Solid State Physics

THERMAL CONDUCTIVITY

Lecture 12

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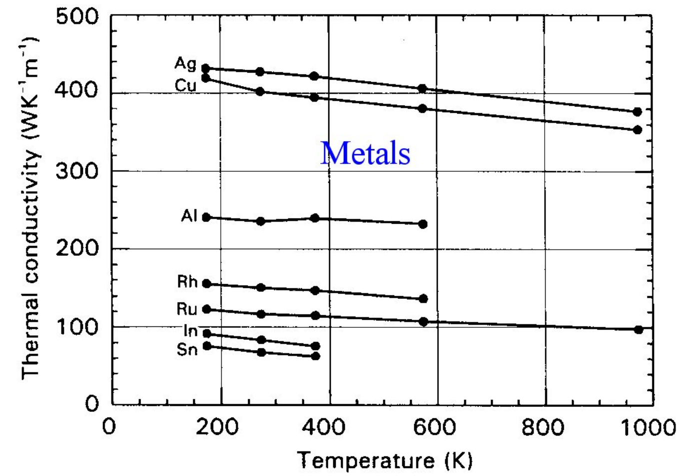
Thermal Conductivity

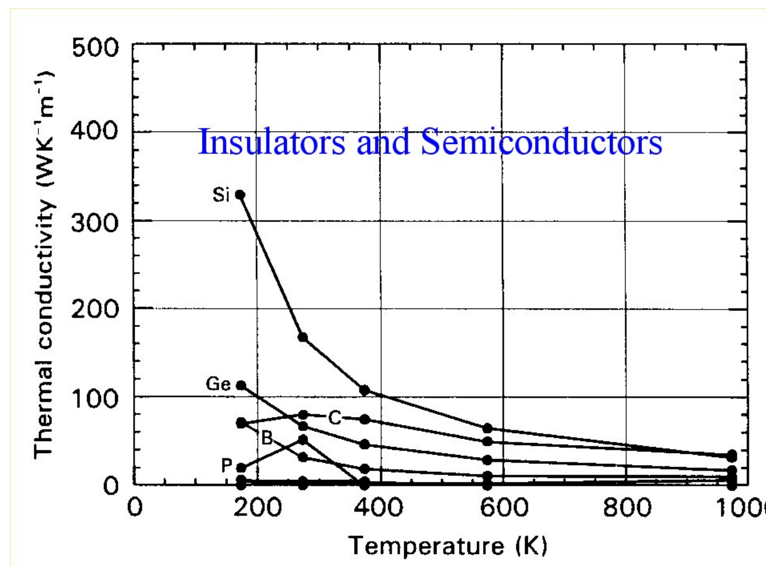
Previous Lecture

- Specific heats always tend to classical limit at high T.
- C_V decreases with decreasing T.
- Einstein model decreases too rapidly at low T
- Debye model gives correct T^3 dependence at low T.
- Debye temperature Θ_{D} as
  - correction factor to get right number of degrees of freedom;
  - fitting parameter.

4.7 Lattice Thermal Conductivity

4.8 Experimental values





- **Different behaviours of metals compared with insulators and semi-conductors;**
- **Very large range of values: for elements at room T**
  - **diamond: up to** $2600\ \mathrm{W\ K^{-1}m^{-1}}$
  - **copper:** $400\ \mathrm{W\ K^{-1}m^{-1}}$
  - **sulphur:** $0.3\ \mathrm{W\ K^{-1}m^{-1}}$

In the following sections we look at thermal conduction by lattice vibrations.

4.8.1 Phonons as particles

If mode k is in its n_kth excited state, as the energy levels are equally spaced, we can regard this as a state with n_k identical excitations in mode k, each with energy $\hbar\omega_k$. We say there are n_k phonons in mode k – exact analogy with photons. The phonon has energy $\hbar\omega_k$ and momentum $\hbar\mathrm{k}$. We can think of the phonon as a particle (*quasiparticle*).

4.8.2 Phonon momentum

The momentum of phonons is rather different to normal momentum. Conservation of momentum is a fundamental property of most systems: it is a result of the fact that the Hamiltonian of a free particle is invariant under translation (p commutes with $\mathcal{H}$). In a crystal, the Hamiltonian is only invariant under translation through a lattice vector R. As a result, momentum in the crystal in only conserved to within an additive constant $\hbar\mathrm{G}$, where G is a reciprocal lattice vector. $\hbar\mathrm{k}$ is not a true momentum of the whole crystal – except at $\mathrm{k} = 0$ when it corresponds to uniform motion of the whole crystal. $\hbar\mathrm{k}$ is called *quasimomentum*.

4.8.3 Phonon interactions

In the harmonic approximation we ignored terms in the Hamiltonian like

$$\sum_{nn'n''} u_n u_{n'} u_{n''} D_{nn'n''},$$

and got normal modes which did not interact. When we look for wave-like solutions, we have terms of the form

$$\sum_{kk'k''}\sum_n A_{kk'k''}\exp(i(\mathbf{k}+\mathbf{k}'+\mathbf{k}'').\mathbf{R}_n),$$

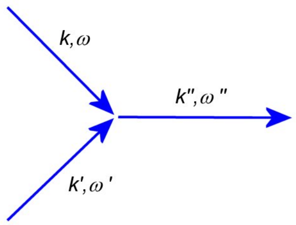
and, as in our discussion of diffraction, the sum will be zero because of phase cancellation unless

$$(\mathbf{k}+\mathbf{k}'+\mathbf{k}'').\mathbf{R}_n = 2m\pi$$

where m is an integer. But if G is a reciprocal lattice vector, $G.\mathrm{R}_n$ is a multiple of 2π, so all we can say is that

$$\mathbf{k}+\mathbf{k}'+\mathbf{k}''+\mathbf{G} = 0.$$

As a result of the anharmonic terms, we have phonon-phonon interactions. Physical explanation: a phonon alters the local atomic spacing, so that another phonon sees a difference in the crystal structure and is scattered by it.



4.8.4 Heat Transport

Treat phonons as a classical gas of particles, transporting energy $\hbar\omega$ at velocity v, the group velocity of the waves. Hot regions have a higher density of phonons than cool regions. Heat flux (energy/area/time) Q:

$$\mathbf{Q} = -\kappa \nabla T,$$

and κ depends on

- **number of particles/volume carrying energy n**
- **specific heat per carrier c_V**
- **carrier velocity v**
- **how far carrier travels before being scattered (mean free path Λ)**

From kinetic theory of gases

$$\kappa = \frac{1}{3} n v c_V \Lambda.$$

Note that nc_V is the specific heat *per volume* – contrast the specific heat *per mole* calculated earlier. Unless the phonons interact with something (are scattered) the thermal conductivity will be infinite.

4.8.5 Boundary scattering

Clearly Λ is limited by the size of the specimen. Generally, the specimen is polycrystalline – Λ is limited by the crystallite size.

4.8.6 Point defect scattering

Any irregularity in the crystal will scatter a wave. An impurity, or even a different isotope, creates an irregularity. The defect size is about that of an atom. But at low temperatures only low-energy, long-wavelength phonons are excited. Scatterer size $<<\ \lambda$ is the condition for Rayleigh scattering $\rightarrow\ \Lambda \propto \lambda^4$. Dominant phonons at temperature T have $k \propto T$, $\lambda \propto T^{-1}$, and at low T the number of phonons $\propto T^3$ suggesting $\kappa \propto T^3 \times T^{-4} = T^{-1}$. More exact treatment

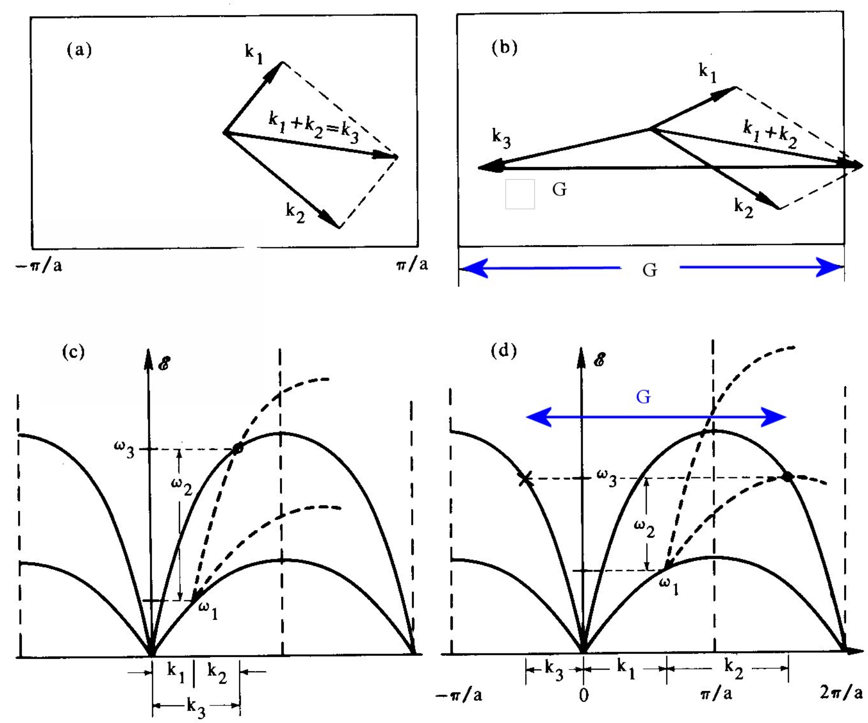
$$\kappa \propto T^{-\frac{3}{2}}.$$

4.8.7 Phonon-phonon scattering

At first glance, expect phonon scattering to preserve thermal current, as energy and momentum are both conserved:

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$$
$$\omega_1 + \omega_2 = \omega_3$$

so even if phonons interact, they continue to carry the energy in the same direction. But remember that the dispersion relation is periodic – this makes a difference.



If the two initial wavevectors add to a new wavevector which is outside the Brillouin zone, they give a new wave with a group velocity in the opposite direction. Usually, subtract G, a reciprocal lattice vector, to get back into the Brillouin zone:

$$\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{G} = \mathbf{k}_3.$$

Such a process is called an *Umklapp* process (German: flip-over) or *U-process*. Processes in which $\mathrm{G} = 0$ are called *N-processes*. Note that for a U-process at least one of the phonons must have $|\mathrm{k}| > \pi/(2a)$ – so very rare at low T. At low T, assume number of phonons with large enough $|\mathrm{k}|$ is $\propto \exp(-\theta/T)$, where θ is a temperature comparable with the Debye temperature. At high T, most of the phonons will have large enough $|\mathrm{k}|$ to give U-processes, and number of phonons $\propto T$.

4.8.8 Combined processes

Assume all the scattering processed are independent. Each process acts independently to reduce the conductivity. Analogous to resistances in series, so

$$\text{total resistance} = \sum_{\text{processes } i} \text{resistance}_i$$

or

$$\kappa = \frac{1}{\sum_i \frac{1}{\kappa_i}}.$$

Look at temperature dependence of terms in

$$\kappa = \frac{1}{3} n v c_V \Lambda;$$

note that v has negligible T dependence.

***High* T: can always have enough phonons for U-processes to dominate,**

- nc_V **independent of T (classical limit)**
- $\Lambda \propto T^{-1}$
- $\kappa \propto T^{-1}$

***Very low* T: U-processes are frozen out, and only have very long-λ phonons so defect scattering small. Boundary scattering dominates:**

- $nc_V \propto T^3$
- Λ **independent of T (geometry)**
- $\kappa \propto T^3$

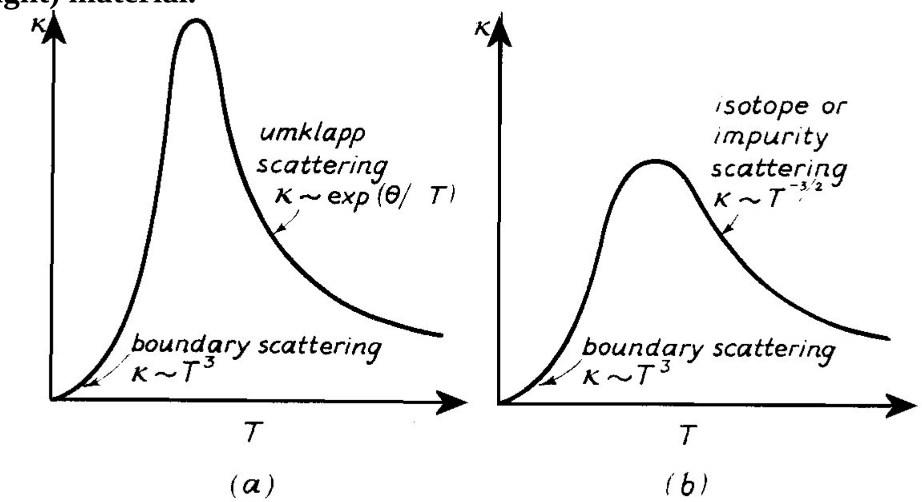
***Low-intermediate* T*, isotopically pure* U-processes dominate:**

- nc_V **only weakly dependent on T compared with**
- $\Lambda \propto \exp(\theta/T)$
- $\kappa \propto \exp(\theta/T$

***Low* T*, impure* defect scattering dominates:**

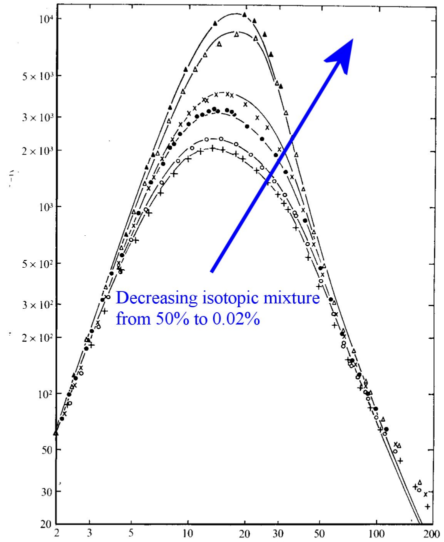
- $nc_V \propto T^3$
- $\Lambda \propto T^{-9/2}$
- $\kappa \propto T^{-3/2}$

Schematic variation of κ with T for isotopically pure (left) or impure (right) material.



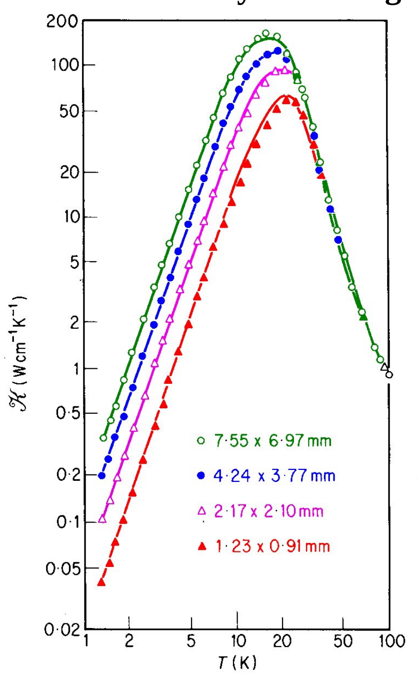
Note steeper rise to higher peak value for pure material.

Thermal conductivity of LiF as function of temperature for varying content of $^6\mathrm{Li}$ isotope.

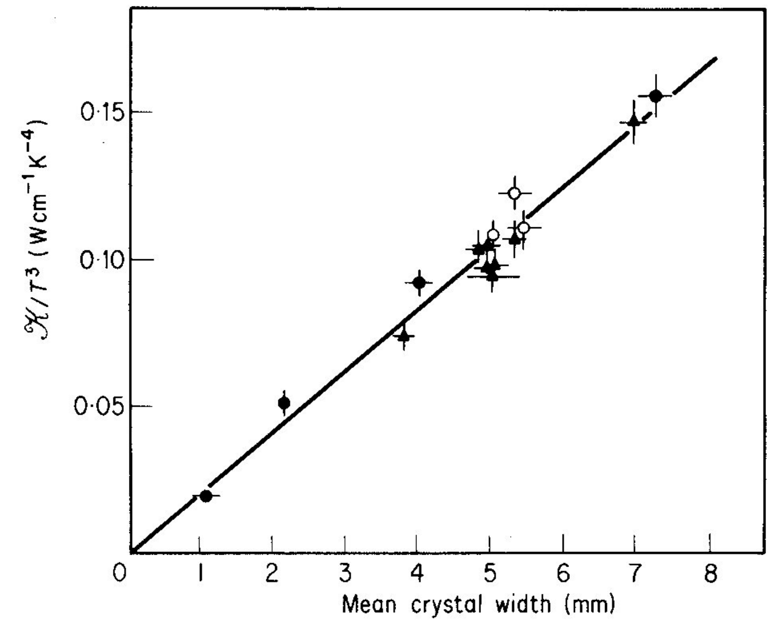


Defect content can be increased by irradiation (e.g. neutron damage in nuclear reactor).

Thermal conductivity of LiF as function of specimen size at low temperature, showing effect of boundary scattering.



Thermal conductivity of LiF plotted as κ/T^3 as function of temperature for low temperature.



Summary

- **Phonon scattering limits thermal conductivity**
- **scattering processes**
  - **phonon-phonon**
  - **phonon-defect**
  - **phonon-grain boundary**