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Soft chemical dimensionality evolutions in complex oxides

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Synthesis of complex oxides is challenging: Coordination, connectivity, cation ratio and oxygen stoichiometry must all be correct but conventional synthesis relying on thermodynamics often addresses a subset of these features at the expense of the others. Thermodynamically stable phases are usually straightforward to realize, while a host of adjacent or similar phases remain out of reach. Chimie douce, or soft chemistry, is a set of tools for low temperature, kinetics-based post-processing to achieve phases outside of thermodynamic stability starting from a stable precursor [1, 2].

Redox-active soft chemistry on complex oxides has mostly focussed on topotactic oxidation and reduction – where oxygen stoichiometry and coordination can be modified, but connectivity and cation ratio must be preserved [3].

Here I will present an additional tool for the soft chemistry toolbox: Redox-active dimensionality evolutions, where quasi-2D layered compounds can be achieved by post-processing their 3D counterparts, and vice versa. For example, evolutions between different members of the same Ruddlesden-Popper series, $A_{n+1}B_nO_{3n+1}$, and the perovskite end compound, ABO_3.

The $LaCoO_3 \rightarrow La_2CoO_4$ evolution is taken as a test case. 3D $LaCoO_3$ is thermodynamically stable, while quasi-2D La_2CoO_4, an interesting strongly correlated magnet, is not. We grow epitaxial single crystal thin films of $LaCoO_3$ by pulsed laser deposition, then post-process to realize the single-layer Ruddlesden-Popper La_2CoO_4. I will present our preliminary work on understanding the fundamental materials chemistry of this evolution by in-situ probes and advanced microscopy, as well as promising solid-gas reaction control knobs and potential for generalization.

References:

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[3] Hayward M. A. "Topochemical reactions of layered transition-metal oxides", Semicond. Sci. Technol. **29** (2014)