TDDFT in linear response

$$v_{ext}(\mathbf{r}, t) = v_{ext}(\mathbf{r}, 0) + \delta v_{ext}(\mathbf{r}, t)$$

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"small"

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$$v_{ext}(\mathbf{r},t) = v_{ext}(\mathbf{r},0) + \boxed{\delta v_{ext}(\mathbf{r},t)}$$

"small"

$$\chi(\mathbf{r},\mathbf{r}',\omega)$$

polarizability :: linear response function

TDDFT in linear response

$$v_{ext}(\mathbf{r},t) = v_{ext}(\mathbf{r},0) + \delta v_{ext}(\mathbf{r},t)$$

$$\chi(\mathbf{r},\mathbf{r}',\omega)$$

polarizability :: linear response function excitations energies

Absorption spectrum

Electron Energy Loss

refraction index

Inelastic X-ray Scattering

Compton Scattering

Reflectivity

Surface differential reflectivity

Reflectance Anisotropy spectroscopy

....

$$v_{ext}(\mathbf{r}, t) = v_{ext}(\mathbf{r}, 0) + \delta v_{ext}(\mathbf{r}, t)$$

$$n(\mathbf{r}, t) = n(\mathbf{r}, 0) + \delta n(\mathbf{r}, t) + \delta^{(2)} n(\mathbf{r}, t) + \ldots$$

$$v_{ext}(\mathbf{r},t) = v_{ext}(\mathbf{r},0) + \delta v_{ext}(\mathbf{r},t)$$

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$$v_{ext}(\mathbf{r},t) = v_{ext}(\mathbf{r},0) + \boxed{\delta v_{ext}(\mathbf{r},t)}$$

$$n(\mathbf{r},t) = n(\mathbf{r},0) + \boxed{\delta n(\mathbf{r},t)} + \delta^{(2)} n(\mathbf{r},t) + \ldots$$

$$\delta n(\mathbf{r},t) = \int d\mathbf{r}' dt' \, \chi(\mathbf{r},\mathbf{r}',t-t') \delta v_{ext}(\mathbf{r}',t')$$

polarizability

polarizability :: density-density response function

$$\chi(\mathbf{r},\mathbf{r}',t-t') = i\,\langle\Psi_0|[\hat{n}(\mathbf{r},t),\hat{n}(\mathbf{r}',t')]|\Psi_0\rangle$$

polarizability :: density-density response function

$$\chi(\mathbf{r},\mathbf{r}',t-t') = i\,\langle\Psi_0|[\hat{n}(\mathbf{r},t),\hat{n}(\mathbf{r}',t')]|\Psi_0\rangle$$

$$\hat{n}(\mathbf{r},t) = e^{-iHt}\hat{n}(\mathbf{r})e^{iHt} \qquad \hat{n}(\mathbf{r}) = \sum_i \delta(\mathbf{r}-\mathbf{r}_i)$$

polarizability :: density-density response function

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$$\chi(\mathbf{r},\mathbf{r}',\omega) = \sum_I \left[\frac{\langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r}')|\Psi_0\rangle}{\omega-(E_I-E_0)+i0^+} - \frac{\langle\Psi_0|\hat{n}(\mathbf{r}')|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r})|\Psi_0\rangle}{\omega+(E_I-E_0)+i0^+}\right]$$

Exercise

polarizability :: density-density response function

$$\chi(\mathbf{r},\mathbf{r}',t-t') = i\,\langle\Psi_0|[\hat{n}(\mathbf{r},t),\hat{n}(\mathbf{r}',t')]|\Psi_0\rangle$$

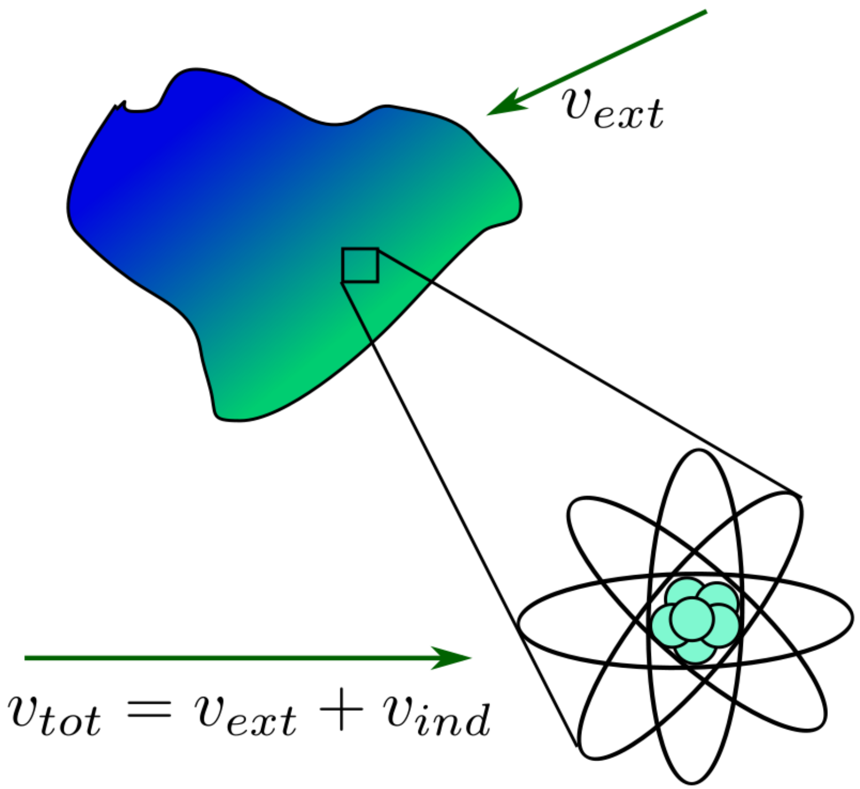
$$\hat{n}(\mathbf{r},t) = e^{-iHt}\hat{n}(\mathbf{r})e^{iHt} \qquad \hat{n}(\mathbf{r}) = \sum_i \delta(\mathbf{r}-\mathbf{r}_i)$$

$$\chi(\mathbf{r},\mathbf{r}',\omega) = \sum_I \left[\frac{\langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r}')|\Psi_0\rangle}{\omega-\underbrace{(E_I-E_0)}_{\Omega_I}+i0^+} - \frac{\langle\Psi_0|\hat{n}(\mathbf{r}')|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r})|\Psi_0\rangle}{\omega+(E_I-E_0)+i0^+}\right]$$

Ω_I excitations energies

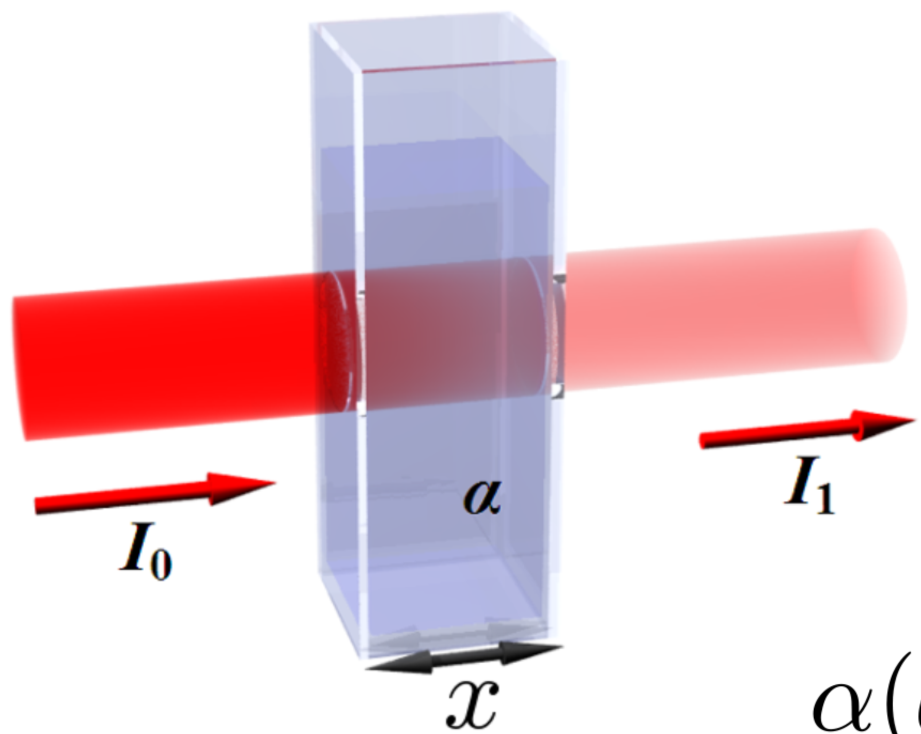
Exercise

Connection to spectroscopies :: inverse dielectric function



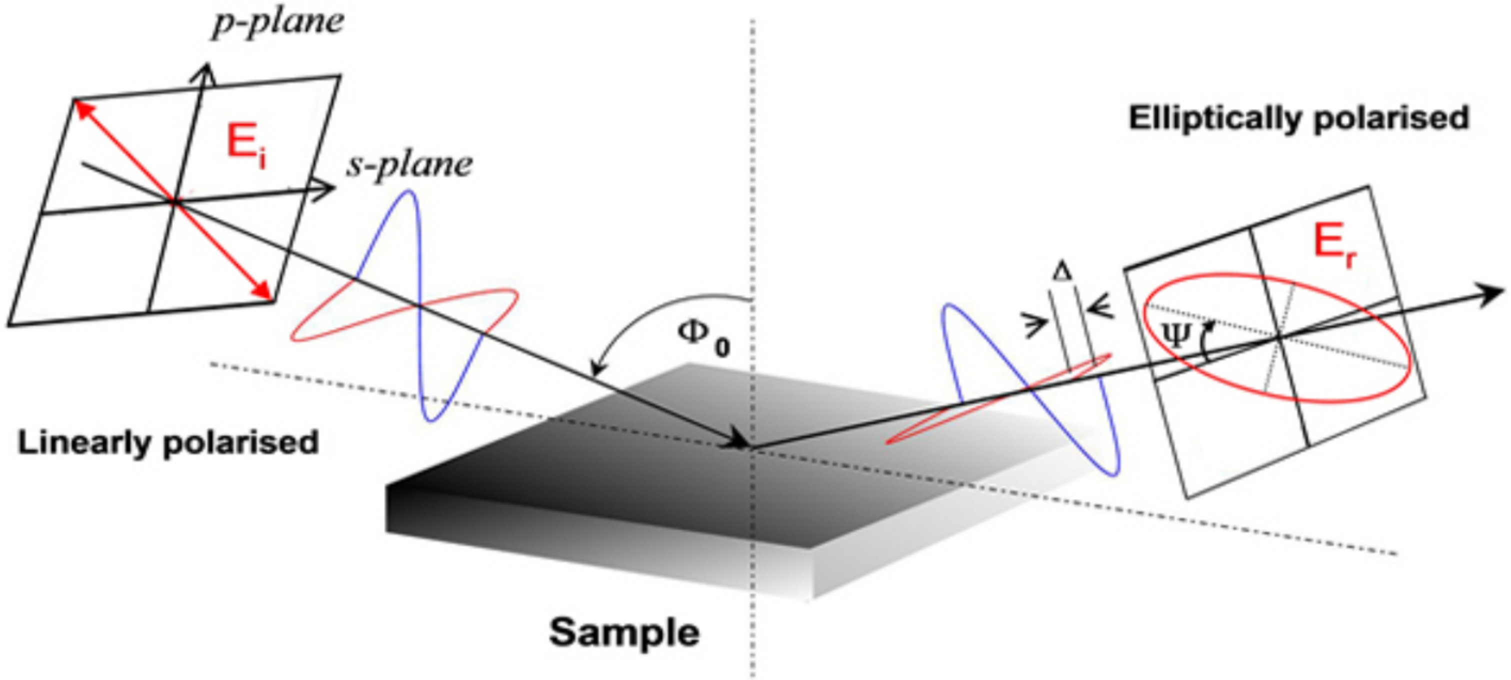
$$v_{tot} = \varepsilon^{-1} v_{ext}$$

Connection to spectroscopies :: optical absorption



$$\alpha(\omega) = \mathrm{Im}\left[\varepsilon_M(\omega)\right]$$

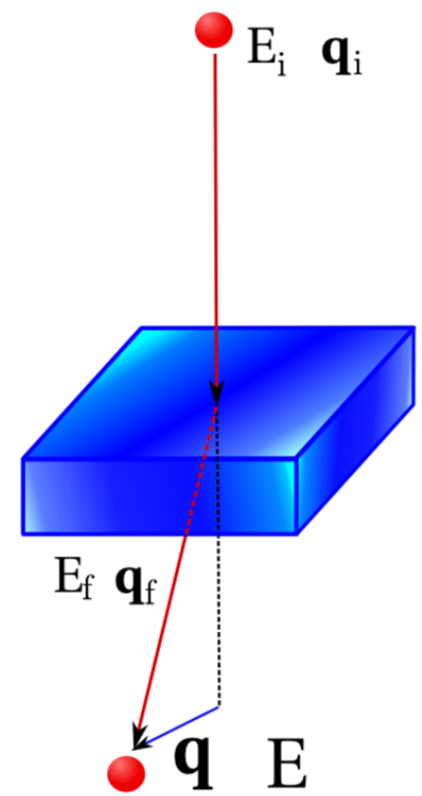
Connection to spectroscopies :: optical absorption



$$\varepsilon_M = \sin^2\Phi + \sin^2\Phi\tan^2\Phi\left(\frac{1-\frac{E_r}{E_i}}{1+\frac{E_r}{E_i}}\right)$$

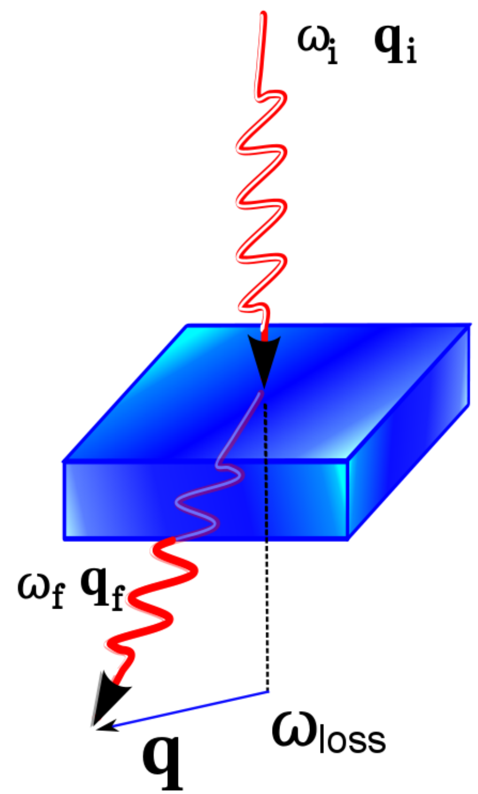
Connection to spectroscopies :: electron energy loss (EELS)

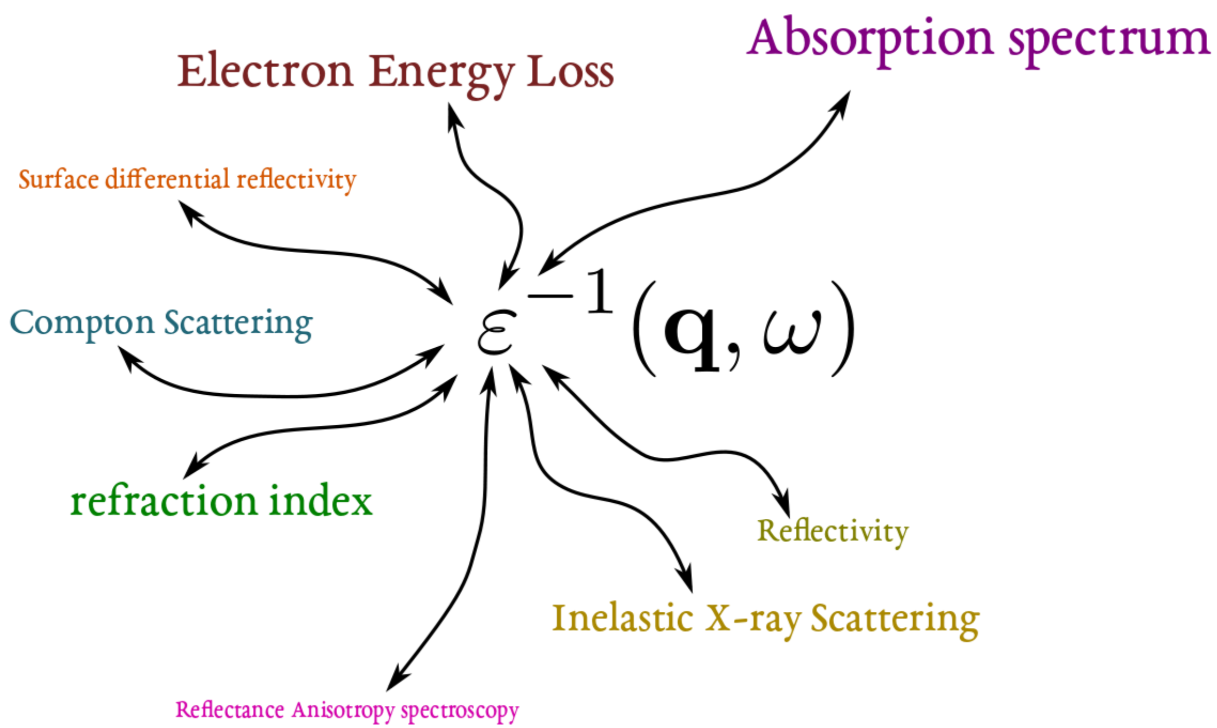
$$\frac{d^2\sigma}{d\Omega d\omega} \propto \mathrm{Im}\left[\varepsilon^{-1}(\mathbf{q},\omega)\right]$$



Connection to spectroscopies :: inelastic X-ray scattering (IXS)

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \mathrm{Im}\left[\varepsilon^{-1}(\mathbf{q},\omega)\right]$$




Absorption spectrum
Electron Energy Loss
Surface differential reflectivity
Compton Scattering
$\varepsilon^{-1}(\mathbf{q}, \omega)$
refraction index
Reflectivity
Inelastic X-ray Scattering
Reflectance Anisotropy spectroscopy

Absorption spectrum

Electron Energy Loss

Surface differential reflectivity

Compton Scattering

$$\varepsilon^{-1}(\mathbf{q},\omega) = 1 + v(\mathbf{q})\chi(\mathbf{q},\omega)$$

refraction index

Reflectivity

Inelastic X-ray Scattering

Reflectance Anisotropy spectroscopy

excitations energies

$$\chi(\mathbf{r},\mathbf{r}',\omega) = \sum_I \left[\frac{\langle \Psi_0 | \hat{n}(\mathbf{r}) | \Psi_I \rangle \; \langle \Psi_I | \hat{n}(\mathbf{r}') | \Psi_0 \rangle}{\omega - (E_I - E_0) + i0^+} - \frac{\langle \Psi_0 | \hat{n}(\mathbf{r}') | \Psi_I \rangle \; \langle \Psi_I | \hat{n}(\mathbf{r}) | \Psi_0 \rangle}{\omega + (E_I - E_0) + i0^+} \right]$$

Polarizability of an independent-particle system

$$\chi(\mathbf{r},\mathbf{r}',\omega) = \sum_I \left[\frac{\langle\Psi_0|\hat{n}(\mathbf{r})|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r}')|\Psi_0\rangle}{\omega - (E_I - E_0) + i0^+} - \frac{\langle\Psi_0|\hat{n}(\mathbf{r}')|\Psi_I\rangle\ \langle\Psi_I|\hat{n}(\mathbf{r})|\Psi_0\rangle}{\omega + (E_I - E_0) + i0^+} \right]$$

Ψ_0 single determinant

Polarizability of an independent-particle system

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Ψ_0 single determinant

Exercise

$$\chi^0(\mathbf{r},\mathbf{r}',\omega) = \sum_{ij} (f_i - f_j) \left[\frac{\psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')}{\omega - \underbrace{(\epsilon_j - \epsilon_i)}_{} + i0^+} - \frac{\psi_i(\mathbf{r})\psi_j^*(\mathbf{r})\psi_i^*(\mathbf{r}')\psi_j(\mathbf{r}')}{\omega + (\epsilon_j - \epsilon_i) + i0^+} \right]$$

$$\delta n = \chi^0 \delta v_{eff} \qquad\qquad \delta n$$

$$\delta n = \chi^0 \delta v_{eff} \qquad\qquad \delta n = \chi \delta v_{ext}$$

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$$\chi \delta v_{ext} \overset{\mathrm{DFT}}{=} \chi^0 \delta v_{eff}$$

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$$\chi \delta v_{ext} \overset{\text{DFT}}{=} \chi^0 \delta v_{eff}$$

$$\delta v_{eff} = \delta v_{ext} + \delta v_H + \delta v_{xc}$$

Dyson equation for the polarizability

$$\chi = \chi^0 + \chi^0 \left[v + f_{xc}\right] \chi$$

- evalu

Dyson equation for the polarizability

$$\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$$

$$\chi(\mathbf{r}, \mathbf{r}', \omega) = \chi^0(\mathbf{r}, \mathbf{r}', \omega) +$$

$$+ \int d\mathbf{r}_1 d\mathbf{r}_2 \chi^0(\mathbf{r}, \mathbf{r}_1, \omega) \left[v(\mathbf{r}_1, \mathbf{r}_2) + f_{xc}(\mathbf{r}_1, \mathbf{r}_2, \omega) \right] \chi(\mathbf{r}_2, \mathbf{r}', \omega)$$

- evalu
- f_{xc}
- appro

Dyson equation for the polarizability

$$\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$$

$$\chi(\mathbf{r}, \mathbf{r}', \omega) = \chi^0(\mathbf{r}, \mathbf{r}', \omega) +$$

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$$f_{xc} = \frac{\delta v_{xc}}{\delta n}$$ exchange-correlation kernel

- evalu
- f_{xc}
- appro

- evaluation of χ knowing χ^0 (ground state calculation)
- f_{xc} functional of the ground-state density
- approximations for f_{xc}

$$\left.\begin{array}{l} \bullet\ f_{xc} = 0 \\[2ex] \bullet\ f_{xc} = \dfrac{\delta v_{xc}^{gs}}{\delta n} \\[2ex] \bullet\ \text{any other } f_{xc} \end{array}\right\} \textit{coherence vs freedom}$$

Practical procedure for χ and ε^{-1} Scal

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- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V_{ion}^{ps}) $o(N^{1 \div 3})$

Practical procedure for χ and ε^{-1} Scalin

- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V_{ion}^{ps}) $o(N^{1\div 3}$
- creation of $\chi^0 = \sum_{ij} \frac{\psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')}{\omega - (\epsilon_j - \epsilon_i) + i0^+}$ $o(N^4)$

Practical procedure for χ and ε^{-1} Scalin

- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V_{ion}^{ps}) $o(N^{1\div3}$
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- determination of $\chi = \chi^0 + \chi^0 \left[v + f_{xc}\right] \chi$ (approx :: f_{xc}) $o(N^{2\div3})$

Practical procedure for χ and ε^{-1} Scaling

- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V^{ps}_{ion}) — $o(N^{1\div3})$
- creation of $\chi^0 = \sum_{ij} \frac{\psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')}{\omega - (\epsilon_j - \epsilon_i) + i0^+}$ — $o(N^4)$
- determination of $\chi = \chi^0 + \chi^0 \left[v + f_{xc}\right] \chi$ (approx :: f_{xc}) — $o(N^{2\div3})$
- evaluation of $\varepsilon^{-1} = 1 + v\chi$

  Absorption spectrum Inelastic X-ray Scattering refraction index Surface differential reflectivity

Practical procedure for χ and ε^{-1} — Scaling

- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V_{ion}^{ps}) — $o(N^{1\div3})$
- creation of $\chi^0 = \sum_{ij} \frac{\psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')}{\omega - (\epsilon_j - \epsilon_i) + i0^+}$ — $o(N^4)$
- determination of $\chi = \chi^0 + \chi^0 [v + f_{xc}] \chi$ (approx :: f_{xc}) — $o(N^{2\div3})$
- evaluation of $\varepsilon^{-1} = 1 + v\chi$

Absorption spectrum Inelastic X-ray Scattering refraction index Surface differential reflectivity
Compton Scattering Reflectivity Electron Energy Loss Reflectance Anisotropy spectroscopy

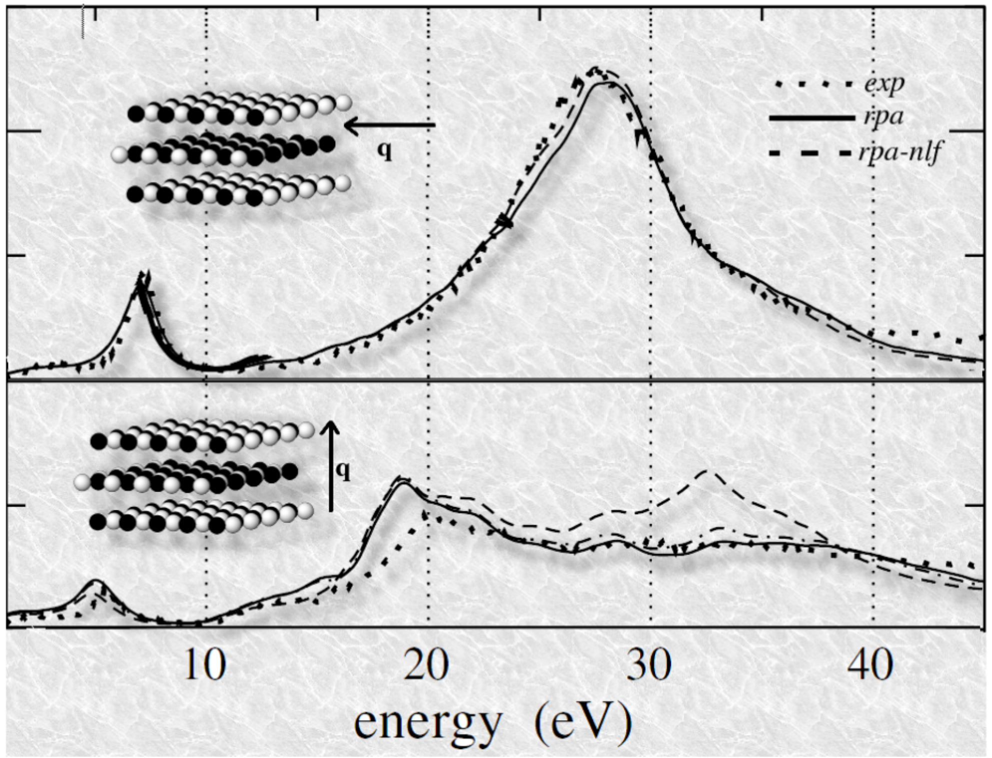
Practical procedure for χ and ε^{-1}

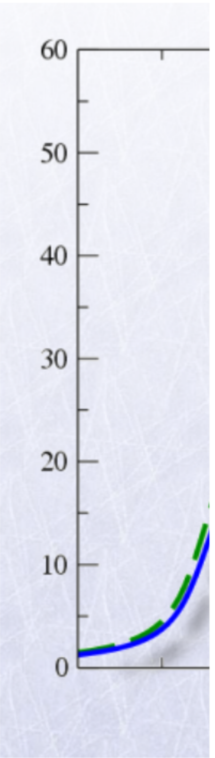
Scaling (with N_{atoms})

- DFT-KS calculation ψ_i, ϵ_i (approx :: v_{xc}, V_{ion}^{ps}) — $o(N^{1\div 3})$
- creation of $\chi^0 = \sum_{ij} \frac{\psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')}{\omega - (\epsilon_j - \epsilon_i) + i0^+}$ — $o(N^4)$
- determination of $\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$ (approx :: f_{xc}) — $o(N^{2\div 3})$
- evaluation of $\varepsilon^{-1} = 1 + v\chi$

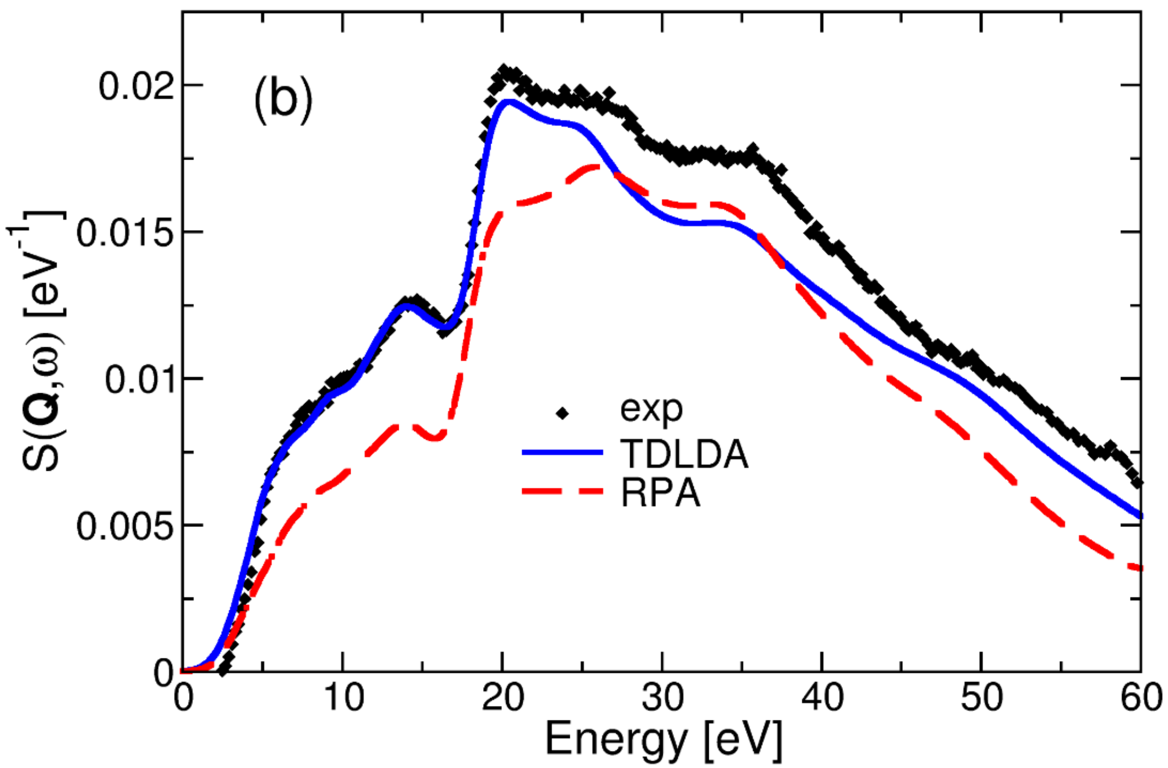
  Absorption spectrum Inelastic X-ray Scattering refraction index Surface differential reflectivity
  Compton Scattering Reflectivity Electron Energy Loss Reflectance Anisotropy spectroscopy

EELS of graphite

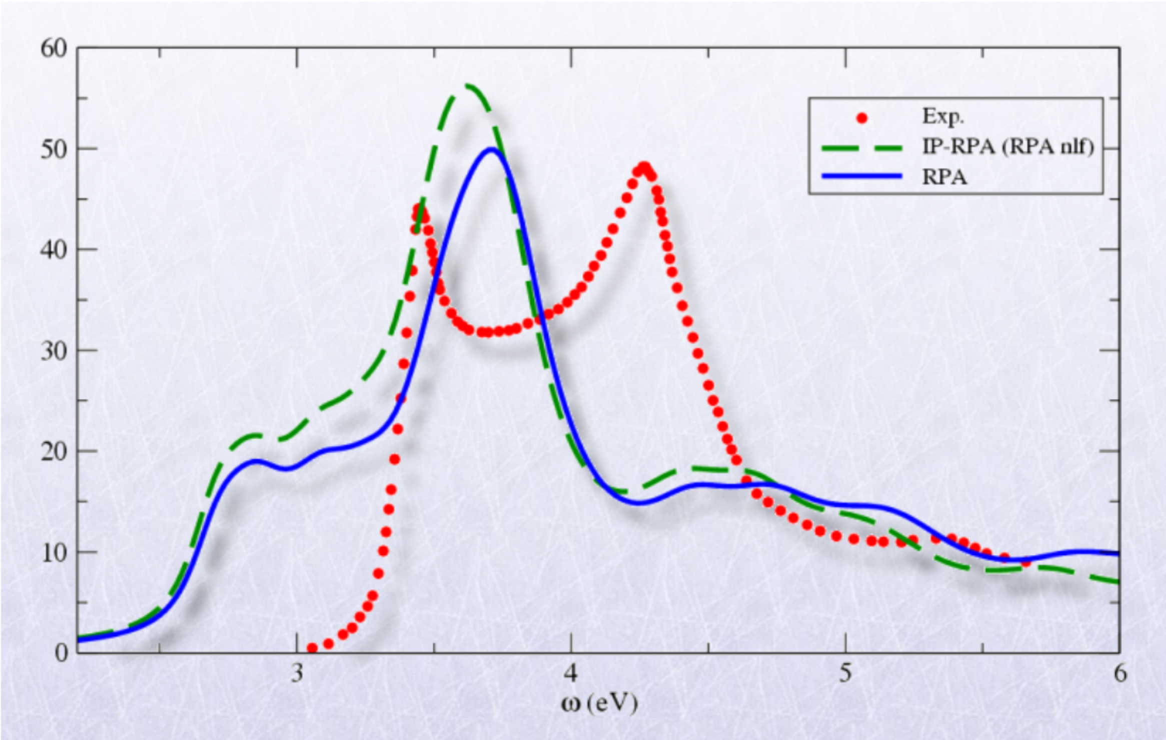




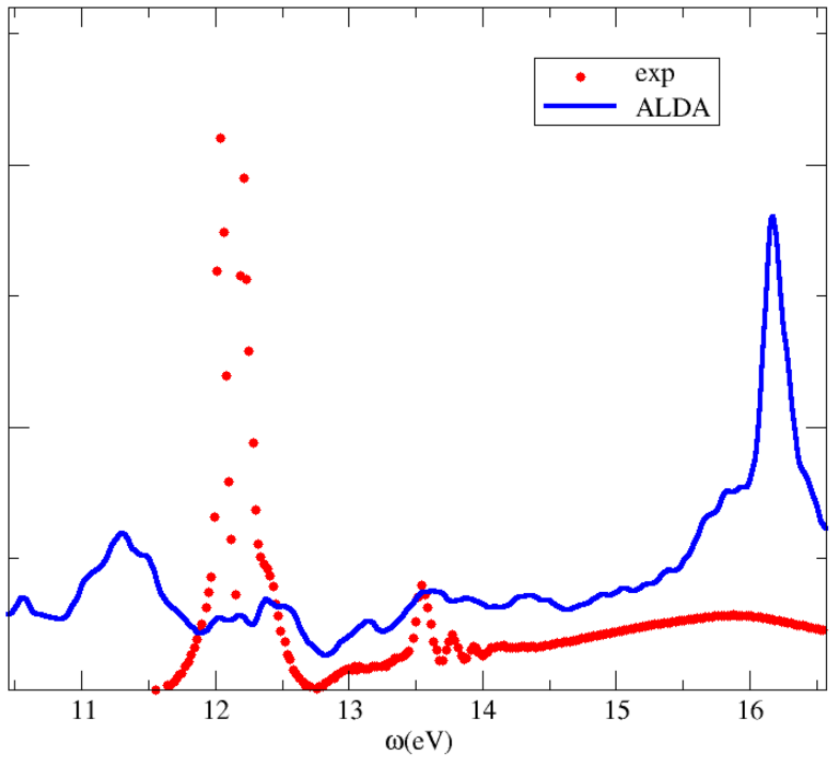
IXS of Silicon

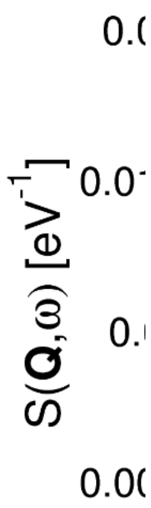


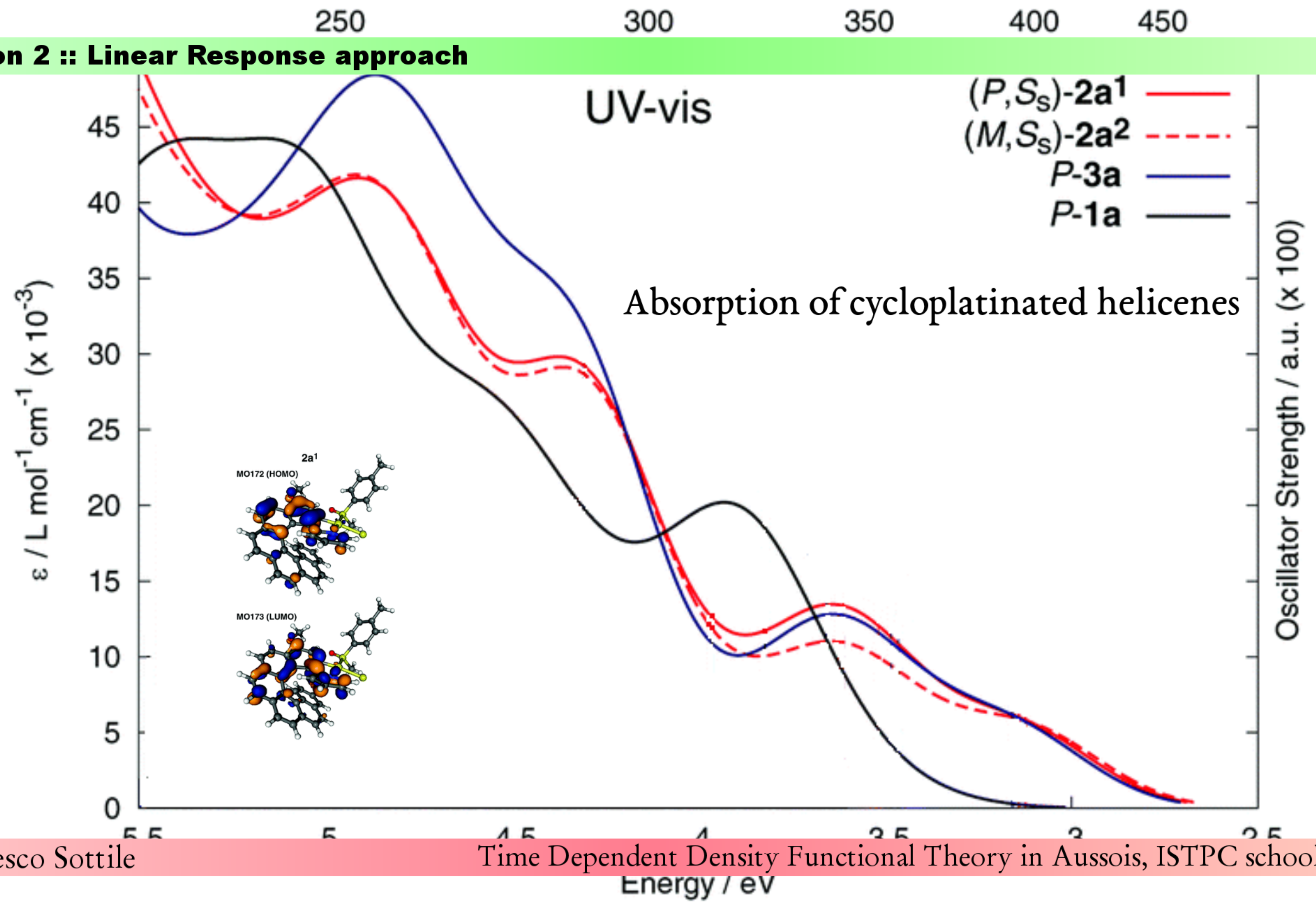
Absorption of Silicon

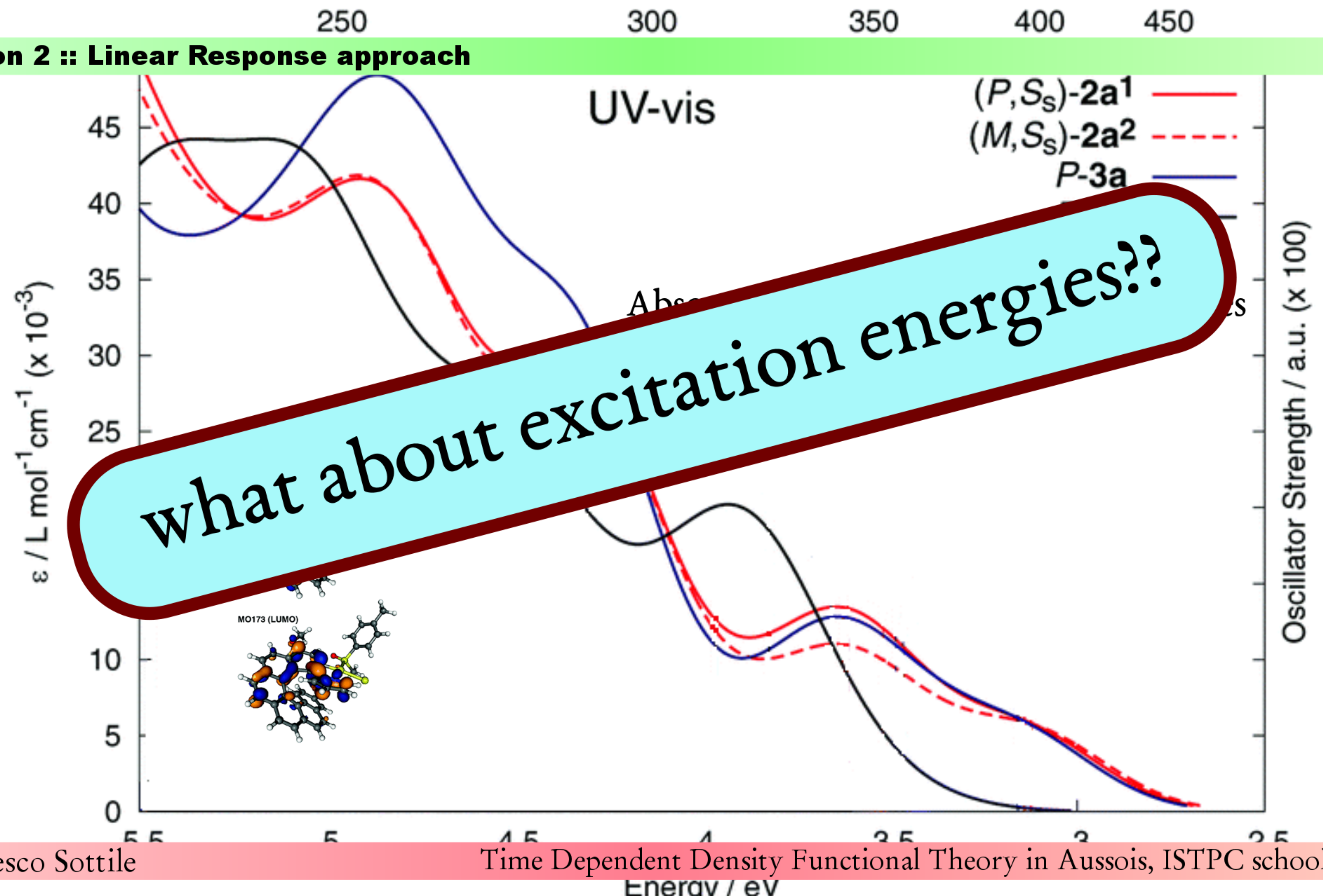


Absorption of Argon

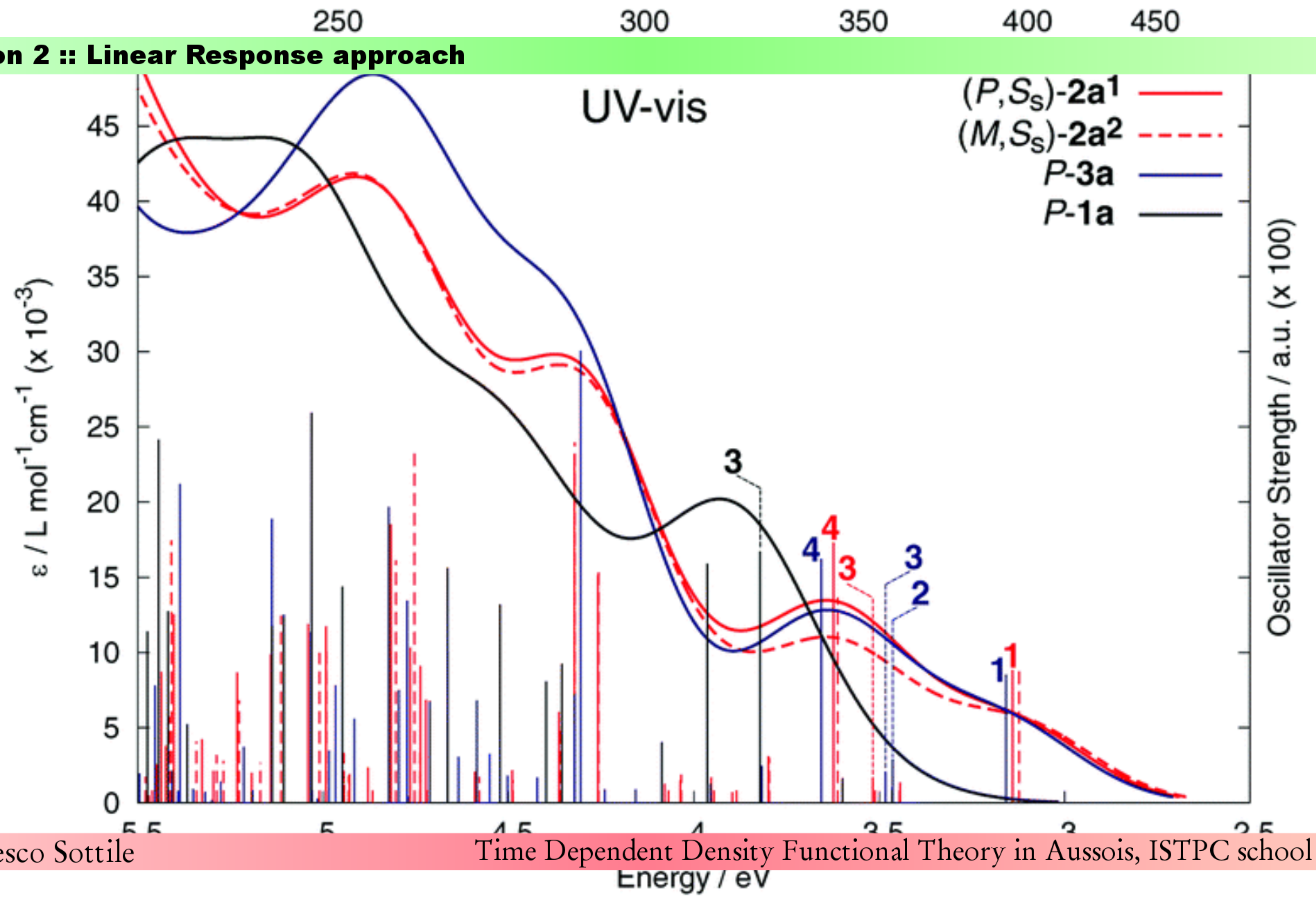





250
300
350
400
450
UV-vis
(P,S_S)-2a1
(M,S_S)-2a2
P-3a
P-1a
Absorption of cycloplatinated helicenes
ε / L mol-1cm-1 (x 10-3)
Oscillator Strength / a.u. (x 100)
45
40
35
30
25
20
15
10
5
0
2a1
MO172 (HOMO)
MO173 (LUMO)
5.5
5
4.5
4
3.5
3
2.5
Energy / eV



what about excitation energies??


UV-vis
250
300
350
400
450
(P,S_S)-2a1
(M,S_S)-2a2
P-3a
P-1a
ε / L mol-1cm-1 (x 10-3)
Oscillator Strength / a.u. (x 100)
Energy / eV
0
5
10
15
20
25
30
35
40
45
3
4
4
3
3
2
1
1

$$\chi(\mathbf{r},\mathbf{r}',\omega) = \chi^0(\mathbf{r},\mathbf{r}',\omega) +$$

$$+ \int d\mathbf{r}_1 d\mathbf{r}_2 \chi^0(\mathbf{r},\mathbf{r}_1,\omega)\left[v(\mathbf{r}_1,\mathbf{r}_2) + f_{xc}(\mathbf{r}_1,\mathbf{r}_2,\omega)\right]\chi(\mathbf{r}_2,\mathbf{r}',\omega)$$

$$\chi(\mathbf{r}, \mathbf{r}', \omega) = \chi^0(\mathbf{r}, \mathbf{r}', \omega) +$$

$$+ \int d\mathbf{r}_1 d\mathbf{r}_2 \chi^0(\mathbf{r}, \mathbf{r}_1, \omega) \left[v(\mathbf{r}_1, \mathbf{r}_2) + f_{xc}(\mathbf{r}_1, \mathbf{r}_2, \omega) \right] \chi(\mathbf{r}_2, \mathbf{r}', \omega)$$

basis change

$$f_{ij}^{kl} = \iint \psi_i^*(\mathbf{r}) \psi_j(\mathbf{r}) \psi_k(\mathbf{r}') \psi_l^*(\mathbf{r}') \, f(\mathbf{r}, \mathbf{r}') \; d\mathbf{r} d\mathbf{r}'$$

$$\chi(\mathbf{r},\mathbf{r}',\omega) = \chi^0(\mathbf{r},\mathbf{r}',\omega) +$$

$$+ \int d\mathbf{r}_1 d\mathbf{r}_2 \chi^0(\mathbf{r},\mathbf{r}_1,\omega)\left[v(\mathbf{r}_1,\mathbf{r}_2) + f_{xc}(\mathbf{r}_1,\mathbf{r}_2,\omega)\right]\chi(\mathbf{r}_2,\mathbf{r}',\omega)$$

basis change

$$f_{ij}^{kl} = \iint \psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_k(\mathbf{r}')\psi_l^*(\mathbf{r}')\, f(\mathbf{r},\mathbf{r}')\; d\mathbf{r}d\mathbf{r}'$$

$$\chi_{ij}^{kl} = \left[\chi^0\right]_{ij}^{kl} + \sum_{mnop}\left[\chi^0\right]_{ik}^{mn}\left[v_{mn}^{op} + \left[f_{xc}\right]_{mn}^{op}\right]\chi_{op}^{kl}$$

$$[\chi^0]_{ij}^{kl} = \frac{(f_i - f_j)\delta_{ik}\delta_{jl}}{\omega - (\epsilon_j - \epsilon_i)}$$

diagonal in ij, kl

$$\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$$

$$\Downarrow$$

Exercise

$$\chi = \left[\left(\chi^0 \right)^{-1} - \left(v + f_{xc} \right) \right]^{-1}$$

$$\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$$

Exercise

$$\chi = \left[\left(\chi^0 \right)^{-1} - \left(v + f_{xc} \right) \right]^{-1}$$

$$\chi = \left[\left(\chi^0 \right)^{-1} - K \right]^{-1}$$

$$\chi =$$

$$\chi_{ij}^{kl}$$

$$\chi = \left[\left(\chi^0\right)^{-1} \right.$$

χ_{ij}^{kl}

$\omega - (\epsilon_j - \epsilon_i)\delta_{ik}\delta_{jl}$

$$\chi = \left[\left(\chi^0\right)^{-1} - K \right]^{-1}$$

χ_{ij}^{kl}

$\omega - (\epsilon_j - \epsilon_i)\delta_{ik}\delta_{jl}$

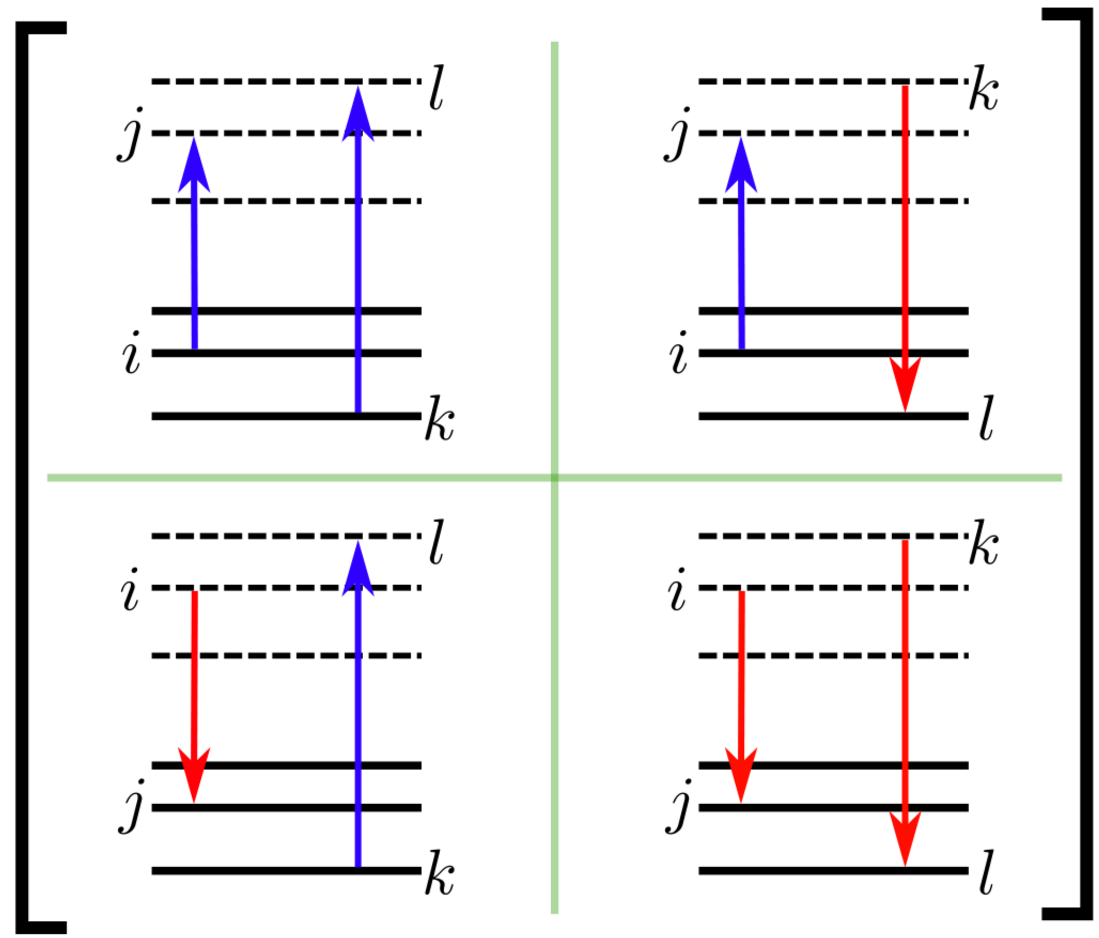
$K_{ij}^{kl} = \iint \psi_i^*(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j^*(\mathbf{r}')K(\mathbf{r},\mathbf{r}')\,d\mathbf{r}d\mathbf{r}'$

$$\chi = \frac{1}{H^{\mathrm{EXC}} - \omega} = \sum_{\lambda\lambda'} \frac{|V_\lambda\rangle\, S_\lambda^{\lambda'}\, \langle V_\lambda|}{E_\lambda - \omega}$$

$$\chi = \frac{1}{H^{\mathrm{EXC}} - \omega} = \sum_{\lambda} \frac{|V_\lambda\rangle \langle V_\lambda|}{E_\lambda - \omega}$$

$$H^{\mathrm{EXC}} = \begin{array}{cc} & ij \\ kl & \begin{bmatrix} (\epsilon_j - \epsilon_i)\delta_{ik}\delta_{jl} & \ddots & K_{ij}^{kl} \\ K_{ij}^{kl} & (\epsilon_j - \epsilon_i)\delta_{ik}\delta_{jl} & K_{ij}^{kl} \\ K_{ij}^{kl} & K_{ij}^{kl} \quad \ddots & (\epsilon_j - \epsilon_i)\delta_{ik}\delta_{jl} \end{bmatrix} \end{array}$$

$$H^{\mathrm{EXC}} = \ {}_{kl}\!\!{}^{ij}$$



$$H^{\mathrm{EXC}} = {}_{kl}{}^{ij}\left[\begin{array}{c|c} \mathbf{A} & \mathbf{B} \\ \hline -\mathbf{B}^{*} & -\mathbf{A}^{*} \end{array}\right]$$

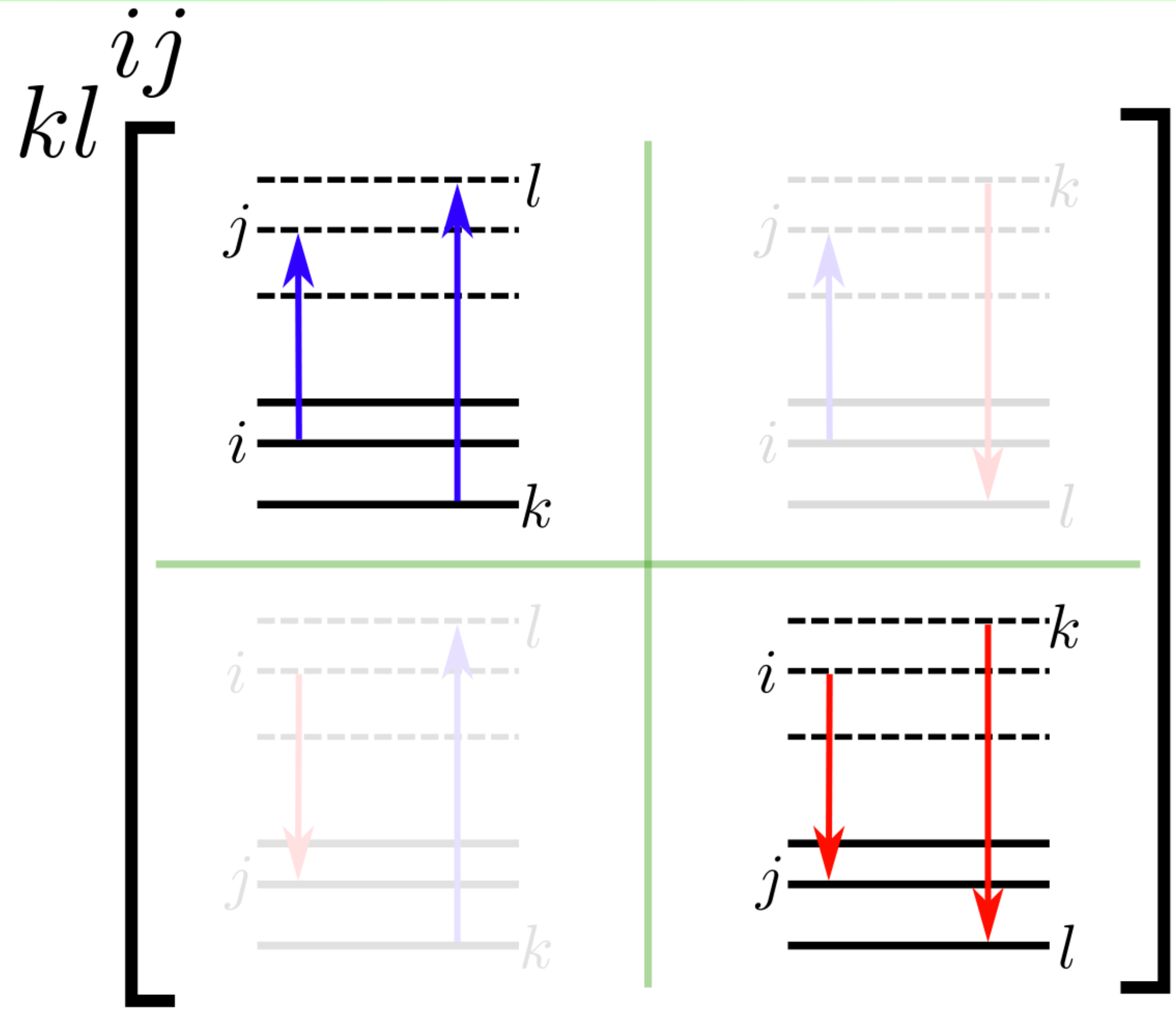
$$\left[\begin{array}{c|c} \mathbf{A} & \mathbf{B} \\ \hline -\mathbf{B}^* & -\mathbf{A}^* \end{array}\right] \begin{bmatrix} \mathbf{X} \\ \mathbf{Y} \end{bmatrix} = E_\lambda \begin{bmatrix} \mathbf{X} \\ \mathbf{Y} \end{bmatrix}$$

$$\left[\begin{array}{c|c} \mathbf{A} & \mathbf{B} \\ \hline \mathbf{B}^* & \mathbf{A}^* \end{array}\right]\left[\begin{array}{c} \mathbf{X} \\ \mathbf{Y} \end{array}\right] = E_\lambda \left[\begin{array}{c|c} 1 & 0 \\ \hline 0 & -1 \end{array}\right]\left[\begin{array}{c} \mathbf{X} \\ \mathbf{Y} \end{array}\right]$$

$$H^{\mathrm{EXC}} = \begin{array}{cc} & ij \\ kl & \left[\begin{array}{c|c} \mathbf{A} & \mathbf{B} \\ \hline -\mathbf{B}^* & -\mathbf{A}^* \end{array}\right] \end{array}$$

Tamm-Dancoff approx

$$H^{\mathrm{EXC}} =$$



Tamm-Dancoff approx

		
$\chi = \chi^0 + \chi^0 \left[v + f_{xc} \right] \chi$	full spectrum N^4 scaling	excitations energies
$\begin{bmatrix} \text{A} & \text{B} \\ \text{-B}^* & \text{-A}^* \end{bmatrix} \begin{bmatrix} \text{X} \\ \text{Y} \end{bmatrix} = E_\lambda \begin{bmatrix} \text{X} \\ \text{Y} \end{bmatrix}$	few excitations energies iterative techniques analysis	N^6 scaling