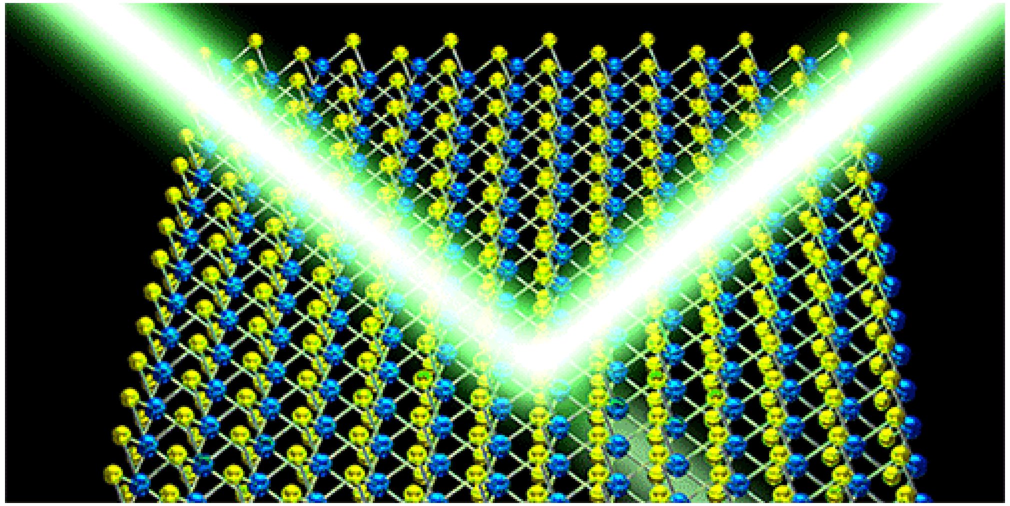
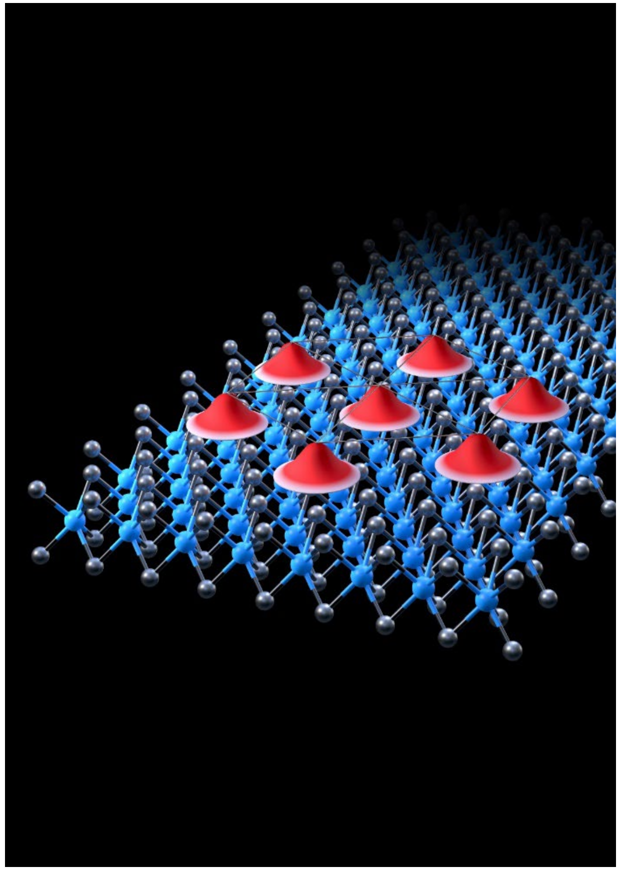
Quantum photonics in 2D materials: polariton-electron interactions

Atac Imamoglu

ETH Zurich





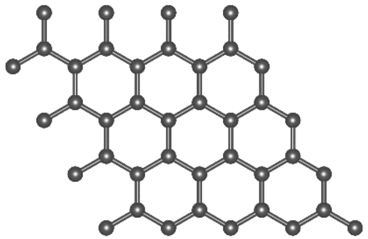
Outline

- Lecture 1: Elementary optical excitations of electron or hole-doped transition metal dichalcogenide (TMD) monolayers
- Lecture 2: Electrical control of optical properties: realization of quantum confined excitons in a monolayer p-i-n junction
- Lecture 3: Cavity-QED with excitons embedded in a degenerate electron system + moire physics with excitons (time permitting)

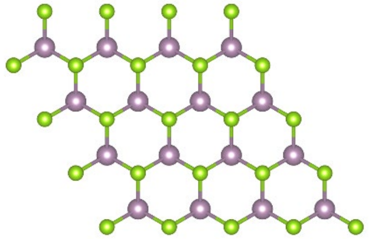
Why are two-dimensional (2D) materials interesting for photonics?

- Ability to use gates to control charging and optical properties: monolayer or bilayer p-i-n diodes, exciton-based light sources

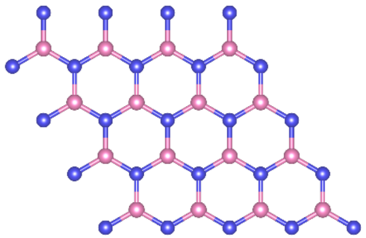
Graphene (metal)



TMD (semiconductor)



hBN (insulator)



Magnet, Superconductor, TI, …

- Layered materials with weak van der Waals bonds between the 2D sheets can be exfoliated down to a monolayer

- Monolayers retain (to a large extent) the properties/functionality of the bulk material.

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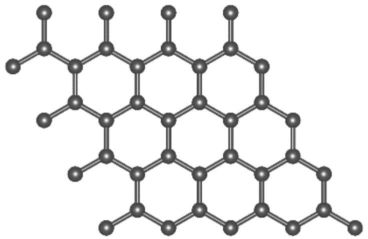

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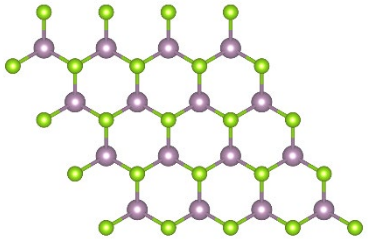

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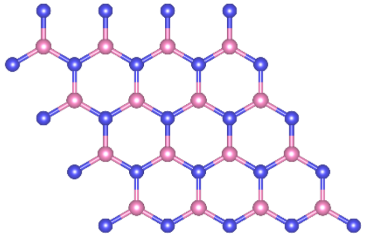
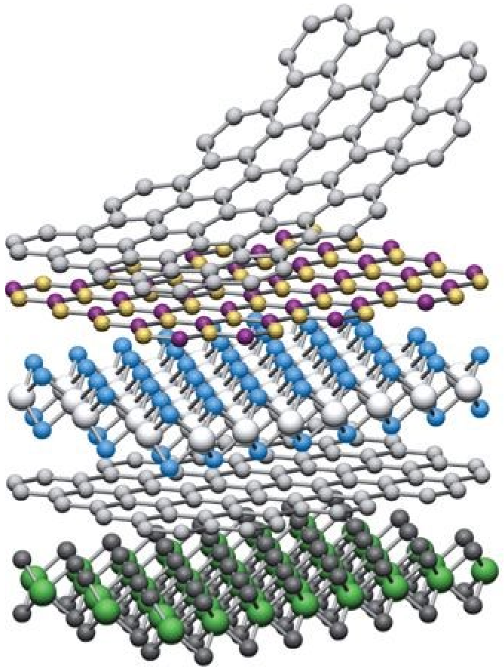


Magnet, Superconductor, TI, …

Van der Waals heterostructure

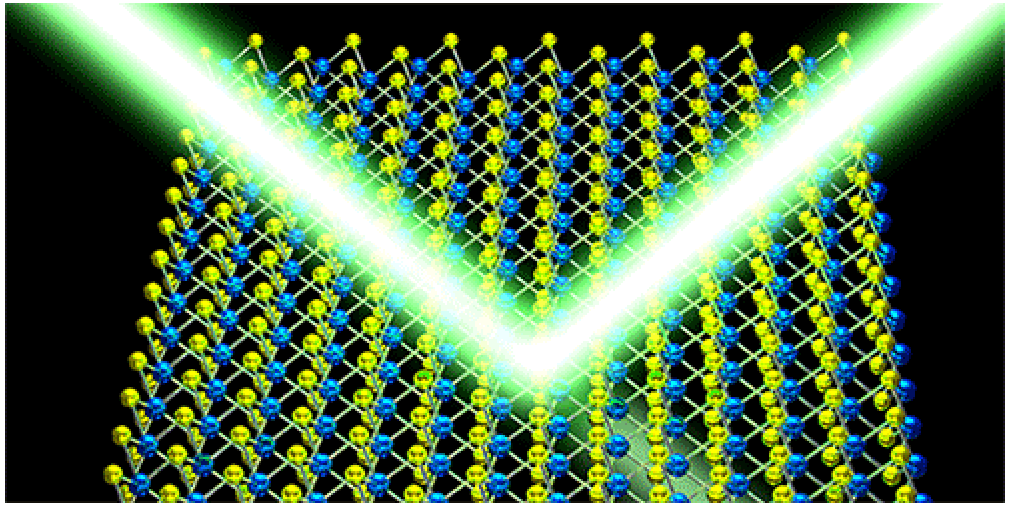


A. K. Geim *et al.*, Nature **499**, 419 (2013)

New (optical) functionality upon stacking different mono-layers

Why are two-dimensional (2D) materials interesting for photonics?

- Ability to use gates to control charging and optical properties: monolayer or bilayer p-i-n diodes, exciton-based light sources
- Very strong linear optical response ensured by tightly bound excitons (small Bohr radius a_B): <u>atomically thin mirror</u>



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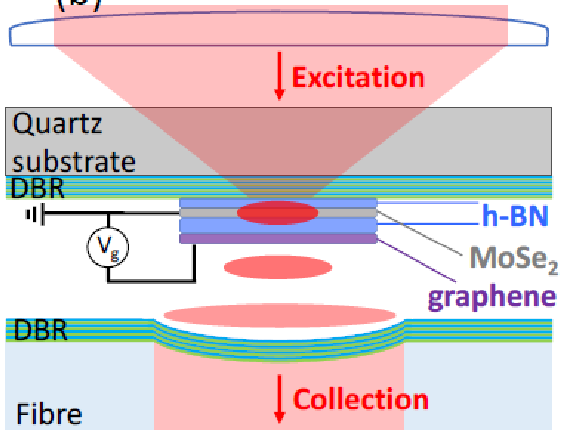
(b)



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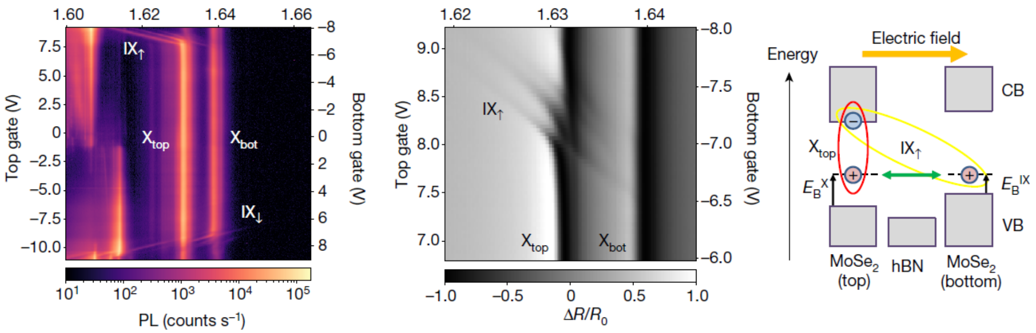

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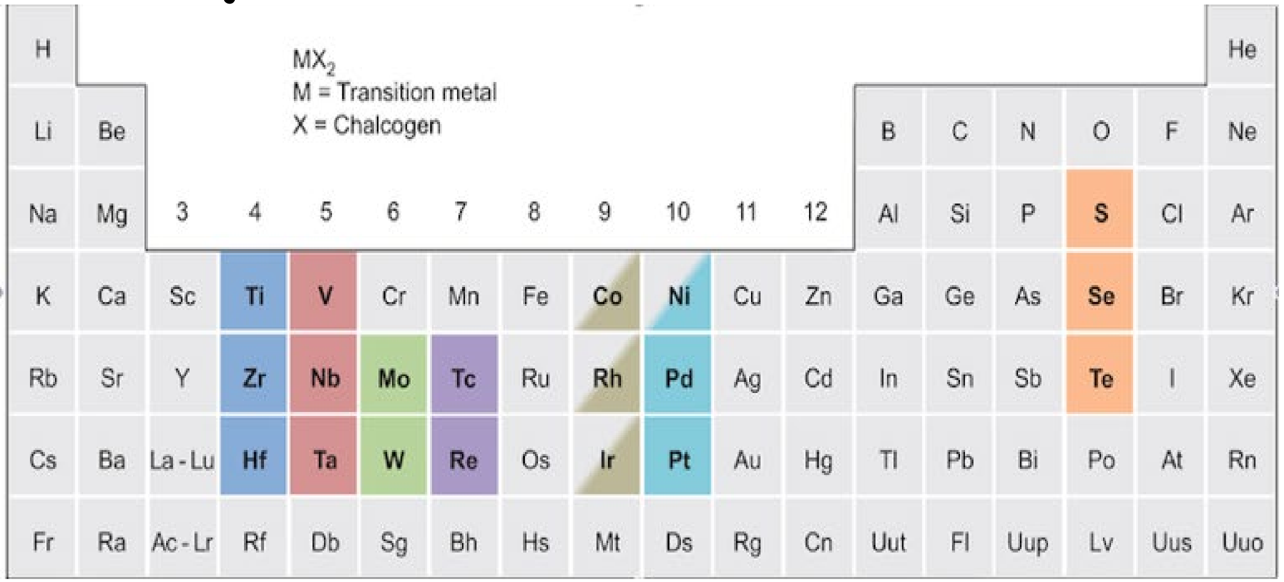
- Layer degree of freedom: long-lived dipolar optical excitations
- Excitons and exciton-polarons sense/detect many-body electronic states: spectroscopy

Materials: Transition metal dichalcogenides (TMD) - layered 2D semiconductors

Formula: MX_2

M = Transition Metal

X = Chalcogen



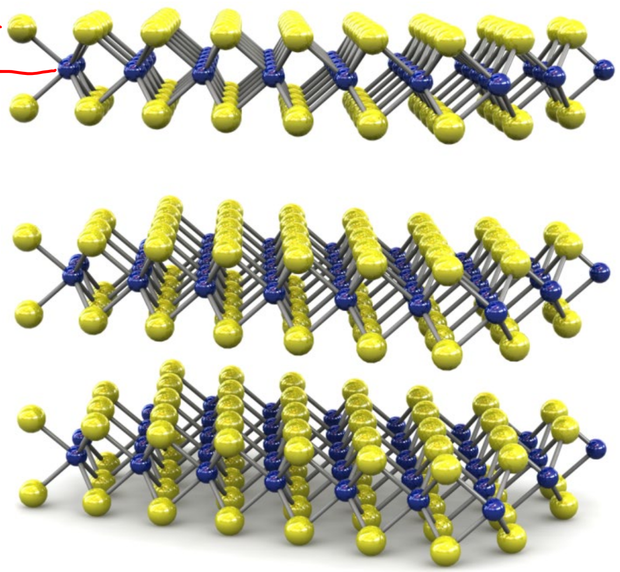
Electrical property	Material
Semiconducting	MoS_2 $MoSe_2$ WS_2 WSe_2 $MoTe_2$ WTe_2
Semimetallic	TiS_2 $TiSe_2$
Metallic, CDW, Superconducting	$NbSe_2$ NbS_2 $NbTe_2$ TaS_2 $TaSe_2$ $TaTe_2$

Layered materials

Se

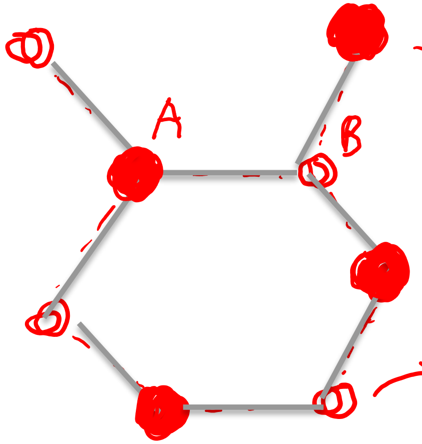
Mo

Se



effective monolayer

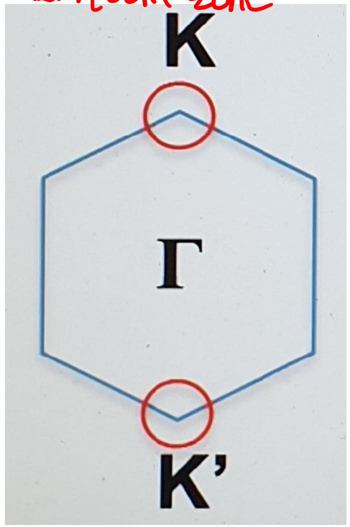
Honeycomb lattice



Mo atom

2 Se atoms

Brillouin zone



How do we understand optical transitions?

A) Description of the band structure using massive Dirac model (direct bandgap)

$$H_\tau = \begin{bmatrix} \Delta/2 & v_F(\tau k_x - i k_y) \\ v_F(\tau k_x + i k_y) & -\Delta/2 \end{bmatrix} \quad \left\{ \begin{array}{l} \tau = +1 \leftrightarrow K \\ \quad\; -1 \leftrightarrow K' \end{array} \right.$$

$\Delta =$ Band-gap.

acts on

$$\begin{bmatrix} A \\ B \end{bmatrix} = \begin{bmatrix} |d_{z^2}\rangle \\ |d_{x^2-y^2}\rangle + i\tau |d_{xy}\rangle \end{bmatrix}$$

Can be justified starting from 6 band tight binding approach

B) Light - matter coupling

- $k_i \longrightarrow k_i - eA_i$ $(i = x, y)$; $\vec{k}$ measured from K, K'
- K valley , RCP light $\vec{A} = \vec{A}_x + i\vec{A}_y = A_0(\hat{x} + i\hat{y})\hat{a}$ + h.c.

$$\Rightarrow H_{RCP} = \begin{bmatrix} \Delta/2 & v_F(k_x - ik_y + 2eA_0\hat{a}) \\ v_F(k_x + ik_y + 2eA_0\hat{a}^{\dagger}) & -\Delta/2 \end{bmatrix}$$

VB couples to CB by annihil. a photon.

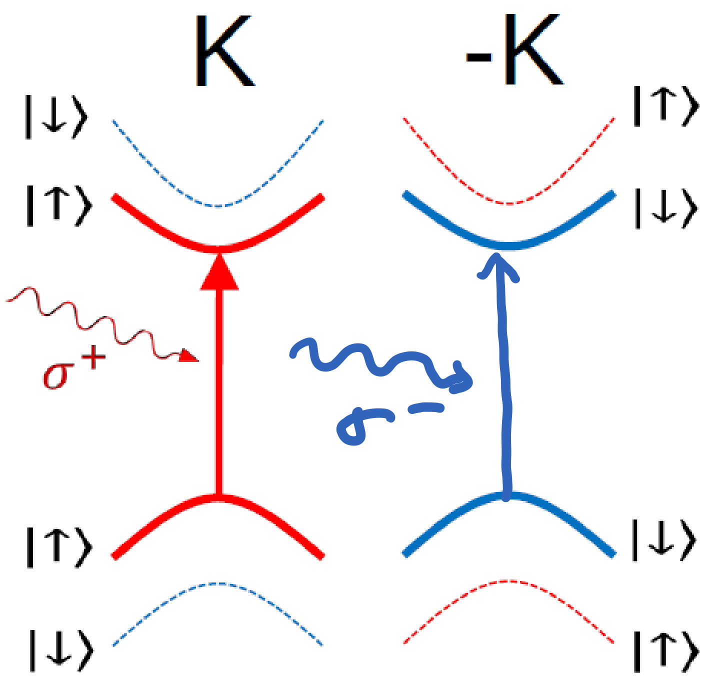
similarly for LCP light (in RWA)

$$H_{LCP} = \begin{bmatrix} \Delta/2 & v_F(k_x - ik_y) \\ v_F(k_x + ik_y) & -\Delta/2 \end{bmatrix}$$

i.e no coupling to LCP light

Simplified band structure

conduction band (CB)



valence band (VB)

$k=0$ CB: d_{z^2}

VB: $d_{x^2-y^2} + i d_{xy}$

- Spin-orbit locks the spin & valley degrees of freedom

- Elementary optical excitations are excitons: bound state of a conduction band electron & a valence band hole

$-\ +$

Excitons in 5 different TMD monolayers

Table 1 Fundamental optoelectronic material parameters of monolayer TMD semiconductors

Material	m_r (m_0)	E_b (meV)	E_{gap} (eV)	κ	r_0 (nm)	r_{1s} (nm)
hBN | MoS_2| hBN	0.275 ± 0.015	221	2.160	4.45	3.4	1.2
hBN | $MoSe_2$| hBN	0.350 ± 0.015	231	1.874	4.4	3.9	1.1
hBN | $MoTe_2$| hBN	0.360 ± 0.040	177	1.352	4.4[a]	6.4	1.3
hBN | WS_2| hBN	0.175 ± 0.007	180	2.238	4.35	3.4	1.8
hBN | WSe_2| hBN[b]	0.20 ± 0.01	167	1.890	4.5	4.5	1.7

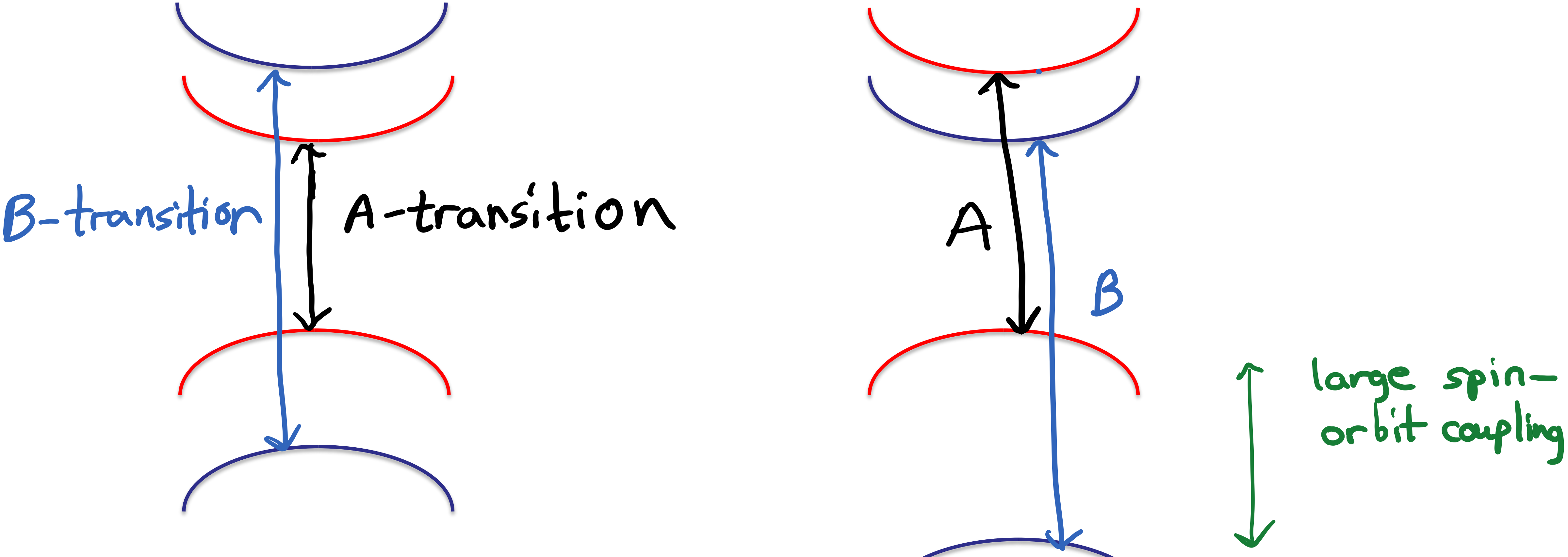
Experimentally determined values of the exciton reduced mass m_r, the 1s exciton binding energy E_b, the free-particle bandgap E_{gap}, the dielectric screening parameters r_0 and κ, and the root-mean-square radius of the 1s exciton r_{1s}. Typical error bars on experimental values of E_b and E_{gap} are ±3 meV. Typical error bars on values of r_0 and r_{1s} are ±0.1 nm, except for $MoTe_2$, where they are ±0.3 nm

[a]The value of κ for $MoTe_2$ is assumed to be 4.4 and is not a fitting parameter (see text for details)

[b]Values for hBN-encapsulated WSe_2 are taken from ref. [27]

Full exciton spectrum: A & B excitons

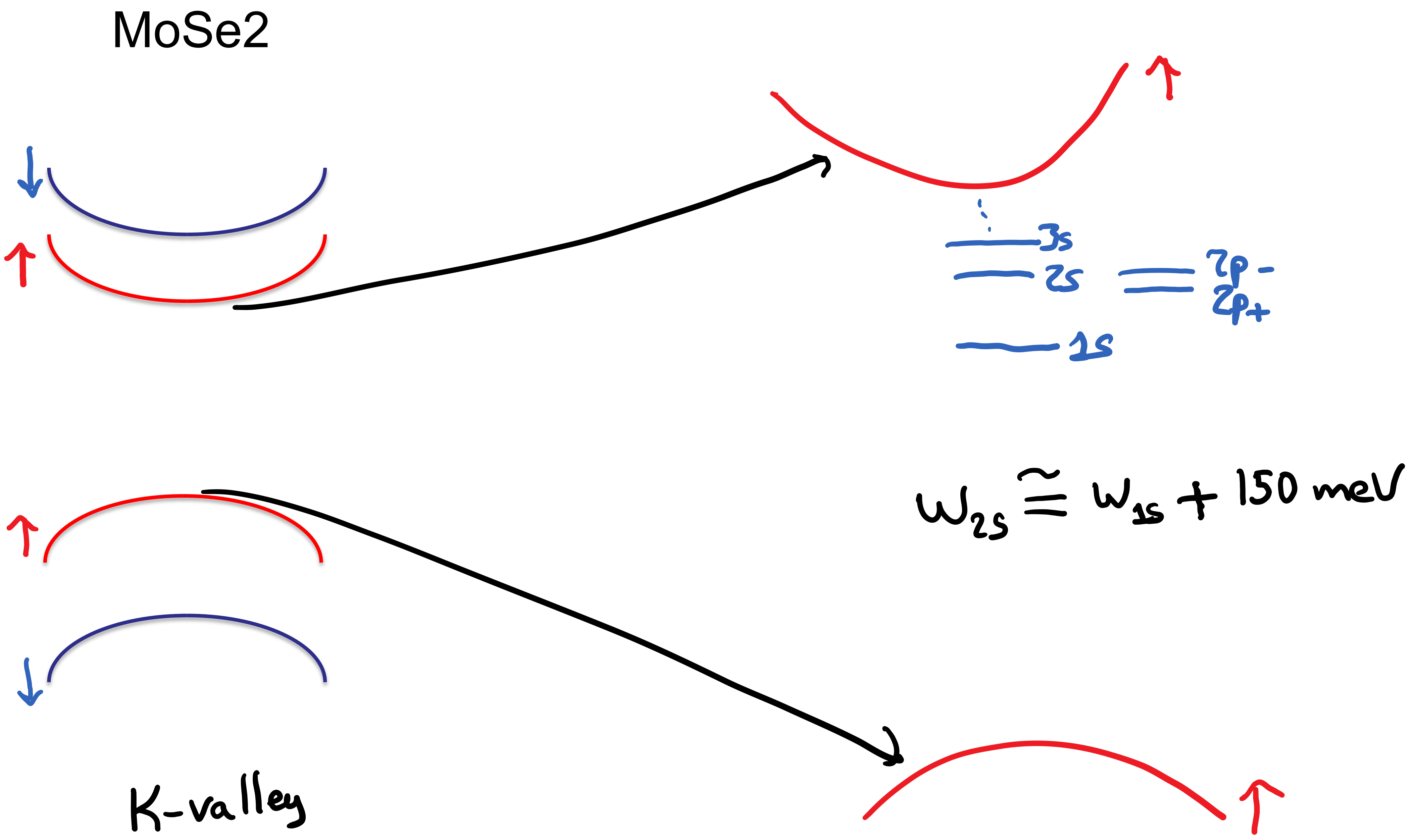
MoSe2 WSe2



$\omega_B \simeq \omega_A + 150\ \text{meV}$ ($MoSe_2$)

$\omega_B \simeq \omega_A + 400\ \text{meV}$ (WSe_2)

Full exciton spectrum: Rydberg excitons



Resonant reflection spectroscopy

- For excitons that are broadened exclusively by radiative decay, $\boxed{\text{resonant transmission} = 0}$

$$\Rightarrow \Gamma_{rad} \propto \frac{1}{a_B^2} \gg \gamma_{dephasing} \Rightarrow R \sim 1.$$

$\Rightarrow$ Highest specular reflection reported $R \sim 0.85$

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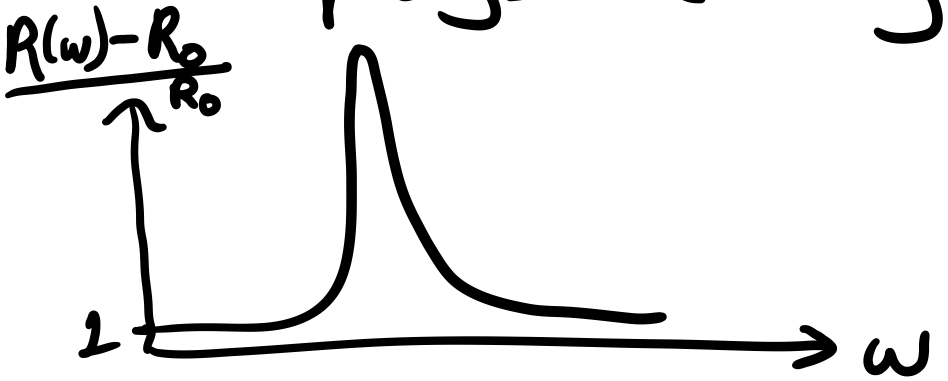
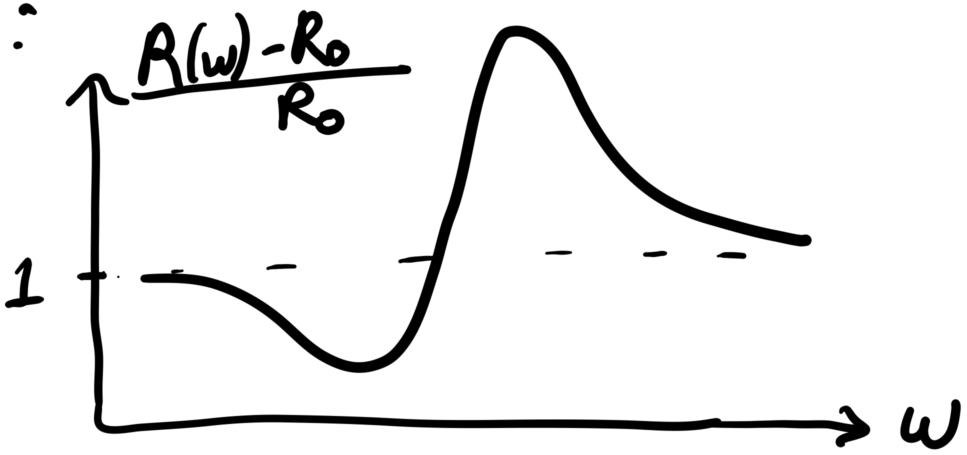
- The thickness of the surrounding material plays a key role:



or



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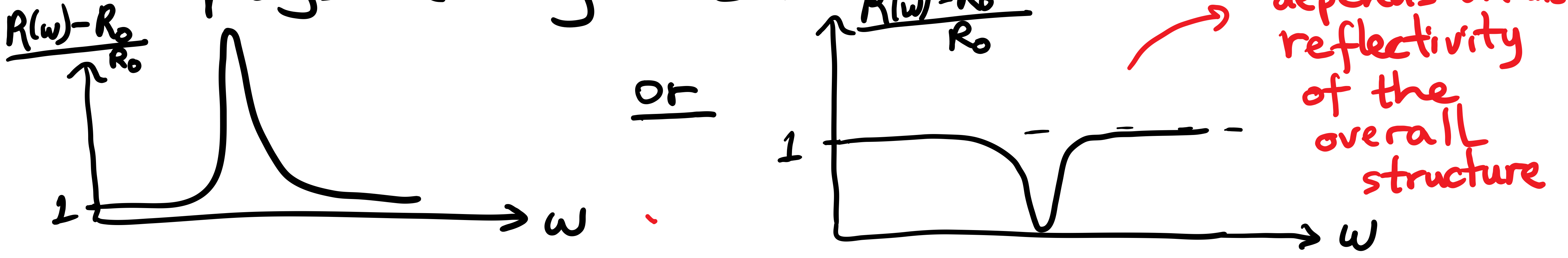
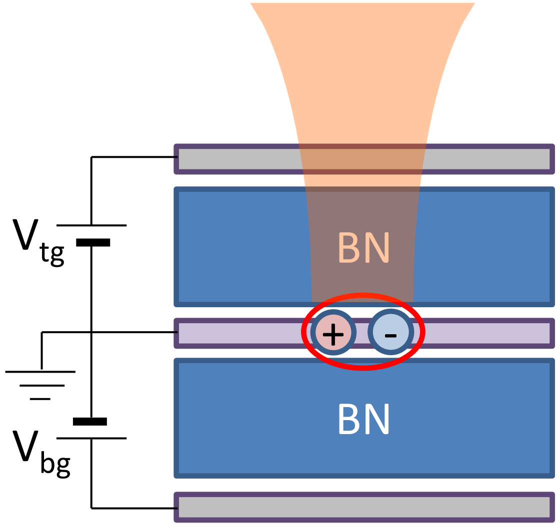


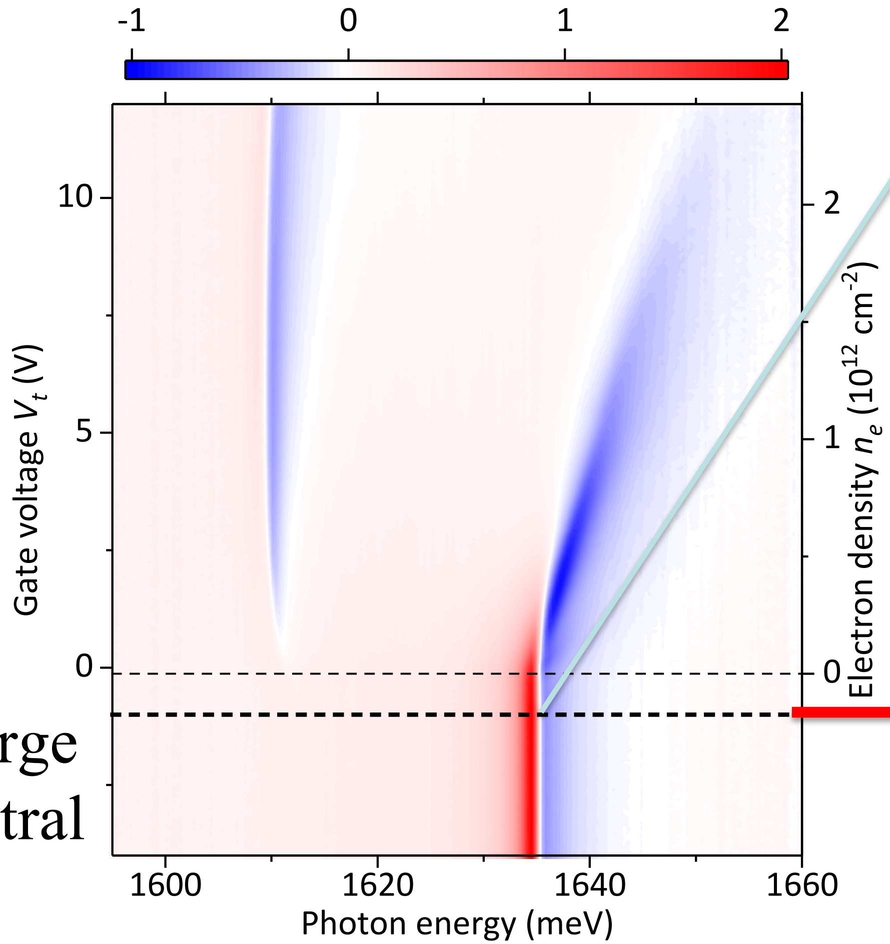
Charge tunable van der Waals heterostructures

- Exfoliation of and stacking of monolayers of semiconducting TMDs and graphene, together with ~50 nm thick insulating boron nitride (BN) layers
- A gate voltage applied between the top/bottom (transparent) graphene gate and the MoSe2 layer allows for tuning the electron/hole density

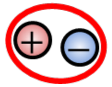


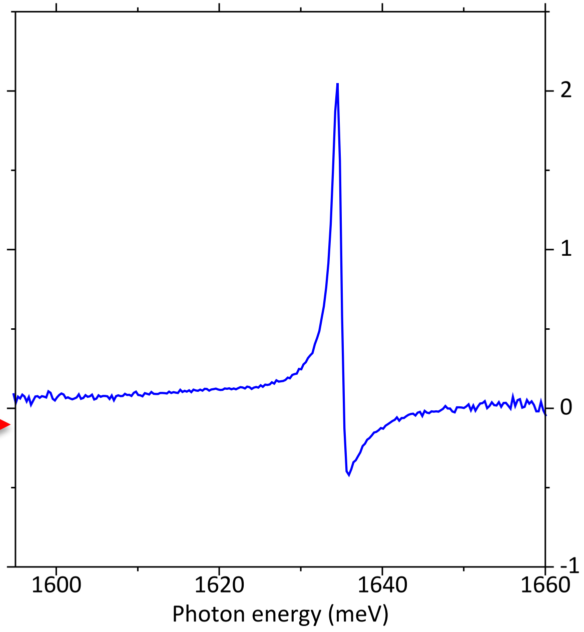
Elementary optical excitations in monolayer $MoSe_2$

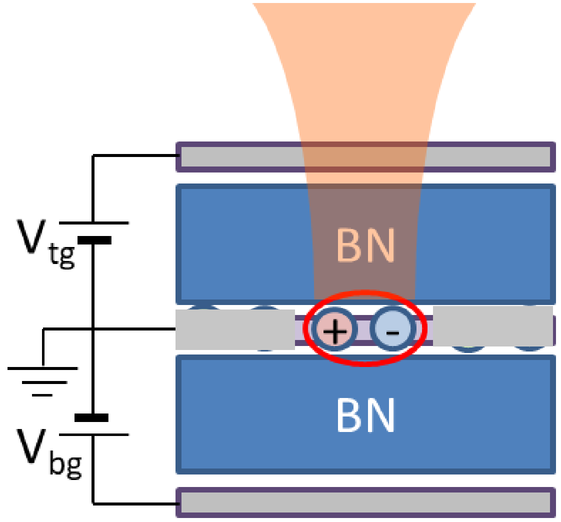
<u>Charge neutrality:</u> tightly bound 1s exciton dominates



charge
neutral

Exciton 

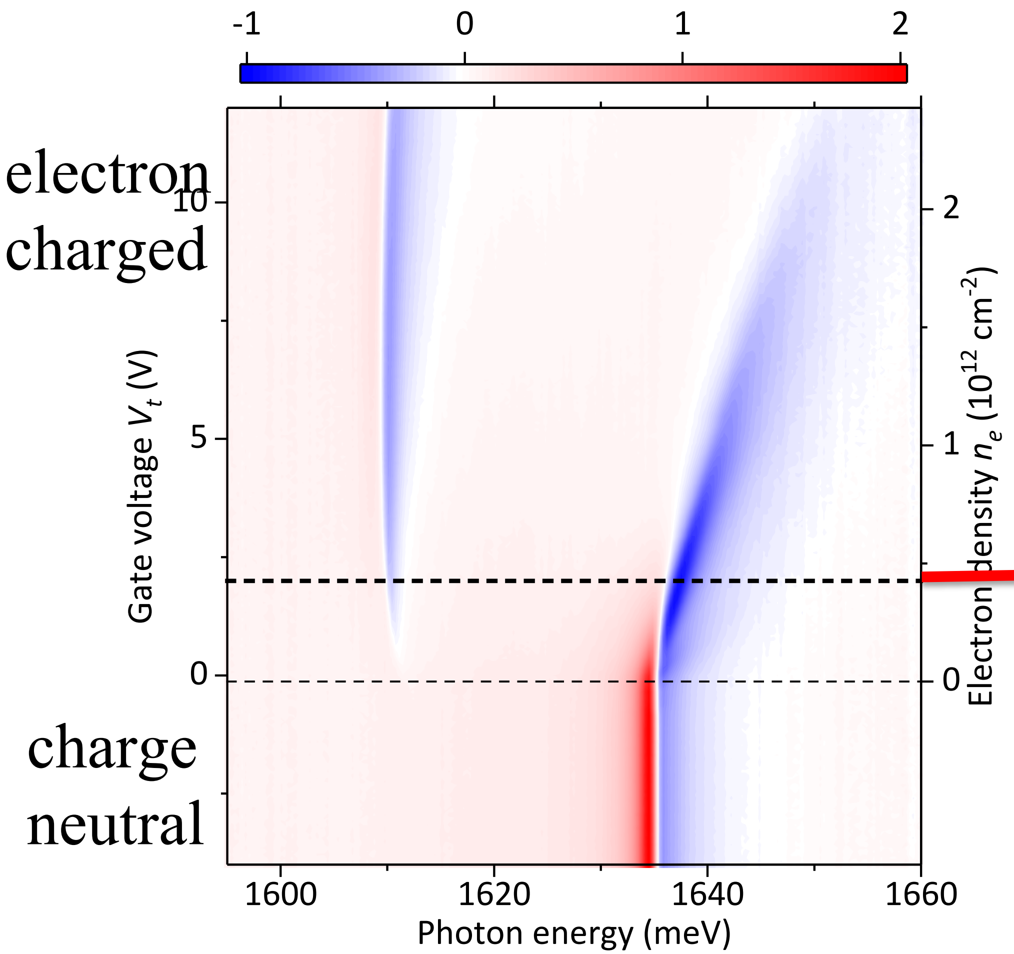


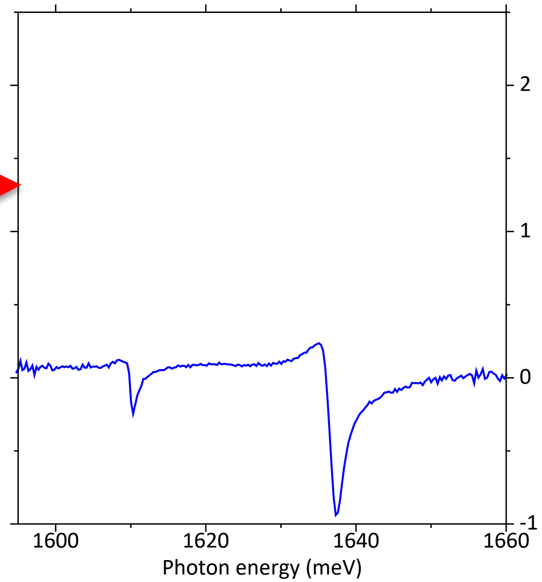


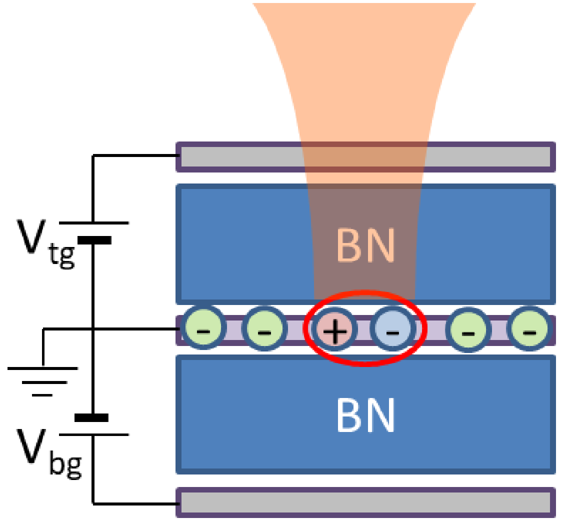
Elementary optical excitations in monolayer $MoSe_2$

<u>Charge neutrality:</u> tightly bound 1s exciton dominates

<u>Finite electron or hole density:</u> spectrum is drastically modified



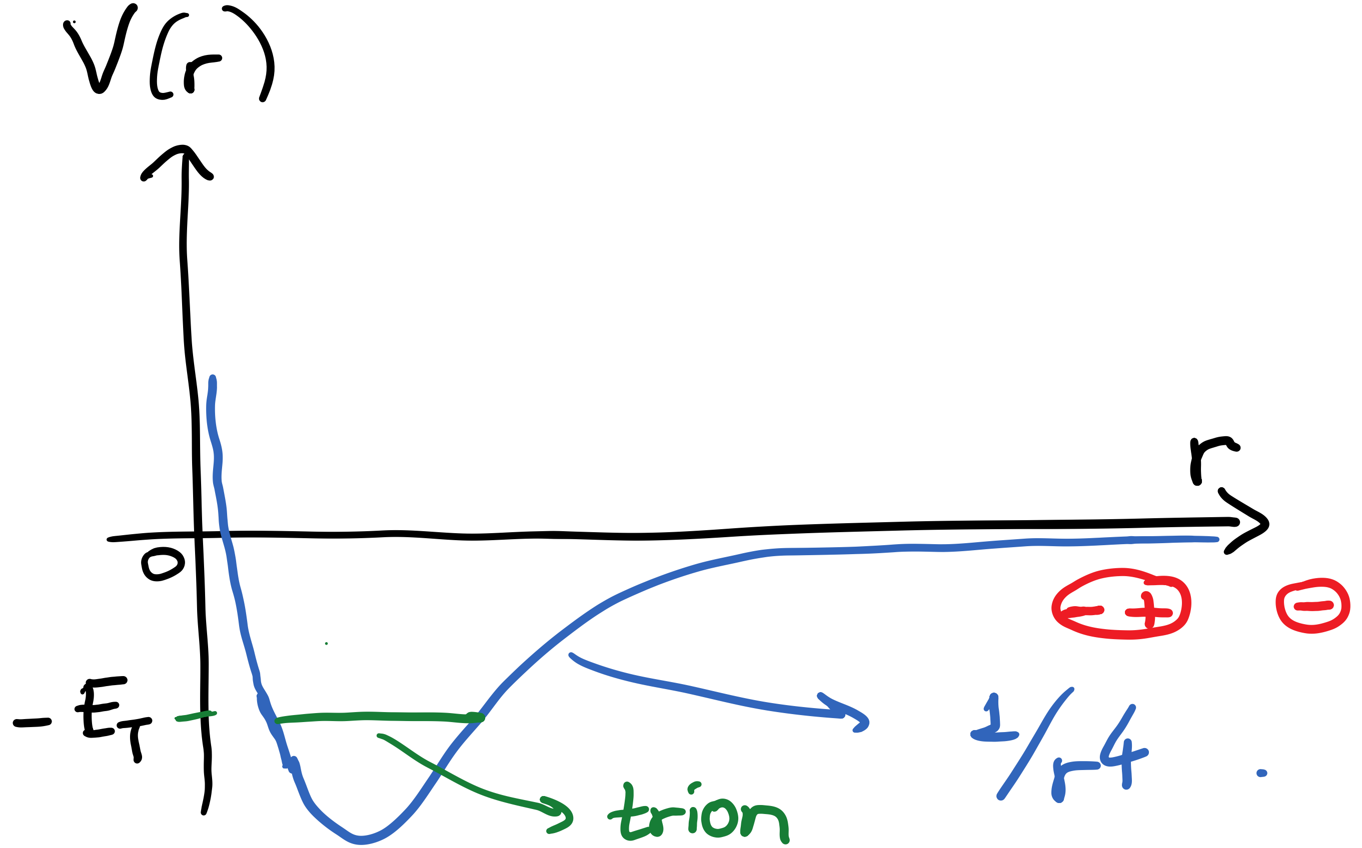




How to understand the modified spectrum: Exciton-electron scattering in a monolayer TMD

- Excitons are neutral bosonic optical excitations (quanta of electronic polarization wave) that interact with itinerant electrons or holes and form a bound molecular state termed "trion"

exciton $-+$
electron $-$
r = separation of an exciton & electron



Exciton as a mobile impurity in a degenerate electron system

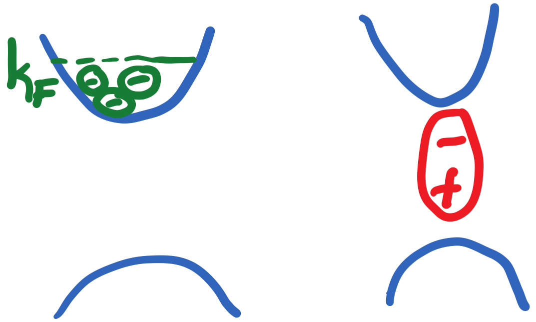
Exciton-electron interactions

- We consider a single exciton, optically injected into a 2D electron system (2DES) with Fermi energy/momentum E_F/k_F

$$H = \sum_{k_1} w_x(k_1)\, x^\dagger_{k_1} x_{k_1} + \sum_{k_2} \epsilon(k_2)\, e^\dagger_{k_2} e_{k_2}$$

$$- V \sum x^\dagger_{k_1+q}\, e^\dagger_{k_2-q}\, e_{k_2}\, x_{k_1}$$

- We assume all electrons in -K valley (optical pumping and an exciton in K valley



- To make the problem tractable, we consider a truncated basis to represent many-body eigenstates:

$$|\Psi_p\rangle = \Big[\underbrace{\phi_p}_{\text{quasi-particle weight}} x_p^{\dagger} + \sum_{k,q} \varphi(k,q) \underbrace{x_{p+q-k}^{\dagger} e_k^{\dagger} e_q}_{\substack{x \text{ generates an electron} \\ (e_k^{\dagger}\ k>k_F)\ \&\ \text{hole}\ (e_q\ q\lesssim k_F) \\ \text{pair}}}\Big]|\underline{0}\rangle$$

Chevy Ansatz

↳ denotes a $T=0$ Fermi sea

↳ Exact eigenstates will have $\lesssim N_e$ electron-hole pair excitations (N_e = # of electrons)

↳ When is it justified to use Chevy?

i) Impurity is very light; then the momentum kick

$x_p^\dagger \to x^\dagger_{p+q-k}$ is energetically costly

ii) Impurity has a short lifetime

applies to TMD excitons.

- To find the spectrum for $p=0$ (optically excitable exciton) we minimize

$$\langle \Psi_{p=0} | H - E | \Psi_{p=0} \rangle$$

i.e. set $\frac{\partial}{\partial \phi_0^*} \langle H - E \rangle = 0$ & $\frac{\partial}{\partial \varphi^*(k,q)} \langle H - E \rangle = 0$

i) $\left[E - \omega_x(0) + V \sum_{q} \right] \phi_0 = -V \sum \varphi(k,q)$

ii) $\left[E - \omega_x(q-k) - \epsilon(k) + \epsilon(q) \right] \varphi(k,q) =$

$$\underbrace{-V \phi_0}_{\text{bare exciton } x_0^+ \text{ scatters an electron-hole pair}} \underbrace{- V \sum_{k'>k_F} \varphi(k',q)}_{\text{exciton scatters an electron with } k'>k_F \text{ to } k>k_F} \underbrace{+ V \sum_{q'\leq k_F} \varphi(k,q')}_{\text{exciton scatters a (Fermi sea) hole from } q' \text{ to } q.}$$

$\rightarrow$ Last term can be neglected

- By letting $\chi(q) = \phi_0 + \sum_k \varphi(k,q)$, we can solve the eqns for $E_{p=0}$

$$E_{p=0} = \omega_x(0) + \underbrace{\sum_{q=0}^{k_F} \left[\frac{1}{V} - \sum_{k=k_F}^{\Omega_c} \frac{1}{E_{p=0} - \omega_x(q-k) - \epsilon(k) + \epsilon(q)} \right]^{-1}}_{\Sigma_x(E_{p=0})}$$

$\Sigma_x(E_{p=0})$: exciton self-energy

$\Rightarrow$ Absorption spectrum :

$$A(\omega) = \mathrm{Im}\left[\frac{1}{\omega + i\eta - \omega_x(0) - \Sigma_x(\omega)} \right]$$

Exciton-polaron-polaritons

- If the 2D material is inside a cavity, then

$$H \rightarrow H + \sum_p \omega_c(p)\, a_p^\dagger a_p + \sum_p g_c(p)\left[a_p^\dagger x_p + x_p^\dagger a_p\right]$$

- Due to steep cavity dispersion, we only need to consider exciton dressing; i.e.

$$|\psi_{n,p}\rangle = \left[\eta_{n,p}\, a_p^\dagger + \phi_{n,p}\, x_p^\dagger + \sum_{k,q} \varphi_n(k,q)\, x^\dagger_{p+q-k}\, e^\dagger_k e_q\right]|FS\rangle$$

yields:

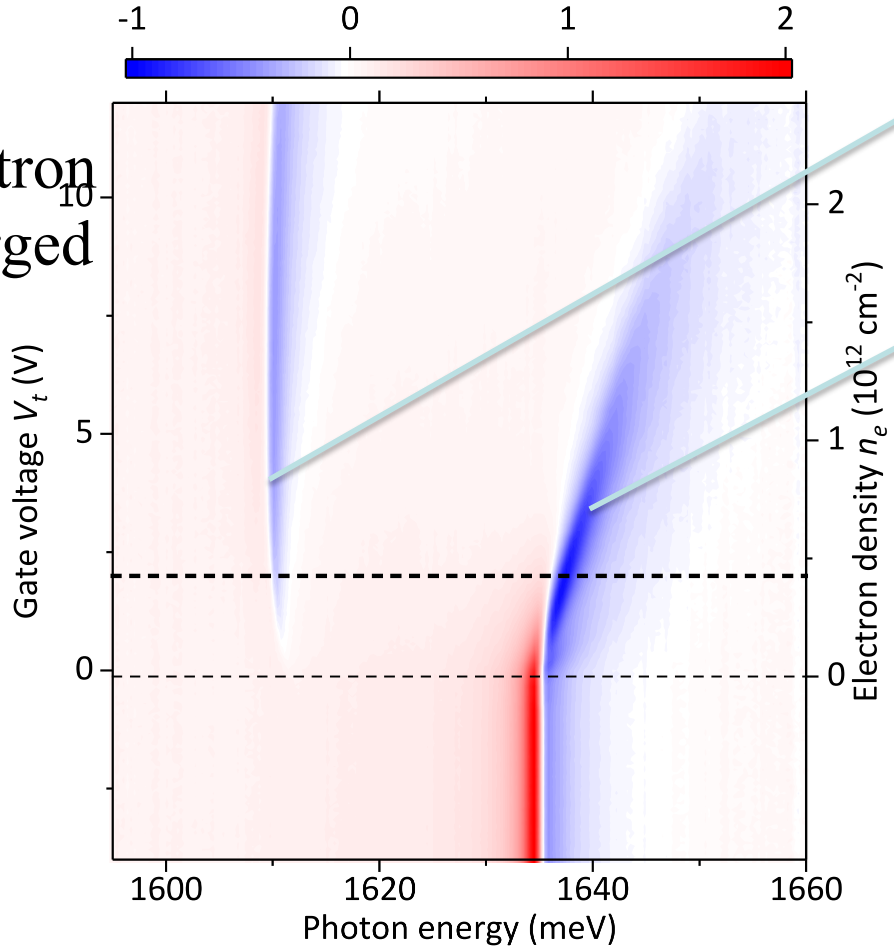
$$E_{n,p} = \omega_x(p) + \Sigma_x(E_{n,p}) + \frac{g_c^2}{E_{n,p} - \omega_c(p)}$$

Elementary optical excitations in monolayer $MoSe_2$

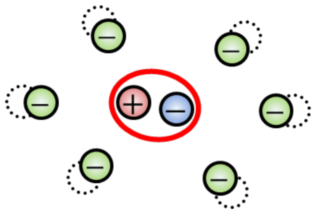
Charge neutrality: tightly bound 1s exciton dominates

Finite electron or hole density: exciton-polarons (many-body excitations)

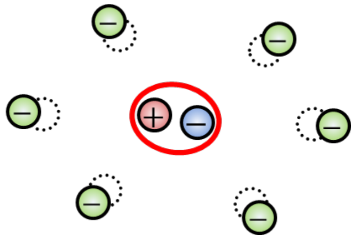
electron charged

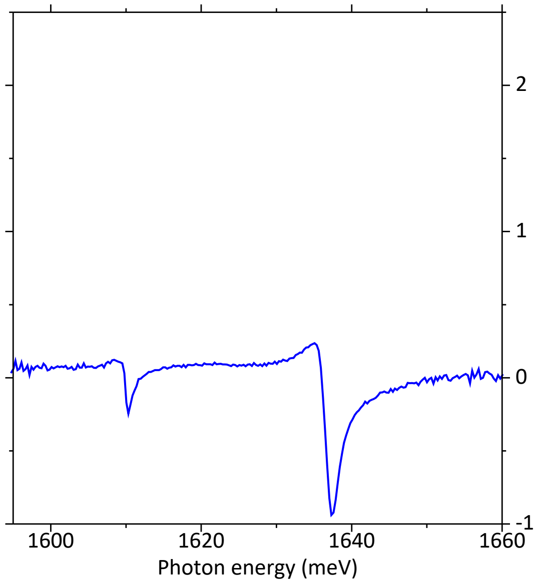


Attractive-polaron (AP)



Repulsive-polaron (RP)





Energy shift of Exciton
(Repulsive polaron)
→ Charge sensing

Polaron vs. Trion : analogy to Tavis-Cummings (TC) model

$\boxed{\text{TC}}$

$|1_c\rangle$ $\updownarrow \Delta$ $|0_c\rangle$ — cavity mode

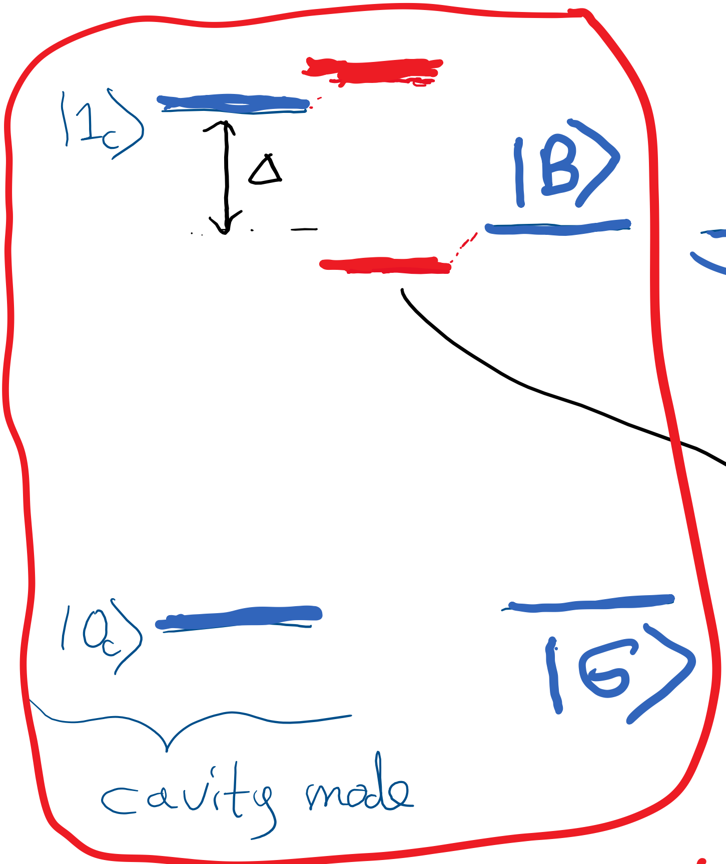
$|e\rangle_1$ $|e_2\rangle$ . . . $|e_N\rangle$

$|g\rangle_1$ $|g\rangle_2$ . . . $|g\rangle_N$

ensemble of N atoms, each with weak coupling to the cavity

Polaron vs. Trion : analogy to Tavis-Cummings (TC) model

$\boxed{\text{TC}}$



N-1 dark states

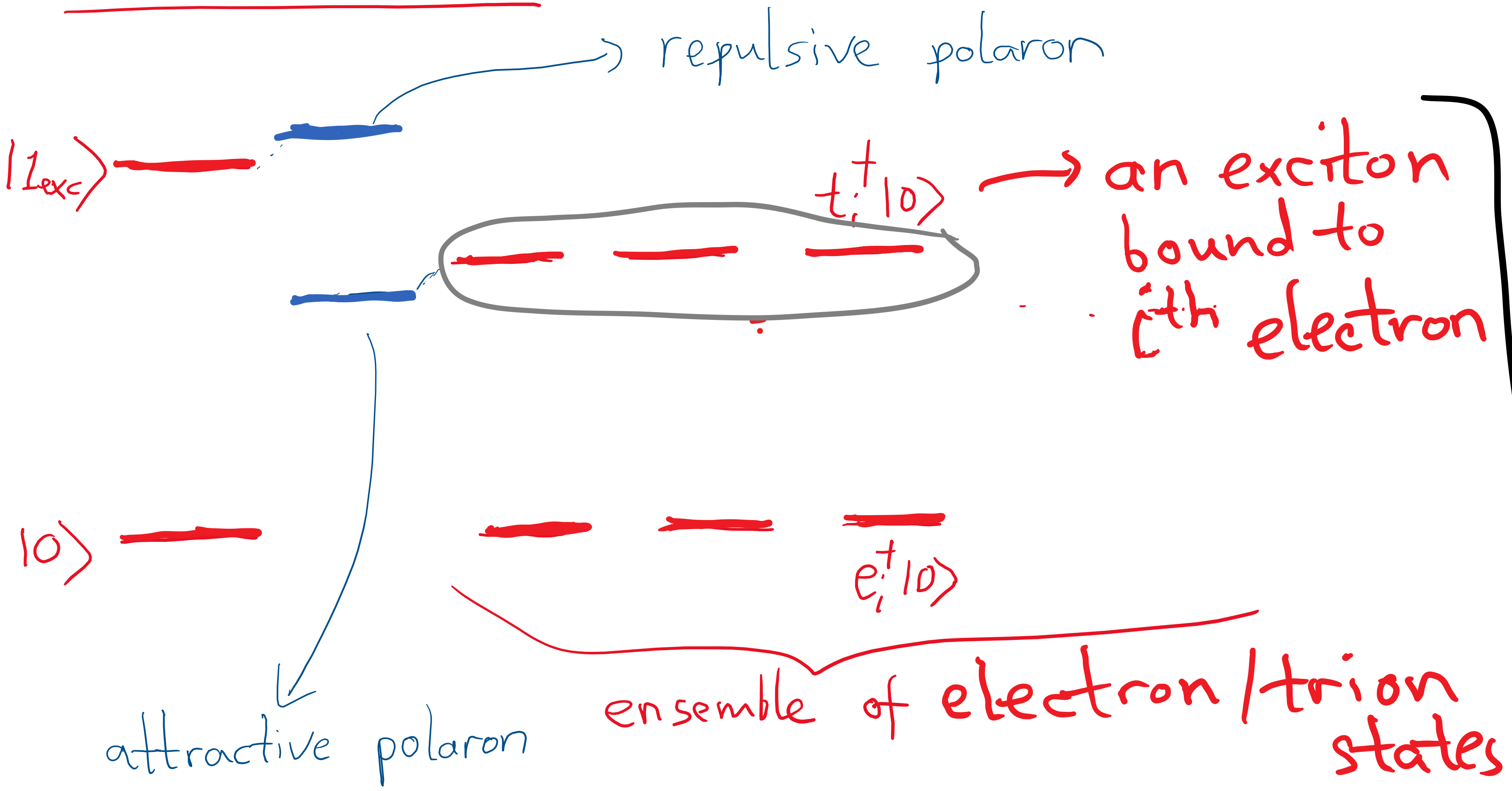
dressed state:

$$|\psi\rangle = \Big[\frac{g_c \sqrt{N}}{\Delta} a_c^\dagger + \overbrace{\frac{1}{\sqrt{N}} \sum_{i=1}^{N} \sigma_{eg}^{i}}^{|B\rangle} \Big] |G\rangle$$

ϕ_0 - quasi-particle weight

cavity only couples to the bright state

Polaron model (limit of localized electrons)



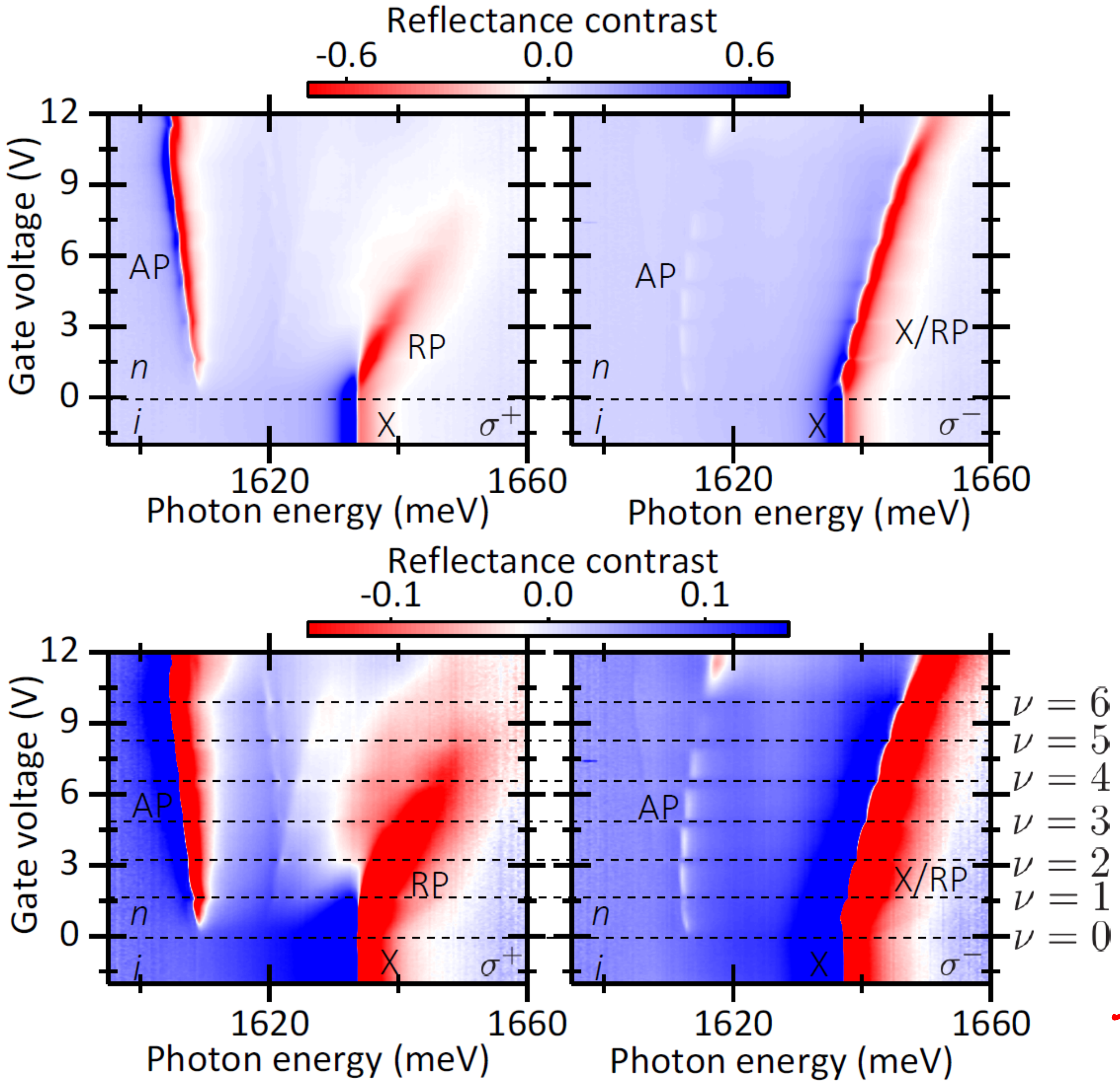
- Polaron formation does not require mobile electrons.
- Attractive polaron energy

$$\boxed{E_{AP} = E_T - \frac{m_e}{m_r} E_F}$$

⇒ Polaron is a superposition of collective (trion) excitation of electrons <u>and</u> exciton.

* Analogy valid: Chevy limit of attrac. polaron

Exciton-polarons at B=14 T: optical signature of IQH states



A filled Landau level shows up as a cusp in RP/exciton energy and a reduction in its linewidth

Energy scales:

$$E_c = \frac{e^2}{\epsilon \ell_B} \sim 30 \text{ meV}$$

$$\omega_c = \frac{eB}{m} \sim 2.2 \text{ meV}$$

→ Shubnikov de Haas oscillations in optical conductivity: precise determination of electron density

Brief summary

- TMD monolayers are outstanding optical materials at low T and when encapsulated by hBN.

- For low electrons densities ($\leq 1x\ 10^{12}\ cm^{-2}$), exciton-electron interactions can be described by treating excitons as robust impurities (i.e. Bohr radius remains almost unchanged).