Hydrodynamic finite-size scaling of the thermal conductivity in glasses

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Introduction - Background

- Thermal conductivity

$$J = -\kappa \nabla T$$

- Linear Response: Quantum Green-Kubo (GK) formalism [1]:

$$\kappa = \frac{1}{VT}\int_0^{\infty} dt \int_0^{1/k_b T} d\lambda \langle \hat{J}(t - i\hbar\lambda)\hat{J}\rangle$$

$\hat{J}$ is the **heat flux** operator. In solid insulators, it depends on **lattice vibrations**:
Single-mode relaxation-time approximation: GK $\longrightarrow$ Quasi Harmonic Green Kubo (**QHGK**) method for **crystals** and **glasses** alike [2-3]:

$$\kappa = \frac{1}{V}\sum_{\mathbf{q}\nu\nu'} C_{\mathbf{q}\nu\nu'} v_{\mathbf{q}\nu\nu'} v_{\mathbf{q}\nu'\nu} \frac{\gamma_{\mathbf{q}\nu} + \gamma_{\mathbf{q}\nu'}}{(\omega_{\mathbf{q}\nu} - \omega_{\mathbf{q}\nu'})^2 + (\gamma_{\mathbf{q}\nu} + \gamma_{\mathbf{q}\nu'})^2}$$

QHGK scales as N^3_{atoms} for glasses at Γ, i.e. $\mathbf{q} = (0,0,0)$. This **drastically limits** our computational ability to systems of a few thousand atoms, that are hardly converged in size. We overcome this issue by mapping the low-energy glassy normal modes to acoustic sound waves, which allows for a simple hydrodynamic extrapolation:

$$\kappa_{\mathrm{hydro}} = \sum_{b=L,T} \frac{c_b^2}{3}\int_0^{\omega_P} C(\omega)\rho_b(\omega)\frac{1}{2\Gamma_b(\omega)}\mathrm{d}\omega + \frac{1}{3V}\sum_{\nu,\nu'}\Theta(\omega_{\nu,\nu'} - \omega_P) C_{\nu\nu'} v_{\nu\nu'} v_{\nu'\nu}\tau_{\nu\nu'}$$

Zoology of glassy vibrations: propagons, diffusons, and locons

Normal modes in glasses can be grouped into low-energy delocalized modes, the **propagons**, and progressively more energetic and more localized modes, **diffuson** and **locons** [4].
This shows in the **Dynamical Structure Factor** (DSF)

$$S_b(\omega, \mathbf{Q}) = \sum_\nu \frac{1}{\pi}\frac{\gamma_\nu}{\gamma_\nu^2 + (\omega - \omega_\nu)^2}|\langle \nu|\mathbf{Q}, b\rangle|^2$$

where $\langle \nu|\mathbf{Q}, b\rangle$ projects the a plane-wave of wavevector $\mathbf{Q}$ and polarization b onto the νth normal mode, and γ_ν is the νth normal mode's linewidth.
Propagons are **almost plane waves**:

$$S_b(\omega, \mathbf{Q}) \propto \frac{2\Gamma_b(Q)}{(\omega - c_b Q)^2 + \Gamma_b^2(Q)}$$

The DSF features an almost linear dispersion with a broadening $\Gamma_b(Q)$.

An effective model for propagons

For propagons one can use a plane-wave basis, so the **energy flux** reads

$$\hat{J} = \frac{1}{V}\sum_{\mathbf{Q},b,b'} J_{\mathbf{Q}}^{bb'} \hat{a}^\dagger_{\mathbf{Q}b}\hat{a}_{\mathbf{Q}b'}$$

The **two-point correlation function** is thus given by:

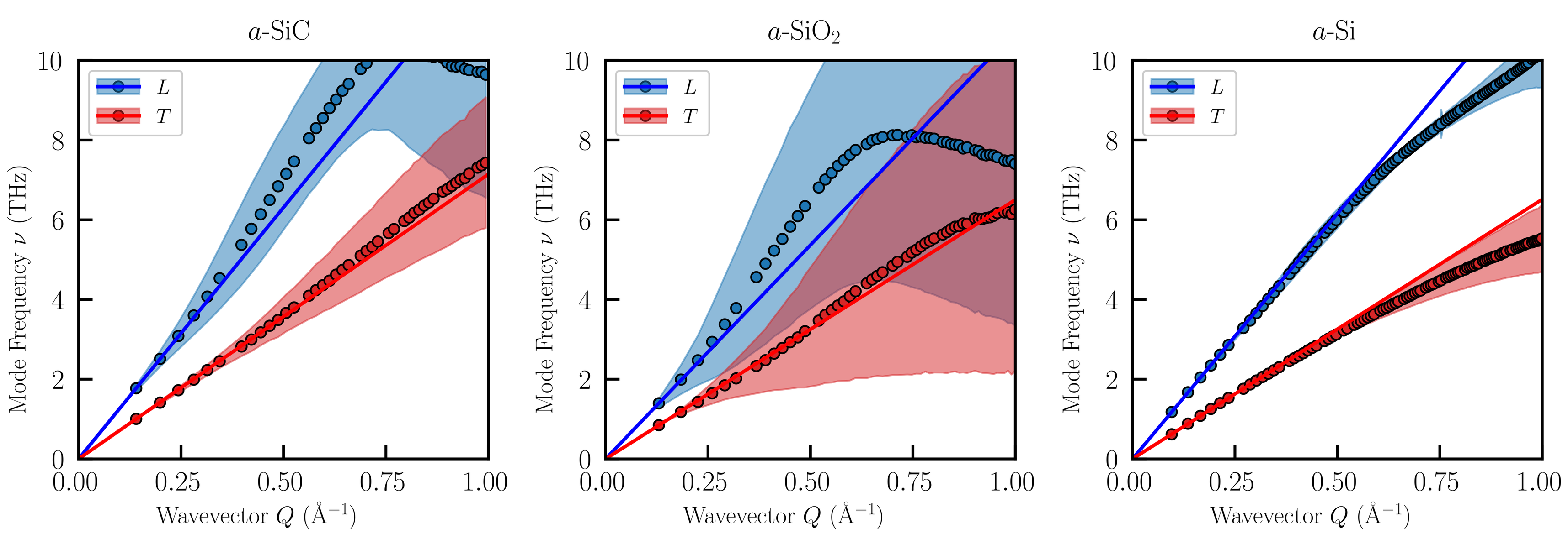
$$\begin{aligned}\langle \hat{a}_{\mathbf{Q}b}(t)\hat{a}^\dagger_{\mathbf{Q}b'}\rangle &\approx \delta_{bb'}\frac{1}{2\pi}\int e^{-i\omega t} S_b(\omega, \mathbf{Q})(n(\omega, T) + 1)\,\mathrm{d}\omega \\ &\approx \delta_{bb'}(n(c_b Q, T) + 1)e^{-ic_b Qt - \Gamma_b(Q)|t|}\end{aligned}$$

that leads to an expression for the **thermal conductivity of propagons** given by

$$\kappa_P = \frac{1}{3V}\sum_{\mathbf{Q},b} C(c_b Q) c_b^2 \frac{1}{2\Gamma_b(Q)}$$

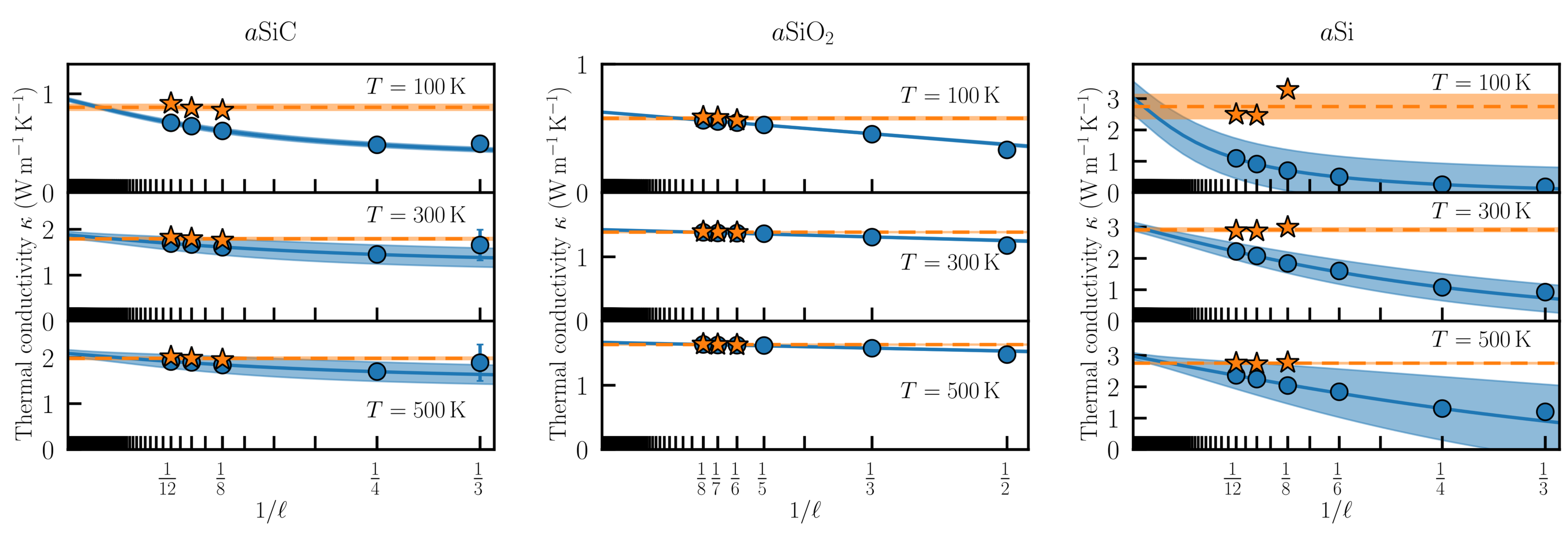
The broadening of the DSF determines the thermal conductivity of propagons. This **includes both anharmonicity and disorder**.

Dynamical Structure Factor for a-SiC, a-SiO$_2$, a-Si



Different materials display different dispersions. Amorphous silica (a-SiO$_2$) is **dominated by the disorder**. Size effects are expected to be quite small, since wave propagation is hindered by the large disorder. In amorphous silicon (a-Si), disorder is **far less prominent**. Here size effects are expected to be important. Amorphous silicon carbide (a-SiC) is something in between the two: it should serve us as a **bridge** between the two extrema.

Hydrodynamic extrapolation of κ

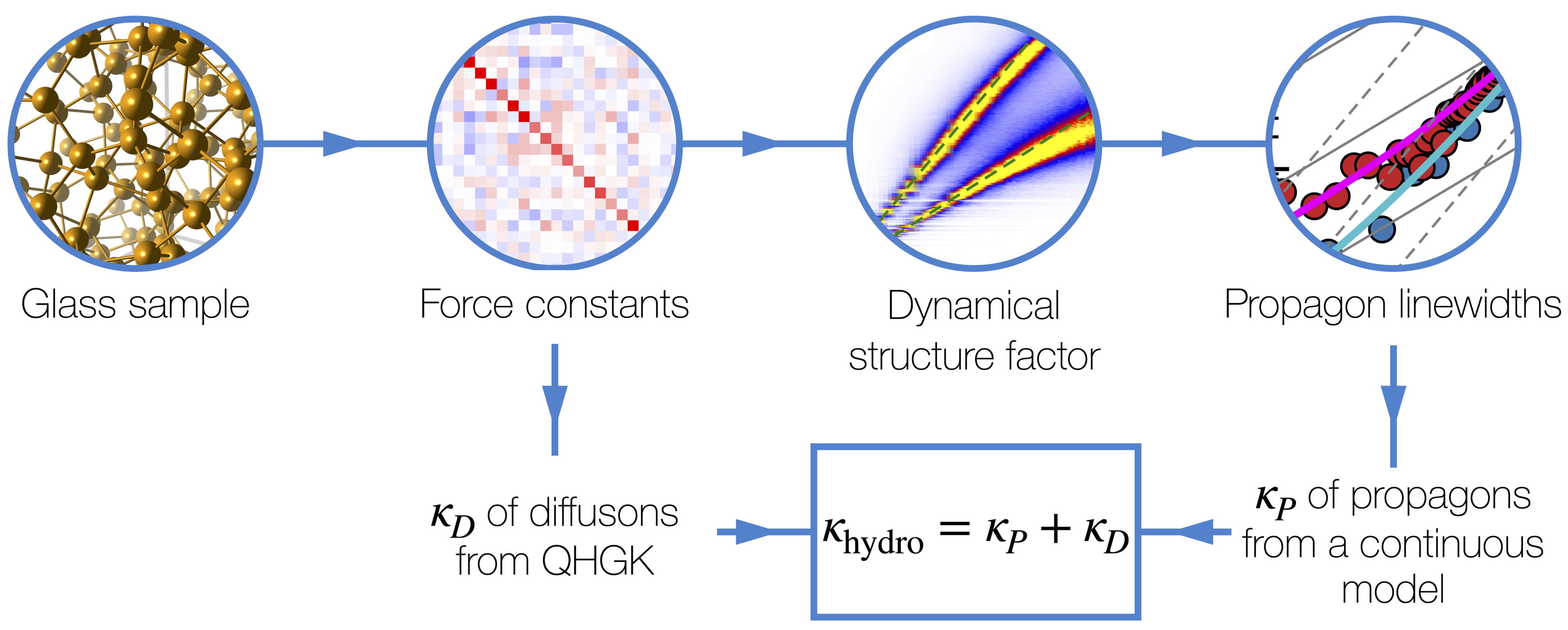


The model for propagons **accurately extrapolates** the infinite-size thermal conductivity of paradigmatic amorphous solids. Depending on the material, models with up-to-few thousand atoms are still required in order to achieve convergence. $\ell \propto N^{1/3}$

Conclusions

- Propagons can be described by a simple Debye-like model of acoustic sound waves with a **lifetime due to both disorder and anharmonic effects**.
- For each branch, the speed of sound and the Q-dependent lifetimes are obtained from the **DSF** of relatively **small models**.
- Through hydrodynamic extrapolation, size convergence is achieved for the thermal conductivity of three paradigmatic glasses.

Summary of the workflow



References

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