

# Electronic Structure of Cs Doped and Undoped FeSe Thin Films

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*Program: 2021 Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials Research Experience for Undergraduates Program at Cornell University (PARADIM REU @ Cornell)*

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*Primary Source of PARADIM REU Funding: Support for PARADIM is provided under NSF Cooperative Agreement No. DMR-2039380 as part of the Materials Innovation Platform Program.*

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## Abstract:

FeSe is a superconducting material whose critical temperature ( $T_C$ ) increases eight-fold when grown as a monolayer on the substrate SrTiO<sub>3</sub> (STO)<sup>1</sup>. The interfacial interaction between FeSe and STO is comprised of multiple effects, any of which could affect the critical temperature. Understanding why this change in  $T_C$  occurs would provide insight into how to increase the  $T_C$  of other superconductors. To work towards understanding this phenomenon, we decided to investigate the effect of electron doping (one of the interfacial effects) on the band structure and superconductivity of FeSe. We doped a multilayer FeSe film with Cs, introducing electrons without any other interfacial effects. The band structure of the doped film was measured using ARPES. The ARPES data showed that there was a superposition of multiple doping states in the film, one of which resembled the monolayer electronic structure.

## Introduction:

Superconductors allow current to flow without any resistance. However, there is currently only one material that is superconducting at room temperature, but only under 39 million psi<sup>2</sup>.

The more that is known about superconducting materials and what affects their critical temperature, the better we can design future materials with possibly higher, maybe even room temperature, critical temperatures.

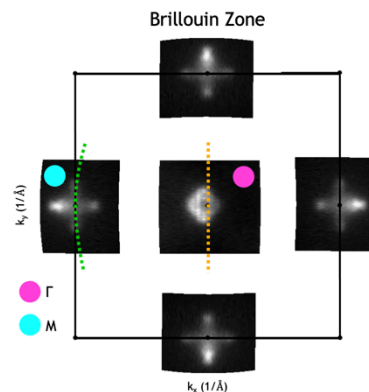
FeSe is a fascinating material to study when looking at superconductivity because of the change its  $T_C$  undergoes when the film is grown a monolayer thick on SrTiO<sub>3</sub> (STO). Bulk-like FeSe has a  $T_C$  of about 8 K<sup>1</sup>. Monolayer FeSe has a  $T_C$  about eight times greater, in the 60 –70 K range<sup>1</sup>. The interfacial interaction between FeSe and STO causes this change in critical temperature<sup>1</sup>. However, this interaction consists of a multitude of effects, so we need to piece apart what exactly causes the change in critical temperature. To do so, we need to isolate each effect and determine its impact on FeSe's electronic structure and superconductivity.

One of the effects that makes up the interfacial interaction is electron doping. There have been other successful attempts at electron doping FeSe, but the films these methods have produced are either incompatible with ARPES or produce poor quality ARPES data. Our

process creates a film that produces clean ARPES data, which we can then use to better analyze and form conclusions about how electron doping affects the electronic structure and superconductivity of bulk FeSe.

## Methods:

Multi-layer FeSe films are synthesized on SrTiO<sub>3</sub> substrates using a dedicated chalcogenide molecular-beam epitaxy (MBE) system. We started by annealing the surface of an STO substrate to make it as smooth as possible. We then loaded the substrate into the MBE, heated it to 600 °C, and degassed it overnight. The next day, we reduced the substrate to growth temperature (420 °C). Fe and Se were co-deposited on the surface for 13:20, resulting in a film that was about 20 unit cells thick. After cooling to room temperature, we deposited



**Figure 1.** A Fermi Surface Map (FSM) of the undoped film. This is a plot of intensity as a function of electron momentum at the Fermi level. The pink dot marks  $\Gamma$  and the blue dot marks M. The dotted line on each feature marks the location of the cross section through that feature.

Cs on the surface for five minutes through solid phase epitaxy, where the Cs intercalates into the holes of the FeSe lattice. We then capped the film with Se and transferred it to the ARPES system, where we took data to capture the 1-Fe UC. This data included Fermi surface maps (FSMs) at  $\Gamma$  and M and cross-section slices through the middle of those features (Fig. 1). We monitored the quality of the substrate and film throughout the growth process using RHEED.

## Results and Discussion:

At  $\Gamma$ , there is a third band,  $\beta'$  in Fig. 2(f), that appears in the Cs doped samples that is not present in the undoped sample. This band is not caused by electron doping. It is caused by the superposition of the undoped band structure onto the doped band structure.

The superposition becomes clearer when we look at  $\Gamma$  differently. The raw data [Fig. 2(a, d)] for both samples looks similar, but the third band is not clearly present. After processing this data, we can see the third band, in light blue, at the lower energies of our doped sample [Fig. 2(c)]. We can also see that the outermost band is almost identical in the undoped and doped data, implying a superposition of the undoped data onto the doped.

Although they are the same at  $\Gamma$ , the two Cs doped samples have different band structures at M [Fig. 3(b, c)]. This is due to superposition, but this time the number of band structures being superimposed differs. In sample A, there are two superimposed band structures: the undoped and a lower doped band structure. Sample B has a third, higher doped band structure superimposed on it as well. The higher doped band structure is where the additional band comes from in sample B that is not in sample A [Fig. 3 (b, c)]. We can also see that the pink band in sample B [Fig. 3(c)] caused by the higher doping mirrors closely the dispersion of the monolayer film [Fig. 3(d)].

To quantify the electron doping in the samples, we used Eq. (1), which relates the area encompassed by the band to the additional electrons/UC. The higher doped phase of sample B has about the same doping as the monolayer FeSe [Fig. 4(c, d)].

$$\text{Electrons/UC} = \frac{2A}{4 * (\frac{\pi}{3.88})^2} = \frac{A}{1.3112} \quad (1)$$

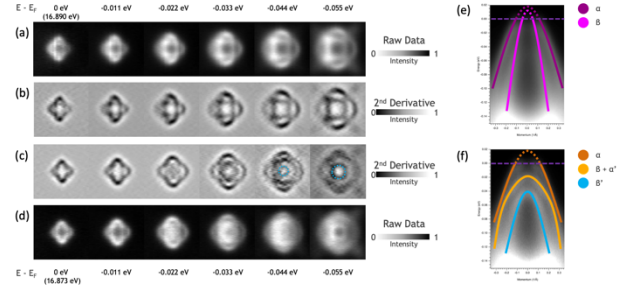
## Conclusions and Next Steps:

We were able to successfully produce an electron doped FeSe film through the solid phase epitaxy of Cs. This method produced a film that gave quality ARPES data. However, because of the superposition we observed, we can infer that this ARPES data was a mixture of phases with discrete doping levels. This is why moving forward, we would like to determine if we can better control the doping to produce a uniformly doped sample with about 0.1 additional electrons/UC. The lack of superposition would create a clearer picture of how the band structure changes with electron doping,

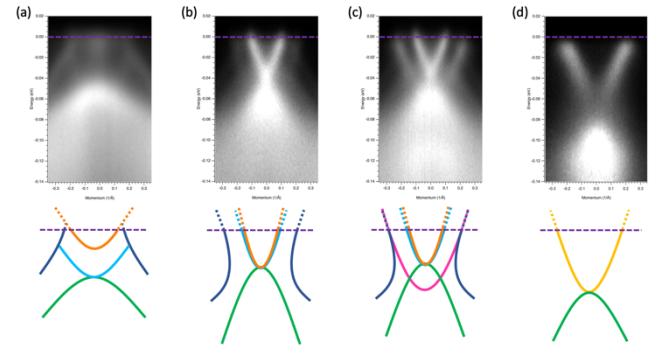
and hopefully resemble the monolayer band structure like the higher doping in doped sample B appears to.

It would also be interesting to investigate why M changes when the sample is doped different amounts, but  $\Gamma$  stays the same.

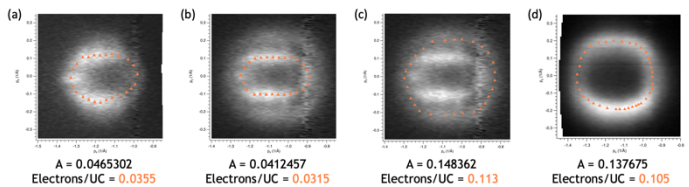
Finally, we would also like to investigate the temperature dependence of the higher doped band structure in doped sample B to determine if it has an increased critical temperature over bulk FeSe.



**Figure 2.** Breakdown of the feature at  $\Gamma$ . (a)–(d) are series of iso energy cuts at  $\Gamma$  which start at the Fermi level and decrease in energy. (a) Raw data progression of  $\Gamma$  in the undoped sample. (b) Processed raw data to accentuate the bands in the undoped sample. (c) Processed raw data for Cs Doped Sample A. The blue circle maps the dispersion of  $\beta'$ . (d) Raw data progression for Cs Doped Sample A. (e) Cross-section of  $\Gamma$  for the undoped sample. The colored lines map the band dispersion. (f) Cross-section of  $\Gamma$  for Cs Doped Sample A.  $\alpha'$  and  $\beta'$  are the shifted versions of  $\alpha$  and  $\beta$  due to electron doping.



**Figure 3.** The cross-section slice through M for 4 different samples. The colored bands below each slice map the band dispersion in the data. The purple dotted line represents the Fermi level of each sample. (a) Undoped. (b) Cs Doped Sample A. (c) Cs Doped Sample B. (d) Monolayer FeSe



**Figure 4.** Outline of the bands at M in each sample's ARPES data at the Fermi level. The size of the bands corresponds to the doping level of the sample. (a) Cs Doped Sample A. (b) Lower doped band structure in Cs Doped Sample B. (c) Higher doped band structure in Cs Doped Sample B. (d) Monolayer FeSe.

## Acknowledgements:

Thank you to Dr. Brendan Faeth for all his help and guidance on this project and for teaching me what it means to be a good scientist and communicator. Thank you to my PI, Darrell Schlom and Jim Overheiser for all their help. And thank you to the National Science Foundation for making this work possible under NSF Cooperative Agreement No. DMR-2039380.

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