SCHOOL OF ENGINEERING

Study of the dynamics of electron–lattice coupling in cuprates and UO_{2+x} to unravel the role of coherent tunneling polaron quantum phases

PI: Steven Conradson

Collaborators: S. Biedron, T. Bolin, M. Fazio (UNM); A. Bishop, A. Hurd, J. Sarrao, A. Taylor, R. Prasankumar (LANL); M. Palmer, J. Tao (BNL); D. Martin (ANL); C.C. Kao, D. Reis (SLAC); M. Karppinen (Aalto U.); R. Springell (U. Bristol); C.Q. Jin (CAP)

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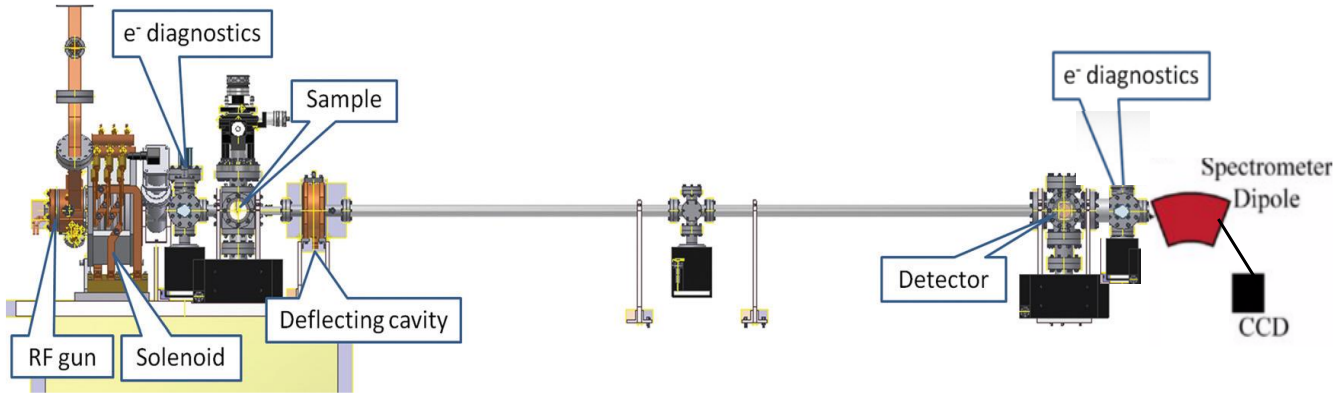
The sample preparation is supported by Steven Conradson.

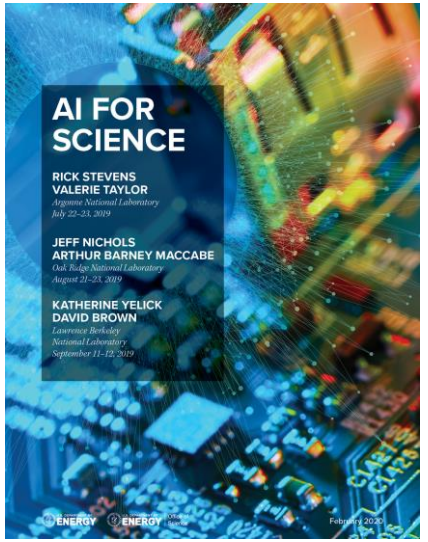
Two DOE facilities are involved: ATF and ALCF

Accelerator Test Facility (ATF)



Argonne Leadership Computing Facility (ALCF)

The combination of machine hardware, advanced computing for simulation, and data science for surrogate modelling, training of neural networks and data analysis is inspired by our past work and our participation on DOE meetings, workshops and reports such as AI for Science (*https://www.anl.gov/ai-for-science-report*).



Special Equipment Requirements and Hazards

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Special equipment:

- None.

Hazards:

- **Depleted uranium** is very mildly radioactive and chemically toxic as a heavy meta. However, the thin film samples of UO_{2+x} to be used contain at most a few hundred micrograms (relative to an average body burden of 20 micrograms from natural sources) and their activity is below background.

- Other potential hazards include the ***laser*** of the MUED instrument and the ***cryogenic system*** necessary to cool the samples to the desired temperatures. We will work with the BNL collaborators to exercise the necessary precautions.

Experimental time request

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CY2021 Time Request

Capability	Setup Hours	Running Hours
UED Facility	10	30

Time Estimate for Remaining Years of Experiment (including CY2021)

Capability	Setup Hours	Running Hours
UED Facility	10	30

Overview of the proposal

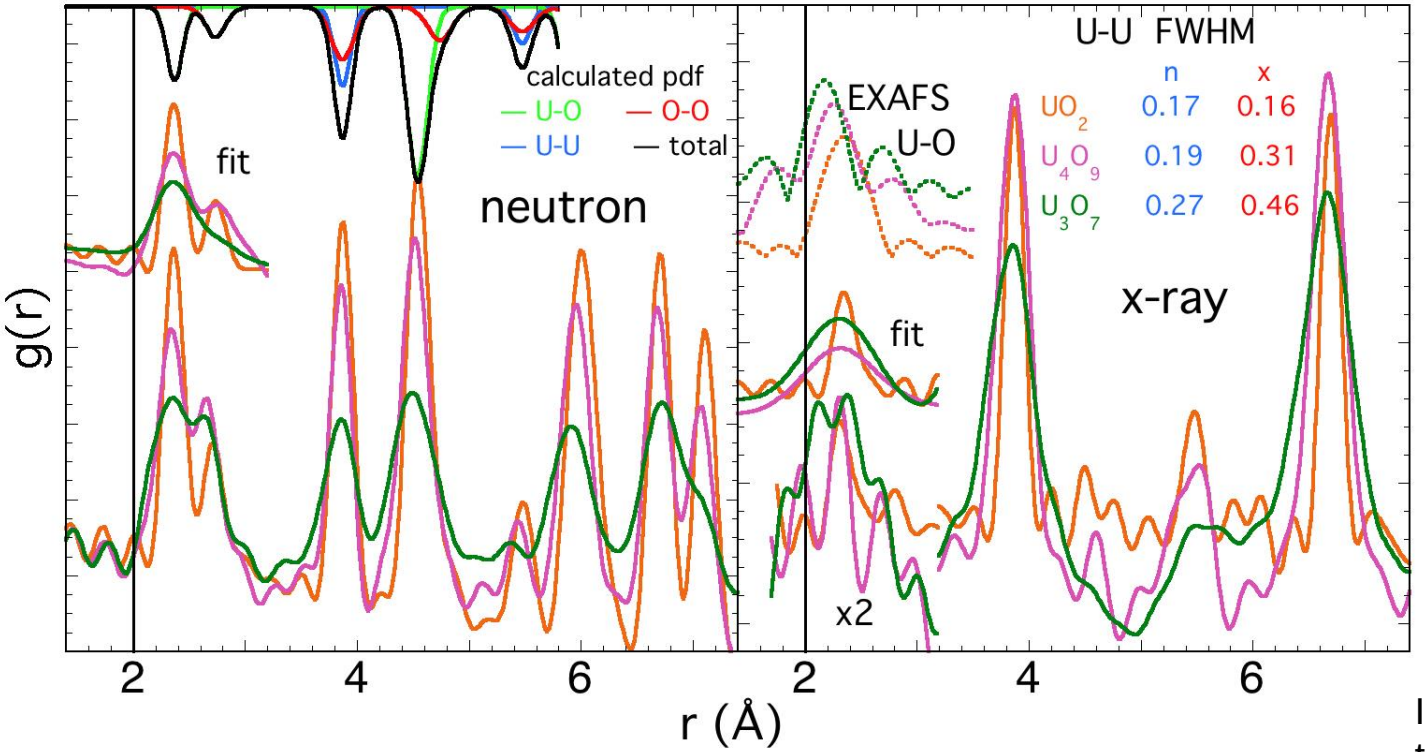
We suspect that a "*synchronized distortion*" in some materials represents hints of original physics responsible for coherent spin states. This distortion lies in the dynamic structure factor in the intermediate electron-lattice coupling regime.

The purpose of this 1-year proposal is to visualize the Internal Quantum Tunneling Polarons (IQTPs) and their domains by obtaining femtosecond-picosecond resolved real space data. We have postulated that in UO_2 the IQTPs oscillate between UO_2 and U_3O_8 / UO_3 type structures, determined by the spacing between the [111] U planes. Similarly, in the cuprates the synchronization that may stabilize the superconductivity to higher temperatures should created ordered domains that we could expect to see on timescales faster than their dissipation.

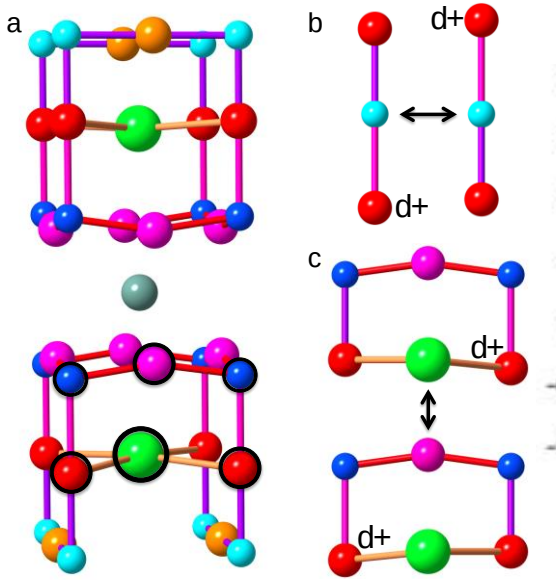
Goal: study the dynamic structure phenomena of cuprates and UO_{2+x} towards an understanding of BEC formation mechanisms and exotic superconductivity as a path forward into realizing devices based on these material systems.

Experience from SSRL XAFS experiments

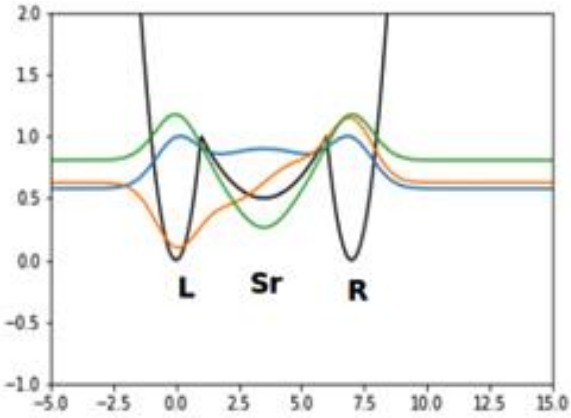
This effect was first revealed at a leading synchrotron by X-ray absorption fine structure (XAFS) that probes local structure and unoccupied local electronic states. Specifically, Internal Quantum Tunneling Polarons (IQTPs), a part of the dynamic structure.



The components of the dynamical structure are identified by the differences between measurements made with x-rays vs. diffraction and those made with neutrons. These are all identical for $UO_{2.00}$ but not for UO_{2+x} including the nearest neighbor O where XAFS and x-ray pdf find O with U-O < 2 Å, indicative of multiply bound U(VI) uranyl species.



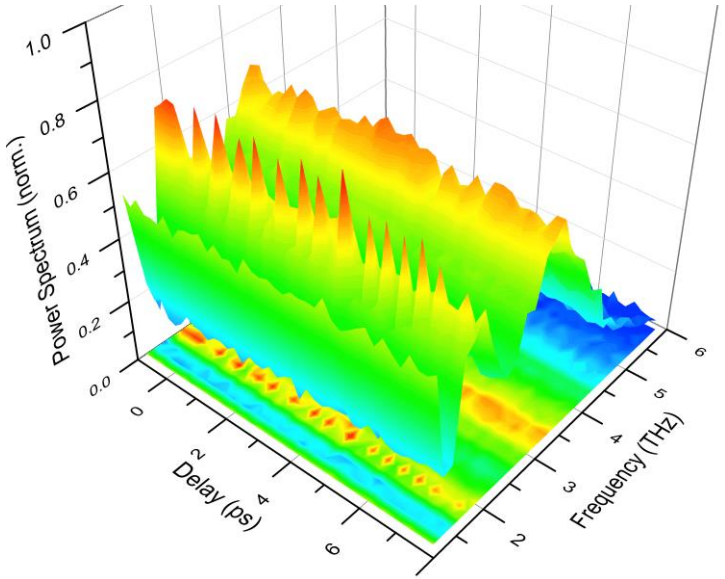
In these mixed valence compounds, the essential component is an *internal quantum tunneling polaron* in which the charge and atom displacement oscillate between two conformations, depicted here in $YSr_2Cu_3O_7$.



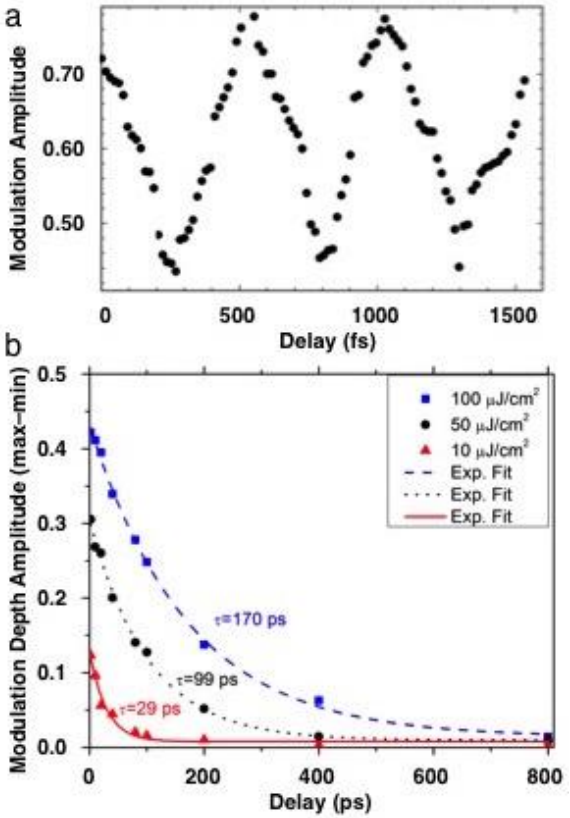
The synchronization of the oscillations causes the formation of a new dynamical phase that forms a triple well potential over the O atom in the CuO_2 plane and the Sr.

Experience from pulse-probe spectroscopy and other techniques

We have postulated that the highly unusual results from femtosecond pulse-probe laser spectroscopy measurements of UO_2 that include exceptional coherence and other unique properties originate in excitation and synchronization of its iqtps found in the combined neutron-x-ray measurements.

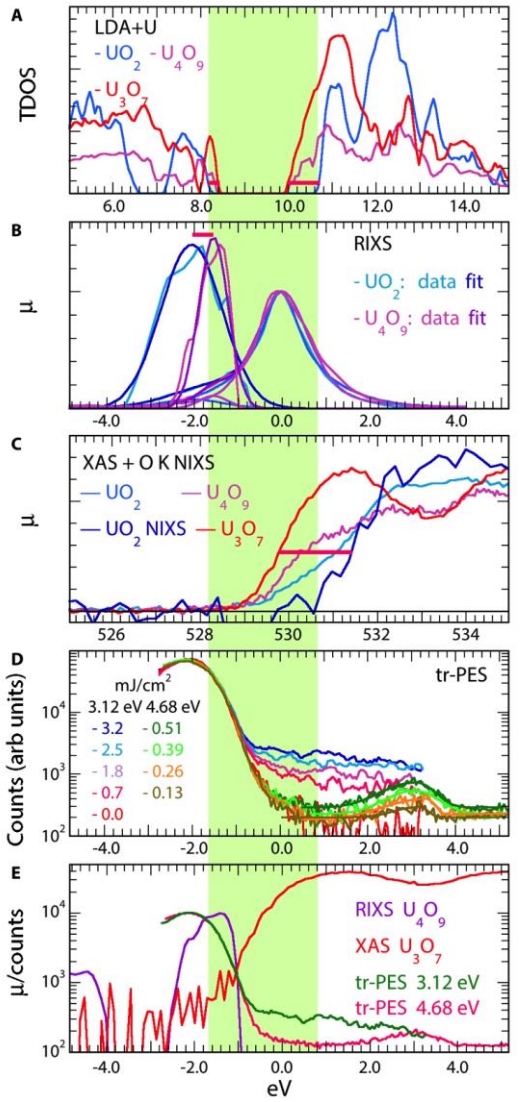


Time domain THz spectroscopy found a 1.8 THz signal that does not belong to UO_2 that remains coherent through hundreds of cycles even at 300 K.



XAS measurement of UO_{2+x} gave a much lower edge for the upper Hubbard band than predicted by DFT calculations, including a tail that may bridge the gap into the lower Hubbard band.

ARPES probe of the U6*d* final state continues to show the gap. In contrast, excitation to the U5*f* final state gives a unique "superthermal" metal, a continuously occupied band through several eV corresponding to 1000s of K, that was stable for several picoseconds.



What's next to complete the picture? - MUED

What MeV electron diffraction can tell us

Spectroscopic and structural probes sensitive to the dynamic structure of IQTPs in static and optically pumped states on sub-picosecond time, nanometer length, and meV-eV energy scales.

What are the structural and electronic properties of cuprates and UO2 that cause them to host IQTPs?

Via our collaborator Dr. Ross Springell at the University of Bristol we can obtain single crystal PVD films of UO_{2+x} for any composition and orientation and with varying degrees of strain induced by the substrate. These films are typically < 100 micrograms, well under the amount where their activity exceeds the noise level.

Characterization of the materials

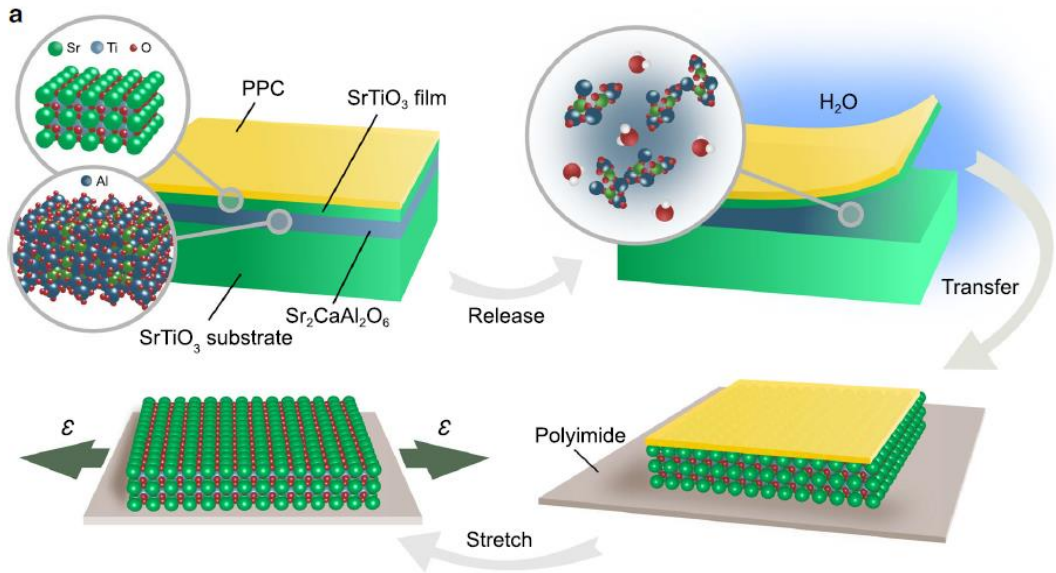
- We will study UO_2 chemically doped with O and photoexcited with controlled composition, and two cuprates $YBa_2Cu_3O_{6.5}$ and $Sr_2CuO_{3.3}$.

- We will also fully characterize these materials systems employing different laser fluences, pulse delays and sample temperature.

- We will leverage advances made on the instrument for improved control of the electron beam (proposals UED-308073, UED-308096).

Sample preparation

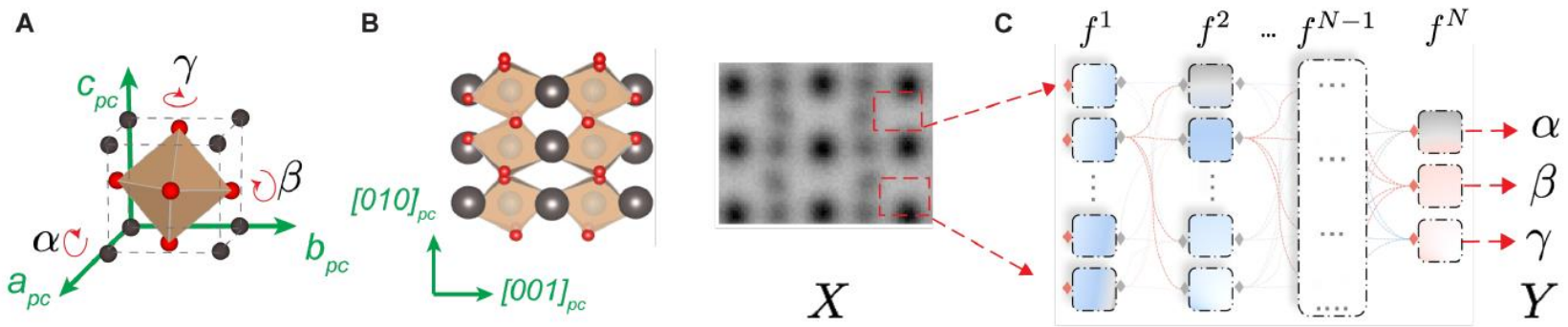
- This is a critical part of our proposal; development of sample preparation methods can extend the applications of the MUED instrument to more material systems.
- We are exploring membrane preparation techniques similar to those used by Harold Hwang, et al. that can be transferred to a wide range of materials.
- In this work, a sacrificial layer of $Sr_2CaAl_2O_6$ was employed which is later dissolved in deionized water.
- The PPC layer used to transfer the membrane is removed by thermal decomposition in O_2.
- Another promising membrane preparation technique: *Kum, Hyun S., et al. "Heterogeneous integration of single-crystalline complex-oxide membranes." Nature 578.7793 (2020): 75-81.*



Xu, Ruijuan, et al. "Strain-Induced Room-Temperature Ferroelectricity in $SrTiO_3$ Membranes." arXiv preprint arXiv:2005.09101 (2020).

As Mariana already said on Monday…

- We will automate of the process for real-time analysis.
- Analysis of collected diffraction patterns with software available at BNL.
- *This is aligned with goals and offline experiments from proposal UED-308073 (M. Fazio).*
- We will also implement artificial neural networked based models to extract relevant information and predict the materials structure.
- Relevant previous work on STEM by Nouamane Laanait (ex ORNL) et. al.:



Laanait, Nouamane, Qian He, and Albina Y. Borisevich. arXiv preprint arXiv:1902.06876 (2019).

As Mariana already said on Monday…

- All collected diffraction patterns will be stored in a newly created database (see UED-308073).
- Team members at LANL have a ‘materials genome-like’ effort under the auspices of the Institute for Materials Science.
- They also have a data repository through the LANL library that they use for all of the LANL Office of Science Data Management Plans (DMPs).
- We will develop a long-term data storage strategy and implementation for archival storage.
- A mirror of the data will be on the Argonne Leadership Computing Facility where this materials data will be used to build models of various material genres to assist with the automated data analysis system based around machine learning.