

Figure S1. EDS maps of individual elements in pristine/ON/OFF state samples, showing no diffusion of La (Ni) into the interlayer.



Figure S2. Correlative EDS-EELS O-K edge data shown in a representative region of the sample in ON state. Al rich regions (box/spectrum in the middle) correspond to weak O-K edge signal. A small pre-peak (spectrum on the right) is also observed, which is reminiscent of the pre-peak observed for a sample in OFF state (Fig. 2d), except that in the OFF state it is much more intense.



Figure S3. Comparison of the oxygen *K* edge signals, after background subtraction using a power law fit and normalization to the continuum at 650 eV. Data correspond to various values of t in the pristine sample. Plural scattering has been deconvolved using the zero-loss peak. The reduced overall intensities for spectra acquired near the interface (e.g. t=2, 4 nm – black and red spectra) when compared to spectra away from the interface (t=17 nm, pink) point to a lower oxygen content near the interface region. This finding strongly suggests that oxygen deficiency could be responsible for the reduction of Ni at the LNO-AlO_x interface.



Figure S4. Current-density vs. Voltage (J-V) hysteresis loops on another representative device. The pristine state transformed to an ON state at \sim +6V. Upon reversing the direction of voltage sweep, the ON state transformed to an OFF state at -4 V, which could be set to ON and reset back to OFF at +4 V and -4 V respectively.