Supplementary Information

Table S1. The La/Ti and Sr/Ti atomic ratios as determined from XEDS analysis for the $La_{0.5-x}Li_{0.5-x}Sr_{2x}TiO_3$ compounds. Unit cell parameters have been calculated on the basis of a cubic cell, symmetry space group P*m-3m*.

Nominal Composition	La/Ti	Sr/Ti	a_c (Å)
$La_{0.437}Li_{0.437}Sr_{0.125}TiO_3$	0.423	0.127	3.8800 (1)
$La_{0.375}Li_{0.37}Sr_{0.25}TiO_3$	0.370	0.26	3.8857 (1)
$La_{0.35}Li_{0.35}Sr_{0.30}TiO_3$	0.351	0.31	3.8864 (1)
$La_{0.25}Li_{0.25}Sr_{0.50}TiO_3$	0.241	0.50	3.8930 (1)

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† *Electronic supplementary information (ESI) available*

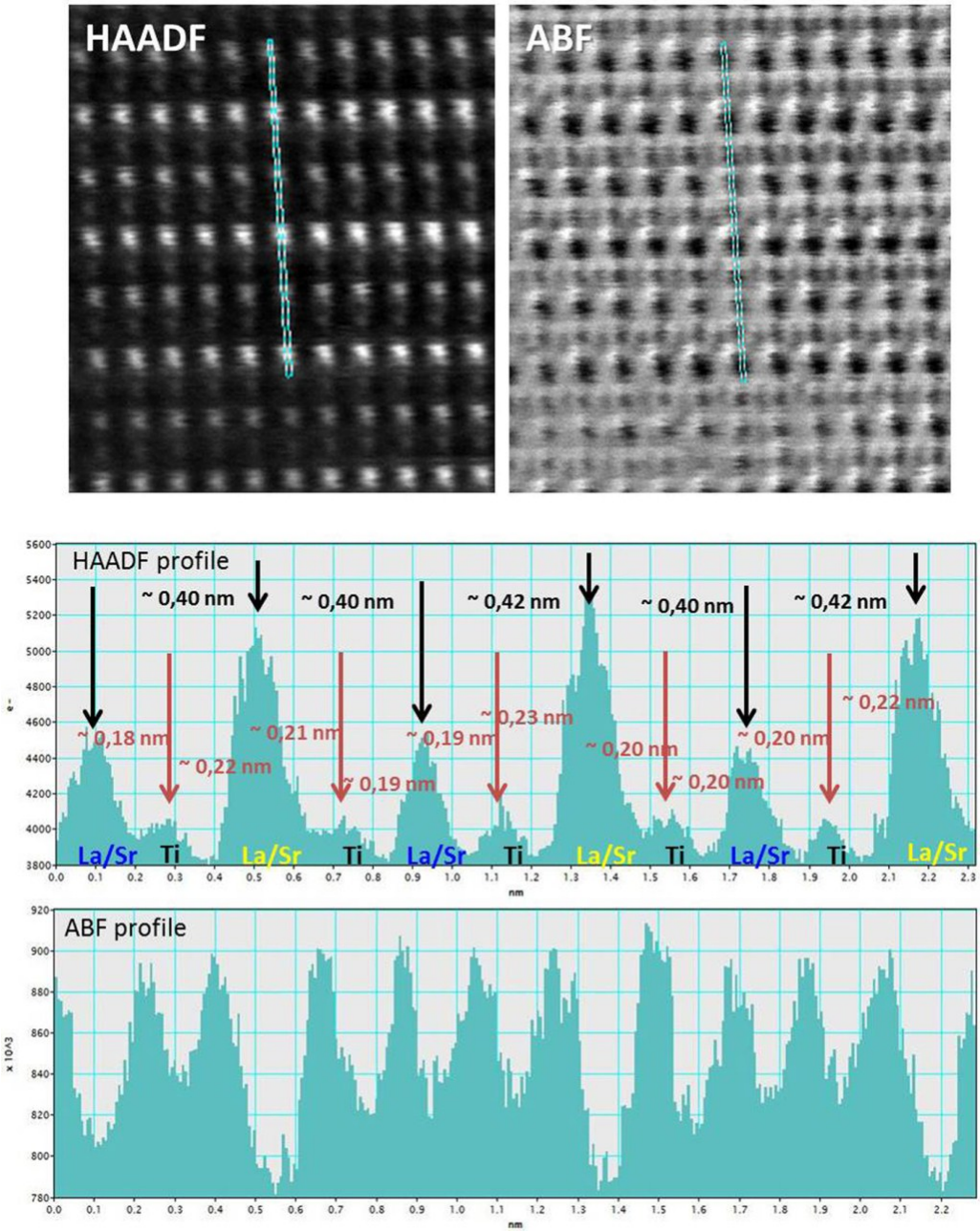

Figure S1. Simultaneous HAADF (left) and ABF (right) images of a region with $2a_c$-type contrast in a crystal of x=0.3 composition, observed along the $[110]_c$ zone axes . Corresponding intensity scan profiles obtained (down) along the selected lines. Distances between neighboring La/Sr and Nb columns demonstrate the displacement of Ti atoms towards La/Sr poor layers (labelled in blue).

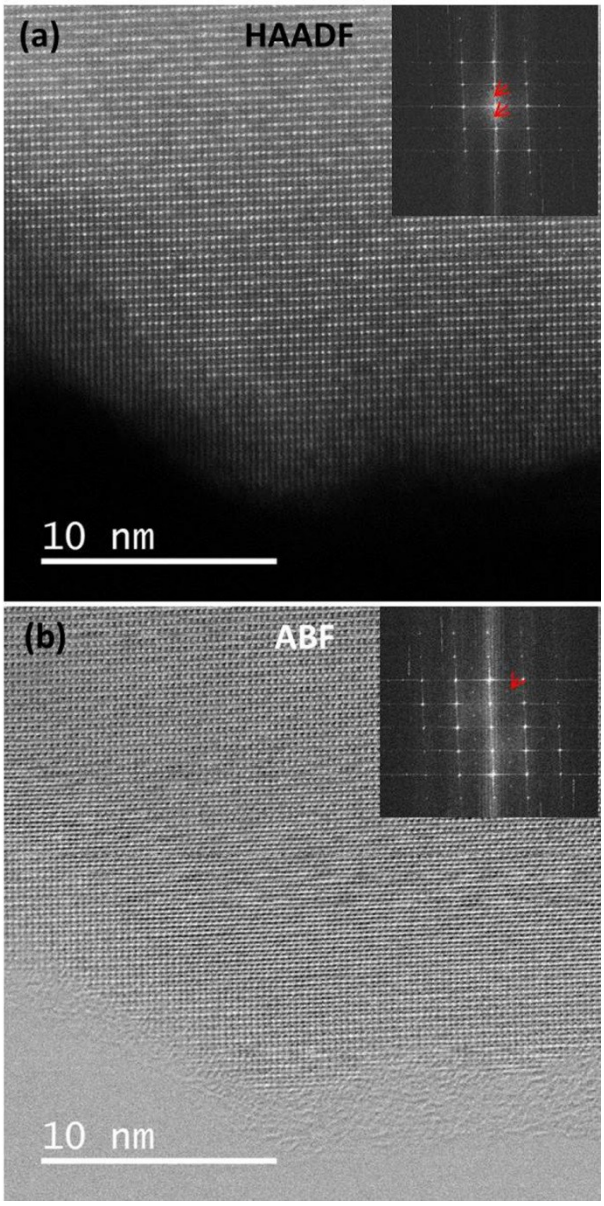

Figure S2. Low magnification HAADF (a) and ABF(b) images of the same area of a crystal of composition x=0.25 and their corresponding Fourier Transforms. FFT of (a) exhibits only extra diffraction maxima at ½<001> while those at ½<111> are not observed and can be only discerned in the FFT of (b).

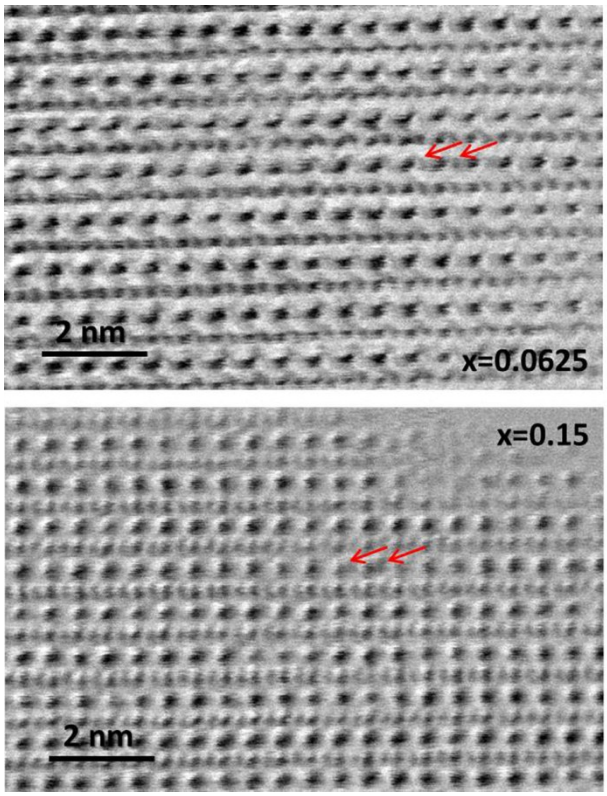

Figure S3. Details of ABF images of crystals of the x=0.0625 (up) and x=0.15 (down). Red arrows point image contrasts placed in the center of the O4 square window or slightly off center that suggests the presence of lithium.

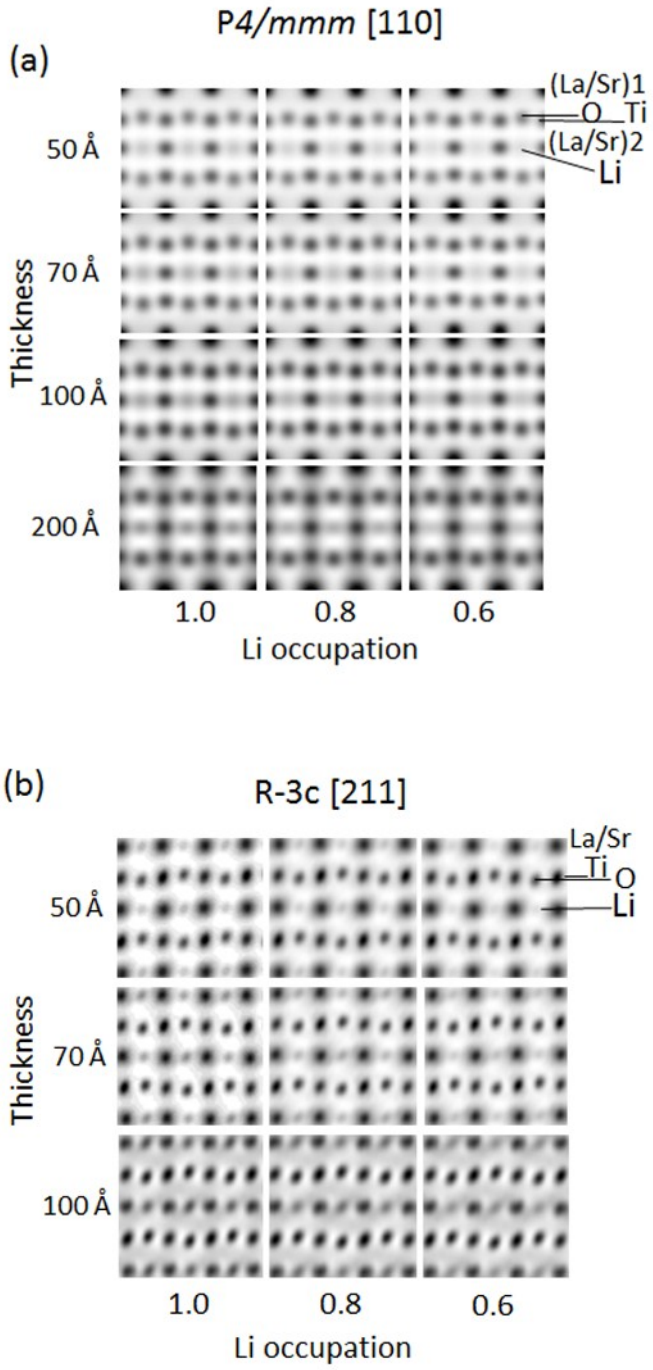

Figure S4. (a) Simulated ABF images corresponding to the P*4/mmm* cell along the [110] zone axis. (b) Simulated ABF images corresponding to the R$\bar{3}c$ cll along the [211] zone axis. Simulations have been performed by using the McTempas software package (version 2.4.25).