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Electronic-Structure Engineering Through Atomic-Scale Strain Control in Geometrically Frustrated Spinel LiV_2O_4

Yu-Mi Wu[1], Ulrike Niemann[1], Minu Kim[1], Yi Wang[1,2], Hidenori Takagi[1] and Peter A. van Aken[1*]

[1.] Max Planck Institute for Solid State Research, Stuttgart, Germany.
[2.] Center for Microscopy and Analysis, Nanjing University of Aeronautics and Astronautics, Nanjing, China.
* Corresponding author: p.vanaken@fkf.mpg.de

In complex oxide systems, the coupling of local atomic configurations and electronic degrees of freedom play a fundamental role in understanding exotic phenomena such as formation of charge ordering, metal-insulator transition and colossal magnetoresistance. Atomic-scale thin film syntheses enable to disentangle these competing interactions and tune novel ground states of materials, which do not exist in bulk crystals. Modifying strain by depositing epitaxial thin films on substrates with different lattice spacing results in a precise control of the local physical behavior. Scanning transmission electron microscopy (STEM) combined with electron energy-loss spectroscopy (EELS) not only provides information on the local atomic structure, but also on the site-specific chemical composition and electronic structure of materials and interfaces. Such direct real-space observations are important to unveil the microscopic origins of macroscopic properties in complex oxides and are essential for possible applications in relevant electronic and spintronic devices.

Here, we demonstrate how strain locally alters physical properties in a geometrically frustrated spinel using STEM-EELS, where epitaxial LiV_2O_4 thin films are grown on $SrTiO_3$ and MgO substrates by pulsed-laser deposition. We find that the V-3*d* electrons redistribute in the lattice and become confined or delocalize depending on the epitaxial strain, hereby generating ordered or disordered patterns. We reveal two competing behaviors of the thin films on the two different substrates, a metallic charge-disordered phase on $SrTiO_3$ and an insulating charge-ordered phase on MgO. Our findings evidence that charge fluctuations can freeze into an insulating “charge-glass”-like state, when epitaxial strain is applied, which relieves the frustration in LiV_2O_4. These results provide a route for understanding, controlling and designing local novel phases in complex oxides heterostructures [1].

References:

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