Real-time Integration Tool for the Comparison of ARPES Data and DFT Band Structure Predictions

Anna Capuano
2022 PARADIM REU Intern @ Cornell
Intern Affiliation: Department of Materials Engineering, Rensselaer Polytechnic Institute

Program: 2022 Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials Research Experience for Undergraduates Program at Cornell University (PARADIM REU @ Cornell)
PARADIM REU Principal Investigator: Prof. Darrell Schlom, Materials Science and Engineering, Cornell University PARADIM REU Mentor: Dr. Brendan Faeth, Cornell University
Primary Sources of PARADIM REU Funding: Support for PARADIM is provided under NSF Cooperative Agreement No. DMR-2039380 as part of the Materials Innovation Platform Program and support for the PARADIM REU is provided under NSF Cooperative Agreement No. DMR- 2150446.
Contact: anna28cap@gmail.com, bdf53@cornell.edu, schlom@cornell.edu
Website: https://www.paradim.org/reu/cornell

Abstract:
Angle Resolved Photoemission Spectroscopy (ARPES) is a premier tool in condensed matter physics for studying quantum materials due to its ability to directly measure momentum-resolved electronic band structure and Fermi surfaces. As a result, ARPES has been a significant aid in the study of high-temperature superconductors, graphene and topological electronic materials. A crucial part of collecting ARPES data is the ability to compare it to theory. Comparing ARPES data to theory is currently inefficient due to both complications in finding the out-of-plane momentum, which leads to the inability to quickly determine location in momentum space during calculations, and the time taken by geometry dependent calculations in order to make the two data formats compatible for comparison. To address this problem we developed and benchmarked an integrated ARPES and Density Functional Theory (DFT) comparison tool that allows the user to rapidly compare the data sets, while also viewing their location in 2D and 3D momentum space.

Introduction:
In a standard ARPES experiment, a spectrometer measures the emission angle and kinetic energy of electrons photoemitted from a sample following excitation from an ultraviolet (UV) photon beam. The crystal momentum in the x and y direction as well as the binding energy can be calculated easily from the emission angles and kinetic energies collected using Equations 1 and 2 below.

$$k_x = \sqrt{(2*(m_e))/\hbar} * \sqrt{KE} * \cos(Q) * (-\cos(\omega) * \sin(\theta) - \cos(\phi) * \cos(\theta) * \sin(\omega)) + \sin(Q) * \sin(\phi) * \sin(\omega)$$

Equation 1.

$$k_y = \sqrt{(2*(m_e))/\hbar} * \sqrt{KE} * \cos(Q) * (-\cos(\phi) * \cos(\theta) * \cos(\omega) + \sin(\theta) * \sin(\omega)) + \sin(Q) * \cos(\omega) * \sin(\phi)$$

Equation 2.

In the equations above, k_x and k_y represent the momentum in the x and y direction respectively, KE is kinetic energy and Q is the analyzer measurement angle. The out-of-plane momentum is found using the inner potential rate in Equation 3 below,

$$k_z = \sqrt{(0.262468 * (KE + v_0)) - (k_x^2) + (k_y^2)}$$

Equation 3.

where v_0 represents the inner potential rate. To determine the out-of-plane momentum, the out-of-plane momentum must be tuned by changing the photon energy. Unfortunately, the lab-based plasma-discharge lamps typically available as UV photon sources only provide a few discrete photon energies corresponding to different plasma emission lines. This means that without the ability to quickly compare the ARPES measurements being taken with theory it becomes a challenge to determine the current location in momentum space and therefore difficult to study specific features in the band structure, specifically for 3D materials. Creating an integration tool that would allow the user to easily and quickly compare the computed band structures from theory to ARPES data collected in real time directly would greatly optimize the collection of ARPES data by creating the ability to reference the location of the cut through the Fermi surface of ARPES data in 3D momentum space while taking measurements.

Methods:
We used Igor Pro Software to add an ARPES and DFT comparison tool to a previously created PARADIM ARPES data collection tool called BlueZone. BlueZone shows the current cut in reference to the Brillouin zone when provided with angles or momentums. To begin this project we augmented BlueZone into BlueZone3D in order to

show the same cut in 3D momentum space when provided with either the inner potential (V_0) or out-of-plane momentum. $SrVO_3$ was used as the material for the benchmark study of this tool due to its cubic symmetry, meaning that the electronic states share 3D behavior. The ARPES data used for the comparison was previously collected and provided by Brendan Faeth. We converted the data to momentum space using BlueZone3D. The theory used for the comparison was DFT data provided by theorists Betul Pamuk and Sri Gudivada, who used Quantum Espresso software to simulate the electronic structure of $SrVO_3$. We converted the DFT data into momentum-space using BlueZone3D.

Results:

We were able to make real time comparisons between converted momentum energy space ARPES data and DFT data for Helium and Krypton photon energies (He-I 21.2 eV, Kr-1 10.03 eV). Using BlueZone3D to determine where the cut was in momentum space, we documented two comparisons for both photon energies. The first is cut A, which refers to the cut directly in the middle of the zone and the second is cut B, which refers to the cut outside of the zone. The comparisons shown in Figure 1 and 2 show the Helium photon energy. This photon energy is the cut in the middle of the $SrVO_3$ Fermi surface. This means that for cut A, a band should be visible since it is a cut through the middle of the surface. Since cut A is in the middle of the Fermi surface and Strontium Vanadate has cubic symmetry, there should also be a band visible for cut B. Figure 1 shows cut A, as seen in the BlueZone3D cut to the left of the figure and the band is visible. Figure 2 shows cut B, since the BlueZone3D cut is out of the zone and a band is still visible.

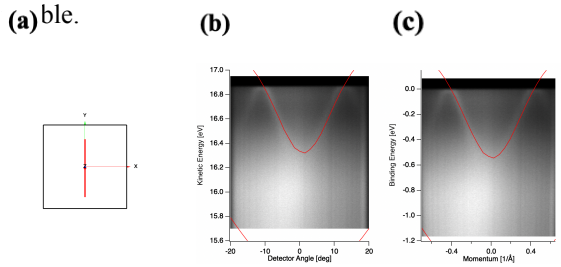

Figure 1. (a) BlueZone3D cut. (b) Detector Angle vs Kinetic Energy and (c) Momentum vs Binding Energy ARPES to DFT comparison of Helium Photon Energy of Strontium Vanadate at θ of 0 degrees, Φ of -0.75 degrees, Kinetic Energy of 12.866 eV and inner potential rate of 10. Red solid lines indicate the extracted DFT band dispersions.

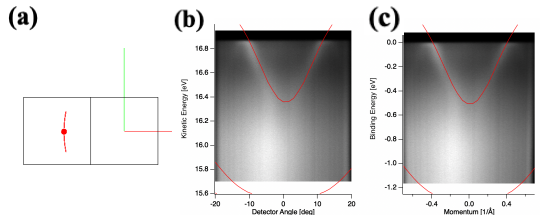

Figure 2. (a) BlueZone3D cut. (b) Detector Angle vs Kinetic Energy and (c) Momentum vs Binding Energy ARPES to DFT comparison of Helium Photon Energy of Strontium Vanadate at θ of 50 degrees, Φ of 0.75 degrees, Kinetic Energy of 12.866 eV and inner potential rate of 10. Red solid lines indicate the extracted DFT band dispersions.

The comparisons shown in Figure 3 and 4 show the Krypton photon energy. This photon energy is the cut on the edge of the $SrVO_3$ Fermi surface. This means that for cut A, a band should be seen, since it cuts through part of the Fermi surface and for cut B, a band shouldn't be seen, since cut A is at the edge of the Fermi surface cut B will be off of the Fermi surface. Figure 3 shows cut A, as seen in BlueZone3D and with the band visible. Figure 4 shows cut B, since the BlueZone3D cut is out of the zone and a band is not very visible.

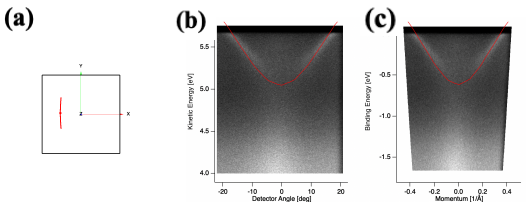

Figure 3. (a) BlueZone3D cut. (b) Detector Angle vs Kinetic Energy and (c) Momentum vs Binding Energy ARPES to DFT comparison of Krypton Photon Energy of Strontium Vanadate at θ of 12 degrees, Φ of -0.25 degrees, Kinetic Energy of 5.662 eV and inner potential rate of 10. Red solid lines indicate the extracted DFT band dispersions.

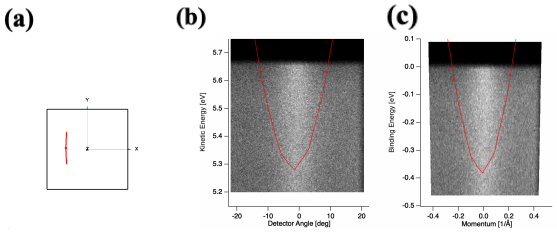

Figure 4. (a) BlueZone3D cut. (b) Detector Angle vs Kinetic Energy and (c) Momentum vs Binding Energy ARPES to DFT comparison of Krypton Photon Energy of Strontium Vanadate at θ of 45 degrees, Φ of -1.25 degrees, Kinetic Energy of 5.662 eV and inner potential rate of 10. Red solid lines indicate the extracted DFT band dispersions.

Future Work and Conclusion:

It is now possible to compare the ARPES and DFT data in real time, but it is still a challenge to find the necessary out-of-plane momentum that would create a fit for all of the photon energies. To make this next step possible, a new capability must be added to the software suite that allows for the viewing of multiple sets of data next to each other. This will allow data to have their parameters tuned at the same time, in order to find an out-of-plane momentum fit.

Acknowledgments:

An enormous thank you to my mentor Brendan Faeth for his guidance through this project. Thank you to my PI, Darrel Schlom and REU program organizer, James Overheiser for making this REU happen. Lastly, thank you to the National Science Foundation for supporting this work under cooperative agreements No. DMR- 2039380 and No. DMR-2150446.

References:

"Igor Pro Version 8 ." *WaveMetrics*, 26 Feb. 2019, https://www.wavemetrics.net/doc/igorman/IgorMan.pdf.