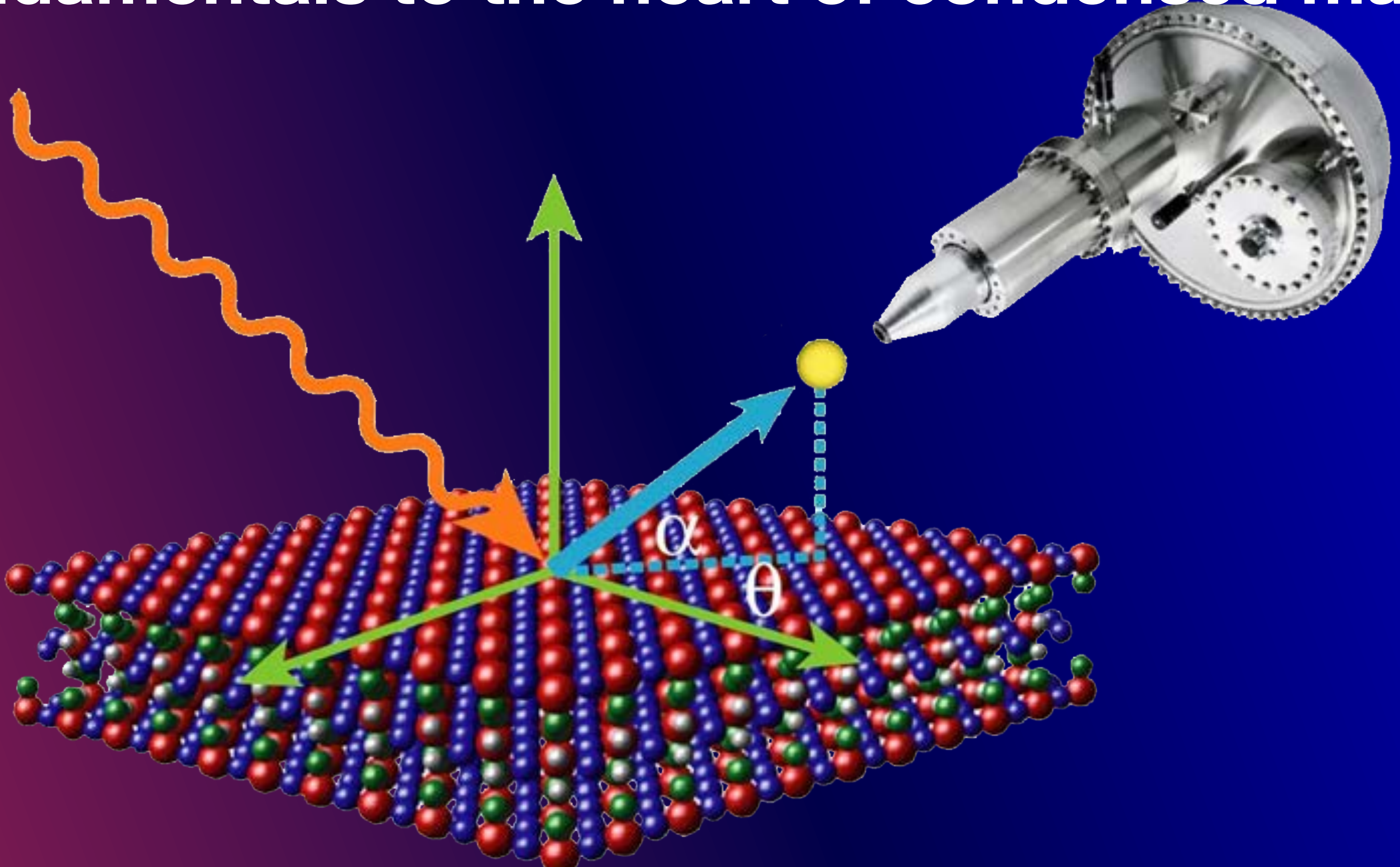
SEVERO OCHOA SCHOOL

Angle Resolved Photoemission Spectroscopy

From fundamentals to the heart of condensed matter



6-7 FEBRERO, 2023

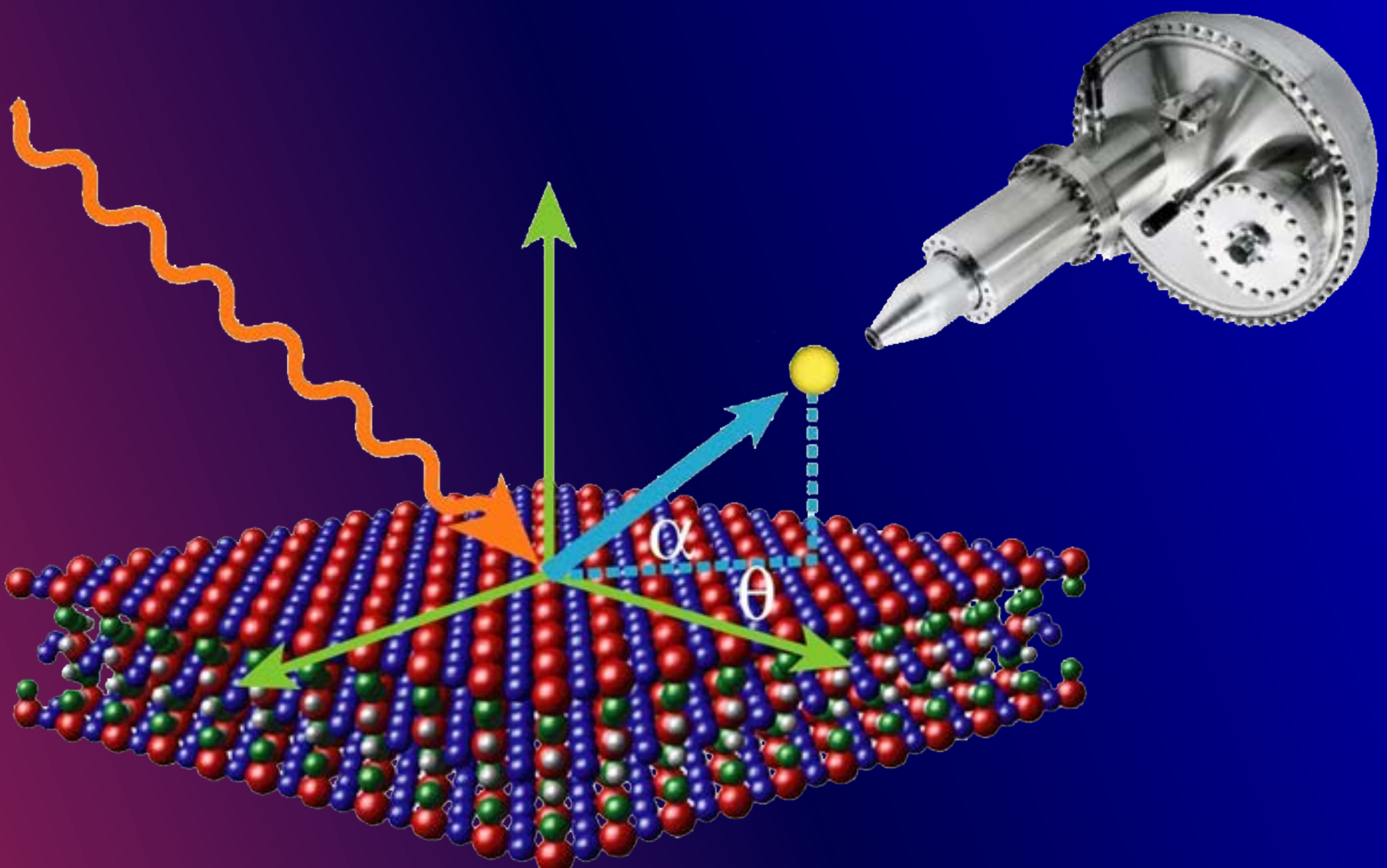
CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

SEVERO OCHOA SCHOOL

INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA

EXCELENCIA SEVERO OCHOA

Lecture #3 : ARPES Studies of Quantum Materials



6-7 FEBRERO, 2023

Concept Question #8

Consider an ARPES experiment being conducting with an electron analyzer resolution of ΔE = 10 meV and an photon bandwidth of ΔE = 2 meV

What is the closest value of the **TOTAL** "effective" energy broadening in the experiment?

A. 10 meV
B. 11 meV
C. 12 meV
D. 13 meV

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Concept Question #12

In your ARPES experiment of a given material, you see a sharp "kink" in the quasiparticle band dispersion, which is a clear signature of some electron-boson interaction.

What information **cannot** be directly obtained from the analysis of your experimental data?

A. The energy of the boson

B. The strength of the electron-boson interaction

C. What kind of boson it is (i.e. phonon, magnon, etc…)

D. None of A, B, and C can be obtained from the data

E. All of A, B, and C can be obtained from the data

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Concept Question #13

Which of the following material properties would be directly observable using ARPES?

1. The exciton binding energy in a semiconductor
2. The strength of electron-phonon interactions in a metal
3. The magnon (spin-wave) dispersion in an antiferromagnet
4. The interacting quasiparticle band structure
5. The electronic band structure as calculated by DFT

A. 2 & 4

B. 1, 3, 4, & 5

C. 2, 3, 4, &5

D. 3, 4, & 5

E. All of the above

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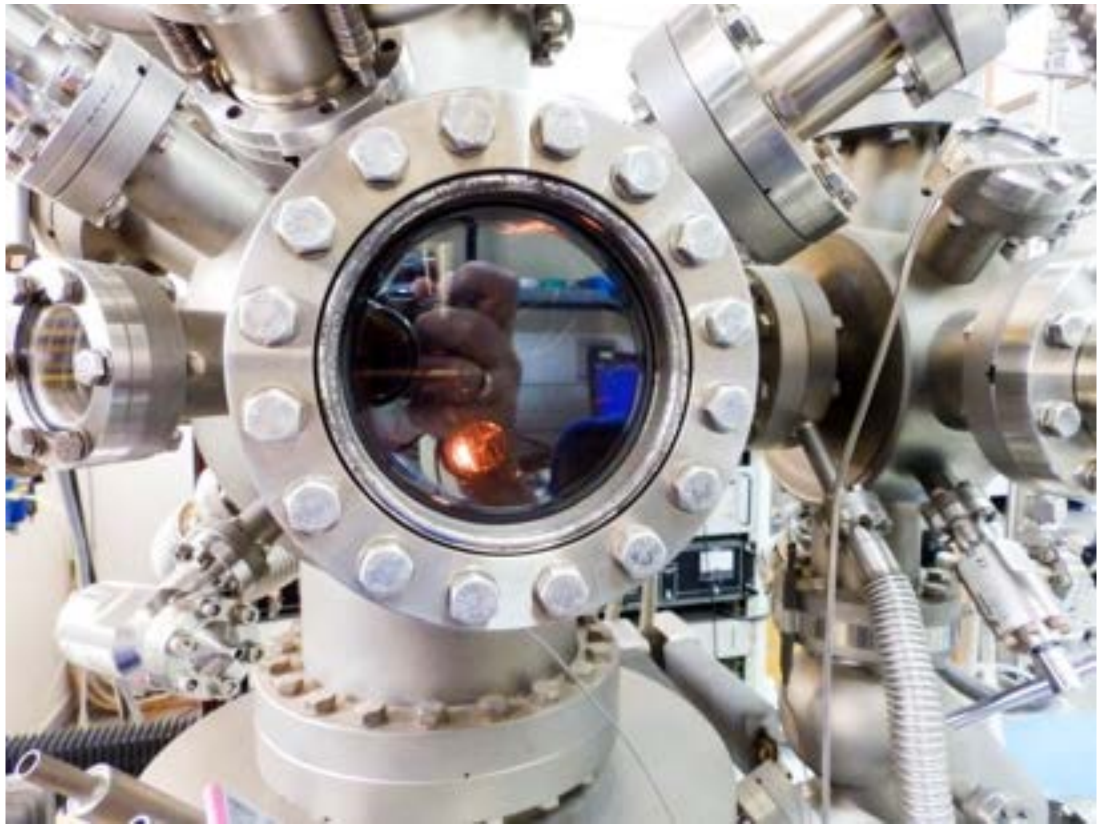
Concept Question #6

ARPES measurements need to take place in ultrahigh vacuum (10^{-10} torr or better). Which of the following is the **most important** factor which determines the level of vacuum needed to perform experiments?



A. The scattering / absorption of photoelectrons traveling inside the chamber

B. The operation of the electron analyzer

C. The absorption of vacuum ultraviolet (VUV) photons used for photoemitting the electrons

D. The scattering of electrons from adsorbed molecules at the sample's surface

E. All of the above are equally important

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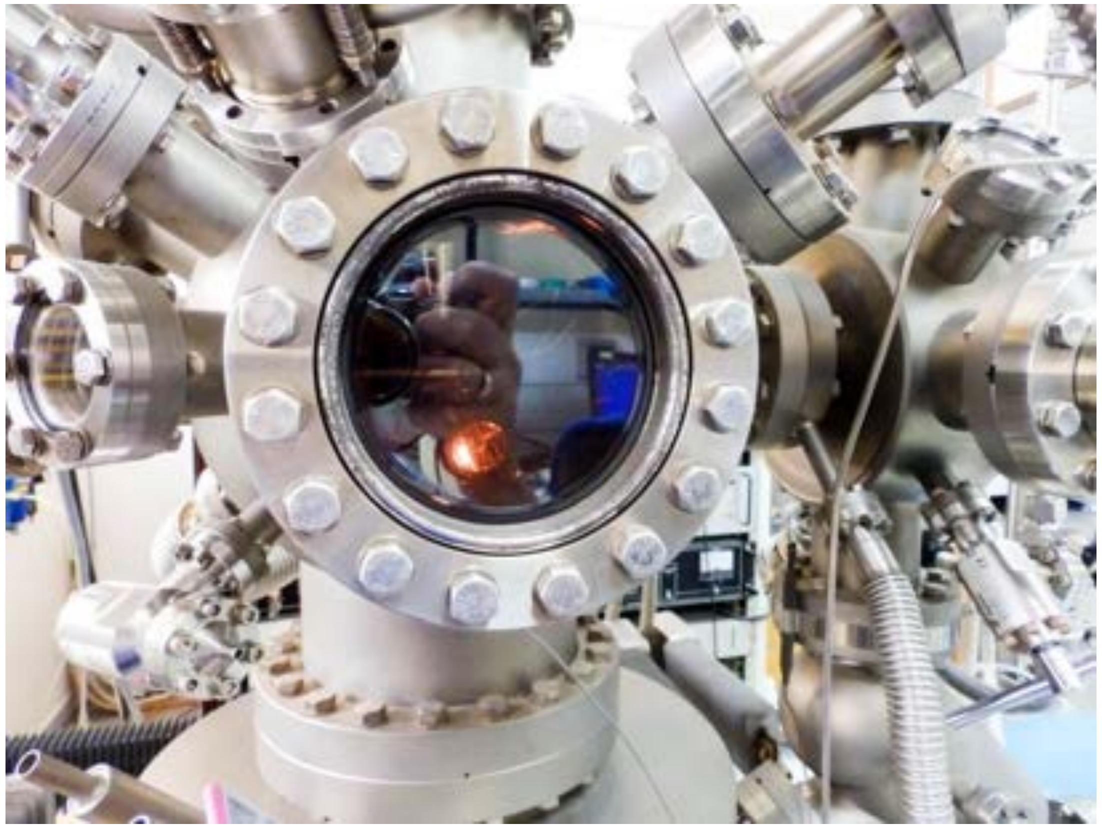

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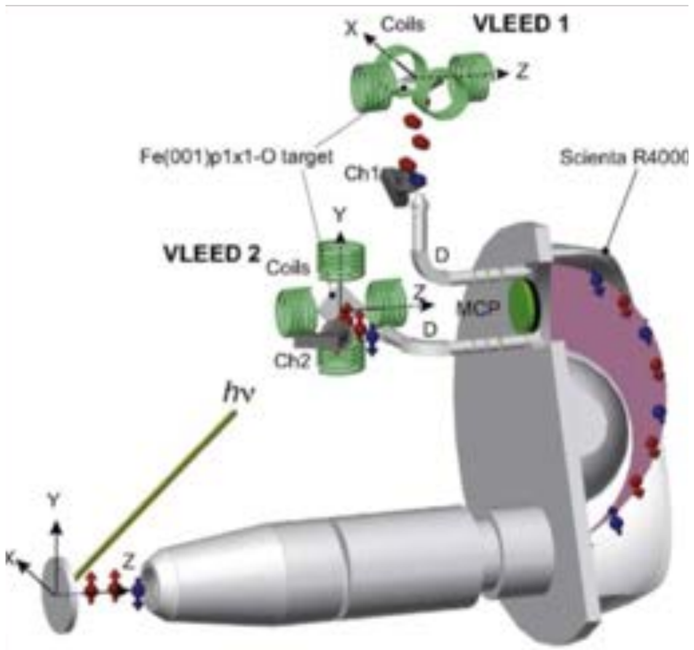
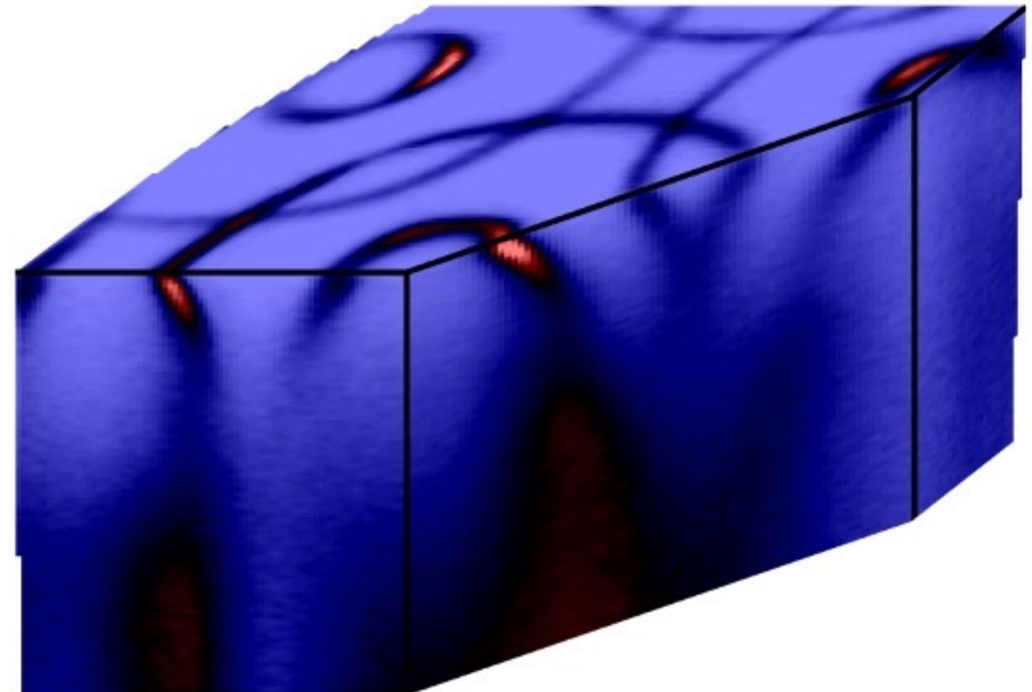
The "Ecosystem" of Modern Photoemission

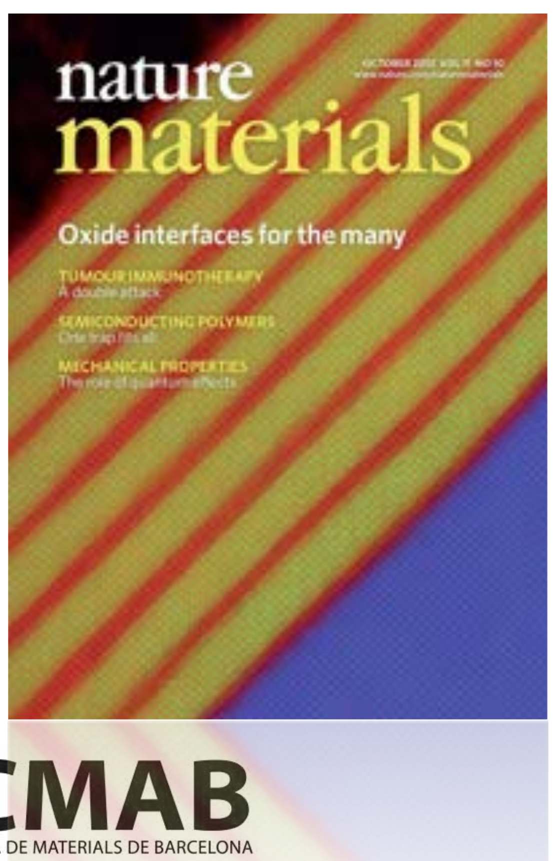
New Instrumentation



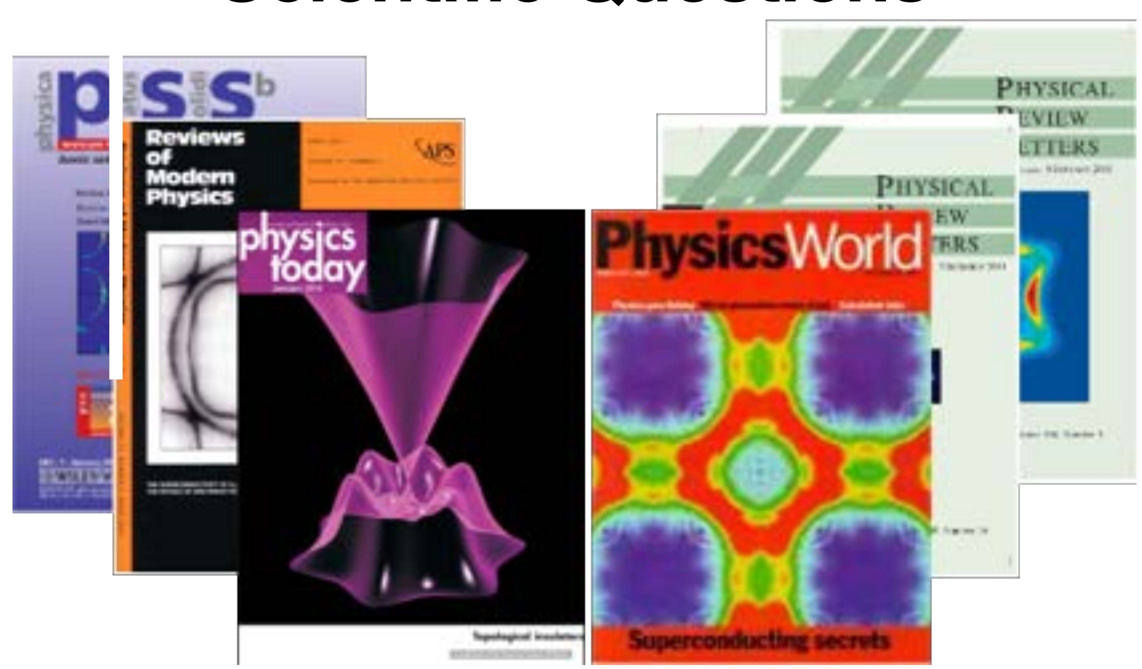
Data Processing



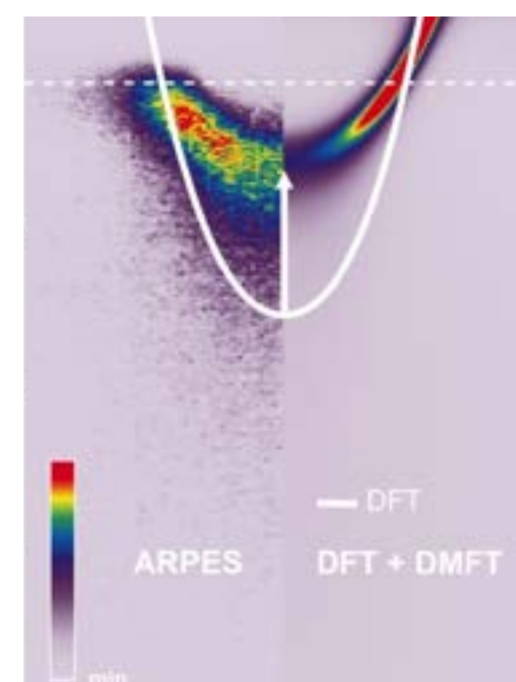
New Materials



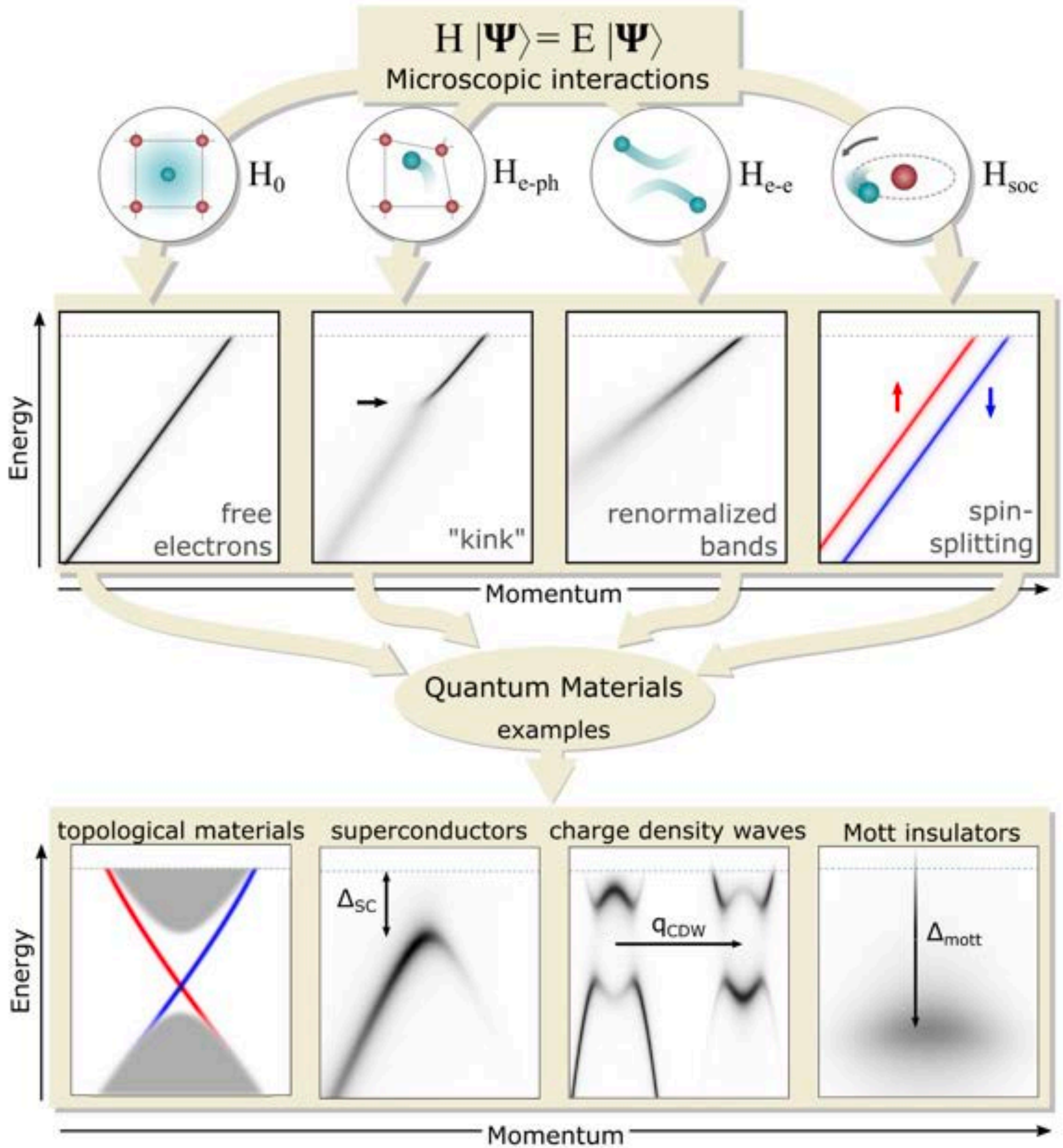
Scientific Questions



State-of-the-art Theory



From Interactions to Quantum Materials



J. A. Sobota, Y. He, and Z.-X. Shen. Reviews of Modern Physics 93, 025006 (2021)

Example #1 : High-Temperature Cuprate Superconductors

- Evolution from the parent Mott insulating state
- *d*-wave superconducting gap
- discovery of the pseudogap

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- “Fermi Surface Engineering” in strained Sr_2RuO_4
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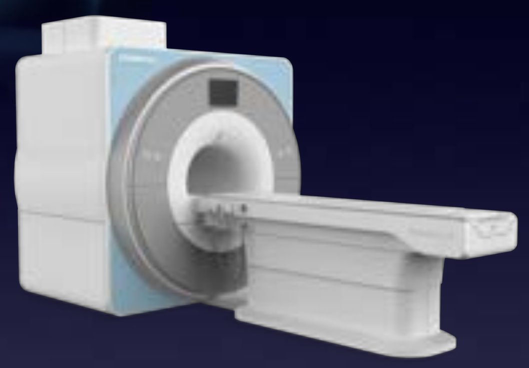
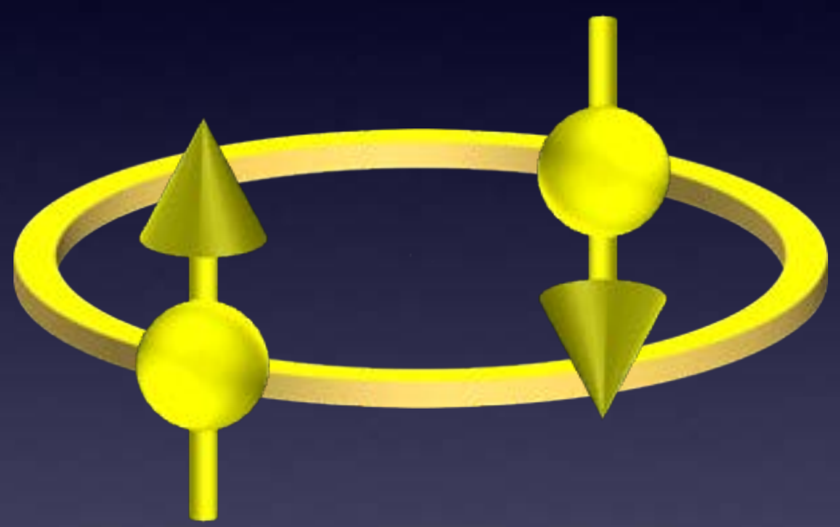
- Pairing enhancement in FeSe / $SrTiO_3$
- Interfacial Electron-Phonon Coupling in FeSe / $SrTiO_3$

superconductivity : macroscopic quantum phenomena

magnetic fields

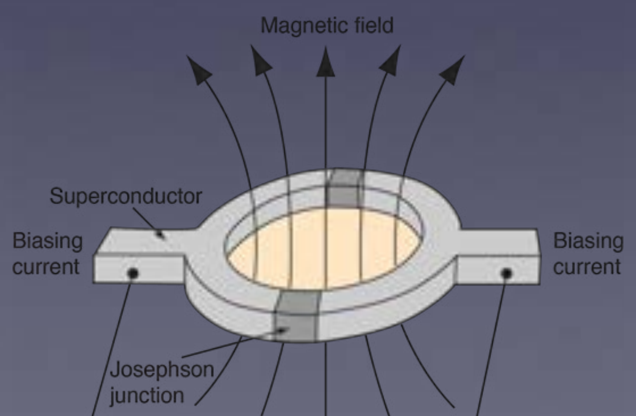


energy applications



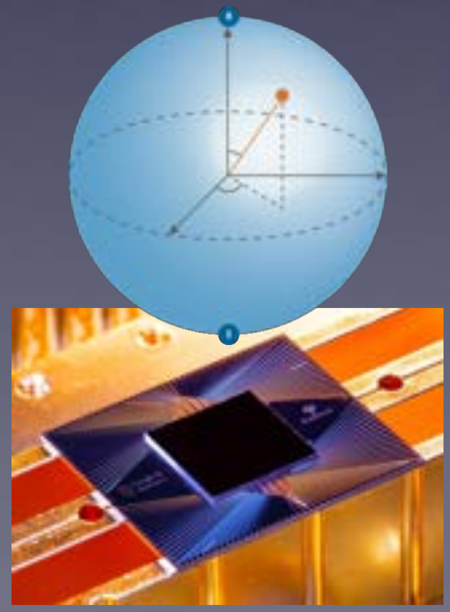
quantum sensors

transition edge sensor

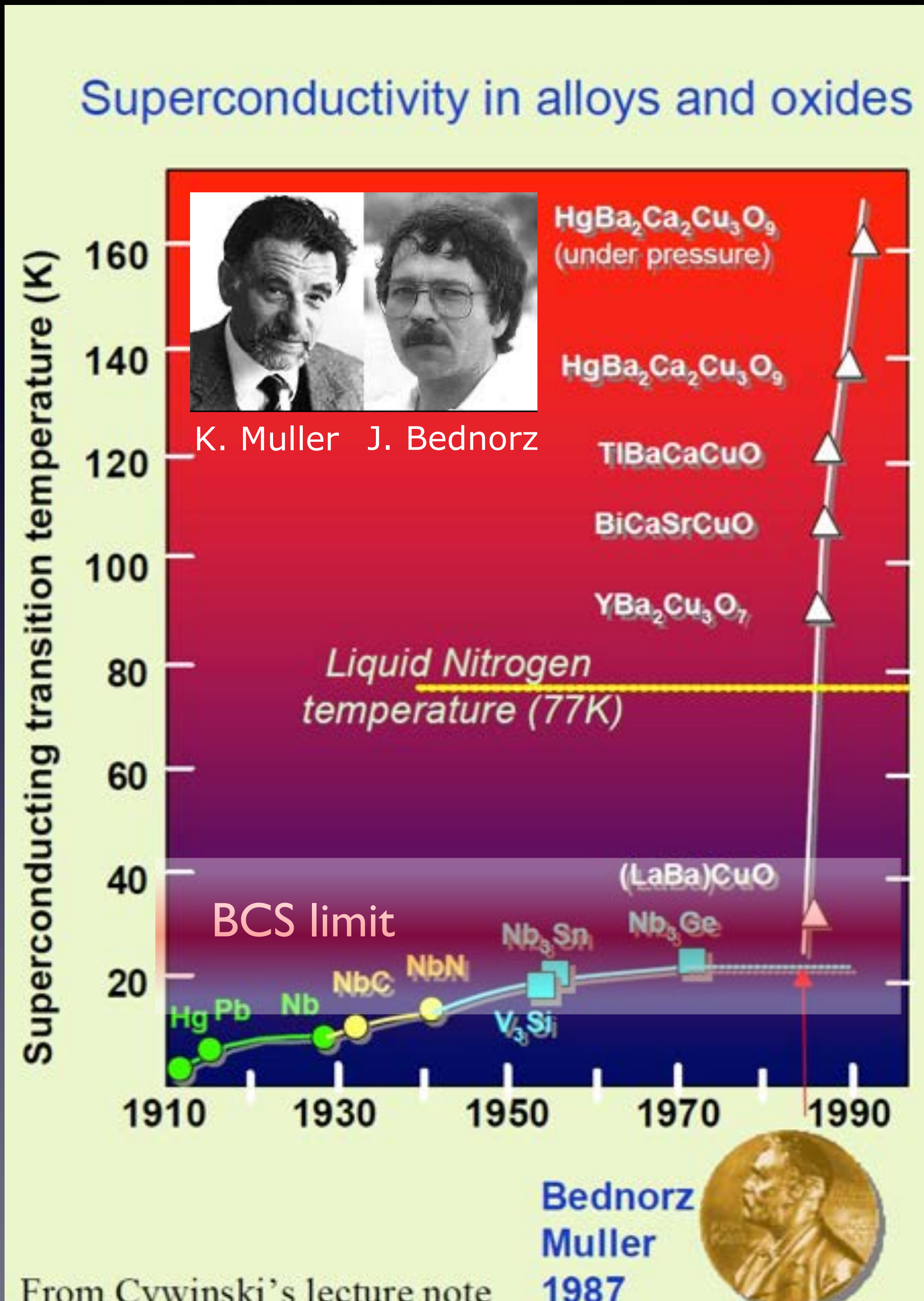


Superconducting Quantum Interference Device (SQUID)

quantum computing

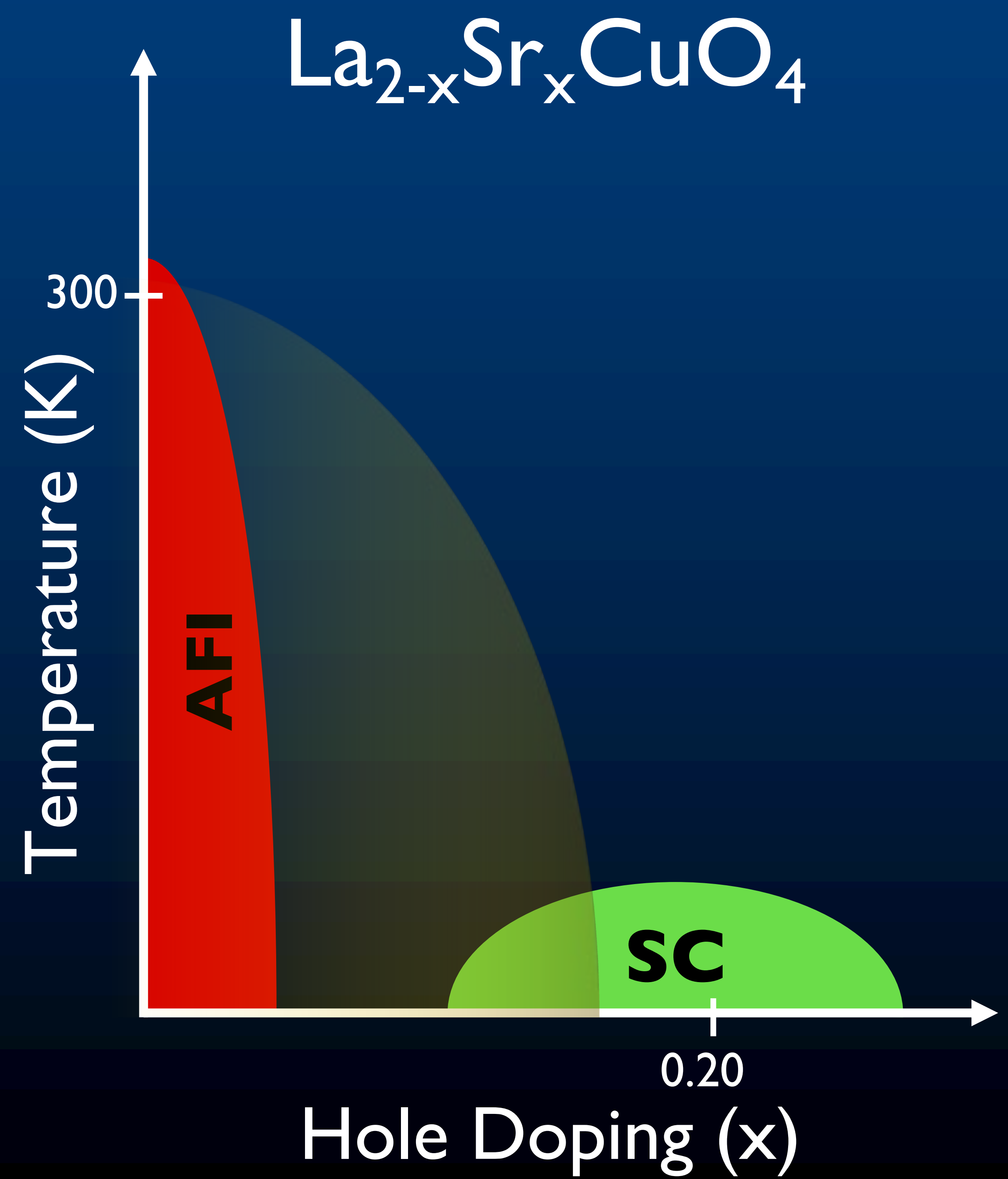


High-Tc Cuprate Superconductors : The “Hydrogen Atom” of Quantum Materials

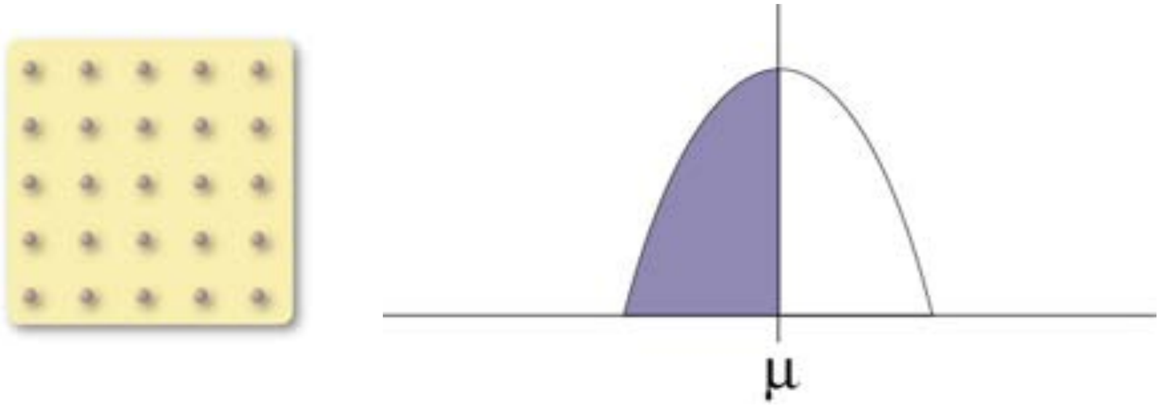


- Conventional ***“independent electron”*** band theory that works so well for materials like silicon fails completely for cuprates
- Jump-started research on the many-body physics (quantum materials) - “physics of the many”
- Motivated the discovery of many other families of “quantum materials”

Many-Body Interactions in Cuprates : Strong Correlations

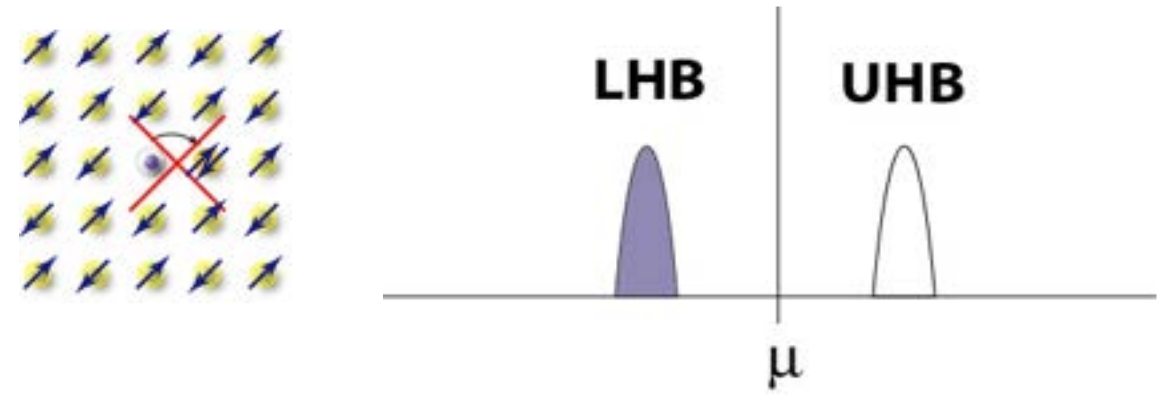


One-Electron Prediction :



Half-filled metal with ~ 3 eV bandwidth

Real Situation :



Mott insulator with ~ 2 eV gap

Breakdown of independent electron paradigm in cuprates

$$\boldsymbol{H\psi = E\psi}$$

$$\boldsymbol{H} = \sum_{i=1}^{N}\left(-\frac{\hbar^2}{2m}\nabla i^2 - Ze2\sum_{R}\frac{1}{|ri-R|}\right) + \frac{1}{2}\sum_{i\neq j}\frac{e2}{|ri-rj|}$$

independent electrons – indeed unreasonable approximation?

John Hubbard

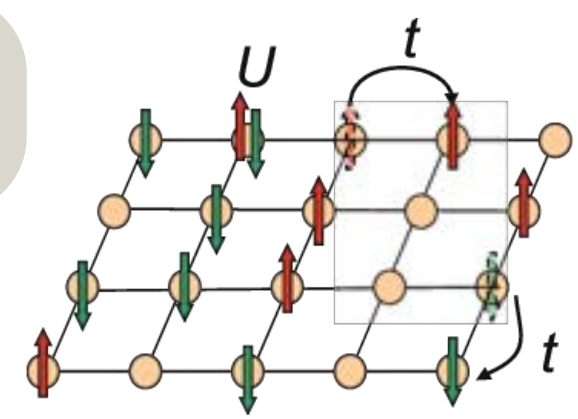
High-Tc is an extension of the long standing problem of insulating oxides – "Mott Insulators"

P.W. Anderson,1987

$$\mathcal{H} = -t\sum_{i,\sigma}\left(c_{i\sigma}^{\dagger}c_{i+1\sigma} + h.c.\right) + U\sum_{i} n_{i\uparrow}n_{i\downarrow}$$

Source of the strong pairing attraction **?**

The "hydrogen atom" model of strongly correlated electrons



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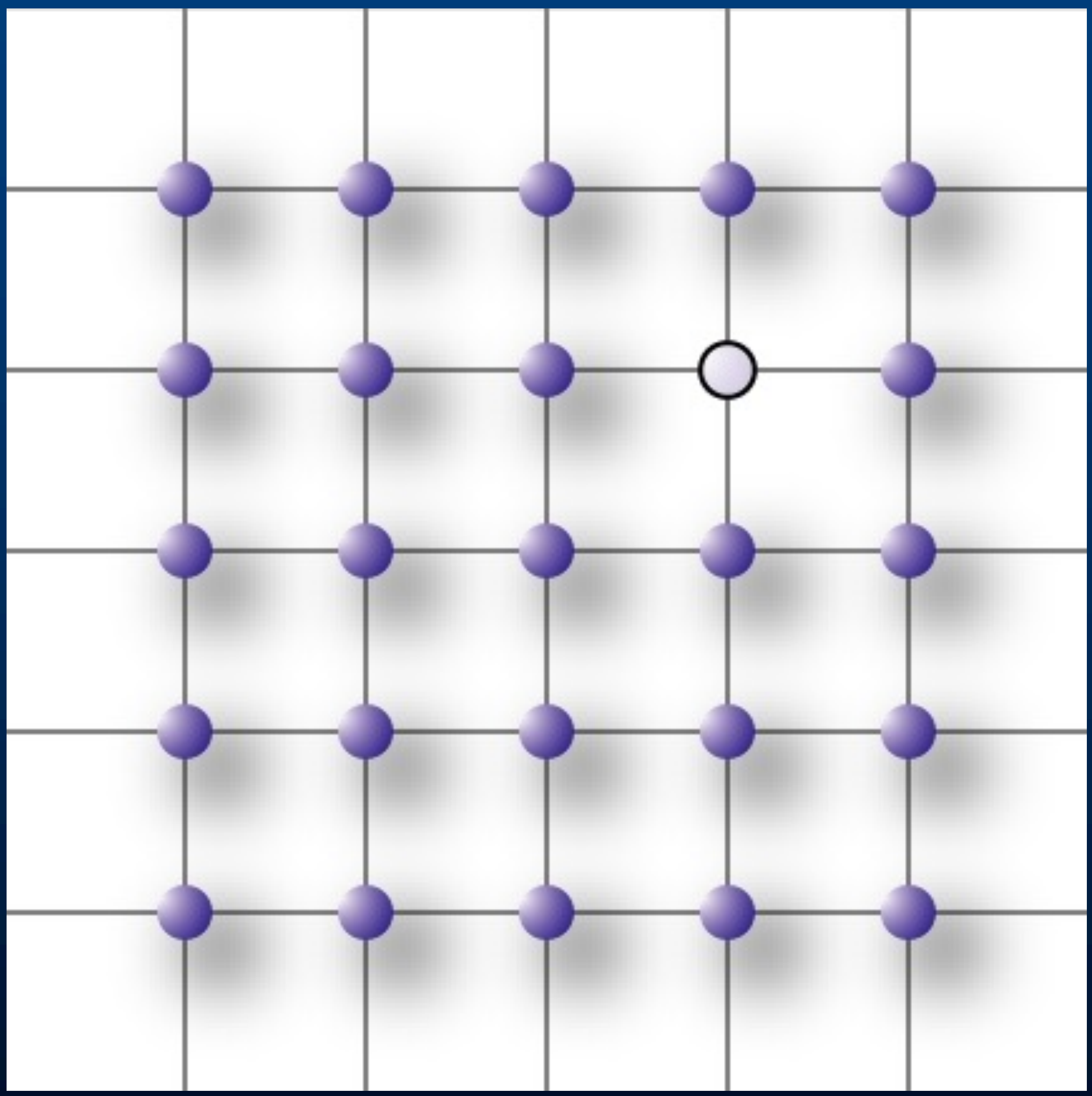
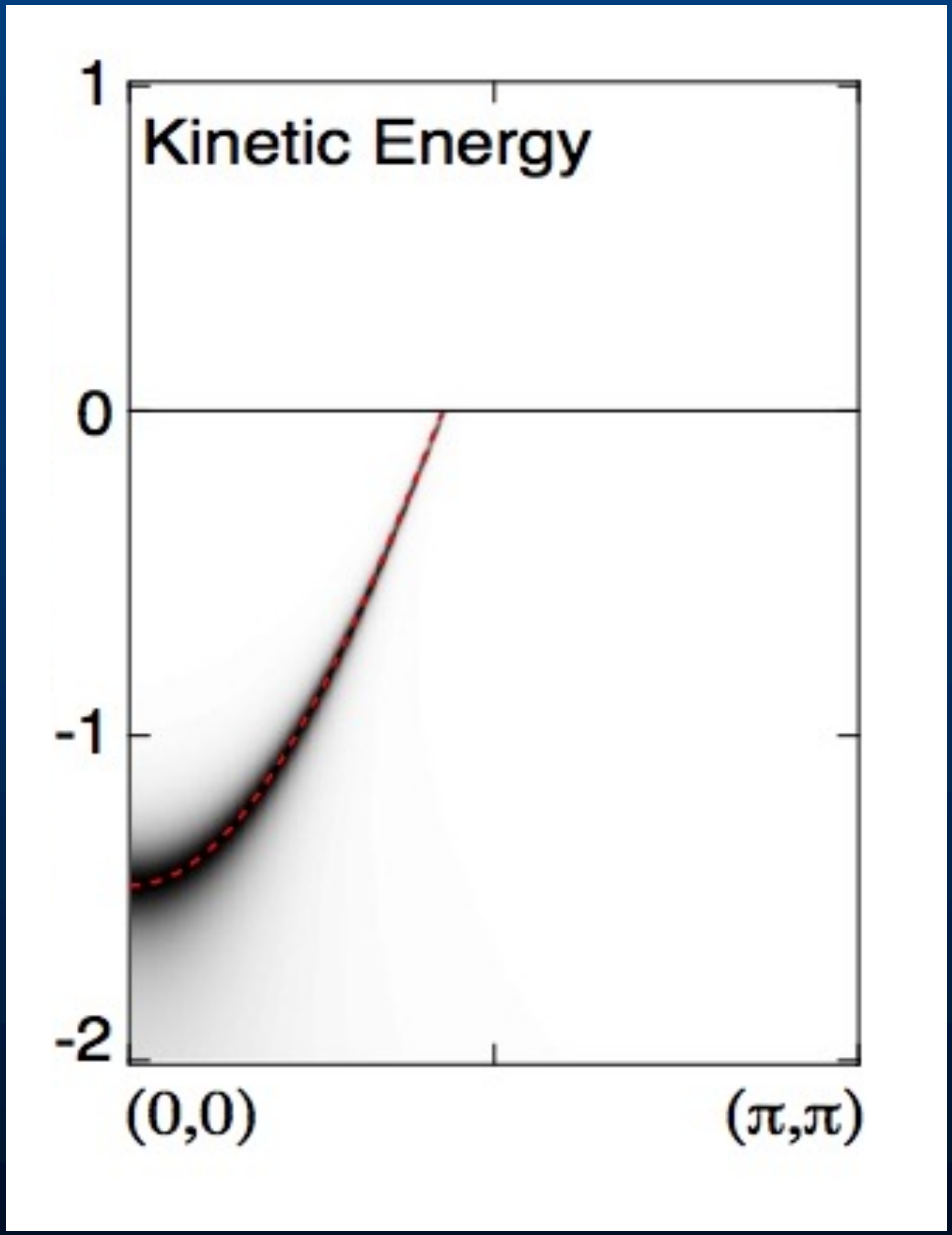
Courtesy of ZX Shen

Band Structure Predictions : Non-Interacting

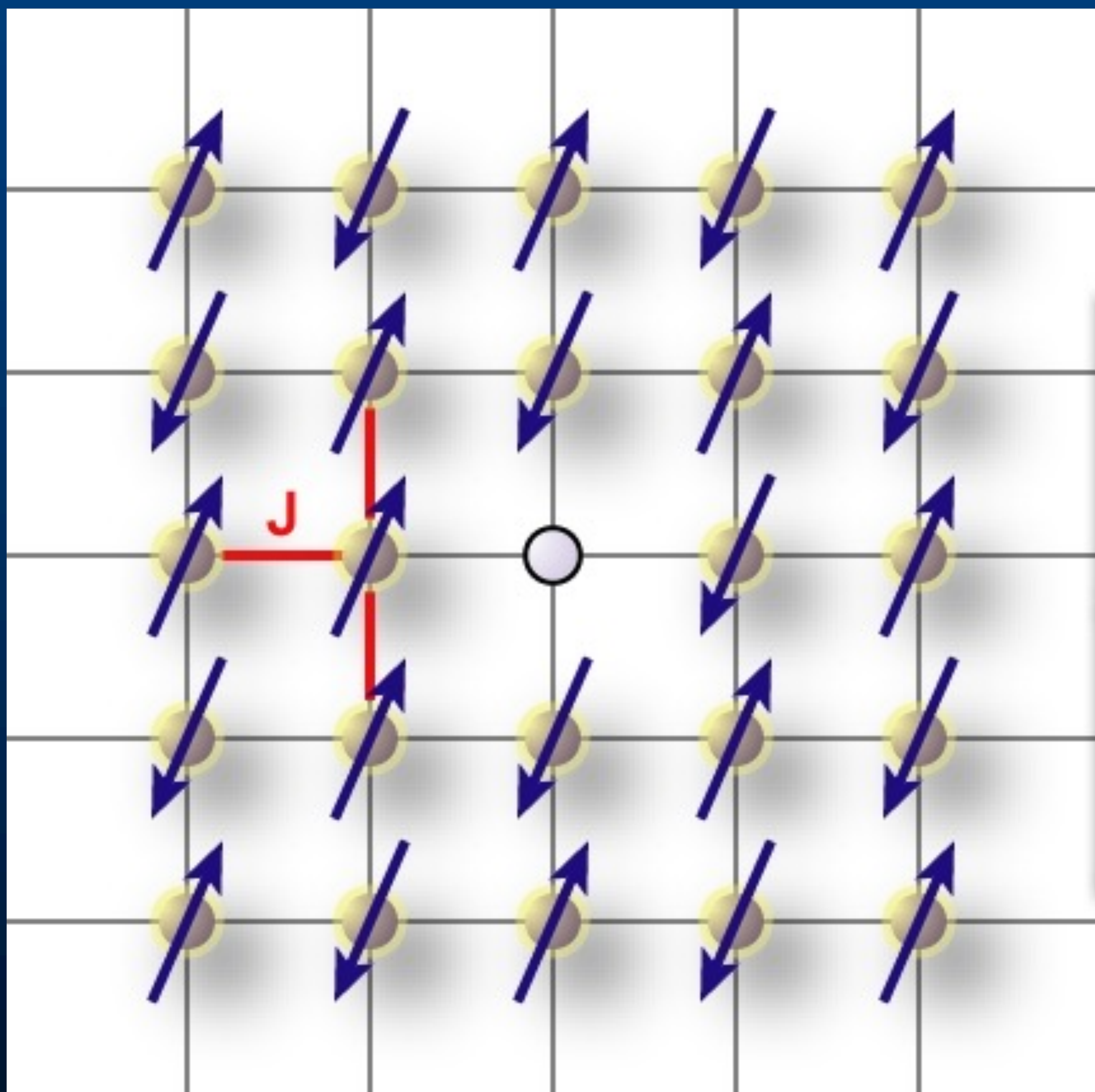
Kinetic Energy Only

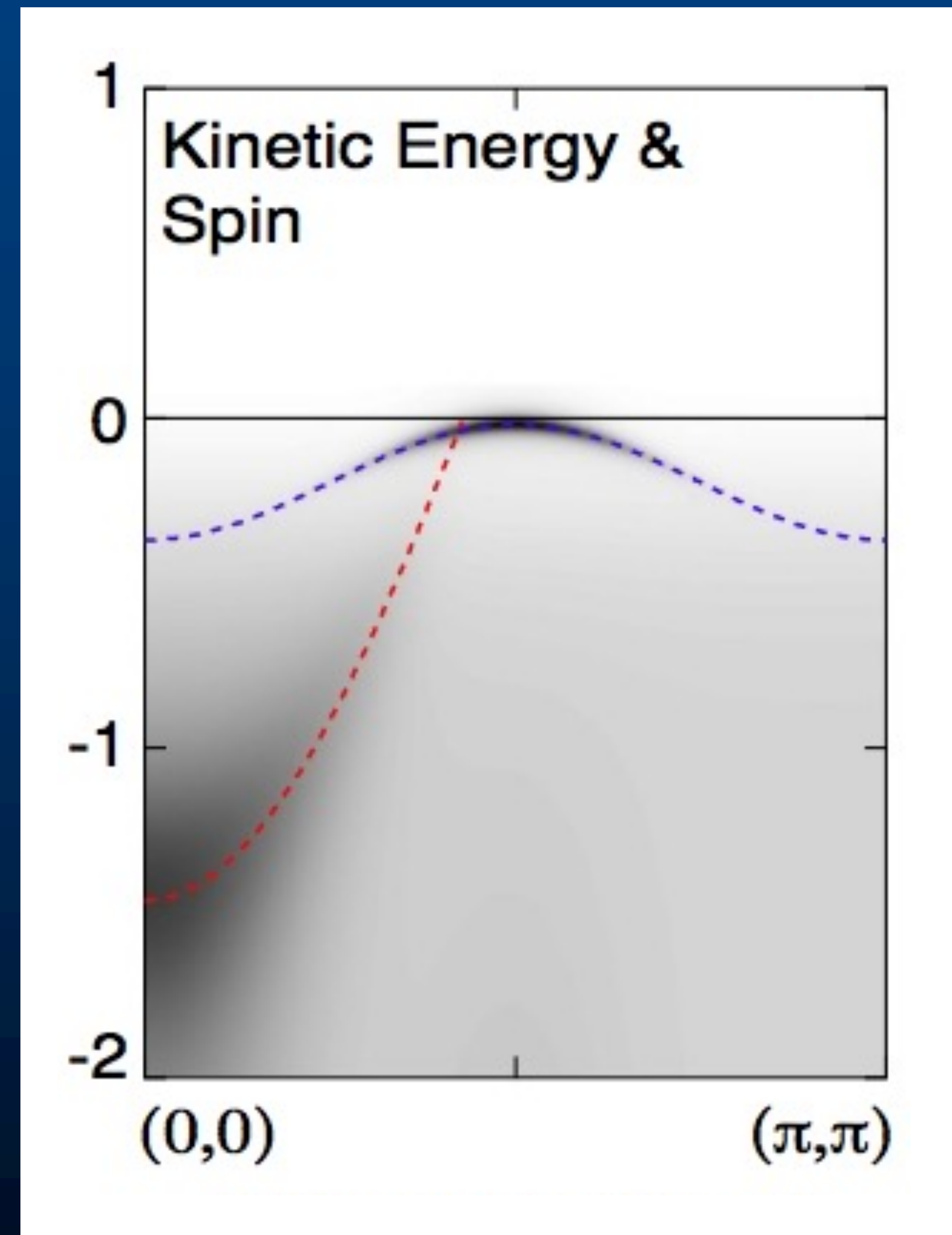




Mott Insulator : Magnetic Interactions

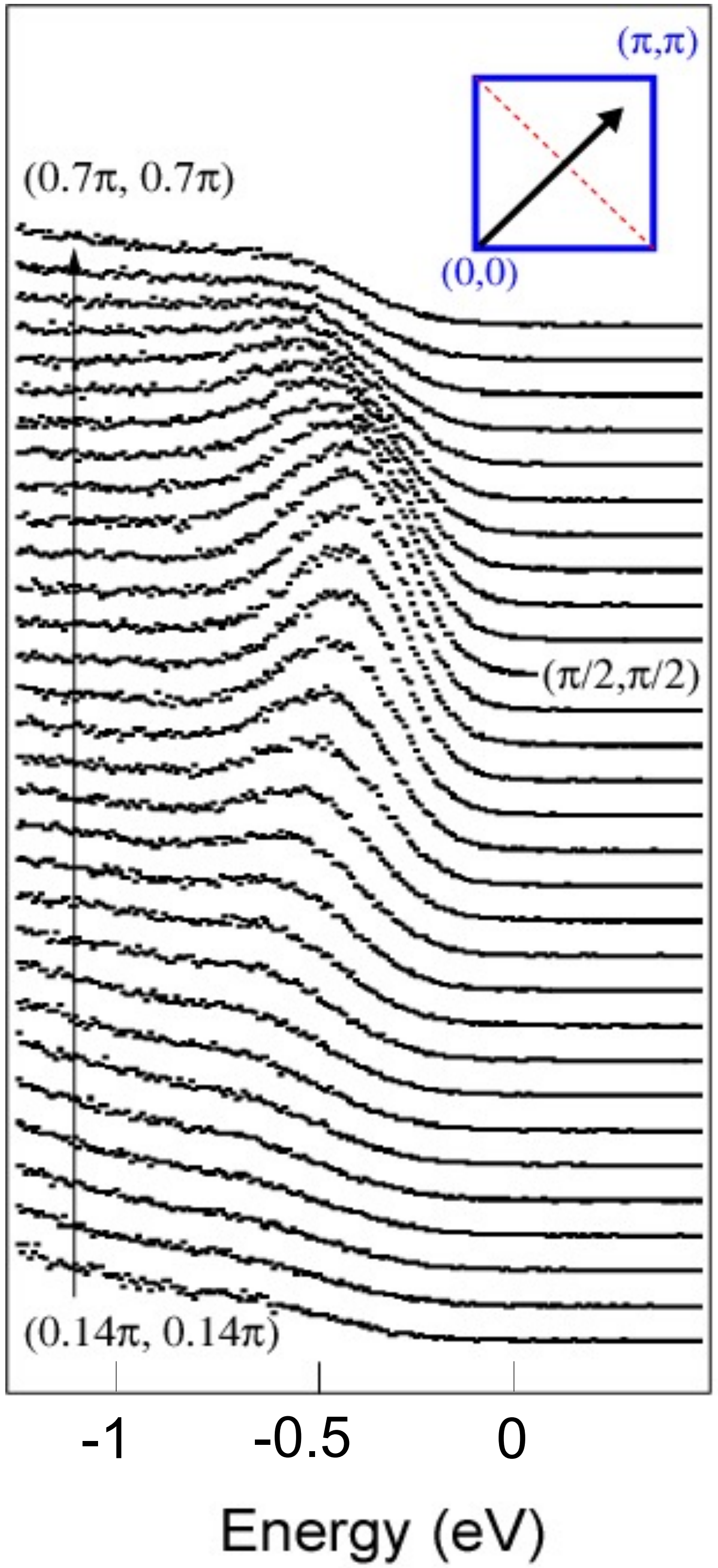
KE & Magnetism

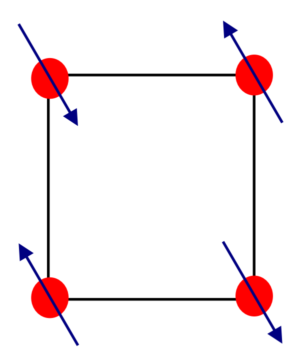


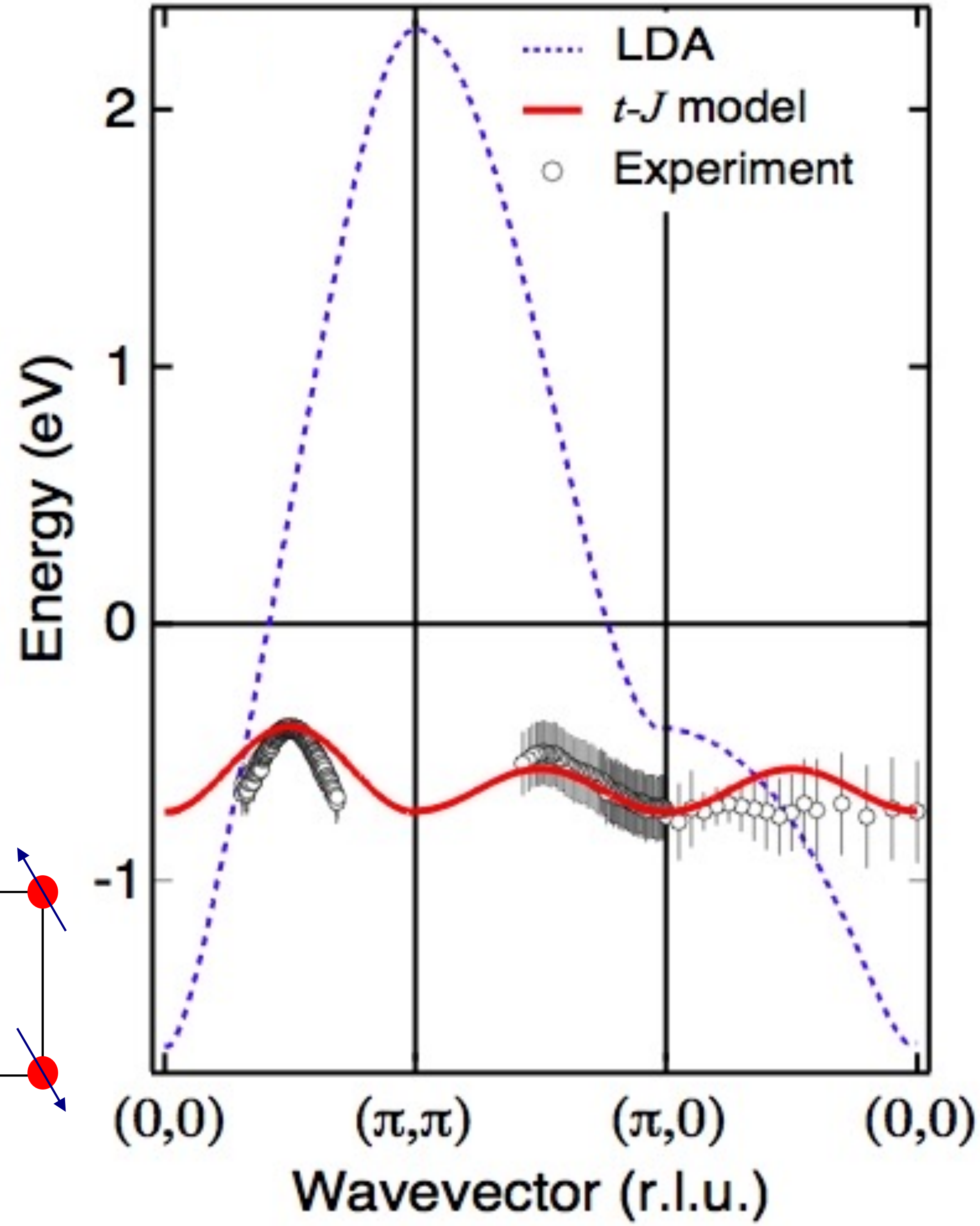


$$t\text{-}J: \quad \mathcal{H} = -t \sum_{\langle ij\rangle,\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + \text{h.c.}) + J \sum_{\langle ij\rangle,\sigma} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4})$$

A Single Hole in the Mott Insulator

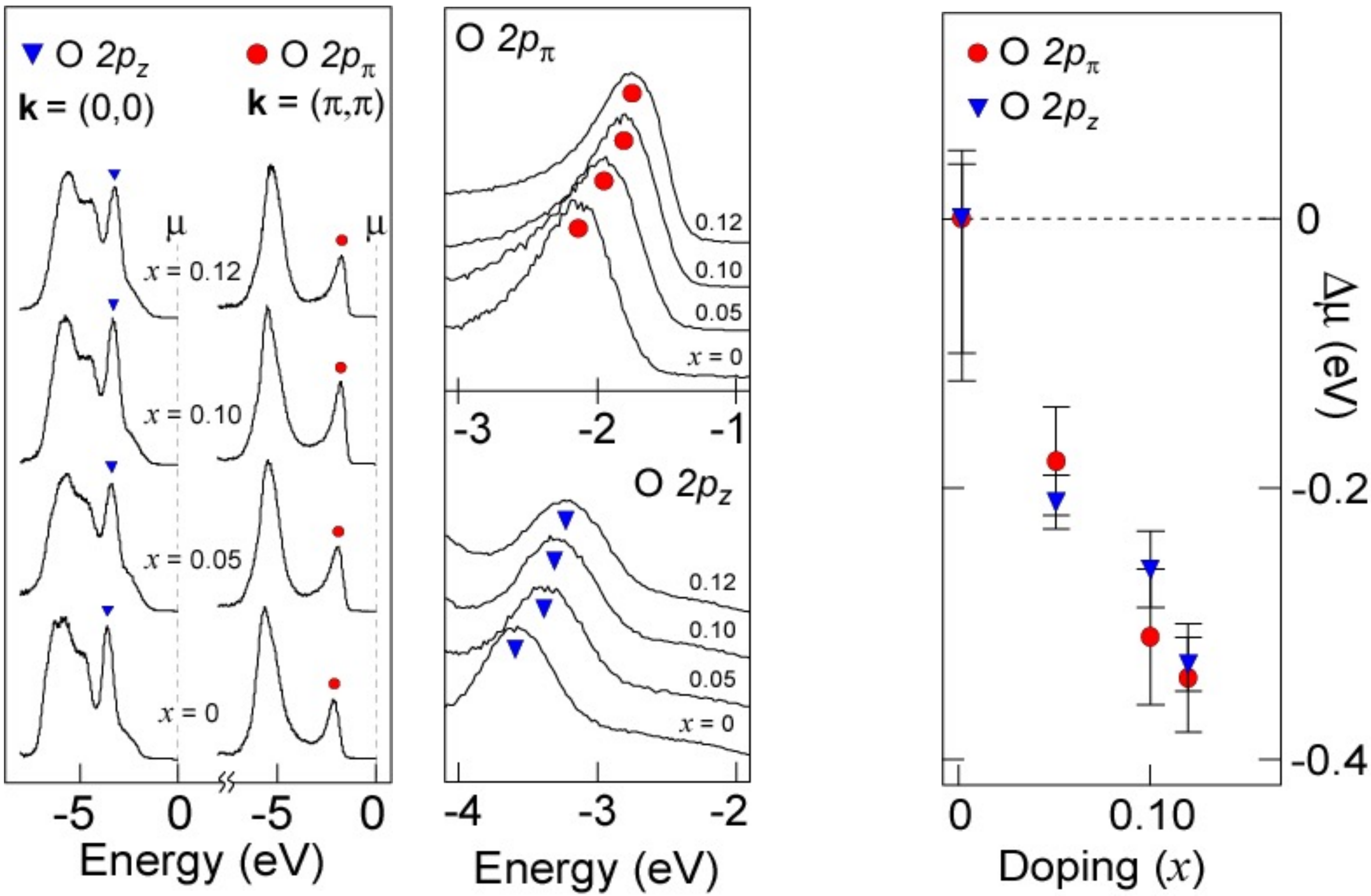




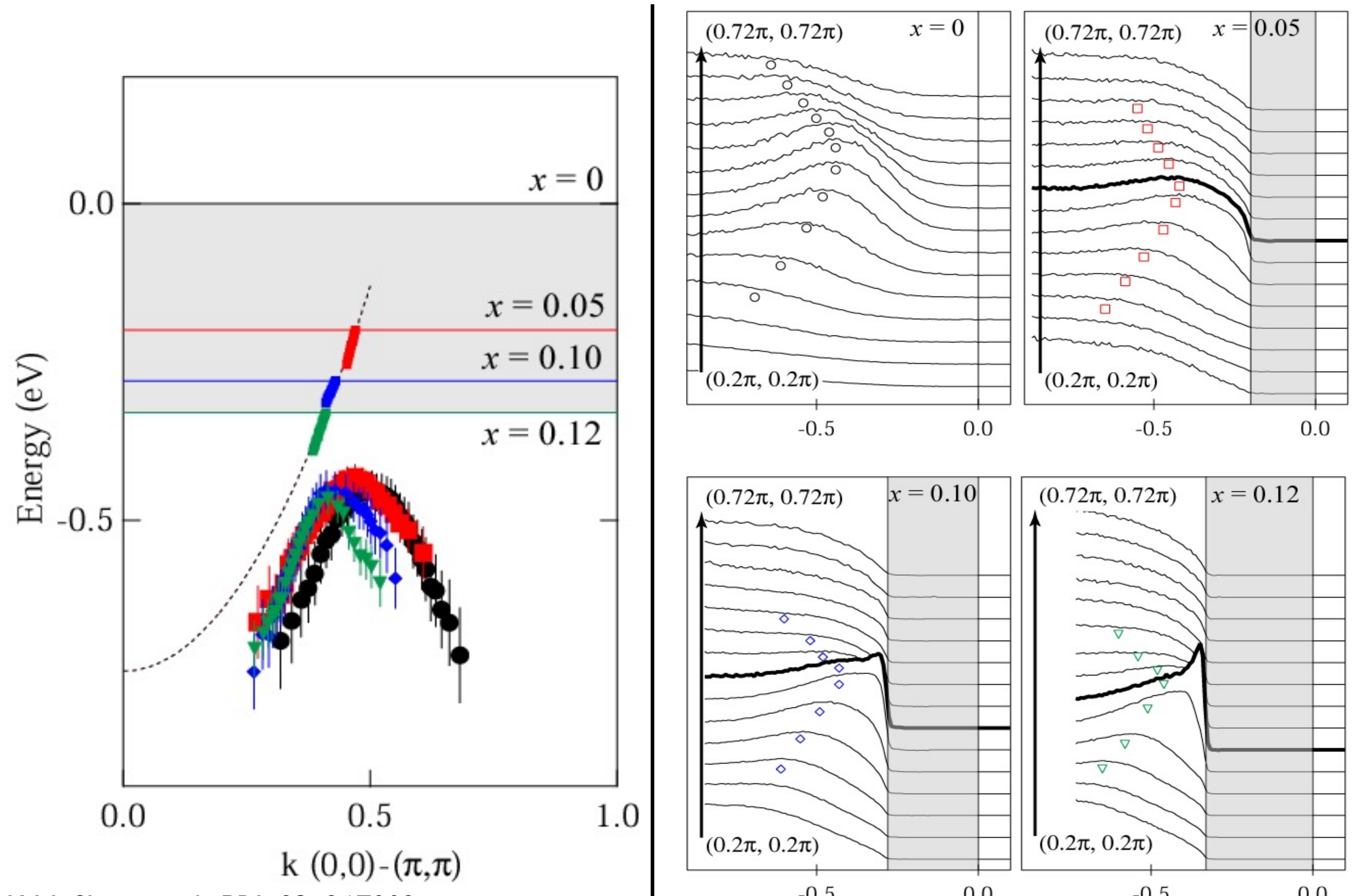


K.M. Shen *et al., Science* **307**,

Chemical Potential Shift : $O2p_\pi$ & $O2p_z$



Evolution of Low Energy States with Doping



K.M. Shen, *et al.*, PRL 93, 267002

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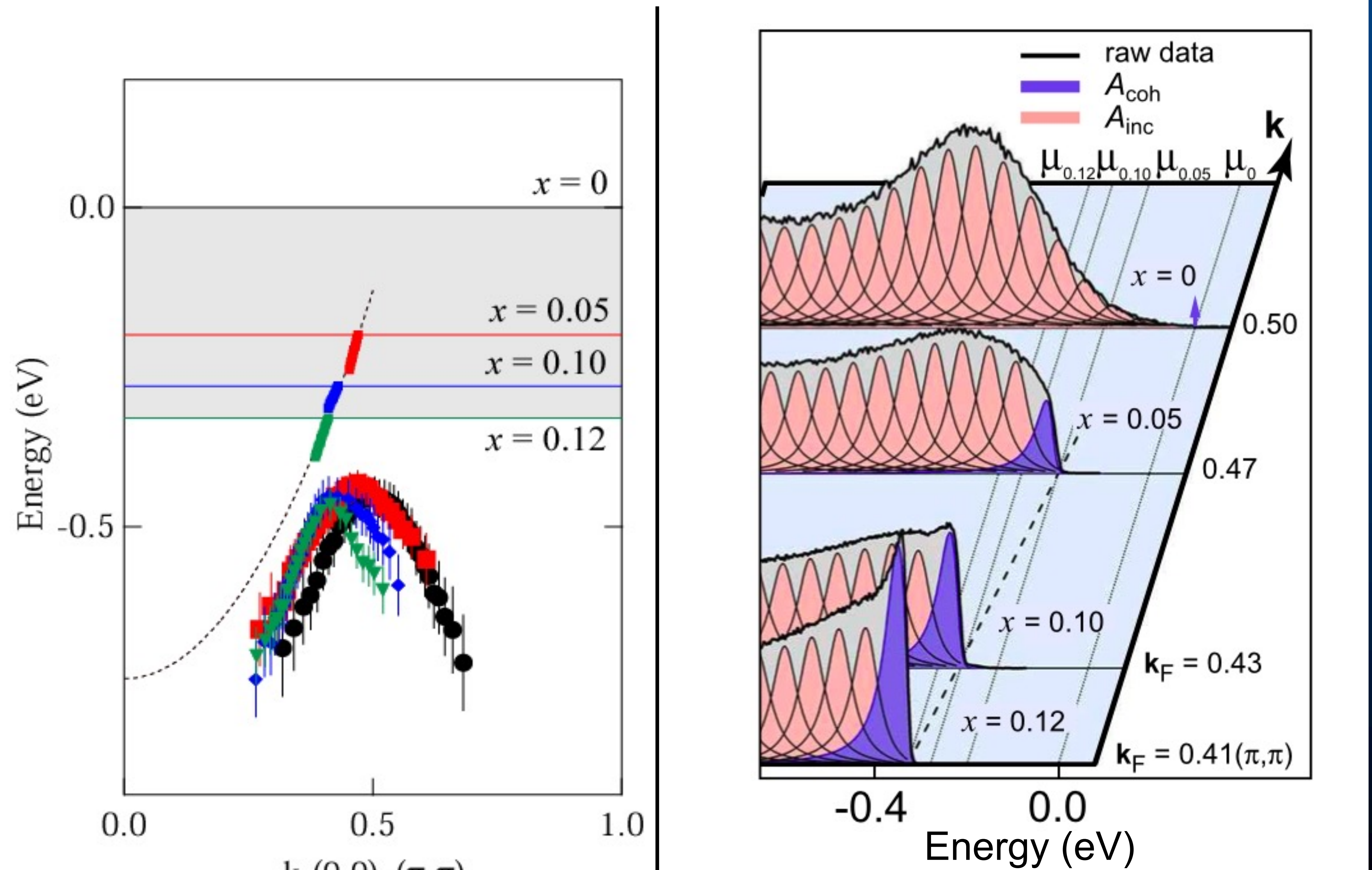
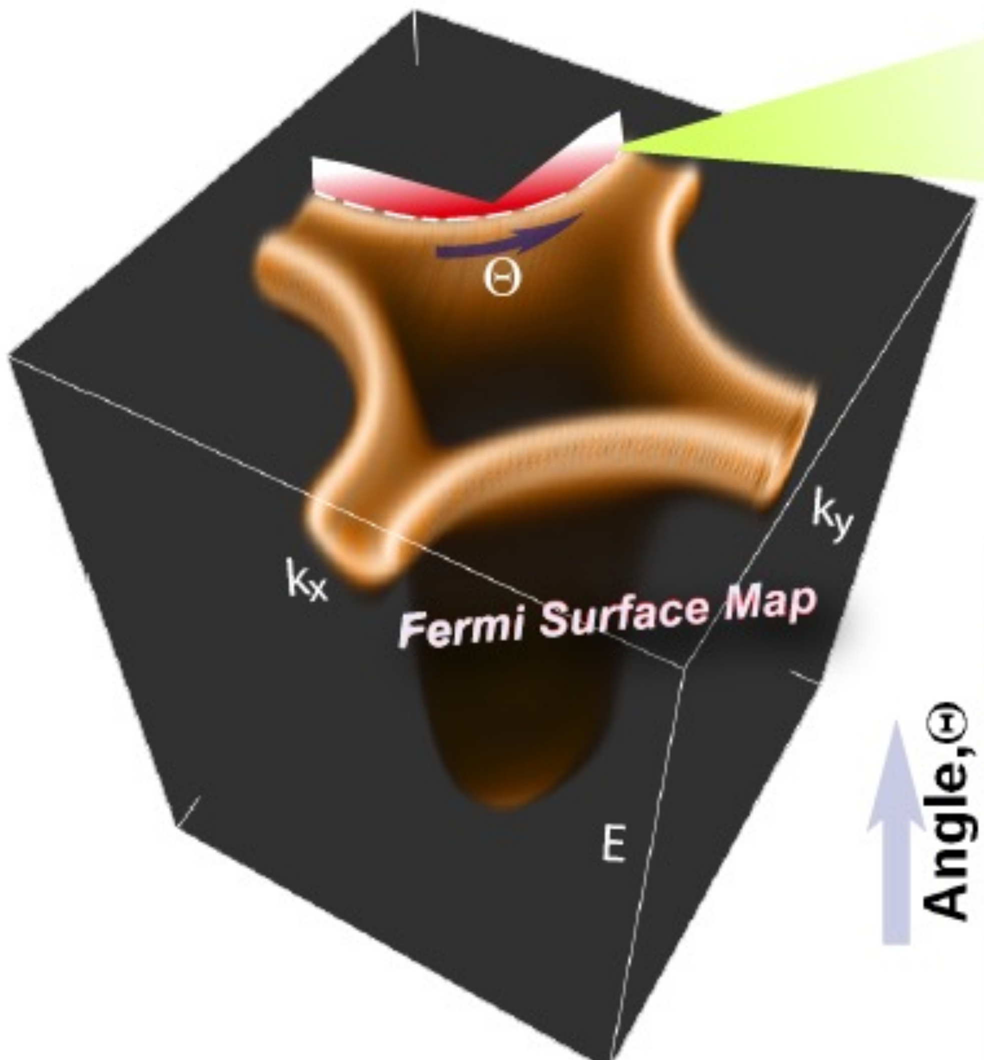
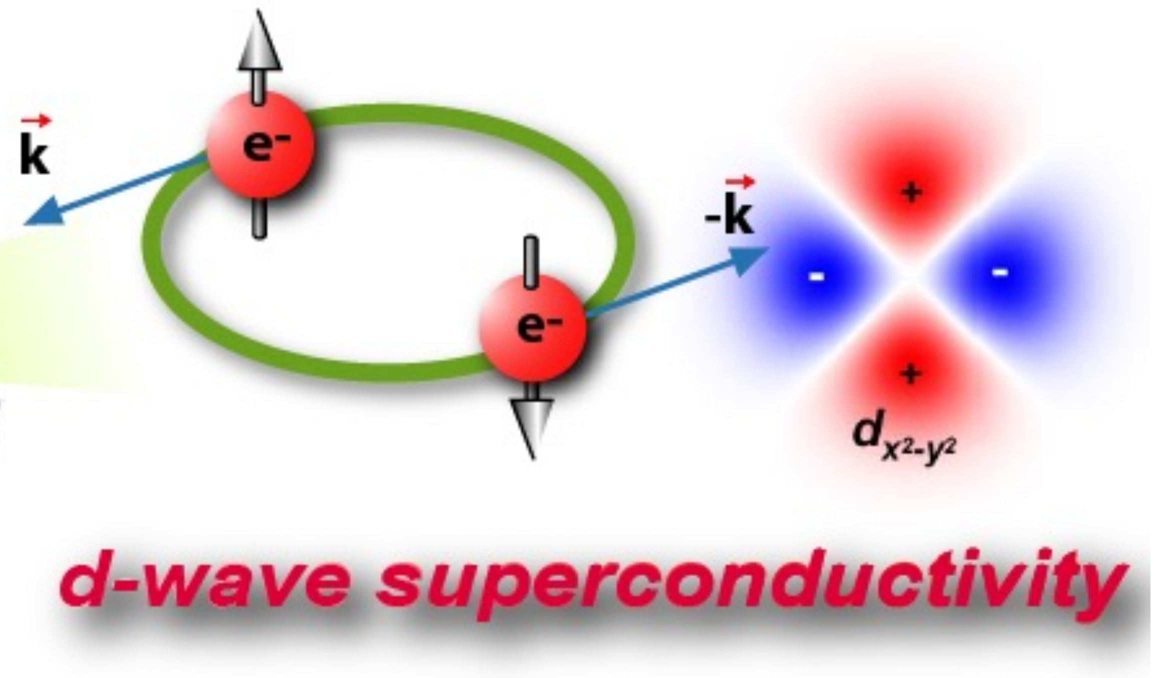
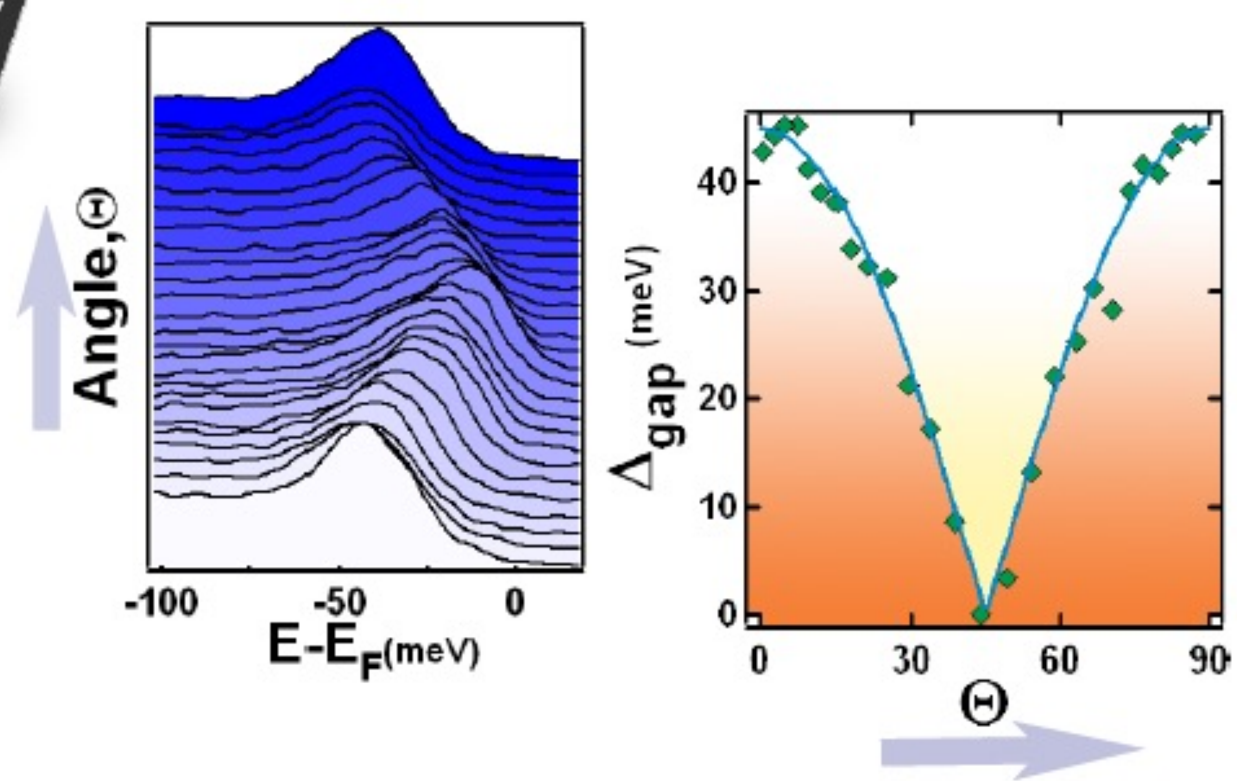


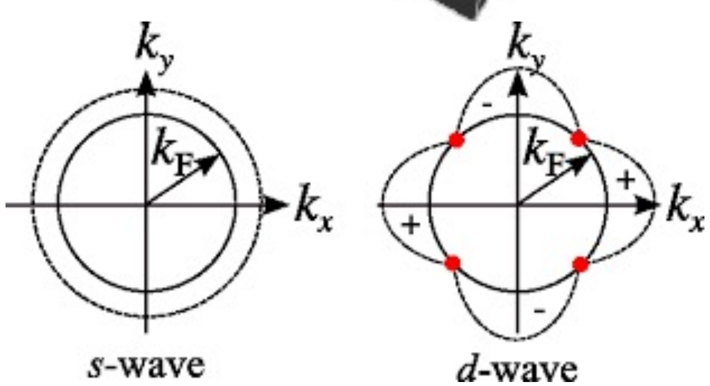
K.M. Shen, *et al.*, PRL 93, 267002

Discovery of "*d*-wave" superconductivity in cuprates









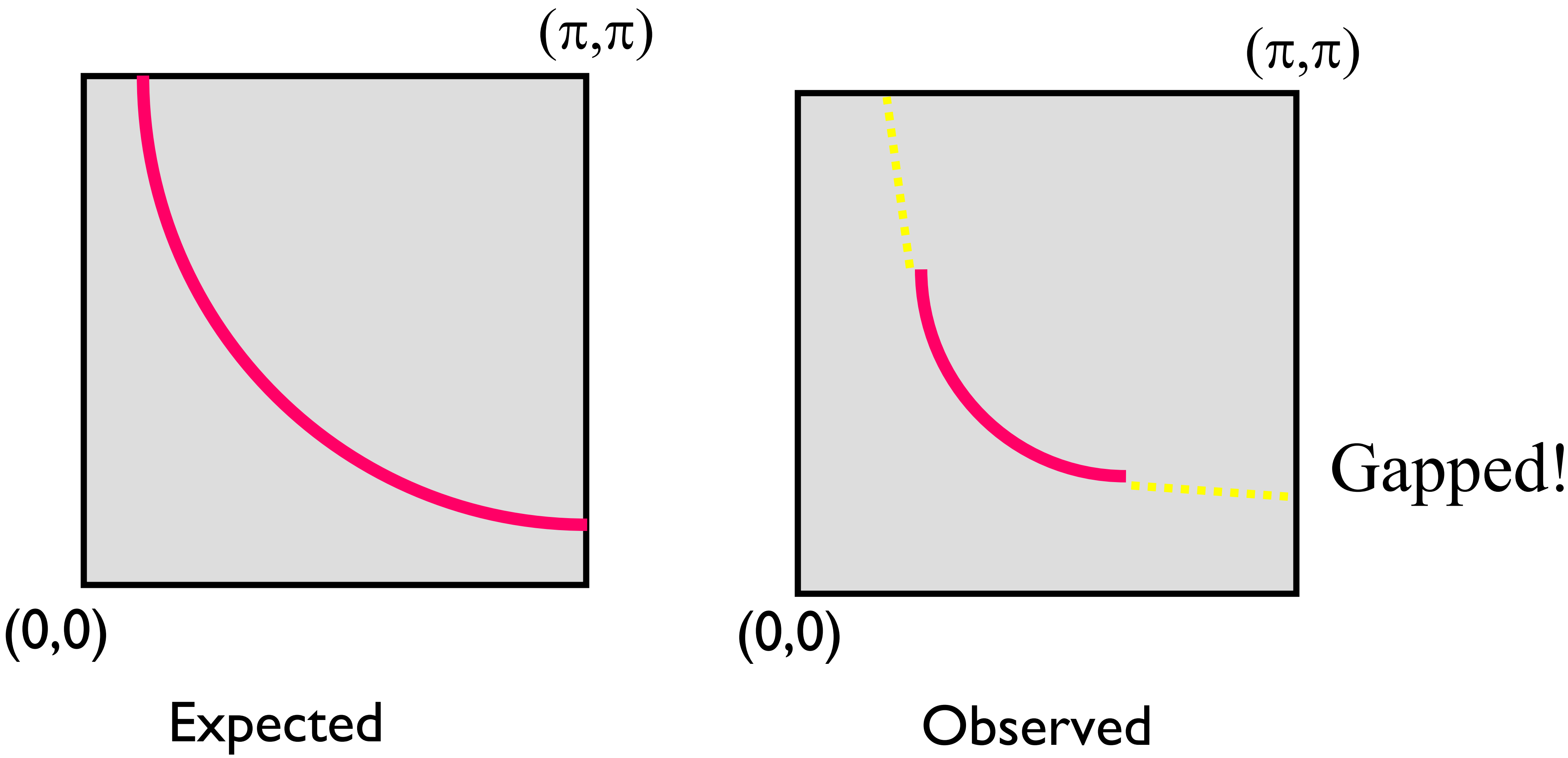
Shen, Dessau, Wells, *et al.*, PRL **70**, 1553 (1993)

P. Bogdanov, Y.L. Chen Ph.D Thesis (2001)

Courtesy of ZX Shen

Discovery of the "pseudogap" in the cuprates

Discontinuous Fermi Surfaces in the Normal State (?!?)



Portion of the Fermi surface gapped, even in the normal state!

D.S. Marshall et al., Phys. Rev. Lett. 76, 4841 (1996)
A.G. Loeser et al. Science **273**, 325 (1996)
H. Ding et al. Nature **382**, 51 (1996)

Courtesy of ZX Shen

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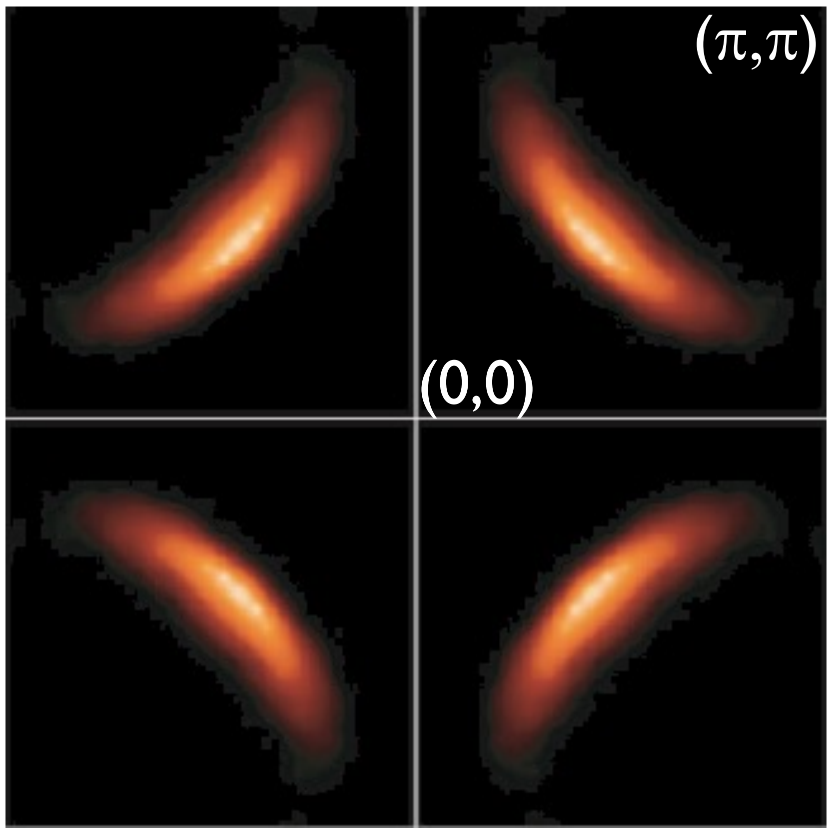
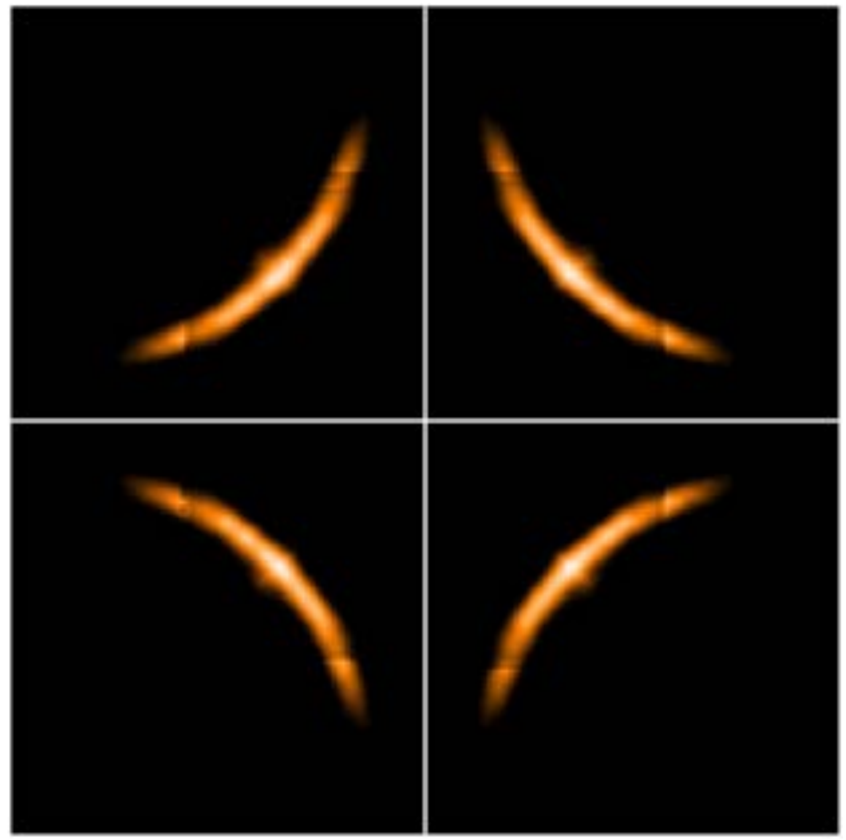


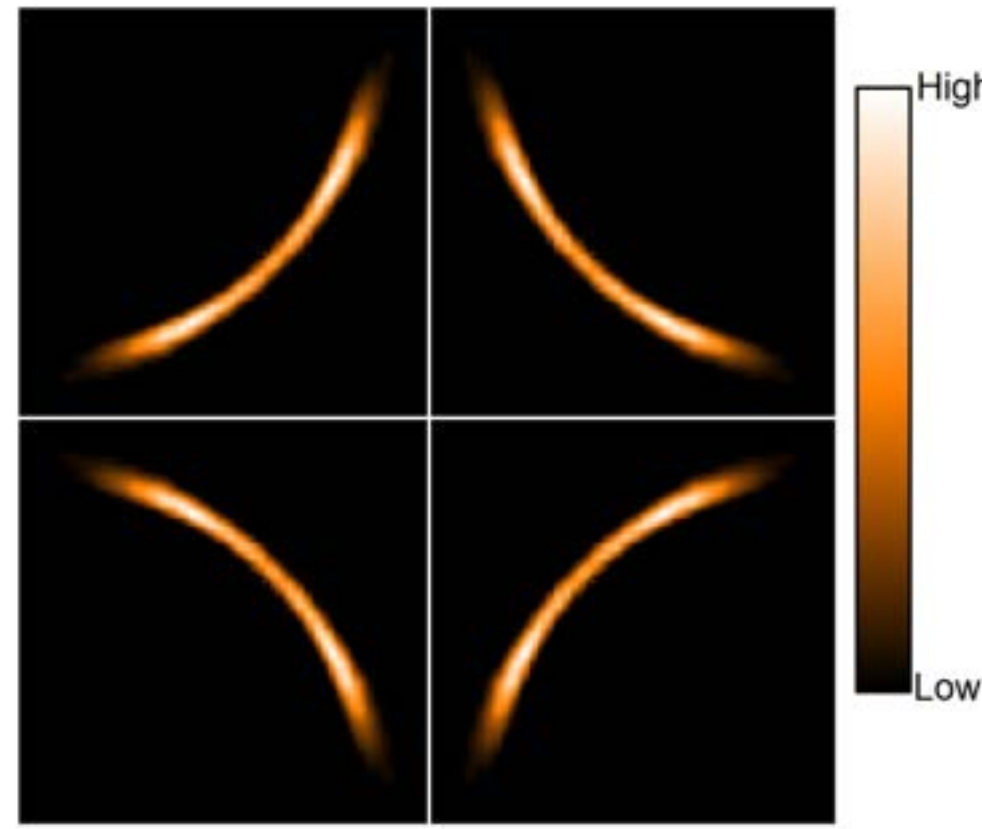
CCOC

K.M. Shen et al.,
Science 307, 901



Bi-2212

W. S. Lee *et al.*
Nature 450, 81



Bi-2201

M. Hashimoto et al.,
Nature Physics 6, 414

Courtesy of ZX Shen

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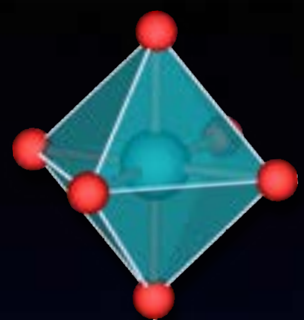
Ruthenate properties are highly tunable with structural changes

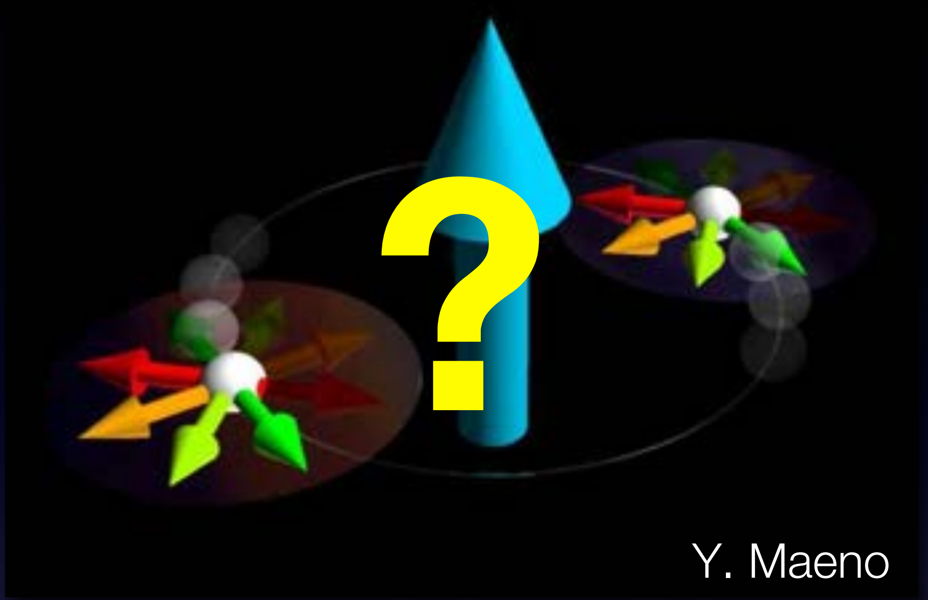


RuO_6 octahedra

Ru^{4+} : $4d^4$

Compound	Dimensionality	Octahedral Connectivity	Properties
Sr_2RuO_4	2D	CORNER	Exotic SC
Ca_2RuO_4	2D	CORNER	AF Mott Insulator
$CaRuO_3$	3D	CORNER	heavy FL
$SrRuO_3$	3D	CORNER	FM Metal
RuO_2	3D	EDGE & CORNER	Metal

ground states can be tuned from metal, AF insulator, FM metal, exotic SC, simply by changing connectivity of RuO_6 octahedra (without doping)

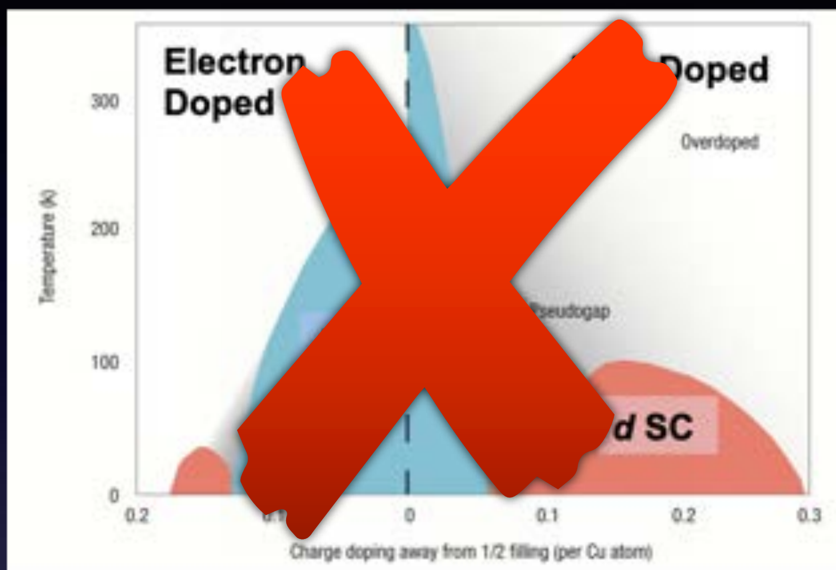


- various experiments (μSR, Kerr rotation) point towards broken time-reversal symmetry
- simple chiral *p*-wave, spin-triplet model called into question by recent experiments
- order parameter is unconventional, but precise nature still up for debate

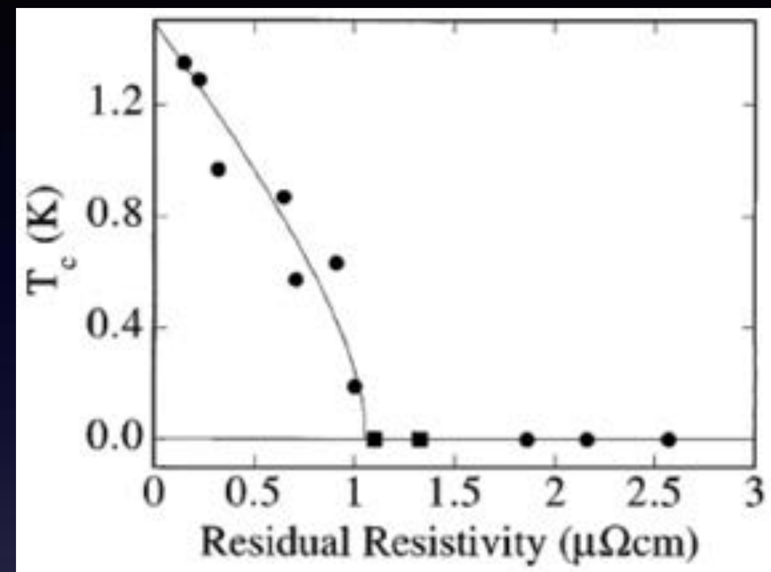
WANTED : clean knobs to control SC in Sr_2RuO_4

traditional approaches like doping and chemical substitution cannot be applied to studying superconductivity of Sr_2RuO_4

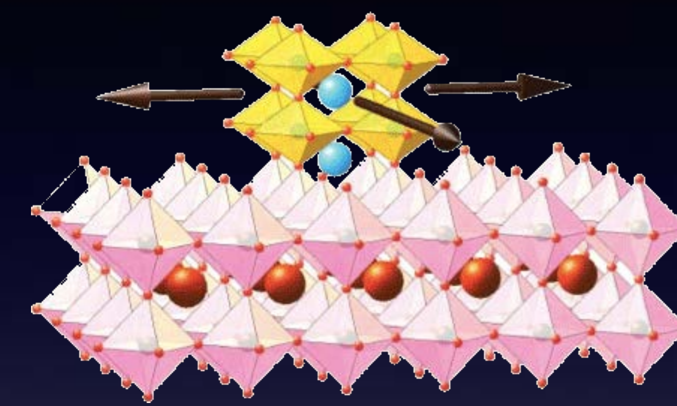
uniaxial or epitaxial (biaxial) strain is a clean alternative



D.A. Bonn, Nature Phys. (2007)



A.P. Mackenzie et. al, *Phys. Rev. Lett.* **80**, 161 (1998)

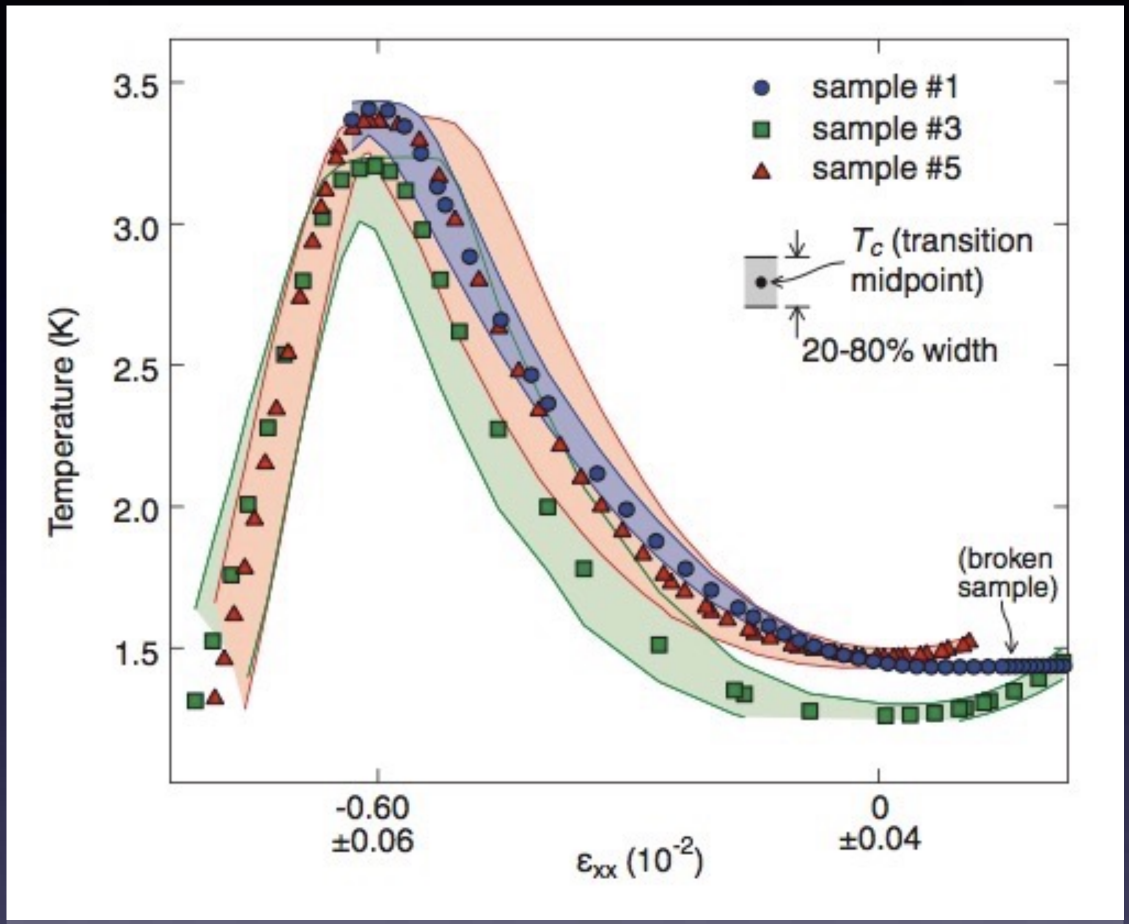


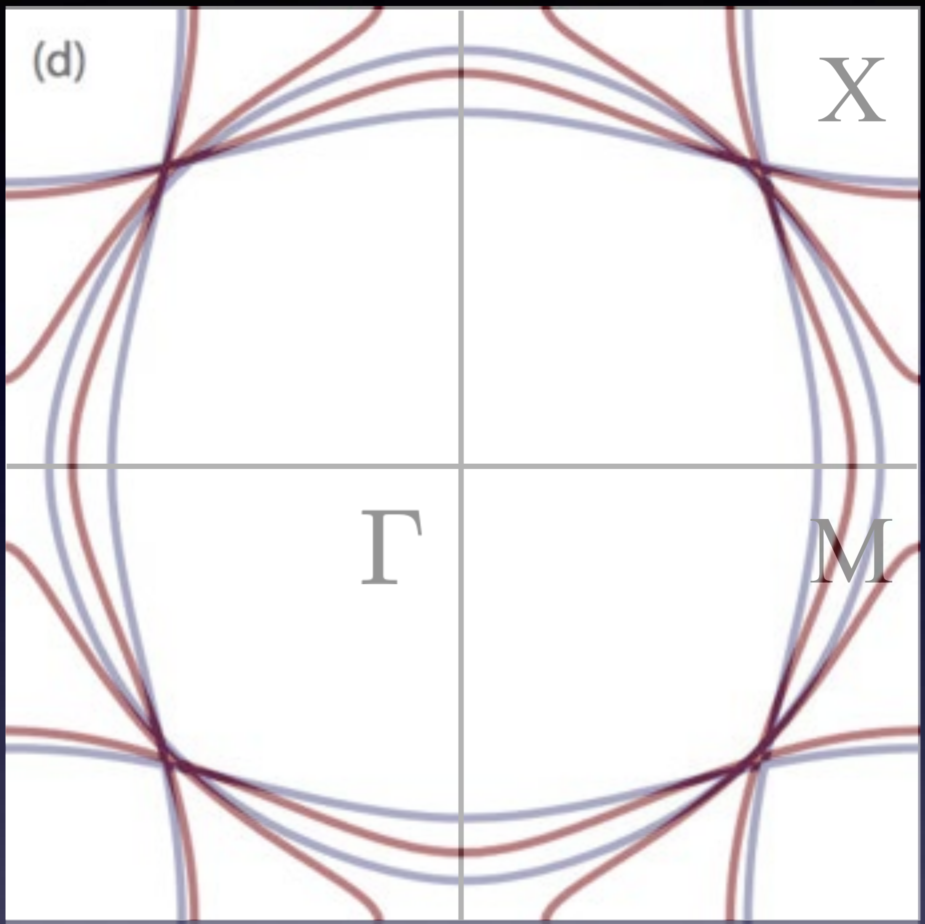
- HTSCs (cuprates, Fe-SC) require doping at the level of 10% to realize SC
- SC is robust at the level of 100,000s of ppm's!
- superconducting coherence lengths ~ 1 nm

- Sr_2RuO_4 is the most disorder-sensitive SC known
- tens to hundreds of ppm's of impurities kills SC
- superconducting coherence lengths ~ 0.1 microns

- strains on the order of a couple percent can be applied
- does not introduce substantial disorder
- can also be implemented in device structures

in-plane uniaxial strain significantly increases T_c in Sr_2RuO_4



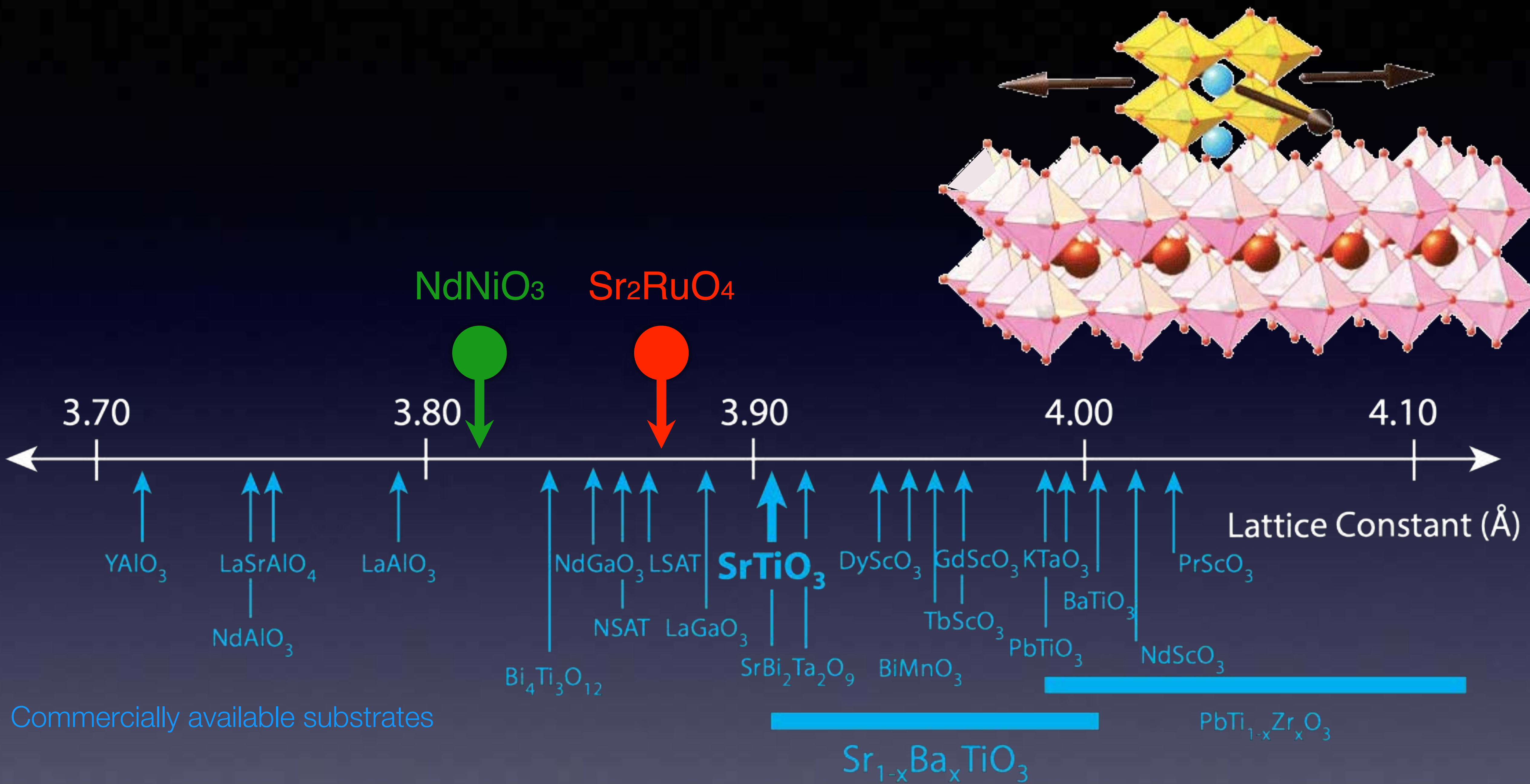


enhancements in T_c may be tied to proximity of van Hove singularity to E_F; proposed that "Lifshitz transition" likely gives rise to the sharp peak in T_c with strain.

How does electronic structure evolve with epitaxial strain?

A. Steppke et al., Science **355**, *eaaf9398*

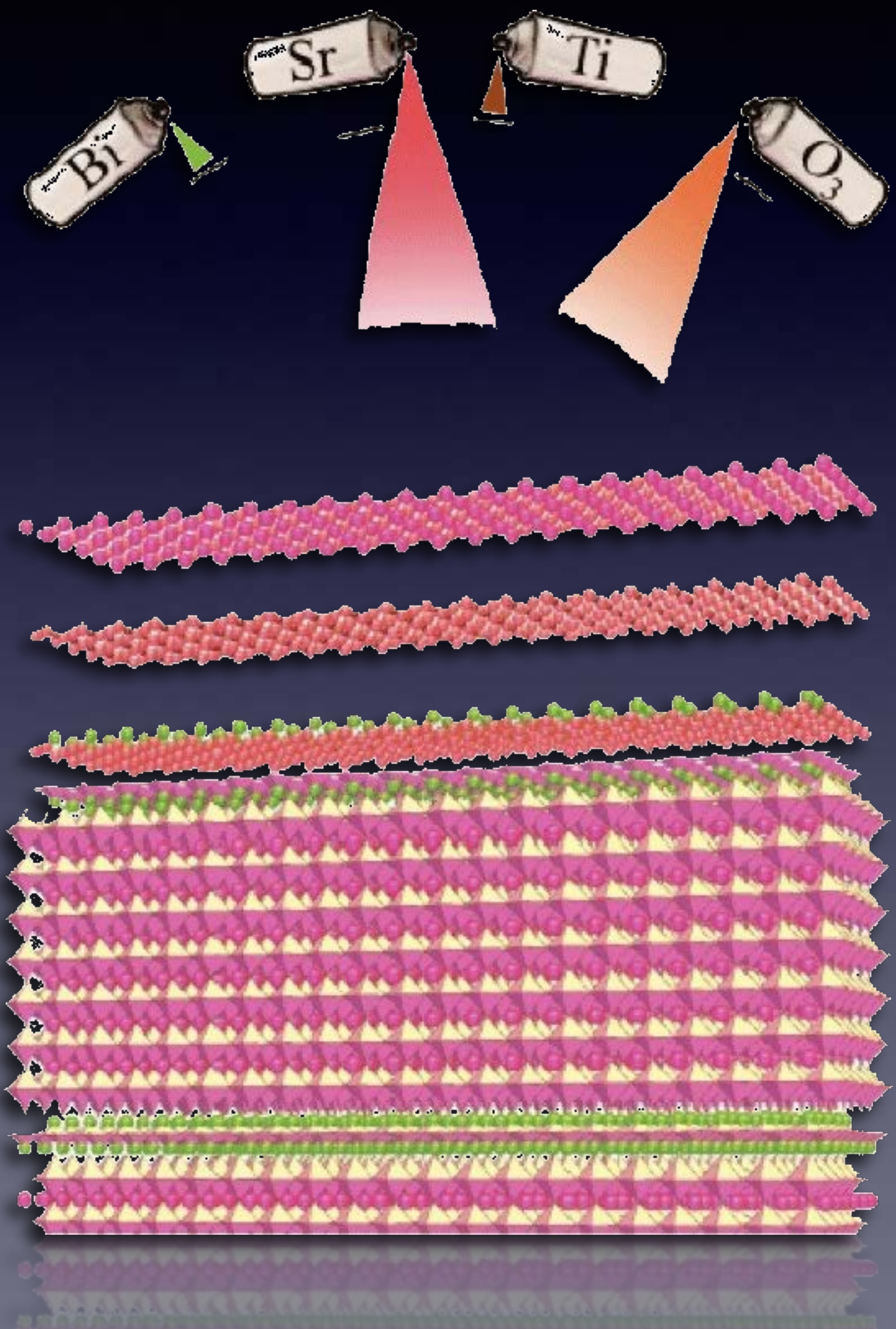
epitaxial strain as a tuning parameter in quantum material heterostructures



- clean tuning parameter (unlike chemical pressure)
- enables most spectroscopies & probes (unlike hydrostatic pressure)
- much larger strains than possible in bulk crystals (and different symmetries), ~3%
- scalable and enables device fabrication (e.g. strained silicon MOSFETs)

molecular beam epitaxy (MBE)

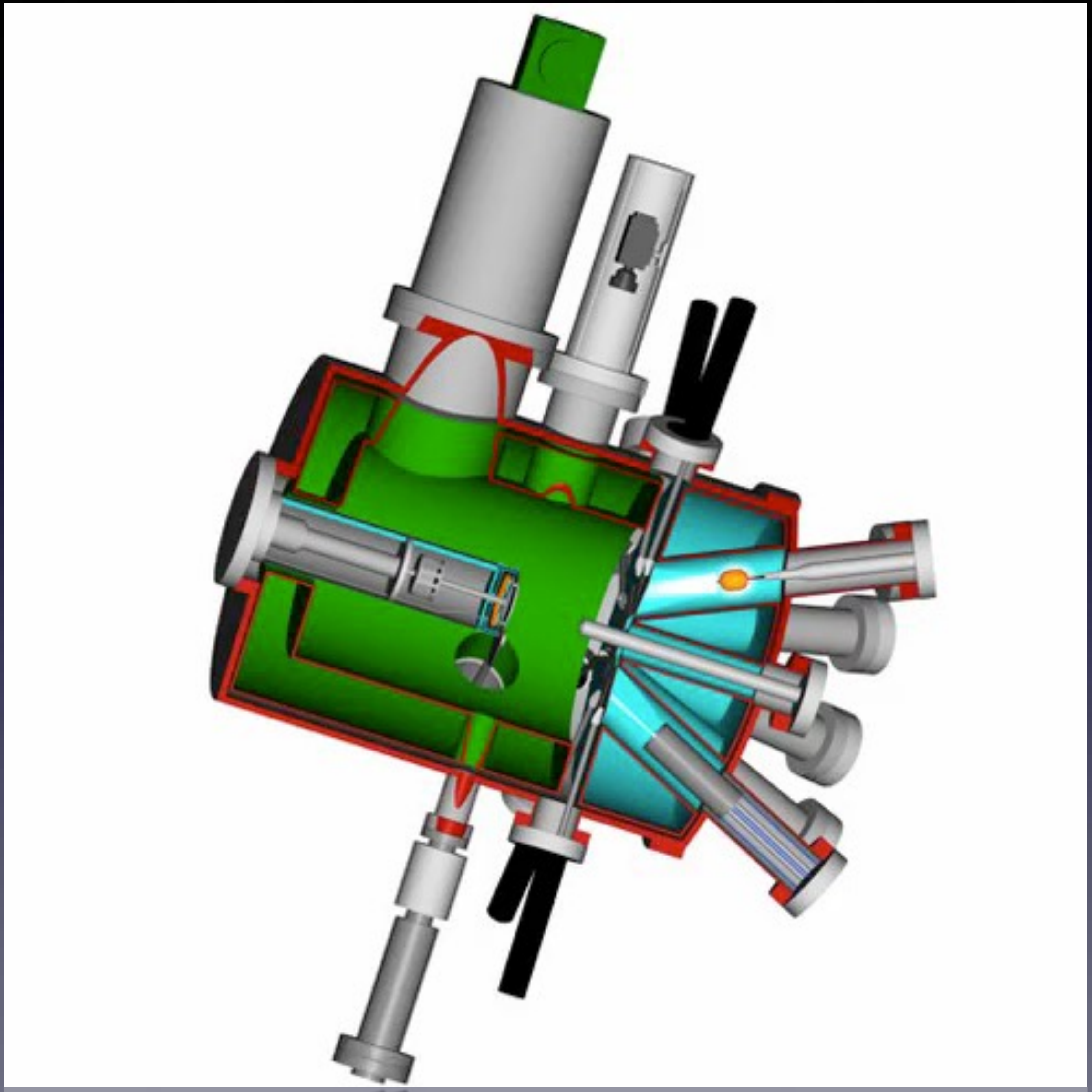
"atomic spray painting"



advantages of MBE

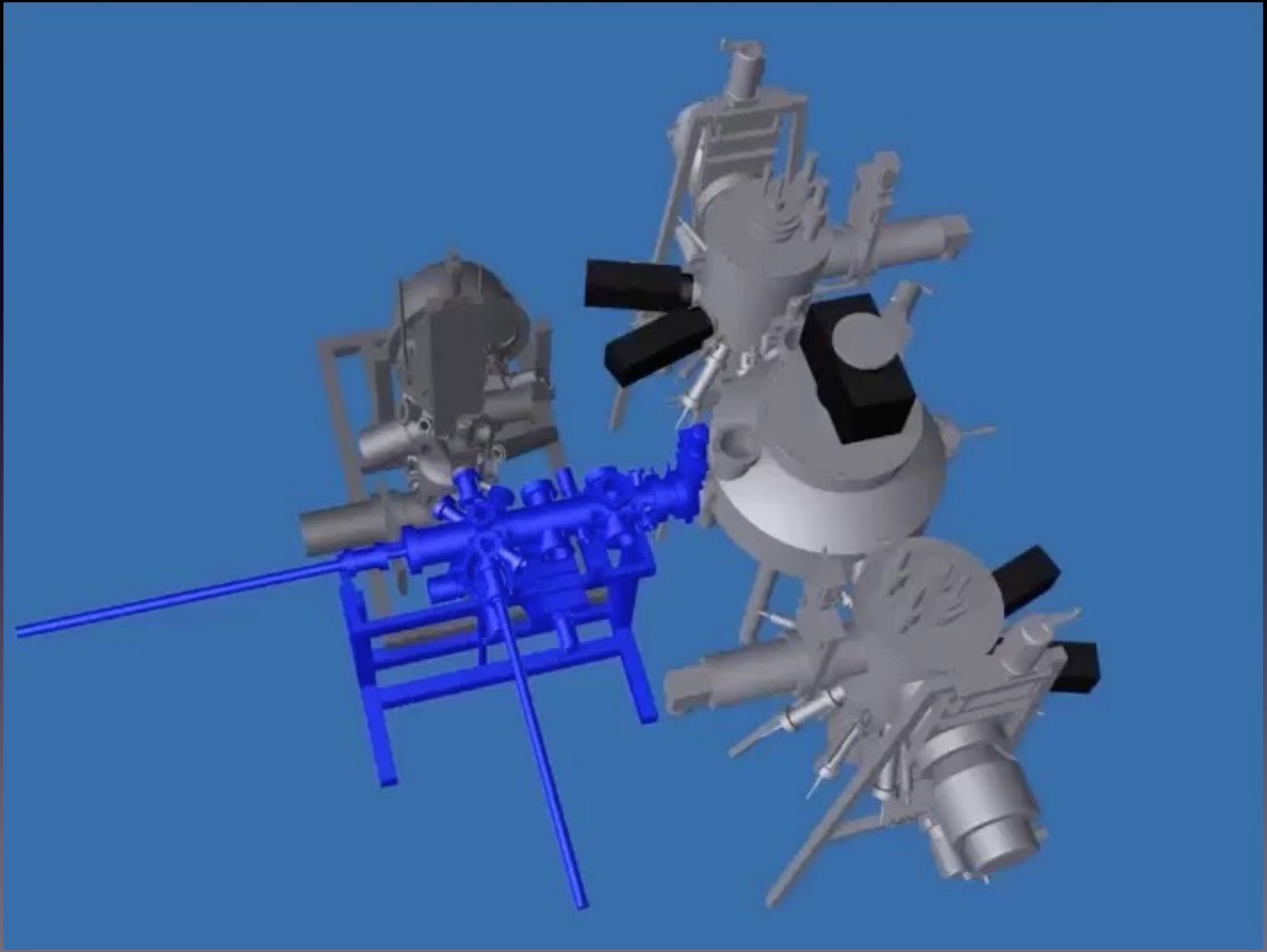
- sub-monolayer control of atomic layers
- can create nearly perfect atomic interfaces, heterostructures, or metastable structures not possible in bulk
- can synthesize materials of extremely high purity
- used for synthesizing laser diodes, LEDs, photovoltaics, etc....

molecular beam epitaxy (MBE)



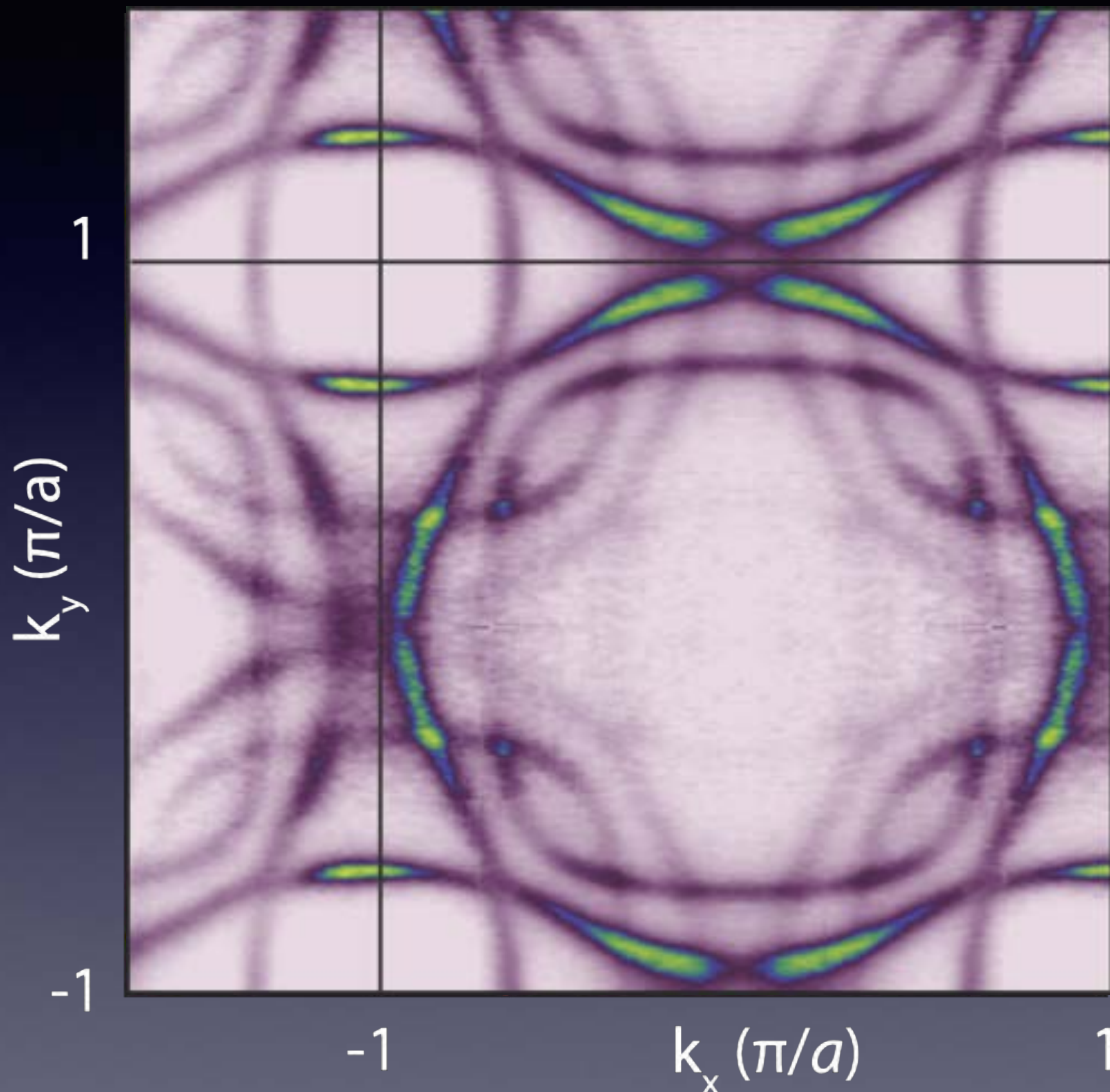
integrated ARPES & MBE system

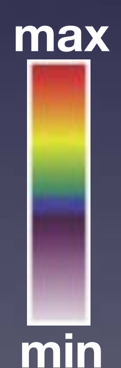

Darrell Schlom



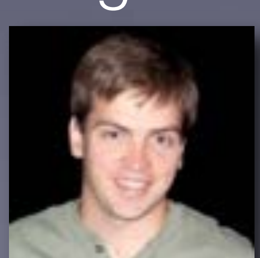
Can tensile strain push the van Hove singularity closer to E_F?

Sr_2RuO_4 single crystal
Sr_2RuO_4 on $SrTiO_3$ (+0.9%)





Bulat Burganov

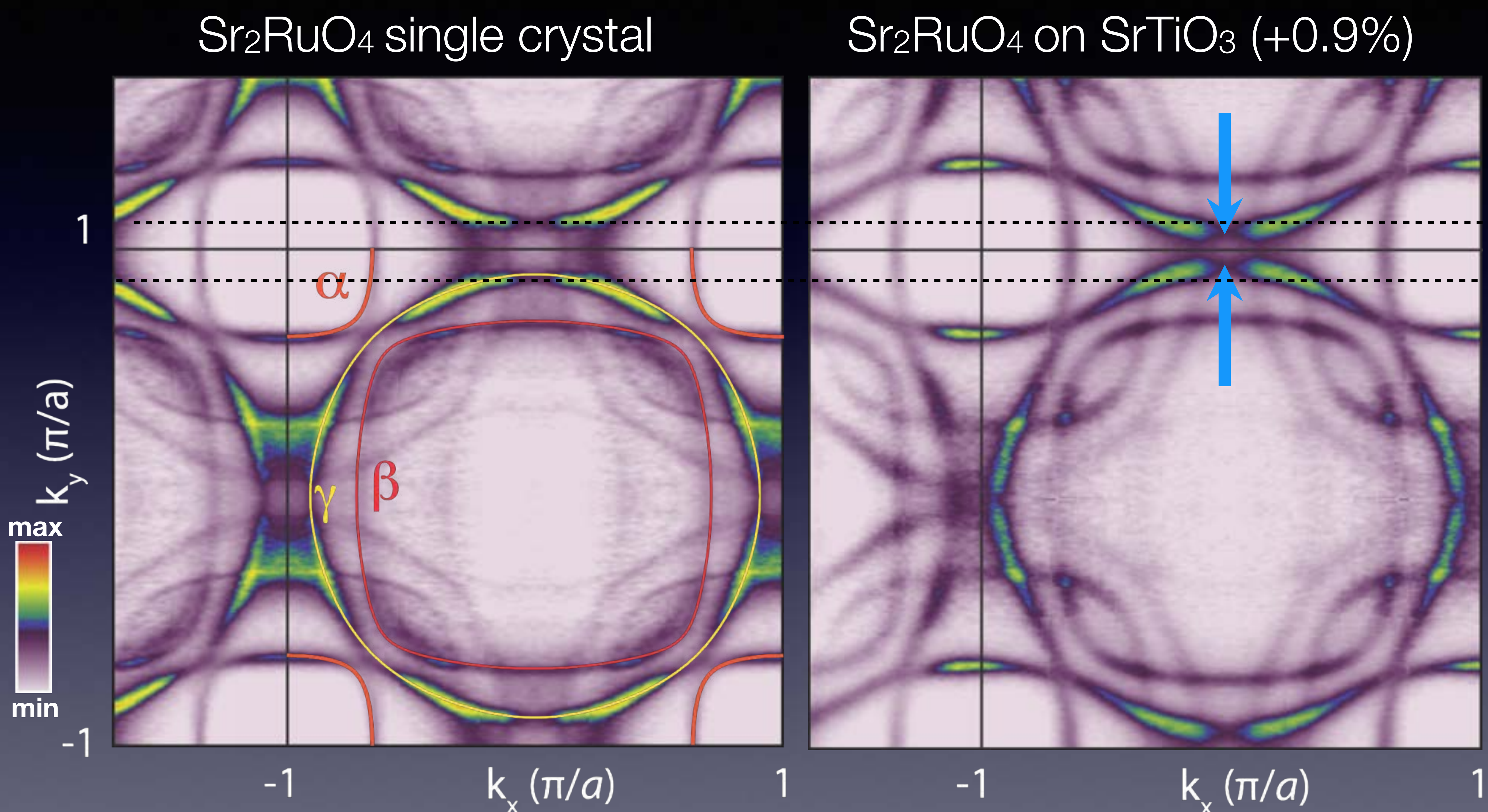


Carolina Adamo

B. Burganov, et al., *Phys. Rev. Lett.* **116**, 197003

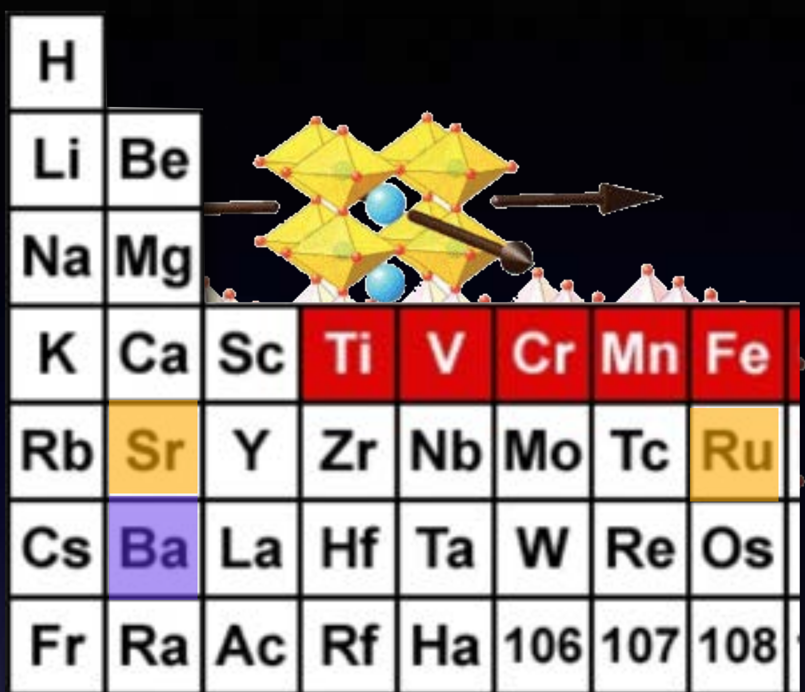
single crystal from A.P. Mackenzie

Can tensile strain push the van Hove singularity closer to E_F?

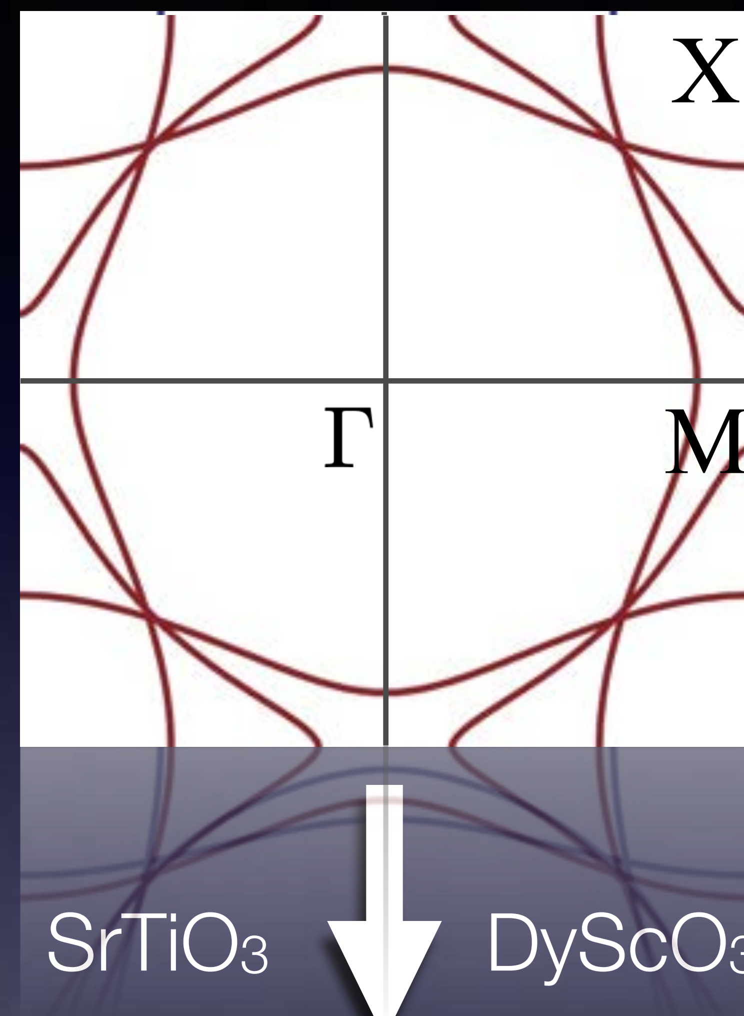


single crystal from A.P. Mackenzie

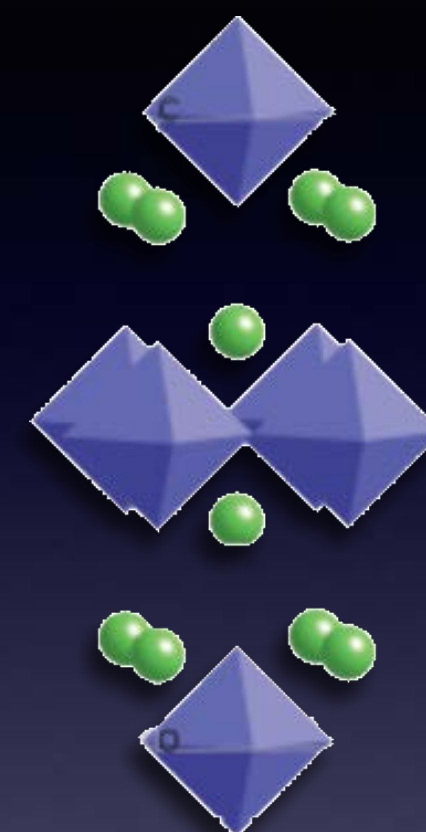
Epitaxial strain to enhance superconductivity in Sr_2RuO_4?

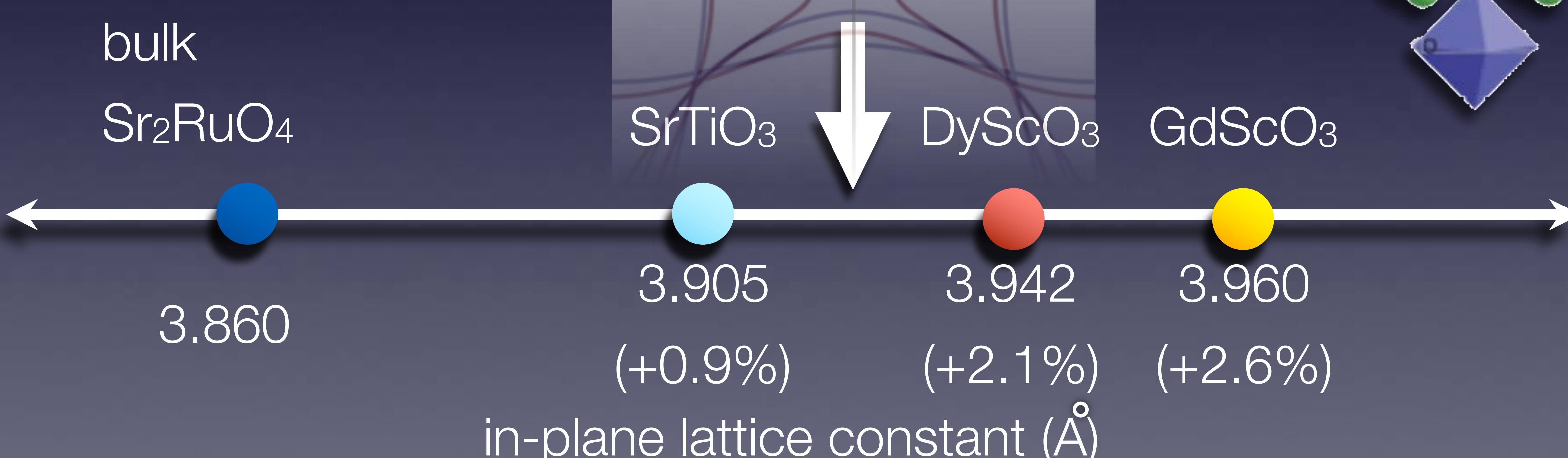


Lifshitz transition



Ba_2RuO_4



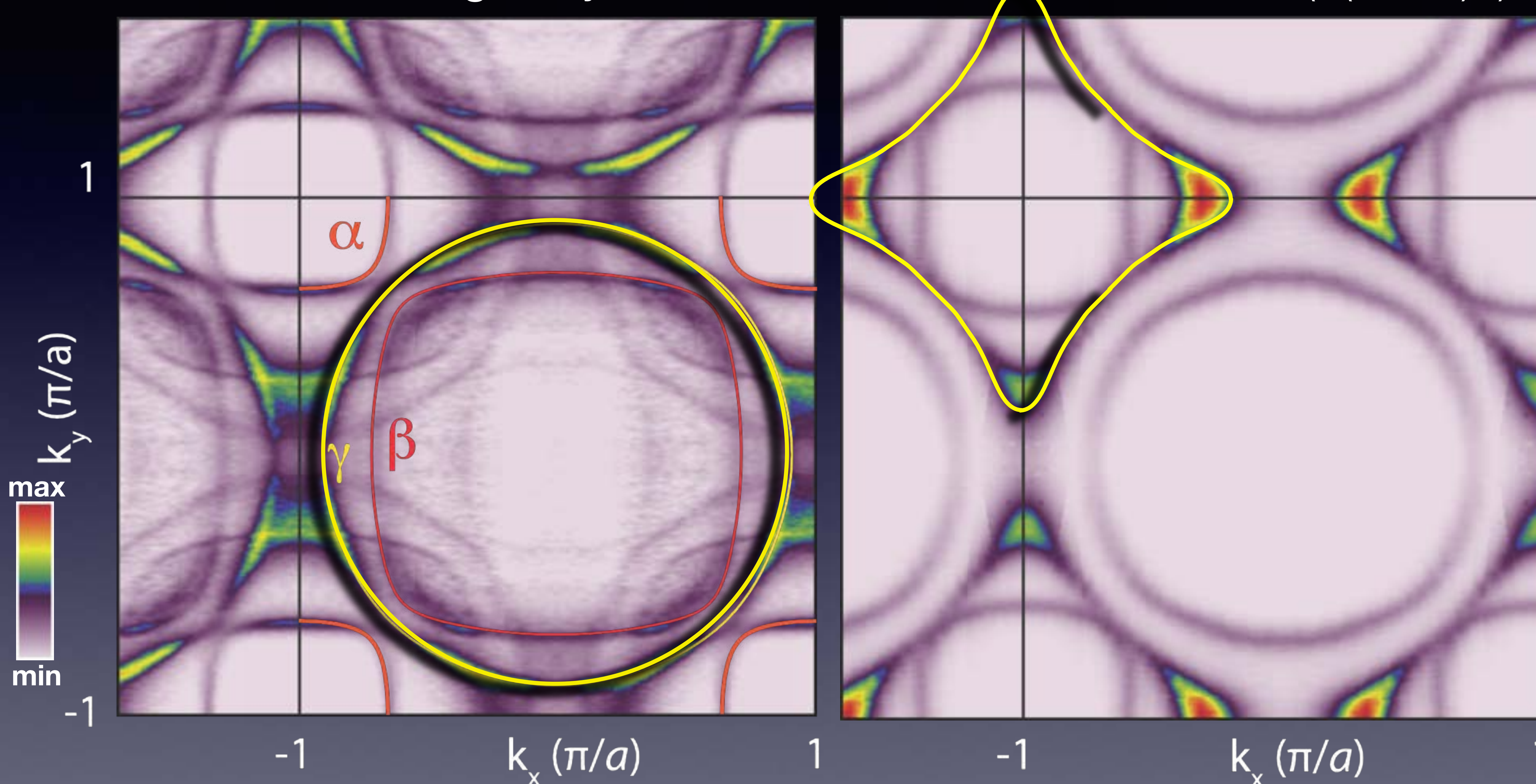


- Ba_2RuO_4 is metastable in bulk but can be epitaxially stabilized

Can tensile strain push the van Hove singularity closer to E_F?

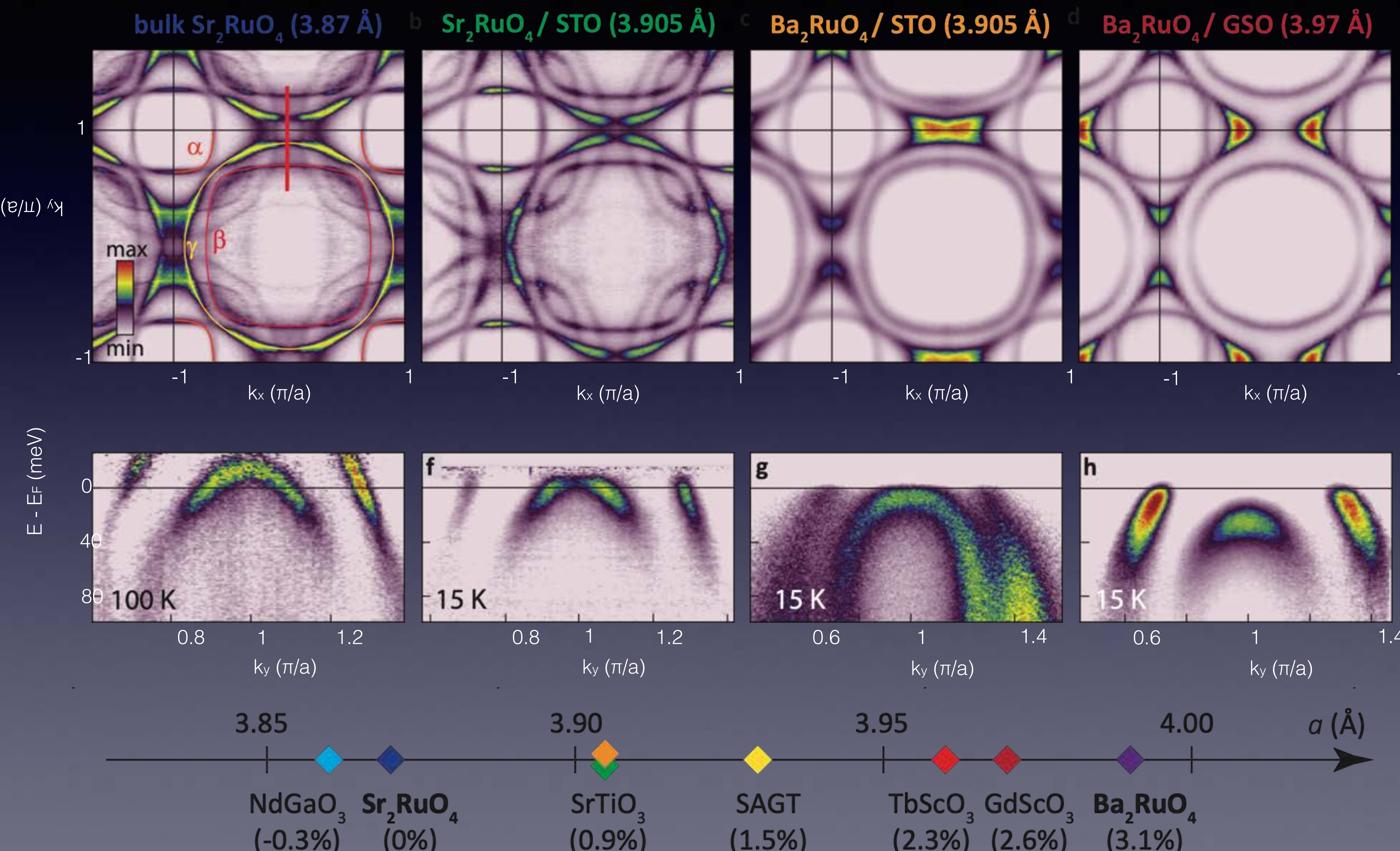
Sr_2RuO_4 single crystal

Ba_2RuO_4 on $SrTiO_3$ (+2.2%)



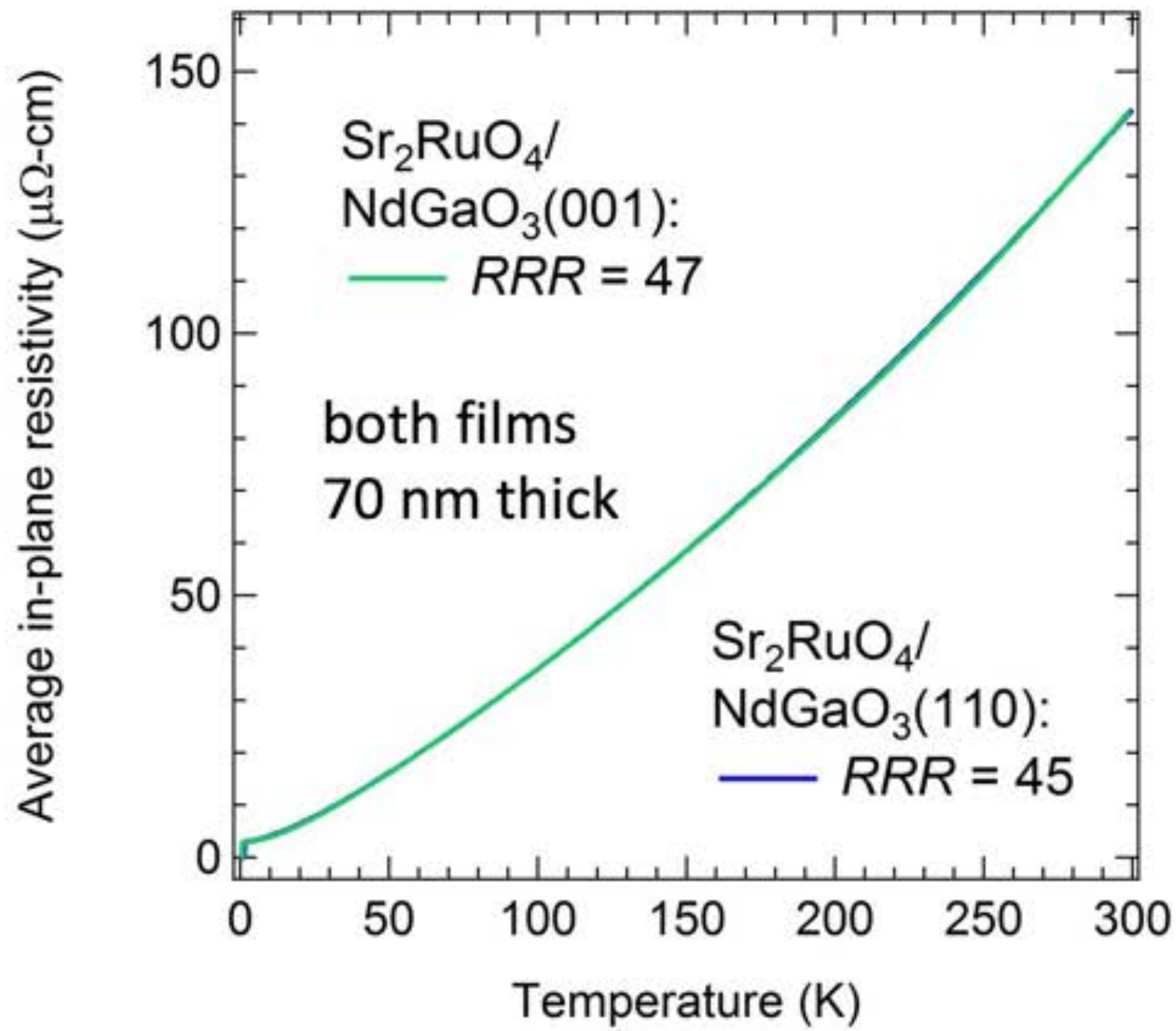
low T Hall coefficient changes sign from negative (Sr_2RuO_4) to positive (Ba_2RuO_4), consistent with ARPES

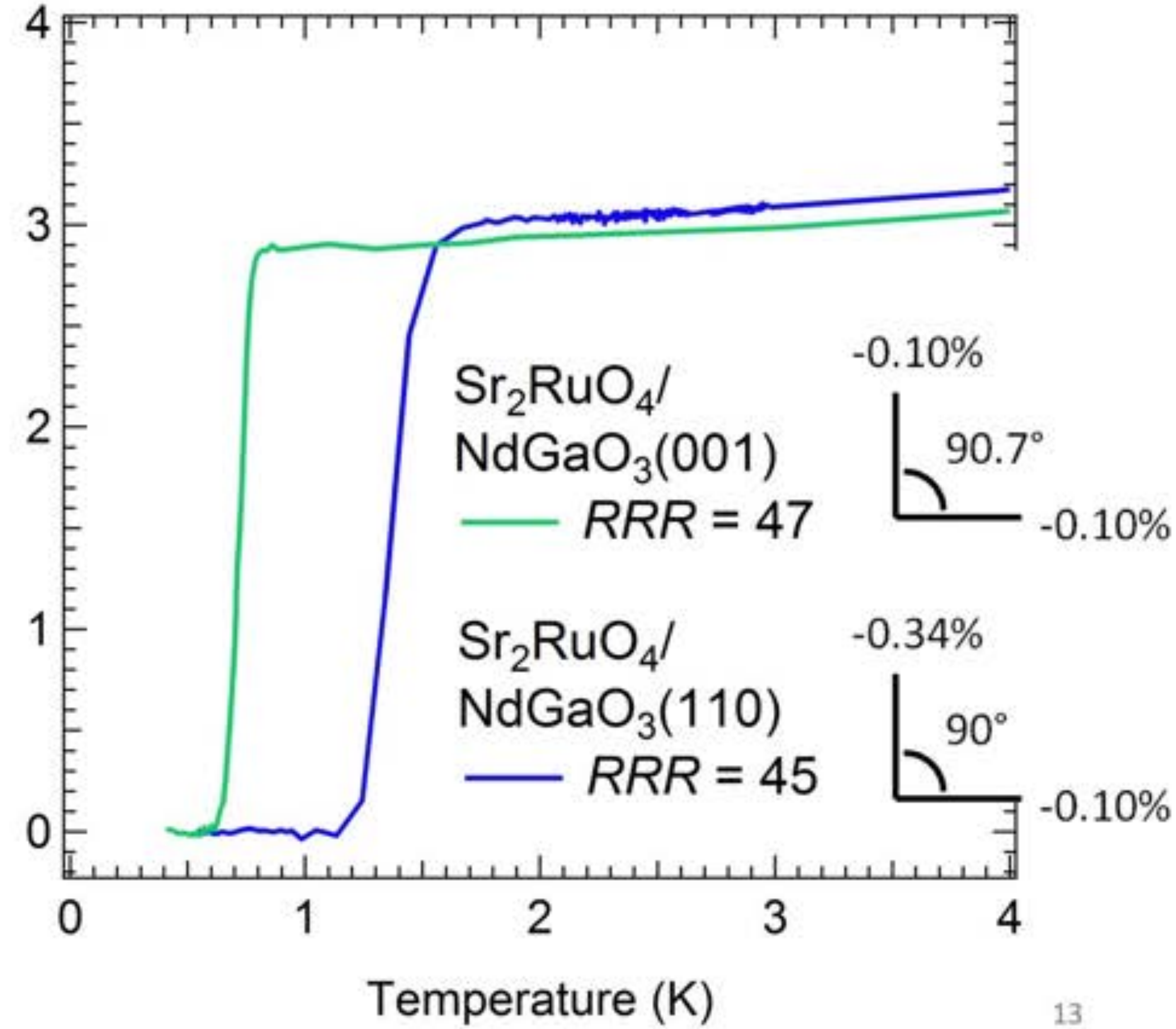
summary of Fermi surface & van Hove singularity evolution with strain



B. Burganov, C. Adamo, et al., *Phys. Rev. Lett.* **116** (2016)

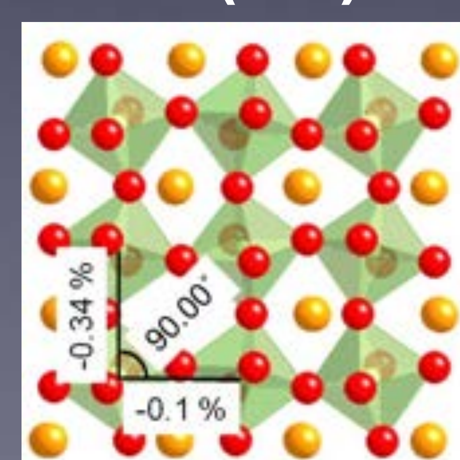
superconductivity depends on orientation of $NdGaO_3$ substrate



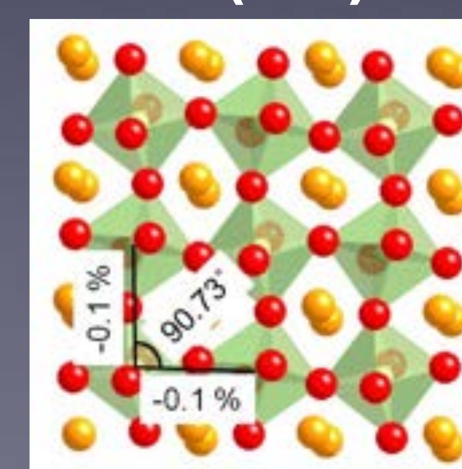


13

$NdGaO_3$ (110) Pbnm



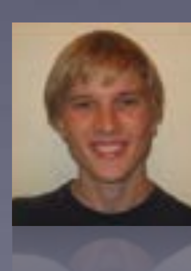
$NdGaO_3$ (001) Pbnm



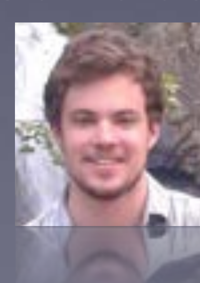
Darrell Schlom

Hari Nair

Jacob Ruf

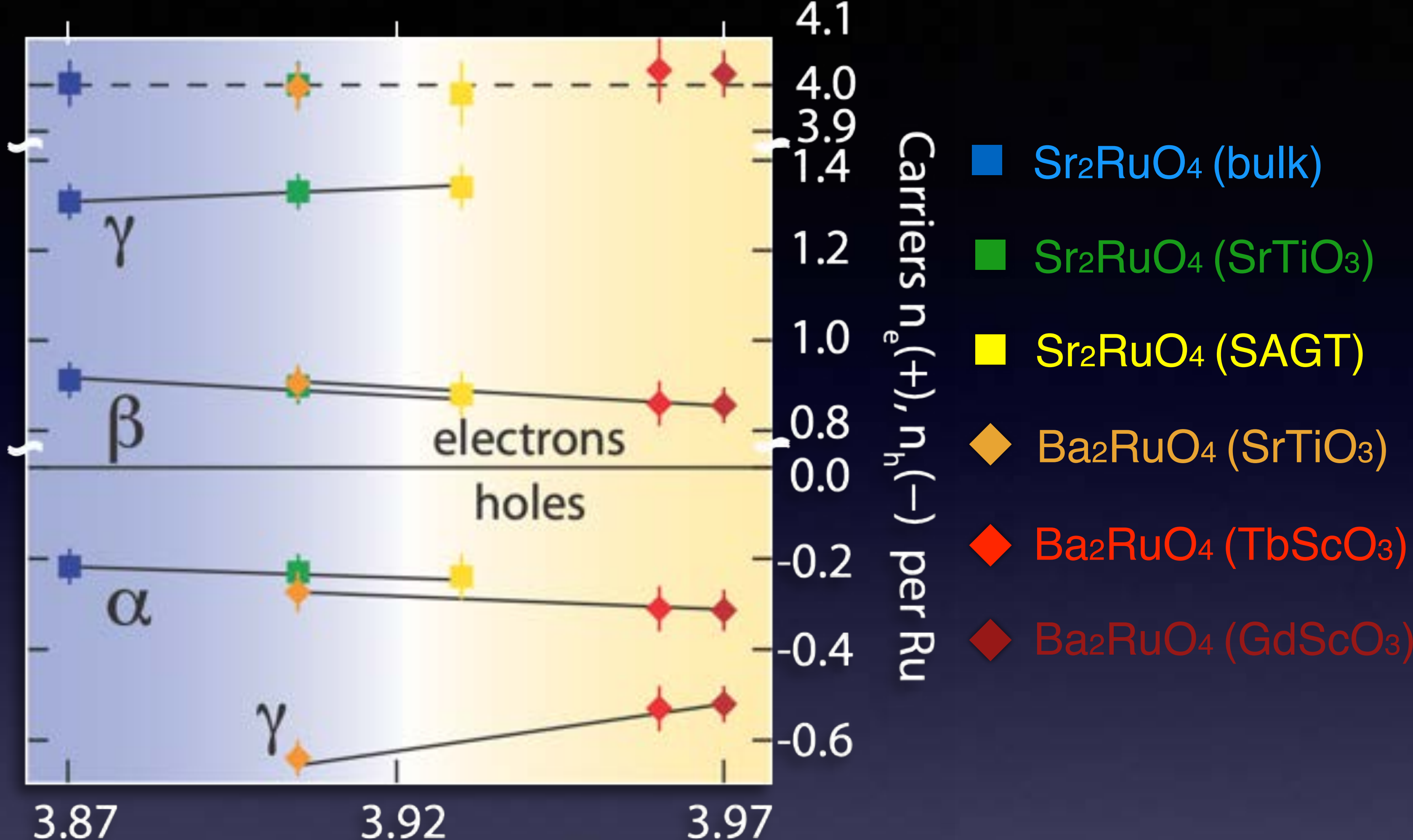


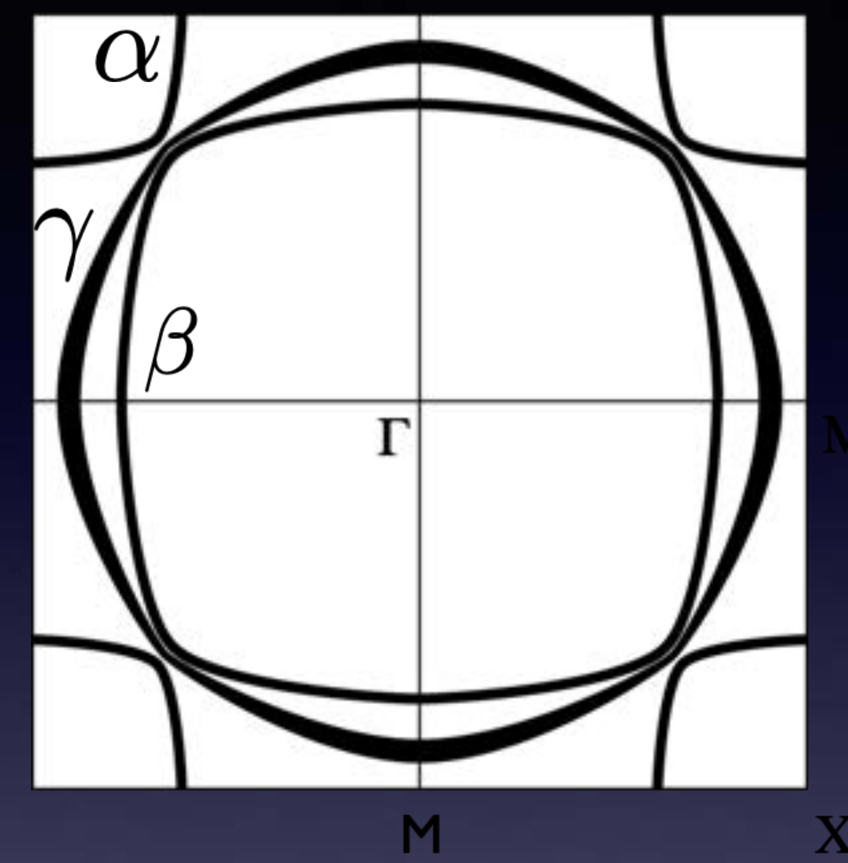
Nate Schreiber



H.P. Nair, J.P. Ruf *et al.*, APL Matr. **6**, 101108

Detailed Luttinger count shows interorbital electron transfer





detailed Luttinger count shows that total number of electrons per Ru remains 4.00 +/- 0.05; electrons are transferred from the 1D d_{yz} & d_{xz} bands to the d_{xy} band