

Visualizing Electronic Structures of Quantum Materials

– By Angle Resolved Photoemission Spectroscopy (ARPES)

PART B: New Frontier in ARPES

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New Frontiers

Complete photo-electron
spectroscopy

$$f(k, E, t, r, \sigma)$$

Momentum

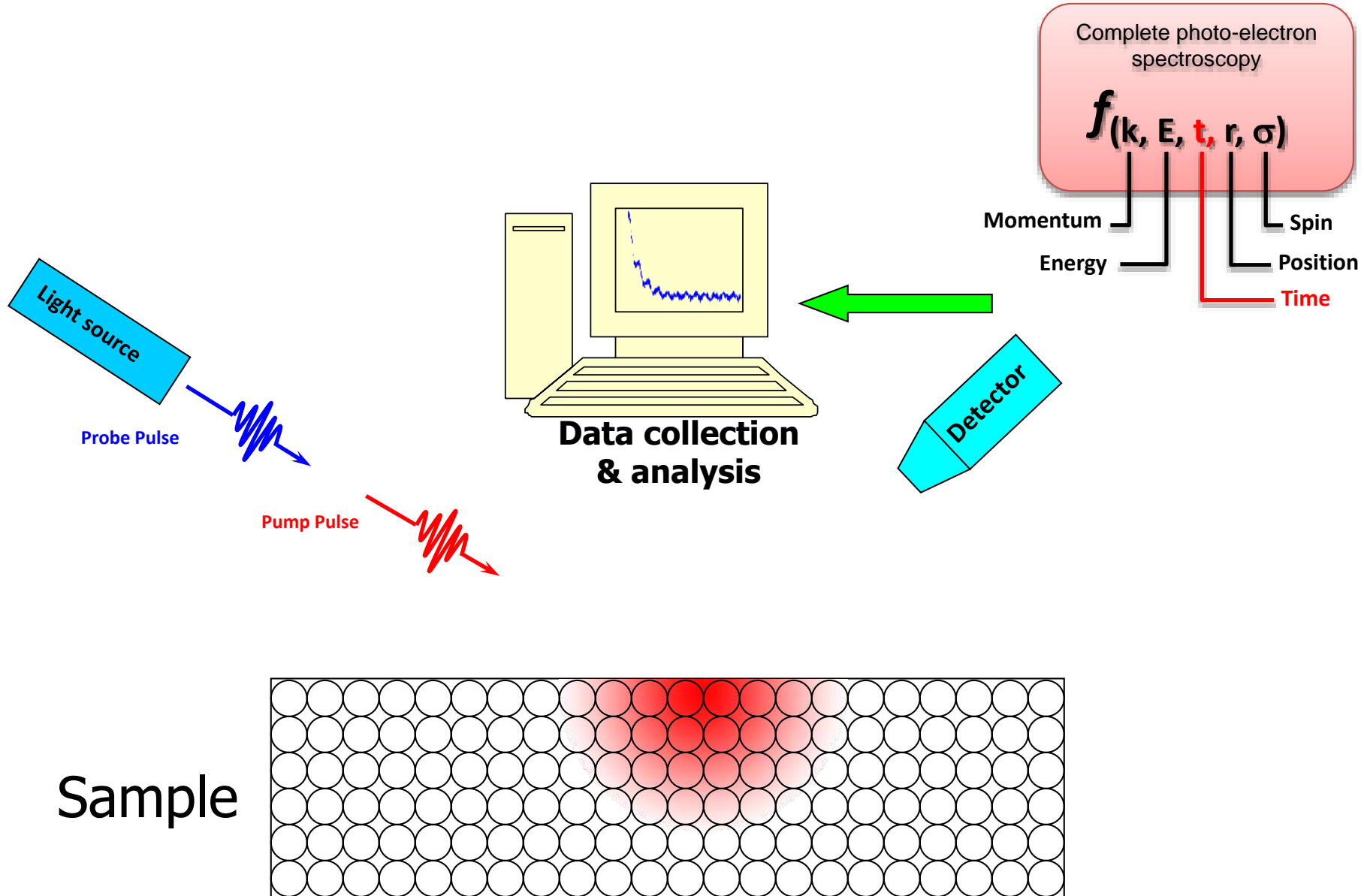
Energy

Spin

Position

Time

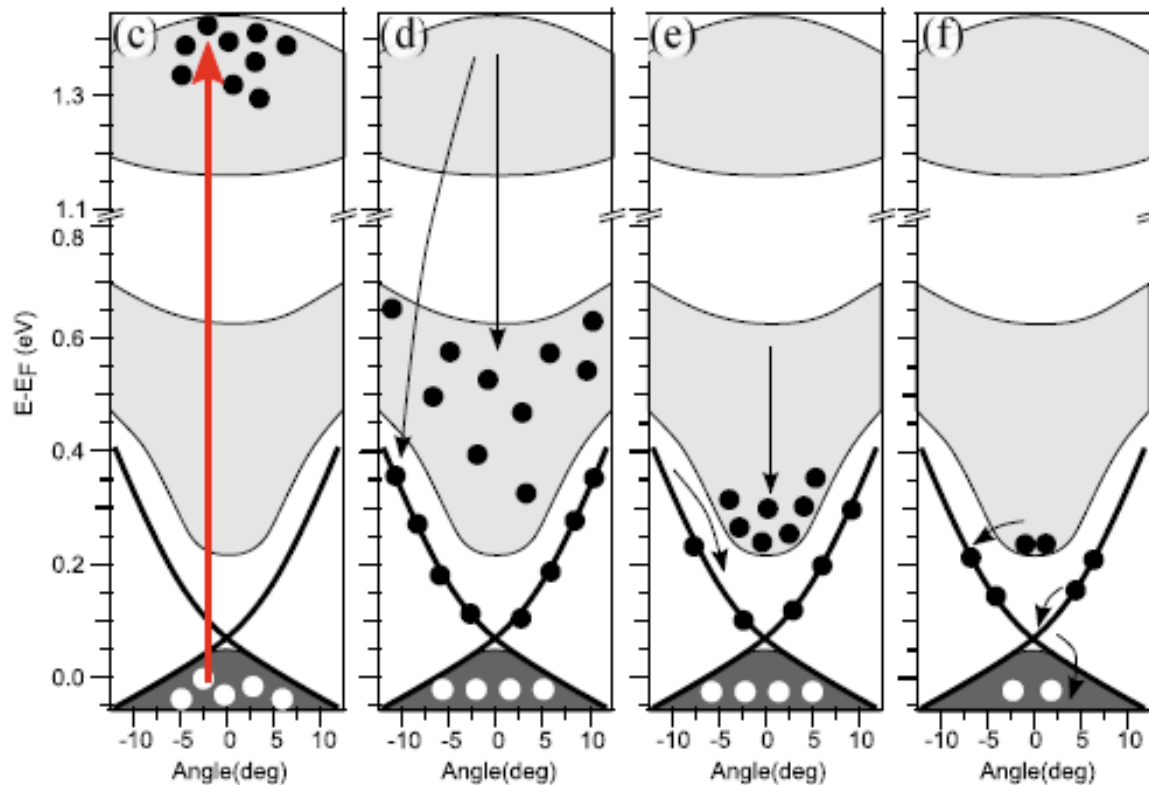
Explore electron dynamics



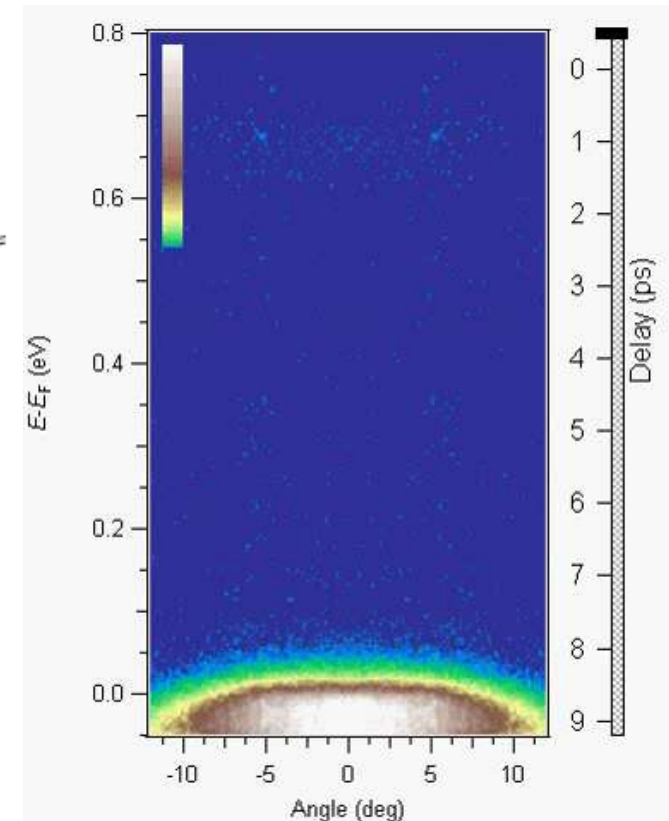
Long lived surface electrons of Bi_2Se_3

J. Sobota, *et al. Phys. Rev. Lett.* **108**, 117403 (2012)

Physical process



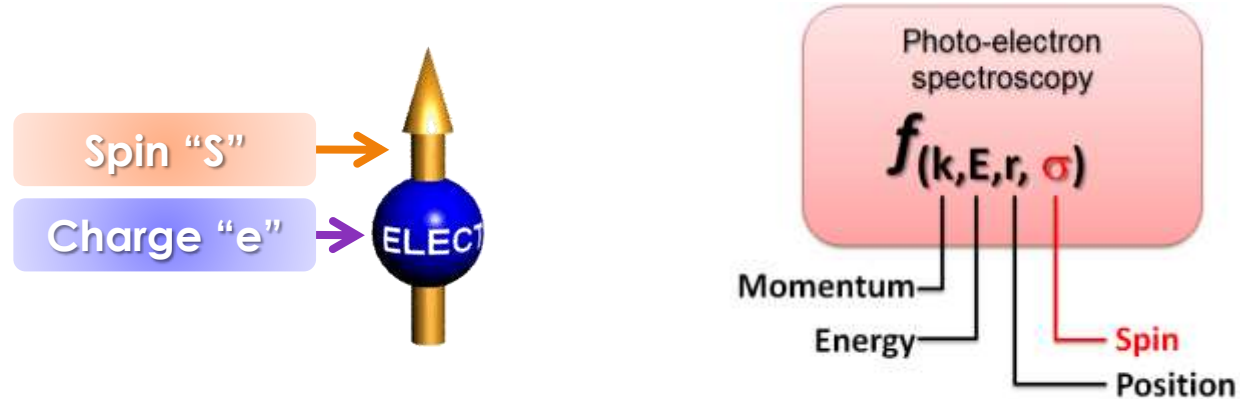
Measurement



Spin resolved ARPES

New frontier

Explore electron spin



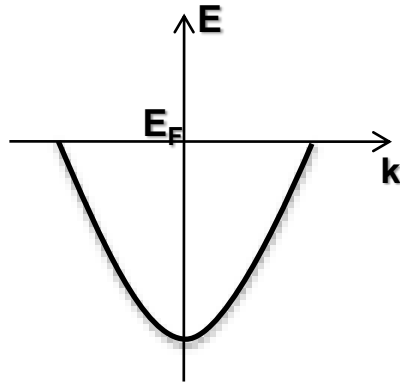
Spin information is important for:

- Topological quantum materials
- CMR materials
- Novel superconductivities
- Multiferroic materials
- Heavy fermion systems
- Spintronics applications

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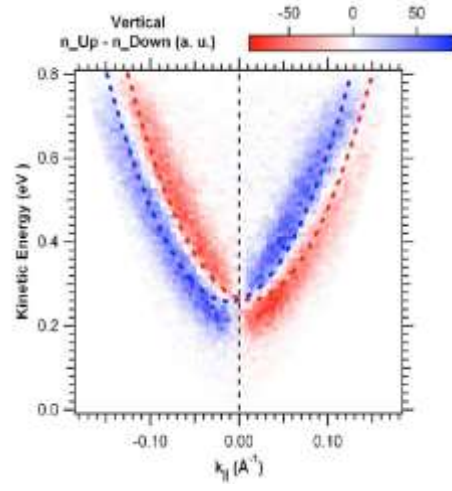
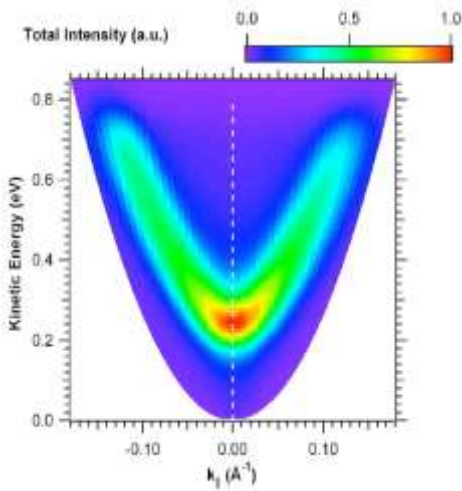
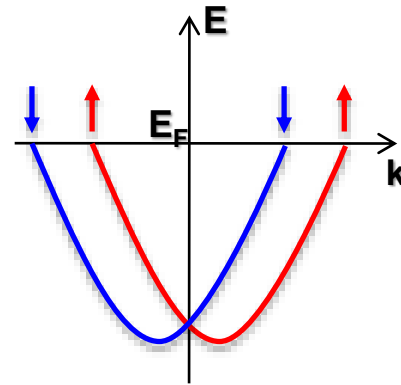
Materials with spin-dependent electronic structure

Free electron dispersion



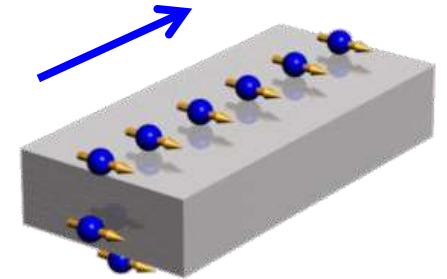
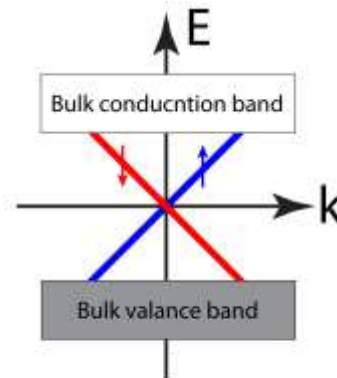
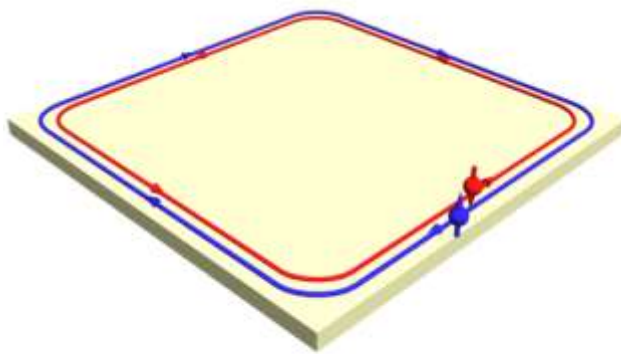
SOC
→

Free electron dispersion

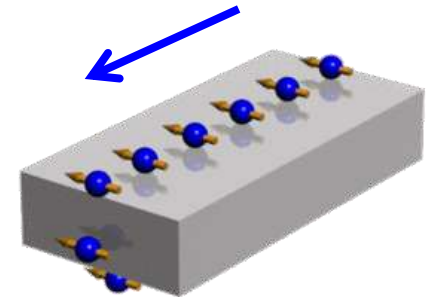
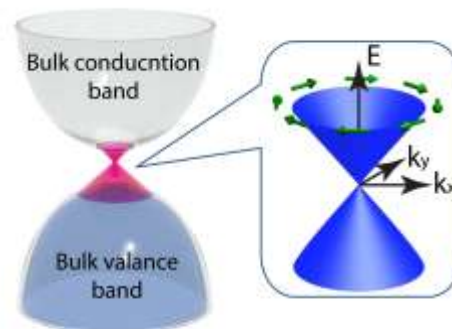
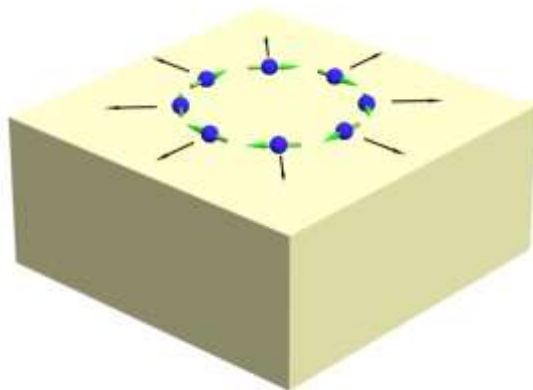


Exotic spin states: Topological insulators

“Locking” of current & spin



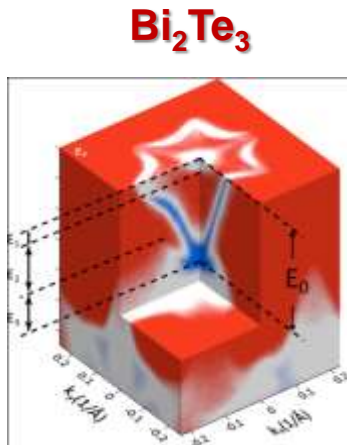
2D Topological insulator (QSH insulator)



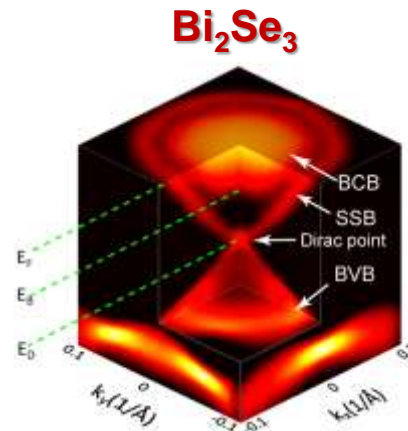
3D Topological insulator

Understand the spin of topological surface electrons

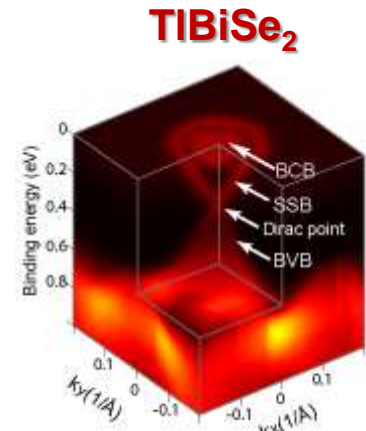
Regular ARPES
3D structure



Y. L. Chen, *et al.*,
Science 325, 178 (2009)

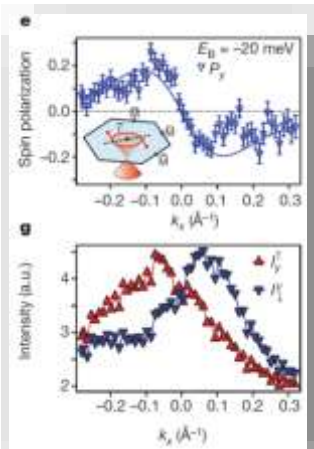


Y. L. Chen, *et al.*,
Science 329, 659 (2010)

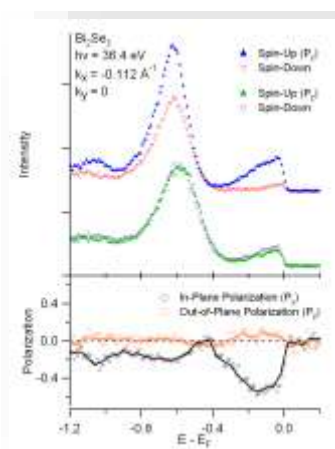


Y. L. Chen, *et al.*,
Phys. Rev. Lett., 105 266401 (2010)

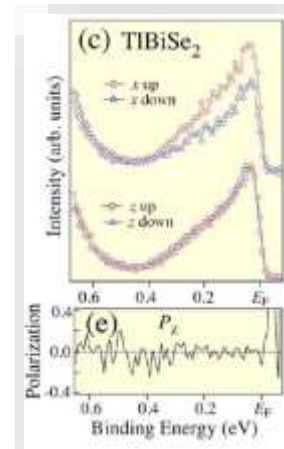
Spin-ARPES
1D structure



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



C. Jozwiak, *et al.*,
Phys. Rev. B, **84**, 165113 (2011)

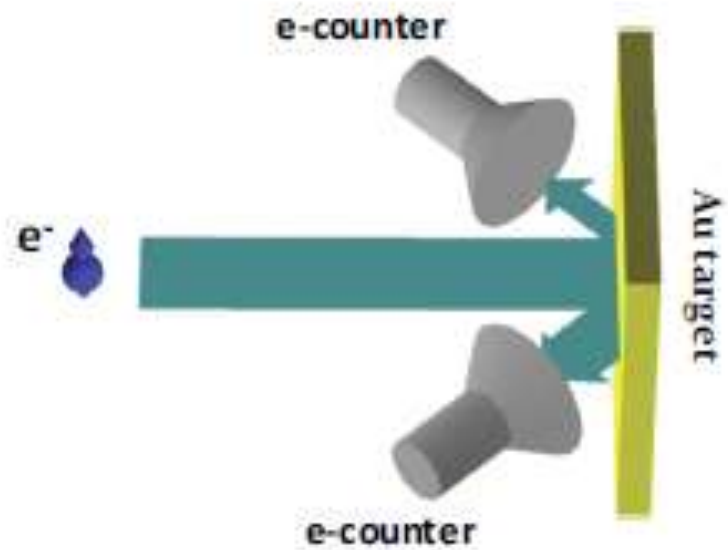
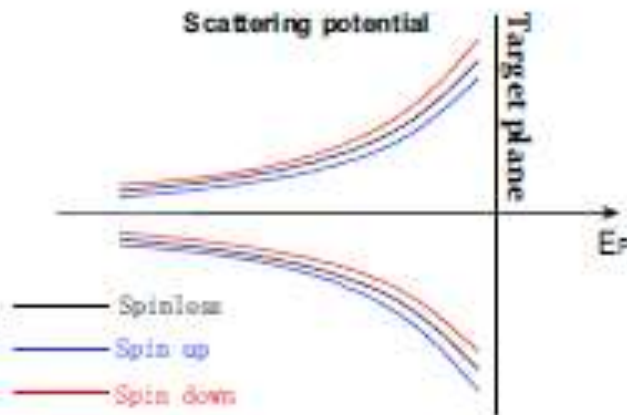
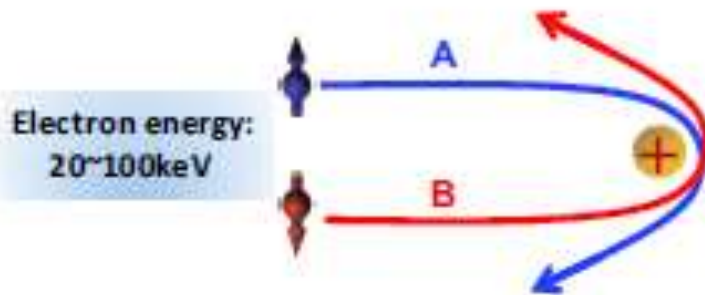


Souma, *et al.*,
Phys. Rev. Lett., 106, 216803 (2011)

Detecting Electron Spin

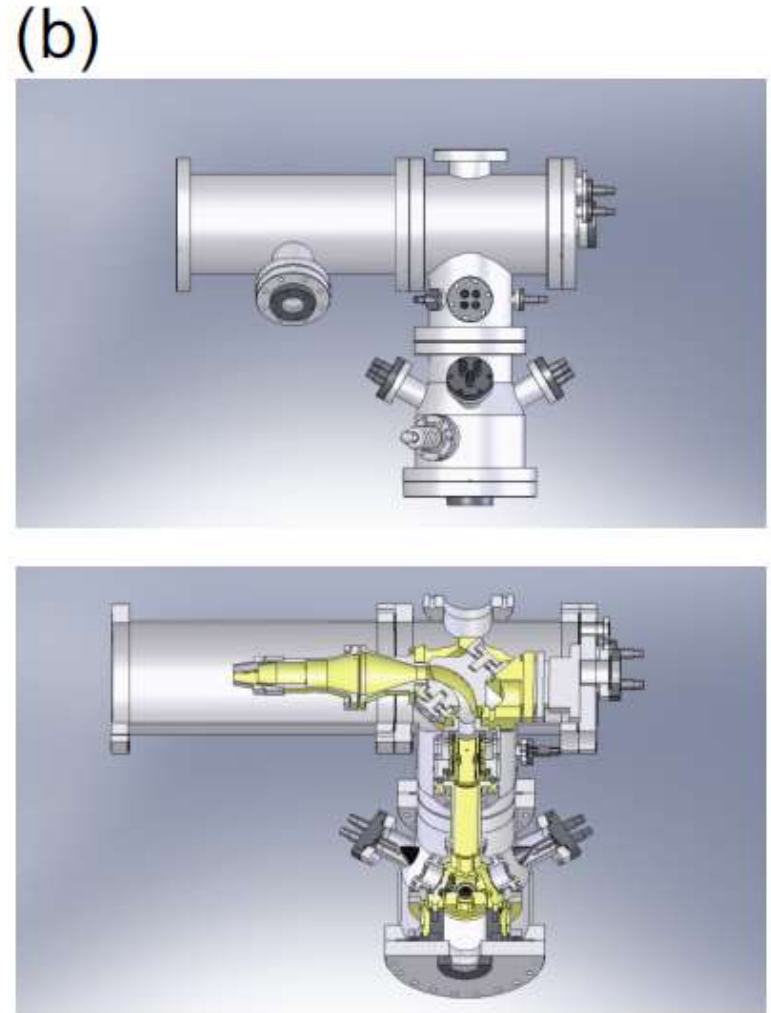
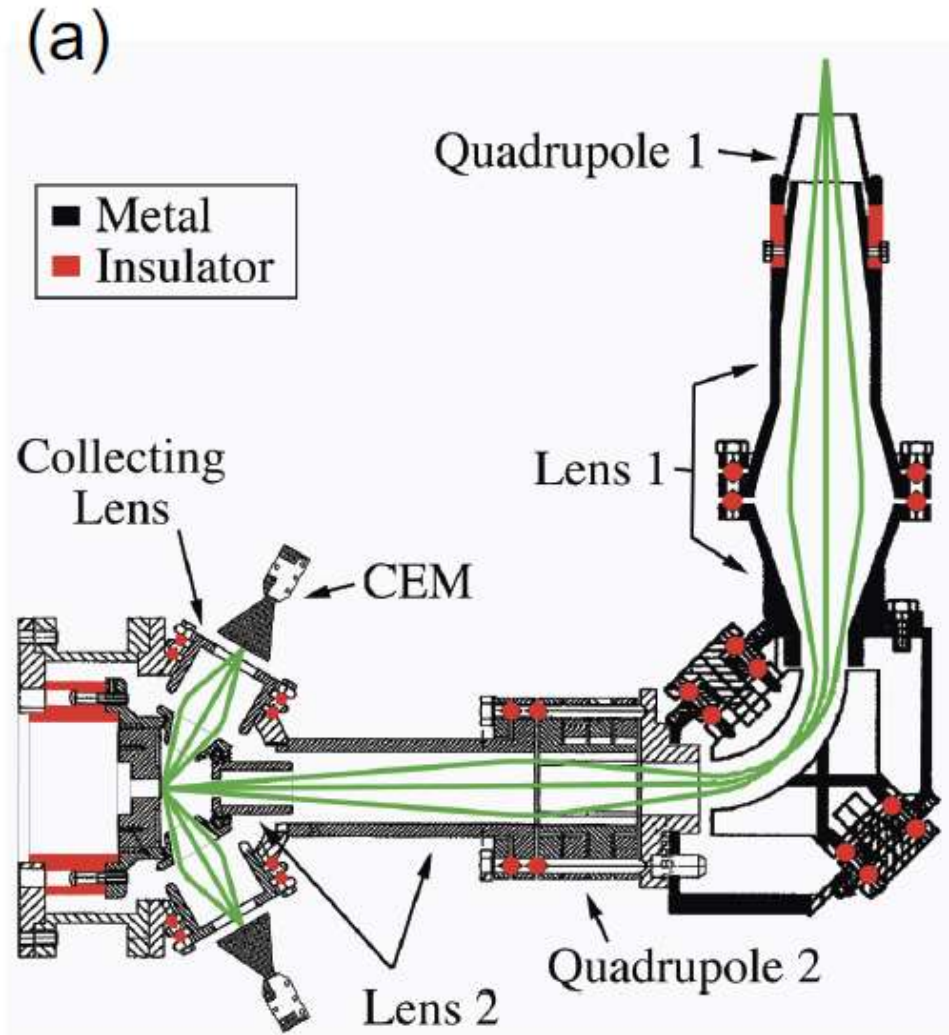
Mott scattering spin polarimeter

Mott Scattering



Detecting Electron Spin

Mott scattering spin polarimeter



Detecting Electron Spin

Figure of Merit

$$A = \frac{I_{left} - I_{right}}{I_{left} + I_{right}} = PS \quad \Rightarrow \quad P = \frac{A}{S} \Rightarrow \Delta P = \frac{\Delta A}{S} = \sqrt{\frac{1}{IS^2}}$$

$$\begin{aligned} \Delta A &= \sqrt{\left(\frac{\partial A}{\partial I_{left}}\right)^2 (\Delta I_{left})^2 + \left(\frac{\partial A}{\partial I_{right}}\right)^2 (\Delta I_{right})^2} \\ &= \sqrt{\left(\frac{2I_{right}}{(I_{left} + I_{right})^2}\right)^2 I_{left} + \left(-\frac{2I_{left}}{(I_{left} + I_{right})^2}\right)^2 I_{right}} \end{aligned}$$

as $I = I_{left} + I_{right}$

$$= \sqrt{\frac{4I_{left}I_{right}}{I^3}}$$

$$4I_{left}I_{right} = I^2(1 - P^2S^2) \Rightarrow 4I_{left}I_{right} \approx I^2$$

$$\approx \sqrt{\frac{1}{I}}$$

$$\Rightarrow \Delta P = \frac{\Delta A}{S} = \sqrt{\frac{1}{IS^2}}$$

Detecting Electron Spin

Figure of Merit

$$\Delta P = \frac{\Delta A}{S} = \sqrt{\frac{1}{IS^2}}$$

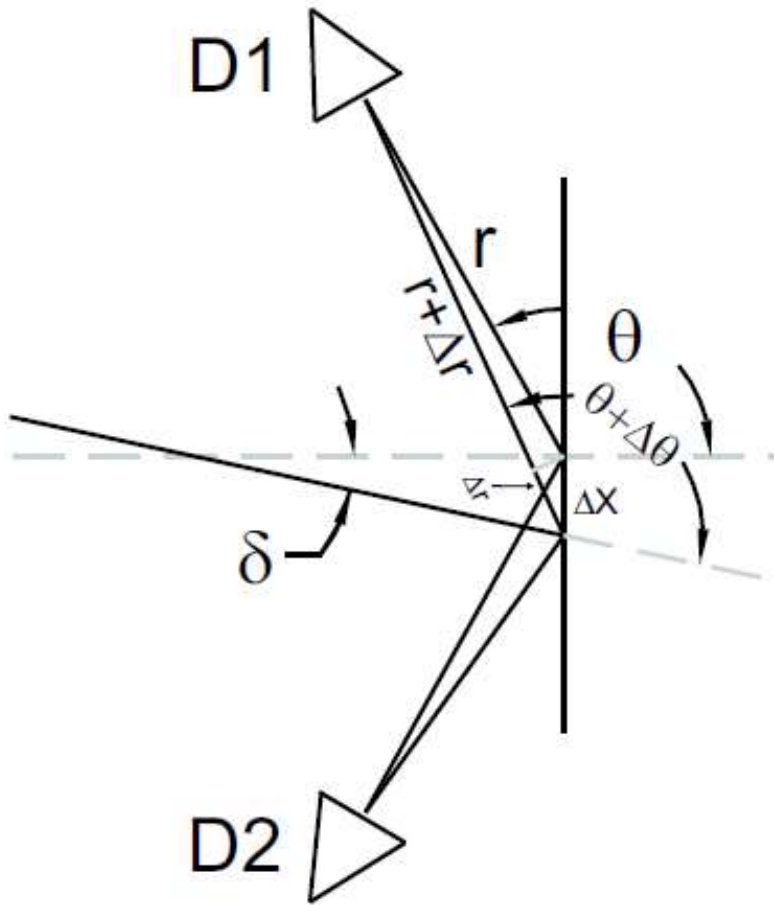
To minimize ΔP , we need to maximize IS^2

So normalized by the total initial flux I_0 , the index is defined as
“Figure of Merit” (FOM)

$$FOM = S^2 \frac{I}{I_0}$$

Detecting Electron Spin

Mott scattering spin polarimeter



$$N^+ = \sqrt{L_{\uparrow}R_{\downarrow}}, \quad N^- = \sqrt{R_{\uparrow}L_{\downarrow}}$$

$$L_{\uparrow} = nNE_l\Omega_l(\Delta r, \Delta\theta)\sigma(\theta + \Delta\theta)(1 + PS(\theta) + P\frac{\partial S}{\partial\theta}\Delta\theta)$$

$$R_{\uparrow} = nNE_r\Omega_r(\Delta r, \Delta\theta)\sigma(\theta + \Delta\theta)(1 - PS(\theta) + P\frac{\partial S}{\partial\theta}\Delta\theta)$$

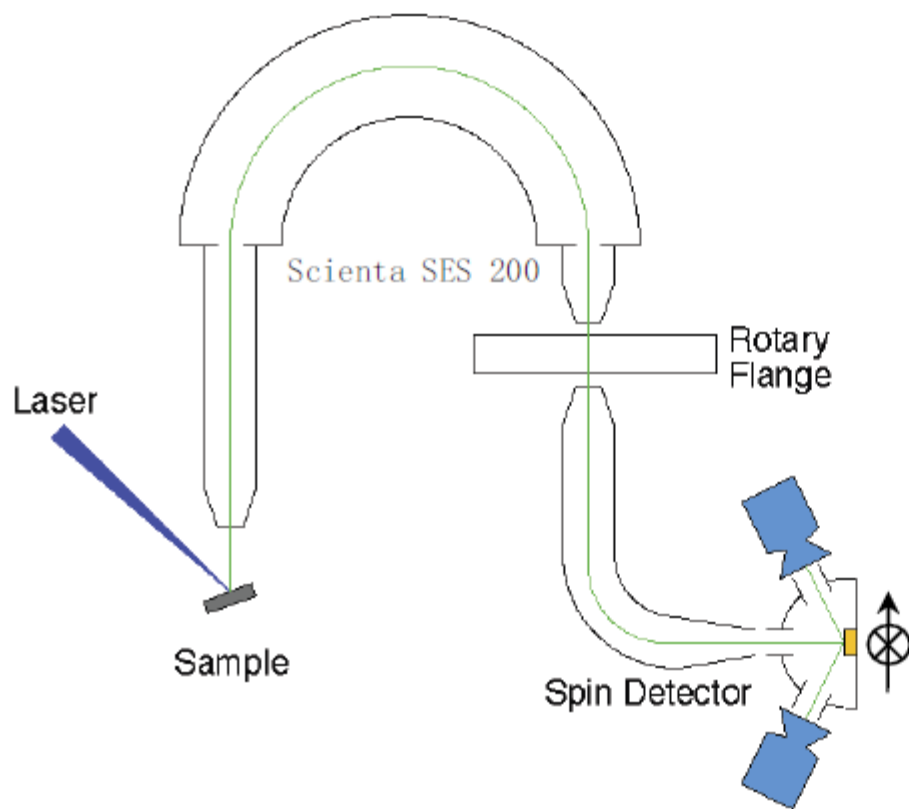
$$L_{\downarrow} = n'NE_l\Omega_l(\Delta r, \Delta\theta)\sigma(\theta + \Delta\theta)(1 - PS(\theta) - P\frac{\partial S}{\partial\theta}\Delta\theta)$$

$$R_{\downarrow} = n'NE_r\Omega_r(\Delta r, \Delta\theta)\sigma(\theta + \Delta\theta)(1 + PS(\theta) - P\frac{\partial S}{\partial\theta}\Delta\theta)$$

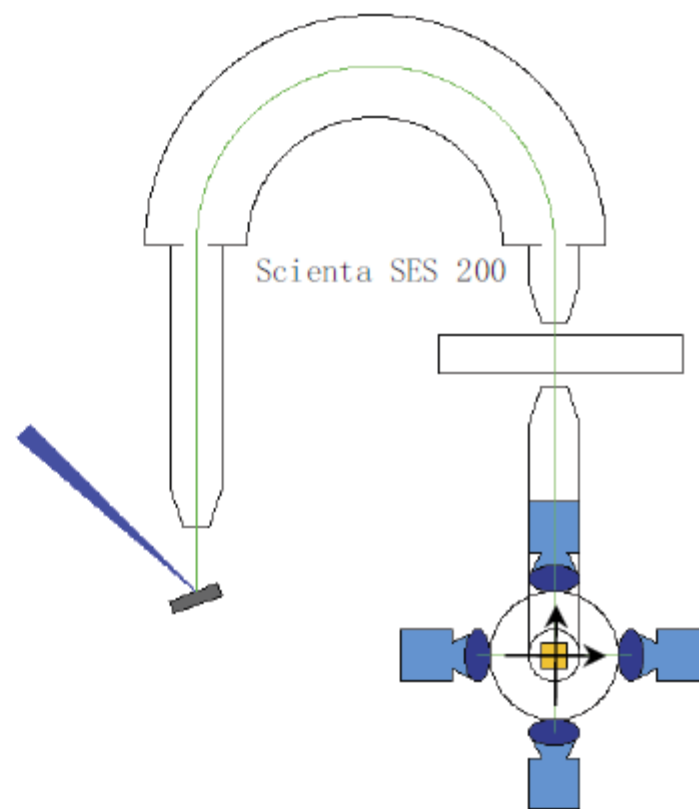
$$A = \frac{N^+ - N^-}{N^+ + N^-} = PS$$

Detecting Electron Spin

Mott scattering spin polarimeter



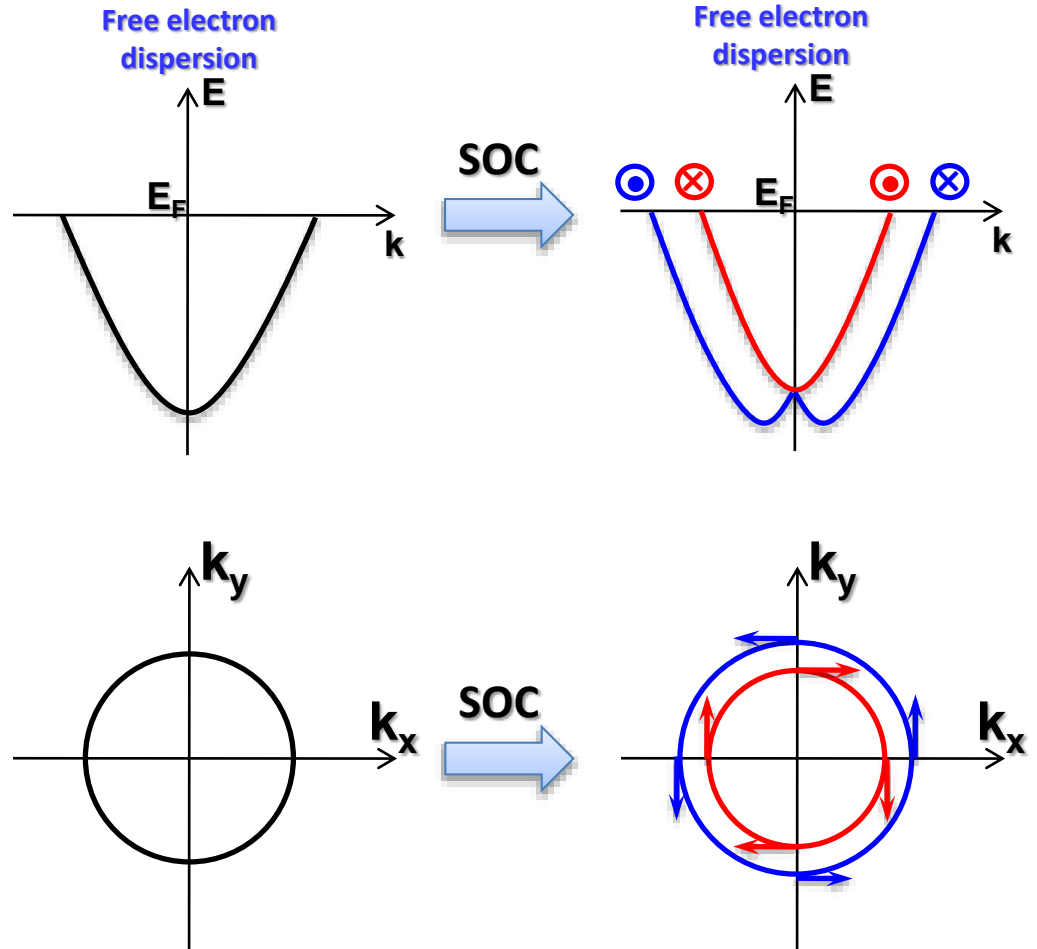
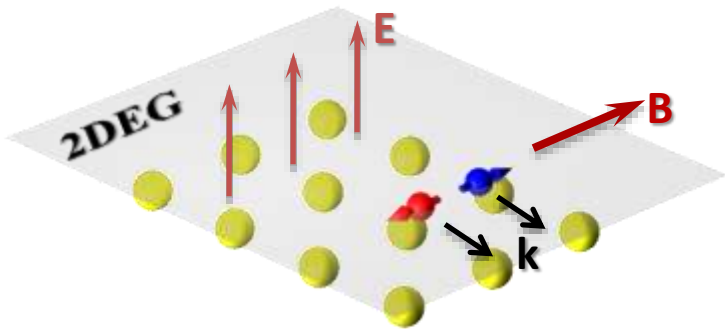
First Arrangement:
Measures P_{\otimes} and P_{\uparrow}



Second Arrangement:
Measures P_{\uparrow} and P_{\rightarrow}

Spin-resolved laser ARPES

Spin-orbital splitting of the surface state band

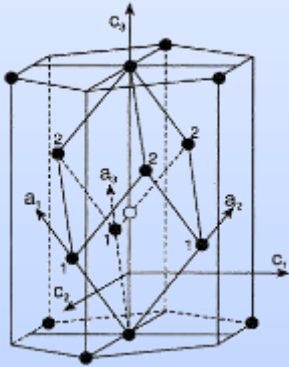


Spin-resolved laser ARPES

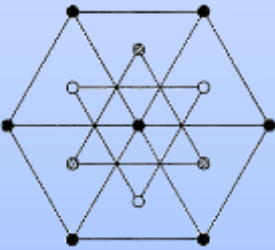
Sb(111) Surface state

Structural Aspect

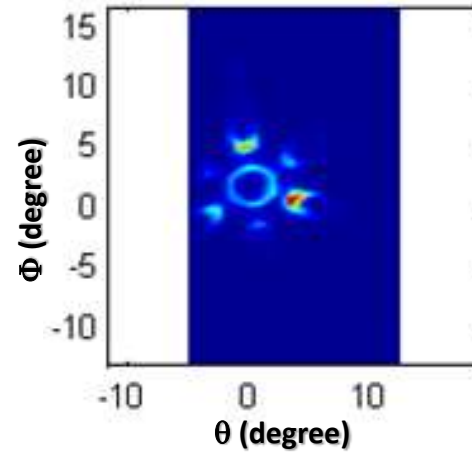
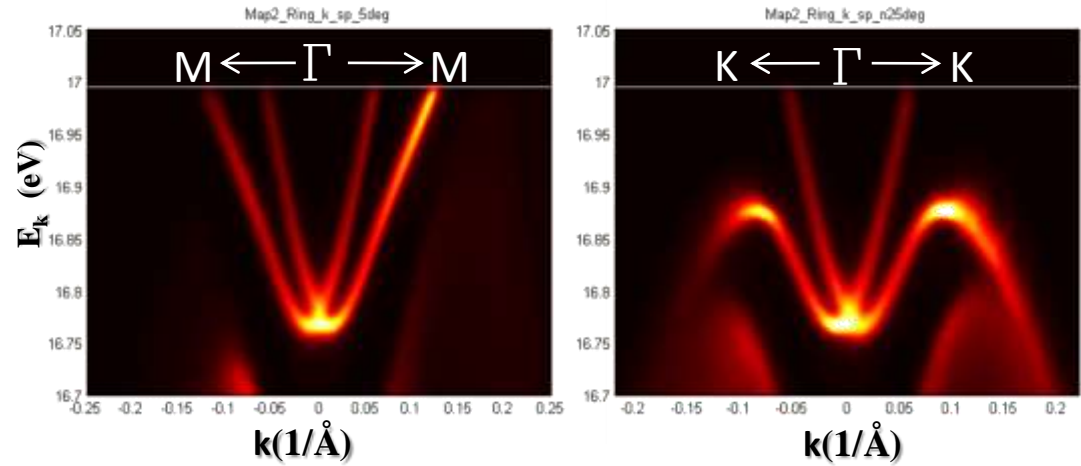
Rhombohedral Lattice



Top View

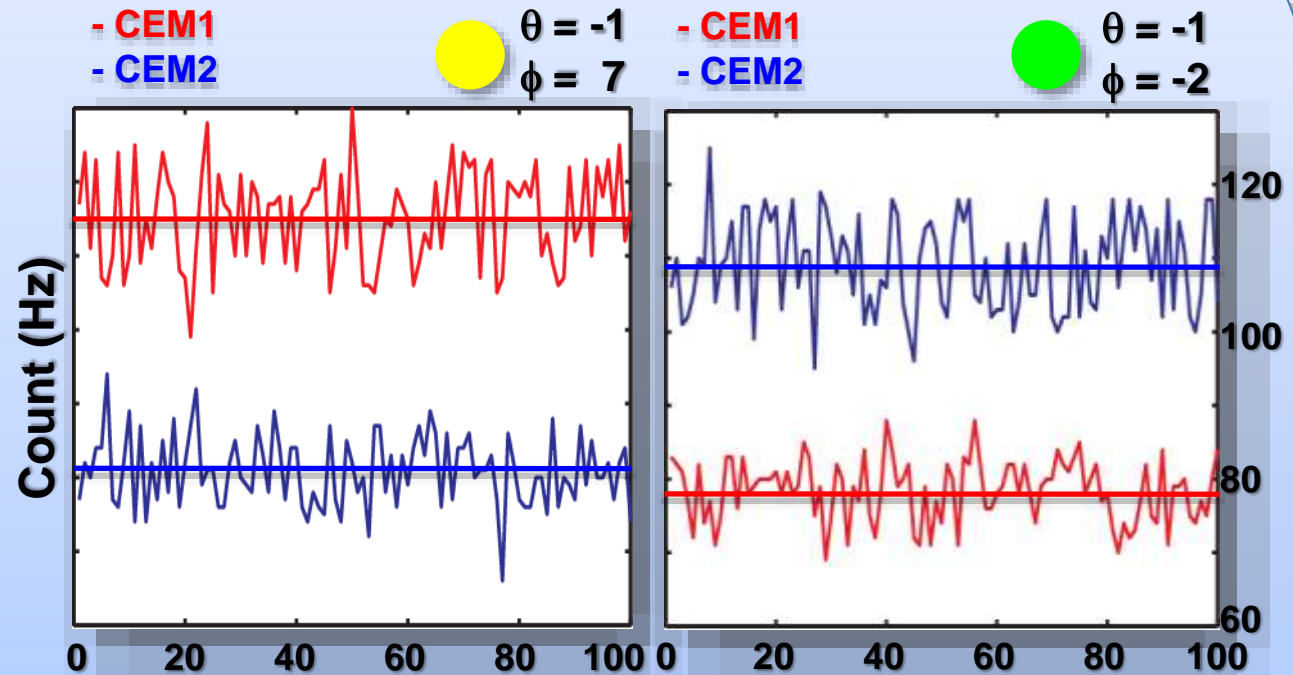
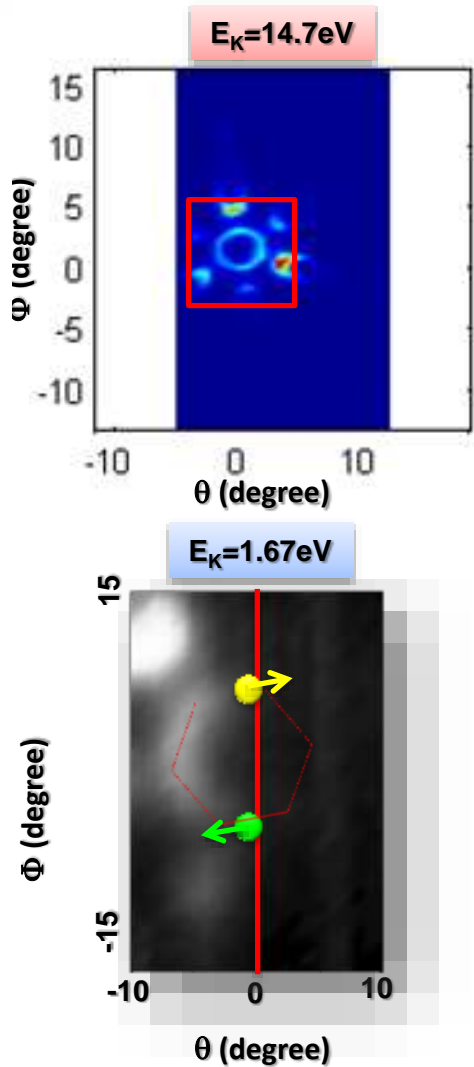


Regular ARPES



Spin-resolved laser ARPES

Spin direction of the FS



$$A = \frac{\overline{C1} - \overline{C2}}{\overline{C1} + \overline{C2}} = 0.175$$

$$P = \frac{A}{S} = 84.7 \pm 8.7\%$$

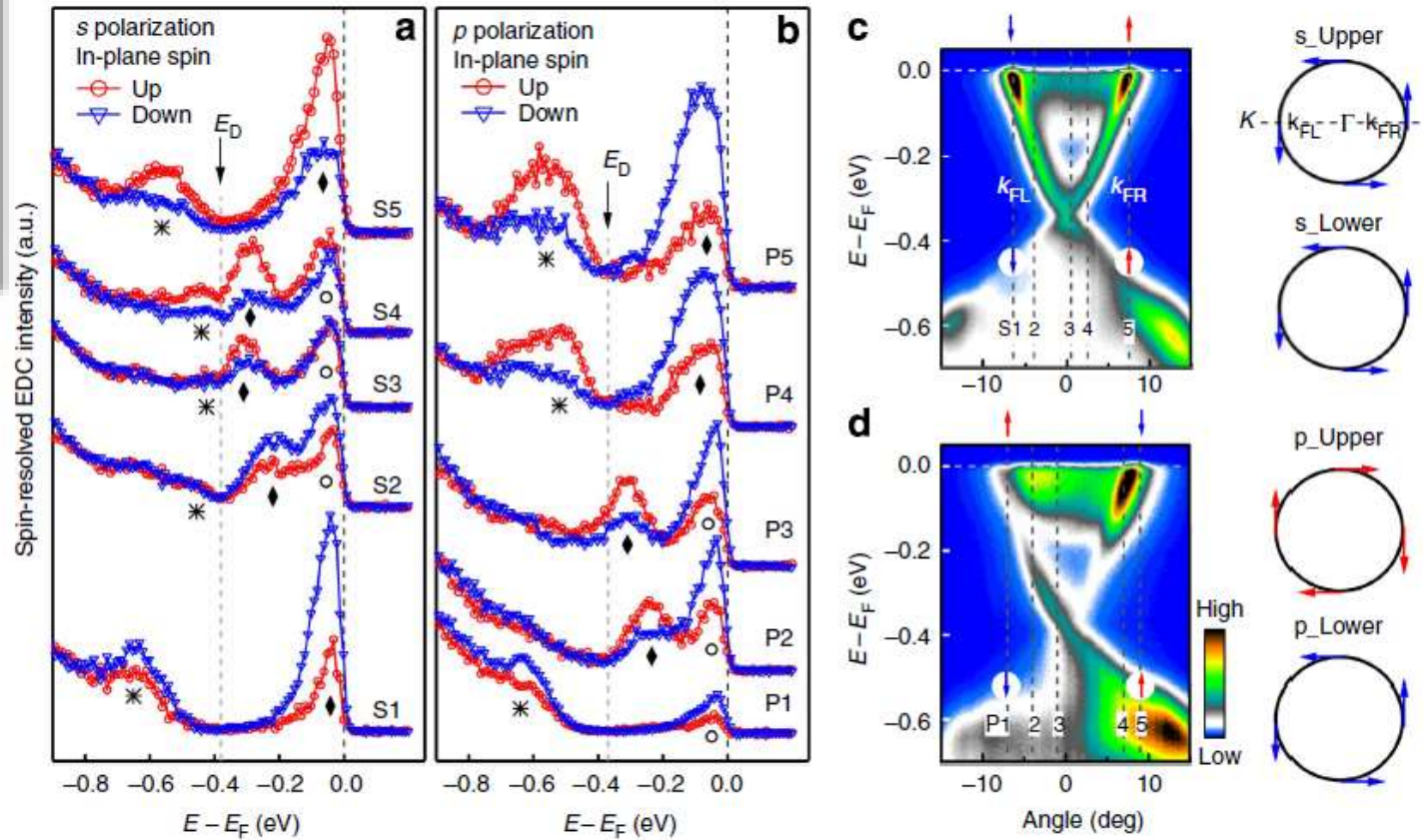
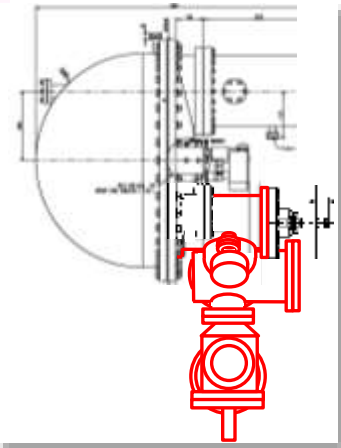
$$A = \frac{\overline{C1} - \overline{C2}}{\overline{C1} + \overline{C2}} = -0.163$$

$$P = \frac{A}{S} = -78.9 \pm 8.1\%$$

Understand the spin of topological surface electrons

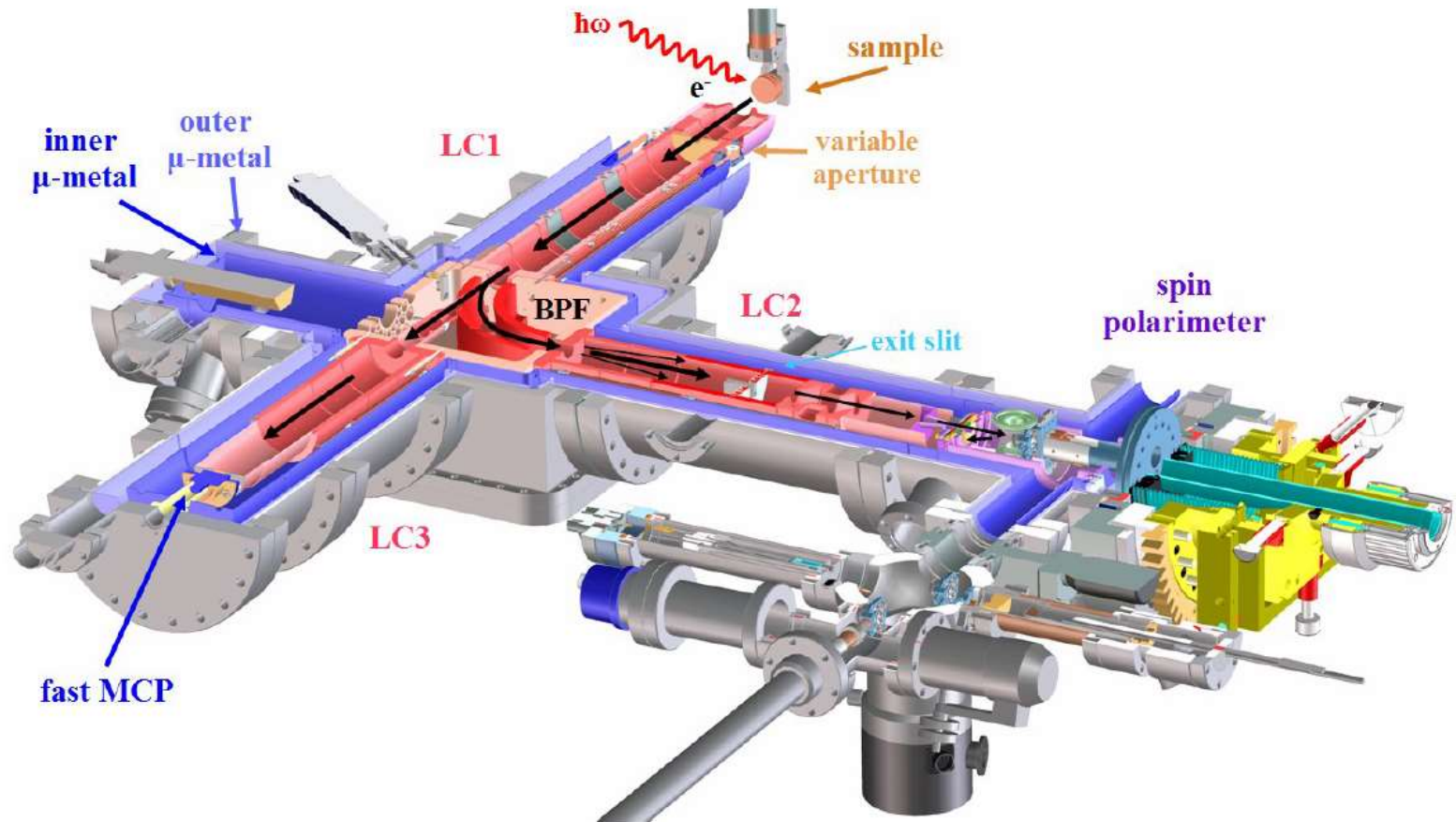
Z. Xie, et. al., *Nature Comm* 5:3382 (2014)

0D Spin detection



Understand the spin of topological surface electrons

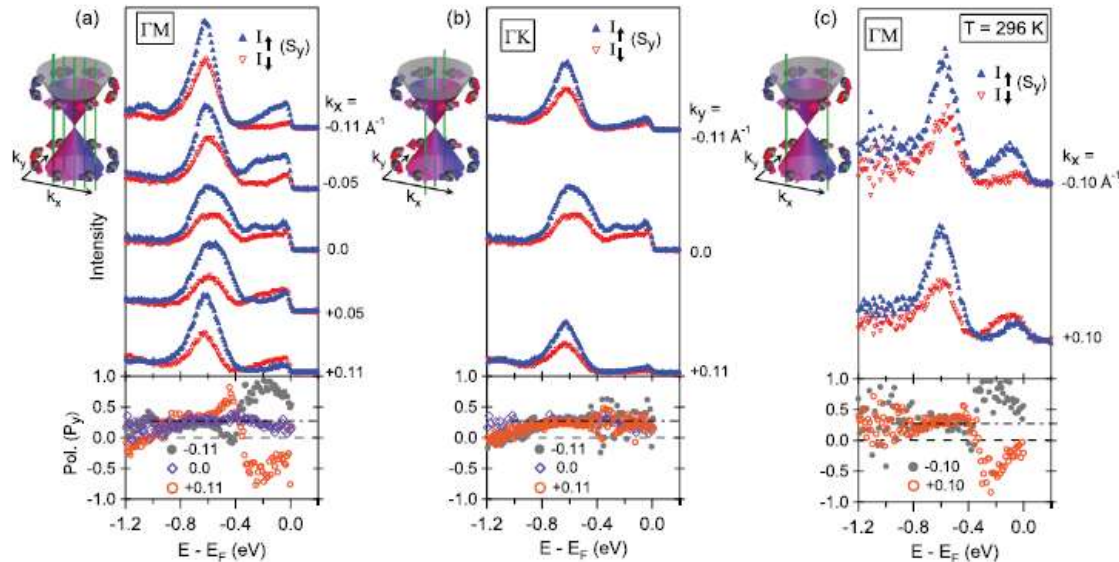
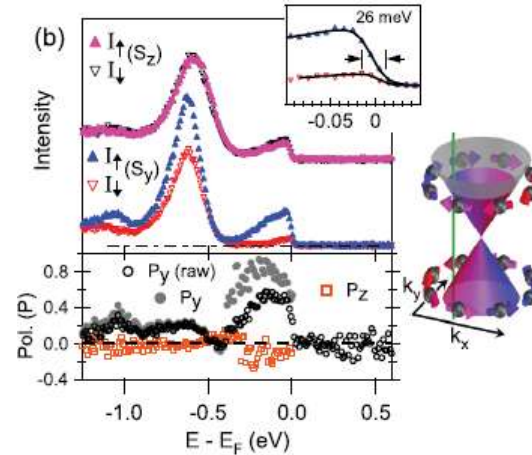
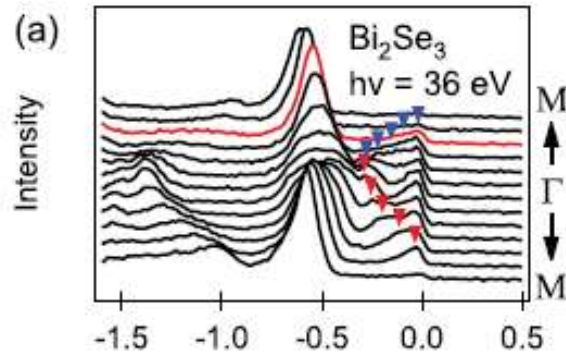
1D Spin detection



Understand the spin of topological surface electrons

C. Jozwiak, et. al., *Phys. Rev. B*. 84, 165113 (2011)

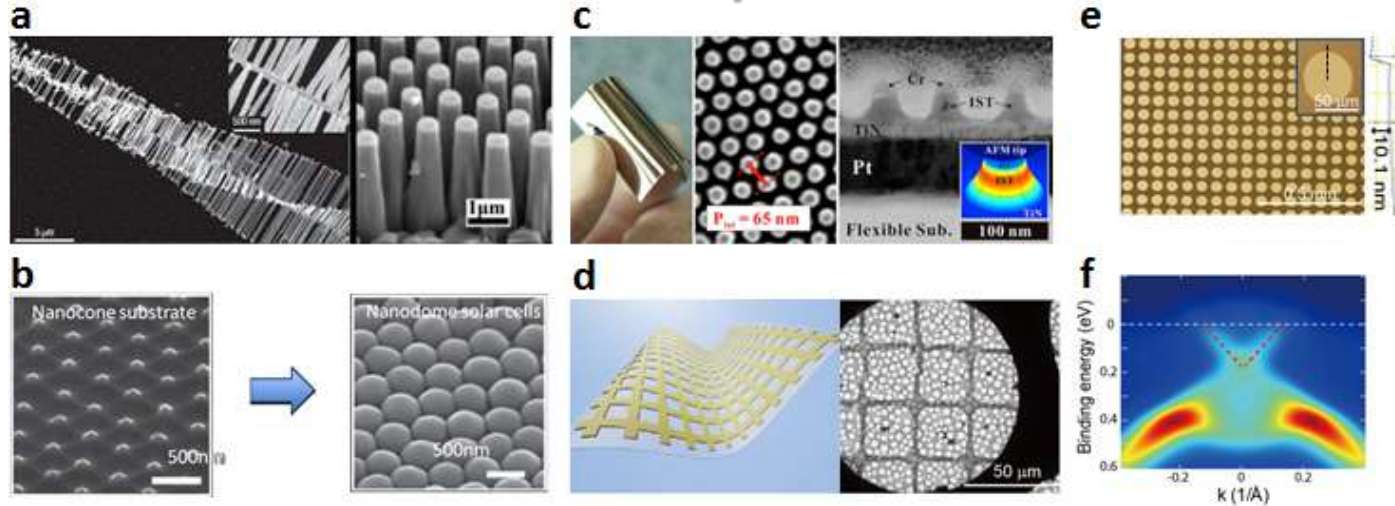
1D Spin detection



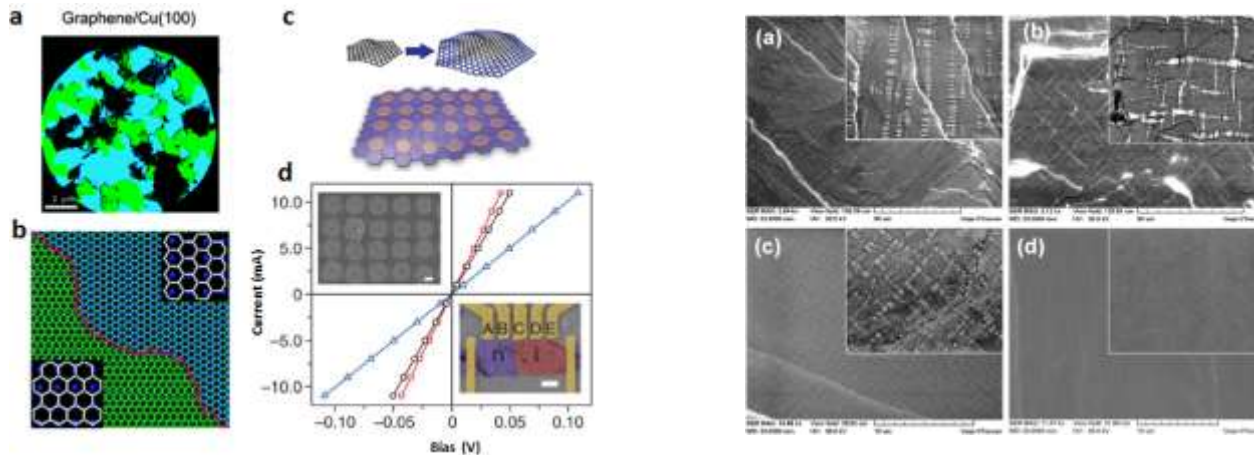
Spatially resolved ARPES

Explore electronic structure with spatial resolution

New mesoscopic materials



Materials with local compositional inhomogeneity

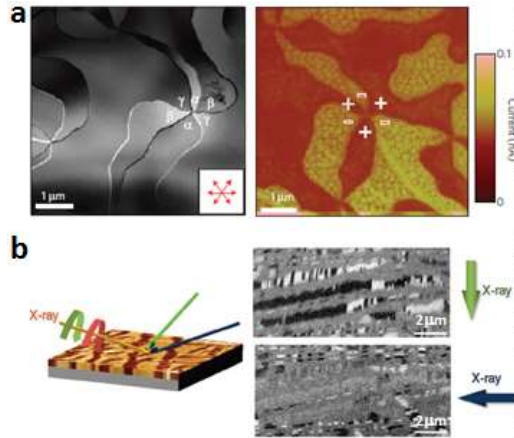


Graphene p-n junction

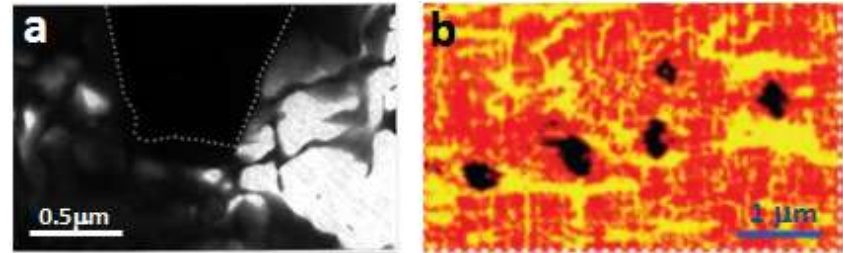
Fe-based superconductor

Explore electronic structure with spatial resolution

Materials with domains

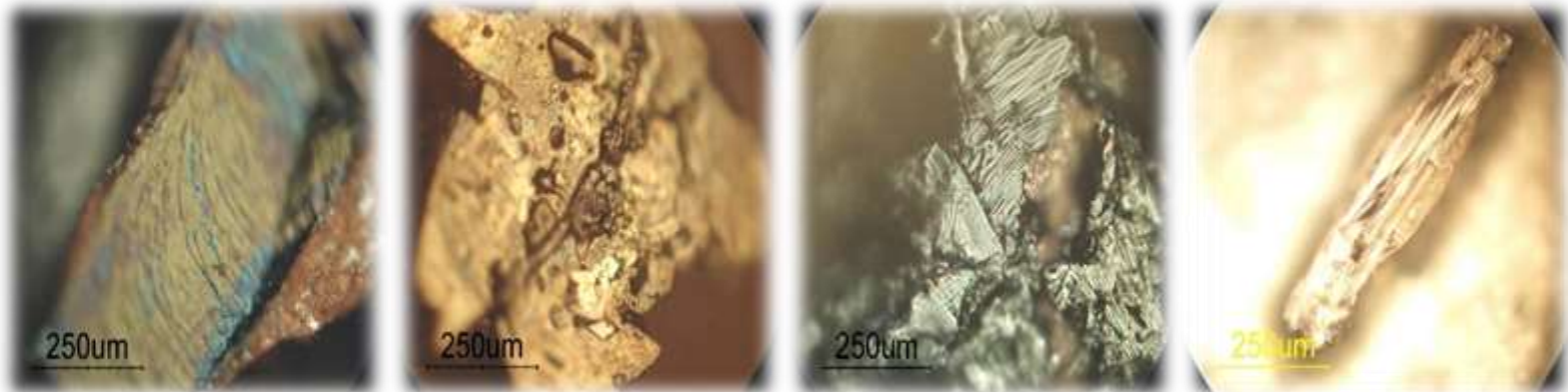


Multi-ferroic materials



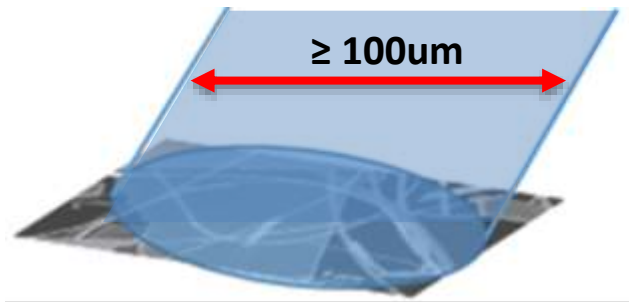
CMR materials

Unfriendly sample surface for traditional ARPES

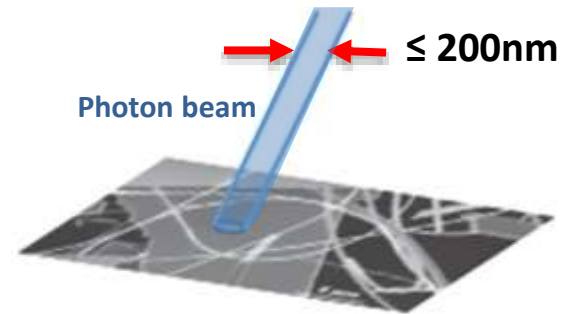


How to achieve nm scale spatial resolution

Regular ARPES



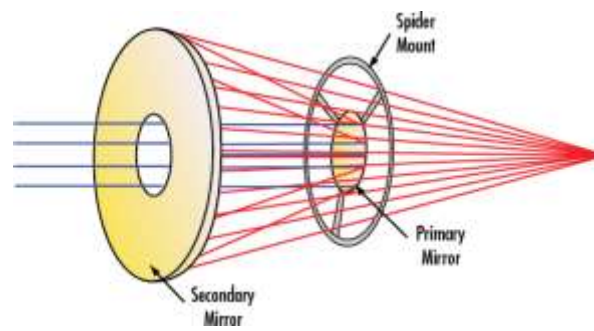
Spatially-resolved ARPES



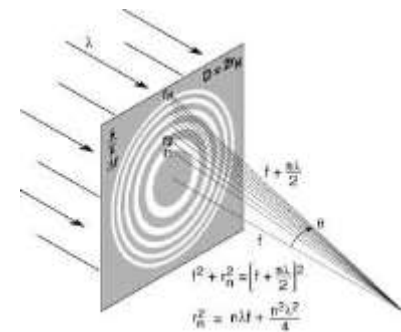
Focus with lens



Schwarzschild objective

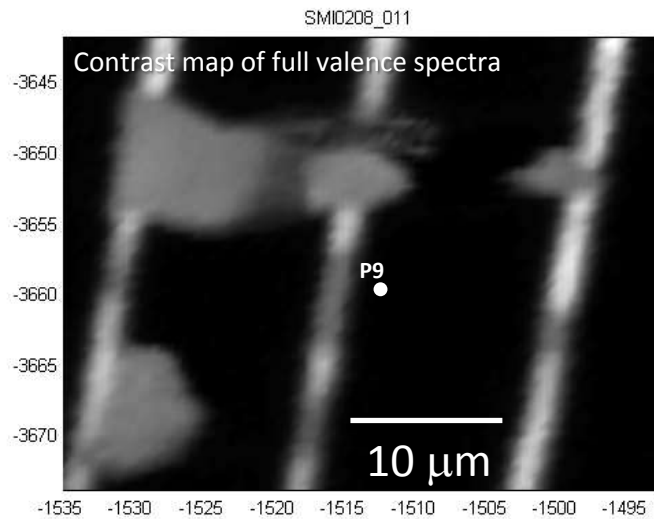


Zone plate



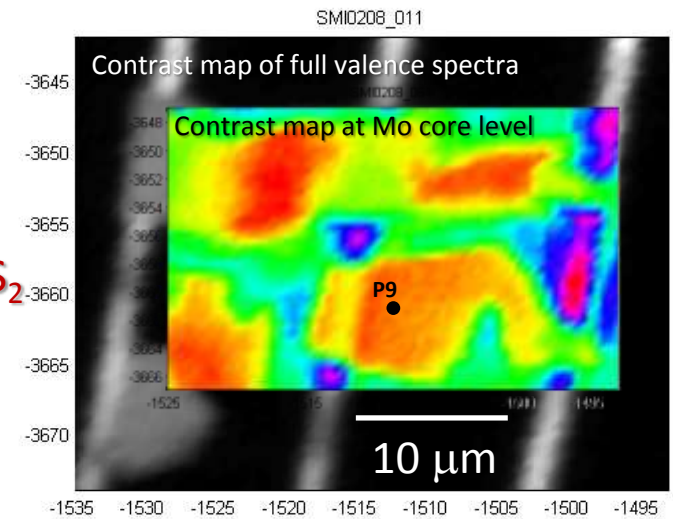
$\lambda_{100\text{eV}} \sim 13\text{nm}$

Preliminary study – element enhanced mapping

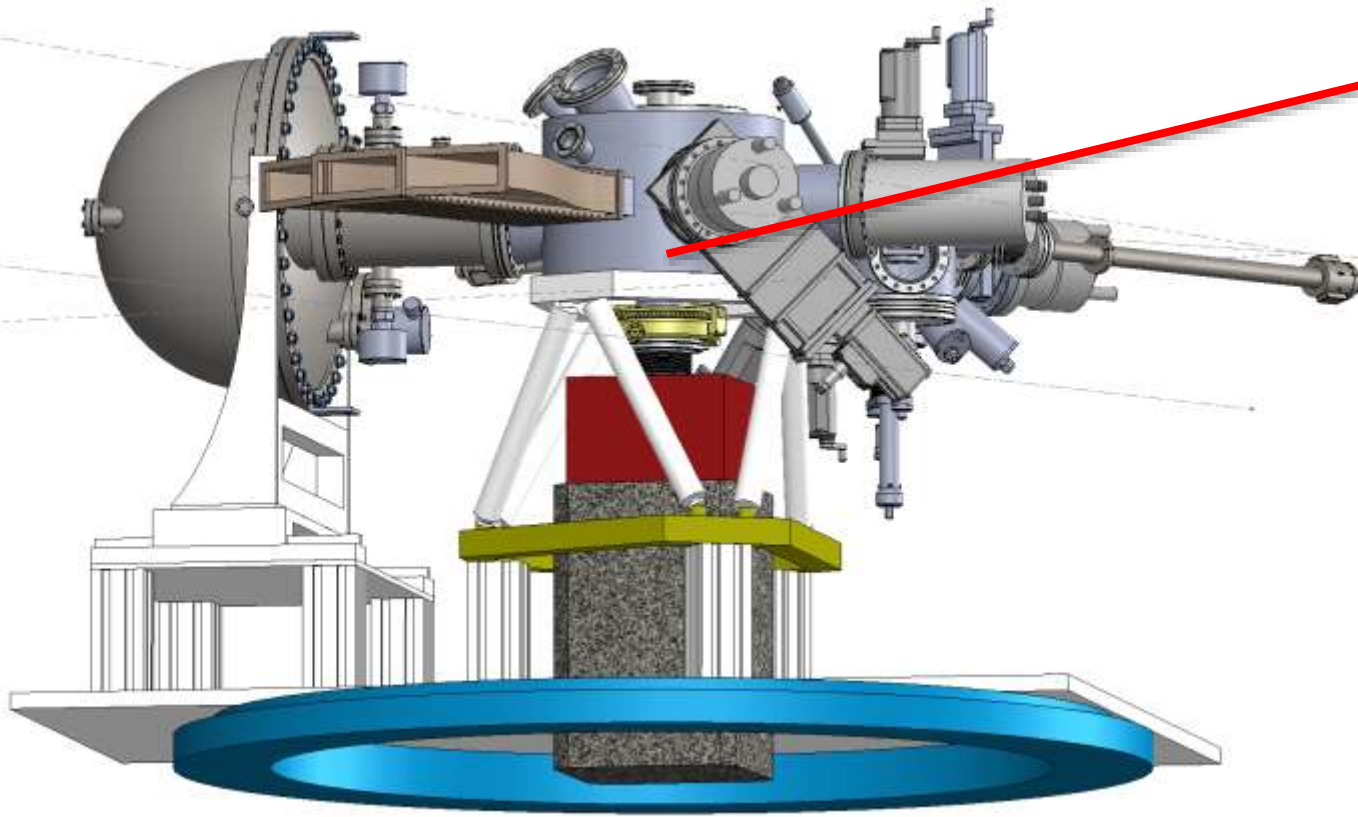


Core level enhanced
contrast image

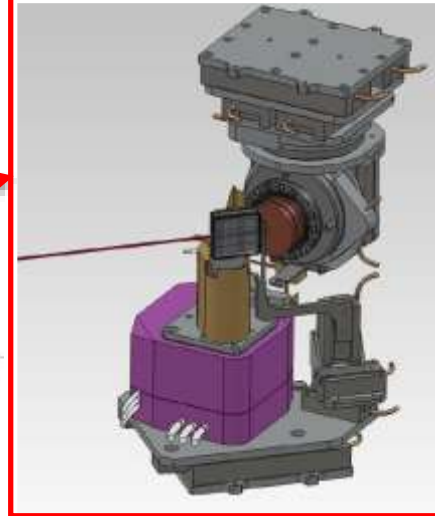
Reveals single layer MoS_2



Conceptual design

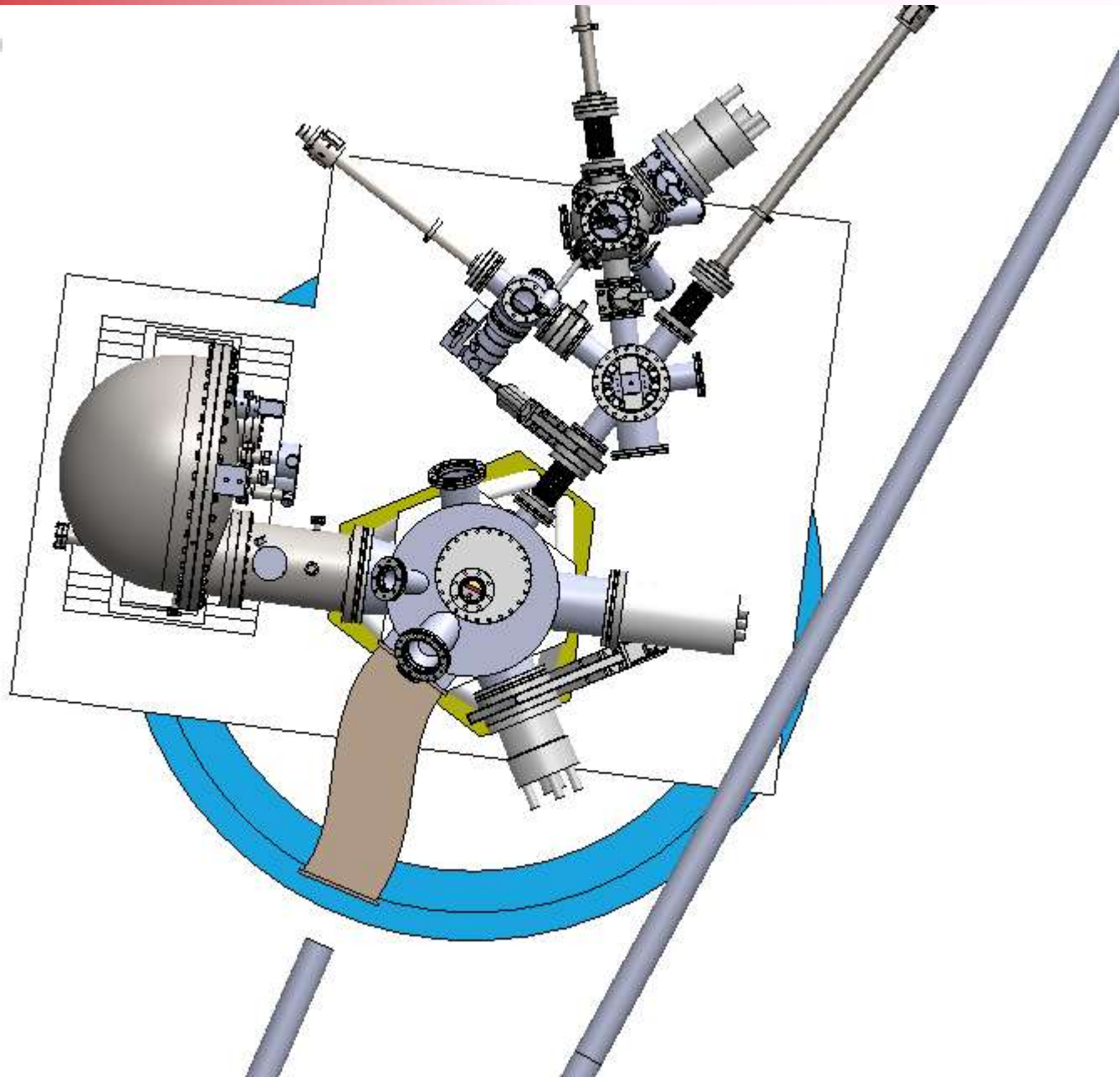


**Sample manipulator
& Zone plate holder**



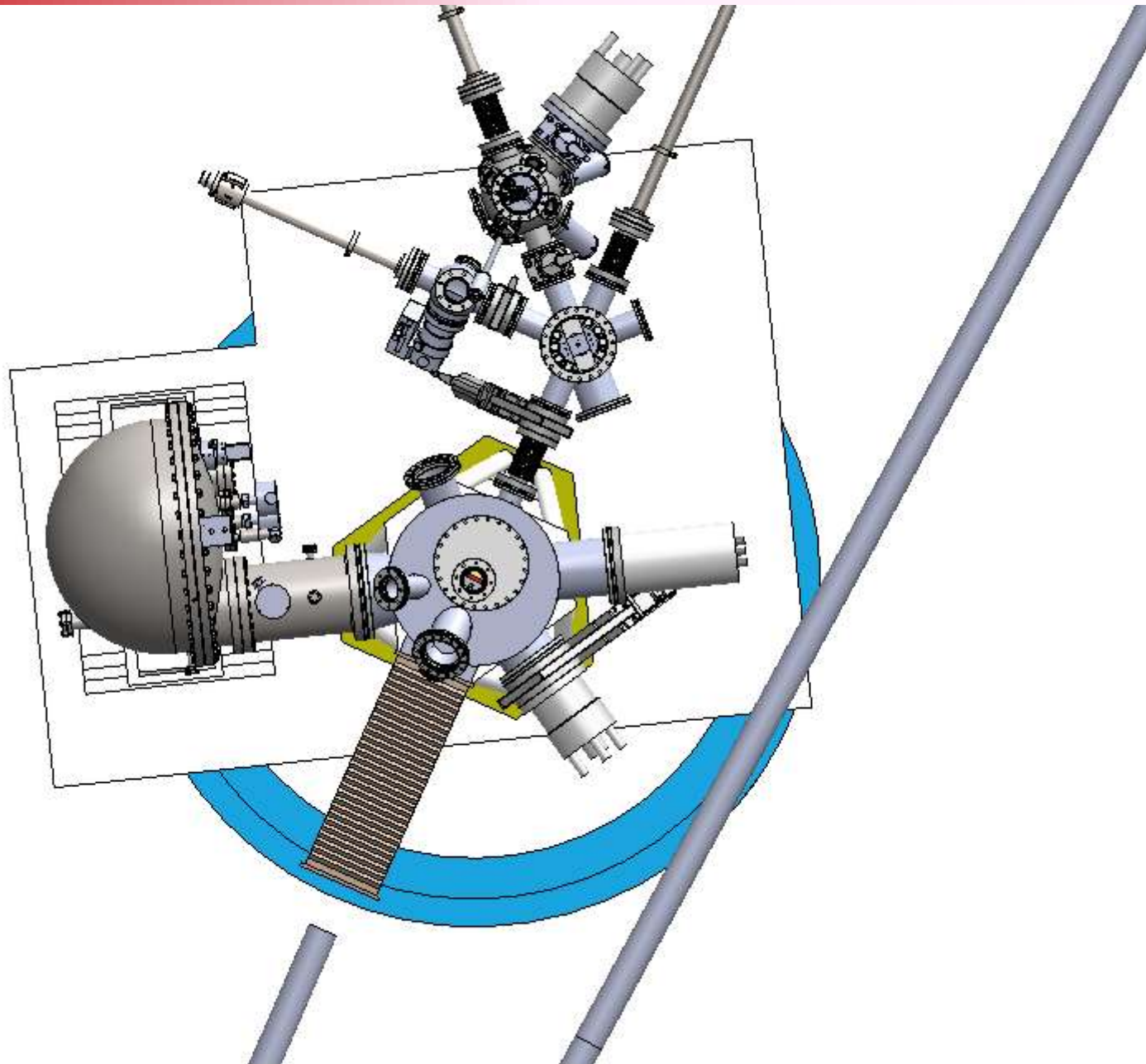
Conceptual design

$$\theta = -15^\circ$$



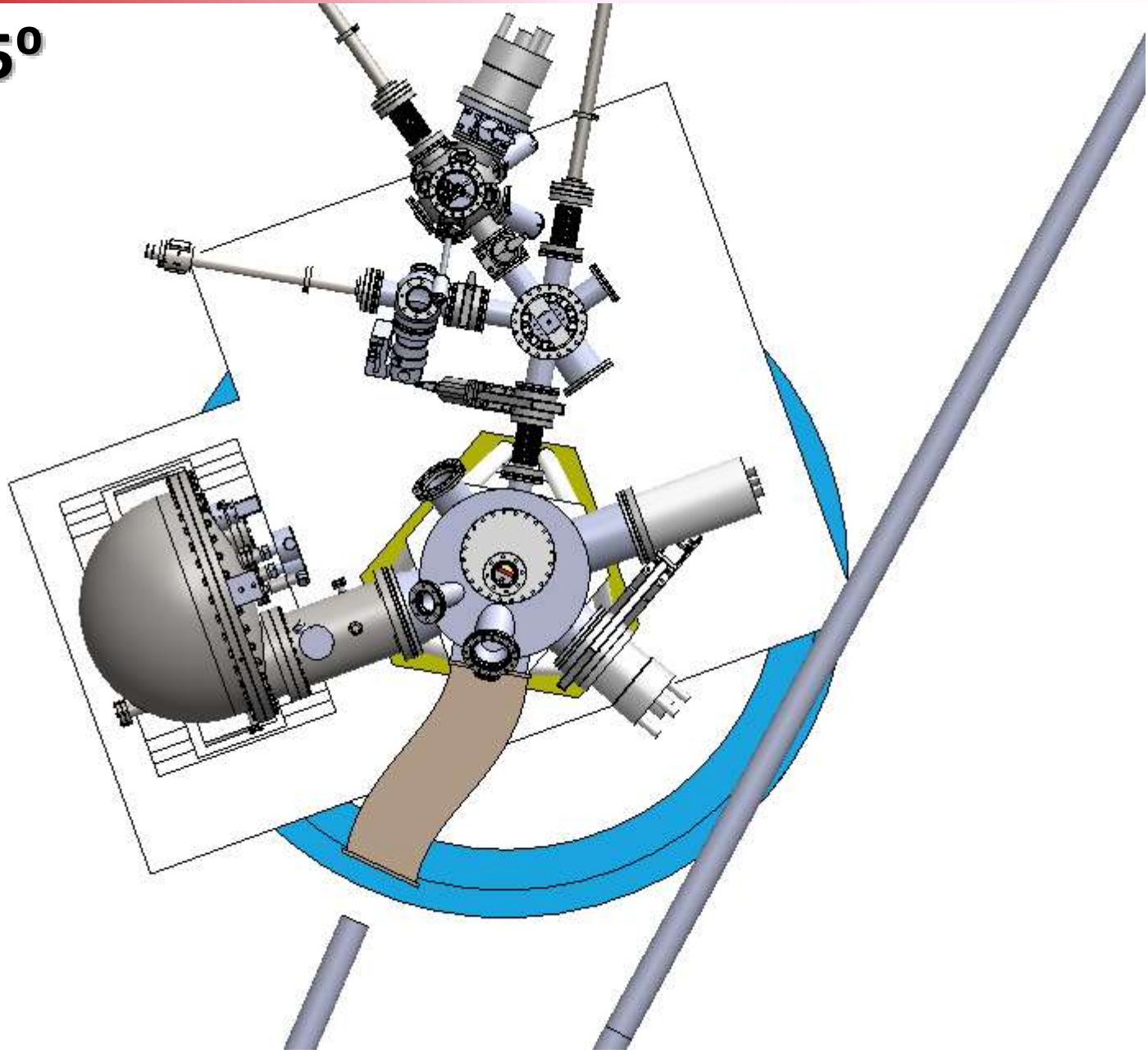
Conceptual design

$$\theta = 0^\circ$$



Conceptual design

$$\theta = 15^\circ$$



The image shows the entrance gate of a park. The gate is a large, modern structure with a curved top and is flanked by two green, triangular-shaped structures. The text 'Thank you!' is overlaid in large, bold, red letters across the center of the gate. The background features a paved path leading into a lush green area with many trees. To the left of the gate, there are several flagpoles with various flags. The overall scene is bright and sunny, suggesting a pleasant day at the park.

Thank you!