Strong Correlations and Unconventional Superconductivity: Towards a Conceptual Framework， 2014/09/22--26, KITP, Santa Barbara

ARPES on Electronic Structure and Superconductivity of FeSe /$SrTiO_3$ Films

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Colleagues and Collaborators

➢ARPES System Development, Maintenance and Improvement;

Chuangtian Chen, Yong Zhu, Guochun Zhang, Xiaoyang Wang
Zuyan Xu, Guiling Wang, Hongbo Zhang, Yong Zhou
Technical Institute of Physics and Chemistry, CAS, China

➢FeSe Thin Films on $SrTiO_3$ (001)

Wenhao Zhang, Yun-Bo Ou, Qing-Yan Wang, Zhi Li, Lili Wang,
Xucun Ma and Qikun Xue, Institute of Physics, CAS & Tsinghua University

➢Theoretical Discussions

Jiangping Hu, Dung-hai Lee, Qianghua Wang, Tao Xiang, Qimiao Si

Outline

- **Distinct Electronic Structure of Superconducting Single-Layer FeSe/$SrTiO_3$ Films;**

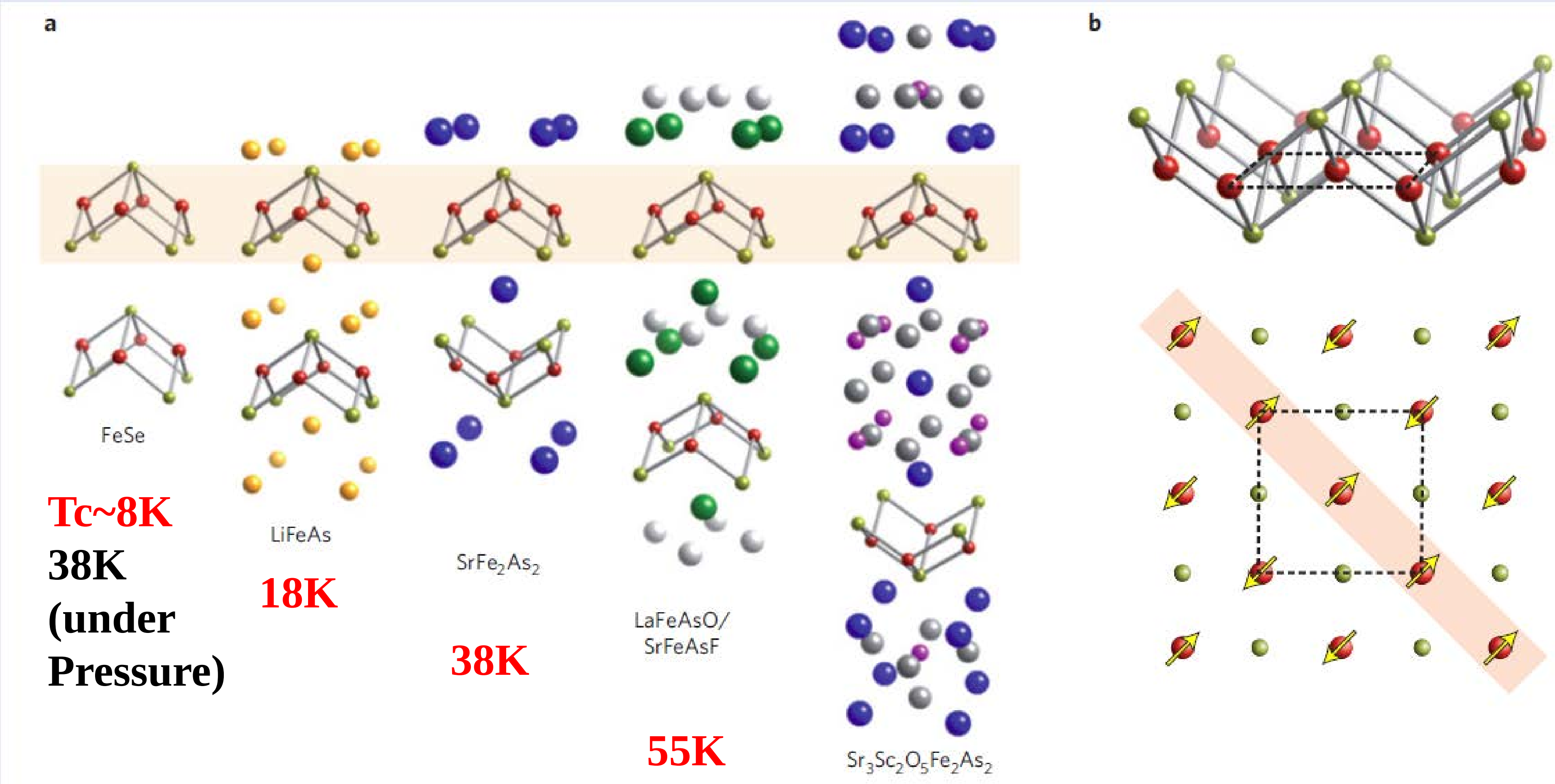
- **Electronic Phase Diagram of Single-Layer FeSe/$SrTiO_3$ Films;**

- **Evidence of an Insulator-Superconductor Transition in Single-Layer FeSe/$SrTiO_3$ Films;**

- **Dichotomy between the Single-Layer and Double-Layer FeSe/$SrTiO_3$ Films.**

Iron-Based Superconductors

“11” “111” “122” “1111” **FeAs or FeSe**

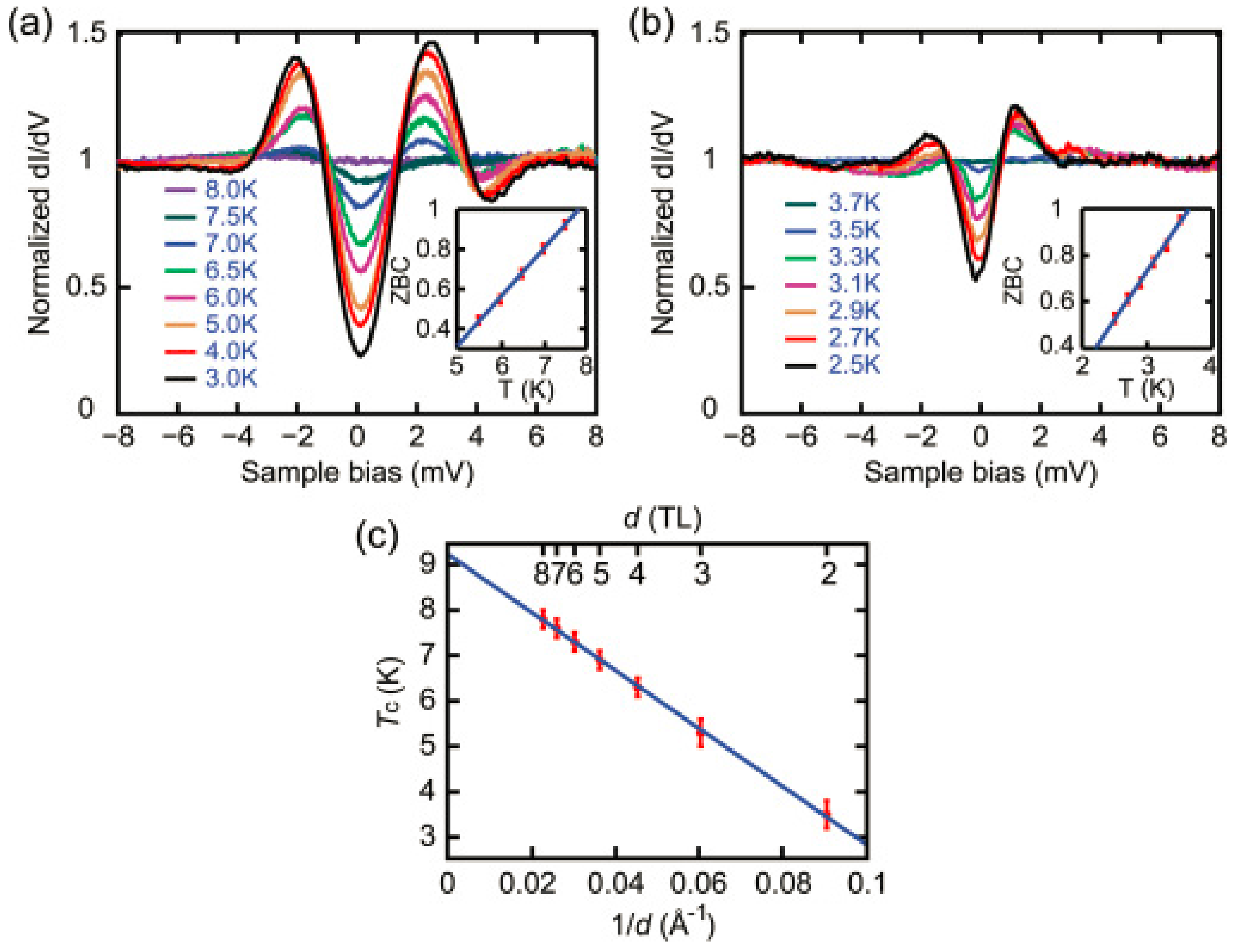


J. Paglione and R. L. Greene, Nature Physics 6(2010)645.

FeSe Films Grown on Graphene

8-Layer

2-Layer



C. L. Song et al., Phys. Rev. B 84 (2011) 020503(R).

Possible High Tc in Single-Layer FeSe Film on $SrTiO_3$

CHIN. PHYS. LETT. Vol. 29, No. 3 (2012) 037402

Interface-Induced High-Temperature Superconductivity in Single Unit-Cell FeSe Films on $SrTiO_3$ *

WANG Qing-Yan(王庆艳)[1,2†], LI Zhi(李志)[2†], ZHANG Wen-Hao(张文号)[1†], ZHANG Zuo-Cheng(张祚成)[1†], ZHANG Jin-Song(张金松)[1], LI Wei(李渭)[1], DING Hao(丁浩)[1], OU Yun-Bo(欧云波)[2], DENG Peng(邓鹏)[1], CHANG Kai(常凯)[1], WEN Jing(文竞)[1], SONG CanLi(宋灿立)[1], HE Ke(何珂)[2], JIA Jin-Feng(贾金锋)[1], JI Shuai-Hua(季帅华)[1], WANG Ya-Yu(王亚愚)[1], WANG Li-Li(王立莉)[2], CHEN Xi(陈曦)[1], MA Xu-Cun(马旭村)[2**], XUE Qi-Kun(薛其坤)[1**]

[1] *State Key Lab of Low-Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084*
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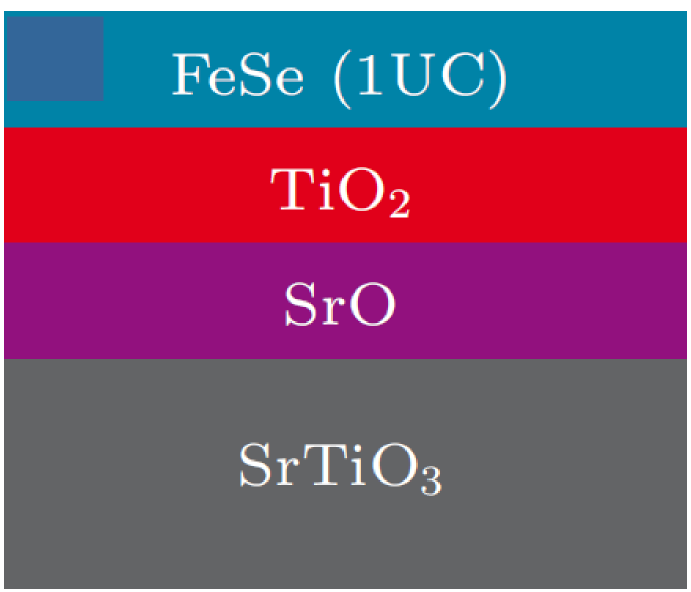
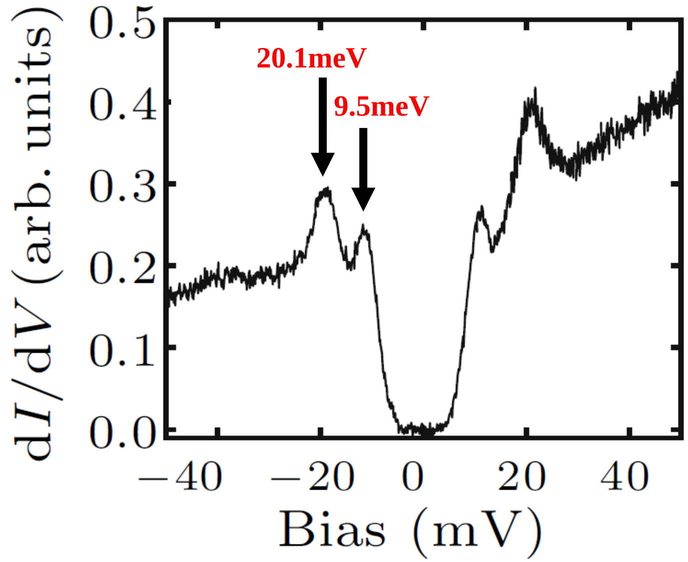
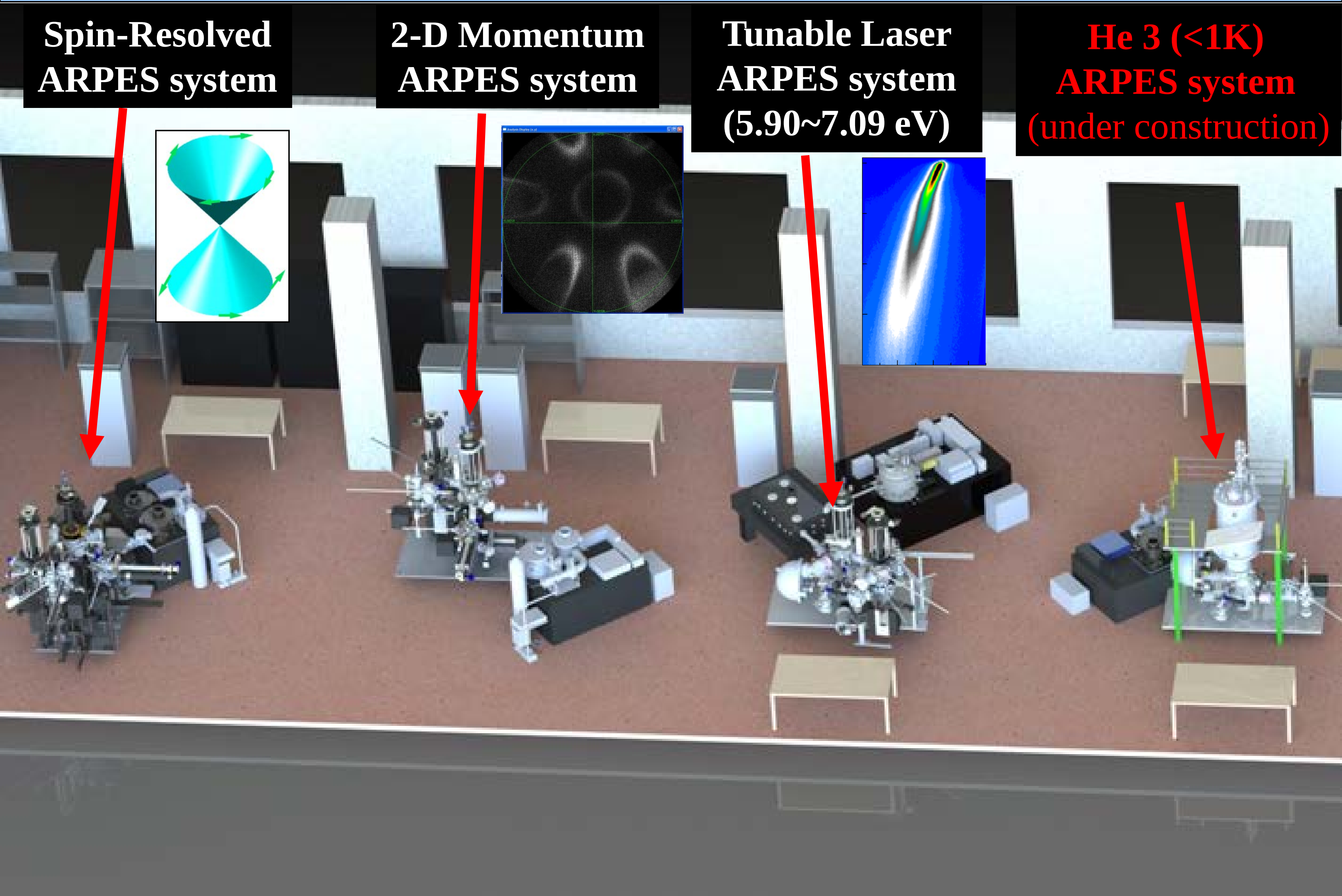
(Received 1 February 2012 and accepted by ZHU Bang-Fen)



$FeSe/SrTiO_3$, Tc ~ 80 K?



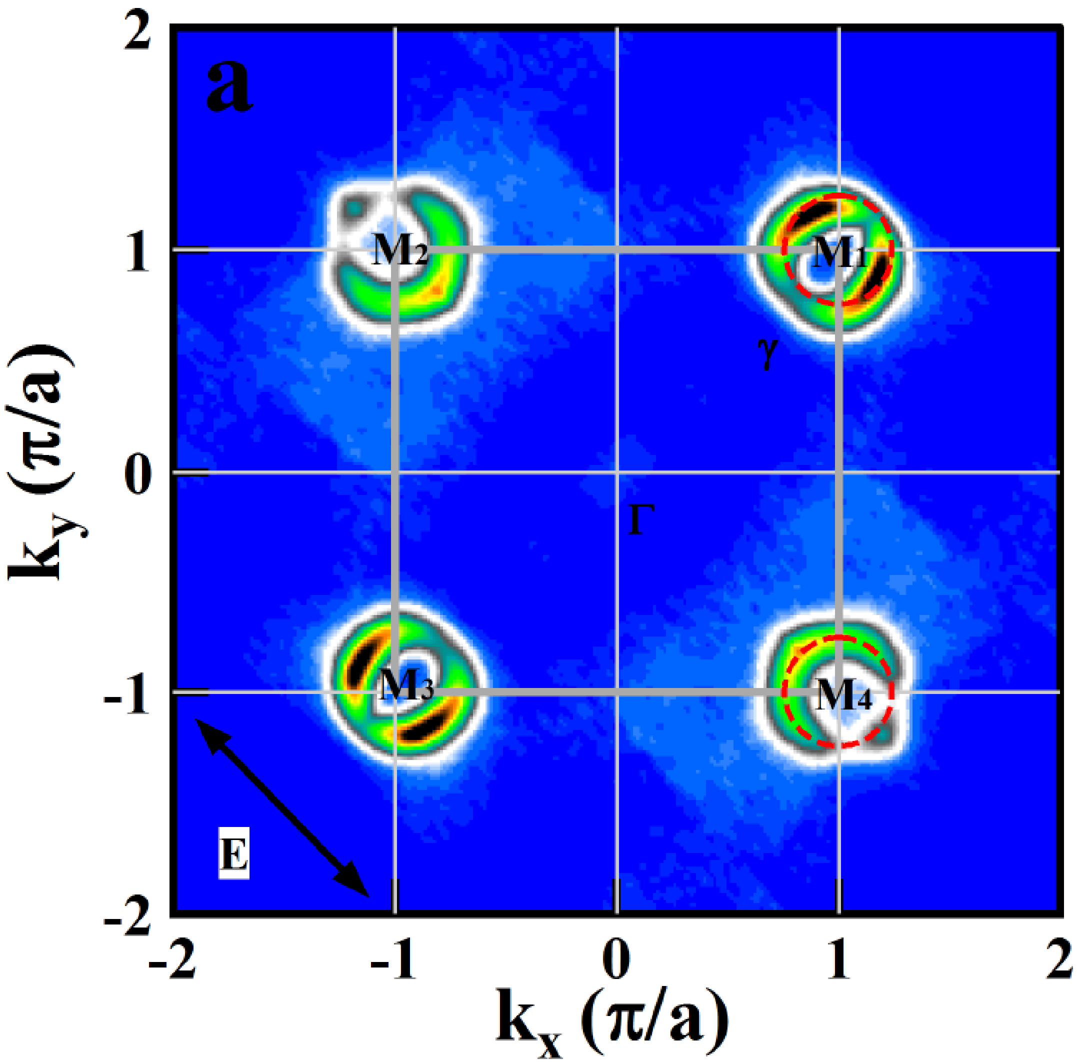
VUV Laser Photoemission Lab at IOP



ARPES on Single-Layer Superconducting $FeSe/SrTiO_3$

D. F. Liu , W. H. Zhao, D. X. Mou, J. F. He, X. C. Ma, Q. K. Xue, X. J. Zhou et al.,
Nature Communications 3, 931 (2012).

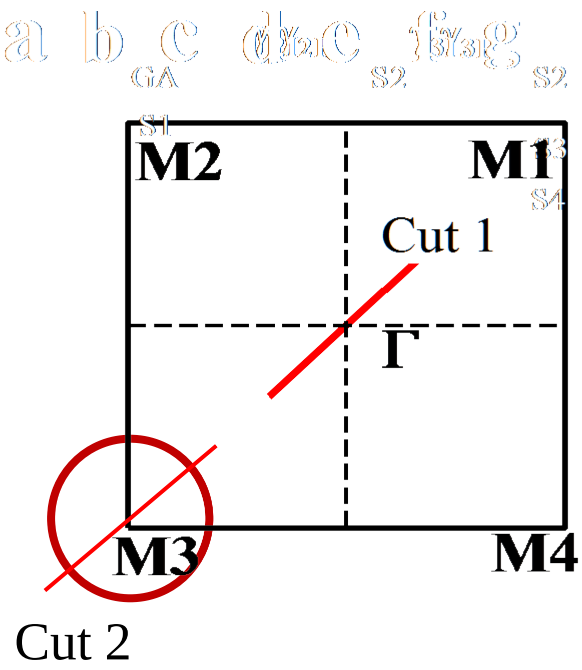
Fermi Surface of Single-Layer Superconducting $FeSe/SrTiO_3$

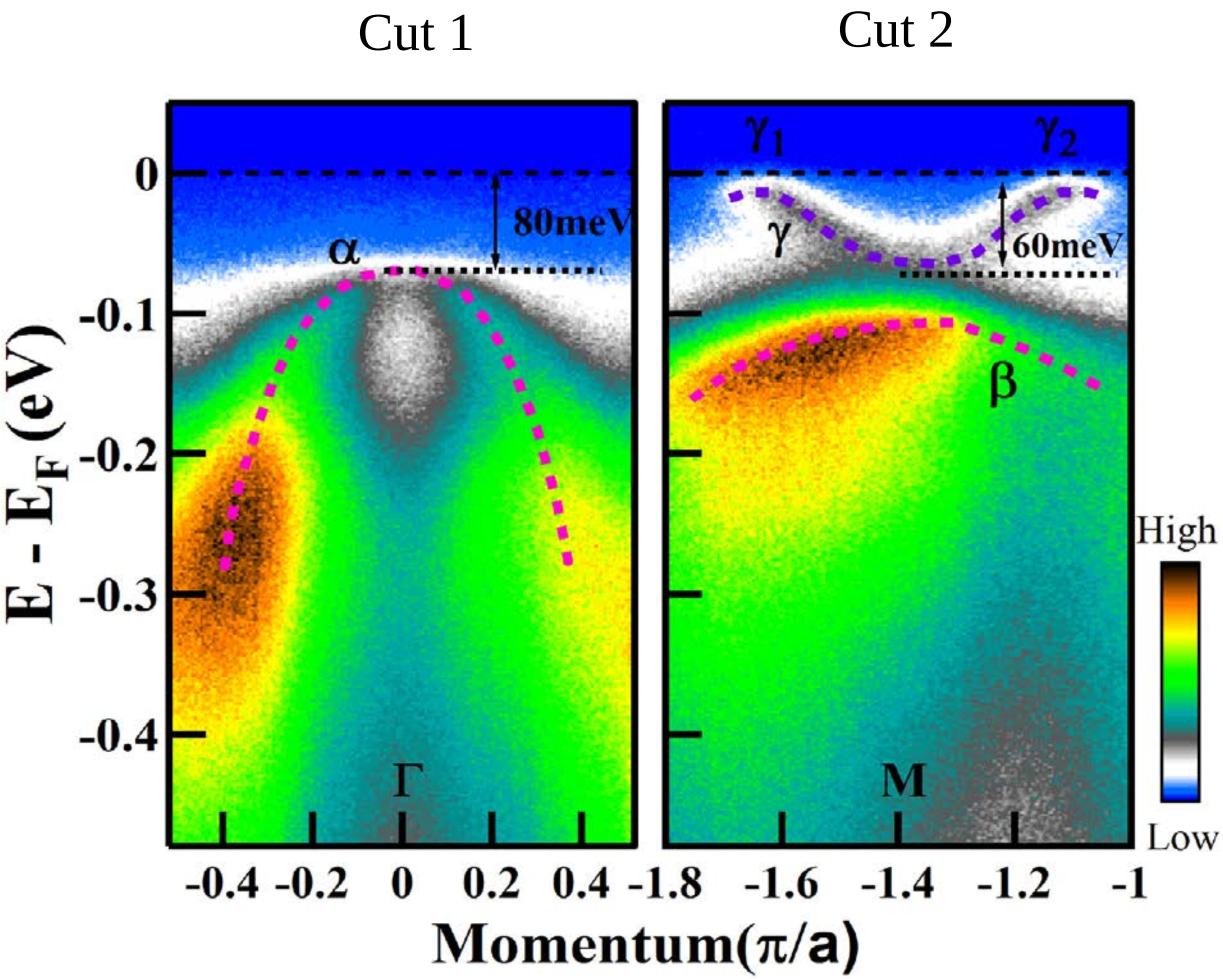


- **Only one Fermi surface γ around M;**
- **No signature of any Fermi surface around Γ.**

D. F. Liu , W. H. Zhao, D. X. Mou, J. F. He, X. C. Ma, Q. K. Xue, X. J. Zhou et al., Nature Communications 3, 931 (2012).

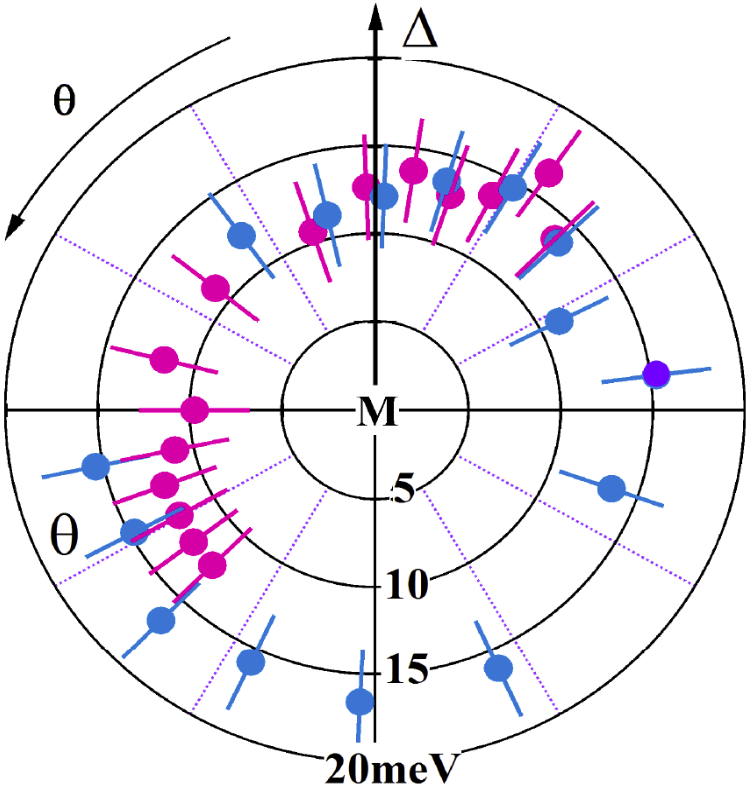
Band Structure of Single-Layer Superconducting $FeSe/SrTiO_3$

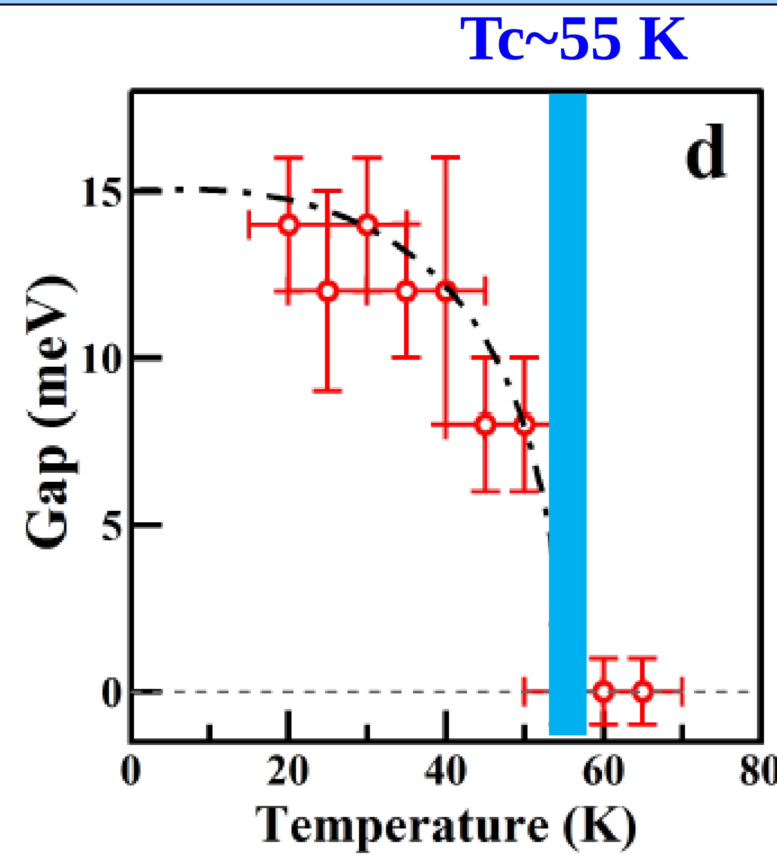




D. F. Liu , W. H. Zhao, D. X. Mou, J. F. He, X. C. Ma, Q. K. Xue, X. J. Zhou et al., Nature Communications 3, 931 (2012).

Momentum and Temperature Dependence of the Energy Gap





Nearly isotropic energy gap;

2D Character → No Gap Node.

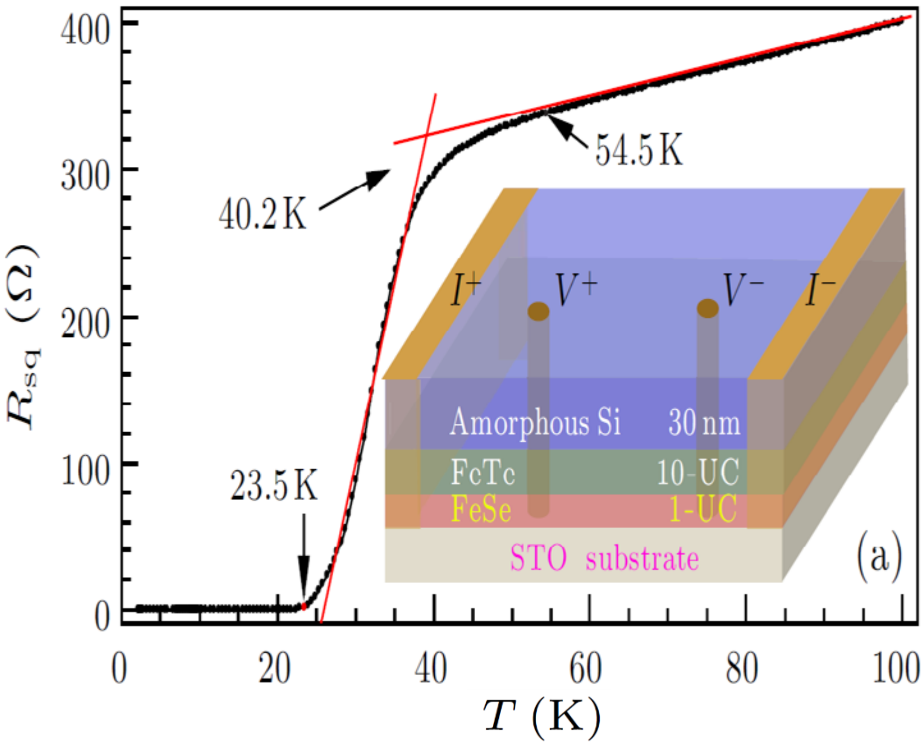
Nearly BCS form;

Tc~55K.

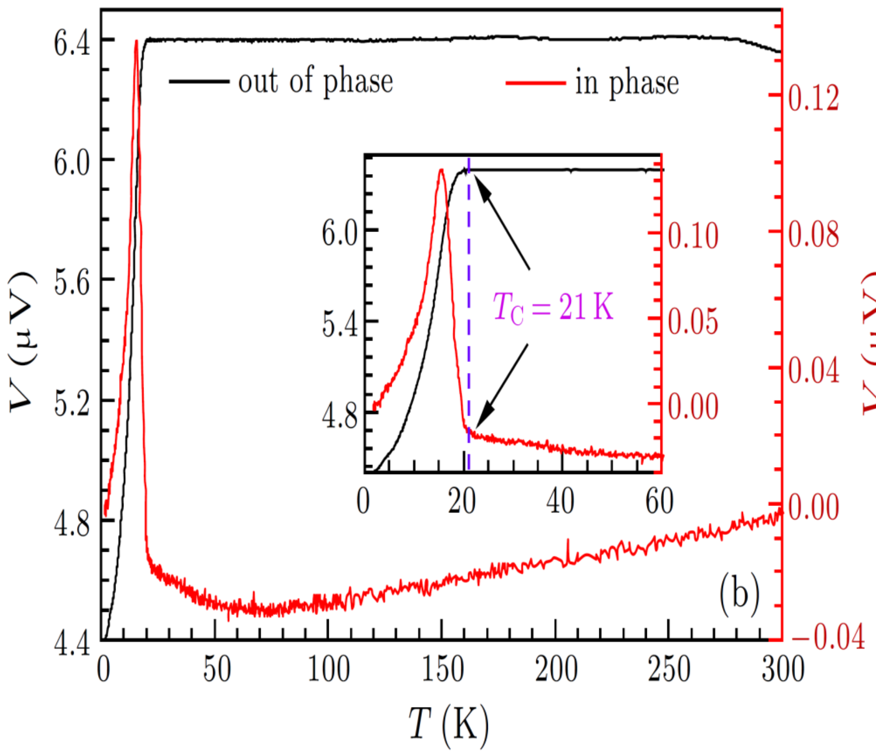
D. F. Liu , W. H. Zhao, D. X. Mou, J. F. He, X. C. Ma, Q. K. Xue, X. J. Zhou et al., Nature Communications 3, 931 (2012).

Transport and Magnetic Measurements on Single-Layer FeSe/$SrTiO_3$ Films

Resistivity



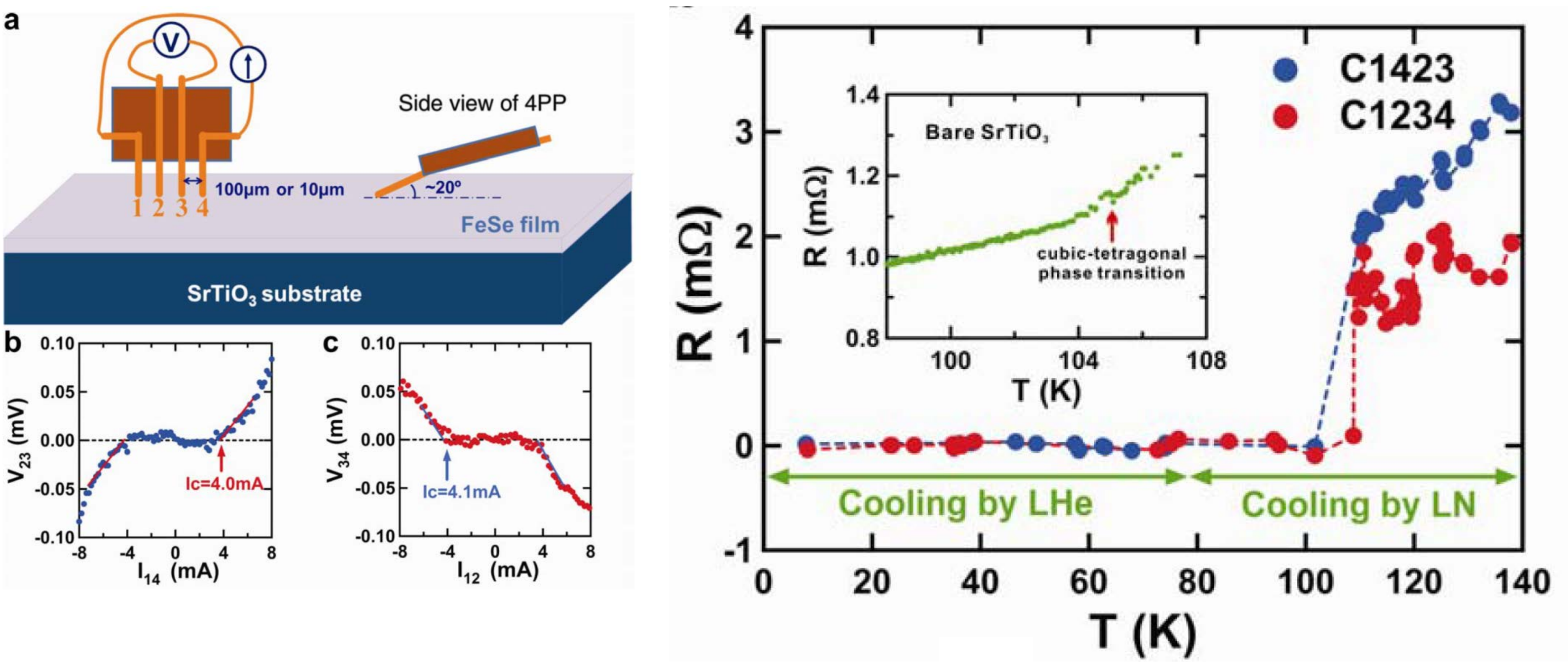
Magnetism



W. H. Zhang, J. Wang, X. C. Ma, Q. K. Xue et al.,
Chinese Physics Letters 31 (2014) 017401.

Superconductivity in single-layer films of FeSe with a transition temperature above 100 K

Jian-Feng Ge[1], Zhi-Long Liu[1], Canhua Liu[1]*, Chun-Lei Gao[1], Dong Qian[1], Qi-Kun Xue[2]*, Ying Liu[1,3], Jin-Feng Jia[1]*



arXiv: 1406.3435

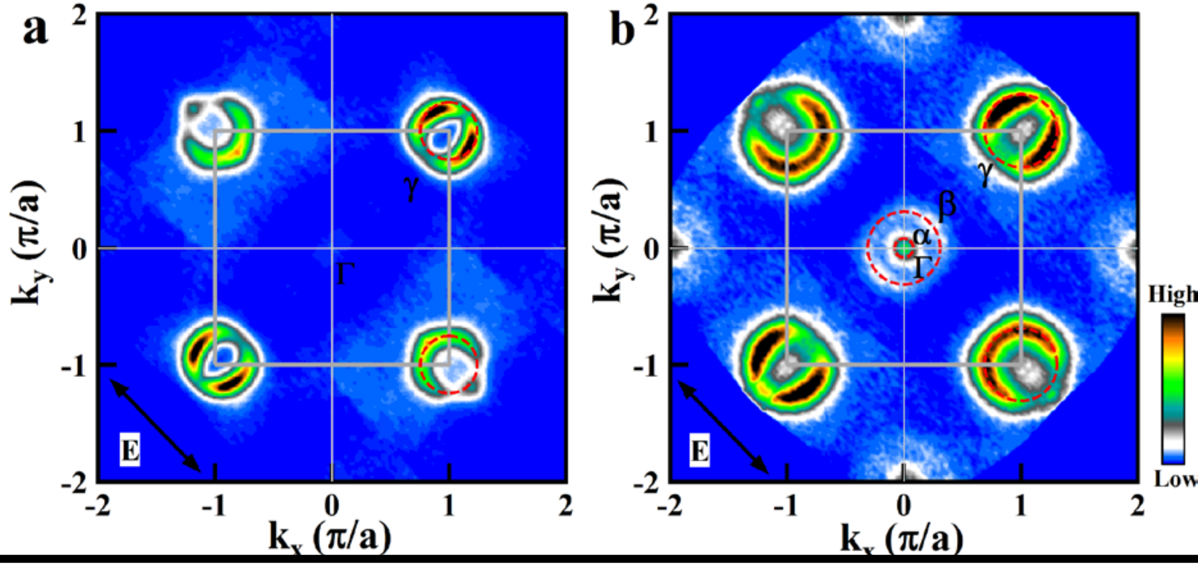
Fermi Surface Topology of the Iron-Based Superconductors

Class I

Single-Layer FeSe

Tc~65 K

D. F. Liu et al.,
Nature Commun.
3(2012)931.



$A_xFe_{2-y}Se_2$

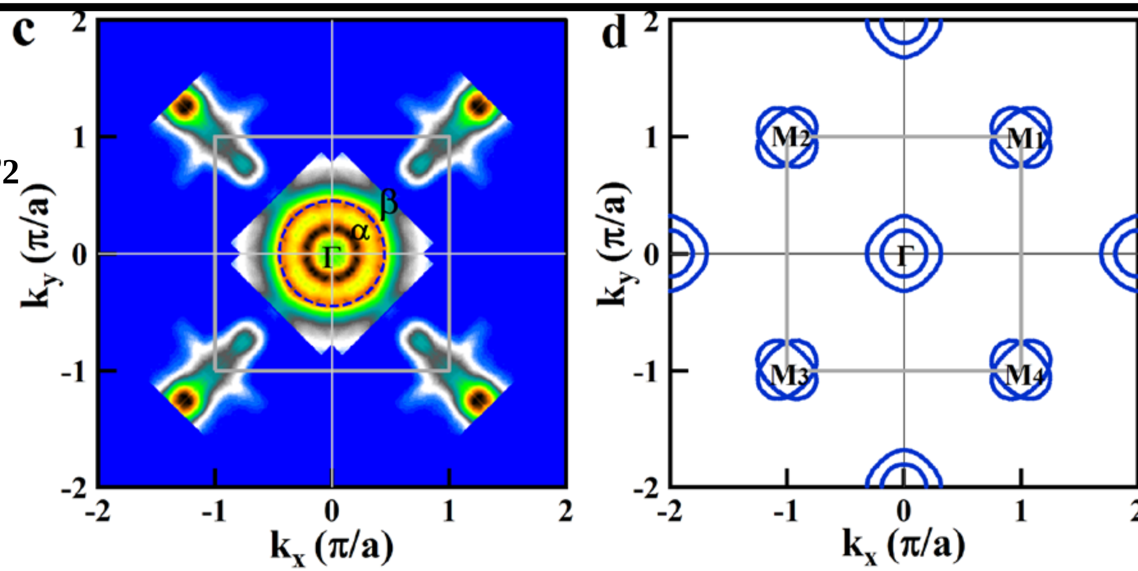
Tc~32 K

D. X. Mou et al.,
Phys. Rev. Lett.
6 (2011) 410.

Class II

$(Ba_{0.6}K_{0.4})Fe_2As_2$

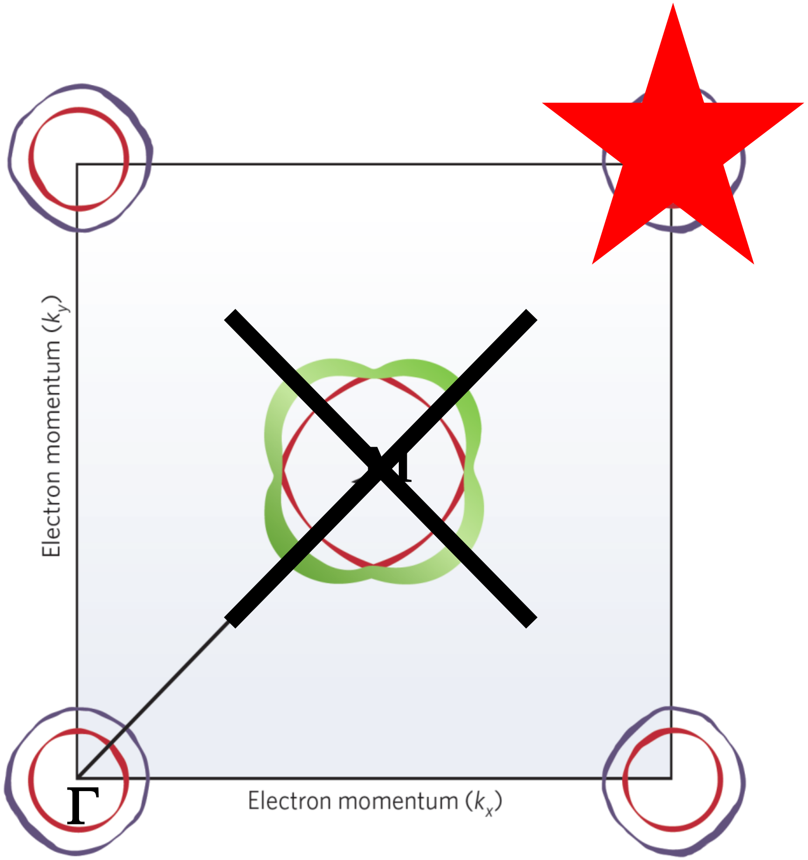
L. Zhao et al.,
Chin. Phys. Lett.
25 (2008) 4402.



Bulk Fe(Se,Te)

A. Tamai et al.,
Phys. Rev. Lett.
104 (2010) 097002.

Implications



1. **Fermi surface topology is not critical to superconductivity?**

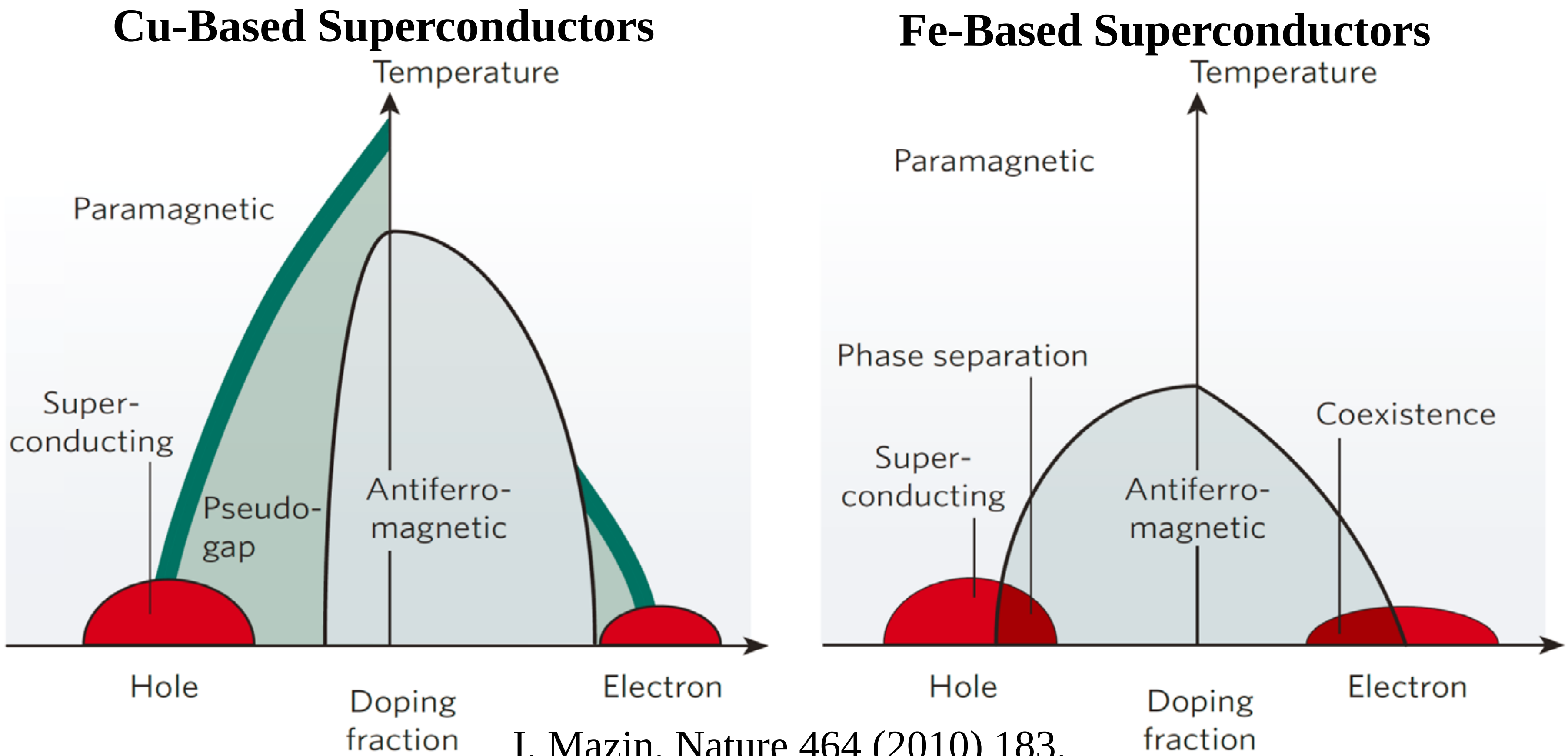
2. **If it is critical, then**
   Hole-Like Fermi Surface near Γ is NOT important;

   Electron-Like Fermi Surface near M is crucial.

Phase Diagram and Electronic Indication of Superconductivity at 65K in Single-Layer FeSe/$SrTiO_3$

S. L. He, J. F. He, W. H. Zhang, L. Zhao, X. C. Ma, Q. K. Xue, X. J. Zhou et al., Nature Materials 12, 605 (2013).

Phase Diagram of High Temperature Superconductors



I. Mazin, Nature 464 (2010) 183.

1. **Doping evolution of electronic structure in single-layer FeSe?**

2. **Can superconductivity of the single-layer FeSe be optimized?**

MBE Preparation of FeSe Films: Two Steps

MBE Film Preparation

Step 1

***As-prepared* FeSe Films**
grown at a low temperature
~300C
(Non-superconducting)

→

Step 2

FeSe Films
annealed at a high temperature
450-~500C
(Superconducting)

Q. Y. Wang et al., Chin. Phys. Lett. 29, 037402 (2012).

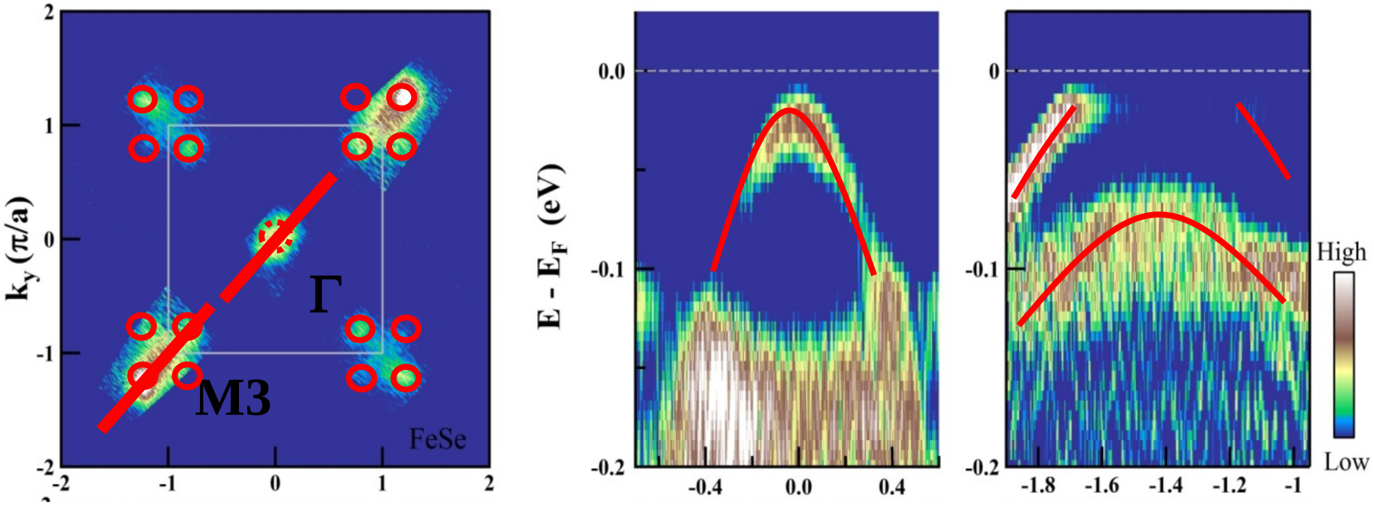
Electronic Structure of

the *As-Prepared* Non-Superconducting

Single-Layer FeSe/$SrTiO_3$ Films ……

“Fermi Surface” and Band Structure of Non-Superconducting Phase

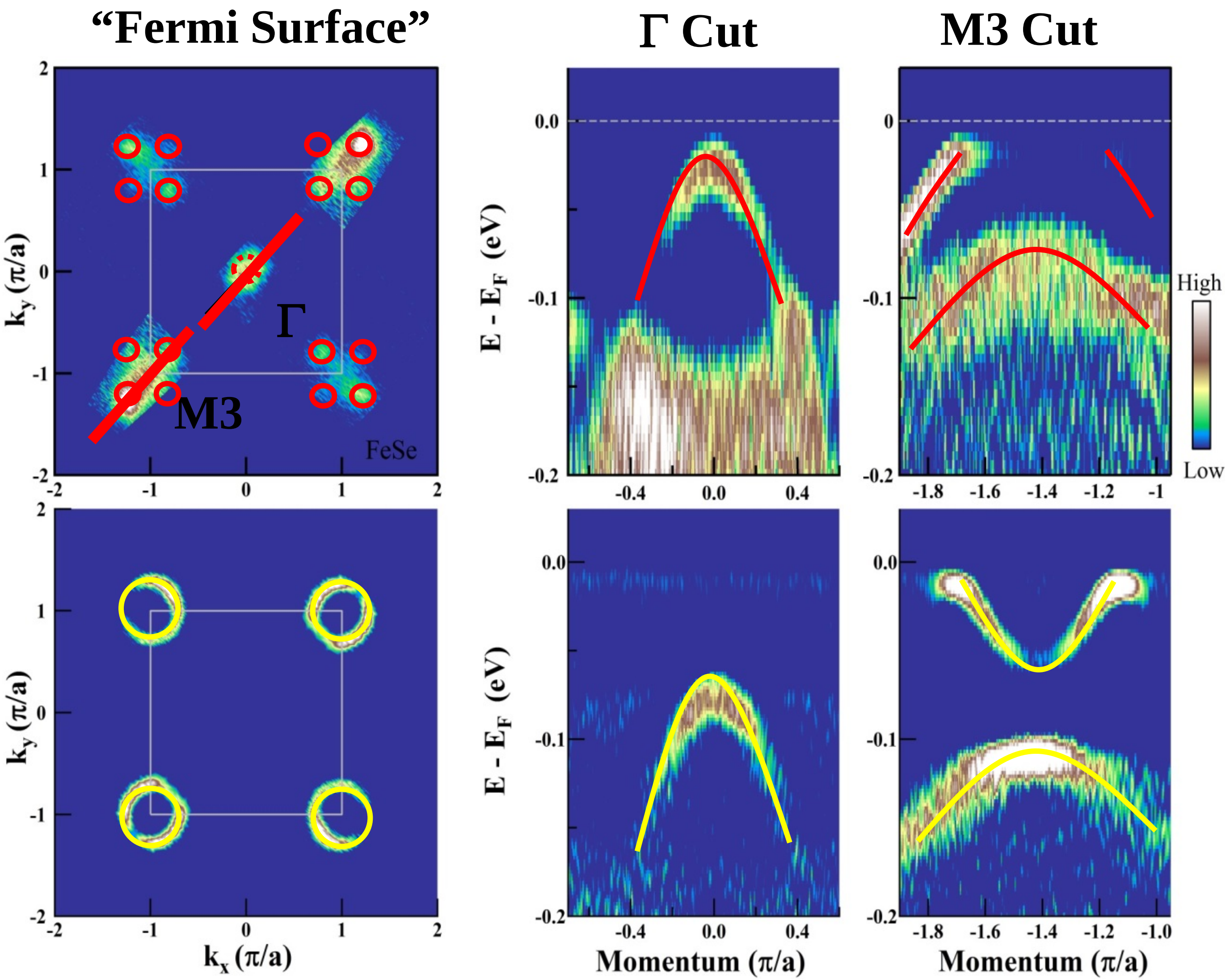
“Fermi Surface” **Γ Cut** **M3 Cut**



As-prepared
Non-superconducting

S. L. He, X. J. Zhou et al., Nature Materials 12, 605 (2013).

Non-Superconducting *vs* Superconducting Phases



As-prepared
Non-superconducting

N Phase

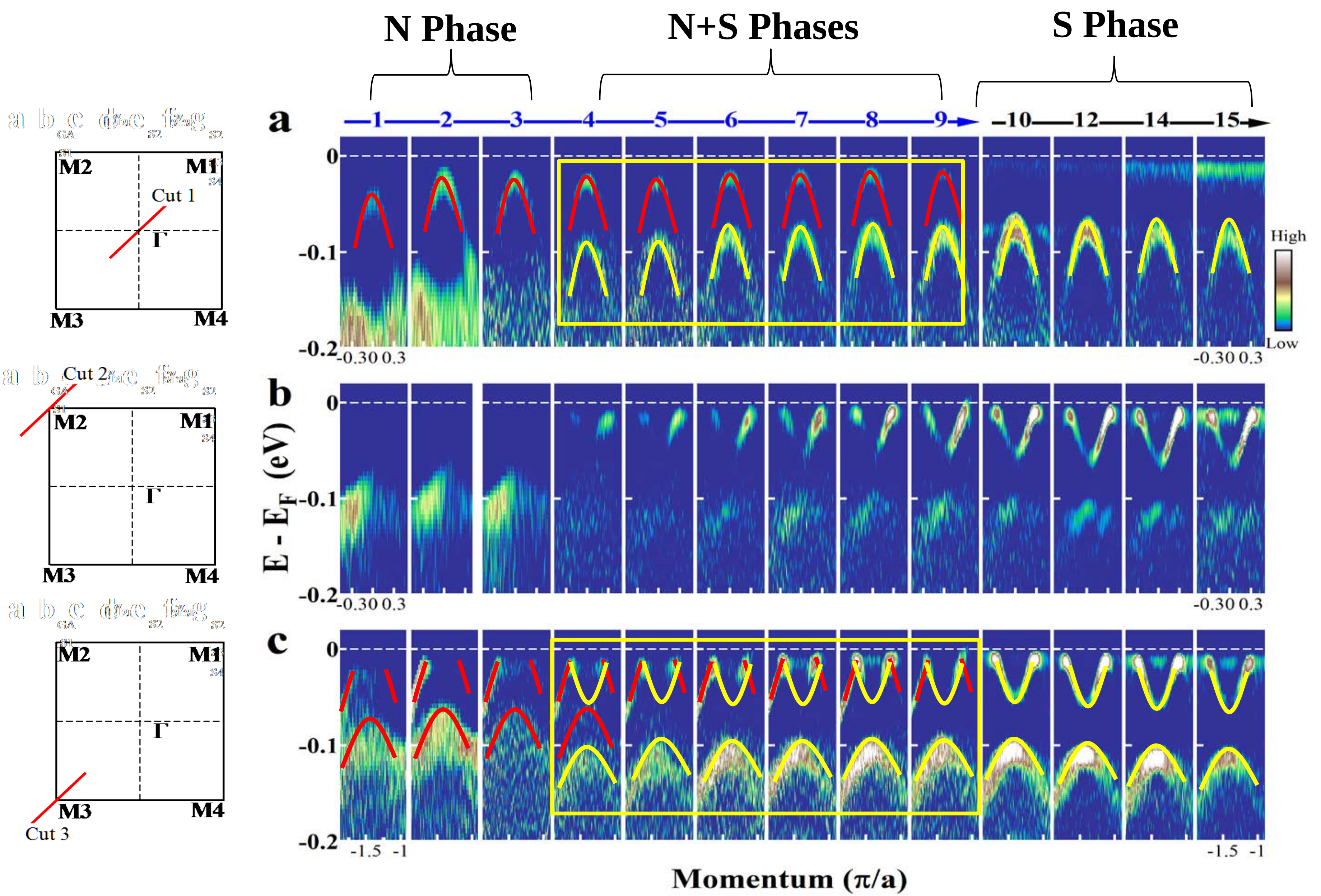
Annealed
Superconducting

S Phase

S. L. He, X. J. Zhou et al., Nature Materials 12, 605 (2013) .

Evolution from N phase to S phase

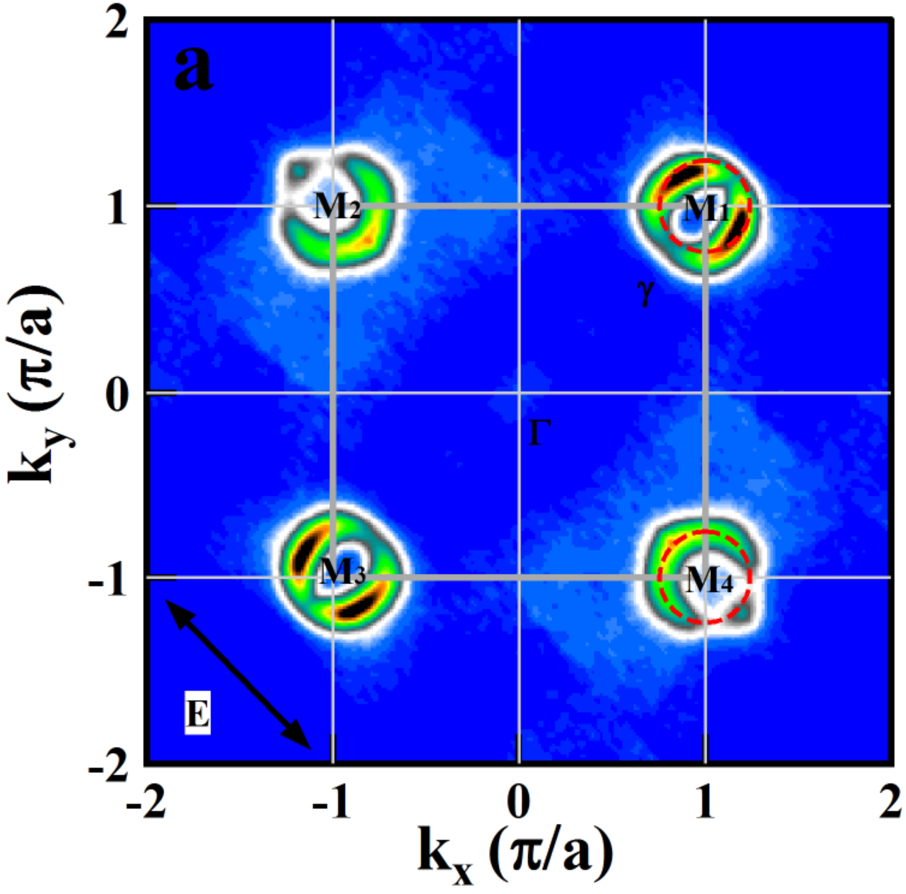
Evolution of Band Structures with Annealing in Single-Layer FeSe/SrTiO$_3$

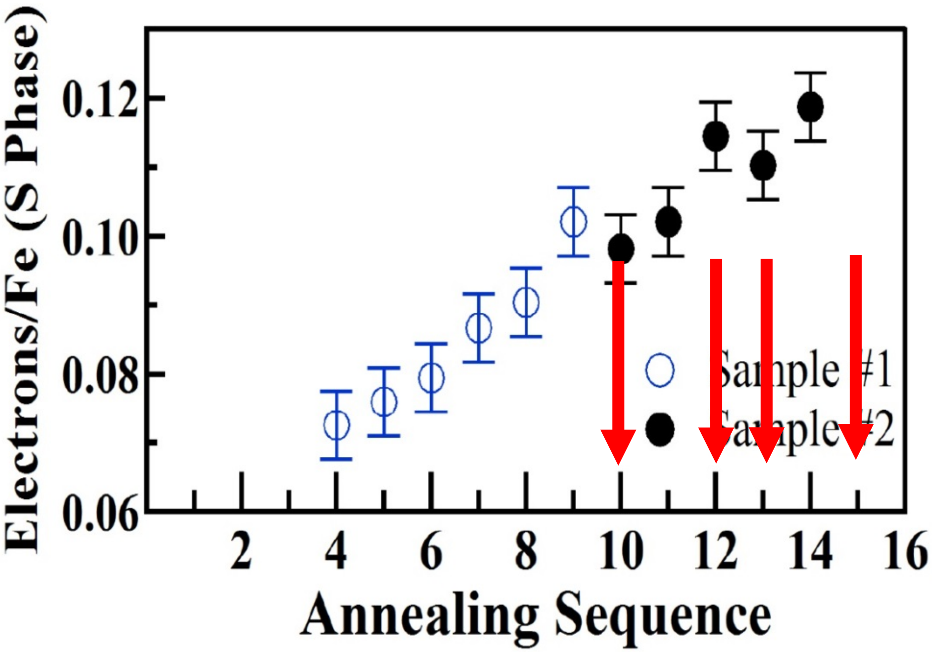


Doping-Evolution of Superconductivity

in the S Phase……

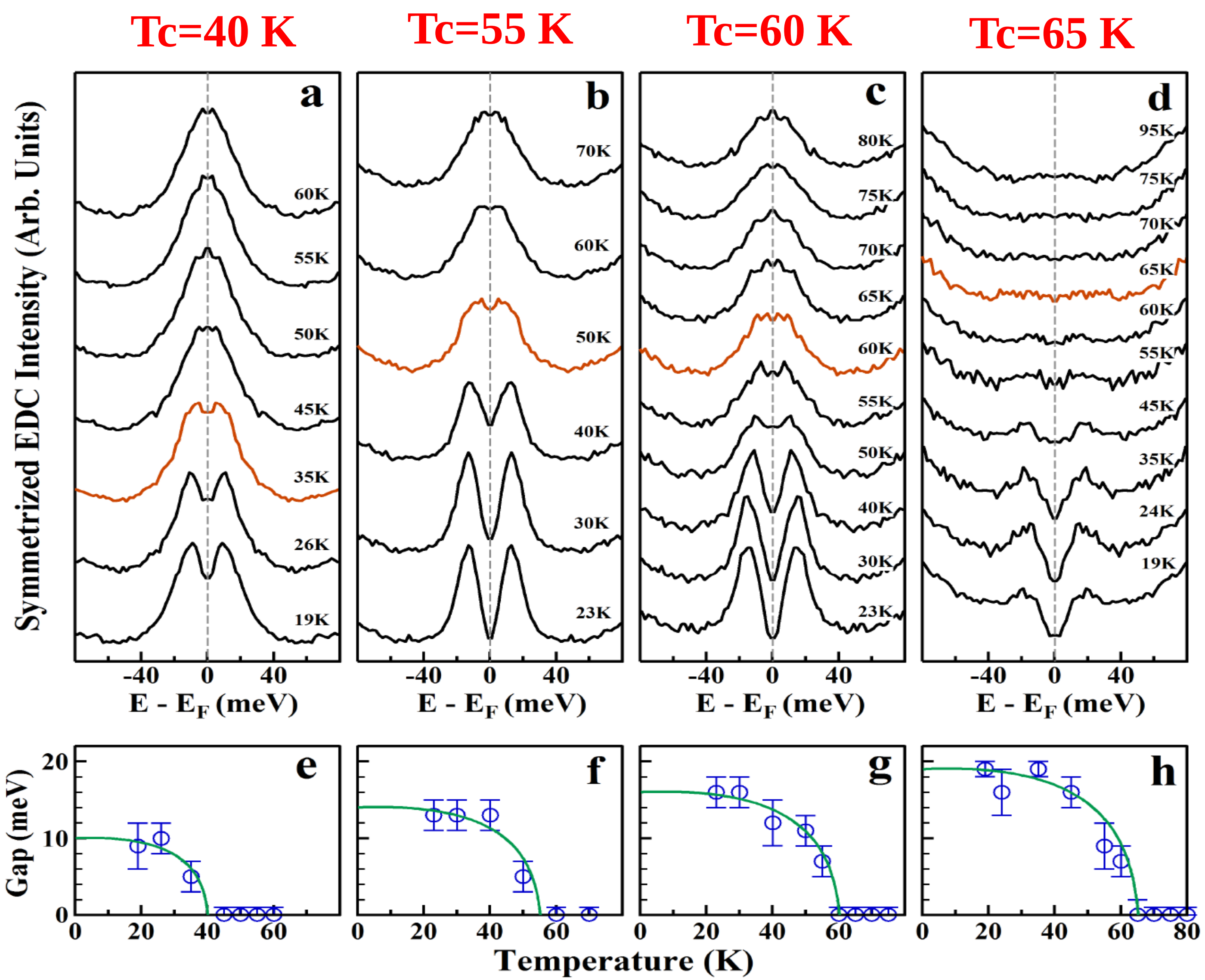
Electron Counting in S Phase in Single-Layer $FeSe/SrTiO_3$



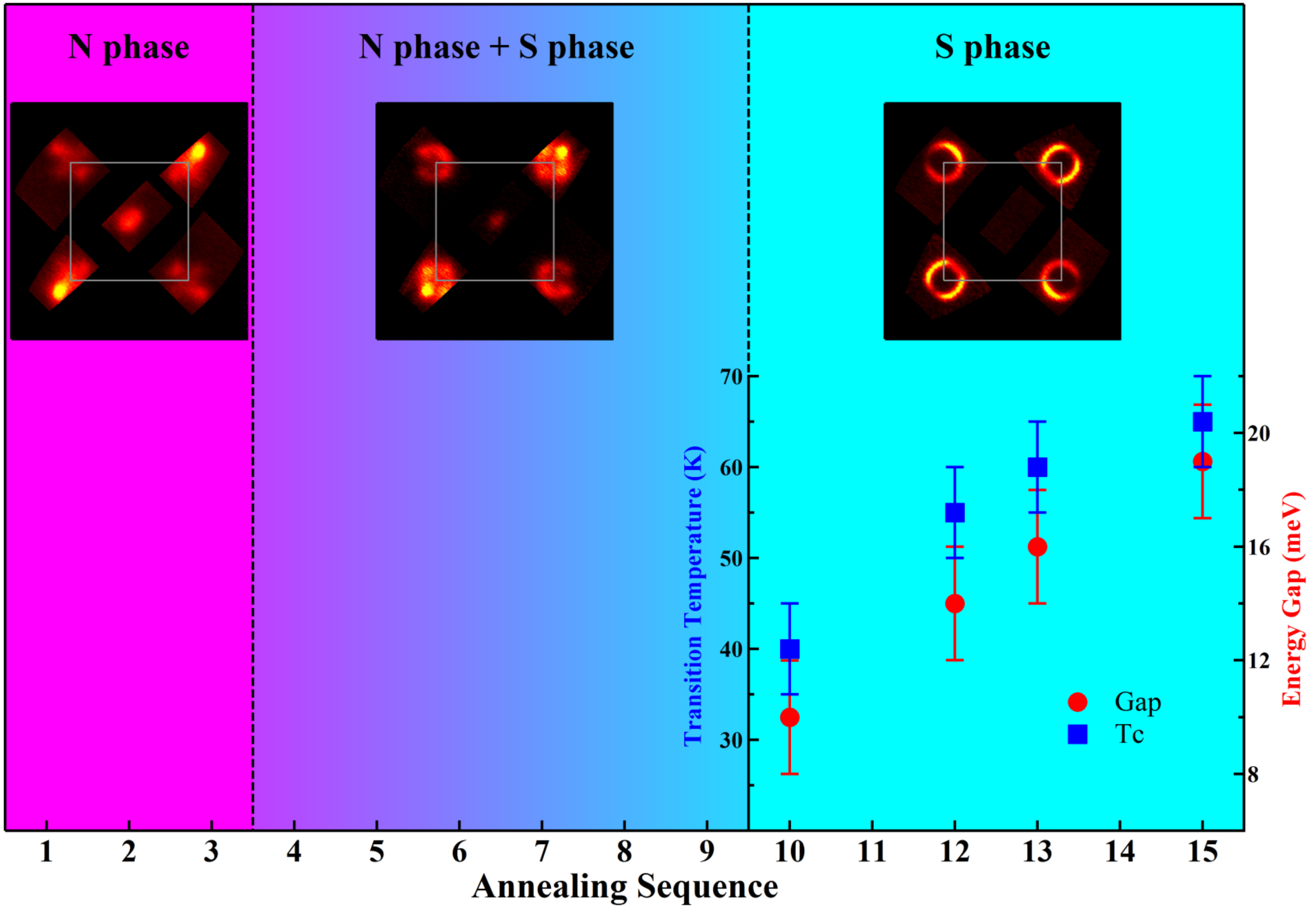


- ➢ **S phase has only an electron-like Fermi surface sheet near M;**
- ➢ **Assuming two Fermi surface sheets near M.**

Energy Gap of Single-Layer FeSe under Different Annealing Conditions



Phase Diagram of Single-Layer $FeSe/SrTiO_3$ Film



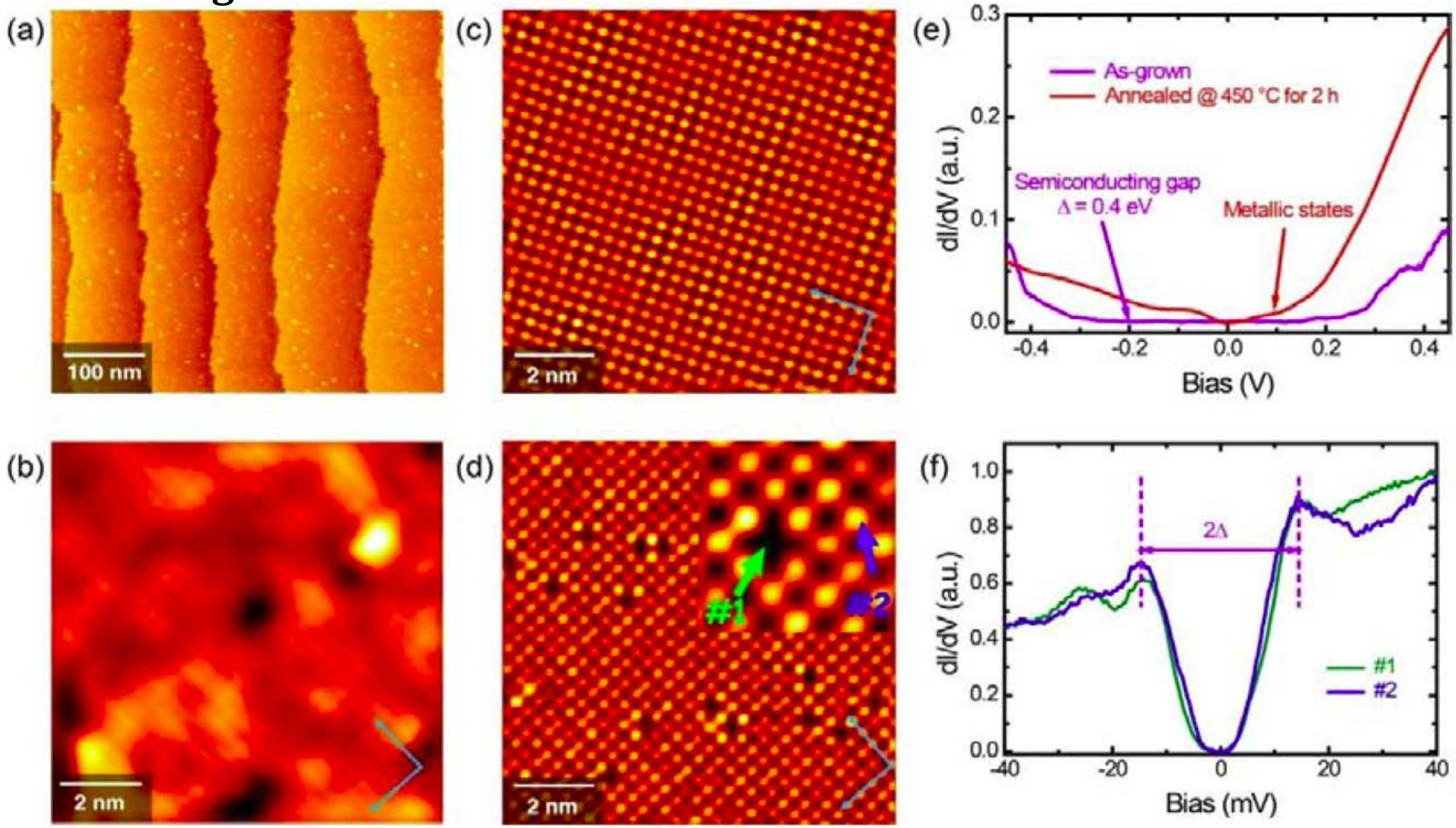
Tc~65 K
Gap ~19 meV

$2\Delta/kT_c$=6~7

S. L. He, X. J. Zhou et al., Nature Materials 12, 605 (2013).

STM/STS on As-Prepared and Vacuum Annealed Single-Layer FeSe/$SrTiO_3$ Film

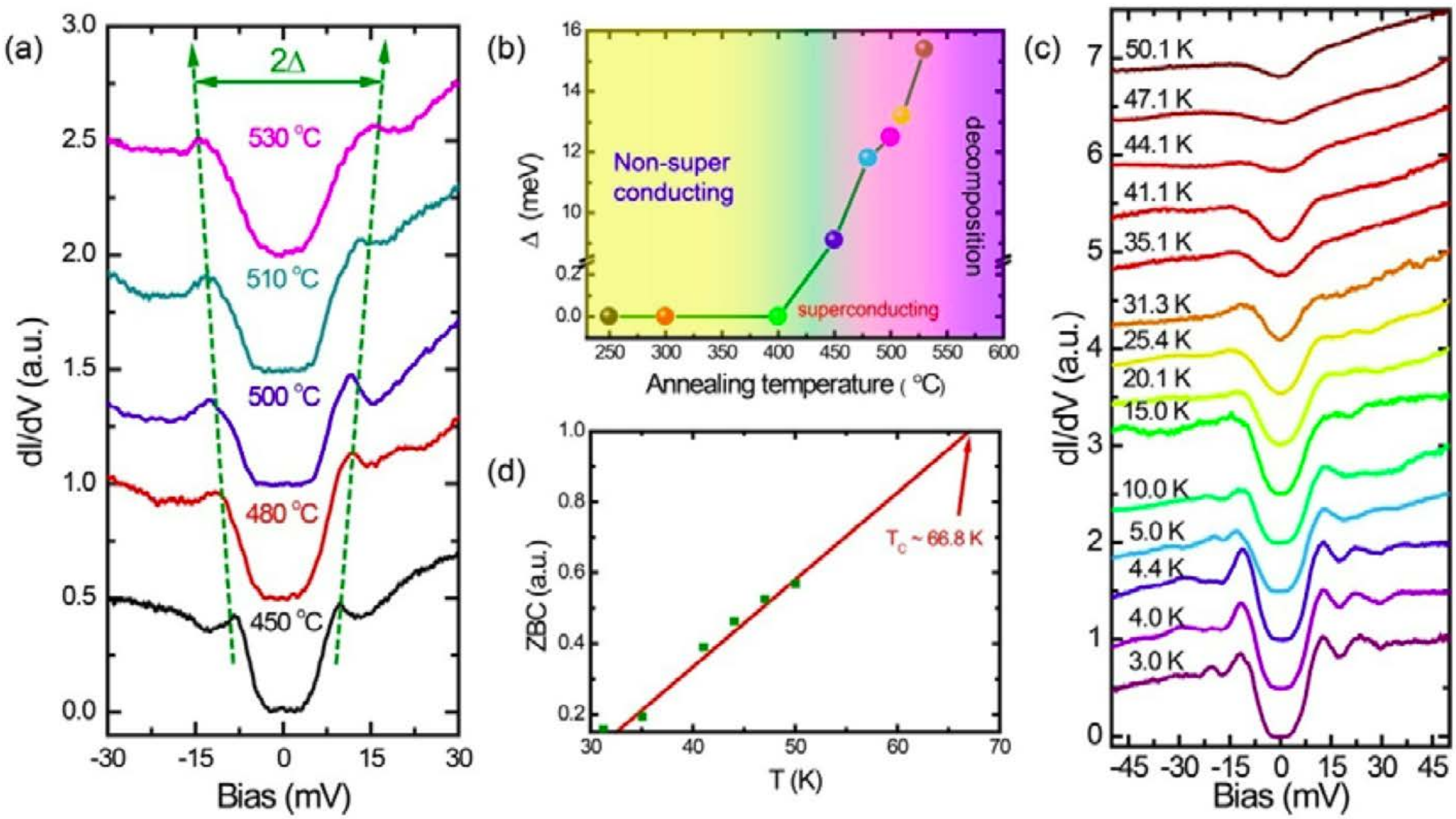
As-grown **Annealed at 400C/2hr**



Annealed at 450C/2hr **Annealed at 530C/2hr**

Wenhao Zhang, Zhi Li, Fangsen Li1, Huimin Zhang, Junping Peng, Chenjia Tang, Ke He, Xi Chen, Lili Wang, Xucun Ma and Qi-Kun Xue, Phys. Rev. B 89, 060506(R) (2014)

Evolution with Annealing for Single-Layer FeSe/$SrTiO_3$ Film



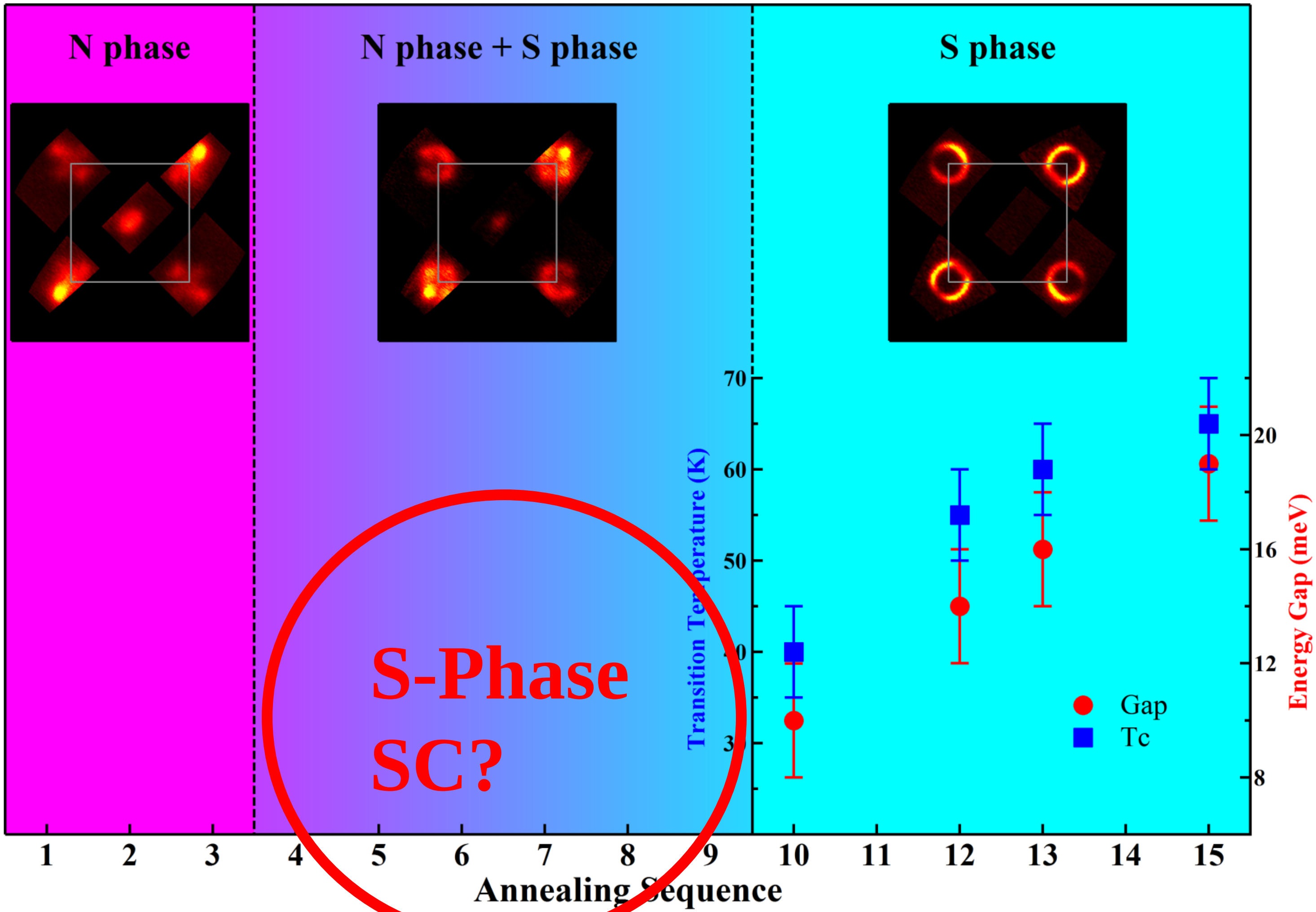
Wenhao Zhang, Zhi Li, Fangsen Li1, Huimin Zhang, Junping Peng, Chenjia Tang, Ke He, Xi Chen, Lili Wang, Xucun Ma and Qi-Kun Xue, Phys. Rev. B 89, 060506(R) (2014)

Electronic Evidence of an Insulator-Superconductor Transition in Singe-Layer FeSe/$SrTiO_3$ Films.

Junfeng He, Xu Liu, Wenhao Zhang, Lin Zhao, X. C. Ma, Q. K. Xue, X. J. Zhou et al.,

arXiv:1401.7115

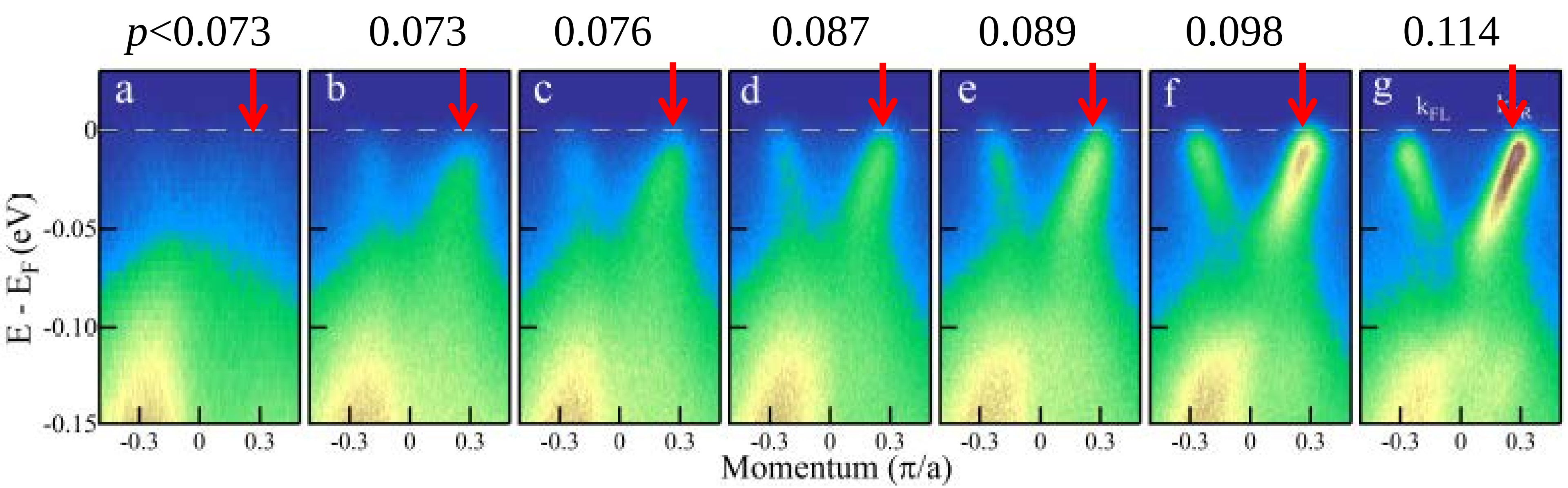
Phase Diagram of Single-Layer $FeSe/SrTiO_3$ Film



Insulator → **Superconductor**

S. L. He, X. J. Zhou et al., Nature Materials 12, 605 (2013).

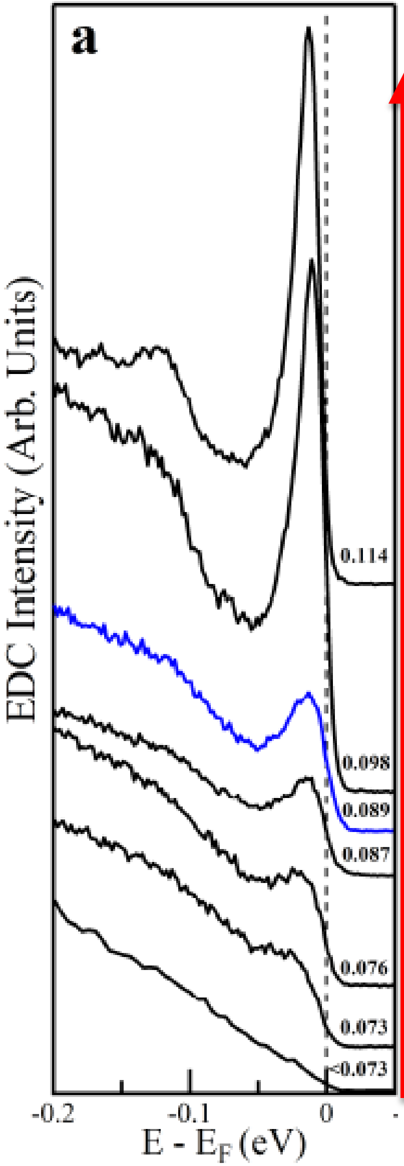
Band Structure Evolution of S Phase with Doping



Spectral weight increases with increasing electron doping.

J. F. He, X. J. Zhou et al., arXiv:1401.7115.

Doping Evolution of EDCs and Energy Gaps in S phase



At high doping
Well-defined
coherence peak

Increase doping
Peak emerges

At low doping
No peak

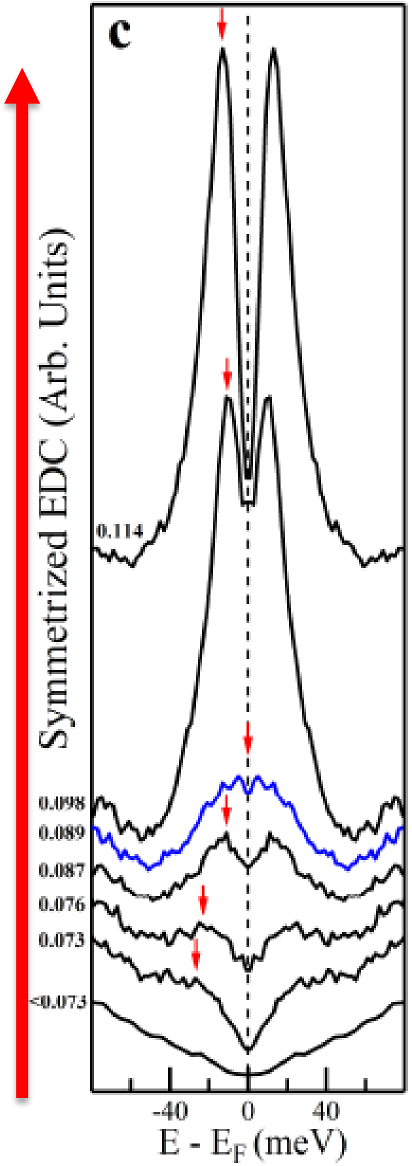
At high doping
Gap opens again

High-Doping Gap

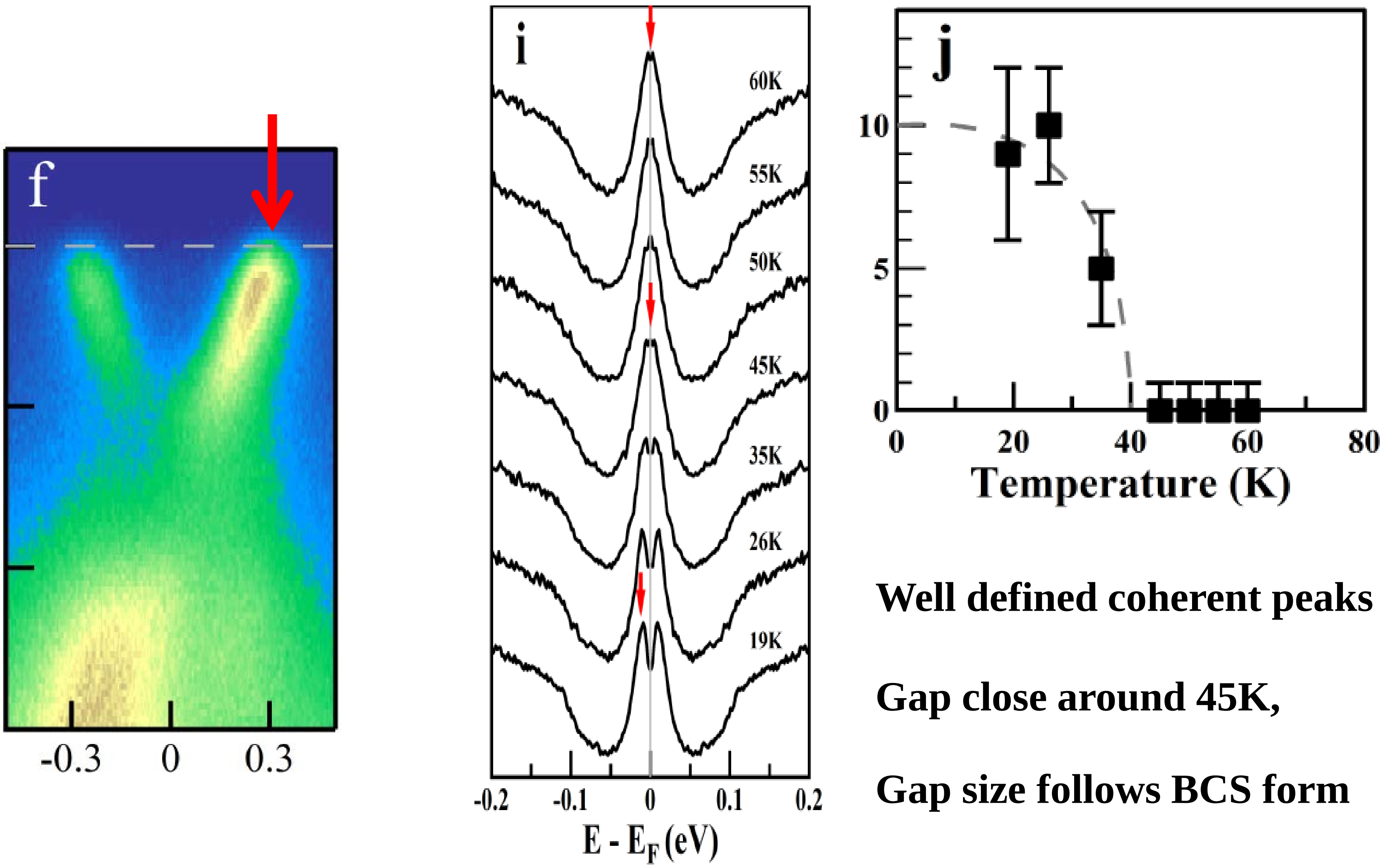
Increase doping
Gap gets smaller
and closes at
doping 0.089

At low doping
Large gap opening

Low-Doping Gap



Temperature Dependence of High-Doping Gap



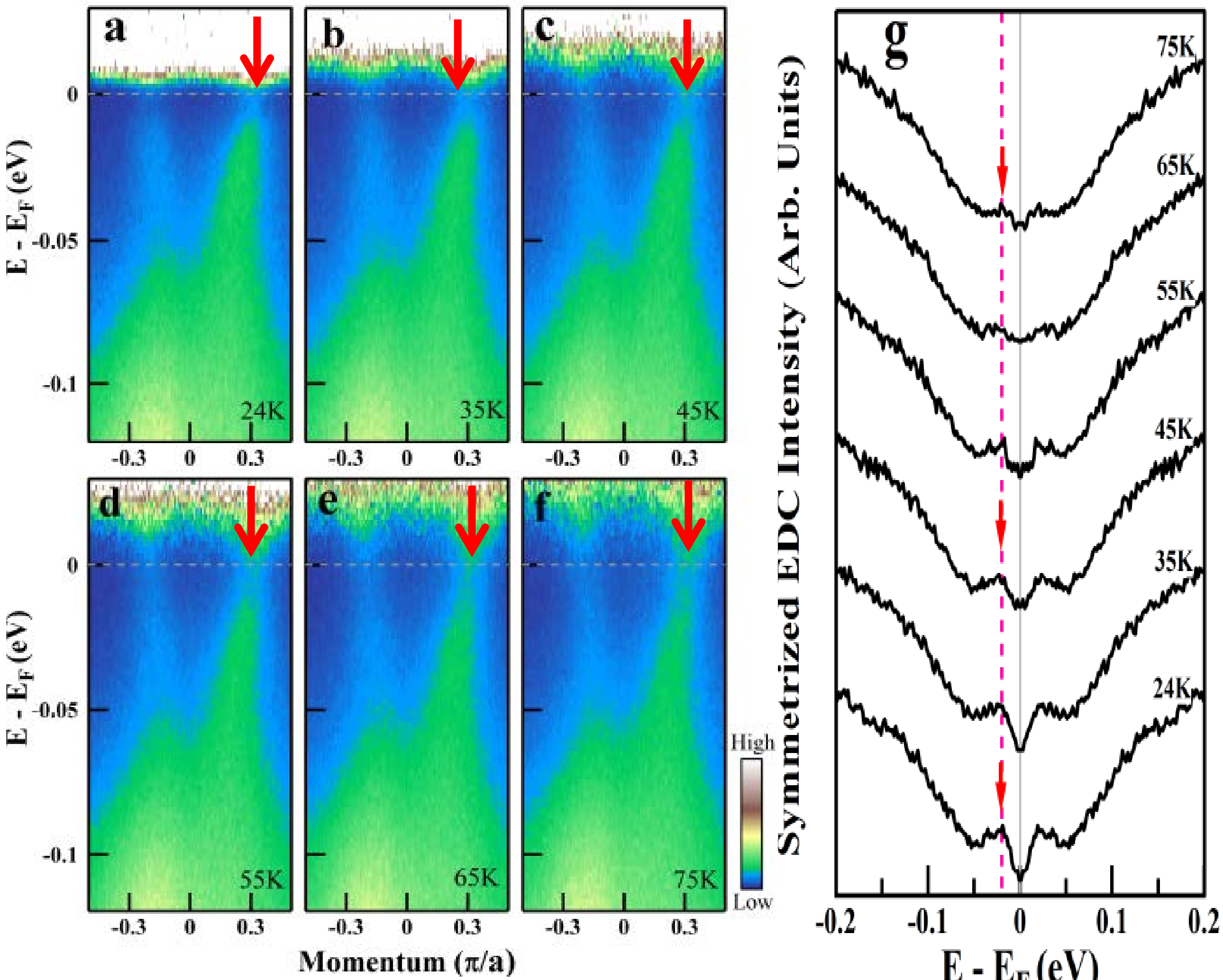
Well defined coherent peaks

Gap close around 45K,

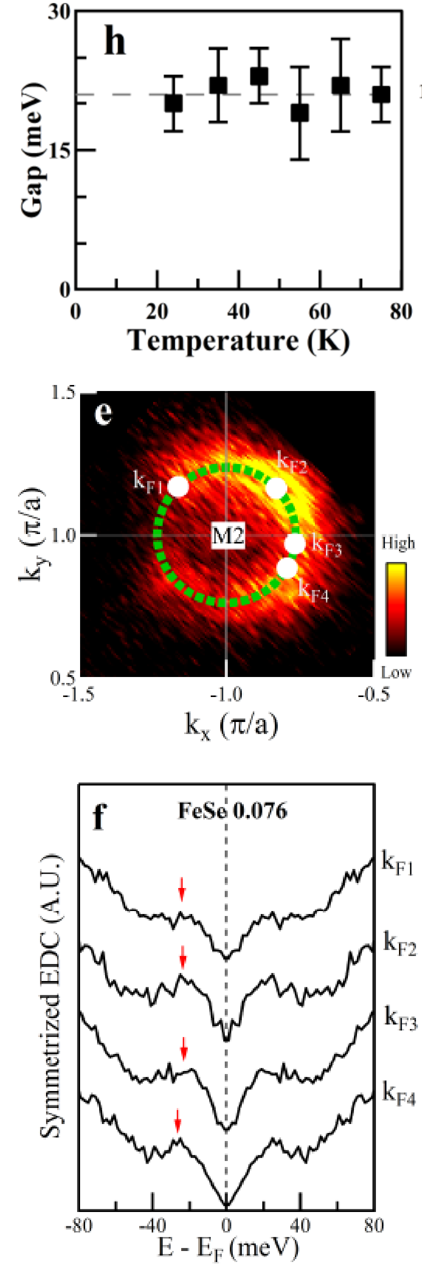
Gap size follows BCS form

High-doping gap is a superconducting gap

Temperature and Momentum Dependence of Low-Doping Gap



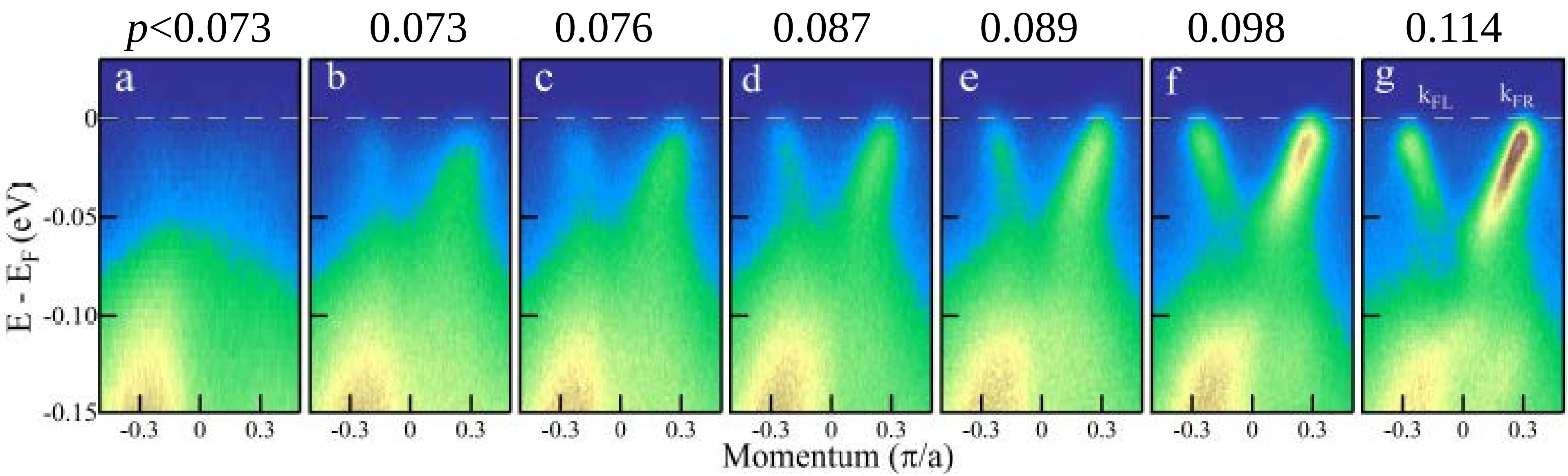
Tempearture dependence of M2 cut



Broad peak, Weak Temperature dependence of the Gap

→ insulating gap

Insulator-Superconductor Transition in S-Phase of Single-Layer $FeSe/SrTiO_3$



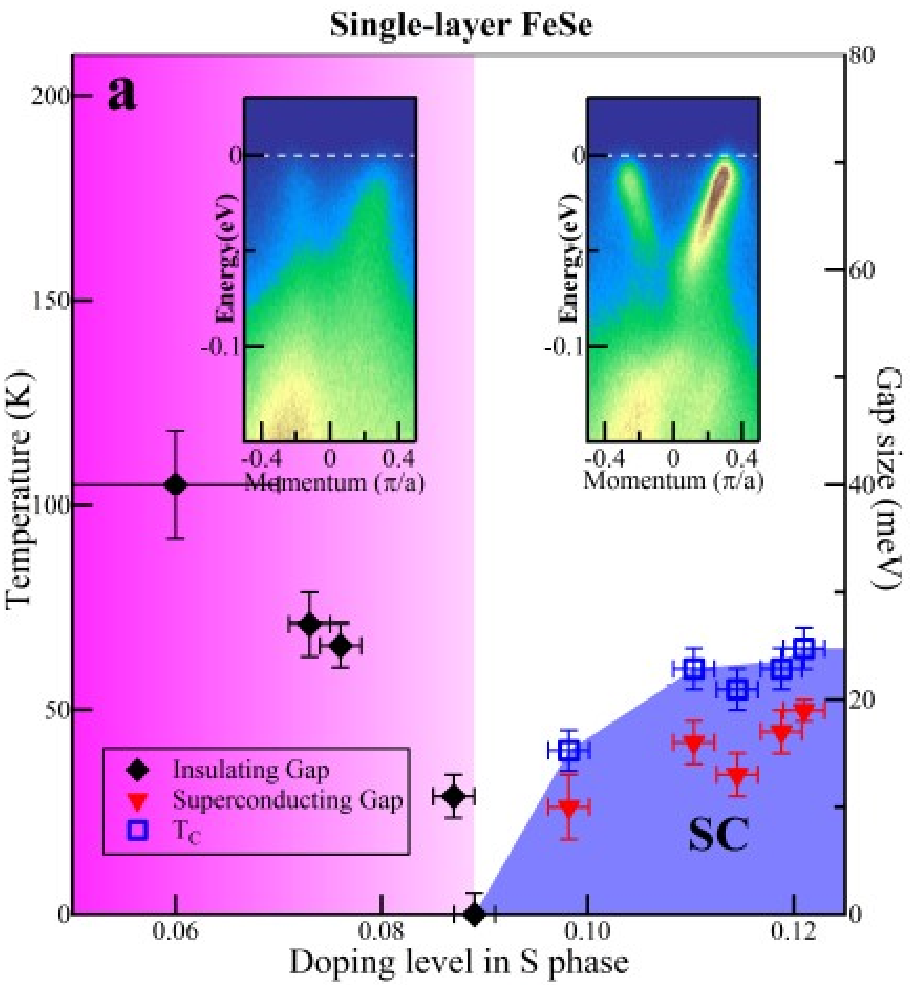
Insulating Gap

Gap closed

Superconducting Gap

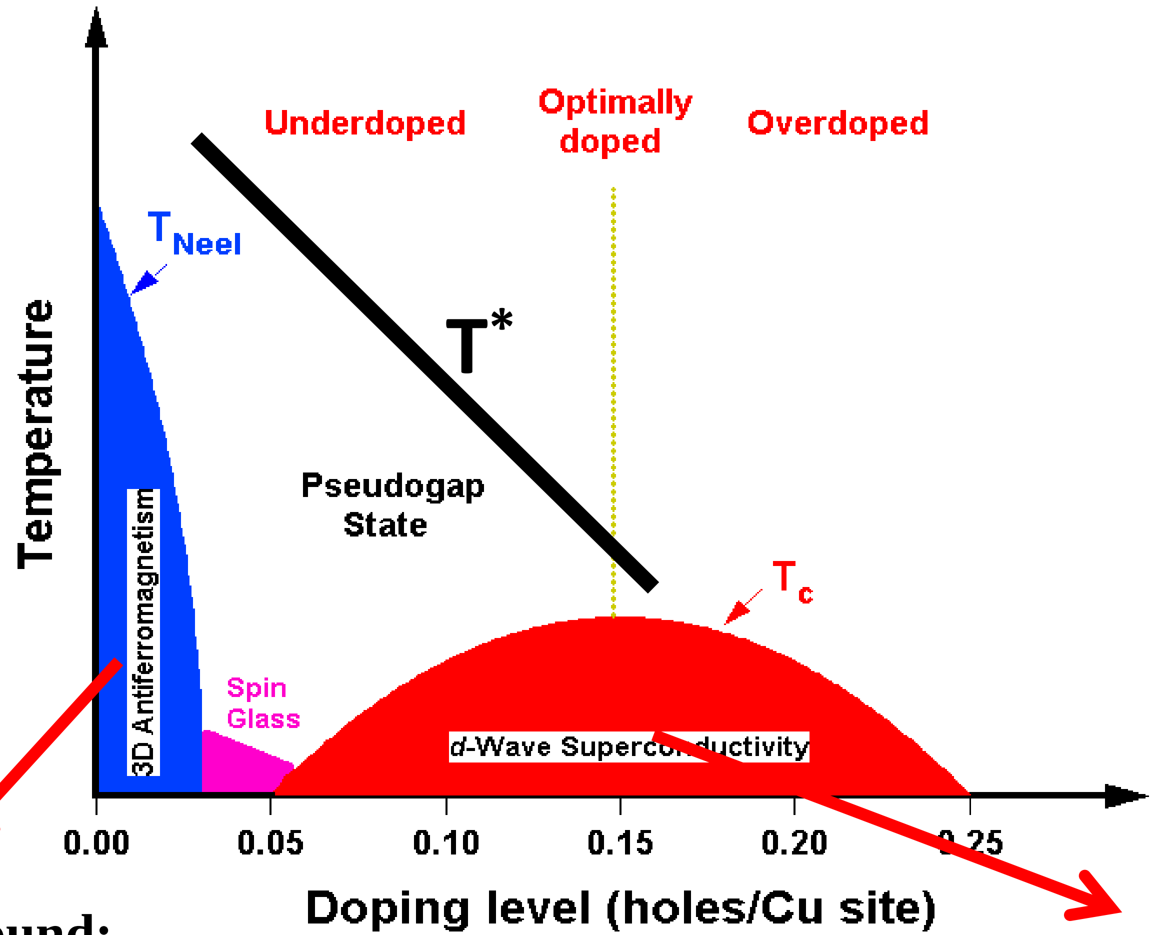
J. F. He, X. J. Zhou et al., arXiv:1401.7115.

Phase Diagram of S-Phase of Single-Layer $FeSe/SrTiO_3$



J. F. He, X. J. Zhou et al., arXiv:1401.7115.

High-Tc Cuprate Superconductors: Doped Mott Insulator

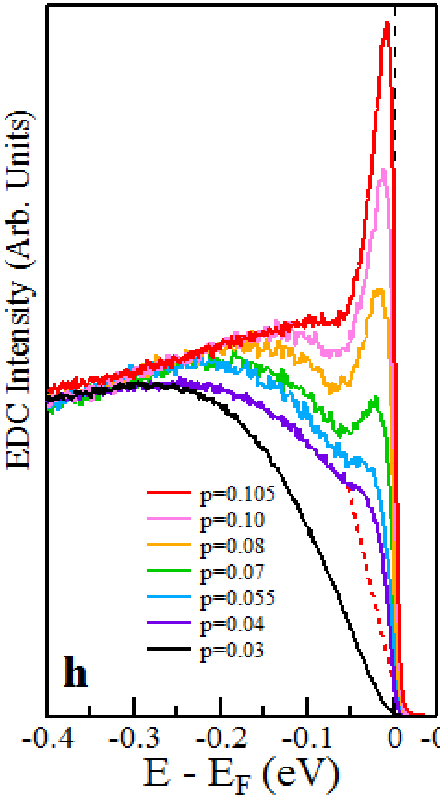


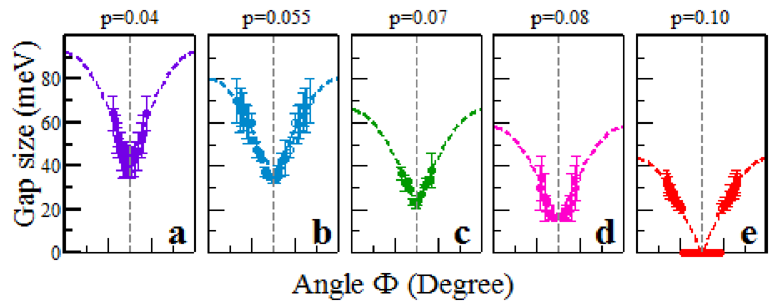
Parent compound:
AFM Mott insulator

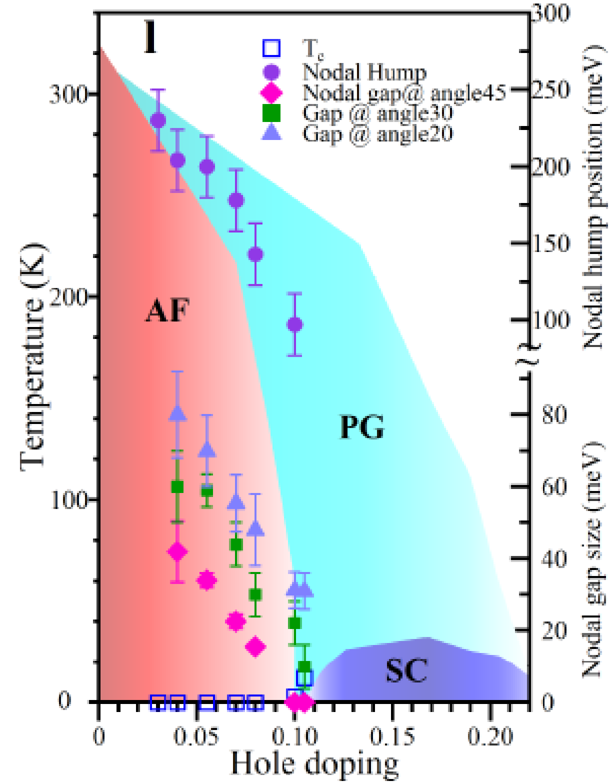
d-wave superconductor

P. A. Lee, N. Nagaosa and X. G. Wen, Rev. Modern Phys. 78, 17 (2006).

Insulator-Superconductor Transition in La-Bi2201





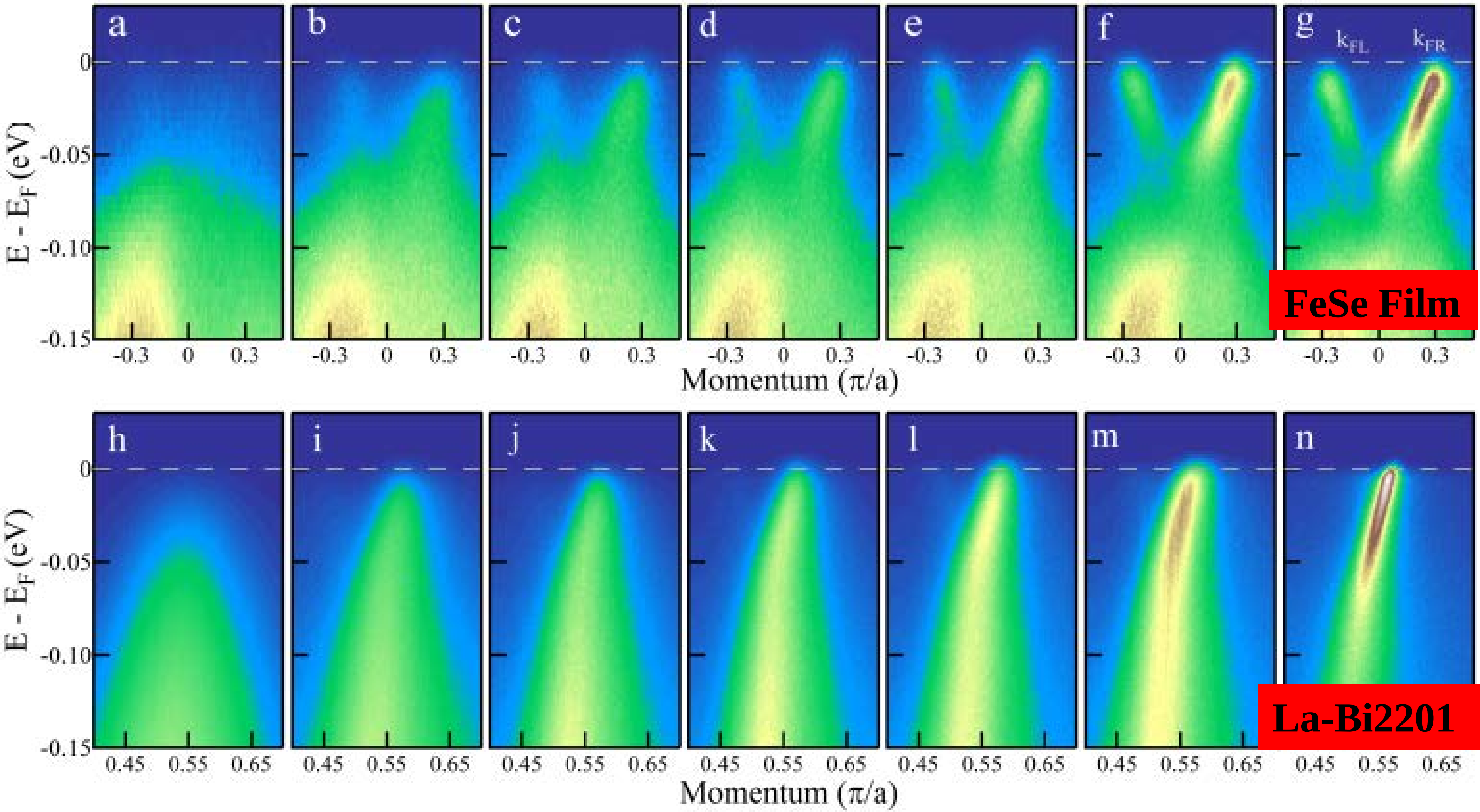


- ➢ **Nodal gap forms below p~0.10;**
- ➢ **Nodal gap decreases with increasing doping and vanishes at p~0.10;**
- ➢ **Near p~0.10, (1). The 3D antiferromagnetic order disappears;**
  (2). Superconductivity starts to emerge;
  (3). Nodal gap gets to zero.

Y. Y. Peng, X. J. Zhou et al., Nature Communications 4, 2459 (2013) .

Similarity between Single-Layer FeSe Film and La-Bi2201

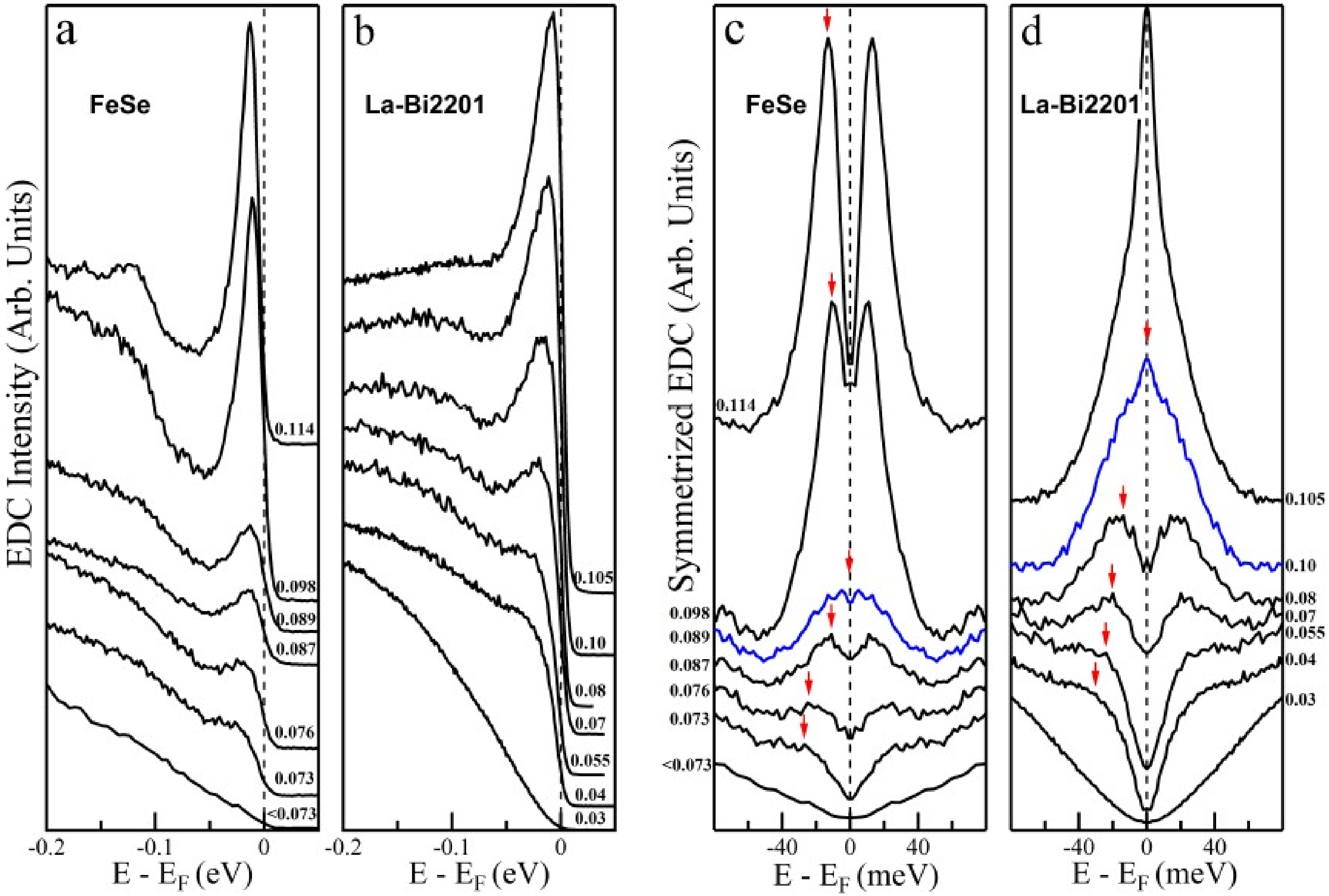
1. Band Structure Evolution



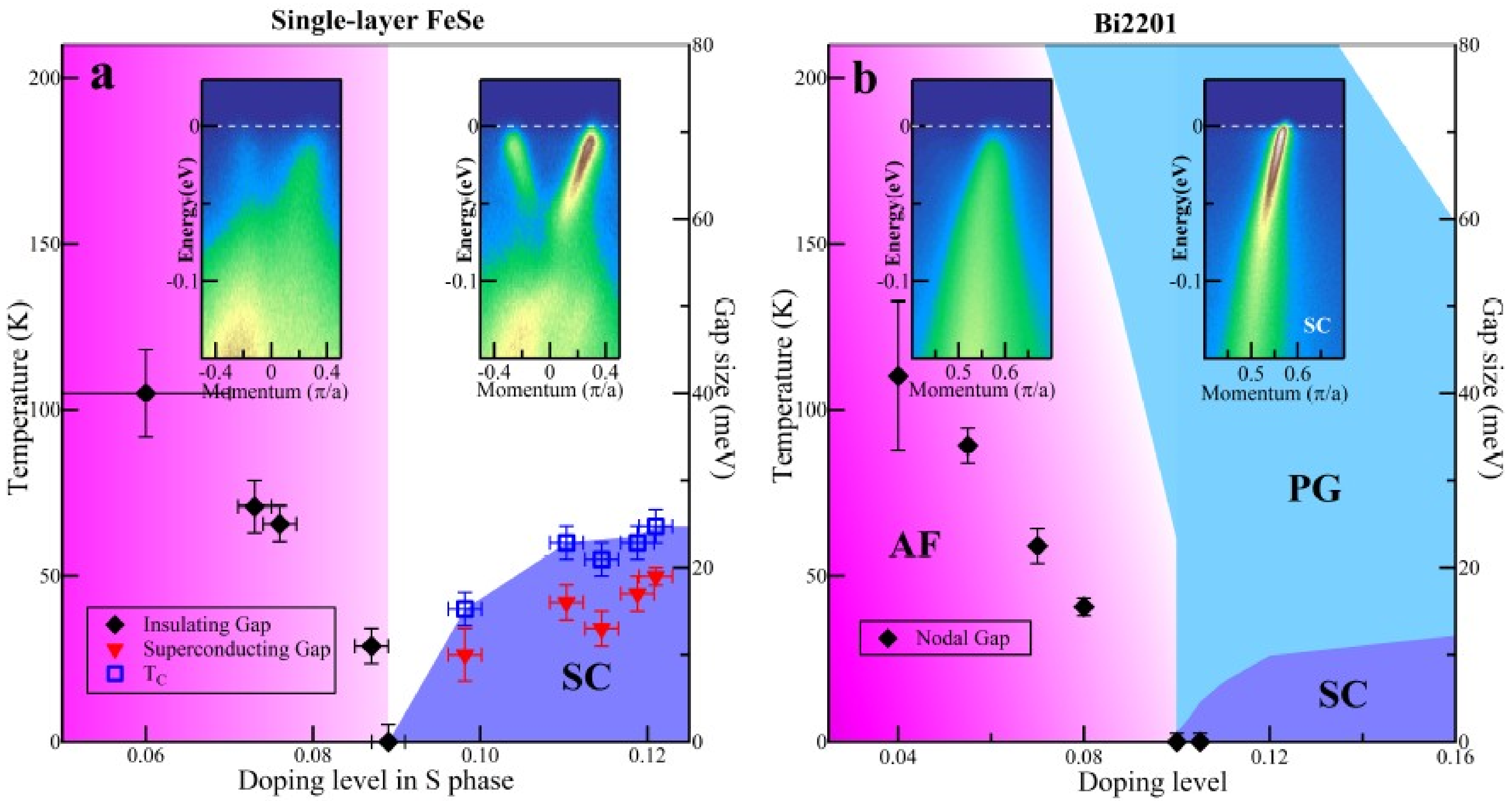
Similarity between Single-Layer FeSe Film and La-Bi2201

2. EDC Cohenerce Peak Evolution

3. Insulating Gap Evolution



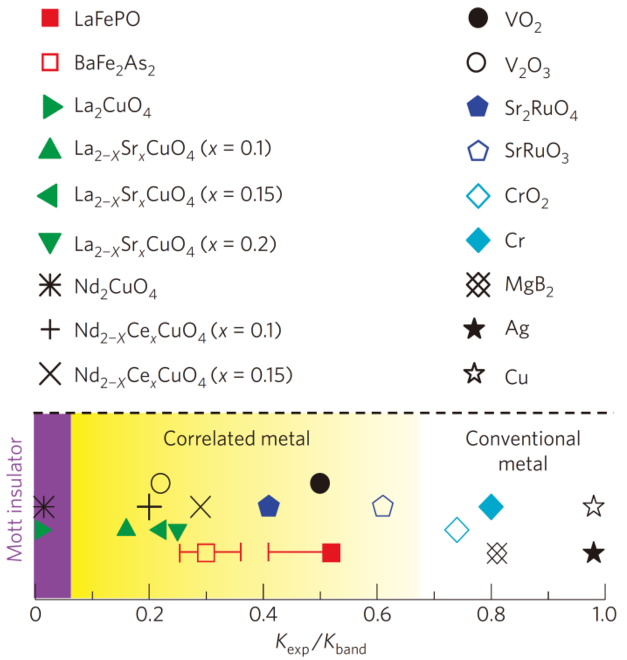
Similar Electronic Phase Diagrams



J. F. He, X. J. Zhou et al., arXiv:1401.7115.

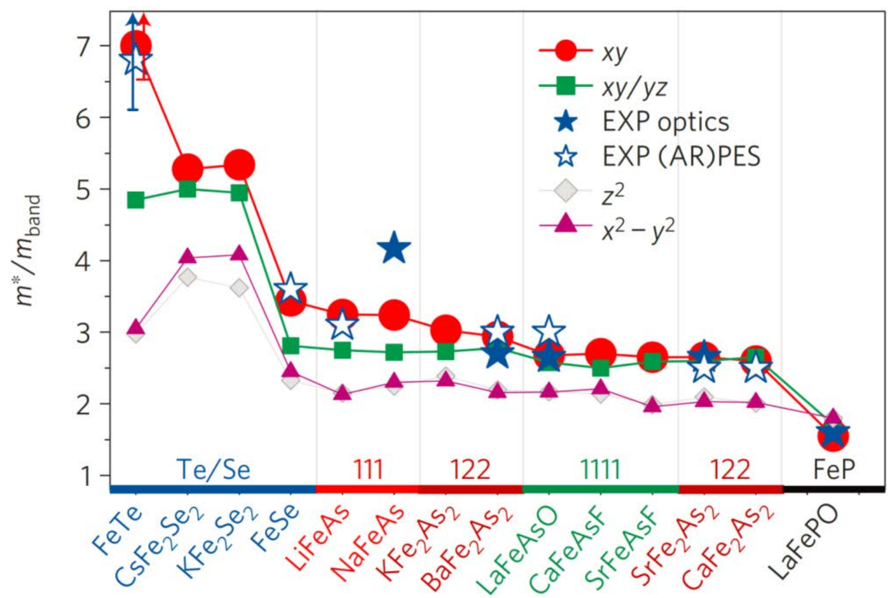
Strong Correlation in Single-Layer FeSe/SrTiO$_3$ Film

1. Existence of Electron Correlation in Fe-based Compounds



M. M. Qazibash et al., Nature Phys. 5, 647 (2009)

2. Stronger Electron Correlation in 11-Based Compounds



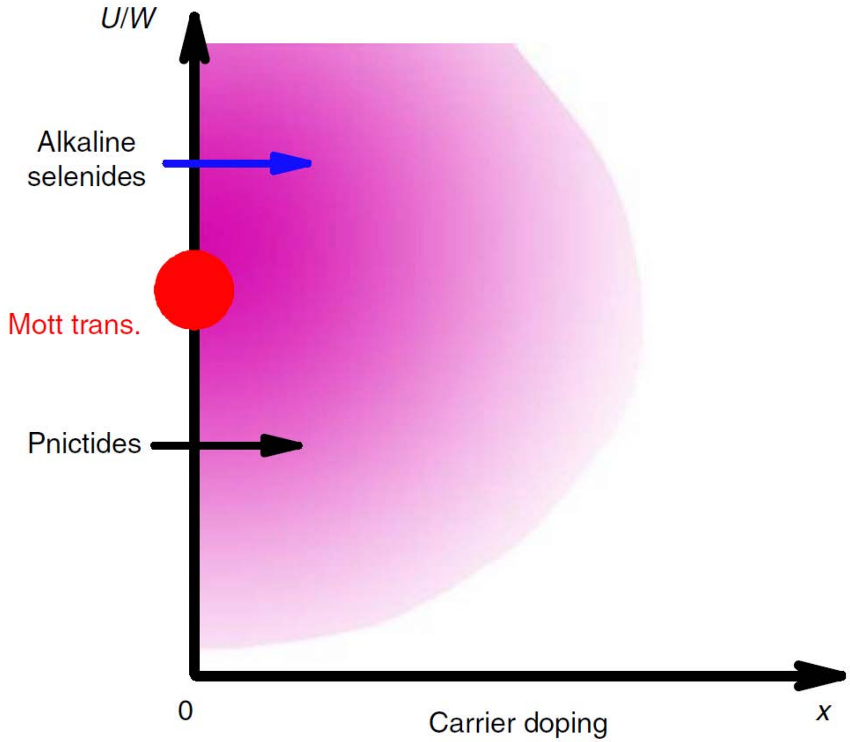
Z. P. Yin et al., Nature Mater. 10, 932 (2011).

3. Enhanced electron correlation due to reduced dimensionality—2D film;

4. Enhanced electron correlation due to tensile strain in single-layer FeSe film (Bulk FeSe: 3.76 A, Single-layer FeSe/SrTiO film: 3.80 A)

Doping of the Orbital-Selective Mott Insulator?

- **Fe-based compounds are on the verge of doped Mott insulator;**
- **Single-Layer FeSe/$SrTiO_3$ has particularly strong electron correlation.**
- **Similarity between Single-layer FeSe/$SrTiO_3$ Film and the underdoepd La-Bi2201.**

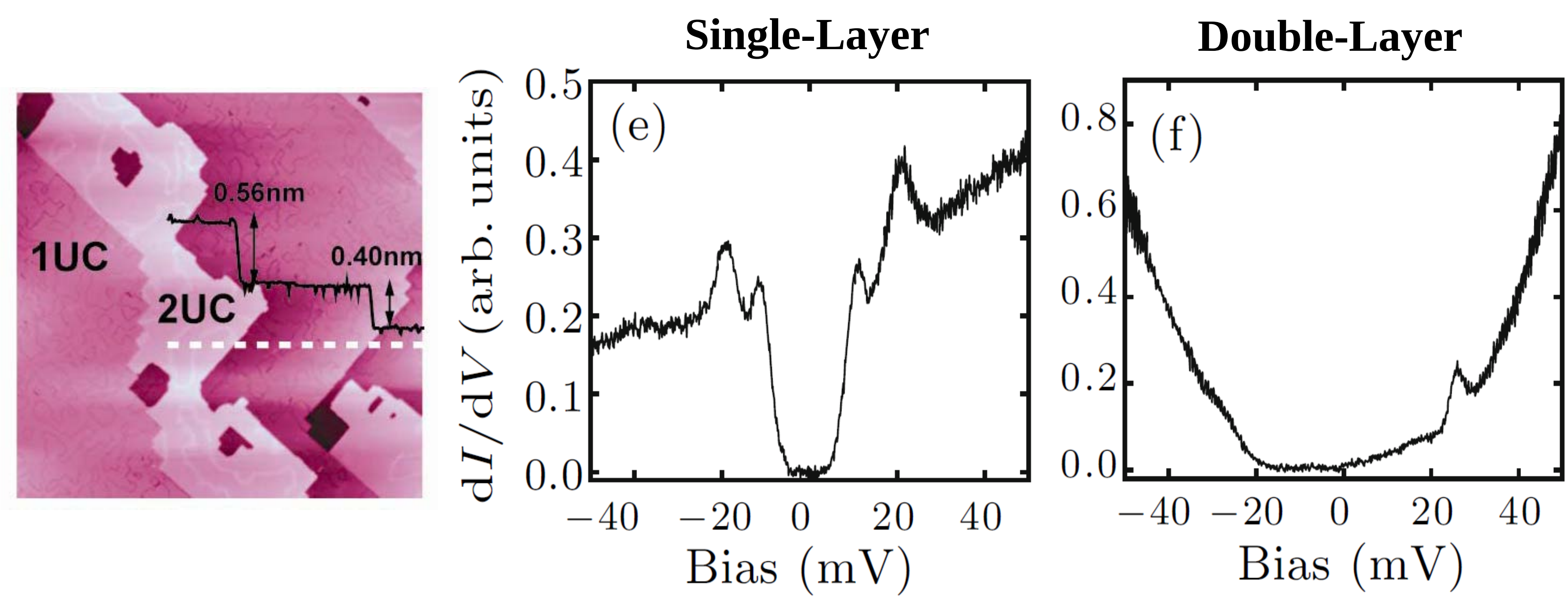


Q. M. Si and E. Abrahams, Phys. Rev. Lett. 101, 076401 (2008).
R. Yu et al., Nature Communications 4, 2783 (2013).
R. Yu and Q. M. Si, Phys. Rev. Lett. 110, 146402 (2013).

Dichotomy of Electronic Structure and Superconductivity between the Single-Layer and Double-Layer FeSe/$SrTiO_3$

X. Liu, D. F. Liu, W. H. Zhang, J. F. He, X. C. Ma, Q. K. Xue, X. J. Zhou et al.,
Nature Communications 5, 5047(2014).

Distinct Behaviors between Single-Layer and Two-Layer FeSe/$SrTiO_3$: STM/STS



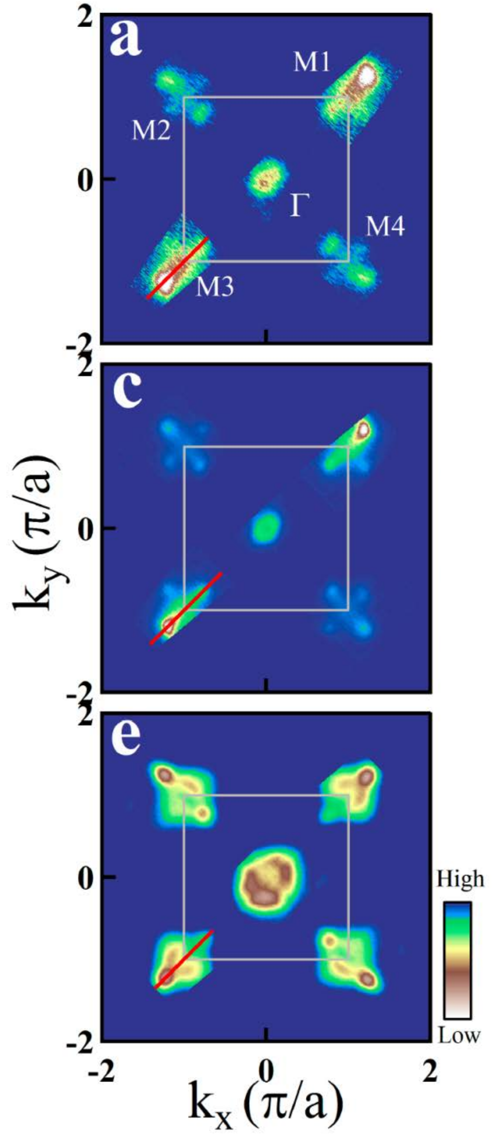
Q. Y. Wang et al., Chin. Phys. Lett. 29, 037402 (2012).

As-prepared single-layer and double-layer $FeSe/SrTiO_3$ are in N phase

Single-Layer $FeSe/SrTiO_3$

Double-Layer $FeSe/SrTiO_3$

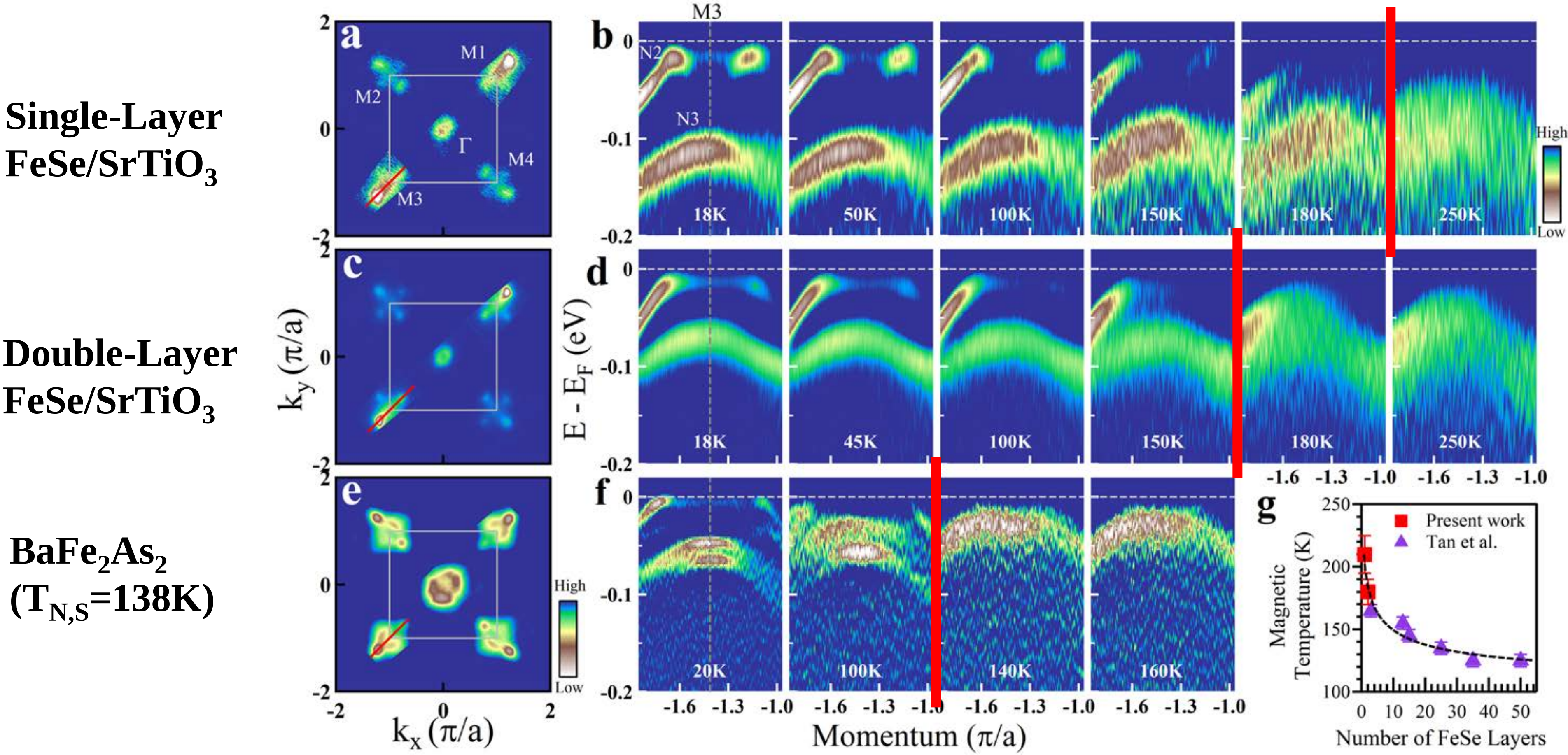
$BaFe_2As_2$



N phase

N phase

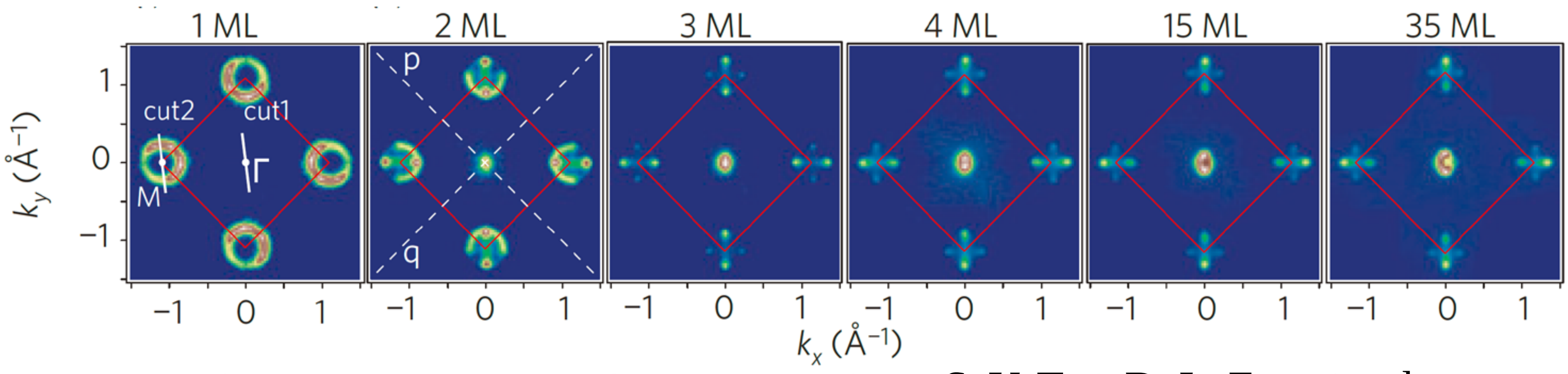
The Fermi surface, M point band and its temperature dependence resemble those in $BaFe_2As_2$ below the magnetic/structural transition



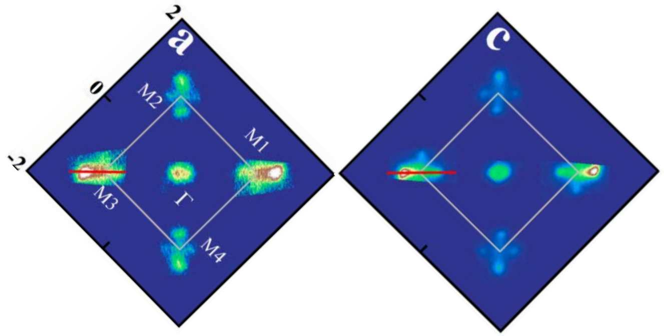
The N Phase in as-prepared FeSe/SrTiO$_3$ is likely magnetic. Further direct measurement is needed.

X. Liu, X. J. Zhou et al., Nature Communications 5, 5047(2014).

N Phase is Present in Single-Layer and Double-Layer $FeSe/SrTiO_3$

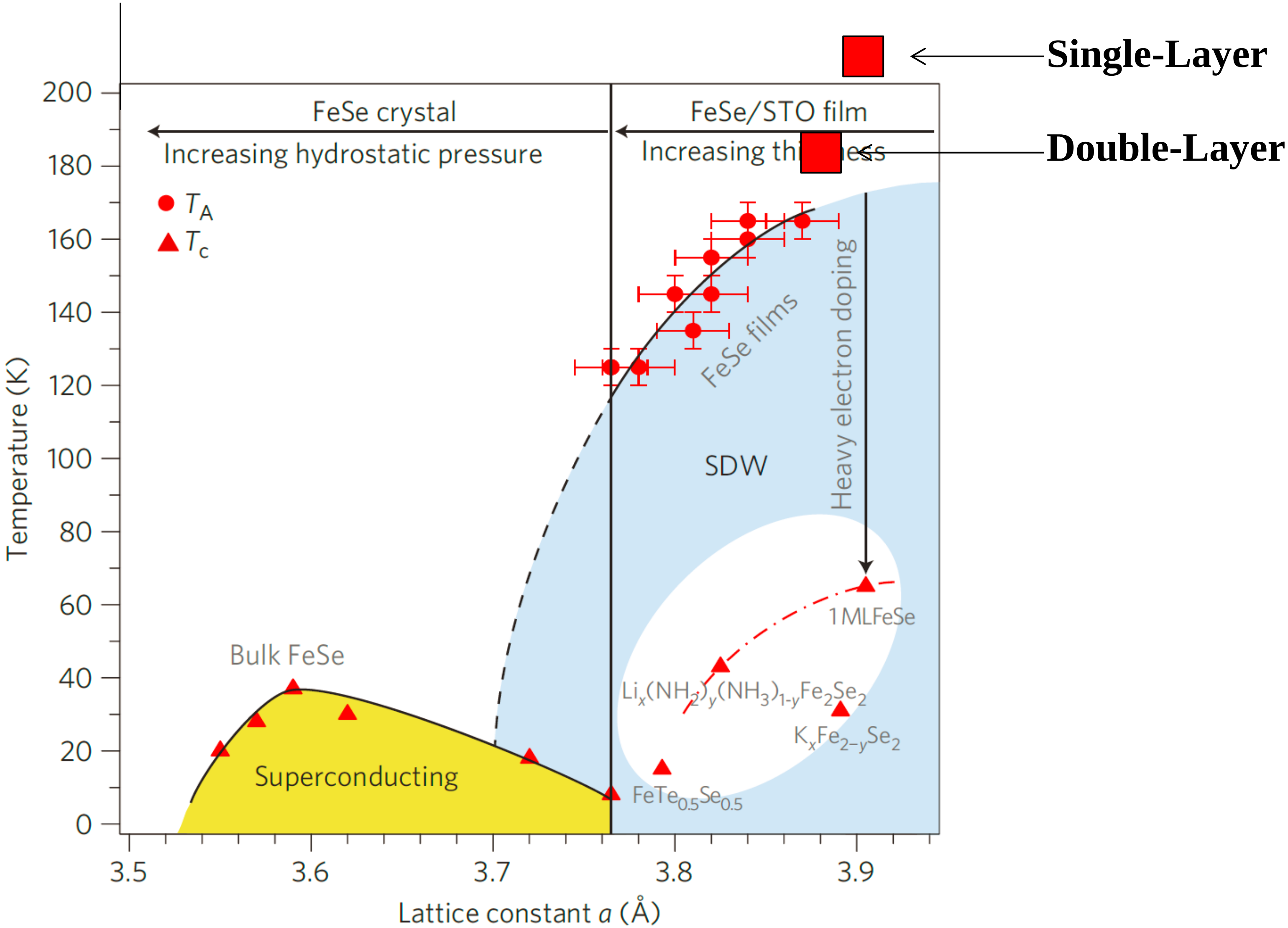


S. Y. Tan, D. L. Feng et al.,
Nature Materials 12 (2013)634.



X. Liu, X. J. Zhou et al.,
Nature Communications 5, 5047(2014).

Transition Temperature *vs* Number of FeSe Layers



S. Y. Tan, D. L. Feng et al., Nature Materials 12 (2013)634;

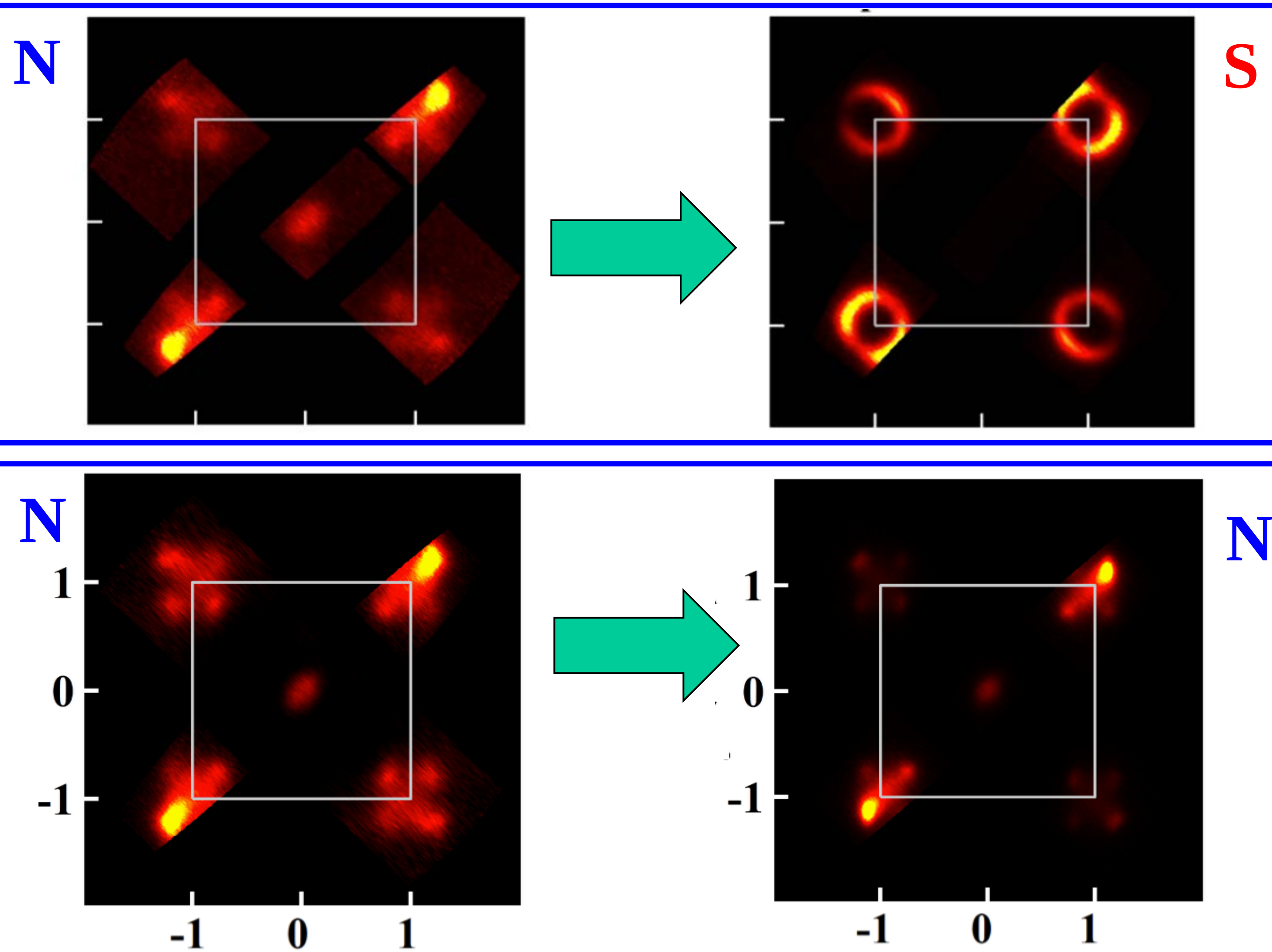
X. Liu, X. J. Zhou et al., Nature Communications 5, 5047(2014).

Distinct Behaviors between Single-Layer and Double-Layer FeSe/$SrTiO_3$

Vacuum Annealed under *Same* Conditions

Single-Layer FeSe/STO

Double-Layer FeSe/STO

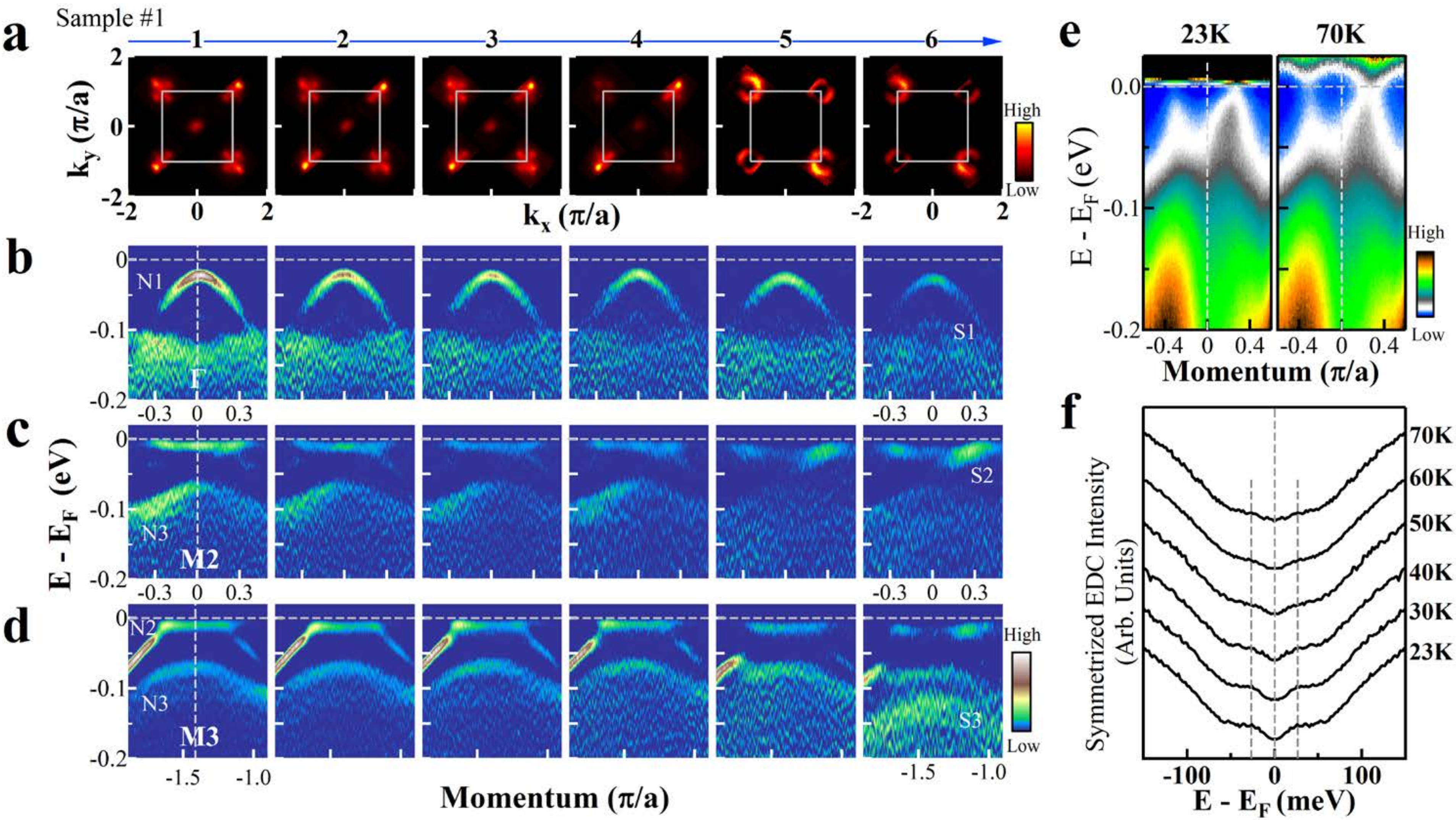


X. Liu, X. J. Zhou et al., Nature Communications 5, 5047(2014).

Let us now enhance the vacuum-annealing condition

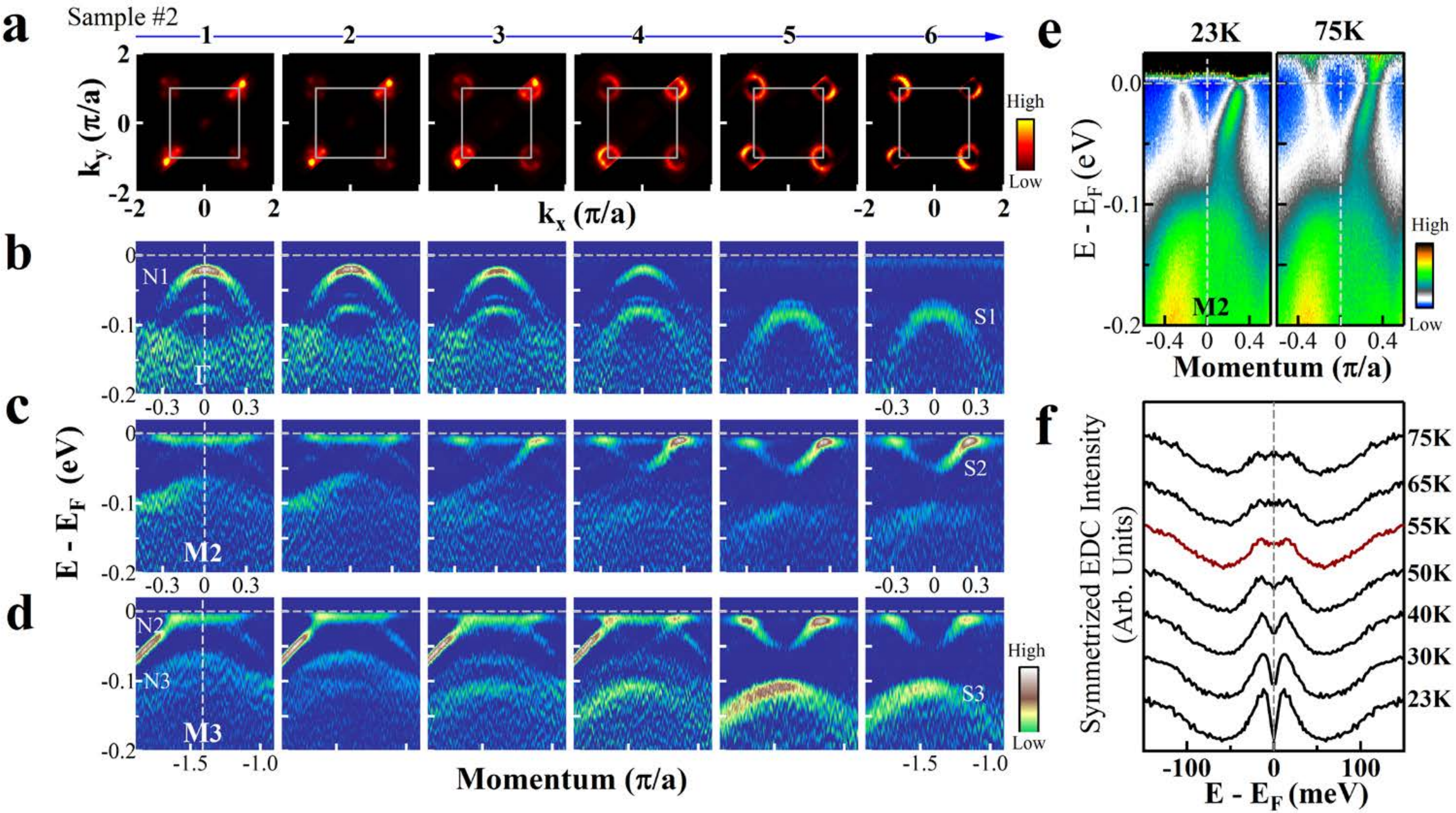
for the double-layer FeSe/$SrTiO_3$ ……

Way 1:
Vacuum Annealing of Double-Layer FeSe/$SrTiO_3$ at 350C for different Times



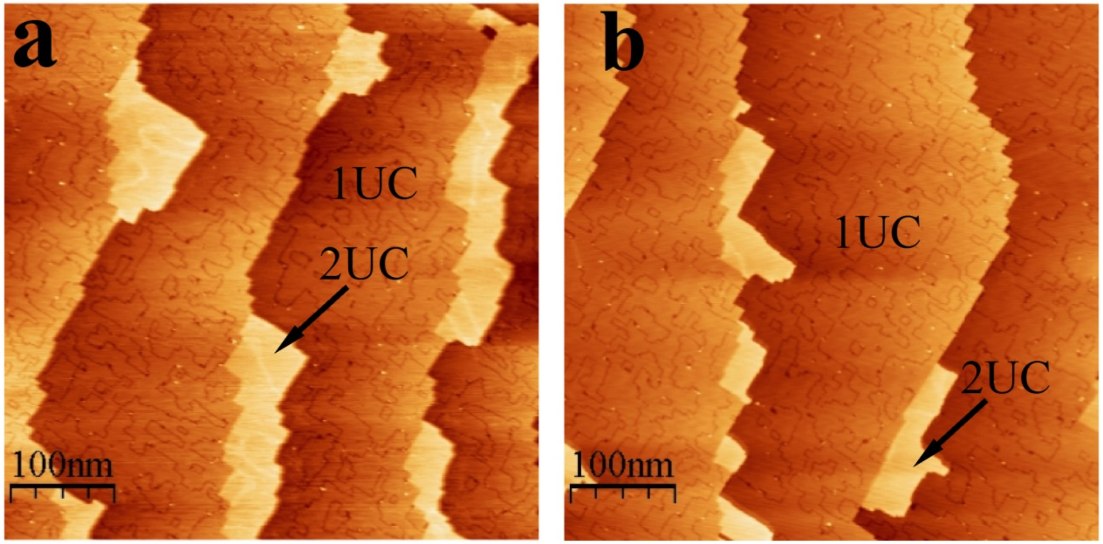
1. The N phase remains dominant after annealing;
2. There is a transition from the N phase to the S phase.

Way 2:
Vacuum Annealing of Double-Layer FeSe/$SrTiO_3$ at Increasing Temperature



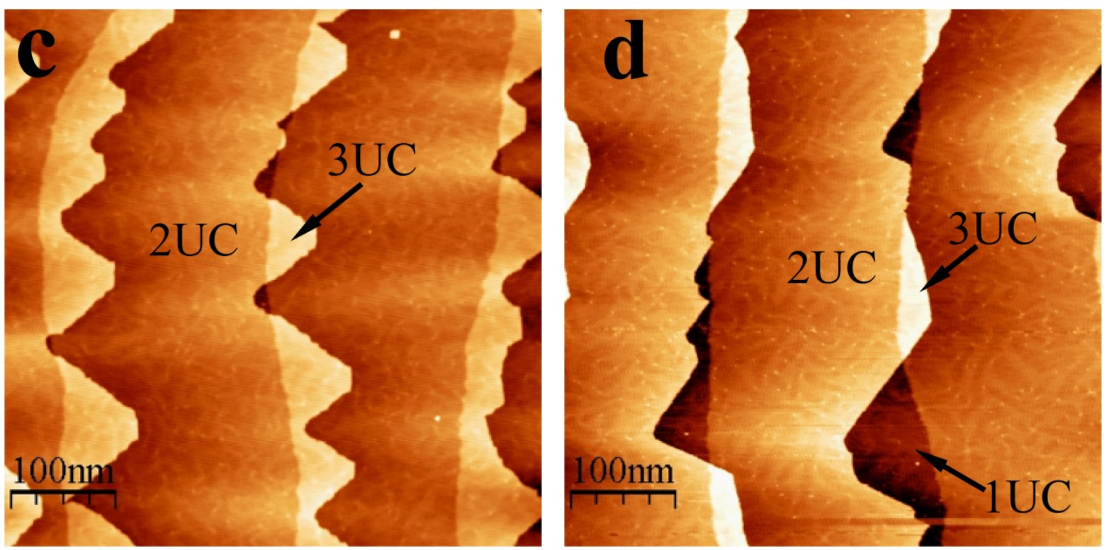
1. The N Phase transforms completely into pure S phase;
2. It becomes superconducting.

STM Observations During Annealing Process



STM images before (a) and after (b) the sample was annealed at 450 C for 15 hours.

No obvious FeSe evaporation

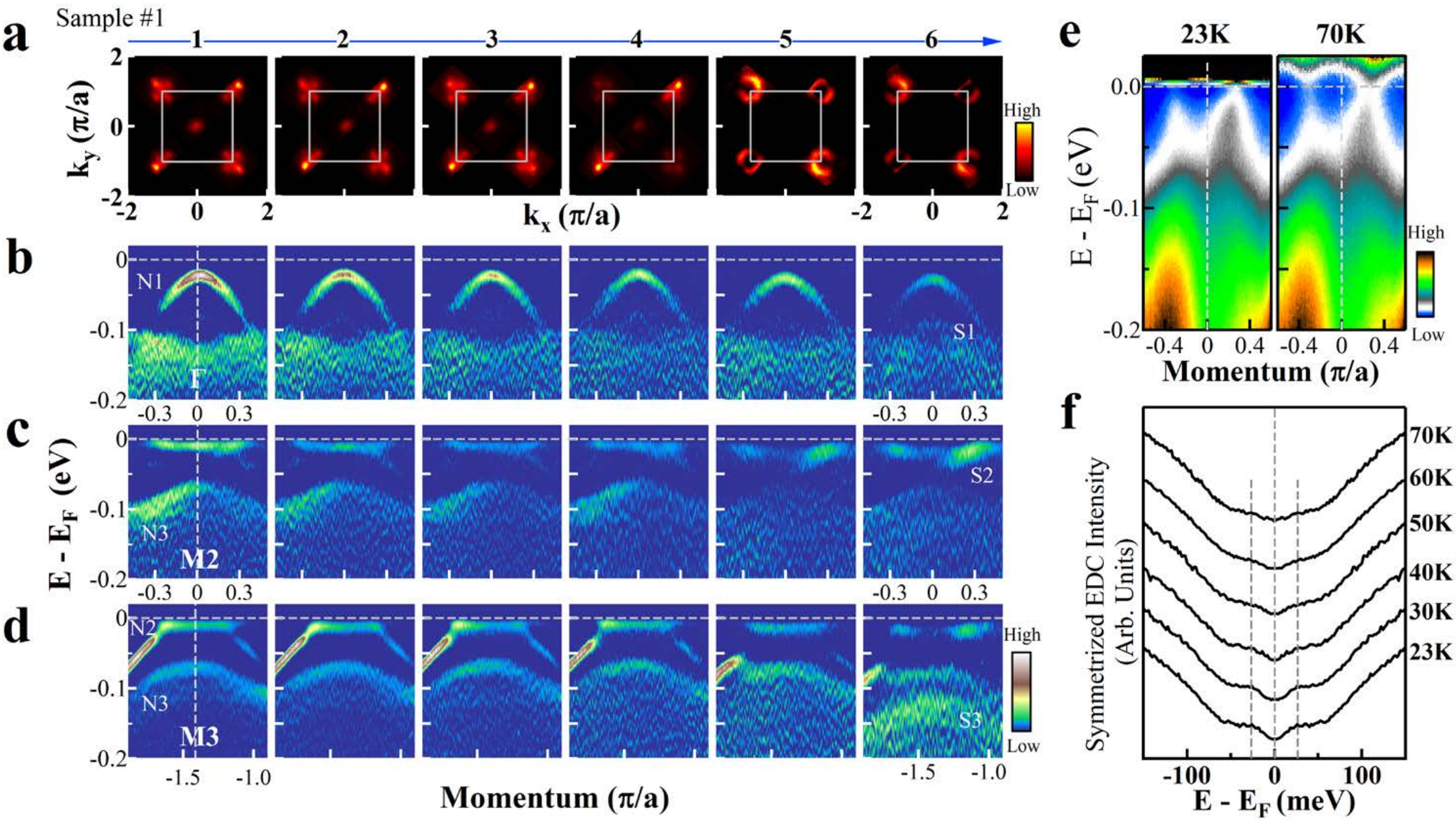


STM images before (c) and after (d) the sample was annealed at 500 C for 3 hours.

Obvious FeSe evaporation

Way 1:

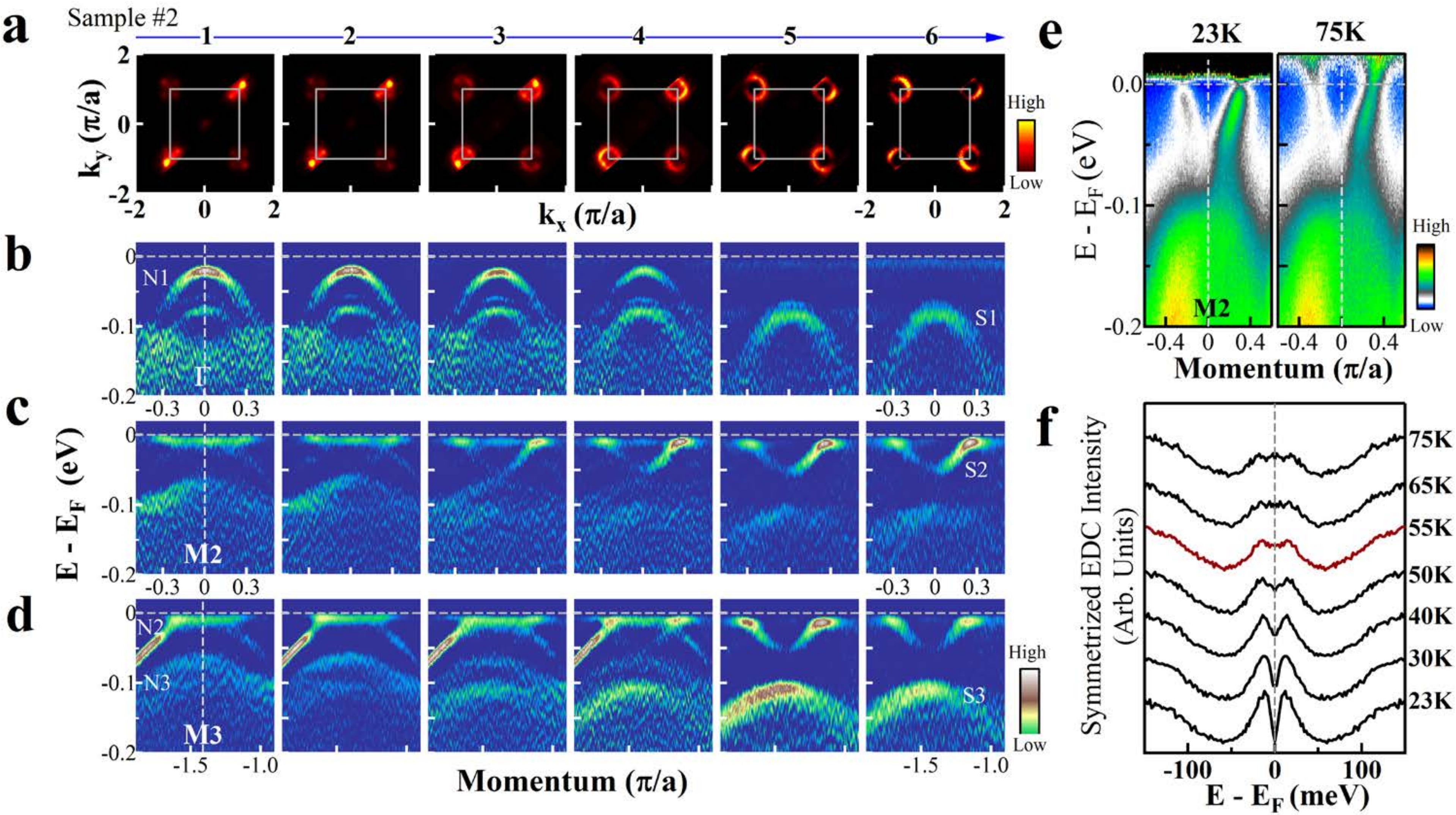
Vacuum Annealing of Double-Layer FeSe/$SrTiO_3$ at 350C for different Times



At 350C, the double-layer FeSe/SrTiO3 film remains intact

Way 2:

Vacuum Annealing of Double-Layer FeSe/$SrTiO_3$ at Increasing Temperature



1. Most likely, wwo-layers may become evaporated into single-layer;

2. The superconductivity may be caused by exposed single-layer region.

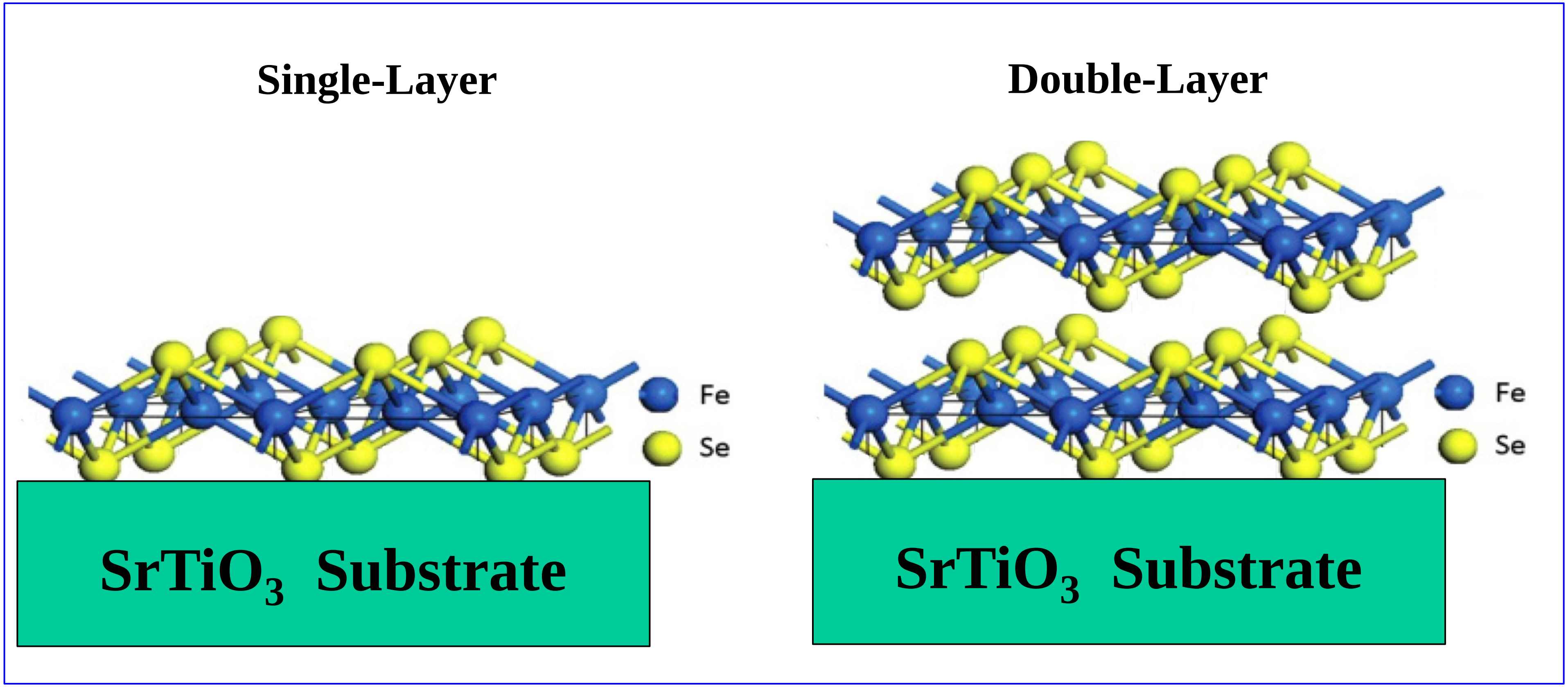
- **It is much harder to transform double-layer from the N phase into the S phase than the single-layer FeSe/SrTiO$_3$.**

- **It is possible to transform double-layer FeSe/SrTiO$_3$ from the N phase into the S phase.**

Origin of Electron-Doping during Annealing

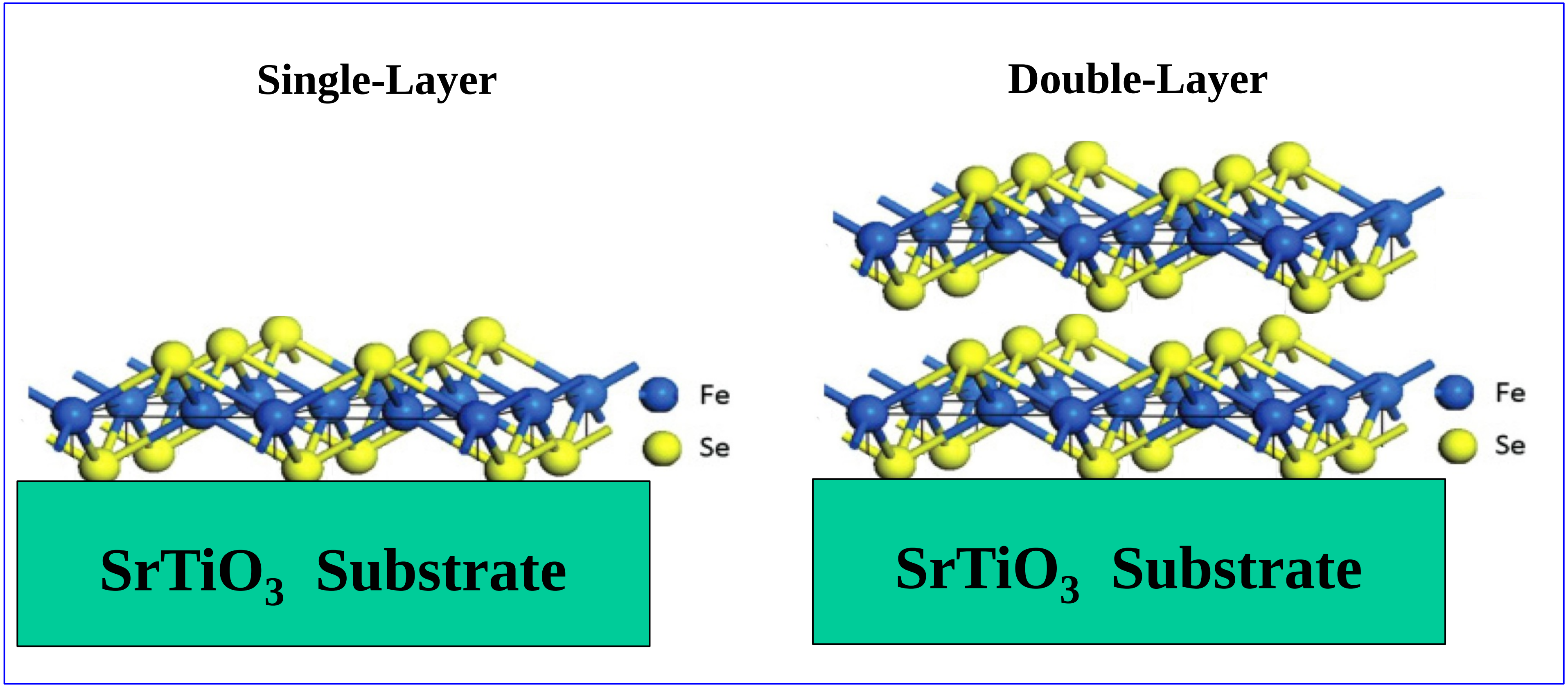
1. **Excess Se removal or Se vacancy Formation in $FeSe_{1-x}$?**
   Less likely

2. **Charge transfer from oxygen loss in $SrTiO_{3-\delta}$?**



Why is Double-Layer $FeSe/SrTiO_3$ Difficult to be Doped?

- **Charge transfer from oxygen loss in $SrTiO_{3-\delta}$;**
- **Charge transfer is shared by two FeSe layers;**
- **The more the number of layers, the more difficult to dope.**



Summary

1. Distinct electronic structure of single-layer FeSe superconductor

Hole-like Fermi surface near Γ is not necessary for high-Tc superconductivity in the Fe-based superconductors.

2. Electronic phase diagram of single-layer FeSe/$SrTiO_3$ established

Two competing and coexisting phases revealed with distinct electronic structures;
Signature of high Tc~65 K observed.

3. Insulator-superconductor transition observed in Single-Layer FeSe/$SrTiO_3$ films

Orbital-selective Mott transition?

4. Dichotomy between single-layer and double-layer FeSe/$SrTiO_3$ Films

Origin of carrier doping from $SrTiO_3$ surface.

Thanks