

Quantum confined excitons in two-dimensional materials

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Co-workers:

Ankur Thureja, Puneet Murty

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Many discussions with:

Rafal Odlziejewski, Richard Schmidt

Eugene Demler

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Congratulations Eugene!

Outline

- Elementary optical excitations of transition metal dichalcogenide (TMD) monolayers
- Electrical control of optical properties: realization of quantum confined excitons in a monolayer p-i-n junction
- Prospects for strongly interacting excitons and polaritons

Materials: Transition metal dichalcogenides (TMD)

- layered 2D semiconductors

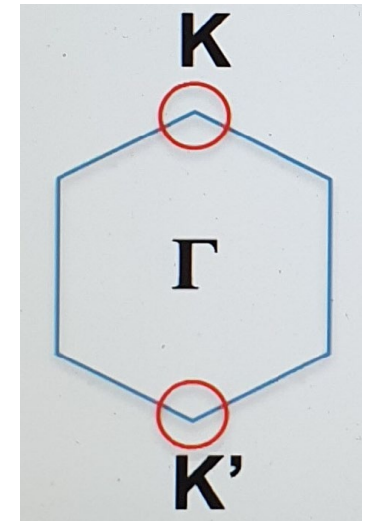
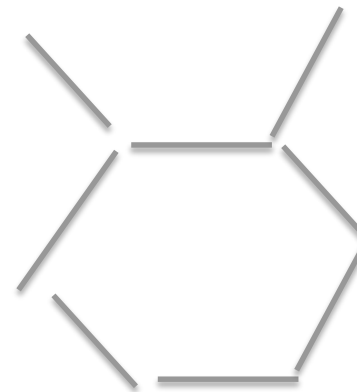
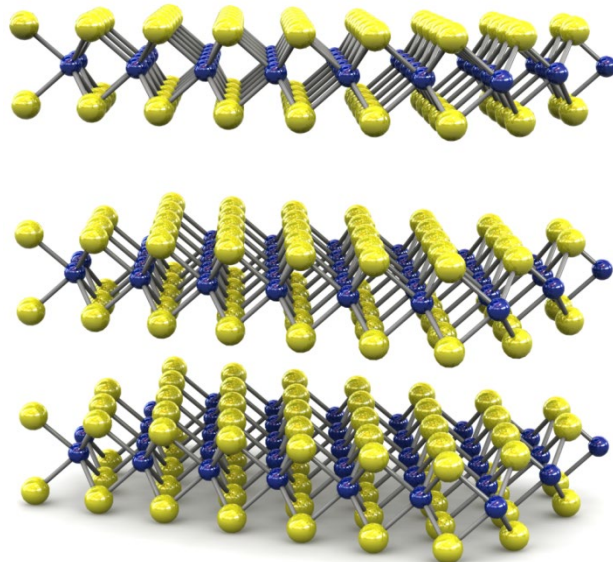
Formula: MX_2

M = Transition Metal
X = Chalcogen

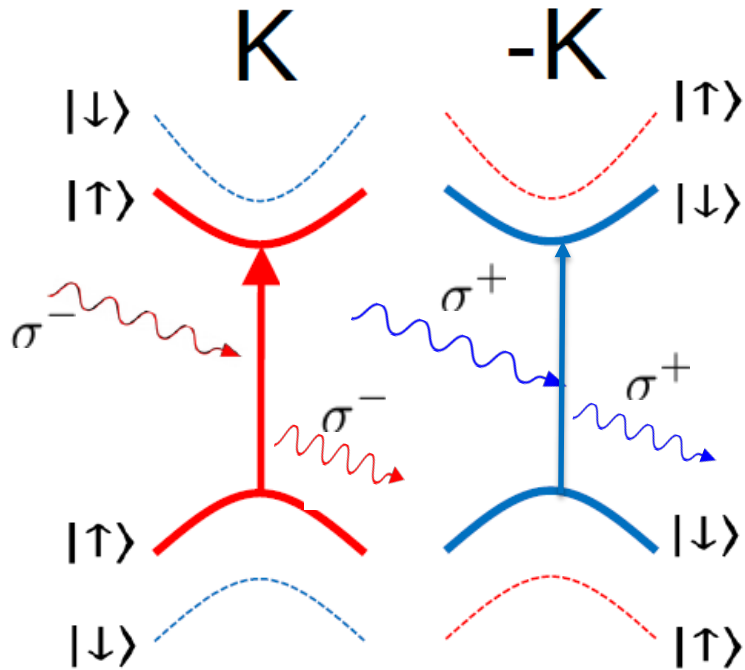
H																	He
Li	Be	MX_2 M = Transition metal X = Chalcogen										B	C	N	O	F	Ne
Na	Mg	3	4	5	6	7	8	9	10	11	12	Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac-Lr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

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



TMD monolayers: direct band-gap semiconductors with a valley degree of freedom

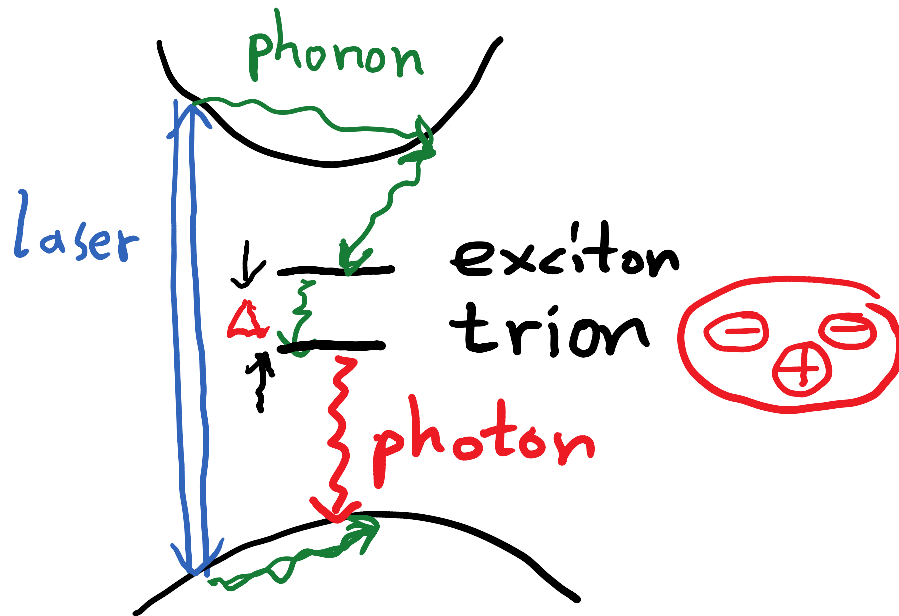


TMD monolayers: direct band-gap semiconductors with
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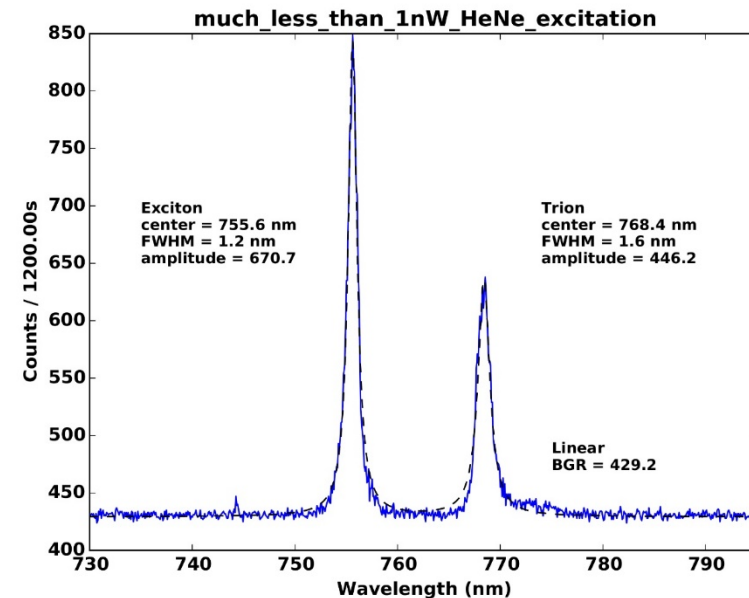
For MoSe_2 in hBN, $E_B = 230$ meV, $a_B = 1.1$ nm

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 very low density electrons (trion osc. str. is much smaller).

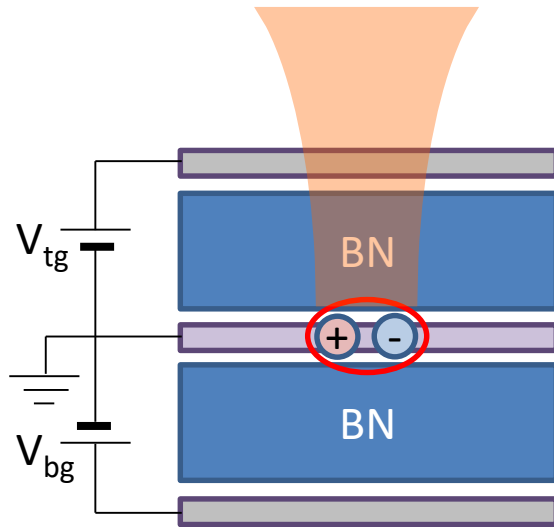


Exciton linewidth of neutral MoSe_2 is comparable to the radiative decay rate

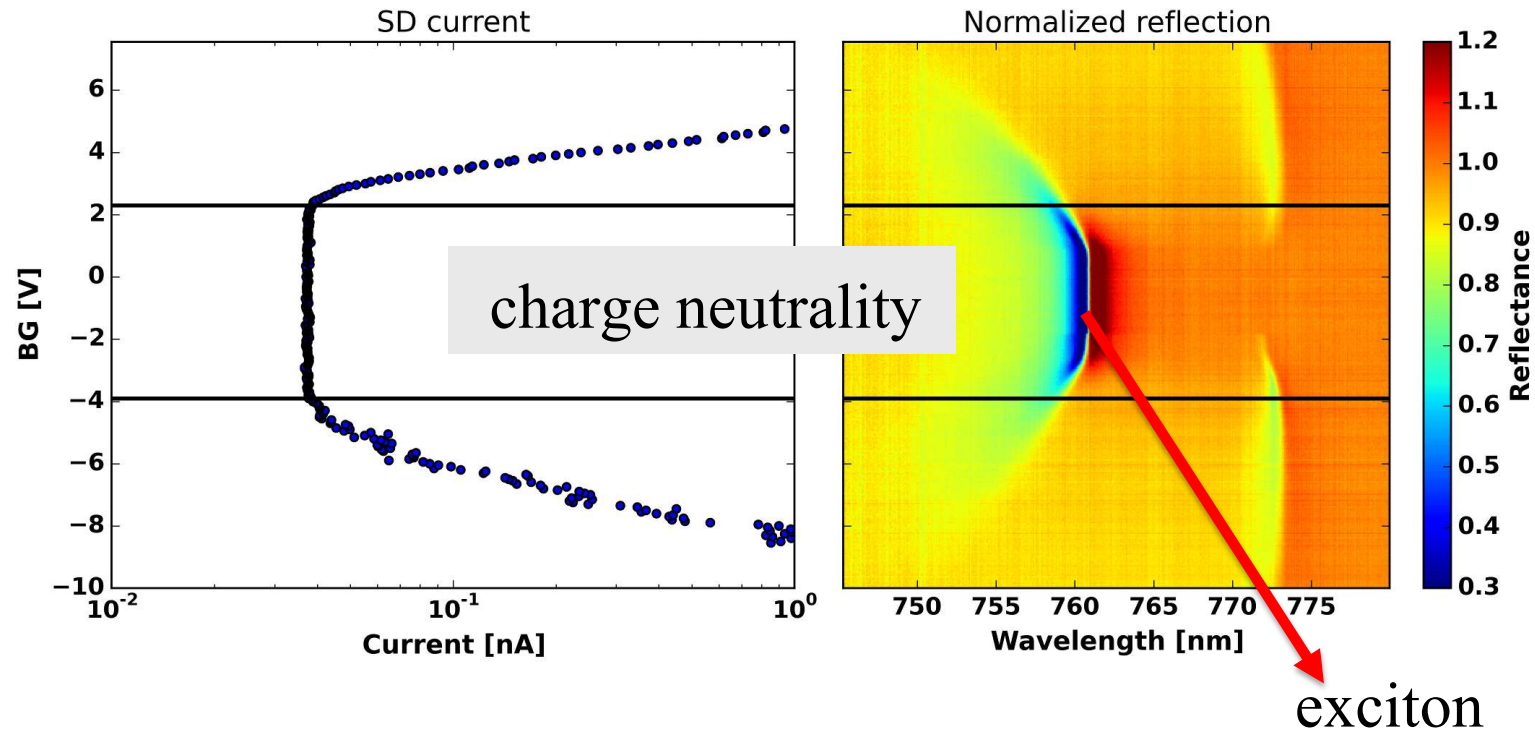


Charge tunable van der Waals heterostructures

- 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



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 – **attractive polaron** - if electrons/holes are present

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 as a mobile impurity in a degenerate electron system

Interacting exciton-electron system (with Eugene)

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

Contact interaction strength determined
by the trion binding energy

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

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Approximate form of eigenstates of H in the truncated Hilbert space:

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

bare exciton

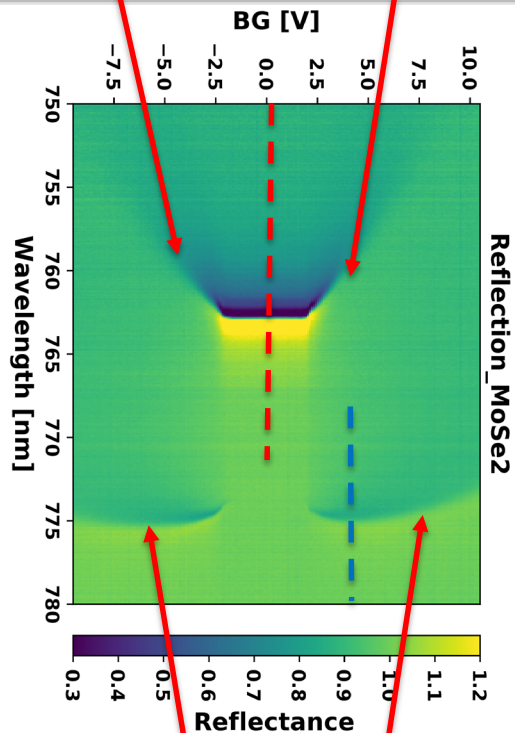
Exciton + Fermi-sea electron-hole pair

Eigenstates with a large spectral weight:

attractive (AP) and repulsive (RP) polarons

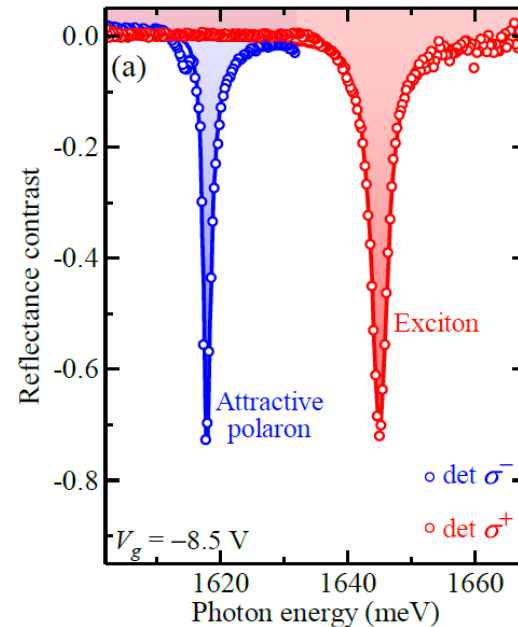
Exciton-(Fermi)-polaron: exciton dynamically screened by itinerant electrons

Repulsive polaron (RP) branch



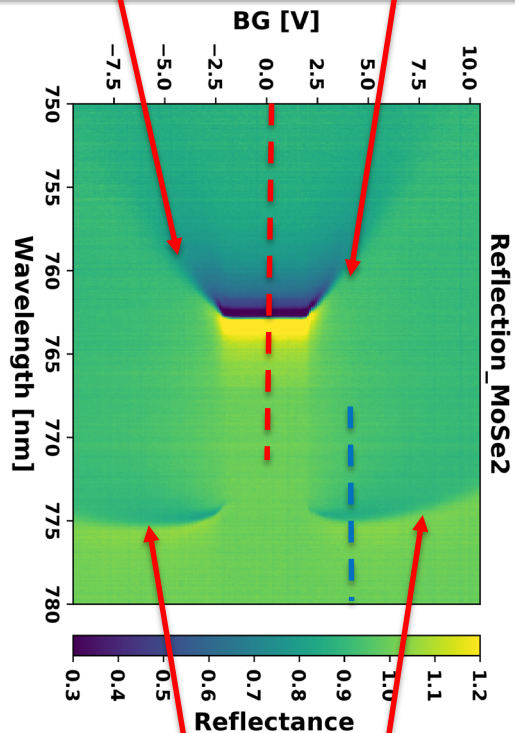
Attractive polaron (AP) branch

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



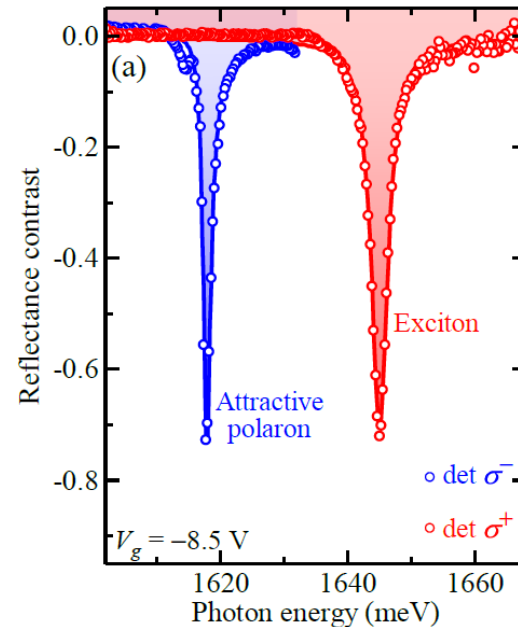
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Note the strong electron (hole) density dependence of repulsive polarons:
Exciton as a «quantum sensor» of correlated electron states

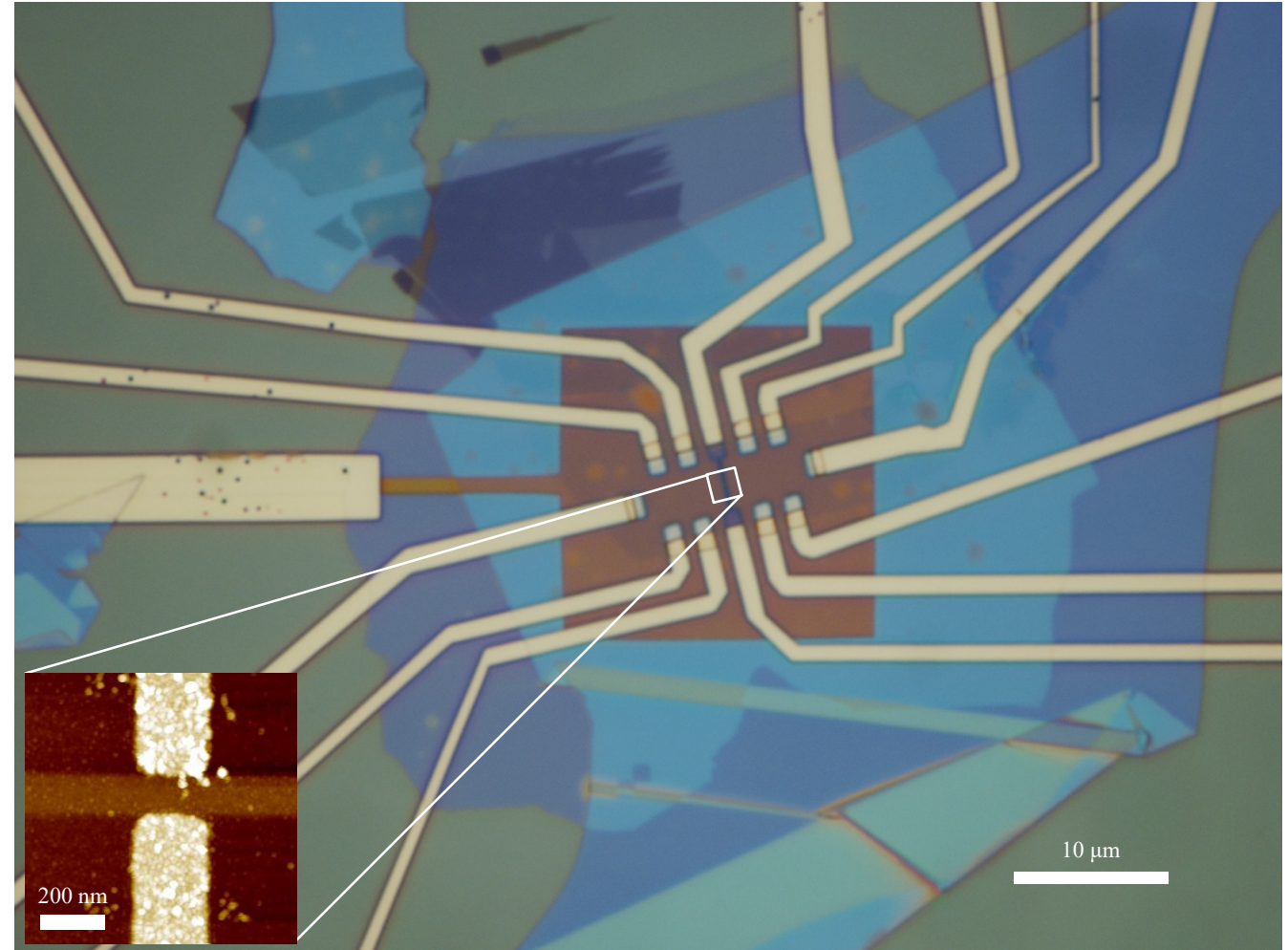
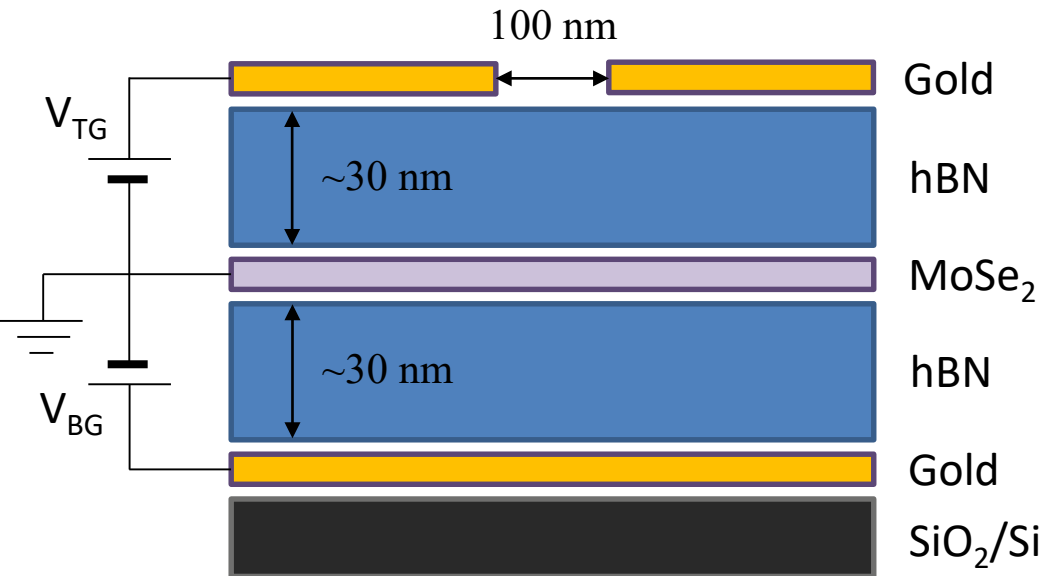
Quantum confinement of neutral excitons using electric fields

Device structure

➤ Stack:

Au back gate/ hBN / 1L

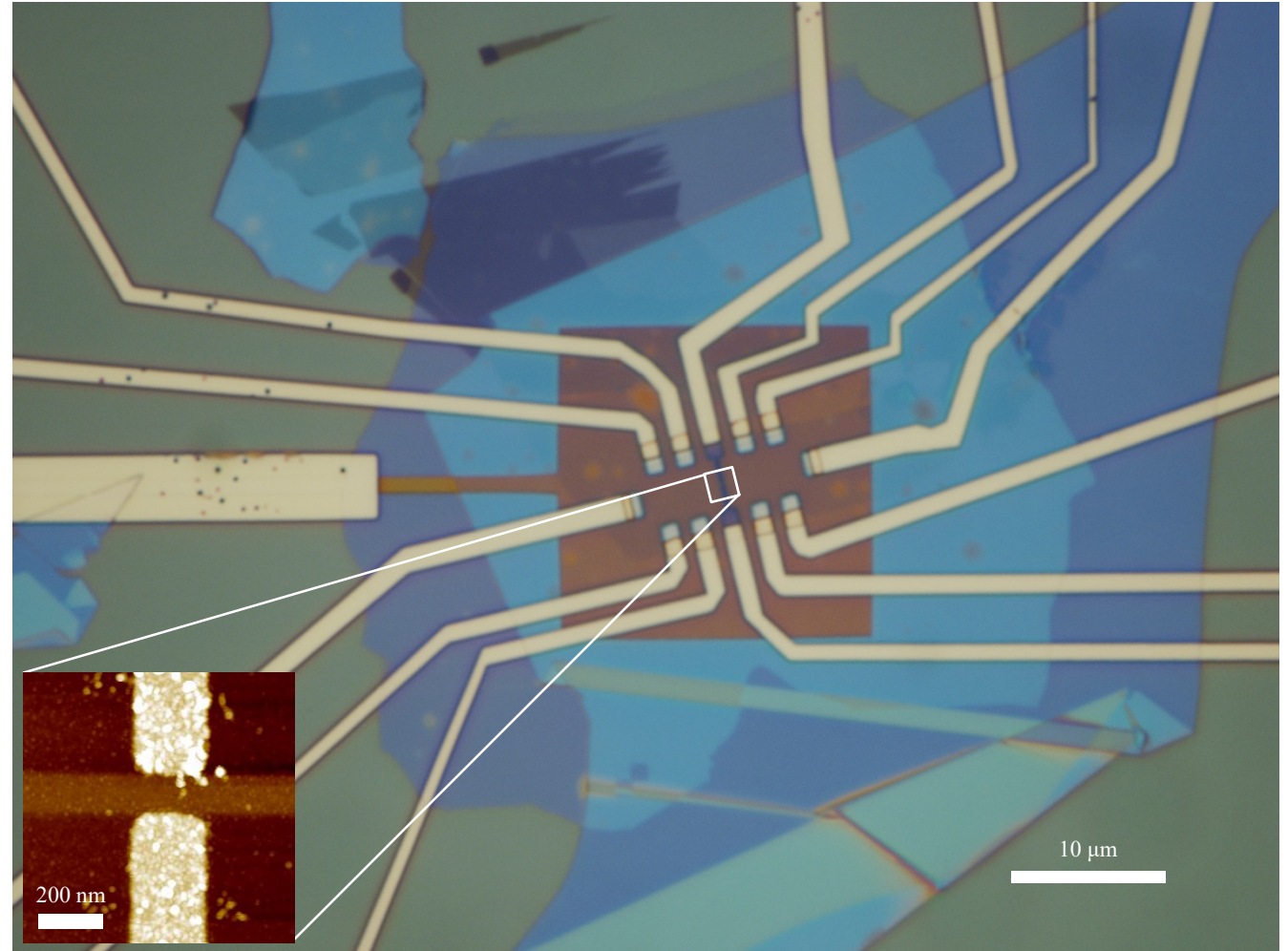
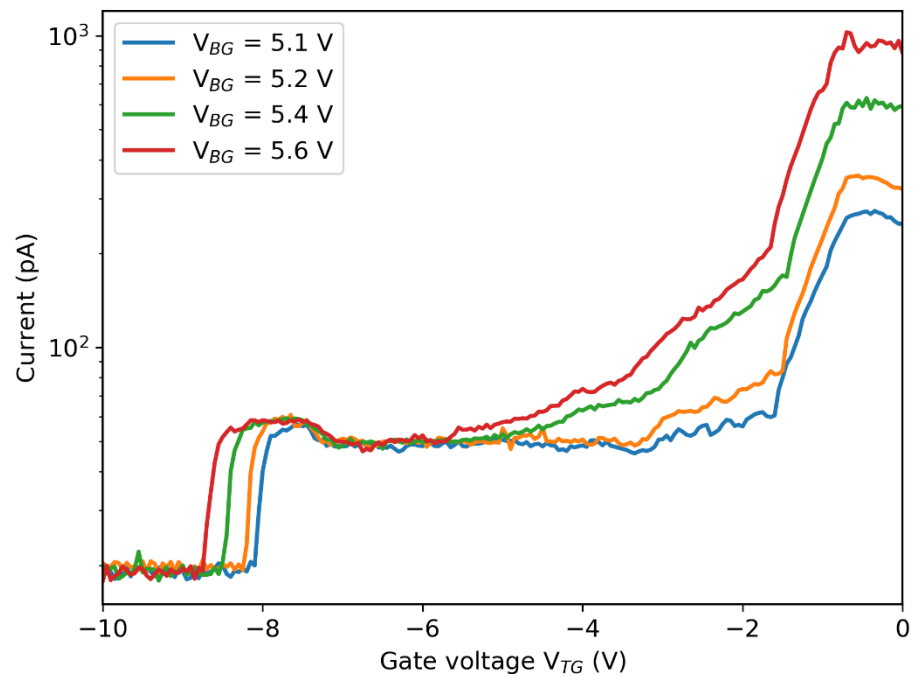
MoSe₂ / hBN / Au split gate



3nm Ti / 7 nm Au split gate ➔ Optically transparent

Quantum confinement of neutral excitons using electric fields

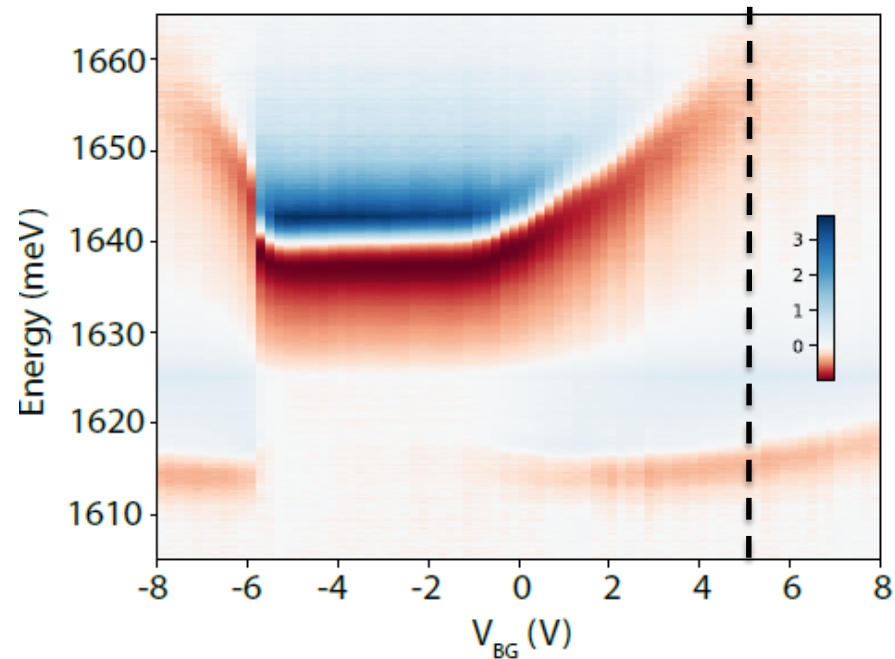
Quantized 2P-current
measurement $T = 4\text{ K}$, $B = 7\text{ T}$



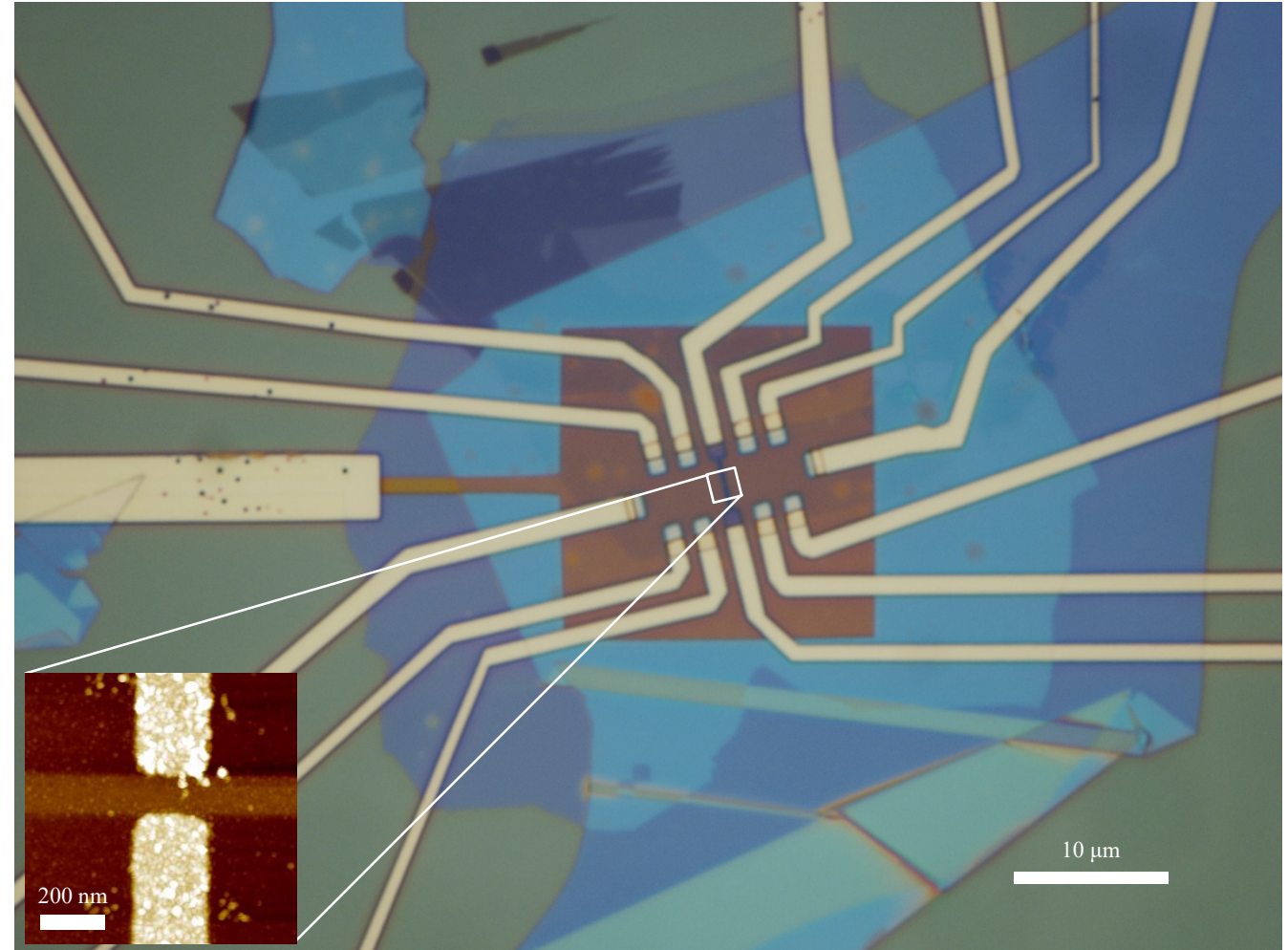
3nm Ti / 7 nm Au split gate \rightarrow Optically transparent

Quantum confinement of neutral excitons using electric fields

Set top gate to 0 V

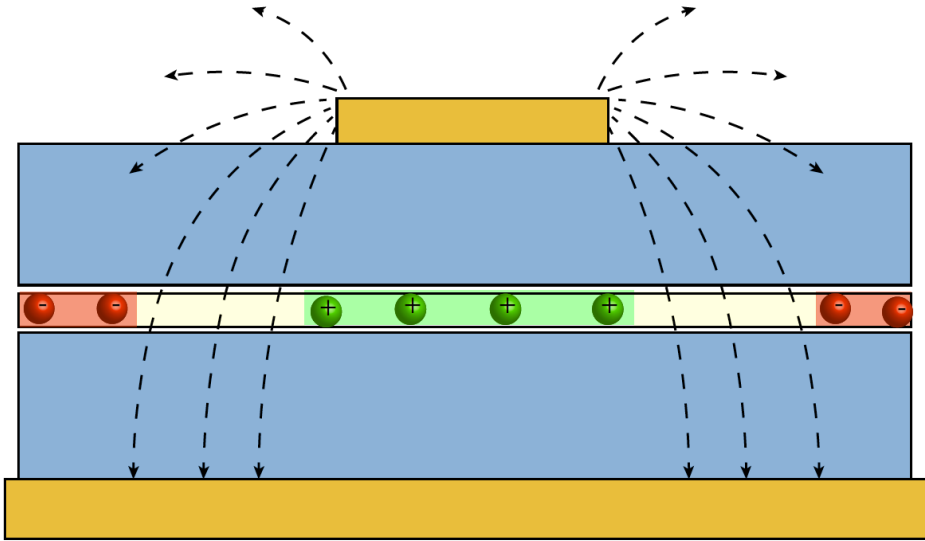


For $V_{BG} = 5$ V (black dashed line),
the sample is electron doped

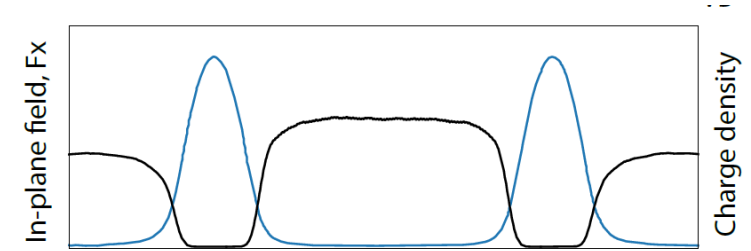


3nm Ti / 7 nm Au split gate ➔ Optically
transparent

Reflection spectrum– Top gate dependence for $V_{BG} = 5V$



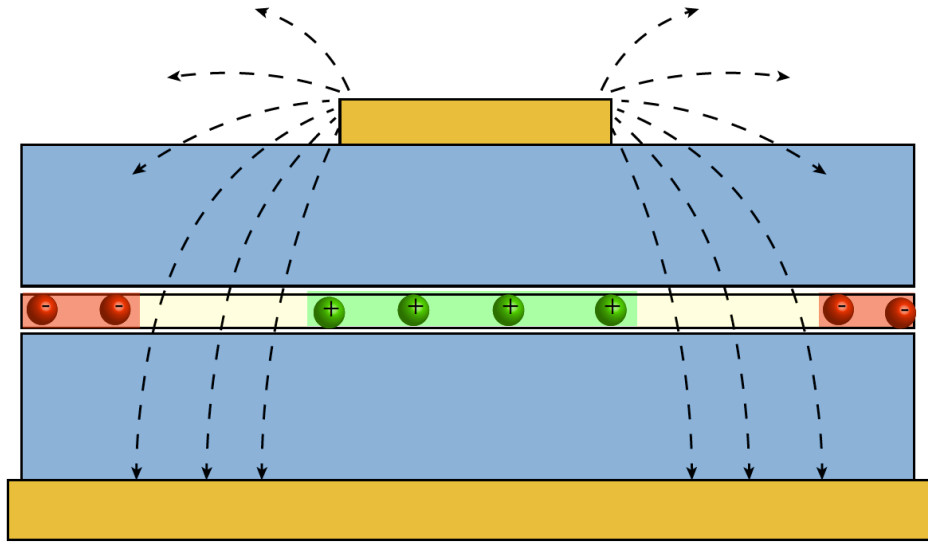
As we reduce V_{TG} we can hole dope under the top gate; there is large electric field in the i-region that separates p- and n-doped regions



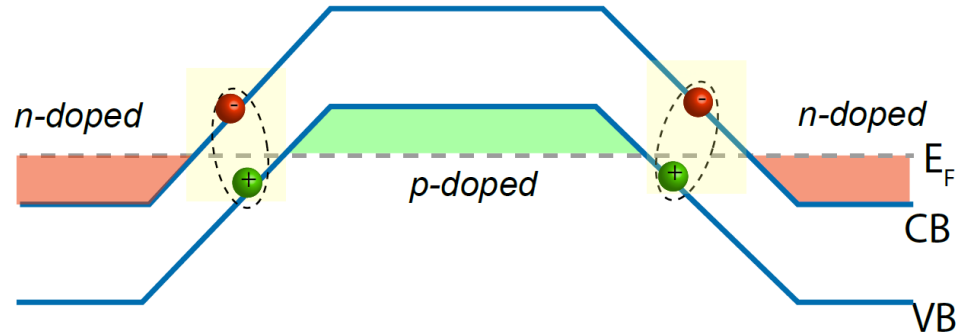
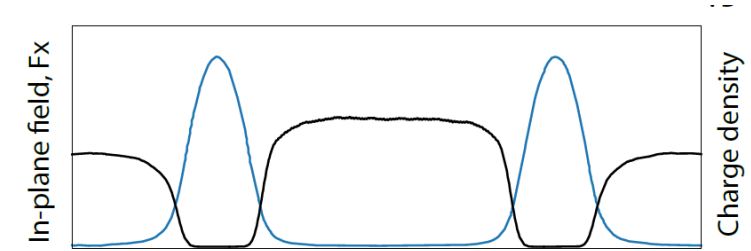
$V_{BG} > 0$: overall electron doping

$V_{TG} + V_{BG} < 0$: hole doping under the top gate

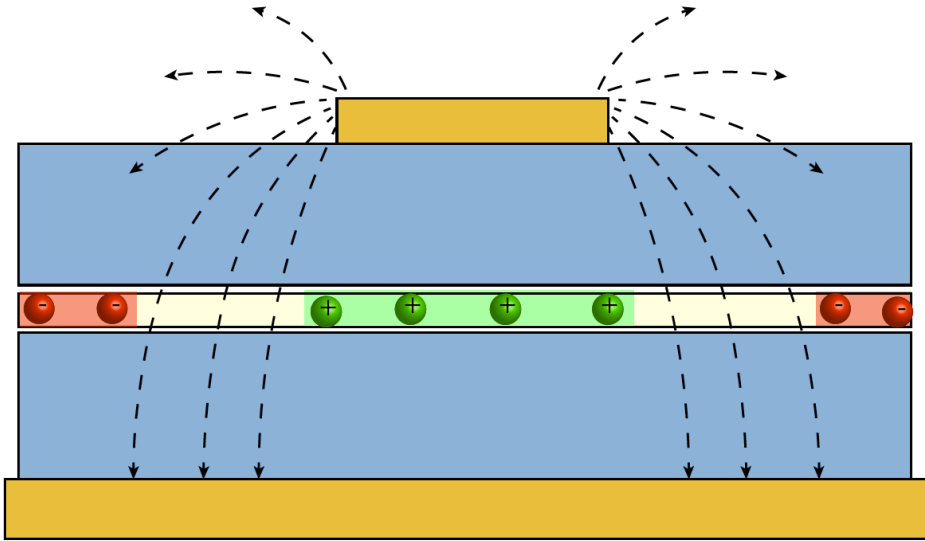
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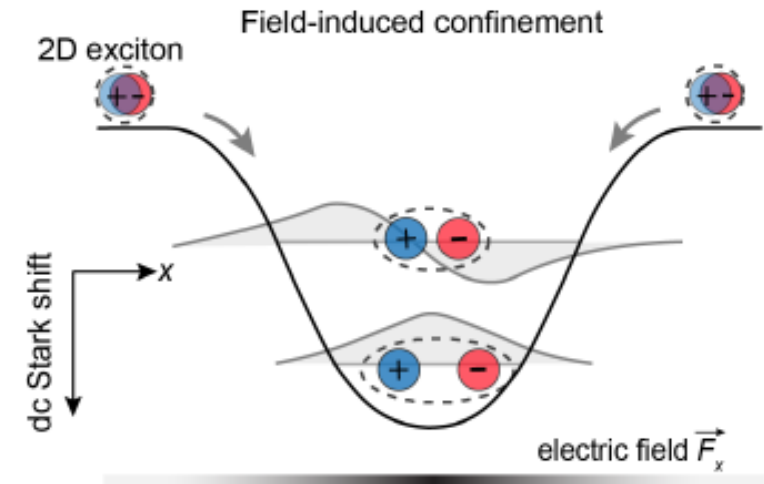
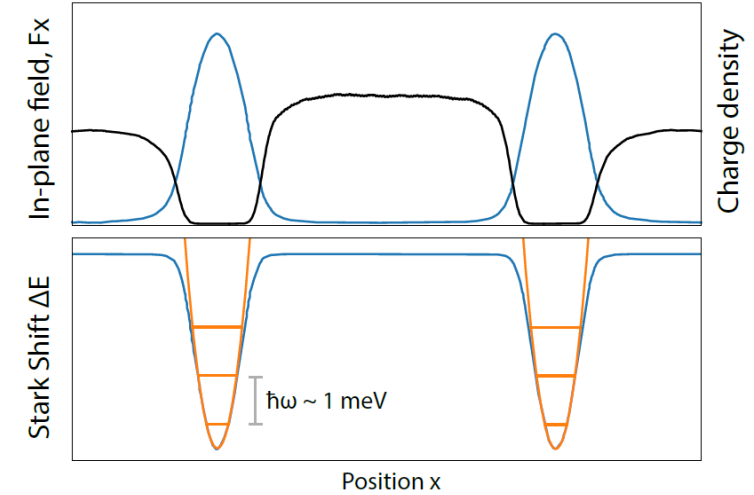


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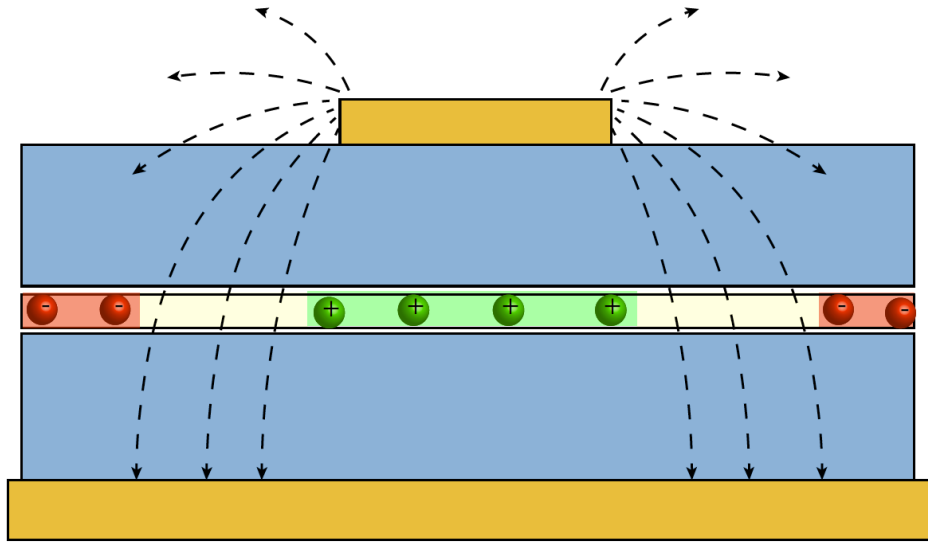


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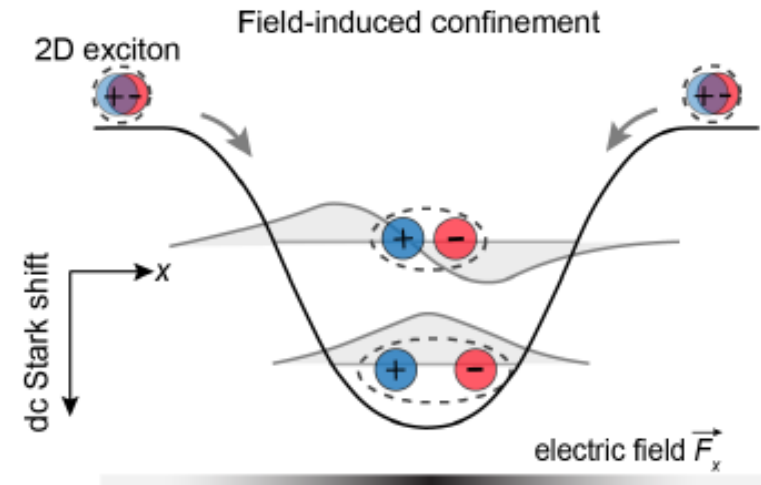
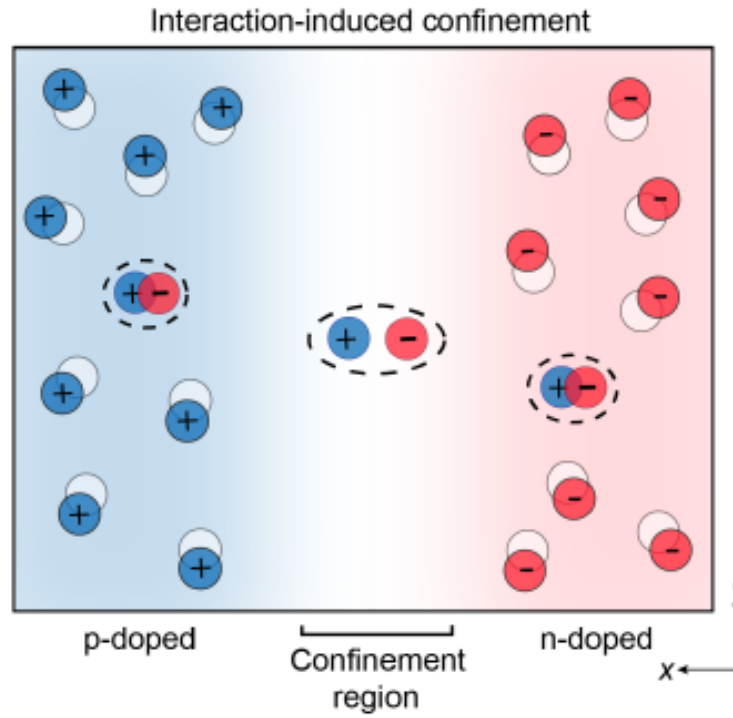
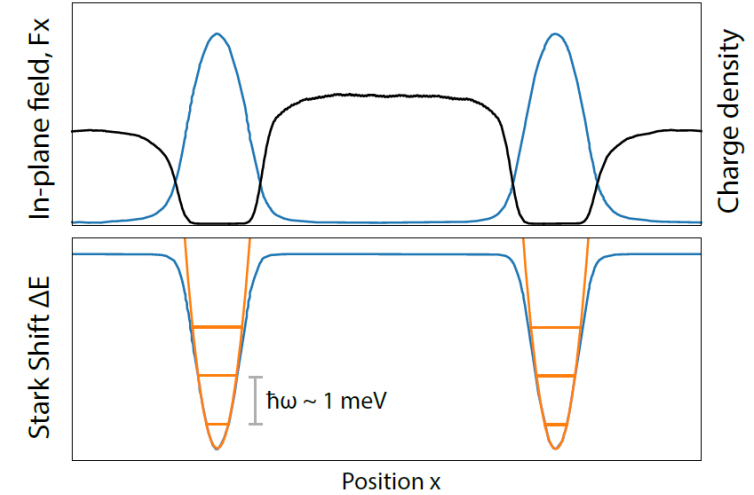
$$\Delta E_S = -\frac{1}{2} \alpha F_x(x)^2$$



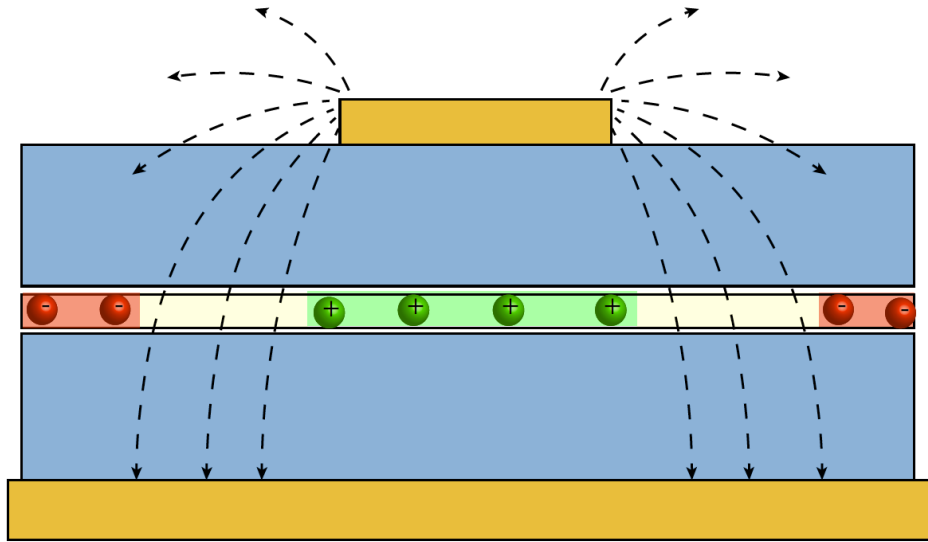
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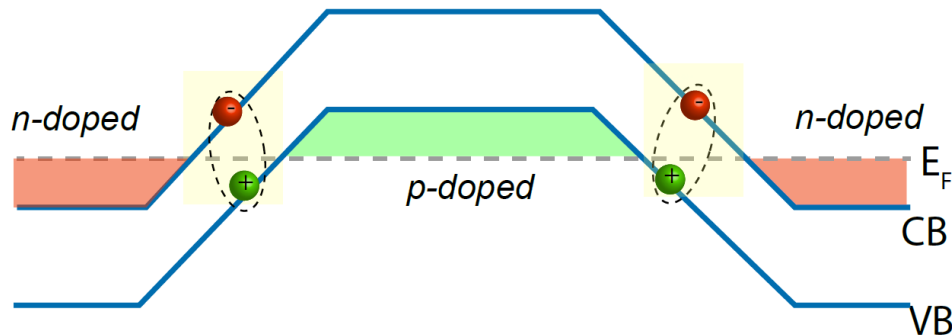
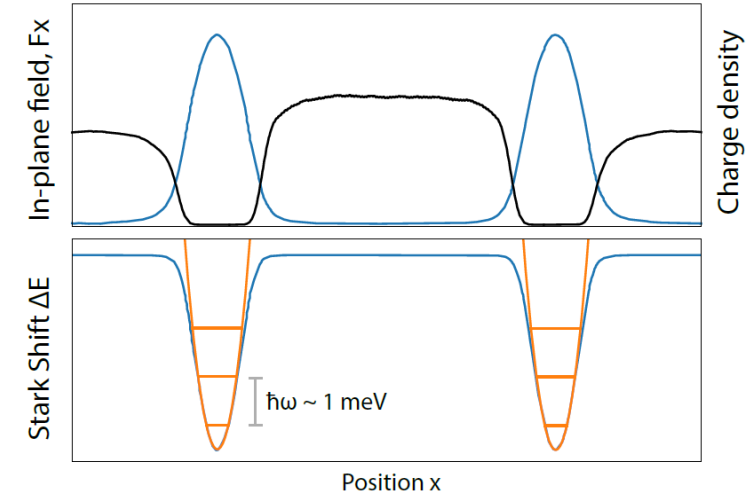
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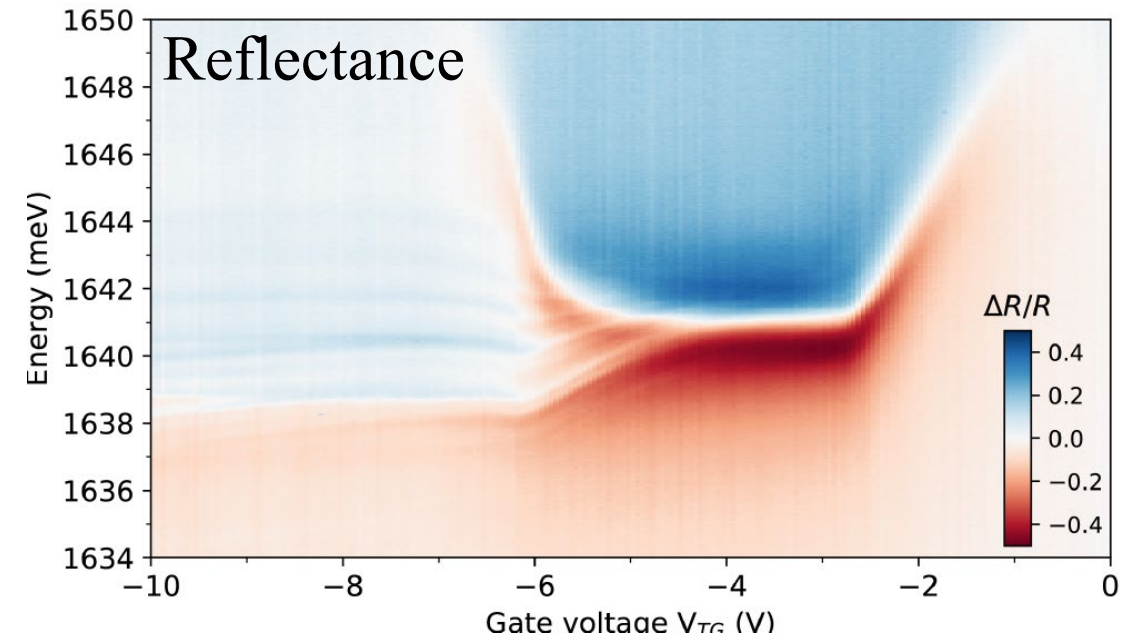
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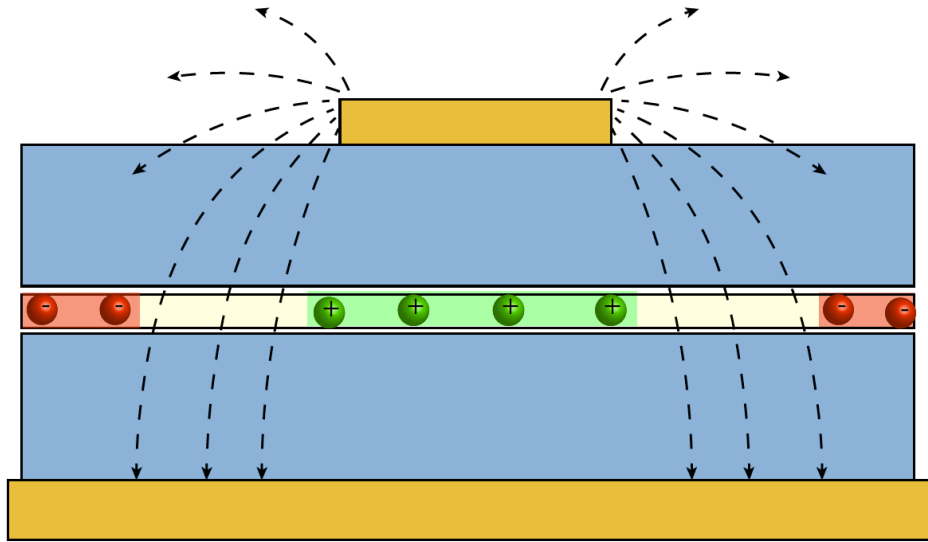
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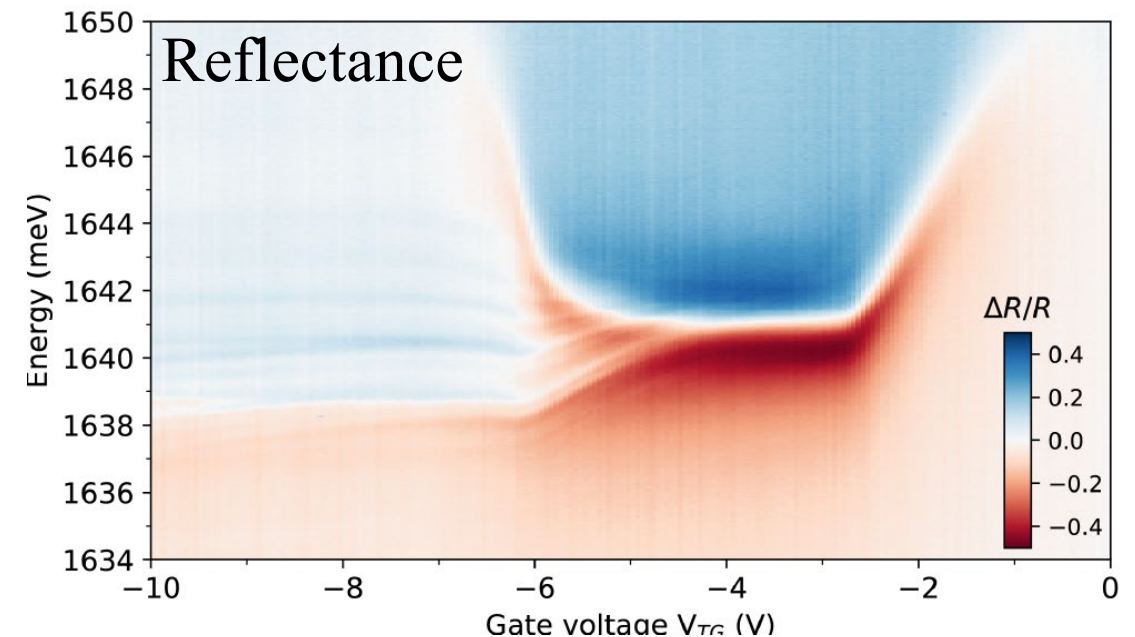
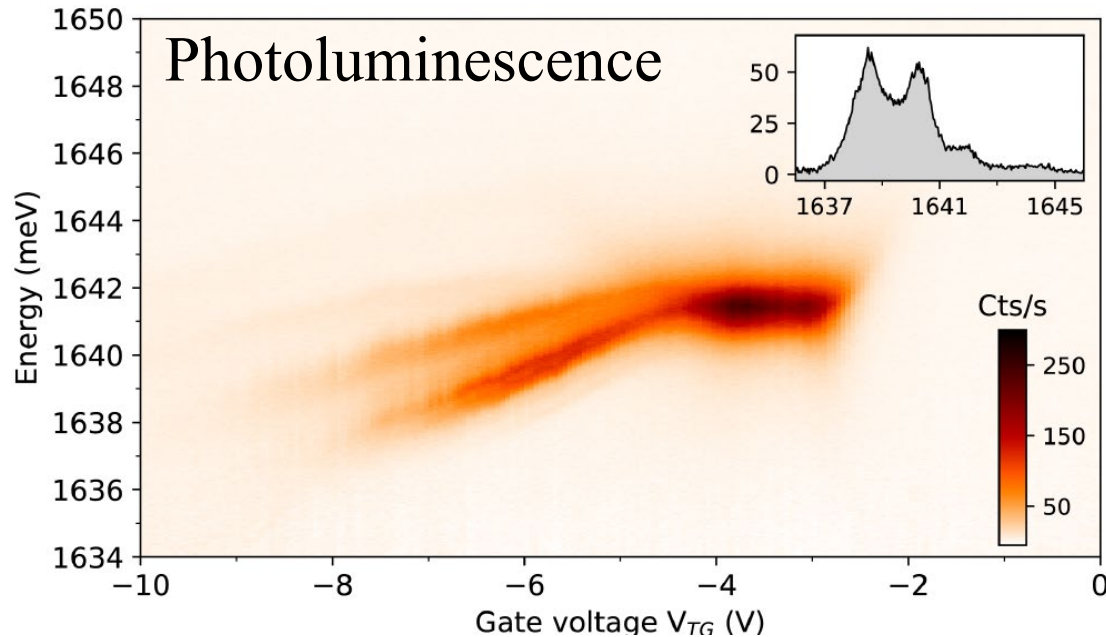
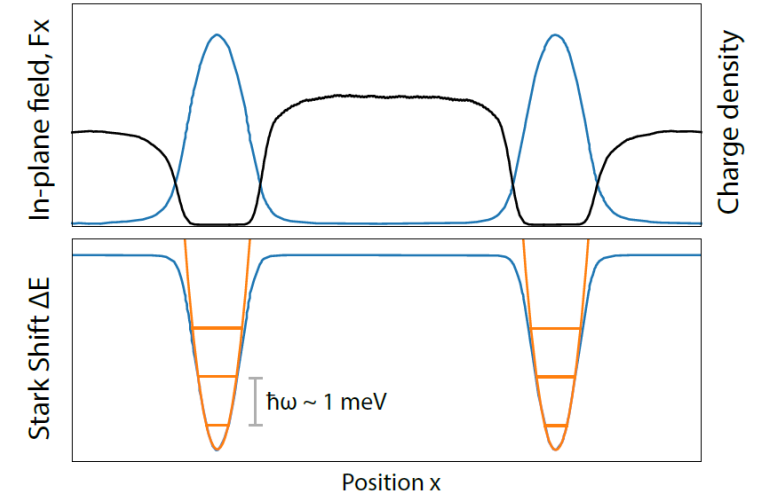
Neutral excitons are confined in the i-region by dc-Stark effect and repulsive exciton-carrier interactions: dipolar excitons in a 1D wire



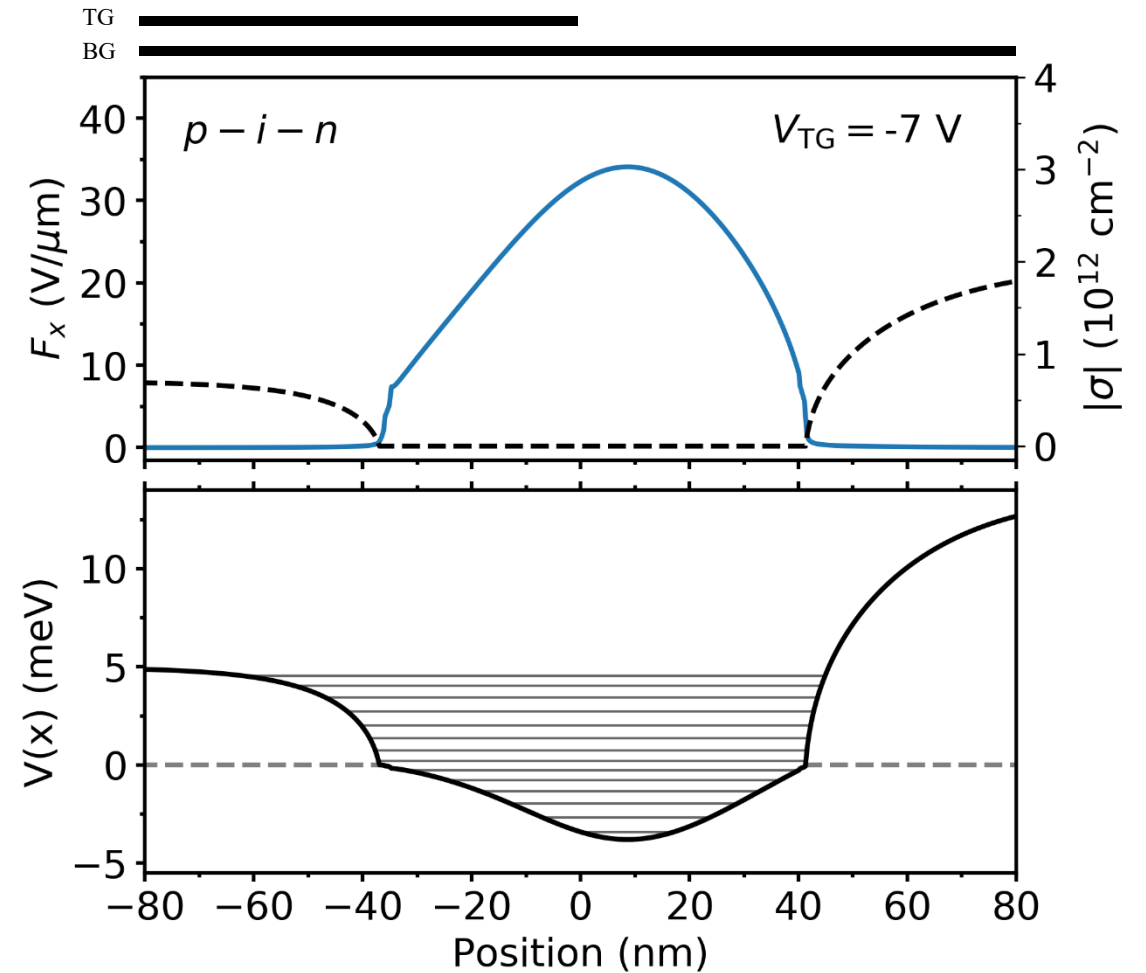
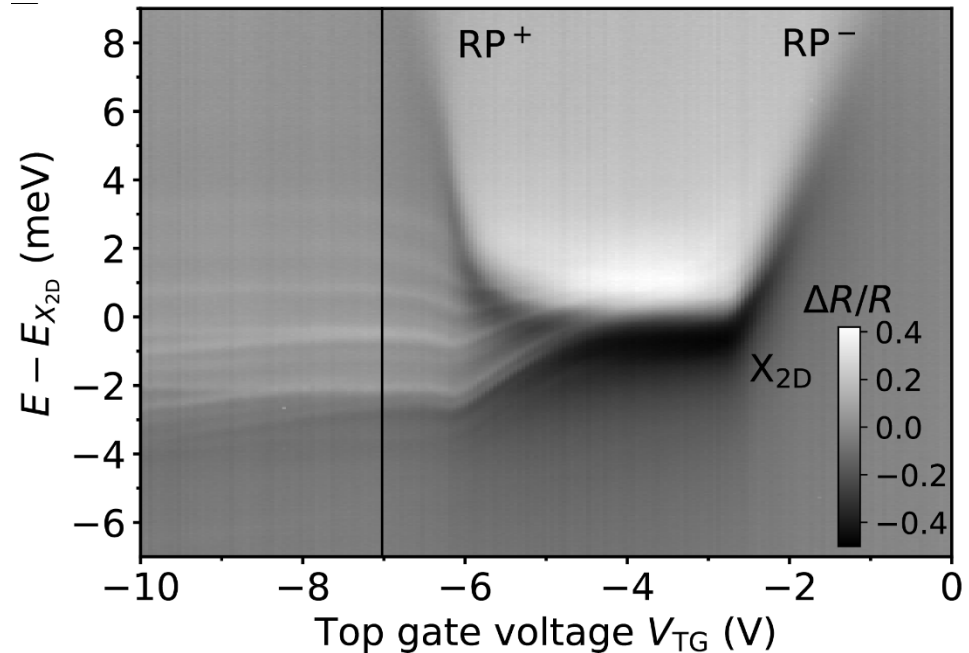
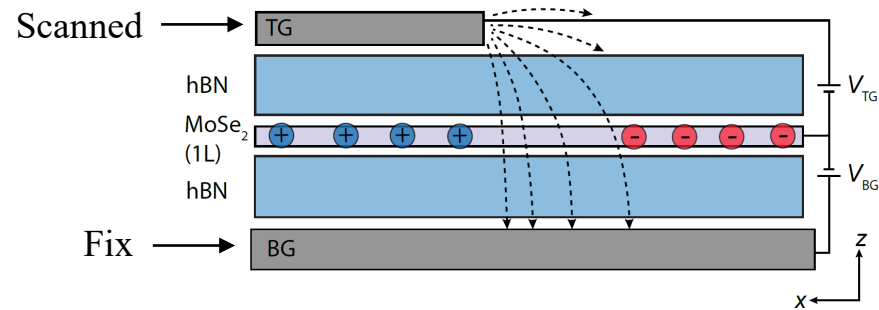
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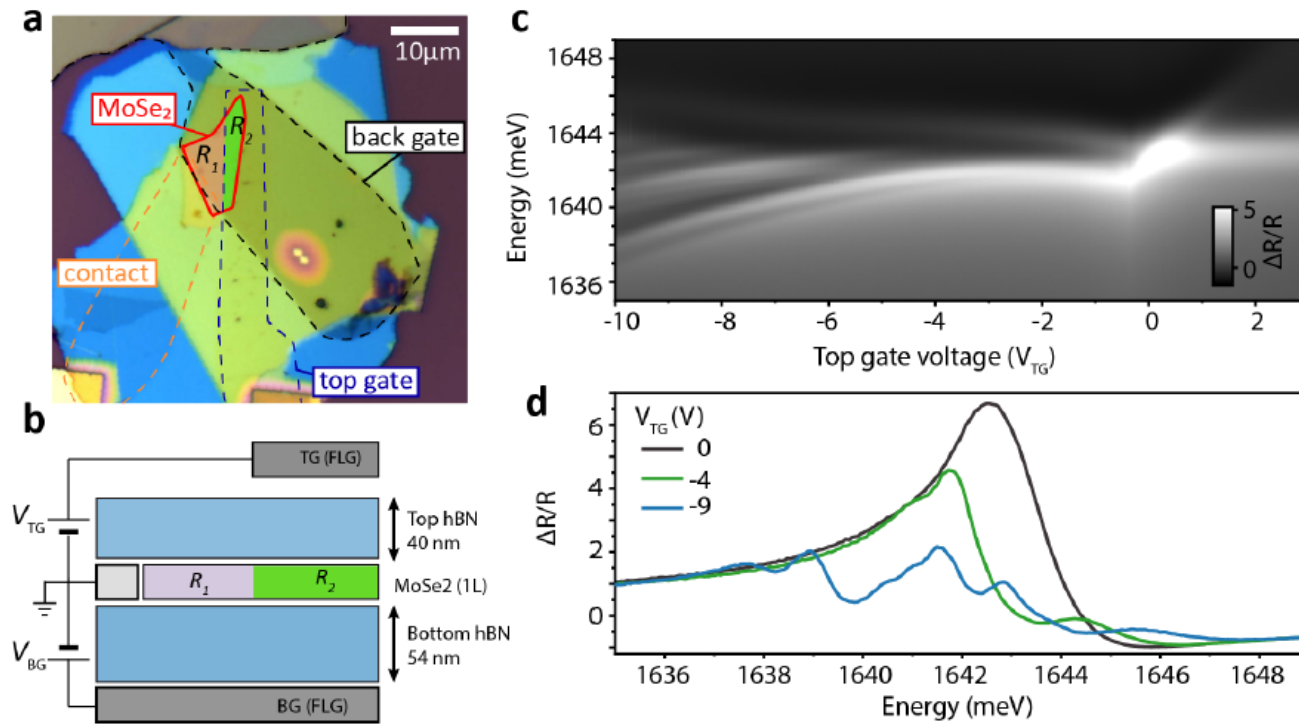


Quantum confined excitons in the p-i-n diode

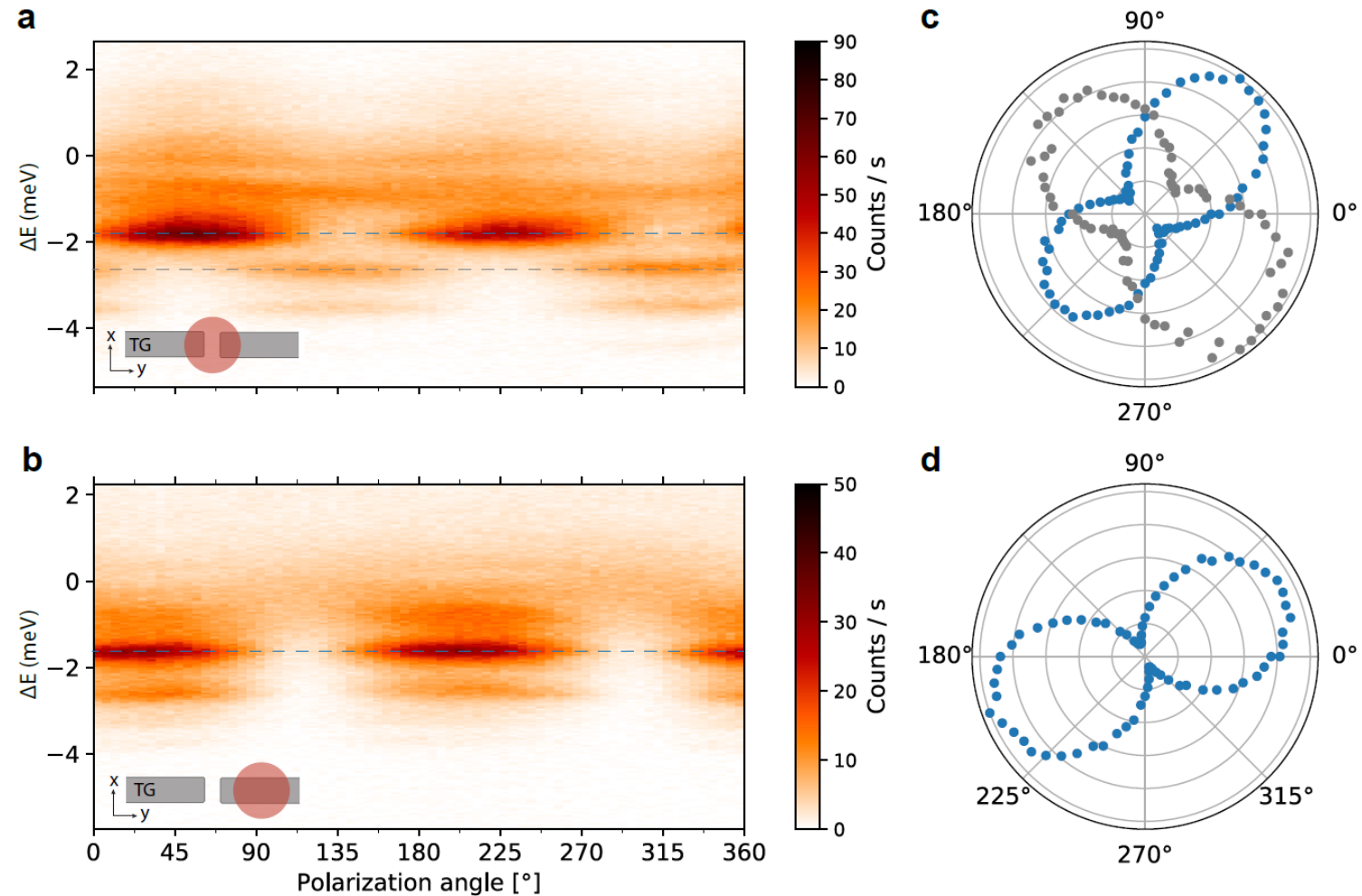


Change in slope of red-shift due to optical charging effects

Quantum confinement of excitons in another device



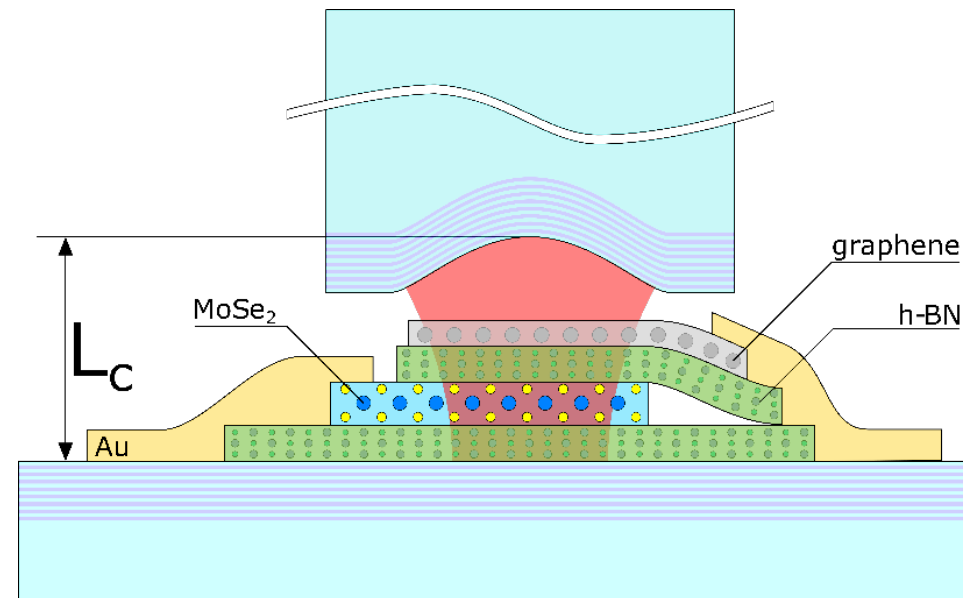
Evidence for 1D confinement: linearly polarized emission



Long-range electron-hole exchange ensures that the exciton emission is polarized along the wire

New directions

- Strongly interacting photons: so far the successful efforts used either 0D emitters (transmons in circuit-QED) or Rydberg excitations from 3D atomic ensembles).
 - a 1D exciton wire in a 0D cavity as a solid-state photonic system with strong polariton interactions in the polariton blockade regime



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- Fully electrically defined and tunable quantum dots in monolayer TMDs.