Improper Ferroelectricity at the Monolayer Limit with Undiminished Curie Temperature in *h*-$LuFeO_3$

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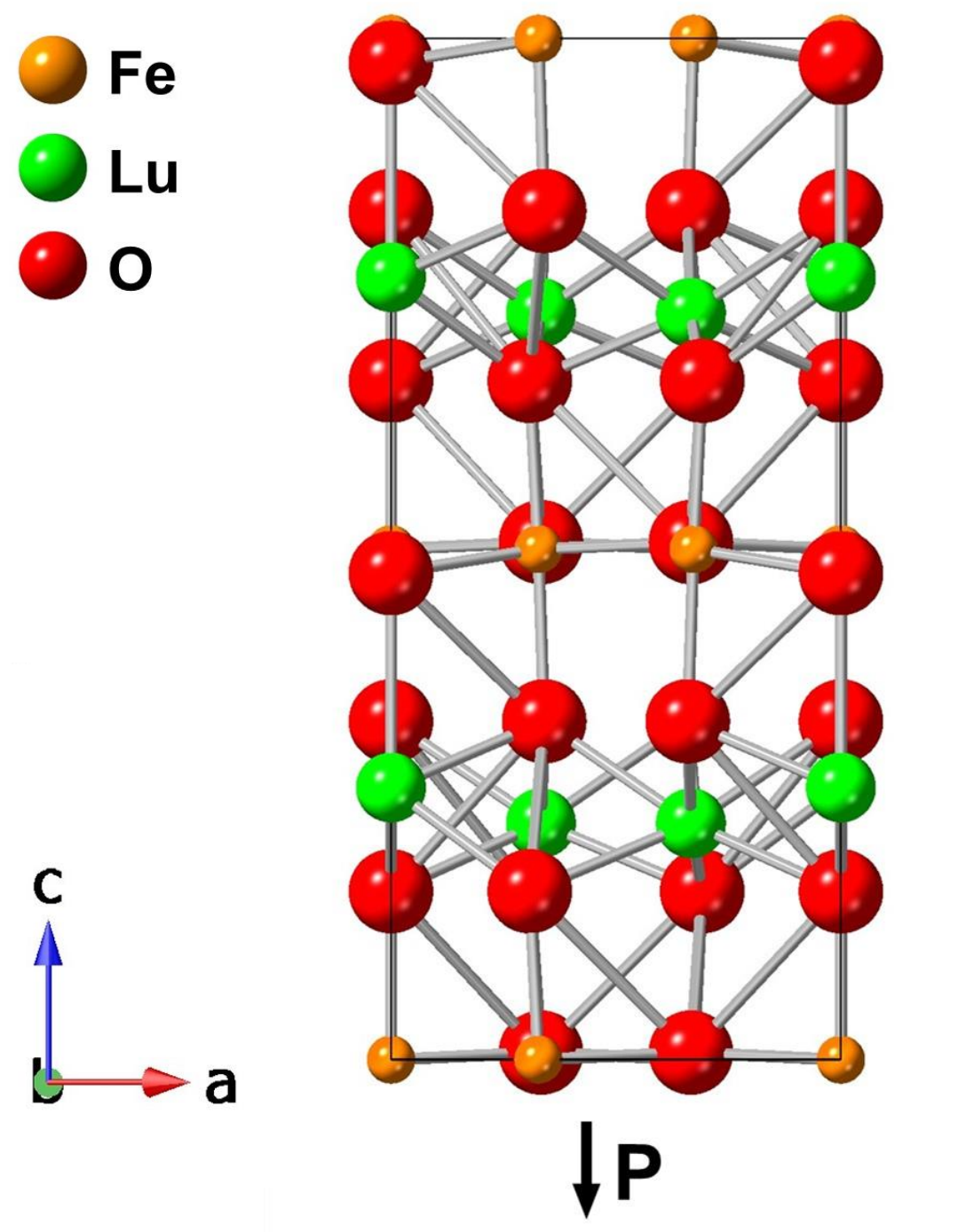
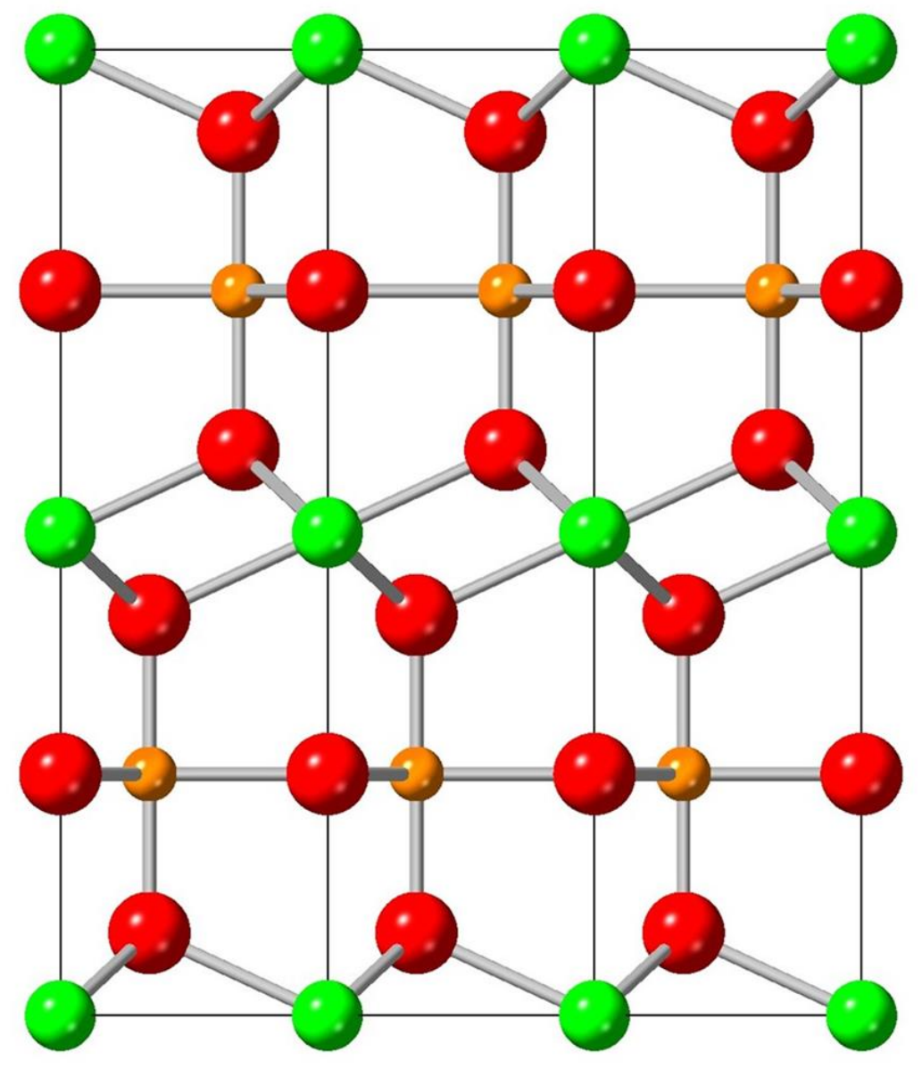
Background

Ferroelectricity:
Spontaneous polarization that can be reversed by an applied electric field
Caused by a phase transition that lowers structural symmetry below Curie temperature, T_C

Proper ferroelectricity:
Polarization is the primary order parameter of the phase transition
Improper ferroelectricity:
Polarization is a secondary order parameter of the phase transition



($T < T_C$, ferroelectric)

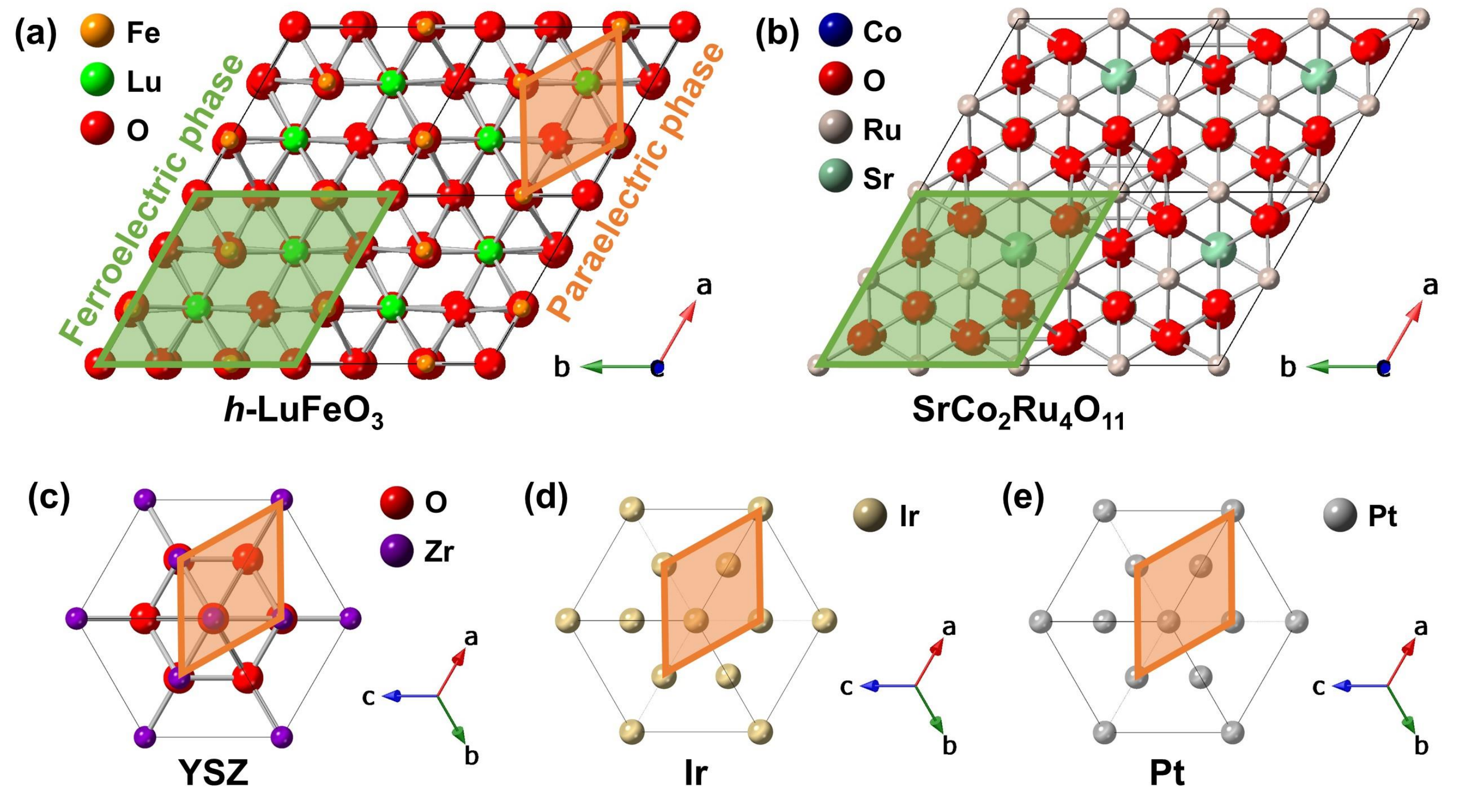


no net **P**
($T > T_C$, paraelectric)

- Experimental measurements [1,2] suggest a critical thickness for ferroelectricity, on the order of several unit cells
- Theoretical calculations [3] posit a lack of critical thickness for improper ferroelectricity

- Our novel bottom electrode, $SrCo_2Ru_4O_{11}$ (SCRO), facilitates ultrathin ferroelectricity in *h*-$LuFeO_3$
- SCRO provides a template conducive to the growth of the ferroelectric phase of *h*-$LuFeO_3$
- Other substrates and bottom electrodes, including:
  - (111) YSZ
  - (111) Ir
  - (111) Pt

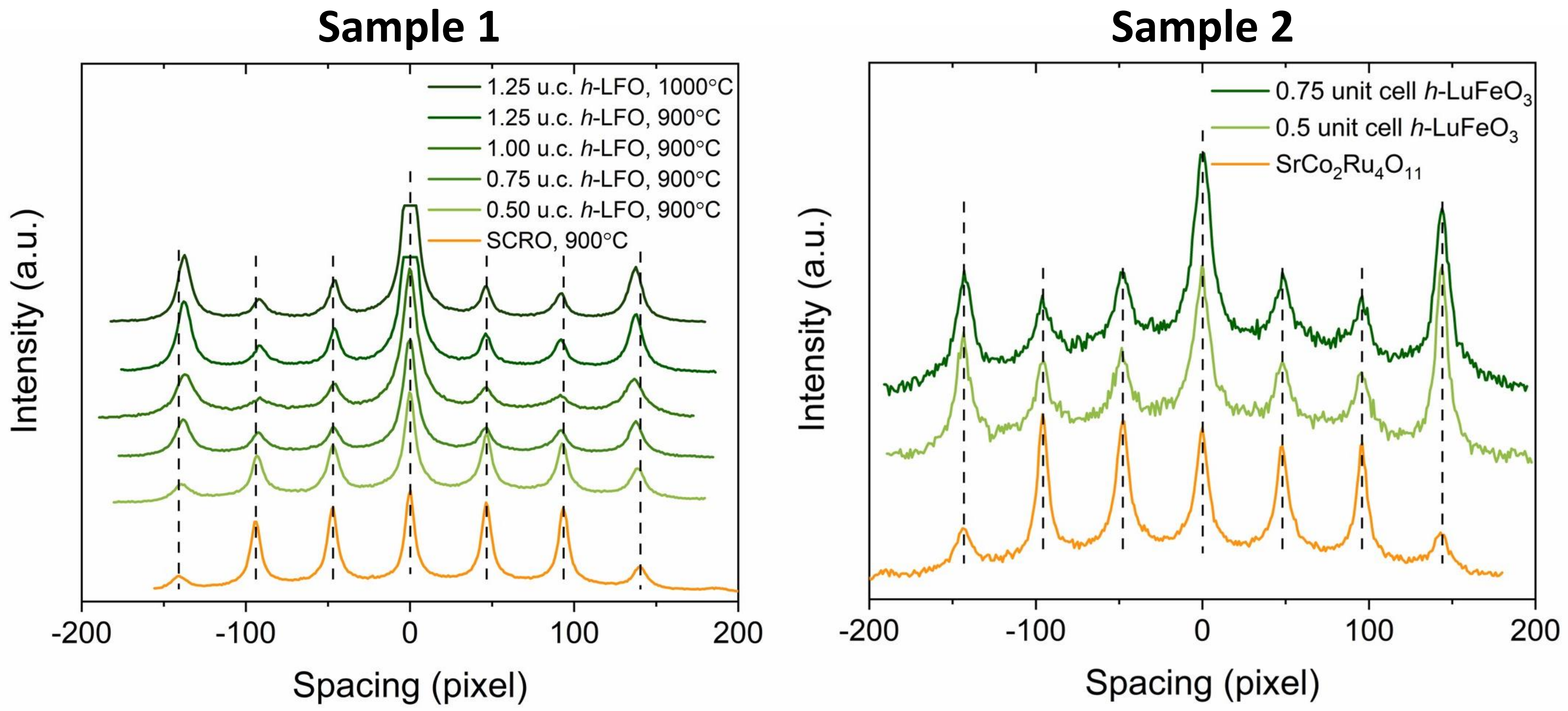
  promote the growth of paraelectric *h*-$LuFeO_3$



Methods

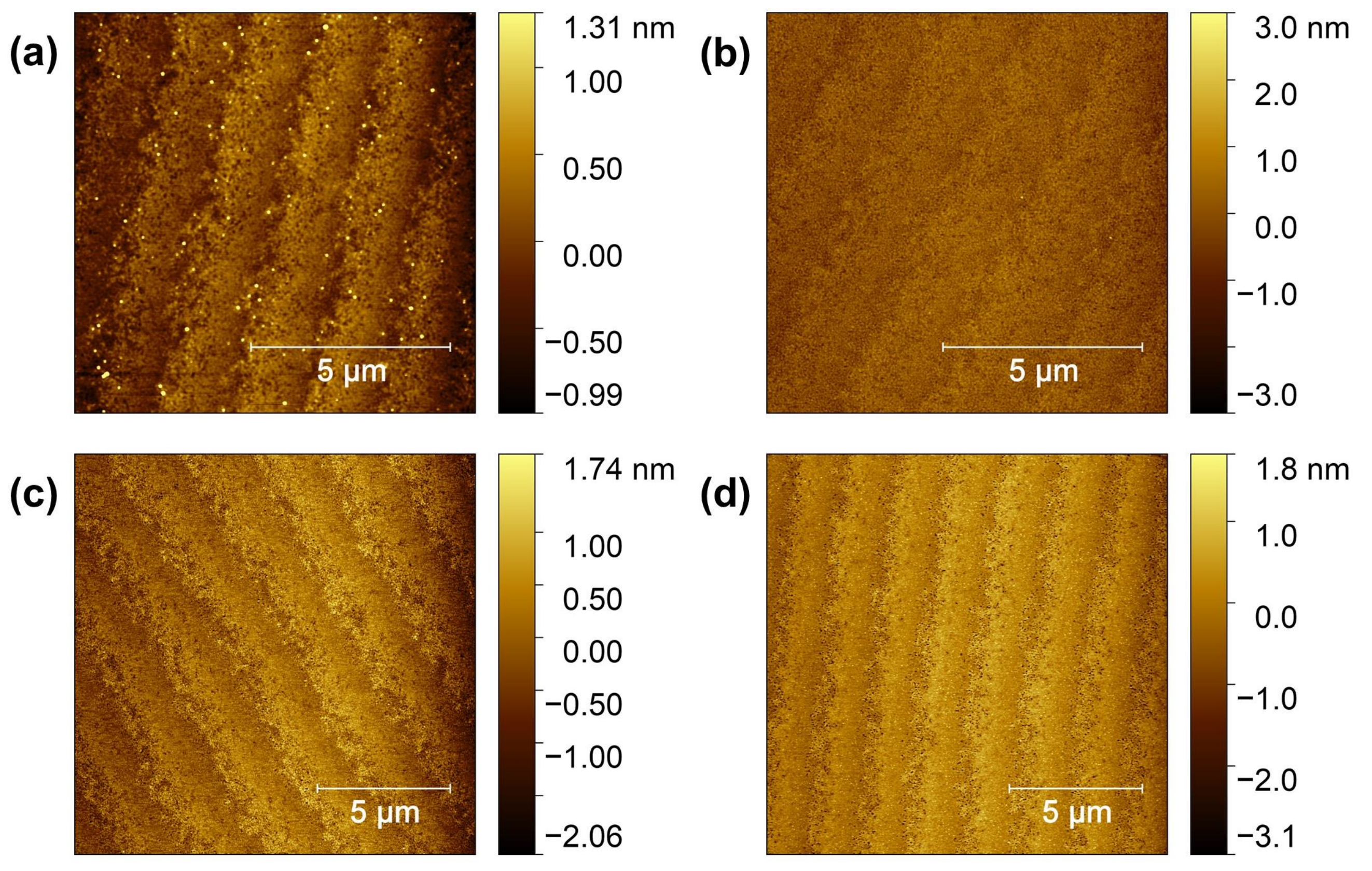
- 16 nm SCRO was grown on $Sr_{1.03}Ga_{10.81}Mg_{0.58}Zr_{0.58}O_{19}$ (SGMZ) substrates with molecular beam epitaxy (MBE)
- *h*-LFO was grown on SCRO via MBE, alternately growing one monolayer of iron oxide or lutetium oxide at a time
- Film quality was monitored via reflection high-energy electron diffraction (RHEED)
- Atomic force microscopy (AFM) images were compared before and after growth to confirm film uniformity

Results



- Line profiles for growth of (a) Sample 1 (1.25 unit cells thick) and (b) Sample 2 (0.75 unit cells thick)
- 2.3% spacing difference between *h*-$LuFeO_3$ and SCRO peaks in Sample 1 supports their identification as different materials
- Ferroelectric tripling is observed in Sample 1 at 0.75 unit cells thick and in Sample 2 at 0.5 unit cells (1 formula unit) thick

AFM images before and after growth: (a) bare SCRO before growth of Sample 1 (RMS=0.33 nm), (b) Sample 1 after growth (RMS=0.43 nm), (c) bare SCRO before growth of Sample 2 (RMS=0.59 nm), (d) Sample 2 after growth (RMS=0.59 nm)



Conclusions

- *h*-$LuFeO_3$ is an improper ferroelectric with no critical thickness and an undiminished phase transition temperature
  - RHEED tripling provides evidence for ferroelectricity at a thickness of one formula unit (Sample 2)
- AFM images show preservation of terraces and low RMS roughness values, indicating smooth, uniform film
- AFM confirms that RHEED tripling is from *h*-$LuFeO_3$ thin film, not uncovered SCRO or thicker islands of *h*-$LuFeO_3$
- Next steps: scanning transmission electron microscopy (STEM) images and polarization switching measurements

We demonstrate that there is **no critical thickness** for **improper ferroelectricity.**

We provide a framework for the **fabrication of ultrathin improper ferroelectrics** via epitaxial engineering.

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References

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