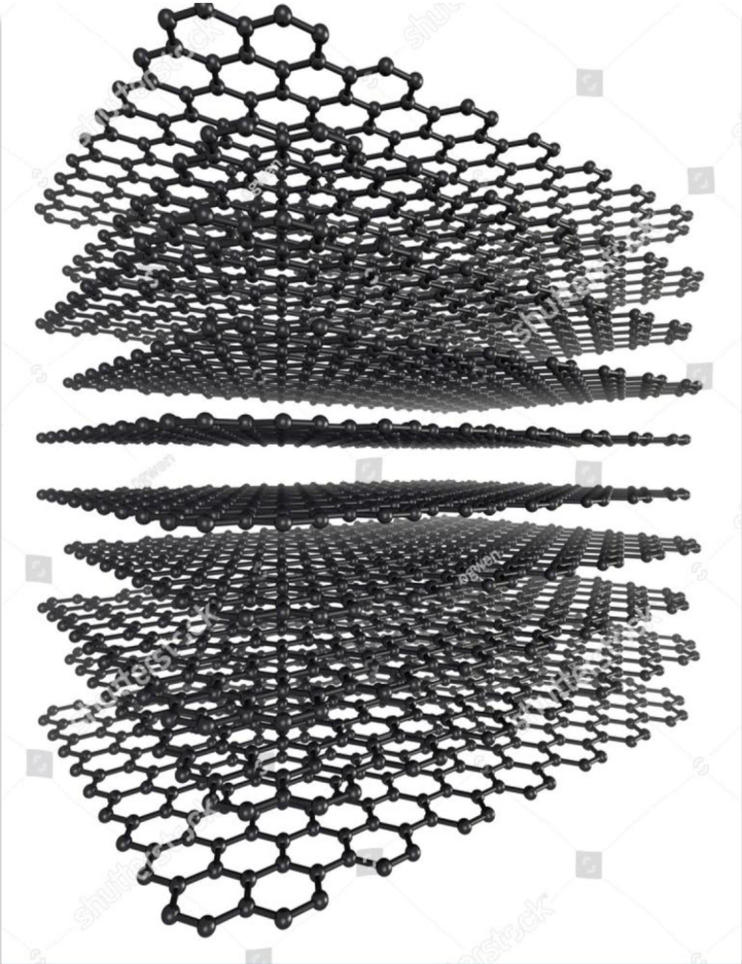
Exciting excitons in layered materials

SARTHAK DAS



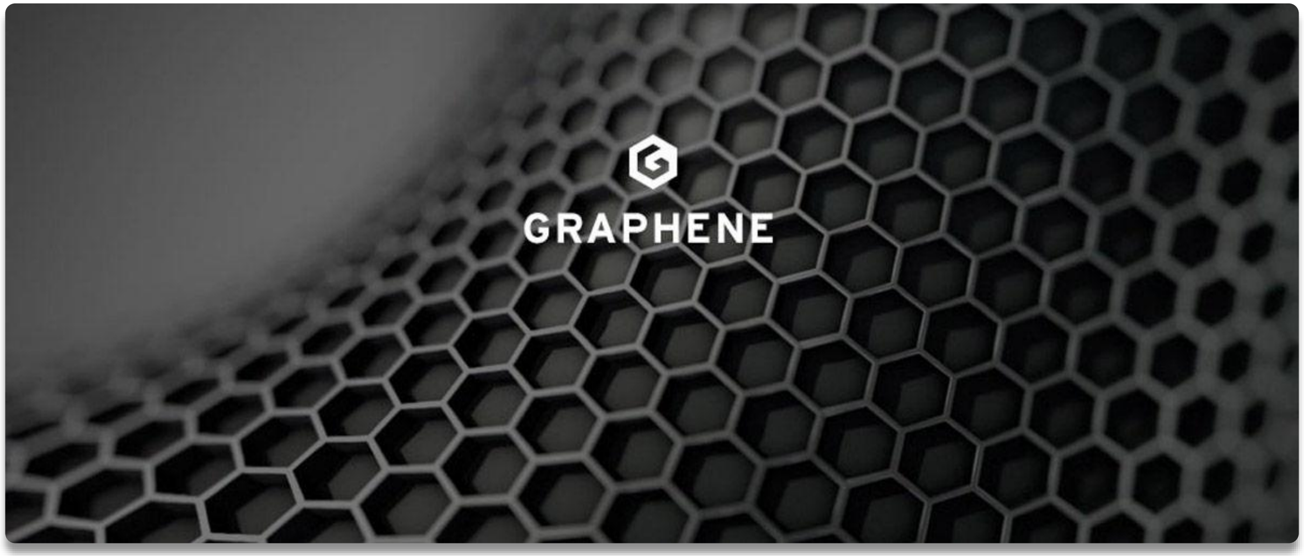
2d materials

GRAPHITE-ALLOTROPE OF CARBON

Ref. Shutterstock

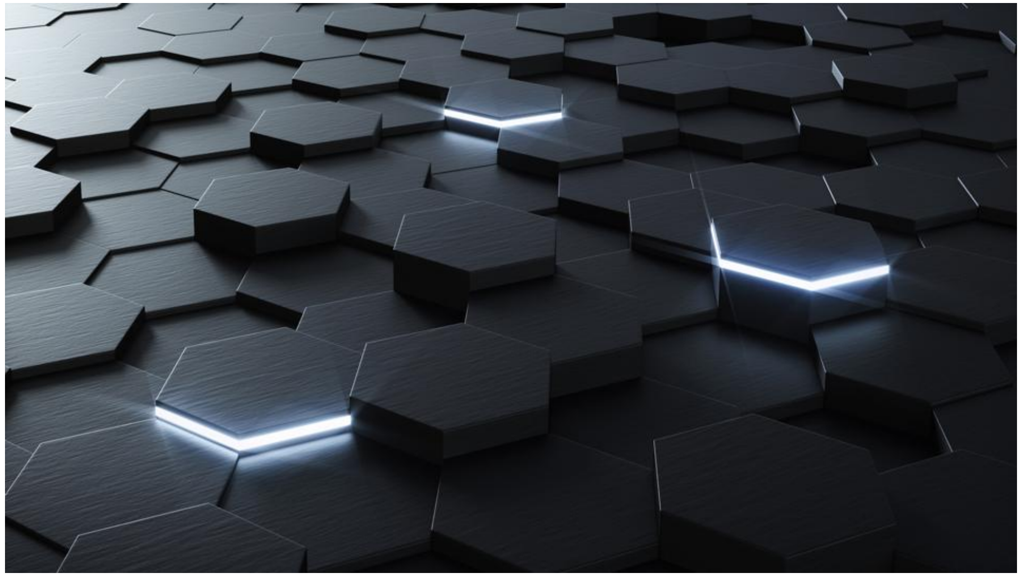
Graphene

- Layered material
- Honeycomb lattice structure
- One atomic thickness 3.4Å
- Unique electrical optical and mechanical properties



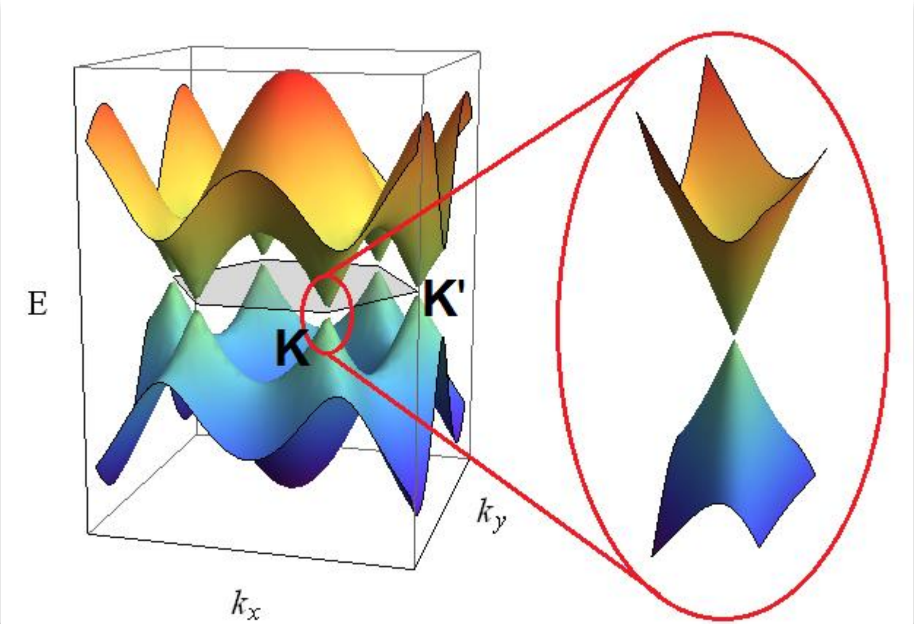
Graphene

- Discovered in 2004
- Lobel prize in 2010



Graphene bandstructure

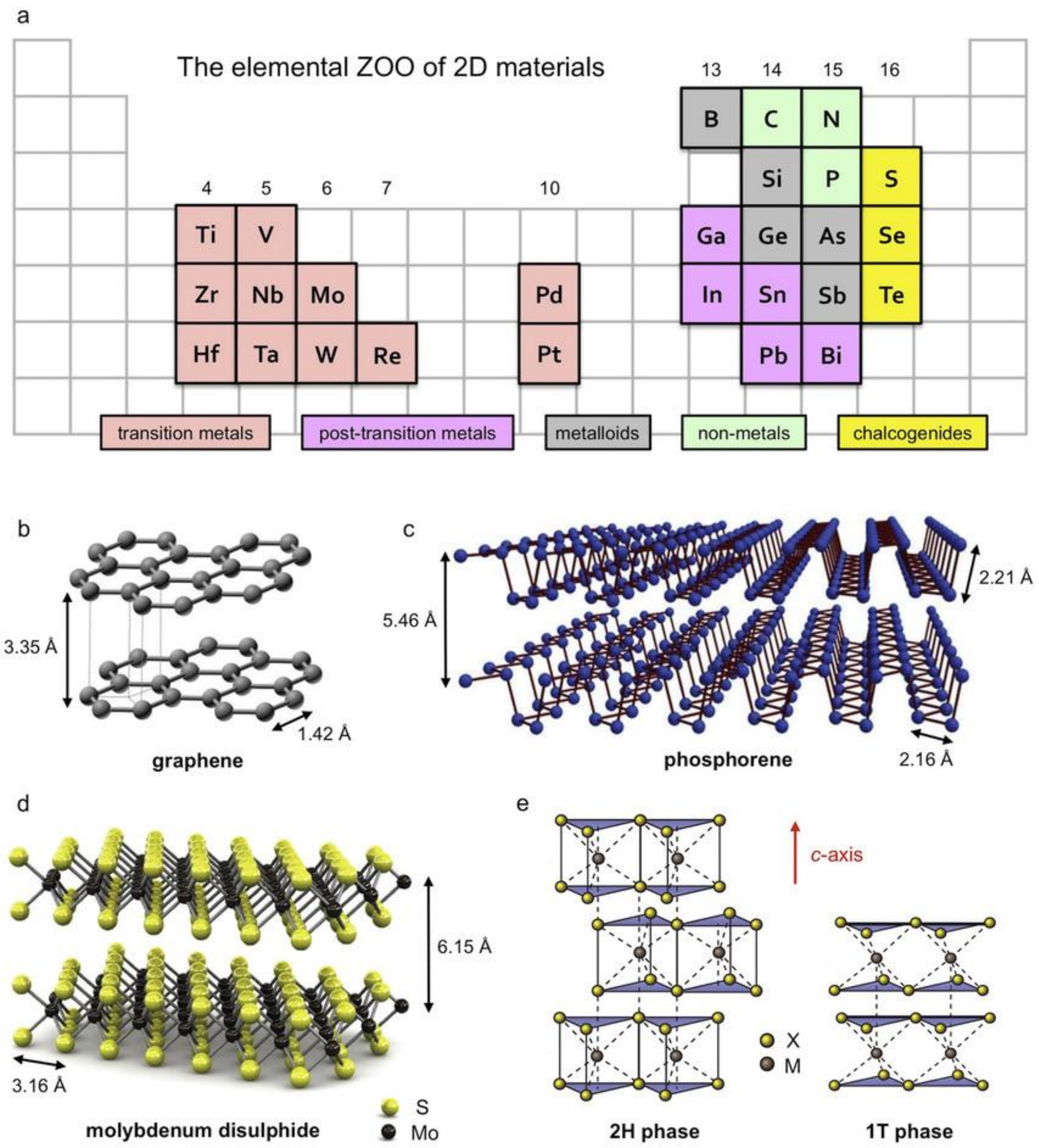
- Hexagonal Brilliouin Zone with three equivalent valleys
- Linear dispersion relation
- Zero band gap material



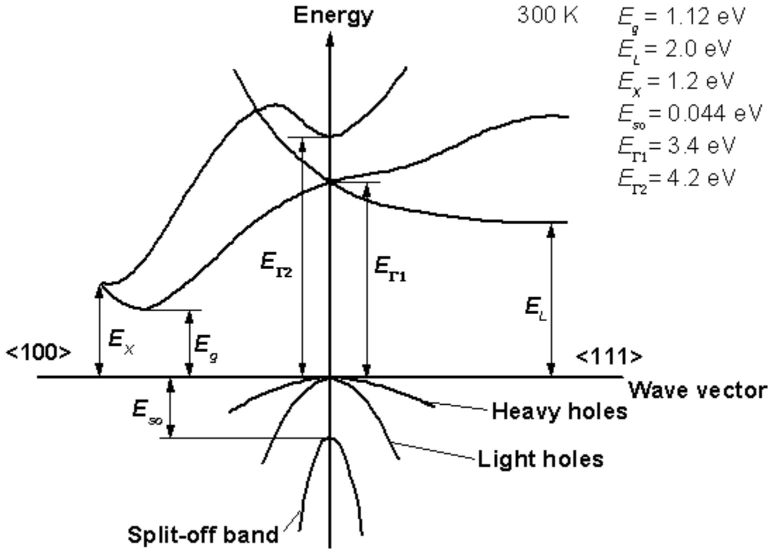
Graphene

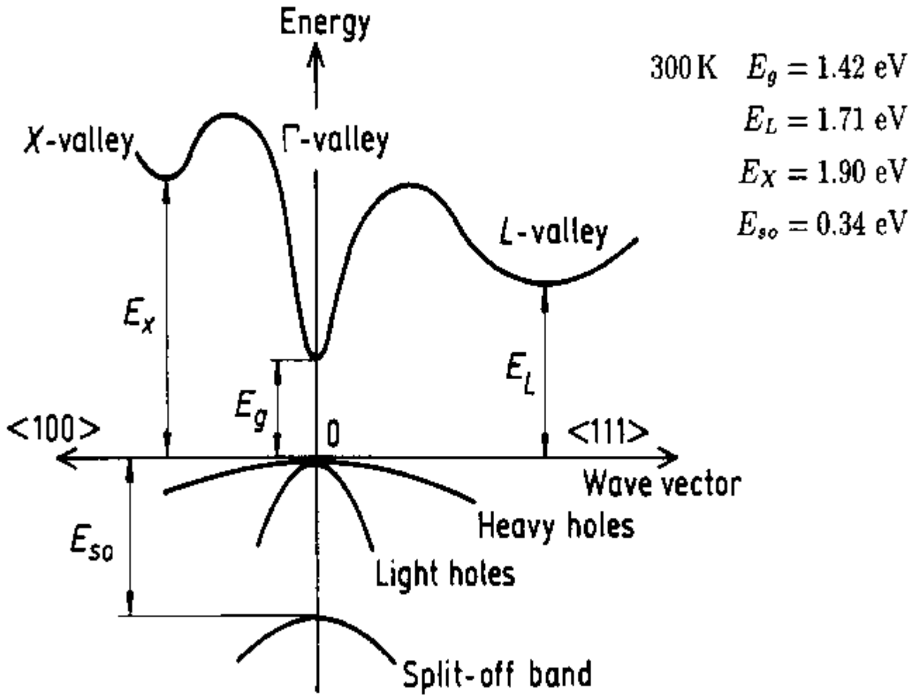
- ✗ Zero band gap material
- 👣 High mobility
- ✓ Flexibility
- ☹ Poor I_{on}/I_{off} ratio

2d materials beyond graphene



Direct and Indirect bandgap semiconductors

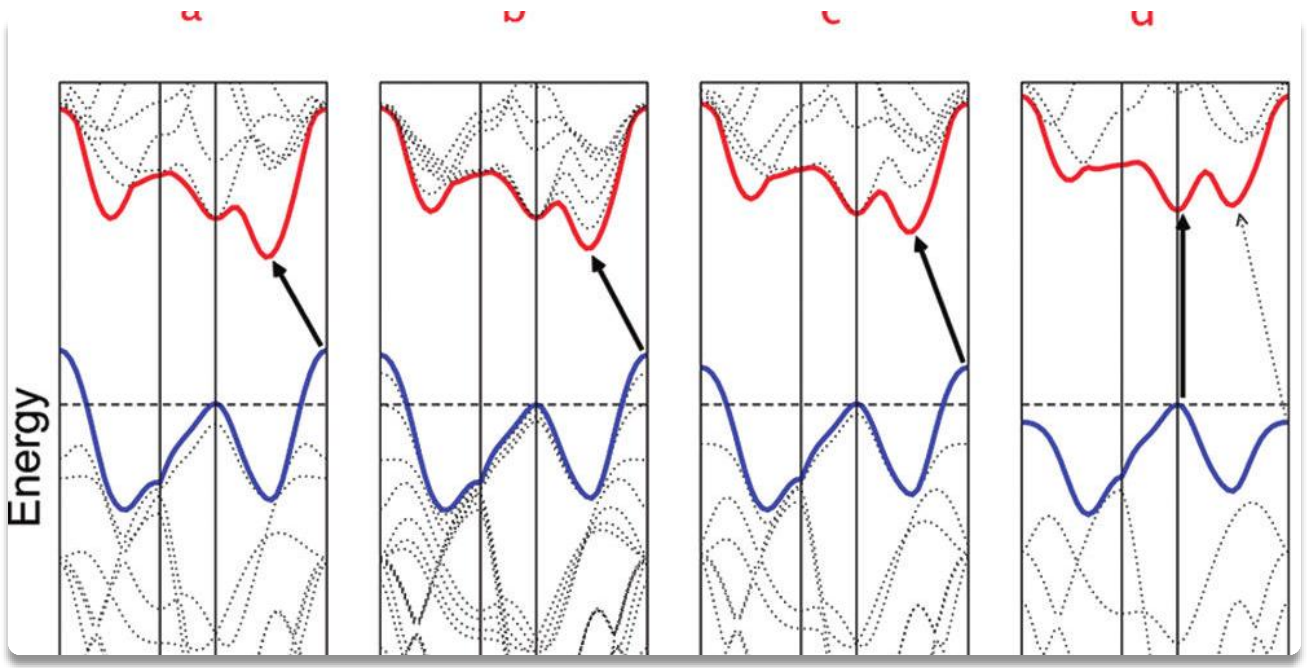




Ref. https://www.physics.byu.edu/research/coltonlab/basics.php

Direct and Indirect bandgap semiconductors

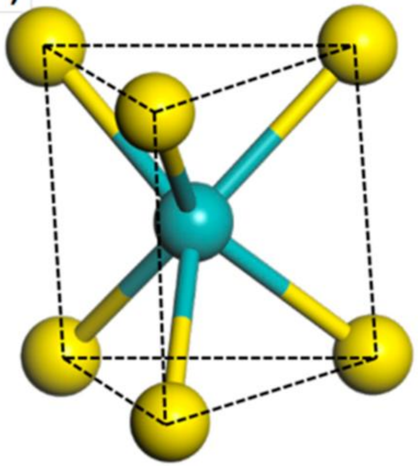
- Indirect bandgap in bulk form
- Direct bandgap in monolayer



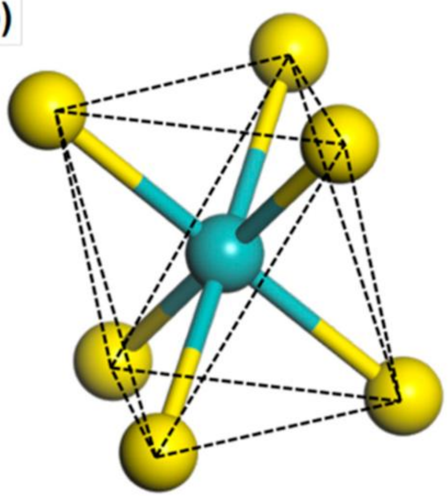
Ref. Splendiani et al.

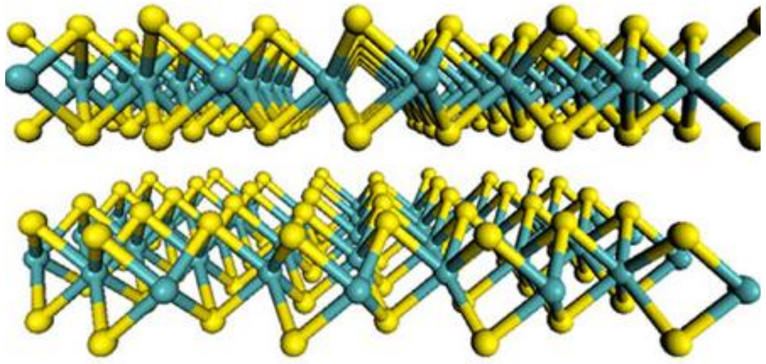
Structure of TMDs

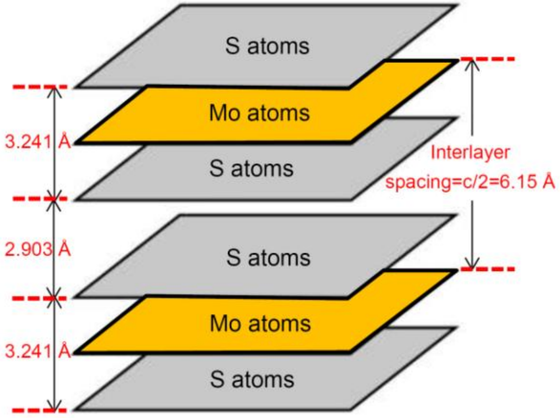
(a)



(b)







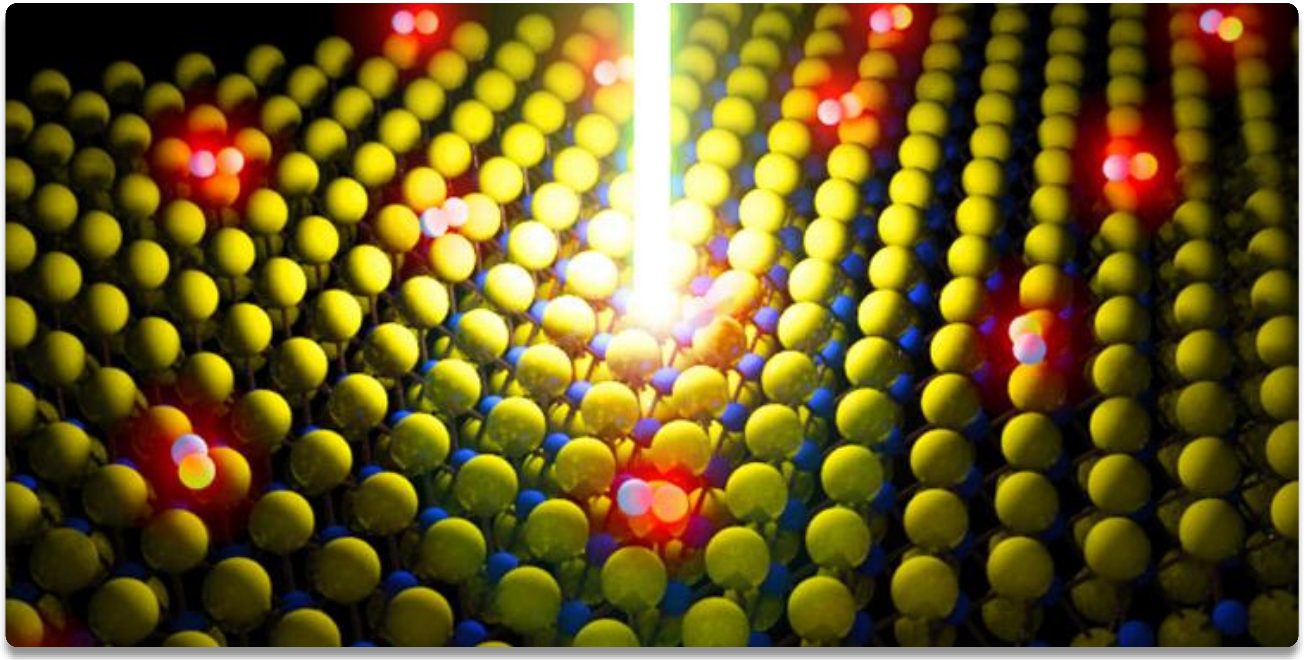
Ref. Wang et al. MIT

Properties of TMDs

- Similar to graphene layered material with ~7Å thickness
- Offers enhanced light-matter interactions
- Tunability of bandgap by external means e.g. electric field, magnetic field, strain, temperature etc.
- Provides the platform to study different quantum mechanical phenomena
- Realization of advanced opto-electronics devices

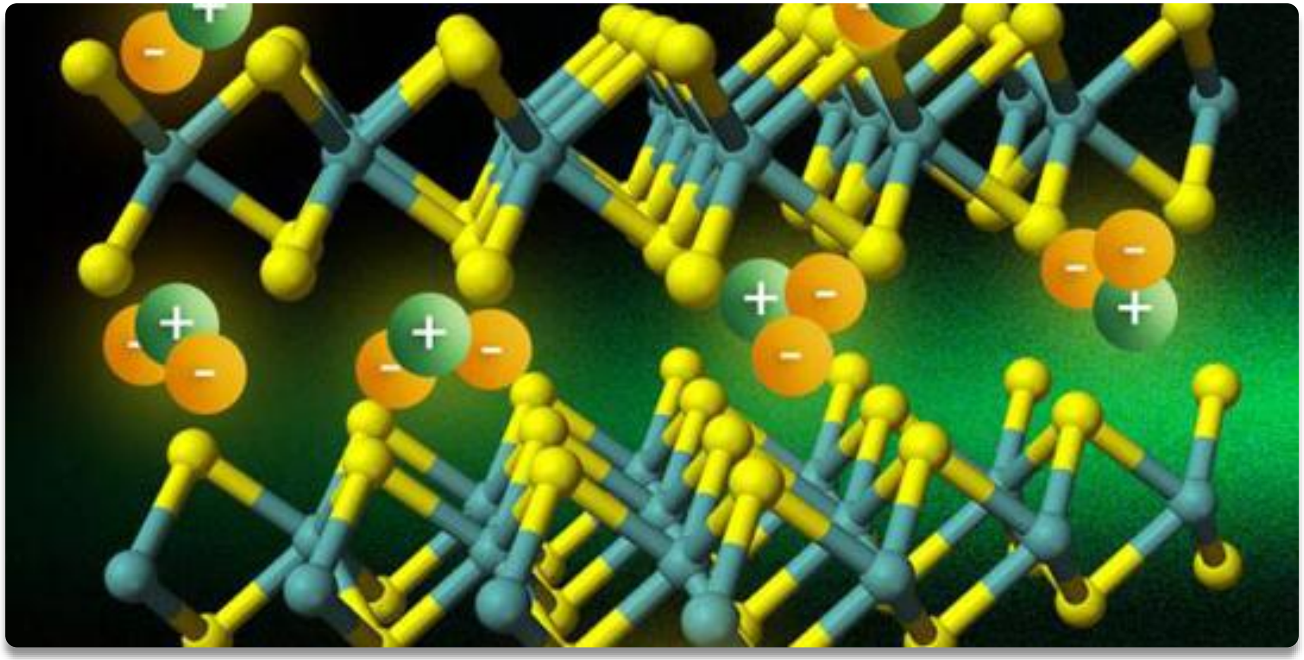
Excitons in TMDs

- Bound electron and hole pair, known as excitons
- Electrically neutral
- Highly confined in a 2D plane
- High binding energy



Ref. Science 2015 UCB

Excitons in TMDs

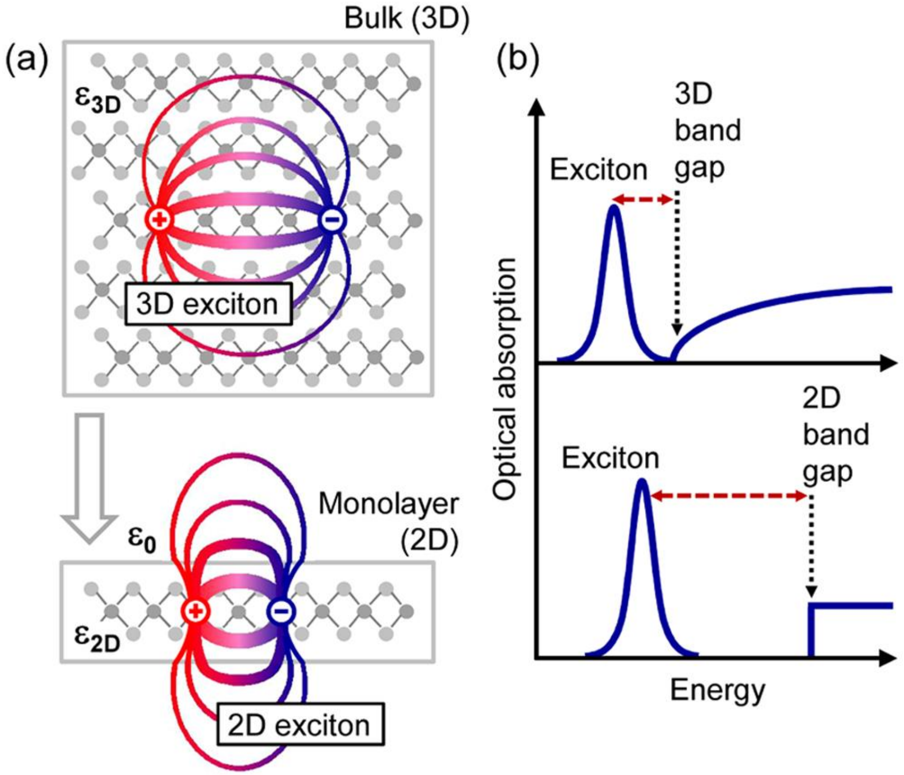


- Small effective Bohr radius
- Higher effective mass
- Reduced dielectric screening

Ref. Jose-Luis Olivares/MIT

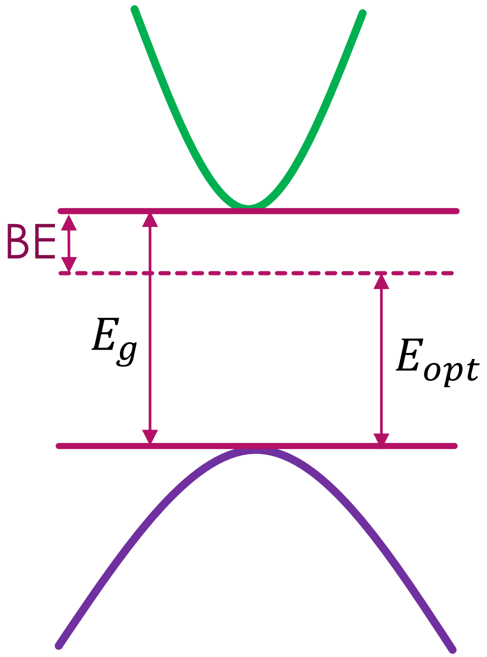
Binding energy of exciton in TMDs

- Dielectric screening is reduced in 2D system compared to the 3D system
- The monolayer excitons are strongly confined to a single layer and experiences reduced screening as the electric field penetrates outside the material.



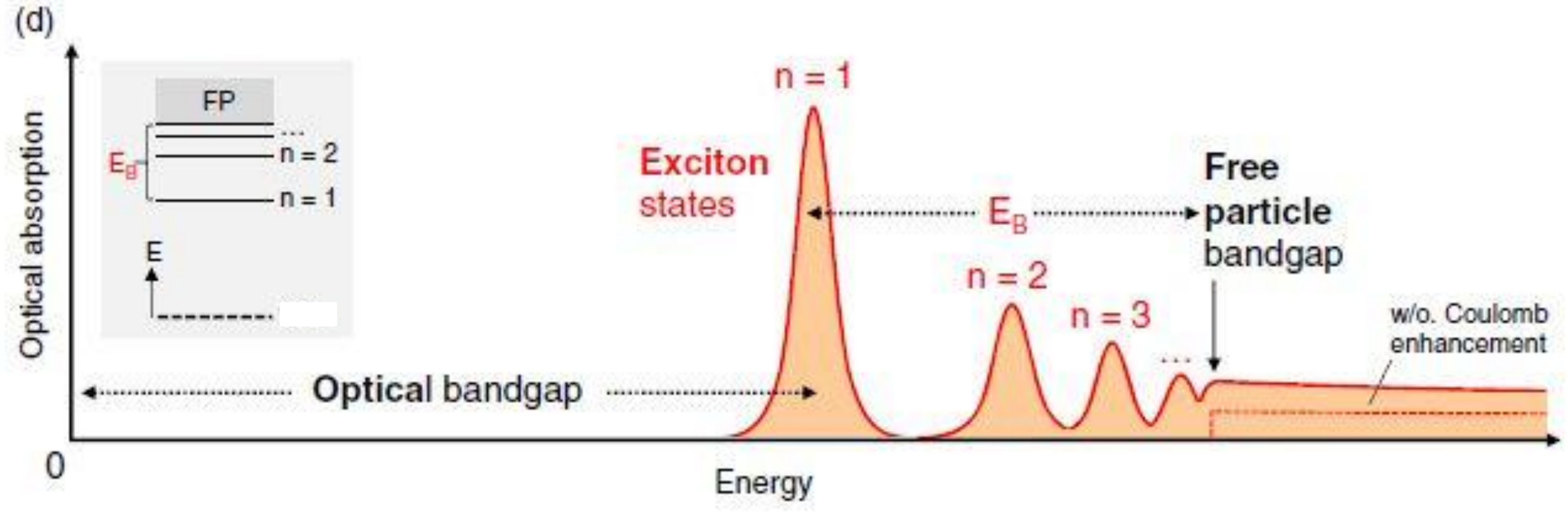
Ref. Chernikov et al. PRL.2014

Free particle bandgap and Optical bandgap



Binding energy=E_g-E_{opt}
E_{opt}= Optical bandgap
E_g= Quasiparticle bandgap

Binding energy of excitons in TMDCs is defined as the energy difference between quasiparticle (QP) bandgap and optical bandgap

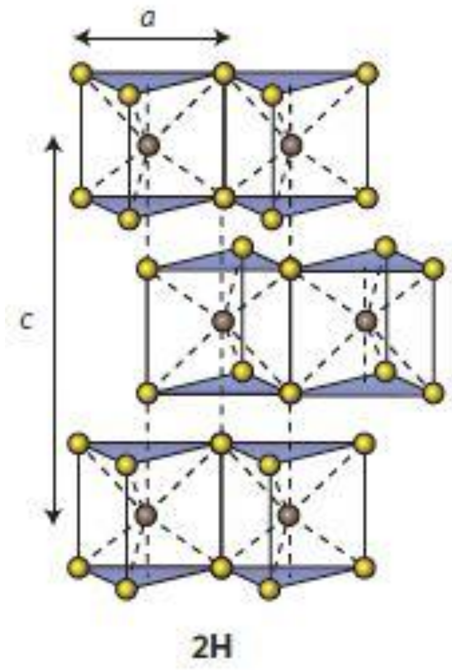


Ref. Wang et al. RevModPhys. 2018

Structure of TMDs

- TMDCs crystal structure consisting of weakly coupled sandwich layers X-M-X, where M atom layer is enclosed within two X layers and atoms in layers are hexagonally packed.

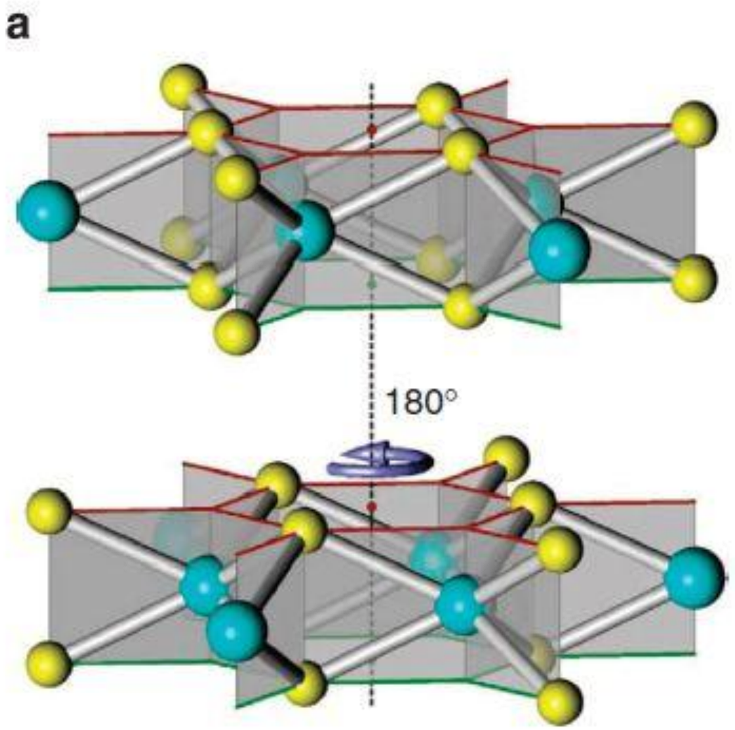
- Breaks the inversion symmetry in monolayer.



[Ref. *Wang et al. Nnano.2012*]

Structure of TMDs

- Group-VI TMDC bilayers are AB stacked i.e. one monolayer sits on another but with 180° rotation. Pristine bilayers are therefore inversion symmetric.



Ref. Gong et al. Ncomms.2013

Quasiparticle bandgap of TMDs

$$H_{2L} = \begin{bmatrix} \mathit{\Delta} & at_i(gk_x + ik_y) & 0 & 0 \\ at_i(gk_x - ik_y) & -gs\lambda & 0 & t_\perp \\ 0 & 0 & \mathit{\Delta} & at_i(gk_x - ik_y) \\ 0 & t_\perp & at_i(gk_x + ik_y) & gs\lambda \end{bmatrix}$$

Δ is the monolayer bandgap,

a is the lattice constant,

t_i is the nearest-neighbour intra-layer hopping,

λ is the spin-valley coupling for holes in monolayer,

$t_\perp$ is the interlayer hopping for holes,

g is the valley degree of freedom ($+1$ for K and -1 for K'), and

s is spin degree of freedom (± 1)

Ref. Gong et al. Ncomms.2013

Quasiparticle bandgap of TMDCs

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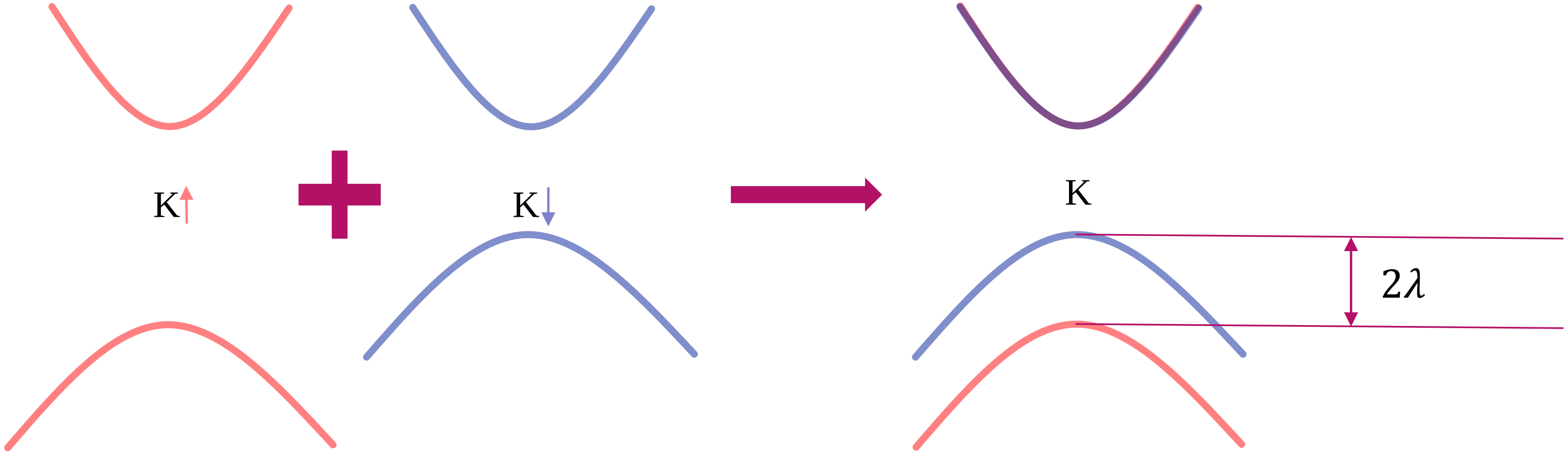


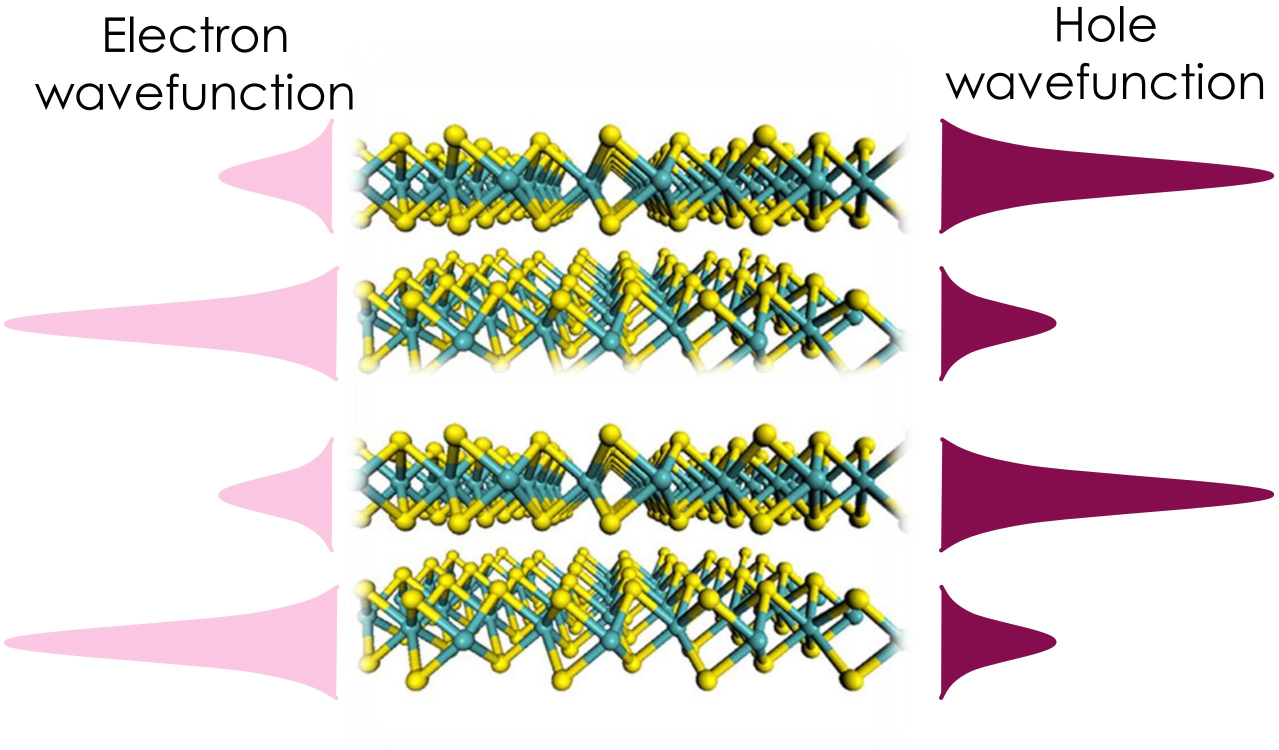
For a given valley two opposite spin configuration

Quasiparticle bandstructure of multilayer TMDCs

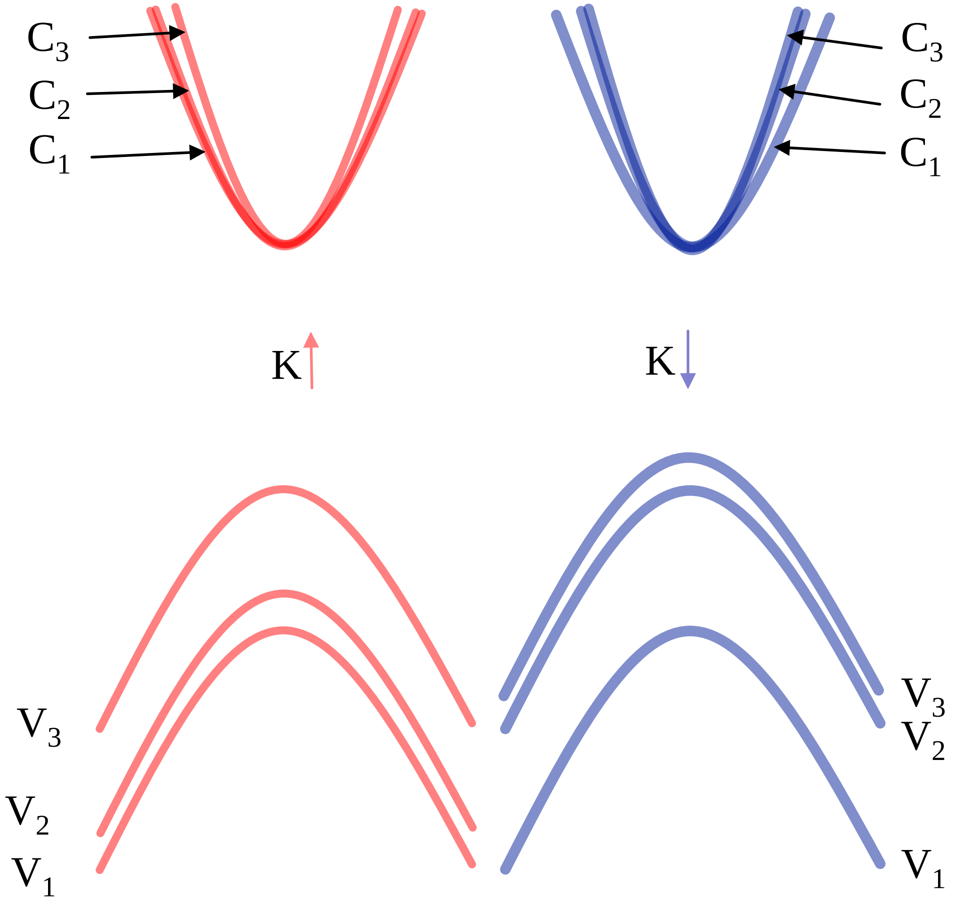
$$H_{4L}=\begin{bmatrix}
\Delta & at_i(gk_x+ik_y) & 0 & 0 & 0 & 0 & 0 & 0\\
at_i(gk_x-ik_y) & -gs\lambda & 0 & t_\perp & 0 & 0 & 0 & 0\\
0 & 0 & \Delta & at_i(gk_x-ik_y) & 0 & 0 & 0 & 0\\
0 & t_\perp & at_i(gk_x+ik_y) & gs\lambda & 0 & t_\perp & 0 & 0\\
0 & 0 & 0 & 0 & \Delta & at_i(gk_x+ik_y) & 0 & 0\\
0 & 0 & 0 & t_\perp & at_i(gk_x-ik_y) & -gs\lambda & 0 & t_\perp\\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & at_i(gk_x-ik_y)\\
0 & 0 & 0 & 0 & 0 & t_\perp & at_i(gk_x+ik_y) & gs\lambda
\end{bmatrix}$$

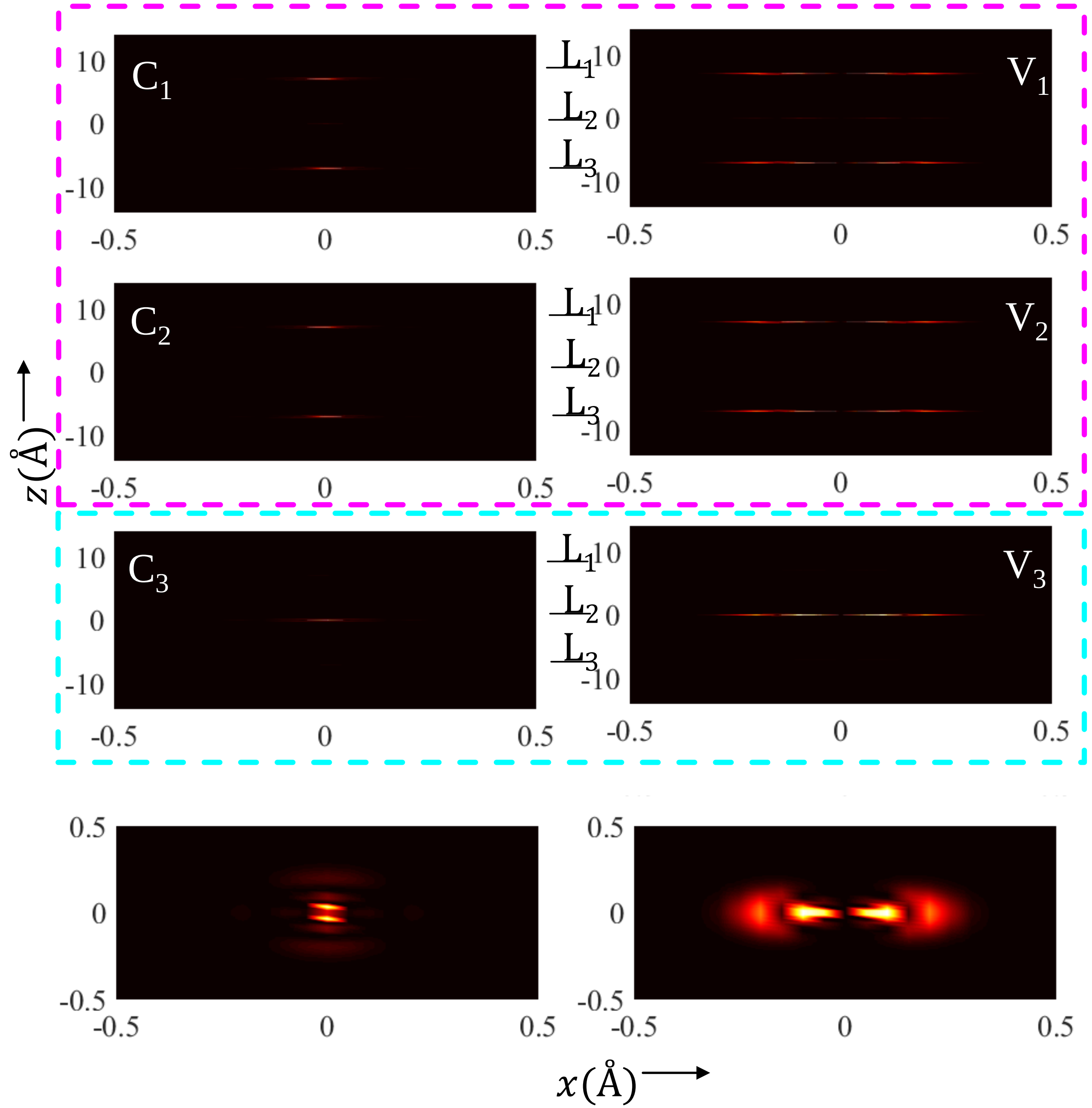
Formation of excitons in multilayer TMDs

- Alternative distribution of electrons and holes in different layers.
- Formation of even and odd pairs.



Layer distribution of wave functions





From QP bandgap to Optical bandgap

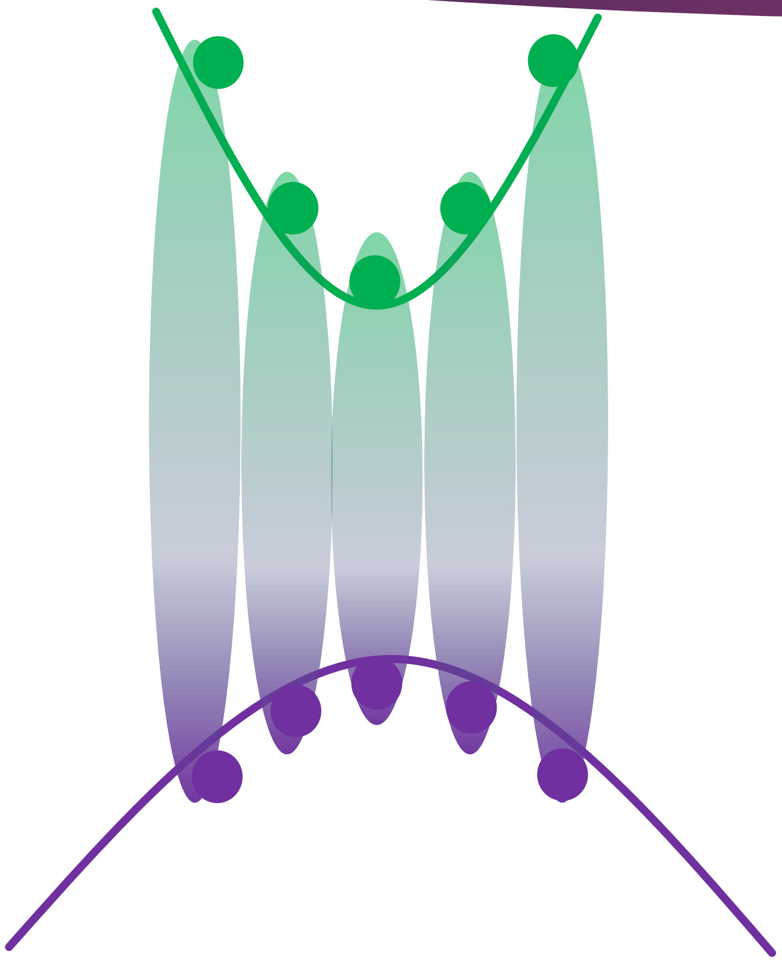
Solve the QP Hamiltonian

Put the values into Bethe Salpeter eq^n

Different energy states Corresponds to 1s,2s,2p…

Get the continuum value i.e the QP bandgap.

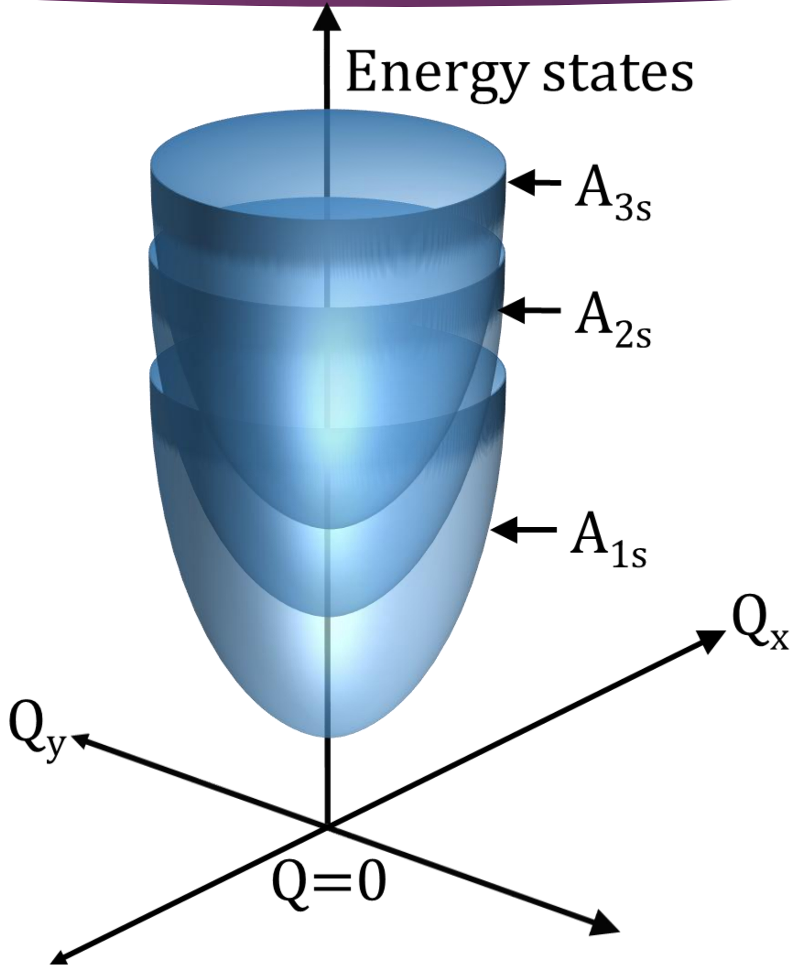
Calculation of optical bandgap



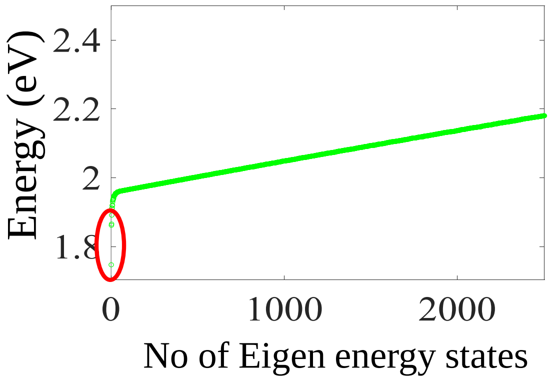
- Here we have considered the direct transitions i.e. Q=0 where Q is the exciton centre of mass momentum.
- We interested to find the k-space distribution of the excitons which is defined as:

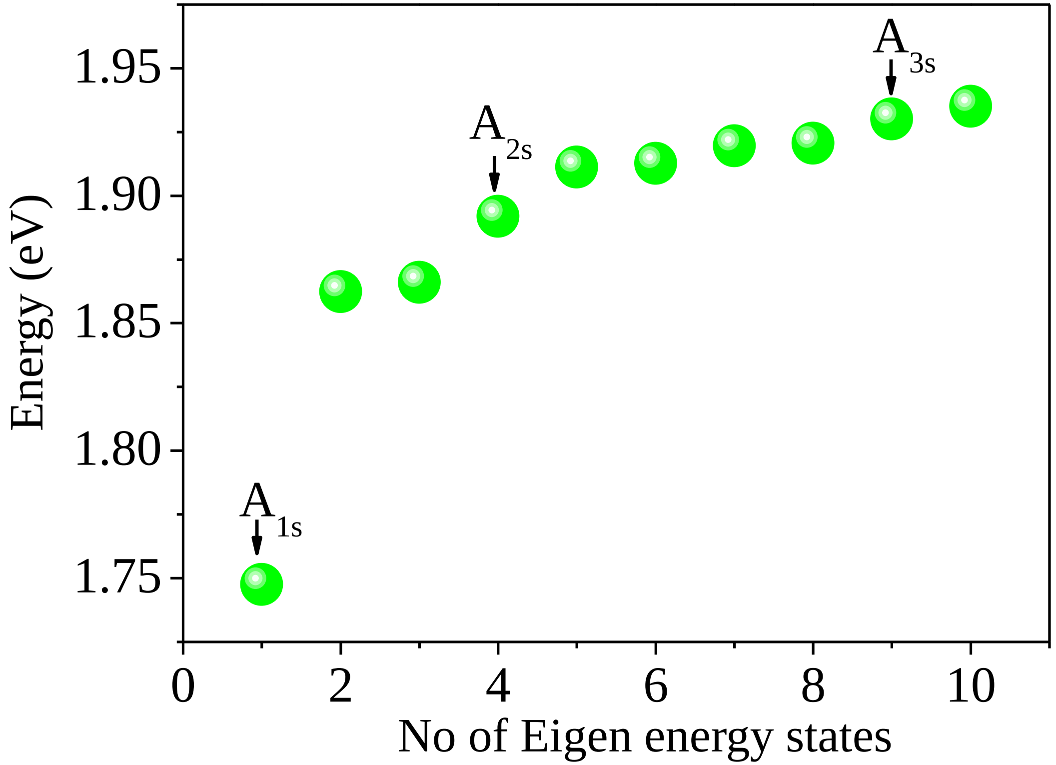
$$P_{\vec{Q}}(\vec{k}) = \sum_{v,c} |\psi_{\vec{Q}}(v,c,\vec{k})|^2.$$

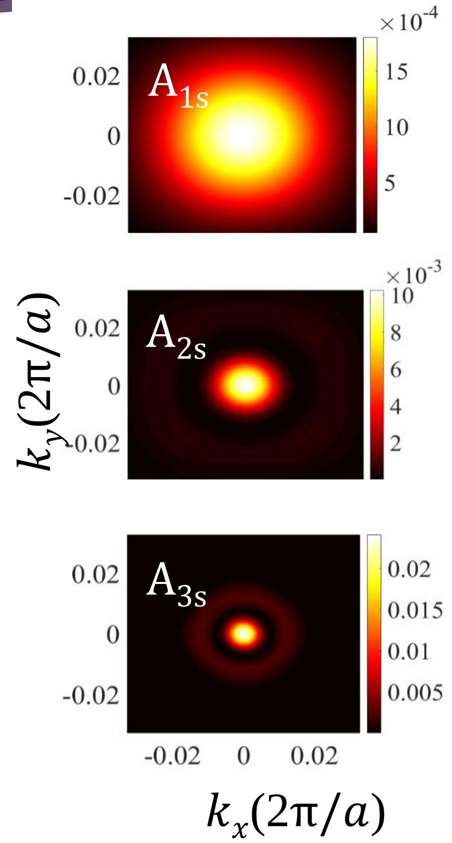
Exciton in Q space



k-space distribution of exciton

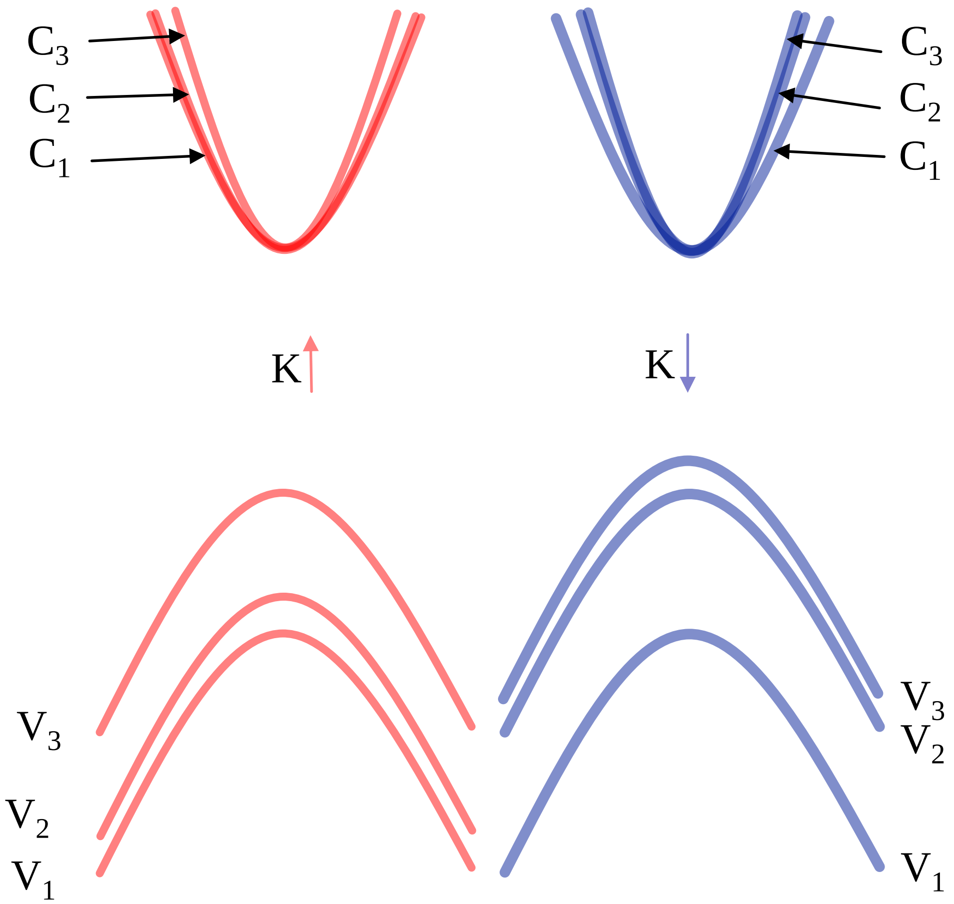






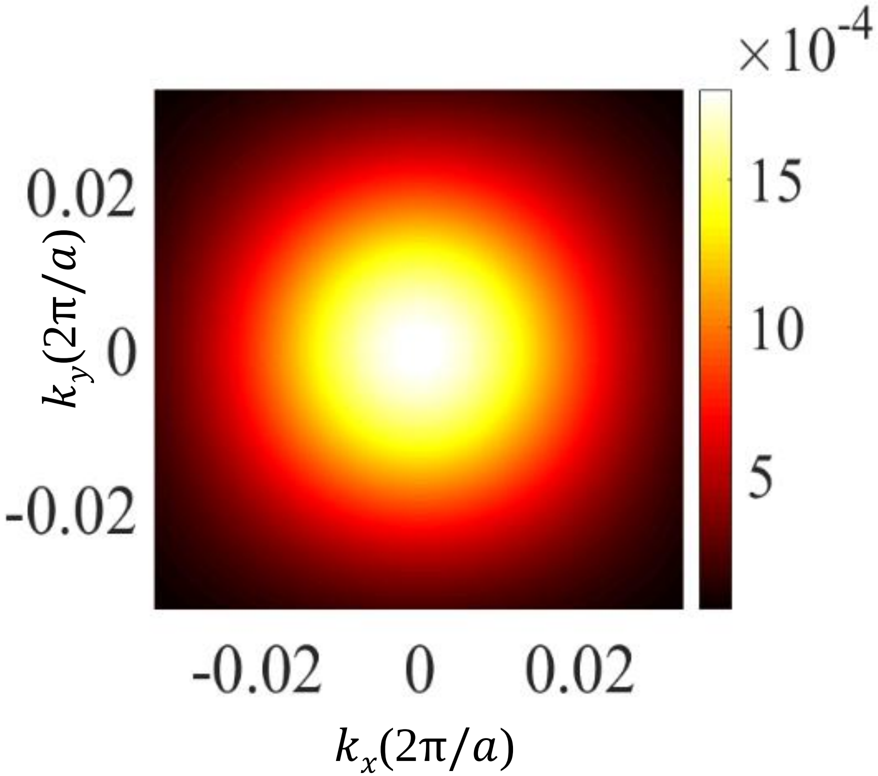
k-space distribution of exciton for different energy states for 1L WSe_2

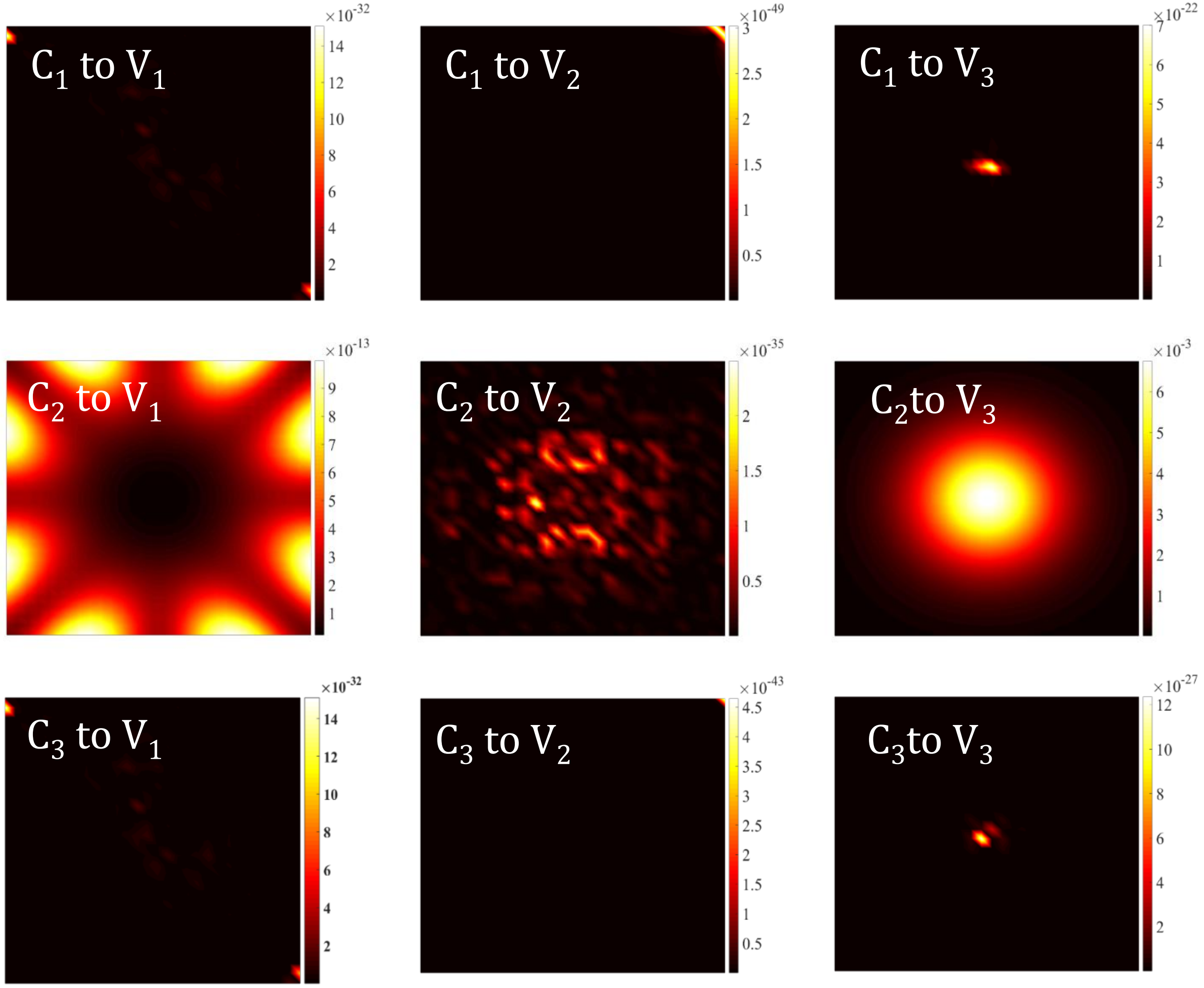
Exciton formation with possible transitions

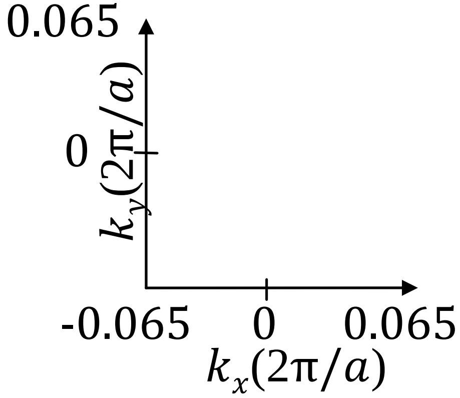


- Even though it appears from the layer distribution of the wave functions that bound exciton can be formed by the transitions from all valence band to conduction band but actually only the top valence band contributes to the exciton formation.

Exciton formation with possible transitions

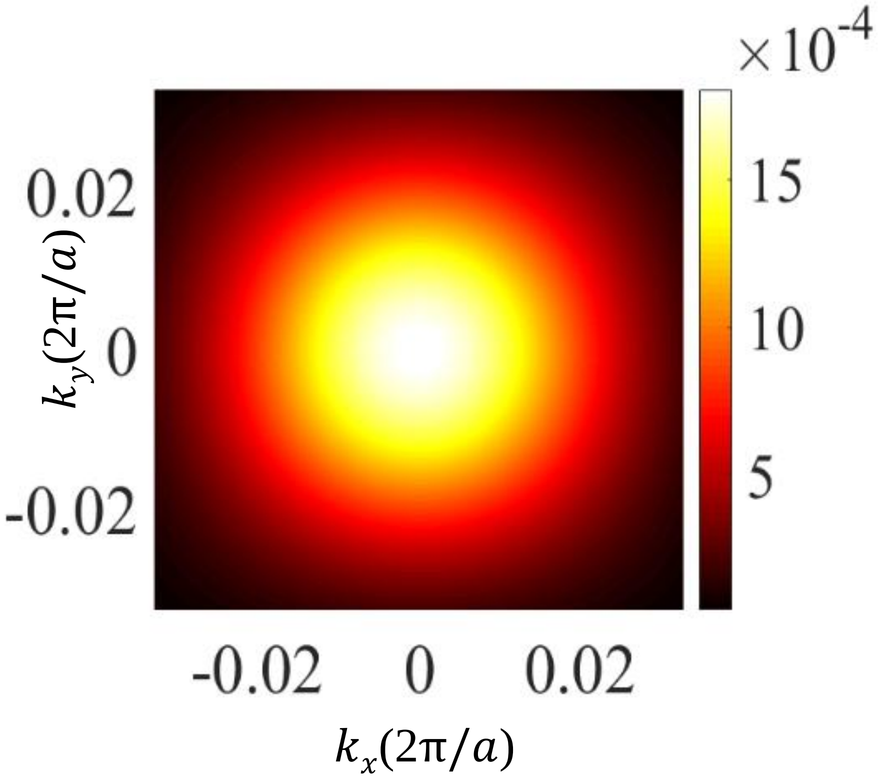


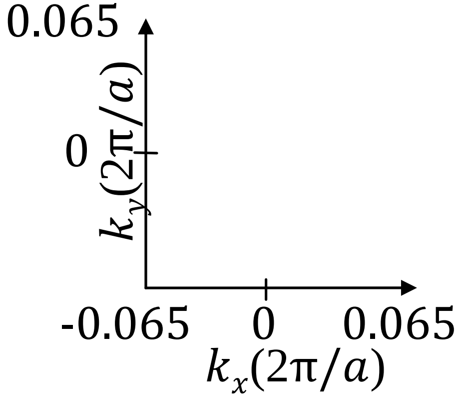


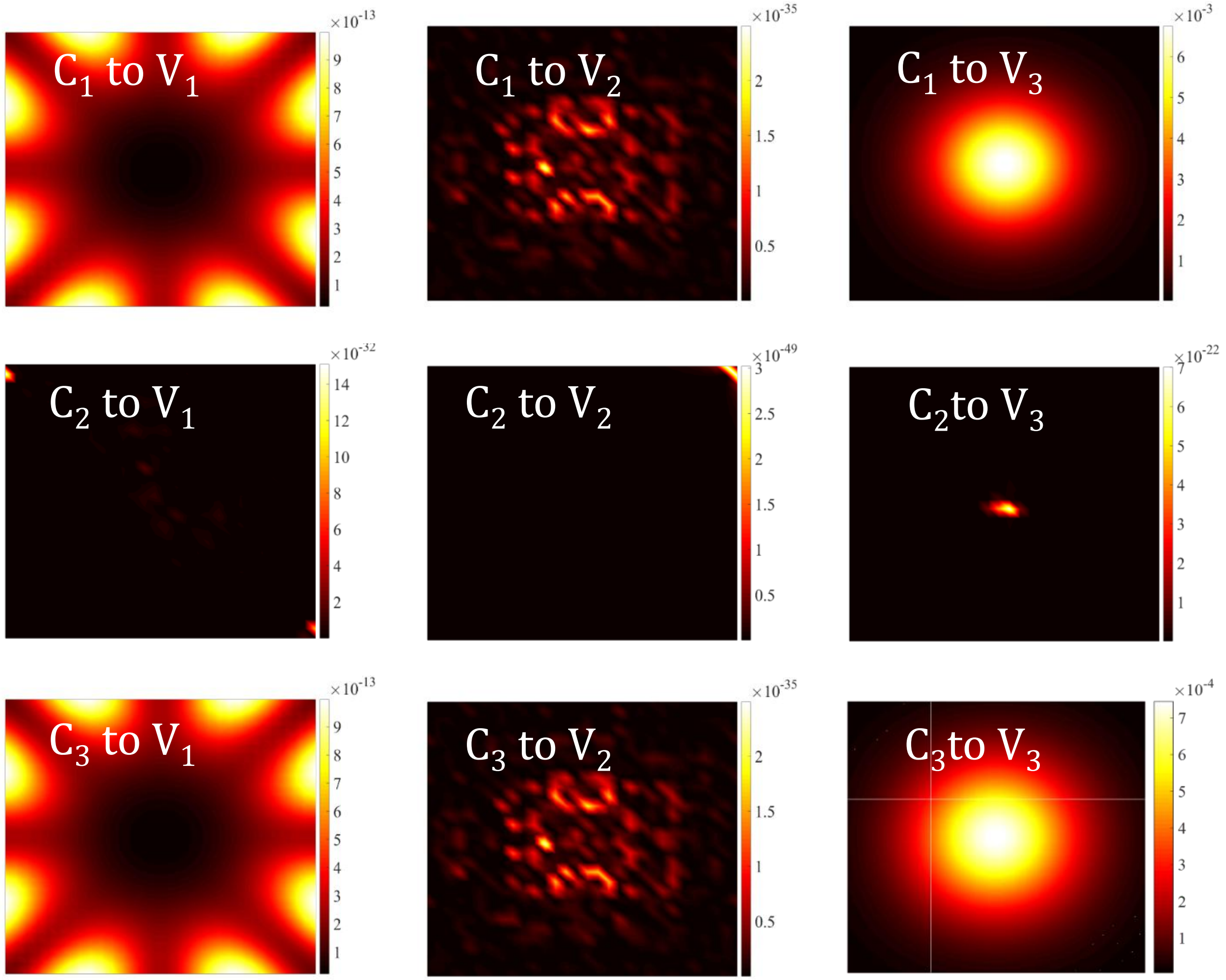


Inter-layer

Exciton formation with possible transitions



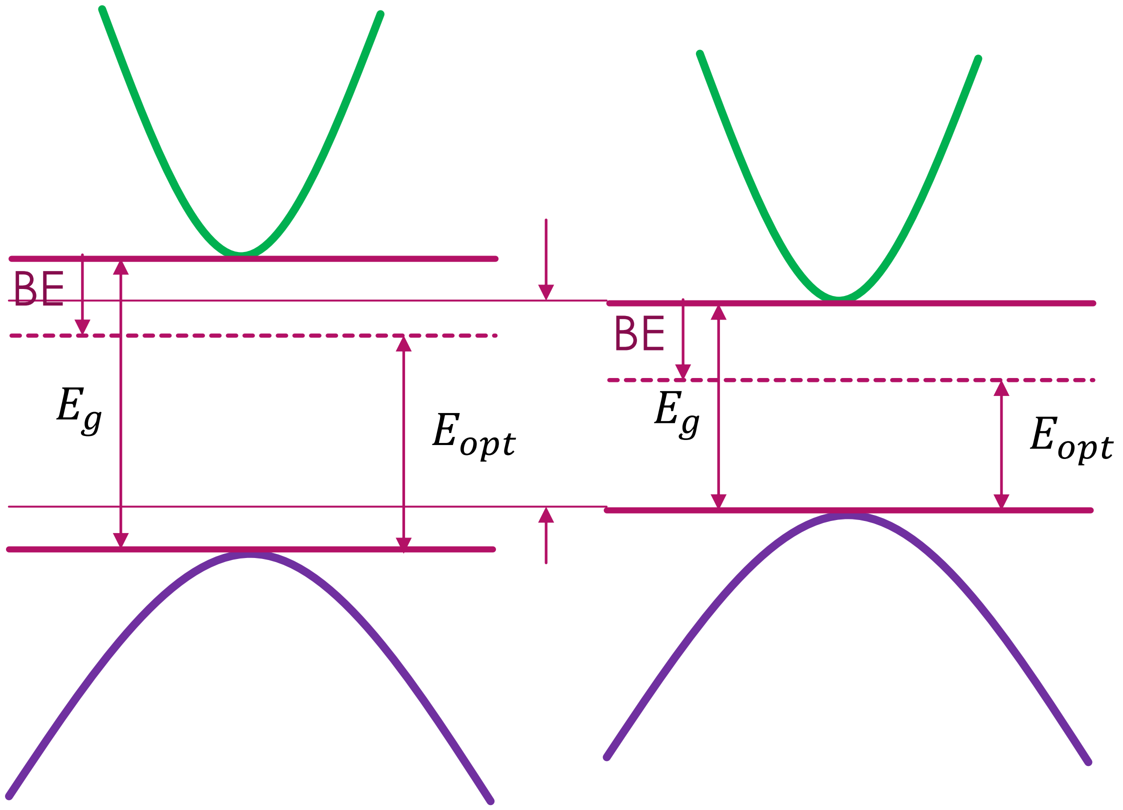




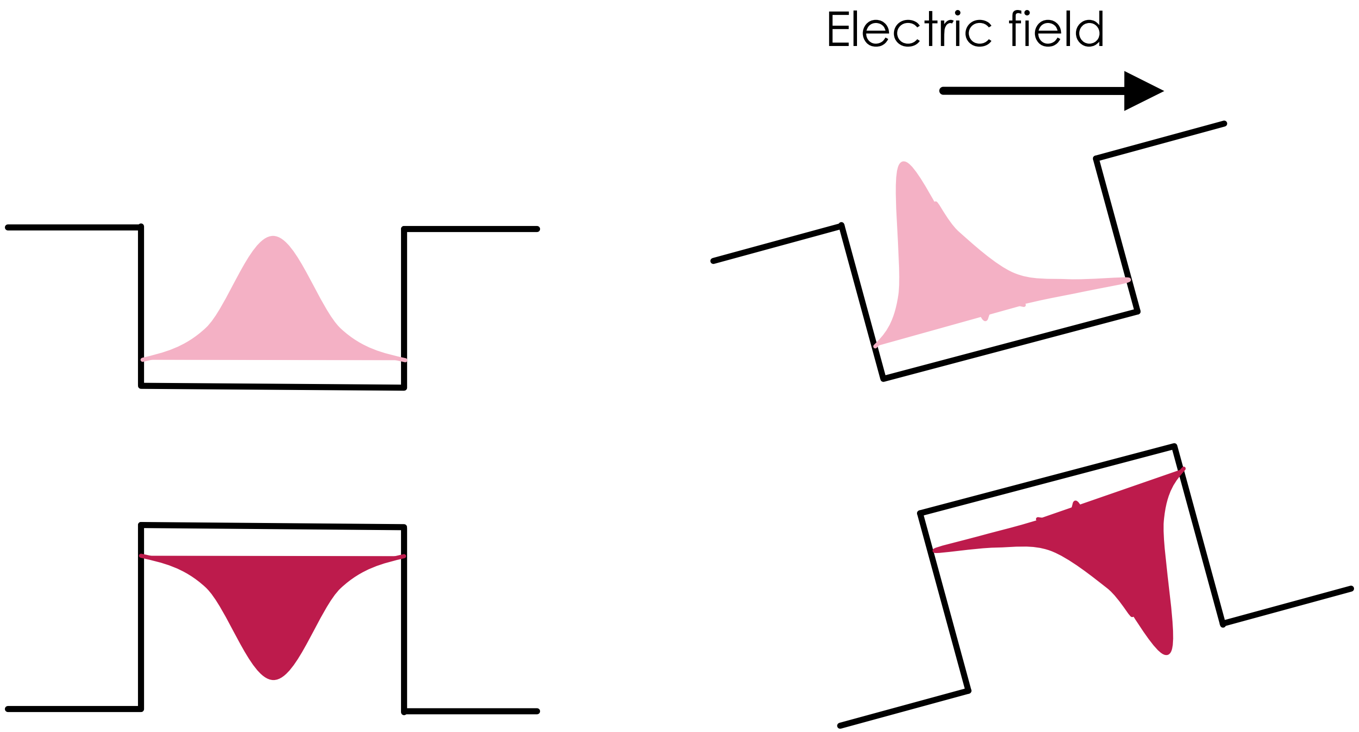
Intra-layer

Quantum Confined Stark Effect

- ***Stark Effect:*** Splitting and shifting of energy states due to external field



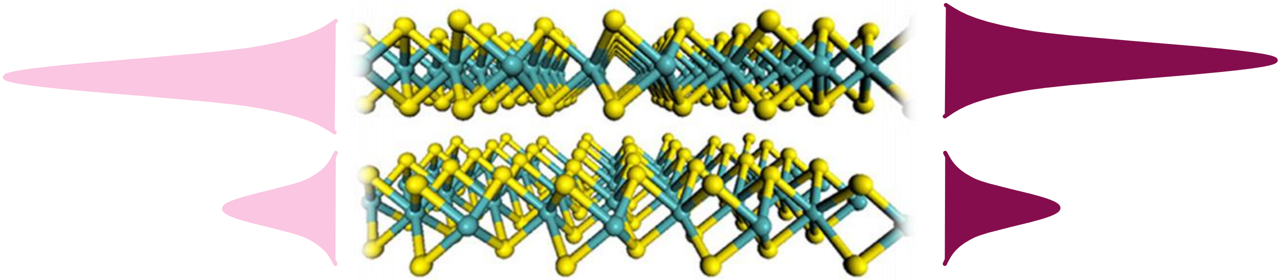
Stark Effect and Quantum Confined Stark Effect (QCSE)



- ***QCSE:*** This is the change in the relative position of wave function with applied electric field in QW structures resulting in reduction emission frequency. (*Miller et al. PRL.1984*)

Intra and inter-layer excitons

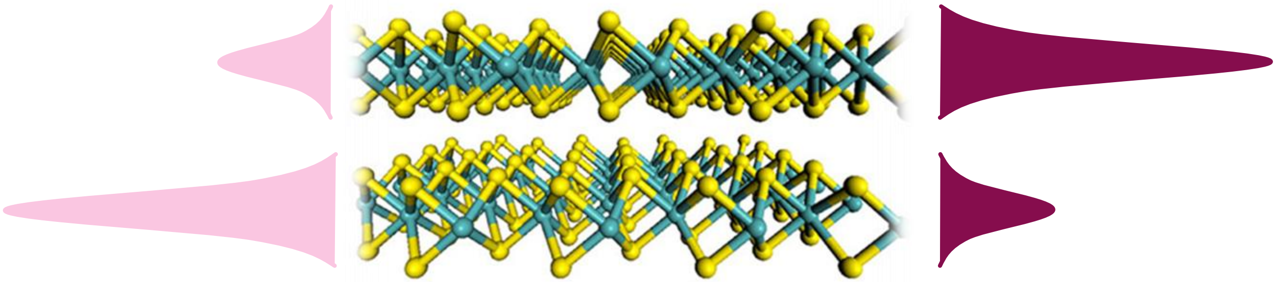
- Depending upon the distribution of carriers in the same layer or in the different layer excitons can be classified as intra and inter-layer excitons



Electron wavefunction

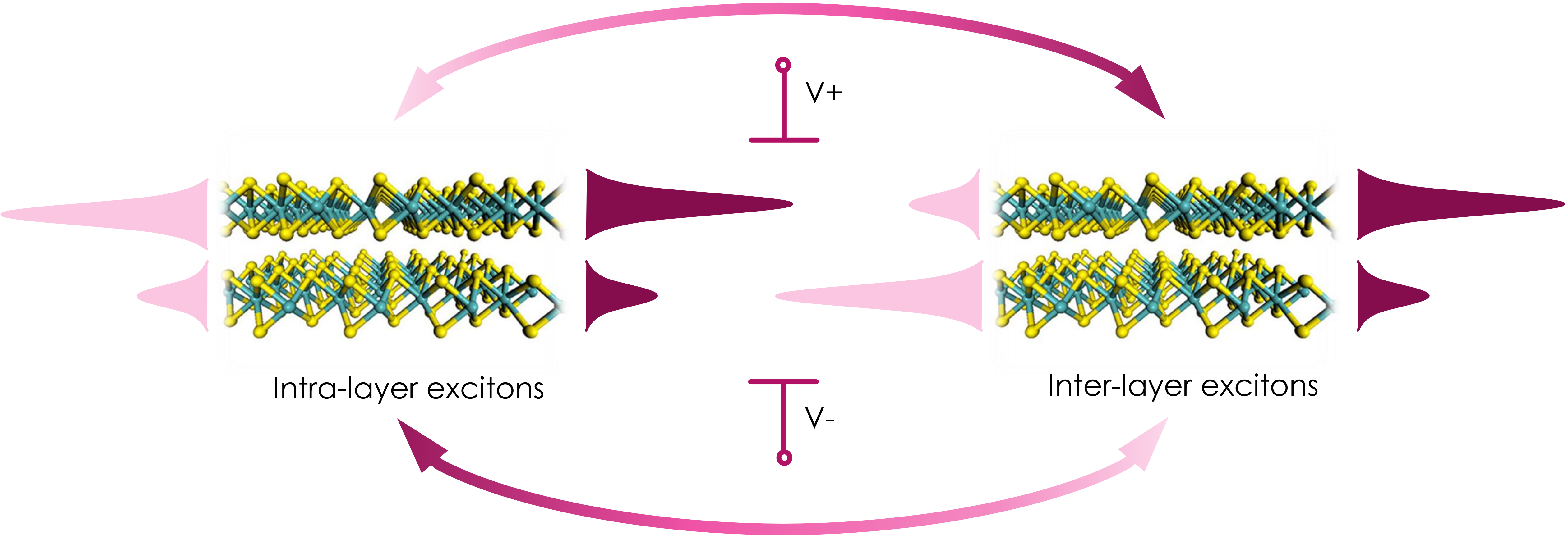
Intra-layer excitons

Hole wavefunction

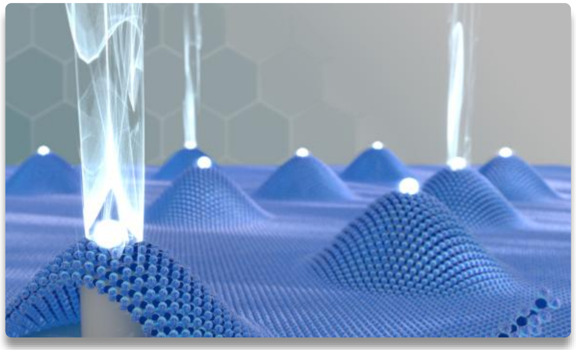


Inter-layer excitons

Tunability with external field

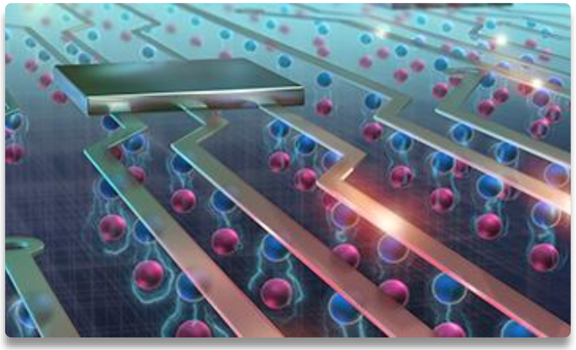


Potential applications



LEDs/SPEs

Recombination of electron and hole pair; gives out the light



Excitonic devices

Create strongly bound electron and hole pair

Photodetectors

Separate the carriers and collect through contacts

Ref. Pawel Latawiec/Harvard University
LANES EPFL
Photodetectors Definitions

THANK YOU