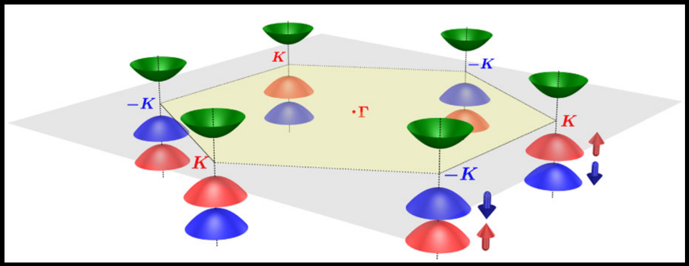
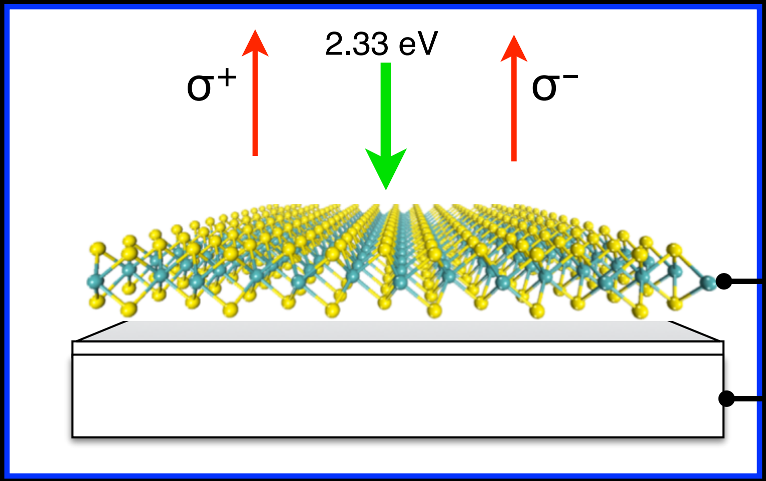
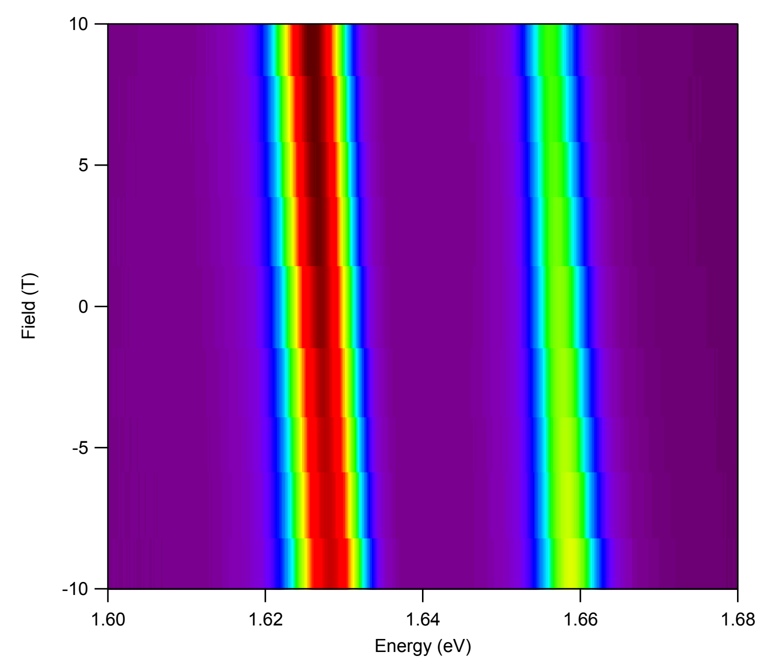
Magneto-spectroscopy of excitons in monolayer transition metal dichalcogenides

Valley splitting and polarization by magnetic field in monolayer $MoSe_2$







Dmitry Smirnov

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NATIONAL HIGH
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Magneto-spectroscopy of excitons in monolayer transition metal dichalcogenides

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Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014).

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James Hone
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DMR-1106225
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DE-FG02-07ER46451

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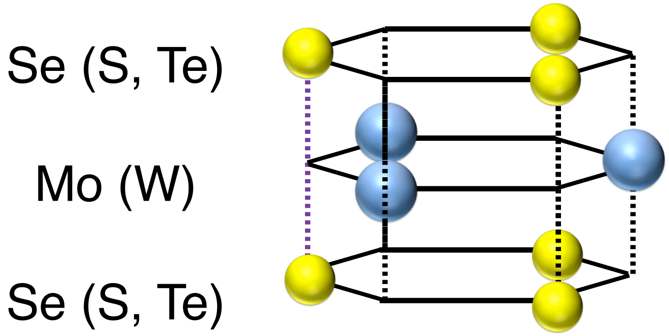
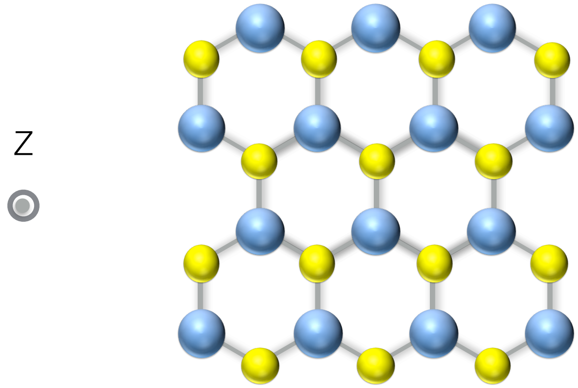
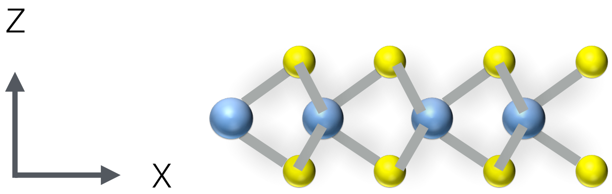
Semiconducting monolayer TMDs

MX$_2$

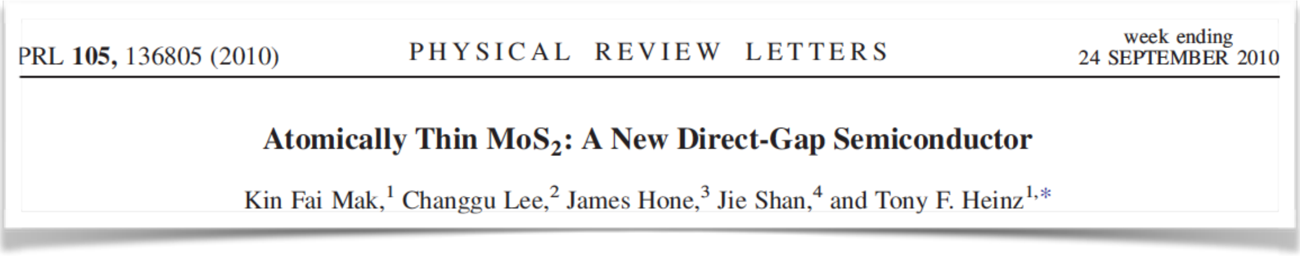






Bulk MoS$_2$: indirect-gap

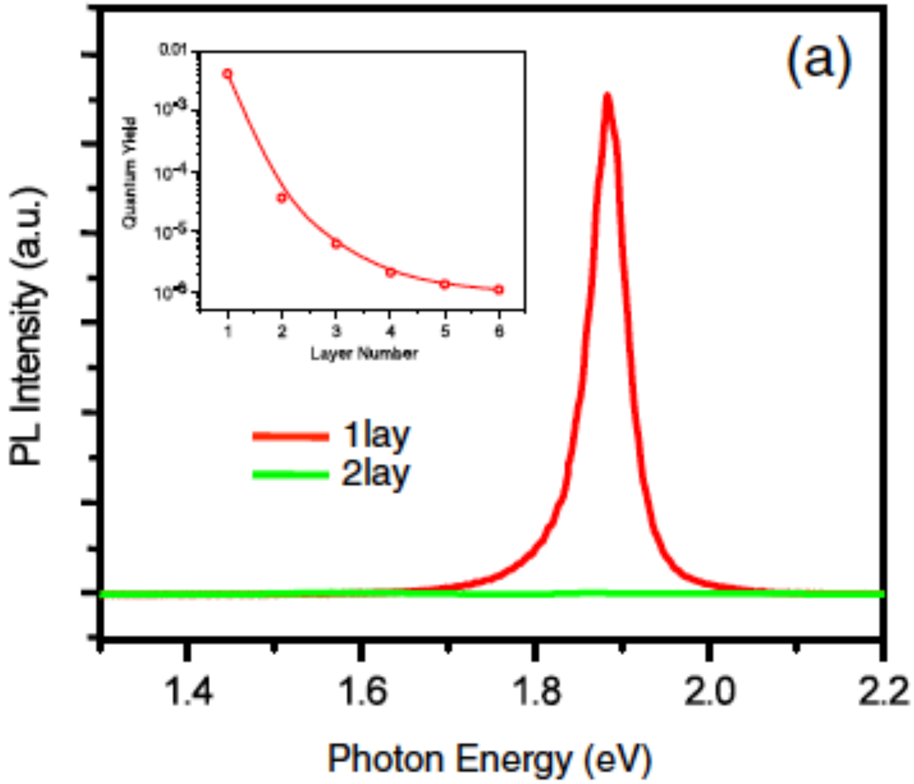
From indirect gap (bulk) to direct bandgap in monolayer



PRL **105,** 136805 (2010)　PHYSICAL REVIEW LETTERS　week ending 24 SEPTEMBER 2010

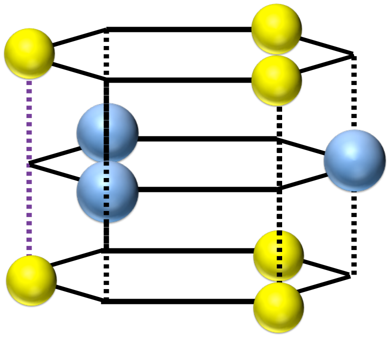
Atomically Thin MoS$_2$: A New Direct-Gap Semiconductor

Kin Fai Mak,[1] Changgu Lee,[2] James Hone,[3] Jie Shan,[4] and Tony F. Heinz[1,*]



Semiconducting monolayer TMDs

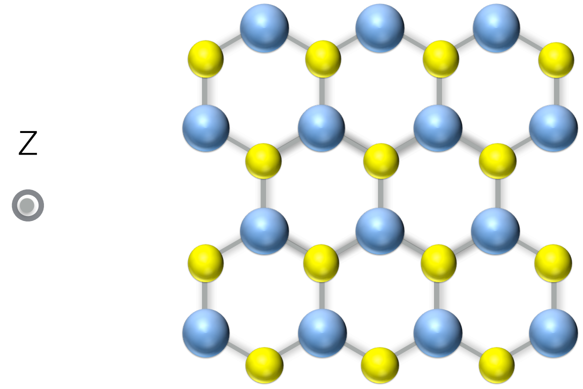
MX_2

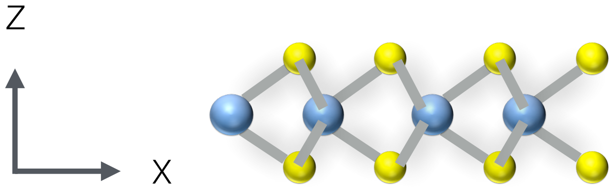


Se (S, Te)

Mo (W)

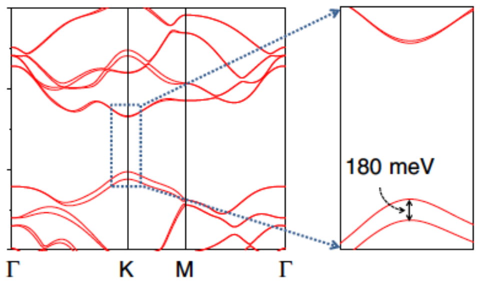
Se (S, Te)



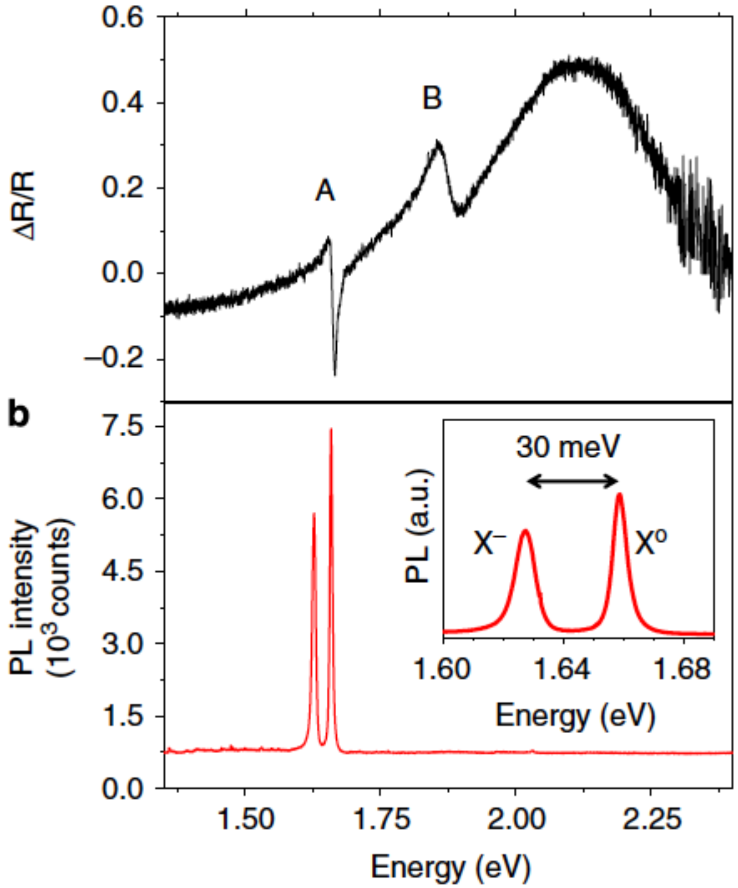


Bulk MoS_2 : 1.3 eV indirect-gap

From indirect gap (bulk) to direct bandgap in monolayer

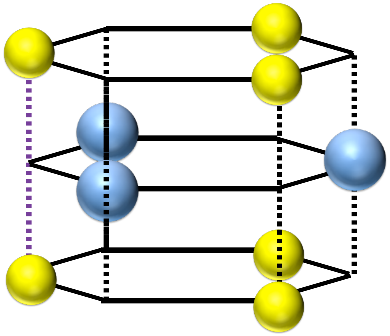


Ross et al. Nature Comm., 4:1474 (2013)



Semiconducting monolayer TMDs

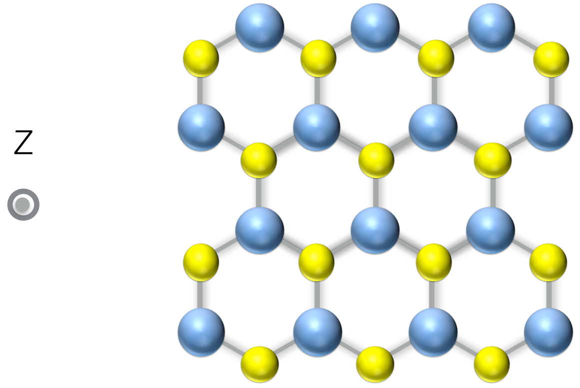
MX_2

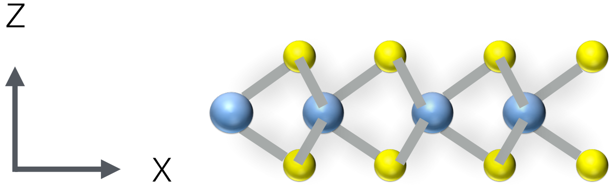


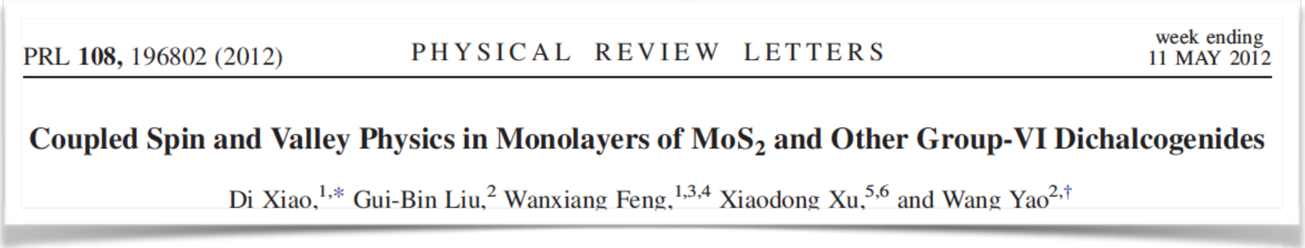
Se (S, Te)

Mo (W)

Se (S, Te)



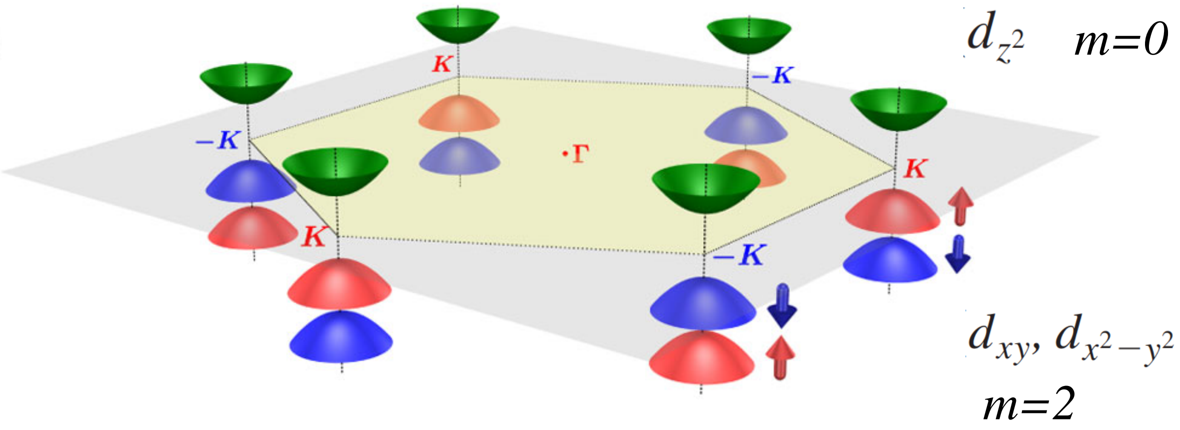




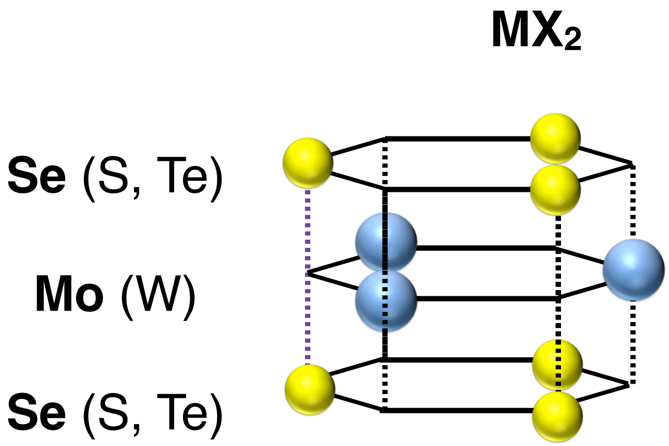
PRL **108,** 196802 (2012) PHYSICAL REVIEW LETTERS week ending 11 MAY 2012

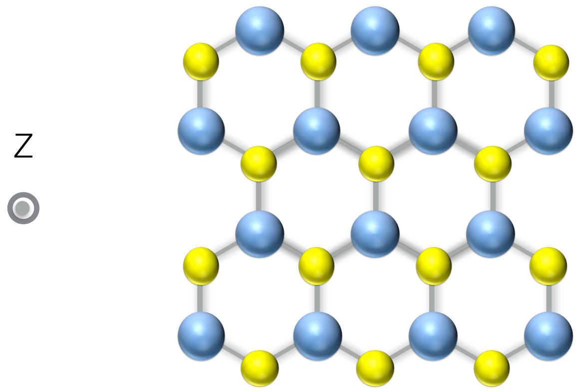
Coupled Spin and Valley Physics in Monolayers of MoS_2 and Other Group-VI Dichalcogenides

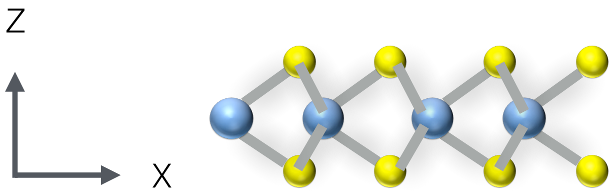
Di Xiao,[1,*] Gui-Bin Liu,[2] Wanxiang Feng,[1,3,4] Xiaodong Xu,[5,6] and Wang Yao[2,†]

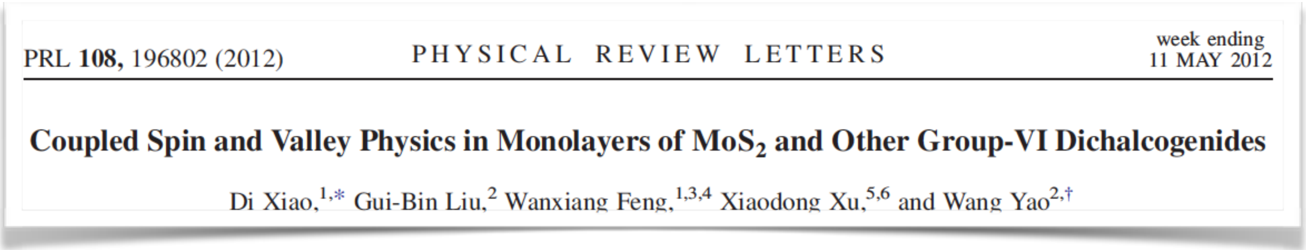


Semiconducting monolayer TMDs







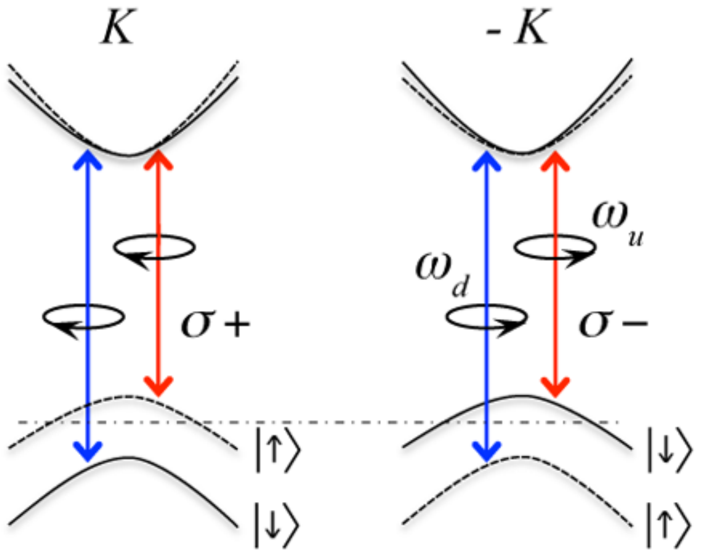


PRL **108,** 196802 (2012) PHYSICAL REVIEW LETTERS week ending 11 MAY 2012

Coupled Spin and Valley Physics in Monolayers of MoS$_2$ and Other Group-VI Dichalcogenides

Di Xiao,[1,*] Gui-Bin Liu,[2] Wanxiang Feng,[1,3,4] Xiaodong Xu,[5,6] and Wang Yao[2,†]

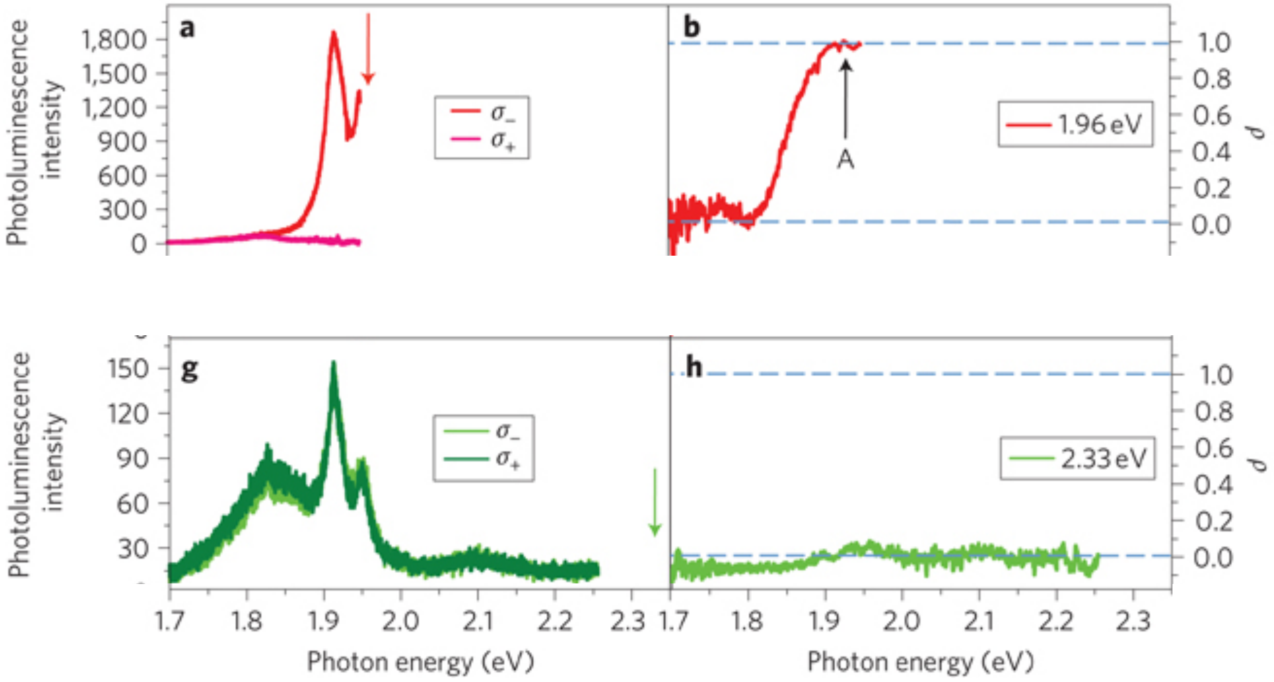
Spin-valley coupling

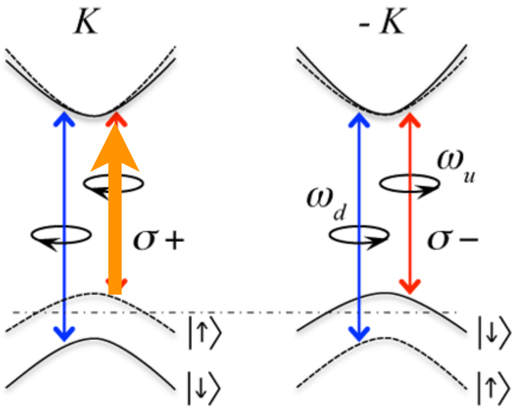


Valley-spin coupling

Circular Polarized PL at Resonance Excitation

- Strong polarization selectivity, preservation of circular state
- Creation of ***transient*** valley population imbalance



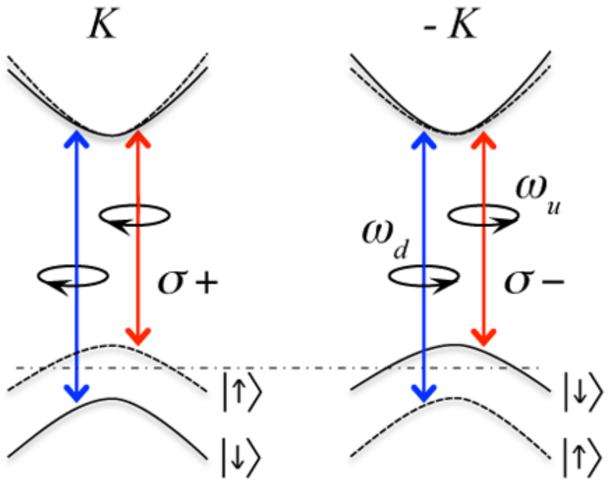


Mak, K. F., He, K., Shan, J., & Heinz, T. F. Nature Nanotechn, 7, 494 (2012)
Also experiments by X. Cui, J. Feng, B. Urbaszek groups

Valley-spin coupling

Circular Polarized PL at Resonance Excitation

- **Strong polarization selectivity, preservation of circular state**
- **Creation of *transient* valley population imbalance**



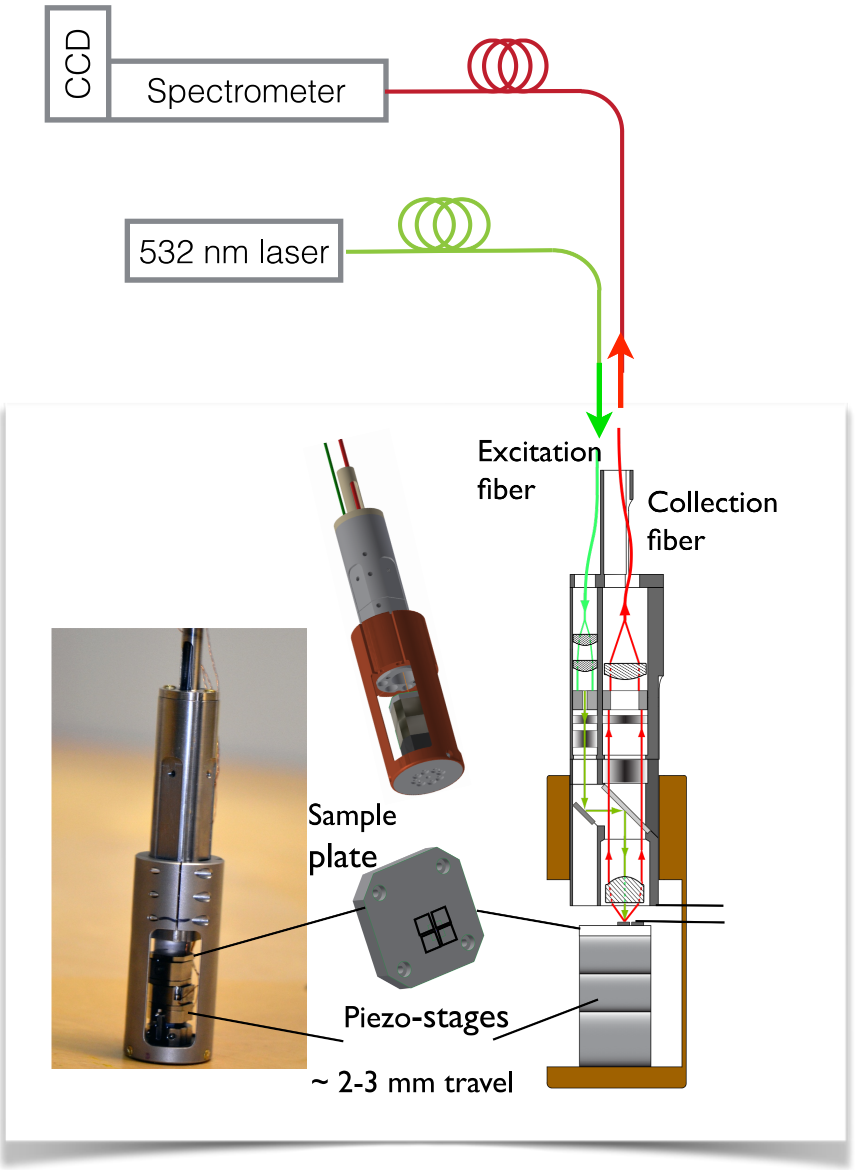
Open questions (motivation):

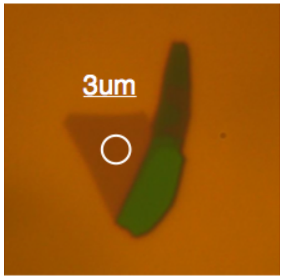
- **How to break the valley degeneracy and control the valley splitting?**
- **How to create and control the steady-state valley polarization**

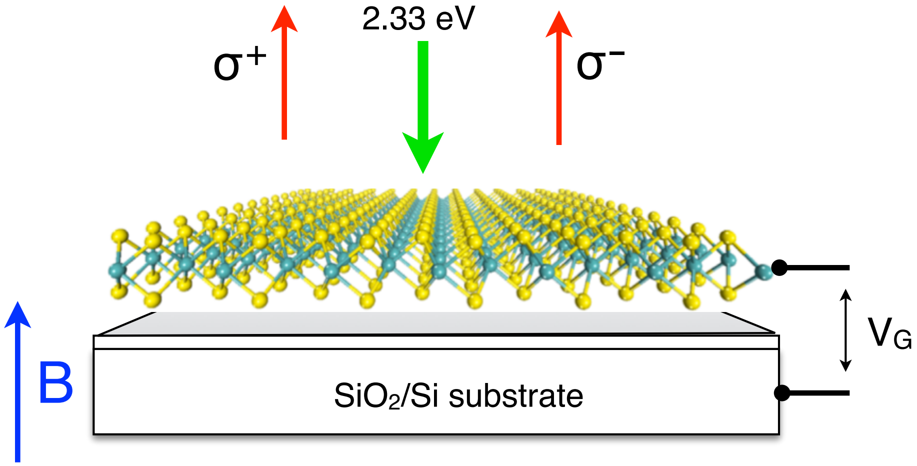
Answer (method):

- **Apply magnetic field and break the time reversal symmetry**

Experimental details

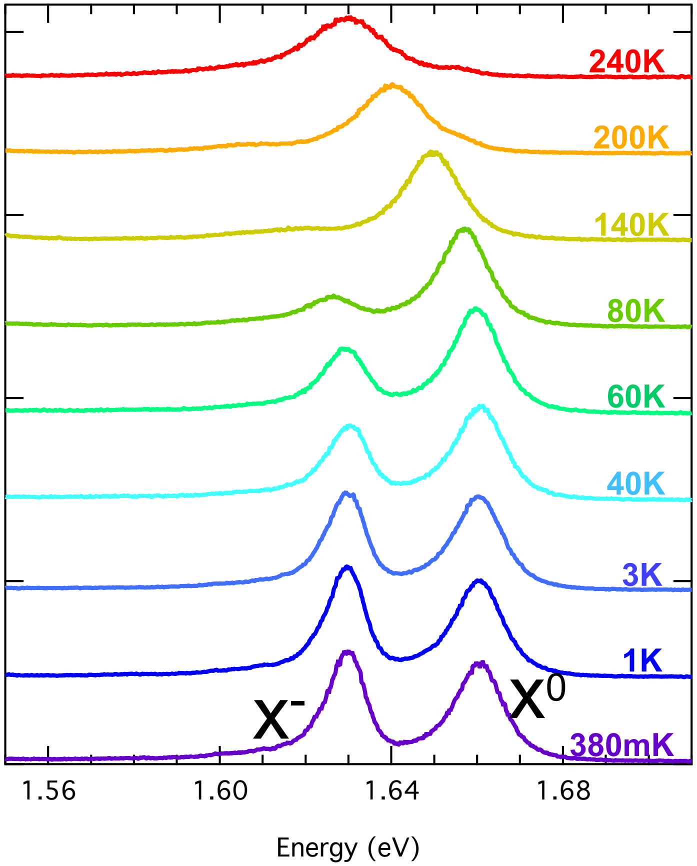




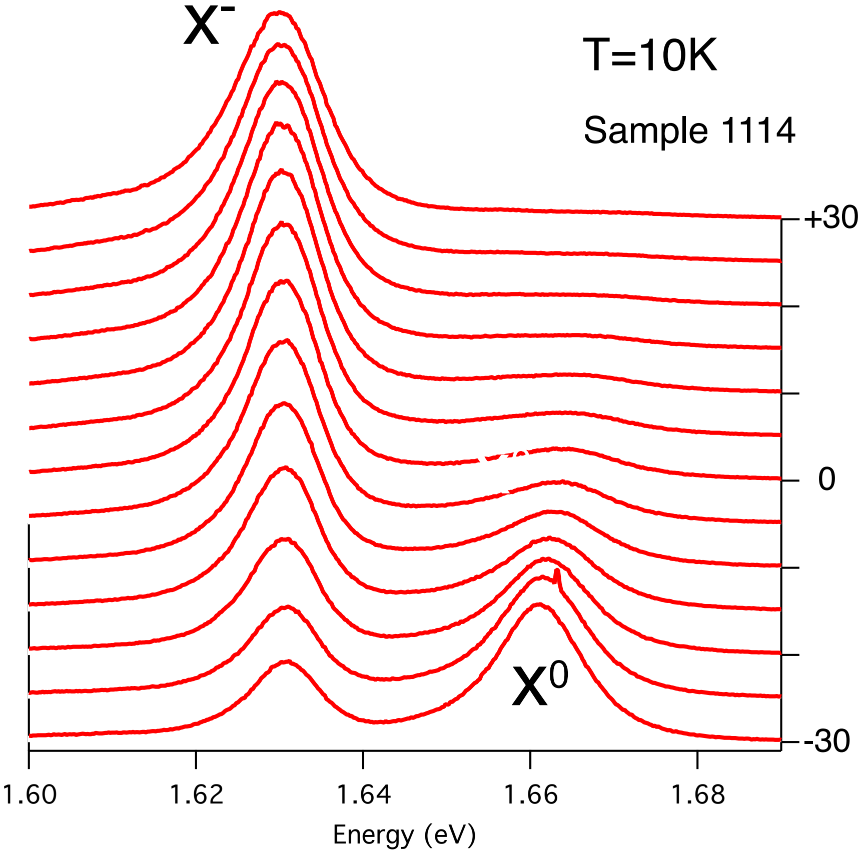


Gate control of neutral and charged excitons in a monolayer MoSe$_2$

Temperature dependence



Gate voltage dependence

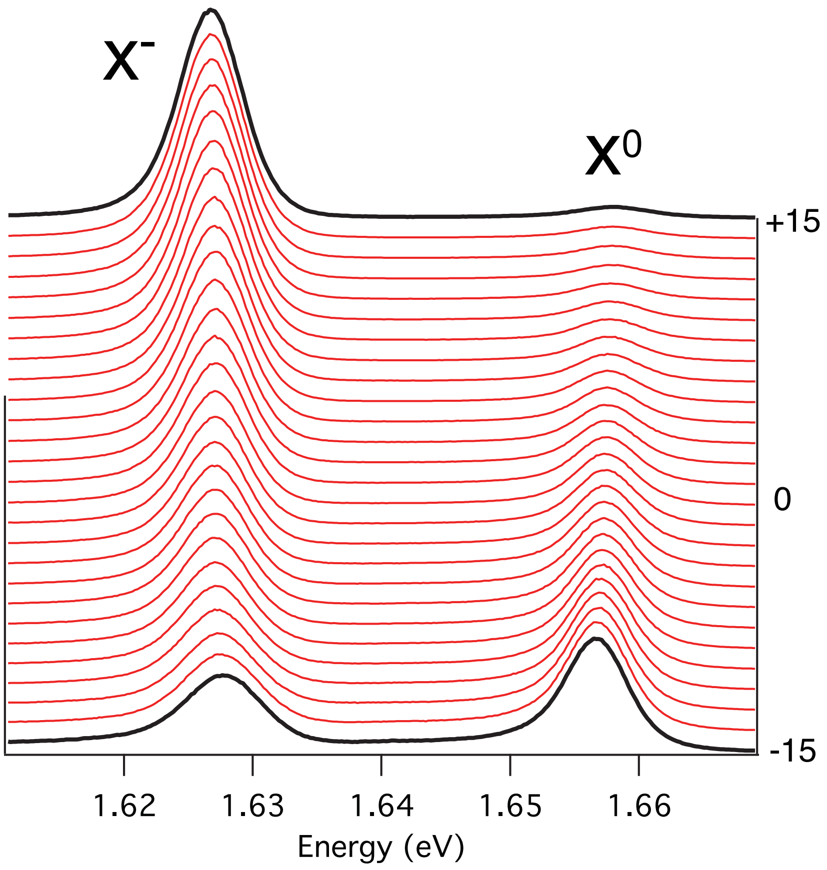


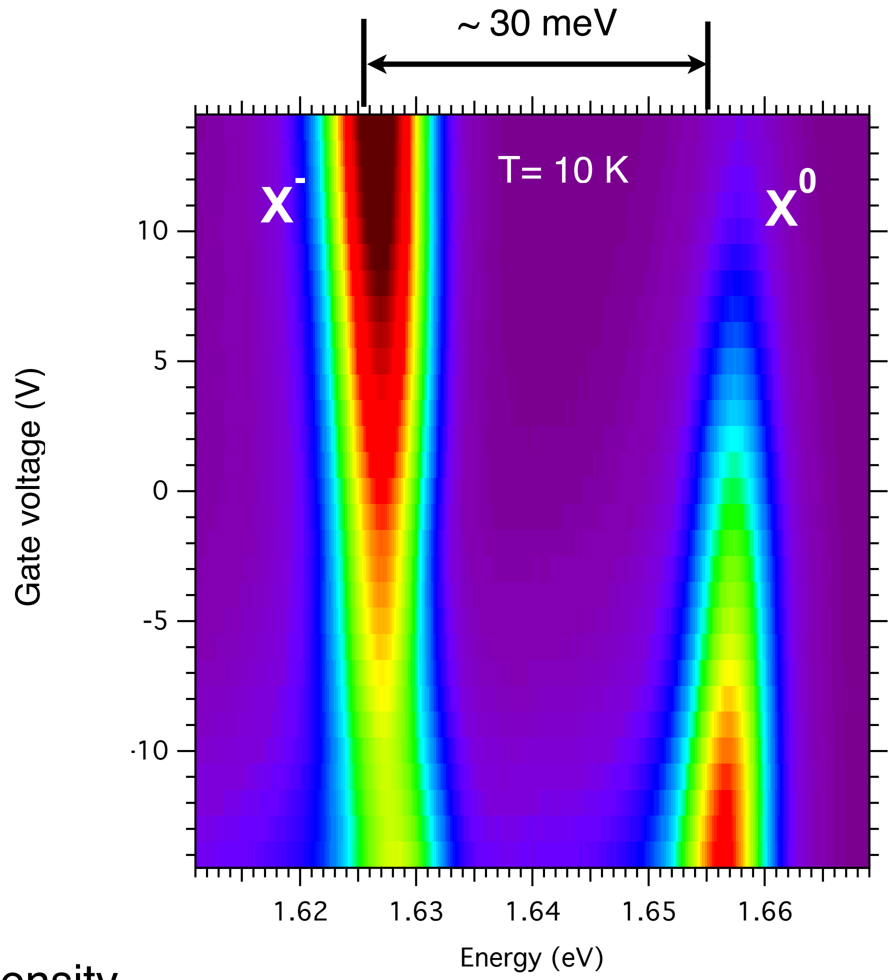
First shown by T.Heinz’ (MoSe$_2$) and X.Xu’s (MoSe$_2$) groups

Mak et al. Nature Mat., **12**, 207 (2013)

Ross et al. Nature Comm., 4:1474 (2013)

Zero-field PL vs gate voltage



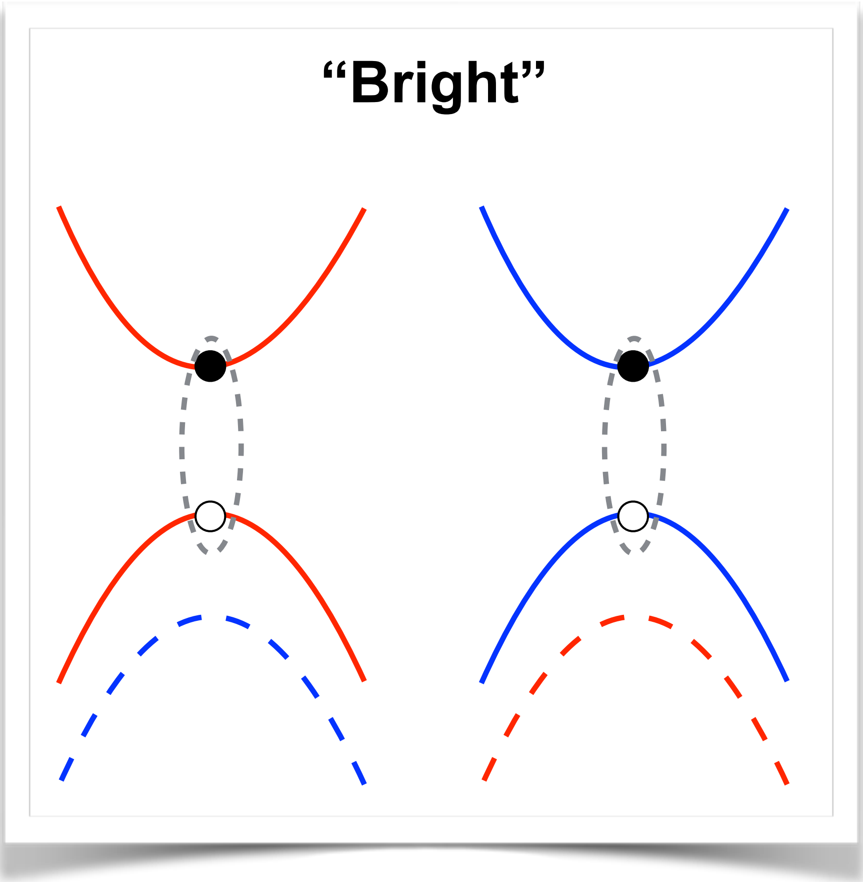


- Low-doping regime : X^- and X^0 have similar intensity
- High-doping regime : X^- dominates

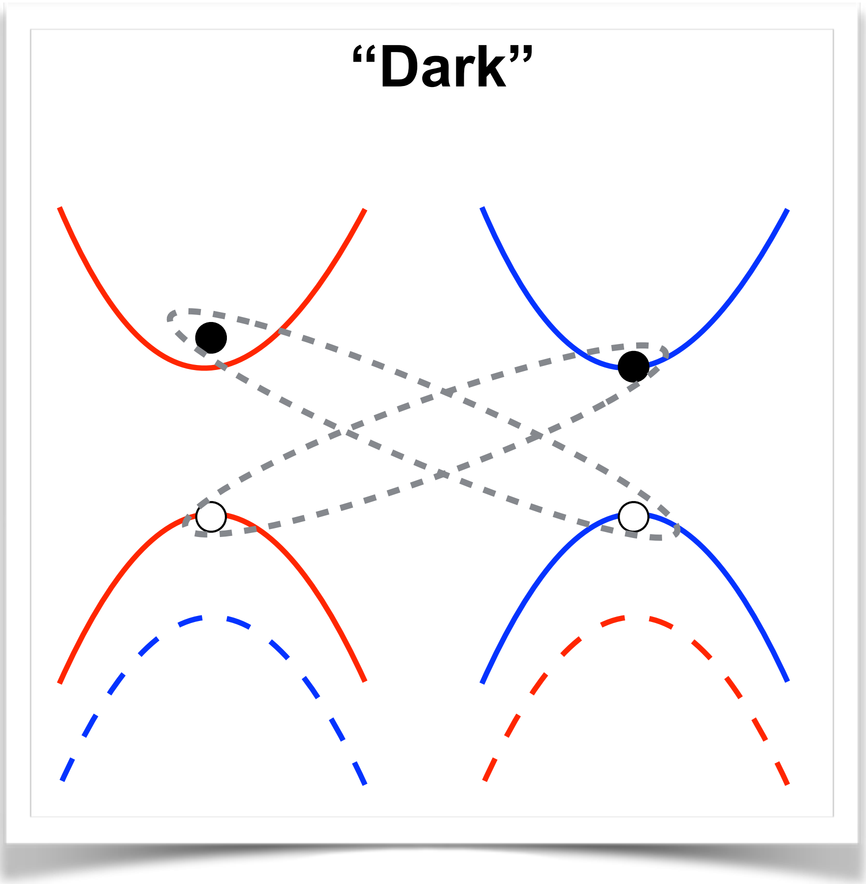
Sample 415

Excitons in a monolayer $MoSe_2$

<u>X0 exciton : neutral exciton</u>



“Bright”

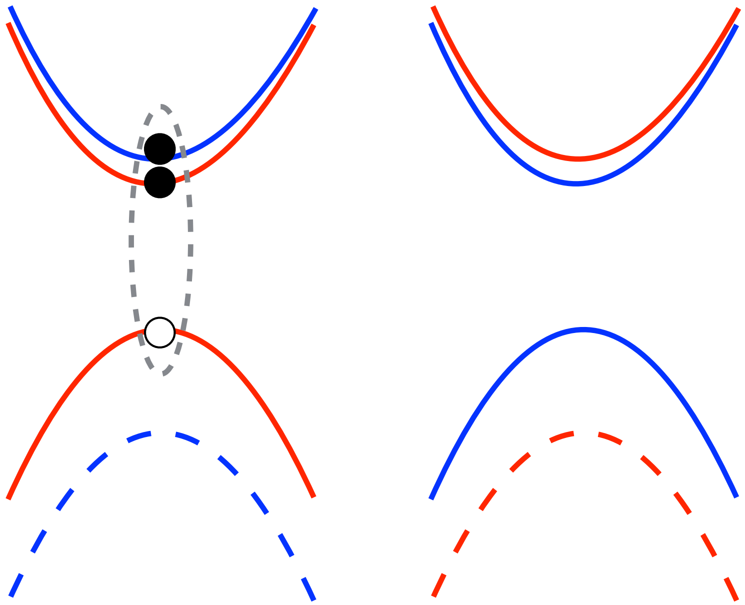


“Dark”

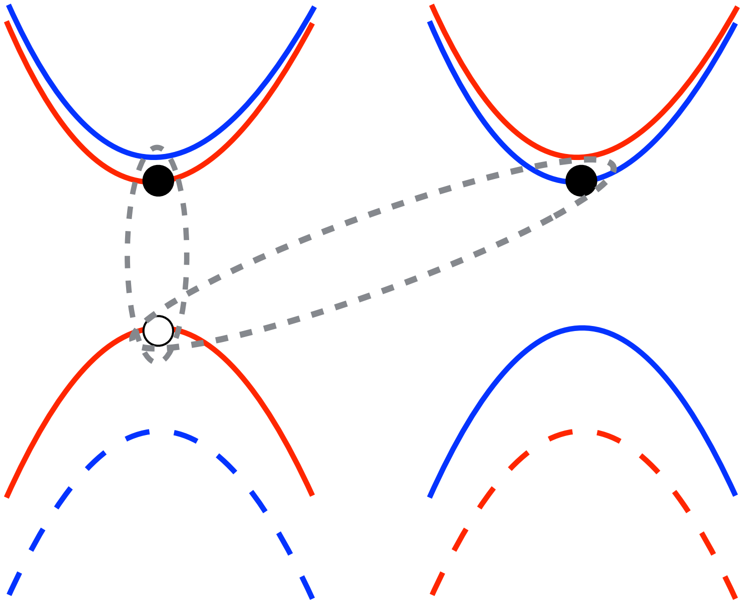
Excitons in monolayer $MoSe_2$

X- exciton : negatively charged trion

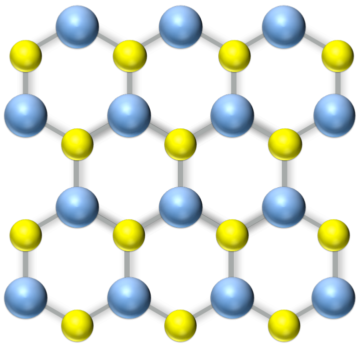
“Bright” intra-valley exciton

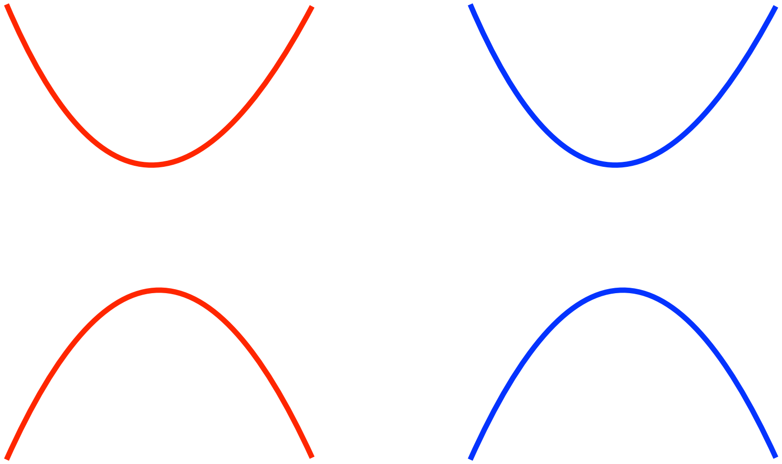


“Bright” inter-valley exciton



Valley Zeeman effect





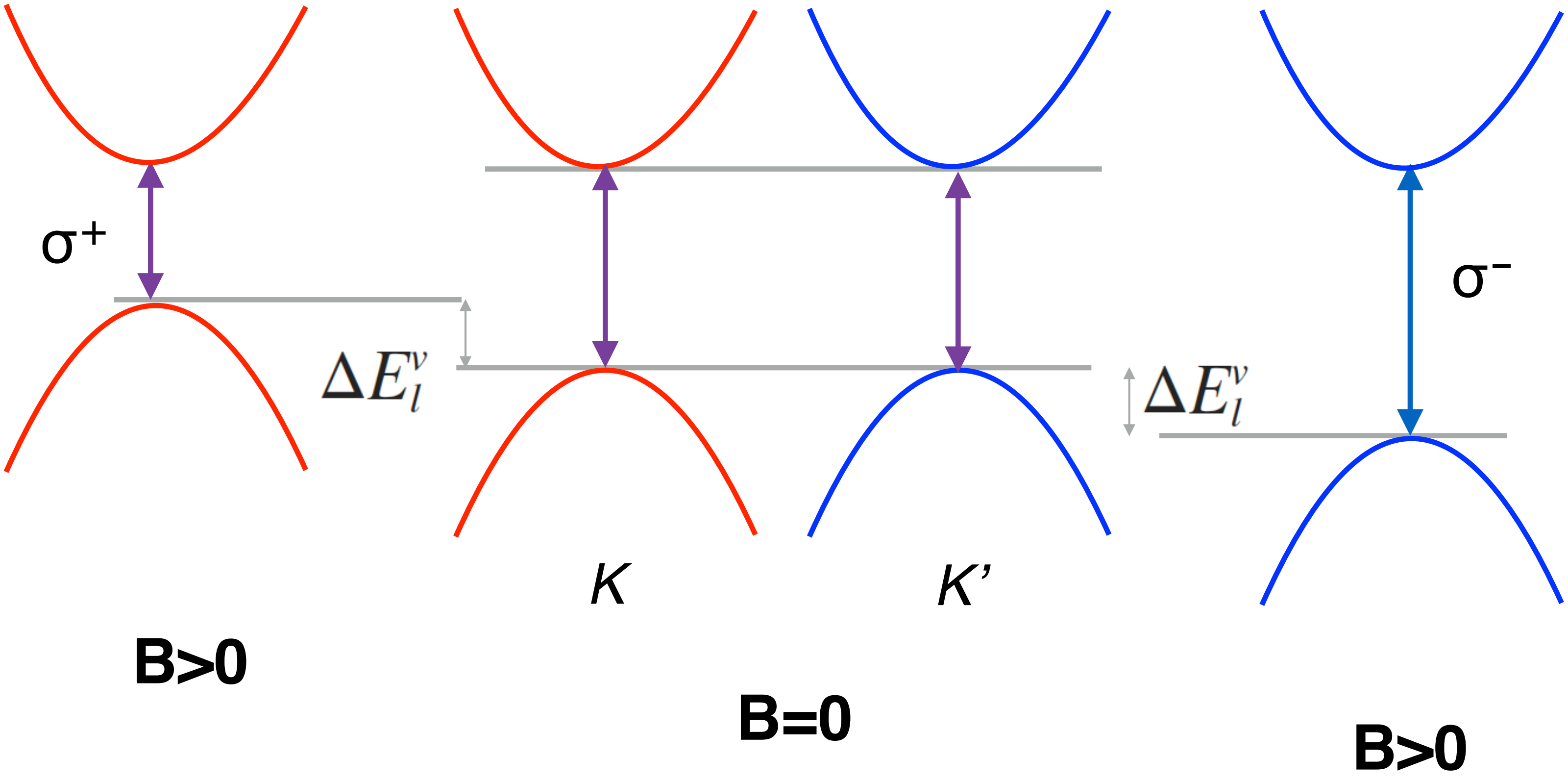
$$\Delta E_Z = E_s^{c/v} + \Delta E_l^{c/v} + \Delta E_k^{c/v}$$

	K valley		K’ valley	
	CB	VB	CB	VB
Spin	$-\mu_B$	$-\mu_B$	$+\mu_B$	$+\mu_B$
Atomic d-orbitals (intracellular)	0	$-2\mu_B$	0	$+2\mu_B$
Phase winding of Bloch function (intercellular) , $\alpha = m_0/m_{C,V}$	$-\alpha\mu_B$	$-\alpha\mu_B$	$+\alpha\mu_B$	$+\alpha\mu_B$

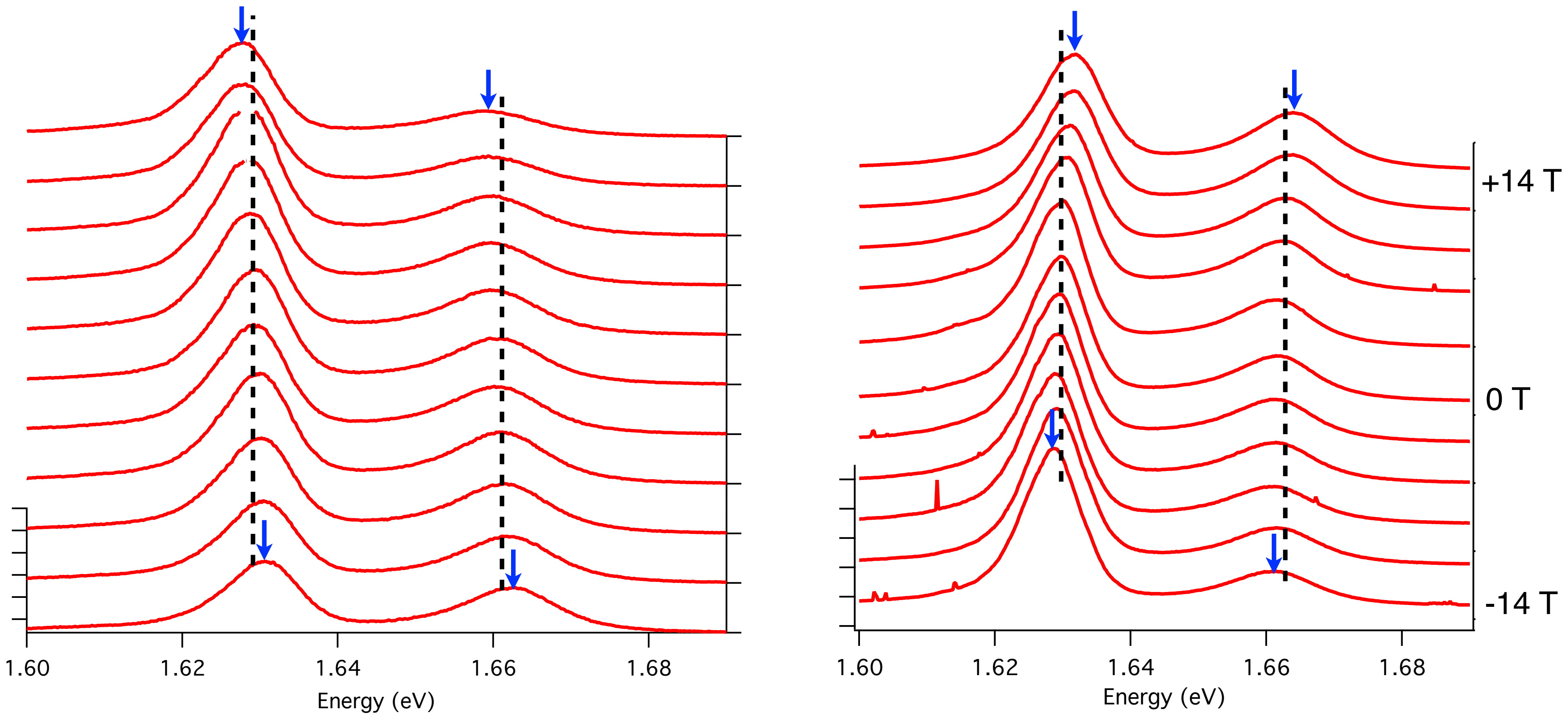
W. Yao, et al. PR B, 2008; X. Xu, et al. Nature Phys., 2014, T. C. Berkelbach et al. PRB, 2013

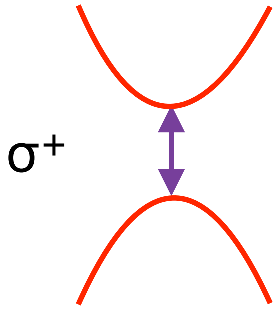
Valley Zeeman effect

$$\Delta E_Z = \cancel{E_s^{c/v}} + \Delta E_l^{c/v} + \cancel{\Delta E_k^{c/v}} \approx 4\mu_B B$$

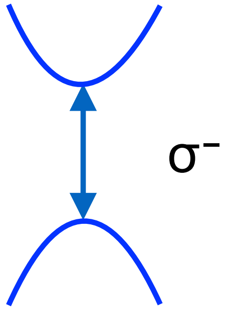


Valley Zeeman effect in a monolayer $MoSe_2$: low carrier density

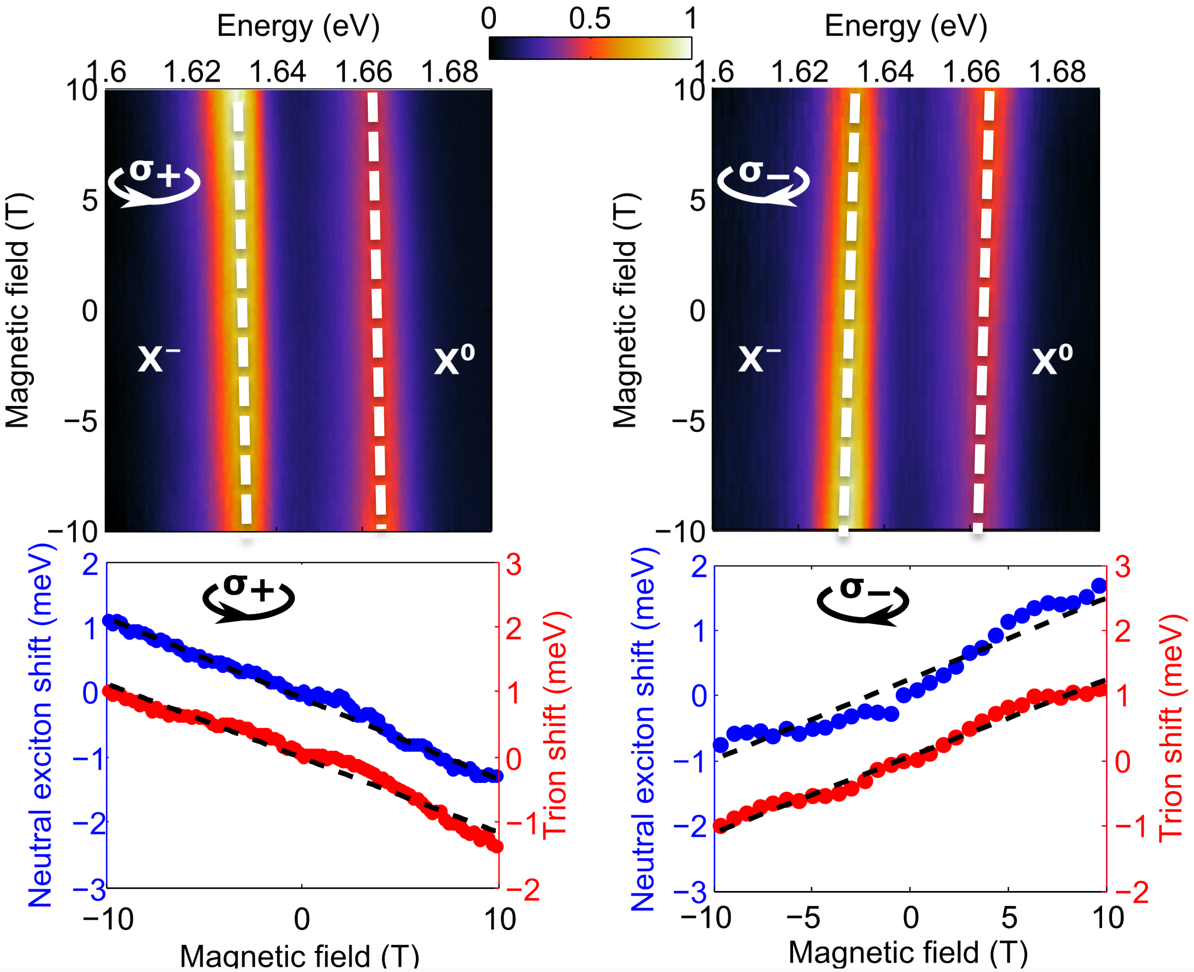




Valley degeneracy is lifted



Zeeman shift of exciton peaks



	Experimental slope
X^0	± 0.12 meV/T
X^-	± 0.12 meV/T

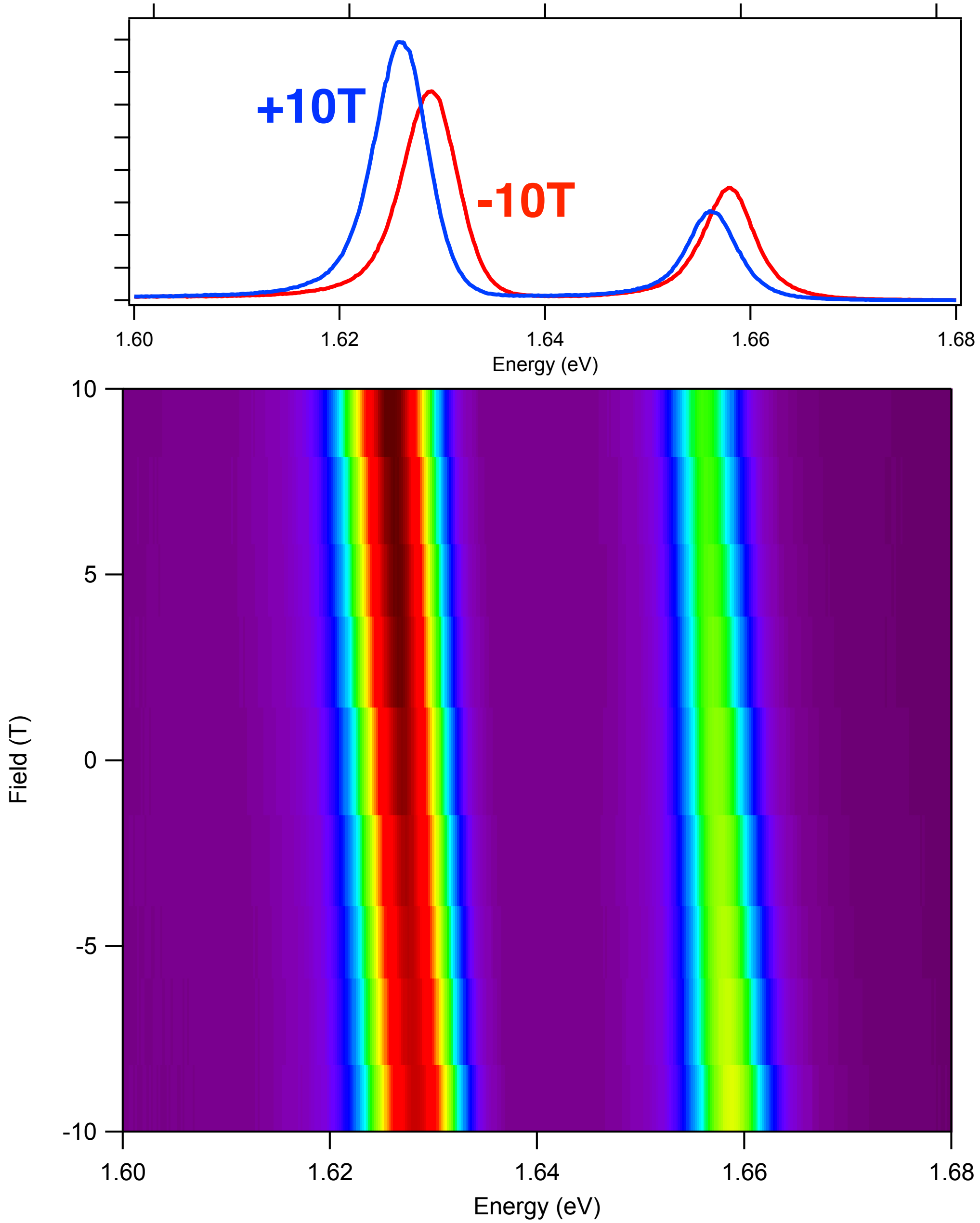
$$2\mu_B = 0.116\ \text{meV/T}$$

$$E_{X0} = E^c - E^v - E^b_{X0}$$
$$E_{X-} = E^c - E^v - E^b_{X0} - E^b_{X-}$$

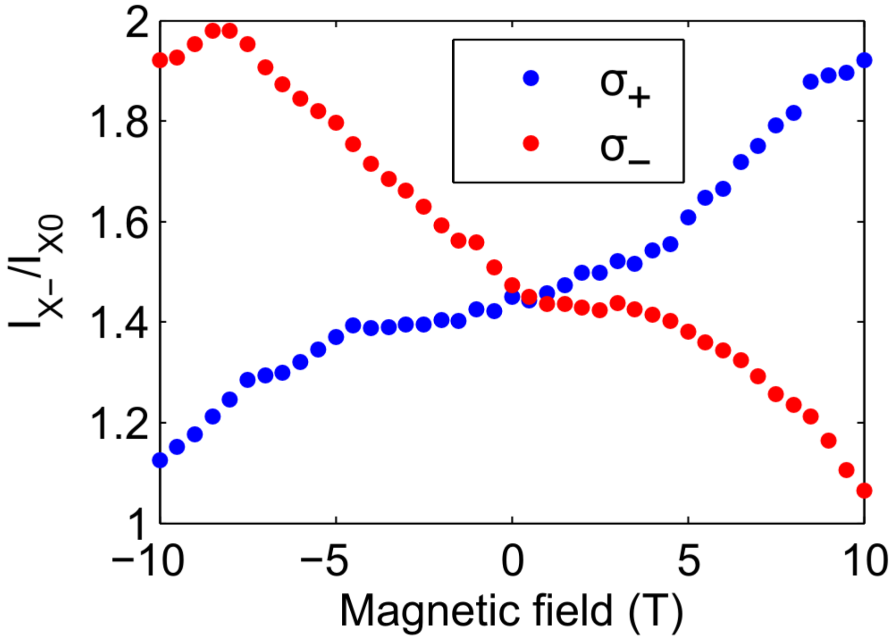
- **Valley degeneracy is lifted due to the contribution from the valence band atomic orbitals, resulting in total Lande factor of 4.1**
- **Binding energies are not influenced by the magnetic field at low densities**

Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014)

Variation of relative intensity

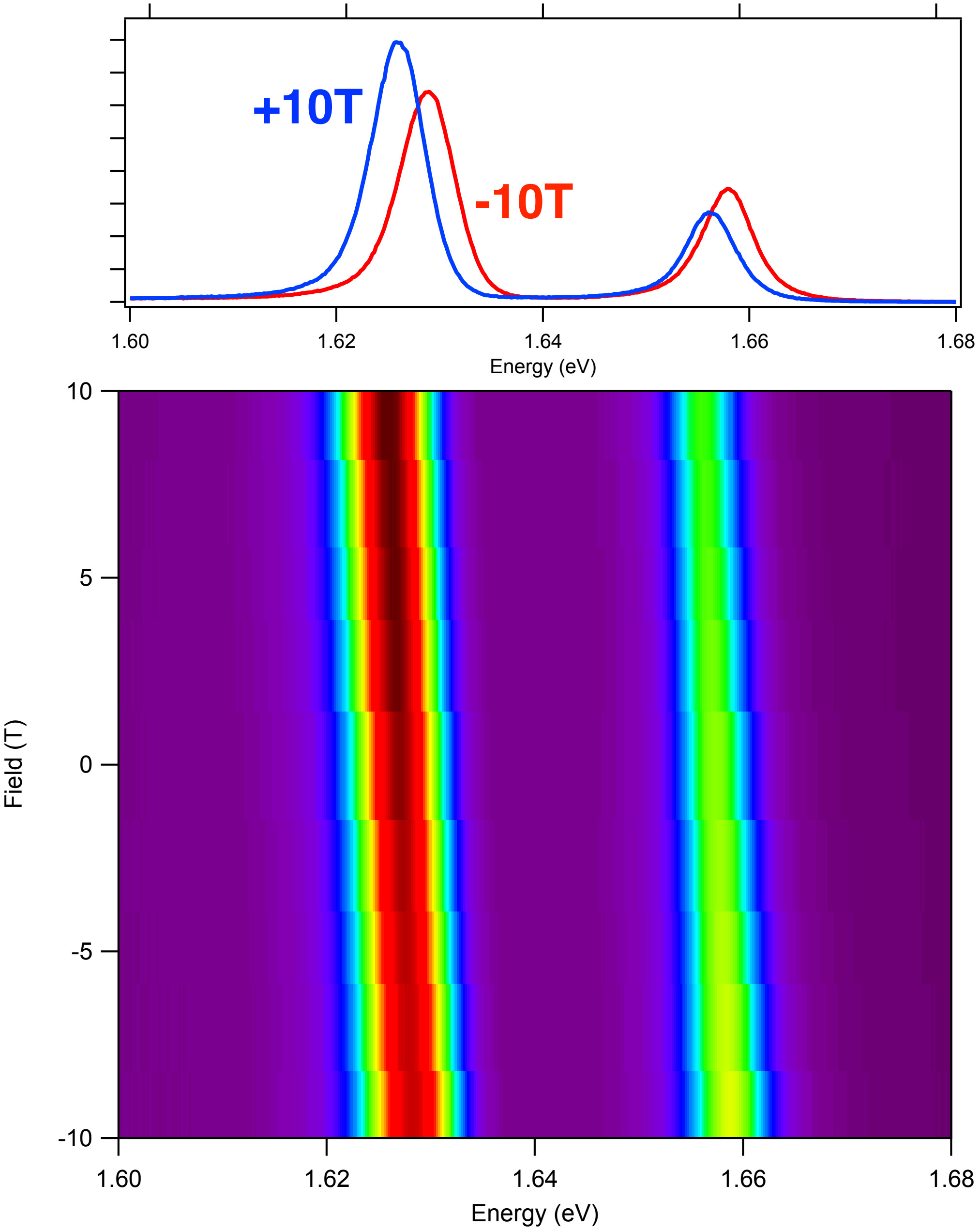


- **The relative intensities of X– and X0 varies monotonically with magnetic field**
- **The trend is reversed for the opposite valleys**



Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014)

Trion configuration



- The relative intensities of X– and X0 varies monotonically with magnetic field
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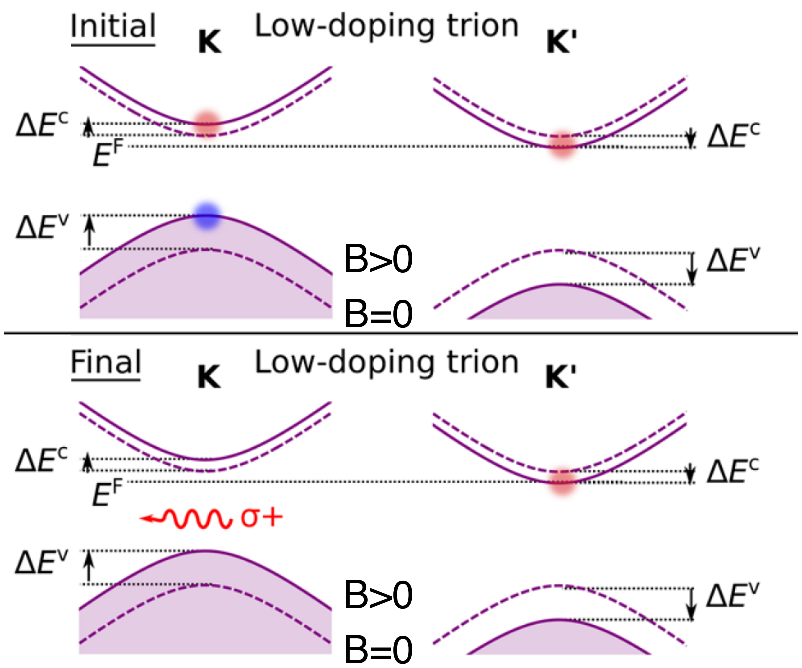
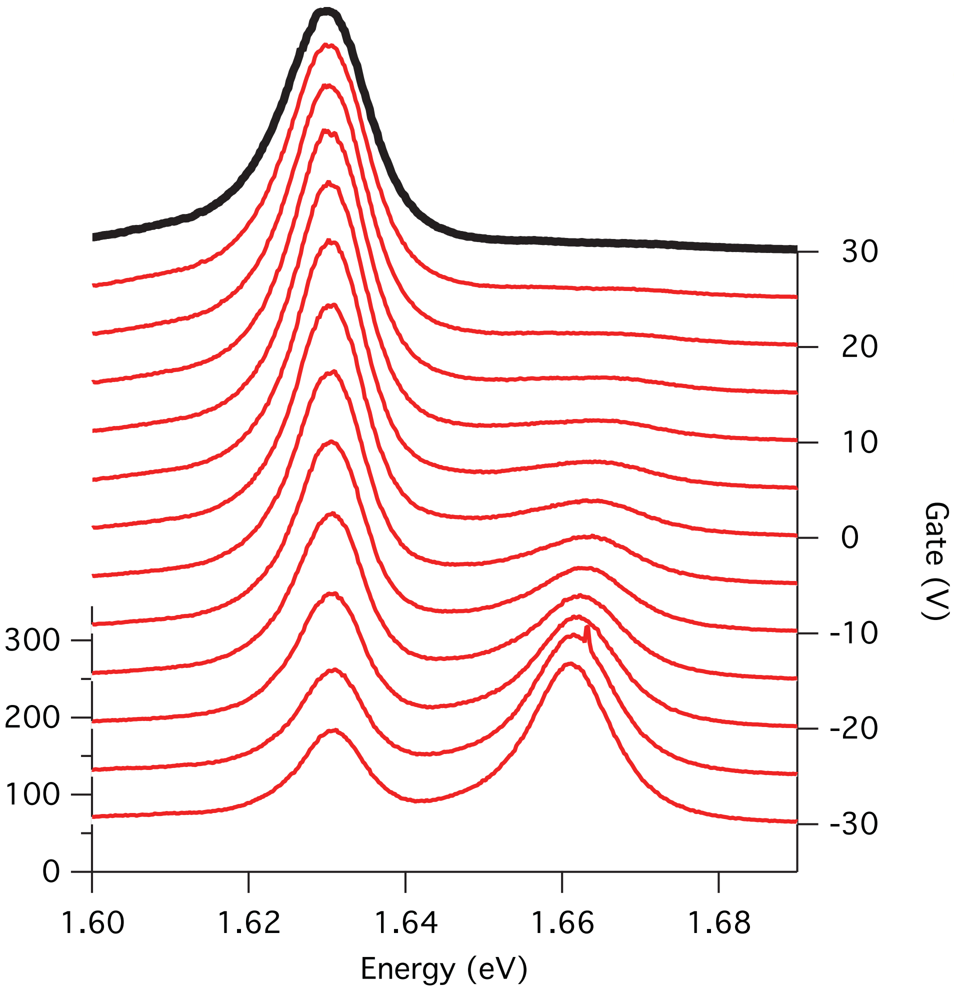
Inter-valley trion

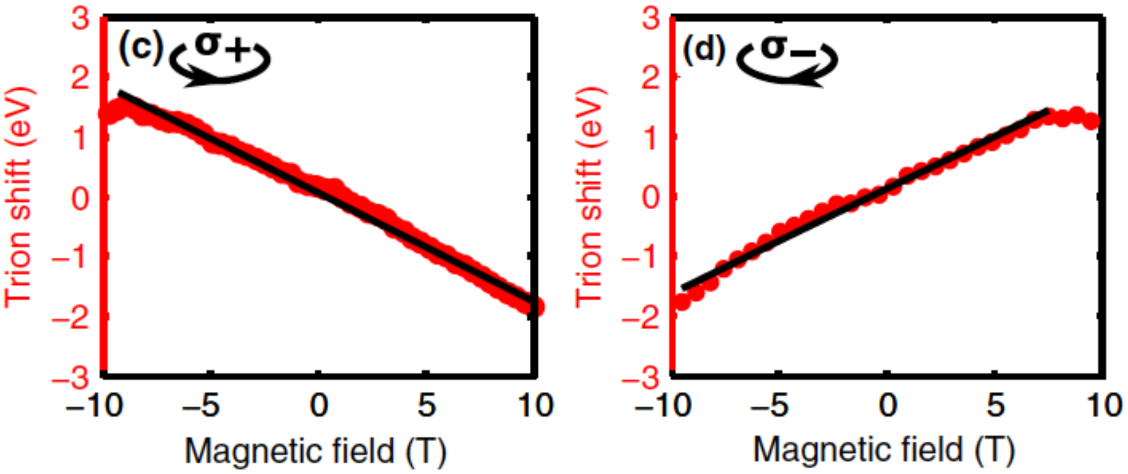


Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014)

Theory: H. Yu, et al. Nat. Commun. **5**, 3876 (2014).

Trion emission at high carrier density

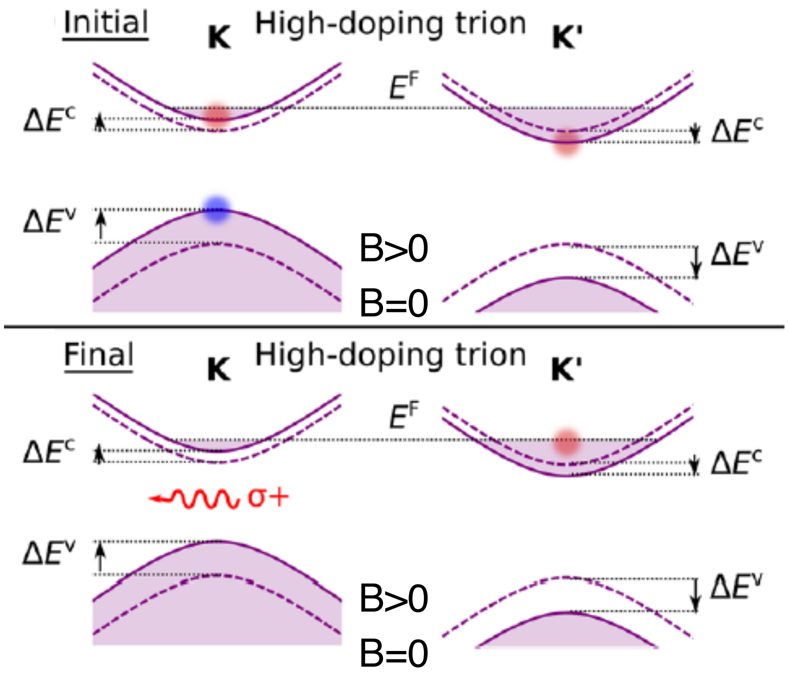


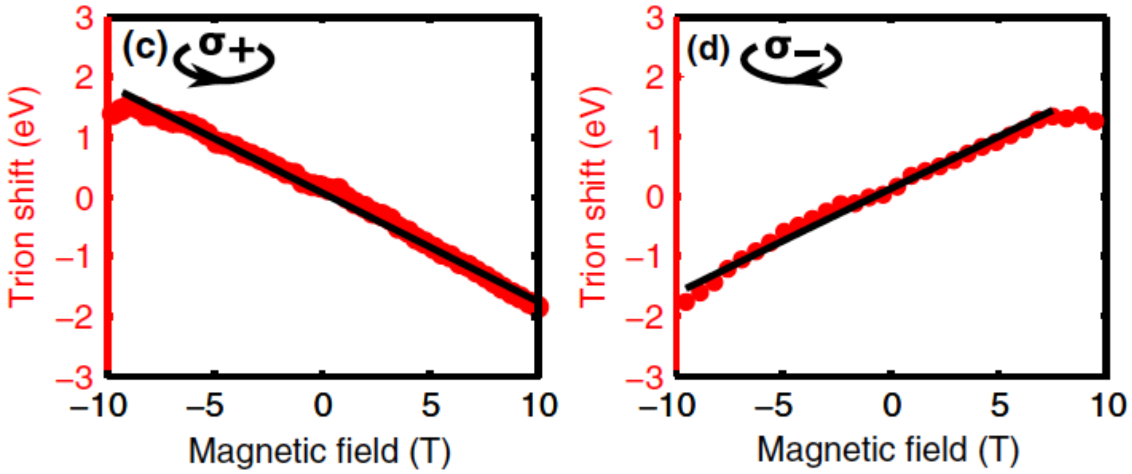


- The slope is **0.18 meV/T**, i.e. **50% increase** compared with **0.12 meV/T** in the regime of low carrier density
- Estimated carrier density of 3×10^{12} would cause the Fermi level to be ~10meV above the CB edge

Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014)

Trion emission at high carrier density





- At $E_F > E_C$, the trion Zeeman shift is expected to follow total VB contribution only ($5\mu_B$), which would result in **0.29 meV/T.**

???

- The slope is **0.18 meV/T**, i.e. **50% increase** compared with **0.12 meV/T** in the regime of low carrier density
- Estimated carrier density of 3×10^{12} would cause the Fermi level to be ~10meV above the CB edge

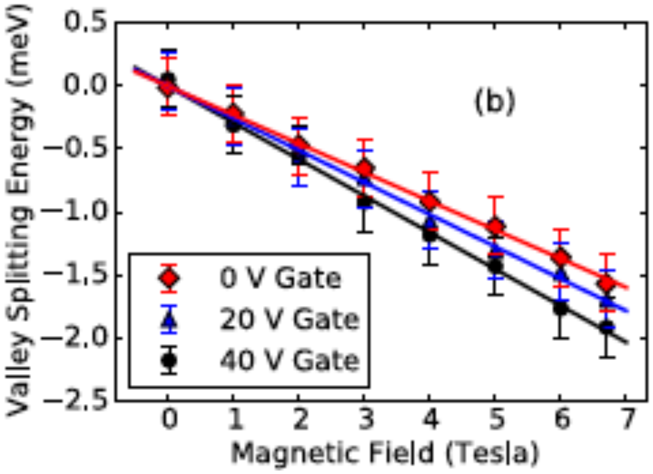
Li, Y., Ludwig, J. et al. Phys. Rev. Lett. **113**, 266804 (2014)

Related works on valley Zeeman effect in monolayer TMDs

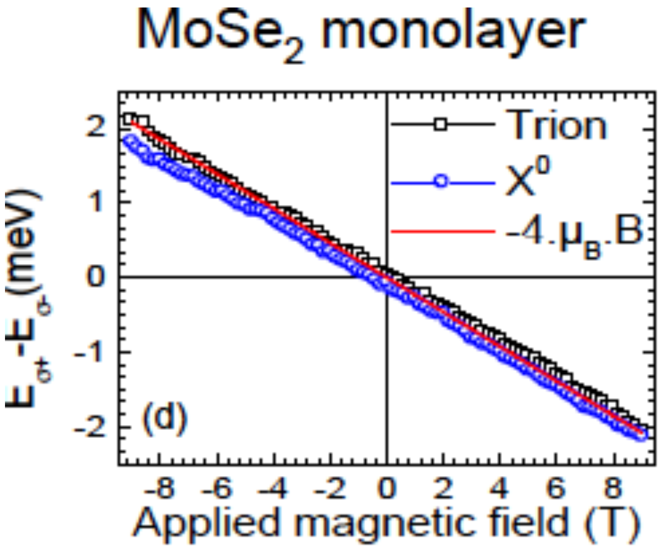
Aivazian, G., et al. (Univ. of Wash.)
Magnetic control of valley pseudospin in monolayer WSe_2.
Nature Physics, **11**(2), 148 (2015)

Srivastava, A.,et al. (ETH, EPFL)
Valley Zeeman effect in elementary optical excitations of monolayer WSe2.
Nature Physics, **11**(2), 141 (2015)

MacNeill, D., et al. (Cornell)
Breaking of Valley Degeneracy by Magnetic Field in Monolayer MoSe2.
Physical Review Letters, **114,** 037401 (2015)



Wang, G., et al. (Toulouse, Ioffe)
Magneto-optics in transition metal diselenide monolayers.
arXiv:1503.04105v1 (2015)



Valley splitting and polarization in monolayer $MoSe_2$

- **Splitting of K/K' valleys by application of perpendicular magnetic field (tuning valley DoF)**
- **Charge imbalance in different valleys for doped samples – creation of steady-state valley polarization**
- **Intervalley configuration is the lower energy state for the trion**
- **Variation in the trion emission energy X-(B) with at high doping (call for more experimental and theoretical studies)**