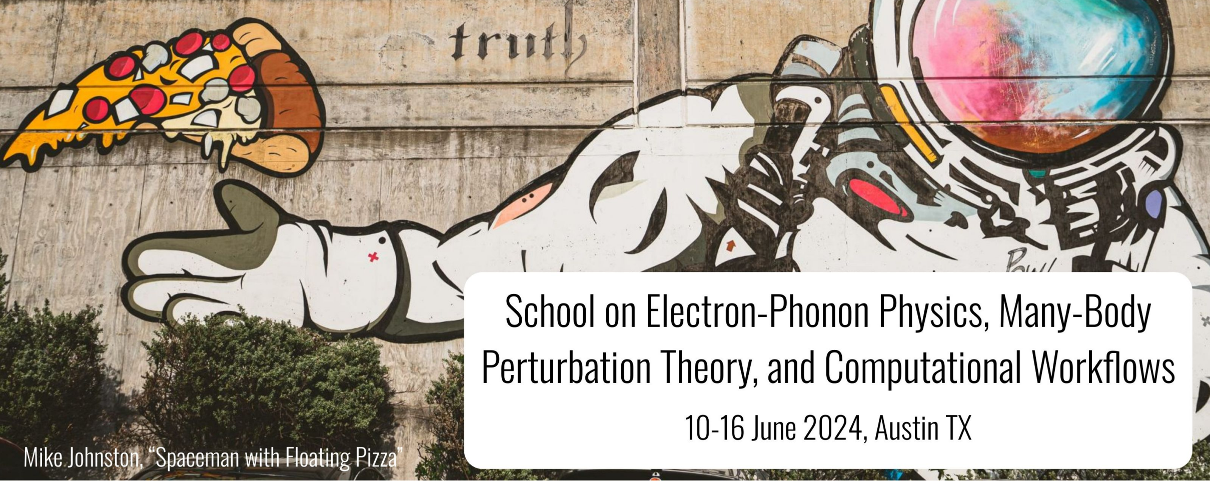


Mike Johnston, "Spaceman with Floating Pizza"

School on Electron-Phonon Physics, Many-Body Perturbation Theory, and Computational Workflows

10-16 June 2024, Austin TX

U.S. DEPARTMENT OF ENERGY

NSF

TACC
TEXAS ADVANCED COMPUTING CENTER

ODEN

Lecture Sat. 5

Quasidegenerate many-body perturbation theory for optical absorption and luminescence

Sabyasachi Tiwari

Oden Institute for Computational Engineering and Sciences

The University of Texas at Austin

Lecture Summary

- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

Lecture Summary

- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

Introduction

- Optoelectronics

- Optoelectronics
  - For energy applications

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed
  - Theory needed to determine best materials

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed
  - Theory needed to determine best materials
  - Quantity to access a material (absorption coefficient: α)

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed
  - Theory needed to determine best materials
  - Quantity to access a material (absorption coefficient: α)
- Optical absorption

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed
  - Theory needed to determine best materials
  - Quantity to access a material (absorption coefficient: α)
- Optical absorption
  - Direct (e.g., GaAs)

Introduction

- Optoelectronics
  - For energy applications
- Material design space is large and theory is needed
  - Theory needed to determine best materials
  - Quantity to access a material (absorption coefficient: α)
- Optical absorption
  - Direct (e.g., GaAs)
  - Indirect phonon-assisted (e.g., Si)

Theory of optical absorption

Full Hamiltonian of a system exposed to electromagnetic radiation can be written as,

$$\hat{H} = \hat{H}_0 + \hat{V}_{\mathrm{ep}} + \hat{V}_{\mathrm{er}}$$

Rev. Mod. Phys. 89, 015003 (2017)

Theory of optical absorption

Full Hamiltonian of a system exposed to electromagnetic radiation can be written as,

$$\hat{H} = \hat{H}_0 + \hat{V}_{\mathrm{ep}} + \hat{V}_{\mathrm{er}}$$

$\hat{V}_{\mathrm{er}} = eA_0\mathbf{e}.\sum_{cv\mathbf{k}} \mathbf{v}_{cv\mathbf{k}}\hat{c}^{\dagger}_{c\mathbf{k}}\hat{c}_{v\mathbf{k}}\cos(\omega t) \rightarrow$ Electron-radiation potential

Rev. Mod. Phys. 89, 015003 (2017)

Theory of optical absorption

Full Hamiltonian of a system exposed to electromagnetic radiation can be written as,

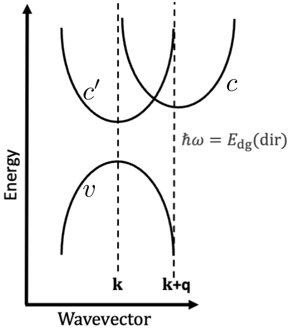
$$\hat{H} = \hat{H}_0 + \hat{V}_{\rm ep} + \hat{V}_{\rm er}$$

$\hat{V}_{\rm ep} = \frac{1}{N^{1/2}} \sum_{mn\mathbf{k}\nu} g_{mn\nu}(\mathbf{k},\mathbf{q}) \hat{c}^{\dagger}_{m\mathbf{k}+\mathbf{q}} \hat{c}_{n\mathbf{k}} (\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu}) \rightarrow$ Electron-phonon potential

Rev. Mod. Phys. 89, 015003 (2017)

Materials whose band gap is direct:

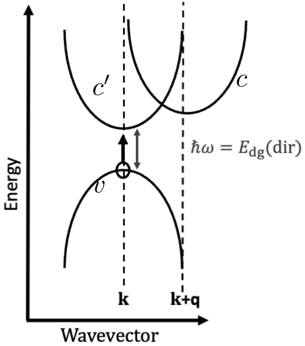
- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\mathrm{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}$



Direct gap materials

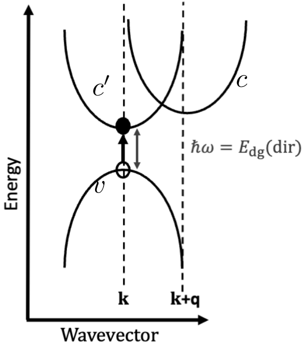
Materials whose band gap is direct:

- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\text{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}} \hat{c}_{v\mathbf{k}}$
- $\hat{c}^{\dagger}_{c'\mathbf{k}} \hat{c}_{v\mathbf{k}} |i_0\rangle$



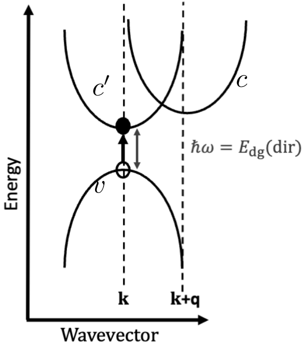
Materials whose band gap is direct:

- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\mathrm{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}$
- $\hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}|i_0\rangle \rightarrow |i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$



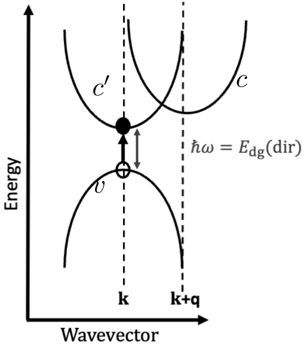
Materials whose band gap is direct:

- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\text{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}} \hat{c}_{v\mathbf{k}}$
- $\hat{c}^{\dagger}_{c'\mathbf{k}} \hat{c}_{v\mathbf{k}} |i_0\rangle \rightarrow |i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle \rightarrow |f\rangle$



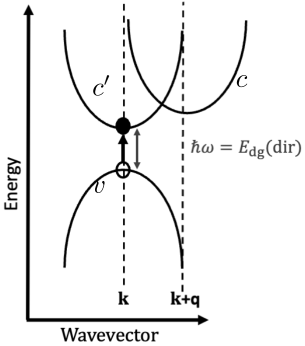
Materials whose band gap is direct:

- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\mathrm{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}$
- $\hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}|i_0\rangle \rightarrow |i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle \rightarrow |f\rangle$
- $\hbar\omega = \epsilon_{c'\mathbf{k}} - \epsilon_{v\mathbf{k}}$



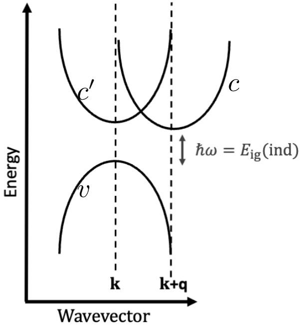
Materials whose band gap is direct:

- The electron-hole pairs are created at the same wavevector
- $\hat{V}_{\text{er}} \propto \hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}$
- $\hat{c}^{\dagger}_{c'\mathbf{k}}\hat{c}_{v\mathbf{k}}|i_0\rangle \rightarrow |i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle \rightarrow |f\rangle$
- $\hbar\omega = \epsilon_{c'\mathbf{k}} - \epsilon_{v\mathbf{k}}$
- Transition rate:
  - $\Gamma_{\text{dir}} = \frac{2\pi}{\hbar}|\langle f|\hat{V}_{\text{er}}|i_0\rangle|^2$



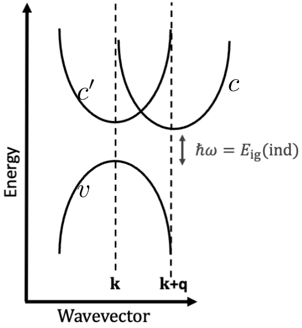
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons



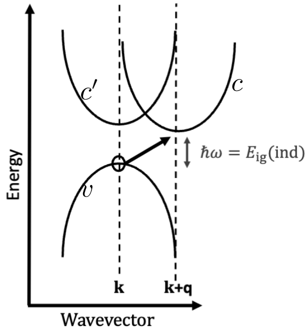
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons



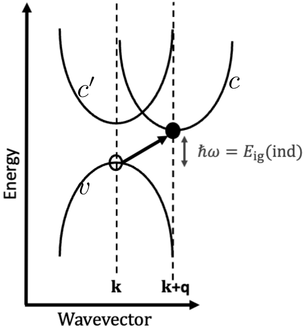
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons



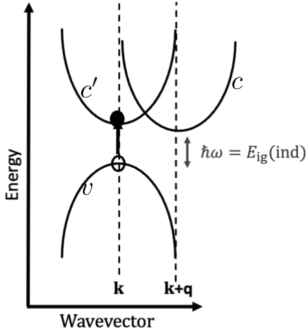
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons
- $\hat{V}_{\mathrm{ep}} \propto \hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}}\hat{c}_{c'\mathbf{k}}(\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})$



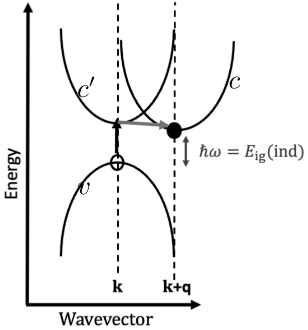
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons
- $\hat{V}_{\rm ep} \propto \hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}}\hat{c}_{c'\mathbf{k}}(\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})$
- $\hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}}\hat{c}_{c'\mathbf{k}}(\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})|i_0 + 1_{c'\mathbf{k}} - 1_{v\mathbf{k}}\rangle$



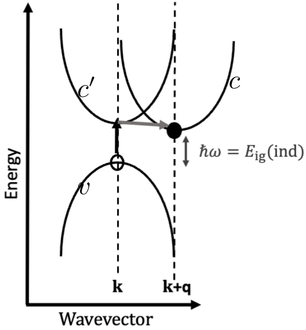
Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons
- $\hat{V}_{\mathrm{ep}} \propto \hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}} \hat{c}_{c'\mathbf{k}} (\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})$
- $\hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}} \hat{c}_{c'\mathbf{k}} (\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu}) |i_0 + 1_{c'\mathbf{k}} - 1_{v\mathbf{k}}\rangle$
  - $|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} \pm 1_{\mp\mathbf{q}\nu}\rangle$
- $\hbar\omega = \epsilon_{c\mathbf{k}+\mathbf{q}} - \epsilon_{v\mathbf{k}} \pm \hbar\omega_{\mathbf{q}\nu}$



Materials whose band gap is indirect:

- The electron-hole pairs are created at different wavevectors
- Extra momentum is needed from phonons
- $\hat{V}_{\rm ep} \propto \hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}}\hat{c}_{c'\mathbf{k}}(\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})$
- $\hat{c}^{\dagger}_{c\mathbf{k}+\mathbf{q}}\hat{c}_{c'\mathbf{k}}(\hat{a}^{\dagger}_{-\mathbf{q}\nu} + \hat{a}_{\mathbf{q}\nu})|i_0 + 1_{c'\mathbf{k}} - 1_{v\mathbf{k}}\rangle$
  - $|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} \pm 1_{\mp\mathbf{q}\nu}\rangle$
- $\hbar\omega = \epsilon_{c\mathbf{k}+\mathbf{q}} - \epsilon_{v\mathbf{k}} \pm \hbar\omega_{\mathbf{q}\nu}$
- Transition rate:
  - $\Gamma_{\rm ind} \propto \left| \frac{\langle f|\hat{V}_{\rm ep}|t\rangle\langle t|\hat{V}_{\rm er}|i_0\rangle}{E_f - E_t} + \frac{\langle f|\hat{V}_{\rm er}|p\rangle\langle p|\hat{V}_{\rm ep}|i_0\rangle}{E_p - E_i} \right|^2$



Indirect gap materials

CHBB Theory:
Second-order perturbation theory

$$\frac{dN_{\mathrm{p}}}{dt} = \frac{2\pi}{\hbar}\frac{e^2A_0^2}{2^2}\frac{1}{N}\sum_{cv\nu,\mathbf{k},\mathbf{q}}^{\eta=\pm1}\left|\mathbf{e}\cdot\left[\mathbf{S}^{(1)}_{cv\nu\eta}(\mathbf{k},\mathbf{q})+\mathbf{S}^{(2)}_{cv\nu\eta}(\mathbf{k},\mathbf{q})\right]\right|^2$$
$$\times\ [n_{\mathbf{q}\nu}+(1+\eta)/2]\delta(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v\mathbf{k}}+\eta\hbar\omega_{\mathbf{q}\nu}-\hbar\omega)$$

Proc. Phys. Soc. A 65, 25 (1952)
Phys. Rev. 95, 559 (1954)
Phys. Rev. Lett. 108, 167402 (2012)

CHBB Theory: Wed. 3. Kioupakis
Second-order perturbation theory

$$\frac{dN_{\rm p}}{dt} = \frac{2\pi}{\hbar}\frac{e^2 A_0^2}{2^2}\frac{1}{N}\sum_{c\upsilon\nu,\mathbf{k},\mathbf{q}}^{\eta=\pm 1}\left|\mathbf{e}\cdot\left[\mathbf{S}^{(1)}_{c\upsilon\nu\eta}(\mathbf{k},\mathbf{q})+\mathbf{S}^{(2)}_{c\upsilon\nu\eta}(\mathbf{k},\mathbf{q})\right]\right|^2$$
$$\times \quad [n_{\mathbf{q}\nu}+(1+\eta)/2]\delta(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{\upsilon\mathbf{k}}+\eta\hbar\omega_{\mathbf{q}\nu}-\hbar\omega)$$

Proc. Phys. Soc. A 65, 25 (1952)
Phys. Rev. 95, 559 (1954)
Phys. Rev. Lett. 108, 167402 (2012)

CHBB Theory:
Second-order perturbation theory

$$-\mathbf{S}^{(1)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{g_{cn\nu}(\mathbf{k},\mathbf{q})\,\mathbf{v}_{nv\mathbf{k}}}{\varepsilon_{c\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}} \ ,$$

$$\mathbf{S}^{(2)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{\mathbf{v}_{cn\mathbf{k}+\mathbf{q}}\,g_{nv\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{n\mathbf{k}+\mathbf{q}} - \varepsilon_{v\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}}$$

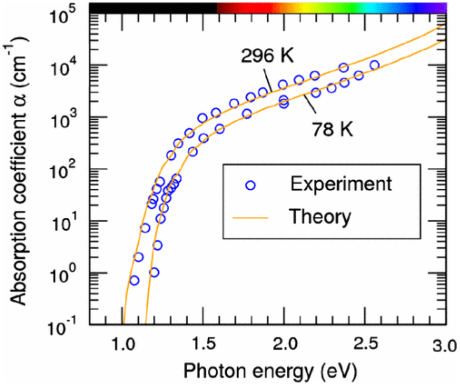
Phys. Rev. Lett. 108, 167402 (2012)

Indirect gap materials

CHBB Theory:
Second-order perturbation theory

$$-\mathbf{S}^{(1)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{g_{cn\nu}(\mathbf{k},\mathbf{q})\,\mathbf{v}_{nv\mathbf{k}}}{\varepsilon_{c\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}},$$

$$\mathbf{S}^{(2)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{\mathbf{v}_{cn\mathbf{k}+\mathbf{q}}\,g_{nv\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{n\mathbf{k}+\mathbf{q}} - \varepsilon_{v\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}}$$



Works well for indirect gap materials

Phys. Rev. Lett. 108, 167402 (2012)

CHBB Theory:
Second-order perturbation theory

$$-\mathbf{S}^{(1)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{g_{cn\nu}(\mathbf{k},\mathbf{q})\,\mathbf{v}_{nv\mathbf{k}}}{\varepsilon_{c\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}},$$

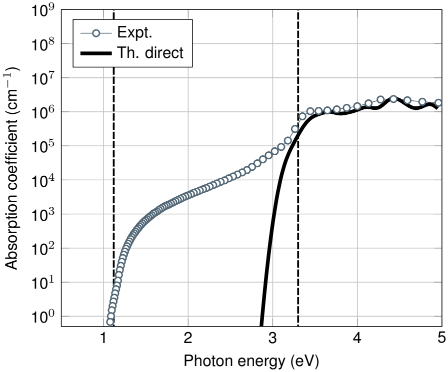
$$\mathbf{S}^{(2)}_{cv\nu\eta}(\mathbf{k},\mathbf{q}) = \sum_n \frac{\mathbf{v}_{cn\mathbf{k}+\mathbf{q}}\,g_{nv\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{n\mathbf{k}+\mathbf{q}} - \varepsilon_{v\mathbf{k}} + \eta\hbar\omega_{\mathbf{q}\nu}}$$

Works well for indirect gap materials

Phys. Rev. Lett. 108, 167402 (2012)

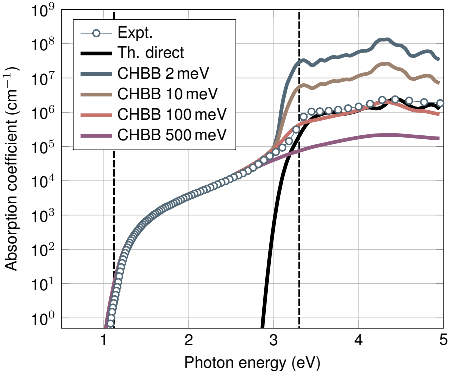
Lecture Summary

- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

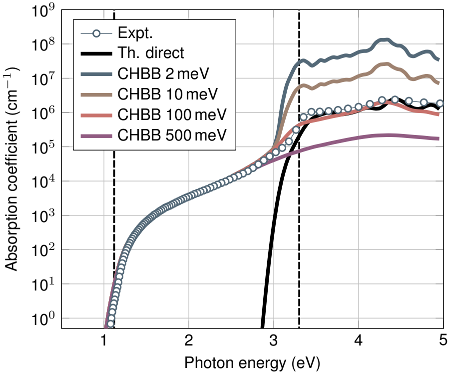


Mon. 1. Giustino

Limitations of the CHBB theory



Mon. 1. Giustino

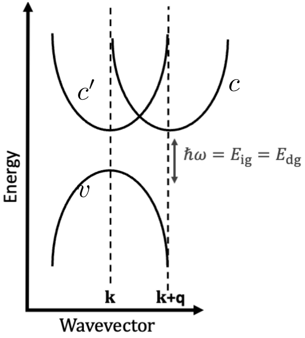


Mon. 1. Giustino

CHBB theory becomes unphysical in the regime of direct absorption

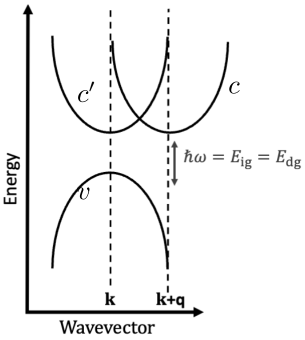
What happens to transition rate?

- The electron-hole pairs can be at the same or at different wavevectors



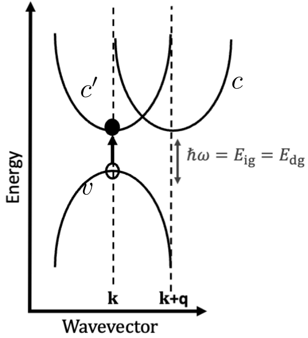
What happens to transition rate?

- The electron-hole pairs can be at the same or at different wavevectors



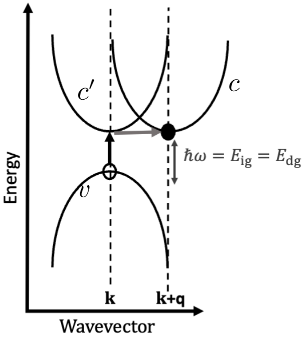
What happens to transition rate?

- The electron-hole pairs can be at the same or at different wavevectors
- $E_t = \epsilon_{c'\mathbf{k}} - \epsilon_{v\mathbf{k}} \approx$



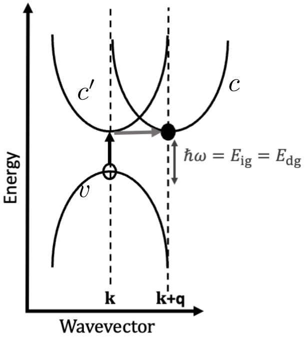
What happens to transition rate?

- The electron-hole pairs can be at the same or at different wavevectors
- $E_t = \epsilon_{c'\mathbf{k}} - \epsilon_{v\mathbf{k}} \approx E_f = \epsilon_{c\mathbf{k+q}} - \epsilon_{v\mathbf{k}} \pm \hbar\omega_{\mathbf{q}\nu}$

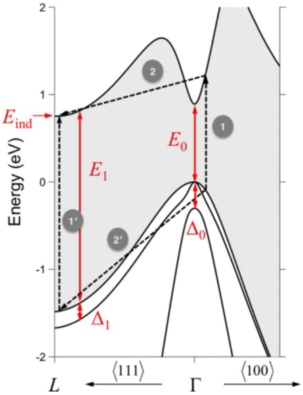


What happens to transition rate?

- The electron-hole pairs can be at the same or at different wavevectors
- $E_t = \epsilon_{c'\mathbf{k}} - \epsilon_{v\mathbf{k}} \approx E_f = \epsilon_{c\mathbf{k+q}} - \epsilon_{v\mathbf{k}} \pm \hbar\omega_{\mathbf{q}\nu}$
  - $\Gamma_{\text{ind}} \propto \left|\frac{1}{E_f - E_t}\right|^2 \to \infty$

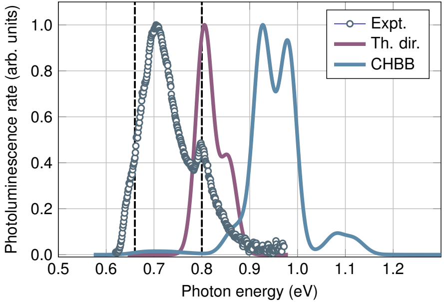


Quasi-direct gap material (Ge)



Phys. Rev. B 98, 165207 (2017)

Quasi-direct gap material (Ge)

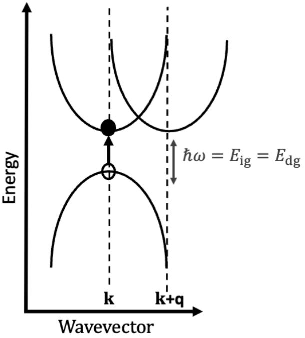


Direct absorption only captures single peak
Indirect absorption peak shifted due to degeneracy from onset

Materials whose direct and indirect gaps are close

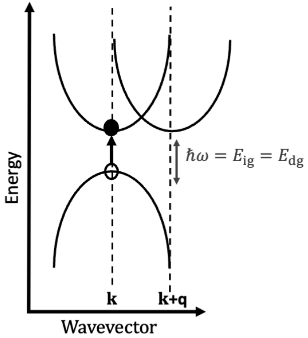
$$\Gamma_{\text{total}} = \Gamma_{\text{dir}} + \Gamma_{\text{ind}} \ ?$$

$|i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$

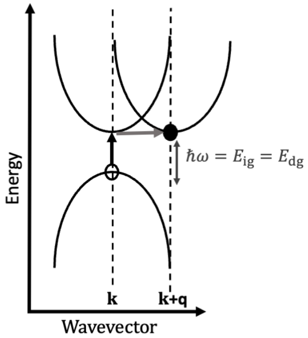


Quasi-direct gap materials

$|i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$

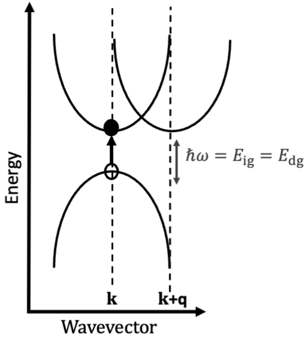


$|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} \pm 1_{\mathbf{q}\nu}\rangle$



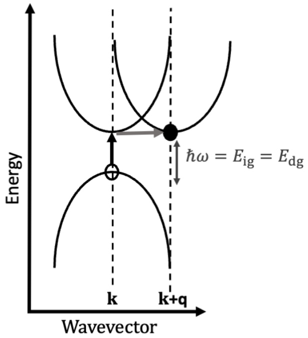
Quasi-direct gap materials

$|i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$



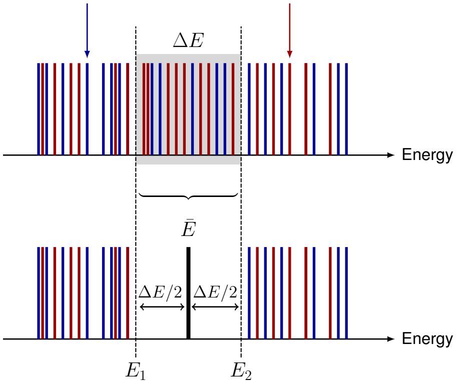
Are degenerate

$|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} \pm 1_{\mathbf{q}\nu}\rangle$



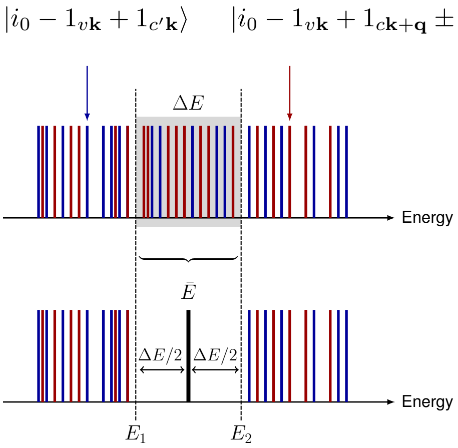
- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

$|i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$



J. Phys. B 7, 2441 (1974)

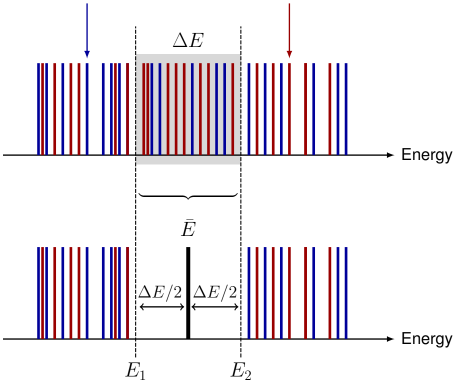
Quasidegenerate many-body perturbation theory



J. Phys. B 7, 2441 (1974)

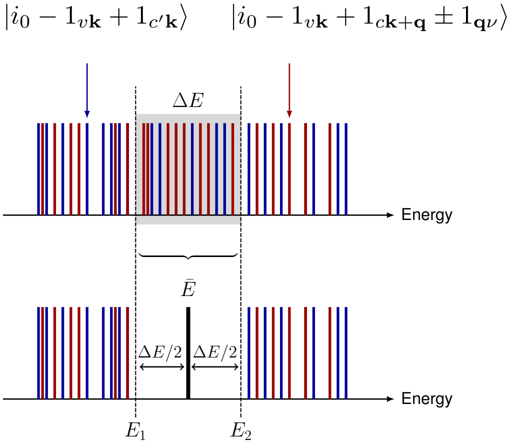
$|i_0 - 1_{v\mathbf{k}} + 1_{c'\mathbf{k}}\rangle$ $|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} \pm 1_{\mathbf{q}\nu}\rangle$

1. Find states in ΔE QD bin



J. Phys. B 7, 2441 (1974)

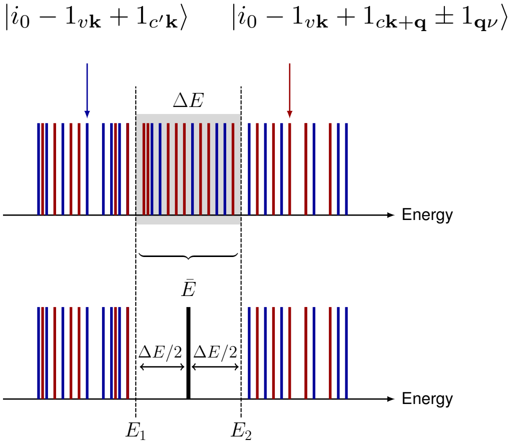
Quasidegenerate many-body perturbation theory



1. Find states in ΔE QD bin
2. $\hat{V}'_{\text{ep}} = \hat{V}_{\text{ep}} - \sum_p (\bar{E} - E_{d_0;p})|d_0;p\rangle\langle d_0;p|$

J. Phys. B 7, 2441 (1974)

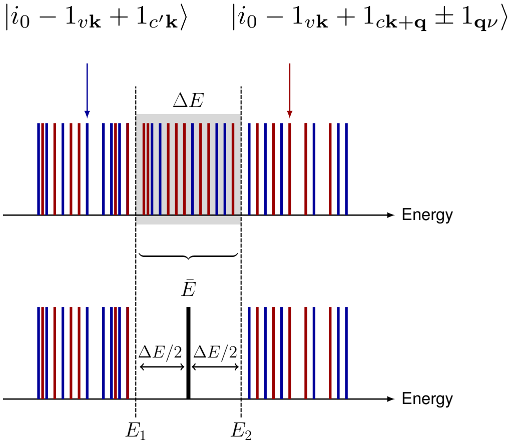
Quasidegenerate many-body perturbation theory



1. Find states in ΔE QD bin
2. $\hat{V}'_{\text{ep}} = \hat{V}_{\text{ep}} - \sum_p (\bar{E} - E_{d_0;p}) |d_0; p\rangle\langle d_0; p|$
3. $\langle d_0; s|\hat{V}'_{\text{ep}}|d_0; p\rangle = \sum_m U_{sm}\lambda_m U^{-1}_{mp}$

J. Phys. B 7, 2441 (1974)

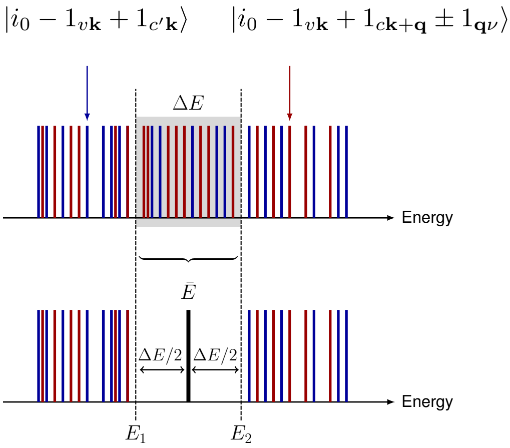
Quasidegenerate many-body perturbation theory



1. Find states in ΔE QD bin
2. $\hat{V}'_{\text{ep}} = \hat{V}_{\text{ep}} - \sum_p (\bar{E} - E_{d_0;p})|d_0;p\rangle\langle d_0;p|$
3. $\langle d_0;s|\hat{V}'_{\text{ep}}|d_0;p\rangle = \sum_m U_{sm}\lambda_m U^{-1}_{mp}$
4. $|f;m\rangle = \sum_p U_{mp}\left[|d_0;p\rangle + \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\text{ep}}|d_0;p\rangle}{\bar{E}-E_{t_0}}|t_0\rangle\right]$

J. Phys. B 7, 2441 (1974)

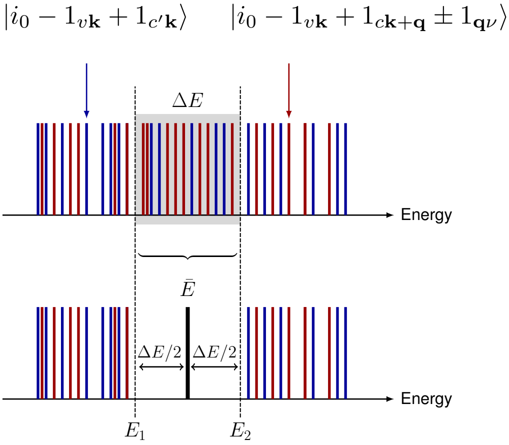
Quasidegenerate many-body perturbation theory



1. Find states in ΔE QD bin
2. $\hat{V}'_{\text{ep}} = \hat{V}_{\text{ep}} - \sum_p (\bar{E} - E_{d_0;p})|d_0;p\rangle\langle d_0;p|$
3. $\langle d_0;s|\hat{V}'_{\text{ep}}|d_0;p\rangle = \sum_m U_{sm}\lambda_m U^{-1}_{mp}$
4. $|f;m\rangle = \sum_p U_{mp}\left[|d_0;p\rangle + \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\text{ep}}|d_0;p\rangle}{\bar{E}-E_{t_0}}|t_0\rangle\right]$
5. Calculate observable for the current bin$\rightarrow$ $\Gamma = \frac{2\pi}{\hbar}|\langle i_0|\hat{V}_{\text{er}}|f;m\rangle|^2\delta(E_f - E_i - \hbar\omega)$

J. Phys. B 7, 2441 (1974)

Quasidegenerate many-body perturbation theory



1. Find states in ΔE QD bin
2. $\hat{V}'_{\text{ep}} = \hat{V}_{\text{ep}} - \sum_p (\bar{E} - E_{d_0;p})|d_0;p\rangle\langle d_0;p|$
3. $\langle d_0;s|\hat{V}'_{\text{ep}}|d_0;p\rangle = \sum_m U_{sm}\lambda_m U^{-1}_{mp}$
4. $|f;m\rangle = \sum_p U_{mp}\left[|d_0;p\rangle + {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\text{ep}}|d_0;p\rangle}{\bar{E}-E_{t_0}}|t_0\rangle\right]$
5. Calculate observable for the current bin$\rightarrow$ $\Gamma = \frac{2\pi}{\hbar}|\langle i_0|\hat{V}_{\text{er}}|f;m\rangle|^2\delta(E_f - E_i - \hbar\omega)$
6. Repeat same steps for all QD bins

J. Phys. B 7, 2441 (1974)

Quasidegenerate many-body perturbation theory

$$
\begin{aligned}
|f;m\rangle = &\sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle \; + \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}-1_{\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} - 1_{\mathbf{q}\nu}\rangle \\
&+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+1_{-\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} + 1_{-\mathbf{q}\nu}\rangle \\
&+ \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle \\
&+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}-1_{\mathbf{q}\nu}} \times {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} - 1_{\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle \\
&+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+1_{-\mathbf{q}\nu}} \times {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} + 1_{-\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle
\end{aligned}
$$

Quasidegenerate many-body perturbation theory

$$|f;m\rangle = \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle + \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}}|i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}}|i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle}{\bar{E} - E_{t_0}}|t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}}|t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}}|t_0\rangle$$

Quasidegenerate many-body perturbation theory

$$|f;m\rangle = \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle + \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}-1_{\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} - 1_{\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+1_{-\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} + 1_{-\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}-1_{\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} - 1_{\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+1_{-\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k+q}} + 1_{-\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

Quasidegenerate many-body perturbation theory

$$|f;m\rangle = \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle + \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} - 1_{\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} + 1_{-\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} - 1_{\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}} \times \sum_{t_0}{}' \frac{\langle t_0|\hat{V}_{\mathrm{ep}}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} + 1_{-\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

Quasidegenerate many-body perturbation theory

$$|f;m\rangle = \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle + \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} - 1_{\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}} |i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} + 1_{-\mathbf{q}\nu}\rangle$$

$$+ \sum_{cv\mathbf{k}} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\rm ep}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}-1_{\mathbf{q}\nu}} \times {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\rm ep}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} - 1_{\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

$$+ \sum_{cv\mathbf{k},\mathbf{q}\nu} U_{m,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+1_{-\mathbf{q}\nu}} \times {\sum_{t_0}}' \frac{\langle t_0|\hat{V}_{\rm ep}|i_0 - 1_{v\mathbf{k}} + 1_{c\mathbf{k}+\mathbf{q}} + 1_{-\mathbf{q}\nu}\rangle}{\bar{E} - E_{t_0}} |t_0\rangle$$

Mixing states outside QD bin

Quasidegenerate many-body perturbation theory

$$\Gamma_{i\to(f;p)} = \frac{\pi e^2 A_0^2}{2\hbar}\Bigg|\mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+\quad N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\;U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}} \qquad \text{(A)}$$

$$+\quad\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k+q}}\,g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k+q}}-\varepsilon_{v'\mathbf{k+q}})}\,\theta_{cv'\mathbf{k+q}} \qquad \text{(B)}$$

$$+\quad\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k+q}}\,g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k+q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \qquad \text{(C)}$$

$$+\quad\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k+q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2 \qquad \text{(D)}$$

$$\times\quad\delta(E_p-E_{i_0}-\hbar\omega)$$

Quasidegenerate many-body perturbation theory

$$\Gamma_{i\to(f;p)} = \frac{\pi e^2 A_0^2}{2\hbar}\Bigg|\mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+\ N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\ U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k+q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\theta_{c'v\mathbf{k}} \qquad \text{(A)}$$

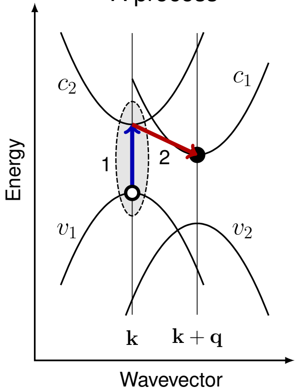
$$+\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k+q}}\,g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k+q}}-\varepsilon_{v'\mathbf{k+q}})}\theta_{cv'\mathbf{k+q}} \qquad \text{(B)}$$

$$+\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k+q}}\,g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k+q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \qquad \text{(C)}$$

$$+\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k+q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2 \qquad \text{(D)}$$

$$\times\ \delta(E_p-E_{i_0}-\hbar\omega)$$

A process



$$\Gamma_{i\to(f;p)} = \frac{\pi e^2 A_0^2}{2\hbar}\Bigg|\mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+\ N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\ U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

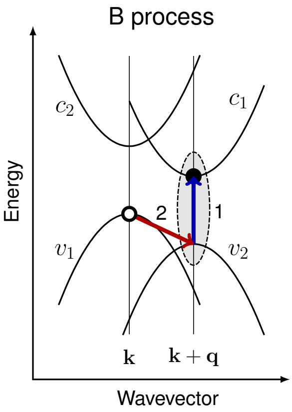
$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\theta_{c'v\mathbf{k}} \qquad \text{(A)}$$

$$+\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\,g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\theta_{cv'\mathbf{k}+\mathbf{q}} \qquad \text{(B)}$$

$$+\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\,g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \qquad \text{(C)}$$

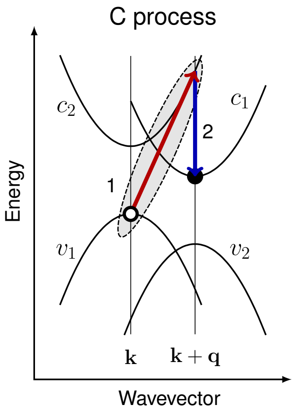
$$+\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2 \qquad \text{(D)}$$

$$\times\ \delta(E_p - E_{i_0} - \hbar\omega)$$



Quasidegenerate many-body perturbation theory

$$
\begin{aligned}
\Gamma_{i\to(f;p)} = \frac{\pi e^2 A_0^2}{2\hbar}\Bigg|\, \mathbf{e}\cdot\sum_{cv\mathbf{k}} \Bigg\{ & U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}} \\
& + N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\; U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}} \\
& \times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}} \qquad \text{(A)} \\
& + \sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\,\theta_{cv'\mathbf{k}+\mathbf{q}} \qquad \text{(B)} \\
& + \sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \qquad \text{(C)} \\
& + \sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2 \qquad \text{(D)} \\
& \times\ \delta(E_p - E_{i_0} - \hbar\omega)
\end{aligned}
$$



$$\Gamma_{i\to(f;p)} = \frac{\pi e^2 A_0^2}{2\hbar}\Bigg|\mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+\; N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\;U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

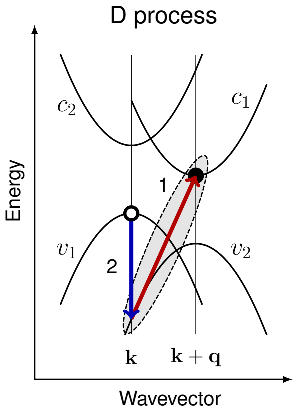
$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\theta_{c'v\mathbf{k}} \qquad \text{(A)}$$

$$+\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\,g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\theta_{cv'\mathbf{k}+\mathbf{q}} \qquad \text{(B)}$$

$$+\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\,g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \qquad \text{(C)}$$

$$+\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2 \qquad \text{(D)}$$

$$\times\;\delta(E_p - E_{i_0} - \hbar\omega)$$



Imaginary dielectric constant

$$
\begin{aligned}
\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega}\frac{1}{\omega^2}\frac{1}{N}\sum_{i_0,p} Z^{-1}\exp\left(-\beta E_{i_0}\right) \times & \left| \mathbf{e}\cdot\sum_{cv\mathbf{k}} \left\{ U^{*}_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} \mathbf{v}_{cv\mathbf{k}} \right.\right. \\
+ \quad N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\; & U^{*}_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}} \\
\times \Bigg[\sum_{c'} \frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}} & \\
+ \sum_{v'} \frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\,\theta_{cv'\mathbf{k}+\mathbf{q}} & \\
+ \sum_{c'} \frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} & \\
\left.\left. + \sum_{v'} \frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \Bigg] \right\} \right|^2 & \\
\times \quad \delta(E_p - E_{i_0} - \hbar\omega) &
\end{aligned}
$$

Imaginary dielectric constant

$$
\begin{aligned}
\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega}\frac{1}{\omega^2}\frac{1}{N}\sum_{i_0,p} Z^{-1}\exp\left(-\beta E_{i_0}\right) \times \Bigg| \mathbf{e}\cdot\sum_{c v \mathbf{k}} \Bigg\{ & U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}} \\
+ \; N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\; U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}} \\
\times \Bigg[\sum_{c'} \frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}} \\
+ \sum_{v'} \frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\,\theta_{cv'\mathbf{k}+\mathbf{q}} \\
+ \sum_{c'} \frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \\
+ \sum_{v'} \frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \Bigg] \Bigg\} \Bigg|^2 \\
\times \; \delta(E_p - E_{i_0} - \hbar\omega)
\end{aligned}
$$

Direct contribution

Imaginary dielectric constant

$$\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega}\frac{1}{\omega^2}\frac{1}{N}\sum_{i_0,p} Z^{-1}\exp\left(-\beta E_{i_0}\right) \times \left| \mathbf{e}\cdot\sum_{cv\mathbf{k}}\left\{ U^{*}_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}} \right.\right.$$

$$+ \; N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\; U^{*}_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

$$\times \left[\sum_{c'} \frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\theta_{c'v\mathbf{k}} \right.$$

$$+ \sum_{v'} \frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\theta_{cv'\mathbf{k}+\mathbf{q}}$$

$$+ \sum_{c'} \frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}$$

$$\left.\left.\left. + \sum_{v'} \frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}} \right]\right\}\right|^2$$

$$\times \;\; \delta(E_p - E_{i_0} - \hbar\omega)$$

Direct contribution

Indirect contribution

Imaginary dielectric constant

$$
\begin{aligned}
\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega} \frac{1}{\omega^2} \frac{1}{N} \sum_{i_0,p} Z^{-1} \exp\left(-\beta E_{i_0}\right) \times \Bigg| \mathbf{e} \cdot \sum_{cv\mathbf{k}} \Bigg\{ U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}} \mathbf{v}_{cv\mathbf{k}} \\
+ \; N^{-1/2} \sum_{\mathbf{q}\nu\eta} \sqrt{n_{\mathbf{q}\nu} + \frac{1+\eta}{2}} \; U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}} \\
\times \Bigg[\sum_{c'} \frac{g_{cc'\nu}(\mathbf{k},\mathbf{q}) \mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0}) - (\varepsilon_{c'\mathbf{k}} - \varepsilon_{v\mathbf{k}})} \theta_{c'v\mathbf{k}} \\
+ \sum_{v'} \frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}} \, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0}) - (\varepsilon_{c\mathbf{k}+\mathbf{q}} - \varepsilon_{v'\mathbf{k}+\mathbf{q}})} \theta_{cv'\mathbf{k}+\mathbf{q}} \\
+ \sum_{c'} \frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}} \, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}} - \varepsilon_{c'\mathbf{k}+\mathbf{q}} - \eta\hbar\omega_{-\eta\mathbf{q}\nu}} \\
+ \sum_{v'} \frac{g_{cv'\nu}(\mathbf{k},\mathbf{q}) \mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}} - \varepsilon_{c\mathbf{k}+\mathbf{q}} - \eta\hbar\omega_{-\eta\mathbf{q}\nu}} \Bigg] \Bigg\} \Bigg|^2 \\
\times \; \delta(E_p - E_{i_0} - \hbar\omega)
\end{aligned}
$$

Direct contribution

Indirect contribution

In presence of both: the many-body states are entangled and cannot be separated

Imaginary dielectric constant

$$\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega}\frac{1}{\omega^2}\frac{1}{N}\sum_{i_0,p} Z^{-1}\exp\left(-\beta E_{i_0}\right) \times \Bigg| \mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{ U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+ \quad N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\; U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}}$$

$$+\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\, g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\,\theta_{cv'\mathbf{k}+\mathbf{q}}$$

$$+\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\, g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}$$

$$+\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2$$

$$\times\quad \delta(E_p - E_{i_0} - \hbar\omega)$$

Direct contribution

Indirect contribution

In presence of both: the many-body states are entangled and cannot be separated

$\Delta E \rightarrow 0$:Perturbation

Imaginary dielectric constant

$$\varepsilon_2(\omega) = \frac{\pi e^2}{\epsilon_0 \Omega}\frac{1}{\omega^2}\frac{1}{N}\sum_{i_0,p} Z^{-1}\exp\left(-\beta E_{i_0}\right)\times \Bigg|\mathbf{e}\cdot\sum_{cv\mathbf{k}}\Bigg\{U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}}}\mathbf{v}_{cv\mathbf{k}}$$

$$+\quad N^{-1/2}\sum_{\mathbf{q}\nu\eta}\sqrt{n_{\mathbf{q}\nu}+\frac{1+\eta}{2}}\;U^*_{p,i_0-1_{v\mathbf{k}}+1_{c\mathbf{k}+\mathbf{q}}+\eta 1_{-\eta\mathbf{q}\nu}}$$

$$\times\Bigg[\sum_{c'}\frac{g_{cc'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{c'v\mathbf{k}}}{(\bar{E}-E_{i_0})-(\varepsilon_{c'\mathbf{k}}-\varepsilon_{v\mathbf{k}})}\,\theta_{c'v\mathbf{k}}$$

$$+\sum_{v'}\frac{\mathbf{v}_{cv'\mathbf{k}+\mathbf{q}}\,g_{v'v\nu}(\mathbf{k},\mathbf{q})}{(\bar{E}-E_{i_0})-(\varepsilon_{c\mathbf{k}+\mathbf{q}}-\varepsilon_{v'\mathbf{k}+\mathbf{q}})}\,\theta_{cv'\mathbf{k}+\mathbf{q}}$$

$$+\sum_{c'}\frac{\mathbf{v}_{cc'\mathbf{k}+\mathbf{q}}\,g_{c'v\nu}(\mathbf{k},\mathbf{q})}{\varepsilon_{v\mathbf{k}}-\varepsilon_{c'\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}$$

$$+\sum_{v'}\frac{g_{cv'\nu}(\mathbf{k},\mathbf{q})\mathbf{v}_{v'v\mathbf{k}}}{\varepsilon_{v'\mathbf{k}}-\varepsilon_{c\mathbf{k}+\mathbf{q}}-\eta\hbar\omega_{-\eta\mathbf{q}\nu}}\Bigg]\Bigg\}\Bigg|^2$$

$$\times\quad\delta(E_p - E_{i_0} - \hbar\omega)$$

Direct contribution

Indirect contribution

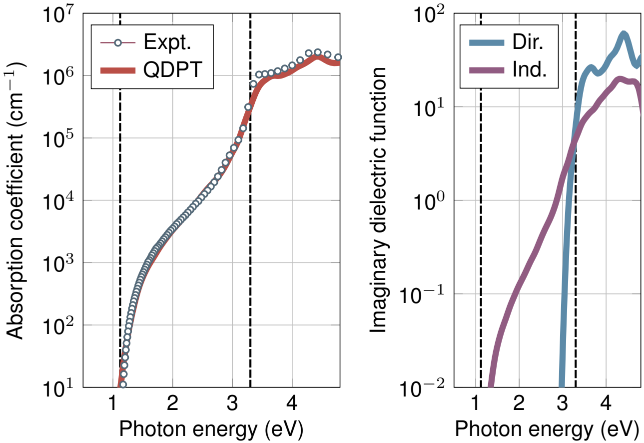
In presence of both: the many-body states are entangled and cannot be separated

$\Delta E \to 0$:Perturbation
$\Delta E \to \infty$:Diagonalization

Lecture Summary

- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

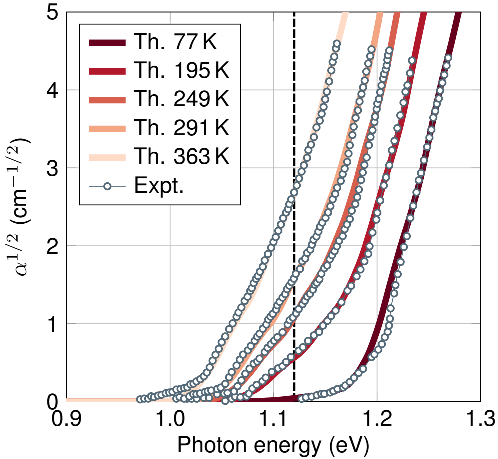
Silicon



Good agreement with experiments

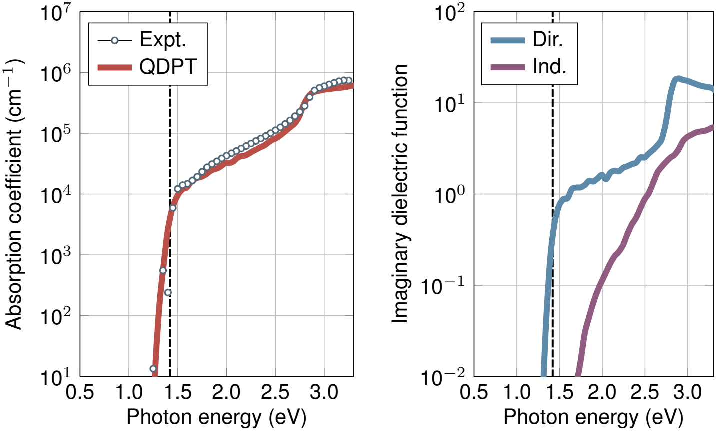
We can also disentangle contributions from direct transitions and phonons

Phys. Rev. B 109, 195127 (2024)



It can recover phonon fine-structure

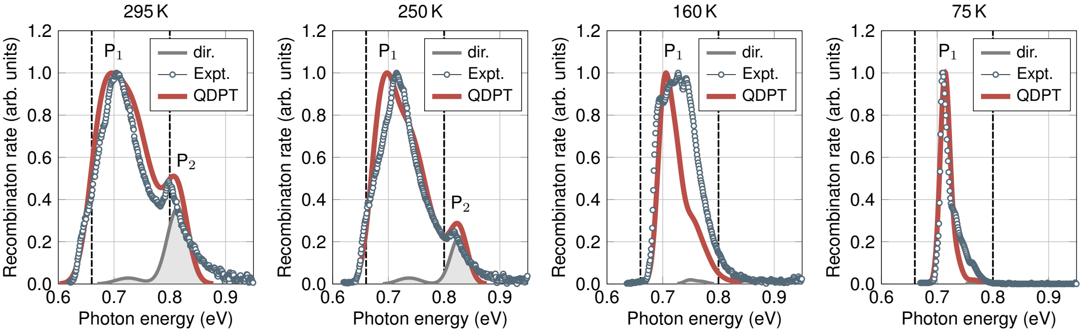
Phys. Rev. 111, 1245 (1958)



Phonons can affect the oscillator strength for higher energies

AIP Adv. 11, 025327 (2021)

Ge (quasi-direct gap)



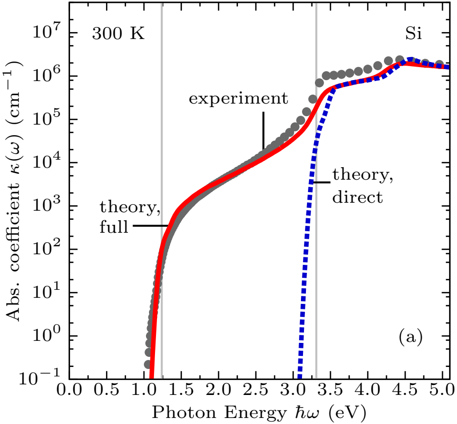
$$R(\omega) = \frac{2n}{\pi c^3} \frac{\omega^3 \varepsilon_2(\omega)}{\exp(\hbar\omega/k_{\mathrm{B}}T)-1}$$

Good agreement with experiments

Phys. Rev. B 101, 195204 (2020)

Alternate methods

- Special displacements method (ZG)
- Fri. 6. Zacharias



Phys. Rev. B 94, 075125 (2016)

Lecture Summary

- Introduction
- Limitations of the CHBB theory
- Quasidegenerate many-body perturbation theory
- Application to materials
- Conclusion

Conclusion

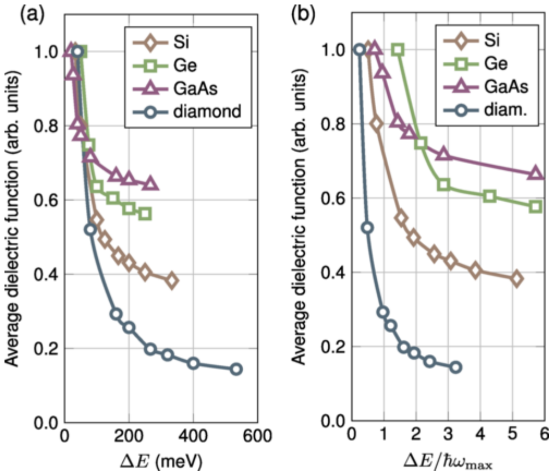
- We have developed a unified theory of optical absorption applicable in all regimes of photon energy
- We applied our method on multiple materials and obtained good agreement with experiments
- QDPT can be easily extended to higher order processes including excitons

References

- S. Tiwari, E. Kioupakis, J. Menendez, and F. Giustino Phys. Rev. B 109, 195127 (2024) [link]
- J. Noffsinger, E. Kioupakis, C. G. Van de Walle, S. G. Louie, and M. L. Cohen, Phys. Rev. Lett. 108, 167402 (2012). [link]

Supplemental Slides

QDPT convergence



QDPT convergence