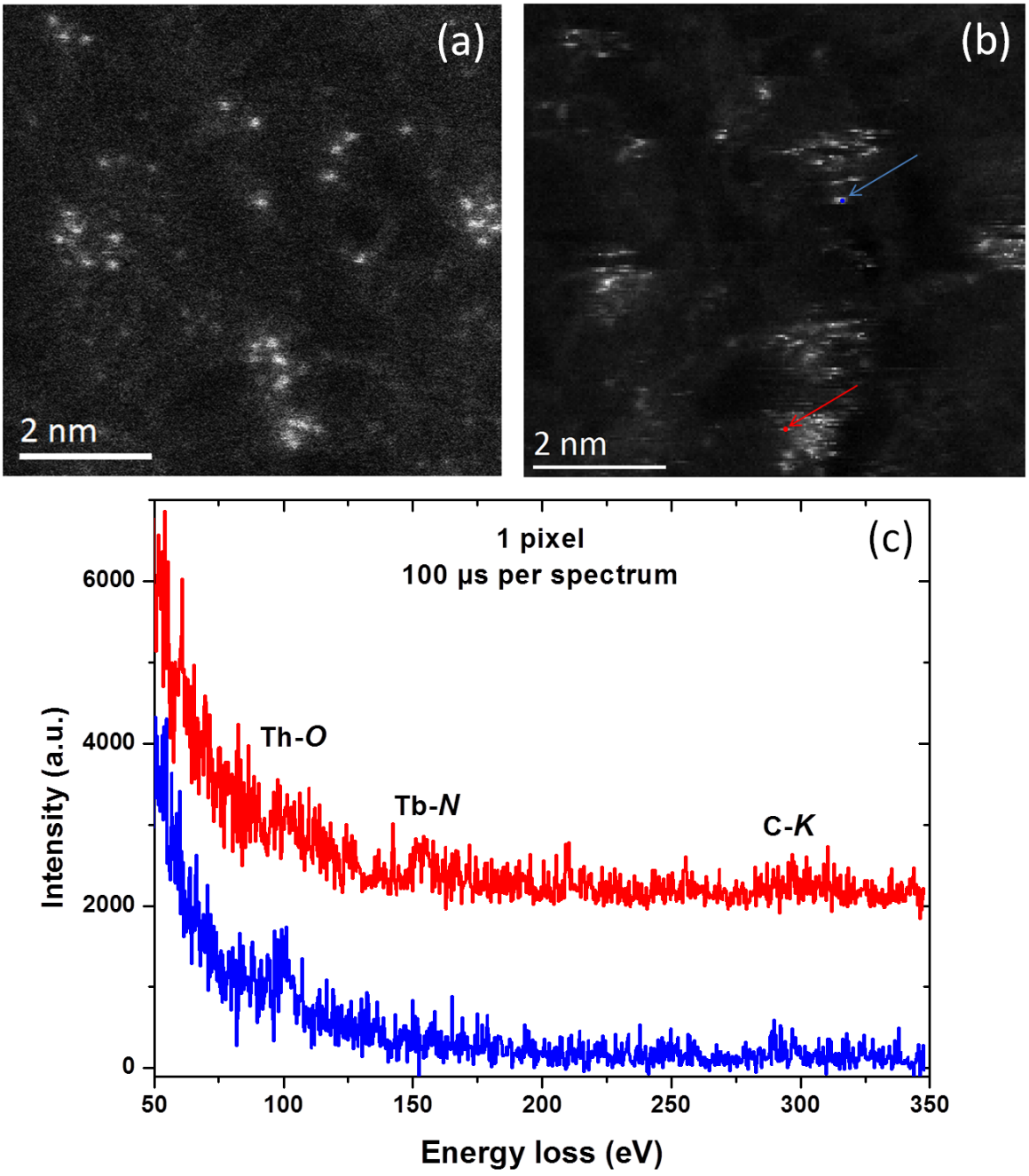

Fig. 1: Imaging and spectroscopy of Th and Tb atoms in rapid motion on a thin carbon foil under the electron beam (60 kV). HAADF images at 2 µs per pixel (a) and at 100 µs per pixel (b). Raw EELS spectra extracted from the SI at two different positions (blue and red) – acquisition time: 100 µs per spectrum (c).