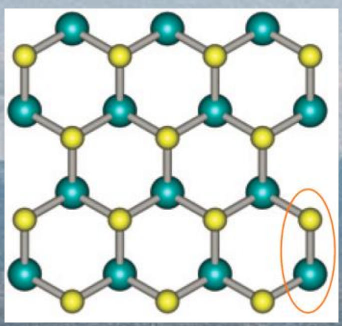
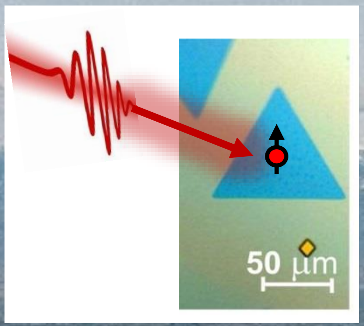
“Valley noise” & valley correlations in monolayer semiconductors





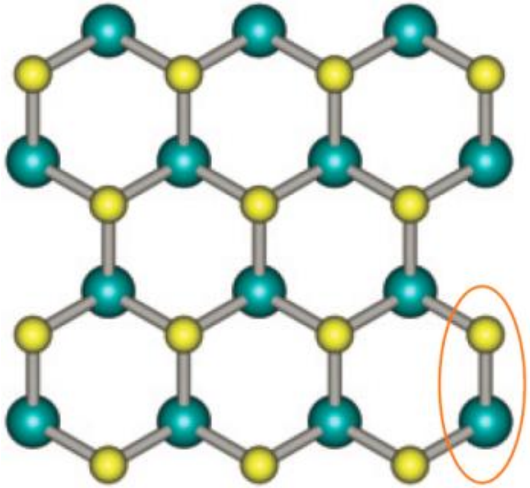
Scott Crooker
National High Magnetic Field Laboratory
Los Alamos National Lab

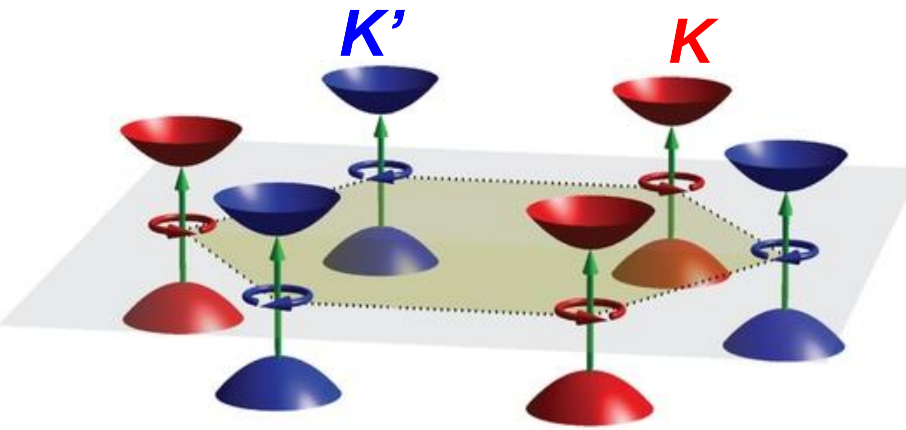
Strongly Correlated Quantum Materials
Santa Fe (April 29- May 3)

NATIONAL HIGH MAGNETIC FIELD LABORATORY

Today: 'Valley noise' & valley-dependent correlations in monolayer semiconductors

Monolayer transition-metal dichalcogenides (MoS_2, WSe_2, etc)





-from X. Xu *et al., Nat. Phys*. **10**, 343 (2014)

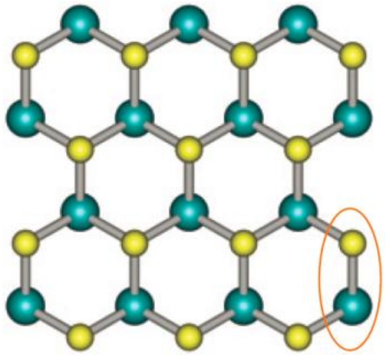
Direct-gap semiconductors!

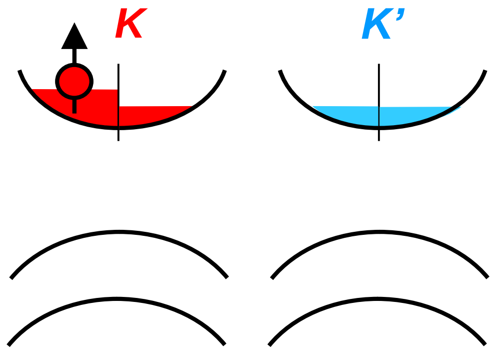
E_{gap}* ~ 1.5 - 2 eV *(infrared – red)

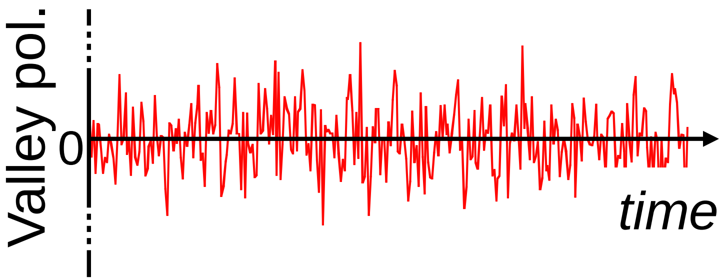
perhaps more

(like graphene… but ^ useful for optoelectronics)

Valley noise & valley correlations in monolayer semiconductors







Outline

- **Monolayer "transition-metal dichalcogenide" semicond.**
  - *optical access to valley degree of freedom*
- **Thermodynamic "valley noise"**
  - *nonperturbative measurement of valley dynamics*
  - *microsecond valley polarization of holes: robust spin-valley locking*
- **Valley correlations in doped monolayers**
  - *intervalley plasmons, optical response in high magnetic fields*

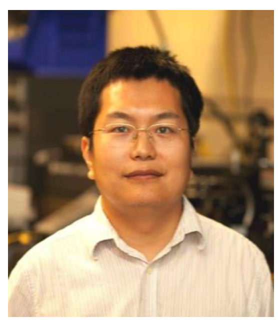
Strongly Correlated Quantum Materials
Santa Fe (April 29- May 3)

Valley noise & valley correlations in monolayer semiconductors

Mateusz Goryca, Andreas Stier, & Scott Crooker *(NHMFL– Los Alamos)*
Nathan Wilson & Xiaodong Xu (*U. Washington*)

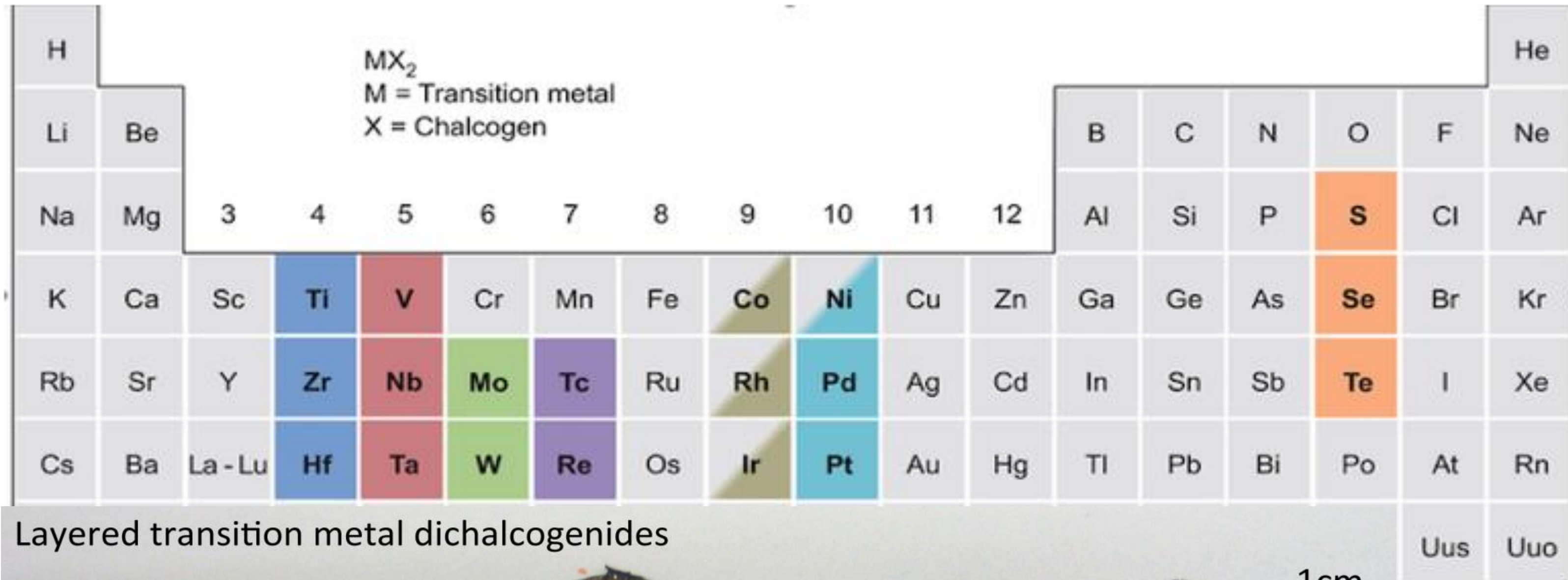
Mateusz Goryca

Andreas Stier



Nathan Wilson & Xiaodong Xu
(University of Washington)

“Transition-Metal Dichalcogenide” (TMD) materials



M. Chhowalla *et.* al. Nature Chemistry **5**, 263–275 (2013).

Bulk TMD semiconductors: MoS_2, $MoSe_2$, WS_2, WSe_2

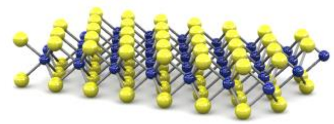
Molybdenite (MoS_2)

So… why all the recent fuss?

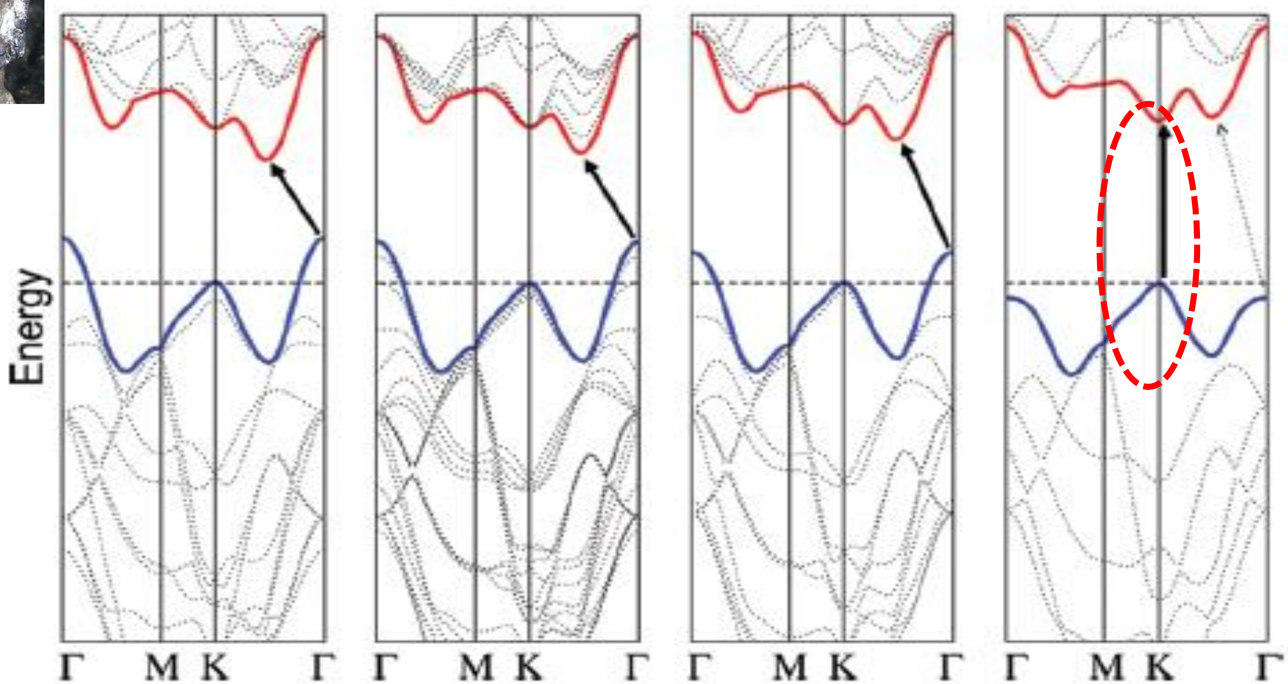
3D to 2D: Indirect- to direct-gap semiconductor

A. Splendiani, F. Wang *et al.*, Nano Letters **10**, 1271 (2010)
K. F. Mak, J. Shan, T. F. Heinz *et al.*, PRL **105**, 136805 (2010)

Decreasing # of MoS_2 layers



Bulk MoS_2 | Quadrilayer | Bilayer | Monolayer



Direct bandgap ~ 1.5 – 2 eV

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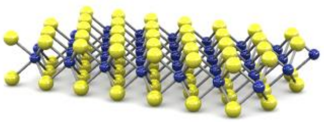
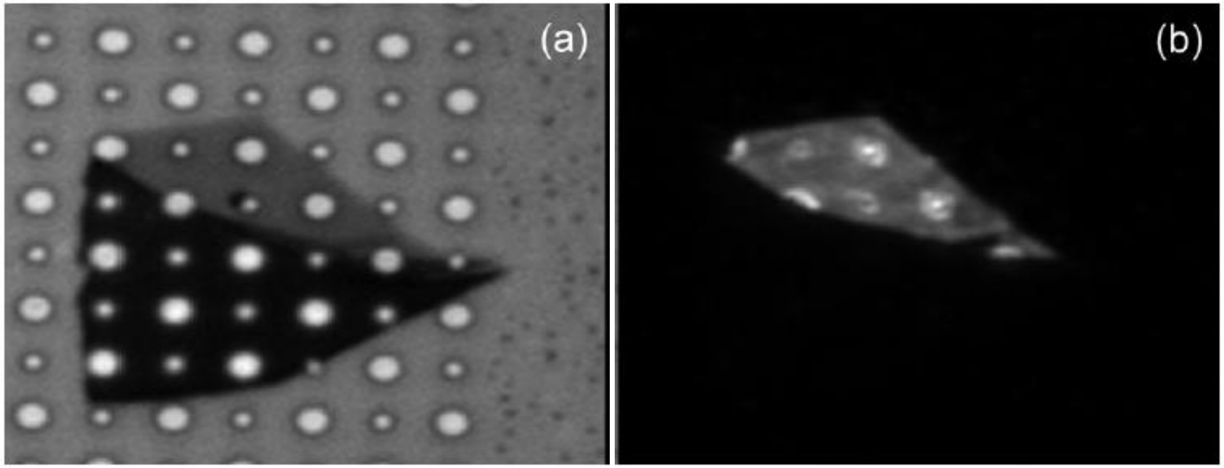
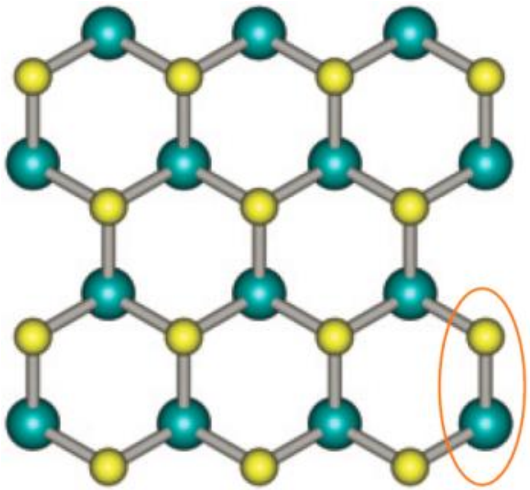


Image (MoS_2) Photoluminescence

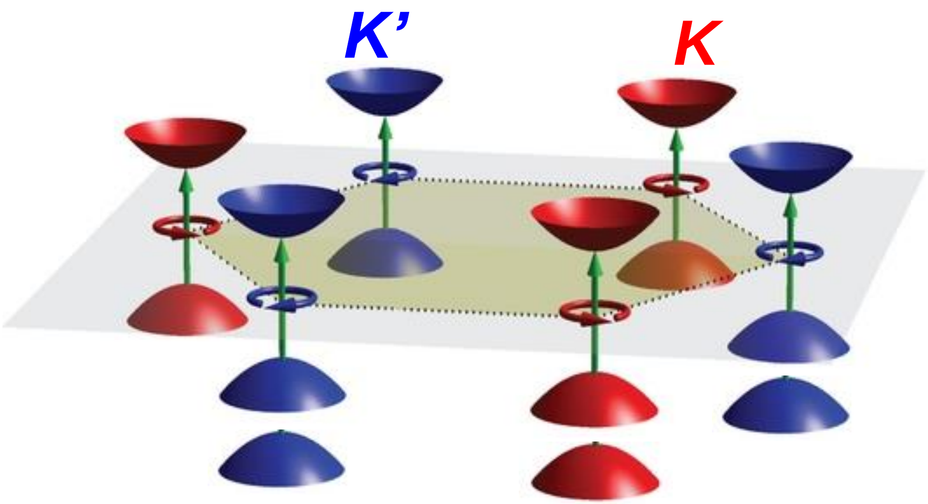


Direct bandgap ~ 1.5 – 2 eV

Motivation: "Valley pseudospin" & valleytronics



-from X. Xu *et al., Nat. Phys*. **10**, 343 (2014)



Key point: Valley-specific optical selection rules

RCP & LCP light couple *selectively* to *K* & *K'* valleys

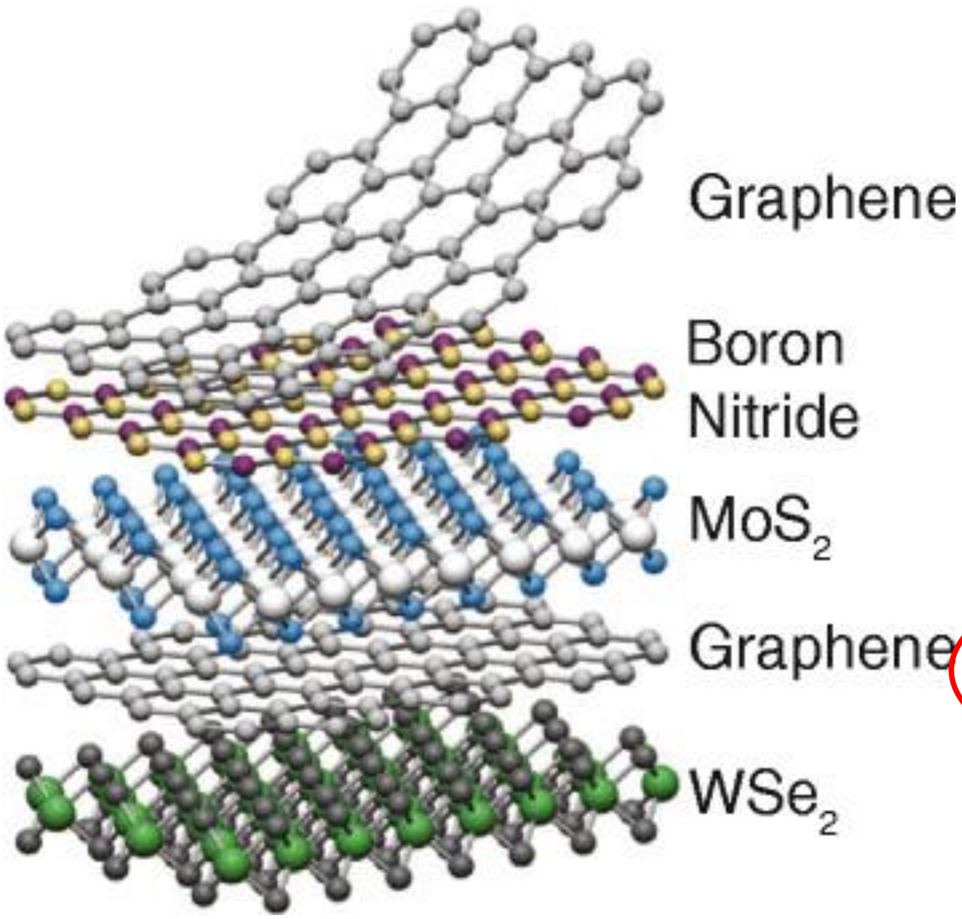
Easy access to valley degrees of freedom

Excellent reviews:

X. Xu, D. Xiao, W. Yao, & T. Heinz, *Nature Physics* **10**, 343 (2014)
K. F. Mak, D. Xiao, & J. Shan, *Nature Photonics* **12**, 451 (2018)

Motivation: van der Waals heterostructures

Opto-electronic ('spin/valleytronic'?) devices by van der Waals assembly of 2D layers



As-yet-unknown:

1) Role of dielectric screening?
   - E_{gap}? *exciton binding?*

2) Exciton (e-h pair) mass?
   - *form of potential V(r)?*

3) Spin & valley dynamics of electrons & holes?

4) Correlations & interactions

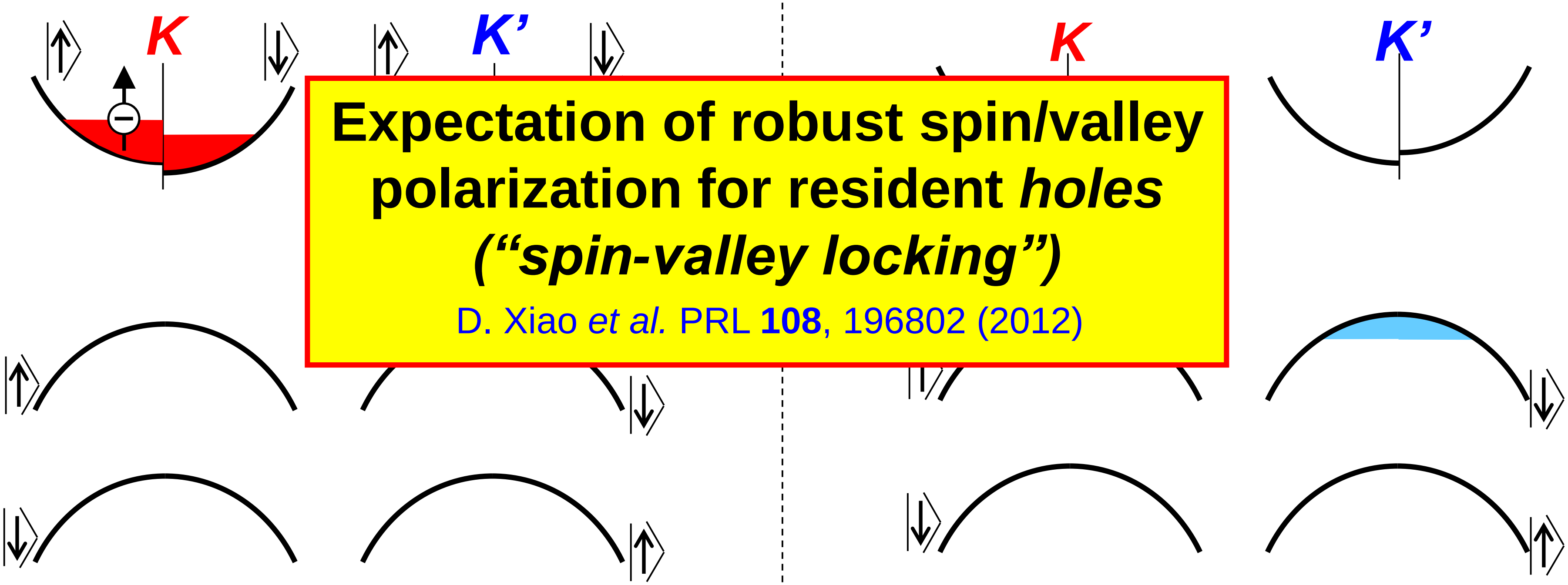
Geim & Grigorieva, Nature **499**, 419 (2013)

A very important question:

Spin/valley scattering timescale of *electrons* & *holes?*

Electron-doped (*n*-type) regime

Hole-doped (*p*-type) regime



Expectation of robust spin/valley polarization for resident *holes*
(*“spin-valley locking”*)

D. Xiao *et al.* PRL **108**, 196802 (2012)

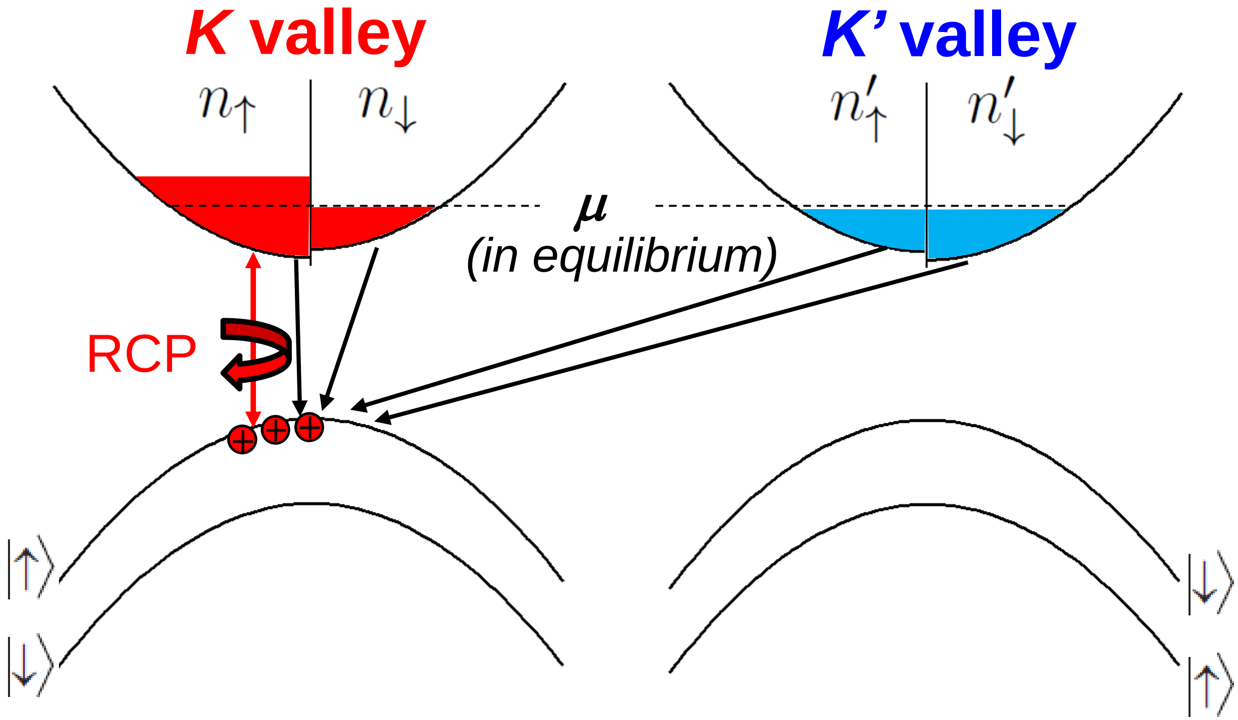
Three pathways for electrons…

1) spin scattering within a valley
2) spin-*conserving* intervalley
3) spin-*flip* intervalley (slow)

ONE pathway for holes…

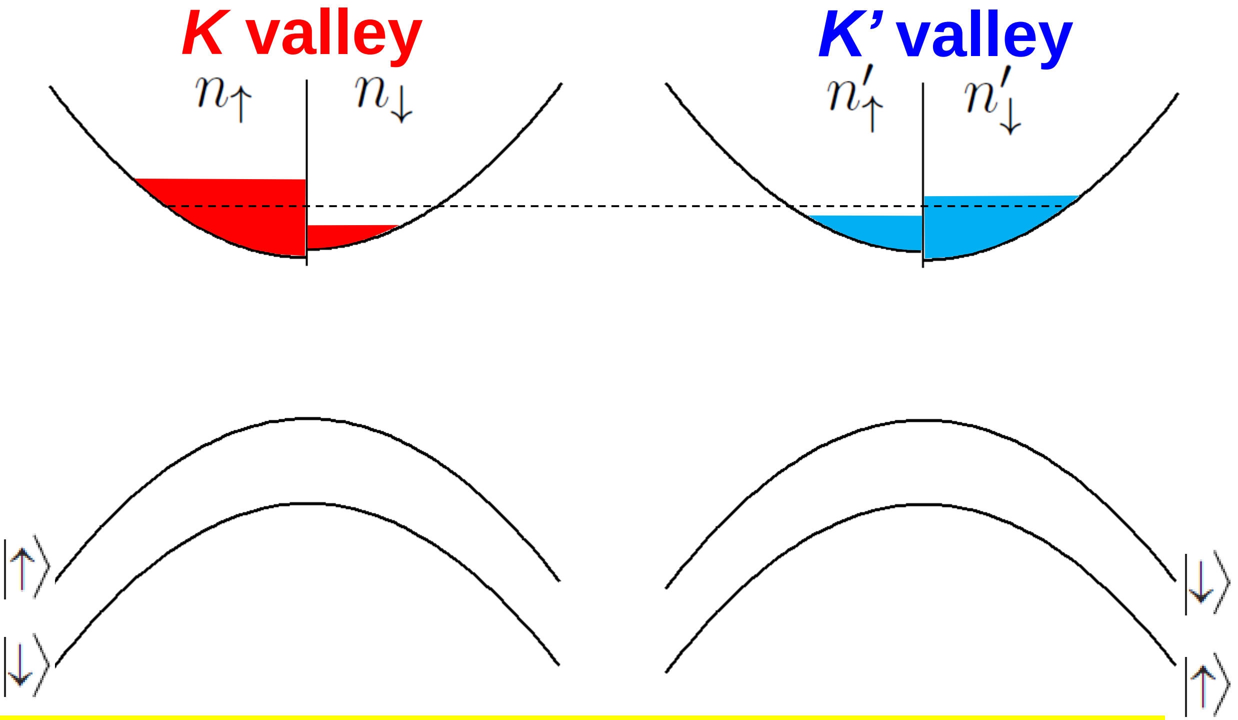
1) spin-*flip* intervalley (slow)

Conventional approach (pump-probe optics)



- ***t<0***: resident carriers in equilibrium ($\mu_\uparrow = \mu_\downarrow = \mu'_\uparrow = \mu'_\downarrow$)
- ***t=0***: pump RCP at 'A' exciton
- ***t~0-100 ps:*** minority carriers recombine with resident carriers *(i.e., excitons & trions form & recombine)*

Conventional approach (pump-probe optics)

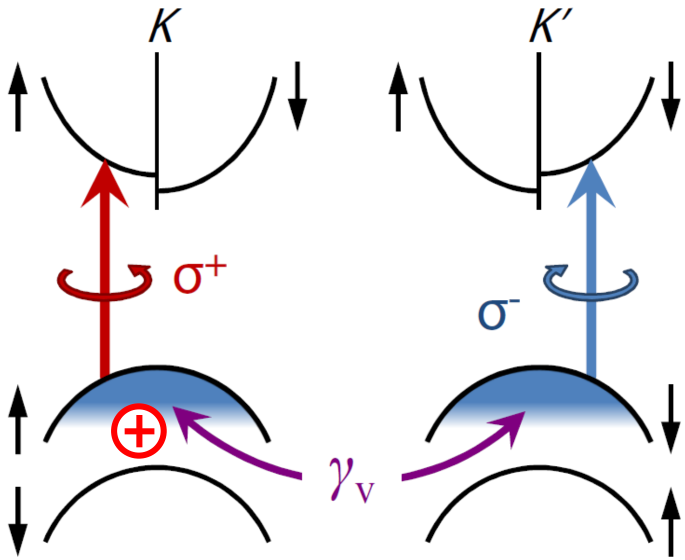


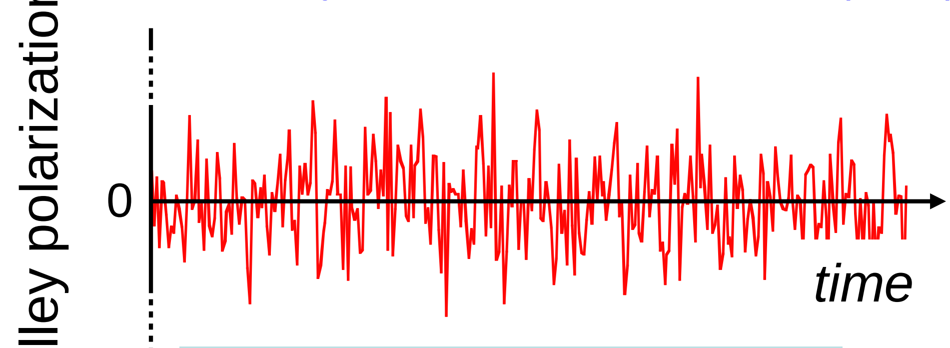
Question:
Can we measure valley dynamics… *without* actually pumping minority carriers?

"Valley Noise Spectroscopy"

M. Goryca, *Sci. Adv.* **5**, eaau4899 (2019)

Consider Fermi sea of holes in thermal equilibrium…





On average: $\langle \delta\theta_F(t) \rangle = 0$

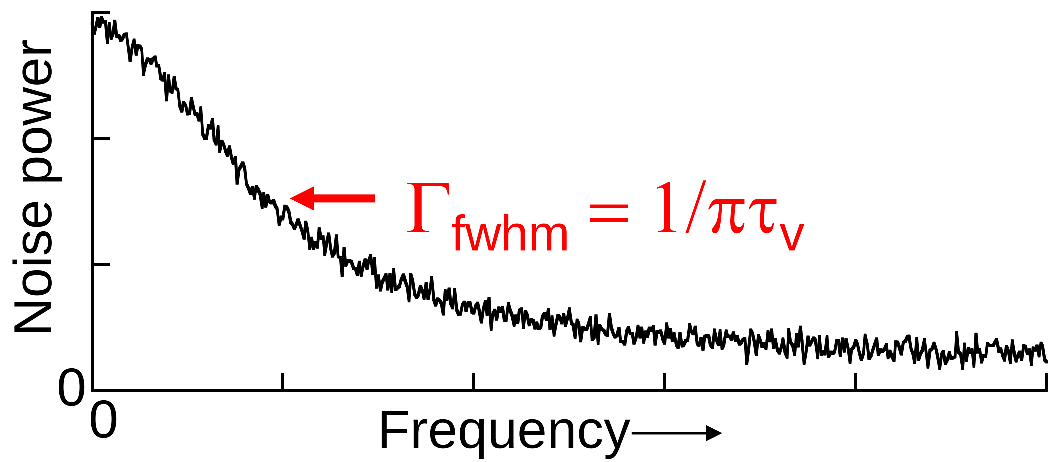
But fluctuations exist: $\langle [\delta\theta_F(t)]^2 \rangle \neq 0$

Valley correlation function: $\langle \delta\theta_F(t)\, \delta\theta_F(0) \rangle$

Spectrum of correlations: "Valley Noise"

- *Width* gives timescales
- *Shape* gives mechanism

etc…

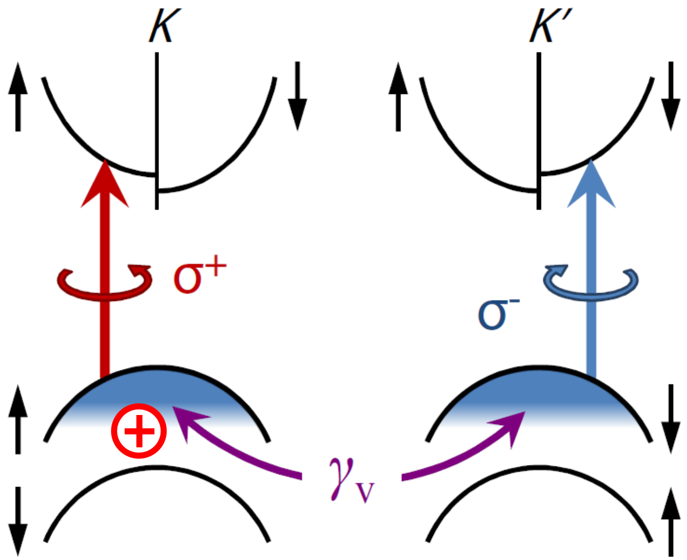


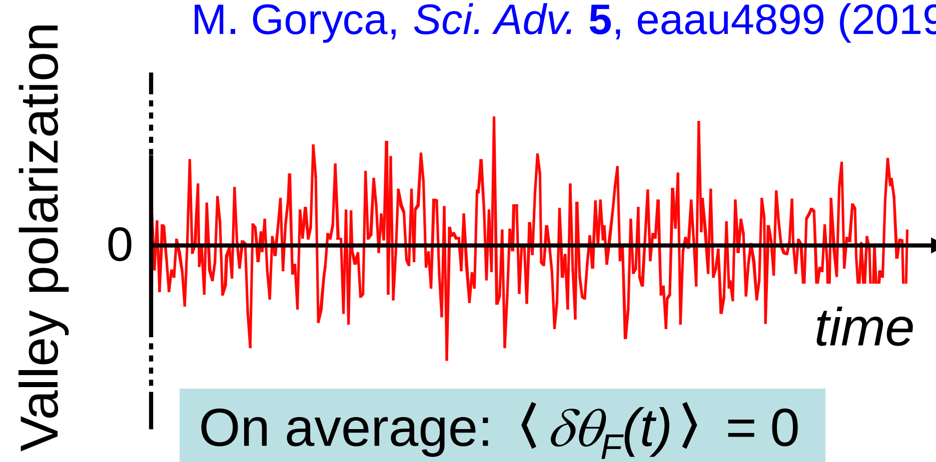
$\Gamma_{\text{fwhm}} = 1/\pi\tau_v$

"Valley Noise Spectroscopy"

Consider Fermi sea of holes in thermal equilibrium…

M. Goryca, *Sci. Adv.* **5**, eaau4899 (2019)





On average: $\langle \delta\theta_F(t) \rangle = 0$

But fluctuations exist: $\langle [\delta\theta_F(t)]^2 \rangle \neq 0$

Valley correlation function: $\langle \delta\theta_F(t)\ \delta\theta_F(0) \rangle$

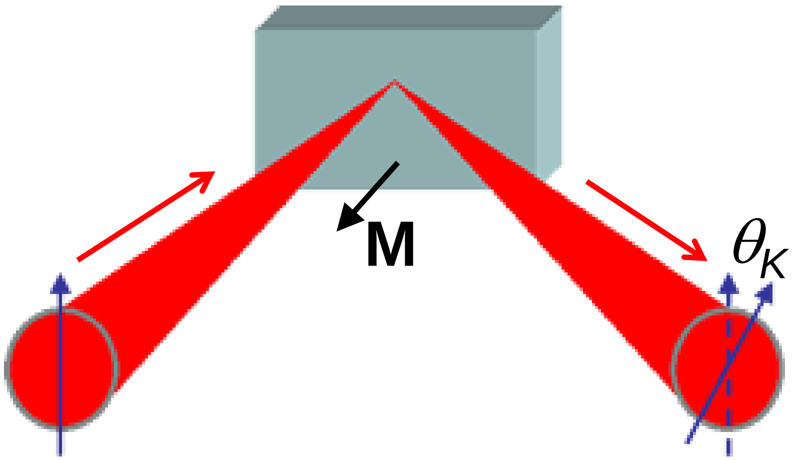
Fluctuation-Dissipation Theorem: $\chi''(\omega)$ = FFT [$\langle \delta\theta_F(t)\ \delta\theta_F(0) \rangle$]

"Linear response of a system to external perturbation (ie, the susceptibility) can be described by the fluctuation properties of the system while in thermal equilibrium".

- *In principle: valley noise alone describes valley dynamics*

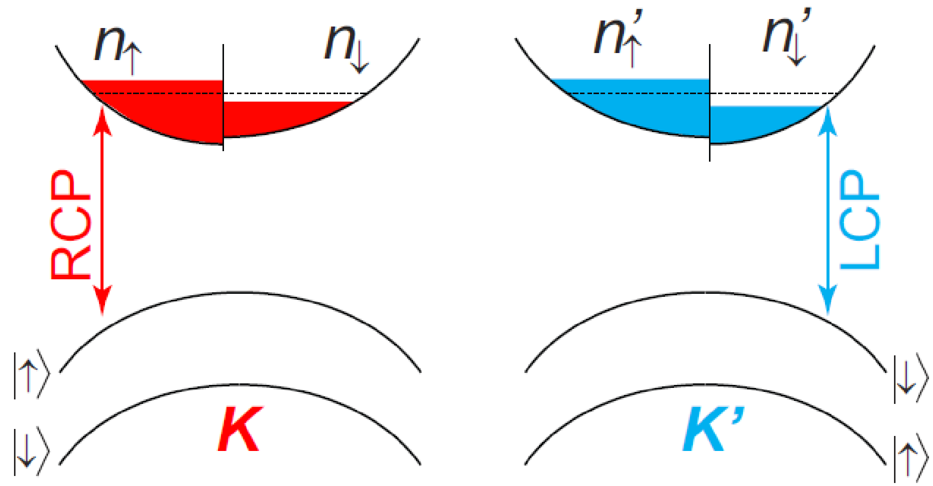
No perturbation, drive, or excitation necessary

Detection: optical Kerr/Faraday rotation:
A direct probe of the carrier valley polarization



Optical Kerr rotation, θ_K

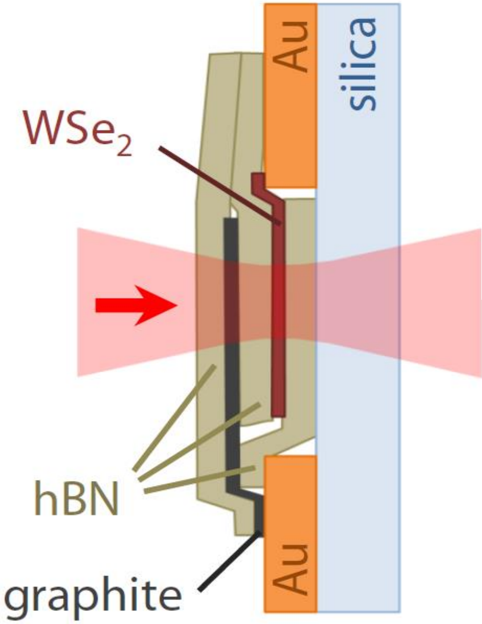
$\propto(\alpha_{RCP}-\alpha_{LCP})$

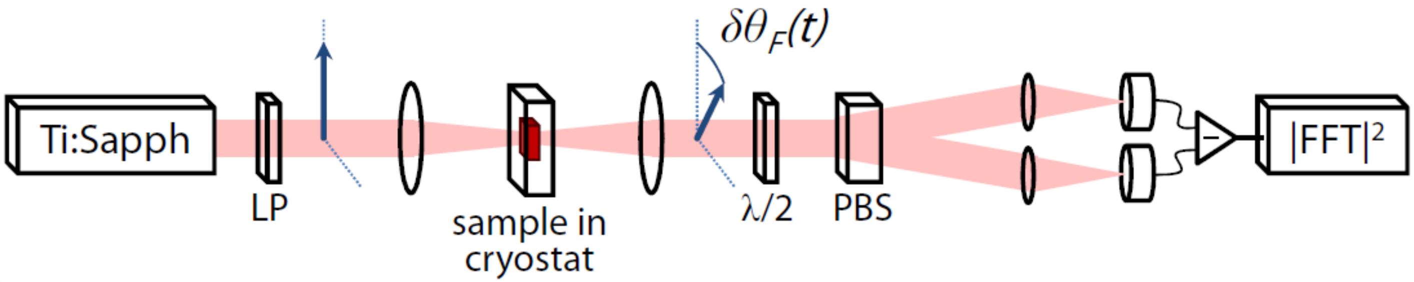


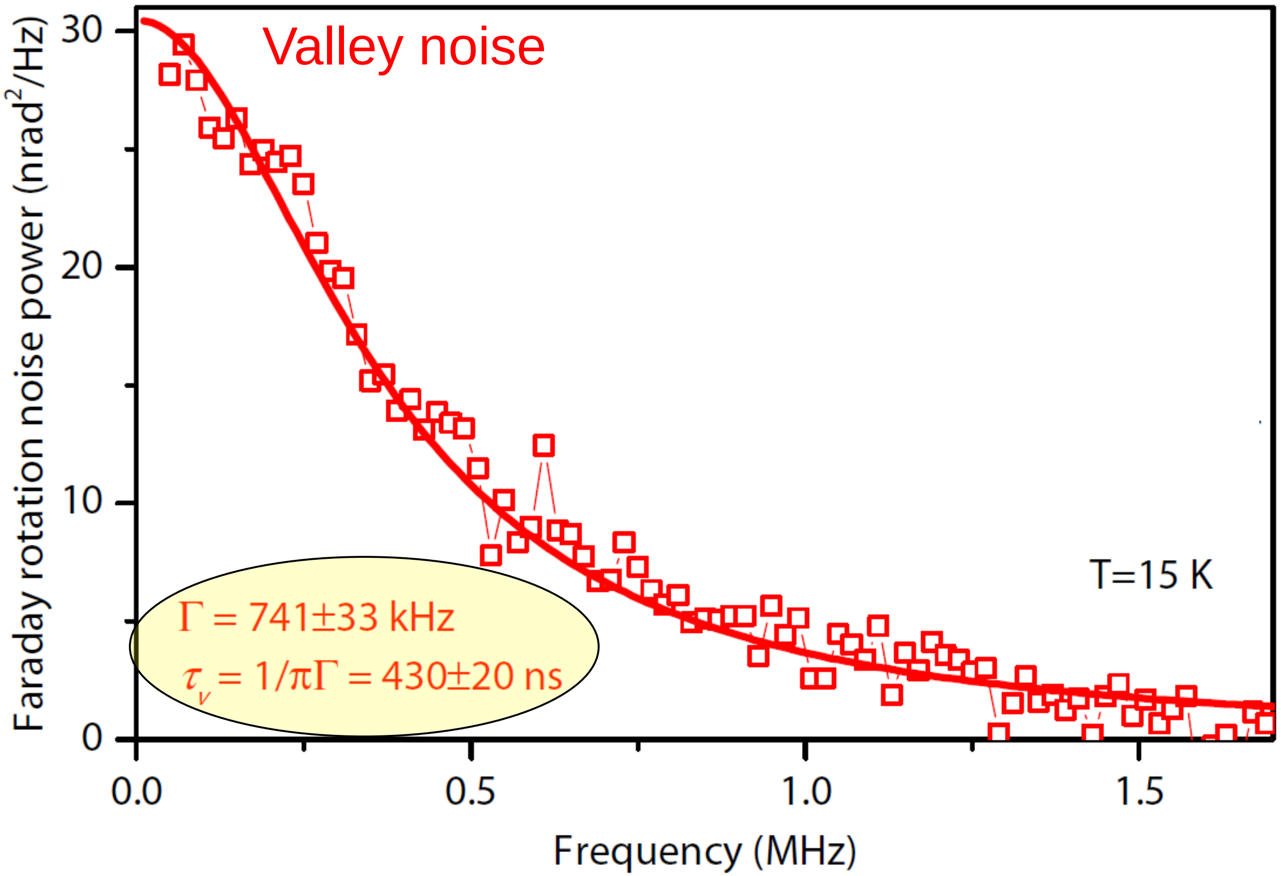
$\theta_K \sim (n_\uparrow - n'_\downarrow) \sim (S_z + N_v)/2$

Measure carrier spin/valley pol., long after minority species are gone

"Listening" to valley noise of holes in monolayer WSe_2



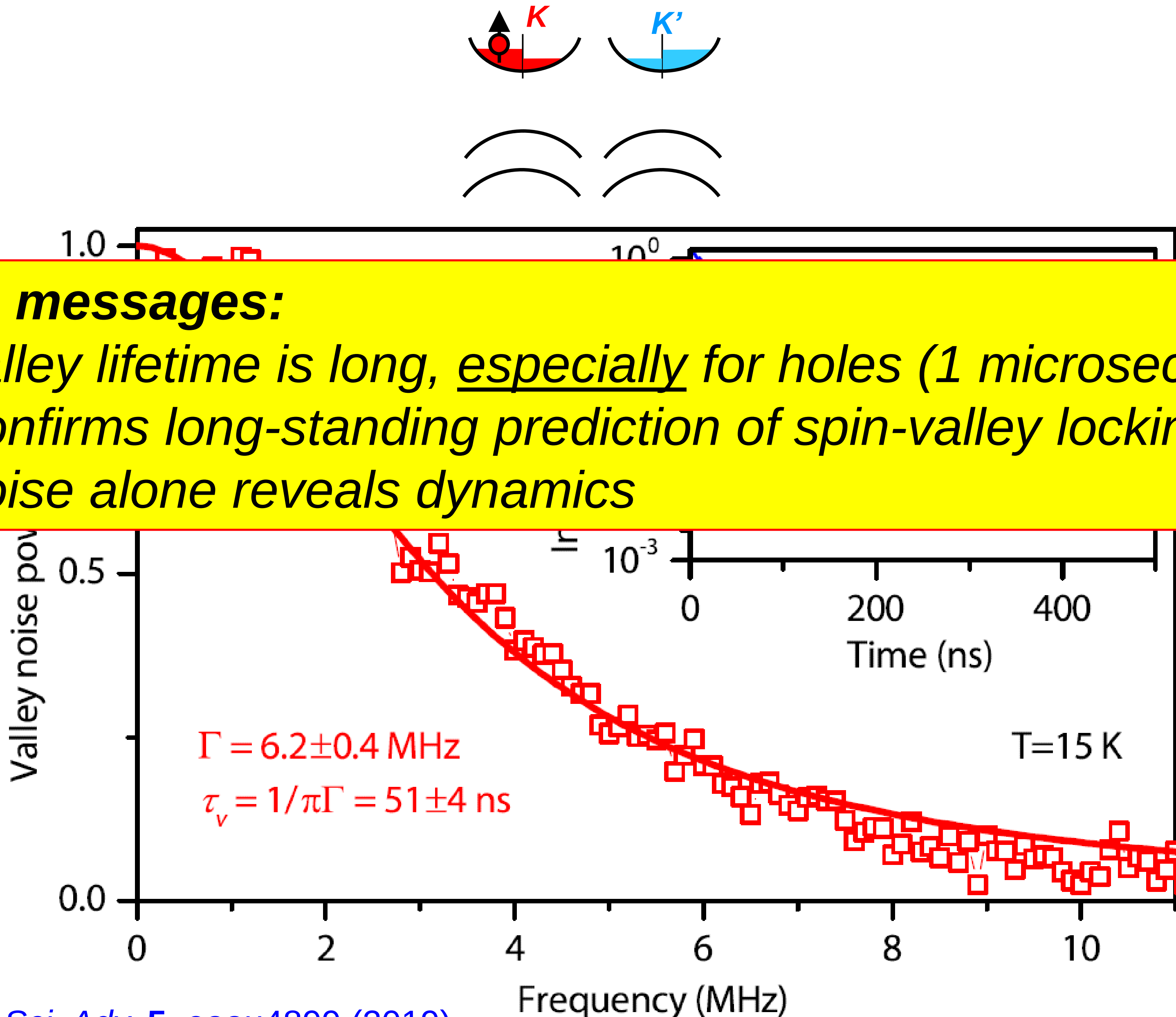




Nathan Wilson
(U Washington)

M. Goryca, *Sci. Adv.* **5**, eaau4899 (2019)

Electron valley noise in *n*-type monolayer WSe_2



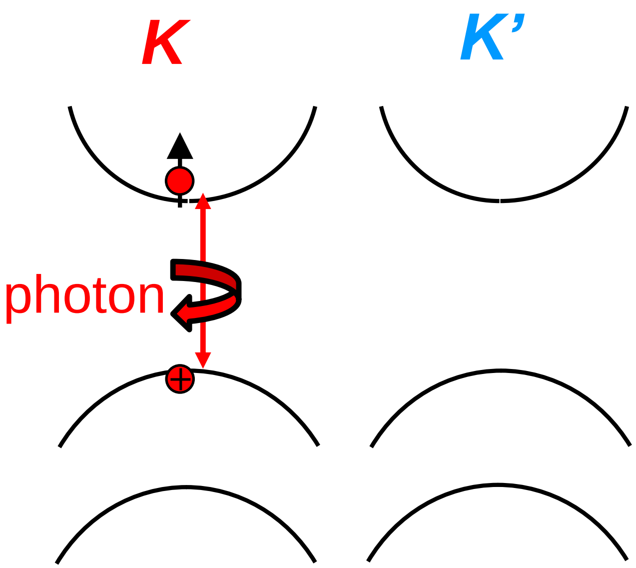
Main messages:

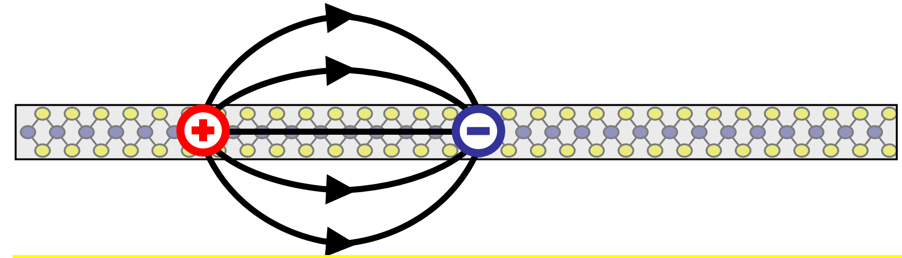
- *Valley lifetime is long, <u>especially</u> for holes (1 microsecond)*
- *Confirms long-standing prediction of spin-valley locking*
- *Noise alone reveals dynamics*

M. Goryca, *Sci. Adv.* **5**, eaau4899 (2019)

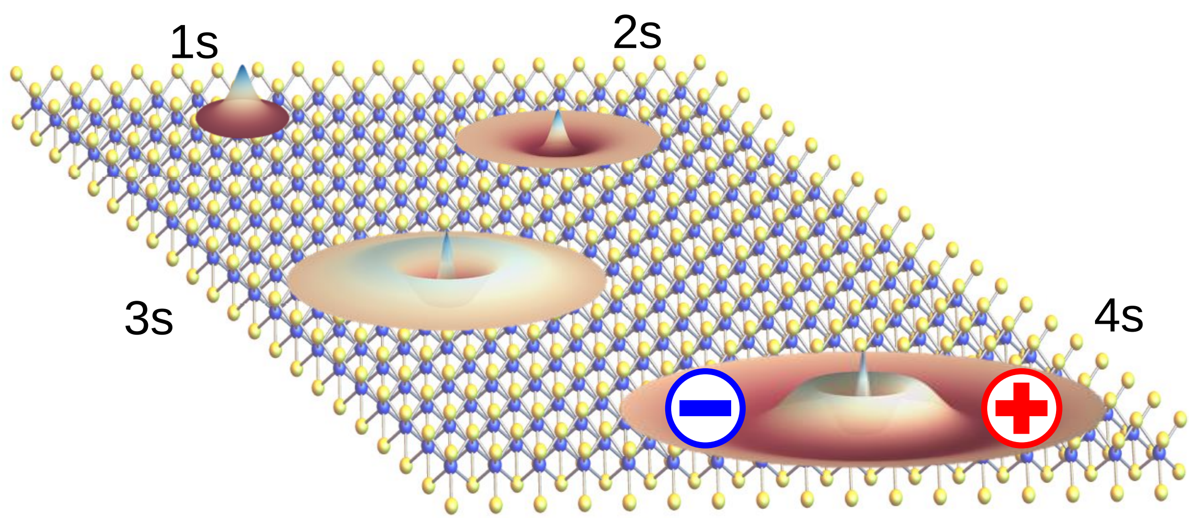
Part II: Electron-hole correlations in monolayer semiconductors

- **Easy: What happens when you put in 1 hole and 1 electron?**
- **Hard: What happens when you put 1 hole into a Fermi sea of ~10^{12} e^-?**



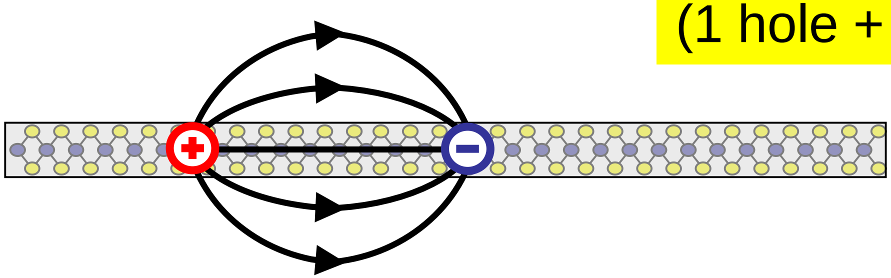


Exciton: Coulomb-bound *e-h* pair
Huge binding energy: 200-500 meV



Magneto-optical spectroscopy of Rydberg excitons in an <u>undoped</u> WSe_2 monolayer

(1 hole + 1 electron)



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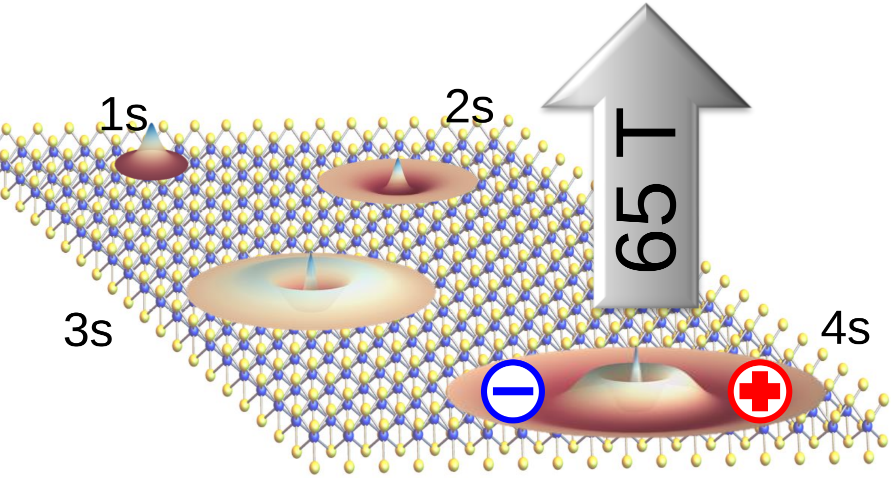

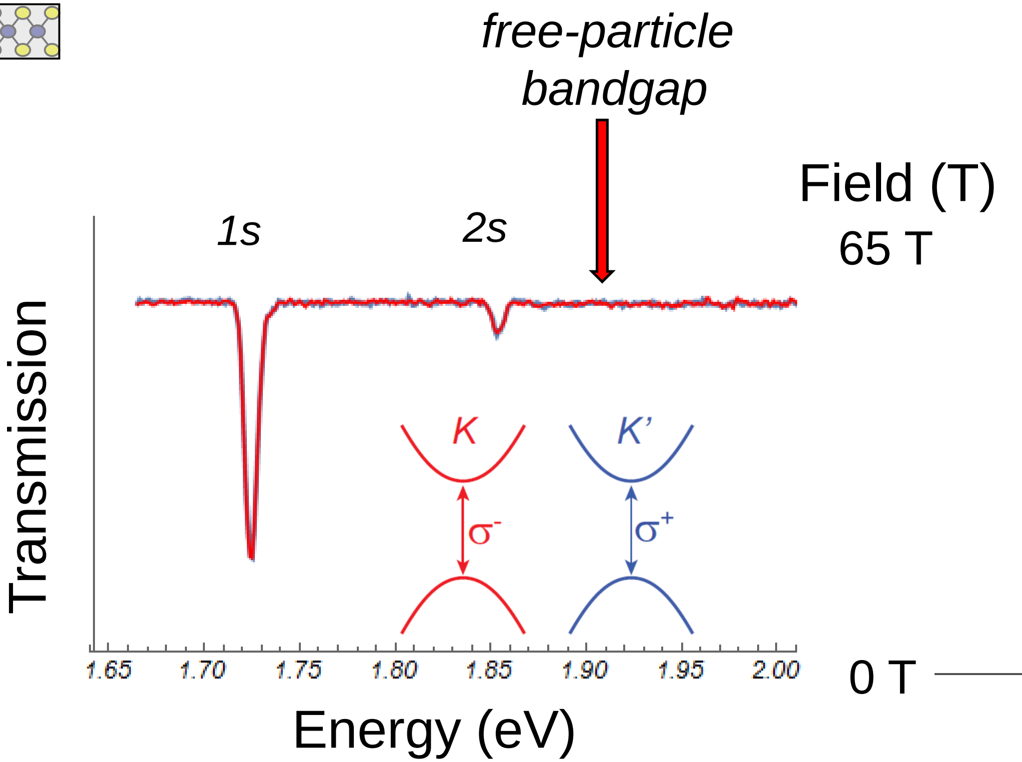

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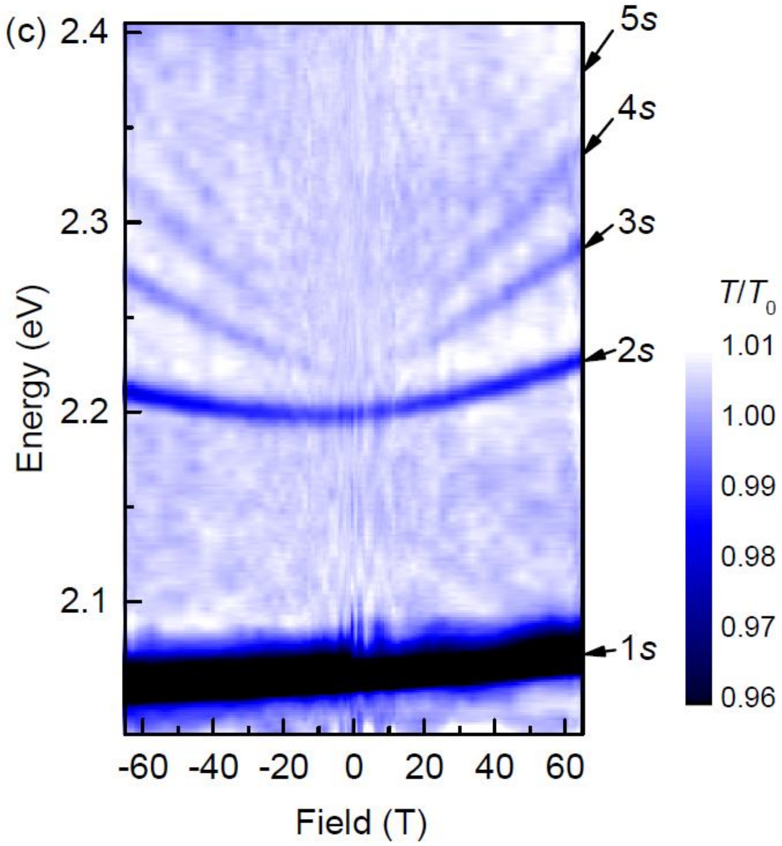
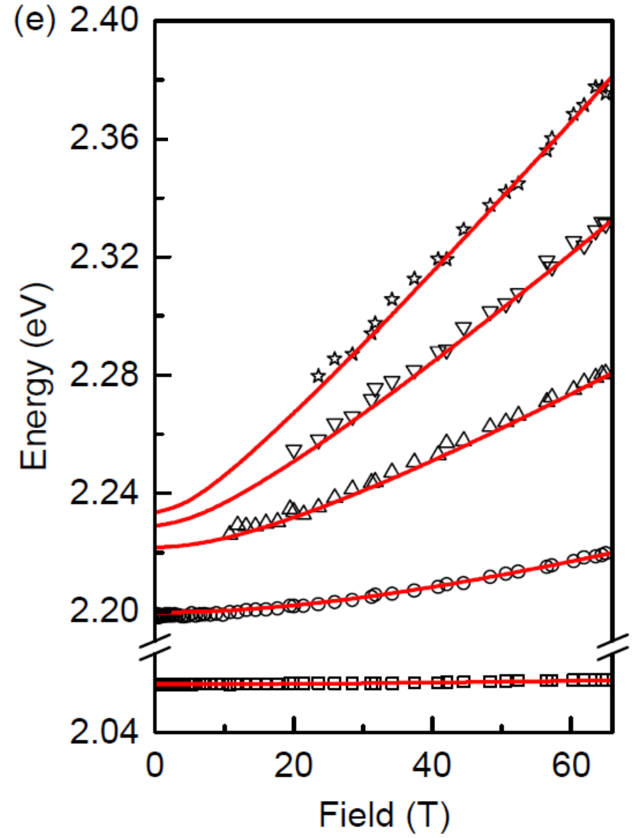
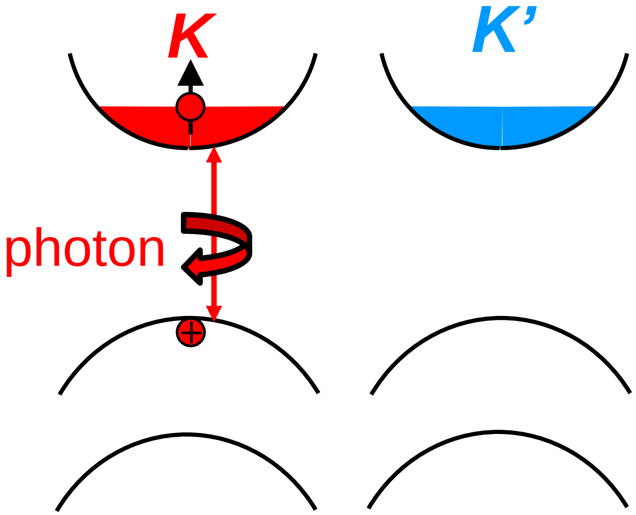


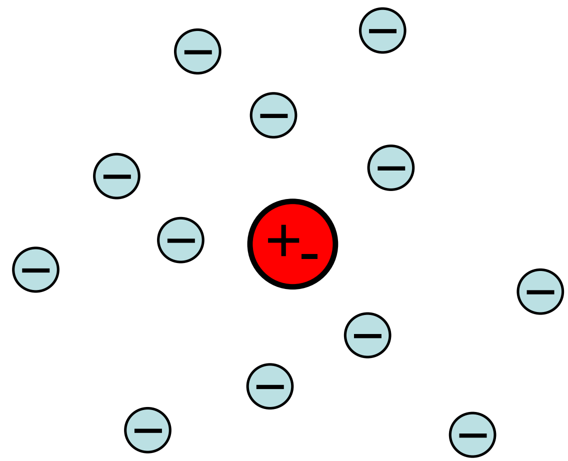


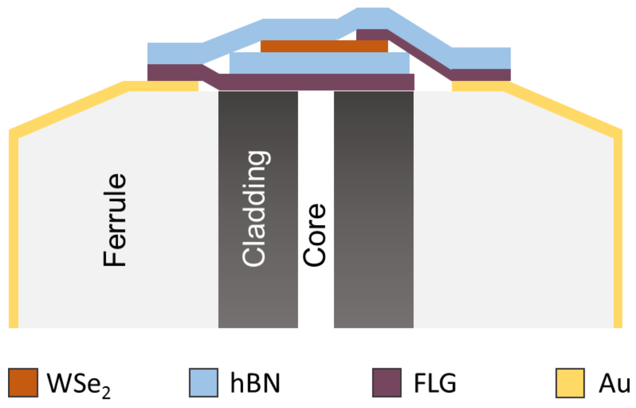
Stier et al., PRL **120**, 057405 (2018)
Goryca et al., arXiv:1904.03238 (2019)

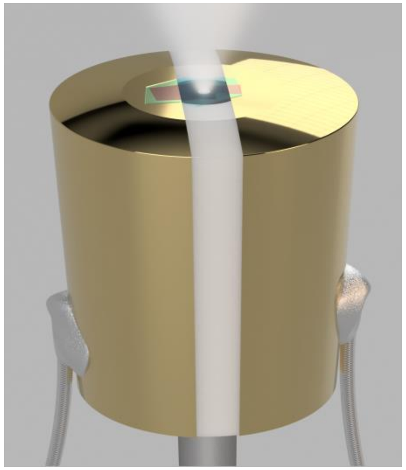
Spectroscopy of a *doped* WSe_2 monolayer

Hard question: 1 hole + *many* electrons (10^{12}-10^{13} /cm^2)



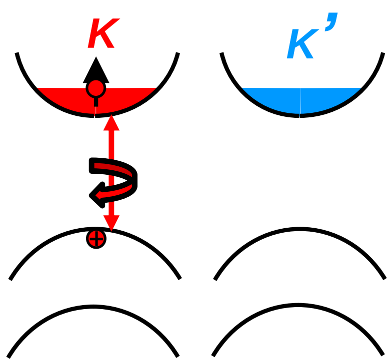


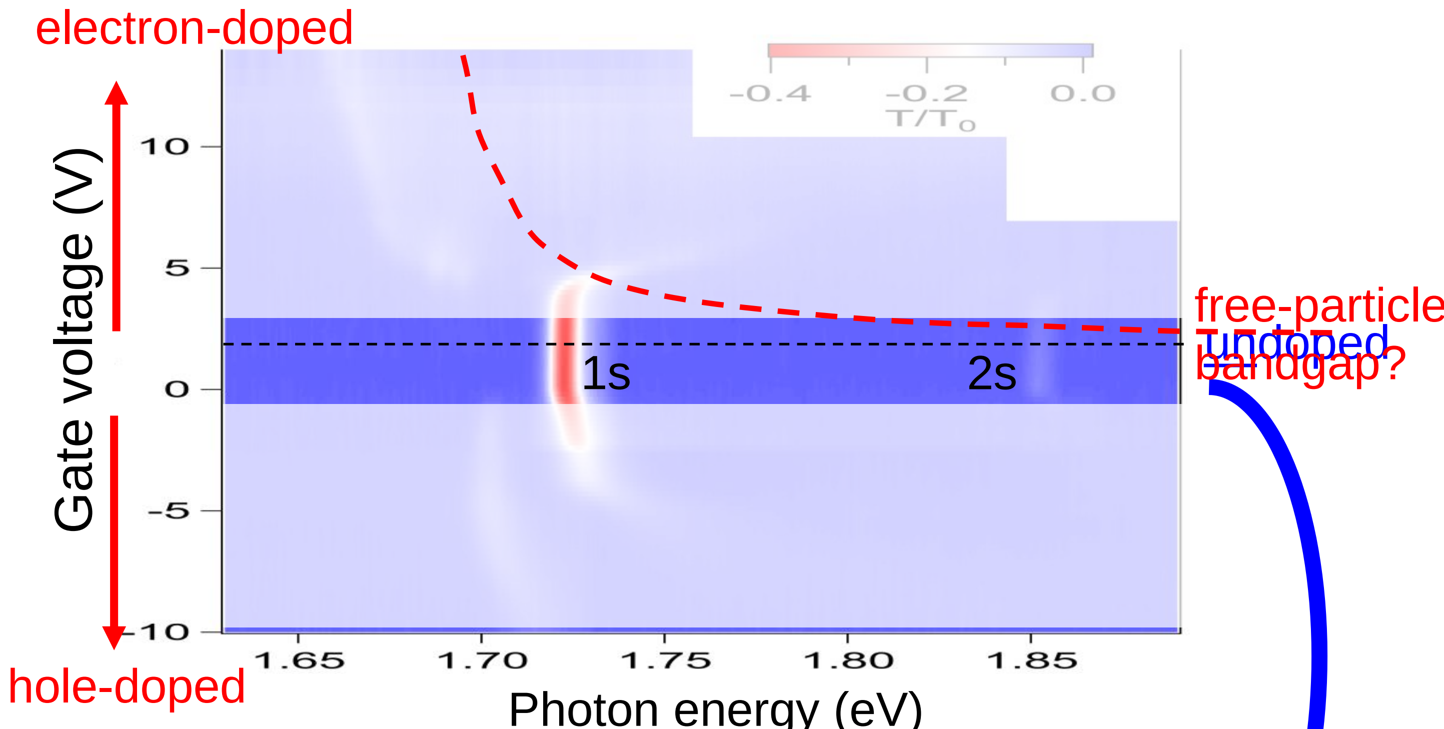


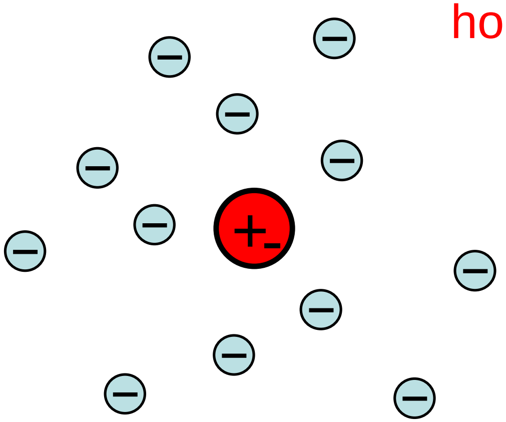


Spectroscopy of a *doped* WSe_2 monolayer

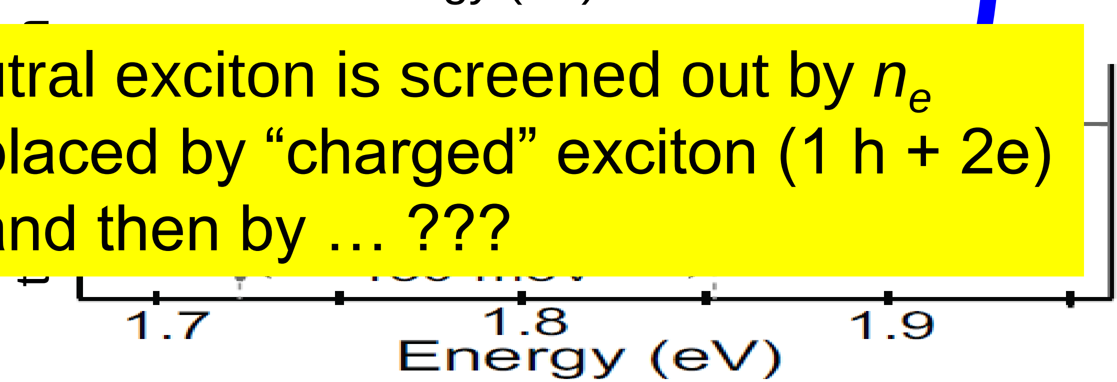
1 hole + *many* electrons (10^{12} - 10^{13} /cm^2)



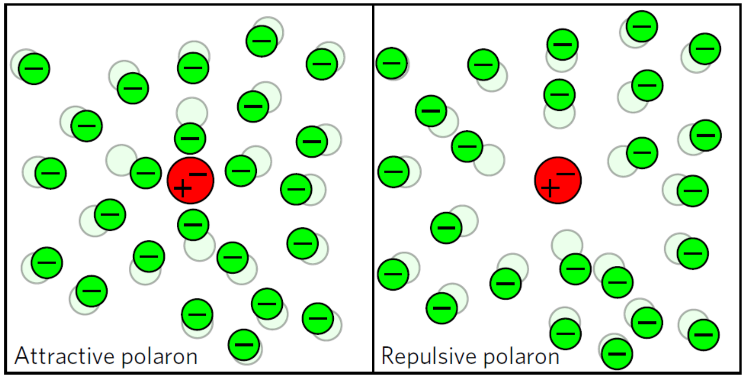


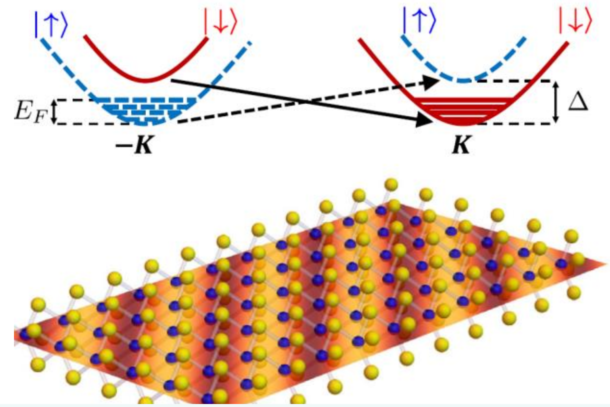


- Neutral exciton is screened out by n_e
- Replaced by "charged" exciton (1 h + 2e)
- … and then by … ???



How is a single hole correlated w/ (many) electrons?



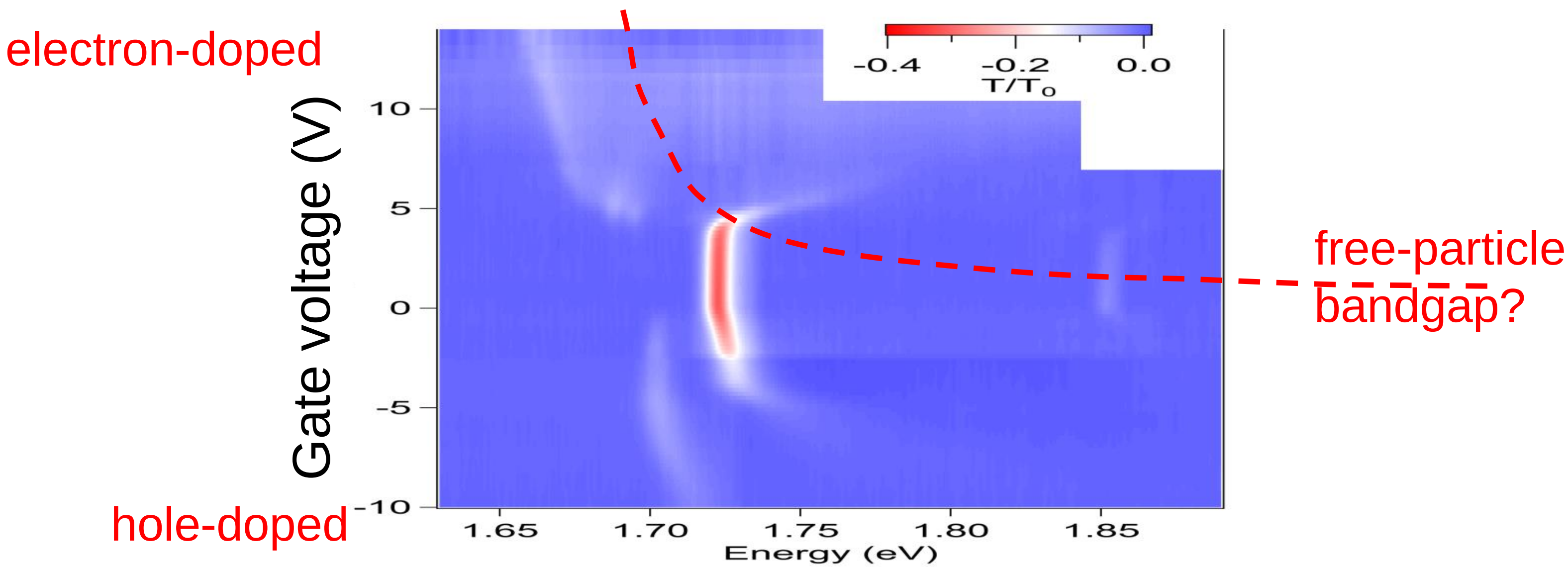


Some models based on picture of attractive/ repulsive *polarons*…

Sidler, Nature Phys. **13**, 255 (2017)
Efimkin, PRB **95**, 035417 (2017)

…while others consider *inter-valley coupling* to short-wavelength *plasmons*

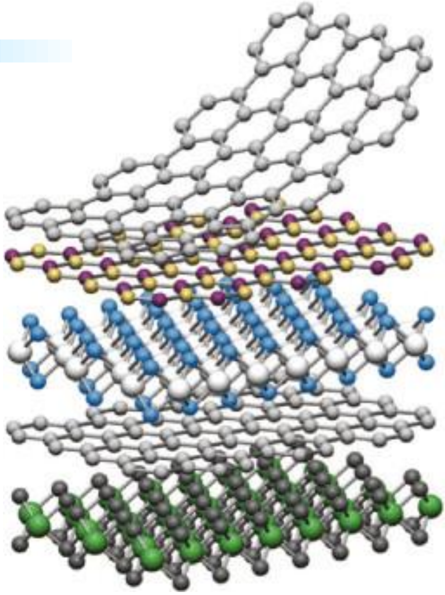
van Tuan, PRX **7**, 041040 (2017)



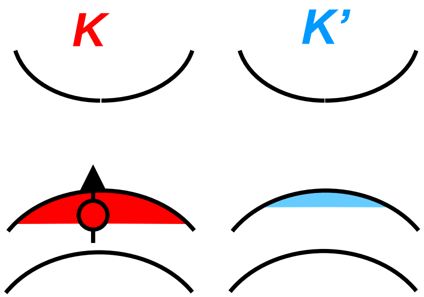
Summary

- **Thermodynamic "valley noise"**
  - *Noise correlations: access to intrinsic valley dynamics*
  - *Very long (~μs) spin/valley polarization of holes*
  - *Validates spin-valley locking prediction*

M. Goryca, *Sci. Adv.* **5**, eaau4899 (2019)



- **Electron & intervalley correlations probed by spectroscopy**
  - *Reveals screening of excitons by 2DEG*
  - *Formation of new bound quasiparticle states*
  - *Intervalley correlations (gapped plasmon spectrum)*



Thank you!

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