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Surface islands in epitaxial Li_xCoO_2 films studied by LEEM/PEEM for different lithium concentrations

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Li_xCoO_2 (LCO) and related intercalation oxides are extensively used in Li-ion batteries as cathode materials and will play a key role in the near future in neuromorphic computing. Most technologically relevant properties of LCO are related to its ability to change its composition from stoichiometric $LiCoO_2$ to Li_xCoO_2, a process that modifies its electronic structure. Epitaxial thin films of Li_xCoO_2 grown on $SrTiO_3$ (STO) were studied at the Nanospectroscopy beamline of Elettra storage ring using Photoemission Electron Microscopy (PEEM) and Low Energy Electron Microscopy (LEEM).

The experiments provided a detailed classification of the sample grains and allowed the identification of islands and wetting layer. In order to understand the electronic and structural changes during lithium deintercalation, the stoichiometric LCO islands were delithiated in situ using preferential Ne sputtering, followed by a thermal treatment to maintain a good surface quality. The pure LCO islands were monitored and the effects of the delithiation on the island size, distribution and surrounding substrate have been characterized. Low Energy Electron Diffraction (LEED) provides a good indicator of the possible deterioration, establishing that the sample surface maintains a good long-range coherence after delithiation, also observed in the Low Energy Electron Microscopy (LEEM) images. The delithiation induces a shift of the valence band position towards the Fermi energy, changes in the Co 3p and Co 2p core level line shapes and in the Co L absorption edge. The changes were monitored after each delithiation cycle and are compared to previous X-ray photoemission measurements on similar samples. The results conclusively indicate a metallization of the sample due to hole doping in the Co t_{2g} levels.

The spatial resolution of PEEM provides further and more detailed information about the behavior of the individual islands of LCO during the metallization. This technique also provides a way to isolate the contribution of the islands from the wetting layer formed between them, which has been observed to contain Li and Co. Changes in the mechanism of lithium deintercalation related to island size and density are also observed and will be discussed.

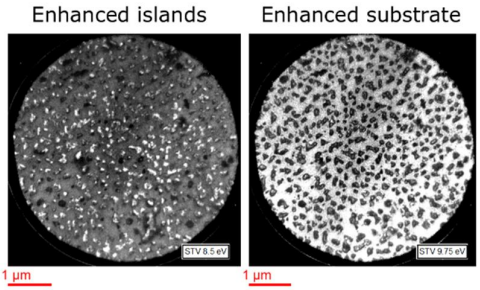

Figure 1. Island identification using PEEM images for different start voltages.