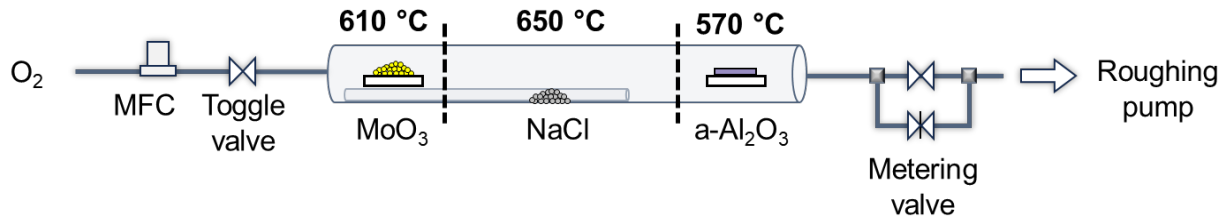
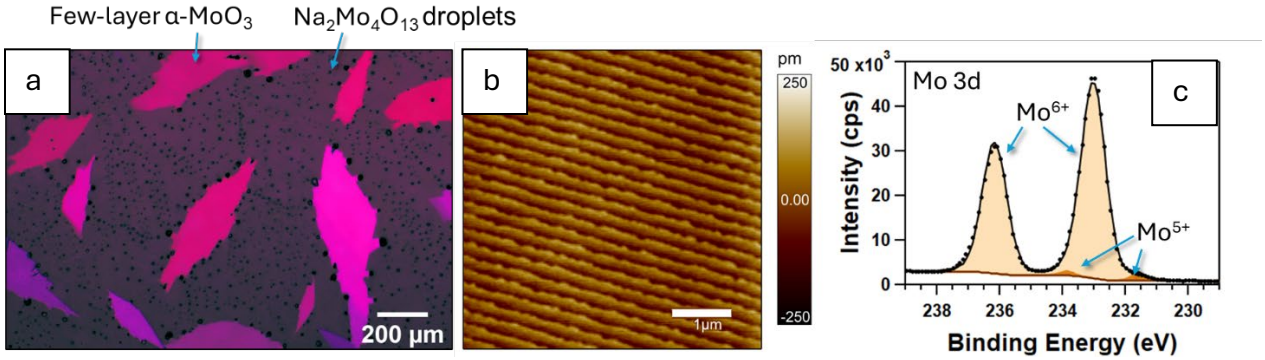
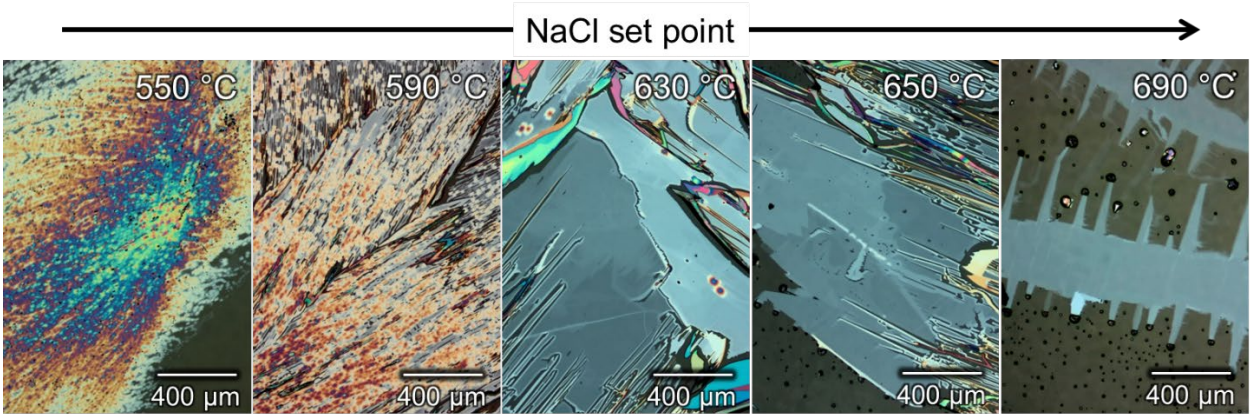


Schematic of alkali-assisted physical vapor transport equipment using a three-zone tube furnace to individually control MoO_3, NaCl, and substrate temperatures.



Overview of α-MoO_3 single crystals grown on *a*-axis sapphire. (a) Polarized optical micrograph indicating absence of grain boundaries within each crystal, and presence of residual $Na_2Mo_4O_{13}$ droplets which served to mediate the crystal growth. (b) Atomic force micrograph showing atomically smooth step-flow growth. (c) X-ray photoelectron spectrum of the Mo 3d core levels indicating primarily 6+ oxidation state, with minimal 5+ likely due to surface hydroxylation.



Optical micrographs of α-MoO_3 growth as a function of NaCl concentration as controlled by modifying the sublimation temperature. Smooth and monocrystalline growth is favored at higher NaCl concentrations.