

Defect Engineering and Anisotropic Modulation of Ionic Transport in Perovskite Solid Electrolyte $\text{Li}_x\text{La}_{(1-x)/3}\text{NbO}_3$

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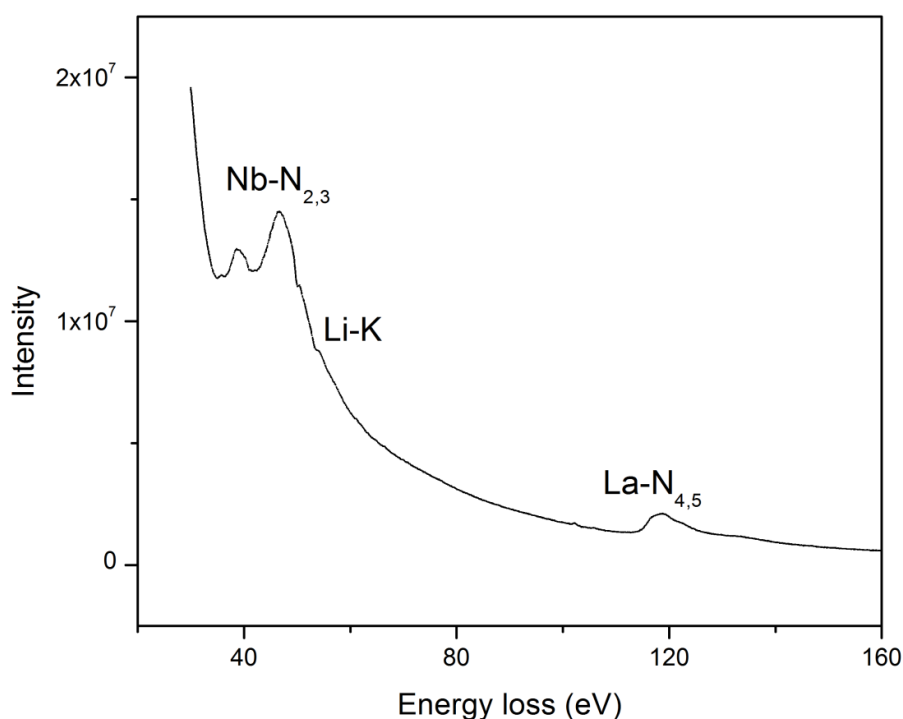


Figure S1. Core-level EELS of solid electrolyte LLNO to show the low content of Li.

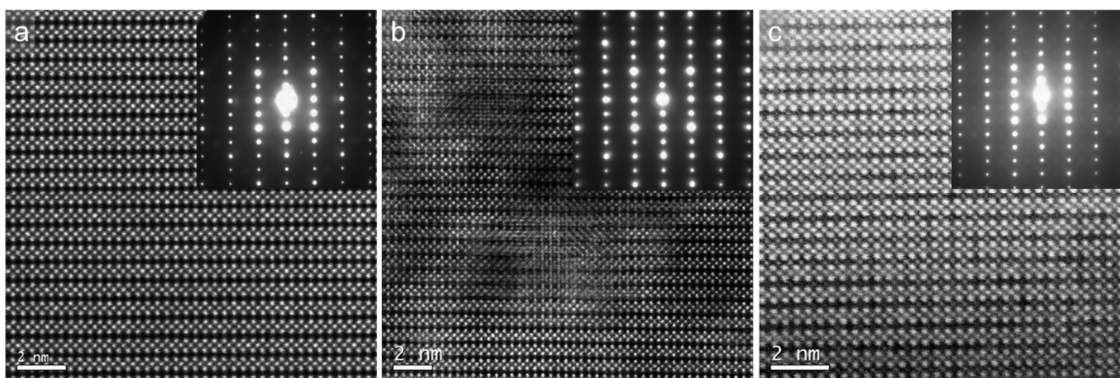


Figure S2. Atomically-resolved HAADF images of electrolytes quenched at different temperatures. (a) pristine non-quenched and 700°C-quenched. (b) 1000°C-quenched. (c) 1300°C-quenched. The corresponding diffraction patterns show no obvious changes except the spot intensity, while the dark patterned spot arrays in 1300°C-quenched indicate the emerging of a ordered metastable structure.

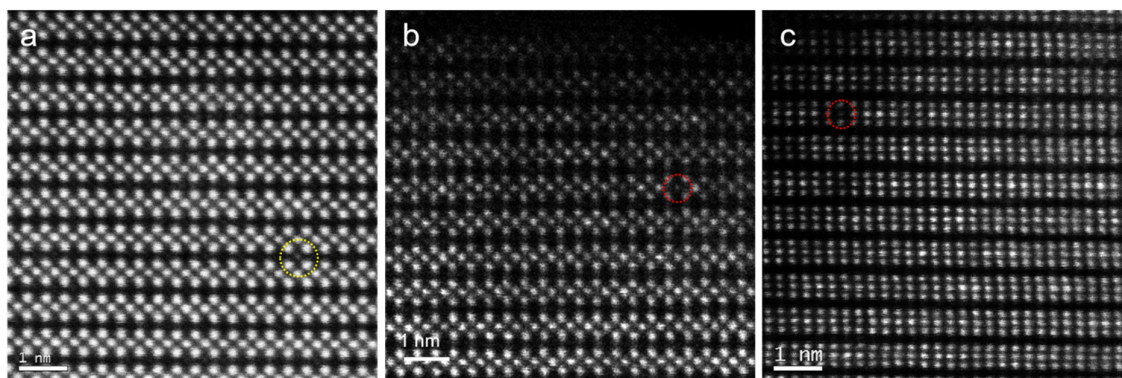


Figure S3. Atomically-resolved HAADF images of 700°C-quenched electrolytes along [100] direction (a,b) and [110] direction (c). Yellow and red circles indicate La interstitials and vacancies, respectively.

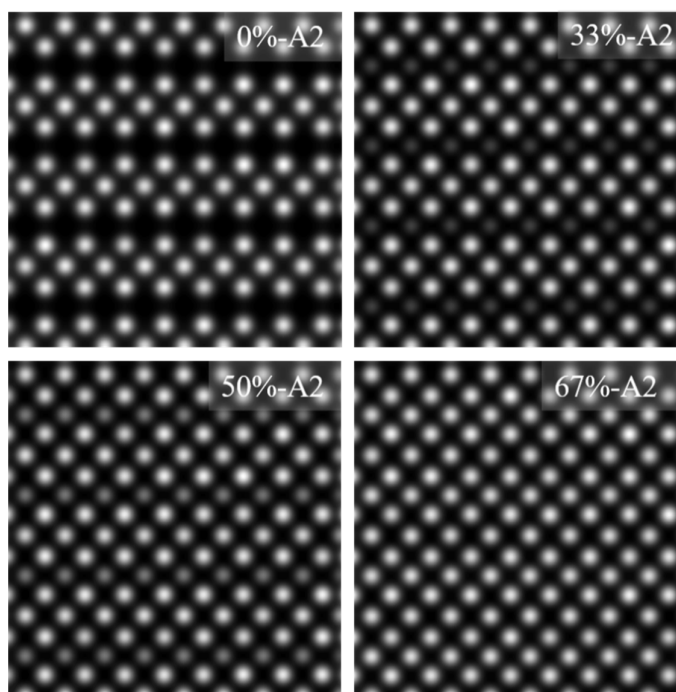


Figure S4. Simulated HAADF image of LLNO electrolytes along [100] direction with different La_{A2} occupancies. Note that 0% occupancy in A2 layer corresponds to the perfect lattice, and the La occupancy in A1 layer in the perfect crystal is 67% and the left 33% are vacancy sites to accommodate Li ions. All these simulations are based on the structures in which La_{A1} occupancy keep as a constant (67%) and La_{A2} occupancy changes.

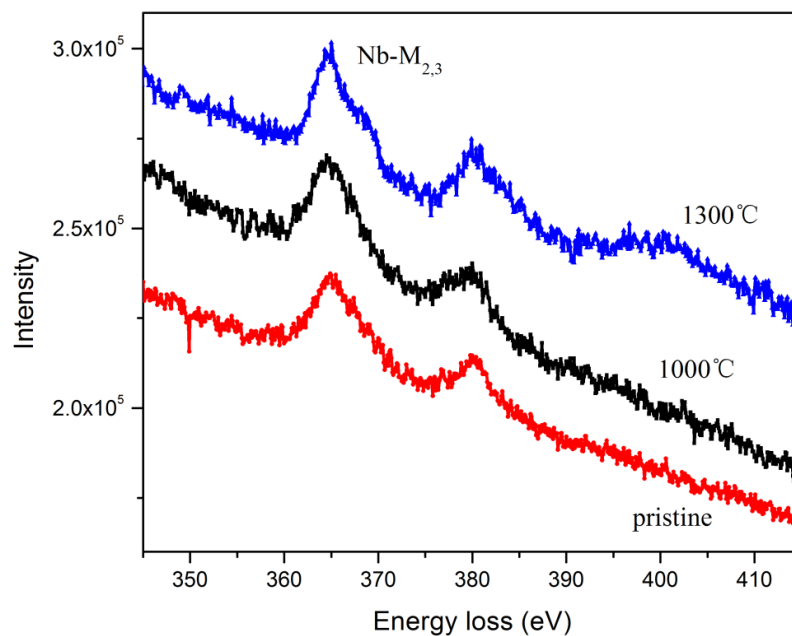


Figure S5. EELS-ELNES of Nb in solid electrolyte LLNO quenched at different temperatures. The intensities of Nb-M₂ at 375eV and Nb-M₁ at 400eV increase with the quenching temperature, resulted from the more coordinated Nb after the atom rearrangement at high temperature.