## Combined imaging and analytical STEM of ultra-thin cuprate films

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Considerable advances in scanning transmission electron microscopy (STEM) have revealed fascinating phenomena in functional complex oxide materials at the atomic scale. Simultaneous acquisition of annular bright-field (ABF) and high-angle annular dark-field (HAADF) STEM images opened up an elegant path for combined imaging of light and heavy elements. Furthermore, combining STEM imaging with advanced analytical methods, such as electron energy-loss spectroscopy (EELS) and energy-dispersive X-ray spectroscopy (EDX), sets an exceptionally powerful approach to study very thin films.

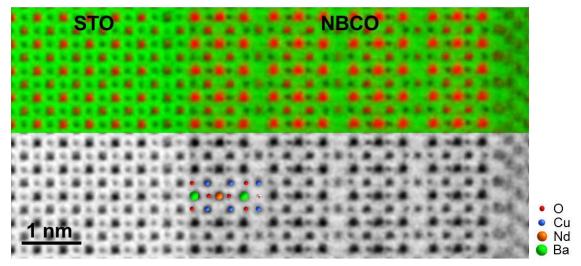
In this work, high quality NdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (NBCO) ultra-thin films (1, 2 and 4 unit cells) have been deposited on TiO<sub>2</sub>-terminated SrTiO<sub>3</sub> (STO) substrates by high oxygen pressure diode sputtering [1]. The layer stacking sequence along the growth direction parallel to the c-axis of NBCO is BaO-CuO<sub>2</sub>-Nd-CuO<sub>2</sub>-BaO-CuO. In NBCO, the Cu atoms are located both in the CuO<sub>2</sub> planes and in the CuO chains that run parallel to the planes and structurally separate the superconducting CuO<sub>2</sub> bilayers. Nd atoms are placed within each bilayer in-between the two CuO<sub>2</sub> planes. Ba atoms are located between the planes and the chains. Major changes of the NBCO's physical properties can appear due to oxygen content fluctuations [2]. For non-stoichiometric NdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub>, x denotes the amount of vacancies present in the CuO chains. The charge balance between the CuO<sub>2</sub> planes and CuO chains affects and effectively dictates  $T_c$  [3].

We have employed atomic-column resolved quantitative STEM imaging combined with forefront analytical techniques to investigate the ultra-thin NBCO films by using an advanced aberration-corrected JEOL JEM-ARM200F microscope equipped with a DCOR probe corrector operated at 200 kV. Simultaneously acquired and overlaid HAADF- and ABF-STEM images of a selected NBCO ultra-thin film on STO (top) and an ABF-STEM image with the superimposed NBCO structural model (bottom) are shown in Figure 1. The improvement of the signal-to-noise ratio as well as the reduction of the image distortions was achieved by multiple frame acquisition [4]. Using such approach, we were able to quantitatively analyse the local cation and anion sub-lattices. In addition, we have performed STEM image simulations in order to correlate the lowest detectable oxygen concentrations with the sample thickness.

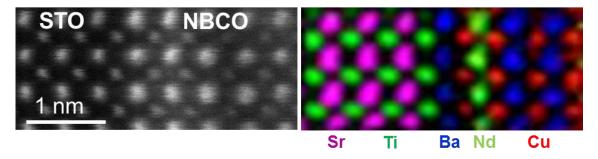
The chemical identity of ultra-thin NBCO films and their interfaces with the STO substrates were studied by combining atomically resolved ADF-STEM imaging with EELS and EDX mapping. An EELS elemental map obtained from a representative NBCO/STO interface is shown in Figure 2. Chemical maps have confirmed that the STO substrate is TiO<sub>2</sub> terminated. Furthermore, surface steps detected on the STO substrate and consequent defects in NBCO films were studied and will be discussed [5].

## References:

- [1] M Salluzzo et al., Phys rev B 78 (2008), 054524.
- [2] H Shaked et al., Phys Rev B 41 (1990), p. 4173.
- [3] RJ Cava et al., Physica C 165 (1990), p. 419.
- [4] Y Wang et al., Ultramicroscopy 168 (2016), p. 46.
- [5] This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 823717 ESTEEM3.



**Figure 1.** Overlay of simultaneously acquired HAADF- (red) and ABF- (green) STEM images (top) of a NBCO ultra-thin film (4 unit cells) grown on STO and ABF-STEM image (bottom) with an overlay of the NBCO structural model.



**Figure 2.** ADF-STEM image of the NBCO/STO interface with a corresponding atomically resolved EELS elemental map.