Supplementary Information

Experimental and simulated SAED patterns of $Li_7La_3Zr_2O_{12}$ in [001] viewing direction are shown in Fig. SX1. There are almost no differences in the diffracted intensities for the SAED patterns. The differences in the "a" parameter between cubic and tetragonal $Li_7La_3Zr_2O_{12}$ amount to only 1.5% which is the accuracy limit of the SAED measurements. Thus, the distinction between two modifications is impossible only from one SAED image.

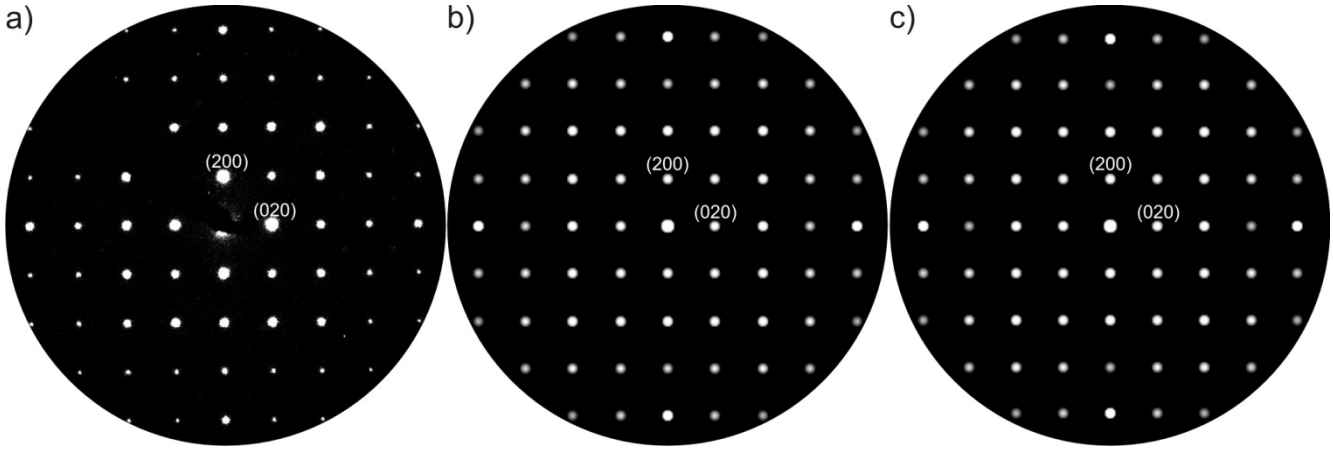

Fig. SX1 (a) Experimental SAED pattern of cubic $Li_xLa_3Zr_2O_{12}$ and simulated SAED patterns of (b) cubic $Li_xLa_3Zr_2O_{12}$ and (c) tetragonal $Li_7La_3Zr_2O_{12}$ along [001] viewing direction.

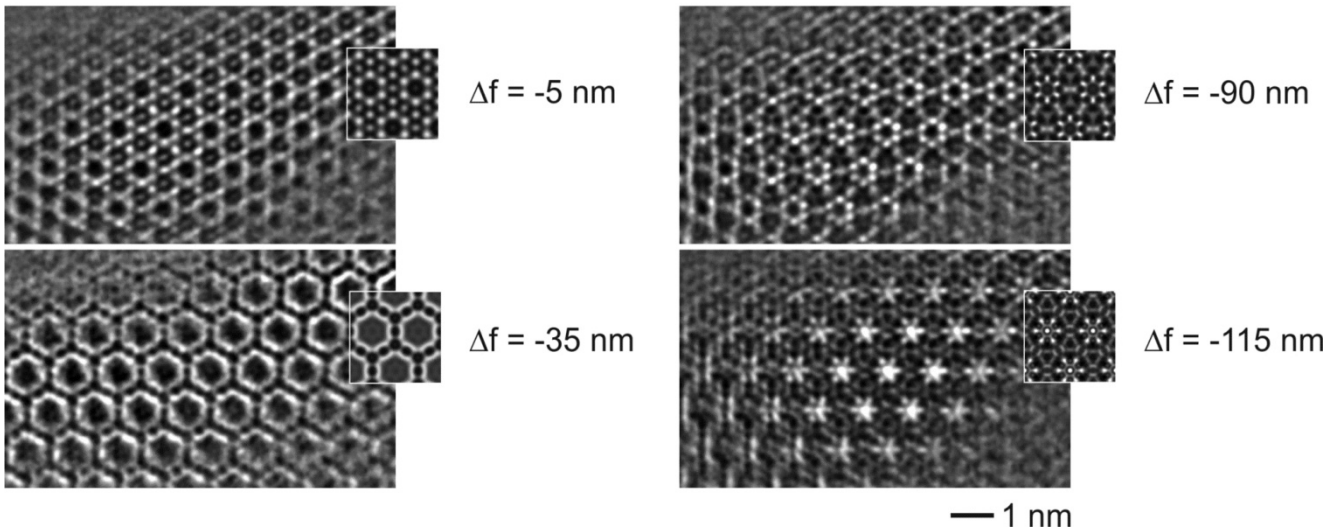

Fig. SX2 Complete focal series for $Li_7La_3Zr_2O_{12}$ with inserted simulated micrographs (zone axis [111], t = 6.7 nm). Focus values specified in the figure.

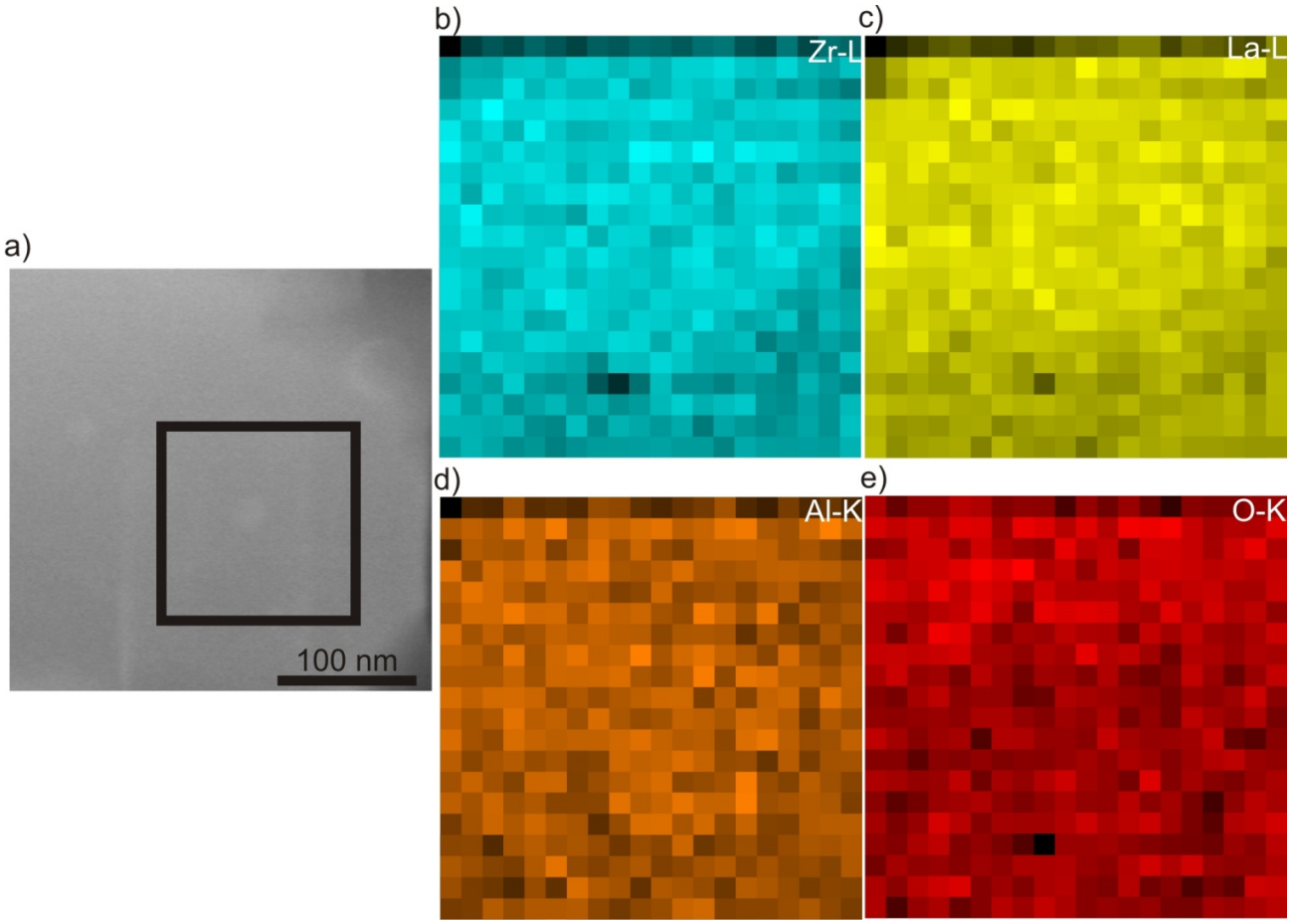

Fig. SX3 (a) STEM-HAADF image and (b)-(e) STEM EDX elemental maps of LLZO sample containing 0.9 wt.% of Al.