Advanced kinetic theory: derivation of Boltzmann equation for u^3 anharmonic processes

Phonon distribution function $n_s(\vec{k}, \vec{r}, t)$

$$\frac{dn_s}{dt} = \frac{\partial n_s}{\partial t} + \frac{\partial n_s}{\partial \vec{r}} \cdot \underbrace{\dot{\vec{r}}}_{\frac{\partial \omega_s}{\partial \vec{k}}} = \underbrace{St\{n\}}_{\text{collision term}}$$

- local density of phonons

$$n(\vec{r}, t) = \sum_s \int \frac{d^3k}{(2\pi)^3} n_s(\vec{k}, \vec{r}, t)$$

- thermal energy density flow

$$\vec{J}(\vec{r}, t) = \sum_s \int \frac{d^3k}{(2\pi)^3} n_s(\vec{k}, \vec{r}, t) \cdot \frac{\partial \omega_s}{\partial \vec{k}} \cdot \hbar\omega_s(\vec{k})$$

etc. etc.

Need to specify collision term $St\{n\}$

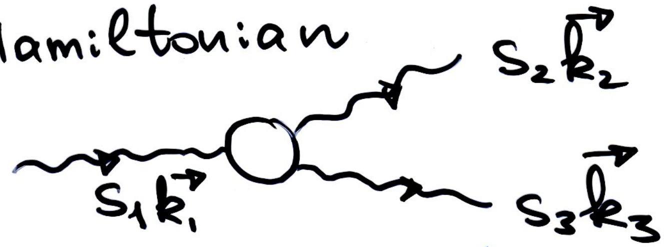
Three-phonon processes

$$H^{(3)} = \frac{1}{3!} \sum_{\vec{R}_1, \vec{R}_2, \vec{R}_3} D^{(3)}_{\mu_1\mu_2\mu_3}(\vec{R}_1 - \vec{R}_2, \vec{R}_1 - \vec{R}_3) \cdot u_{\mu_1}(\vec{R}_1)\, u_{\mu_2}(\vec{R}_2)\, u_{\mu_3}(\vec{R}_3)$$

Consider (just) 3 acoustic modes. Then

$$\vec{u}(\vec{R}) = \frac{1}{\sqrt{N}} \sum_{\vec{k}=1}^{N} \sum_{s=1}^{3} \sqrt{\frac{\hbar}{2M\omega_s(\vec{k})}} \left(a_{\vec{k}s} + a^{+}_{-\vec{k}s}\right) \cdot \vec{\epsilon}_s(\vec{k}) \exp(i\vec{k}\cdot\vec{R})$$

Derive the Hamiltonian describing



$$\boxed{\vec{k}_1 = \vec{k}_2 + \vec{k}_3 + \vec{K}}$$

$$\sim a^{+}_{s_3\vec{k}_3} a^{+}_{s_2\vec{k}_2} a_{s_1\vec{k}_1}$$

$$\text{Part of } H^{(3)} \doteq \Omega(\vec{k}_1, \vec{k}_2, \vec{k}_3) \cdot \frac{a^{+}_{s_3\vec{k}_3} a^{+}_{s_2\vec{k}_2} a_{s_1\vec{k}_1}}{\sqrt{N}\cdot\left(\omega_{s_1}(\vec{k}_1)\,\omega_{s_2}(\vec{k}_2)\,\omega_{s_3}(\vec{k}_3)\right)^{1/2}}$$

$$\Omega = \frac{1}{(2M)^{3/2}} \underbrace{D^{(3)}_{\mu_1\mu_2\mu_3}(\vec{k}_2, \vec{k}_3)}_{\text{Fourier-transform}} \epsilon_{s_1\mu_1}(\vec{k}_1)\, \epsilon_{s_2\mu_2}(\vec{k}_2)\, \epsilon_{s_3\mu_3}(\vec{k}_3)$$

$$\frac{1}{3!}\sum_{R_1R_2R_3} \hat{D}^{(3)}(R_1-R_2, R_1-R_3) \prod_{i=1}^{3} \left(\frac{\hbar}{2M\omega_{s_i}(k_i)}\right)^{1/2} \cdot \frac{1}{N^{3/2}} \cdot$$

$$\cdot\ \vec{\epsilon}_{s_1}(\vec{k}_1)\, \vec{\epsilon}_{s_2}(-\vec{k}_2)\, \vec{\epsilon}_{s_3}(-\vec{k}_3)$$

$$\cdot\ \exp\left(i k_1 \cdot R_1 - i k_2 \cdot R_2 - i k_3 \cdot R_3\right)$$

①. $\vec{\epsilon}_s(-\vec{k}) = \epsilon_s(\vec{k})$ $\quad \vec{k} \to -\vec{k}$

symmetry

② We should exploit crystal transl. symm.

$$\{\vec{R}_1, \vec{R}_2, \vec{R}_3\} \xrightarrow{\hat{A}} \{\vec{R}, \vec{R}', \vec{R}_0\}$$

new variables

$$\vec{R} = \vec{R}_1 - \vec{R}_2 \quad \vec{R}' = \vec{R}_1 - \vec{R}_3, \quad \vec{R}_0 = \vec{R}_1$$

$$\hat{A} = \begin{pmatrix} 1 & -1 & 0 \\ 1 & 0 & -1 \\ 1 & 0 & 0 \end{pmatrix} \qquad |\det \hat{A}| = 1$$

new variables are indeed independent

$$\sum_{R,R',R_0} \hat{D}^{(3)}(R,R')\cdot\exp\{ik_1\cdot R_0 - ik_2\cdot(R_0-R) - ik_3(R_0-R')\}$$

$$R = R_1 - R_2 = R_0 - R_2$$
$$R' = R_1 - R_3 = R_0 - R_3$$

$$\boxed{\begin{array}{ll} R_2 = R_0 - R & \\ R_3 = R_0 - R' & R_1 = R_0 \end{array}}$$

$$\Rightarrow \left[\sum_R \sum_{R'} \hat{D}^{(3)}(R,R') \exp(ik_2\cdot R + ik_3 R')\right] \times$$

$$\times \boxed{\sum_{R_0} \exp(i R_0 [k_1 - k_2 - k_3])}$$

$$\sum_{\vec{R}} e^{i\vec{k}\cdot\vec{R}} = \begin{cases} N, & \vec{k} \in RL \quad (e^{i\vec{k}\cdot\vec{R}} = 1 \ \forall \vec{R}) \\ \emptyset, & \vec{k} \notin RL \end{cases}$$

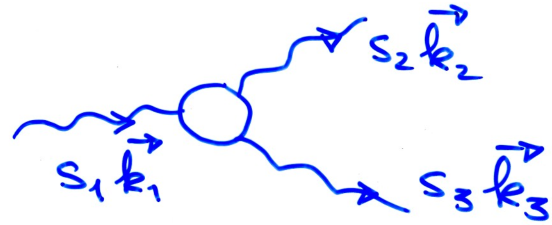
$$\Rightarrow \vec{k}_1 - \vec{k}_2 - \vec{k}_3 = \vec{K}$$

$$\boxed{\vec{k}_1 = \vec{k}_2 + \vec{k}_3 + \vec{K}}$$

conservation of quasi-momentum up to $\vec{K}$

Collision term

"out" contribution



Probability per unit time

$$dW_{out} = \frac{2\pi}{\hbar}\left| \langle n_{s_1}(\vec{k}_1)-1, n_{s_2}(\vec{k}_2)+1, n_{s_3}(\vec{k}_3)+1 | \right.$$

Golden rule

$$\left. H^{(3)} | n_{s_1}(\vec{k}_1), n_{s_2}(\vec{k}_2), n_{s_3}(\vec{k}_3)\rangle \right|^2$$

$$\cdot\, \delta(\hbar\omega_{s_1}(\vec{k}_1) - \hbar\omega_{s_2}(\vec{k}_2) - \hbar\omega_{s_3}(\vec{k}_3))$$

$$\cdot\, \boxed{\frac{d\vec{k}_2 V}{(2\pi)^3}}$$

density of final states

$$\langle n+1 | a^\dagger | n\rangle = \sqrt{n+1} \qquad \langle n-1 | a | n\rangle = \sqrt{n}$$

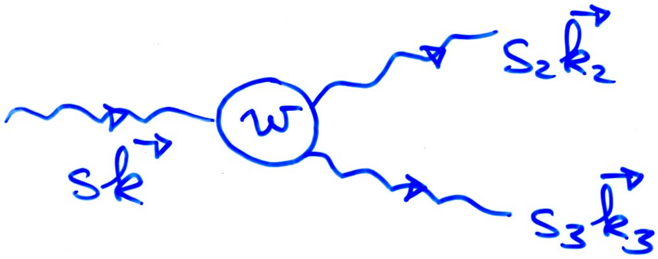
$$\boxed{dW_{out} = w \cdot N_1 (N_2+1)(N_3+1)\cdot \delta(\hbar\omega_1 - \hbar\omega_2 - \hbar\omega_3)\frac{d^3k_2}{(2\pi)^3}}$$

$$w = w(s_1\vec{k}_1; s_2\vec{k}_2, s_3\vec{k}_3) = \frac{2\pi v}{\omega_1\omega_2\omega_3}|\Omega|^2$$

$v = V/N$, primitive cell volume

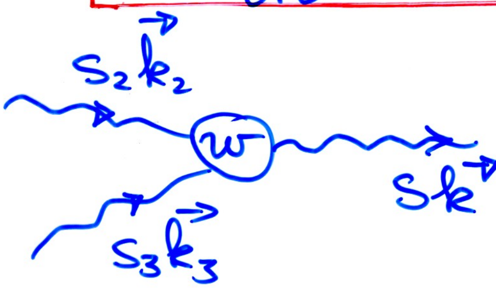
$N_i \equiv n_{s_i}(\vec{k}_i)$, $\omega_i = \omega_{s_i}(\vec{k})$

"In" and "out" contributions

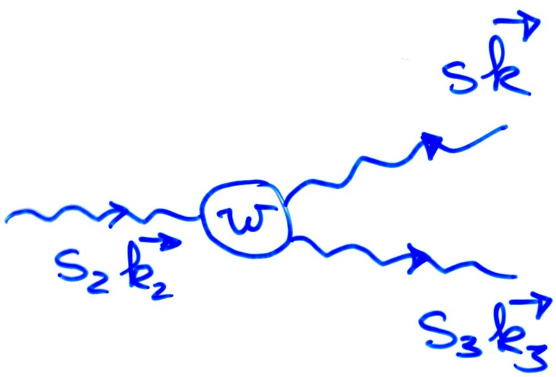


out

$$\boxed{(s\vec{k}) \underset{\text{in}}{\overset{\text{out}}{\rightleftarrows}} (s_2\vec{k}_2) + (s_3\vec{k}_3)}$$

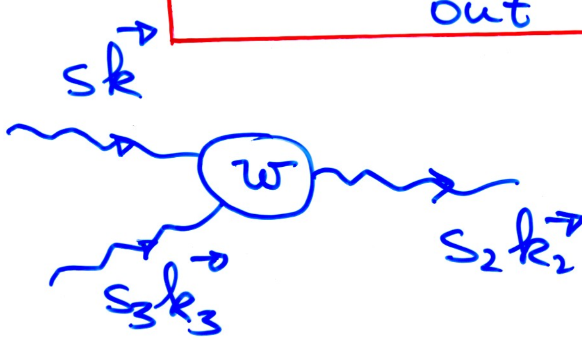


in



in

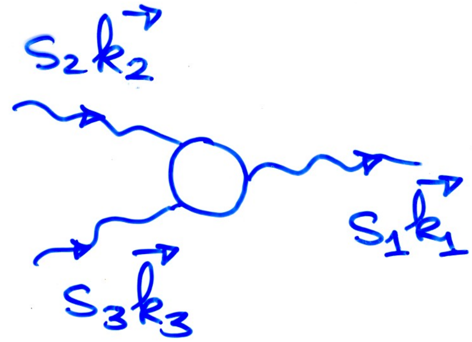
$$\boxed{(s_2\vec{k}_2) \underset{\text{out}}{\overset{\text{in}}{\rightleftarrows}} (s\vec{k}) + (s_3\vec{k}_3)}$$



out

Collision term

"in" contribution



$$\boxed{dW_{in} = w\cdot(N_1+1)N_2N_3\cdot\delta(\hbar\omega_1-\hbar\omega_2-\hbar\omega_3)\frac{d^3k_2}{(2\pi)^3}}$$

same as in dW_{out} // $t\to -t$ //

$$St\{n\} \quad \frac{dn_s(\vec{k};\vec{r};t)}{dt}$$

$$St\{n\} = \int\Big\{\frac{1}{2}\sum_{s_2 s_3} w(s\vec{k};s_2\vec{k}_2,s_3\vec{k}_3)\cdot\delta(\omega-\omega_2-\omega_3)$$
$$\times\left[(N+1)N_2N_3 - N(N_2+1)(N_3+1)\right]$$

$$\boxed{(s\vec{k})\rightleftarrows(s_2\vec{k}_2)+(s_3\vec{k}_3):}$$

$$+\sum_{s_2s_3} w(s_2\vec{k}_2;s\vec{k},s_3\vec{k}_3)\,\delta(\omega_2-\omega-\omega_3)$$
$$\times\left[N_2(N+1)(N_3+1)-(N_2+1)NN_3\right]\Big\}\frac{d^3k_2}{(2\pi)^3}$$

$$\boxed{(s_2\vec{k}_2)\rightleftarrows(s\vec{k})+(s_3\vec{k}_3):}$$

$\hbar = 1$

Boltzmann equation:

$$\frac{\partial n_s(\vec{k},\vec{r},t)}{\partial t} + \frac{\partial n_s}{\partial \vec{r}} \cdot \frac{\partial \omega_s}{\partial \vec{k}} = St\{n\}$$

Very important general properties:

① Equilibrium (Planck's) distribution is stationary:

$$N_s^0 = n_s^0(\vec{k}) = \frac{1}{\exp\left(\frac{\hbar\omega_s(\vec{k})}{k_B T}\right) - 1} \equiv N^0(\omega)$$

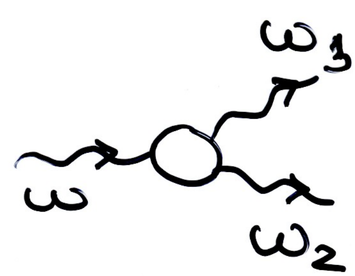
$$St\{N^0\} = 0 \Rightarrow \frac{\partial N^0}{\partial t} = 0 .$$

Indeed,

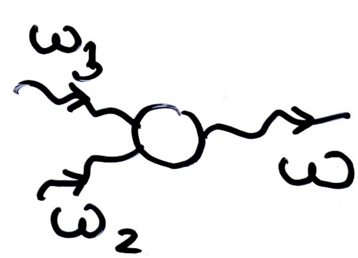
$$N^0(N_2^0+1)(N_3+1) = (N^0+1)N_2^0 N_3^0 \cdot e^{\frac{-\omega+\omega_2+\omega_3}{T}}$$

check!

But $\boxed{\omega = \omega_3 + \omega_2}$!



⇒ "In" and "Out" terms cancel



② Consider

$$\tilde{N}_s^\circ = \frac{1}{\exp\left(\frac{\hbar\omega_s(\vec{k}) - \hbar\vec{V}_0\cdot\vec{k}}{k_B T}\right) - 1}$$

$\vec{V}_0$ is some constant (velocity)

describes the drift of the whole phonon gas with velocity $\vec{V}_0$ relative to the lattice

$$\tilde{N}^\circ(\tilde{N}_2^\circ+1)(\tilde{N}_3^\circ+1) = (\tilde{N}^\circ+1)\tilde{N}_2^\circ\tilde{N}_3^\circ e^{\frac{-\omega+\omega_2+\omega_3}{T}} \times$$

$$\times \exp\left(\frac{(\vec{k}-\vec{k}_2-\vec{k}_3)\cdot\vec{V}_0}{T}\right)$$

If $\boxed{\vec{k} = \vec{k}_2+\vec{k}_3}$: only N processes

$St\{\tilde{N}^\circ\} = 0 \Rightarrow \tilde{N}^\circ$ stationary!

when crystal momentum conserved

thermal energy density current

$$\vec{J} = \sum_s \int \frac{d^3k}{(2\pi)^3} \tilde{N}_s^0(\vec{k}) \frac{\partial \omega_s(\vec{k})}{\partial \vec{k}} \cdot \hbar\omega_s(\vec{k})$$

$$\tilde{N}_s^0(\vec{k}) = \frac{1}{\exp\left(\frac{\hbar\omega_s(\vec{k}) - \hbar\vec{V}_0\cdot\vec{k}}{k_B T}\right) - 1}$$

1. $\underbrace{\omega_s(\vec{k}) = \omega_s(-\vec{k})}_{\text{even}}$ ($t \to -t$ symmetry!) not crystal symm.

2. $\frac{\partial \omega_s}{\partial \vec{k}}$ odd

3. $\tilde{N}_s^0(\vec{k})\Big|_{\vec{V}_0=0} = N_s^0(\vec{k})$ even

$$\vec{J}\Big|_{\vec{V}_0=0} = 0$$

$$\boxed{\vec{J}\Big|_{\vec{V}_0\neq 0} \neq 0}$$ and stationary

In the absence of U processes
$\kappa = \infty$ (R. Peierls 1929)

Linearized stationary Boltzmann equation for $T = T(\vec{r})$

$$N(\vec{k}, \vec{r}) = N^\circ(\vec{k}) + \delta N(\vec{k}, \vec{r})$$

small: $\delta N \ll N^\circ$

$$\frac{\partial N}{\partial \vec{r}} \cdot \underbrace{\frac{\partial \omega(\vec{k})}{\partial \vec{k}}}_{\equiv \vec{u}} = \frac{\partial N}{\partial T} \cdot (\vec{u} \cdot \vec{\nabla}) T = St\{N^\circ + \delta N\}$$

Linearize in δN:

$$\boxed{\frac{\partial N^\circ}{\partial T} (\vec{u} \cdot \vec{\nabla}) T = I(\delta N)}$$

keep in $St\{N^\circ + \delta N\}$ only terms $\sim \delta N$

linear integral equation for $\delta N(\vec{k}, \vec{r})$

To solve it, we need to exploit properties of $w = w(\vec{k}_1, \vec{k}_2 \vec{k}_3)$

. . .

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