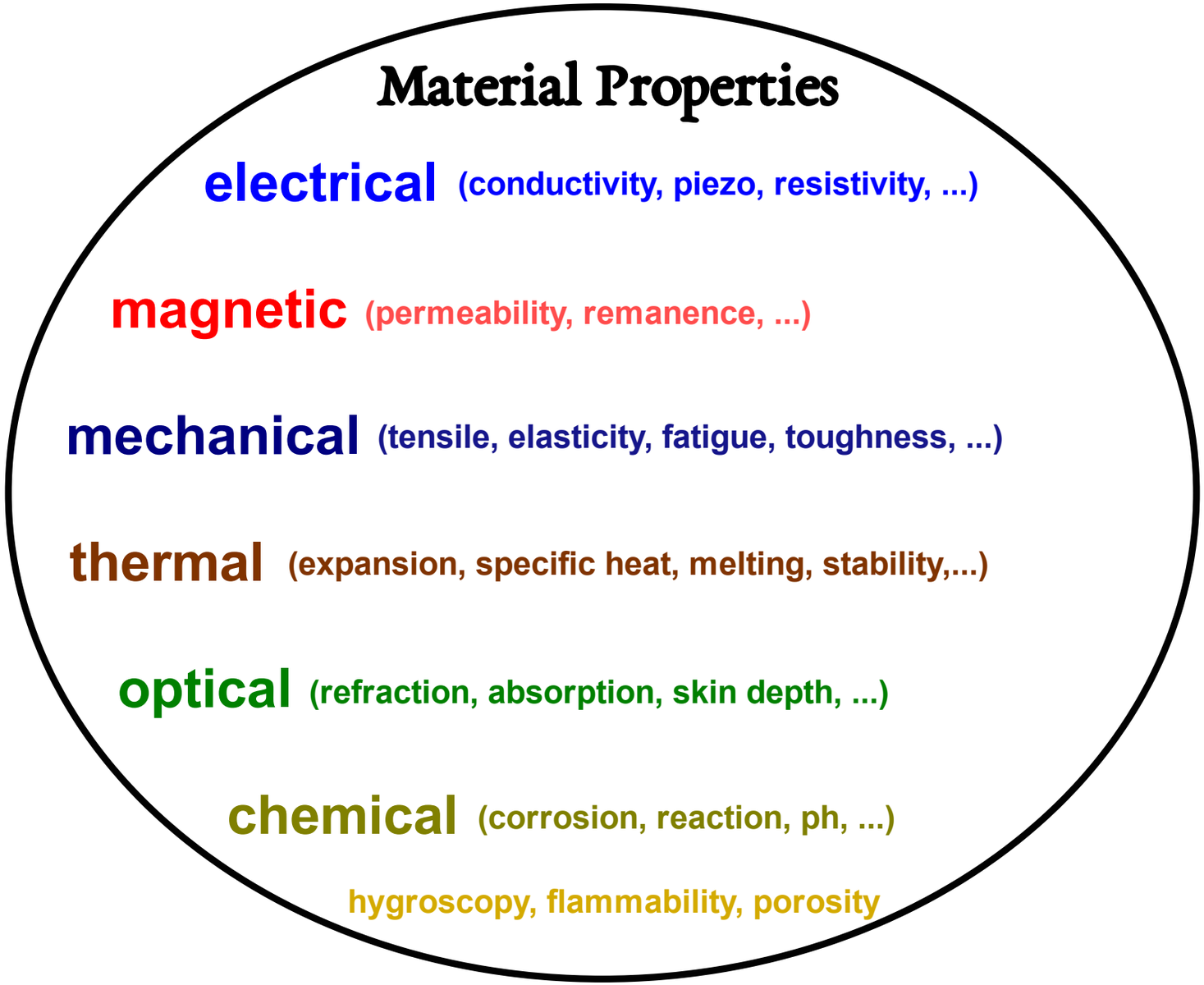
(Ab initio) Theoretical approaches for photovoltaics

Francesco Sottile

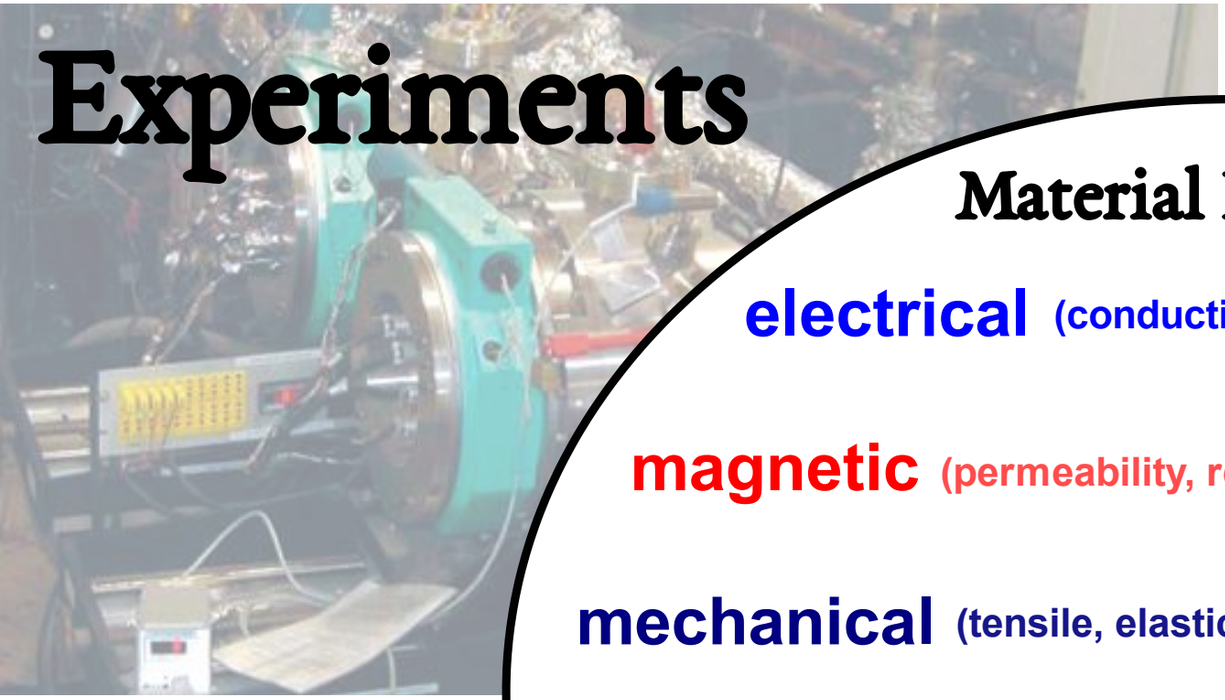
Theoretical Spectroscopy Group, LSI, Ecole Polytechnique

4 July 2024 - Ecole Polytechnique

Space Photovoltaics for Energy Conversion in extra-terrestrial environment Workshop


Material Properties
electrical (conductivity, piezo, resistivity, ...)
magnetic (permeability, remanence, ...)
mechanical (tensile, elasticity, fatigue, toughness, ...)
thermal (expansion, specific heat, melting, stability,...)
optical (refraction, absorption, skin depth, ...)
chemical (corrosion, reaction, ph, ...)
hygroscopy, flammability, porosity

Experiments



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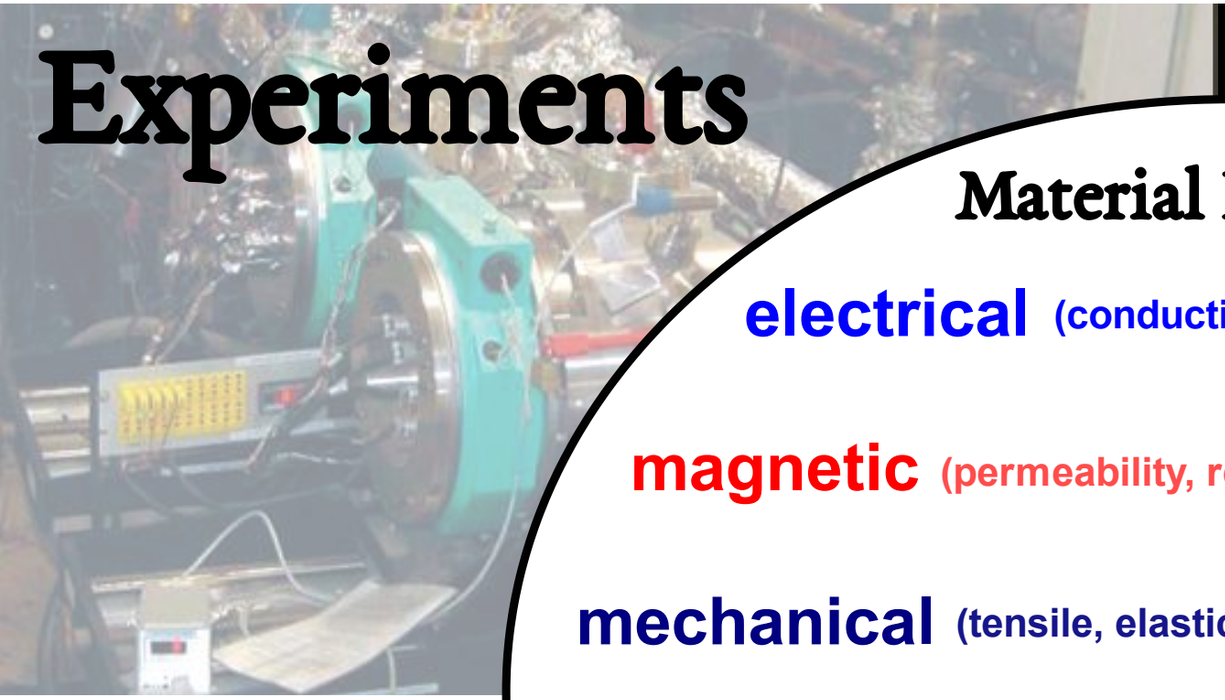

Theory

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2\mathrm{d}\omega_e} \propto \mathrm{Im} \sum_{\substack{vv' \\ cc'c''c''' \\ \mu\mu'\mu''\mu'''}} \left[\tilde{\rho}^*_{\mu v} \cdot \chi^{c'\mu'}_{c\mu}(\omega_i) \cdot \tilde{\rho}_{c'\mu'}\right]^* \chi^{c''v'}_{cv}(\omega_i) \left[\tilde{\rho}^*_{\mu'v'} \cdot \chi^{c'''\mu'''}_{c''\mu''}(\omega_i) \cdot \tilde{\rho}_{c'''\mu'''}\right]$$

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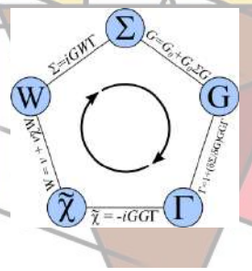

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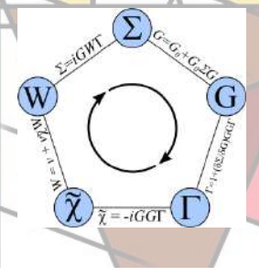

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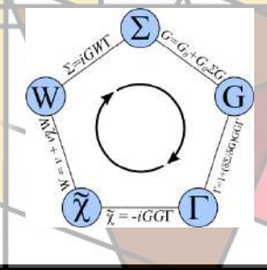

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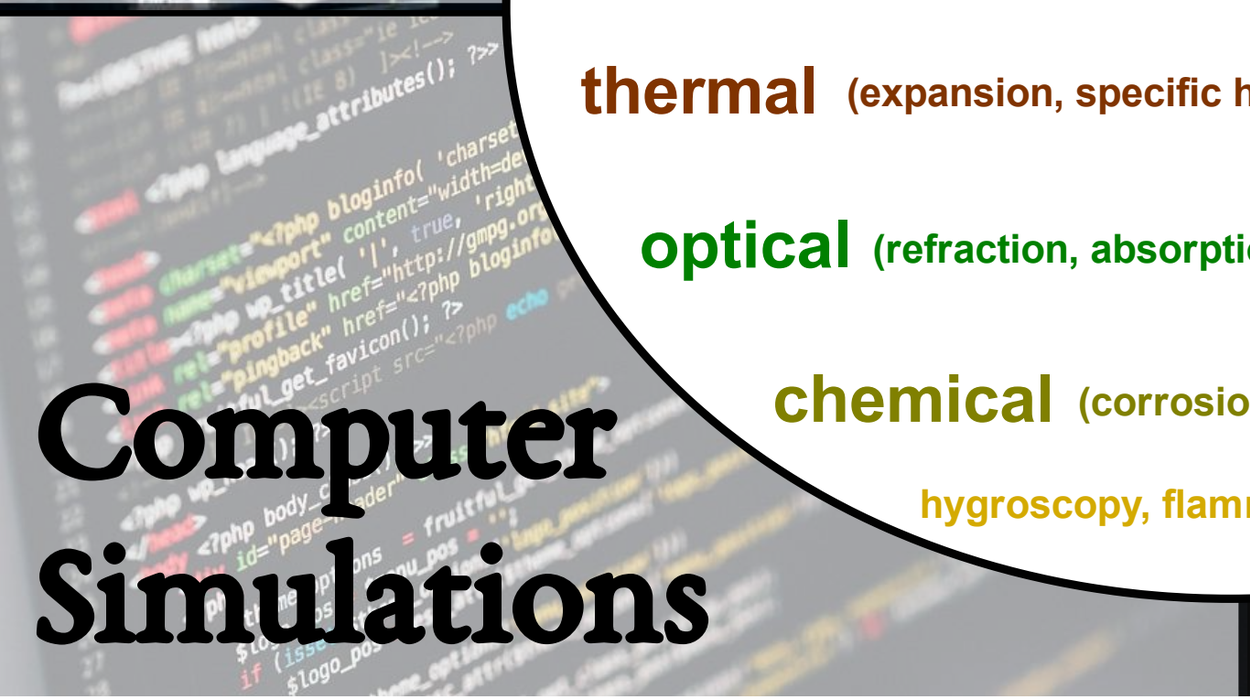

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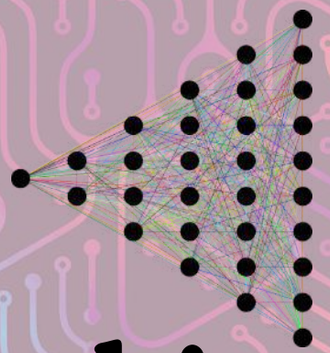


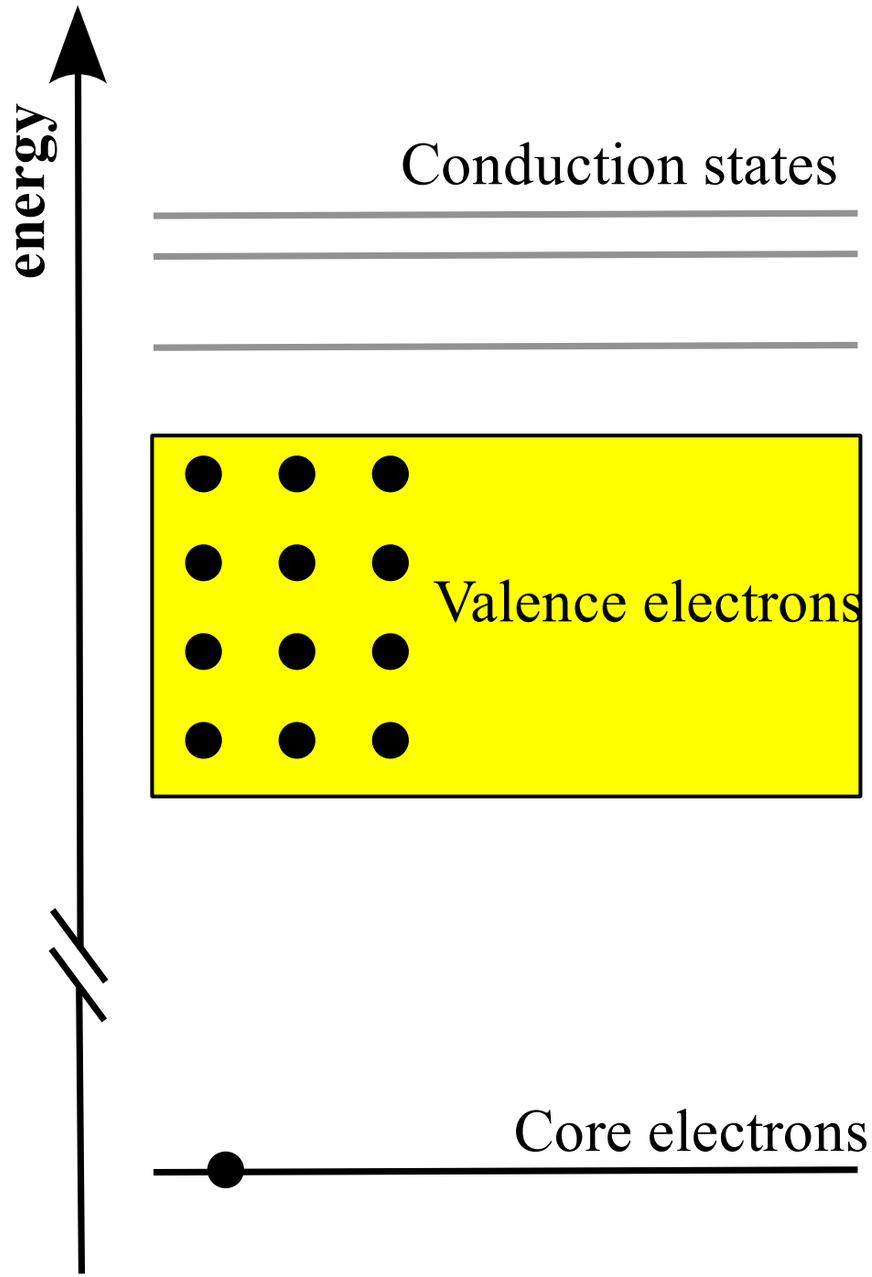
Machine Learning

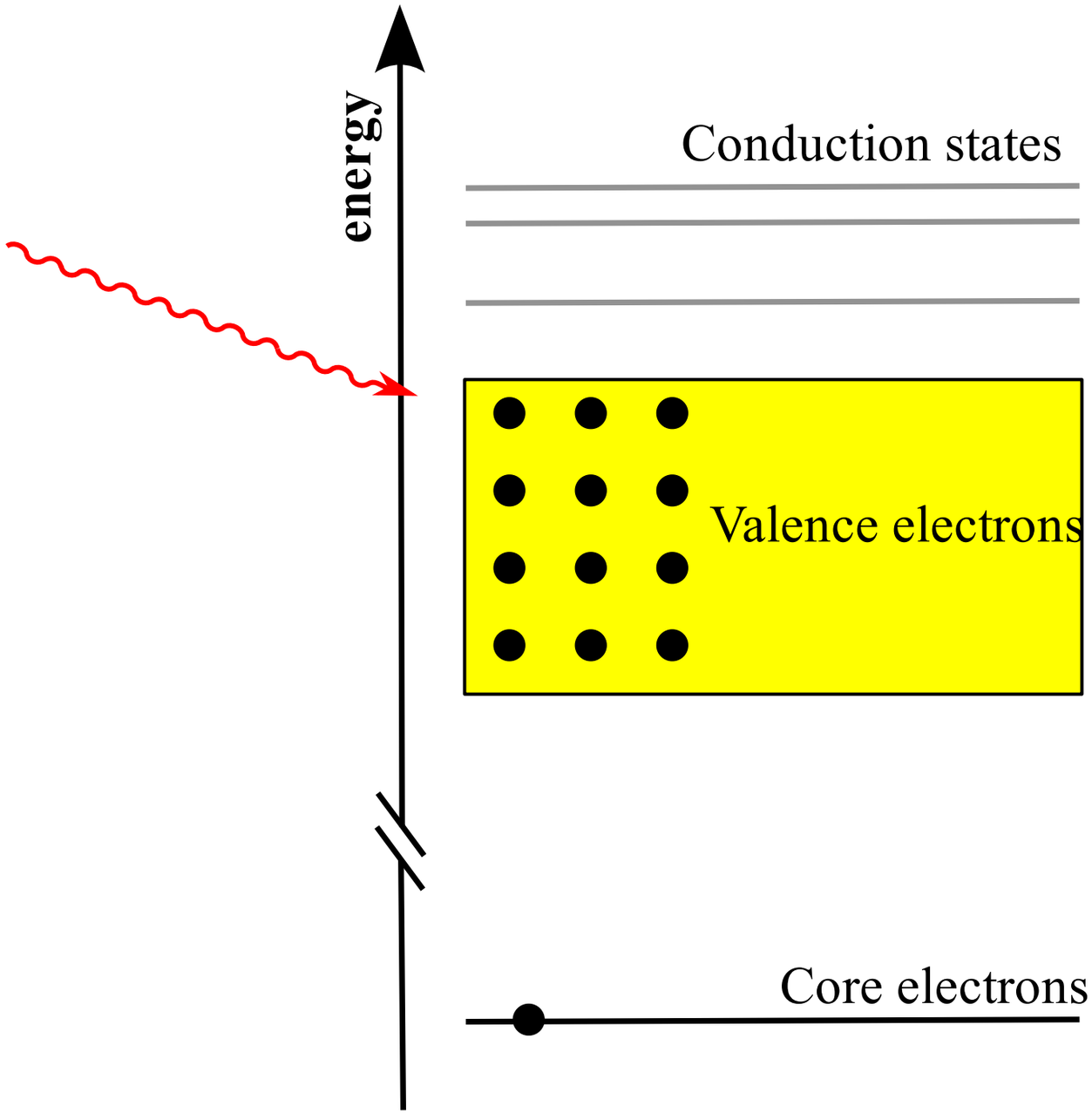
Theoretical Spectroscopy Group

ETSF

theoretical developments for electronic properties

- develop theory and formula
- devise new approximations
- propose new algorithm
- implement in computer codes


energy
Conduction states
Valence electrons
Core electrons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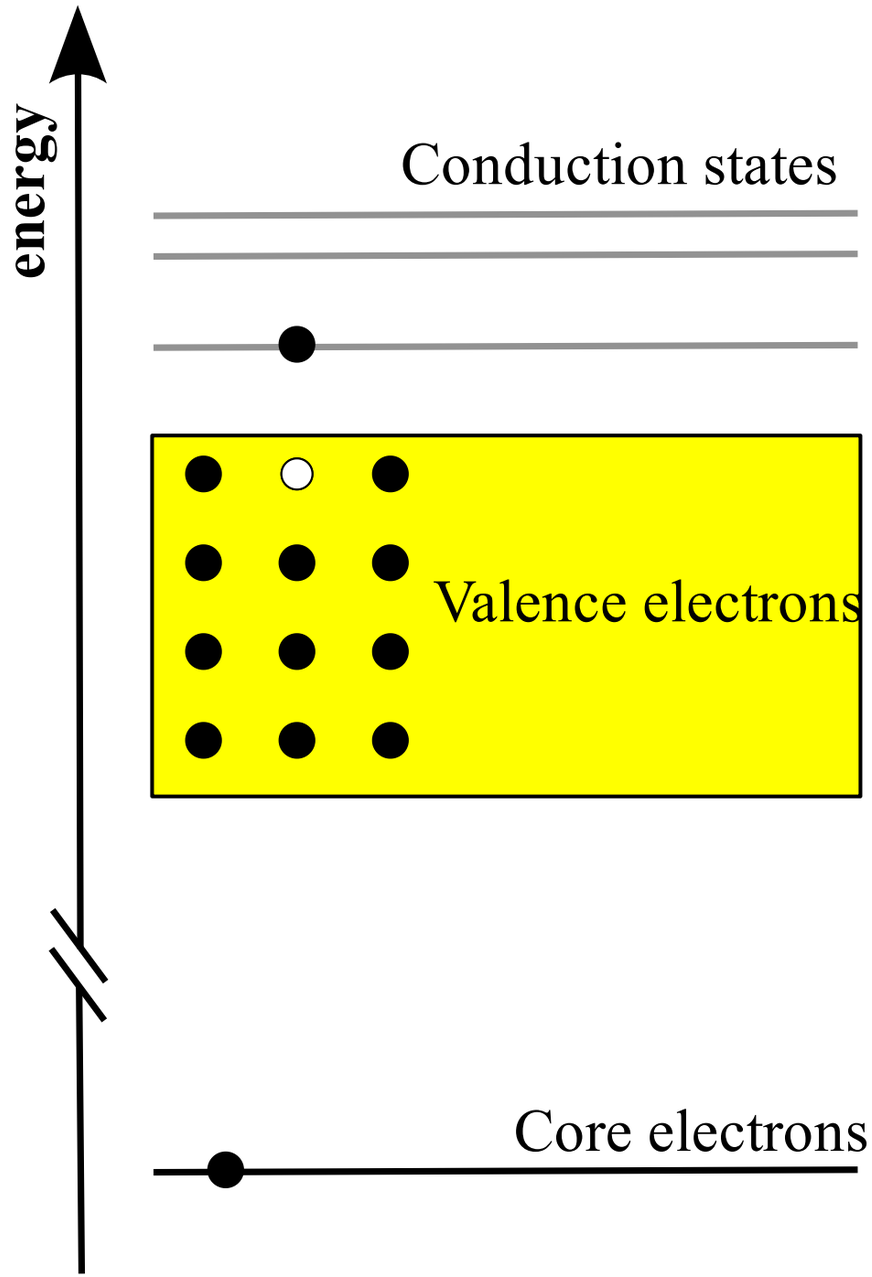
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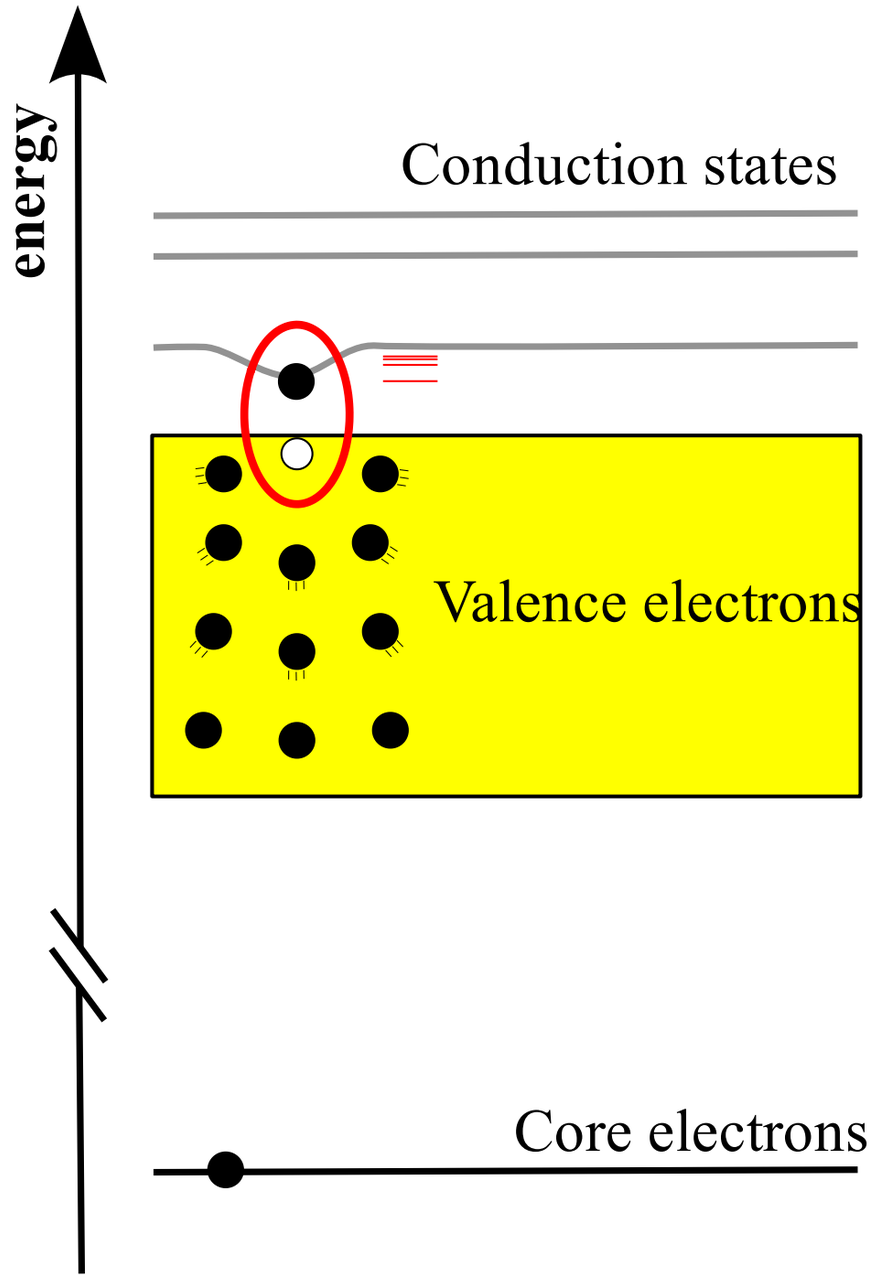
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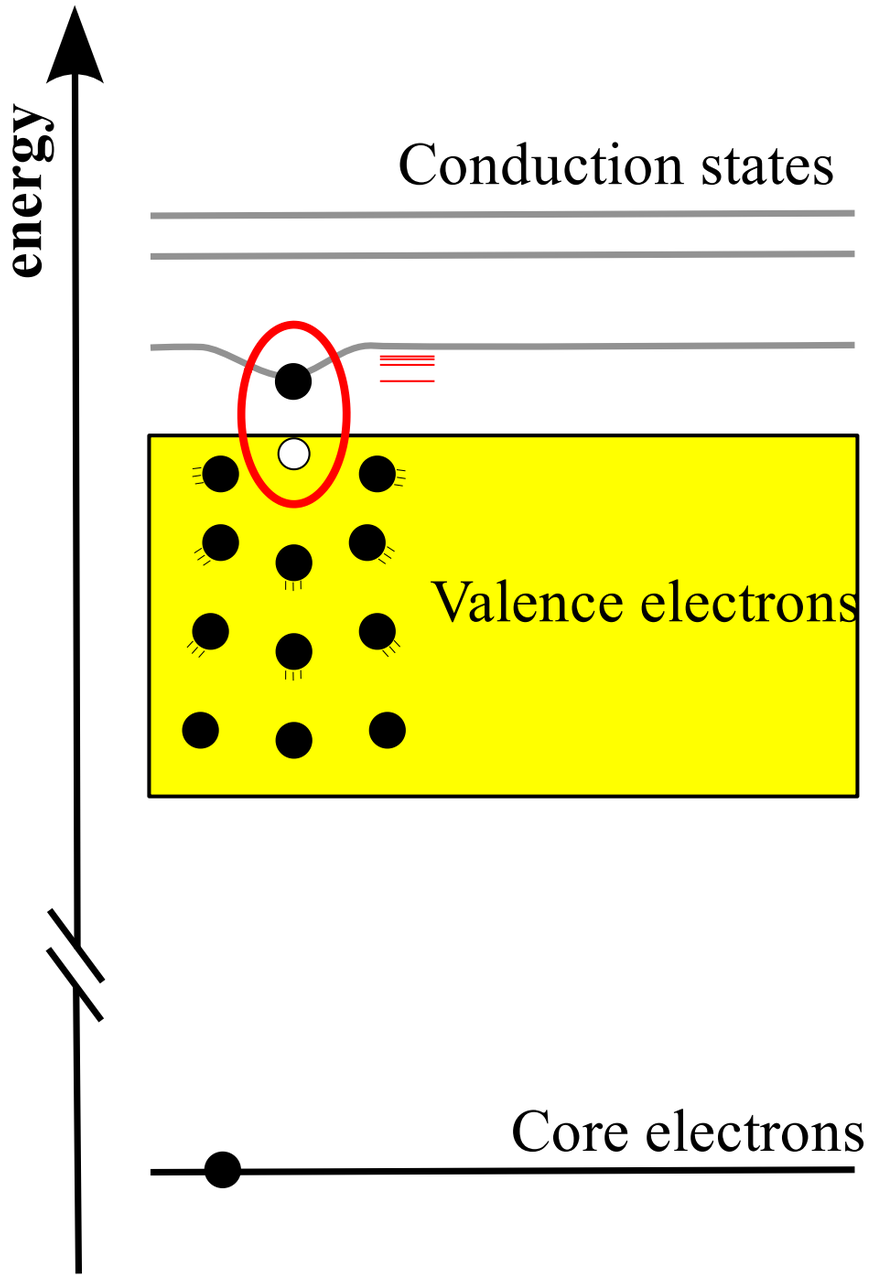


excitons (electron-hole pairs)

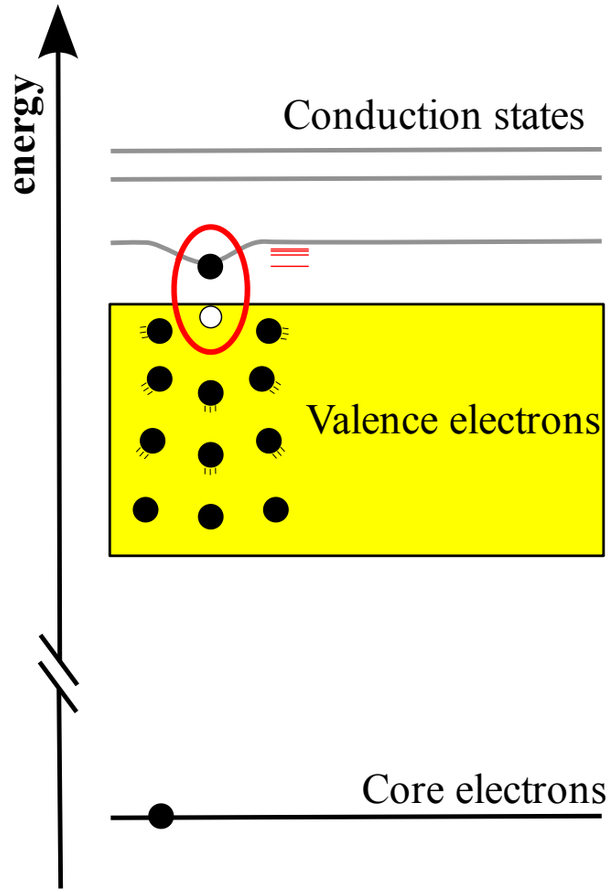
collective modes

beyond one-particle

beyond mean-field

Outline

- (our) Approach to electronic excitations
- Results for absorption spectroscopy
- Advantages, limitations, opportunities



(our) Approach to electronic excitations :: *ab initio*

~~model the system/Hamiltonian~~
~~model the interaction~~
~~model the space topology~~

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$$V_{e-e} = \sum_{i<j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} \qquad V_{e-Z} = \sum_{i,j} \frac{Ze^2}{|\mathbf{r}_i - \mathbf{R}_j|}$$

exponentially hard !!

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Green's functions functional theory $G(\mathbf{r}, \mathbf{r}', \omega)$

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polinomially hard

via functionals

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known interaction (Coulomb)

exponentially hard !!

polinomially hard

via functionals

Density functional theory (DFT) $\rho(\mathbf{r})$

Green's functions functional theory $G(\mathbf{r}, \mathbf{r}', \omega)$

unknown effective potentials $V_{xc}([\rho], \mathbf{r})$

$\Sigma_{xc}(\mathbf{r}, \mathbf{r}', \omega)$

ab initio functional approach (DFT, GFT)

approximated potentials

polinomially scaling

$$A(\omega)$$

$$\varepsilon(\omega)$$

$$\chi^{(n)}(\omega_1, .., \omega_n)$$

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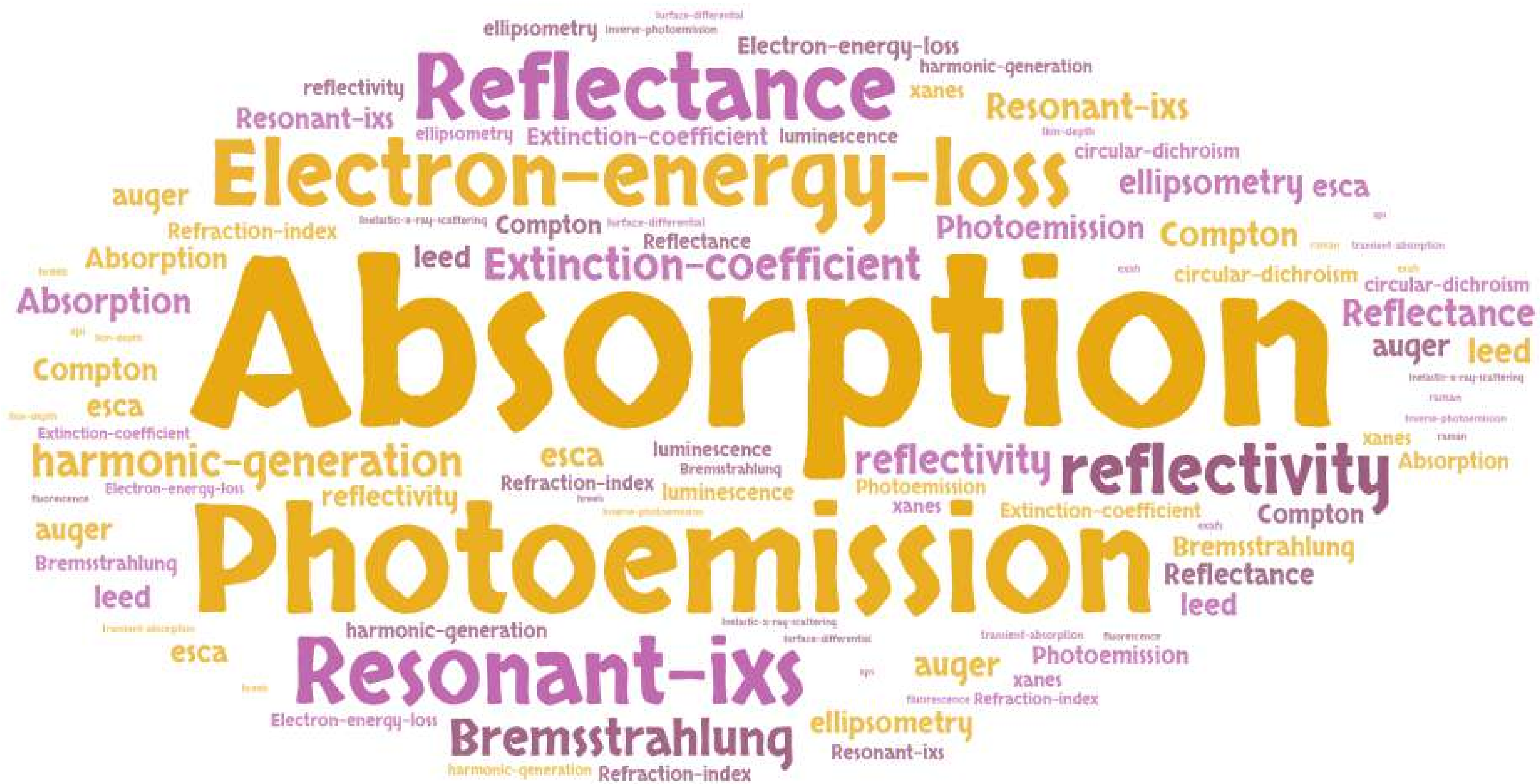

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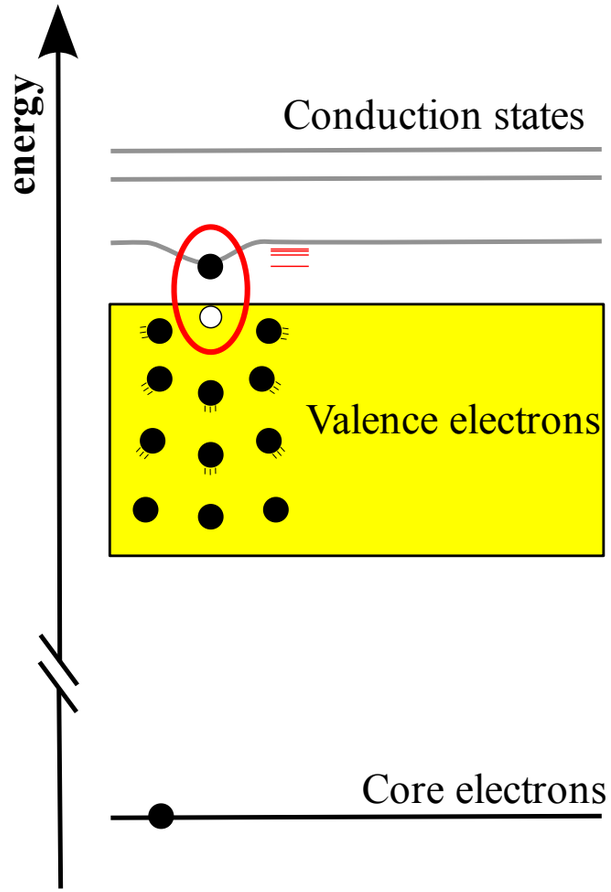
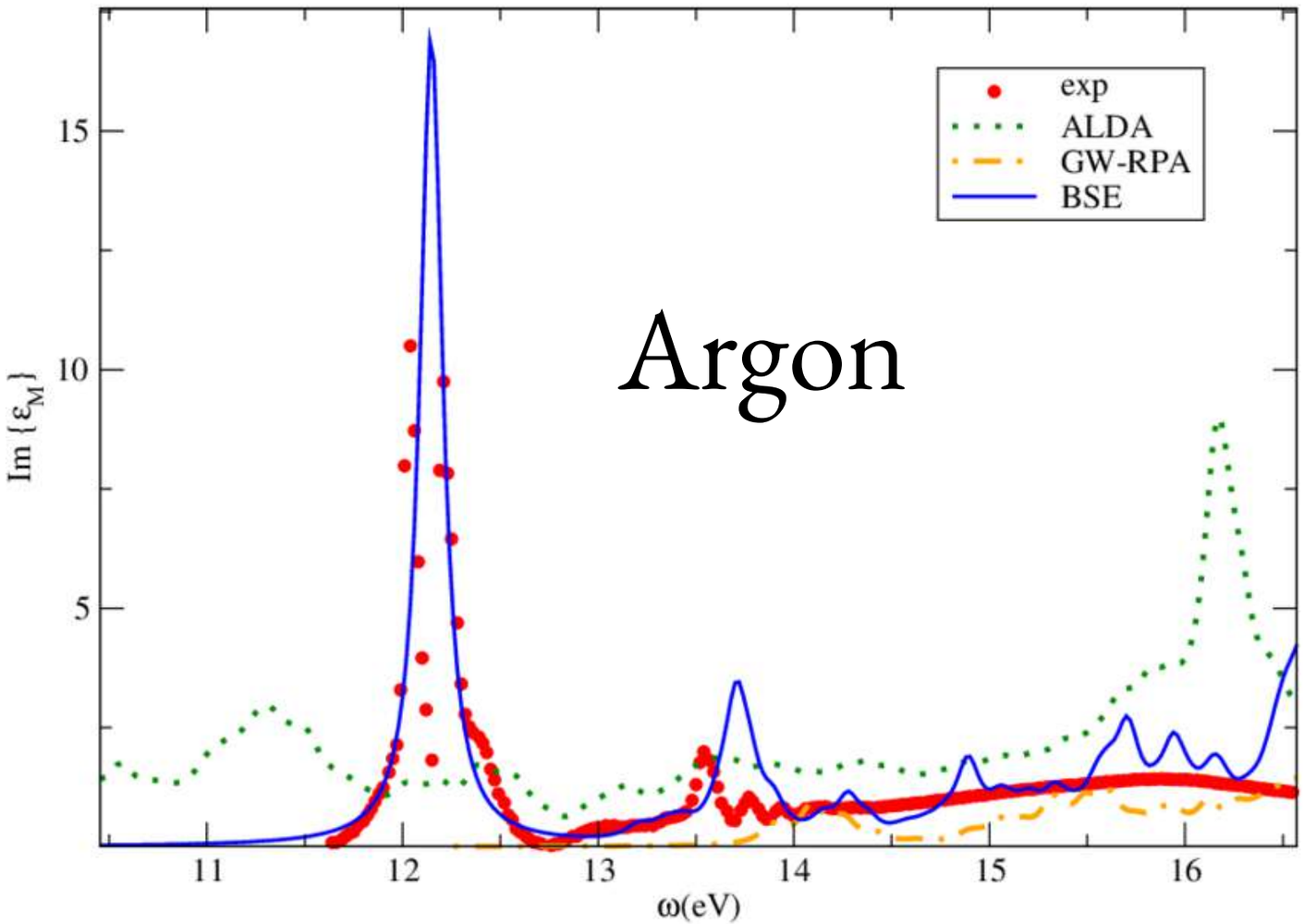
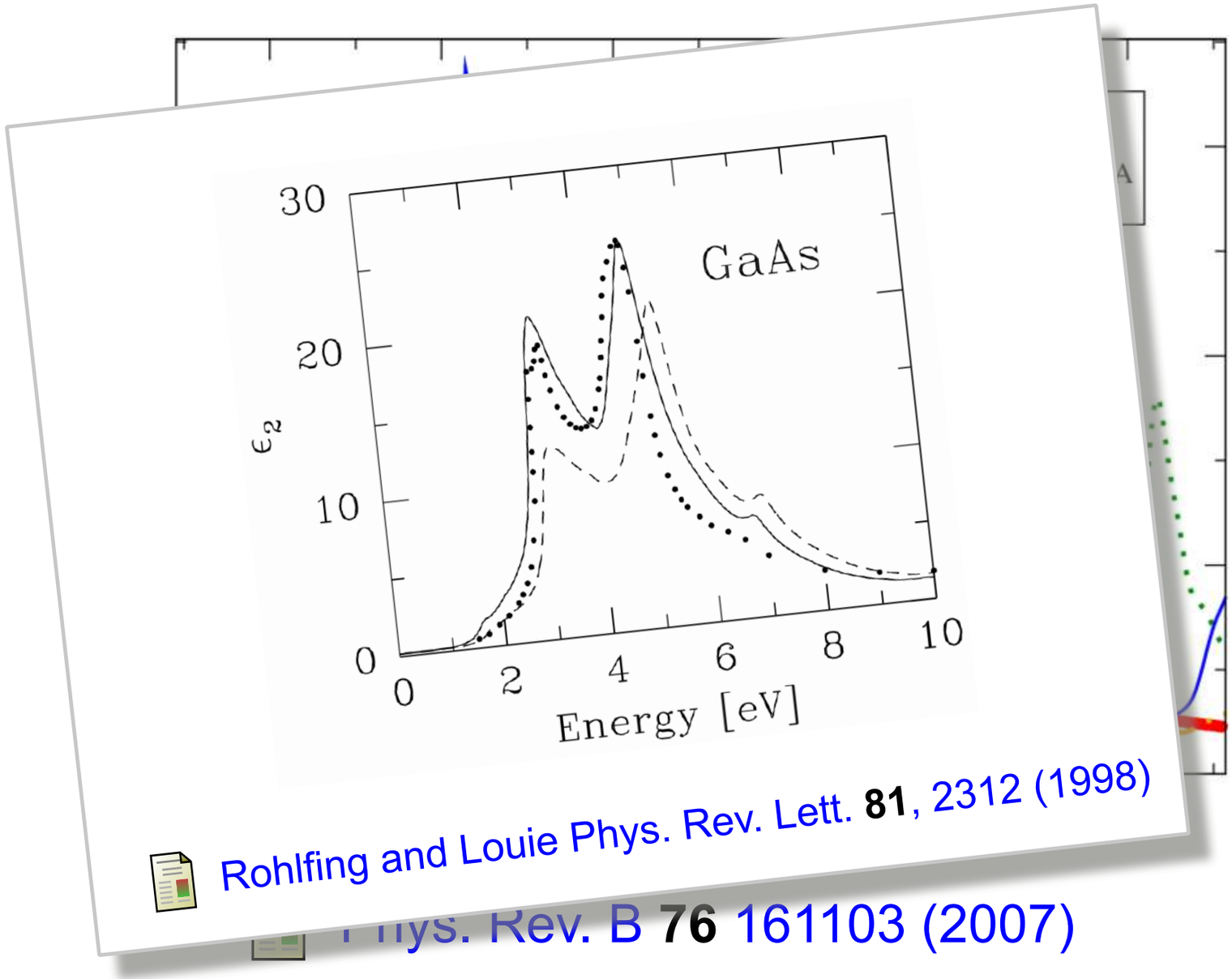




Phys. Rev. B **76** 161103 (2007)



Rohlfing and Louie Phys. Rev. Lett. **81**, 2312 (1998)

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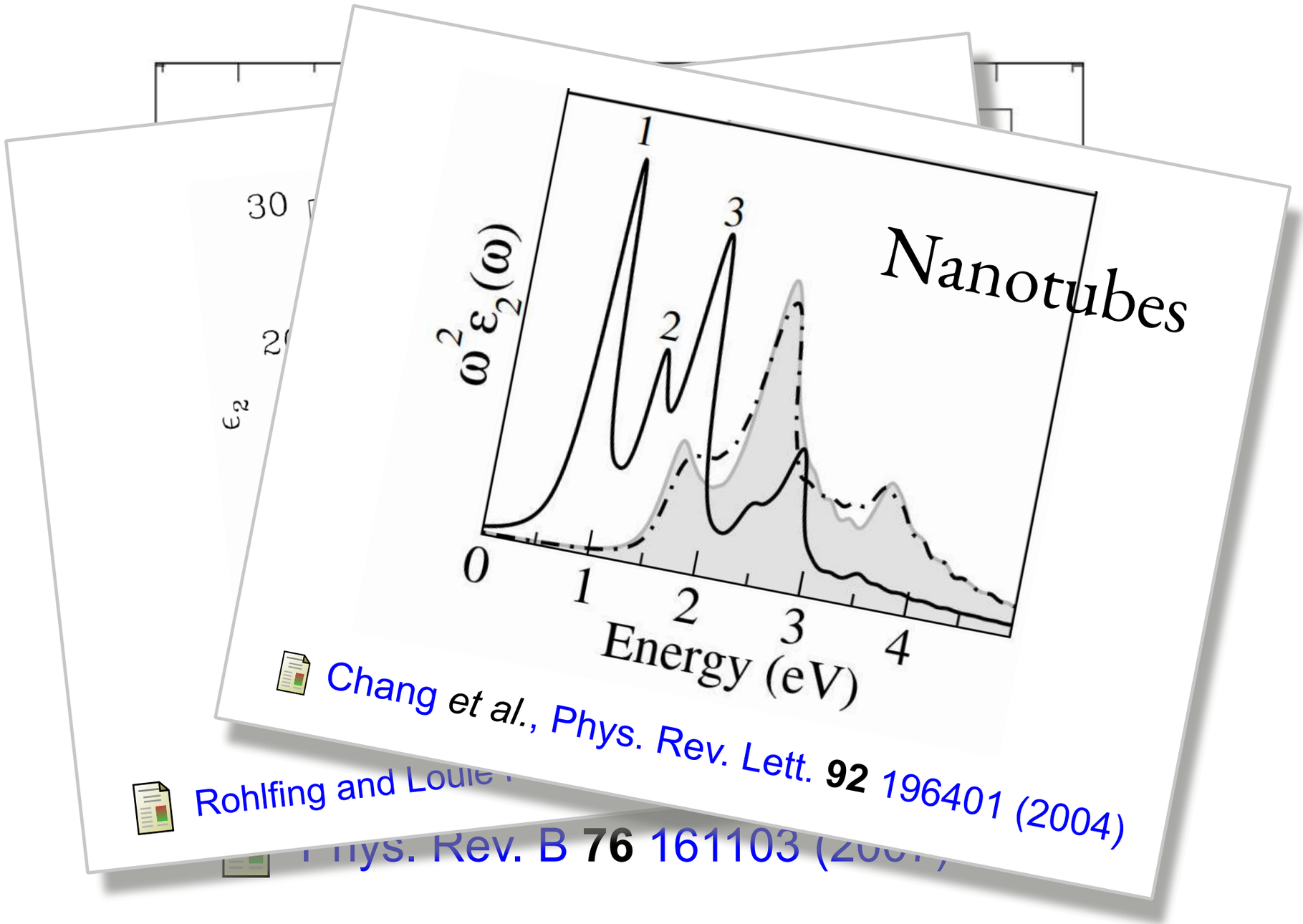
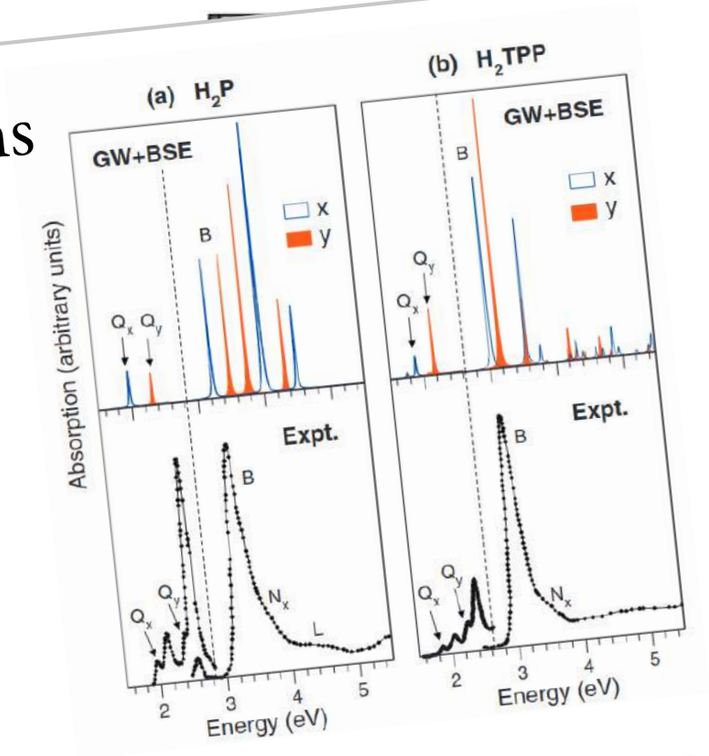

Nanotubes
$\omega^2\varepsilon_2(\omega)$
1
2
3
0
1
2
3
4
Energy (eV)
Chang et al., Phys. Rev. Lett. 92 196401 (2004)
Rohlfing and Louie
Phys. Rev. B 76 161103 (2007)

Porphyrins



Palummo *et al.*, J. Chem. Phys. **131** 084102 (2009)

Chang *et al.*

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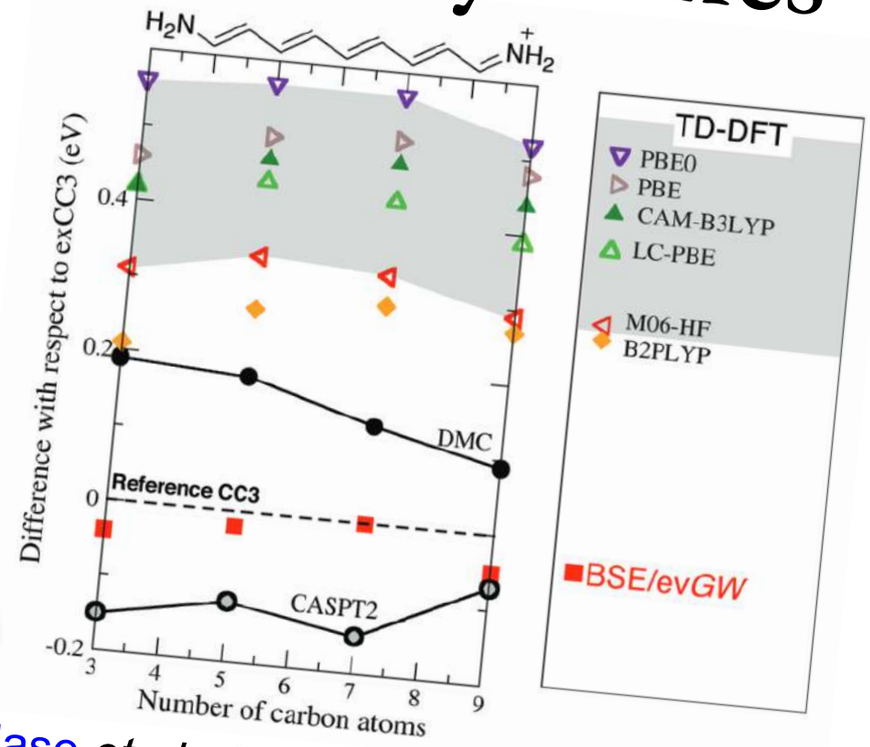
(2004)

streptocyanines



Blase *et al.* Chem. Soc. Rev. **47**, 1022 (2018)

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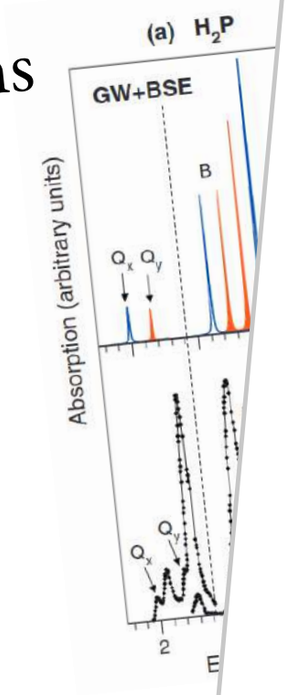
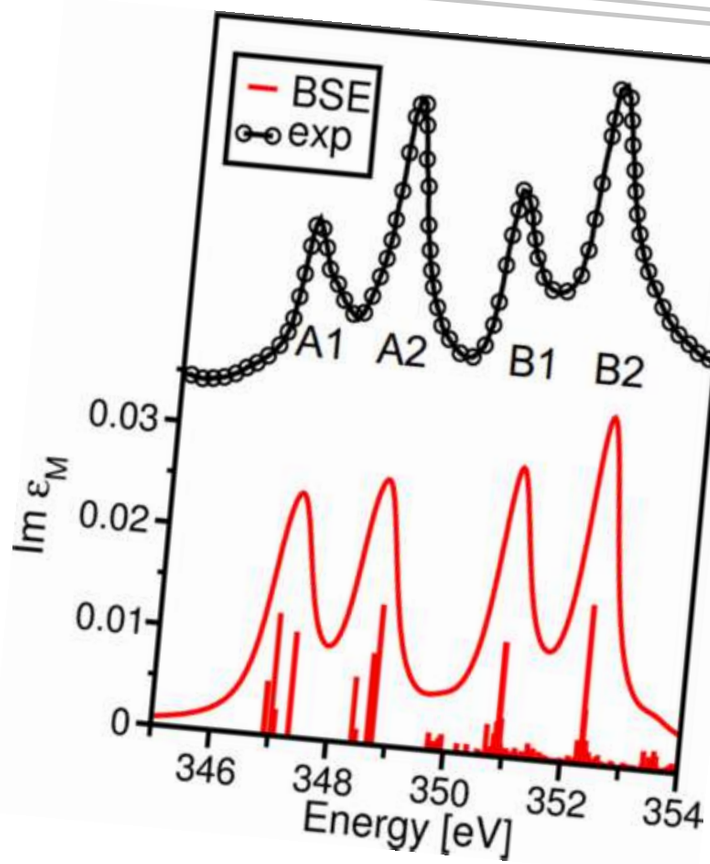


Palummo *et al.*, J.

CaO
Ca L-edge

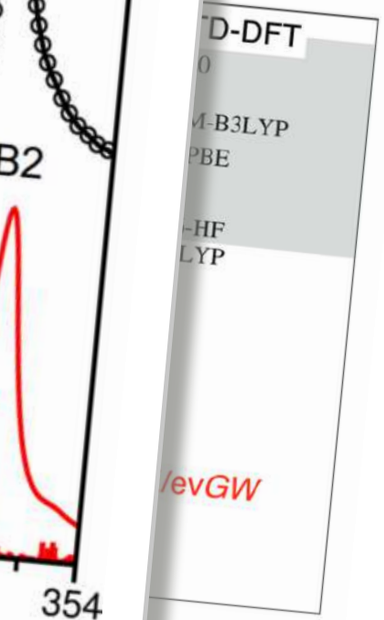


Vorwerk *et al.*, Phys. Rev. B **95**, 155121 (2017)

Rohlfing and

Phys. Rev. B **76** 161103 (2007)

nes



7, 1022 (2018)

4)

DFT/GFT approach to electronic excitations :: successful

- it captures the physics of the electron-hole interaction

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PCCP

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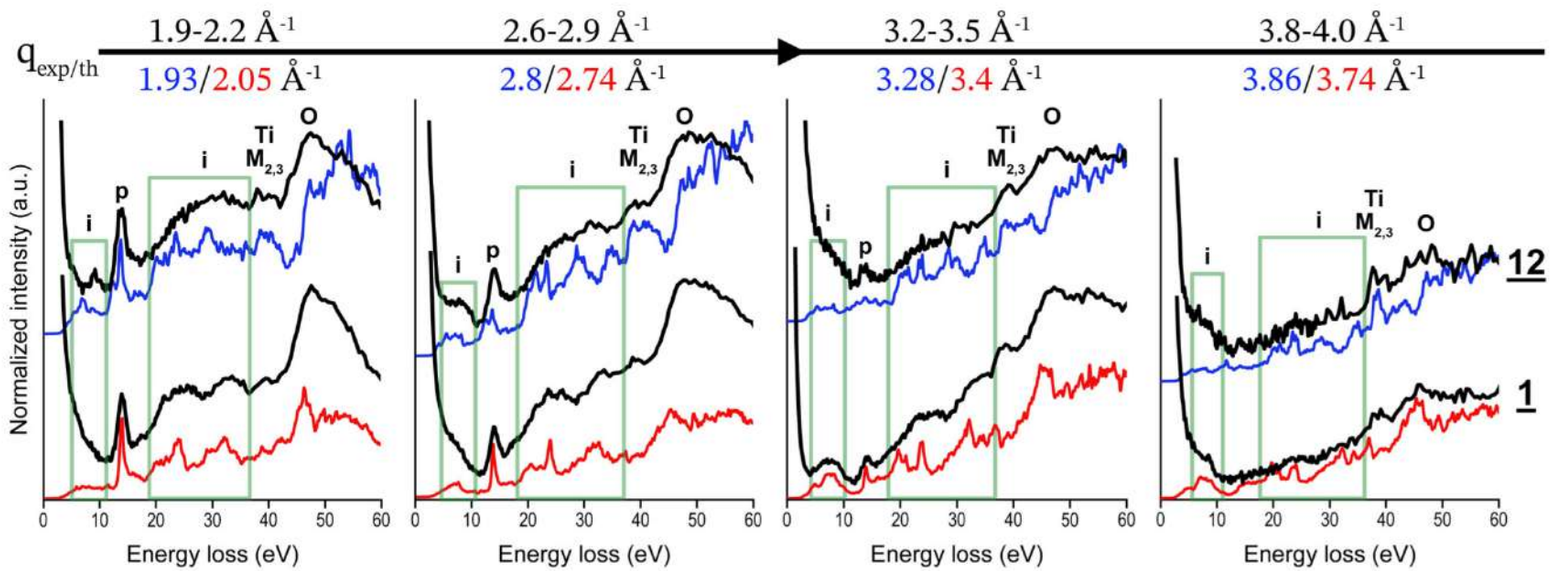
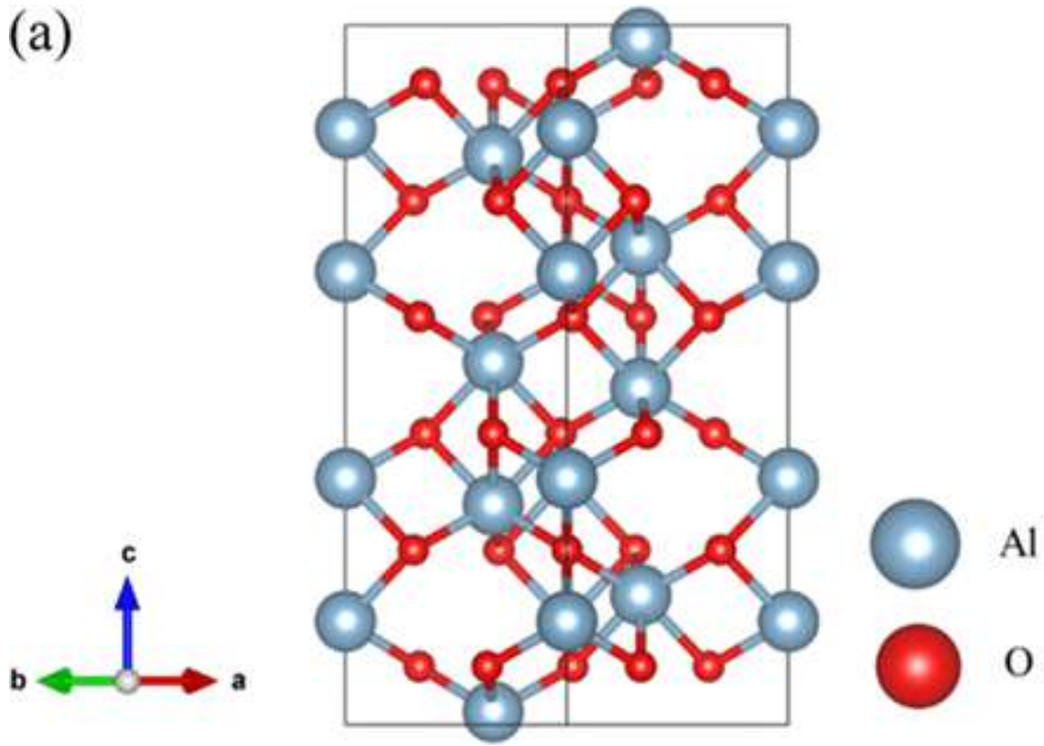
Cite this: *Phys. Chem. Chem. Phys.*, 2024, **26**, 4363

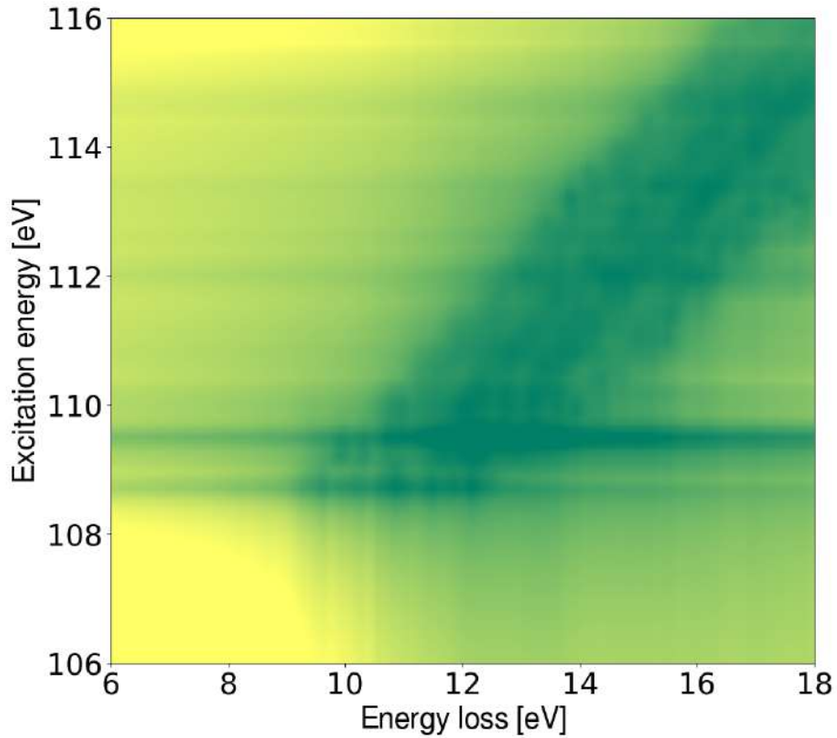
Non-resonant inelastic X-ray scattering for discrimination of pigments†

Lauren Dalecky,[a] Francesco Sottile,[b] Linda Hung,[c] Laure Cazals,[a] Agnès Desolneux,[d] Aurélia Chevalier,[e] Jean-Pascal Rueff[fg] and Loïc Bertrand*[a]



Preliminar RIXS of Al_2O_3 at $L_{2,3}$ edge of Al





M.L.Urquiza, M.Gatti, F.Sottile Phys. Rev. B **109**, 115157 (2024)

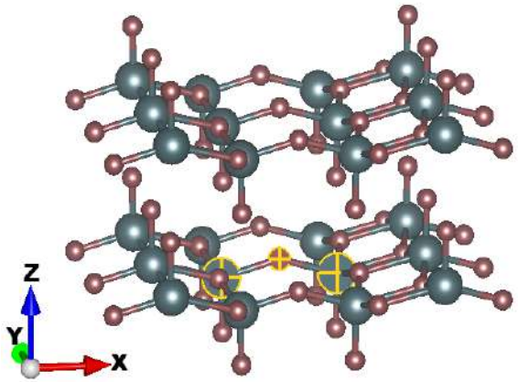
- Beamtime for Abs and RIXS in L_1 and $L_{2,3}$ edge of Al at SOLEIL (A.Nicolau)
- Beamtime for time-dependent RIXS in hBN at FERMI (M.Malvestuto)

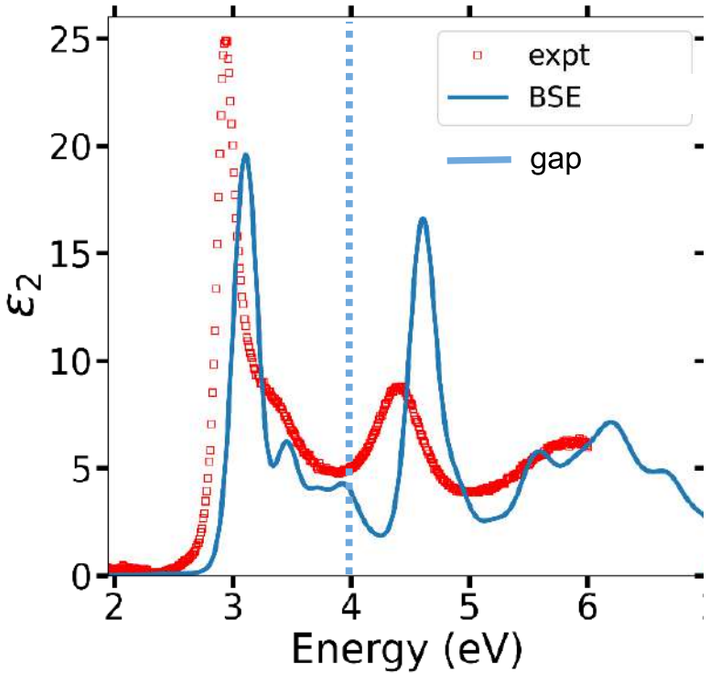
DFT/GFT approach to electronic excitations :: successful

- it captures the physics of the electron-hole interaction
- it can (automatically) profit from extensions
- *ab initio* $\longrightarrow$ predictions
- analysis tools (why? how? who is responsable?)

Excitons in V_2O_5

Why V_2O_5?
- ★ layered, complex material
- ★ applications in photovoltaics
- ★ optical properties not understood





Gorelov *et al.* npj Comput. Mat. **8**, 94 (2022)

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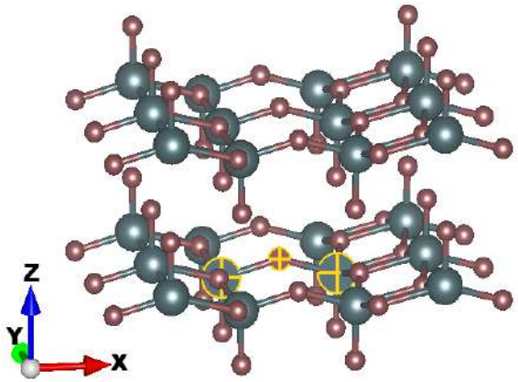
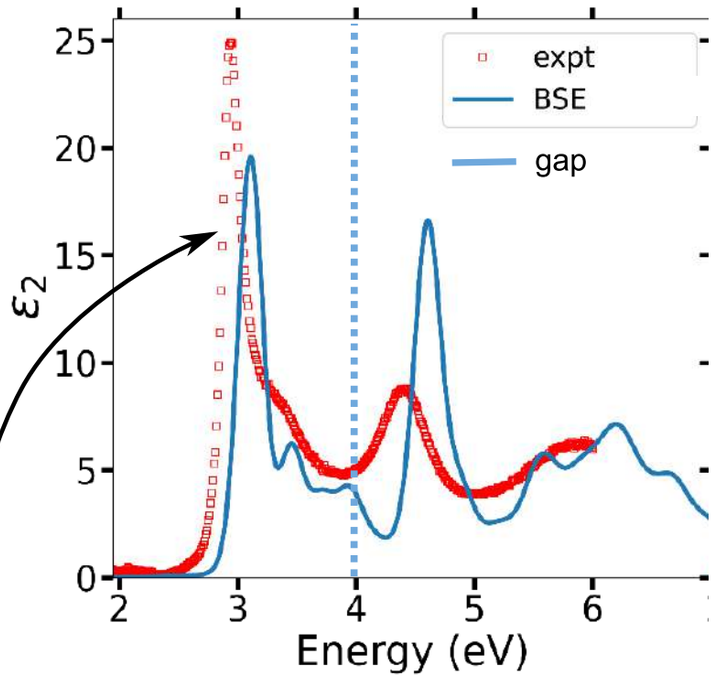
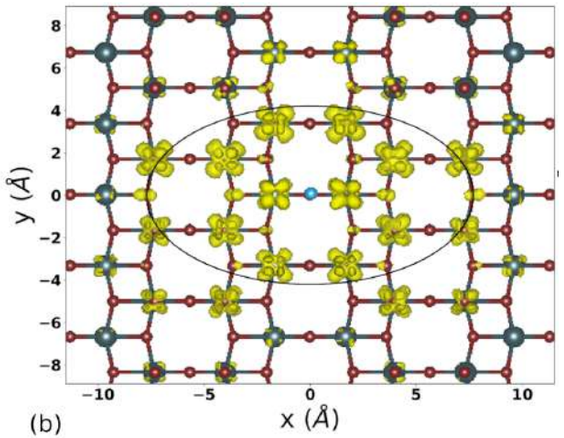




Exciton wave function



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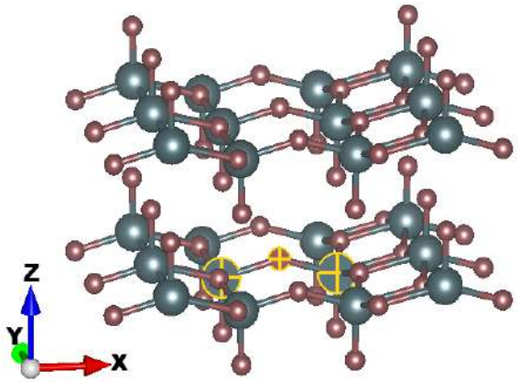
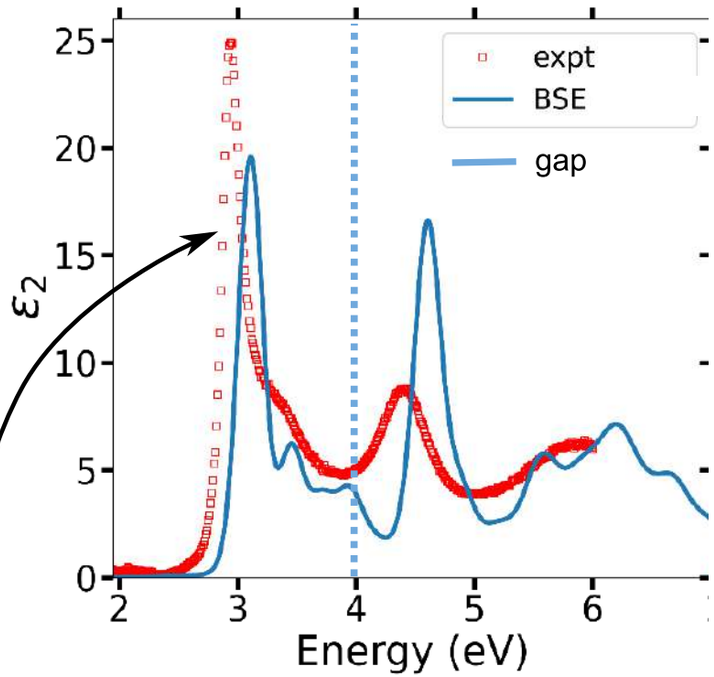
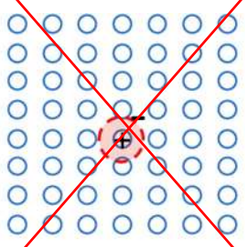


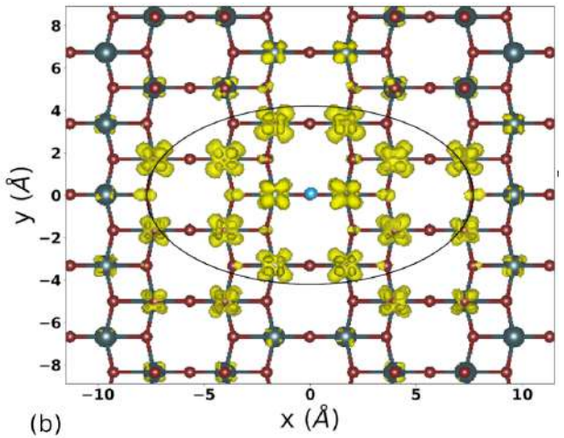


Exciton wave function

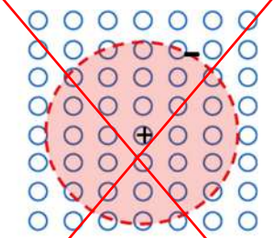
Textbook



Frenkel exciton
Binding energy ~1 eV



Textbook



Wannier-Mott exciton
Binding energy ~10 meV

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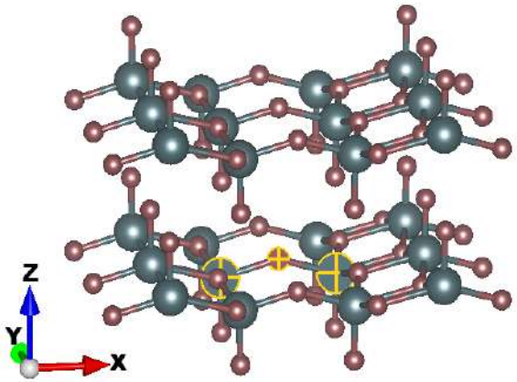
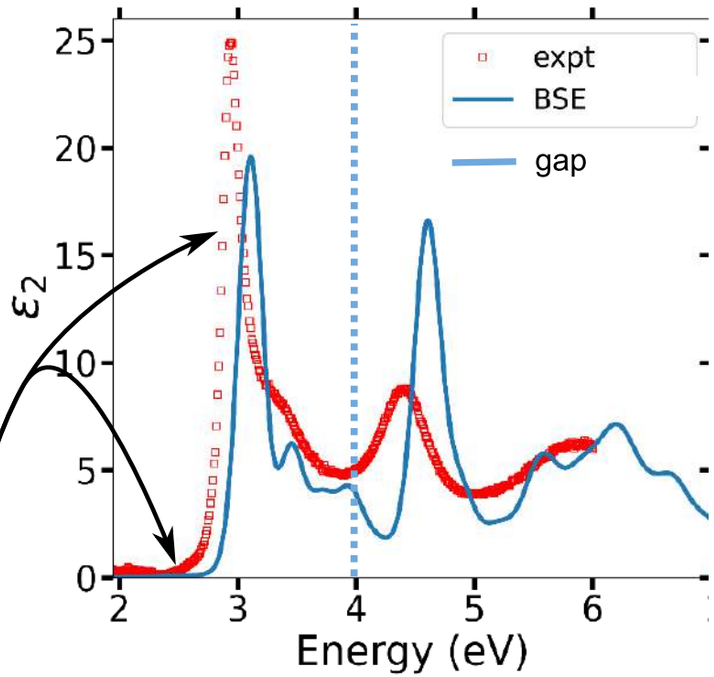
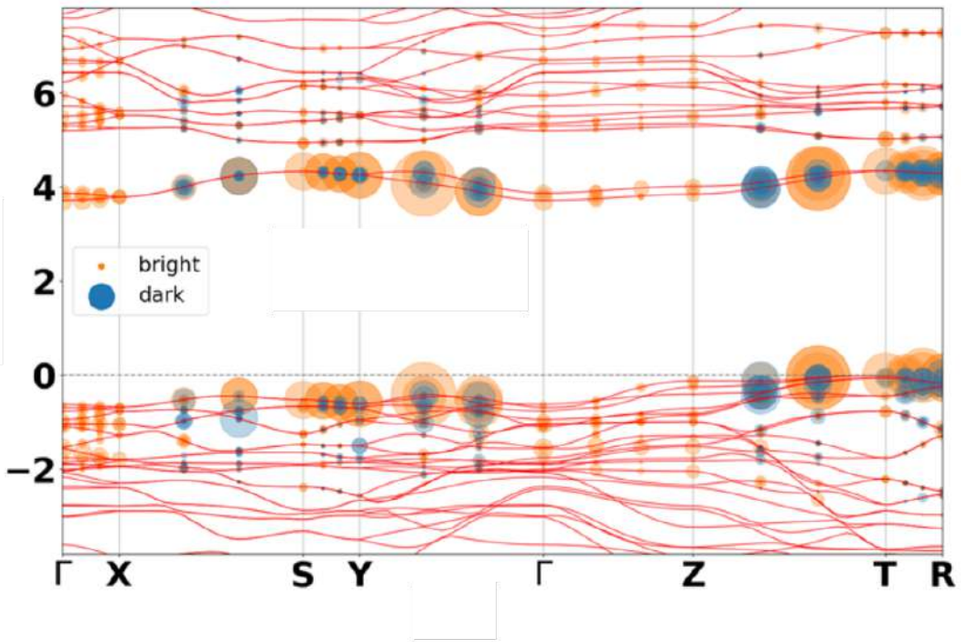
Excitons in V_2O_5

Why V_2O_5?
- ★ layered, complex material
- ★ applications in photovoltaics
- ★ optical properties not understood







origin of the excitons

(bright and dark)

from band-structure analysis

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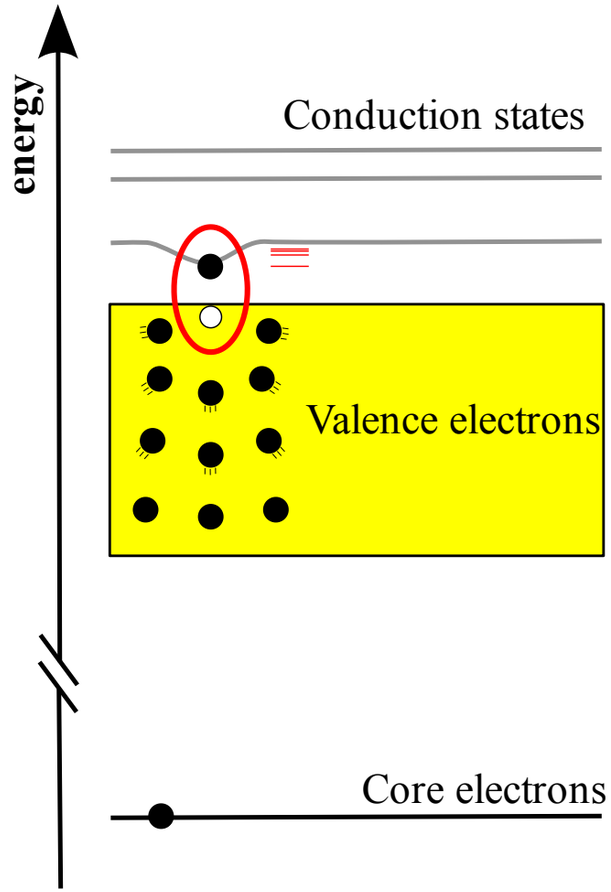
DFT/GFT approach to electronic excitations :: successful

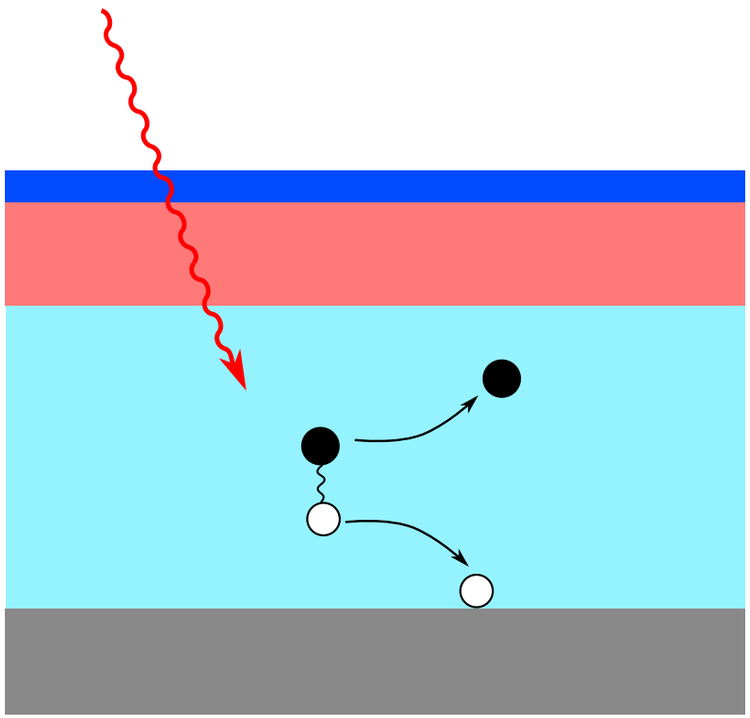
- it captures the physics of the electron-hole interaction
- it can (automatically) profit from extensions
- *ab initio* $\longrightarrow$ predictions
- analysis tools (why? how? who is responsable?)

Outline

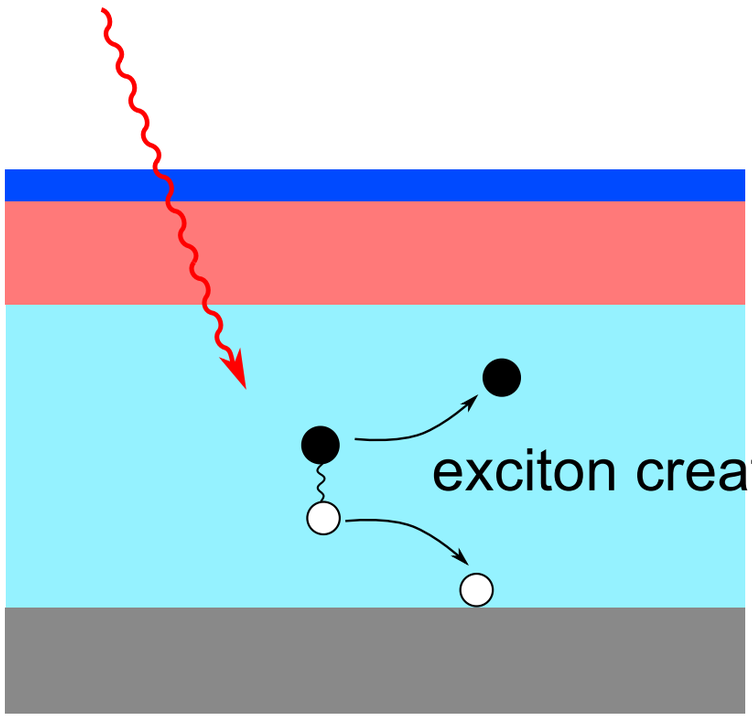
- (our) Approach to electronic excitations
- Results for absorption spectroscopy
- Advantages, limitations, opportunities



DFT/GFT approach to electronic excitations :: very far from ....

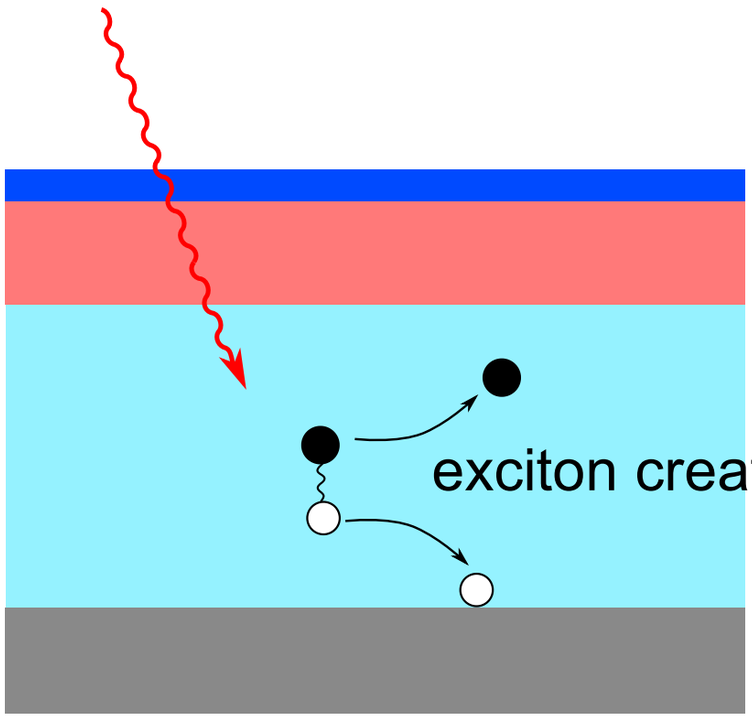


DFT/GFT approach to electronic excitations :: very far from ....



exciton creation and separation (and migration) of charges

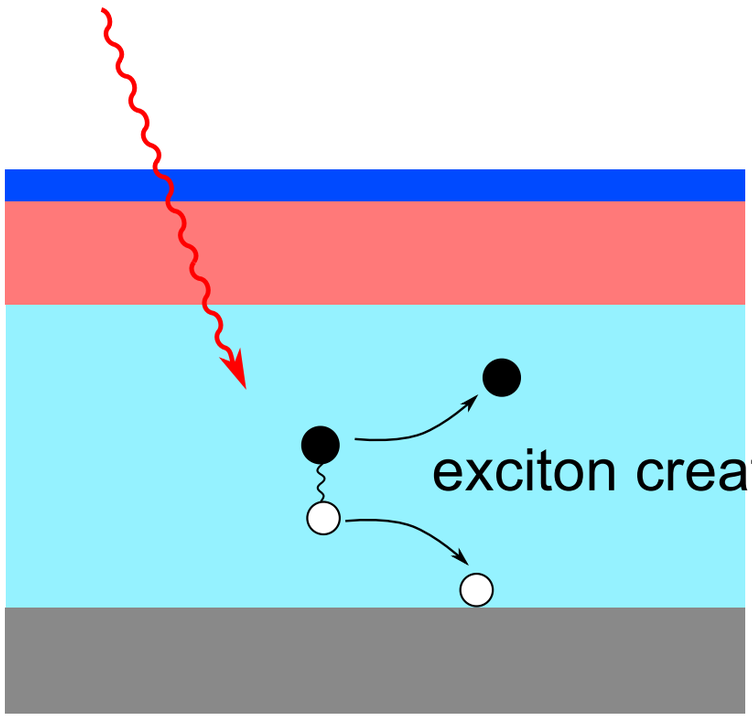
DFT/GFT approach to electronic excitations :: very far from ....



exciton creation and separation (and migration) of charges

avoid recombination

DFT/GFT approach to electronic excitations :: very far from ....

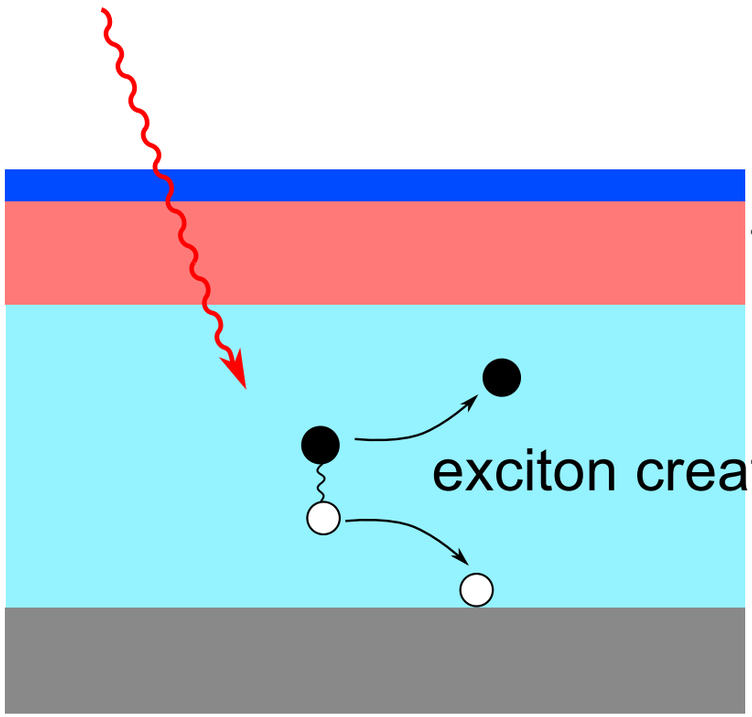


exciton creation and separation (and migration) of charges

avoid recombination

interface (structure, matching, band alignment)

DFT/GFT approach to electronic excitations :: very far from ....



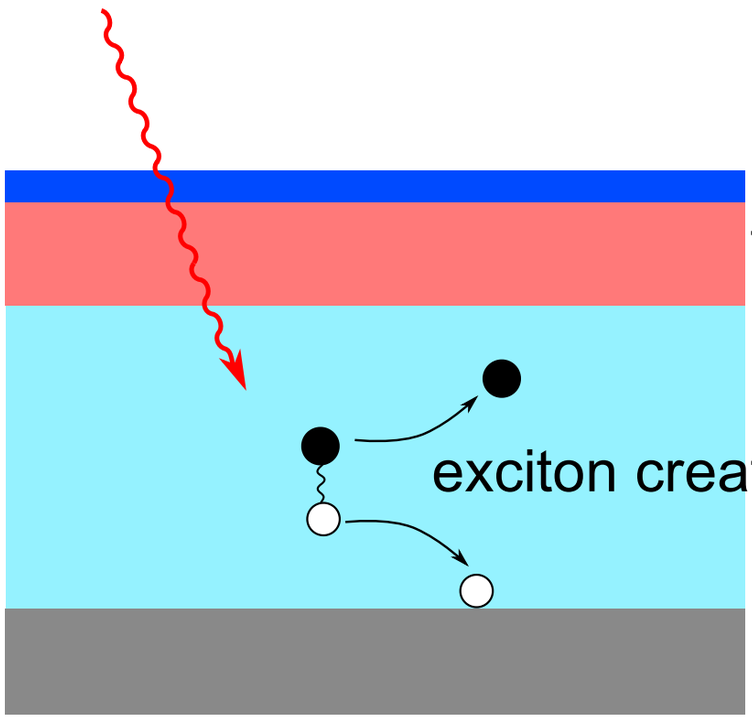
transparent and conducting material

exciton creation and separation (and migration) of charges

avoid recombination

interface (structure, matching, band alignment)

DFT/GFT approach to electronic excitations :: very far from ....



transparent and conducting material

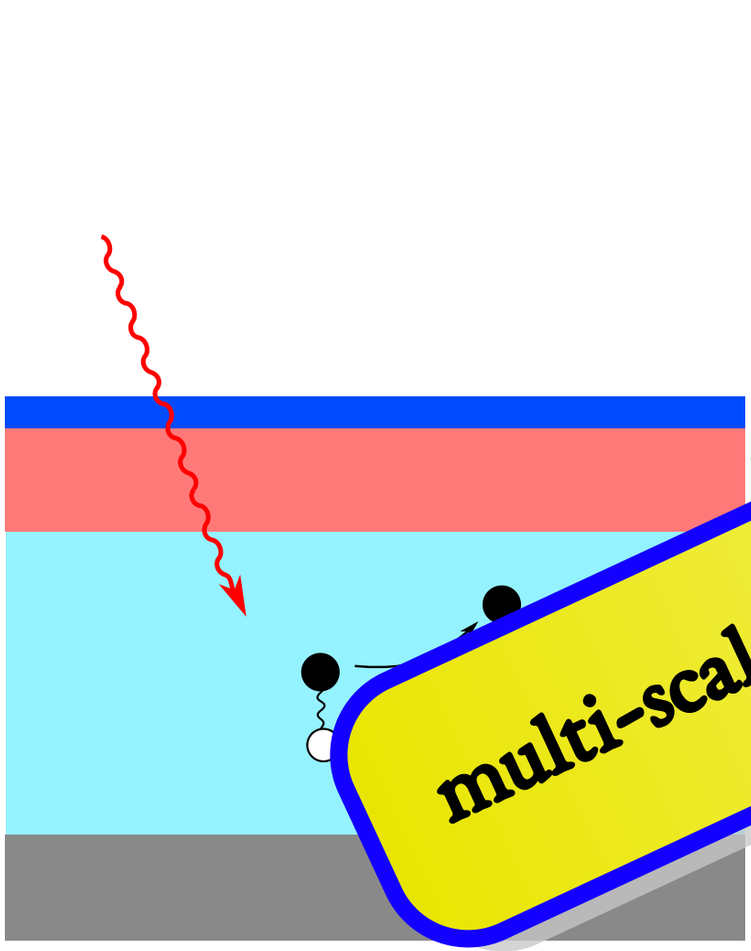
exciton creation and separation (and migration) of charges

avoid recombination

interface (structure, matching, band alignment)

temperature, strain, hygrometry

DFT/GFT approach to electronic excitations :: very far from ....



multi-scale(approx,approach) methods needed

tran… …material

…eparation (and migration) of charges

avoid recombination

interface (structure, matching, band alignment)

temperature, strain, hygrometry

Opportunities and challenges

- Stronger synergy between theory and experiment
  - benchmarking and devise better approximations
- Reinforce connection with math and computer scientists
  - better algorithms
- Exploit next generation tools
  - new architectures (GPU, TPU, vector engines)
  - high throughput procedures
  - machine learning