Break-down of the self-consistent Born approximation for LA+LO phonon interactions in quantum dots - An absorption spectra study

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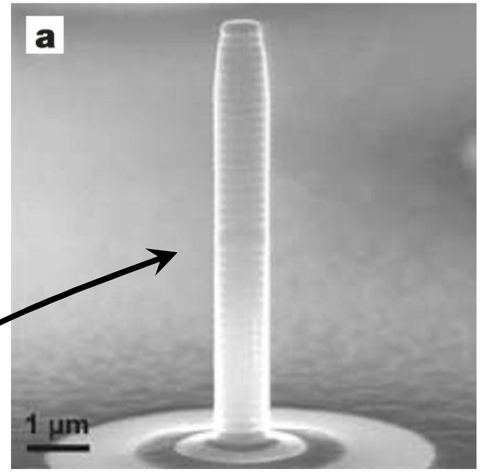
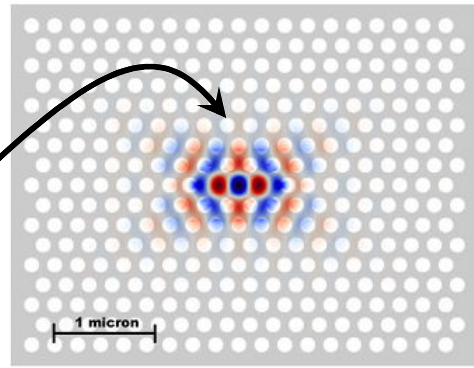
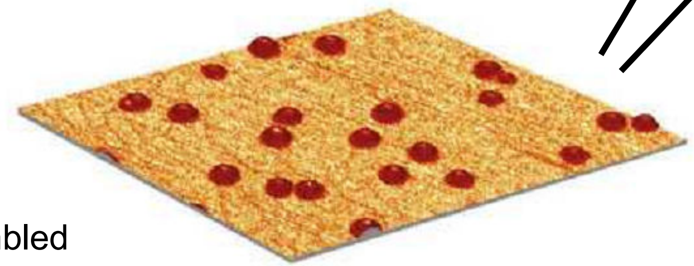
Solid state quantum optics

- Great potential within various quantum technologies
  - Quantum computing
  - Quantum cryptography
  - ….
- Why solid state?
  - Scalability
  - Control over material properties through design
- Single-photon sources
  - Only one!
  - On-demand
  - Indistinguishability
- Challenges
  - Coherence times are short
  - Theoretical description is difficult due to the inherent many-body environment
  - ….



Optical microcavities



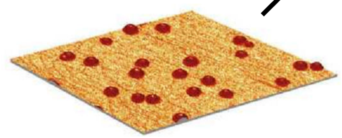


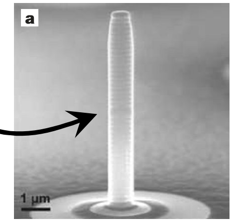
Self-assembled
quantum dots

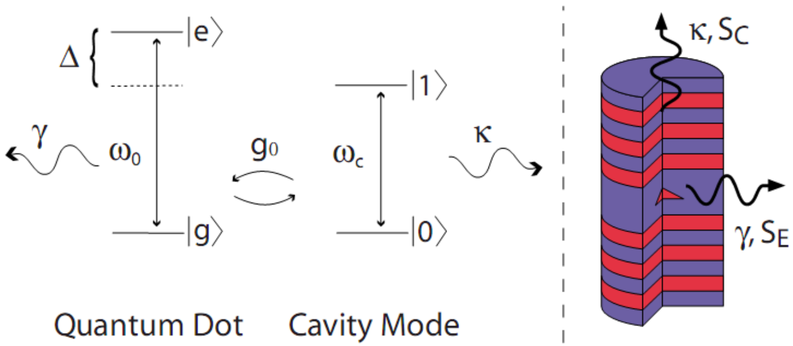
Experiments

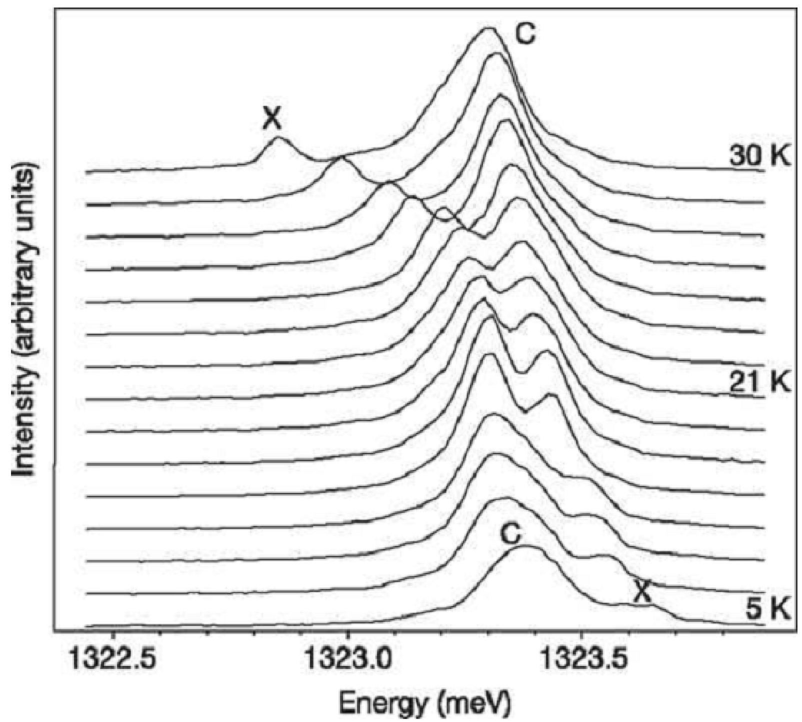
- What is measured = what do we need to calculate
  - Emission spectra

$$S(\boldsymbol{R},\omega_S) \propto \int_{-\infty}^{+\infty} dt' \int_{-\infty}^{+\infty} dt e^{-i\omega_S(t'-t)} \langle E^{(-)}(\boldsymbol{R},t')E^{(+)}(\boldsymbol{R},t)\rangle$$









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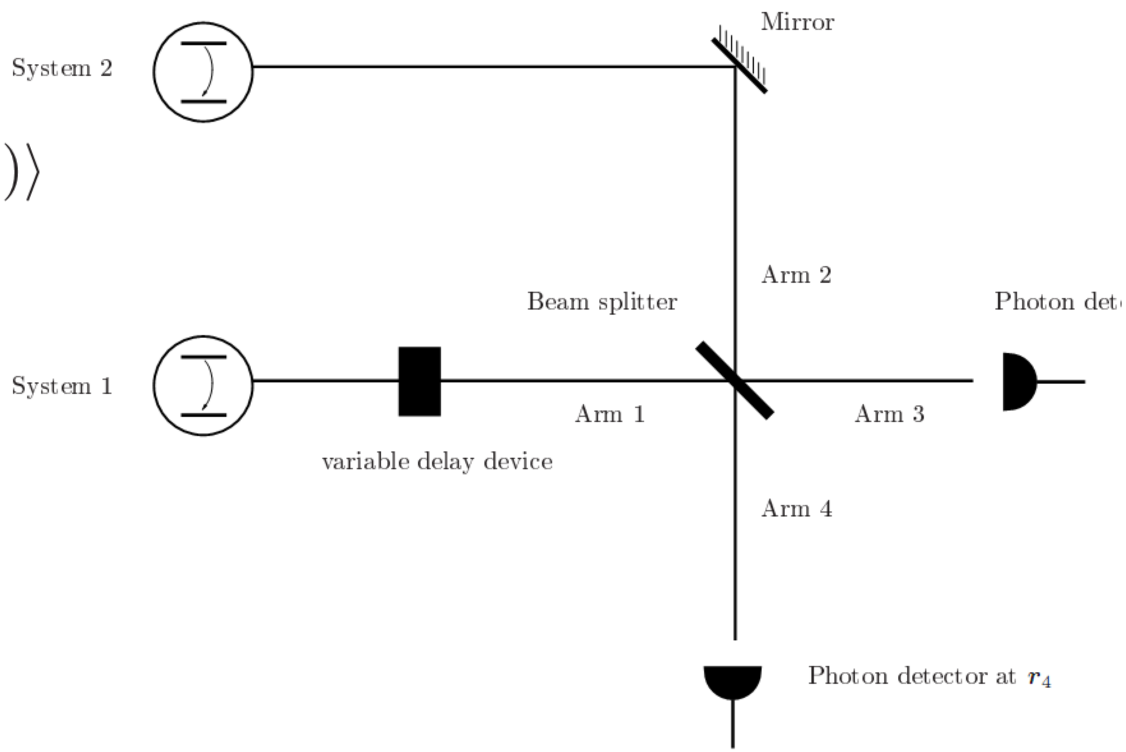
  - Two-photon interference

$$G_{34}^{(2)}(t_3,t_4) = \langle a^\dagger(t_3)a(t_3)\rangle \langle a^\dagger(t_4)a(t_4)\rangle$$
$$- \langle a^\dagger(t_3)a(t_4)\rangle \langle a^\dagger(t_4)a(t_3)\rangle$$
$$+ \langle a^\dagger(t_3)a^\dagger(t_4)a(t_4)a(t_3)\rangle$$



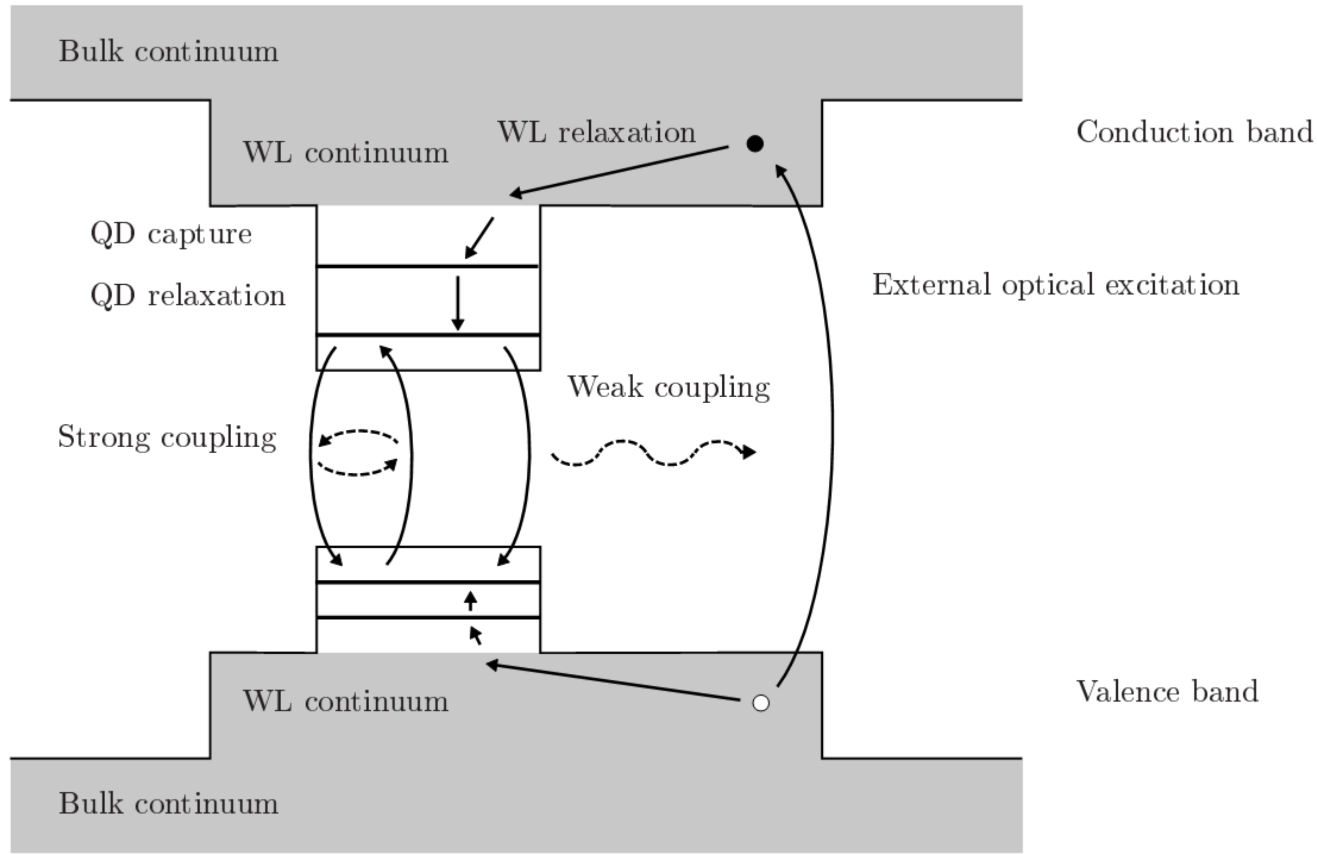
- The full two-time GF is essential
  - Not “only” a computational device for the one-time GF

Outline

- Motivation for using NEGF
- Model physical system
- NEGF theory
- Numerics
- Results and discussion
- Summary

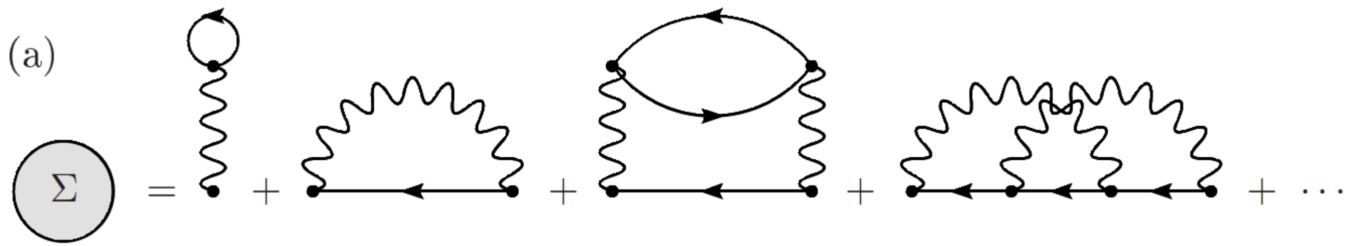
Several interactions

- Typical quantum optical experiment
  - Many interactions are involved
  - All are important
- Formalism needs to be able to handle all of these
- This talk will focus on the phonon interaction

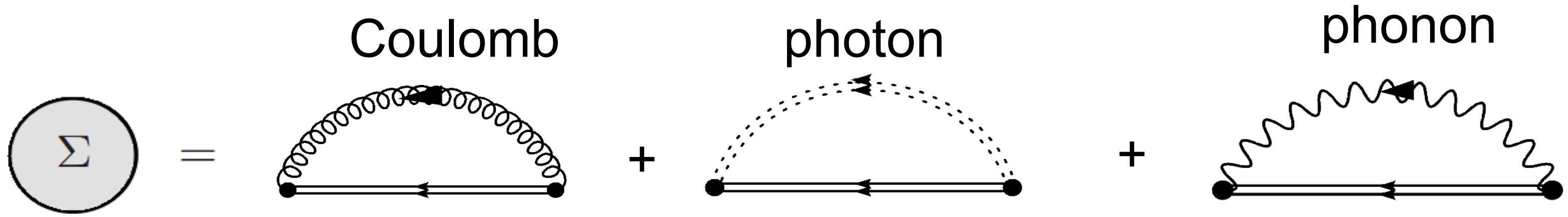


(My) Motivation for using NEGF

- Provides a systematic and consistent framework for many-body problems
  - Systematic in the sense that it is a possible to incrementally improve your theory



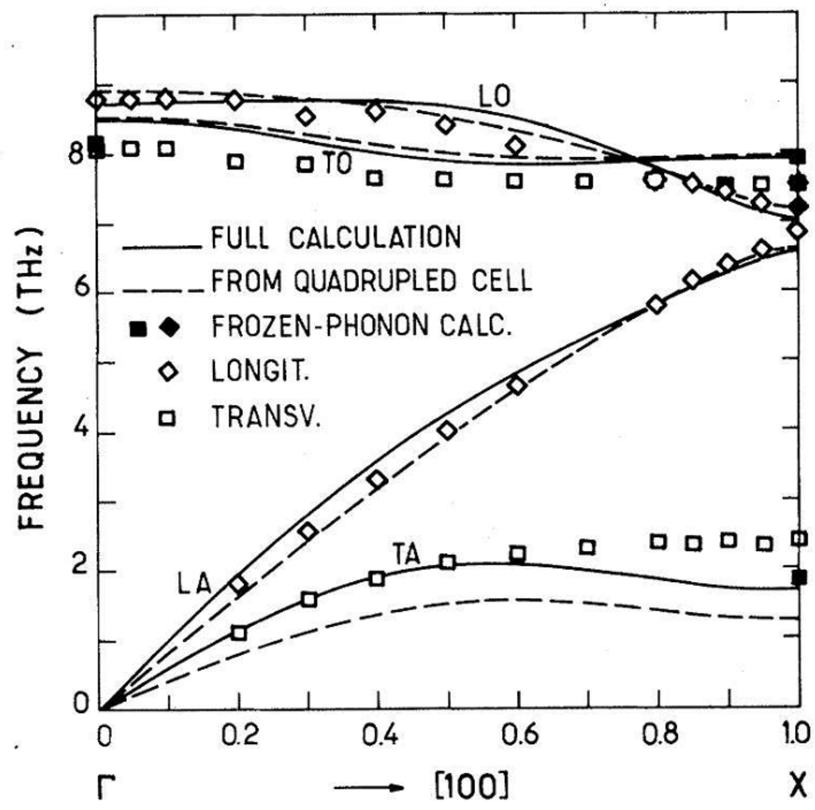
  - Easy inclusion of multiple interaction mechanisms.
    - Simply add standard self-energies that are known to work for the specific interaction



- "The cookbook" for NEGF should be readily applicable

Focus of this talk

- Focus on electronic system and phonons
  - The two most important branches in III-V semiconductors
  - LO = longitudinal optical
    - “Dispersionless”
  - LA = longitudinal acoustical
    - Linear dispersion
- The combined LO+LA interaction has yielded the greatest surprise



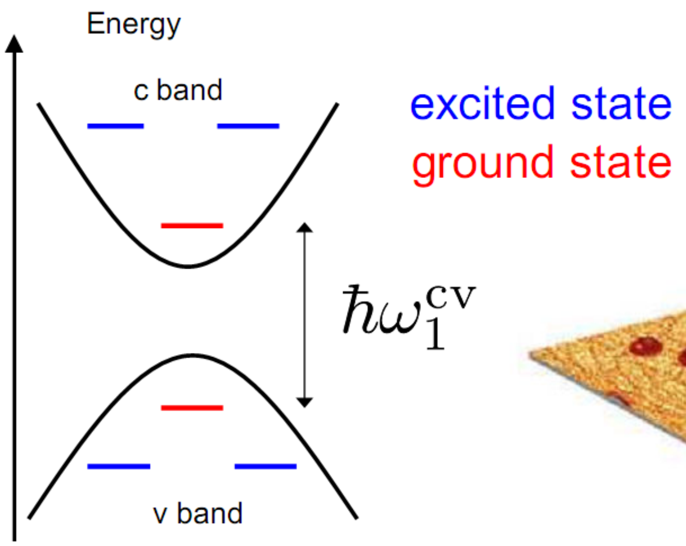
Phonon band diagram
for bulk GaAs

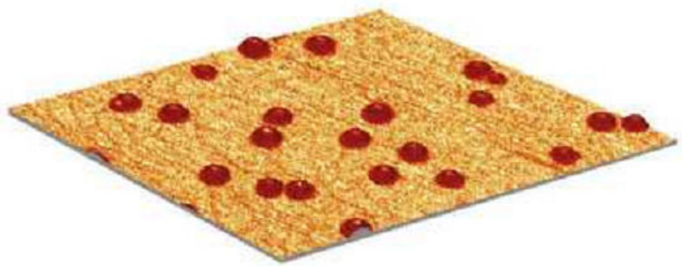
K. Kunc and R. M. Martin, Phys. Rev. Lett. **48**, 406 (1982).

Model of physical system

- Electronic system
  - Confined level modeled as harmonic oscillator states
  - No quasi-continuum included
    - Partly justified by low temperature and focus on confined states

QD level structure





- Phononic system
  - Bulk modes
  - LO phonons: Frohlich Hamiltonian

$$H_{\mathrm{e-LO}} = \sum_{\nu\nu'\boldsymbol{q}} M_{\nu\nu'}^{\boldsymbol{q}} c_\nu^\dagger c_{\nu'} (b_{-\boldsymbol{q}}^\dagger + b_{\boldsymbol{q}}), \quad M_{\nu\nu'}^{\boldsymbol{q}} = \frac{M}{qV^{\frac{1}{2}}} \langle \nu | e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} | \nu' \rangle$$

  - LA phonons: Deformation potential interaction

$$M_{\nu\nu'}^{\boldsymbol{q}} = D_{\nu\nu'} \frac{1}{V^{\frac{1}{2}}} \left(\frac{\hbar}{2\rho_{\mathrm{den}} c_{\mathrm{s}}} \right)^{\frac{1}{2}} q^{\frac{1}{2}} \langle \nu | e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} | \nu' \rangle$$

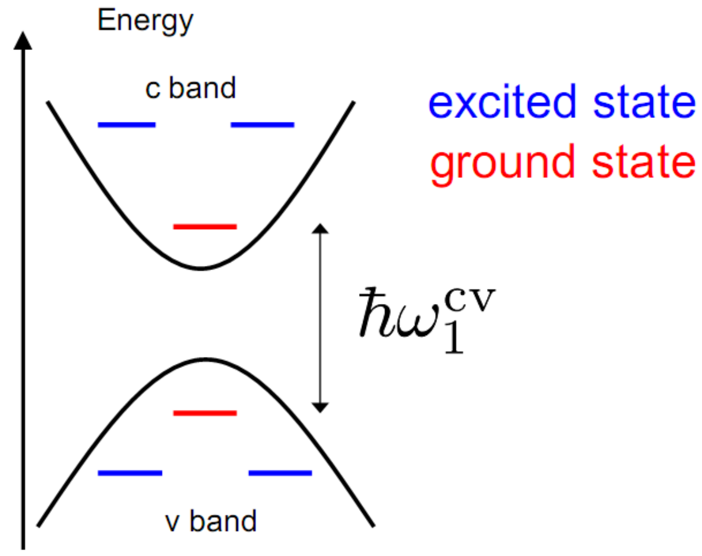
See e.g. Mahan

Notation and self-energies

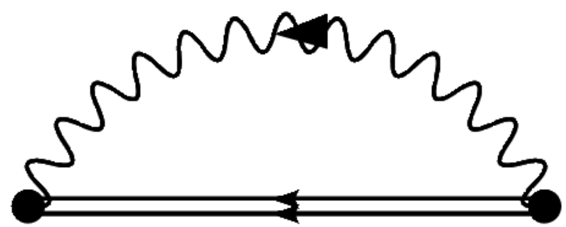
- Notation
  - b = conduction (c), valence (v)
  - α = ground, excited
  - We focus on ground states

$$\nu = (b, \alpha)$$

QD level structure



- Self-energies
  - Self-consistent Born approximation (SCBA)
  - Reasons and justifications:
    - Simplest possible
    - Often used in literature
    - Conserving approximation
    - For metals Migdal's theorem applies, maybe not for semicondonductors



- Hartree diagrams are not needed for linear response

Linear response theory

- Goal: The linear absorption spectra
  - Gives information on spectral features and can be calculated exactly for some systems

$$P(t) = \int_{-\infty}^{t} dt' \chi(t-t') E(t') \qquad \chi(\omega) = \frac{P(\omega)}{E(\omega)}$$

- Calculate polarization from equal time G^<

$$P(t) = -\sum_{\alpha} d_\alpha [\rho_\alpha^{\mathrm{cv}}(t) + \rho_\alpha^{\mathrm{vc}}(t)]$$

$$G_\alpha^{bb',<}(t,t) = i\hbar^{-1} \langle c_{b',\alpha}^\dagger(t) c_{b,\alpha}(t) \rangle = i\hbar^{-1} \rho_\alpha^{bb'}(t)$$

Kadanoff-Baym Equation

$$H = H_0 + H_\mathrm{i} + U(t)$$

- Solve equal-time KBE in the GKBA

$$\begin{aligned} i\hbar\partial_t \boldsymbol{G}^<(t,t) &= \left[\boldsymbol{H}_0(t) + \boldsymbol{\Sigma}^\mathrm{s}(t)\right]\boldsymbol{G}^<(t,t) - \boldsymbol{G}^<(t,t)\left[\boldsymbol{H}_0(t) + \boldsymbol{\Sigma}^\mathrm{s}(t)\right] \\ &+ \int_{-\infty}^{t} dt_1 \left[\boldsymbol{\Sigma}^>(t,t_1)\boldsymbol{G}^<(t_1,t) - \boldsymbol{\Sigma}^<(t,t_1)\boldsymbol{G}^>(t_1,t) - \boldsymbol{G}^>(t,t_1)\boldsymbol{\Sigma}^<(t_1,t) + \boldsymbol{G}^<(t,t_1)\boldsymbol{\Sigma}^>(t_1,t)\right] \end{aligned}$$

- Generalized Kadanoff-Baym Approximation (GKBA)

$$\boldsymbol{G}^\gtrless(t,t') = i\hbar\left[\boldsymbol{G}^r(t,t')\boldsymbol{G}^\gtrless(t',t') - \boldsymbol{G}^\gtrless(t,t)\boldsymbol{G}^a(t,t')\right] = \begin{cases} i\hbar\boldsymbol{G}^r(t,t')\boldsymbol{G}^\gtrless(t',t'), & t > t' \\ -i\hbar\boldsymbol{G}^\gtrless(t,t)\boldsymbol{G}^a(t,t'), & t' > t \end{cases}$$

- Retarded GFs for GKBA
  - We use diagonal equilibrium retarded GFs

KBE in frequency space

- EOM for optical polarization is linear in linear response
  - Populations maintain equil. values, 0 or 1
- Fourier transforming simplifies solution process
  - Iterative procedure maybe necessary

$$\omega_\alpha^{\mathrm{cv}} = \omega_\alpha^{\mathrm{c}} - \omega_\alpha^{\mathrm{v}}$$

$$(\hbar\omega - \hbar\omega_\alpha^{\mathrm{cv}})\rho_\alpha^{\mathrm{cv}}(\hbar\omega) = -\Gamma_\alpha^{\mathrm{DD}}(\hbar\omega)\rho_\alpha^{\mathrm{cv}}(\hbar\omega) + \sum_{\alpha_1}\Gamma_{\alpha\alpha_1}^{\mathrm{OD}}(\hbar\omega)\rho_{\alpha_1}^{\mathrm{cv}}(\hbar\omega) + U_\alpha^{\mathrm{cv}}(\hbar\omega)$$

$$\Gamma_\alpha^{\mathrm{DD}}(\hbar\omega) = i\hbar\sum_{\alpha_1}\left[K_{\alpha\alpha_1,\alpha_1\alpha}^{\mathrm{cc}}(\hbar\omega) + K_{\alpha\alpha_1,\alpha\alpha_1}^{\mathrm{vv}}(\hbar\omega)\right]$$

$$\Gamma_{\alpha\alpha_1}^{\mathrm{OD}}(\hbar\omega) = i\hbar\left[K_{\alpha\alpha_1,\alpha_1\alpha}^{\mathrm{cv}}(\hbar\omega) + K_{\alpha\alpha_1,\alpha\alpha_1}^{\mathrm{cv}}(\hbar\omega)\right]$$

$$K_{\alpha_3\alpha_4,\alpha_1\alpha_2}^{b_3b_4}(t) = G_{\alpha_1}^{\mathrm{c},r}(t)\left[G_{\alpha_2}^{\mathrm{v},r}(t)\right]^* D_{\alpha_3\alpha_4}^{b_3b_4,>}(t)$$

Lorke et al., PRB (2006)
Stauber et al., PRB (2006)

Retarded GF for GKBA

- Use equilibrium version, should be justified in linear response
  - Spectral properties are not changed much by the weak pulse
  - $G^r \sim (\text{external field})^2$
- In equilibrium two-time => one-time (difference time, $\tau = t - t'$)
  - EOM becomes

$$i\hbar\partial_\tau G^r_{\nu\nu'}(\tau) = \delta(\tau)\delta_{\nu\nu'} + \hbar\omega_{\nu'} G^r_{\nu\nu'}(\tau) + \int_0^\tau d\tau_1 \sum_{\nu_1} G^r_{\nu\nu_1}(\tau_1)\Sigma^r_{\nu_1\nu'}(\tau - \tau_1)$$

- Simplification: Only diagonal GFs are conidered

$$G^{bb',x}_{\alpha\alpha'}(\tau) = G^{b,x}_{\alpha}(\tau)\delta_{bb'}\delta_{\alpha\alpha'}, \quad \Sigma^{bb',r}_{\alpha\alpha'}(\tau) = \Sigma^{b,r}_{\alpha}(\tau)\delta_{bb'}\delta_{\alpha\alpha'}$$

Equilibrium retarded GF

- After some semiconductor specific considerations we get

$$\lambda_c = > \text{ and } \lambda_v = <$$

$$i\hbar\partial_\tau G^{b,r}_{\alpha}(\tau) = \delta(\tau) + \hbar\omega^b_\alpha G^{b,r}_\alpha(\tau) + \int_0^\tau d\tau_1 G^{b,r}_\alpha(\tau_1)\sum_{\alpha_1} G^{b,r}_{\alpha_1}(\tau-\tau_1) D^{bb,\lambda_b}_{\alpha\alpha_1}(\tau-\tau_1)$$

- Effective phonon GF
  - Sum of both phonon branches

Wave vector

Branch index

$$D^{\gtrless}_{\nu_2\nu\nu'\nu_1}(t,t') = i\hbar\sum_\mu M^\mu_{\nu\nu_1} M^{\bar{\mu}}_{\nu_2\nu'} D^{0,\gtrless}_{\mu\bar{\mu}}(t,t') \qquad \bar{\mu} = (-\boldsymbol{q},\lambda)$$

$$D^{0,>}_{\mu\mu'}(t,t') = -i\hbar^{-1}\left\{e^{i\omega_\mu(t-t')}n_B(\hbar\omega_\mu) + e^{-i\omega_\mu(t-t')}[n_B(\hbar\omega_\mu)+1]\right\}\delta_{\bar{\mu}',\mu},$$

$$D^{0,<}_{\mu\mu'}(t,t') = -i\hbar^{-1}\left\{e^{-i\omega_\mu(t-t')}n_B(\hbar\omega_\mu) + e^{i\omega_\mu(t-t')}[n_B(\hbar\omega_\mu)+1]\right\}\delta_{\bar{\mu}',\mu}.$$

Numerics for retarded GF

- "Time-stepping friendly" transformation
  - Removes delta function and free evolution

$$G_\alpha^{b,r}(\tau) = -i\hbar^{-1}\theta(\tau)e^{-i\omega_\alpha^b\tau}\mathcal{G}_\alpha^b(\tau)$$

- Predictor-corrector method employed for stepping
  - Only two source evals per time step despite being 4th order
  - First few steps can not be made using PC (use Euler)
  - Requires a bit of bookeeping

$$\partial_t u(t) = f(t)$$

$$\text{predictor}: \quad u_{n+1} = u_n + \frac{h}{12}(23f_n - 16f_{n-1} + 5f_{n-2}) + \mathcal{O}(h^4),$$

$$\text{corrector}: \quad u_{n+1} = u_n + \frac{h}{12}(5f_{n+1} + 8f_n - f_{n-1}) + \mathcal{O}(h^4).$$

- Memory integrals
  - Performed using the trapezoid rule (Simpson rule made not diff.)

$$\int_{hn}^{h(n+1)} dt u(t) = \frac{h}{2}(u_n + u_{n+1}) + \mathcal{O}(h^3)$$

Exact solution of IBM

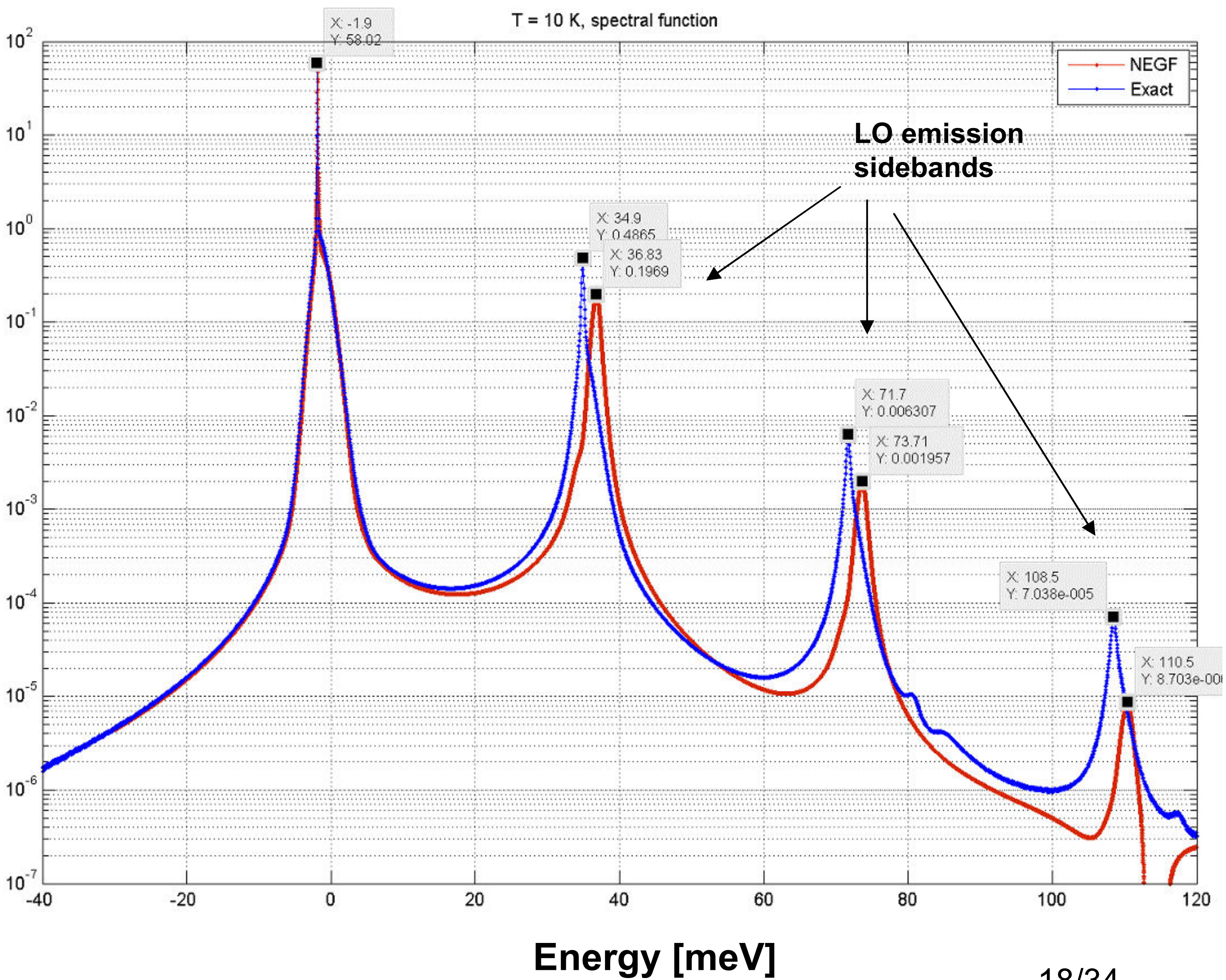
- It is well-known that the Independent Boson Model is exactly solvable
- Provides an important benchmark for equil. retarded GF for one-level systems and absorption spectra for two-level systems

$$G^r(t) = -i\hbar^{-1}\theta(t)e^{-i\omega_\alpha t - \hbar^{-2}\int_0^t dt' \int_0^{t'} dt'' D^>(t'')}$$

See e.g. Mahan

LO+LA – Spectral function

- Comparison with exact solution
  - LO+LA phonons
  - One-level system
  - ZPL nicely captured
  - Magnitude and position of sidebands are off
  - Sidebands are not polaron shifted
- Conclusion:
  - SCBA captures overall equilibrium spectral features well



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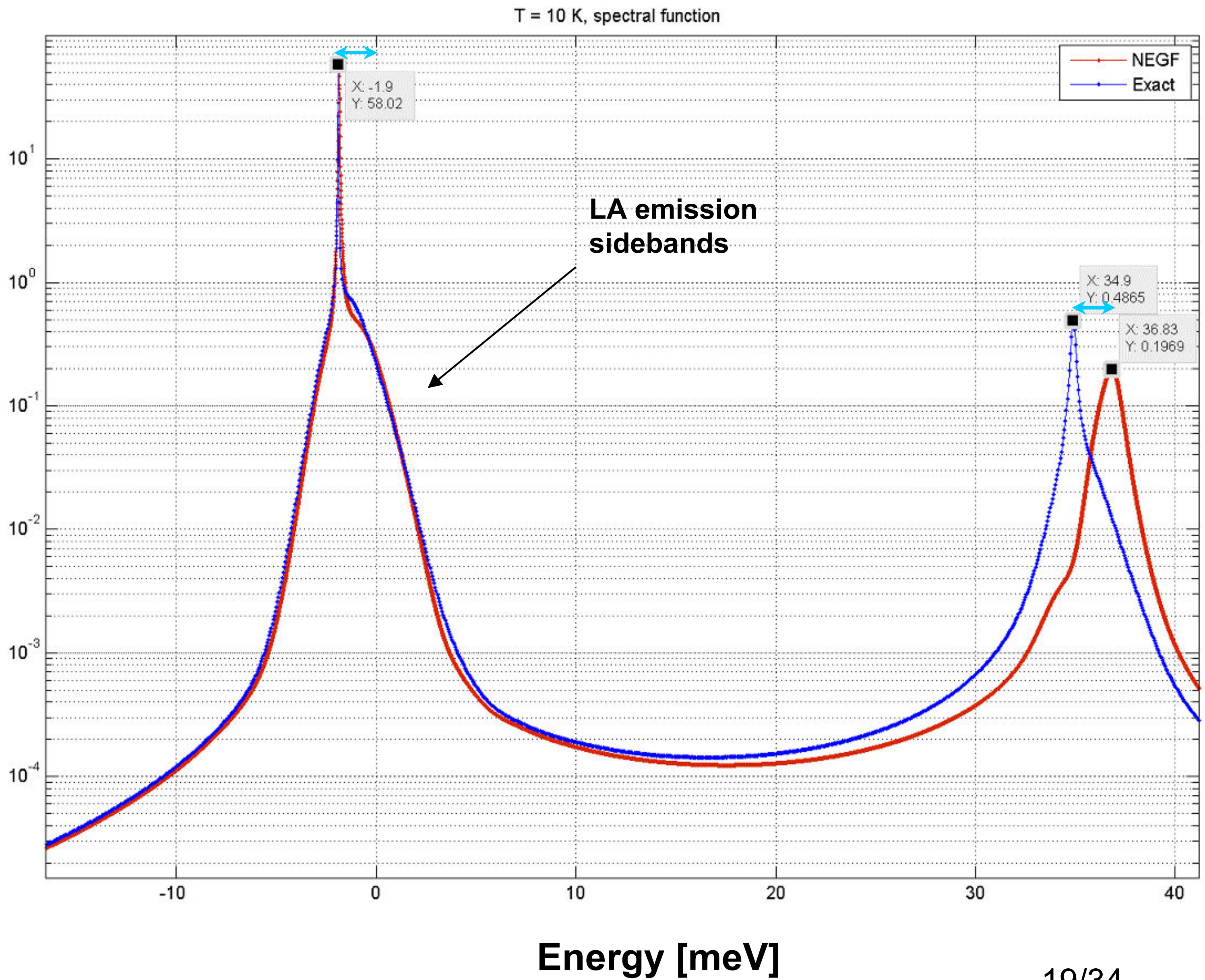
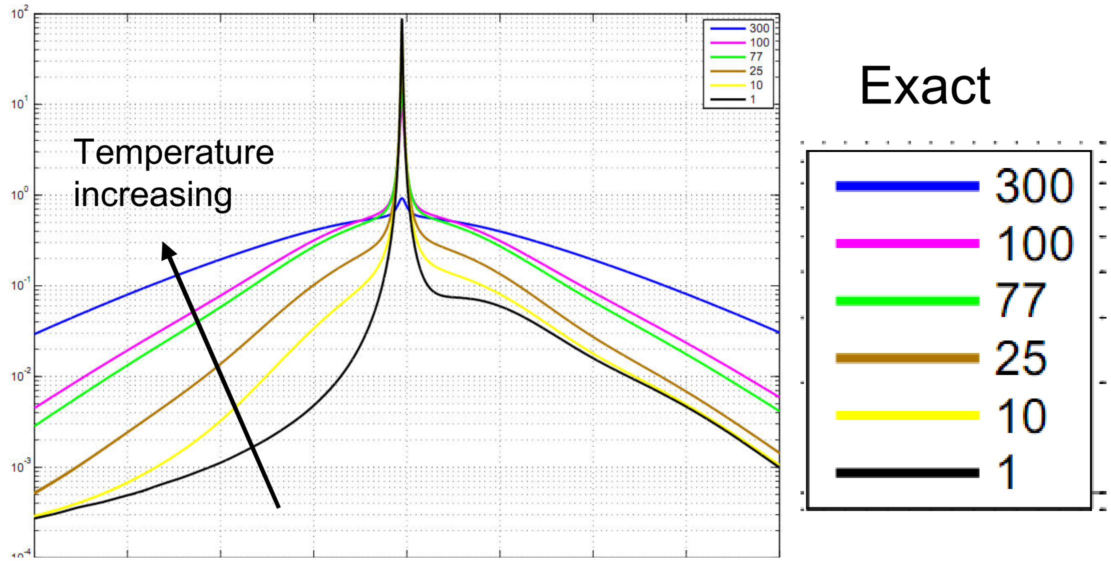
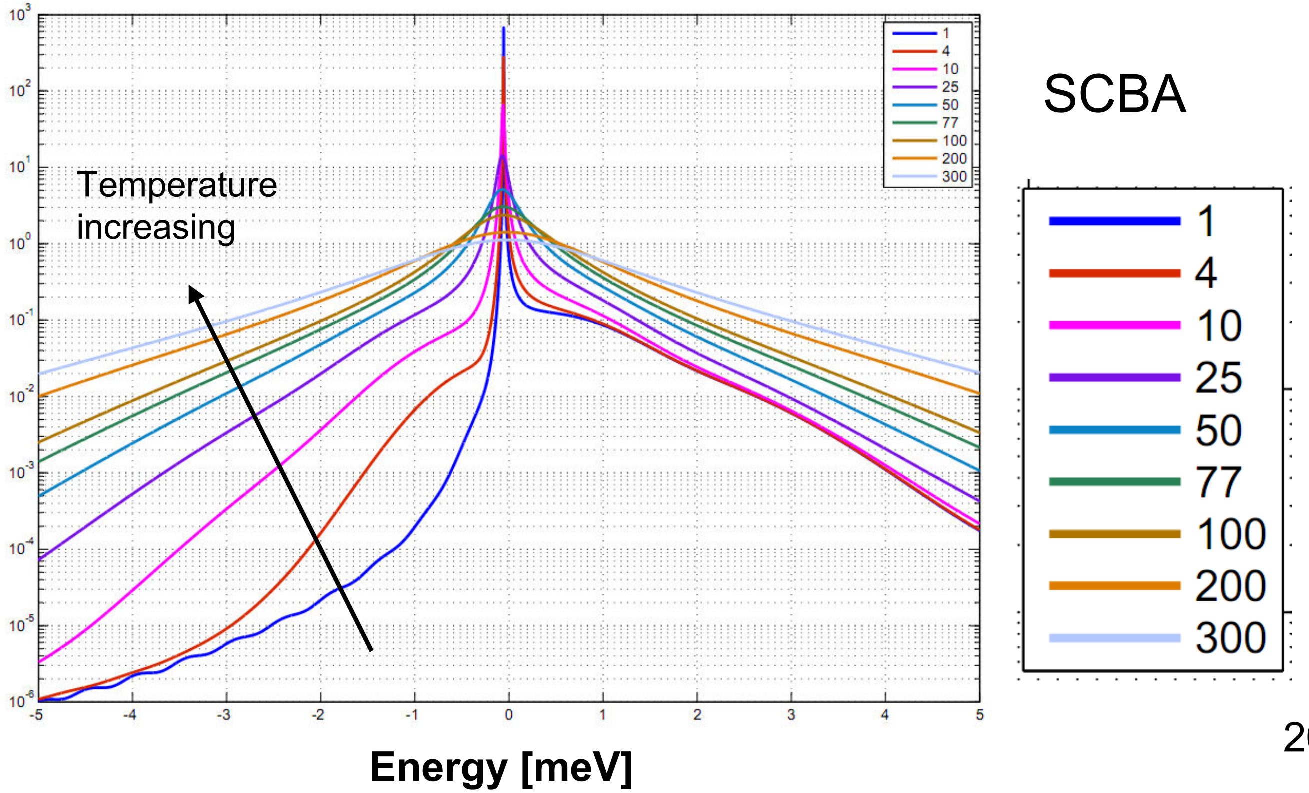


LA only – Absorption spec.

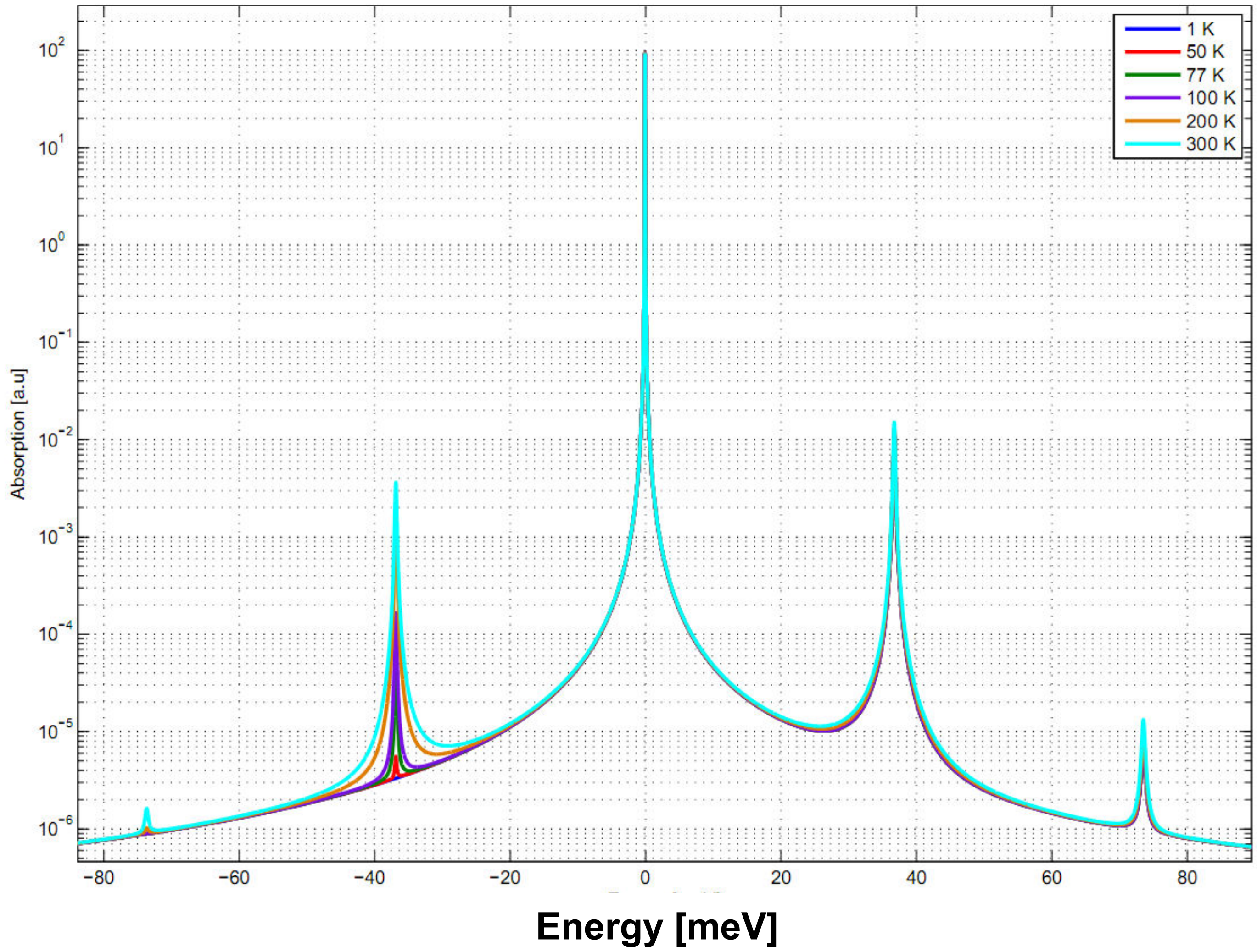
- Exact vs SCBA
  - Two-level system
  - Emission/absorption sidebands are clearly seen
- Conclusion
  - SCBA reproduces the exact solution fairly well





LO only – Absorption spec.

- Exact
  - Two-level system
  - Emission/absorptio n sidebands are clearly seen
  - Textbook result

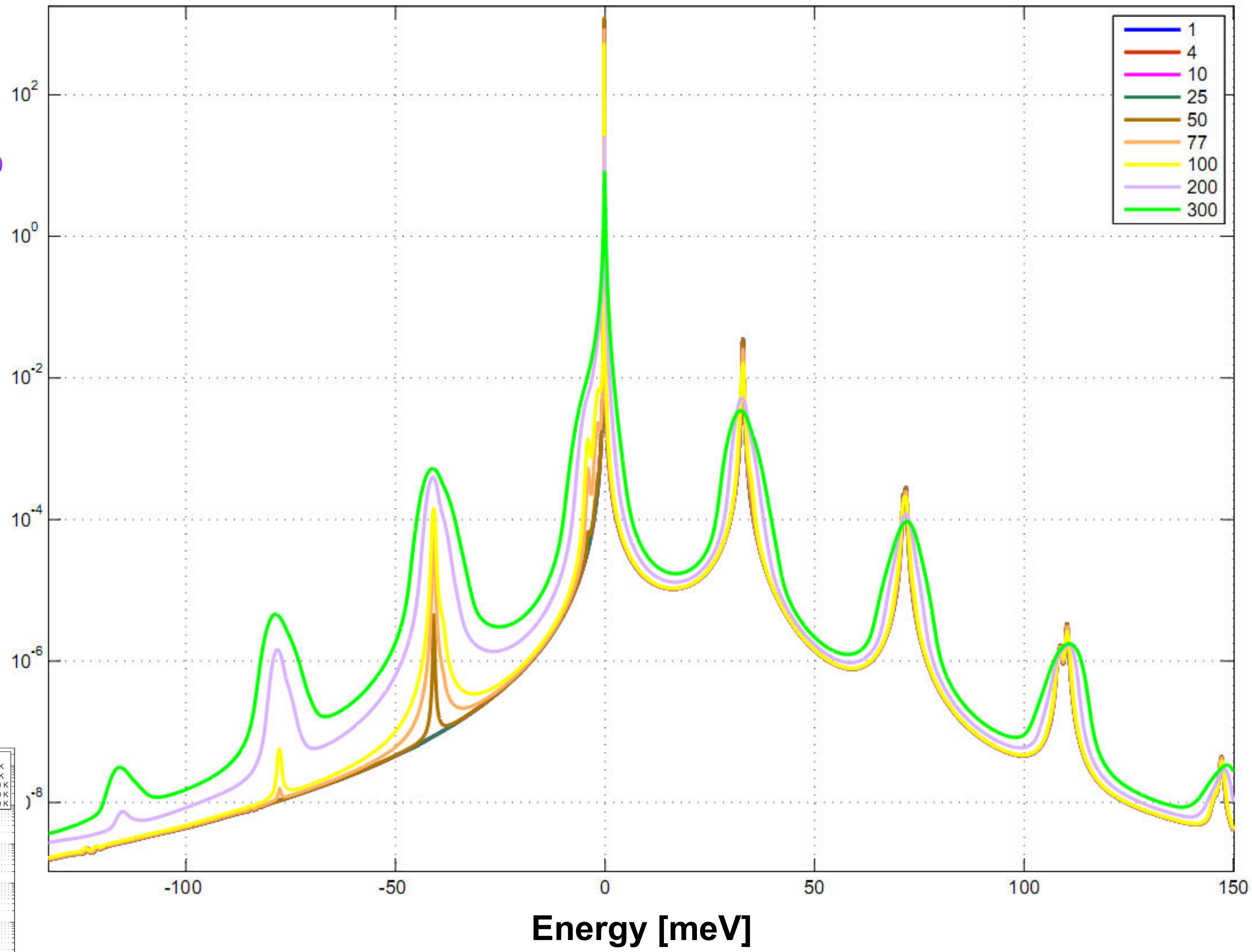


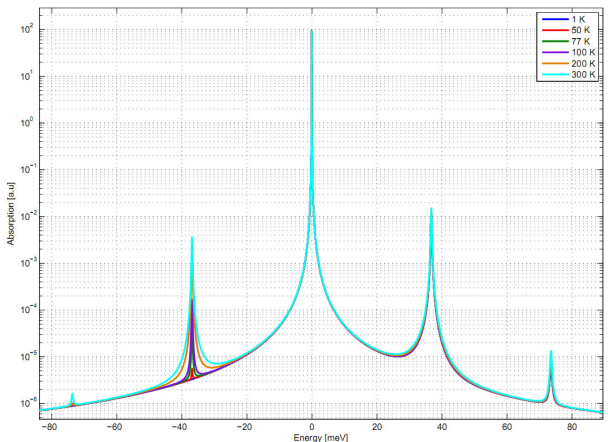
$$P(t) = \frac{1}{i\hbar} e^{-\lambda} e^{-i(\epsilon-\Delta)t} \times \sum_n \frac{1}{n!} \left[\left(\frac{M^2 (N_{\mathrm{LO}}+1)}{(\hbar\omega_{\mathrm{LO}})^2} \right)^n e^{-in\omega_{\mathrm{LO}}t} + \left(\frac{M^2 N_{\mathrm{LO}}}{(\hbar\omega_{\mathrm{LO}})^2} \right)^n e^{in\omega_{\mathrm{LO}}t} \right]$$

Eq. from phd thesis of J. Seebeck

LO only – Absorption spec.

- SCBA
  - Two-level system
  - Emission/absorption sidebands are clearly seen
  - At high T we see artifacts (artificial broadening)
  - Low T is more accurate
  - Why care about such small features?





Stauber et al., PRB (2000)

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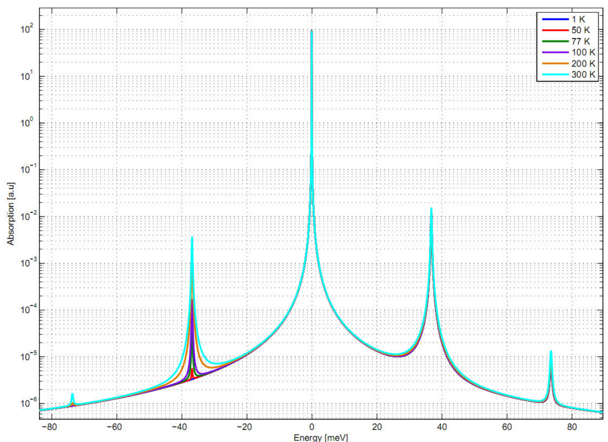

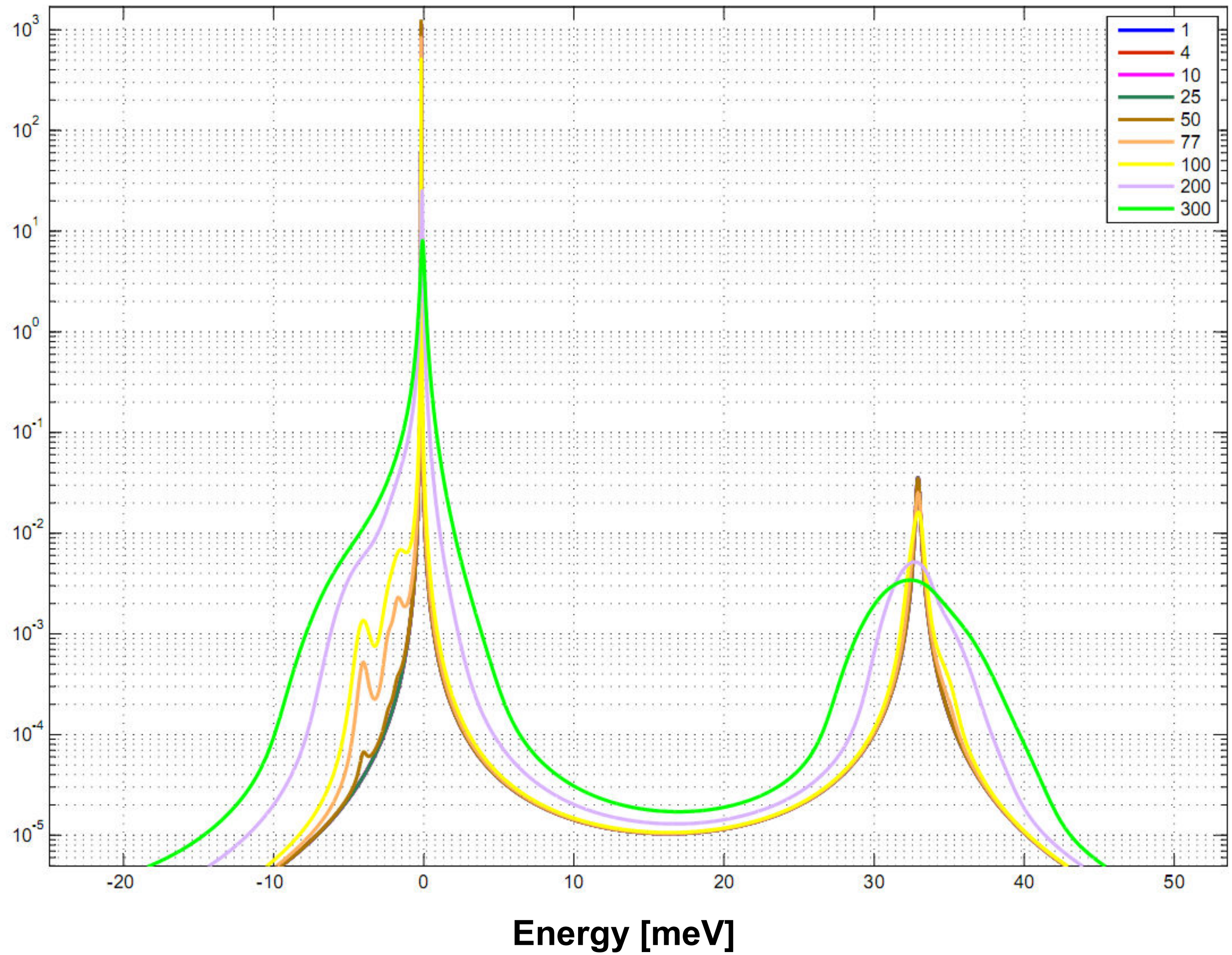

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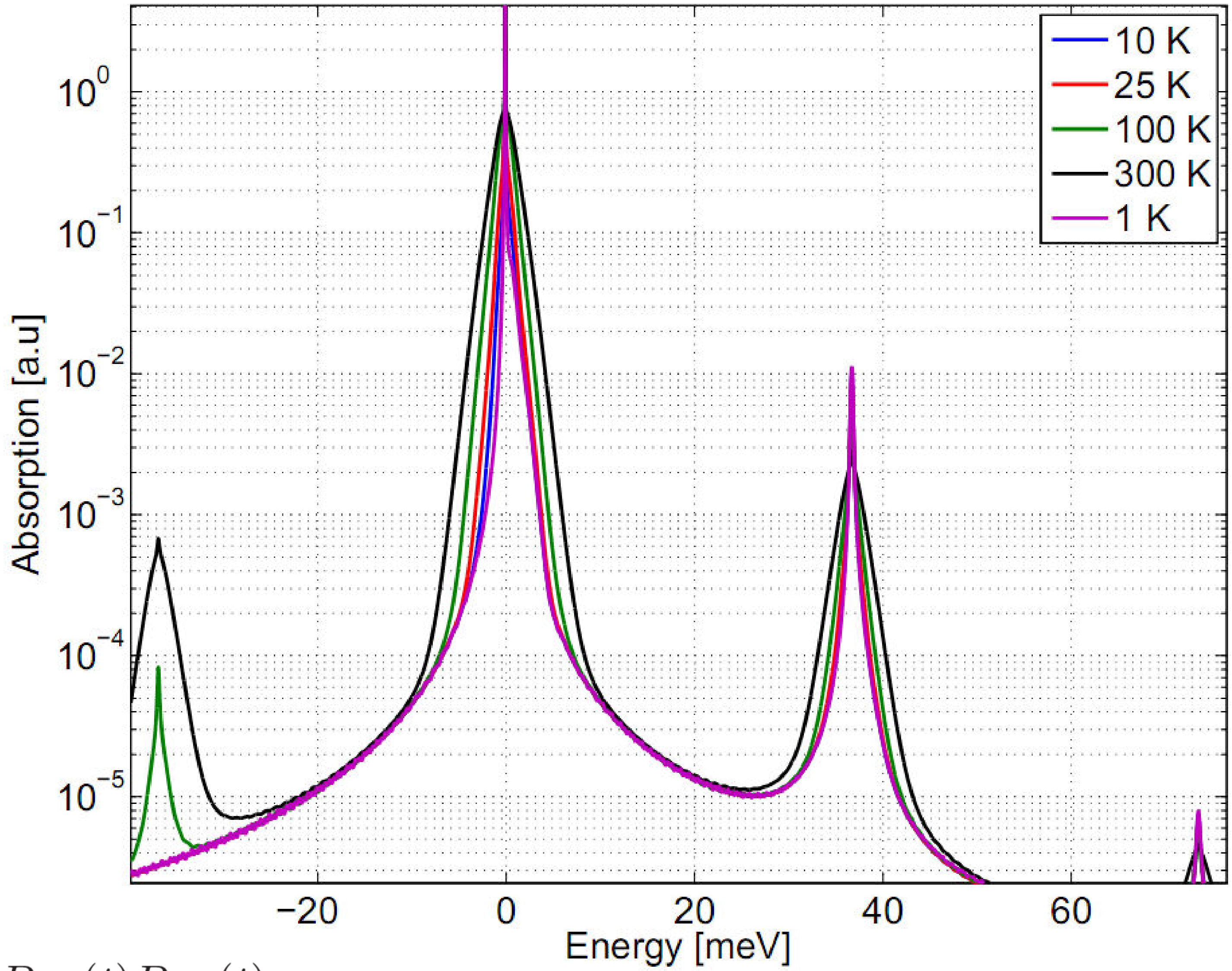
LO+LA – Absorption spec.

- Exact
  - Two-level system
  - Frequency convolution between LO and LA spectra



$$P(t) \propto e^{f_{\rm LO}(t)+f_{\rm LA}(t)} = P_{\rm LA}(t)P_{\rm LO}(t) \Longrightarrow$$

$$P(\omega) \propto \int d\omega_1 P_{\rm LO}(\omega_1)P_{\rm LA}(\omega-\omega_1)$$

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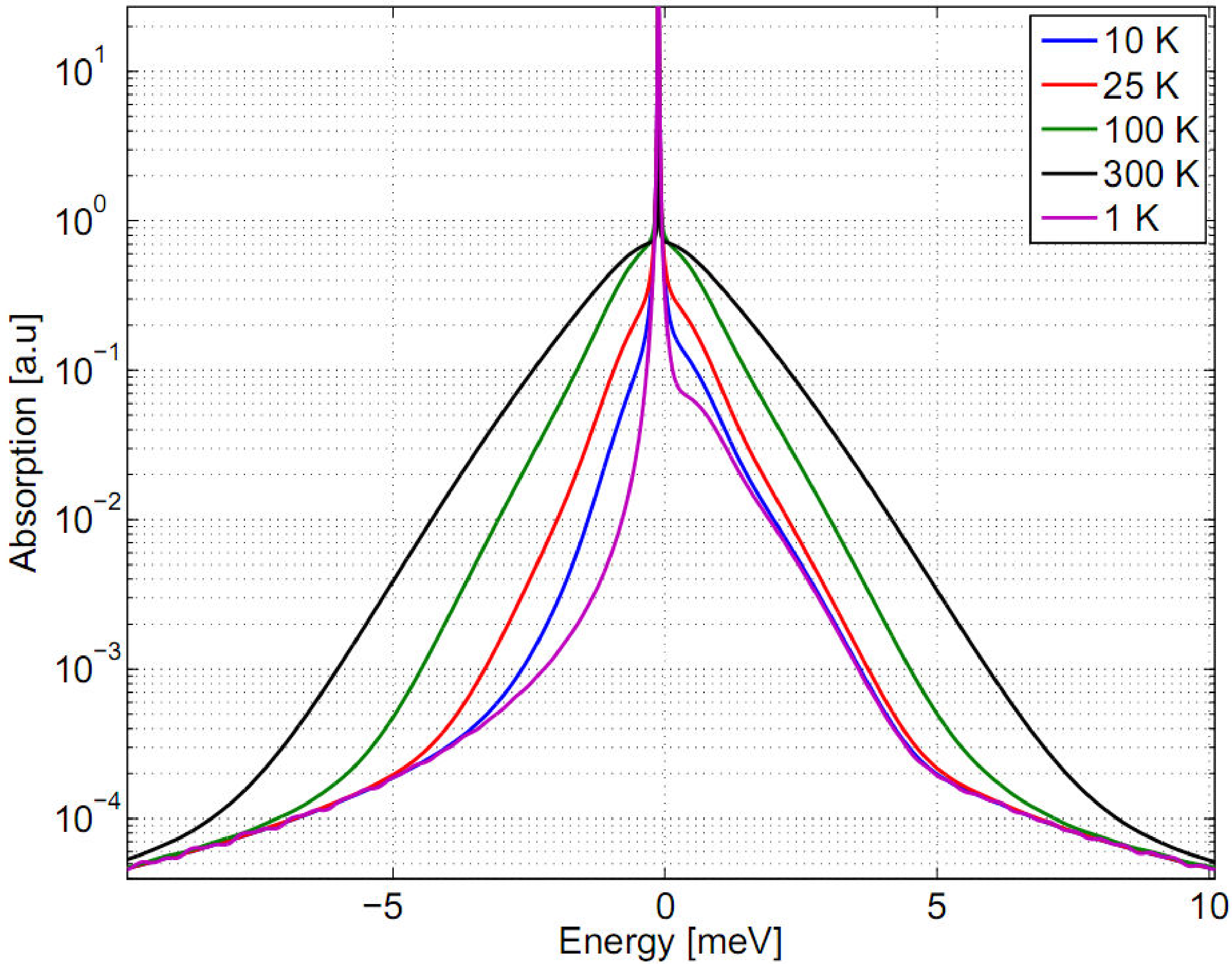

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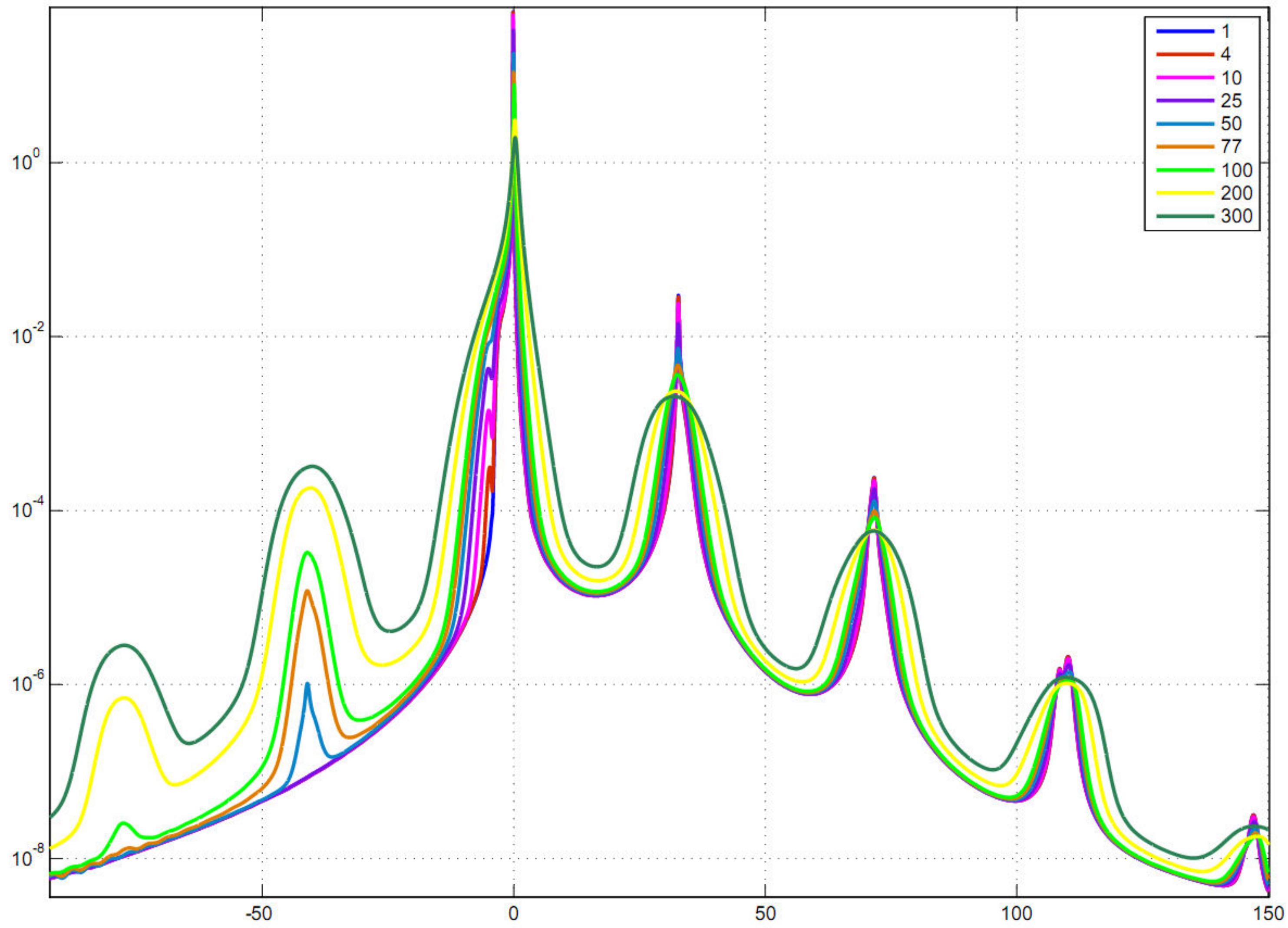
LO+LA – Absorption spec.

- SCBA
  - Two-level system
  - Rough overall structure looks ok
  - Focus on ZPL…

- Conclusion
  - LA sidebands are misplaced for ZPL
  - Looks better for LO sidebands



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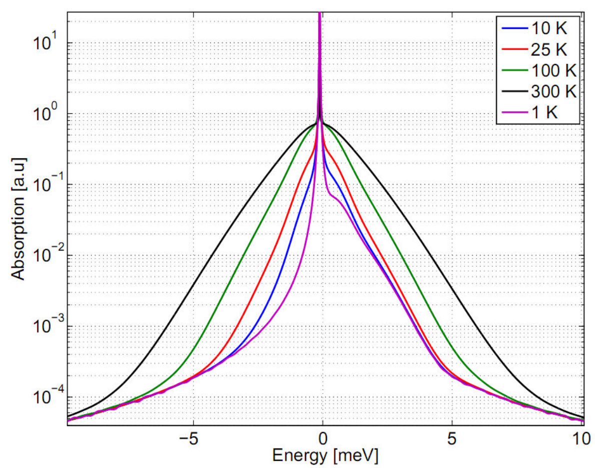
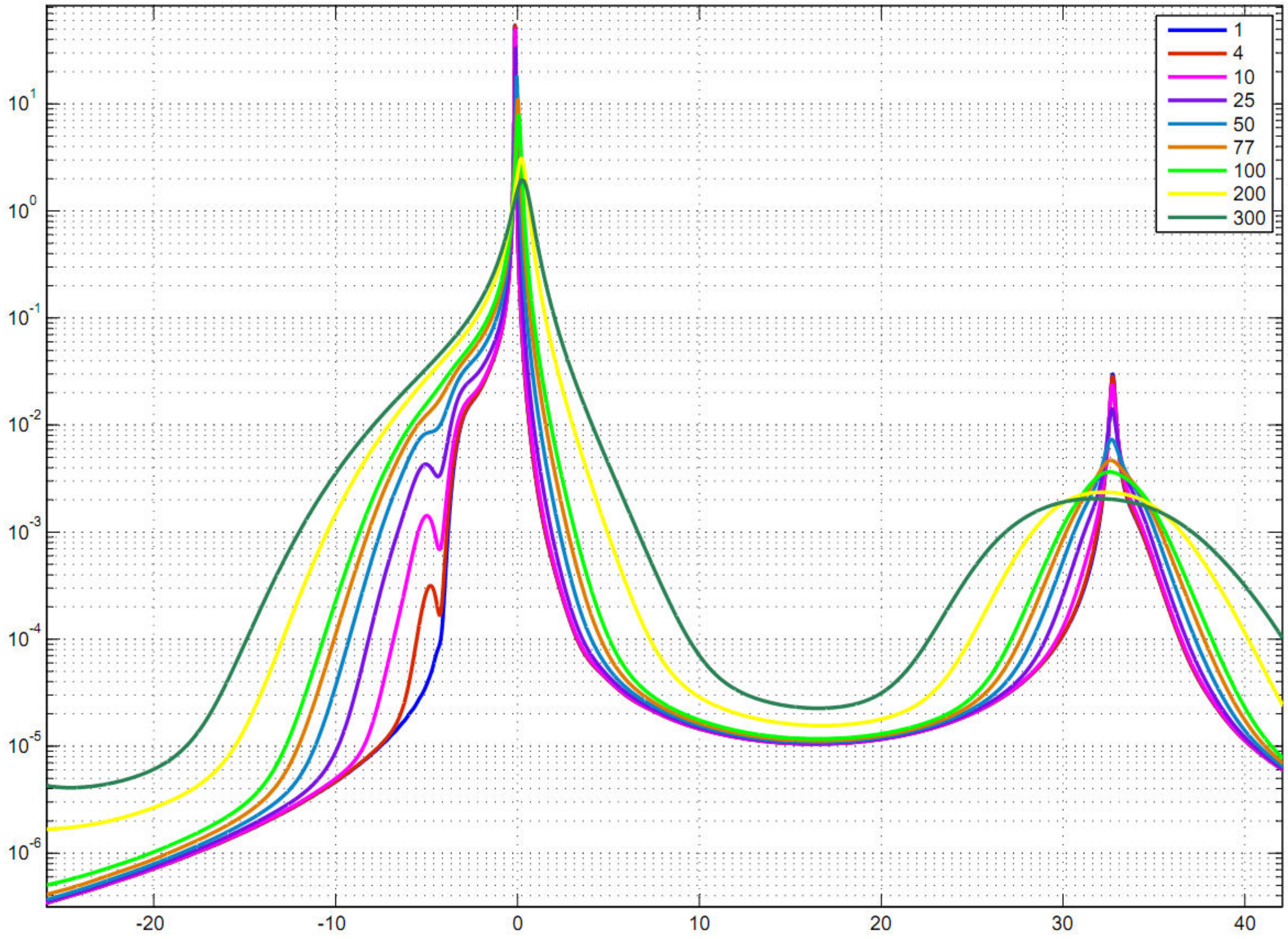
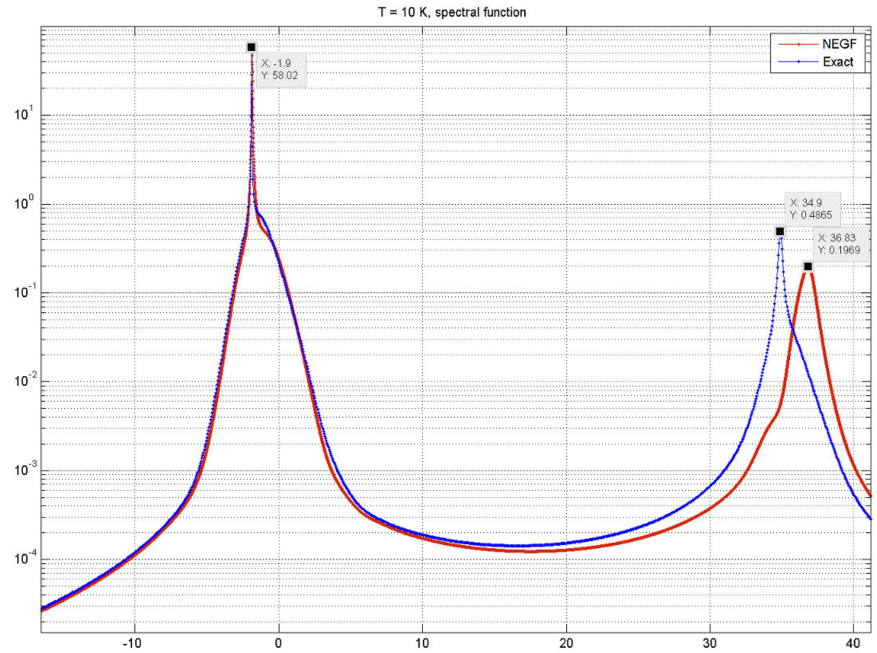
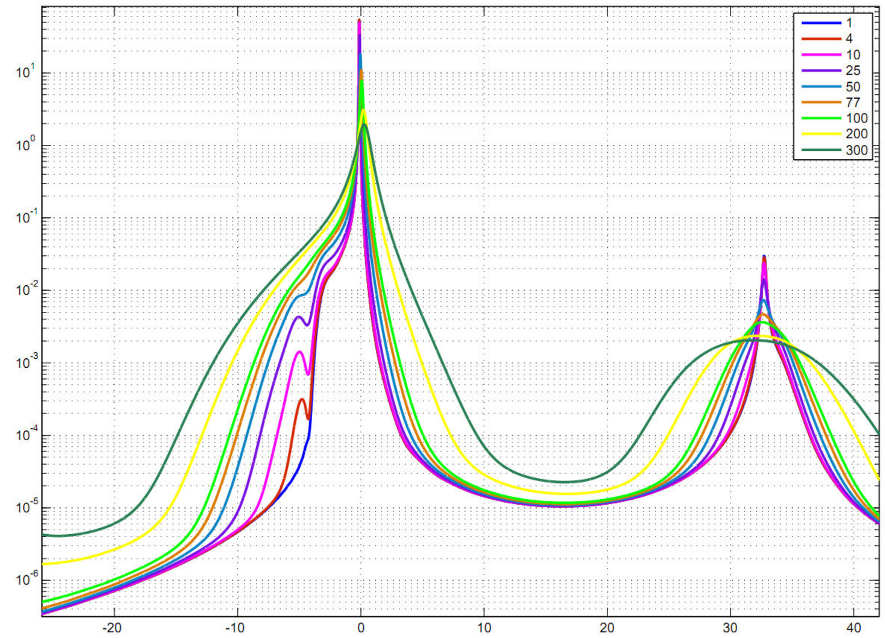




Sum up of observations

- SCBA in equilibrium
  - Performs reasonably well for both individual phonon branches and the combination
  - Sidebands are however not polaron shifted
- SCBA in linear response
  - Performs well for LA phonons at all T
  - Performs well for LO phonons at low T
  - For LO+LA it fails at all T, especially for LA sideband position

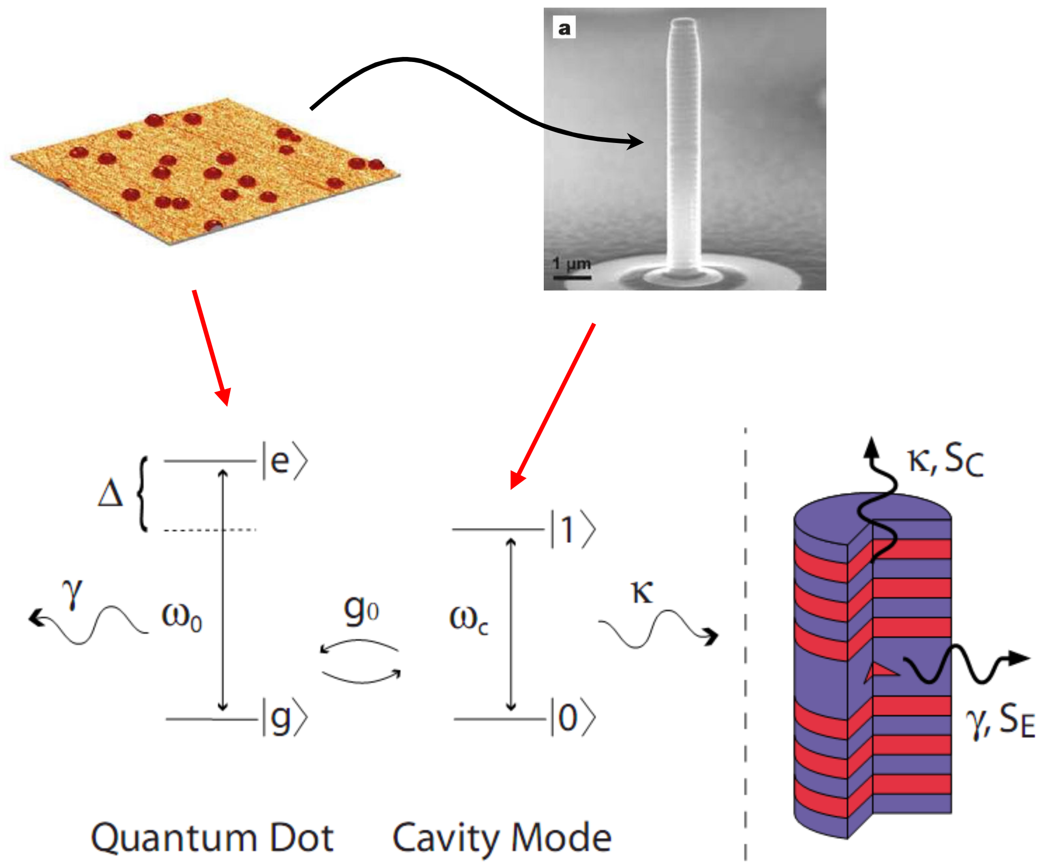




Why care (so much) about sidebands?

- Consider a basic cavity-QED system
  - QD and cavity interact
  - Allow for detuning

- Standard lore
  - Everything is symmetric wrt detuning
  - What about phonon sidebands?
  - They are very small!



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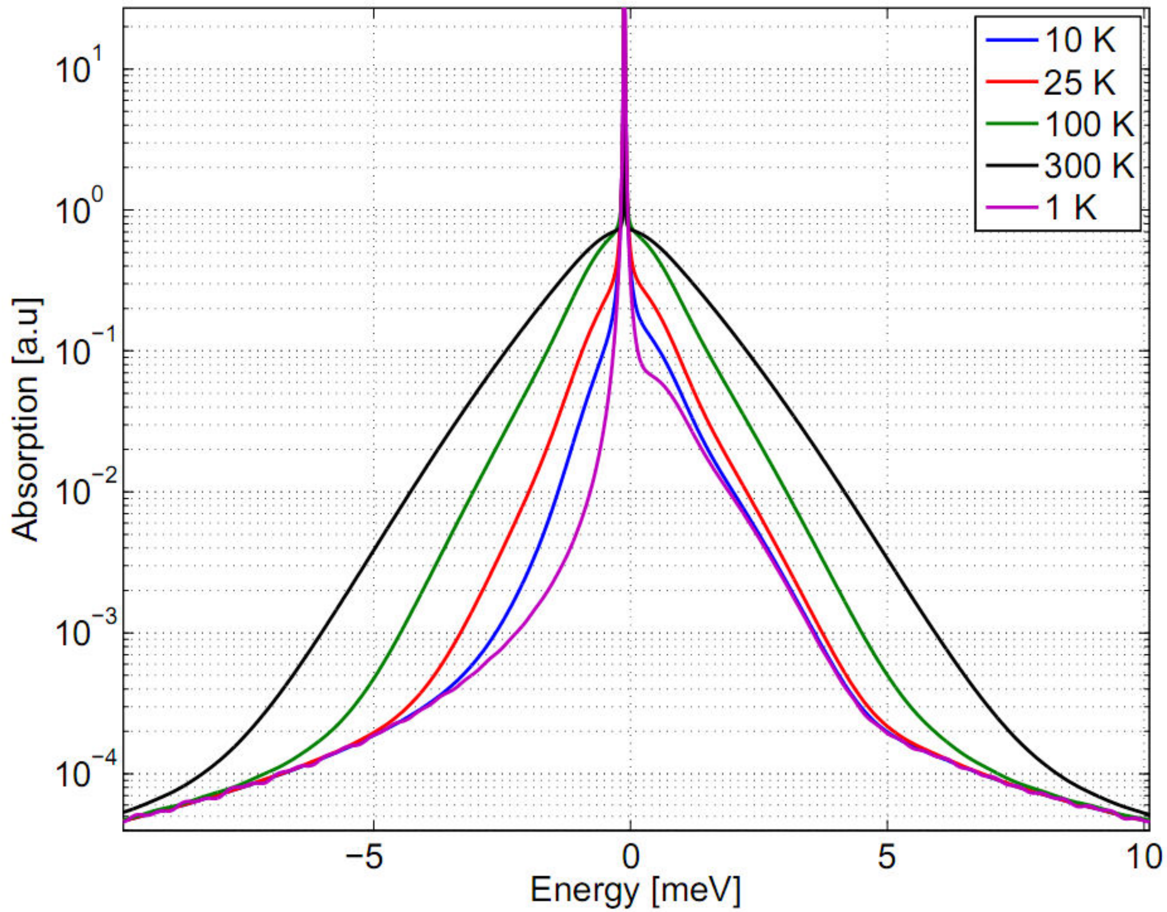
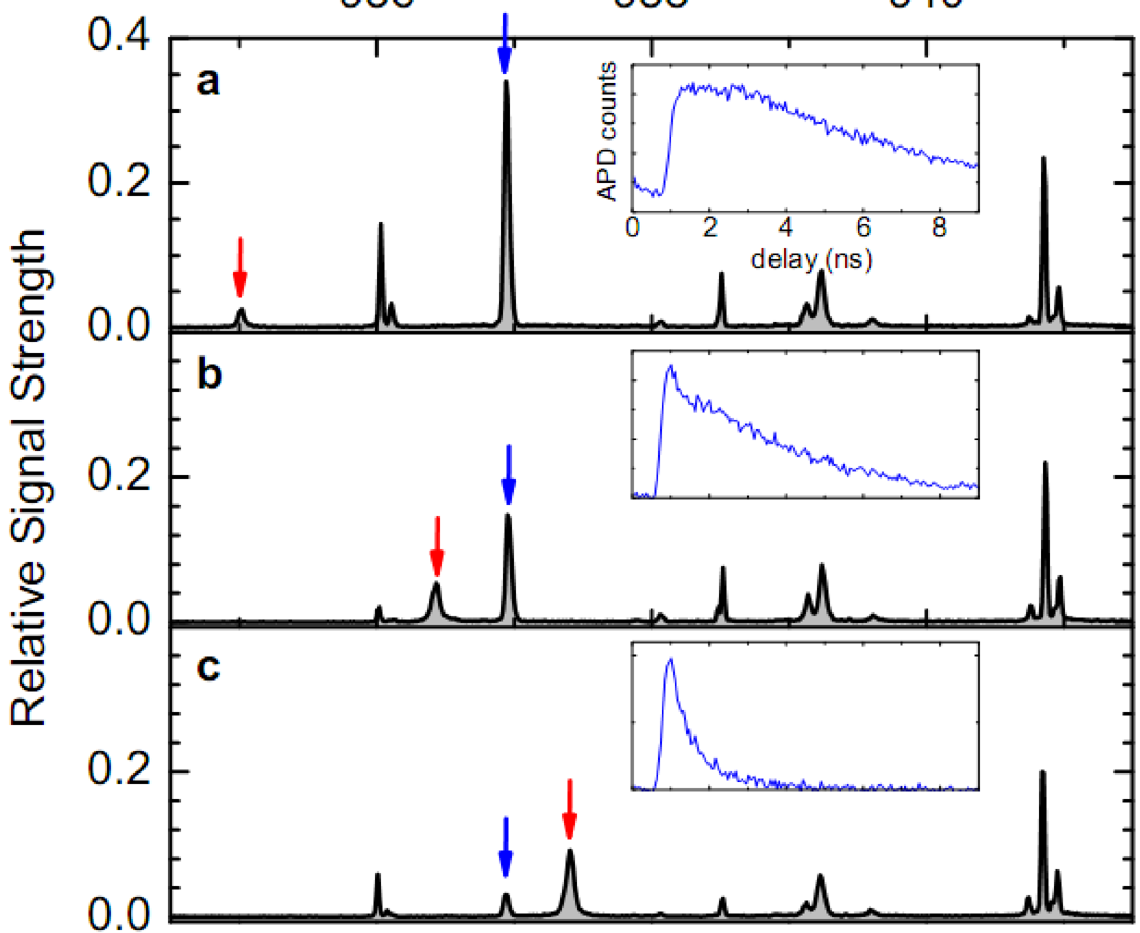
  - QD and cavity interact
  - Allow for detuning

- Standard lore
  - Everything is symmetric wrt detuning
  - What about phonon sidebands?
  - They are very small!



Asymmetries

- Recent experiments observe asymmetry wrt detuning
  - Strongly coupled QD and cavity
  - Lifetimes and emission spectra
  - Are phonons responsible?



Red arrow: Cavity

Blue arrow: QD

Asymmetries

- Recent experiments observe asymmetry wrt detuning
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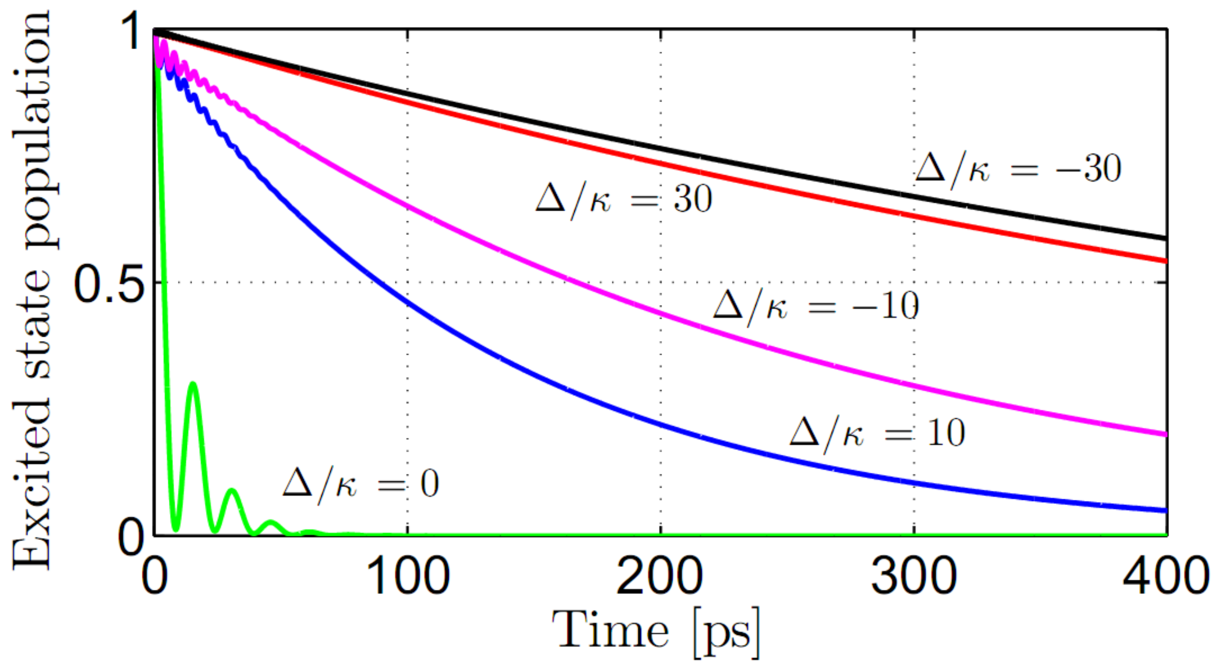
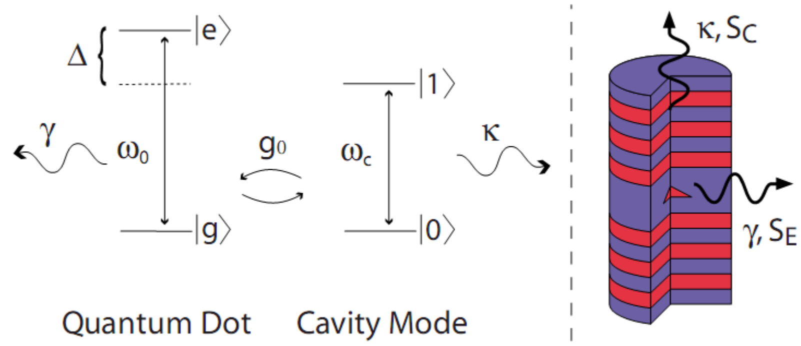
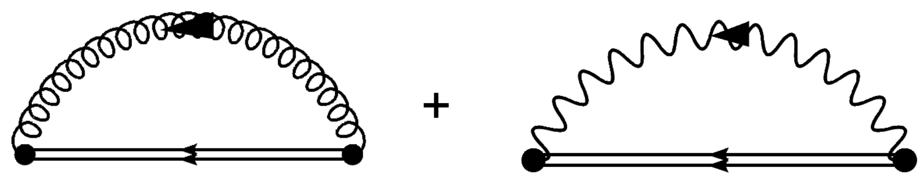


- Phonon model
  - QD, cavity, and LA phonons
  - Sidebands are Purcell enhanced due to cavity!
  - Misplaced sidebands would matter a lot for this case
  - Two-time quantities can be found using the Quantum Regression Theorem



Summary

- SCBA fails for multiple interactions
  - The “mixed processes” introduced through self-consistency are not sufficient



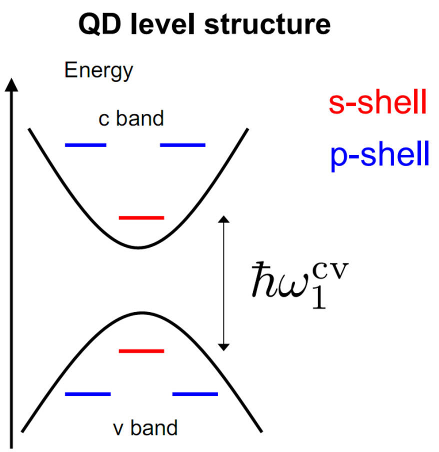
  - The present issue LO+LA issue seems to be related to the transition to non-equilibrium
    - LO+LA works in equilibrium
    - LO <u>or</u> LA works in non-equilibrium
    - LO+LA does not work in non-equilibrium

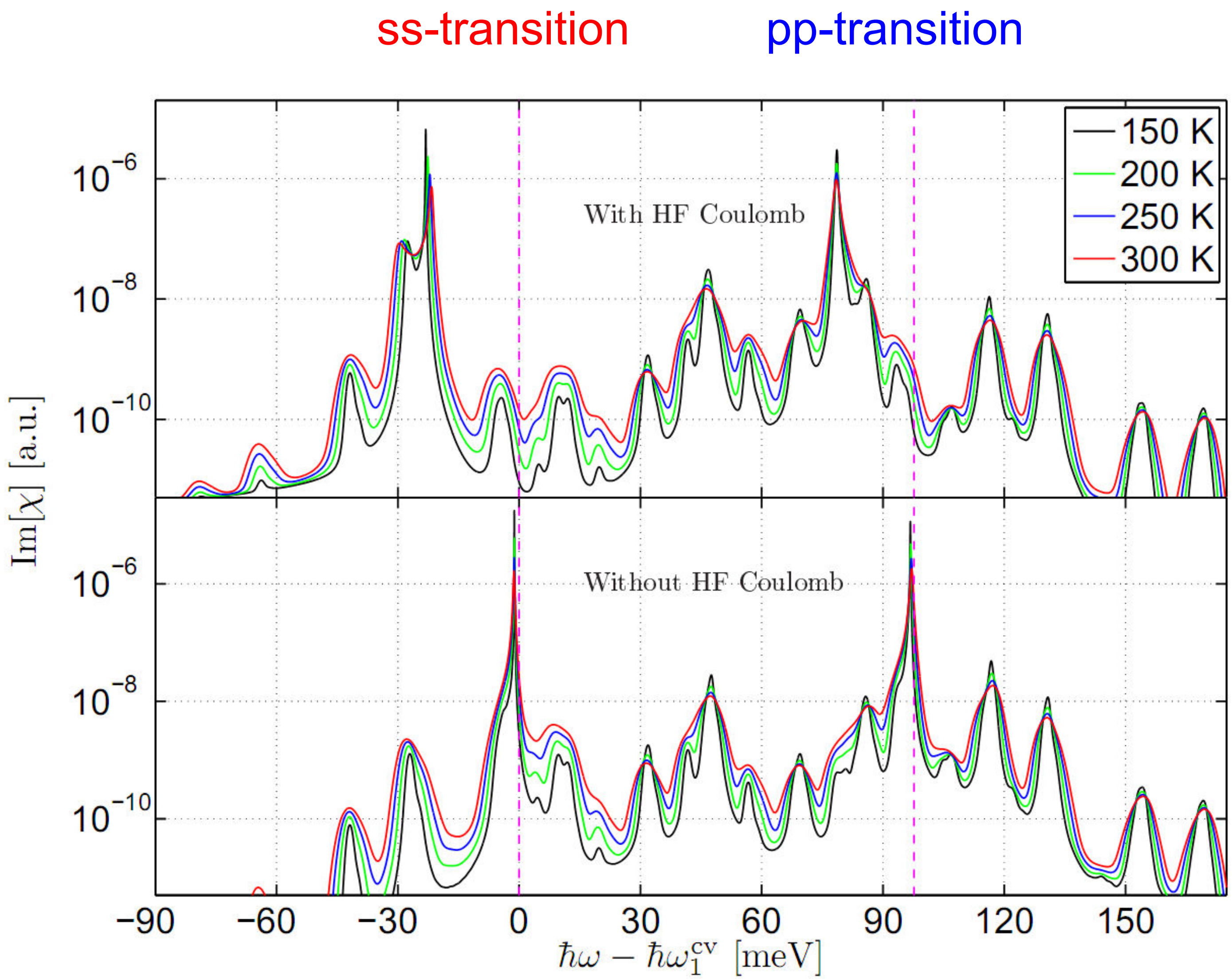
- The solutions “of course” lies in selecting an appropriate self-energy
  - But how?
  - To complicate further. How to couple to Coulomb or even photons?

Thank you for your attention!

Known issues with the SCBA

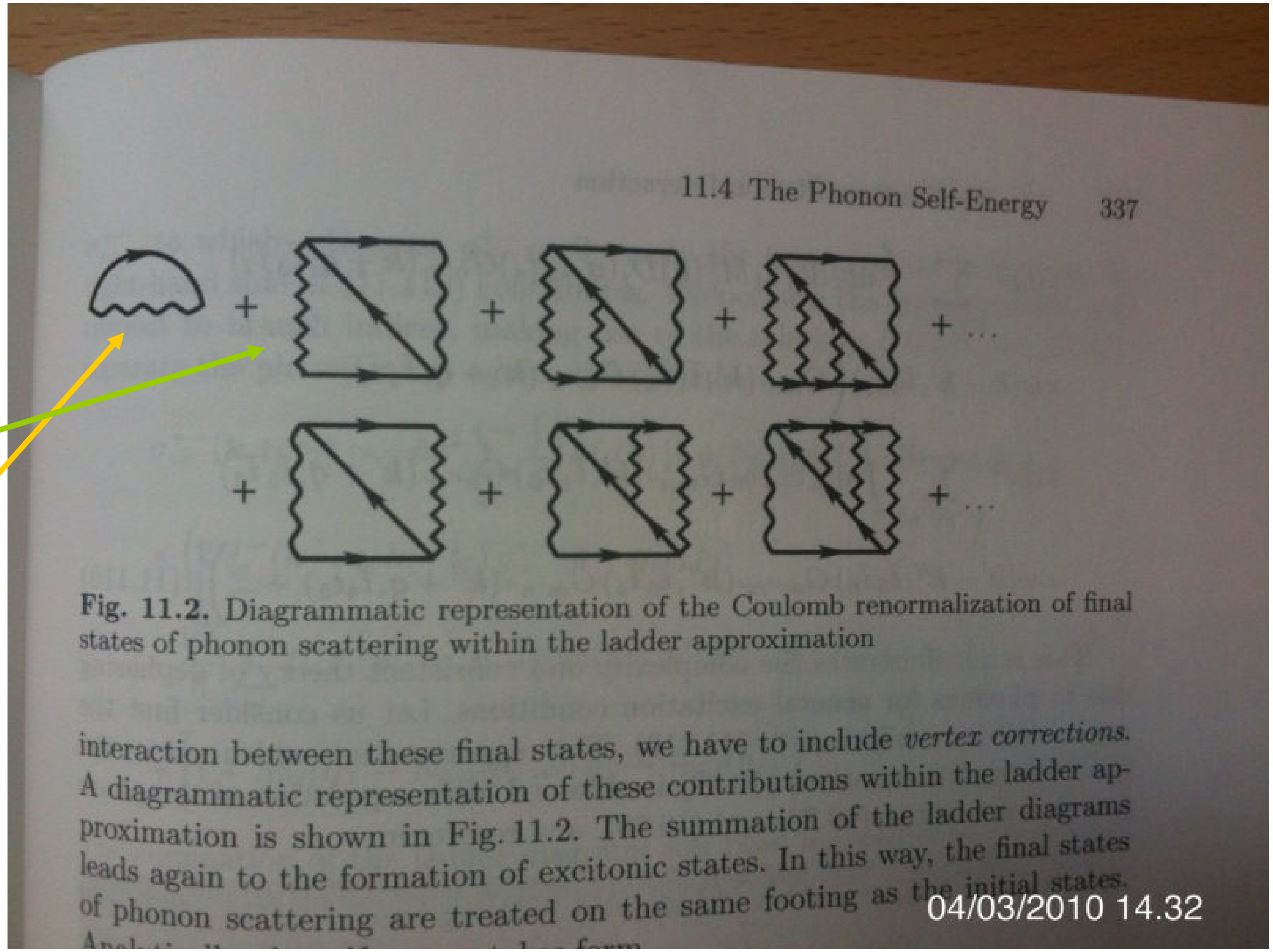
- Phonon sidebands are not exciton shifted, only ZPL is





Known issues with the SCBA

- Solution to non-shifting sidebands
  - W. Schäfers textbook
  - Leads to the solution of a two-particle polarization function



Coulomb line

phonon line

Results – retarded GF

- Spectral densities : LO+LA
  - Multi-level QD
  - Hybridization from off-diag. coupling
  - Sidebands from diag. coupling
  - No exact solution for comparison

$$A^b_\alpha(\omega) = -2\mathrm{Im}\left[G^{b,r}_\alpha(\omega)\right]$$