2022 PARADIM Summer School on MBE & ARPES

Frontiers in ARPES

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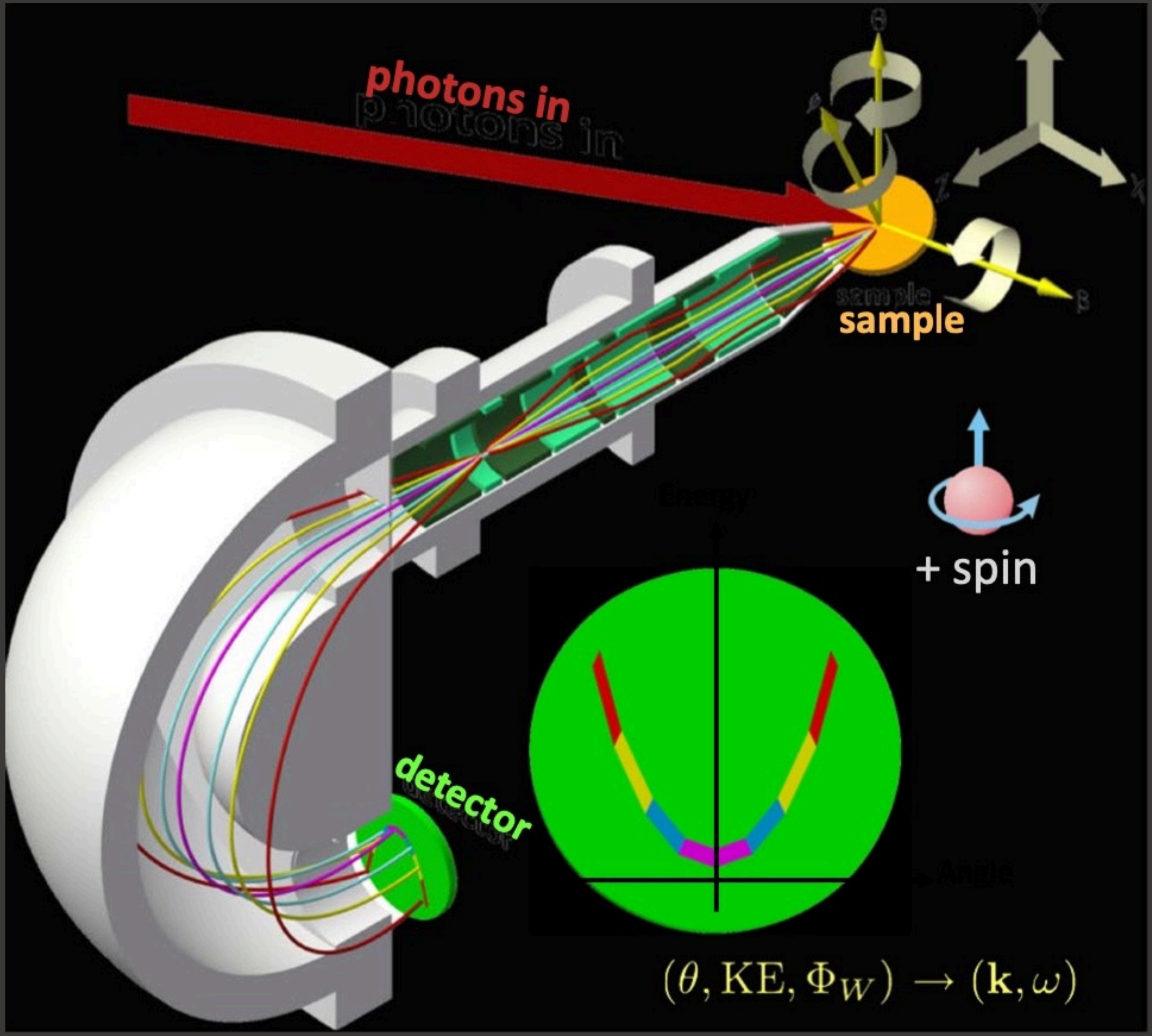
Cornell University
June 17, 2022

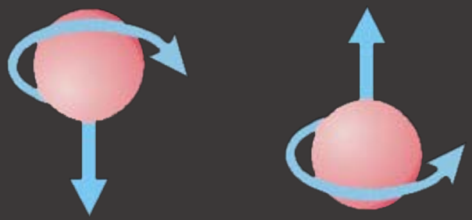
Acknowledgements : Luca Moreschini

ARPES + something else!

1. Spin detection
2. Time-resolution
3. Spatial resolution

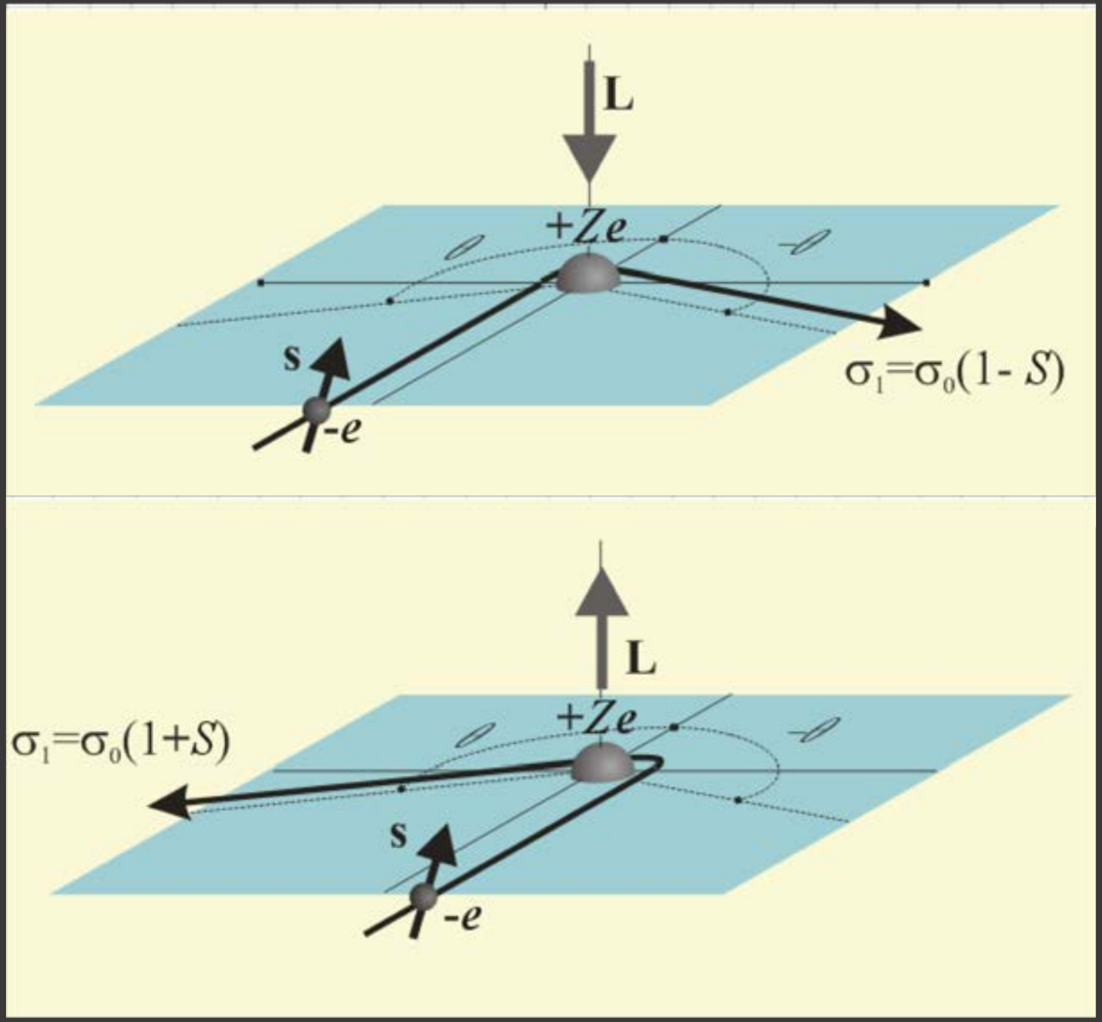


Spin polarimetry



Mott scattering

+ stability, - efficiency

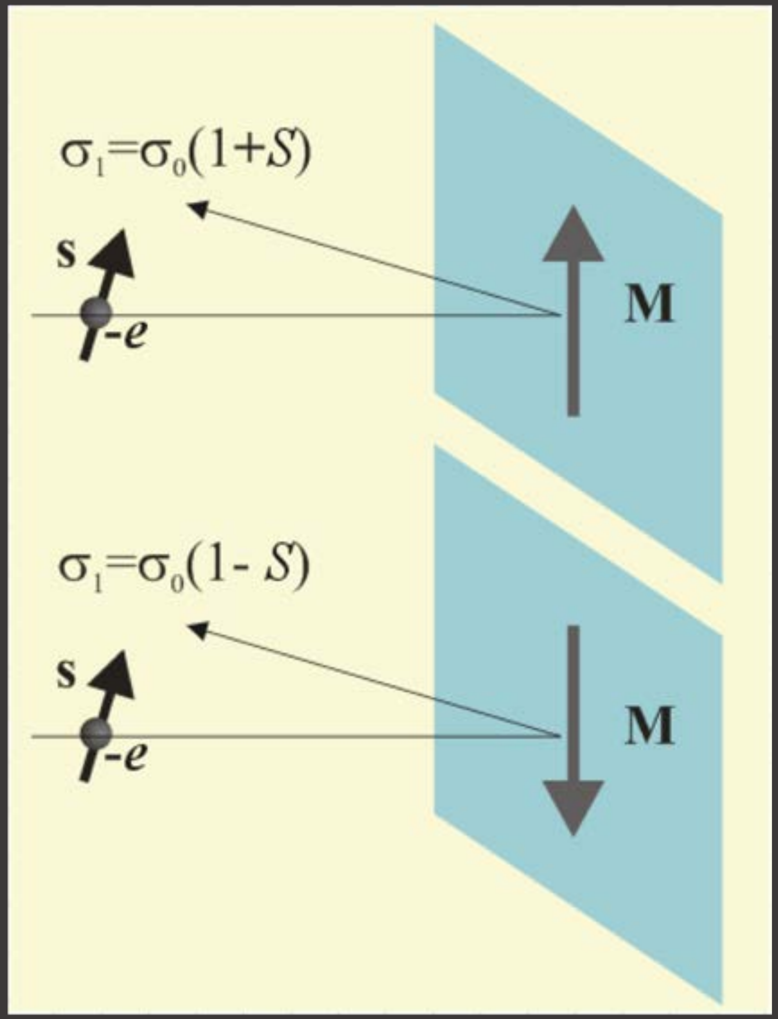


coupling between the atomic orbital momentum and the spin of the electron

different cross section
for two different scattering directions

exchange scattering (VLEED)

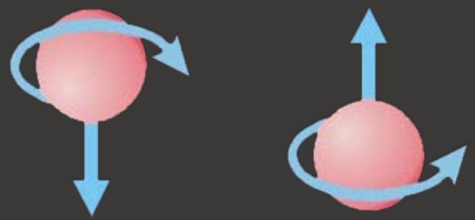
- stability, + efficiency



coupling between the ferromagnet magnetic moment and the spin of the electron

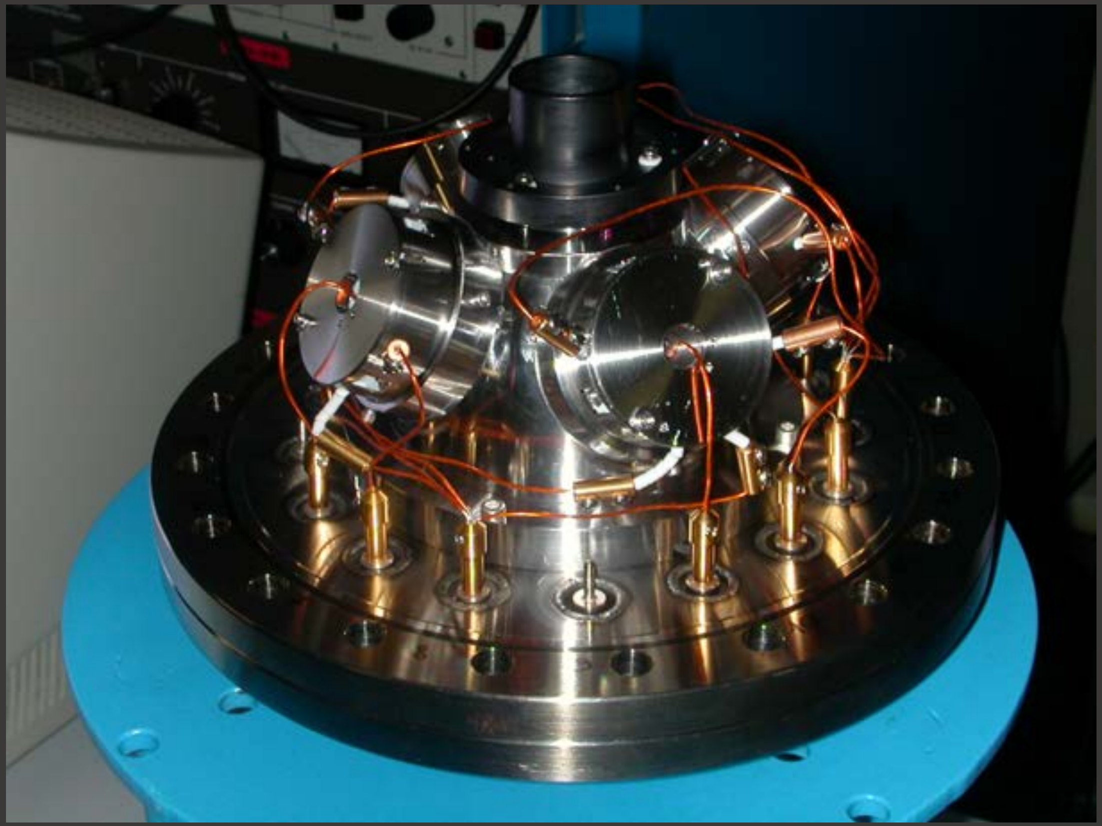
different reflectivity
for two opposite magnetization directions

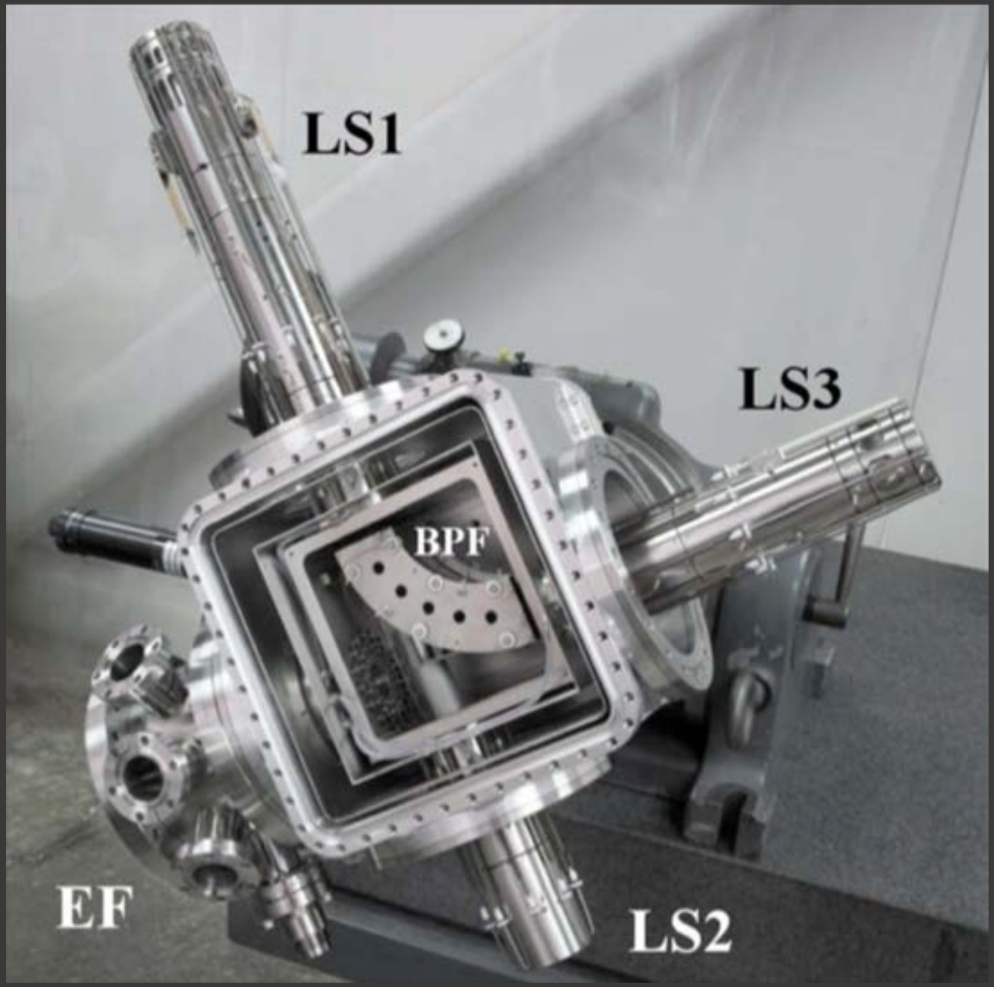
Spin polarimetry



Mott scattering
+ stability, - efficiency

exchange scattering (VLEED)
- stability, + efficiency





different cross section for two different scattering directions

L. Moreschini *et al.*, *Rev. Sci. Instrum.* ***79***, *033905 (2008)*

different reflectivity for two opposite magnetization directions

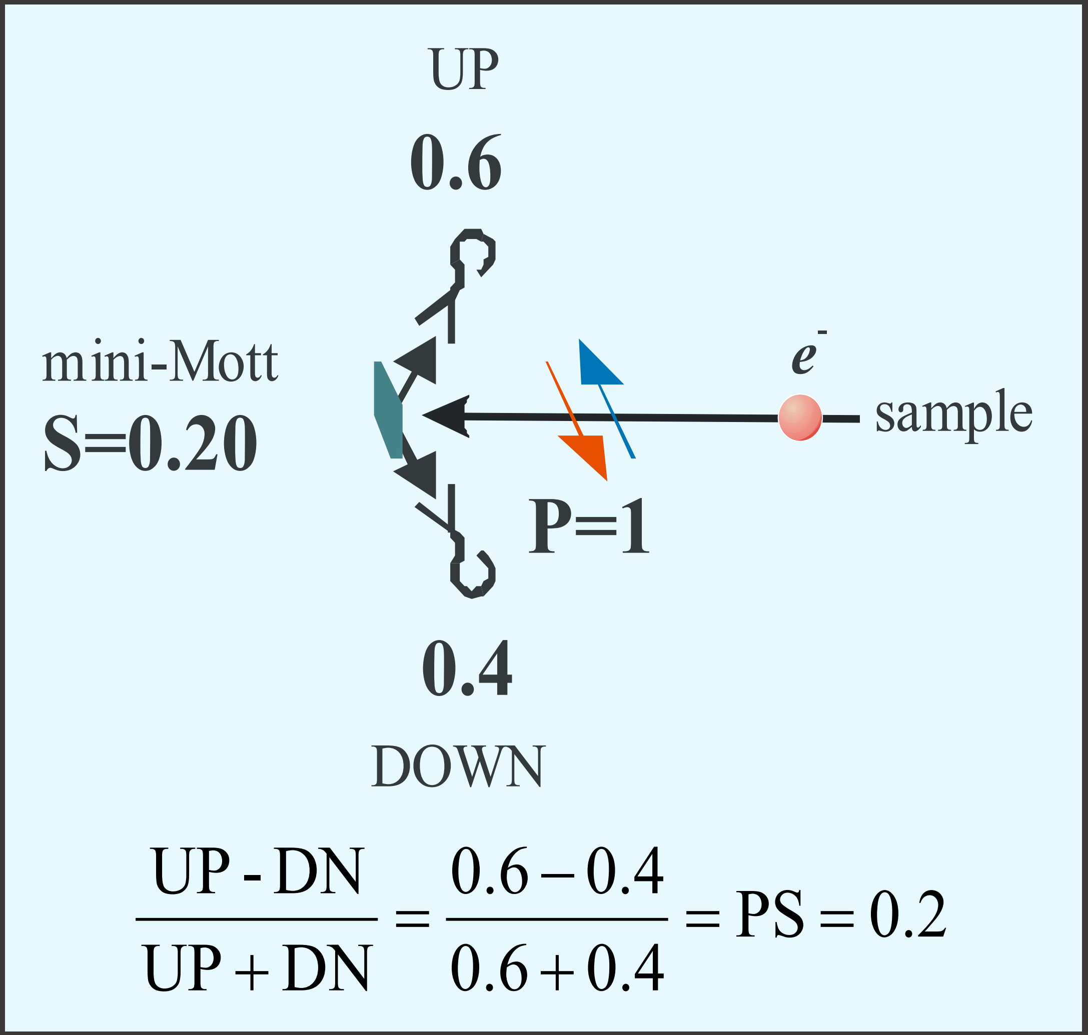
C. Jozwiak *et al.*, *Rev. Sci. Instrum.* ***81***, *053904 (2010)*

Spin polarimetry

measured asymmetry: $A = \frac{I_1 - I_2}{I_1 + I_2} = P * S$ S: Sherman function

error-bar on polarization: $\Delta P \propto \frac{1}{\sqrt{N_0 \eta}}$ $\eta = S^2 \frac{N}{N_0}$ **figure of merit**

Why getting good quality spin-resolved spectra is difficult



$$\frac{\mathrm{UP} - \mathrm{DN}}{\mathrm{UP} + \mathrm{DN}} = \frac{0.6 - 0.4}{0.6 + 0.4} = \mathrm{PS} = 0.2$$

The efficiency of the polarimeters is low

Mott	exchange
$\frac{N}{N_0} \approx 10^{-3}$	$\frac{N}{N_0} \approx 10^{-1} - 10^{-2}$

The Sherman function is small

Mott	exchange
$S < 0.2$	$S < 0.4$

J.Kessler, *Polarized electrons* (1976)

Electron analyzers for spin polarimetry

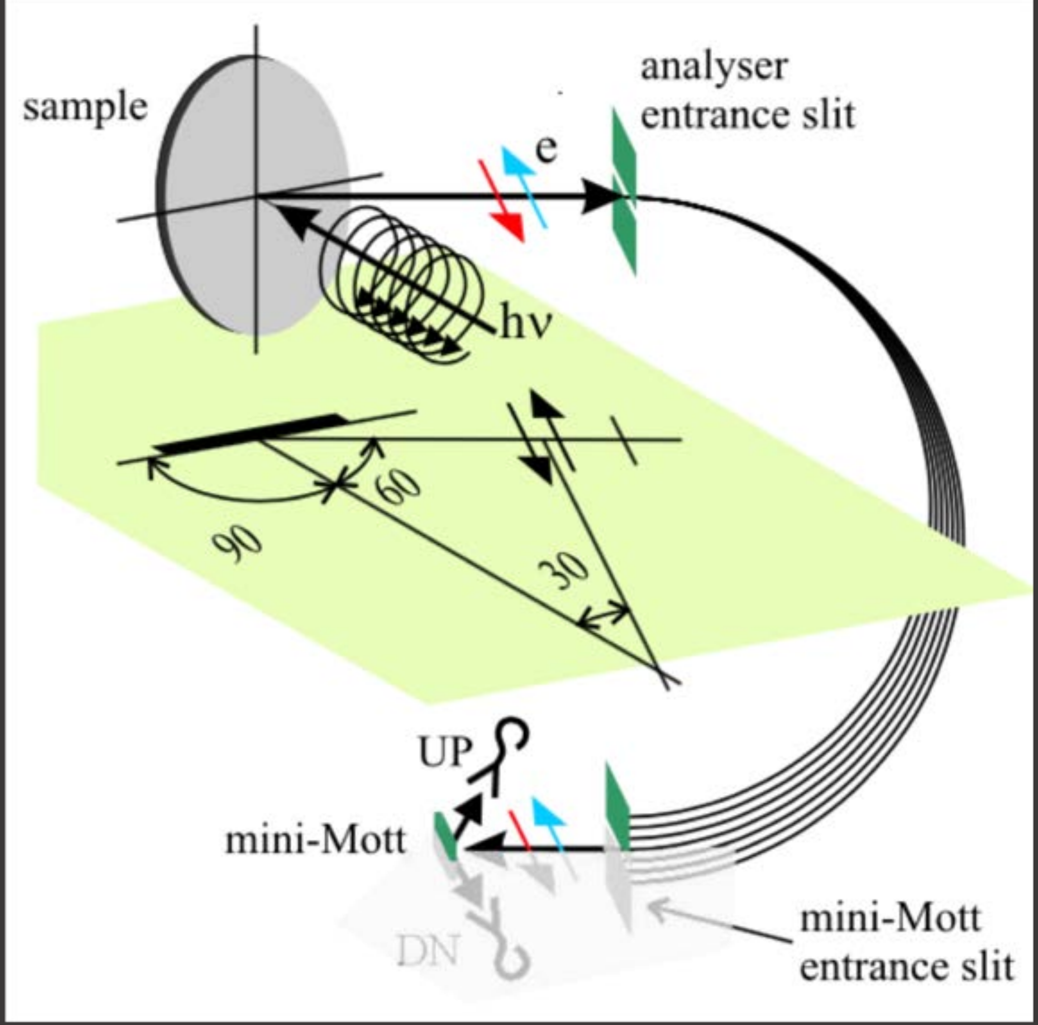
From hemispherical analyzers to time-of-flight analyzers

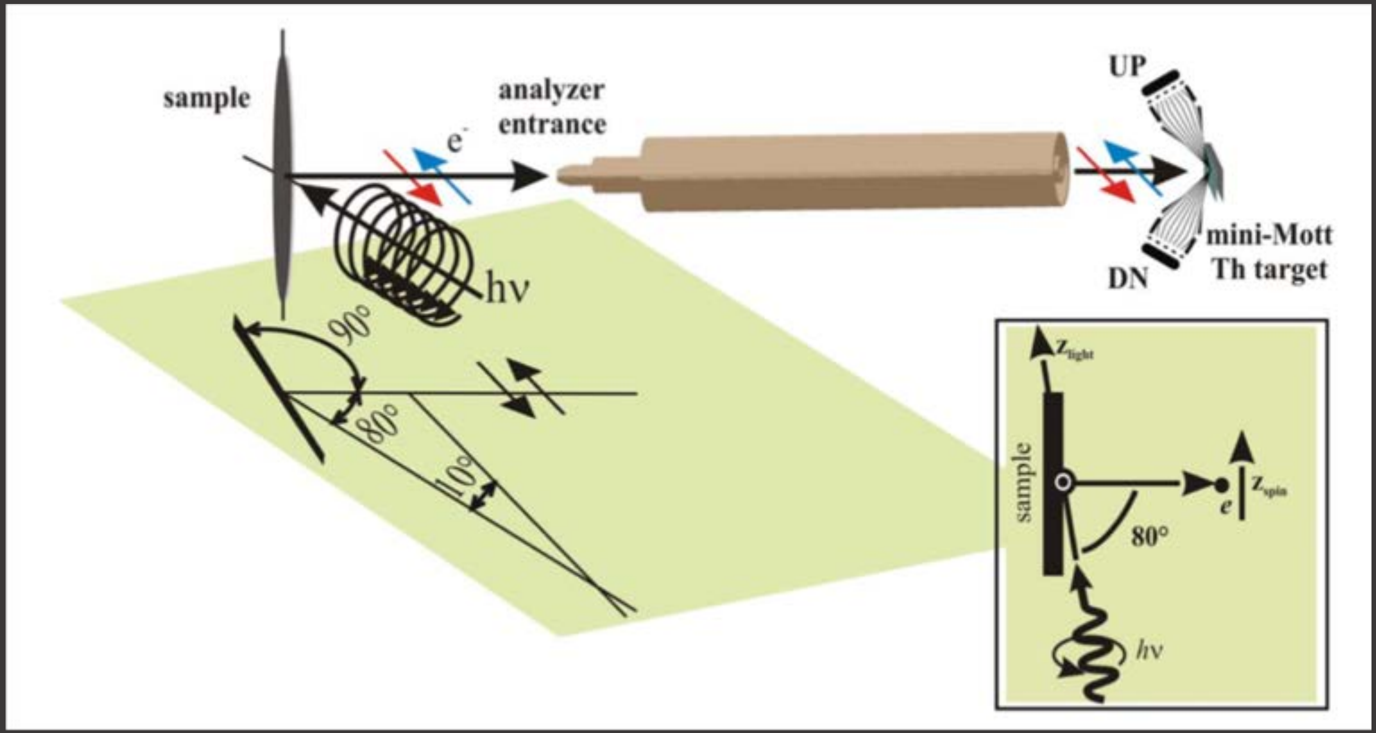
Hemispherical EA-Mott

+ resolution/stability vs hν
- efficiency (serial acquisition)

TOF-Mott

resolution/stability vs hν
+ efficiency (parallel acquisition)

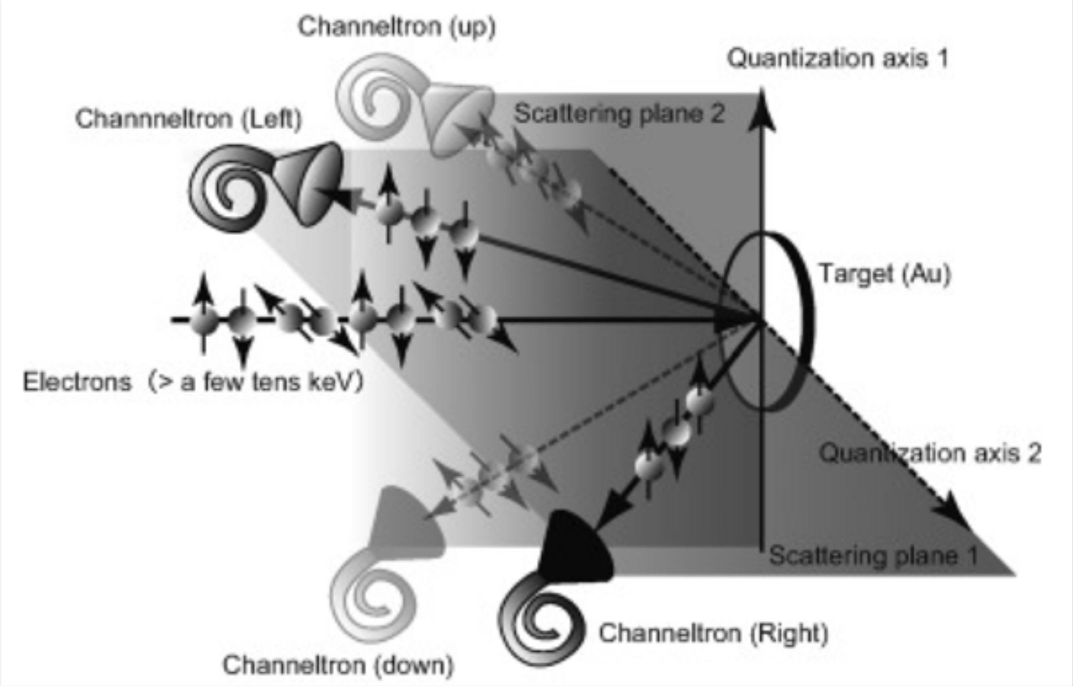




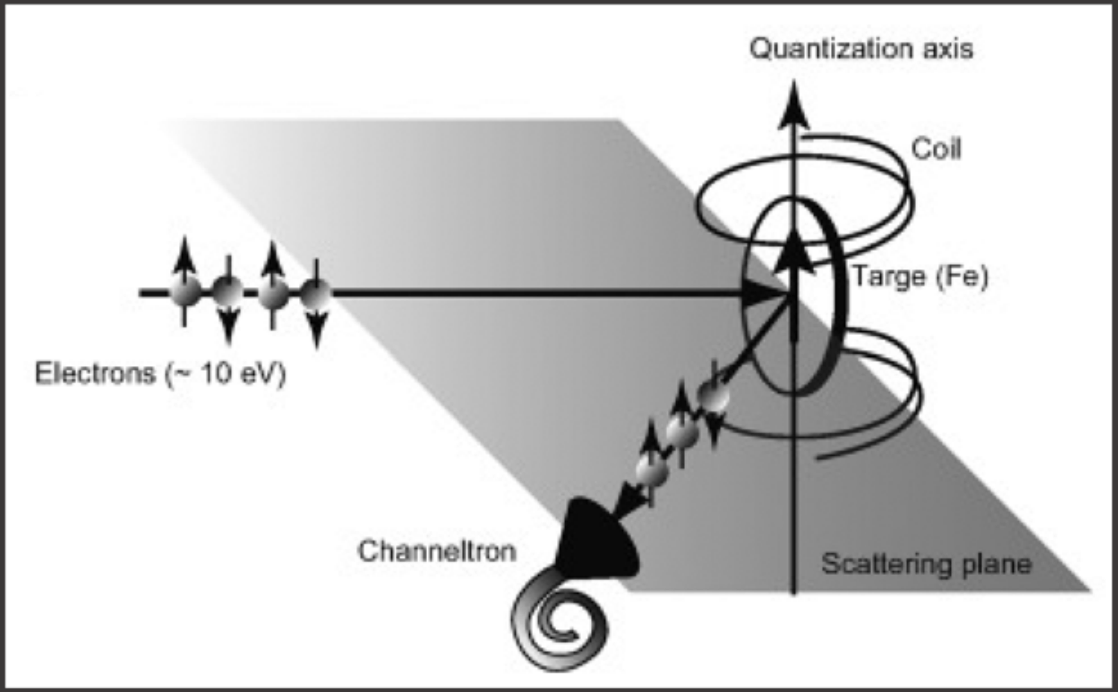
G. Ghiringhelli *et al.*, *Rev. Sci. Instrum.* ***70***, *4225 (1999)* L. Moreschini *et al.*, *Rev. Sci. Instrum.* ***79***, *033905 (2008)*

Coupling spin detection and ARPES

Mott polarimetrs
two axes in parallel

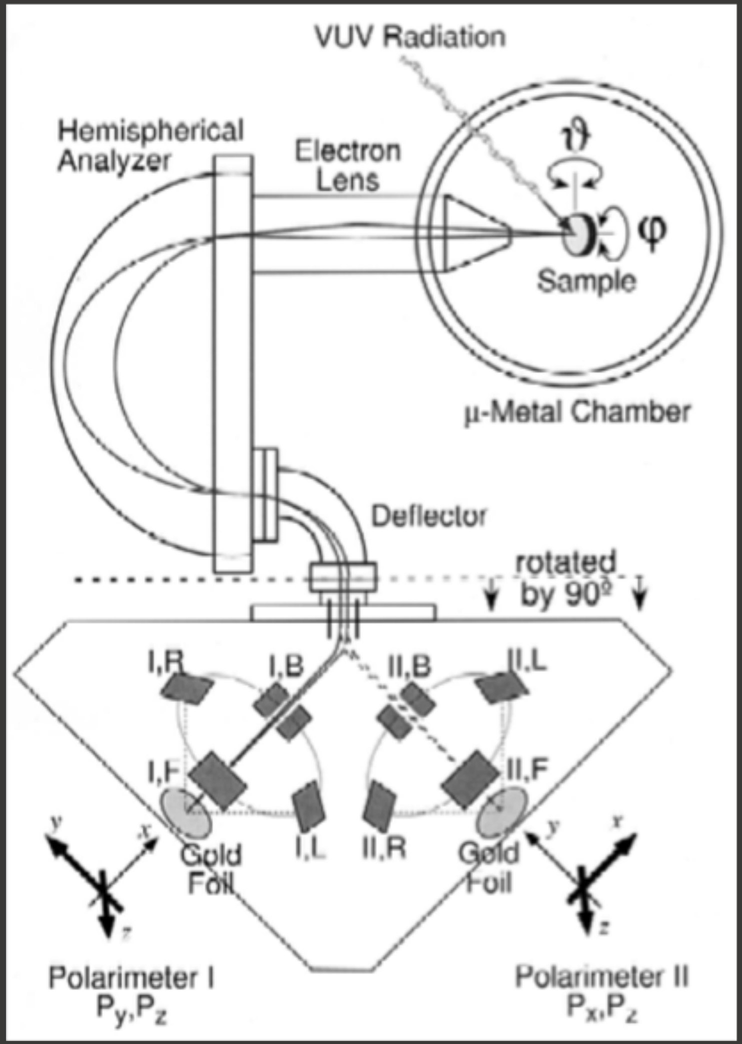


VLEED polarimeters
one axis at a time



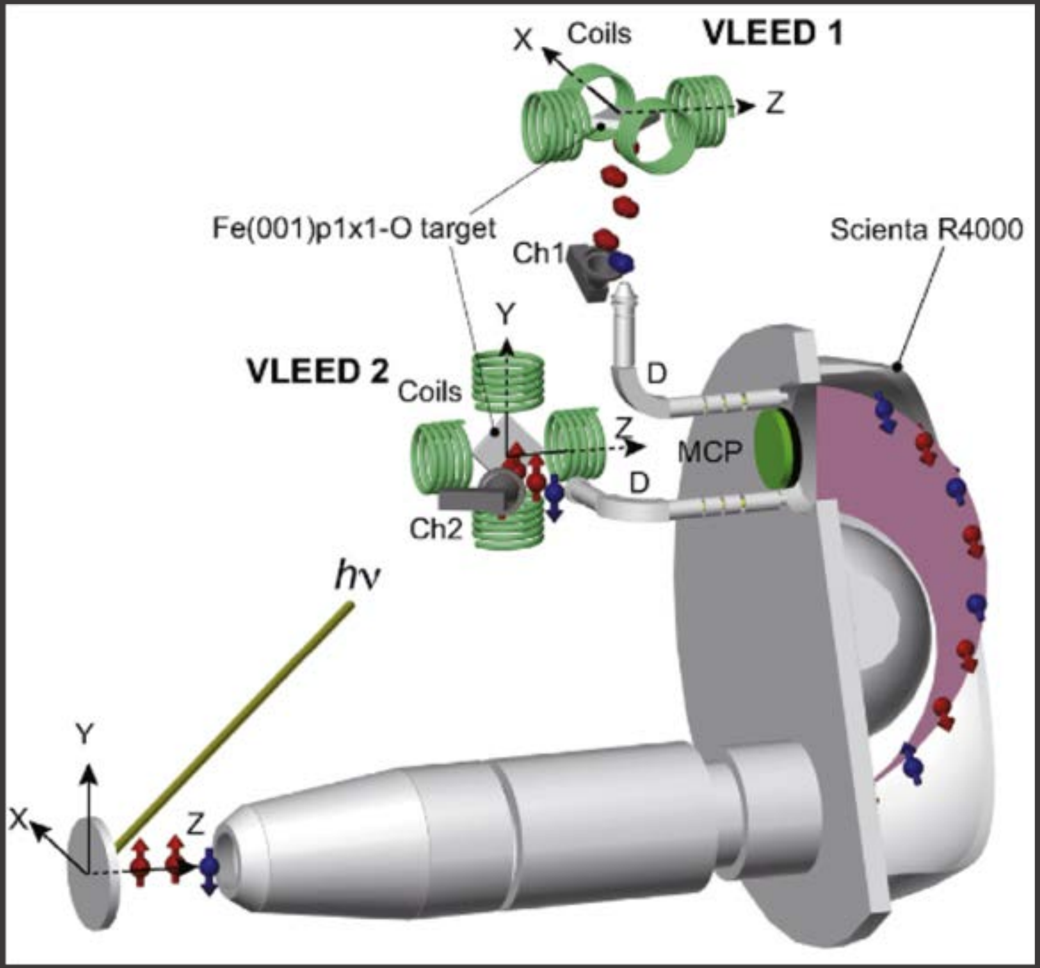
T. Okuda *et al.*, *Rev. Sci. Instrum.* ***201***, *23 (2015)*

Coupling spin detection and ARPES - a few examples



Hemispherical EA + Mott

M. Hoesch *et al.*, *JESRP* ***124****, 263 (2002)*



Hemispherical EA + VLEED

T. Okuda *et al.*, *Rev. Sci. Instrum.* ***82****, 103302 (2011)*
T. Okuda *et al.*, *Rev. Sci. Instrum.* ***201****, 23 (2015)*

Spin polarized electrons in solids (1)
Rashba-Bychkov systems

SO coupling only lifts spin-degeneracy when inversion symmetry is broken

basic symmetry properties in solids:

- space inversion symmetry
- time inversion symmetry

→ band structure is spin-degenerate

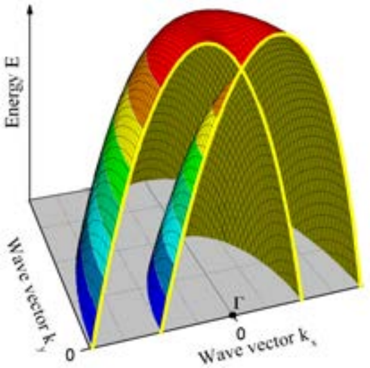
$$\left.\begin{array}{ll}\text{time inversion:} & E(\vec{k},\uparrow)=E(-\vec{k},\downarrow)\\ \text{space inversion:} & E(\vec{k},\uparrow)=E(-\vec{k},\uparrow)\end{array}\right\}\Longrightarrow E(\vec{k},\uparrow\downarrow)=E(\vec{k},\downarrow\uparrow)$$

breaking of inversion symmetry:

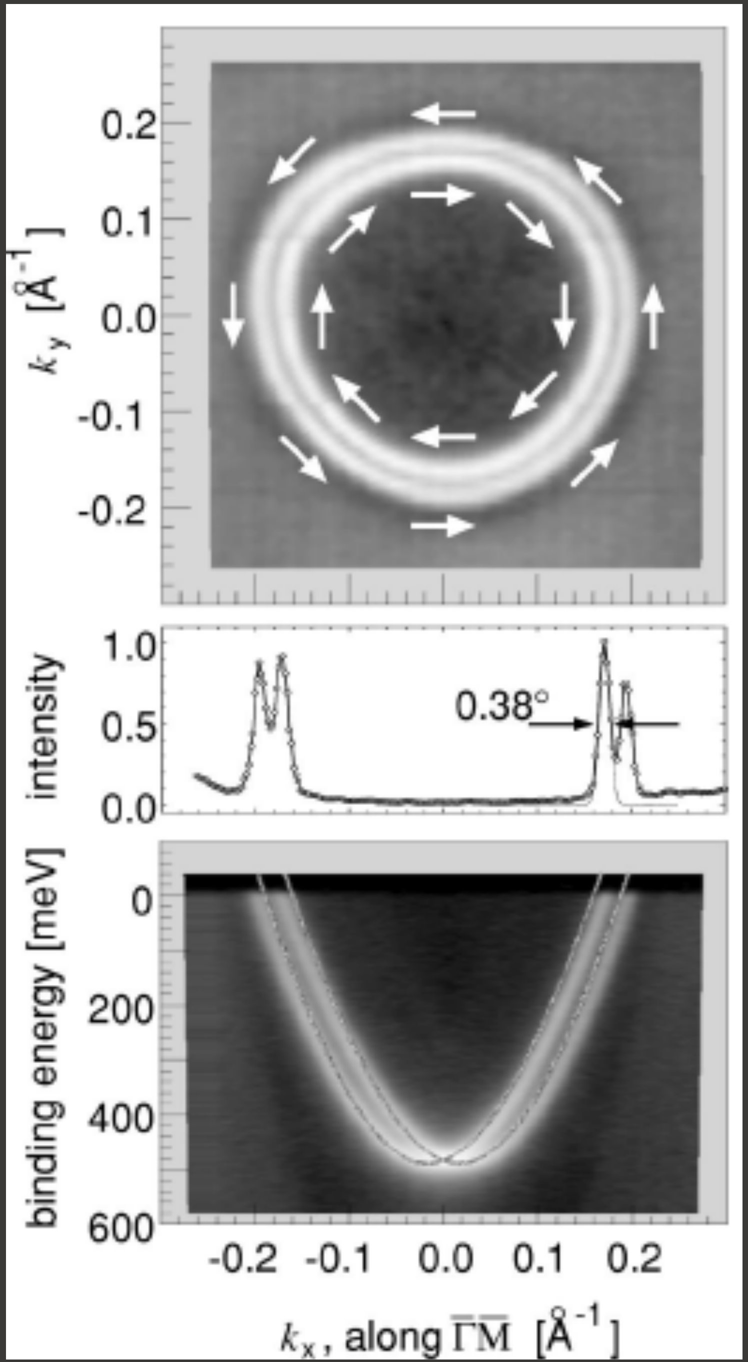
- magnetic fields (time)
- crystals with no inversion center (3D)
- surfaces/interfaces (2D)

→ spin-degeneracy is lifted

Rashba systems - examples



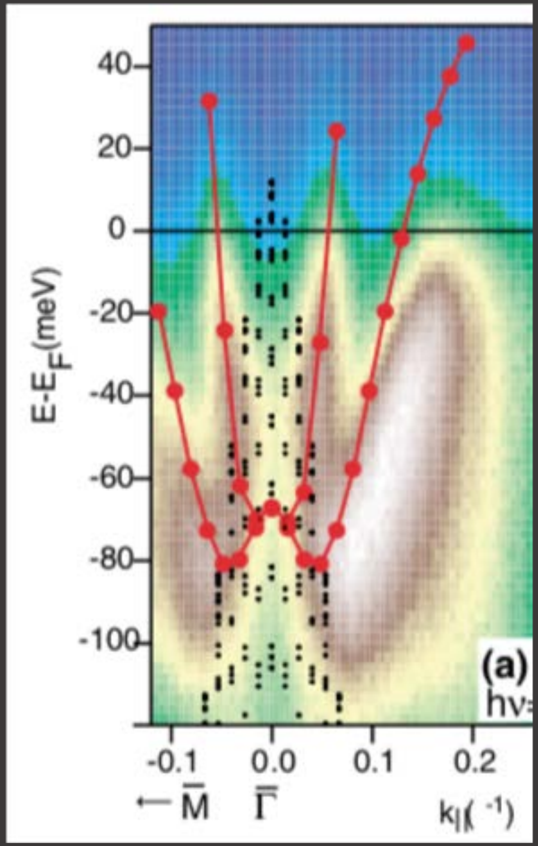
Au(111)



Shockley states in noble metals

G. Nicolay *et al.*, Phys. Rev. B 65, 33407 (2001)

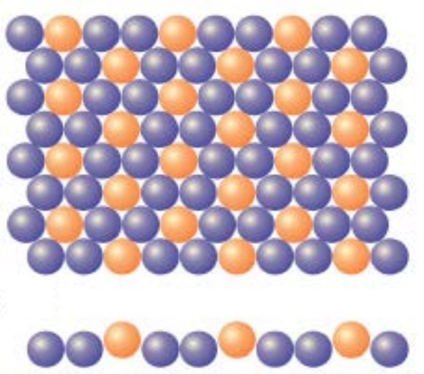
Bi (111)

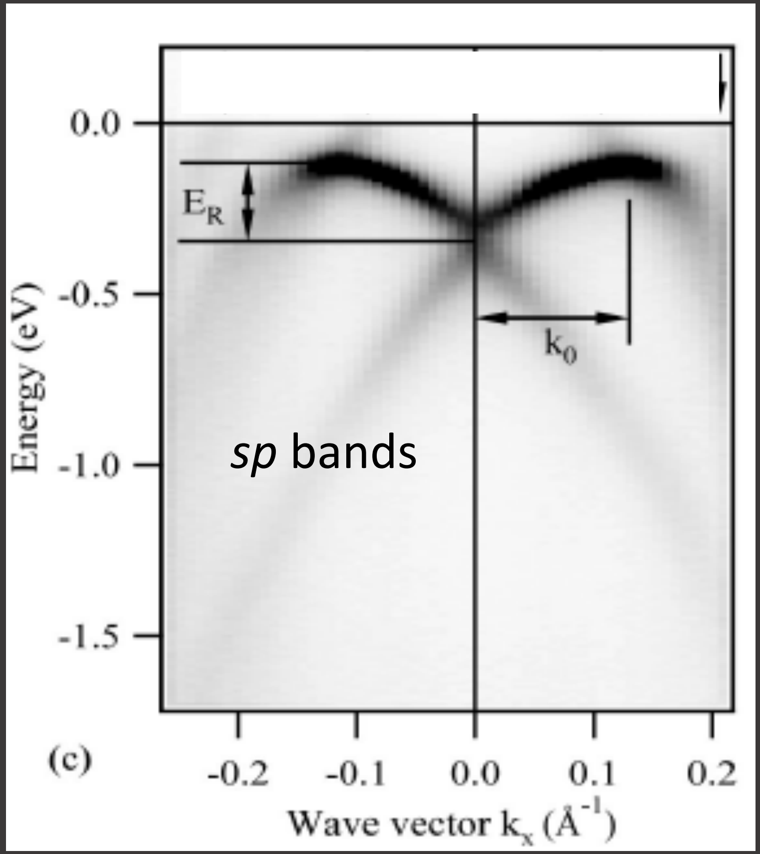


surface states in heavy metals

Y. M. Koroteev *et al.*,
Phys. Rev. Lett. **93**, 046403 (2004)

Bi/Ag(111)

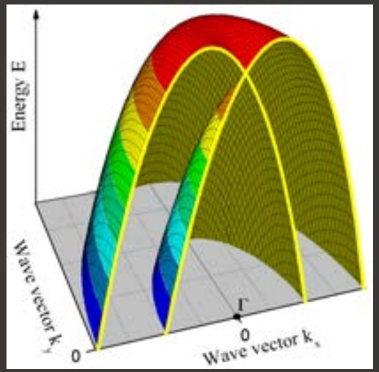




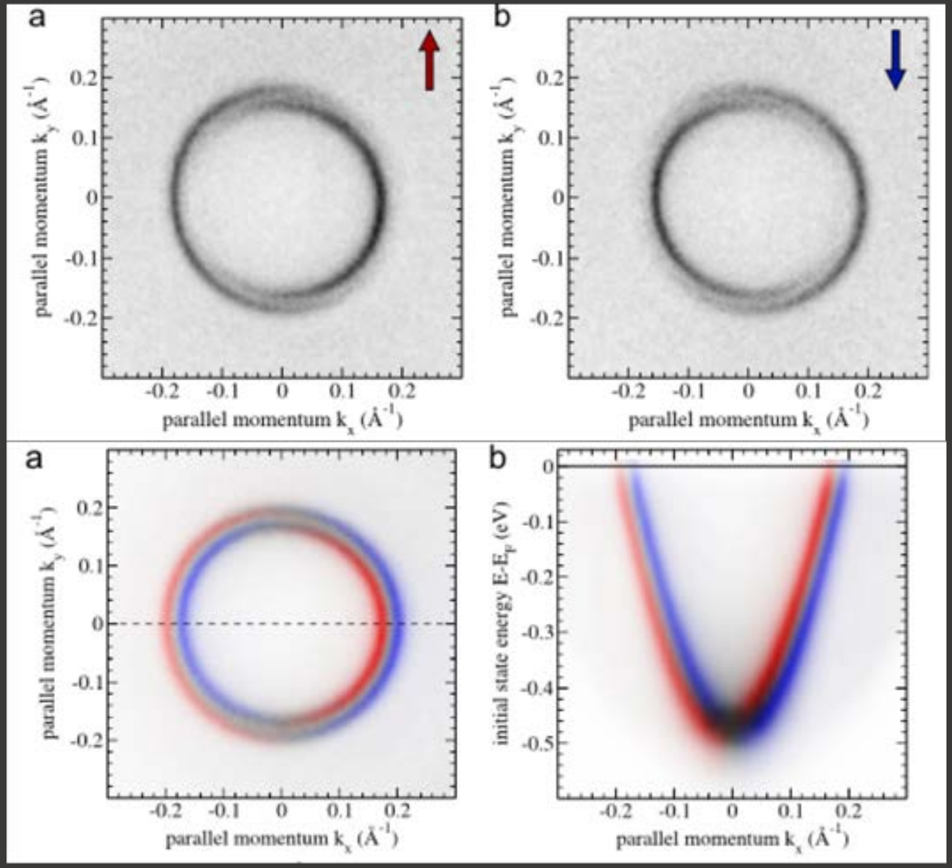
interface states in surface alloys

C. Ast *et al.*, Phys. Rev. Lett. **98**, 186807 (2007)

Rashba systems - examples



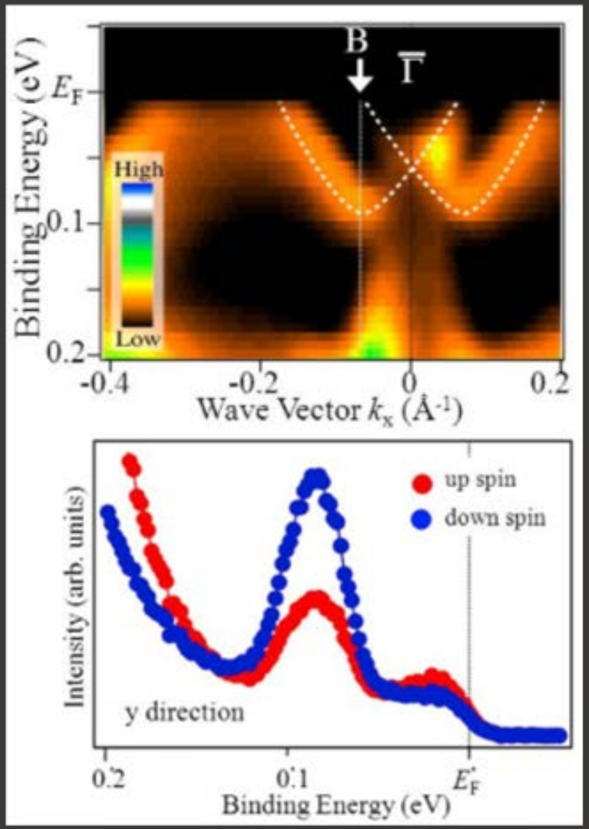
Au(111)



Shockley states in noble metals

C. Tusche *et al.*, Ultramicroscopy, **159**, 520 (2015)

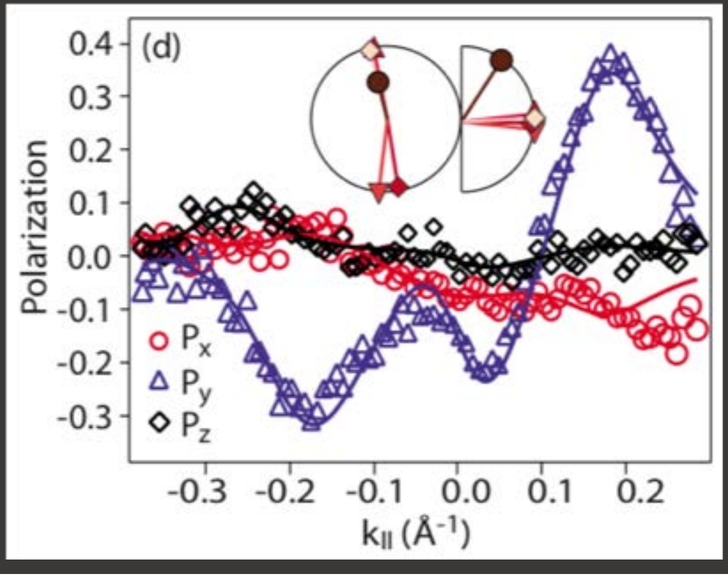
Bi (111)

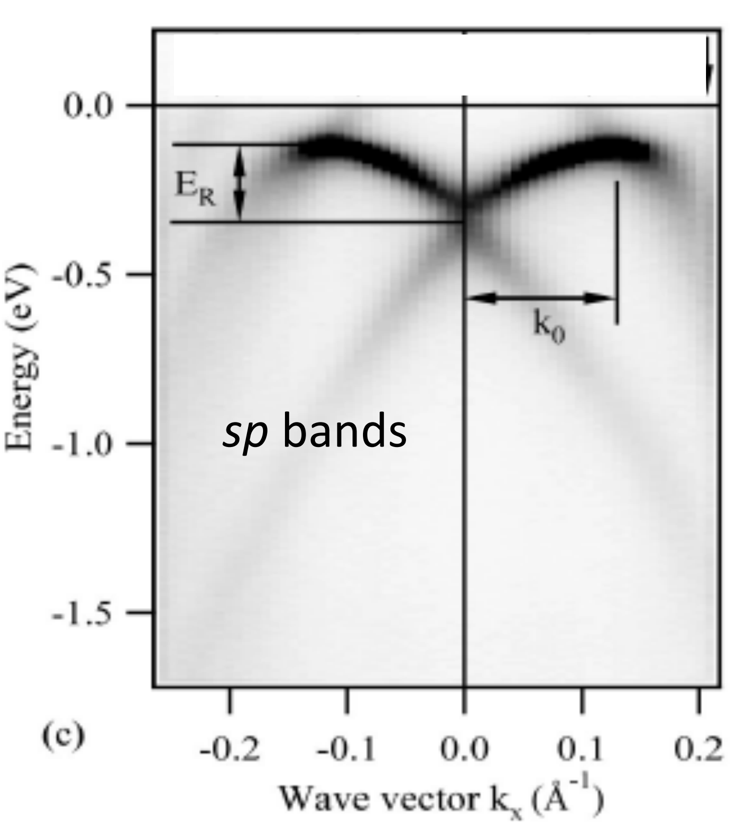


surface states in heavy metals

A Takayama *et al.*,
New J. Phys. **16** 055004 (2014)

Bi/Ag(111)





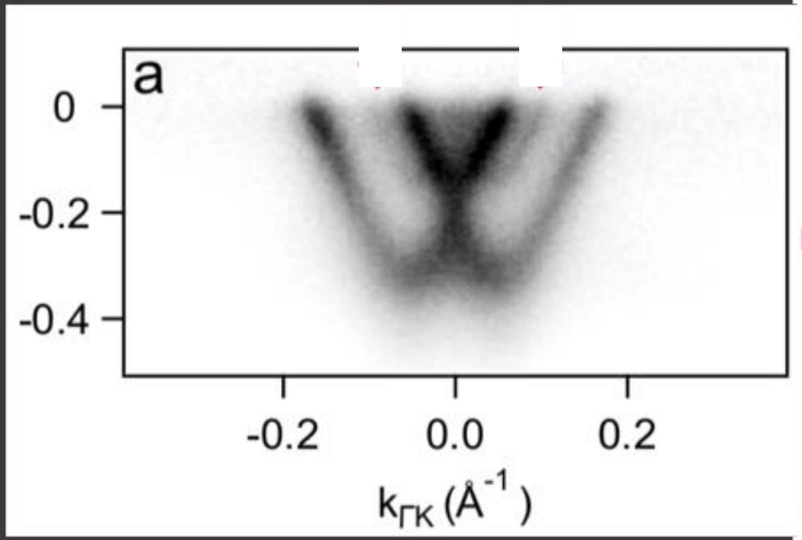
interface states in surface alloys

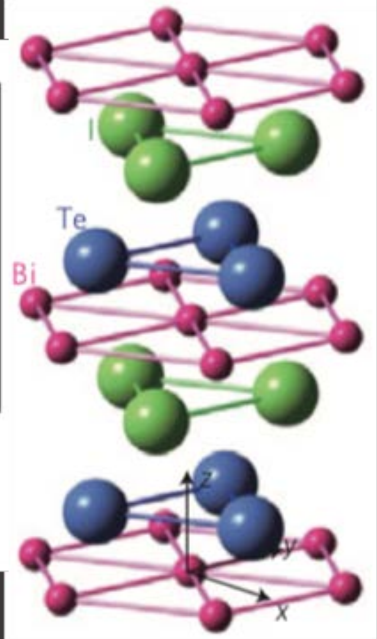
F. Meier *et al.*, Phys. Rev. B **77**, 165431 (2008)

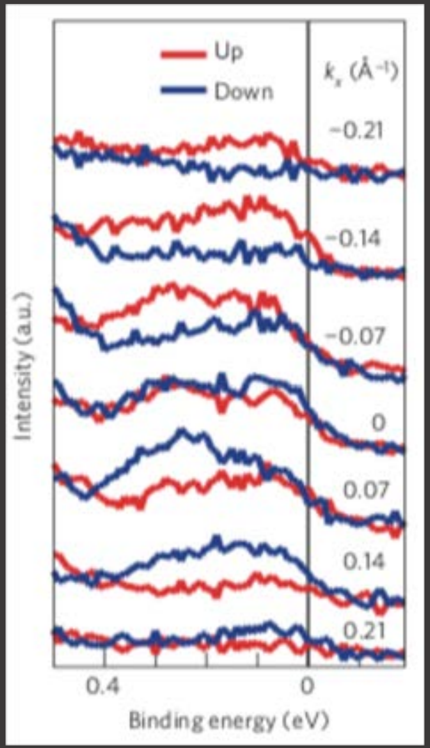
Rashba systems – examples

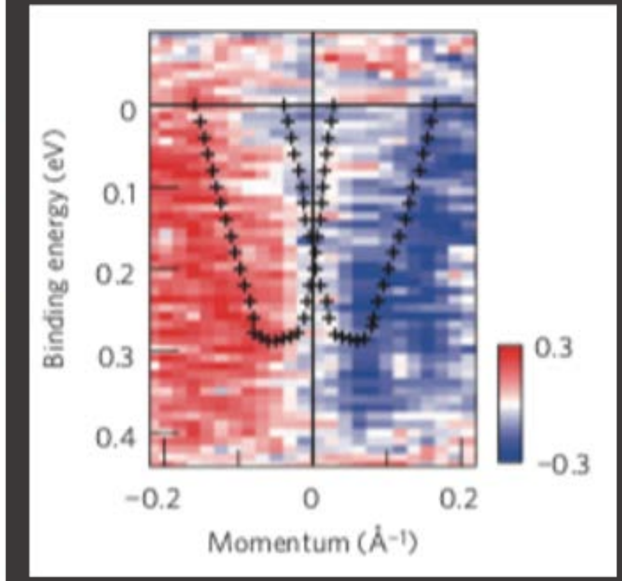
also in bulk! (Dresselhaus effect)

BiTeI





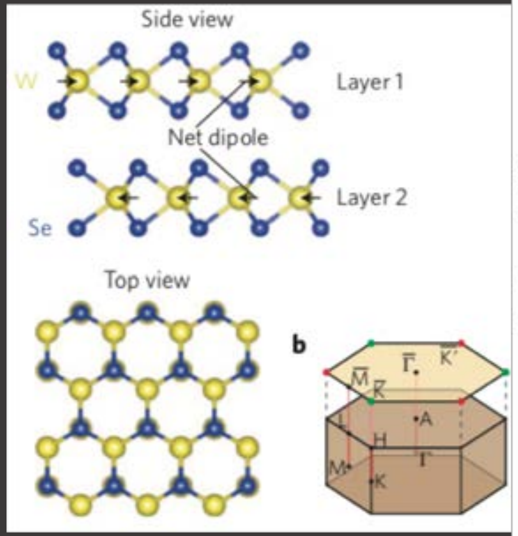


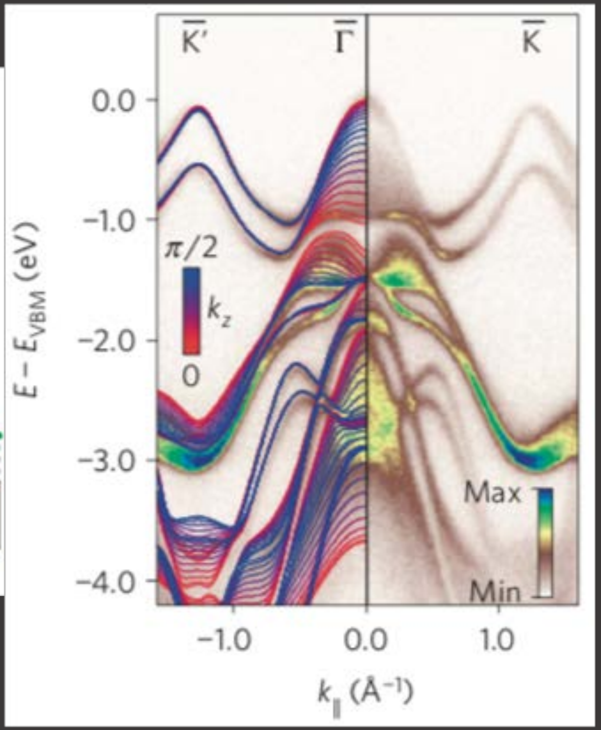


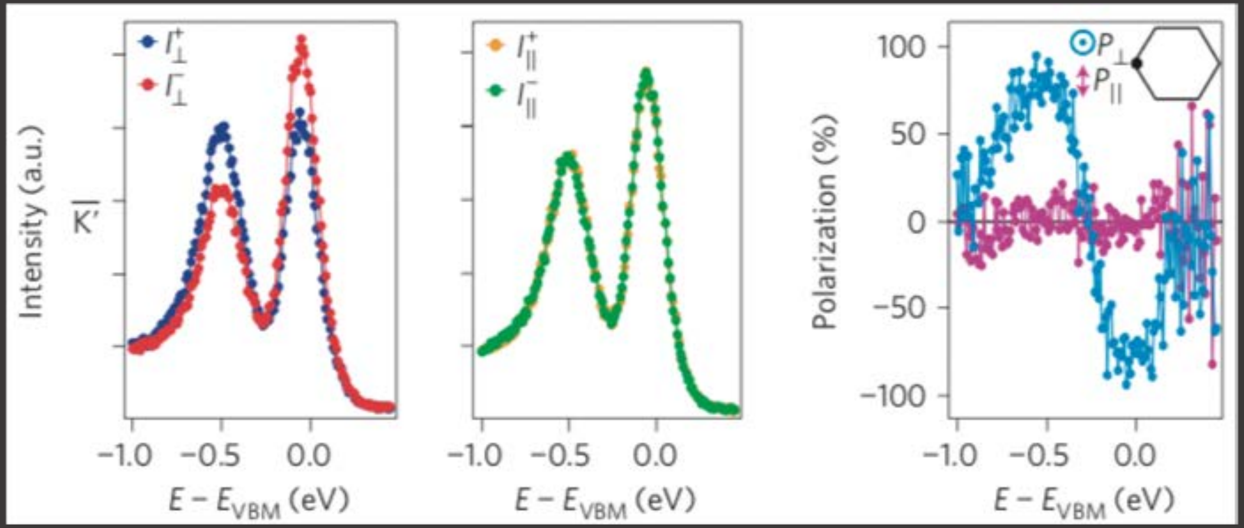
A. Crepaldi *et al.*, Phys. Rev. Lett. **109**, 096803 (2012)
K. Ishizaka *et al.*, Nature Mater. **10**, 521 (2011)

and also in centrosymmetric crystals if the wavefunctions are localized in a portion of the unit cell

WSe_2

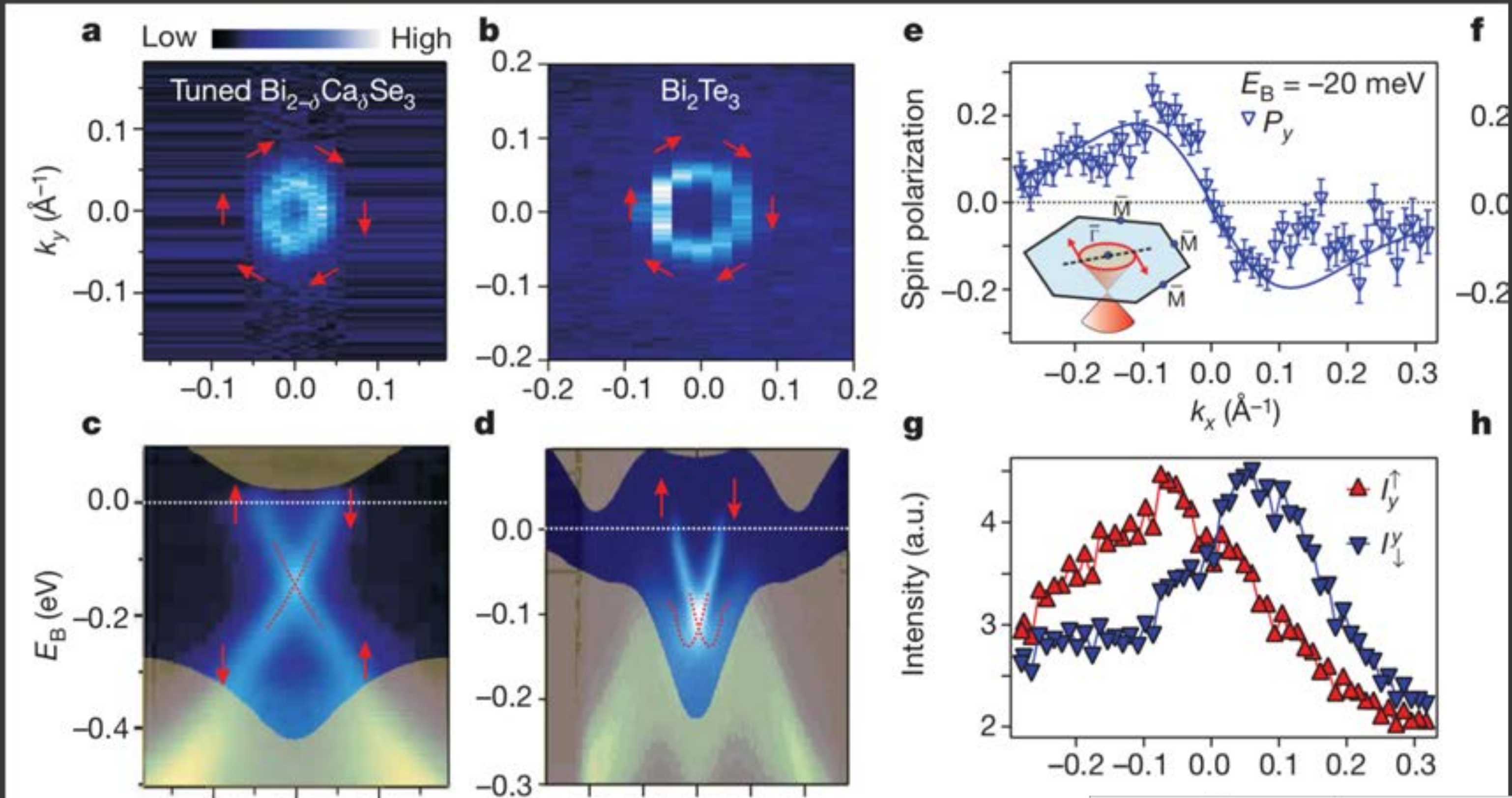






J. M. Riley *et al.*, Nature Phys. **10**, 835 (2014)

Spin polarized electrons in solids (2)
Topological insulators

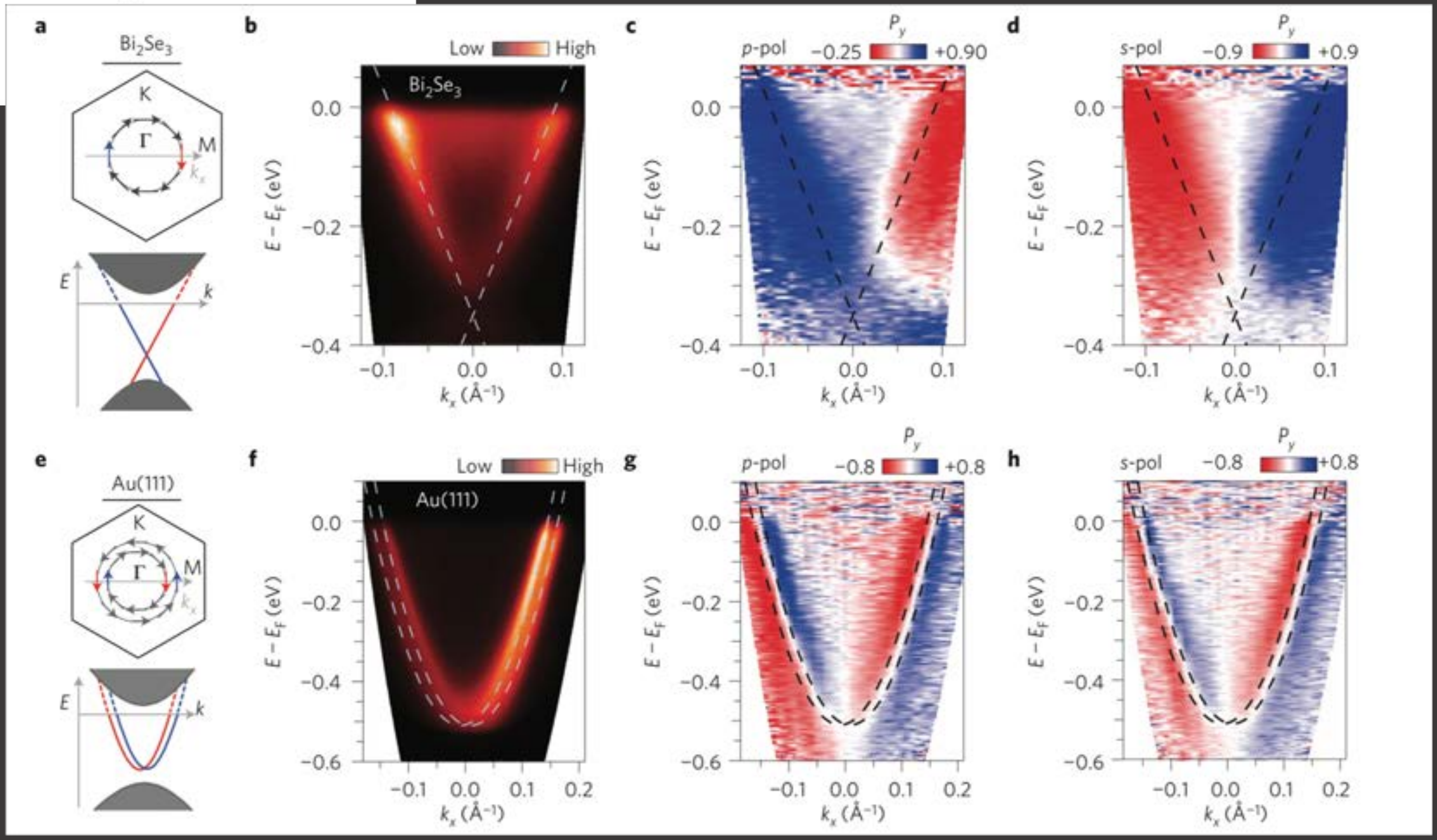


D. Hsieh *et al.*, Nature **460**, 1101 (2009)

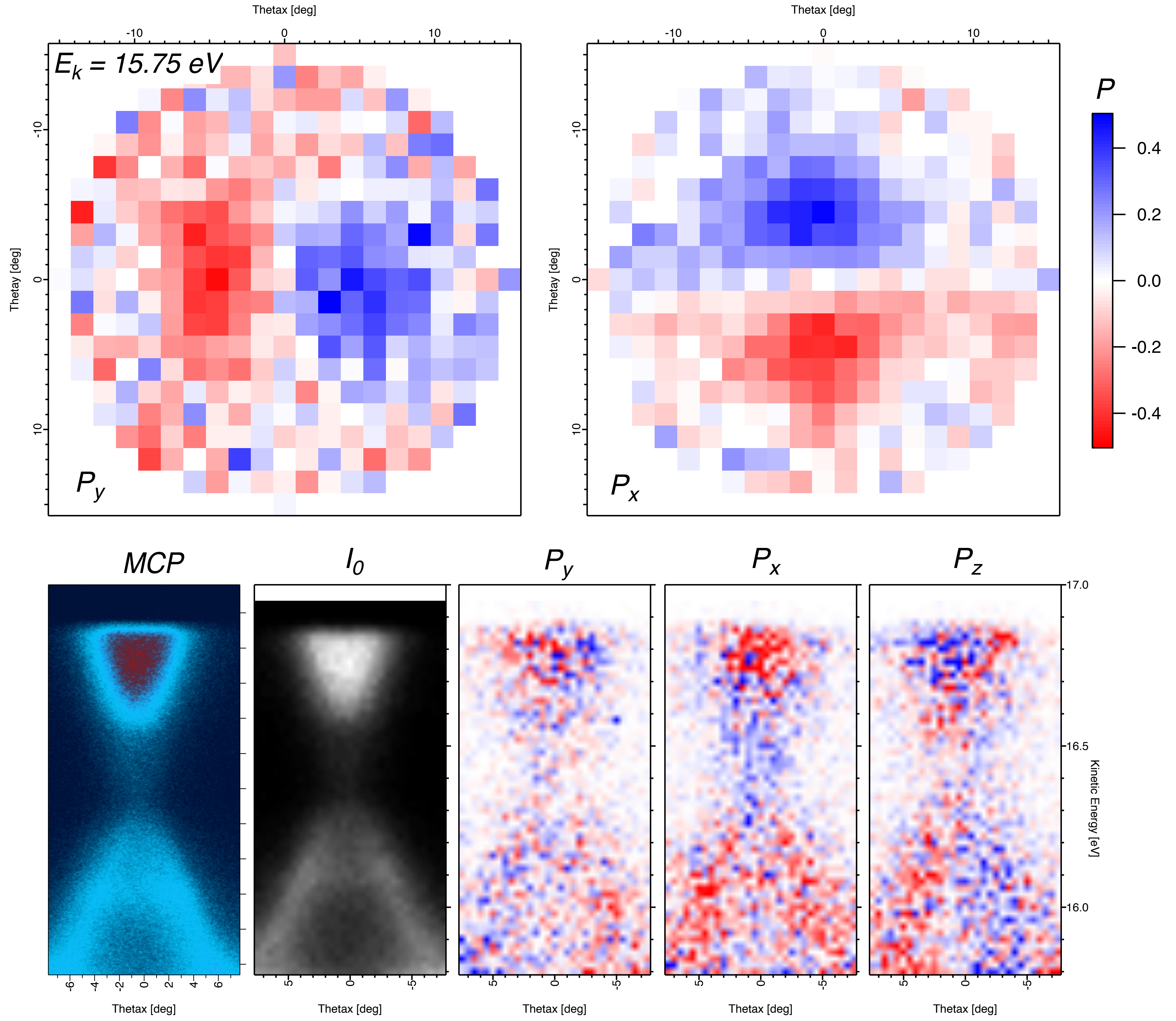
from a plain vanilla picture

to a more involved one

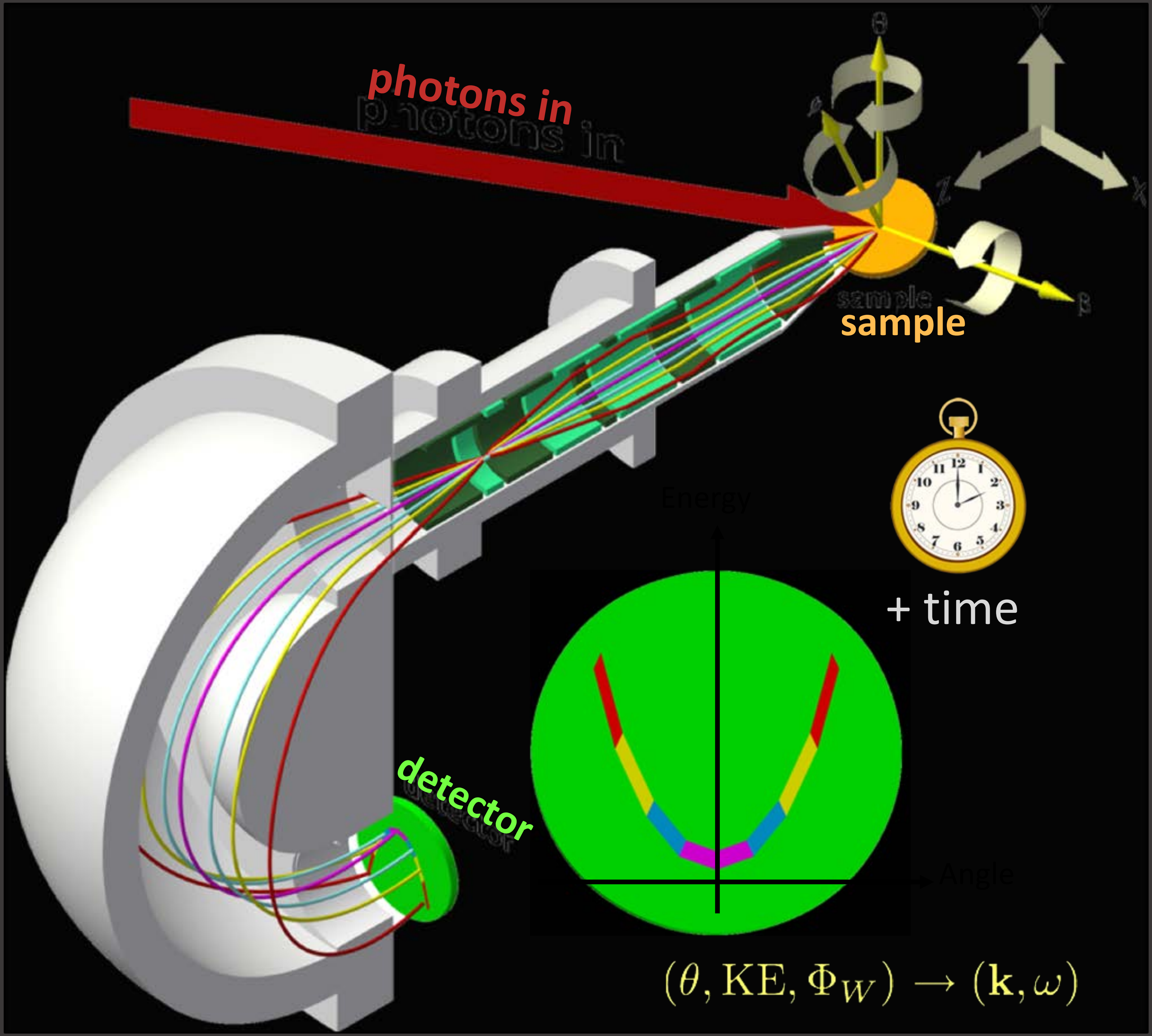
C. Jozwiak *et al.*, Nature Phys. **9**, 293 (2013)



Spin-Resolved ARPES using VLEED detection

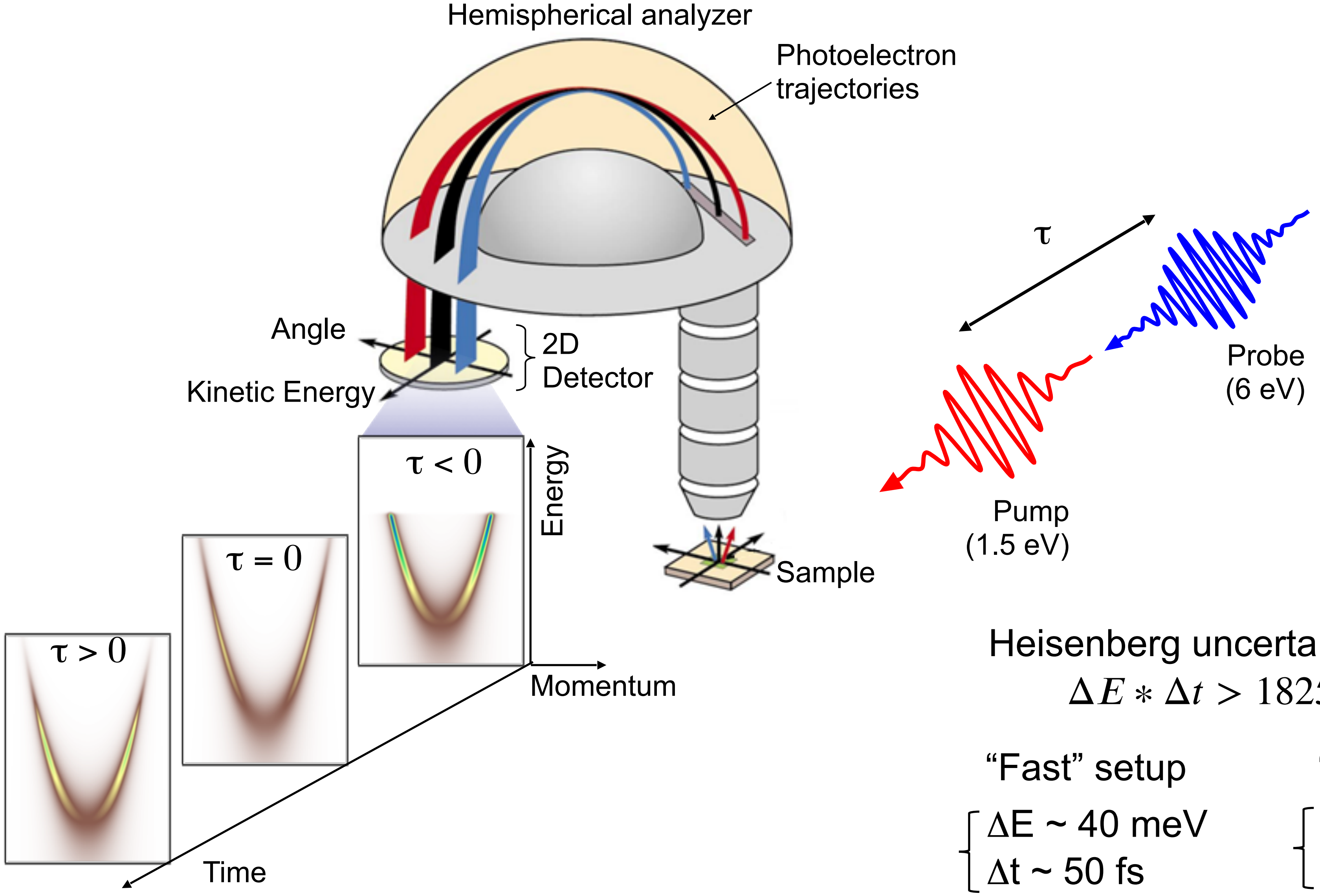


Angle-resolved photoelectron spectroscopy
+ something else



$$(\theta, \mathrm{KE}, \Phi_W) \to (\mathbf{k}, \omega)$$

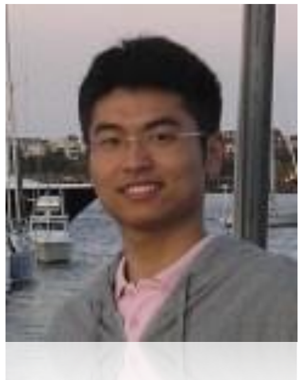
Time-Resolved ARPES : Visualizing Electron Dynamics



Heisenberg uncertainty principle:

$$\Delta E * \Delta t > 1825 \text{ meV.fs}$$

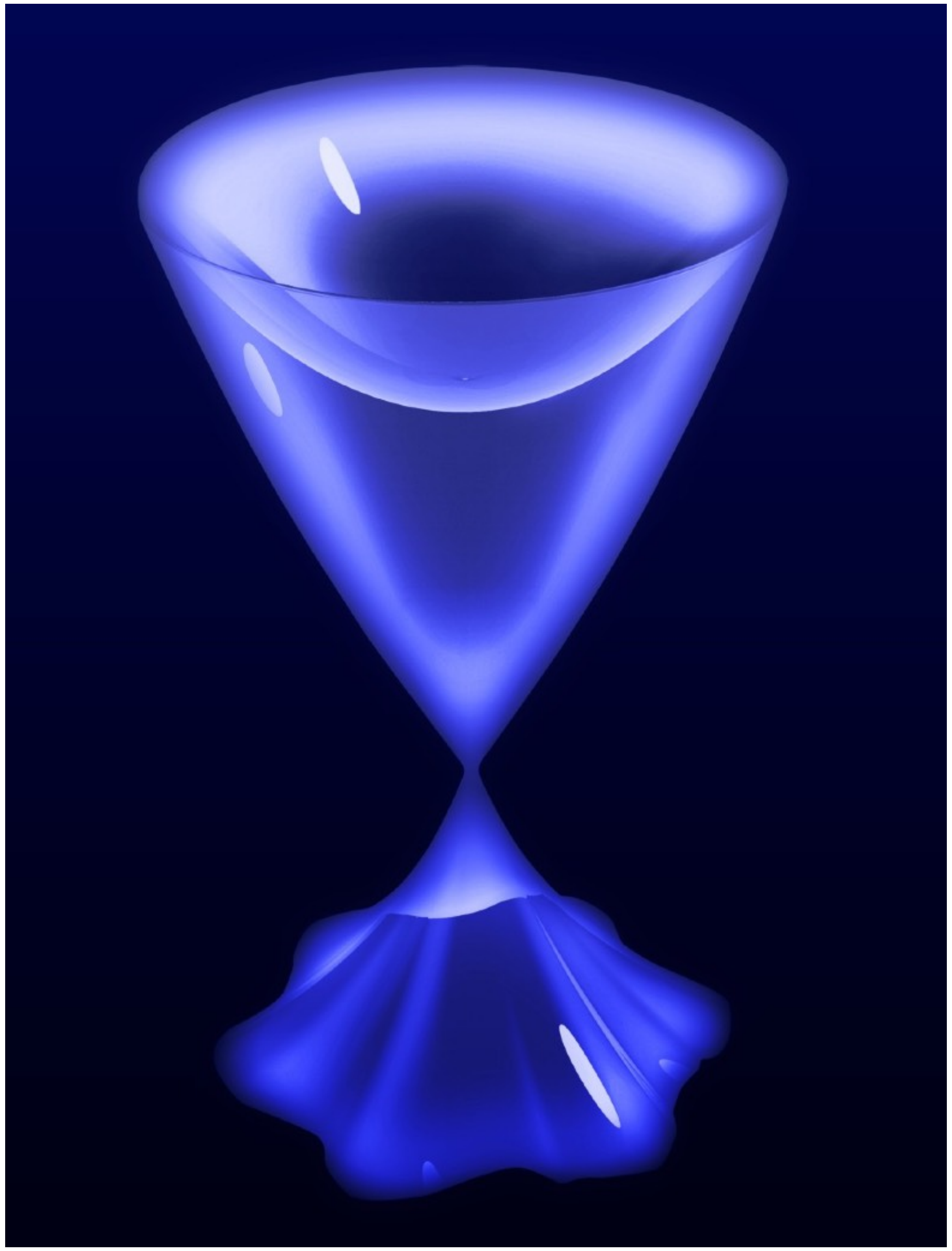
"Fast" setup	"Slow" setup
$\Delta E \sim 40$ meV	$\Delta E \sim 20$ meV
$\Delta t \sim 50$ fs	$\Delta t \sim 100$ fs

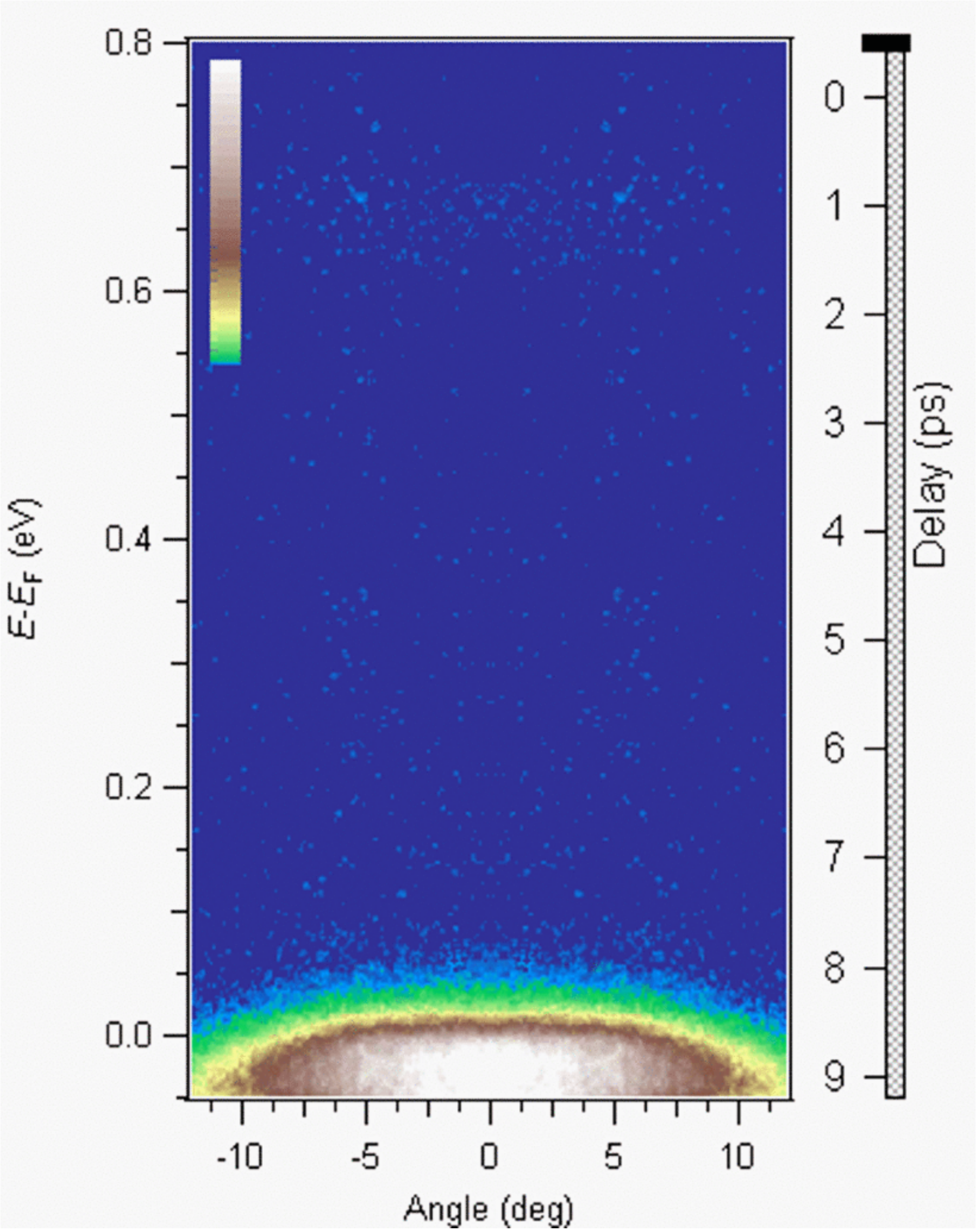


Slides courtesy of Shuolong Yang (U. Chicago)

Surface states in topological insulators

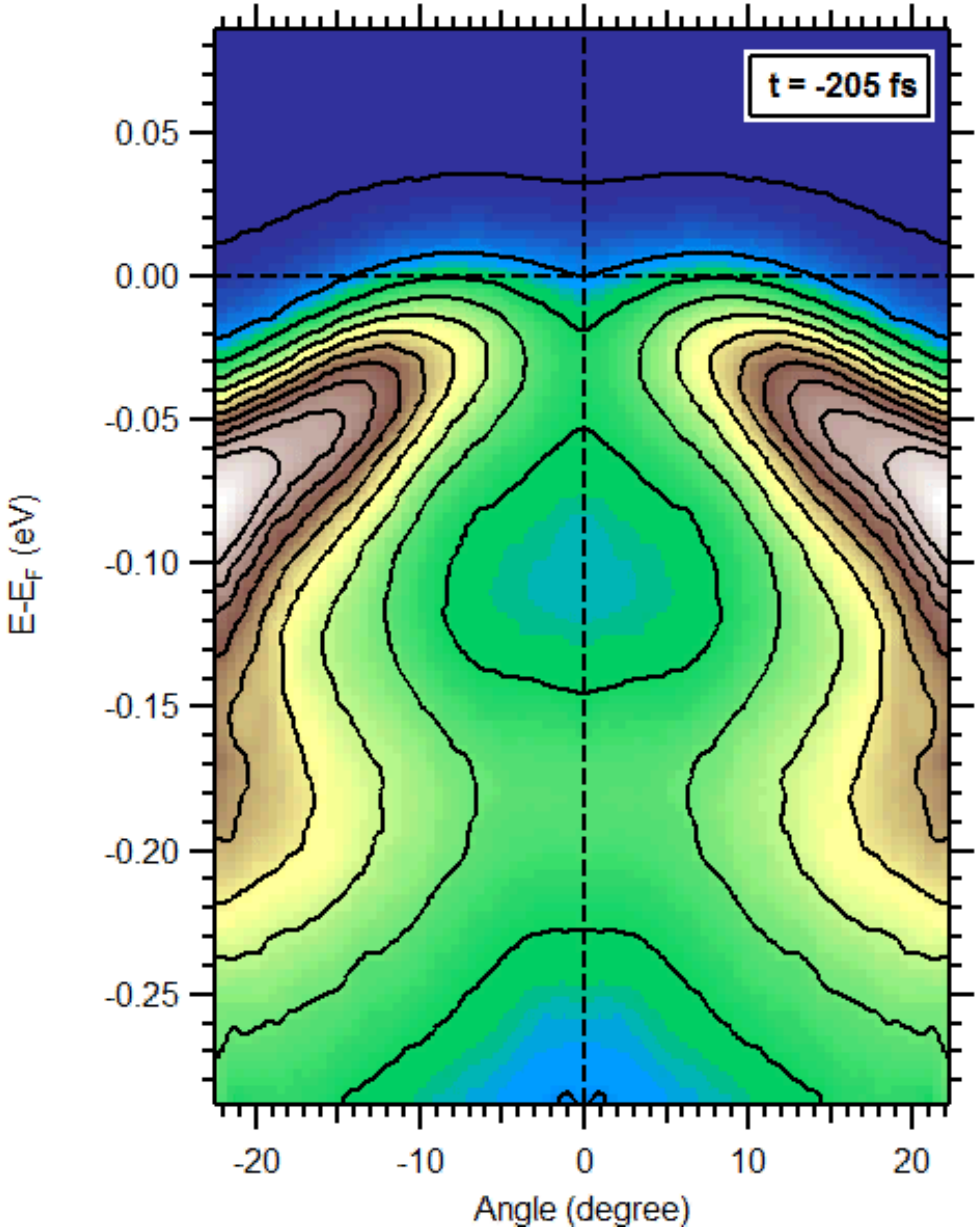
Topological insulator Bi_2Se_3



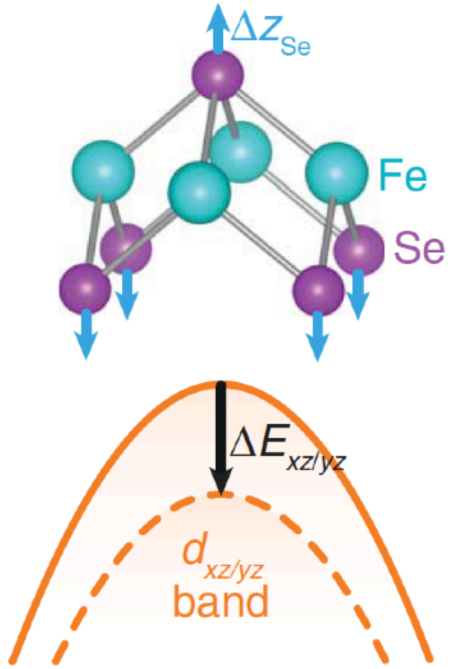


Sobota and Yang et al., PRL 108, 117403 (2012)

Coherent vibration of FeSe electronic bands



Perturbation launches coherent modes



Gerber* and Yang* *et al. Science* **357,** 71 (2017)

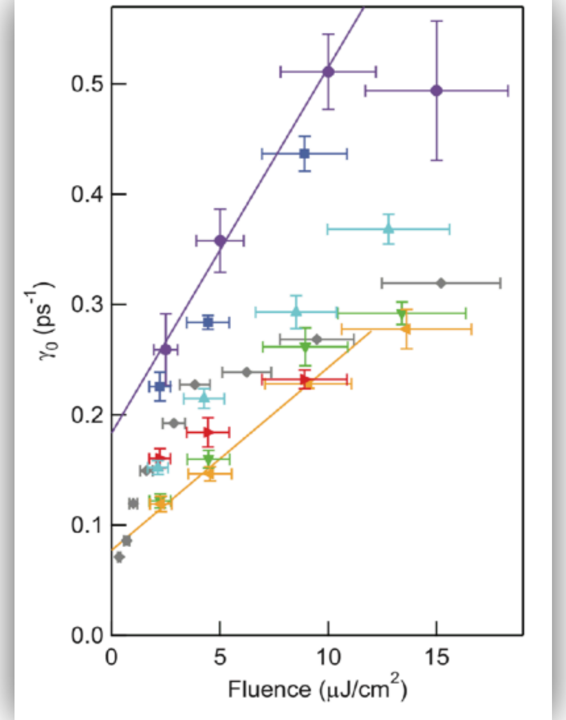
What is Time-Resolved ARPES good for?

trARPES tool set

- Resolving lifetimes
- Launching collective excitations
- Promoting electrons above E_F
- Transiently modifying emergent properties

⋮

Cuprates [1]



Material physics problems

- Complex scattering processes
- Collective modes
- Unoccupied band structure
- Material control

⋮

[1] Smallwood *et al. Science* **336**, 1137 (2012) [2] Schmitt *et al. Science* **321,** 1649 (2008)
[3] Sobota *et al. Phys. Rev. Lett.* **111**, 136802 (2013) [4] Mahmood *et al. Nat. Phys.* Advance Online Publication (2016)

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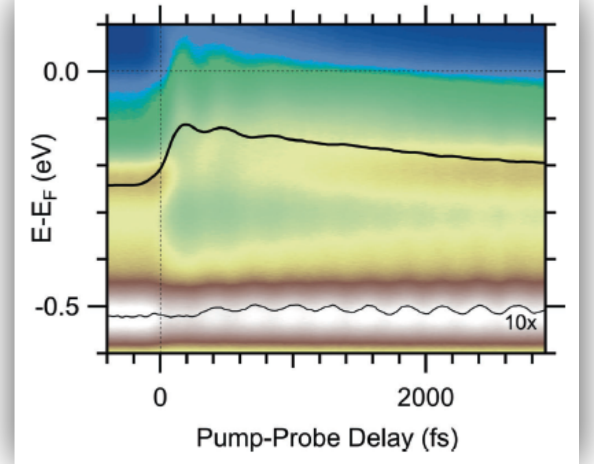
$TbTe_3$ [2]



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[1] Smallwood *et al. Science* **336**, 1137 (2012) [2] Schmitt *et al. Science* **321,** 1649 (2008)
[3] Sobota *et al. Phys. Rev. Lett.* **111**, 136802 (2013) [4] Mahmood *et al. Nat. Phys.* Advance Online Publication (2016)

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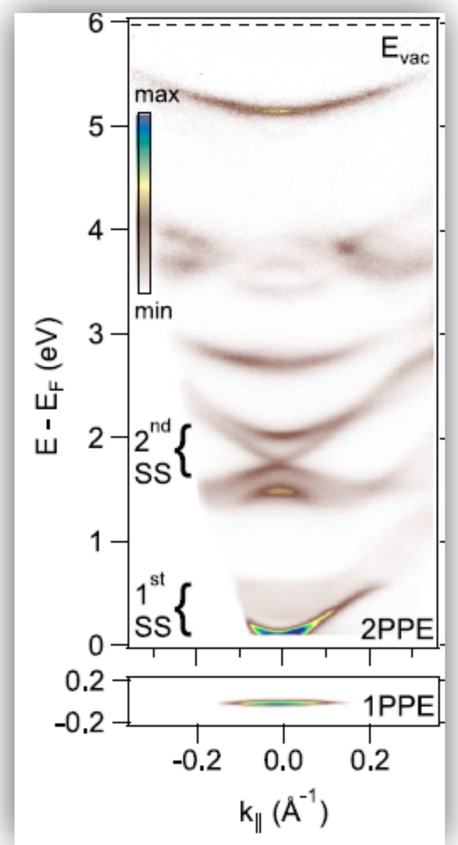
- ⋮

Bi_2Se_3 [3]



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- ⋮

[1] Smallwood *et al. Science* **336**, 1137 (2012) [2] Schmitt *et al. Science* **321,** 1649 (2008)
[3] Sobota *et al. Phys. Rev. Lett.* **111**, 136802 (2013) [4] Mahmood *et al. Nat. Phys.* Advance Online Publication (2016)

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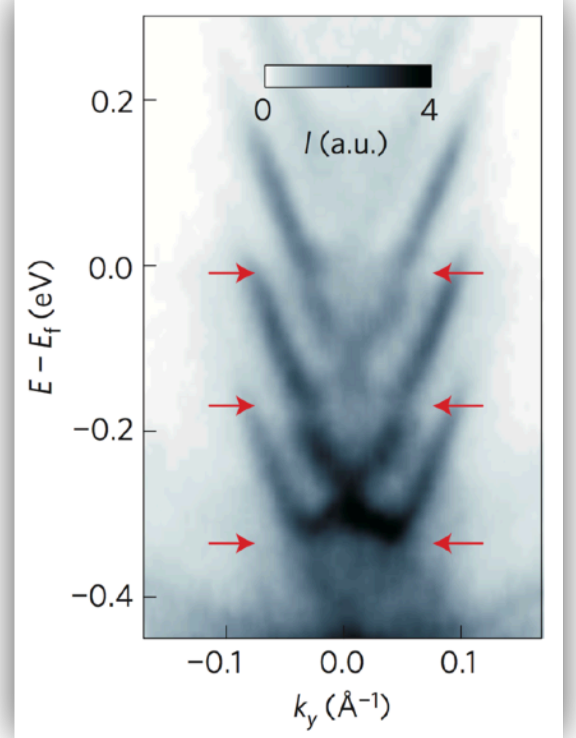
- ⋮

Bi_2Se_3 [4]



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- ⋮

[1] Smallwood *et al. Science* **336**, 1137 (2012) [2] Schmitt *et al. Science* **321,** 1649 (2008)
[3] Sobota *et al. Phys. Rev. Lett.* **111**, 136802 (2013) [4] Mahmood *et al. Nat. Phys.* Advance Online Publication (2016)

TR-ARPES
some useful reviews

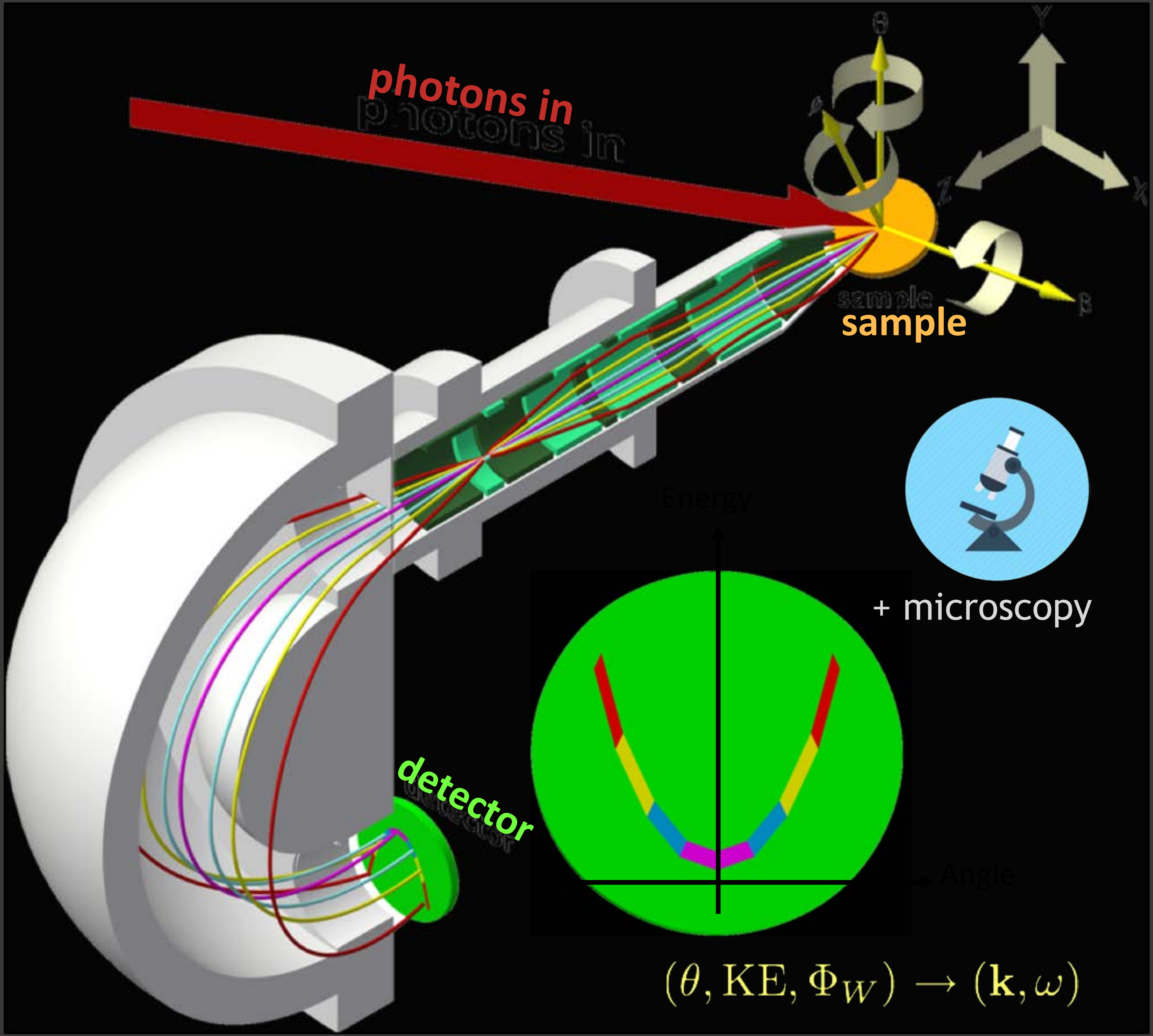
experiment

S. Mathias *et al.*, J. of Phys.: Conf. Ser. **148**, 012042 (2009)
U. Bovensiepen and P. S. Kirchmann, Laser Photonics Rev. **6**, 589 (2012)
C. Giannetti *et al.*, Adv. in Phys. **65**, 58 (2016)
C. L. Smallwood *et al.*, Europhys. Lett. **115**, 27001 (2016)

theory

H. Aoki *et al.*, Rev. Mod. Phys. **86**, 779 (2014)
A. F. Kemper *et al.*, Ann. Phys. 1600235 (2017)

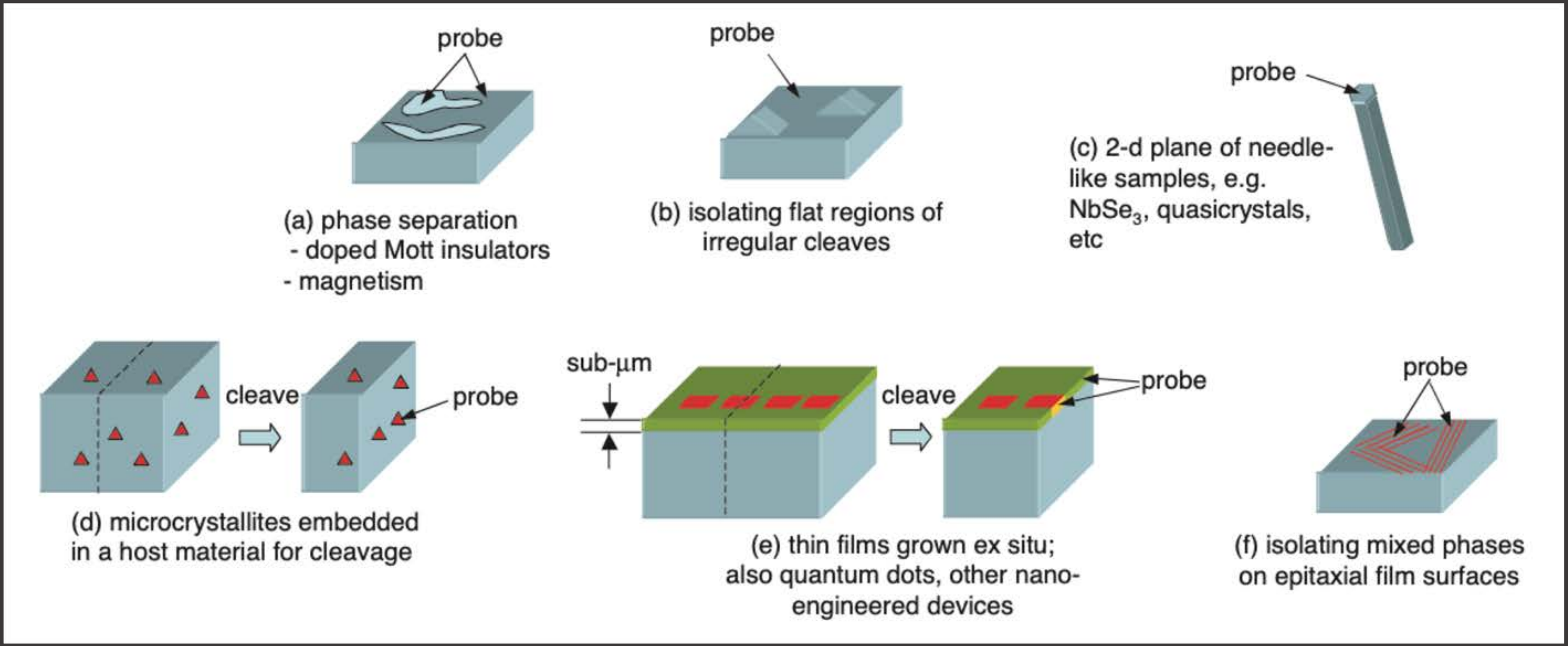
Angle-resolved photoelectron spectroscopy
+ something else



+ microscopy

$$(\theta, \mathrm{KE}, \Phi_W) \rightarrow (\mathbf{k}, \omega)$$

The case for going smaller



(a) phase separation
- doped Mott insulators
- magnetism

(b) isolating flat regions of irregular cleaves

(c) 2-d plane of needle-like samples, e.g. $NbSe_3$, quasicrystals, etc

(d) microcrystallites embedded in a host material for cleavage

(e) thin films grown ex situ; also quantum dots, other nano-engineered devices

(f) isolating mixed phases on epitaxial film surfaces

ackn. E. Rotenberg

TR-ARPES
some useful reviews

experiment

S. Mathias *et al.*, J. of Phys.: Conf. Ser. **148**, 012042 (2009)
U. Bovensiepen and P. S. Kirchmann, Laser Photonics Rev. **6**, 589 (2012)
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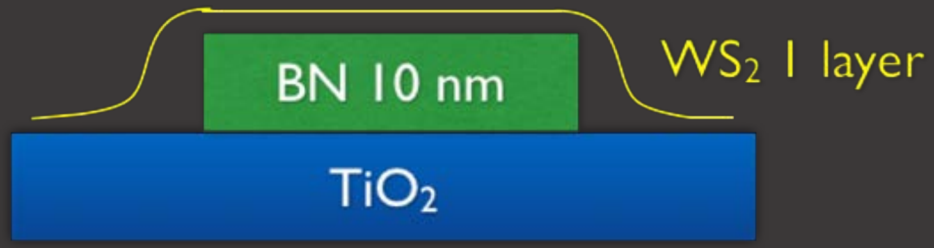
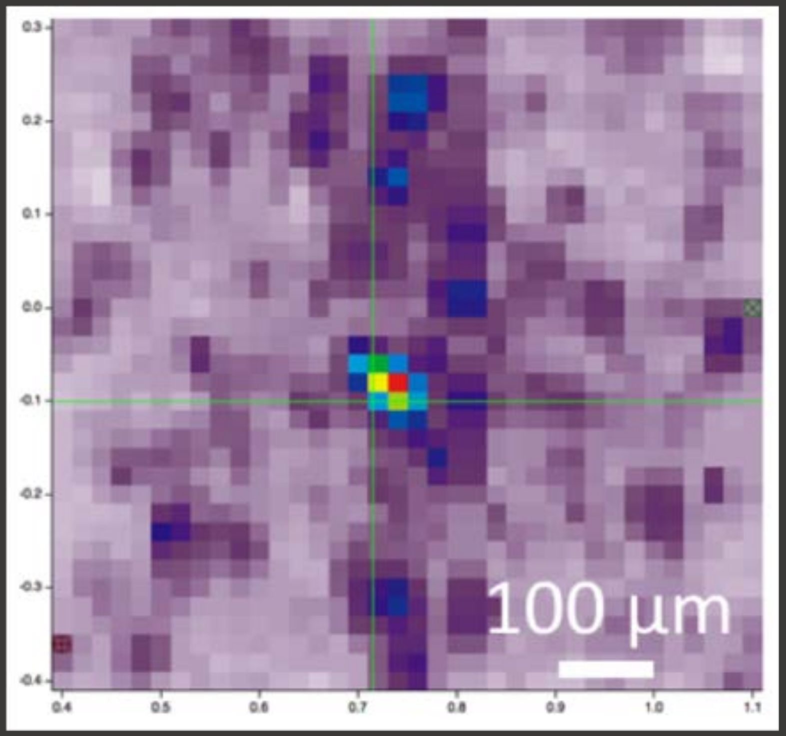
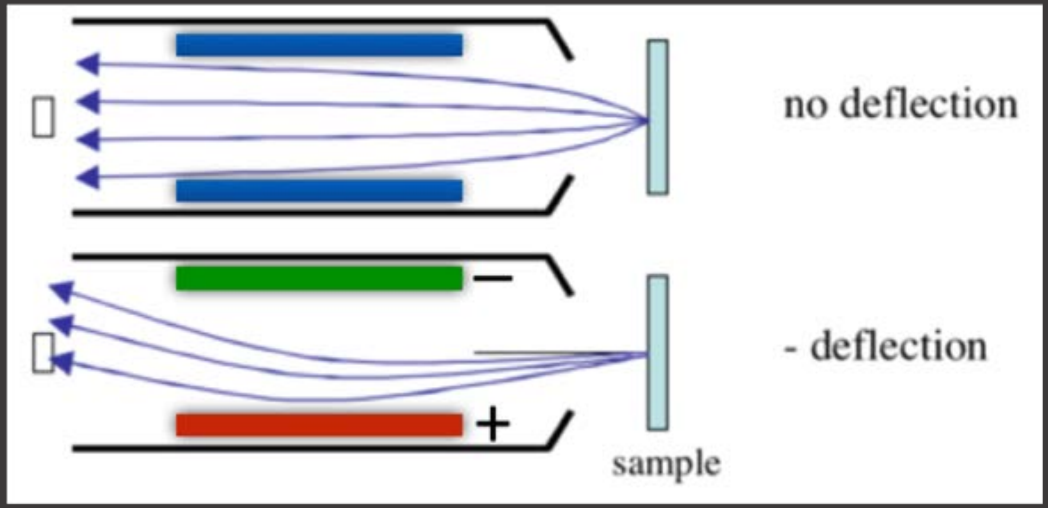
theory

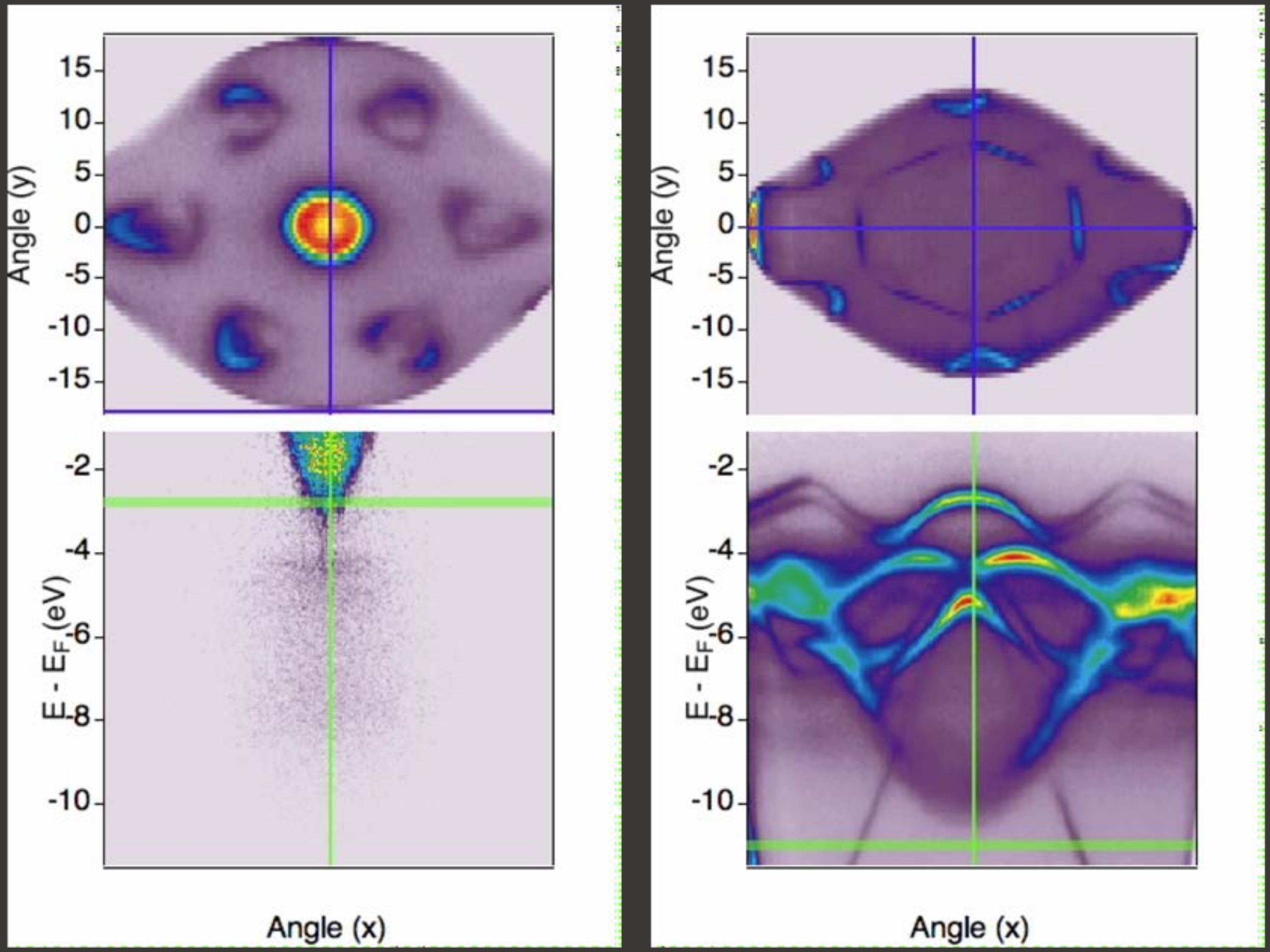
H. Aoki *et al.*, Rev. Mod. Phys. **86**, 779 (2014)
A. F. Kemper *et al.*, Ann. Phys. 1600235 (2017)

Take 1 – μARPES with deflectors





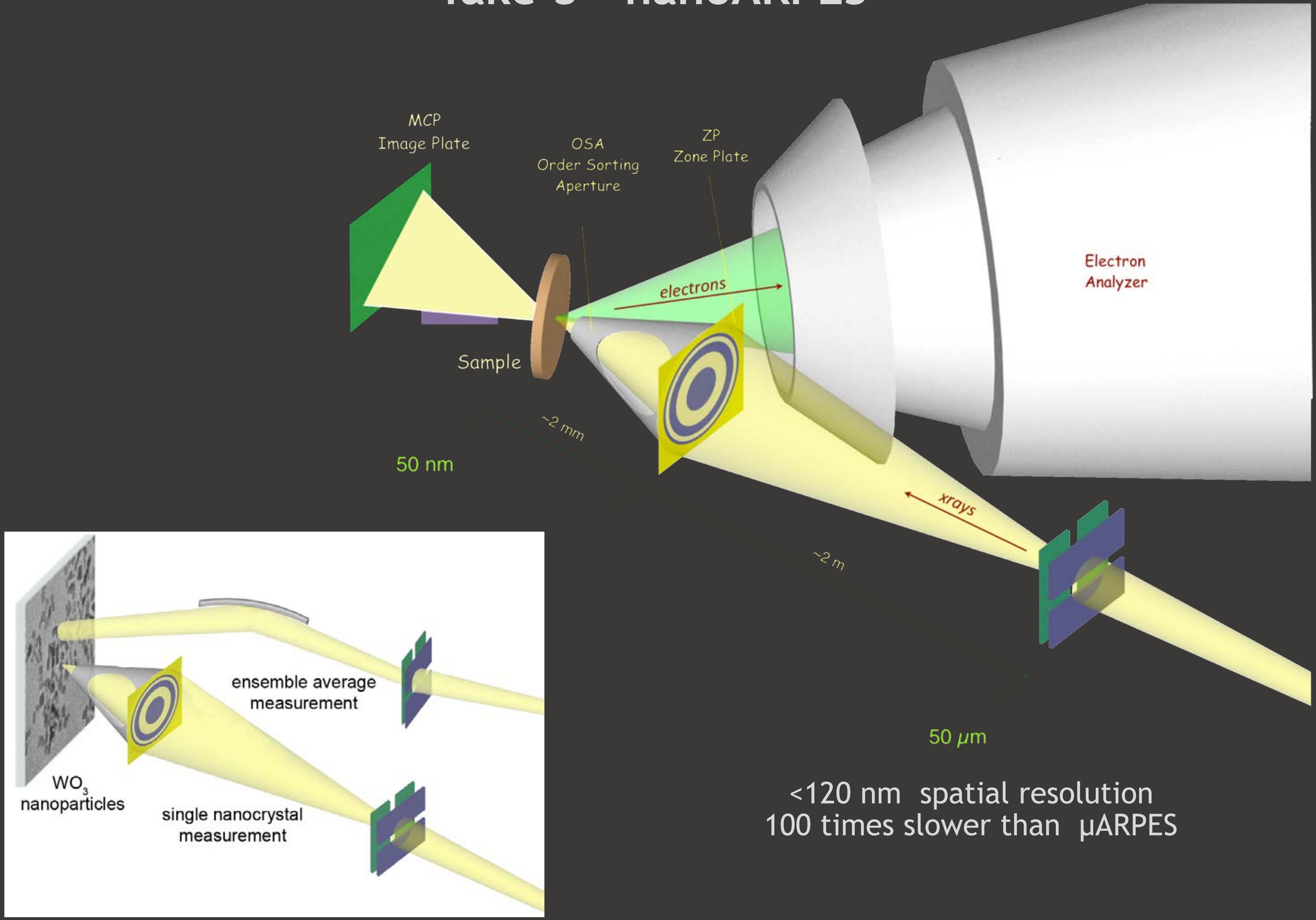




data: J. Katoch and S. Ulstrup

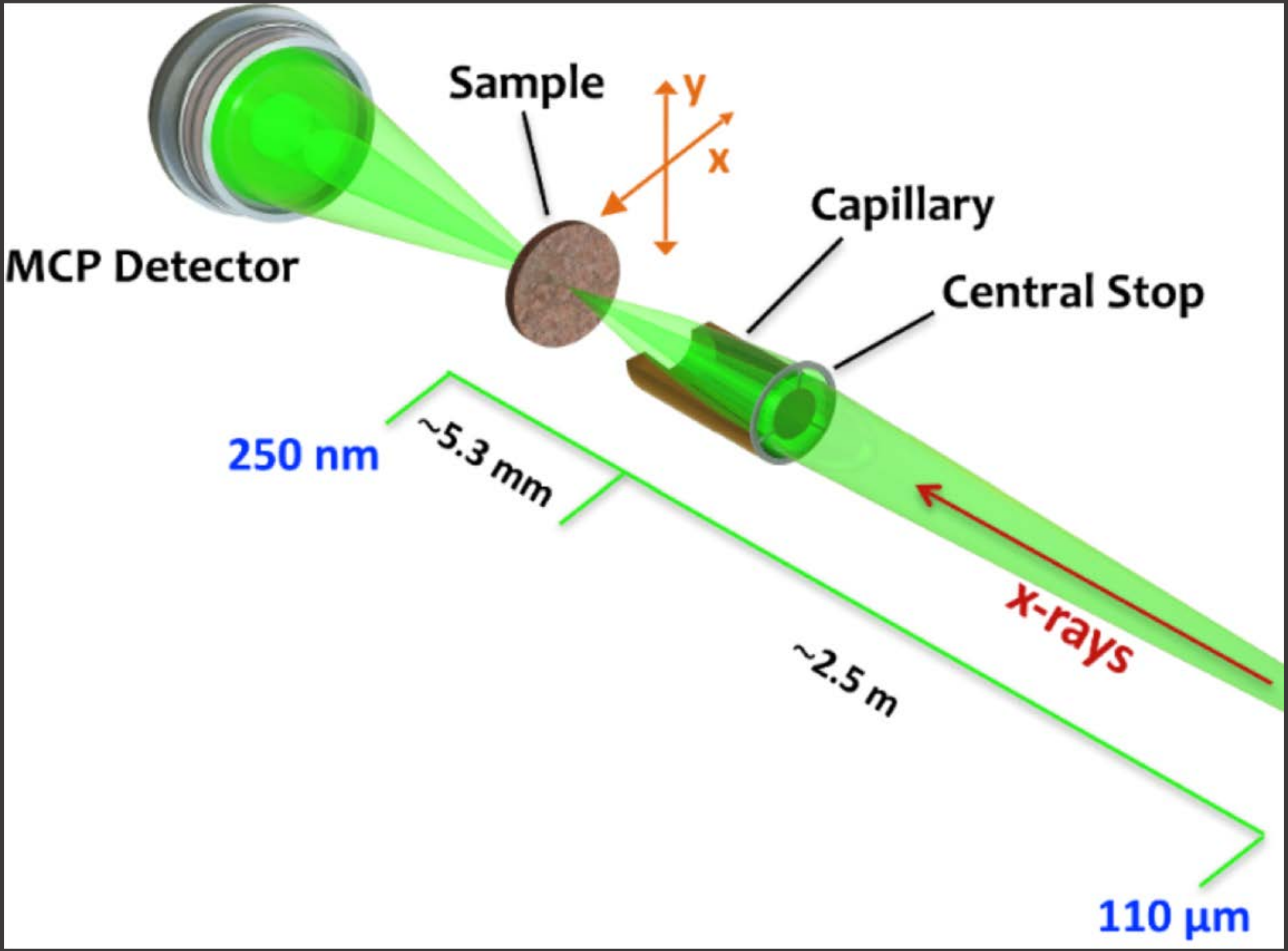
ackn. E. Rotenberg, A. Bostwick, R. Koch

Take 3 - nanoARPES



<120 nm spatial resolution
100 times slower than μARPES

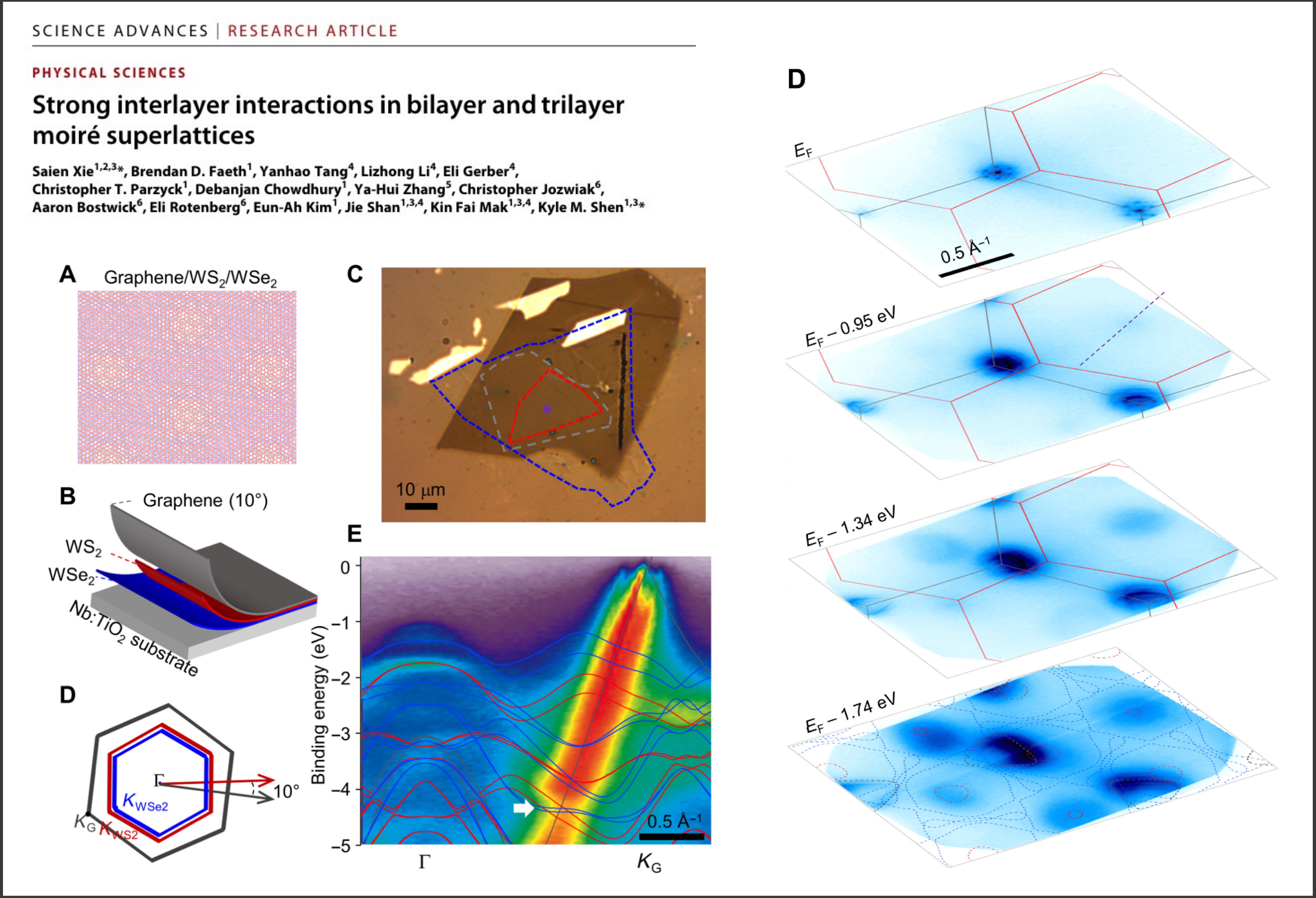
(almost) nanoARPES with KB optics



In collaboration with SIGRAY

R. J. Koch *et al.*, Synchrotron Radiation News **31**, 50 (2018)

Spatially Resolved ARPES : on micro-structures



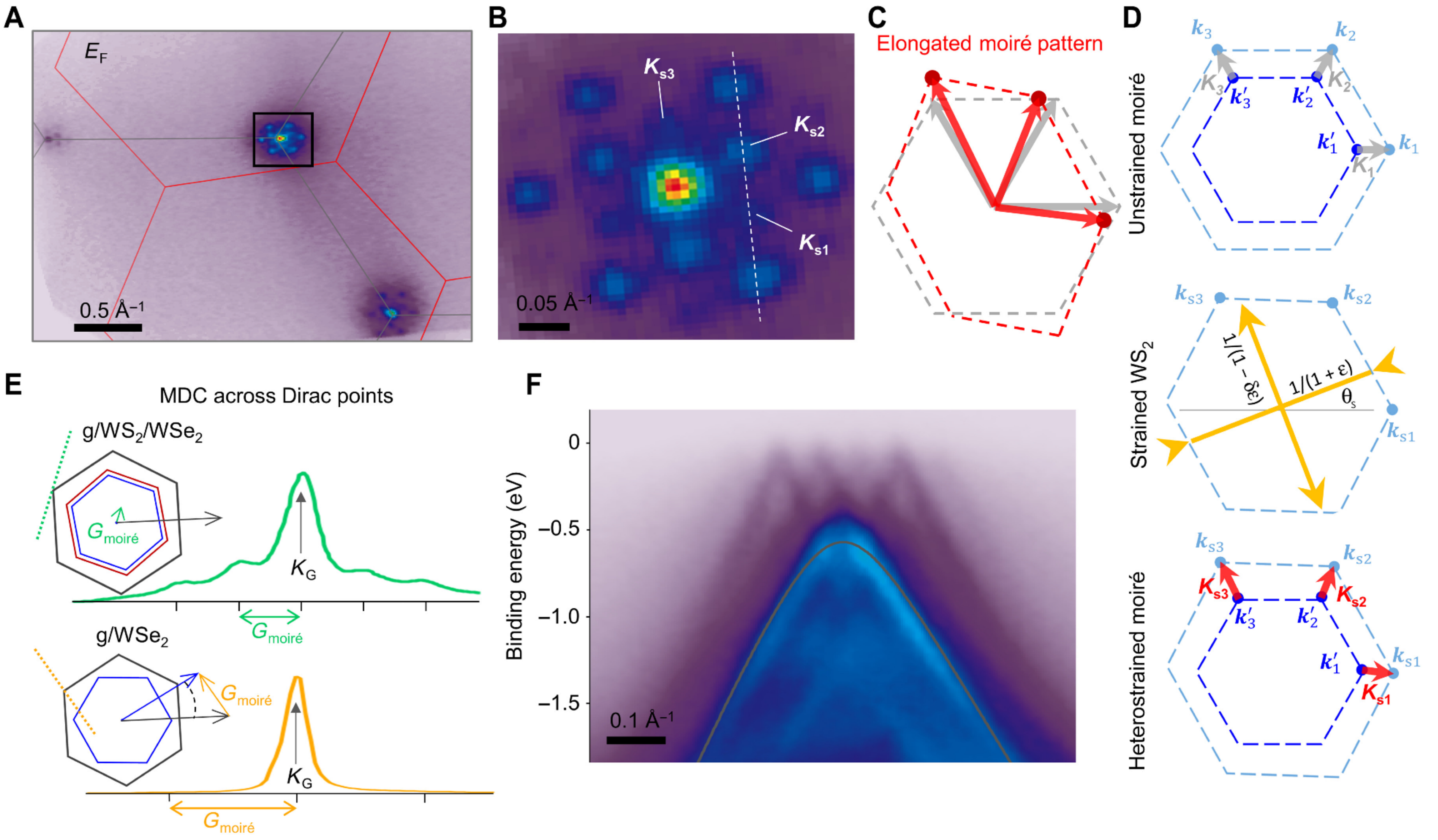
SCIENCE ADVANCES | RESEARCH ARTICLE

PHYSICAL SCIENCES

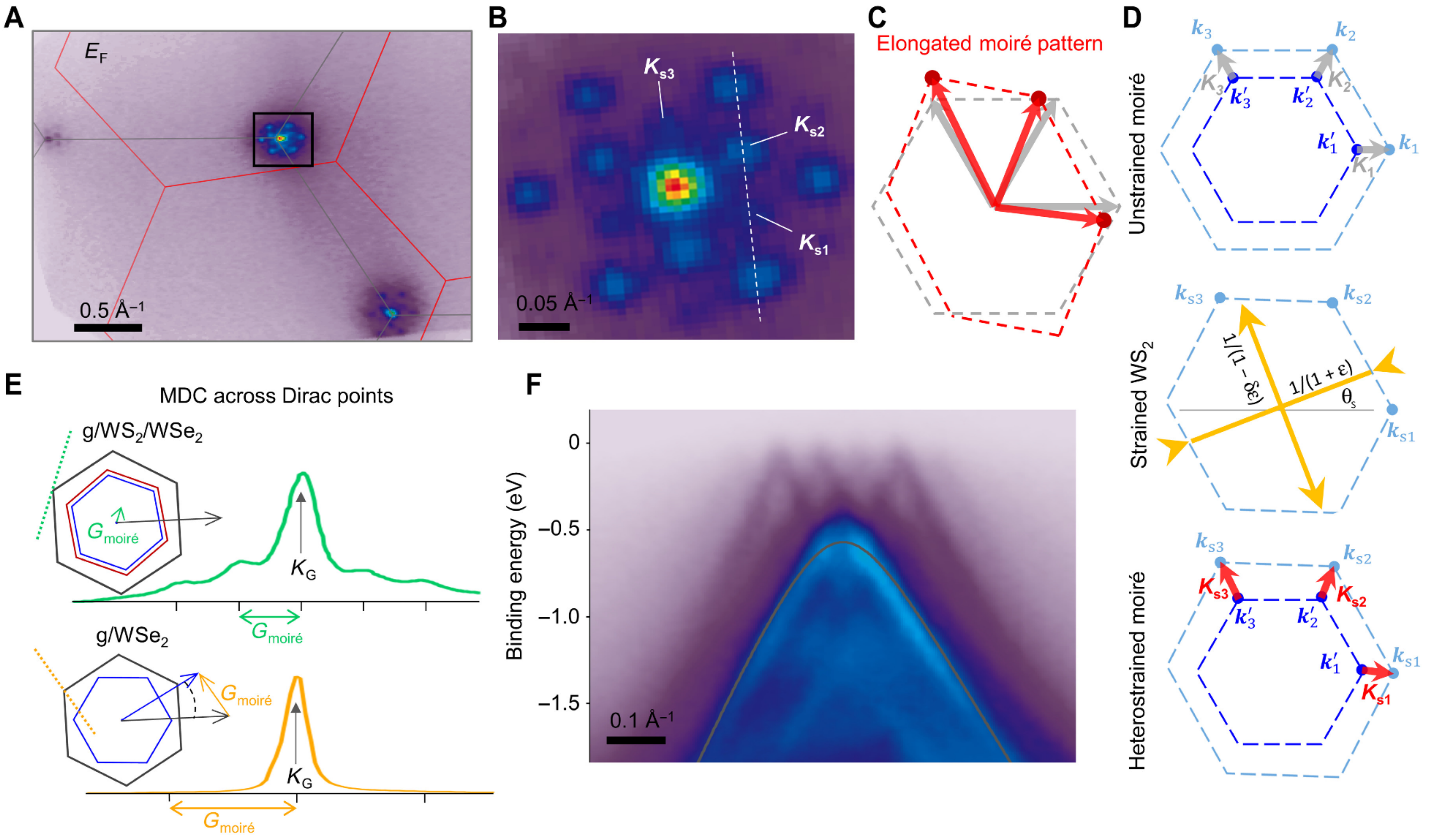
Strong interlayer interactions in bilayer and trilayer moiré superlattices

Saien Xie[1,2,3]*, Brendan D. Faeth[1], Yanhao Tang[4], Lizhong Li[4], Eli Gerber[4], Christopher T. Parzyck[1], Debanjan Chowdhury[1], Ya-Hui Zhang[5], Christopher Jozwiak[6], Aaron Bostwick[6], Eli Rotenberg[6], Eun-Ah Kim[1], Jie Shan[1,3,4], Kin Fai Mak[1,3,4], Kyle M. Shen[1,3]*

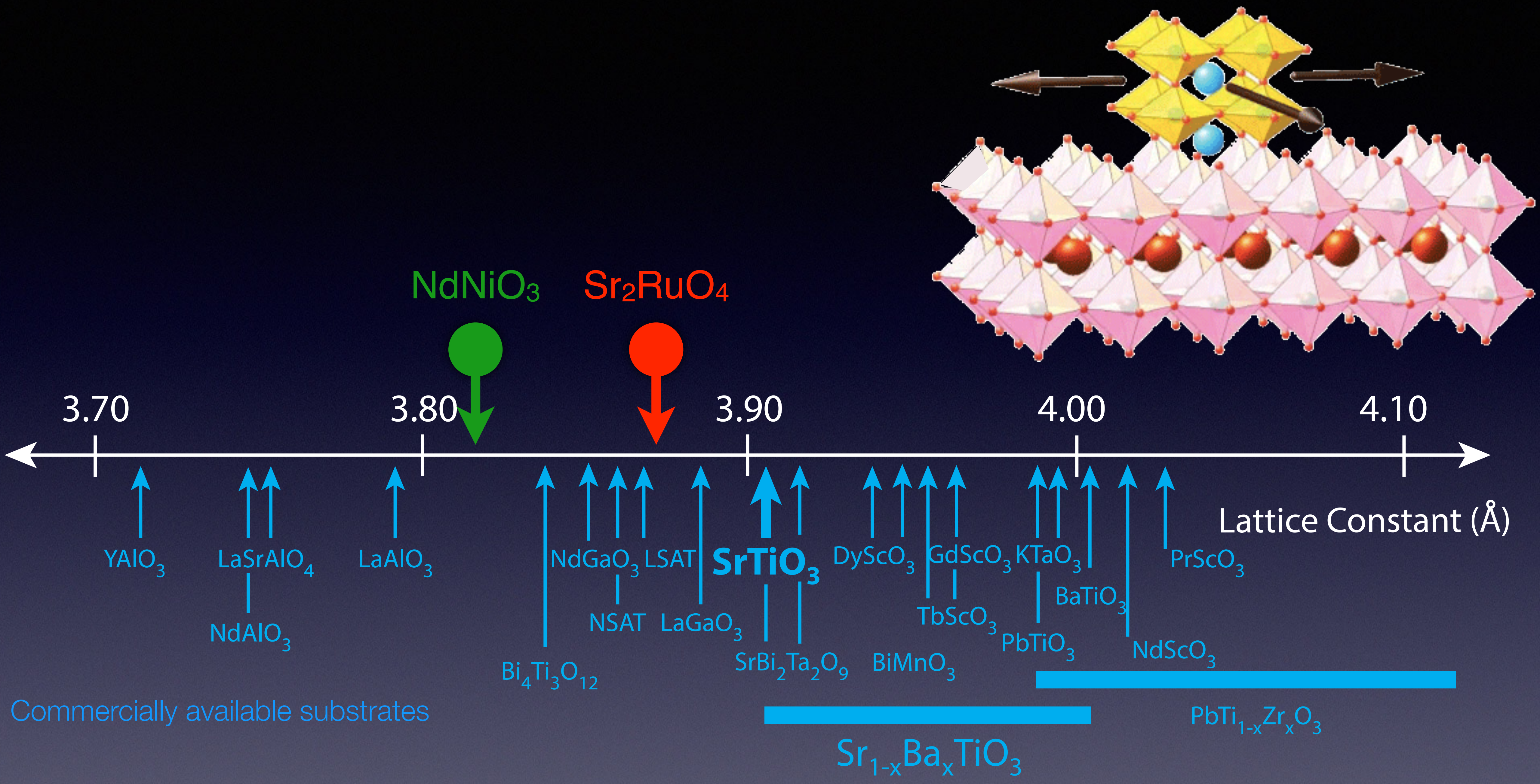
Spatially Resolved ARPES : on micro-structures



Spatially Resolved ARPES : on micro-structures



epitaxial strain as a tuning parameter in quantum material heterostructures



$NdNiO_3$ Sr_2RuO_4

3.70 3.80 3.90 4.00 4.10

Lattice Constant (Å)

$YAlO_3$ $LaSrAlO_4$ $NdAlO_3$ $LaAlO_3$ $Bi_4Ti_3O_{12}$ $NdGaO_3$ LSAT NSAT $LaGaO_3$ **$SrTiO_3$** $SrBi_2Ta_2O_9$ $DyScO_3$ $BiMnO_3$ $GdScO_3$ $TbScO_3$ $KTaO_3$ $PbTiO_3$ $BaTiO_3$ $NdScO_3$ $PrScO_3$

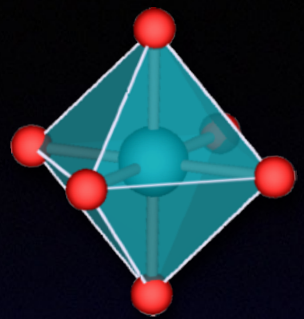
$Sr_{1-x}Ba_xTiO_3$

$PbTi_{1-x}Zr_xO_3$

Commercially available substrates

- 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)

Ruthenate properties are highly tunable with structural changes

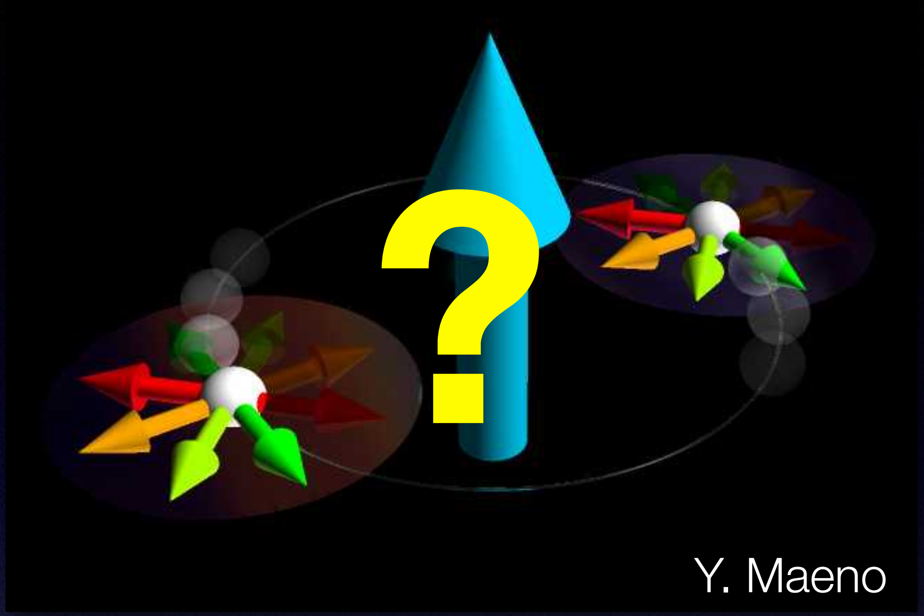


RuO_6 octahedra

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

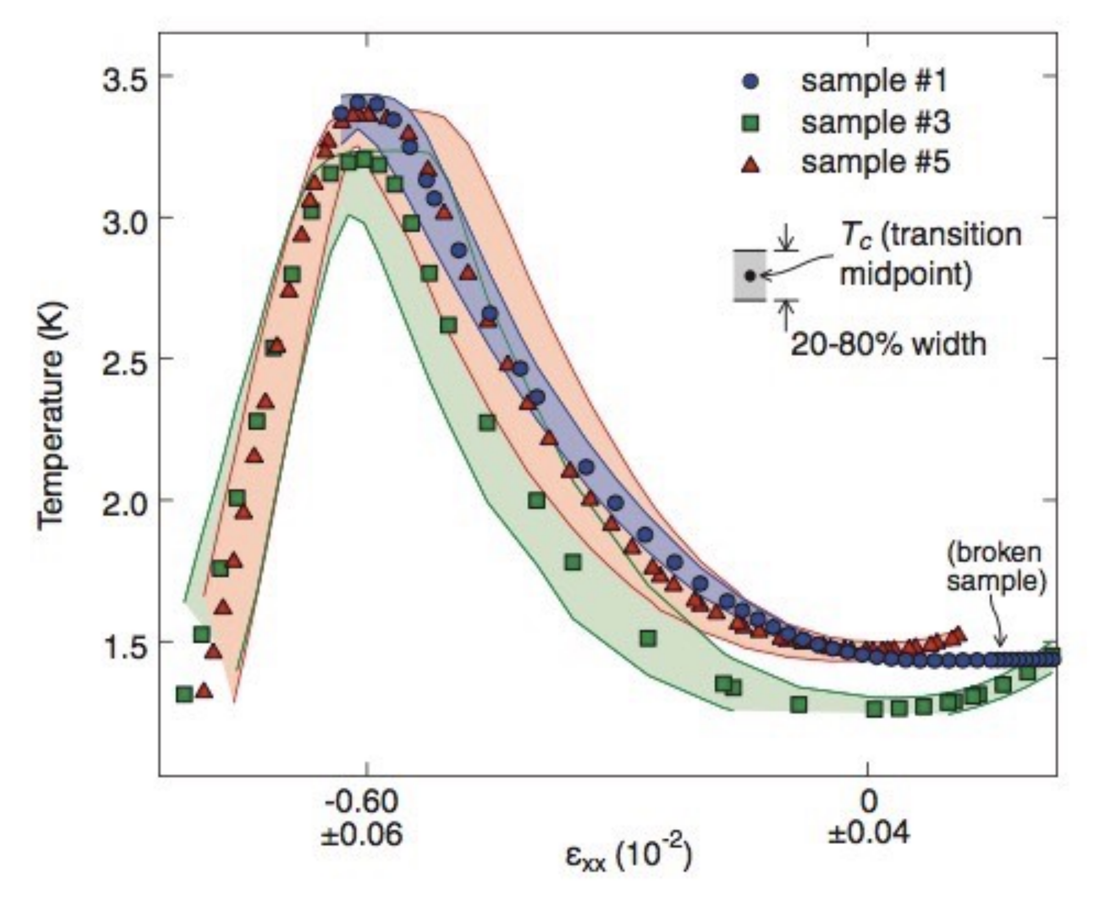
ground states can be tuned from metal, AF insulator, FM metal, exotic SC, simply by changing connectivity of RuO_6 octahedra (without doping)

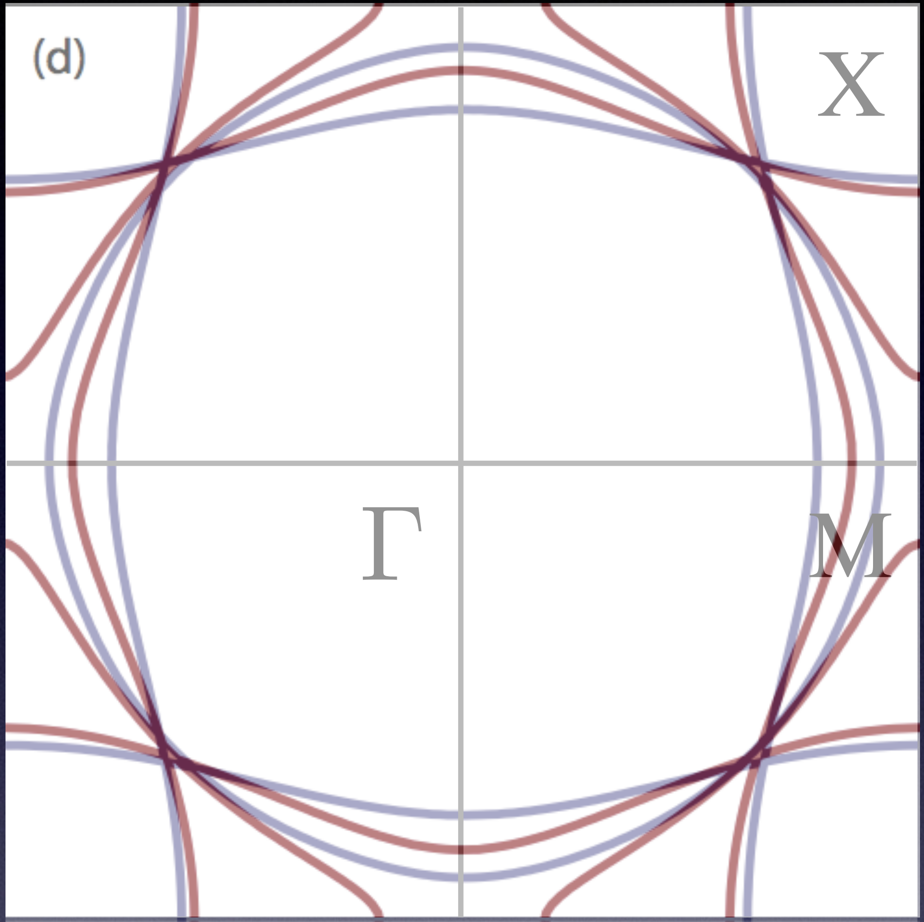


Y. Maeno

- 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

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.

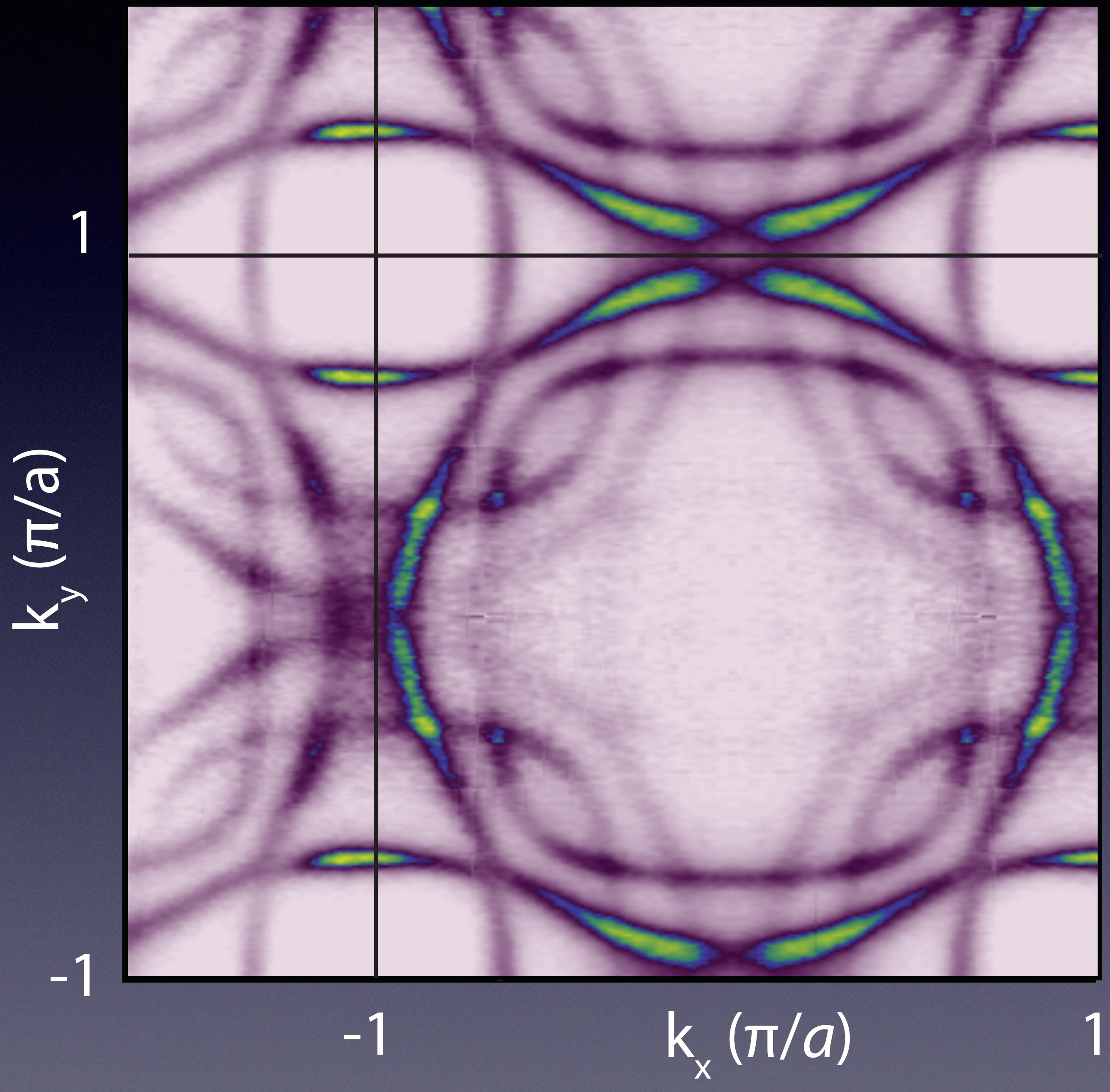
How does electronic structure evolve with epitaxial strain?

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

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%)



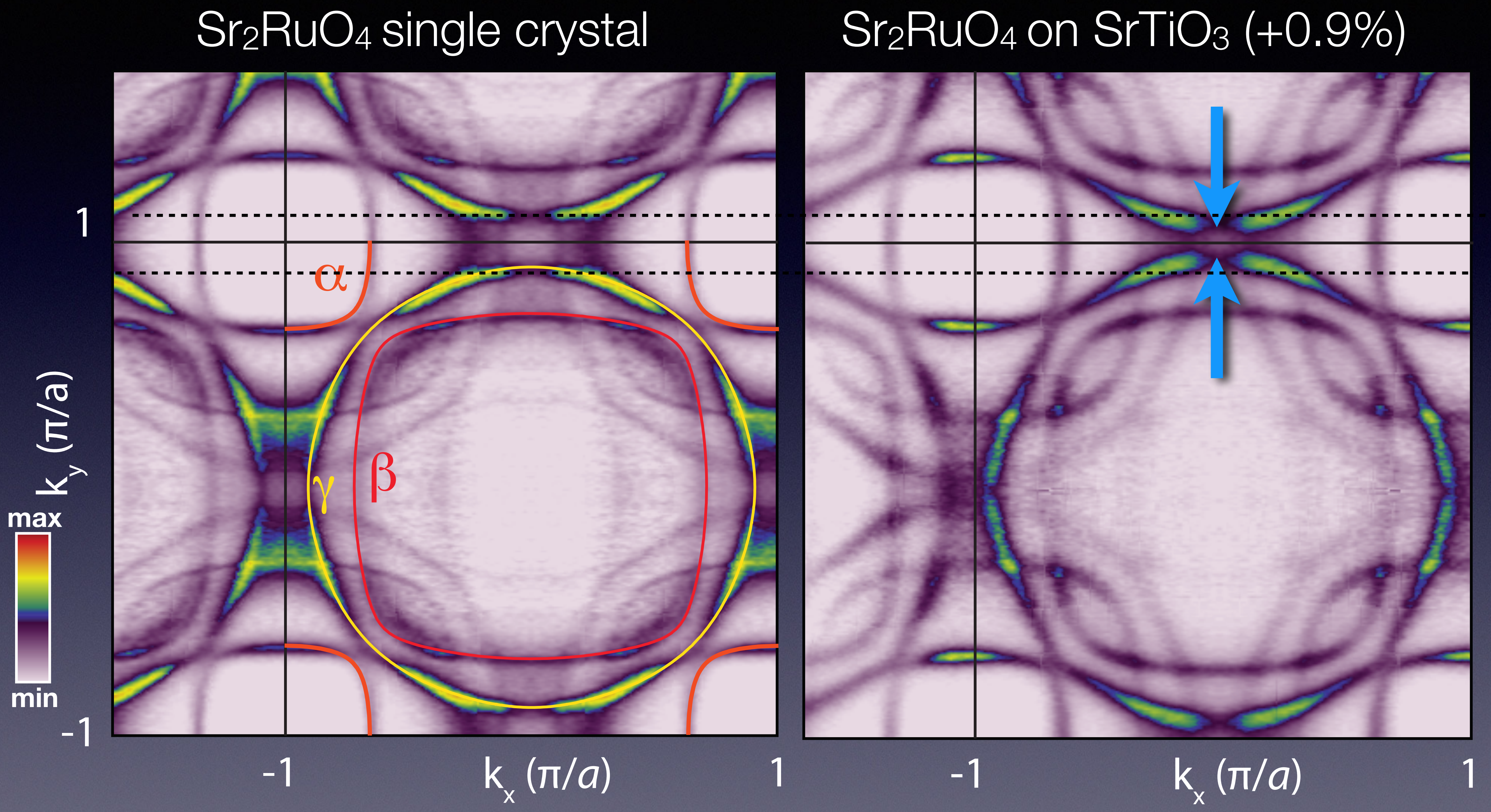
Bulat
Burganov

Carolina
Adamo

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

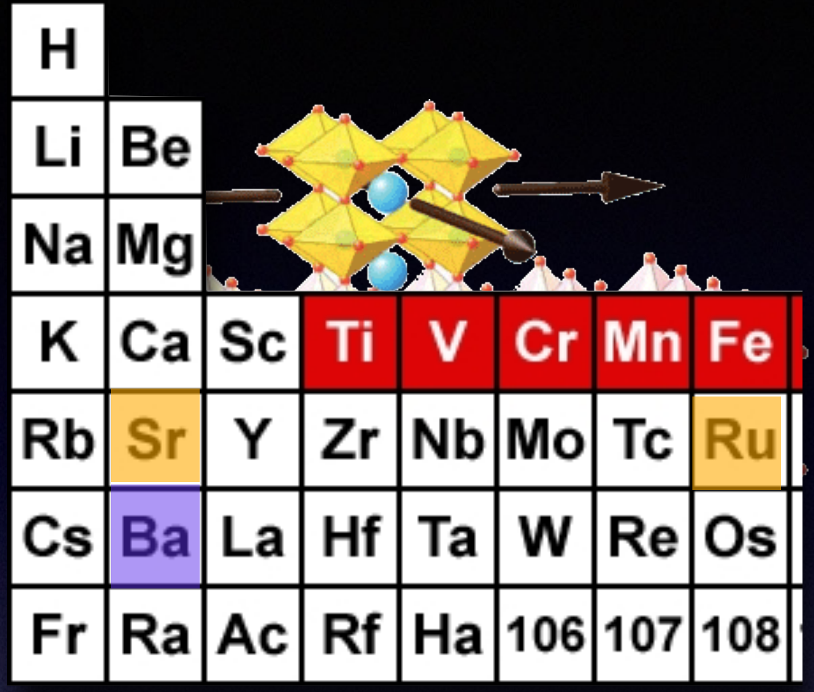
single crystal from A.P. Mackenzie

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

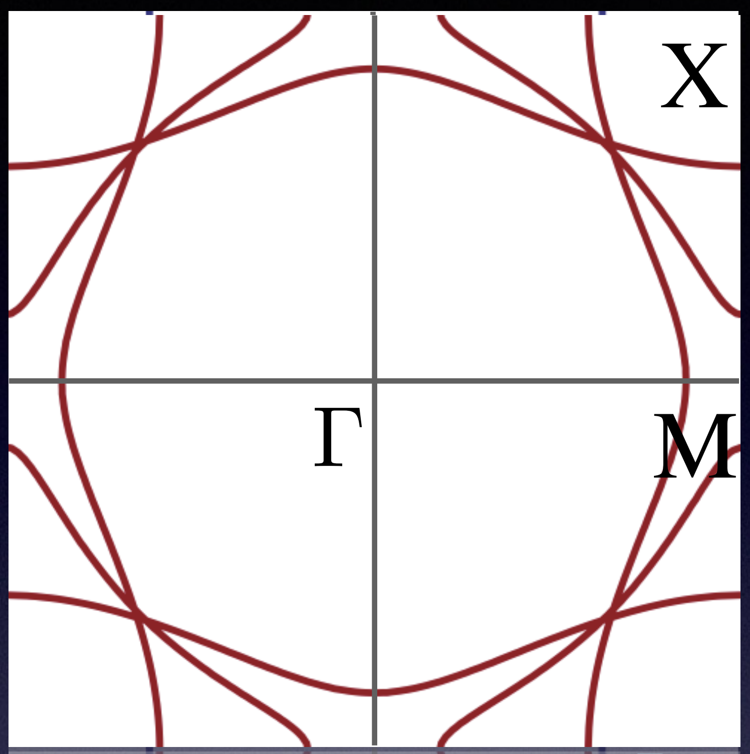


single crystal from A.P. Mackenzie

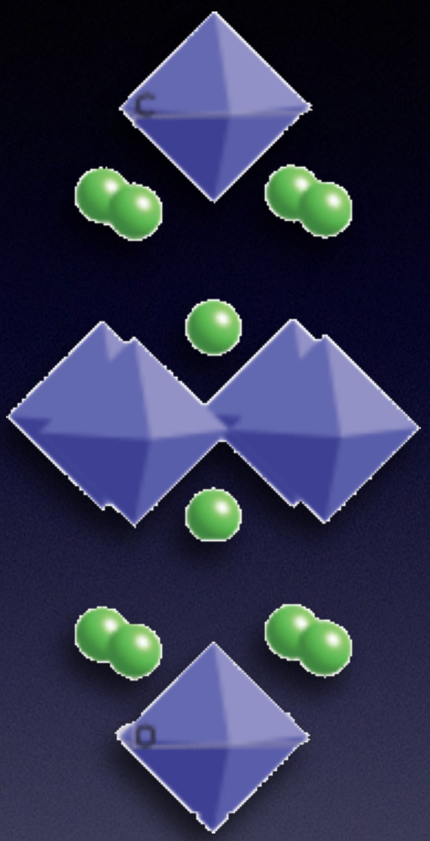
Epitaxial strain to enhance superconductivity in Sr_2RuO_4?



Lifshitz transition



Ba_2RuO_4



bulk

Sr_2RuO_4

$SrTiO_3$

Ba_2RuO_4

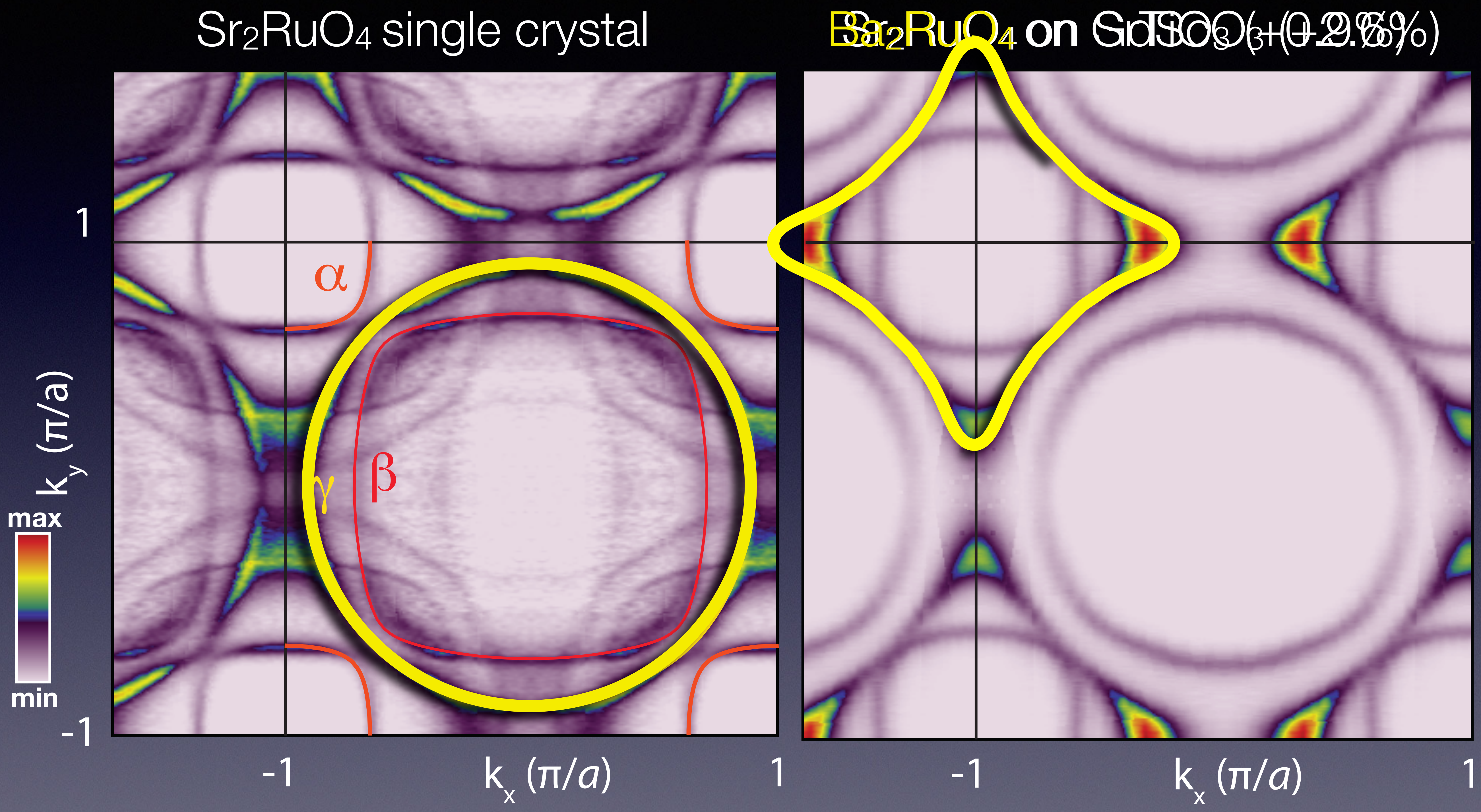
$DyScO_3$ $GdScO_3$

3.860	3.905	3.942	3.960
	(+0.9%)	(+2.1%)	(+2.6%)

in-plane lattice constant (Å)

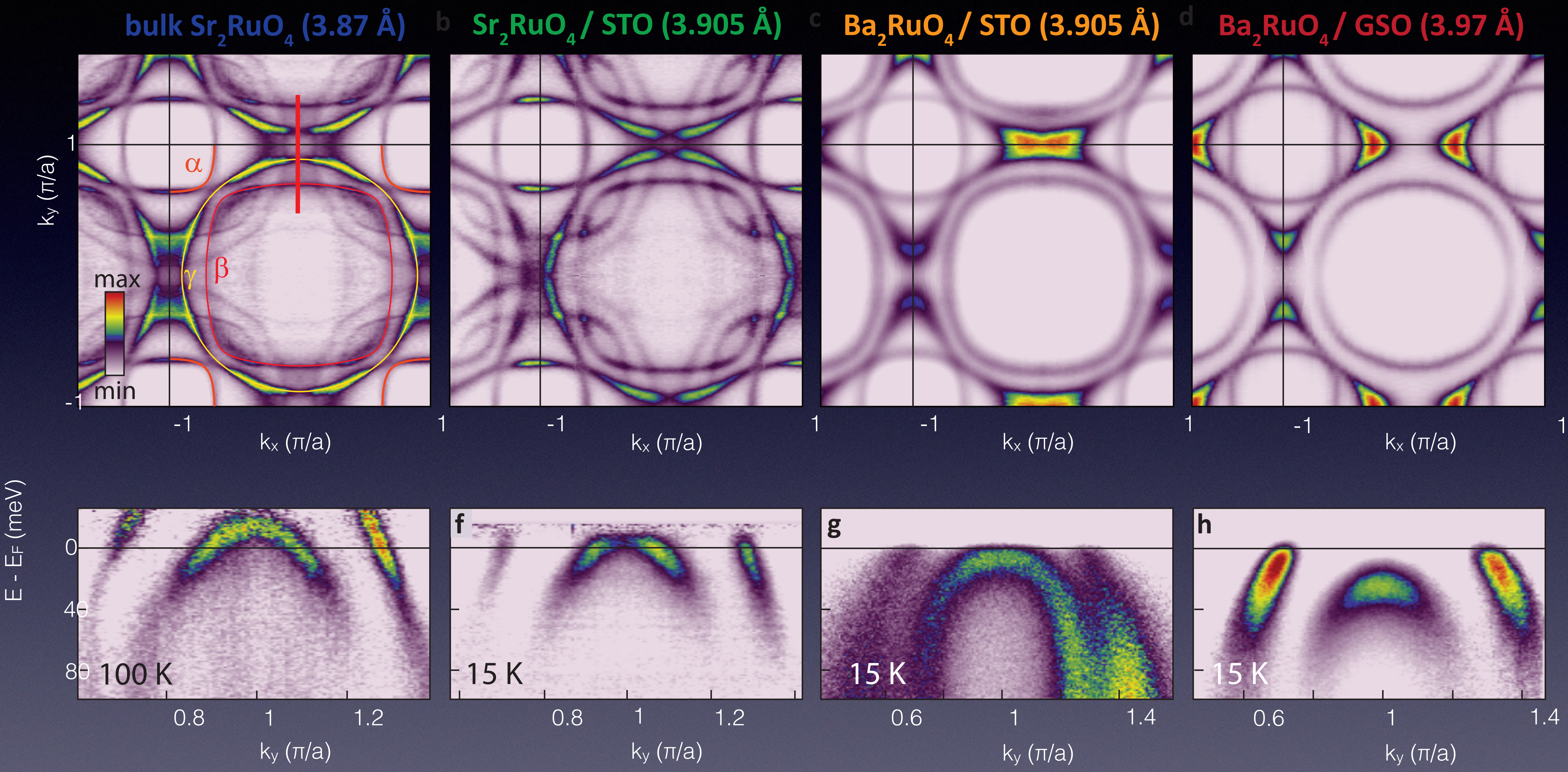
- Ba_2RuO_4 is metastable in bulk but can be epitaxially stabilized

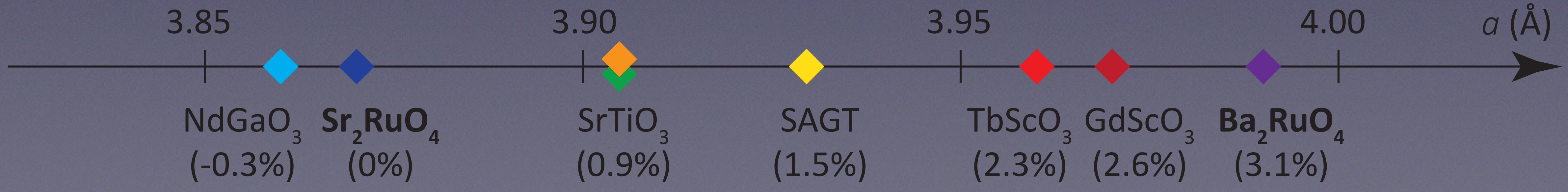
Can tensile strain push the van Hove singularity closer to E_F?



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

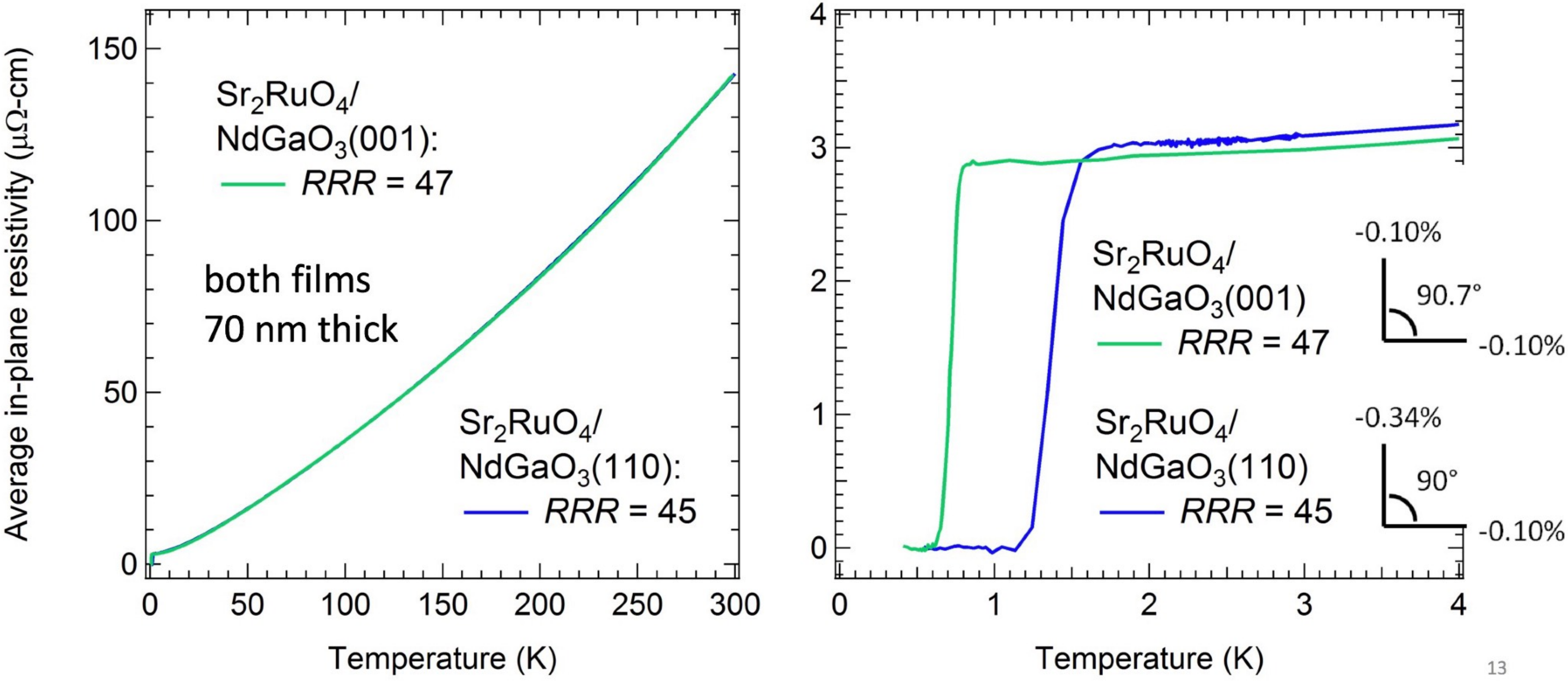
summary of Fermi surface & van Hove singularity evolution with strain





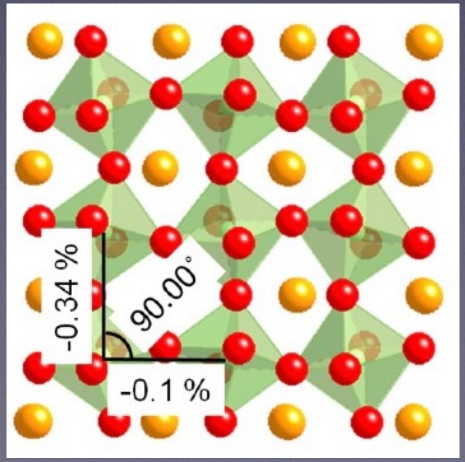
B. Burganov, C. Adamo, et al., *Phys. Rev. Lett.* **116** (2016)

superconductivity depends on orientation of $NdGaO_3$ substrate



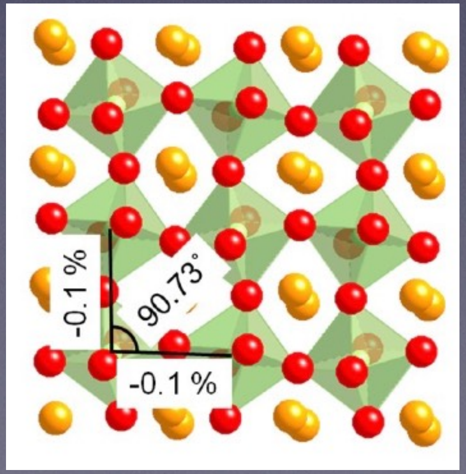
$NdGaO_3$ (110)

Pbnm



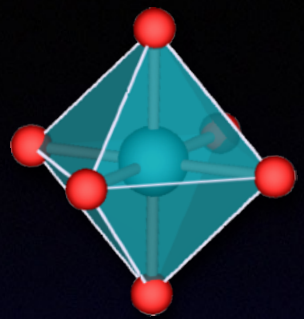
$NdGaO_3$ (001)

Pbnm



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

Ruthenate properties are highly tunable with structural changes



RuO_6 octahedra

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

Compound	Dimensionality	Octahedral Connectivity	Properties
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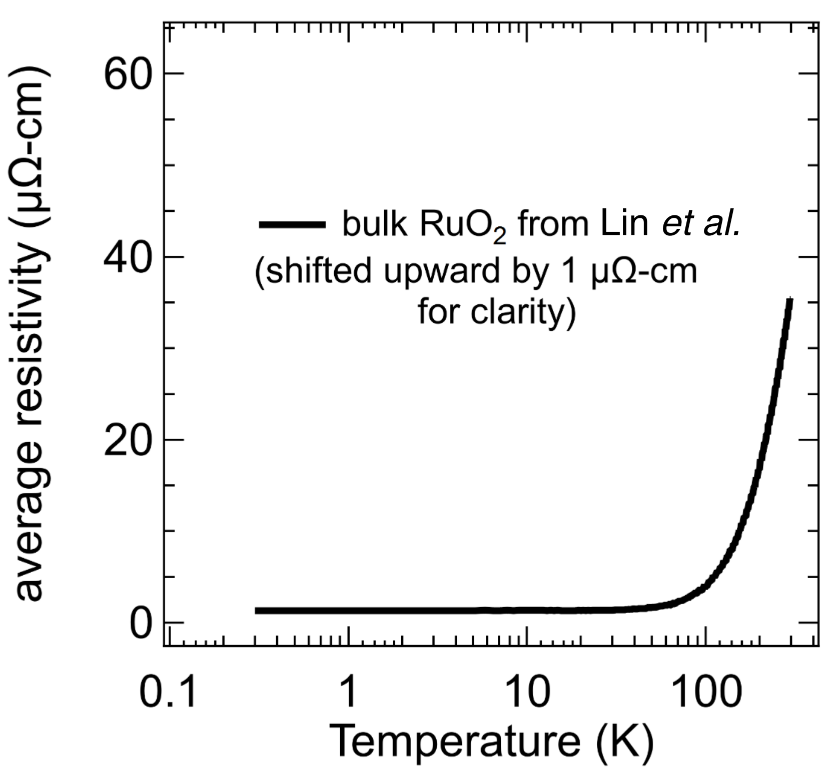
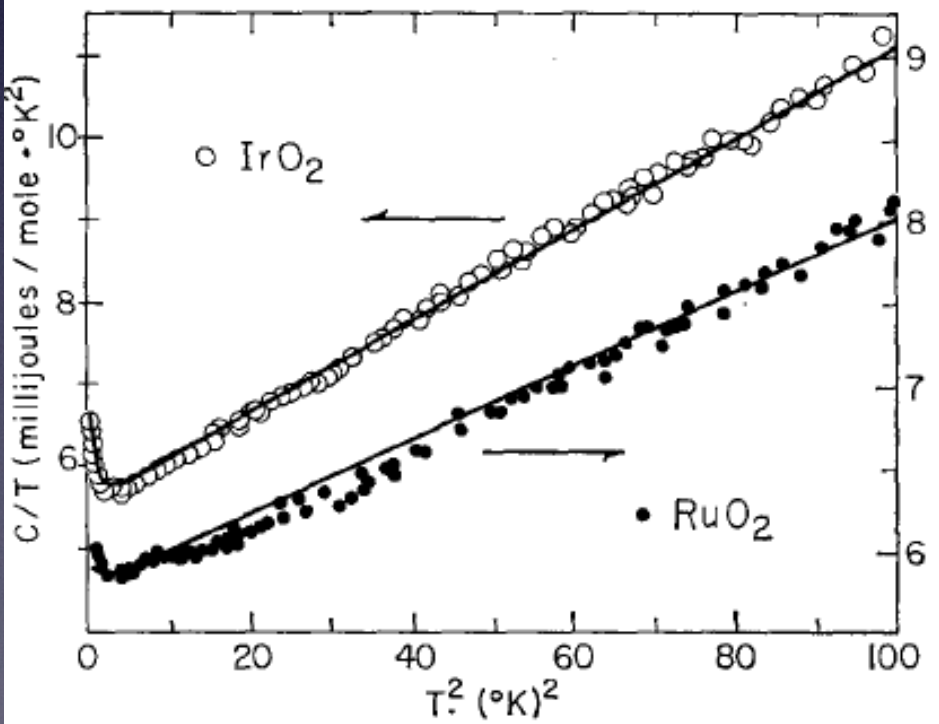
$SrRuO_3$	**3D**	**CORNER**	FM Metal
RuO_2	**3D**	**EDGE & CORNER**	Metal

- $\gamma \sim 6$ mJ / mol K^2 (1.4x DFT)
- $\rho_0 \sim 0.3$ $\mu\Omega$-cm
- modestly correlated Fermi liquid

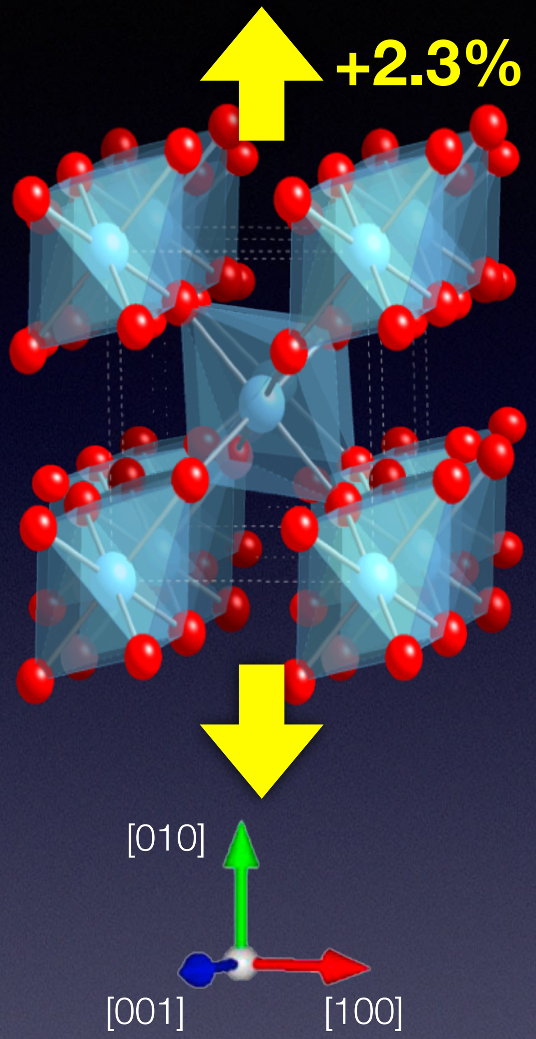




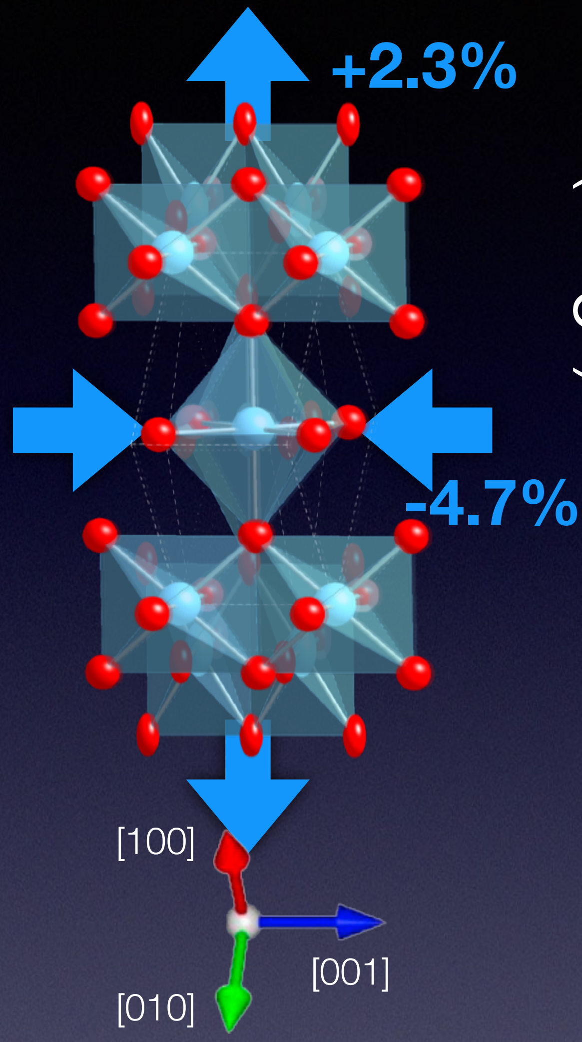
J. J. Lin *et al.*, J. Phys.: Condens. Matter 16, 8035 (2004)
W. D. Ryden et al., Physics Letters A 26, 209 (1968)
B. C. Passenheim & D. C. McCollum, J. Chem. Phys. 51, 320 (1969)
W. D. Ryden & A. Lawson, J. Chem. Phys. 52, 6058 (1970)

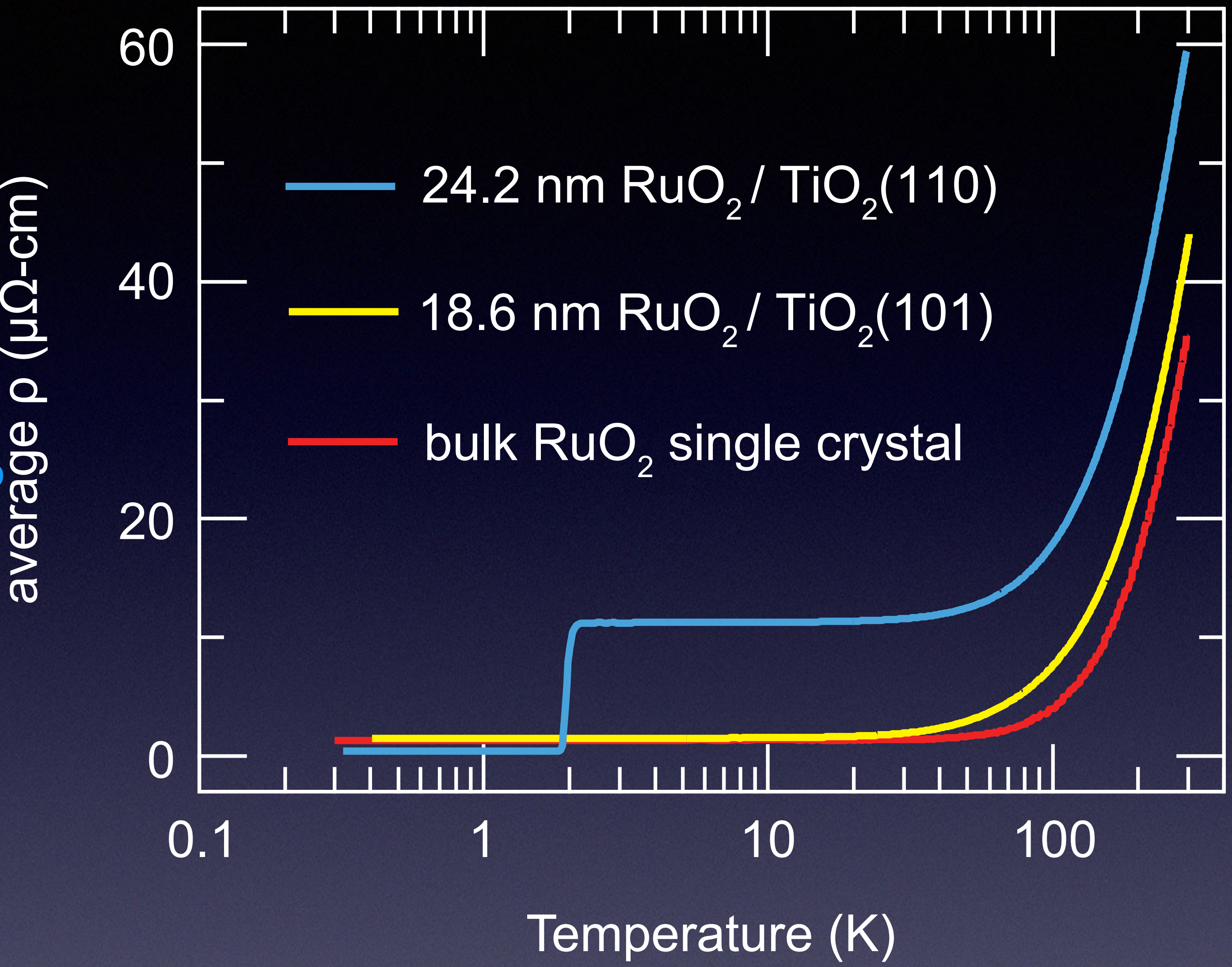
Large, anisotropic epitaxial strain induces superconductivity in RuO_2

RuO_2 on TiO_2 (101)



RuO_2 on TiO_2 (110)

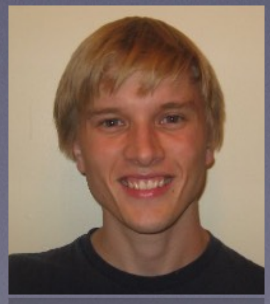




- Superconductivity observed for highly strained RuO_2 films grown on (110) TiO_2 substrates
- Films grown along the (101) direction do not exhibit any evidence for SC, nor do “relaxed”, thick, bulk-like films of RuO_2 / TiO_2 (110)

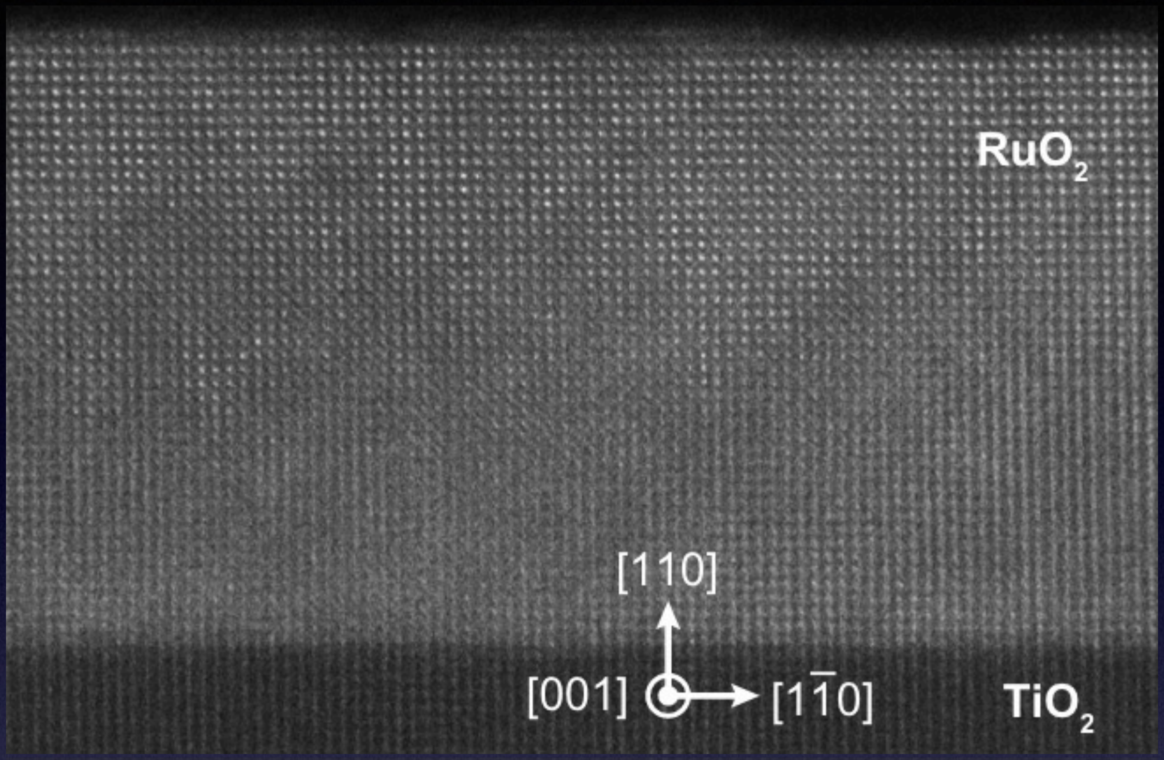
Hanjong
Paik

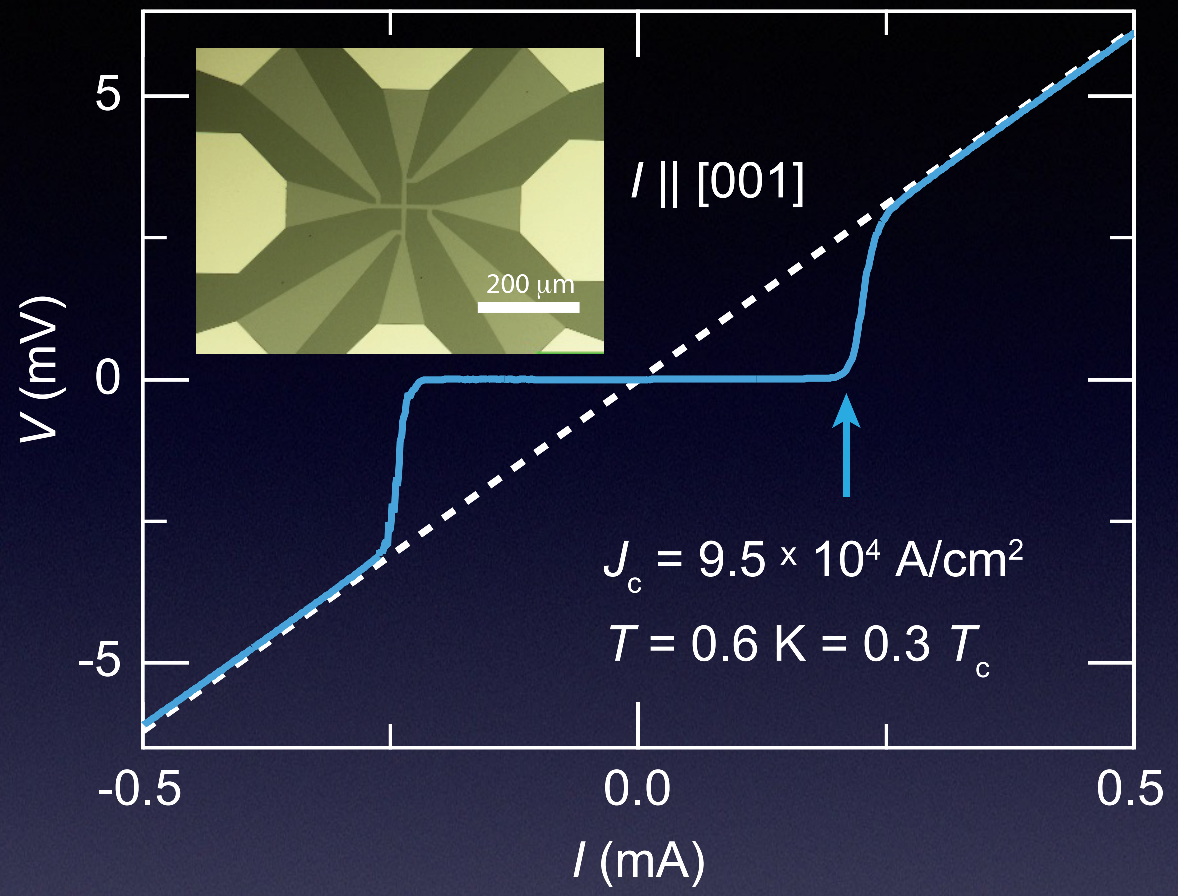
Jacob
Ruf



J.P. Ruf, H. Paik *et al.*, *Nature Comm.* 12, 59 (2021)

Superconductivity in RuO$_2$ / TiO$_2$ (110) is of an intrinsic nature



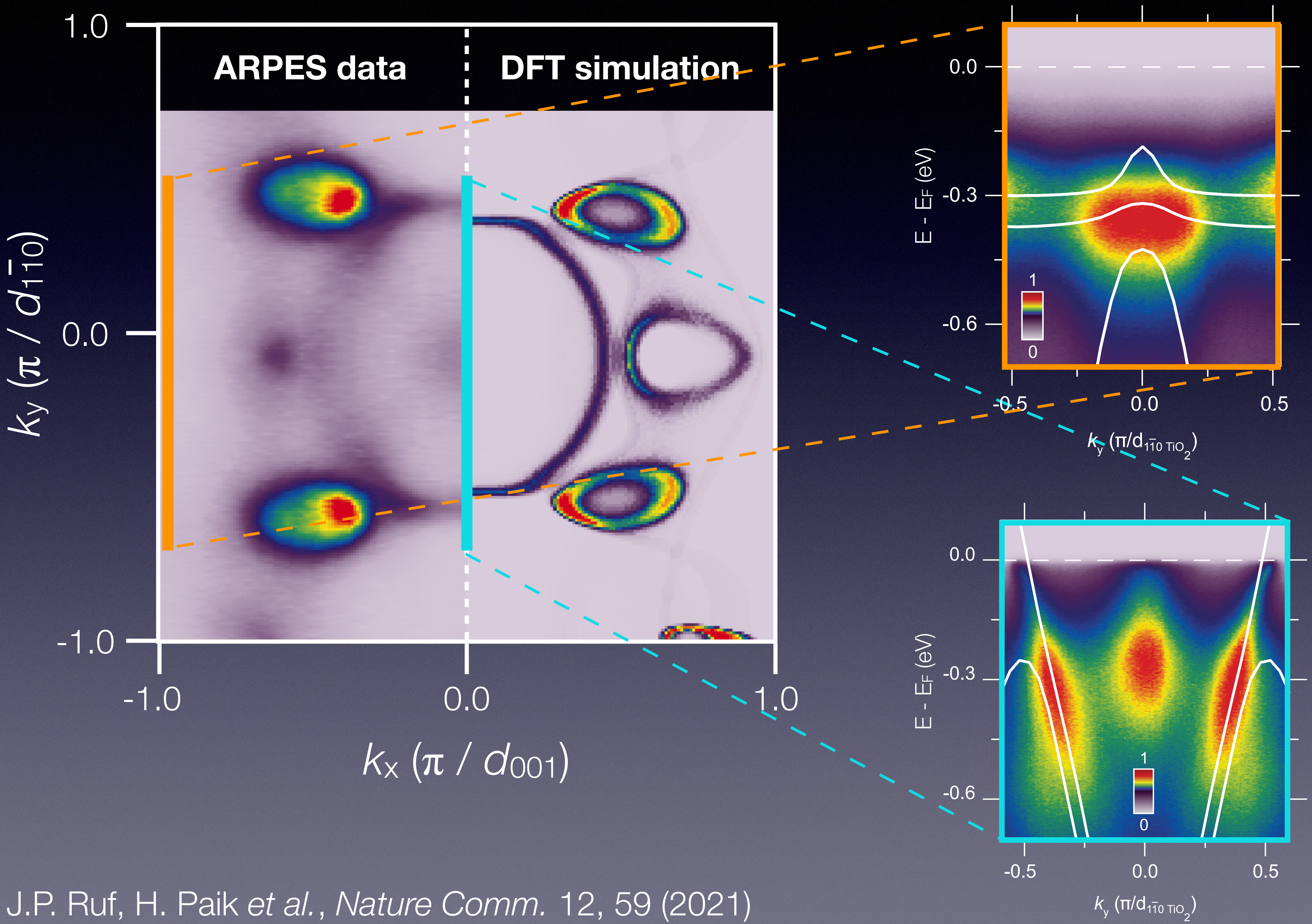


$J_c = 9.5 \times 10^4$ A/cm^2

$T = 0.6$ K $= 0.3\ T_c$

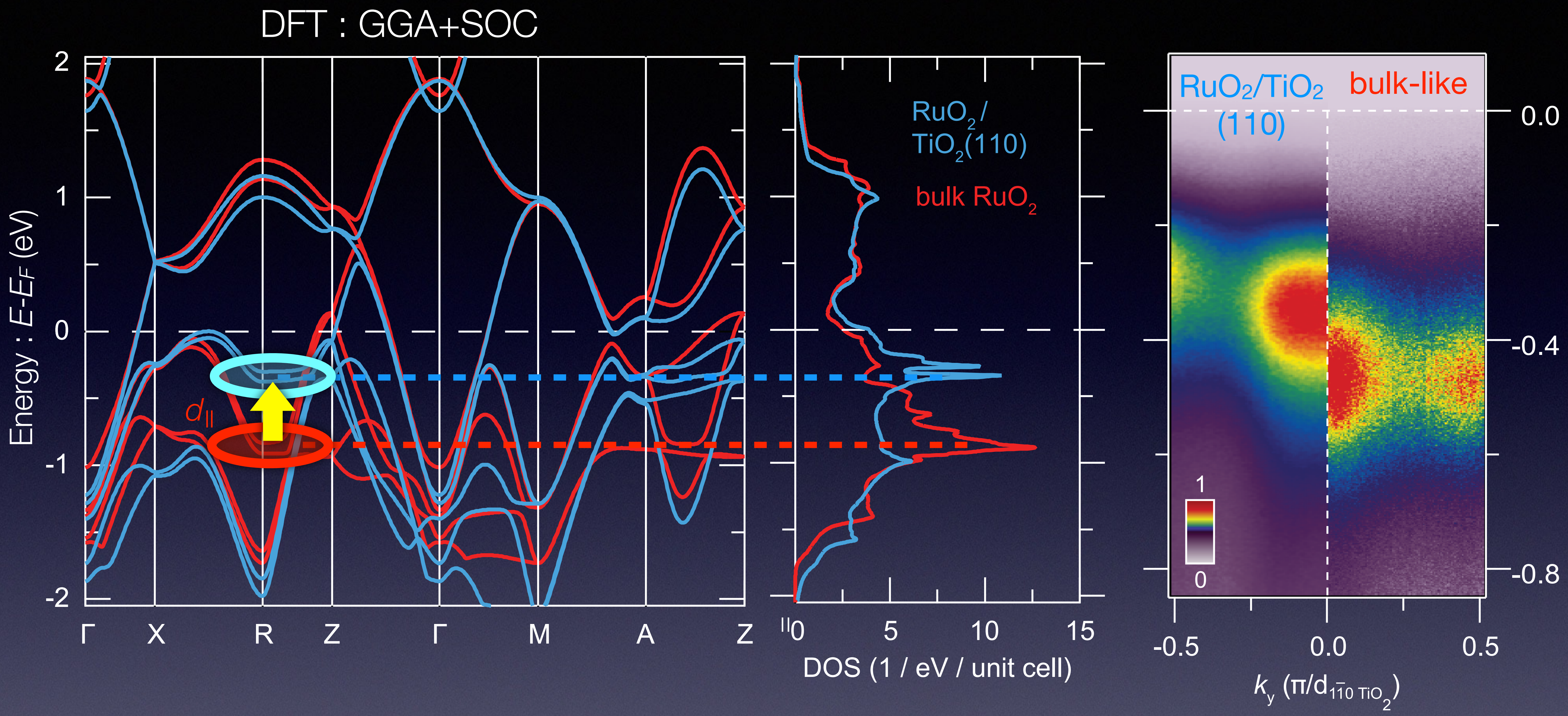
- Highly crystalline samples (TEM, x-ray) rules out possibility of secondary / filamentary impurity phases
- Large J_c's (critical current densities) on patterned devices indicate superconductivity is inherent to the entire RuO$_2$ film, not just the interface(s)

ARPES measurements of electronic structure agree well with DFT



J.P. Ruf, H. Paik *et al.*, *Nature Comm.* 12, 59 (2021)

Epitaxial strain significantly increases DOS near E_F



- $d_{||}$ "flat bands" (R-Z) shifted upwards by anisotropic strain, increasing DOS at E_F
- large, anisotropic strain dramatically affects electronic structure; Migdal-Eliashberg calculations also suggest that calculated T_c should be increased by at least 20x
- first-known "transmutation" of material not known to be SC into a superconductor by strain
- nature of superconductivity still a matter of active investigation. What about larger strains?