Anharmonic Contributions To Lattice Thermal Conductivity

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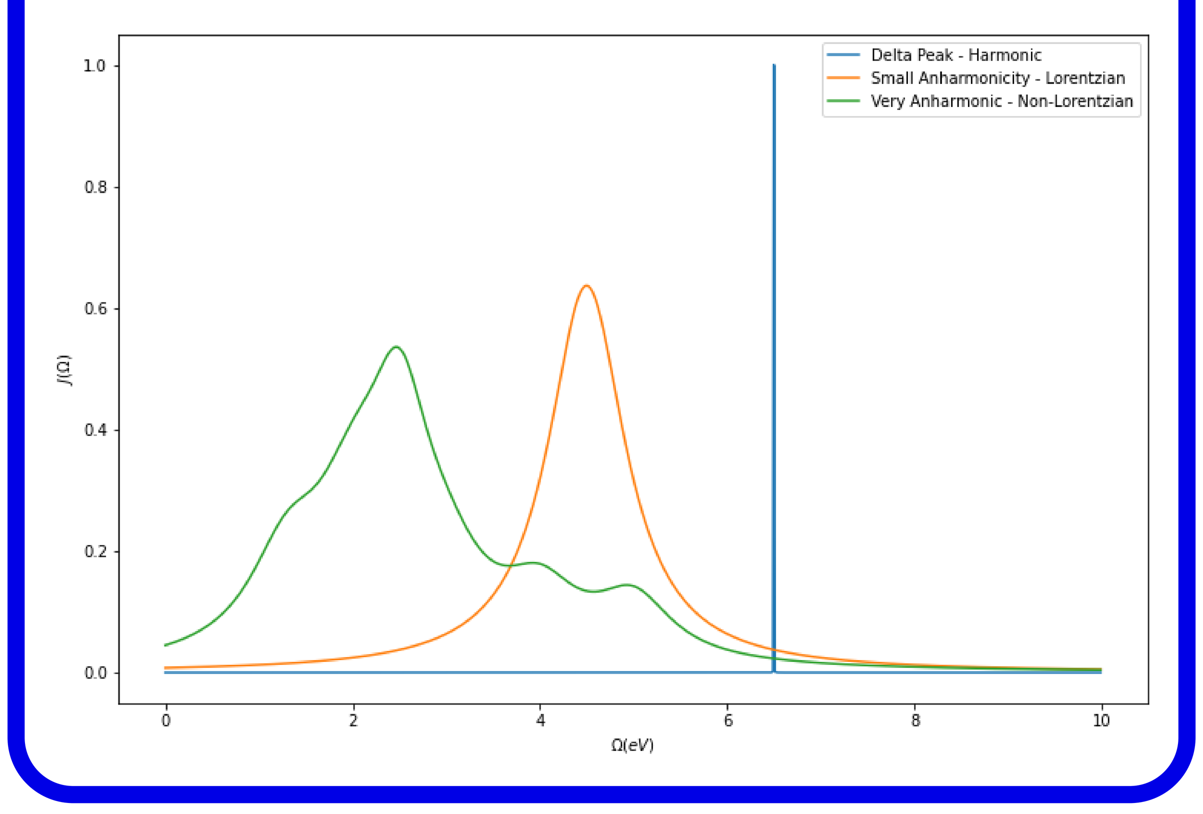
1. Introduction

Anharmonic thermal transport in insulators can be approached in several ways of varying complexity [1, 2], from independent phonon models to a more rigorous treatment with inter-branch coupling and memory included [3]. All current approaches rely, however, on the renormalization of the harmonic expression to include anharmonic effects. In this work we use an explicitly anharmonic heat current to derive the thermal transport coefficients, and find their higher order anharmonic contributions.

2. Quasi-harmonic Approaches

- Green-Kubo: $\kappa = \frac{V}{k_B T^2} \int_0^\infty \langle \mathbf{J}(t)\mathbf{J}(0)\rangle dt$.
- Harmonic heat current: $\mathbf{J}_{ha} = \sum_{ab} \mathbf{v}_{ab}\omega_a B_a A_b$, $\mathrm{q} = (\mathbf{q}, s_q)$, $B_q = a_q - a^\dagger_{\bar{q}}$, $A_q = a_q + a^\dagger_{\bar{q}}$.
- Thermal transport coefficients: $\kappa = \frac{1}{2V}\sum_{ab} \mathbf{v}_{ab} \otimes \mathbf{v}_{ab} c_{ab} \tau_{ab}$.
- Anharmonicity is introduced in τ as:
  - RTA [4]: $\tau_a = \frac{1}{2\Gamma_a}$ → diagonal in momentum and mode.
  - QHGK [1]: $\tau_{\mathbf{a}s_a s_b} = \frac{\Gamma_{\mathbf{a}s_a} + \Gamma_{\mathbf{a}s_b}}{(\omega_{\mathbf{a}s_a} - \omega_{\mathbf{a}s_b})^2 + (\Gamma_{\mathbf{a}s_a} + \Gamma_{\mathbf{a}s_b})^2}$ → no longer diagonal in mode.
  - Γ calculated via Fermi golden rule → no memory effects.
- Anharmonicity renormalizes harmonic results, but no purely anharmonic contributions.

3. Spectral functions



4. Anharmonic Heat Current

- AHC: $\mathbf{J}_{anh} = \sum_{abc} \mathbf{v}_{abc}\omega_a B_a A_b A_c$.
- $\mathbf{v}_{abc}$ generalizes $\mathbf{v}_{ab}$ to the 3-phonon case.
- $\mathbf{J} = \mathbf{J}_{ha} + \mathbf{J}_{anh}$.
- $\kappa \propto \langle (\mathbf{J}_{ha}(t) + \mathbf{J}_{anh}(t))(\mathbf{J}_{ha} + \mathbf{J}_{anh})\rangle$.
- Purely anharmonic contributions will appear.

5. Decoupling Scheme

- 4-point correlation function $\langle B_a(t)A_b(t)B_c A_d\rangle$ leads to the quasi-harmonic results.
- 5 and 6-point correlation functions lead to new contributions.
- Decoupling scheme:

$$\langle B_a(t)A_b(t)A_c(t)B_d A_e A_f\rangle \approx \langle B_a(t)B_d\rangle\langle A_b(t)A_e\rangle\langle A_c(t)A_f\rangle$$
$$+\text{other 2-pt}$$
$$+\langle B_a(t)A_b(t)B_d\rangle\langle A_c(t)A_e A_f\rangle$$
$$+\text{other 3-pt} .$$

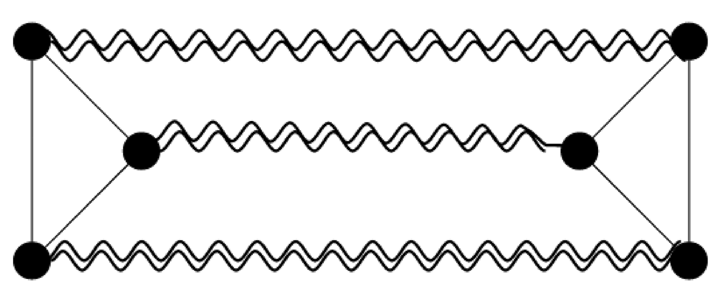
- Since we can write all 3-point functions in terms of 2-point ones, all contributions will still be written in terms of dressed-phonons.

6. Anharmonic Contributions

- In order to evaluate the Kubo integral, we transform the correlation functions into phonon spectral functions $J(\Omega)$:

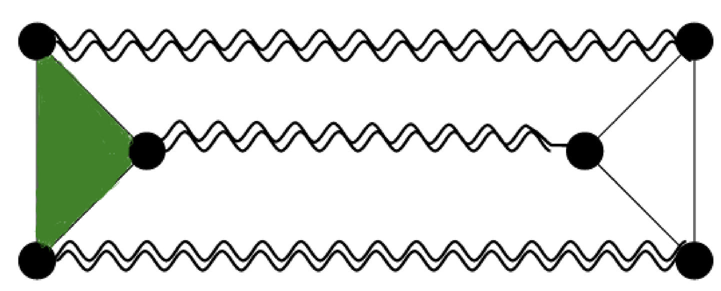
$$\langle X_a(t)Y_b^\dagger\rangle = \int J_{ab}^{XY}(\Omega)(n(\Omega)+1)e^{\Omega\tau} d\Omega .$$

- By convolution of these spectral functions we obtain the purely anharmonic contributions to κ:



$$\kappa_{anh-anh}^{2-pt} \propto J^{AA}_{-(\mathbf{a}+\mathbf{b})s_c s_f}(\Omega) * J^{BB}_{\mathbf{a}s_a s_d}(\Omega') * J^{AA}_{\mathbf{b}s_b s_e}(\Omega'')$$

- Simple 2-momentum dependency.
- Main contribution to anharmonic κ.



$$\kappa_{anh-anh}^{3-pt} \propto (S^{\mathrm{III}}_{ab}(\Omega) J^{AB}_{(\mathbf{a}+\mathbf{b})s_\mu s_d}(\Omega)) * (\mathcal{S}^{\mathrm{I}}_{-(\mathbf{a}+\mathbf{b})s_c}(\Omega') J^{AA}_{-\mathbf{e}s_\nu s_e}(\Omega')) * (J^{AA}_{-(\mathbf{a}+\mathbf{b}-\mathbf{e})s_\eta s_f}(\Omega''))$$

- 3-momentum dependency, $J(\Omega)$ filtered by $S(\Omega)$ and $\mathcal{S}(\Omega)$.

$$\kappa_{anh-ha} \propto (S^{\mathrm{III}}_{ab}(\Omega) J^{AB}_{(\mathbf{a}+\mathbf{b})s_\mu s_d}(\Omega)) * (J^{AA}_{-(\mathbf{a}+\mathbf{b})s_c s_e}(\Omega'))$$

- 2-momentum dependency, corrects κ_{ha}.

$$\kappa_{ha-anh} \propto (\mathcal{S}^{\mathrm{IV}}_{a}(\Omega) J^{AB}_{-\mathbf{c}s_\mu s_c}(\Omega)) * (J^{AA}_{(\mathbf{a}+\mathbf{c})s_\nu s_d}(\Omega')) * (J^{AA}_{-\mathbf{a}s_b s_e}(\Omega''))$$

- 2-momentum dependency, but with $J(\Omega)$ filtered by $\mathcal{S}(\Omega)$.

7. Conclusions and Outlook

- Pure anharmonic contributions appear as new combinations of single phonon spectral functions. Memory is always present.
- Correction to the 2 phonon expression arises from cross-correlation of $\mathbf{J}_{ha}$ and $\mathbf{J}_{anh}$.

References

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