

Handout 21

Phonon Thermal Statistics and Heat Capacities

In this lecture you will learn:

- Phonon occupation statistics
- Bose-Einstein distribution
- Phonon density of states in 1D, 2D, and 3D
- Phonon thermal energy and heat capacity of solids



Peter Debye
Born: 1884 (Netherlands)
Died: 1966 (Ithaca, NY)

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A Single Lattice Wave Mode

Consider the Hamiltonian of just a **single** lattice wave mode:

$$\hat{H} = \hbar \omega(\vec{q}) \left(\hat{a}^+(\vec{q}) \hat{a}(\vec{q}) + \frac{1}{2} \right)$$

Its eigenstates, and the corresponding eigenenergies, are:

$$|n\rangle \quad \{ \text{where } n = 0, 1, 2, 3, \dots \}$$

$$\begin{aligned} \hat{H}|n\rangle &= \hbar \omega(\vec{q}) \left(\hat{a}^+(\vec{q}) \hat{a}(\vec{q}) + \frac{1}{2} \right) |n\rangle \\ &= \hbar \omega(\vec{q}) \left(n + \frac{1}{2} \right) |n\rangle \\ &= E(n) |n\rangle \end{aligned}$$

The state $|n\rangle$ corresponds to “ n ” phonons in the lattice wave mode

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A Single Lattice Wave Mode in Thermal Equilibrium

$$\hat{H}|n\rangle = \hbar \omega(\vec{q}) \left(n + \frac{1}{2} \right) |n\rangle = E(n) |n\rangle$$

Thermal Equilibrium

In thermal equilibrium, let $P(n)$ be the probability that there are “ n ” phonons in this lattice wave mode

$P(n)$ must be related to the energy corresponding to the “ n ” phonons:

$$P(n) \propto e^{-\frac{E(n)}{KT}} = e^{-\frac{\hbar \omega(\vec{q})(n+1/2)}{KT}} \quad \longrightarrow \quad (1)$$

$P(n)$ must be normalized properly:

$$\sum_{n=0}^{\infty} P(n) = 1 \quad \longrightarrow \quad (2)$$

(1) and (2) give:

$$P(n) = e^{-\frac{\hbar \omega(\vec{q})n}{KT}} \left[1 - e^{-\frac{\hbar \omega(\vec{q})}{KT}} \right]$$

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Bose-Einstein Distribution

The probability distribution given by,

$$P(n) = e^{-\frac{\hbar \omega(\vec{q})n}{KT}} \left[1 - e^{-\frac{\hbar \omega(\vec{q})}{KT}} \right]$$

is called the Bose-Einstein distribution

Average Phonon Number:

One can calculate the average phonon number in equilibrium:

$$\langle n \rangle = \sum_{n=0}^{\infty} n P(n) = \frac{1}{e^{\hbar \omega(\vec{q})/KT} - 1}$$

Average phonon number in any lattice wave mode depends on the phonon energy

Limiting Cases:

$$KT \gg \hbar \omega(\vec{q}) \Rightarrow \langle n \rangle = \frac{1}{e^{\hbar \omega(\vec{q})/KT} - 1} \approx \frac{KT}{\hbar \omega(\vec{q})} \Rightarrow \langle E \rangle = \hbar \omega(\vec{q}) \langle n \rangle \approx KT$$

$$KT \ll \hbar \omega(\vec{q}) \Rightarrow \langle n \rangle = \frac{1}{e^{\hbar \omega(\vec{q})/KT} - 1} \approx e^{-\hbar \omega(\vec{q})/KT}$$

↑
Classical
equipartition
theorem

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Classical Equipartition Theorem

Every independent quadratic term in position or momentum in the expression for the energy of a system has an average value equal to $KT/2$ at temperature T

- Only holds when classical statistics apply - which is generally the case at high enough temperatures

Example: A Free Particle in 1D

$$E = \frac{p_x^2}{2m} \Rightarrow \langle E \rangle = \frac{1}{2} KT$$

Example: A Free Particle in 3D

$$E = \frac{p_x^2}{2m} + \frac{p_y^2}{2m} + \frac{p_z^2}{2m} \Rightarrow \langle E \rangle = \frac{3}{2} KT$$

Example: A Classical Simple Harmonic Oscillator in 1D

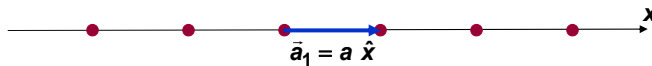
$$E = \frac{p_x^2}{2m} + \frac{1}{2} kx^2 \Rightarrow \langle E \rangle = KT$$

Example: A Single Lattice Wave Mode of a 1D Crystal

$$E = \frac{N}{2M} P(\vec{q}, t) P^*(\vec{q}, t) + \frac{NM}{2} \omega^2(\vec{q}) U(\vec{q}, t) U^*(\vec{q}, t) \Rightarrow \langle E \rangle = KT$$

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Acoustic Phonons in 1D: Density of States



Consider acoustic phonons in a N -primitive-cell 1D crystal of length L :

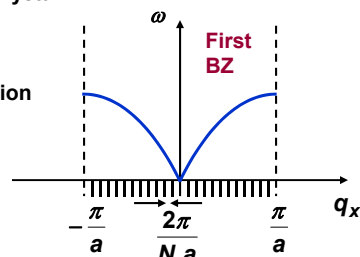
$$L = N a$$

First we need to figure out how to convert a summation over all lattice wave modes of the form:

$$\sum_{q_x \text{ in FBZ}}$$

into an integral for the form:

$$\int_{-\pi/a}^{\pi/a} dq_x$$



We now that there are N different allowed wavevector values in FBZ (in interval $2\pi/a$) So in interval dq_x there must be $(Na/2\pi) dq_x$ different wavevector values:

$$\Rightarrow \sum_{q_x \text{ in FBZ}} \rightarrow Na \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi} \rightarrow L \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi}$$

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Acoustic Phonons in 1D: Density of States

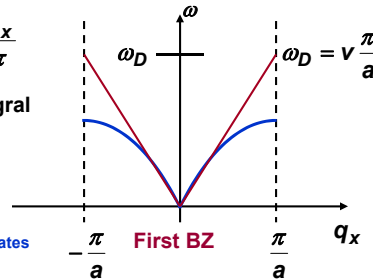
$$\Rightarrow \sum_{q_x \text{ in FBZ}} \rightarrow Na \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi} \rightarrow L \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi}$$

Now we need to figure out how to convert an integral of the form:

$$L \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi}$$

into an integral over frequency of the form:

$$L \int_0^{\omega_D} d\omega g_{1D}(\omega) \quad \leftarrow \text{Density of states}$$



We need to know the dispersion of the phonons. We approximate it by a linear function:

$$\omega = v q_x$$

Therefore:

$$L \int_{-\pi/a}^{\pi/a} \frac{dq_x}{2\pi} \rightarrow 2L \int_0^{\pi/a} \frac{dq_x}{2\pi} \rightarrow \frac{L}{\pi} \int_0^{\omega_D} \left| \frac{dq_x}{d\omega} \right| d\omega \rightarrow L \int_0^{\omega_D} d\omega \frac{1}{\pi v}$$

The density of states function $g_{1D}(\omega)$ is the number of phonon modes per unit frequency interval per unit length:

$$g_{1D}(\omega) = \frac{1}{\pi v}$$

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Acoustic Phonons in 1D: Debye Frequency

We know that:

$$\sum_{q_x \text{ in FBZ}} 1 = N$$

Since:

$$\sum_{q_x \text{ in FBZ}} 1 = L \int_0^{\omega_D} d\omega g_{1D}(\omega)$$

We must have:

$$L \int_0^{\omega_D} d\omega g_{1D}(\omega) = N \quad \rightarrow (1)$$

Since:

$$\omega_D = v \frac{\pi}{a} \quad g_{1D}(\omega) = \frac{1}{\pi v}$$

It can be verified that (1) above holds

The frequency ω_D is called the Debye frequency (after Peter Debye - Cornell University). It is chosen to ensure that the total number of phonon modes are conserved when going from q-space integrals to frequency domain integrals. In 1D this is automatic.

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Acoustic Phonons in 2D: Density of States

Consider acoustic phonons in a N -primitive-cell 2D crystal of area A

We need to go from a q -space integral to a frequency integral:

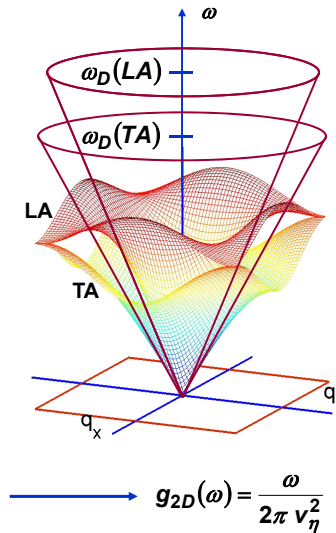
$$\sum_{\vec{q} \text{ in FBZ}} \rightarrow A \int_0^{\omega_D} d\omega g_{2D}(\omega)$$

We need to know the dispersion for the 2 acoustic phonon bands. We assume that for both phonon bands the dispersion is linear:

$$\omega = v_\eta q \quad \{ \eta = 1, 2 \text{ for LA, TA} \}$$

For each phonon band we get:

$$\begin{aligned} \sum_{\vec{q} \text{ in FBZ}} &\rightarrow A \int_{\text{FBZ}} \frac{d^2 \vec{q}}{(2\pi)^2} \rightarrow A \int_0^{\omega_D} \frac{2\pi q dq}{(2\pi)^2} \\ &\rightarrow \frac{A}{2\pi} \int_0^{\omega_D} q \left| \frac{dq}{d\omega} \right| d\omega \rightarrow A \int_0^{\omega_D} d\omega \frac{\omega}{2\pi v_\eta^2} \end{aligned}$$



The question is what is ω_D ?

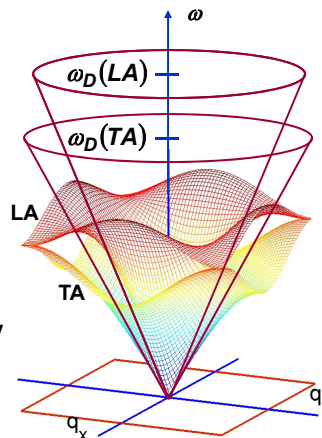
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Acoustic Phonons in 2D: Debye Frequency

To find ω_D we count and conserve the total number of phonon modes in each band:

$$\begin{aligned} \sum_{\vec{q} \text{ in FBZ}} &= N \\ \Rightarrow A \int_0^{\omega_D} d\omega \frac{\omega}{2\pi v_\eta^2} &= N \\ \Rightarrow A \frac{\omega_D^2}{4\pi v_\eta^2} &= N \\ \Rightarrow \omega_D &= \sqrt{4\pi v_\eta^2 \frac{N}{A}} \end{aligned}$$

Each phonon band has a different Debye frequency



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Acoustic Phonons in 3D: Density of States

Consider acoustic phonons in a N -primitive-cell 3D crystal of volume V

We need to go from a q -space integral to a frequency integral:

$$\sum_{\vec{q} \text{ in FBZ}} \rightarrow V \int_0^{\omega_D} d\omega g_{3D}(\omega)$$

We need to know the dispersion for the 3 acoustic phonon bands. We assume that for all 3 phonon bands the dispersion is linear:

$$\omega = v_\eta q \quad \{ \eta = 1,2,3 \text{ for LA,TA,TA} \}$$

For each phonon band we get:

$$\begin{aligned} \sum_{\vec{q} \text{ in FBZ}} &\rightarrow V \int_{\text{FBZ}} \frac{d^3 \vec{q}}{(2\pi)^3} \rightarrow V \int_0^{\omega_D} \frac{4\pi q^2 dq}{(2\pi)^3} \\ &\rightarrow \frac{V}{2\pi^2} \int_0^{\omega_D} q^2 \left| \frac{dq}{d\omega} \right| d\omega \rightarrow V \int_0^{\omega_D} d\omega \frac{\omega^2}{2\pi^2 v_\eta^3} \longrightarrow g_{3D}(\omega) = \frac{\omega^2}{2\pi^2 v_\eta^3} \end{aligned}$$

The question is what is ω_D ?

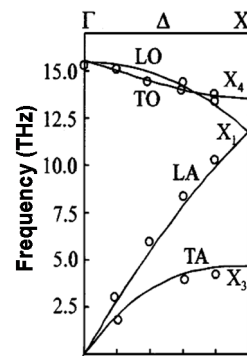
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Acoustic Phonons in 3D: Debye Frequency

To find ω_D we count and conserve the total number of phonon modes in each band:

$$\begin{aligned} \sum_{\vec{q} \text{ in FBZ}} &= N \\ \Rightarrow V \int_0^{\omega_D} d\omega \frac{\omega^2}{2\pi^2 v_\eta^3} &= N \\ \Rightarrow V \frac{\omega_D^3}{6\pi^2 v_\eta^3} &= N \\ \Rightarrow \omega_D &= \left(6\pi^2 v_\eta^3 \frac{N}{V} \right)^{1/3} \end{aligned}$$

Each phonon band has a different Debye frequency



Silicon Phonon Bands

Silicon:

In Silicon the TA phonon velocity is 5.86 km/s. The corresponding Debye frequency is 13.4 THz. The LA phonon velocity is 8.44 km/s. The corresponding Debye frequency is 19.3 THz.

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Acoustic Phonons in 3D: Thermal Energy

Consider acoustic phonons in a N -primitive-cell 3D crystal of volume V

Also assume that all three acoustic phonon modes have the same velocity (for simplicity)

$$\omega = v q \quad \{ \text{for LA, TA, TA} \}$$

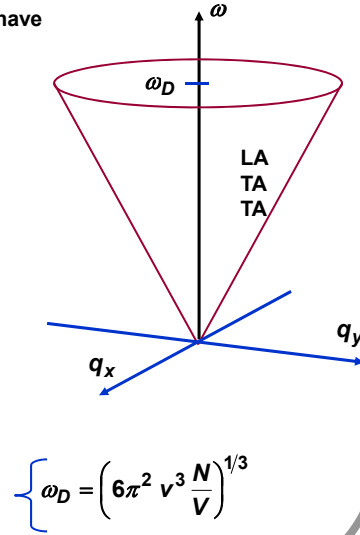
Then for each phonon band we have:

$$g_{3D}(\omega) = \frac{\omega^2}{2\pi^2 v^3}$$

The energy u of the lattice per unit volume at temperature T can be written as:

$$\begin{aligned} u &= 3 \times \frac{1}{V} \sum_{\vec{q} \text{ in FBZ}} \hbar \omega(\vec{q}) \langle n_{\vec{q}} \rangle \\ &= 3 \times \int_0^{\omega_D} d\omega g_{3D}(\omega) \frac{\hbar \omega}{e^{\hbar \omega / KT} - 1} \\ &= \frac{3\hbar}{2\pi^2 v^3} \int_0^{\omega_D} d\omega \frac{\omega^3}{e^{\hbar \omega / KT} - 1} \end{aligned}$$

3 degenerate phonon bands



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Acoustic Phonons in 3D: Thermal Energy

$$u = \frac{3\hbar}{2\pi^2 v^3} \int_0^{\omega_D} d\omega \frac{\omega^3}{e^{\hbar \omega / KT} - 1}$$

Define dimensionless variable "x" as:

$$x = \frac{\hbar \omega}{KT} \quad \Rightarrow \quad x_D = \frac{\hbar \omega_D}{KT}$$

To get:

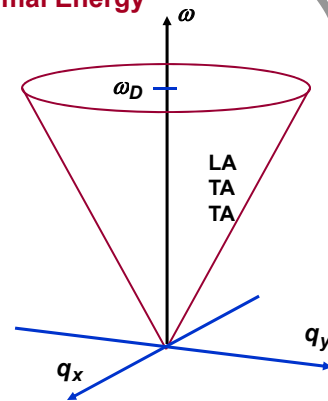
$$u = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \int_0^{x_D} dx \frac{x^3}{e^x - 1}$$

Case I: $KT \ll \hbar \omega_D \Rightarrow x_D \gg 1$

$$u = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \int_0^{\infty} dx \frac{x^3}{e^x - 1} = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \left(\frac{\pi^4}{15} \right) = \frac{\pi^2 (KT)^4}{10 (\hbar v)^3}$$

Specific Heat or Heat Capacity:

$$C = \frac{du}{dT} = \frac{2\pi^2 K^4 (T)^3}{5 (\hbar v)^3} \quad \longrightarrow \quad \text{Debye's famous } T^3 \text{ law}$$



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Acoustic Phonons in 3D: Debye Temperature

The low temperature limit:

$$KT \ll \hbar\omega_D$$

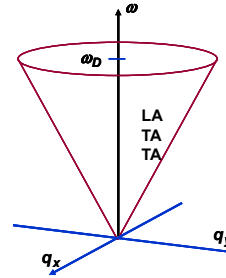
can also be written as:

$$T \ll \theta_D$$

Where θ_D is the Debye temperature:

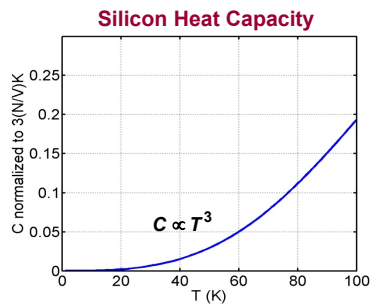
$$\theta_D = \frac{\hbar\omega_D}{K}$$

The Debye frequency thus defines a natural temperature scale for the phonon energetics



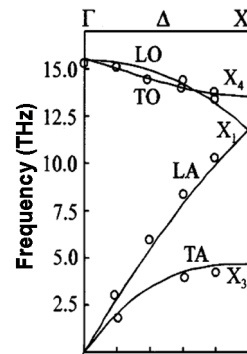
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Silicon Heat Capacity



In silicon where the Debye frequency for TA phonons is 13.4 THz, the corresponding Debye temperature is 643 K. The Debye frequency for LA phonons is 19.3 THz and the corresponding Debye temperature is 926 K

The T^3 law for heat capacity holds well in Silicon for temperatures less than 50 K (much less than the Debye temperature of any phonon band)



Silicon Phonon Bands

$$\omega_D(LA) = 19.3 \text{ THz}$$

$$\theta_D(LA) = 926 \text{ K}$$

$$\omega_D(TA) = 13.4 \text{ THz}$$

$$\theta_D(TA) = 643 \text{ K}$$

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Acoustic Phonons in 3D: Classical Equipartition Theorem

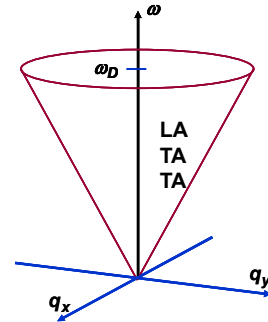
Case II: $KT \gg \hbar\omega_D \Rightarrow T \gg \theta_D$

$$u = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \int_0^{x_D} dx \frac{x^3}{e^x - 1}$$

$$\Rightarrow e^x - 1 \approx x$$

$$u = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \int_0^{x_D} dx x^2 = \frac{3}{2\pi^2 (\hbar v)^3} (KT)^4 \frac{x_D^3}{3}$$

$$= \frac{KT}{2\pi^2 v^3} \omega_D^3 = 3 \left(\frac{N}{V} \right) KT \quad \left\{ \omega_D = \left(6\pi^2 v^3 \frac{N}{V} \right)^{1/3} \right.$$

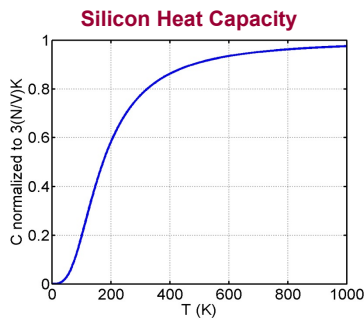


Physical explanation: There are N/V phonon modes per band per unit volume and each mode has energy equal to KT as per the **classical equipartition theorem**

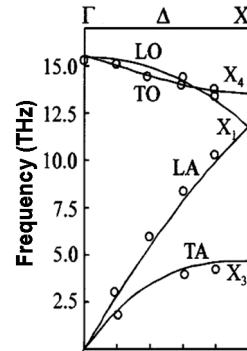
Specific Heat: $C = \frac{du}{dT} = 3 \left(\frac{N}{V} \right) K \longrightarrow$ **Dulong and Petit Law (1819)**

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Silicon Heat Capacity



The Heat capacity approaches $3(N/V)K$ as the temperature exceeds the Debye temperature of all acoustic phonon bands



Silicon Phonon Bands

$$\omega_D(LA) = 19.3 \text{ THz}$$

$$\theta_D(LA) = 926 \text{ K}$$

$$\omega_D(TA) = 13.4 \text{ THz}$$

$$\theta_D(TA) = 643 \text{ K}$$

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Optical Phonons in 1D: Einstein Model and Density of States

Consider optical phonons in a N -primitive-cell 1D crystal of length L

Let the optical phonon frequency be $\omega_{LO}(\vec{q})$

We want to be able to write:

$$\sum_{q_x \text{ in FBZ}} = L \int_0^{\omega_{LO}} d\omega g_{1D}(\omega)$$

We suppose that all optical phonon modes in FBZ have the same frequency ω_{LO} (i.e. the phonon band is completely flat - Einstein model):

$$\Rightarrow g_{1D}(\omega) \propto C \delta(\omega - \omega_{LO}) \quad \longrightarrow \quad \text{What is } C?$$

We know that:

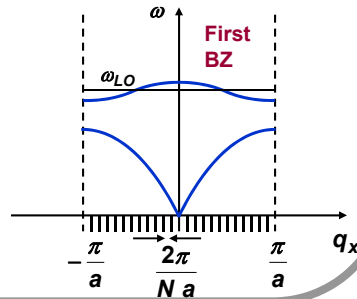
$$\sum_{q_x \text{ in FBZ}} = N$$

Therefore:

$$L \int_0^{\omega_{LO}} d\omega g_{1D}(\omega) = N \quad \Rightarrow \quad C = \frac{N}{L}$$

Finally:

$$g_{1D}(\omega) = \frac{N}{L} \delta(\omega - \omega_{LO})$$



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Optical Phonons in 2D and 3D: Einstein Model

Consider optical phonons in a N -primitive-cell 2D (or 3D) crystal of area A (or volume V)

For each optical phonon band we want to be able to write:

$$\sum_{\vec{q} \text{ in FBZ}} = A \int_0^{\omega_{LO}} d\omega g_{2D}(\omega) \quad \text{or} \quad \sum_{\vec{q} \text{ in FBZ}} = V \int_0^{\omega_{LO}} d\omega g_{3D}(\omega)$$

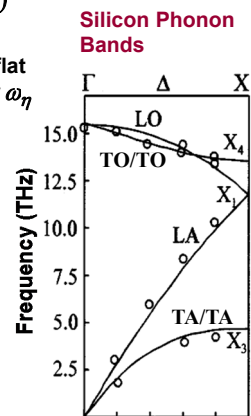
We suppose that each optical phonon band is completely flat and every phonon mode in a band has the same frequency ω_η

$$\omega = \omega_\eta \quad \{ \eta = 1, 2, 3 \text{ for LO, TO, TO}$$

$$g_{2D}(\omega) = \frac{N}{A} \delta(\omega - \omega_\eta) \quad \text{or} \quad g_{3D}(\omega) = \frac{N}{V} \delta(\omega - \omega_\eta)$$

One can check that the number of phonon modes per band is conserved:

$$A \int_0^{\omega_{LO}} d\omega g_{2D}(\omega) = N \quad \text{or} \quad V \int_0^{\omega_{LO}} d\omega g_{3D}(\omega) = N$$



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Heat Capacity of Optical Phonons in 2D: Einstein Model

Consider a material with two atoms per primitive cell in 2D
 ⇒ There must be 2 optical phonons bands in 2D (LO and TO)

Suppose the optical phonon frequencies are ω_{LO} and ω_{TO} . Assuming Einstein model:

$$g_{2D-LO}(\omega) = \frac{N}{A} \delta(\omega - \omega_{LO}) \quad g_{2D-TO}(\omega) = \frac{N}{A} \delta(\omega - \omega_{TO})$$

Total energy per unit area in both the optical phonon modes is:

$$\begin{aligned} u &= \frac{1}{A} \sum_{\vec{q} \text{ in FBZ}} \hbar \omega_{LO}(\vec{q}) \langle n_{\vec{q}} \rangle + \frac{1}{A} \sum_{\vec{q} \text{ in FBZ}} \hbar \omega_{TO}(\vec{q}) \langle n_{\vec{q}} \rangle \\ &= \int_0^{\infty} d\omega g_{2D-LO}(\omega) \frac{\hbar \omega}{e^{\hbar \omega / KT} - 1} + \int_0^{\infty} d\omega g_{2D-TO}(\omega) \frac{\hbar \omega}{e^{\hbar \omega / KT} - 1} \\ &= \frac{N}{A} \frac{\hbar \omega_{LO}}{e^{\hbar \omega_{LO} / KT} - 1} + \frac{N}{A} \frac{\hbar \omega_{TO}}{e^{\hbar \omega_{TO} / KT} - 1} \end{aligned}$$

The heat capacity is:

$$C = \frac{du}{dT} = \frac{N}{A} K \frac{e^{\hbar \omega_{LO} / KT}}{(e^{\hbar \omega_{LO} / KT} - 1)^2} \left(\frac{\hbar \omega_{LO}}{KT} \right)^2 + \frac{N}{A} K \frac{e^{\hbar \omega_{TO} / KT}}{(e^{\hbar \omega_{TO} / KT} - 1)^2} \left(\frac{\hbar \omega_{TO}}{KT} \right)^2$$

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Appendix: Classical Equipartition Theorem

According to the **canonical ensemble** of statistical physics, a system at temperature T will have energy E with the probability given by:

$$P(E) = \frac{1}{Z} e^{-\frac{E}{KT}}$$

The constant Z is determined by adding the probabilities for all possible states of the system and equating the result to unity

1D Example: Consider a free particle in 1D with the energy given by:

$$E = \frac{p_x^2}{2m}$$

The probability that the particle at temperature T will have momentum p_x is then:

$$P(p_x) = \frac{1}{Z} e^{-\frac{p_x^2 / 2m}{KT}}$$

We must have:

$$\int_{-\infty}^{\infty} dp_x P(p_x) = 1 \Rightarrow Z = \sqrt{2\pi mKT}$$

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Appendix: Classical Equipartition Theorem

So we finally have for the probability distribution of the particle momentum:

$$P(p_x) = \frac{1}{\sqrt{2\pi mKT}} e^{-\frac{p_x^2/2m}{KT}}$$

The average energy of the particle is then:

$$\int_{-\infty}^{\infty} dp_x \frac{p_x^2}{2m} P(p_x) = \frac{1}{2}KT$$

General Proof:

Consider a system whose total energy can be written in terms of various independent momenta and displacements as follows:

$$E = \sum_j a_j p_j^2 + \sum_j b_j u_j^2$$

The probability that the system will have some specific values for all the displacements and momenta is:

$$P(p_1, p_2, \dots, u_1, u_2, \dots) = \frac{1}{Z} e^{-\frac{\sum_j a_j p_j^2 + \sum_j b_j u_j^2}{KT}}$$

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Appendix: Classical Equipartition Theorem

$$P(p_1, p_2, \dots, u_1, u_2, \dots) = \frac{1}{Z} e^{-\frac{\sum_j a_j p_j^2 + \sum_j b_j u_j^2}{KT}}$$

The constant Z is determined by requiring:

$$\int_{-\infty}^{\infty} \prod_j (dp_j) \prod_r (du_r) P(p_1, p_2, \dots, u_1, u_2, \dots) = 1$$

It then follows that the average value of any one particular quadratic term in the expression for the total energy of the system is:

$$\langle a_n p_n^2 \rangle = \int_{-\infty}^{\infty} \prod_j (dp_j) \prod_r (du_r) a_n p_n^2 P(p_1, p_2, \dots, u_1, u_2, \dots) = \frac{1}{2}KT$$

$$\langle b_n u_n^2 \rangle = \int_{-\infty}^{\infty} \prod_j (dp_j) \prod_r (du_r) b_n u_n^2 P(p_1, p_2, \dots, u_1, u_2, \dots) = \frac{1}{2}KT$$

The above results follow from the properties of standard Gaussian integrals

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