Minimising Thermal Conduction in Model Materials With Disorder on Different Length Scales

Abstract

Some of this draft will go in the final report, some of it into the literature review. I particularly like how the introduction goes

1 Introduction

It is a well known fact by now that one of the biggest crises facing humanity is the phenomenon of climate change. From researchers to diplomats, influential people around the world have called for urgent action to form a sustainable future [1], with many countries, such as the United Kingdom, setting net zero targets by 2050. [2] [3]

Net zero is achieved when emitted greenhouse gases to the atmosphere are exactly offset by removals. There are many ways of reaching this goal, including carbon capture and clean energy. Whilst the latter is still a rapidly growing sector with lots of potential, ever-growing energy demands push researchers to other techniques, such as improving energy efficiency; the energy losses must be minimised.

Energy is primarily dissipated as heat, with some additional losses in the form of sound waves. This is extremely common in metals, which are good thermal conductors. Conversely, thermal insulators, which are materials vital for the building sector, retain their heat; this makes them a key point of interest in minimising energy losses. Currently, thermal insulation techniques in buildings predominantly use fiberglass ($\kappa = 0.04 \mathrm{W\,m^{-1}\,K^{-1}}$) [4], which traps air within its glass fibers [5]. Foamy materials, which leverage air to maintain heat, are also used [6]. However, these materials are bulky, therefore their production is detrimental for net zero. A reduction of the thermal conductivity from the current ultra-low value of $\kappa \sim 10^{-1} \mathrm{W\,m^{-1}\,K^{-1}}$ to $\kappa \sim 10^{-3} \mathrm{W\,m^{-1}\,K^{-1}}$ would decrease the thermal insulation coatings in new buildings from 10 cm to 1mm. Such a material would be much more sustainable to produce, carrying significant implications for a net zero future.

The thermal conductivity κ is a constant relating the steady-state flow of heat J (also referred to as the thermal current density) to the temperature gradient ∇T, over a material. The relation follows Fourier's law [7], and is given in arbitrary dimension as

$$J = -\kappa \nabla T \tag{1}$$

Minimising the thermal conductivity of materials is an incredibly challenging task due to the underlying physics regarding Heisenberg's Uncertainty Principle. Additionally, modelling many-body interactions between carriers remains a computationally difficult endeavour.

2 Theory

2.1 Phonons

Atoms can never reach zero energy, even at absolute zero, due to Heisenberg's Uncertainty Principle setting a minimum bound on the position and momentum. Therefore, every particle will always have an associated oscillatory motion, no matter the conditions. It is for this reason that the thermal conductivity of a material can never be zero. In a lattice, due to the interatomic potential coupling, the atoms' motions are very highly connected; we say the lattice is vibrating. Such lattice vibrations are commonly referred to as "phonons", which are quasiparticles that can also transfer heat. Therefore, phonons are vital in understanding thermal properties of solids, such as thermal conductivity.

2.2 Anharmonic approximation and phonon scattering

To mathematically motivate thermal conductivity, we approximate the interatomic potential up to the anharmonic $(r-a)^3$ term. This term describes both thermal expansion and also momentary lattice contractions/expansions, which cause phonons to interact with each other. There are two types of phonon interactions: Normal (N), and Umklapp (U) scattering events.

(I am going to add a figure here, I would produce it by myself, but cause this is a literature review I think it would be better to reference a figure from an article)

In Normal scattering, the resulting wavevector lies inside the first Brillouin zone (1BZ), producing a phonon travelling in the same direction. In contrast, the resulting wavevector from Umklapp scattering lies outside the 1BZ, and is hence translated back to 1BZ due to the periodicity of the lattice. Consequently, the phonon now travels in the opposite direction; the flow of heat is now backwards. Therefore, Umklapp events describe thermal resistivity, whilst Normal events describe thermal conductivity.

2.3 Phonon gas model

The phonon gas model trivially treats phonons as gaseous particles, thus using the kinetic theory of gases to understand thermal transport. It is the simplest approach to describing the thermal conductivity of crystalline materials. This was first derived by Who? in Year? (I need to find this out)

By thermostatting two ends of a solid at different temperatures, we can assess the net rate of flow of energy across the temperature gradient due to phonon scattering events. Using this, we can firstly derive the thermal current density, and subsequently the thermal conductivity, which is found to be

$$\kappa = \frac{1}{3}\bar{v}l\tilde{C}_v \tag{2}$$

where $\bar{v}$ is the average phonon velocity (which would just be the speed of sound in this case), l is the mean free path and $\tilde{C}_v$ is the specific heat capacity of the material.

This approach is very robust for finding the thermal conductivity for a crystal on the surface level. However, it assumes a constant $\tilde{C}_v$, l and $\bar{v}$; additionally, it does not consider distinguishing between Normal and Umklapp scattering. Therefore, a more sophisticated description will be needed for a greater accuracy.

2.4 Einstein's model and Debye's improvements

Einstein set out to improve on (Who?'s) model of thermal conductivity in 1911 [8]. Einstein's key insight was to instead consider the solid as a collection of quantum harmonic oscillators, where the harmonic oscillators were between the first three nearest neighbours of atoms [9].

Although Einstein was correct to treat the problem as a set of harmonic oscillators, his assumption that all phonon modes shared the same frequency was incorrect. Debye improved on Einstein's models by assuming that the oscillation frequencies ω had a linear dependence on the phonon modes k, following the equation $\omega = v_s k$, where v_s is the speed of sound. In 1992, Cahill et al. modified Einstein's minimum thermal conductivity by using Debye's assumptions instead [9], to obtain the following relation for three dimensions

$$\kappa_{\min} = \left(\frac{\pi}{6}\right)^{1/3} k_B n^{1/3} \sum_i v_i \left(\frac{T}{\Theta_i}\right)^2 \int_0^{T/\Theta_i} \frac{x^3 e^x}{(e^x-1)^2}\, dx \tag{3}$$

where $k_B = 1.38 \times 10^{-23} \mathrm{J\,K^{-1}}$ is the Boltzmann constant, n is the number density, v_i is the acoustic phonon velocity, T is the temperature in Kelvin, and Θ_i is the Debye temperature.

Although this modified model is a vast improvement to the basic phonon gas model, it still does not account for the two types of phonon scatterings.

2.5 Peierls' model (rough notes)

In 1929, Peierls

$$\kappa = \frac{1}{3} \int C(\omega)\, \nu^2(\omega)\, \tau(\omega)\, d\omega \tag{4}$$

$$C(\omega) = \hbar\omega\, N(\omega)\, \frac{\partial f}{\partial T} \tag{5}$$

Although the Peierls model remains one of the most sophisticated models to find κ, it fails spectacularly to describe the thermal conductivity of disordered, or "amorphous" materials.

2.6 Allen-Feldman theory (rough notes)

Concerns disordered materials, which are known for exhibiting very low thermal conductivity

Locons, propagons, diffusons are the three types of coupling here

Propagons and diffusons I believe contribute to thermal conductivity, especially propagons. Fairly self explanatory given its name. Daniel said diffusons didn't contribute to thermal conductivity in his report but other literature says otherwise? Maybe I read it wrong

Locons in particular have zero thermal conductivity because they are 'localised' atoms. Is this called Anderson localisation? The localisation insulates because the phonons themselves are basically 'localised', they scatter off the boundaries of the local region so they're confined to that place. Meaning they can't propagate through. That would suggest to me that if we do want to minimise thermal conductivity we should look for methods which achieve many body localisation. I think to do that we need a critical energy, and also introduce vastly different masses (defects) like how Daniel did. Superlattices to introduce disorder across lengths. Perhaps try in lower dimensions and obtain a respectable result there

2.7 Attempts to unify (rough notes)

One of the papers Andrew sent covered this in detail, and it was really nice to see something which could unify all these theories. It was at a level far beyond my comprehension skills unfortunately, regarding condensed matter physics. Off diagonal elements in the velocity operator matrix represent the disordered terms (AF model), diagonals represent Peierls' model (crystals). They do start with the Born-Oppenheimer approx, Daniel and Andrew mentioned this

Disordered phonons behave like waves? Whilst crystal phonons behave like particles. Am I remembering that correctly?

3 Project Aims and Objectives

Using molecular dynamics simulations to model materials with disorder on different length scales, I think there's a Python dimension to this aswell with regards to the mesoscale

3.1 LAMMPS

3.2 Possible routes

4 Conclusion

References

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