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STEM Imaging, Monochromated EELS, and Theory of Natural and Artificial Superlattices

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As the length-scales of materials decrease, heterogeneities associated with interfaces approach the importance of the surrounding materials. The importance of interfaces, or interface-like planes, has led to extensive studies combining electron energy-loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM) that relate local atomic structure to emergent electronic and magnetic properties of superlattices. Previous studies have focused on electronic characterization because the energy resolution limitations of STEM-EELS precluded losses of tens to hundreds of meV. Energy resolution is now less limiting, allowing for local characterization of lower-energy-loss electronic excitations and atomic vibrations in materials with closely spaced interfaces.

A cross-over exists where the description of repeating crystals separated by interfaces loses meaning and the structure is better described by a single crystal structure. Examples include artificially grown short period superlattices [1] or the naturally occurring Ruddlesden-Popper crystal structure [2]. Here we combine STEM imaging, monochromated EELS, and density-functional-theory (DFT) calculations to explore the cross-over of hierarchical lattices such as $SrTiO_3$-$CaTiO_3$ superlattices and Ruddlesden-Popper $Ba_{n+1}Zr_nS_{3n+1}$. In Figure 1 we show period-dependent vibrational response of $SrTiO_3$-$CaTiO_3$ superlattices that is related to the local atomic arrangements of oxygen octahedra tilting. In Figure 2, we show energy-losses resulting from the superlattice structure of Ruddlesden-Popper $Ba_3Zr_2S_7$ [3]

References:

[1] E.R. Hoglund, *et al.*, Nature. **601** 7894 (2022) pp. 1–9.

[2] N.M. Dawley, *et al.*, Appl. Phys. Lett. **118** 9 (2021) 091904-6.

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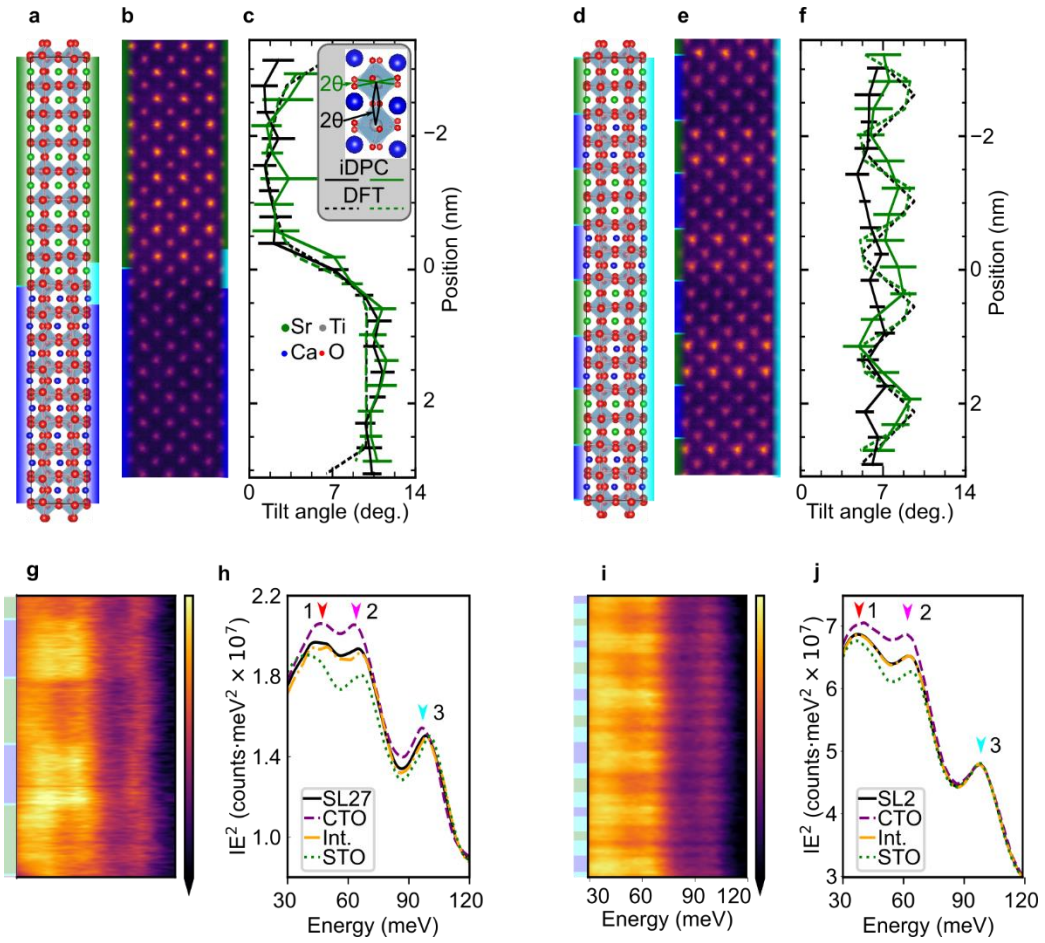

Figure 1. $SrTiO_3$-$CaTiO_3$ large period superlattice (a) atom model, (b) HAADF, and (c) tilt angle compared to small period superlattice (d) atom model, (e) HAADF, and (f) tilt angle. Vibrational EELS line scans in the (g) large and (i) small period superlattices with layer averaged spectra shown in (h) and (i).

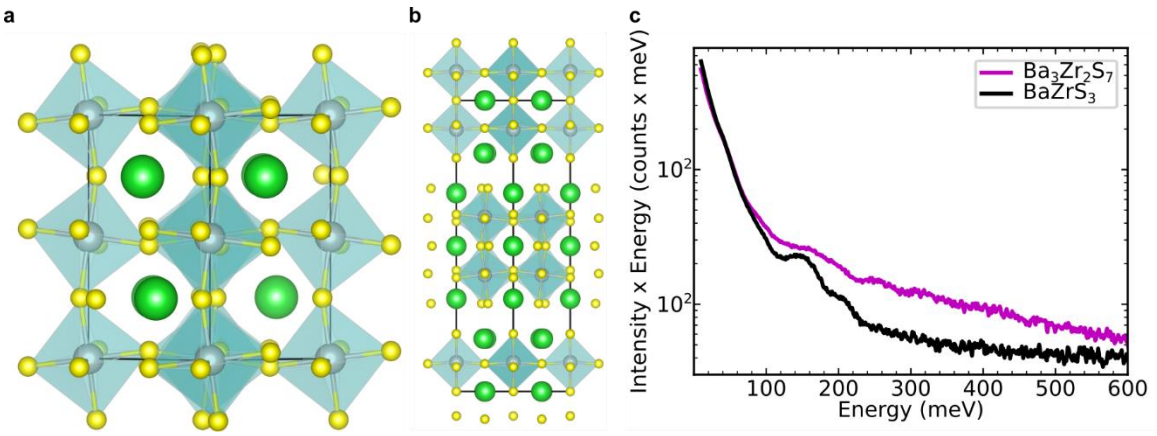

Figure 2. (a) Perovskite $BaZrS_3$ and (b) $Ba_3Zr_2S_7$ Ruddlesden-Popper models and the low energy loss spectra.

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