Quantum optics and magnetism in 2D materials

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Presented by: Martin Kroner

Motivation:

A new regime of cavity-QED with nonperturbative interactions between photons/polaritons and itinerant electrons or magnons?

Outline

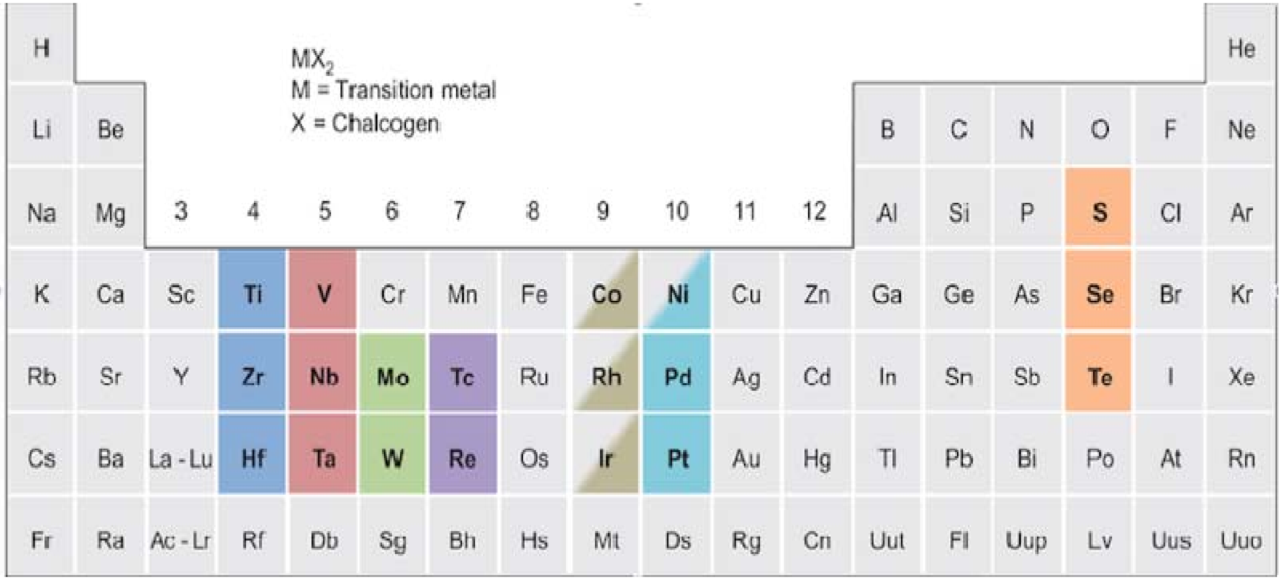
1) Strong light-matter coupling in semiconducting transition metal dichalcogenide (TMD) monolayers:
   - Realization of an atomically thick mirror using monolayer $MoSe_2$
   - Observation of robust exciton-polaritons

2) Strong exciton-electron interaction in TMDs:
   - <u>exciton-polarons</u> as elementary many-body optical excitations
   - nonequilibrium dynamics of a mobile quantum impurity

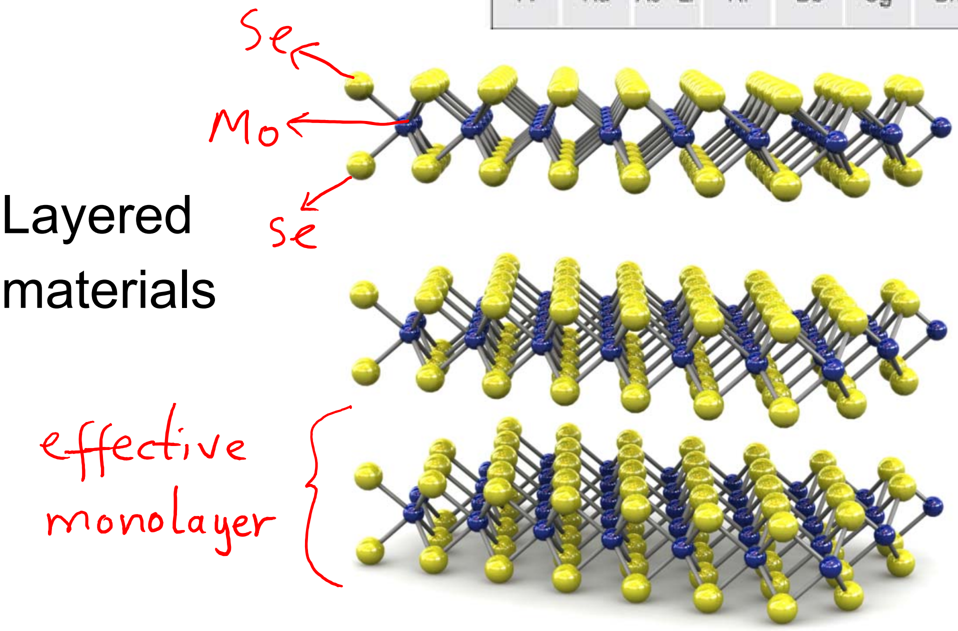
3) Giant valley/spin susceptibility in monolayer $MoSe_2$

A new class of 2D materials: Transition metal dichalcogenides (TMD)

Formula: MX_2
M = Transition Metal
X = Chalcogen



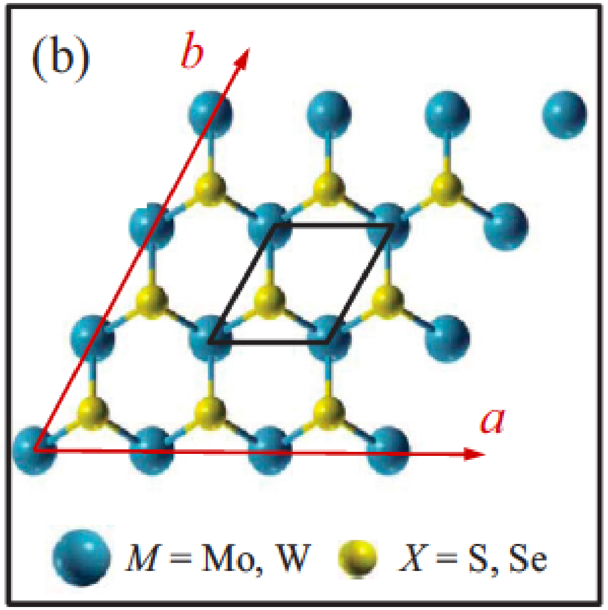
Layered materials



Electrical property	Material
Semiconducting	MoS_2 $MoSe_2$ WS_2 WSe_2 $MoTe_2$ WTe_2
Semimetallic	TiS_2 $TiSe_2$
Ferromagnetic	CrI_3, $CrBr_3$
Metallic, CDW, Superconducting	$NbSe_2$ NbS_2 $NbTe_2$ TaS_2 $TaSe_2$ $TaTe_2$

Transition metal dichalcogenides (TMD)

- Monolayer TMD has a honeycomb lattice
- Unlike graphene, inversion symmetry is broken
- Valley semiconductor: physics in ±K valleys

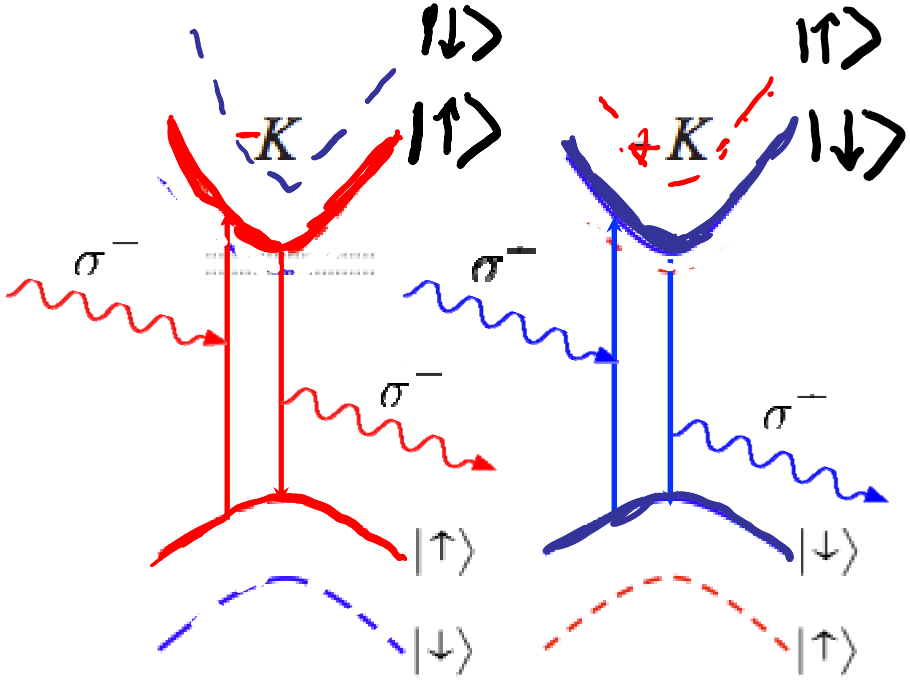


$$H_K = v\begin{pmatrix} \Delta/2v & p_x - ip_y \\ p_x + ip_y & -\Delta/2v \end{pmatrix}$$

(unlike graphene, 2-band model only provides a qualitative description)

- Monolayers of TMDs can be combined with other 2D materials to make van der Waals heterostructures with novel properties

2D semiconductor with optically addressable valley pseudospin degree of freedom



±K valleys respond to ±σ polarized light
→ optical valley addressability

Finite Berry curvature leads to valley Hall effect & modifies optical spectrum

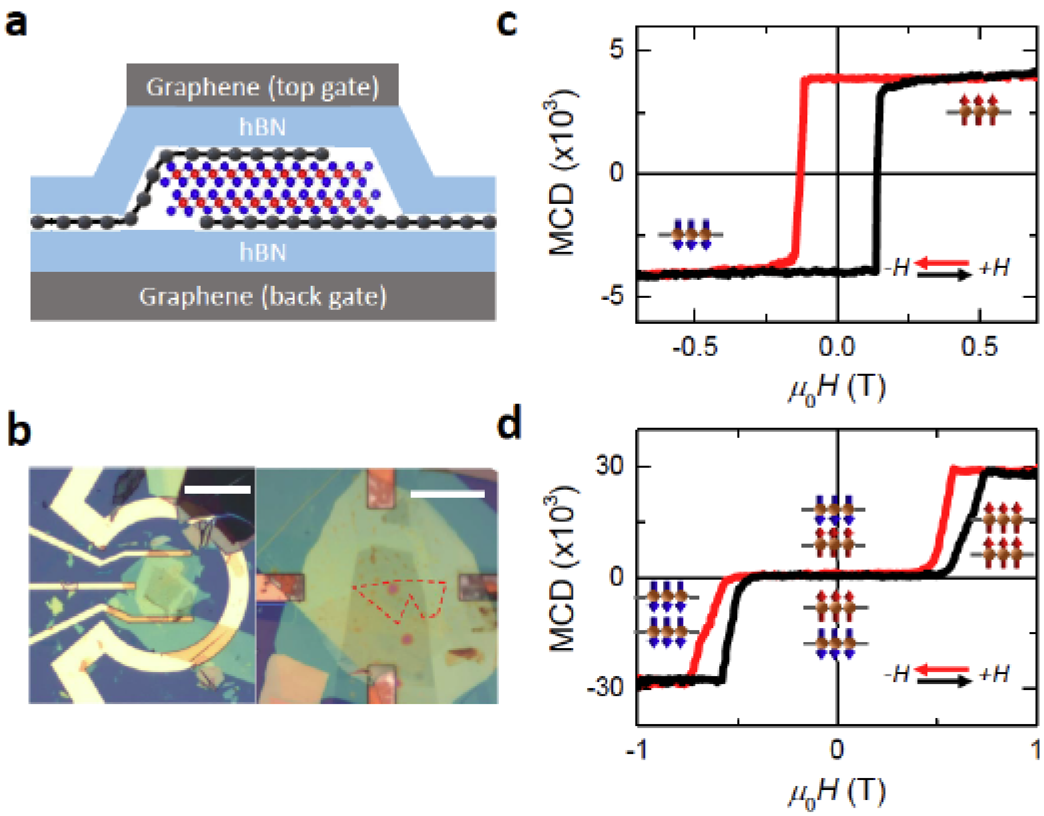
Spin-orbit leads to spin-valley locking

For this talk: a 2D charge tunable valley semiconductor with strongly bound excitons – emphasis on interactions

Pioneering work: Heinz, Xu

Magnetic 2D materials

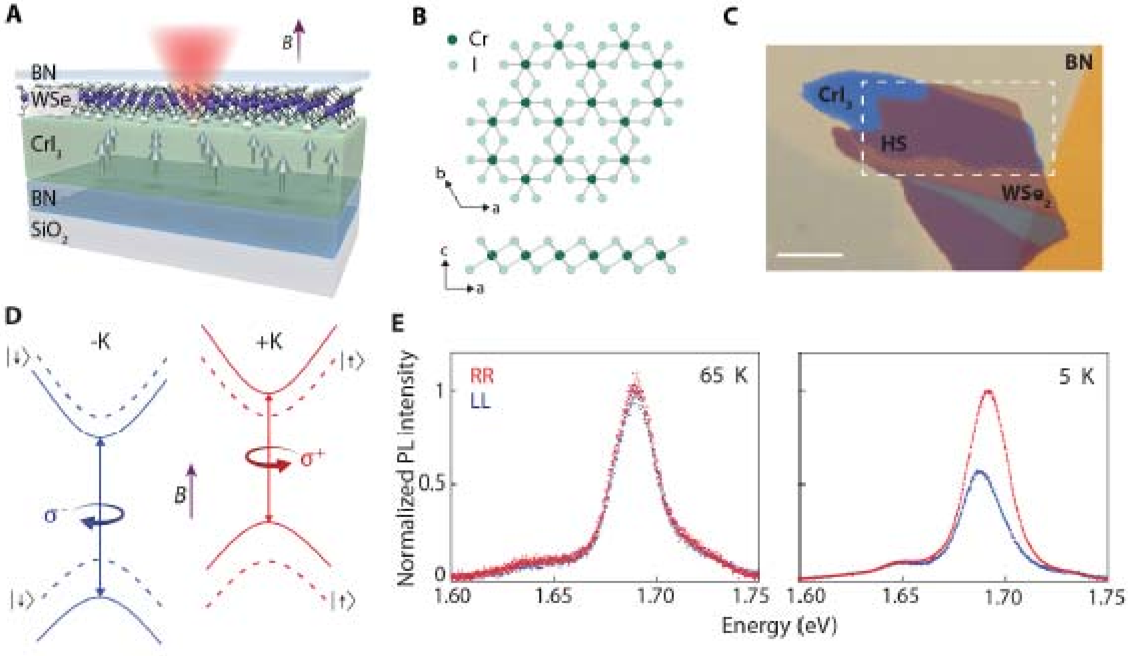
(Mak & Shan)



- Magnetic material with a bandgap of 2 eV

TMD-magnetic heterostructures

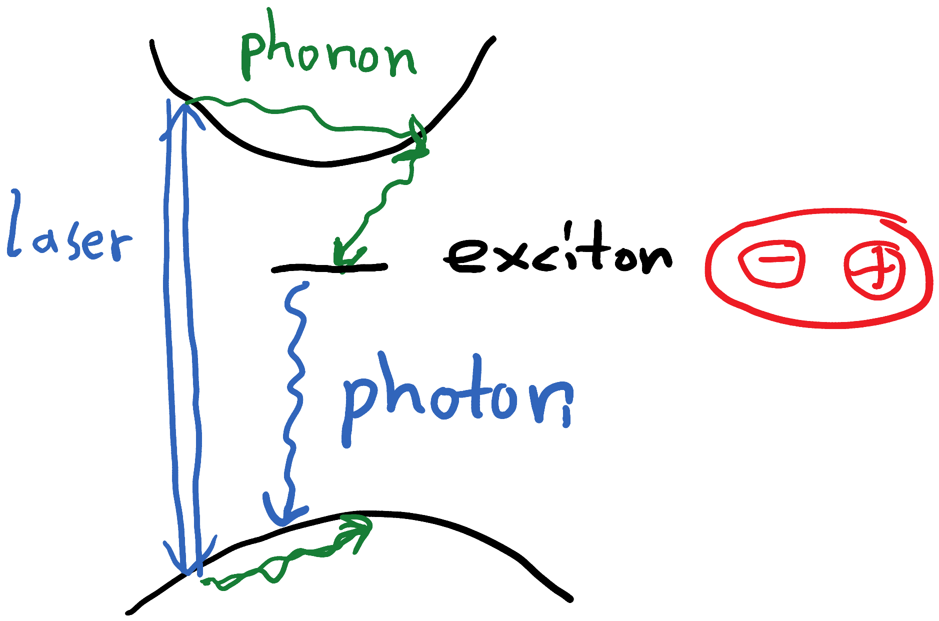
(Xu)



- Exchange fields up to 13 T in WSe2

Photoluminescence (PL) from 2D materials

- Due to strong Coulomb interactions, electrons and holes form strongly bound states before they recombine: PL is dominated by decay of an exciton.



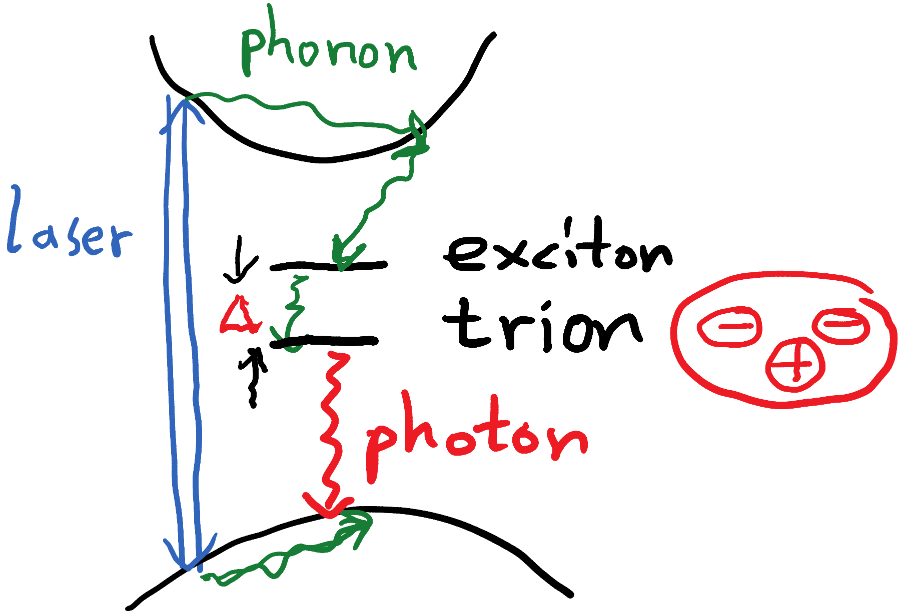
Exciton binding energy:

~ 10 meV, band-gap ~1.5 eV (GaAs)

~ 0.5 eV, band-gap ~ 2.0 eV (TMD)

Photoluminescence (PL) from 2D materials

- Due to strong Coulomb interactions, electrons and holes form strongly bound states before they recombine: PL is dominated by decay of an exciton or a trion if QW has localized electrons



Exciton binding energy:

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~ 0.5 eV, band-gap ~ 2.0 eV (TMD)

Exciton+electron form a trion, an H- like molecule with binding energy Δ ~ 1 meV (GaAs) ~ 25 meV (TMD)

Photoluminescence (PL) from 2D materials

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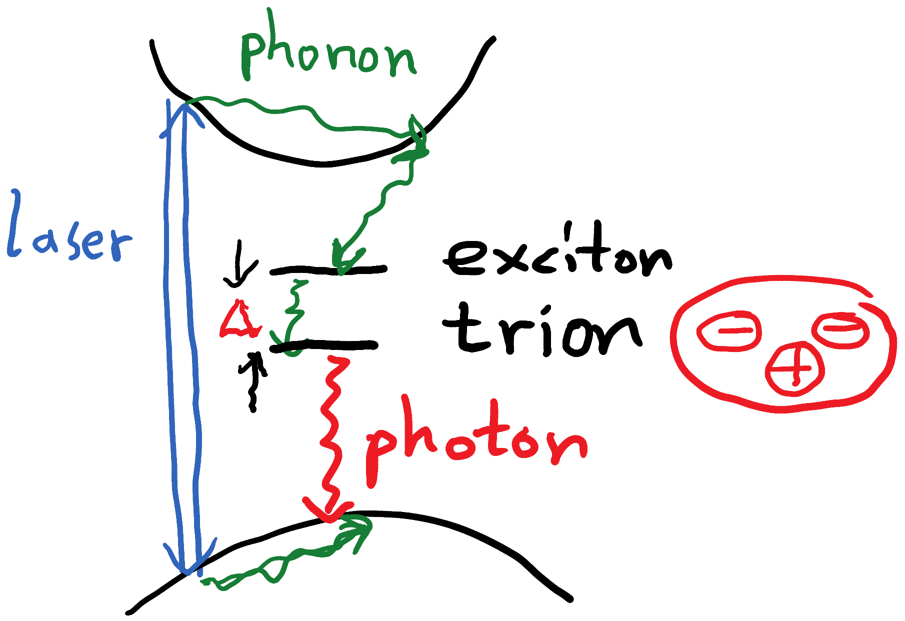
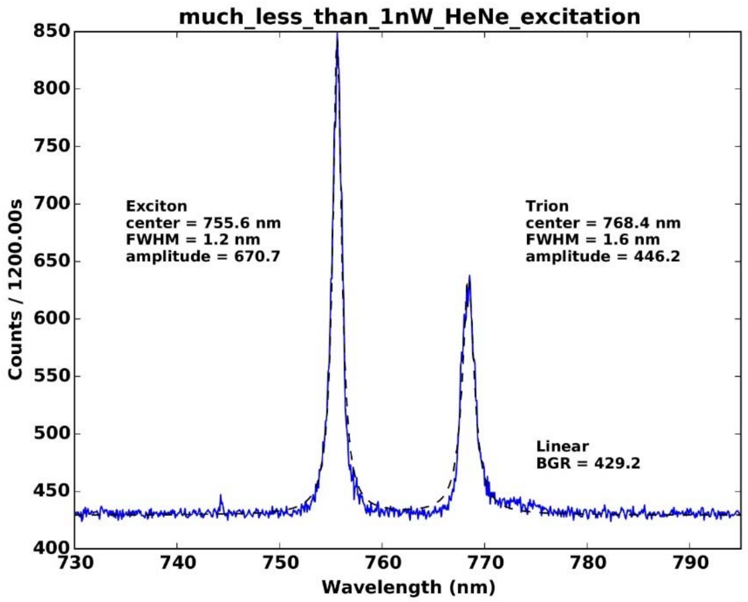


Exciton linewidth of MoSe2 in hBN is comparable to the radiative decay rate



Implications of strong exciton binding
≡ small Bohr radius a_B

- TMD excitons couple very strongly to resonant photons:

  - ultrafast /sub-ps radiative decay rate ($\sim 1/a_B^2$) $\Gamma_{rad} \sim 1.5$ meV

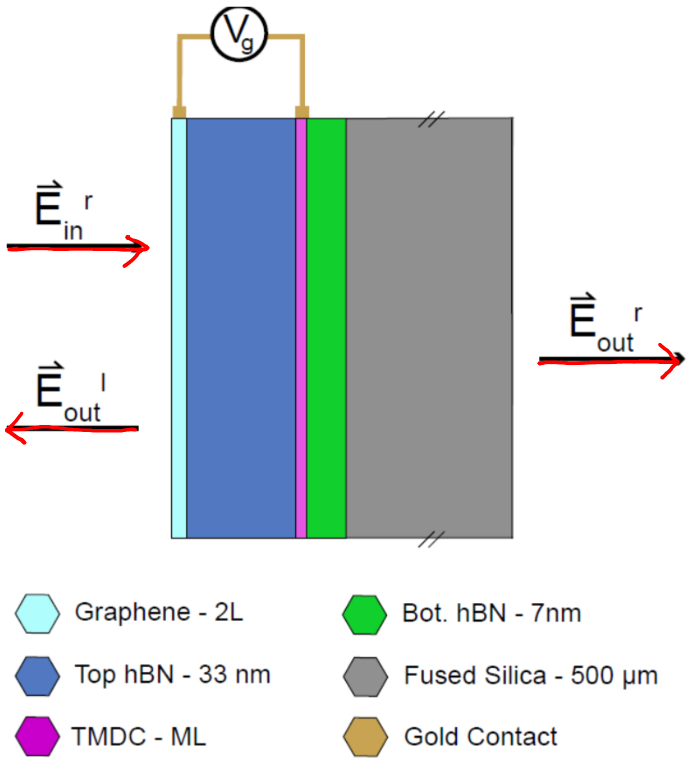
  - strong reversible coupling to cavities ($\sim 1/a_B$) $g \sim 10\text{-}40$ meV

- State-of-the art TMD monolayers have nearly radiative decay limited exciton linewidths

  - resonant coherent light scattering - not incoherent absorption!

Monolayer MoSe$_2$: atomicallly thin mirror?

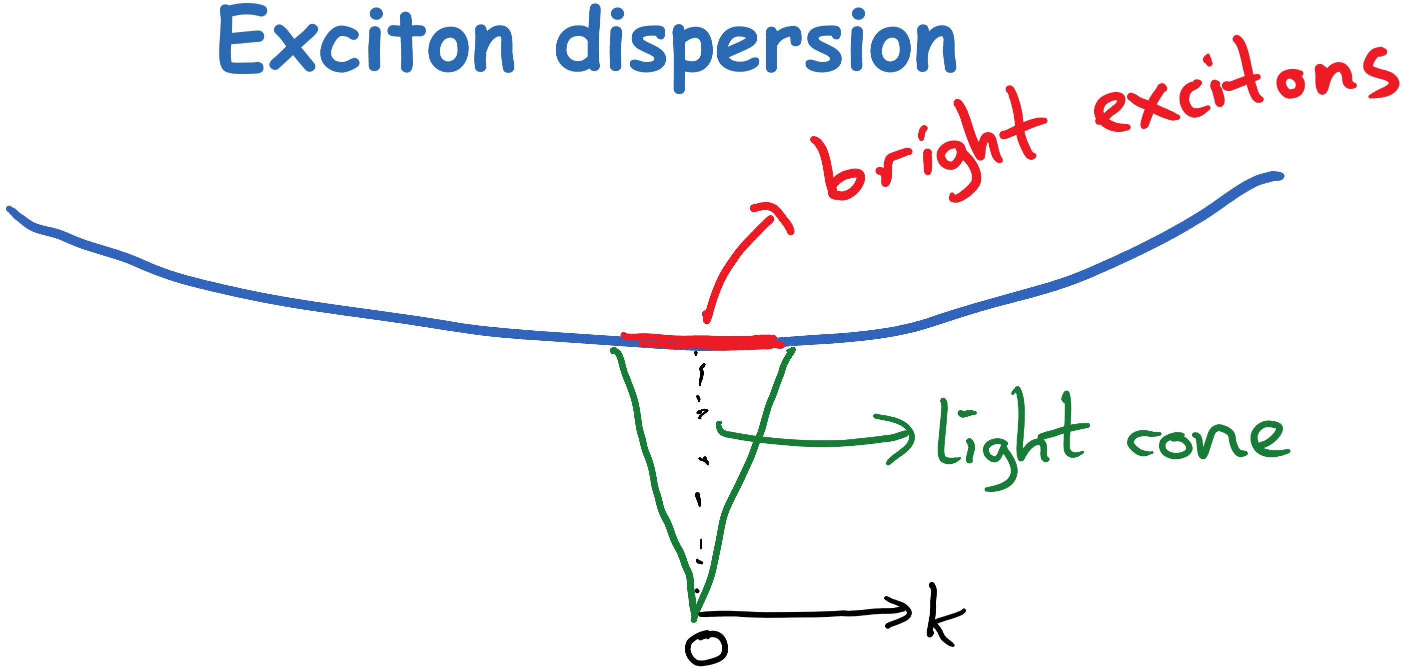
In-plane momentum conservation ensures that that for radiatively broadened 2D exciton resonance, incident resonant light experiences perfect 100% specular reflection



- High reflection or extinction of transmission on resonance only possible for spontaneous emission broadened excitons
- Equivalent to a single atom coupled to a 1D reservoir/waveguide

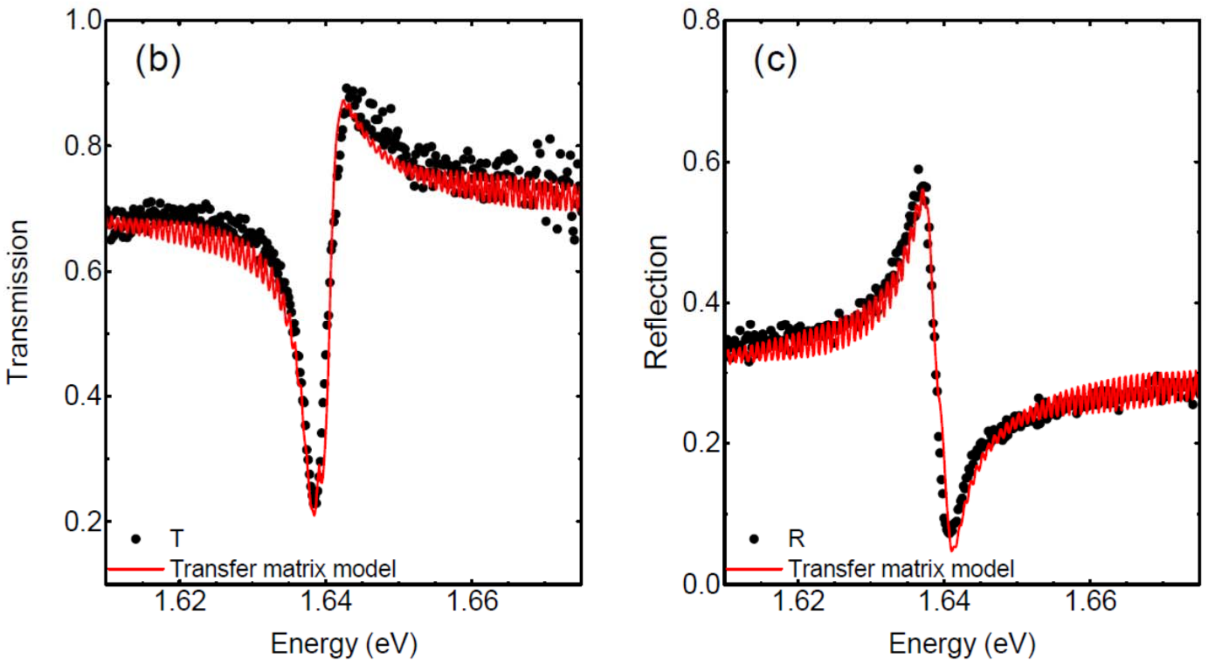
Theory: Zeytinoglu et al. arxiv 1701.08228; related work on atomic arrays: Adams & Lukin-Yelin groups

Exciton dispersion



- Incident photons with in-plane momentum k generate excitons with identical momentum (translational invariance)
- Secondary field generated by excitons interferes with the external field to modify transmission and lead to reflection
- Only excitons within the light cone couple to light; disorder scatters excitons to dark states – leads to real absorption

Realization of an atomicallly thin mirror

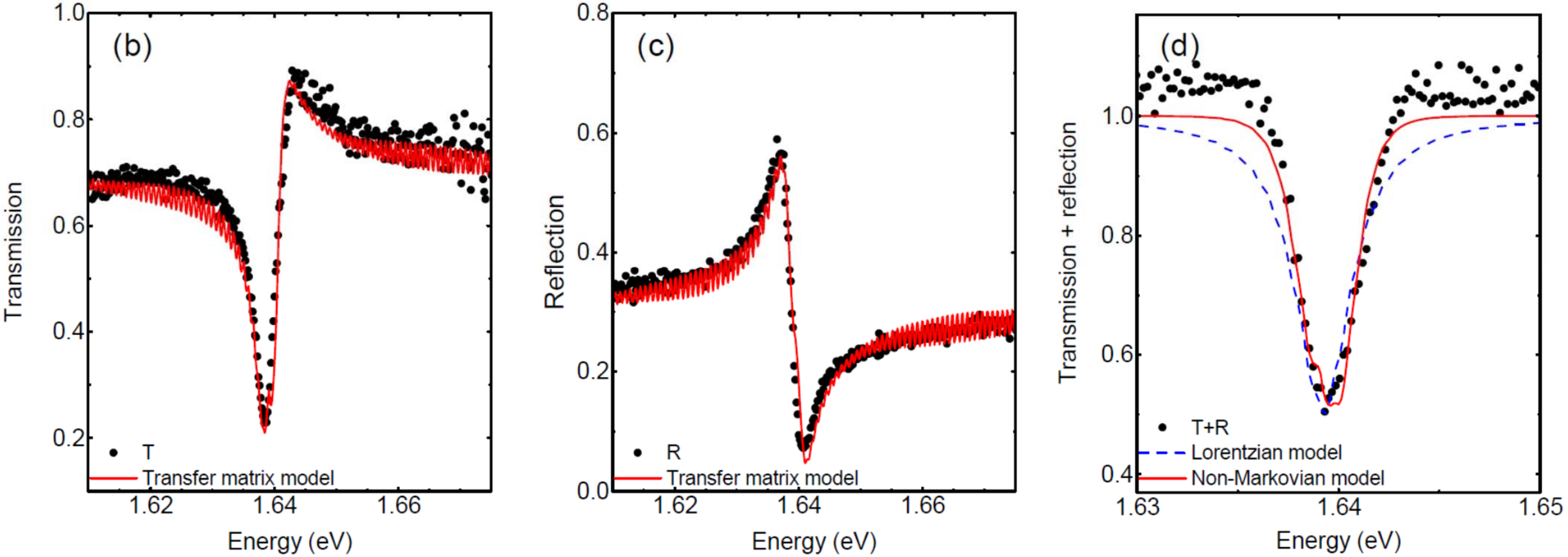


90% extinction of transmission
45% peak monolayer reflection

→ Demonstrates predominantly radiatively broadened excitons

(see also Kim-Lukin-Park & Shan-Mak results – up to 80% reflection)

Realization of an atomicallly thin mirror



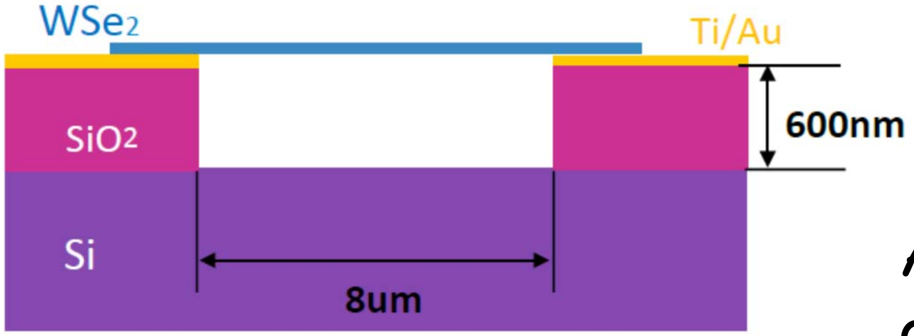
Sum of specular reflection & transmission: R+T

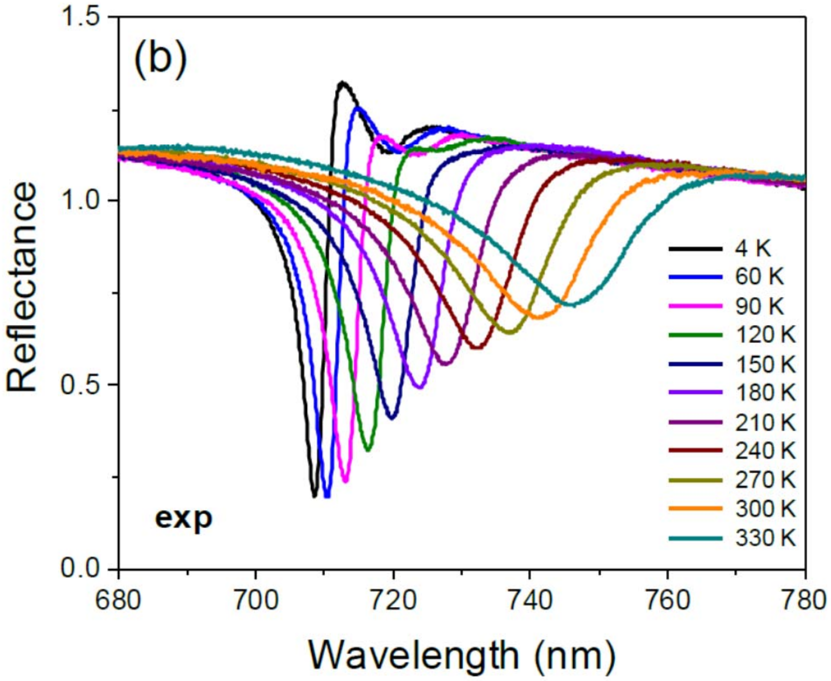
→ For pure radiative decay, we should have R+T = 1, for all ω_{inc}

→ Also, for pure inhomogeneous broadening R+T =1, for all ω_{inc}

→ R+T lineshape is non-Lorentzian – due to scattering into $k \neq 0$

A suspended atomically thin mirror (Mak & Shan)

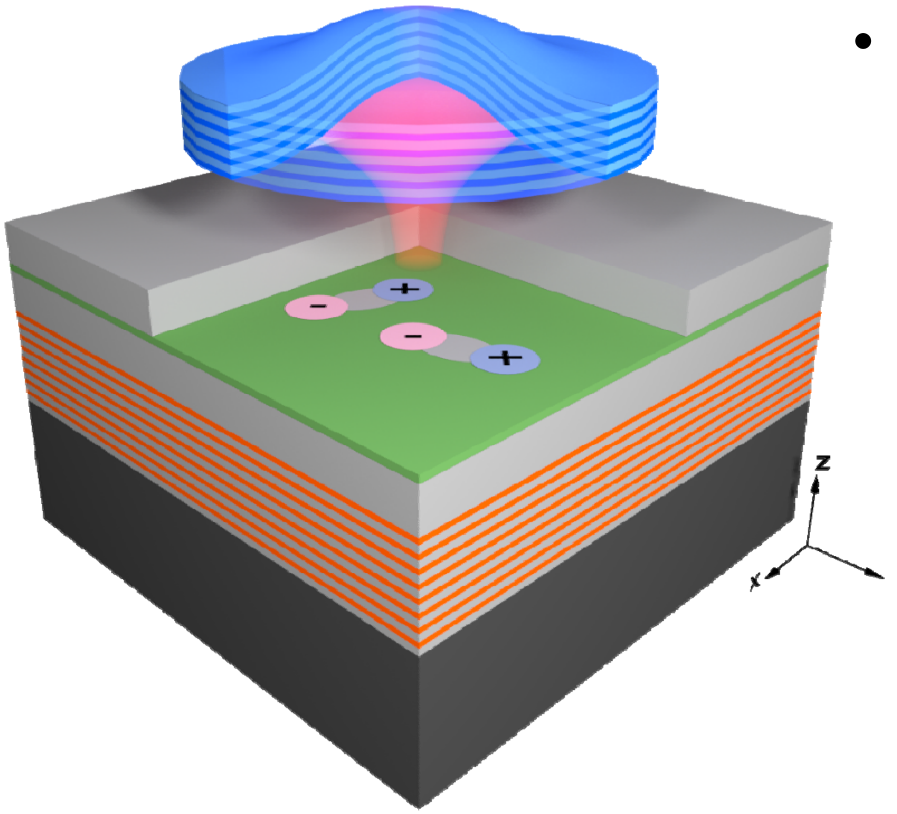


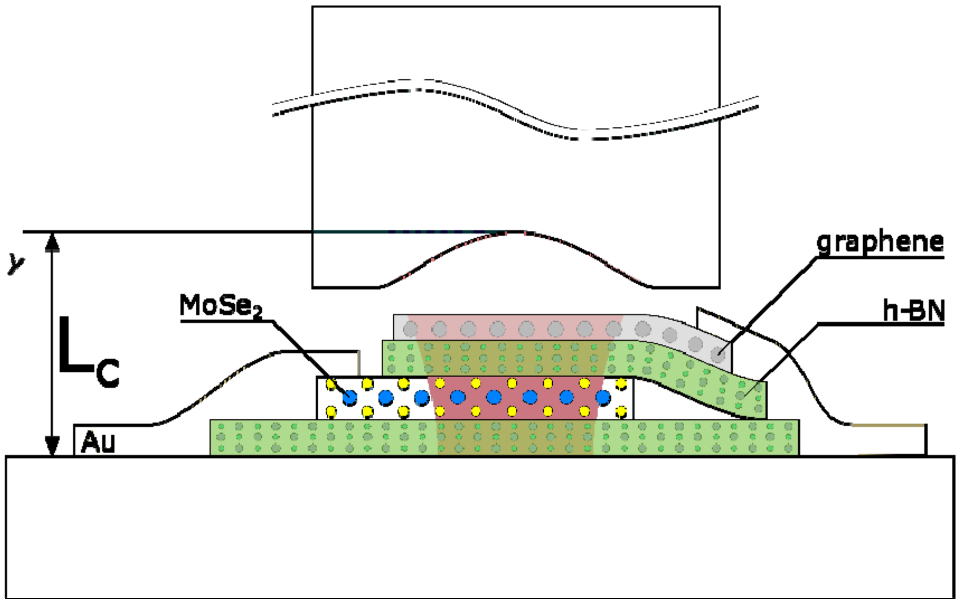


A new paradigm for optomechanics

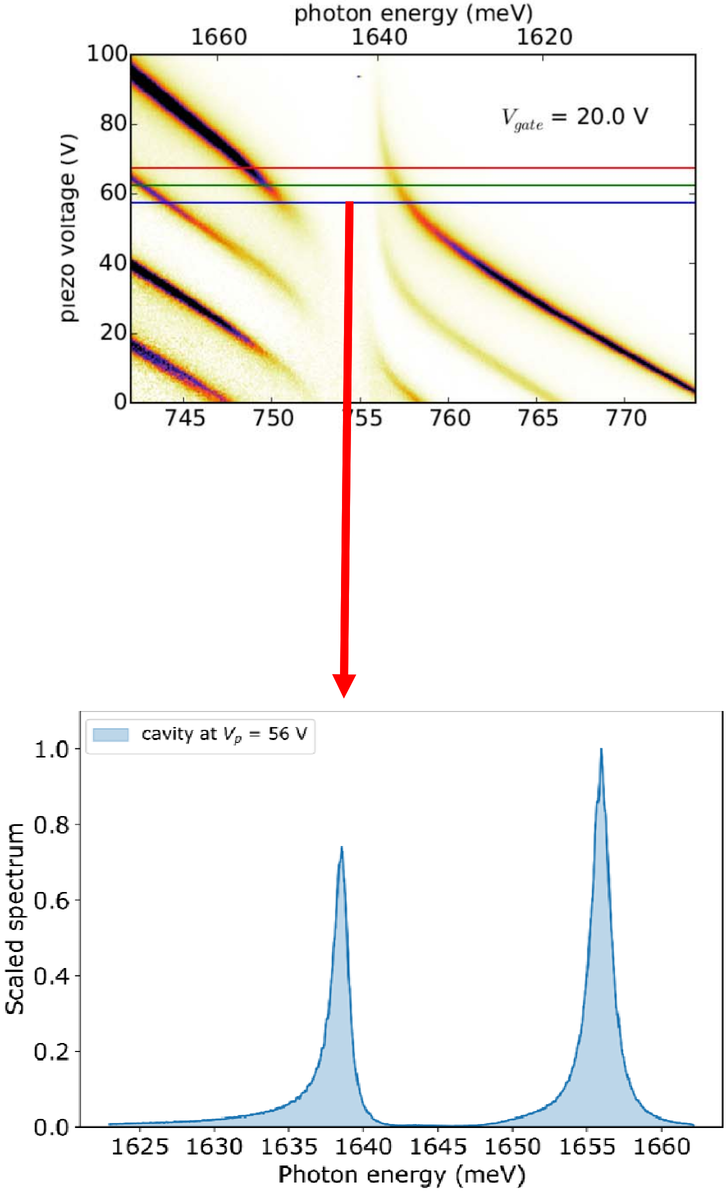
Cavity-polaritons with 2D materials

- Tunable vacuum field strength and long cavity lifetime allowing for high-precision spectroscopy
- Versatile platform for cavity-QED with any material system





Strong coupling regime



- Large normal mode splitting: Ω_R = 17 meV – new elementary excitations: exciton-polaritons
- Maximum reported splitting > 50 meV

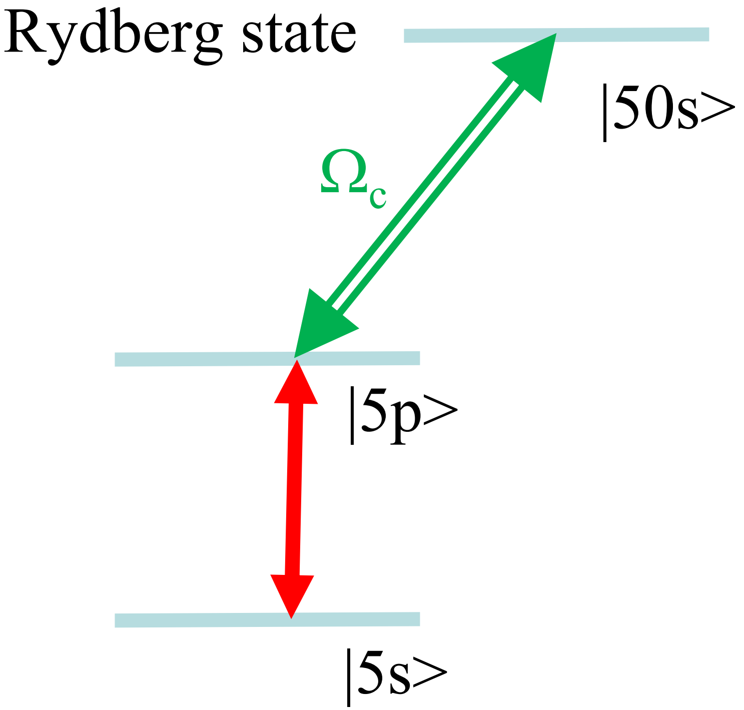
Earlier results:

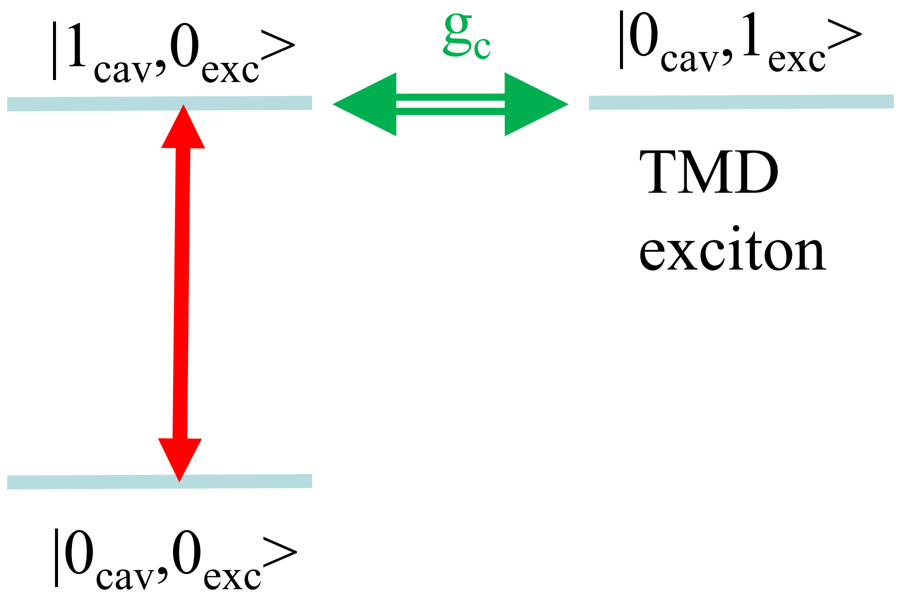
Menon, Tartakovskii

Rydberg blockade analog

Rydberg EIT/blockade

Polariton blockade





- Short exciton lifetime is irrelevant: the decay rate of polaritons is determined exclusively by their cavity nature – the more exciton like the polariton is, the longer the decay time

Optical excitations out of a 2DES

- Contrary to common wisdom, it is not possible to observe a (sharp) trion peak in absorption or emission from an ideal degenerate 2DES

- Direct formation of a trion in absorption is $\propto$ to the probability of finding a k~0 exciton in a strongly bound trion $\propto (k_{photon}a_B)^2$
- The radiative decay of a k=0 (lowest energy) trion has to produce a k=0 electron – Pauli-blocking when $E_F > 0$ (not the case for localized electrons)

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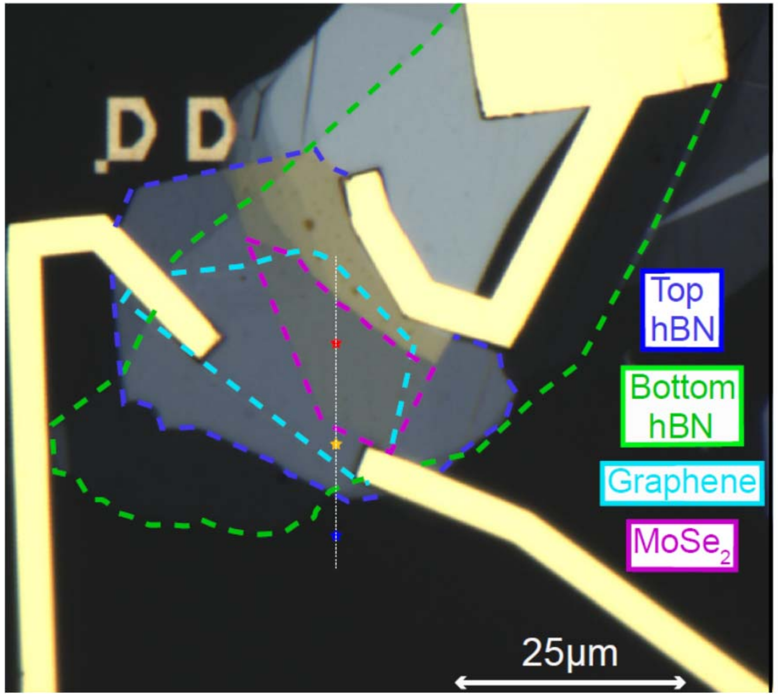
➢This talk: proper description of the optical excitation spectrum is provided by many-body excitations termed exciton-polarons

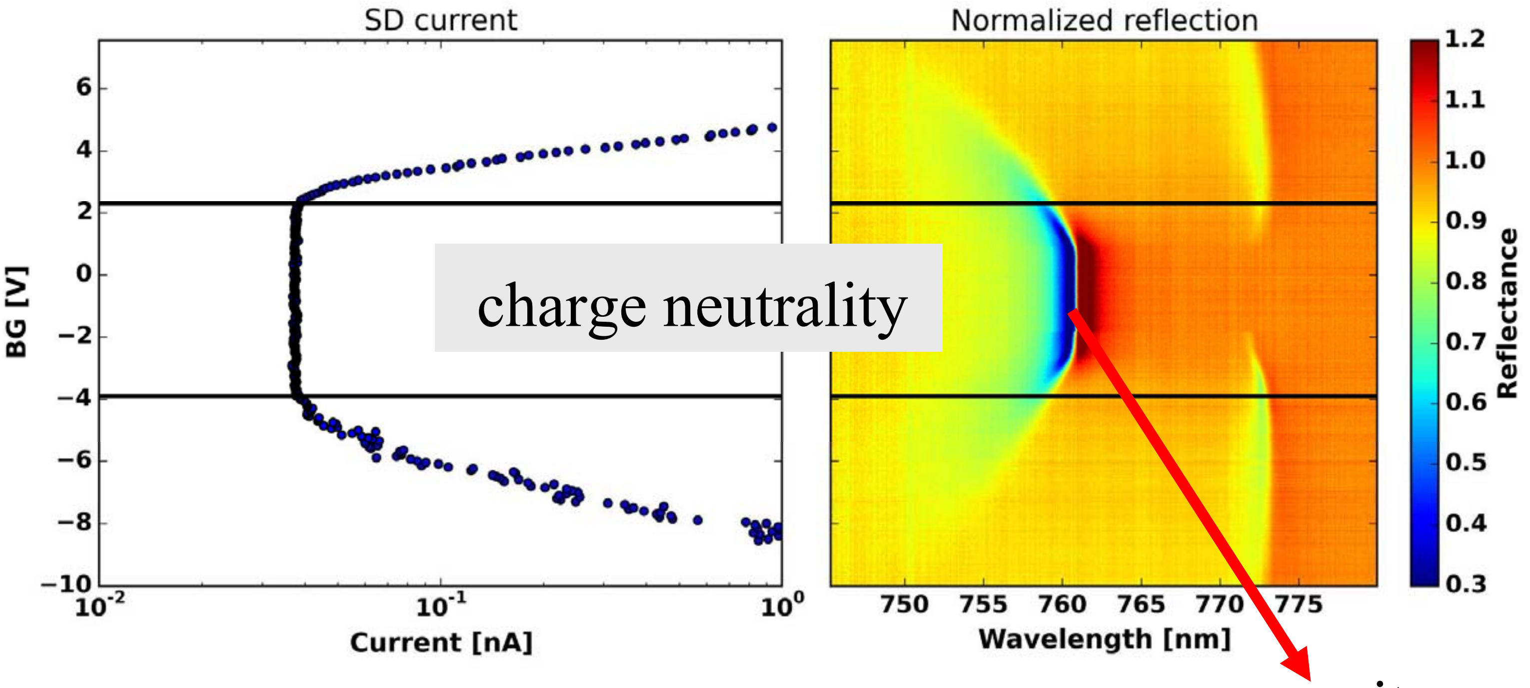
→ exciton as a finite-mass impurity in 2DES

Electrical control of optical properties



- A van der Waals heterostructure incorporating a graphene layer on top of hBN/MoSe2/hBN layers allow for controlling charge density
- Ideal for investigating exciton-electron interactions

Carrier density dependent reflection



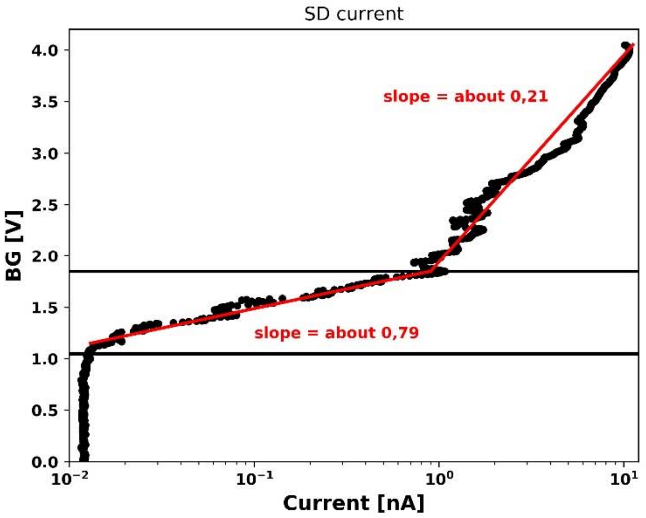
charge neutrality

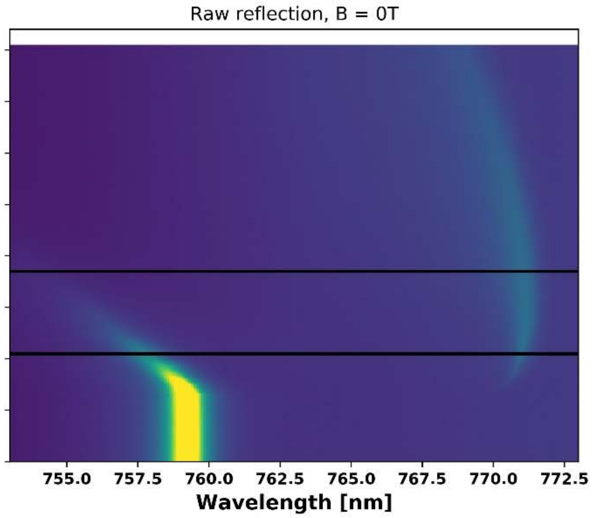
exciton

- Sharp increase in conductance indicates free carriers
- Reflection is strongly modified as electrons or holes injected

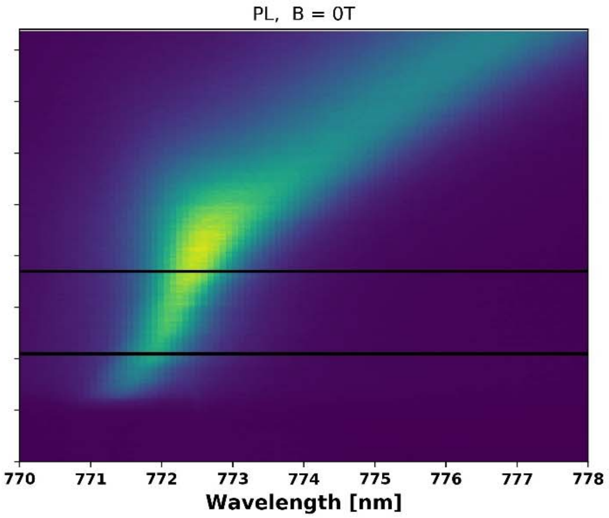
Carrier density dependent reflection & emission (PL)

Electron doped regime





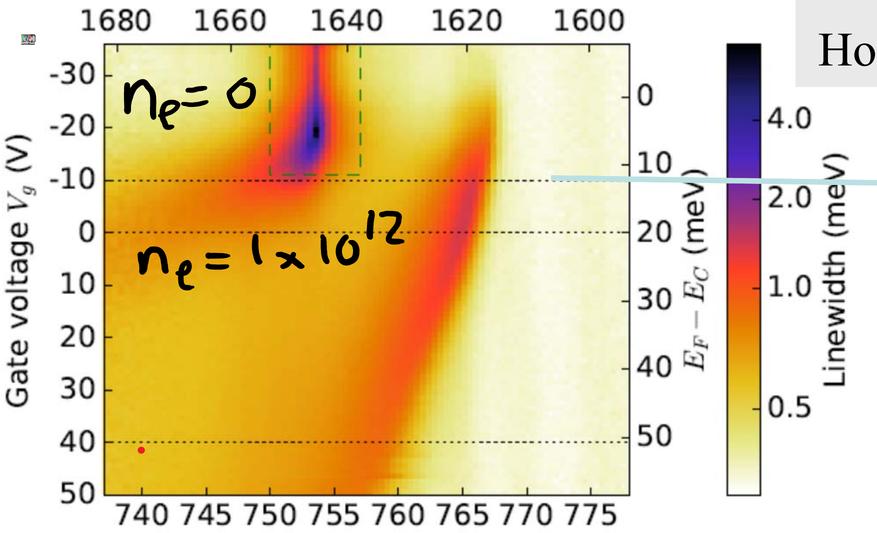
absorption



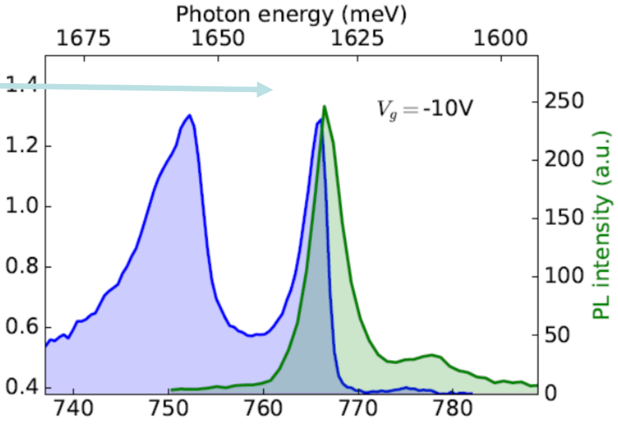
emission

- Sharp increase in conductance indicates free carriers
- Absorption & emission are different for high electron density

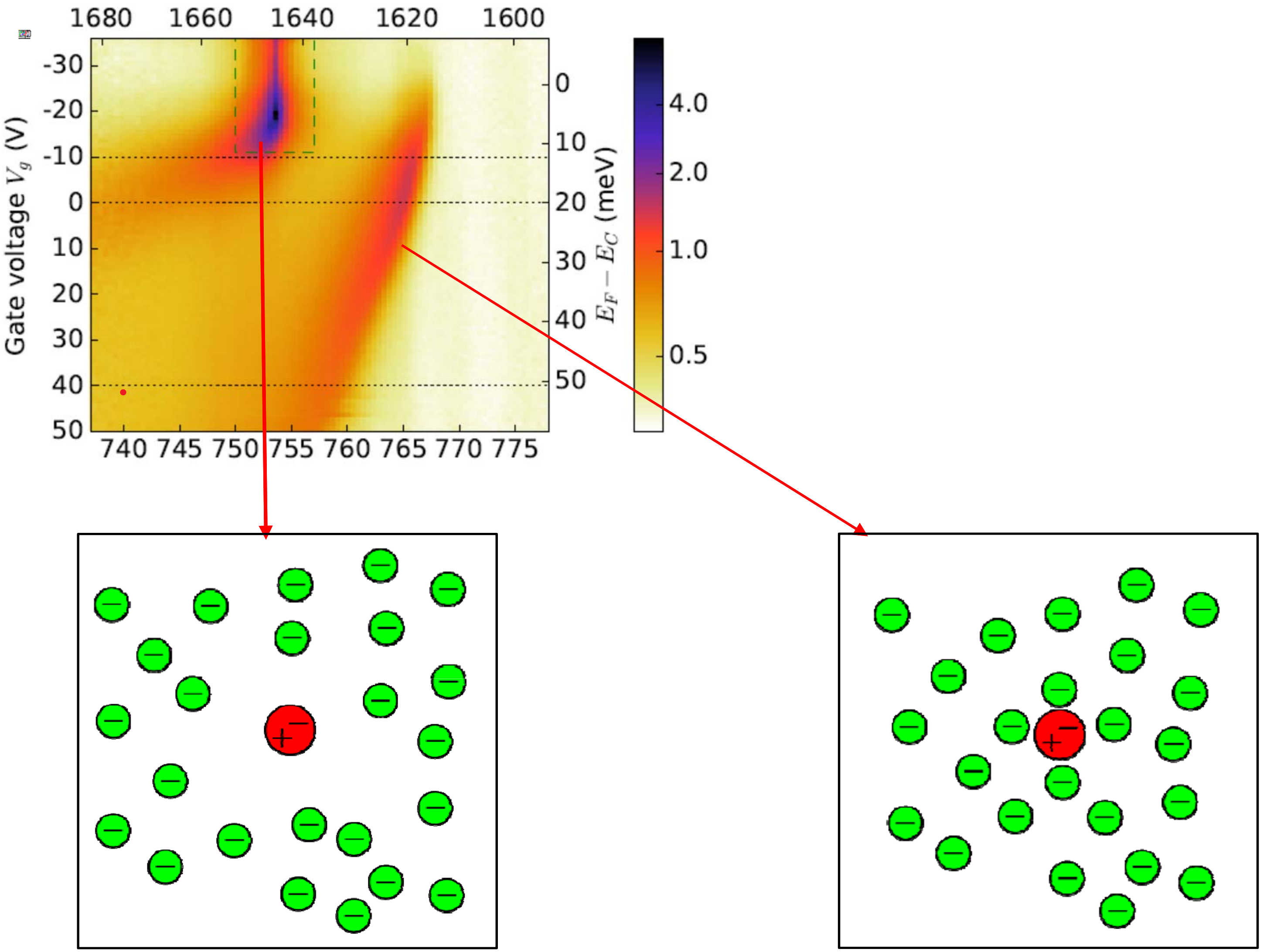
Fermi energy dependence of absorption spectrum



Horitontal line-cut: absorption (blue) + PL (green)



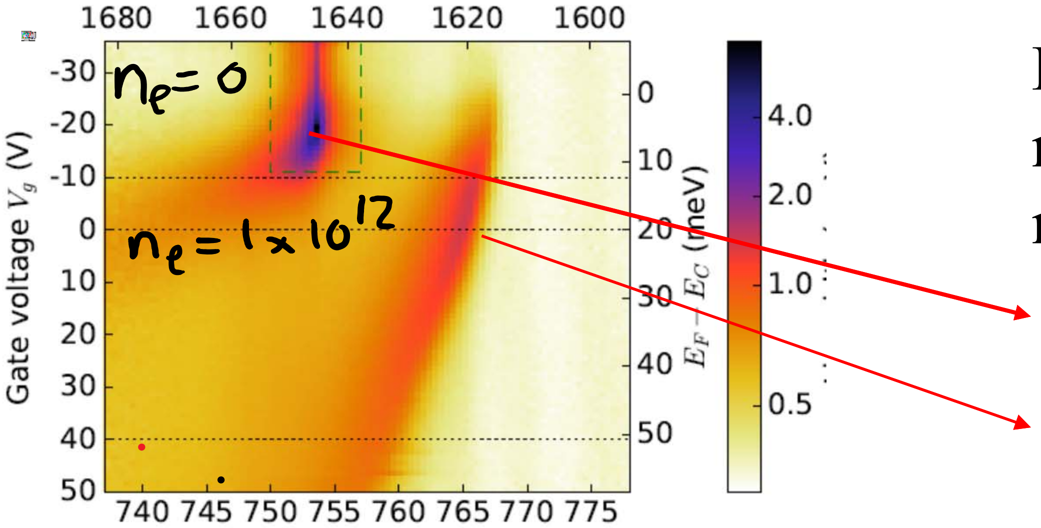
Fermi energy dependence of absorption spectrum



repulsive polaron

attractive polaron

Fermi energy dependence of the spectrum



Differential absorption measurement from a $MoSe_2$ monolayer

Repulsive polaron

Attractive polaron

Due to exciton-electron interactions, elementary optical excitations are exciton-polarons

$$|\Psi\rangle = \alpha\,|\text{bare exciton}\rangle + \beta\,\left|\begin{array}{l}\text{exciton dressed with}\\ \text{electron screening cloud}\end{array}\right\rangle$$

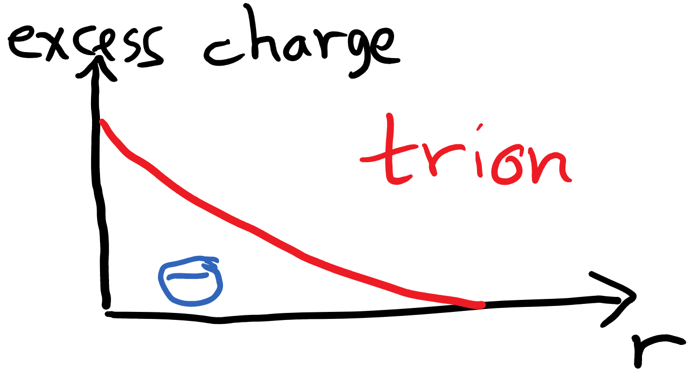
↓ ensures strong light coupling

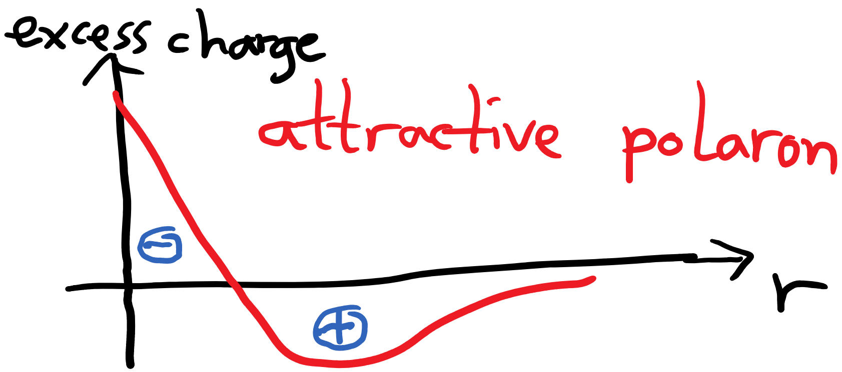
↓ Leads to red (attr. pol.) or blue (rep. pol) shifted resonance

Trion vs. attractive polaron

Trion : H^- like bound state of an exciton and electron

- Assume an exciton at $r = 0$





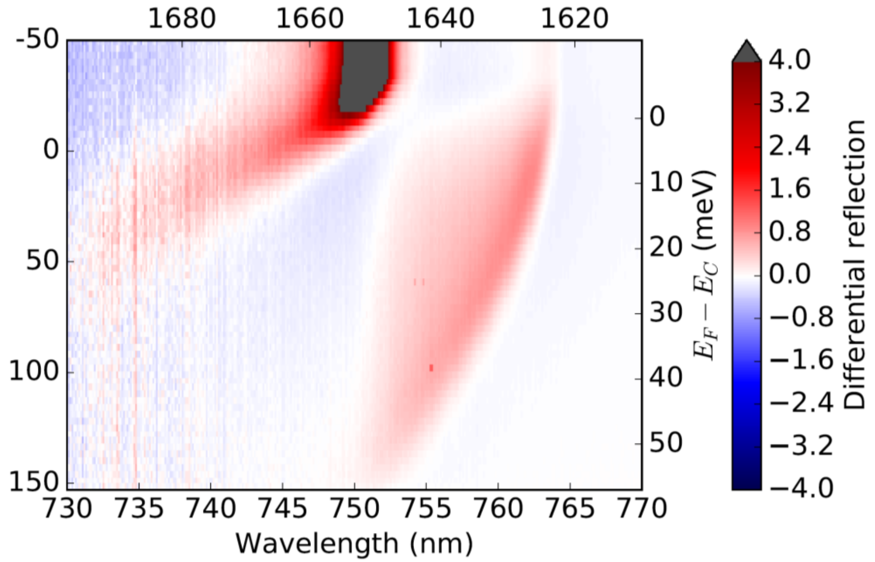
net charge = -e
small oscillator strength

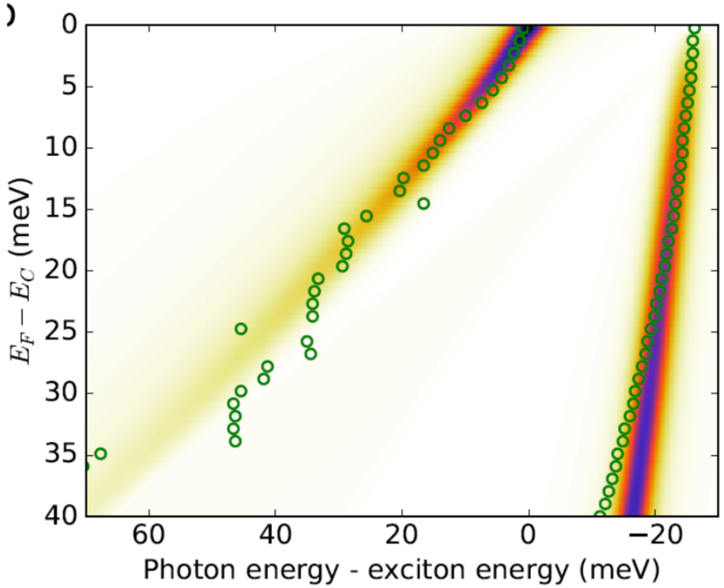
charge neutral !
strong optical coupling

$$\boxed{|\Psi\rangle = \left(x_0^\dagger \phi_0 + \sum_{k>k_F,\, q<k_F} \phi_{kq} c_k^\dagger c_q x_{k-q}^\dagger \right) |0_x\rangle |FS\rangle}$$

Ansatz describes polaron & trion

Chevy Ansatz vs experiments



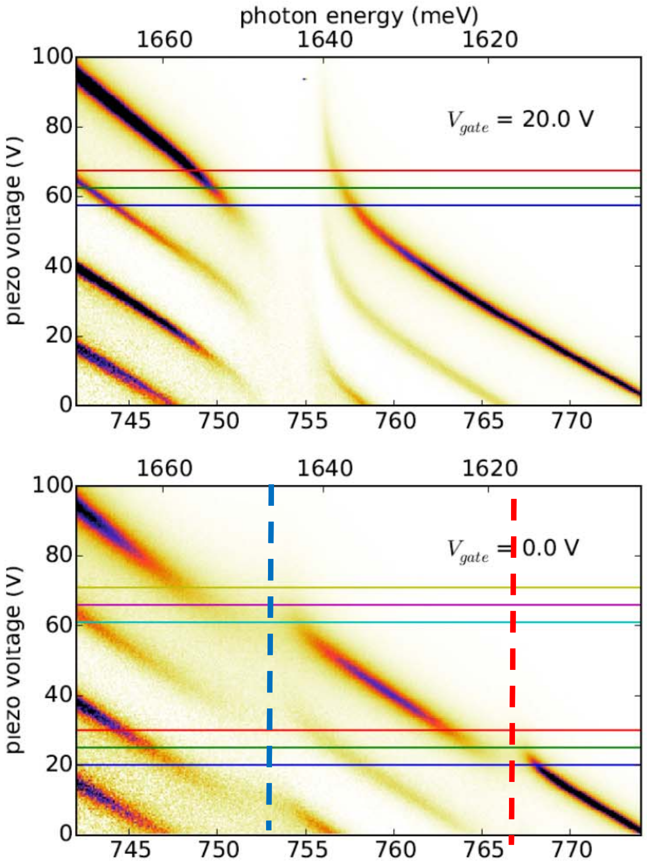


- simple Chevy Ansatz captures the repulsion of the two (polaron) resonances remarkably well (no fit parameters for splitting)

- The overall blue shift of the excitonic resonances due to phase space filling, screening and bandgap renormalization is a fit parameter.

(Sidler et al., Nat. Phys. 2017, Efimkin-MacDonald PRB 2017)

Strong cavity coupling



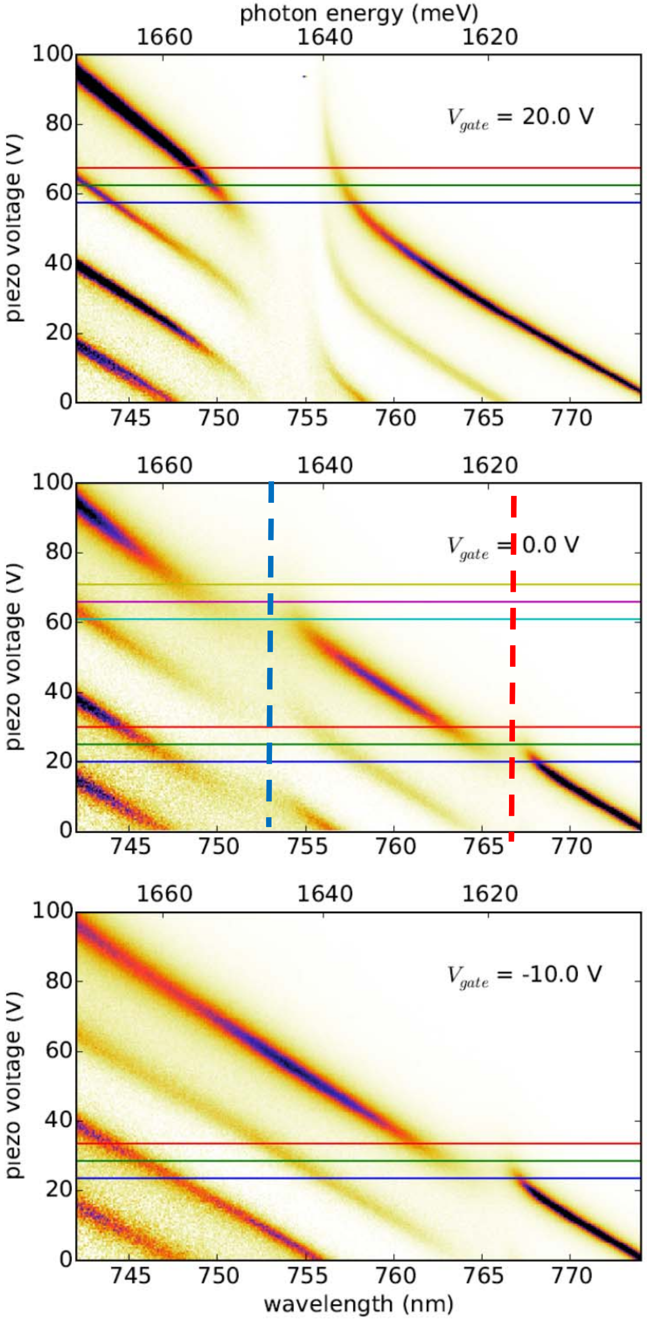
Monolayer is depleted of free electrons: only exciton resonance is visible: $\Omega_R = 18$ meV

exciton-polaritons

Fermi energy $E_F < E_T, \Omega_R$: both attractive & repulsive polarons are observable

exciton-polaron-polaritons

Strong cavity coupling



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Fermi energy $E_F < E_T, \Omega_R$: both attractive & repulsive polarons are observable

Observation of polaron physics with an ultra-light mass (polariton) impurity

$E_F \sim E_T, \Omega_R$: only attractive-polaron polariton is observable: $\Omega_R = 7$ meV

(Sidler et al. Nat. Phys. 2017)

New physics and applications

- Transport of polaritons (dressed with electrons) using external electric & magnetic fields (with F. Pientka, R. Schmidt & E. Demler)
- Polaritons mediating attractive interactions between electrons: light induced superconductivity (V. Ginzburg, W. Little, A. Kavokin)
- Polaritons dressed with anyons in FQHE regime
- Electrons mediating interactions between polaritons: a new method to enhance photon-photon interactions?

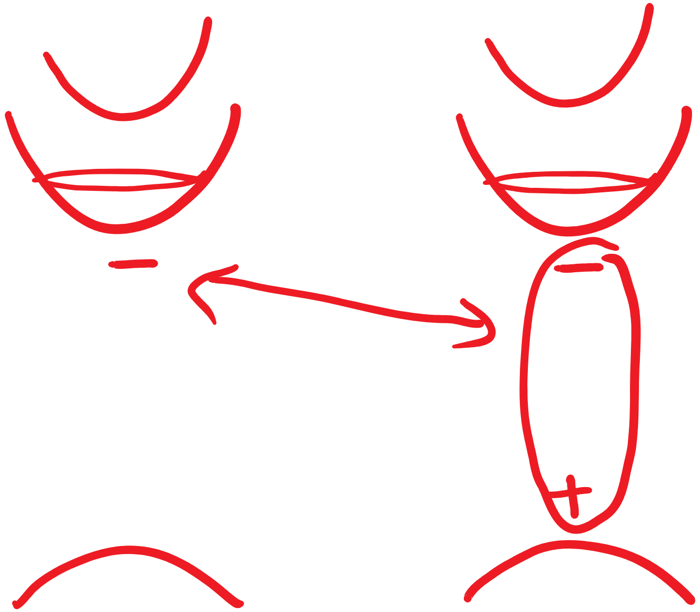
Giant spin susceptibility of TMD monolayers

- In comparison to GaAs, TMD monolayers exhibit:
  - large effective mass (small kinetic energy)
  - reduced screening (large exchange energy gain from spin/valley polarization)

- Itinerant ferromagnetism?

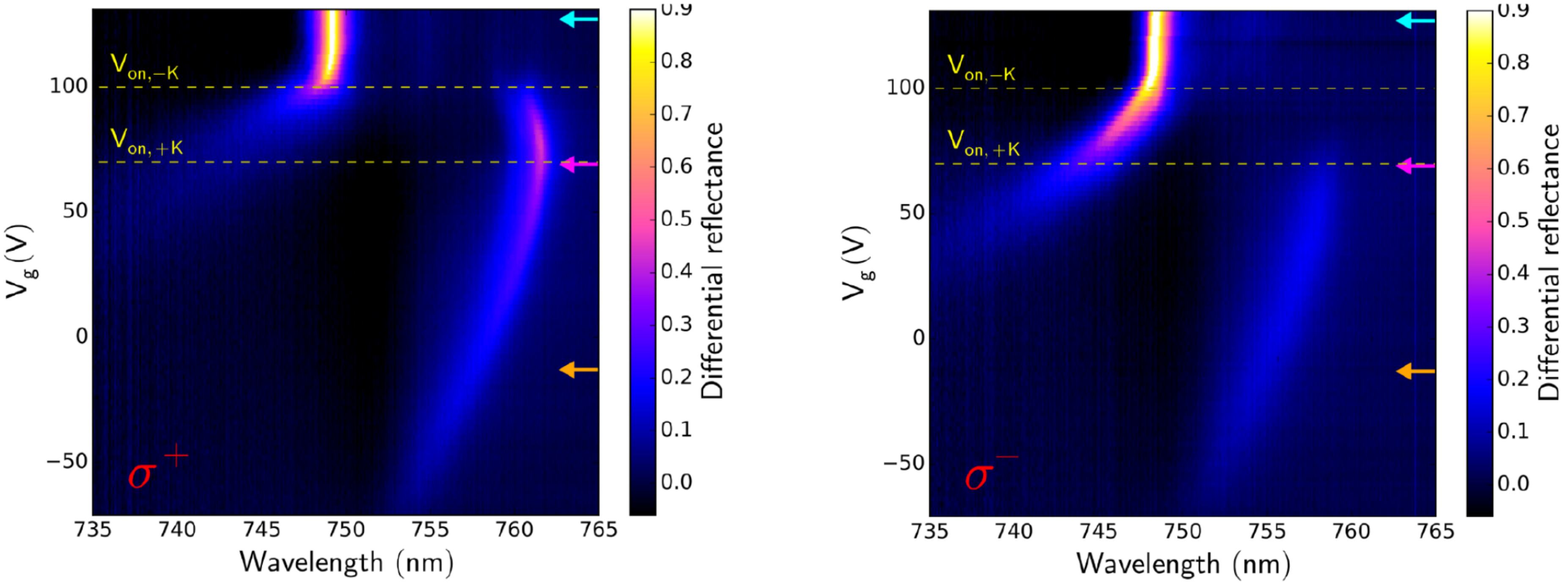
Resonant optical detection of electron valley polarization

- Trion and attractive polaron formation is only possible if the exciton and the electron occupy different valleys ↔ inter-valley trion.
- If electrons are valley polarized, trion formation and polaron absorption/emission is observed only for a single polarization



Intra-valley trion in $MoSe_2$ is triplet – and is not bound

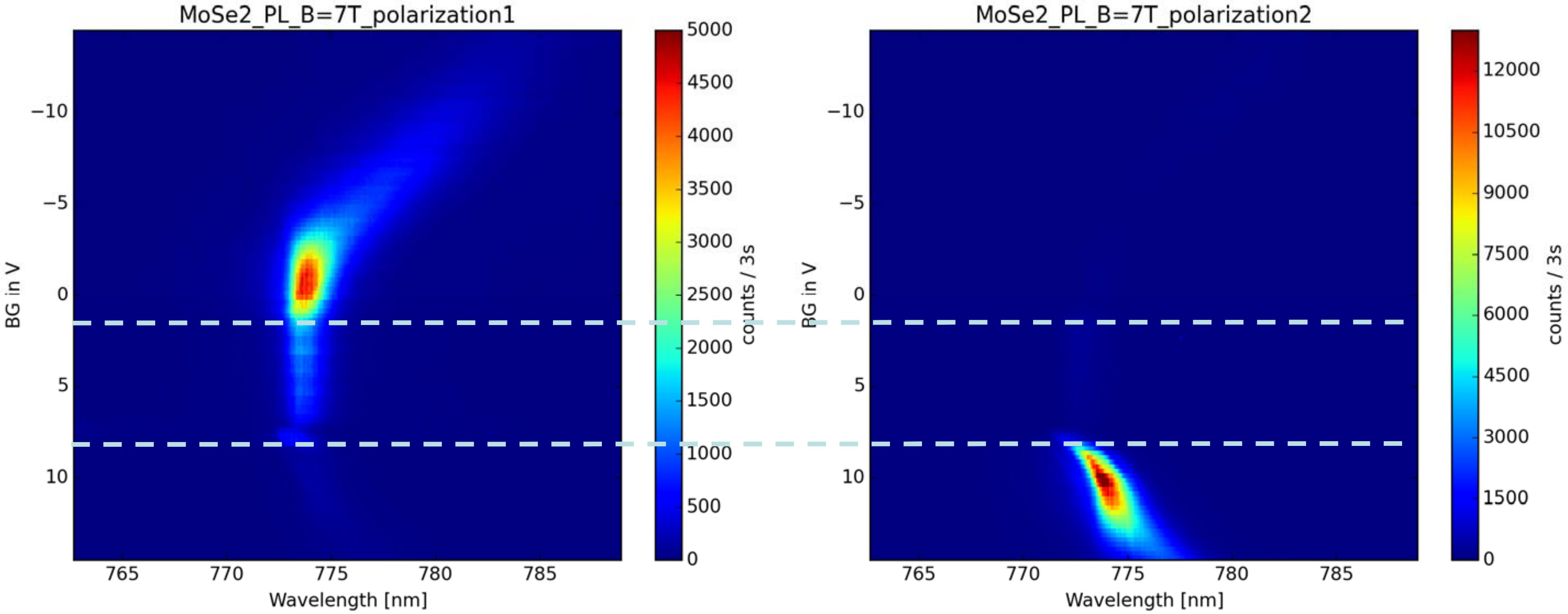
Gate voltage dependent reflection at B=7T



- For 100V > Vgate > 70V, σ+ reflection is dominated by attractive polaron whereas that of σ– by exciton
- The electron density needed to observe σ– attractive polaron line is 1.6×10^{12} cm^{-2}
- In the absence of interactions, this would have required $g_{elec} = 38$!

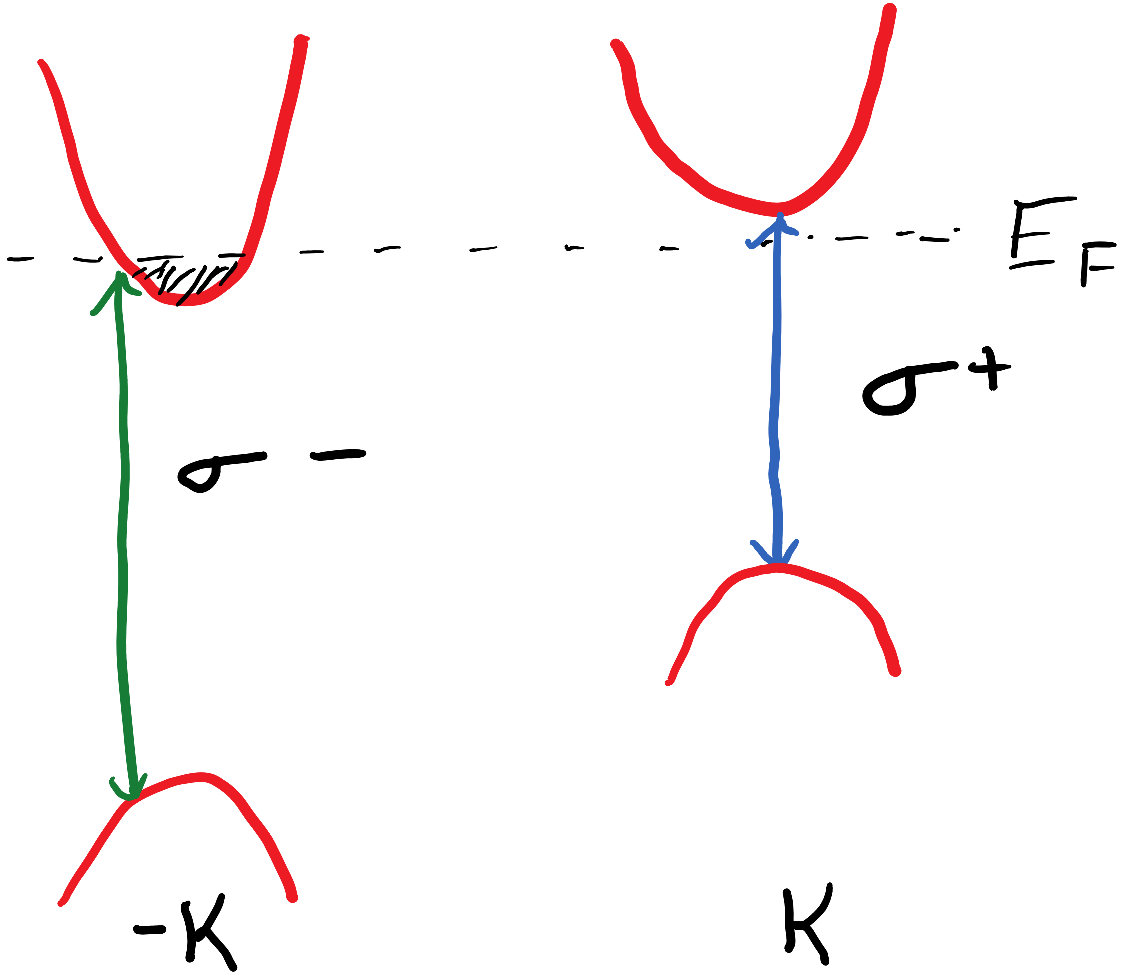
PL: electron and hole doping

σ+ polarized PL σ- polarized PL



- For B= 7T, electrons are valley polarized in –K valley and holes are valley polarized in +K valley
- Hole-polaron PL is a factor of 2.5 stronger than electron-polaron!

How do we understand the B=7 Tesla Photoluminescence spectrum?



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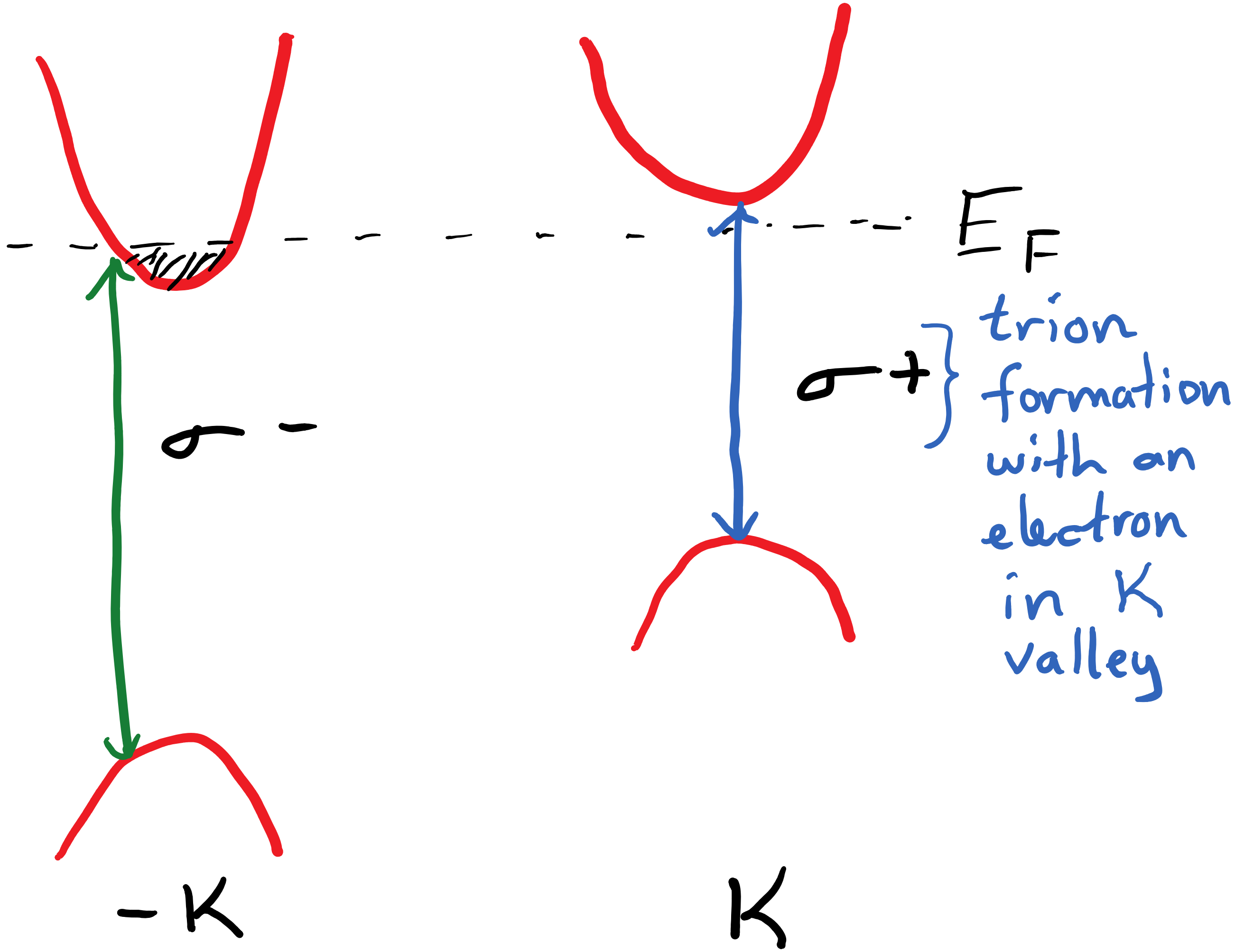

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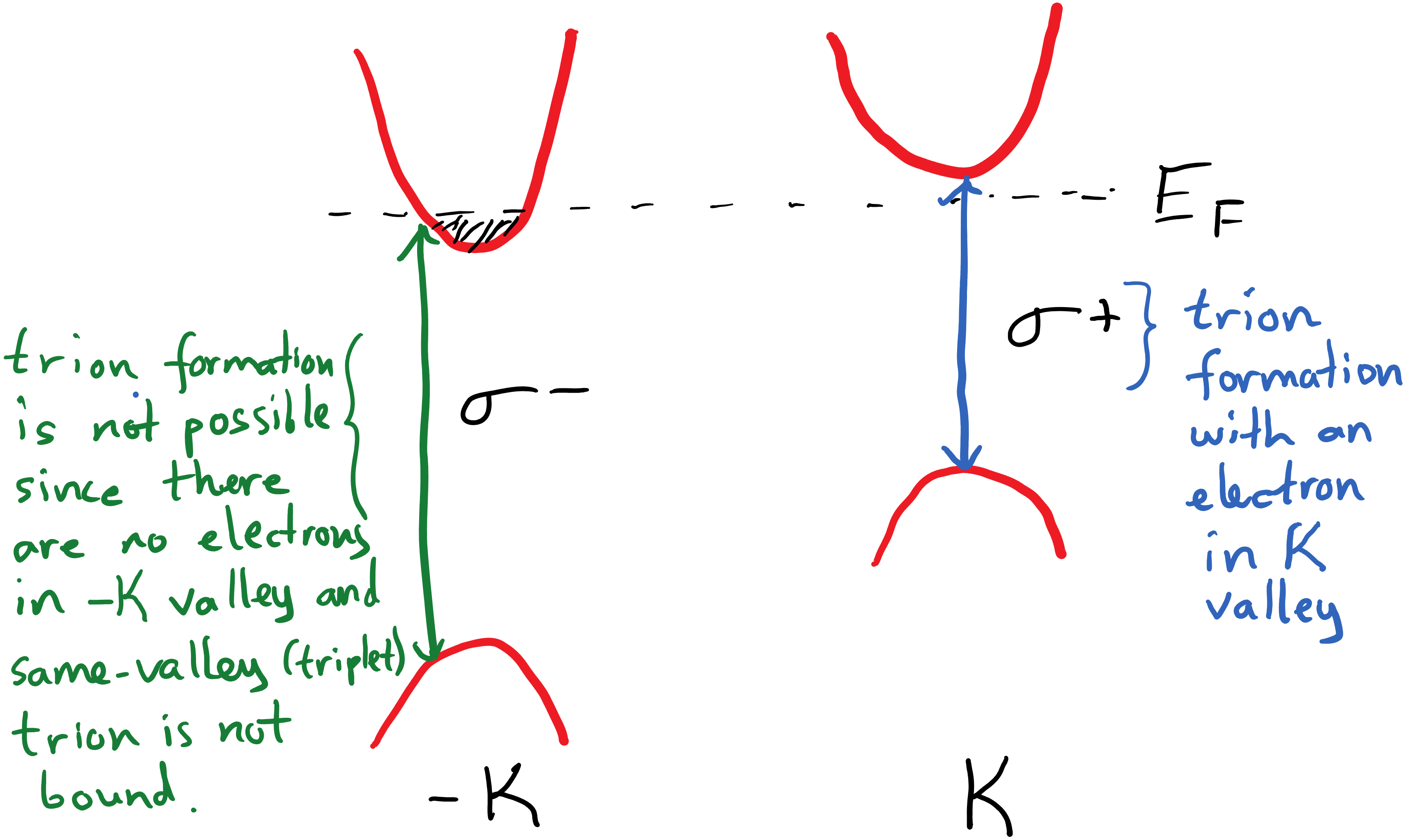

trion formation is not possible since there are no electrons in -K valley and same-valley (triplet) trion is not bound.

trion formation with an electron in K valley

Super-paramagnetic response of MoSe2

- In contrast to GaAs, $MoSe_2$ has a high electron mass and reduced screening: exchange energy gain from valley/spin polarization could exceed kinetic energy cost - towards Stoner instability?
- No magnetization for B= 0T
  - but saturation for B > 5T

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