Ab initio study of the capacity loss in Over-Lithiated Oxides cathode material for Li-ion batteries

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The Over-Lithiated-Oxides (OLO), composed of the layered structure $x\mathrm{Li_2MnO_3}(1\text{-}x)\mathrm{LiMO_2}$ (M=Mn,Fe,Co, Ni), are shown to deliver much higher capacity than traditional layered oxides, due to the presence of the $\mathrm{Li_2MnO_3}$ phase. However, a long voltage "plateau" around 4.5 V and a large capacity loss (over 100 mAh g^{-1}) are observed during the first charge-discharge cycle. Although the microscopic origins are still under dispute and investigation, some mechanisms to explain this strange phenomenon have been proposed based on experimental findings, including O^{2-} ion migration, lattice densification, and Li^+/H^+ exchange. Through our *ab initio* density-functional theory (DFT) simulations, we will investigate mechanisms from atomic-scale models to analyze the stable configurations and the variation in the electronic structure with $Li^+/O^{2-}/M^{n+}$ extraction. Based on atomic scale understanding of those mechanisms, we will discuss some helpful methods to overcome the capacity loss problem.

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