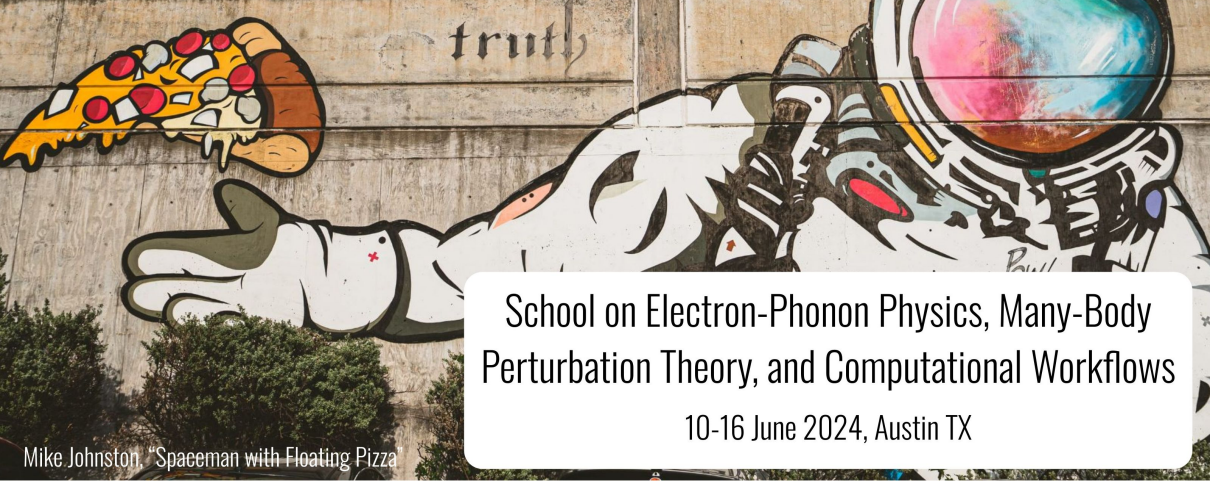

Mike Johnston, "Spaceman with Floating Pizza"

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

10-16 June 2024, Austin TX

U.S. DEPARTMENT OF ENERGY

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TEXAS ADVANCED COMPUTING CENTER

ODEN

Lecture Tue.2

Many-body methods for electron-phonon interactions

Feliciano Giustino

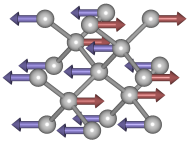
Oden Institute & Department of Physics

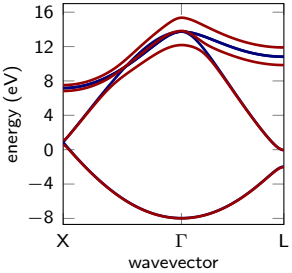
The University of Texas at Austin

Lecture Summary

- Limitations of Rayleigh-Schrödinger perturbation theory
- Many-body Hamiltonian
- Green's function and the spectral function
- Electron-phonon self-energy
- Electron mass enhancement and lifetimes
- How do phonons arise from the many-body Schrödinger equation

Limitations of DFT and Rayleigh-Schrödinger perturbation theory

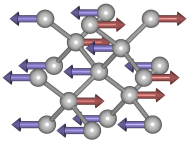


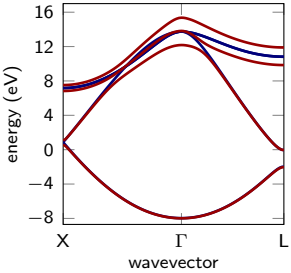


Kohn-Sham equations from Lec. Mon.1

$$-\frac{\hbar^2}{2m_\mathrm{e}}\nabla^2\,\psi_n(\mathbf{r}) + V_\mathrm{SCF}(\mathbf{r};\boldsymbol{\tau}_1,\boldsymbol{\tau}_2,\cdots)\,\psi_n(\mathbf{r}) = \varepsilon_n\,\psi_n(\mathbf{r})$$

Limitations of DFT and Rayleigh-Schrödinger perturbation theory





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- Adiabatic Born-Oppenheimer approximation

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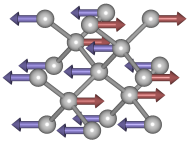

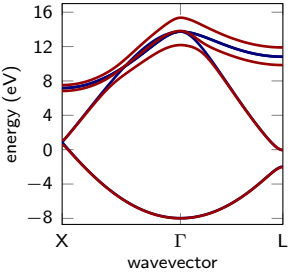

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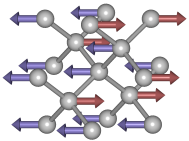

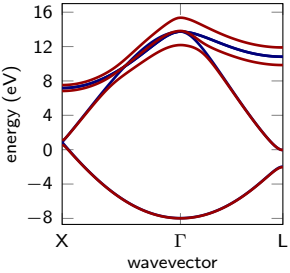

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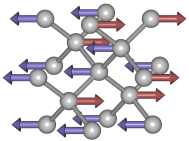
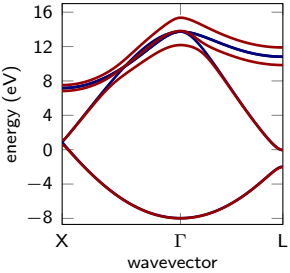
- Electron-phonon interactions depend on XC functional

Limitations of DFT and Rayleigh-Schrödinger perturbation theory





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- Phonons have infinite lifetimes in metals

Phenomena beyond Rayleigh-Schrödinger perturbation theory

→ Lec. Thu.2 Giustino

Polaron liquid at the $SrTiO_3(001)$ surface

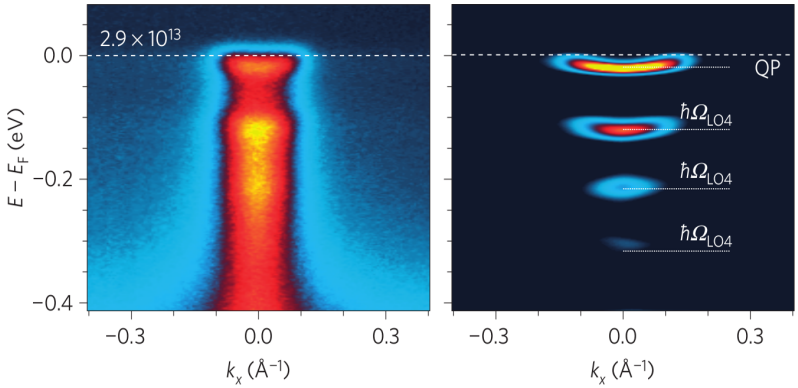

Figure from Wang et al, Nat. Mater. 15, 835 (2016)

Phenomena beyond Rayleigh-Schrödinger perturbation theory

→ Lec. Wed.2 Margine

Andreev reflection spectroscopy of superconducting MgB_2

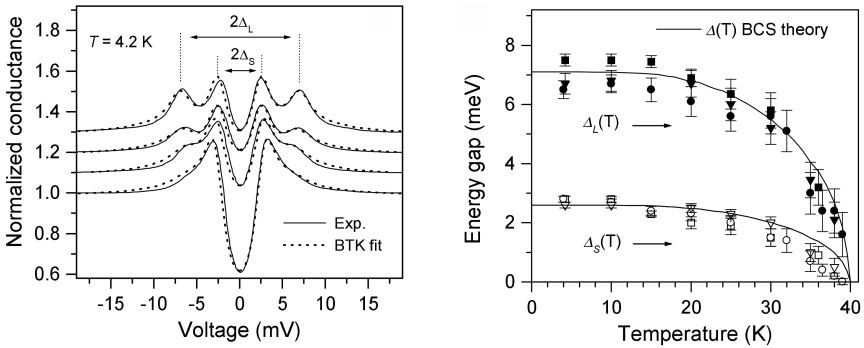

Figure from Szabó et al, Phys. Rev. Lett. 87, 137005 (2001)

Phonon softening and lifetimes in B-doped diamond

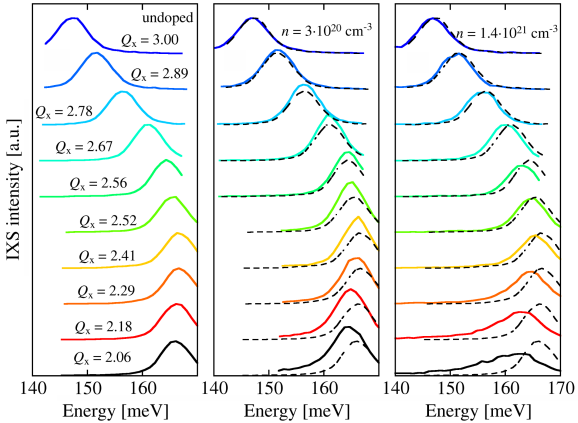

Figure from Caruso et al, Phys. Rev. Lett. 119, 017001 (2017)

Many-body Schrödinger equation for electrons and nuclei

electrons

nuclei

interaction

$$\left[-\frac{\hbar^2}{2m_e} \sum_i \nabla_i^2 + \sum_{i>j} v(\mathbf{r}_i, \mathbf{r}_j) \right.$$

$$-\frac{\hbar^2}{2M_\kappa} \sum_\kappa \nabla_\kappa^2 + \sum_{\kappa>\kappa'} Z_\kappa Z_{\kappa'} \, v(\boldsymbol{\tau}_\kappa, \boldsymbol{\tau}_{\kappa'})$$

$$\left. -\sum_{i,\kappa} Z_\kappa \, v(\mathbf{r}_i, \boldsymbol{\tau}_\kappa) \right] \Psi = E_{\text{tot}} \Psi$$

$\mathbf{r}_i$ electron

$\boldsymbol{\tau}_\kappa$ nucleus

$$v(\mathbf{r}, \mathbf{r}') = \frac{e^2}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|}$$

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- Electrons and vibrations must be described **on the same footing**

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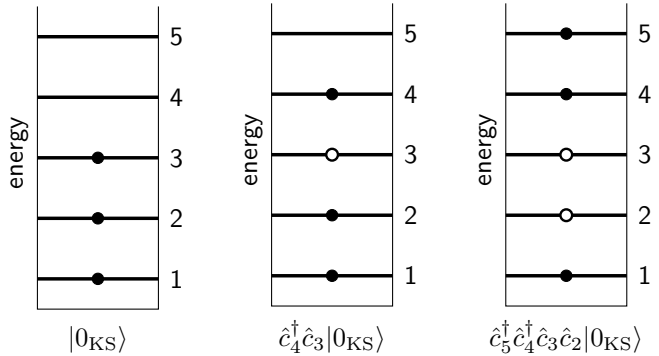
- Electrons and vibrations must be described **on the same footing**
- The many-body Schrödinger equation is **impractical** for calculations or EPIs in **solids**

Field operators

Many-electron wavefunction as a linear combination of Slater determinants

$$\Psi(\mathbf{r}_1, \mathbf{r}_2, \cdots) = A|0_{\mathrm{KS}}\rangle + \sum_{mn} B_{mn}\, \hat{c}_m^\dagger \hat{c}_n |0_{\mathrm{KS}}\rangle + \sum_{mnpq} C_{mnpq}\, \hat{c}_m^\dagger \hat{c}_n^\dagger \hat{c}_p \hat{c}_q |0_{\mathrm{KS}}\rangle + \cdots$$



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Operators in second quantization

$$\sum_i V(\mathbf{r}_i) = \sum_{mn} V_{mn}\, \hat{c}_m^\dagger \hat{c}_n$$

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Hamiltonian in field-theoretic formulation

$$\hat{H} = \hat{T}_{\mathrm{e}} + \hat{T}_{\mathrm{n}} + \hat{U}_{\mathrm{en}} + \hat{U}_{\mathrm{ee}} + \hat{U}_{\mathrm{nn}}$$

$$\hat{n}_{\mathrm{e}}(\mathbf{r}) = \hat{\psi}^{\dagger}(\mathbf{r})\hat{\psi}(\mathbf{r})$$

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Electron-nucleus interaction $$\hat{U}_{\mathrm{en}} = \int d\mathbf{r}\, d\mathbf{r}'\, \hat{n}_{\mathrm{e}}(\mathbf{r})\hat{n}_{\mathrm{n}}(\mathbf{r}')v(\mathbf{r},\mathbf{r}')$$

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Electron-electron interaction $$\hat{U}_{\mathrm{ee}} = \frac{1}{2}\int d\mathbf{r}\, d\mathbf{r}'\, \hat{n}_{\mathrm{e}}(\mathbf{r})\left[\hat{n}_{\mathrm{e}}(\mathbf{r}') - \delta(\mathbf{r}-\mathbf{r}')\right] v(\mathbf{r},\mathbf{r}')$$

Time evolution of field operators and Dyson orbitals

Ground state of N-electron system $\hat{H}|N\rangle = E_N|N\rangle$

Time evolution of field operators and Dyson orbitals

Ground state of N-electron system	$\hat{H}\|N\rangle = E_N\|N\rangle$
s-th excited state of $N+1$-electron system	$\hat{H}\|N+1,s\rangle = E_{N+1,s}\|N+1,s\rangle$

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Heisenberg time evolution of field operator	$\hat{\psi}(\mathbf{x},t) = e^{i\hat{H}t/\hbar}\,\hat{\psi}(\mathbf{x})\,e^{-i\hat{H}t/\hbar}$

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Exercise $\quad \langle N|\hat{\psi}(\mathbf{x},t)|N+1,s\rangle$

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$$= \langle N|e^{iE_N t/\hbar}\,\hat{\psi}(\mathbf{x})\,e^{-iE_{N+1,s}t/\hbar}|N+1,s\rangle$$

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Exercise

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&= \langle N|e^{iE_N t/\hbar}\,\hat{\psi}(\mathbf{x})\,e^{-iE_{N+1,s}t/\hbar}|N+1,s\rangle \\
&= \underbrace{\langle N|\,\hat{\psi}(\mathbf{x})|N+1,s\rangle}_{f_s(\mathbf{x})\ \text{Dyson orbital}}\,e^{-i\varepsilon_s t/\hbar}
\end{aligned}$$

The Green's function at zero temperature

Time-ordered Green's function

Wick's time-ordering operator

$$G(\mathbf{x}t, \mathbf{x}'t') = -\frac{i}{\hbar}\langle N|\, \hat{T}\, \hat{\psi}(\mathbf{x}t)\, \hat{\psi}^{\dagger}(\mathbf{x}'t')|N\rangle$$

The Green's function at zero temperature

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$\left|\right.$ electron in $\mathbf{x}'$ at time t' $\left.\right\rangle$

The Green's function at zero temperature

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Consider $t > t'$ (electron added to ground state)

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The Green's function at zero temperature

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$$
\begin{aligned}
G(\mathbf{x}t, \mathbf{x}'t') &= -\frac{i}{\hbar}\langle N|\, \hat{\psi}(\mathbf{x}t)\, \hat{\psi}^{\dagger}(\mathbf{x}'t')|N\rangle \\
&= -\frac{i}{\hbar}\langle N|\, e^{i\hat{H}t/\hbar}\, \hat{\psi}(\mathbf{x})\, e^{-i\hat{H}t/\hbar}\; e^{i\hat{H}t'/\hbar}\, \hat{\psi}^{\dagger}(\mathbf{x}')\, e^{-i\hat{H}t'/\hbar}|N\rangle
\end{aligned}
$$

The Green's function at zero temperature

Consider $t > t'$ (electron added to ground state)

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\begin{aligned}
G(\mathbf{x}t, \mathbf{x}'t') &= -\frac{i}{\hbar}\langle N|\, \hat{\psi}(\mathbf{x}t)\, \hat{\psi}^\dagger(\mathbf{x}'t')|N\rangle \\
&= -\frac{i}{\hbar}\langle N|\, e^{i\hat{H}t/\hbar}\, \hat{\psi}(\mathbf{x})\, e^{-i\hat{H}t/\hbar}\; e^{i\hat{H}t'/\hbar}\, \hat{\psi}^\dagger(\mathbf{x}')\, e^{-i\hat{H}t'/\hbar}|N\rangle \\
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&= -\frac{i}{\hbar}\sum_s f_s(\mathbf{x}) f_s^*(\mathbf{x}') e^{-i\varepsilon_s(t-t')/\hbar}
\end{aligned}
$$

The spectral function

Carry out the same operation for $t < t'$ and Fourier transform

$$G(\mathbf{x}, \mathbf{x}', \omega) = \sum\nolimits_s \frac{f_s(\mathbf{x}) f_s^*(\mathbf{x}')}{\hbar\omega - \varepsilon_s \mp i0^+} \qquad \mp \text{ occ/unocc}$$

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From the Green's function we can obtain the spectral (density) function

$$A(\mathbf{x}, \omega) = \frac{1}{\pi} \left| \mathrm{Im}\, G(\mathbf{x}, \mathbf{x}, \omega) \right| = \sum_s |f_s(\mathbf{x})|^2 \, \delta(\hbar\omega - \varepsilon_s)$$

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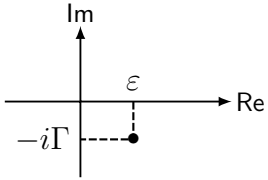
The spectral function is the many-body (local) density of states

The spectral function: Broadening

Example: a single complex pole $\varepsilon - i\Gamma$



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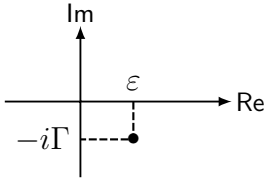

$$G(\mathbf{x},\mathbf{x},t-t') = -\frac{i}{\hbar}\left|f_s(\mathbf{x})\right|^2 e^{-i\varepsilon(t-t')/\hbar} e^{-\Gamma(t-t')/\hbar}$$

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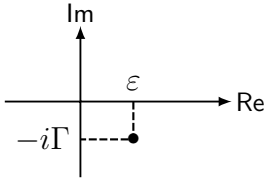

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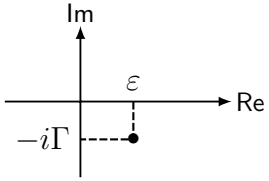

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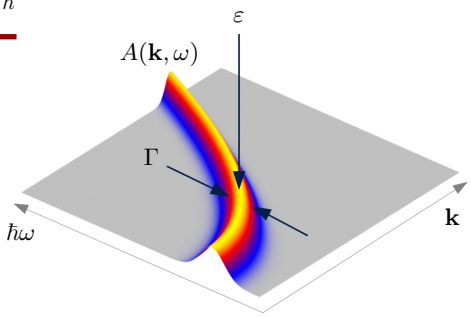

The spectral function: Coherent and incoherent structures

$$A(\mathbf{k},\omega) = \frac{1}{\pi}\,|\mathrm{Im}\,G(\mathbf{k},\omega)|$$

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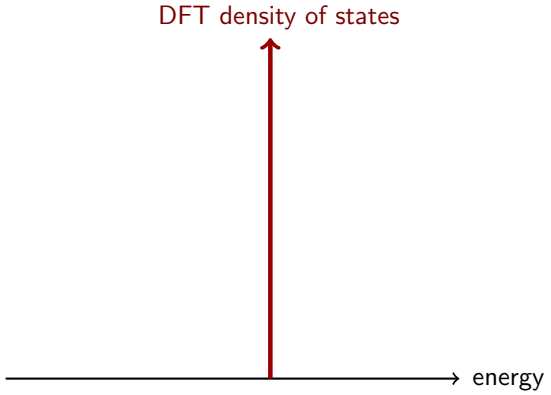

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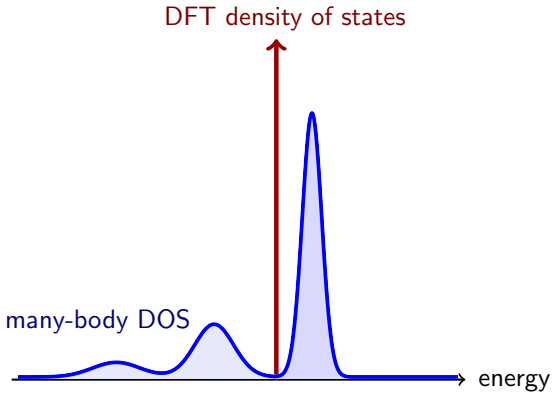

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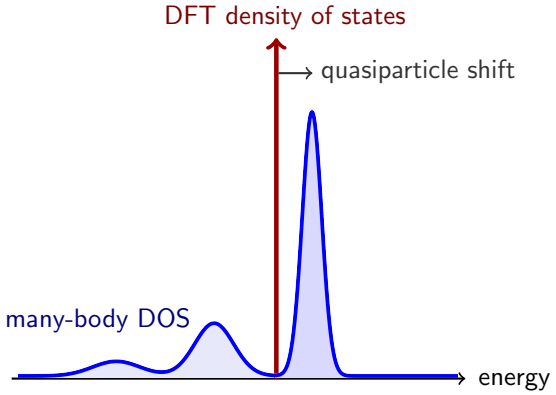

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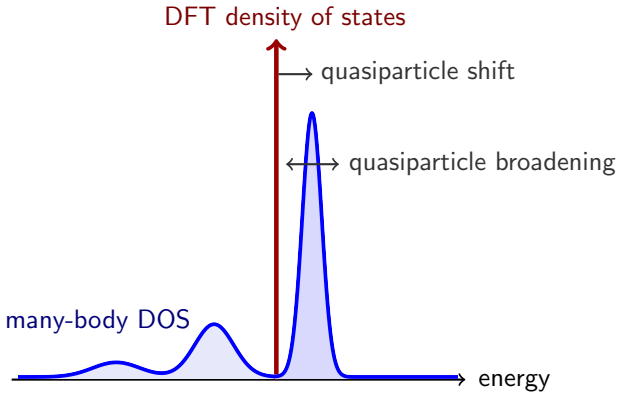

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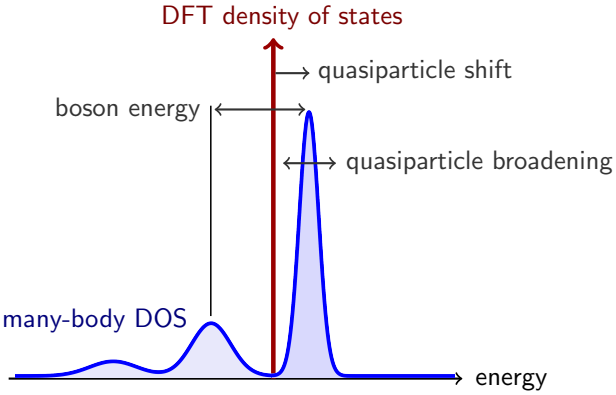

How to calculate the Green's function

Heisenberg time evolution

$$\hat{\psi}(\mathbf{x}, t) = e^{i\hat{H}t/\hbar}\, \hat{\psi}(\mathbf{x})\, e^{-i\hat{H}t/\hbar}$$

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total charge, electrons & nuclei

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Equation of motion for Green's function

$$\left[i\hbar\frac{\partial}{\partial t_1} + \frac{\hbar^2}{2m_\mathrm{e}}\nabla_1^2\right]G(12) + \frac{i}{\hbar}\int d3\, v(13)\,\langle \hat{T}\,\hat{n}(3)\,\psi(1)\,\psi^\dagger(2)\,\rangle = \delta(12)$$

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4 field operators

Nuclear potential + Hartree + Fock + 2-particle Green's function

Dyson equation

Nuclear potential + Hartree

$$V_{\rm tot}(1) = \int d2\, v(12) \langle \hat{n}(2) \rangle$$

2-particle Green's function rewritten using self-energy Σ

$$\left[i\hbar \frac{\partial}{\partial t_1} + \frac{\hbar^2}{2m_{\rm e}} \nabla_1^2 - V_{\rm tot}(1) \right] G(12) - \int d3\, \Sigma(13)\, G(32) = \delta(12)$$

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Express the Green's function in terms of Dyson's orbitals

$$\left[-\frac{\hbar^2}{2m_{\mathrm{e}}} \nabla^2 + V_{\mathrm{tot}}(\mathbf{r}) \right] f_s(\mathbf{x}) + \int d\mathbf{x}'\, \Sigma(\mathbf{x}, \mathbf{x}', \varepsilon_s/\hbar)\, f_s(\mathbf{x}') = \varepsilon_s f_s(\mathbf{x})$$

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Sources of electron-phonon interaction

Dyson equation

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Express the Green's function in terms of Dyson's orbitals

self-consistent eigenvalue

$$\left[-\frac{\hbar^2}{2m_{\rm e}} \nabla^2 + V_{\rm tot}(\mathbf{r}) \right] f_s(\mathbf{x}) + \int d\mathbf{x}'\, \Sigma(\mathbf{x}, \mathbf{x}', \varepsilon_s/\hbar)\, f_s(\mathbf{x}') = \varepsilon_s f_s(\mathbf{x})$$

Sources of electron-phonon interaction

How to calculate the electron-phonon self-energy

Electron self-energy from **Hedin-Baym**'s equations

$$\Sigma(12) = i\hbar \int d(34)\, G(13)\, \Gamma(324) W(41^+)$$

Green's function

Vertex

Screened Coulomb interaction

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Vertex

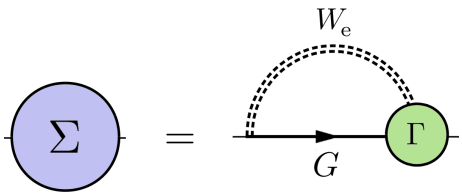
Screened Coulomb interaction

$$W = W_{\mathrm{e}} + W_{\mathrm{ph}}$$

$$W_{\mathrm{e}}(12) = \int d3\, \epsilon_{\mathrm{e}}^{-1}(13) v(32)$$

Reduces to standard GW method + **screening from nuclei**

Diagrammatic representation of the self-energy

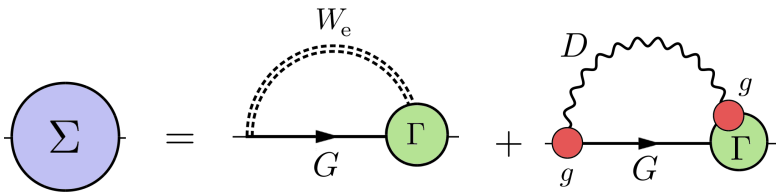


Standard GW self-energy

→ Lec. Thu.1 Louie

Figure from FG, Rev. Mod. Phys. 89, 015003 (2017)

Diagrammatic representation of the self-energy



Standard GW self-energy

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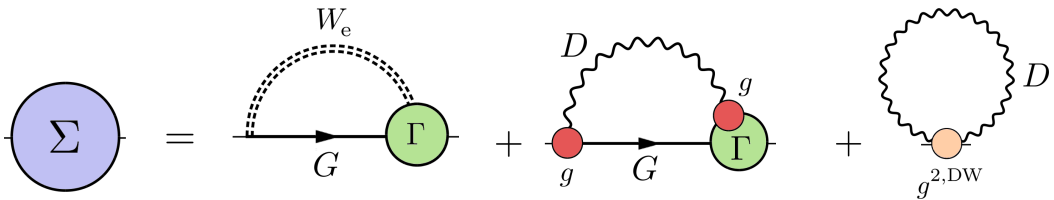

Standard GW self-energy → Lec. Thu.1 Louie

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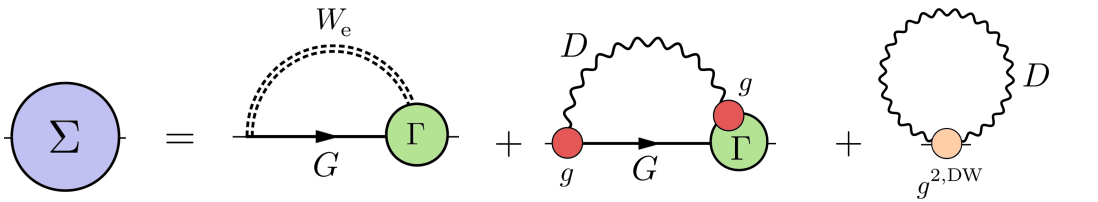

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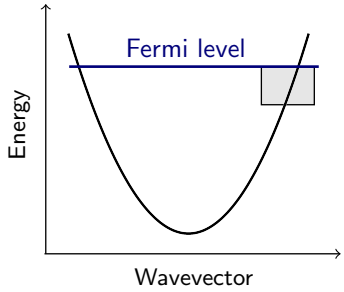
Figure from FG, Rev. Mod. Phys. 89, 015003 (2017)

Fan-Migdal self-energy using Kohn-Sham states and DFPT phonons

$$\Sigma^{\mathrm{FM}}_{n\mathbf{k}}(\omega) = \frac{1}{\hbar}\sum_{m\nu}\int \frac{d\mathbf{q}}{\Omega_{\mathrm{BZ}}} \left|g_{mn\nu}(\mathbf{k},\mathbf{q})\right|^2$$
$$\times\left[\frac{1 - f_{m\mathbf{k}+\mathbf{q}} + n_{\mathbf{q}\nu}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}}/\hbar - \omega_{\mathbf{q}\nu} + i\eta} + \frac{f_{m\mathbf{k}+\mathbf{q}} + n_{\mathbf{q}\nu}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}}/\hbar + \omega_{\mathbf{q}\nu} + i\eta}\right]$$

Example: Interaction with dispersionless phonon



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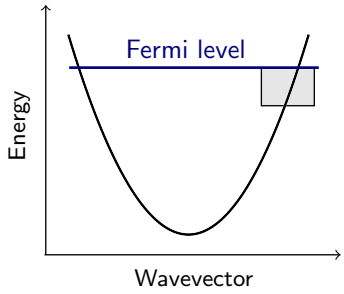

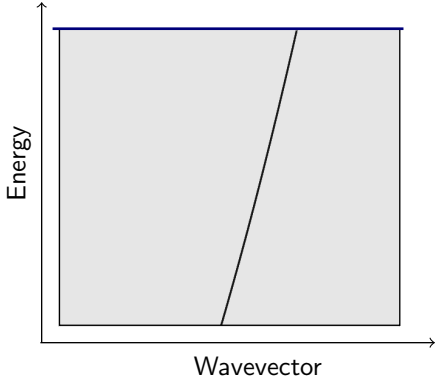

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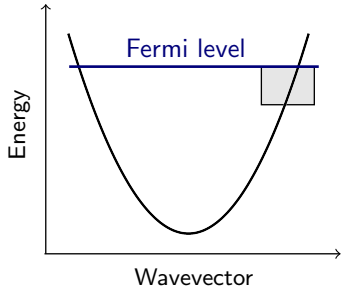
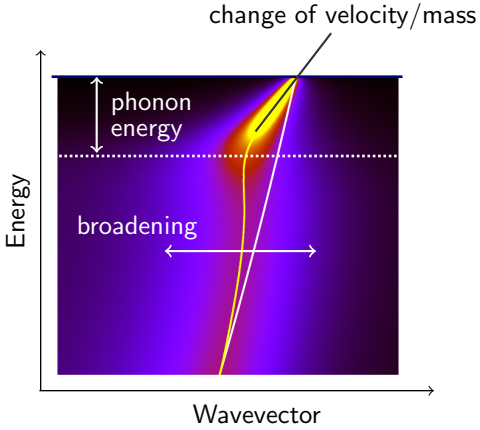
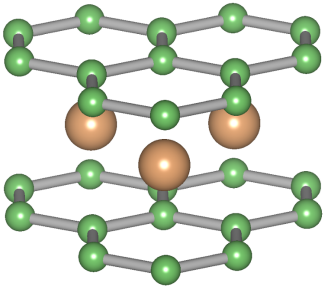




Example from experiments: Velocity renormalization in MgB_2



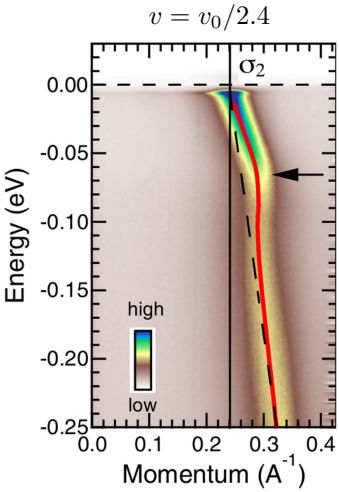


Figure from Mou et al, Phys. Rev. B 91, 140502(R) (2015)

Quasiparticle shift and broadening

Quasiparticle approximation: Approximate the Green's function as sum of comples poles

$$A(\mathbf{k},\omega) = \sum_n Z_{n\mathbf{k}} \frac{1}{\pi} \frac{\Gamma_{n\mathbf{k}}}{(\hbar\omega - E_{n\mathbf{k}})^2 + \Gamma_{n\mathbf{k}}^2}$$

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$$E_{n\mathbf{k}} = \varepsilon_{n\mathbf{k}} + \mathrm{Re}\,\Sigma_{n\mathbf{k}}(E_{n\mathbf{k}}/\hbar)$$ quasiparticle energy

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$$E_{n\mathbf{k}} = \varepsilon_{n\mathbf{k}} + \mathrm{Re}\,\Sigma_{n\mathbf{k}}(E_{n\mathbf{k}}/\hbar) \qquad \text{quasiparticle energy}$$

$$\Gamma_{n\mathbf{k}} = Z_{n\mathbf{k}}\,\mathrm{Im}\,\Sigma_{n\mathbf{k}}(E_{n\mathbf{k}}/\hbar) \qquad \text{quasiparticle broadening}$$

$$Z_{n\mathbf{k}} = \frac{1}{1 - \frac{1}{\hbar}\left.\frac{\partial \mathrm{Re}\Sigma_{n\mathbf{k}}(\omega)}{\partial\omega}\right|_{\omega=E_{n\mathbf{k}}/\hbar}} \qquad \text{quasiparticle strength}$$

The mass enhancement parameter

Take $\mathbf{k}$-Hessian of the quasiparticle energy $E_{n\mathbf{k}}$ to find mass renormalization*

$$M^*_{n\mathbf{k}} = (1 + \lambda_{n\mathbf{k}})\, m^*_{n\mathbf{k}}$$

*These expressions are for the electron gas; more complicated expressions are found in other cases

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$\lambda_{n\mathbf{k}}$ is the mass enhancement parameter

$$\lambda_{n\mathbf{k}} = -\frac{1}{\hbar} \left. \frac{\partial\, \mathrm{Re}\Sigma_{n\mathbf{k}}(\omega)}{\partial \omega} \right|_{\omega = E_{n\mathbf{k}}/\hbar}$$

*These expressions are for the electron gas; more complicated expressions are found in other cases

Electron lifetimes

→ Lec. Wed.1 Poncé

$$\tau_{n\mathbf{k}} = \frac{\hbar}{2\Gamma_{n\mathbf{k}}} = \frac{\hbar}{2\,|Z_{n\mathbf{k}}\mathrm{Im}\,\Sigma_{n\mathbf{k}}(E_{n\mathbf{k}}/\hbar)|}$$

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Common approximation: Replace $E_{n\mathbf{k}}$ by $\varepsilon_{n\mathbf{k}}$ and set $Z_{n\mathbf{k}} = 1$

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Common approximation: Replace $E_{n\mathbf{k}}$ by $\varepsilon_{n\mathbf{k}}$ and set $Z_{n\mathbf{k}} = 1$

$$\frac{1}{\tau_{n\mathbf{k}}} = \frac{2\pi}{\hbar}\sum_{m\nu}\int\frac{d\mathbf{q}}{\Omega_{\mathrm{BZ}}}|g_{nm\nu}(\mathbf{k},\mathbf{q})|^2$$
$$\times[(1 - f_{m\mathbf{k}+\mathbf{q}} + n_{\mathbf{q}\nu})\delta(\varepsilon_{n\mathbf{k}} - \hbar\omega_{\mathbf{q}\nu} - \varepsilon_{m\mathbf{k}+\mathbf{q}}) \quad \text{phonon emission}$$
$$+ (f_{m\mathbf{k}+\mathbf{q}} + n_{\mathbf{q}\nu})\delta(\varepsilon_{n\mathbf{k}} + \hbar\omega_{\mathbf{q}\nu} - \varepsilon_{m\mathbf{k}+\mathbf{q}})] \quad \text{phonon absorption}$$

Identical to Fermi golden rule formula

Example: Mass enhancement and lifetimes in $MAPbI_3$

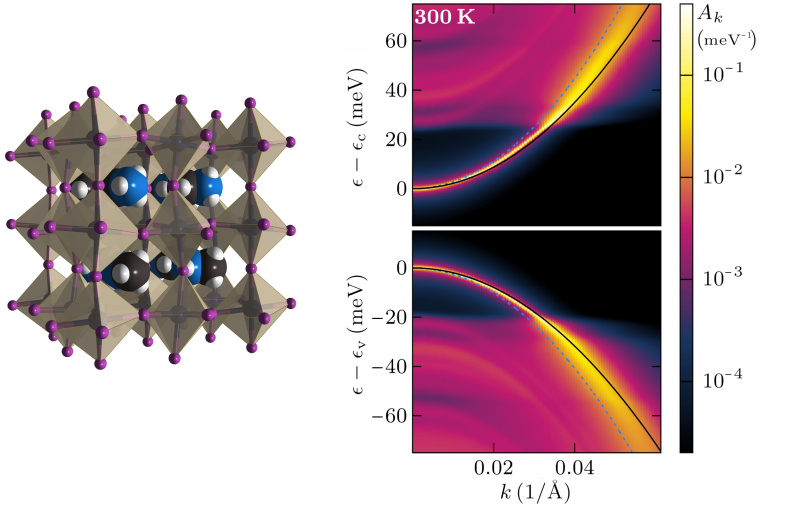
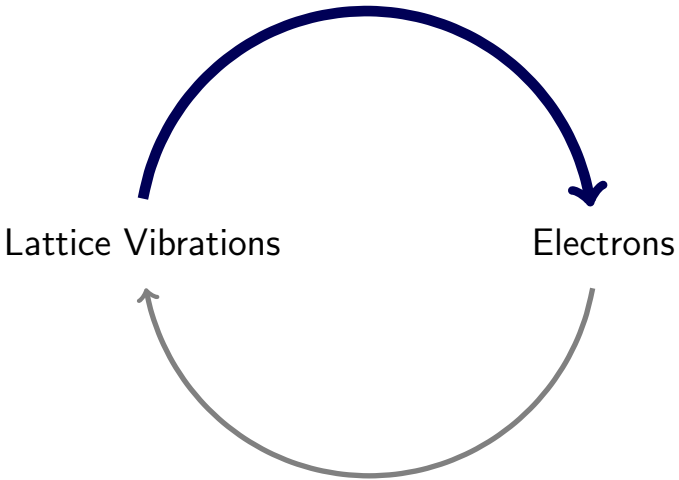


Figure adapted from Schlipf et al, Phys. Rev. Lett. 121, 086402 (2018)

Mutual interaction between electrons and vibrations



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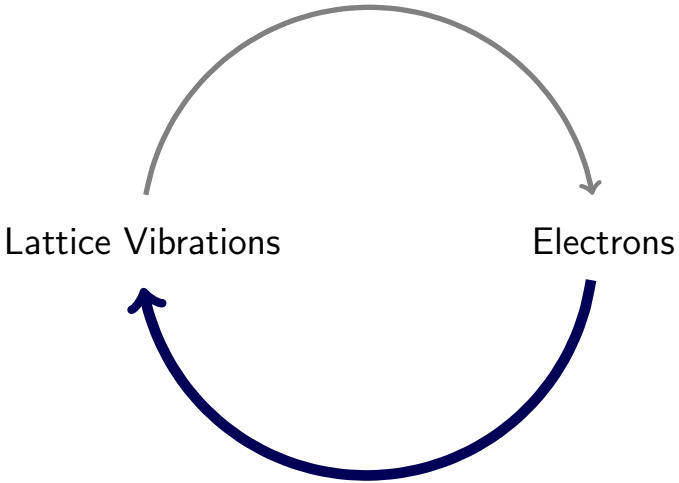

Many-body Schrödinger equation for electrons and nuclei (again)

electrons

nuclei

interaction

$$\left[-\frac{\hbar^2}{2m_e} \sum_i \nabla_i^2 + \sum_{i>j} v(\mathbf{r}_i, \mathbf{r}_j) - \frac{\hbar^2}{2M_\kappa} \sum_\kappa \nabla_\kappa^2 + \sum_{\kappa>\kappa'} Z_\kappa Z_{\kappa'} \, v(\boldsymbol{\tau}_\kappa, \boldsymbol{\tau}_{\kappa'}) - \sum_{i,\kappa} Z_\kappa \, v(\mathbf{r}_i, \boldsymbol{\tau}_\kappa) \right] \Psi = E_{\text{tot}} \Psi$$

$\mathbf{r}_i$ electron

$\boldsymbol{\tau}_\kappa$ nucleus

$$v(\mathbf{r}, \mathbf{r}') = \frac{e^2}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|}$$

Interatomic force constants in density functional perturbation theory

Approximation: Born-Oppenheimer + classical nuclei

$$E_{\rm tot} = \langle\Psi| \left[-\frac{\hbar^2}{2m_e}\sum_i \nabla_i^2 + \sum_{i>j} v(\mathbf{r}_i,\mathbf{r}_j) + \sum_{\kappa>\kappa'} Z_\kappa Z_{\kappa'}\, v(\boldsymbol{\tau}_\kappa,\boldsymbol{\tau}_{\kappa'}) \quad - \sum_{i,\kappa} Z_\kappa\, v(\mathbf{r}_i,\boldsymbol{\tau}_\kappa) \right] |\Psi\rangle$$

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Forces (Hellman-Feynman)

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Force constants

$$\frac{\partial^2 E_{\rm tot}}{\partial \boldsymbol{\tau}_\kappa \partial \boldsymbol{\tau}_{\kappa'}} = \frac{\partial^2 U_{\rm nn}}{\partial \boldsymbol{\tau}_\kappa \partial \boldsymbol{\tau}_{\kappa'}} + \int d\mathbf{r}\; \frac{\partial n_{\rm e}}{\partial \boldsymbol{\tau}_\kappa}\, \frac{\partial V_{\rm en}}{\partial \boldsymbol{\tau}_{\kappa'}} + \int d\mathbf{r}\; n_{\rm e}\, \frac{\partial^2 V_{\rm en}}{\partial \boldsymbol{\tau}_\kappa \partial \boldsymbol{\tau}_{\kappa'}}$$

→ Lec. Mon.2 Giannozzi

Phonons from a field-theoretic perspective

Key quantity to study phonons in a many-body framework:

displacement-displacement correlation function

$$D_{\kappa\alpha,\kappa'\alpha'}(tt') = -\frac{i}{\hbar}\langle N|\hat{T}\,\Delta\hat{\tau}_{\kappa\alpha}(t)\,\Delta\hat{\tau}^{\dagger}_{\kappa'\alpha'}(t')|N\rangle$$

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Heisenberg time evolution of atomic displacements

$$i\hbar\frac{d}{dt}\Delta\hat{\boldsymbol{\tau}}_{\kappa}(t) = [\Delta\hat{\boldsymbol{\tau}}_{\kappa}(t), \hat{H}]$$

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Heisenberg time evolution of atomic displacements

$$i\hbar\frac{d}{dt}\Delta\hat{\boldsymbol{\tau}}_{\kappa}(t) = [\Delta\hat{\boldsymbol{\tau}}_{\kappa}(t), \hat{H}]$$

Make it look like Newton's equation by taking 2nd derivative

$$M_{\kappa}\frac{d^2\Delta\hat{\boldsymbol{\tau}}_{\kappa}}{dt^2} = \underbrace{-\frac{M_{\kappa}}{\hbar^2}[[\Delta\hat{\boldsymbol{\tau}}_{\kappa}, \hat{H}], \hat{H}]}_{\text{dimensions of force}}$$

Many-body interatomic force constants

Equation of motion for the displacement correlation function
(using single atomic mass M for simplicity)

$$M \frac{\partial^2}{\partial t^2} \mathbf{D}(tt') =$$

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$$\Pi_{\kappa\alpha,\kappa'\alpha'}(\omega) = \frac{\partial^2}{\partial\tau_{\kappa\alpha}\,\partial\tau_{\kappa'\alpha'}}\int d\mathbf{r}\;\epsilon_{\mathrm{e}}^{-1}(\boldsymbol{\tau}_\kappa,\mathbf{r},\omega)\,\frac{e^2 Z_\kappa Z_{\kappa'}}{4\pi\epsilon_0|\mathbf{r}-\boldsymbol{\tau}_{\kappa'}|} - \text{(static force)}$$

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$\mathbf{\Pi}(\omega)$ contains the interatomic force constants resulting from the Coulomb interaction between nuclei screened by the **electronic dielectric matrix** $\epsilon_{\rm e}(\mathbf{r},\mathbf{r}',\omega)$

Many-body vibrational eigenfrequencies

Equation of motion in frequency domain

$$M\,\omega^2\,\mathbf{D}(\omega) = \mathbf{I} + \mathbf{\Pi}(\omega)\,\mathbf{D}(\omega)$$

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Solve for $\mathbf{D}$: Phonon Green's function in Cartesian coordinates

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The poles of the Green's function are the phonon eigenfrequencies:

$$\Omega(\omega) = \omega$$

where $\Omega^2(\omega)$ is an eigenvalue of $\mathbf{\Pi}(\omega)/M$

Connection with density-functional perturbation theory

$$\mathbf{\Pi}(\omega) = \frac{\partial^2}{\partial \boldsymbol{\tau}_\kappa \partial \boldsymbol{\tau}_{\kappa'}} \int d\mathbf{r}\ \epsilon_{\mathrm{e}}^{-1}(\boldsymbol{\tau}_\kappa, \mathbf{r}, \omega)\, \frac{e^2 Z_\kappa Z_{\kappa'}}{4\pi\epsilon_0 |\mathbf{r} - \boldsymbol{\tau}_{\kappa'}|} - \text{(static force)}$$

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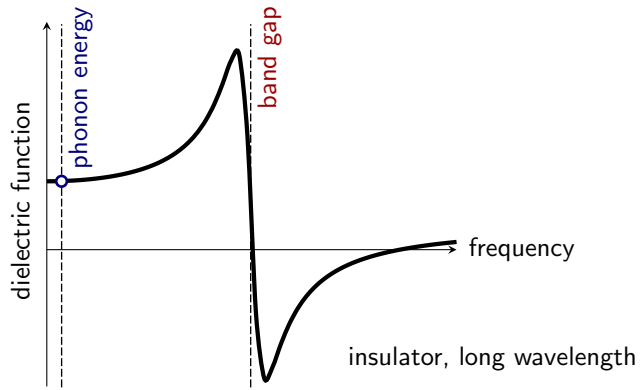

Connection with density-functional perturbation theory

We call **adiabatic** self-energy the $\mathbf{\Pi}$ evaluated using the **static** screening

(phonon period so long that electrons have time to complete relax after an atomic displacement)

$$\mathbf{\Pi}^{\mathrm{A}} \overset{\mathrm{def}}{=} \mathbf{\Pi}\left(\omega = 0\right)$$

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After rearranging:

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replace with DFT electron density

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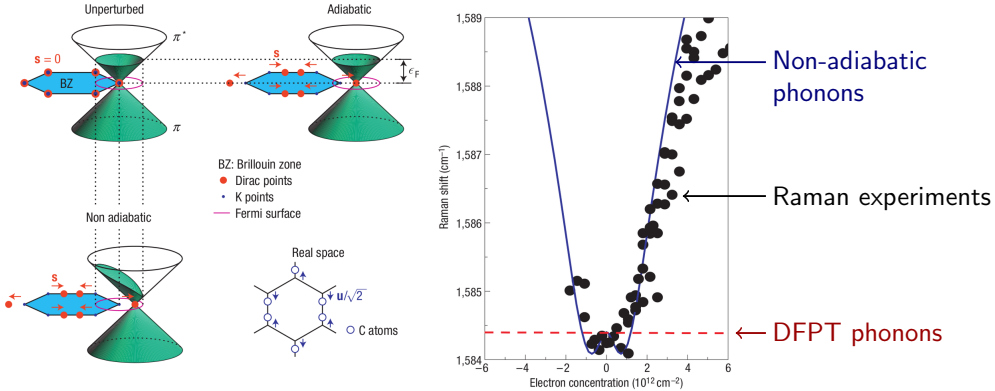
replace with DFT electron density

$$\mathbf{\Pi}^{\mathrm{A}} = \frac{\partial^2 E_{\mathrm{tot}}}{\partial \boldsymbol{\tau}_\kappa\, \partial \boldsymbol{\tau}_{\kappa'}}$$

Same as DFPT in slide 29

Phonons beyond the Born-Oppenheimer approximation

Example: Non-adiabatic Kohn-anomaly in graphene



Figures from Pisana et al, Nat. Mater. 6, 198 (2007)

Take-home messages

- Quantum field theory provides a rigorous framework to study electron-phonon physics
- The Fan-Migdal self-energy yields electron mass enhancement and electron lifetimes
- Phonon frequencies are deeply connected with the electronic dielectric matrix
- The many-body framework reduces to DFPT under suitable approximations

Further reading

- FG, Rev. Mod. Phys. 89, 015003 (2017) [link]
- Stefanucci, van Leeuwen, Perfetto, Phys. Rev. X 13, 031026 (2023) [Link]
  generalization of this lecture to out-of-equilibrium case
- Baym, Ann. Phys. 14, 1 (1961) [Link]
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- Grimvall, The electron-phonon interaction in metals, 1981 (North-Holland)