Simulating ARPES Measurements of Quantum Materials

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Abstract:

Angle-resolved photoemission spectroscopy (ARPES) is frequently used in solid-state physics to probe the electronic structures of materials. The calculation of ARPES intensity, however, is a nontrivial matter, as the intensity depends on factors such as the polarization of incident light, the experimental geometry, and the orbital character of the energy bands. The recently developed Python package *chinook* provides a computational framework for calculating this intensity given the tight-binding model of a material. The simulation of ARPES measurements using this software would allow for direct comparisons between theoretical band structure calculations and experimental ARPES results. Here we outline a procedure for adapting tight-binding models from the computer program Wannier90 for use in *chinook*, and we present the results from an ARPES simulation performed in *chinook* using this approach.

Introduction:

The primary input for *chinook* is a tight-binding model, which can be defined through the specification of an orbital basis and a tight-binding Hamiltonian [1]. The orbital basis used in *chinook* can be expressed as:

$$\phi_a(r,\theta,\phi) = R_a(r) \sum_{lm} c_{lm}^a Y_l^m(\theta,\phi), \quad (1)$$

where each c_{lm}^a is a complex-valued constant. Given this basis, the Hamiltonian can be written as:

$$H_{ij}(\vec{k}) = \sum_n t_{ij}^n e^{i\vec{k}\cdot\vec{R}_{ij}^n}, \quad (2)$$

where i and j are the indices of the corresponding basis states. Within this framework, the user has the option of specifying the radial wavefunctions $R_a(r)$, the spherical harmonics projections c_{lm}^a, the hopping parameters t_{ij}^n, and the connecting vectors $\vec{R}_{ij}^n$. Using this tight-binding model, the software can then calculate the photoemission intensity given the parameters of an ARPES experiment.

Summary of Research:

Density functional theory (DFT) calculations for CoSe were performed with the Quantum Espresso package [2], and the corresponding tight-binding model was constructed using Wannier90 [3]. The hopping parameters and corresponding connecting vectors were written to a data file, which could then be opened and read with Python for use in *chinook*.

Specifying the orbital basis, however, was not as straightforward. Wannier90 uses a basis of maximally localized Wannier functions; for each of the orbitals, a 3D grid containing the value of the function at each point in real space can be written to a data file, but this grid must be converted into the form of Eq. (2) in order to be used in *chinook*. Although strictly speaking the Wannier functions cannot be expressed analytically, we sought to calculate an approximate form of each orbital for the purposes of our ARPES simulations.

To accomplish this, we implemented Misner's algorithm [4] for calculating the spherical harmonic decomposition of a function at some fixed radius given a grid of points. For each of the data grids exported from Wannier90, we performed this calculation over a range of radii, starting from the center of the orbital and extending to the edge of the grid; for each value of the radius, we also computed the average absolute value of the Wannier function over the corresponding spherical shell. After normalizing each set of spherical harmonic
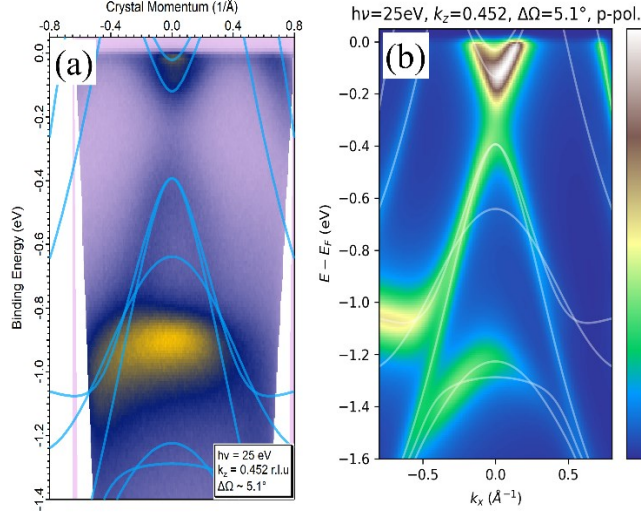
Figure 1: Comparison between the experimental (a) and simulated (b) ARPES spectra along the same cut in momentum space for CoSe, with the calculated band structure plotted on top.

coefficients, we took the weighted average of the decompositions, using the average absolute values of the function as the weights. This yielded approximate values for the c_{lm}^a coefficients in Eq. (2).

We were then able to calculate the radial wavefunctions $R_a(r)$ by simply rearranging Eq. (2):

$$R_a(r) = \frac{\phi_a(r, \theta, \phi)}{\sum_{lm} c_{lm}^a Y_l^m(\theta, \phi)}. \qquad (3)$$

By iterating over the same range of radii as before and computing the average value of $R_a(r)$ over each spherical shell, we were able to reconstruct a numerical form of each radial wavefunction, which could then be passed into *chinook*.

Results and Conclusions:

An example of an experimental ARPES measurement and the corresponding simulated result for CoSe are provided in Fig. 1. In both Fig. 1a and Fig. 1b, the band structure has been shifted upwards by 75 meV to better fit the experimental intensities; however, even after this shift, the bright spot at ~-0.9 eV in Fig. 1a does not appear to match any of the calculated bands. Although the origin of this intensity is not immediately obvious just from looking at Fig. 1a, the simulated result in Fig. 1b suggests that this bright spot corresponds to the two lower bands at ~-1.3 eV in the calculated band structure. The discrepancy in the locations of these bands can likely be explained by the failure of DFT to account for orbital-dependent electronic correlations. This example demonstrates how the simulation of ARPES measurements can facilitate the interpretation of experimental results and offer insight into the limitations of our theoretical calculations.

Future Work:

Although we were able to obtain useful results for CoSe using the outlined procedure, we do not expect this approach to be universally applicable. We therefore intend to repeat this process for a range of other materials in order to determine which classes of materials can be accurately simulated using this approach. In the long term, we hope to implement a set of scripts in Python to streamline the process of importing tight-binding models from Wannier90 into *chinook*.

References:

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