Physique des Matériaux et Nanostructures

10th International ABINIT developer workshop
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Cumulant expansion

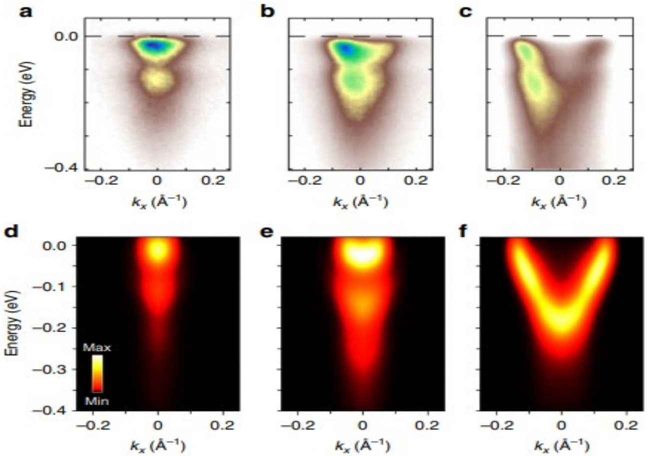
Joao Abreu[1], Matteo Giantomassi[2], Matthieu Verstraete[1]

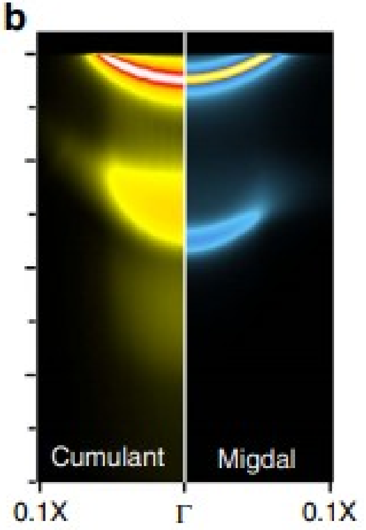
[1] nanomat/QMAT/CESAM and Physics Department, University of Liège
[2] UCLouvain, Institute of Condensed Matter and Nanosciences (IMCN)

Summary

Motivation

TiO2





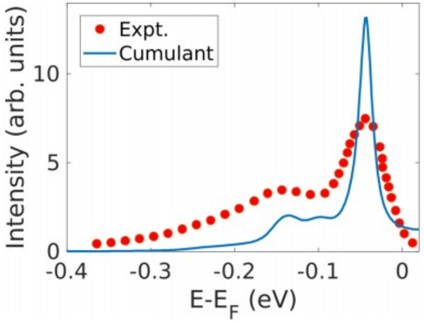
General Properties

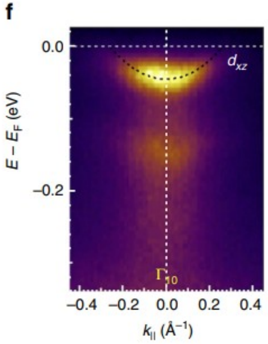
- Phonon limits electron mobility
- Temperature dependent band structures
- Zero-point renormalization of the band gap
- Thermal and electrical conductivities

- Polaron binding energy
- QP Broadening

Cumulant Expansion

SrTiO3





Many-Body Perturbation Theory

The Green's Function is a mathematical tool
to deal with particle interactions when including excitations

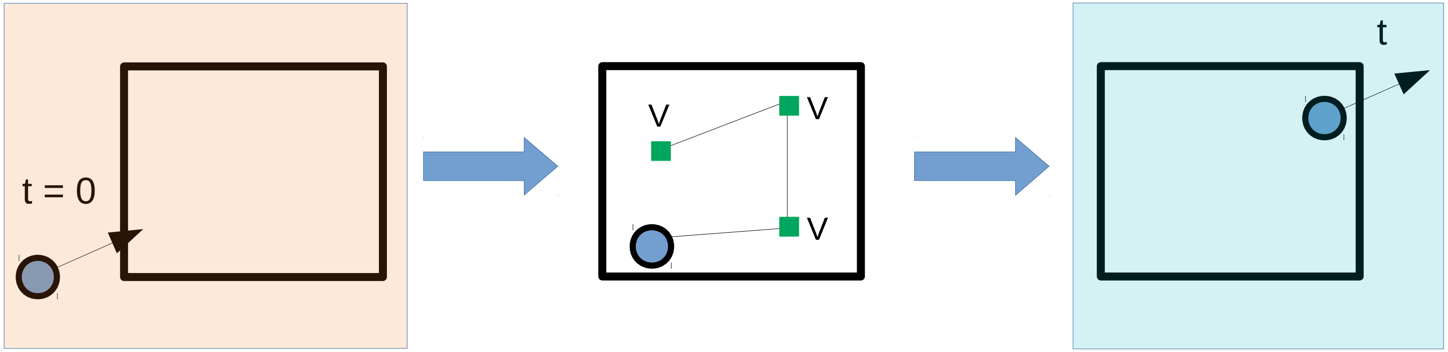
$$H_0 = \sum_k \varepsilon_0 c_k^+ c_k + \sum_q \omega_q a_q^+ a_q$$

$$V = \sum_{kq} g_{kq} c_k^+ c_k \left(a_q^+ + a_q\right)$$

Retarded Green's Function

$$H = H_0 + V$$

$$G_k(t) = -i\,\theta(t) \left\langle c_k(t)\, c_k^+(0) \left[1 + \Pi_i \int_0^t dt_i\, V(t_i)\right] \right\rangle$$



Interacting Green's Function

$$G_k(t) = -i\theta(t)\left\langle c_k(t)c_k^+(0)\left[1+\Pi_i \int_0^t dt_i V(t_i)\right]\right\rangle$$

Dyson Fan-Migdal

$$G_k = G_k^0 + G_k^0 \Sigma_k G_k$$

$$G_k = \frac{1}{\left(G_k^0\right)^{-1} + \Sigma_k}$$

Cumulant expansion

$$G_k = G_k^0 e^{C(t)}$$

$$C(t) = FT\, G_k^0 \Sigma G_k^0$$

Higher orders of interaction →

$$C(t) = \int d\omega \frac{1}{\pi} \left|\Im m \Sigma_k(\omega)\right| \frac{e^{i\omega t} + i\omega t - 1}{\omega^2}$$

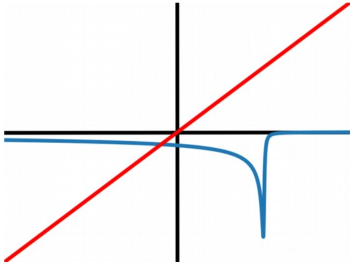
One satellite (Frohlich)
Wrong polaron binding energy
Low broadening at high T

$$A_k(\omega) = \Im m G_k(\omega)$$

Generates satellites (Frohlich)
Shift Quasi-particle peak
Renormalizes Quasi-particle

Energy renormalization

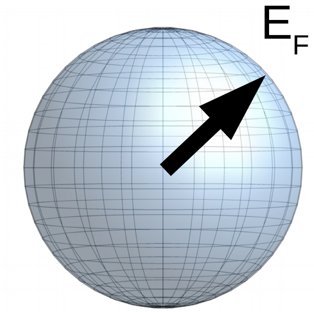
Self-consistent: $$\varepsilon^{SC} = \varepsilon^{KS} + \Re\,\Sigma\left(\varepsilon^{SC}\right)$$



Linear approximation: $$\varepsilon^{Linear} = \varepsilon^{KS} + Z\,\Re\,\Sigma\left(\varepsilon^{KS}\right)$$

$$Z = \left(1 - \left.\frac{\partial\,\Sigma(\omega)}{\partial\,\omega}\right|_{\omega=\varepsilon^{KS}}\right)^{-1}$$

On-the-mass-shell: $$\varepsilon^{OMS} = \varepsilon^{KS} + \Re\,\Sigma\left(\varepsilon^{KS}\right)$$

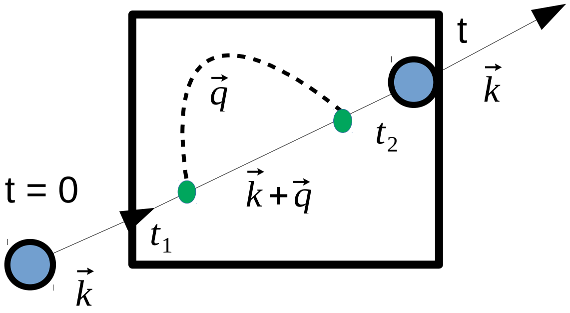


Self-Energy

$k = n\vec{k}$

$q = j\vec{q}$

Fan-Migdal

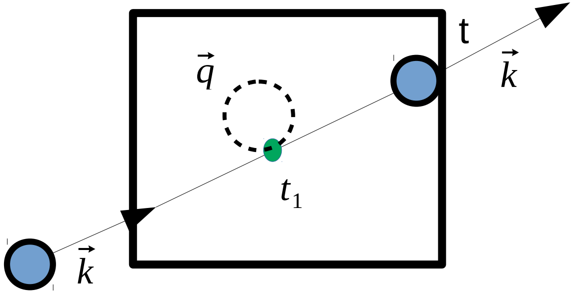


$$\Sigma_k^{FM}(\omega) = i\int \frac{d\omega'}{2\pi} \sum_{mq} |g_{mkq}|^2 D_q^0(\omega') G^0_{m\vec{k}+\vec{q}}(\omega-\omega')$$

$$= \frac{1}{N_q} \sum_{mq}^{BZ} |g_{mkq}|^2 \times \left(\frac{n_q + 1 - f_{m\vec{k}+\vec{q}}}{\omega - \varepsilon_{m\vec{k}+\vec{q}} - \omega_q + i\eta} + \frac{n_q + f_{m\vec{k}+\vec{q}}}{\omega - \varepsilon_{m\vec{k}+\vec{q}} + \omega_q + i\eta} \right)$$

- Dynamic
- Singularities occur at electronic ± phonon energies

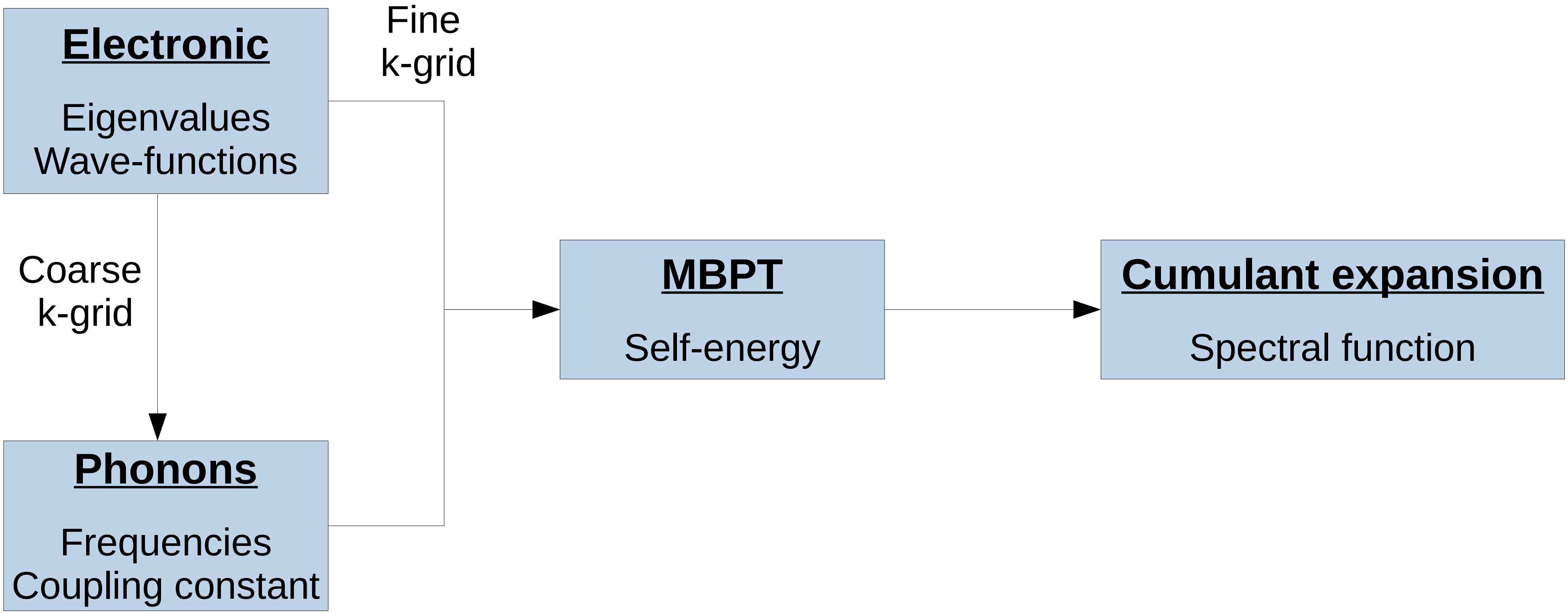
Debye-Waller



$$\Sigma_k^{DW} = i\int \frac{d\omega'}{2\pi} \sum_{mq} |g^{DW}_{mkq}|^2 \frac{2n_q + 1}{\varepsilon_k - \varepsilon_{m\vec{k}}}$$

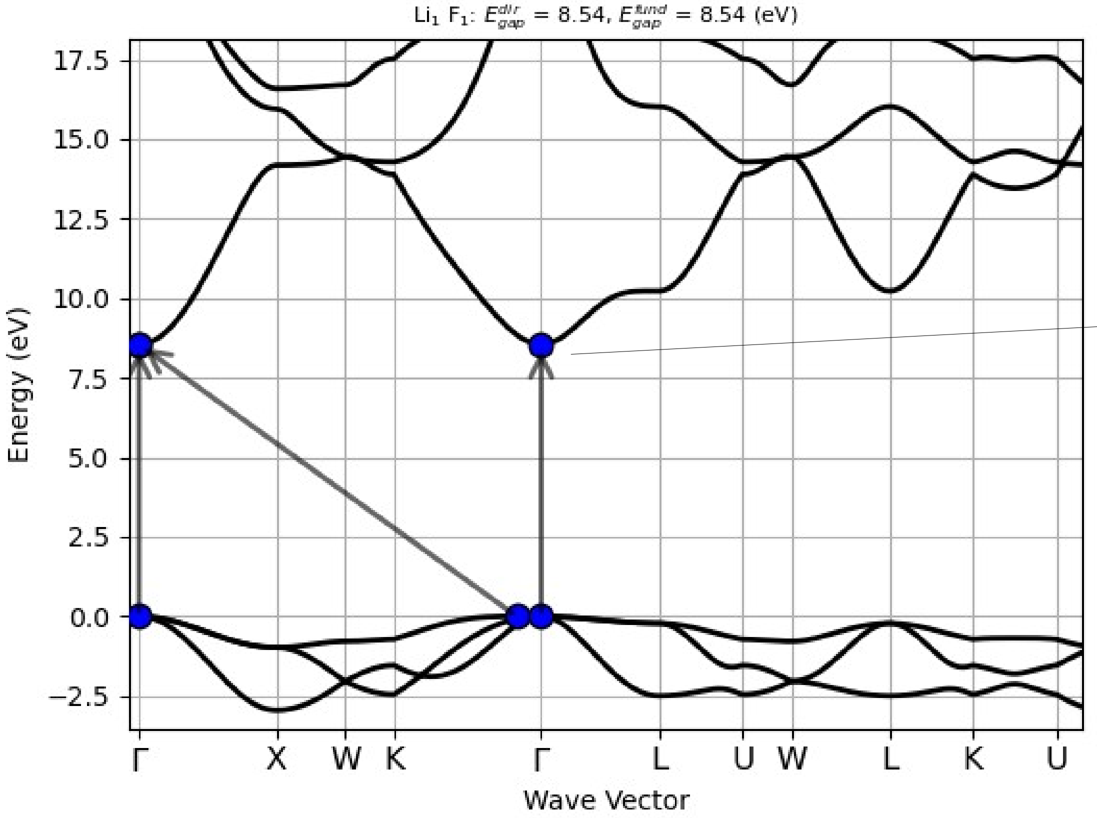
- Static
- Increases with number of phonons

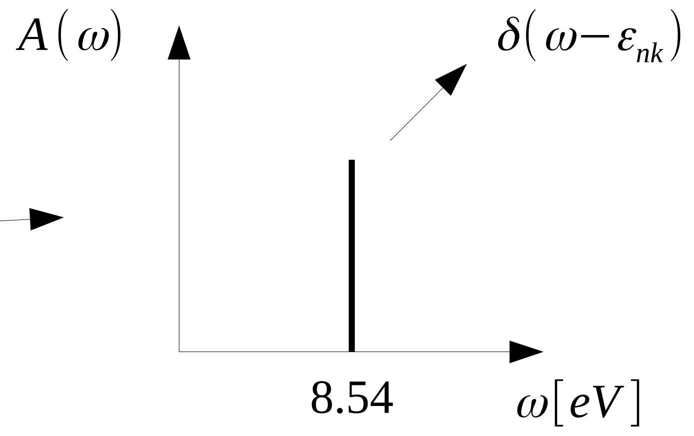
Calculations



Electronic Band Structure

LiF

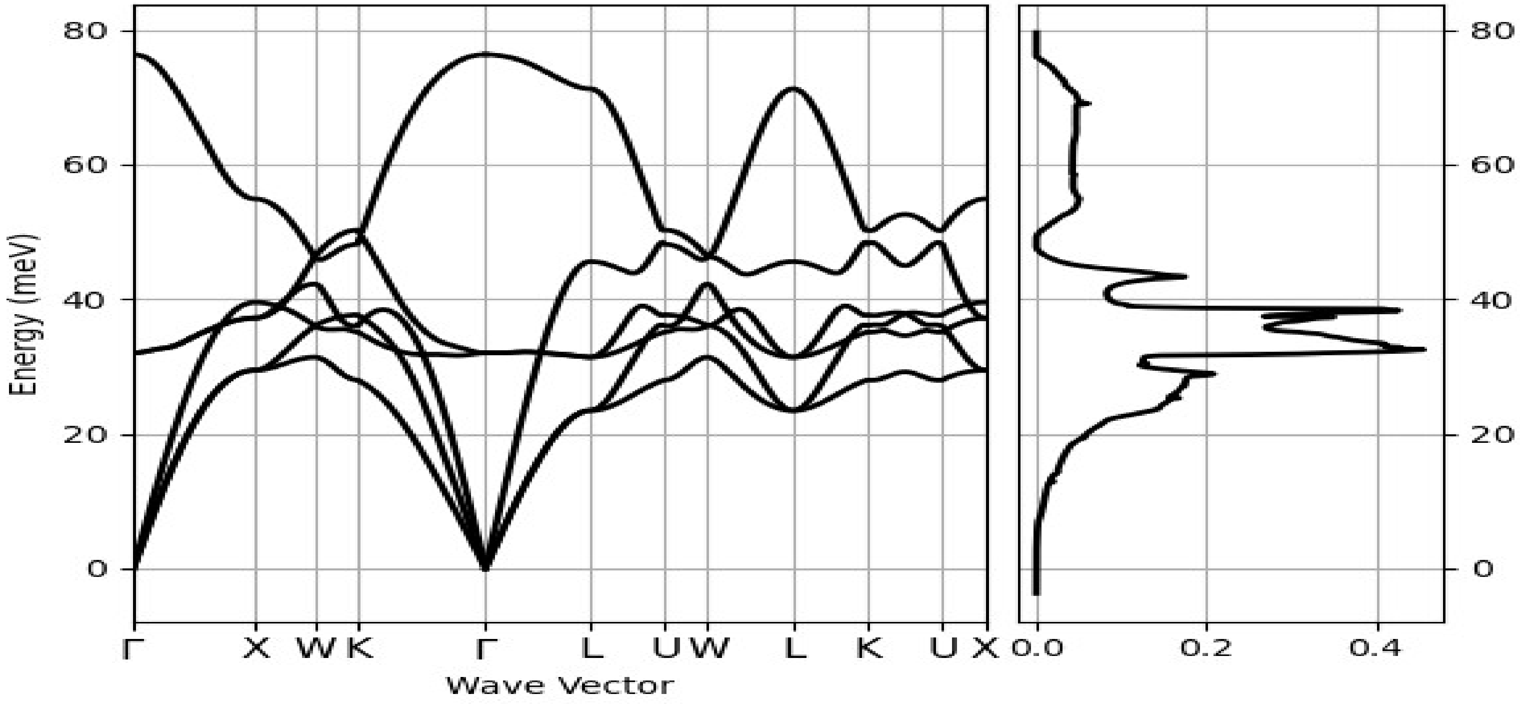




- Phonons → # k-points = # q-points
- EPH → dense k- and q- points

Phonon band structure

LiF – 8x8x8 q-point grid



EPH calculations

Self-energy

$$\Sigma_k^{FM}(\omega) = \frac{1}{N_q} \sum_m \sum_q^{BZ} |g_{mkq}|^2 \times \left(\frac{n_q + 1 - f_{m\vec{k}+\vec{q}}}{\omega - \varepsilon_{m\vec{k}+\vec{q}} - \omega_q + i\eta} + \frac{n_q + f_{m\vec{k}+\vec{q}}}{\omega - \varepsilon_{m\vec{k}+\vec{q}} + \omega_q + i\eta} \right)$$

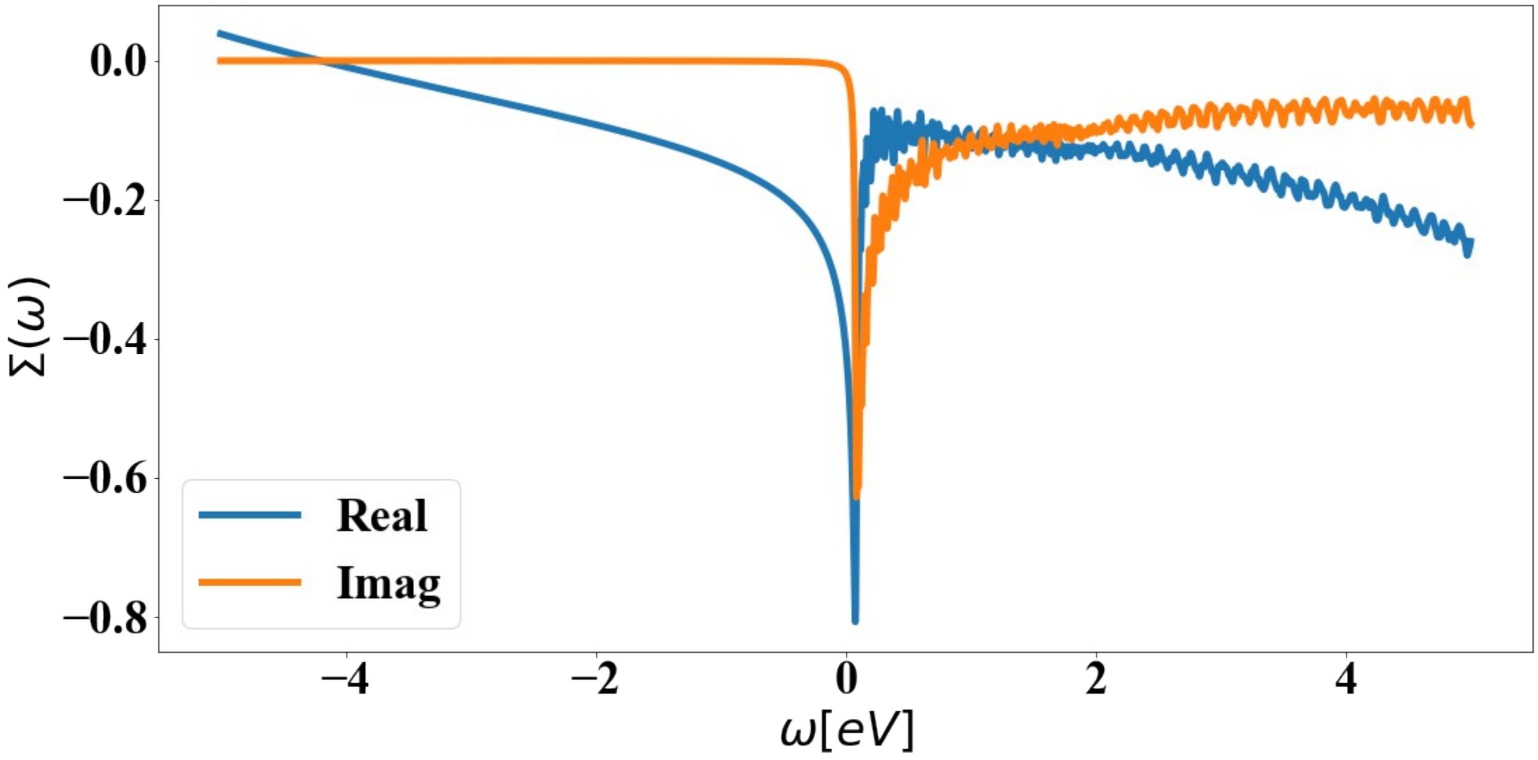
Important input variables

- **eph_task** = 4 (self-energy calculations)
- **eph_stern** = sternheimer
- **eph_ngqpt_fine** = interpolation
- **zcut** = infinitesimal number (below ω_{LO})

- **nfreqsp** = number frequency domain
- **freqspmax** = max frequency
- **freqspmin** = min frequency

Self-Energy

LiF



Convergence

Zcut = 0.01 eV
 # Bands = 10 (stern)
K and q = 96x96x96

Polar material

Peak at phonon ω_{LO}

Implementation of the Cumulant Expansion

$$C_k(t)=\int d\omega \frac{1}{\pi}\left|\Im m \Sigma_k^{FM}(\omega)\right| \frac{e^{i\omega t}+i\omega t-1}{\omega^2}$$

$$G_k(t)=-i\theta(t) e^{i(\varepsilon_k^{KS}+\Sigma_k^{DW})t} e^{C_k(t)}$$

$$G_k(\omega)=\int dt\, e^{i\omega t} G_k(t)$$

$$A_k(\omega)=-\frac{1}{\pi}\Im G_k(\omega)$$

Debug

Important input variables

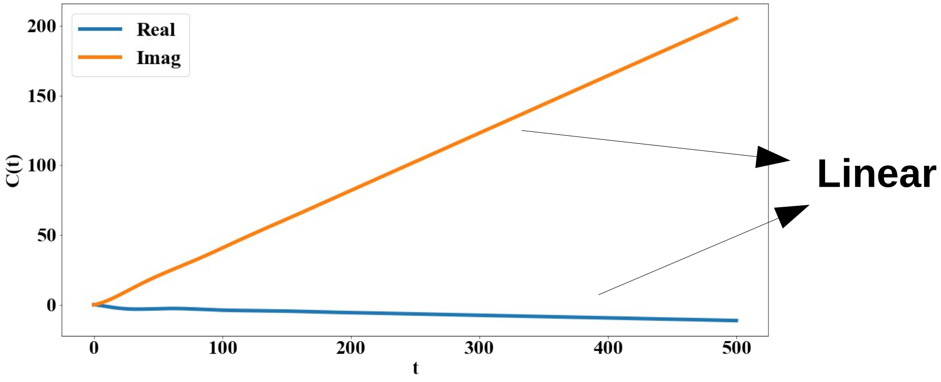
- **eph_task** = 9 (cumulant calculations)
- **tolcum** = Finding tmax

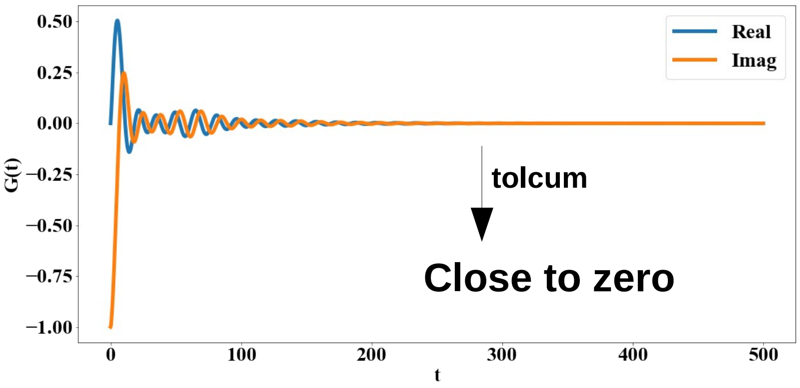
Kramers-Kroning relation

$$\Re e \Sigma_k^{FM}(\varepsilon^{KS})=-P\int_{-\infty}^{\infty} d\omega \frac{1}{\pi} \frac{\left|\Im m \Sigma_k^{FM}(\omega)\right|}{\omega}$$

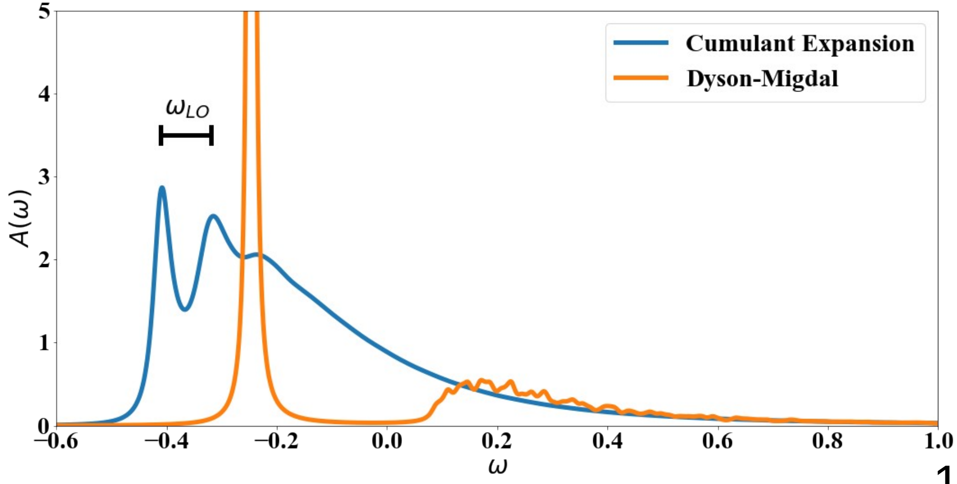
Langreth, Physical Review B 1:471 (1969)
Gumhalter, Physical Review B 72:165406 (2005)
Kubo, Journal of the Physical Society of Japan, 17(7):1100 (1962)
Nery et al, Physical Review B, 97:115145 (2018)

Cumulant Expansion





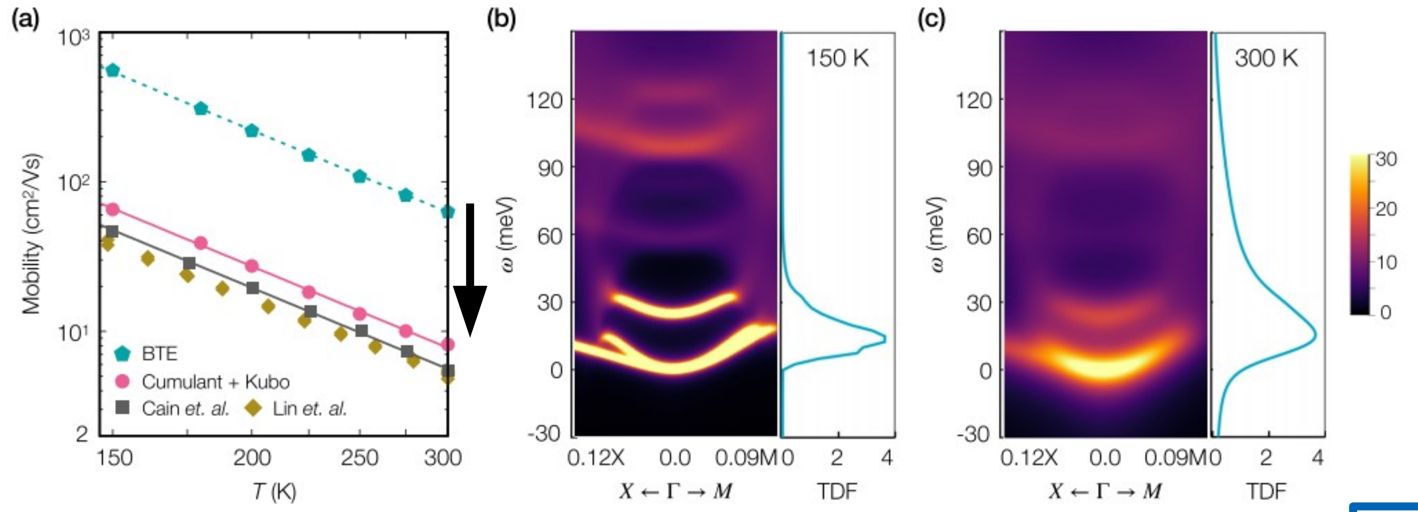
- **C(t)** - Linear behaviour at long time
  ~ π Im $\Sigma(\varepsilon^{KS})$ t
- **G(t)** - Damping
  ~ exp(- π Im $\Sigma(\varepsilon^{KS})$ t)
- **A(w)**
  - Correction of the polaron binding energy
  - Renormalization of the energy



Work in progress:

Kubo-Greenwood formalism:

$$\sigma_{\alpha\beta}(\omega)=\frac{\pi}{\Omega}\int d\omega'\frac{f(\omega')-f(\omega'+\omega)}{\omega}\sum_{n\vec{k}} v^{\alpha}_{n\vec{k}} v^{\beta}_{n\vec{k}} A_{n\vec{k}}(\omega') A_{n\vec{k}}(\omega'+\omega)$$

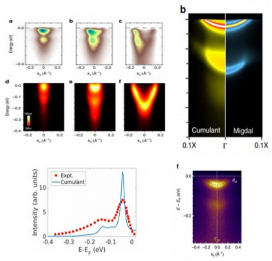


- QP broadening increases
- Life-time decreases
- Mobility decreases

SrTiO3

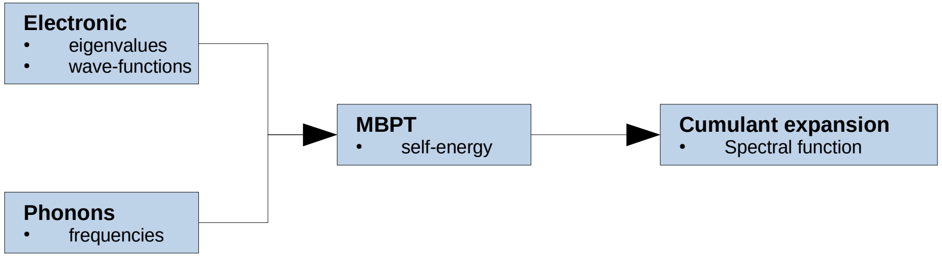
J.-J. Zhou, M. Bernardi, Physical Review Research 1, 033138 (2019)

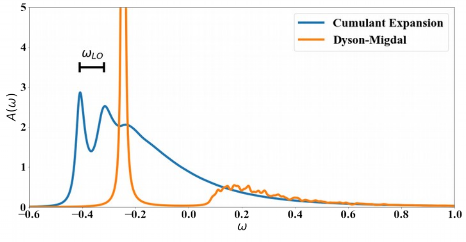
Summary



Experimental measurements comparable with cumulant expansion

Calculations to be able to calculate cumulant expansion





Accurate spectral function description

Going beyond Boltzmann transport equation