

Many-body optical excitations in solid-state systems:

a degenerate Bose-Fermi mixture consisting of excitons/polaritons and itinerant electrons

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Optical excitations in the presence of itinerant electrons or holes

- **A generic system with a long history in both pure and applied physics:**
 - Fermi-edge singularity (an optically generated immobile valence hole)
 - Semiconductor lasers or LEDs

Optical excitations in the presence of itinerant electrons or holes

- **A generic system with a long history in both pure and applied physics:**
 - Fermi-edge singularity (an optically generated immobile valence hole)
 - Semiconductor lasers or LEDs
- **The new angle provided by the new two-dimensional (2D) materials:**
 - Elementary optical excitations are strongly bound excitons which can be treated as rigid, mobile impurities in a Fermi sea of electrons or holes
 - Easy control of degeneracy of both electrons (through electrical gates) and excitons (through resonant lasers) – Bose-Fermi mixtures
 - Strictly nonequilibrium dynamics due to short exciton lifetime

Motivation for investigating degenerate electron-exciton system

- **Control of electron-electron interactions:**
 - Polariton mediated superconductivity in a 2D semiconductor
 - Optical control of electronic phase transitions in steady-state
- **Enhancement of exciton-exciton interactions:**
 - Photon blockade and strongly interacting photonic systems
 - Manipulation of photonic excitations using electric or magnetic fields

Outline

- Introduction
- Elementary optical excitations of transition metal dichalcogenide (TMD) monolayers
- Dipolar excitons in TMD monolayers
- Optical gain by stimulated polaron-electron scattering

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

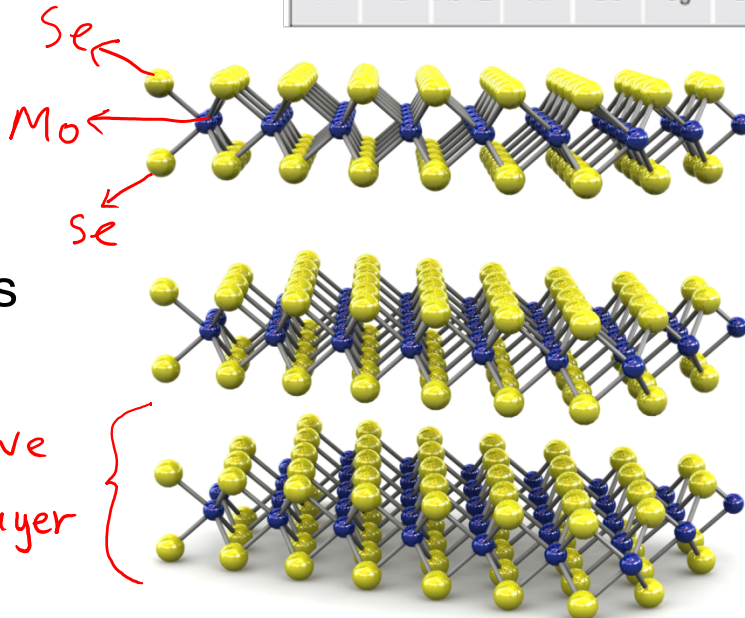
Formula: MX_2

M = Transition Metal
X = Chalcogen

H																	He
Li	Be	MX_2 M = Transition metal X = Chalcogen										B	C	N	O	F	Ne
Na	Mg	3	4	5	6	7	8	9	10	11	12	Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac-Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo

Layered
materials

effective
monolayer



**Electrical
property**

Semiconducting

Semimetallic

Ferromagnetic

Metallic, CDW,
Superconducting

Material

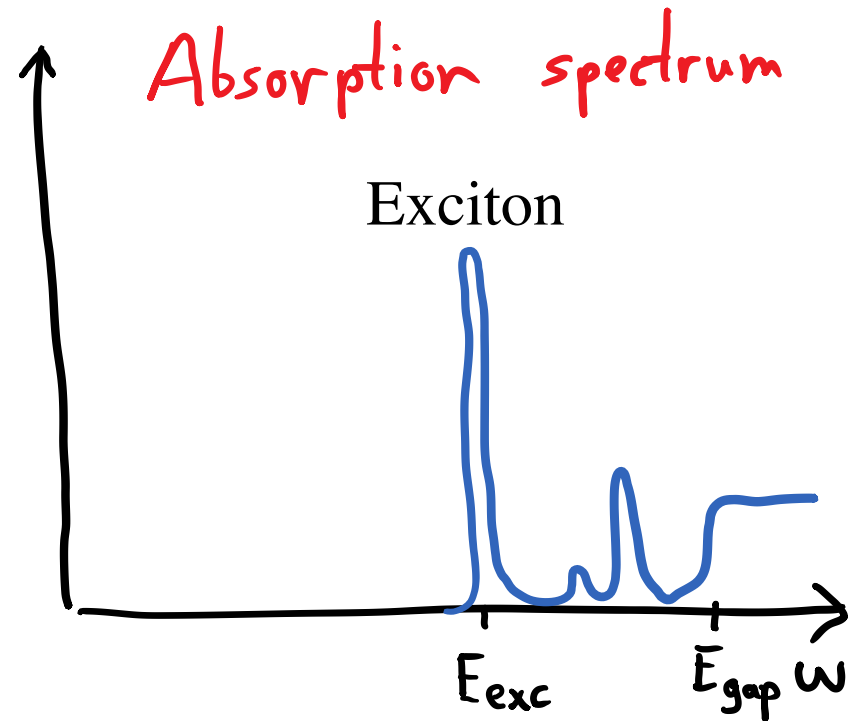
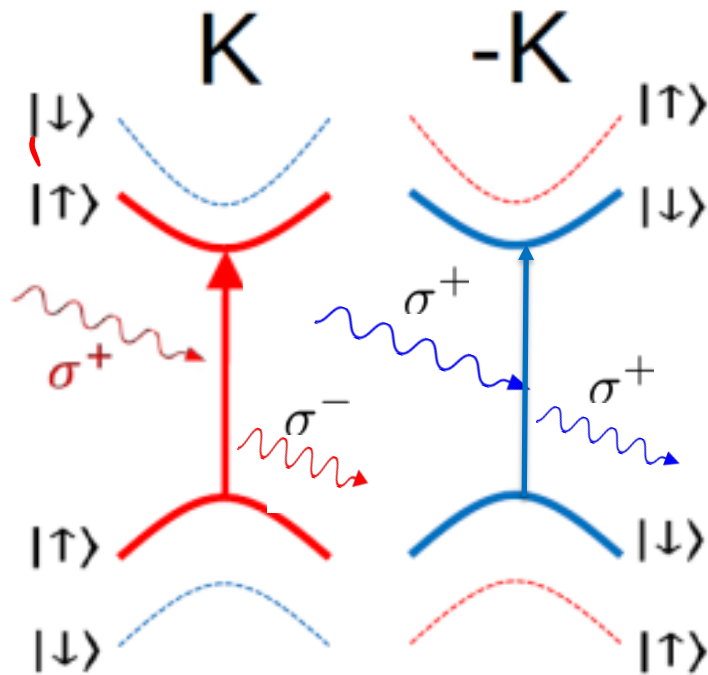
MoS_2 MoSe_2 WS_2
 WSe_2 MoTe_2 WTe_2

TiS_2 TiSe_2

CrI_3 , CrBr_3

NbSe_2 NbS_2 NbTe_2
 TaS_2 TaSe_2 TaTe_2

TMD monolayers: semiconductors with optically addressable valley degree of freedom



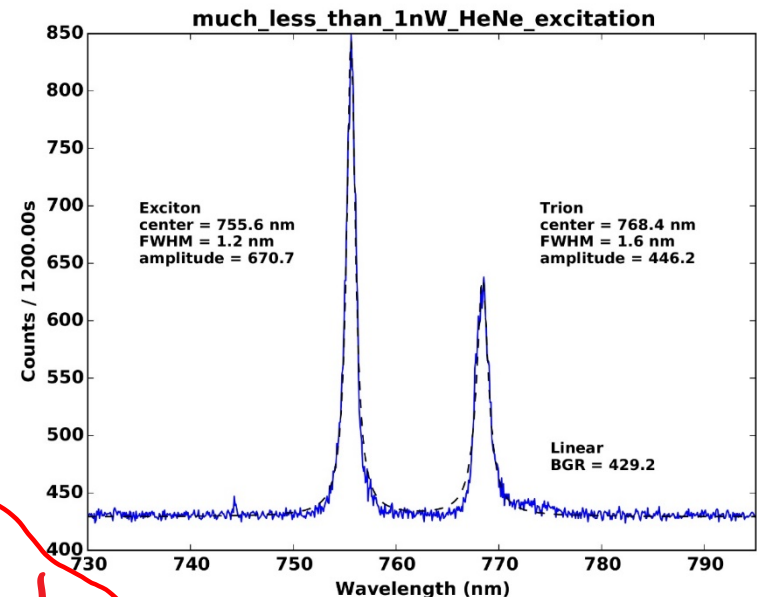
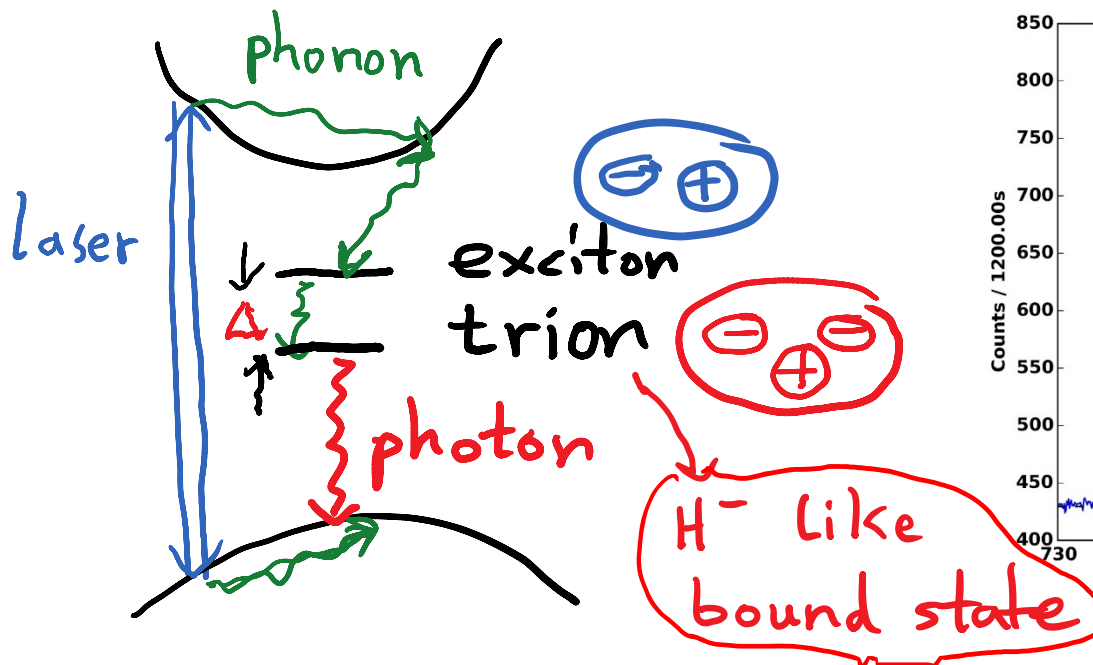
Reduced dielectric screening leads to ultrastrong exciton binding energy

Pioneering work: Heinz, Xu

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 low density localized electrons (trion oscillator strength is much smaller).

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

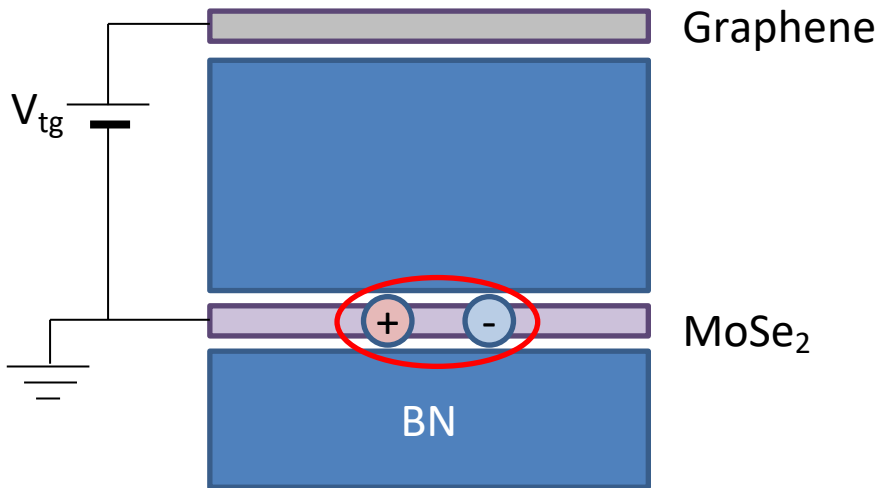


Implications of strong exciton binding $\equiv$ small Bohr radius a_B

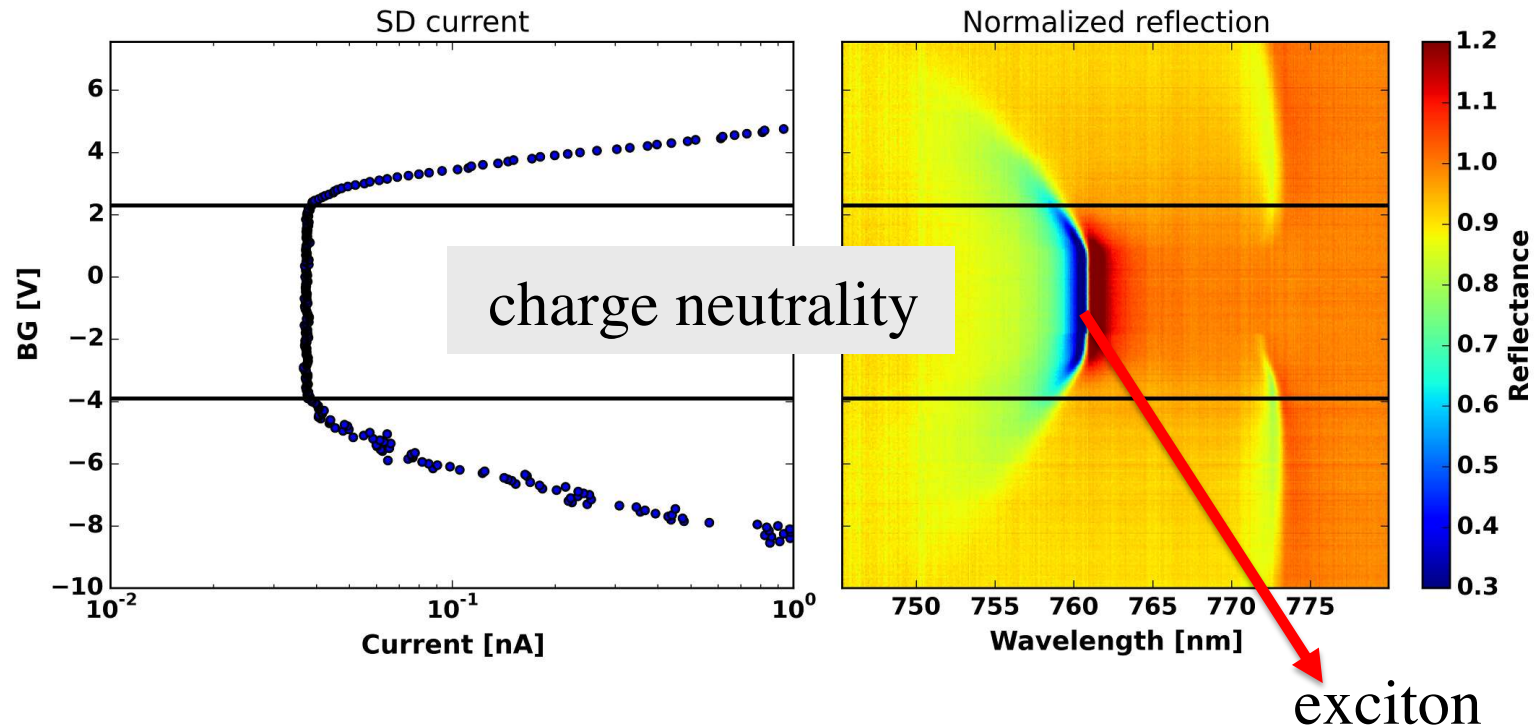
- TMD excitons couple very strongly to resonant photons:
 - ultrafast /sub-ps radiative decay rate ($\sim 1/a_B^2$) $\Gamma_{\text{rad}} \sim 1 \text{ meV}$
 - strong reversible coupling to cavities ($\sim 1/a_B$) $g \sim 10\text{-}30 \text{ meV}$
- State-of-the art TMD monolayers are insensitive to disorder and have nearly radiative decay limited exciton linewidths
- Optical response of charge-neutral TMDs is predominantly linear;
 - monolayers are near-perfect mirrors ($R > 80\%$ observed)
 - difficult to realize polariton lasers, polariton blockade

Charge tunable van der Waals heterostructures

- Exfoliation of and stacking of monolayers of semiconducting TMDs and graphene, together with ~ 10 nm thick insulating boron nitride (BN) layers
- A gate voltage applied between the top (transparent) graphene gate and the MoSe₂ layer allows for tuning the electron/hole density



Carrier density dependent reflection



- Sharp increase in conductance indicates free carriers
- Reflection is strongly modified as electrons or holes injected
- A new red-shifted resonance if electrons/holes are present

(prior work in GaAs: Rapaport, Bloch,...)

Interacting exciton-electron system

$$H = \sum_k \epsilon_k c_k^\dagger c_k + \sum_k \omega_k x_k^\dagger x_k + \sum_q V_q c_{k+q}^\dagger c_k x_{k'-q}^\dagger x_{k'}$$

rigid exciton
assumption

$$\epsilon_k = \frac{\hbar^2 k^2}{2m_e}$$

$$\omega_k = \frac{\hbar^2 k^2}{2m_x} + e_F$$

$$\frac{1}{V_q} = \frac{1}{V} = - \sum_k \frac{1}{e_B + \hbar^2 k^2 / (2\mu)}$$

↓
molecule (trion)
binding energy

Interacting exciton-electron system

$$H = \sum_k \epsilon_k c_k^\dagger c_k + \sum_k \omega_k x_k^\dagger x_k + \sum_q V_q c_{k+q}^\dagger c_k x_{k'-q}^\dagger x_{k'}$$

$$\epsilon_k = \frac{\hbar^2 k^2}{2m_e} \quad \omega_k = \frac{\hbar^2 k^2}{2m_x} + e_F \quad \frac{1}{V_q} = \frac{1}{V} = - \sum_k \frac{1}{e_B + \hbar^2 k^2 / (2\mu)}$$

Approximate eigenstates of H in the truncated Hilbert space:

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

Polaron Chevy-Ansatz

(prior work: Suris)

exciton

Exciton + Fermi-sea electron-hole pair

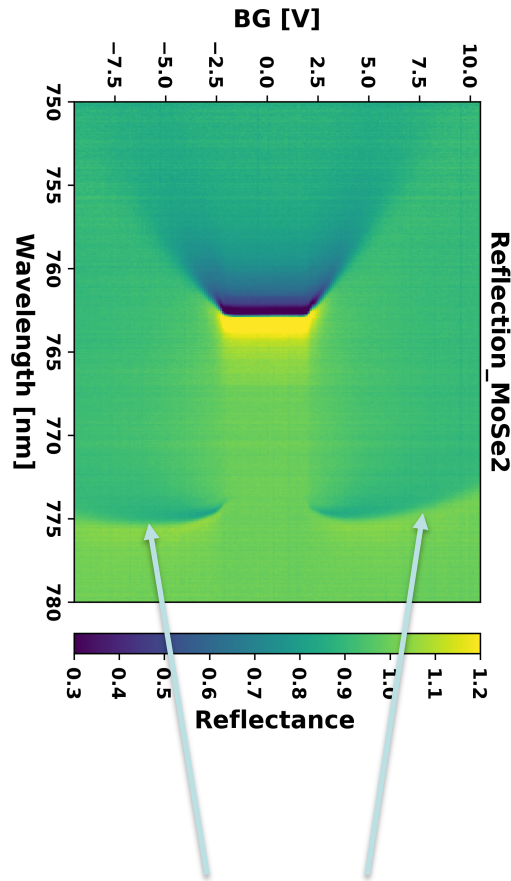
quasi
particle :
weight

$$\phi_0 \sim k_F a_T$$

ensures that polaron has
finite oscillator strength.

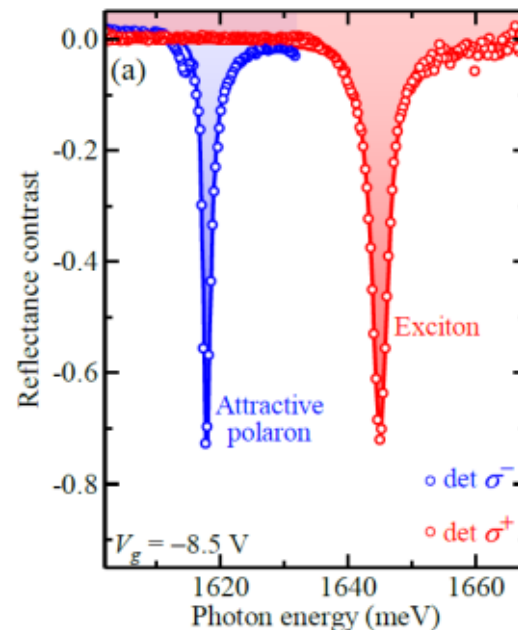
a_T : trion
Bohr rad.

Fermi energy dependence of the spectrum



Attractive polaron branch

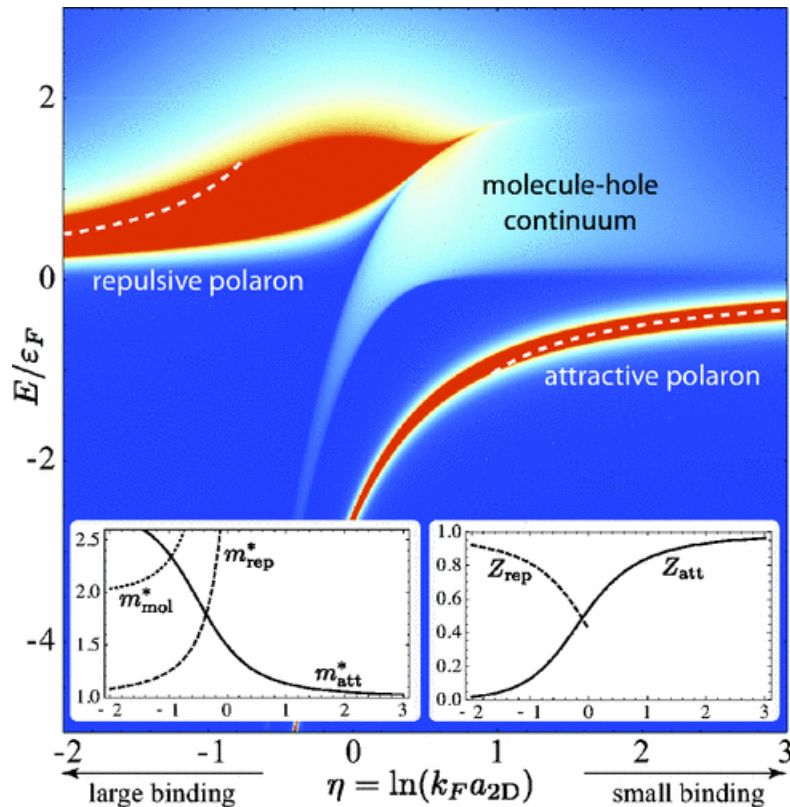
- Existence of a bound trion state is crucial for polaron formation
- For vanishing electron density, attractive polaron & trion are indistinguishable
- Excitons in K-valley can only be dressed by electrons in K'-valley



A comparison of Fermi-polaron physics in cold atom and 2D semiconductor systems

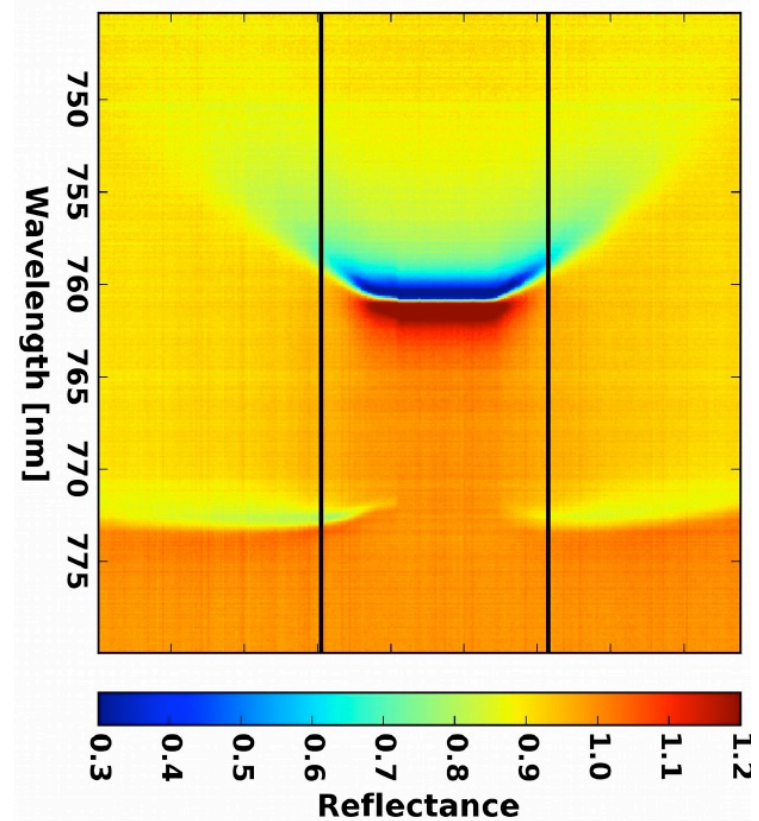
Cold atoms

vary scattering length



2D Semiconductors

vary electron density

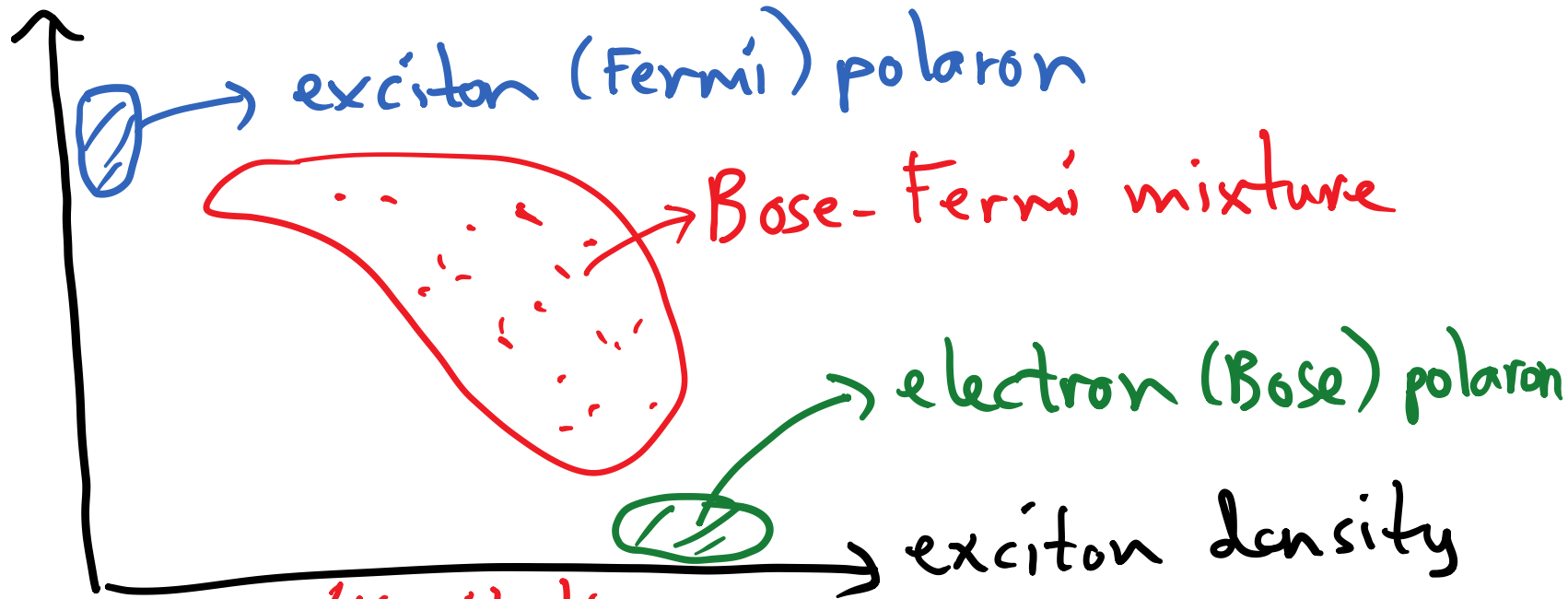


Impurity created by: RF pulse (atoms)

resonant laser (TMDs)

From Fermi to Bose polarons

electron density



Features/Limitations:

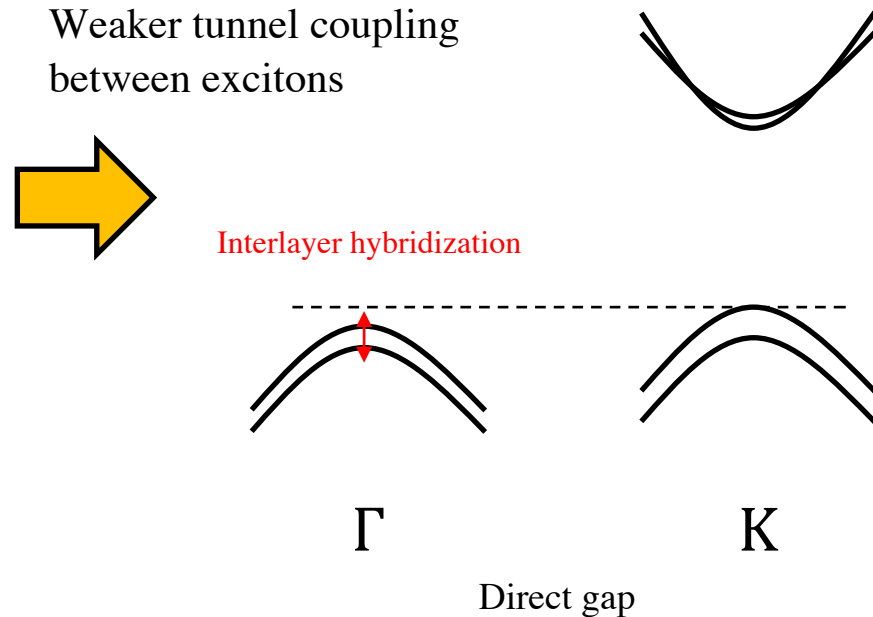
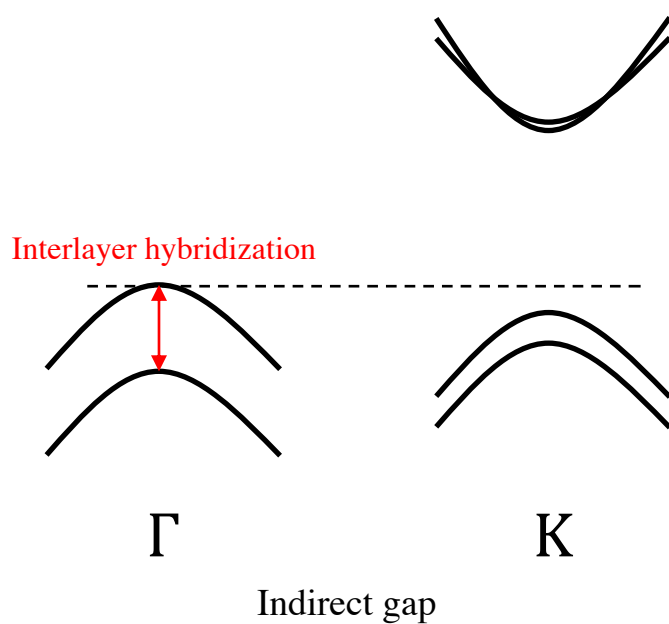
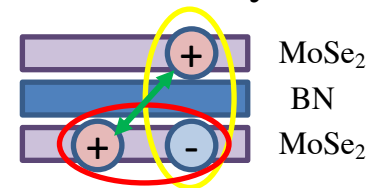
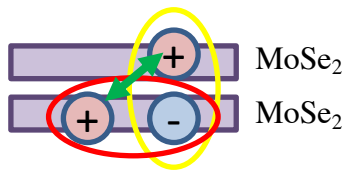
- $E_F \gg kT$ - but exciton decay rate is large
- Rigid exciton assumption
- electron-electron interactions

A new tool for controlling electron-exciton interactions:

Dipolar excitons in Bilayer MoSe₂ + Monolayer BN

Homobilayer TMDs are indirect bandgap, leading to inferior optical properties

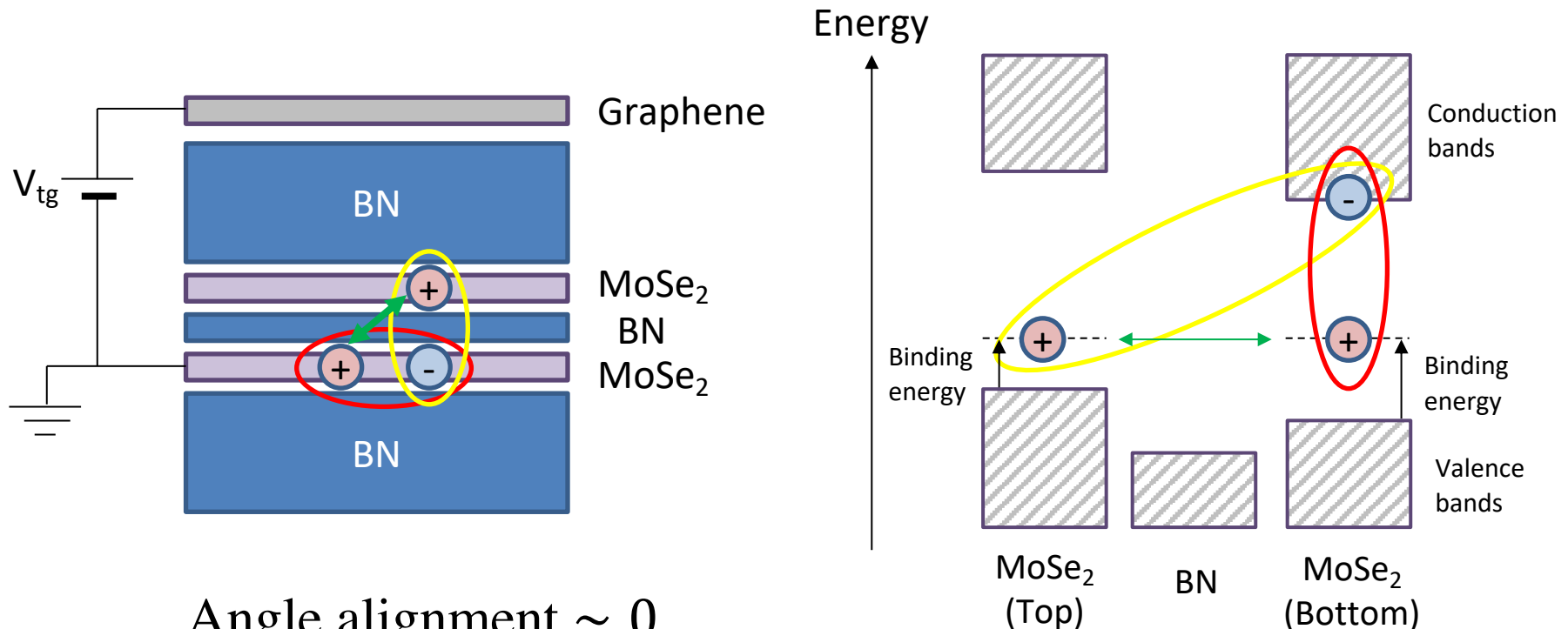
Tunnel coupling in Γ valley leading to an indirect bandgap can be suppressed by a BN layer inbetween MoSe₂ layers



Enhanced dipole moment

Gate controlled bilayer MoSe₂

- Hybridization of intra- and inter-layer excitons at $V_g > 0$ through electron tunneling and for $V_g < 0$ through hole tunneling



Angle alignment ~ 0

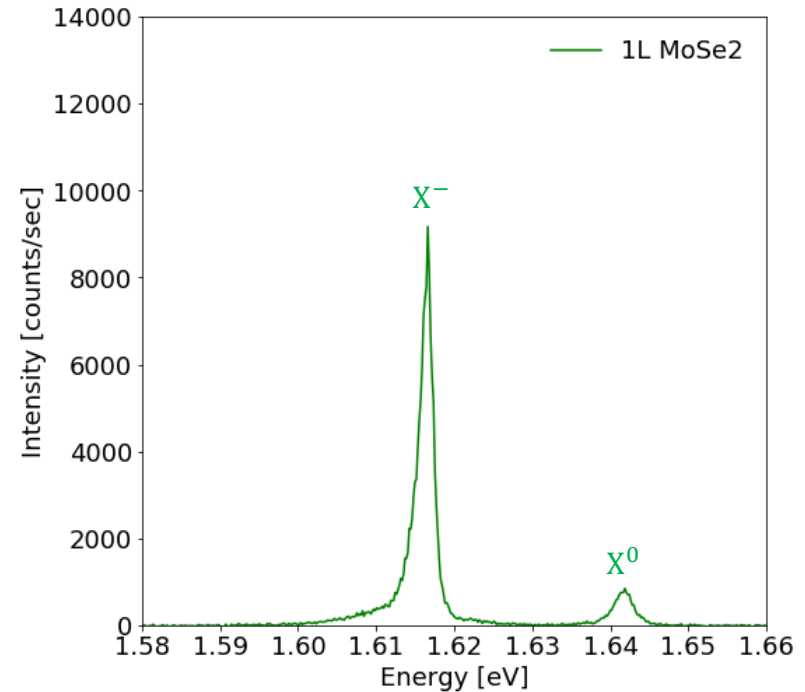
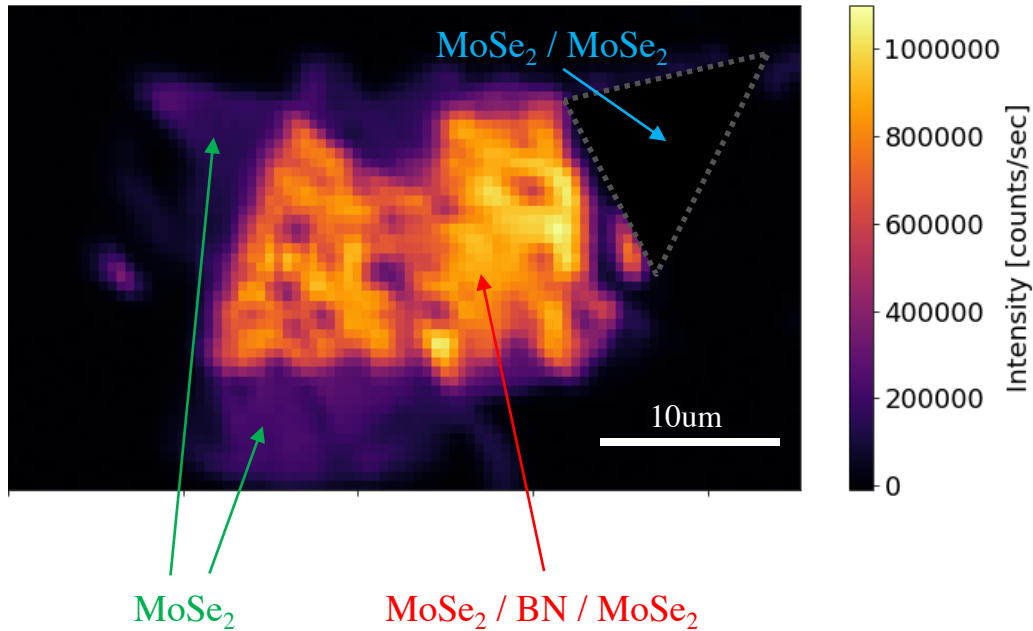
Tear and stack technique

(also used for twisted bilayer graphene)

Spatial map of photoluminescence

Spatial map of total PL

(PL integrated over 1.58 – 1.66 eV)



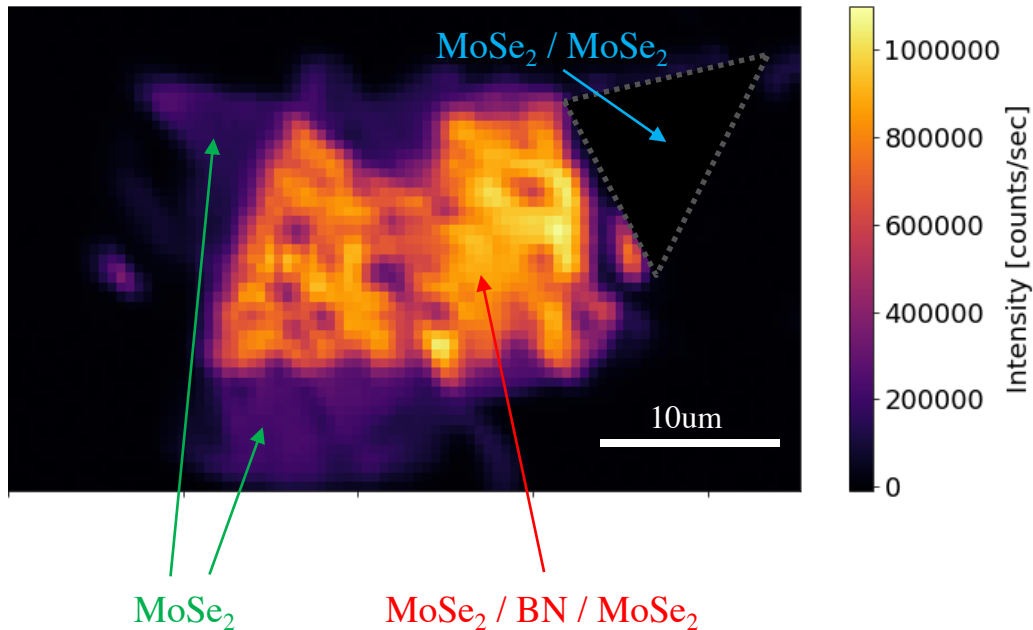
T = 4.2K

633nm HeNe laser excitation (400nW)

Spatial map of photoluminescence

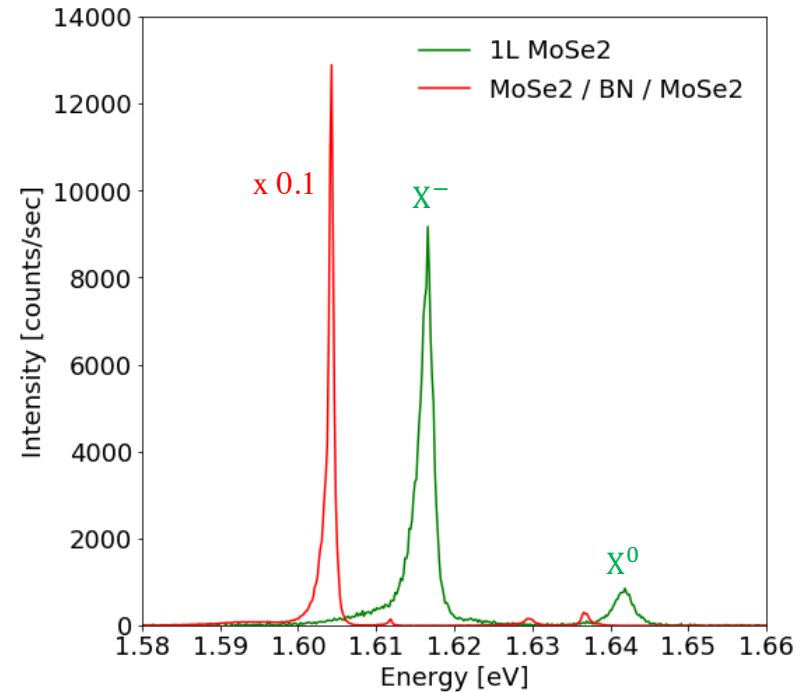
Spatial map of total PL

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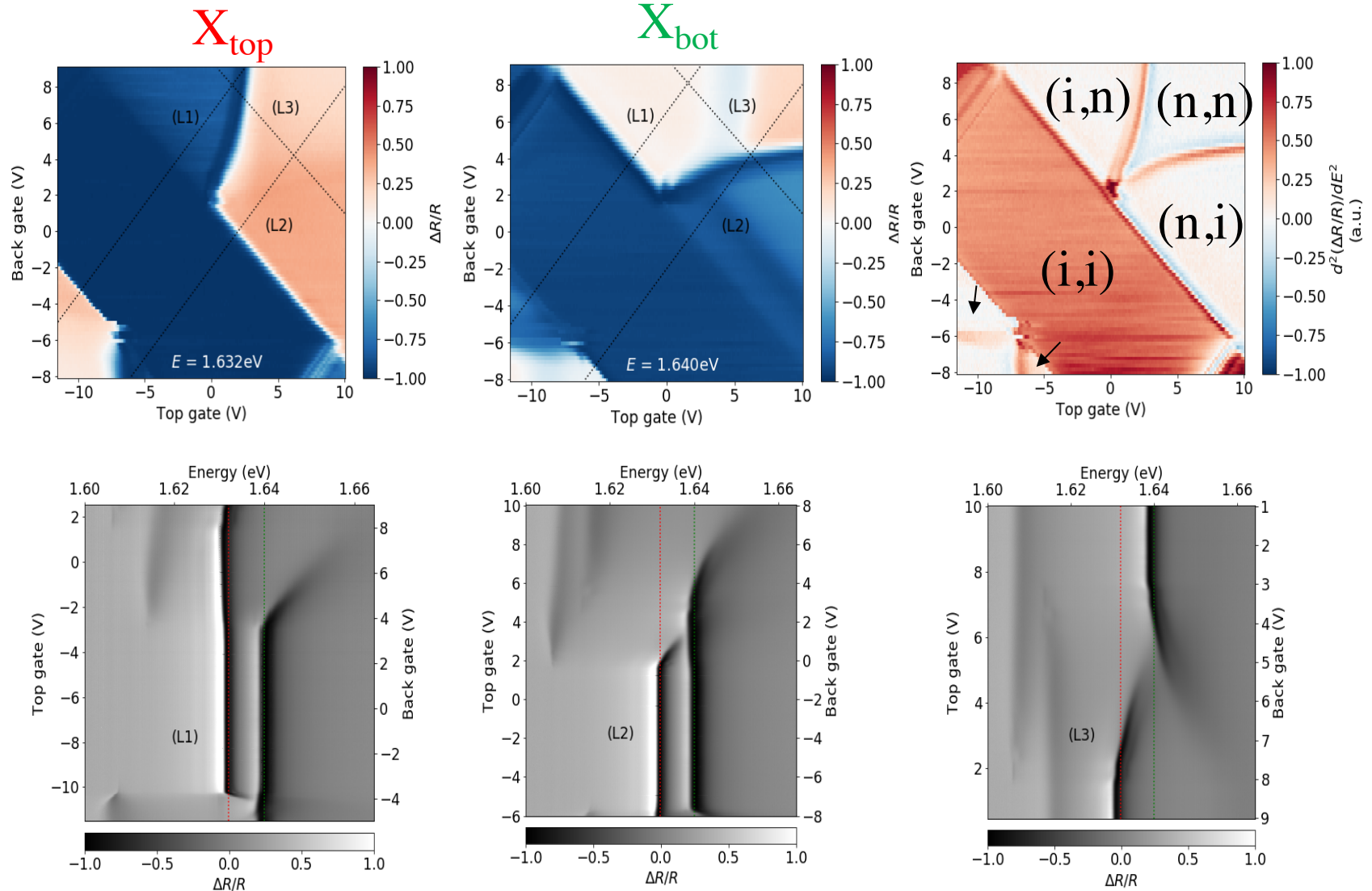
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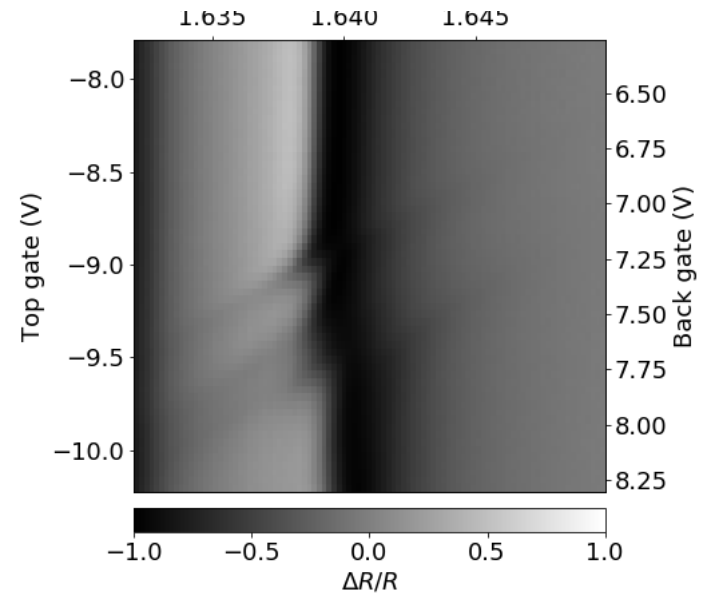
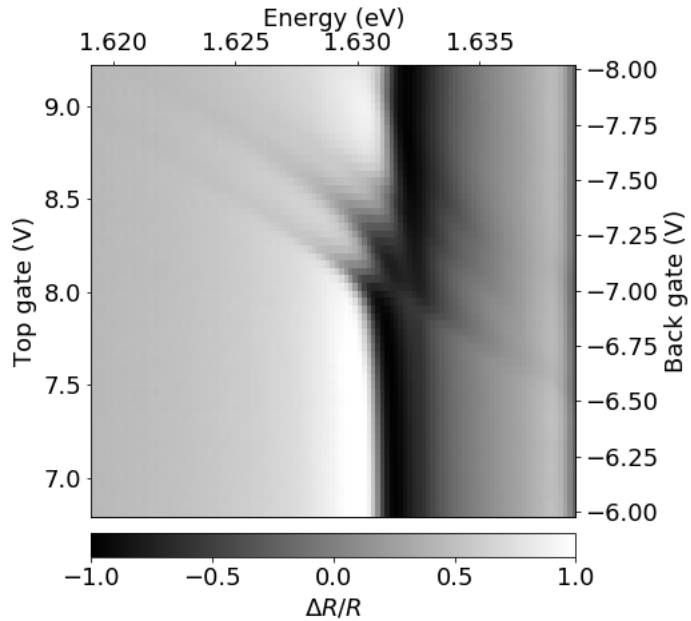
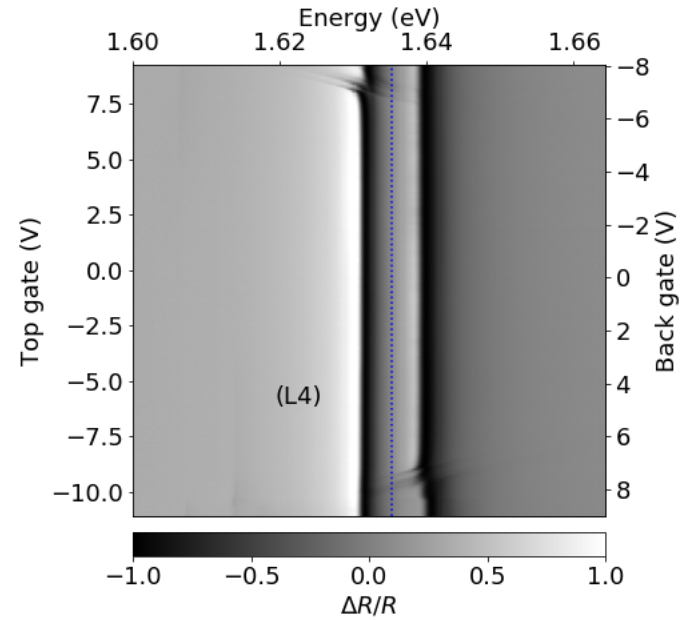
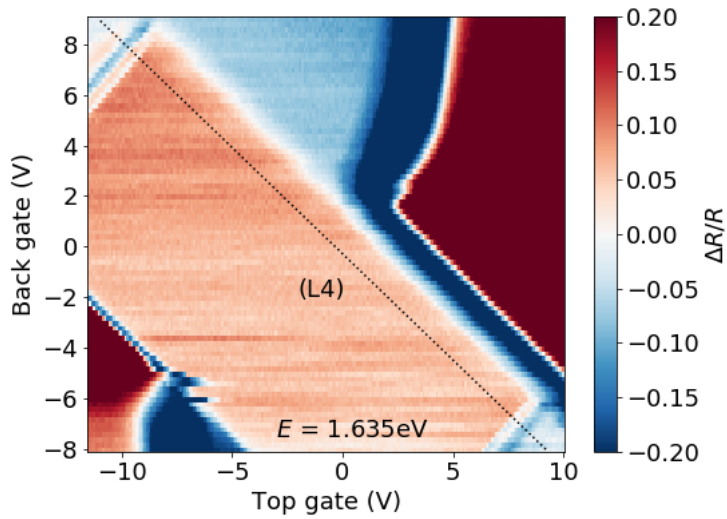


Gate dependence of differential reflectance

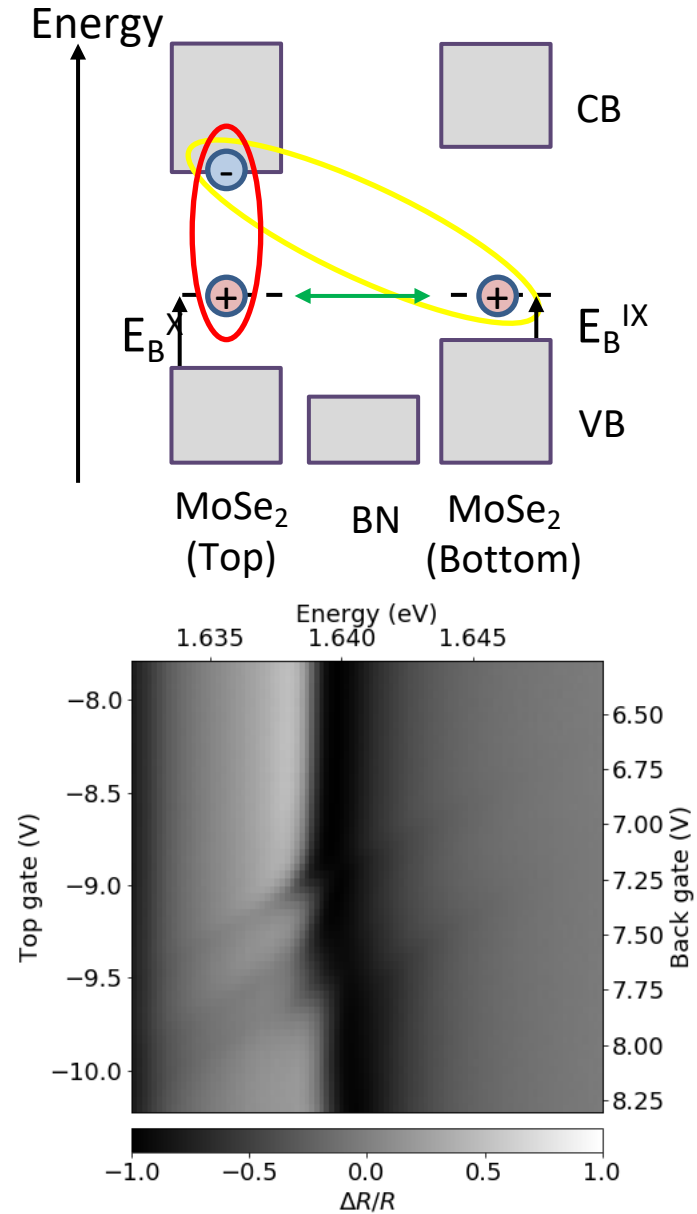
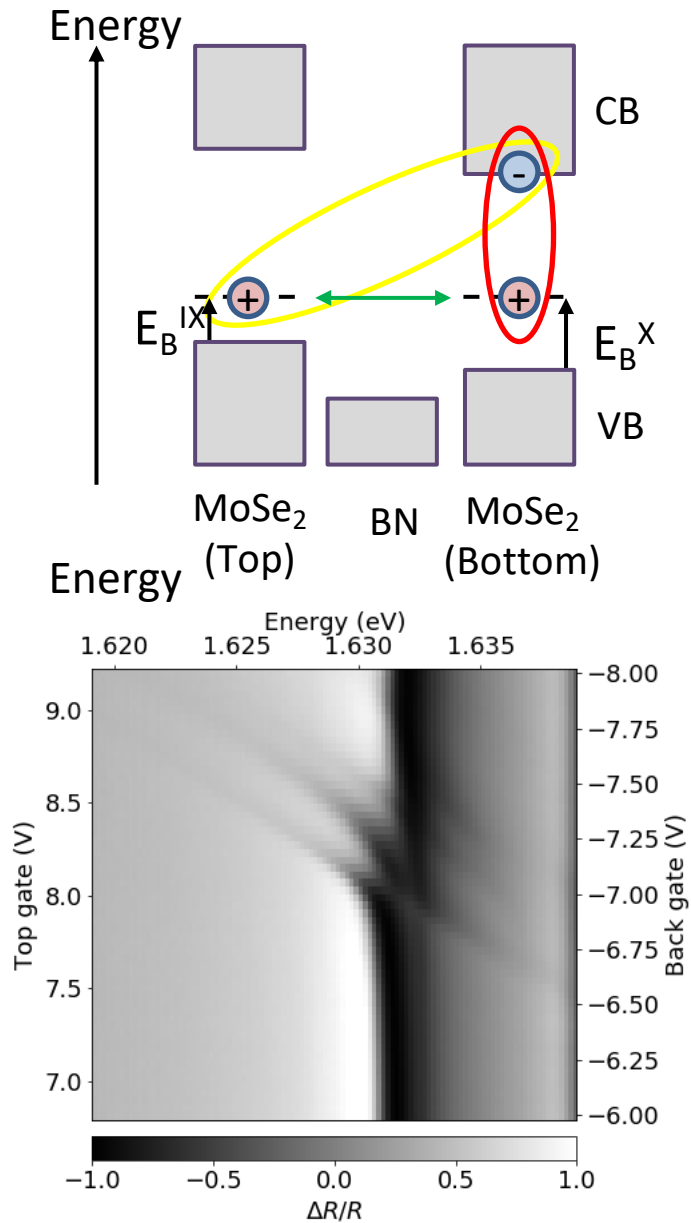
- Two layers have slightly different neutral exciton energies
- Optical reflection map of charging plateaus



Dipolar excitons

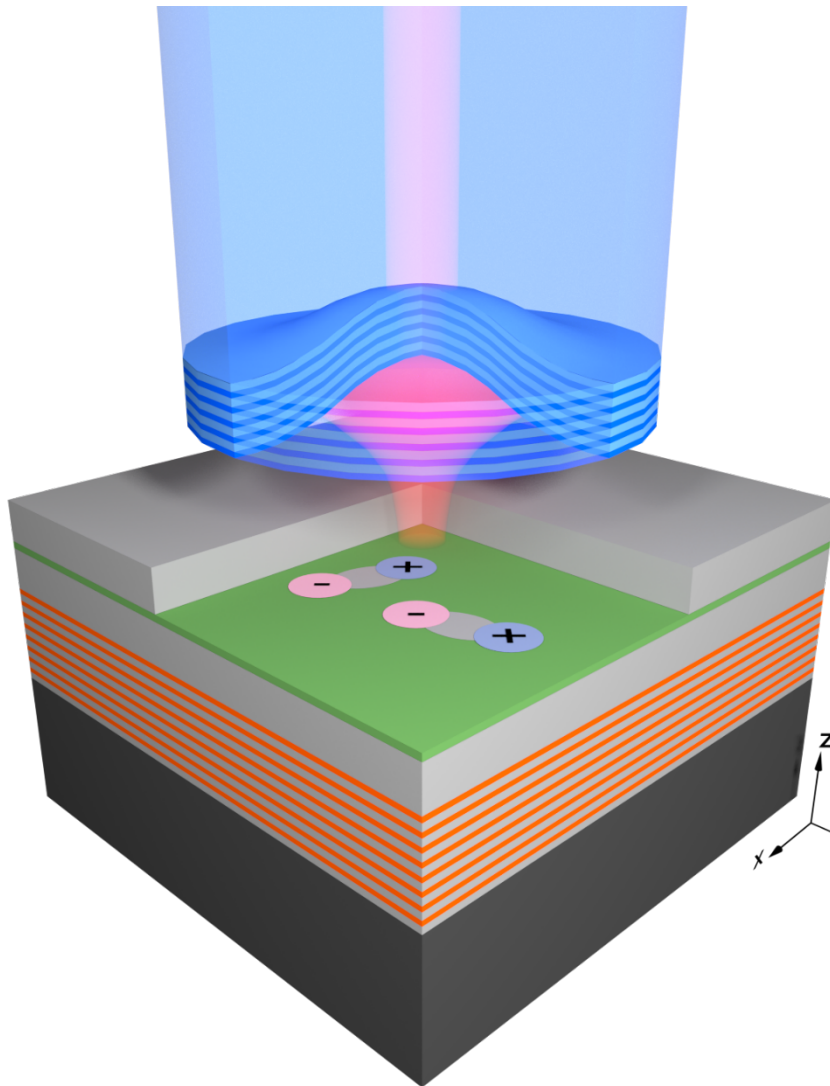


Dipolar excitons

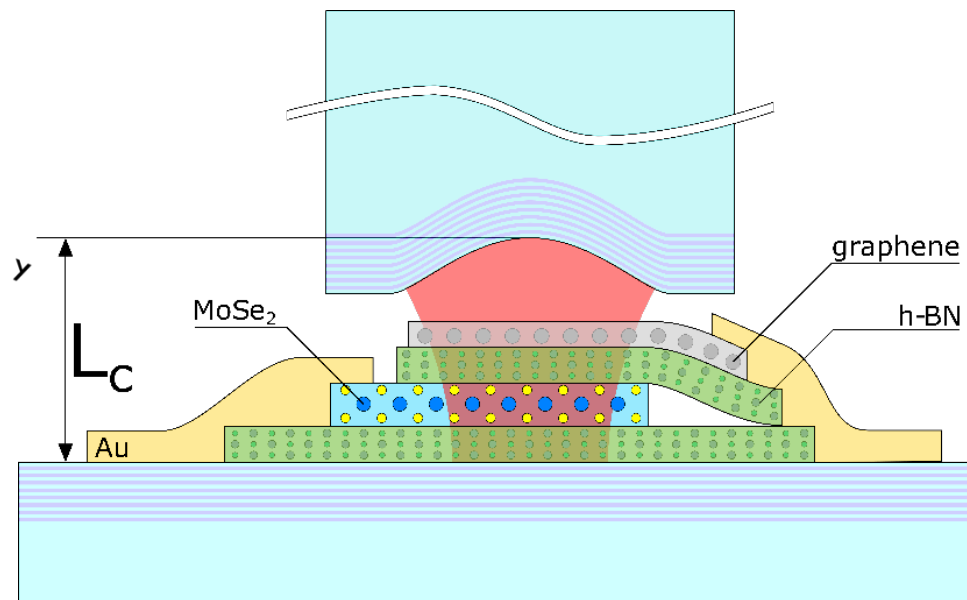


- Clear anti-crossing – but 3 indirect exciton lines!

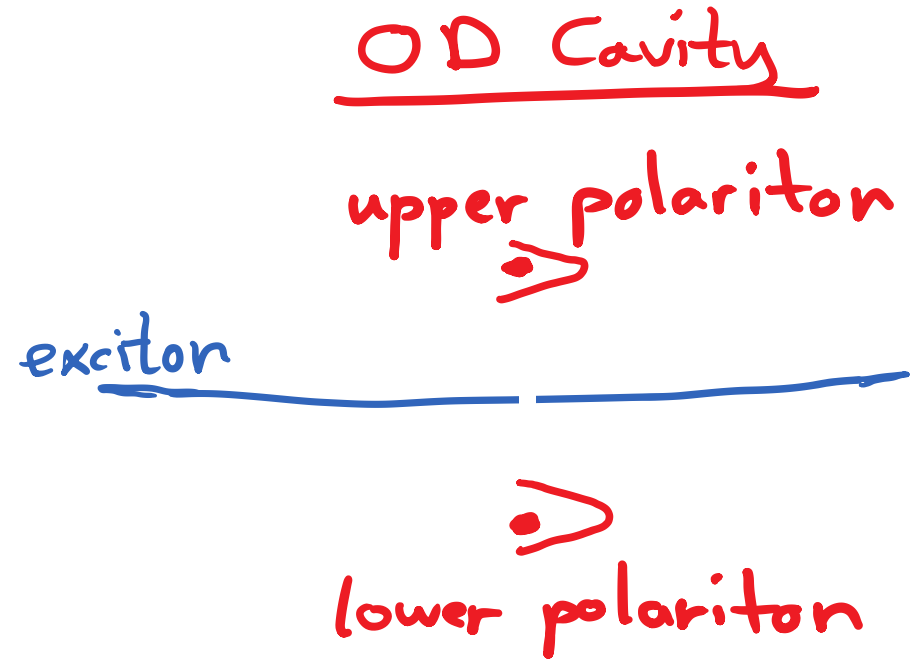
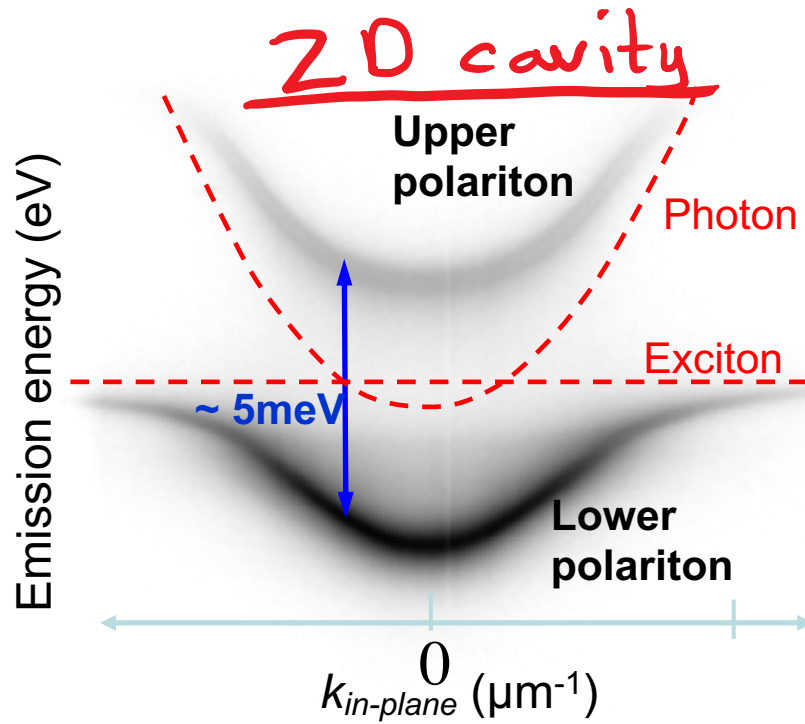
Cavity-polaritons with 2D materials



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

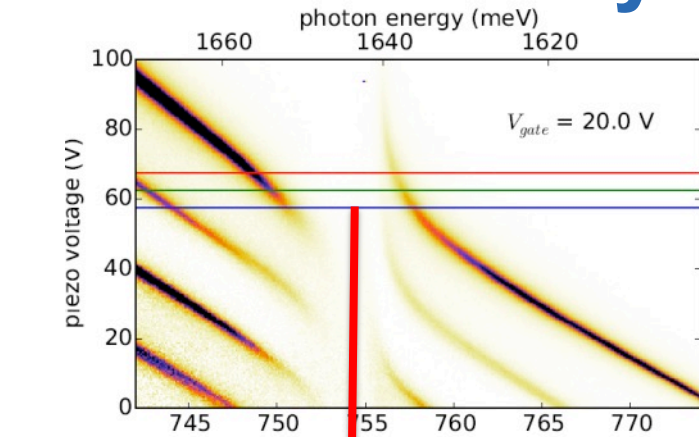


Cavity-polaritons with 2D materials

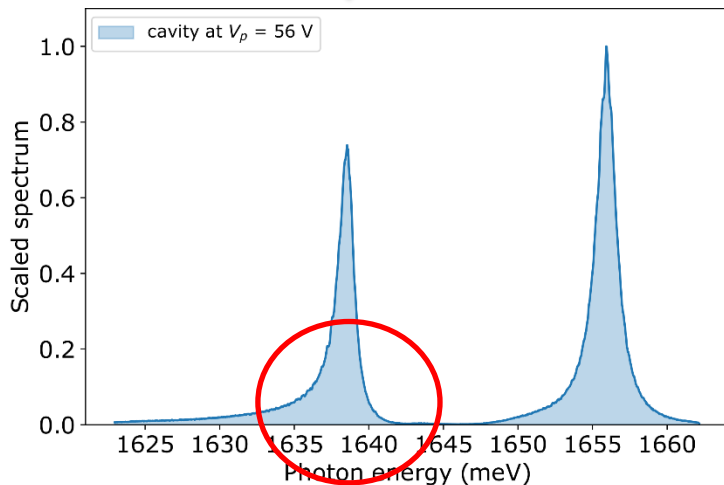


$$|\Psi_{\text{polariton}}\rangle = \alpha |\text{exciton}\rangle + \beta |\text{cavity photon}\rangle$$

TMD in a cavity: strong coupling regime



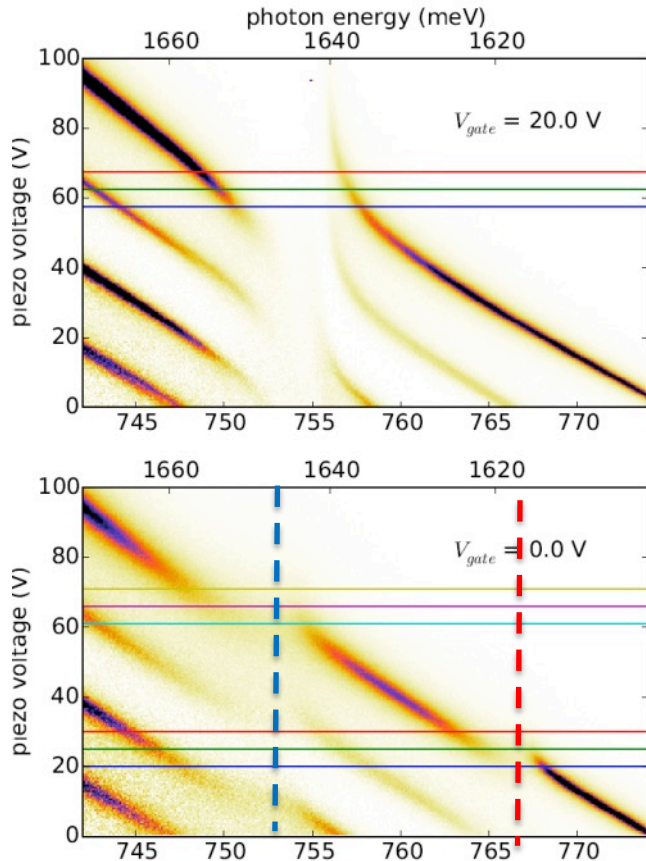
- Large normal mode splitting:
 $\Omega_R = 17 \text{ meV}$ — **elementary excitations:**
exciton-polaritons
- Maximum reported splitting $> 40 \text{ meV}$
- Lower polariton exhibits asymmetric lineshape due to quantum interference: EIT-analog



Earlier results:

Menon, Tartakovskii

Exciton-polaritons in the presence of a 2DES



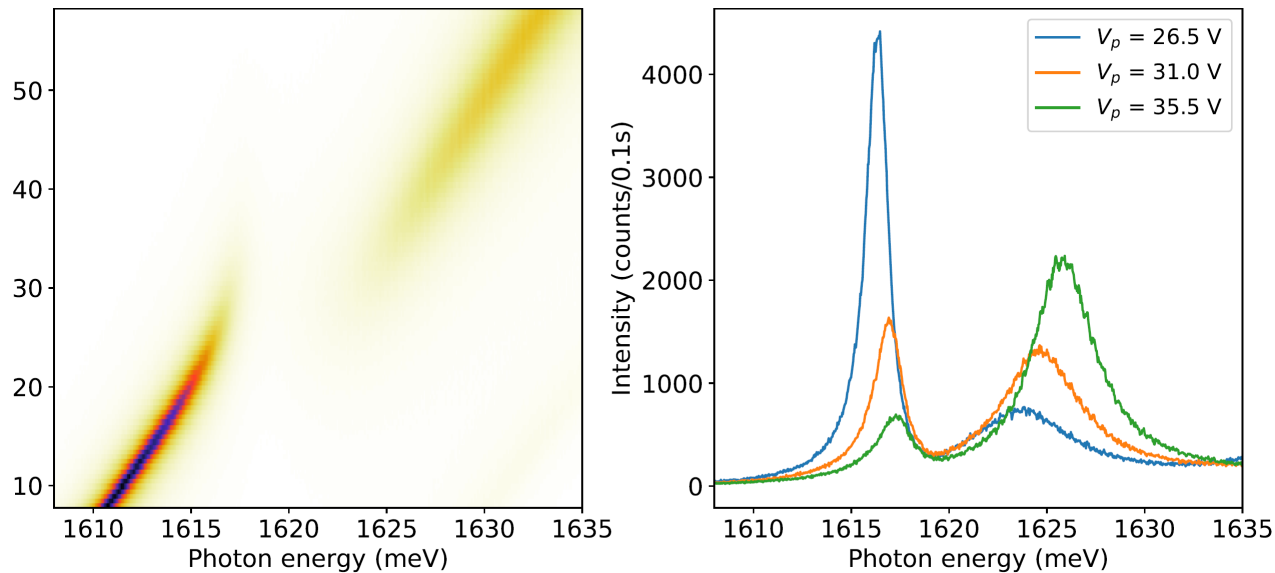
Monolayer is depleted of free electrons: only exciton resonance is visible: $\Omega_R = 18$ meV

exciton-polaritons

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

exciton-polaron-polaritons

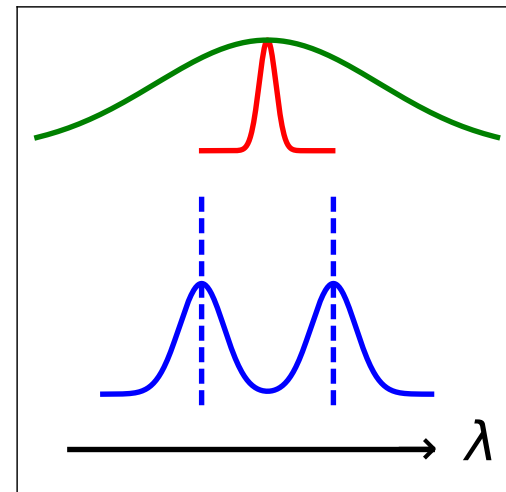
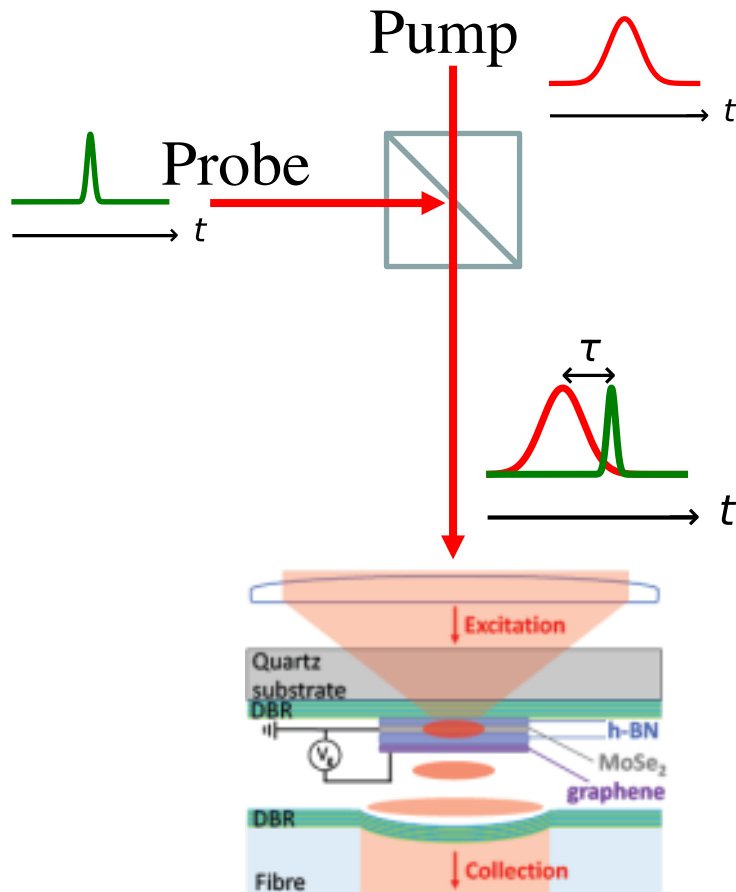
Attractive-polaron-polaritons



- Strong asymmetry between the lower and upper polaritons
- Efficient relaxation (only) of upper polaritons by Fermi sea
- Nonperturbative coupling between polarons and cavity-mode, or, between polaritons and electrons: enhanced nonlinearity?

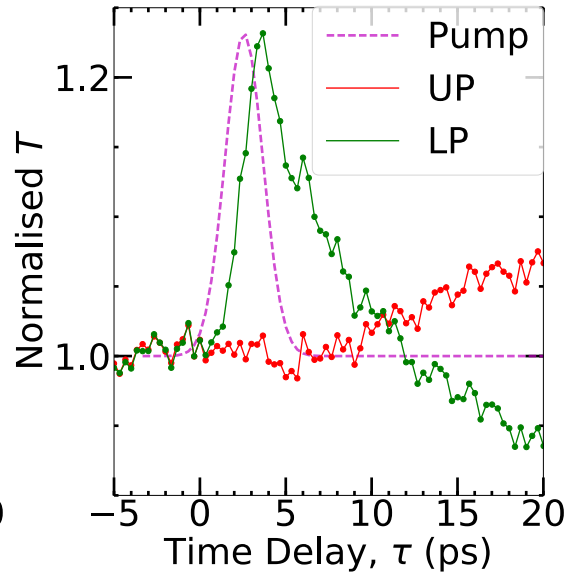
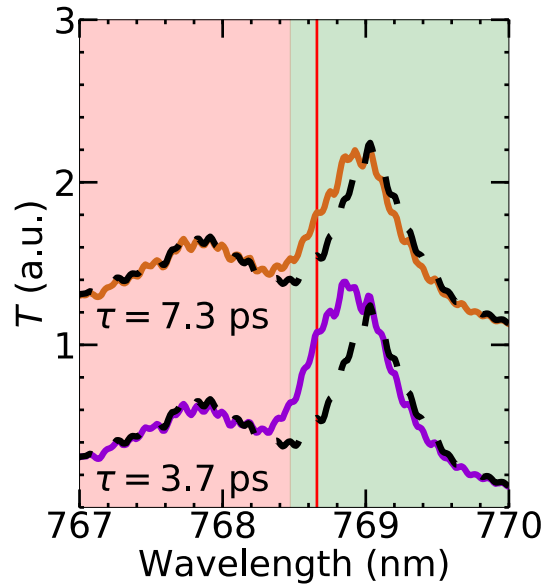
Going beyond linear response: Pump-probe experiments with TMD polaritons

- Broadband probe transmission as a function of time delay between 3 ps H-polarized pump and 0.5 ps V-polarized probe pulses
- Response of lower polaritons (LP) upon pumping in between the two branches



Both pump and probe lasers generate polaron-polaritons in distinct modes

Going beyond linear response: Pump-probe experiments with TMD polaritons



$$n_e = 8 \times 10^{11} \text{ cm}^{-2}$$

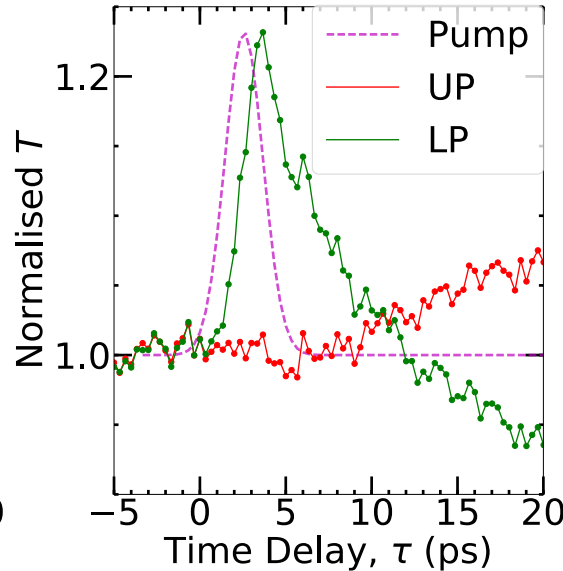
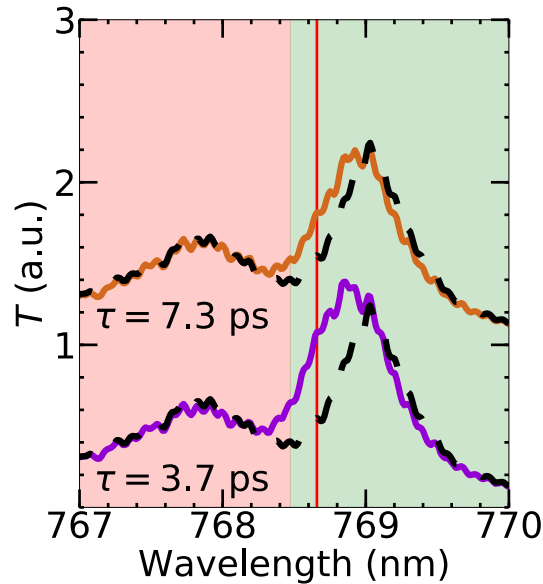
Polariton density:

$$(8 \pm 1) \times 10^{10} \text{ cm}^{-2}$$

enhanced polariton-polariton interactions:

$$U \sim 1 \text{ } \mu\text{eV } \mu\text{m}^2$$

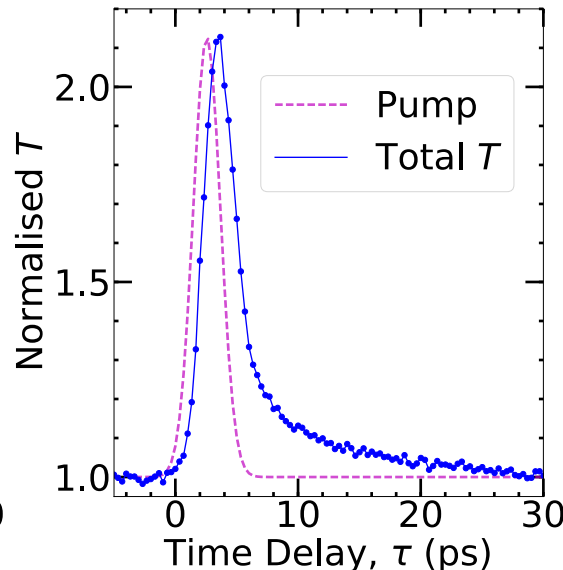
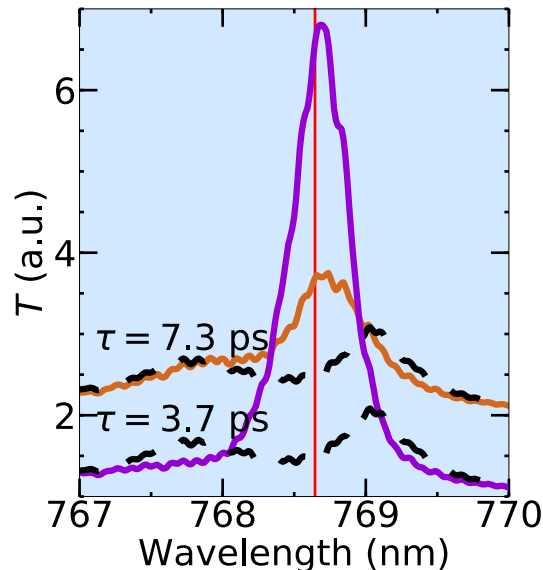
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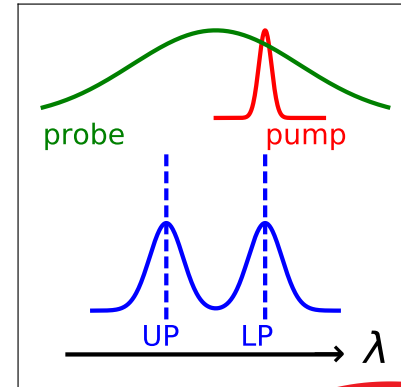
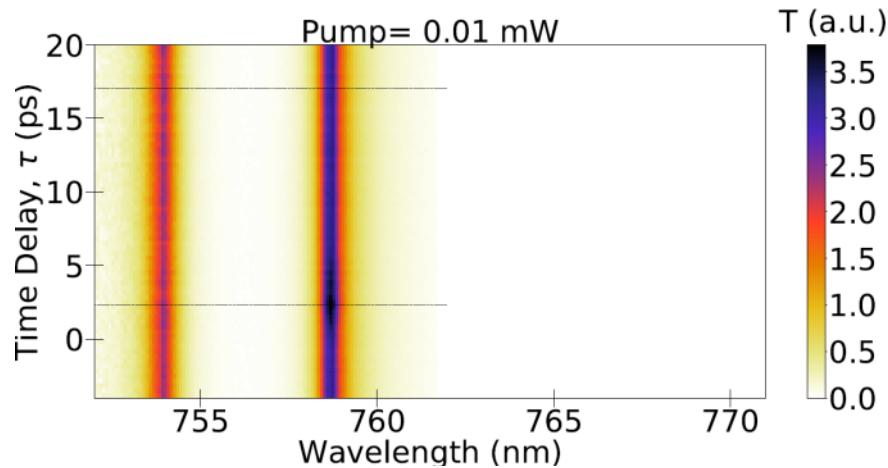
$$(8 \pm 1) \times 10^{10} \text{ cm}^{-2}$$



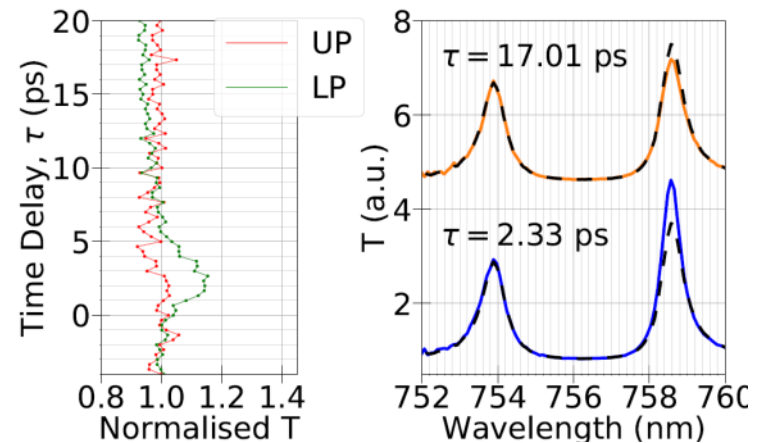
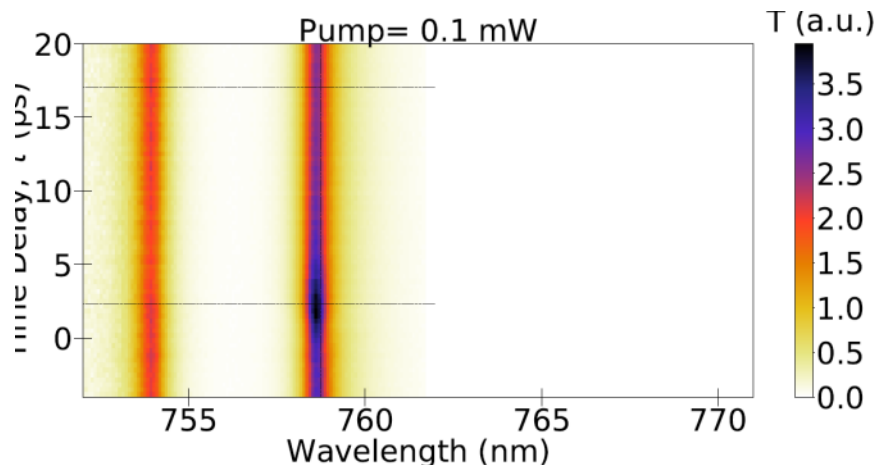
$$(8 \pm 1) \times 10^{11} \text{ cm}^{-2}$$

- Bose-enhanced cooling of pump-induced polarons
- Amplification of probe field by stimulated polaron-electron scattering

Pump-probe experiments with TMD polaritons (in the absence of electrons)

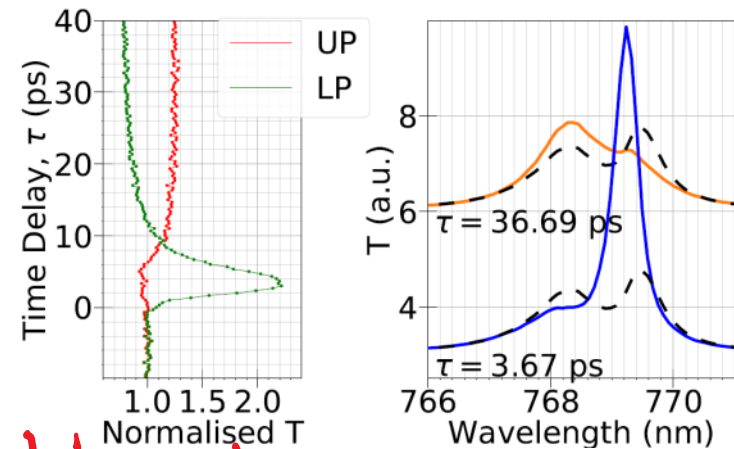
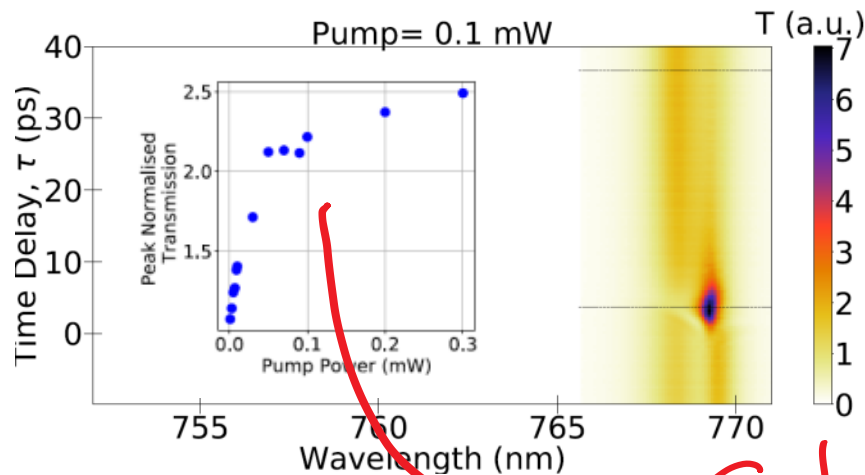
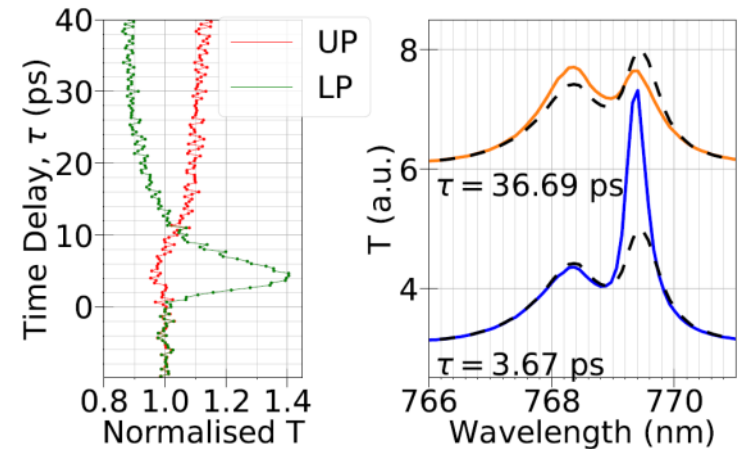
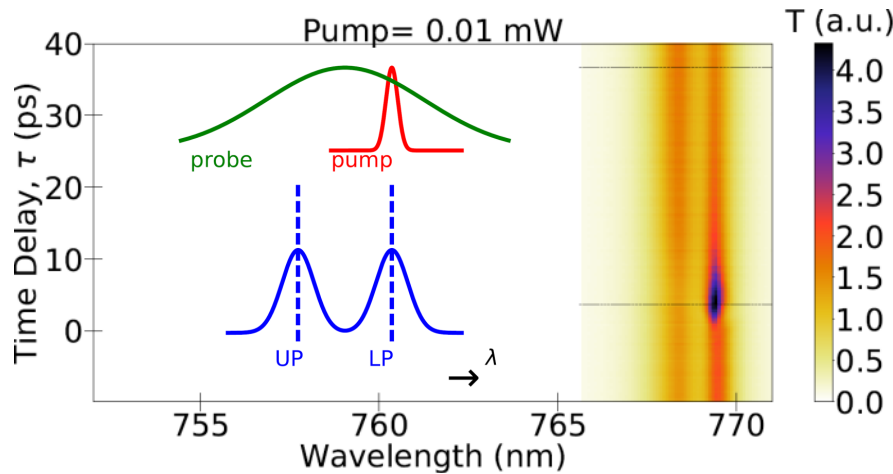


No change in pump laser transmission even for $n_{\text{pol}} = 1 \times 10^{12} \text{ cm}^{-2}$



Pump-probe with TMD polaron-polaritons

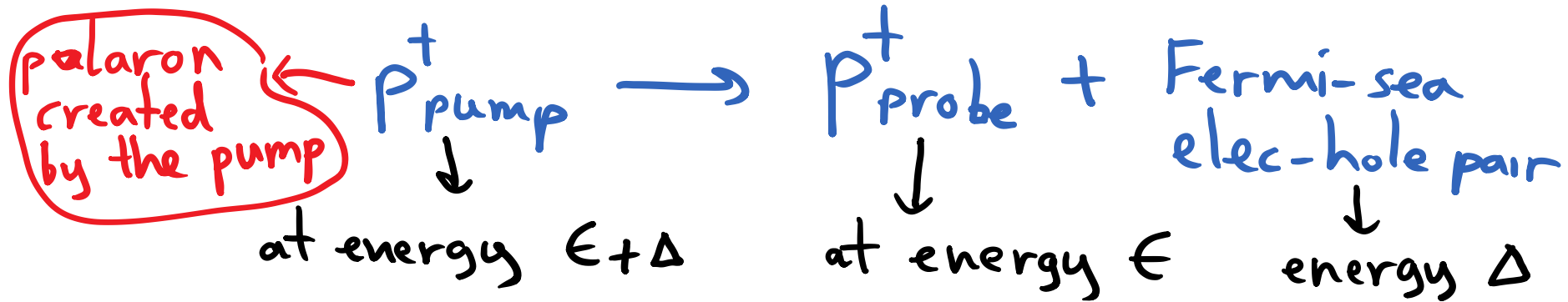
Response of exciton-polaron-polaritons upon pumping lower polariton (LP)



→ Saturation when $n_{elec} \approx n_{polar}$

How do we understand the optical gain?

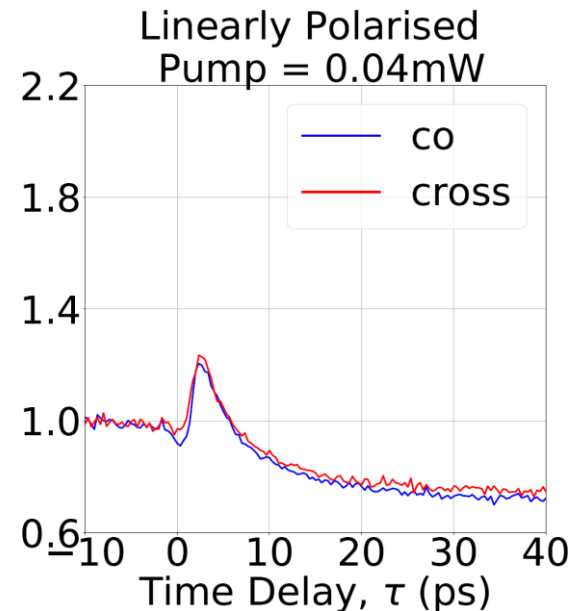
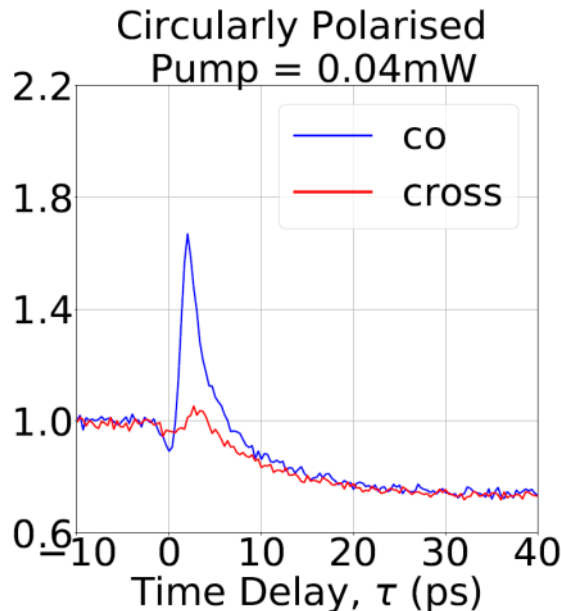
- Residual interaction between polarons & electrons lead to :



- Effective population inversion :

$$n_{\text{pump}} > \frac{kT}{\Delta}$$

Polarization dependent gain and relaxation of valley coherence



- Probe transmission at LP resonance upon pumping UP
- Circularly polarized excitation (K-valley) ensures that there is vanishing stimulated scattering for in cross-circularly polarized probe (K'-valley)
- Linearly polarized excitation (K+K') results in equal stimulated scattering into K+K' and K-K' polaron-polaritons:
 - ➡ Loss of valley coherence due to polaron-electron scattering!

How do we understand loss of valley coherence?

i) σ^+ excitation

$$|\text{polaron}_K\rangle \rightarrow |\text{polaron}_K\rangle \hat{e}_{K'}^+ \hat{e}_K$$

↳ generation of elec-hole pairs cannot flip valley.

ii) Linearly polar. excitation

$$|\text{pol}_K\rangle + |\text{pol}_{K'}\rangle \rightarrow |\text{pol}_K\rangle e_{K'}^+ e_K + |\text{pol}_{K'}\rangle e_K^+ e_K$$

Low mom. scattering
→ entanglement. ✓

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[O. Cotlet](#) (theory)

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Eugene Demler (Harvard)

Falko Pientka (Dresden)

Richard Schmidt (MPQ)