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National Synchrotron Radiation Research Center

Angle-resolved photoemission spectroscopy of quantum materials

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Outline

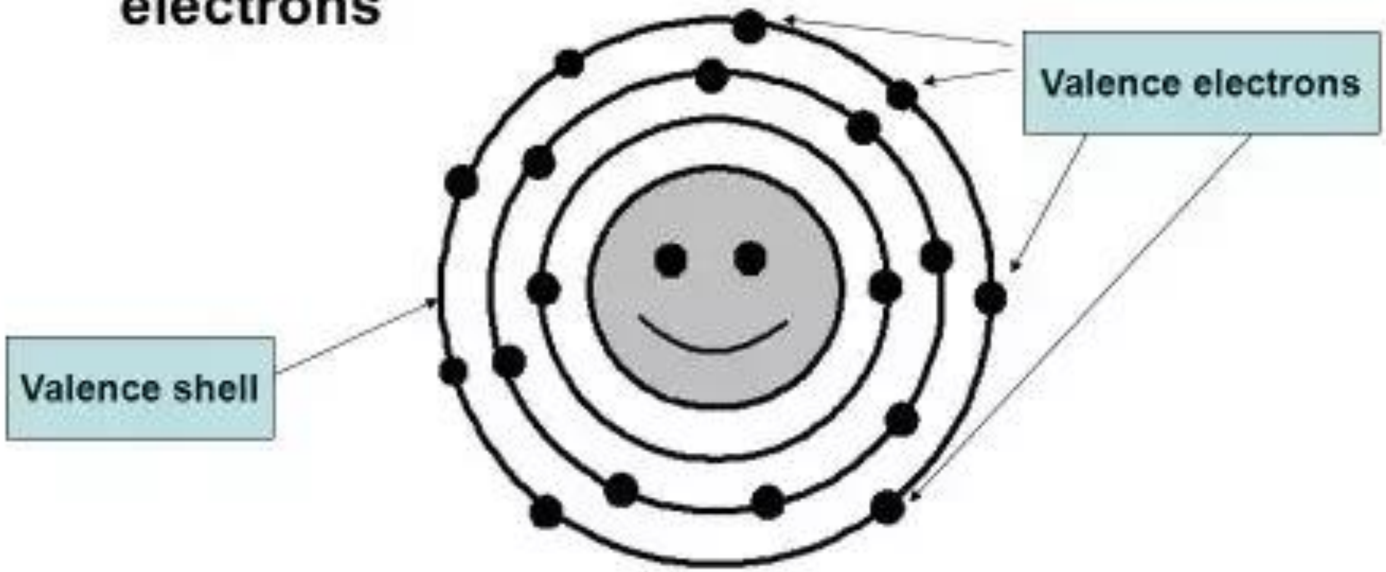
- Introduction to angle-resolved photoemission spectroscopy (ARPES)
  - *The concept of energy band.*
  - *The principle of ARPES.*
  - *Scientific case : cuprates, graphene, TMDs and topological insulators.*
- Probe the electronic structure of emergent materials
  - *Scientific opportunities in nanoARPES.*
  - *The challenge and design concept of nanoARPES.*
  - *Beamline and end station design.*
- Summary

Formation of Bands

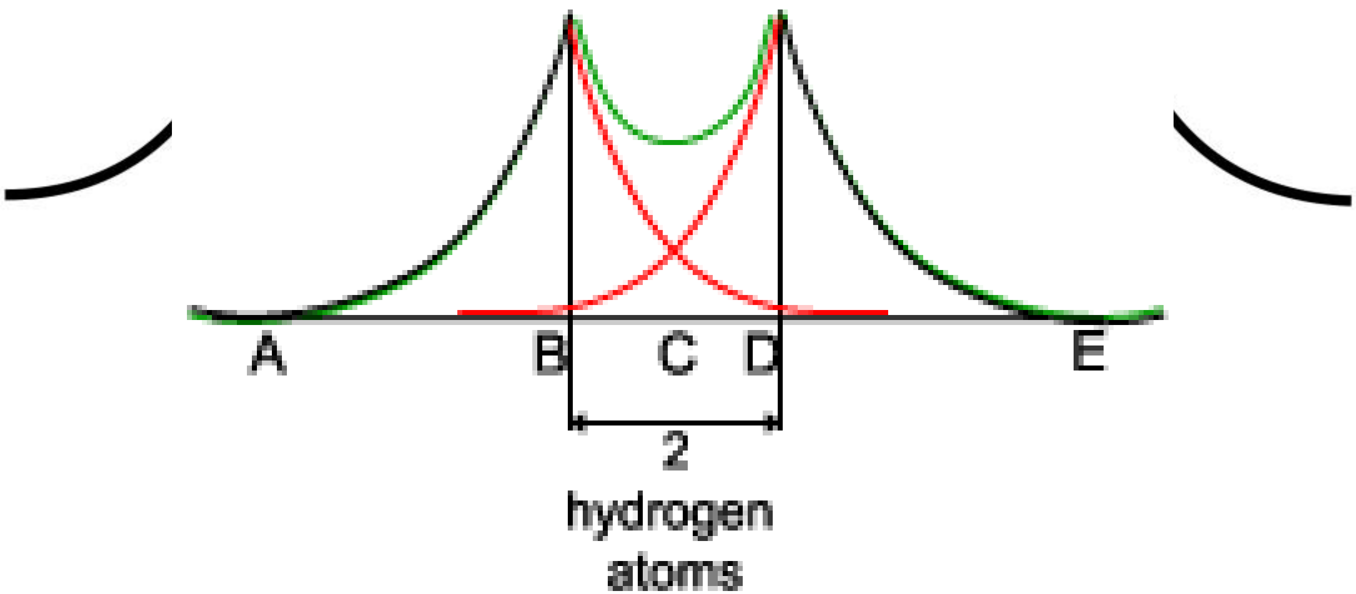
Valence Electrons

– The outer shell is called the **valence shell.**

– Electrons in the outer shell are called **valence electrons**

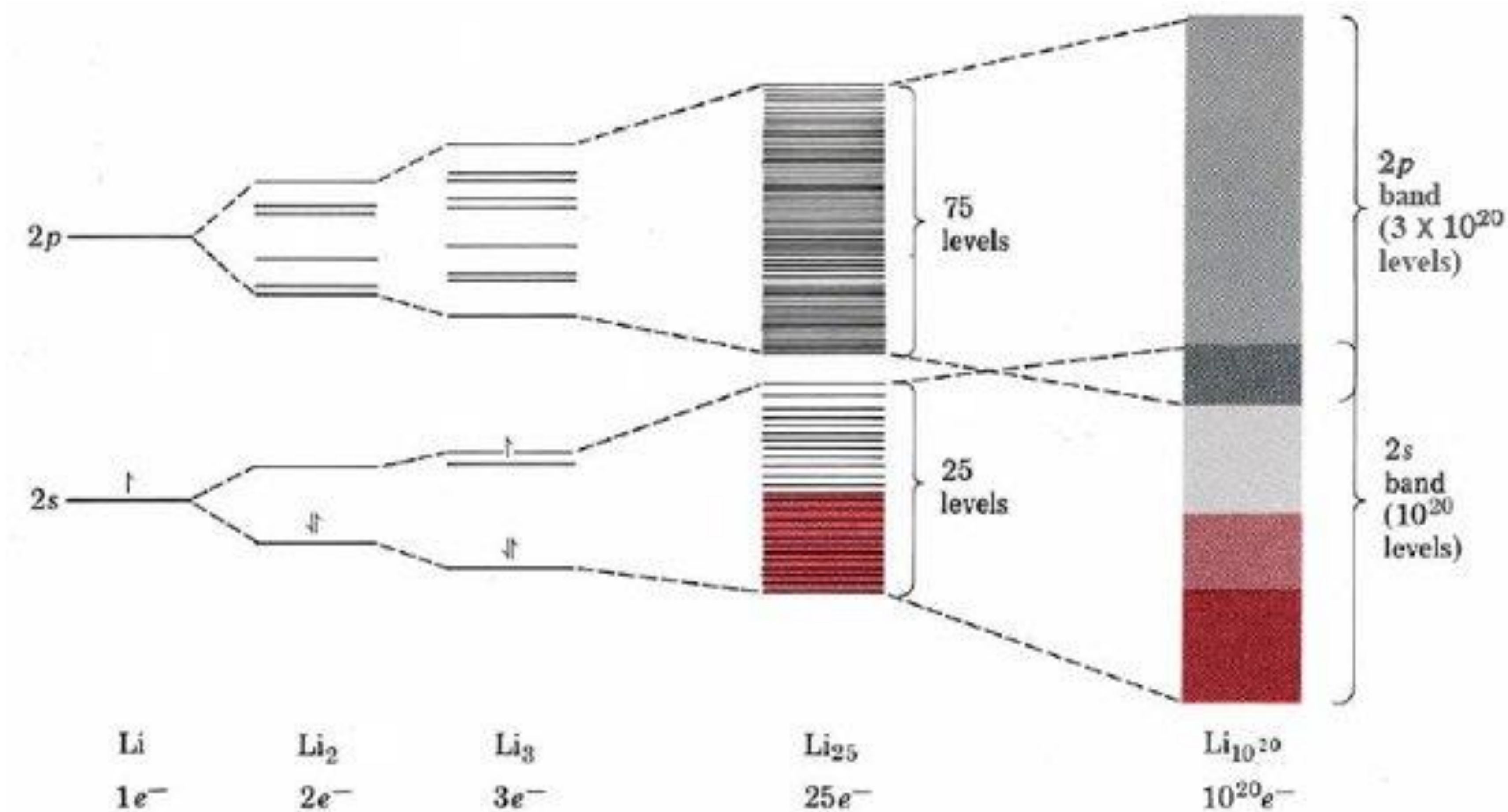


Concept of forming energy bands



Formation of a molecular orbital

Formation of Bands



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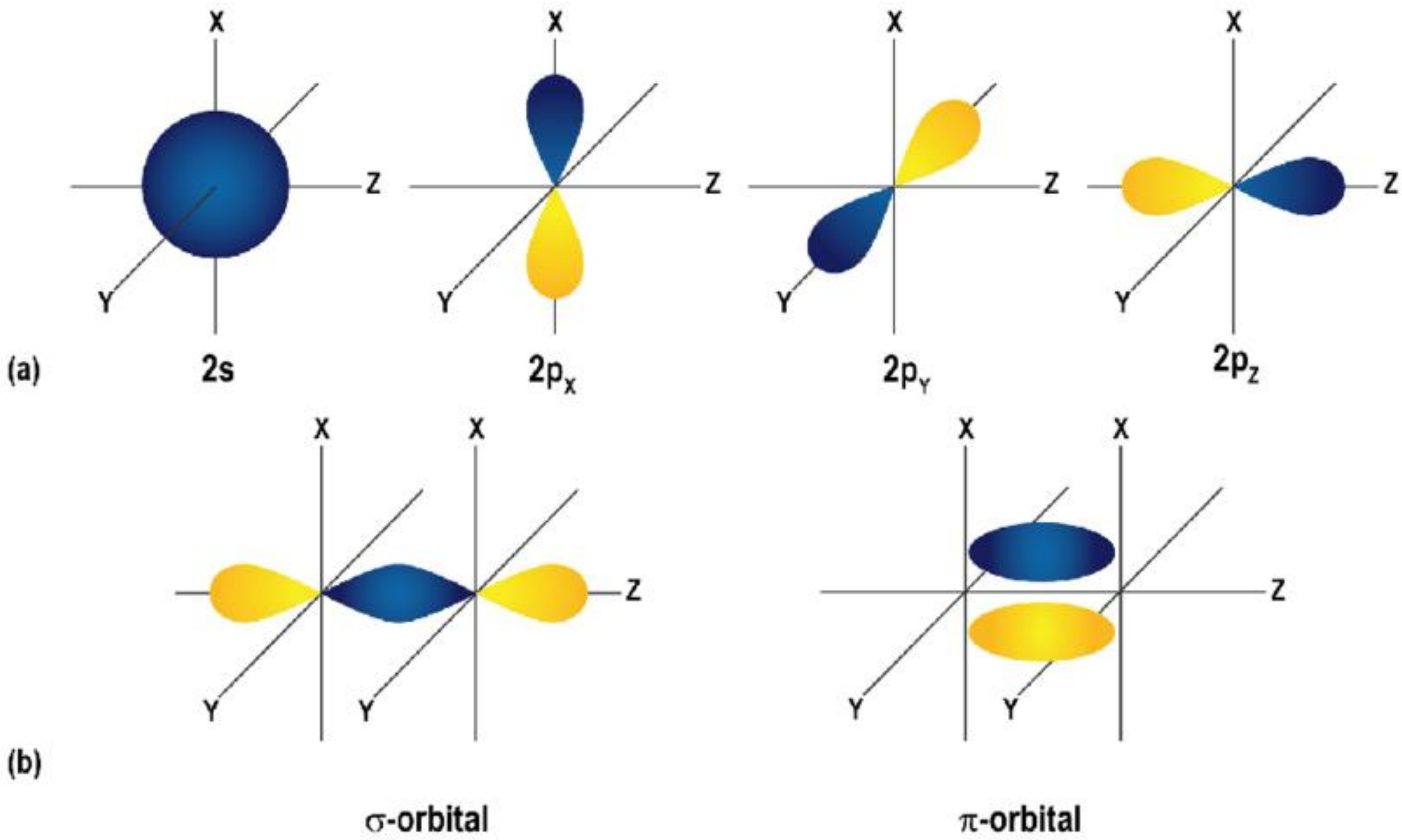
What we are interested in ....



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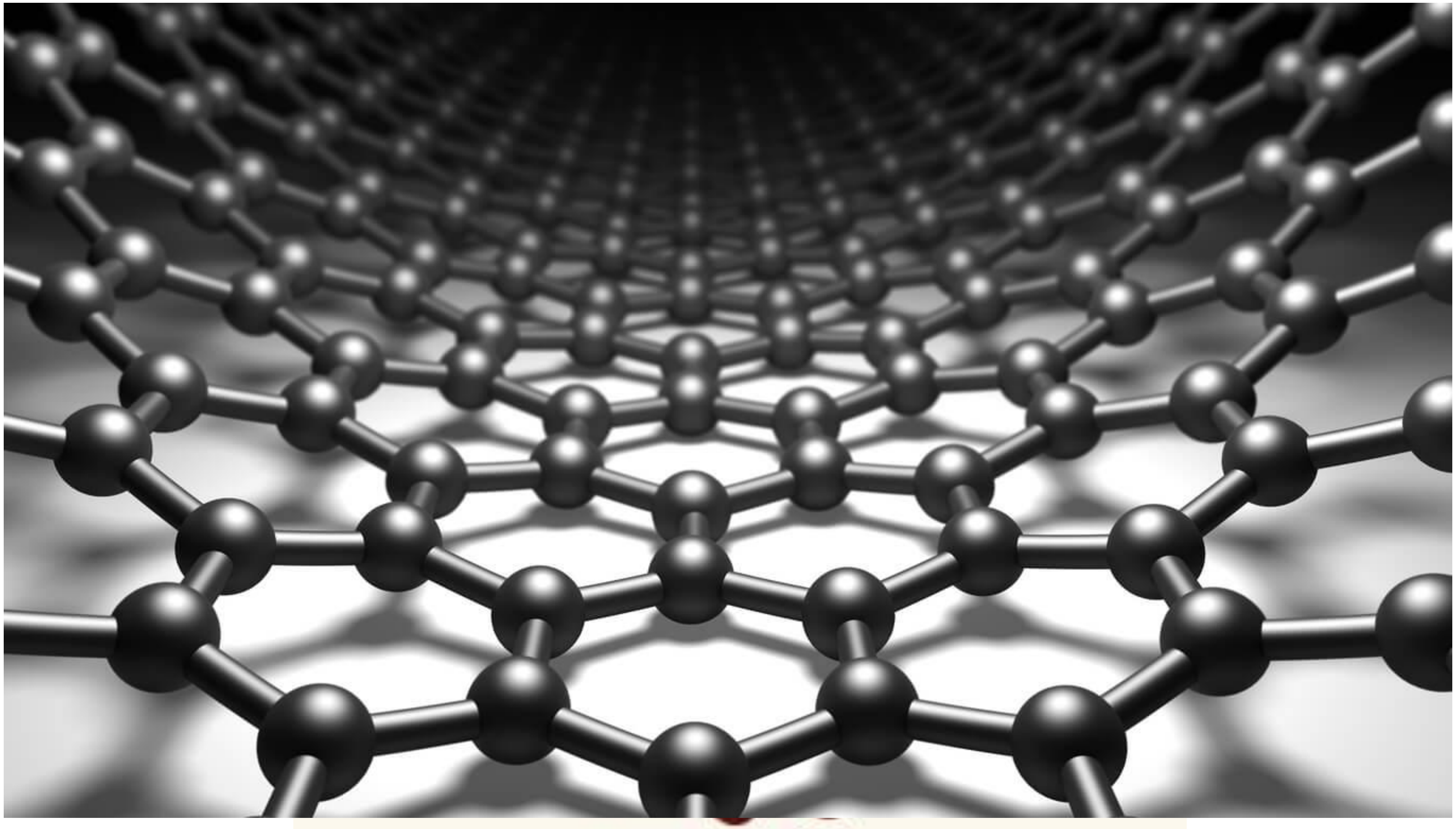
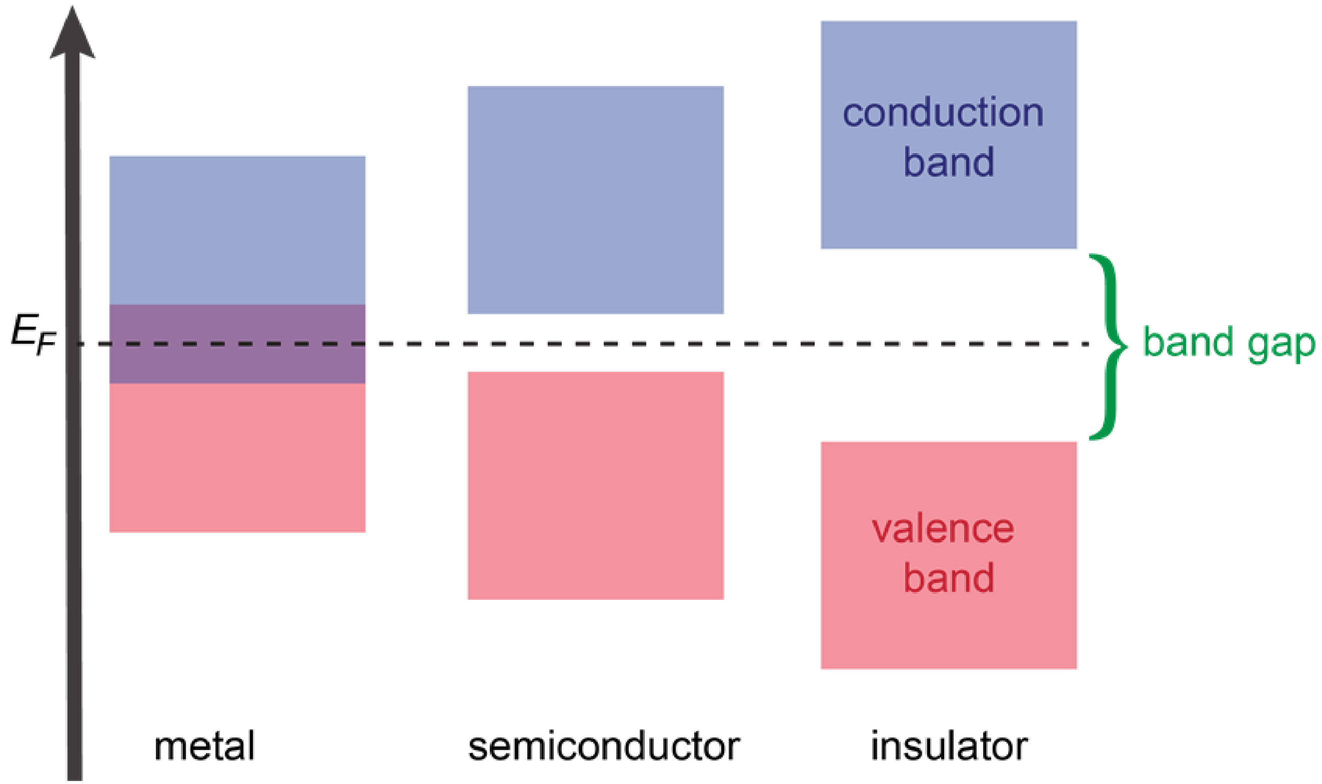
Wijeratne, Kosala, Conducting Polymer Electrodes for Thermogalvanic Cells (2018)

What we are interested in ....



Energy Band Diagram



Fermi-Dirac Distribution

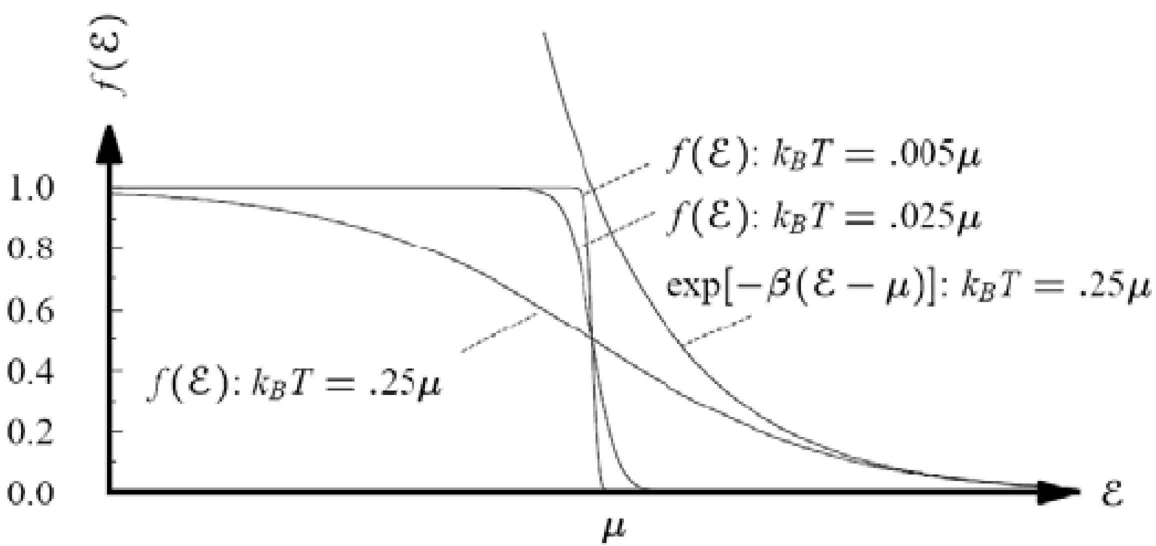
Almost every electronic transport property of solids is proportional to $D(\mathcal{E}_F)$.

Thermal Properties of Free Electron Gas:

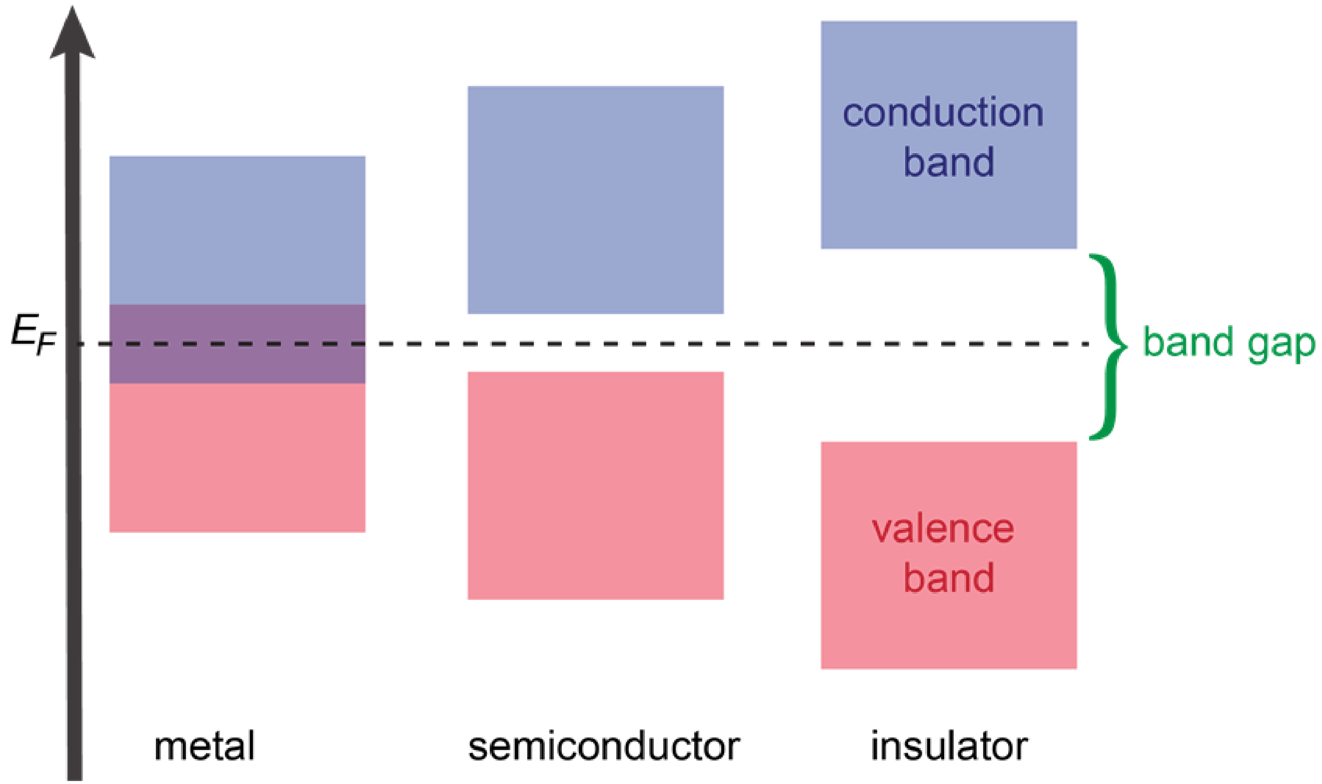
Fermi function
(Fermi-Dirac distribution)

$$f(\mathcal{E}) = \frac{1}{e^{(\mathcal{E}-\mu)/k_B T} + 1}$$

RT ~ 25 meV
4KT broadening ~ 100 meV



Energy Band Diagram



Band Theory

Two approximations

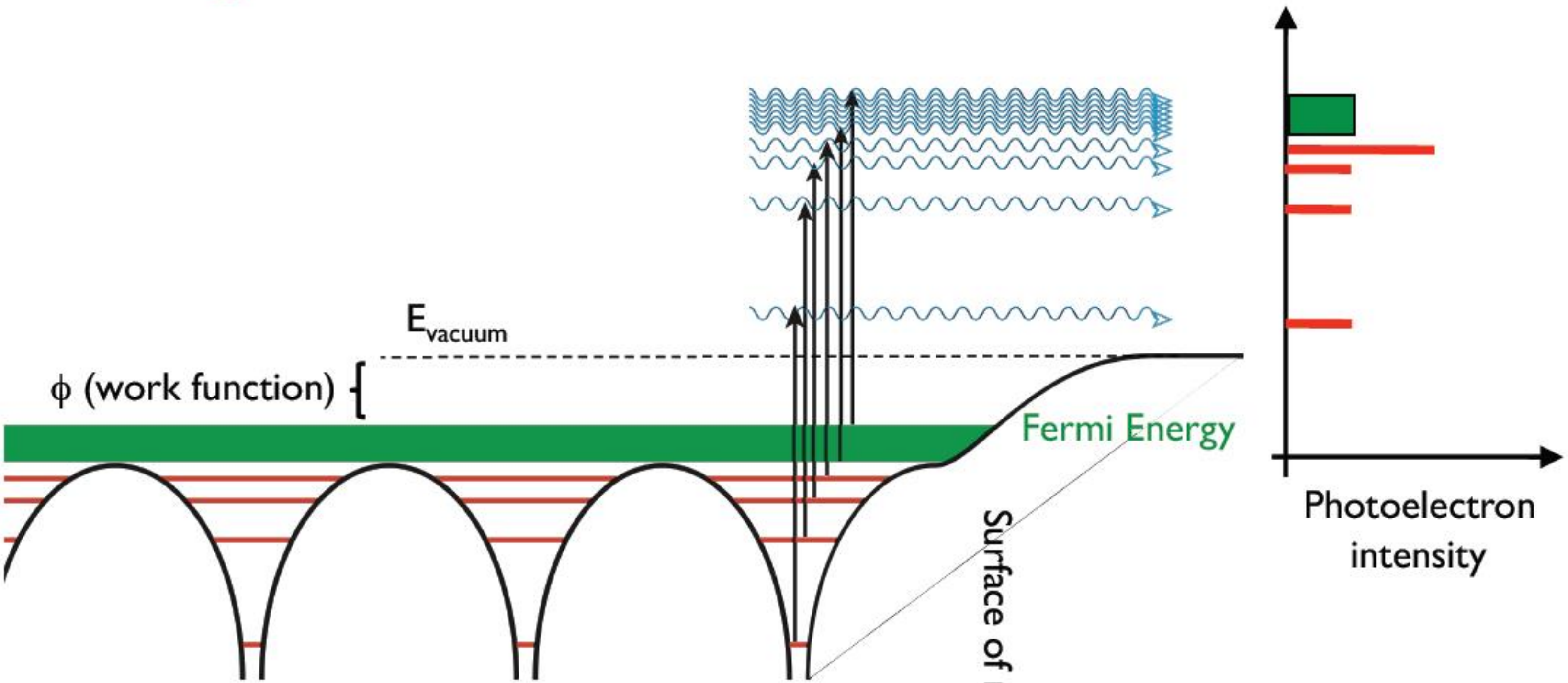
Nearly free electrons. Electrons are non-interacting in a periodic crystal potential which is relatively weak and can be treated as a perturbation. As in the free-electron-gas model, they are still subject to the Pauli exclusion principle.

Free electron gas :
The interactions between electrons and between electrons and nuclei are turned off, subject only to the Pauli exclusion principle.

Tightly-bonding approximation

Electrons are tightly bound to particular atoms, overlapping only weakly with neighbors.

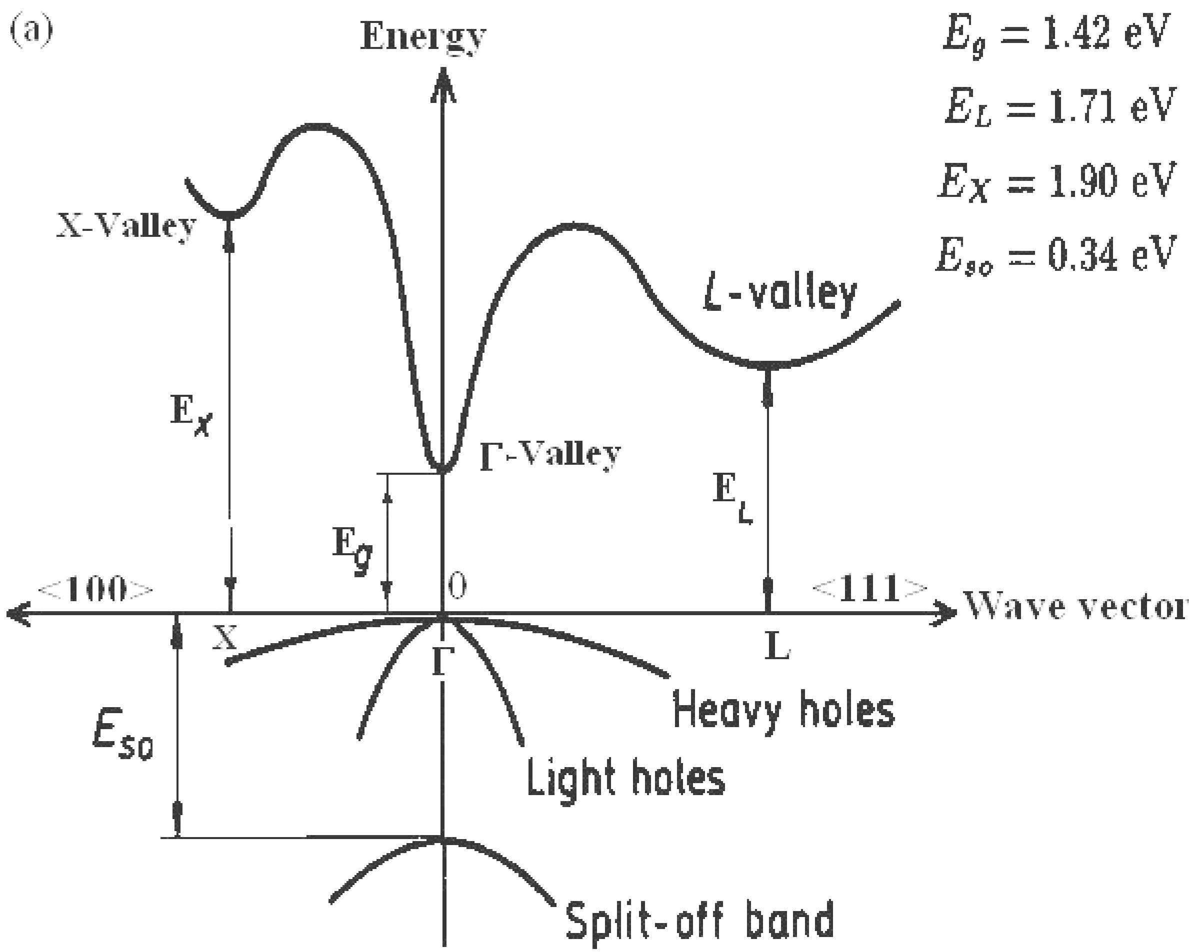
- Deeply bound "*core*" electrons remain basically unchanged
- Outermost "*valence*" electrons hybridize forming continuous "*energy bands*"



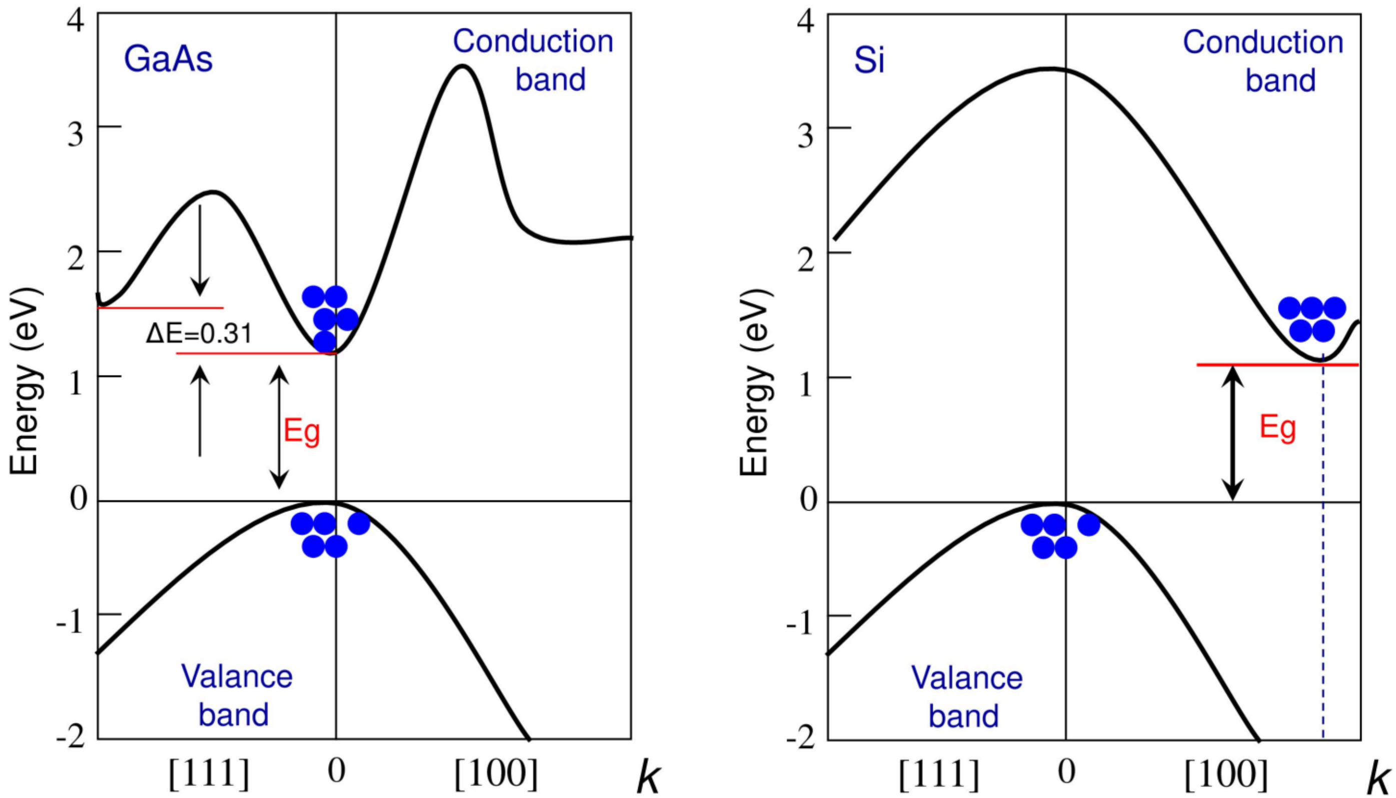
Na metal (Ne + one 3s electron)

Kyle Shen, IGERT Lecture 2008

The main features of the E-k relation of GaAs

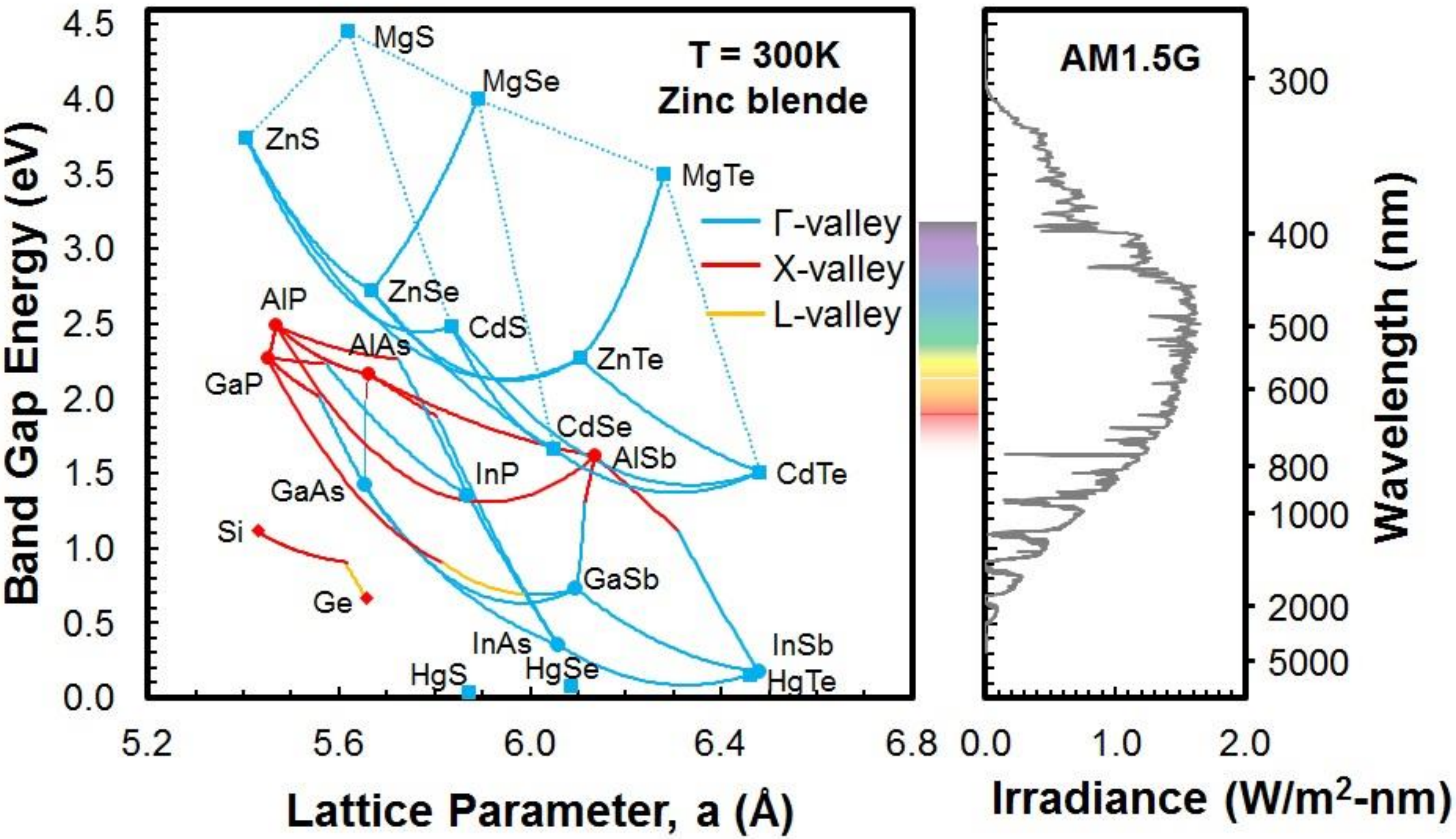


Direct and Indirect Band Gap

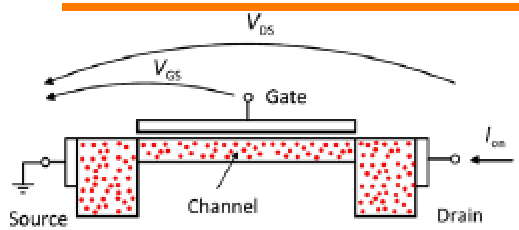


Energy band structures of *GaAs* and *Si*

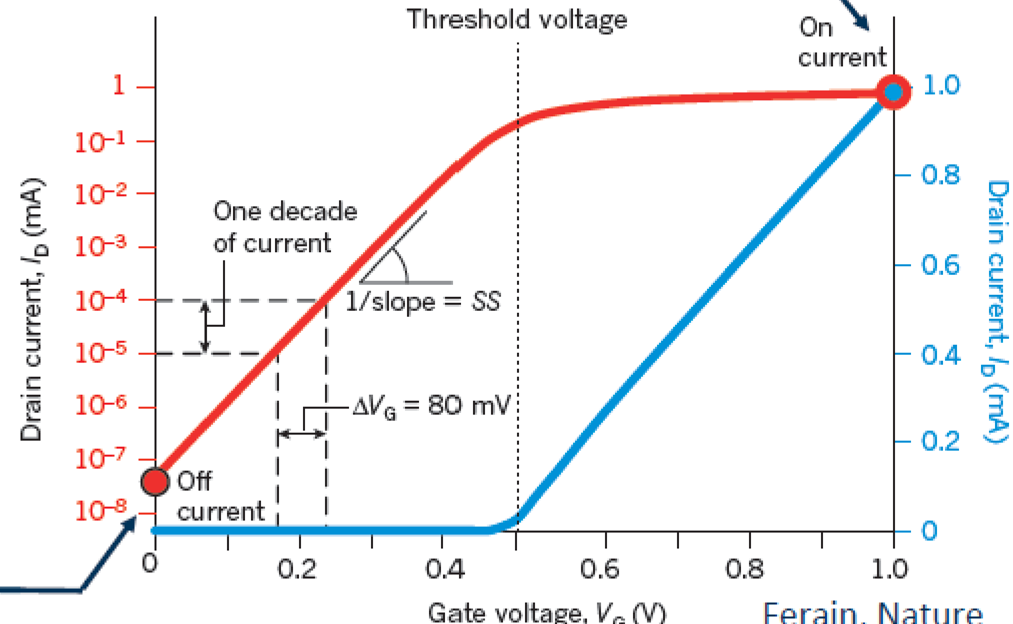
Band gap value of semiconductor



FET Basics – Digital CMOS



On-state

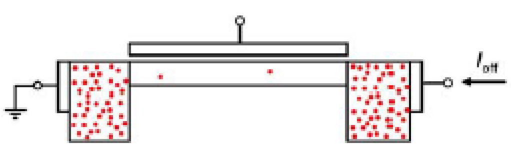


Ferain, Nature 479, 310 (2011).

Requirements for logic

- High on-off ratio I_{on}/I_{off} $10^4 \ldots 10^7$.
- High I_{on} (high speed).
- Low I_{off} (low static power).
- Steep slope in sub-threshold, i.e., small *SS*.

Off-state



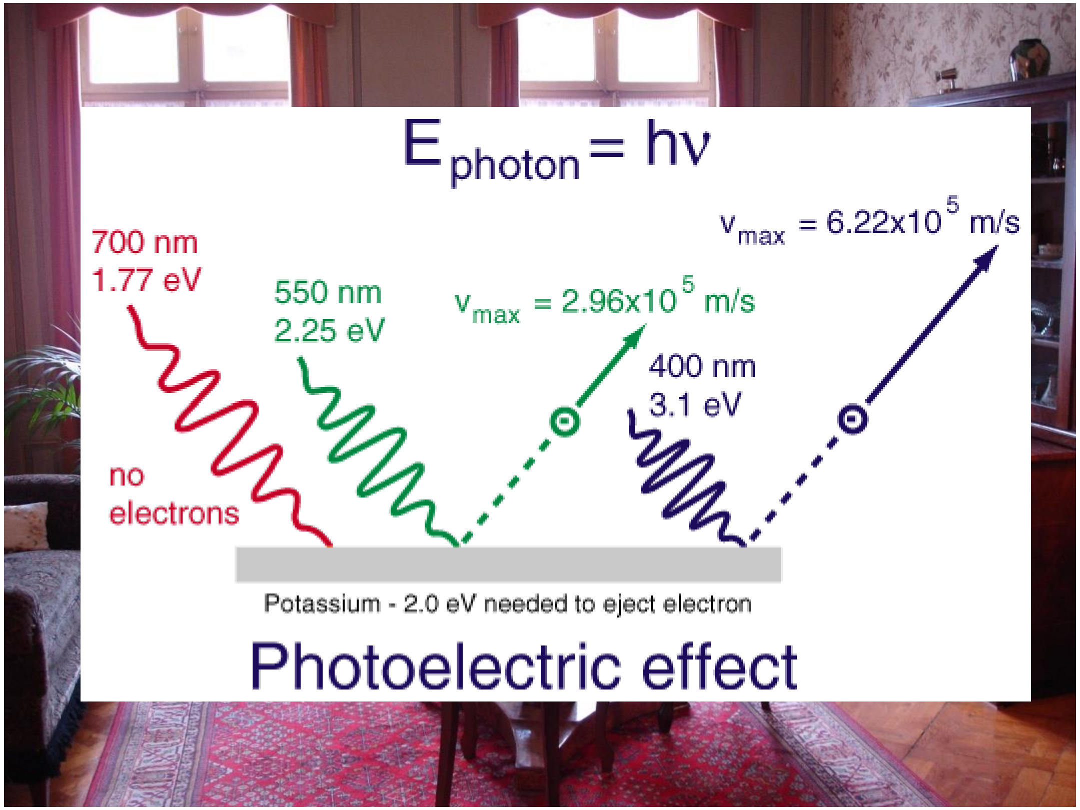
Long channels: $I_{off} \propto \exp \dfrac{-E_G}{m\, k_B\, T}$

A sizeable gap is mandatory!

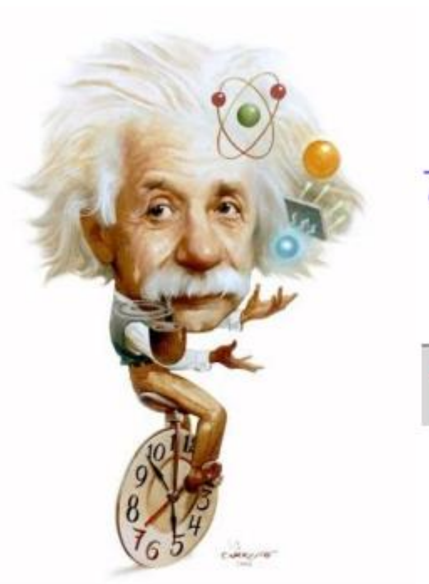
E_G :1~3 eV

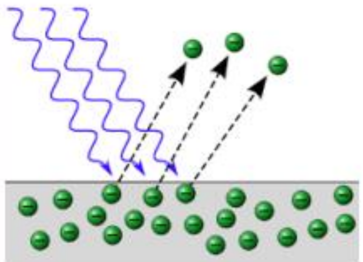
How to probe the band structure of solids

Photoelectric effect



Photoelectric effect --- > Photoemission





The Nobel Prize in Physics 1981

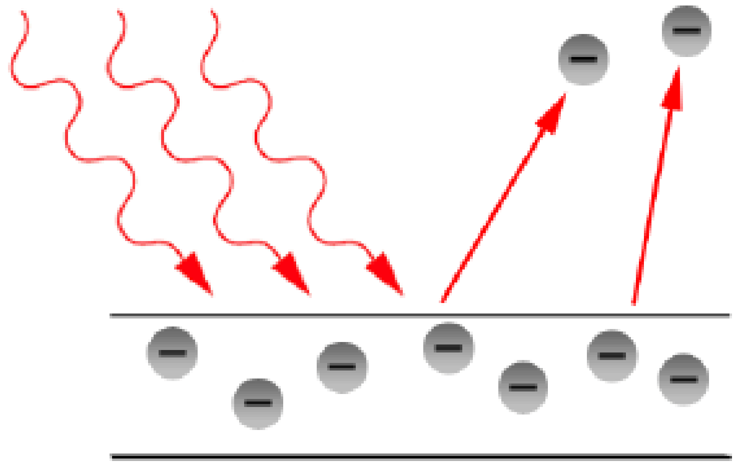
Nicolaas Bloembergen	Arthur Leonard Schawlow	Kai M. Siegbahn
Prize share: 1/4	Prize share: 1/4	Prize share: 1/2

The Nobel Prize in Physics 1981 was divided, one half jointly to Nicolaas Bloembergen and Arthur Leonard Schawlow *"for their contribution to the development of laser spectroscopy"* and the other half to Kai M. Siegbahn *"for his contribution to the development of high-resolution electron spectroscopy"*.

The Principle of Photoemission Spectroscopy

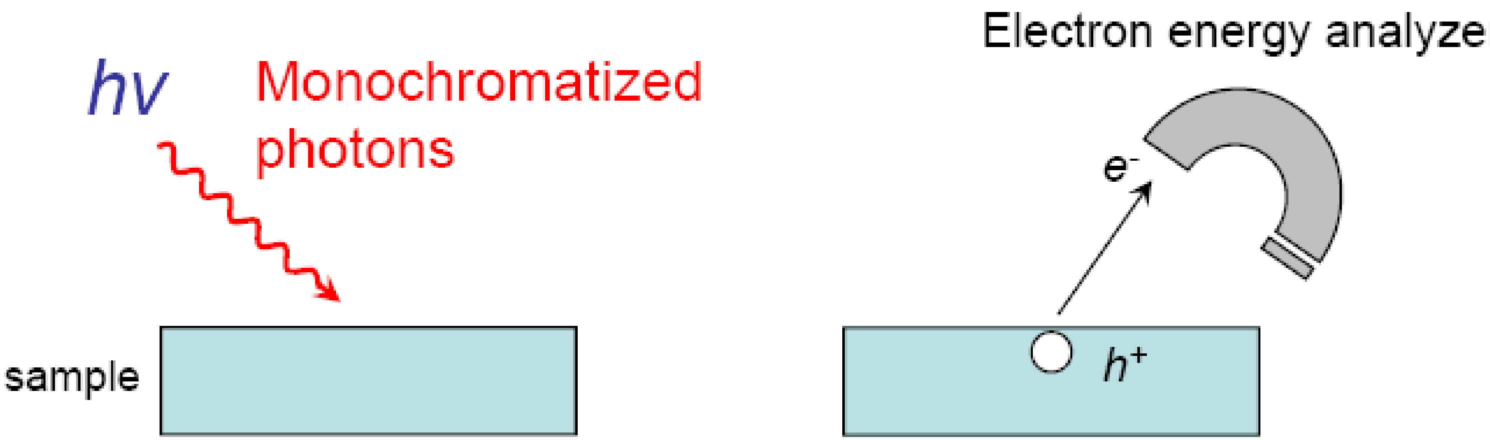
First ESCA spectrometer at the
Department of Physics in Uppsala.

What is photoemission?



Photon in –> electron out (emission)

What is photoemission spectroscopy? (photoelectron spectroscopy) (PES)



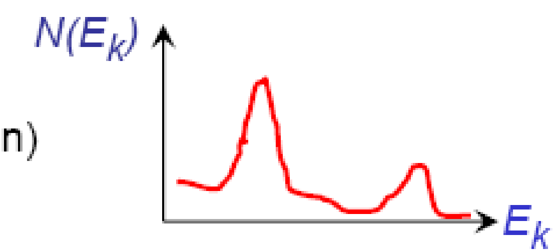
Initial state: ground (neutral) state

Final state: hole (excited) state

Conservation of energy

$\boldsymbol{E_k = h\nu + E_i - E_f}$ (most general expression)

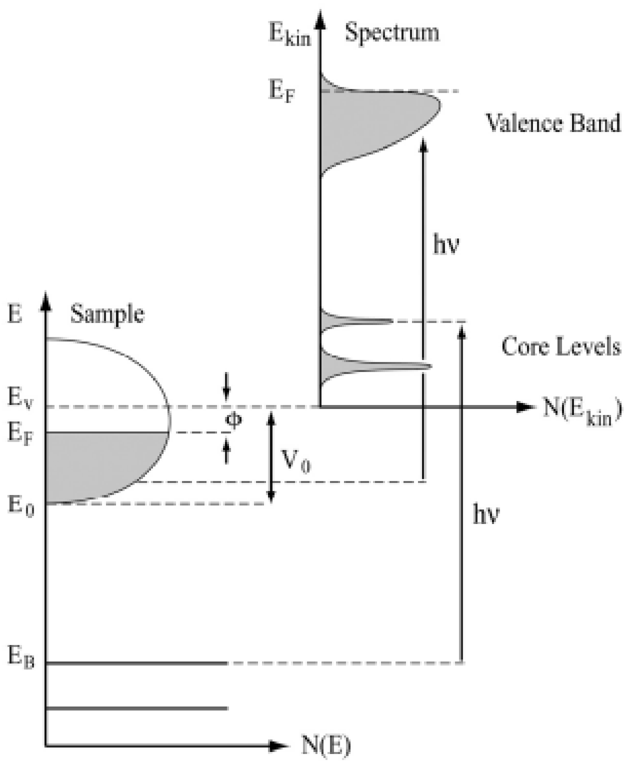
$\boldsymbol{E_k}$: photoelectron kinetic energy
$\boldsymbol{E_i}$ (N) : total initial state system energy
$\boldsymbol{E_f}$ $(N\text{-}1)$: total final state system energy



Energy Distribution Curve (EDC) (Spectrum)

Conservation of energy in PES

Energetics in PES



Hufner, Damascelli

$E_k = h\nu - E_B - \phi$

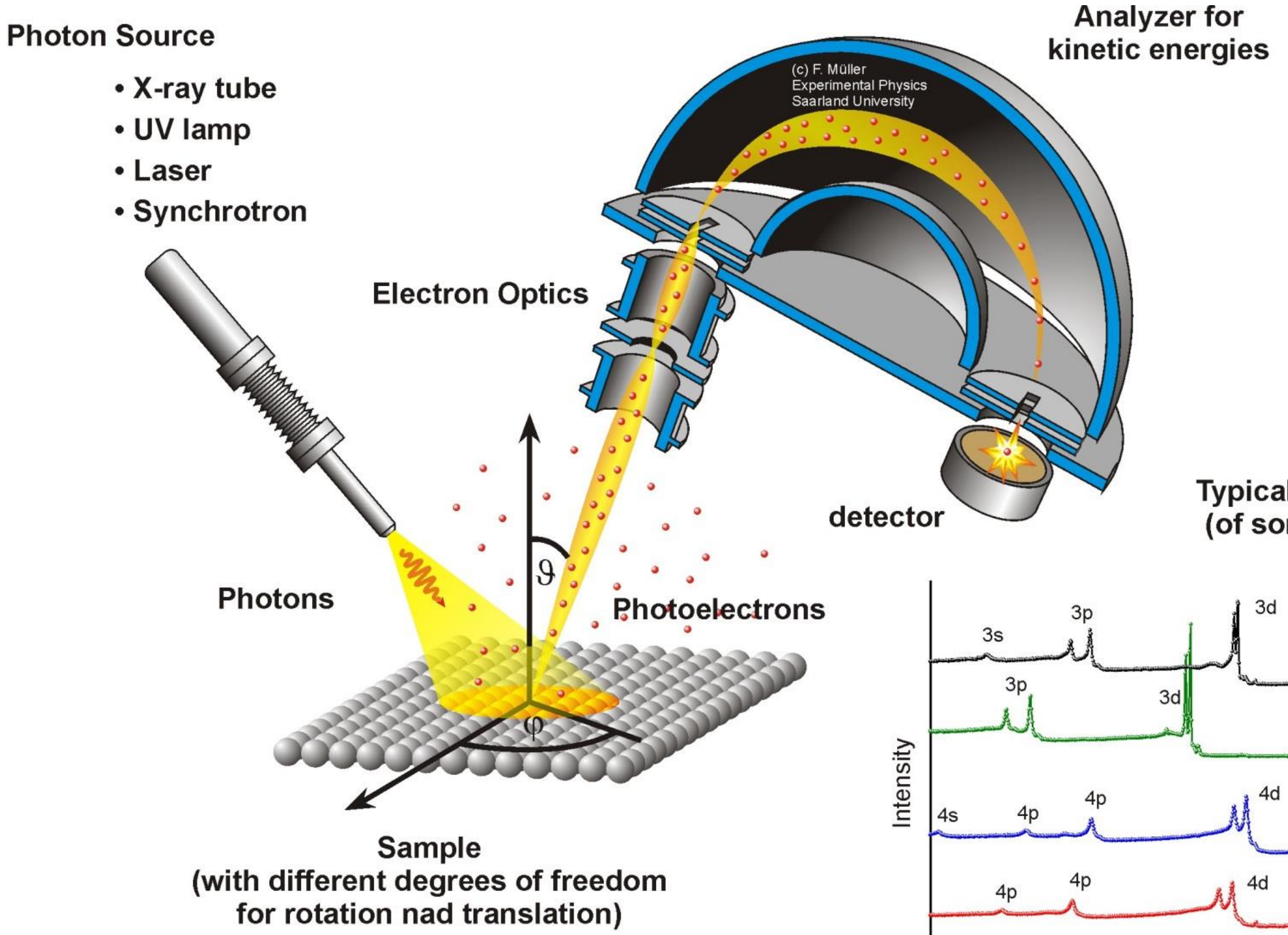
Conservation of energy

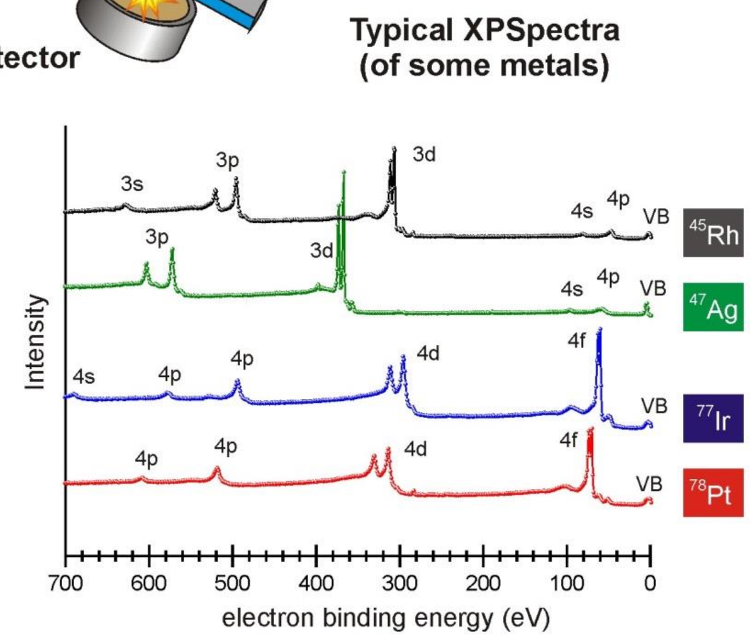
E_v : vacuum (energy) level
E_F : Fermi (energy) level
$\phi = E_v - E_F$: work function
E_0 : bottom of valence band
$V_0 = E_v - E_0$: inner potential

E_k^{max} marks E_F in spectra
E_B measured relative to $E_F = 0$

Usually fixed photon energy
scanning not needed

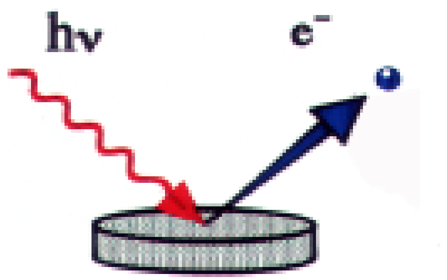
The Principle of Photoemission Spectroscopy





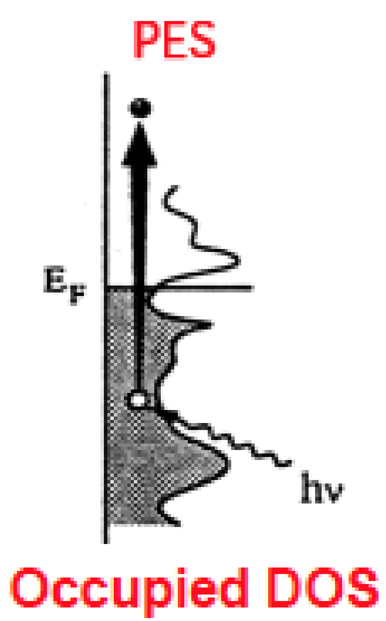
What is AIPES?

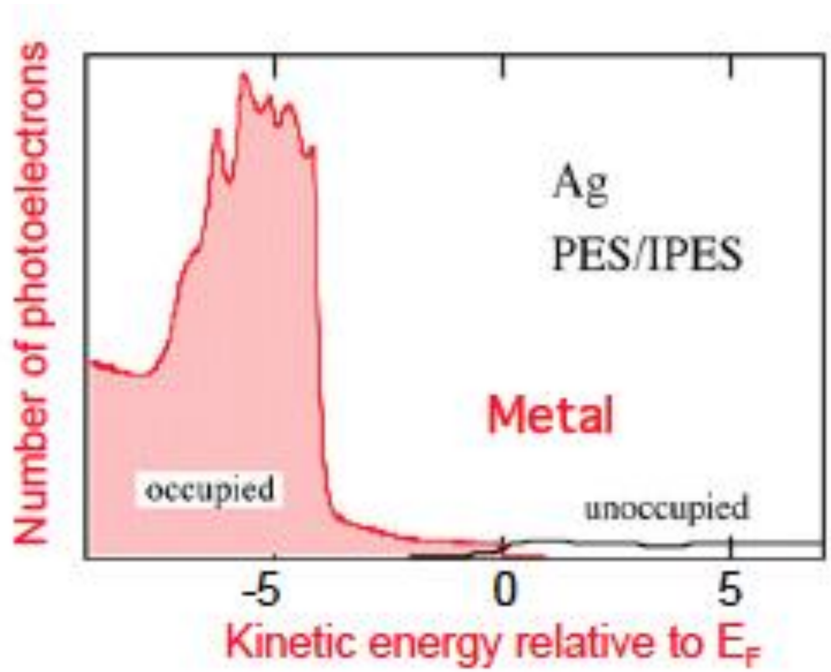
Angle-Integrtaed Photoemission Spectroscopy



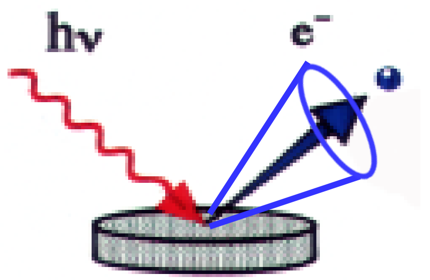
Conservation of Energy

$$\boxed{E_{ph} = E_b + \Phi + E_k}$$





Angle-Integrtaed Photoemission Spectroscopy



Conservation of Energy

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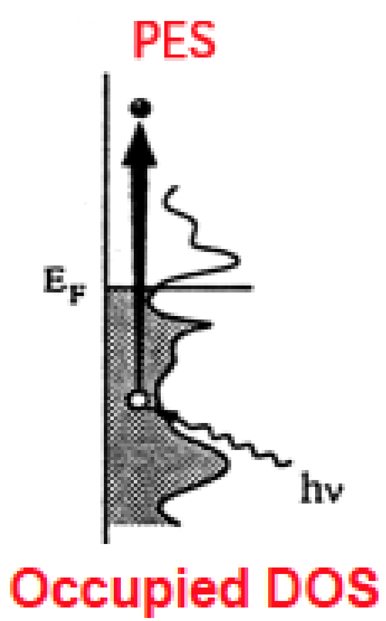
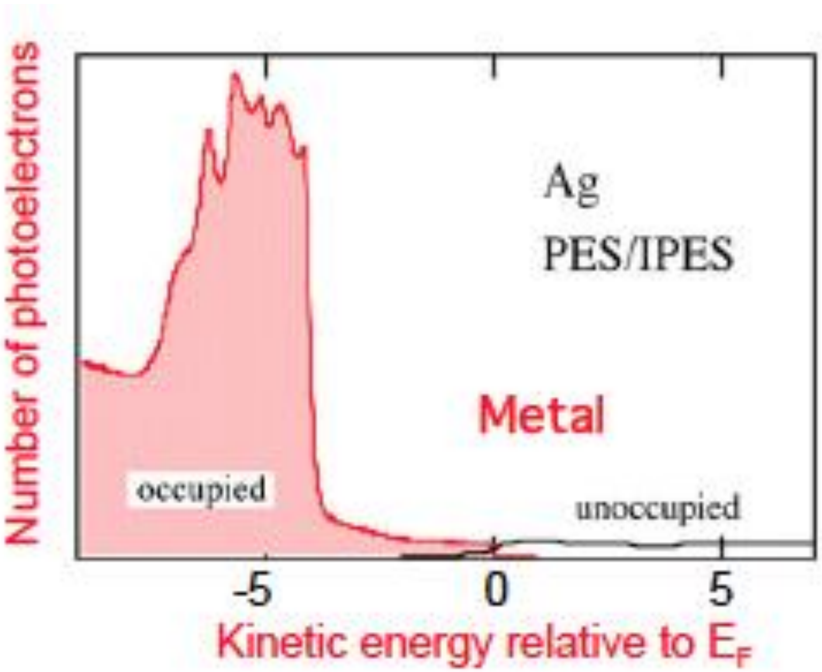
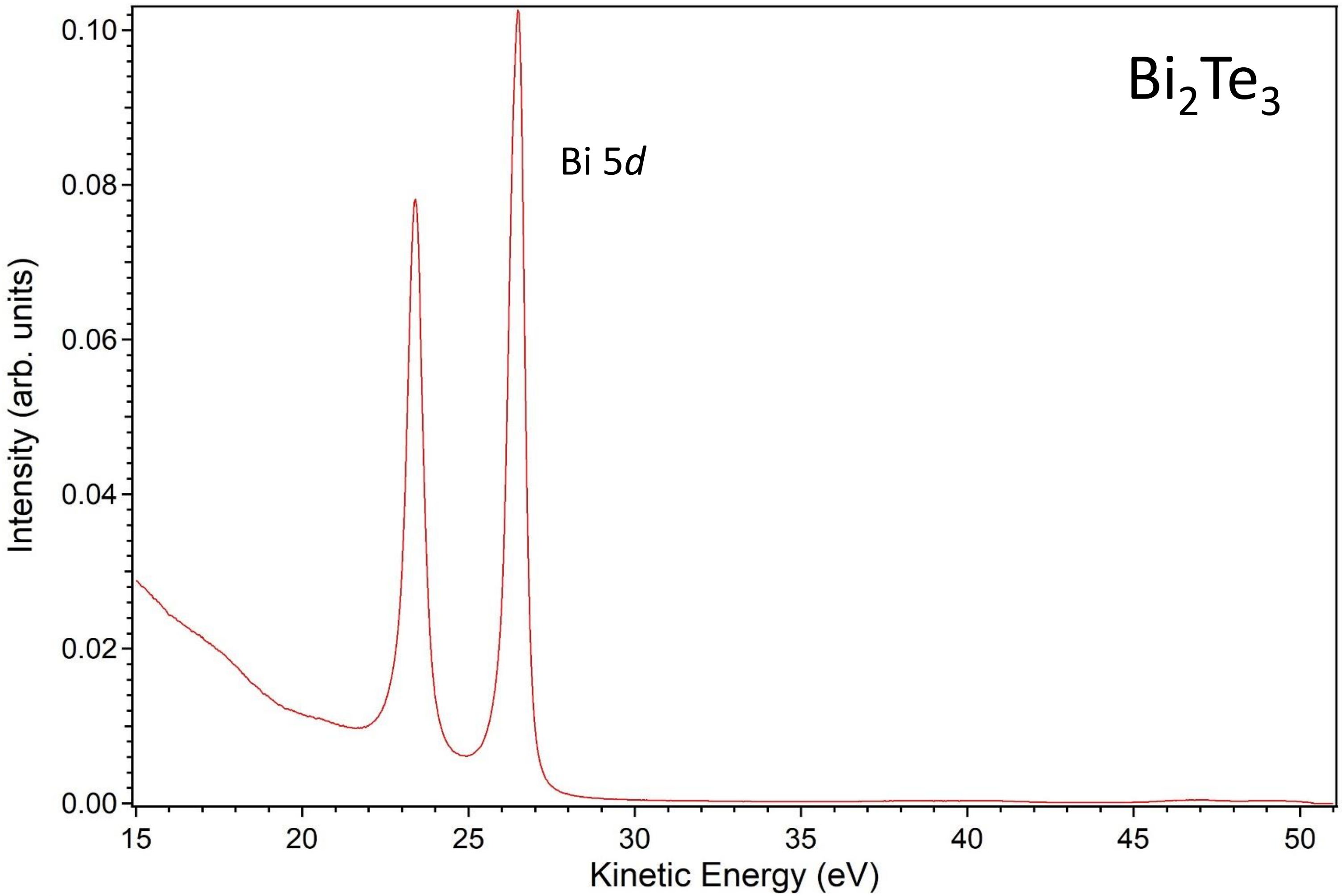
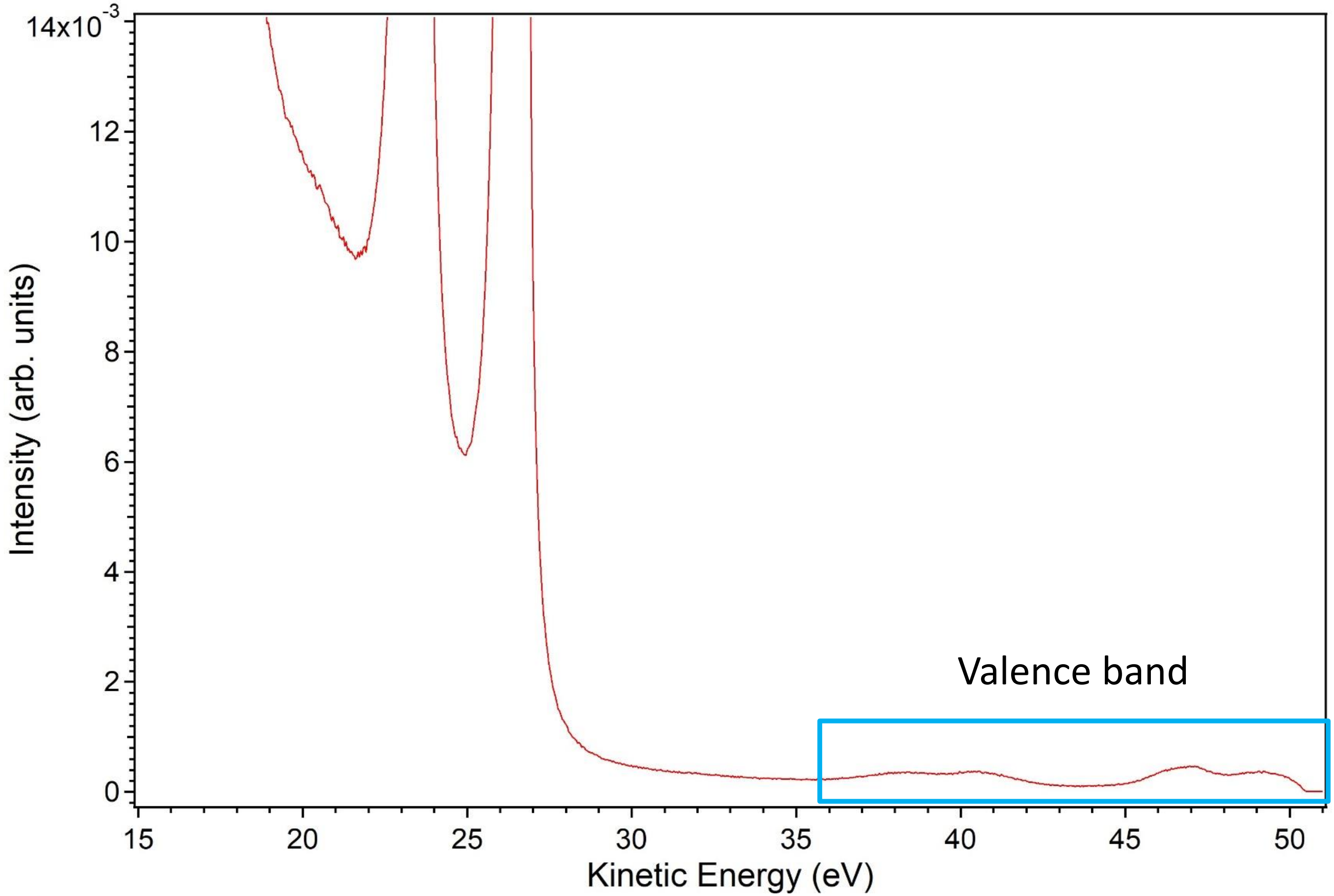




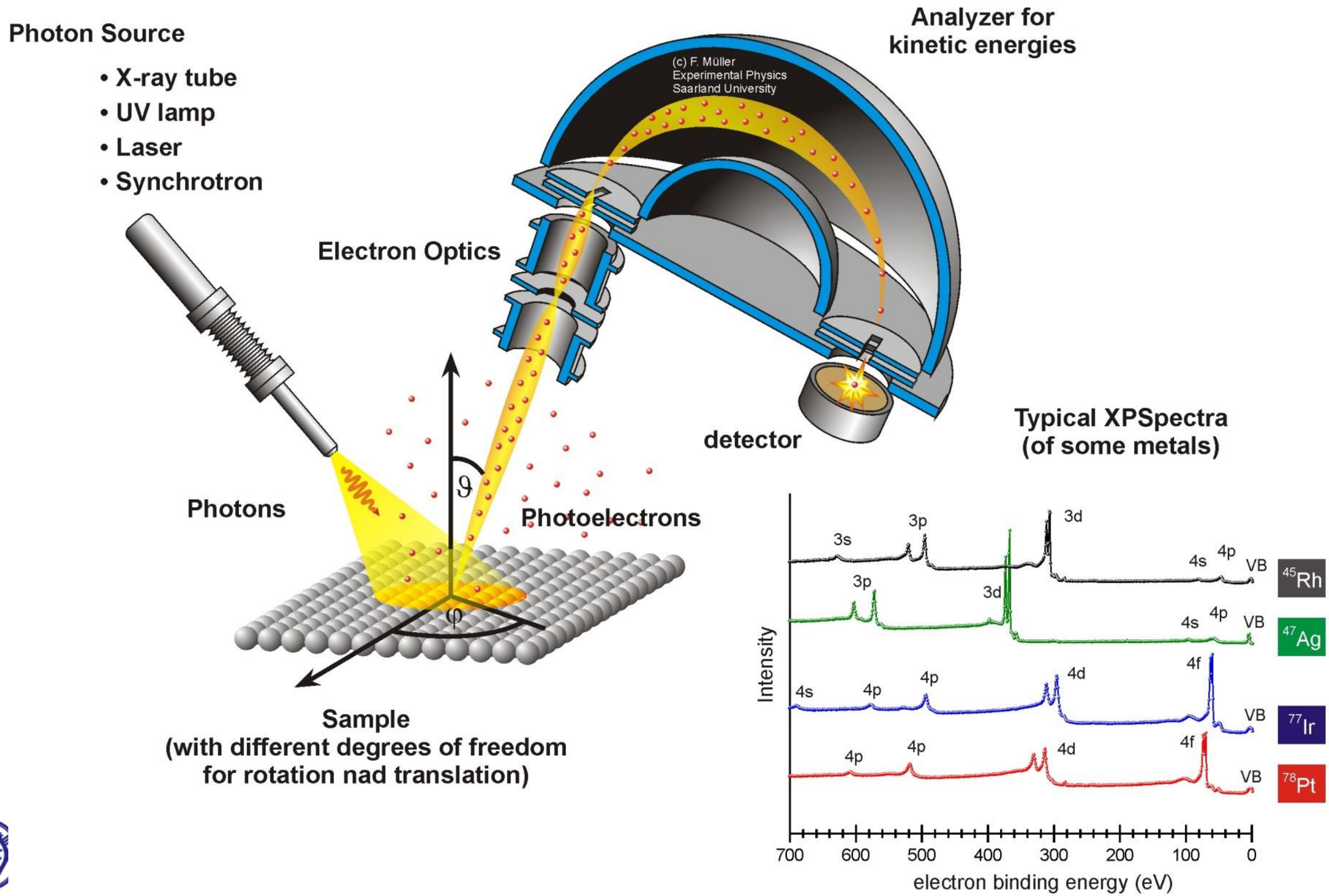
Energy Distribution Curve (EDC)



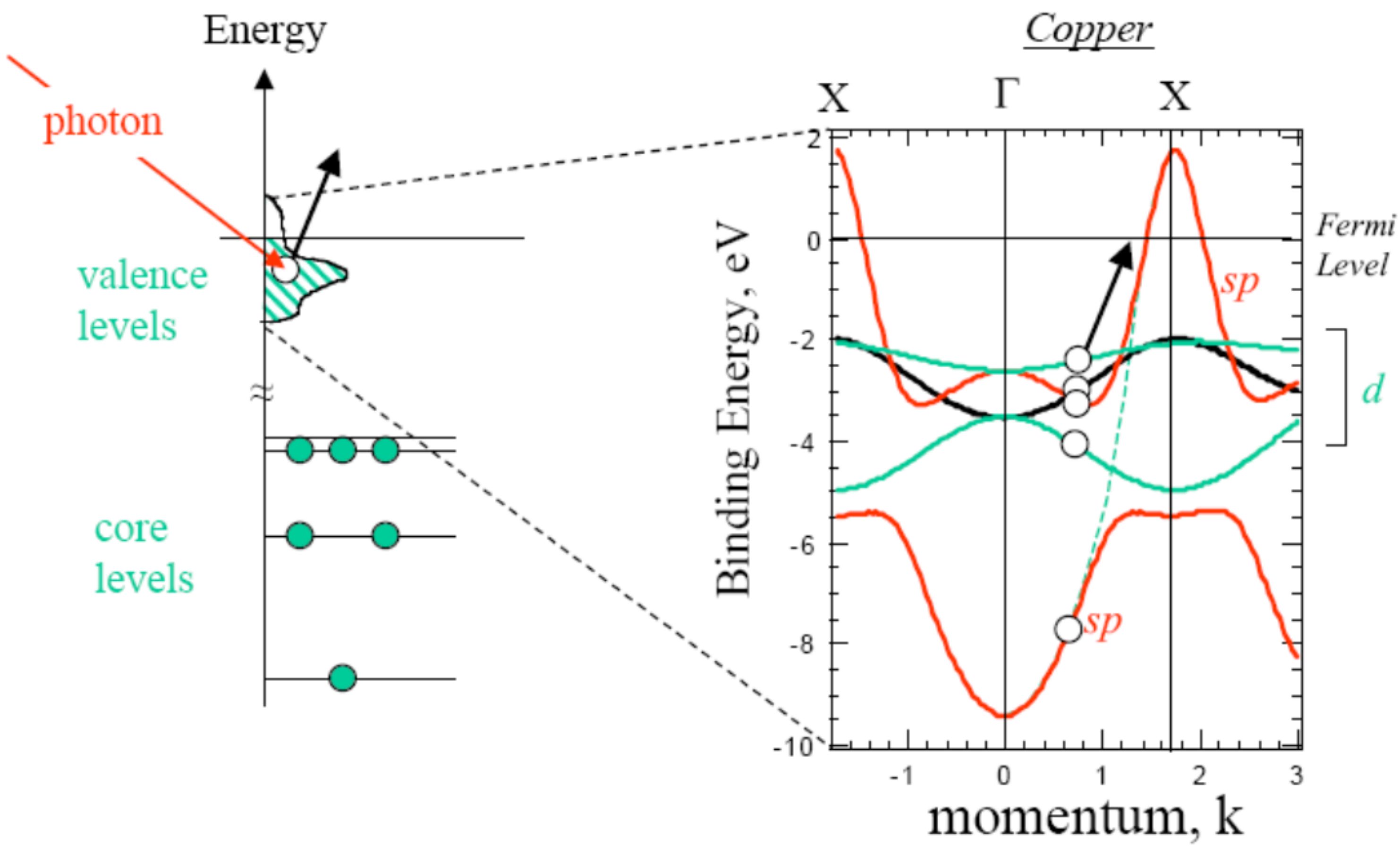
Energy distribution curve (EDC)



The Principle of Photoemission Spectroscopy

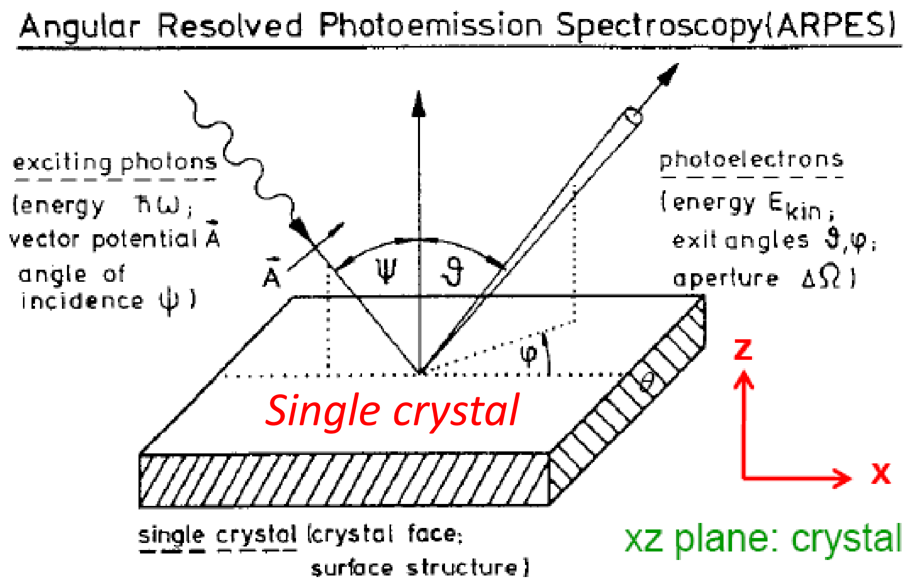


what is angle-resolved photoemission spectroscopy (ARPES)



What is ARPES?

Angle-Resolved Photoemission Spectroscopy



xz plane: crystal symmetry plane

Electron emission angle: Θ

Photon incident angle: ψ, *s*- and *p*-polarization

$$k_{//} = \sqrt{\frac{2m}{\hbar^2}E_k} \cdot \sin\theta$$

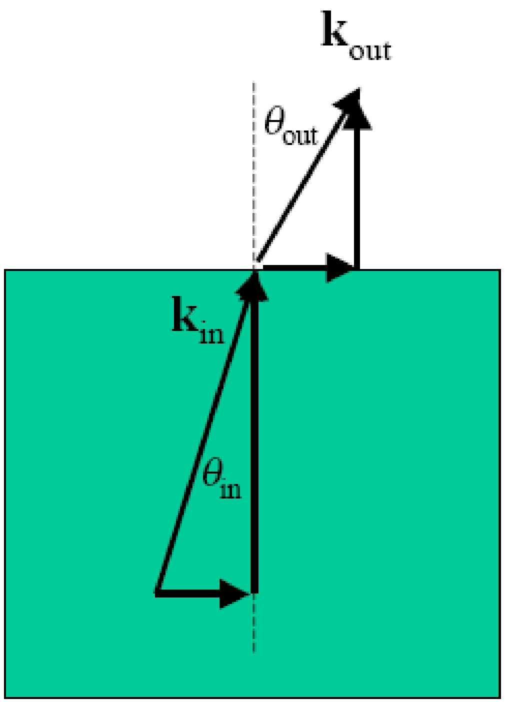
$$k_{//}(\text{Å}^{-1}) = 0.5123\sqrt{E_k(eV)} \cdot \sin\theta$$

$k_{||}$(inside) = $k_{||}$(outside)

Conservation of liner momentum

Important for 3D and 2D band mapping

Conservation of linear momentum parallel to surface



Kinematic relations

$$k_{out} = \sqrt{\frac{2m}{\hbar^2} E_{kin}}$$

$$k_{in} = \sqrt{\frac{2m}{\hbar^2} (E_{kin} + V_0)}$$

$$k_{out,\|} = k_{in,\|} \equiv k_{\|}$$

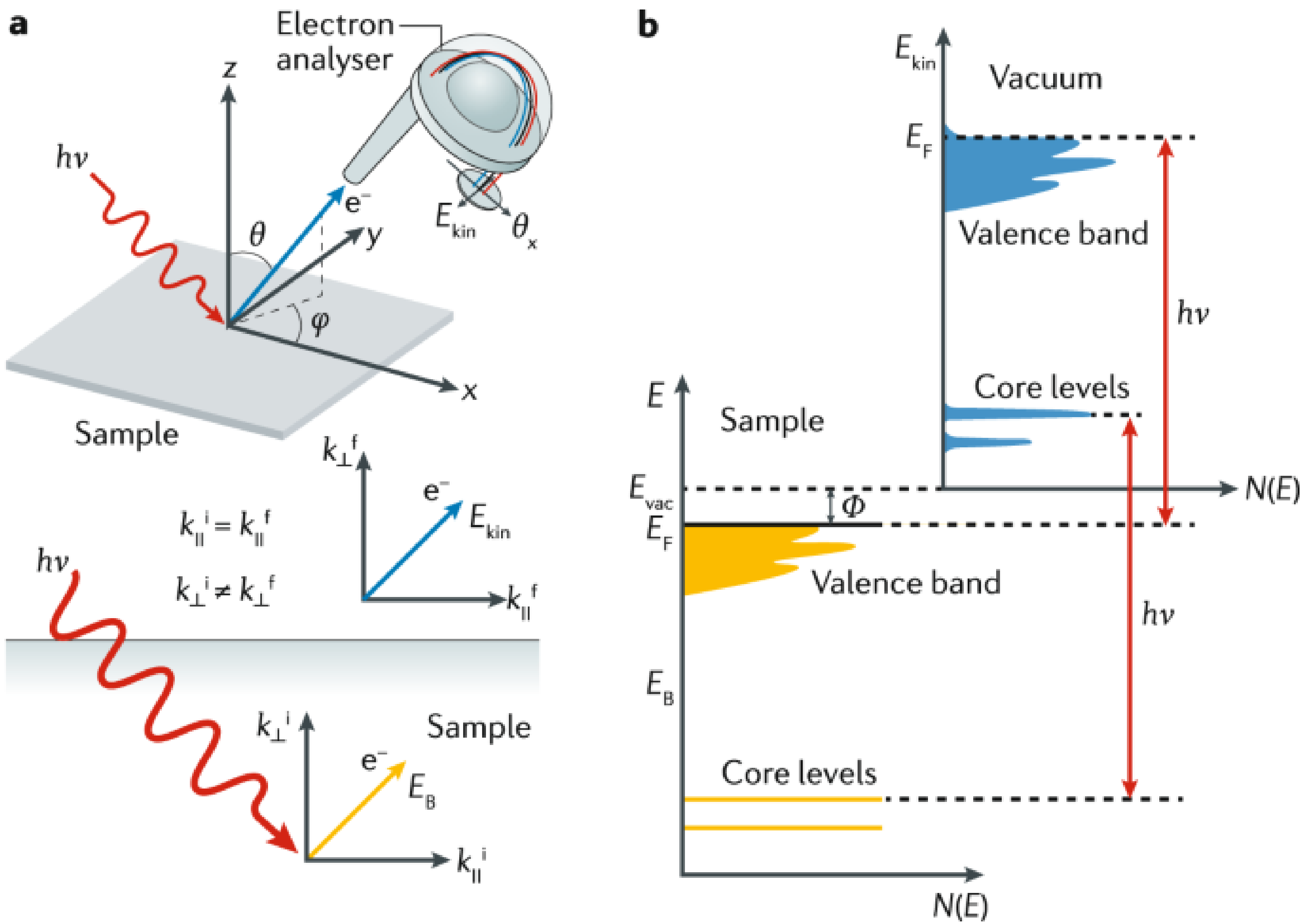
"Snell's Law"

$$k_{\|} = \sin\theta_{out} \sqrt{\frac{2m}{\hbar^2} E_{kin}} = \sin\theta_{in} \sqrt{\frac{2m}{\hbar^2} (E_{kin} + V_0)}$$

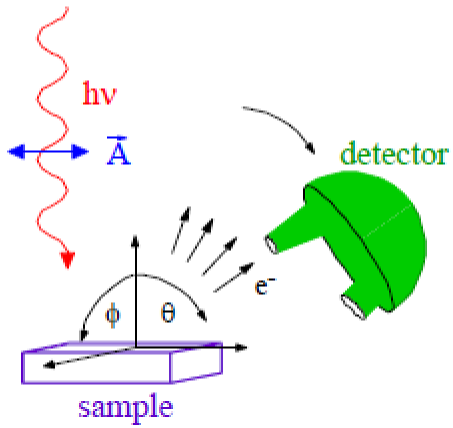
Critical angle for emission

$$(\sin\theta_{out})_{\max} = \sqrt{\frac{E_{kin}}{E_{kin} + V_0}}$$

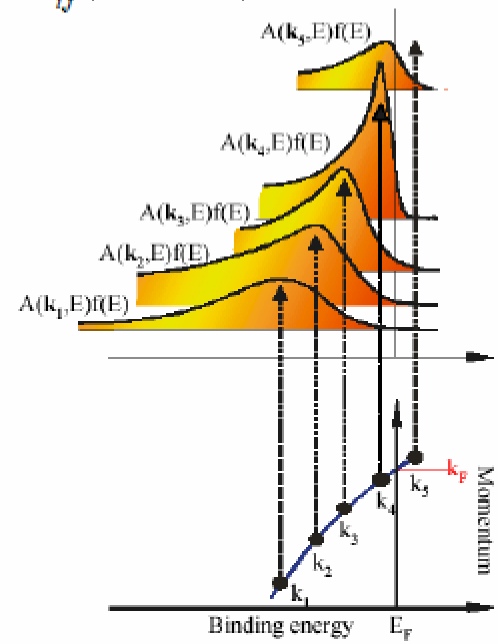
what is ARPES?



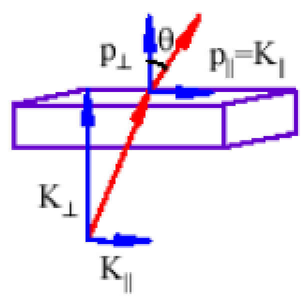
What is ARPES?



$$\text{Intensity} \propto \sum_{i,f} \left| \left\langle f \middle| \vec{\mathbf{p}} \bullet \vec{\mathbf{A}} \middle| i \right\rangle \right|^2 A(\vec{\mathbf{k}}, E) f(E)$$



Electron momentum Parallel to the surface is conserved



- Low photon energy provides better momentum resolution, but the covering range of BZ is also small.
- We expect to study the electronic structure of solids at VUV region (10 eV~ 100 eV).

$$k_{\perp} = 0.5123\sqrt{(E_{kin}\cos^2\theta + V_0)}$$

$$k_{//} = 0.5123\sqrt{E_{kin}}\sin\theta$$

Light sources and terminology

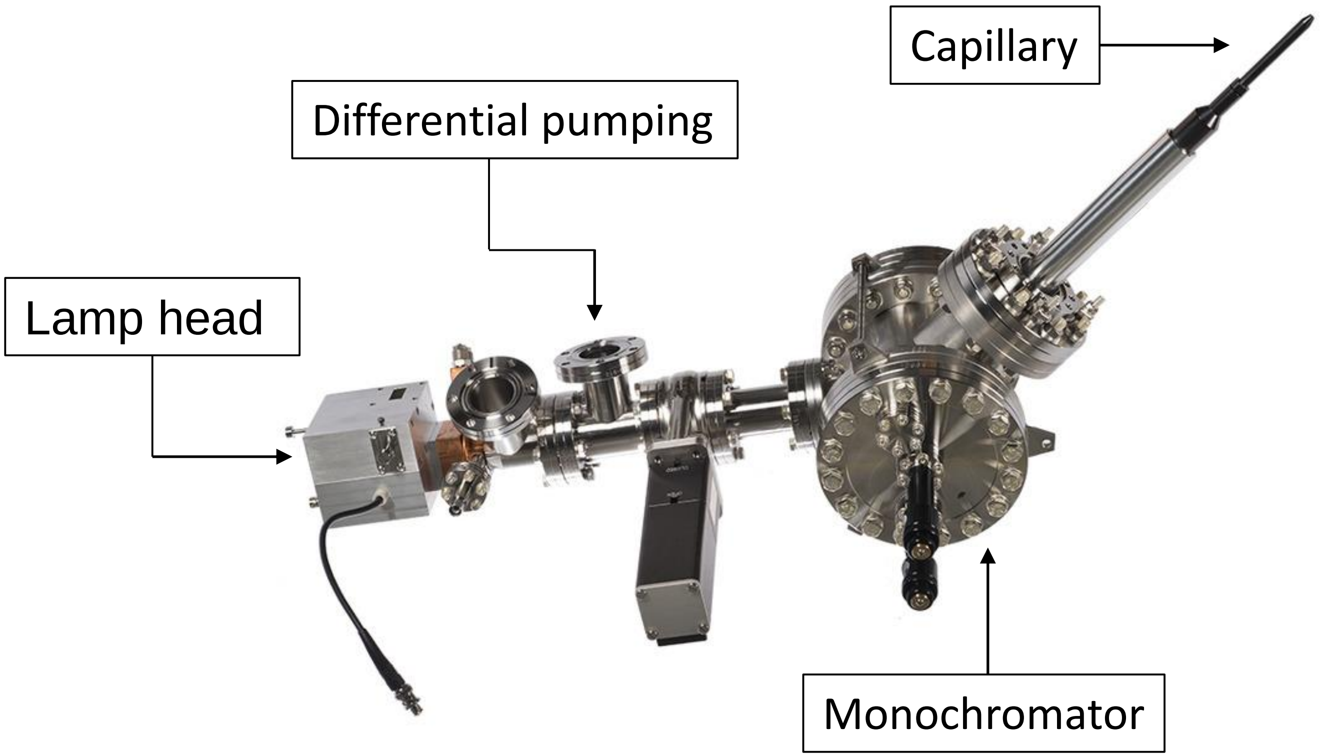
- **Ultraviolet Photoemission Spectroscopy (UPS)**
  - UV He lamp (21.2 eV, 40.8 eV)
  - Laser : 6 eV (BBO), 8 eV (KBBF), 11 eV (gas cell) or HHG (High harmonic generation)
  - Valence band PES, direct electronic state info.

- **X-ray Photoemission Spectroscopy (XPS)**
  (Electron Spectroscopy for Chemical Analysis) (ESCA)
  - X-ray gun (Al: 1486.6 eV, Mg: 1253.6 eV)
  - core level PE, indirect electronic state info
  - chemical analysis

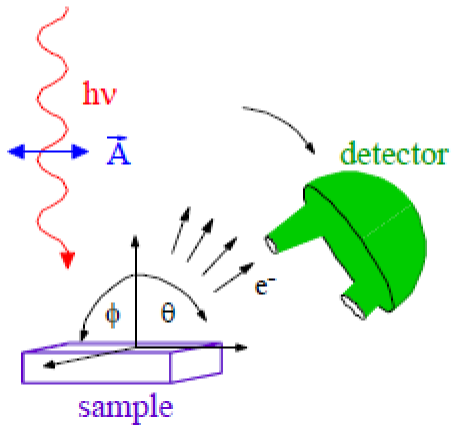
- **Synchrotron radiation**
  - continuous tunable wavelength
  - valance band and core level

Light sources and terminology

UV lamp (He I$_\alpha$ 21.2 eV, He II$_\alpha$ 40.8 eV)



What is ARPES?



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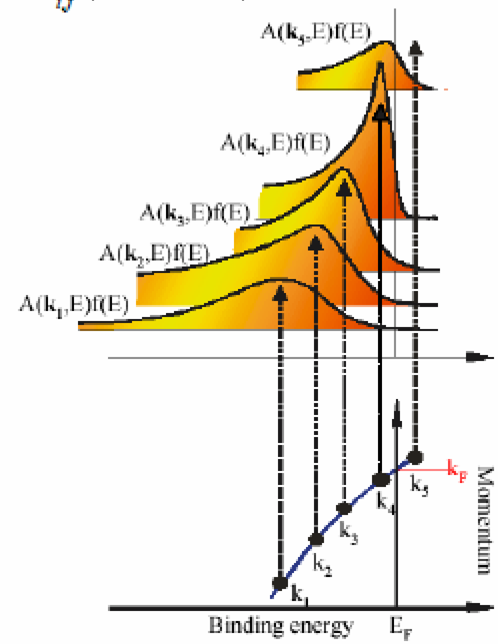

Electron momentum
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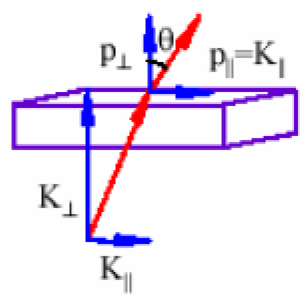

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Light sources and terminology

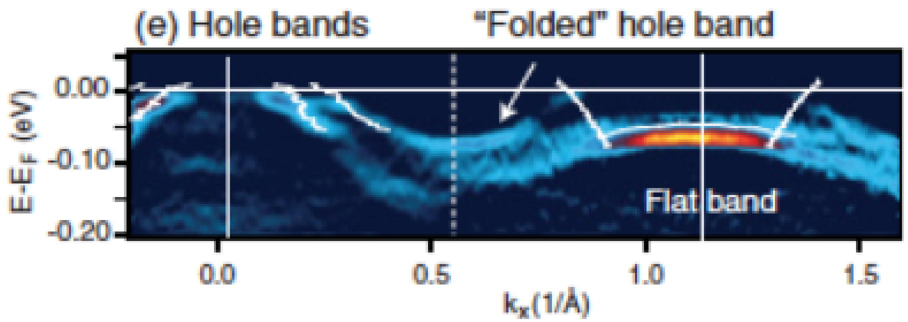
Photon energy

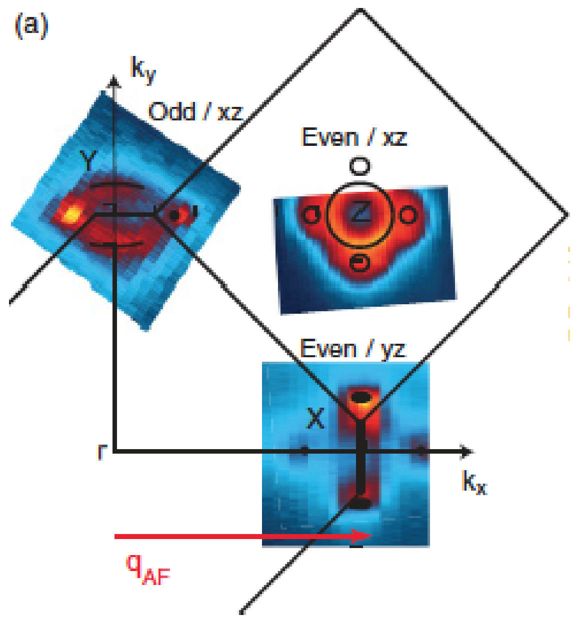
- defines the detectable area of Energy

$$E_{kin} = \hbar\nu - \phi$$

- defines the accessible area of BZ

$$k_{//} = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\theta$$

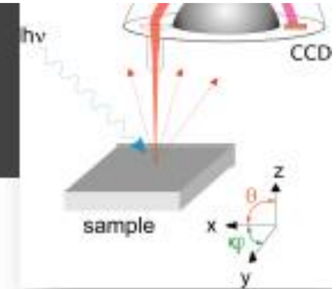




Phys. Rev. B 84, 014509 (2011)

Light sources and terminology

Photon energy



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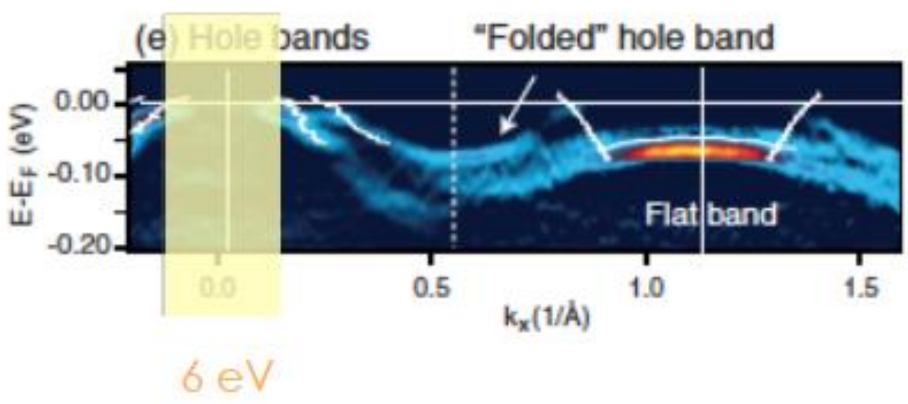

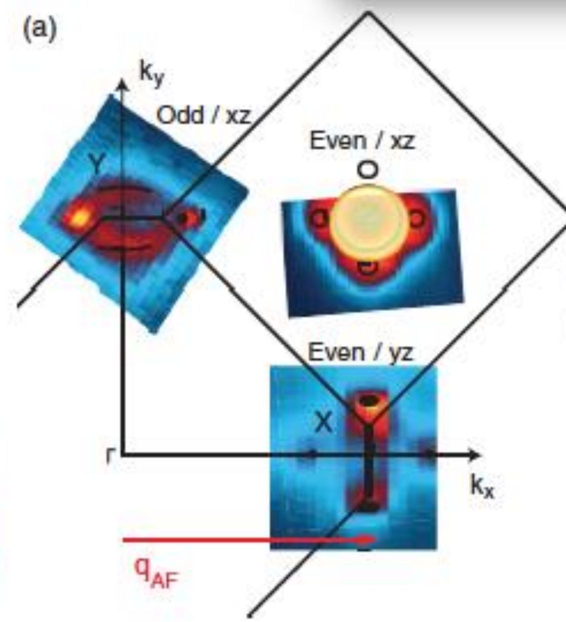

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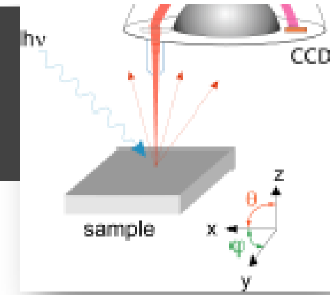
Photon energy



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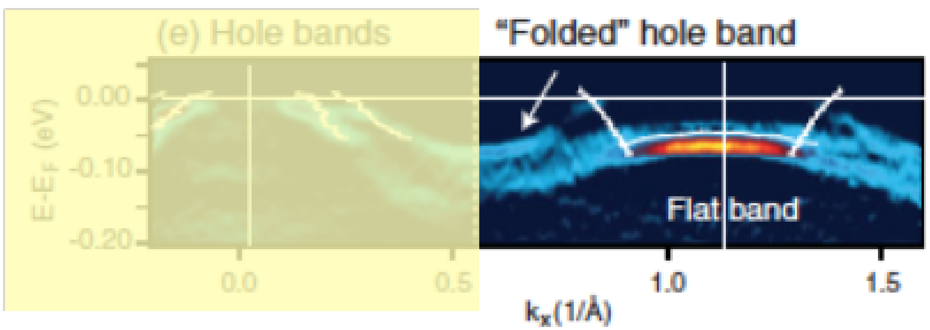
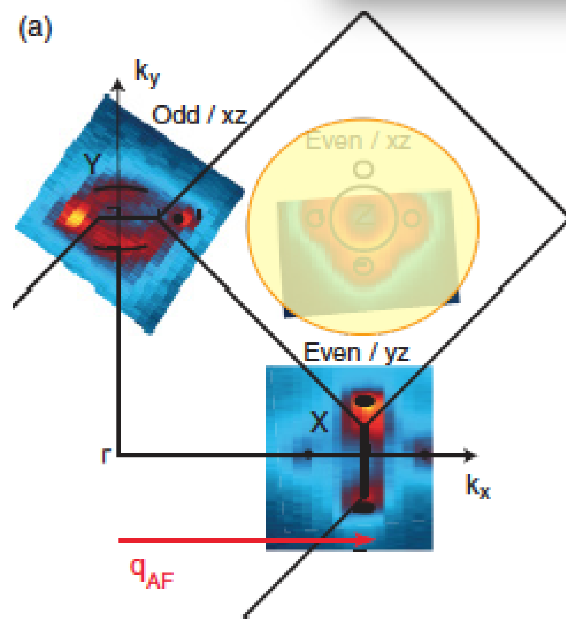


20 eV



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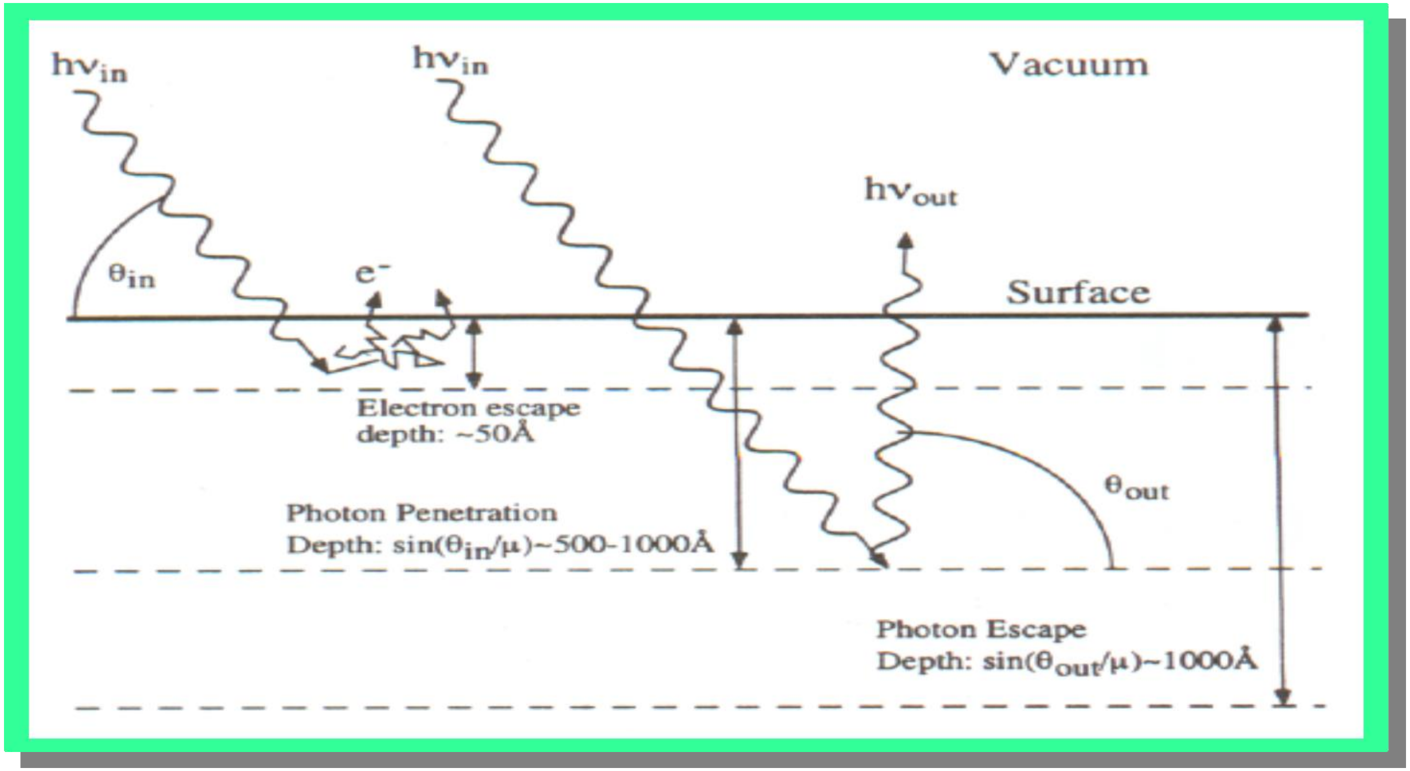
Light sources and terminology



The powerful spectroscopic tools such as XPS and UPS might be limited in in-situ chemical analysis because of the short penetration depth of electrons.

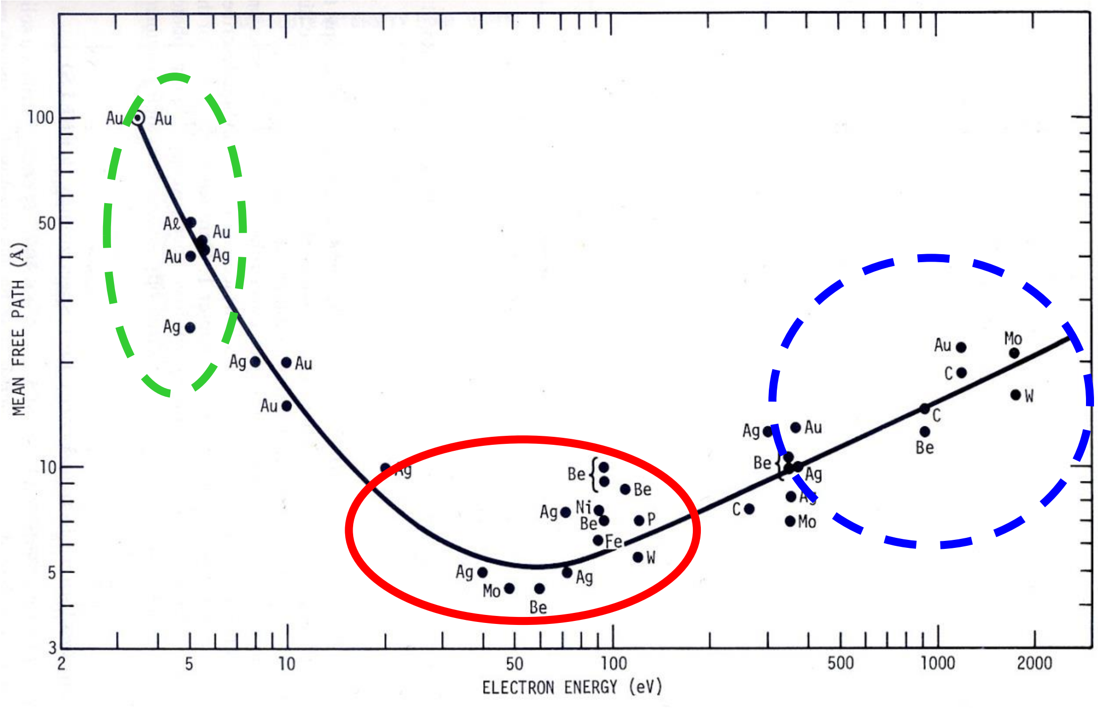
Light sources and terminology

Why are electrons so useful as probes of surfaces?

Or

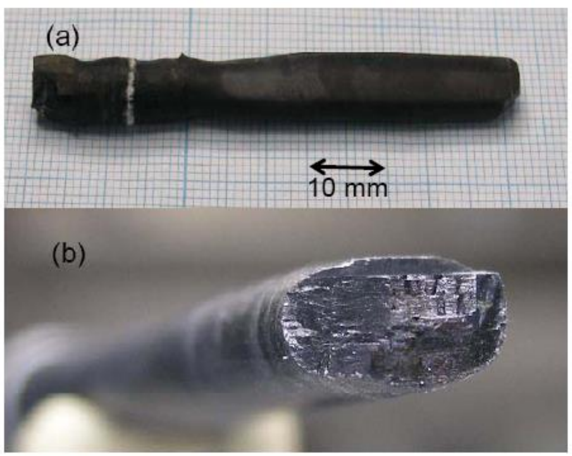
Not so useful for studying bulk properties !!

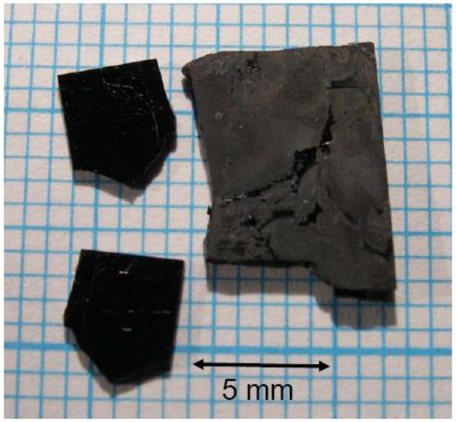
Minimum due to electron-electron scattering, mainly plasmons
PES is a surface sensitive technique! (requires UHV)
High energy photoemission: several keV to increase bulk sensitivity



Light sources and terminology

- UHV environment : better than 1×10^{-10} Torr
- Single crystals or *in-situ* growth thin films
- Conductors or semiconductors
- Tunable photon energies





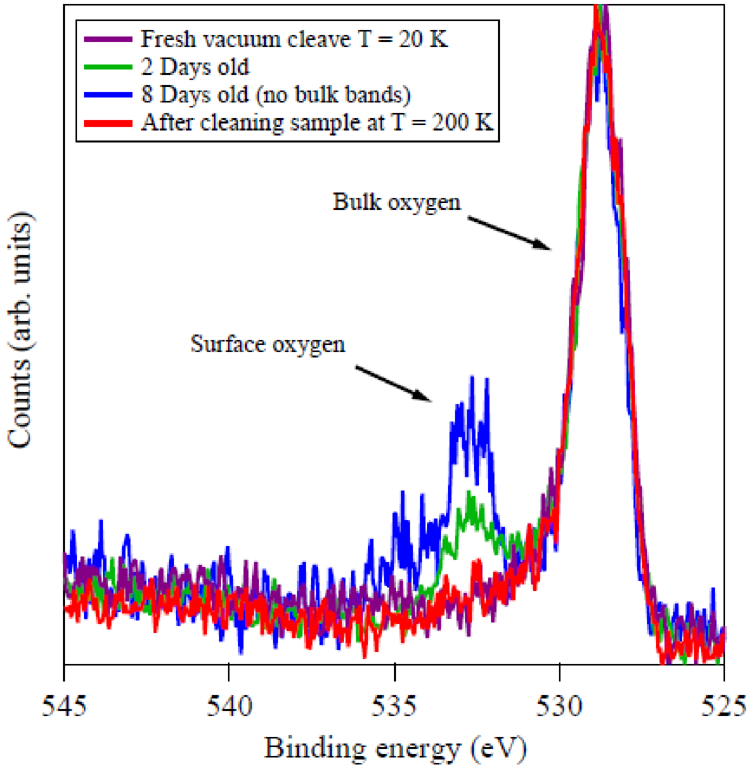


Figure 5.2: (color) The oxygen 1s peaks from Bi2212 at different times after the cleave. A constant background was subtracted from each spectrum to allow direct comparison. The peak derived from bulk oxygen is stable over time, while the surface oxygen peak grows as more oxygen sticks to the cold surface.

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HC Hsu, Ph.D. Thesis NTNU(2010)
Koralek, U. Colorado Ph.D. Thesis (2007)

Light sources and terminology

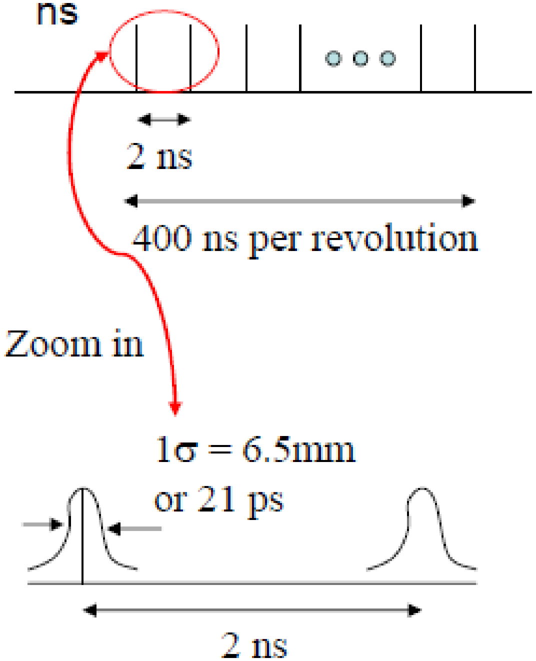
- **Ultraviolet Photoemission Spectroscopy (UPS)**
  - UV He lamp (21.2 eV, 40.8 eV)
  - Laser : 6 eV (BBO), 8 eV (KBBF), 11 eV (gas cell) or HHG (High harmonic generation)
  - Valence band PES, direct electronic state info.

- **X-ray Photoemission Spectroscopy (XPS)**
  (Electron Spectroscopy for Chemical Analysis) (ESCA)
  - X-ray gun (Al: 1486.6 eV, Mg: 1253.6 eV)
  - core level PE, indirect electronic state info
  - chemical analysis

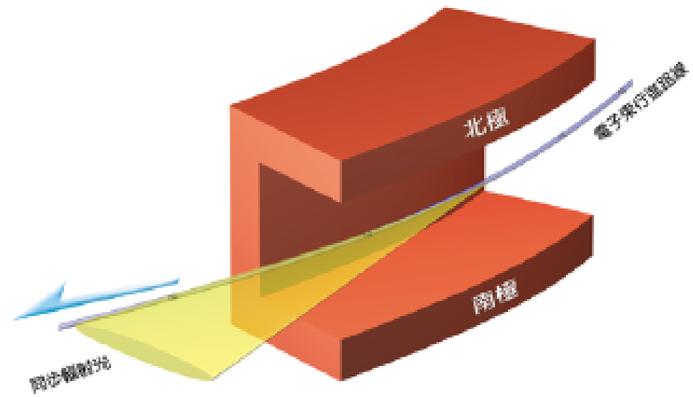
- **Synchrotron radiation**
  - continuous tunable wavelength
  - valance band and core level

Basic Parameters of Taiwan Light Source

- Interval between bunches: 2ns
- Bunch length (1σ@1.6MV): 6.5 mm
- Bunch duration (1σ): 21 ps
- SC Cavity length: 24 cm
- Flight time through SC cavity: 0.8 ns



- Bunch current (180/200 filling to I_{avg}=300 mA): 1.67 mA/bunch or $4.17*10^9$ electrons/bunch



- Critical energy of SR

$$Ec(keV) = 0.665\ E^2\ (GeV)\ B(T)$$

8 GeV X-Ray Free Electron Laser Facility at SPring-8

European XFEL

Taiwan Photon Source and Taiwan Light Source

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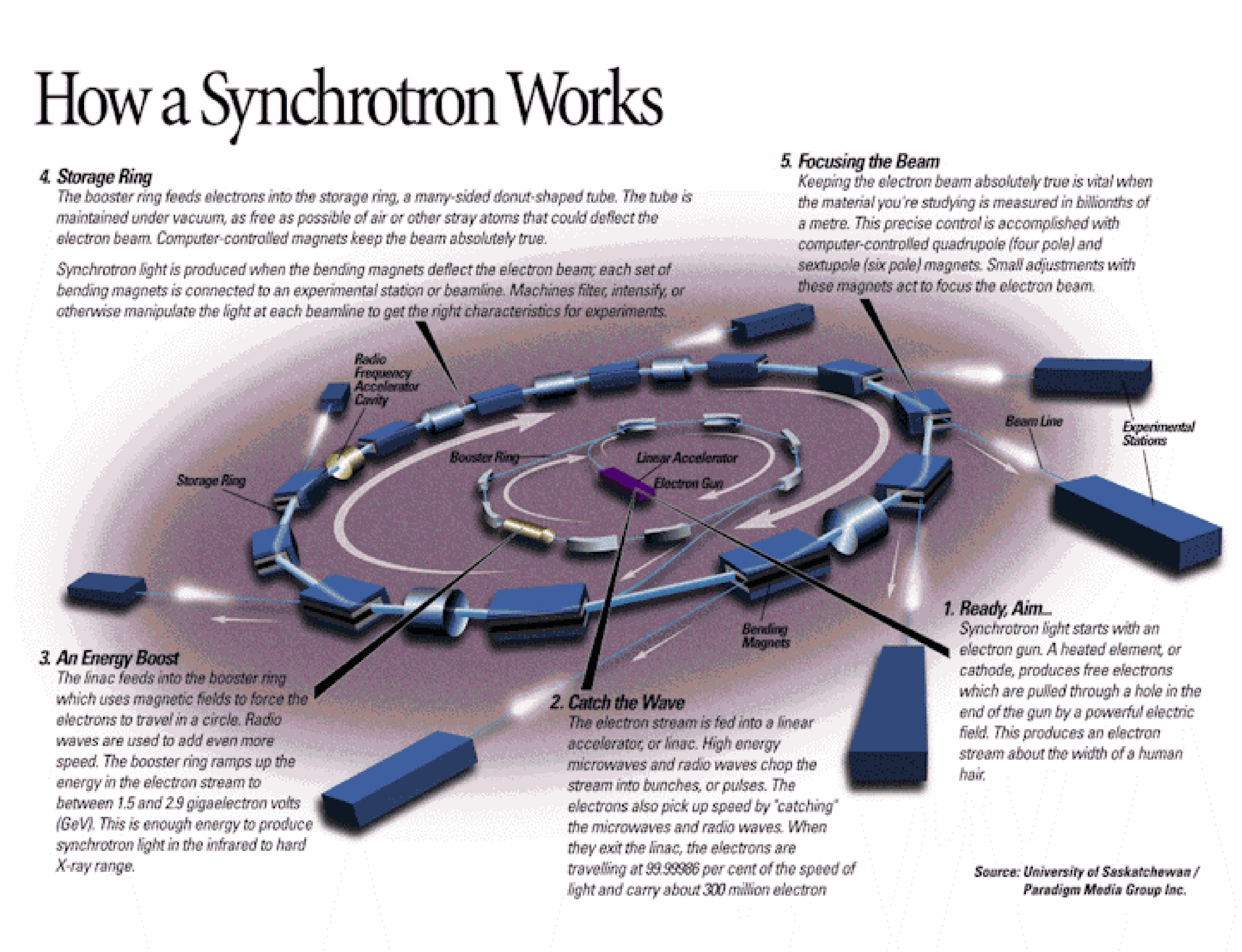
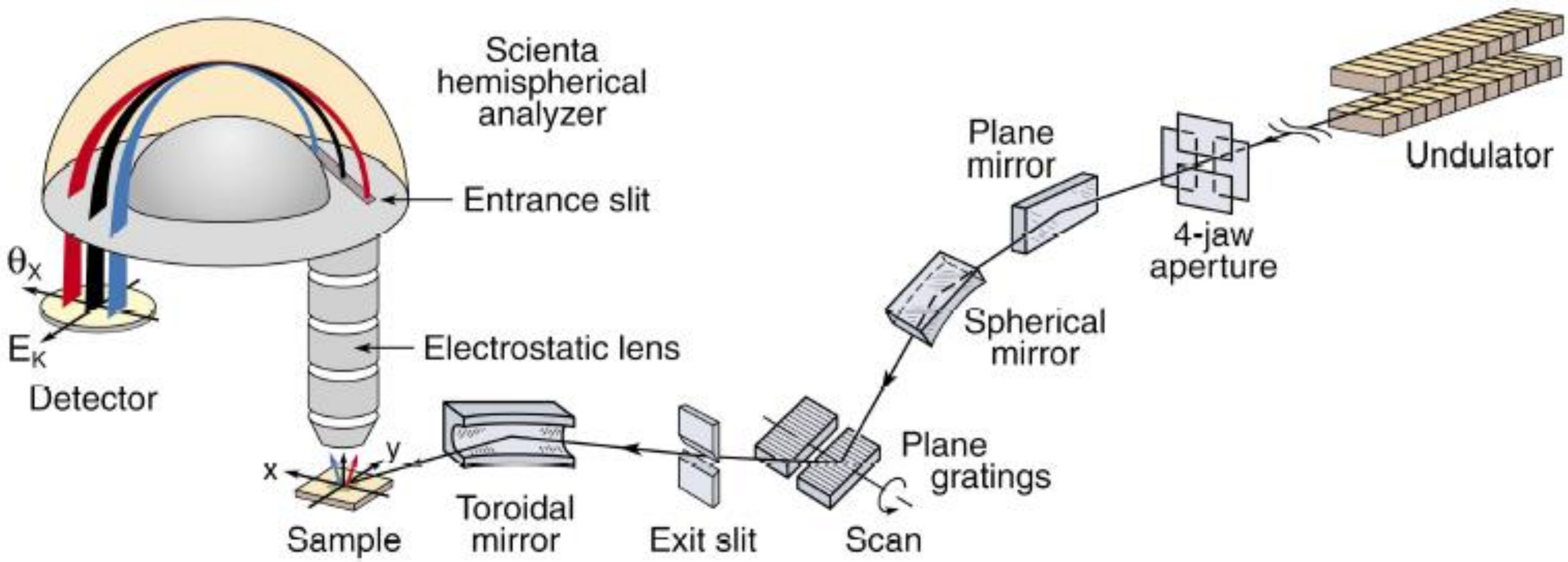
Light sources and terminology

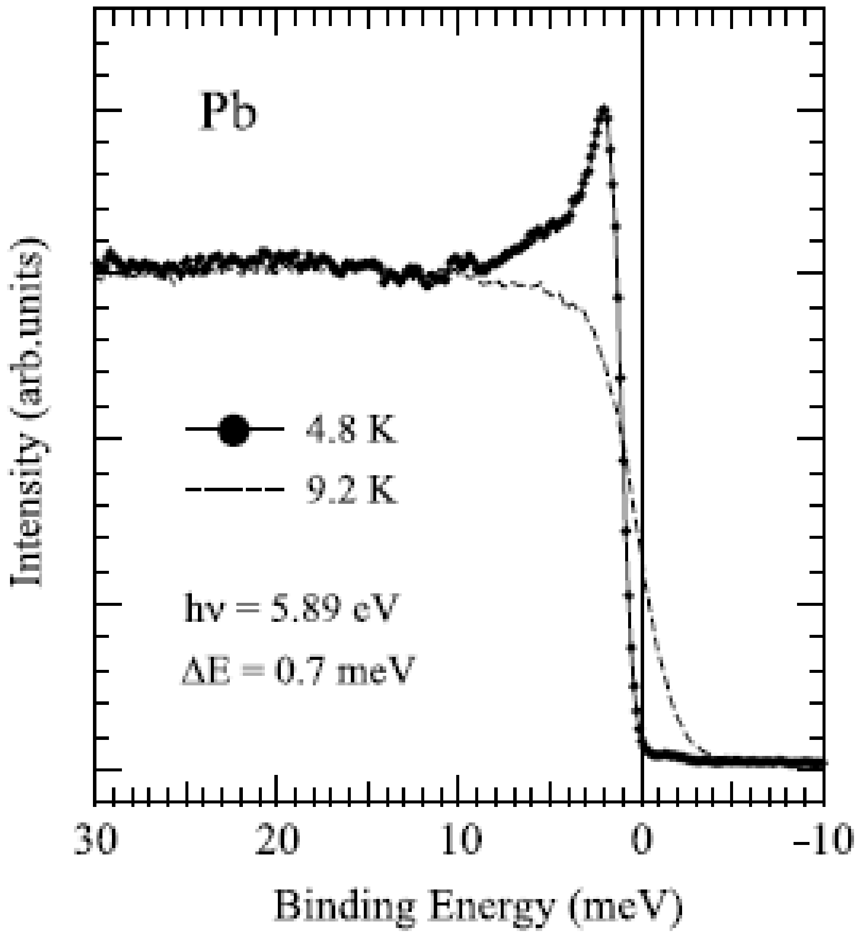


Light sources and terminology



Scientific Case : High- Tc Superconductor

Superconducting gap



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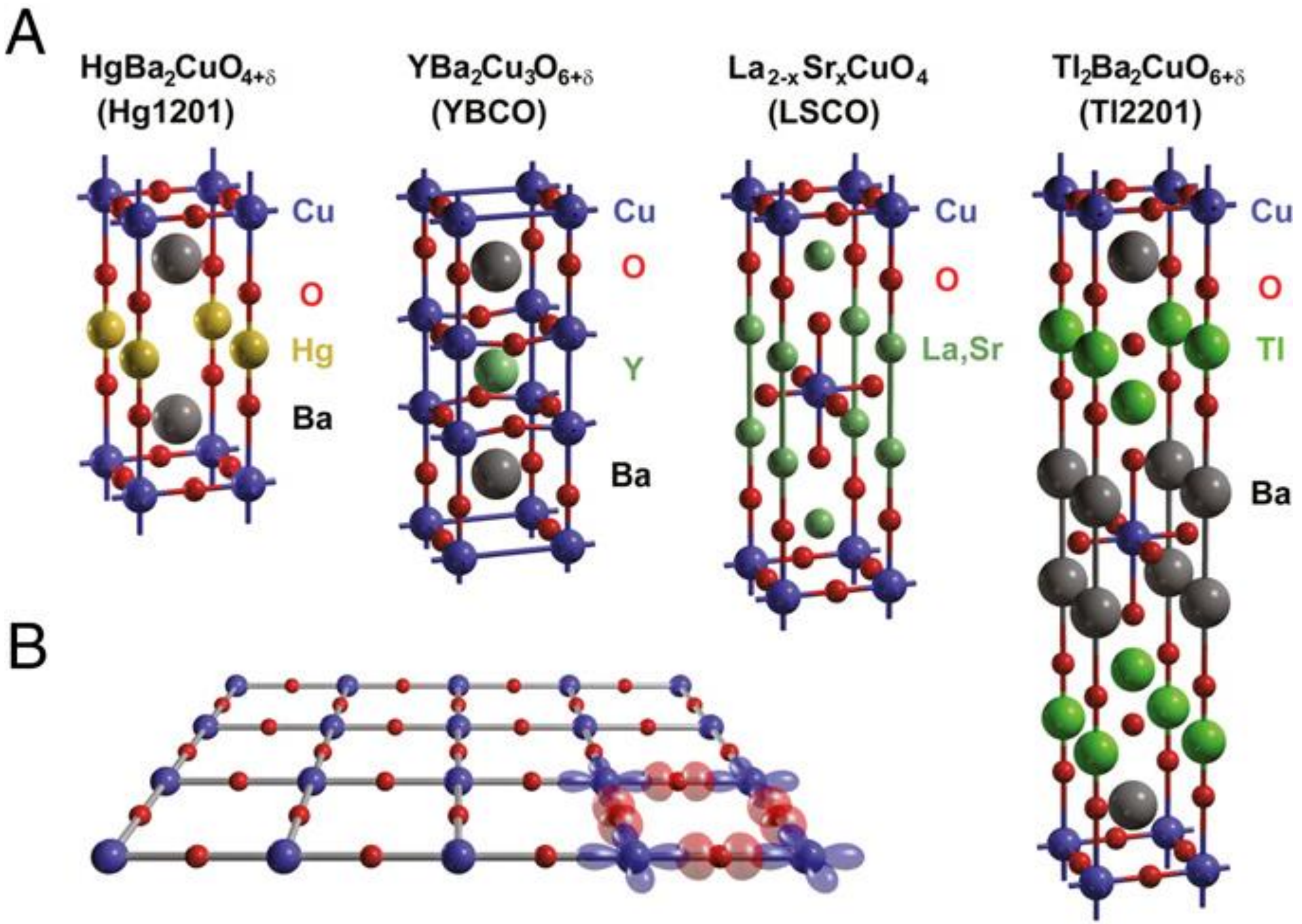
Hufner, Very high resolution photoelectron spectroscopy

Scientific Case : High- Tc Superconductor



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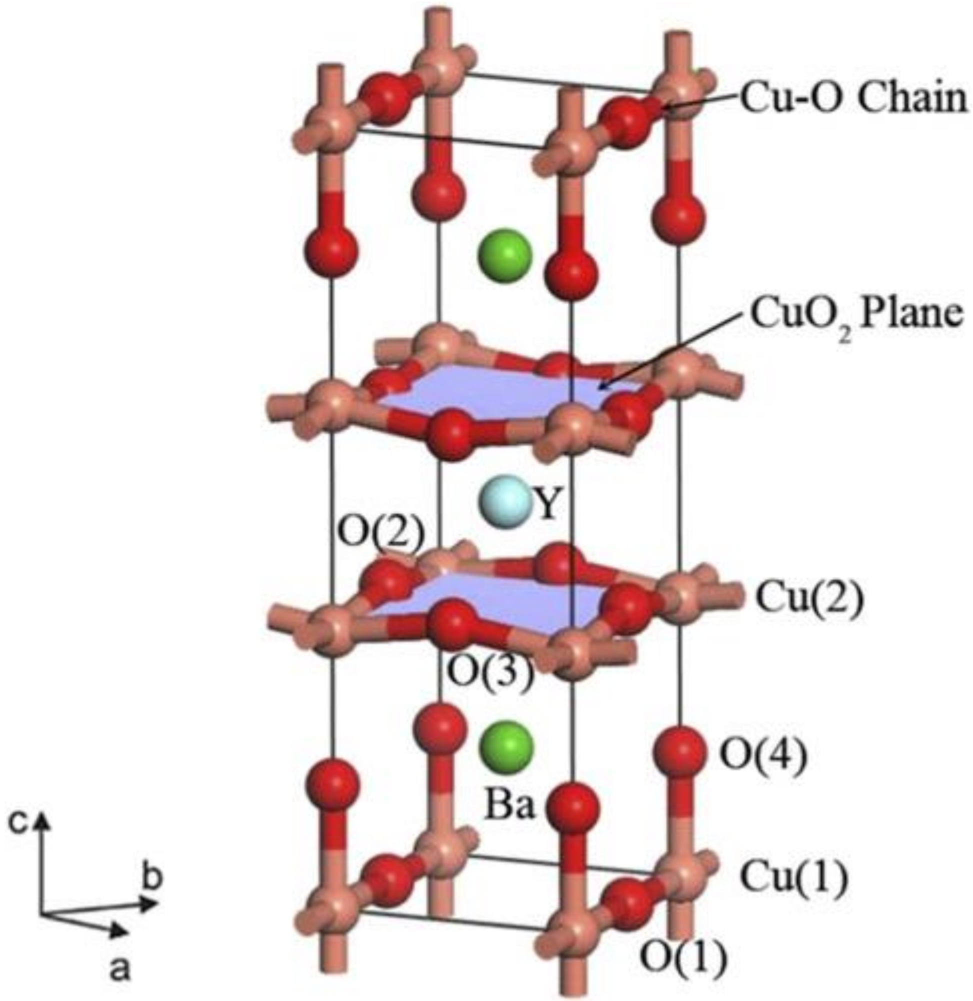
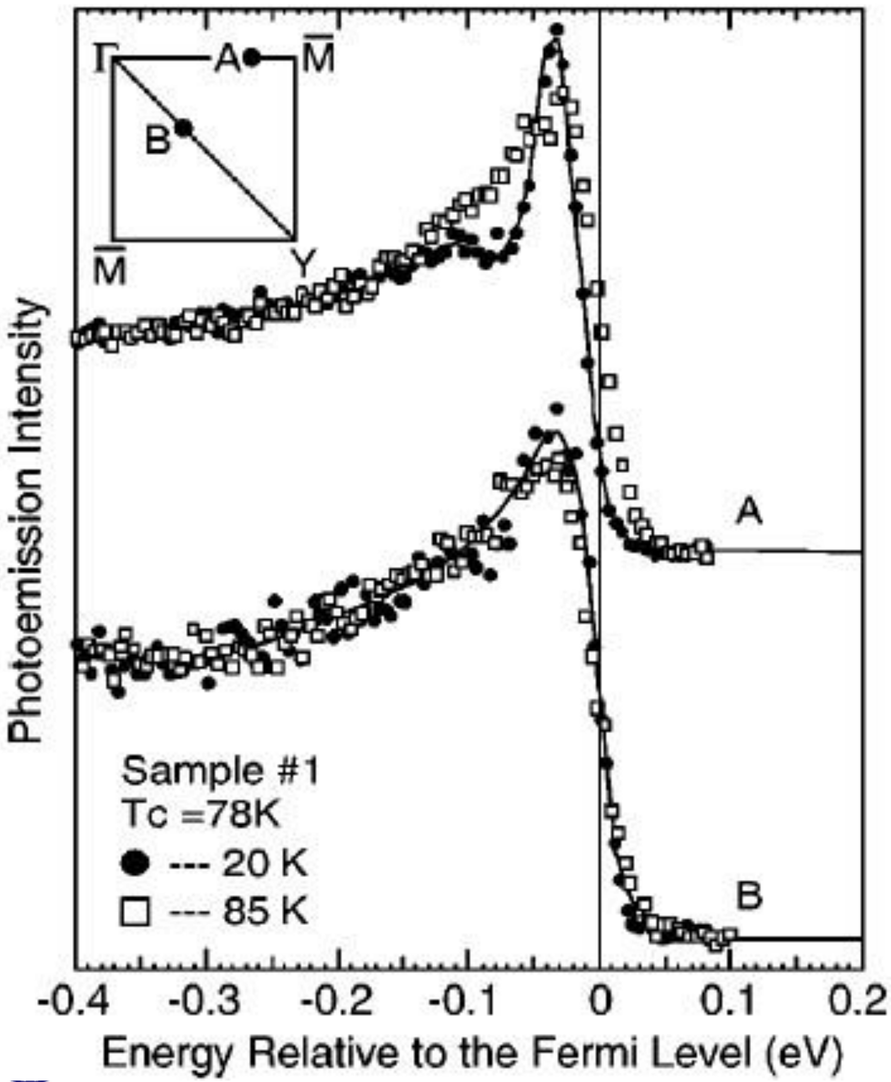
Scientific Case : High- Tc Superconductor



Scientific Case : High- Tc Superconductor

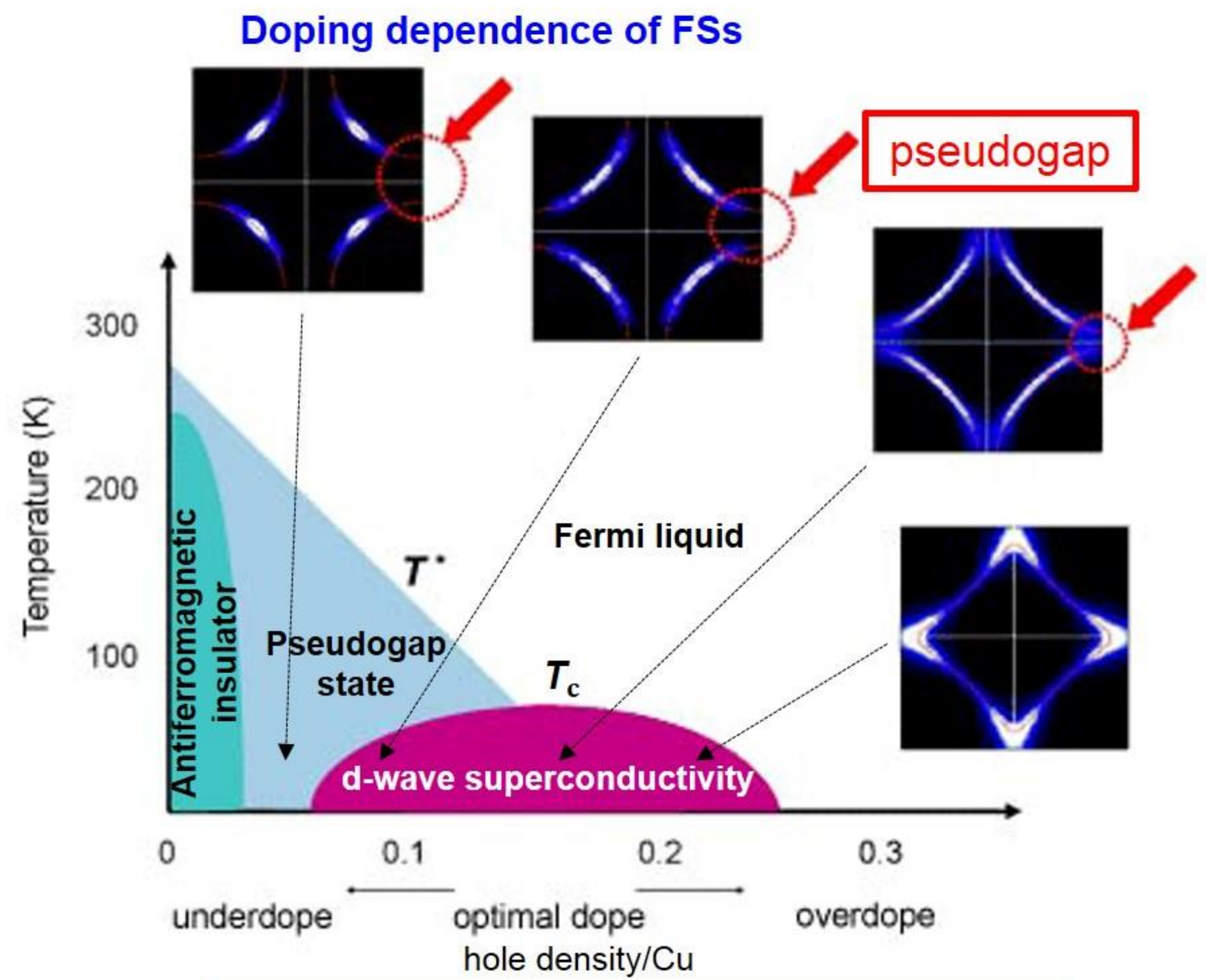


Overdoped Bi-2212 sample at two different momenta in the Brillouin zone.

These results strongly suggest that the superconducting gap is anisotropic and, in particular, consistent with a d-wave order parameter (*Scalapino, 1995*). Together with the microwave penetration depth results (*Hardy et al., 1993*), this direct evidence for gap anisotropy played a major role in the early debate on the pairing symmetry (*Levi, 1993*).

ZX Shen, PRL (1993)

Scientific Case : High- Tc Superconductor



Phase diagram of hole-doped high-Tc superconductors

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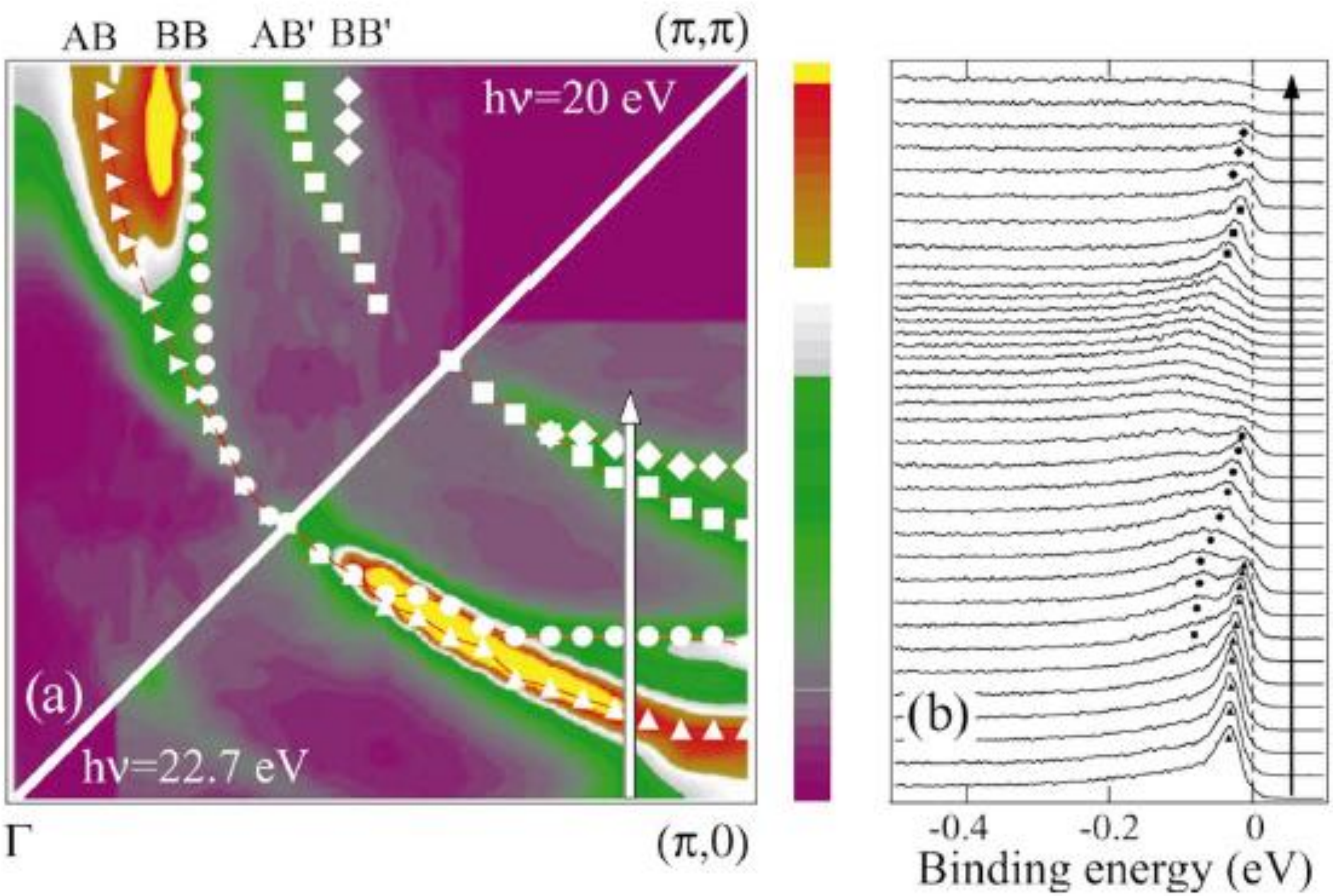
Scientific Case : High- Tc Superconductor

Bilayer band splitting in overdoped Bi-2212 (Tc = 65 K)



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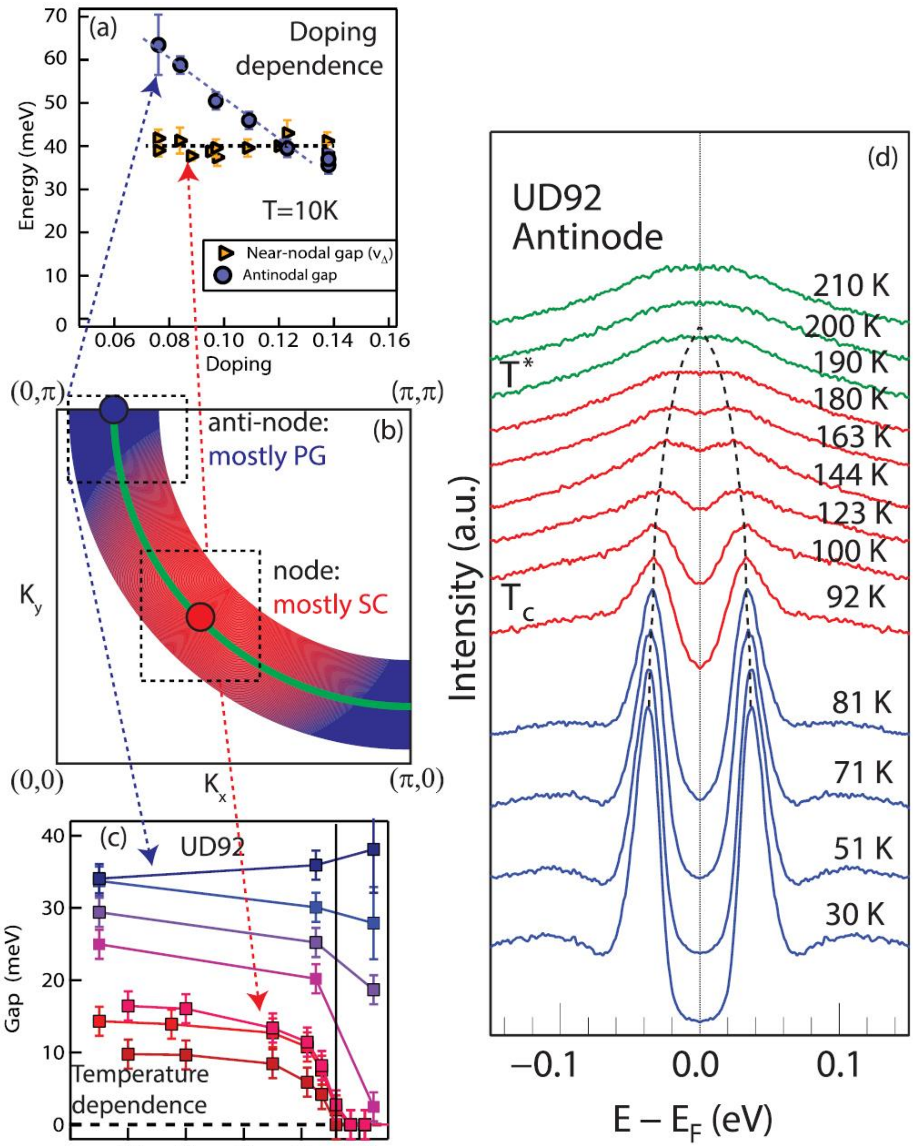
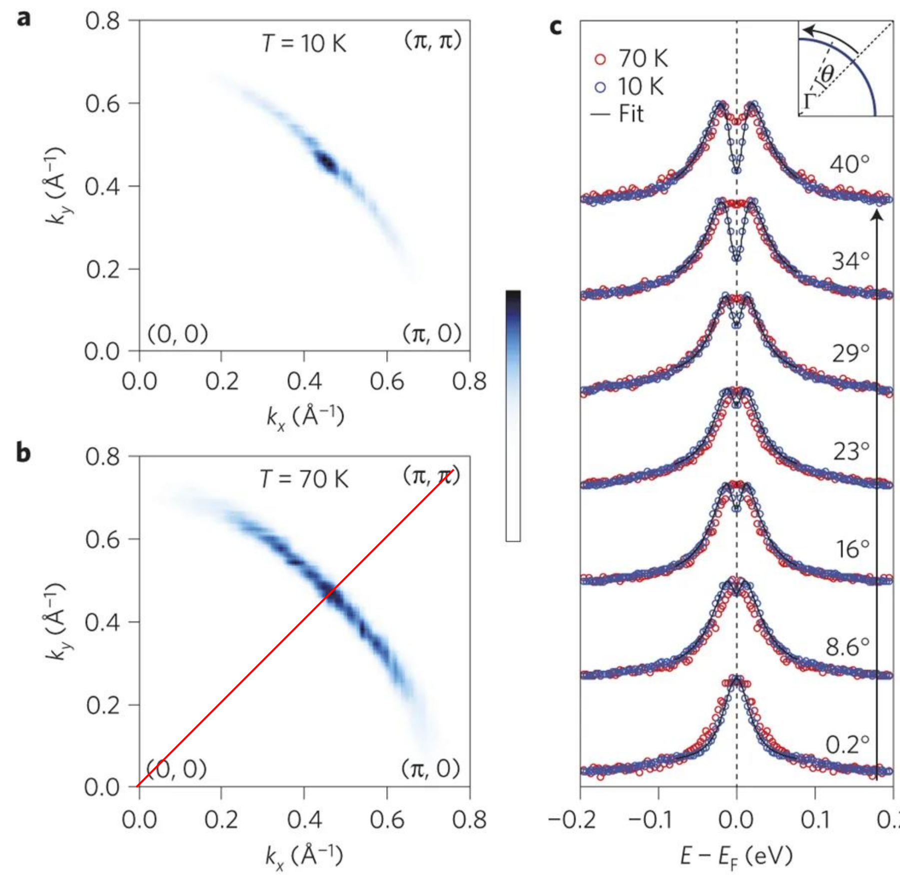
DL Feng, PRL (2001)

Scientific Case : High- Tc Superconductor



Sr_2IrO_4



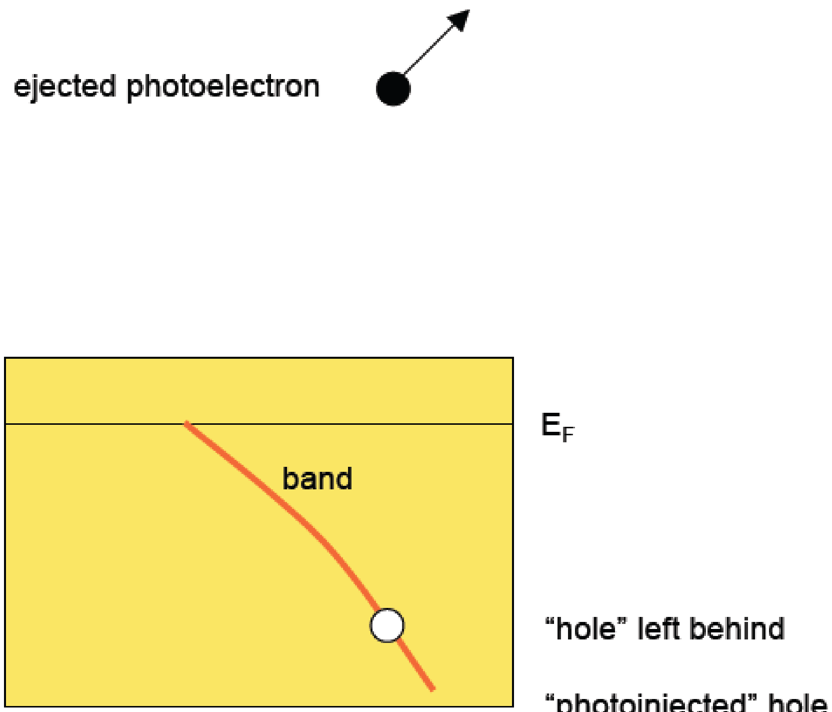
Kim et al., Nat. Phys. (2015)

Scientific Case : High- Tc Superconductor

$$I \propto \sum_{f,i,\mathbf{k}} \left| \langle \phi_f | H_{int} | \phi_i \rangle \right|^2 A(\mathbf{k},E)$$

Matrix element

Spectra function



we believe the properties of the photoelectron reflect the properties of the injected hole

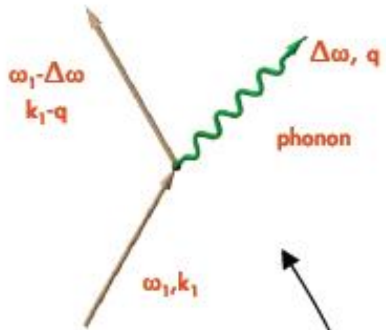
namely,

we can learn about the scattering and lifetime of the injected hole

Eli Rotenberg, ALS summer school

Scientific Case : High- Tc Superconductor

The carriers have a finite lifetime due to absorption and emission of phonons and other excitations

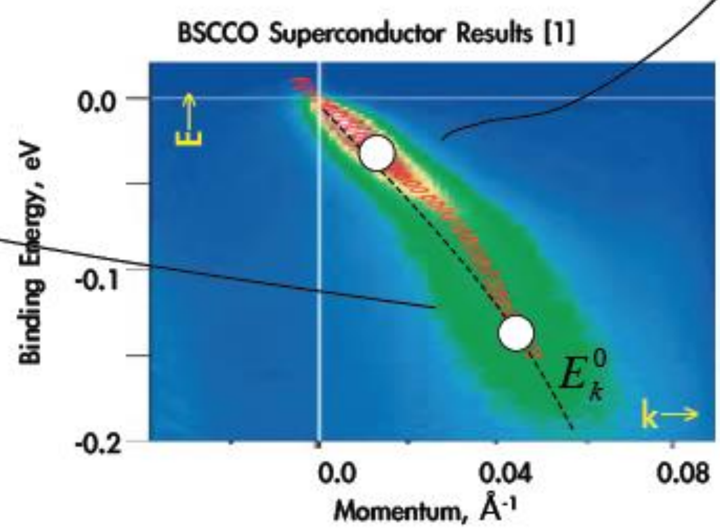


electron emits a phonon : change of energy **ω** and momentum *k*

The measured state is broader in momentum and energy

electron emits and reabsorbs a phonon :Change of mass and slope

The mass of carrier is increased



there is no direct way to probe these processes except through ARPES measurments

Eli Rotenberg, ALS summer school

Scientific Case : High- Tc Superconductor

The quantity determined in ARPES experiments is the single-particle spectral function

$$G(k,\omega)=\frac{1}{\omega-\varepsilon_k-\Sigma(k,\omega)}$$

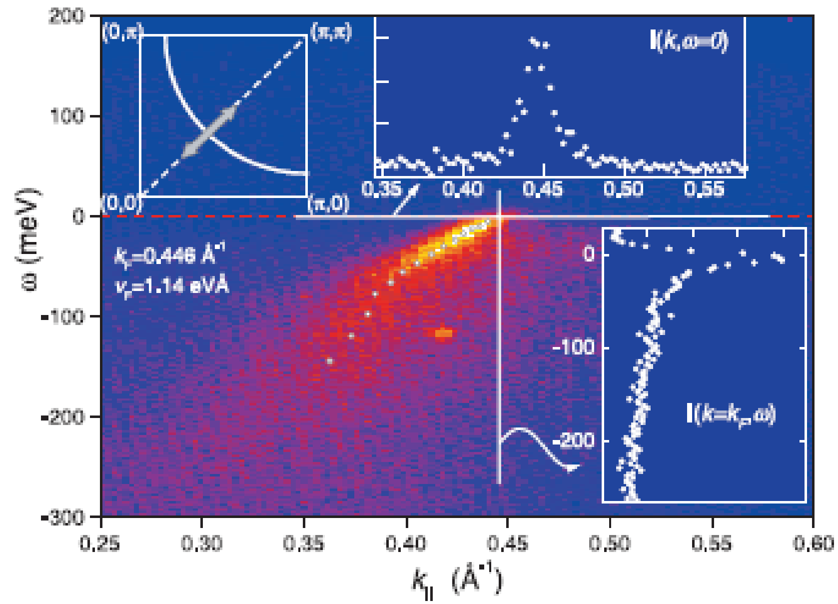
$$A(k,\omega)=\frac{\operatorname{Im}\Sigma(k,\omega)}{[\omega-\varepsilon_k-\operatorname{Re}\Sigma(k,\omega)]^2+[\operatorname{Im}\Sigma(k,\omega)]^2}$$

$$\Sigma=\operatorname{Re}\Sigma+i\operatorname{Im}\Sigma$$

Dispersion:
***E-k* Relation (Velocity; Effective mass etc.)**

Scattering rate (Lifetime)

Optimally doped Bi-2212 cuprate



$$v_k\Delta k=\frac{v_k}{l}=\left|2\operatorname{Im}\Sigma(k,\omega)\right|$$

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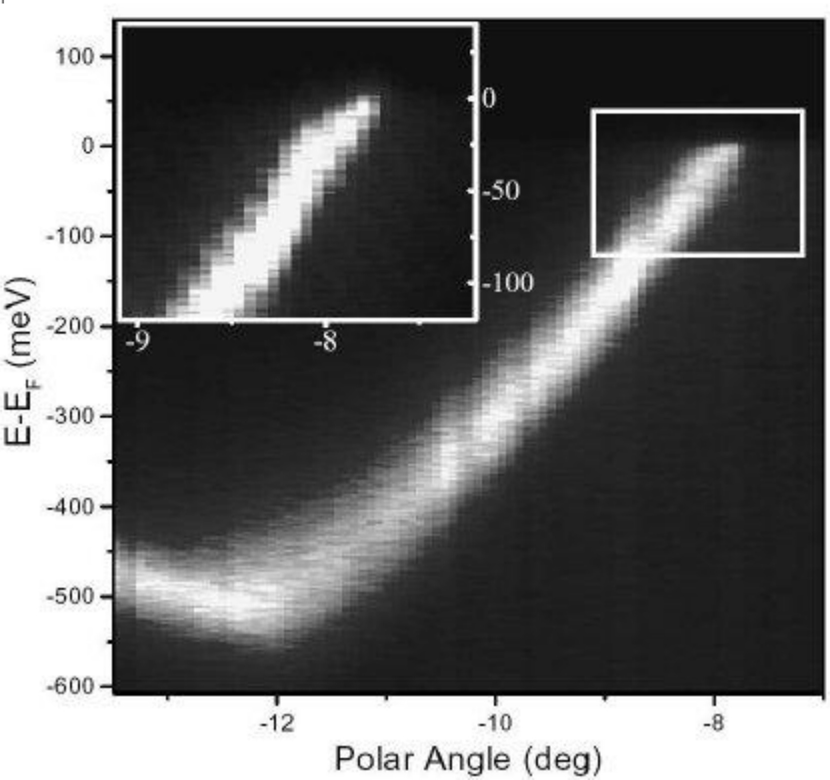
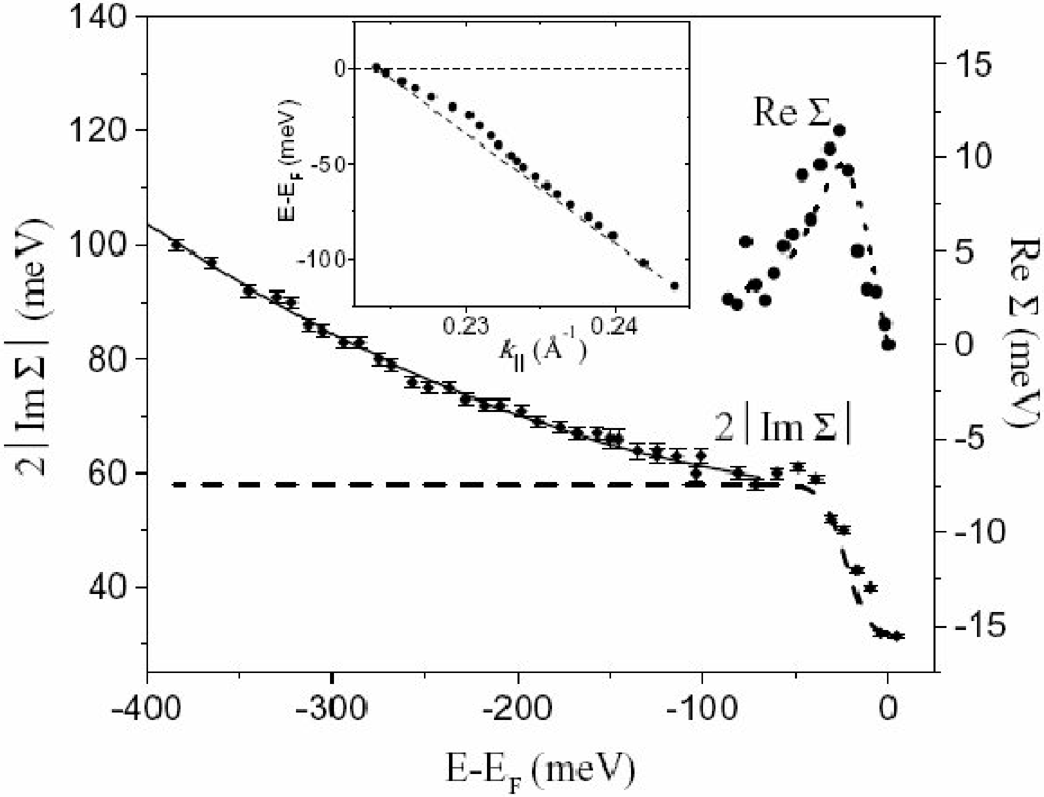
T. Valla et al., Science (2000)

Scientific Case : High- Tc Superconductor

Line shape analysis





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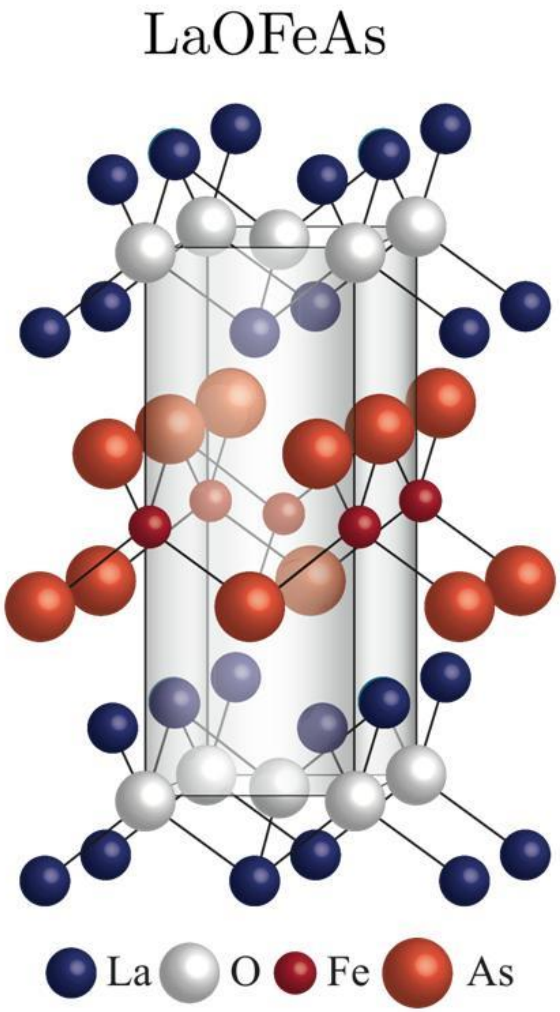
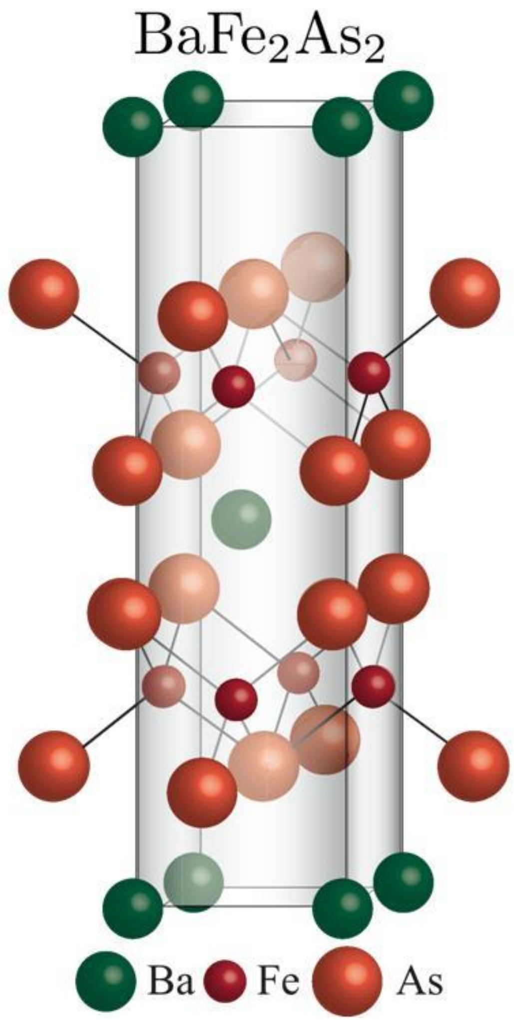
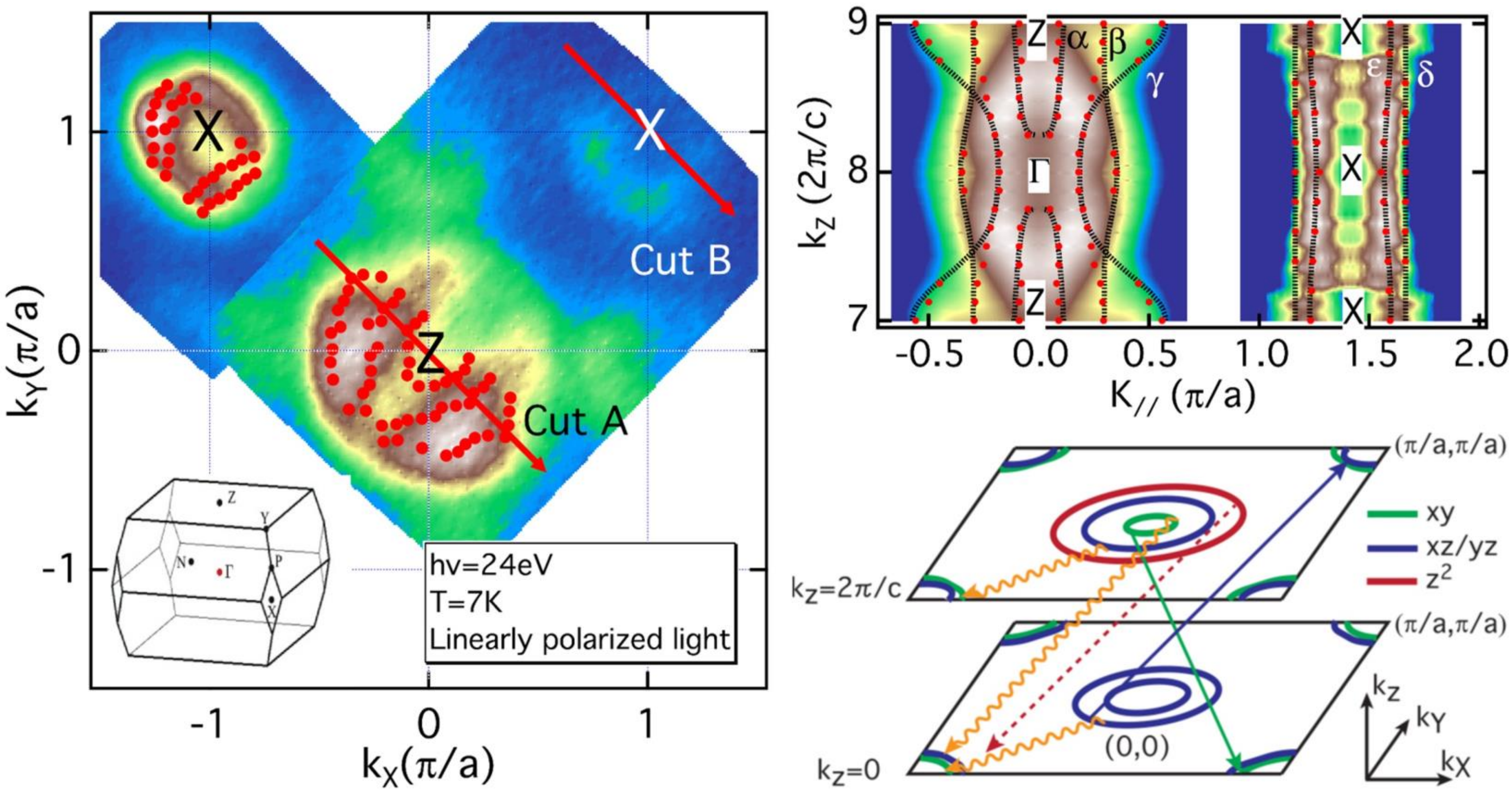
Scientific Case : High- Tc Superconductor





Scientific Case : High- Tc Superconductor

FS shapes of $SrFe_2(As_{0.65}P_{0.35})_2$ and
k_z dependence of the FSs

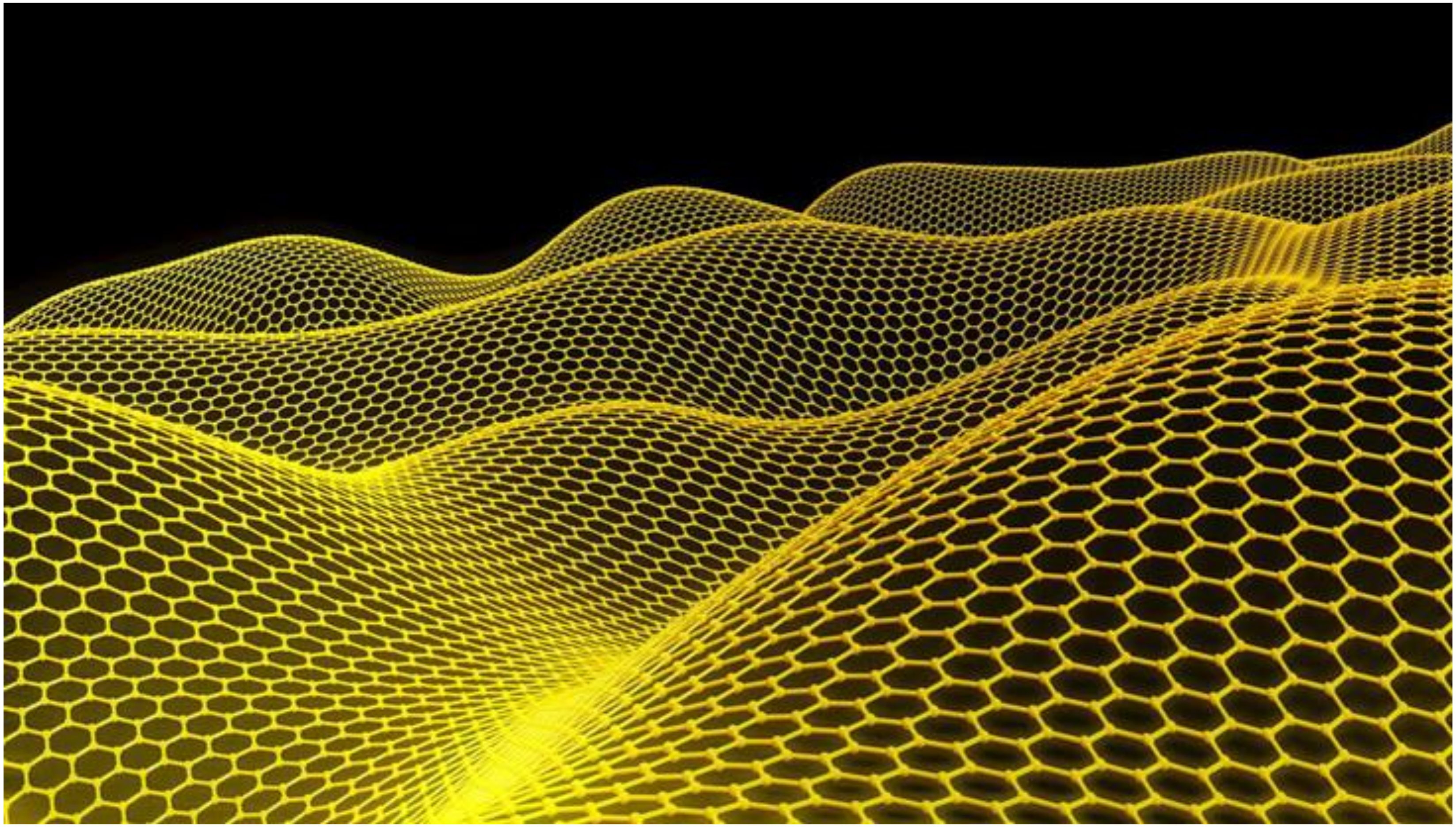


$$k_{\perp} = 0.5123\sqrt{(E_{kin}\cos^2\theta + V_0)}$$

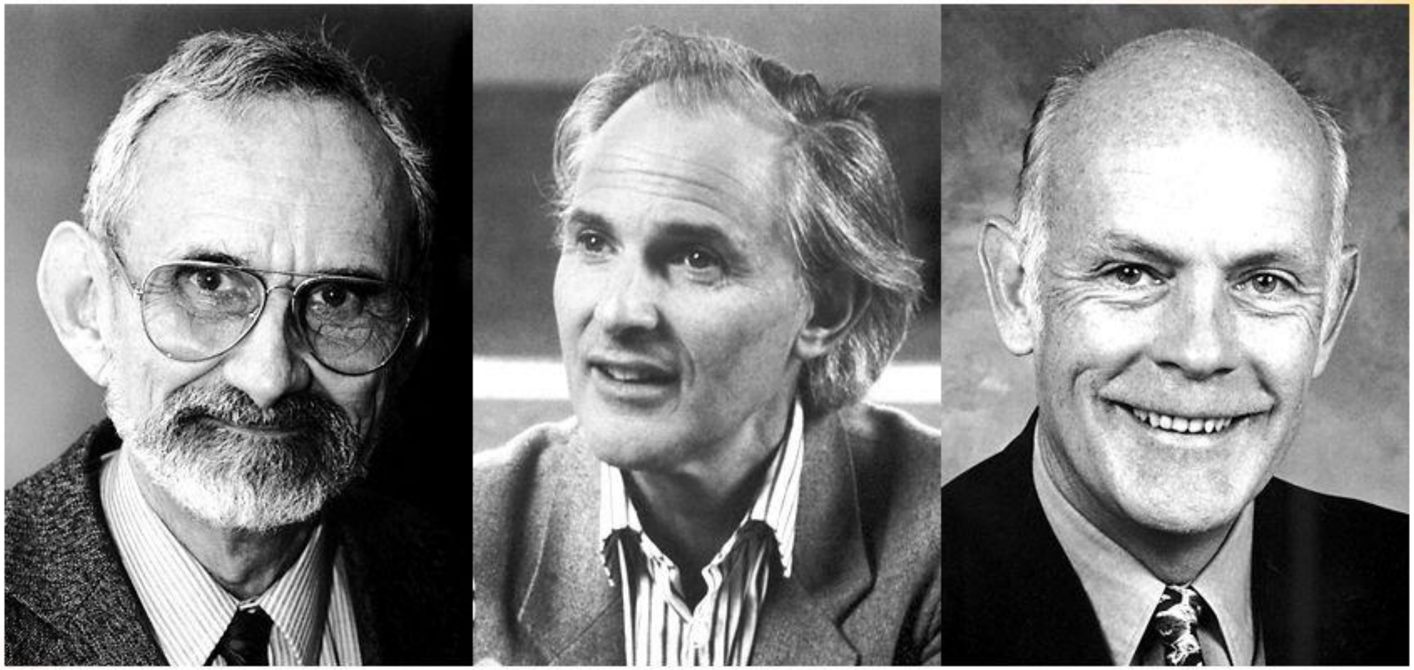
$$k_{//} = 0.5123\sqrt{E_{kin}}\sin\theta$$

Suzuki et al., PRB (2017)

Scientific case : Graphene



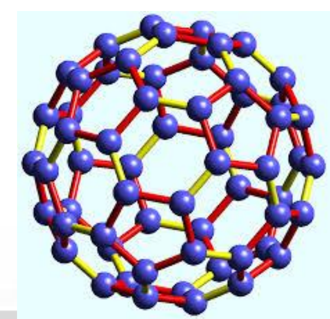
Scientific Case : Graphene



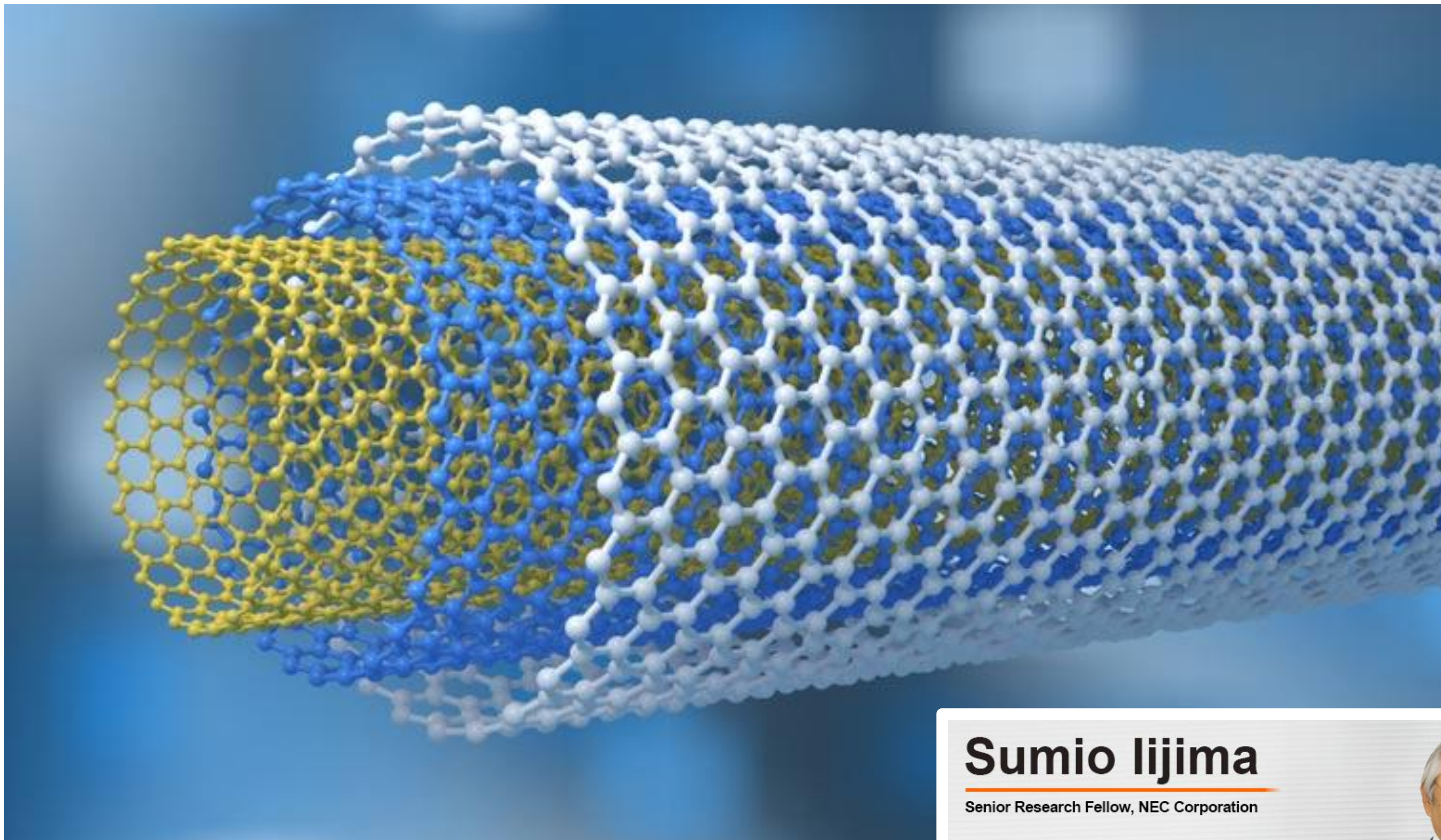
R.F. Curl H.W. Kroto R.E. Smalley

Discovered C_{60} in 1985

Awarded Nobel prize for Chemistry in 1996



Scientific Case : Graphene



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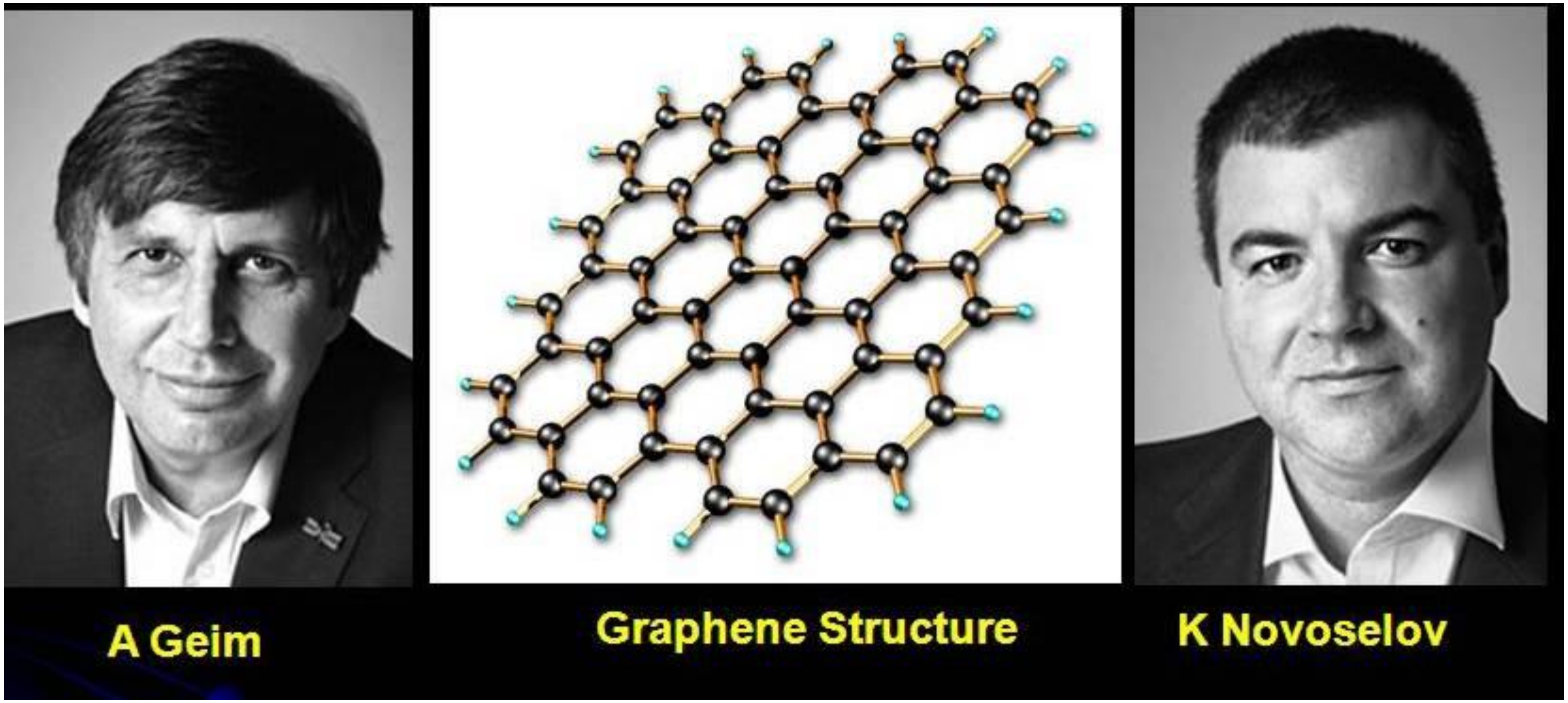
Scientific Case : Graphene



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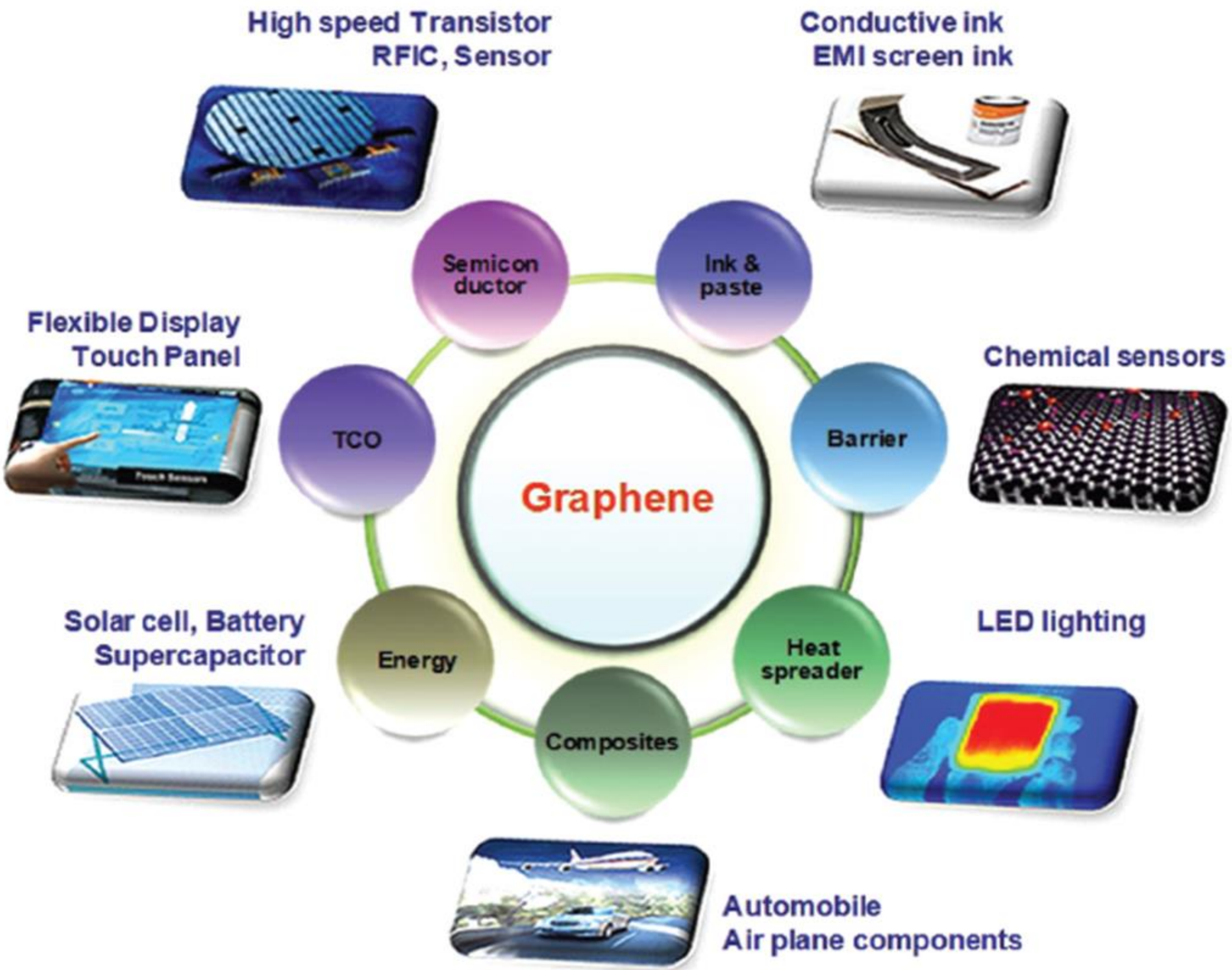
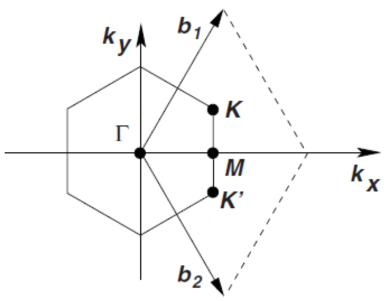
Scientific Case : Graphene

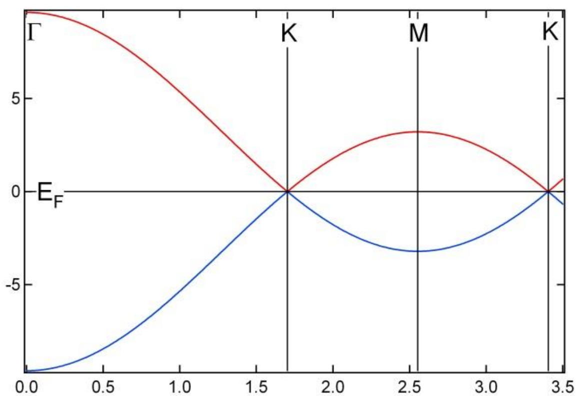


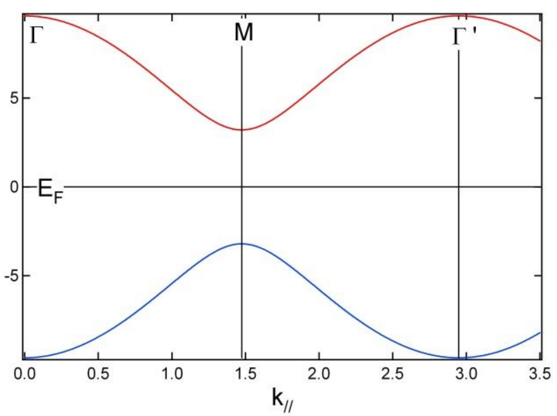
Scientific Case : Graphene

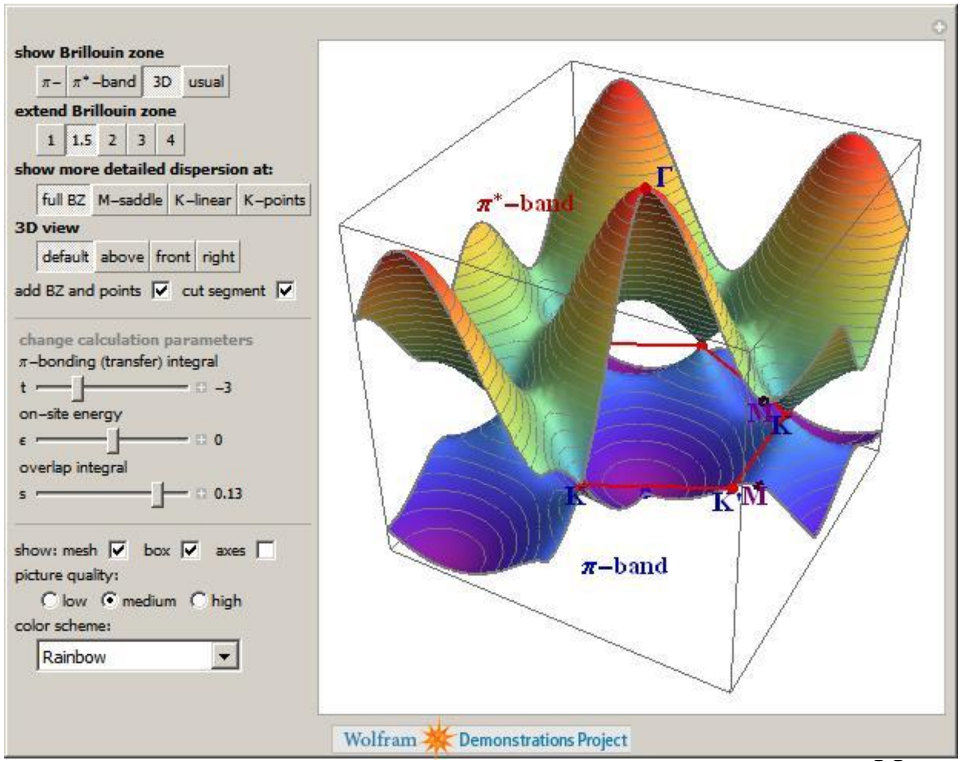


$$\boldsymbol{b}_1 = \frac{2\pi}{3a}(1, \sqrt{3}), \quad \boldsymbol{b}_2 = \frac{2\pi}{3a}(1, -\sqrt{3})$$

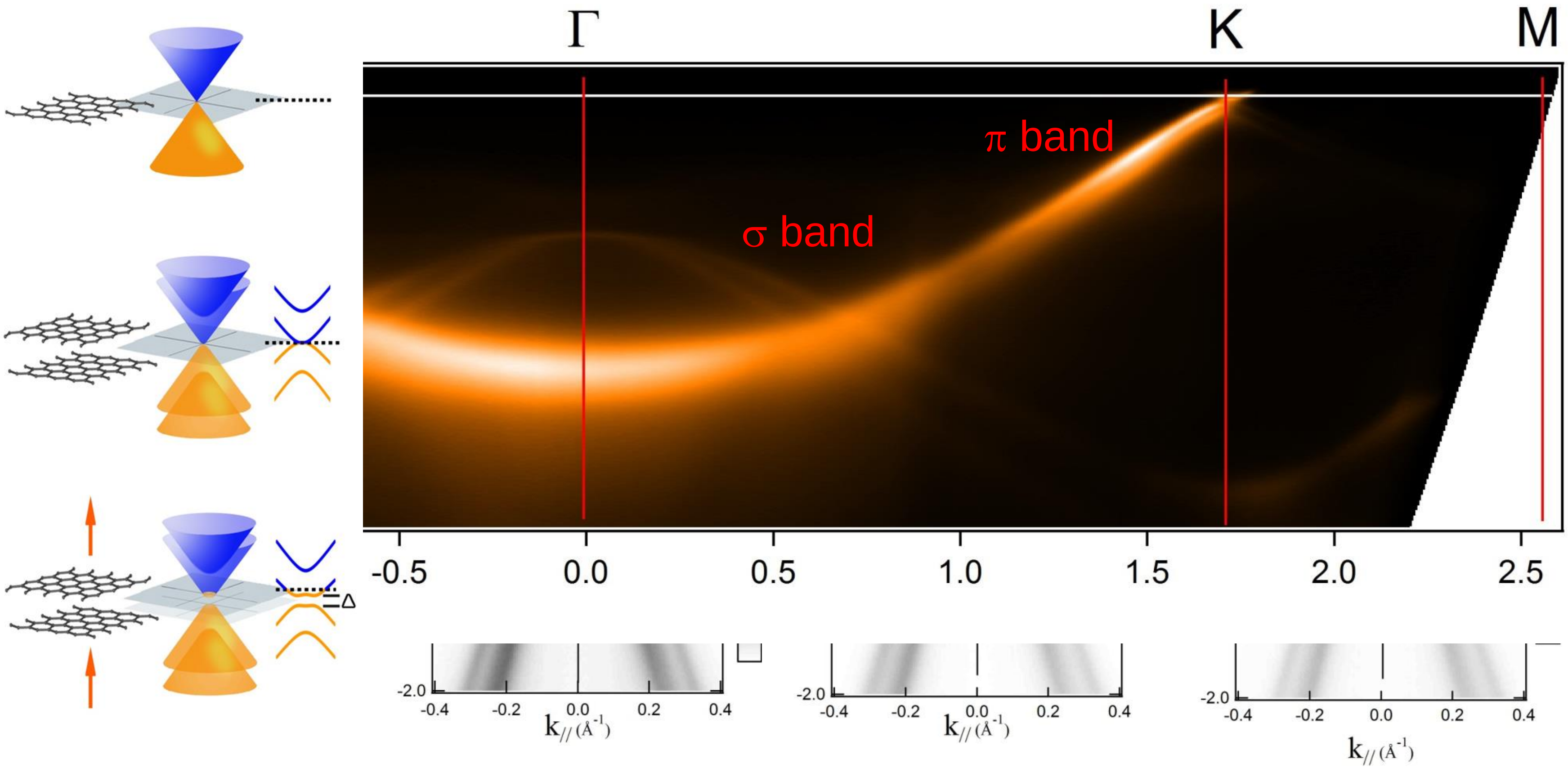
$$\boldsymbol{K} = \left(\frac{2\pi}{3a}, \frac{2\pi}{3\sqrt{3}a}\right), \quad \boldsymbol{K}' = \left(\frac{2\pi}{3a}, -\frac{2\pi}{3\sqrt{3}a}\right)$$







What we expect to observe in ARPES



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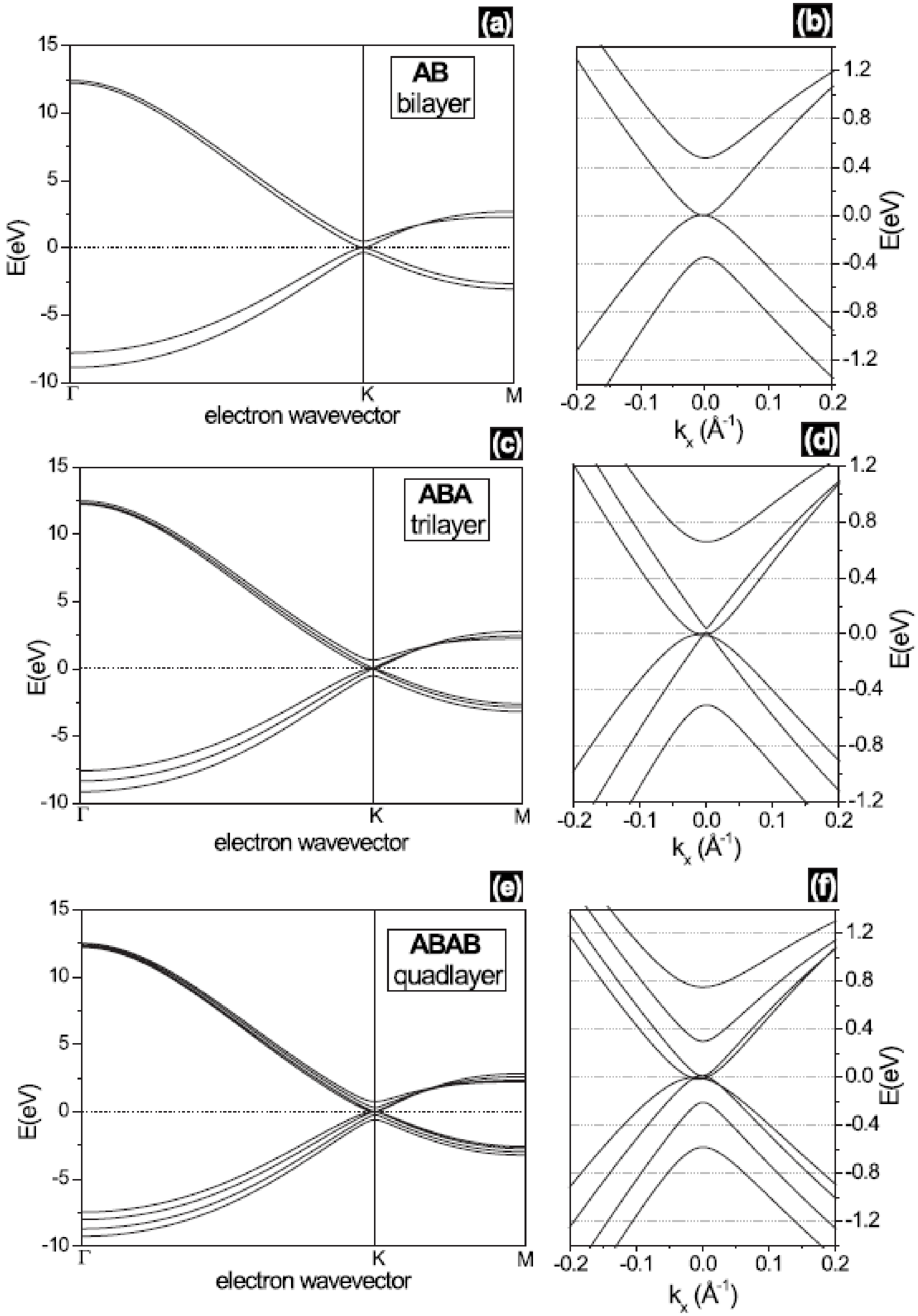
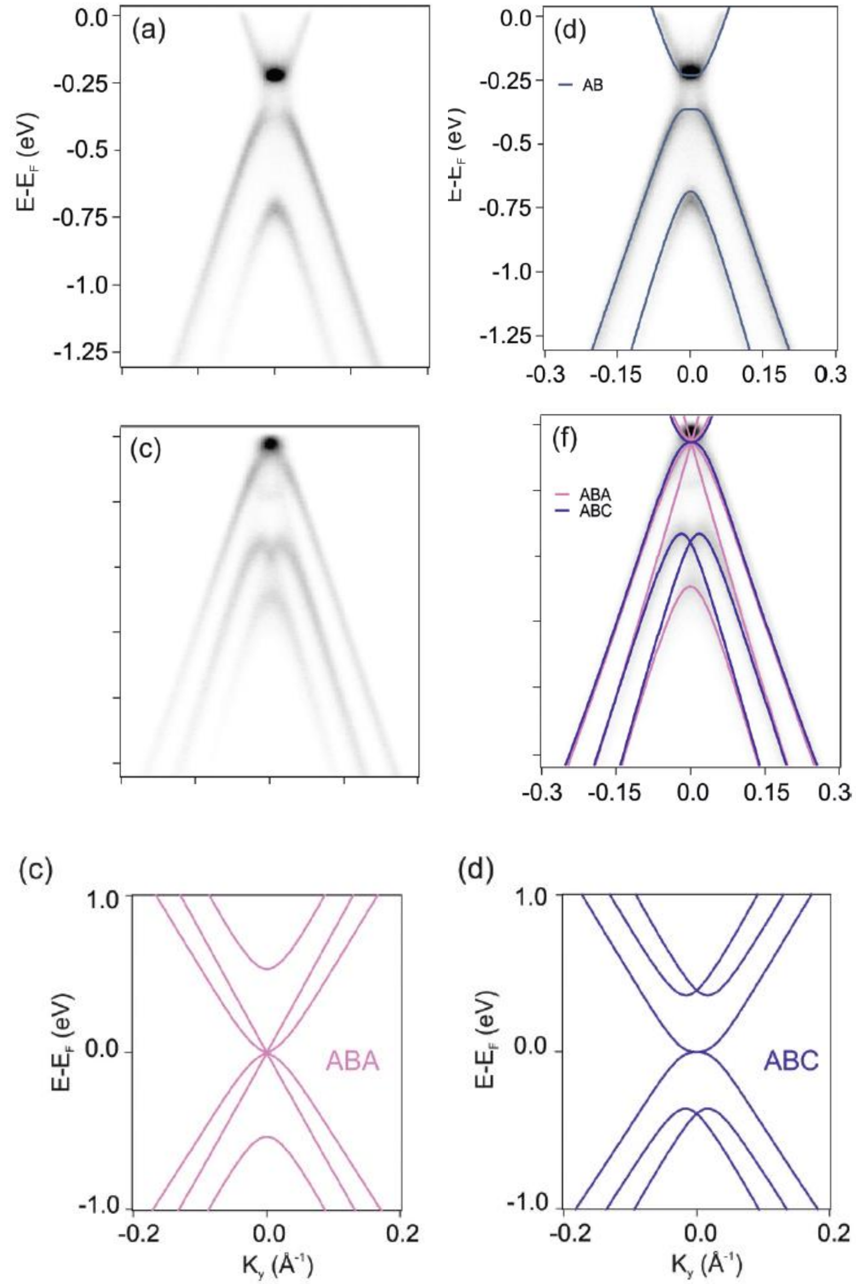
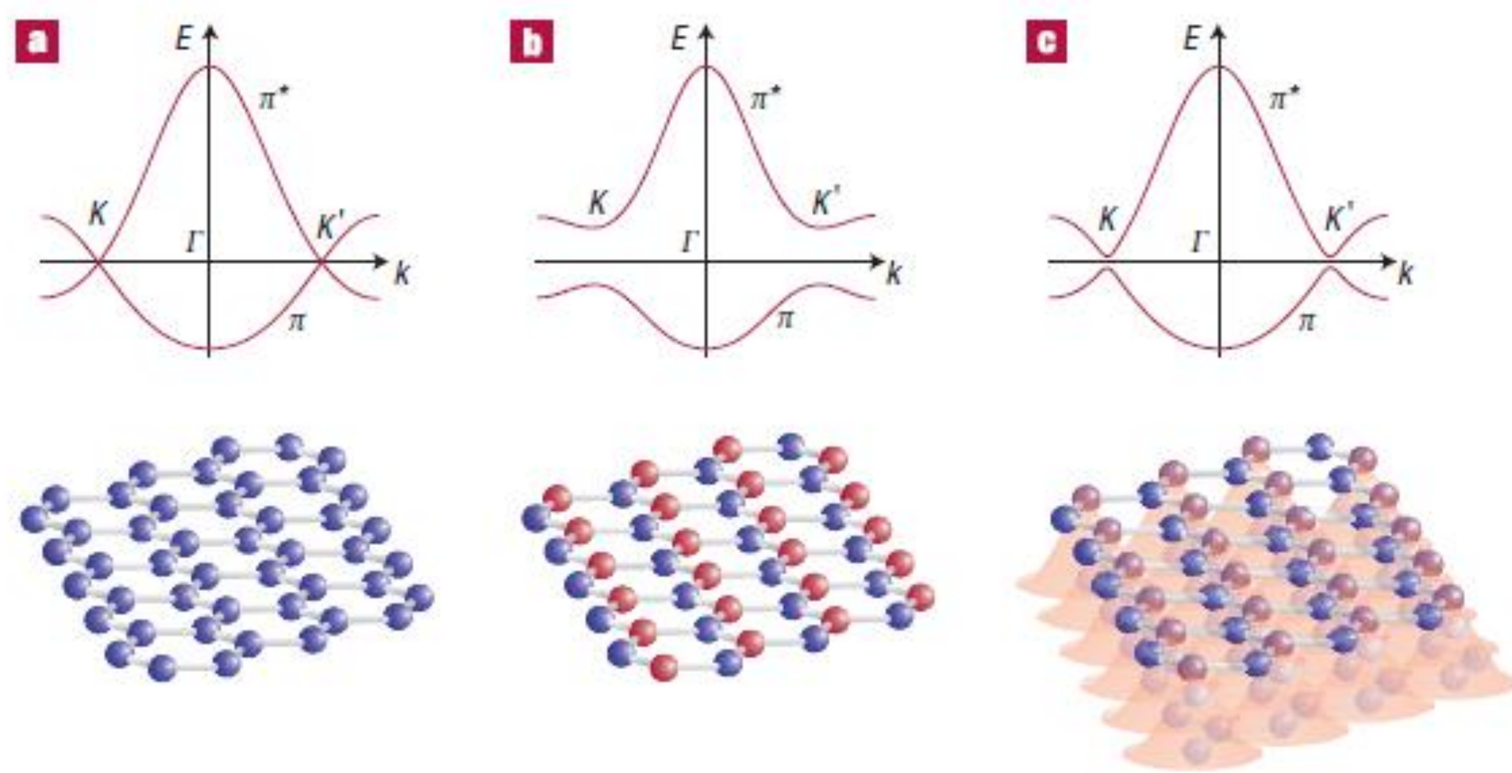




Coletti et al., PRB (2013)

Mind the gap of graphene



Graphene

h-BN

Graphene/SiC

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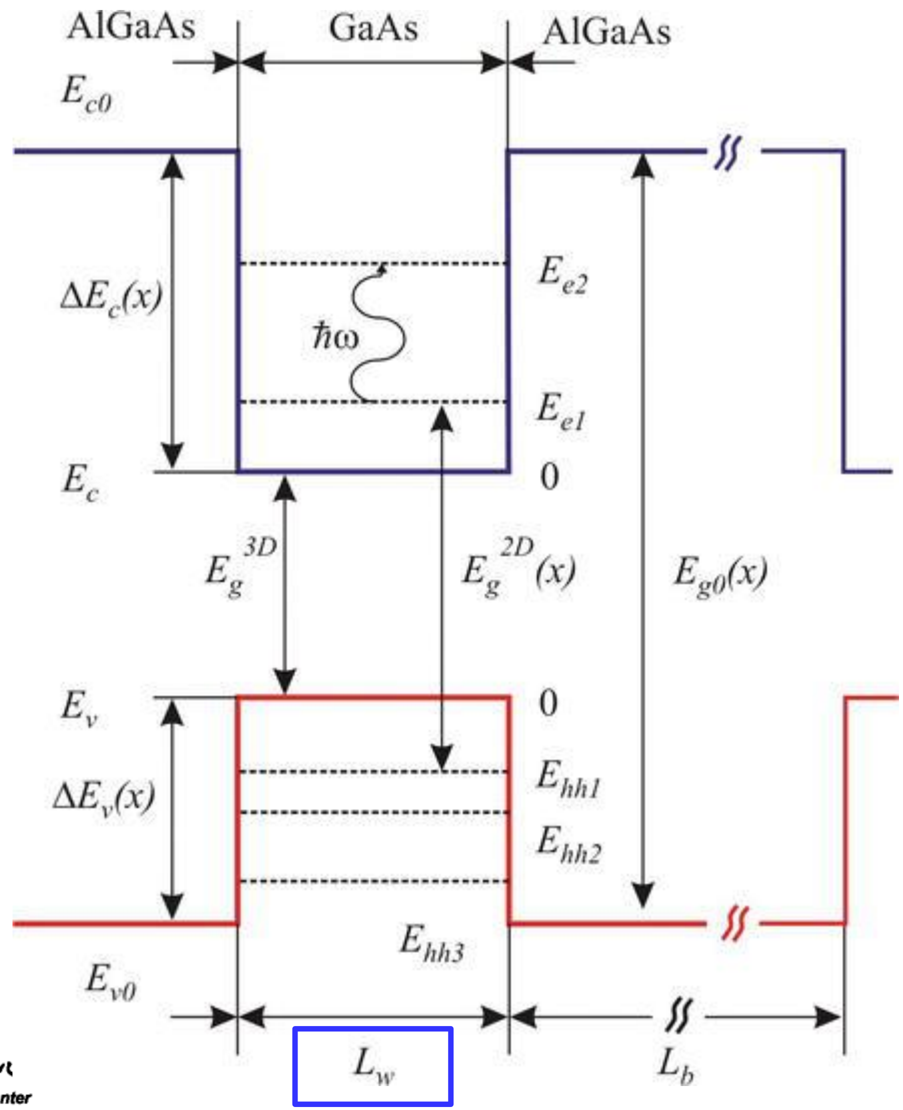
Reduce the thickness of materials : quantum well



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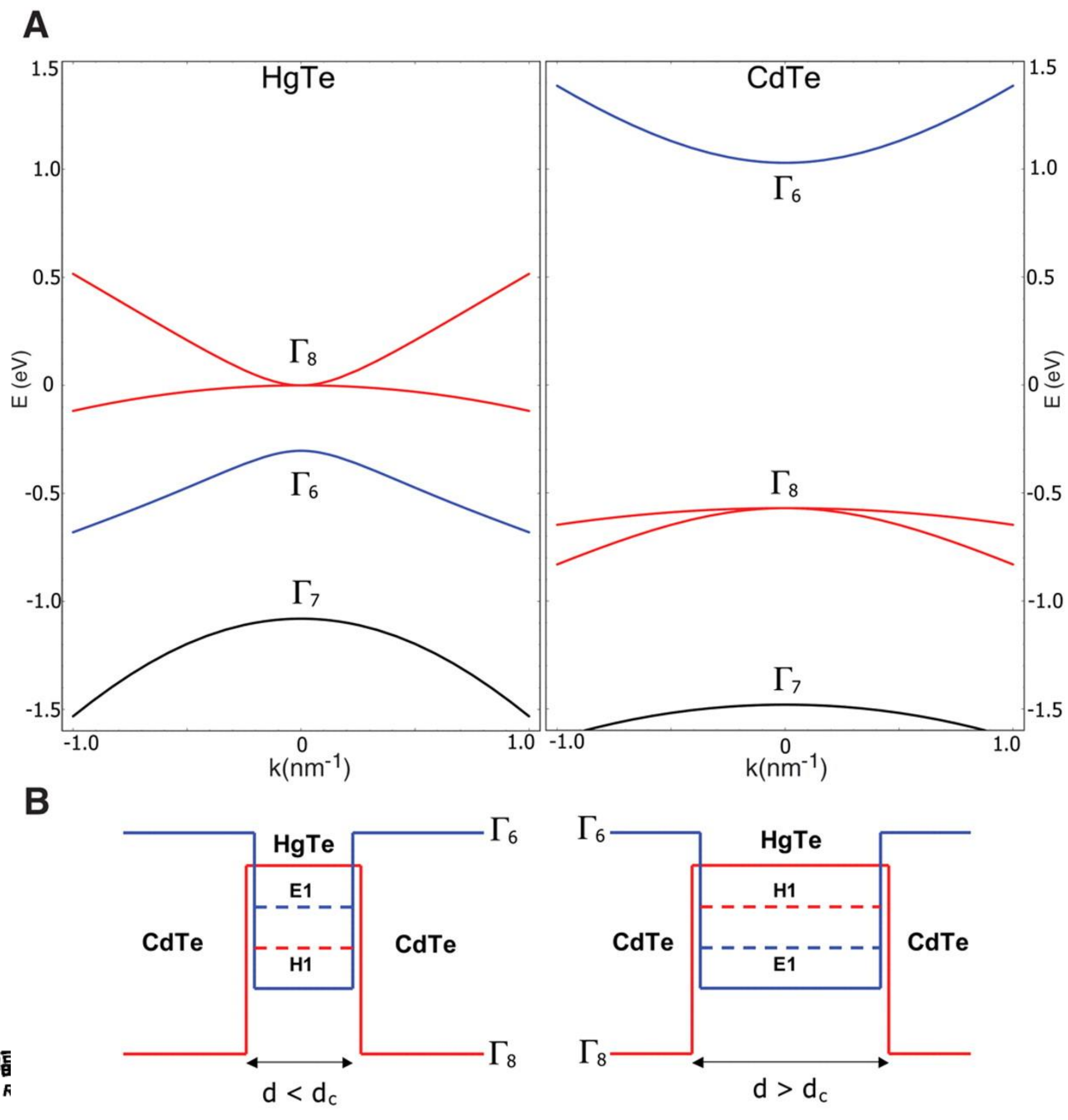
Quantum well : quantum spin Hall effect (QSHE)



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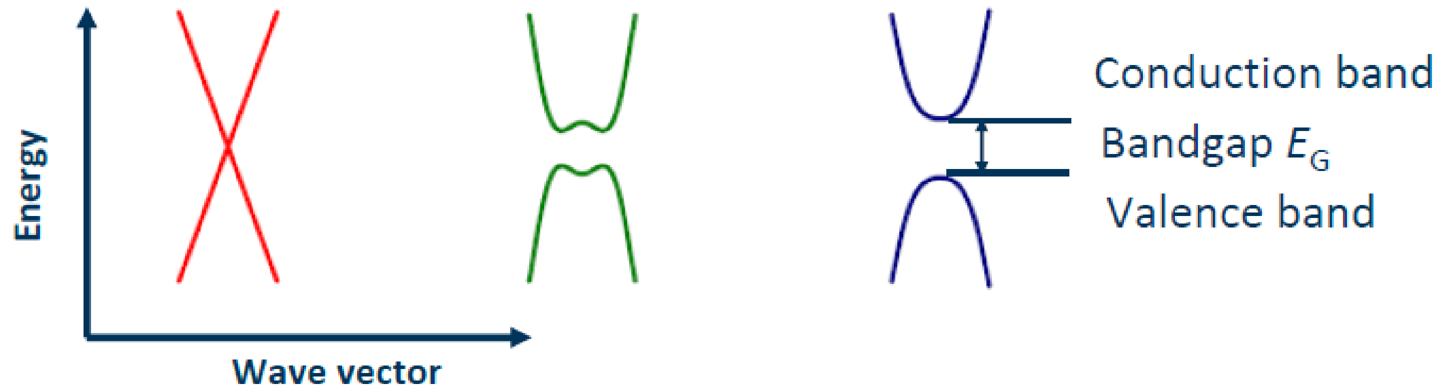
If we reduce the thickness of materials to several atomic layer,

how the band structure change?

Motivation for studying 2D materials

2D Materials – An (Incomplete) Overview



X-enes
- Graphene
- Silicene
- Germanene

No gap, E_G = 0! This is really a pitty, since the missing gap causes serious problems for transistors.

BLG

$E_G \leq 130$ meV Too narrow for logic transistors.

X-enes
- Phosphorene
- Stanene
- GNRs

MX-enes
- Sc_2CF_2
- $TiCO_2$

X-anes
- Graphane
- Silicane
- Germanane

2D TMDs
- MoS_2, $MoSe_2$, $MoTe_2$
- WS_2, WSe_2, WTe_2

etc., etc.

Many of these materials have a gap E_G = 0.5... 2.5 eV, perfect for transistors.

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Frank Schwierz, Nanoscale (2015)

ARPES – Characterization of emergent quantum materials

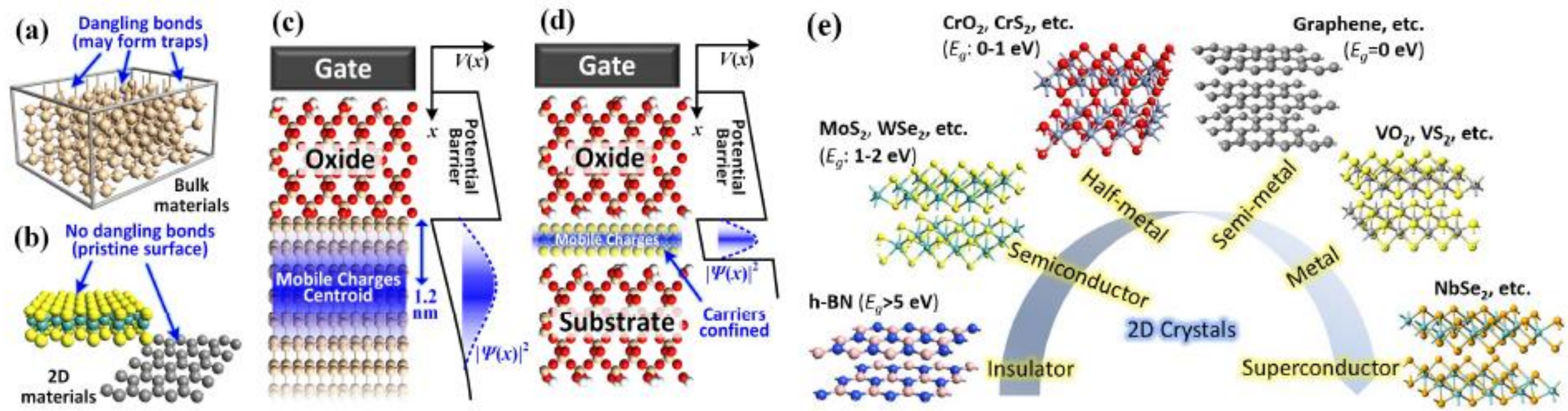

Figure 1. Schematic illustrating advantages of 2D materials: surfaces of **(a)** 3D and **(b)** 2D materials. The pristine interfaces (without out-of-plane dangling bonds) of 2D materials help reduce the interface traps. Mobile charge distribution in **(c)** 3D and **(d)** 2D crystals used as channel materials. The carrier confinement effect in 2D materials leads to excellent gate electrostatics. **(e)** Various types of 2D materials from insulator to superconductor. E_g denotes the band gap.

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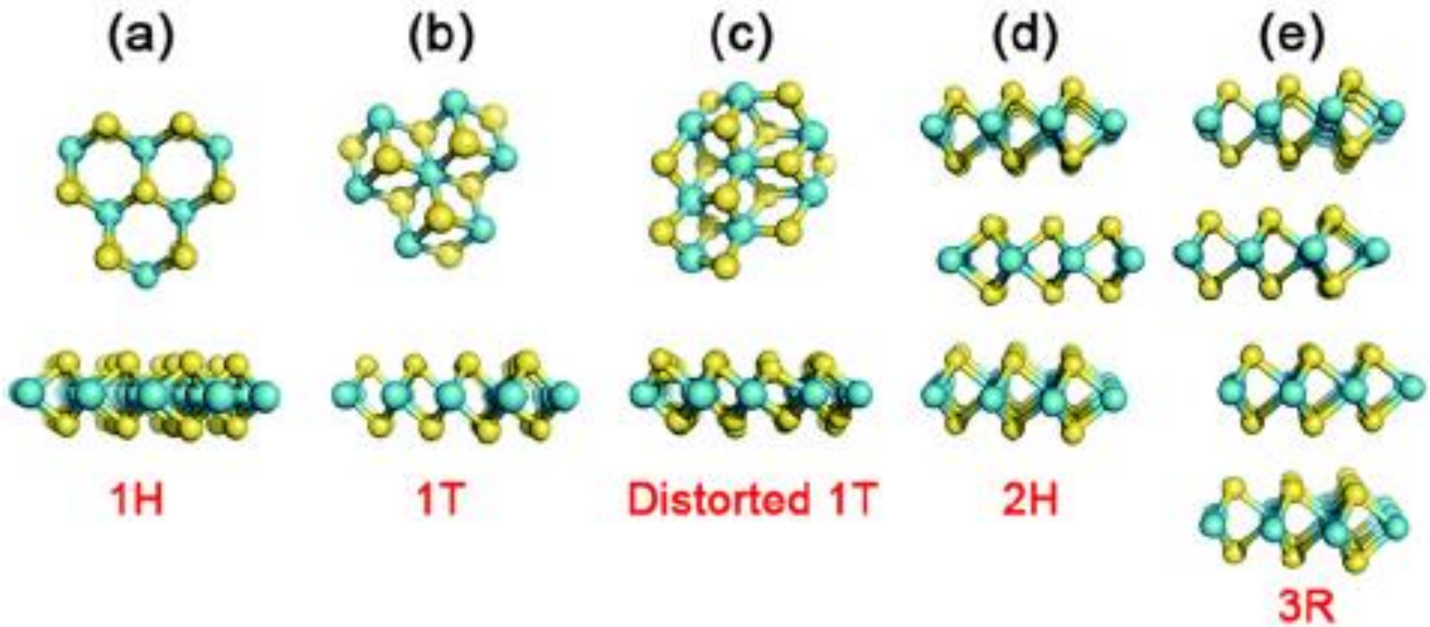
Transition Metal Dichalcogenides (TMD)

2D TMD MX_2
M : Mo, W
X : S, Se, Te



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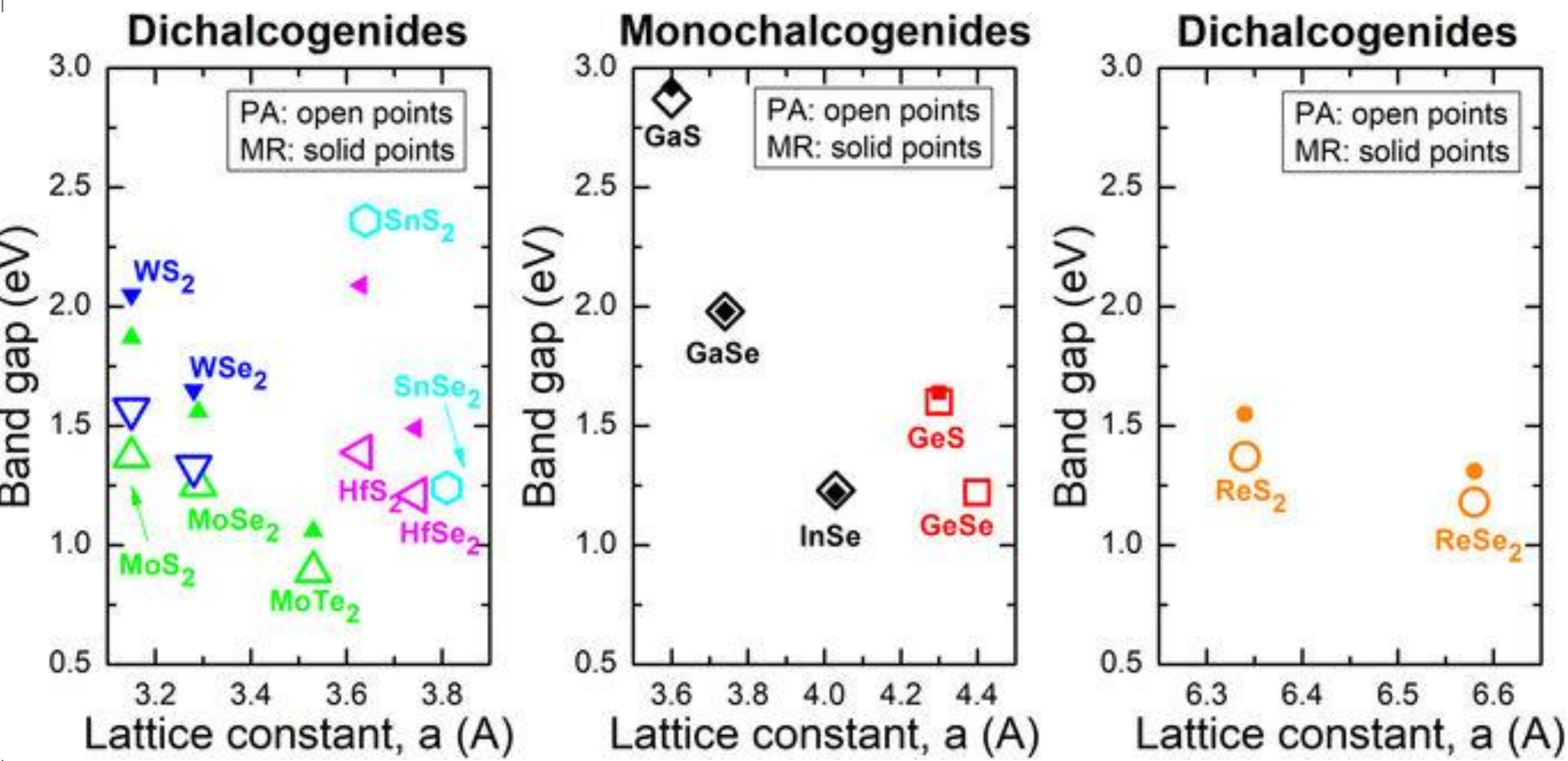
Lin et. al., 2D Materials (2016)

Transition Metal Dichalcogenides



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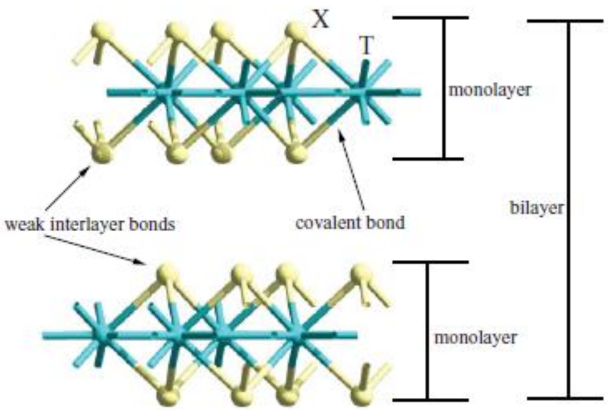
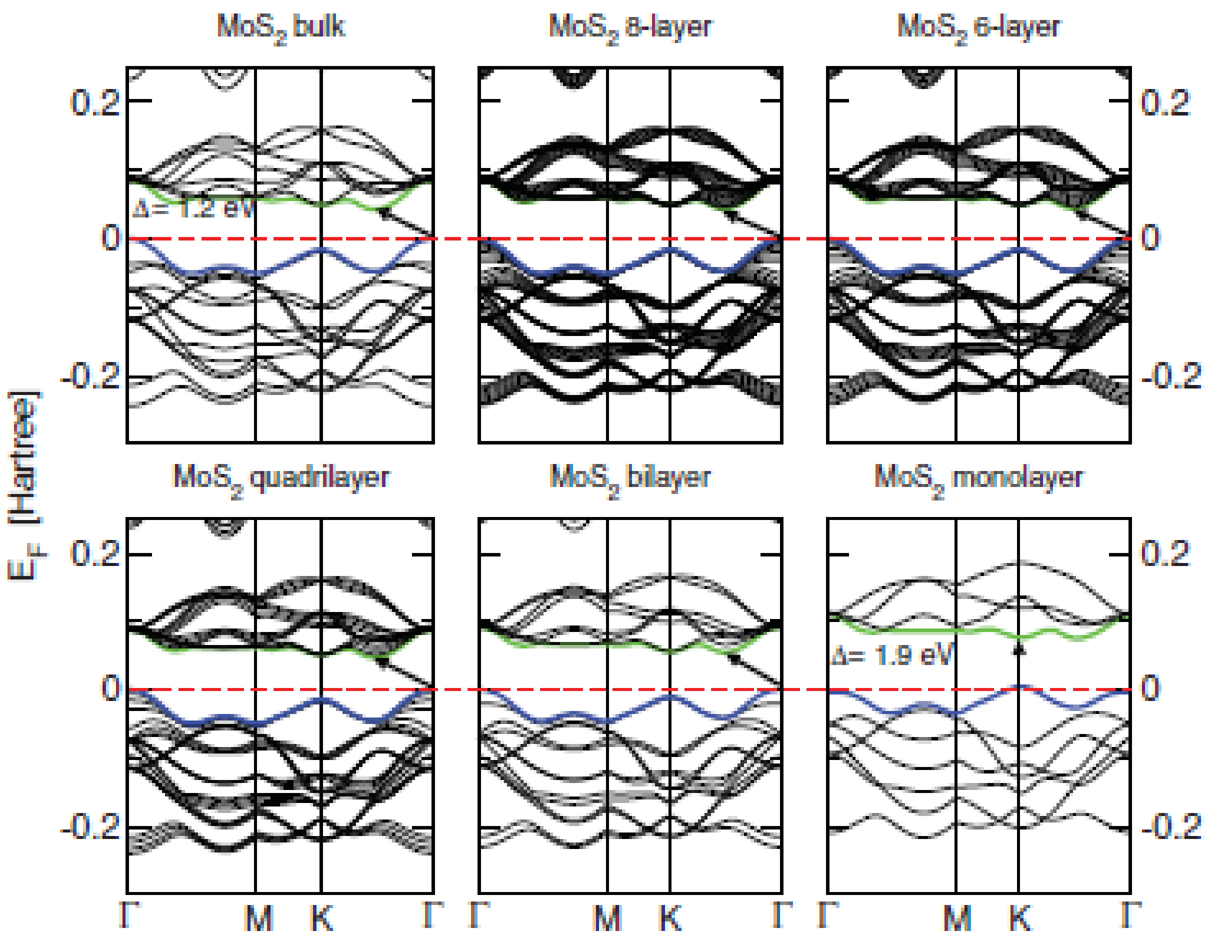
Zelewski et al., Scientific Reports (2017)

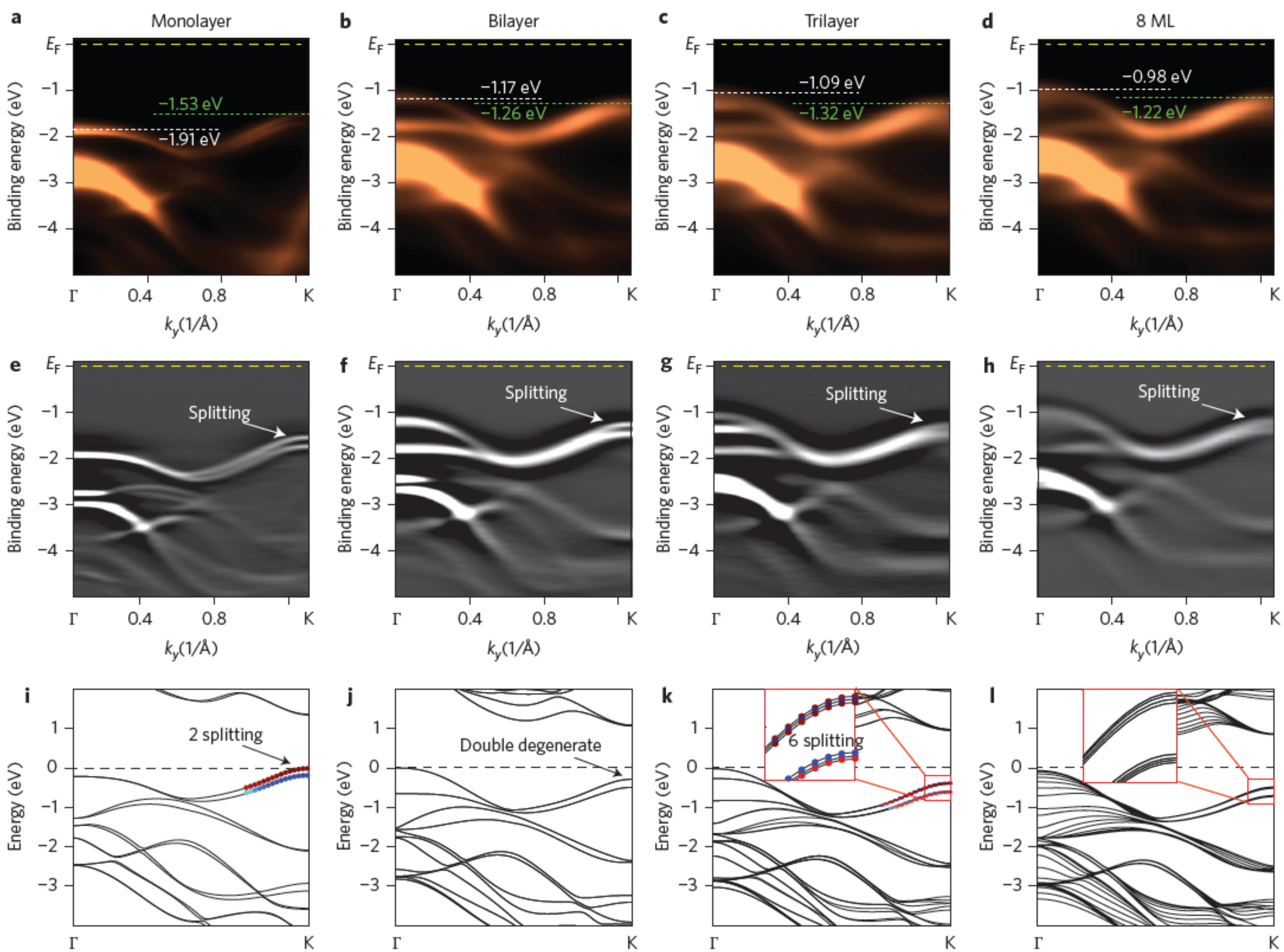
Transition Metal Dichalcogenides : MoS_2





Kuc et. al., PRB (2011)

Transition Metal Dichalcogenides : $MoSe_2$



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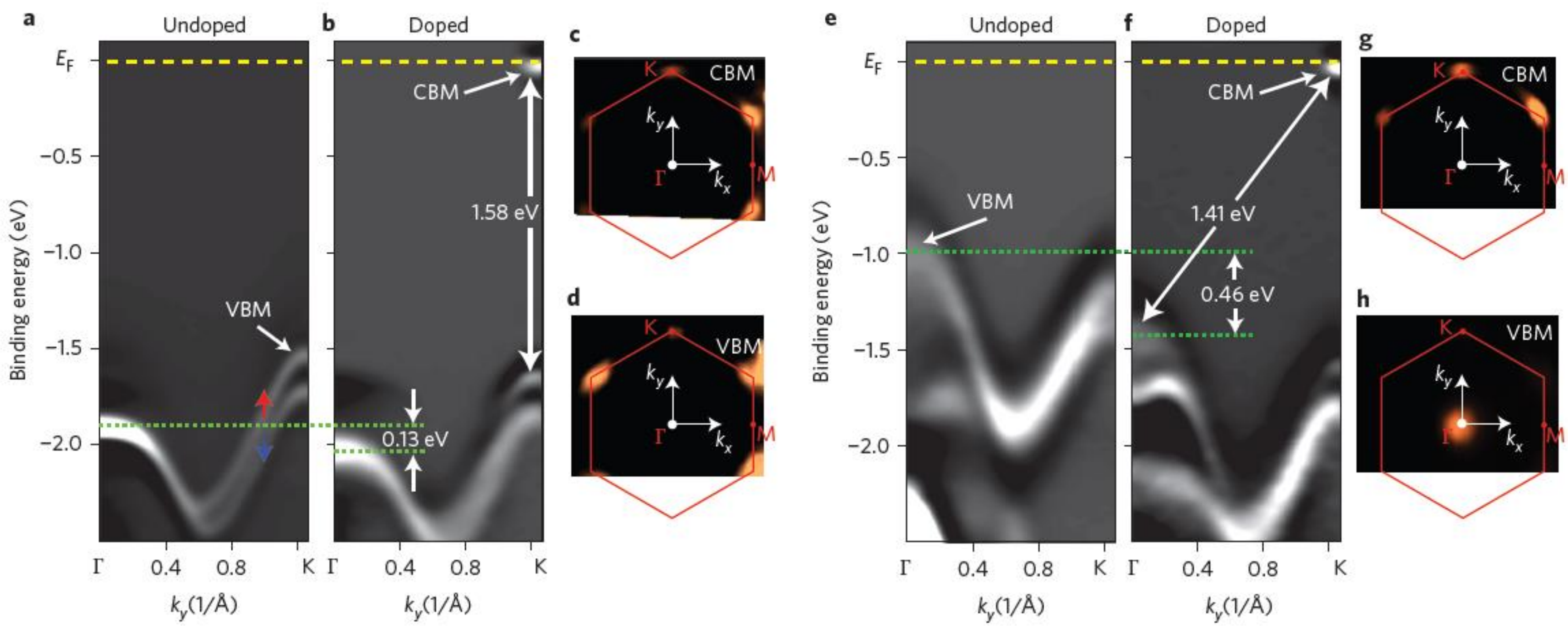
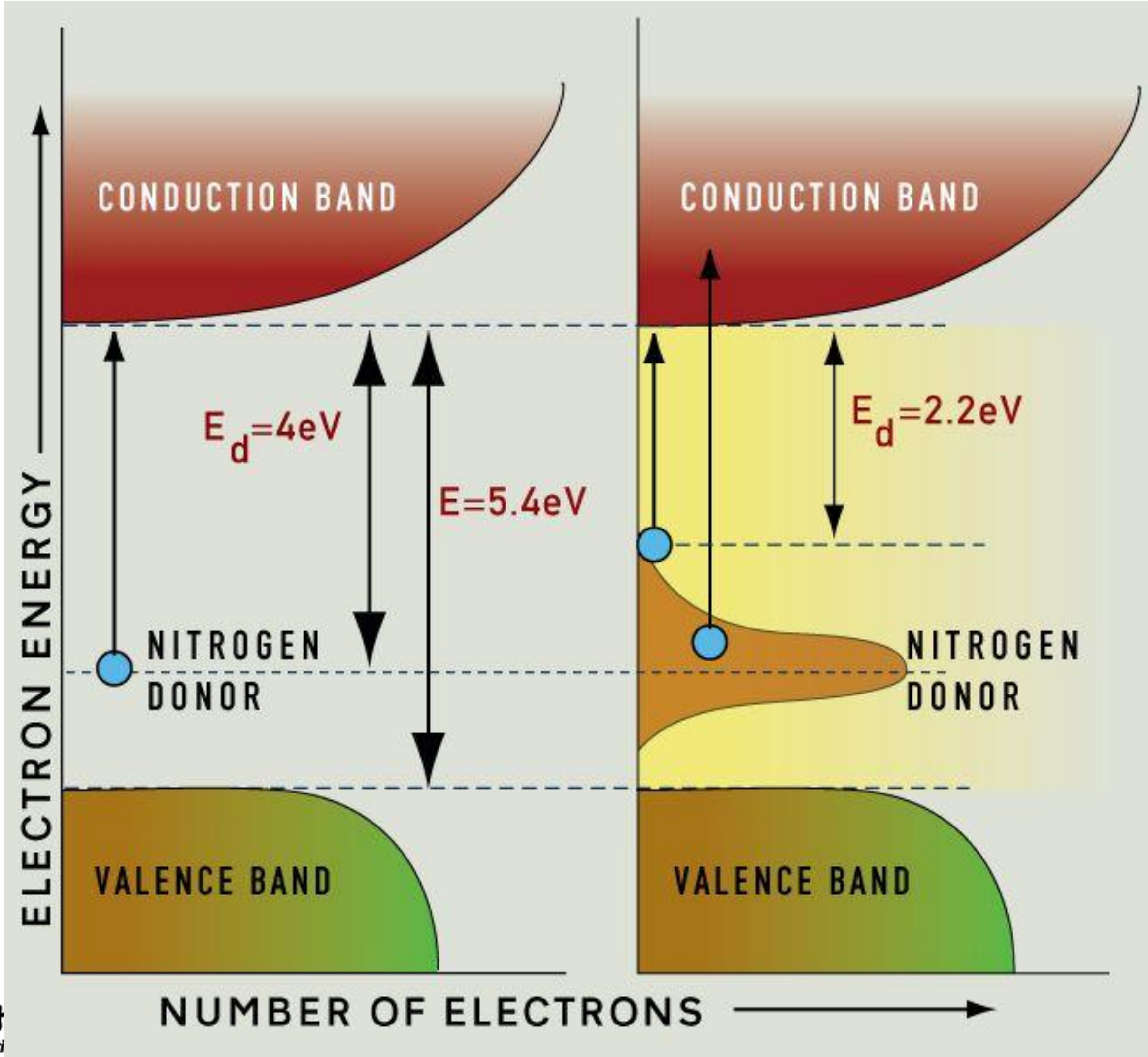
Zhang et al., Nat. Nano. (2013)

Transition Metal Dichalcogenides :$MoSe_2$



Defect engineering of semiconductor



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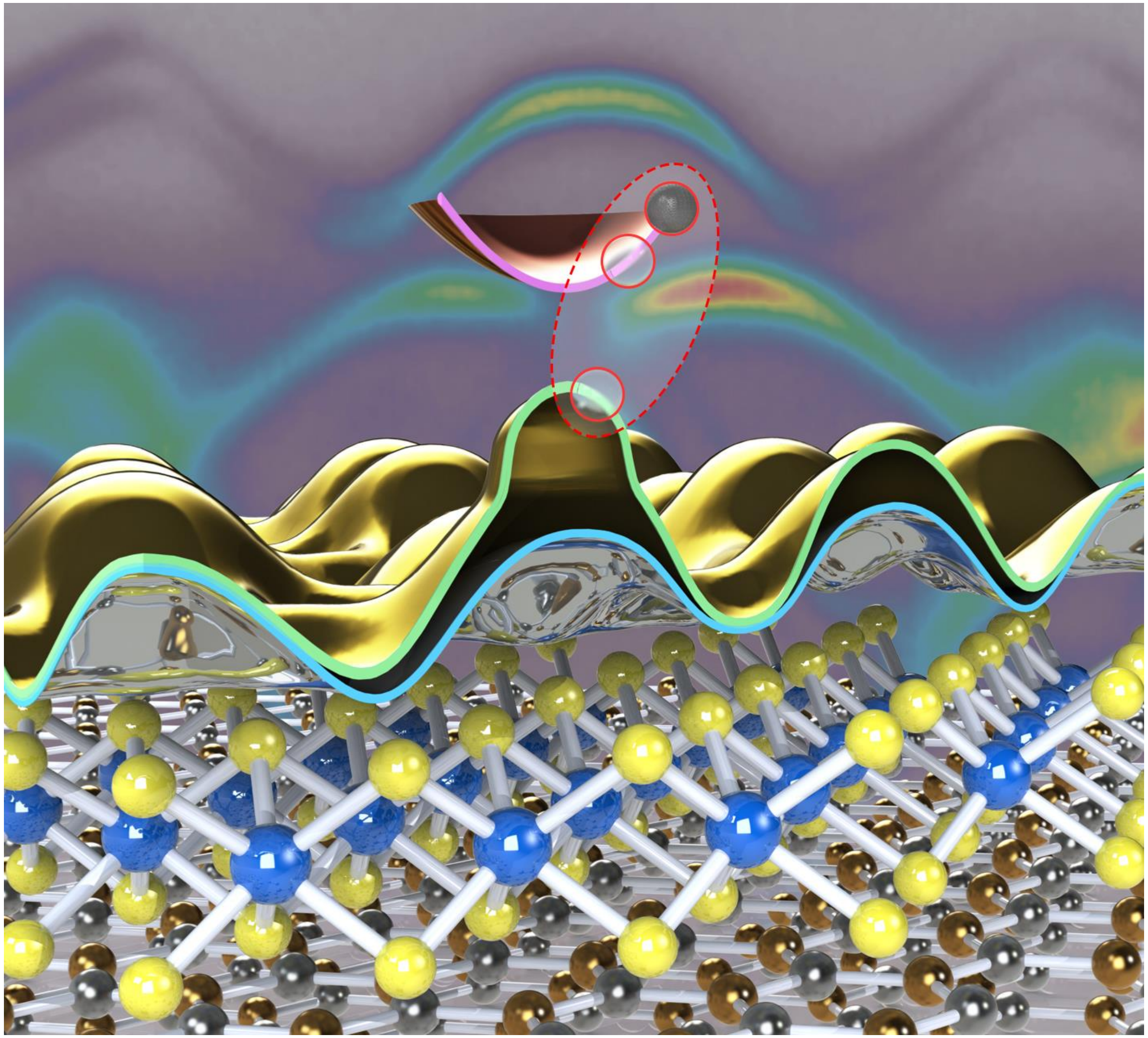
Tuning the Electronic Structure of a 2D Material



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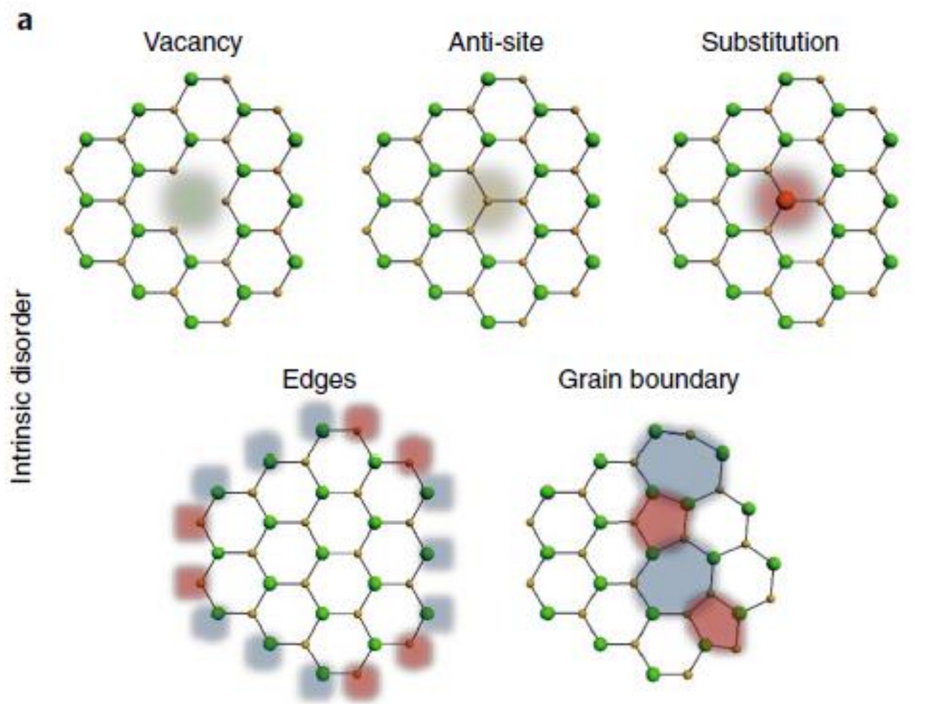
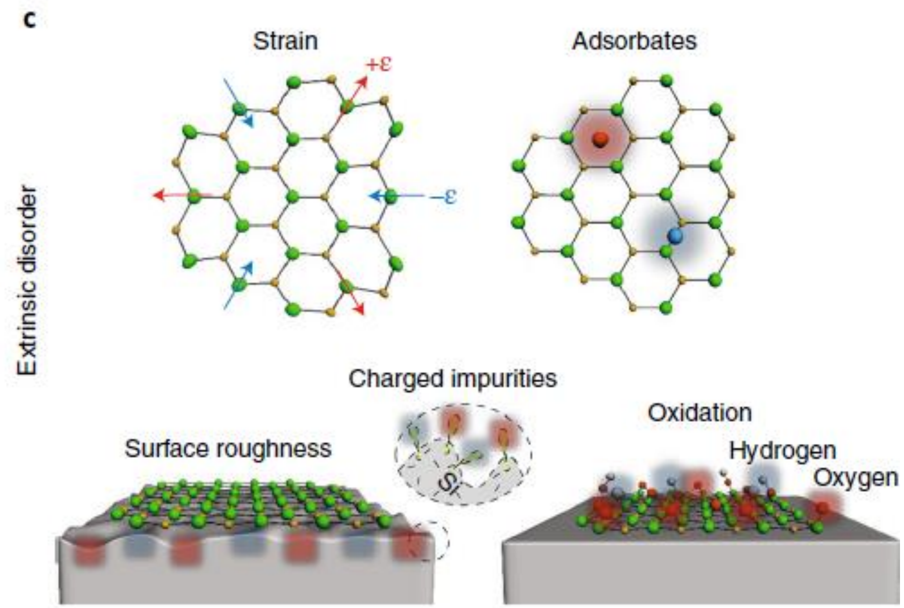
Transition Metal Dichalcogenides : Defect engineering

Intrinsic

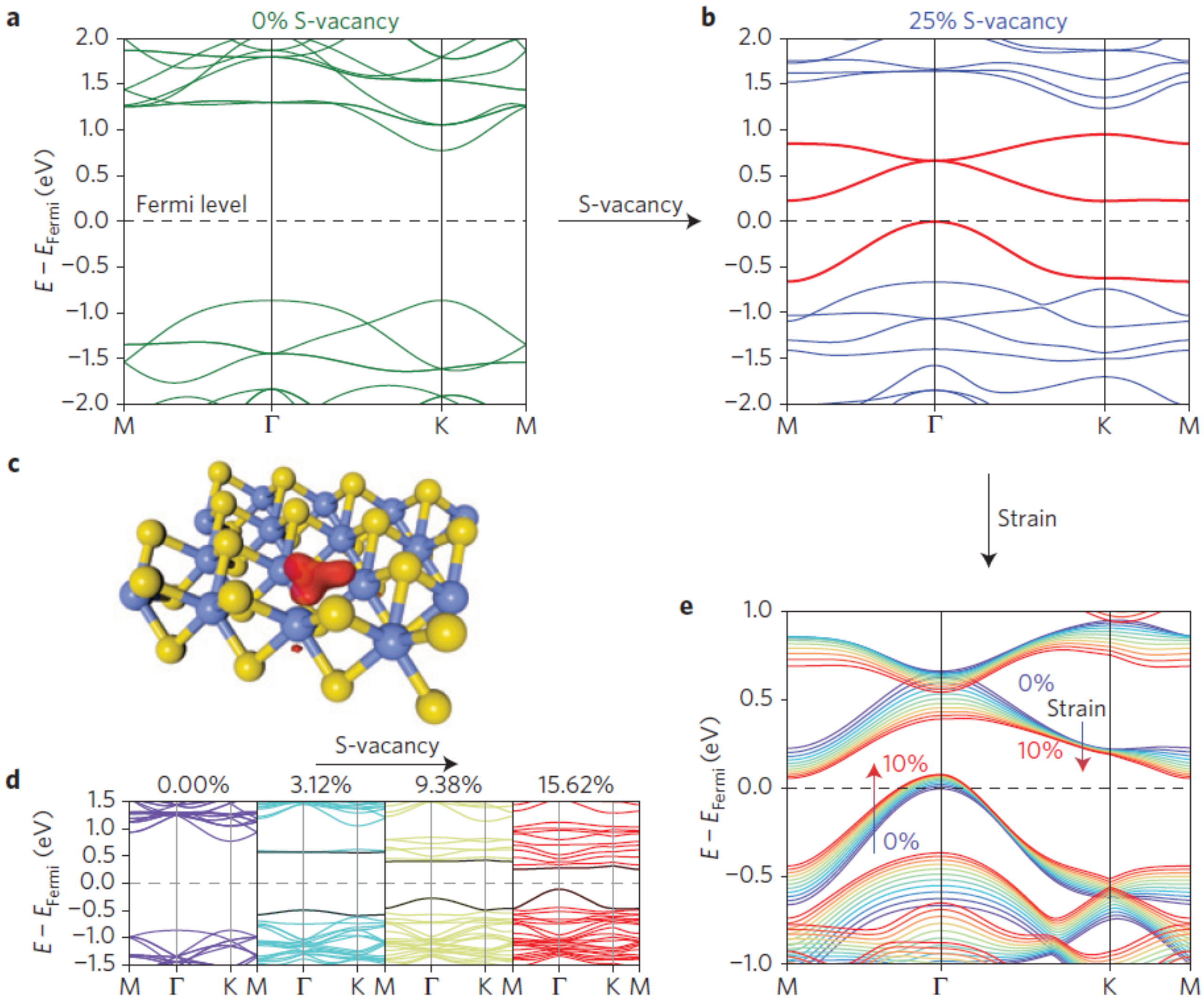


Extrinsic

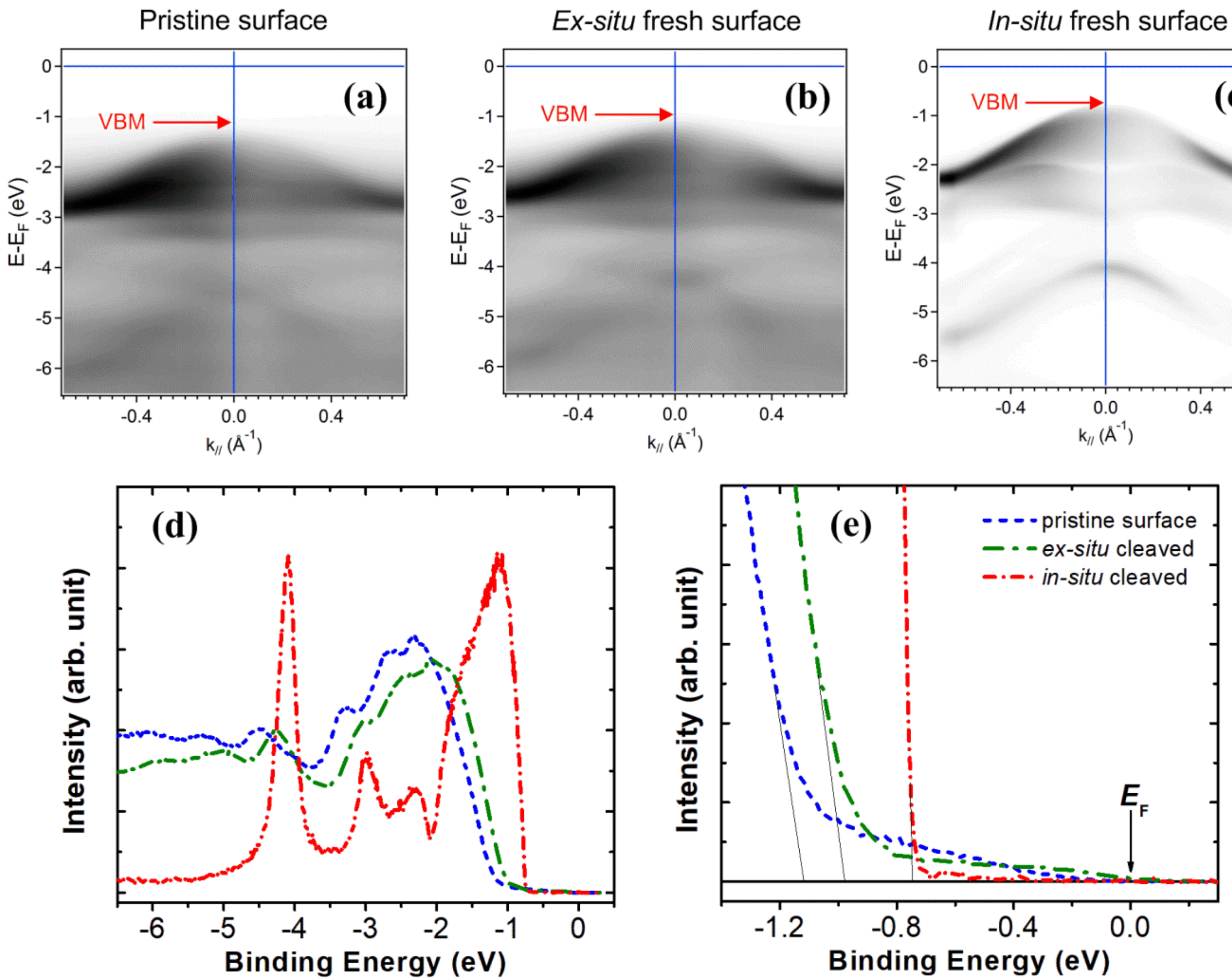


Rhodes et al., Nat. Mater. (2019)

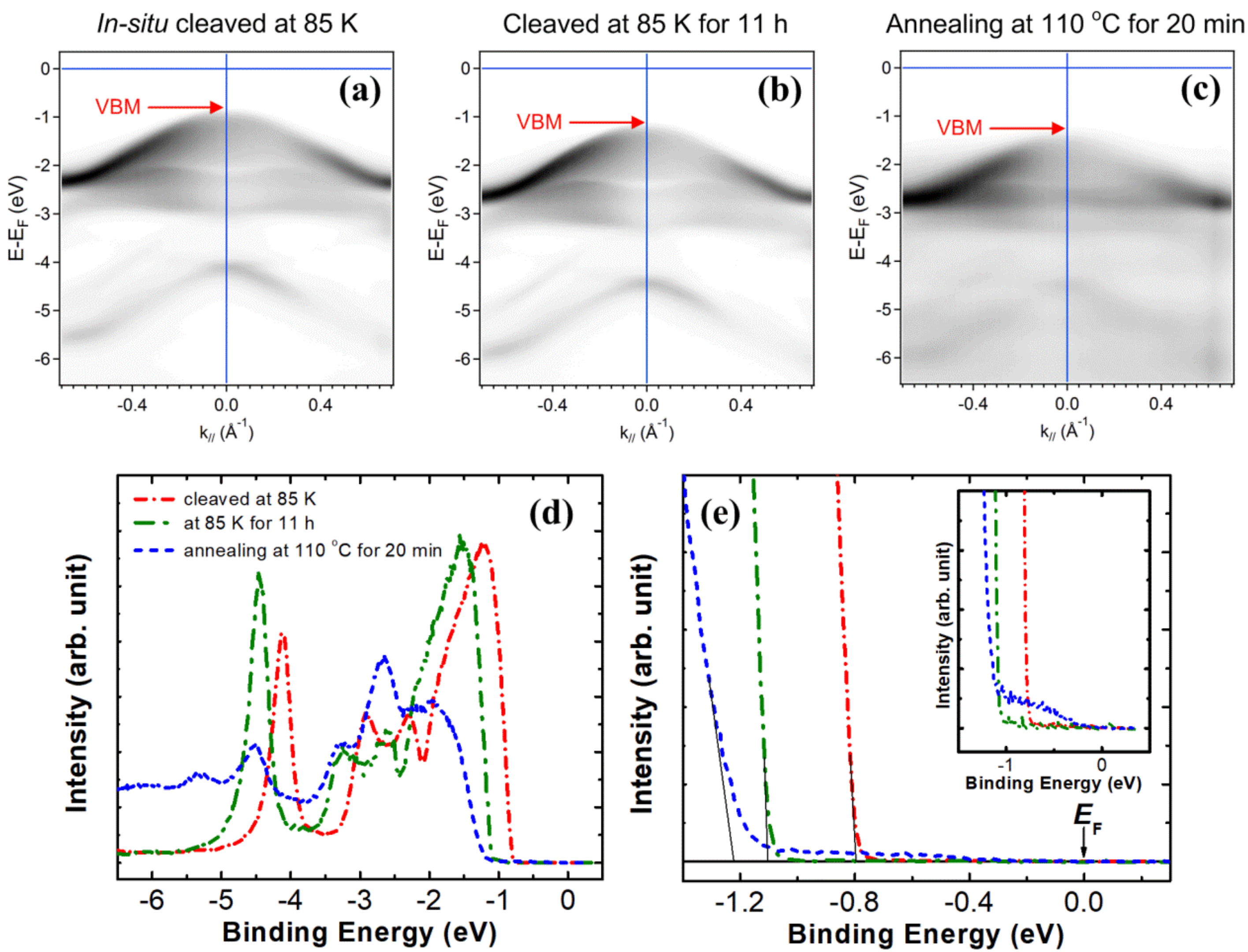
S-vacancy and strain on the band structure of MoS_2



The band structure of MoS_2 single crystal with varied environment



S-vacancy and strain on the band structure of MoS_2



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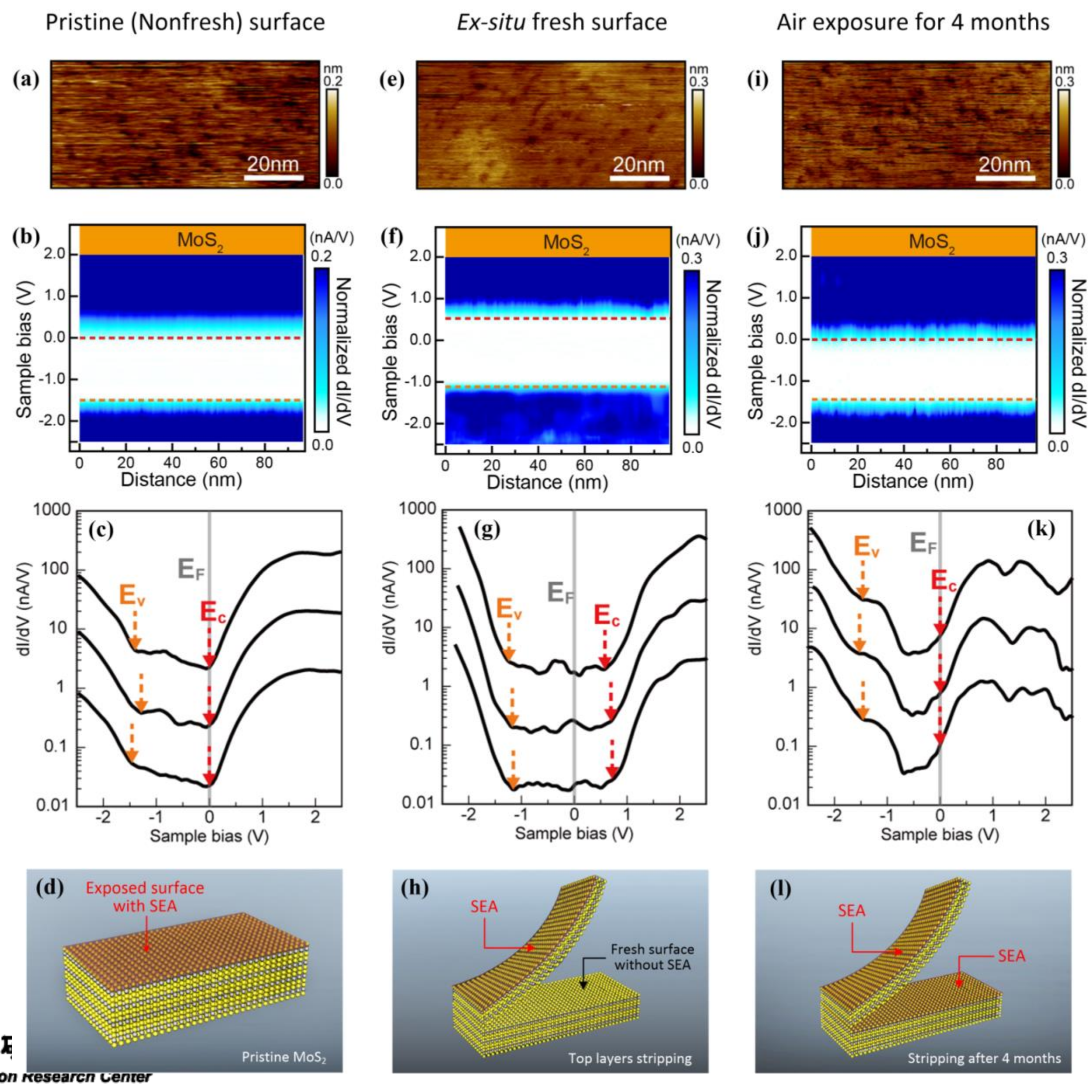
STM characterization of a MoS_2 single crystal



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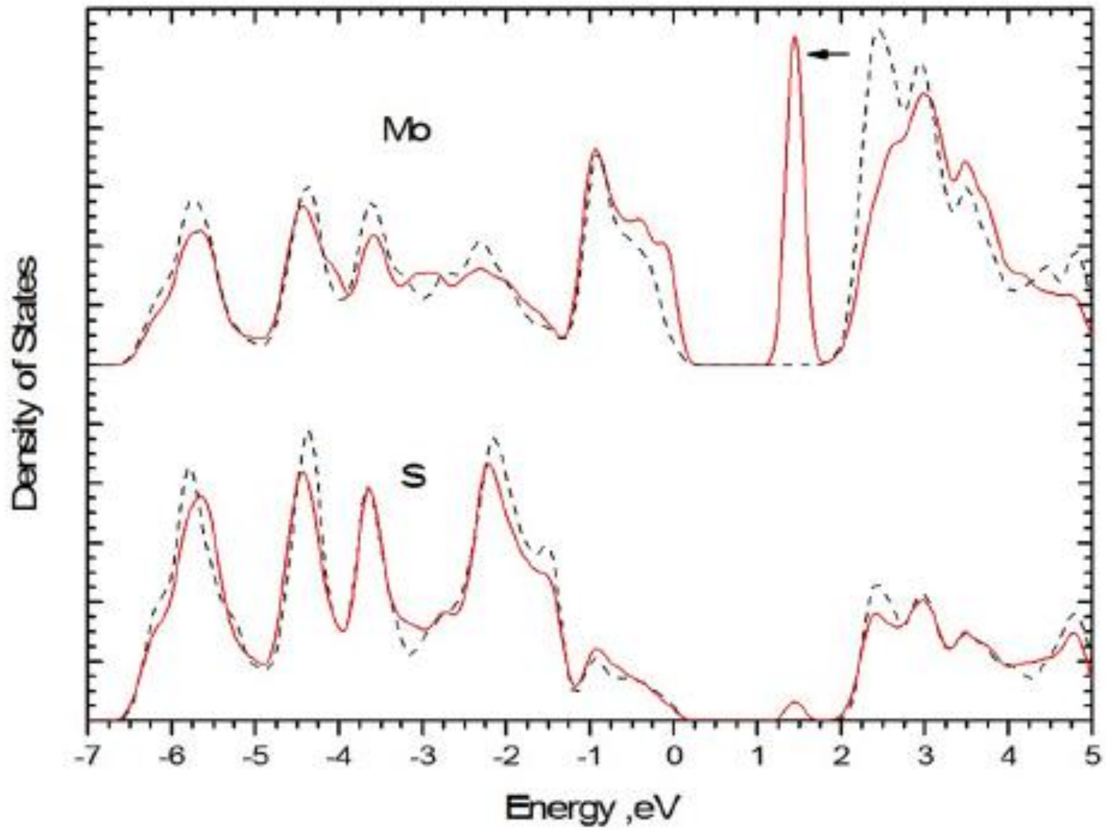
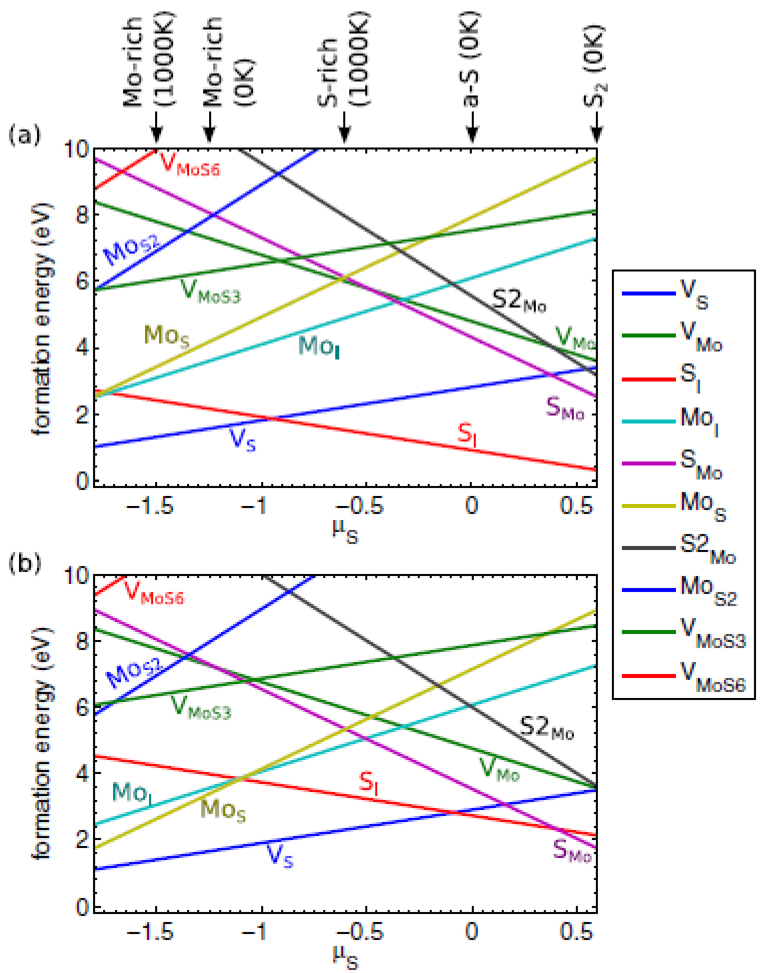
Sulfur vacancies in monolayer MoS_2 and its electrical contacts

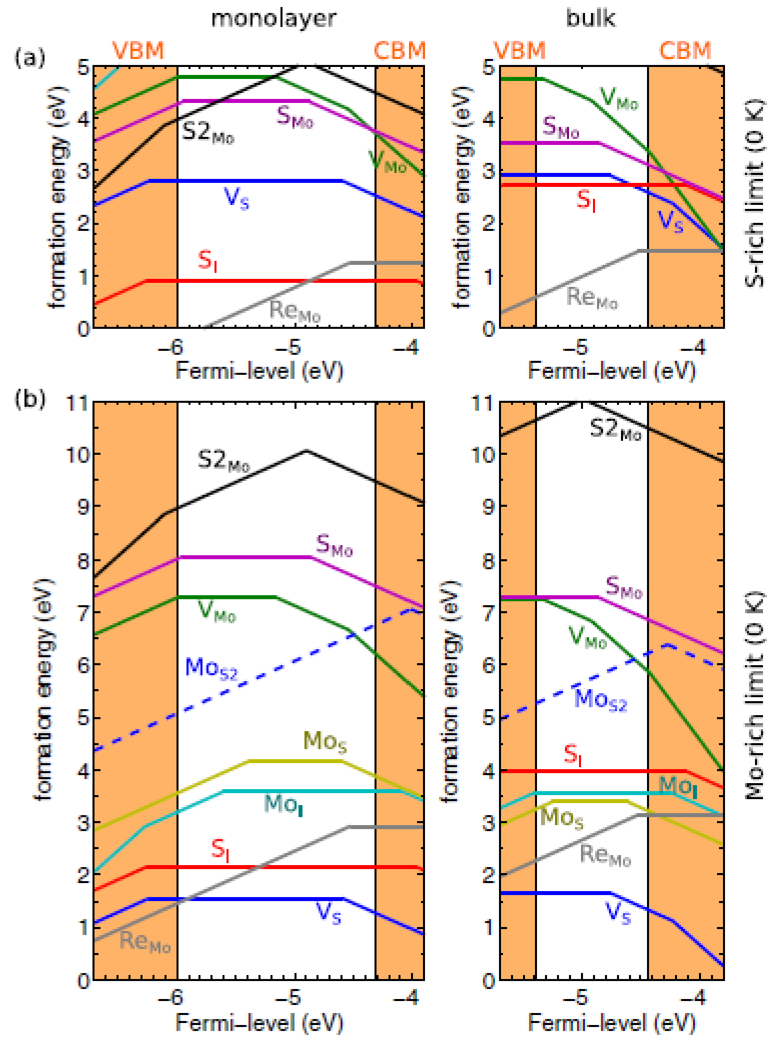
S vacancy has a formation energy of 2.35 eV in S-rich condition
Mo vacancy has a formation energy of 8.02 eV in Mo-rich condition



Liu et al., APL (2013)

Formation energies as a function of the Fermi level





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Komsa et al., PRB (2015)

Bandgap, Mid-Gap States, and Gating Effects in MoS_2

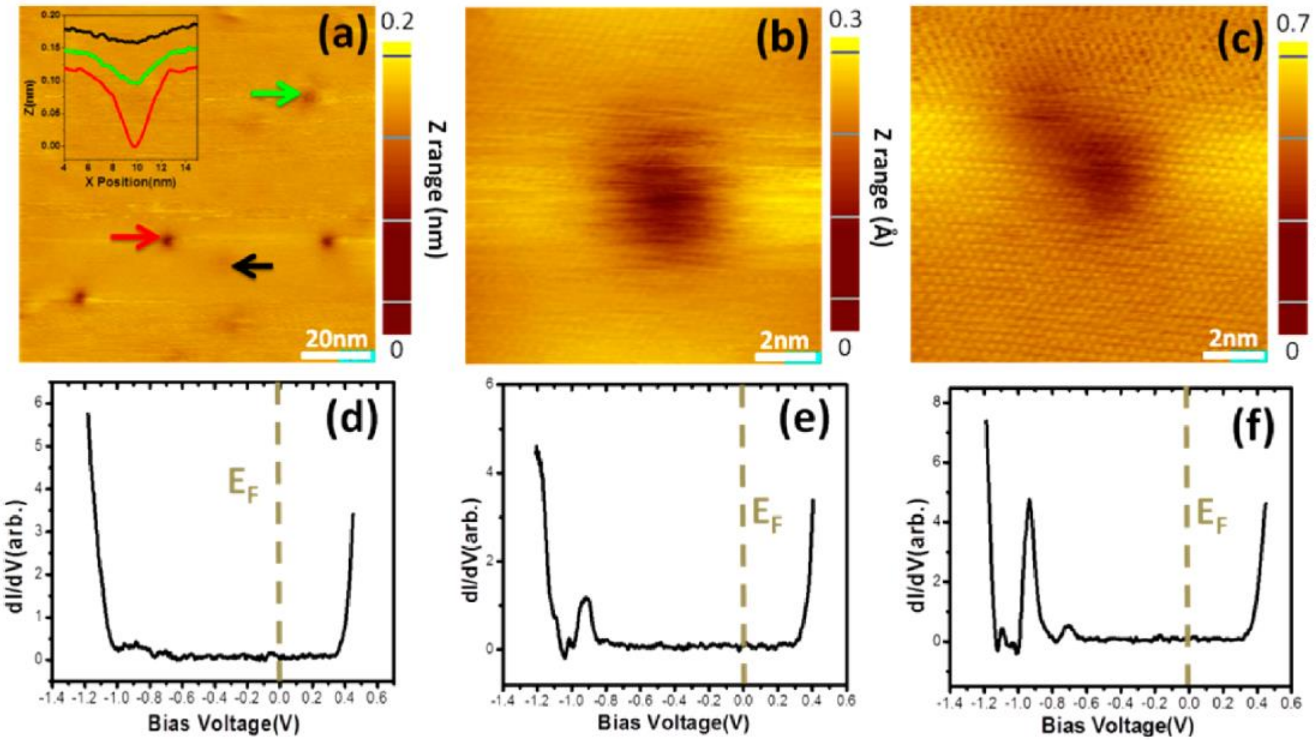
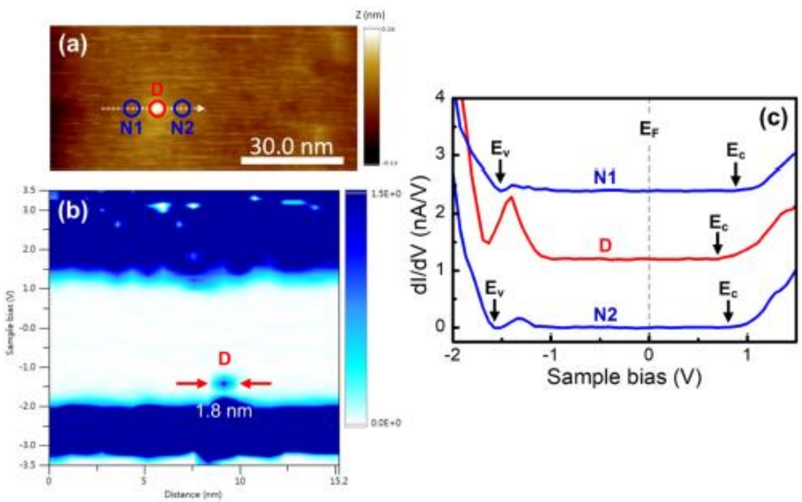


Figure 4. (a) STM constant current topography image of a 50 × 50 nm area at V_b = 1.4 V and I = 20 pA on bulk MoS_2 showing three types of defects with different apparent depths as indicated by the height profiles in the inset. (b) Constant current STM image on an isolated defects. (V_b = 1.2 V and I = 20 pA). (c) Constant current STM atomic resolution image on a twin-defect. (V_b = 1.2 V and I = 20 pA). (d) dI/dV spectrum taken far from any defect. (e) dI/dV spectrum on isolated defect in (b) shows a pronounced in-gap resonance near −0.94 V. The Fermi level is ~0.35 eV below the CB edge. The measurement was referenced to a set-point with tunneling resistance 15 GΩ (bias voltage V_b = −1.5 V and tunneling current I = 100 pA). (f) dI/dV spectrum on the twin-defect in (c) shows a pronounced in-gap resonance near −0.94 V and a satellite peak at −0.7 V. Parameters are the same as in (e).

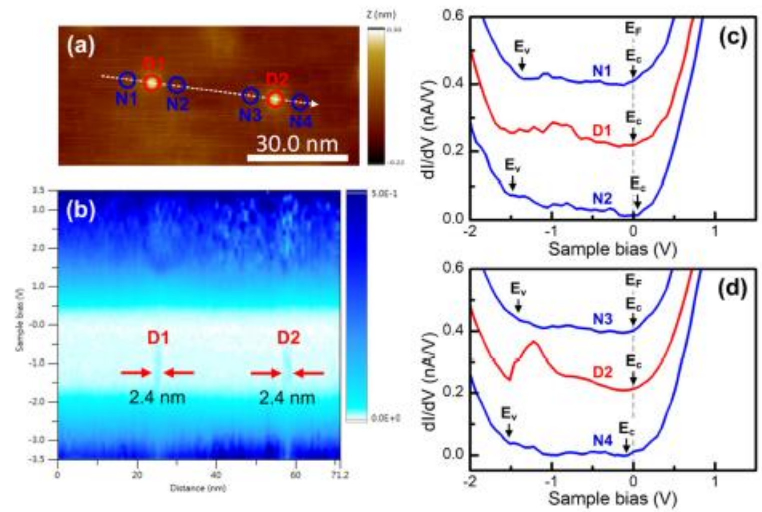
Lu et al., Nano Lett. (2014)

Evidence from STS measurement

Fresh surface



Nonfresh surface



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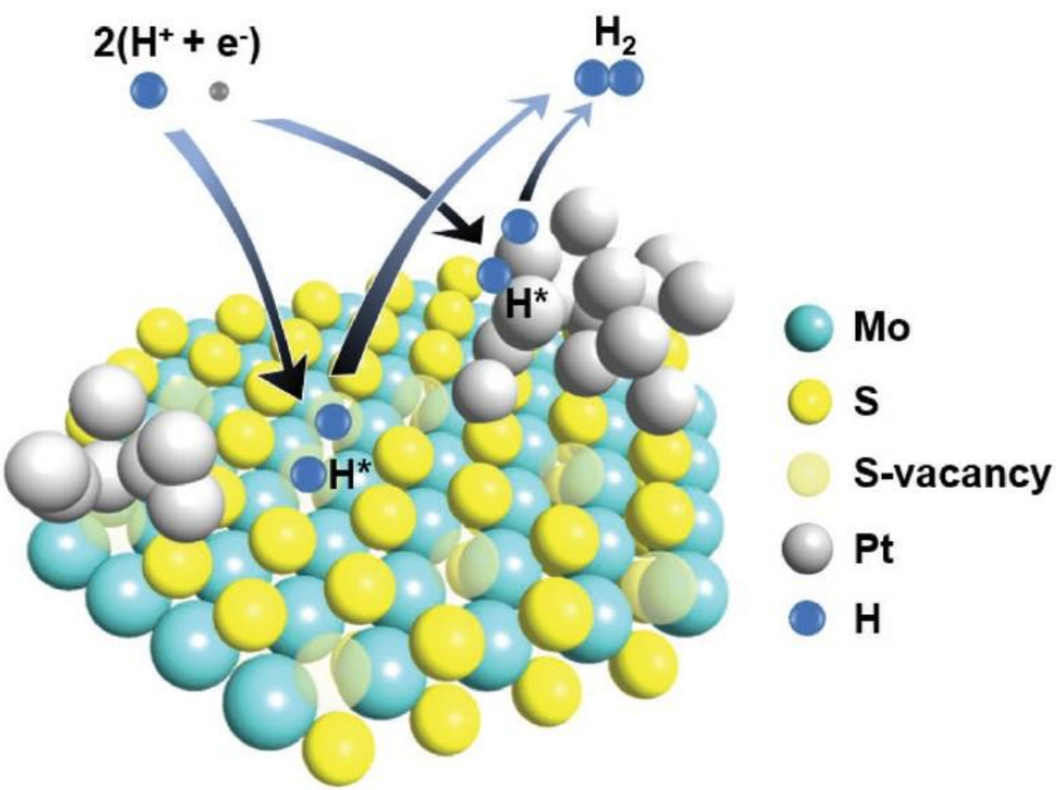
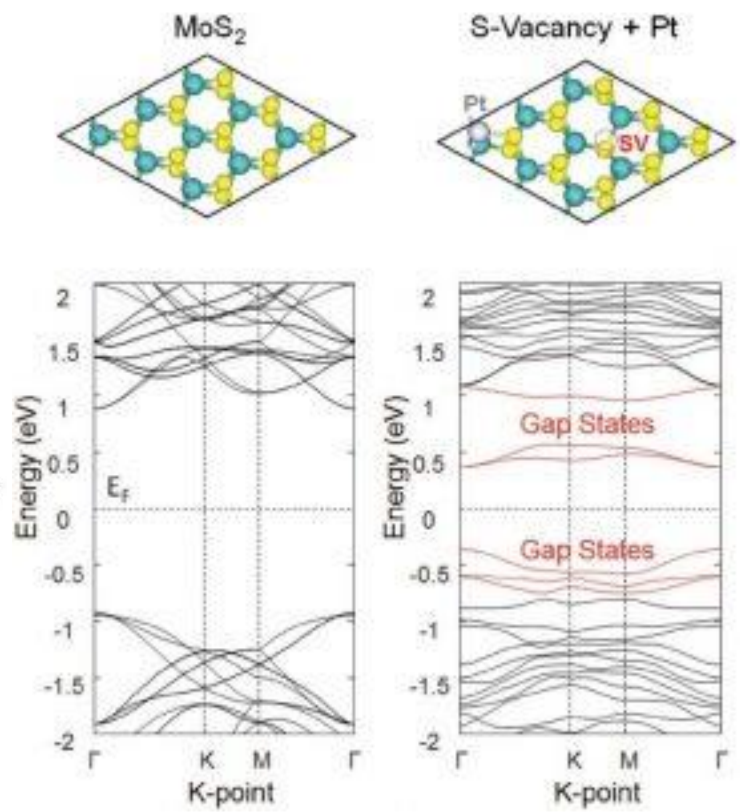
Siao et al., Nat. Comm. (2018)

Engineering the Catalytic Performance of TMDs

Hydrogen evolution reaction (HER)

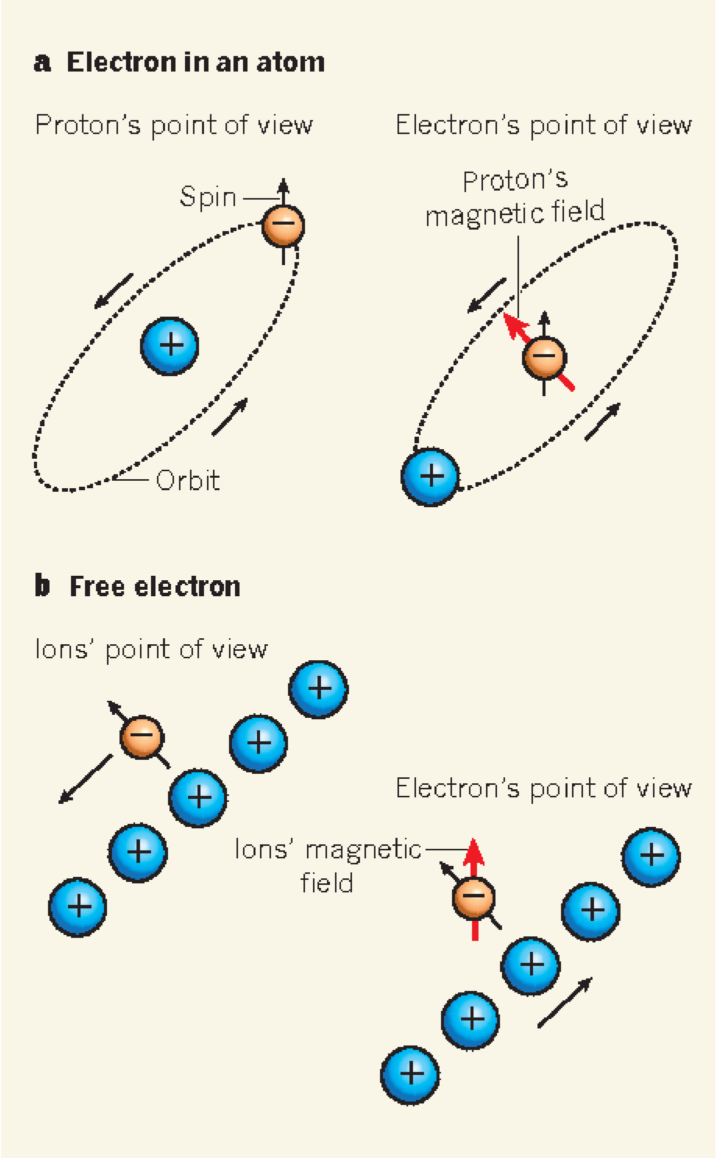




Topological insulators

Spin-orbital interaction

Concept of spin-orbit coupling



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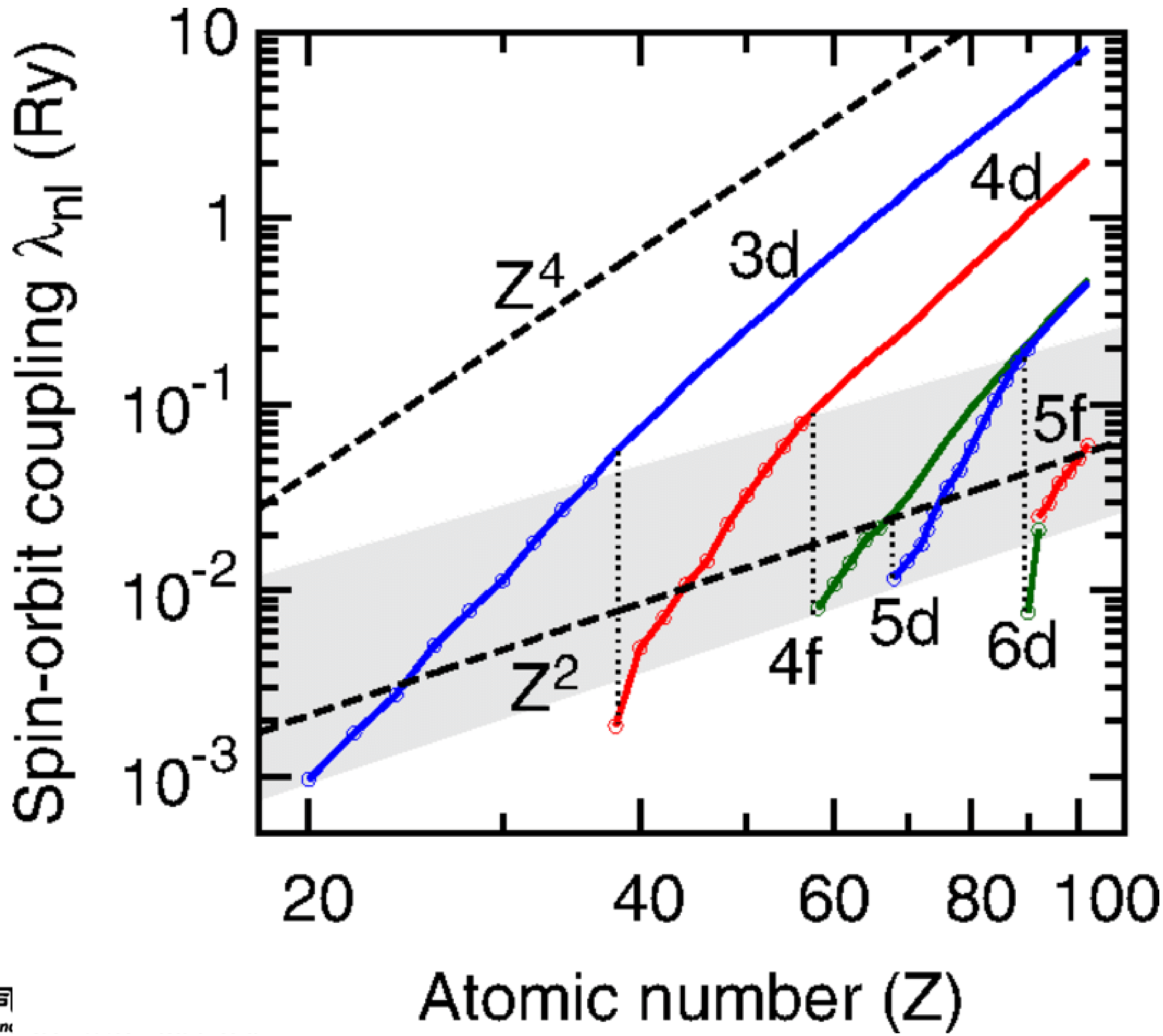
Spin-orbital interaction



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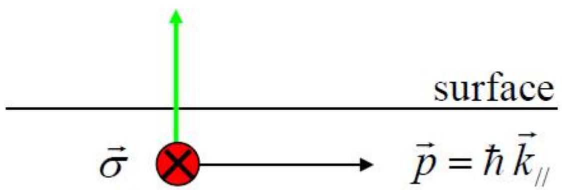
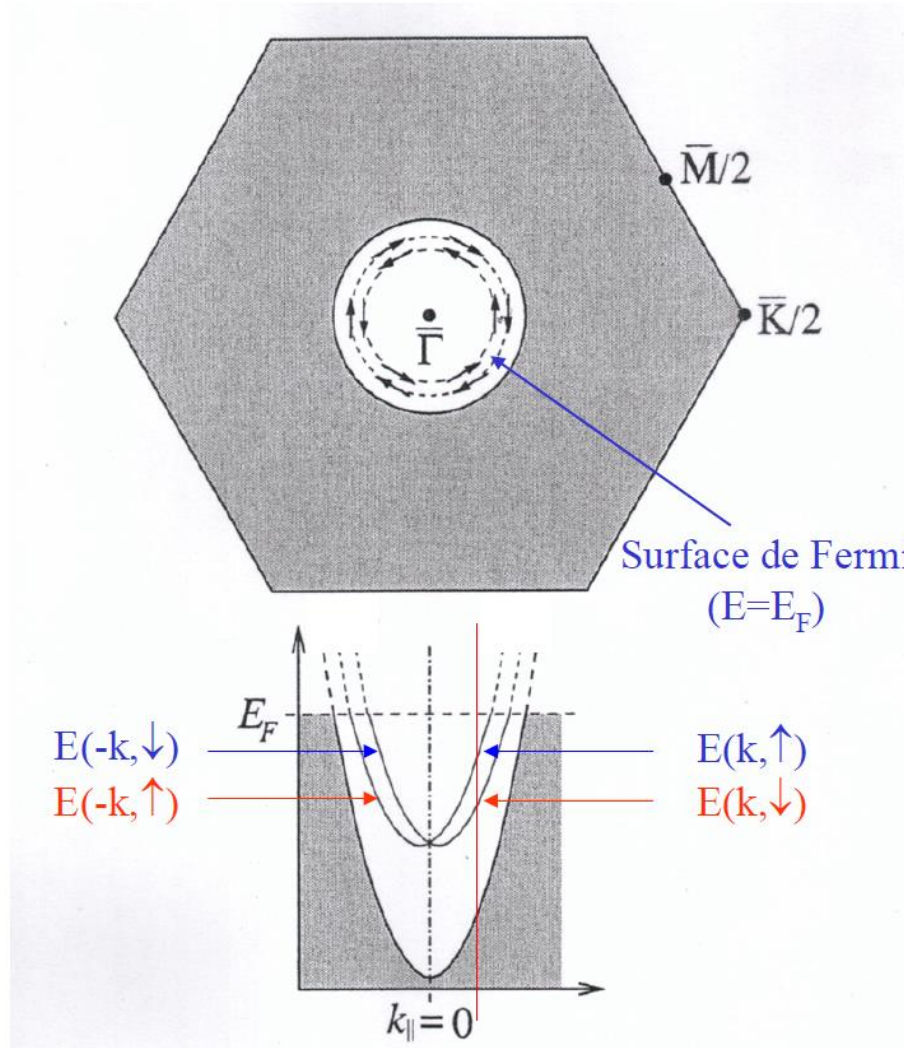
Au(111) : The inversion symmetry is broken at the surface

$$H_{so} = \frac{\hbar}{4m^2c^2}(\vec{\nabla} V \wedge \vec{p}).\vec{\sigma}$$

$$\vec{\nabla} V = \frac{dV}{dz}\vec{e}_z$$



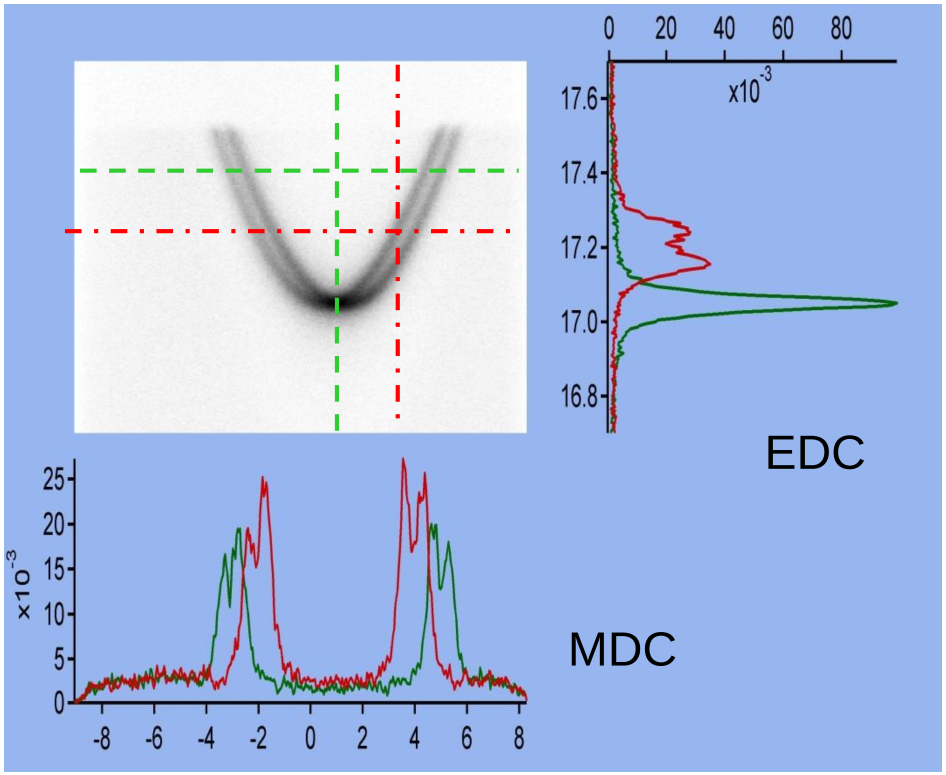


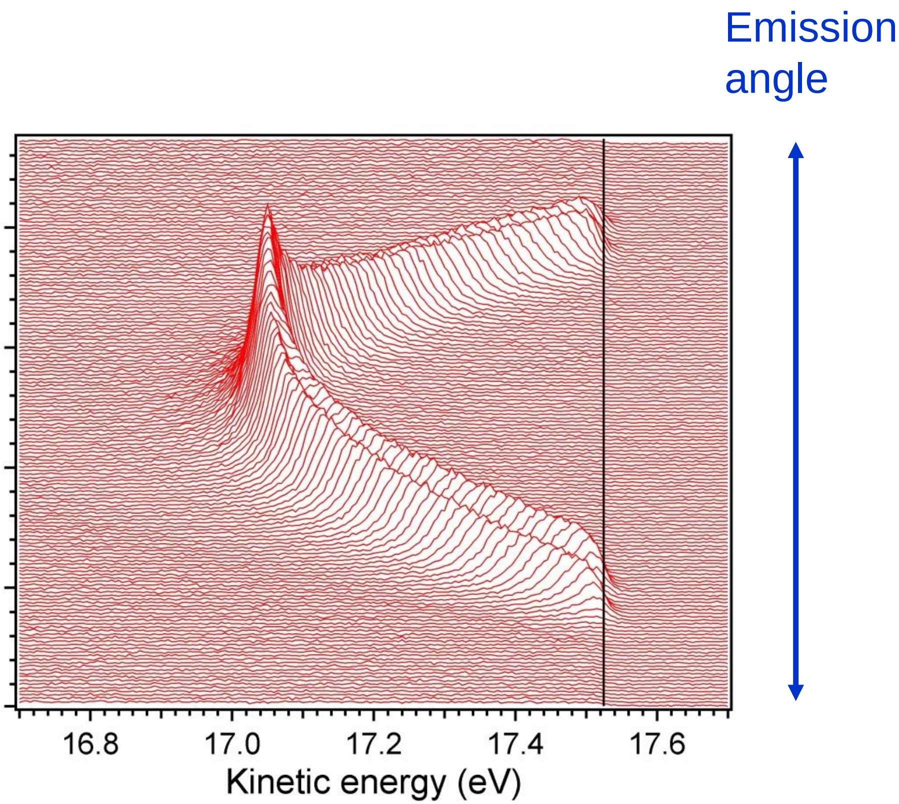
In a nearly free electron picture,
$\vec{\nabla} V$ is perpendicular to the surface

→ two spin polarized SS bands

Rashba Splitting

Accumulate spectra of Rashba effect on Au(111) as the angle is scanned





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Scientific Cases: Topological protected surface state

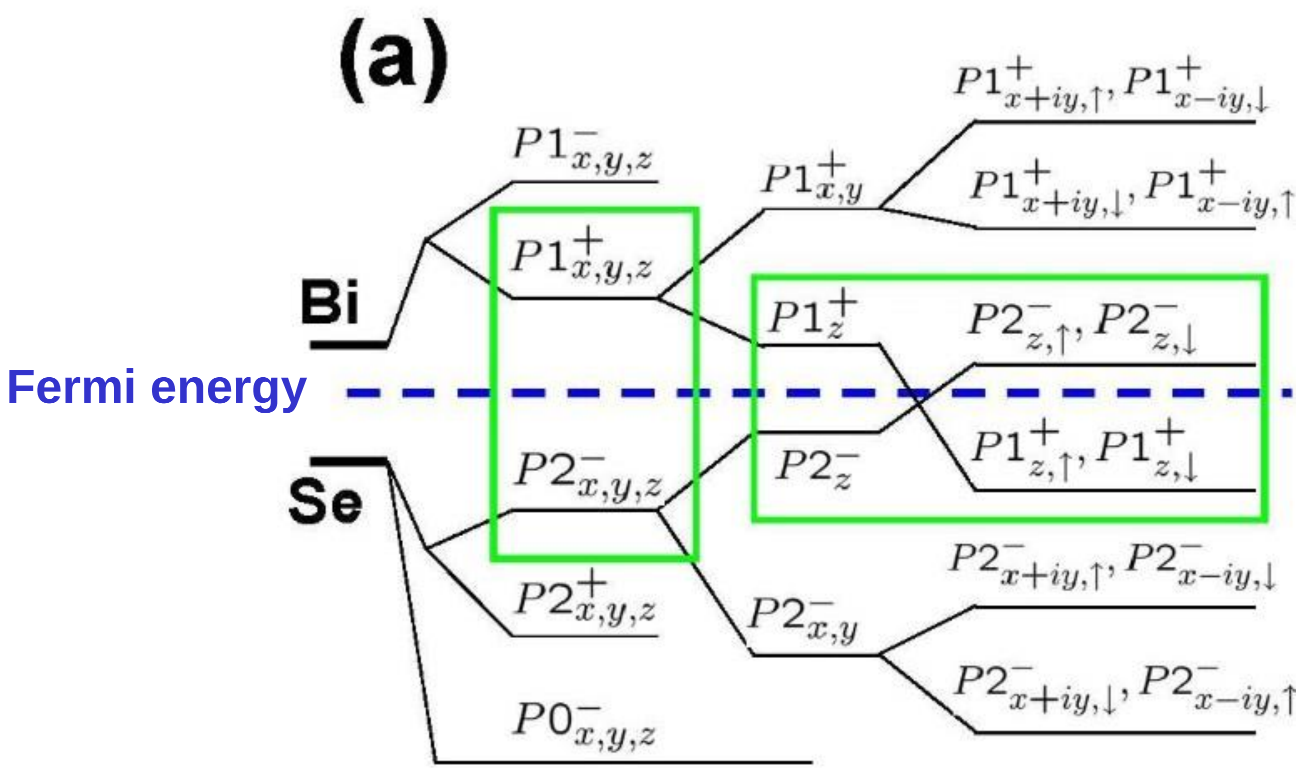
Shou-Cheng Zhang (1963-2018)

Strong spin-orbital interaction

Heavy elements

10	11	12	13	14	15	16	17	18
			5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
			13 Al Aluminium	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
78 Pt Platinum	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson

Scientific Cases: Topological protected surface state



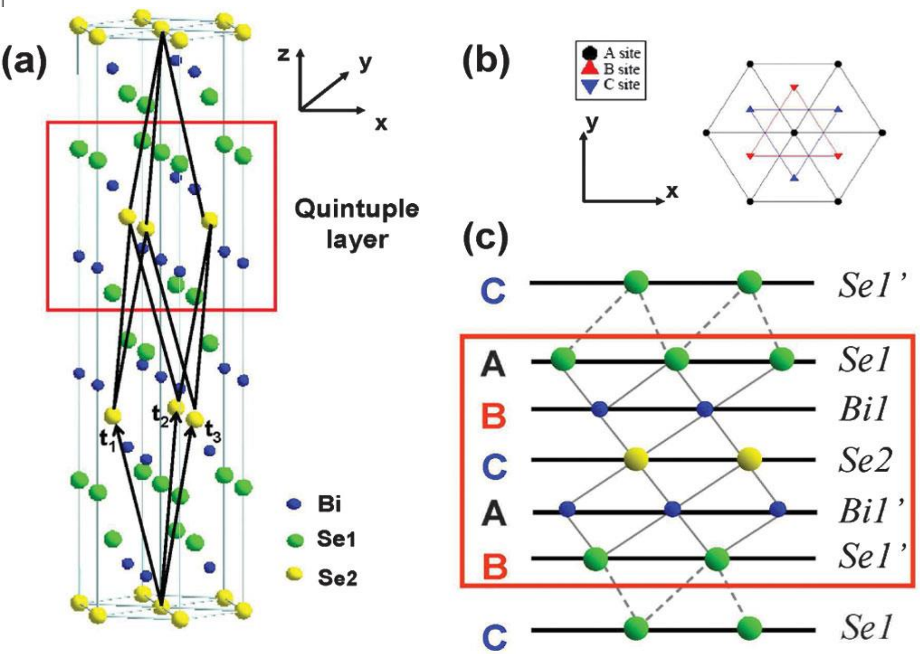
parity difference

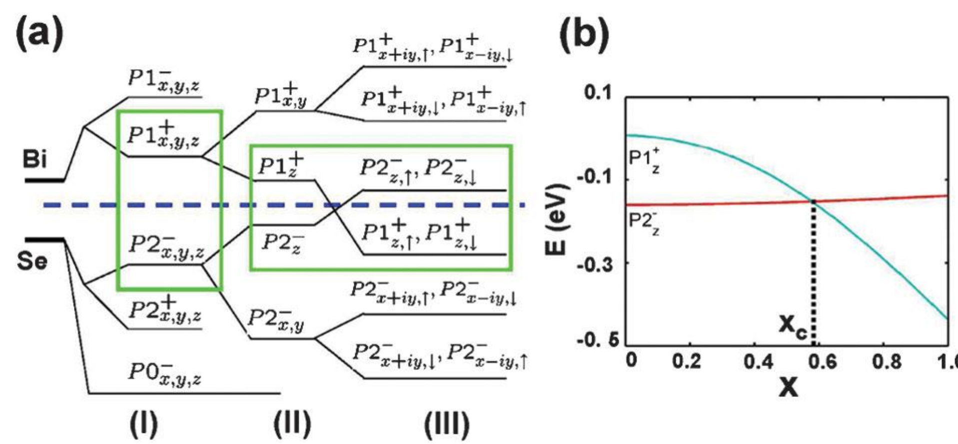
crystal field splitting

spin-orbital coupling

Xiao-Liang Qi and Shou-Cheng Zhang, RMP (2011)

3D topological insulators





Strong spin-orbit coupling
Induce the band inversion

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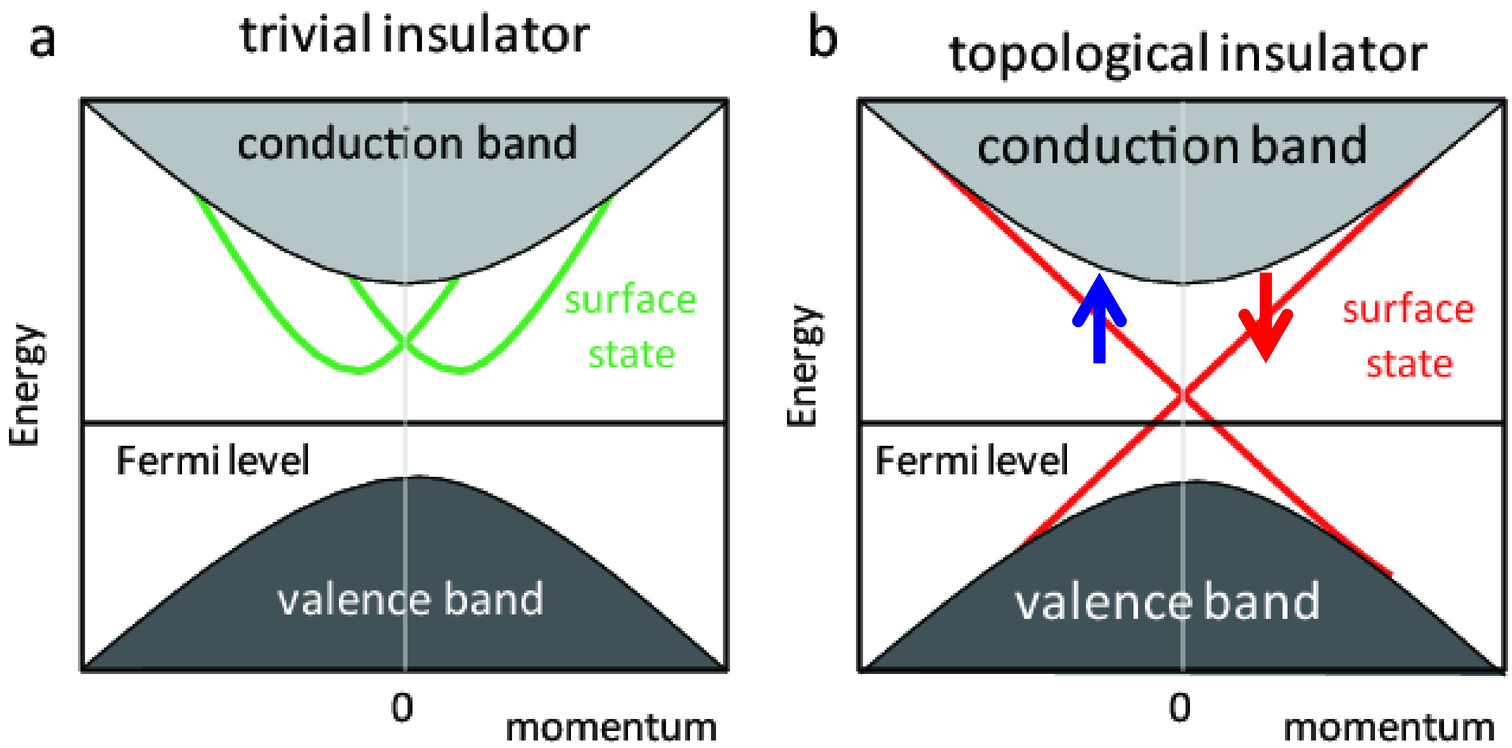
Xiao-Liang Qi and Shou-Cheng Zhang, RMP (2011)

Scientific Cases: Topological protected surface state



time-reversal symmetry protected

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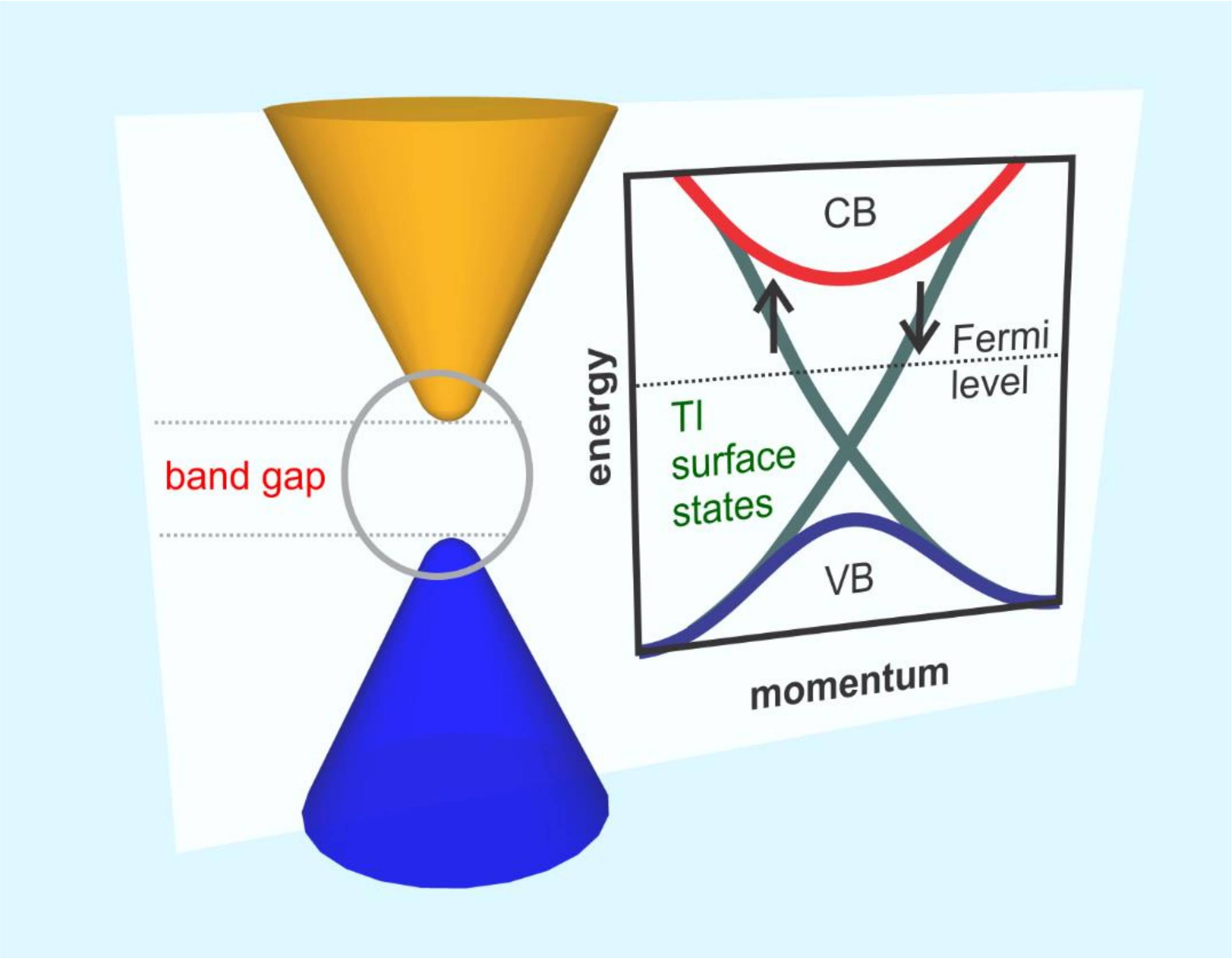
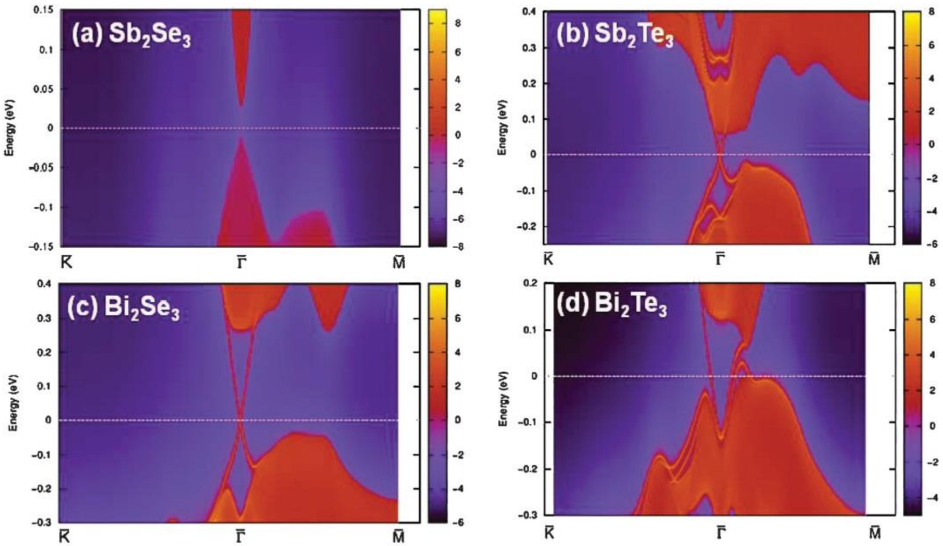
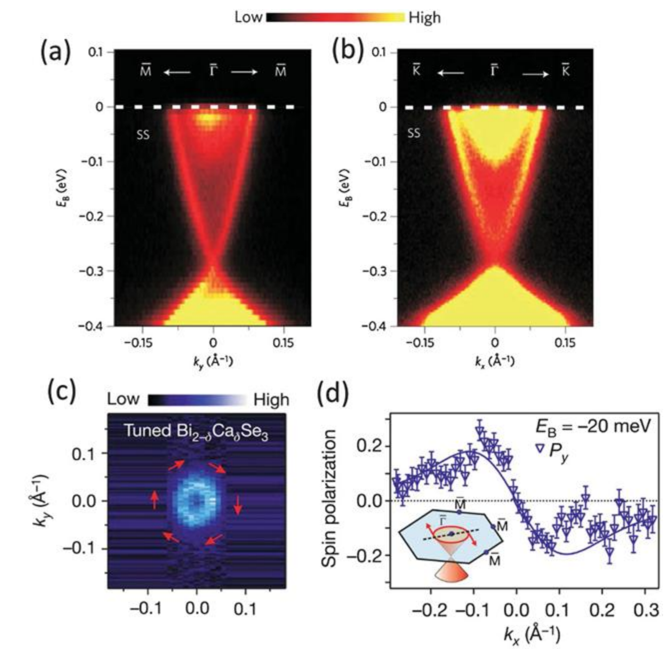
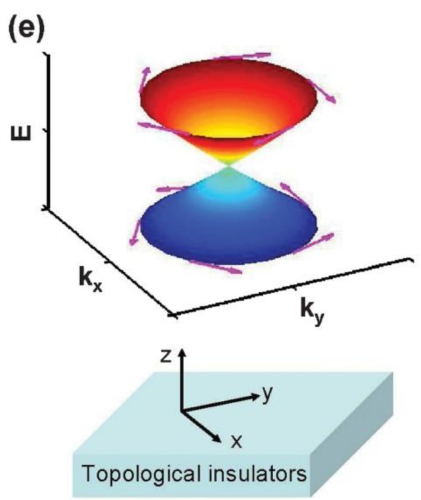
Scientific Cases: Topological protected surface state

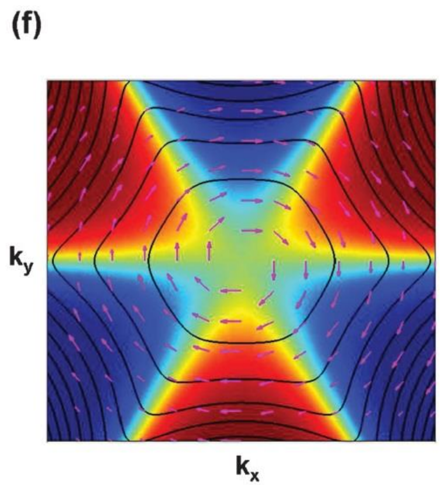


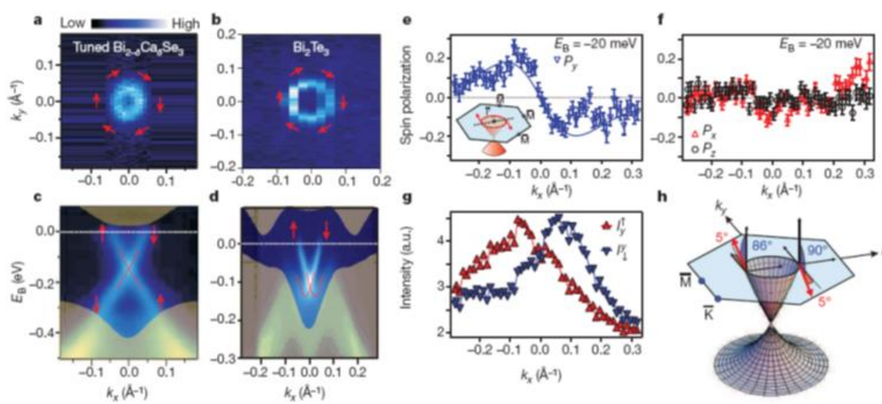
Topological protected surface state











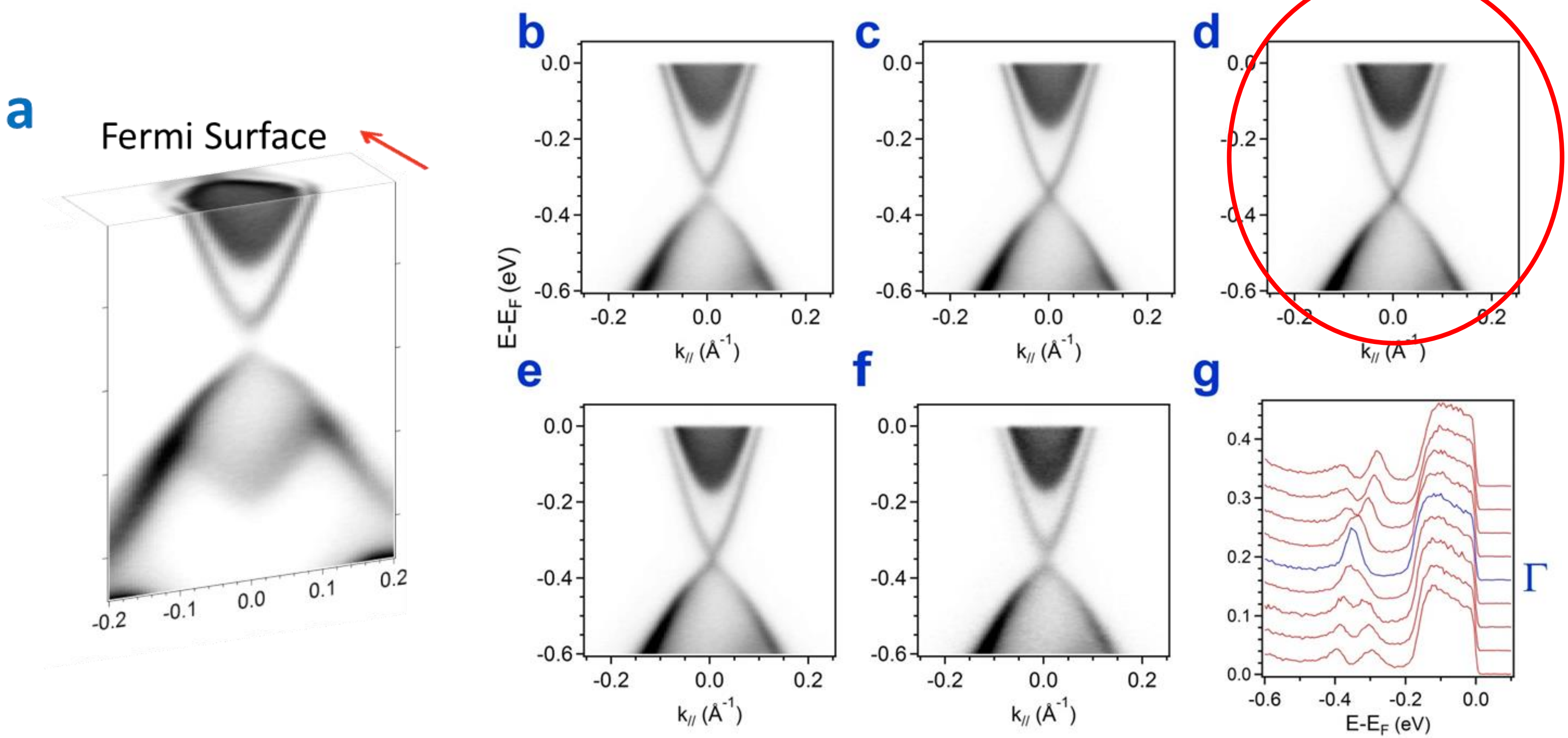
Chen et al, Science (2009)

Xia et al., Nature Physics (2009)

Xiao-Liang Qi and Shou-Cheng Zhang, RMP (2011)

Hsieh et al., Nature (2009)

Topological Insulators probed by ARPES



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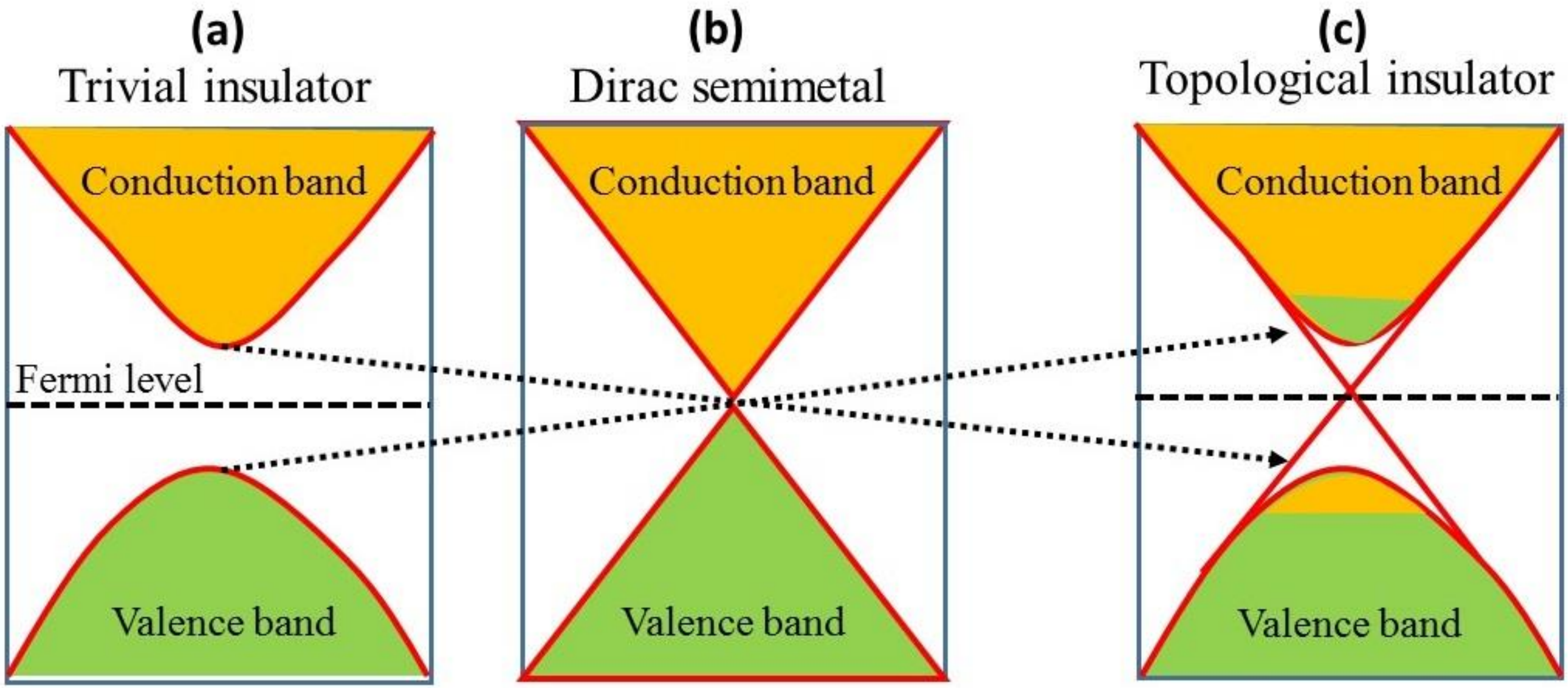
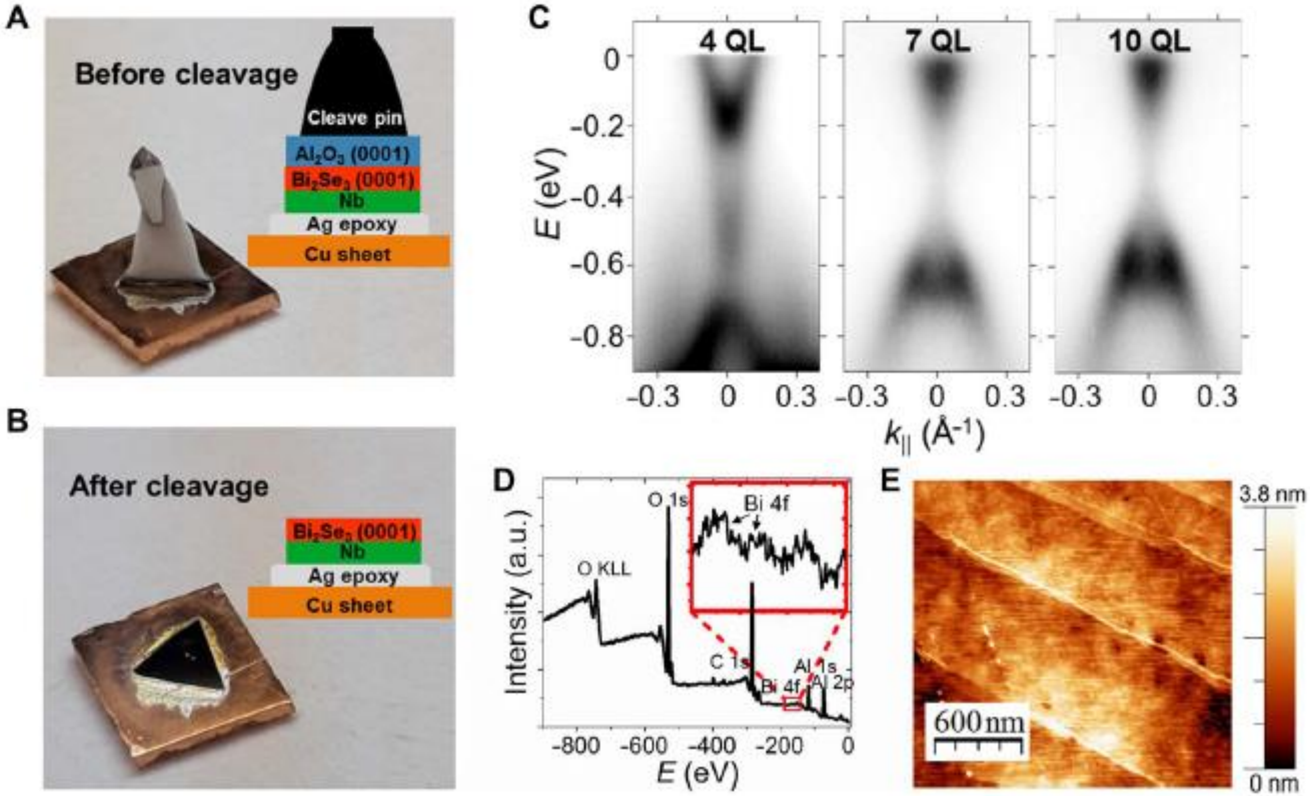
Scientific Cases: Dirac semimetal and Weyl semimetal



Proximity effect in topological insulators

Superconducting pairing of topological surface states



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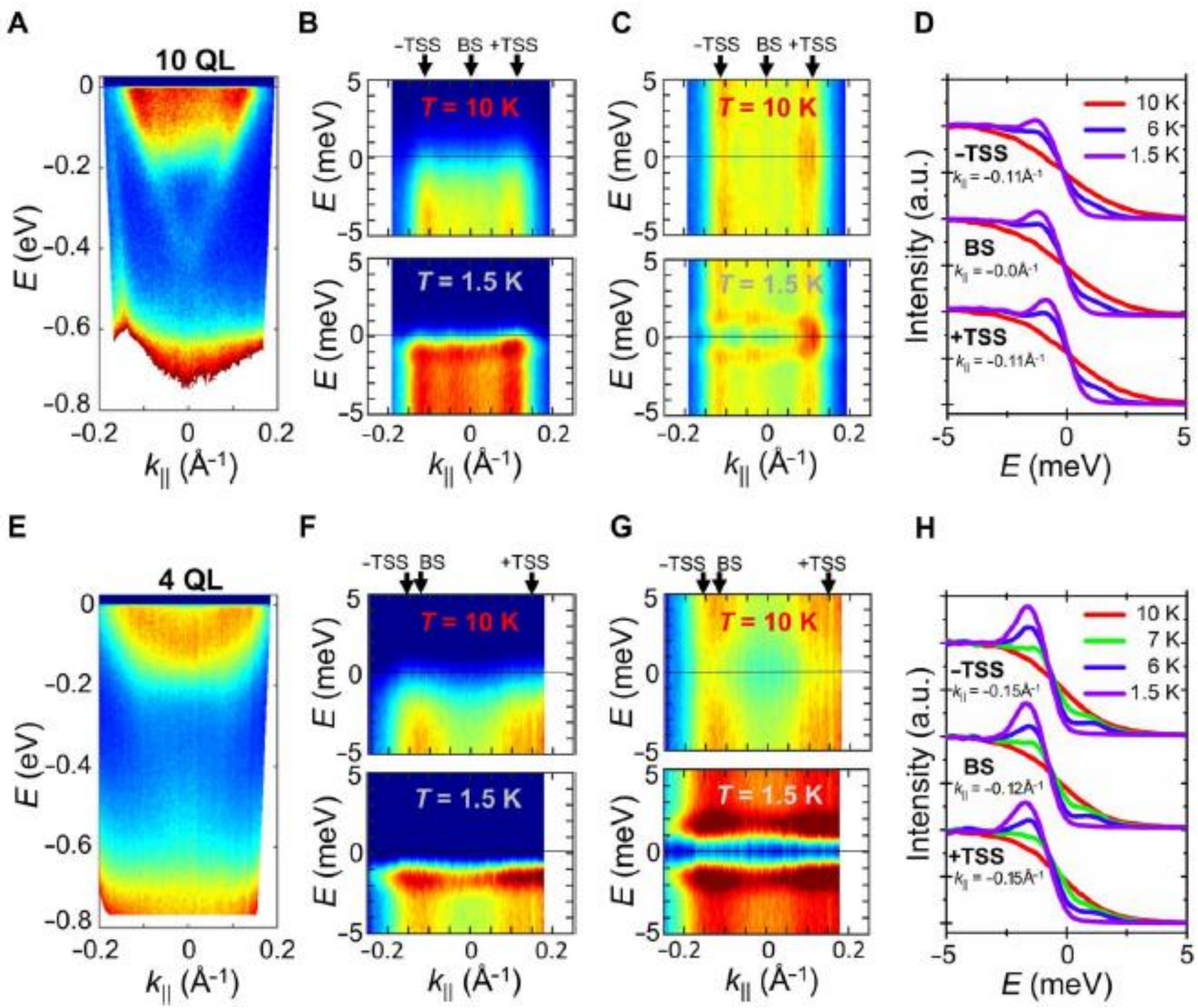
Flötotto et al. Science Advance (2018)

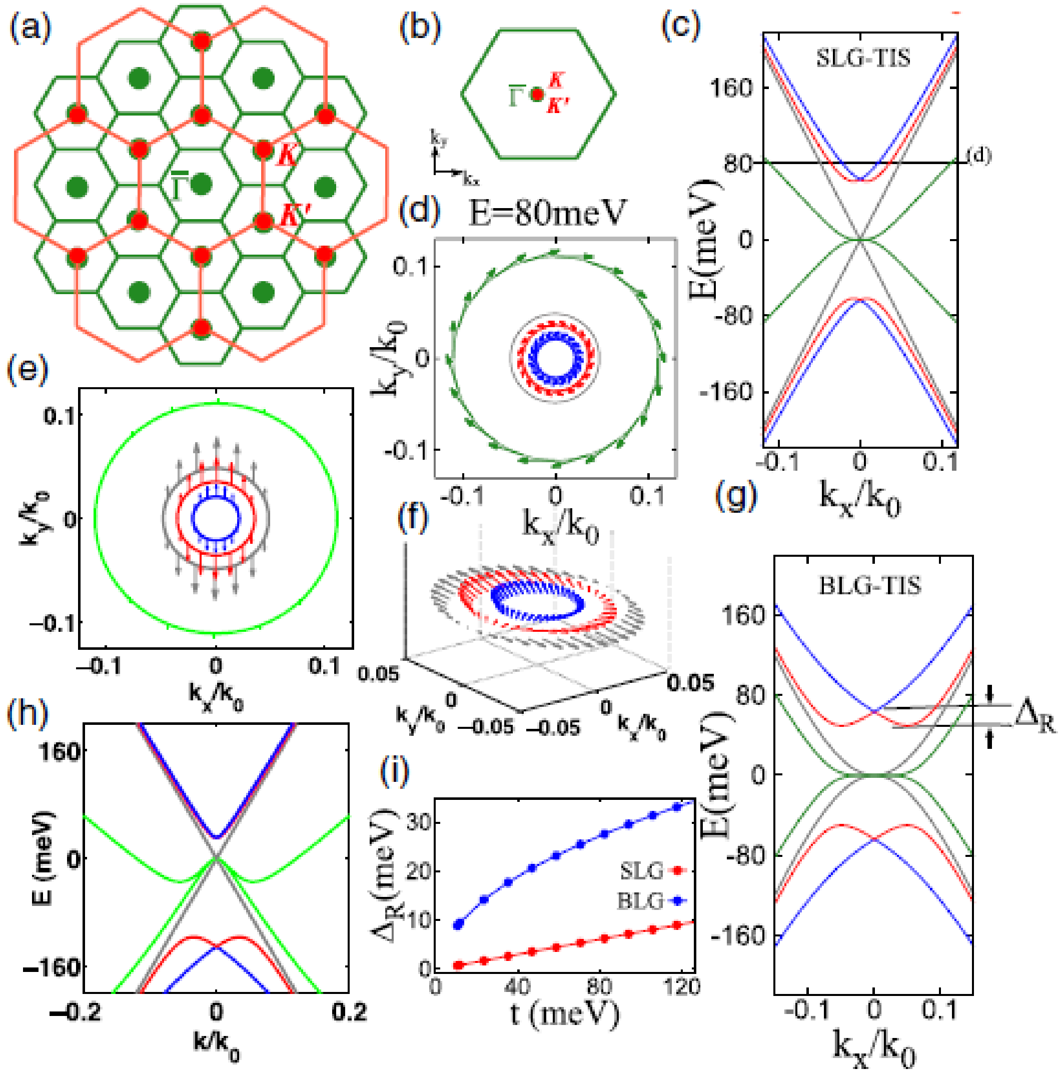
Proximity effect in topological insulators



Flötotto et al. Science Advance (2018)

Proximity effect in topological insulators

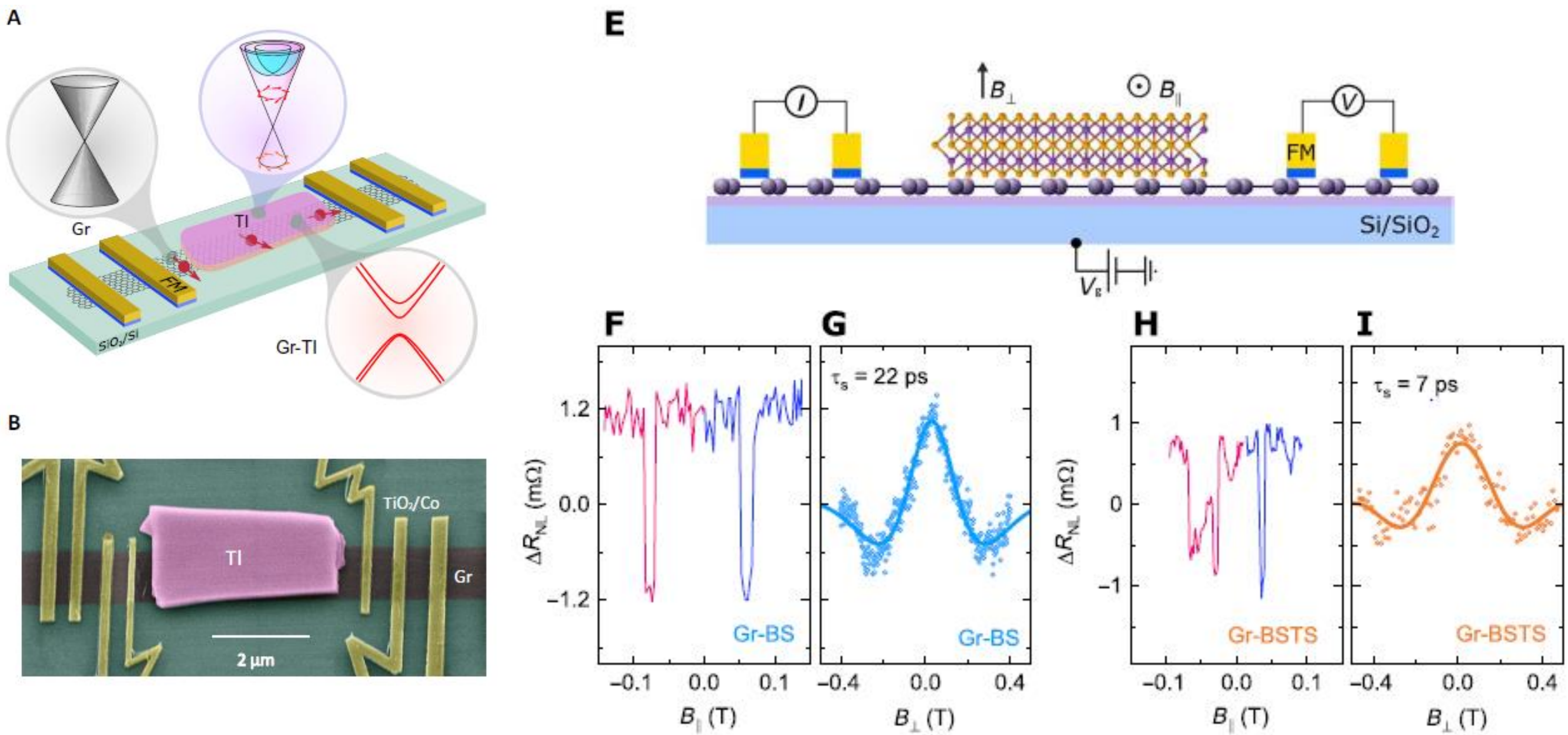
Proximity Effect in Graphene–Topological-Insulator Heterostructures



Junhua Zhang, PRL (2014)

Proximity effect in Gr-Topological insulator heterostructure

A Gr-TI heterostructure channel and ferromagnetic (FM) tunnel contacts for spin injection and detection in a nonlocal transport geometry



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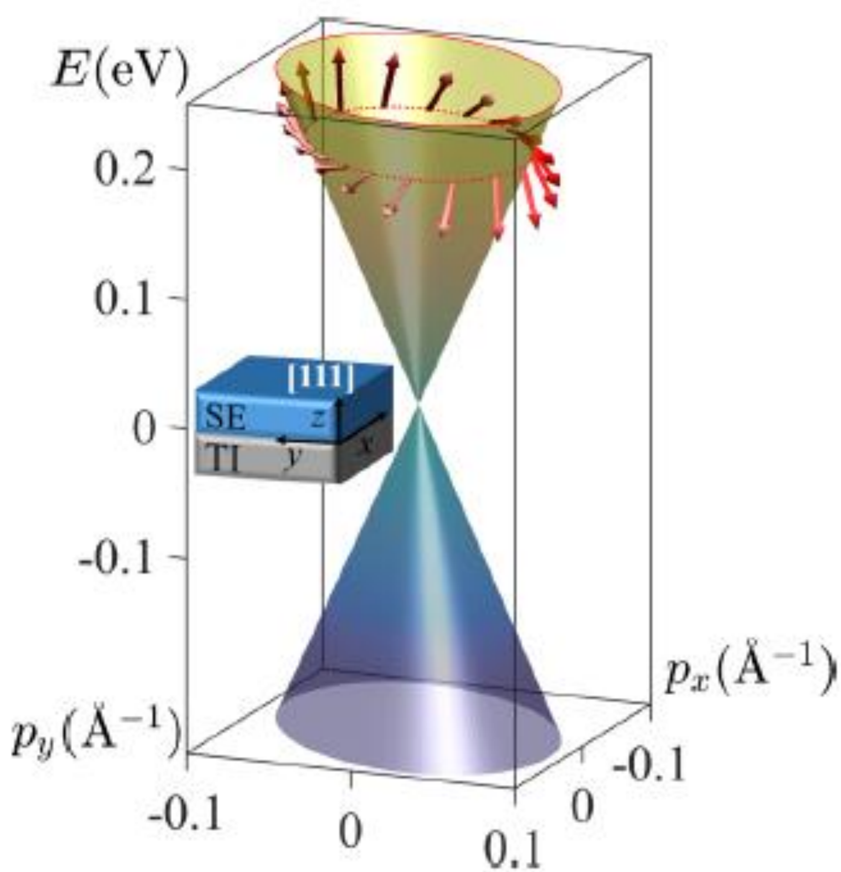
Khokhriakov et al., Science Advance (2018)

Can we control the spin texture with SE/TI heterostructure……



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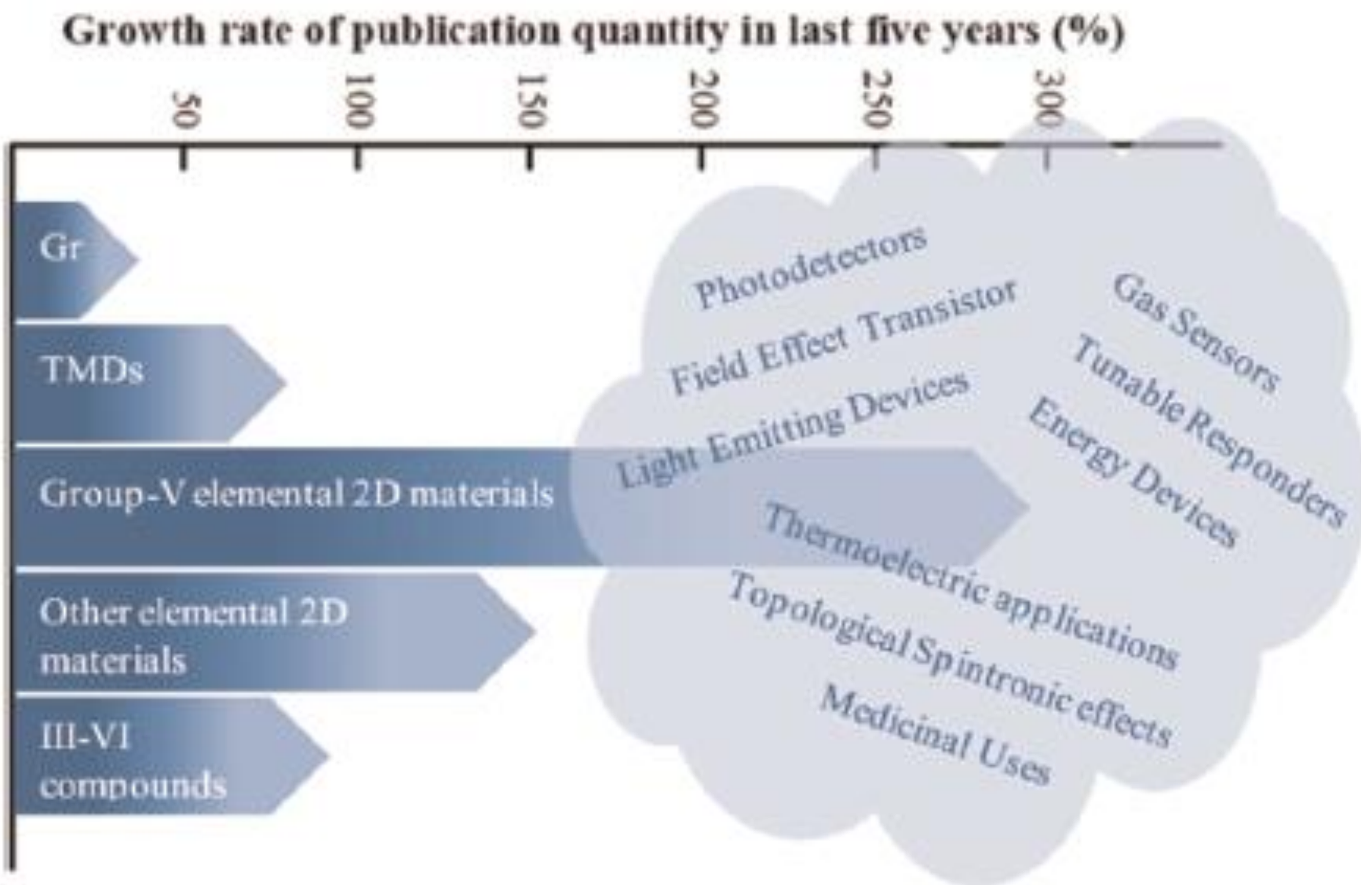
Mahmoud M. Asmar et al., PRB (2017)

Motivation for studying TI hybrid nanostructure

Group ▶ / Period ▼	10	11	12	13	14	15	16	17	18
2				5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
3				13 Al Aluminium	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
4	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
5	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
6	78 Pt Platinum	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
7	110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson

2D Group V monoelemental monolayer



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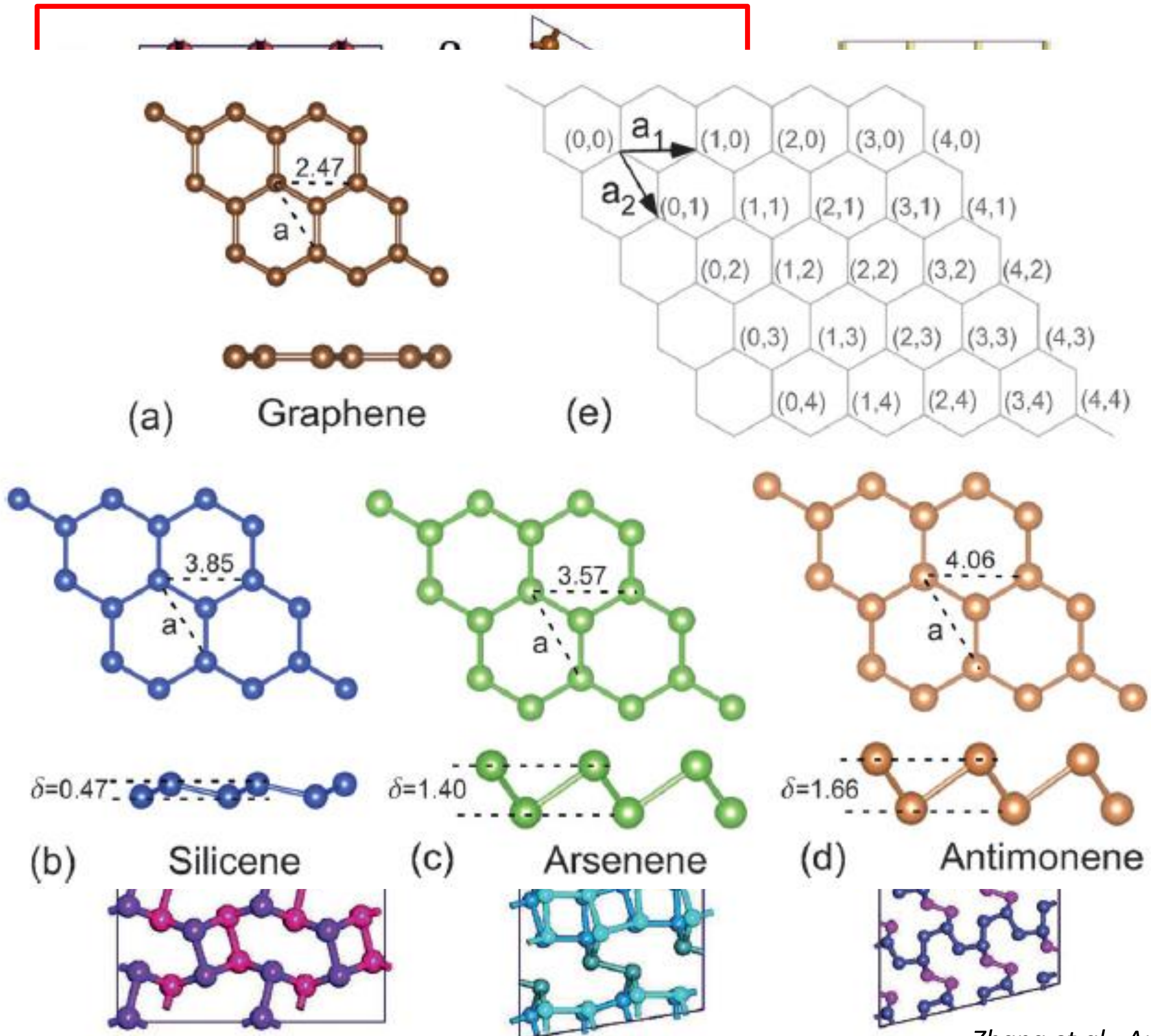
Wu et al., npj 2D Materials and Applications (2020)

Structure configuration of 2D group V monoelemental materials



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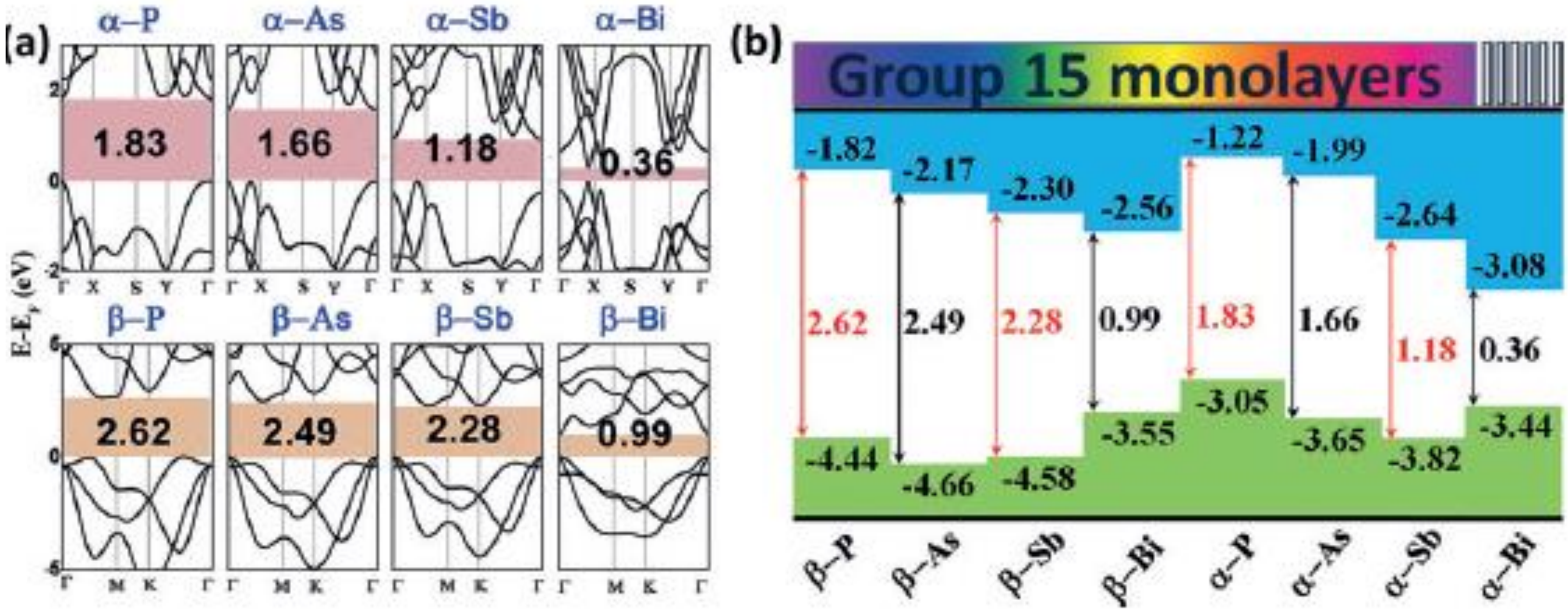
Zhang et al., Angew. Chem. (2015)
Yang, Phys. Chem. Chem. Phys (2015)

Band gap of group 15 monolayers



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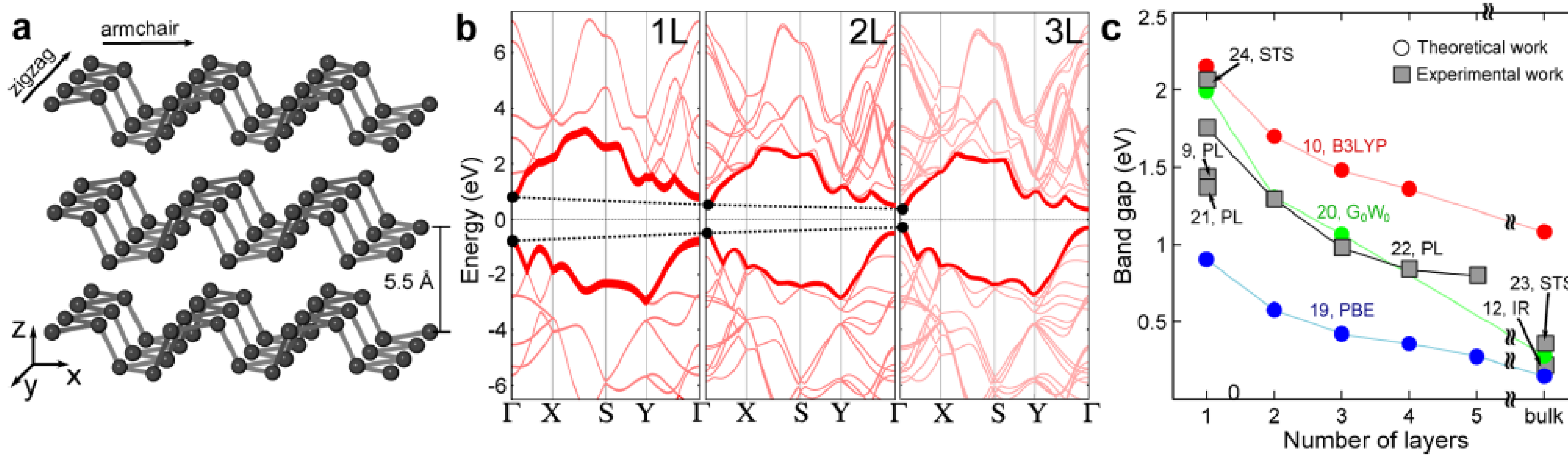
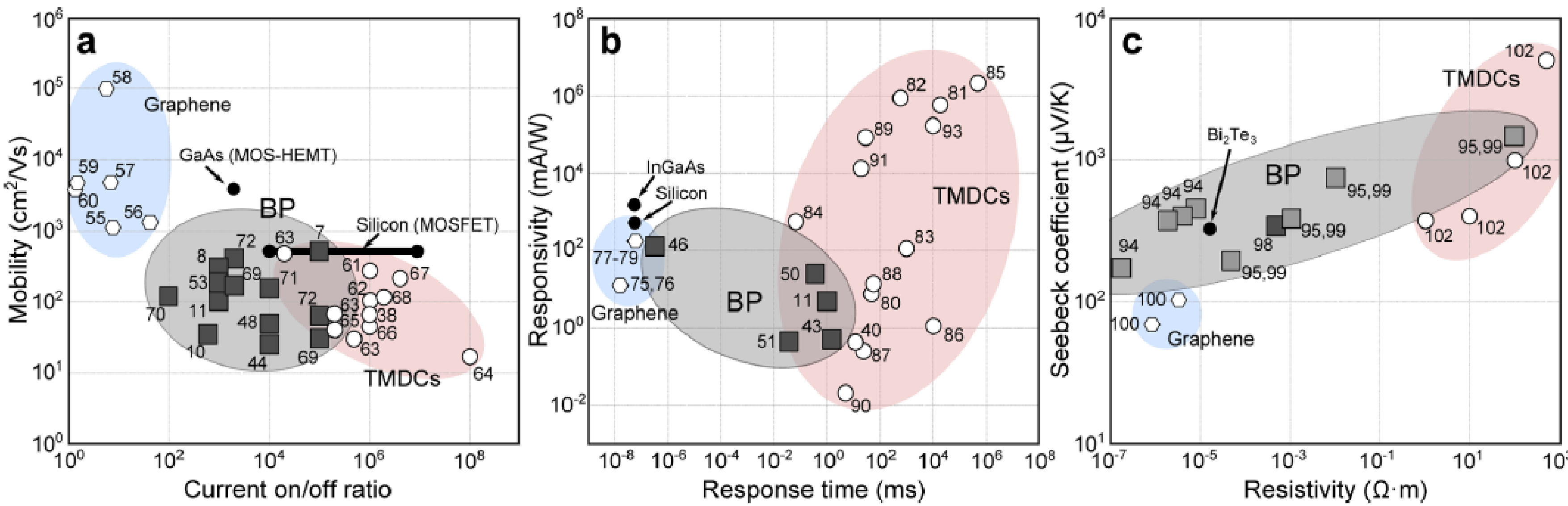
Zhang et al., Angew. Chem. (2015)

The electronic structure of black phosphorus

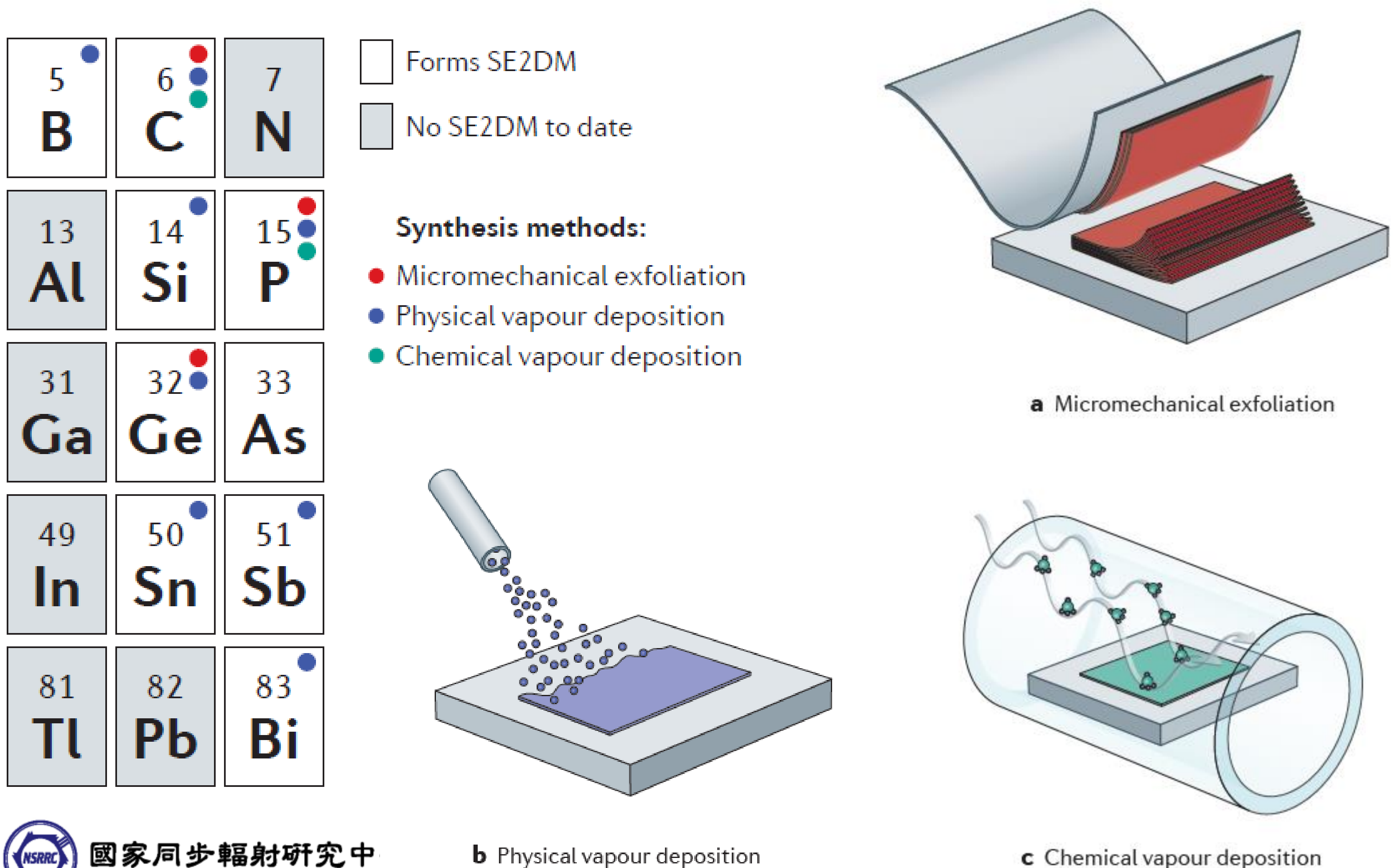
Black phosphorus (BP)





Catellanoas-Gomez, J. Phys. Chem. Lett. (2015)

Synthesis and chemistry of elemental 2D materials



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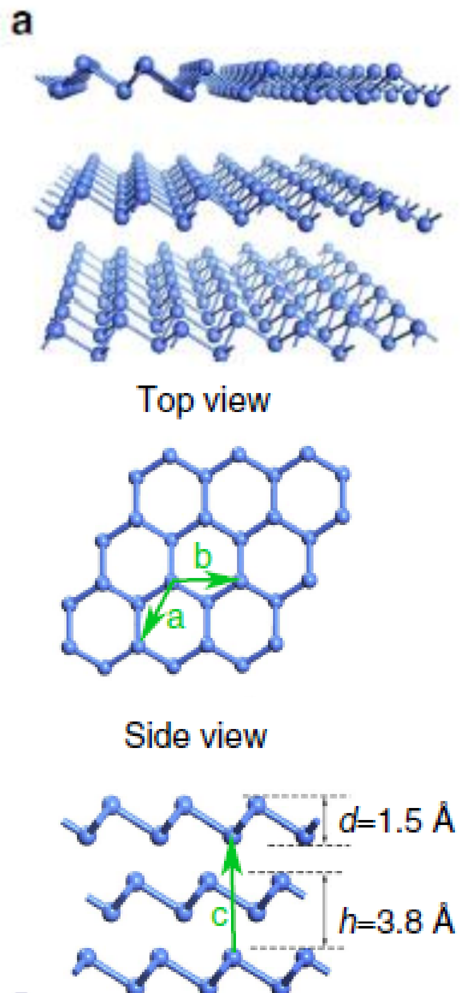
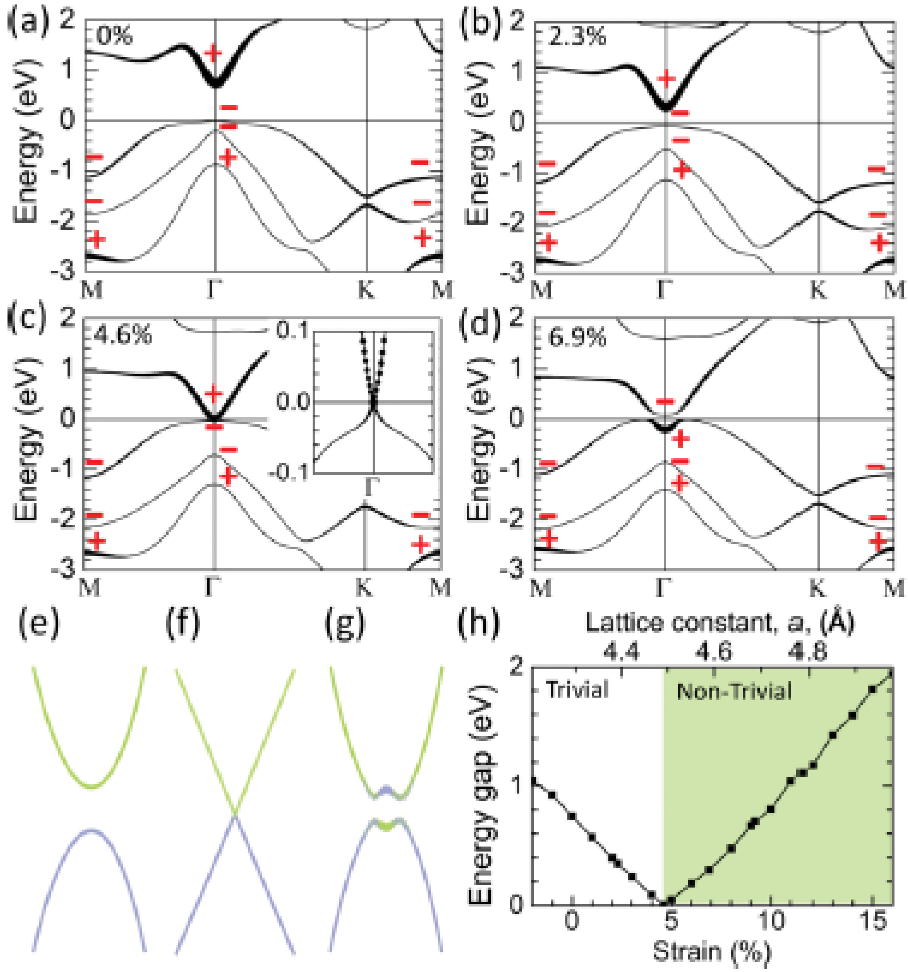
Mannix, Nat. Rev. Chem. (2017)

A topological phase transition in Sb bilayer





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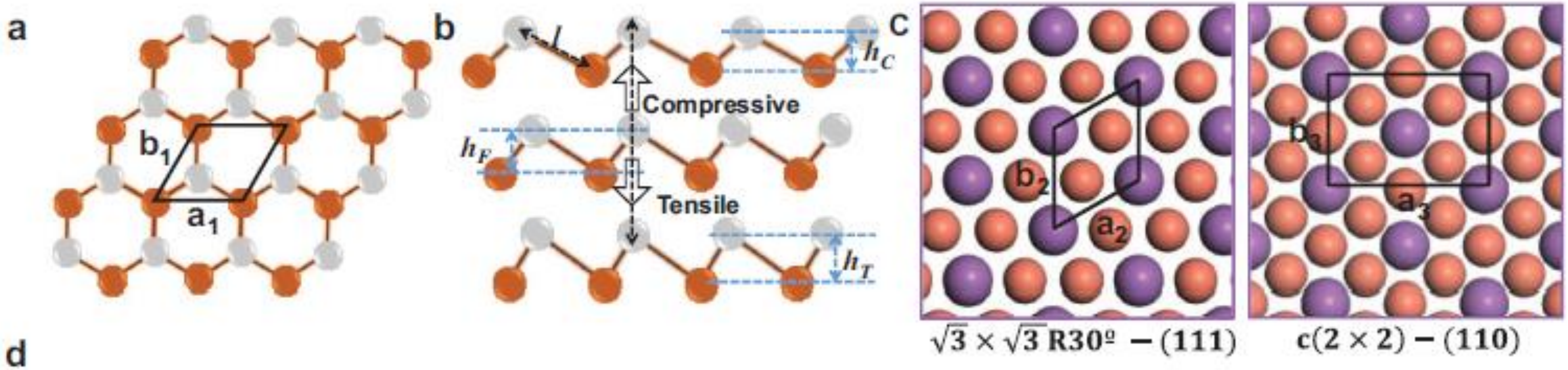
Chuang et al., APL (2013)
Ji et al., Nat. Comm. (2016)

Synthesis and chemistry of elemental 2D materials



d

β-antimonene	Free-standing	Tensile Strain	Ultra-flat	Tensile Strain	Compressive Strain
Supporting Substrate	No	$PdTe_2$	Ag(111)	Sb-Cu(111)	Sb-Cu(110)
Lattice Constant (a, Å)	4.12	4.13	5.01	4.43	3.84
Buckling Height (h, Å)	1.65	1.65	0	1.42	1.78
Sb-Sb bond length (l, Å)	2.89	2.89	2.89	2.93	2.95
References	2c	10c	10e	This work	This work

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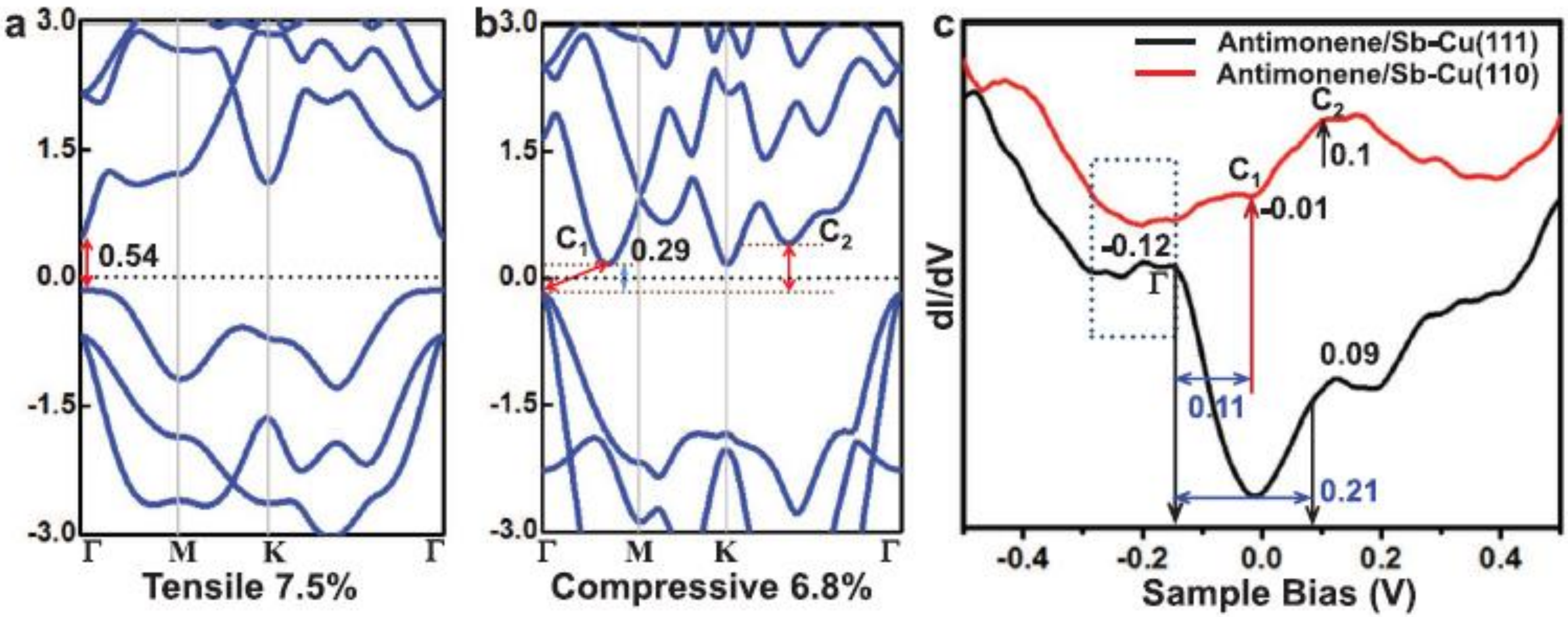
Tianchao Niu et al., Adv. Mater. (2019)

Synthesis and chemistry of elemental 2D materials

Cu(111) : 4.43 Å
Cu(100) : 3.84 Å



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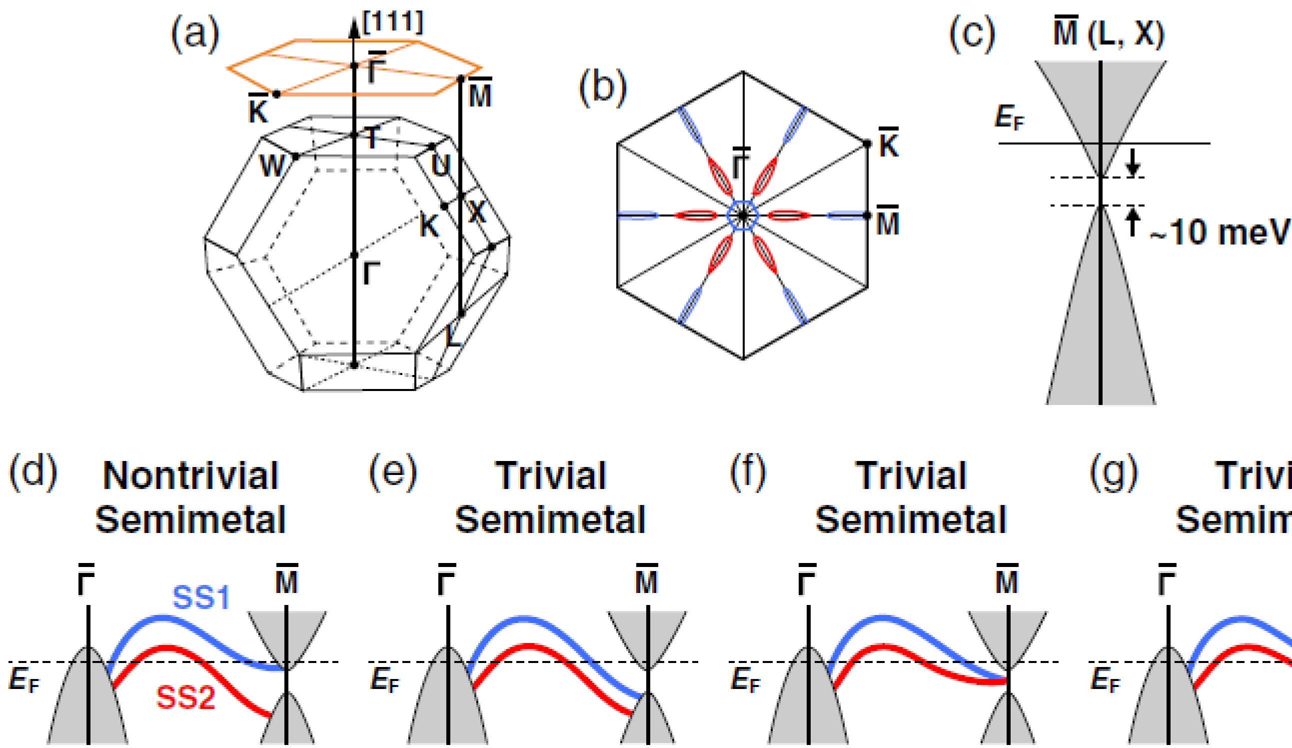
Tianchao Niu et al., Adv. Mater. (2019)

Nontrivial Topology of Pure Bismuth



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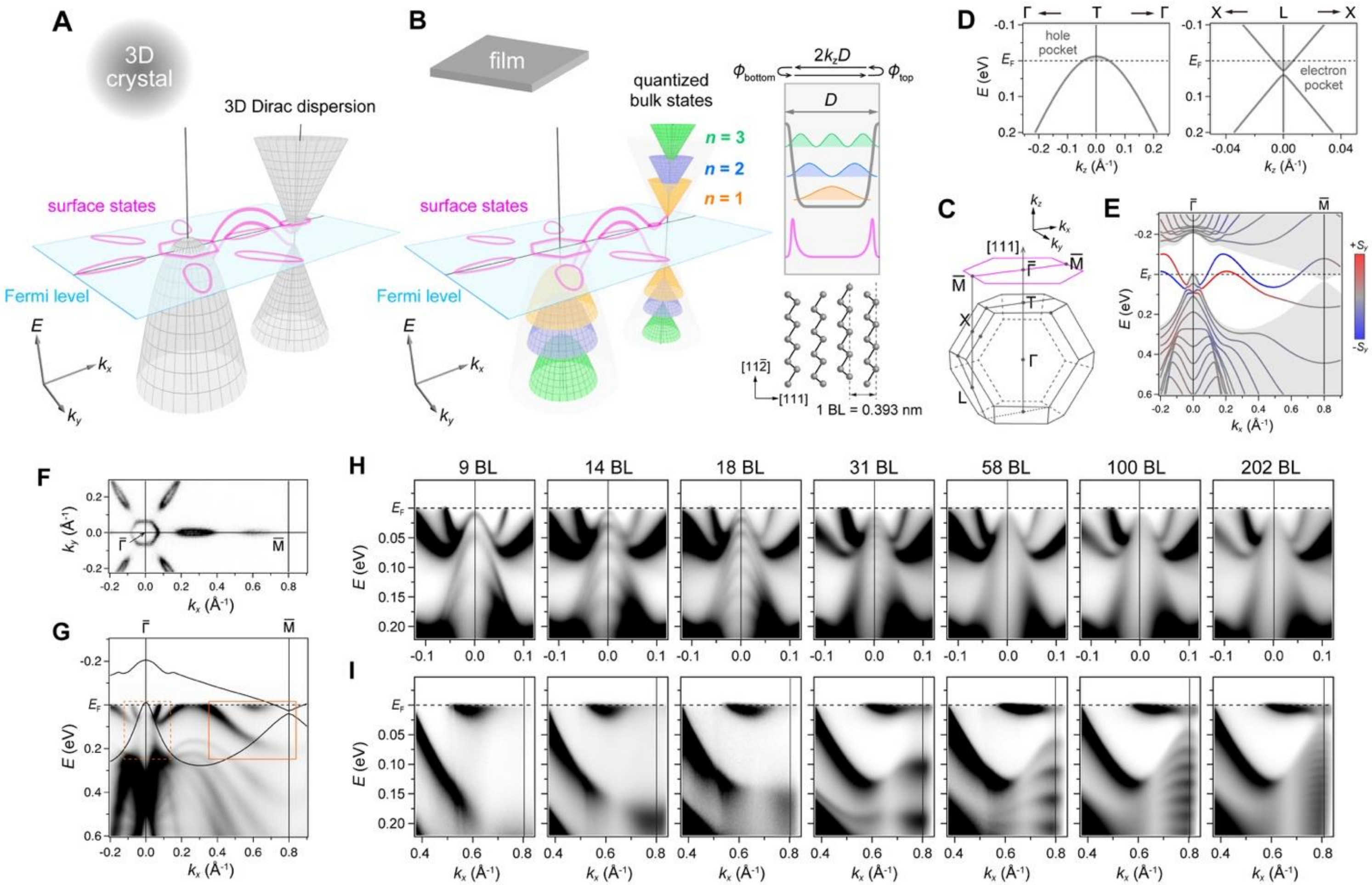
Ito et al. PRL (2016)

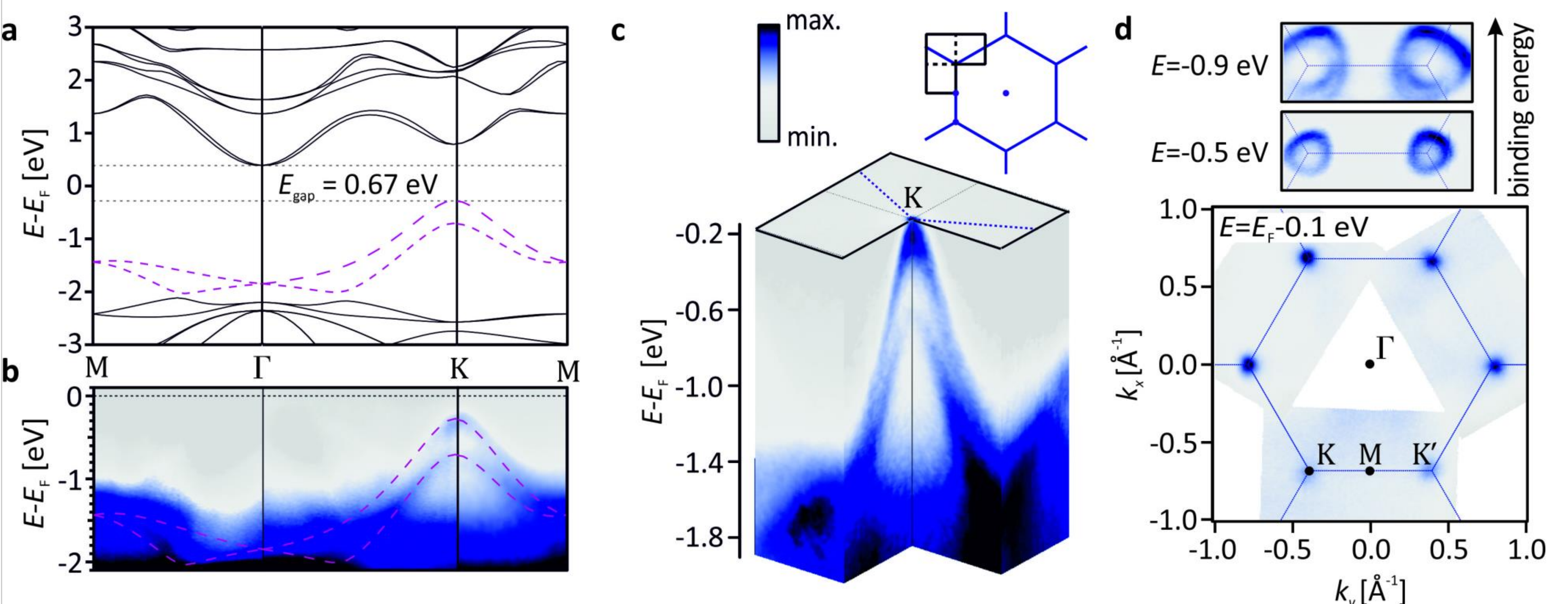
Nontrivial Topology of Pure Bismuth



Ito et al. Science Advance (2020)

Nontrivial Topology of Pure Bismuth

Bismuthene on a SiC Substrate



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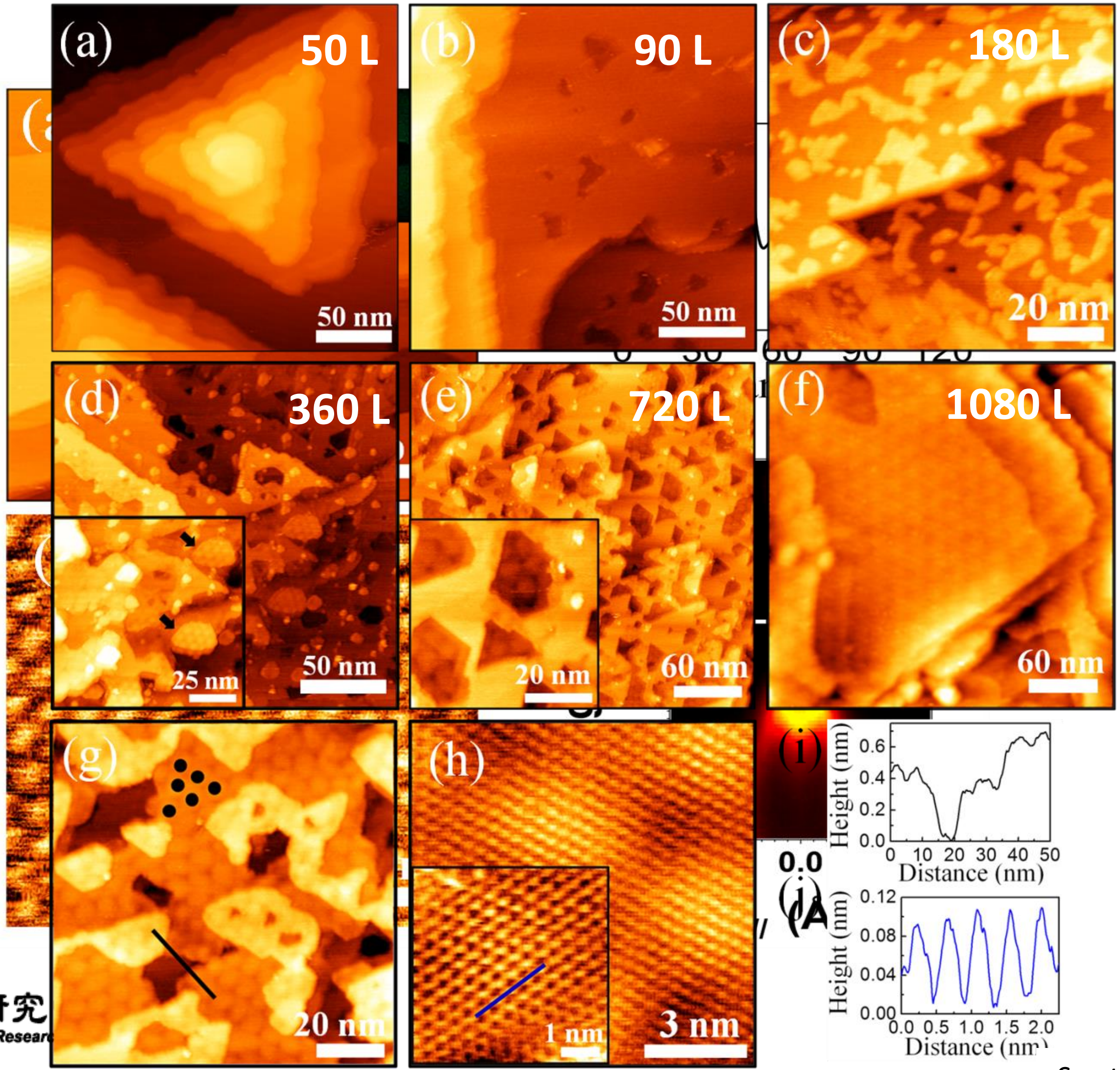
Reis et al. Science (2017)

The fabrication of bismuthene and antimonene on TIs

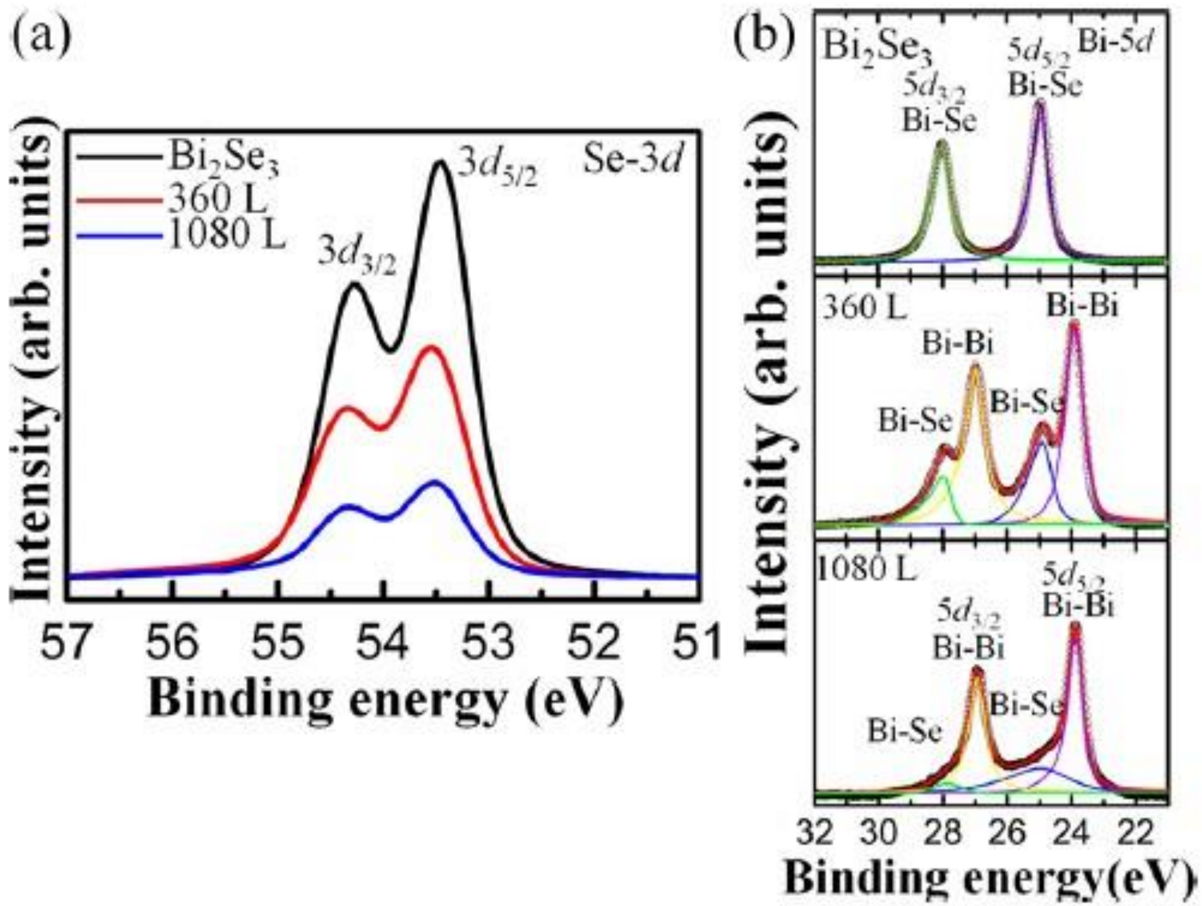
The fabrication of bismuthene on TIs

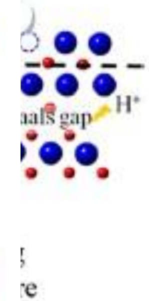
Bi-BLs/Bi_2Se_3 prepared with the atomic hydrogen etching method



Su et al., Chem. Mater. (2017)

The fabrication of bismuthene on TIs





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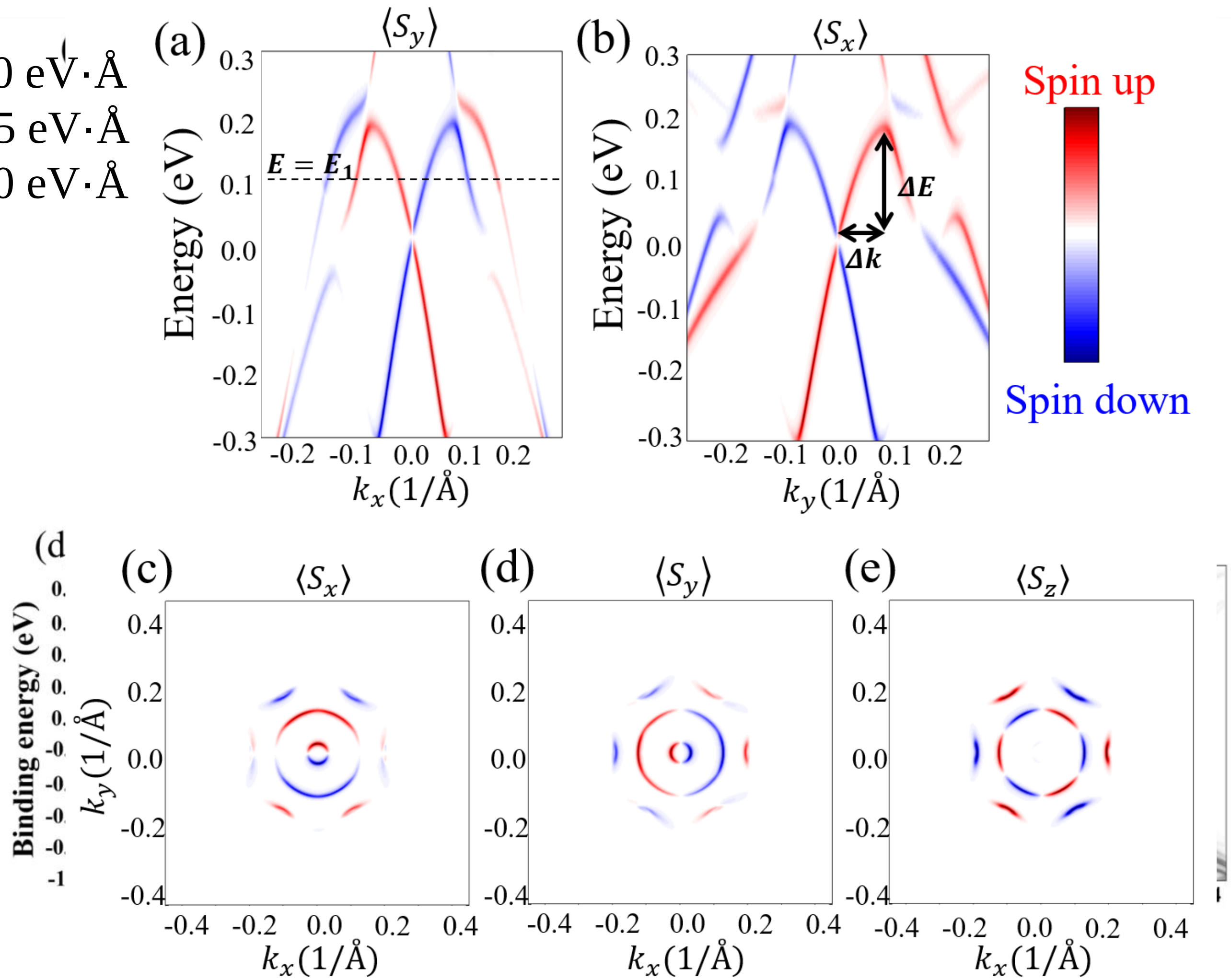
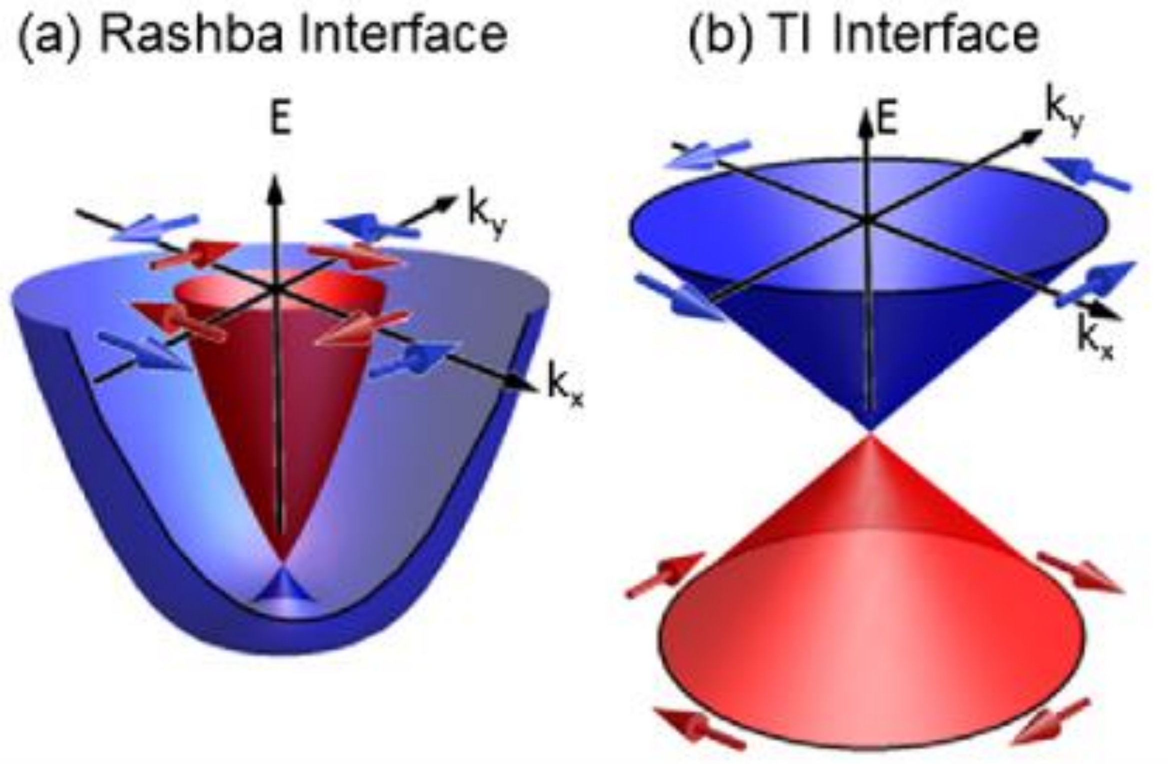
The fabrication of bismuthene on TIs

$$\alpha = \frac{2\Delta E}{\Delta k} \sim 4.5\ \mathrm{eV \cdot \AA}$$

Bi/Ge(111)~1.80 eV·Å
Bi/Ag(111)~3.05 eV·Å
BiTeI ~3.80 eV·Å

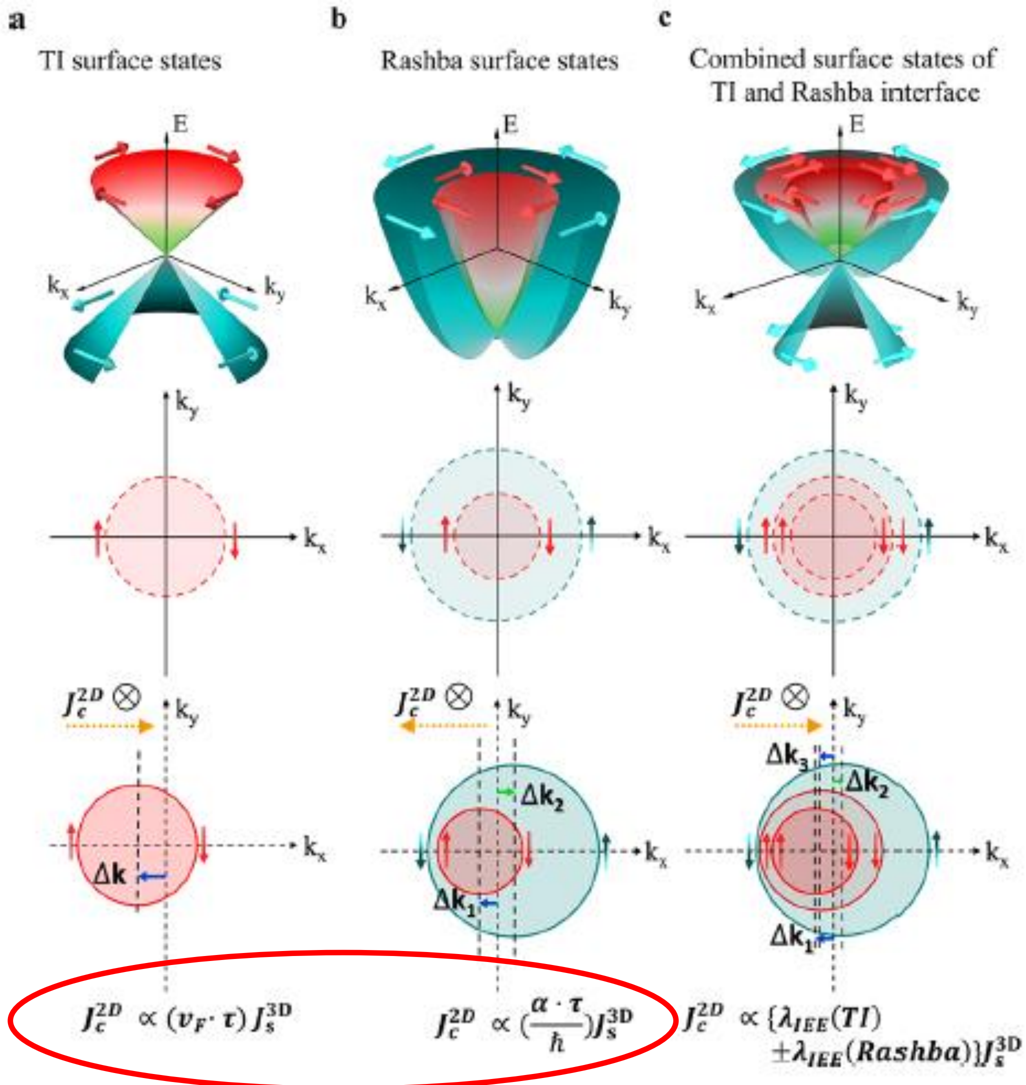


Proximity effect in topological insulators



J.-C. Rojas-S'anchez et al., PRL (2016)

Tunable Spin-to-Charge Conversion in TI hybrid-structure



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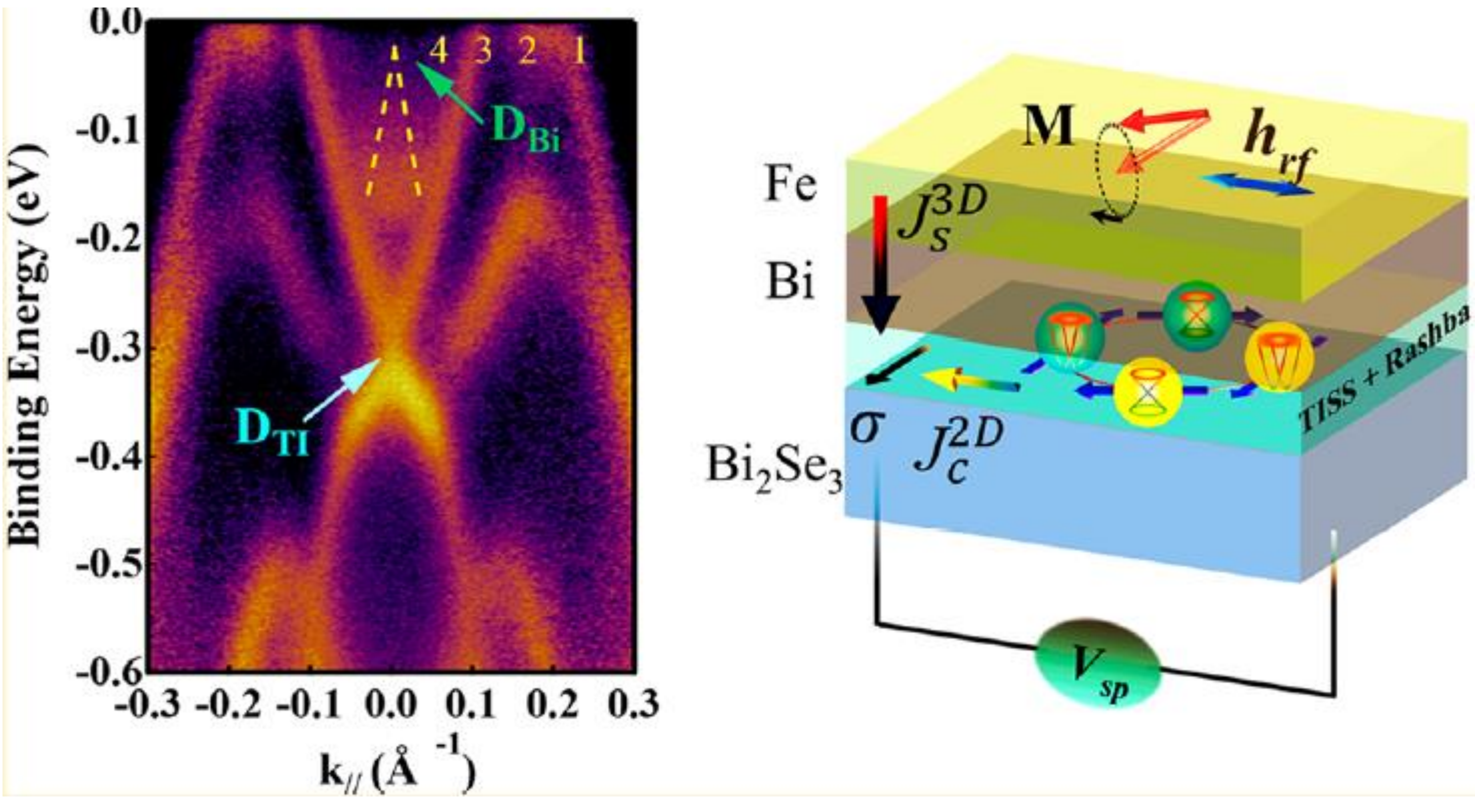
Rui Sun etal., Nano Lett. (2019)

Tunable Spin-to-Charge Conversion in TI hybridstructure

Incorporating a "second" spin-splitting band, a Rashba interface formed by inserting a bismuth interlayer between the ferromagnet and the Bi_2Se_3 (i.e., ferromagnet/Bi/ Bi_2Se_3 heterostructure), λ_{IEE} shows a pronounced increase (up to 280 pm) compared with that in pure TIs.



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Rui Sun etal., Nano Lett. (2019)

Future and outlook

- *Spin and dynamic behavior*
  - *Topological insulator, 2D materials, superconductors, complex oxides, graphene-based materials….*
  - *Time-resolved ARPES.*
  - *Spin-resolved ARPES*

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  - *Single crystals with broken surface after in-situ cleaving.*
  - *Gating Effects in 2D Materials*

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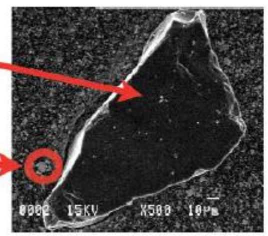
Nano Everywhere

Materials of interest often contain intrinsic, extrinsic, and designed nanoscale features.

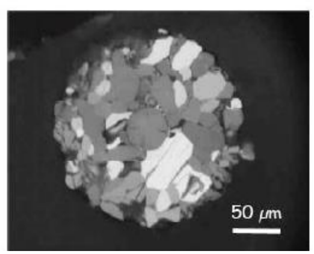
Need rapid electronic structure mapping at the nanoscale

Large MgB_2 single crystal ~200 μm- available two years after discovery

10 μm - available shortly after discovery

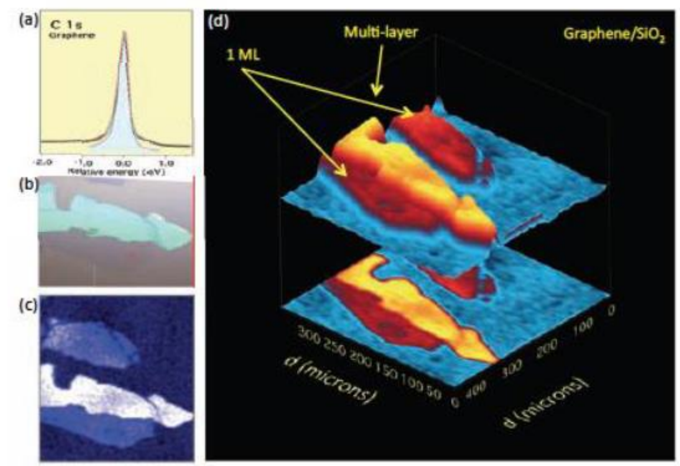


MgB_2 wire - available few months after discovery



IOWA STATE UNIVERSITY

Multi-thickness domains existed in an exfoliated graphene flake



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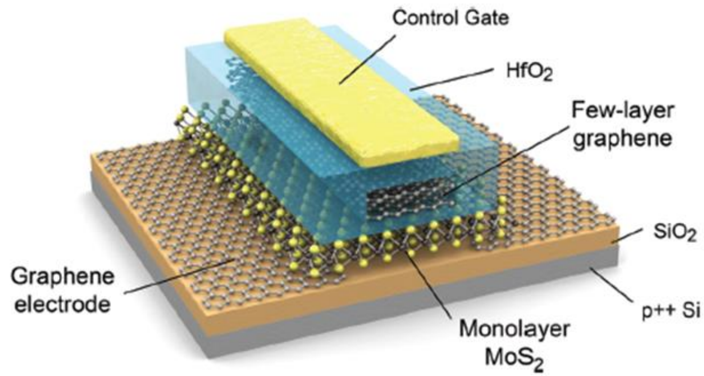
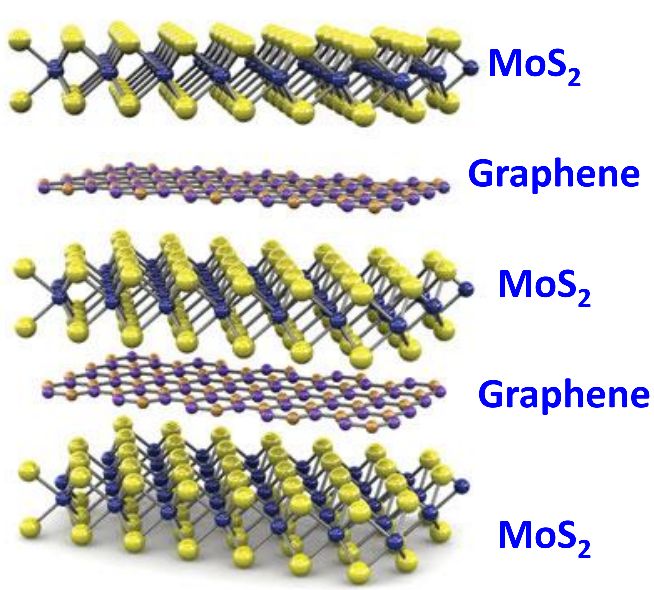
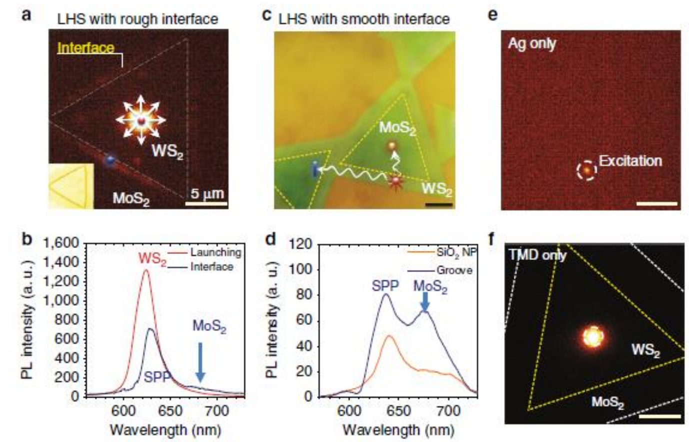
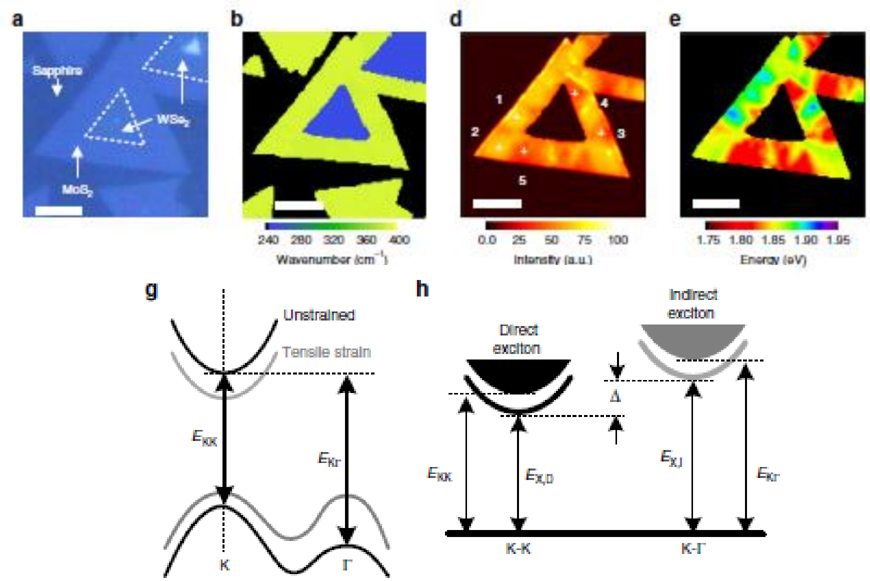
Ref : Avila, Synchrotron Radiation News (2014)

Van der Waals Heterostructures





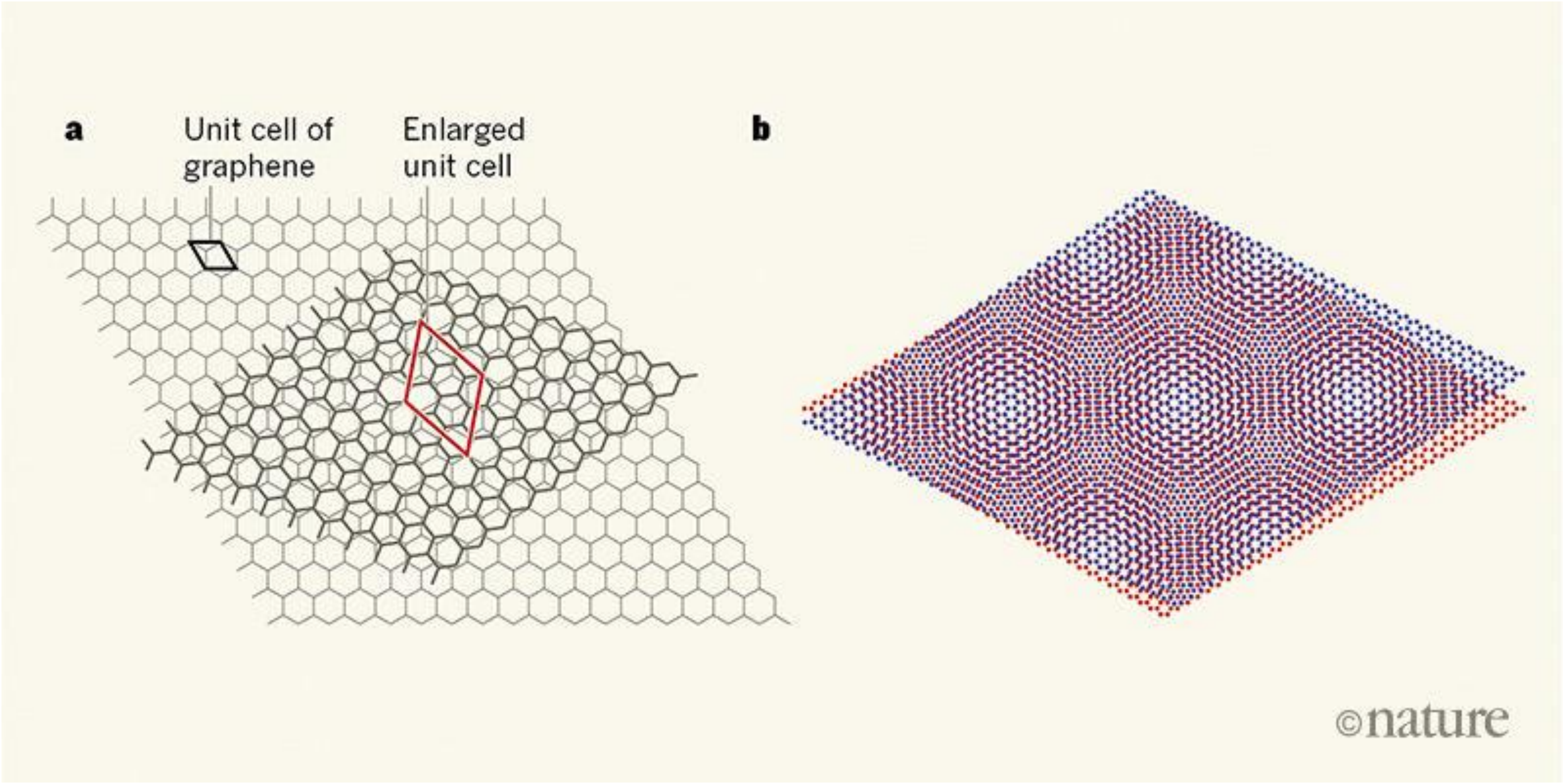




Ref : Shi et al., Nat. Comm. (2017)
Hsu et al., Nat Comm. (2017)

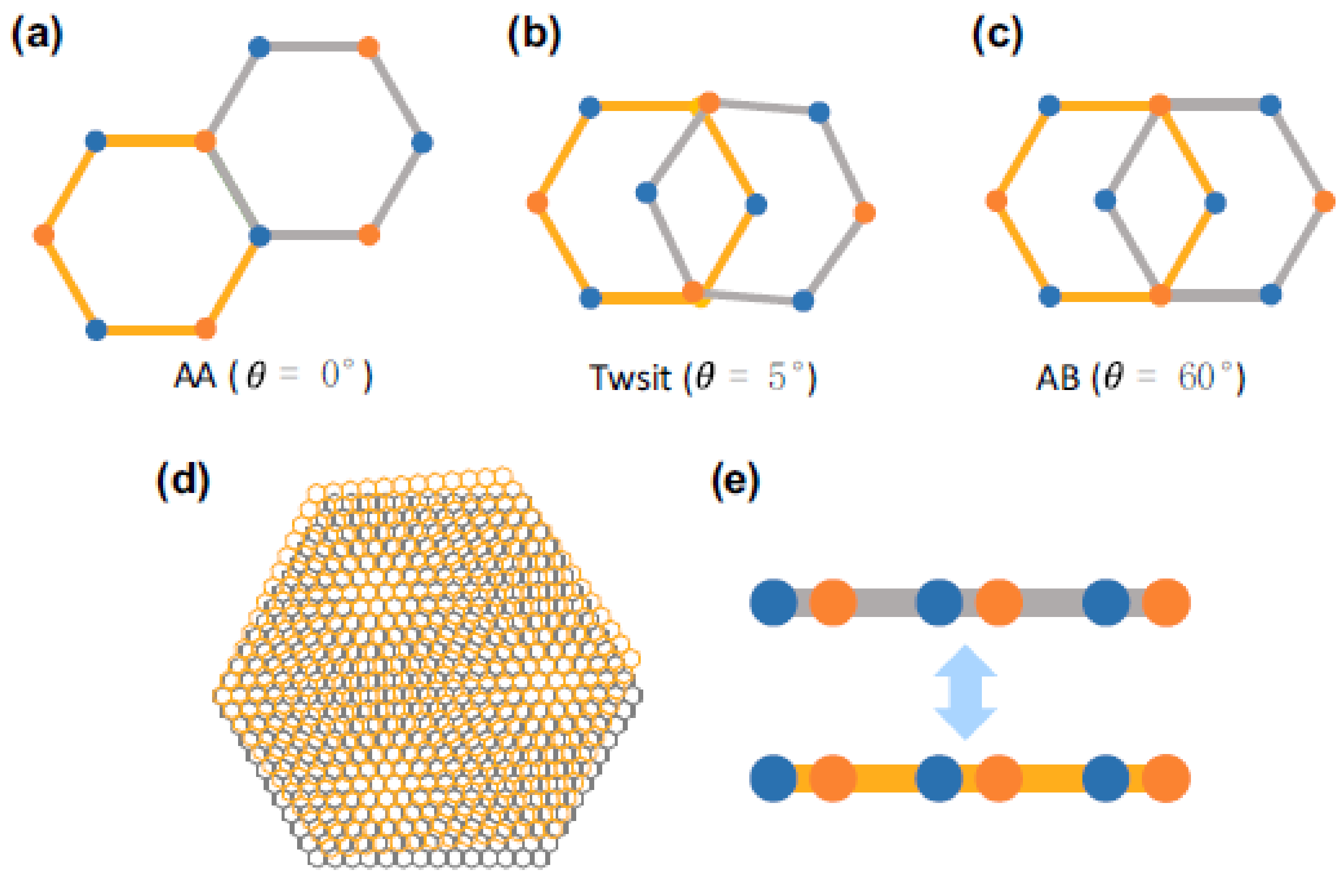
Magic Angle in Twisted Graphene

['Magic' Twist in Stacked Graphene Reveals Potentially Powerful Superconducting Behavior]



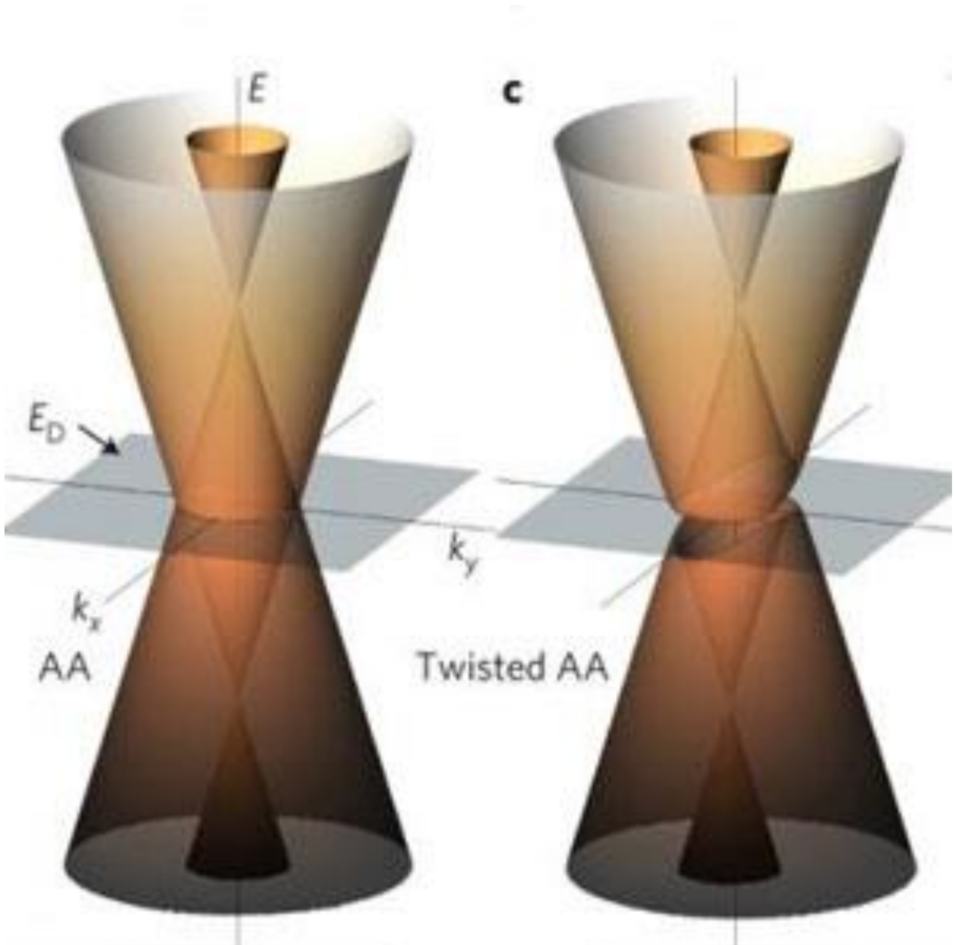
Scientific Cases: Twisted Bilayer Graphene

Moiré patterns and enhanced interlayer coupling in 2D twisted bilayer materials.



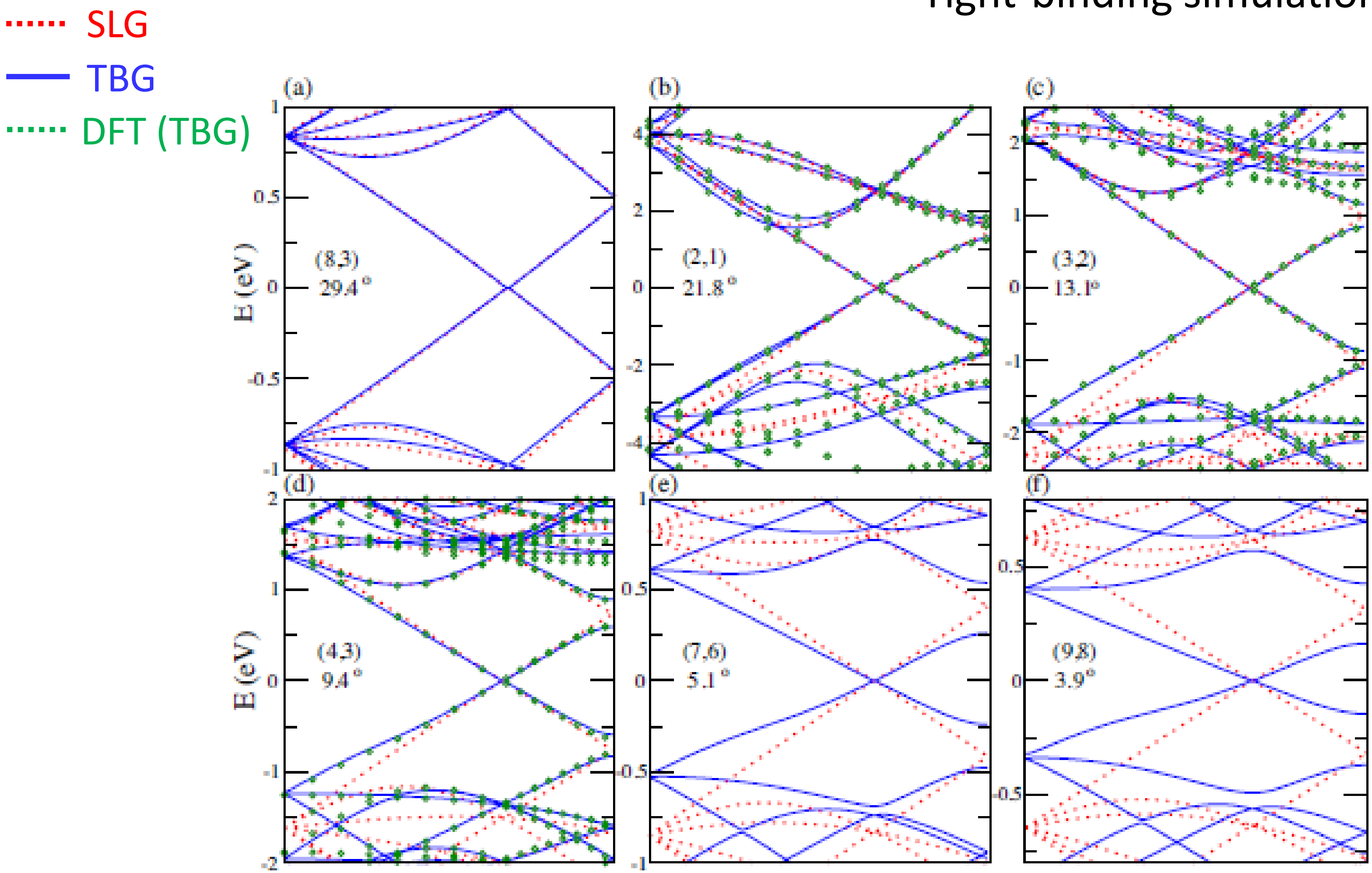
Twist (theta=5°)

Scientific Cases: Twisted Bilayer Graphene (TBG)



Scientific Cases: Twisted Bilayer Graphene (TBG)

Tight-binding simulation of TBG



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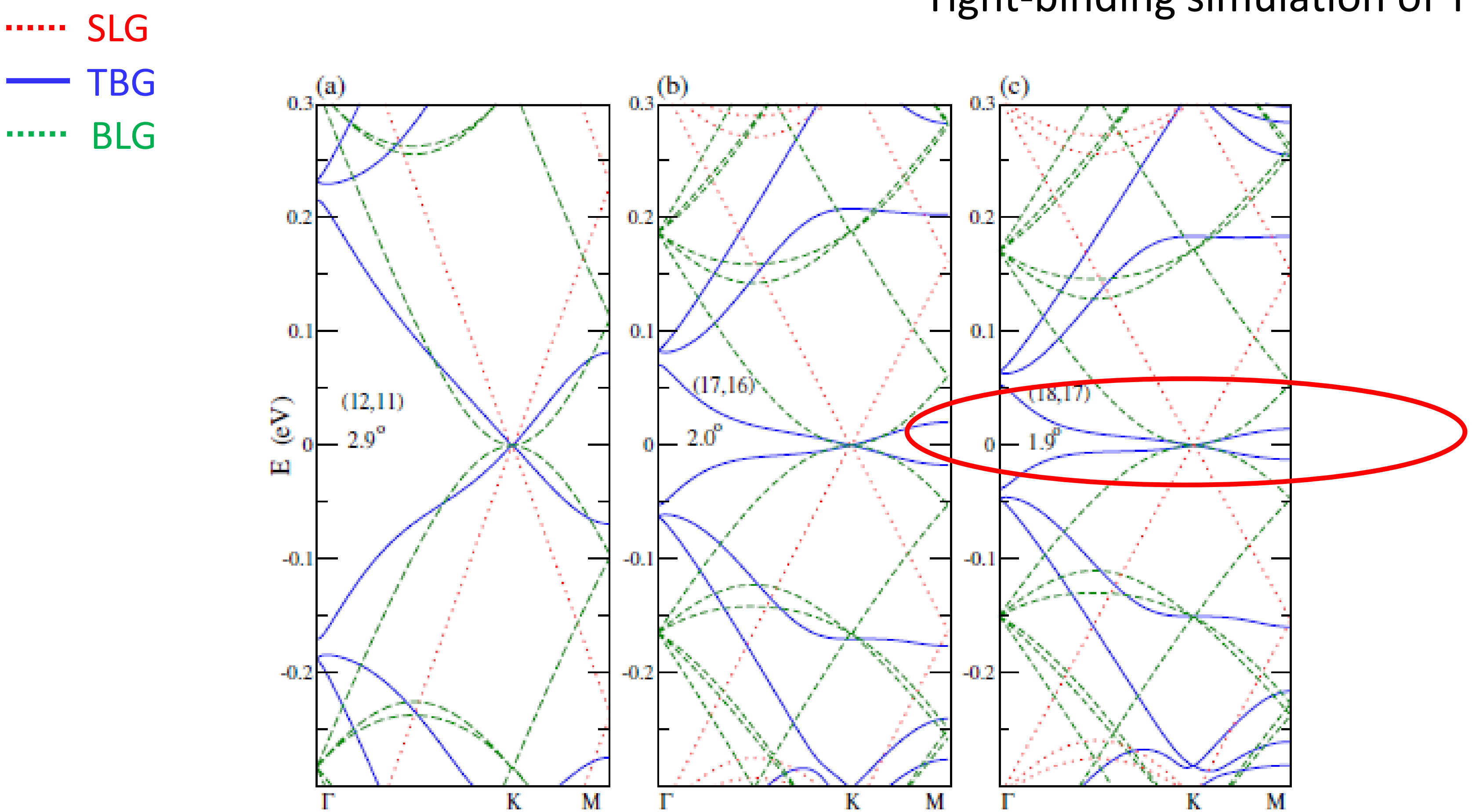
Tight-binding simulation of TBG



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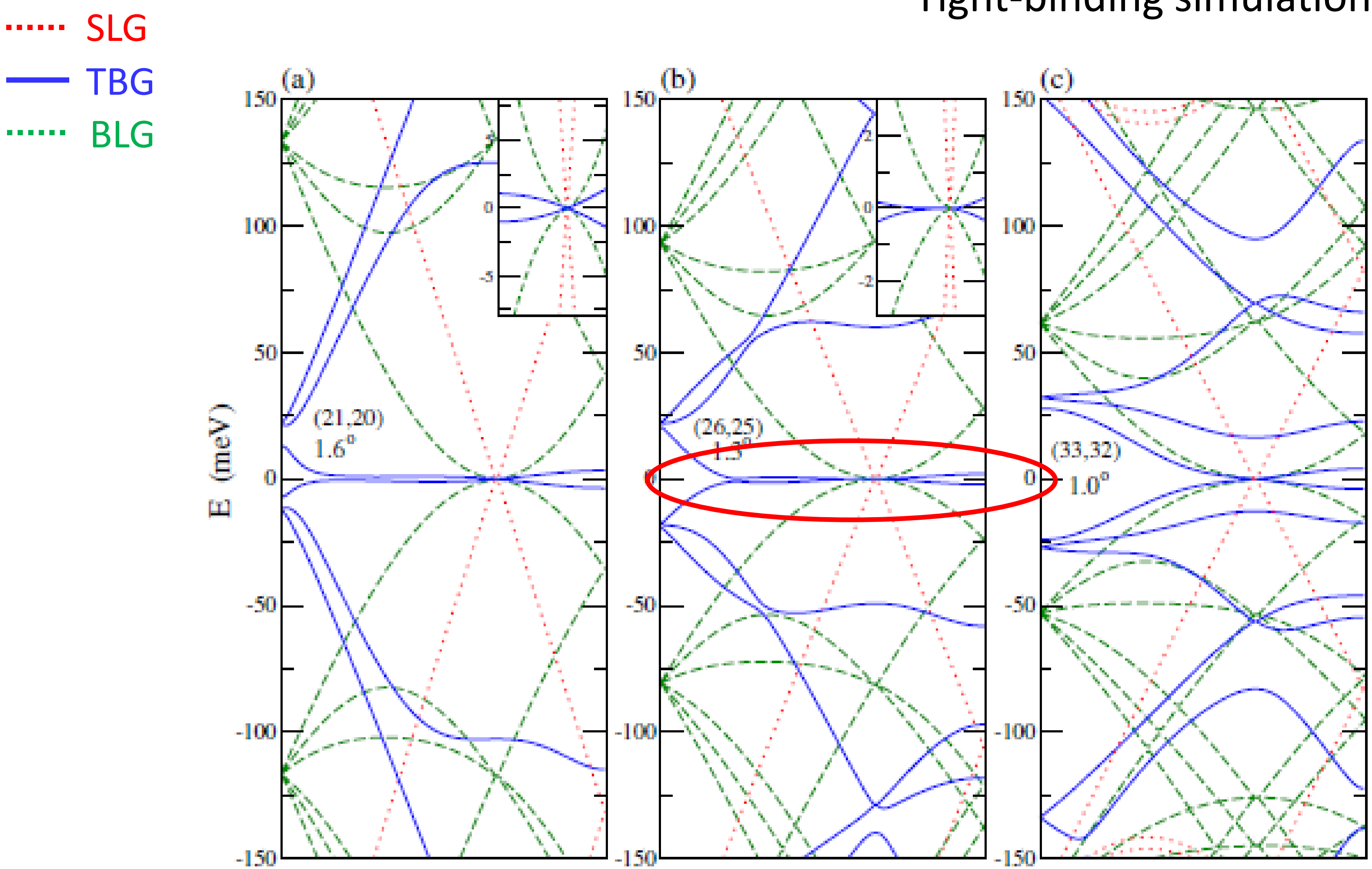
Tight-binding simulation of TBG



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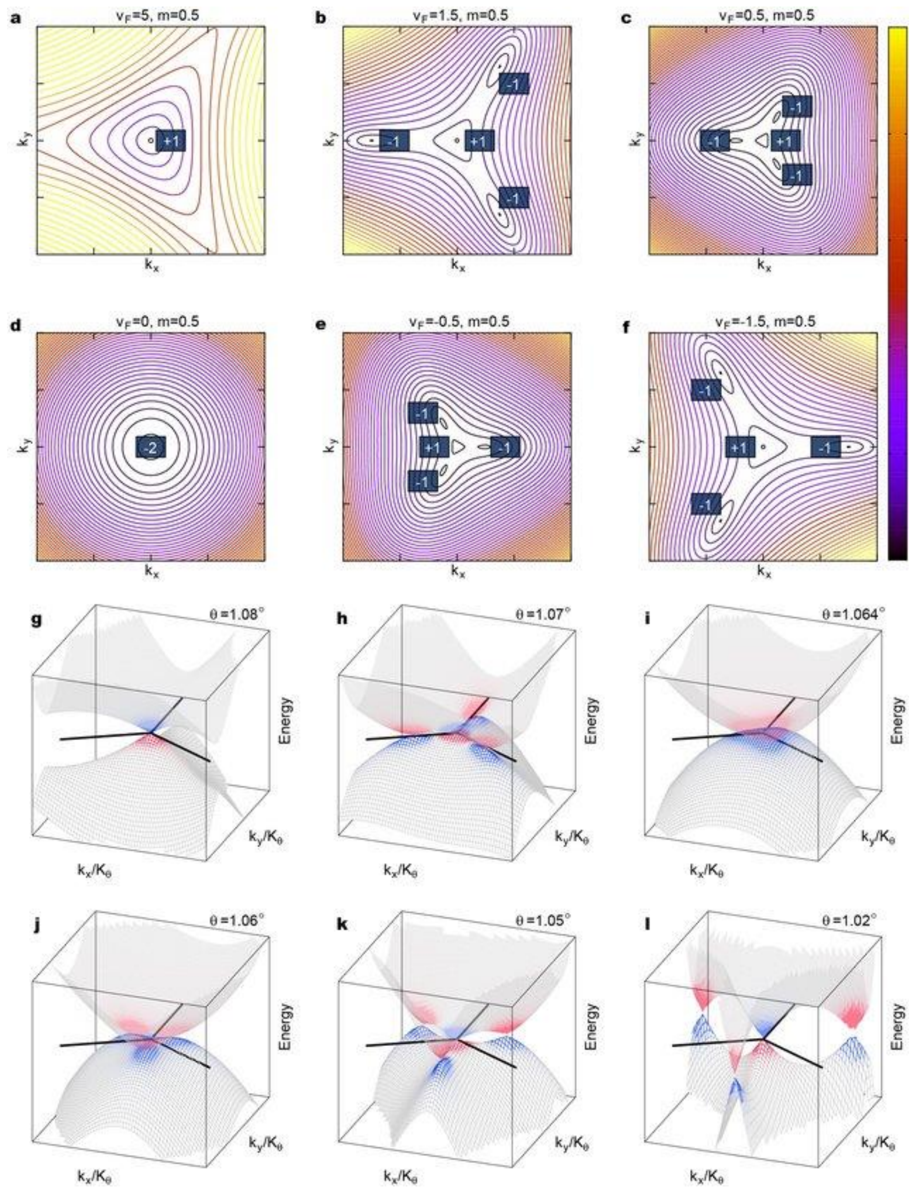
Scientific Case : Twisted Bilayer Graphene (TBG)

The simulated band structure of TBG



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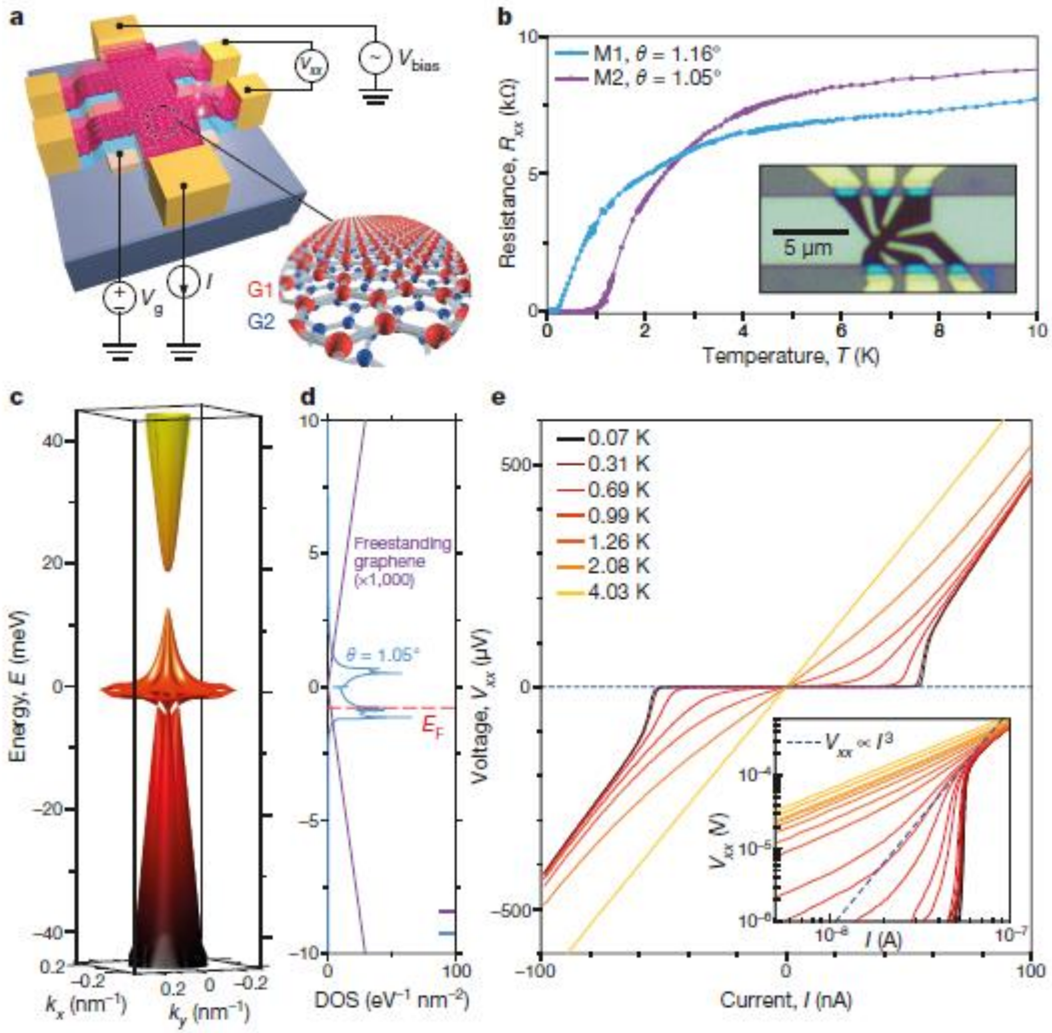
Scientific Case : Magic Angle Twisted Bilayer Graphene (MATBG)



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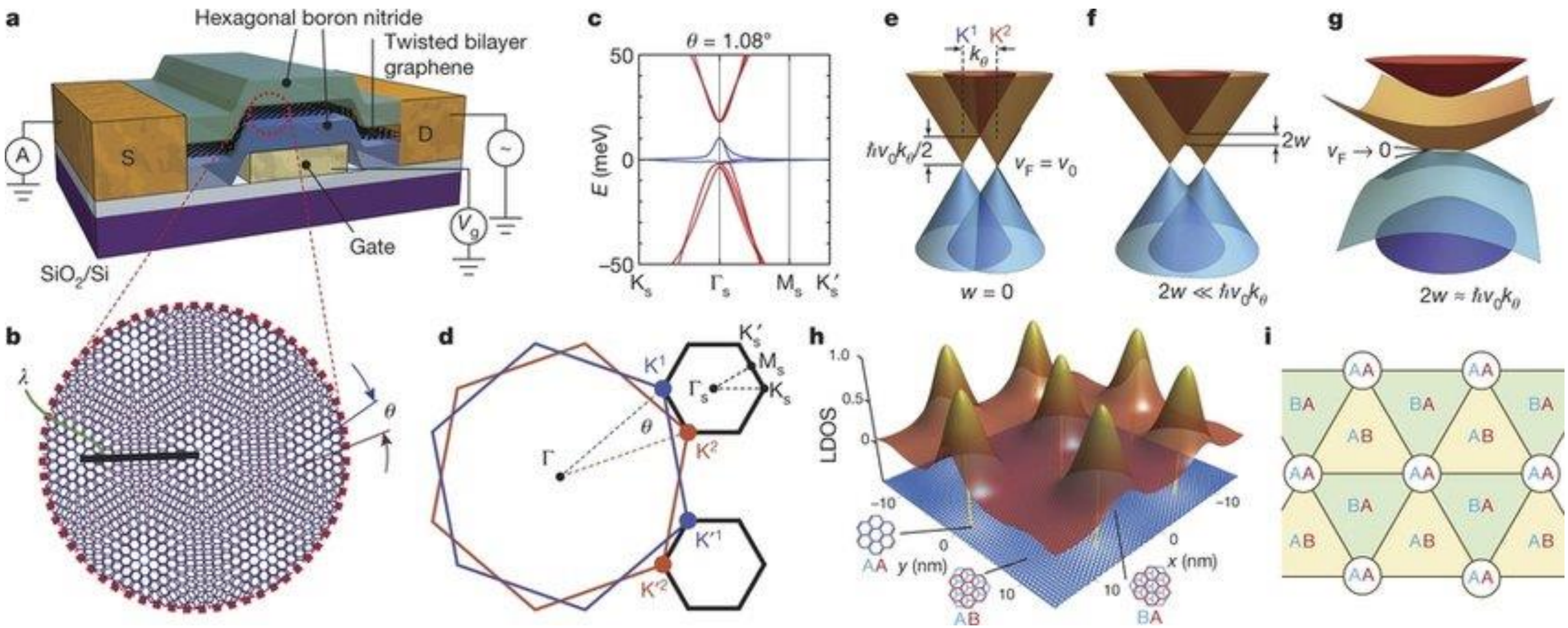
Cao et al., Nature (2018)

Scientific Case : Magic Angle Twisted Bilayer Graphene (MATBG)

The simulated band structure of twisted bilayer graphene



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Cao et al., Nature (2018)

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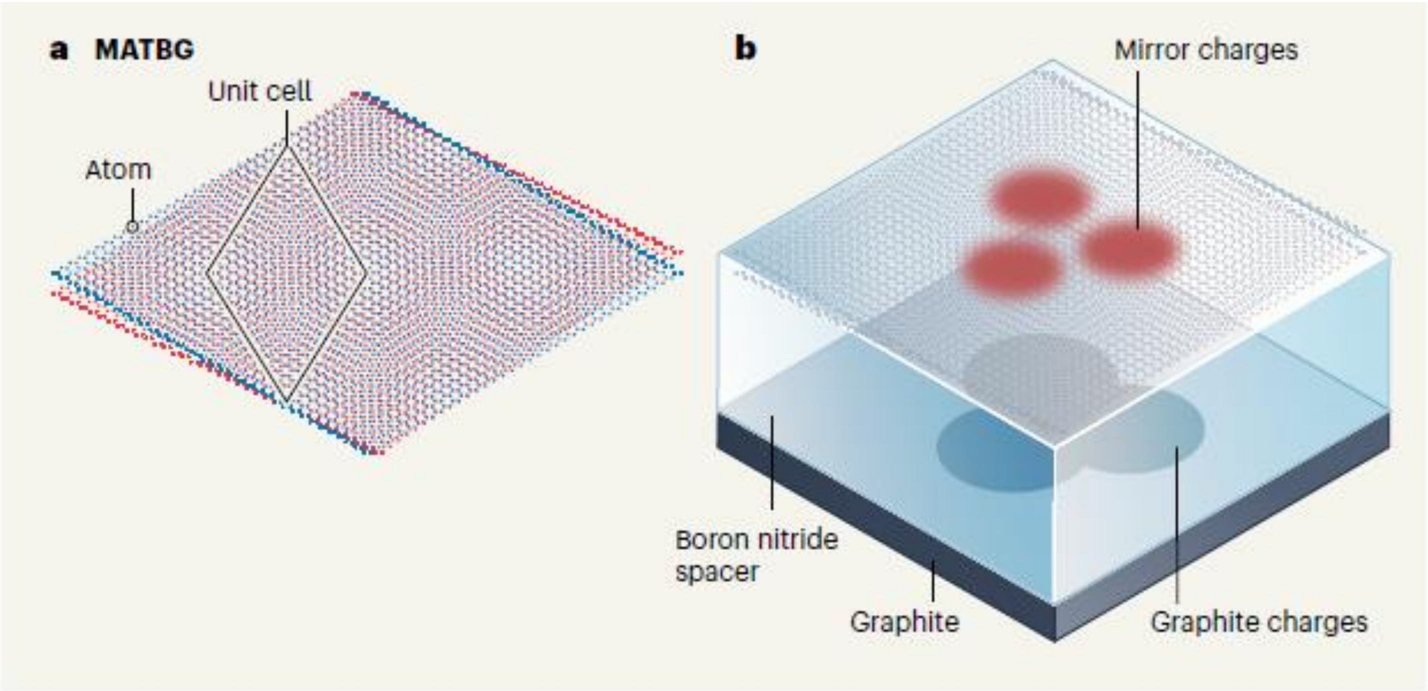

Figure 1 | Dielectric engineering in magic-angle twisted bilayer graphene (MATBG). a, MATBG comprises two layers of graphene (2D sheets of carbon atoms) that are stacked with their honeycomb lattices slightly rotated out of alignment. The atoms form a moiré pattern in which the spatial extent of the unit cell (the smallest repeating unit) is as large as 15 nanometres. **b**, Stepanov *et al.*[4] report an experiment in which boron nitride acts as a dielectric (insulating) spacer between MATBG and a graphite layer. Mirror charges (charges of opposite sign to those in the graphite) are induced in the MATBG. When the thickness of the spacer is less than the spatial extent of the unit cell, the interaction strength of electrons in the MATBG is substantially altered. Arora *et al.*[5] report an experiment in which a tungsten diselenide layer is included in a related set-up (not shown).

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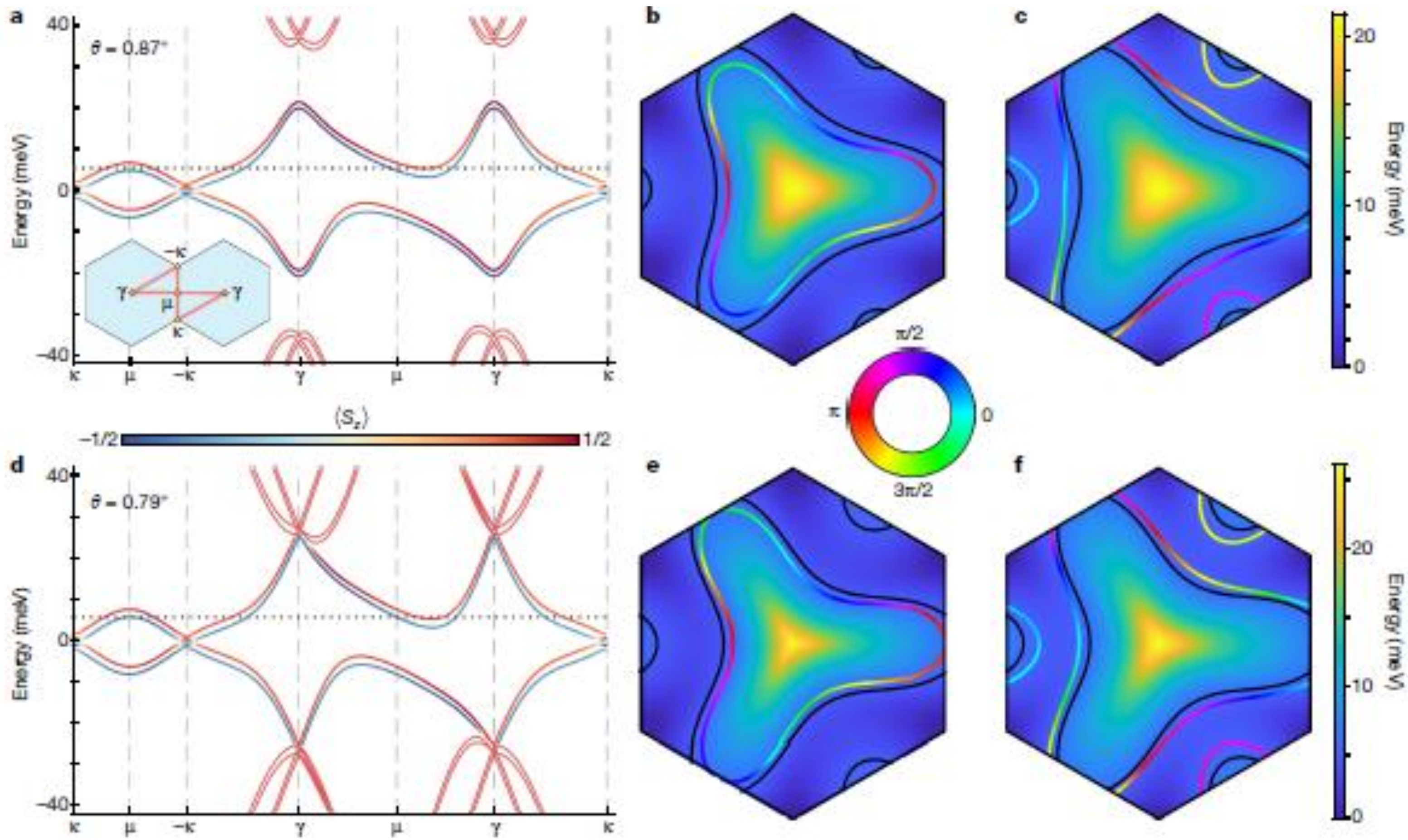
Thomale., Nature (2020)

Scientific Case : Superconductivity in MATBG stabilized by WSe2



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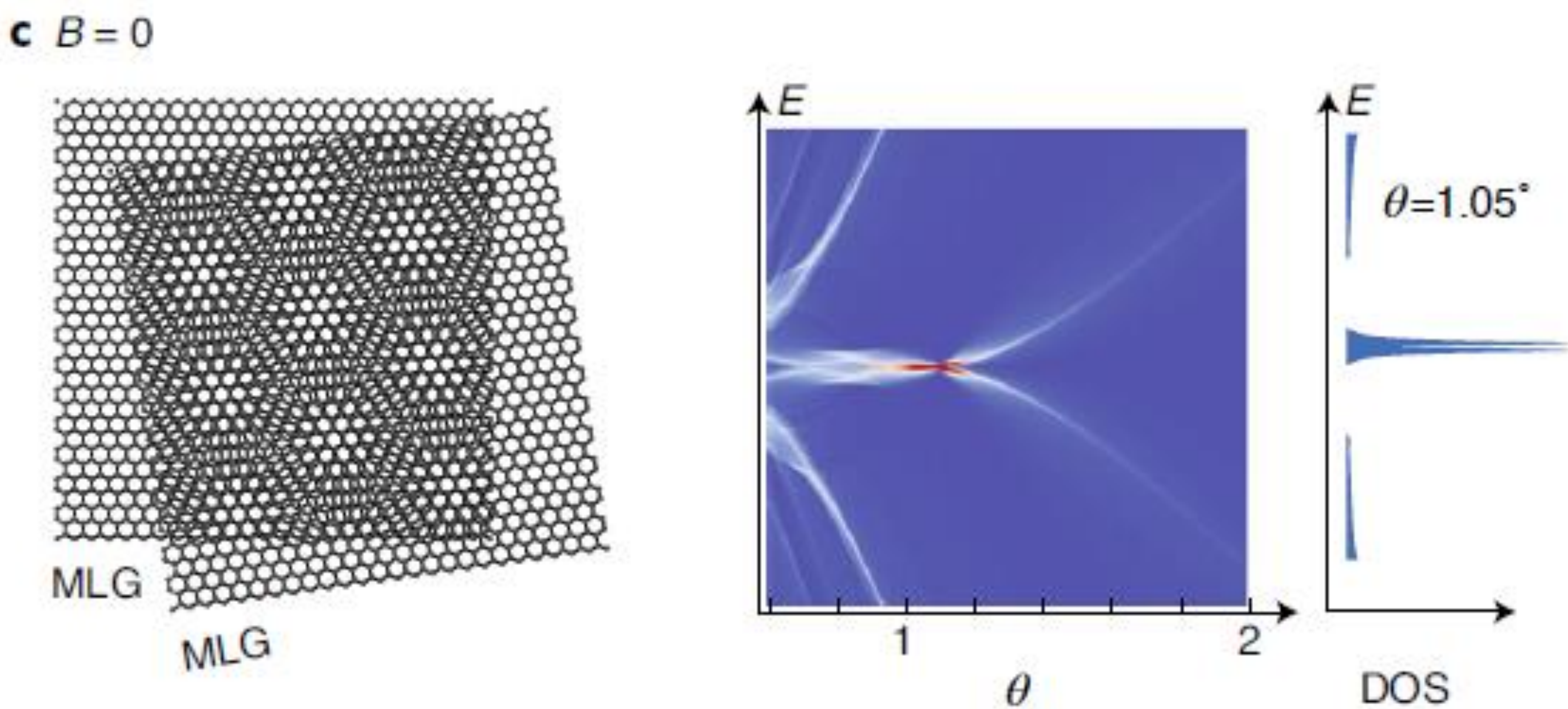
Arora., Nature (2020)

Scientific Case : misaligned Single Layer Graphene on ML h-BN



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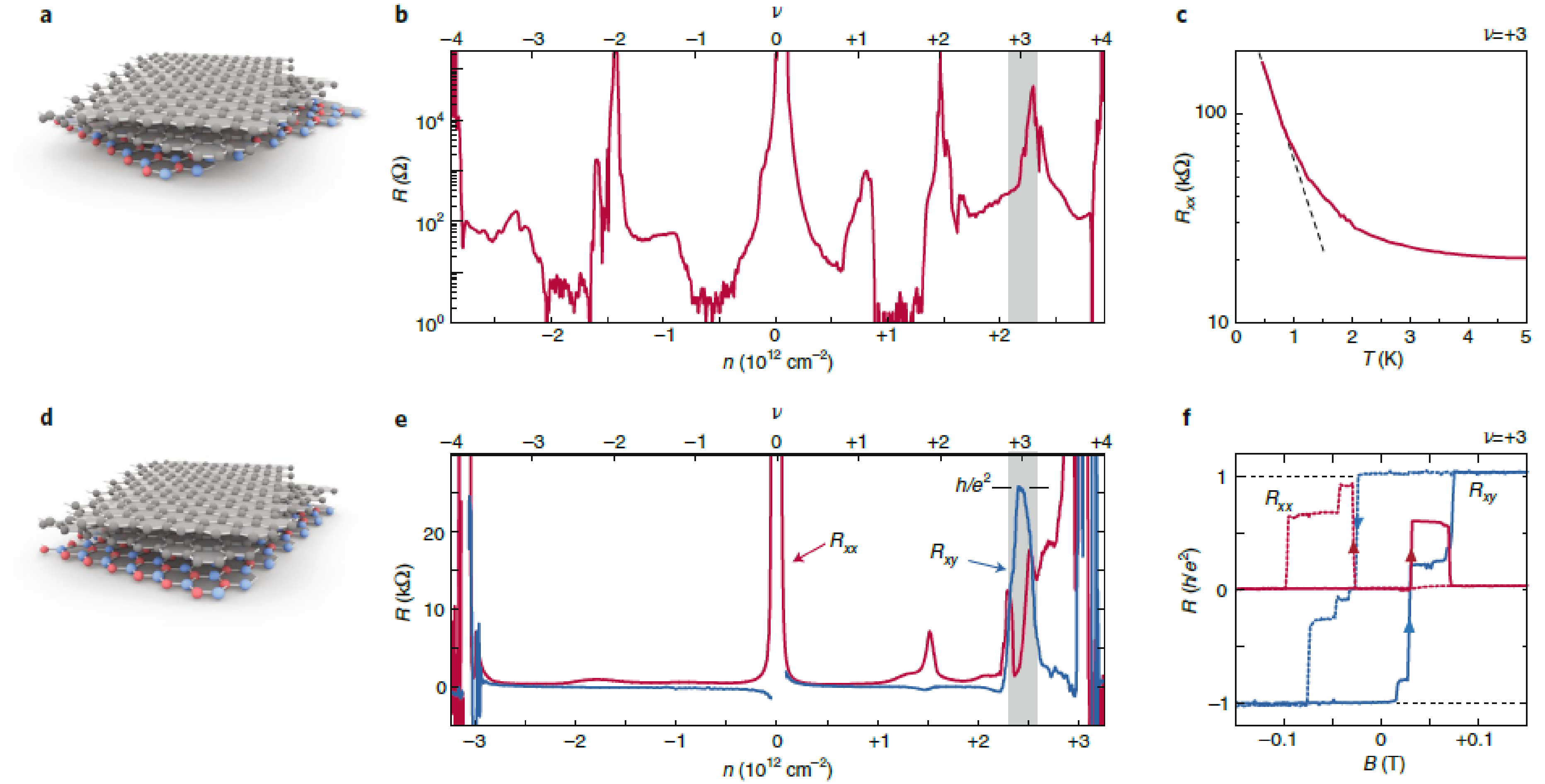
Balents., Nature Physics (2020)

Scientific Case : misaligned Single Layer Graphene on ML h-BN



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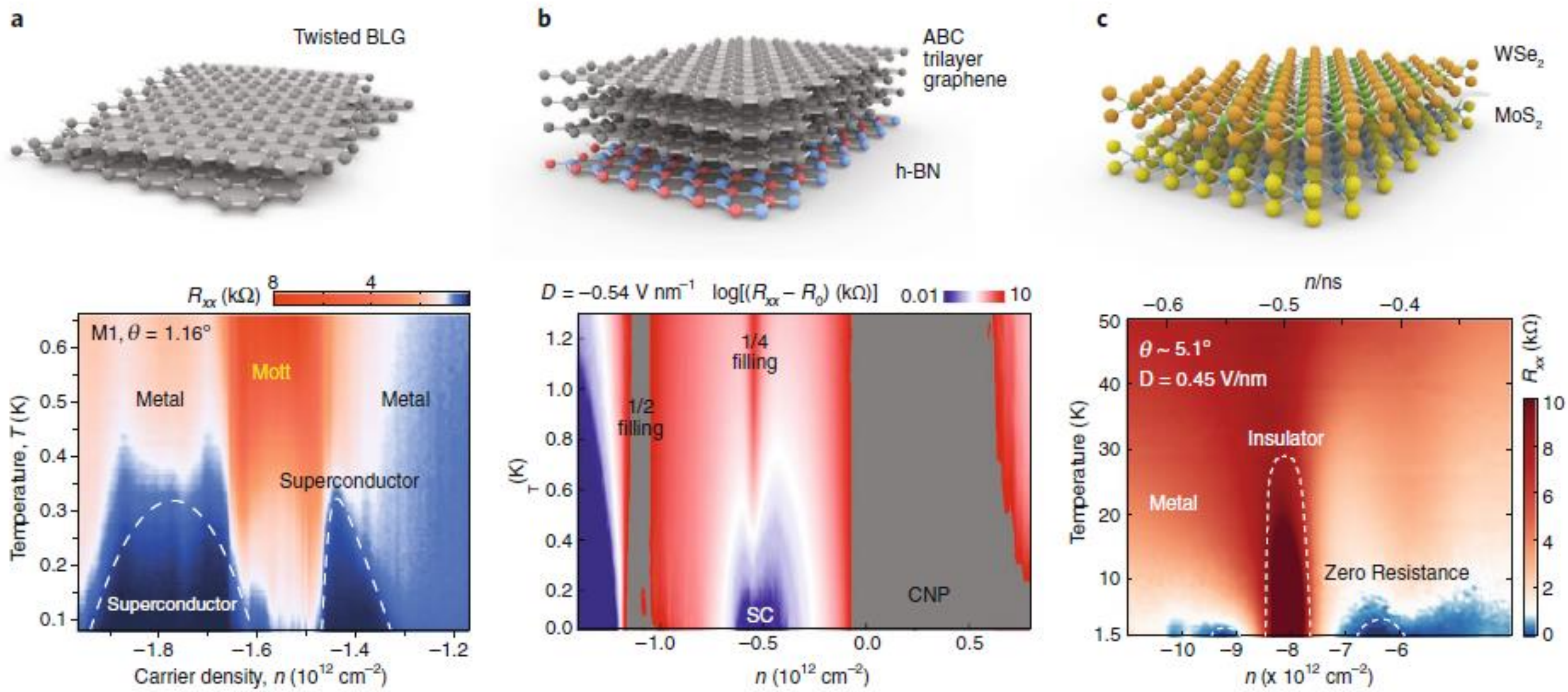
Balents., Nature Physics (2020)

Scientific Case : Proliferation of zero-field narrow-band systems



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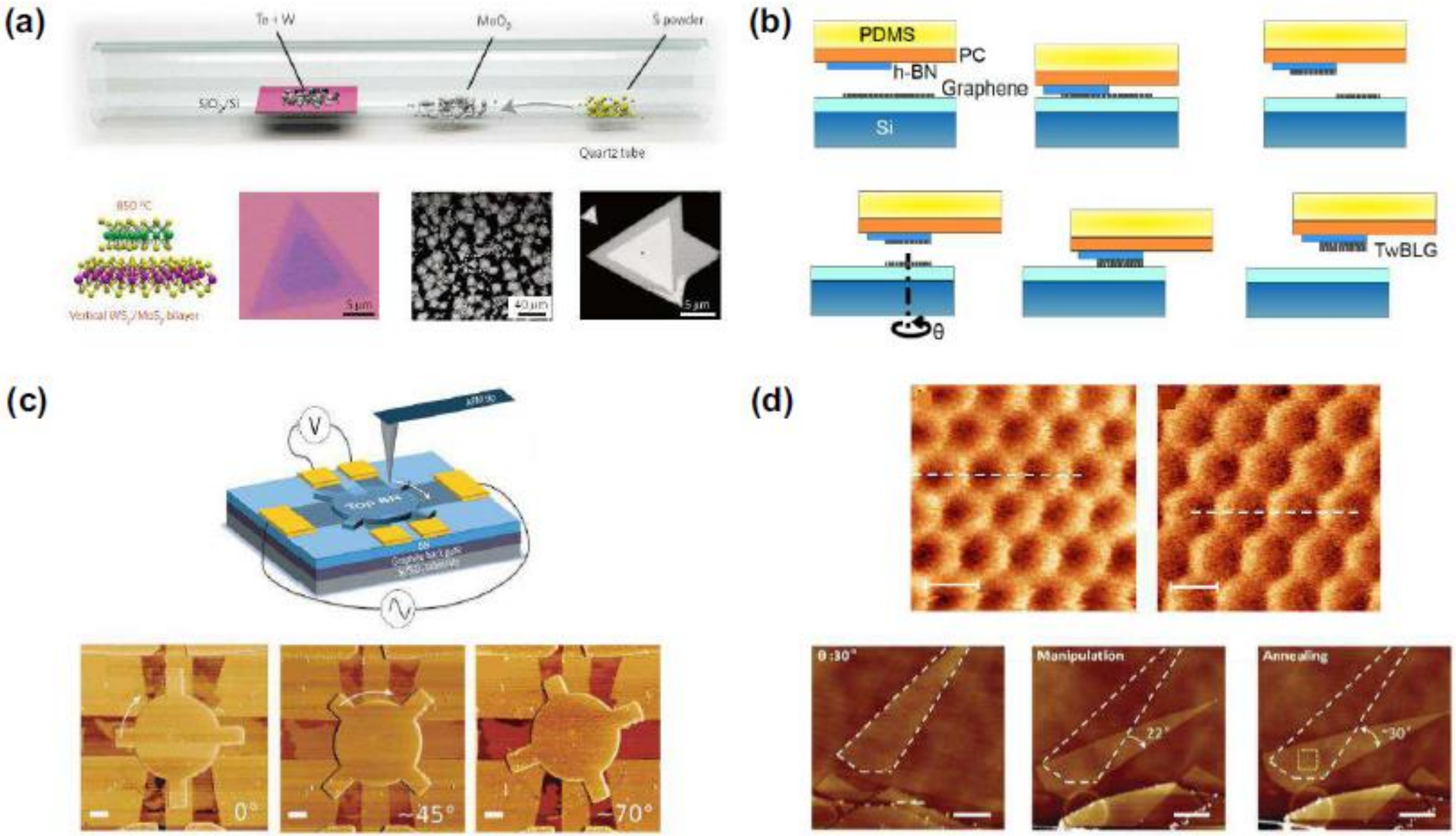
Balents., Nature Physics (2020)

Scientific Case : Fabrication of van der Waals heterostructure



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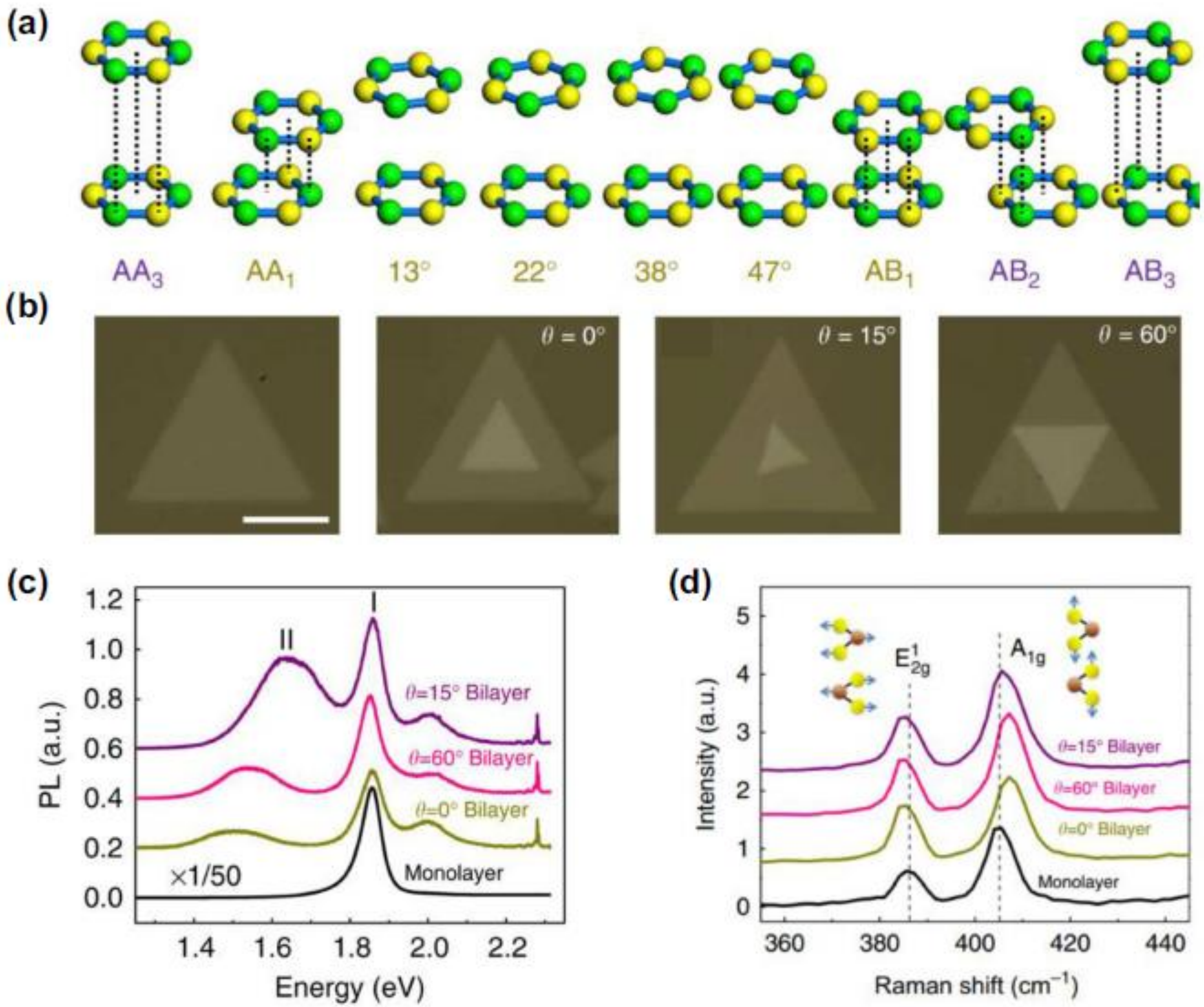
Scientific Case : Fabrication of van der Waals heterostructure

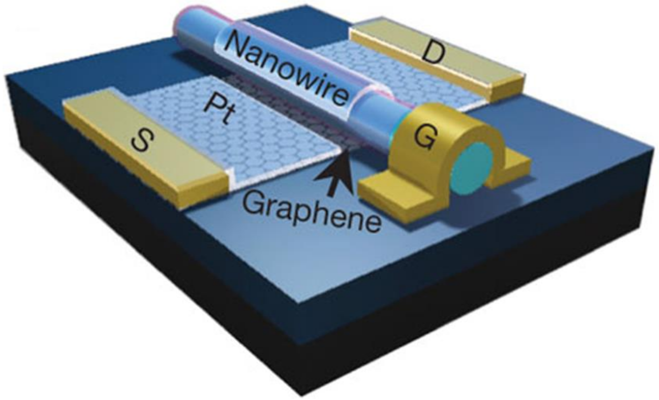


Liu et al., Nat. Comm (2014)

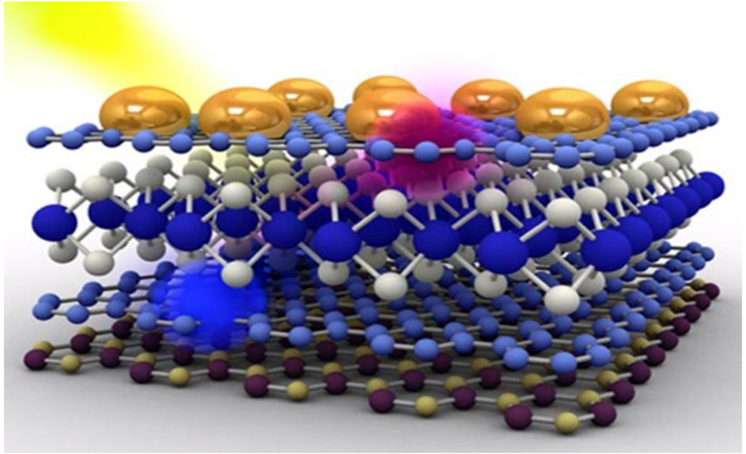
With spatial and momentum k information

Conduct ARPES experiment with smaller beam size

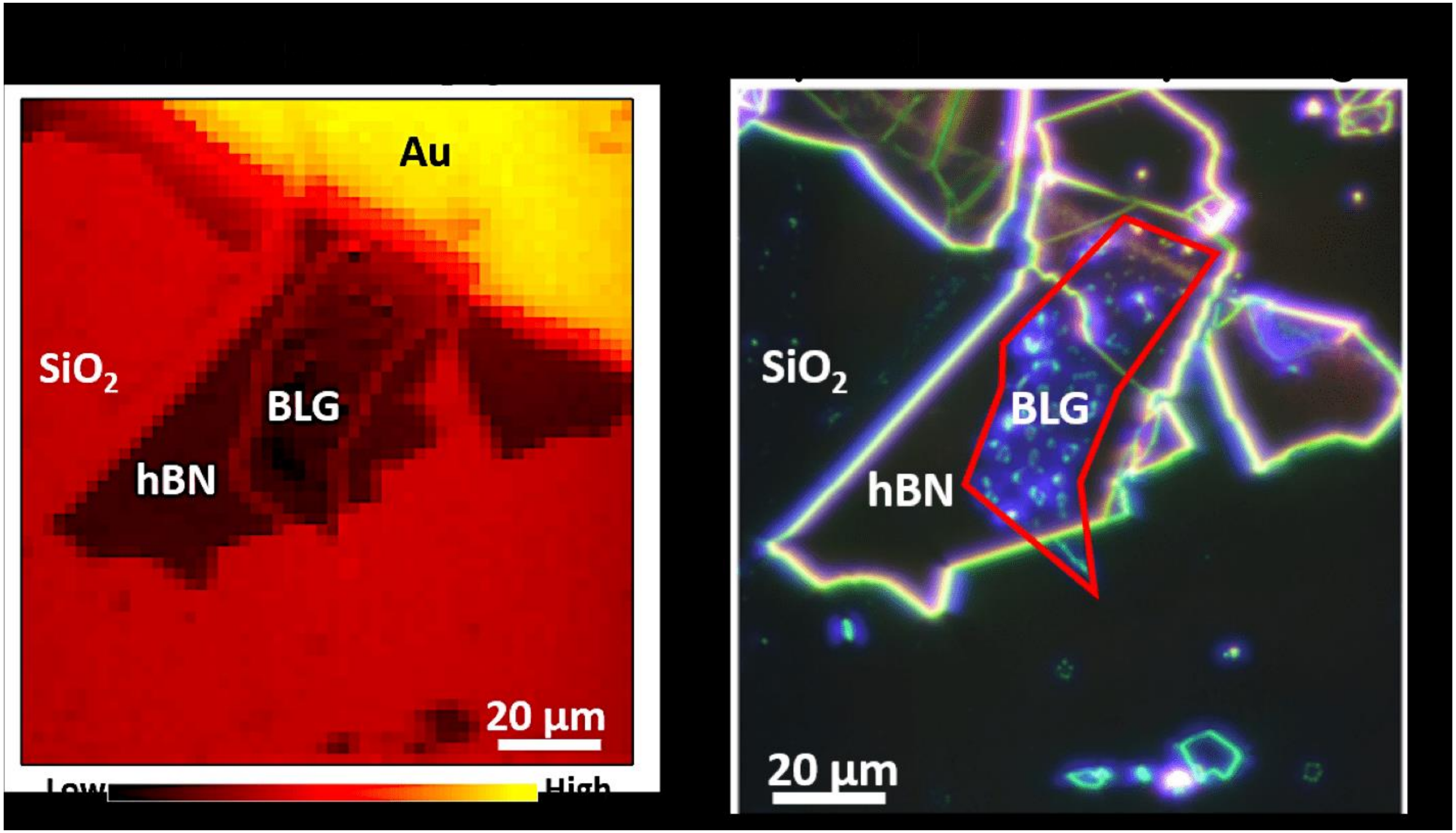
Electronic and Chemical imaging of Nanowires and patterned Samples



Band structure of transition metal dichalcogenides and their correlation effects



Magic angle in twisted Graphene



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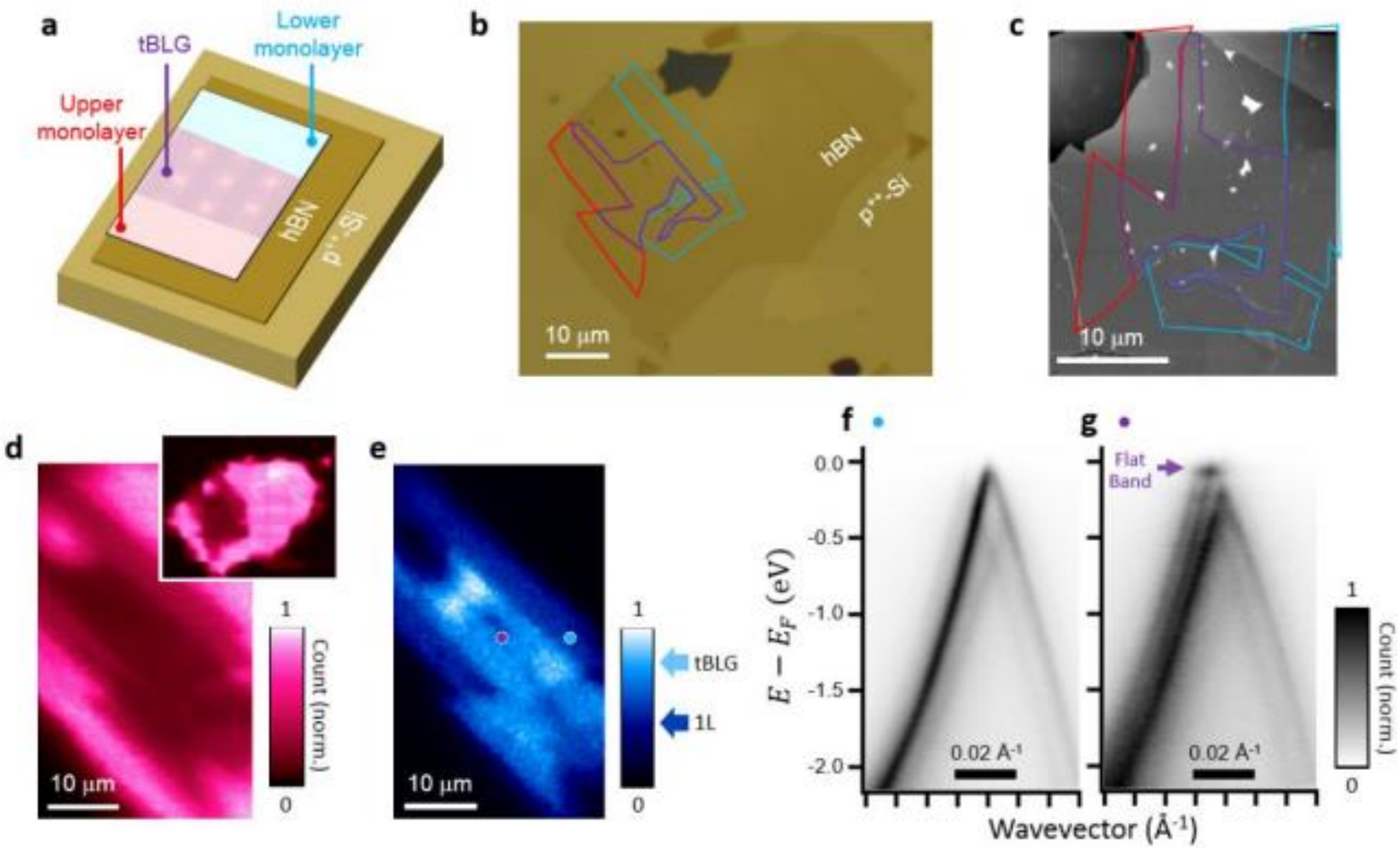
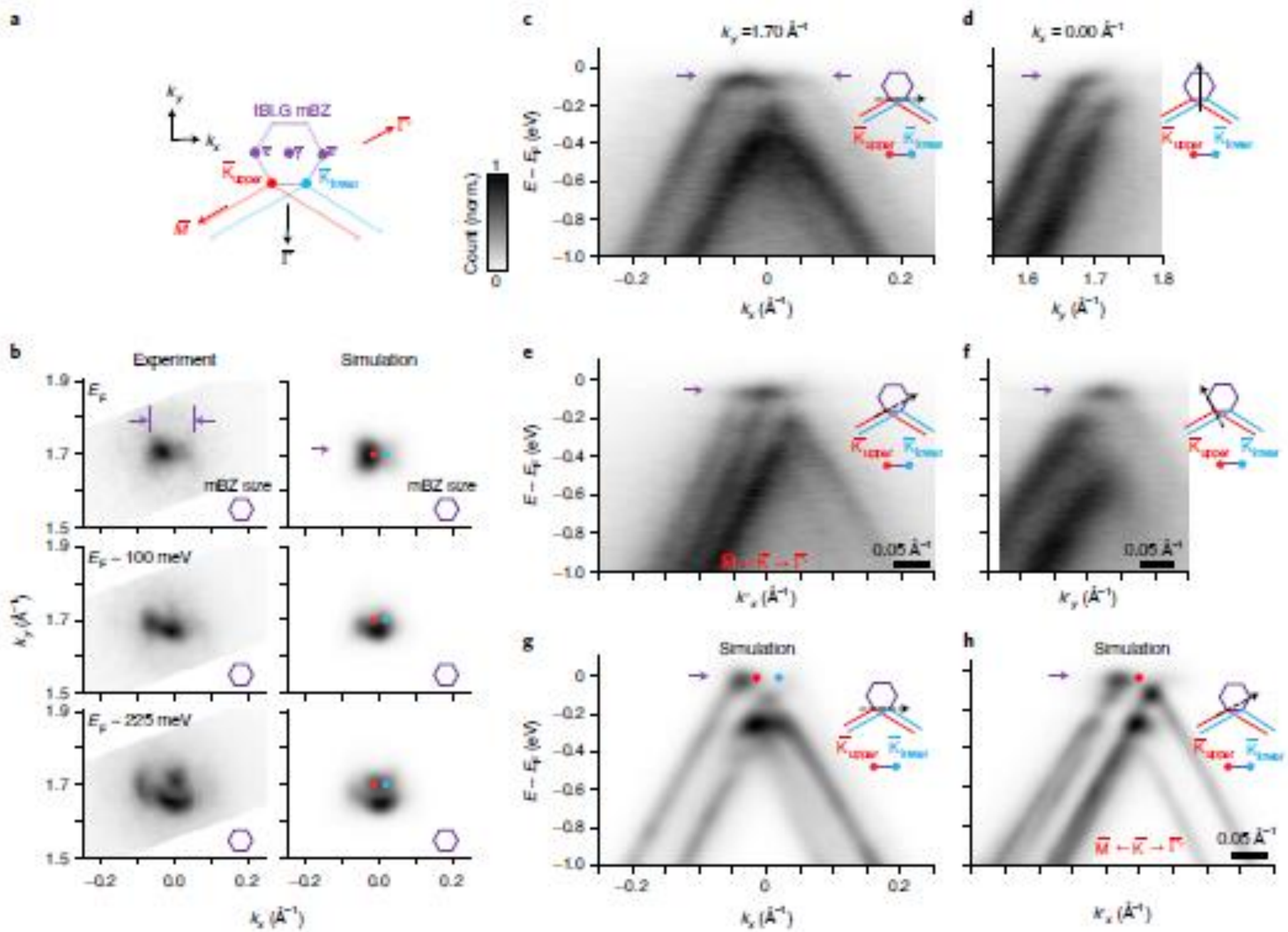


Ref : Utama et al., Nat. Phys. (2020)

Magic angle in twisted Graphene



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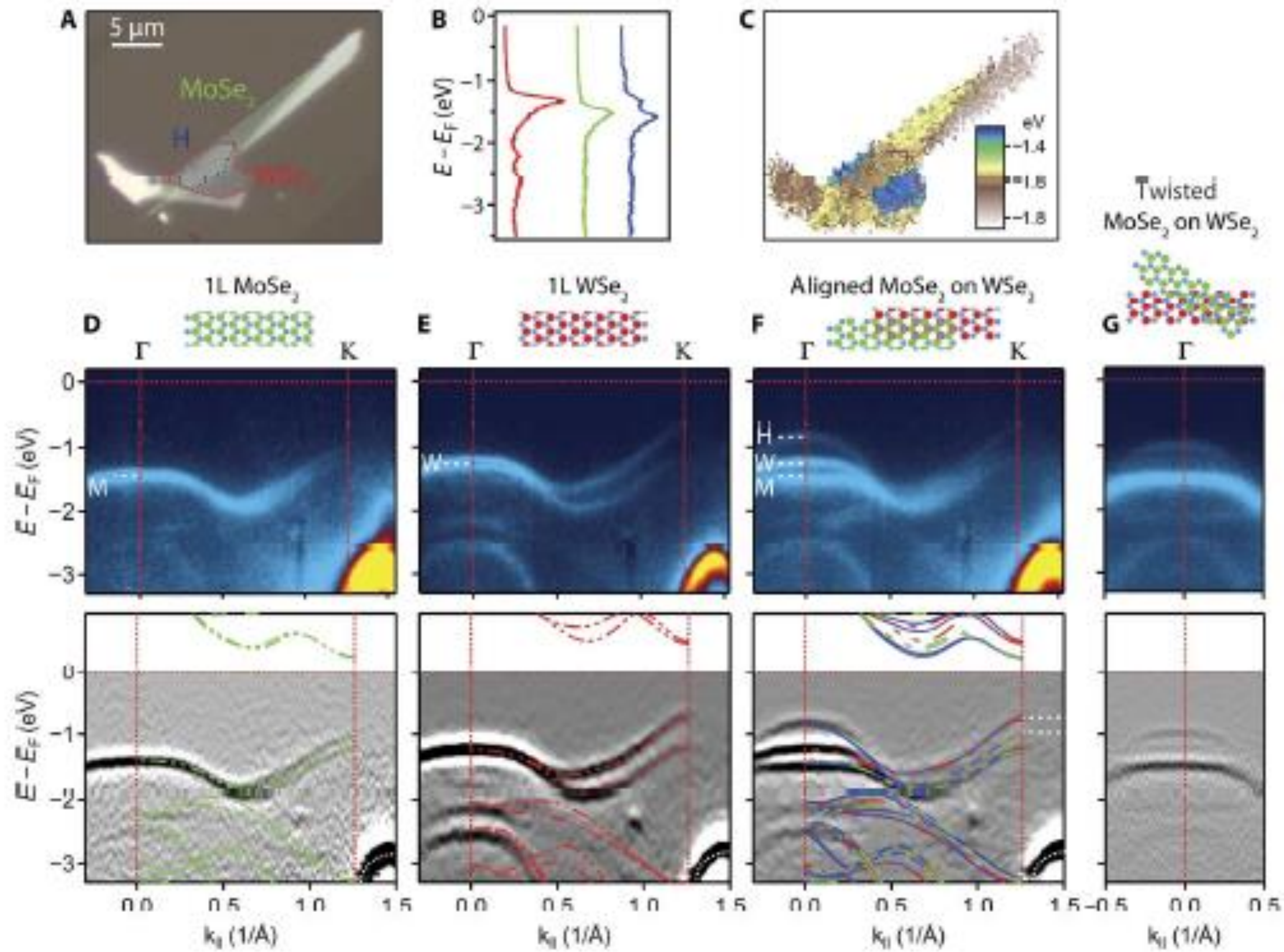
Van der Waals Heterostructures



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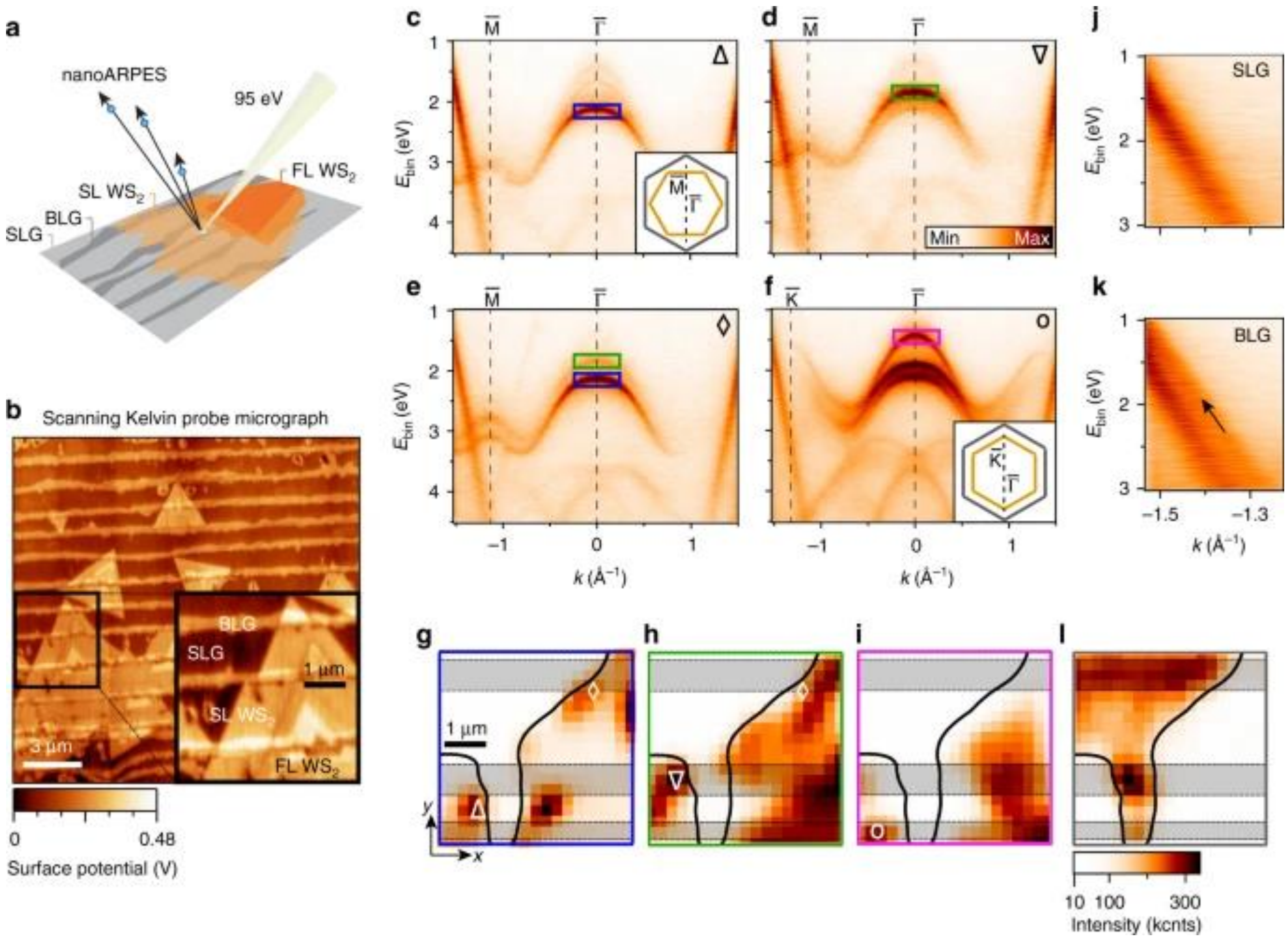
Ref : Wilson et al., Sci. Adv. (2020)

Van der Waals Heterostructures



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Ref : Ulstrup et al., Nat Comm. (2017)

Future and outlook

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  - *Topological insulator, 2D materials, superconductors, complex oxides, graphene-based materials….*
  - *Time-resolved ARPES.*
  - *Spin-resolved ARPES*

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  - *Novel materials fabricated with the CVD method, micro-mechanic exfoliation… usually have smaller domain size in the early stage of material discovery. (van der Waals heterostructure, twisted bilayer graphene, 2D materials)*
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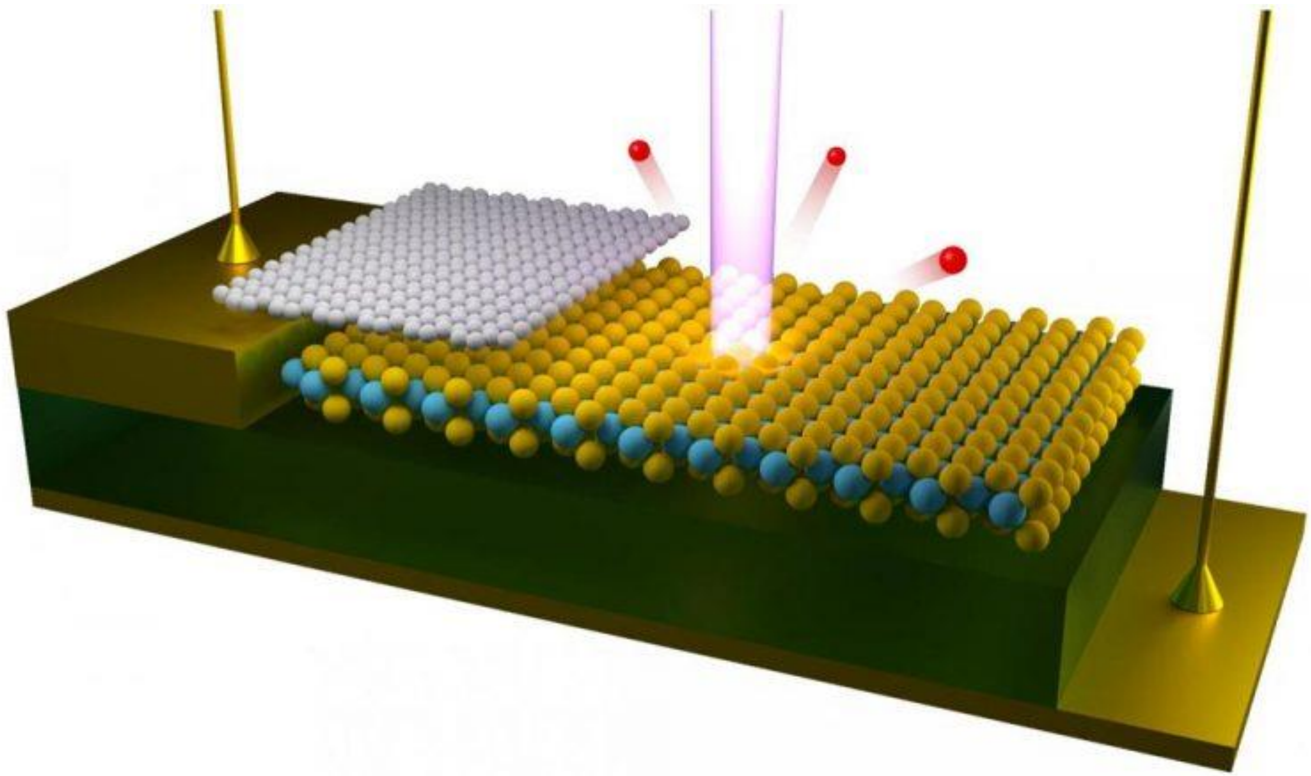
  - *Gating Effects in 2D Materials*

Gating Effects in 2D Materials

First-ever visualizations of electrical gating effects on electronic structure



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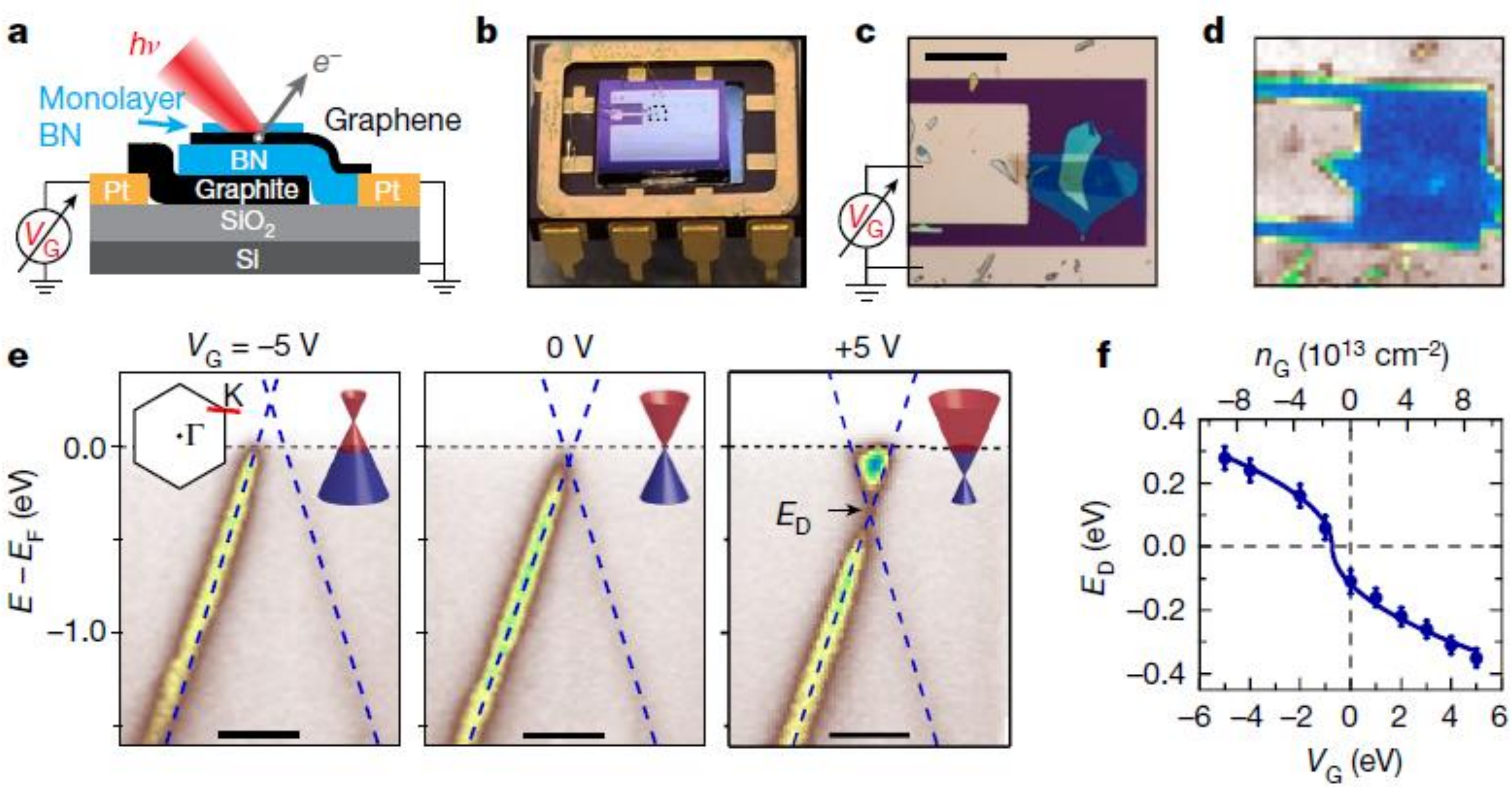
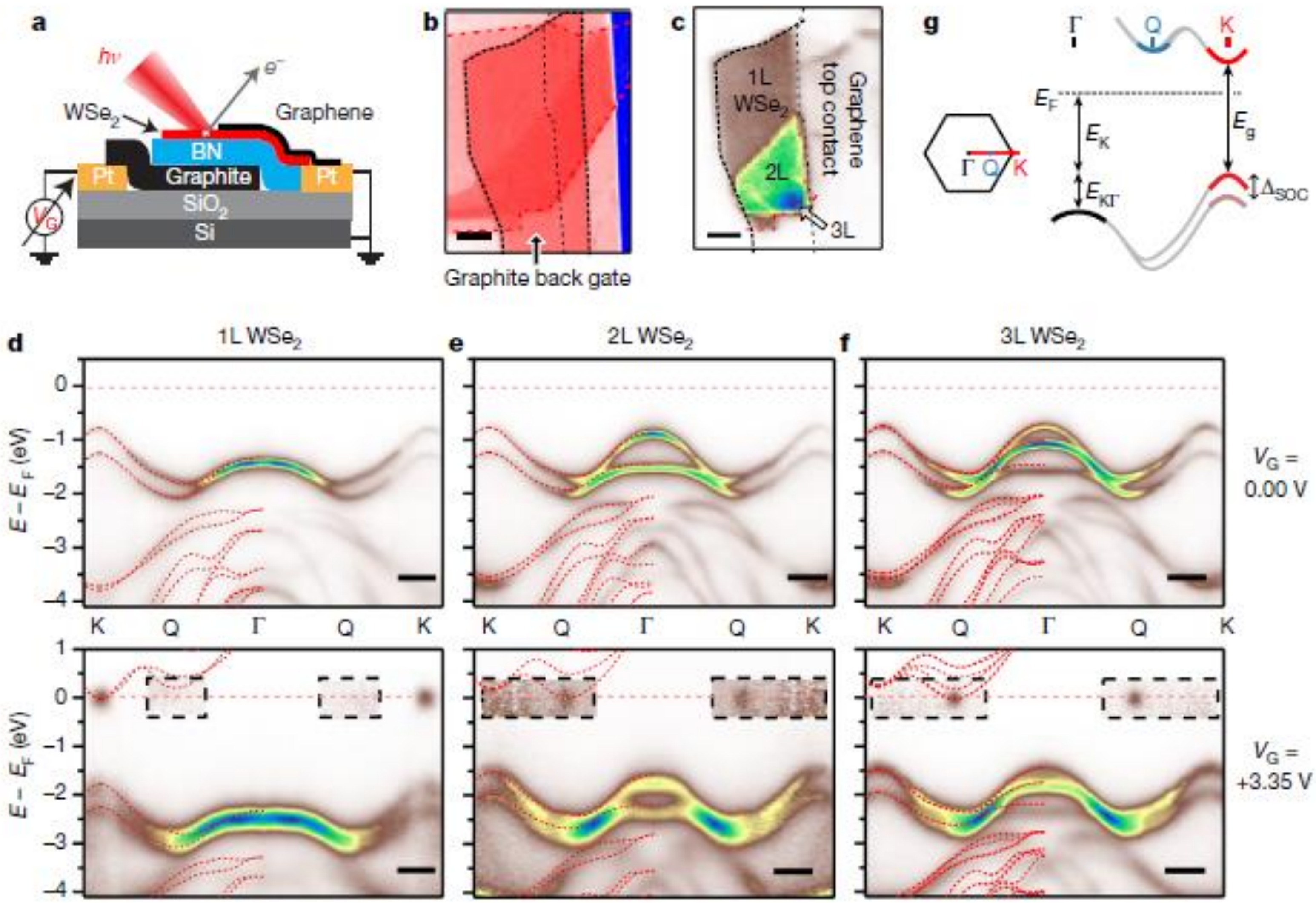
Gating Effects in 2D Materials



Paul V. Nguyen et al., Nature (2019)

Gating Effects in 2D Materials



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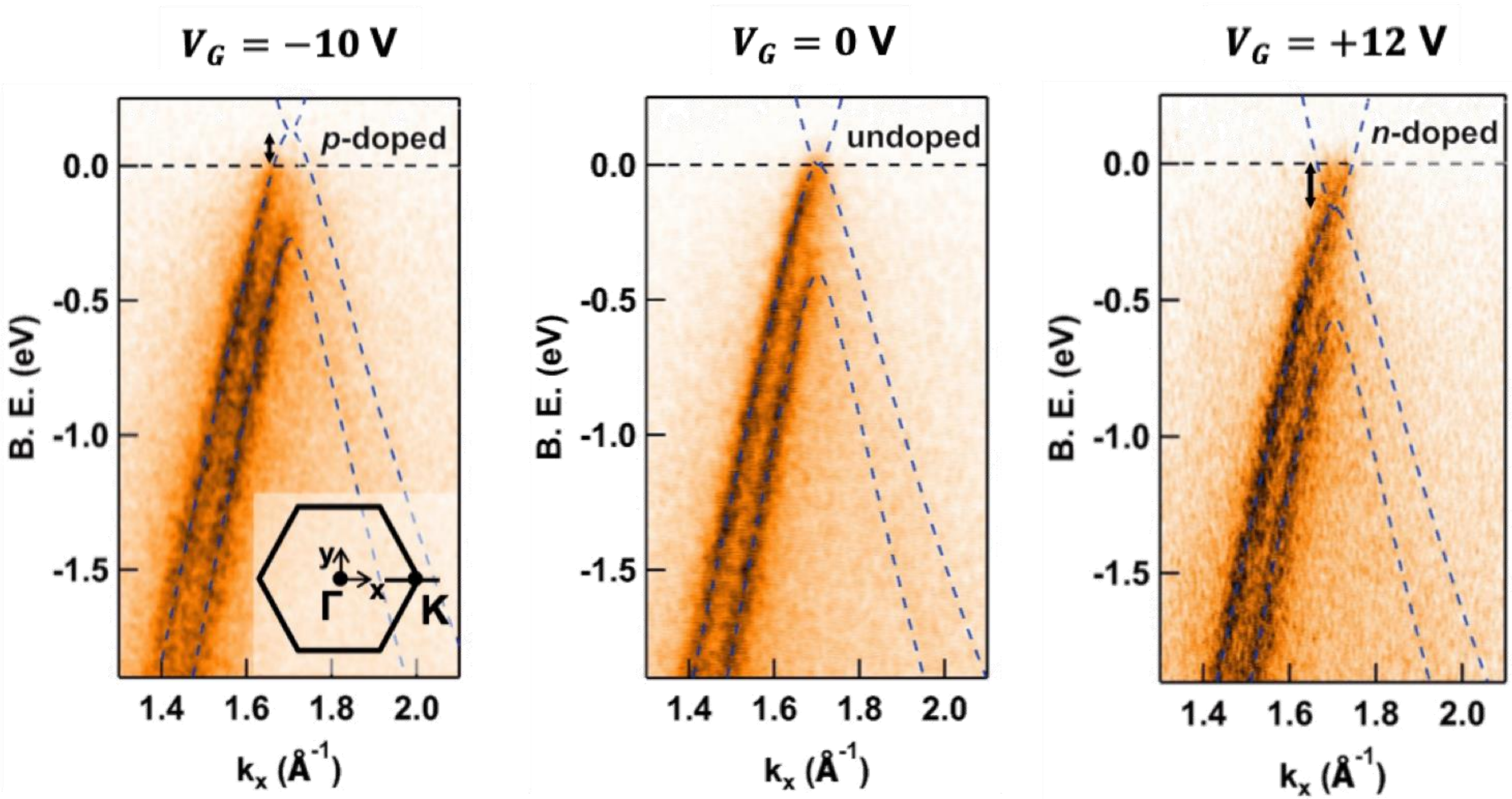
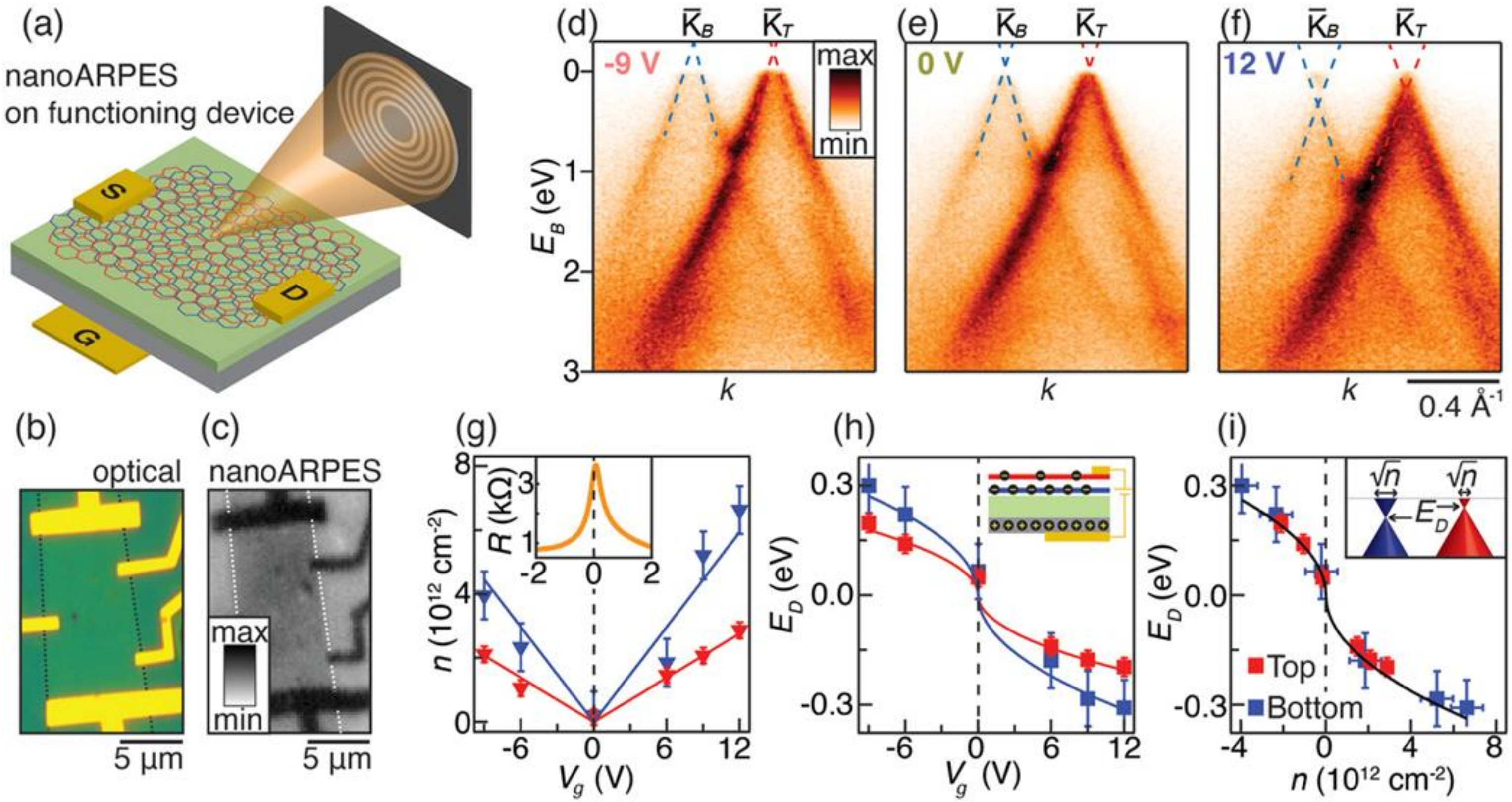


Joucken et al., Nano Letters (2019)

Gating Effects in 2D Materials



Jones et al., Adv. Mater. (2020)

Beamlines in light sources worldwide for micro- or nano-ARPES

valence band
nanoARPES

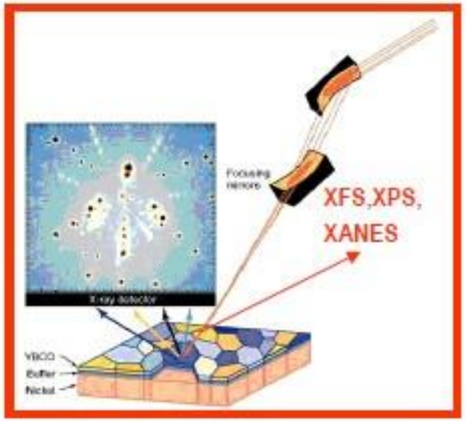
operating:
Elettra, Soleil, DLS

planning/commissioning:
TPS, NSLS-2

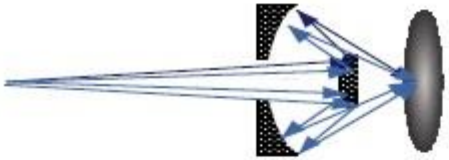
nanoARPES : X-ray focusing optics

X-ray focusing optics:
zone plates, mirrors, capillaries

Zone Plate optics – circular grating with decreasing width: from ~ 200 to ~ 10000 eV
Monochromatic:
Resolution achieved 15 nm in transmission

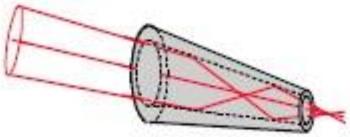


KP-B mirrors each focusing in one direction: soft & hard X-rays: ~ 100 nm
Soft & hard x-rays!
achromatic focal point, easy energy tunability, comfortable working distance
Resolution ≲ 100 nm



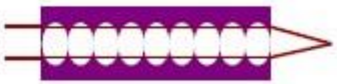
Normal incidence:
spherical mirrors with multilayer interference coating (Schwarzschild Objective)
Monochromatic, good for E < 100eV
Resolution: best ~ 100 nm

Capillary: multiple reflection concentrator



Hard x-rays ~ 8-18 keV
Resolution: > 3000 nm

Refractive lenses



Hard x-rays ~ 4-70 keV
Resolution: > 1000 nm

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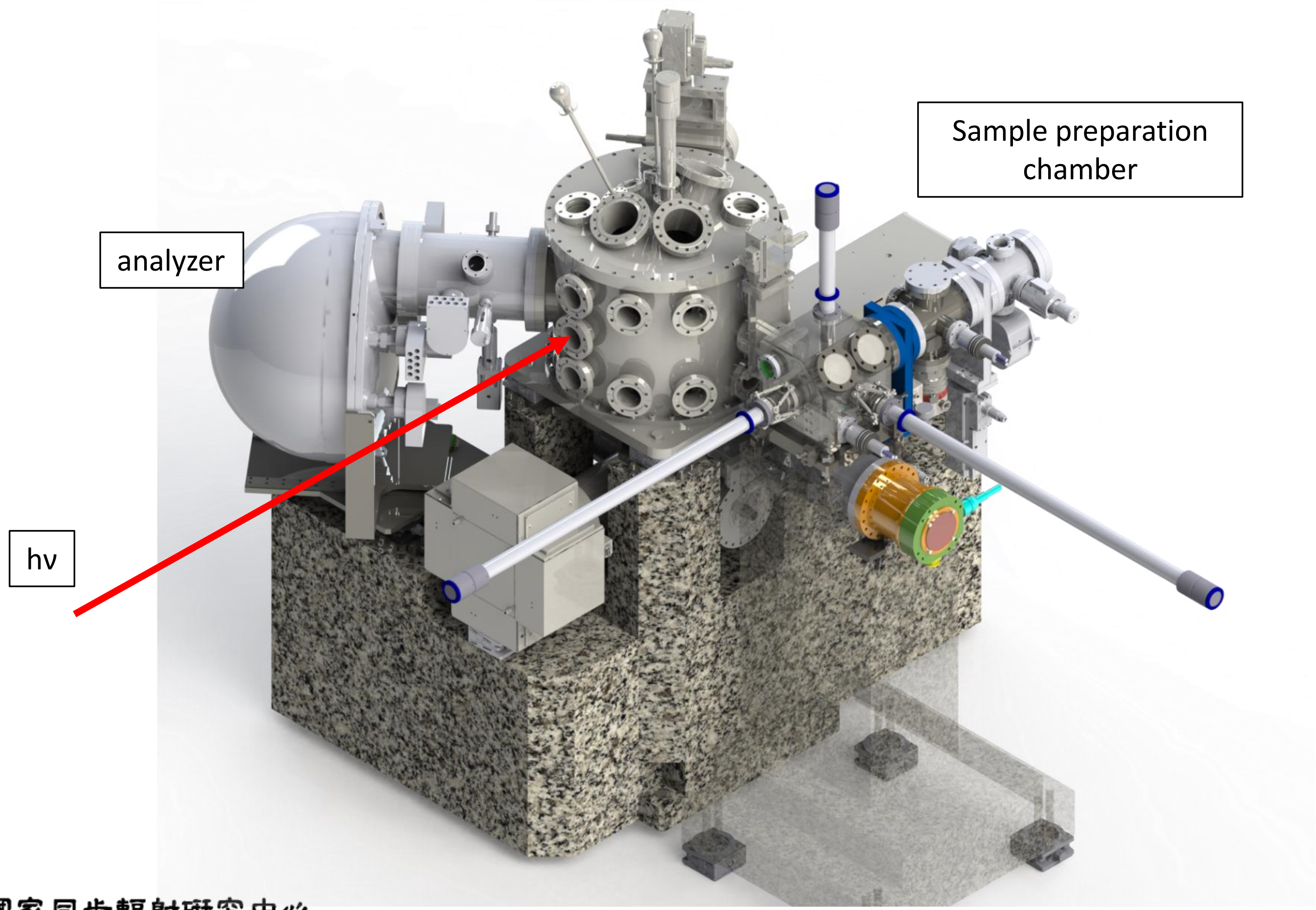
The end station design of NanoARPES Branch



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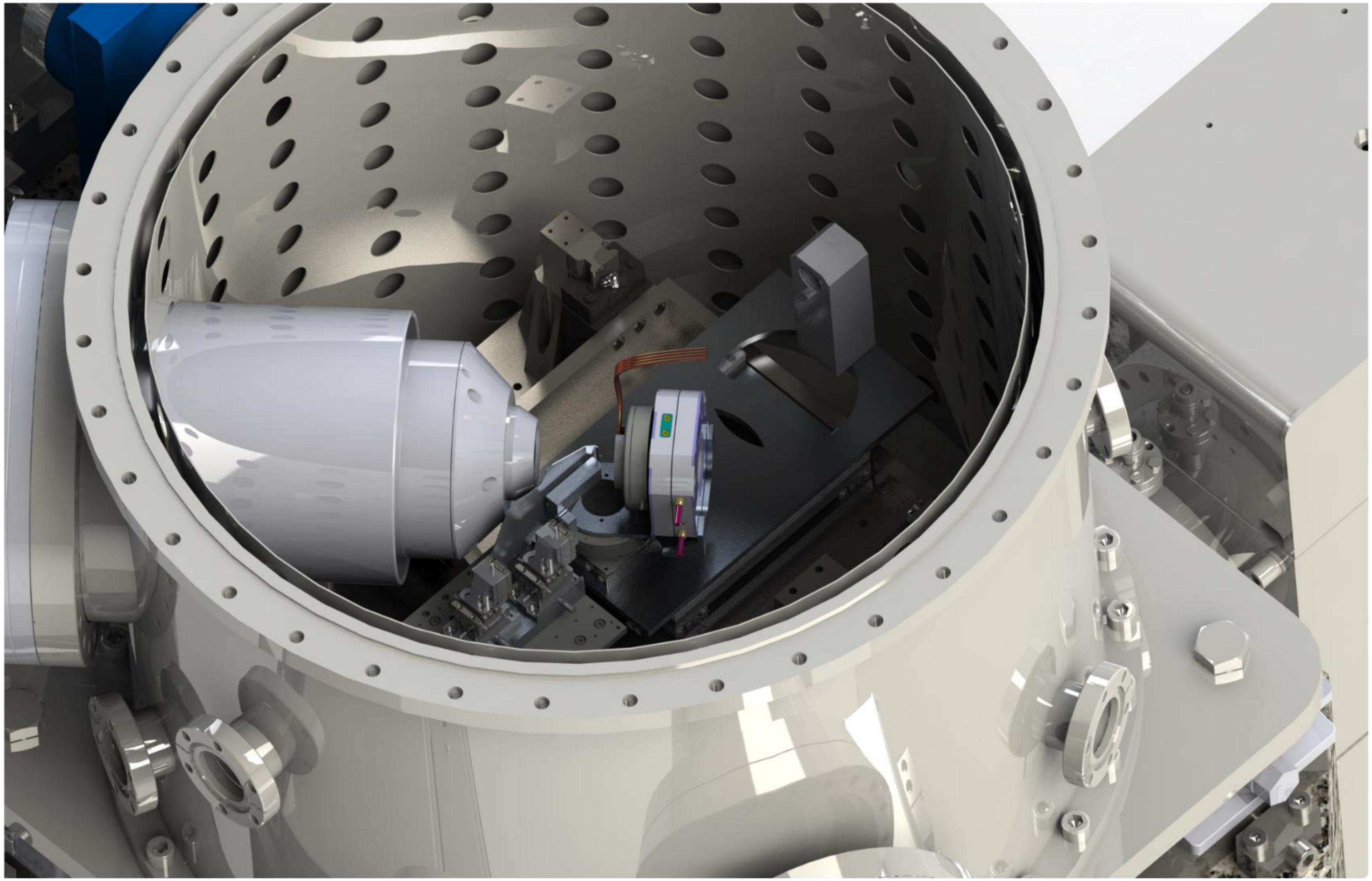

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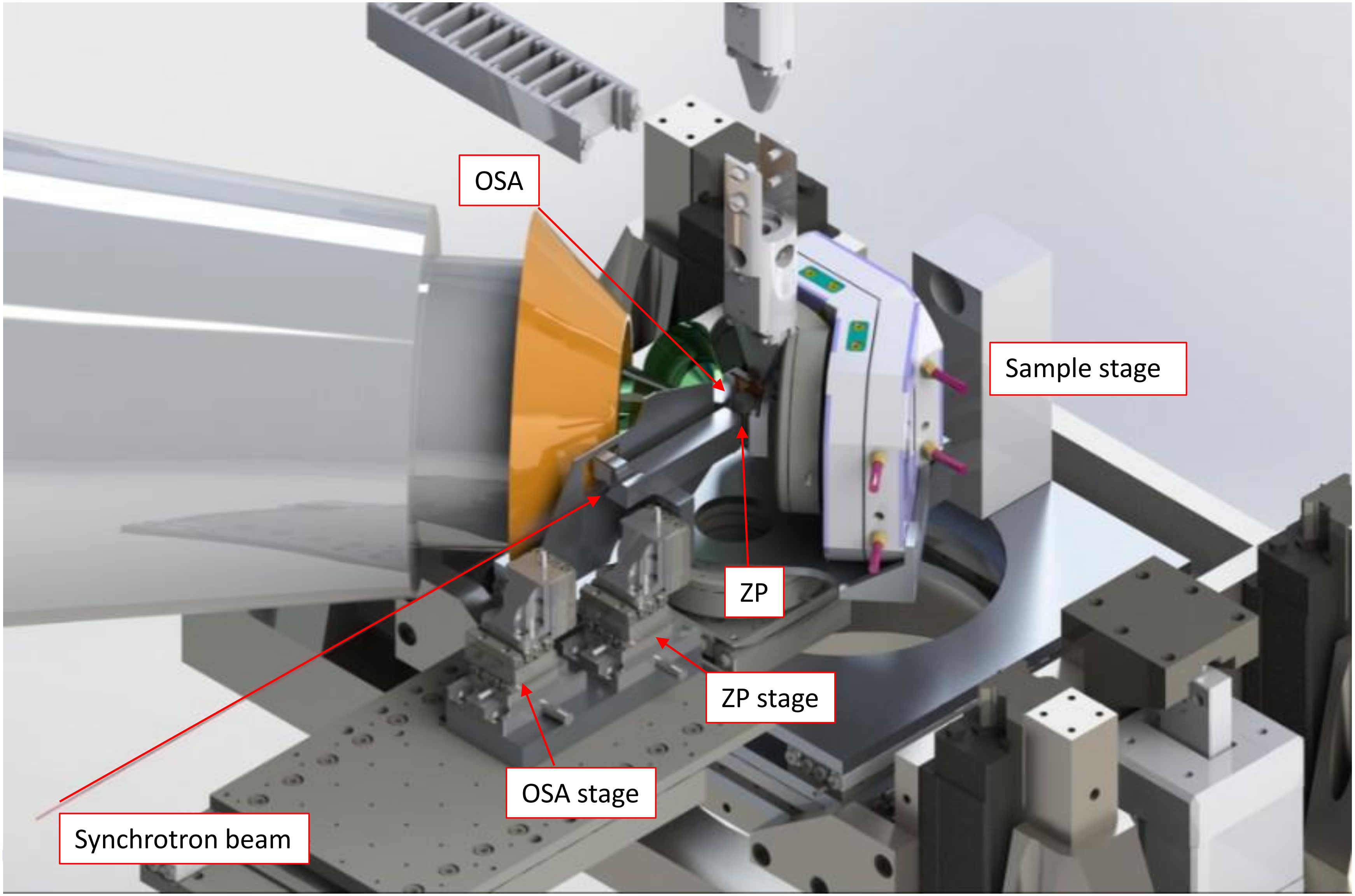

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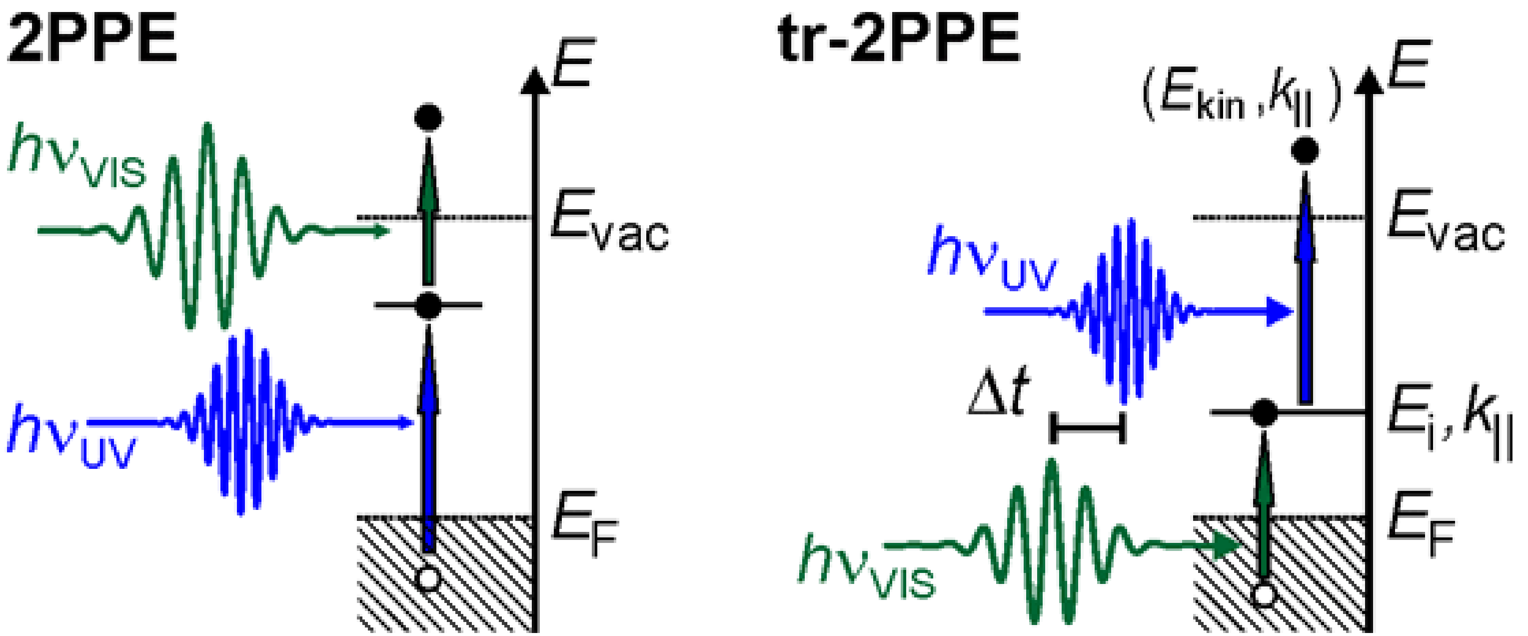
Time-resolved ARPES

Table 1. Some typical laser light sources used in ARPES.

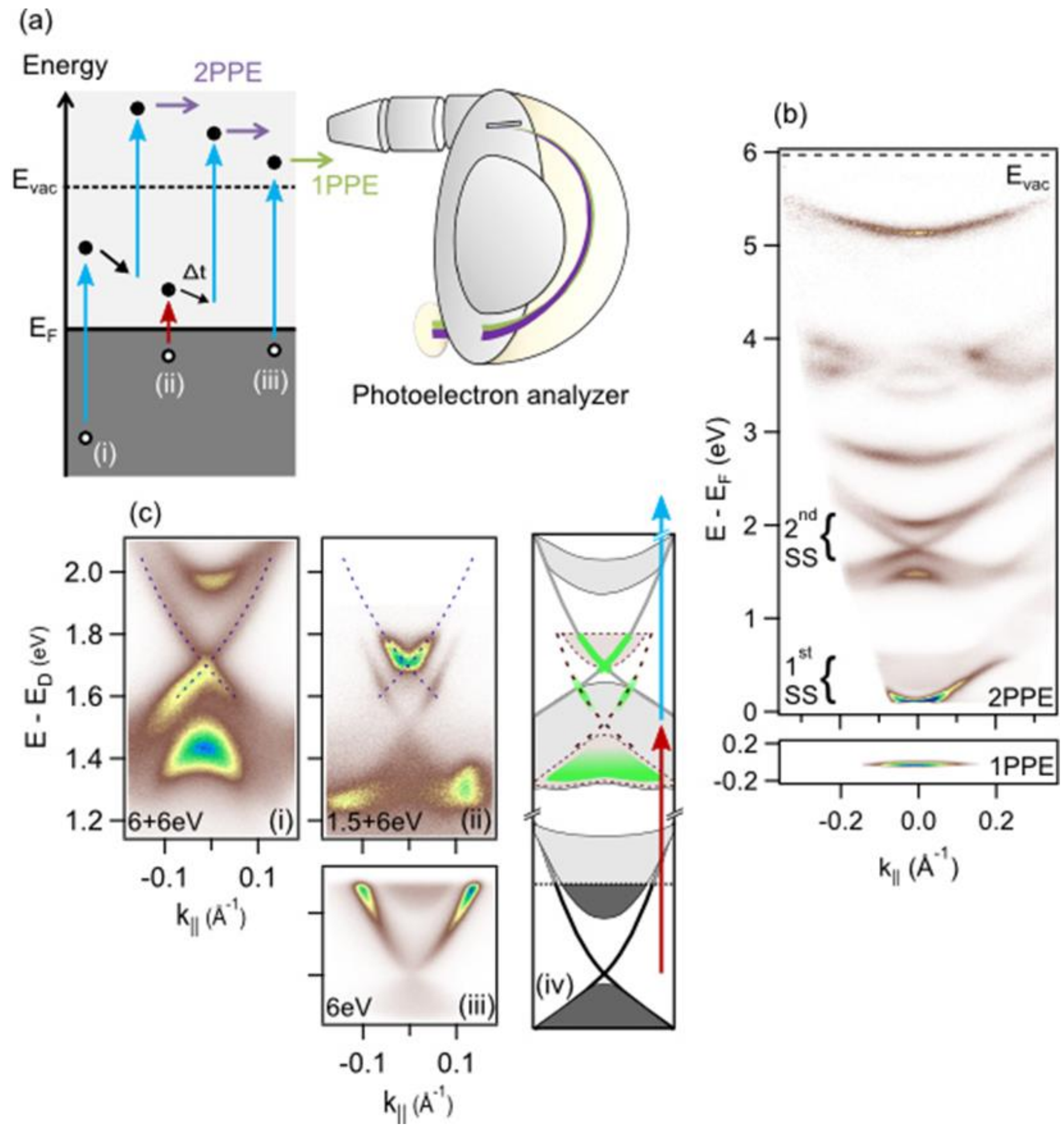
Laser catagory	Generation	Application	Pho. energy (eV)	Pulse width (ps, fs)	Rep. rate (kHz, MHz)	Max.Pho. flux (photons s^{-1})	Energy res. (meV)	Tem. Res. (fs)	References	Remarks
Quasi-CW	NLO crystal SFG+SHG	High-res ARPES	7	~10 ps	80 MHz	1.5×10^{15}	0.26 meV	/	Liu *et al* [21]	(a)
			6	~70 fs (seed)	100 MHz	$\sim 10^{15}$	4.7 meV	/	Koralek *et al* [20]	(b)
			7	~10 ps	120 MHz	Unknown	0.025 meV	/	Okazaki *et al* [186]	(c)
			5.3–7	5 ps	76 MHz	$\sim 10^{14}$	Unsecified	/	Jiang *et al* [87]	(d)
CW	NLO crystal SFG+SHG	High-res ARPES	6.05	Infinite	Infinite	1×10^{15}	0.01	/	Tamai *et al* [37]	(e)
			6.49	Infinite	Infinite	1.25×10^{15}	$\sim 10^{-7}$ meV	/	Scholz *et al* [54]	(f)
Pulsed laser	NLO crystal SFG+SHG	Tr-ARPES	1.5,6	50 fs, 160 fs	80 MHz	Unspecified	<22 meV	163 fs	Sobota *et al* [41]	(g)
			1.5,6.04	35 fs, 55 fs	250 kHz	$\sim 10^{13}$	40 meV	65 fs	Faure *et al* [40]	
			1.48, 5.92	170 fs,-	250 kHz	Unspecified	⩾10.5 meV	⩾240 fs	Ishida *et al* [149]	
HHG	Noble gas HHG	Tr-ARPES	1.58, 15–40	40 fs, 100 fs	10 kHz	3.6×10^{17}	90 meV@35.6 eV	125 fs	Frietsch *et al* [43]	(h)
			1.6, 22.1	30 fs, 11 fs	10 kHz	Unspecified	170 meV	13 fs	Rohde *et al* [42]	(i)
			1.57, 20.4	30 fs	1 kHz	Unspecified	Unspecified	30 fs	Petersen *et al* [31]	
Mod./ Reson. type HHG	Mixed rare gas	High-res ARPES	10.5	10 ps,	0.2–8 MHz	9×10^{12}	<1 meV	/	Berntsen *et al* [75]	(j)
			10.9	100 ps,	1–20 MHz	$\times 10^{13}$	<2 meV	/	Yu He *et al* [76]	(k)
FEL	Long undulator	Tr-ARPES	26–300	30–150 ps	<10 Hz	Very high	300 meV	700 fs	Hellmann *et al* [59]	

Time-resolved ARPES



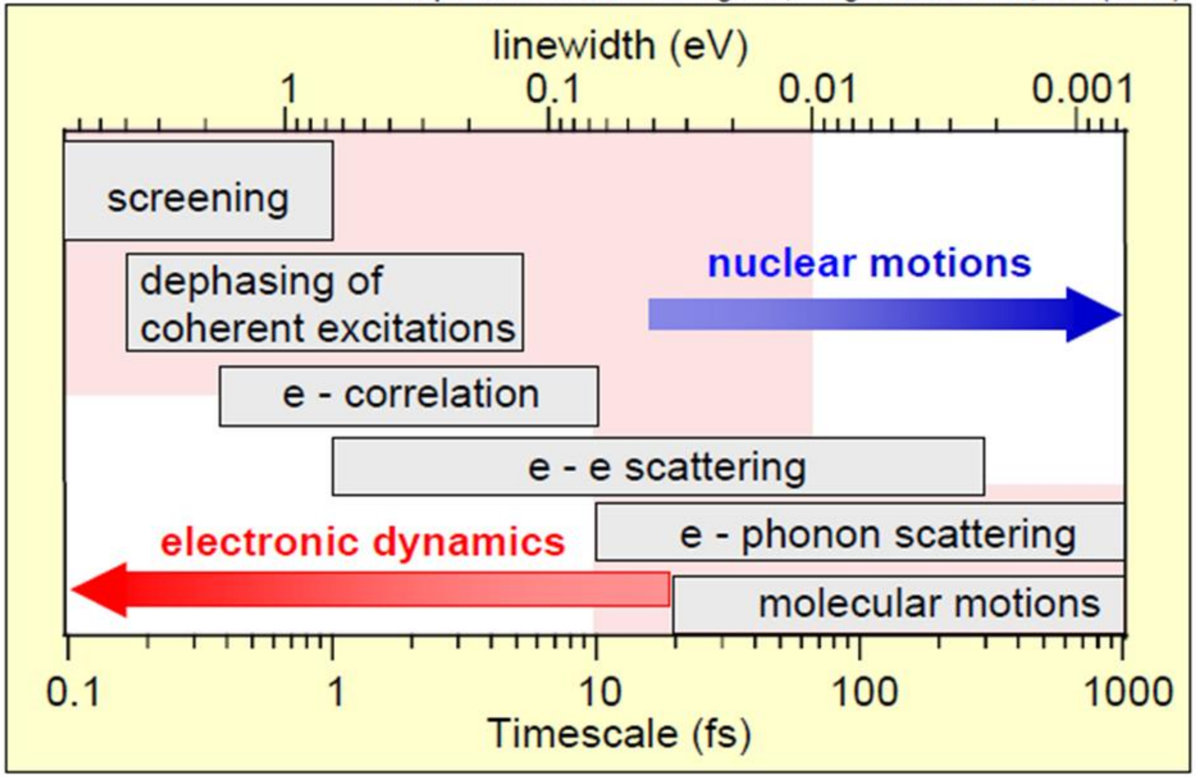
Time-resolved ARPES

Probing unoccupied states of Bi_2Se_3 with 2PPE



Time-resolved ARPES

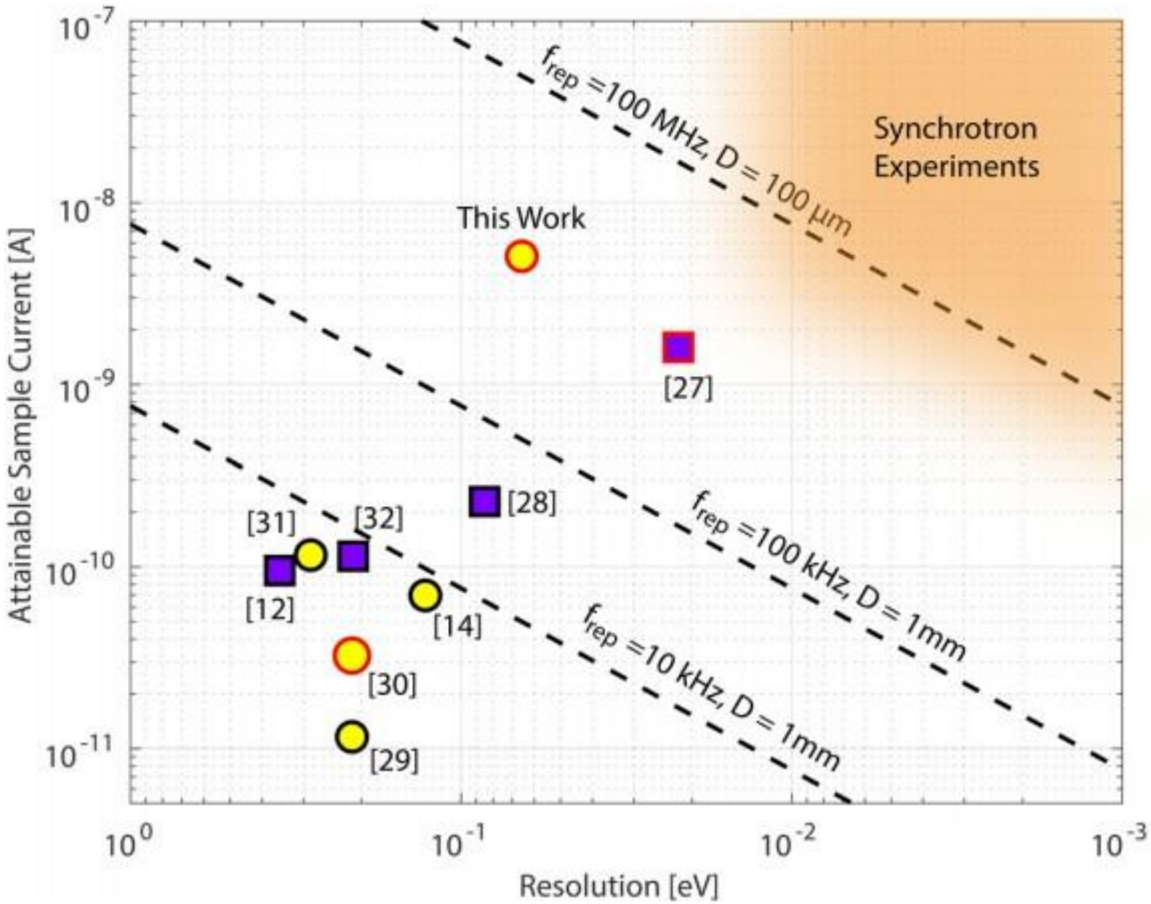
adapted from Petek and Ogawa, Prog. Surf. Sci. **56**, 239 (1997)



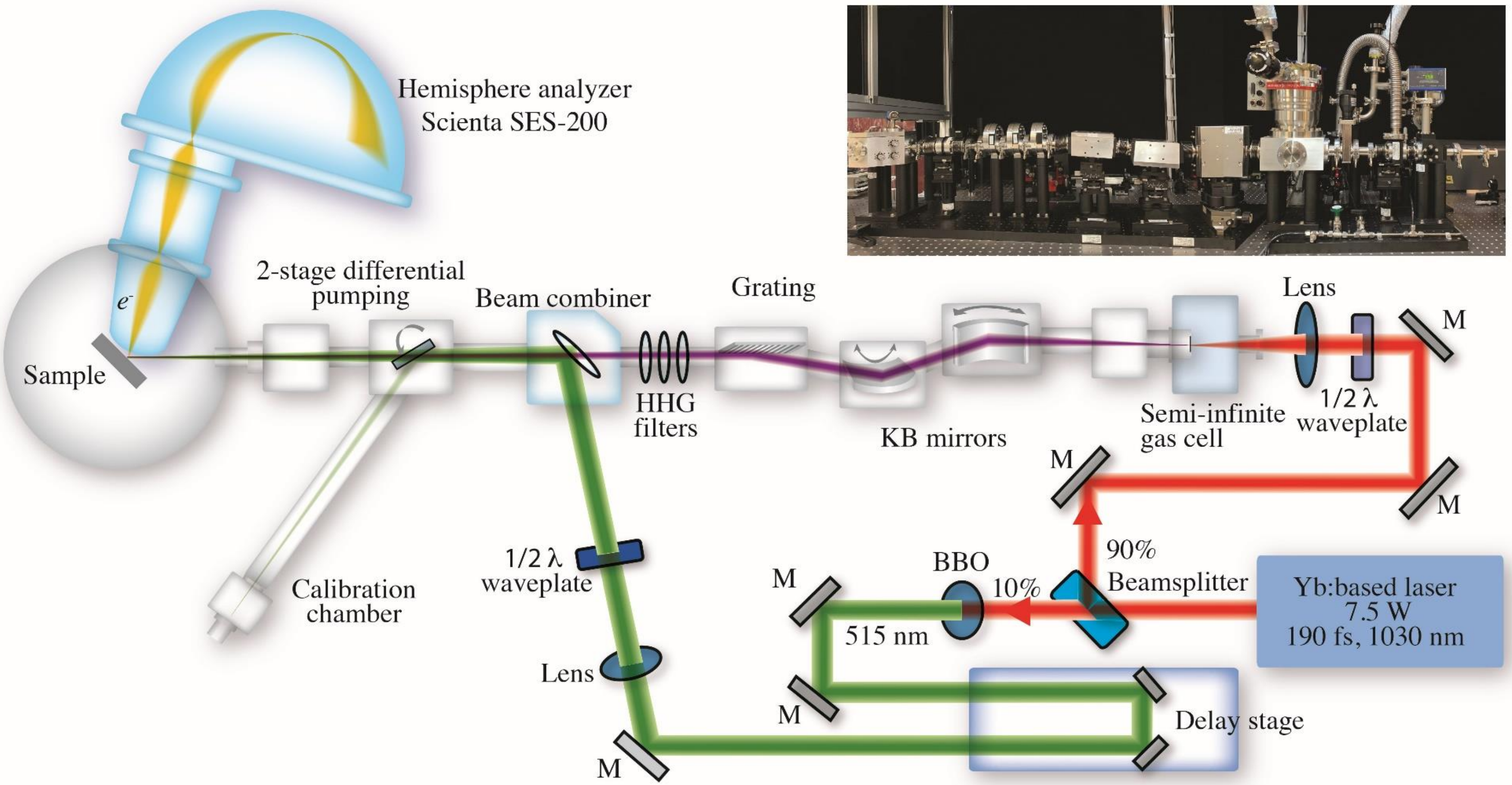
- Hierarchy of time scales: from as to ps
- Separation of electronic and nuclear dynamics

Time-resolved ARPES

High repetition rate/ lower photon flux in single pulse



Time-resolved ARPES



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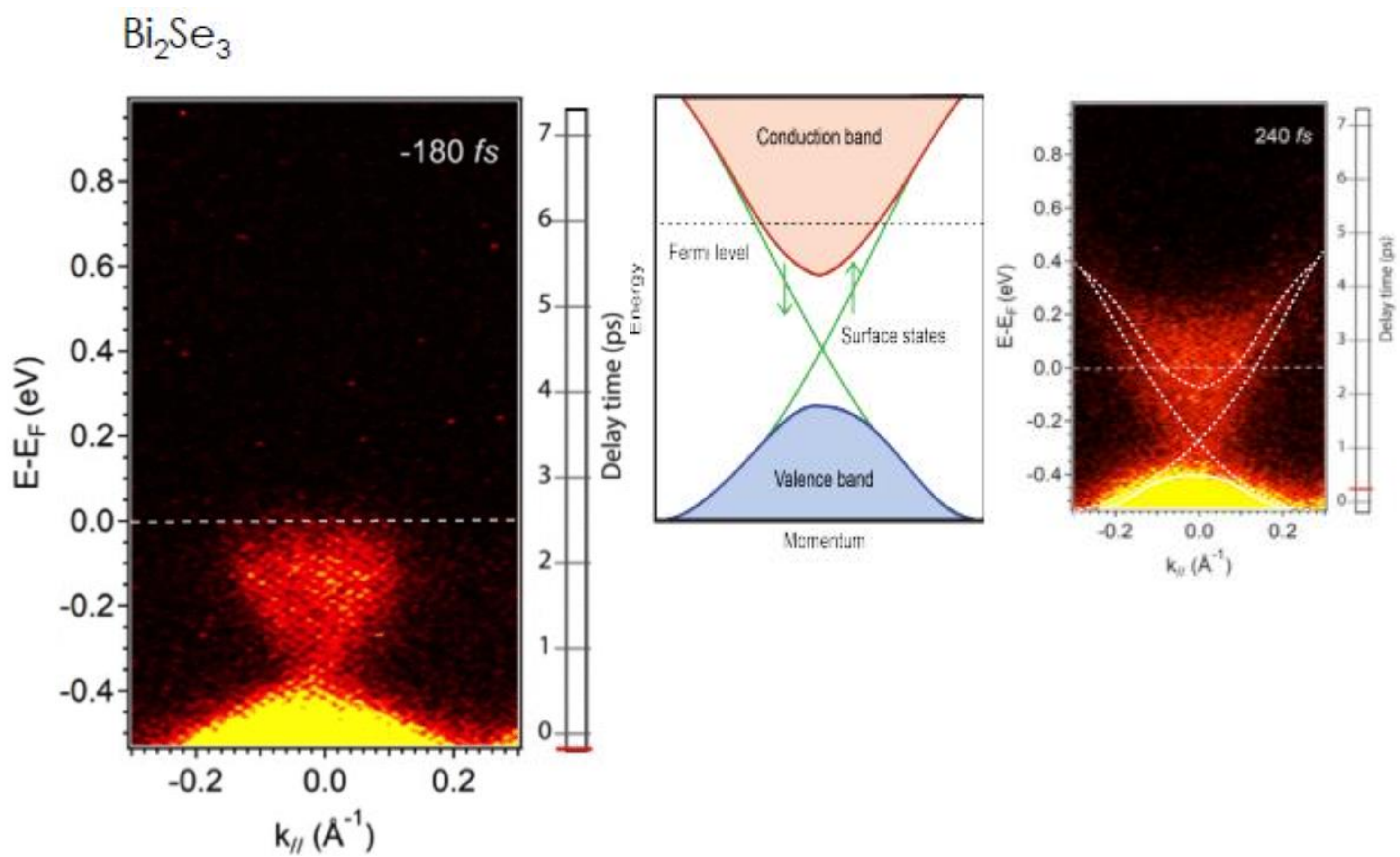
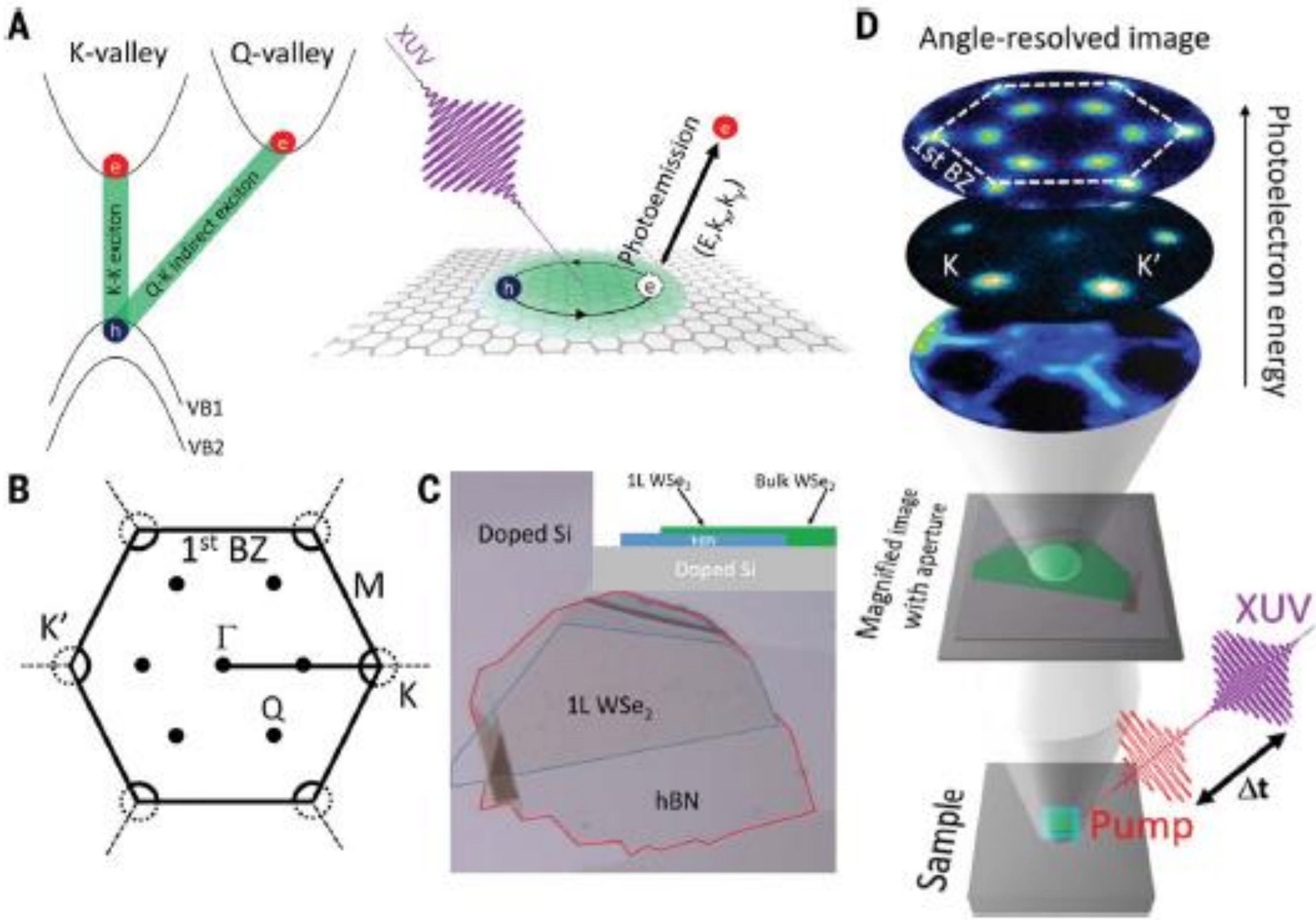
Time-resolved ARPES



Excitons and their dynamics in atomically thin semiconductors



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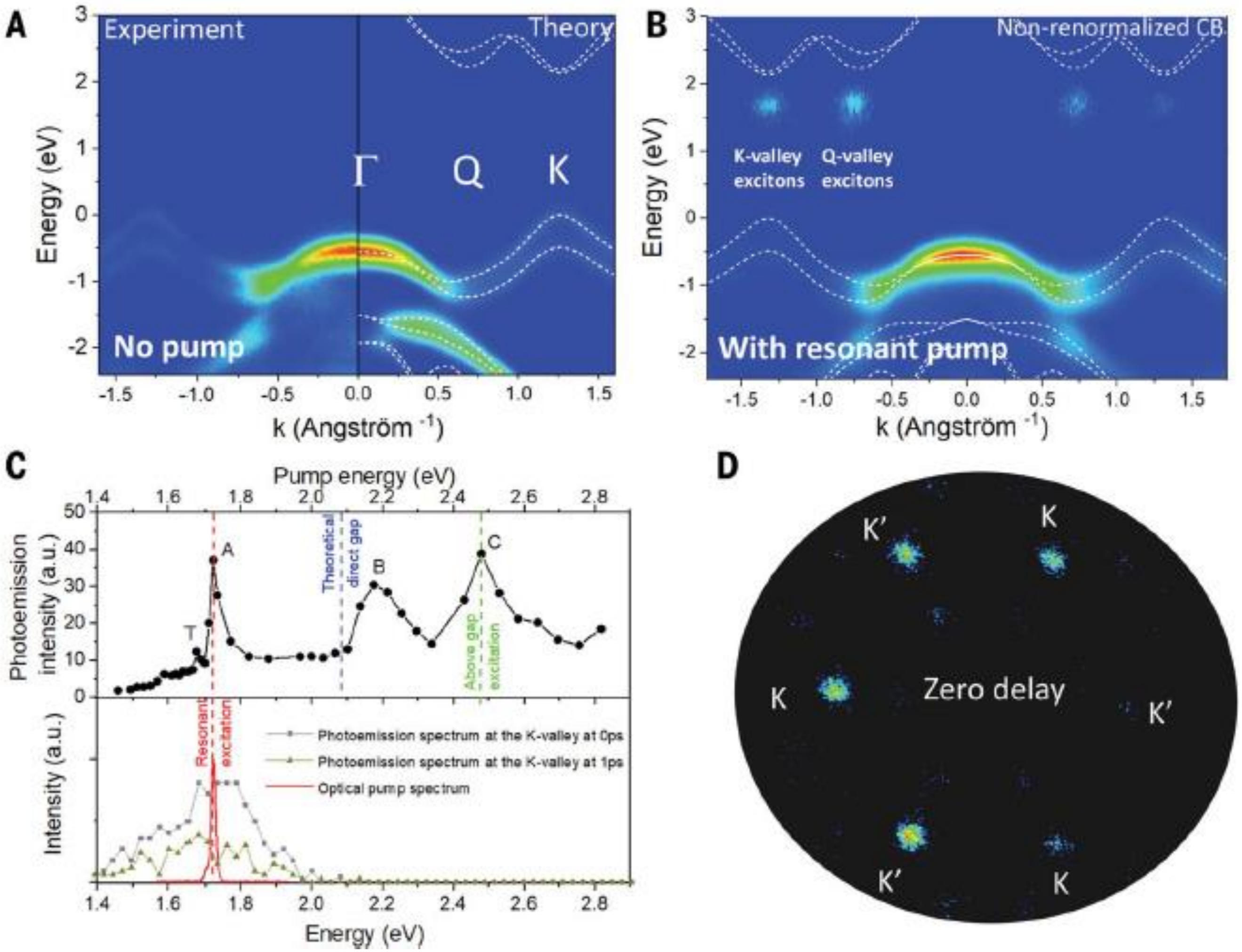
Madeo et al., Science (2020)

Excitons and their dynamics in atomically thin semiconductors



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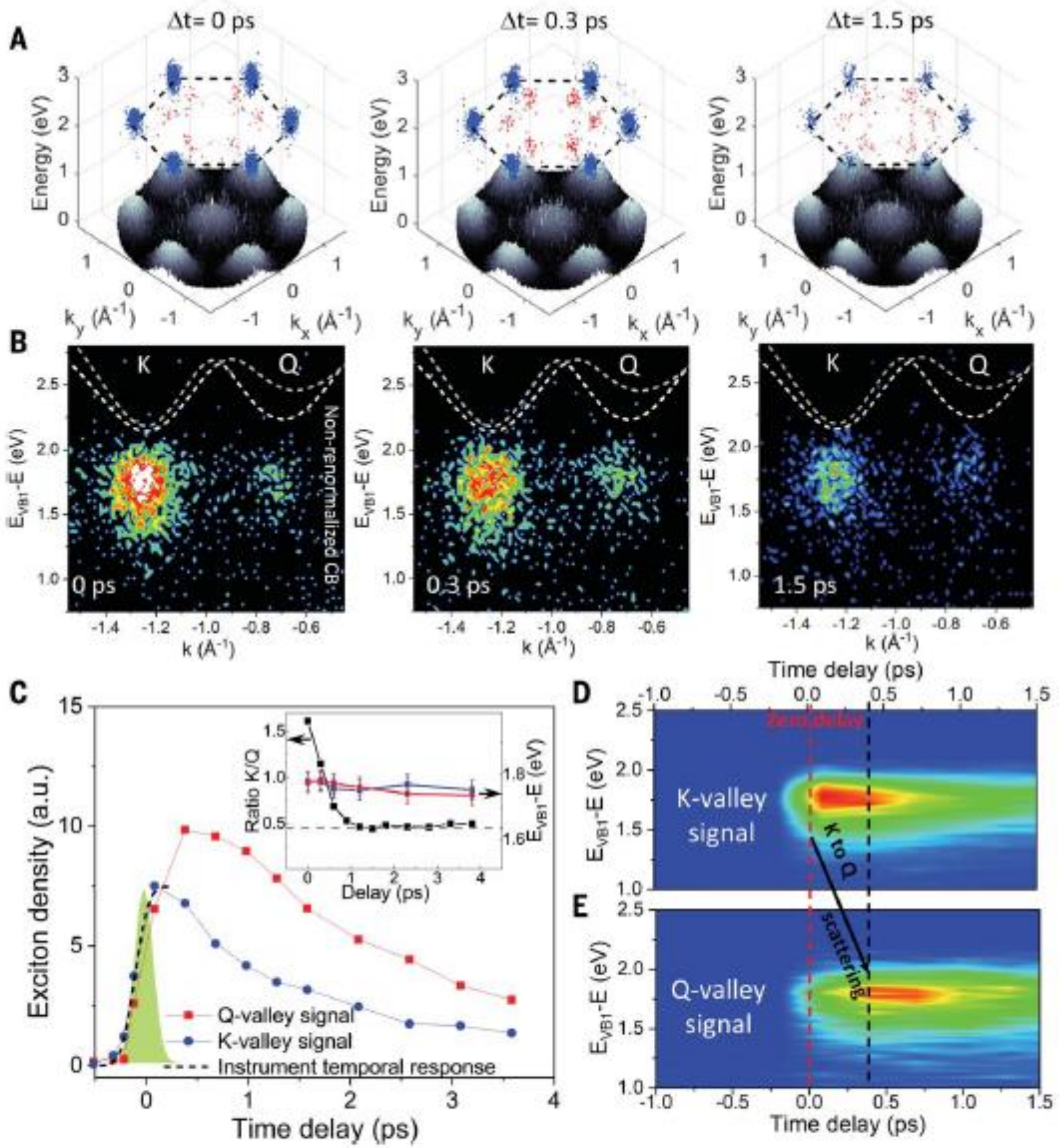
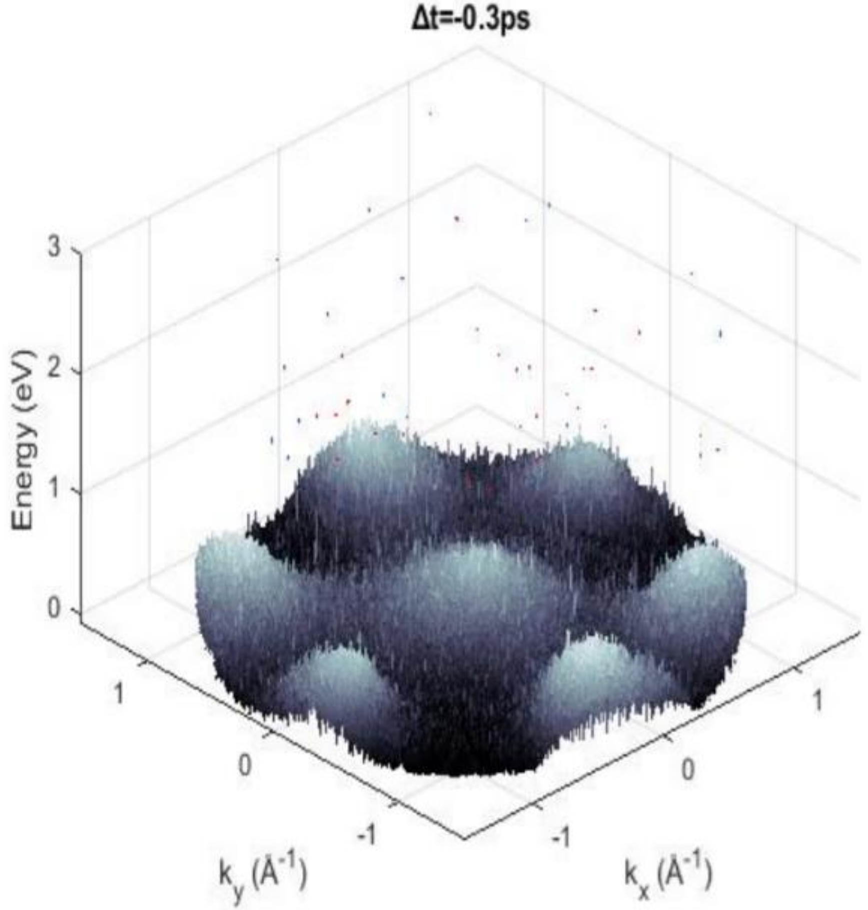
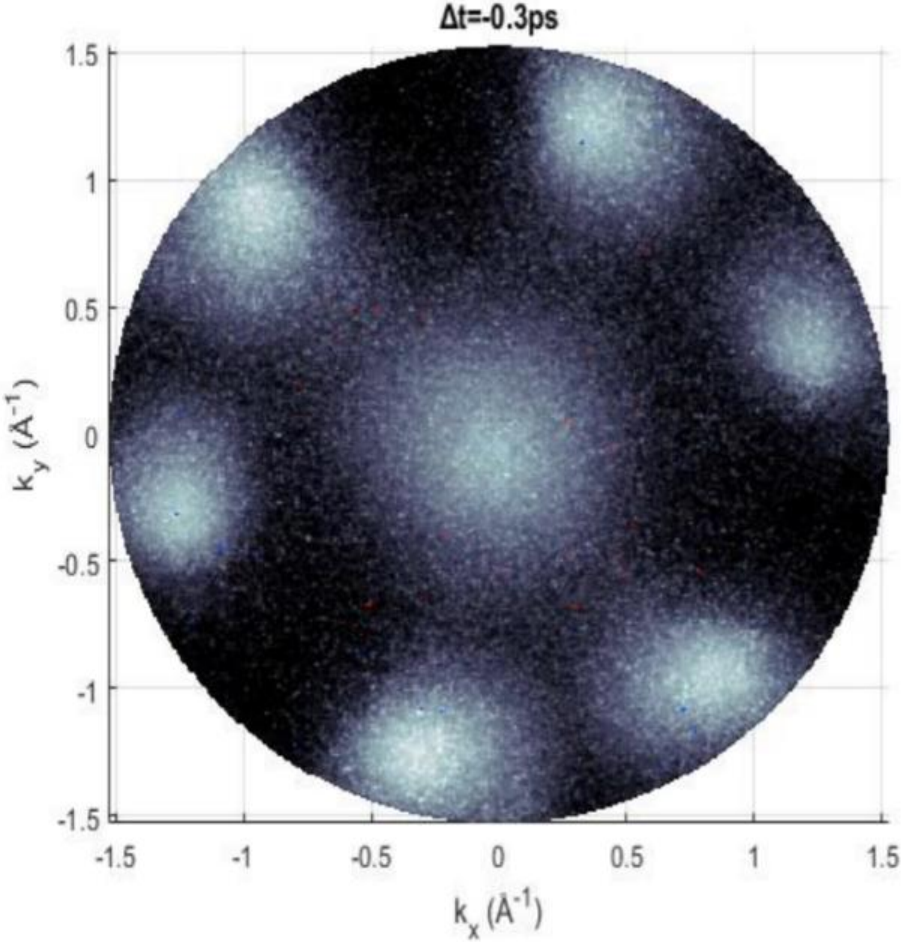
Excitons and their dynamics in atomically thin semiconductors

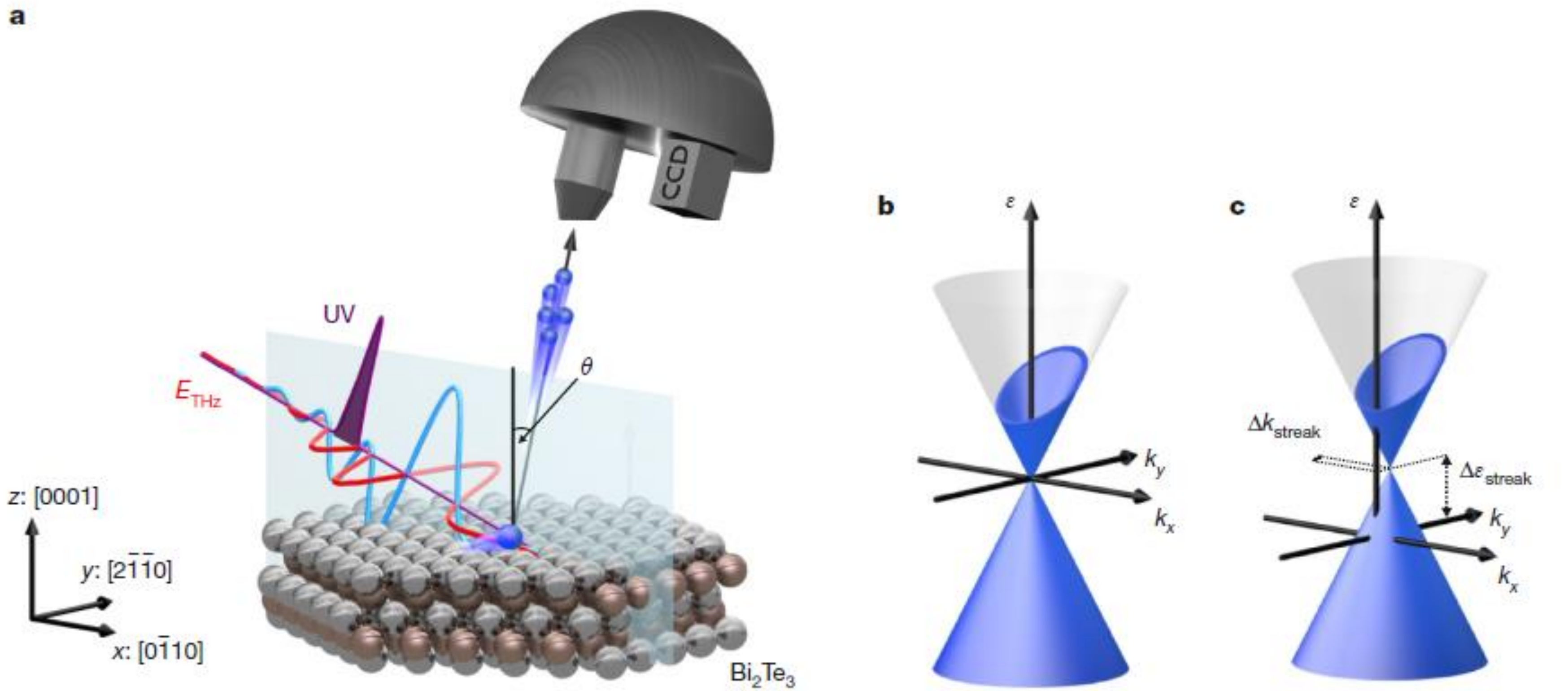


Above Bandgap Excitation





Time-resolved ARPES : THz exciting source



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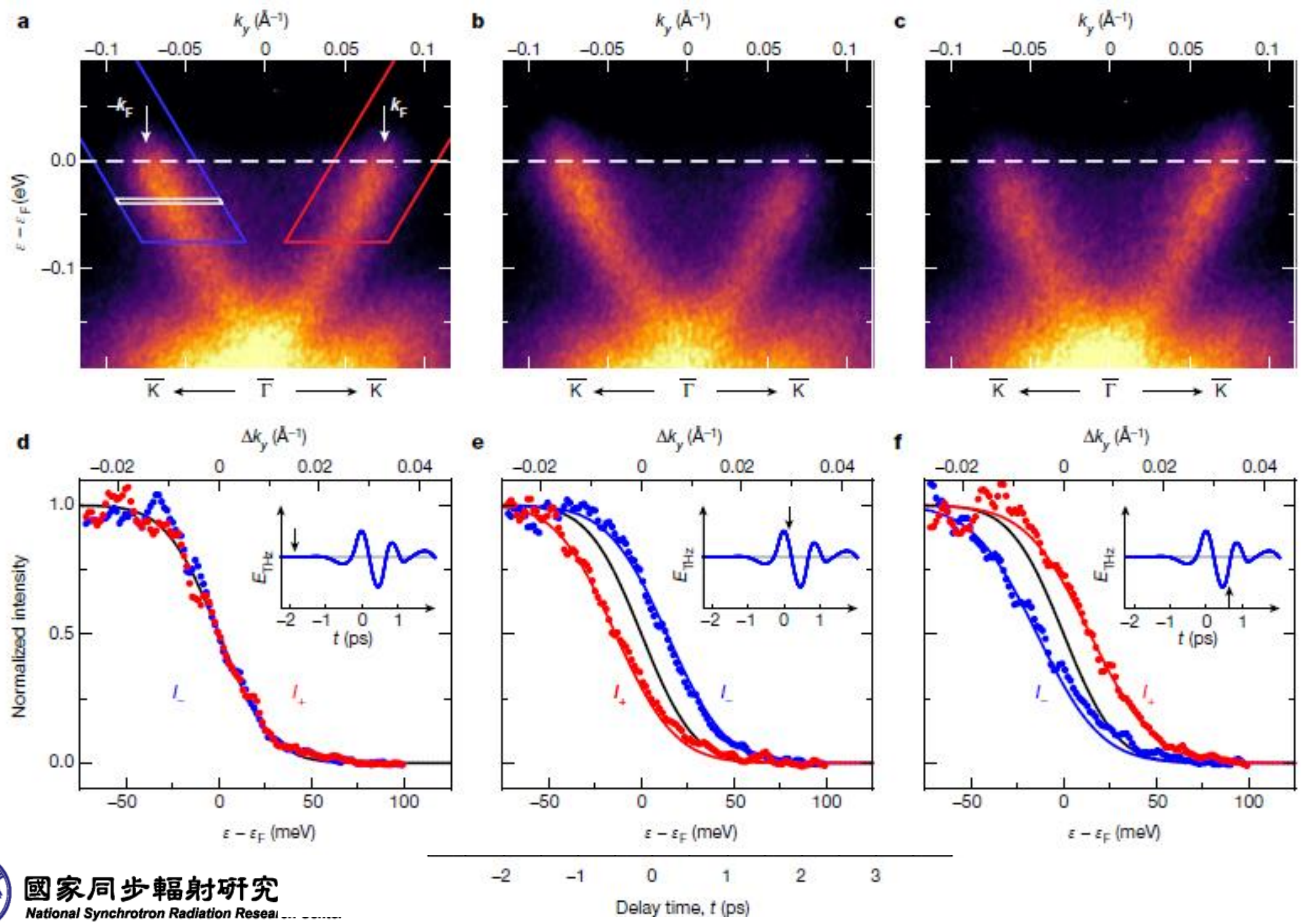
Reimann et al., Nature (2018)

Time-resolved ARPES :THz exciting source



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Reimann et al., Nature (2018)

Taiwan Photon Source and Taiwan Light Source

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