Novel optoelectronic properties of 2D materials:

Ultrafast Optical Studies of Valley States in 2D Transition Metal Dichalcogenides

Apr. 12th, 2017

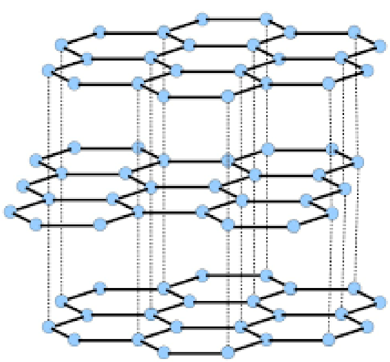
Jonghwan Kim

Department of Physics, UC Berkeley & LBNL

Department of Materials Science & Engineering, POSTECH

Atomically Thin 2D Crystals

Crystal structure



Graphene isolation

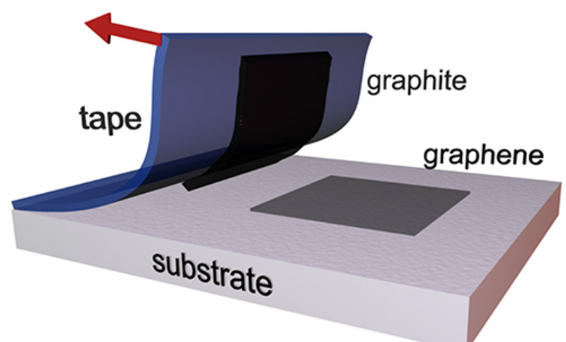
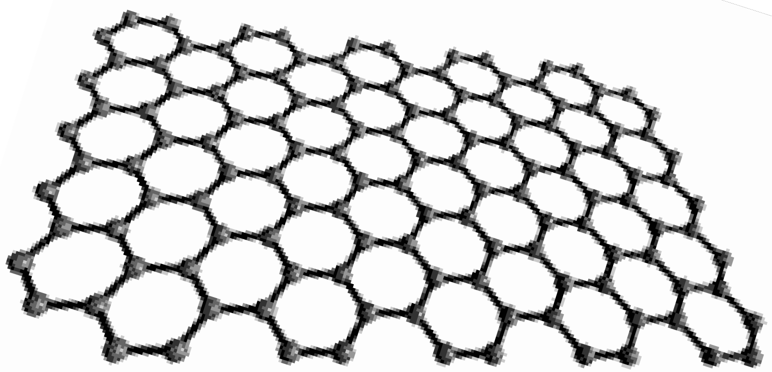


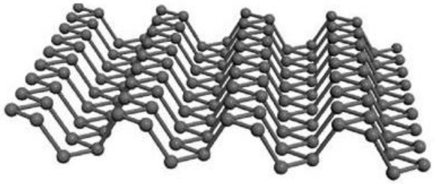
Image from GLAB

Atomically Thin 2D Crystals

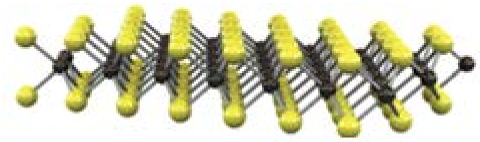


Graphene

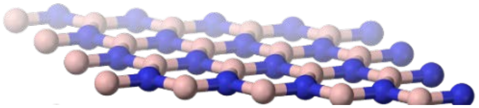
Single atomic layer of carbon



Phosphorene



MoS_2



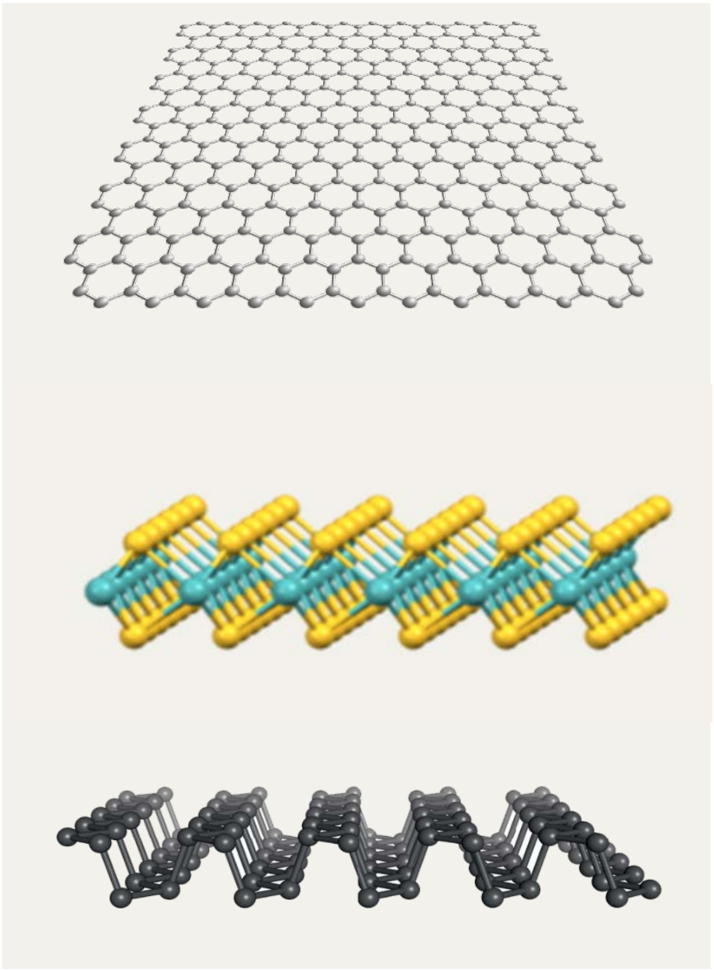
h-BN

Semiconductors

Metal: $NbSe_2$, TaS_2, WTe_2

Novel electronic states in the exact 2D materials!

Atomically Thin 2D Crystals



Graphene

Massless Dirac fermion
(Record high mobility)

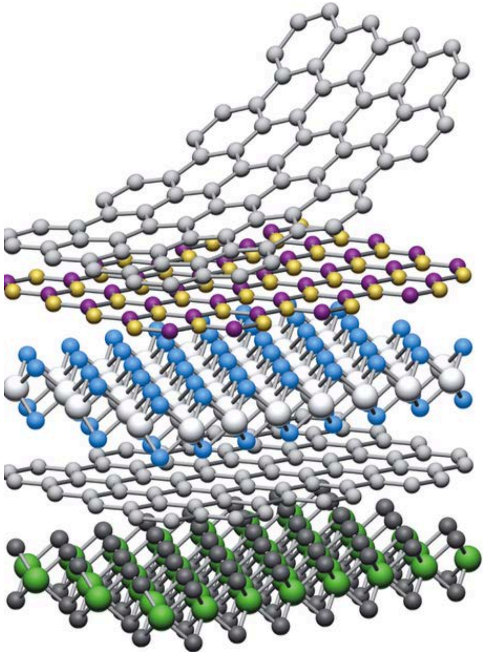
MoS_2

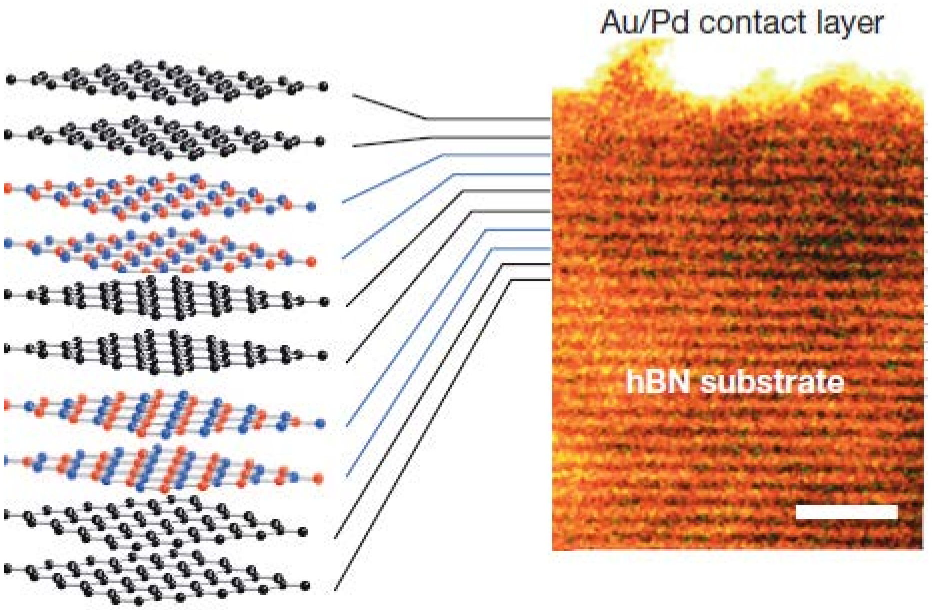
Valley electronic state
(Future electronic memory)

Phosphorene

Interlayer interaction
(Tunable direct bandgap
visible to zero-band gap)

van der Waals 2D Heterostructures





<u>Atomically engineered material for new physics</u>

Geim, Nature, **499**, 419 (2013)

Outline

1. New information carrier

   : Valley state in 2D transition metal dichalcogenides (TMD)

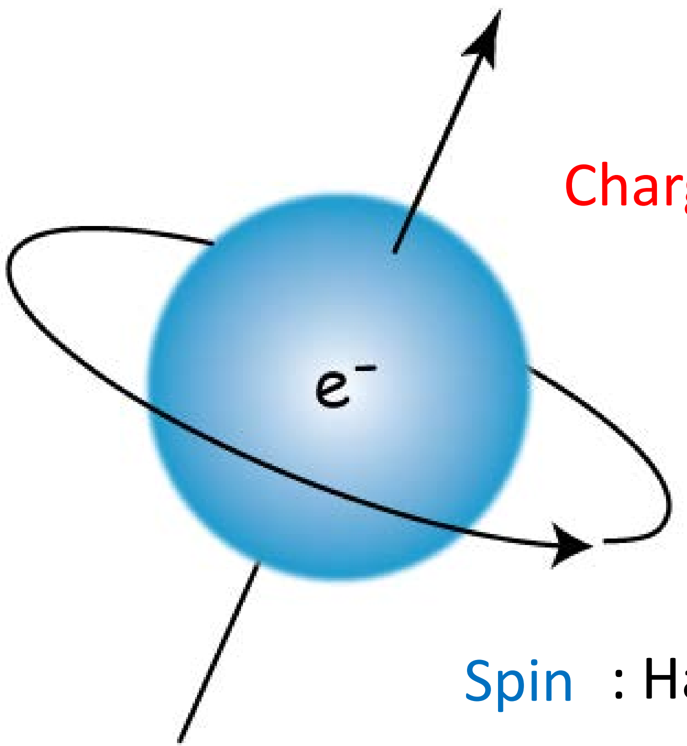
2. Valley information manipulation

   : Ultrafast and strong pseudomagnetic field in TMD monolayer

3. Valley information lifetime

   : Ultralong valley polarization in TMD heterostructures

Degree of Freedom in Electrons

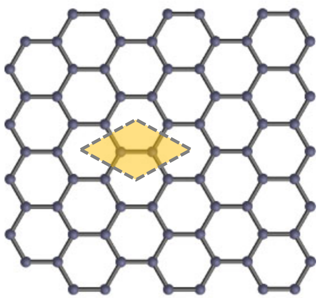


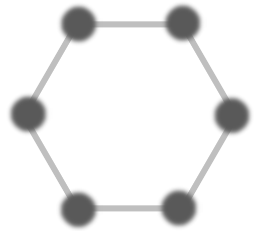
Charge : Field effect transistor

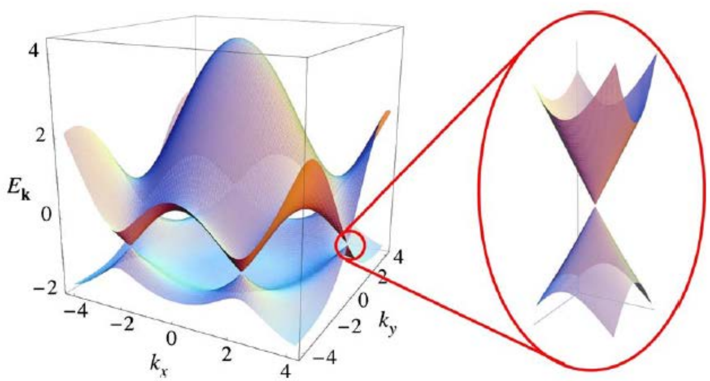
Spin : Hard disk drive

Valley Degree of Freedom

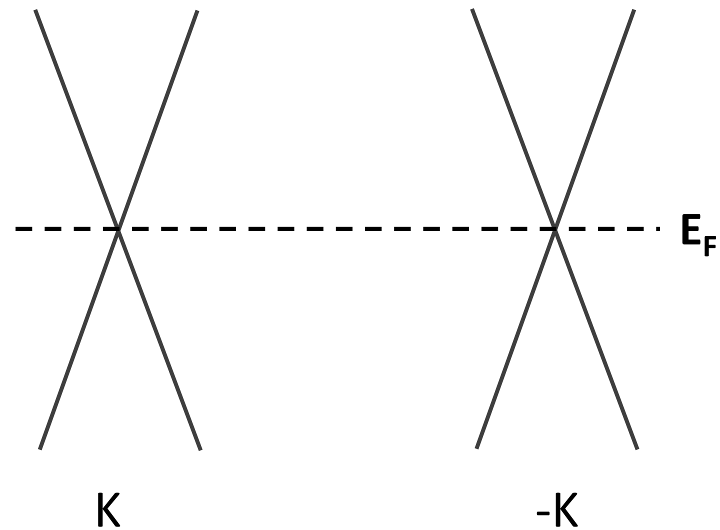
Graphene





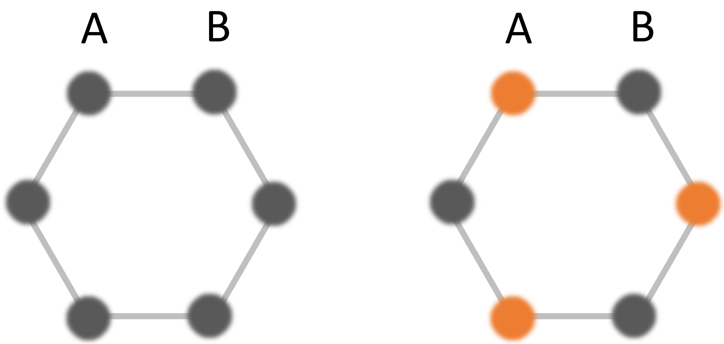


Time-reversal pair



Access to Valley State

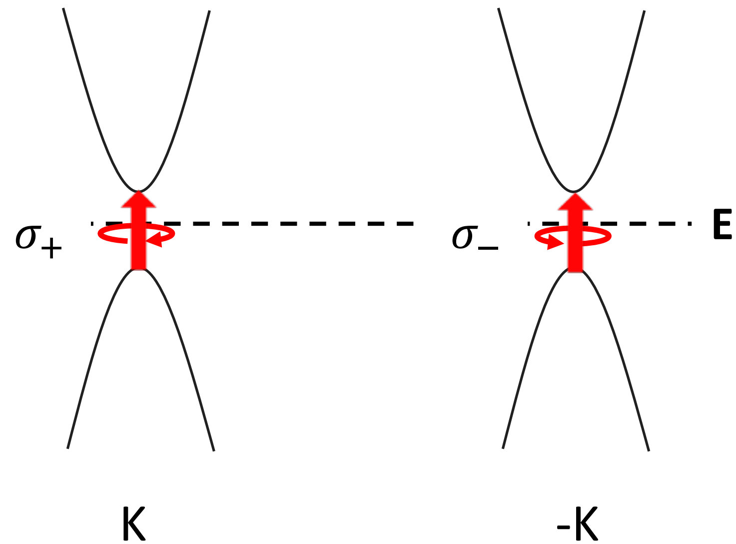
Inversion symmetry: Zero magnetic moment



Time reversal symmetry : $\mu_K = -\mu_{-K}$

Inversion symmetry : $\mu_K = \mu_{-K}$

$\mu_K = 0, \mu_{-K} = 0$

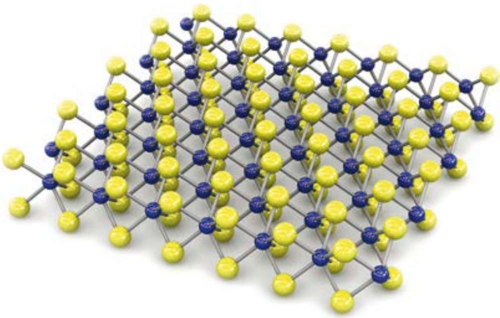


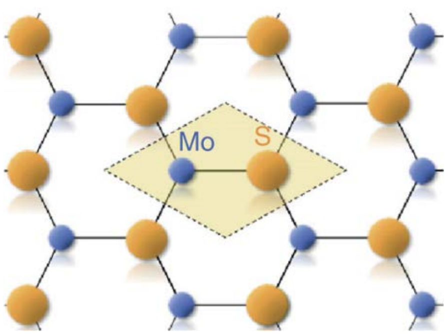
$\mu_K = -\mu_{-K} \neq 0$

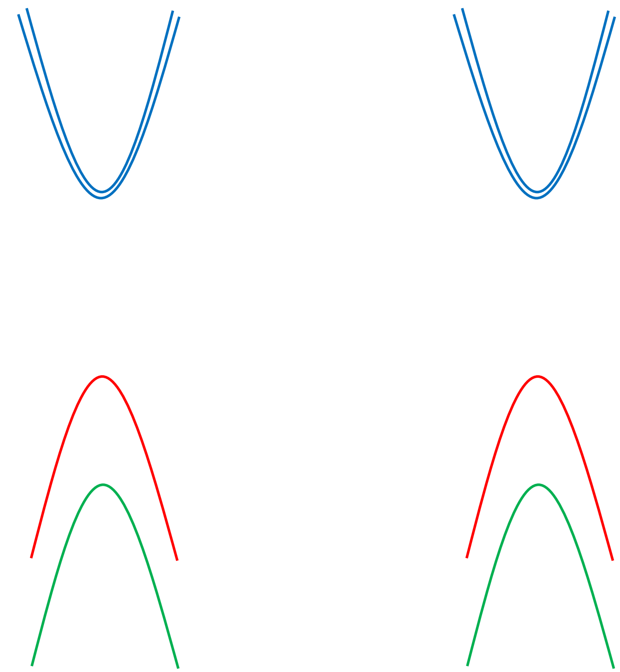
Helicity-dependent light absorption

TMD Monolayer

MX_2: M = Mo, W; X = S, Se
MoS_2, $MoSe_2$, WS_2, WSe_2







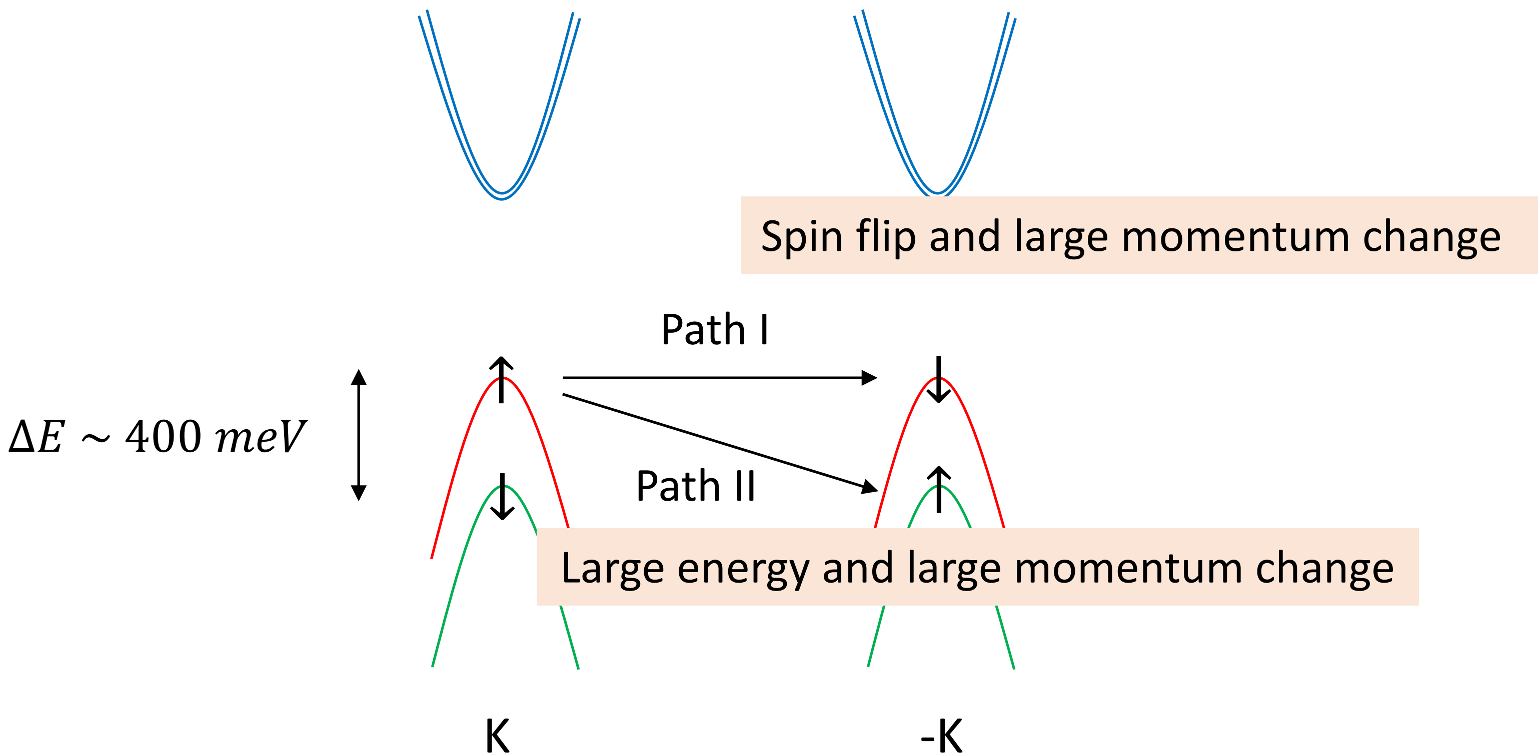
K

-K

Explicitly broken inversion symmetry

Valley State in 2D TMD

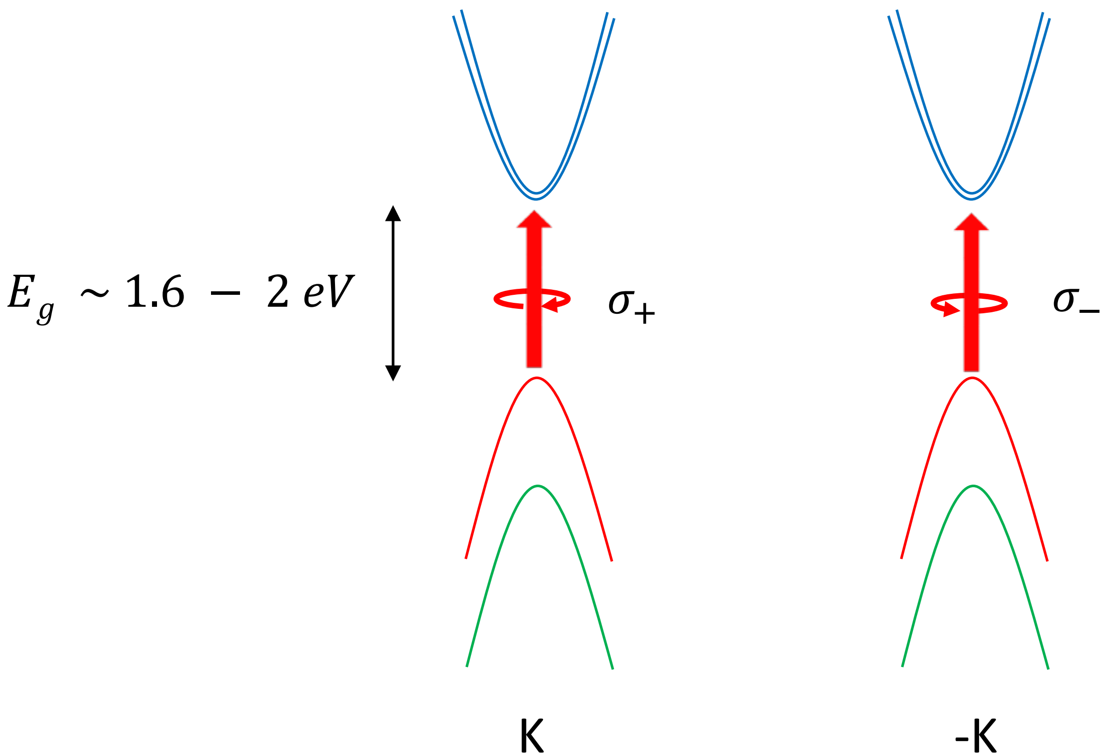
Strong SO coupling: Spin-Valley locking



Possibly ultralong spin/valley lifetime!

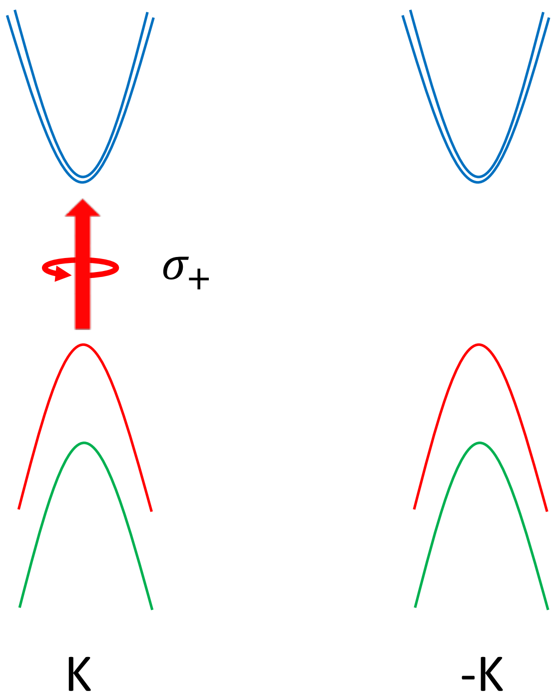
Valley State in 2D TMD

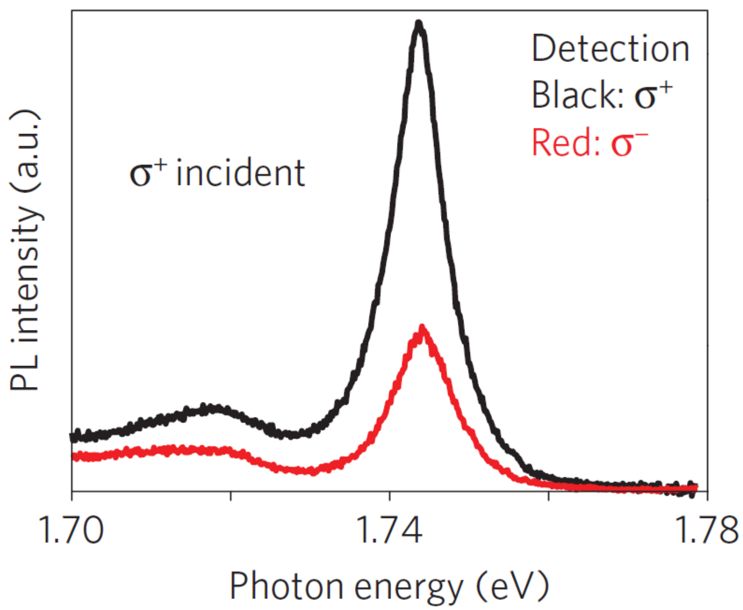
Direct bandgap semiconductor at (near) visible frequency



Convenient valley control with helicity of visible photon

Valley Polarization Control

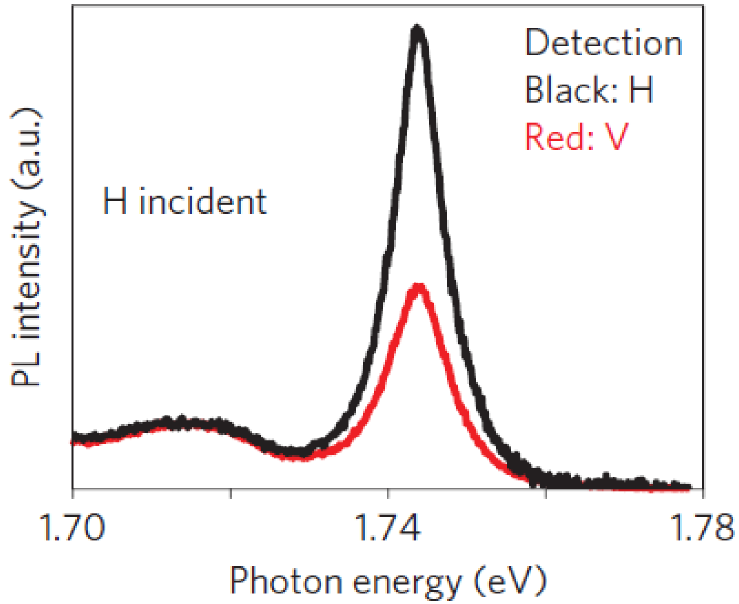


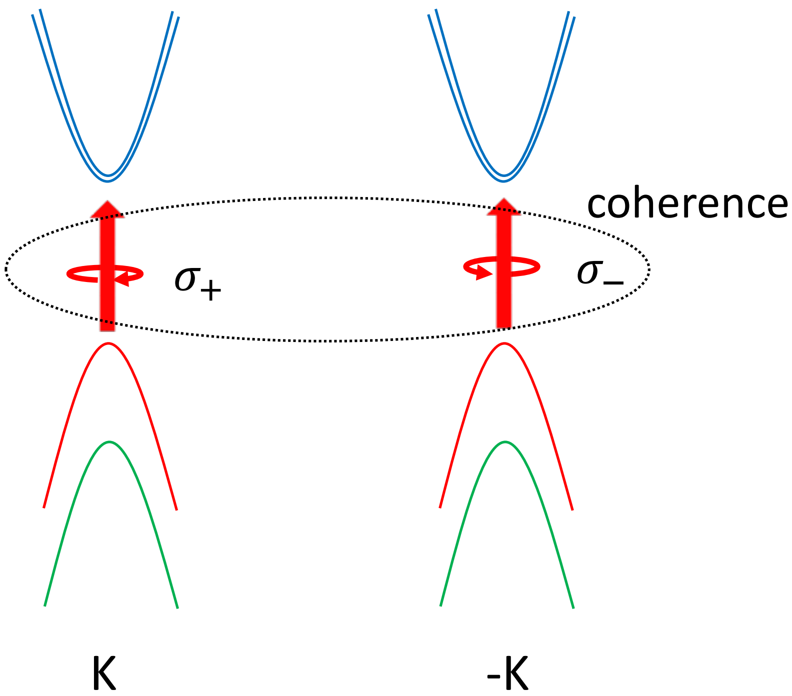


Heinz (Stanford), Cui (HKU), Xu (U. Washington), Feng (PKU)

Review: X. Xu, et. al., Nature Physics **10**, 343 (2014)

Valley Coherence Control

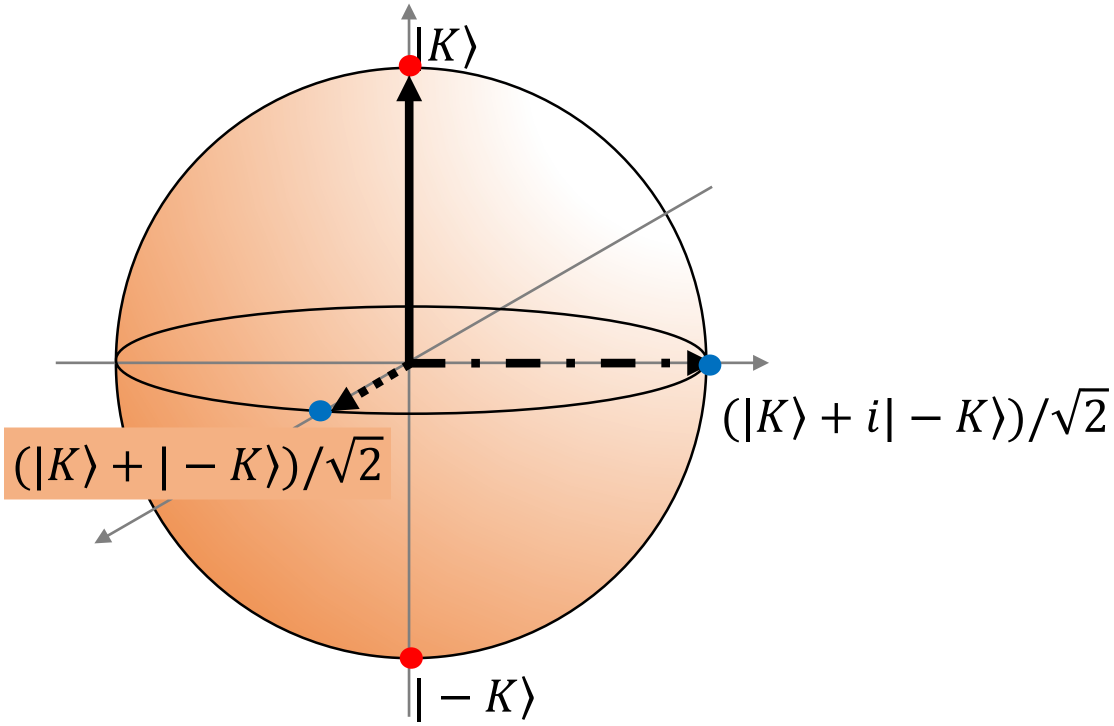




X. Xu, et. al., Nature Physics **10**, 343 (2014)

Valley Information

Bloch sphere for coherent valley polarization



Write: Polarization-controlled photoexcitation, spin-polarized carrier injection

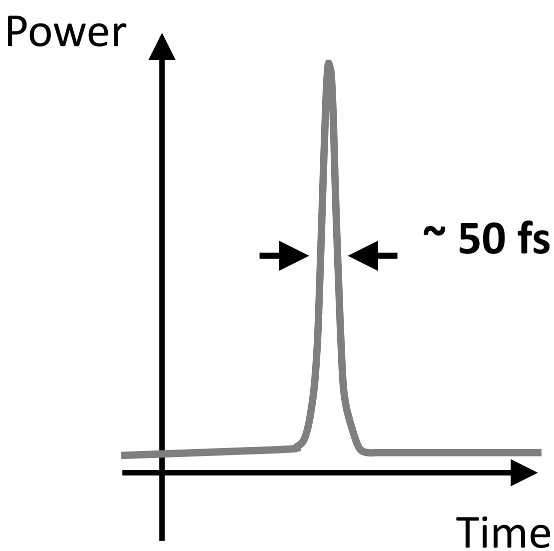
Read: Photoluminescence, circular dichroism, valley Hall effect

1. Manipulation of valley information?

2. Lifetime of valley information?

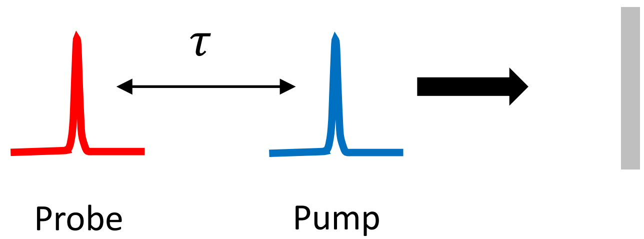
Femtosecond Optics

Femtosecond optical pulse



1. Ultrafast time resolution: fs time resolution

atomic motion : $\Delta t \sim$ 1 nm / 1000 m/s = 1 ps



2. Ultra high peak power

peak power : 1 mJ / 100 fs = 10 GW
(note. 4 GW for nuclear power plant)

Using light to control matter

Nonlinear optical phenomena

3. Ultra broadband tunability

Terahertz,	infrared,	visible	to UV
Free electron Superconductor	Phonon	Electronic transition	Photoemission

Outline

1. New information carrier

: Valley state in 2D transition metal dichalcogenides (TMD)

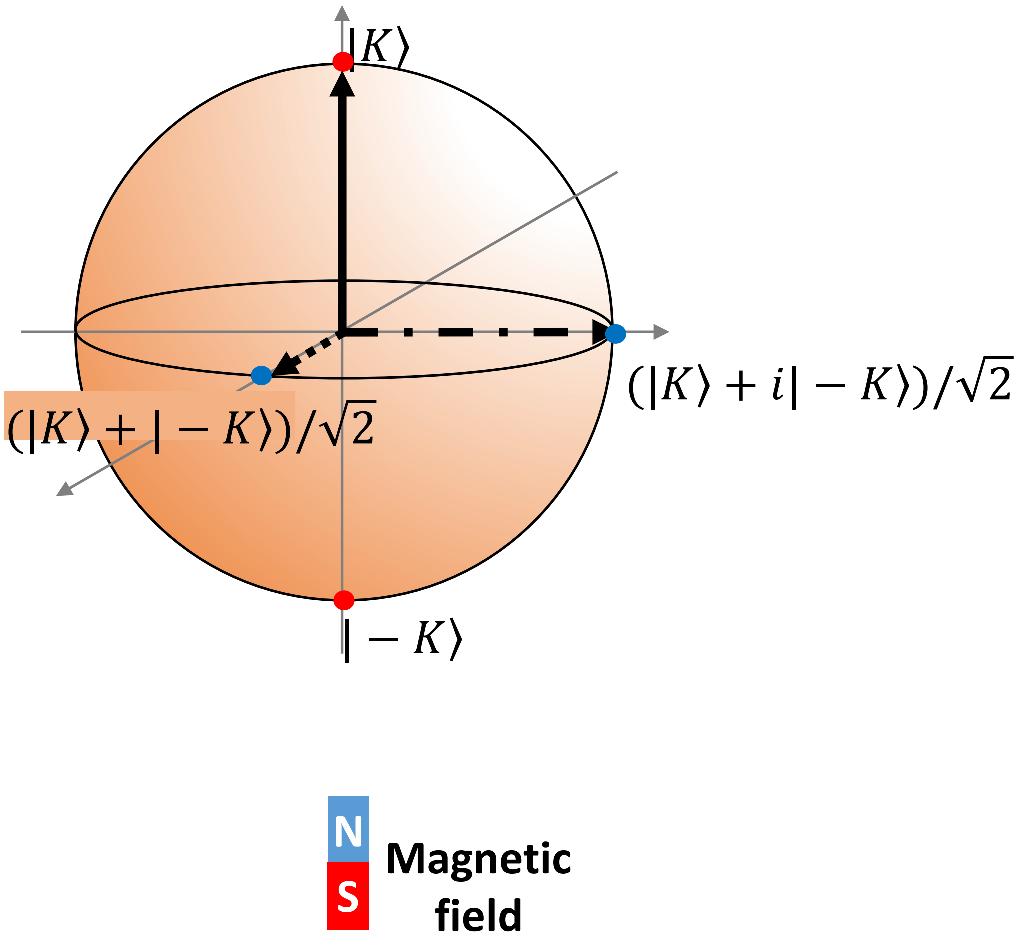
2. Valley information manipulation

: Ultrafast and strong pseudomagnetic field in TMD monolayer

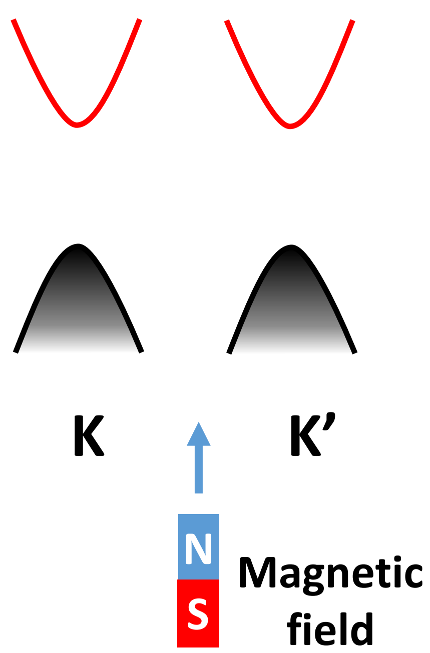
3. Valley information lifetime

: Ultralong valley polarization in TMD heterostructures

Spin/Valley Manipulation

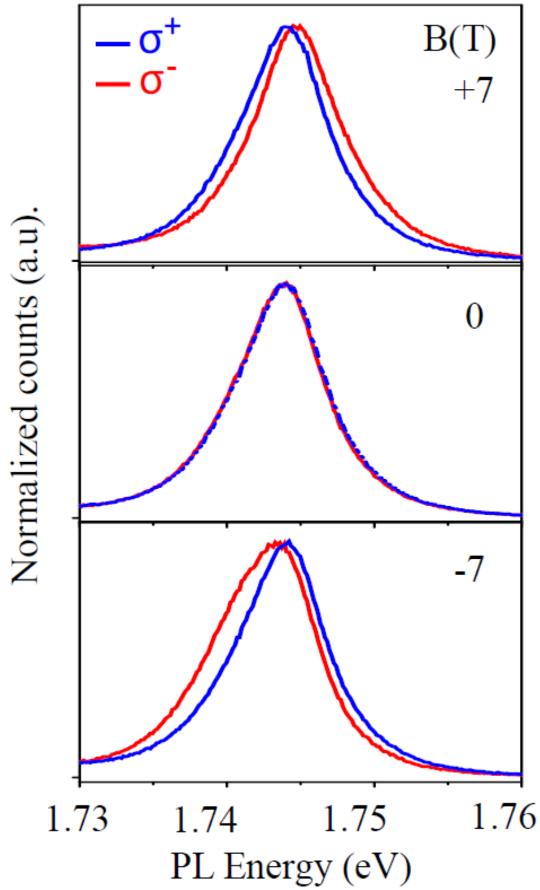


Break valley degeneracy



Spin/Valley Manipulation

Valley Zeeman effect observation

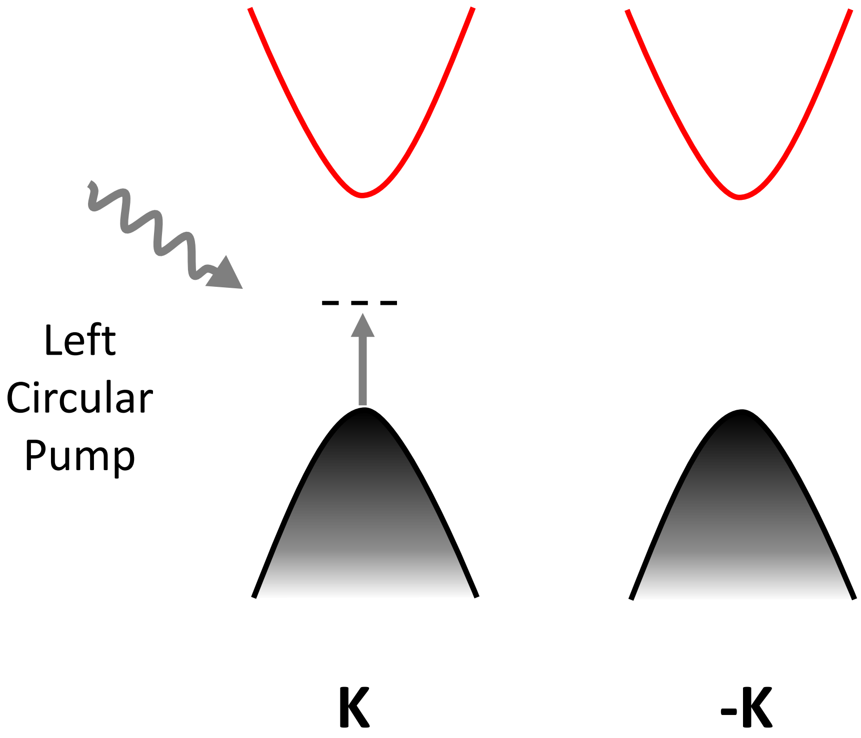


8 T ~ 1 meV

Y. Li, *et. al.*, PRL **113**, 266804 (2014)
A. Srivastava, *et. al.*, Nature Physics **11**, 141 (2015)
G. Avivazan, *et. al.*, Nature Physics **11**, 148 (2015)
D. Macneill, *et. al.*, PRL **114**, 037401 (2015)

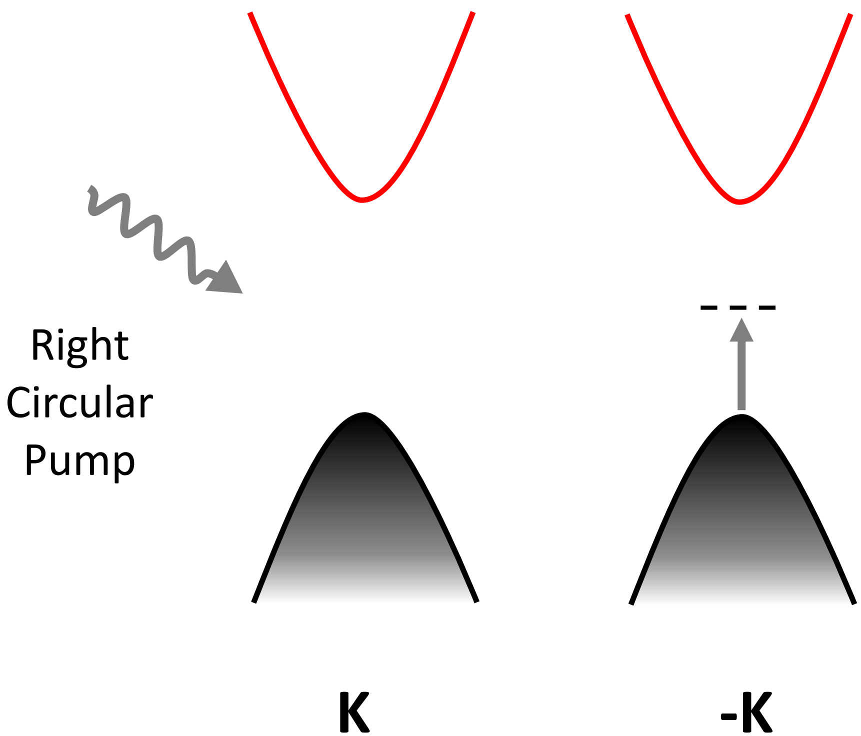
Ultrafast and Efficient Valley Control

Non-resonant femtosecond pulse with circular polarization



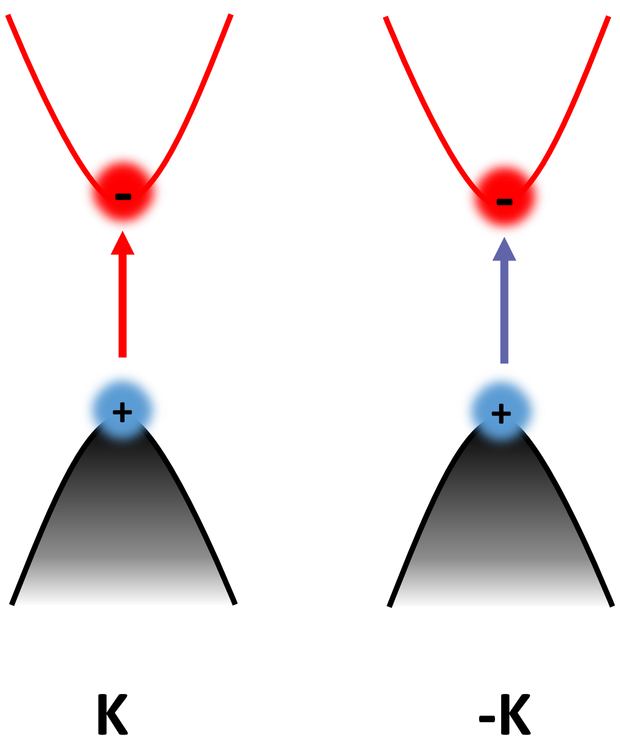
Ultrafast and Efficient Valley Control

Non-resonant femtosecond pulse with circular polarization

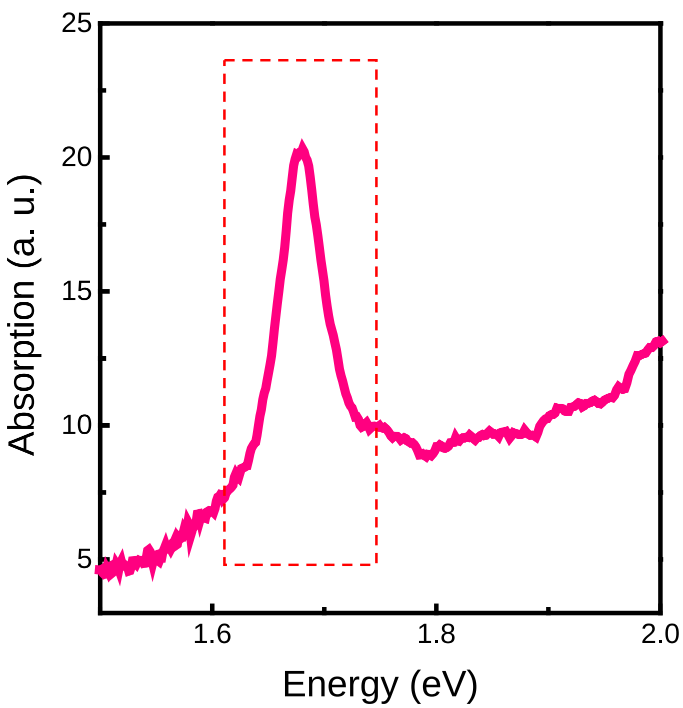


Optical Stark Effect: Pseudomagnetic field (Atomic physics)

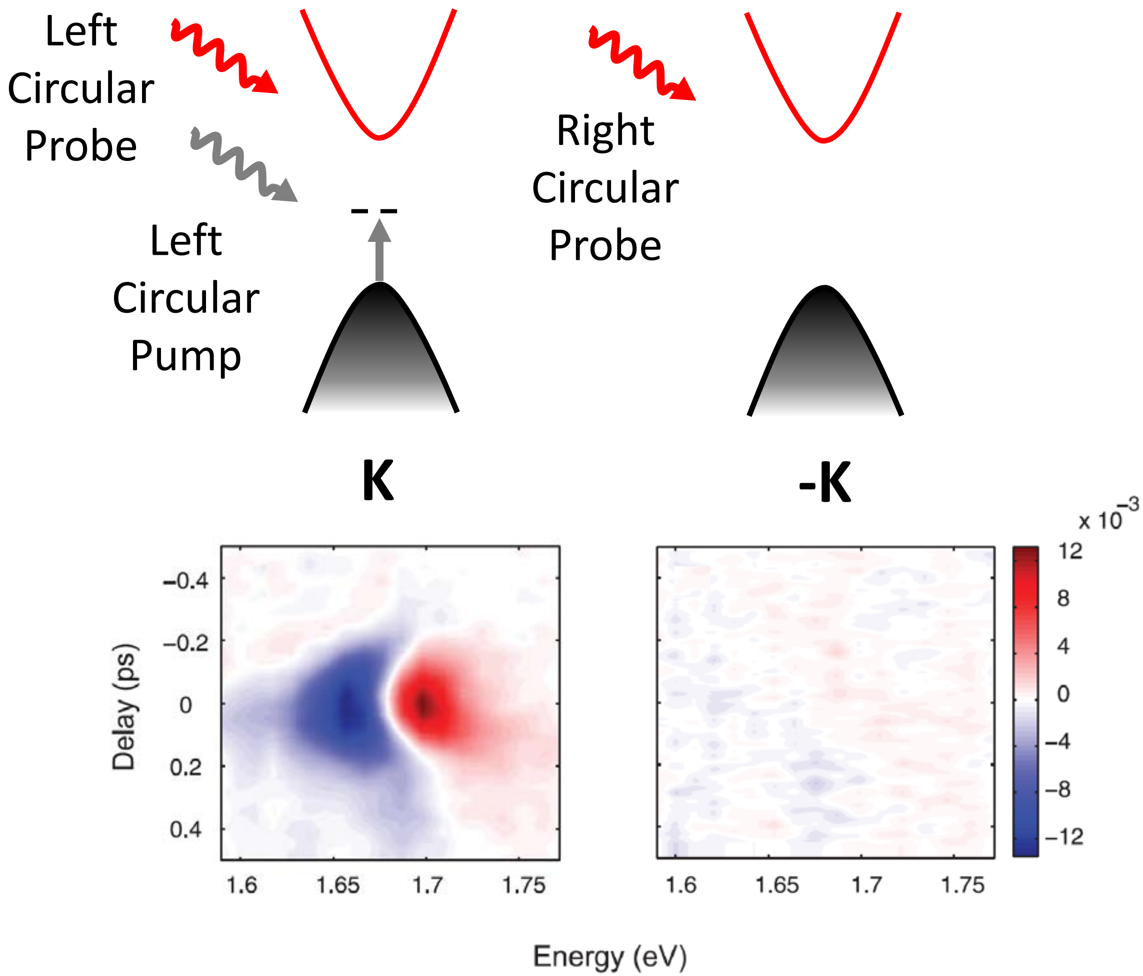
Sample and Absorption Spectrum



Absorption at 1.68 eV

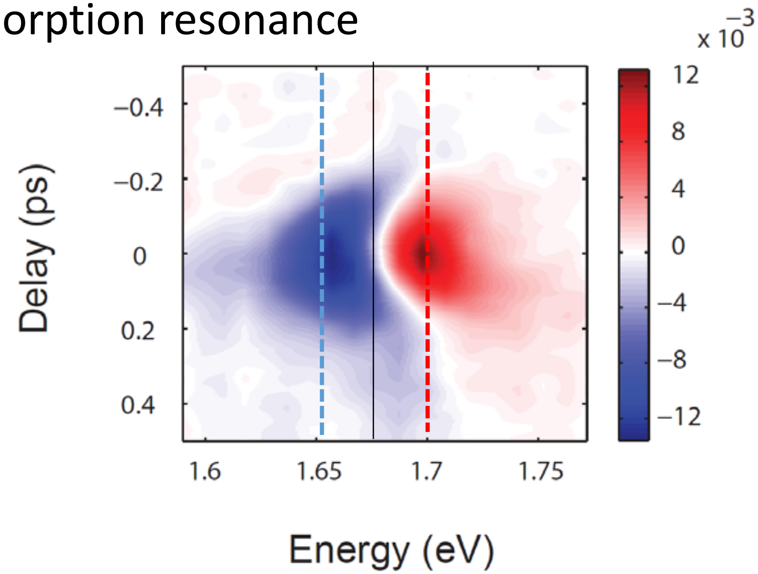


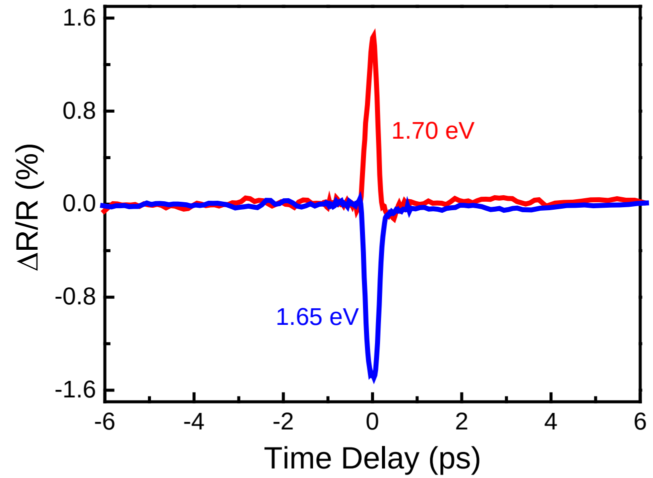
Transient Absorption Spectrum



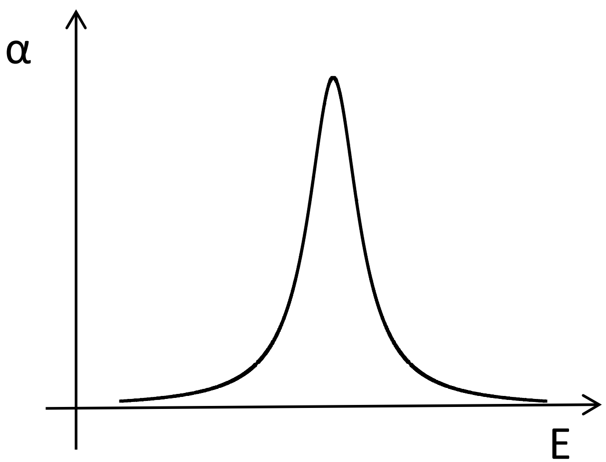
J. Kim*, X. Hong*, *et. al.*, Science **346,** 1205 (2014)

Absorption resonance

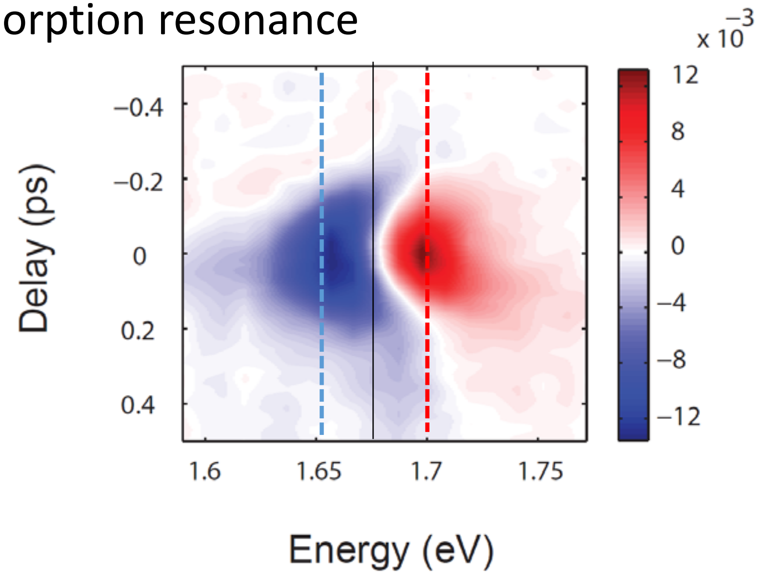


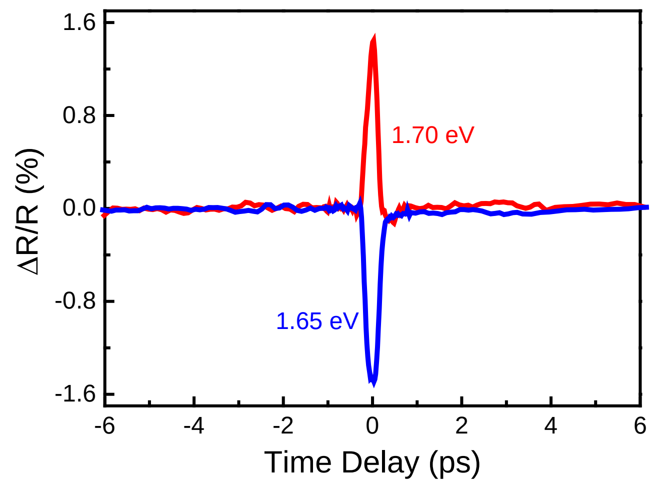


Instantaneous response

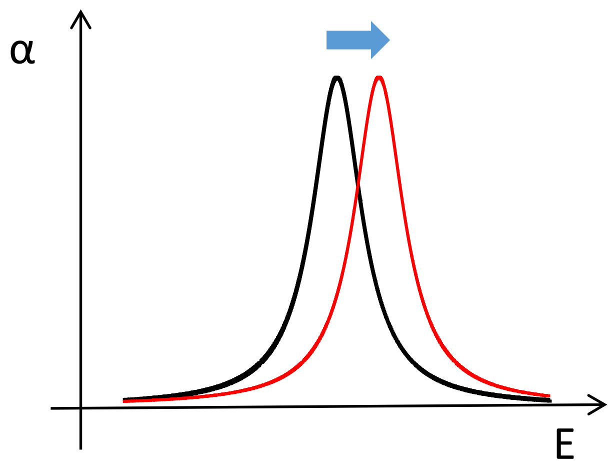


Absorption resonance

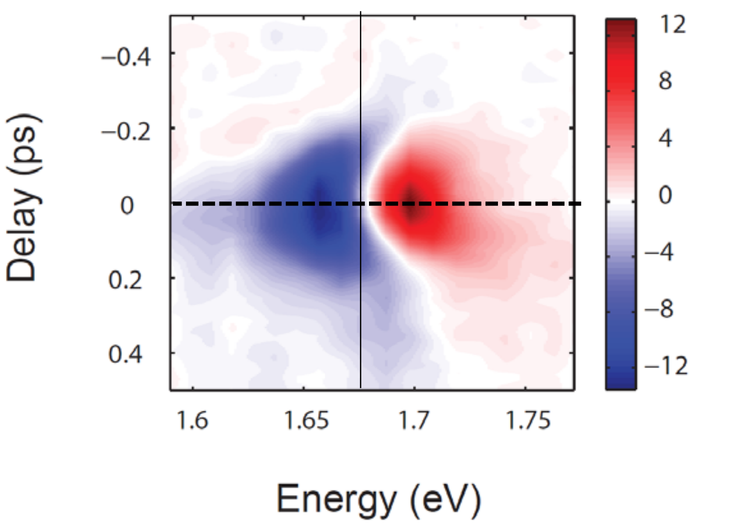


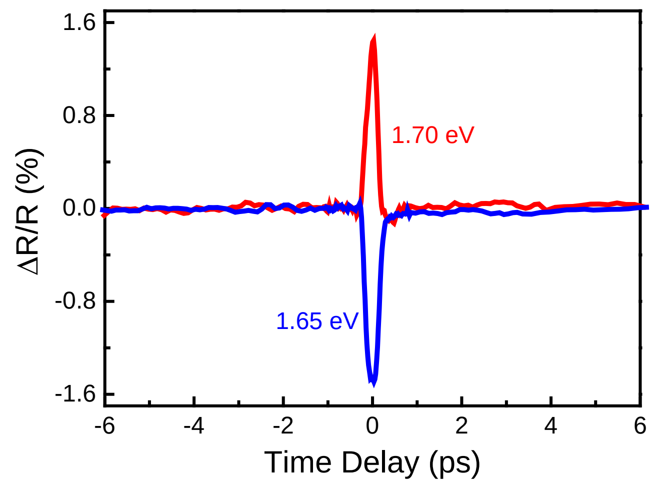


Instantaneous response

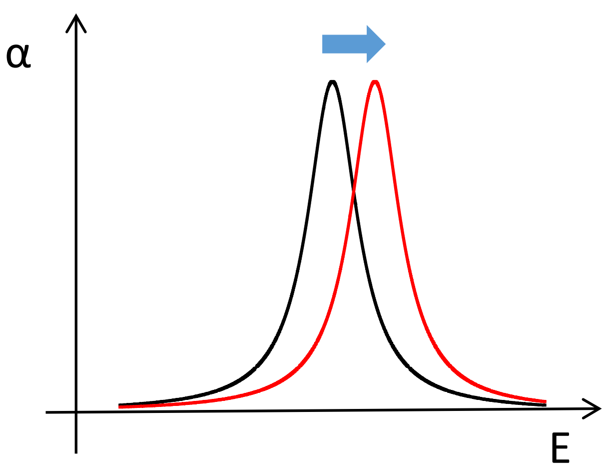


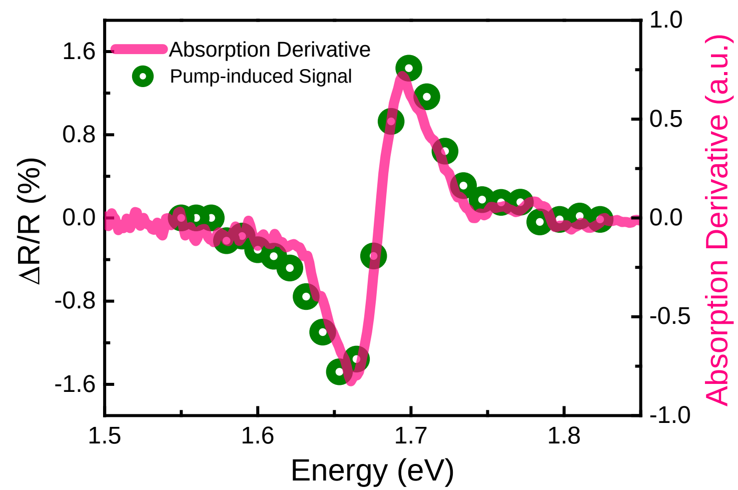
Absorption resonance





Instantaneous response





Energy blueshift : 4 meV

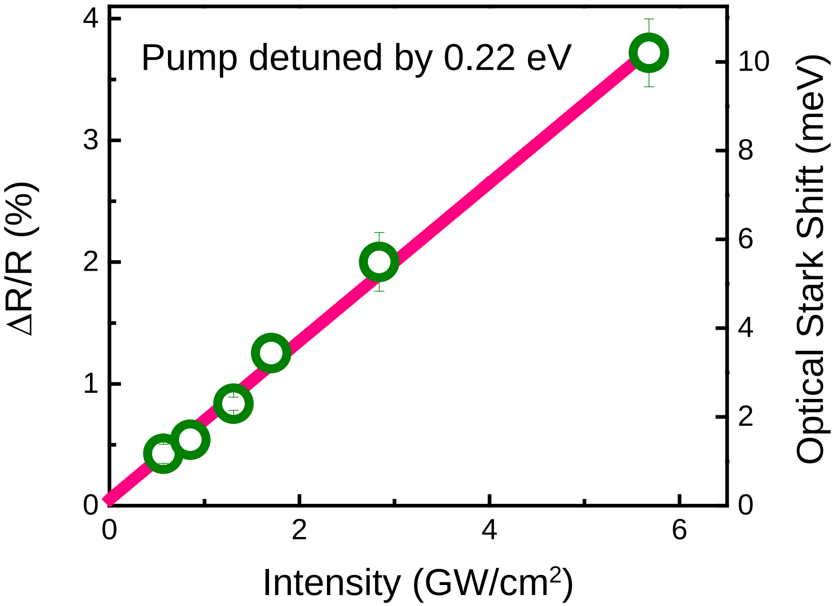
Pump Power and Detuning Dependence

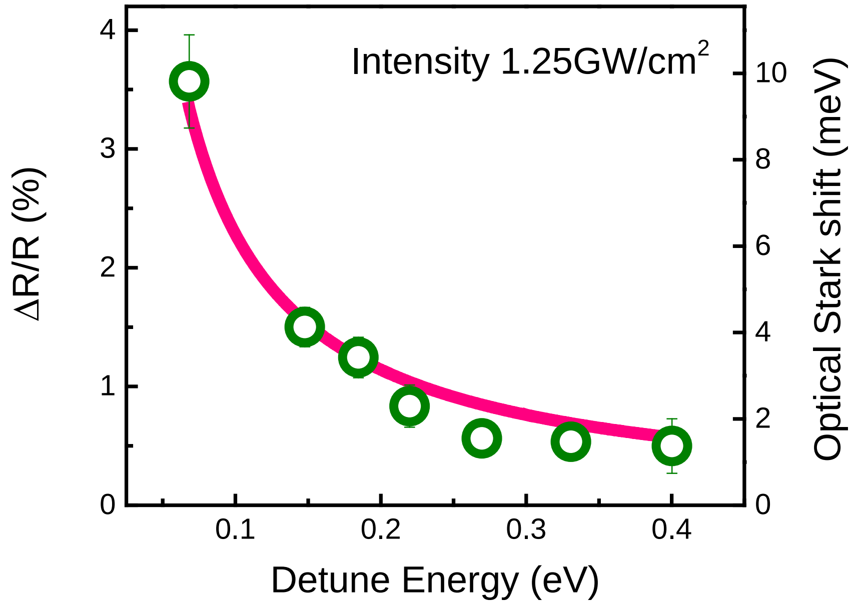
Energy shift : $\delta(\hbar\omega) = \dfrac{2S \cdot E_p^2}{\hbar\Omega}$

S : *optical Stark effect coefficient*

E_p : *Electric field of pump pulse*

Ω : *pump energy detuning*





Estimation of Pseudo-magnetic Field

Pseudomagnetic field : $$B_{eff} = \frac{\Delta E}{2 g_{ex} \mu_B}$$

g-factor of valley exciton in WSe2 : $g_{ex} \sim 1.5$ (theory)

$\boldsymbol{\Delta E = 10\ meV}$ **:** $\boldsymbol{B_{eff} \sim 60\ T}$

PRL **113** 266804 (2014)

PRL **114**, 037401 (2015)

Nature Physics **10**, 343 (2014)

PRB **88**, 085440 (2013)

Nature Physics **11**, 141-147 (2015)

Nature Physics **11**, 148-152 (2015)

Outline

1. New information carrier

   : Valley state in 2D transition metal dichalcogenides (TMD)

2. Valley information manipulation

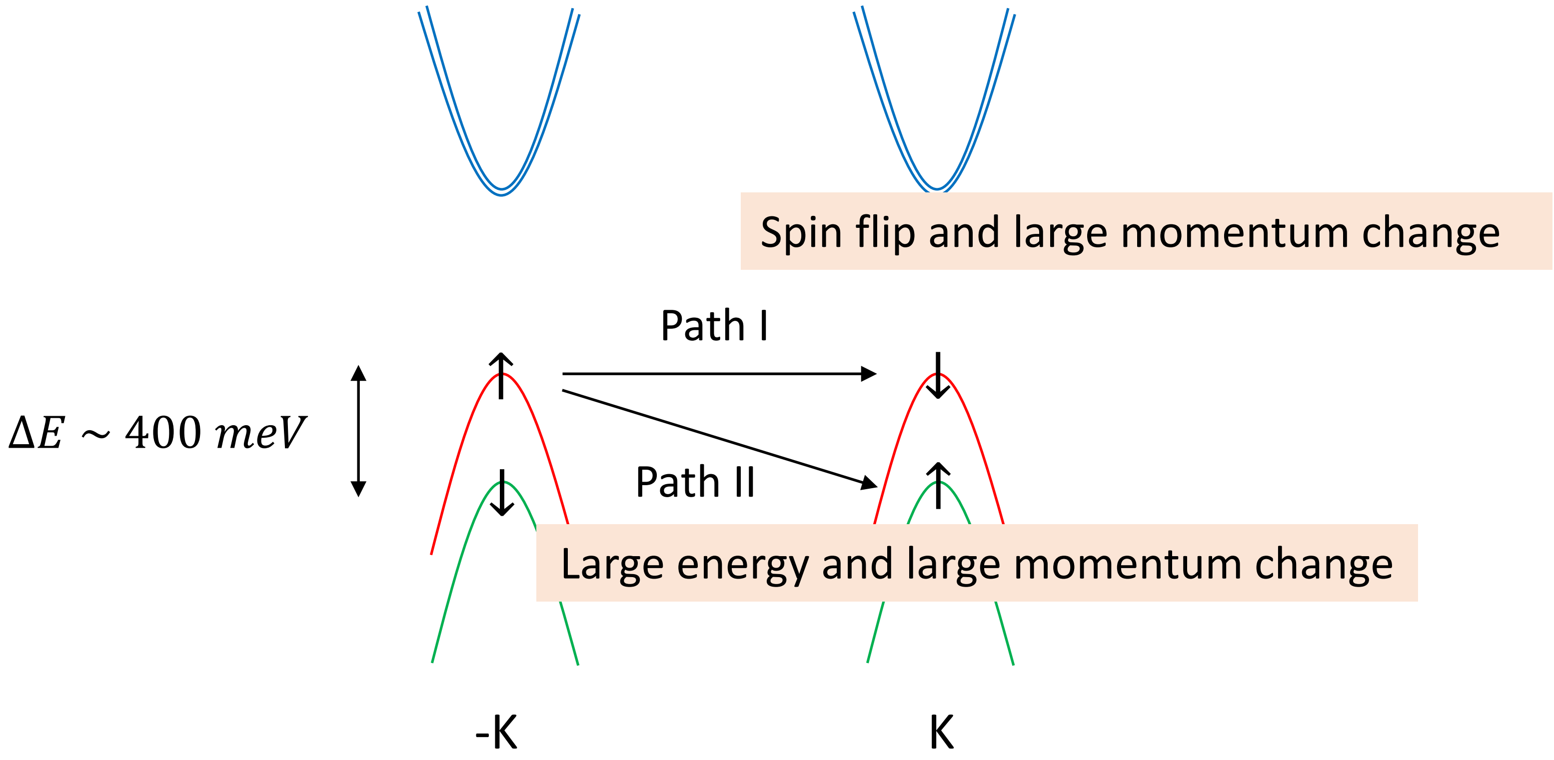
   : Ultrafast and strong pseudomagnetic field in TMD monolayer

3. **Valley information lifetime**

   : Ultralong valley polarization in TMD heterostructures

Valley State in 2D TMD

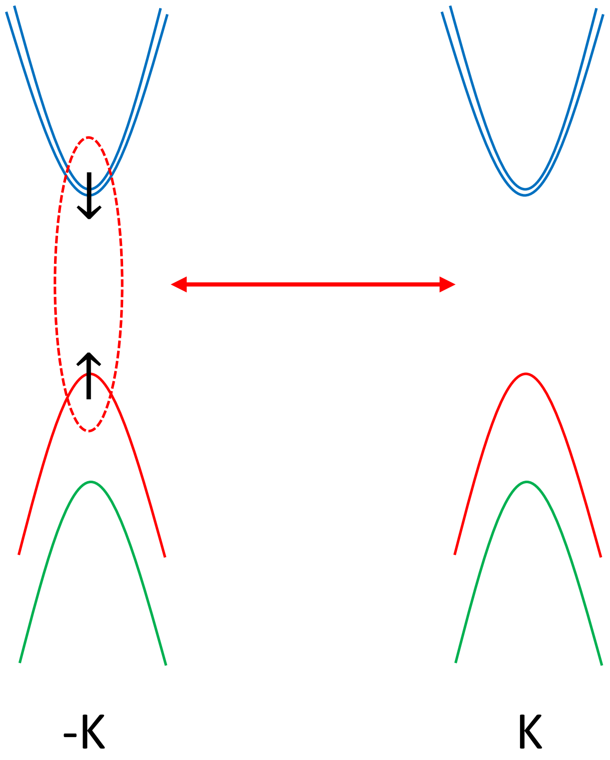
Strong SO coupling: Spin-Valley locking



Possibly ultralong spin/valley lifetime!

Valley State in 2D TMD

Exciton flips valley state fast!

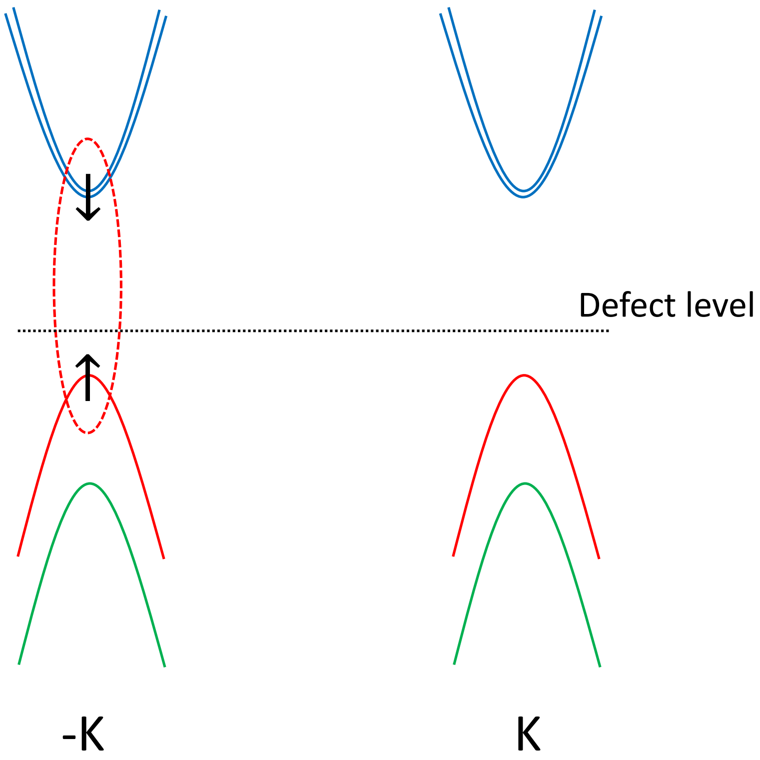


Maialle, Silva and Shan, 1993
Yao group, 2014
Wu group, 2014

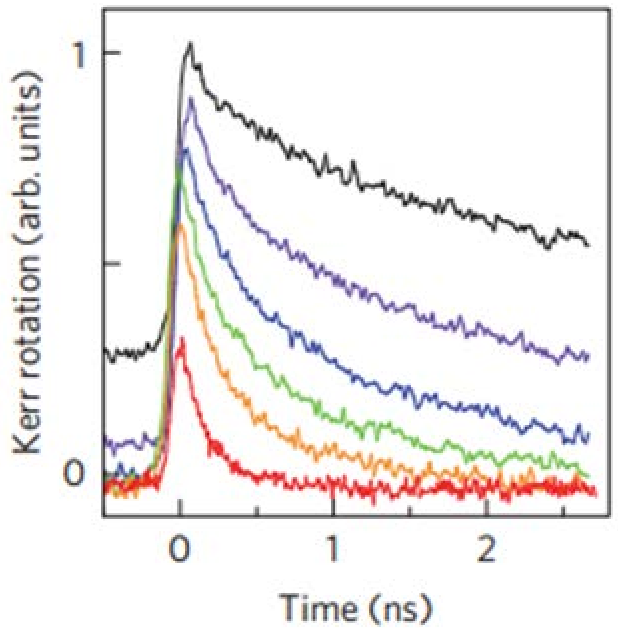
Exchange interaction: ~ 300 fsec

Valley State in 2D TMD

What if we can break exciton and leave only carriers? : Resident carrier



nsec valley lifetime



Lifetime limited by defect and low valley polarization

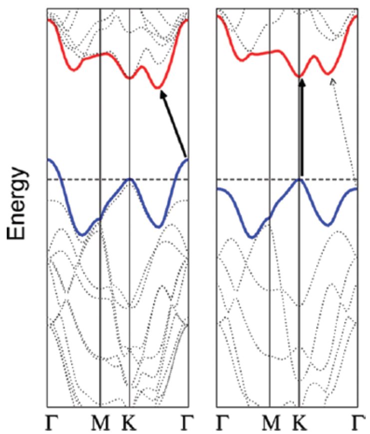
S. Crooker 2015

‘Ultrafast’ and ‘intrinsic’ process for exciton dissociation?

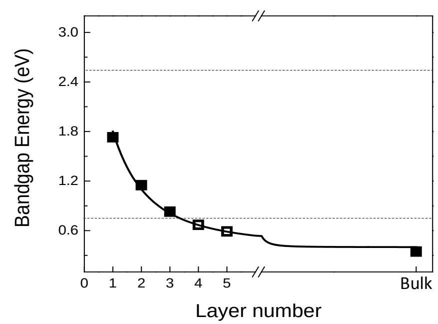
Controlling Electronic Structure in vdW crystals

Indirect to direct gap transition (MoS2)

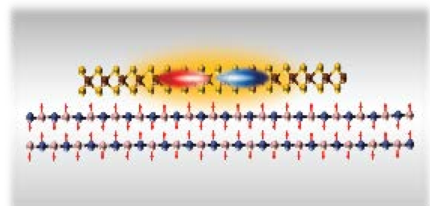
2 layer **1 layer**

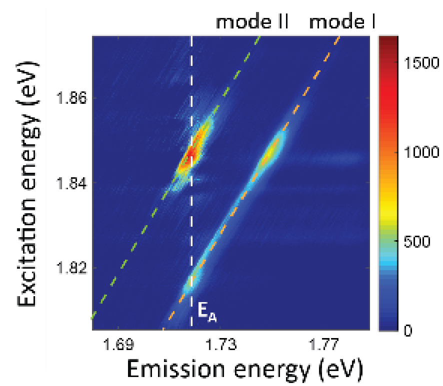


Direct bandgap 1.7 – 0.3 eV (Phosphorene)



Interlayer electron-phonon Interaction (WSe2/hBN)





A. Splendiani, **J. Kim** *et. al. Nano Lett.* **10**, 1271 (2010)

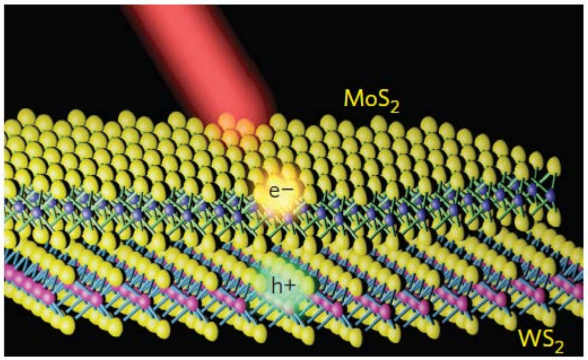
Also, Fai Mak *et. al.* PRL (2010)

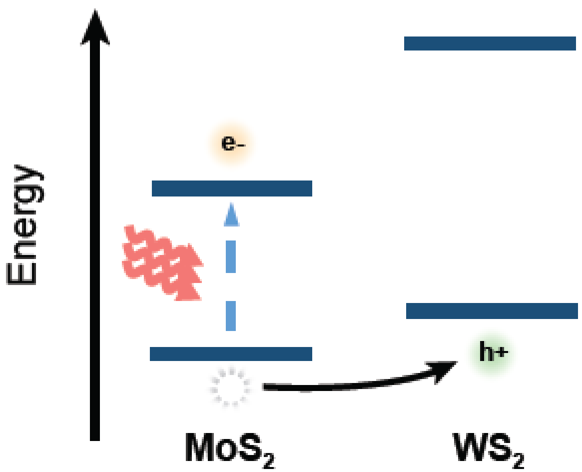
L. Li*, **J. Kim***, C. Jin* *et. al, Nature Nano* ***12***, 21 (2017)

C. Jin*, **J. Kim*** *et. al, Nature Physics* ***13***, 127 (2017)

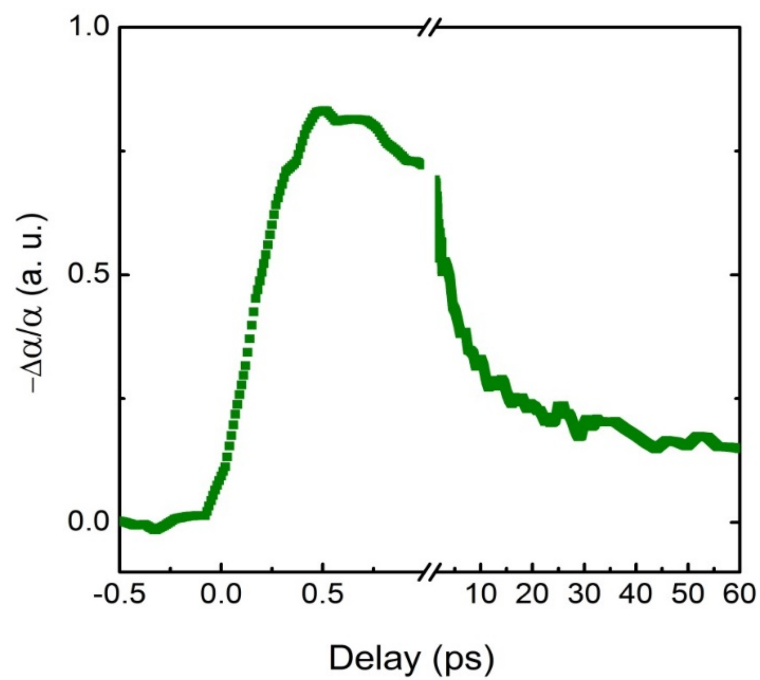
Controlling Carrier Dynamics

Ultrafast charge separation in TMD heterostructure





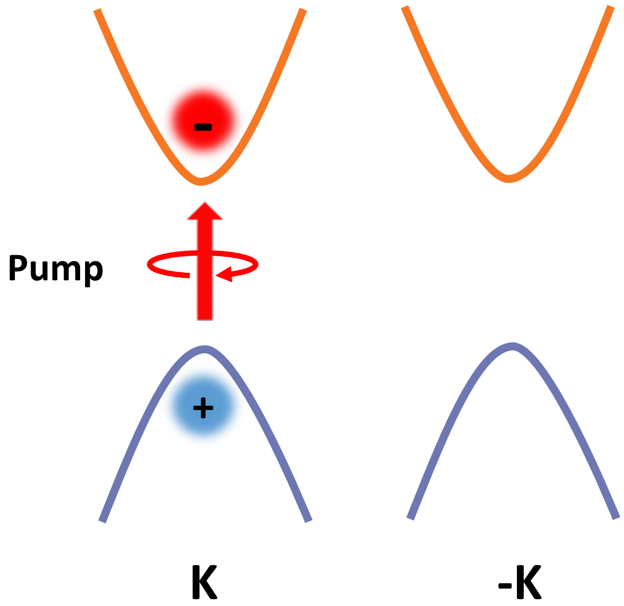
Charge transfer dynamics



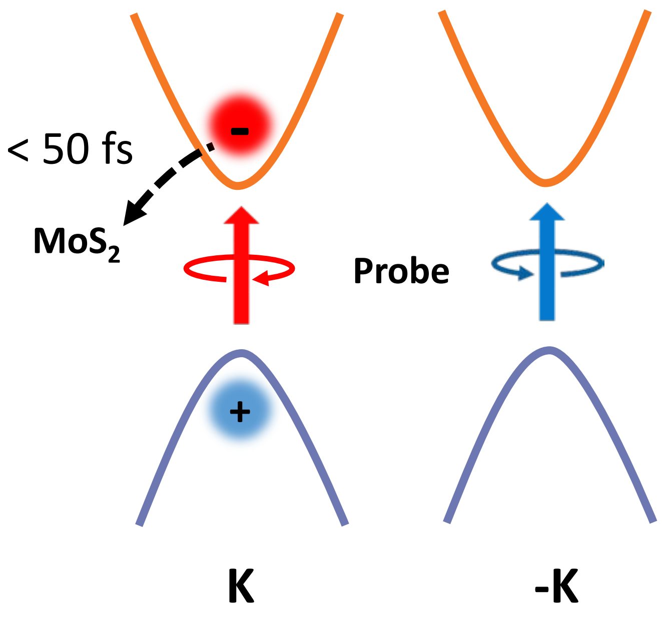
Charge transfer time < 50 fs

X. Hong*, **J. Kim***, S. Shi*, *et. al.*, Nature Nanotechnology **9**, 682 (2014)

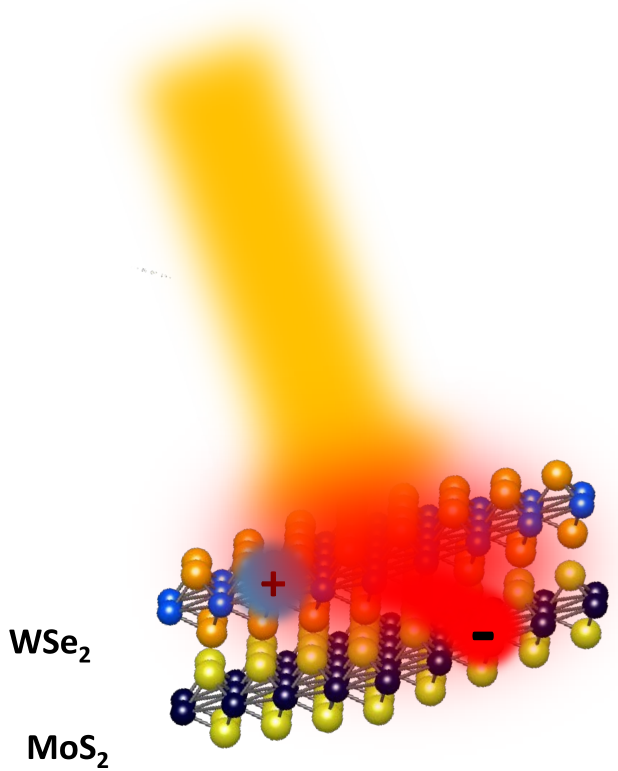
Generation of Valley-Polarized Holes

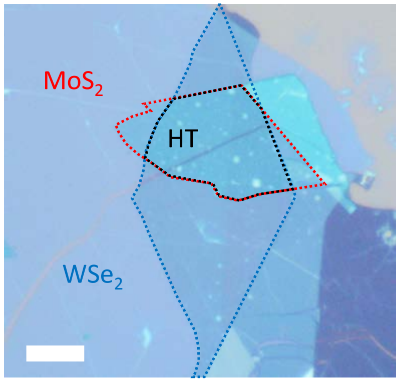


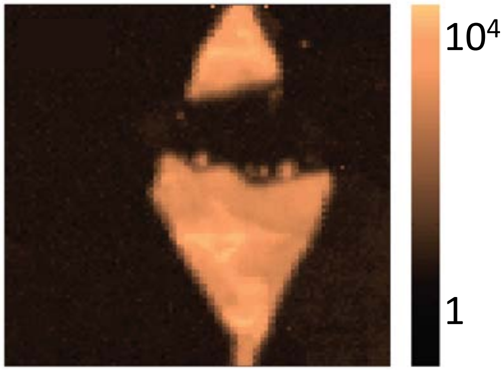
Generation of Valley-Polarized Holes



Generation of Valley-Polarized Holes

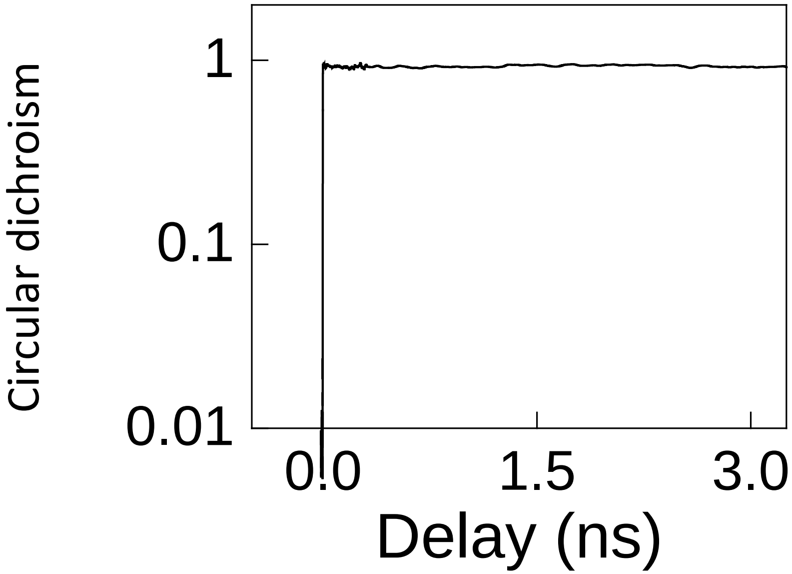


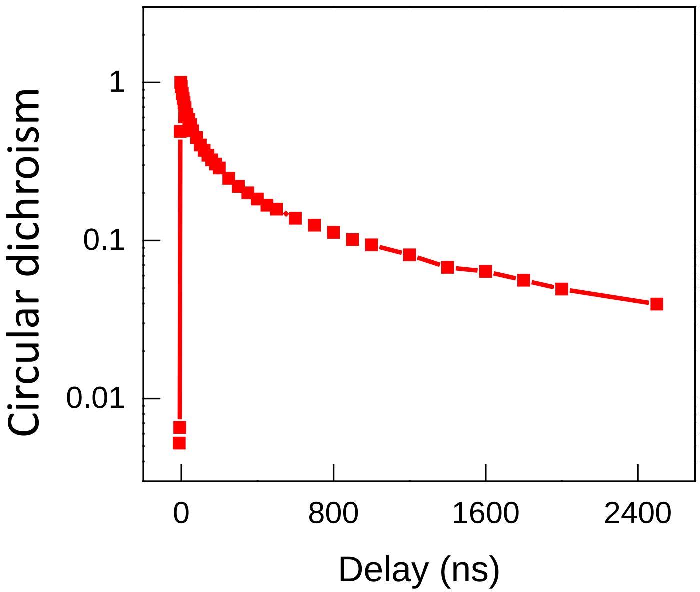




<u>J. Kim</u>*, C. Jin*, *et. al.* (Under review)

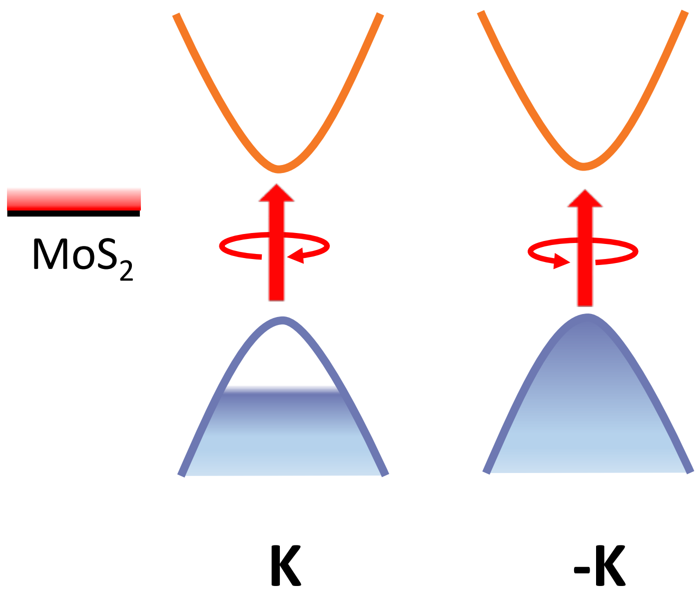
Valley-polarized Hole Dynamics





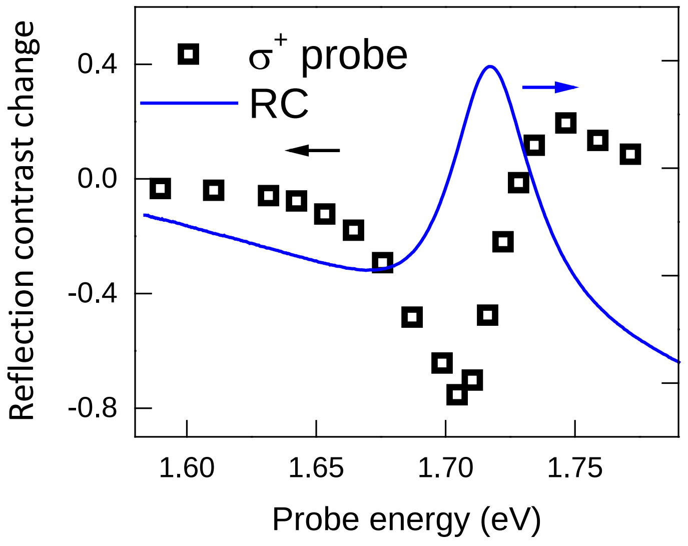
Microsecond valley lifetime! : Orders of magnitude longer than previous report

Valley Polarization Analysis

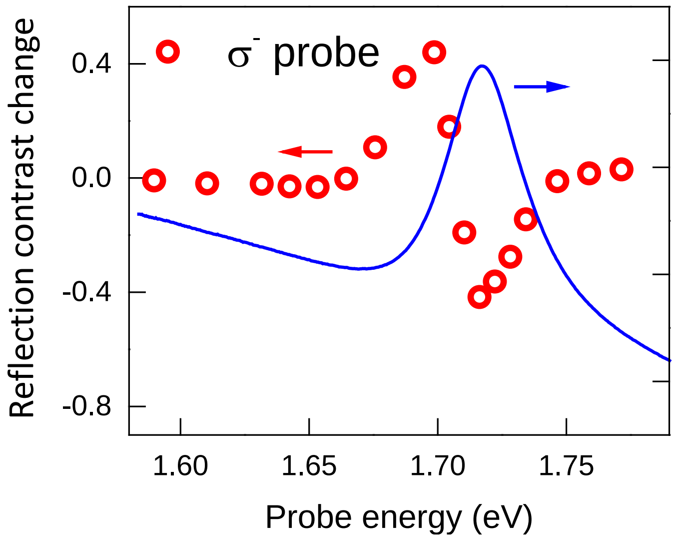


Carrier population at a valley induces “oscillator strength decrease”.

Valley Polarization Analysis



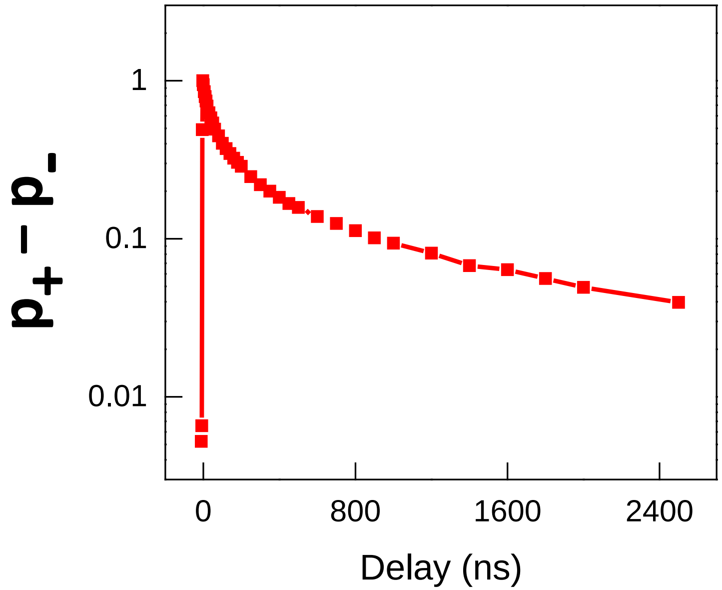
Large oscillator strength decrease

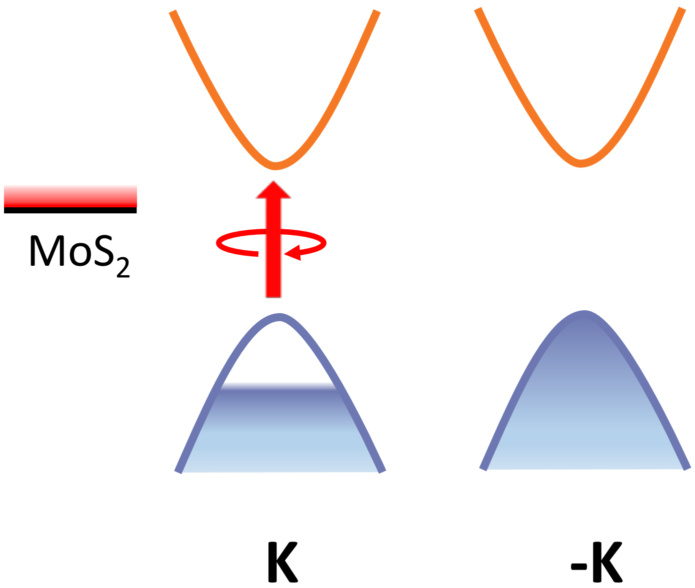


Pure resonance shift

Oscillator strength decrease occurs only at K valley: 100 % valley polarization

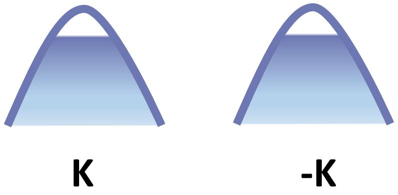
Valley Relaxation Mechanism



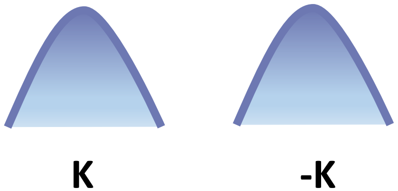


$p_+ - p_- = 0$ means:

1) Intervalley scattering:

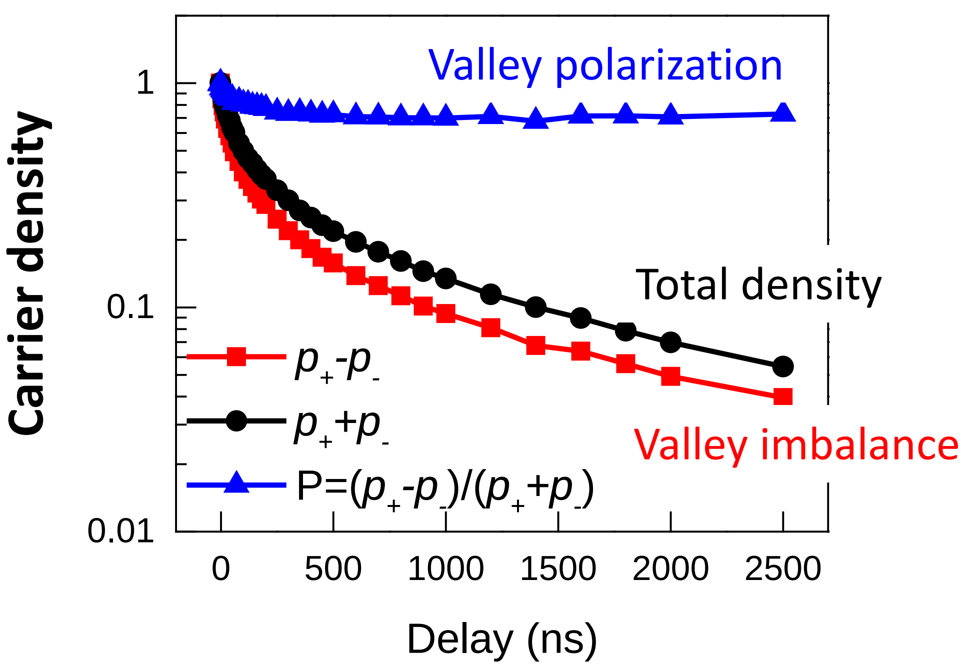


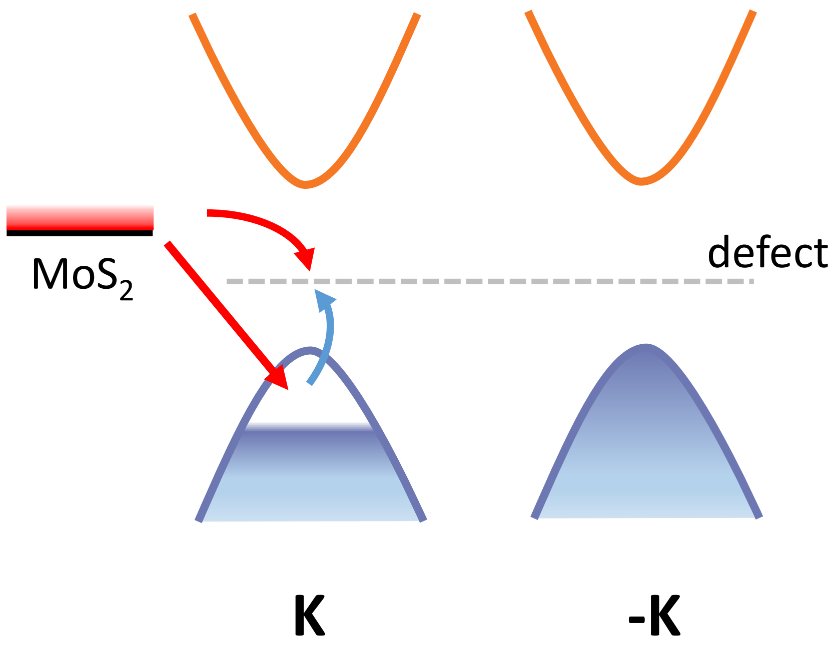
2) Population relaxation:



Valley Relaxation Mechanism

Population relaxation dominates valley relaxation

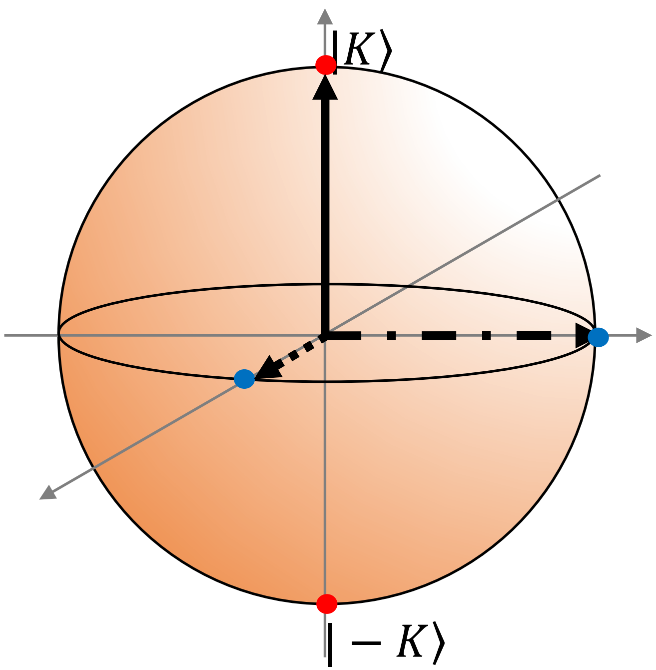




Intervalley scattering rate > 40 microsecond!

Summary

Valley state in 2D TMD: Novel information carrier



1. Valley state manipulation

Optical Stark effect:
Ultrafast pseudomagnetic field > 60 T

2. Valley lifetime

Ultrafast charge transfer
process in TMD heterostructure:
Lifetime > 1 us (can be longer than 40 us)

Acknowledgement

Advisor: Prof. Feng Wang

Wang group

Chenhao Jin

Xiaoping Hong

Sufei Shi

Zhiwen Shi

Arizona State Univ.

Prof. Sefaattin Tongay

KAUST

Prof. Lance Li

Peking Univ.

Prof. Yanfeng Zhang

Collaborations in Berkeley:

Prof. Steve Louie, Felipe H. da Jornada, Diana Y. Qiu

Prof. Ron Shen

Prof. Alex Zettl, Dr. Jongmin Yuk (now in KAIST)

Prof. Junqiao Wu, Dr. Joonki Suh

Prof. Michael Crommie, Dilon Wong

Thank you!