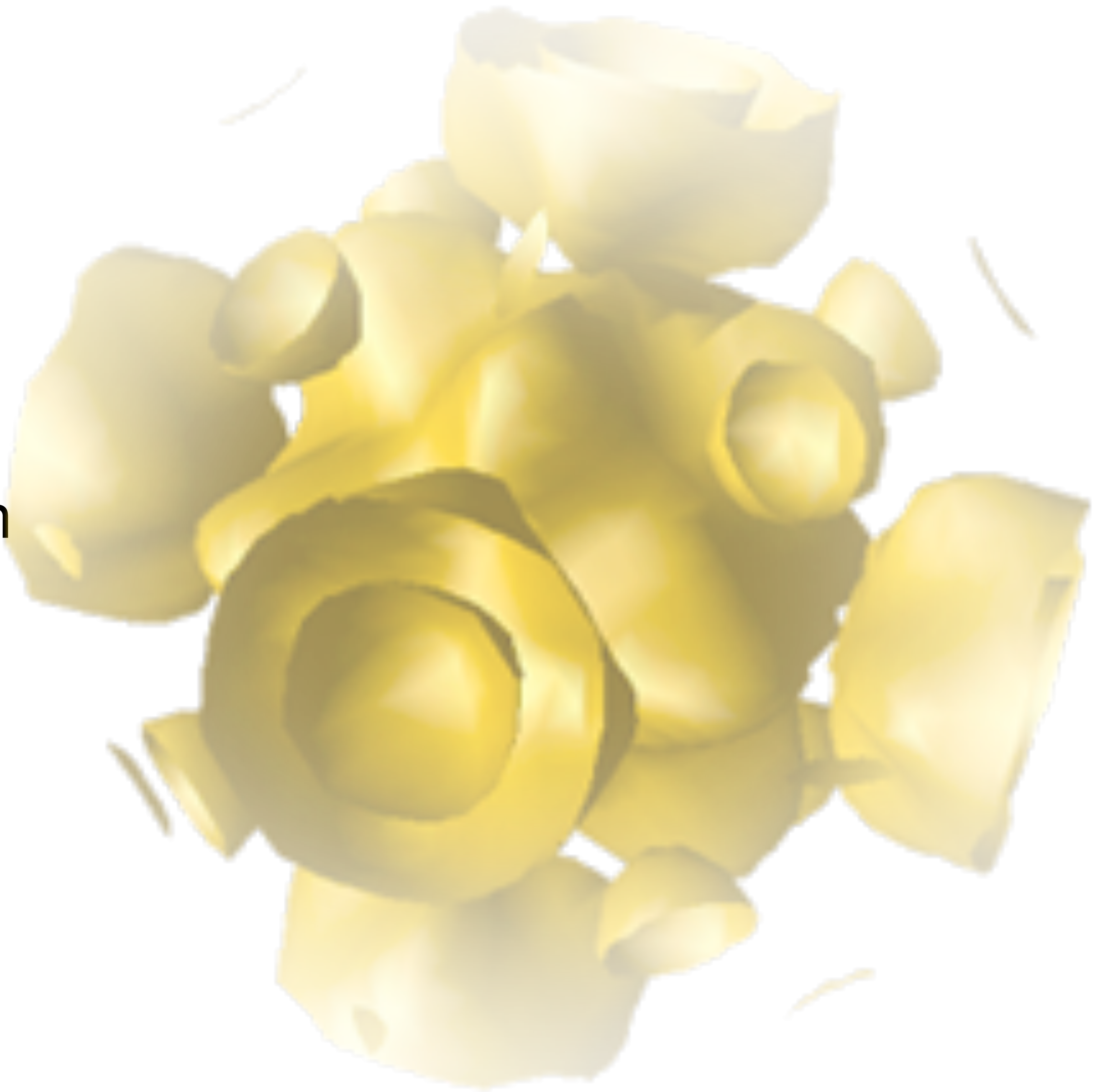
BSE in `exciting`

Solve the **B**ethe-**S**alpeter **E**quation

Benedikt Maurer, HU Berlin

Outline

- BSE
- Implementation
- Usage
- Features
- News



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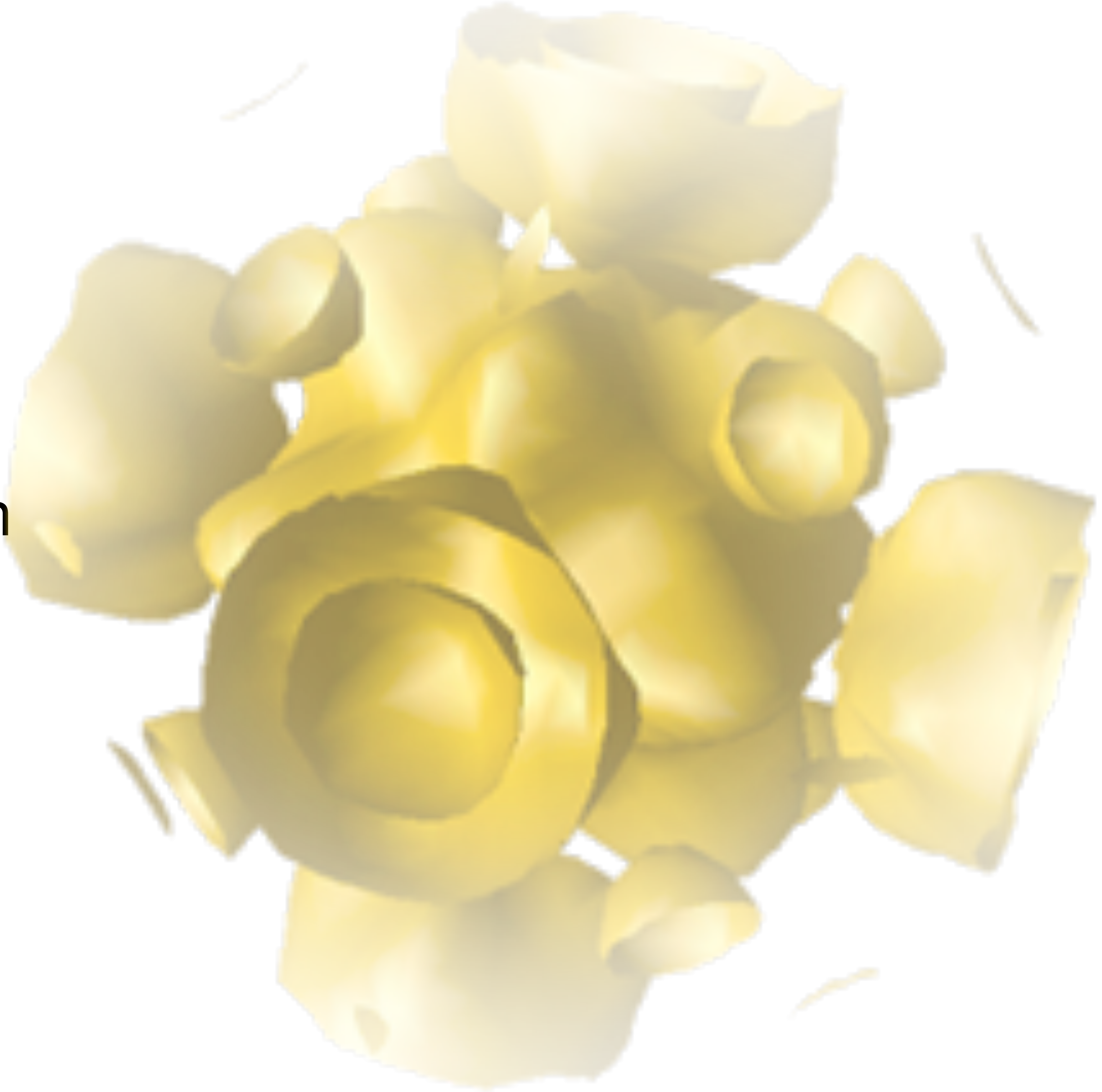
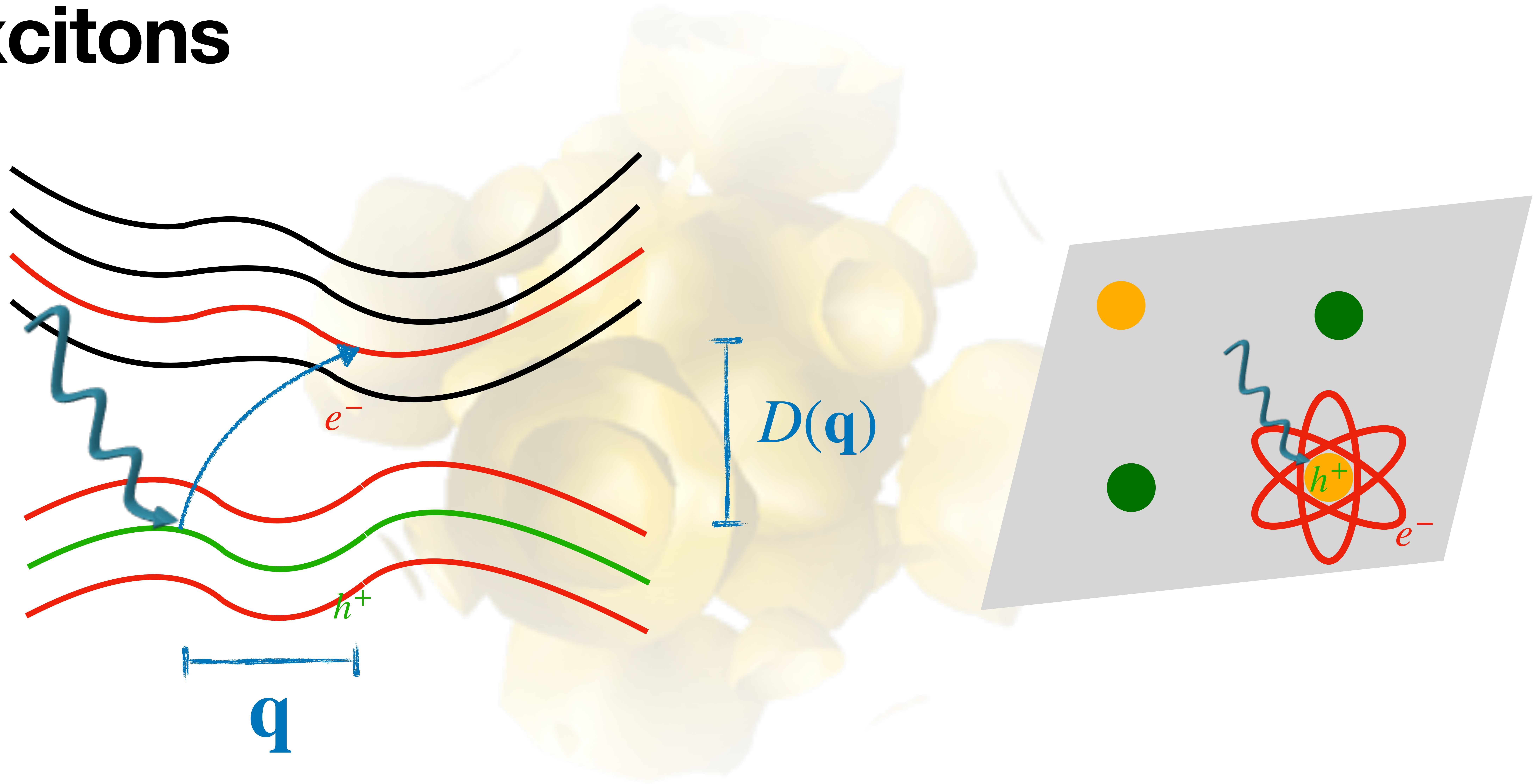
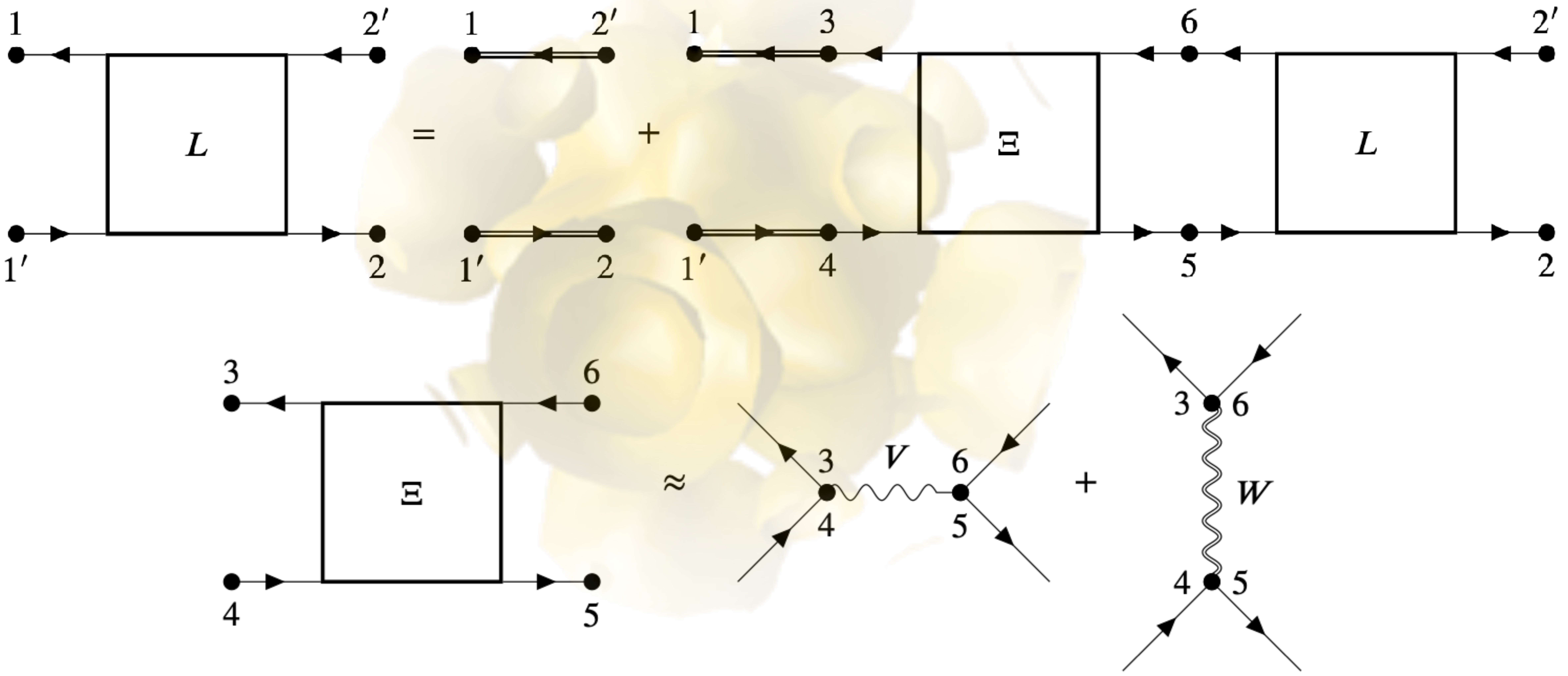


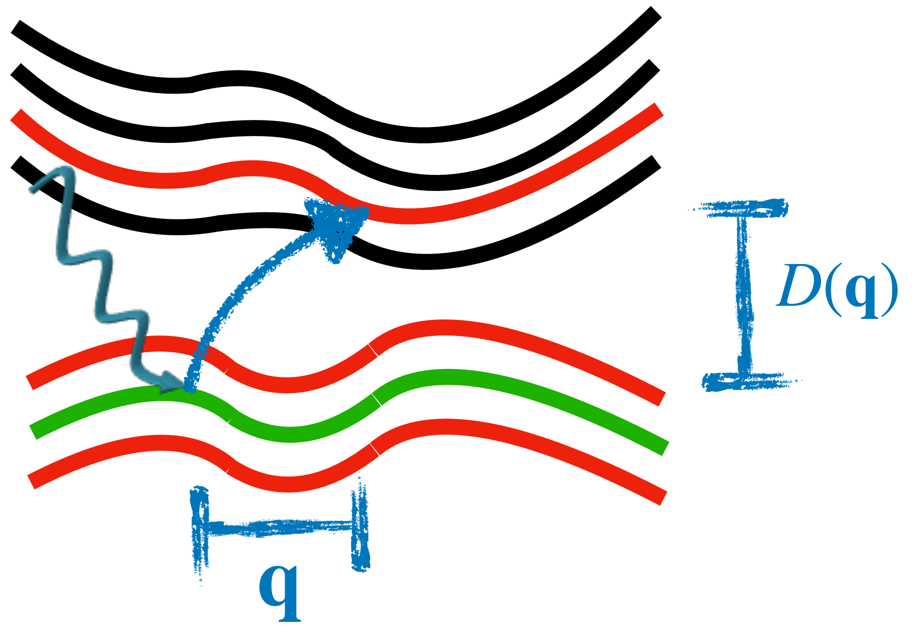
Excitons



$e - h$ Correlation Function



From L to H



$$\Rightarrow L(\mathbf{q},\omega) = \left[L_0^{-1}(\mathbf{q}) - \Xi(\mathbf{q})\right]^{-1}$$

$$\Rightarrow L_0^{-1}(\mathbf{q},\omega) = \left[\begin{pmatrix} D(\mathbf{q}) & 0 \\ 0 & D(\mathbf{q}) \end{pmatrix} - \omega \begin{pmatrix} \mathbf{I} & 0 \\ 0 & -\mathbf{I} \end{pmatrix}\right]$$

$$\Rightarrow L(\mathbf{q},\omega) = -\left[H(\mathbf{q}) - \omega\Delta\right]^{-1}$$

$$\Upsilon^{r}_{ou\mathbf{k},\mathbf{q}}(\mathbf{r},\mathbf{r}') = \psi_{o\mathbf{k}+\frac{\mathbf{q}}{2}}(\mathbf{r})\psi^{*}_{u\mathbf{k}-\frac{\mathbf{q}}{2}}(\mathbf{r}')$$

$$H(\mathbf{q}) = \begin{pmatrix} A(\mathbf{q}) & \cancel{B(\mathbf{q})} \\ \cancel{B(\mathbf{q})} & A(\mathbf{q}) \end{pmatrix}$$

$$\mathbf{q} = \mathbf{0}$$

TDA

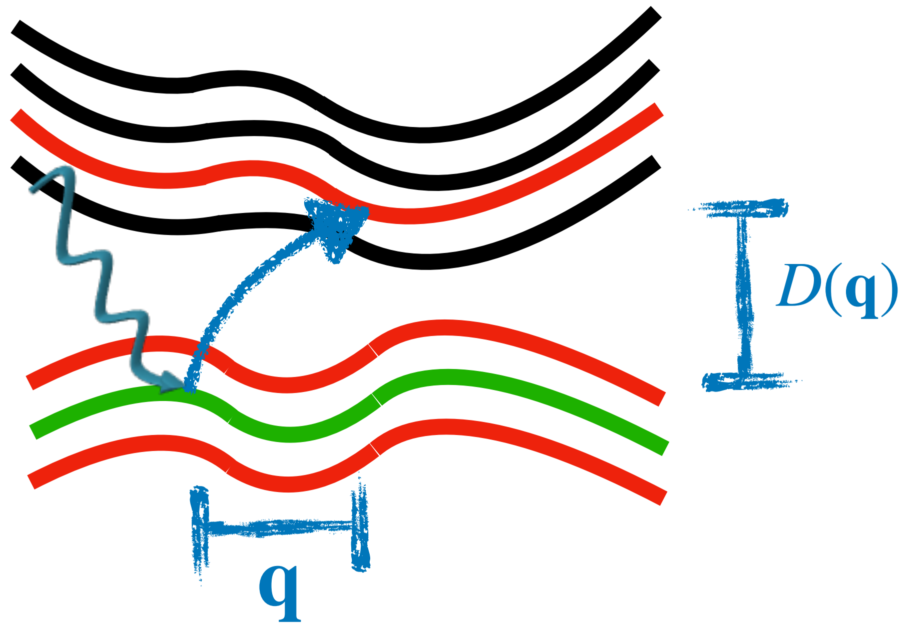
$$A(\mathbf{q}) = D(\mathbf{q}) + 2\gamma_x V^{rr}(\mathbf{q}) - \gamma_c W^{rr}(\mathbf{q})$$

$$\Rightarrow H := A(\mathbf{0})$$

~~$B(\mathbf{q}) = 2\gamma_x V^{rr}(\mathbf{q}) - \gamma_c W^{ra}(\mathbf{q})$~~

$$H|\phi^{\lambda}\rangle = E^{\lambda}|\phi^{\lambda}\rangle$$

The Bethe-Salpeter Hamiltonian



$$D_{ou\mathbf{k},o'u'\mathbf{k}'} = (\epsilon_{u\mathbf{k}} - \epsilon_{o\mathbf{k}})\delta_{oo'}\delta_{uu'}\delta_{\mathbf{k}\mathbf{k}'}$$

$$V_{ou\mathbf{k},o'u'\mathbf{k}'} = \iint d^3\mathbf{r}d^3\mathbf{r}'\,\psi^*_{o\mathbf{k}}(\mathbf{r})\,\psi_{u\mathbf{k}}(\mathbf{r})\,V(\mathbf{r},\mathbf{r}')\,\psi_{o'\mathbf{k}'}(\mathbf{r}')\,\psi^*_{u'\mathbf{k}'}(\mathbf{r}')$$

$$W_{ou\mathbf{k},o'u'\mathbf{k}'} = \iint d^3\mathbf{r}d^3\mathbf{r}'\,\psi_{o\mathbf{k}}(\mathbf{r})\,\psi^*_{o'\mathbf{k}'}(\mathbf{r})\,W(\mathbf{r},\mathbf{r}')\,\psi_{u'\mathbf{k}'}(\mathbf{r}')\,\psi^*_{u\mathbf{k}}(\mathbf{r}')$$

$$\Rightarrow \sum_{o'u'\mathbf{k}'} H^{BSE}_{ou\mathbf{k},o'u'\mathbf{k}'}A^{\lambda}_{o'u'\mathbf{k}'} = E^{\lambda}A^{\lambda}_{ou\mathbf{k}}$$

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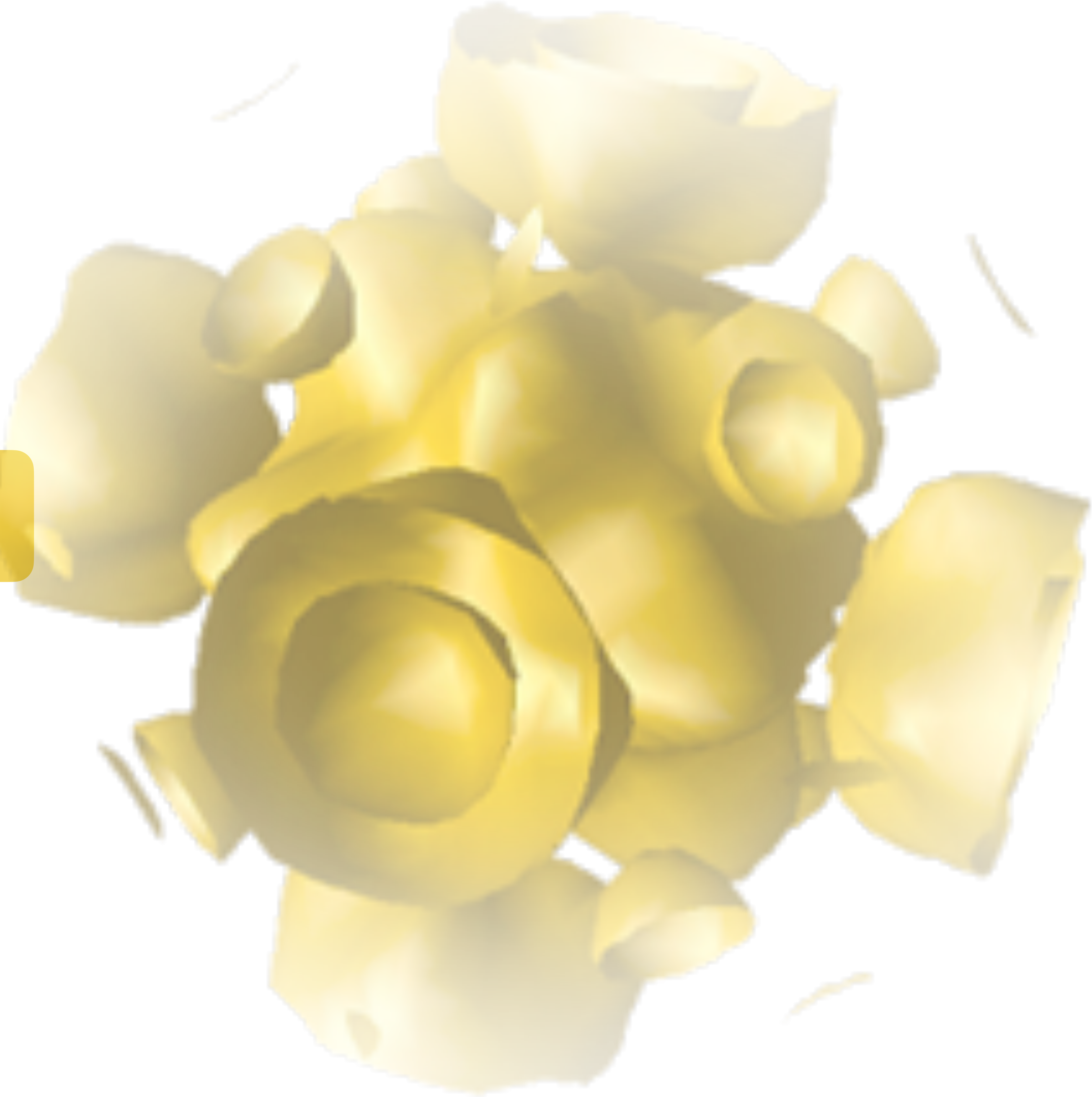
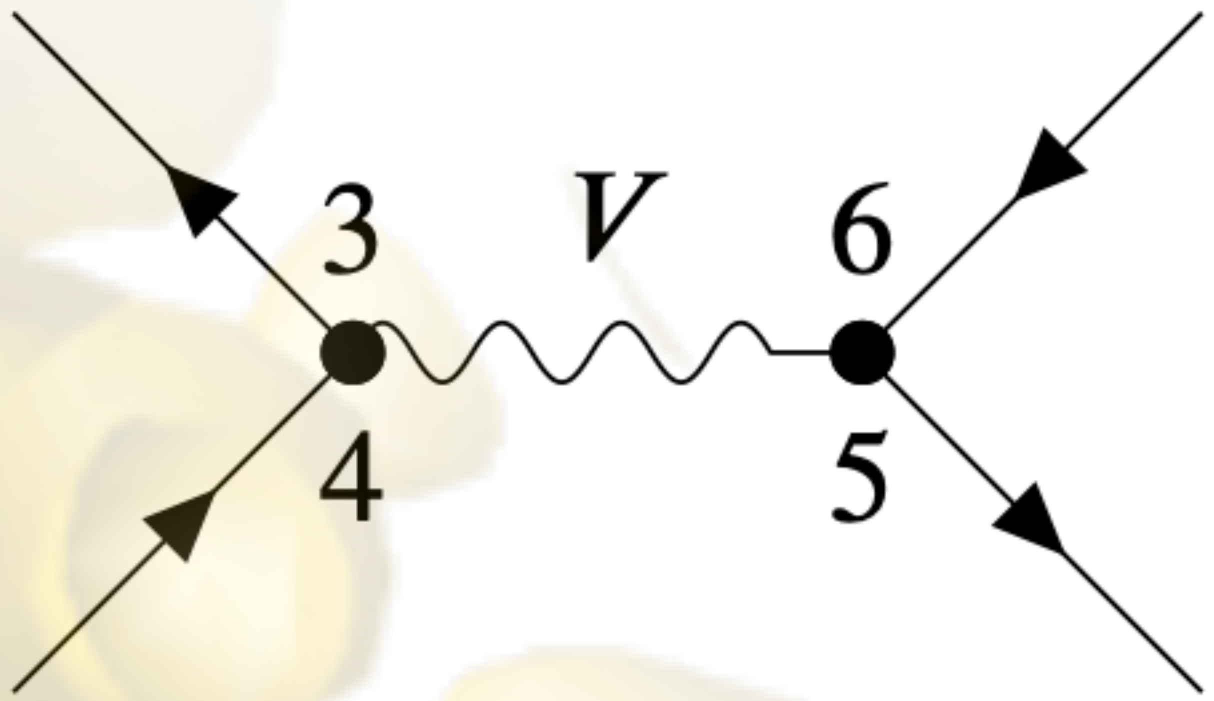
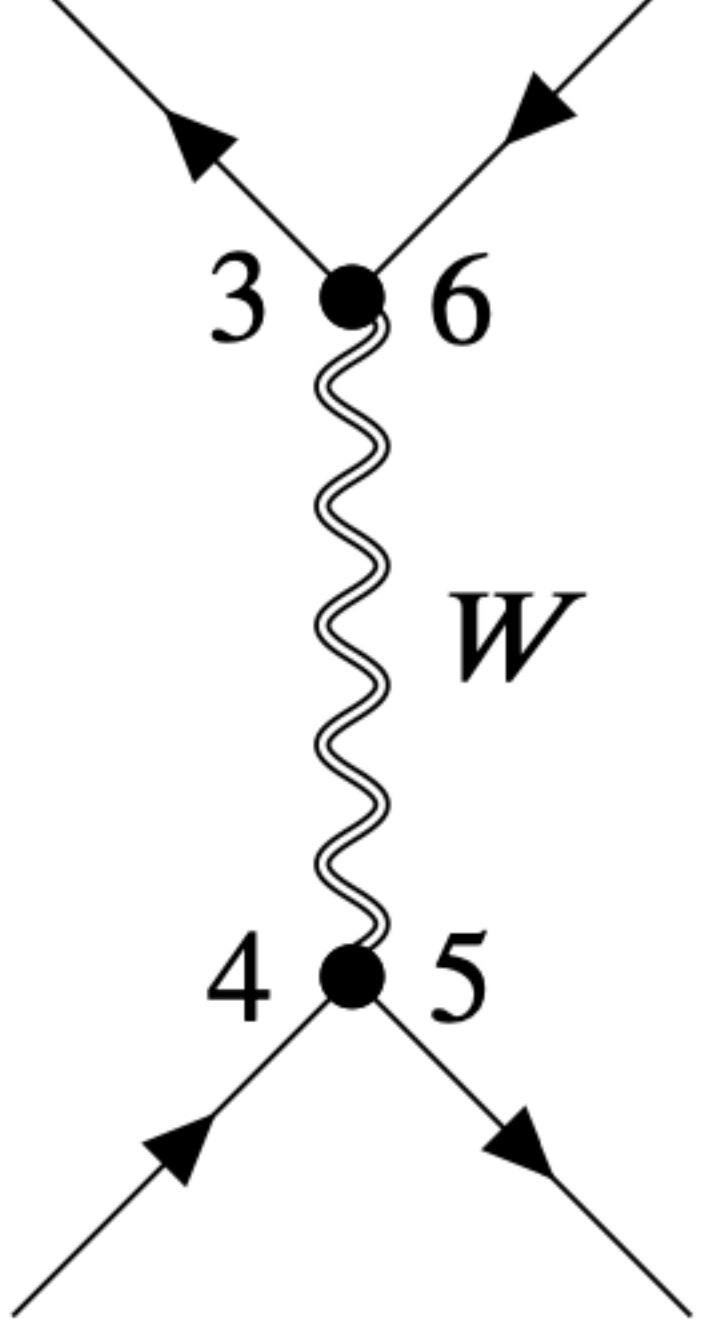


Interaction Kernels

$$M_{ij\mathbf{k}}(\mathbf{G},\mathbf{q}) = \langle i\mathbf{k}\,|\,e^{-i(\mathbf{k}+\mathbf{G})\mathbf{q}}\,|\,j(\mathbf{k}+\mathbf{q})\rangle$$





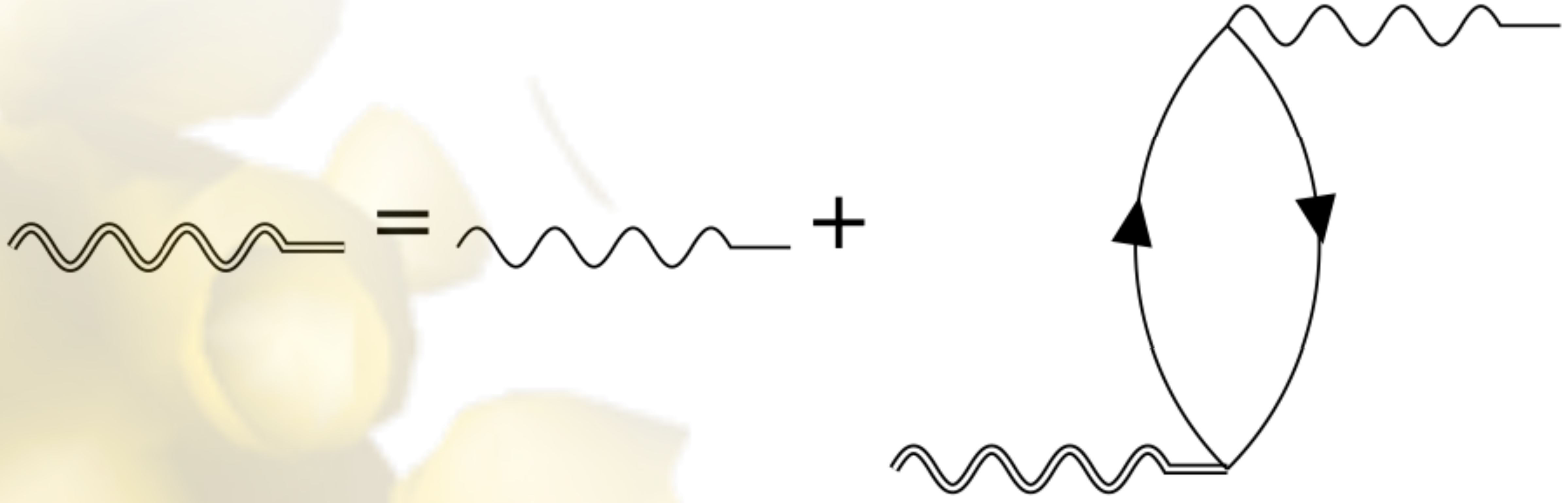
$$V_{ou\mathbf{k},o'u'\mathbf{k}'} = \sum_{\mathbf{G}}^{|\mathbf{G}+\mathbf{q}|_{max}} M^{*}_{ou\mathbf{k}}(\mathbf{G},\mathbf{0})\tilde{V}_{\mathbf{q}=\mathbf{0}}(\mathbf{G})M^{*}_{ou\mathbf{k}}(\mathbf{G},\mathbf{0})$$

$$W_{ou\mathbf{k},o'u'\mathbf{k}'} = \sum_{\mathbf{G}\mathbf{G}'}^{|\mathbf{G}+\mathbf{q}|_{max}} M^{*}_{oo'\mathbf{k}}(\mathbf{G},\mathbf{k}'-\mathbf{k})(\mathbf{G})\tilde{W}_{\mathbf{k}-\mathbf{k}'}(\mathbf{G},\mathbf{G}')M^{*}_{uu'\mathbf{k}}(\mathbf{G},\mathbf{k}'-\mathbf{k})$$

C. Vorwerk, B. Aurich, C. Cocchi, C. Draxl, Electronic Structure, (2019)

Screening

$$\tilde{V}_{\mathbf{q}}(\mathbf{G}) = \frac{1}{\Omega} \frac{4\pi}{|\mathbf{G} + \mathbf{q}|}$$



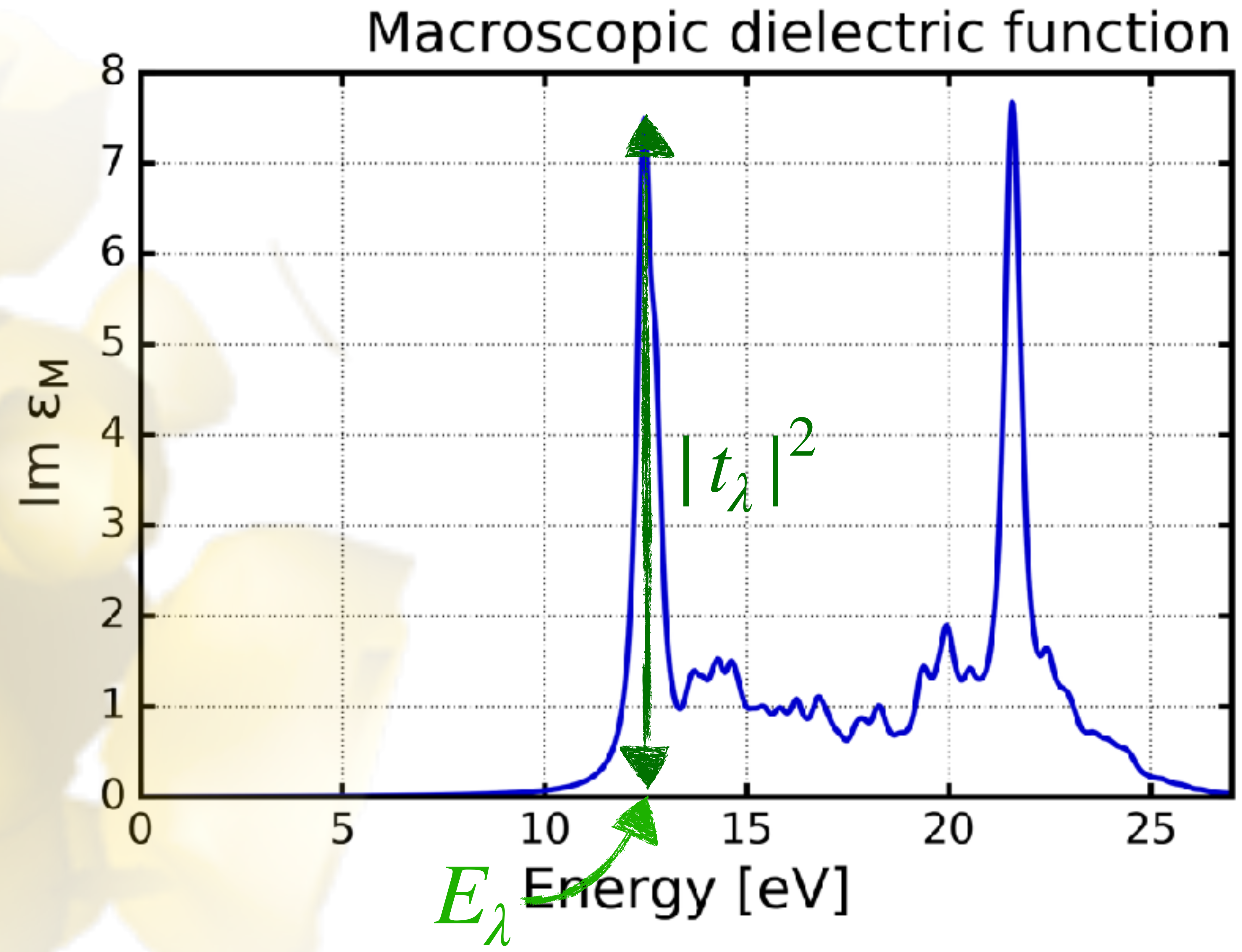
$$\tilde{W}_{\mathbf{q}}(\mathbf{G}, \mathbf{G}') = V_{\mathbf{q}}(\mathbf{G}) \left[\varepsilon^{RPA}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega = 0) \right]^{-1}$$

$$\varepsilon^{RPA}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = \delta_{\mathbf{G}\mathbf{G}'} - \sum_{ij\mathbf{k}} \frac{f(\epsilon_{j\mathbf{k}+\mathbf{q}}) - f(\epsilon_{i\mathbf{k}})}{\epsilon_{j\mathbf{k}+\mathbf{q}} - \epsilon_{i\mathbf{k}} - \omega} \left[M_{ij\mathbf{k}}(\mathbf{G}, \mathbf{q}) \right]^* M_{ij\mathbf{k}}(\mathbf{G}', \mathbf{q})$$

C. Vorwerk, B. Aurich, C. Cocchi, C. Draxl, Electronic Structure, (2019)

Post Processing

$$D_{ou\mathbf{k},j} = i\frac{\langle \psi_{o\mathbf{k}} | p_j | \psi_{u\mathbf{k}} \rangle}{\epsilon_{u\mathbf{k}} - \epsilon_{o\mathbf{k}}}$$

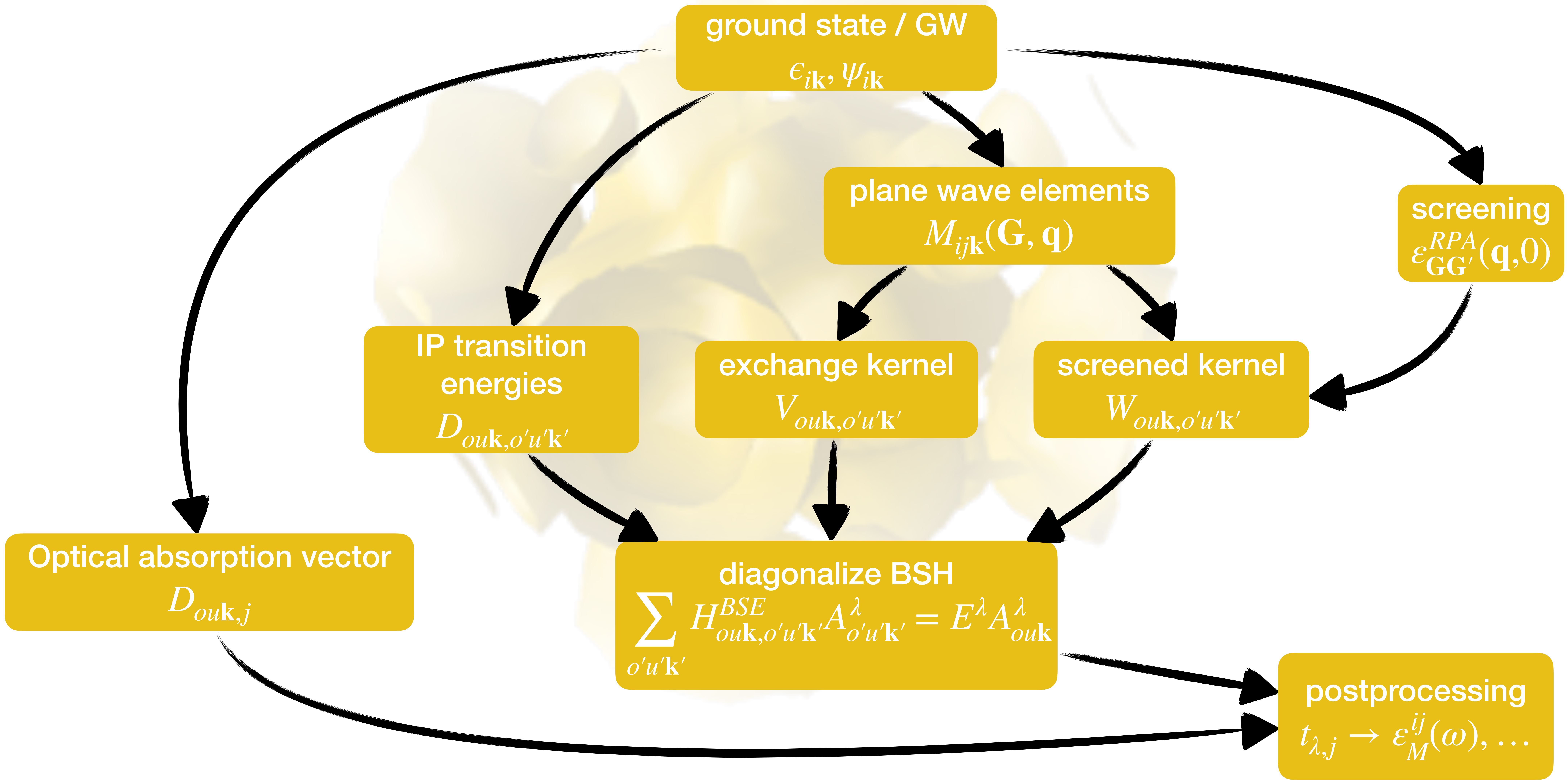


$$t_{\lambda,j} = -\, i \sum_{uo\mathbf{k}} A^{\lambda}_{uo\mathbf{k}} D_{ou\mathbf{k},j}$$

$$\varepsilon^{ij}_{M}(\omega) = \delta_{ij} - 4\pi \sum_{\lambda} \left(\frac{t^{*}_{\lambda,i} t_{\lambda,j}}{\omega - E_{\lambda} + i\delta} + \frac{t^{*}_{\lambda,i} t_{\lambda,j}}{-\omega - E_{\lambda} + i\delta} \right)$$

C. Vorwerk, B. Aurich, C. Cocchi, C. Draxl, Electronic Structure, (2019)

Workflow in `exciting`



C. Vorwerk, B. Aurich, C. Cocchi, C. Draxl, Electronic Structure, (2019)

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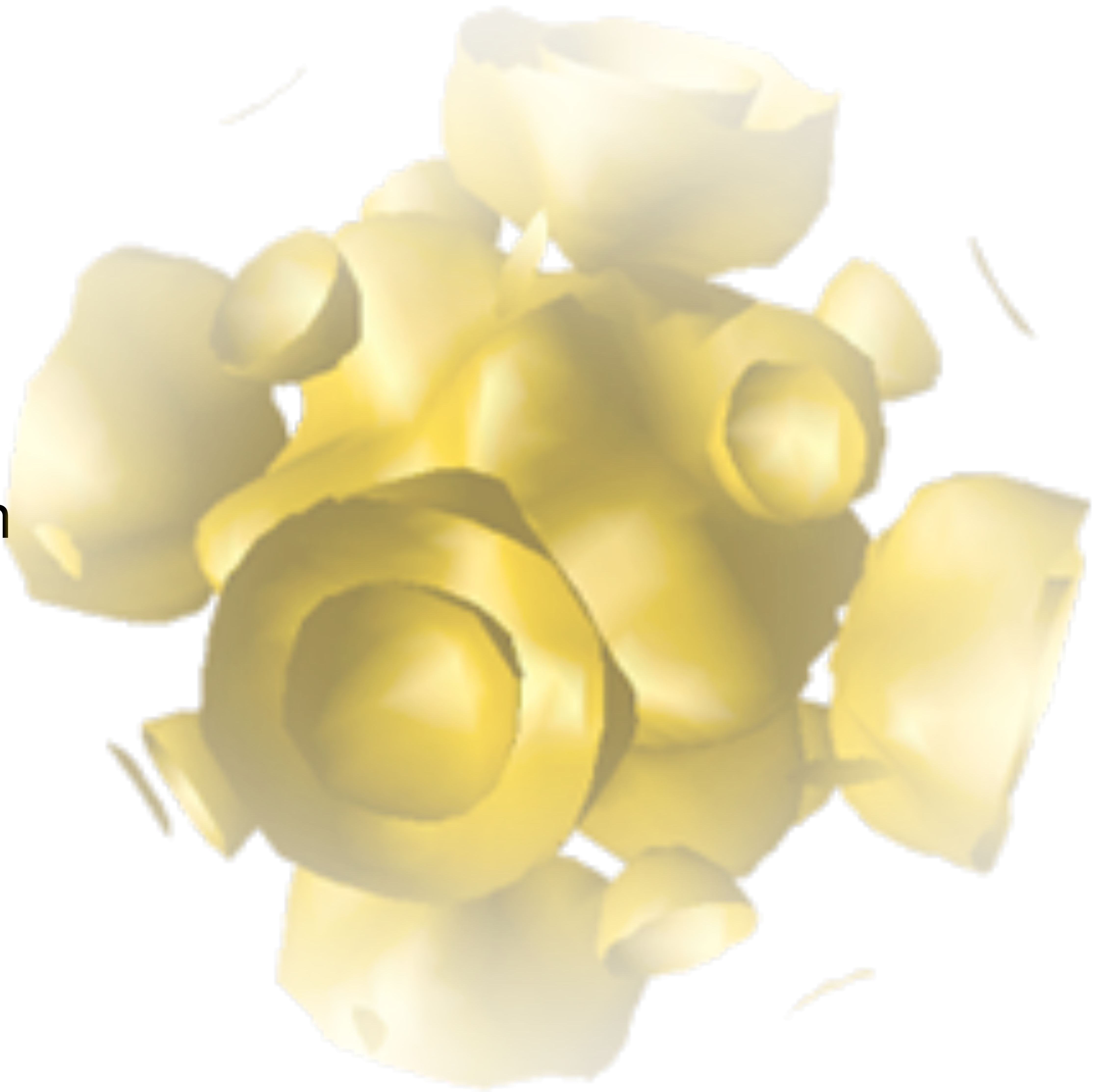

input.xml

- General settings
- Settings for ω
- Settings for the screening
- Settings for the BSE
- Momentum transfer

input.xml

```
...
   <xs
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      ngridk="3 3 3"
      vkloff="0.097 0.273 0.493"
      ngridq="3 3 3"
      nempty="30"
      gqmax="2.5"
      broad="0.007"
      scissor="0.20947"
      tappinfo="true"
      tevout="true">

      <energywindow
         intv="0.0 1.0"
         points="1200"/>

      <screening
         screentype="full"
         nempty="100"/>

      <BSE
         bsetype="singlet"
         nstlbse="1 5 1 4"  />

      <qpointset>
         <qpoint>0.0 0.0 0.0</qpoint>
      </qpointset>

   </xs>
...
```

Parameters

$$H = D + 2\gamma_x V - \gamma_c W$$

$\rightarrow \gamma_x = \gamma_c = 1$

$\epsilon_{i\mathbf{k}}, \psi_{i\mathbf{k}}$ → $i = 1,\ldots,$nempty

$$D_{ou\mathbf{k},o'u'\mathbf{k}'} = (\epsilon_{u\mathbf{k}} - \epsilon_{o\mathbf{k}})\delta_{oo'}\delta_{uu'}\delta_{\mathbf{k}\mathbf{k}'} + \Delta$$

$$V_{ou\mathbf{k},o'u'\mathbf{k}'} = \sum_{\mathbf{G}}^{|\mathbf{G}+\mathbf{q}|_{max}} M^*_{ou\mathbf{k}}(\mathbf{G},\mathbf{0})\tilde{V}_{\mathbf{q}=0}(\mathbf{G})M^*_{ou\mathbf{k}}(\mathbf{G},\mathbf{0})$$

plane wave cutoff

$$W_{ou\mathbf{k},o'u'\mathbf{k}'} = \sum_{\mathbf{G}\mathbf{G}'}^{|\mathbf{G}+\mathbf{q}|_{max}} M^*_{oo'\mathbf{k}}(\mathbf{G},\mathbf{k}'-\mathbf{k})(\mathbf{G})\tilde{W}_{\mathbf{k}-\mathbf{k}'}(\mathbf{G},\mathbf{G}')M^*_{uu'\mathbf{k}}(\mathbf{G},\mathbf{k}'-\mathbf{k})$$

→ $i,j = 1,\ldots,$nempty

$$\varepsilon^{RPA}_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega=0) = \delta_{\mathbf{G}\mathbf{G}'} - \sum_{ij\mathbf{k}} \frac{f(\epsilon_{j\mathbf{k}+\mathbf{q}}) - f(\epsilon_{i\mathbf{k}})}{\epsilon_{j\mathbf{k}+\mathbf{q}} - \epsilon_{i\mathbf{k}}} \left[M_{ij\mathbf{k}}(\mathbf{G},\mathbf{q})\right]^* M_{ij\mathbf{k}}(\mathbf{G}',\mathbf{q})$$

input.xml

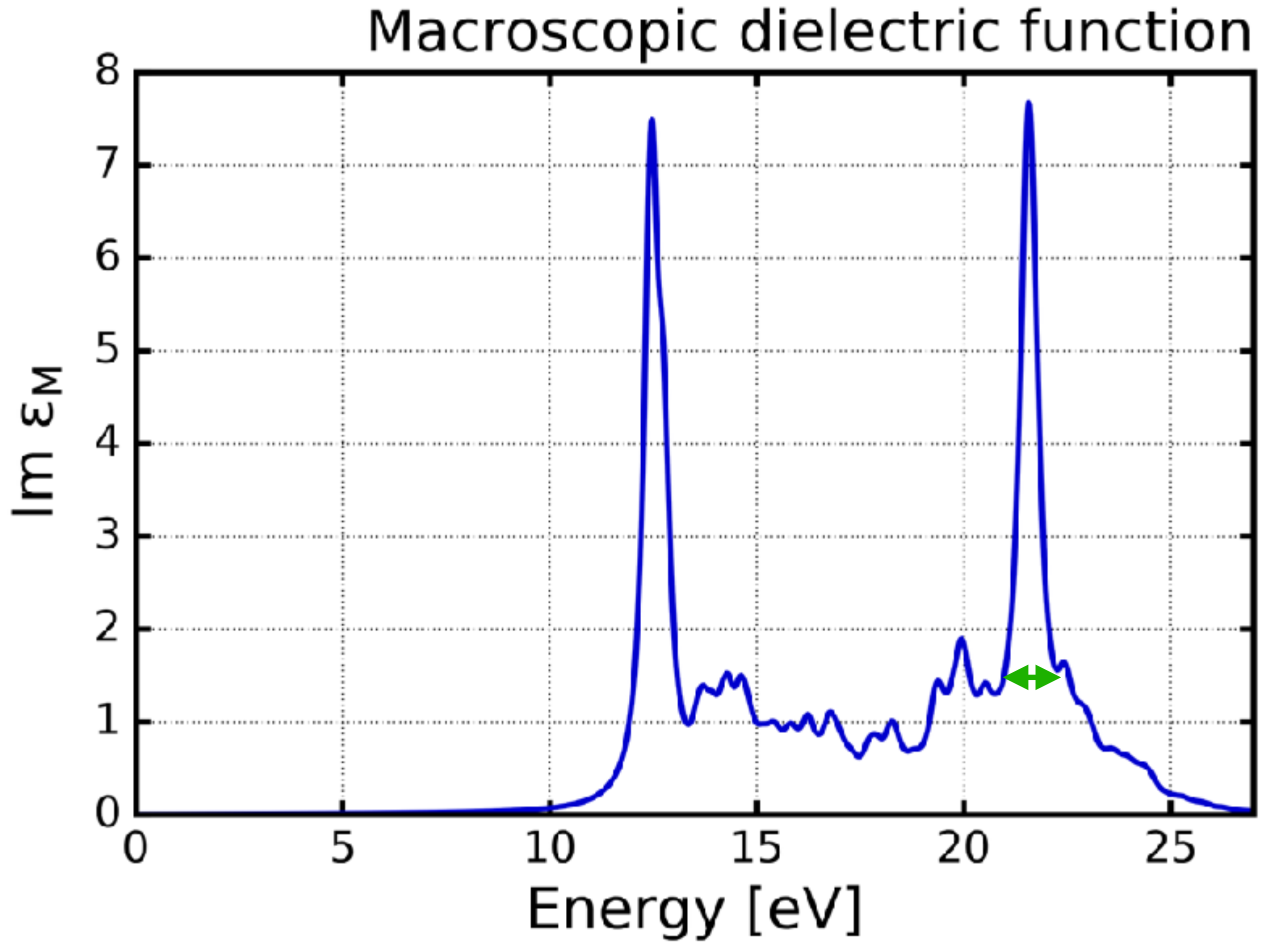
```
...
  <xs
     xstype="BSE"
     ngridk="3 3 3"
     vkloff="0.097 0.273 0.493"
     ngridq="3 3 3"
     nempty="30"
     gqmax="2.5"
     broad="0.007"
     scissor="0.20947"
     tappinfo="true"
     tevout="true">

     <energywindow
        intv="0.0 1.0"
        points="1200"/>

     <screening
        screentype="full"
        nempty="100"/>

     <BSE
        bsetype="singlet"
        nstlbse="1 5 1 4" />

     <qpointset>
        <qpoint>0.0 0.0 0.0</qpoint>
     </qpointset>

  </xs>
...
```

Parameters

Γ must be always first!!!

$$L(\blacksquare,\blacksquare) = -\left[H(\blacksquare) - \blacksquare\Delta\right]^{-1}$$

$$\varepsilon_M^{ij}(\blacksquare) = \delta_{ij} - 4\pi \sum_{\lambda} \left(\frac{t^*_{\lambda,i} t_{\lambda,j}}{\blacksquare - E_\lambda + i\delta} + \frac{t^*_{\lambda,i} t_{\lambda,j}}{-\blacksquare - E_\lambda + i\delta} \right)$$



`intv` $\rightarrow [\omega_{min}, \omega_{max}]$

`points` $\rightarrow \#\omega$

input.xml

```xml
...
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     xstype="BSE"
     ngridk="3 3 3"
     vkloff="0.097 0.273 0.493"
     ngridq="3 3 3"
     nempty="30"
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        screentype="full"
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     <BSE
        bsetype="singlet"
        nstlbse="1 5 1 4" />

     <qpointset>
        <qpoint>0.0 0.0 0.0</qpoint>
     </qpointset>

  </xs>
...
```

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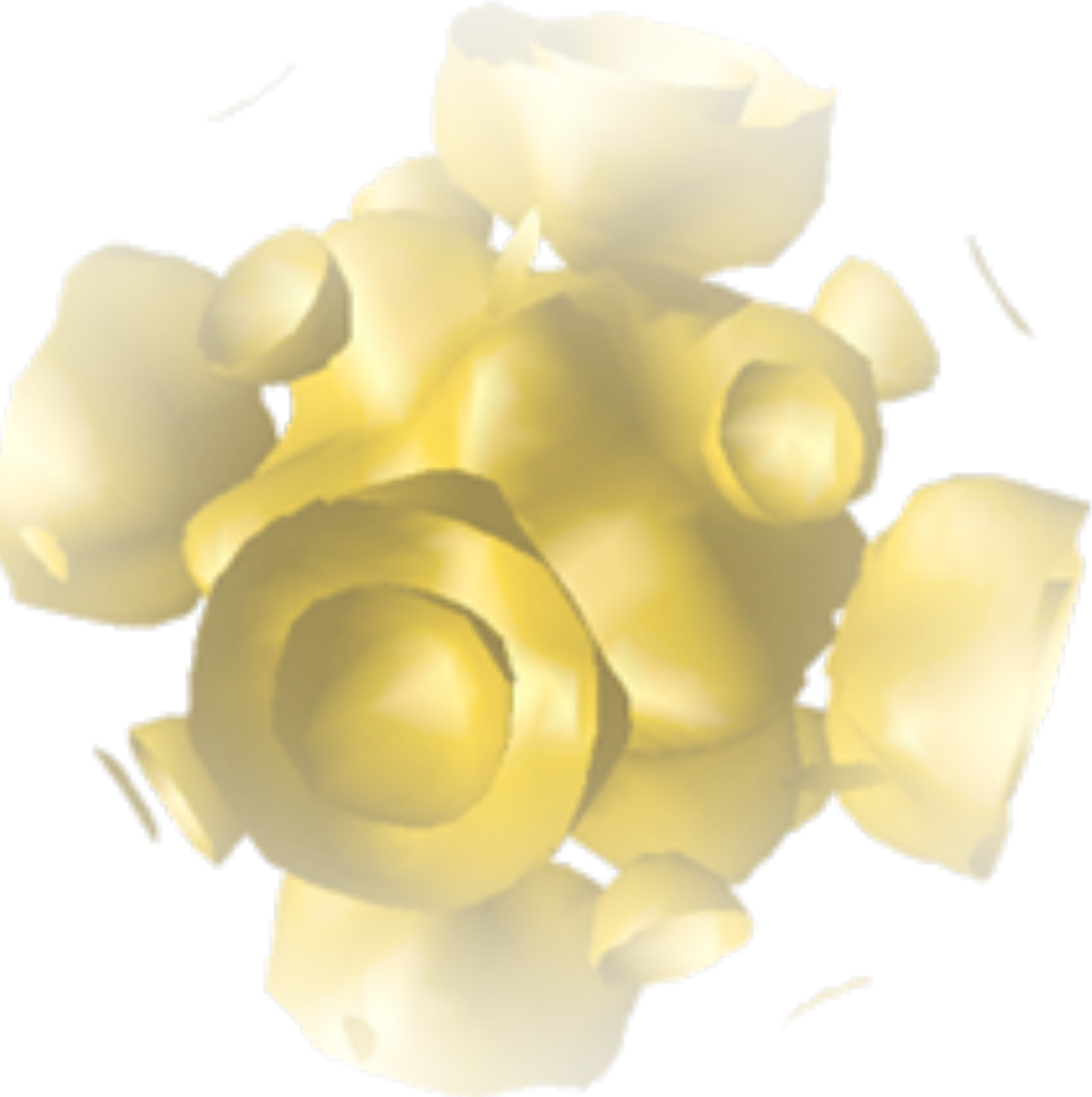
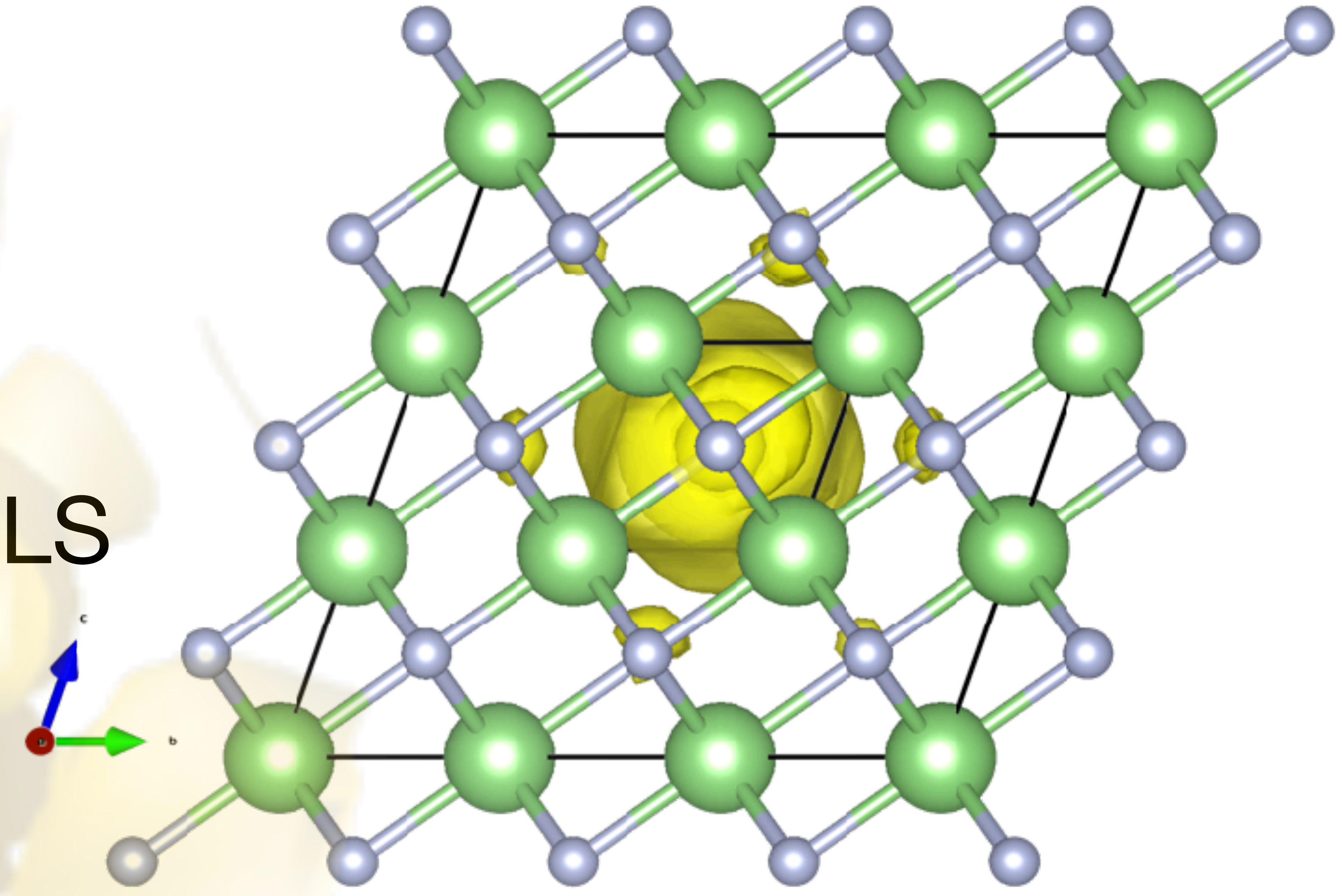


Features

- TDA and non TDA $\Rightarrow B(\mathbf{q}) \neq 0$
- X-ray excitations → XAS, XES, XANES, EELS
- Exciton analysis tools
- Finite momentum transfer $\Rightarrow \mathbf{q} \neq 0$
- Additive screening
- Fast solver $\rightarrow \mathcal{O}(N_k \log N_k + N_e^2)$ New!!!
- Detailed tutorials for everything



BSE:

- 【b】 Excited states from BSE
- 【a】 Exciton analysis and visualization
- 【a】 X-ray absorption spectra using BSE
- 【a】 X-ray emission spectra
- 【a】 q-dependent BSE calculations
- 【a】 Additive screening for interface systems

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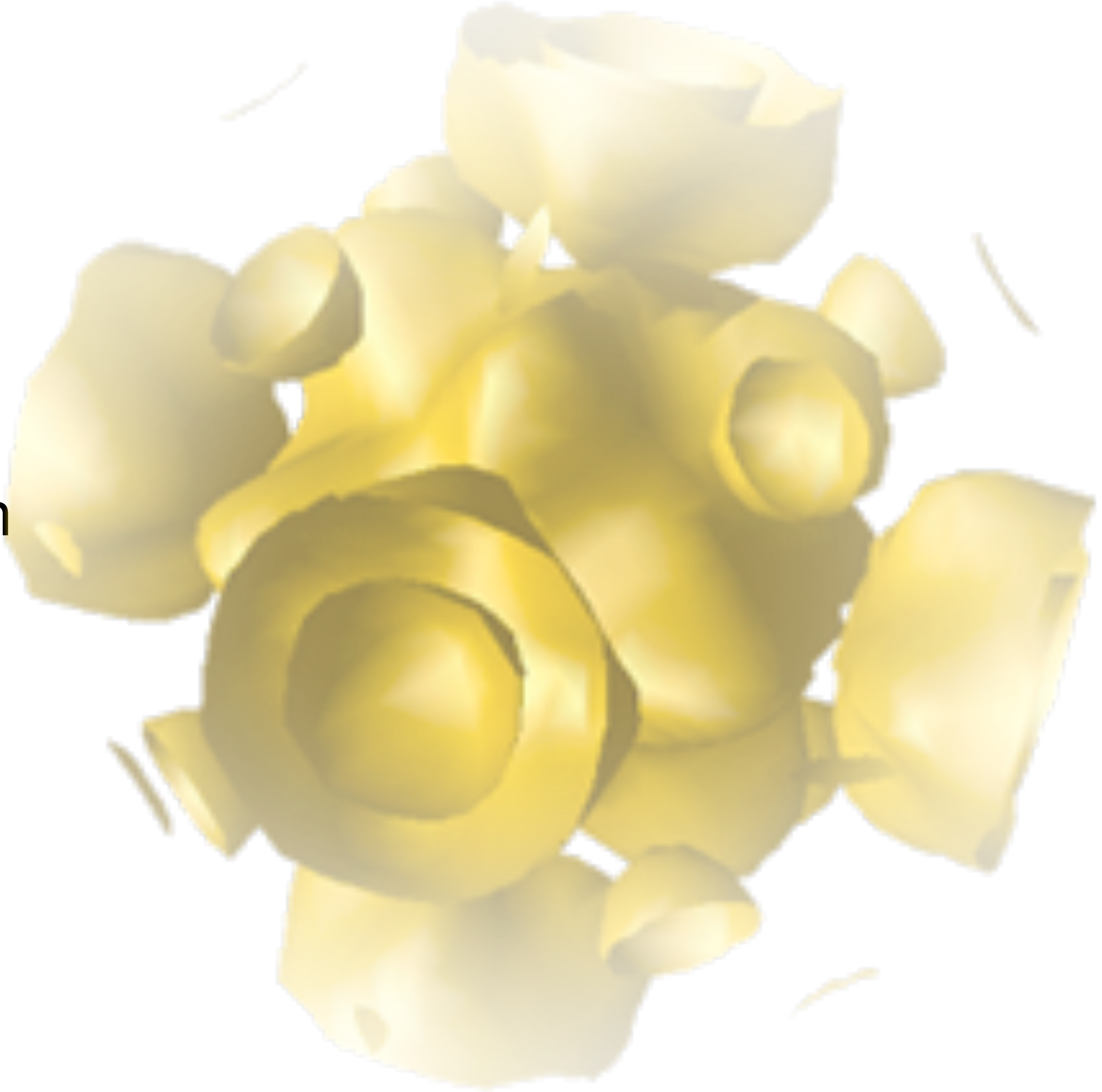
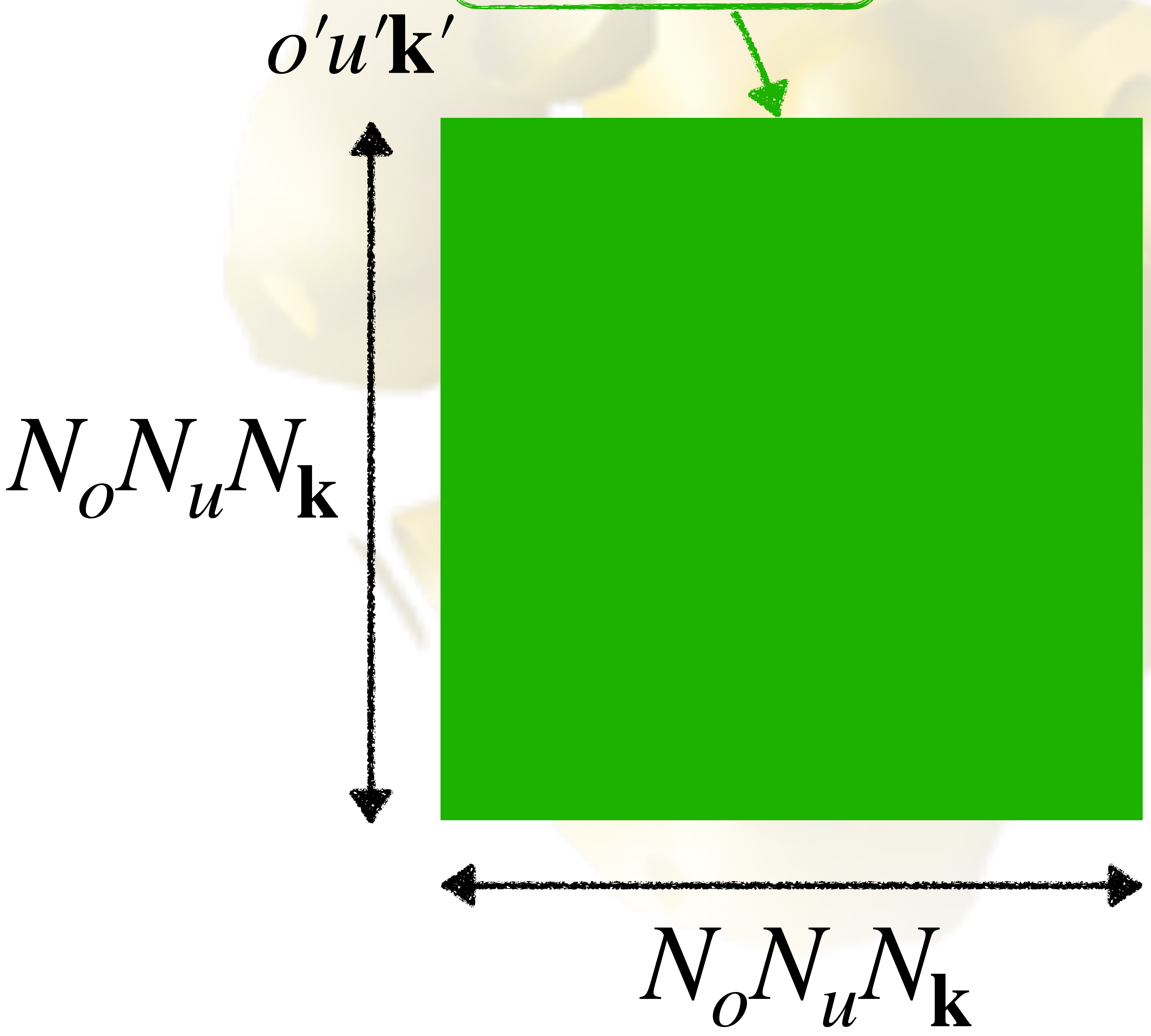


Scaling problem

$$\sum_{o'u'\mathbf{k}'} H^{BSE}_{ou\mathbf{k},o'u'\mathbf{k}'} A^{\lambda}_{o'u'\mathbf{k}'} = E^{\lambda} A^{\lambda}_{ou\mathbf{k}}$$



Setting the Hamiltonian up:
$\mathcal{O}(N_o^2 N_u^2 N_\mathbf{k}^2)$

Diagonalizing the Hamiltonian:
$\mathcal{O}(N_o^3 N_u^3 N_\mathbf{k}^3)$

$$\psi_{i\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{i\mathbf{k}}(\mathbf{r})$$

Scaling problem

$N_o N_u N_{\mathbf{k}}$

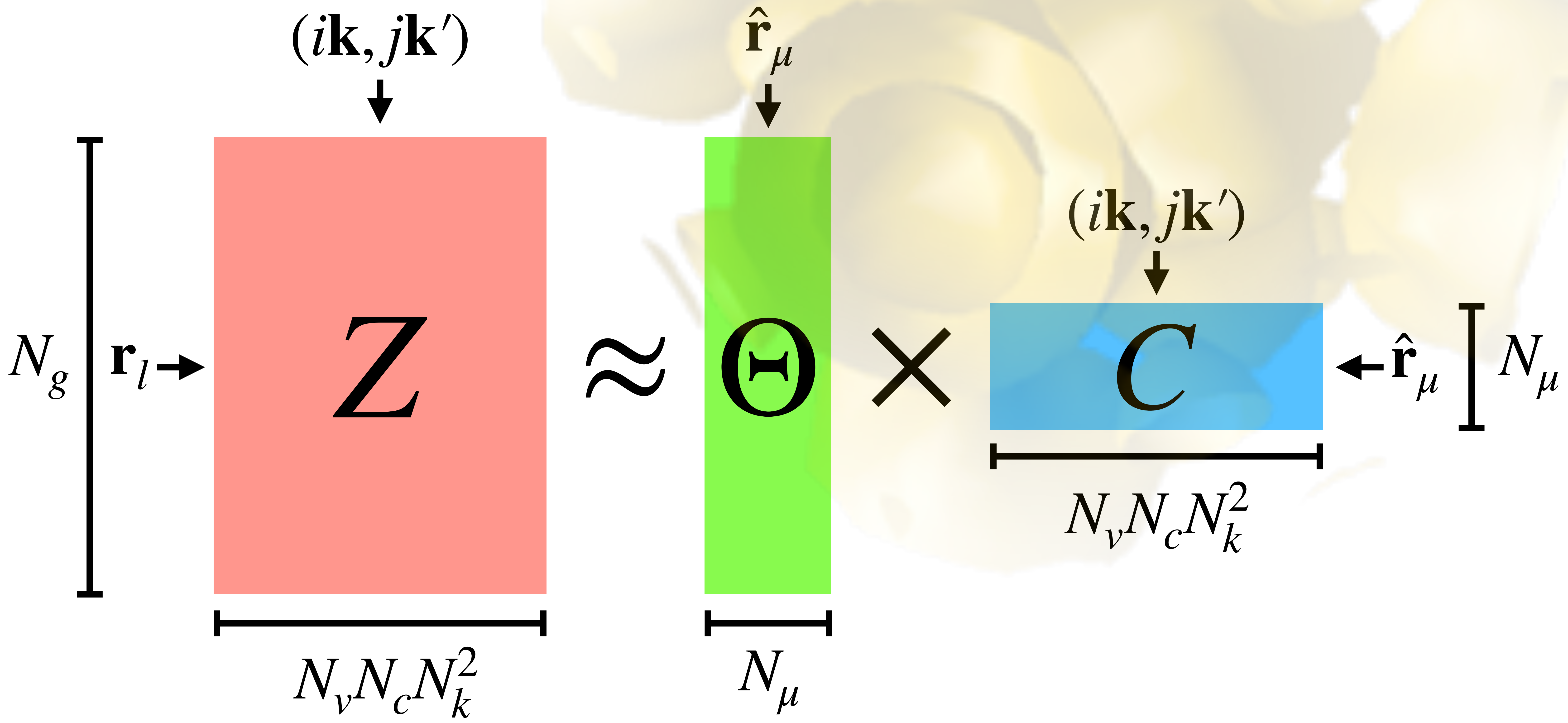
$$V_{ou\mathbf{k},o'u'\mathbf{k}'} = \frac{1}{N_k^2} \int_{\Omega^l \times \Omega^l} d\mathbf{r} d\mathbf{r}' u^*_{u\mathbf{k}}(\mathbf{r}) u_{o\mathbf{k}}(\mathbf{r}) V(\mathbf{r}, \mathbf{r}') u^*_{o'\mathbf{k}'}(\mathbf{r}') u_{u'\mathbf{k}'}(\mathbf{r}')$$

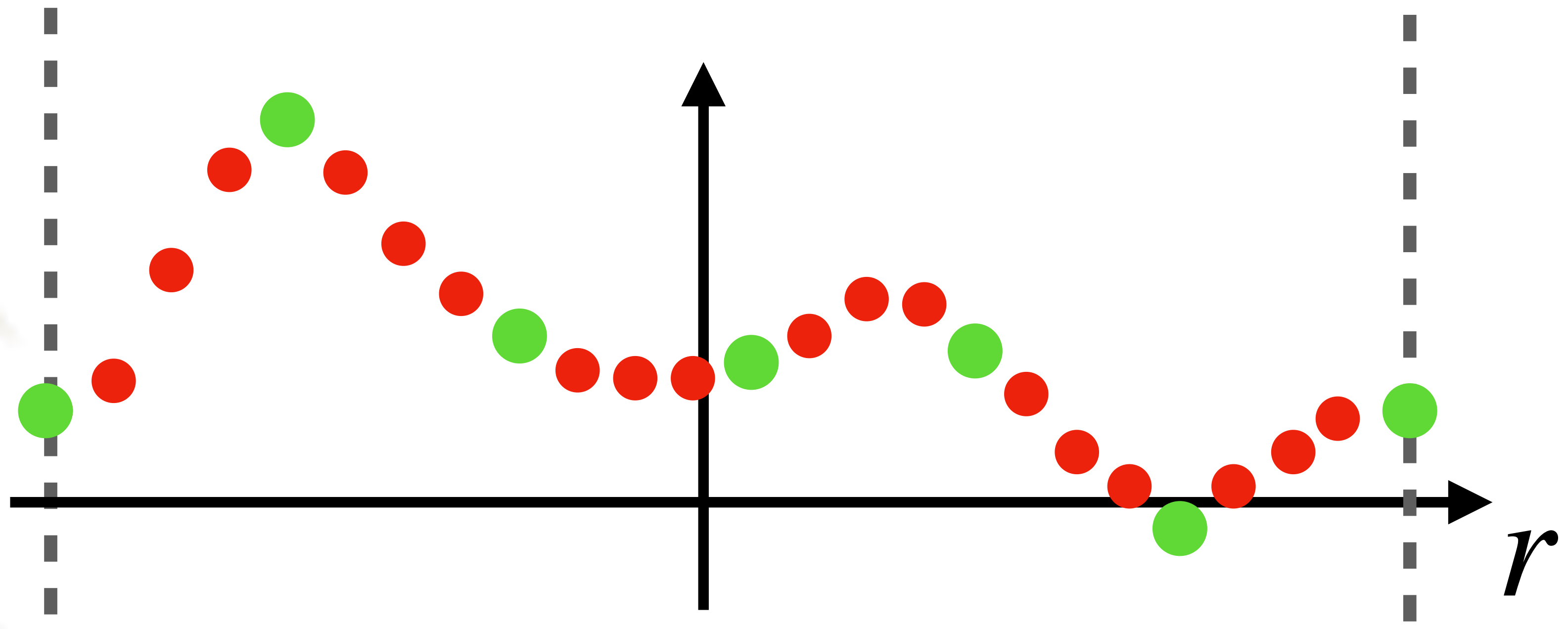
$$W_{ou\mathbf{k},o'u'\mathbf{k}'} = \frac{1}{N_k^2} \int_{\Omega^l \times \Omega^l} d\mathbf{r} d\mathbf{r}' e^{-i(\mathbf{k}-\mathbf{k}')(\mathbf{r}-\mathbf{r}')} u^*_{u\mathbf{k}}(\mathbf{r}) u_{u'\mathbf{k}'}(\mathbf{r}) W(\mathbf{r}, \mathbf{r}') u^*_{o'\mathbf{k}'}(\mathbf{r}') u_{o\mathbf{k}}(\mathbf{r}')$$

$N_o^2 N_{\mathbf{k}}^2$ $\quad$ $N_u^2 N_{\mathbf{k}}^2$

ISDF

$$Z_{i\mathbf{k},j\mathbf{k}'}(\mathbf{r}) := u^*_{i\mathbf{k}}(\mathbf{r})u_{j\mathbf{k}'}(\mathbf{r}) \approx \sum_{\mu=1}^{N_\mu} \zeta_\mu(\mathbf{r})u^*_{i\mathbf{k}}(\mathbf{r}_\mu)u_{j\mathbf{k}'}(\mathbf{r}_\mu)$$





$\{\hat{\mathbf{r}}_\mu\}_{i=1}^{N_\mu} \subset \{\mathbf{r}_i\}_{i=1}^{N_g}$

$N_\mu \ll N_g$

➡Least squares approximation:

$$\Theta = ZC^*(CC^*)^{-1}$$

Compressed Interaction Kernels

$$V_{ou\mathbf{k},o'u'\mathbf{k}'} \approx \frac{1}{N_k^2} \sum_{\mu,\nu} u^*_{u\mathbf{k}}(\mathbf{r}_\mu) u_{o\mathbf{k}}(\mathbf{r}_\mu) \left\{ \boxed{\int_{\Omega^l \times \Omega^l} dr dr' \zeta_\mu^{*V}(\mathbf{r}) V(\mathbf{r},\mathbf{r}') \zeta_\nu^V(\mathbf{r}')} \right\} u^*_{u'\mathbf{k}}(\mathbf{r}_\nu) u_{o'\mathbf{k}}(\mathbf{r}_\nu)$$

$$\equiv \tilde{V}_{\mu\nu} \Rightarrow \mathcal{O}((N_\mu^V)^2 N_r^2)$$

$$W_{ou\mathbf{k},o'u'\mathbf{k}'} \approx \frac{1}{N_k^2} \sum_{\mu,\nu} u^*_{u\mathbf{k}}(\mathbf{r}_\mu) u_{u'\mathbf{k}'}(\mathbf{r}_\mu) \left\{ \boxed{\int_{\Omega^l \times \Omega^l} dr dr' \zeta_\mu^{*W_u}(\mathbf{r}) W_{\mathbf{k}-\mathbf{k}'}(\mathbf{r},\mathbf{r}') \zeta_\nu^{W_o}(\mathbf{r}')} \right\} u^*_{o'\mathbf{k}'}(\mathbf{r}_\nu) u_{o\mathbf{k}}(\mathbf{r}_\nu)$$

$$\equiv \tilde{W}_{\mathbf{k}-\mathbf{k}',\mu\nu} \Rightarrow \mathcal{O}(N_k N_\mu^{W_o} N_\mu^{W_u} N_r^2)$$

Lanczos for

diagonalization $\Rightarrow H \cdot X \Rightarrow \mathcal{O}(N_k \log N_k + N_e^2)$

Results


Optical Absor
N_k
Im ε
ω [eV]
wall time [hrs]
wall time [mins]
N_k
N_k
N_k

input.xml

Choose the **solver**: **direct** or **fastBSE**

$$u_{i\mathbf{k}}(\mathbf{r})\bar{u}_{j\mathbf{k}'}(\mathbf{r}) \approx \sum_{\mu=1}^{N_\mu} \zeta_\mu(\mathbf{r})\, u_{i\mathbf{k}}(\hat{\mathbf{r}}_\mu)\,\bar{u}_{j\mathbf{k}'}(\hat{\mathbf{r}}_\mu)$$

$$V_{ou\mathbf{k},o'u'\mathbf{k}'} \approx \frac{1}{N_k^2} \sum_{\mu,\nu}^{N_\mu^V} u^*_{u\mathbf{k}}(\hat{\mathbf{r}}_\mu)\, u_{o\mathbf{k}}(\hat{\mathbf{r}}_\mu)\, \tilde{V}_{\mu\nu}\, u^*_{u'\mathbf{k}}(\hat{\mathbf{r}}_\nu)\, u_{o'\mathbf{k}}(\hat{\mathbf{r}}_\nu)$$

$$W_{ou\mathbf{k},o'u'\mathbf{k}'} \approx \frac{1}{N_k^2} \sum_{\mu}^{N_\mu^W u} \sum_{\nu}^{N_\mu^W o} u^*_{u\mathbf{k}}(\hat{\mathbf{r}}_\mu)\, u_{u'\mathbf{k}'}(\hat{\mathbf{r}}_\mu)\, W_{\mathbf{k}-\mathbf{k}',\mu\nu}\, u^*_{o'\mathbf{k}'}(\hat{\mathbf{r}}_\nu)\, u_{o\mathbf{k}}(\hat{\mathbf{r}}_\nu)$$

Maximum Lanczos iterations

```
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    nstlbse="1 4 1 20"
    solver="fastBSE"/>

<fastBSE
    rsampling="20 20 20"
    nisdf="80 200 200"
    lanczosmaxits="200"/>
```

Thank you!

C. Vorwerk, B. Aurich, C. Cocchi, C. Draxl, Electronic Structure, (2019)

F. Henneke , L. Lin, C. Vorwerk, C. Draxl, R. Klein, C. Yang Commun. Appl. Math. Comp. Sci. ***15****, 89 (2020)*