

# Numerical method for the calculation of excitonic effects in the optoelectronic responses of transition metal dichalcogenide monolayers

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# Outline

## ① Overview

- Monolayer transition metal dichalcogenide systems
- Exciton in 2D semiconductors

## ② Theoretical method

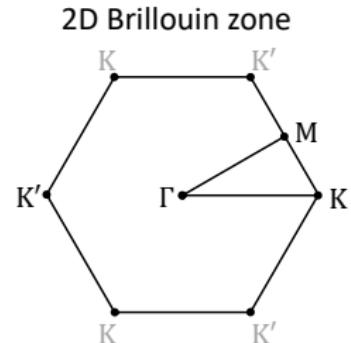
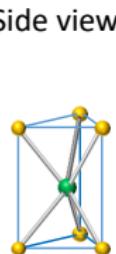
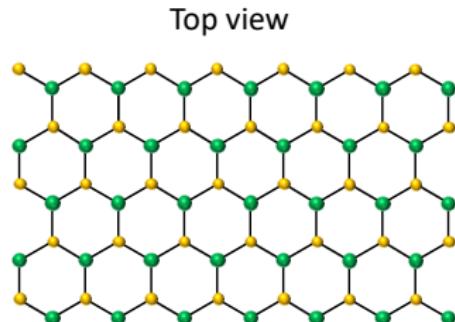
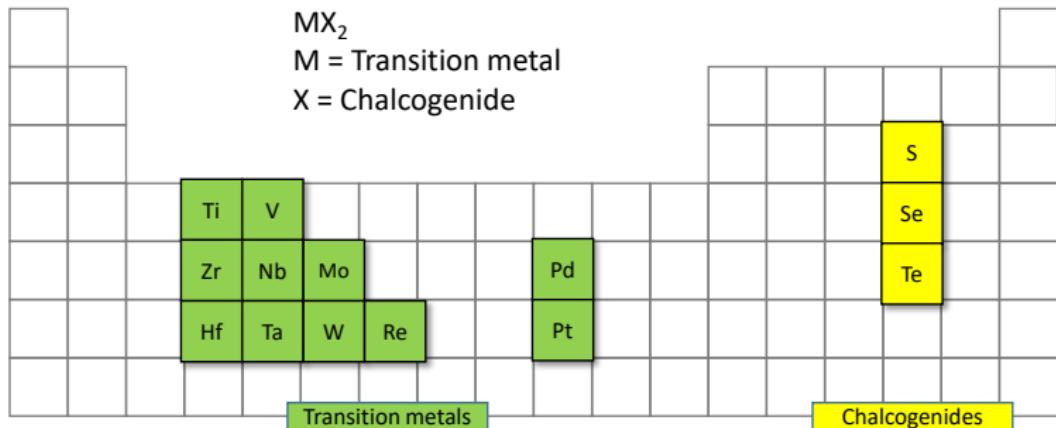
- Three-band tight-binding model
- Multi-band semiconductor Bloch equations

## ③ Numerical method

- Algorithm for time integrating the multi-band semiconductor Bloch equations in the entire 2D Brillouin zone
- Parallel computing on CPU and GPU using Fortran with OpenMP Device Offload

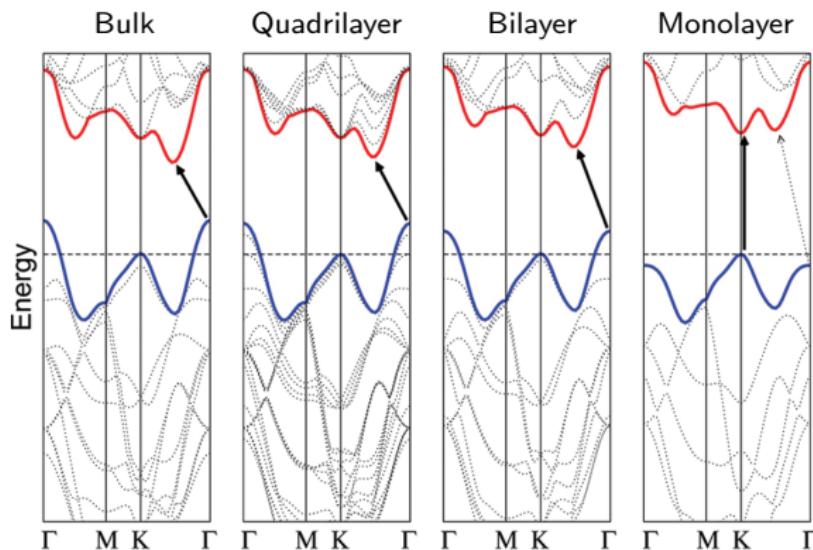
## ④ Model verification: Calculation of linear absorption spectrum and exciton binding energy

## Transition metal dichalcogenide monolayers



# Transition metal dichalcogenide monolayers

Transition metal dichalcogenide (TMD) monolayers are semiconductors with a direct band gap

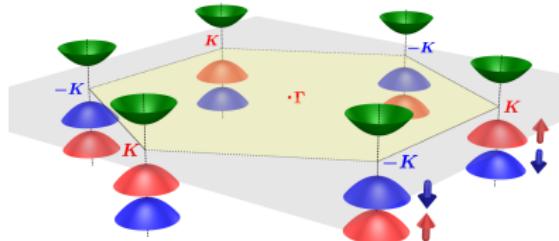


Ab initio band structures of bulk MoS<sub>2</sub> and ultrathin MoS<sub>2</sub> layers<sup>1</sup>

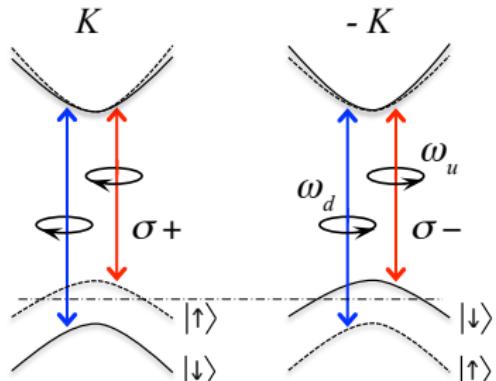
<sup>1</sup>Figure adapted from ref. A. Splendiani *et al.*, *Nano Lett.* **10**, 1271-1275 (2010)  
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# Transition metal dichalcogenide monolayers

TMD monolayers have a large spin splitting at band valleys



Schematic drawing of the band structure at the band edges



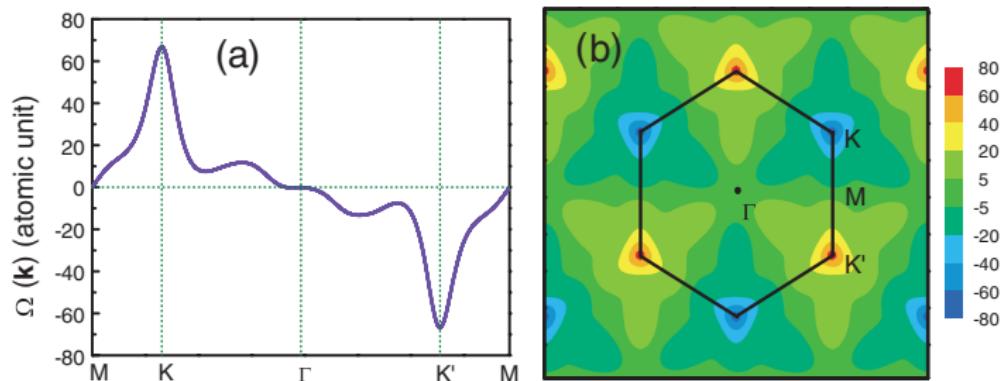
Valley and spin optical transition selection rules

Inversion asymmetry together with strong spin-orbit coupling (SOC) leads to a spin splitting of hundreds meV at the band valleys  
⇒ Coupled spin and valley physics<sup>2</sup>

<sup>2</sup>Figures adapted from ref. D. Xiao *et al.*, Phys. Rev. Lett. **108**, 196802 (2012)  
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# Transition metal dichalcogenide monolayers

TMD monolayers possess interesting Berry curvature effects<sup>3</sup>



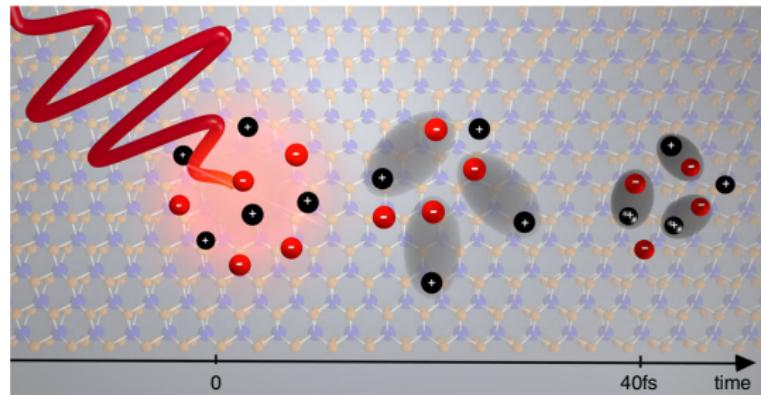
The Berry curvatures of monolayer MoS<sub>2</sub> along the high-symmetry lines (a) and in 2D  $k$ -space (b)<sup>4</sup>

<sup>3</sup>Berry curvature is a local manifestation of the geometric phase properties of the wave functions in the parameter space

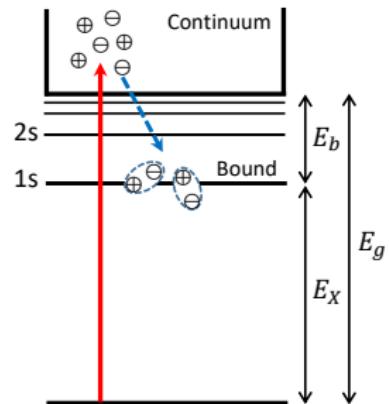
<sup>4</sup>Figure adapted from ref. W. Feng *et al.*, Phys. Rev. B **86**, 165108 (2012) (©2012 American Physical Society)

# Exciton in 2D semiconductors

Exciton is a pair of electron and hole bound together by Coulomb attraction

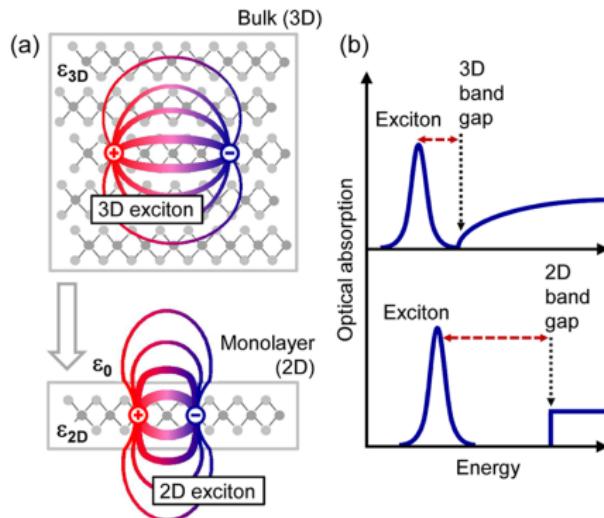


Exciton formation process after photo-injection of free electron-hole pairs<sup>5</sup>



<sup>5</sup>Figure adapted from ref. C. Trovatello *et al.*, Nat. Commun. **11**, 5277 (2020)  
(©2020 Springer Nature)

# Exciton in 2D semiconductors

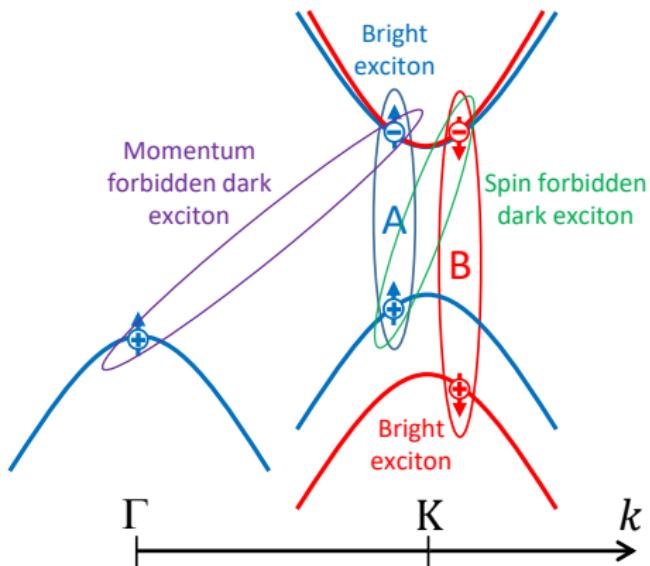


Strong quantum confinement and weak dielectric screening lead to an increase of exciton binding energy in semiconducting 2D materials, e.g.  $E_b = 0.32 \pm 0.04$  eV for monolayer  $\text{WS}_2$ <sup>6</sup>,  $E_b = 0.22 \pm 0.1$  eV for monolayer  $\text{MoS}_2$ <sup>7</sup>

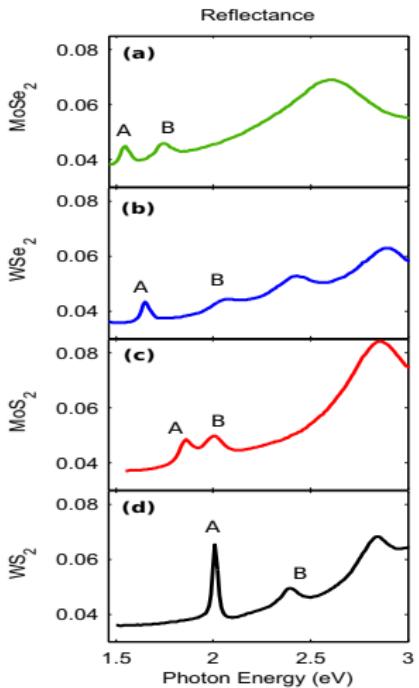
<sup>6</sup>Figure and data adapted from ref. A. Chernikov *et al.*, Phys. Rev. Lett. **113**, 076802 (2014) (©2014 American Physical Society)

<sup>7</sup>C. Zhang *et al.*, Nano Lett. **14**, 2443-2447 (2014)

# Exciton in 2D semiconductors



Different exciton types in TMD monolayers



Measured optical response of TMD monolayers<sup>8</sup>

<sup>8</sup>Figure adapted from ref. Y. Li *et al.*, Phys. Rev. B **90**, 205422 (2014) (©2014 American Physical Society)

# Three-band tight-binding model

Time-independent Schrödinger equation for an electron in the crystal:

$$\left[ -\frac{\hbar^2 \nabla^2}{2m} + U_0(\mathbf{r}) \right] |\psi_{\lambda, \mathbf{k}}(\mathbf{r})\rangle = \varepsilon_{\lambda, \mathbf{k}} |\psi_{\lambda, \mathbf{k}}(\mathbf{r})\rangle$$

Tight-binding (TB) wave function:

$$|\psi_{\lambda, \mathbf{k}}(\mathbf{r})\rangle = \sum_{\alpha} C_{\alpha}^{\lambda}(\mathbf{k}) \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} |\phi_{\alpha}(\mathbf{r} - \mathbf{R})\rangle$$

The basis consists of three  $d$ -orbitals of the  $M$  atom:

$$|d_{z^2} \uparrow\rangle, |d_{xy} \uparrow\rangle, |d_{x^2-y^2} \uparrow\rangle, |d_{z^2} \downarrow\rangle, |d_{xy} \downarrow\rangle, |d_{x^2-y^2} \downarrow\rangle$$

$C_{\alpha}^{\lambda}(\mathbf{k})$  is given by the equation:

$$\sum_{\beta} [H_{\alpha\beta}^{\text{TB}}(\mathbf{k}) - \varepsilon_{\lambda, \mathbf{k}} S_{\alpha\beta}(\mathbf{k})] C_{\beta}^{\lambda}(\mathbf{k}) = 0$$

# Three-band tight-binding model

Overlap matrix elements:

$$S_{\alpha\beta}(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \simeq \delta_{\alpha\beta}$$

TB Hamiltonian matrix elements:

$$H_{\alpha\beta}^{\text{TB}}(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \left[ -\frac{\hbar^2 \nabla^2}{2m} + U_0(\mathbf{r}) \right] | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle$$

Model including SOC with up to third-nearest-neighbor (TNN) hoppings<sup>9</sup>:

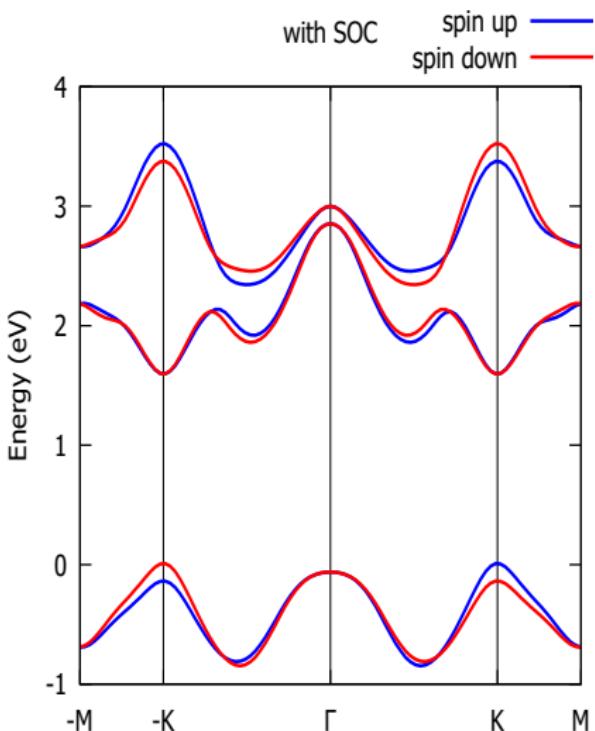
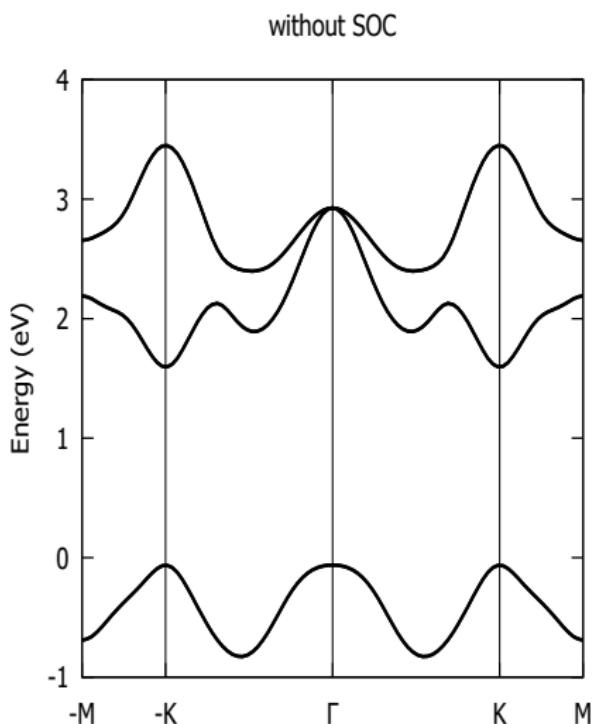
$$H^{\text{TB}}(\mathbf{k}) = \begin{bmatrix} V_0 & V_1 & V_2 & 0 & 0 & 0 \\ V_1^* & V_{11} & V_{12} + i\lambda & 0 & 0 & 0 \\ V_2^* & V_{12}^* - i\lambda & V_{22} & 0 & 0 & 0 \\ 0 & 0 & 0 & V_0 & V_1 & V_2 \\ 0 & 0 & 0 & V_1^* & V_{11} & V_{12} - i\lambda \\ 0 & 0 & 0 & V_2^* & V_{12}^* + i\lambda & V_{22} \end{bmatrix}$$

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<sup>9</sup>G. Liu, W. Shan, Y. Yao, W. Yao, and D. Xiao, Phys. Rev. B **88**, 085433 (2013)

# Three-band tight-binding model

## TB band structure of monolayer MoS<sub>2</sub>



# TB model in the presence of electric and magnetic fields

TB Hamiltonian matrix elements change to

$$H_{\alpha\beta}^{\text{TB}}(\mathbf{k}) = \sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \left[ \frac{(-i\hbar\boldsymbol{\nabla} + e\mathbf{A}(\mathbf{r}))^2}{2m} + U_0(\mathbf{r}) - e\Phi(\mathbf{r}) + \frac{e\hbar}{2m} \boldsymbol{\sigma} \cdot \mathbf{B} \right] | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle$$

It is possible to add a phase factor to the basis function

$$\begin{aligned} & \left[ \frac{(-i\hbar\boldsymbol{\nabla} + e\mathbf{A}(\mathbf{r}))^2}{2m} + U_0(\mathbf{r}) \right] e^{i\theta_{\mathbf{R}}} | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \\ &= e^{i\theta_{\mathbf{R}}} \left[ \frac{(-i\hbar\boldsymbol{\nabla} + e\mathbf{A}(\mathbf{r}) + \hbar\boldsymbol{\nabla}\theta_{\mathbf{R}})^2}{2m} + U_0(\mathbf{r}) \right] | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \end{aligned}$$

By choosing  $\theta_{\mathbf{R}} = -\frac{e}{\hbar} \int_{\mathbf{R}}^{\mathbf{r}} \mathbf{A}(\mathbf{r}') \cdot d\mathbf{r}'$  (Peierls substitution) one has

$$\begin{aligned} H_{\alpha\beta}^{\text{TB}}(\mathbf{k}) &= \sum_{\mathbf{R}} e^{\frac{ie}{\hbar} \int_0^{\mathbf{R}} \mathbf{A}(\mathbf{r}') \cdot d\mathbf{r}'} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \left[ -\frac{\hbar^2\boldsymbol{\nabla}^2}{2m} + U_0(\mathbf{r}) \right] | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \\ & - e \sum_{\mathbf{R}} e^{\frac{ie}{\hbar} \int_0^{\mathbf{R}} \mathbf{A}(\mathbf{r}') \cdot d\mathbf{r}'} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \Phi(\mathbf{r}) | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \\ & + \frac{e\hbar}{2m} \mathbf{B} \cdot \sum_{\mathbf{R}} e^{\frac{ie}{\hbar} \int_0^{\mathbf{R}} \mathbf{A}(\mathbf{r}') \cdot d\mathbf{r}'} e^{i\mathbf{k}\cdot\mathbf{R}} \langle \phi_{\alpha}(\mathbf{r}) | \boldsymbol{\sigma} | \phi_{\beta}(\mathbf{r} - \mathbf{R}) \rangle \end{aligned}$$

# Many-body Hamiltonian

Hamiltonian of the crystal in the presence of a light field:

$$H = H_0 + H_{e-L} + H_{e-e}$$

where

$$H_0 = \sum_i \left[ -\frac{\hbar^2 \nabla_i^2}{2m} + U_0(\mathbf{r}_i) \right], \quad H_{e-L} = \sum_i e \mathbf{E}(t) \cdot \mathbf{r}_i, \quad H_{e-e} = \sum_{i,j} \frac{e^2}{\epsilon |\mathbf{r}_i - \mathbf{r}_j|}$$

In second quantization:

$$H_0 = \sum_{\lambda, \mathbf{k}} \varepsilon_{\lambda, \mathbf{k}} a_{\lambda, \mathbf{k}}^\dagger a_{\lambda, \mathbf{k}}$$

$$H_{e-L} = e \mathbf{E}(t) \cdot \sum_{\lambda, \lambda', \mathbf{k}, \mathbf{k}'} \mathbf{r}_{\lambda \lambda'}(\mathbf{k}, \mathbf{k}') a_{\lambda, \mathbf{k}}^\dagger a_{\lambda', \mathbf{k}'}$$

$$H_{e-e} = \frac{1}{2} \sum_{\lambda_1, \lambda_2, \lambda_3, \lambda_4, \mathbf{k}, \mathbf{k}', \mathbf{q}} V_{\lambda_1 \lambda_2 \lambda_3 \lambda_4}(\mathbf{k}, \mathbf{k}', \mathbf{q}) a_{\lambda_1, \mathbf{k} + \mathbf{q}}^\dagger a_{\lambda_2, \mathbf{k}' - \mathbf{q}}^\dagger a_{\lambda_3, \mathbf{k}'} a_{\lambda_4, \mathbf{k}}$$

# Position and momentum matrix elements

Position matrix elements:

$$\mathbf{r}_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}') = \langle \psi_{\lambda, \mathbf{k}}(\mathbf{r}) | \mathbf{r} | \psi_{\lambda', \mathbf{k}'}(\mathbf{r}) \rangle = (\boldsymbol{\xi}_{\lambda\lambda'}(\mathbf{k}) + i\delta_{\lambda\lambda'} \boldsymbol{\nabla}_{\mathbf{k}}) \delta(\mathbf{k} - \mathbf{k}')$$

Transition dipole matrix elements:

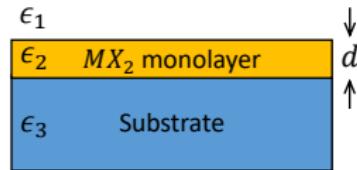
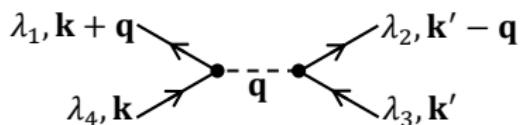
$$\boldsymbol{\xi}_{\lambda\lambda'}(\mathbf{k}) = -\frac{i\hbar}{m} \frac{\mathbf{p}_{\lambda\lambda'}(\mathbf{k})}{\varepsilon_{\lambda}(\mathbf{k}) - \varepsilon_{\lambda'}(\mathbf{k})}, \quad \lambda \neq \lambda'$$

Momentum matrix elements:

$$\begin{aligned} \mathbf{p}_{\lambda\lambda'}(\mathbf{k}) &= \langle \psi_{\lambda, \mathbf{k}}(\mathbf{r}) | -i\hbar \boldsymbol{\nabla} | \psi_{\lambda', \mathbf{k}}(\mathbf{r}) \rangle \\ &\simeq \frac{m}{\hbar} \sum_{\alpha, \beta} C_{\alpha}^{\lambda*}(\mathbf{k}) (\boldsymbol{\nabla}_{\mathbf{k}} H_{\alpha\beta}^{\text{TB}}(\mathbf{k})) C_{\beta}^{\lambda'}(\mathbf{k}) \end{aligned}$$

# Coulomb matrix elements

Coulomb matrix elements:



$$\begin{aligned} V_{\lambda_1 \lambda_2 \lambda_3 \lambda_4}(\mathbf{k}, \mathbf{k}', \mathbf{q}) &= \langle \psi_{\lambda_1, \mathbf{k}+\mathbf{q}}(\mathbf{r}) \psi_{\lambda_2, \mathbf{k}'-\mathbf{q}}(\mathbf{r}') | \frac{e^2}{\epsilon |\mathbf{r} - \mathbf{r}'|} | \psi_{\lambda_3, \mathbf{k}'}(\mathbf{r}') \psi_{\lambda_4, \mathbf{k}}(\mathbf{r}) \rangle \\ &\simeq \frac{2\pi e^2}{\epsilon L^2} \frac{1}{|\mathbf{q}|} \sum_{\alpha} C_{\alpha}^{\lambda_1*}(\mathbf{k} + \mathbf{q}) C_{\alpha}^{\lambda_4}(\mathbf{k}) \sum_{\beta} C_{\beta}^{\lambda_2*}(\mathbf{k}' - \mathbf{q}) C_{\beta}^{\lambda_3}(\mathbf{k}') \end{aligned}$$

The effective dielectric constant of  $\text{MX}_2$  monolayers can be described by<sup>10</sup>

$$\epsilon(\mathbf{q}) = \epsilon_2 \frac{1 - \frac{(1 - \epsilon_2/\epsilon_1)(1 - \epsilon_2/\epsilon_3)}{(1 + \epsilon_2/\epsilon_1)(1 + \epsilon_2/\epsilon_3)} e^{-2qd}}{\left[1 - \frac{1 - \epsilon_2/\epsilon_1}{1 + \epsilon_2/\epsilon_1} e^{-qd}\right] \left[1 - \frac{1 - \epsilon_2/\epsilon_3}{1 + \epsilon_2/\epsilon_3} e^{-qd}\right]}$$

<sup>10</sup>C. Zhang *et al.*, Phys. Rev. B **89**, 205436 (2014); L. V. Keldysh, Pis'ma Zh. Eksp. Teor. Fiz. 29, 716 (1979)

# Multi-band semiconductor Bloch equations

Reduced density matrix elements:

$$\rho_{\lambda\lambda'}(\mathbf{k}) = \langle a_{\lambda',\mathbf{k}}^\dagger a_{\lambda,\mathbf{k}} \rangle$$

Using the Heisenberg equation of motion and treating the many-body Coulomb interaction in the time-dependent Hartree-Fock approximation we obtain the multi-band semiconductor Bloch equations (SBE)

$$\begin{aligned} \frac{d}{dt} \rho_{\lambda\lambda'}(\mathbf{k}) &= -\frac{i}{\hbar} (\varepsilon_{\lambda,\mathbf{k}} - \varepsilon_{\lambda',\mathbf{k}}) \rho_{\lambda\lambda'}(\mathbf{k}) \\ &\quad - i \sum_{\mu} (\Omega_{\lambda\mu}(\mathbf{k}) \rho_{\mu\lambda'}(\mathbf{k}) - \rho_{\lambda\mu}(\mathbf{k}) \Omega_{\mu\lambda'}(\mathbf{k})) \\ &\quad + \frac{e}{\hbar} \mathbf{E}(t) \cdot \nabla_{\mathbf{k}} \rho_{\lambda\lambda'}(\mathbf{k}) + \left( \frac{d}{dt} \rho(\mathbf{k}) \bigg|_{\text{scat}} \right)_{\lambda\lambda'} \end{aligned}$$

Generalized Rabi frequency:

$$\Omega_{\lambda\lambda'}(\mathbf{k}) = \frac{1}{\hbar} [e \mathbf{E}(t) \cdot \boldsymbol{\xi}_{\lambda\lambda'}(\mathbf{k}) - \Sigma_{\lambda\lambda'}(\mathbf{k})]$$

# Multi-band semiconductor Bloch equations

Time-dependent Hartree-Fock self energy:

$$\begin{aligned}\Sigma_{\lambda\lambda'}(\mathbf{k}) &= \sum_{\mu,\nu,\mathbf{k}'} V_{\mu\lambda\nu\lambda'}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) \rho_{\nu\mu}(\mathbf{k}') \\ &= \sum_{\substack{\mu=\nu, \mathbf{k}' \\ \mu \in \text{VB}}} V_{\mu\lambda\mu\lambda'}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) + \sum_{\substack{\mu=\nu, \mathbf{k}' \\ \mu \in \text{VB}}} V_{\mu\lambda\mu\lambda'}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) (1 - \rho_{\mu\mu}(\mathbf{k}')) \\ &\quad + \sum_{\substack{\mu=\nu, \mathbf{k}' \\ \nu \in \text{CB}}} V_{\nu\lambda\nu\lambda'}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) \rho_{\nu\nu}(\mathbf{k}') + \sum_{\mu \neq \nu, \mathbf{k}'} V_{\mu\lambda\nu\lambda'}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) \rho_{\nu\mu}(\mathbf{k}')\end{aligned}$$

Electric field of light:

$$\mathbf{E}(t) = \hat{\mathbf{e}} E_{\text{env}}(t) e^{i\omega_L t} + c.c$$

Initial condition at  $t = -\infty$ :

$$\rho_{\lambda\lambda'}(\mathbf{k}) = \begin{cases} 1 & \lambda = \lambda' \in \text{VB} \\ 0 & \text{otherwise} \end{cases}$$

# Optoelectronic responses

From the solutions of SBE we can compute optoelectronic responses

Carrier density:

$$N_e(t) = \frac{1}{L^2} \sum_{\lambda \in \text{CB}, \mathbf{k}} \rho_{\lambda\lambda}(\mathbf{k}), \quad N_h(t) = \frac{1}{L^2} \sum_{\lambda \in \text{VB}, \mathbf{k}} (1 - \rho_{\lambda\lambda}(\mathbf{k}))$$

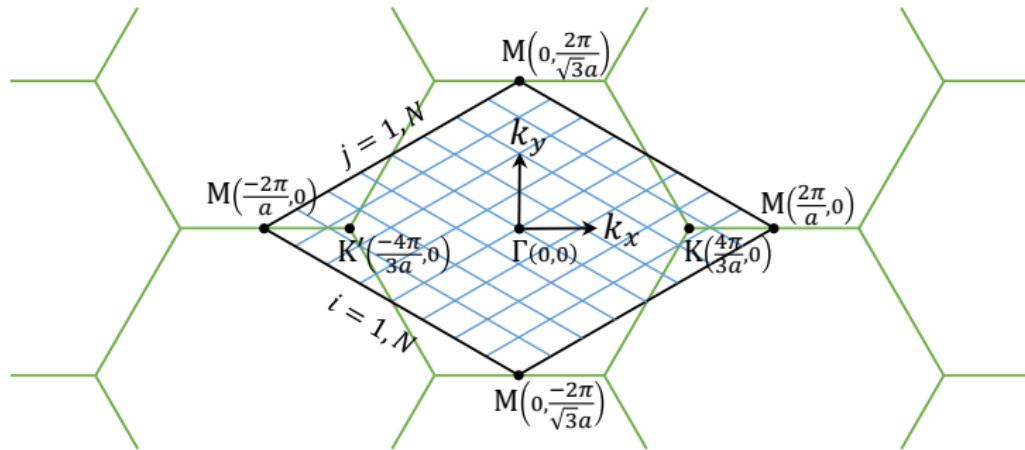
Photocurrent response:

$$\mathbf{J}(t) = \frac{e}{L^2 m} \sum_{\lambda, \mathbf{k}} \mathbf{p}_{\lambda\lambda}(\mathbf{k}) \rho_{\lambda\lambda}(\mathbf{k})$$

Interband polarization response:

$$\mathbf{P}(t) = \frac{e}{L^2} \sum_{\lambda, \lambda' \neq \lambda, \mathbf{k}} \boldsymbol{\xi}_{\lambda\lambda'}(\mathbf{k}) \rho_{\lambda'\lambda}(\mathbf{k})$$

# Discretization of the k-space using a non-Cartesian grid



Sum over  $k$ -states:

$$\sum_{\mathbf{k}} f(\mathbf{k}) \rightarrow \frac{L^2}{4\pi^2} \iint_{\text{BZ}} f(k_x, k_y) dk_x dk_y \rightarrow \frac{L^2}{4\pi^2} \frac{\sqrt{3}}{2} \Delta k^2 \sum_{i,j=1}^N f(k_x(i, j), k_y(i, j))$$

# Discretization of the k-space using a non-Cartesian grid

Let  $N$  be the number of  $k$ -points in one dimension

The number of  $k$ -points in two dimensions is  $N^2$

Coulomb matrix couples all two-dimensional wave vectors  $\mathbf{k}$  and  $\mathbf{k}'$

$$V_{\lambda_1 \lambda_2 \lambda_3 \lambda_4}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) = \frac{2\pi e^2}{\epsilon L^2} \frac{1}{|\mathbf{k}' - \mathbf{k}|} \sum_{\alpha} C_{\alpha}^{\lambda_1*}(\mathbf{k}') C_{\alpha}^{\lambda_4}(\mathbf{k}) \sum_{\beta} C_{\beta}^{\lambda_2*}(\mathbf{k}) C_{\beta}^{\lambda_3}(\mathbf{k}')$$

The computational amount of the Coulomb matrix scales as  $N^4$

Parallel computing can be used to speed up calculations

## Temporal discretization for the numerical solutions of SBE

The semiconductor Bloch equations (SBE) take the form of an initial value problem as follows

$$\frac{dy}{dt} = f(t, y), \quad y(t_0) = y_0$$

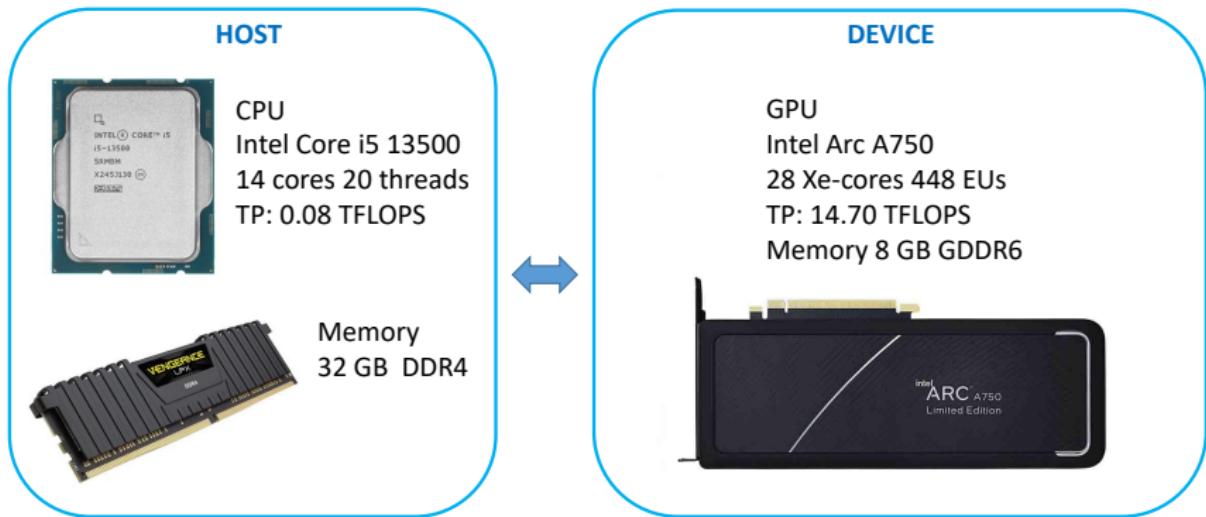
Differential equations of this type can be solved using the Runge-Kutta method  $\text{RK}_4$

Let  $\Delta t$  be the time step for numerical integration

$$\Delta t \ll T_L$$

$T_L$  is the period of the incident light wave

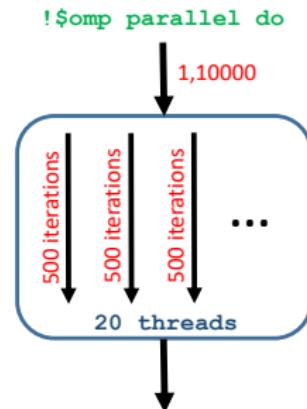
# Hardware for parallel computing



# Parallelism in Intel OpenMP Fortran

Creating a parallel code on the host

```
real :: a(100),b(100),c(100,100)
do i=1,100
  a(i)=1.
  b(i)=1.
end do
 !$omp parallel do
 do i=1,100
   do j=1,100
     c(i,j) = a(i)*b(j)
   end do
 end do
 !$omp end parallel do
 write(*,*)c
```



All CPU threads execute calculations in parallel

# Parallelism in Intel OpenMP Offload Fortran

## Offloading the parallel code to a target device

```
real :: a(100),b(100),c(100,100)
do i=1,100
  a(i)=1.
  b(i)=1.
end do

!$omp target map(to:a,b) map(tofrom:c)
 !$omp parallel do
  do i=1,100
    do j=1,100
      c(i,j) = a(i)*b(j)
    end do
  end do
 !$omp end parallel do
 !$omp end target

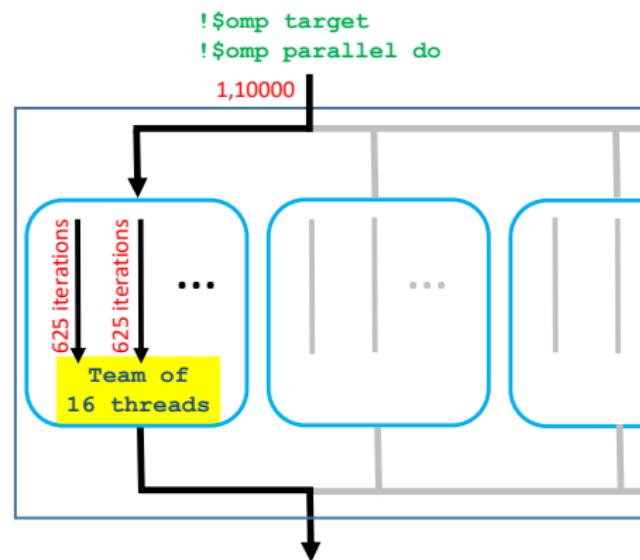
write(*,*)

```

Host code

Device code

Host code



Only one streaming multiprocessor (Xe core) is used for parallel computing

# Parallelism in Intel OpenMP Offload Fortran

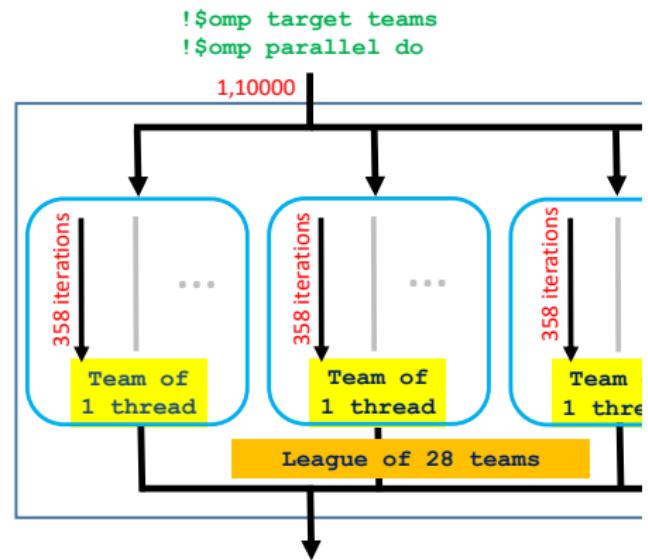
Creating a league of teams where the primary thread of each team executes calculations

```
real :: a(100),b(100),c(100,100)
do i=1,100
  a(i)=1.
  b(i)=1.
end do
!$omp target teams map(to:a,b)
map(tofrom:c)
 !$omp parallel do
  do i=1,100
    do j=1,100
      c(i,j) = a(i)*b(j)
    end do
  end do
 !$omp end parallel do
 !$omp end target teams
 write(*,*),c
```

Host code

Device code

Host code



# Parallelism in Intel OpenMP Offload Fortran

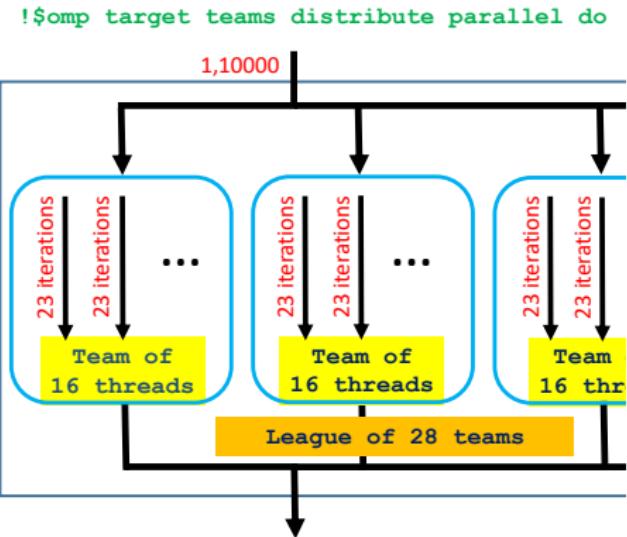
## Distributing calculations to all GPU Execution Units

```

real :: a(100),b(100),c(100,100)
do i=1,100
  a(i)=1.
  b(i)=1.
end do                                         Host code

!$omp target teams distribute
parallel do map(to:a,b) map(tofrom:c)
  do i=1,100
    do j=1,100
      c(i,j) = a(i)*b(j)
    end do
  end do
!$omp end target teams distribute
parallel do
  write(*,*)c

```



# Numerical calculation of linear absorption spectrum

In the weak excitation limit,  $\rho_{cc}(\mathbf{k}) \simeq 0$ ,  $\rho_{vv}(\mathbf{k}) \simeq 1$ , the interband coherence  $\rho_{vc}(\mathbf{k})$  is obtained by

$$\begin{aligned} \frac{d}{dt} \rho_{vc}(\mathbf{k}) &= \frac{i}{\hbar} \left( \varepsilon_{c,\mathbf{k}} - \varepsilon_{v,\mathbf{k}} + \frac{i\hbar}{T_2} \right) \rho_{vc}(\mathbf{k}) + \frac{i}{\hbar} e \mathbf{E}(t) \cdot \boldsymbol{\xi}_{vc}(\mathbf{k}) \\ &\quad - \frac{i}{\hbar} \sum_{c,v,\mathbf{k}'} V_{v'cc'v}(\mathbf{k}, \mathbf{k}', \mathbf{k}' - \mathbf{k}) \rho_{v'c'}(\mathbf{k}') \end{aligned}$$

Optical polarization:

$$\mathbf{P}(t) = \frac{e}{L^2} \sum_{c,v,\mathbf{k}} \boldsymbol{\xi}_{cv}(\mathbf{k}) \rho_{vc}(\mathbf{k})$$

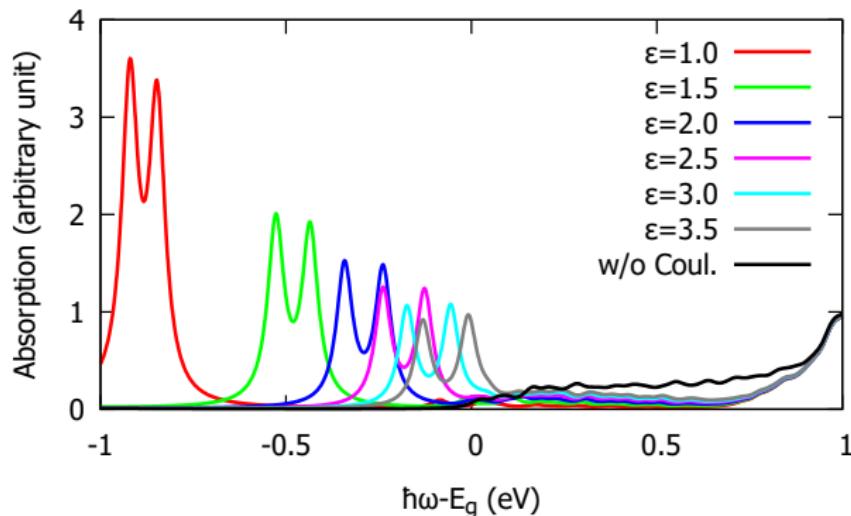
In the linear optical regime:

$$\mathbf{e} \cdot \mathbf{P}(\omega) = \chi(\omega) E(\omega)$$

Linear absorption coefficient:

$$\alpha(\omega) \propto \text{Im} \chi(\omega) = \text{Im} \left[ \frac{\mathbf{e} \cdot \mathbf{P}(\omega)}{E(\omega)} \right]$$

# Numerical calculation of linear absorption spectrum



Calculated linear absorption spectrum of MoS<sub>2</sub> monolayer for different  $\epsilon$

$\epsilon$	1.0	1.5	2.0	2.5	3.0	3.5
$E_b$ (eV)	0.92	0.53	0.34	0.24	0.17	0.13

Exciton binding energy extracted from the linear absorption spectrum

# Summary

- We study the optoelectronic responses of optically excited monolayer TMD systems
- Our approach is based on multi-band semiconductor Bloch equations in combination with a three-band tight-binding model
- The Coulomb interaction between electrons is taken into account using the time-dependent Hartree-Fock approximation
- We numerically solve the dynamical equations in the entire two-dimensional Brillouin zone
- Our Fortran programs are developed for parallel computation on both CPU and GPU
- The initial result of calculating the linear absorption spectrum is presented