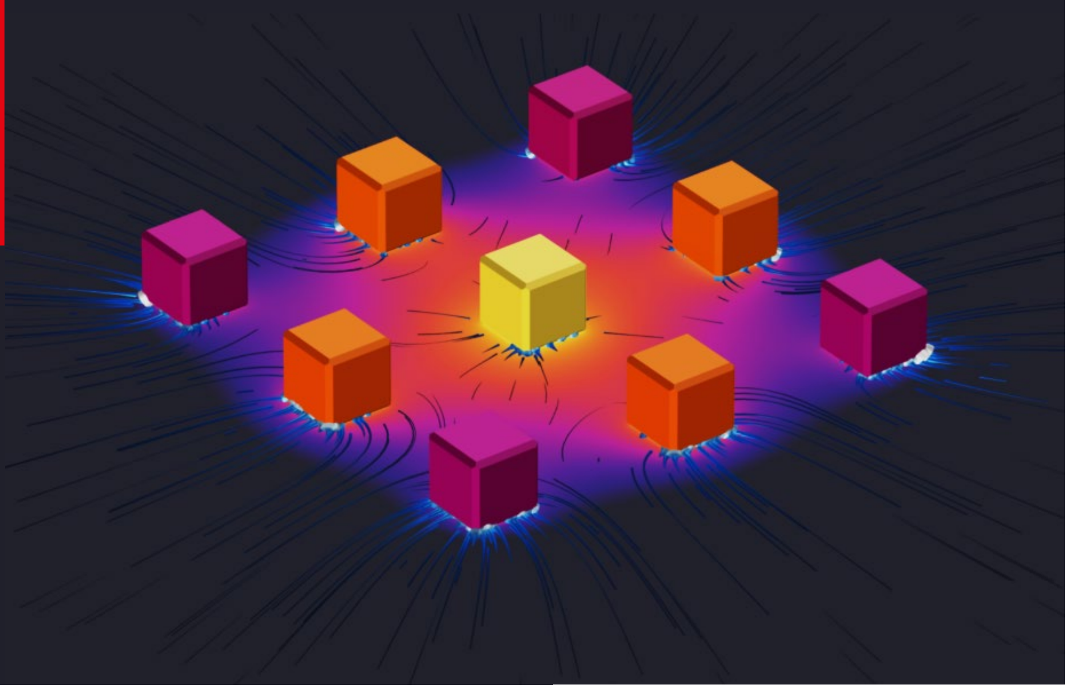
Nanoscale Heat Transfer (and Energy Conversion) ME469

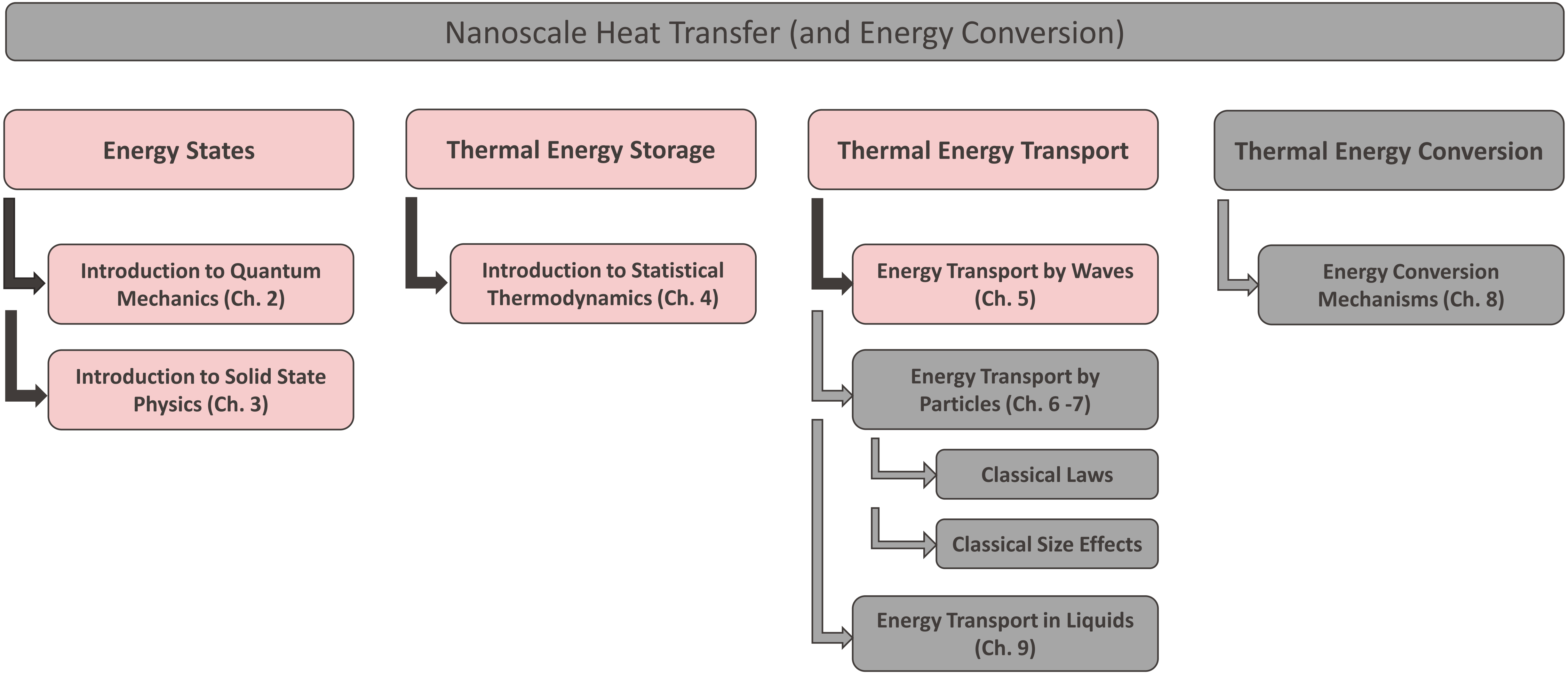
Instructor: Giulia Tagliabue



École polytechnique fédérale de Lausanne

Spring Semester

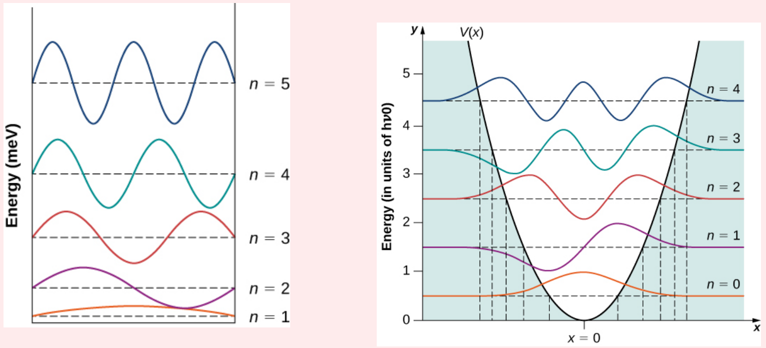
What we covered so far…



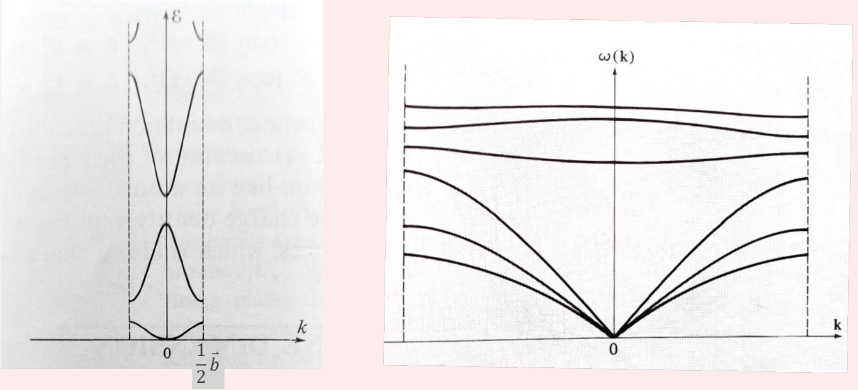
- Nanoscale Heat Transfer (and Energy Conversion)
  - Energy States
    - Introduction to Quantum Mechanics (Ch. 2)
    - Introduction to Solid State Physics (Ch. 3)
  - Thermal Energy Storage
    - Introduction to Statistical Thermodynamics (Ch. 4)
  - Thermal Energy Transport
    - Energy Transport by Waves (Ch. 5)
    - Energy Transport by Particles (Ch. 6 -7)
      - Classical Laws
      - Classical Size Effects
    - Energy Transport in Liquids (Ch. 9)
  - Thermal Energy Conversion
    - Energy Conversion Mechanisms (Ch. 8)

From Quantum States to Macroscopic Properties

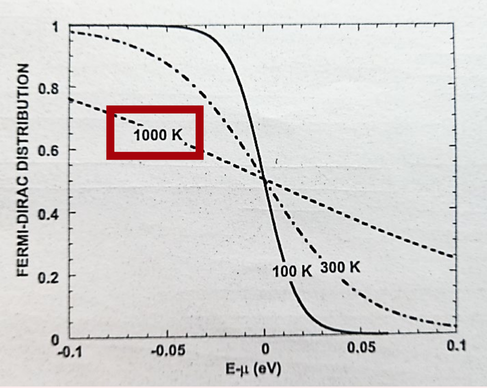
Localized potential



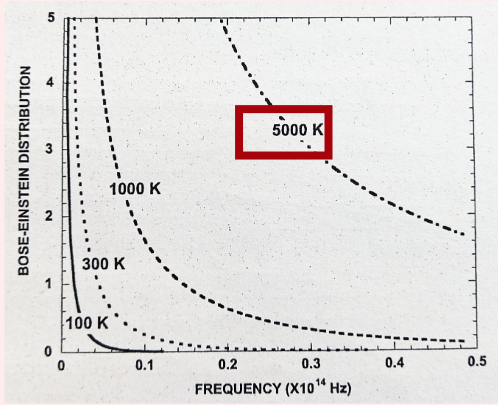
Periodic potential

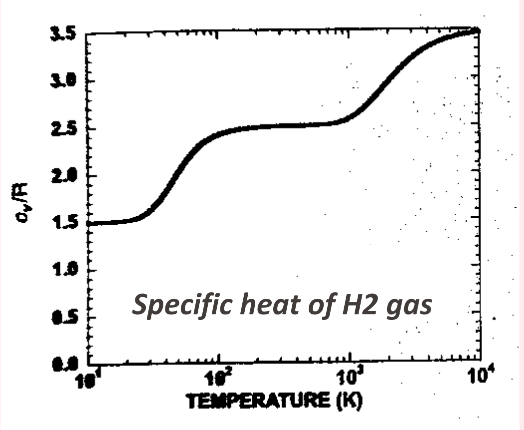


Fermi Dirac Distribution

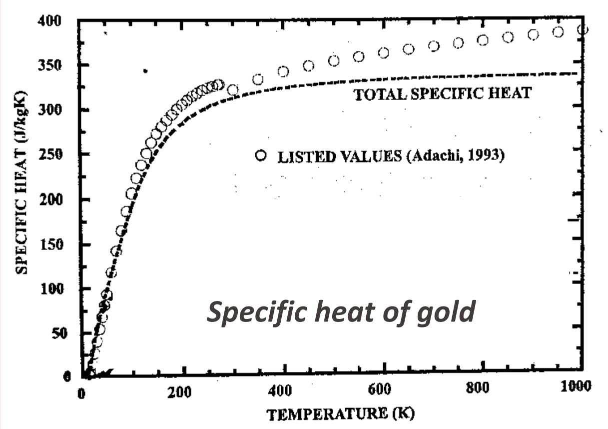


Bose Einstein Distribution





Specific heat of H2 gas



Specific heat of gold

Allowed quantum states (steady-state Schrodinger eqn)

Connect quantum states and energy levels with the temperature

Probability that matter will be in a given quantum state when it is at equilibrium. (Statistical Thermodynamics)

Investigate the properties of matter at finite temperatures

Macroscopic properties and their dependence on temperature

Internal Energy and Specific Heat

To calculate average quantities we recall that: $\langle X\rangle = \sum_{s=1}^{\Omega} X(s)P(s)$

Therefore, for the average internal energy we will have: $U = \sum_{i} E_i P(E_i)$ $\quad P(E_i) = e^{-\frac{E_i}{\kappa_B T}}/Z$ $\quad P(E_i, N_i) = e^{\frac{N_i\mu - E_i}{\kappa_B T}}/\mathcal{F}$

We recall that the constant-volume specific heat **per unit volume** is defined as: $C_V = \frac{1}{V}\left(\frac{\partial U}{\partial T}\right)_V$

Thus the constant-volume specific heat **per unit mass** is defined as: $c_V = \frac{C_V}{\rho}$ and the heat capacity is $\mathbb{C} = mc_V = \mathrm{V}C_V = \left(\frac{\partial U}{\partial T}\right)$

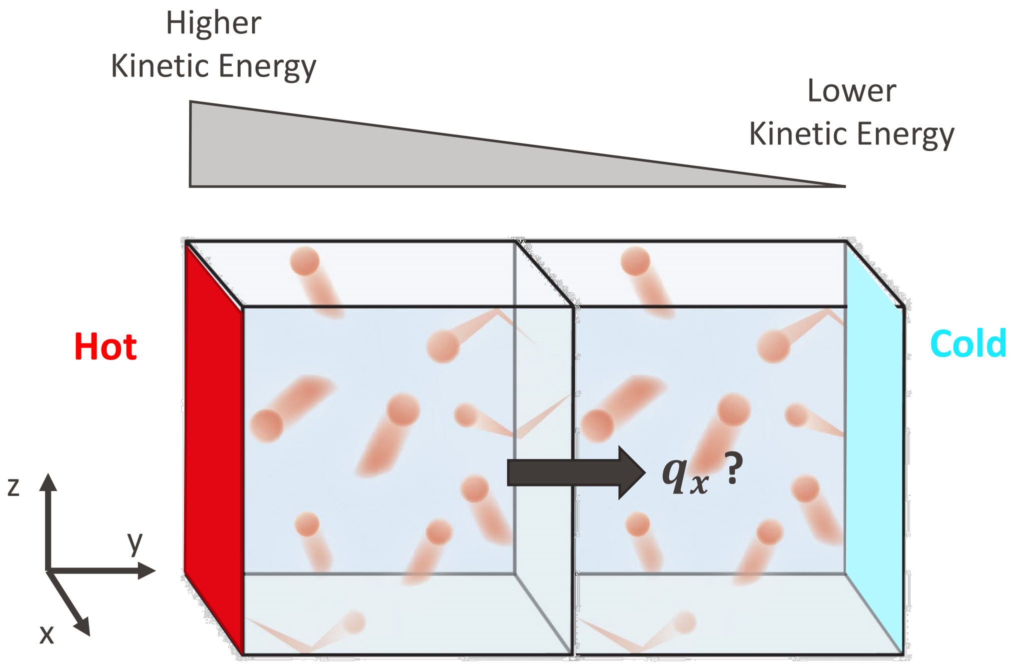
We will now consider different particles and follow the procedure:

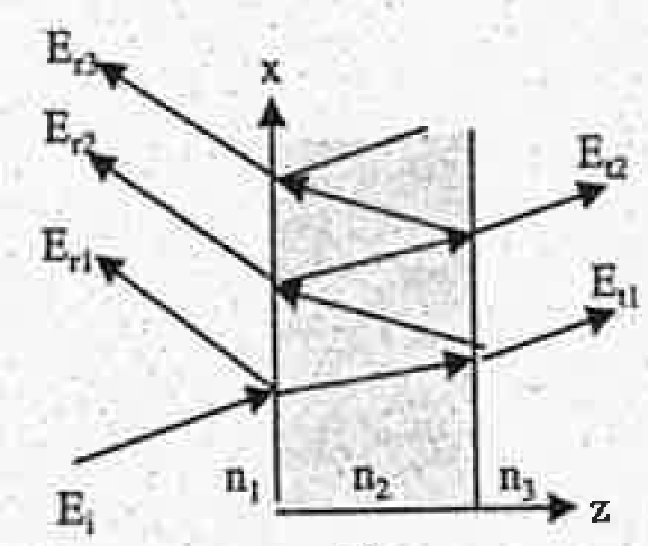
- Pick the statistical distribution function (fermionic/bosonic particles)
- Calculate the total internal energy
- Calculate C_V

Internal Energy and Specific Heat

Particle	**Internal Energy**	**Specific Heat**
Gases	$U = \sum_i E_i P(E_i) = \kappa_B T^2 \frac{\partial lnZ_N}{\partial T}$	$C_V = C_{V,t} + C_{V,r} + C_{V,v} + C_{V,e}$ $C_{V,t} = \frac{1}{V}\frac{3N\kappa_B}{2}$, $C_{V,r} = \frac{N\kappa_B}{V}$ $\mathit{for}\; T \gg \theta_r,$ $C_{V,v} = \frac{N\kappa_B}{V}\frac{\theta_v^2}{T^2}\frac{e^{\theta_v/T}}{\left(e^{\theta_v/T}-1\right)^2} \approx \frac{N\kappa_B}{V}$ for $T \gg \theta_v$, $C_{V,e} = 0$
Electrons	$U(T) = \int Ef(E,T,\mu)D(E)dE$	$C_{V,e}(T) = \frac{1}{2}\pi^2 n_e \kappa_B \frac{T}{T_f} \propto T$
Phonons	$U(\omega) = \int_0^{\omega_D} \hbar\omega f(\omega,T)\, D(\omega) d\omega$	$C_{p,D}(T) = \frac{36\pi^4\kappa_B}{15}\frac{N}{V}\left(\frac{T}{\theta_D}\right)^3 \propto T^3$ (low T) $C_{p,E}(T) = \frac{N_p\kappa_B N'}{V}\frac{\left(\frac{\hbar\omega_E}{\kappa_B T}\right)^2 exp\left(\frac{\hbar\omega_E}{\kappa_B T}\right)}{\left[exp\left(\frac{\hbar\omega_E}{\kappa_B T}\right)-1\right]^2} \sim const$ (high T)
Photons	$U_\omega = \hbar\omega f(\omega,T)\, D(\omega)$	$I_\omega = \frac{\hbar}{4\pi^3 c^2}\frac{\omega^3}{exp\left(\frac{\hbar\omega}{\kappa_B T}\right) - 1}$

Energy Transport





Particle View

Wave View

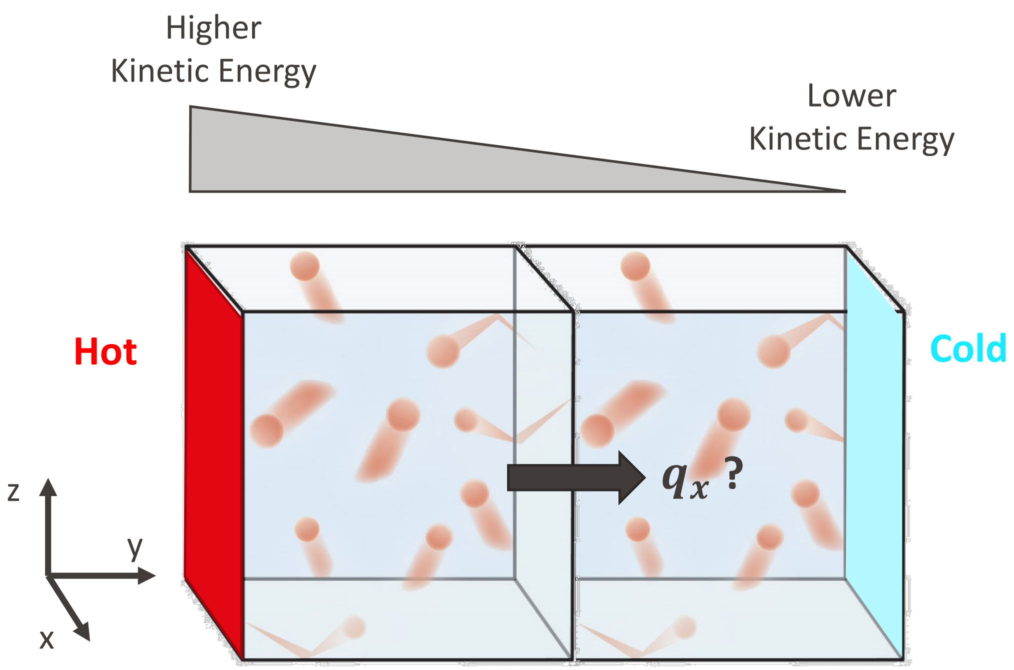
Energy and velocity of a carrier

Statistical distribution

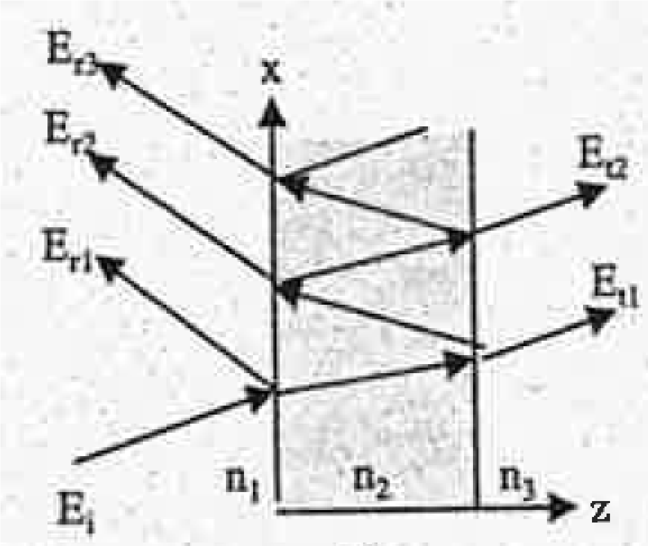
Reflectivity and Transmissivity

Interference phenomena

Energy Transport



Particle View



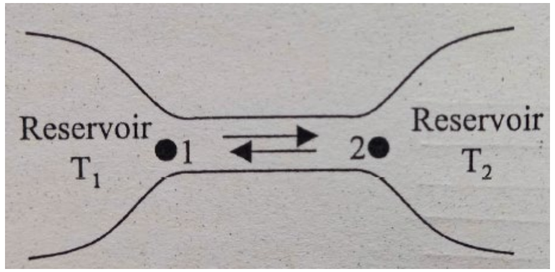
Wave View

How can we describe transport in a particle view accounting for the wave nature?
When and how does the wave-behavior affects energy transport?

In This Lecture …

- **Energy transfer in Nanostructures (Landauer formalism)**
  - Thermal boundary resistance and Universal quantum thermal conductance
  - Near field heat transfer and Superlattices
- From Waves to Particles
  - Wave packets and group velocity
  - Coherence lengths and heat transfer models

Energy Transfer in Nanostructures – Landauer Formalism



In the *Landauer formalism* the transport is seen as a transmission process.

In other words, if we know the energy and velocity distribution as well as transmissivity of a given particle, we can calculate the particle flux and from this the energy transport.

Let's consider for example the case of heat transfer between two reservoirs. We know that heat is carried primarily by phonons, therefore we will write:

To get energy density

Energy of one carrier

Statistical distribution

$$q_{1\to2} = \sum_{p}\left[\frac{1}{V}\overbrace{\sum_{k_{x1}=-k_{max}}^{k_{max}}\ \sum_{k_{y1}=-k_{max}}^{k_{max}}\ \sum_{k_{z1}=0}^{k_{max}}}^{\text{Sum over all wavevectors}} v_{z1}\tau_{12}\underbrace{Ef(E,T_1)}_{\text{Average energy per quantum state}}\right] = \sum_{p=1}^{3m}\left[\frac{1}{V}\sum_{k_{x1}=-k_{max}}^{k_{max}}\ \sum_{k_{y1}=-k_{max}}^{k_{max}}\ \sum_{k_{z1}=0}^{k_{max}} v_{z1}\hbar\omega\tau_{12}f(\omega,T_1)\right]$$

Sum over all polarization

Propagation in the positive z-direction

velocity

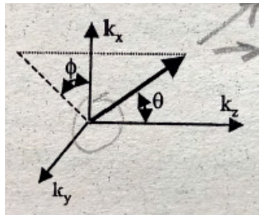
transmissivity

where

$$k_{max} = \frac{\pi}{a}$$

m=atoms per basis

$$v_{z1} = v_1 cos\theta$$



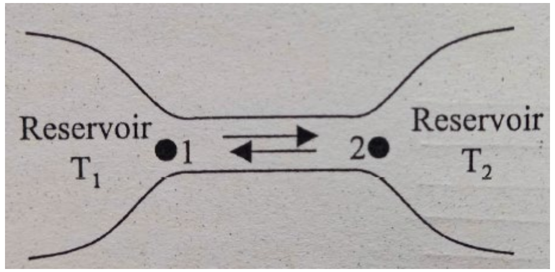
Debye frequency

Solid angle

Using the density of states we can turn the summation in an integral: $$q_{1\to2} = \iint_{\Omega_1>2\pi}\left[\int_0^{\omega_{D1}} v_{z1}\hbar\omega\tau_{12}f(\omega,T_1)\frac{D_1(\omega)}{4\pi}d\omega\right]d\Omega$$

All solid angles for positive z-direction

Energy Transfer in Nanostructures – Landauer Formalism



We note that there will be also a flux from reservoir 2 to reservoir 1. The NET heat flux between the reservoirs is:

$$q = q_{1\to 2} - q_{2\to 1} = \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_{z1} \hbar\omega \tau_{12} f(\omega, T_1) \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega - \iint_{\Omega_2 < 2\pi} \left[\int_0^{\omega_{D1}} v_{z2} \hbar\omega \tau_{21} f(\omega, T_2) \frac{D_2(\omega)}{4\pi} d\omega \right] d\Omega$$

At thermal equilibrium $T_1 = T_2$:

$$0 = \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_{z1} \hbar\omega \tau_{12} f(\omega, T_1) \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega - \iint_{\Omega_2 < 2\pi} \left[\int_0^{\omega_{D1}} v_{z2} \hbar\omega \tau_{21} f(\omega, T_1) \frac{D_2(\omega)}{4\pi} d\omega \right] d\Omega$$

This is called a detailed balance and enables us to express the flux only based on the properties of one reservoir plus the transmissivity:

$$q = q_{1\to 2} - q_{2\to 1} = \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_{z1} \hbar\omega \tau_{12} \big(f(\omega, T_1) - f(\omega, T_2) \big) \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega$$

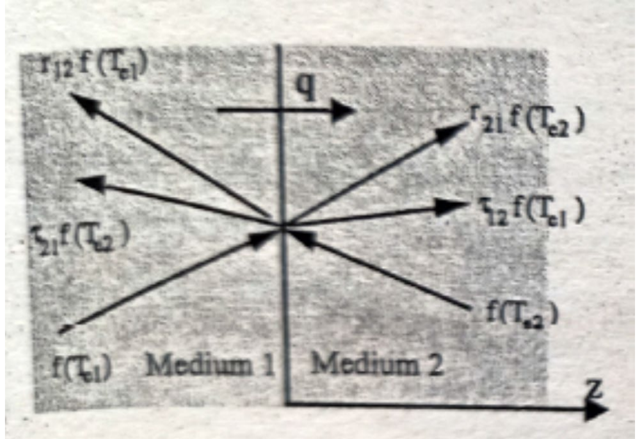
For a small difference in temperature:

$$q = q_{1\to 2} - q_{2\to 1} = (T_1 - T_2) \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_{z1} \hbar\omega \tau_{12} \frac{\partial f}{\partial T} \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega = K\Delta T$$

where $K = thermal\ conductance\ [W m^{-1} K^{-1}]$

We note that the critical quantity to be calculate is the transmissivity τ_{12}. When the particles travel without scattering, i.e. collisions, which is also called ballistic transport, the transmissivity can be calculated based on the previous discussion of waves transport across interfaces. However, in most cases, scattering renders the calculation very complex and the Landauer formalism less useful. Yet this approach proves most useful for the study of transport in nanostructures.
Let’s now see a few cases.

Thermal Boundary Resistance



Let’s consider the heat flux at the interface between two materials at respective temperatures $T_1 > T_2$.
Phonons travelling towards the interface from both sides will be transmitted and reflected.
If we take an isotropic material with only one atom per basis, based on our previous analysis we can write:

$$q = q_{1\to 2} - q_{2\to 1} = (T_{e1} - T_{e2}) \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_1 cos\theta_1 \hbar\omega\tau_{12} \frac{\partial f}{\partial T} \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega = \frac{T_{e1} - T_{e2}}{R_e}$$

➡ $R_e = thermal\ boundary\ resistance$

T_e are the phonon temperatures. However, we are in a **highly non-equilibrium situation** and it is hard to correctly define a temperature.

Indeed, at the interface we have three distinct groups of phonons:

- Incoming phonons with temperature T_{e1}
- Reflected phonons with energy distribution depending on the convolution of the incoming phonons and the interface reflectivity
- Transmitted phonons with an energy distribution depending on the convolution of the phonons arriving from the medium 2 and the interface transmittivity

It is then clear that, even for an ideal interface, there will be a thermal boundary resistance as long as reflection occurs at such interface.
For nearly ideal interfaces, the order of magnitude of R_e is:

$$R_e \sim \frac{1}{Cv}$$

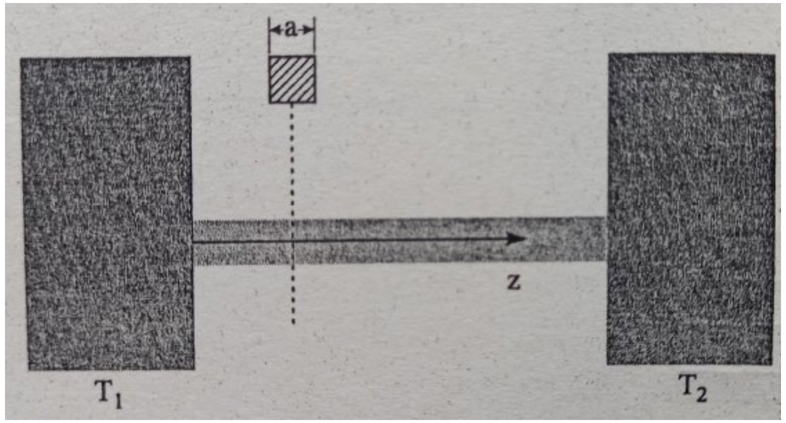
$C = volumetric\ specfic\ heat \approx 10^6\ J\ m^{-3} K^{-1}\ @300\ K$

$v = phonon\ speed \approx 10^3\ m\ s^{-1}\ @300\ K$

➡ $R_e \sim 10^{-8} - 10^{-9} K m^2 W^{-1}$

This result is consistent with experimental data. Non-ideal interfaces will have larger values.
Although this value is very small for a single interface, for nanoscale system with a large number of interfaces this resistance becomes dominant. Importantly, multilayer systems can exhibit distinct behaviors due to interference effects.
In macro-scale systems, this value can also be large due to the absence of perfect contact.

Universal Quantum Thermal Conductance



Let’s now consider a nanowire with square cross section that connects two thermal reservoirs.
Let’s neglect internal scattering and assume that the phonon transmissivity is unity for each mode.
We now want to calculate the thermal conductance of the nanowire:

$$K = \frac{q_{12}}{T_1 - T_2} \quad \text{where} \quad q_{12} = \sum_{p=1}^{3m}\left[\frac{1}{V}\sum_{k_{x1}=-k_{max}}^{k_{max}}\ \sum_{k_{y1}=-k_{max}}^{k_{max}}\ \sum_{k_{z1}=0}^{k_{max}} v_{z1}\hbar\omega\left(f(\omega,T_1) - f(\omega,T_2)\right)\right]$$

We thus need to compute the allowable wavevectors in this system. In the xy plane the wire acts as a 2D potential well and therefore the allowable modes are:

$$k_x = 2\pi\frac{m}{2a} = \frac{\pi m}{a},\ k_y = \frac{n\pi}{a}\ (m, n = \pm 1, \pm 2, \ldots)$$

$$\omega = ck = c\sqrt{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{a}\right)^2 + k_z^2}$$

$$v_{z1} = \frac{d\omega}{dk_z} = group\ velocity$$

➡ At low T and for the first few modes (small m,n)

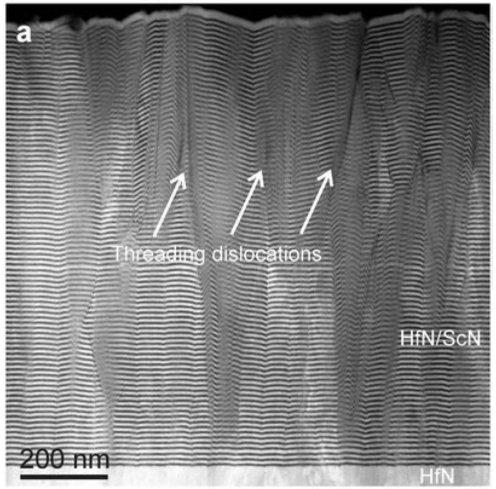
$$K_1 = \frac{\pi^2 \kappa_B^2 T}{3h}$$

Quantum thermal conductance of each modes

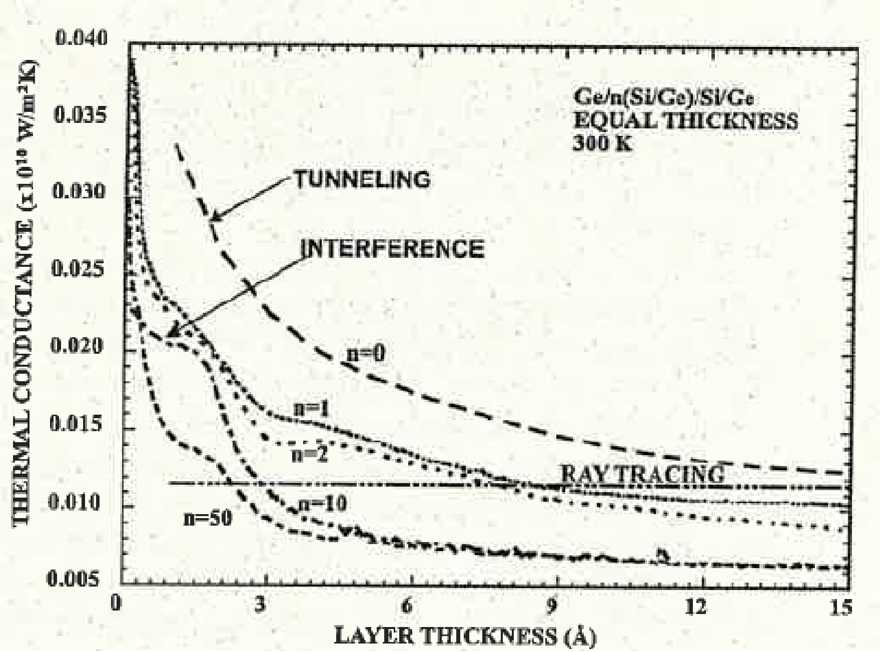
We observe that this conductance does NOT depend on the material properties. Thus it is the same for all the materials.
This quantum thermal conductance has been observed experimentally [1], although recently deviations have been recently reported [2]

[1] https://www.sciencedirect.com/science/article/abs/pii/S0749603697905619; [2] https://www.nature.com/articles/s41467-018-06791-0

Super-lattices



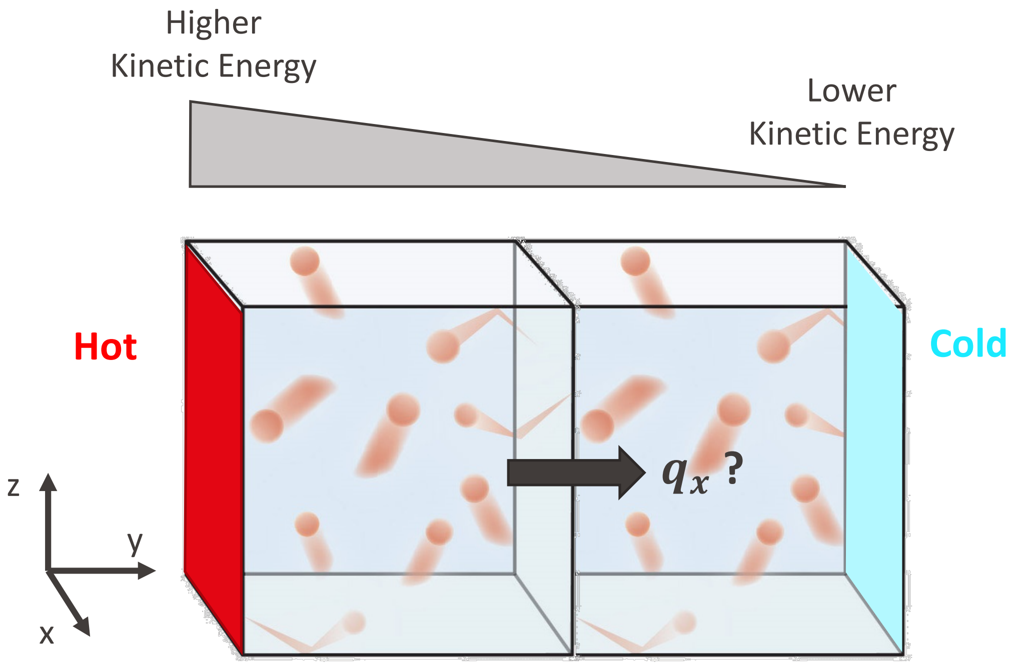
https://www.nature.com/articles/srep46092

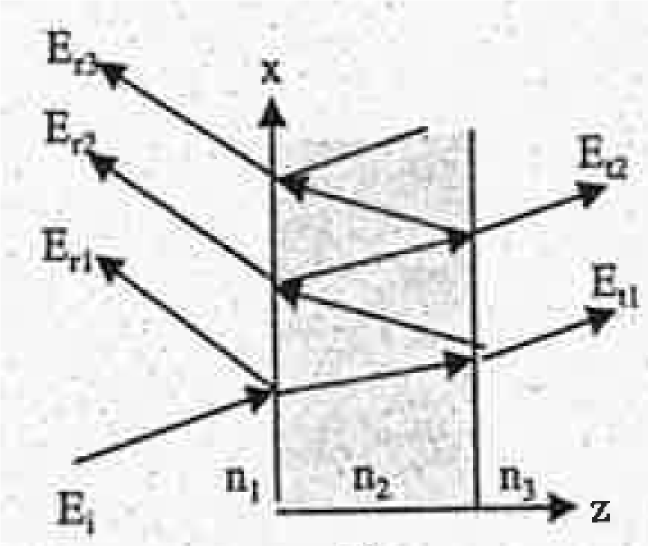


Phonons have the same statistics as photons and similar phenomena can be observed. In particular, when a superlattice is created, i.e. a periodic structure with repeated thin films of different materials, interference effects (analogous to Bragg-reflector effects) and tunneling effects can alter the thermal conductance.

Under the **assumption of no scattering**, the thermal conductance of a superlattice can be computed and signatures of tunneling and interference have been identified.

Energy Transport





Particle View

Wave View

How can we describe transport in a particle view accounting for the wave nature?

When and how does the wave-behavior affects energy transport?

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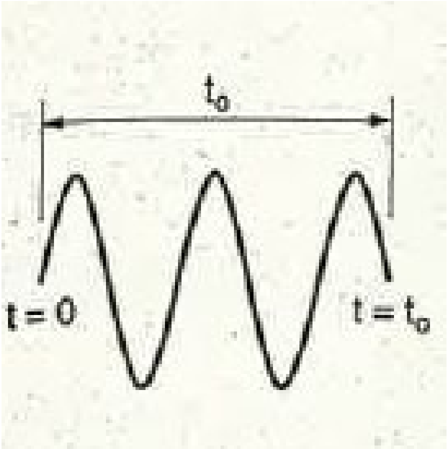
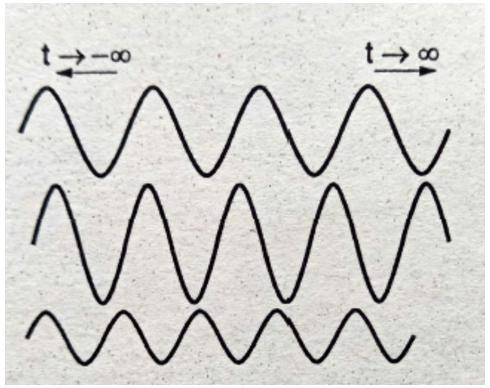
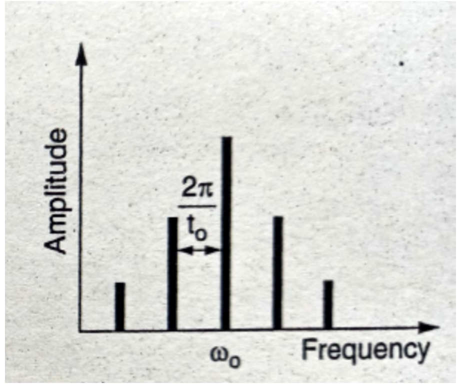
    - Thermal boundary resistance and Universal quantum thermal conductance
    - Near field heat transfer and Superlattices
- **From Waves to Particles**
    - Wave packets and group velocity
    - Coherence lengths and heat transfer models

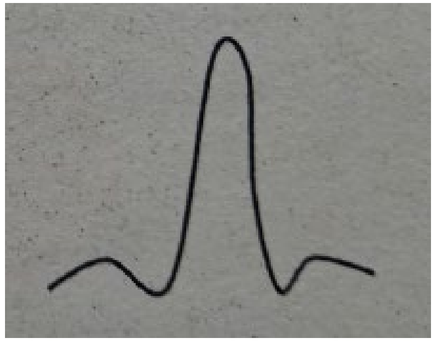
From Wave to Particle – Wave Packets and Group Velocity

Wave Packet



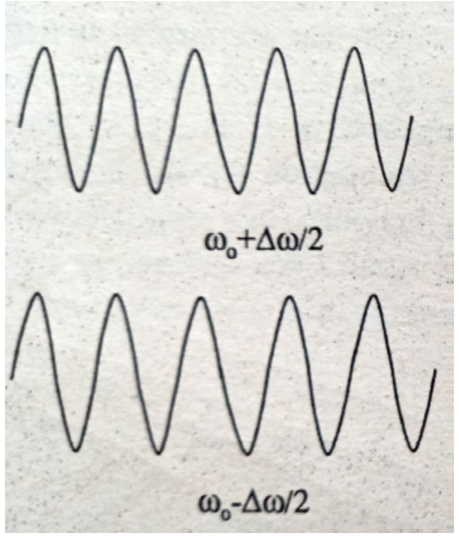


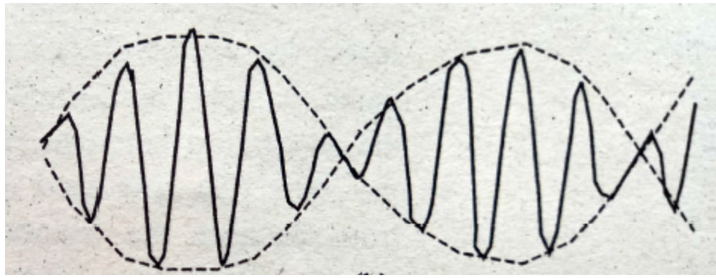




From Wave to Particle – Wave Packets and Group Velocity

Wave Packet HP: $\Delta\omega \ll \omega$





$$\begin{aligned} E_y(x,t) &= a\cos\left[\left(\omega_0-\frac{\Delta\omega}{2}\right)t-\left(k_0-\frac{\Delta k}{2}\right)x\right] \\ &\quad + a\cos\left[\left(\omega_0+\frac{\Delta\omega}{2}\right)t-\left(k_0-\frac{\Delta k}{2}\right)x\right] \\ &= 2a\cos(\Delta\omega t-\Delta k x)\cos(\omega_0 t-k_0 x) \end{aligned}$$

Energy propagation:

$$S(x,t)=\frac{4a^2 n}{c_0\mu}\cos^2\boxed{(t\,\Delta\omega-x\,\Delta k)} \Rightarrow v_{g,x}=\frac{\Delta\omega}{\Delta k}$$

Group velocity

In the most general case:

$$\boxed{\mathbf{v}_g=\nabla_{\mathbf{k}}\omega}=\frac{\partial\omega}{\partial k_x}\hat{\mathbf{k}}_x+\frac{\partial\omega}{\partial k_y}\hat{\mathbf{k}}_x+\frac{\partial\omega}{\partial k_z}\hat{\mathbf{k}}_z$$

!!! The group velocity corresponds to the energy propagation velocity only if $\Delta\omega \ll \omega$.

From Wave to Particle – Wave Packets and Group Velocity

Electrons

$$E = \frac{(\hbar \mathbf{k})^2}{2m}$$

$$E = \hbar\omega$$

$$\omega = E/\hbar \quad ➡ \quad v = \frac{\omega}{k} \qquad v_g = \frac{\partial\omega}{\partial k} \quad ➡$$

Phase velocity

$$v = \frac{\hbar k}{2m}$$

Group velocity

$$v_g = \frac{\hbar k}{m}$$

We observe that it is the group velocity, and not the phase velocity, that satisfies the de Broglie relationship: $p = \hbar k$

However, in crystals we have that the wavevectors determined by he Von Karmann boundary conditions do not satisfy the classical momentum relationship:

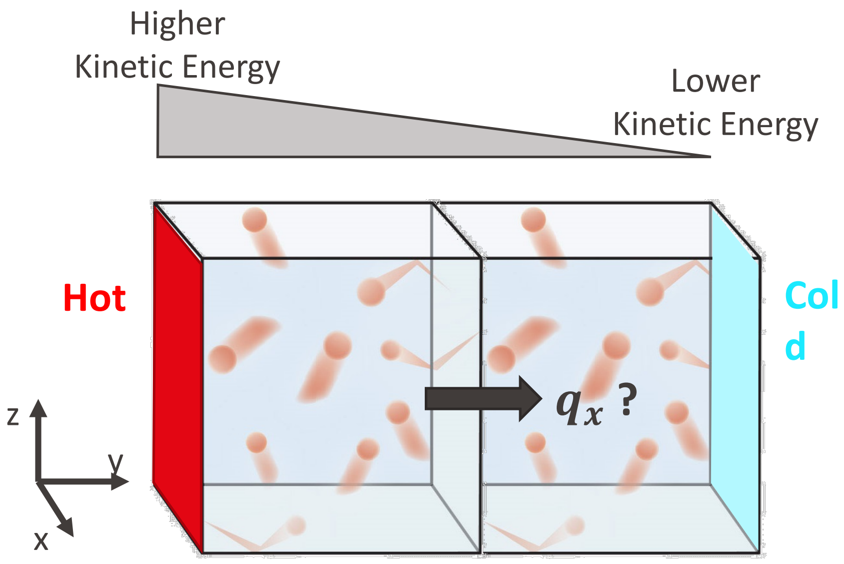
$$quantum\ p = \hbar k \neq m v_g = classical\ p$$

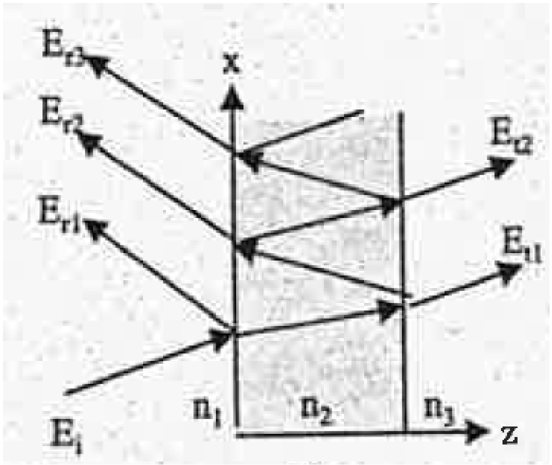
Thus we call $p = \hbar k = crystal\ momentum$

And we use it to satisfy the momentum conservation rules and calculate the external force field: $F_{ext} = \frac{\partial(\hbar k)}{\partial t}$

Indeed the periodic potential exerts its force onto the electrons. When using the crystal momentum however, one carries on the calculation as if the electrons were NOT subject to this internal field of the crystal. The group velocity defines the actual speed of motion of the electrons.

Energy Transport



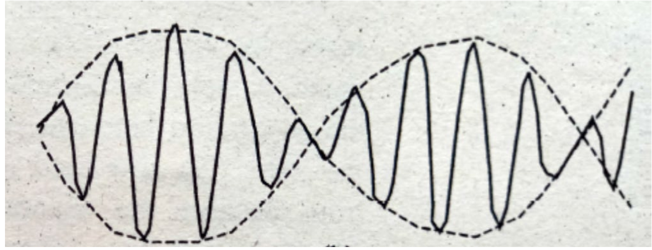


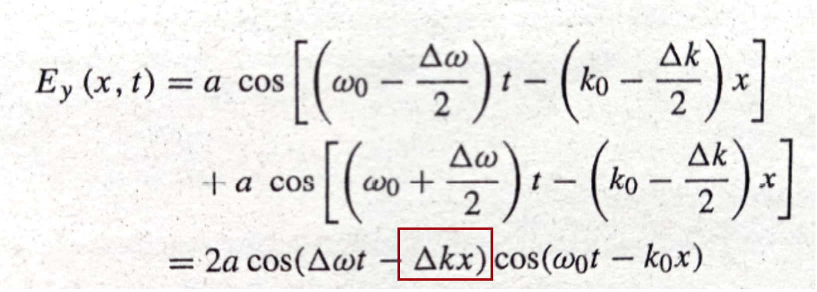
Should a wave or particle model be used to compute energy transport?

- If the phase of the carriers is important (interference effects), transport is coherent and the wave approach must be used (**ballistic transport**)
- If the phase of the carriers in unimportant, a particle approach can be used (**diffuse transport**)
- In between we have the **partially coherent regime**

Therefore, it is critical to **determine a coherence length** for the different types of waves in order to determine the most suitable energy transport model. We will now consider the three cases of electromagnetic waves, electron waves and acoustic waves.

Coherence Lengths – Electromagnetic Waves





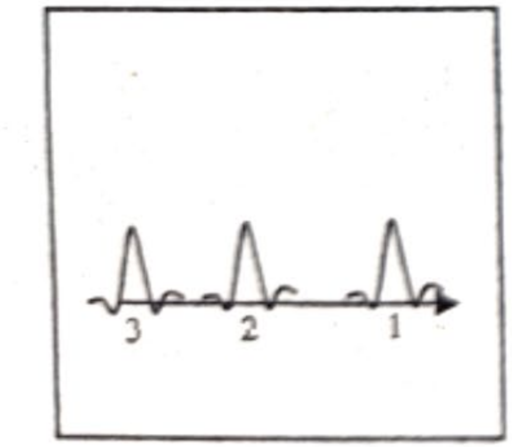
$$\begin{aligned} E_y(x,t) &= a\cos\left[\left(\omega_0 - \frac{\Delta\omega}{2}\right)t - \left(k_0 - \frac{\Delta k}{2}\right)x\right] \\ &\quad + a\cos\left[\left(\omega_0 + \frac{\Delta\omega}{2}\right)t - \left(k_0 - \frac{\Delta k}{2}\right)x\right] \\ &= 2a\cos(\Delta\omega t - \boxed{\Delta k x)}\cos(\omega_0 t - k_0 x) \end{aligned}$$

The spatial spread of a wave-packet is: $\Delta x \cdot \Delta k \approx 2\pi$ ➡ $\Delta x = l_c \approx \dfrac{2\pi}{\Delta k} = \dfrac{2\pi}{\Delta\omega/c} = \dfrac{c}{\Delta\nu}$

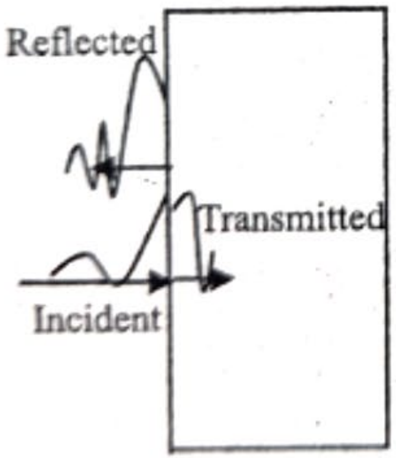
As a rule of thumb, give a spatial domain of characteristic dimension d:

- **Coherent/ballistic transport** : $l_c > d$
- **Incoherent / diffusive transport** : $l_c < d$
- **Partially coherent regime** : $l_c \approx d$

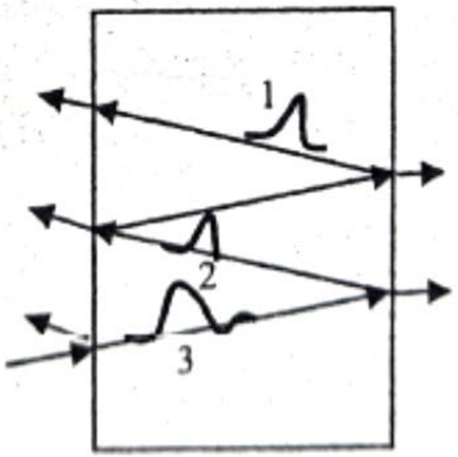
Coherence Lengths – Electromagnetic Waves



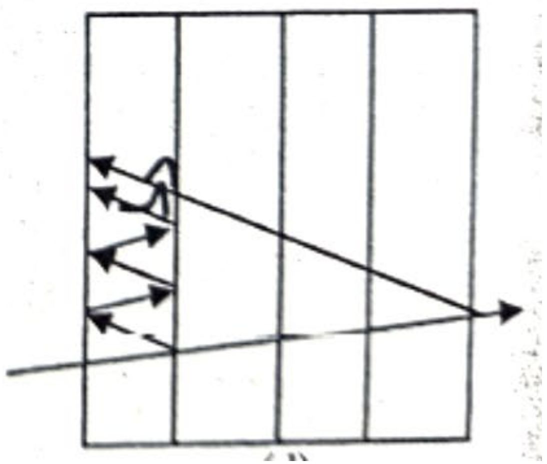
Different photons emitted by a source have no fixed phase relationship. Hence they do not interfere when they overlap and behave just like point particles.



At an interface, the incident and reflected wave packets have a fixed phase relationship. Hence they can interfere and a wave model is necessary.



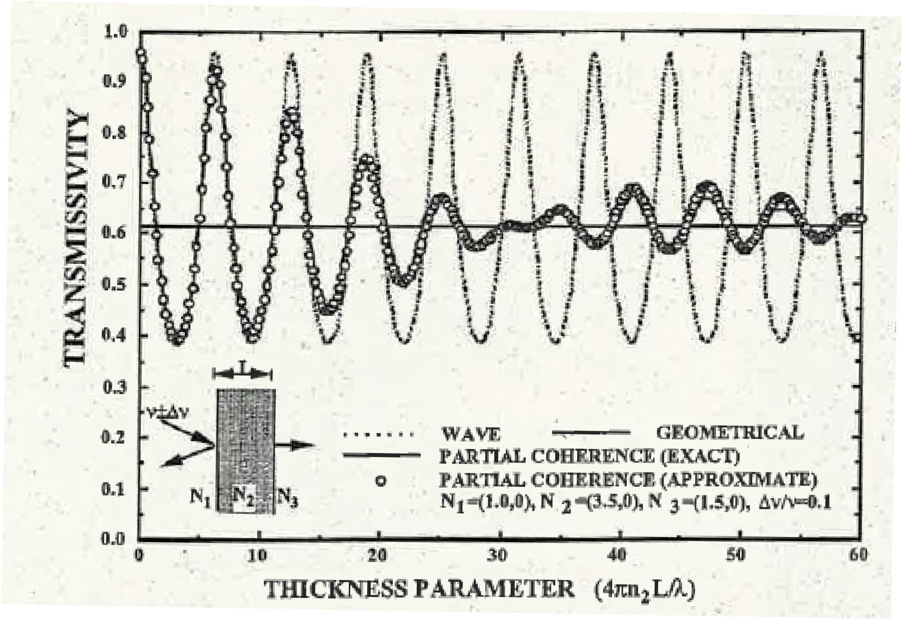
In a thick film ($d \gg l_c$), transmitted and reflected photons can overlap but they do not have a fixed phase relationship, hence they only transiently interfere. They can be treated as particles (ray tracing)



In a multilayer structure, the same wave packet is split many times and therefore can interfere with its previous reflection. A wave model is necessary.

Although the coherence length is a reasonable indicator of the type of regime, it can fail when multiple interfaces are involved.

Coherence Lengths – Electromagnetic Waves



To calculate the transmissivity or reflectivity of a wave packet, it is possible to perform a Fourier decomposition, calculate the transmissivity and reflectivity for each component and then obtain for the wave packet:

$$\bar{\tau} = \frac{J_t}{J_i} = \frac{\int_0^{\Delta\omega} \tau(\omega) J_i(\omega) d\omega}{\int_0^{\Delta\omega} J_i(\omega) d\omega}$$

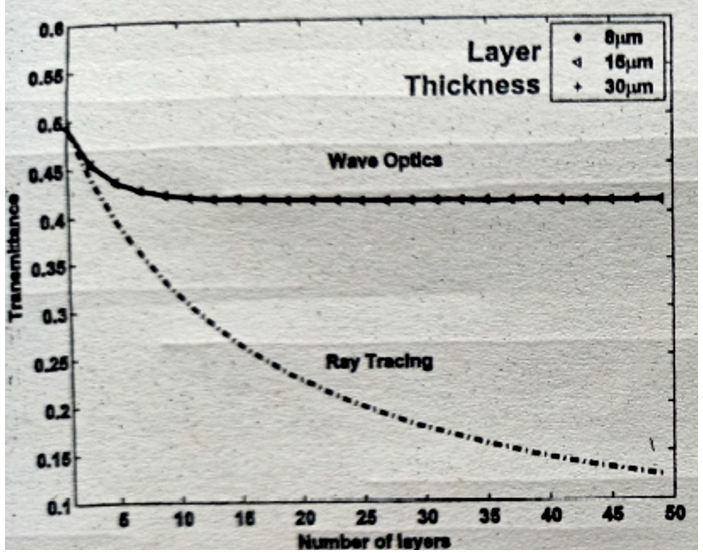
$$\overline{R} = \frac{J_t}{J_i} = \frac{\int_0^{\Delta\omega} R(\omega) J_i(\omega) d\omega}{\int_0^{\Delta\omega} J_i(\omega) d\omega}$$

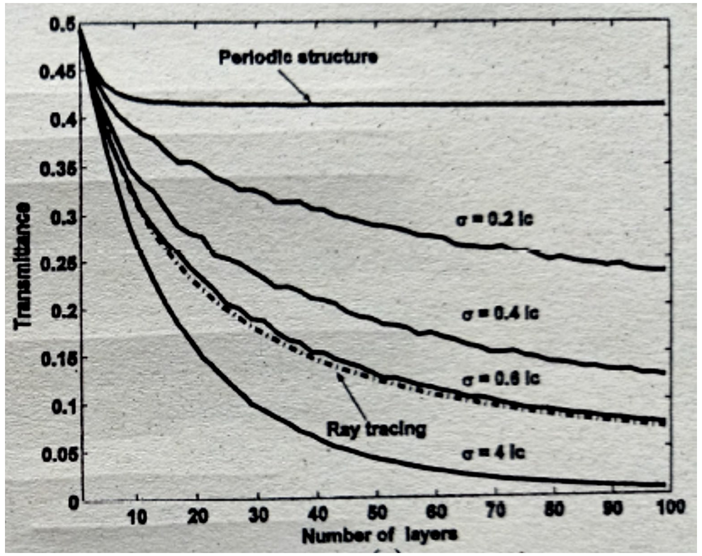
Where $J_i = Poynting\ vector$ and $\Delta\omega$ is the spectral width of the incident photon.

For the blackbody thermal radiation the energy spread is of the order of $k_B T$ therefore the thermal coherence length is

$$\ell_c = 0.15 \frac{hc}{\kappa_B T}$$

Coherence Lengths – Electromagnetic Waves





For Bragg reflectors the transmissivity calculated with the transfer matrix method reaches a constant value after a certain number of layer, independently of the thickness of the layers, while the ray-tracing method, which would be more suitable for a diffusive regime, shows a constantly decaying transmissivity value.

Important parameters defining the most suitable model will be (i) surface roughness, (ii) non-parallel surfaces (iii) thickness variations.

Randomness plays a very important role in multilayer structures. Indeed, random thickness variations can result in a phenomenon called *Anderson localization* which entails complete cancellation of waves at certain frequencies due superposition.

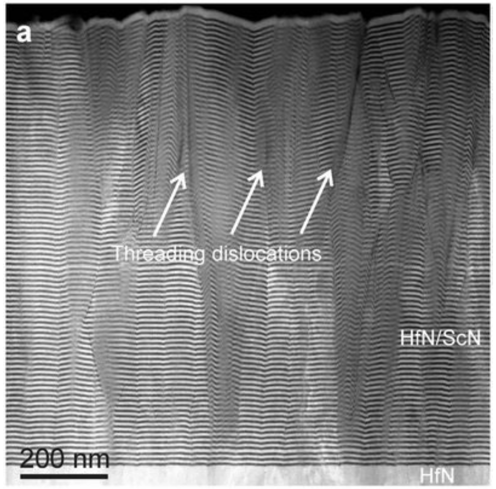
This result in wave localization at certain frequencies with a subsequent decrease in the transmissivity. This is shown in the graph on the left where, as a function of the standard deviation of the randomness, the behavior can go from a wave picture to a ray-tracing.

Localization is challenging to achieve in three-dimensional structures and often surface roughness can make even 2D structure effectively become 3D.

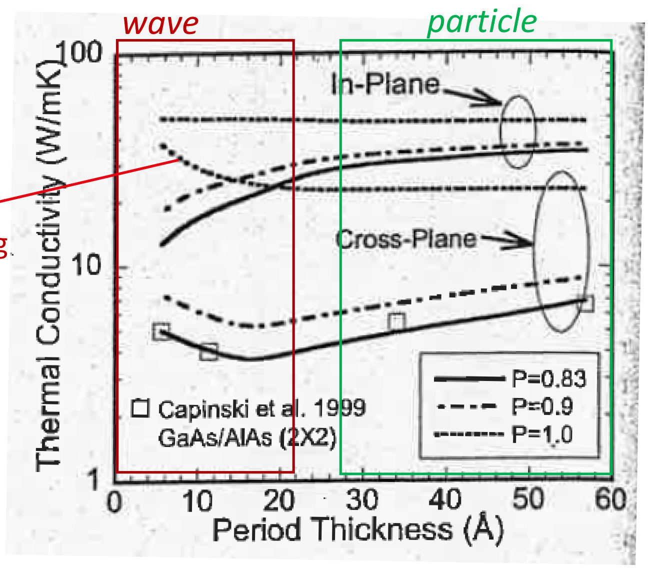
Coherence Lengths – Acoustic Waves

Each phonon has an energy $E = \hbar\omega$ (quantum oscillator) and we also know that in the Debye approximation we have $\omega = a\sqrt{K/mk}$. The dominant phonon wavelength is given by the Wien's displacement law for phonons: $\lambda_{p,peak} = \frac{hv_s}{4.965 k_B T}$. At room temperature the phonon wavelength is of the order of $\approx 10 - 20$ Å .

As a consequence, roughness present at the interface can easily cause scattering of phonons. Therefore, although scattering of phonons with boundaries and impurities is elastic, interface roughness tends to randomize the phase and the particle approach is likely to be valid in most situations. Nonetheless, it has been shown that in superlattices thermal conductivities both in-plane and out-of-plane are significantly reduced compared to the bulk values. Also, they depend on the structure of the superlattice.



https://www.nature.com/articles/srep46092



Photon tunneling increases k_{cross}

P = interface specularity parameter (specularly reflected phonons are assumed to be coherent)

Phonon-phonon scattering is an inelastic process and its mean free path can be very long ($2500 - 2300$ Å).

[1] https://www.sciencedirect.com/science/article/pii/S1631070516300871?via%3Dihub

Coherence Lengths – Electron Waves

While photon scattering is mostly an ELASTIC process (same energy and wavelength) ,

electron scattering can be both ELASTIC and INELASTIC (change in both direction and energy).

- Scattering with impurities and boundaries is typically elastic. Nonetheless, the random location of impurities can introduce sufficient randomness in the phase to make the particle approach valid
- Scattering with phonons is inelastic. The location of the scattering event is ever changing and hence he phase becomes randomized, making the particle approach valid.

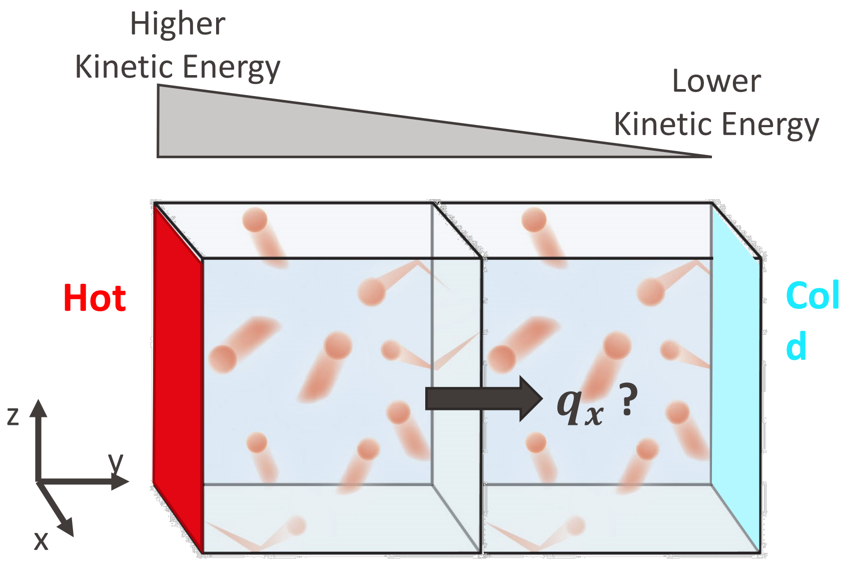
For electrons it is possible to define three critical lengths:

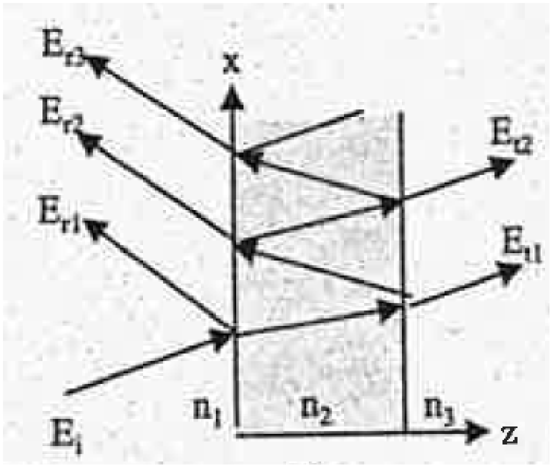
- Mean free path: average distance between successive scattering events $\Lambda \approx v_F \tau$ Where $\tau = relaxation\ time, v_F = fermi\ velocity$
  $\Lambda_{in} \approx v_F \tau_\phi$ Where $\tau_\phi = phase\ breaking\ time$ (inelastic scattering mean free path)
- Phase coherence length (Thouless length): depends on the diffusivity $\Lambda_\phi \approx \left(a\tau_\phi\right)^{1/2}$ Where $a = electron\ diffusivity \approx v_F^2 \tau$
- Thermal length: due to the thermal broadening $\propto \kappa_B T$ $\Lambda_T \approx (a\hbar/\kappa_B T)^{1/2}$

For $\Lambda_T > \Lambda_\phi$ ➡ Inelastic scattering is the dominant phase destroying mechanism. If the structure characteristic length $L > \Lambda_\phi$ then no quantum states are formed and therefore the particle approach should be used.

For $\Lambda_T < \Lambda_\phi$ ➡ Thermal excitation is the dominant dephasing mechanism. If $L > \Lambda_T$ the wave and particle approach will converge to the same result (except for periodic structures).

Energy Transport

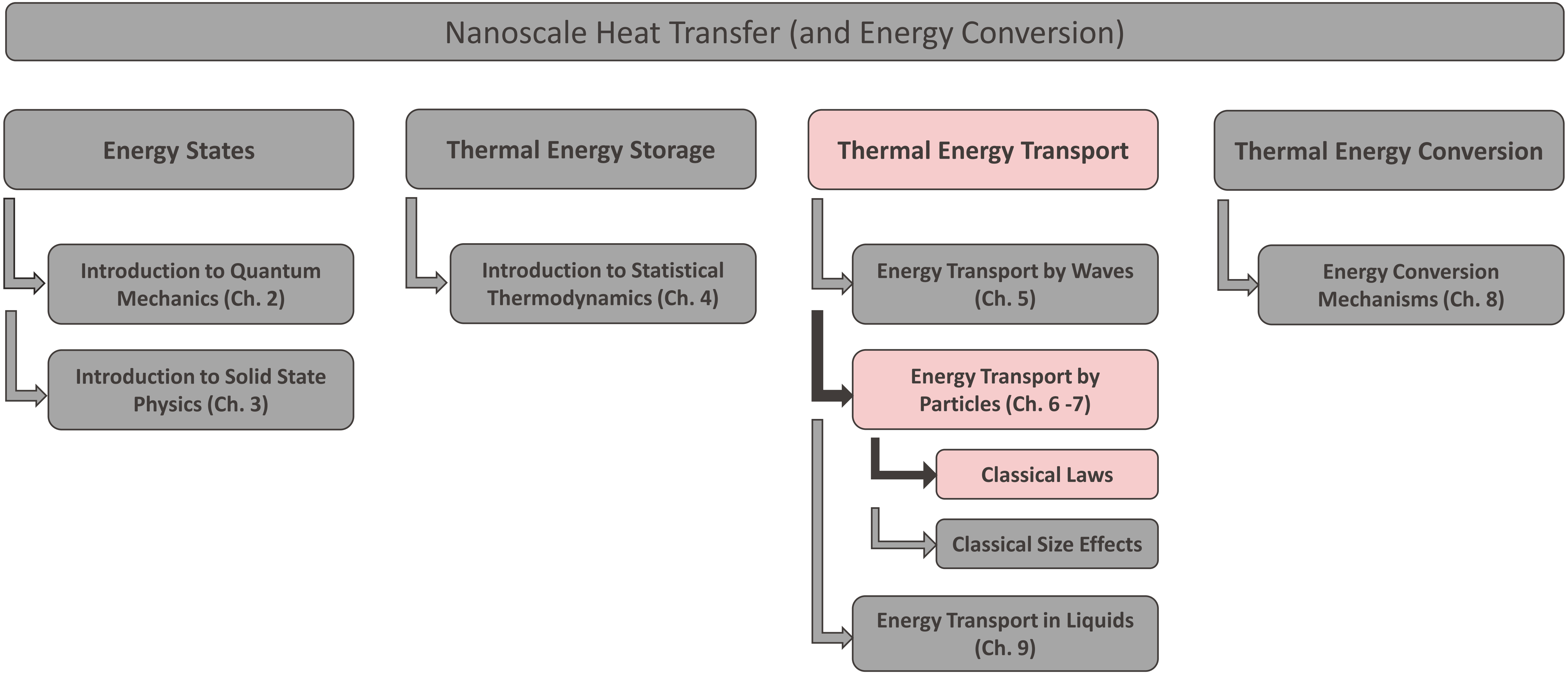




- If the phase of the carriers is important (interference effects), transport is coherent and the wave approach must be used (**ballistic transport**)
- If the phase of the carriers in unimportant, a particle approach can be used (**diffuse transport**)
- In between we have the **partially coherent regime**

Therefore, it is critical to **determine a coherence length** for the different types of waves in order to determine the most suitable energy transport model. We will now consider the three cases of electromagnetic waves, electron waves and acoustic waves.

Next Lecture



Nanoscale Heat Transfer (and Energy Conversion)

Energy States
- Introduction to Quantum Mechanics (Ch. 2)
- Introduction to Solid State Physics (Ch. 3)

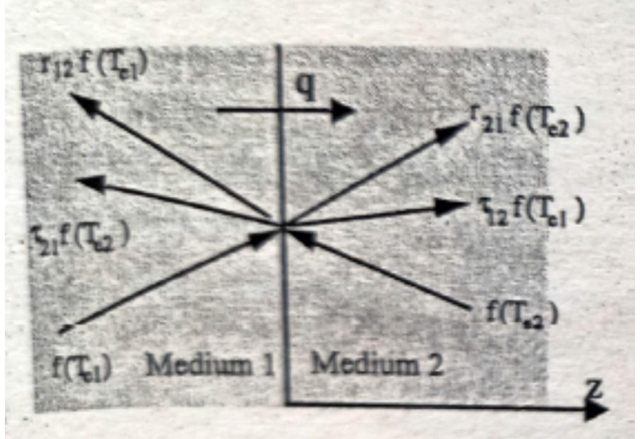
Thermal Energy Storage
- Introduction to Statistical Thermodynamics (Ch. 4)

Thermal Energy Transport
- Energy Transport by Waves (Ch. 5)
- Energy Transport by Particles (Ch. 6 -7)
  - Classical Laws
  - Classical Size Effects
- Energy Transport in Liquids (Ch. 9)

Thermal Energy Conversion
- Energy Conversion Mechanisms (Ch. 8)

Supplementary

Thermal Boundary Resistance



Let's consider the heat flux at the interface between two materials at respective temperatures $T_1 > T_2$.
Phonons travelling towards the interface from both sides will be transmitted and reflected.
If we take an isotropic material with only one atom per basis, based on our previous analysis we can write:

$$q_{1\to 2} = \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_{z1} \hbar\omega \tau_{12} f(\omega, T_{e1}) \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega$$

$$q = q_{1\to 2} - q_{2\to 1} = (T_{e1} - T_{e2}) \iint_{\Omega_1 > 2\pi} \left[\int_0^{\omega_{D1}} v_1 cos\theta_1 \hbar\omega \tau_{12} \frac{\partial f}{\partial T} \frac{D_1(\omega)}{4\pi} d\omega \right] d\Omega = \frac{T_{e1} - T_{e2}}{R_e}$$

➡ $R_e = thermal\ boundary\ resistance$ ➡ $$\frac{1}{R_e} = \frac{1}{4\pi} \int_0^{2\pi} d\phi_1 \int_0^{\pi/2} d\theta_1 \int_0^{\omega_{D1}} v_1 cos\theta_1 sin\theta_1 \hbar\omega \tau_{12}(\omega, \phi_1, \theta_1) \frac{\partial f(\omega, T)}{\partial T} D_1(\omega) d\omega$$

For our isotropic material ➡ $$\frac{1}{R_e} = \frac{1}{2} \int_0^1 \mu d\mu_1 \int_0^{\omega_{D1}} v_1 C_1(\omega) \tau_{12}(\omega, \mu_1)\, d\omega \qquad \mu = cos\theta \qquad C_1(\omega) = \hbar\omega \frac{\partial f(\omega, T)}{\partial T} D_1(\omega)$$

- The above expression can be calculated using the Snell's law for phonon only if phonon scattering at the interface is specular and elastic (no change in frequency). This requirements are satisfied only at very low temperatures when the phonon wavelengths are long. In this regime we observe:

$$C(\omega) \propto T^3 \quad ➡ \quad R_e \propto \frac{1}{T^3}$$

- At room temperature the phonon wavelength is of the order of $\approx 10 - 20$ Å . As a consequence, roughness present at the interface can easily cause scattering of phonons. Hence, specular and elastic conditions are no longer satisfied and we need a model for the transmissivity at high T to be able to calculate the boundary resistance.

Thermal Boundary Resistance

Diffuse mismatch model – phonons emerging from the interface bear no relationship with their origin

$$R_{12} = \tau_{21} \text{ or } 1 - \tau_{12} = \tau_{21}$$

HP. linear dispersion of acoustic phonons $\omega = a\sqrt{K/m}k$

➡ $$\tau_{d12} = \frac{1/v_2^2}{1/v_1^2 + 1/v_2^2}$$

(Low T approximation)

➡ $$\tau_{d12}(T_e) = \frac{v_2 U_2(T_e)}{v_1 U_1(T_e) + v_2 U_2(T_e)} = \frac{1}{1 + [v_1 U_1 / v_2 U_2]}$$

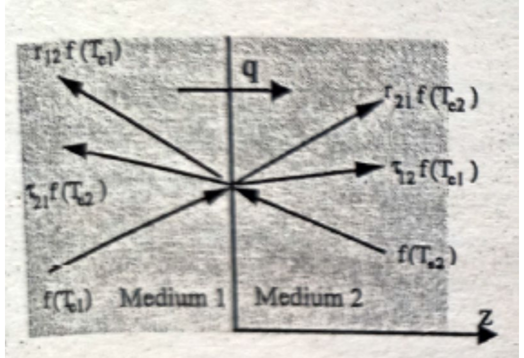
(High T approximation)

$$U = \int_0^{\omega_D} \hbar\omega f(\omega, T_e) D(\omega) d\omega = \int_0^{T_e} C(T) dT$$

We observe that this approximation cannot be valid when the two materials are very similar and fails completely when the two materials are equal, i.e. we are considering an imaginary interface within the same material. Indeed in this limit the thermal boundary resistance should be zero but none of these models capture this correctly.

This leads us to an important observation.

Thermal Boundary Resistance – Equilibrium Temperature



Previously we have written:

$$q = \frac{T_{e1} - T_{e2}}{R_e}$$

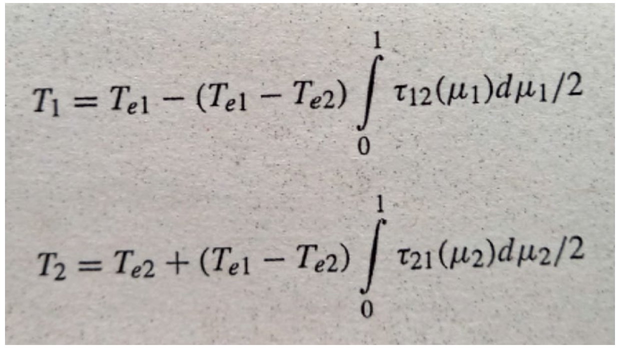
Where T_e are the phonon temperatures. Yet, we now realizes that we are in a **highly non-equilibrium situation** and it is hard to correctly define a temperature.

Indeed, at the interface we have three distinct groups of phonons:

- Incoming phonons with temperature T_{e1}
- Reflected phonons with energy distribution depending on the convolution of the incoming phonons and the interface reflectivity
- Transmitted phonons with an energy distribution depending on the convolution of the phonons arriving from the medium 2 and the interface transmittivity

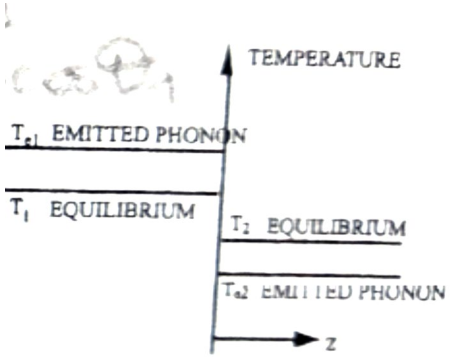
Therefore the phonon energy spectra at the interface are very different from those of the incoming phonons and cannot be represented by an equilibrium distribution with a single temperature T.

Instead, assuming that the phonons adiabatically approach an equilibrium condition, we define an **equivalent equilibrium temperature**, which is a measure of the local energy density rather than the spectral characteristics of the energy distribution:

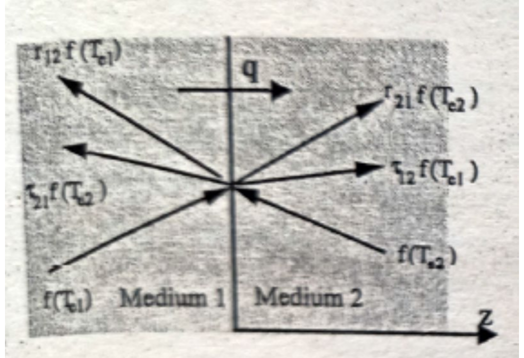


$$T_1 = T_{e1} - (T_{e1} - T_{e2}) \int_0^1 \tau_{12}(\mu_1) d\mu_1/2$$

$$T_2 = T_{e2} + (T_{e1} - T_{e2}) \int_0^1 \tau_{21}(\mu_2) d\mu_2/2$$



Thermal Boundary Resistance – Equilibrium Temperature



We thus re-write:

$$\Re = \frac{T_1 - T_2}{q} = \frac{2[1 - \langle \int_0^1 \tau_{12}(\mu_1) d\mu_1 + \int_0^1 \tau_{21}(\mu_2) d\mu_2 \rangle / 2]}{\int_0^1 [\int \tau_{12}(\mu_1) v_1 C_1(\omega) d\omega] \mu_1 d\mu_1}$$

In practice, we can rarely measure T_e and therefore the thermal boundary resistance is obtained on the basis of the equivalent thermal equilibrium temperature. This must be taken into account when analyzing the experimental data with the models discussed above.

It is then clear that, even for an ideal interface, there will be a thermal boundary resistance as long as reflection occurs at such interface.
For nearly ideal interfaces, the order of magnitude of R_e is:

$$R_e \sim \frac{1}{Cv}$$

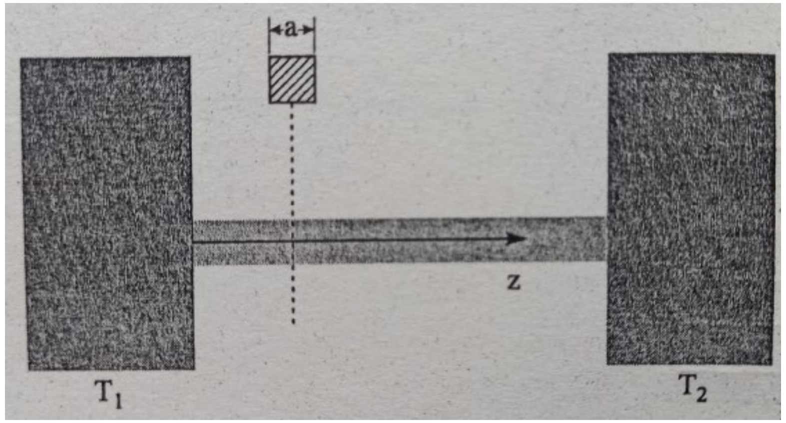
$C = volumetric\ specfic\ heat \approx 10^6\ J\ m^{-3} K^{-1}\ @300\ K$

$v = phonon\ speed \approx 10^3\ m\ s^{-1}\ @300\ K$

➡ $R_e \sim 10^{-8} - 10^{-9} K m^2 W^{-1}$

This result is consistent with experimental data. Non-ideal interfaces will have larger values.
Although this value is very small for a single interface, for nanoscale system with a large number of interfaces this resistance becomes dominant. Importantly, multilayer systems can exhibit distinct behaviors due to interference effects.
In macro-scale systems, this value can also be large due to the absence of perfect contact.

Universal Quantum Thermal Conductance



Let’s now consider a nanowire with square cross section that connects two thermal reservoirs.
Let’s neglect internal scattering and assume that the phonon transmissivity is unity for each mode.
We now want to calculate the thermal conductance of the nanowire:

$$K = \frac{q_{12}}{T_1 - T_2}$$

where

$$q_{12} = \sum_{p=1}^{3m}\left[\frac{1}{V}\sum_{k_{x1}=-k_{max}}^{k_{max}}\;\sum_{k_{y1}=-k_{max}}^{k_{max}}\;\sum_{k_{z1}=0}^{k_{max}} v_{z1}\hbar\omega\left(f(\omega,T_1)-f(\omega,T_2)\right)\right]$$

We thus need to compute the allowable wavevectors in this system. In the xy plane the wire acts as a 2D potential well and therefore the allowable modes are:

$$k_x = 2\pi\frac{m}{2a} = \frac{\pi m}{a},\; k_y = \frac{n\pi}{a}\;(m, n = \pm 1, \pm 2, \ldots)$$

$$\omega = ck = c\sqrt{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{a}\right)^2 + k_z^2}$$

$$v_{z1} = \frac{d\omega}{dk_z} = group\ velocity$$

$$q_{12} = 3\frac{1}{2\pi}\sum_{m,n}\int v_{z1}\hbar\omega\left(f(\omega,T_1)-f(\omega,T_2)\right)dk_z = 3\frac{1}{2\pi}\sum_{m,n}\int \hbar\omega\left(f(\omega,T_1)-f(\omega,T_2)\right)d\omega$$

$$q_{12} = 3\frac{(T_1 - T_2)}{2\pi}\sum_{m,n}\int_{\omega_{mn}}^{\omega_{max}}\hbar\omega\frac{\partial f(\omega,T)}{\partial T}d\omega$$

Where

$$\omega_{mn} = c\sqrt{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{a}\right)^2}$$

➡ $$K = \frac{3}{2\pi}\sum_{m,n}\int_{\omega_{mn}}^{\omega_{max}}\hbar\omega\frac{\partial f(\omega,T)}{\partial T}d\omega$$