Electronic excitations via Inelastic X-ray Scattering: Green's functions approach

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IXS2022

The 12th International Conference on Inelastic X-ray Scattering

- Green's functions approach to spectra

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- Excitonic effects in (non-)resonant, coherent IXS

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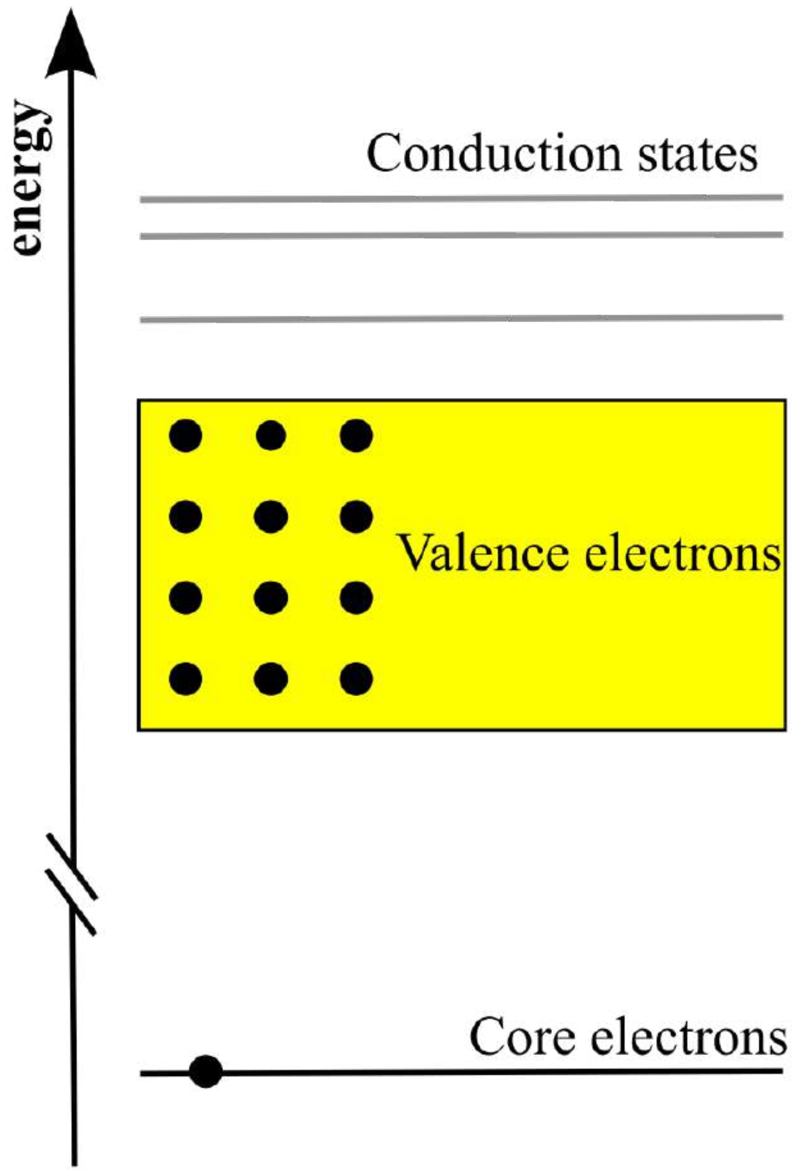
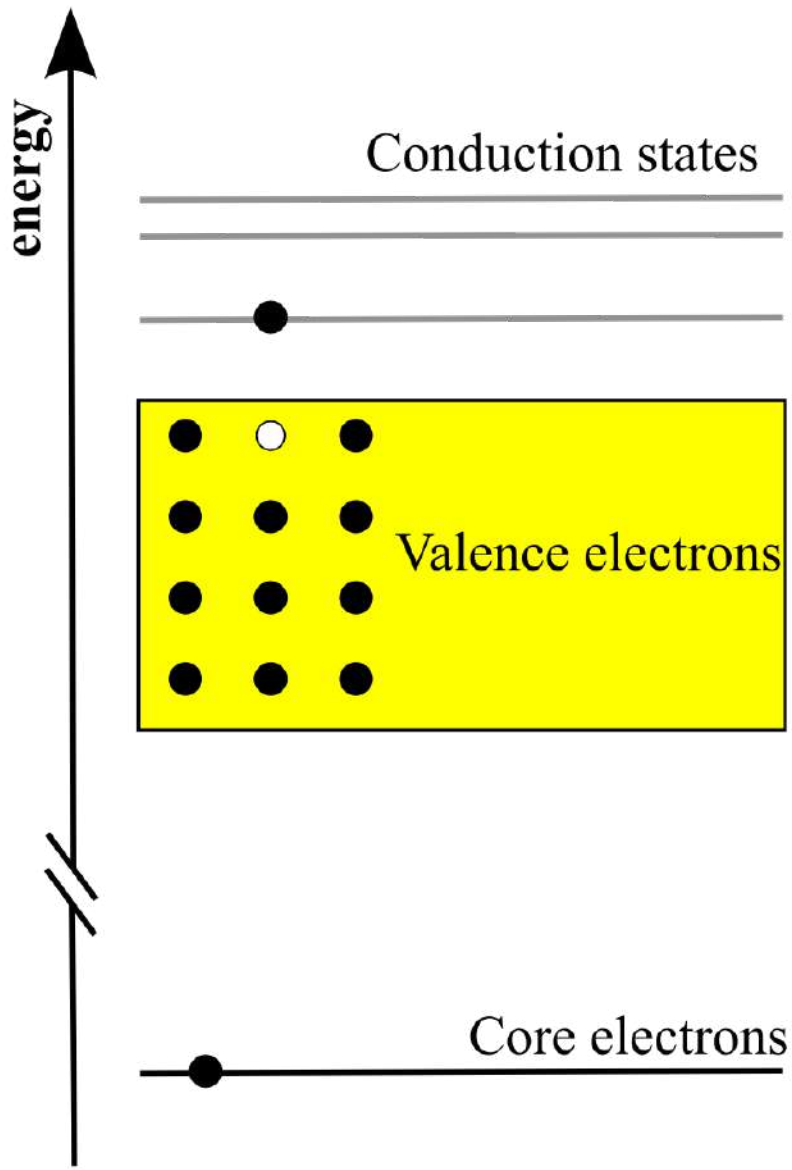
- Coherence in RIXS


energy
Conduction states
Valence electrons
Core electrons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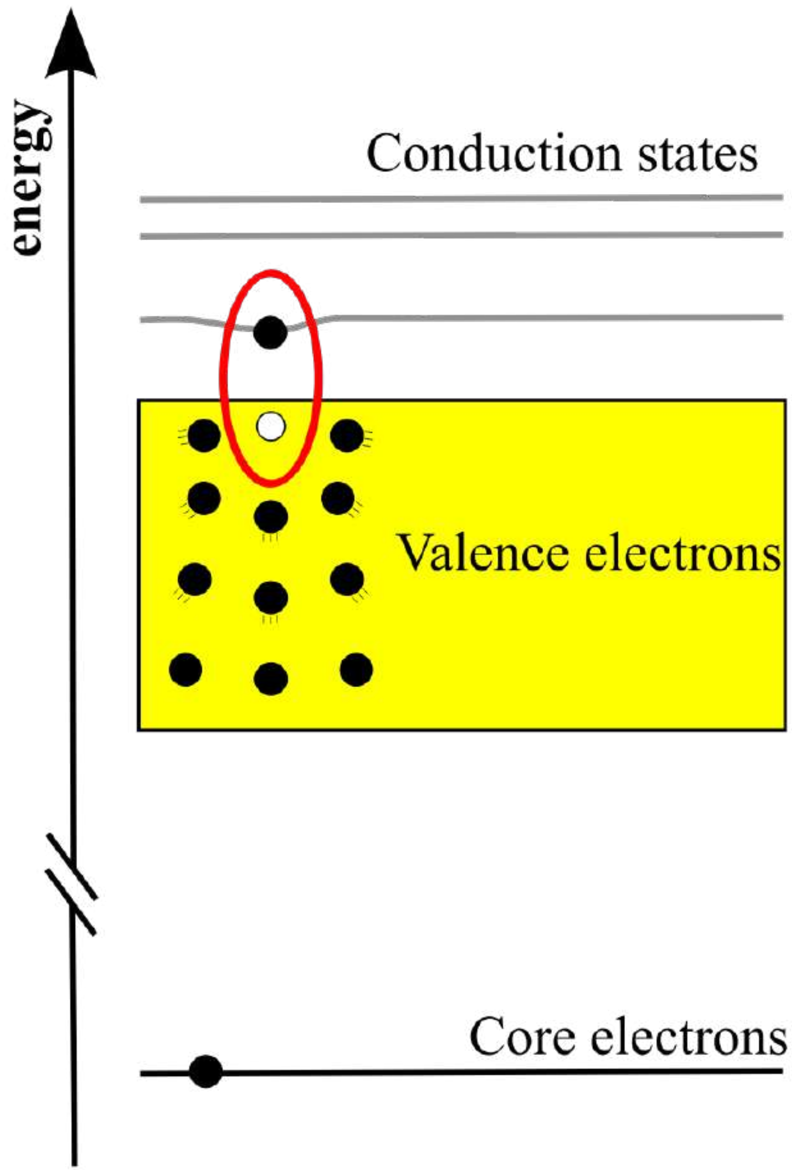

energy
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The Bethe-Salpeter Equation

$$L(1,2,3,4) = L^0(1,2,3,4) + L^0(1,2,5,6)\left[v(5,7)\delta(5,6)\delta(7,8) + i\frac{\delta\Sigma(5,6)}{\delta G(7,8)}\right]L(7,8,3,4)$$

$$1 = (\mathbf{r}_1, t_1, \sigma_1)$$

The Bethe-Salpeter Equation

$$2, 3, 4) = L^0(1, 2, 3, 4) + L^0(1, 2, 5, 6)\left[v(5, 7)\delta(5, 6)\delta(7, 8) + i\frac{\delta\Sigma(5, 6)}{\delta G(7, 8)}\right] L(7, 8$$

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$$-iL(1, 2, 1^+, 2^+) = \chi(1, 2) = \frac{\delta n(1)}{\delta V_{\text{ext}}(2)}$$

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Excitations energies

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Electron Energy Loss

Excitations energies

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Inelastic X-ray Scattering

The Bethe-Salpeter Equation

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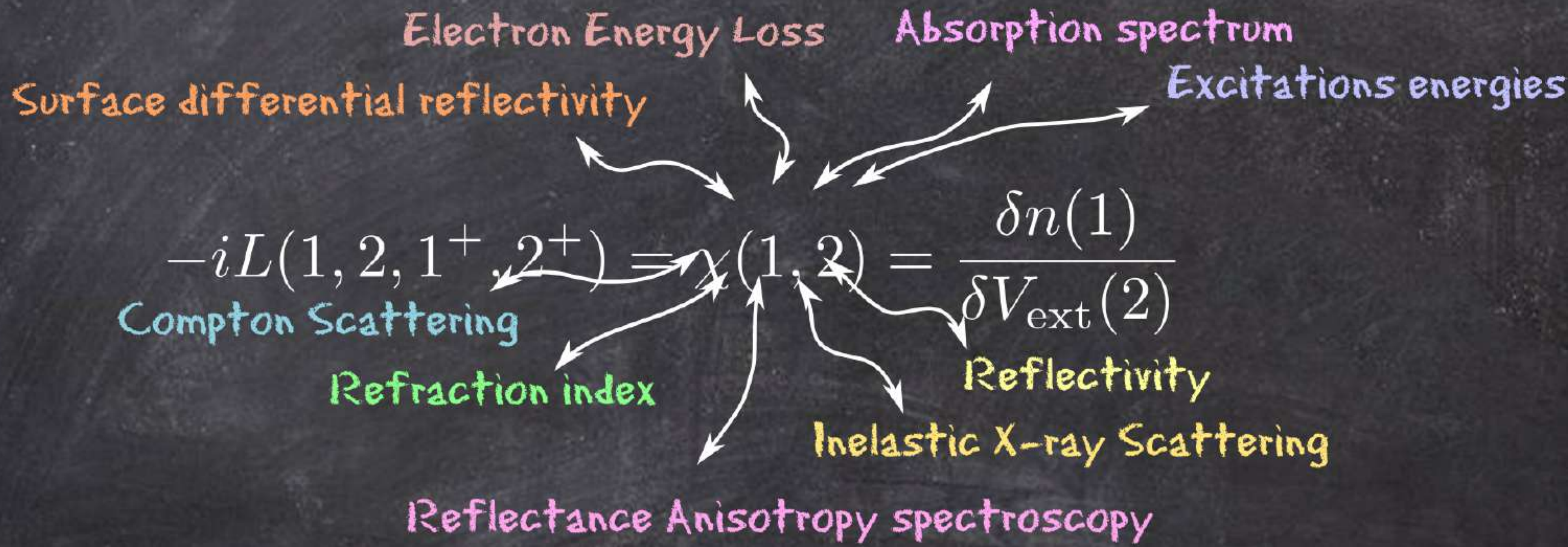

$$H^{\mathrm{BSE}} A_\lambda = E_\lambda A_\lambda$$

$$\sum_{p'h'} \begin{pmatrix} A & C \\ -C^* & -A^* \end{pmatrix}_{php'h'} \begin{pmatrix} X_\lambda^{p'h'} \\ Y_\lambda^{p'h'} \end{pmatrix} = E_\lambda \begin{pmatrix} X_\lambda^{ph} \\ Y_\lambda^{ph} \end{pmatrix}$$

particle-hole space

BSE, TDDFT, RPA

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$$\sum_{t'} \begin{pmatrix} A & C \\ -C^* & -A^* \end{pmatrix}_{tt'} \begin{pmatrix} X_\lambda^{t'} \\ Y_\lambda^{t'} \end{pmatrix} = E_\lambda \begin{pmatrix} X_\lambda^{t} \\ Y_\lambda^{t} \end{pmatrix}$$

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Tamm-Dancoff approx

$$\sum_{t'} \begin{pmatrix} A & \cancel{C} \\ -\cancel{C}^* & -A^* \end{pmatrix}_{tt'} \begin{pmatrix} X_\lambda^{t'} \\ Y_\lambda^{t'} \end{pmatrix} = E_\lambda \begin{pmatrix} X_\lambda^{t} \\ Y_\lambda^{t} \end{pmatrix}$$

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$$\chi_M(\omega) = \sum_\lambda \frac{\left| \sum_{t=vc} X_\lambda^{vc} \langle c|\hat{\mathbf{d}}|v\rangle \right|^2}{\omega - E_\lambda + i\eta}$$

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BSE

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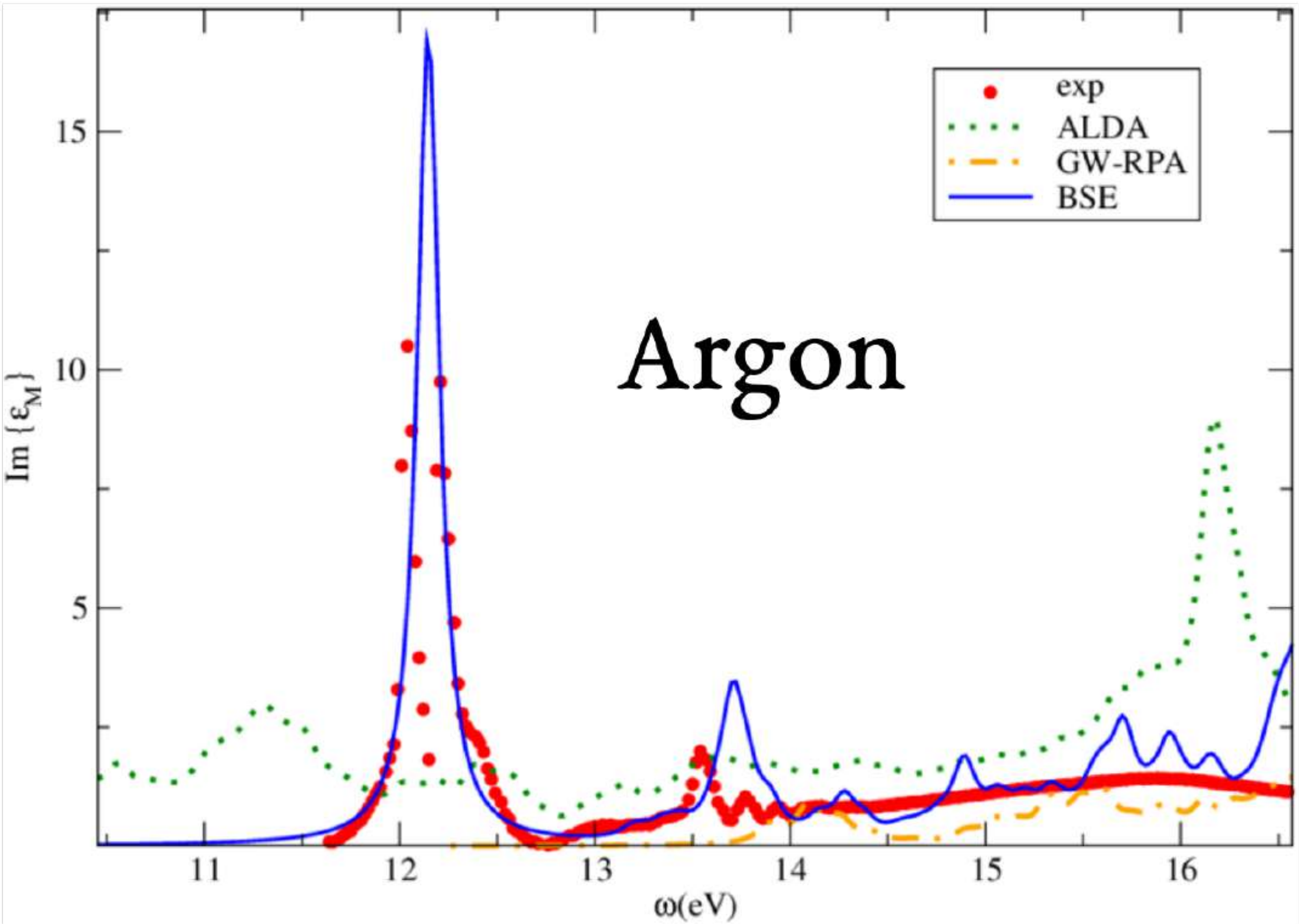
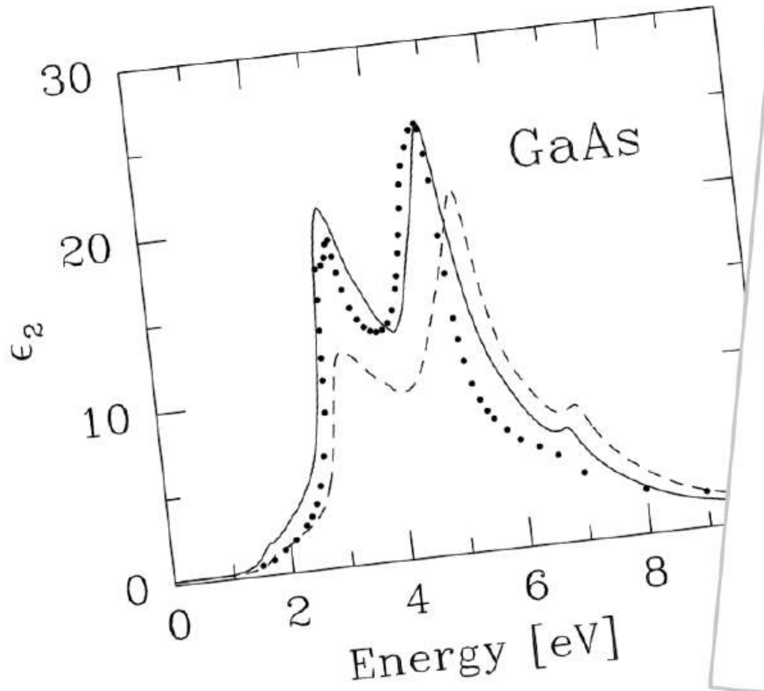
BSE

$$\chi_M^0(\omega) = \sum_{vc} \frac{\left| \langle c|\hat{\mathbf{d}}|v\rangle \right|^2}{\omega - (\varepsilon_c - \varepsilon_v) + i\eta}$$

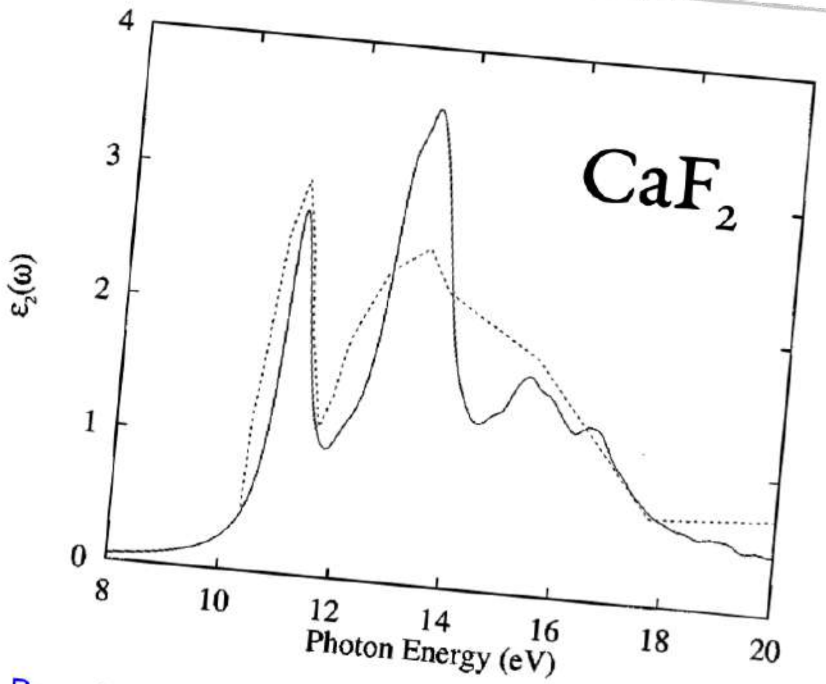
IPA



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

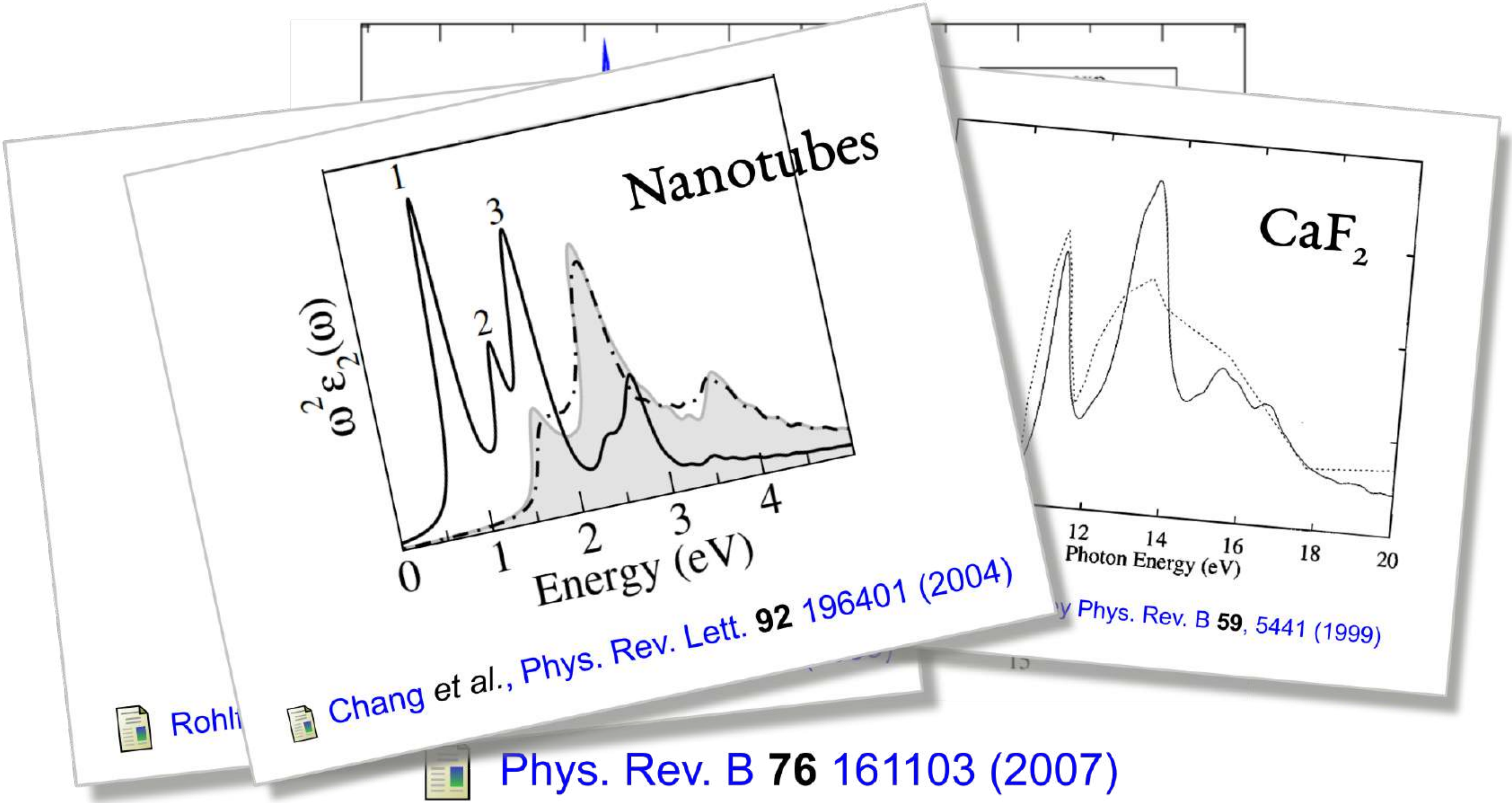


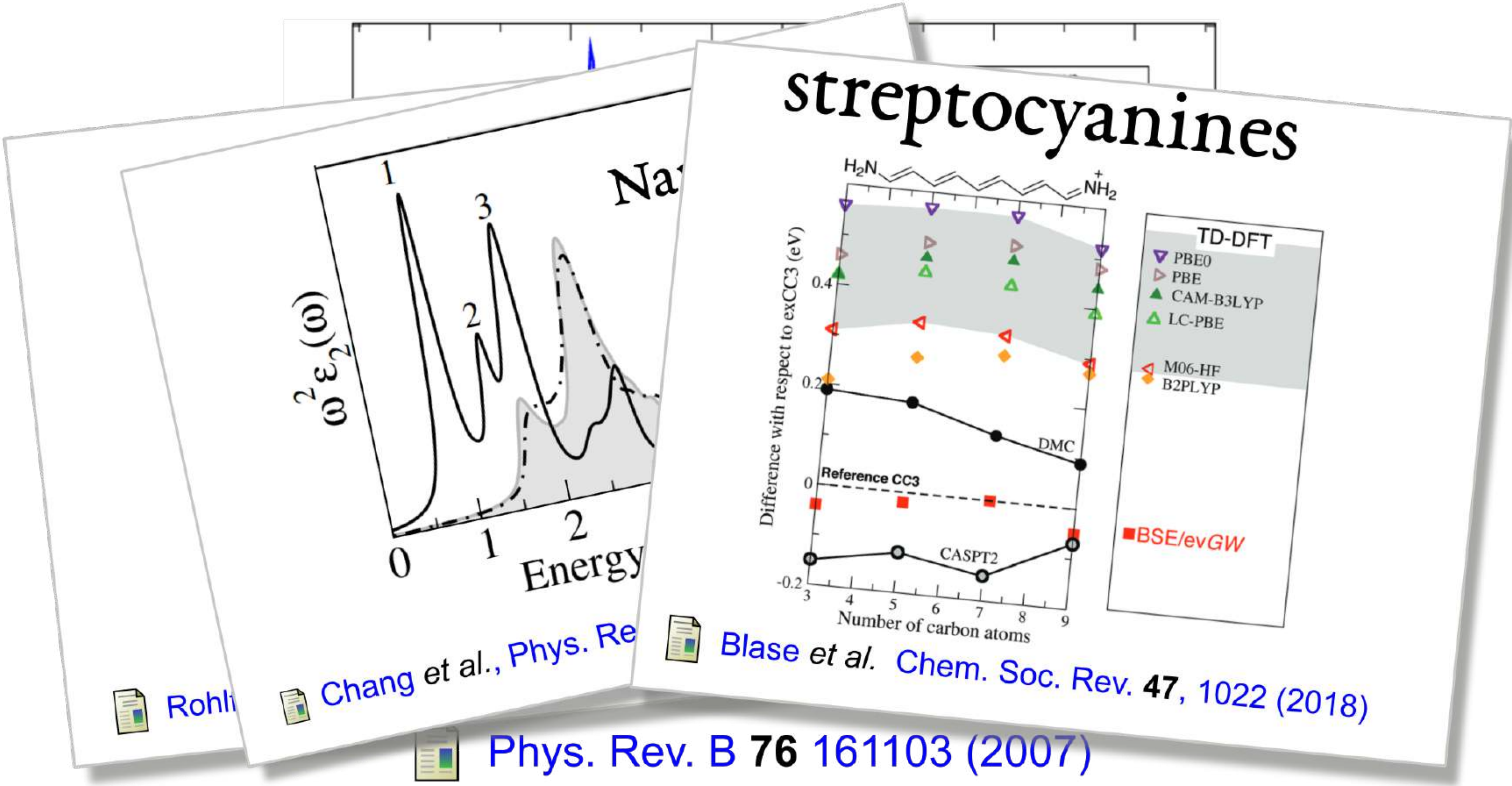
Rohlfing and Louie Phys. Rev. Lett. **81**, 2312 (1998)



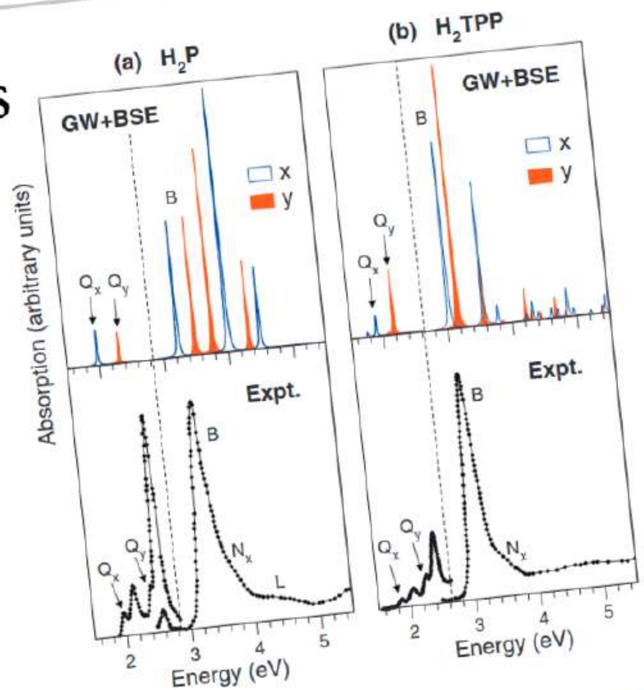
Benedict and Shirley Phys. Rev. B **59**, 5441 (1999)

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


Nanotubes
1
2
3
$\omega^2\varepsilon_2(\omega)$
0
1
2
3
4
Energy (eV)
Chang et al., Phys. Rev. Lett. 92 196401 (2004)
Rohl
CaF2
12
14
16
18
20
Photon Energy (eV)
Phys. Rev. B 59, 5441 (1999)
Phys. Rev. B 76 161103 (2007)

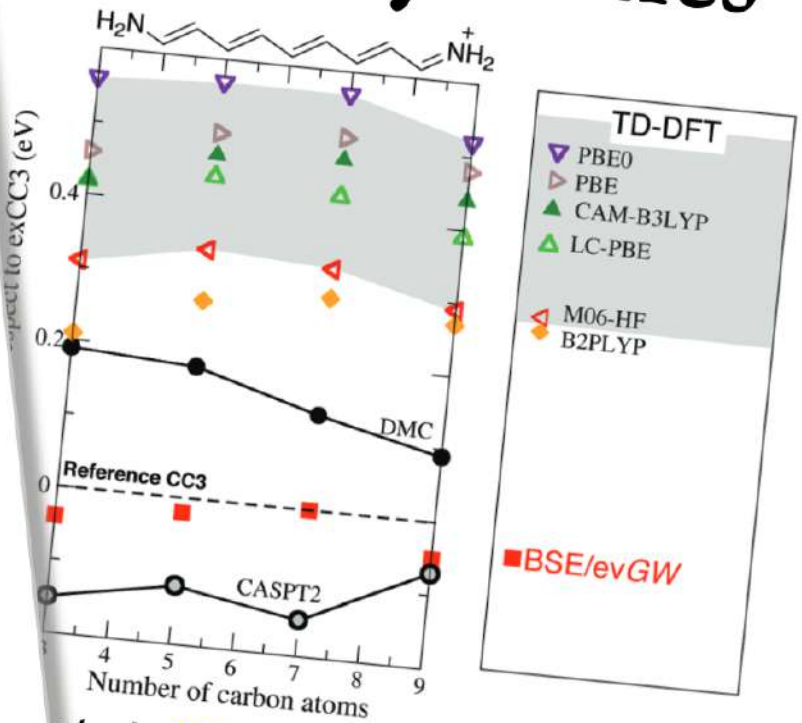

streptocyanines
H_2N
$\overset{+}{N}H_2$
TD-DFT
PBE0
PBE
CAM-B3LYP
LC-PBE
M06-HF
B2PLYP
BSE/evGW
DMC
Reference CC3
CASPT2
Difference with respect to exCC3 (eV)
Number of carbon atoms
Blase et al. Chem. Soc. Rev. 47, 1022 (2018)
$\omega^2\varepsilon_2(\omega)$
Energy
Chang et al., Phys. Re
Rohl
Phys. Rev. B 76 161103 (2007)

Porphyrins



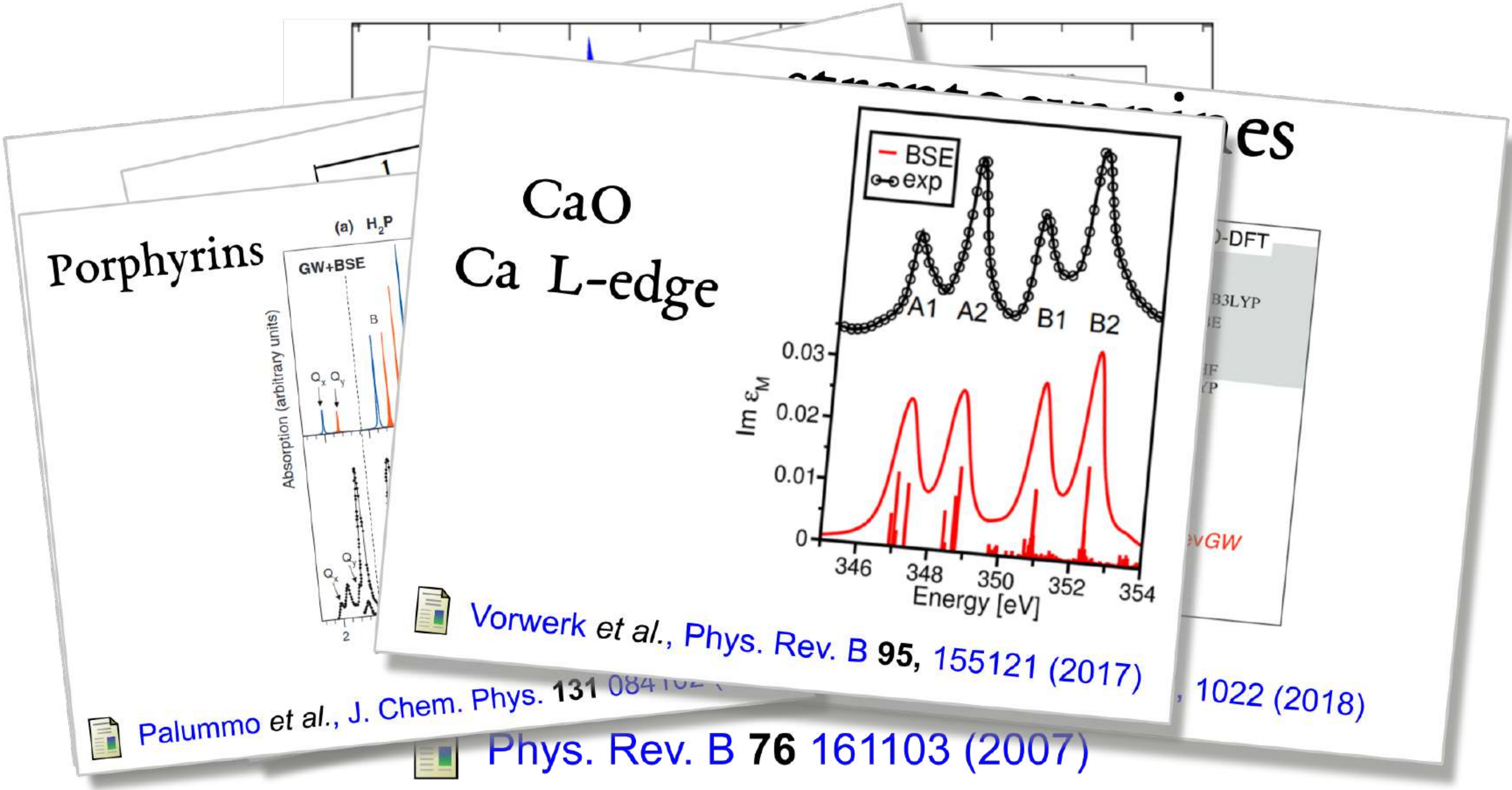
Palummo *et al.*, J. Chem. Phys. **131** 084102 (2009)

streptocyanines



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

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


CaO
Ca L-edge
BSE
exp
A1
A2
B1
B2
0.03
0.02
0.01
0
Im ε_M
346
348
350
352
354
Energy [eV]
Vorwerk et al., Phys. Rev. B 95, 155121 (2017)
Porphyrins
(a) H_2P
GW+BSE
Absorption (arbitrary units)
Palummo et al., J. Chem. Phys. 131 084102
Phys. Rev. B 76 161103 (2007)
-DFT
B3LYP
evGW
, 1022 (2018)
es

BSE :: accurate for absorption spectra (and excitation energies)

BSE :: accurate for absorption spectra (and excitation energies)

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

Bethe-Salpeter Equation - finite momentum transfer

$$S(\mathbf{q},\omega) \propto \chi_M(\mathbf{q},\omega) = \sum_\lambda \frac{\left|\sum_{vc} A_\lambda^{vc,\mathbf{q}} \langle c|e^{i\mathbf{q}\cdot\mathbf{r}}|v\rangle\right|^2}{\omega - E_\lambda(\mathbf{q}) + i\eta}$$

- Green's functions approach to spectra
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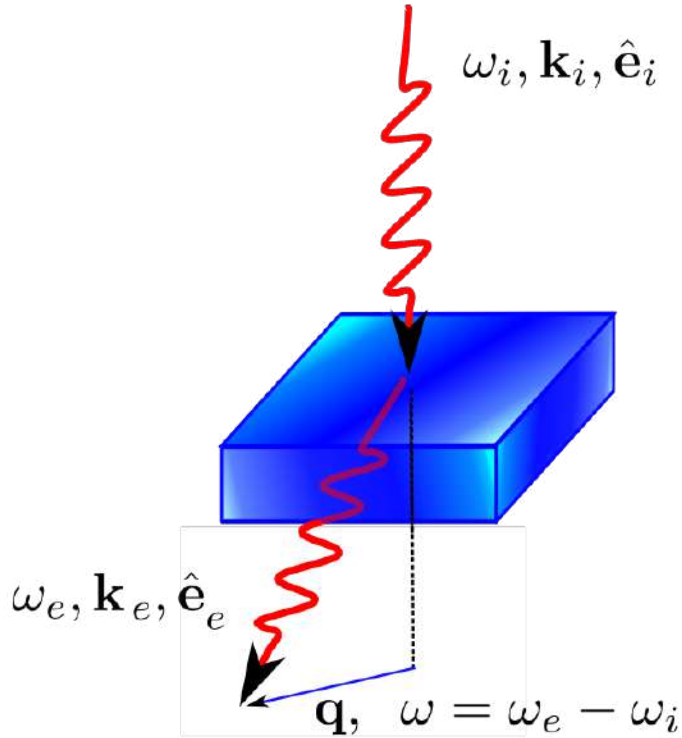
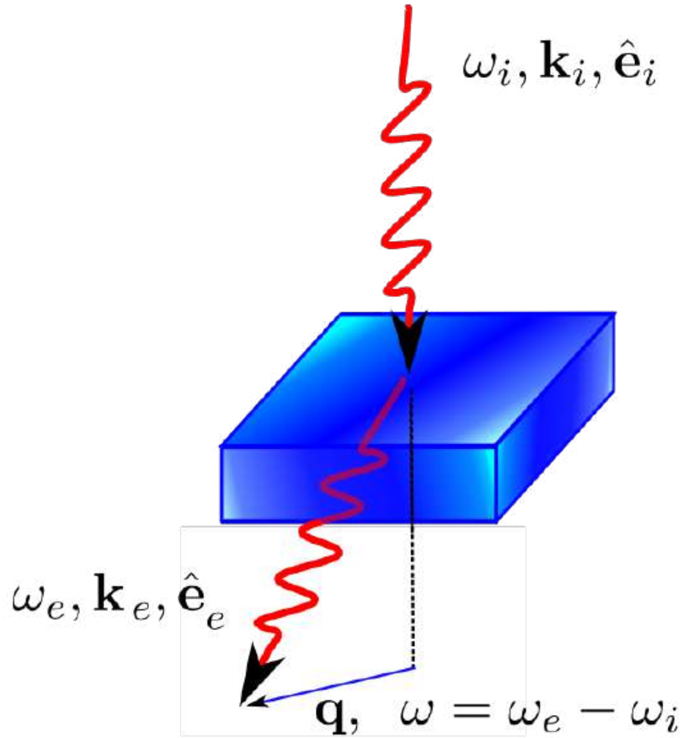
- Coherence in RIXS

X-ray scattering



$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2 \mathrm{d}\omega_e} \propto \sum_f \left| \langle f|e^{i\mathbf{q}\cdot\mathbf{r}}|0\rangle + \frac{\langle f|e^{-i\mathbf{k}_f\cdot\mathbf{r}}\nabla|n\rangle \; \langle n|e^{i\mathbf{k}_i\cdot\mathbf{r}}\nabla|0\rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta\left(\omega - (E_f - E_0)\right)$$

X-ray scattering



non-Resonant IXS

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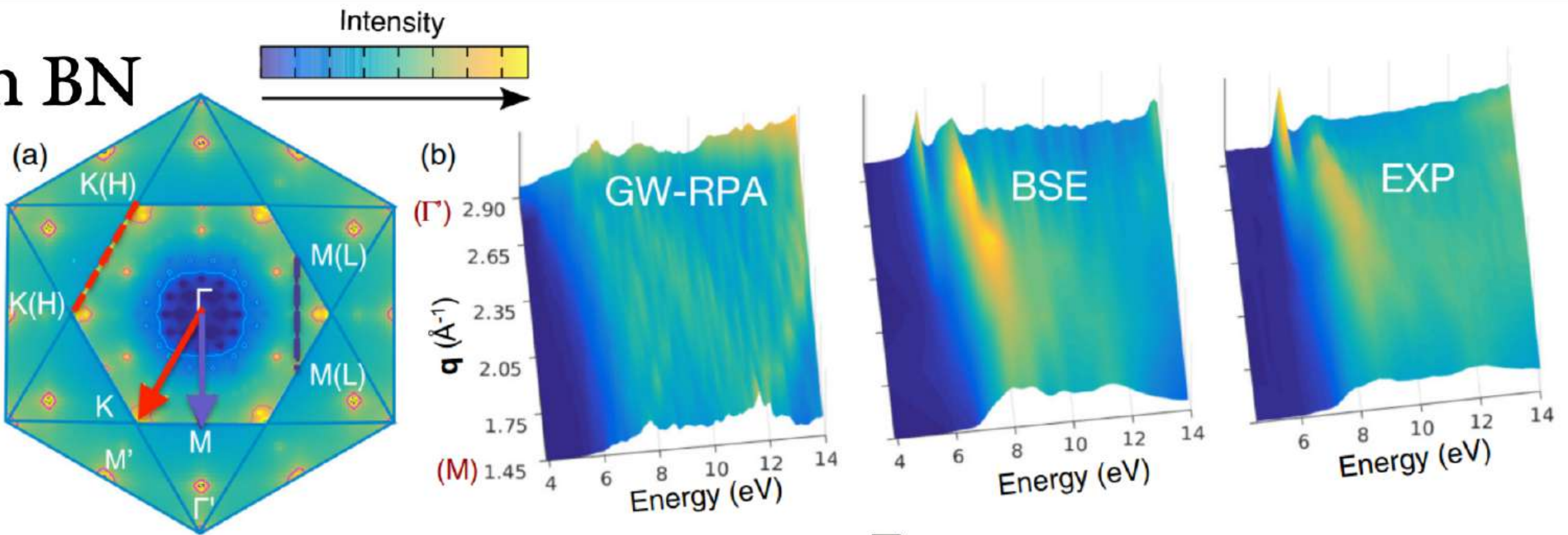
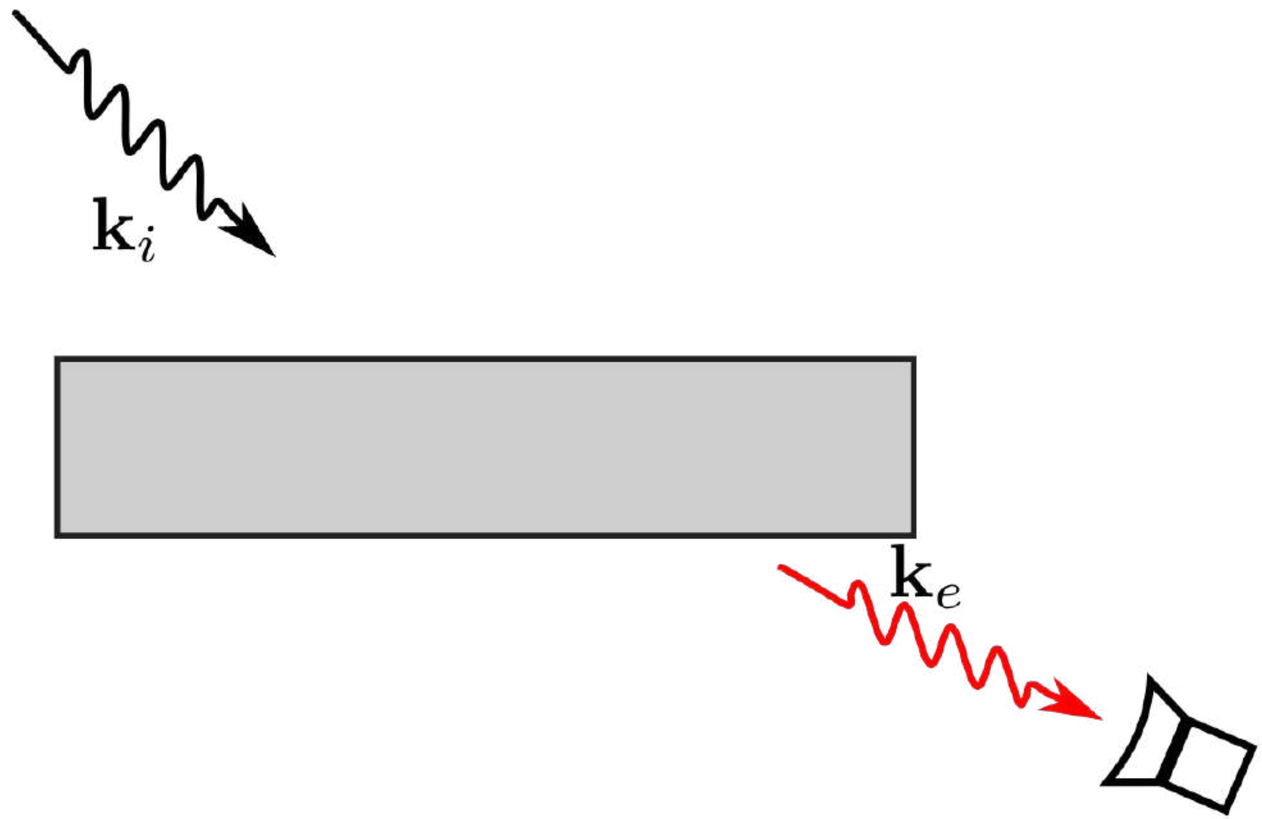
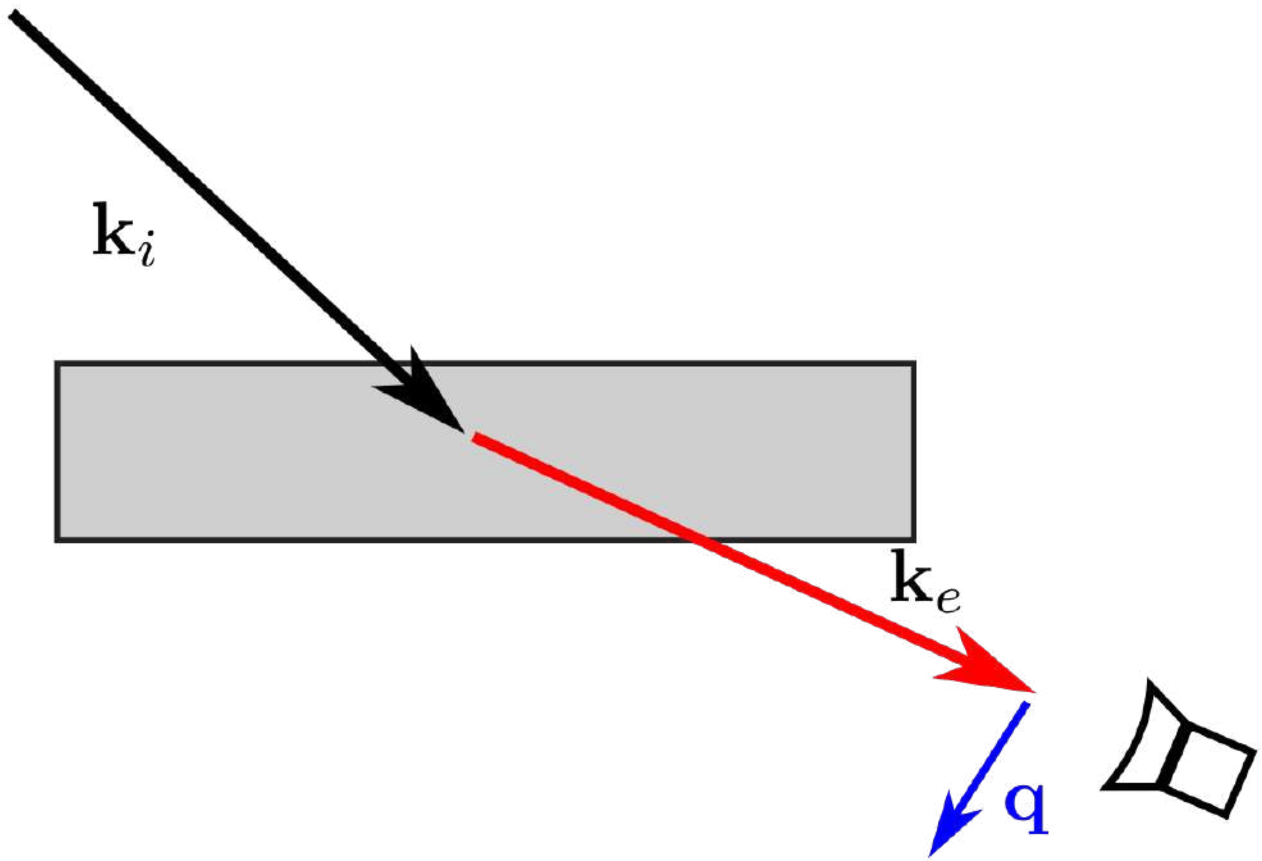
h BN



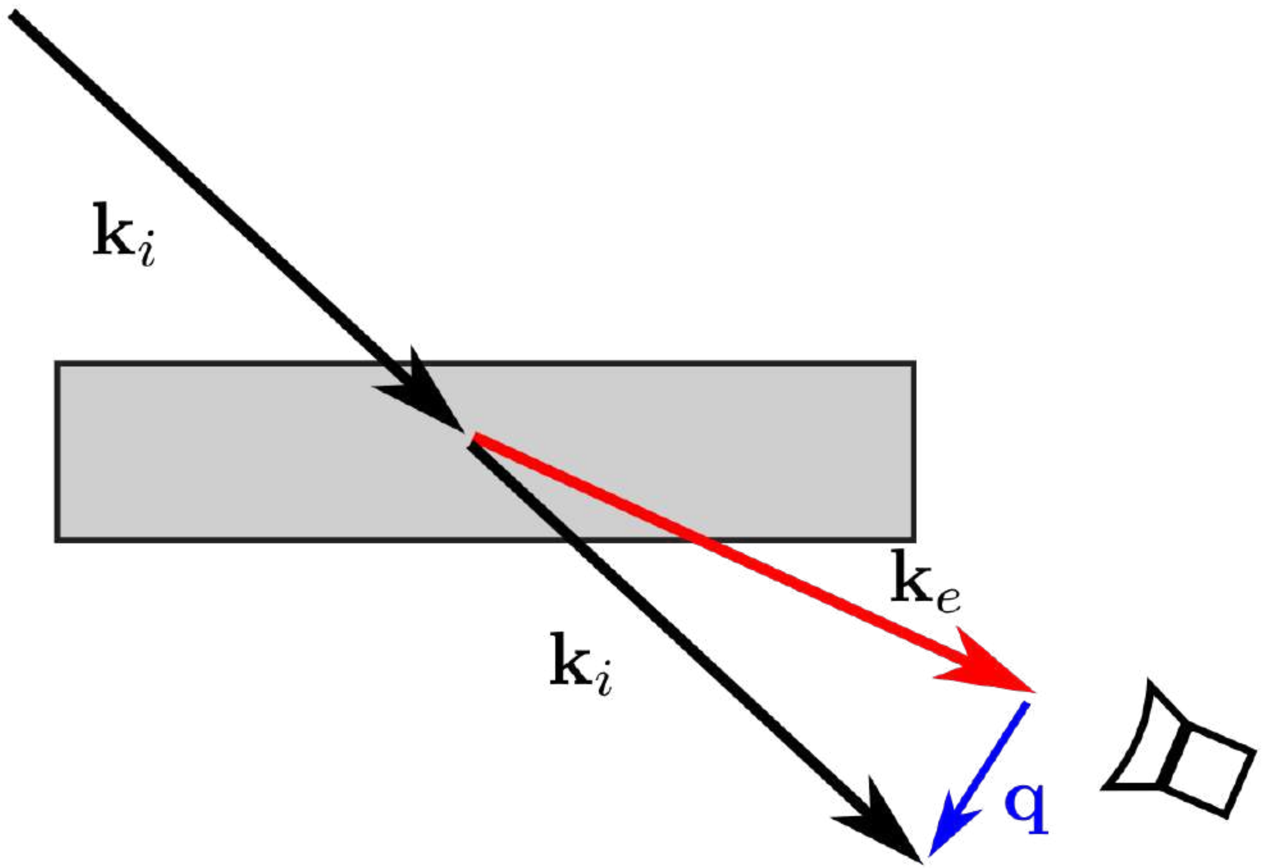
Fugallo *et al.* Phys. Rev. B **92**, 165122 (2015)

Galambosi *et al.* Phys. Rev. B **83**, 081413(R) (2011)

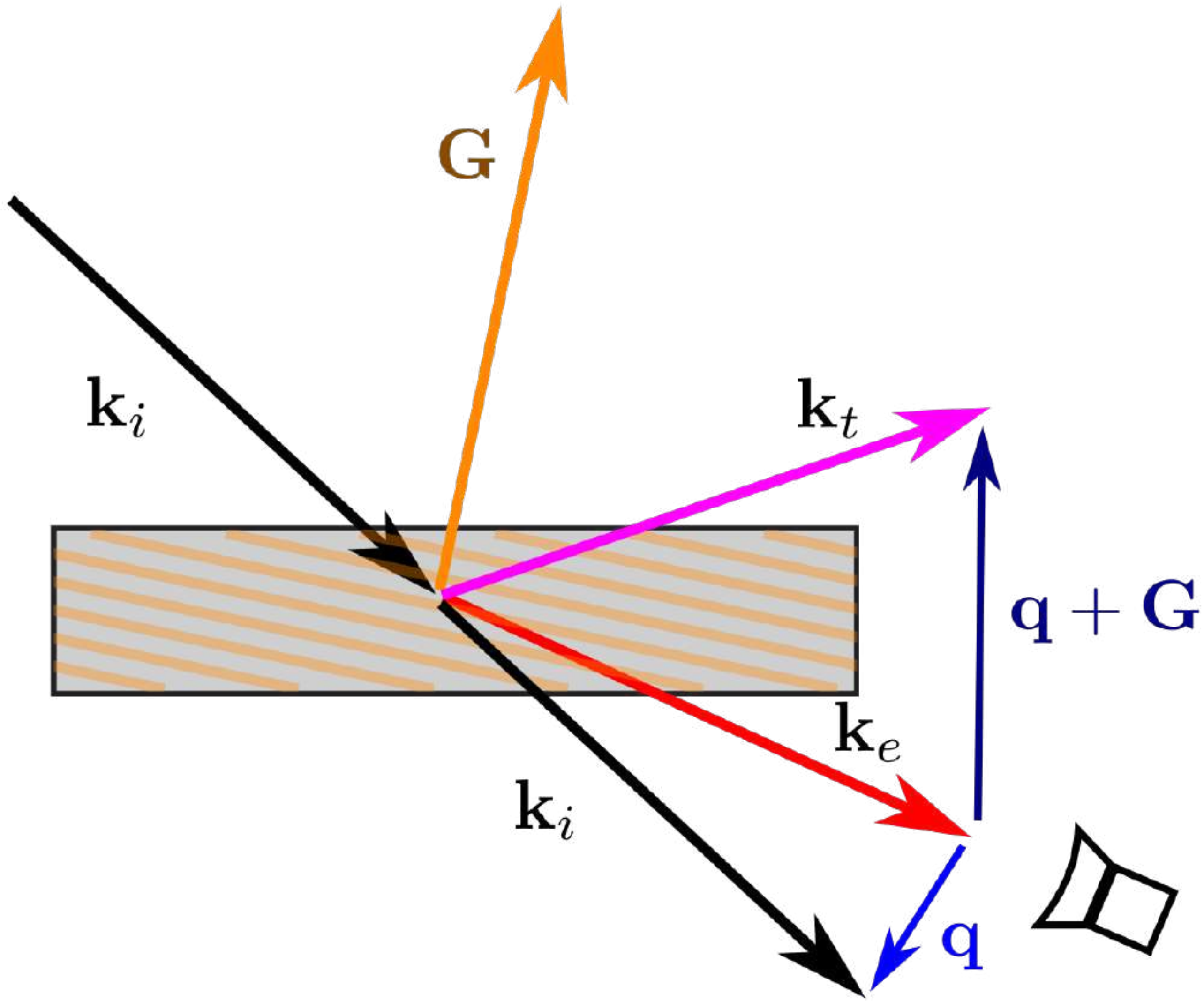

$\mathbf{k}_i$
$\mathbf{k}_e$



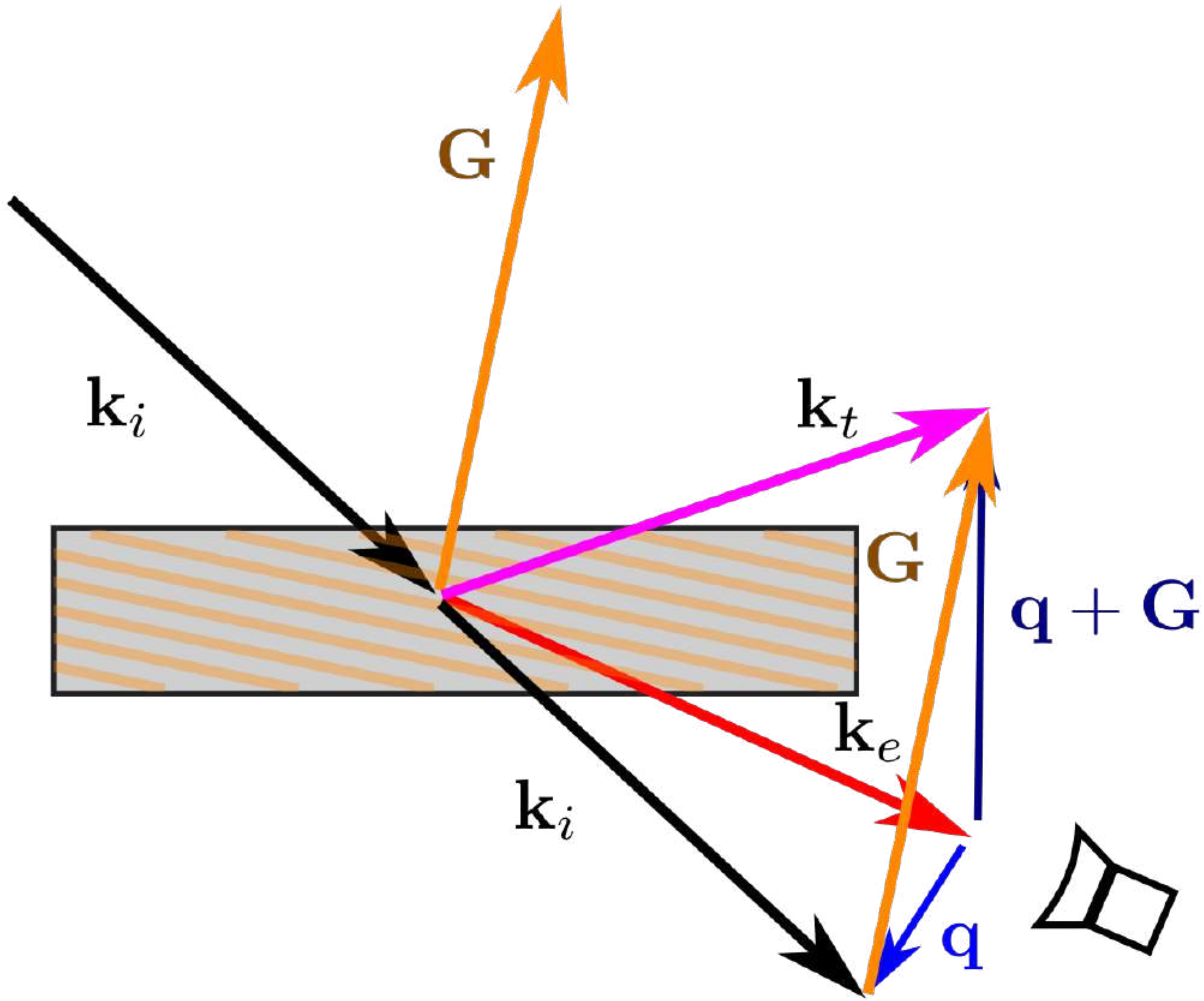
$\mathbf{k}_i$
$\mathbf{k}_e$
$\mathbf{q}$



$\mathbf{k}_i$
$\mathbf{k}_e$
$\mathbf{k}_i$
$\mathbf{q}$

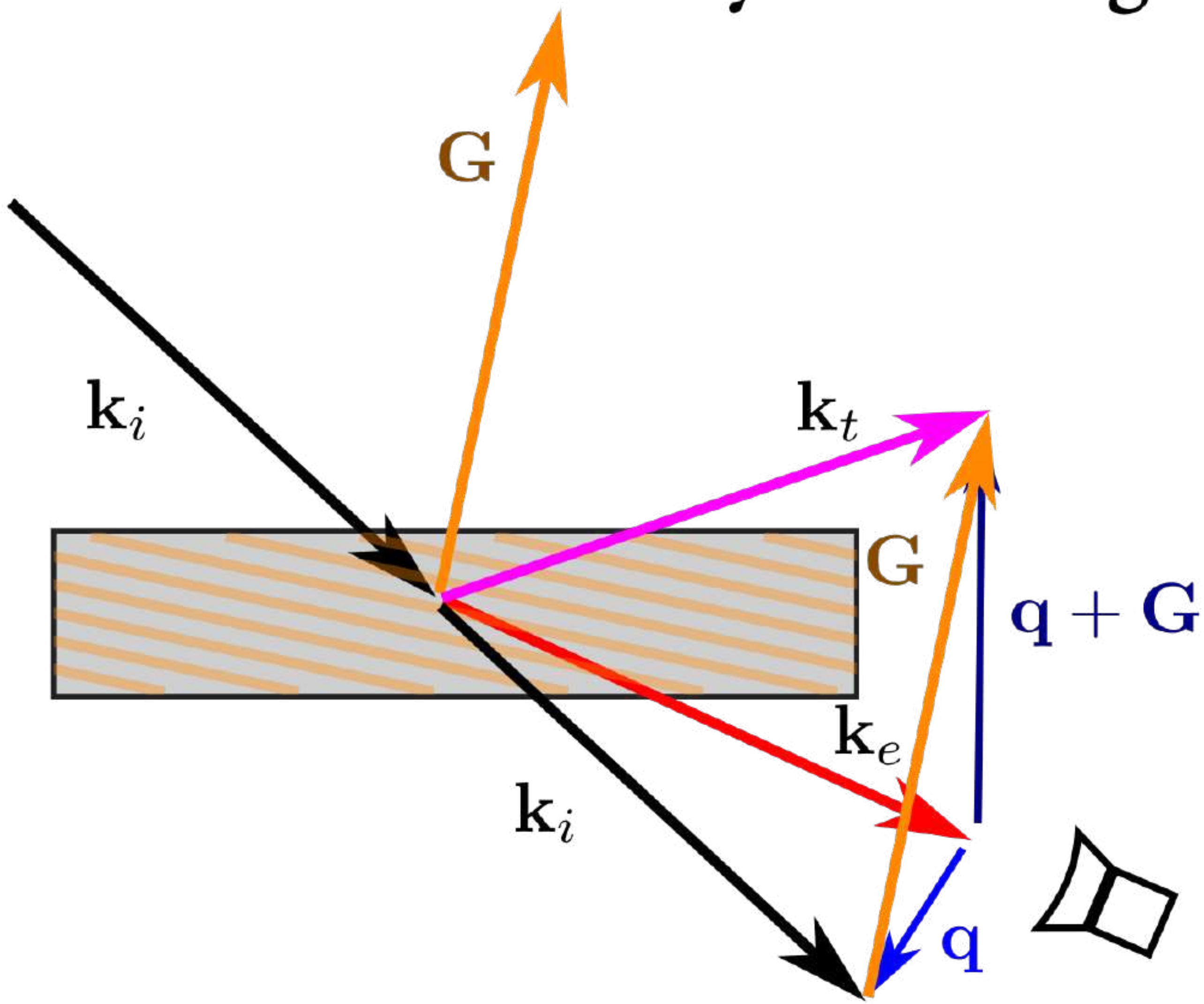


$\mathbf{G}$
$\mathbf{k}_i$
$\mathbf{k}_t$
$\mathbf{q} + \mathbf{G}$
$\mathbf{k}_e$
$\mathbf{k}_i$
$\mathbf{q}$



G
$\mathbf{k}_i$
$\mathbf{k}_t$
G
$\mathbf{q}+\mathbf{G}$
$\mathbf{k}_e$
$\mathbf{k}_i$
$\mathbf{q}$

Coherent Inelastic X-ray scattering



Coherent Inelastic X-ray scattering

$$\chi(\mathbf{q}, \mathbf{q} + \mathbf{G}, \omega) = \sum_{\lambda\lambda'} \frac{\sum_{vc} A_{\lambda}^{vc,\mathbf{q}} \langle c|e^{-i\mathbf{q}\cdot\mathbf{r}}|v\rangle \, S_{\lambda\lambda'}^{-1} \sum_{v'c'} A_{\lambda}^{*,v'c',\mathbf{q}} \langle v'|e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}|c'\rangle}{\omega - E_{\lambda}(\mathbf{q}) + i\eta}$$

Coherent Inelastic X-ray scattering

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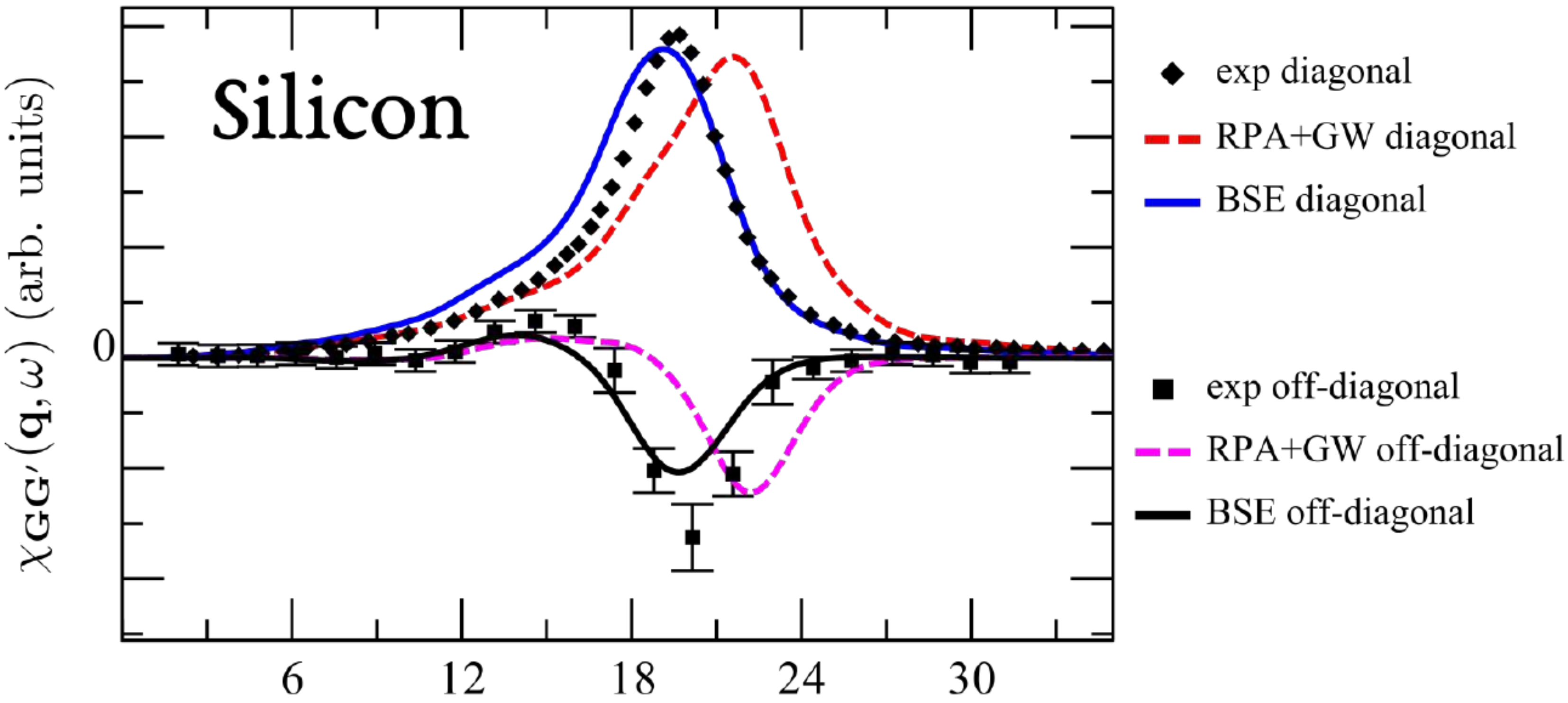
Igor Reshetnyak *et al.* Phys. Rev. Research **1**, 032010(R) (2019)



Schülke and Kaprolat, Phys. Rev. Lett. **67**, 879 (1991).

Weissker *et al.* Phys. Rev. B **81**, 085104 (2010).

Coherent Inelastic X-ray scattering

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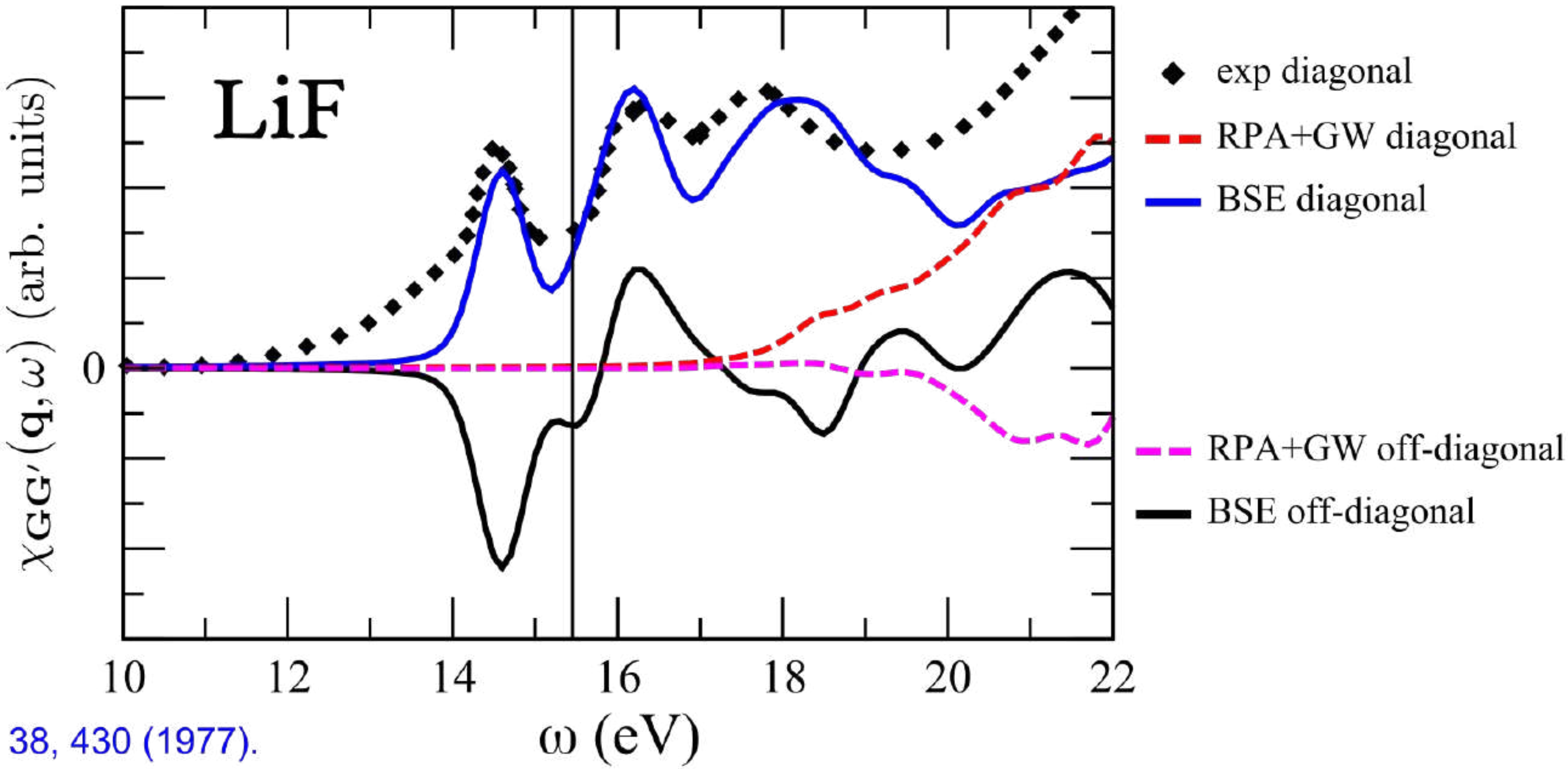

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Fields *et al.* Phys. Rev. Lett. 38, 430 (1977).

Coherent Inelastic X-ray scattering

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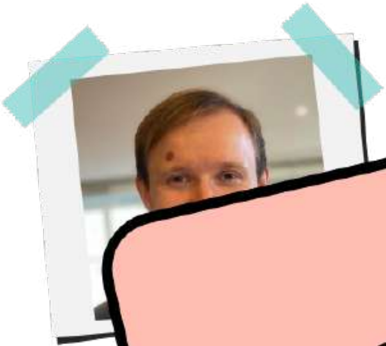

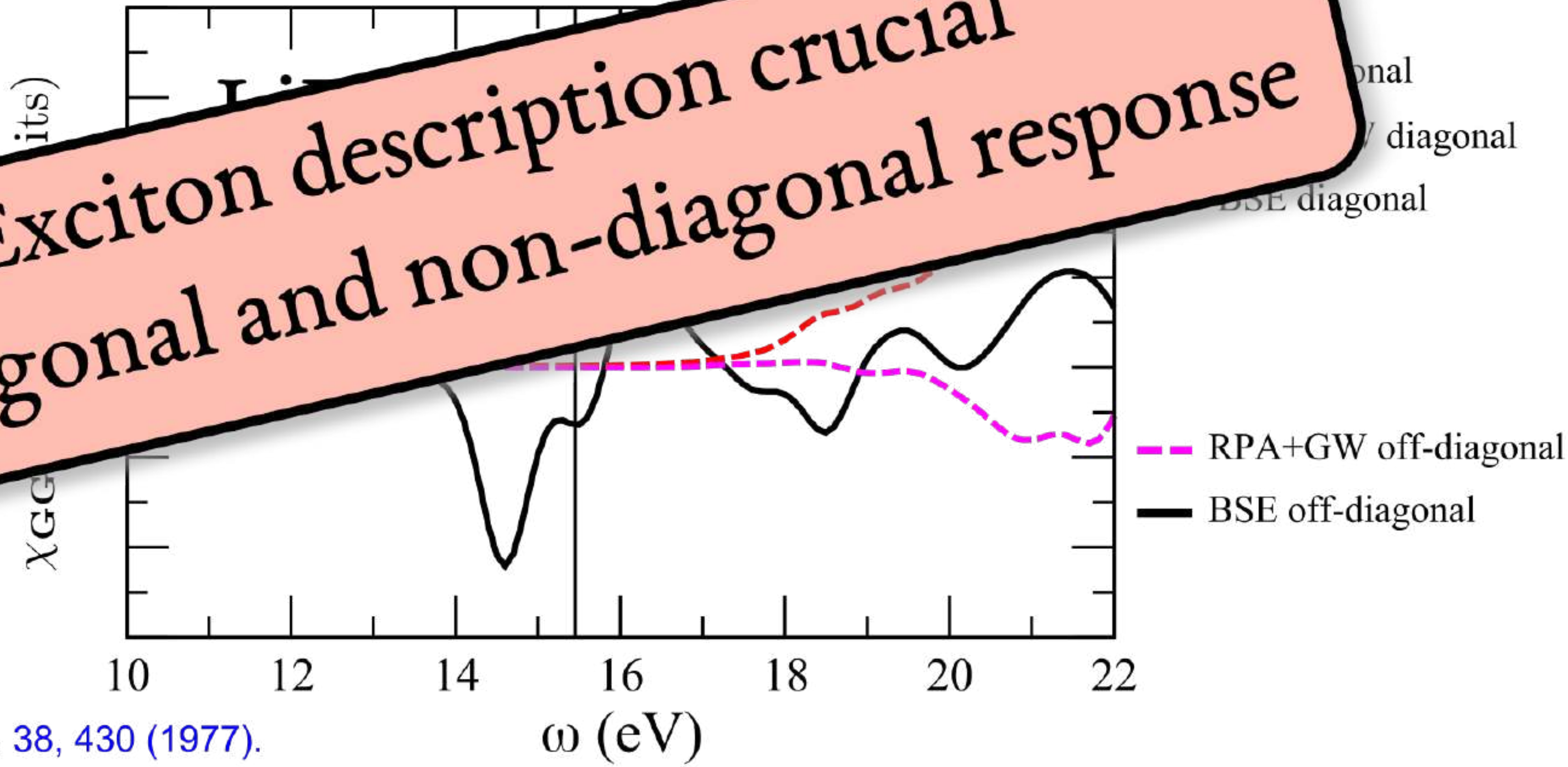

Exciton description crucial for diagonal and non-diagonal response

Igor Re… Phys. R… 032010(…

Fields *et al.* Phys. Rev. Lett. 38, 430 (1977).

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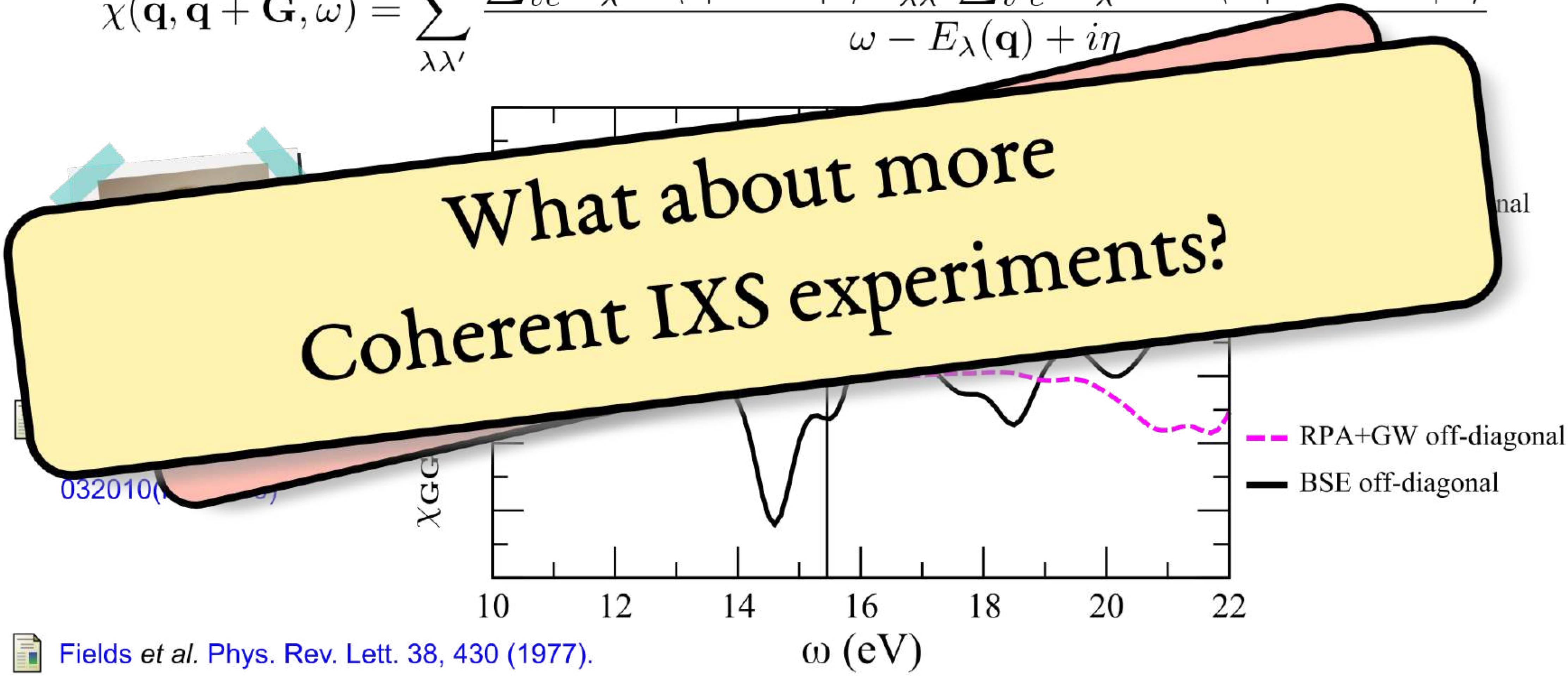
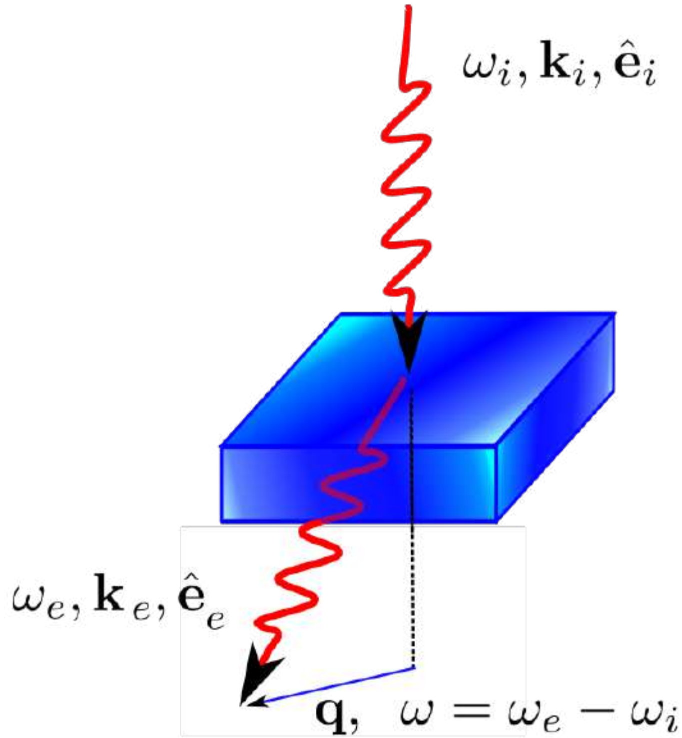


Fields *et al.* Phys. Rev. Lett. 38, 430 (1977).

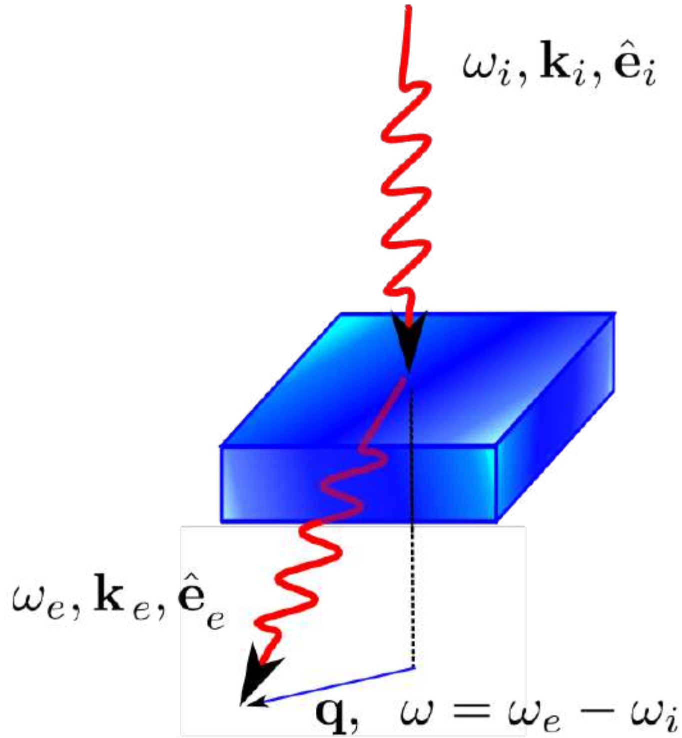
X-ray scattering



non-Resonant IXS

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2 \mathrm{d}\omega_e} \propto \sum_f \left| \langle f|e^{i\mathbf{q}\cdot\mathbf{r}}|0\rangle + \frac{\langle f|e^{-i\mathbf{k}_f\cdot\mathbf{r}}\nabla|n\rangle \; \langle n|e^{i\mathbf{k}_i\cdot\mathbf{r}}\nabla|0\rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta\left(\omega - (E_f - E_0)\right)$$

X-ray scattering



non-Resonant IXS

Resonant IXS

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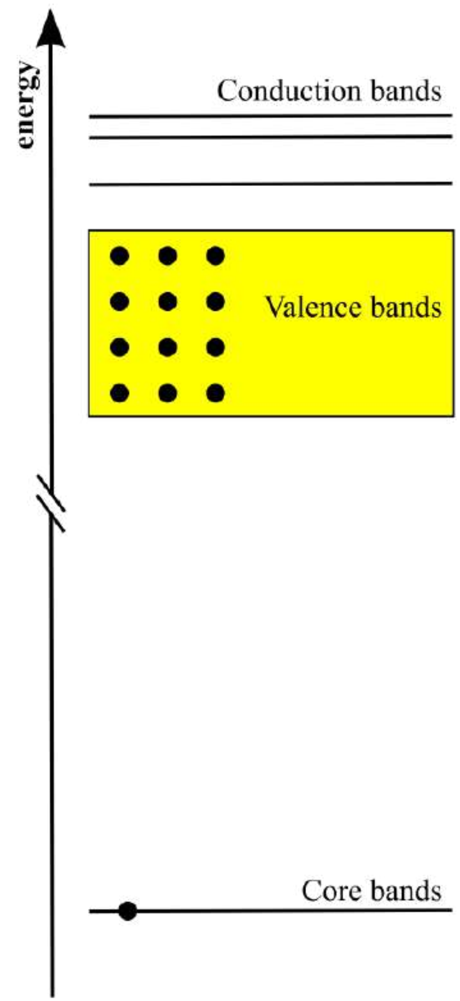
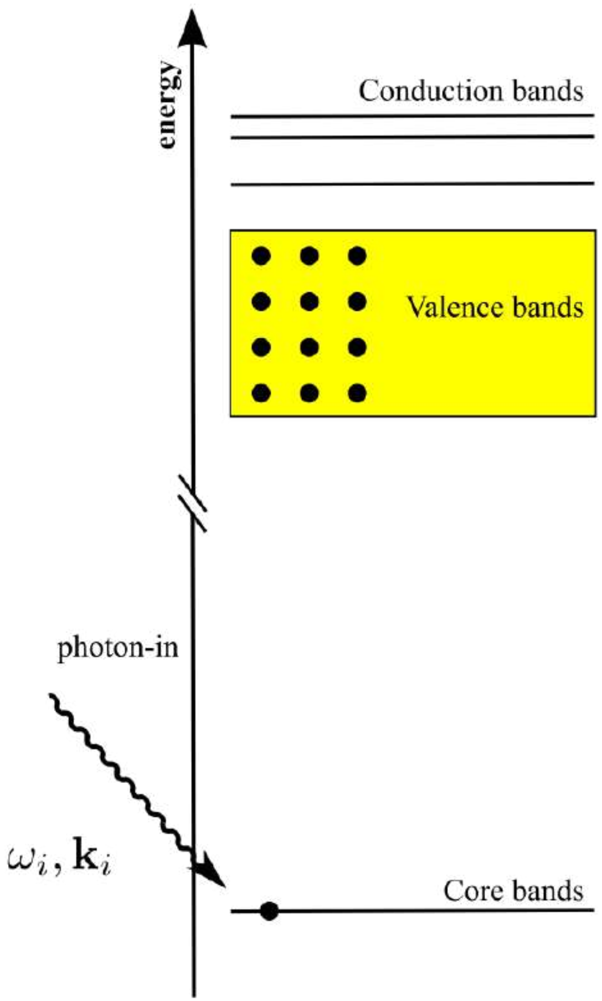
Resonant IXS

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2 \mathrm{d}\omega_e} \propto \sum_f \left| \sum_n \frac{\langle f | e^{-i\mathbf{k}_f \cdot \mathbf{r}} \nabla | n \rangle \ \langle n | e^{i\mathbf{k}_i \cdot \mathbf{r}} \nabla | 0 \rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta \left(\omega - (E_f - E_0) \right)$$

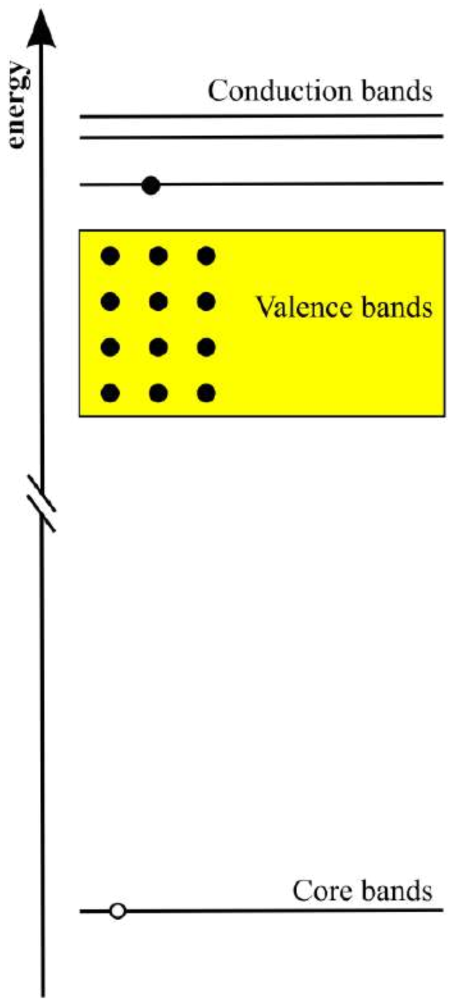
Initial state

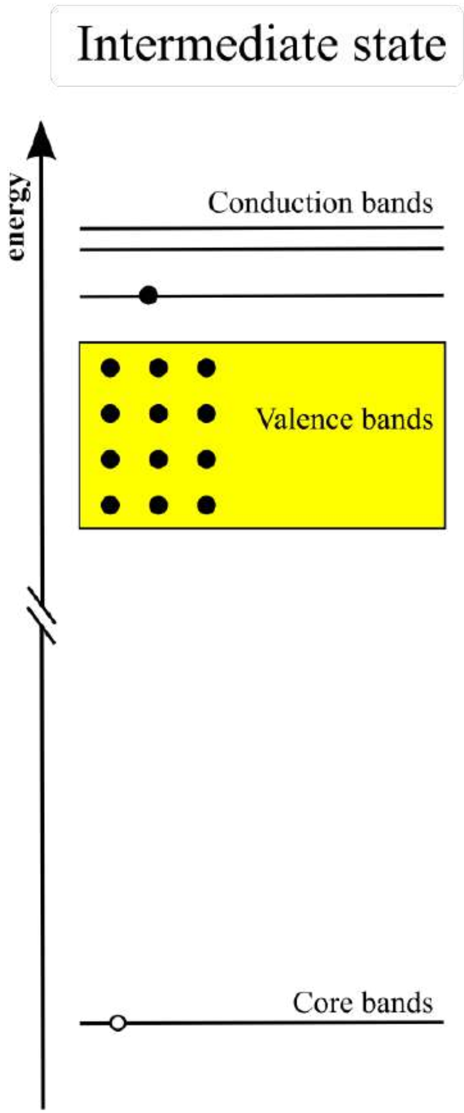


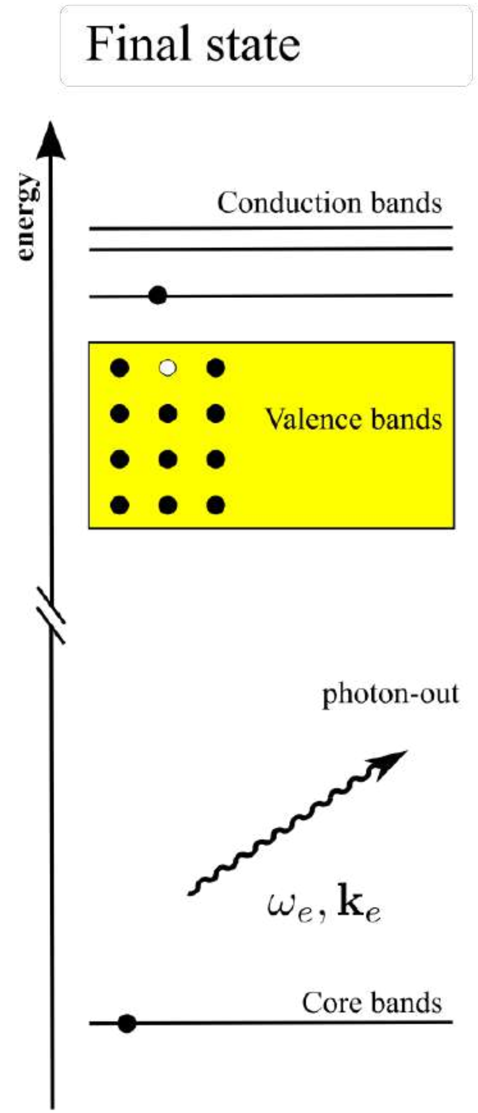
Initial state



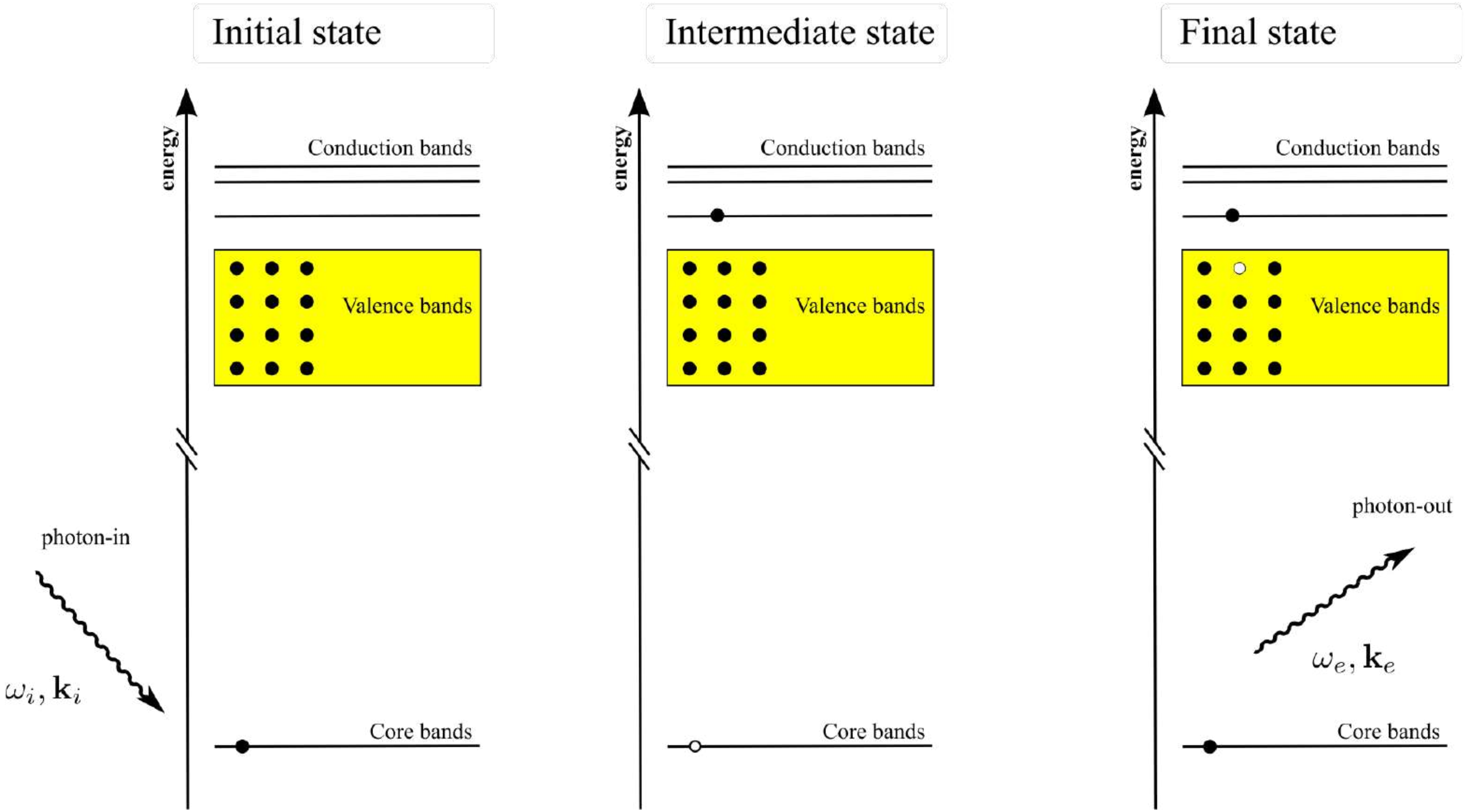
Initial state




Intermediate state
energy
Conduction bands
Valence bands
Core bands



Final state
energy
Conduction bands
Valence bands
photon-out
$\omega_e, \mathbf{k}_e$
Core bands



Initial state
energy
Conduction bands
Valence bands
photon-in
$\omega_i, \mathbf{k}_i$
Core bands
Intermediate state
energy
Conduction bands
Valence bands
Core bands
Final state
energy
Conduction bands
Valence bands
photon-out
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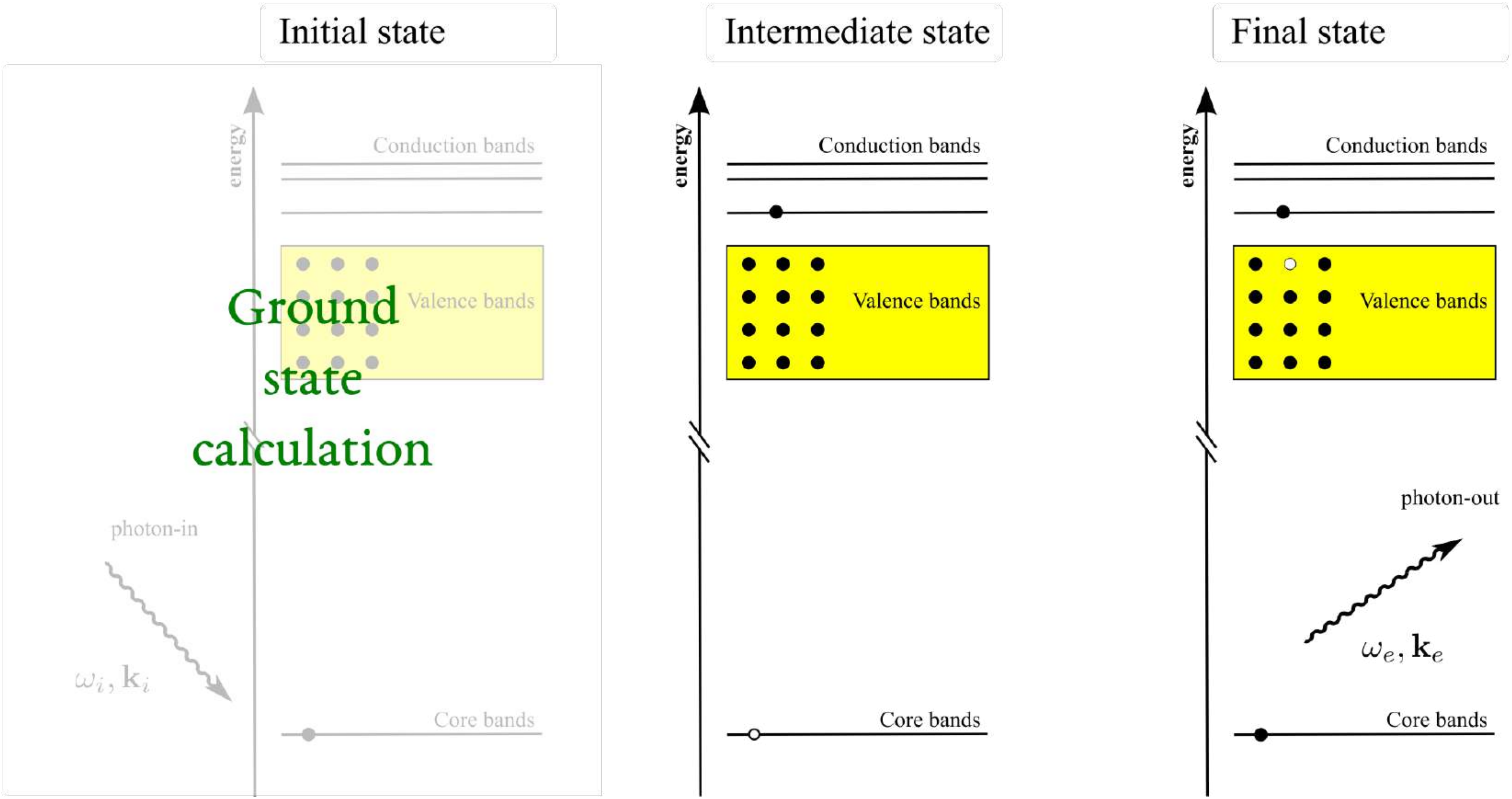
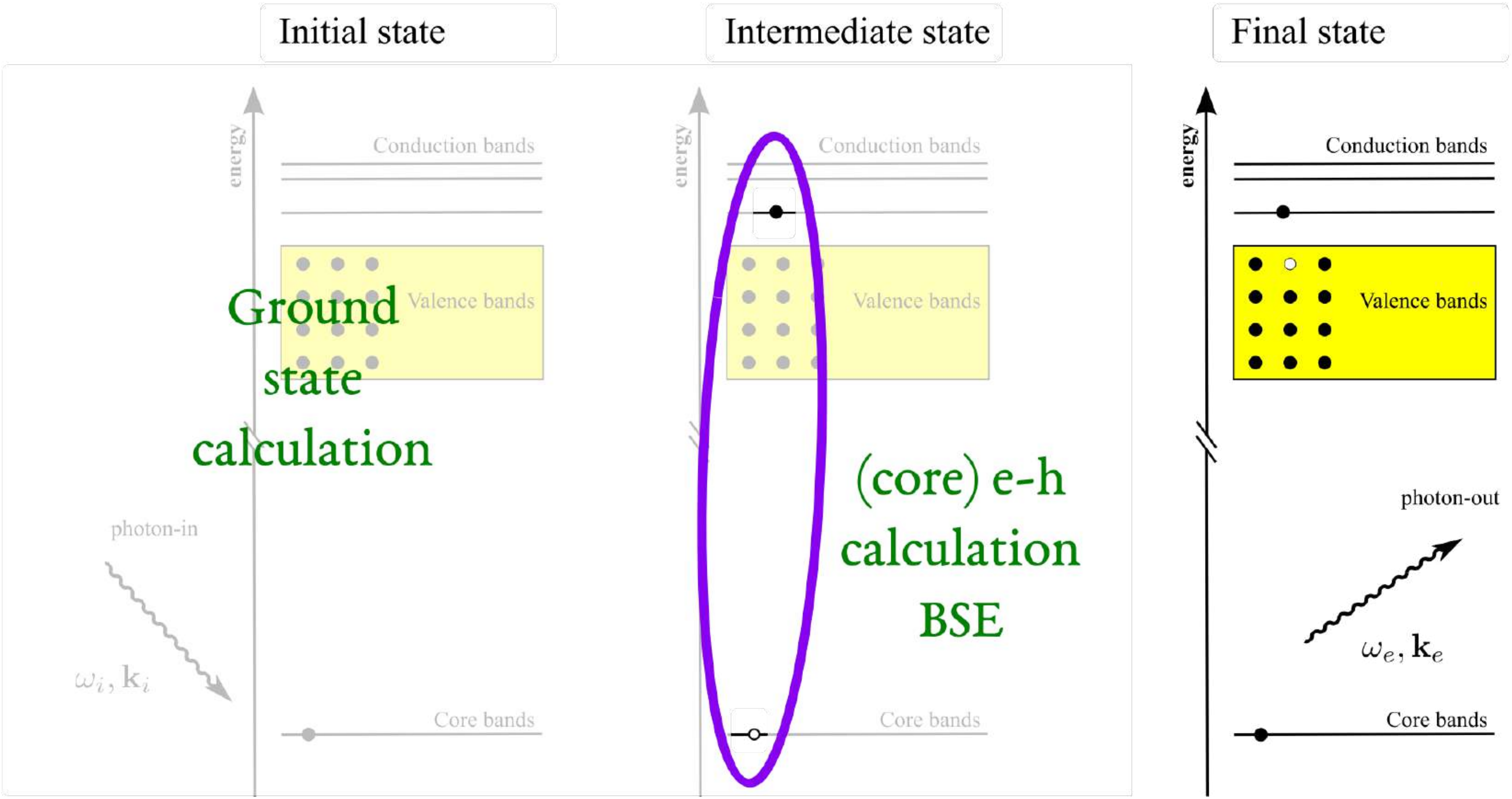
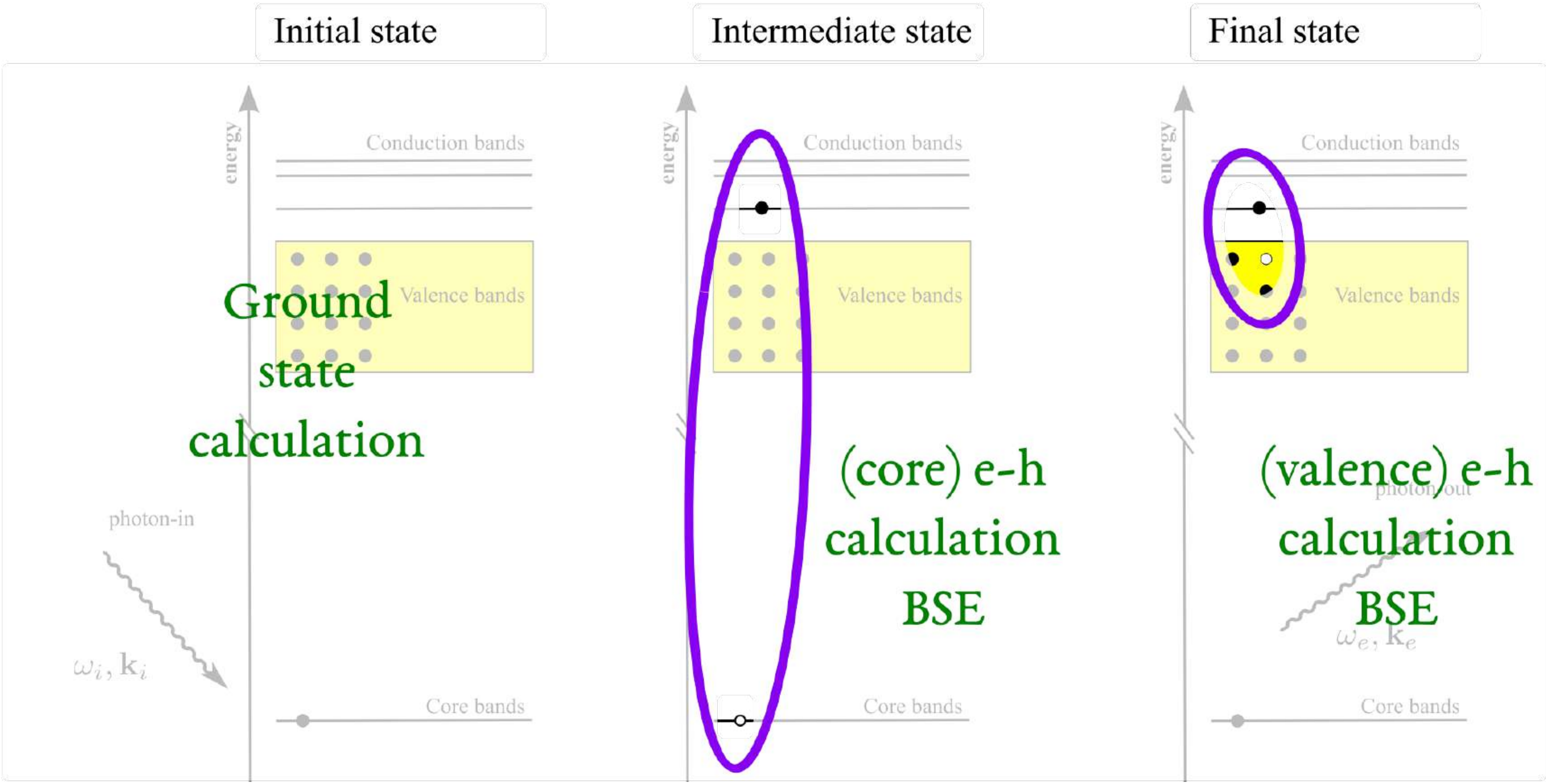
Core bands



Initial state
Intermediate state
Final state
energy
Conduction bands
Valence bands
Ground state calculation
photon-in
$\omega_i, \mathbf{k}_i$
Core bands
energy
Conduction bands
Valence bands
Core bands
energy
Conduction bands
Valence bands
photon-out
$\omega_e, \mathbf{k}_e$
Core bands



Initial state
Intermediate state
Final state
energy
Conduction bands
Valence bands
Core bands
Ground state calculation
photon-in
$\omega_i, \mathbf{k}_i$
energy
Conduction bands
Valence bands
Core bands
(core) e-h calculation BSE
energy
Conduction bands
Valence bands
Core bands
photon-out
$\omega_e, \mathbf{k}_e$



Initial state
Intermediate state
Final state
energy
Conduction bands
Valence bands
Core bands
photon-in
$\omega_i, \mathbf{k}_i$
photon-out
$\omega_e, \mathbf{k}_e$
Ground state calculation
(core) e-h calculation BSE
(valence) e-h calculation BSE

Resonant Inelastic X-ray scattering
in BSE

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2\mathrm{d}\omega_e} \propto \sum_f \left| \sum_n \frac{\langle f|e^{-i\mathbf{k}_f\cdot\mathbf{r}}\nabla|n\rangle\ \langle n|e^{i\mathbf{k}_i\cdot\mathbf{r}}\nabla|0\rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta\left(\omega - (E_f - E_0)\right)$$

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

Vinson *et al.*, Phys. Rev. B **94**, 035163 (2016)

Geondzhian and Gilmore, Phys. Rev. B **98**, 214305 (2018)

Shirley, Phys. Rev. Lett. **80**, 794 (1998)

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Vinson *et al.*, Phys. Rev. B **94**, 035163 (2016)

Geondzhian and Gilmore, Phys. Rev. B **98**, 214305 (2018)

Shirley, Phys. Rev. Lett. **80**, 794 (1998)

Resonant Inelastic X-ray scattering

via excitonic pathways in BSE

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2\mathrm{d}\omega_e} \propto \sum_f \left| \sum_n \frac{\langle f|e^{-i\mathbf{k}_f\cdot\mathbf{r}}\nabla|n\rangle\ \langle n|e^{i\mathbf{k}_i\cdot\mathbf{r}}\nabla|0\rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta\left(\omega - (E_f - E_0)\right)$$

$$H^{\mathrm{BSE}} A^{vc}_{\lambda_o} = E_{\lambda_o} A^{vc}_{\lambda_o}$$

$$H^{\mathrm{BSE}} A^{\mu c}_{\lambda_c} = E_{\lambda_c} A^{\mu c}_{\lambda_c}$$

$$t^{(1)}_{\lambda_c} = \sum_{\mu c} A^{\mu c}_{\lambda_c} \tilde{\rho}_{\mu c}$$

$$t^{(2)}_{\lambda_c \lambda_o} = \sum_{\mu v c} A^{*,vc}_{\lambda_o} \tilde{\rho}_{\mu v} A^{\mu c}_{\lambda_c}$$

Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)

Resonant Inelastic X-ray scattering

via excitonic pathways in BSE

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2\mathrm{d}\omega_e} \propto \sum_f \left| \sum_n \frac{\langle f|e^{-i\mathbf{k}_f\cdot\mathbf{r}}\nabla|n\rangle\ \langle n|e^{i\mathbf{k}_i\cdot\mathbf{r}}\nabla|0\rangle}{\omega_i - (E_n - E_0)} \right|^2 \times \delta\left(\omega - (E_f - E_0)\right)$$

$$\downarrow$$

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega_2\mathrm{d}\omega_e} \propto \mathrm{Im} \sum_{\lambda_o} \frac{\left| \sum_{\lambda_c} \frac{t^{(2)}_{\lambda_c\lambda_o} t^{(1)}_{\lambda_c}}{E_{\lambda_c} - \omega_i - i\eta} \right|^2}{E_{\lambda_o} - (\omega_i - \omega_e) - i\eta}$$

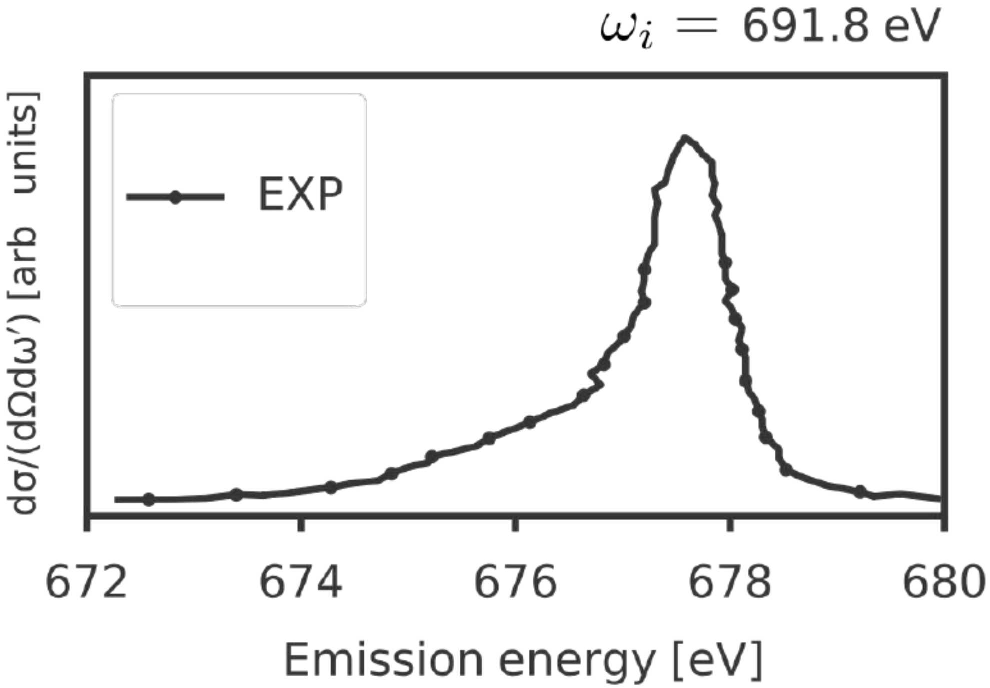
$$t^{(1)}_{\lambda_c} = \sum_{\mu c} A^{\mu c}_{\lambda_c} \tilde{\rho}_{\mu c}$$

$$t^{(2)}_{\lambda_c\lambda_o} = \sum_{\mu v c} A^{*,vc}_{\lambda_o} \tilde{\rho}_{\mu v} A^{\mu c}_{\lambda_c}$$

Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)

Resonant Inelastic X-ray scattering

via excitonic pathways in BSE



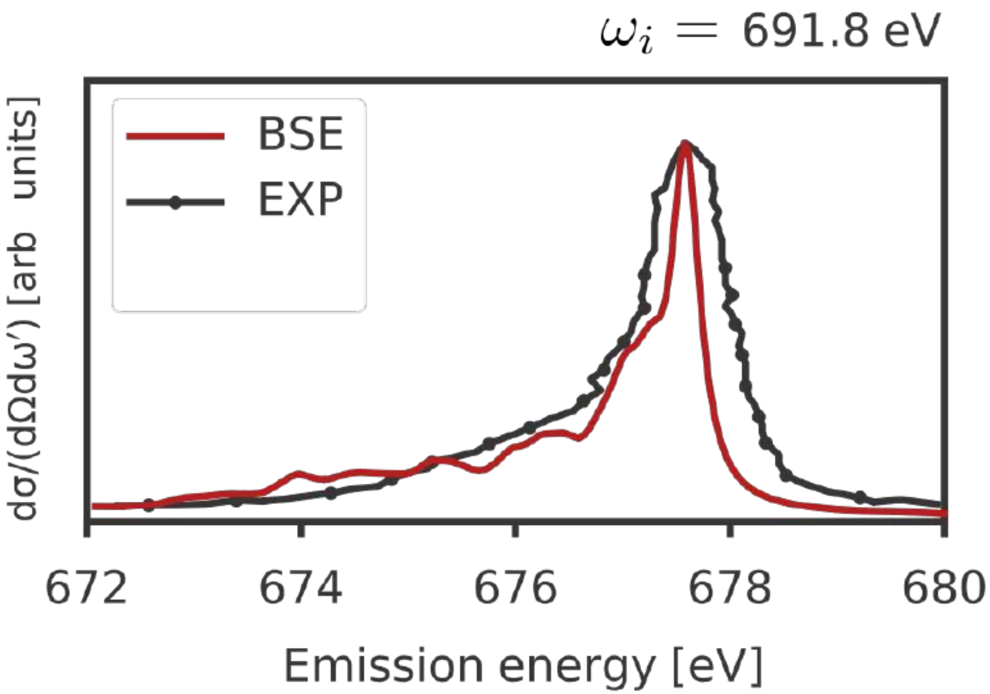
LiF

Kikas *et al.*, Phys. Rev. B **70**, 085102 (2004)

Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)

Resonant Inelastic X-ray scattering

via excitonic pathways in BSE



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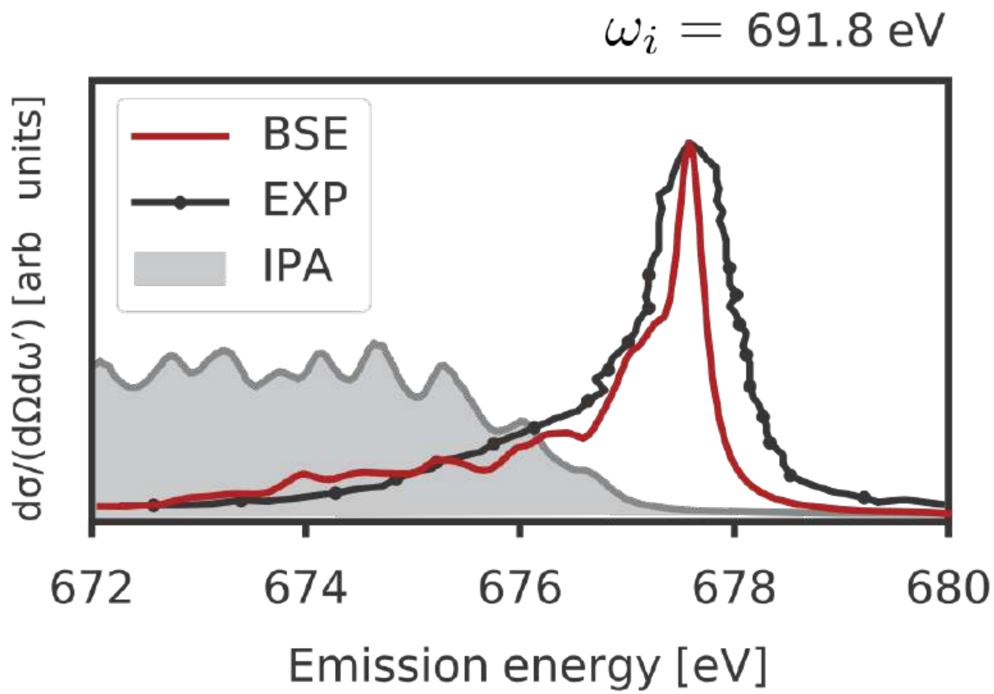
Resonant Inelastic X-ray scattering
via excitonic pathways in BSE



LiF

Kikas *et al.*, Phys. Rev. B **70**, 085102 (2004)

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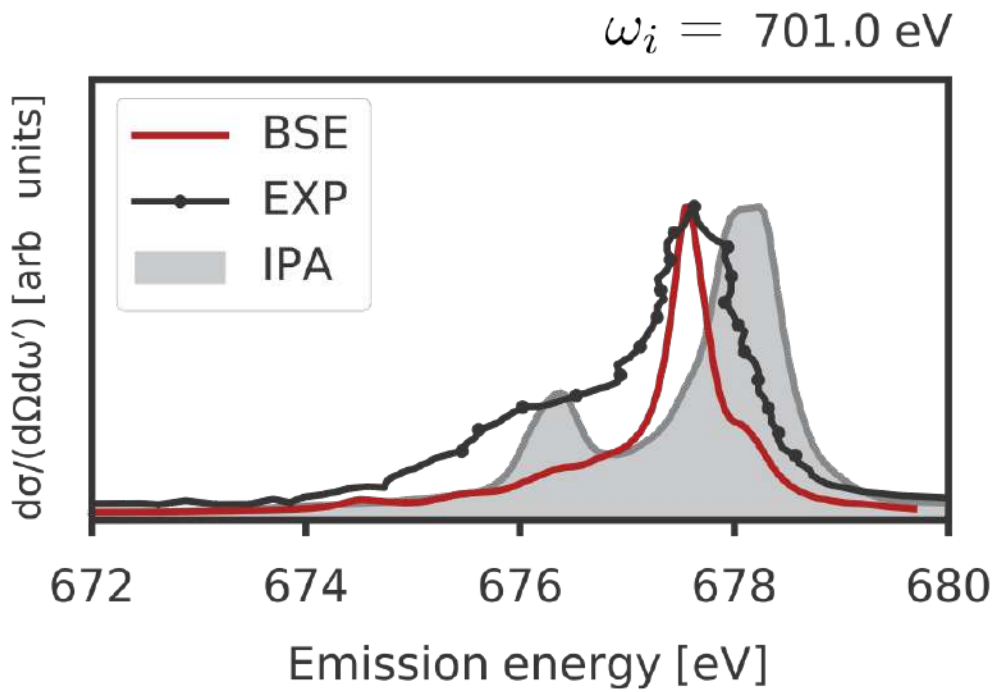

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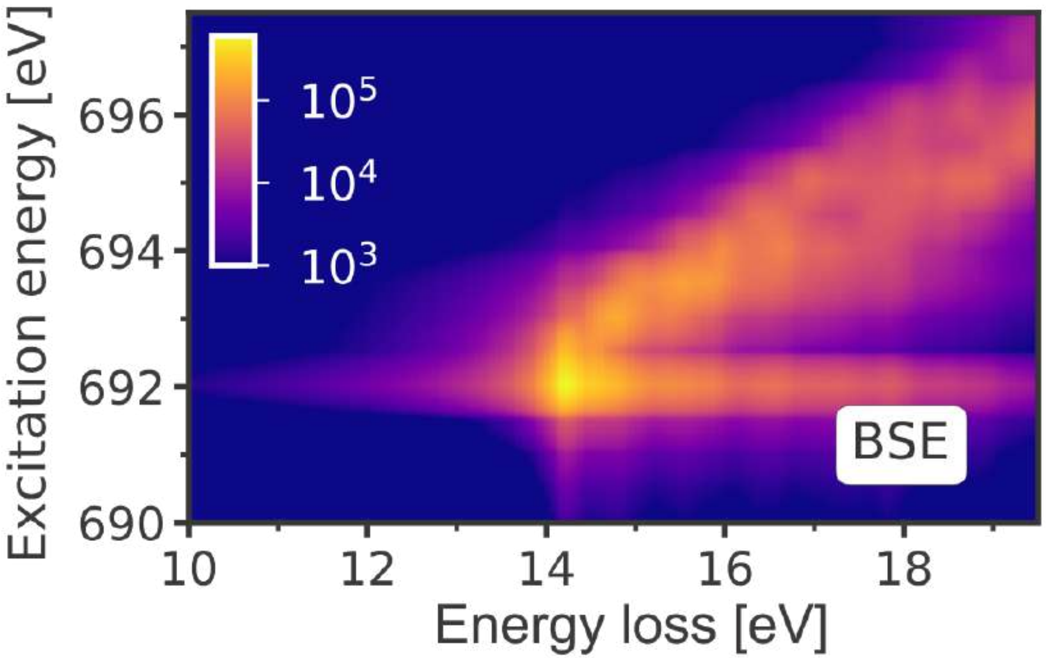
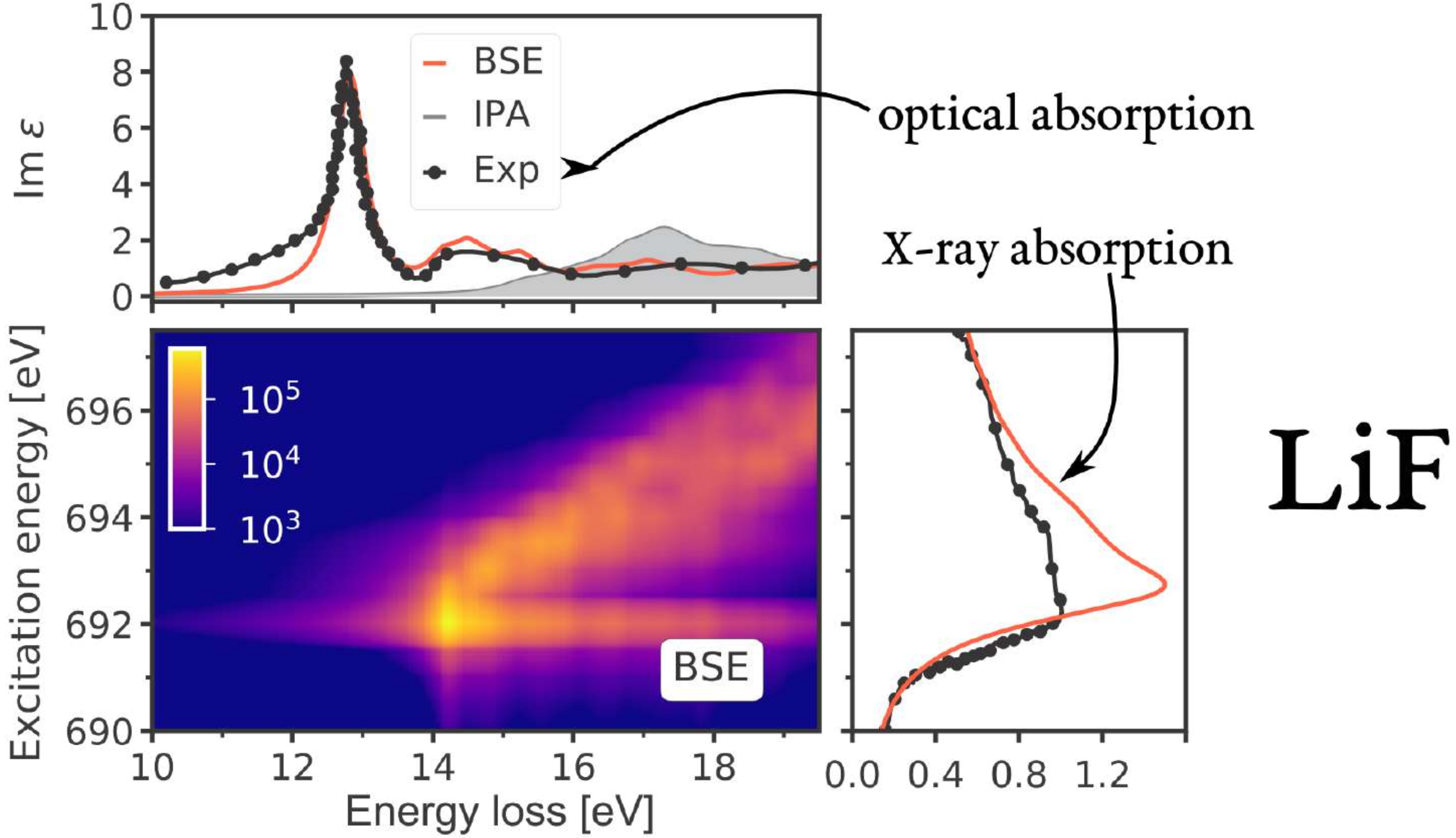
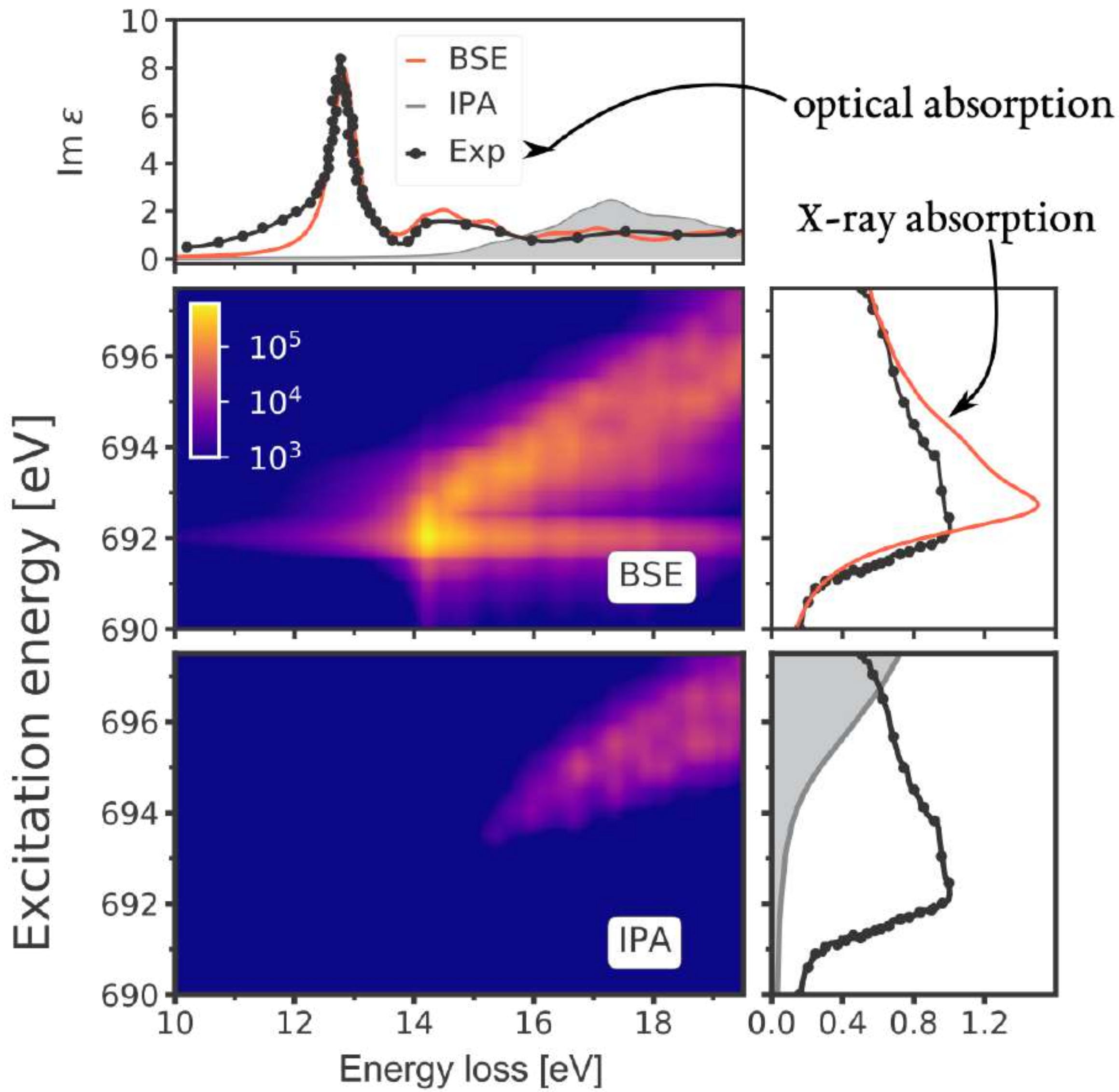


LiF

Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)



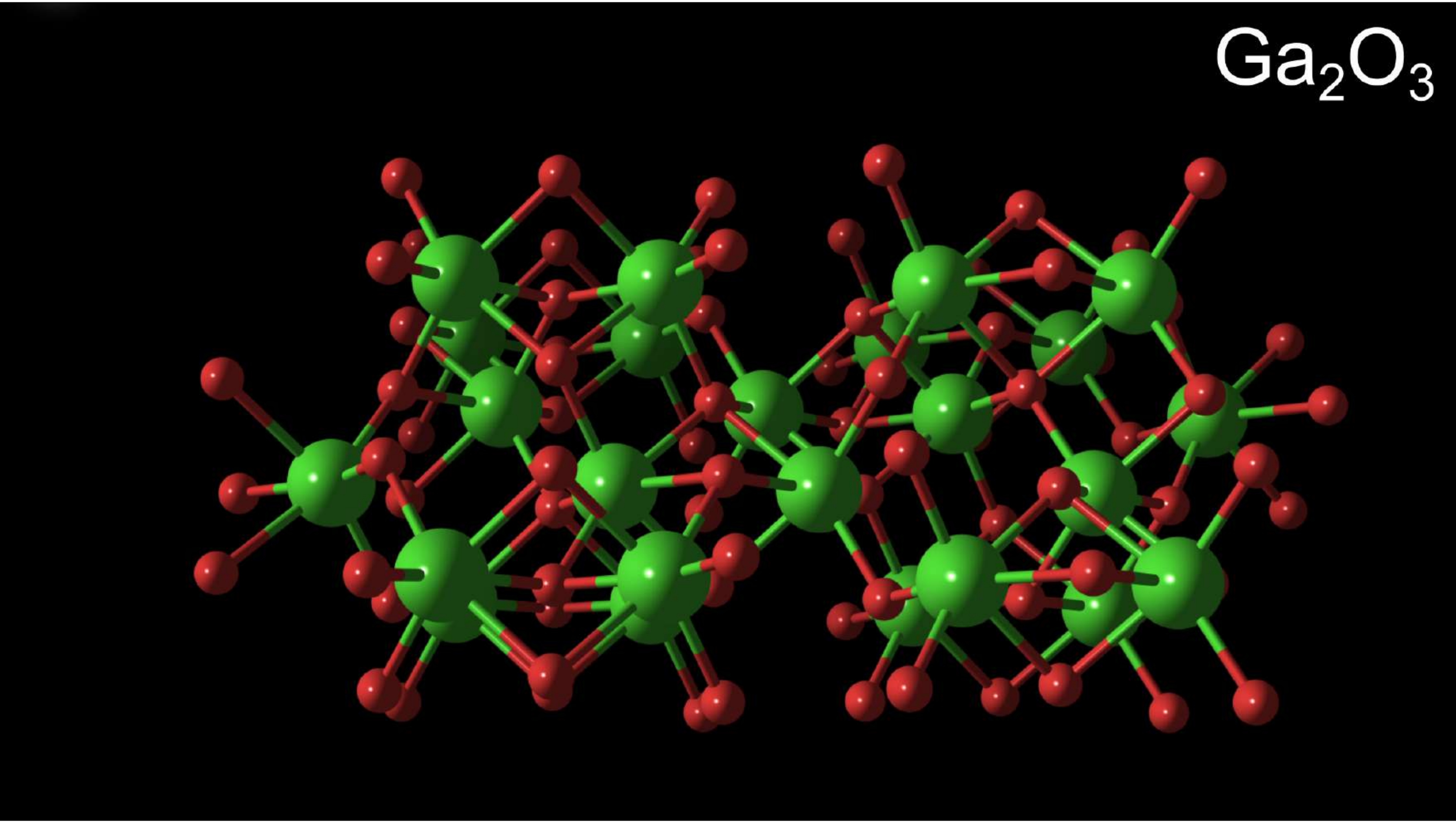
Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)

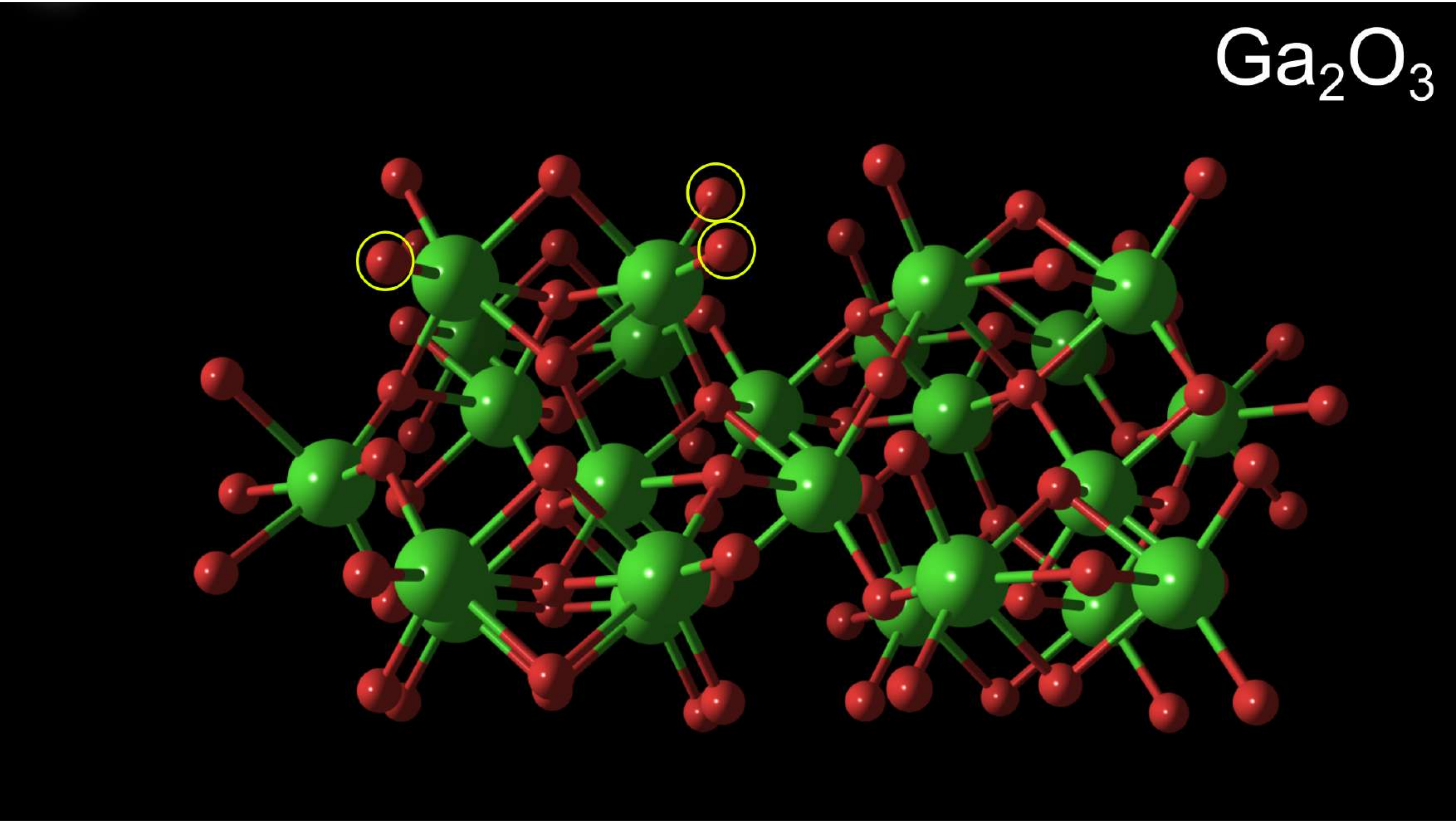


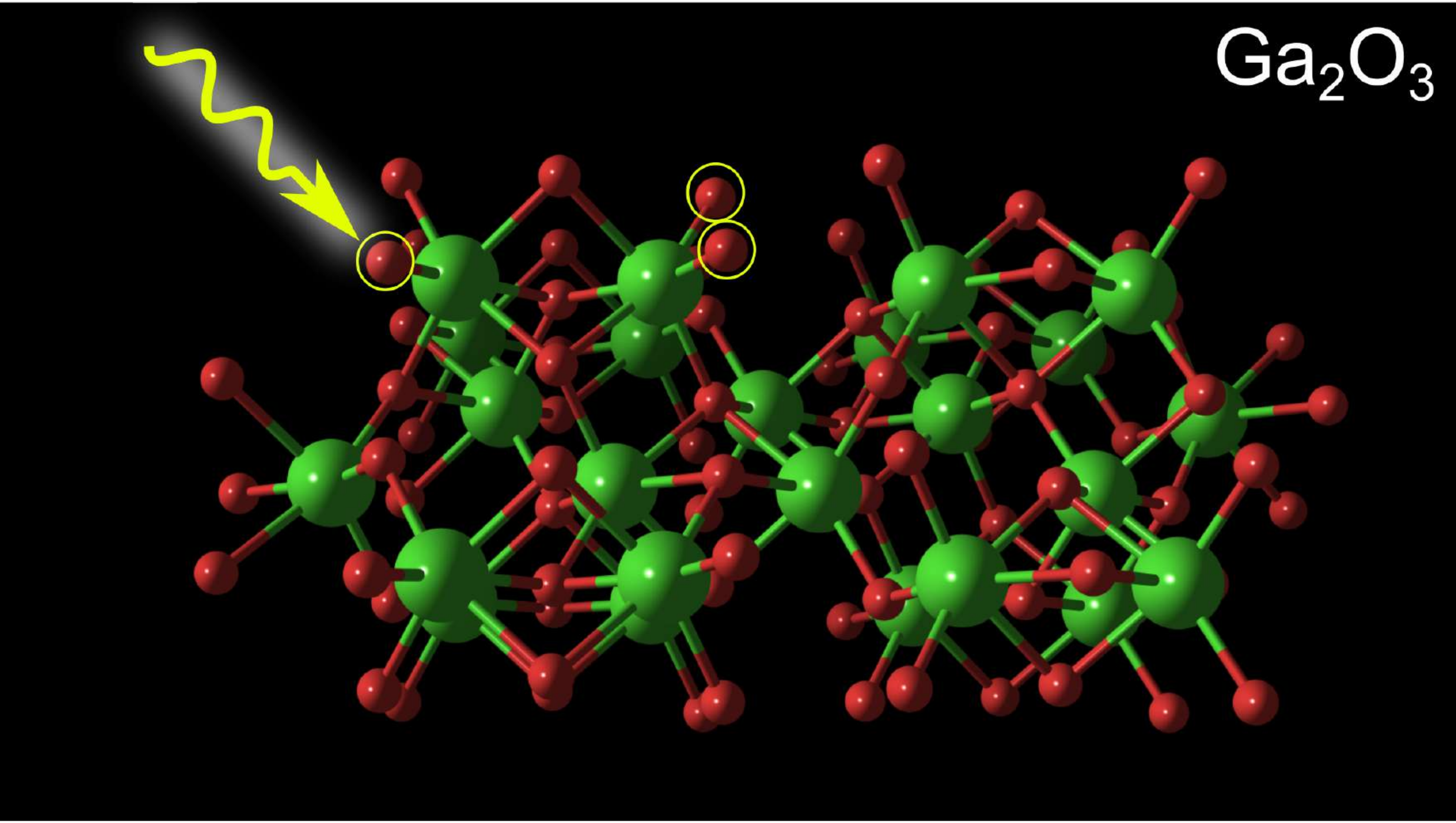
LiF

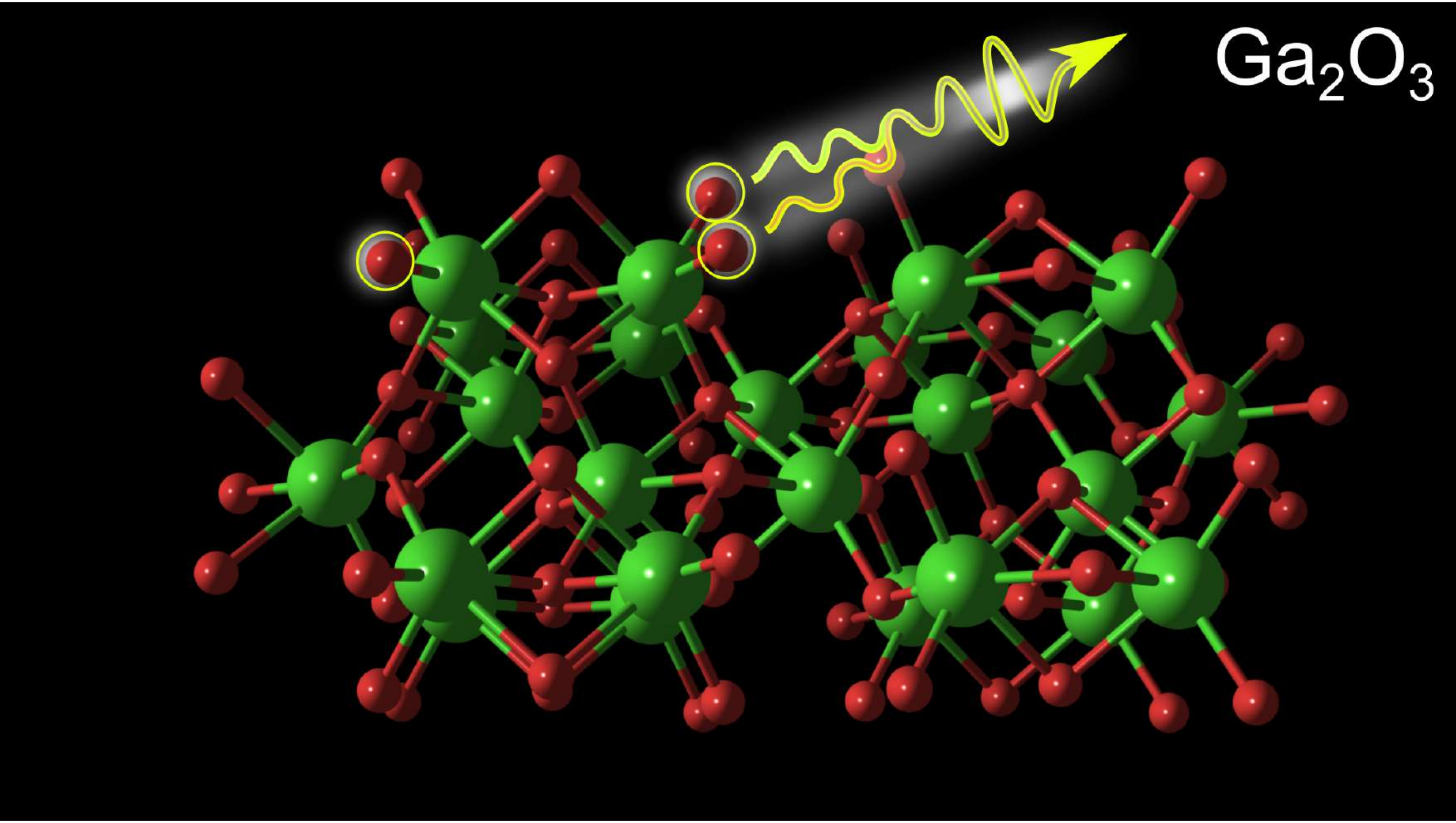
Vorwerk, Sottile, Draxl, Phys. Rev. Research **2**, 042003(R) (2020)

- Green's functions approach to spectra
- Excitonic effects in (non-)resonant, coherent IXS
- Coherence in RIXS

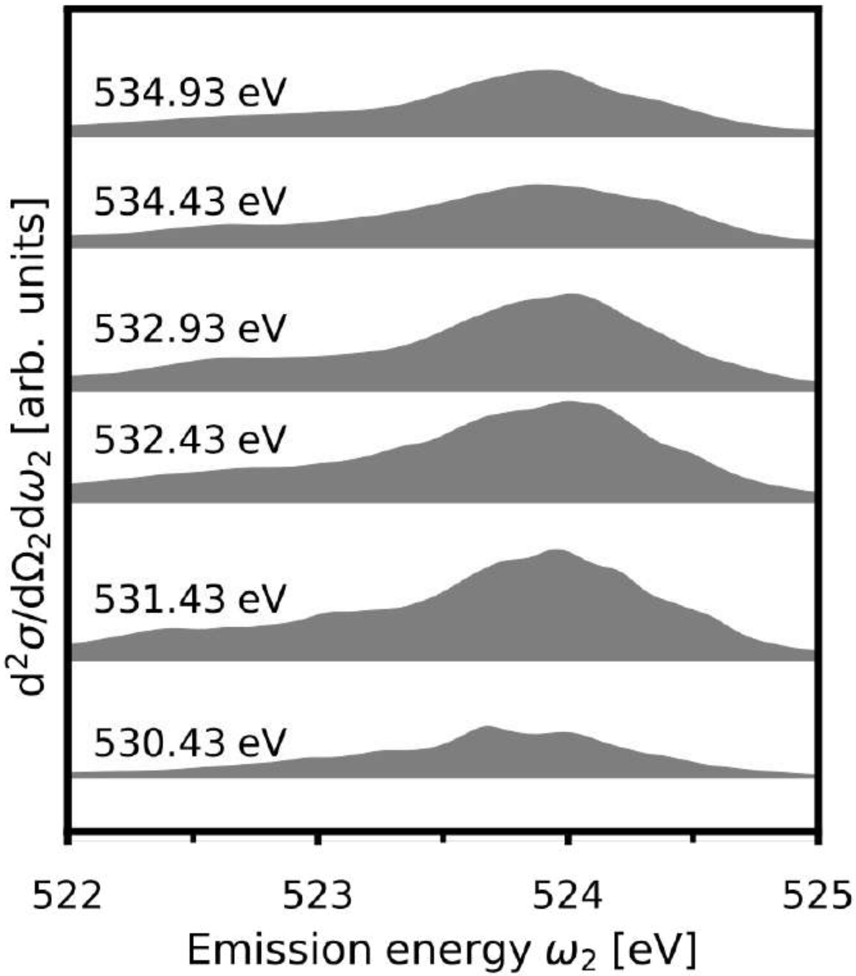

Ga_2O_3


Ga_2O_3

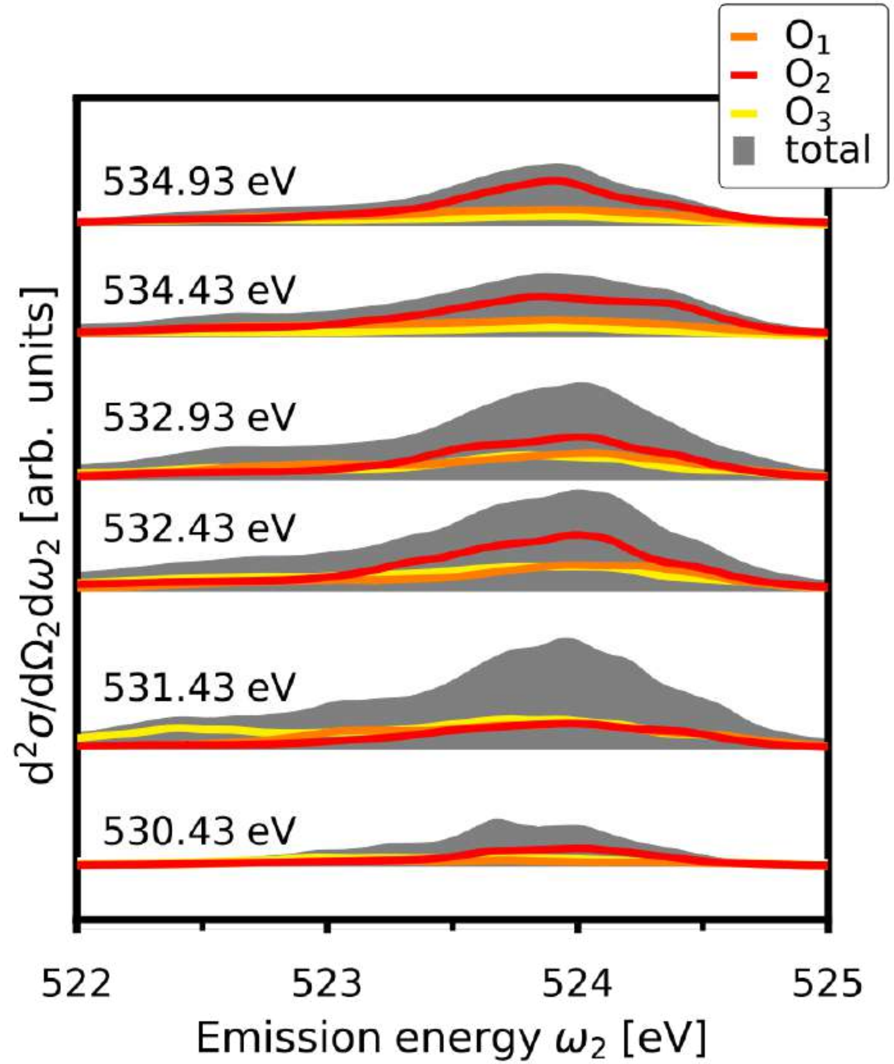

Ga_2O_3


Ga_2O_3

O-K Ga_2O_3

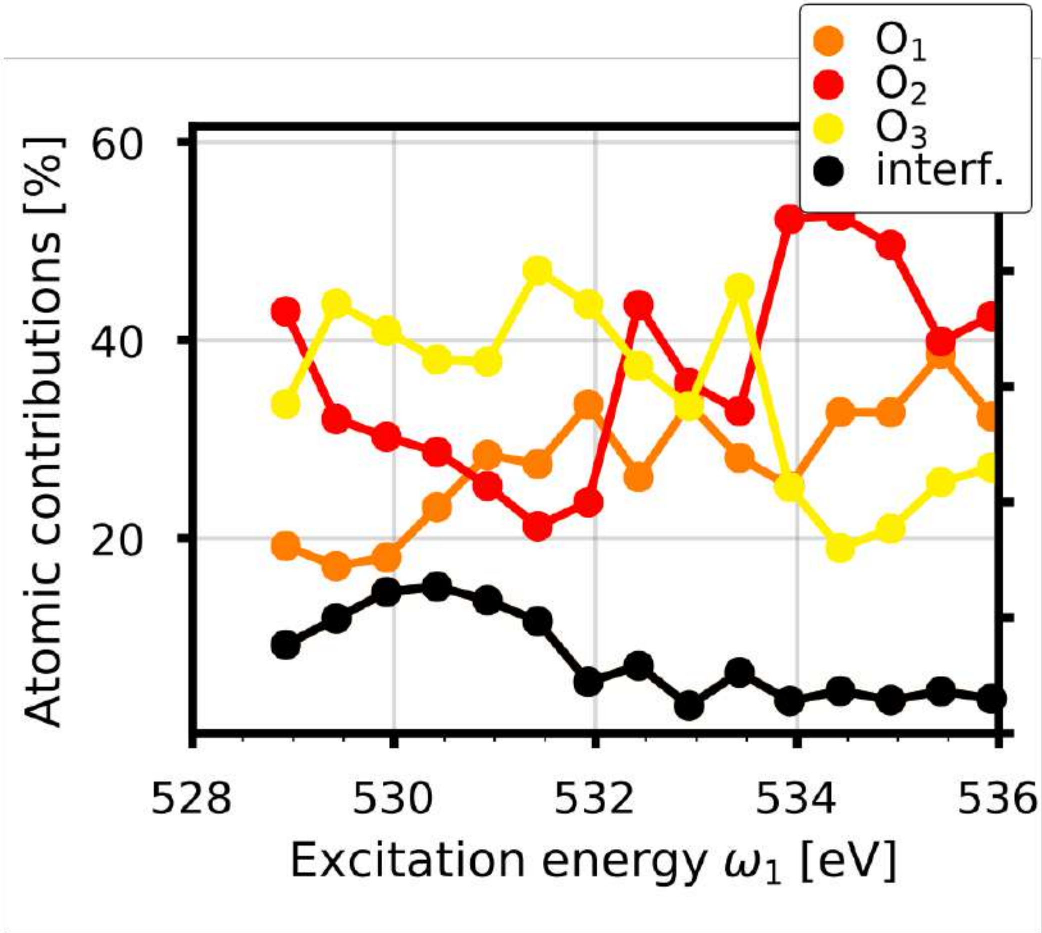

534.93 eV
534.43 eV
532.93 eV
532.43 eV
531.43 eV
530.43 eV
d²σ/dΩ₂dω₂ [arb. units]
522
523
524
525
Emission energy ω₂ [eV]

O-K Ga_2O_3

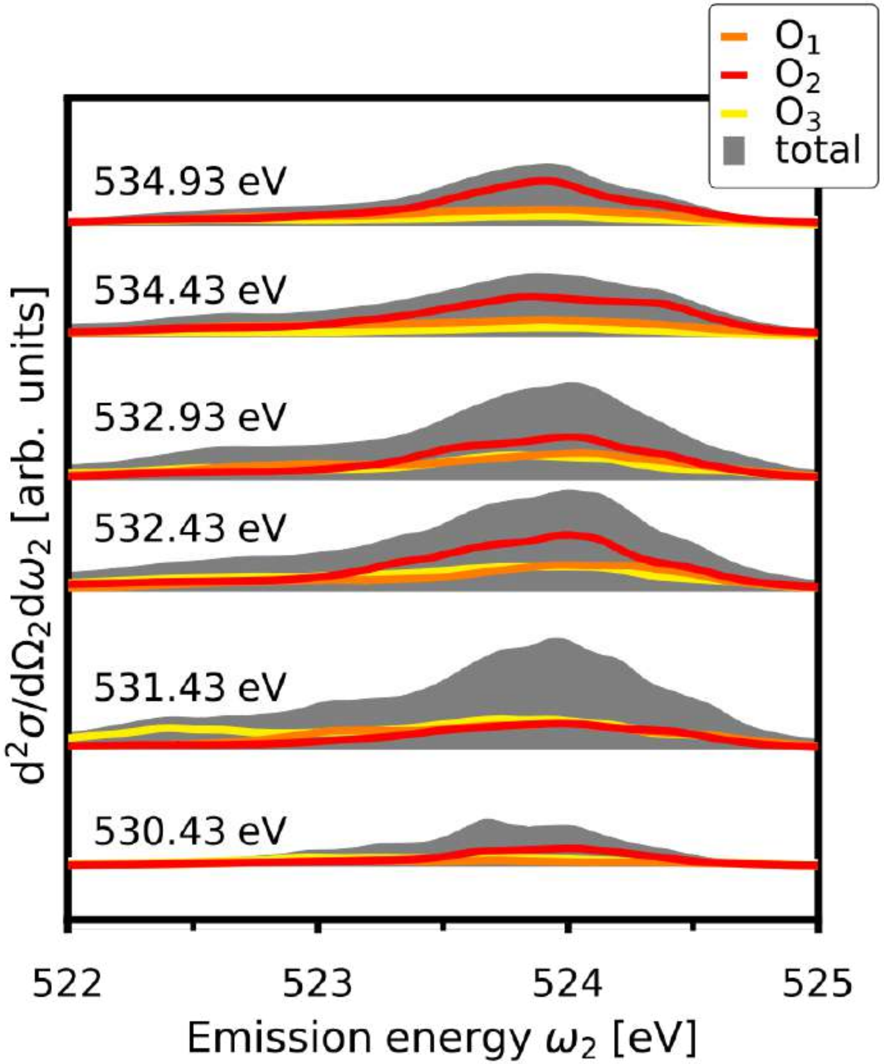


O1
O2
O3
total
534.93 eV
534.43 eV
532.93 eV
532.43 eV
531.43 eV
530.43 eV
d2σ/dΩ2dω2 [arb. units]
522
523
524
525
Emission energy ω2 [eV]

O-K Ga_2O_3



O1
O2
O3
interf.
Atomic contributions [%]
60
40
20
528
530
532
534
536
Excitation energy ω1 [eV]



O1
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534.93 eV
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d²σ/dΩ2dω2 [arb. units]
522
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- Green's functions approach to spectra
- Excitonic effects in (non-)resonant, coherent IXS
- Coherence in RIXS

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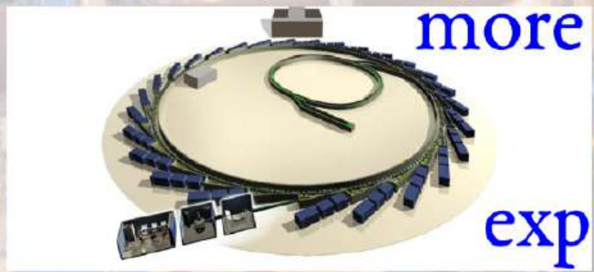

- Coherence in RIXS

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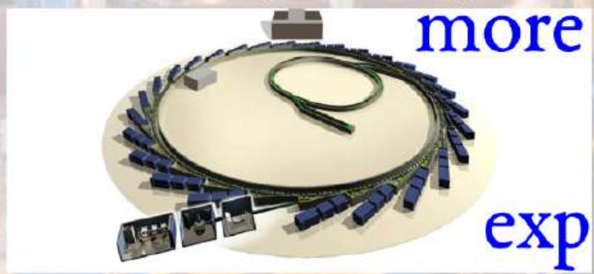
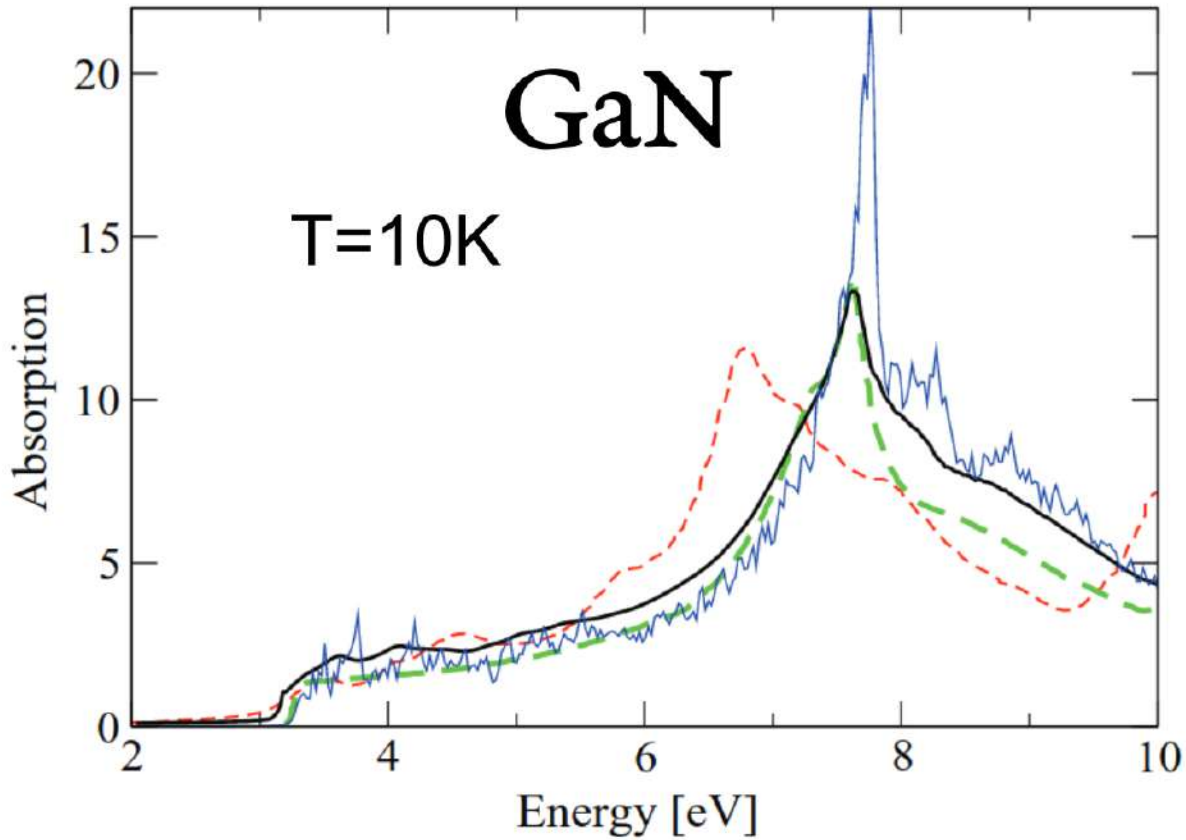


- Coherence in RIXS

Temperature electron-phonon



Kawai *et al.* Phys. Rev. B **89**, 085202 (2014)

Evidence of ideal excitonic insulator in bulk MoS_2 under pressure

S. Samaneh Ataei[a,1], Daniele Varsano[a,1], Elisa Molinari[a,b], and Massimo Rontani[a,2]

PNAS 2021 Vol. 118 No. 13 e2010110118 https://doi.org/10.1073/pnas.2010110118

PHYSICAL REVIEW B, VOLUME 65, 155332

Bethe-Salpeter equation for magnetoexcitons in quantum wells

Z. G. Koinov*
Department of Physics & Astronomy, University of Texas at San Antonio, San Antonio, Texas 78249
(Received 10 December 2001; published 11 April 2002)

PRL **116,** 196804 (2016) PHYSICAL REVIEW LETTERS week ending 13 MAY 2016

Three-particle correlation from a Many-Body Perspective: Trions in a Carbon Nanotube

Thorsten Deilmann,* Matthias Drüppel, and Michael Rohlfing

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