Using Inverse Photoemission to Study Complex Oxides

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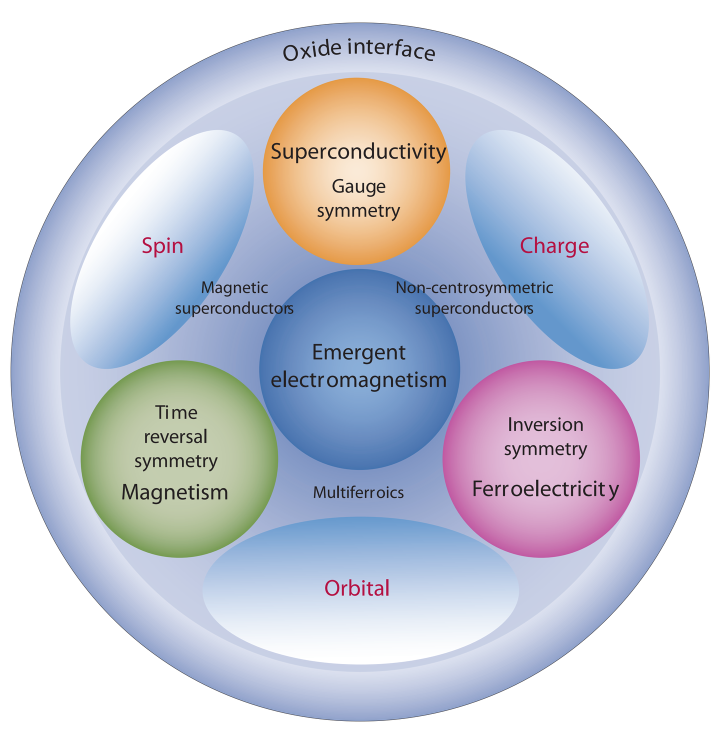
Complex Oxides: Next-Generation Materials

What Are Complex Oxides?

- Materials which are composed of at least two types of metal ions along with oxygen.
- Metal and oxygen ions tend to organize into ordered lattices
- Example: $La_{2-x}Sr_xCuO_4$ forms CuO_2 planes separated by layers of (La,Sr)O
- Many interesting materials belong to this family, such as the high-temperature superconducting cuprates



Hwang et al., Nature Mater. **11**, 103 (2012)

Why Study Complex Oxides?

- Strong interplay between various degrees of freedom: charge, spin, orbital, and lattice
- Competing effects create a large variety of interesting phases, as shown on left
- Some examples: high-temperature superconductivity, metal-insulator phase transitions, multiferroicity (coupling of electric and magnetic polarizations)
- These phases can be used in many kinds of devices, such as transistors, non-volatile memory, solar cells, etc

K-resolved Inverse Photoemission (KRIPES)

What is Inverse Photoemission?

Emission of photon from a material which absorbs an incoming electron.

Momentum (K)-Resolved:

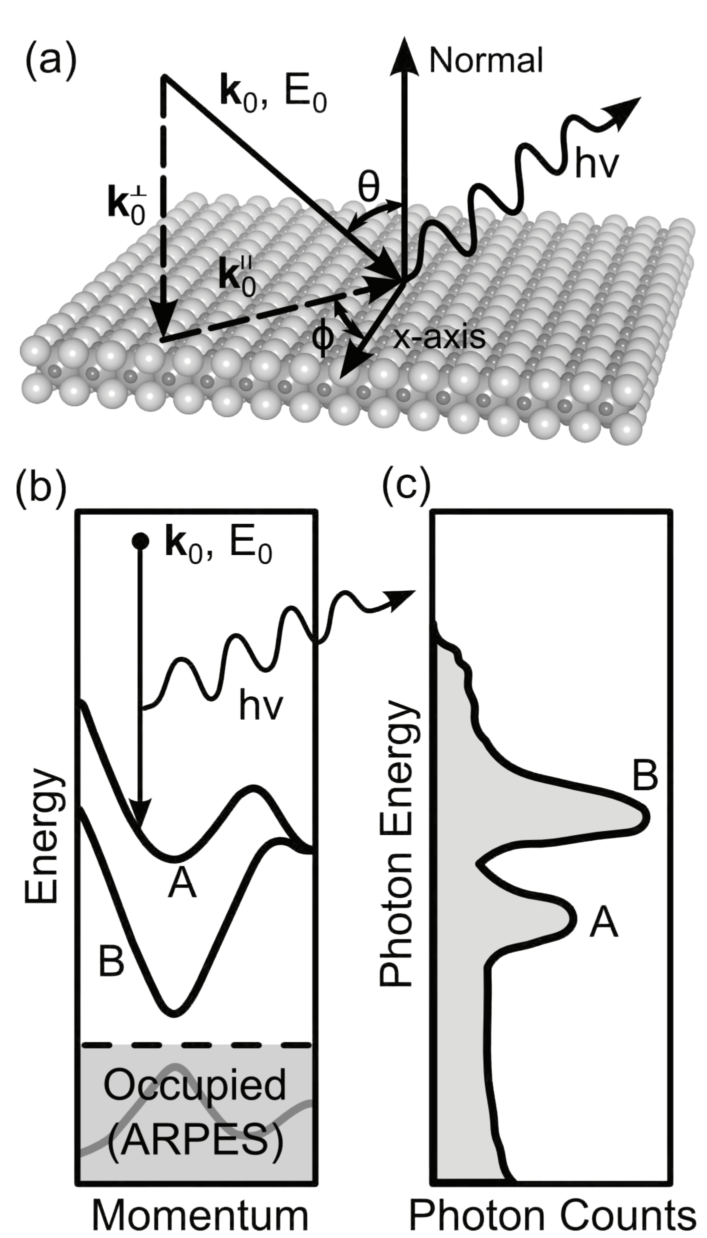
Given incident electron energy (E_0) and momentum (k_0) and measuring outgoing photon energy (hv), can determine unoccupied electronic band structure

Studying on Complex Oxides:

- Strong interactions in complex oxides makes them difficult to fully model
- Important quantities like bandgap size not accurate from computational methods
- ARPES and KRIPES directly measure the electronic band structure in materials and understand their electronic properties

KRIPES:

- Many ARPES experiments already exist studying complex oxides, but no KRIPES experiments
- ARPES only measures occupied band structure, can't see unoccupied bands
- KRIPES only method to directly measure unoccupied band structure and provide important information about bandgap size, nature of excitations, etc



Schematic of KRIPES
Image courtesy of Ed Lochocki

KRIPES Chamber, Under Construction

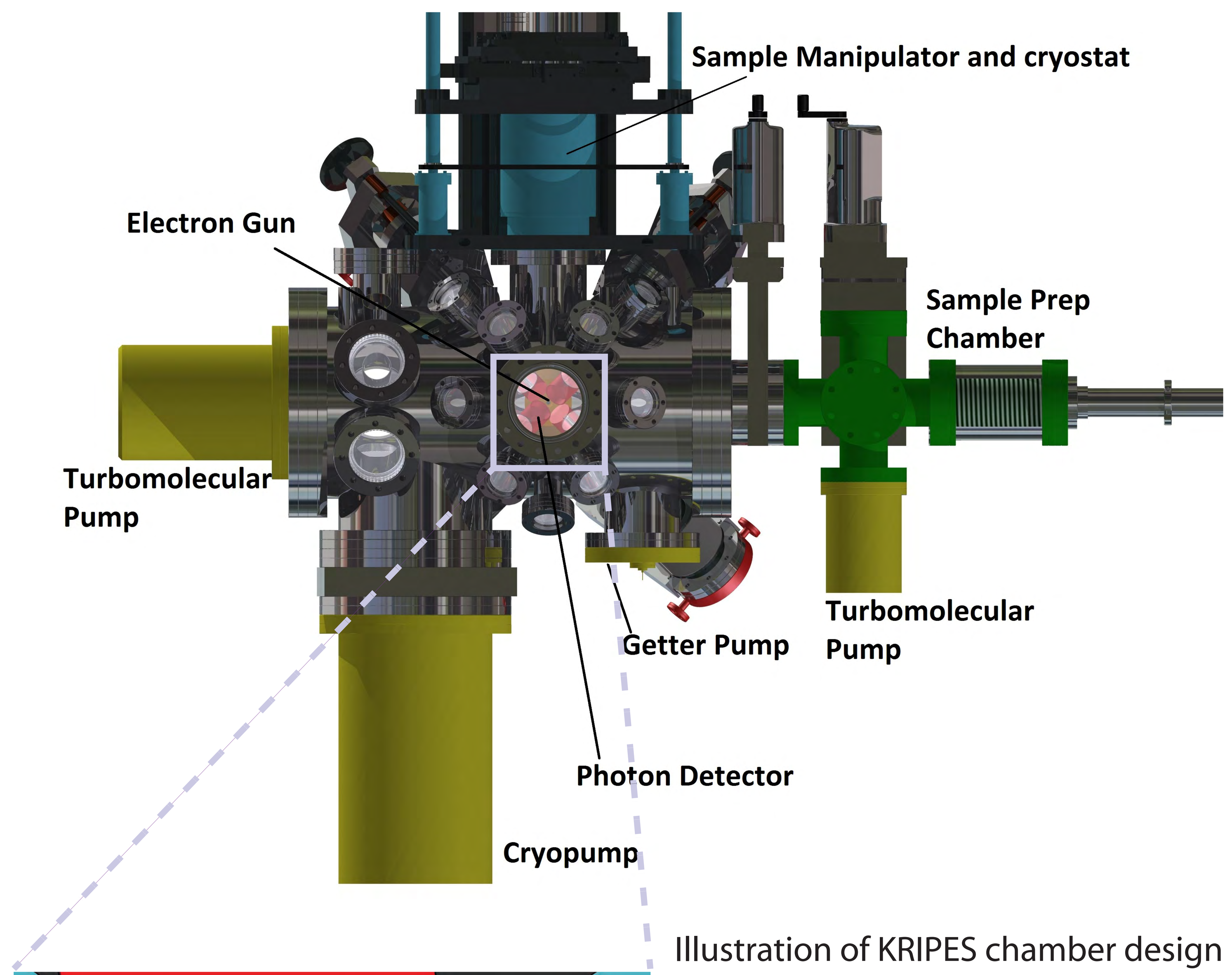


Illustration of KRIPES chamber design

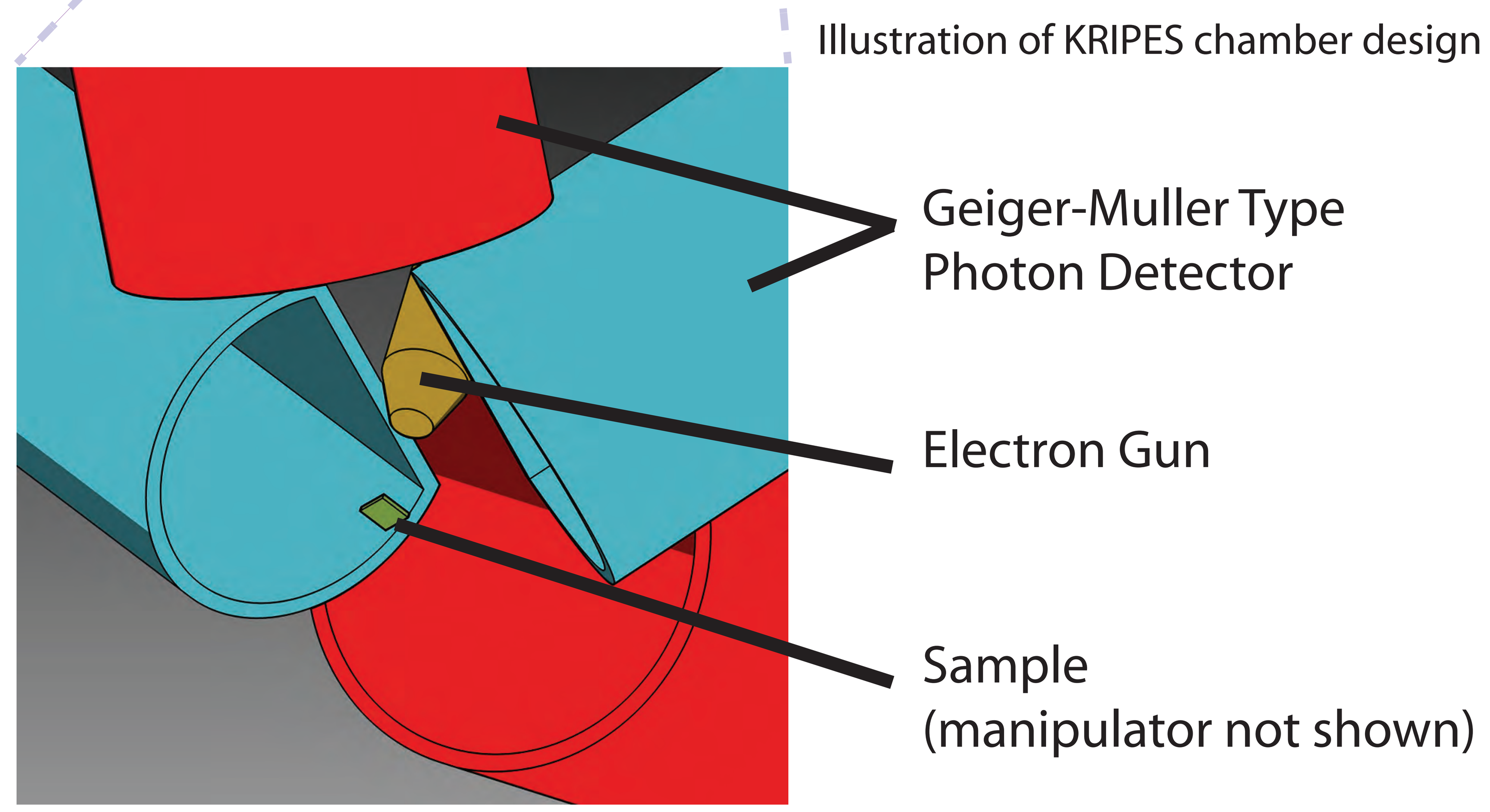


Illustration of detector geometry.

Apparatus Capabilities

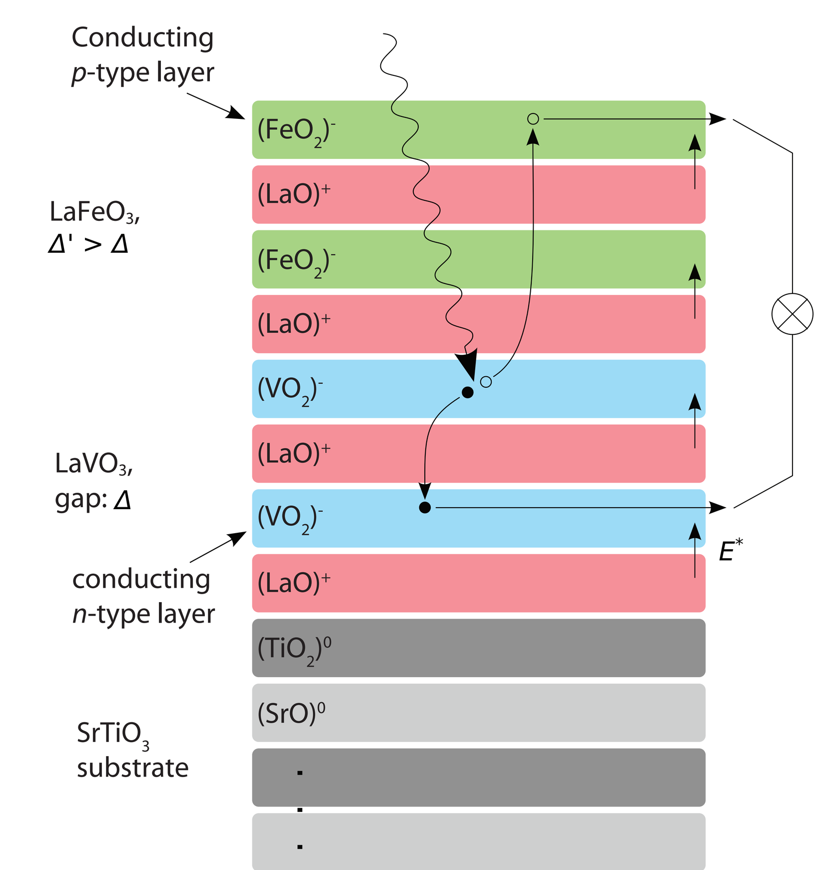
Design Needs:

- Inverse photoemission process has small cross section
- Photon emission rate is very small compared to photoemission (ARPES)
- Detector geometry needs to capture large percentage of emitted light
- Chamber must support extremely high vacuum (10^{-12} torr)

Final Design Specifications:

- Manipulator with six axes of motion (x, y, z and all 3 rotation axes) for fully capturing momentum space
- Four detectors to capture 85% of total emitted photons
- Variable energy resolution from 85 meV to 350 meV to optimize measurements for high count rate vs high energy resolution
- Multiple pumps to achieve UHV, mu-metal shields for stray magnetic fields
- Closed-cycle cryostat for very long measurement times

Complex Oxides for Sustainability - Efficient Solar Cells



Schematic of proposed solar cell heterostructure
Assmann et al, Phys. Rev. Lett. **110**, 078701 (2013)

Proposed Structure

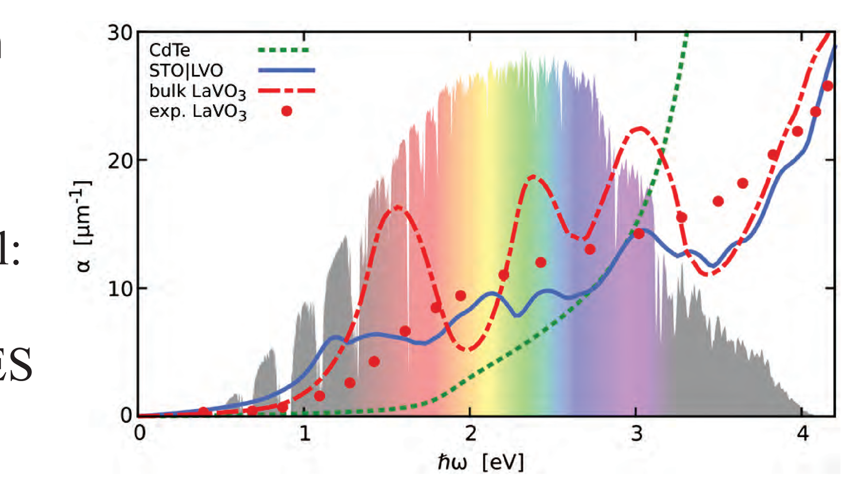
Alternate layers of $LaVO_3$ and $LaFeO_3$ grown on top of $SrTiO_3$.

Advantages:

- Intrinsic electric field to separate photoexcited carriers
- Band gap in ideal energy range for capturing sunlight
- Metallic interfaces a natural method to extract excited carriers
- Multi-layer structure allows use of materials with varied gaps to more efficiently capture sunlight

Growth and Characterization

- Use molecular beam epitaxy to grow proposed structure
- Determine viability for solar cell:
- Measure size and structure of bandgap using ARPES and KRIPES
- Measure unoccupied density of states
- Measure metallic nature of oxide interface



Calculated absorption of light in LVO on STO, compared with that of CdTe, a compound widely used in current high-efficiency solar cells.

Also from Assmann et al.

Conclusion

Understanding the properties of oxides and oxide interfaces is key to using them for applications. Towards that end, we have designed and begun building a KRIPES system which will fill an important gap in experimental efforts towards that understanding. One of the first projects we will pursue with the new system is the growth and characterization of a newly proposed complex oxide solar cell heterostructure, as the KRIPES system is especially suited for characterizing bandgaps, an important property of solar cell materials.

Acknowledgements

H.W. acknowledges support from the NSF IGERT program (DGE-0903653). This material is based upon work supported by the National Science Foundation Graduate Research Fellowship under Grant No. DGE-1144153 and by a grant from the Army Research Office.