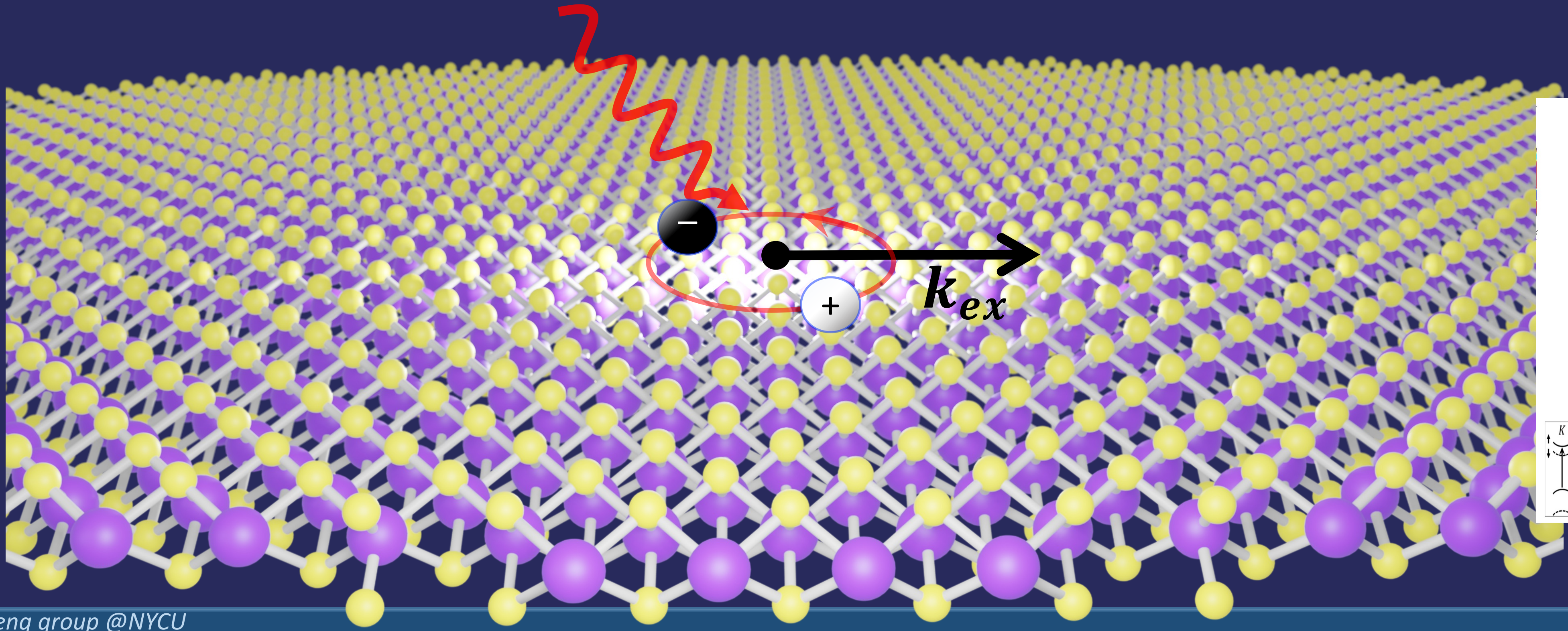
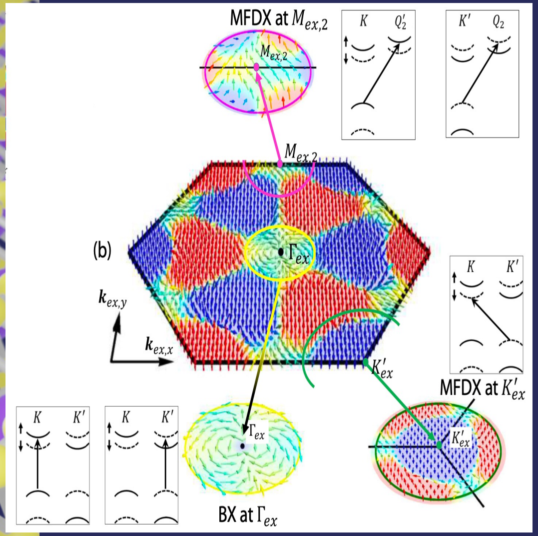
Full-Zone Landscape of momentum excitons in atomically thin transition metal dichalcogenides

Shun-Jen Cheng (鄭舜仁)

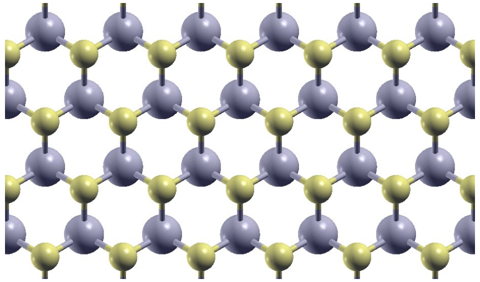
Department of Electrophysics, National Yang Ming Chiao Tung University, Taiwan





Valley exciton in transition-metal dichalcogenides (TMDs)

Top view

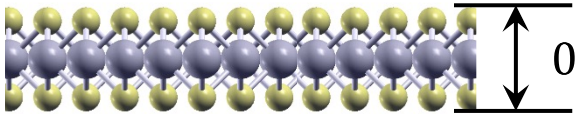


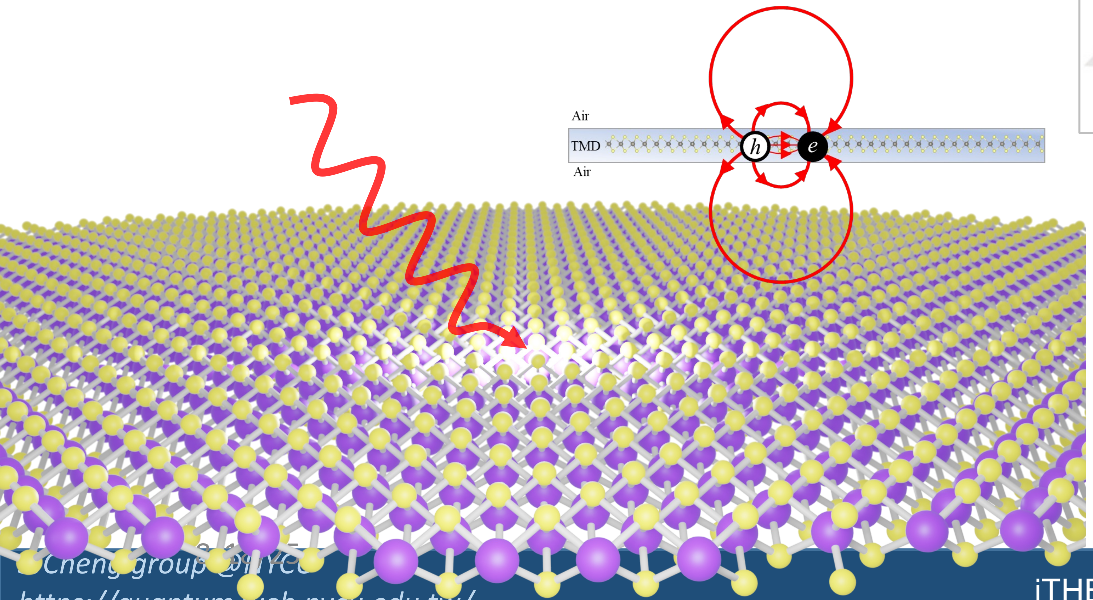
$\boldsymbol{MX_2}$,

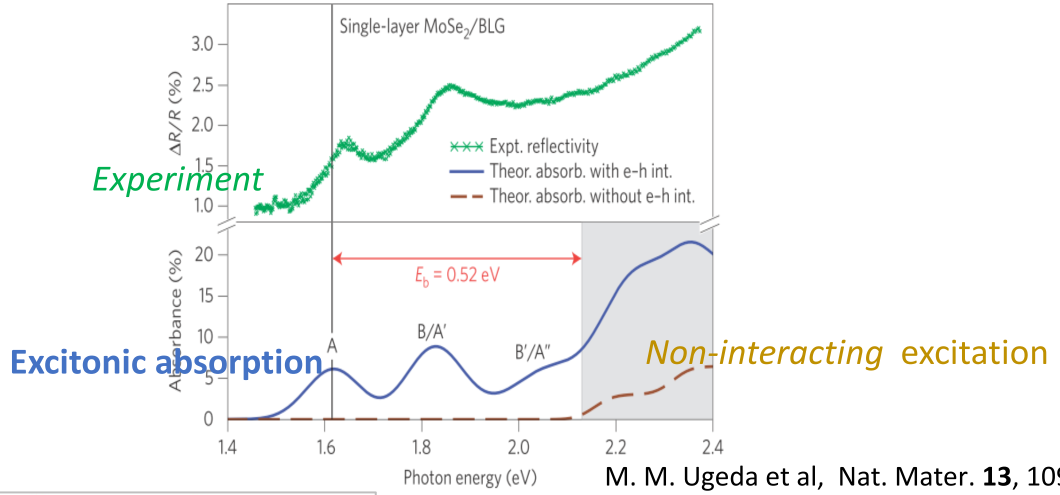
$M = Mo, W$,

$X = S, Se$

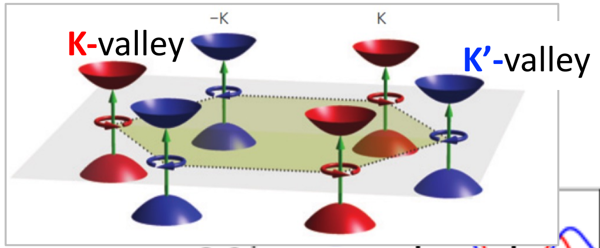
Side view

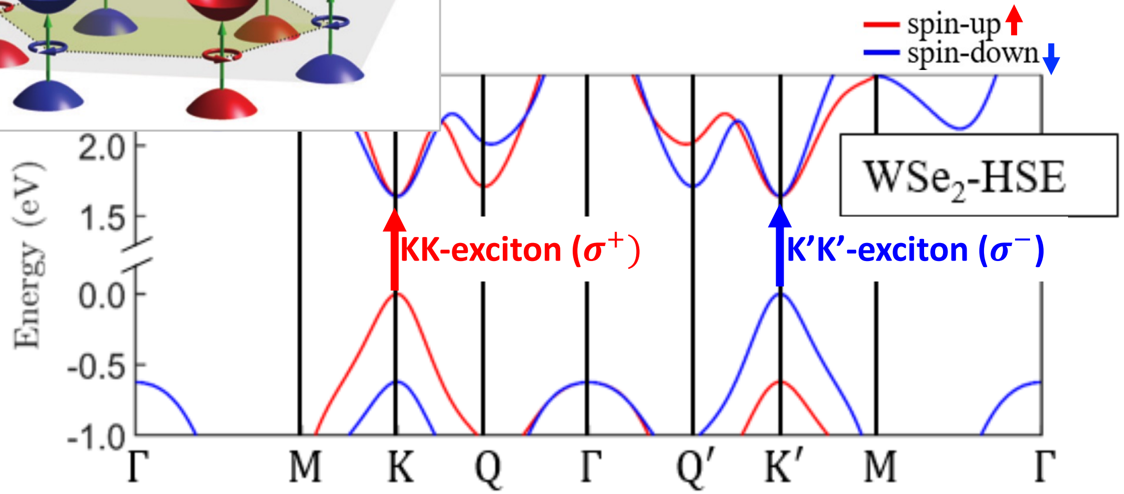






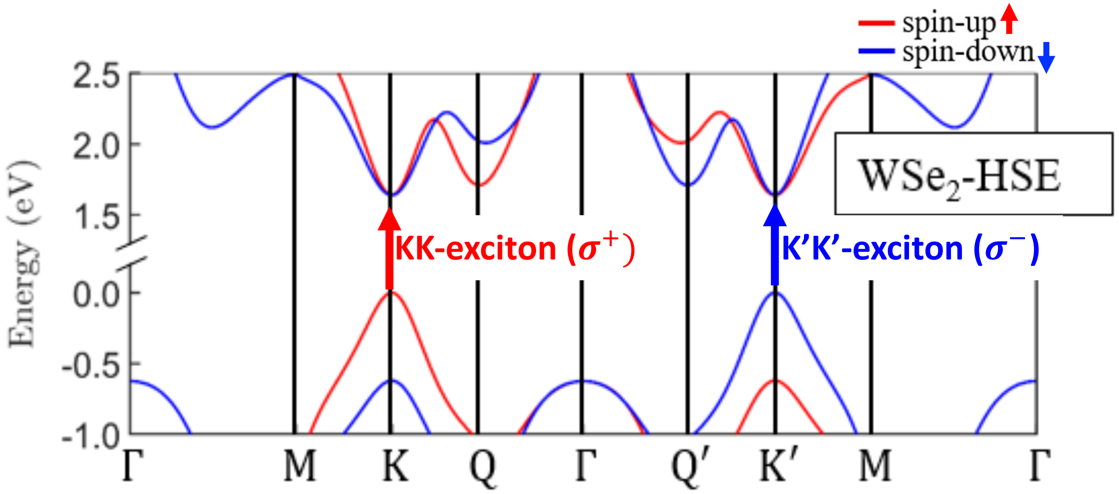
M. M. Ugeda et al, Nat. Mater. **13**, 1091 (2014).

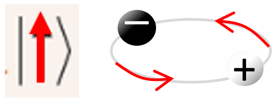


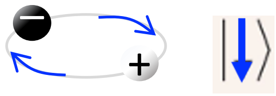


H. Yu, et al. Nat. Sci. Rev. **2**, 57 (2015).

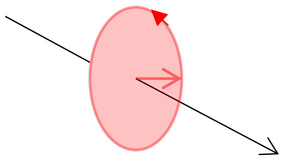
Valley exciton in transition-metal dichalcogenides (TMDs)



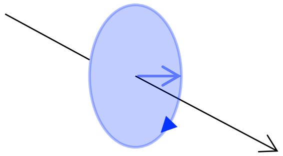




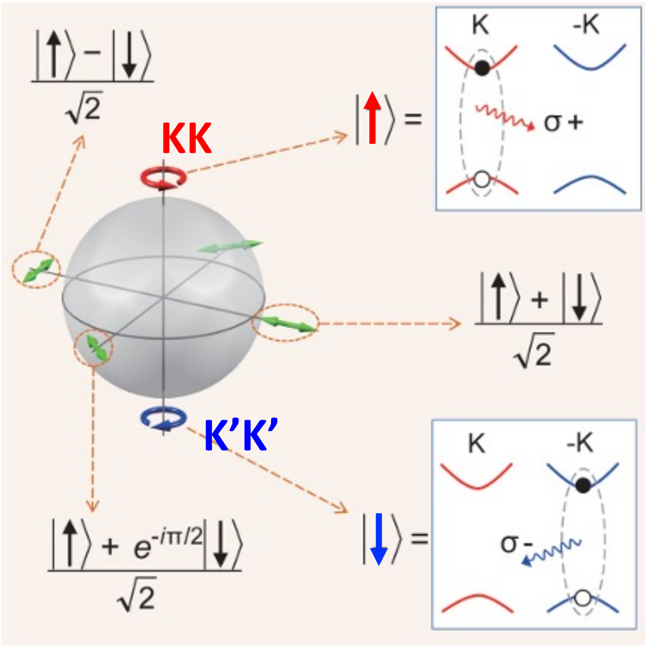
σ^+-polarization



σ^--polarization



Valley pseudospin



H. Yu, et al. Nat. Sci. Rev. **2**, 57 (2015).

Valley polarization of exciton complexes in 2D TMDs

ACS NANO
Cite This: ACS Nano 2019, 13, 14107−14113
www.acsnano.org

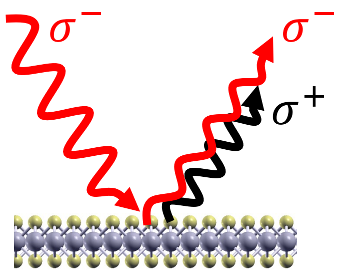
Momentum-Dark Intervalley Exciton in Monolayer Tungsten Diselenide Brightened *via* Chiral Phonon

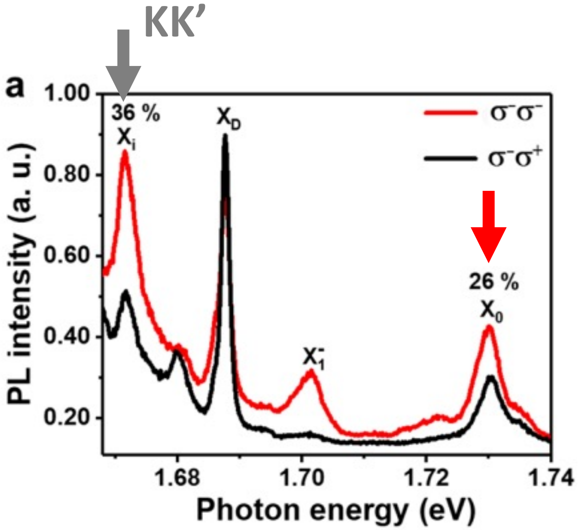
Zhipeng Li,[†,‡,^] Tianmeng Wang,[‡,^] Chenhao Jin,[§,^] Zhengguang Lu,[∥,⊥] Zhen Lian,[‡] Yuze Meng,[‡,#] Mark Blei,[○] Mengnan Gao,[%] Takashi Taniguchi,[▽] Kenji Watanabe,[▽] Tianhui Ren,[*,†] Ting Cao,[◆,¶] Sefaattin Tongay,[○] Dmitry Smirnov,[∥] Lifa Zhang,[*,%] and Su-Fei Shi[*,‡,&]

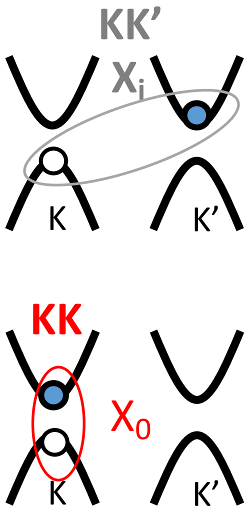
NATURE COMMUNICATIONS | (2020) 11:618 |

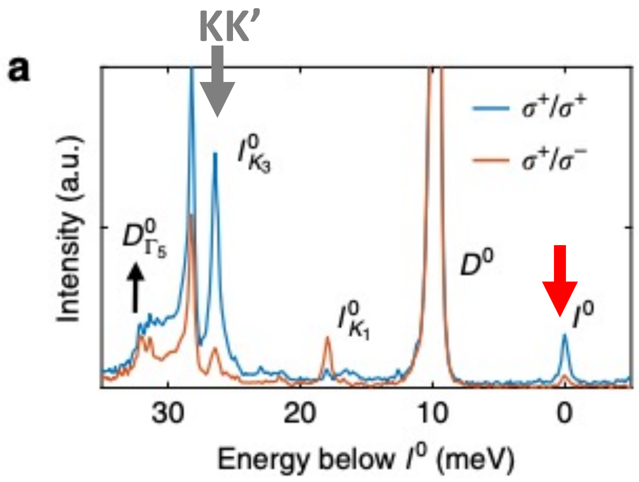
Valley phonons and exciton complexes in a monolayer semiconductor

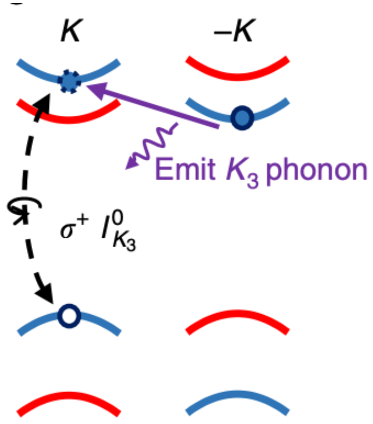
Minhao He[1,10], Pasqual Rivera[1,10], Dinh Van Tuan[2], Nathan P. Wilson[1], Min Yang[2], Takashi Taniguchi[3], Kenji Watanabe[3], Jiaqiang Yan[4,5], David G. Mandrus[4,5,6], Hongyi Yu[7], Hanan Dery[2,8*], Wang Yao[7*] & Xiaodong Xu[1,9*]



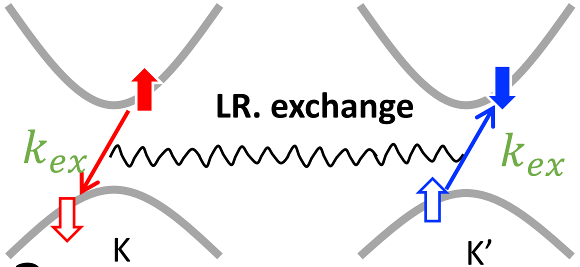






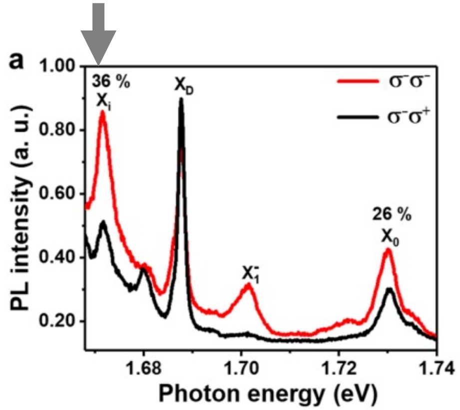


Momentum-dependent valley polarization of exciton



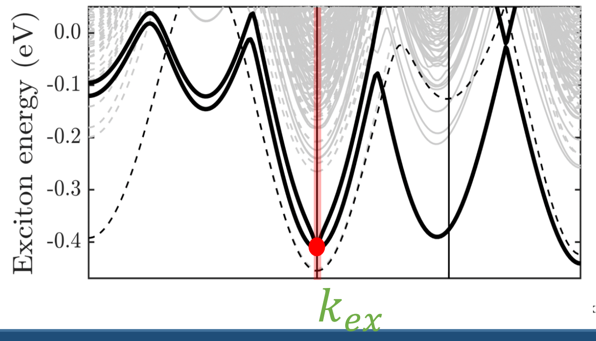
Questions:

- Why the degree of valley polarization of **BX** is **not high**?

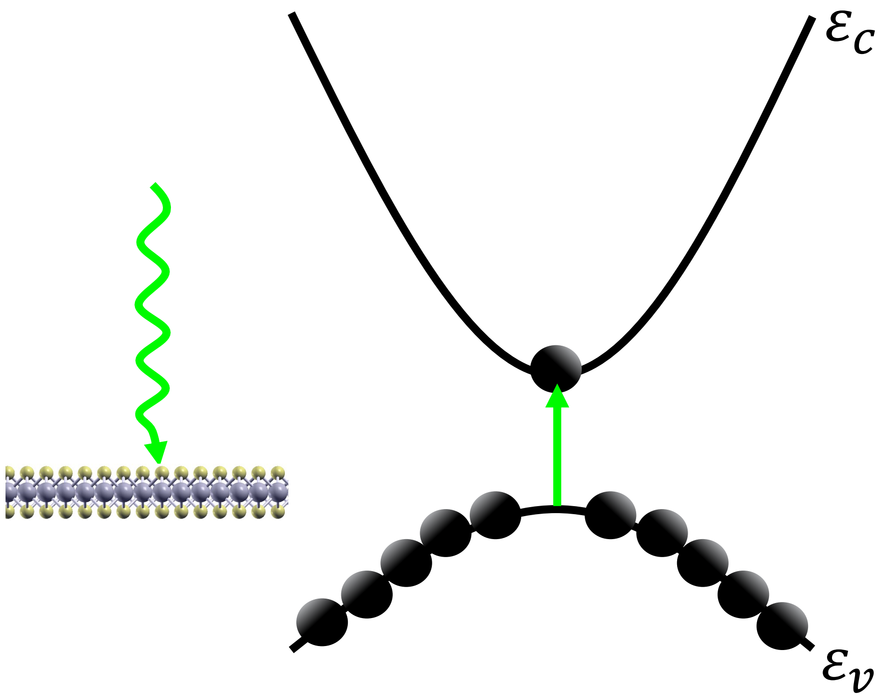


- Why the degree of valley polarization of **intervalley FM-DX** is **higher** than the BX ?

- How is the exciton-momentum ($\boldsymbol{k_{ex}}$)-dependences of the valley polarization of exciton?



Exciton: a many-body problem



Exciton: A many-body ($(N_v - 1) + 1$) problem

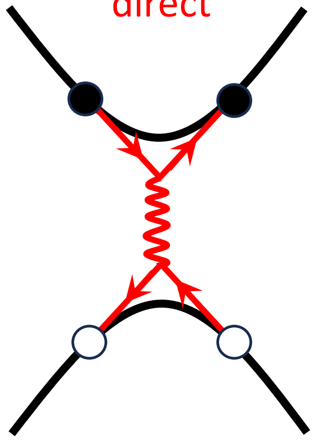
Exciton state: $|S\,\boldsymbol{k_{ex}}\rangle = \Sigma_{\boldsymbol{k}} A_{S\boldsymbol{k_{ex}}}(\boldsymbol{k})\hat{c}^{\dagger}_{k+\boldsymbol{k_{ex}}}\hat{h}^{\dagger}_{k}|0\rangle$

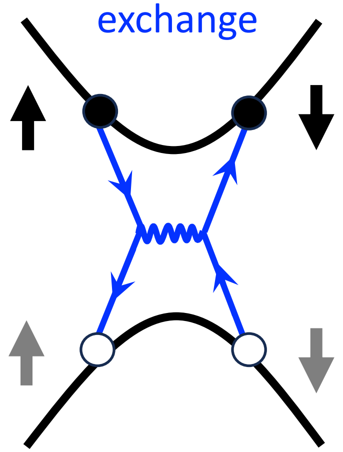
Bethe-Salpeter equation (BSE):

$$(\epsilon_{ck+\boldsymbol{k_{ex}}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{k_{ex}}}(\boldsymbol{k}) - \frac{1}{\mathcal{A}}\sum_{\boldsymbol{k'}} U_{\boldsymbol{k_{ex}}}(\boldsymbol{k},\boldsymbol{k'})A_{S\boldsymbol{k_{ex}}}(\boldsymbol{k'}) = E^{X}_{S\boldsymbol{k_{ex}}}A_{S\boldsymbol{k_{ex}}}(\boldsymbol{k})$$

$$U_{\boldsymbol{k_{ex}}}(\boldsymbol{k},\boldsymbol{k'}) = W^{d}_{\boldsymbol{k_{ex}}}(\boldsymbol{k},\boldsymbol{k'}) - V^{x}_{\boldsymbol{k_{ex}}}(\boldsymbol{k},\boldsymbol{k'})$$

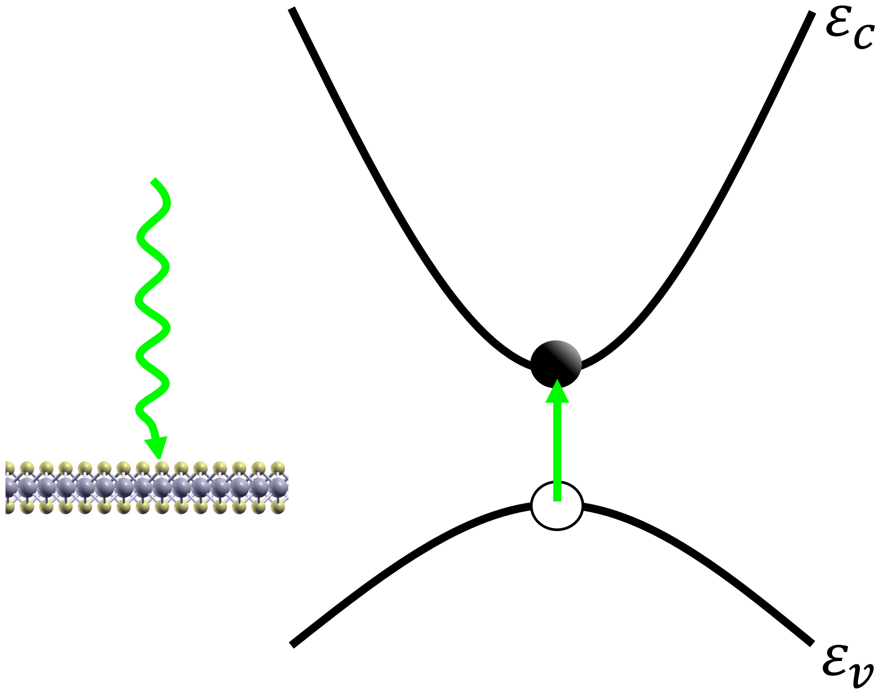
direct exchange





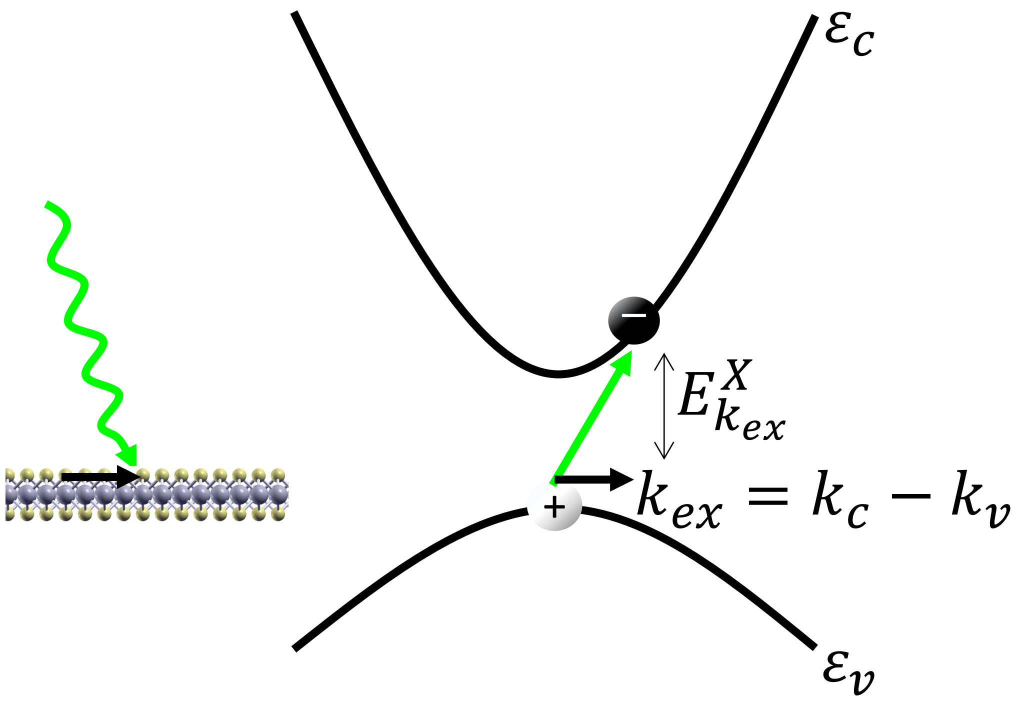
- Qiu, D. Y.; Cao, T.; Louie, S. G. PRL (2015)
- G. H. Peng, et al., ***SJC***. Nano Lett. **19**, 2299 (2019).

Exciton: a many-body problem

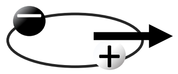


An exciton

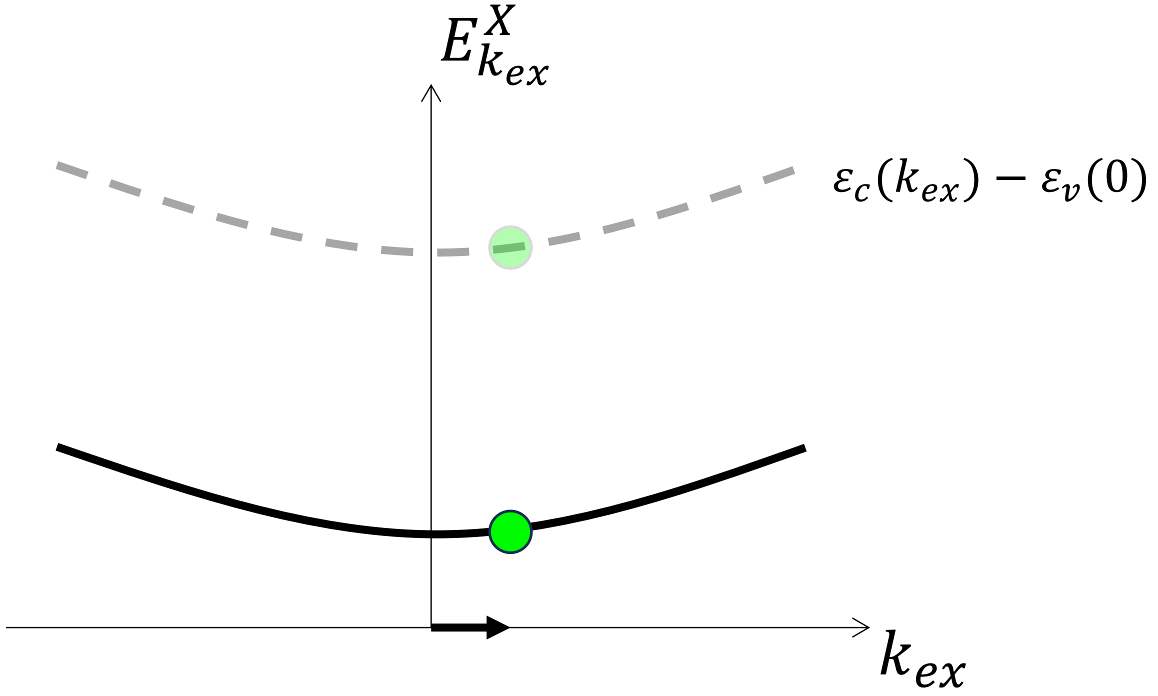
Exciton band: direct interaction



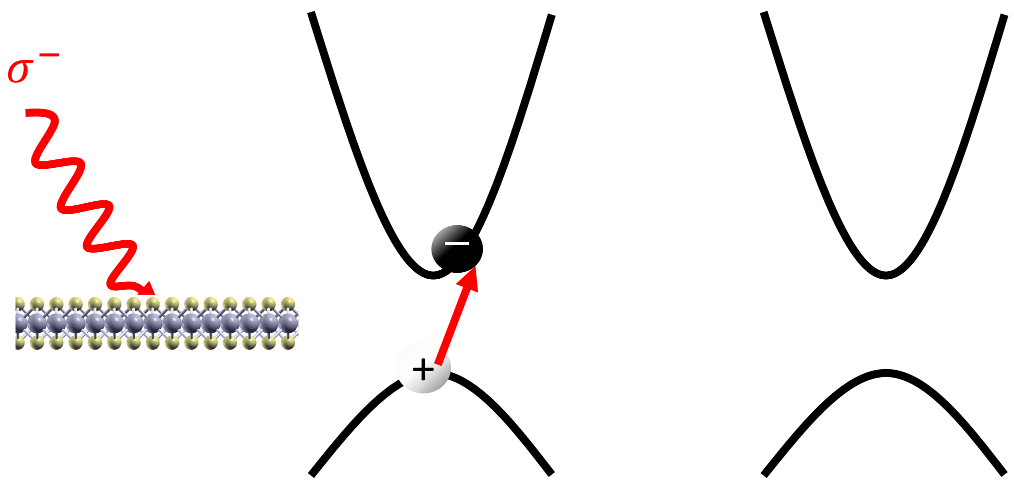
An exciton with finite momentum P_{CM}

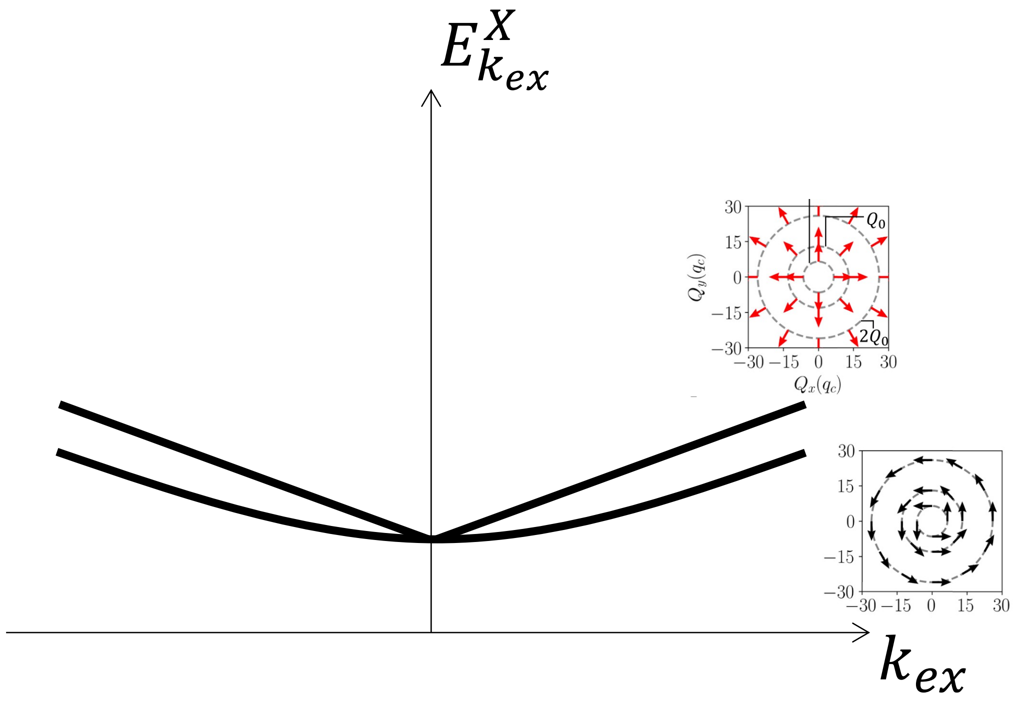


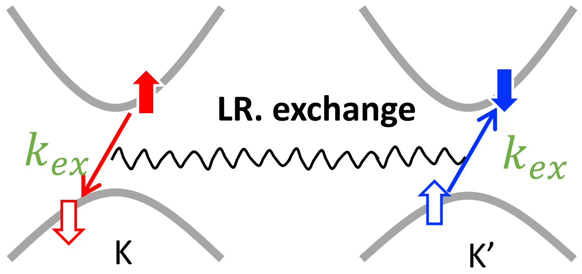
$P_{CM} = \hbar k_{ex}$



Exciton band dispersions $E^X_{k_{ex}}$ vs. k_{ex}

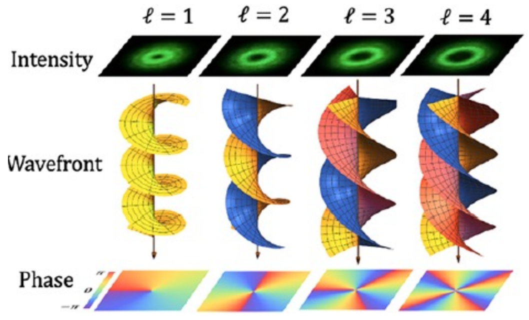

σ^-

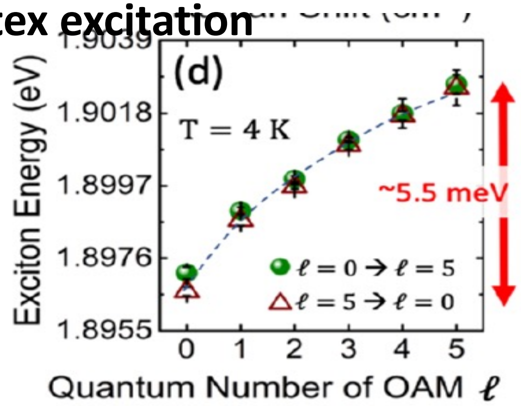

$E^X_{k_{ex}}$
k_{ex}
$Q_y(q_c)$
$Q_x(q_c)$
Q_0
$2Q_0$

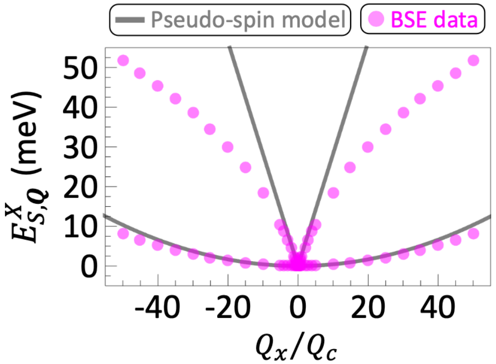

LR. exchange
k_{ex}
k_{ex}
K
K'

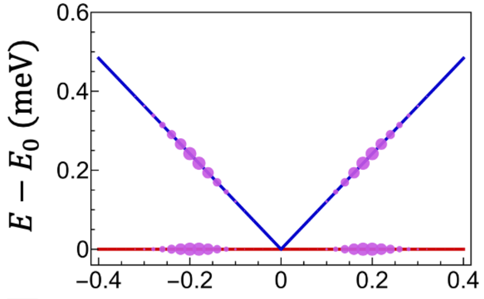
TH Lu@NTNU YW Lan@NTNU

PL using the optical vortex excitation









Electron energy loss spectra

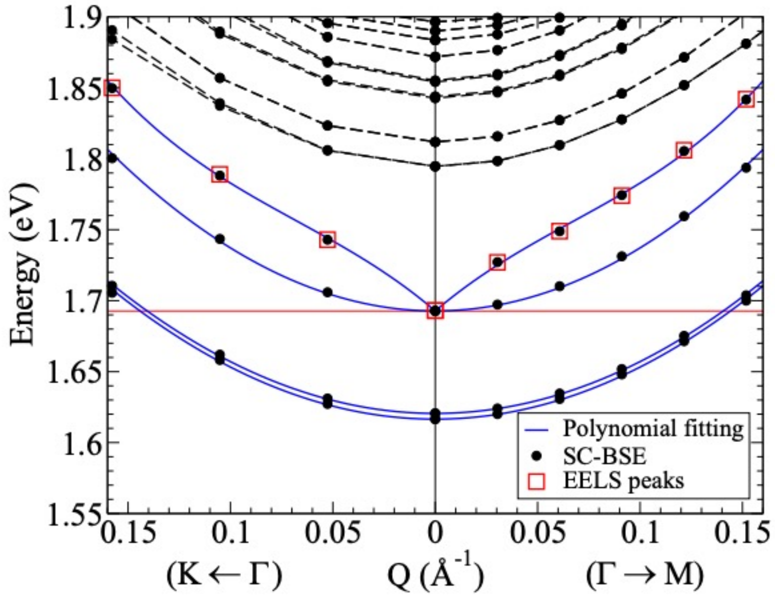
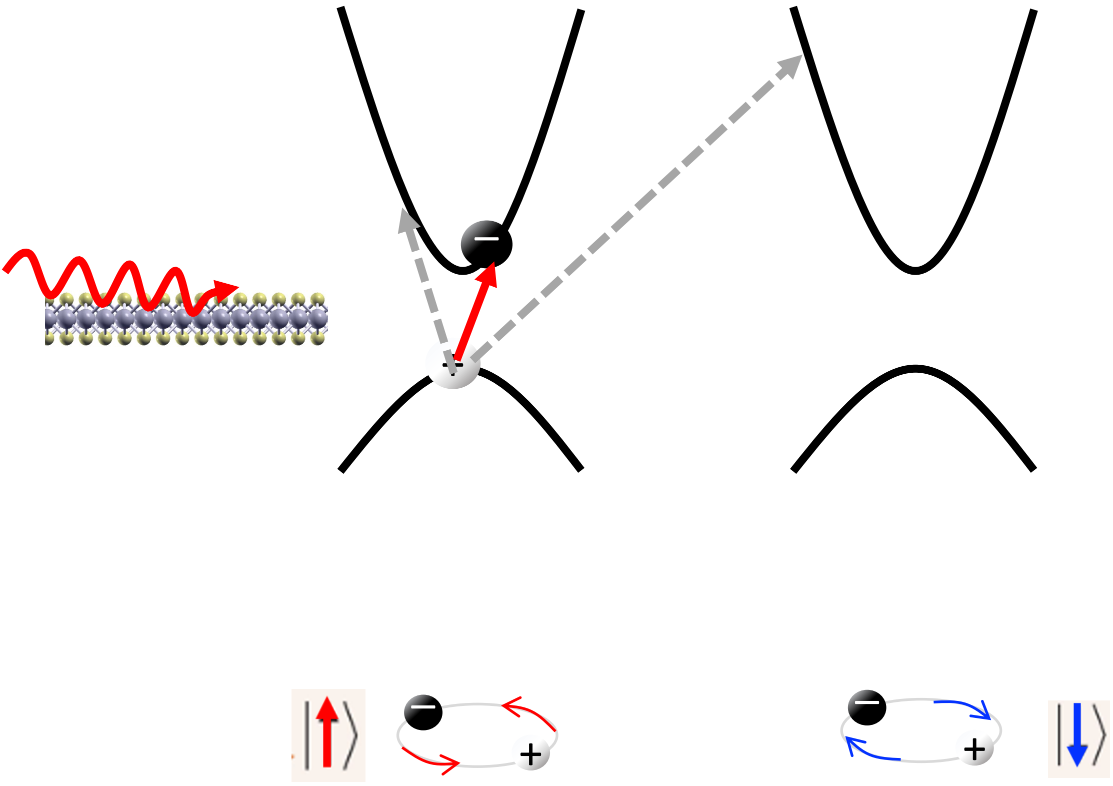
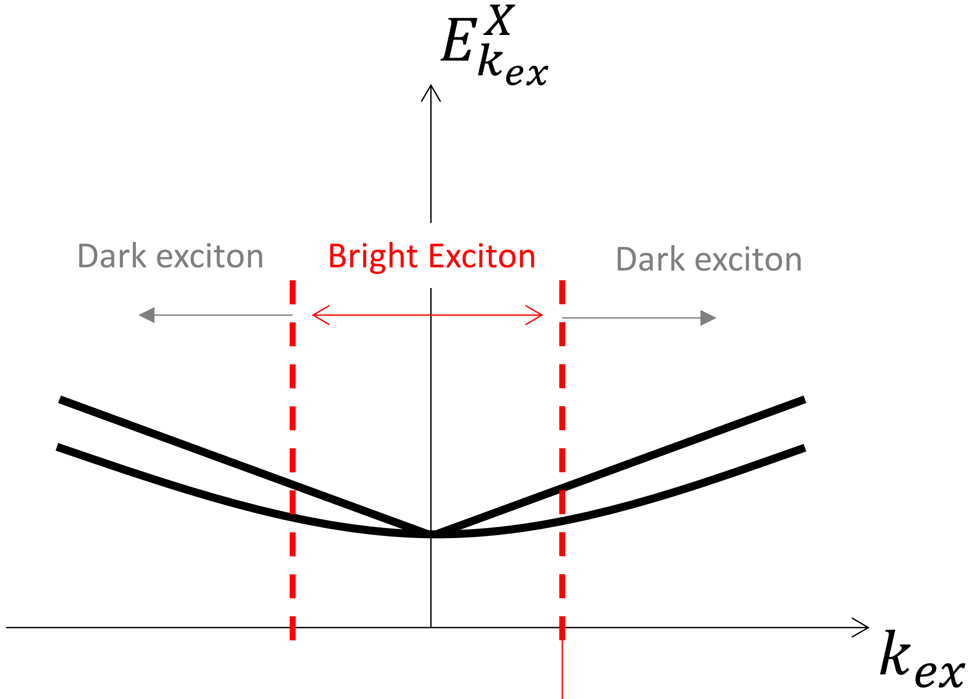


FIG. 4. The exciton band structure and the calculated EELS peaks dispersion of the A exciton of the WSe_2 monolayer. The black

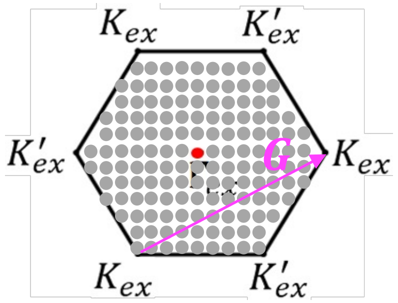
n, G. Y. Guo, PRB 110, 205417 (2024)

K Simbulan et al. SJC, THLu, YWLan, ACS Nano 15, 3481 (2021)
GH Peng, et al. SJC, PRB **106**, 155304 (2022)

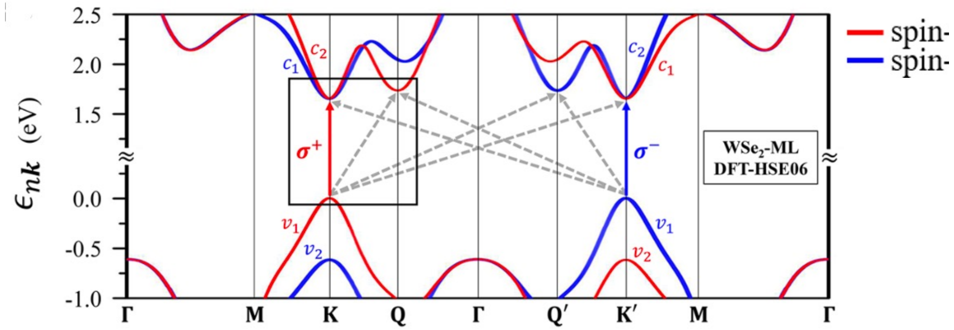


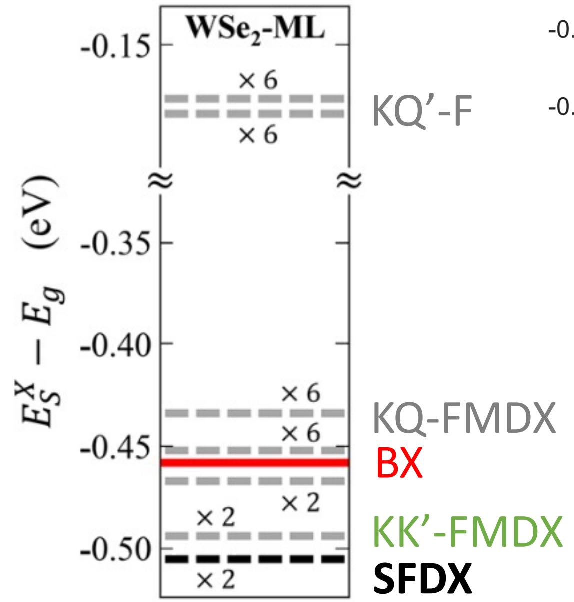


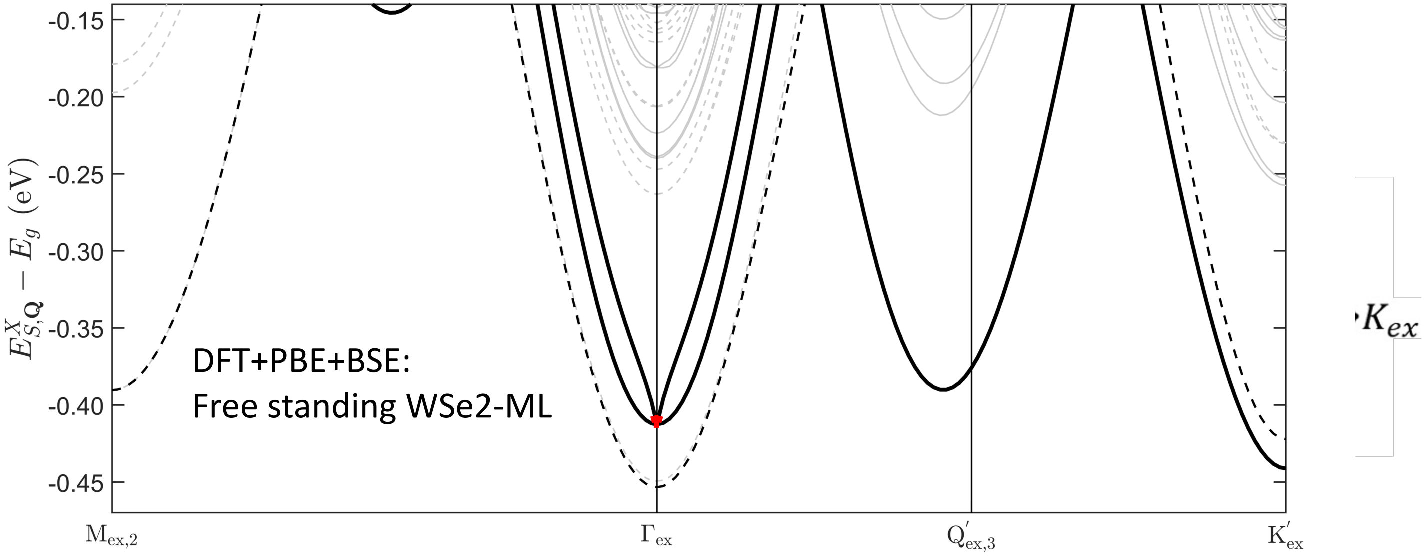
$q_c = \frac{2\pi}{\lambda} \ll G = \frac{2\pi}{a_0}$

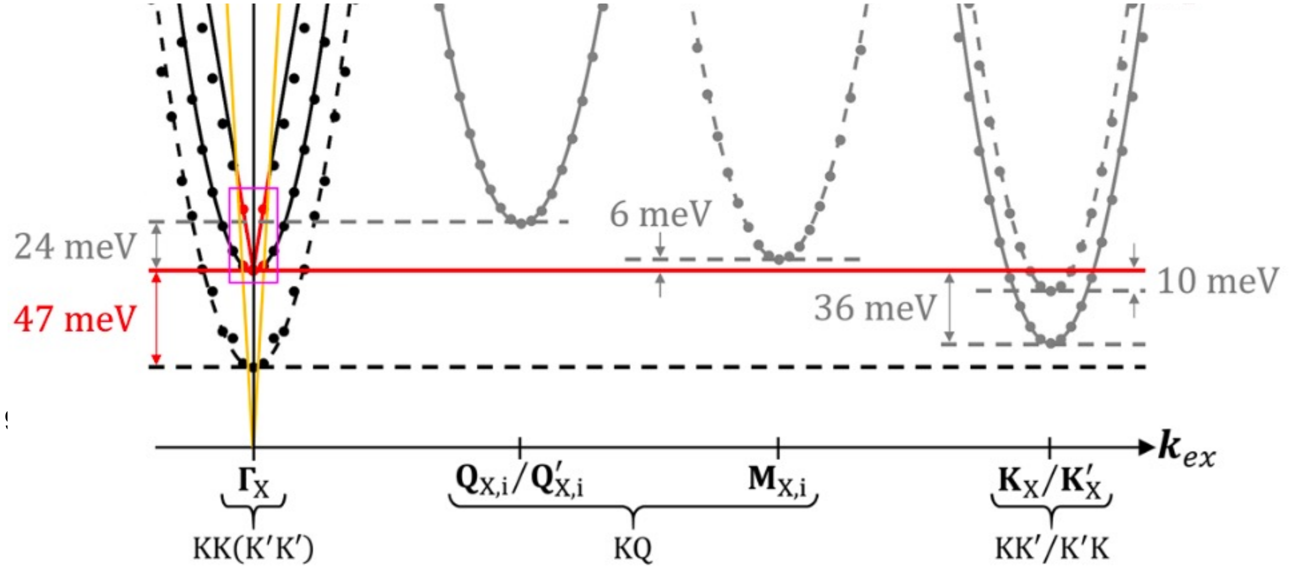


Finite momentum dark excitons (FM-DXs) in 2D TMDs





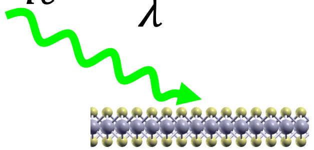


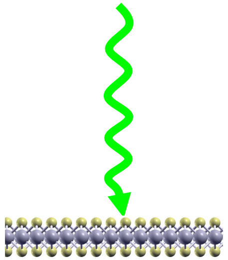


G. H. Peng,, et al. SJC, Nano Lett. **19**, 2299 (201

Finite momentum dark excitons (FM-DXs) in 2D TMDs

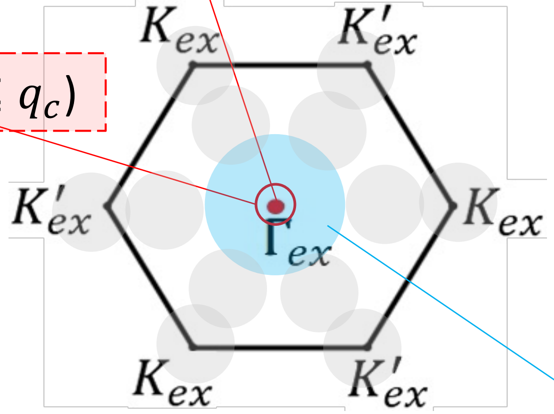
$$q_c = \frac{2\pi}{\lambda}$$





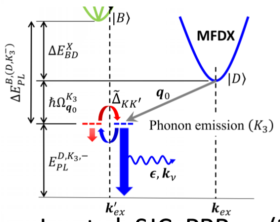
Bright exciton ($k_{ex} = 0$)

Finite-momentum bright exciton ($k_{ex} \leq q_c$)



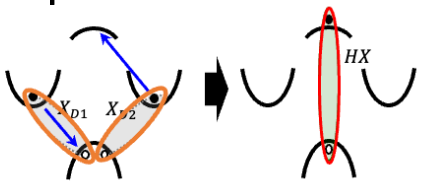
Inter-valley momentum dark exciton ($k_{ex} \gg q_c$)

Phonon-assisted PL



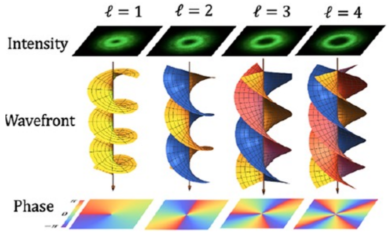
Lo et al. SJC, PRRes. (2021)

Upconversion PL via EEA

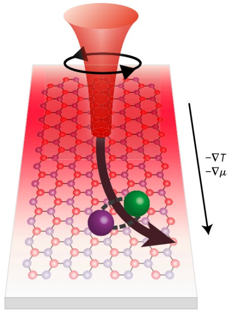


Y Chen et al. SJC, SYChen, Nat. Comm (2025)

Optical vortex excitation



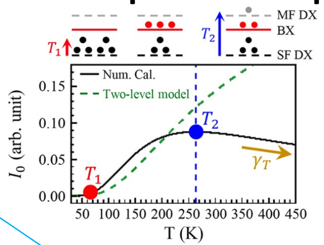
K Simbulan et al., ACS Nano (2021)



Exciton Hall effect

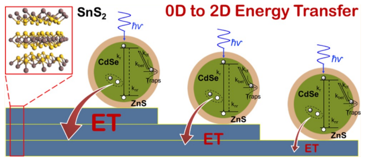
M. Onga et al. Y. Iwasa, Nature Mat. (2017)

Temperature-dependent PL



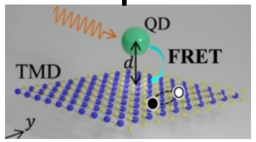
Peng,, et al., SJC, Nano Lett. (2019).

Intra-valley momentum dark exciton ($k_{ex} > q_c$)



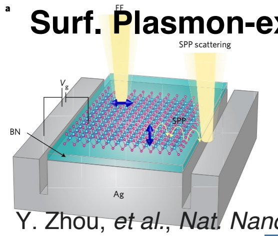
H Zang, et al., *ACS Nano*, **10**, 4790 (2016).

Nano-optics: Energy transfer



Npj: 2D Mat. **7**, 51 (2023)

Surf. Plasmon-exciton conversion



Y. Zhou, *et al*., *Nat. Nanot.* **12**, 856 (2017).

DFT-based Bethe-Salpeter equation (BSE) theory for exciton in 2D systems

VASP, **Quantum Espresso**, BerkeleyGW,…

DFT

Solve Kohn-Sham Eq. In DFT

GW

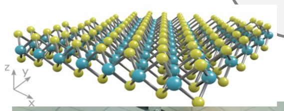
construct Green's function G

construct screening W (RPA)

Green's function Self-consistency

solve the Dyson equation

Improved spectrum



BSE

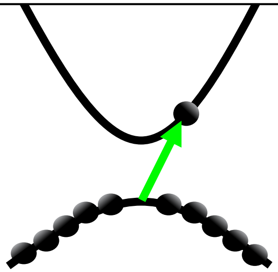
static $\epsilon((\mathbf{q}, \omega = 0)$

construct Kernel K

solve the Bethe-Salpeter equation

Antonios M. Alvertis,
"*On Exciton–Vibration and Exciton–Photon Interactions in Organic Semiconductors*", *Springer Verlag*. ISSN 2190-5053

Exciton:
A many-body ($N_{tot} = (N_v - 1) + 1$) problem



A finite-momentum ($\boldsymbol{k}_{ex}$) exciton state:

$$|S\boldsymbol{k}_{ex}\rangle = \Sigma_{\boldsymbol{k}} A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k})\hat{c}^{\dagger}_{k+k_{ex}}\hat{h}^{\dagger}_{k}|0\rangle$$

Bethe-Salpeter equation (BSE): Sham, L. J.; Rice, T. M. Phys. Rev. 144, 708 (1966).

$$[\epsilon_{c,\boldsymbol{k}+\boldsymbol{k}_{ex}} - \epsilon_{v,\boldsymbol{k}}]\, A_{S,\boldsymbol{k}_{ex}}(vc\boldsymbol{k}) + \sum_{v'c'\boldsymbol{k}'} U_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')\, A_{S,\boldsymbol{k}_{ex}}(v'c'\boldsymbol{k}') = E_{S,\boldsymbol{k}_{ex}} A_{S,\boldsymbol{k}_{ex}}(vc\boldsymbol{k})$$

Coulomb kernel:

$$U_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') = V^{d}_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') - V^{x}_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')$$

Direct interaction:

$$V^{d}_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') = \int d^3\boldsymbol{r}_1 d^3\boldsymbol{r}_2\, \psi^{*}_{c,\boldsymbol{k}+\boldsymbol{k}_{ex}}(\boldsymbol{r}_1)\, \psi_{v,\boldsymbol{k}}(\boldsymbol{r}_2)\, W(\boldsymbol{r}_1, \boldsymbol{r}_2)\, \psi^{*}_{v',\boldsymbol{k}'}(\boldsymbol{r}_2)\, \psi_{c',\boldsymbol{k}'+\boldsymbol{k}_{ex}}(\boldsymbol{r}_1)$$

Exchange interaction:

$$V^{x}_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') = \int d^3\boldsymbol{r}_1 d^3\boldsymbol{r}_2\, \psi^{*}_{c,\boldsymbol{k}+\boldsymbol{k}_{ex}}(\boldsymbol{r}_1)\, \psi_{v,\boldsymbol{k}}(\boldsymbol{r}_1)\, V(\boldsymbol{r}_1 - \boldsymbol{r}_2)\, \psi^{*}_{v',\boldsymbol{k}'}(\boldsymbol{r}_2)\, \psi_{c',\boldsymbol{k}'+\boldsymbol{k}_{ex}}(\boldsymbol{r}_2)$$

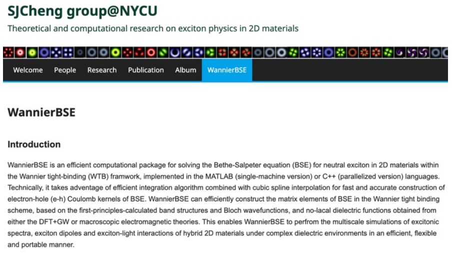
$$V(r_1 - r_2) = \frac{e^2}{4\pi\epsilon_0|r_1 - r_2|}$$

$$W(r_1 - r_2) = \int d^3r\, \epsilon^{-1}(r_1, r')V(r' - r_2)$$

$\epsilon^{-1}(r, r')$: inverse dielectric function

WannierBSE: an efficient methodology for solving a DFT-based BSE in WTB scheme

GH Peng et al SJC, Nano Lett (2019)


SJCheng group@NYCU

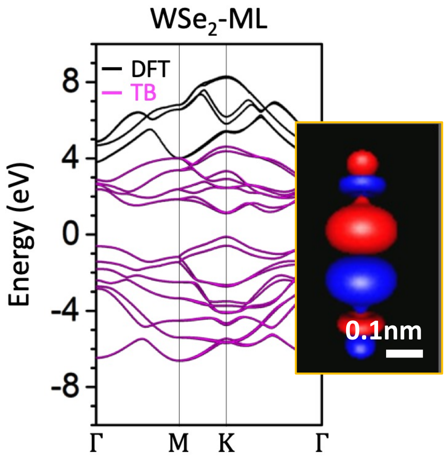
Theoretical and computational research on exciton physics in 2D materials

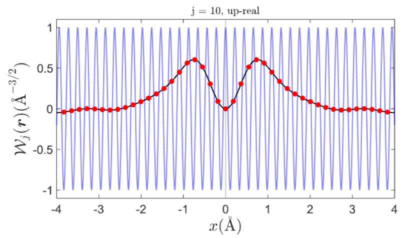
Welcome | People | Research | Publication | Album | WannierBSE

WannierBSE

Introduction

WannierBSE is an efficient computational package for solving the Bethe-Salpeter equation (BSE) for neutral exciton in 2D materials within the Wannier tight-binding (WTB) framwork, implemented in the MATLAB (single-machine version) or C++ (parallelized version) languages. Technically, it takes adventage of efficient integration algorithm combined with cubic spline interpolation for fast and accurate construction of electron-hole (e-h) Coulomb kernels of BSE. WannierBSE can efficiently construct the matrix elements of BSE in the Wannier tight binding scheme, based on the first-principles-calculated band structures and Bloch wavefunctions, and no-lacal dielectric functions obtained from either the DFT+GW or macroscopic electromagnetic theories. This enables WannierBSE to perfrom the multiscale simulations of excitonic spectra, exciton dipoles and exciton-light interactions of hybrid 2D materials under complex dielectric environments in an efficient, flexible and portable manner.





DFT calculation (QE, VASP,…)

↓

Wannierization (Wannier90)
1. Max. Localized Wannier func.
2. Wannier Tight-binding model

↓

$$H^{KS}|\psi_{n,\boldsymbol{k}}\rangle = \epsilon_{n\boldsymbol{k}}|\psi_{n,\boldsymbol{k}}\rangle$$

$$|\mathcal{W}_{j\boldsymbol{R}}\rangle \equiv \frac{1}{\sqrt{N}}\sum_{\boldsymbol{k}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}}\sum_{n} U_{ni}^{(\boldsymbol{k})}|\psi_{n,\boldsymbol{k}}\rangle$$

$$H_{ij}^{TB}(\boldsymbol{k}) = \sum_{\boldsymbol{R}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}} t_{ij}(\boldsymbol{R})$$

$$t_{ij}(\boldsymbol{R}) = \langle\mathcal{W}_{j\boldsymbol{R}=\boldsymbol{0}}|H^{KS}|\mathcal{W}_{j\boldsymbol{R}}\rangle$$

↓

Discretized with non-uniform $\boldsymbol{k}$-grids

$\boldsymbol{k} = \{\boldsymbol{k}_1, \boldsymbol{k}_2, \dots\}$

↓

Efficient Adaptive integrations

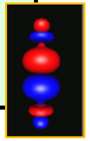
$$\mathcal{W}_{j\boldsymbol{R}} \rightarrow \varrho_{j'j}^{(\boldsymbol{R})} \rightarrow \Xi_{j'j\ell'\ell}^{\boldsymbol{R},\boldsymbol{R}'}$$

$$U_{\boldsymbol{k}_{ex}} = V_{\boldsymbol{k}_{ex}}^{d} + V_{\boldsymbol{k}_{ex}}^{x}$$

Coulomb kernel

Exchange term

$$C_j^{(n)}(\boldsymbol{k}), e^{i\boldsymbol{k}\cdot\boldsymbol{R}}, \Xi_{j'j\ell'\ell}^{\boldsymbol{R},\boldsymbol{R}'}$$
$$\downarrow$$
$$V_{\boldsymbol{k}_{ex}}^{x}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')$$

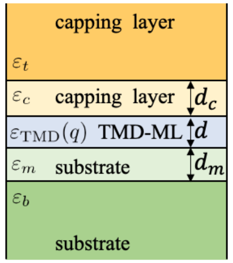


Screened Direct term

$$C_j^{(n)}(\boldsymbol{k}), e^{i\boldsymbol{k}\cdot\boldsymbol{\tau}_j}, \varepsilon(\boldsymbol{q})$$
$$\downarrow$$
$$V_{\boldsymbol{k}_{ex}}^{d}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')$$

Multi-scaled electromagnetic theory

$\varepsilon(\boldsymbol{q})$



Bethe-Salpeter equation (BSE)

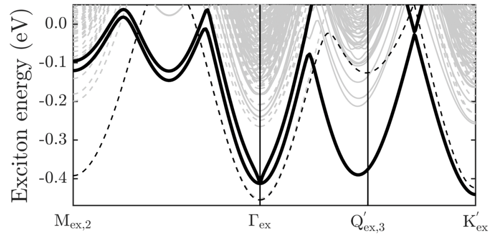
$$|S\boldsymbol{k}_{ex}\rangle = \Sigma_{\boldsymbol{k}} A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k})\hat{c}_{k+k_{ex}}^{\dagger}\hat{h}_{k}^{\dagger}|0\rangle$$

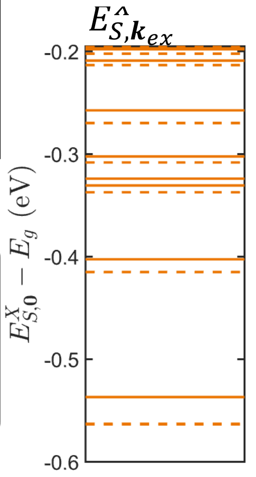
$$(\epsilon_{c\boldsymbol{k}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{k}_{ex}}(vc\boldsymbol{k}) - \frac{1}{\mathcal{A}}\sum_{v'c'\boldsymbol{k}'} U_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')A_{S\boldsymbol{k}_{ex}}(v'c'\boldsymbol{k}') = E_{S\boldsymbol{k}_{ex}}^{X} A_{S\boldsymbol{k}_{ex}}(vc\boldsymbol{k})$$

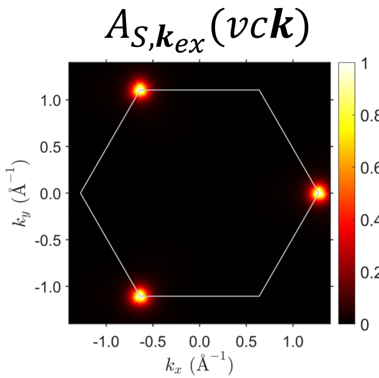
↓

Diagonalization of BSE matrix

Output:
1. **Exciton bands: $E_{S,\boldsymbol{k}_{ex}}^{X}$**
2. **Exciton wave func. $A_{S,\boldsymbol{k}_{ex}}(vc\boldsymbol{k})$**
3. **Exciton dipoles**





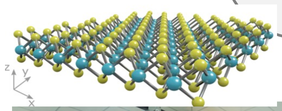


DFT-BSE vs. *DFT-WannierBSE*

VASP, Quantum Espresso, BerkeleyGW,…

DFT — Solve Kohn-Sham Eq. In DFT

GW
- construct Green's function G
- construct screening W (RPA)
- solve the Dyson equation
- Green's function Self-consistency
- Improved spectrum

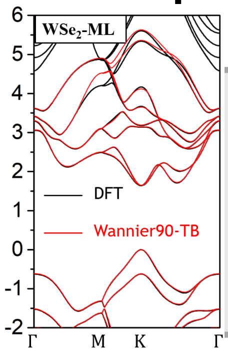


BSE
- static $\epsilon((\mathbf{q}, \omega = 0)$
- construct Kernel K
- solve the Bethe-Salpeter equation

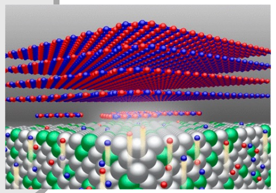
Antonios M. Alvertis,
"*On Exciton–Vibration and Exciton–Photon Interactions in Organic Semiconductors*", *Springer Verlag*. ISSN 2190-5053

DFT — Solve Kohn-Sham Eq. In DFT

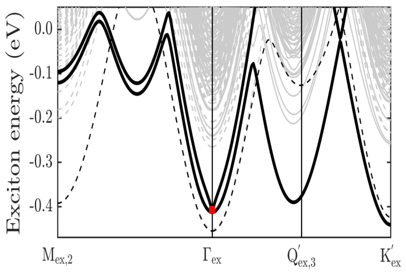
(GW) — GW (Optional)



WTB
- Wannierization (WANNIER90)
- Wannier Tight Binding (WTB)



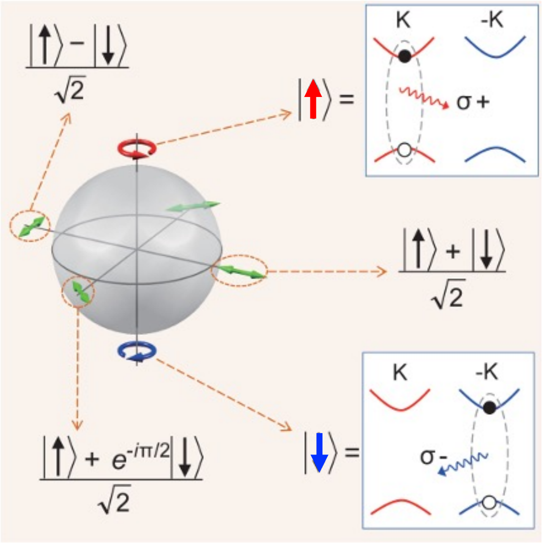
WannierBSE
- $\epsilon(\mathbf{q}, \omega = 0)$ ← Classical EM theory
- K (adaptive integration algorithm)
- Solve BSE



https://quantum.web.nycu.edu.tw/wannierbse/

G. H. Peng, P Y. Lo, et al. SJC, Nano Lett. (2019)

Momentum-dependent valley polarization (pseudo-spin) of valley exciton



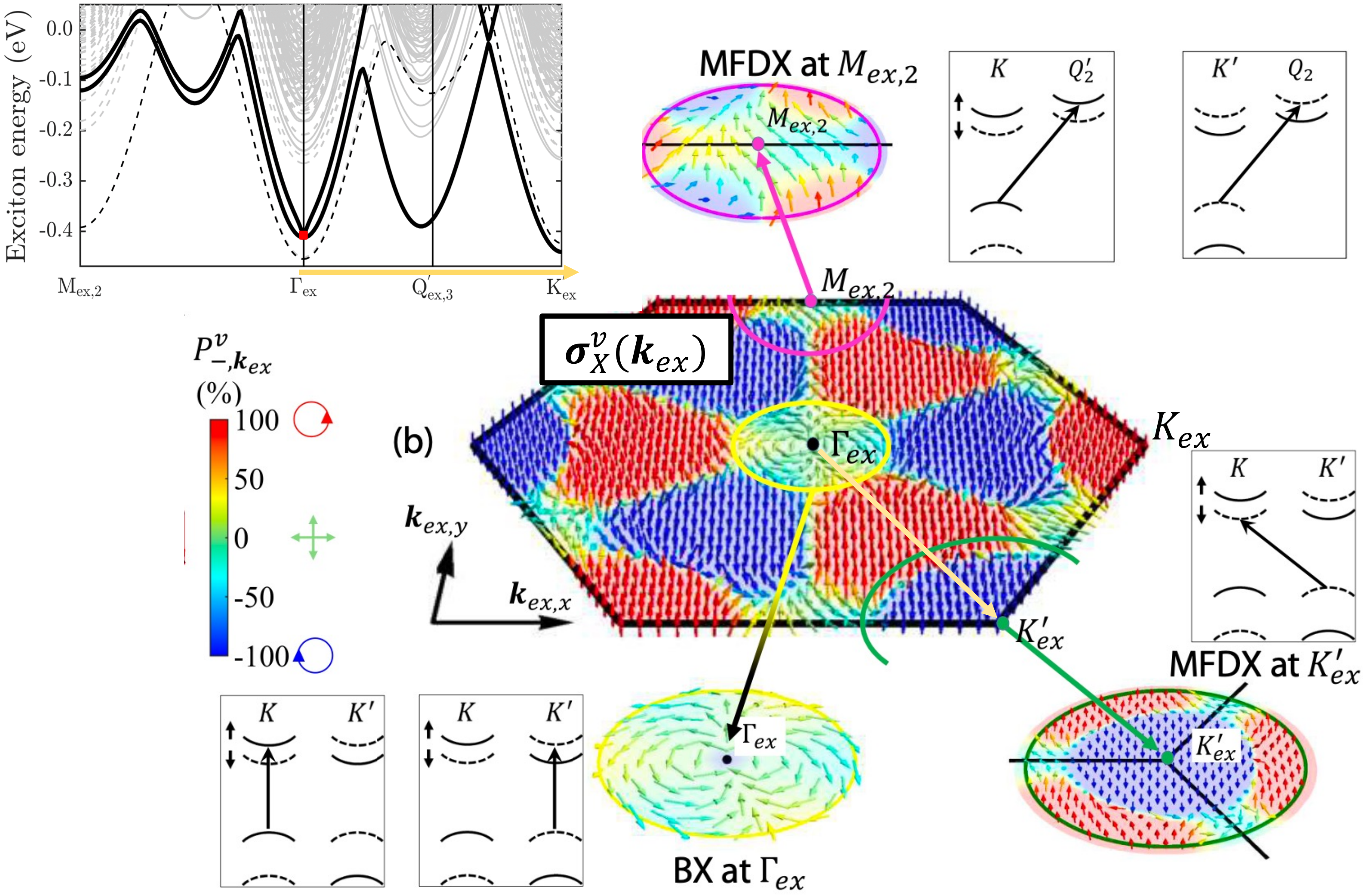
Fit the DFT-WannierBSE-calculated FM-DX states to the pseudo-spin model:

$$|S;\boldsymbol{k}_{ex}\rangle \approx \alpha_S^K(\boldsymbol{k}_{ex})|K;\boldsymbol{k}_{ex}\rangle + \alpha_S^{K'}(\boldsymbol{k}_{ex})|K';\boldsymbol{k}_{ex}\rangle$$

$$\boxed{H_{eff}^X(\boldsymbol{k}_{ex}) = \boldsymbol{\Omega}_X(\boldsymbol{k}_{ex}) \cdot \boldsymbol{\sigma}_X^v(\boldsymbol{k}_{ex})}$$

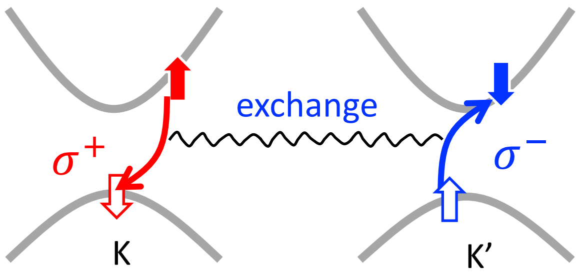
$\boldsymbol{\sigma}_X^v$: valley pseudo-spin of k_{ex}-exciton

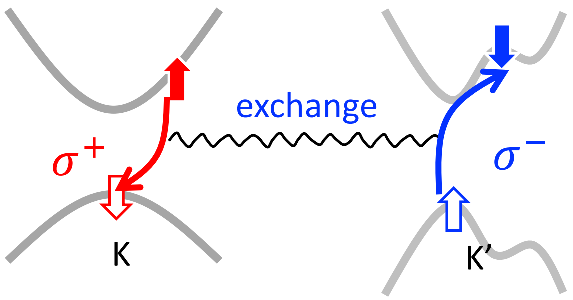
Valley polarization: $P_{sk_{ex}}^v \equiv \frac{\alpha_S^K - \alpha_S^{K'}}{\alpha_S^K + \alpha_S^{K'}}$



PY Lo, et al. SJC, P.R. Research 3, 043198 (2021)

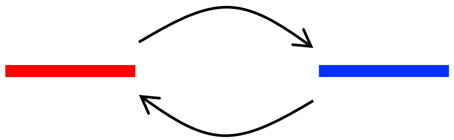
Electron-hole exchange interaction: intervalley transfer of valley exciton

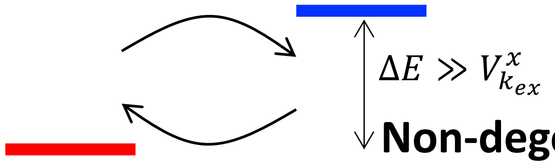




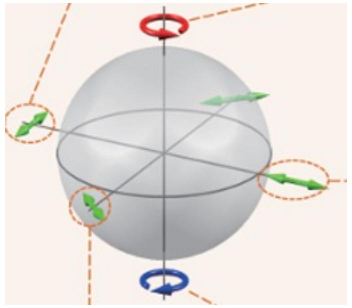
$V^{x,LR}_{k_{ex}} \sim 1\text{meV}$

Degenerate valley states





Non-degenerate valley states

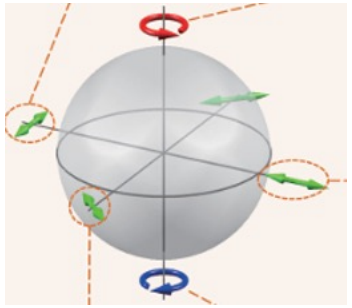


$|\pm\rangle \propto (|{\color{red}\Uparrow\Downarrow}\rangle \pm |{\color{blue}\Uparrow\Downarrow}\rangle)$

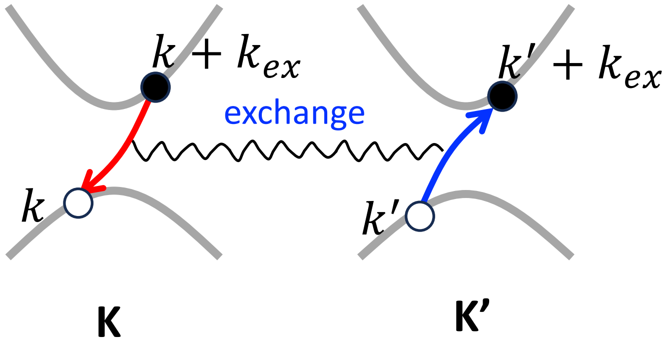
$(\sigma^+ \;\pm\; \sigma^-) \propto \boldsymbol{\pi_{x/y}}$

$|-\rangle \approx |{\color{red}\Uparrow\Downarrow}\rangle \quad {\color{red}\sigma^+}$

$|+\rangle \approx |{\color{blue}\Uparrow\Downarrow}\rangle \quad {\color{blue}\sigma^-}$



Momentum-dependent valley depolarization: symmetry analysis

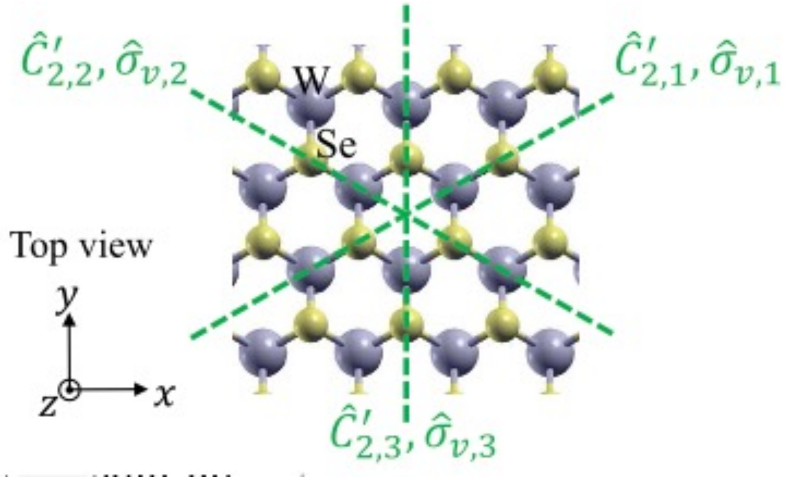


So, Let's derive the condition for the degeneracy of finite-momentum excited states:

$$\epsilon_{c,\boldsymbol{k}+\boldsymbol{k}_{\mathrm{ex}}} - \epsilon_{v,\boldsymbol{k}} = \epsilon_{c,\boldsymbol{k}'+\boldsymbol{k}_{\mathrm{ex}}} - \epsilon_{v,\boldsymbol{k}'}$$

TMD-MLs : $D_{3h} = \{E, C_3, C_3^{-1}, \sigma_h, S_3, S_3^{-1}, C'_{2,1}, C'_{2,2}, C'_{2,3}, \sigma_{v,1}, \sigma_{v,2}, \sigma_{v,3}\}$

Symmetry of 2D TMD ensures : $\epsilon_{n,U\boldsymbol{k}} = \epsilon_{n,\boldsymbol{k}}$ with $U \in Q = D_{3h}$

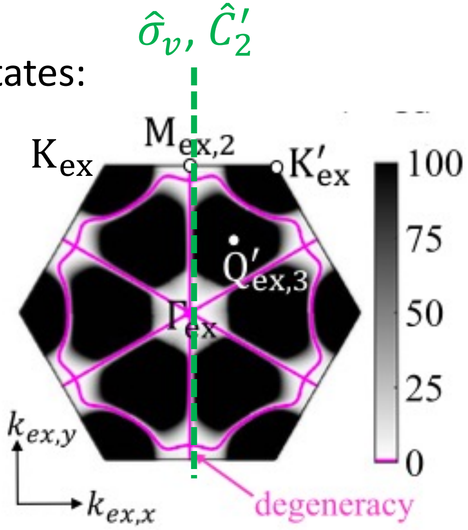


From $\epsilon_{v,\boldsymbol{k}} = \epsilon_{v,\boldsymbol{k}'}$ and $\epsilon_{c,\boldsymbol{k}+\boldsymbol{k}_{\mathrm{ex}}} = \epsilon_{c,\boldsymbol{k}'+\boldsymbol{k}_{\mathrm{ex}}}$
We find the condition of degeneracy of momentum exciton states:

$$\boxed{\boldsymbol{k}' = U\boldsymbol{k} \quad \text{and} \quad \boldsymbol{k}_{\mathrm{ex}} = U\boldsymbol{k}_{\mathrm{ex}}}$$

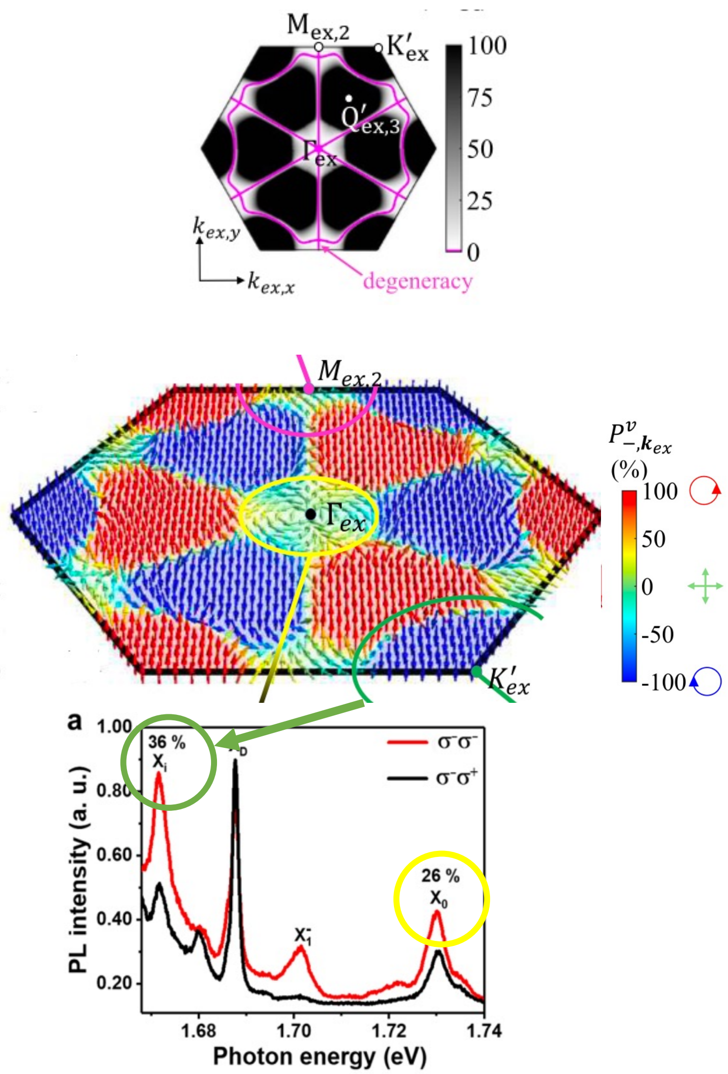
Accordingly, we find that the finite momentum exciton States should be degenerate only if

$$\boldsymbol{k}_{\mathrm{ex}} \in \overline{\Gamma_{\mathrm{ex}}\mathrm{M}_{\mathrm{ex},i}} \text{ with } i = 1,2,3$$



P.-Y. Lo, G.-H. Peng, et al SJC, Phys. Rev. Research **3**, 043198 (2021).

Optical polarization of Phonon-assisted PL from intervalley DXs



In the 2nd-order perturbation theory, the phonon-assisted indirect transition rate reads:

$$\Gamma_i^{(2)} = \frac{2\pi}{\hbar}\sum_f \left|\sum_m \frac{\langle f|\hat{H}_{X-\nu}|m\rangle\langle m|\hat{H}_{X-ph}|i\rangle}{E_i - E_m + i0^+}\right|^2 \delta\left(E_f - E_i\right),$$

Exciton-phonon interaction:

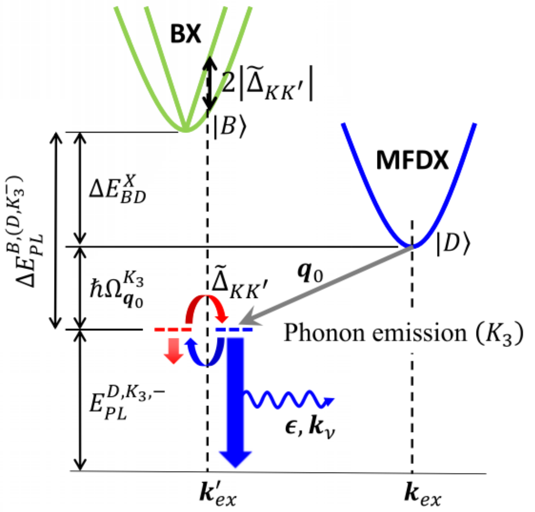
$$\hat{H}_{X-ph} = \sum_{S'\boldsymbol{k}'_{ex}}\sum_{S\boldsymbol{k}_{ex}}\sum_{\lambda\boldsymbol{q}} \hbar g^{\lambda,\boldsymbol{q}}_{S',\boldsymbol{k}'_{ex};S,\boldsymbol{k}_{ex}} \hat{X}^{\dagger}_{S',\boldsymbol{k}'_{ex}}\hat{X}_{S,\boldsymbol{k}_{ex}}\left(\hat{b}_{\lambda,\boldsymbol{q}} + \hat{b}^{\dagger}_{\lambda,\boldsymbol{q}}\right)$$

optical polarization of an indirect phonon-assisted transition via the BX intermediate states $P_D^{o(2)}$ can be related to the valley polarization of the initial MFDX state P_D^v by

negligible

$$P_D^{o(2)} \approx P_D^v\{1 - \cancel{4\mathrm{Re}[\widetilde{\beta}_{BD}^{KK'}(\boldsymbol{k}_{v,\parallel})(\alpha_D^K)^*\alpha_D^{K'}]}\},$$

where $\widetilde{\beta}_{BD}^{KK'} \equiv -\frac{\widetilde{\Delta}_{KK'}(\boldsymbol{k}_{v,\parallel})}{\Delta E_{BD}^X + \hbar\Omega_{\boldsymbol{q}_0}^{K_3}}$ arises from the EHEI $\widetilde{\Delta}_{KK'}$



For 2D TMD, $|\widetilde{\boldsymbol{\beta}}_{\boldsymbol{BD}}^{\boldsymbol{KK'}}| \sim \mathbf{10^{-2}}$ is negligible, with $|\widetilde{\Delta}_{BD}^{KK'}| \sim$ 1meV and $\Delta E_{BD}^X + \hbar\Omega \approx$ 55meV

$P_D^{o(2)} \equiv \frac{I_D^{(2)}(\epsilon_+) - I_D^{(2)}(\epsilon_+)}{I_D^{(2)}(\epsilon_+) + I_D^{(2)}(\epsilon_+)}$ is the degree of optical **polarization of the indirect PL**, $I_D^{(2)} \propto \Gamma_D^{(2)}$, of $|D\rangle$.

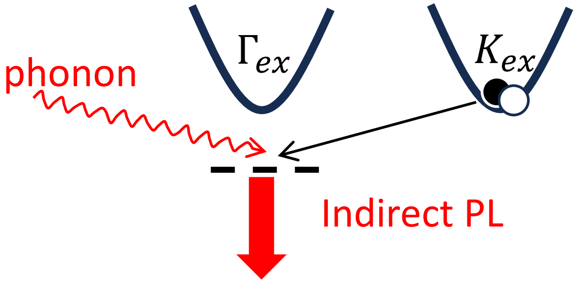
P_D^v is the degree of **valley polarization** of the intervalley momentum dark state $|D\rangle$.

$$\Rightarrow \boxed{P_D^{o(2)} \approx P_D^v}$$

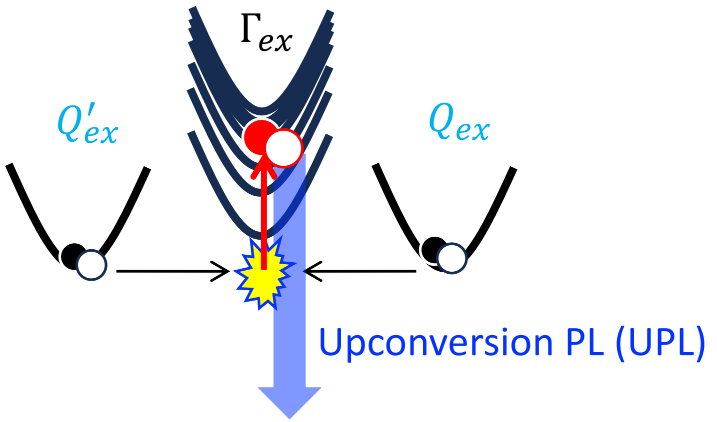
P.-Y. Lo, G.-H. Peng, et al SJC, Phys. Rev. Research **3**, 043198 (2021).

Efficient up-conversion PL via exciton-exciton annihilation (EEA)

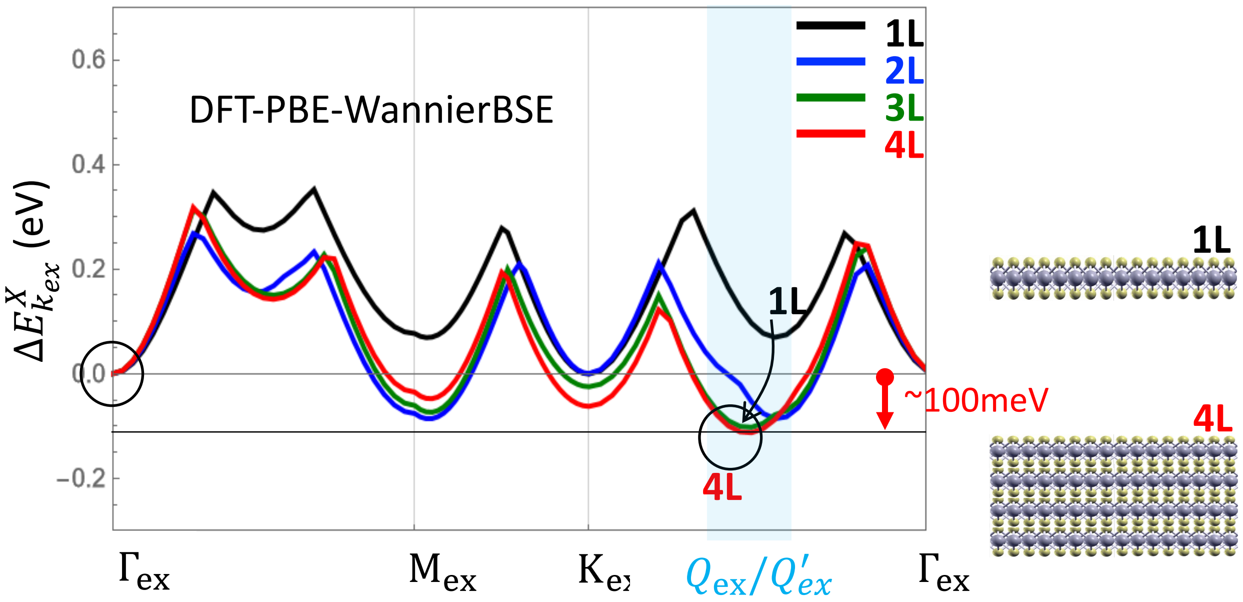
Phonon-assisted PL from FMDXs



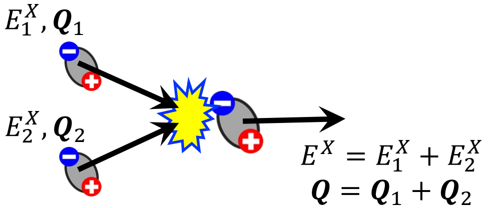
UPL from FMDXs via EEA

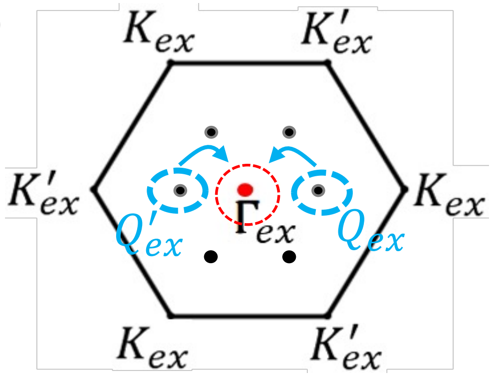


Full-zone exciton bands of multilayer WSe2

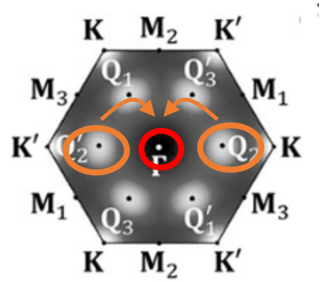


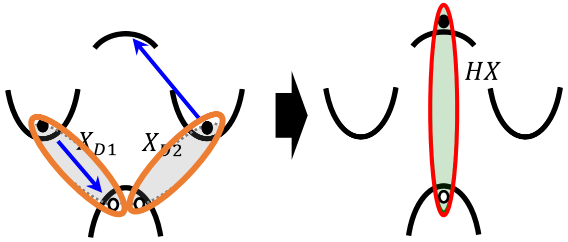
Exciton-exciton annihilation (EEA)





Efficient up-conversion PL (UPL) of intervalley FMDXs via EEA





X-XX-coupling Hamiltonian:

$$H^X = H_C^X + H_{NC}^X$$

P. Hawrylak *et al.*, PRB **54**, 11397 (1996)

M, Korkusinski, O Voznyy, PH. PRB (2011)

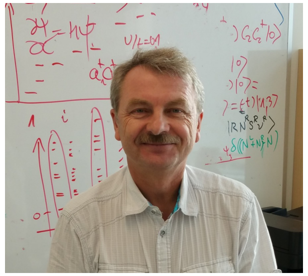
Effective EEA-Hamiltonian:

$$\hat{H}_X^{\mathrm{EEA}} = \frac{1}{2}\sum_{S,S_1,S_2}\sum_{\mathbf{Q}_1,\mathbf{Q}_2} W^{X,\mathrm{EEA}}_{S\mathbf{Q};S_1\mathbf{Q}_1,S_2\mathbf{Q}_2}\hat{X}^\dagger_{S\mathbf{Q}}\hat{X}_{S_1\mathbf{Q}_1}\hat{X}_{S_2\mathbf{Q}_2} + h.c.,$$

$$\Gamma^{\mathrm{EEA}}_{S\mathbf{Q};S_1\mathbf{Q}_1,S_2\mathbf{Q}_2} = \frac{2\pi}{\hbar}\left|W^{X,\mathrm{EEA}}_{S\mathbf{Q};S_1\mathbf{Q}_1,S_2\mathbf{Q}_2}\right|^2 \bar{n}^X_{S_1\mathbf{Q}_1}\bar{n}^X_{S_2\mathbf{Q}_2}\,\delta\left(E^{HX}_{S\mathbf{Q}} - E^X_{S_1\mathbf{Q}_1} - E^X_{S_2\mathbf{Q}_2}\right).$$

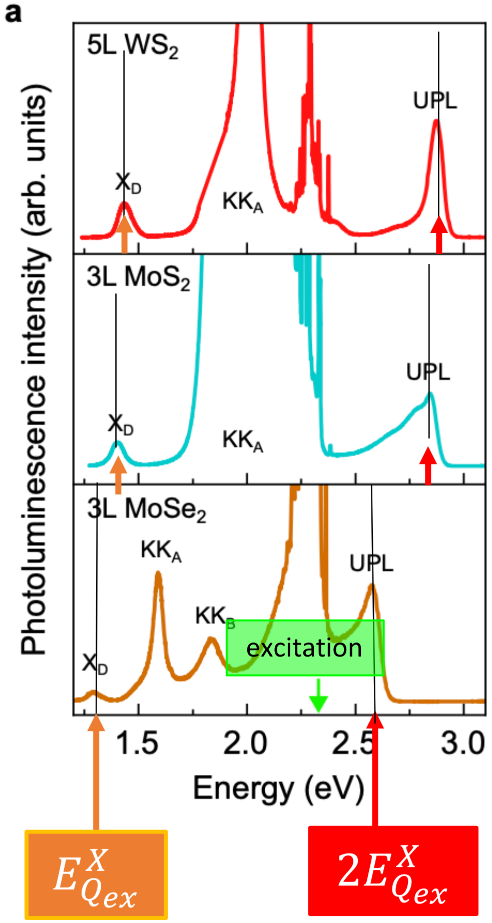
WannierBSE

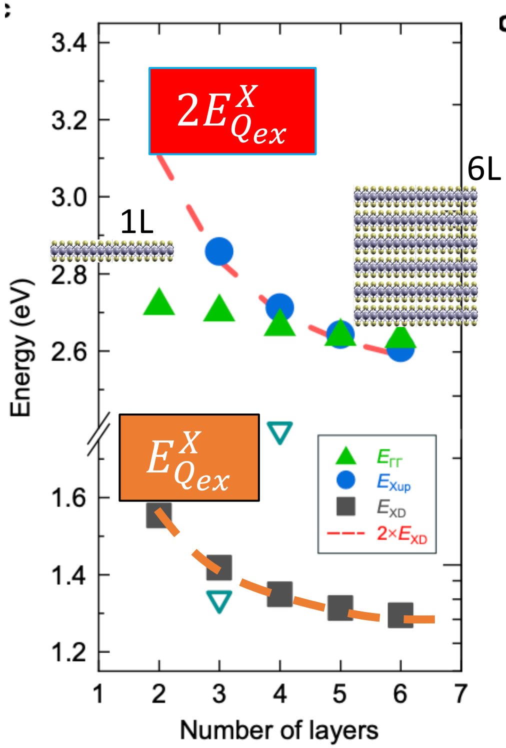

PYLo@NYCU

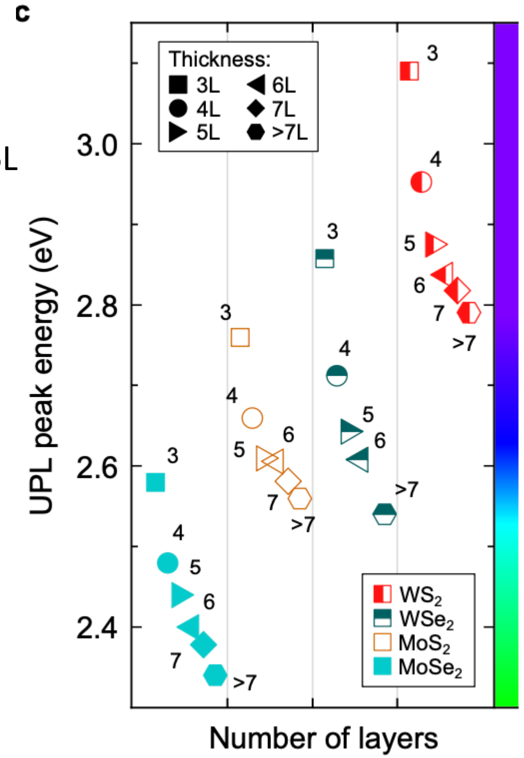

P Hawrylak@UOttawa


SY Chen@NTU


YH Chen
@Monash U





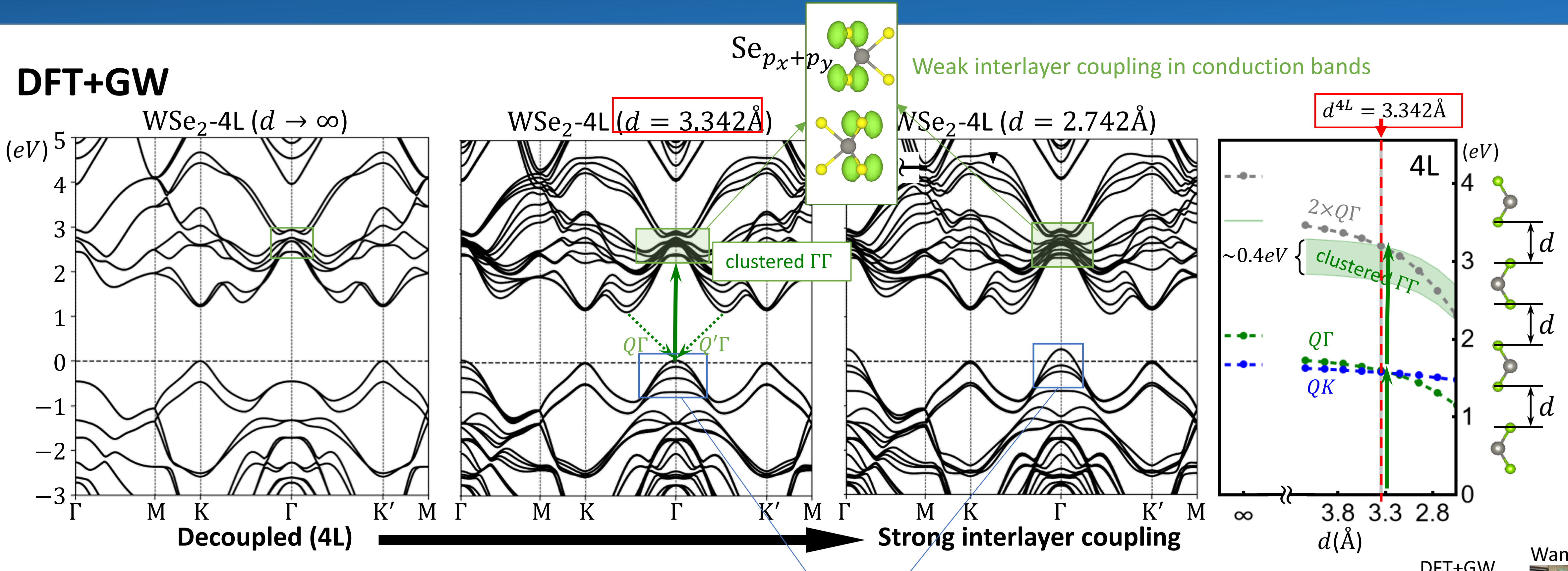


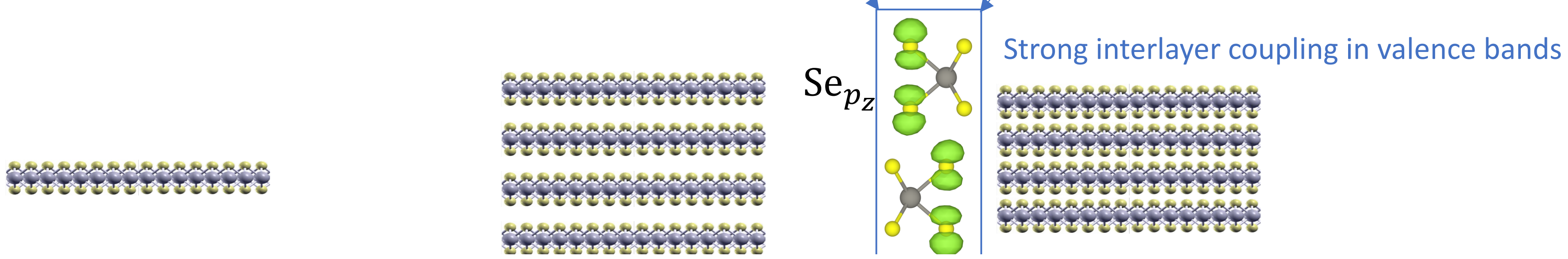
- The UPL energy is 2x of that of the weak indirect PL from intervalley dark excitons.
- The UPL emerges as the thickness of TMDs >3L.
- The upconversion efficiency increases as increasing the thickness of TMDs.
- The UPL features are generic for all multi-layer TMDs.

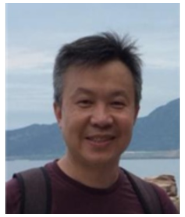
YHC, PYL,…, P Hawrylak, HCH,..,SJC, M Fuhrer, SYC Nat Comm.16,2935 (2025)

UPL of multi-layer TMDs: $2\times Q\Gamma \rightarrow \Gamma\Gamma$

DFT+GW







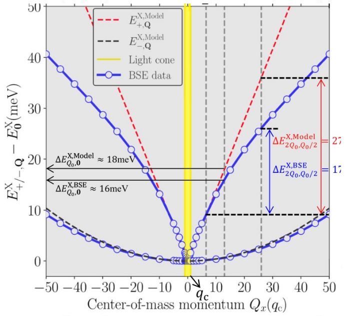
YHC, PYL,…, P Hawrylak, HCH,..,SJC, M Fuhrer, SYC Nat Comm.16,2935 (2025)

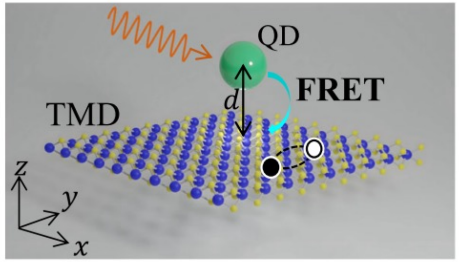
Essential role of intravalley FM-DX in nano-optics: High efficient energy transfers

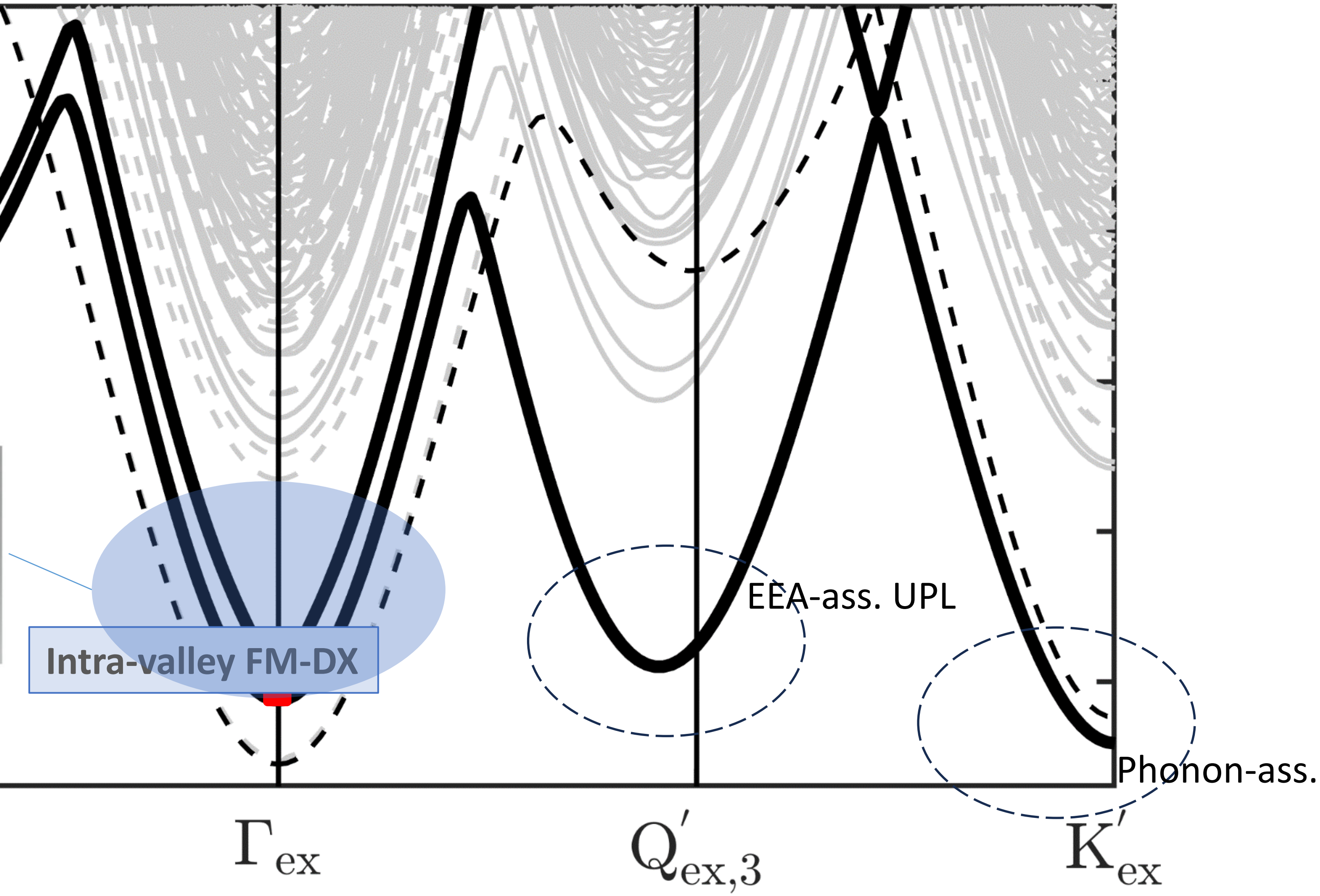
CWLuo@NYCU CLLin@NYCU JD Lin, YCTai@NYCU

- J.D. Lin et al. SJC, Npj 2D Mat. (2023)
- YC Tai, et al, SJC, CW Luo, Nano Lett. (2023)

DX-mediated fast energy transfer

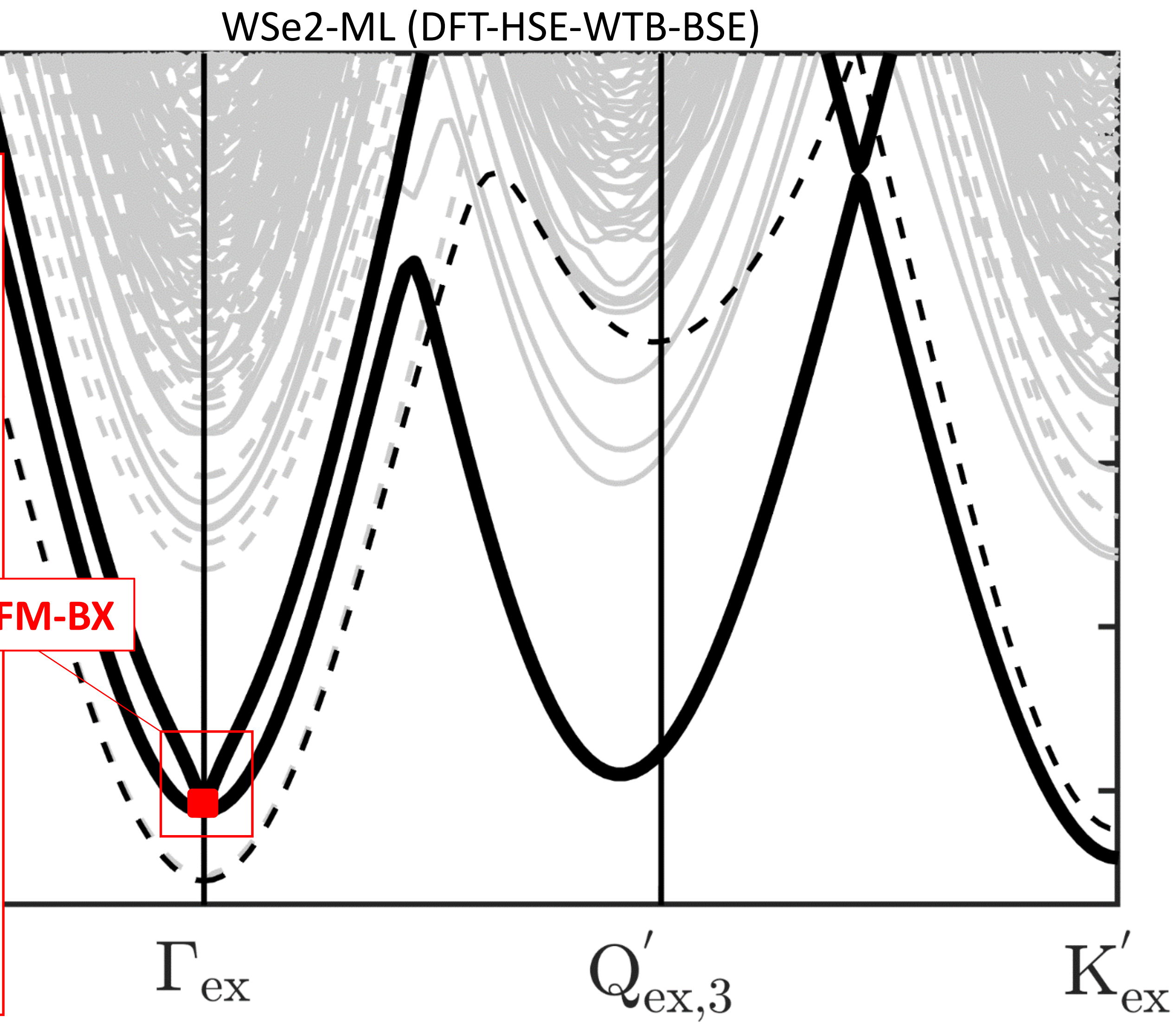




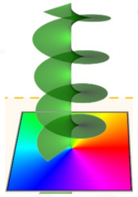


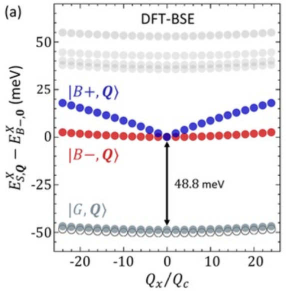
Twisted-light-controlled photoexcitation of intra-valley FM-BXs

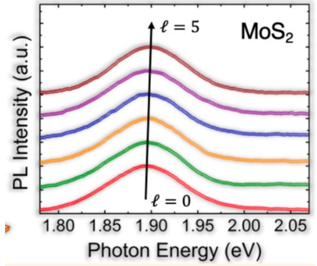
GH Peng et al SJC, Nano Lett (2019)



Twisted light with OAM







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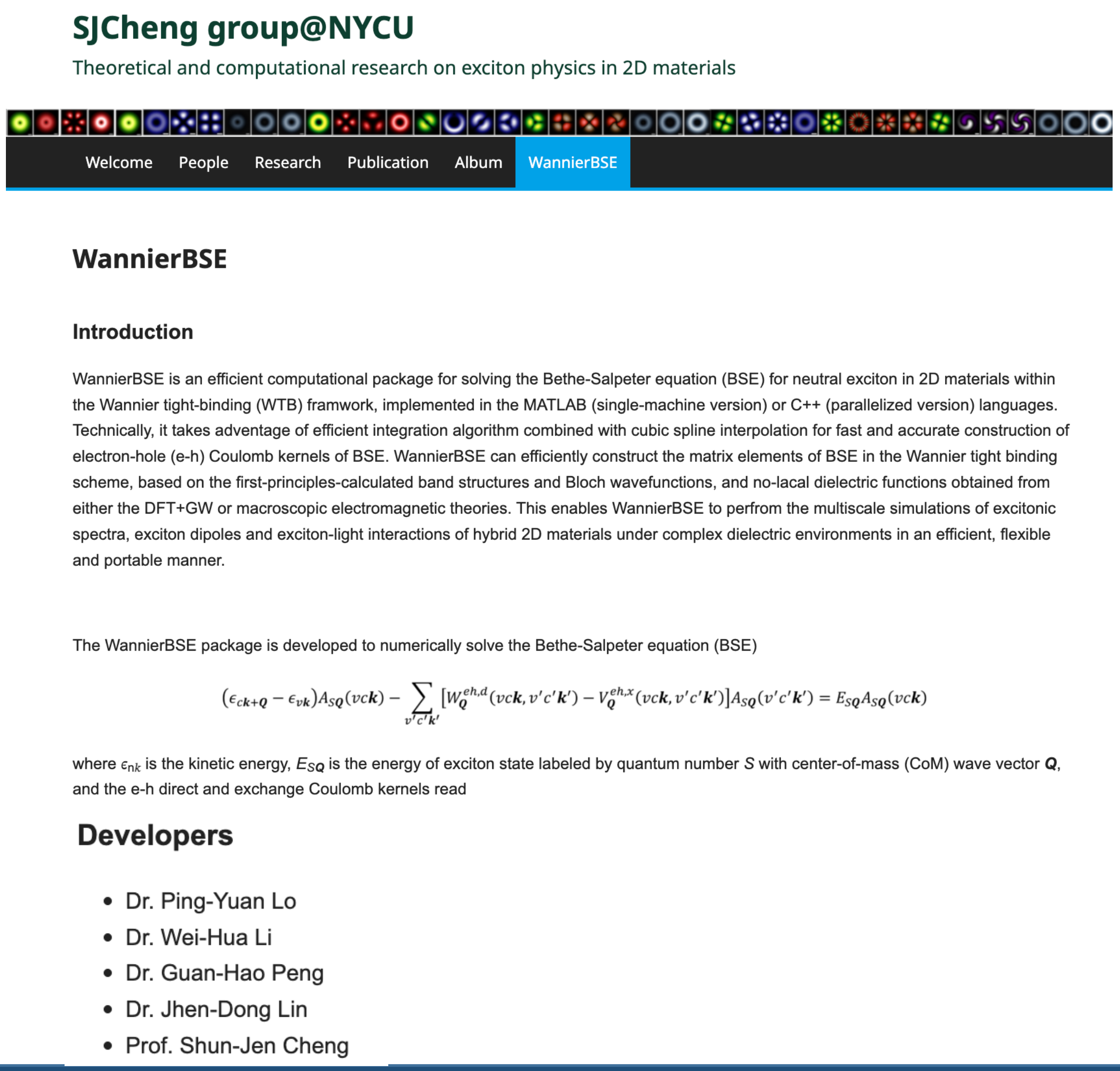
GHPeng, OSanchez@NYCU

- K Bryan, SJC, THL, YWL, ACS Nano 2021
- GH Peng, et al. JC, PRB 2022
- O Sanchez, et al. SJC, ACS Nano 2024
- YR Chen, SJC, THL, YWL, ACS Nano 2025

Summary

- We have developed a DFT-based BSE solver (**WannierBSE**) in the Wannier tight-binding (WTB) framework that serves as a multi-scaled computational platform for efficient simulation of exciton complexes in 2D materials under multi-layer dielectric environment.
- We simulate and analyze the landscape of the momentum-dependent valley polarization of TMD monolayer and interpret the observed high degree of polarization of intervalley exciton, in terms of the inherent crystal symmetry of TMD-ML.
- Using WannierBSE, we show that a pair of intervalley Q_{ex}/Q'_{ex} dark excitons of multi-layer TMDs are likely coupled to a high-lying bright single exciton ones via the exciton-exciton-annihilation (EEA), leading to the upconversion PLs (UPL).

https://quantum.web.nycu.edu.tw/wannierbse/


SJCheng group@NYCU

Theoretical and computational research on exciton physics in 2D materials

Welcome | People | Research | Publication | Album | WannierBSE

WannierBSE

Introduction

WannierBSE is an efficient computational package for solving the Bethe-Salpeter equation (BSE) for neutral exciton in 2D materials within the Wannier tight-binding (WTB) framwork, implemented in the MATLAB (single-machine version) or C++ (parallelized version) languages. Technically, it takes advantage of efficient integration algorithm combined with cubic spline interpolation for fast and accurate construction of electron-hole (e-h) Coulomb kernels of BSE. WannierBSE can efficiently construct the matrix elements of BSE in the Wannier tight binding scheme, based on the first-principles-calculated band structures and Bloch wavefunctions, and no-lacal dielectric functions obtained from either the DFT+GW or macroscopic electromagnetic theories. This enables WannierBSE to perfrom the multiscale simulations of excitonic spectra, exciton dipoles and exciton-light interactions of hybrid 2D materials under complex dielectric environments in an efficient, flexible and portable manner.

The WannierBSE package is developed to numerically solve the Bethe-Salpeter equation (BSE)

$$(\epsilon_{c\boldsymbol{k}+\boldsymbol{Q}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{Q}}(vc\boldsymbol{k}) - \sum_{v'c'\boldsymbol{k}'}[W_{\boldsymbol{Q}}^{eh,d}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') - V_{\boldsymbol{Q}}^{eh,x}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')]A_{S\boldsymbol{Q}}(v'c'\boldsymbol{k}') = E_{S\boldsymbol{Q}}A_{S\boldsymbol{Q}}(vc\boldsymbol{k})$$

where ϵ_{nk} is the kinetic energy, $E_{S\boldsymbol{Q}}$ is the energy of exciton state labeled by quantum number S with center-of-mass (CoM) wave vector $\boldsymbol{Q}$, and the e-h direct and exchange Coulomb kernels read

Developers

- Dr. Ping-Yuan Lo
- Dr. Wei-Hua Li
- Dr. Guan-Hao Peng
- Dr. Jhen-Dong Lin
- Prof. Shun-Jen Cheng

Acknowledgement

NSTC 國家科學及技術委員會 National Science and Technology Council

NARLabs 財團法人國家實驗研究院 NCHC 國家高速網路與計算中心 National Center for High-performance Computing

Exciton theory:

National Yang Ming Chiao Tung University ***(NYCU)***

- Guan-Hao Peng
- Oscar J G Sanchez
- Wei-Hua Li
- Ping-Yuan Lo




OAM light in 2D materials (exp)

National Taiwan Normal University ***(NTNU)***

- Prof. Yan-Wen Lan
- Prof. Ting-Hua Lu


Exciton in 2D materials (exp)

National Taiwan University (**NTU**)

- Dr. Shao-Yu Chen

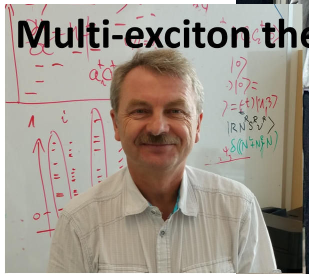
OAM in plasmonics (exp):

Leibniz Institute of Photonic Technology **(IPHT),** Germany

- Prof. Jer-Shing Huang

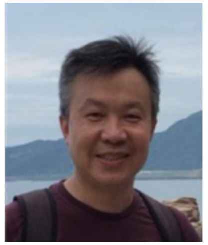
Dr. YH Chen
@Monash U

Multi-exciton theory



P.Hawrylak
@UOttawa






Pump-probe optics
Prof. C. W. Luo
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STM
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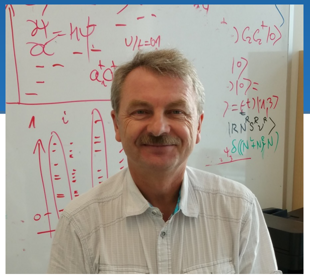
(DFT+GW):
H C. Hsueh
Tamkang Univ.

DFT:
C. K. Yang
Natl Chengchi U.

Thank you for your attention !!

WannierBSE

PYLo@NYCU

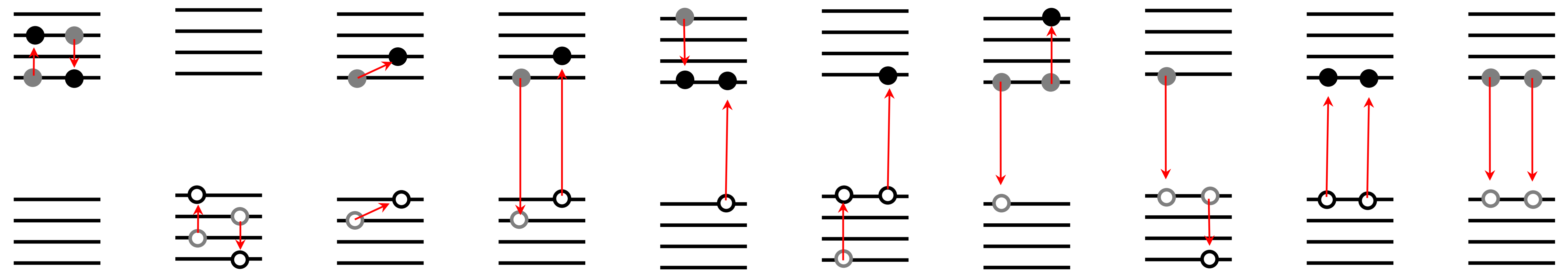


P Hawrylak@UOttawa

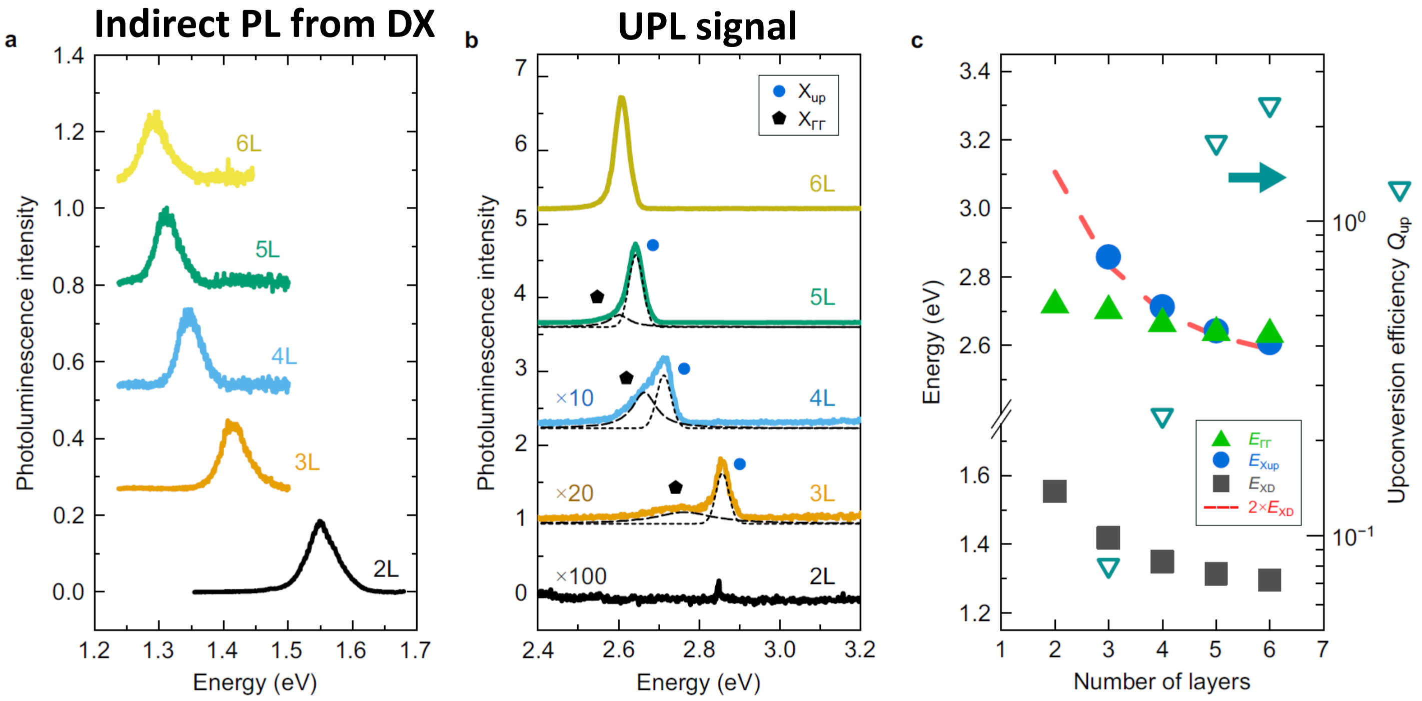
$$\hat{H}_C = \underbrace{\sum_j \epsilon_j \hat{c}_j^\dagger \hat{c}_j}_{(1)} - \underbrace{\sum_\alpha \epsilon_\alpha \hat{h}_\alpha^\dagger \hat{h}_\alpha}_{(2)} + \underbrace{\frac{1}{2}\sum_{j,k,k',j'} W^{ee}_{jkk'j'} \hat{c}_j^\dagger \hat{c}_k^\dagger \hat{c}_{k'} \hat{c}_{j'}}_{(3)} + \underbrace{\frac{1}{2}\sum_{\alpha,\beta,\beta',\alpha'} W^{hh}_{\alpha'\beta'\beta\alpha} \hat{h}_\alpha^\dagger \hat{h}_\beta^\dagger \hat{h}_{\beta'} \hat{h}_{\alpha'}}_{(4)} - \sum_{j,\alpha,\alpha',j'} \Big[\underbrace{W^{eh,d}_{j\alpha'\alpha j'}}_{(5)} - \underbrace{V^{eh,x}_{j\alpha' j'\alpha}}_{(6)}\Big] \hat{c}_j^\dagger \hat{h}_\alpha^\dagger \hat{h}_{\alpha'} \hat{c}_{j'}$$

$$\hat{H}_{NC} = -\underbrace{\sum_{j,k,k',\alpha} W^{eeh\leftarrow e}_{kj\alpha k'} \hat{h}_\alpha^\dagger \hat{c}_j^\dagger \hat{c}_k^\dagger \hat{c}_{k'}}_{(7)} - \underbrace{\sum_{j,\beta',\beta,\alpha} W^{ehh\leftarrow h}_{\beta' j\alpha\beta} \hat{c}_j^\dagger \hat{h}_\alpha^\dagger \hat{h}_\beta^\dagger \hat{h}_{\beta'}}_{(8)} - \underbrace{\sum_{j,\alpha',k',j'} W^{e\leftarrow eeh}_{j\beta' k' j'} \hat{c}_j^\dagger \hat{c}_{j'} \hat{c}_k \hat{h}_{\alpha'}}_{(9)} - \underbrace{\sum_{\alpha',\beta',\beta,j'} W^{h\leftarrow ehh}_{\beta'\alpha' j'\beta} \hat{h}_\beta^\dagger \hat{h}_{\beta'} \hat{h}_{\alpha'} \hat{c}_{j'}}_{(10)}$$
$$+ \underbrace{\frac{1}{2}\sum_{j,k,\beta,\alpha} V^{XX,+}_{jk\beta\alpha} \hat{c}_j^\dagger \hat{c}_k^\dagger \hat{h}_\beta^\dagger \hat{h}_\alpha^\dagger}_{(11)} + \underbrace{\frac{1}{2}\sum_{\alpha',\beta',k',j'} V^{XX,-}_{\alpha'\beta'k'j'} \hat{h}_{\alpha'} \hat{h}_{\beta'} \hat{c}_{k'} \hat{c}_{j'}}_{(12)}$$

(3) e-e (4) h-h (5) e-h dir. (6) e-h exch. (7) Auger eeh<-e (8) Auger ehh<-h (9) Auger e<-eeh (10) Auger h<-ehh (11) XX creation (12) XX annihilation



Layer dependence of UPL in few-layer TMDs

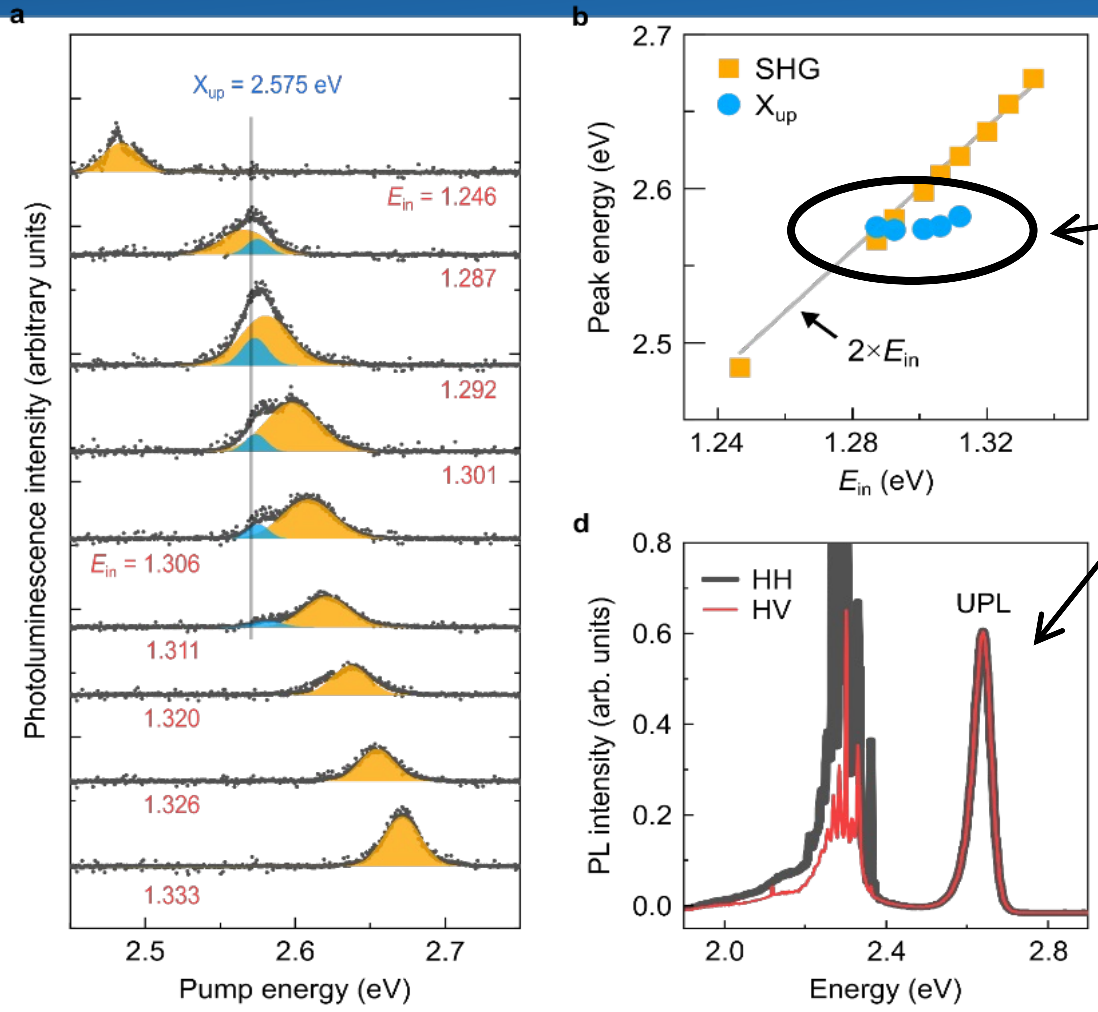


- The UPL energy is 2x of indirect PL from dark excitons
- The UPL emerges for 3L and thicker TMDs
- The upconversion efficiency increases as TMDs becomes thicker

Possibilities of UPL:

- Second harmonic generation (SHG)?
- Phonon-assisted UPL?
- Auger recombination?
- Exciton-exciton annihilation (EEA)?

UPL via second harmonic generation (SHG)?



Independent of pumping energy

Independent of the polarization of pumping light

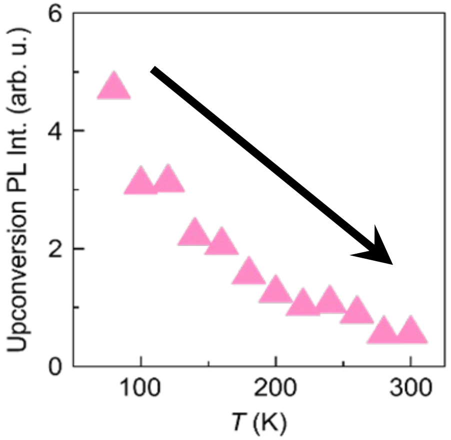
Second harmonic generation

Sample: WSe_2-5L

Is the UPL a phonon-assisted PL ?

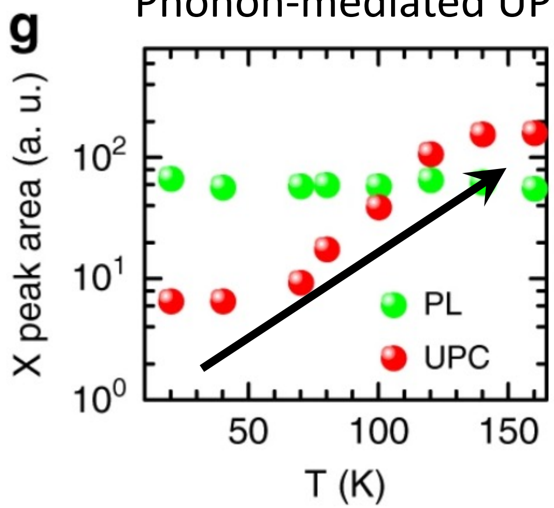
Temperature dependence of the UPL

This work



➡ ~~Phonon-assisted~~ UPL

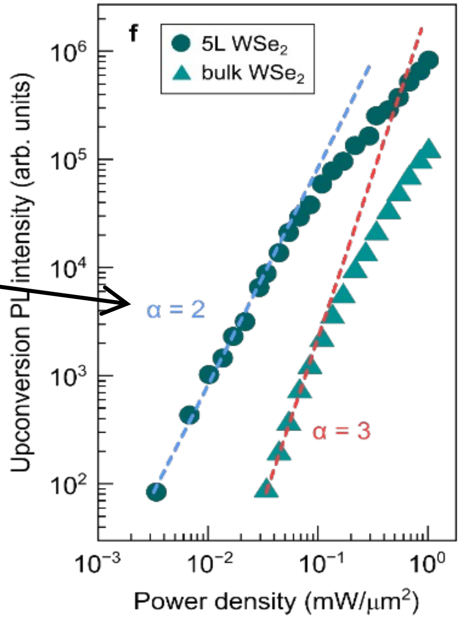
Phonon-mediated UPL

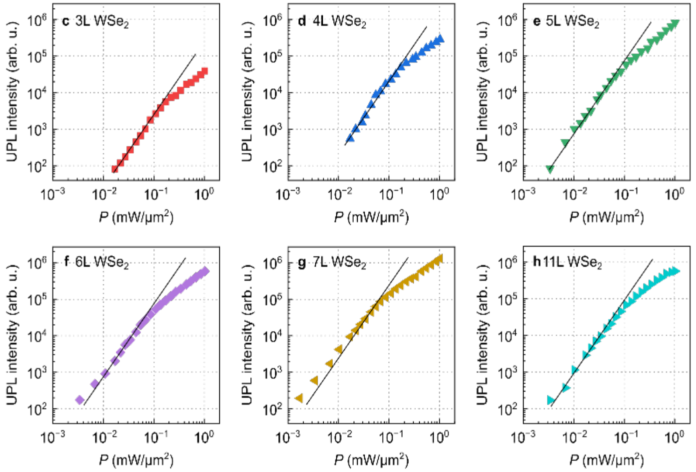


J. Jadczak *et al*. *Nat. Commun.* **10**, 107 (2019)

Power dependence of the UPL in few-layer TMDs

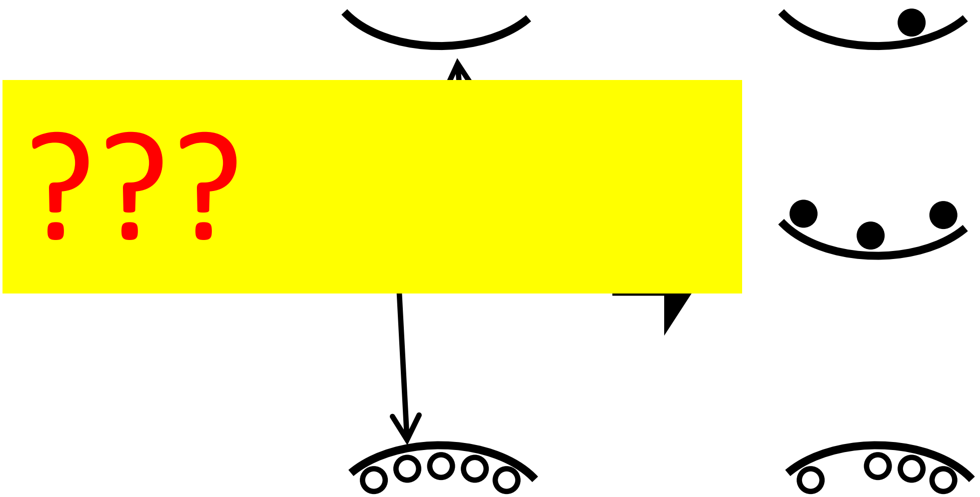
Exciton effects!!





($\alpha = 2$ for black lines)

Auger recombination

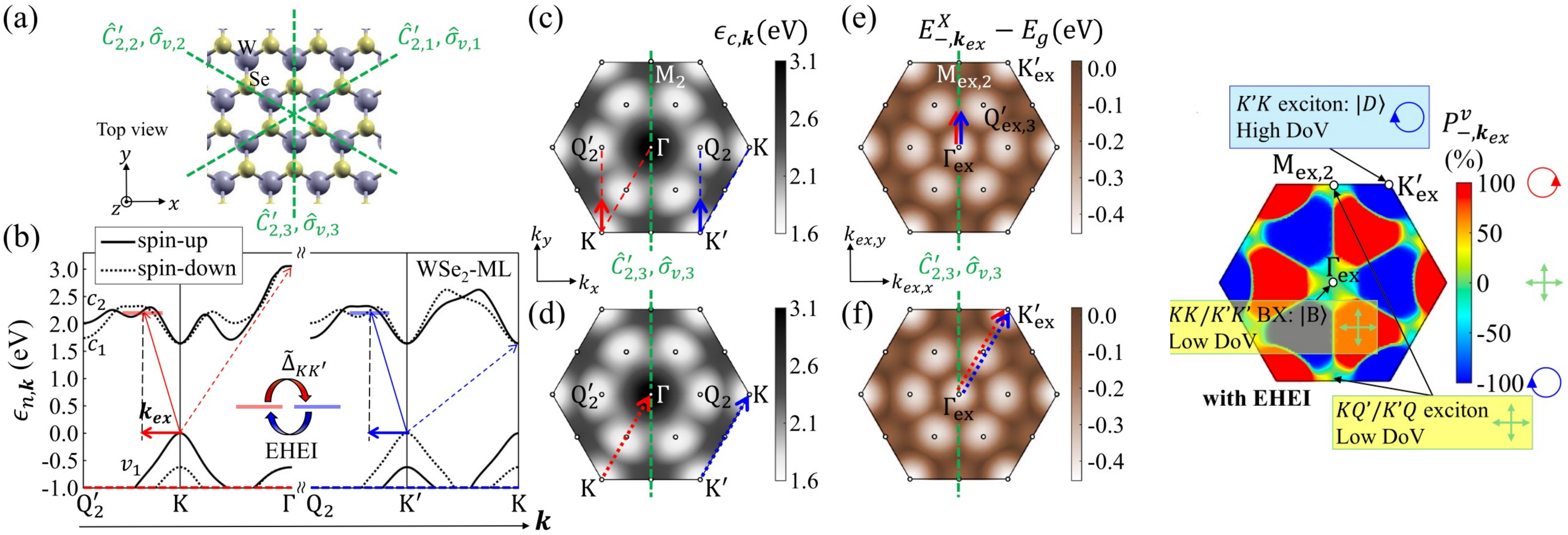


Assume $\bar{n}_e = \bar{n}_h = \bar{n}$, and are unbounded (no exciton effects)

Probability of e-h recombination $\propto \bar{n}^2$,

Probability of finding another electron to be scattered $\propto \bar{n}$

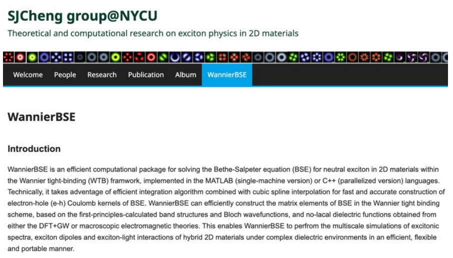
Power dependence of Auger recombination $\propto \bar{n}^3$



P.-Y. Lo, G.-H. Peng, W.-H. Li, Y. Yang, and S.-J. Cheng, Phys. Rev. Research **3**, 043198 (2021).

WannierBSE: a multi-scaled BSE-solver

GH Peng et al SJC, Nano Lett (2019)


SJCheng group@NYCU

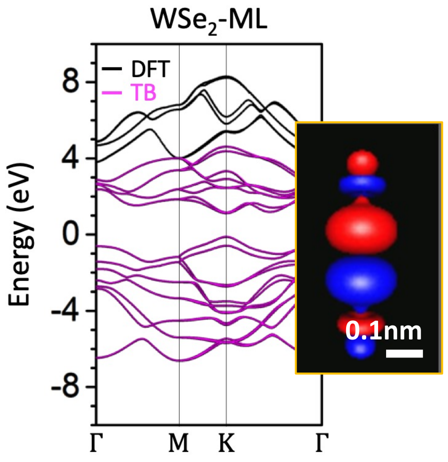
Theoretical and computational research on exciton physics in 2D materials

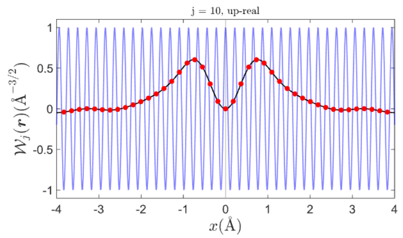
Welcome People Research Publication Album WannierBSE

WannierBSE

Introduction

WannierBSE is an efficient computational package for solving the Bethe-Salpeter equation (BSE) for neutral exciton in 2D materials within the Wannier tight-binding (WTB) framwork, implemented in the MATLAB (single-machine version) or C++ (parallelized version) languages. Technically, it takes adventage of efficient integration algorithm combined with cubic spline interpolation for fast and accurate construction of electron-hole (e-h) Coulomb kernels of BSE. WannierBSE can efficiently construct the matrix elements of BSE in the Wannier tight binding scheme, based on the first-principles-calculated band structures and Bloch wavefunctions, and no-lacal dielectric functions obtained from either the DFT+GW or macroscopic electromagnetic theories. This enables WannierBSE to perfrom the multiscale simulations of excitonic spectra, exciton dipoles and exciton-light interactions of hybrid 2D materials under complex dielectric environments in an efficient, flexible and portable manner.





DFT calculation (QE, VASP,…)

Wannierization (Wannier90)
1. Max. Localized Wannier func.
2. Wannier Tight-binding model

$$H^{KS}|\psi_{n,\boldsymbol{k}}\rangle = \epsilon_{n\boldsymbol{k}}|\psi_{n,\boldsymbol{k}}\rangle$$

$$|\mathcal{W}_{j\boldsymbol{R}}\rangle \equiv \frac{1}{\sqrt{N}}\sum_{\boldsymbol{k}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}} \sum_{n} U_{ni}^{(\boldsymbol{k})}|\psi_{n,\boldsymbol{k}}\rangle$$

$$H_{ij}^{TB}(\boldsymbol{k}) = \sum_{\boldsymbol{R}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}} t_{ij}(\boldsymbol{R})$$

$$t_{ij}(\boldsymbol{R}) = \langle\mathcal{W}_{j\boldsymbol{R}=\boldsymbol{0}}|H^{KS}|\mathcal{W}_{j\boldsymbol{R}}\rangle$$

Discretized with non-uniform $\boldsymbol{k}$-grids
$\boldsymbol{k} = \{\boldsymbol{k}_1, \boldsymbol{k}_2, \dots\}$

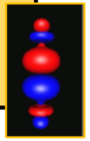
Efficient Adaptive integrations

$$\mathcal{W}_{j\boldsymbol{R}} \to \varrho_{j'j}^{(\boldsymbol{R})} \to \Xi_{j'j\ell'\ell}^{\boldsymbol{R},\boldsymbol{R}'}$$

$U_{\boldsymbol{k}_{ex}} = V_{\boldsymbol{k}_{ex}}^{d} - V_{\boldsymbol{k}_{ex}}^{x}$

Coulomb kernel

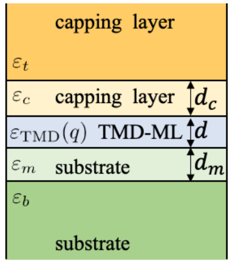
Exchange term
$C_j^{(n)}(\boldsymbol{k}), e^{i\boldsymbol{k}\cdot\boldsymbol{R}}, \Xi_{j'j\ell'\ell}^{\boldsymbol{R},\boldsymbol{R}'}$
$\downarrow$
$V_{\boldsymbol{k}_{ex}}^{x}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')$



Screened Direct term
$C_j^{(n)}(\boldsymbol{k}), e^{i\boldsymbol{k}\cdot\boldsymbol{\tau}_j}, \varepsilon(\boldsymbol{q})$
$\downarrow$
$V_{\boldsymbol{k}_{ex}}^{d}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')$

Multi-scaled electromagnetic theory

$\varepsilon(\boldsymbol{q})$

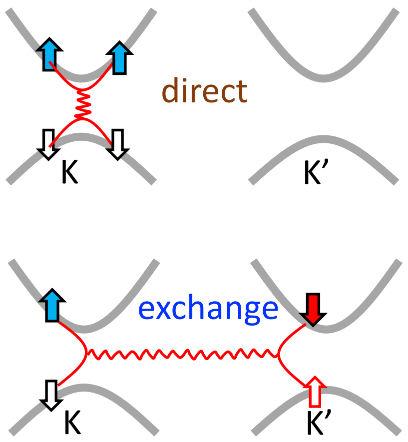


Bethe-Salpeter equation (BSE)

$$|S\boldsymbol{k}_{ex}\rangle = \Sigma_{\boldsymbol{k}} A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k})\hat{c}_{\boldsymbol{k}+\boldsymbol{k}_{ex}}^{\dagger}\hat{h}_{\boldsymbol{k}}^{\dagger}|0\rangle$$

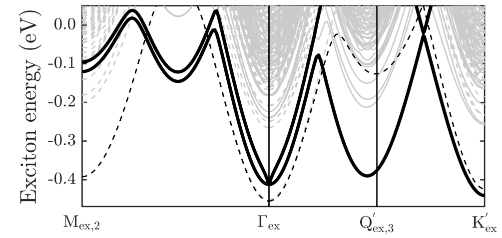
$$(\epsilon_{c\boldsymbol{k}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{k}_{ex}}(vc\boldsymbol{k}) - \frac{1}{\mathcal{A}}\sum_{v'c'\boldsymbol{k}'} U_{\boldsymbol{k}_{ex}}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')A_{S\boldsymbol{k}_{ex}}(v'c'\boldsymbol{k}') = E_{S\boldsymbol{k}_{ex}}^{X} A_{S\boldsymbol{k}_{ex}}(vc\boldsymbol{k}),$$

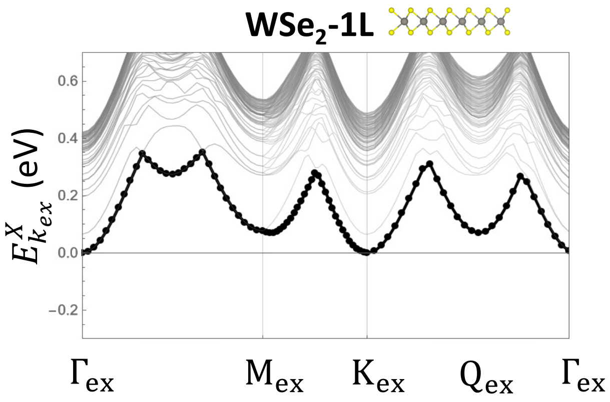
$\boxed{U_{\boldsymbol{k}_{ex}} = V_{\boldsymbol{k}_{ex}}^{d} - V_{\boldsymbol{k}_{ex}}^{x}}$

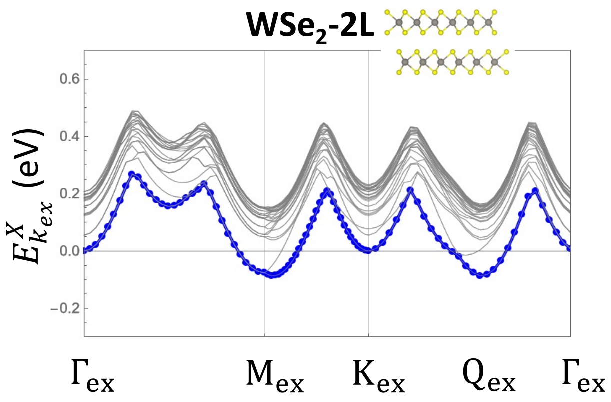


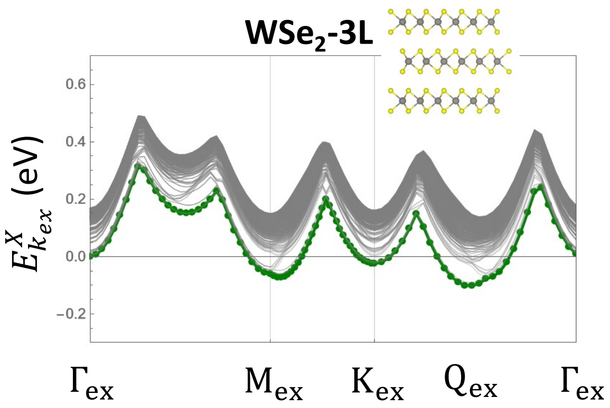
Diagonalization of BSE matrix

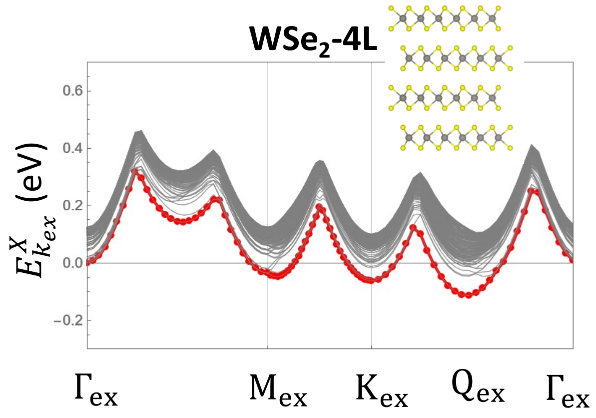
Output:
1. **Exciton bands: $E_{S,\boldsymbol{k}_{ex}}^{X}$**
2. **Exciton wave func. $A_{S,\boldsymbol{k}_{ex}}(vc\boldsymbol{k})$**
3. **Exciton dipoles**

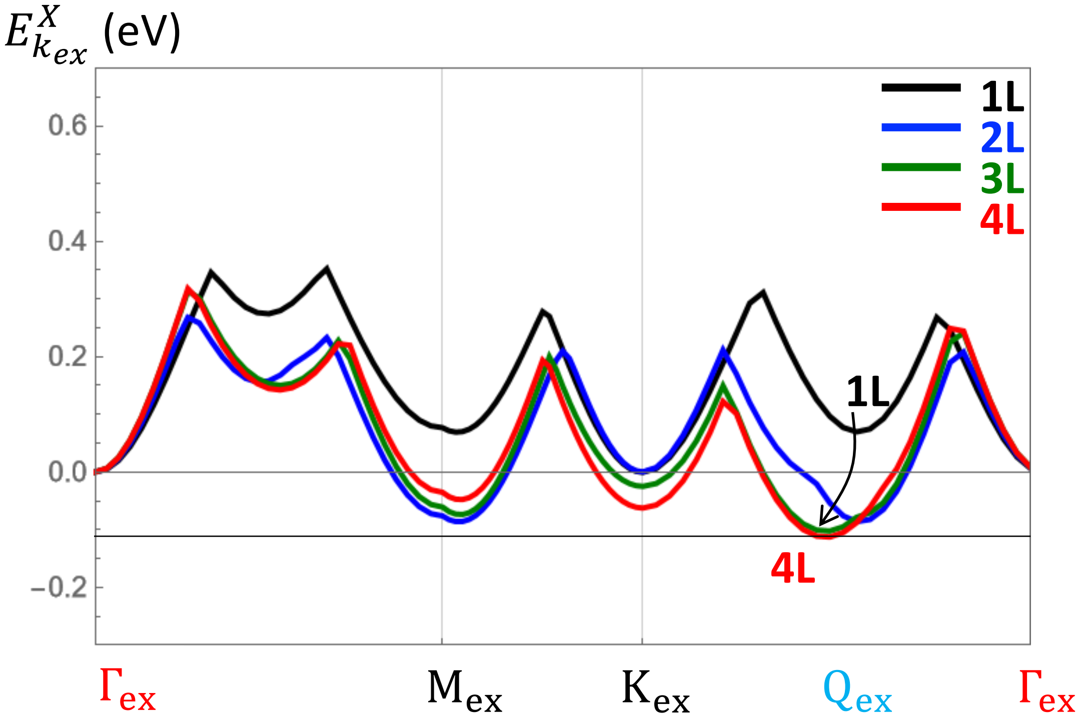


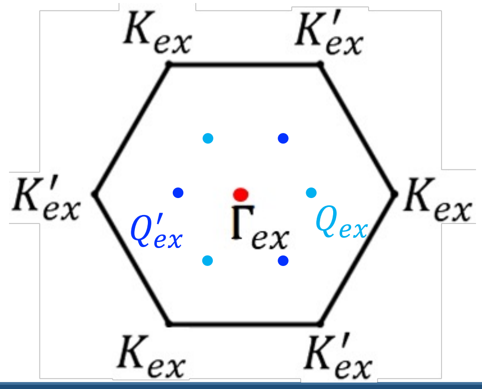

WSe$_2$-1L
$E^X_{k_{ex}}$ (eV)
0.6
0.4
0.2
0.0
−0.2
Γex
Mex
Kex
Qex
Γex


WSe$_2$-2L
$E^X_{k_{ex}}$ (eV)
0.6
0.4
0.2
0.0
−0.2
Γex
Mex
Kex
Qex
Γex


WSe$_2$-3L
$E^X_{k_{ex}}$ (eV)
0.6
0.4
0.2
0.0
−0.2
Γex
Mex
Kex
Qex
Γex

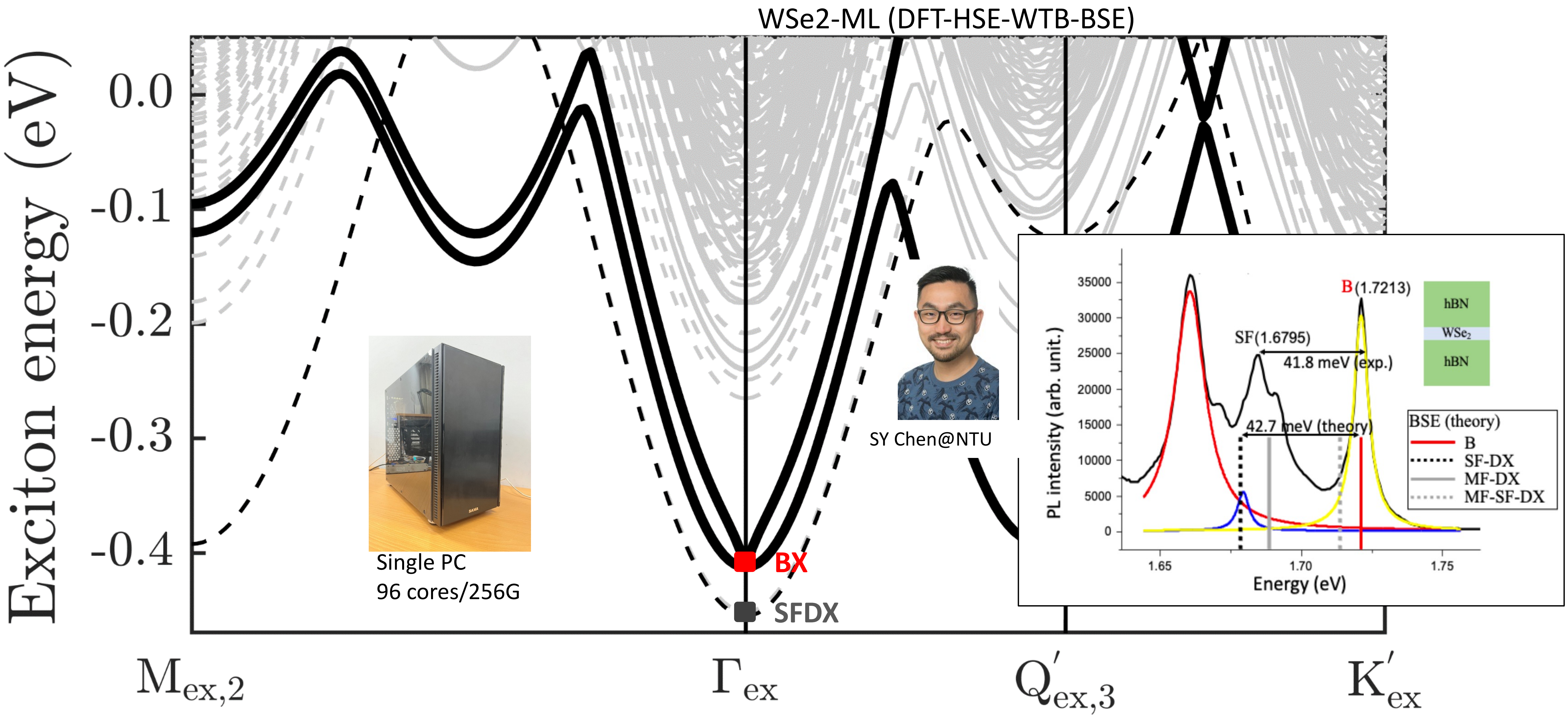

WSe$_2$-4L
$E^X_{k_{ex}}$ (eV)
0.6
0.4
0.2
0.0
−0.2
Γex
Mex
Kex
Qex
Γex


$E^X_{k_{ex}}$ (eV)
1L
2L
3L
4L
0.6
0.4
0.2
0.0
−0.2
1L
4L
Γex
Mex
Kex
Qex
Γex

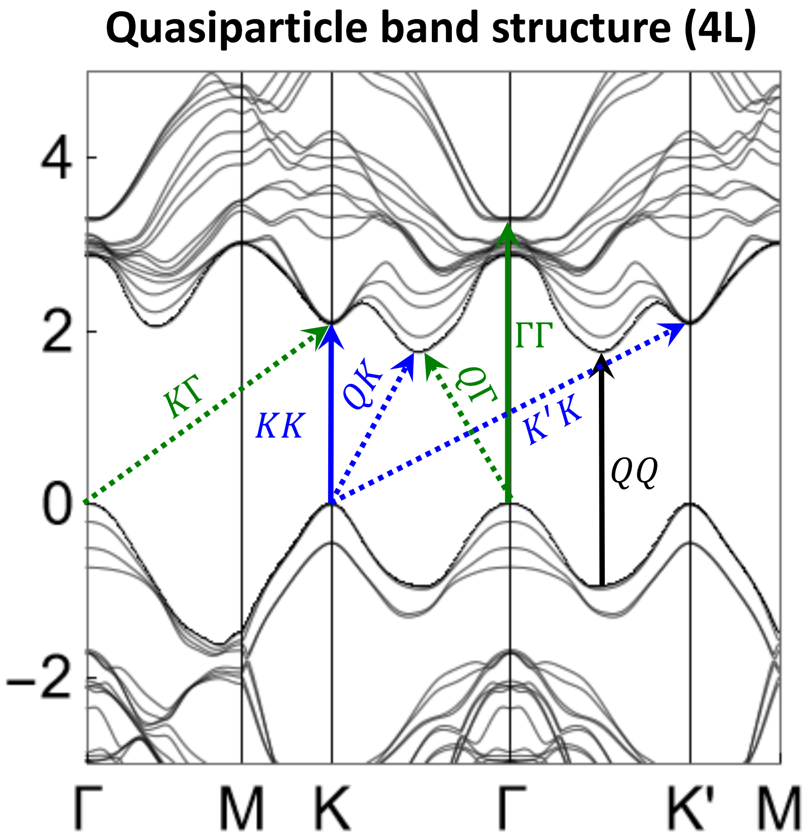

K_{ex}
K'_{ex}
K'_{ex}
K_{ex}
Q'_{ex}
Γ_{ex}
Q_{ex}
K_{ex}
K'_{ex}

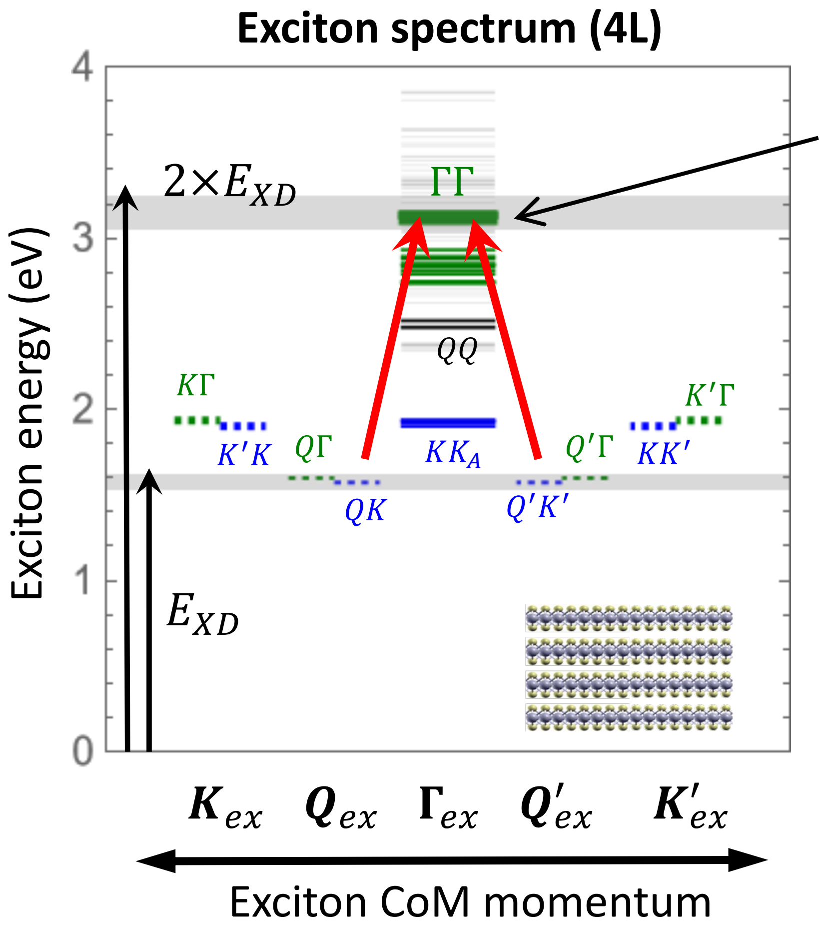
Exciton fine structure: BX and SFDX

GH Peng et al SJC, Nano Lett (2019)

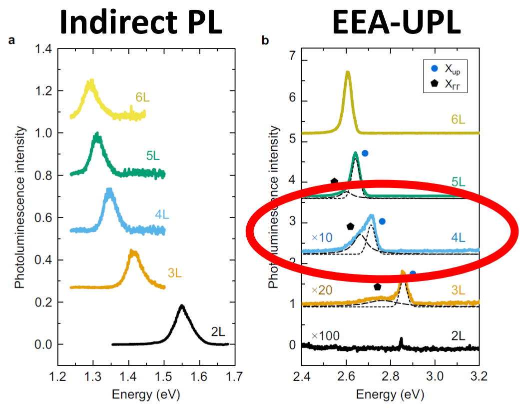


Viable pathways for resonant EEA in $\mathbf{WSe_2}$-4L

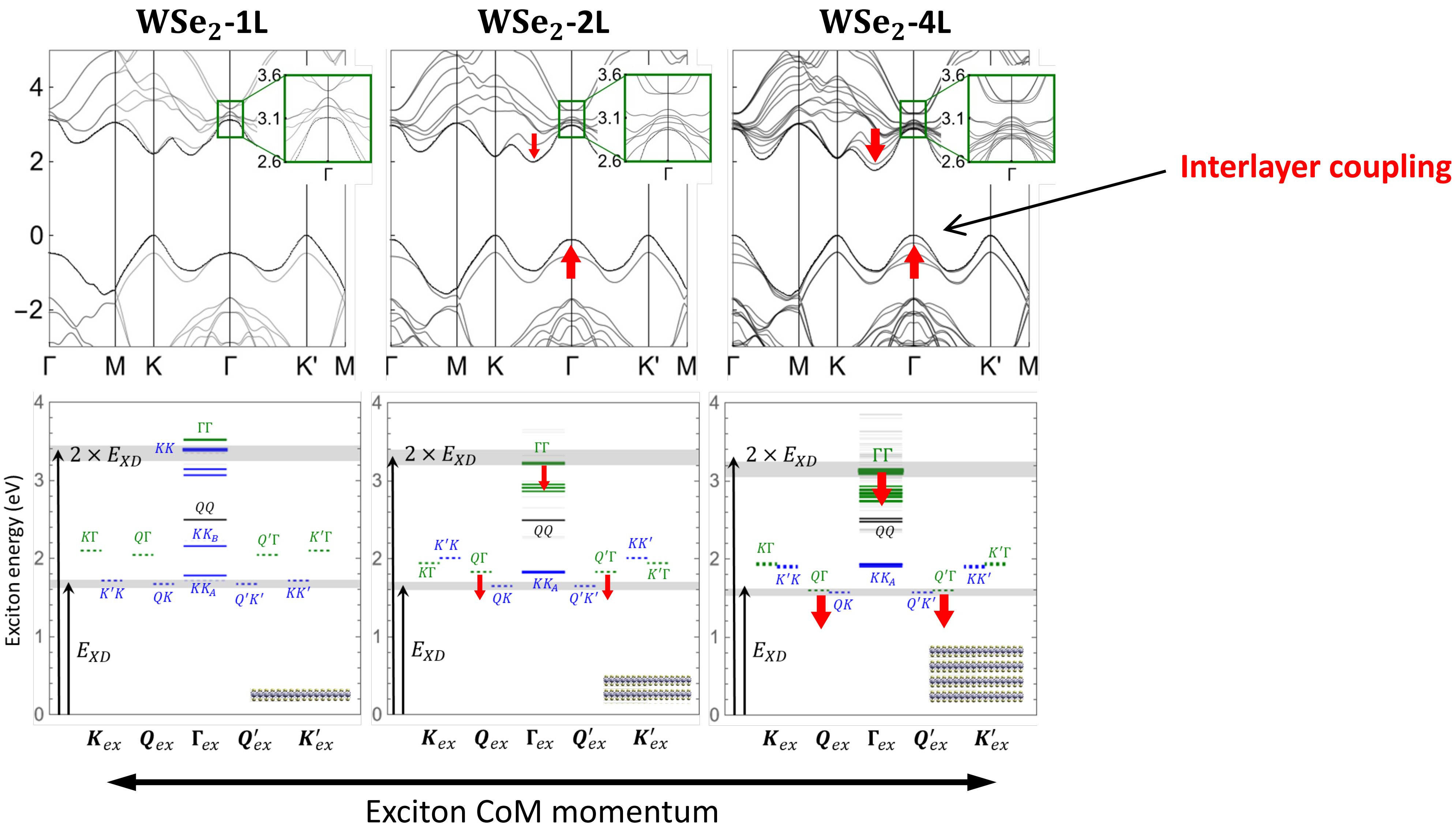




A cluster of ΓΓ exciton states in 4L WSe_2 fulfill the criteria of resonant EEA!!



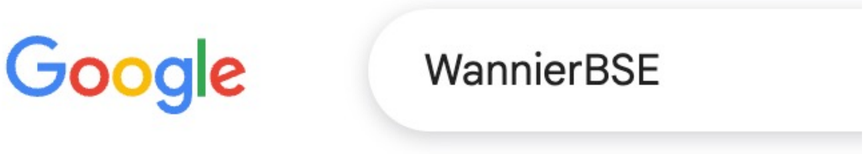
Evolution of quasiparticle band structures & exciton spectra



Multi-lyer more pronounced EEA

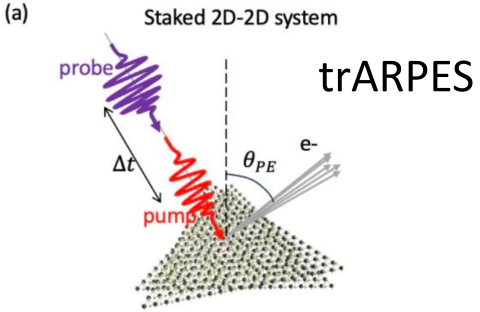
Summary

- We have developed a computational methodology for establishing and solving a DFT-based BSE in Wannier tight-binding (WTB) scheme (*WannierBSE*), which enable the quantitatively precise simulation of the energy bands, exciton dipoles, exciton wave functions of exciton complexes in 2D materials with drastically reduced numerical cost. The DFT-WTB-BSE solver serves as a multi-scaled computational platform for the computational and theoretical studies of exciton complexes in 2D materials, which can straightforwardly integrate the microscopic DFT and macroscopic electro-magnetic theory.
- Employing the *WannierBSE*, we have performed the theory-experiment-joint research on the intriguing physics of bright-, dark-, gray-, and finite-momentum excitons in variety. In this project, the code will be a useful tool for the theoretical simulation of the trARPES on the 2D systems.



***Subject* (研究主題):** 國家科學及技術委員會 NSTC National Science and Technology Council

Theory-experiment-joint studies of momentum- and time-resolved spectroscopy for exciton dynamics in hybrid-structured 2D materials (二維複合材料中激子動力學之時間與動量解析光譜研究)



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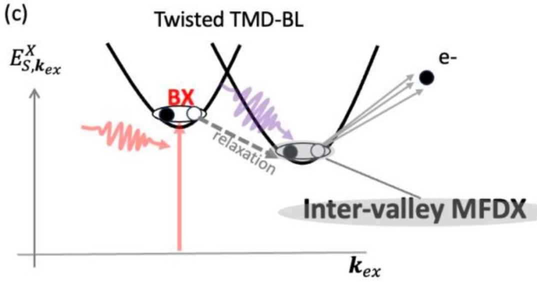

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WannierBSE

WannierBSE is an efficient computational package for solving the Bethe-Salpeter equation (BSE) for neutral exciton in 2D materials within the Wannier tight- ...

Developers

- Dr. Ping-Yuan Lo
- Dr. Wei-Hua Li
- Dr. Guan-Hao Peng
- Dr. Jhen-Dong Lin
- Prof. Shun-Jen Cheng

SJCheng group@NYCU

Theoretical and computational research on exciton physics in 2D materials

Welcome | People | Research | Publication | Album | WannierBSE

WannierBSE

Introduction

WannierBSE is an efficient computational package for solving the Bethe-Salpeter equation (BSE) for neutral exciton in 2D materials within the Wannier tight-binding (WTB) framwork, implemented in the MATLAB (single-machine version) or C++ (parallelized version) languages. Technically, it takes advantage of efficient integration algorithm combined with cubic spline interpolation for fast and accurate construction of electron-hole (e-h) Coulomb kernels of BSE. WannierBSE can efficiently construct the matrix elements of BSE in the Wannier tight binding scheme, based on the first-principles-calculated band structures and Bloch wavefunctions, and no-lacal dielectric functions obtained from either the DFT+GW or macroscopic electromagnetic theories. This enables WannierBSE to perfrom the multiscale simulations of excitonic spectra, exciton dipoles and exciton-light interactions of hybrid 2D materials under complex dielectric environments in an efficient, flexible and portable manner.

The WannierBSE package is developed to numerically solve the Bethe-Salpeter equation (BSE)

$$(\epsilon_{c\boldsymbol{k}+\boldsymbol{Q}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{Q}}(vc\boldsymbol{k}) - \sum_{v'c'\boldsymbol{k}'}\left[W_{\boldsymbol{Q}}^{eh,d}(vc\boldsymbol{k}, v'c'\boldsymbol{k}') - V_{\boldsymbol{Q}}^{eh,x}(vc\boldsymbol{k}, v'c'\boldsymbol{k}')\right]A_{S\boldsymbol{Q}}(v'c'\boldsymbol{k}') = E_{S\boldsymbol{Q}}A_{S\boldsymbol{Q}}(vc\boldsymbol{k})$$

where ϵ_{nk} is the kinetic energy, $E_{S\boldsymbol{Q}}$ is the energy of exciton state labeled by quantum number S with center-of-mass (CoM) wave vector $\boldsymbol{Q}$, and the e-h direct and exchange Coulomb kernels read

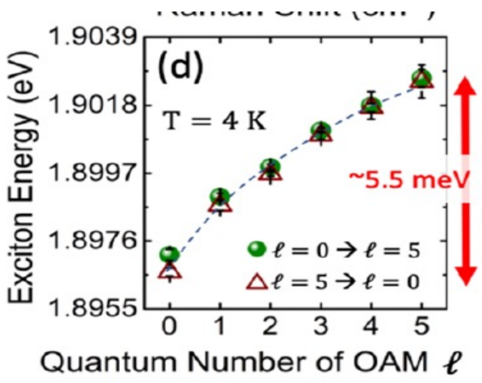
Summary

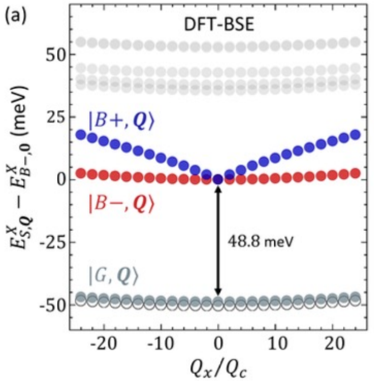
- We have presented a comprehensive theoretical study of the light-matter **interaction between OAM-TLs and exciton in TMD-MLs** by using the efficient in-house code to solve the **DFT-WTB-BSE.**

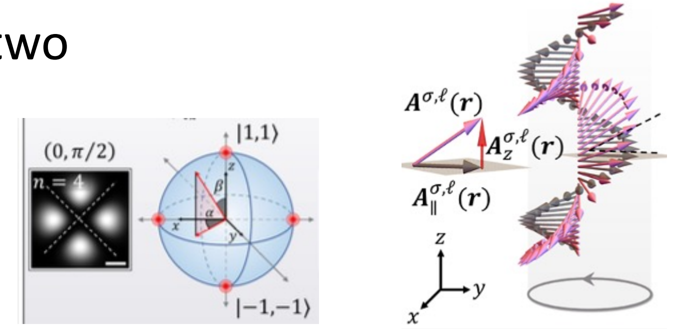
- We show that, in the electrical dipole approximation, a incident TL couples the **center-of mass motion** of bright exciton (BX) in a TMD-ML, and selectively photo-excites **finite-momentum excitons**, leading to the blue-shifted PLs with increasing the OAM of TL and formation of **localized exciton wave packet**.

- Due to the **photonic spin-orbital interaction**, a TL possesses **the longitudinal field**, which enhances the photo-excitation of spin-forbidden **gray exciton (GX).**

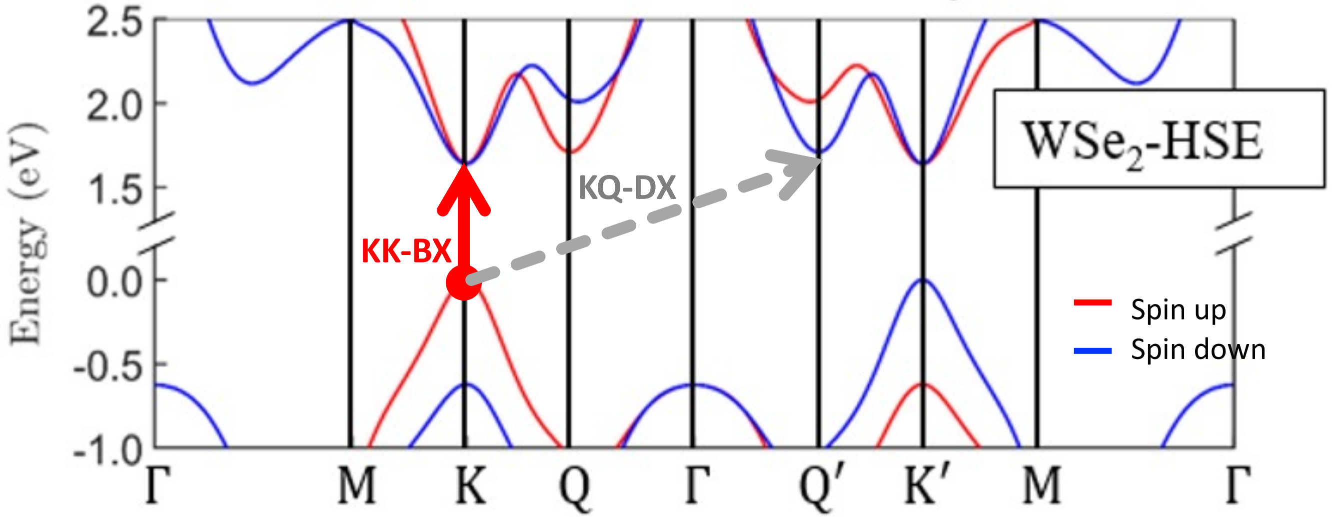
- Employing a **vector vortex beam (VVB)** formed by the superposition of two distinct TLs with opposite AM to excite a TMD-ML enable the **angular momentum imprint** of the **GX** states.

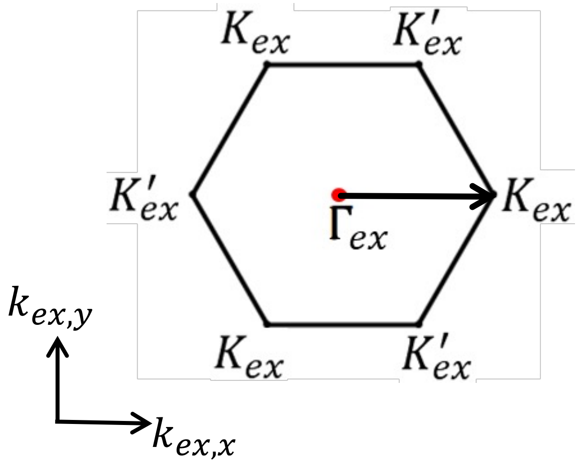


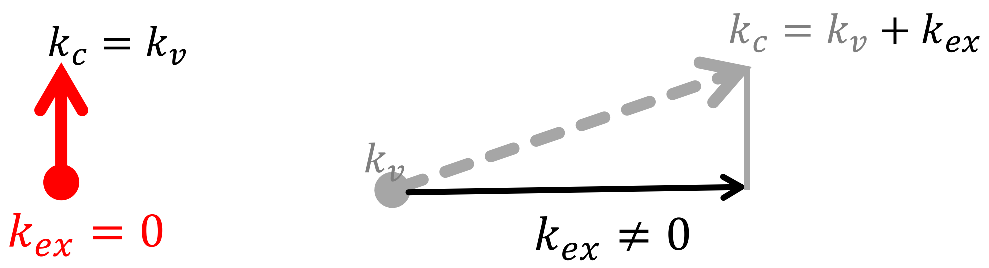


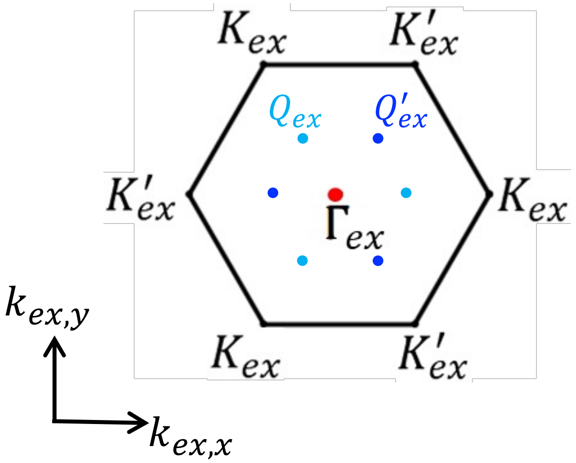


G. H. Peng, et al. SJC, Nano Lett. 19, 2299 (2019).

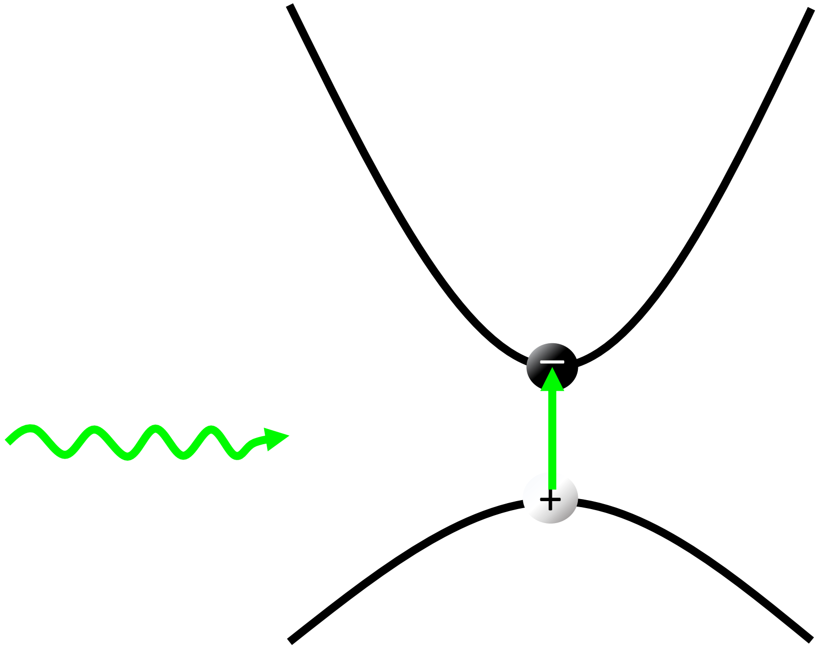




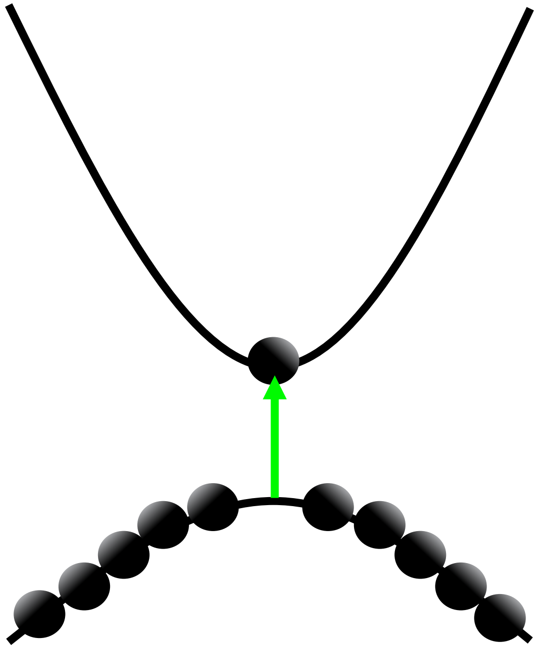




Exciton: a many-body problem



A naive picture: two-body problem



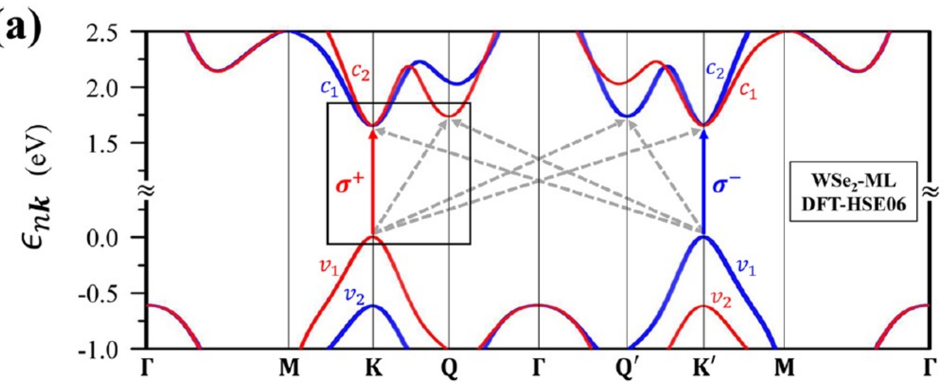
Reality: A many-body ($N_{tot} = (N_v - 1) + 1$) problem

Exciton state: $|S\boldsymbol{k}_{ex}\rangle = \Sigma_{\boldsymbol{k}} A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k})\hat{c}^{\dagger}_{k+k_{ex}}\hat{h}^{\dagger}_{k}|0\rangle$

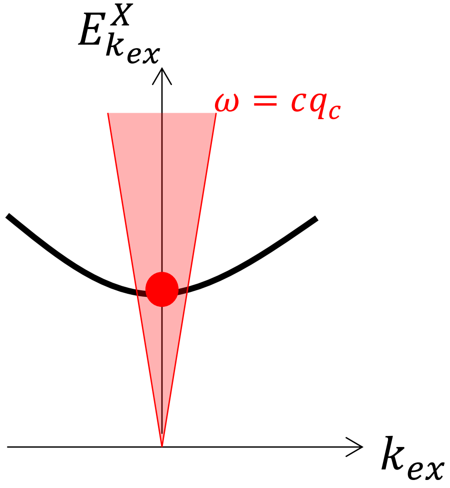
Bethe-Salpeter equation (BSE):

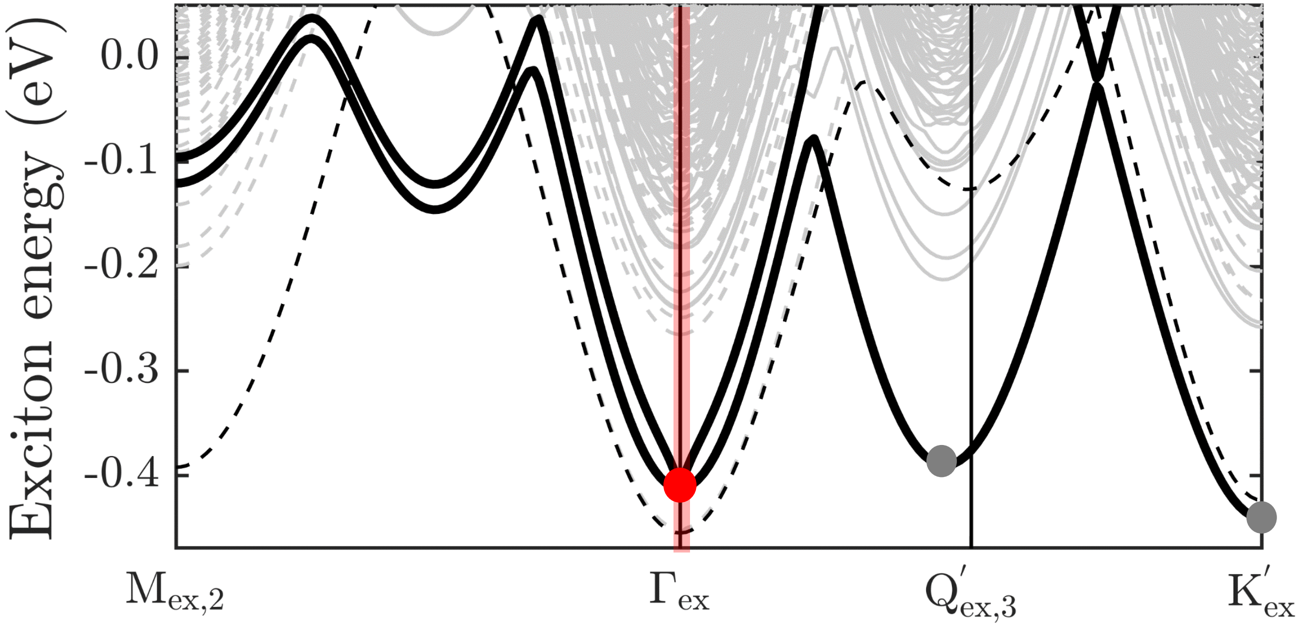
$$(\epsilon_{c\boldsymbol{k}} - \epsilon_{v\boldsymbol{k}})A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k}) - \frac{1}{\mathcal{A}}\sum_{\boldsymbol{k}'} U_{\boldsymbol{k}_{ex}}(\boldsymbol{k},\boldsymbol{k}')A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k}') = E^{X}_{S\boldsymbol{k}_{ex}}A_{S\boldsymbol{k}_{ex}}(\boldsymbol{k})$$

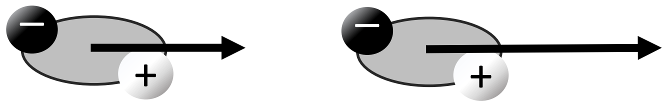
Exciton with finite momentum



G. H. Peng, **PYL**, et al. Nano Lett. **19**, 2299 (2019).

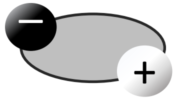






$k_{ex} \leq q_c \equiv \omega/c$

Intervalley dark excitons (**DXs**)



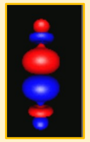
Bright exciton (BX)

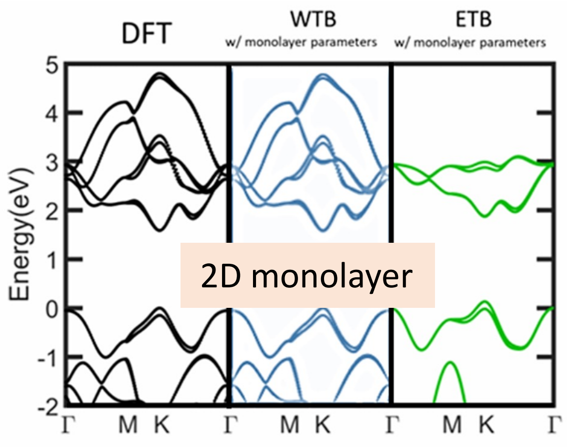
Dark exciton physics:

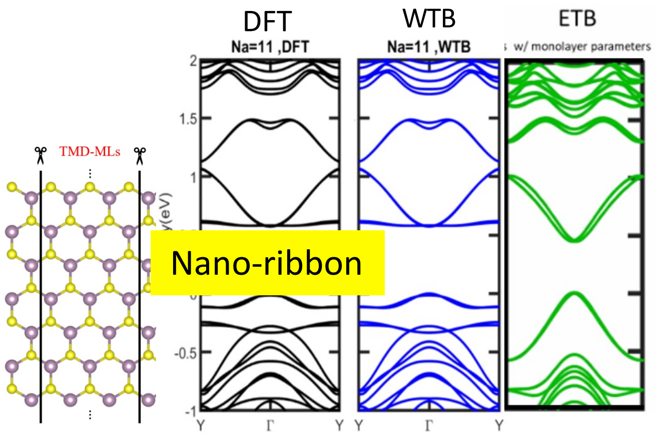
Theoretical challenges: --exciton band

-- generalized LMT beyond BX

Band theories: ETB vs. DFT vs. WTB

	ETB	WTB (WANNIER90)	DFT (Q. Espresso, …)
Band theories	Empirical tight-binding theory (**ETB**)	DFT-based Wannier tight binding (**WTB**) theory	Density-functional theory (**DFT**)
Applicable scales	Atomistic	Atomistic and microscopic	Atomistic and microscopic
Numerical cost	**Low cost**	**Low cost**	**High cost**
Advantages/ Disadvantages	• **Empirical** parameters, **not physics-based**. • **Non-transferrable** and **non-universal** parameters $\vert\phi_i\rangle$	• An ab-initio method • Deliver full microscopic **wave functions**	• An ab-initio method • Deliver full microscopic **wave functions** 





ETB:

$$H_{ij}^{TB}(\boldsymbol{k}) = \sum_{\boldsymbol{R}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}} t_{ij}(\boldsymbol{R})$$

WTB:

$$H_{i,j}^{\lambda}(\boldsymbol{k}) = \sum_{\boldsymbol{R}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}} H_{i,j}^{\lambda}(\boldsymbol{R}),$$