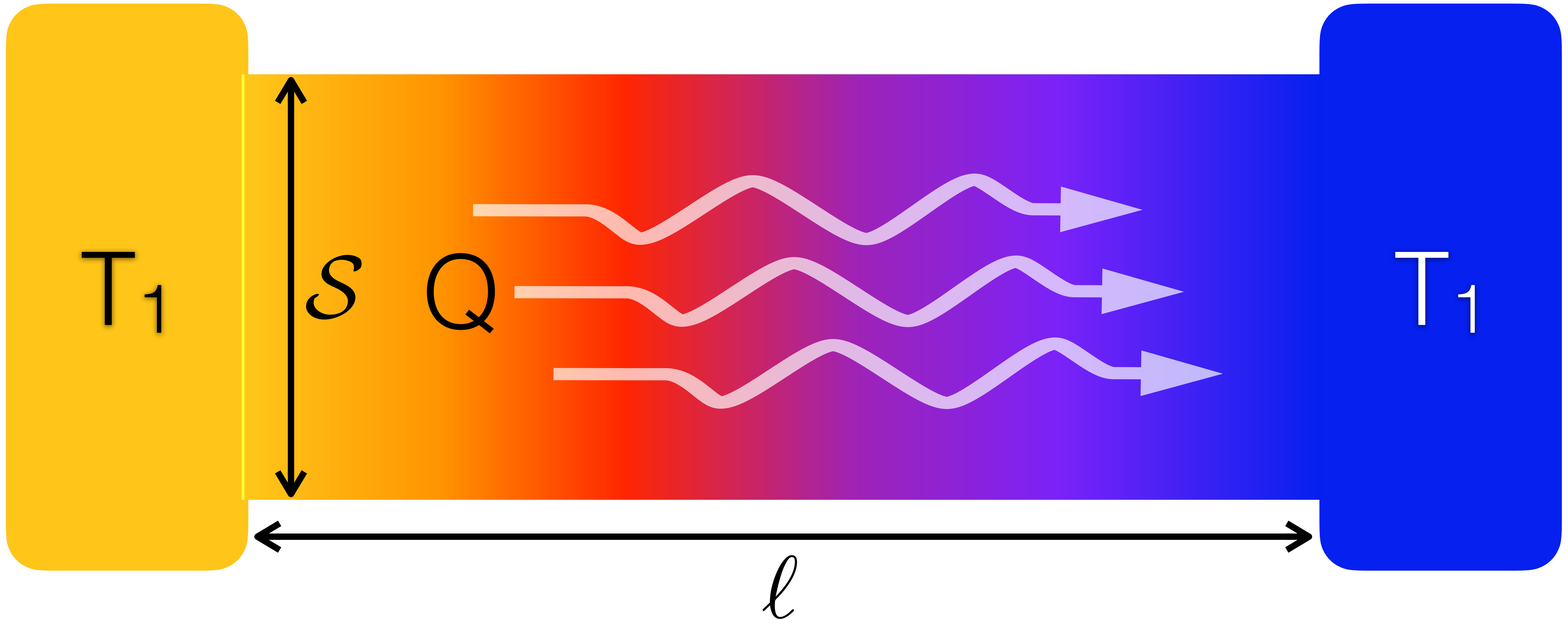
heat transport in ill-condensed matter

Stefano Baroni
Scuola Internazionale Superiore di Studi Avanzati
Trieste — Italy

Frontiers in Thermal and Electronic Transport in Materials: a Tribute to Nicola Bonini, King's College London, April 11-12, 2024

disclaimer

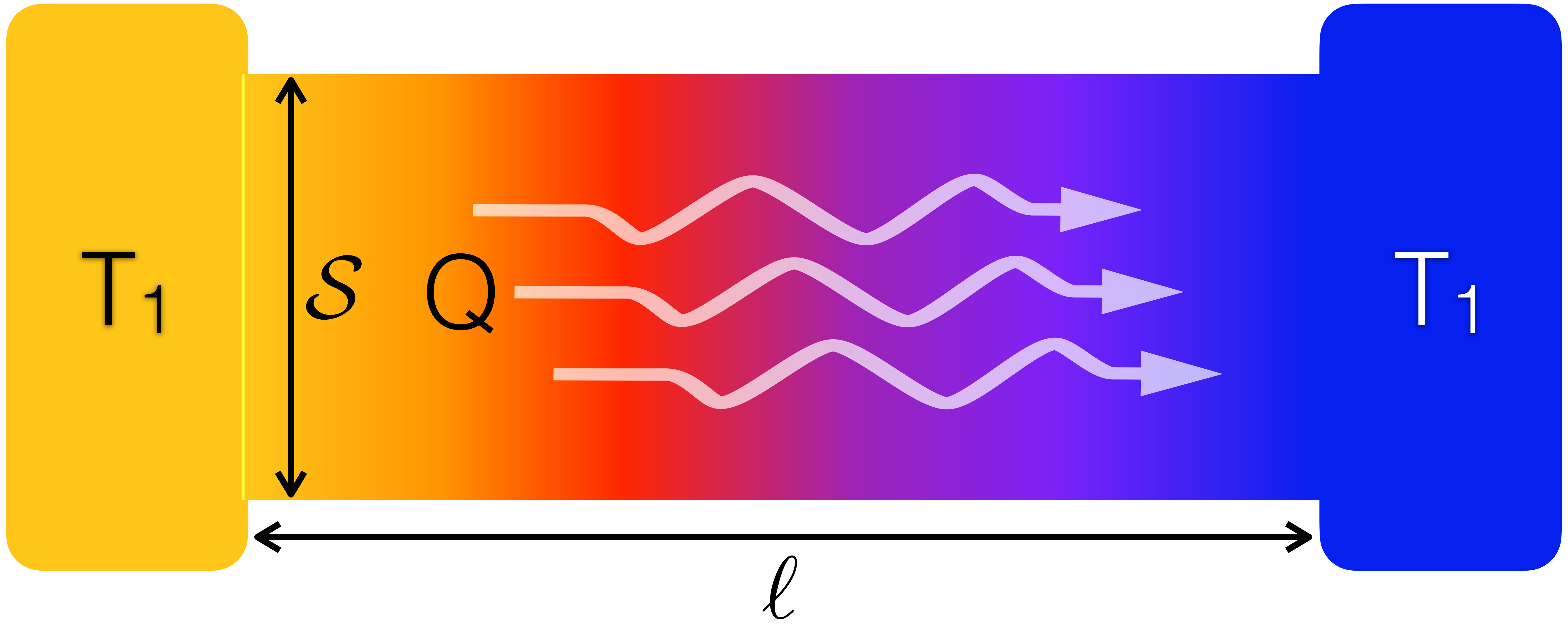
what heat transport is all about



$$\frac{1}{\mathcal{S}}\frac{dQ}{dt} = -\kappa\frac{(T_2 - T_1)}{\ell}$$

heat flows from warmth to coolth
as time flows from the past to the future

what heat transport is all about



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Fourier Law

$$\boldsymbol{J} = -\kappa \nabla T$$

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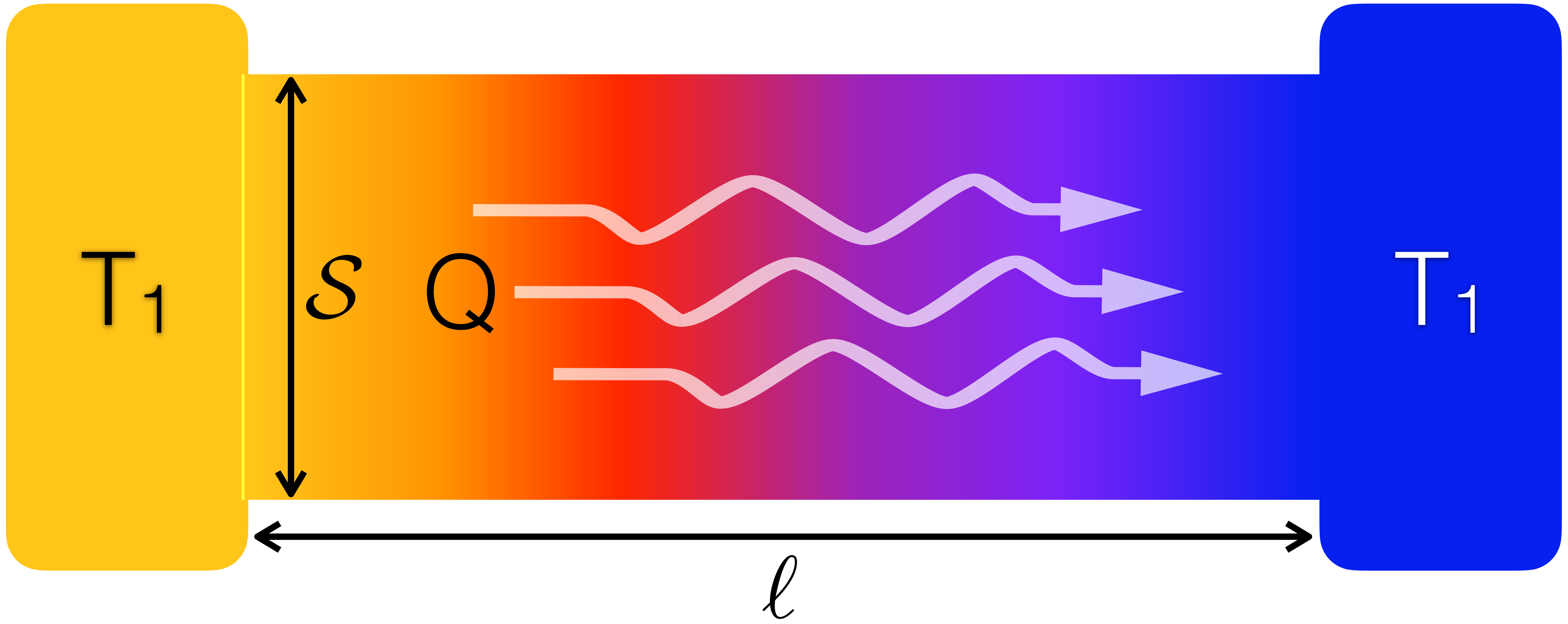

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Fourier Law

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Green-Kubo

$$\kappa = \frac{\Omega}{3k_B T^2}\int_0^\infty \langle \boldsymbol{J}(t)\cdot\boldsymbol{J}(0)\rangle dt$$

heat transport in solid (non-diffusive) media

$$\begin{aligned}\boldsymbol{J} &= \frac{d}{dt}\sum_n \boldsymbol{R}_i e_i \\ &= \sum_i \left(\dot{\boldsymbol{R}}_i e_i + \boldsymbol{R}_i \dot{e}_i\right)\end{aligned}$$

heat transport in solid (non-diffusive) media

$$\boldsymbol{J} = \sum_i \left(\dot{\boldsymbol{R}}_i e_i + \boldsymbol{R}_i \dot{e}_i \right)$$

$$\boxed{\boldsymbol{R}_i = \boldsymbol{R}_i^\circ + \boldsymbol{u}_i}$$

$$= \sum_i \boldsymbol{R}_i^\circ \dot{e}_i + \frac{d}{dt} \sum_i \boldsymbol{u}_i e_i$$

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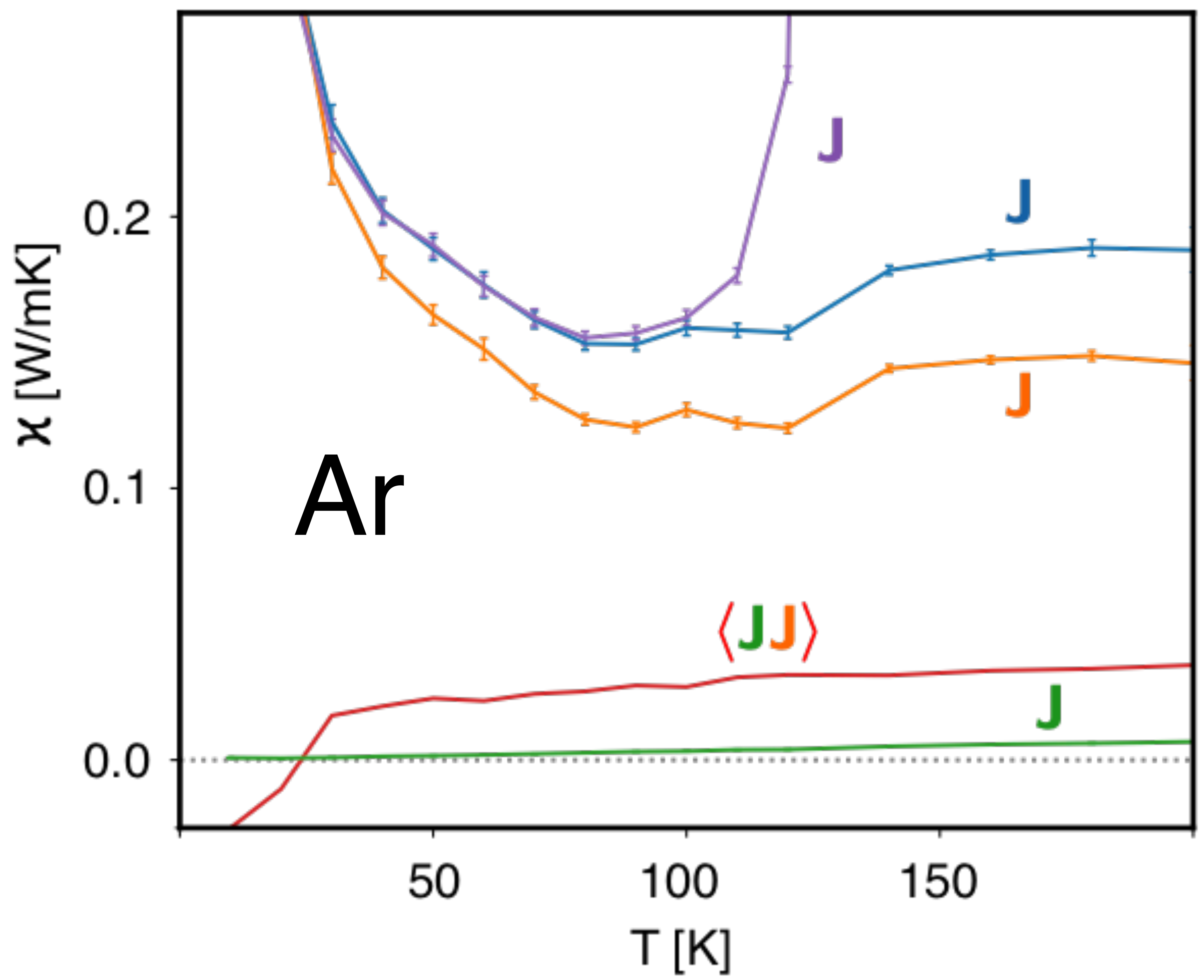

heat transport in solid (non-diffusive) media

$$\boldsymbol{J} = \sum_i \boldsymbol{R}_i^\circ \dot{e}_i$$

$$e_i \approx \frac{1}{2}\sum_\alpha \dot{u}_{i\alpha}^2 + \frac{1}{2}\sum_{\alpha\beta,j\neq i} \Phi_{i\alpha}^{j\beta} u_{i\alpha} u_{j\beta}$$

$$J_\alpha \approx \frac{1}{2}\sum_{ij\beta\gamma} \left(R_{i\alpha}^\circ - R_{j\alpha}^\circ\right) \Phi_{i\beta}^{j\gamma} u_{i\beta} \dot{u}_{j\gamma}$$

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$$\begin{aligned} J_\alpha &\approx \frac{1}{2} \sum_{ij\beta\gamma} \left(R_{i\alpha}^\circ - R_{j\alpha}^\circ \right) \Phi_{i\beta}^{j\gamma} u_{i\beta} \dot{u}_{j\gamma} \\ &= \sum_{nm} v_{nm}^\alpha \sqrt{\omega_n \omega_m} q_n p_m \end{aligned}$$

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first real-space moments of the inter-atomic force constants

heat transport in solid (non-diffusive) media

$$J_\alpha = \sum_{nm} v_{nm}^{\alpha} \sqrt{\omega_n \omega_m} q_n p_m$$

$$\kappa \propto \int_0^\infty dt \int dq_\circ dp_\circ \underbrace{J(q_t p_t) J(q_\circ p_\circ)}_{\text{4-th order polynomial}} \underbrace{\mathrm{e}^{-\beta H(q_\circ p_\circ)}}_{\text{Gaussian}}$$

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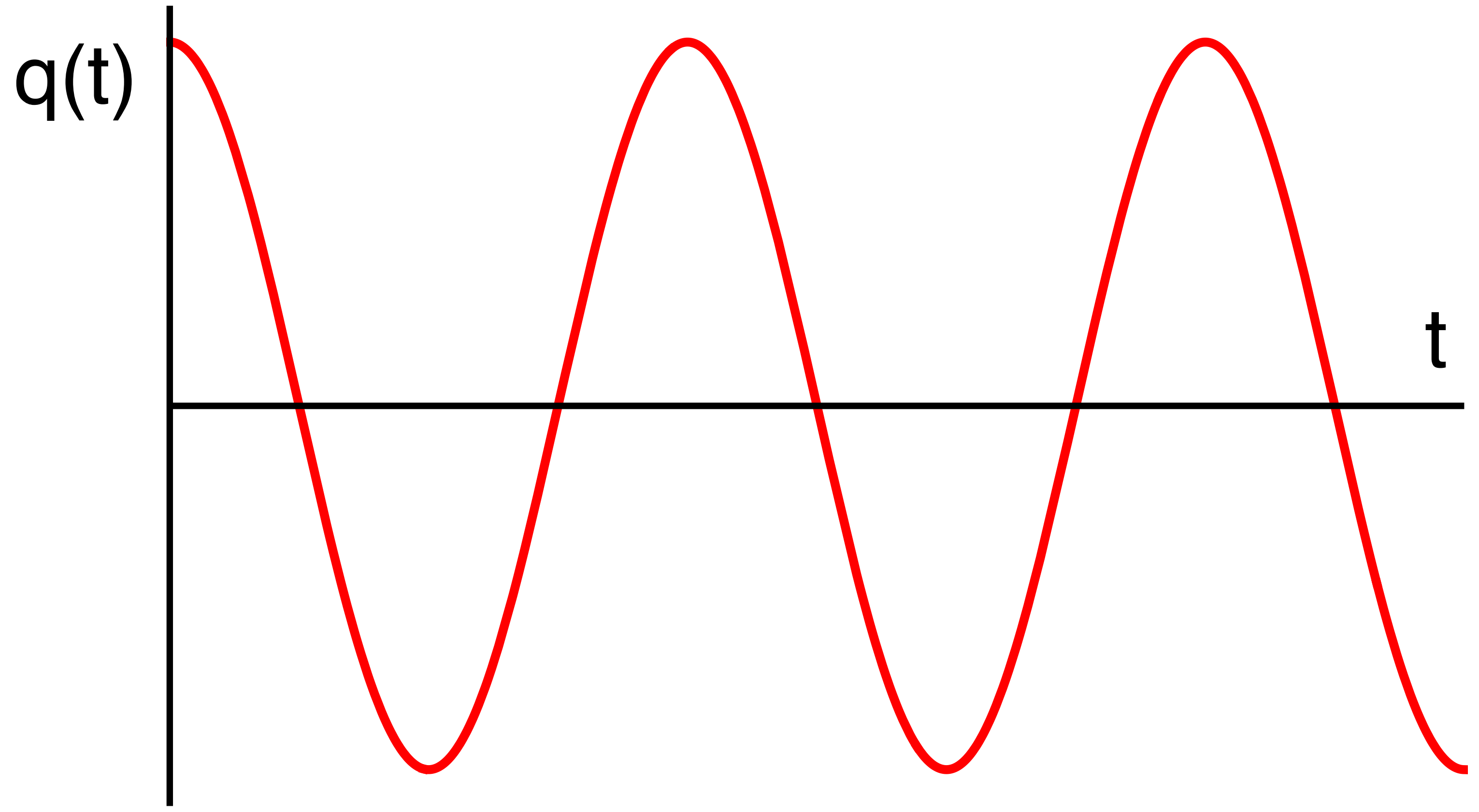

$\longrightarrow$

$$\kappa = \infty$$

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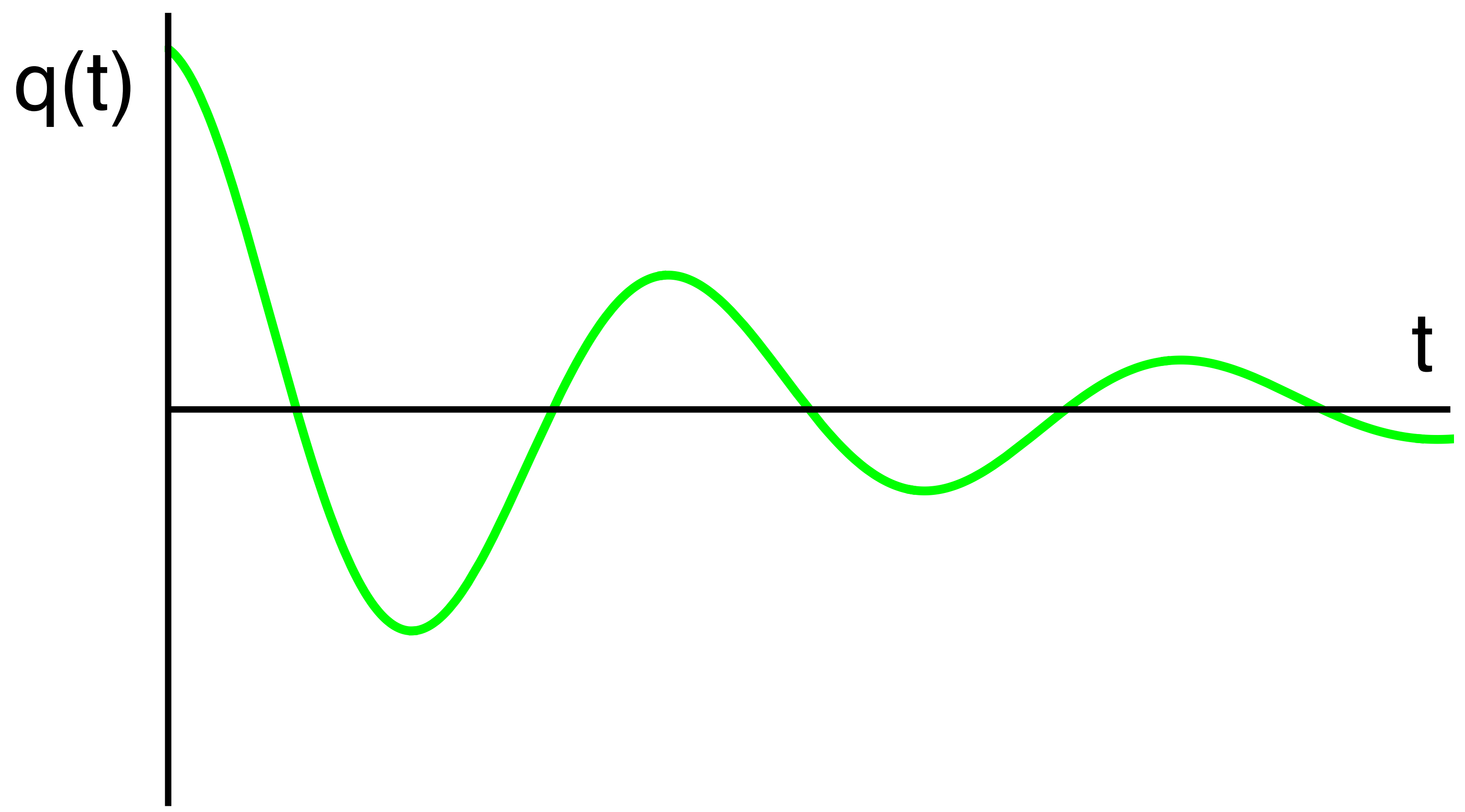

$$\omega \mapsto \omega + i\gamma \quad \longrightarrow$$

$$\kappa < \infty$$

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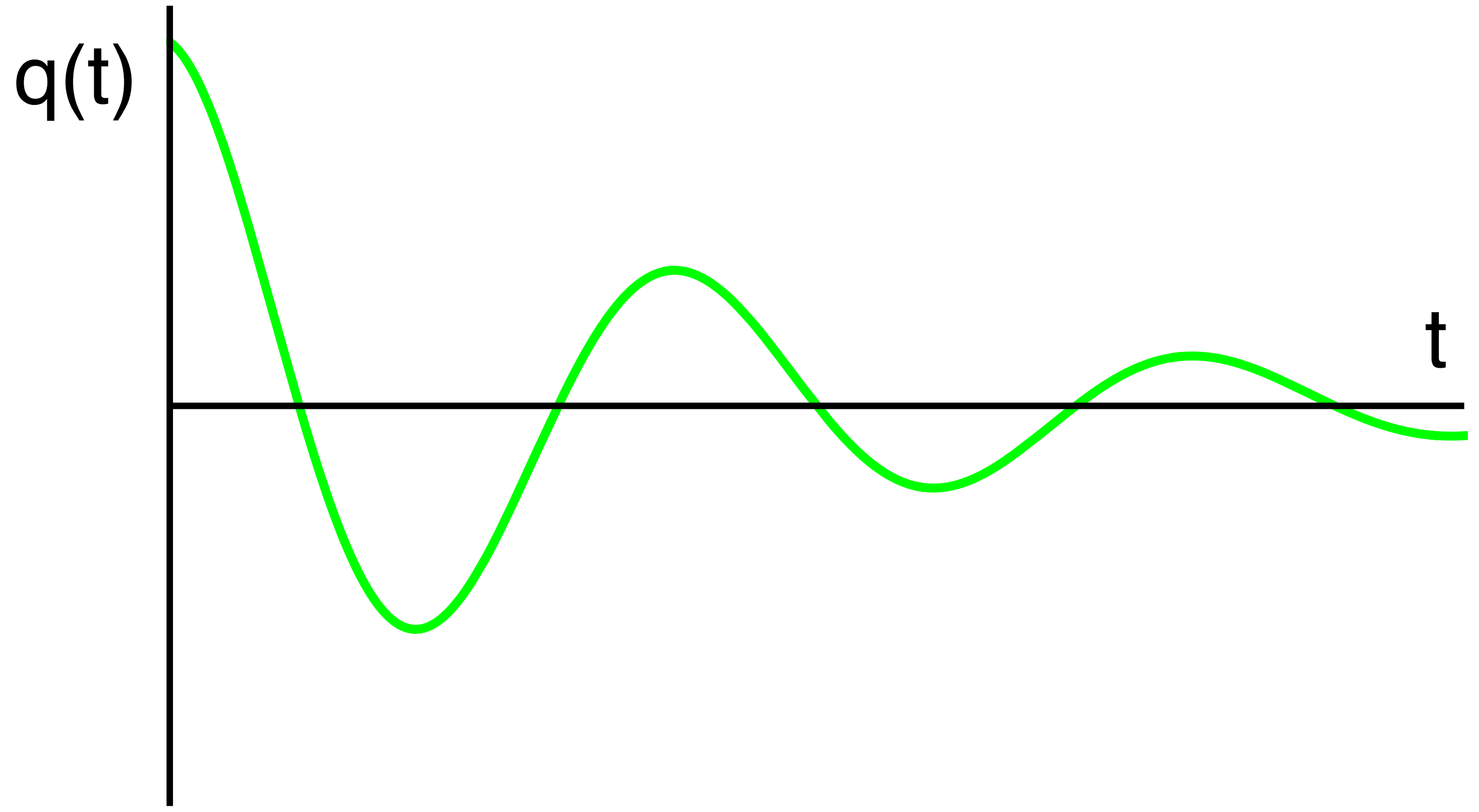

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$$\boxed{\kappa = \frac{1}{V} \sum_{nm} c_{nm} \big(v_{nm}\big)^2 \tau^\circ_{nm}}$$

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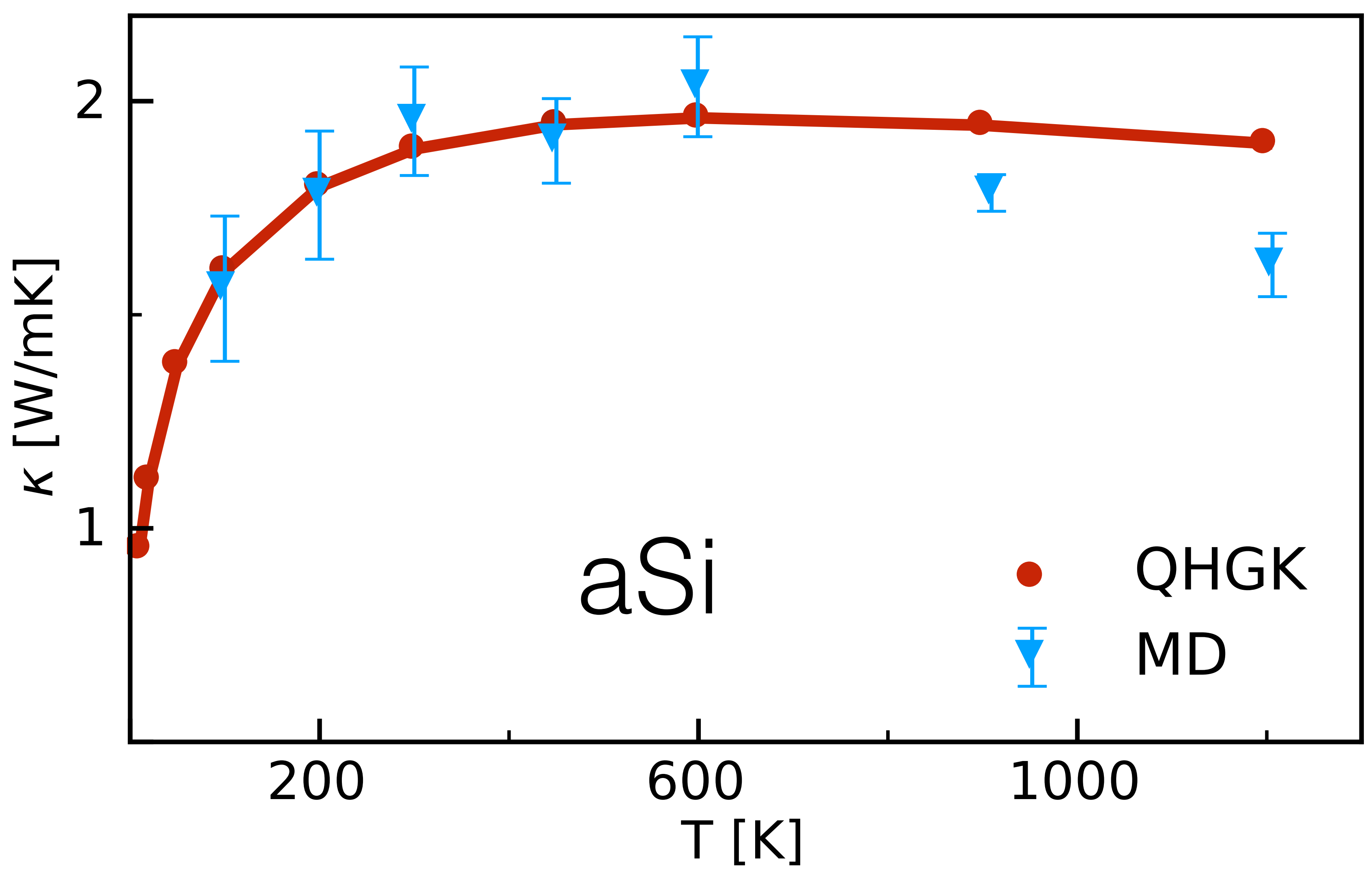
small γ

$$\approx \pi \delta(\omega_n - \omega_m)$$

$$\approx k_B \left(\frac{\hbar\omega_n}{k_B T}\right)^2 \frac{1}{\left(\mathrm{e}^{\frac{\hbar\omega_n}{k_B T}} - 1\right)^2}$$

heat transport in solid (non-diffusive) media

$$\kappa = \frac{1}{V}\sum_{nm} c_{nm}\left(v_{nm}\right)^2 \tau^\circ_{nm}$$



heat transport in solid (non-diffusive) media

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in crystals

$$v_{nn'} = v_n(\boldsymbol{q})\delta_{\nu\nu'}\delta_{\boldsymbol{q}\boldsymbol{q}'}$$

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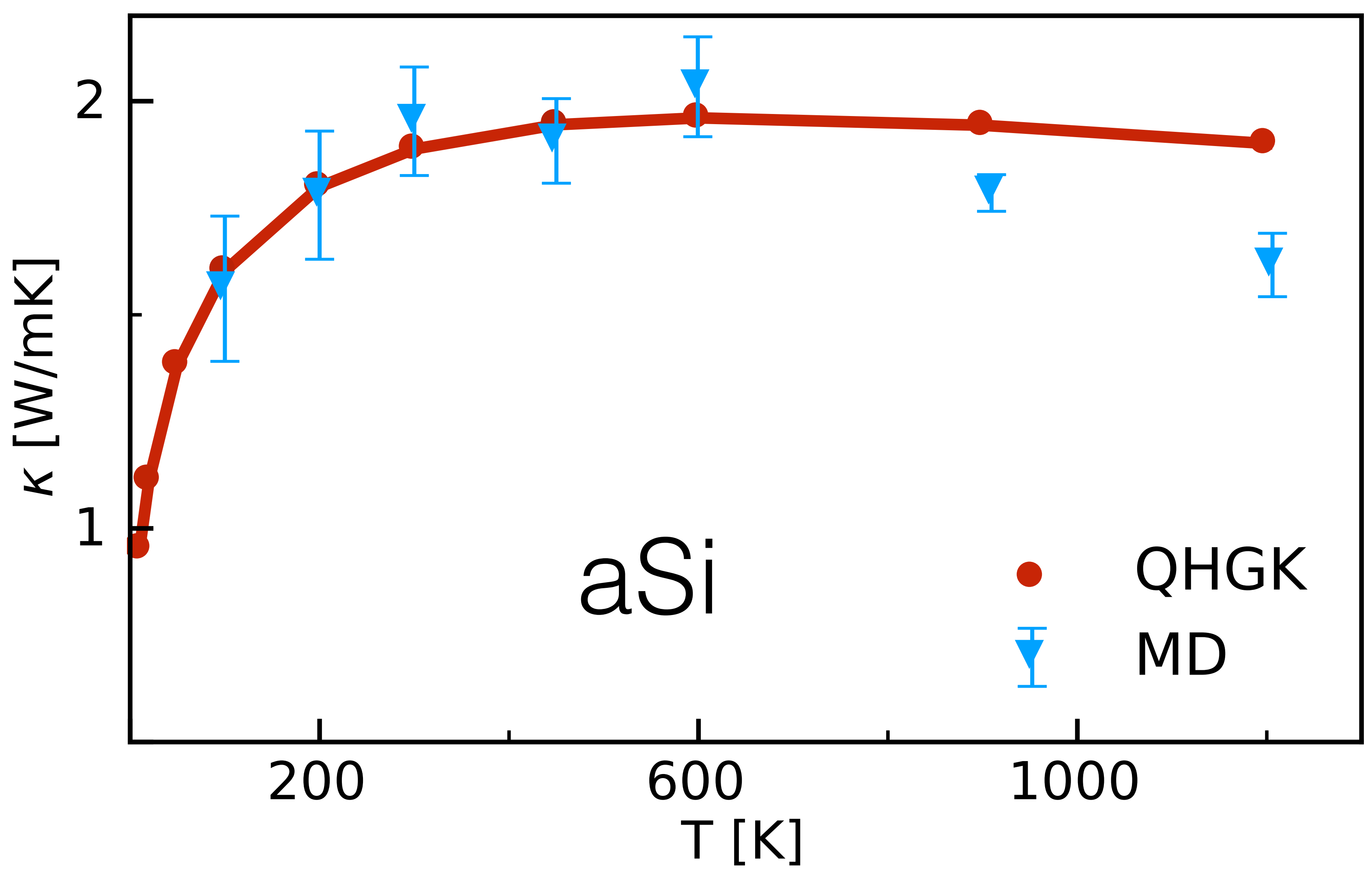

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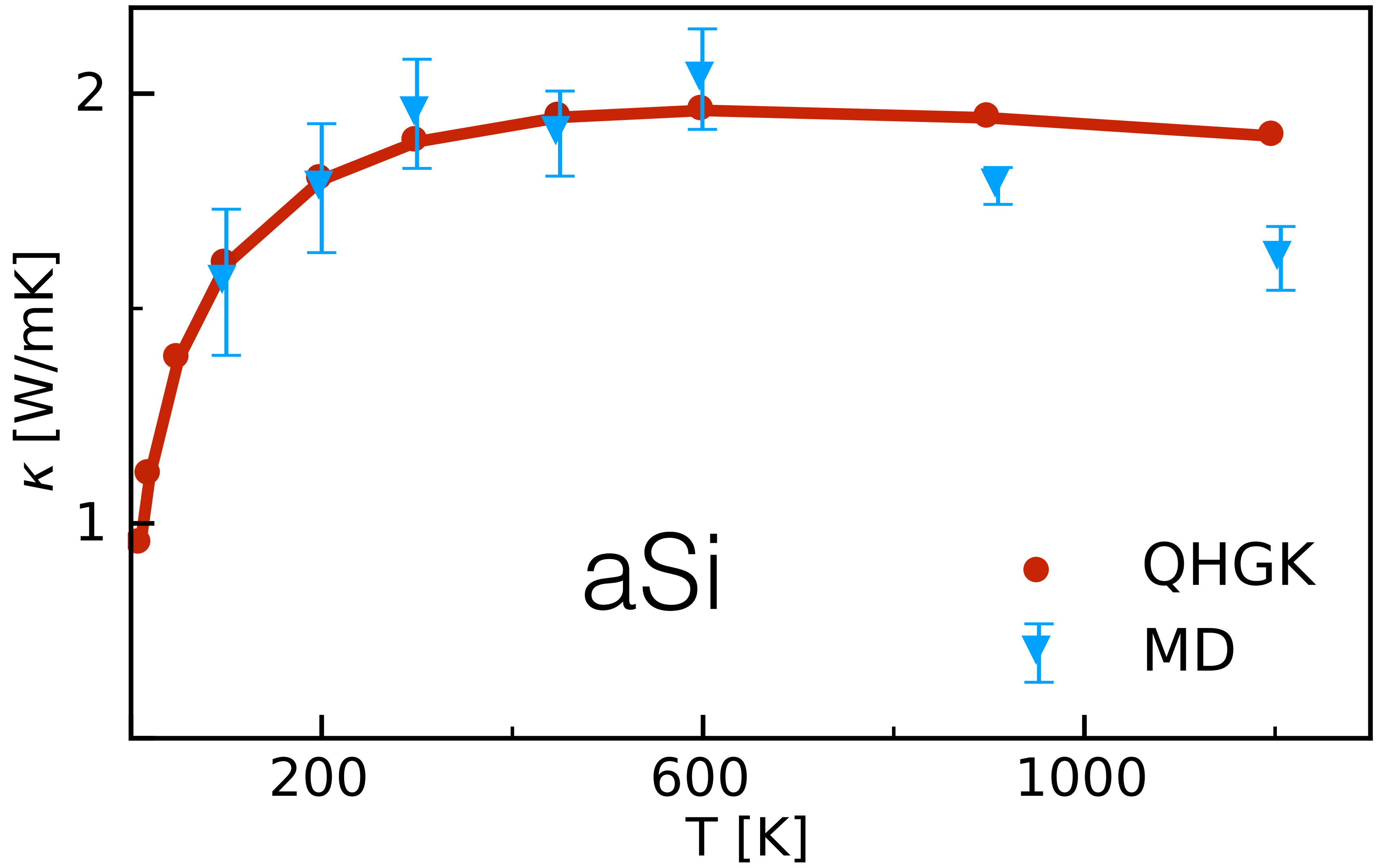

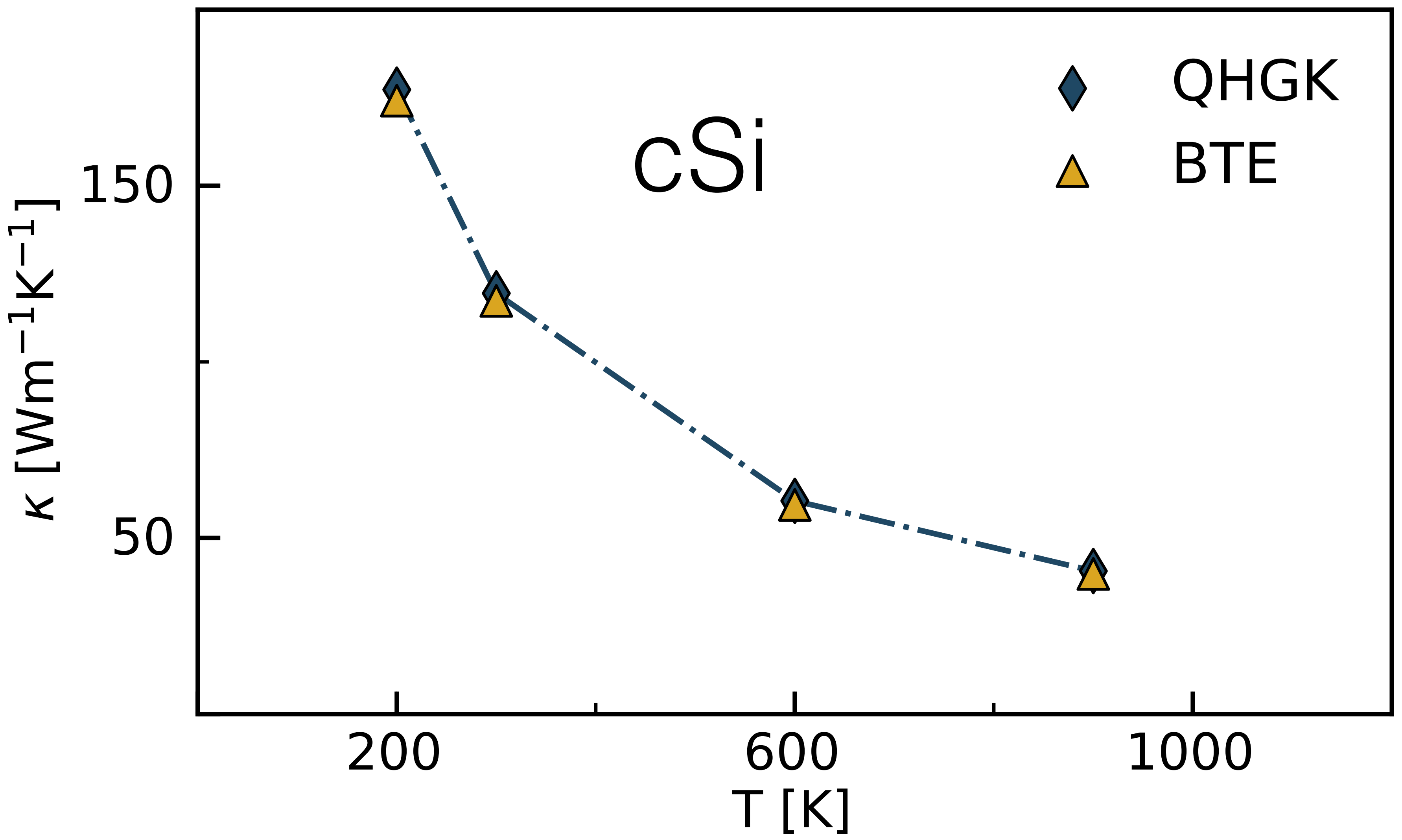

Isaeva, L., Barbalinardo, G., Donadio, D. & Baroni, S. *Modeling heat transport in crystals and glasses from a unified lattice-dynamical approach,* Nat. Commun. 10, 3853 (2019).

Simoncelli, M., Marzari, N. & Mauri, F. *Unified theory of thermal transport in crystals and disordered solids,* Nat. Phys. 15, 809–813 (2019).

heat transport in glasses

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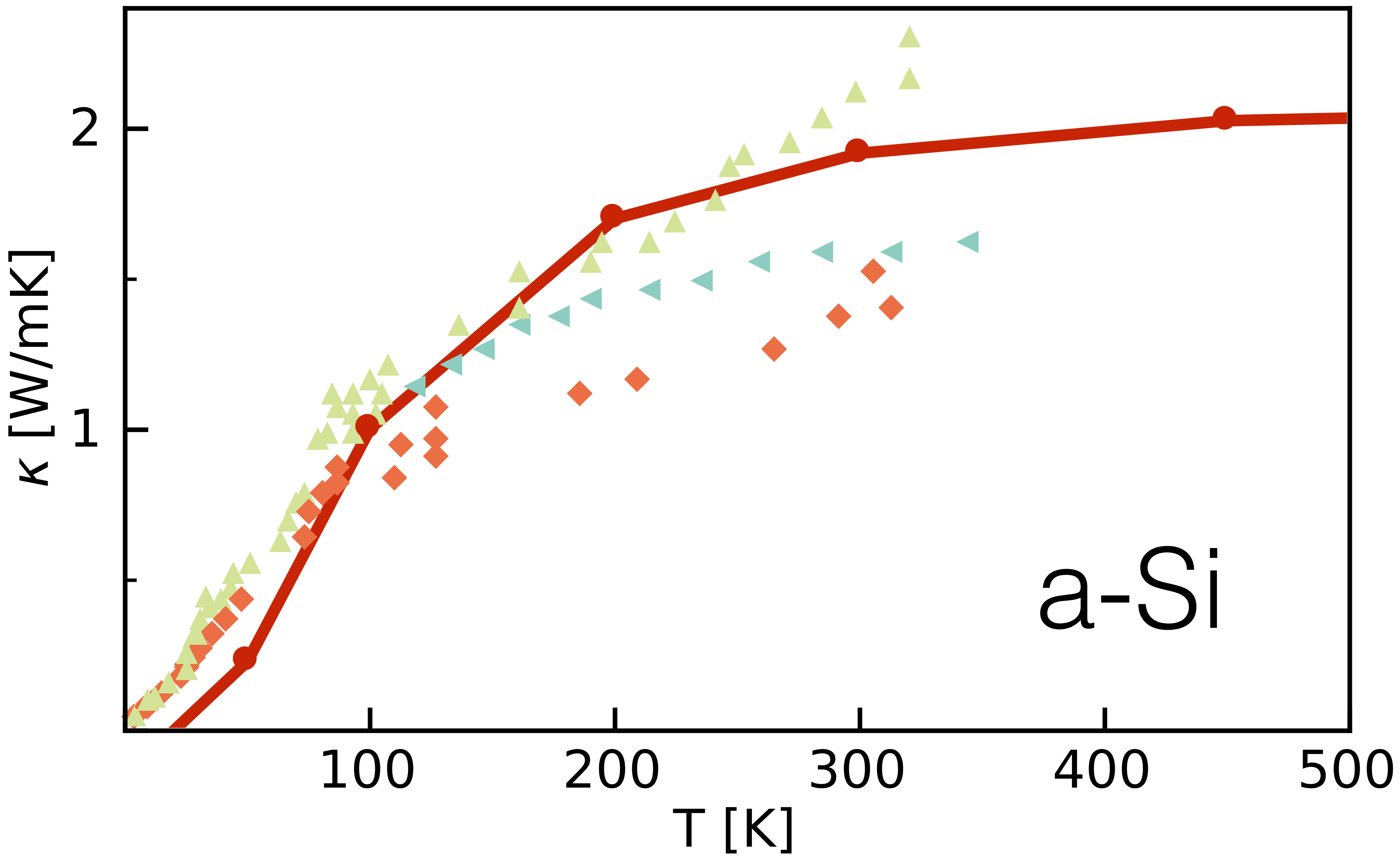

quantum
QHGK

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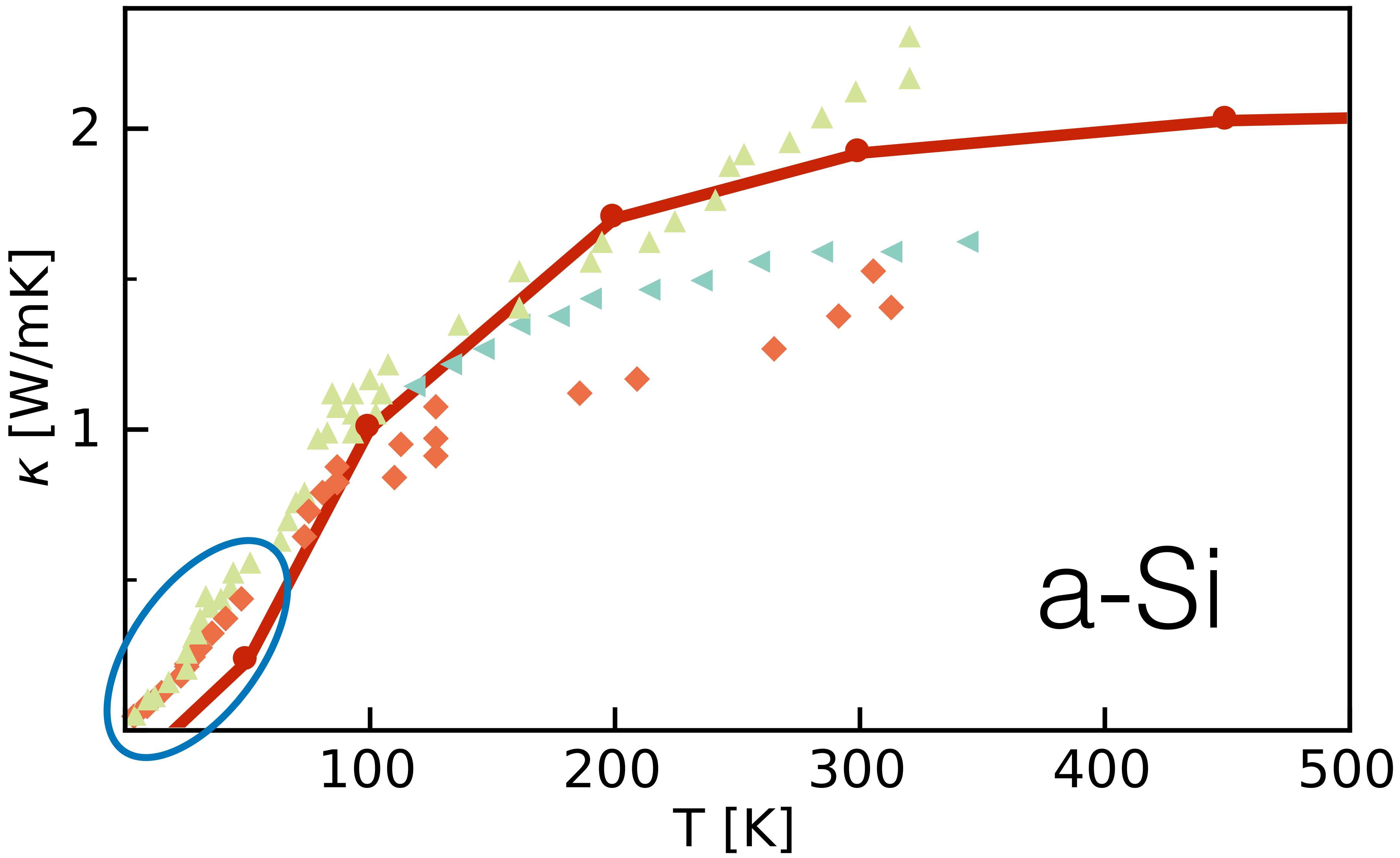

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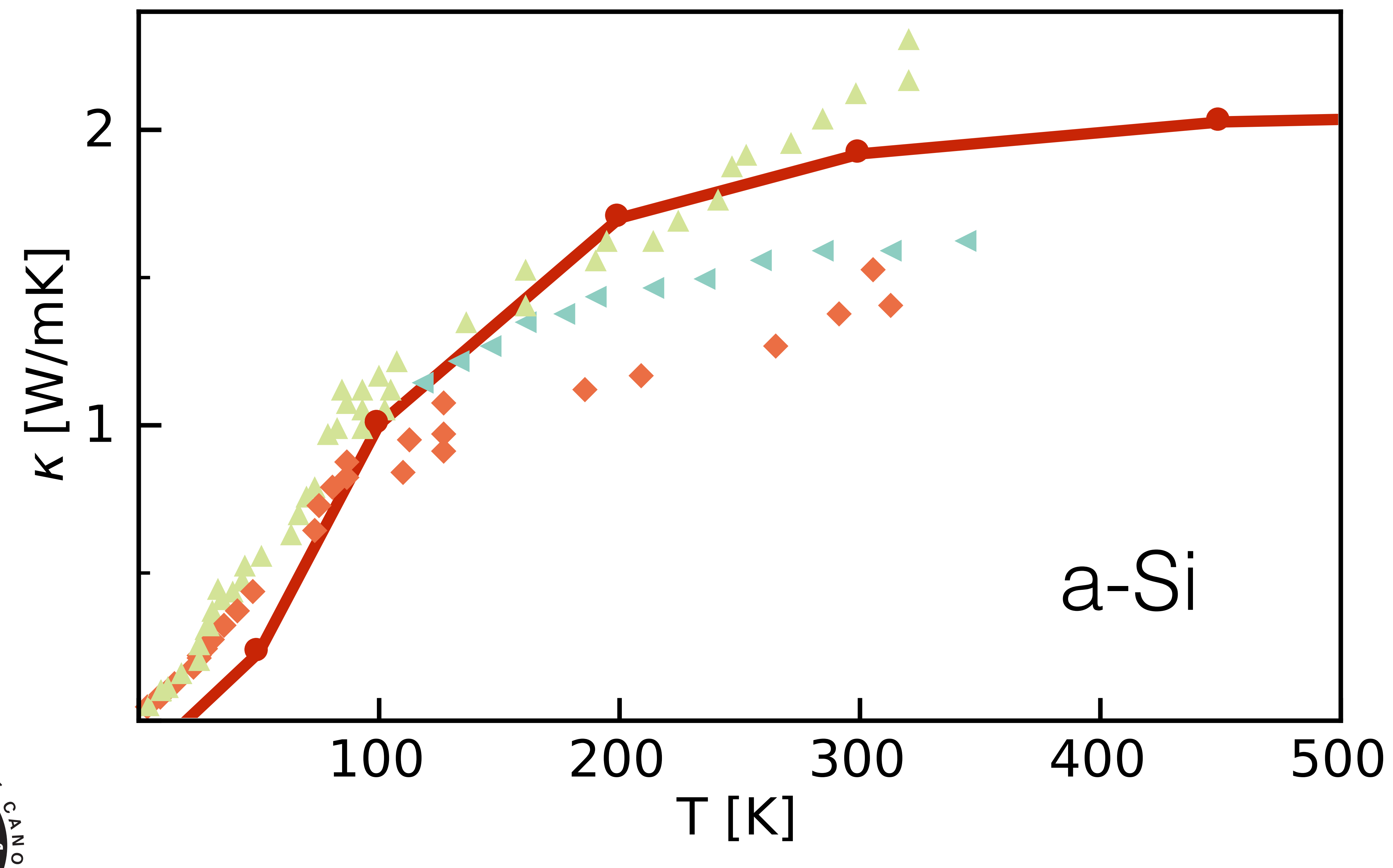
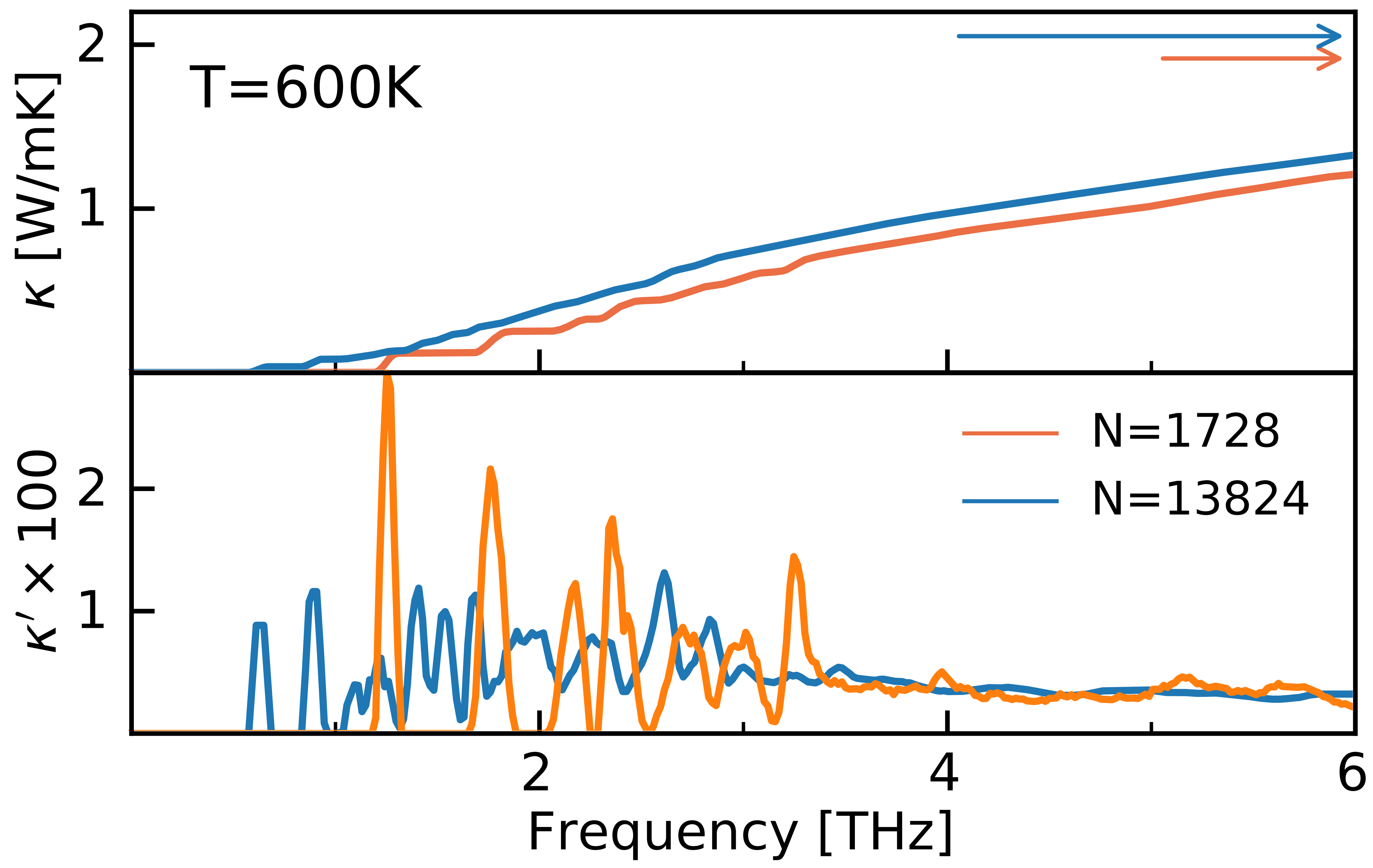
heat transport in glasses

$$\kappa = \frac{1}{V} \sum_{nm} c_{nm} \left(v_{nm} \right)^2 \tau^\circ_{nm}$$

$$\kappa'(\omega) = \frac{1}{V} \sum_{nm} \delta(\omega - \omega_n) c_{nm} \left(v_{nm} \right)^2 \tau^\circ_{nm}$$

$$\kappa(\omega) = \int_0^\omega \kappa'(\omega') d\omega'$$





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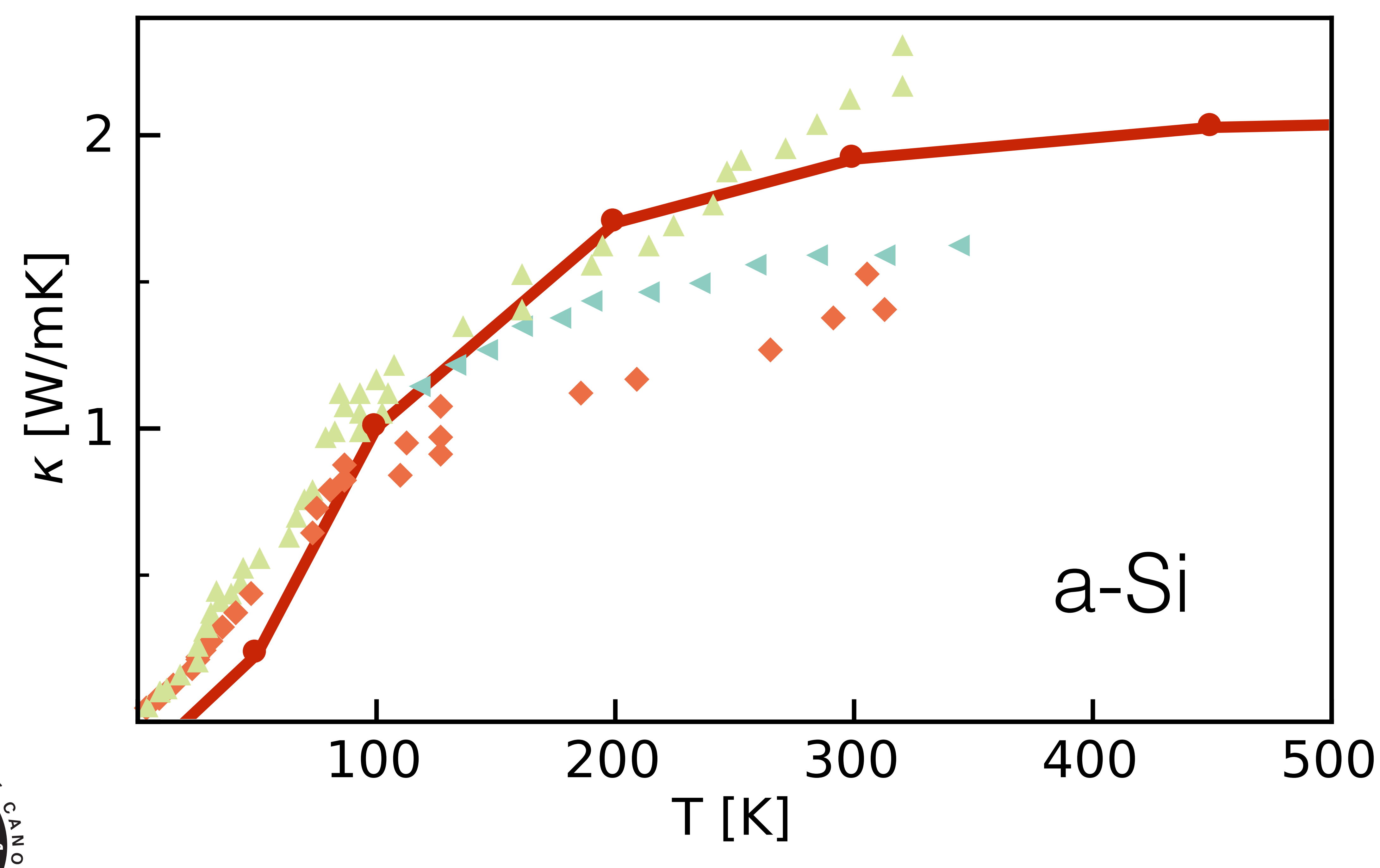

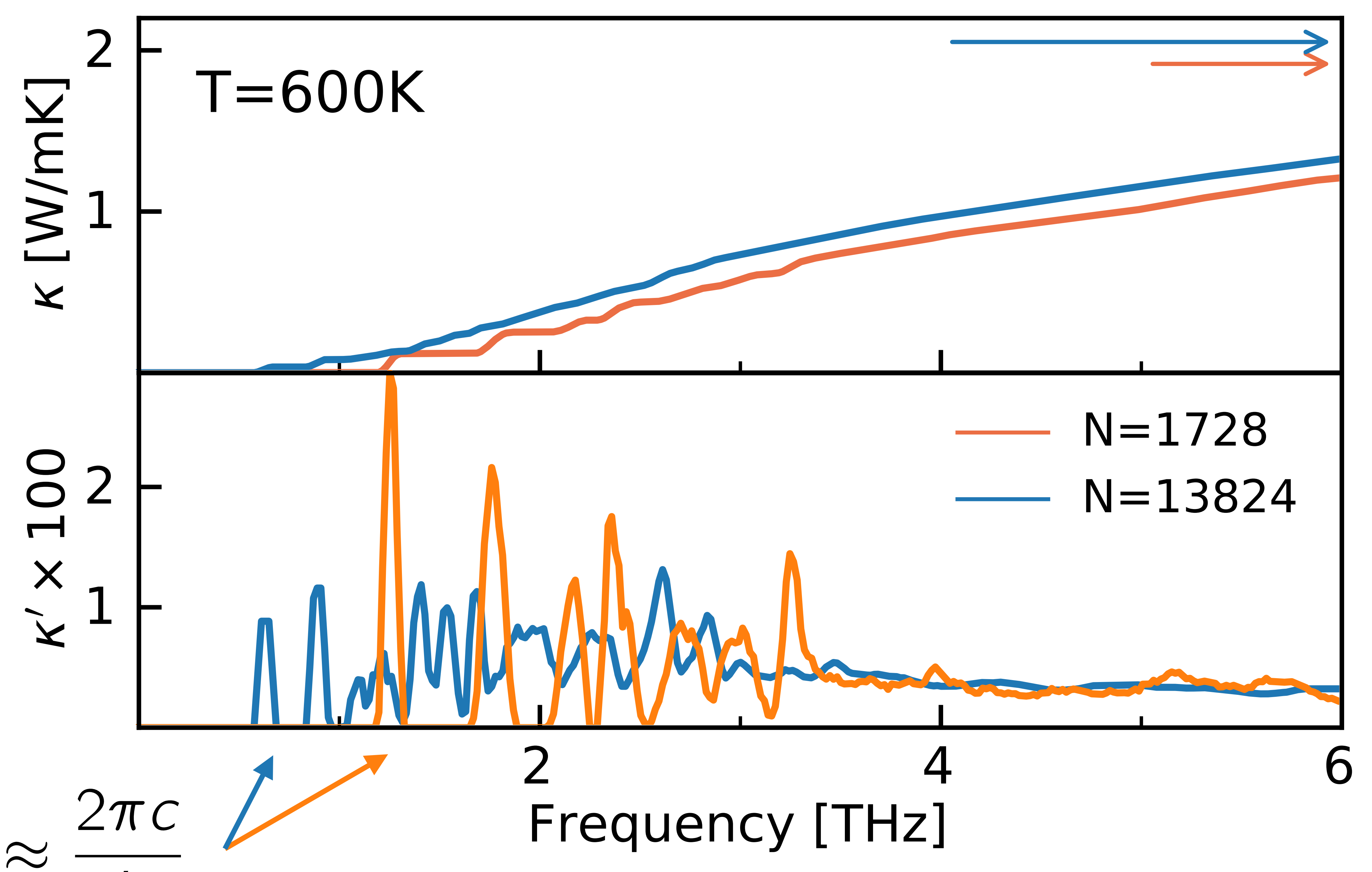

$$\omega_{min} \approx \frac{2\pi c}{L}$$

heat transport in glasses

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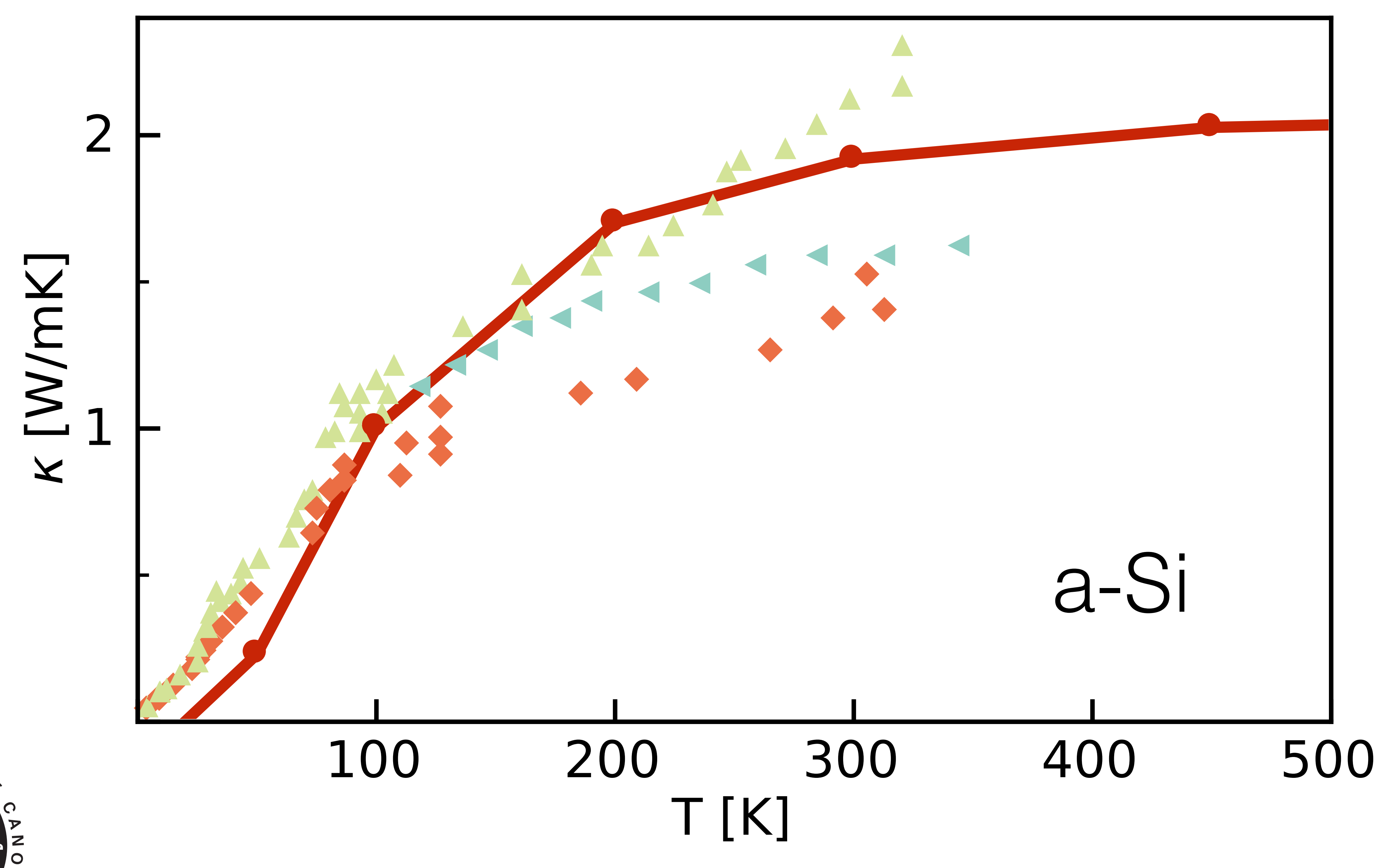

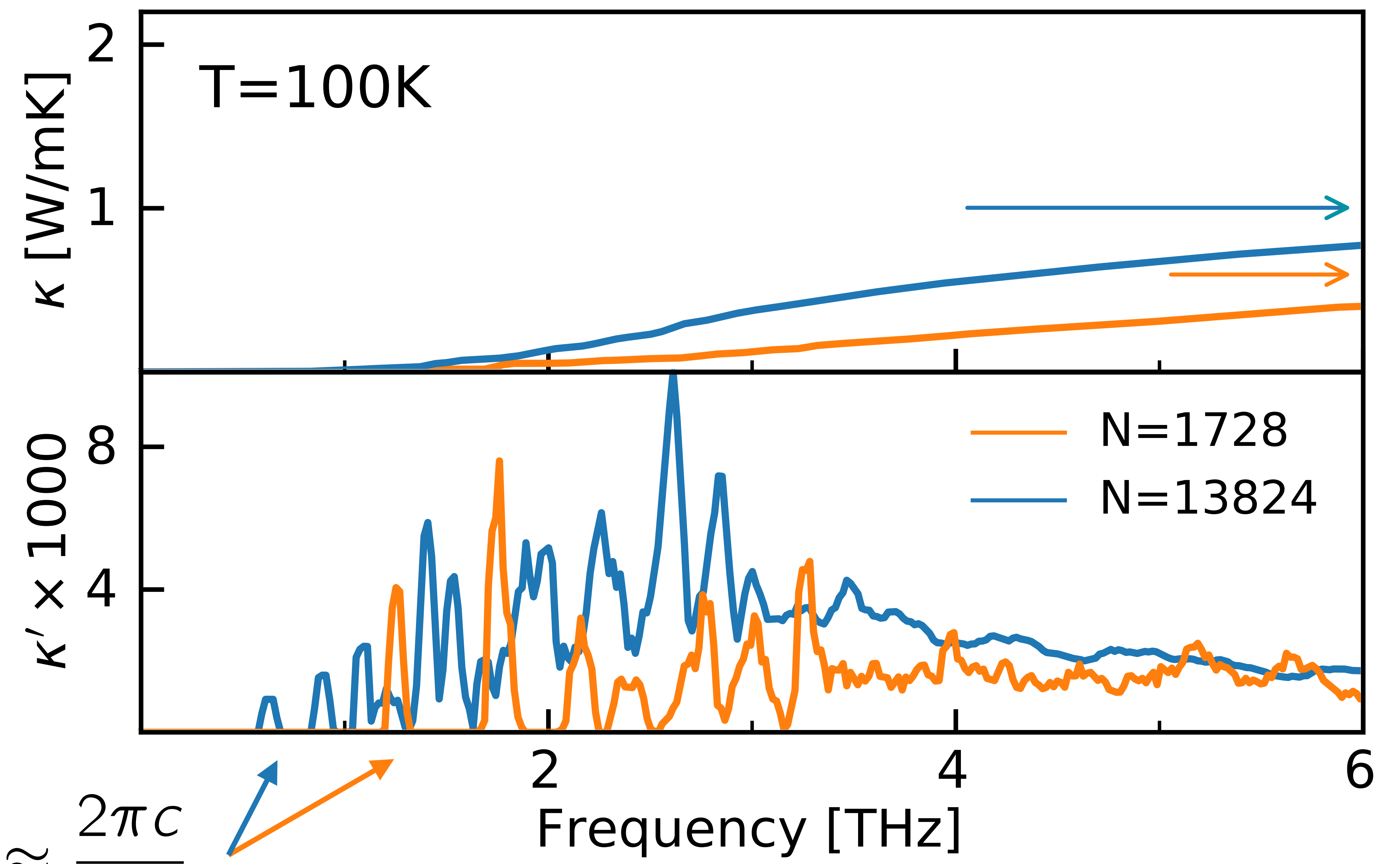

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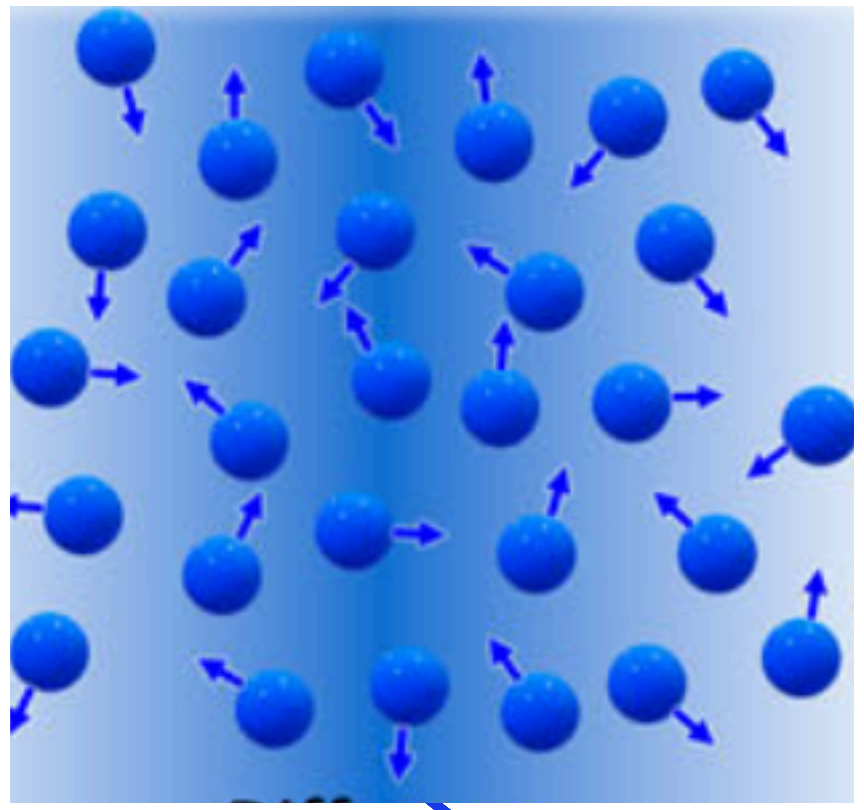
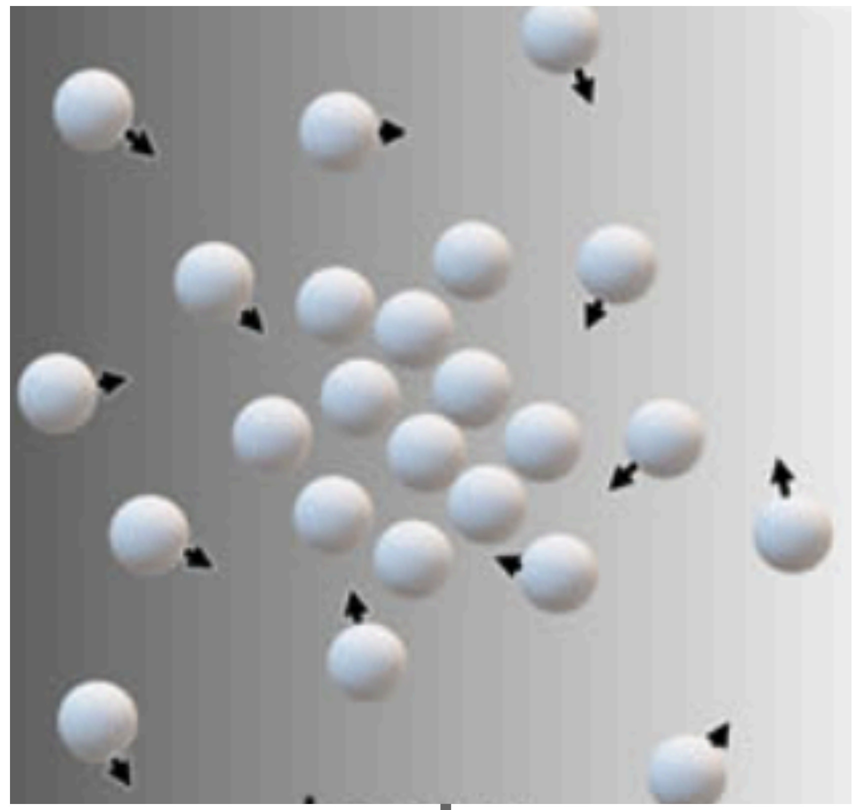
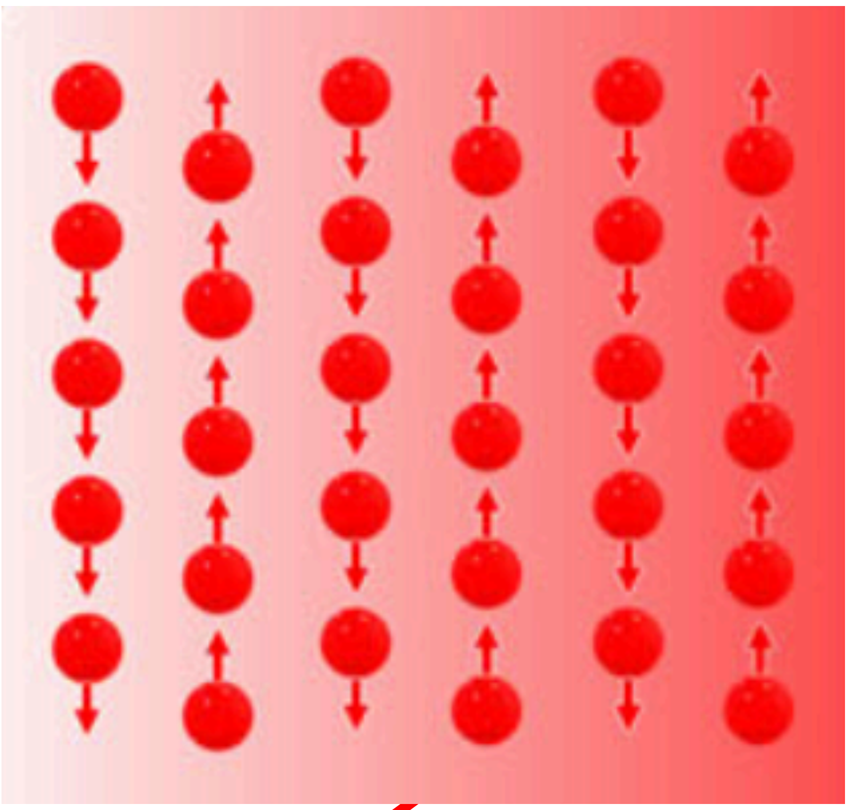
finite-size scaling

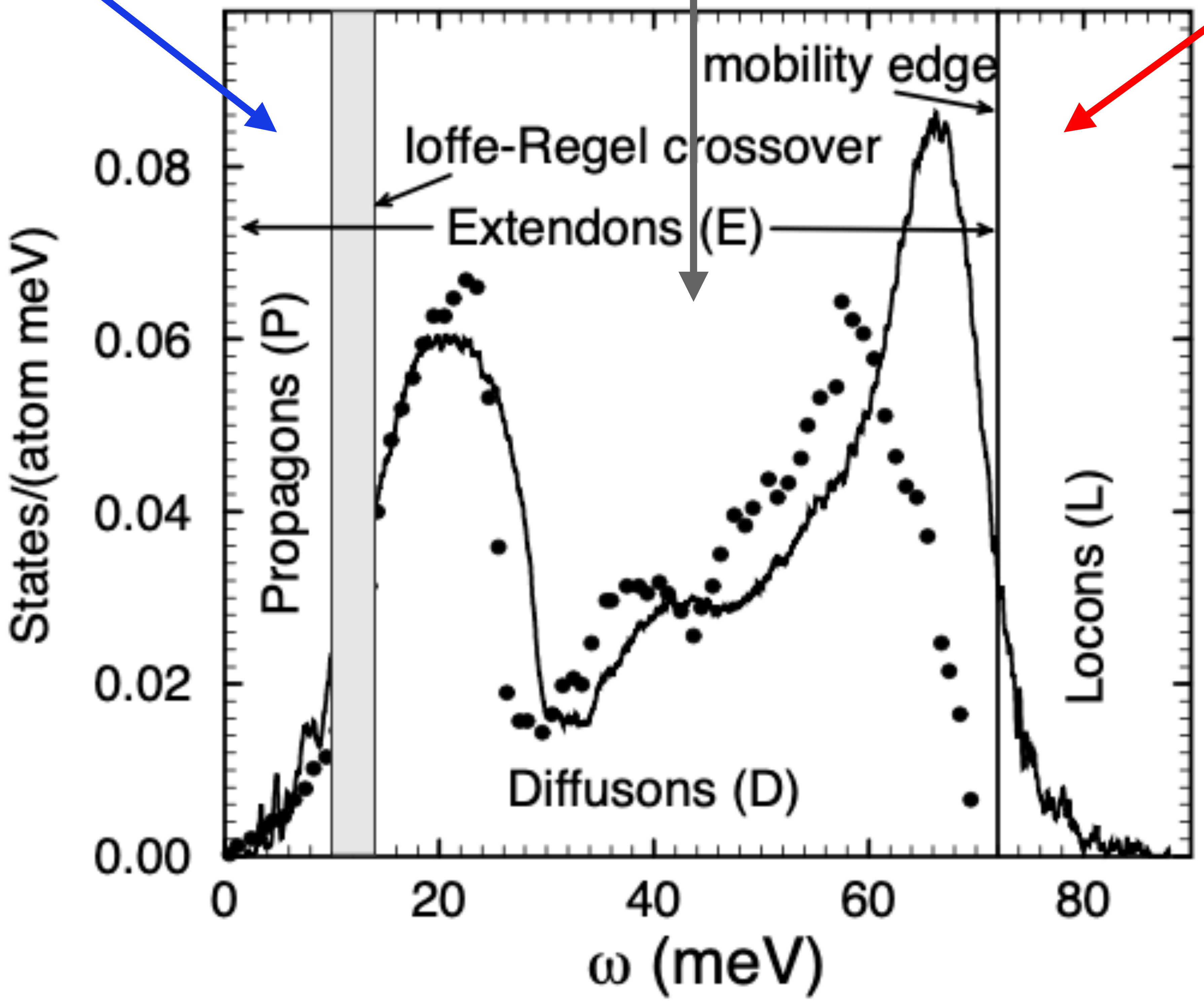






sound waves

localised vibrations



finite-size scaling

Dynamical Structure Factor

$$
\begin{aligned}
S(\boldsymbol{q},\omega) &= -\frac{1}{\pi}\mathrm{Im}G(\boldsymbol{q},\omega) \\
&= \frac{1}{\pi}\sum_n \frac{\gamma_n}{(\omega_n-\omega)^2+\gamma_n^2}|\langle e_n|\boldsymbol{q}\rangle|^2
\end{aligned}
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finite-size scaling

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$$= \frac{A(\boldsymbol{q})}{\pi}\frac{\Gamma(\boldsymbol{q})^2}{\left(\omega - \Omega(\boldsymbol{q})\right)^2 + \Gamma(\boldsymbol{q})^2}$$

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anharmonicity disorder

(blue sky)

finite-size scaling

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anharmonicity disorder

(blue sky)

Lanczos recursion

MA PER SEGUIR VIRTUTE E CANOSCENZA

finite-size scaling

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anharmonicity disorder

(blue sky)

Lanczos recursion

$$S(\boldsymbol{q},\omega) = -\frac{1}{\pi}\mathrm{Im}\left\langle \boldsymbol{q}\middle|\left((\omega+i\epsilon)^2-D\right)^{-1}\middle|\boldsymbol{q}\right\rangle$$
$$= -\frac{1}{\pi}\cfrac{1}{(\omega+i\epsilon)^2-a_0-\cfrac{b_1^2}{(\omega+i\epsilon)^2-a_1-\cfrac{b_2^2}{\ddots}}}$$

finite-size scaling

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anharmonicity disorder

(blue sky)

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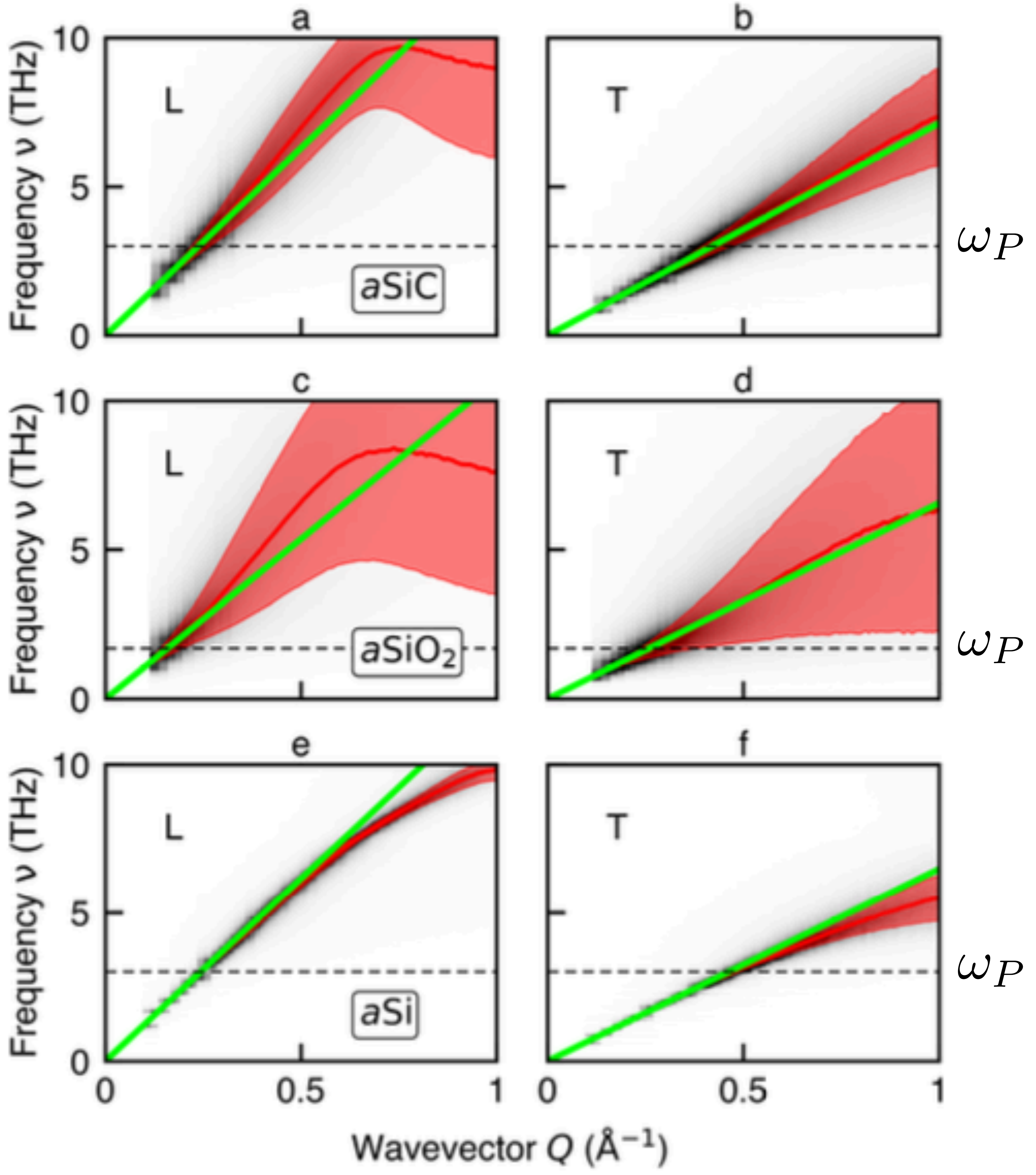
$$\begin{aligned}
|\xi_0\rangle &= |\boldsymbol{q}\rangle \\
b_0|\xi_{-1}\rangle &= 0 \\
a_n &= \langle\xi_n|D|\xi_n\rangle \\
a_{n+1}|\xi_{n+1}\rangle &= (D-a_n)|\xi_n\rangle - b_n|\xi_{n-1}\rangle \\
b_{n+1} &= \langle\xi_n|D|\xi_{n+1}\rangle
\end{aligned}$$

dozens of thousands of atoms easily manageable

finite-size scaling

Dynamical Structure Factor

$$S(\boldsymbol{q},\omega) = \frac{A(\boldsymbol{q})}{\pi} \frac{\Gamma(\boldsymbol{q})^2}{(\omega - \Omega(\boldsymbol{q}))^2 + \Gamma(\boldsymbol{q})^2}$$



finite-size scaling

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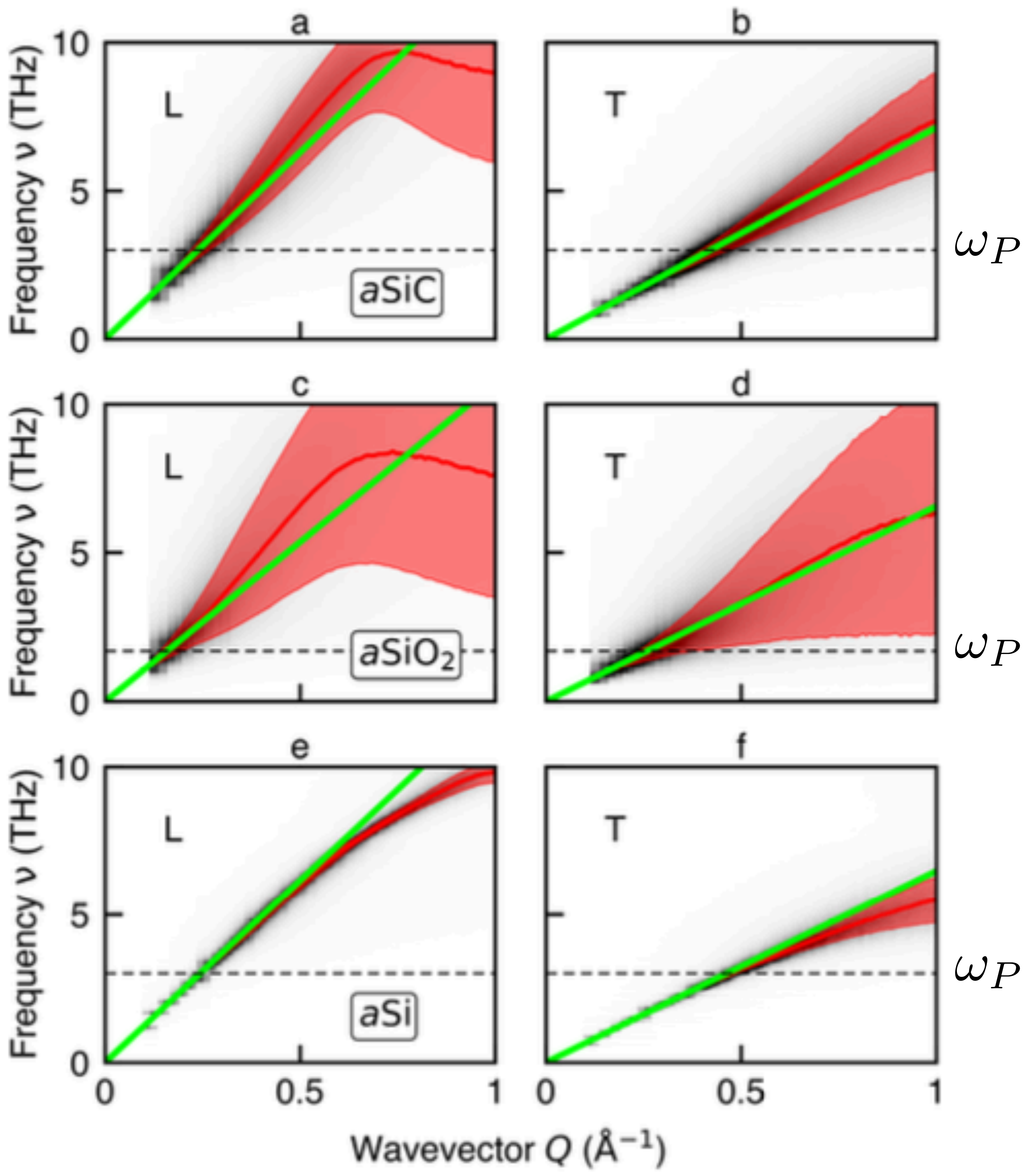
Heat conductivity

$$\kappa = \frac{1}{V}\sum_{nm\in P} c_{nm}\left(v_{nm}\right)^2\tau^{\circ}_{nm} + \frac{1}{V}\sum_{nm\notin P} c_{nm}\left(v_{nm}\right)^2\tau^{\circ}_{nm}$$



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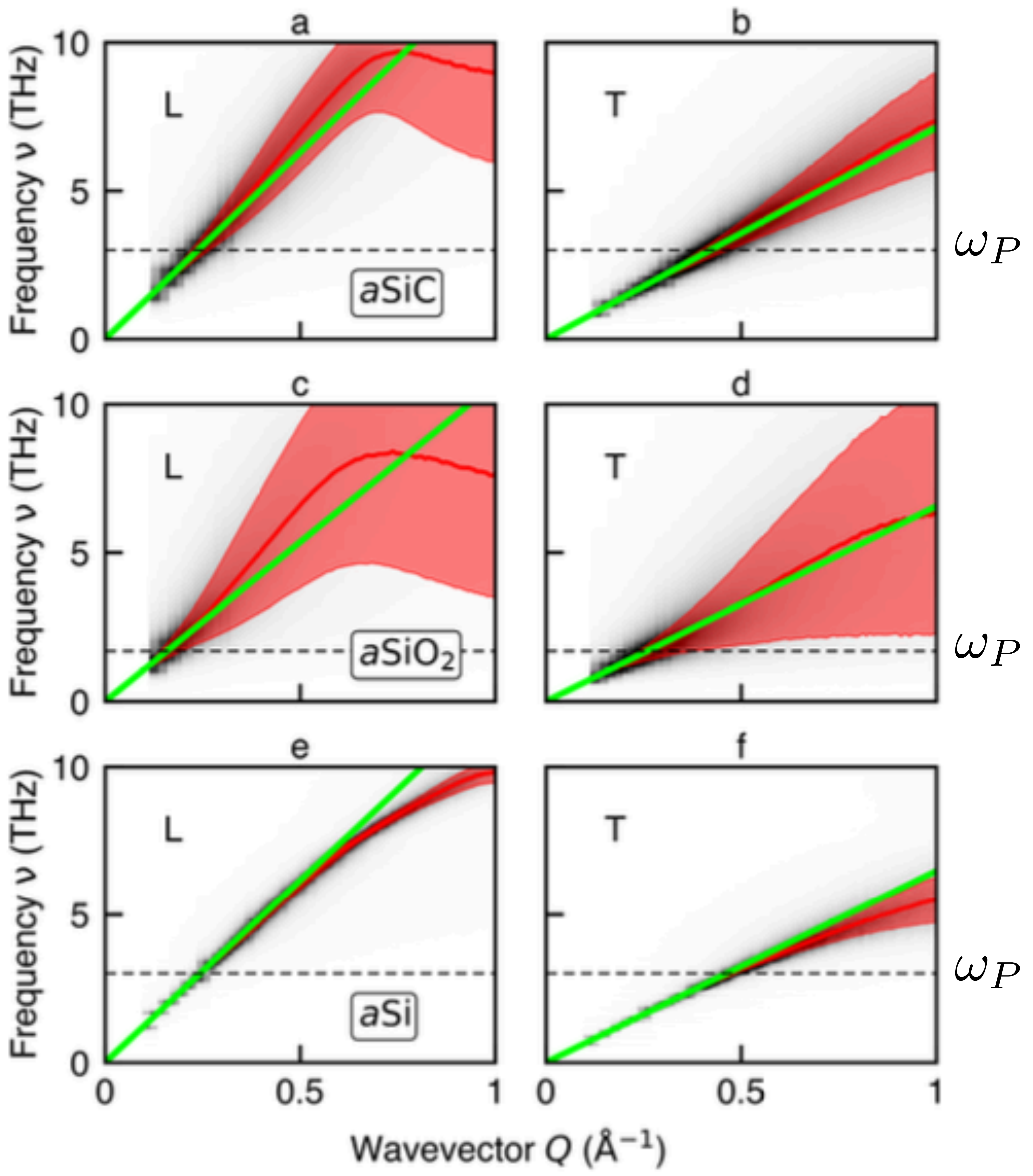
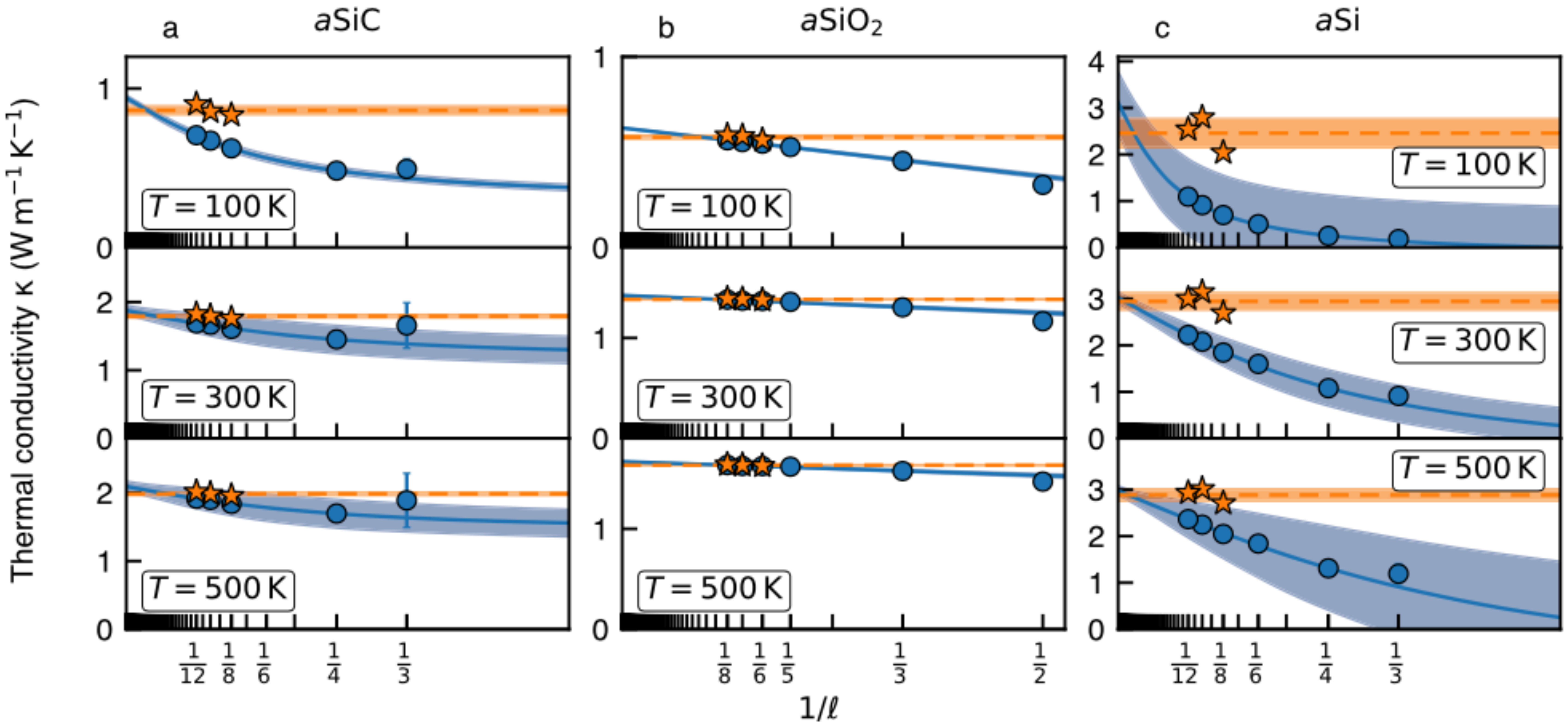
$$= \underbrace{\frac{1}{2\pi^2 c}\int_0^{\omega_P}\frac{\omega^2 C(\omega)}{2\Gamma(\omega)}d\omega}_{\kappa_P} + \underbrace{\frac{1}{V}\sum_{nm\notin P} c_{nm}\left(v_{nm}\right)^2\tau^{\circ}_{nm}}_{\kappa_{LD}}$$



finite-size scaling

hydrodynamic extrapolation

$$\kappa = \underbrace{\frac{1}{2\pi^2 c}\int_0^{\omega_P} \frac{\omega^2 C(\omega)}{2\Gamma(\omega)}\,d\omega}_{\kappa_P} + \underbrace{\frac{1}{V}\sum_{nm\notin P} c_{nm}\left(v_{nm}\right)^2 \tau^{\circ}_{nm}}_{\kappa_{LD}}$$



the $V \to \infty$ limit of the Allen-Feldman harmonic model

$$\kappa^{AF} = \frac{\pi}{V} \sum_{nm} C_n (v_{nm})^2 \delta(\omega_n - \omega_m)$$

the $V \to \infty$ limit of the Allen-Feldman harmonic model

$$
\begin{aligned}
\kappa^{AF} &= \frac{\pi}{V}\sum_{nm} C_n (v_{nm})^2 \delta(\omega_n - \omega_m) \\
&\approx \frac{1}{V}\sum_{nm} C_n (v_{nm})^2 \frac{\eta}{(\omega_n - \omega_m)^2 + \eta^2}
\end{aligned}
$$

the $V \to \infty$ limit of the Allen-Feldman harmonic model

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$$\kappa_P^{AF} = \frac{1}{2\pi^2 c}\int_{\omega_{min}}^{\omega_P} \frac{\omega^2 C(\omega)}{b\omega^4 + \eta} d\omega$$

$$\frac{2\pi c}{L}$$

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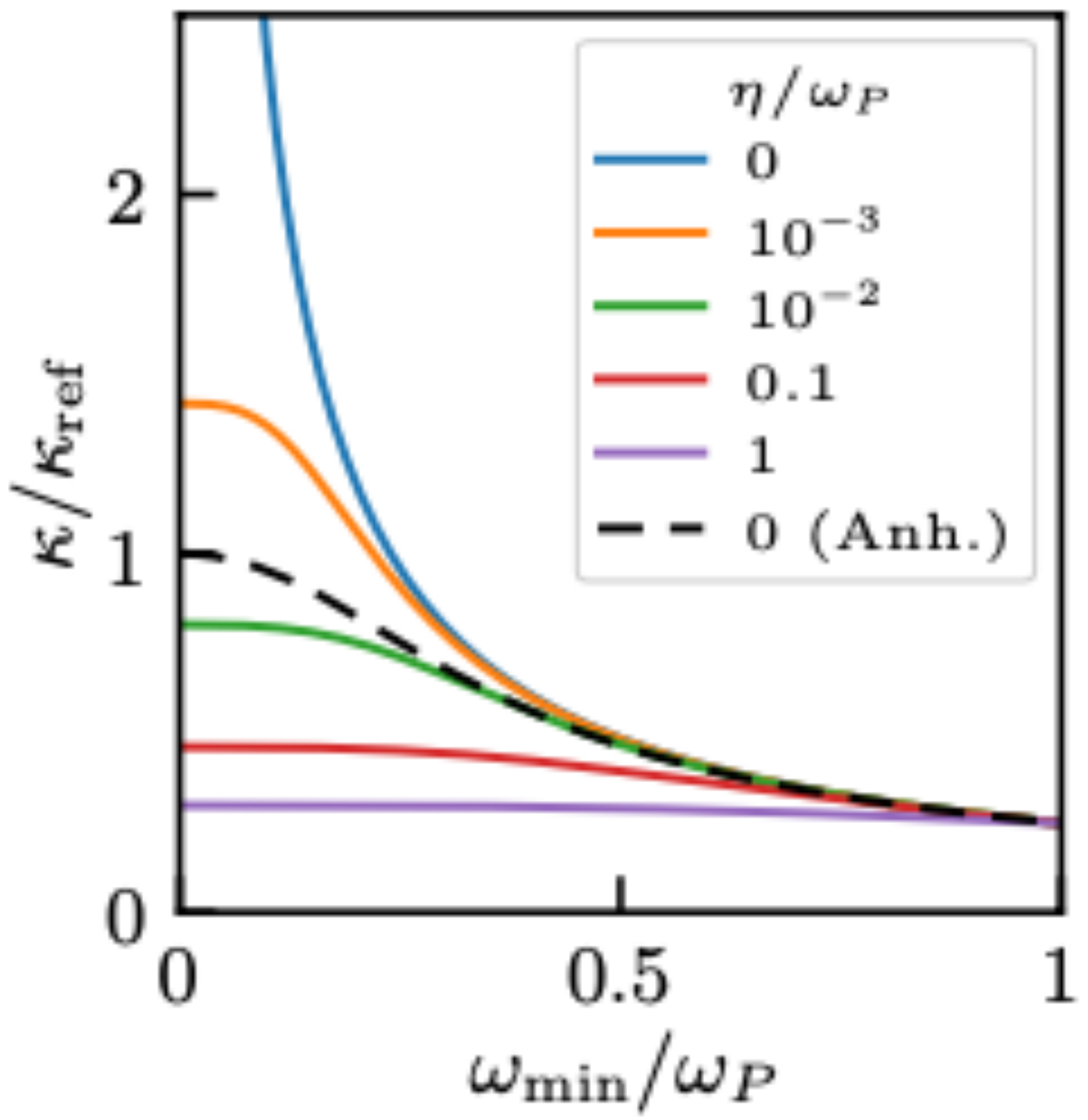

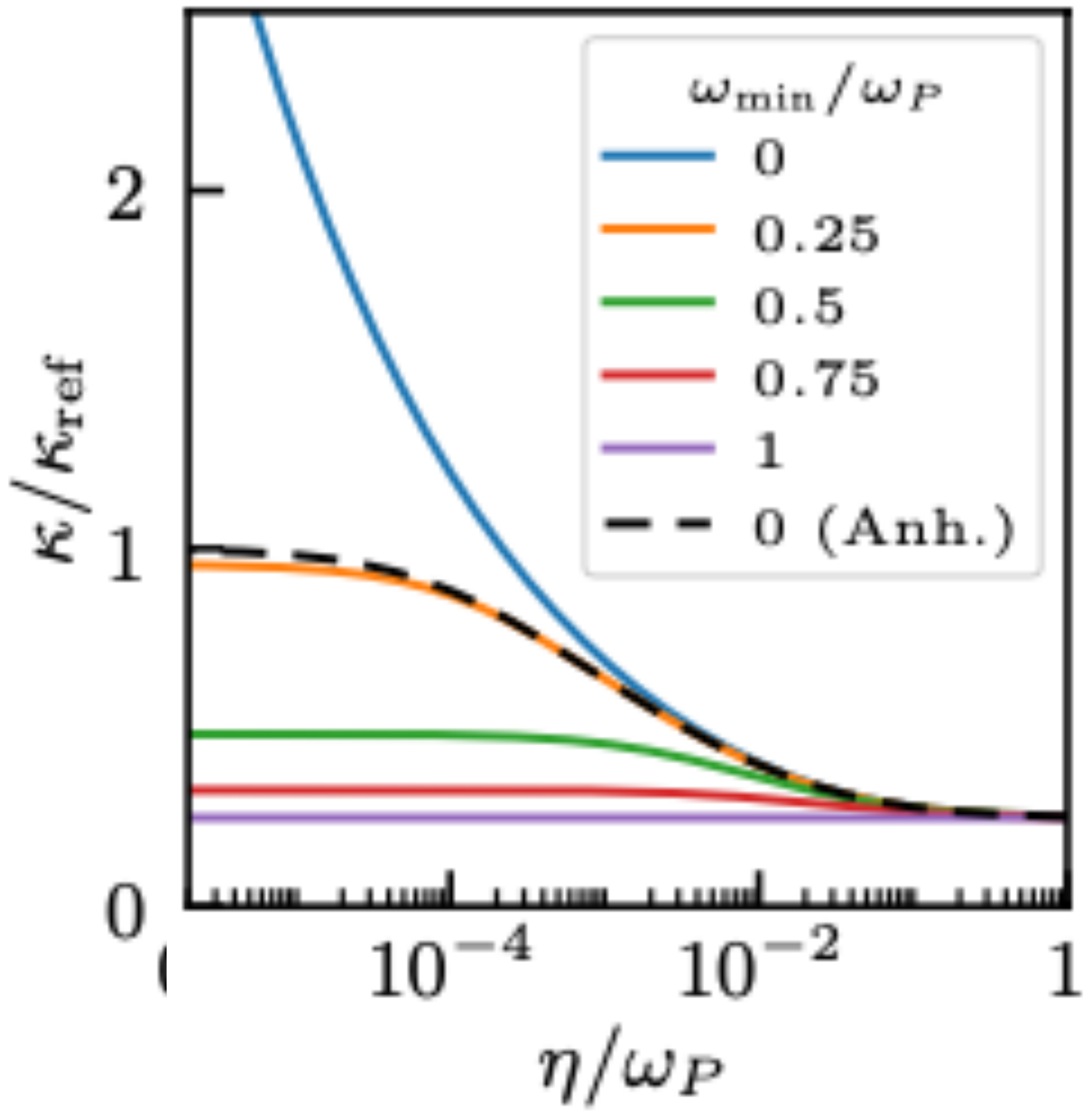

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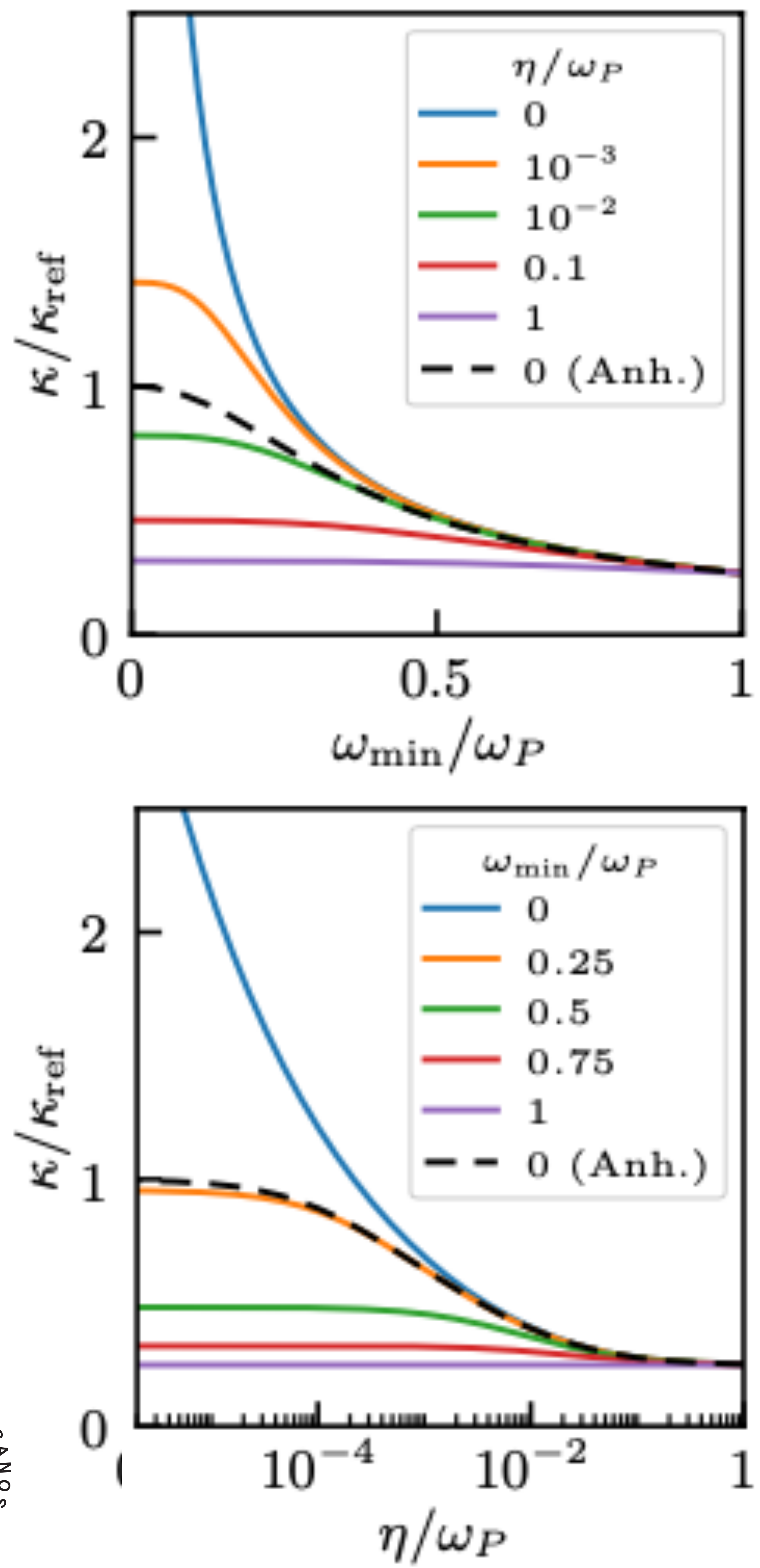
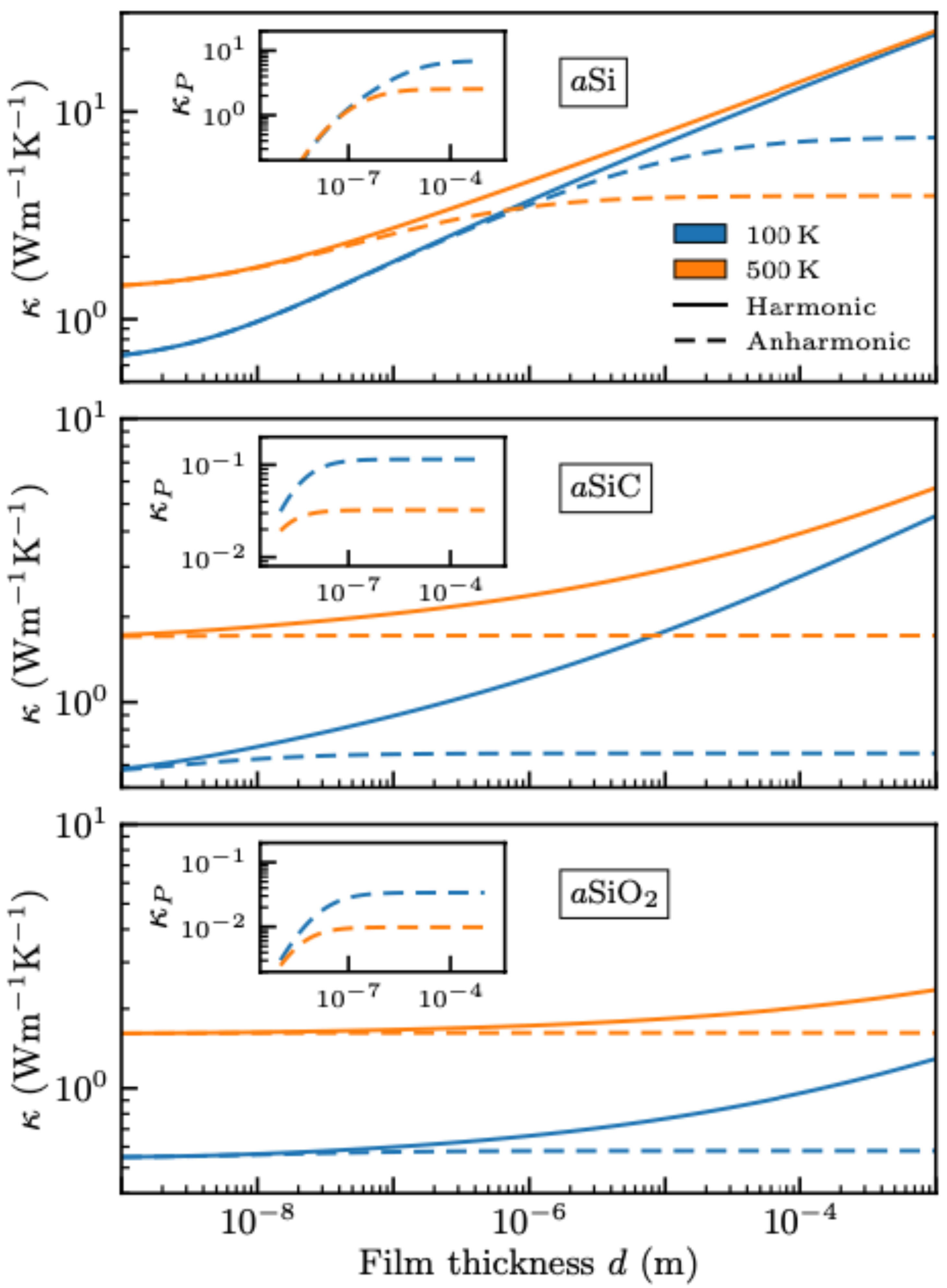
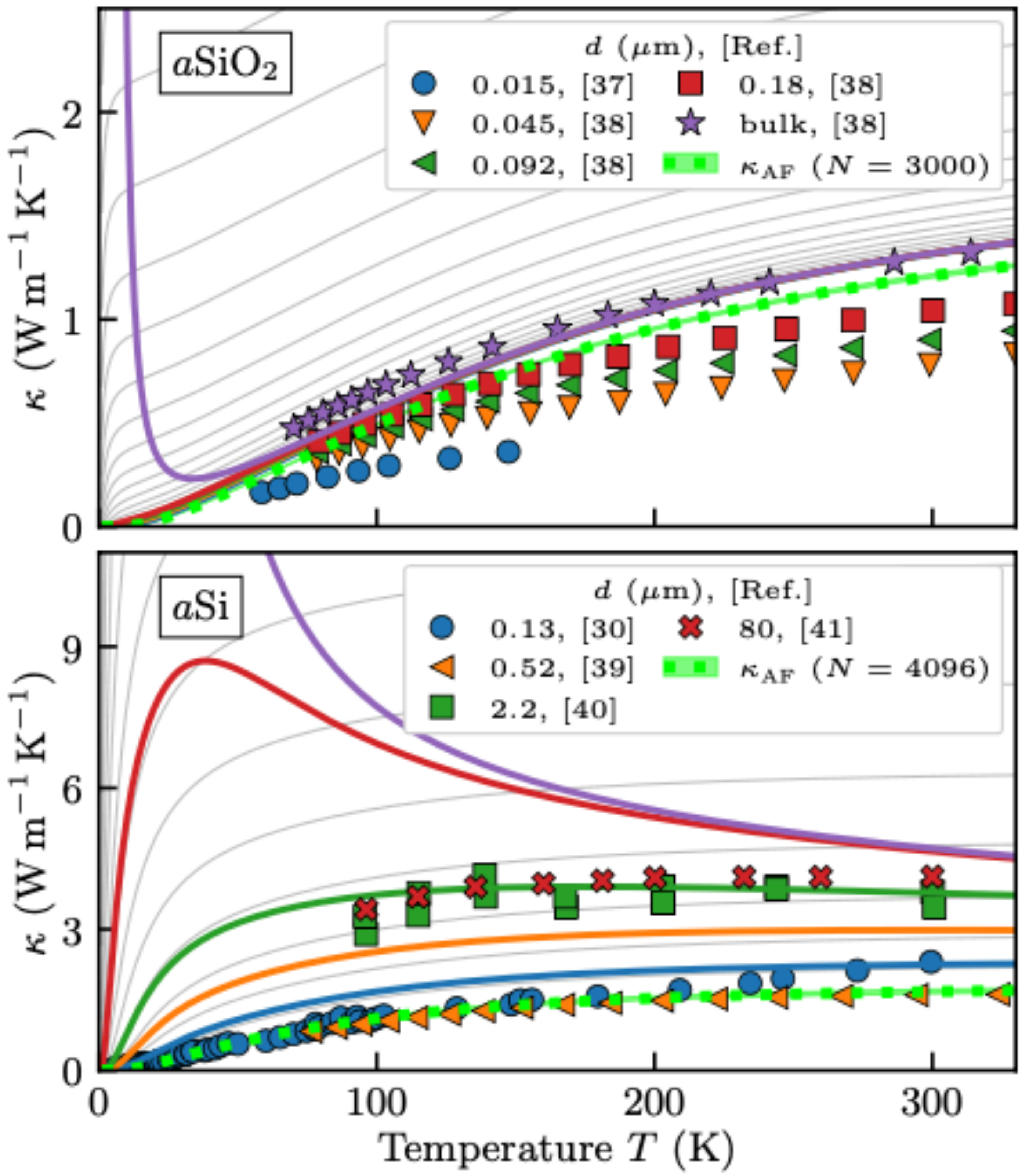
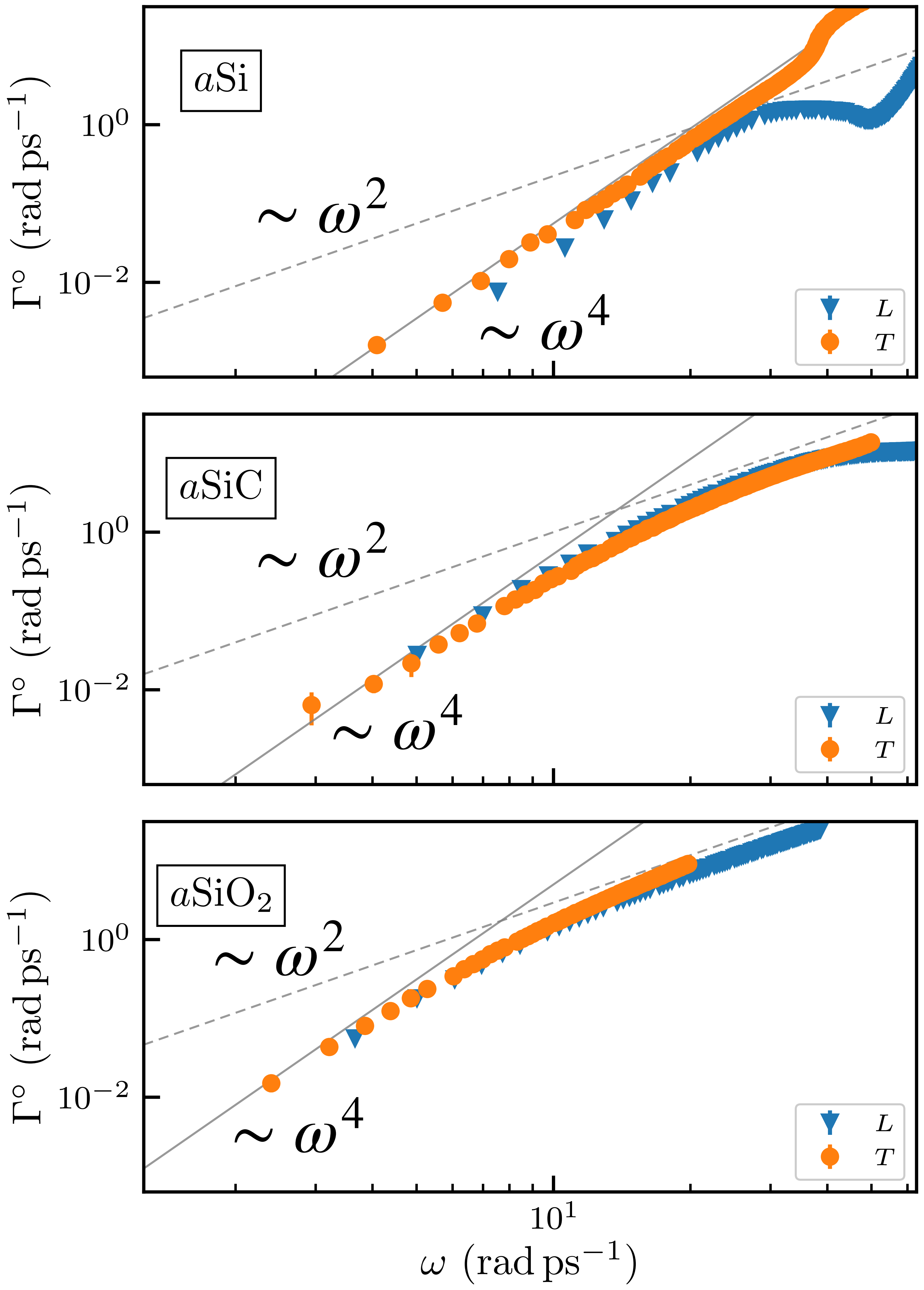






sound damping at low frequency

harmonic damping, $\Gamma(\omega)$



- an $\omega^4 \to \omega^2$ crossover is observed in the sound damping coefficient, as estimated from the harmonic DSF
- $\omega_{XO}[\mathrm{Si}] > \omega_{XO}[\mathrm{SiC}] > \omega_{XO}[\mathrm{SiO_2}]$
- experimentally, one observes two frequency crossovers, $\omega^2 \to \omega^4 \to \omega^2$:
  - the **first, temperature-dependent,** is due to the weakness of anharmonic effects at low temperature;
    - the **second, temperature-independent,** is possibly due to LT mixing determined by the line broadening itself

conclusions

- the combination of the Green-Kubo theory of linear response with the (quasi-) harmonic approximation for lattice vibrations allows us to bridge and naturally extend the Boltzmann-Peierls kinetic theory of heat transport in crystals with the Allen-Feldman harmonic model in glasses

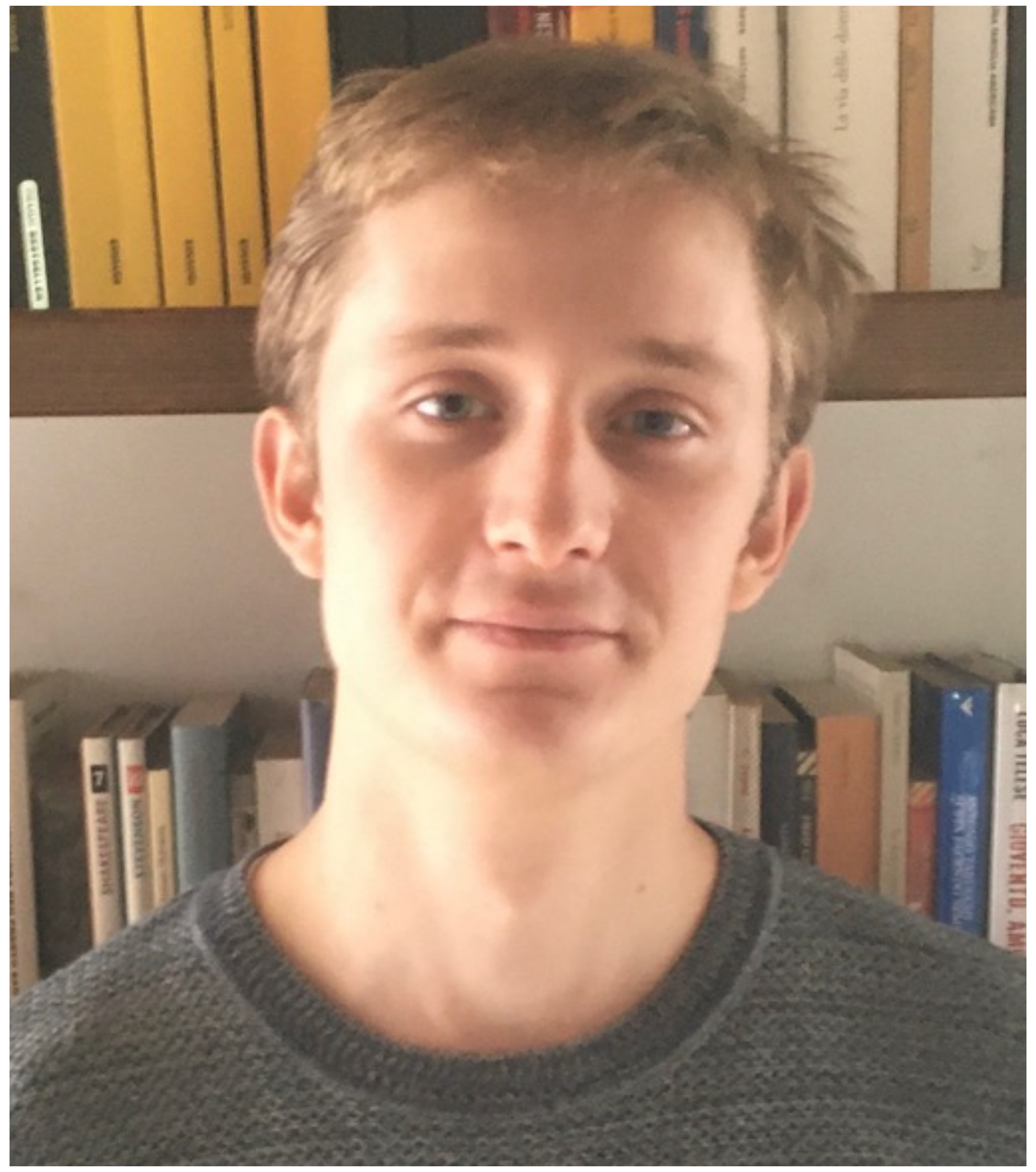
- results from computer simulations performed on finite glass models can be easily and accurately extrapolated to the thermodynamic limit, leveraging hydrodynamic arguments

- the Allen-Feldman harmonic model of heat transport in glasses is inevitably affected by an infrared singularity when the thermodynamic limit is performed properly; a finite conductivity can only result from finite-size finite effects, which can inadvertently mimic boundary scattering in experiments

- anharmonic effects are essential to predict a finite conductivity in bulk glasses

thanks to:

Paolo Pegolo
(EPFL)



Enrico Drigo
(SISSA)

Alfredo Fiorentino
(SISSA)

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Alfredo Fiorentino, Enrico Drigo, Stefano Baroni, Paolo Pegolo

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Centro Nazionale di Ricerca in HPC,
Big Data and Quantum Computing

QUANTUM ESPRESSO
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MAX


farewell, Nicola!


farewell, Nicola!
thanks to Nicola Marzari for sharing his photo archive with me