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Role of matrix elements in observation of valley-selective linear dichroism in bilayer MoS_2

Charlotte Sanders

Phys. Rev. B **100** (2019) 241406(R); *Phys. Rev. B* **100** (2019) 235423

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Elettra Sincrotrone, SuperESCA

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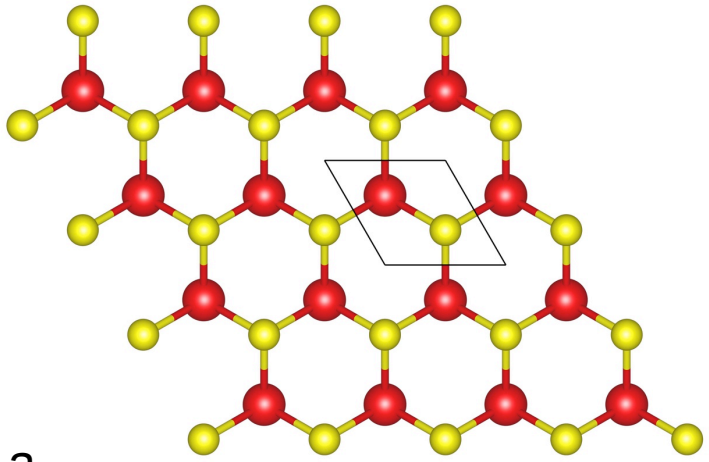
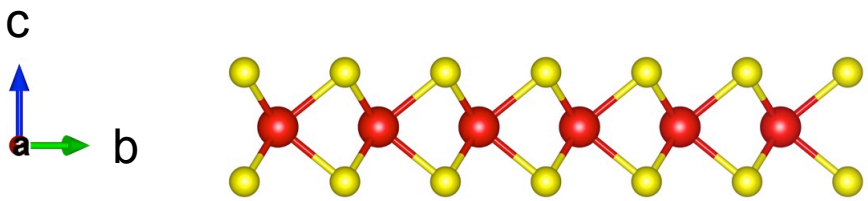
MoS_2 trigonal prismatic structure

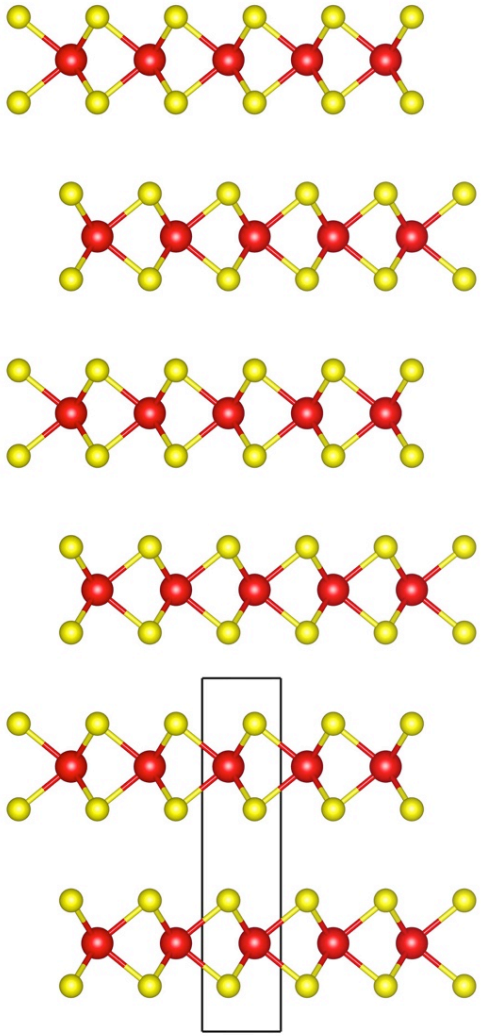
a = 3.15 Å



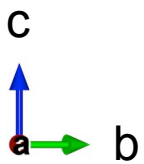
Single-layer "1H"



Bulk 2H



a = 3.15 Å
c = 12.3 Å



Mo

S

van-der-Waals bonded layers

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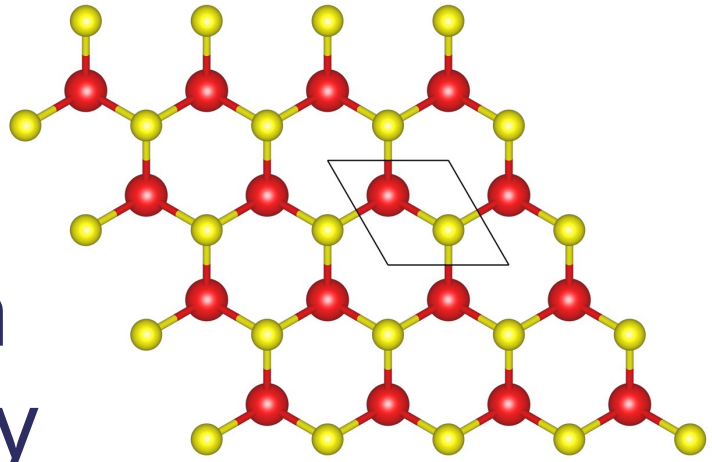
MoS_2 trigonal prismatic structure

Inversion symmetry lifted



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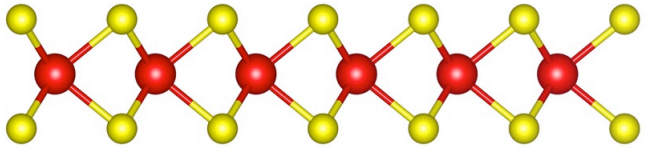

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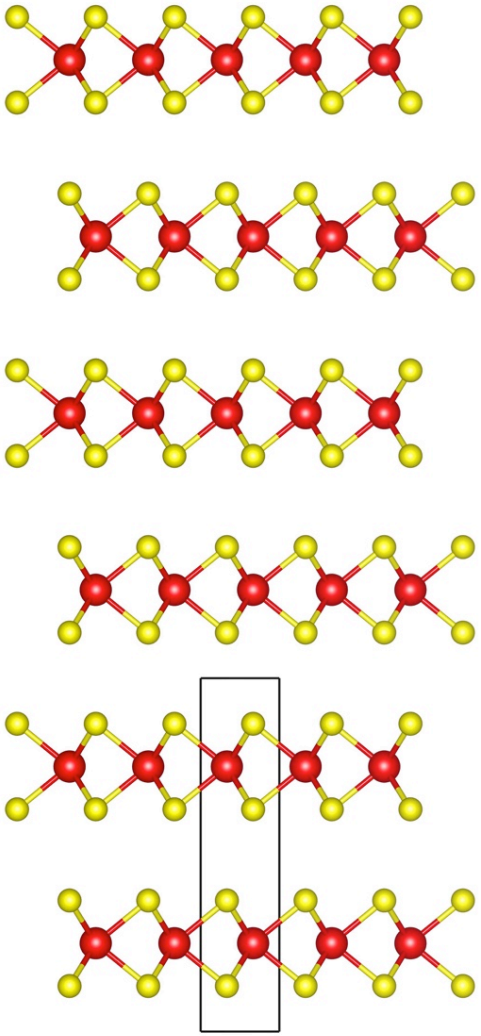

Mo

S

Inversion symmetric

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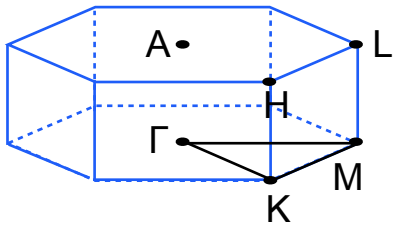
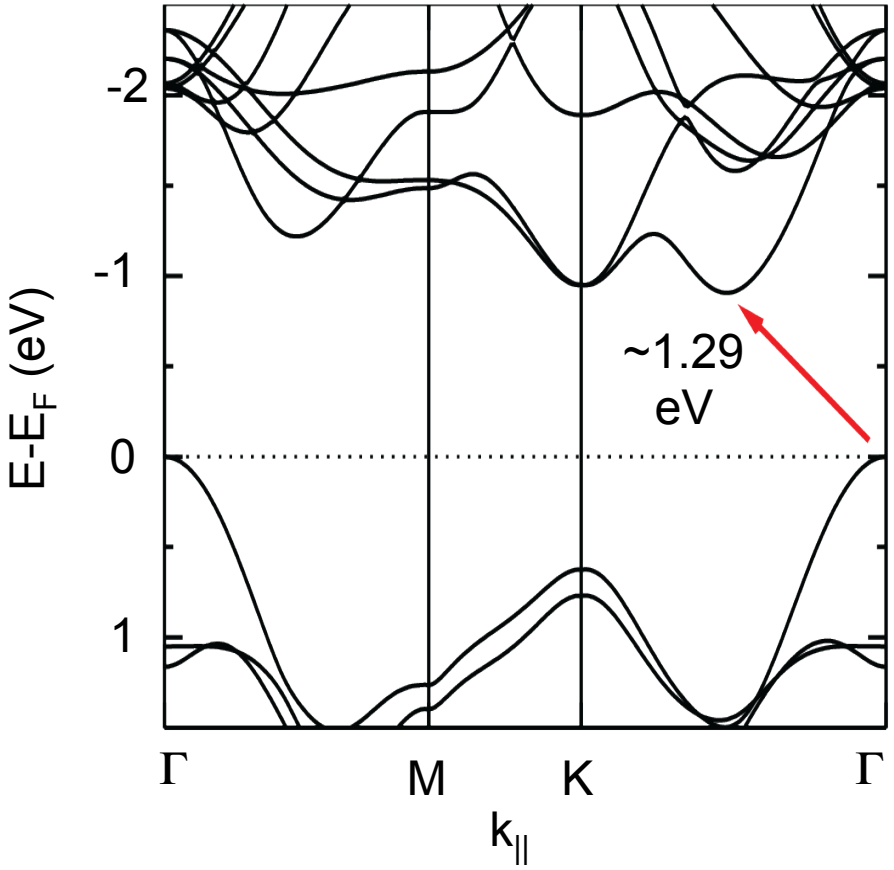
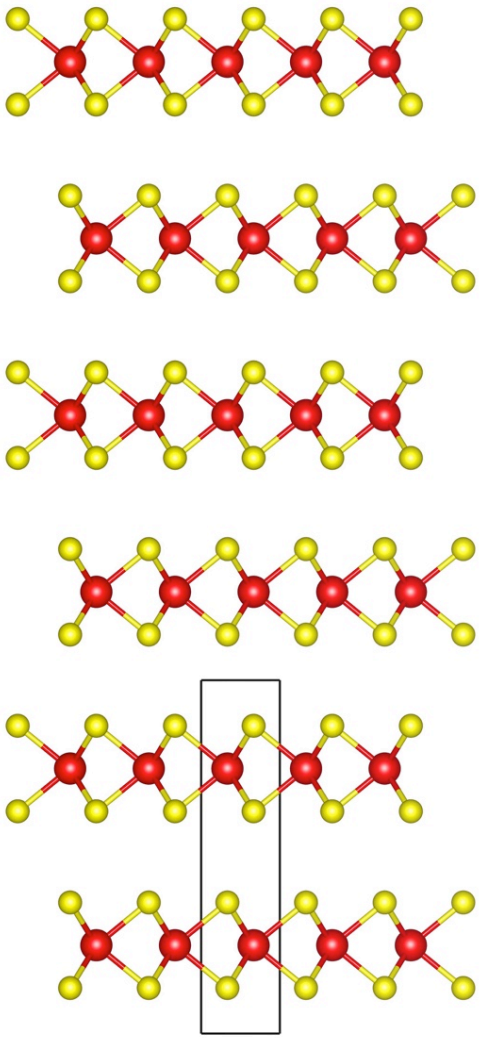
Bulk dispersion







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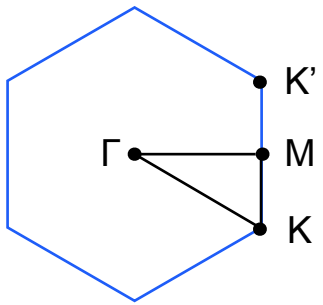
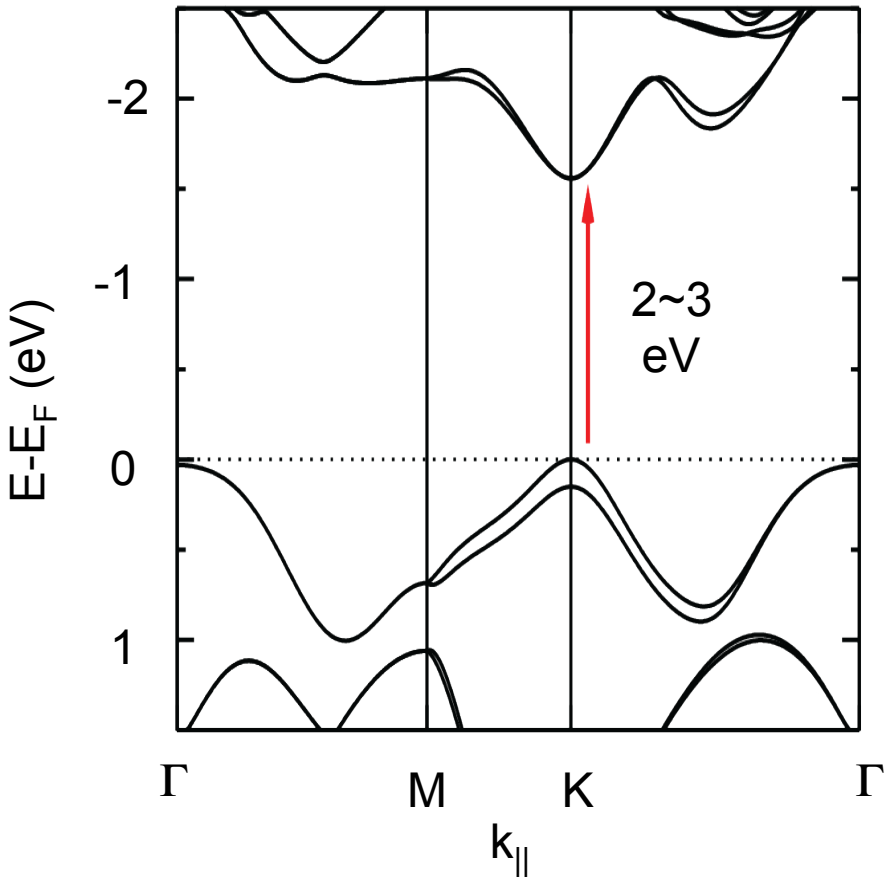
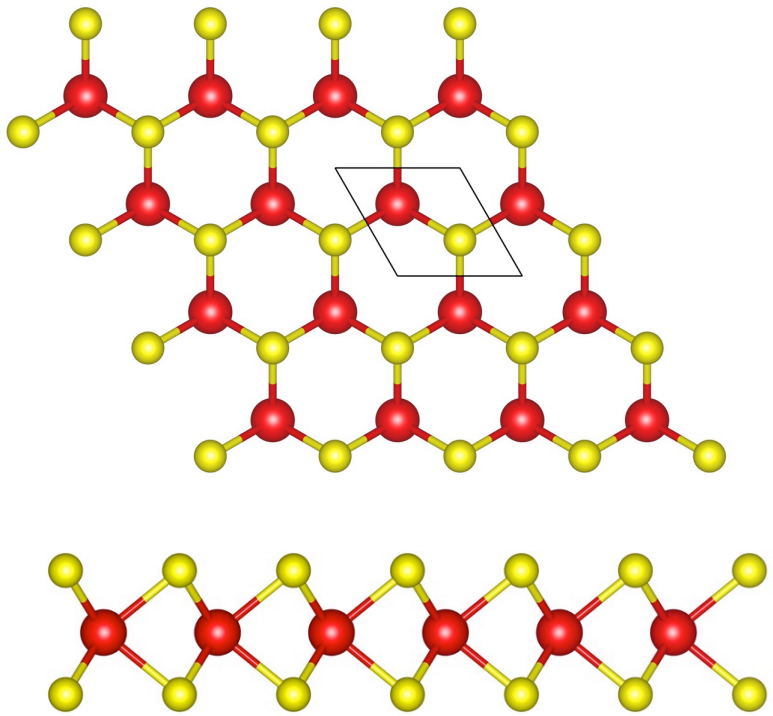
Single-layer dispersion







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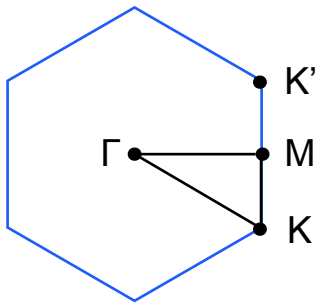
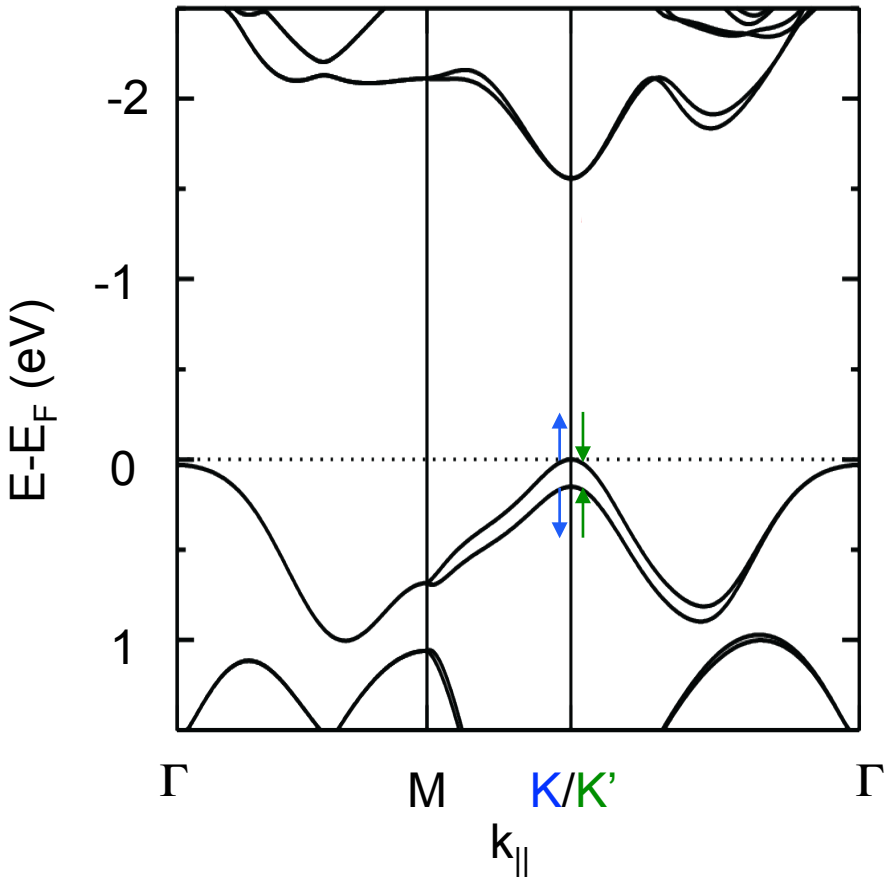
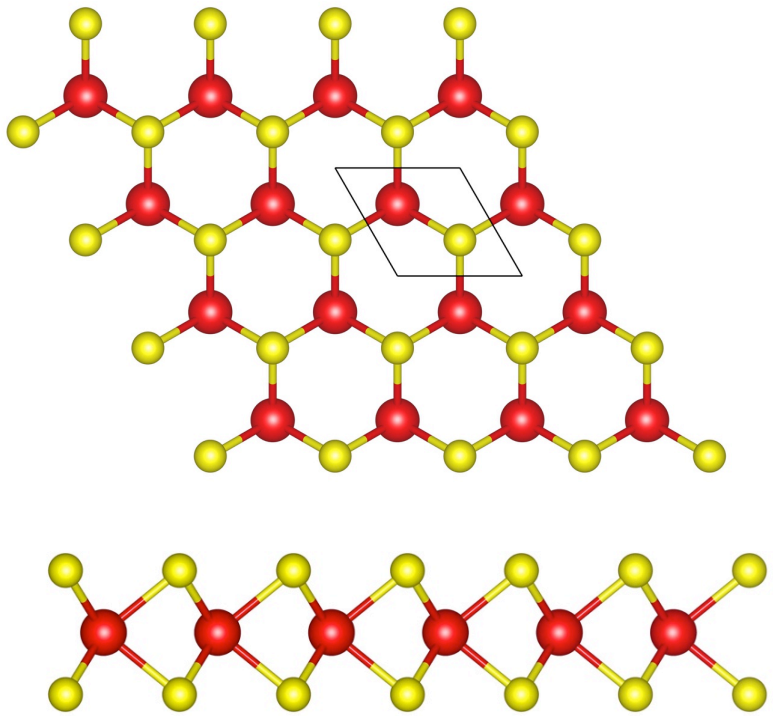
Single-layer dispersion







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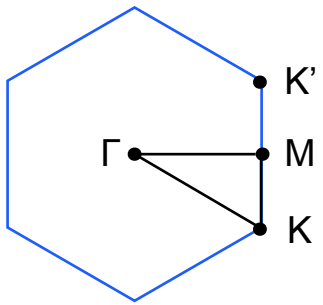
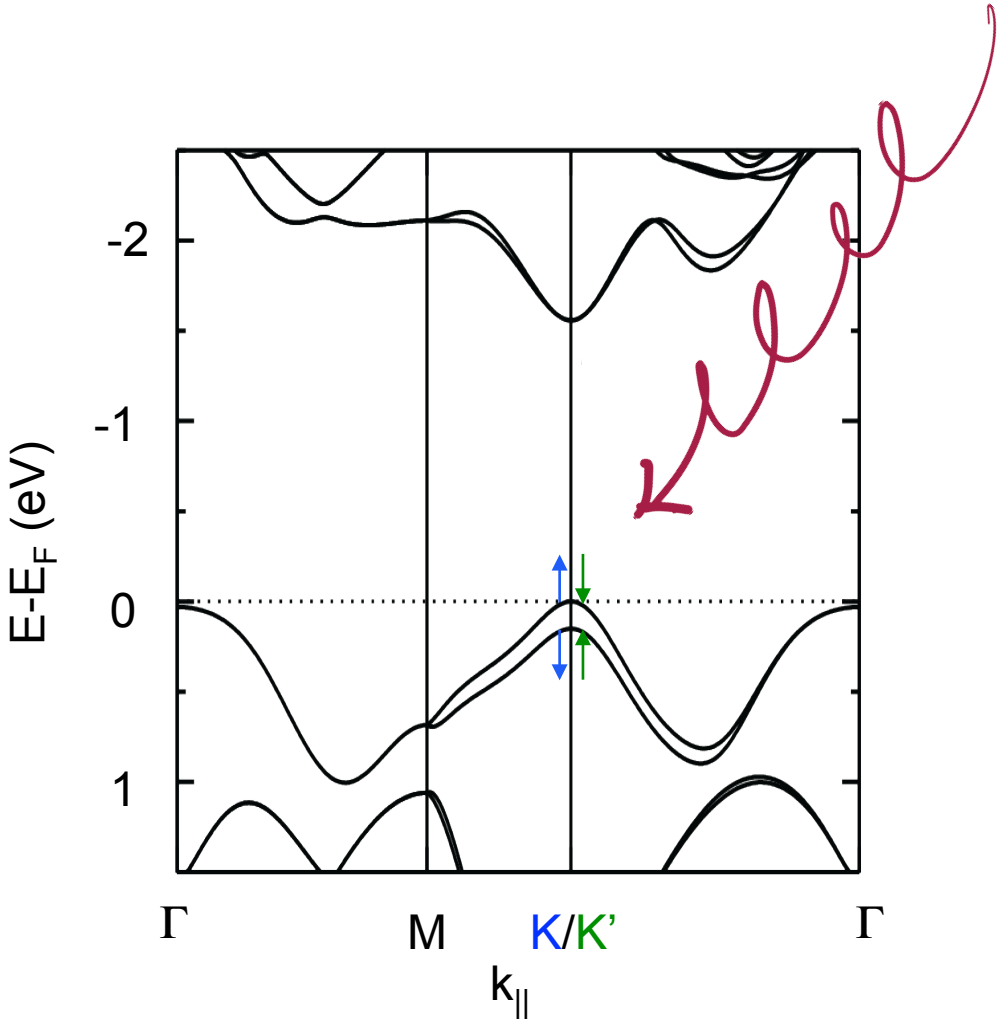
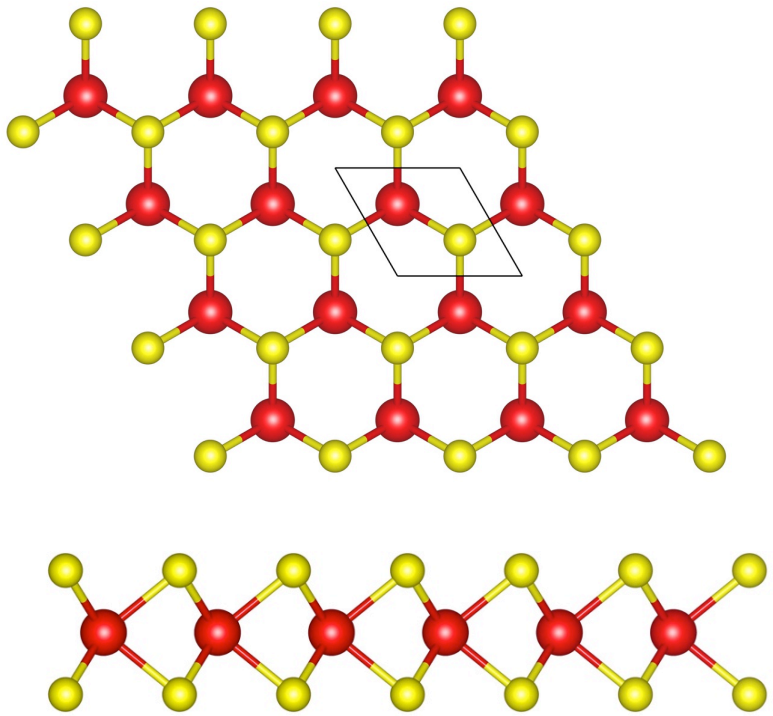
Single-layer: circular dichroism







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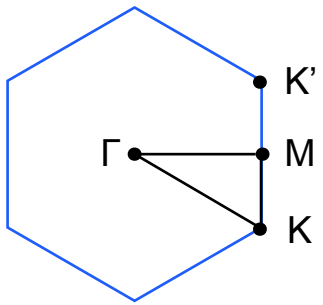
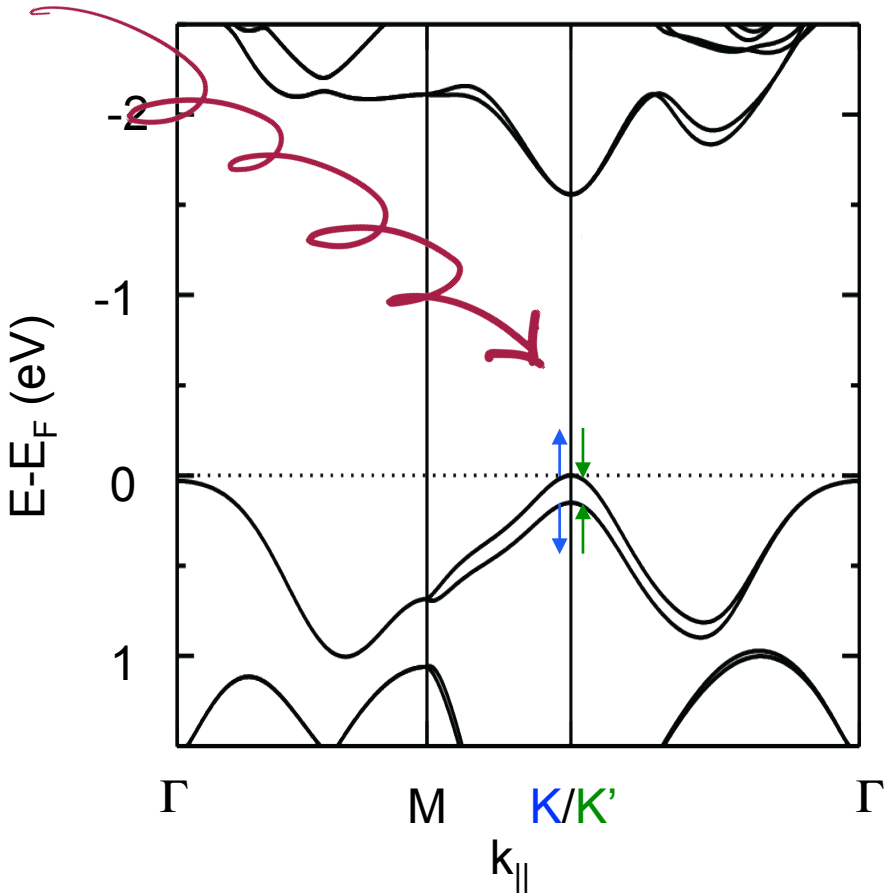
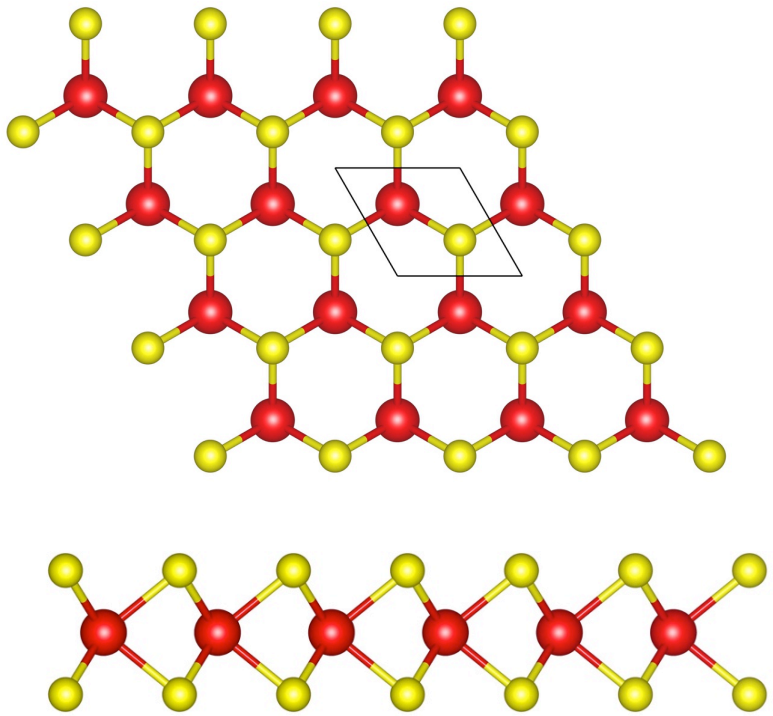
Single-layer circular dichroism







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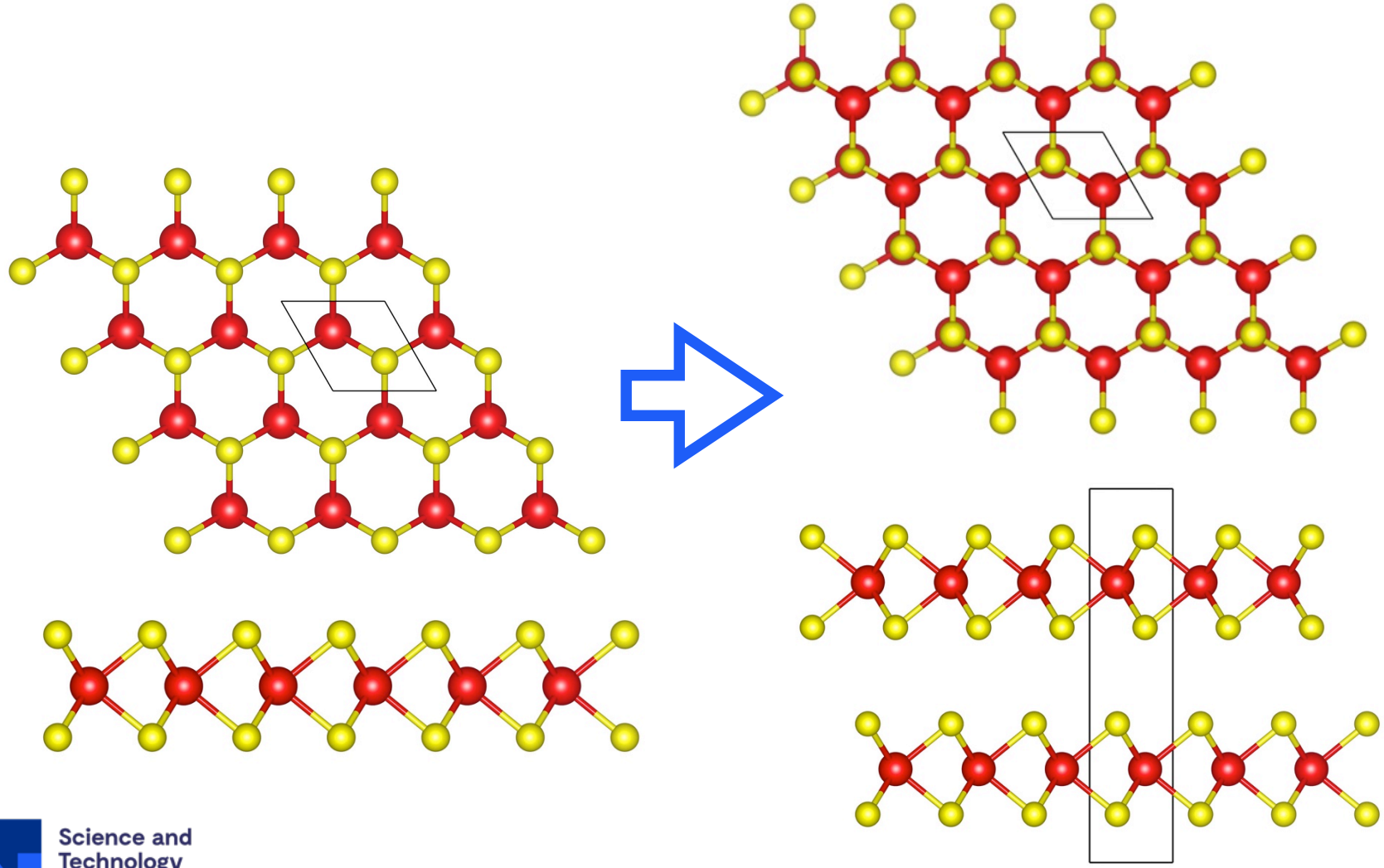
MoS_2 bilayer



Inversion symmetry restored

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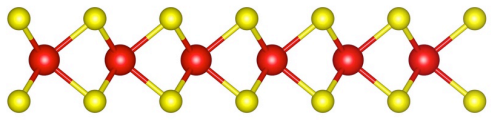
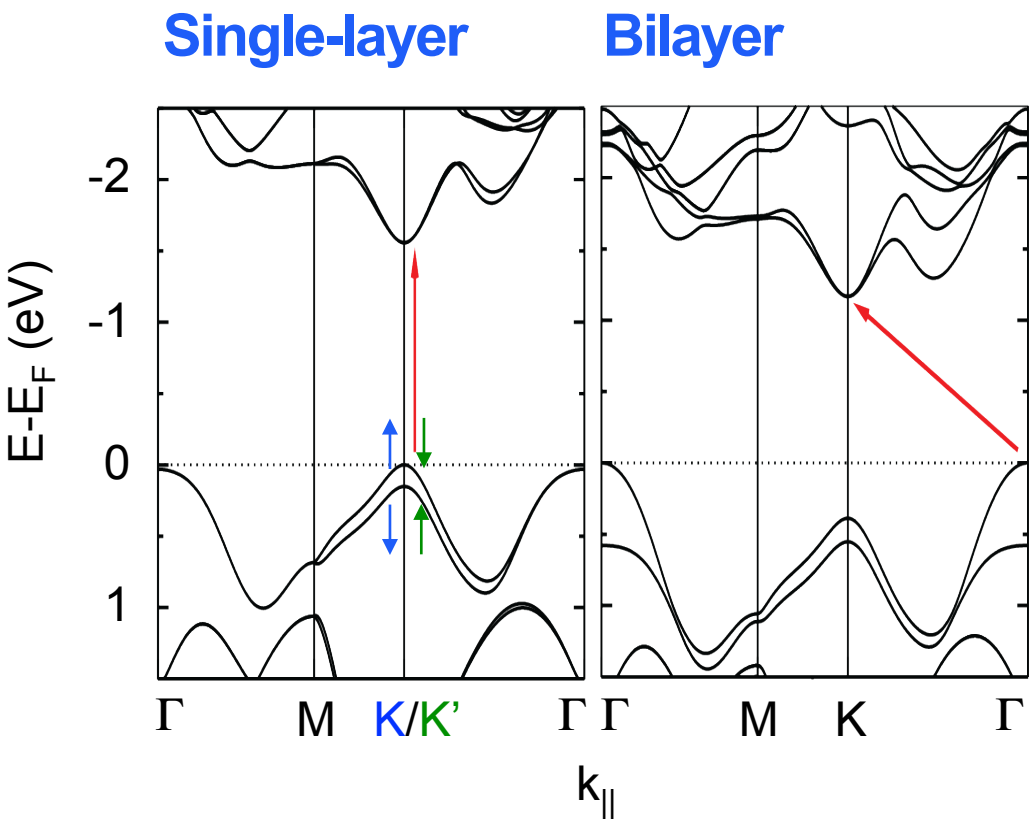
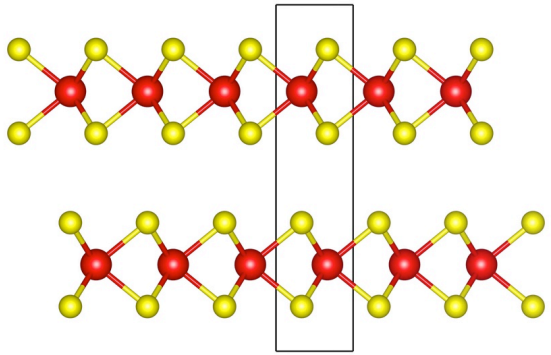
Bilayer dispersion







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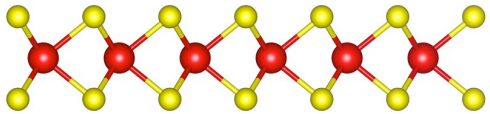
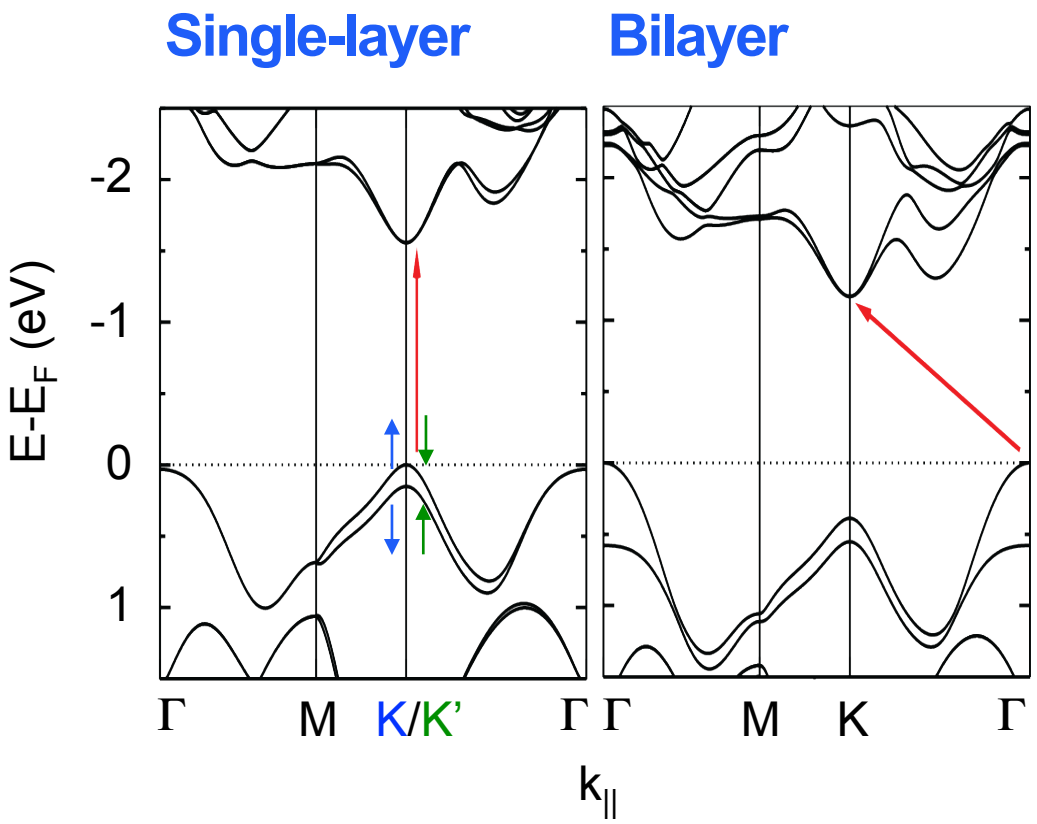
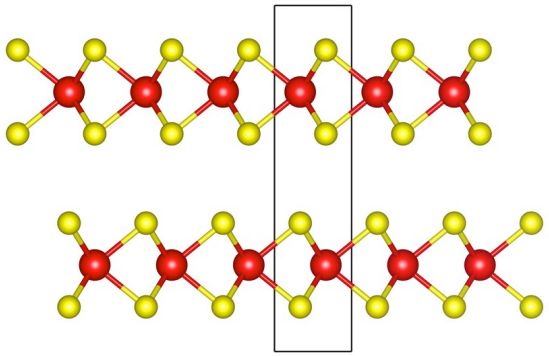
Bilayer dispersion







- Inversion symmetry restored
- Spin polarization lost
- Circular dichroism lost

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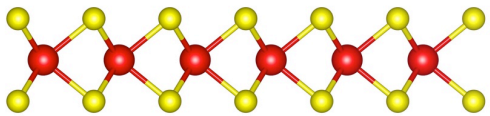
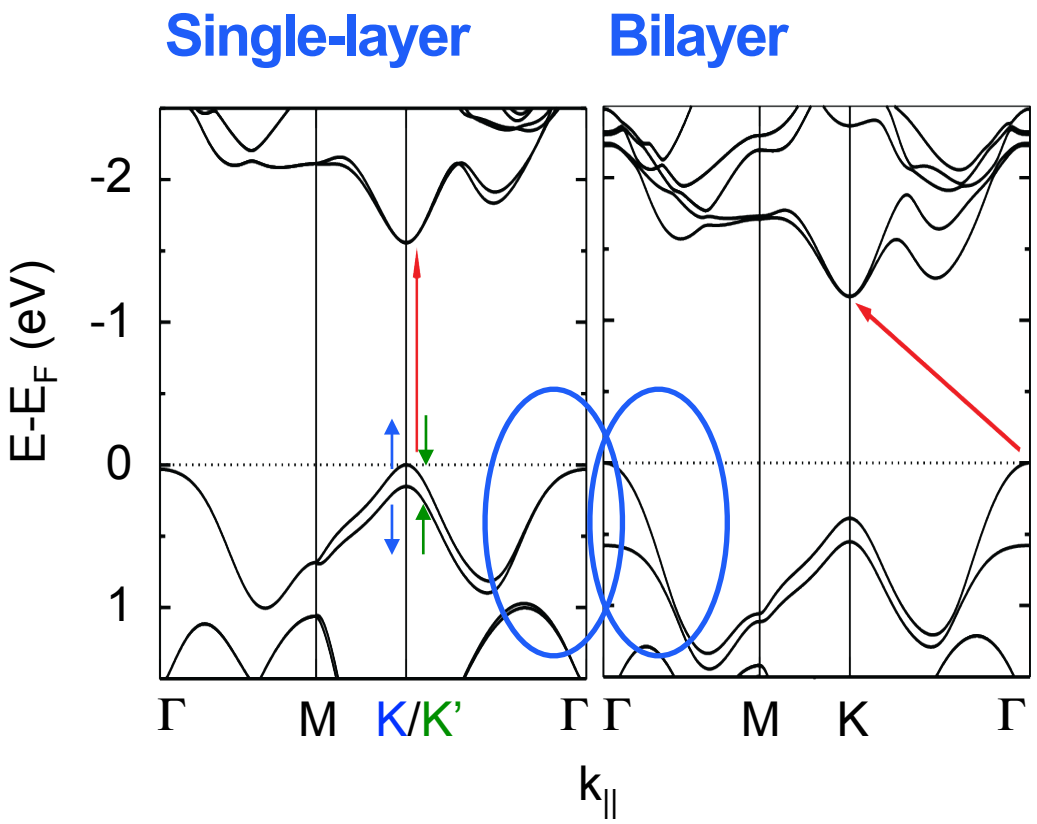
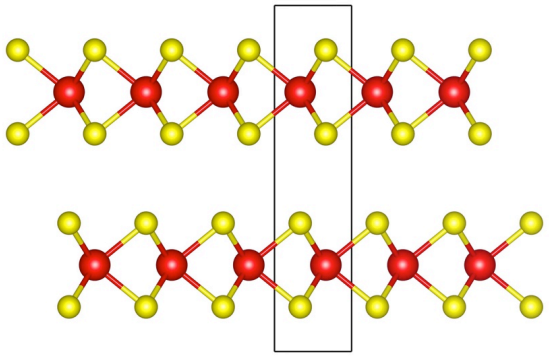
Bilayer dispersion







- Inversion symmetry restored
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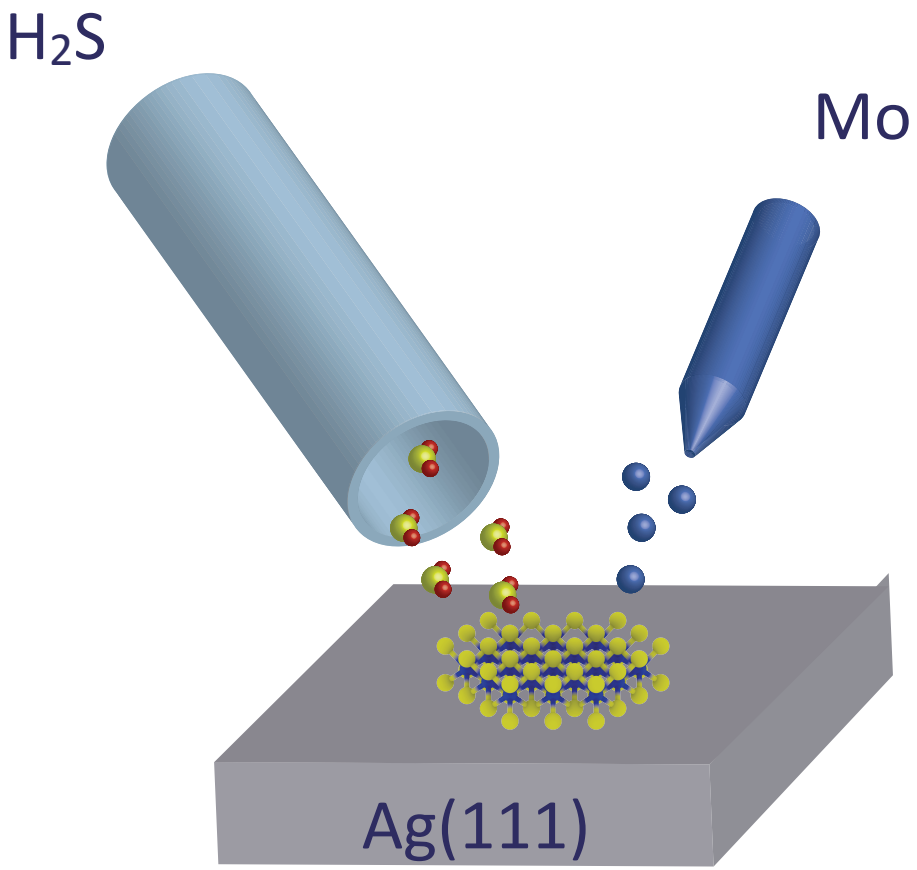
Sample preparation & characterization

Samples: MoS_2/Ag(111)

- Mo evaporation in H_2S atmosphere
- Controlled annealing
- Ultra-high-vacuum base pressure



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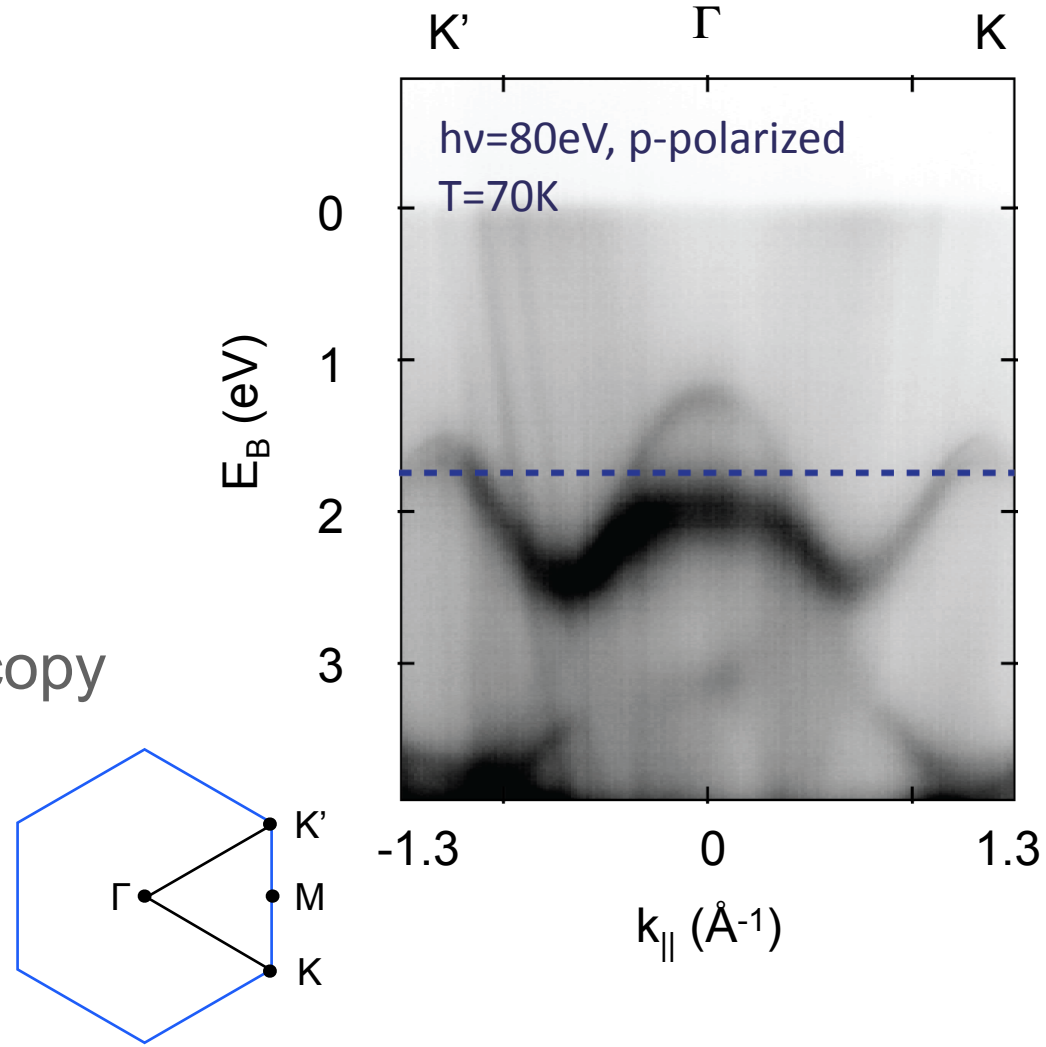
Sample preparation & characterization

Samples: MoS_2/Ag(111)

- Mo evaporation in H_2S atmosphere
- Controlled annealing
- Ultra-high-vacuum base pressure

Preliminary characterization

- X-ray photoelectron diffraction
- Angle-resolved photoemission spectroscopy (ARPES)



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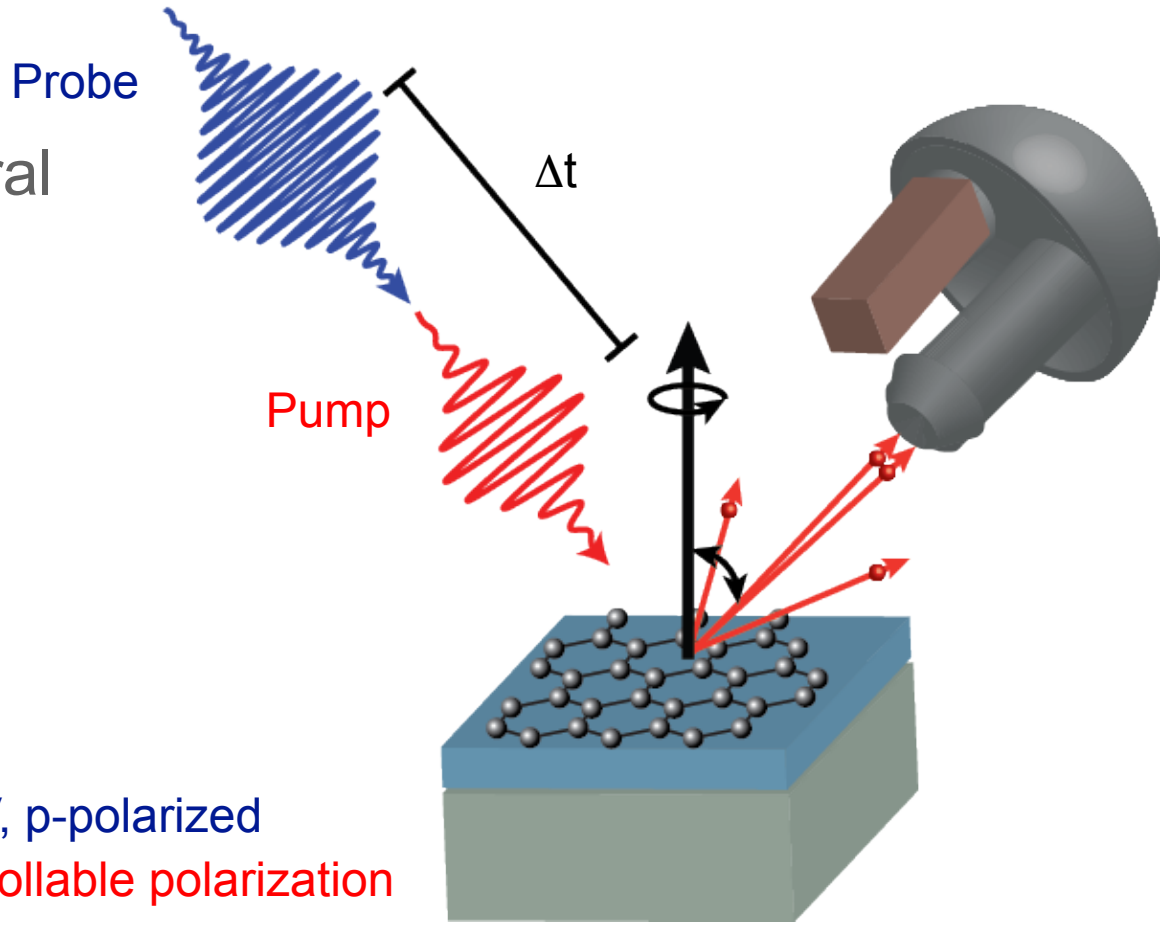
Measurement of excited state

Pump-probe ARPES

- Artemis User Facility (U.K. Central Laser Facility)
- Tunable infrared pump
- Extreme ultraviolet probe generated by high-harmonic generation



Probe: hv=32.5eV, p-polarized

Pump: 2 eV, controllable polarization

central laser facility

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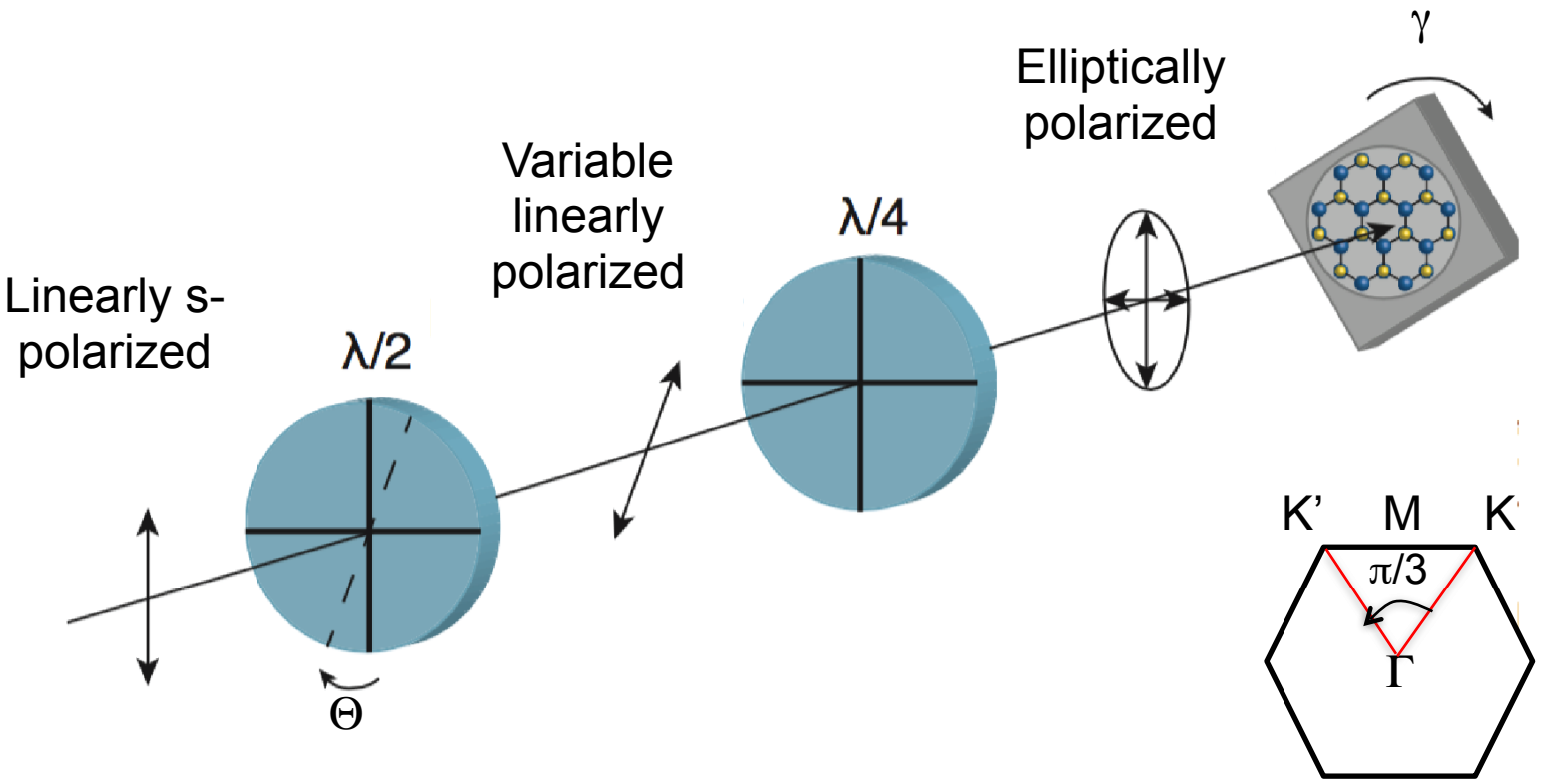
Pump polarization



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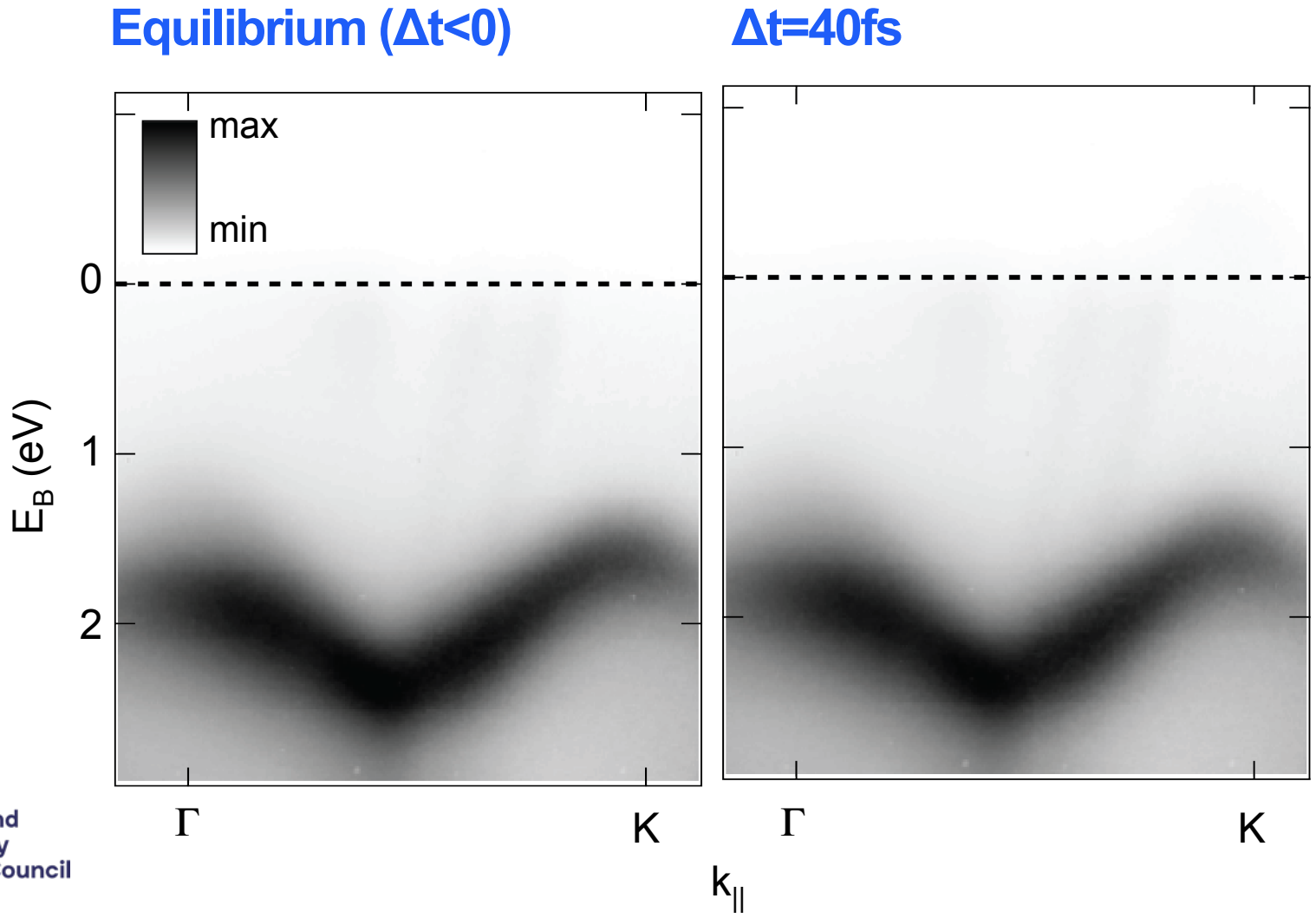
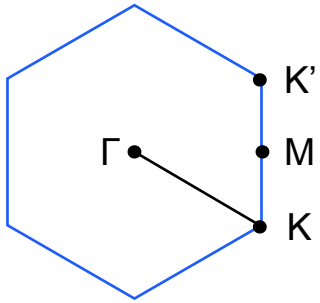
Measurement of excited state





Room temperature

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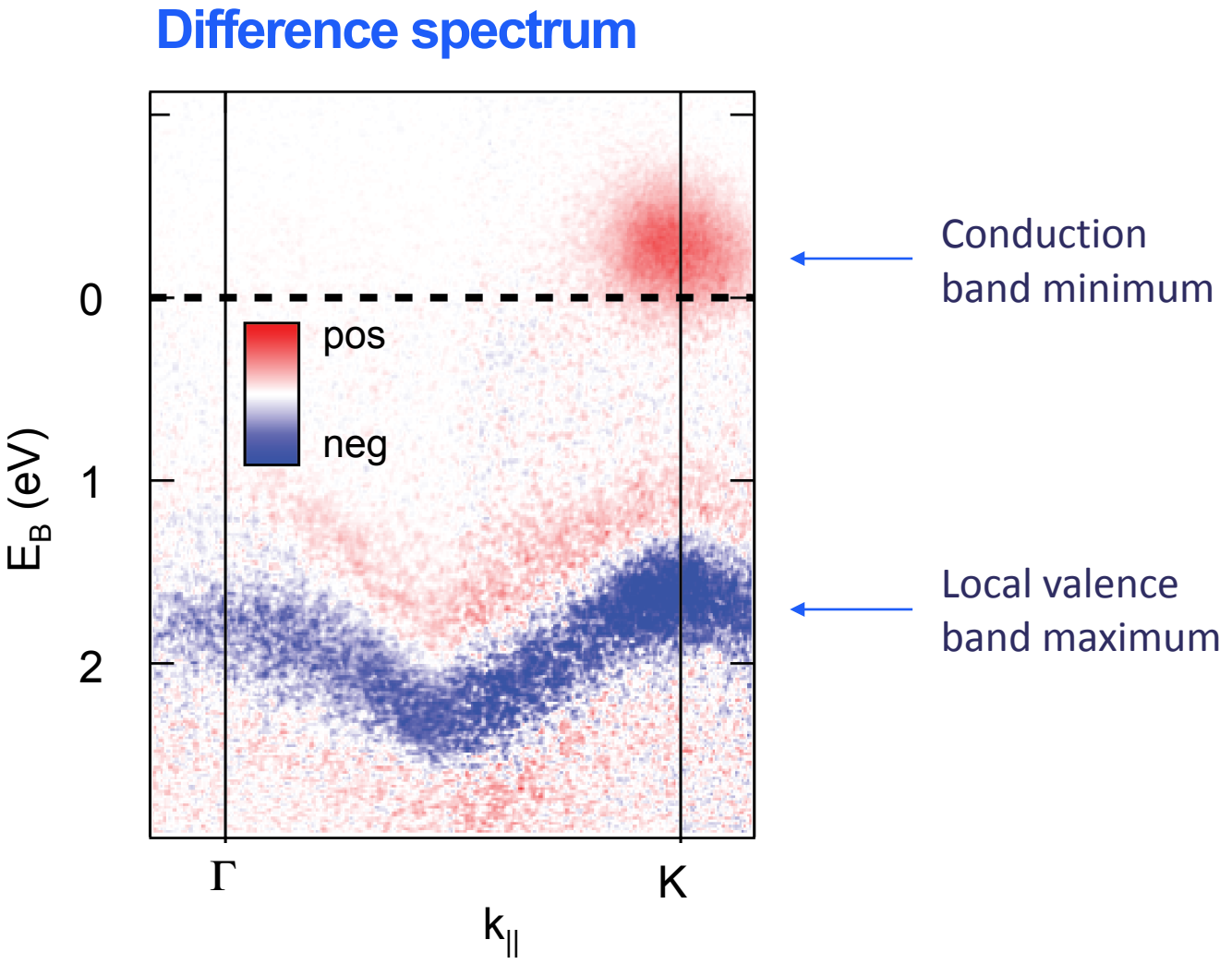
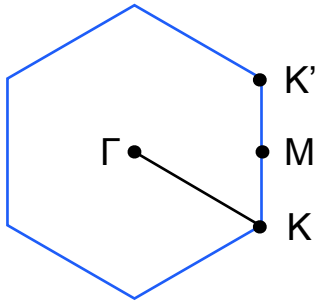
Measurement of excited state





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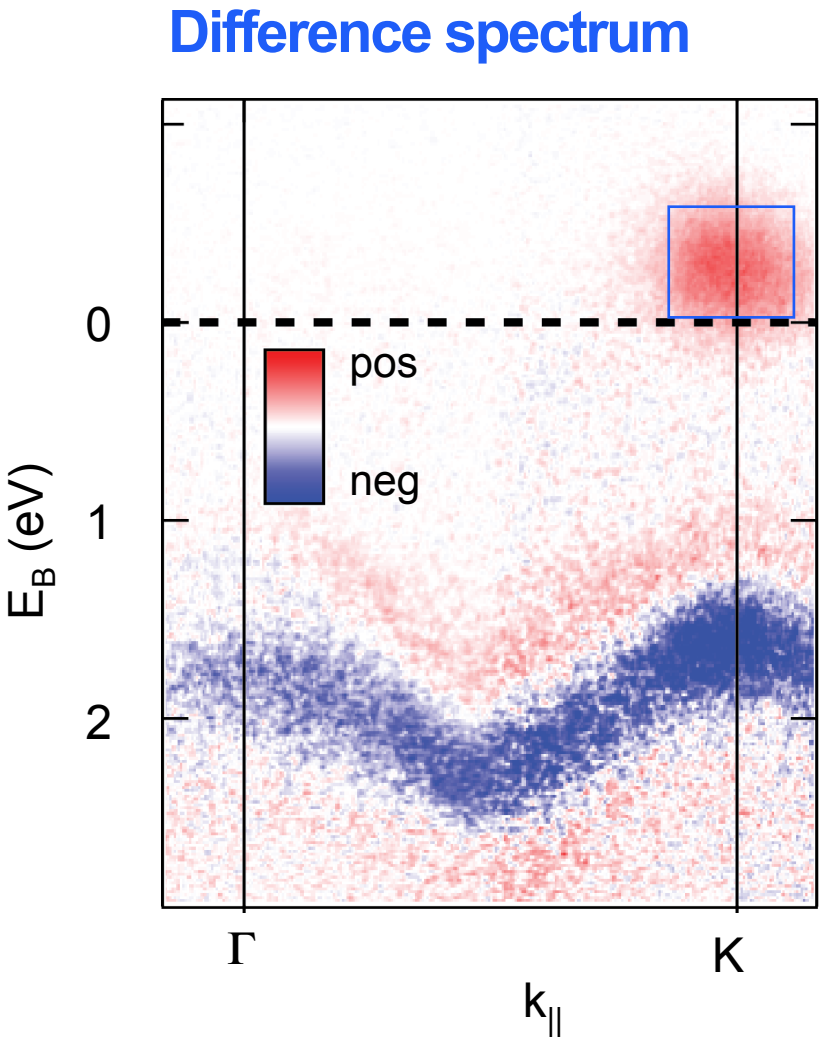
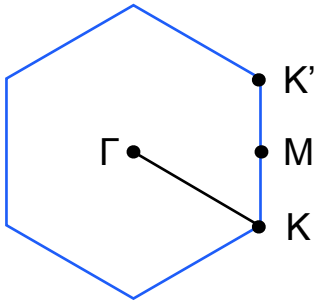
Measurement of excited state





Conduction band minimum

Local valence band maximum

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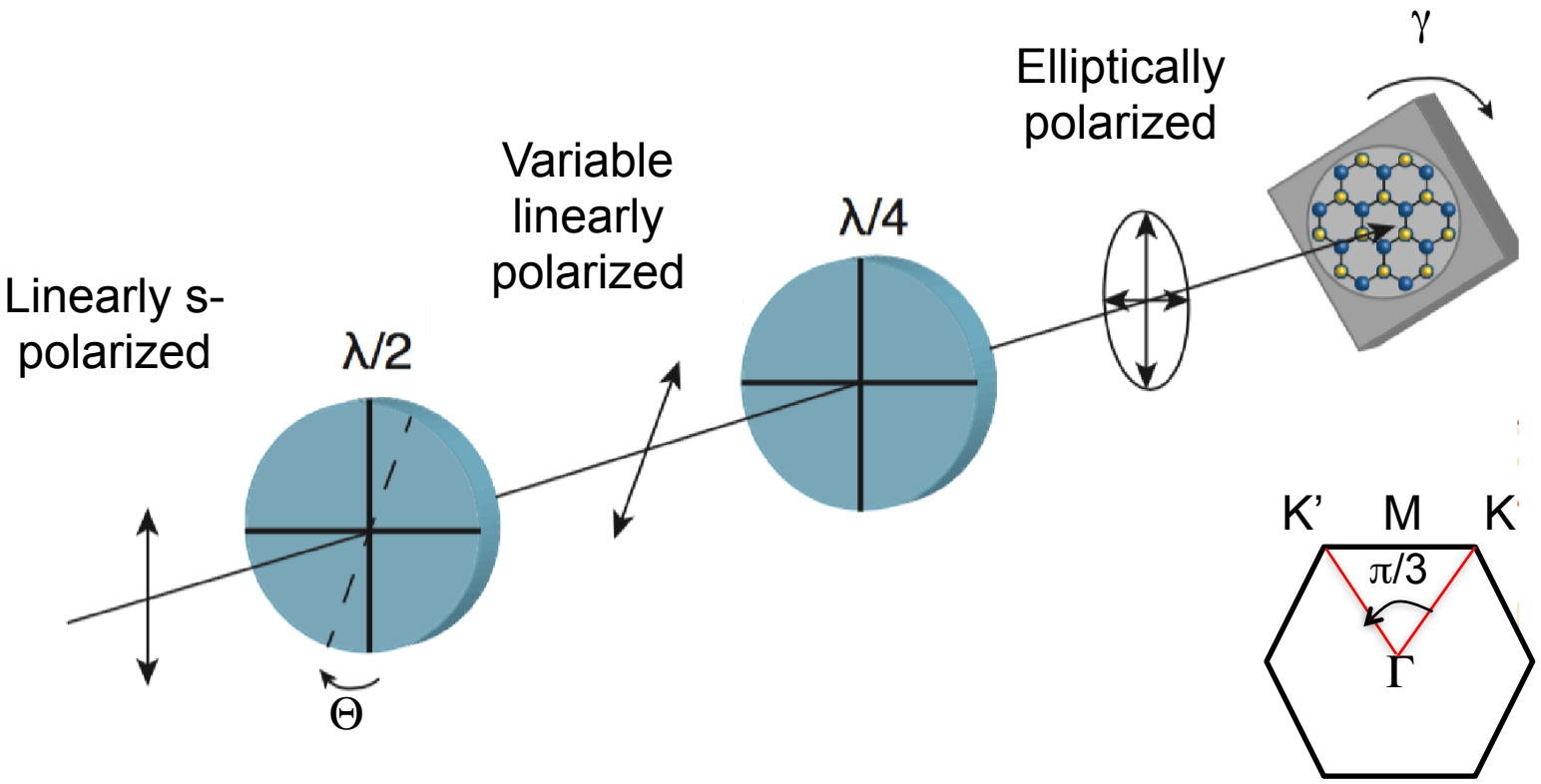
Measurement of excited state



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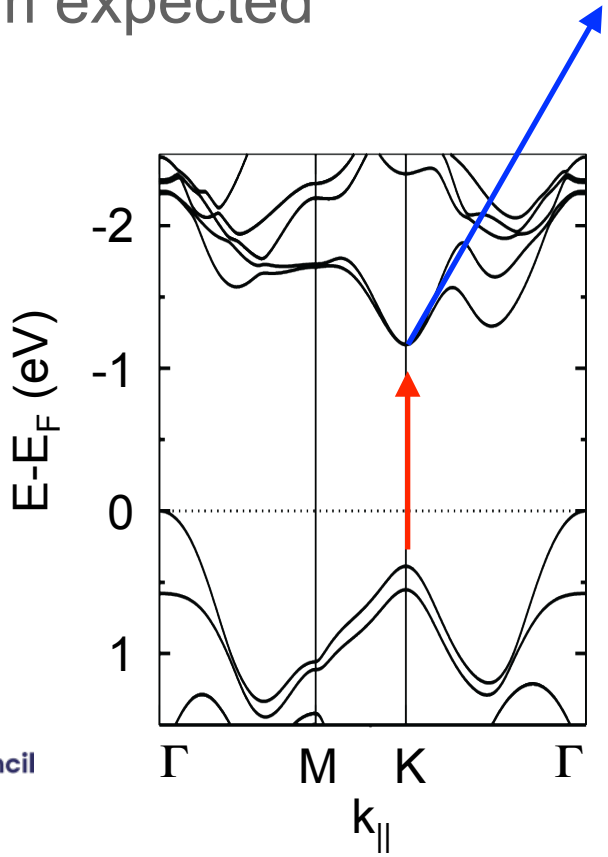
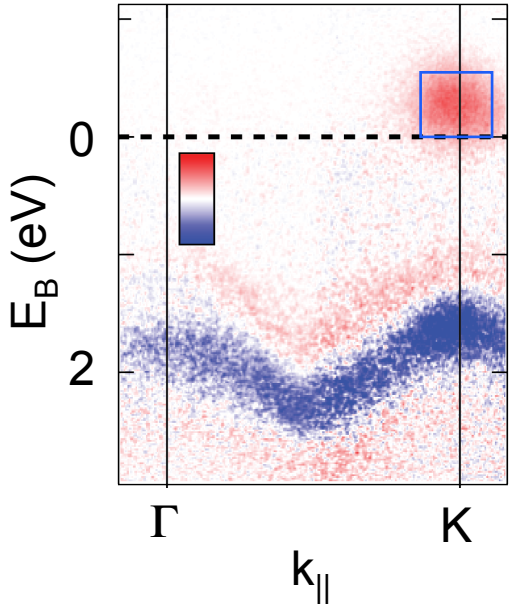
Naïve expectation

- Bilayer is inversion symmetric
- K & K' equivalent
- No dichroism expected





Pump-probe depends on more than one process

- Excitation from the excited state to the final state
- Interband transition

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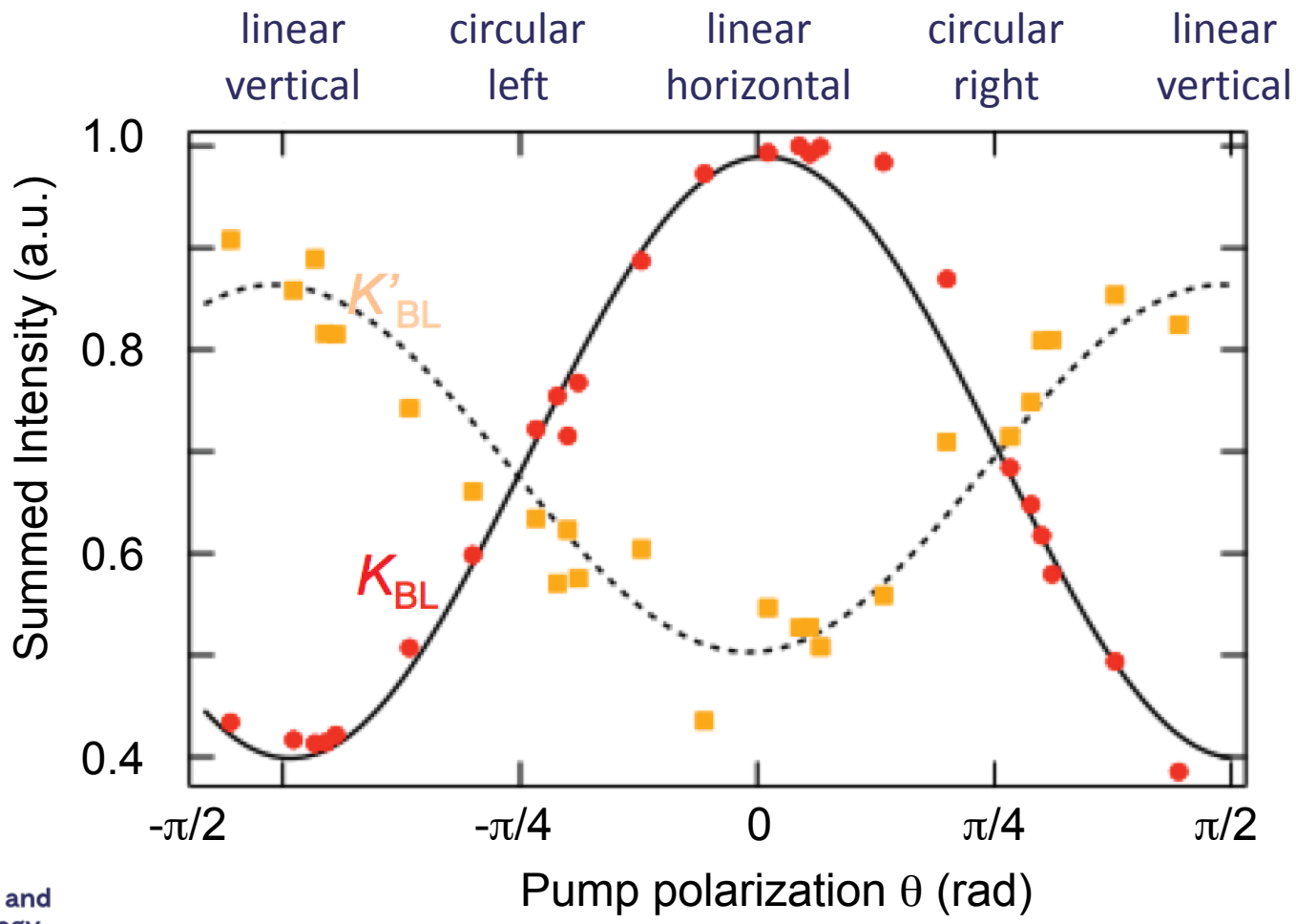
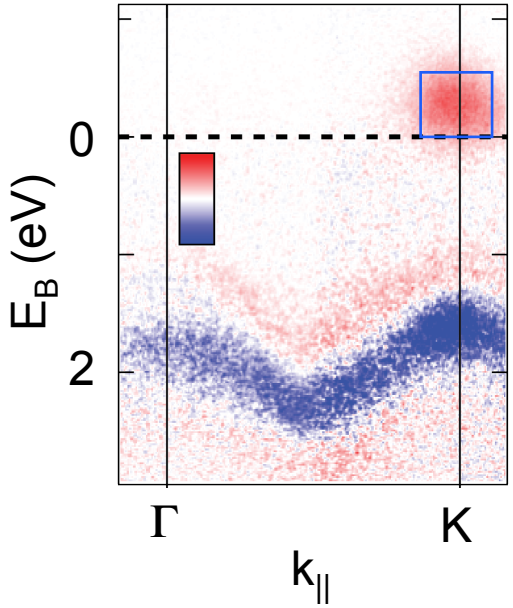
Experimental results





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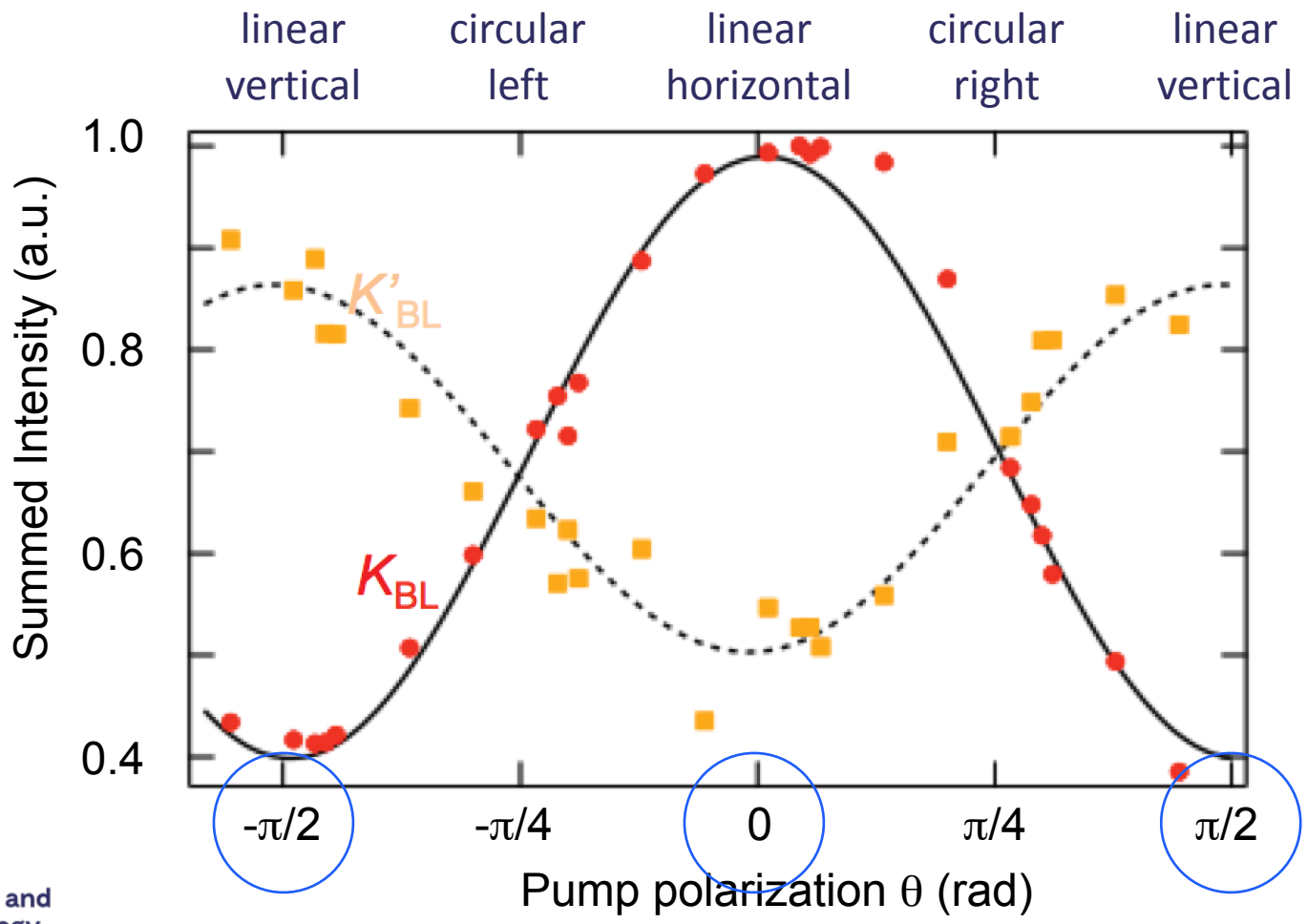
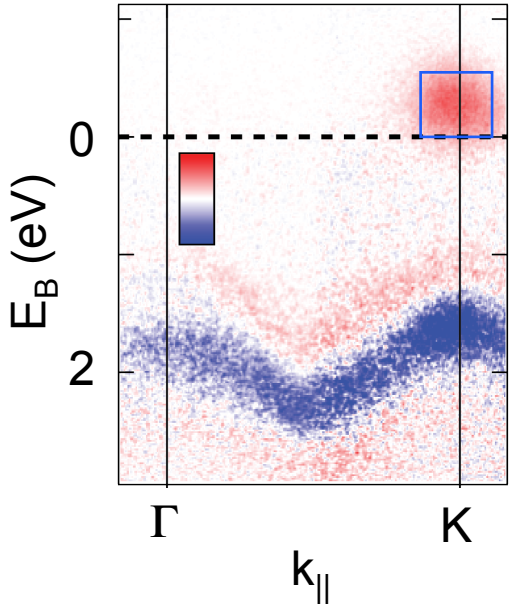
Unexpected dichroism





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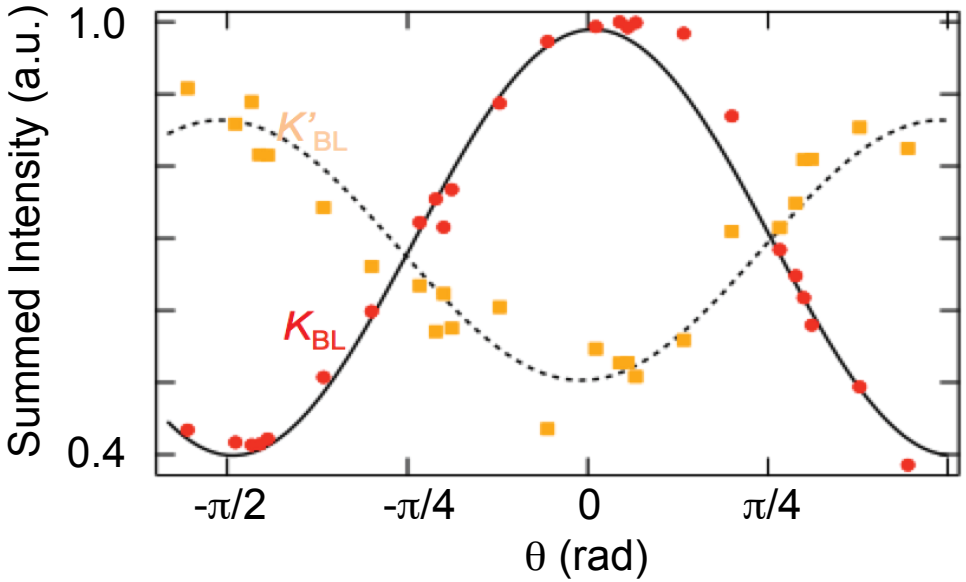
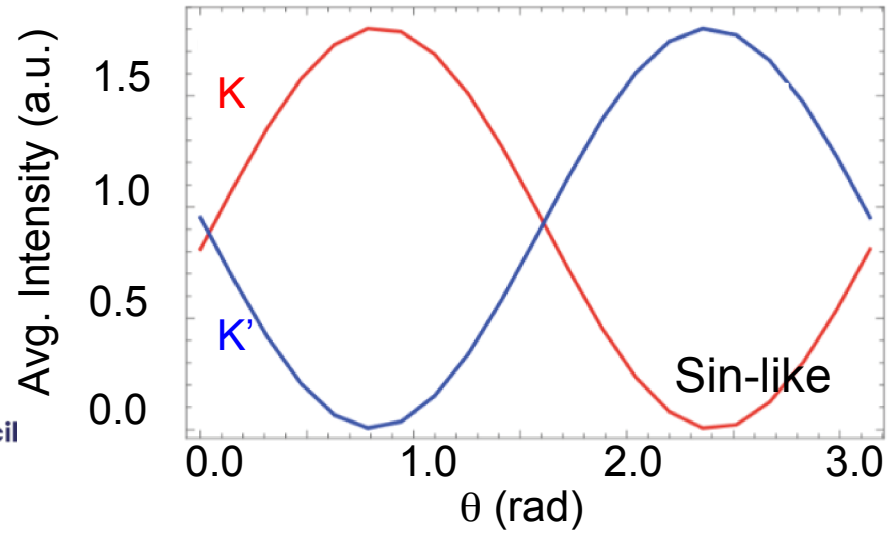
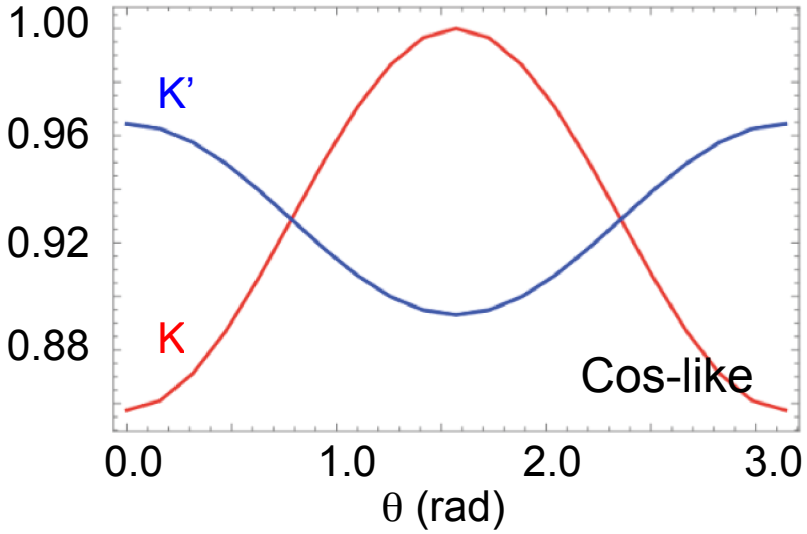
Fitting of observed dichroism



Fitting to a sum of cos and sin terms:
cos term ≈ 4 x sin term





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Theory picture

k p Hamiltonian:

- Bilayer case calculated from simple 4-band model
  - 1 valence & 1 conduction band, top & bottom layers
  - Layer hybridization taken into account
- ***q***: wave vector measured from K
  - φ: azimuthal angle associated with **q**
- τ_z, s_z = +/-1: valley & spin indices

Hamiltonian for single-layer MoS_2

$$\hat{H}_{BL} = \begin{bmatrix} \hat{H}_{SL} & \hat{H}_{\perp} \\ \hat{H}_{\perp}^{\dagger} & \hat{H}_{SL} \end{bmatrix}$$

Interlayer interaction

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Theory picture

Pump polarization

Bilayer Hamiltonian

Matrix element: interband transition

$$M_{cv}(\vec{q},\theta) = < \psi_c(\vec{q}) \,|\, \hat{\epsilon}(\theta) \cdot \nabla_{\vec{q}} \hat{H}_{BL} \,|\, \psi_v(\vec{q}) >$$

yields the excited state population

$$f^{exc}(\vec{q},\theta) \propto 1 + f_{linear}(\vec{q})\cos(2\theta) + f_{circular}(\vec{q})\sin(2\theta)$$

f_{linear} and $f_{circular}$: prefactors describing the relative weight of linear and circular dichroic terms;
dependent on spin, valley

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Theory picture

$$M_{cv}(\vec{q},\theta) = < \psi_c(\vec{q}) \,|\, \hat{\epsilon}(\theta) \cdot \nabla_{\vec{q}} \hat{H}_{BL} \,|\, \psi_v(\vec{q}) >$$

$$f^{exc}(\vec{q},\theta) \propto 1 + f_{linear}(\vec{q})\cos(2\theta) + f_{circular}(\vec{q})\sin(2\theta)$$

$$f_{linear}(q) \approx 2\frac{t_1^2 - 2(\hbar\omega_0)E_{gap}}{E_{gap}^2} q^2 \cos(2\phi)$$

f_{linear} and $f_{circular}$: prefactors describing the relative weight of linear and circular dichroic terms

t_1: intralayer hopping parameter

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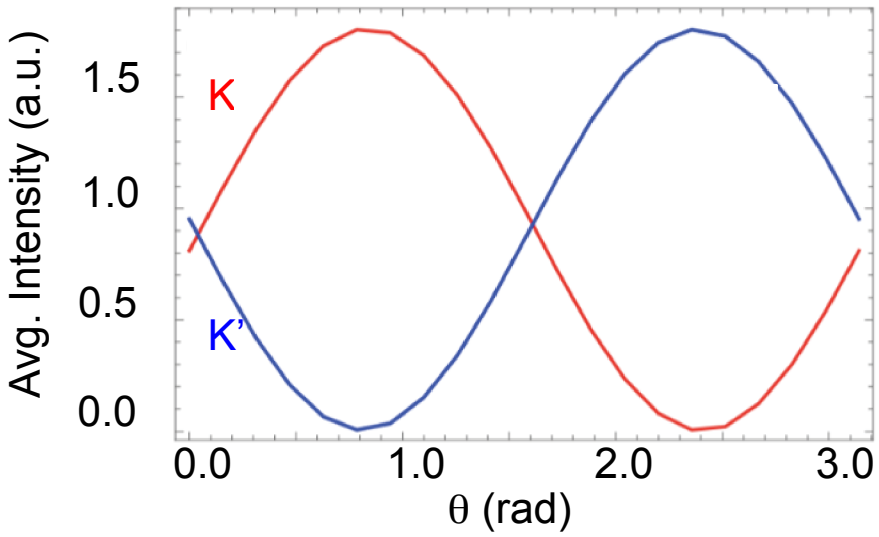
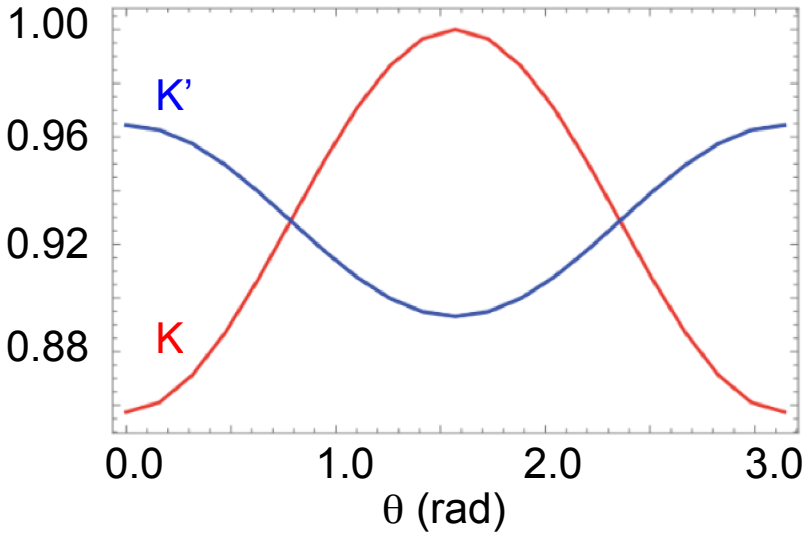
Theory picture

Sine dependence



Cosine dependence



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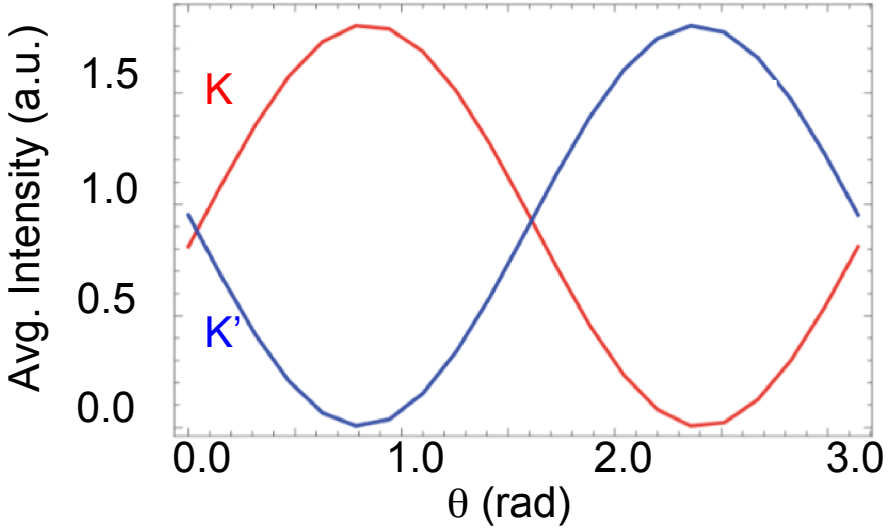
Theory picture

Sine dependence



Inversion-symmetry-broken, circular dichroism

Sin term associated with various symmetry-breaking, such as:

- Presence of single-layer regions on sample
- Substrate effects
- Layer-dependent probe sensitivity
- Possible layer pseudospin

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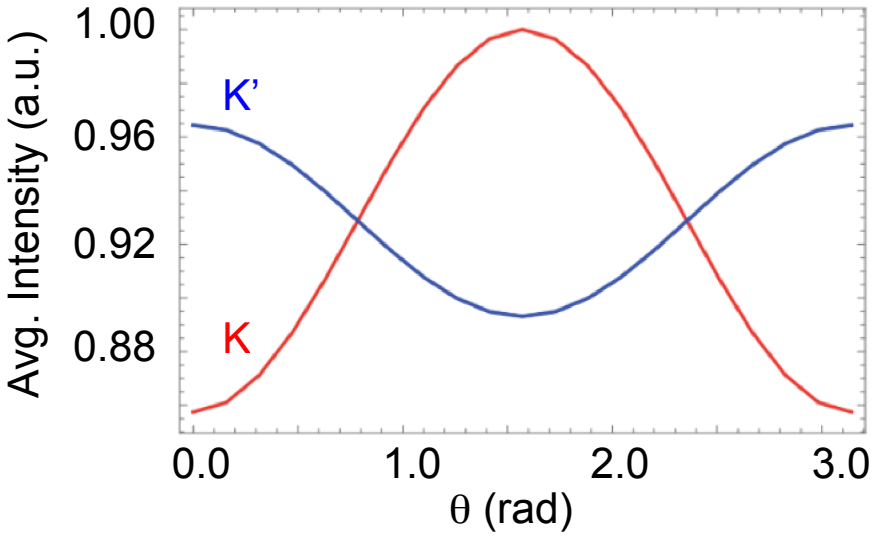
Theory picture

Cos term:

- Related to matrix element of pumped excitation
  - Linear dichroism not exclusive to this material system
- Arises from intralayer rather than interlayer hopping
- Previously overlooked because of the circular dichroism in single-layer MoS_2

Cosine dependence



Linear dichroism

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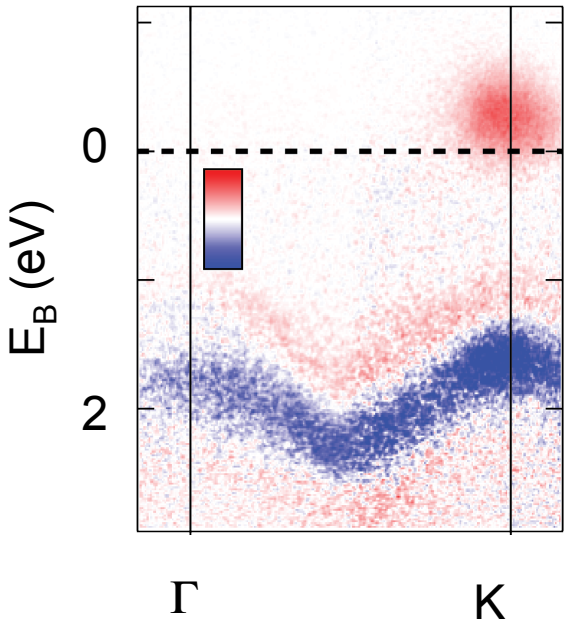
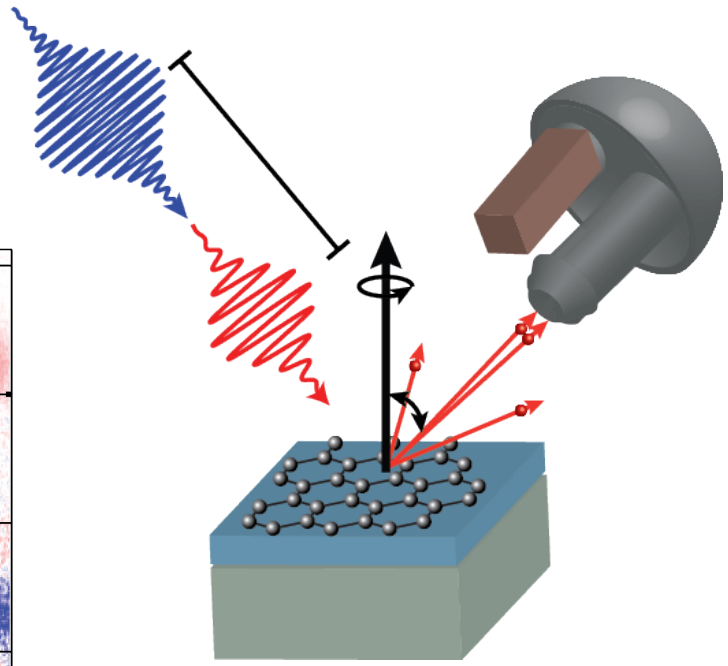
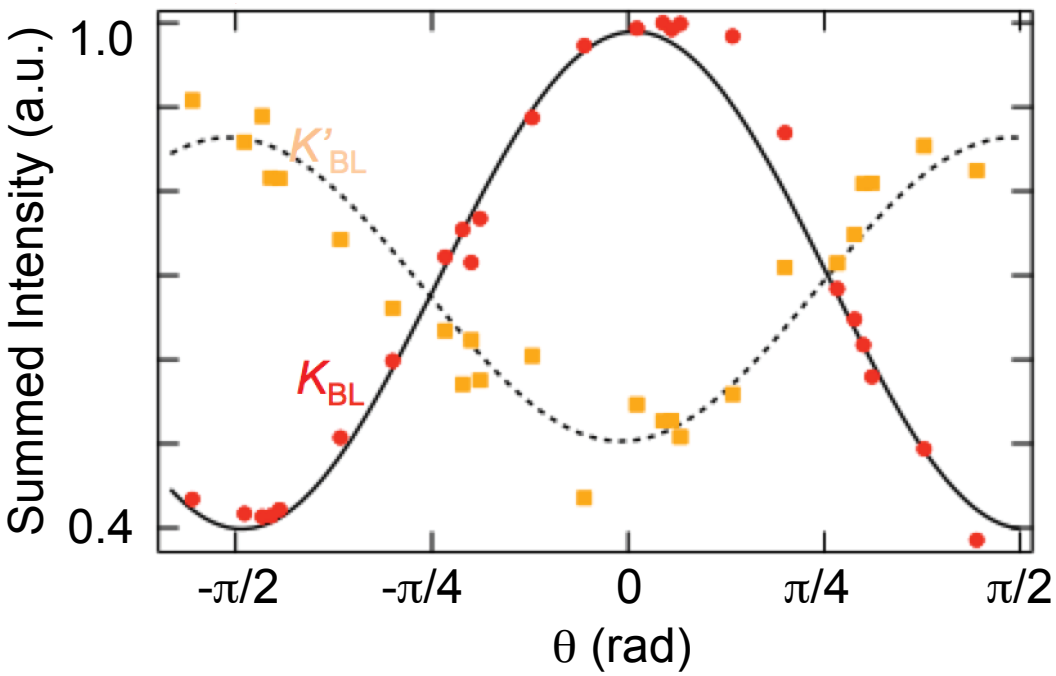
Summary







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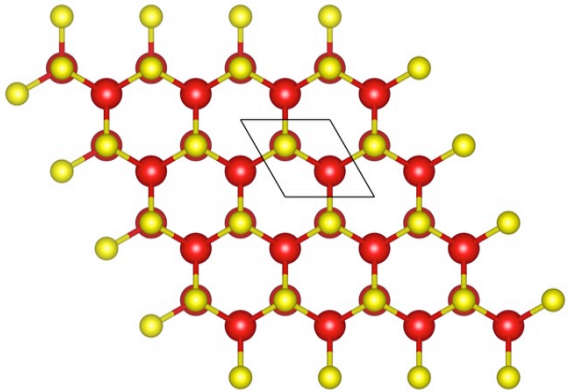

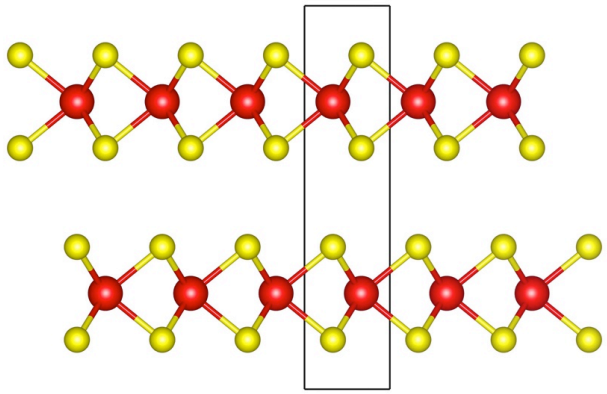

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Thank you

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