Semi-automatic Identification and Quantification of Point Defects on Quantum Materials from Atomic-resolution Electron Microscopy Data

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Abstract: Quantum materials have the potential to revolutionize the world of electronics, however, much is still unknown about the phenomena that induce their exotic properties. Understanding how a material's structure impacts the properties of interest requires close analysis of its nanoscale structure. Here, we use scanning transmission electron microscopy data to characterize the low symmetry C2/m atomic scale structure of La3Cd2As6, which exhibits a vacancy ordering in the cadmium sites. This system is of special interest because it was found to be a narrow-gap semiconductor with a band width of 105 meV. Disorder-free narrow-gap semiconductors are of interest because of their potential in fields such as thermoelectricity and dark matter detection. Furthermore, the La3Cd2As6 system showed a thirteen order of magnitude increase in electrical resistivity upon cooling which suggests that the system has a remarkably clean insulating ground state. In this work, a semi-automated analysis procedure is presented consisting of identifying the cadmium vacancy sites, measuring their image intensity to identify whether there are atoms occupying these sites, and then comparing the intensities in the vacancy sites with the neighboring cadmium sites to quantify the degree of partial occupancy. Identifying these point defects may help understand the origin of the material's remarkable electronic properties.

Methods: *Finding the Cadmium Atomic Rows:* The first step in the analysis process is to locate the atomic row of interest, in this case of the $La_3Cd_2As_6$, the cadmium atomic row. To facilitate processing, I began by rotating the HAADF STEM image so that the atomic rows ran vertically. Next utilizing a python package "photoutils" I implemented atom tracking and row finding functions to identify the rows of the largest atoms in the system. Once these were recognized I used relative pixel positions to identify the start and end point locations of the atomic rows of cadmium, the element of interest.

Analyzing Line Profiles and its Challenges: The recognition of a partial occupancy is illustrated by the instance of a peak where we expect a trough. To find instances of partial occupancies I used existing peak finding functions to recognize the peaks within a certain desired range. Using a set threshold to find instances of partial occupancies was problematic because the average intensity of the peaks changed as we advanced through the image. To overcome this challenge, I designed a method to adjust the range threshold for each line profile that considers the difference in sample thickness across the sample.

Quantification: In the analysis, the motivation for the quantification of the partial occupancy is to see if we were able to recognize patterns across multiple images of a sample that might give us insight into the rise of this material's exotic electronic properties. To do so a normalization was necessary to counteract the impact of the increasing thickness across an image. To overcome this challenge, I implemented this intensity normalization formula.

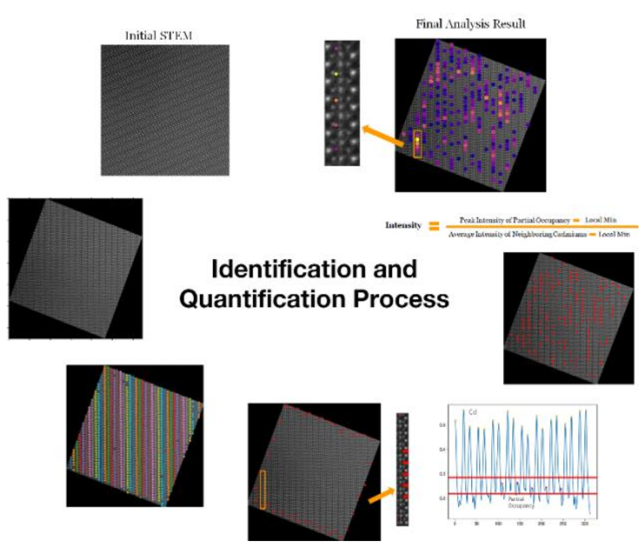

Figure 1. Illustration of the identification and quantification process

Results

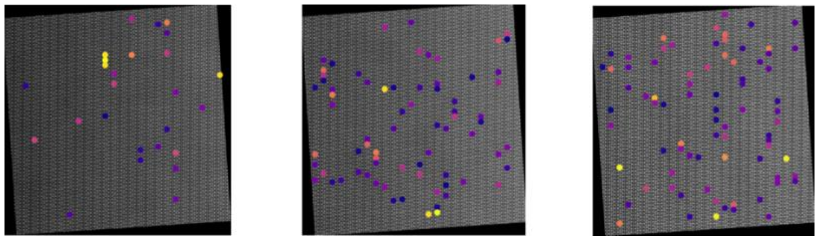

Figure 2: STEM data of the La3Cd2As6 system after its partial occupancy sites have been identified and quantified, ordered in increasing thickness sample.

After a close examination of the quantification trends, we saw that the instances of partial occupancies increase as the thickness of the sample increased. This trend correlates to initial intuitions about interstitial point defects because we can see the impact of more atomic layers of the sample. However, to really understand how this can possibly correlate to physical properties more research and analysis is needed.

Conclusions

After designing a semi-automatic system with replicable results across a multitude of HAADF STEM data sets. We found that there is a quantifiable increase in clustering of partial occupancies as the thickness of a sample increases. As well as a significant presence of point defects was observed despite this material being a clean insulating ground state. This goes against the general intuition that a good insulating ground state is free of point defects and is a subject of further research.

Future Work

Continue to study the relationship between partial occupancies and the rise of exotic electronic properties by conducting similar analysis.

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