Tuning the physical properties of transition metal complex oxides via oxygen off-stoichiometry

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Ionic liquid gating (ILG) method

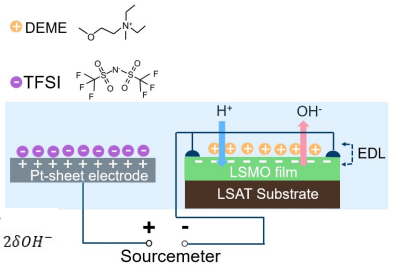
ILG principle:

1.Electrostatic doping: charge depletion induced by electric double layer (EDL).

2.Insertion or removal of ions induced by the electrical field.

3.Electrochemical Reaction:

$$La_{0.7}Sr_{0.3}[Mn^{3+}_{0.7}Mn^{4+}_{0.3}]O_3 + 2\delta e^- + \delta H_2O \leftrightarrow La_{0.7}Sr_{0.3}[Mn^{2+}_{x}Mn^{3+}_{2\delta-x}Mn^{3+}_{0.7-x}Mn^{4+}_{0.3-(2\delta-x)}]O_{3-\delta} + 2\delta OH^-$$



Lattice structure variation by ILG (XRD)

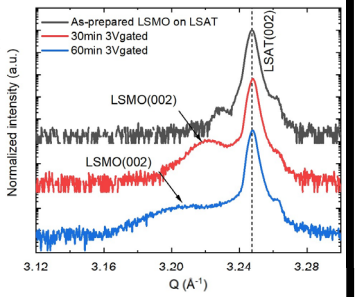
Structural variation:

1.Lattice expansion after 30min ILG at 3V.

2. “plateau”-like feature after 60min.

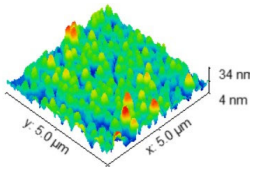
3.Coexistence of many sub-systems with a wide distribution of lattice parameters?

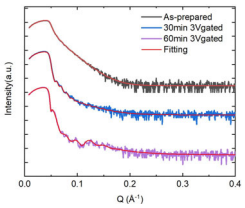
4. What causes this non-uniformity? Can it be solved?



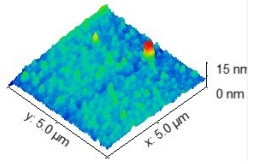
Surface and thickness change (AFM, XRR)

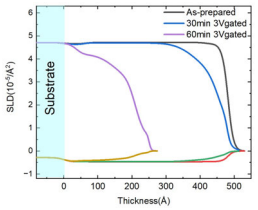
As-prepared:



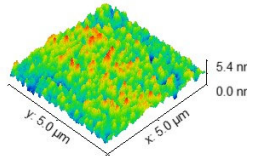


30min 3V:





60min 3V:

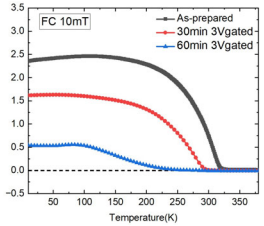


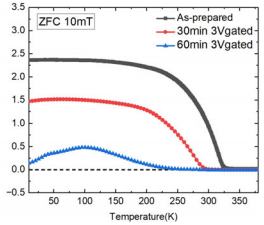
1.AFM: reduced roughness after ILG， from 3.11nm (AS) to 0.63nm (30min) and to 0.44nm (60min).

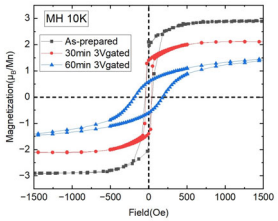
2.XRR: reduced film thickness and SLD.

3.ILG induced etching?

Magnetic properties (SQUID)







1.FC: Lower Tc and reduced magnetization triggered by hydrogen insertion or oxygen vacancy formation?

PNR and NRA measurements are need!

2.ZFC: Antiferromagnetic behavior after 60min gating?

3. MH: after 60min gating, magnetization reversal by incoherent rotation? Indicating sub-systems?

1.Diffuse scattering from surface roughness or from sub-systems?

GALAXI WAXS