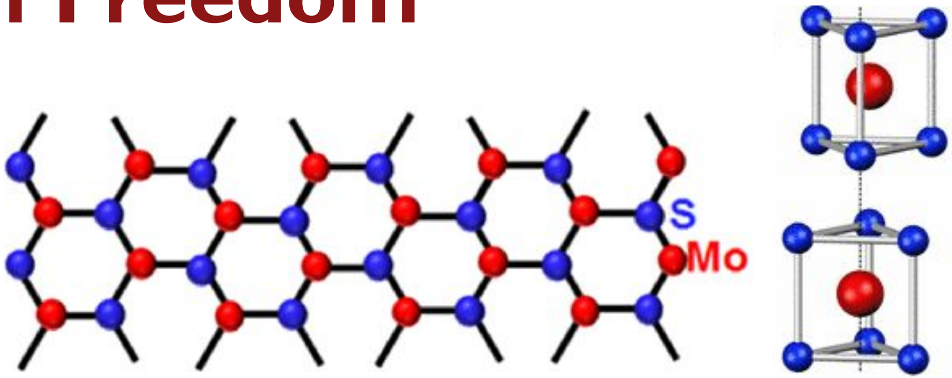
Control of Valley Polarization in Monolayer MoS_2 by Optical Helicity

Ben Safvati

APPPHYS 204 - Quantum Materials

The Valley Degree of Freedom

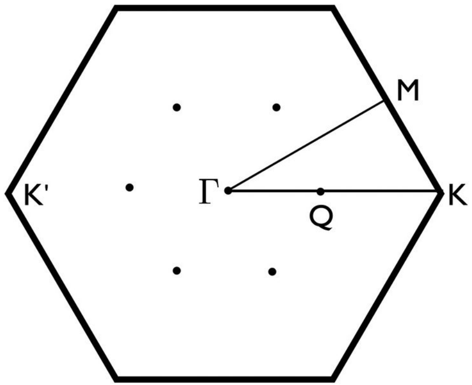
- Band structures of certain materials contain energy extrema called "valleys."

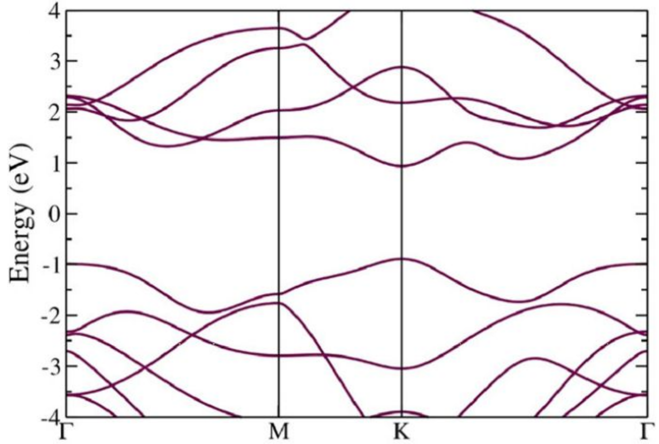


For MoS_2:

- 2 degenerate valleys at K, K' points, opposite in momentum space.

- Direct ~1.8 eV band gap.





The Valley Degree of Freedom

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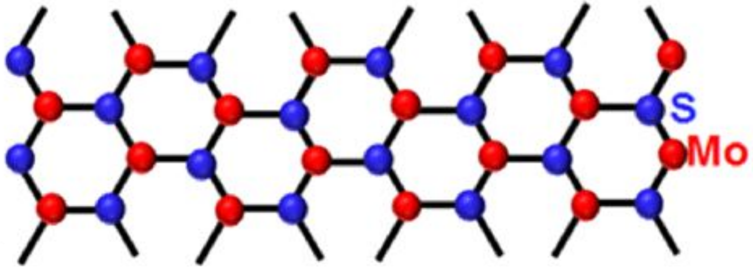

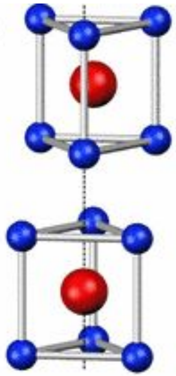

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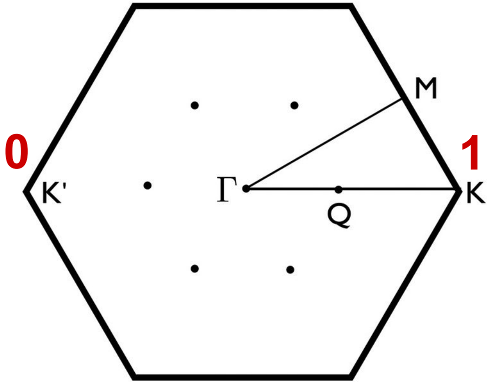
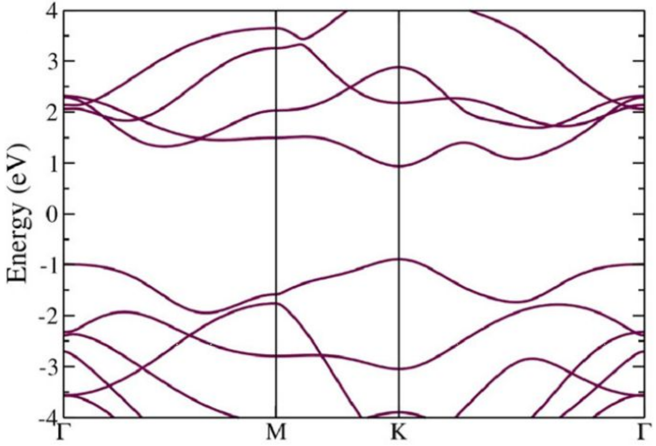
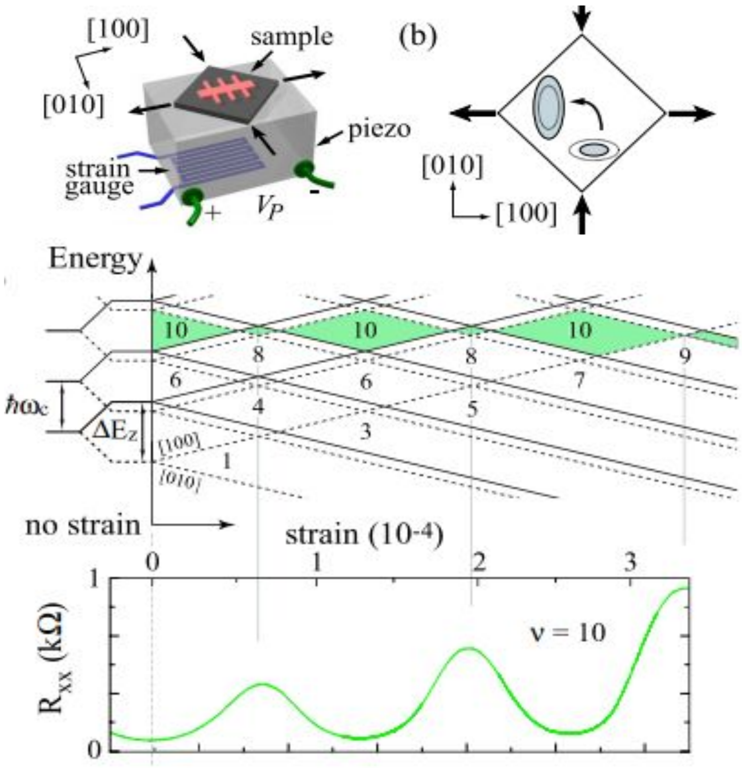




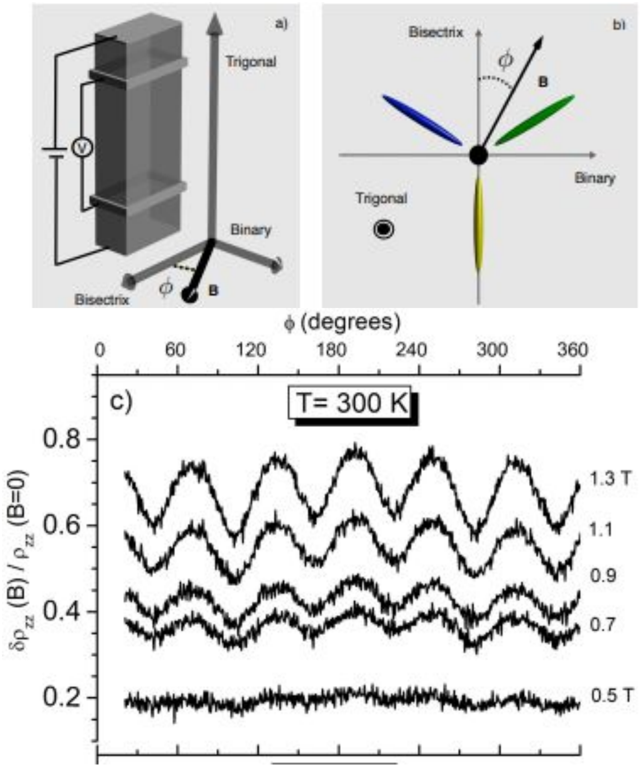
Prior Work on Valley Polarization

Strain-induced valley splitting of FQHE states in AlAs



Bishop, N. C. et al. *Phys. Rev. Lett.* 98, 266404 (2007).

Valley valve in Bi with rotating B field

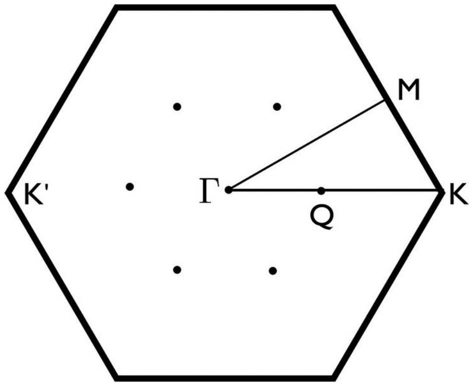


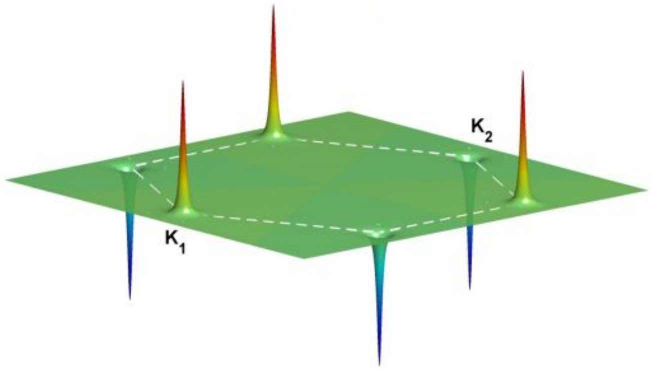
Zhu, Z., Collaudin, A., Fauqué, B., Kang, W. & Behnia, K. *Nature Phys.* 8, 89–94 (2012).

MoS_2 Symmetry Considerations

- Consider valley-differentiated orbital magnetic moment $m_v \propto$ valley index $\boldsymbol{\tau} = \pm 1$.
  - $\boldsymbol{\tau}$ odd under time reversal and spatial inversion
  - m_v odd under time reversal, but even under spatial inversion.
- Thus, m_v can only be nonzero for inversion symmetry breaking

  ⇒ spatial inversion symmetry breaking is necessary to observe valley-contrasting magnetic moment.

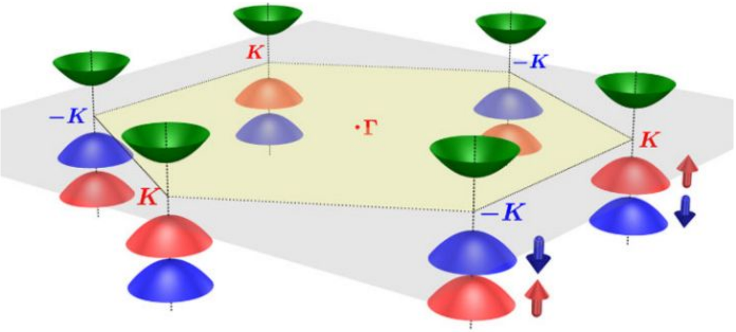




TMD Minimal Band Model

Symmetry-adapted basis states
between band-edge electrons

$$|\phi_c\rangle = |d_{z^2}\rangle, \qquad |\phi_v^\tau\rangle = \frac{1}{\sqrt{2}}(|d_{x^2-y^2}\rangle + i\tau|d_{xy}\rangle)$$



Spin-Valley Locking

Related by
time reversal

k·p Hamiltonian
expanded around K point

$$\hat{H}_0 = at(\tau k_x\hat{\sigma}_x + k_y\hat{\sigma}_y) + \frac{\Delta}{2}\hat{\sigma}_z$$

Spin-orbit coupling
expanded around K point

$$H_{soc} = \lambda\tau\frac{\hat{\sigma}_z - 1}{2}\hat{s}_z$$

Chiral Optical Selection Rules

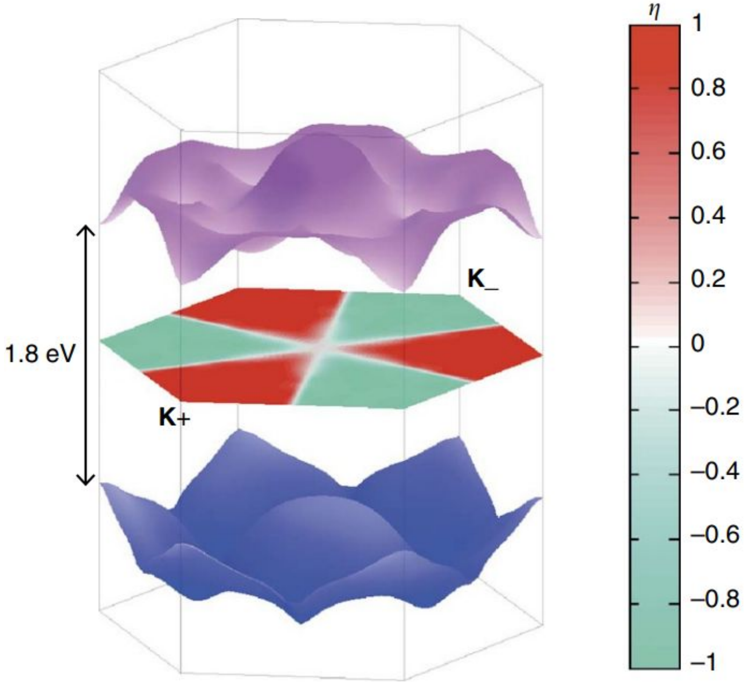
Optical interband transition elements

$$\mathcal{P}_{\pm}(\boldsymbol{k}) \equiv \mathcal{P}_x(\boldsymbol{k}) \pm i\mathcal{P}_y(\boldsymbol{k})$$

$$\mathcal{P}_{\alpha}(\boldsymbol{k}) \equiv m_0 \langle u_c(\boldsymbol{k}) | \frac{1}{\hbar} \frac{\partial \hat{H}}{\partial k_\alpha} | u_v(\boldsymbol{k}) \rangle$$

$$|\mathcal{P}_{\pm}(\boldsymbol{k})|^2 = \frac{m_0^2 a^2 t^2}{\hbar^2}\left(1 \pm \tau \frac{\Delta'}{\sqrt{\Delta'^2 + 4a^2t^2k^2}}\right)^2.$$

$\Delta' \ggg atk$ ⇒ **Nearly perfect valley selection rules!**



$$\eta(\mathbf{k}, \omega_{cv}) = \frac{|\mathcal{P}_+^{cv}(\mathbf{k})|^2 - |\mathcal{P}_-^{cv}(\mathbf{k})|^2}{|\mathcal{P}_+^{cv}(\mathbf{k})|^2 + |\mathcal{P}_-^{cv}(\mathbf{k})|^2}$$

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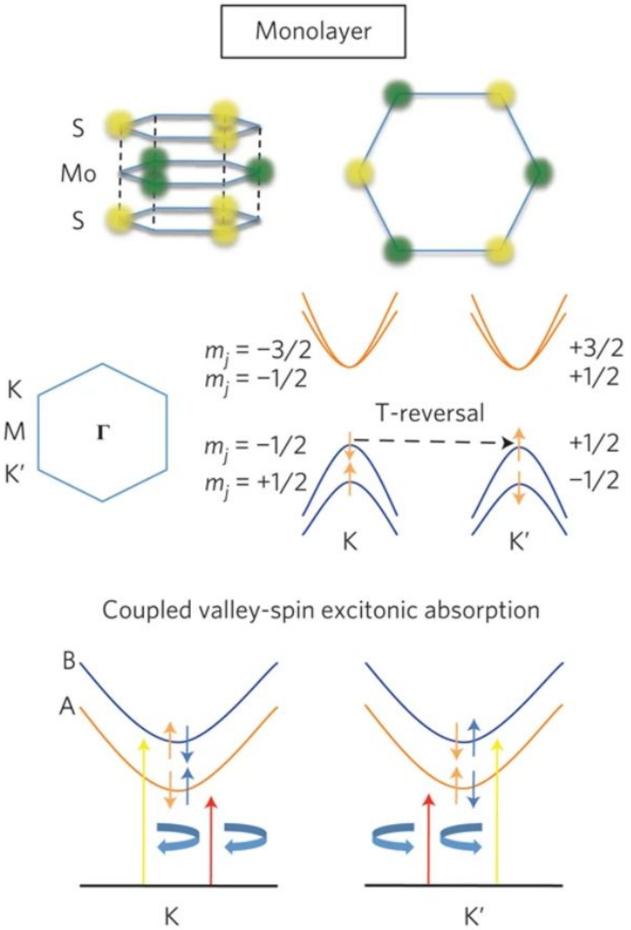
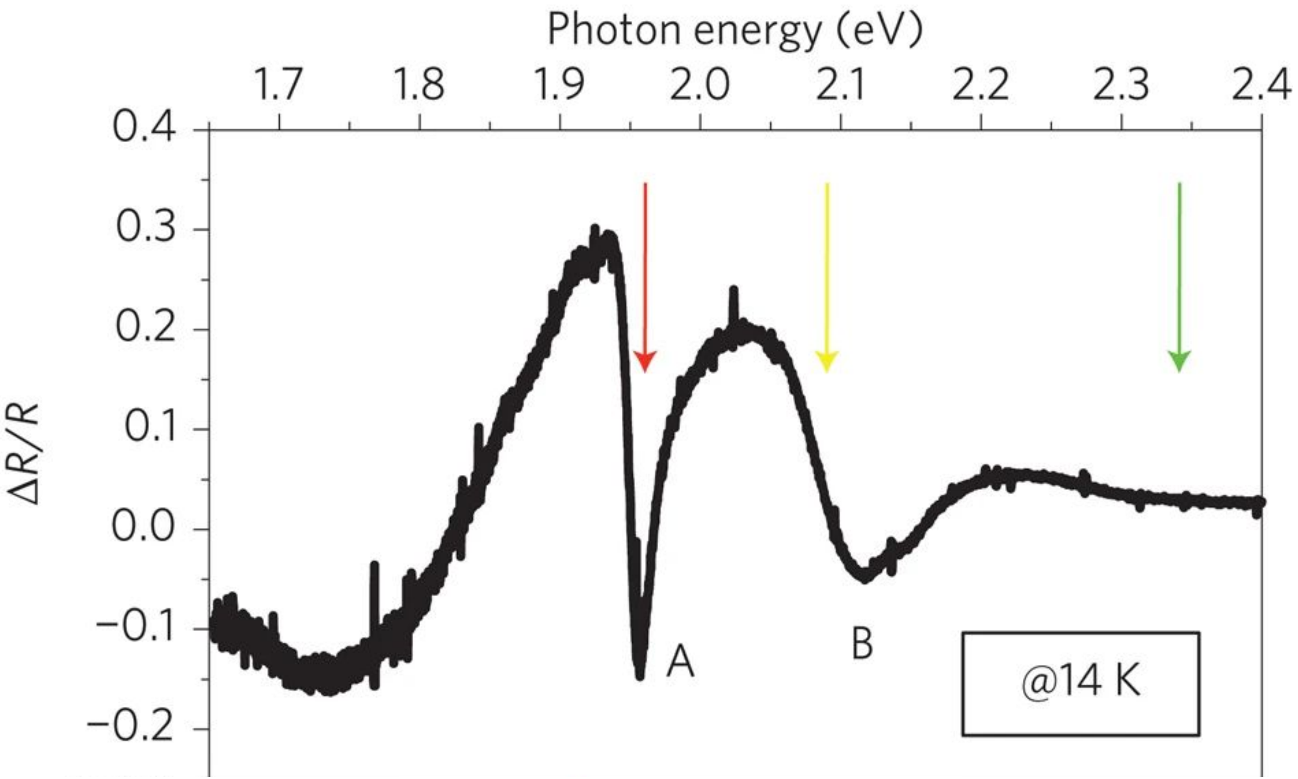
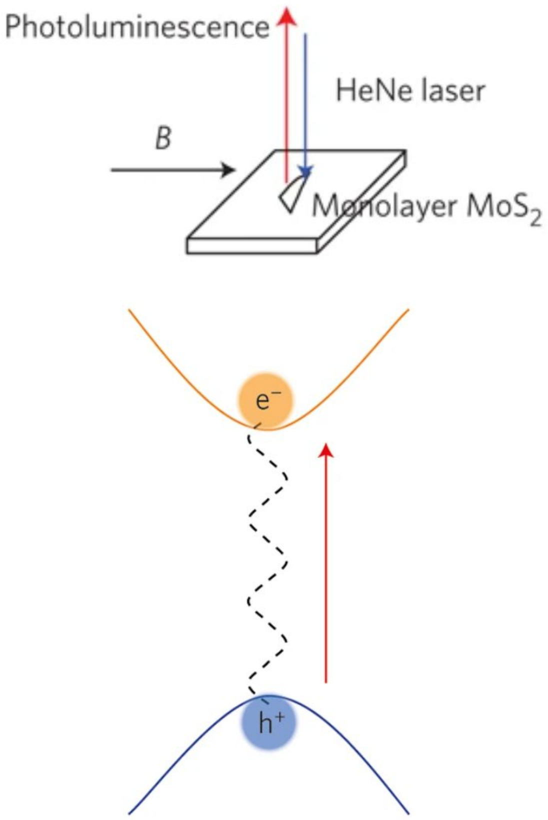


Mak, K., He, K., Shan, J. *et al. Nature Nanotech* 7, 494–498 (2012).

Excitonic Absorption Spectrum





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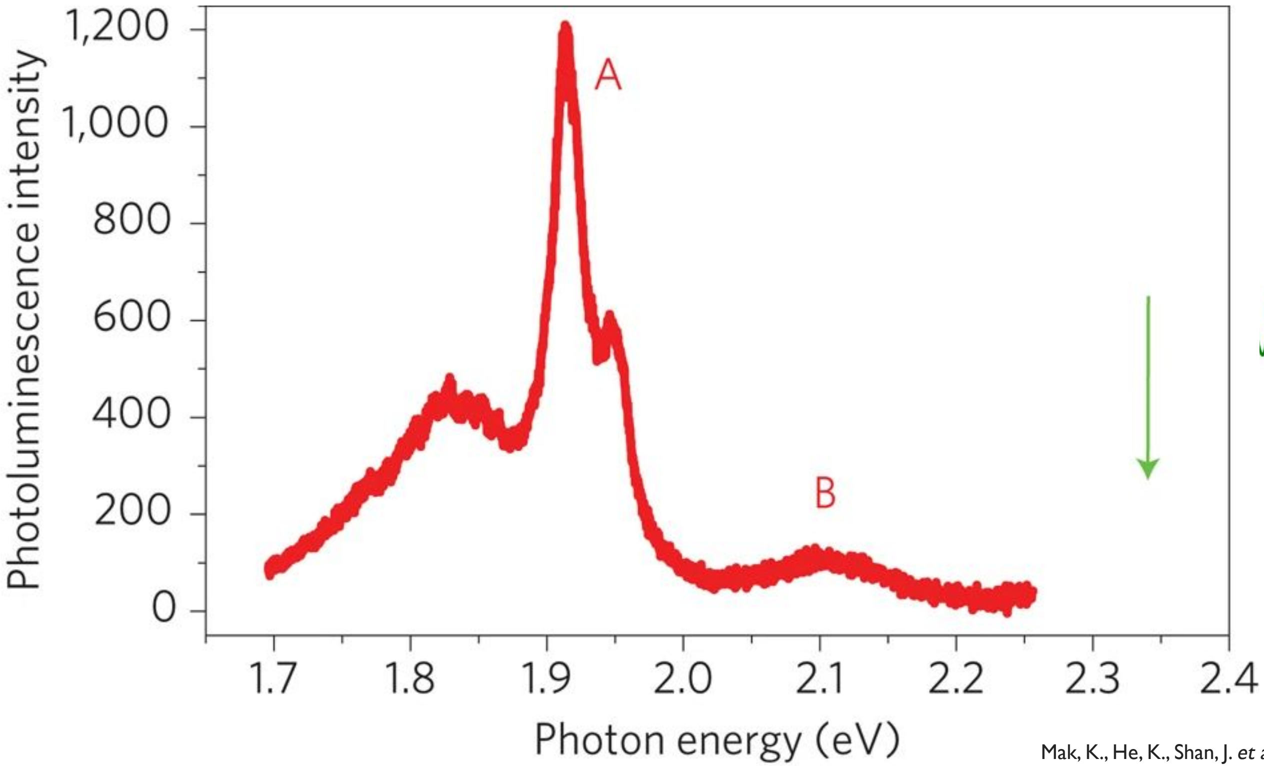
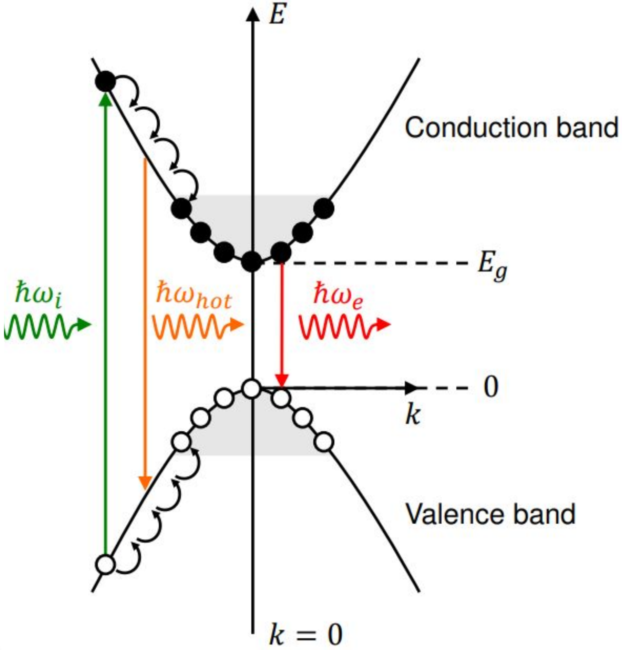
Excitonic Photoluminescence Spectrum





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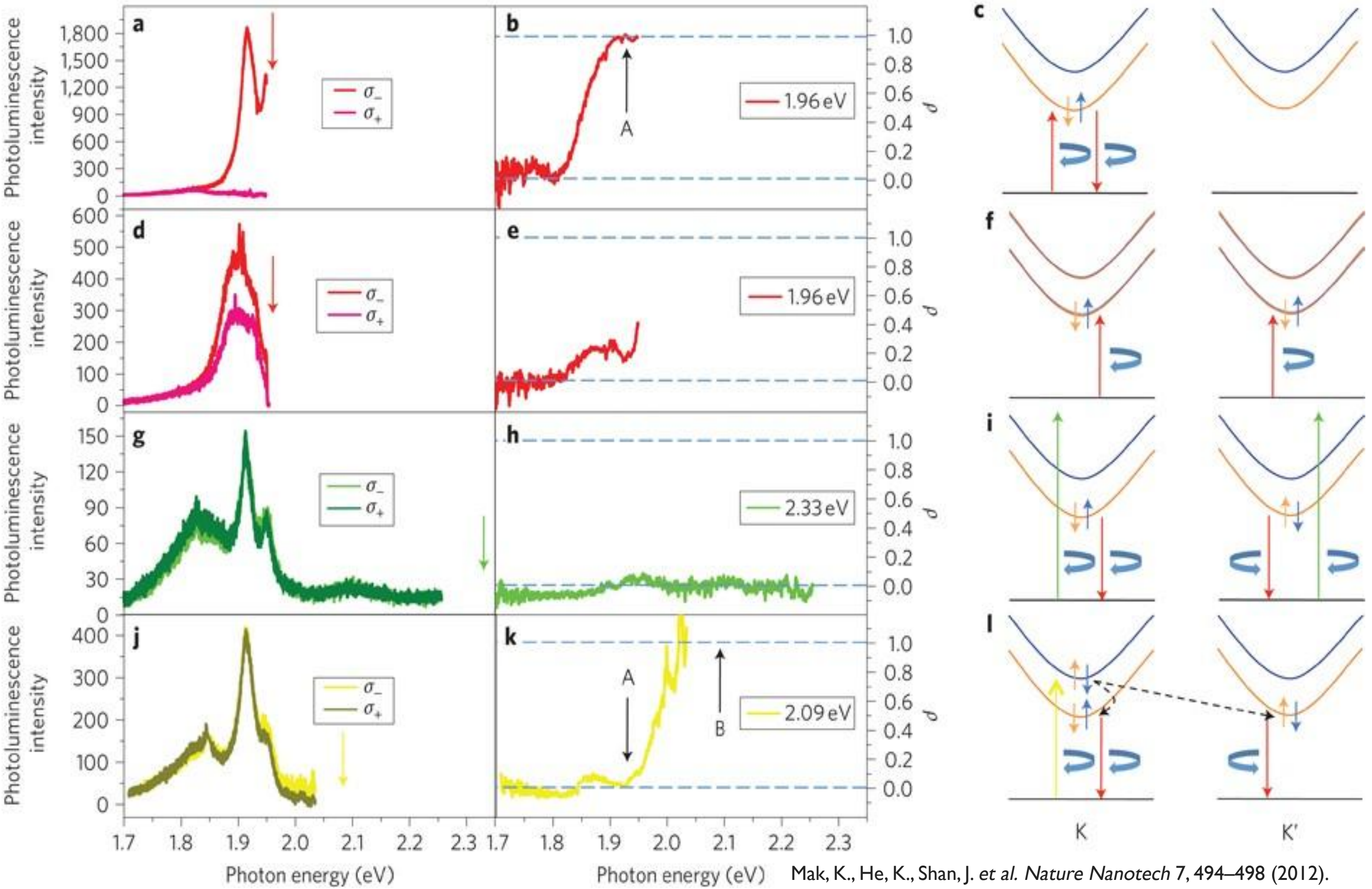
A Exciton Resonance
Perfect helicity

Bilayer MoS_2
No valley polarization

Off-Resonance
Simultaneous population

B exciton Resonance
Perfect helicity
(hot luminescence)



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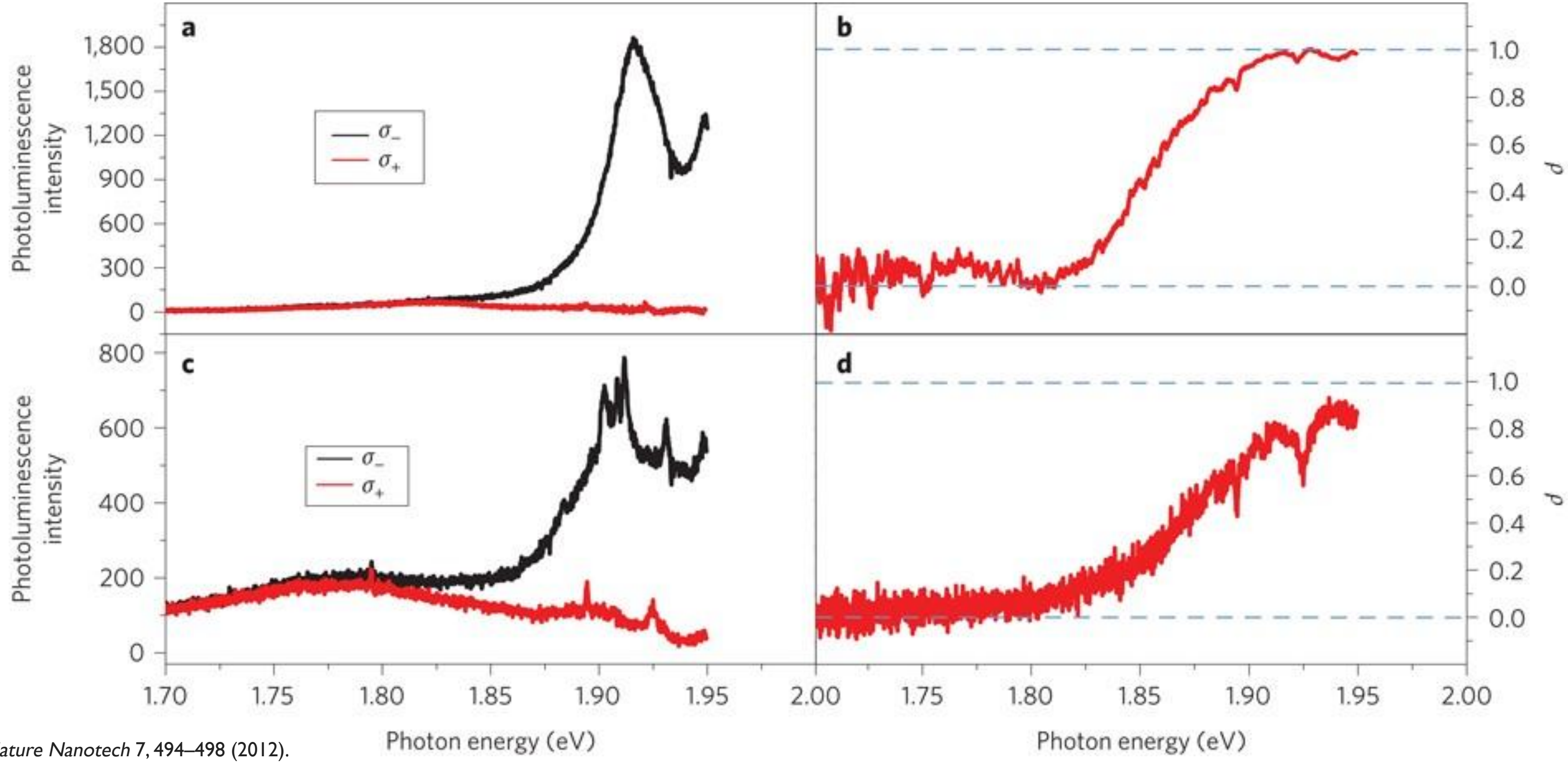
Substrate-Independent Optical Helicity

h-BN

SiO_2/Si



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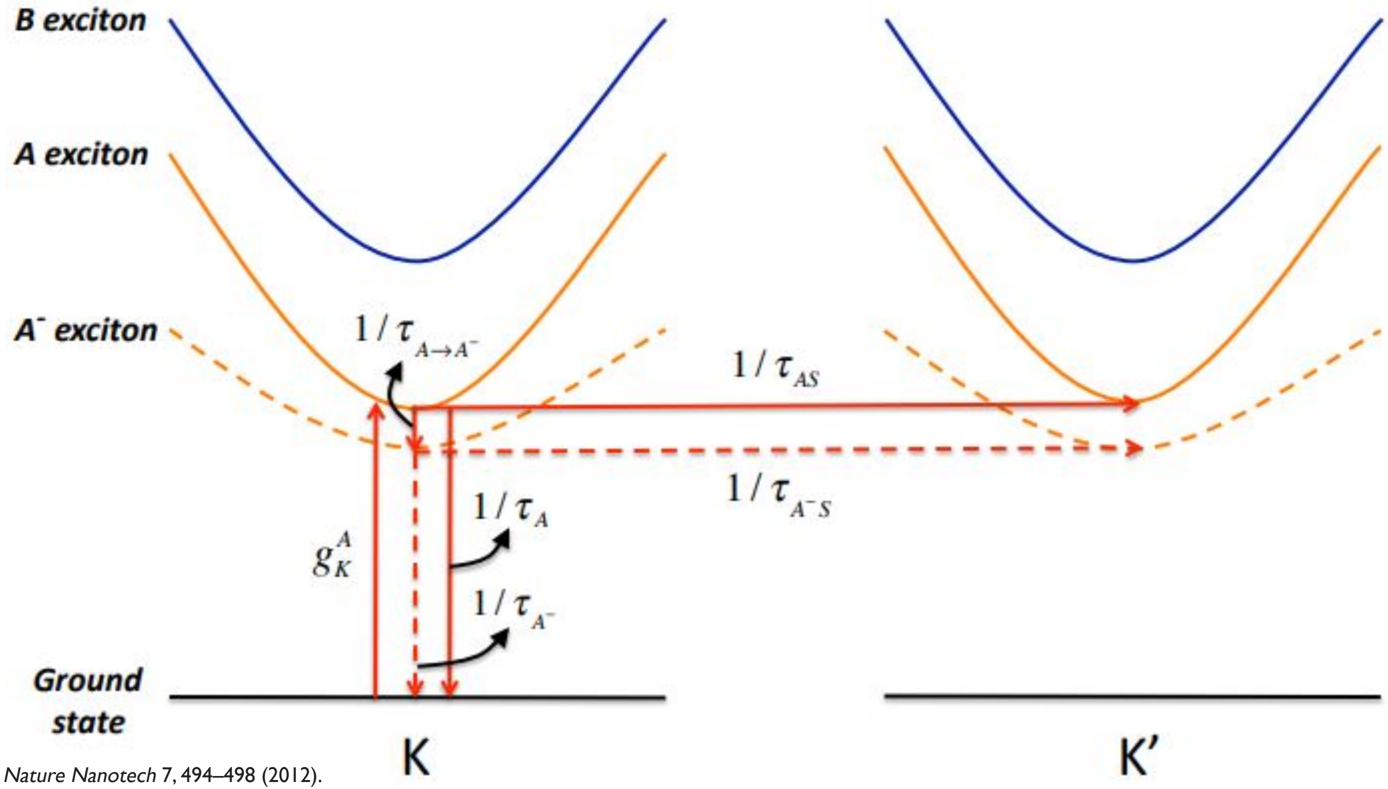
Future Research: Relaxation Mechanisms



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Thank you!

Chiral Optical Selection Rules