Exciting excitons in layered materials

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2d materials

GRAPHITE-ALLOTROPE OF CARBON
Graphene

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

- Discovered in 2004
- Nobel prize in 2010
Graphene bandstructure

- Hexagonal Brillouin 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

Direct and Indirect bandgap semiconductors

- Indirect bandgap in bulk form
- Direct bandgap in monolayer

Ref. Splendiani et al.
Structure of TMDs
Properties of TMDs

- Similar to graphene layered material with ~7 Å 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
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
Binding energy of excitons in TMDCs is defined as the energy difference between quasiparticle (QP) bandgap and optical bandgap.

\[ \text{Binding energy} = E_g - E_{\text{opt}} \]

- \( E_g \) = Quasiparticle bandgap
- \( E_{\text{opt}} \) = 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.

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

\[ H_{2L} = \begin{bmatrix}
\Delta & at_i(gk_x + ik_y) & 0 & 0 \\
-\frac{g s \lambda}{t_{\perp}} & -gs\lambda & 0 & t_{\perp} \\
0 & 0 & \Delta & at_i(gk_x - ik_y) \\
0 & t_{\perp} & at_i(gk_x + ik_y) & gs\lambda 
\end{bmatrix} \]

\( \Delta \) is the monolayer bandgap,
\( a \) is the lattice constant,
\( t_i \) is the nearest-neighbour intra-layer hopping,
\( \lambda \) 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

For a given valley two opposite spin configuration
Quasiparticle band structure of multilayer TMDCs

\[ H_L = \]

\[
\begin{bmatrix}
\Delta & \Delta & t_1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\Delta & \Delta & t_1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
t_1 & t_1 & \Delta & \Delta & \Delta & \Delta & 0 & 0 & 0 & 0 \\
t_1 & t_1 & \Delta & \Delta & \Delta & \Delta & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta & \Delta & \Delta & \Delta \\
\end{bmatrix}
\]
Formation of excitons in multilayer TMDs

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

Electron wavefunction

Hole wavefunction
Layer distribution of wave functions
From QP bandgap to Optical bandgap

Solve the QP Hamiltonian

Put the values into Bethe Salpeter eq\textsuperscript{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:

$$P_Q(\vec{k}) = \sum_{v,c} |\psi_Q(v,c,\vec{k})|^2.$$
Exciton in Q space

Energy states

$A_{3s}$

$A_{2s}$

$A_{1s}$

$Q_x$

$Q_y$

$Q=0$
k-space distribution of exciton

k-space distribution of exciton for different energy states for 1L WSe$_2$
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

$\mathbf{C}_1$ to $\mathbf{V}_1$

$\mathbf{C}_2$ to $\mathbf{V}_1$

$\mathbf{C}_3$ to $\mathbf{V}_1$

$\mathbf{C}_1$ to $\mathbf{V}_2$

$\mathbf{C}_2$ to $\mathbf{V}_2$

$\mathbf{C}_3$ to $\mathbf{V}_2$

$\mathbf{C}_1$ to $\mathbf{V}_3$

$\mathbf{C}_2$ to $\mathbf{V}_3$

$\mathbf{C}_3$ to $\mathbf{V}_3$
Exciton formation with possible transitions

Intra-layer
**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)*

![Diagram showing Stark Effect and Quantum Confined Stark Effect (QCSE)]
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.

Intra-layer excitons
- Electron wavefunction
- Intra-layer excitons
- Hole wavefunction

Inter-layer excitons
Tunability with external field

Intra-layer excitons

Inter-layer excitons

\[ V^+ \]

\[ V^- \]
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