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Charge carrier mobility, quality factor and thermoelectric efficiency

Andrei Novitskii, Academic Research Center for Energy Efficiency, NUST MISIS
Email: novitskiy@misis.ru
@anovitzkij

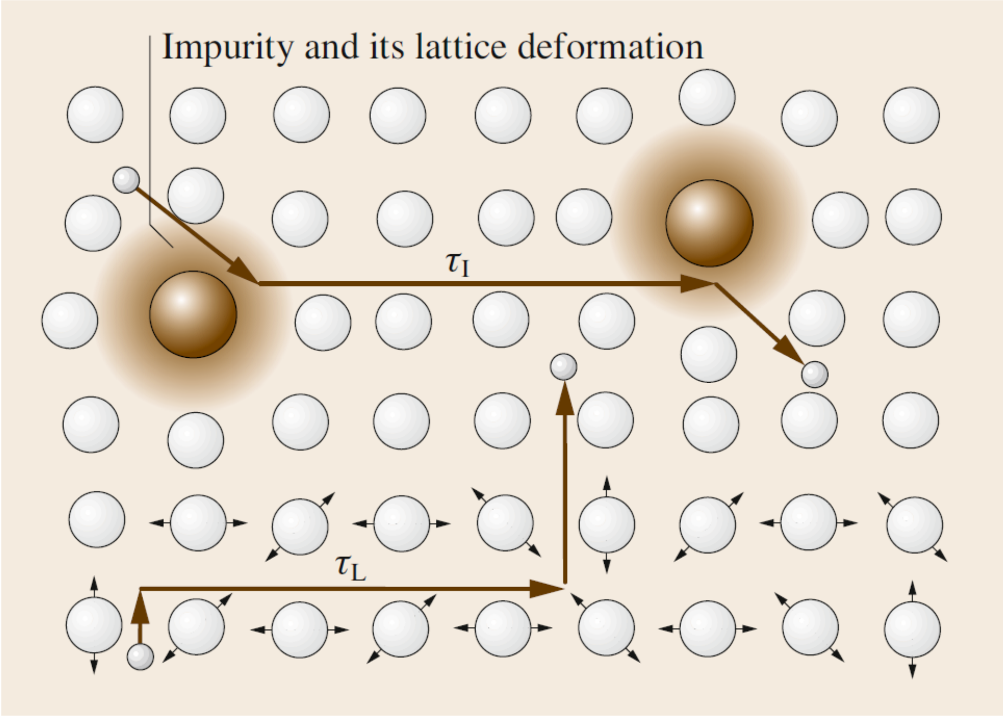
Charge carrier (drift) mobility

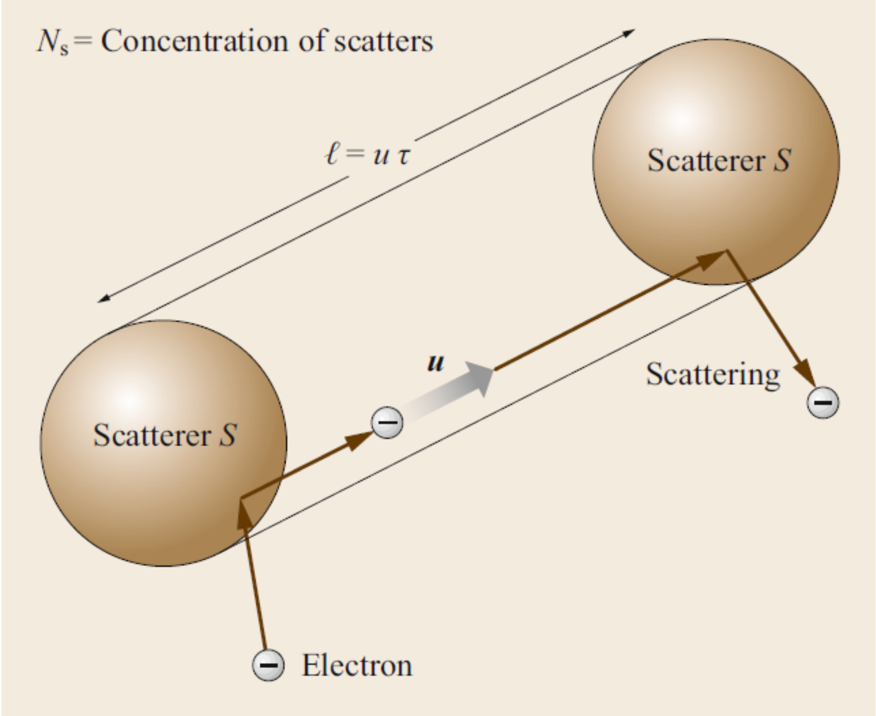
Let's remember what is the carrier mobility?[1]

$$v_d = \frac{e\tau}{m_I^*}E = \mu_d E \text{ and } \sigma = en\mu_d$$

Why is that so important?

scattering





Charge carrier mobility

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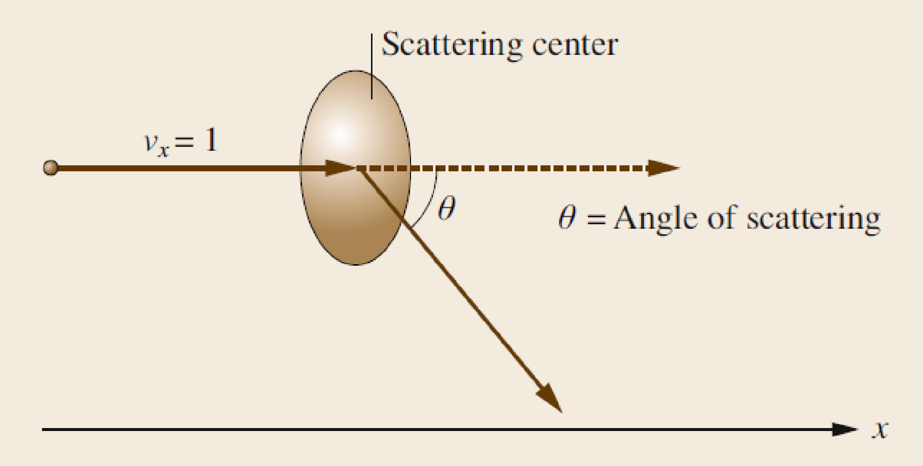
In materials with multiple scattering mechanisms affecting the carrier mean free path, the temperature dependence of charge carrier mobility is complex and total μ can be evaluated by Matthiessen's rule,[2,3] which assumes scattering channels are independent of each other

$$\mu^{-1} = \sum_i \mu_i^{-1}$$

where μ_i represents the mobility of a specified scattering mechanism.
The dominant scattering mechanism can be determined using log-log plot from the temperature dependence of the Hall mobility fitted by a power-law $\mu \propto T^m$, where m is the scattering factor of the carriers.



Electrical conductivity

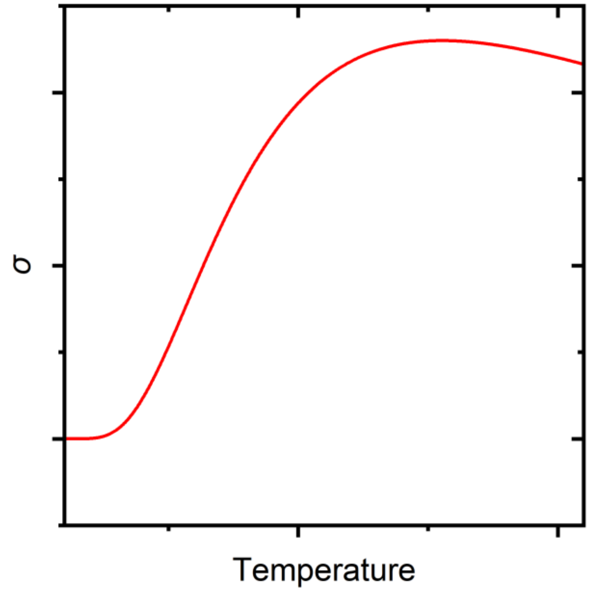
$$\sigma(T) = en(T)\mu(T)$$

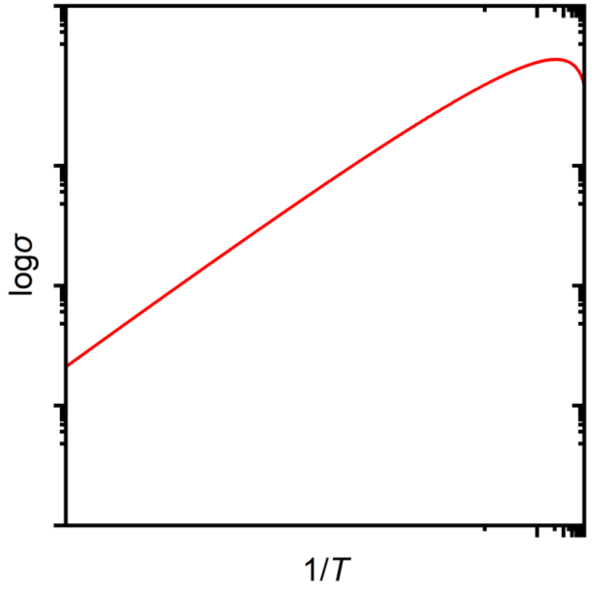
For semiconductors:

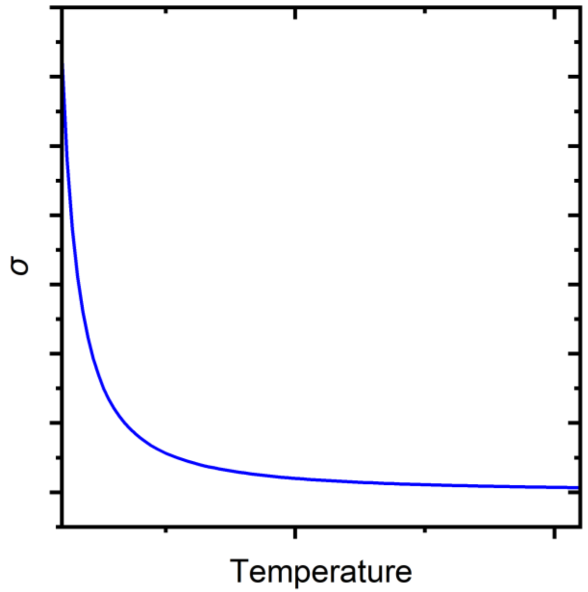
$$n(T) = N_c e^{-\frac{E_C - F}{k_B T}},\ \mu(T) \propto T^m$$

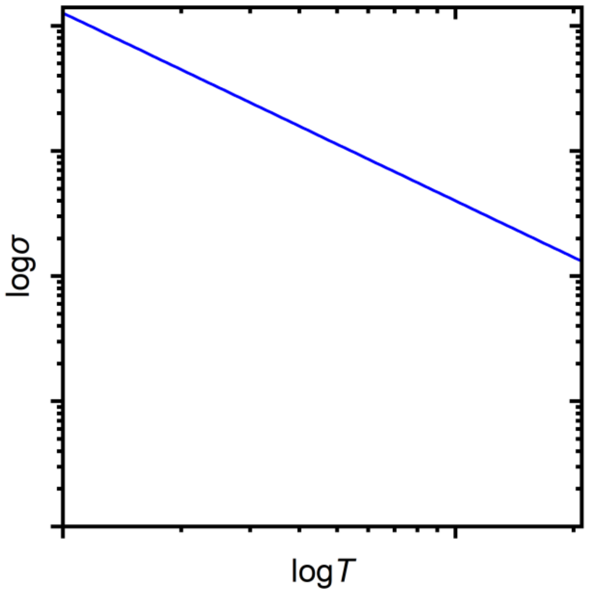
For metals:

$$n = \frac{1}{3\pi^2}\left(\frac{2m_0 E}{\hbar^2}\right)^{3/2},\ \mu(T) \propto T^m$$









Charge carrier mobility

For a single parabolic band semiconductor, the Hall mobility can be represented by

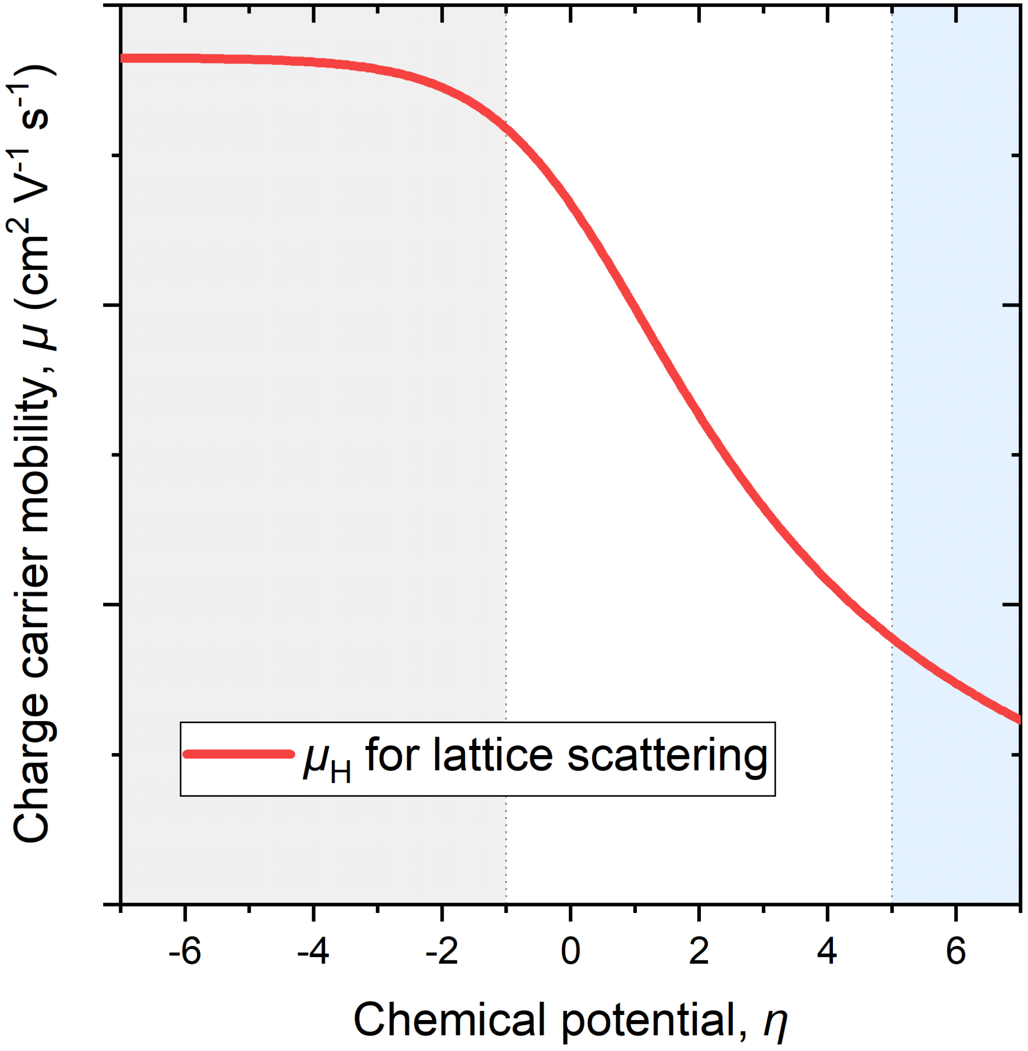
$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

$$F_j(\eta) = \int_0^{\infty} \frac{\varepsilon^j}{1 + e^{\varepsilon - \eta}} d\varepsilon$$

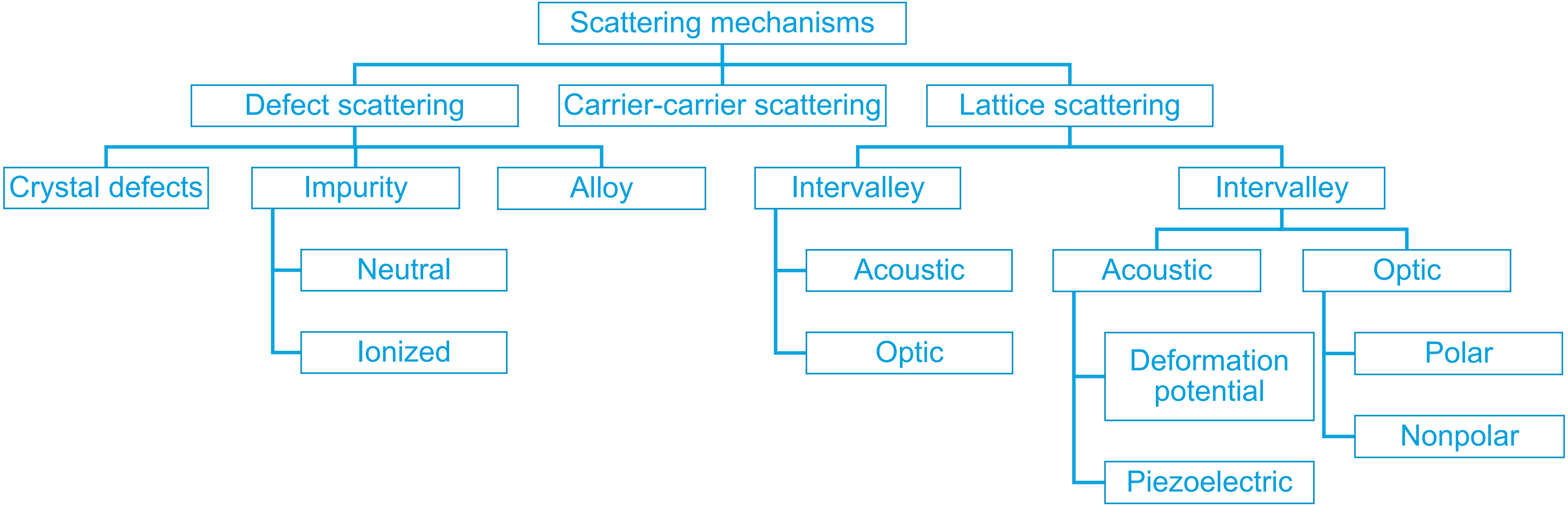
here μ_0 is typically called as the carrier concentration independent mobility parameter or free mobility parameter. μ_0 value can be obtained through the analysis of the Hall mobility data.

Degenerate limit: $\mu_H \propto \frac{\mu_0}{(m^*)^2 T n^{1/3}}$

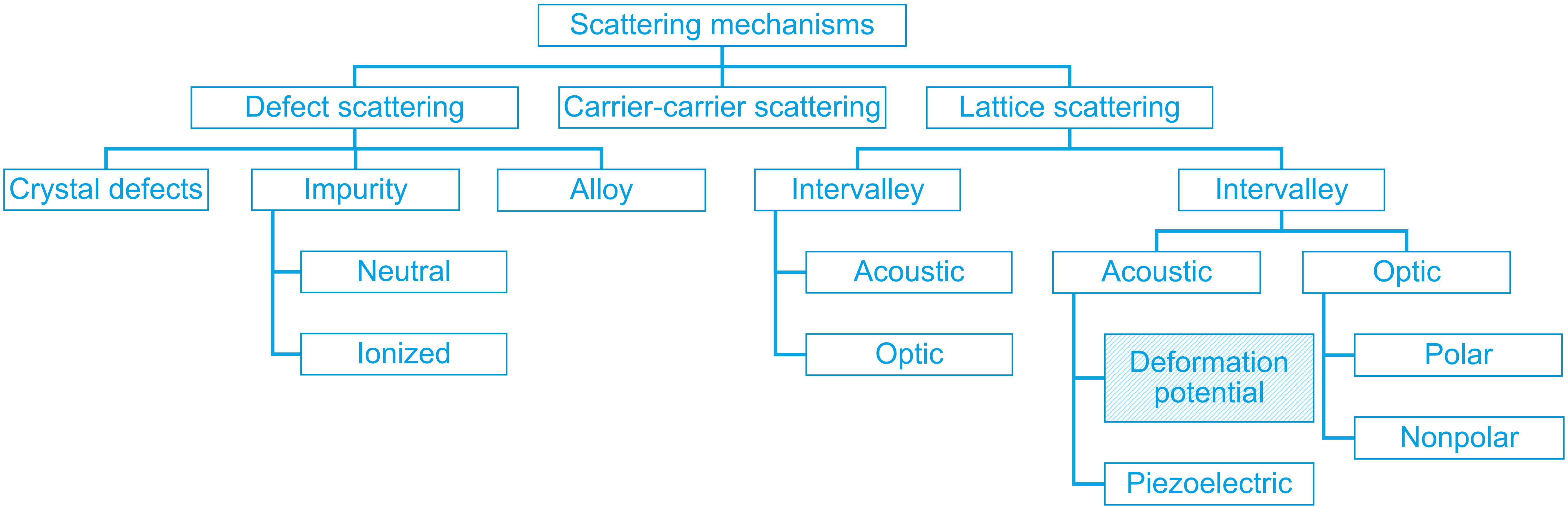
Non-degenerate limit: $\mu_H = \frac{\sqrt{\pi}}{2} \mu_0$



Scattering mechanisms



Scattering mechanisms



Acoustic phonon scattering

$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

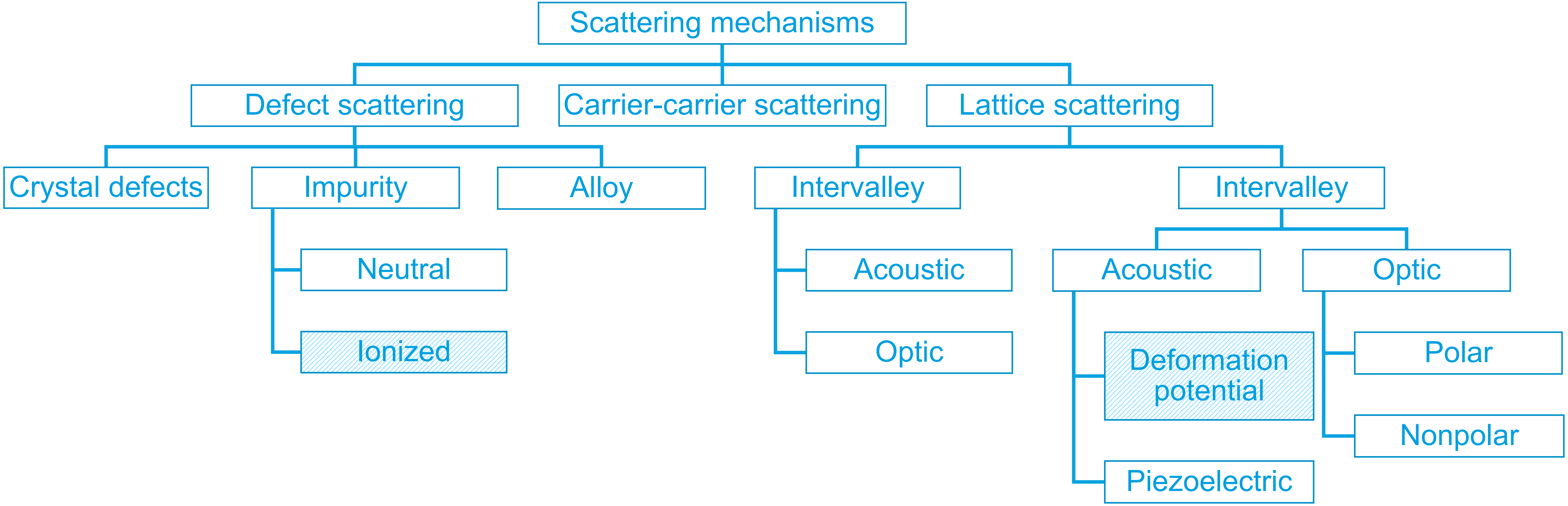
For acoustic phonon scattering:

$$\mu_0 = \frac{e\pi\hbar^4 C_{ll}}{\sqrt{2}(k_B T)^{3/2}(m_b^*)^{3/2} m_I^* \Delta_{def}^2}$$

where $\hbar$ is the reduced Planck's constant, C_{ll} is the elastic constant for longitudinal vibrations ($C_{ll} = dv_l^2$, where d is the density, v_l is the longitudinal component of sound velocity), m_b^* is the effective mass of a single valley, m_I^* is the inertial effective mass (for the isotropic spherical case $m_b^* = m_I^*$), and Δ_{def} is the deformation potential characterizing the carrier-phonon interaction.[2,4]

It should be mentioned, that C_{ll}, m^* and Δ_{def} can also depend on temperature.

Scattering mechanisms



Ionized impurity scattering

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$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

For ionized impurity scattering:

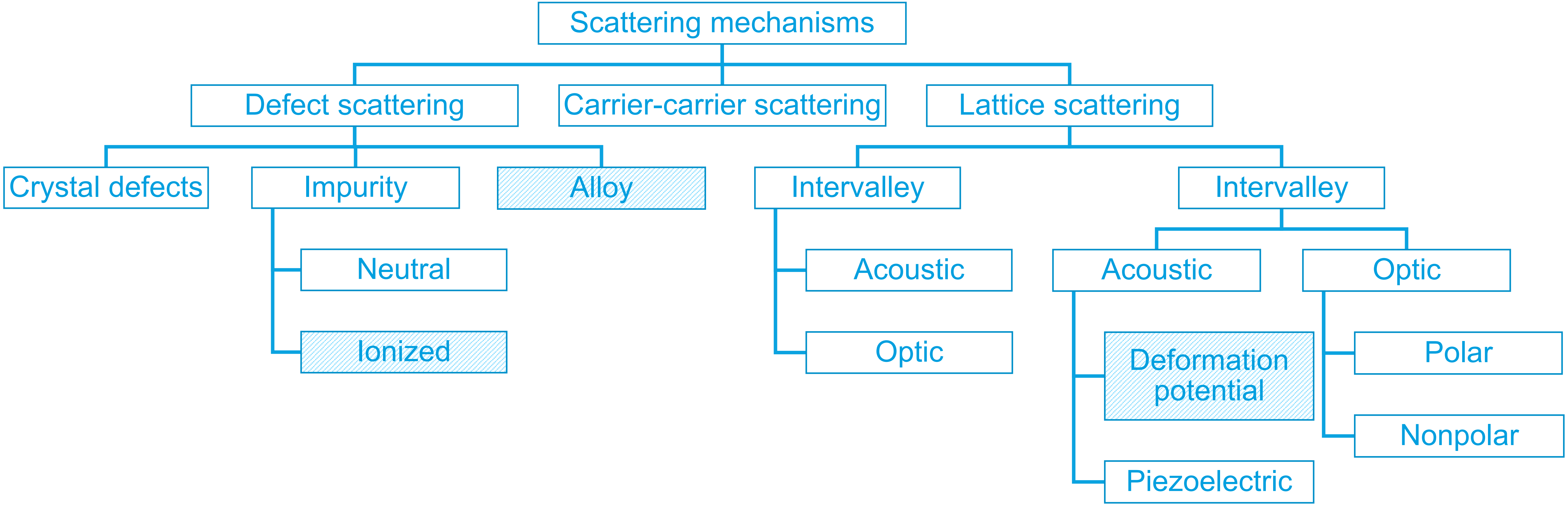
$$\mu_0 = \frac{8\sqrt{2}k_B^{3/2}}{\pi^{3/2}e^3} \frac{(\varepsilon\varepsilon_0)^2}{N_I (m_d^*)^{1/2} Z^2} \frac{T^{3/2}}{\ln\left[1 + \left(\frac{3\varepsilon\varepsilon_0 k_B T}{N_I^{1/3} Ze}\right)^2\right]}$$

This expression for the mobility encompasses the Conwell-Weisskopf and Brooks-Herring.[2,4]

It is sufficient when

$$T^2 \ll \frac{N_I^{2/3} Z^2 e^4}{9(\varepsilon\varepsilon_0)^2 k_B^2}$$

Scattering mechanisms



Alloy disorder scattering

$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

For alloy disorder scattering:

$$\mu_0 = \frac{16e\hbar^4}{9\sqrt{2}z(1-z)(k_B T)^{1/2}} \frac{N_0}{(m_d^*)^{5/2} U^2}$$

where z is fractional concentration of the solid solution and N_0 is the number of atom per unit volume. The potential energy fluctuation caused by alloy disorder is characterized by U, which is analogous the deformation potential Δ_{def}.[2,5]

Scattering mechanisms

- Scattering mechanisms
  - Defect scattering
    - Crystal defects
    - Impurity
      - Neutral
      - Ionized
    - Alloy
  - Carrier-carrier scattering
  - Lattice scattering
    - Intervalley
      - Acoustic
      - Optic
    - Intervalley
      - Acoustic
        - Deformation potential
        - Piezoelectric
      - Optic
        - Polar
        - Nonpolar

Piezoelectric potential scattering

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$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

For piezoelectric potential scattering:

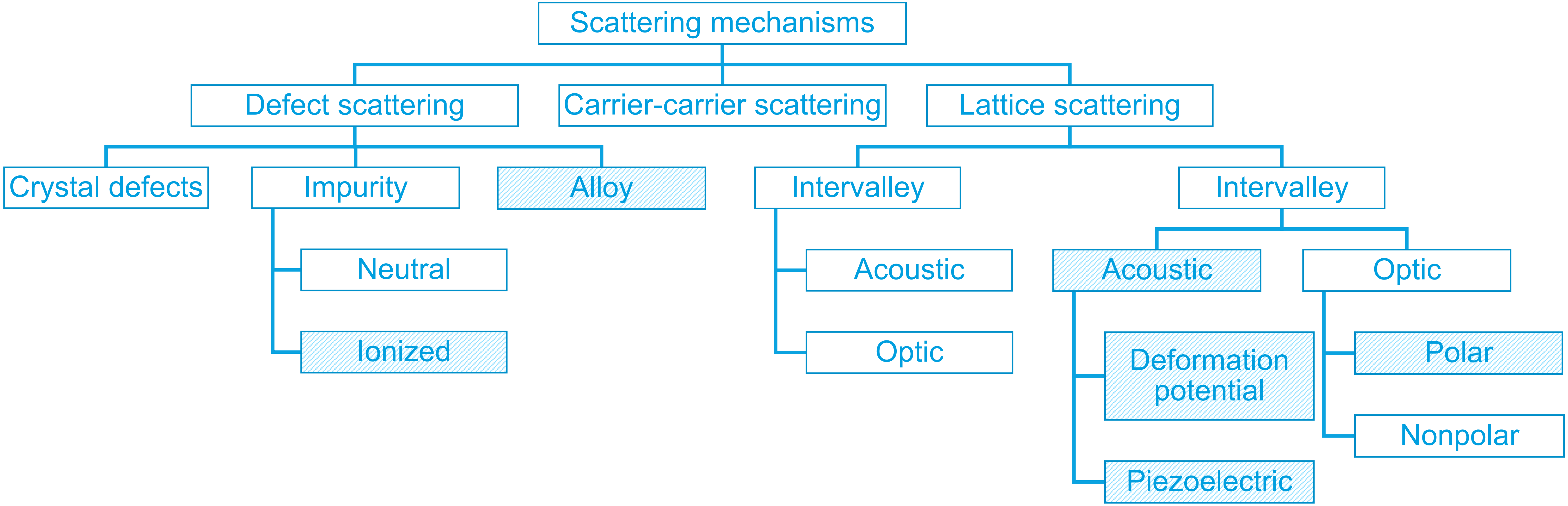
$$\mu_0 = \frac{16\sqrt{2\pi}}{3} \frac{\hbar \varepsilon \varepsilon_0}{(m^*)^{3/2} e K^2} (k_B T)^{-1/2}$$

with

$$K = \frac{e_p^2 / C_{ll}}{\varepsilon \varepsilon_0 + e_p^2 / C_{ll}}$$

e_p being the piezoelectric coefficient. In strongly ionic crystals, e.g. II–VI semiconductors, the piezoelectric scattering can be stronger than the deformation potential scattering.[2]

Scattering mechanisms



Polar optical scattering

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$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

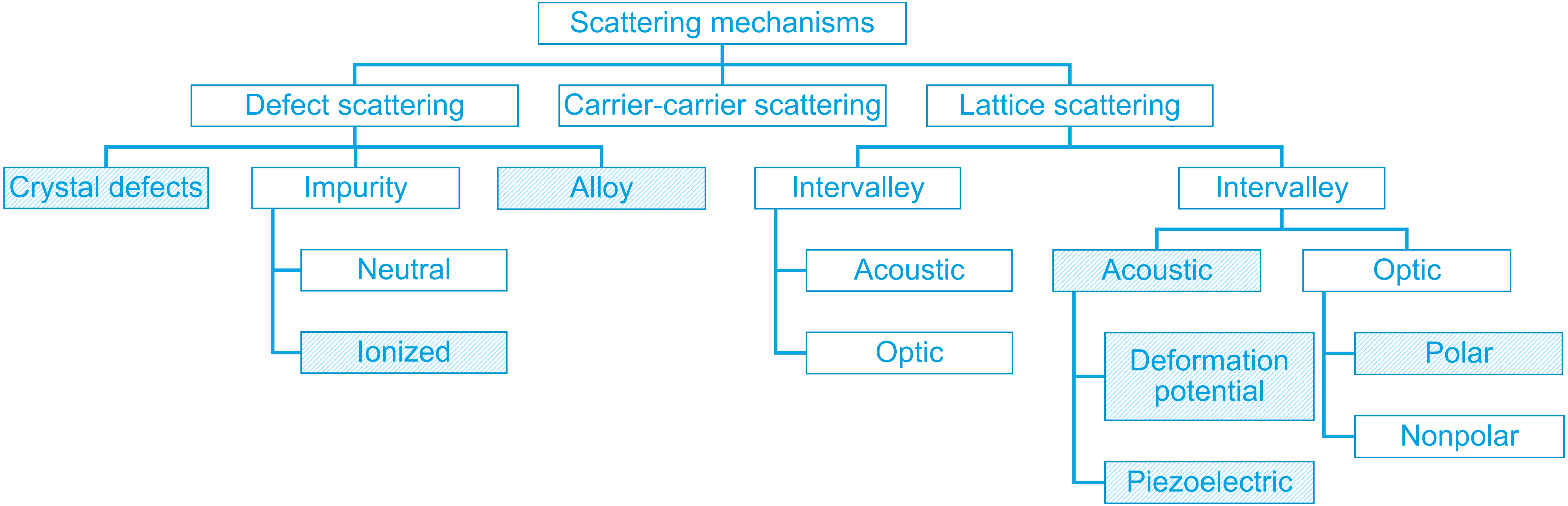
For polar optical scattering (where $T \ll \theta_D$):

$$\mu_0 = \frac{e}{2m^* \alpha_p \omega_0} e^{\theta_D/T}$$

where in the scattering mechanism the absorbed or emitted phonon energy $\hbar\omega_0$ is comparable to the thermal energy of the carriers,[2] α_p is the dimensionless polar constant:

$$\alpha_p = \frac{1}{137}\sqrt{\frac{m^* c^2}{2k_B\theta_D}}\left(\frac{1}{\varepsilon(\infty)} - \frac{1}{\varepsilon(0)}\right)$$

Scattering mechanisms



Dislocation scattering

$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

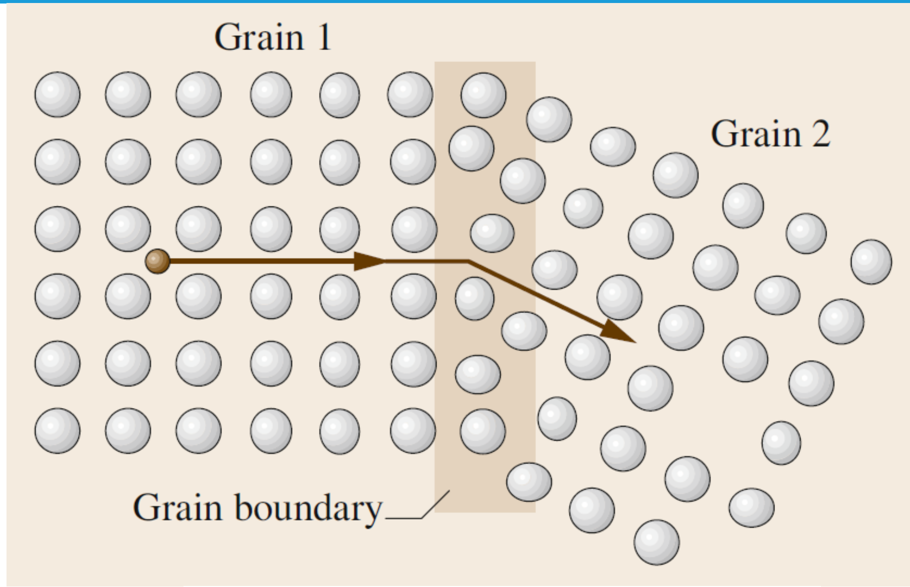
For dislocation scattering (for *n*-type semiconductor):

$$\mu_0 = \frac{30\sqrt{2\pi}(\varepsilon\varepsilon_0)^2 d^2}{N_{disl} e^3 f^2 L_D (m^*)^{1/2}} (k_B T)^{3/2}$$

d being the average distance of acceptor centers along the dislocation line, f their occupation rate, N_{disl} the area density of dislocations and $L_D = \sqrt{\varepsilon k_B T/(e^2 n)}$ the Debye screening length. Dislocations can contain charge centers and thus act as scattering centers. The deformation has introduced acceptor-type defects reducing the mobility in particular at low temperatures (similar to ionized impurity scattering).[2]

Grain boundaries scattering

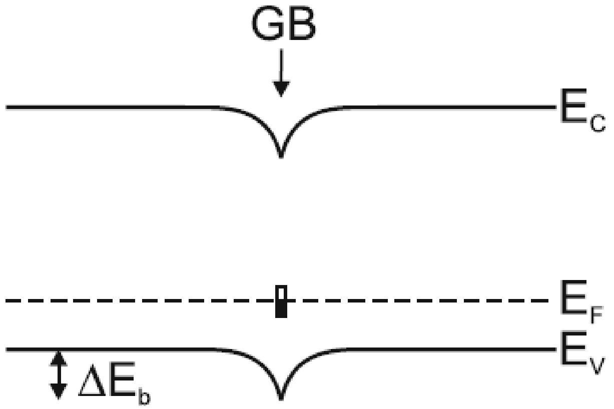
$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$



For grain boundaries scattering:

$$\mu_0 = \frac{eL_G}{\sqrt{8m^*\pi k_B}} T^{-1/2} e^{-\Delta E_b/(k_B T)}$$

where L_G is the grain size. Grain boundaries contain electronic traps whose filling depends on the doping of the bulk of the grains. Charges will be trapped in the grain boundaries and a depletion layer will be created.[2,4]

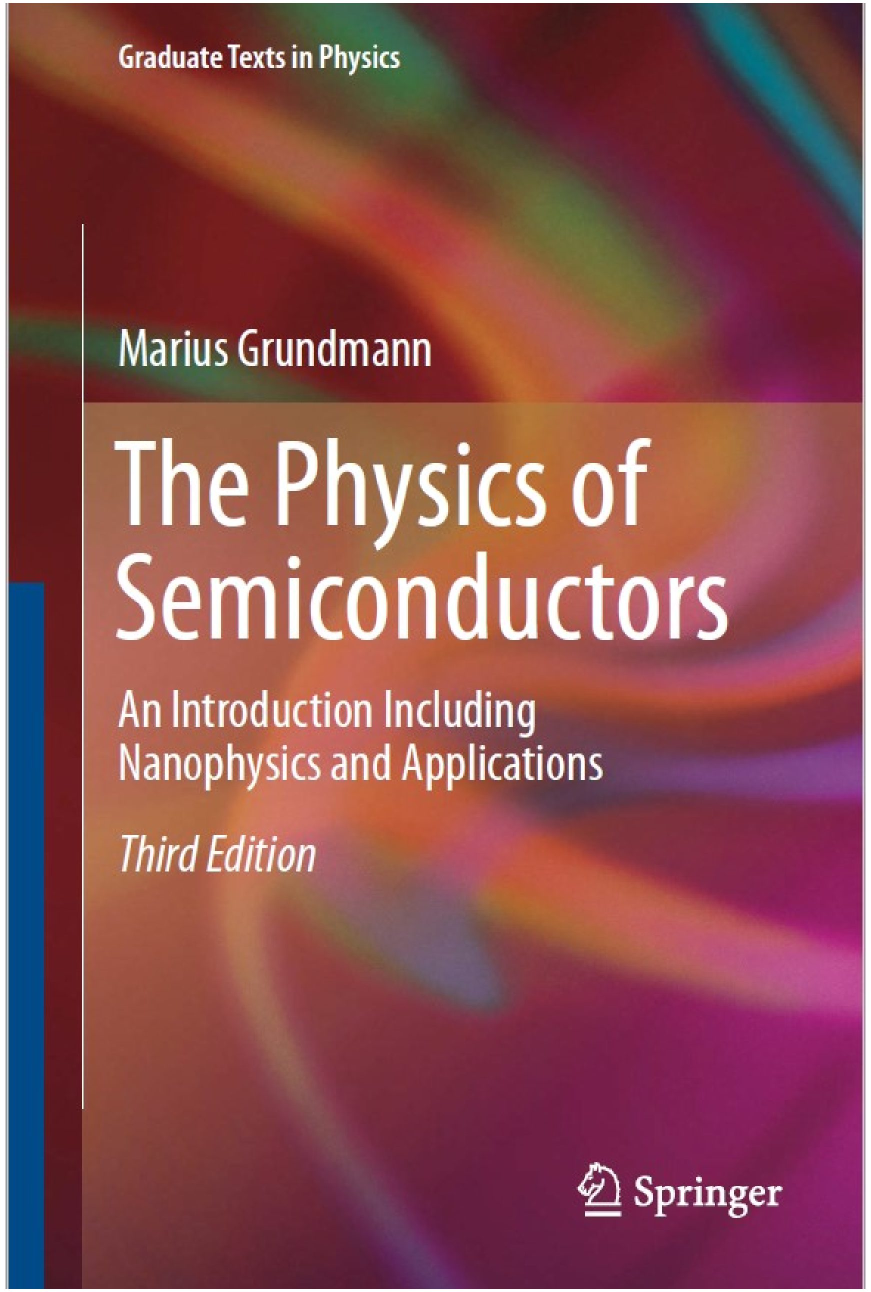


At low doping, the grains are fully depleted and all free carriers are trapped in the grain boundaries. This means low conductivity, however, no electronic barrier to transport exists. At intermediate doping, traps are partially filled and the partial depletion of the grain leads to the creation of an electronic barrier ΔE_b hindering transport since it must be overcome *via* thermionic emission. At high doping the traps are completely filled and the barrier vanishes again.[4]

Scattering mechanisms

- Scattering mechanisms
  - Defect scattering
    - Crystal defects
      - $m_{GB} = -1/2$
      - $m_{disl} = 3/2$
    - Impurity
      - Neutral: $m = 0$
      - Ionized: $m = 3/2$
    - Alloy: $m = -1/2$
  - Carrier-carrier scattering
  - Lattice scattering
    - Intervalley
      - Acoustic
      - Optic
    - Intervalley
      - Acoustic
        - Deformation potential: $m = -3/2$
        - Piezoelectric: $m = -1/2$
      - Optic
        - Polar: $m = 1/2$
        - Nonpolar

Further reading

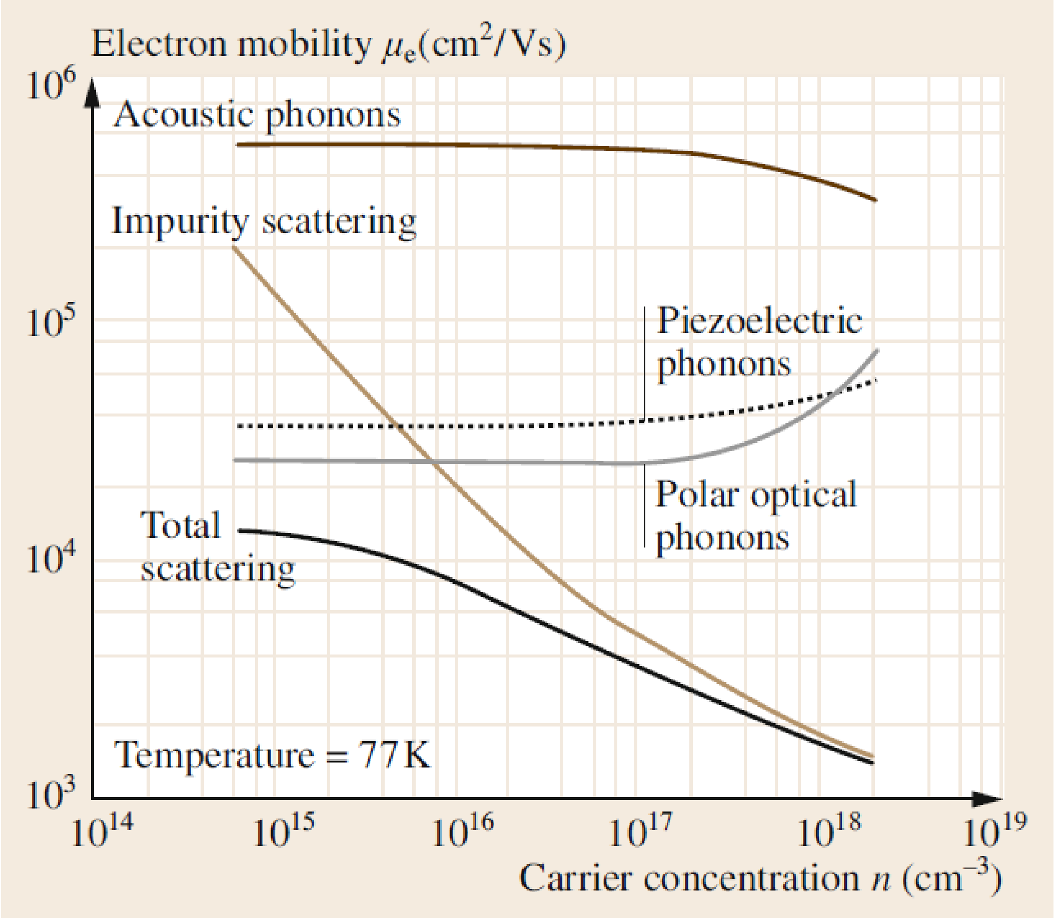


Charge carrier mobility

$$\mu_0^{-1} = \sum_i \mu_{0,i}^{-1} = \frac{1}{\mu_{0,ph}} + \frac{1}{\mu_{0,ion}} + \frac{1}{\mu_{0,gb}} + \cdots$$

$$\mu_H(\eta) = \mu_0 \frac{\left(\frac{3}{2} + 2r\right) F_{2r+1/2}(\eta)}{\left(\frac{3}{2} + r\right) F_{r+1/2}(\eta)}$$

$$n(\eta) = 4\pi \left(\frac{2m_d^* k_B T}{h^2}\right)^{3/2} F_{1/2}(\eta)$$



Charge carrier mobility

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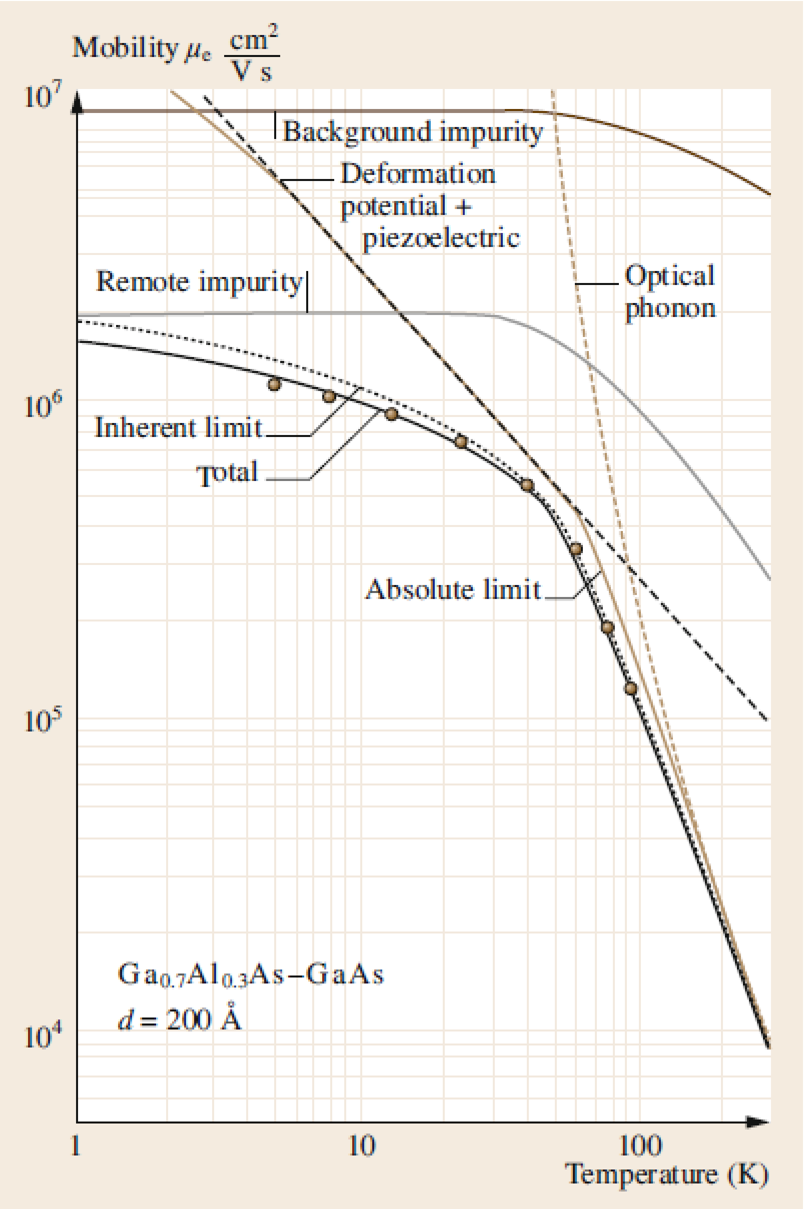

Electrical conductivity

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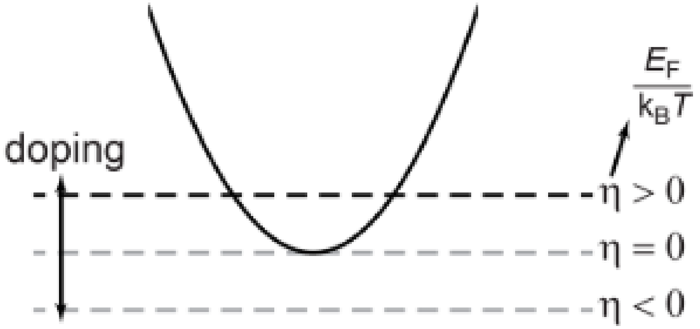
Now it is possible to calculate the electrical conductivity (don't forget that $\mu(\eta) = \mu_H(\eta)/r_H(\eta)$):

$$\sigma(\eta) = en(\eta)\mu(\eta) = \frac{8\pi e(2m_d^* k_B T)^{3/2}}{3h^3}\mu_0(r + \frac{3}{2})F_{r+1/2}(\eta)$$

here σ_{E_0} is the magnitude of conductivity for a given η (describes the conductive "quality" of charge carriers in the material) called transport parameter:

$$\sigma_{E_0} = \frac{8\pi e(2m_e k_B T)^{3/2}}{3h^3}\mu_0\left(\frac{m_d^*}{m_e}\right)^{3/2} = \frac{8\pi e(2m_e k_B T)^{3/2}}{3h^3}\mu_w$$

$\mu_w = \mu_0\left(\frac{m_d^*}{m_e}\right)^{3/2}$ is the weighted mobility.



For acoustic phonon scattering:

$$\sigma(\eta) = \sigma_{E_0}\ln(1 + e^{\eta})$$

Reminder: $\mu_0 = e\tau_0/m_I^*$ and $m_d^* \approx m_S^*$ for acoustic phonon scattering.

Electrical conductivity

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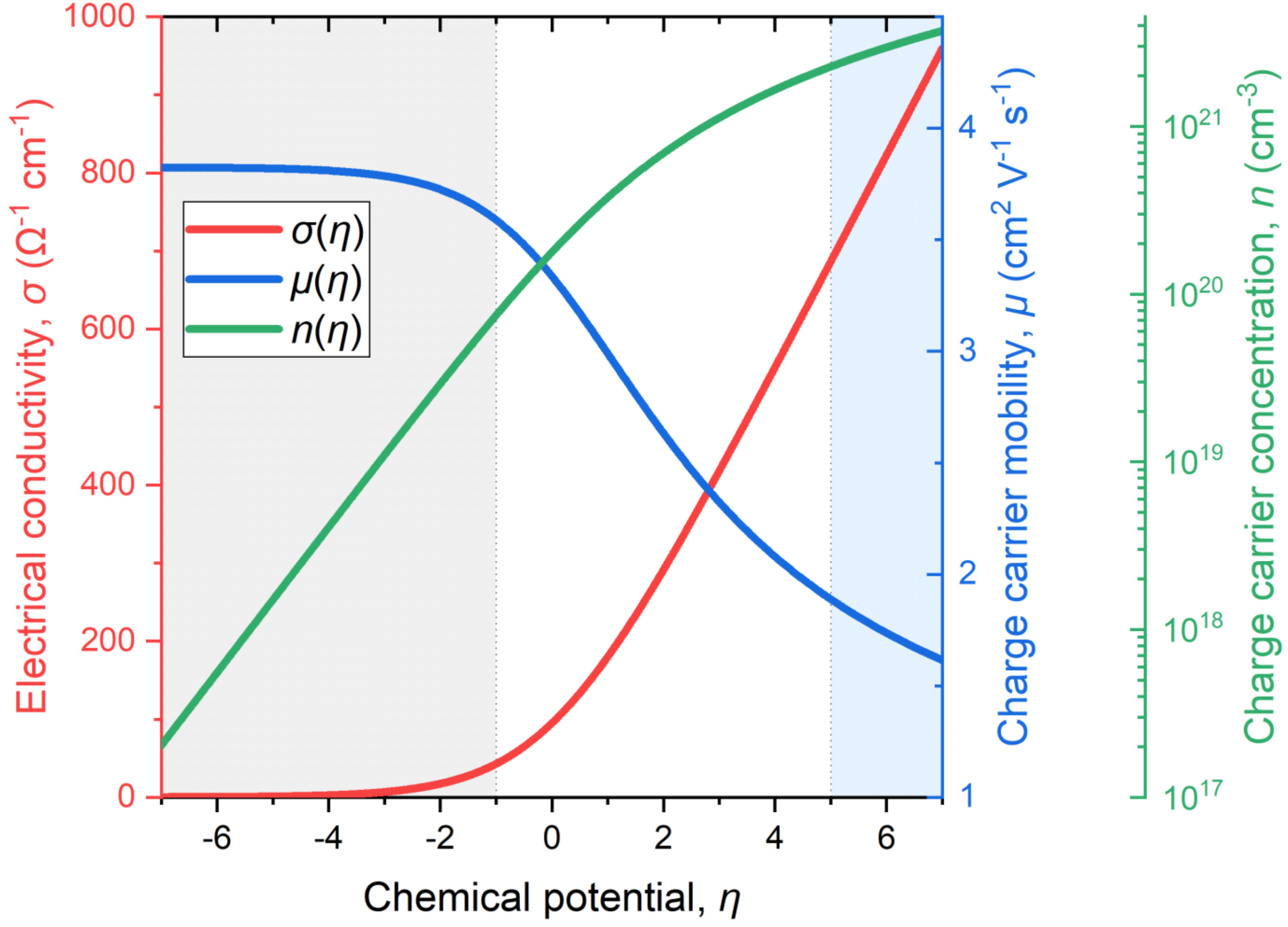
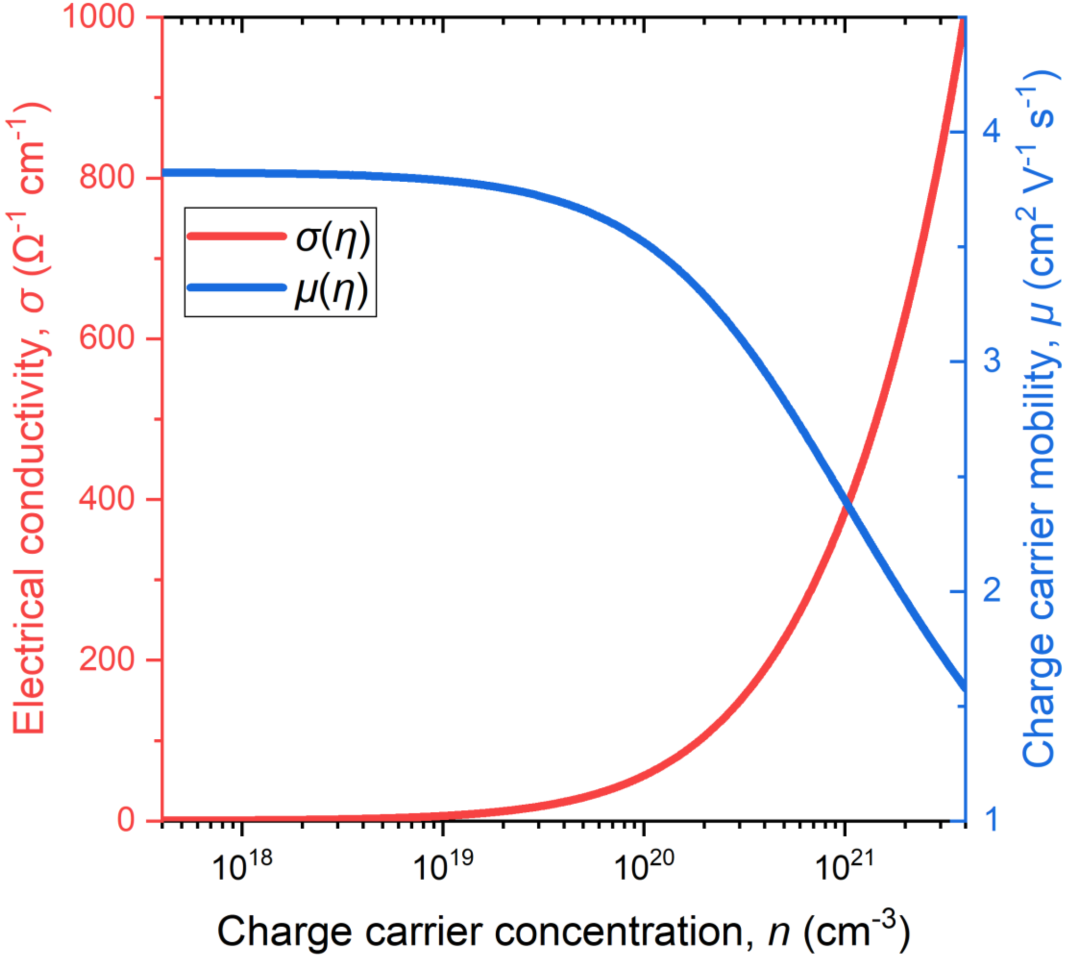
$$\sigma(\eta) = \sigma_{E_0}\ln(1+e^{\eta}),\ \sigma_{E_0} = \frac{8\pi e(2m_e k_B T)^{3/2}}{3h^3}\mu_0\left(\frac{m_d^*}{m_e}\right)^{3/2},\ \mu_H(\eta) = \mu_0\frac{\left(\frac{3}{2}+2r\right)F_{2r+1/2}(\eta)}{\left(\frac{3}{2}+r\right)F_{r+1/2}(\eta)},\ \mu_0 = \frac{e\pi\hbar^4 C_{ll}}{\sqrt{2}(k_B T)^{3/2}(m_b^*)^{3/2} m_I^* \Delta_{def}^2}$$





Thermoelectric efficiency

Using calculated m^*, μ_0, and κ_l it is possible to calculate a ‘theoretical’ zT to estimate the optimum carrier density at a particular temperature (from the plot of the zT versus n):

$$zT = \frac{\alpha^2 \sigma T}{\kappa_l + \kappa_e} = \frac{\alpha^2}{\frac{\kappa_l}{\sigma T} + L}$$

Considering all that we know:

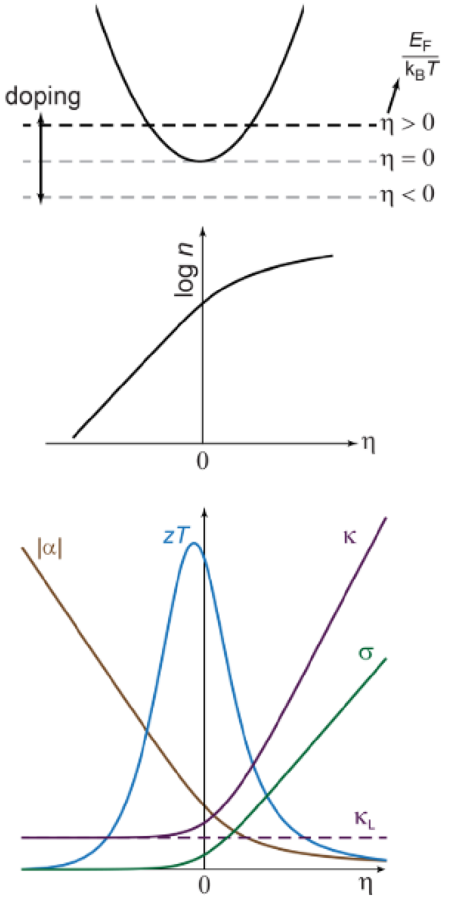
$$zT(\eta) = \frac{\alpha^2(\eta)}{\frac{\kappa_l}{T\sigma_{E_0} \ln(1+e^{\eta})} + L(\eta)} = \frac{\alpha^2(\eta)}{(\psi(\eta)\beta)^{-1} + L(\eta)}$$

here

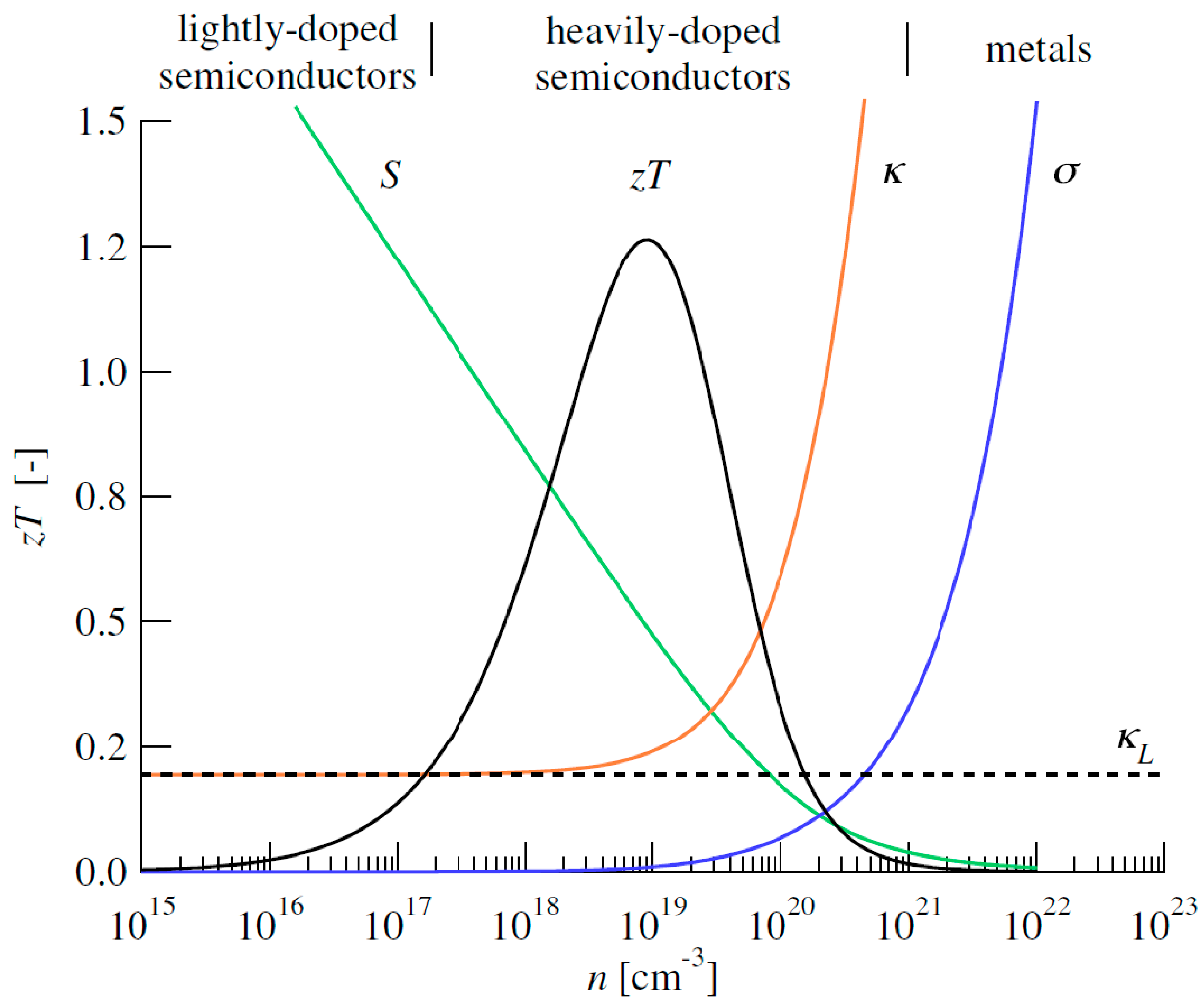
$$\psi(\eta) = \frac{8\pi e}{3}\left(\frac{2m_e k_B}{h^2}\right)^{3/2} F_{r+1/2}(\eta)$$

and

$$\beta = \frac{\mu_0 \left(m^*/m_e\right)^{3/2} T^{5/2}}{k_l}$$



Summary


lightly-doped semiconductors
heavily-doped semiconductors
metals
S
zT
κ
σ
κ_L
zT [-]
1.5
1.2
1.0
0.8
0.5
0.2
0.0
10^15
10^16
10^17
10^18
10^19
10^20
10^21
10^22
10^23
n [cm^-3]

Quality factor

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Again:

$$zT(\eta) = \frac{\alpha^2(\eta)}{\frac{\kappa_l}{T\sigma_{E_0}\ln(1+e^\eta)} + L(\eta)} = \frac{\alpha^2(\eta)}{\frac{(k_B/e)^2}{B\ln(1+e^\eta)} + L(\eta)}$$

$B = \left(\frac{k_B}{e}\right)^2 \frac{\sigma_{E0}}{\kappa_l} T = \left(\frac{k_B}{e}\right)^2 \frac{8\pi e(2m_e k_B)^{3/2}}{3h^3} \frac{\mu_w}{\kappa_l} T^{5/2}$ is the quality factor.

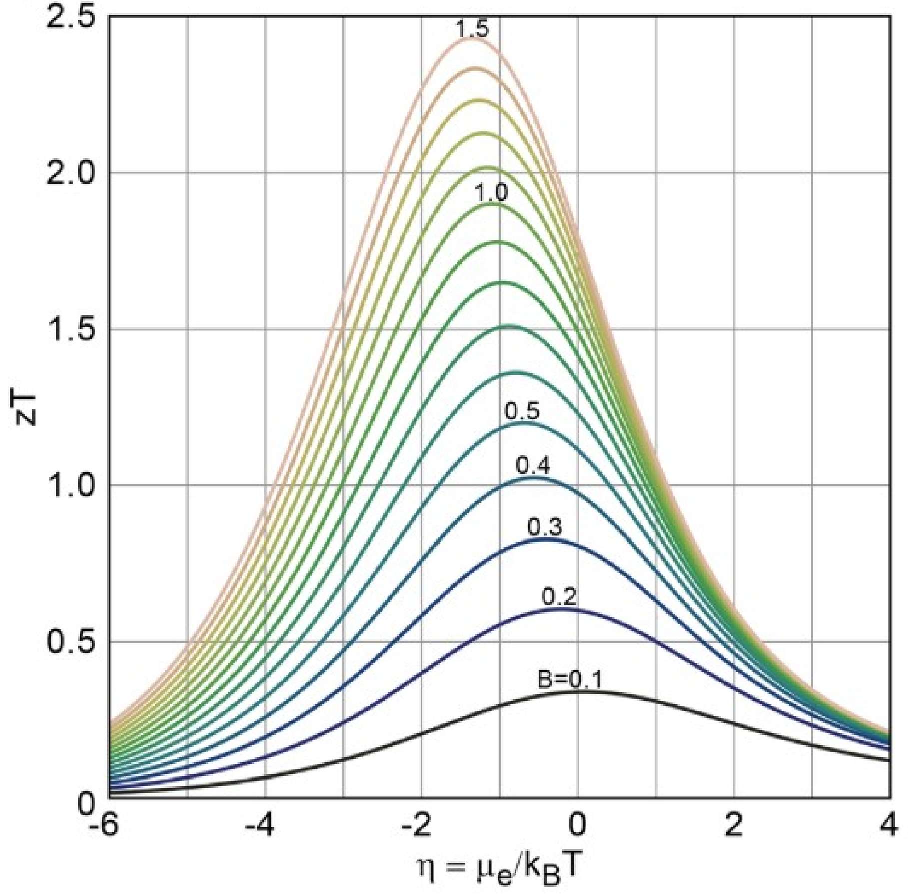
For example, for acoustic phonon scattering $\tau_0 \propto 1/(m_b^*)^{3/2}$

and considering that $m_d^* = N_v^{2/3} m_b^*$, $\mu_0 = \frac{e\tau_0}{m_I^*}$

the quality factor $B \propto N_v/(m_I^* \kappa_l)$

💡 B is materials parameter that might depend on temperature but not doping.

For more details see Ref. [6].



Electronic quality factor

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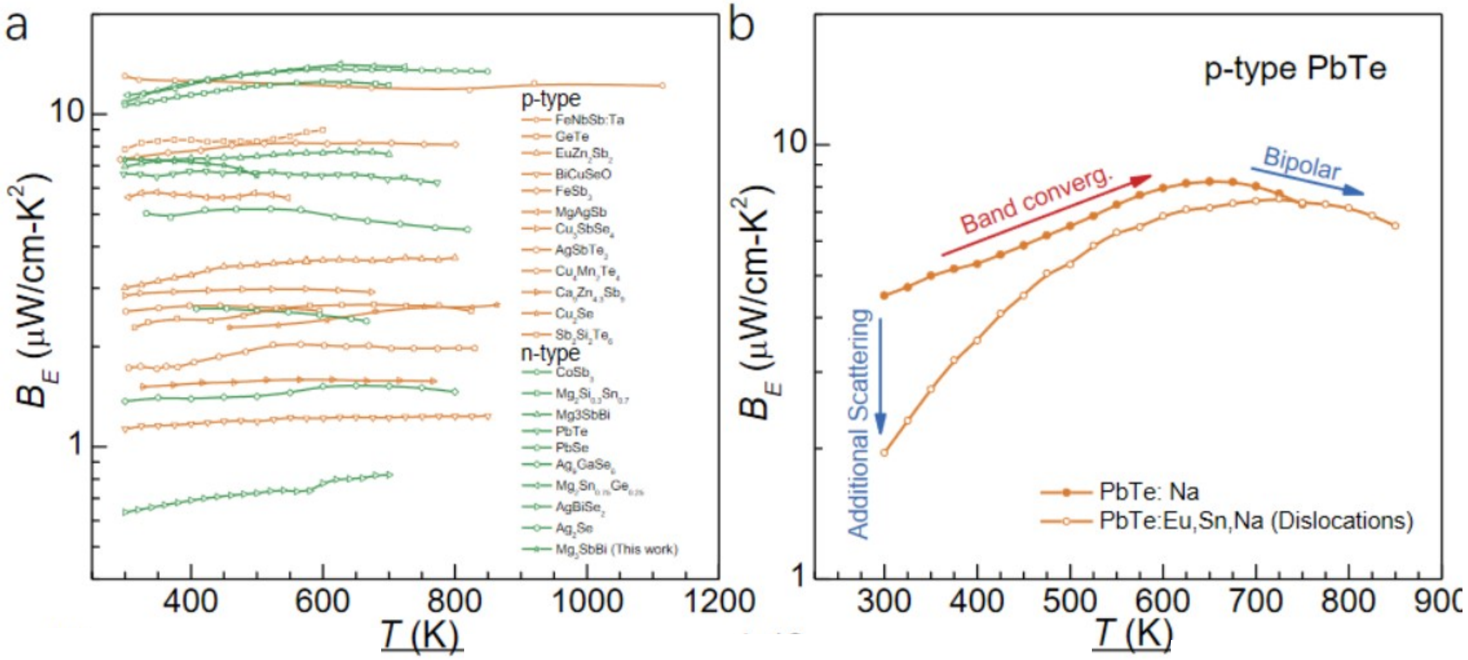
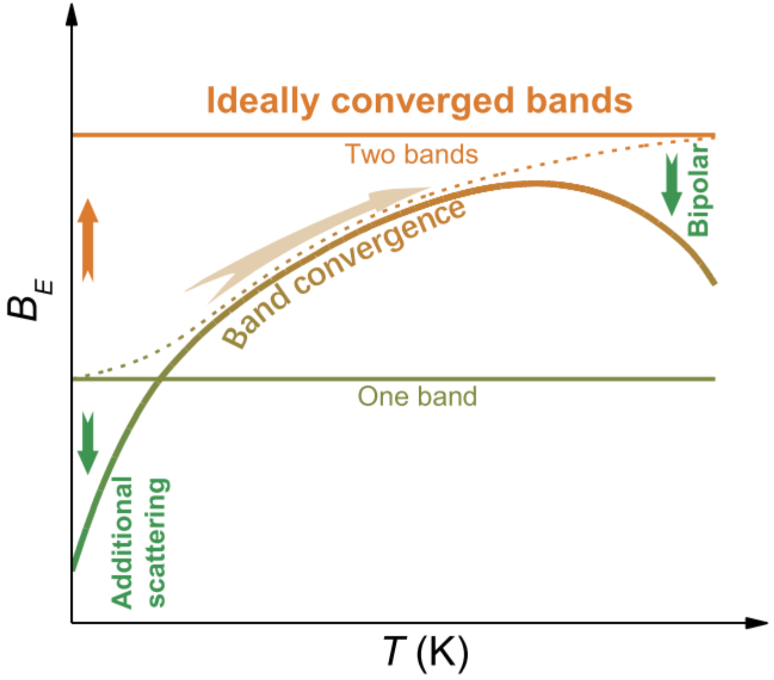
$B_E = \left(\frac{k_B}{e}\right)^2 \sigma_{E_0}$ is the electronic quality factor related to the transport parameter.[7]

Reminder: $\sigma_{E_0} = \frac{8\pi e(2m_e k_B T)^{3/2}}{3h^3}\mu_0\left(\frac{m_d^*}{m_e}\right)^{3/2}$ is a material constant for a good thermoelectric because the mobility tends to depend as $T^{-3/2}$, which cancels the temperature dependence here.





Summary

$$zT(\eta) = \frac{\alpha^2(\eta)}{\frac{(k_B/e)^2}{B\ln(1+e^{\eta})}+L(\eta)}$$

$$\alpha(\eta) = \pm\frac{k_B}{e}\left(\frac{(r+5/2)F_{r+3/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)}-\eta\right)$$

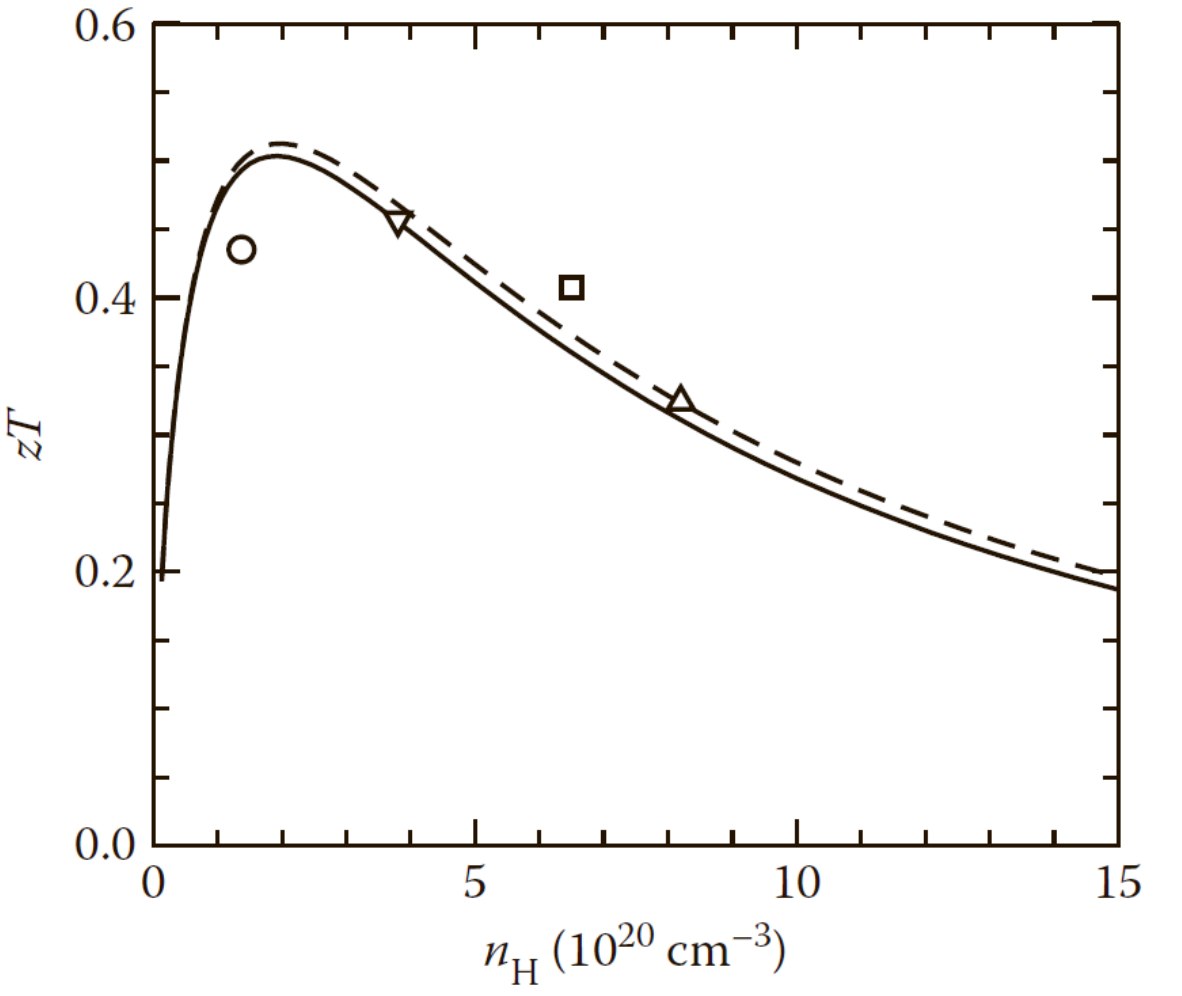
$$B = \left(\frac{k_B}{e}\right)^2\frac{8\pi e(2m_e k_B)^{3/2}}{3h^3}\frac{\mu_w}{\kappa_l}T^{5/2}$$

$$L(\eta) = \left(\frac{k_B}{e}\right)^2\left(\frac{(r+7/2)F_{r+5/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)}-\left[\frac{(r+5/2)F_{r+3/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)}\right]^2\right)$$

At the same time

$$n(\eta) = 4\pi\left(\frac{2m_d^* k_B T}{h^2}\right)^{3/2}F_{1/2}(\eta)$$

$$n_H(\eta) = n(\eta)r_H(\eta),\ r_H(\eta) = \frac{3}{2}F_{1/2}(\eta)\frac{\left(\frac{3}{2}+2r\right)F_{2r+1/2}(\eta)}{\left(\frac{3}{2}+r\right)^2F_{r+1/2}^2(\eta)}$$



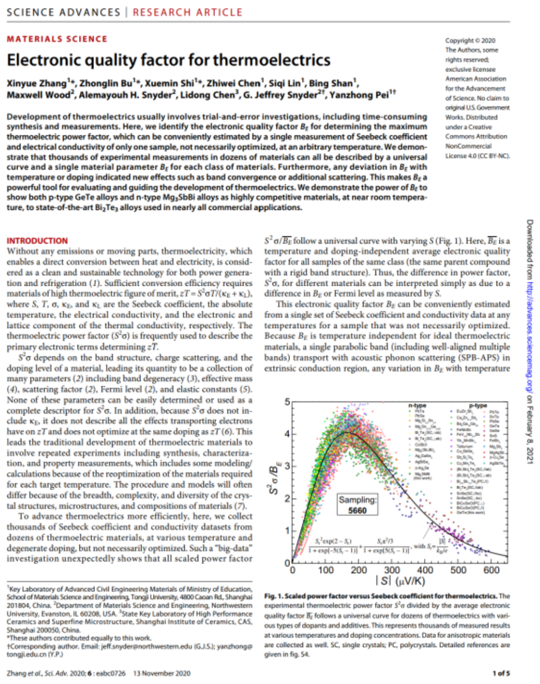
zT as function of doping requires only μ_w and κ_l.

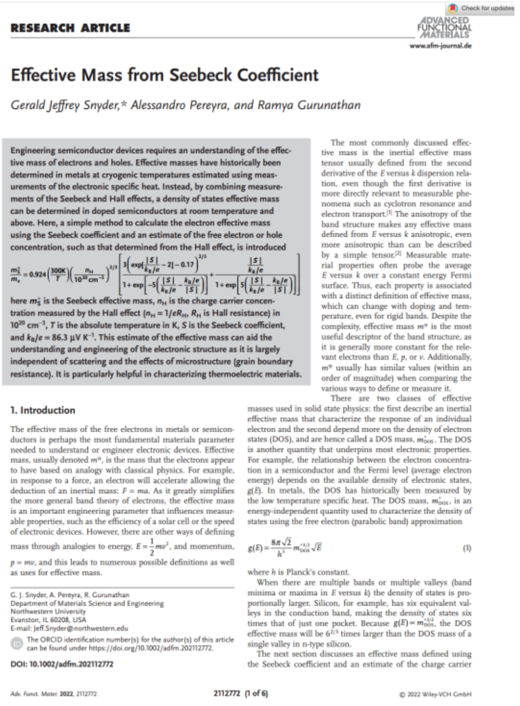
It is enough to measure one sample (α, σ, κ and n_H) to draw the entire $zT = f(n_H)$ curve.

How to calculate?

One way is to use all the formulas that were presented in this and previous lecture as it is (assuming acoustic phonon scattering $r = -1/2$, for example). Thus, the value of η can be found from the experimental value of α considering $\alpha(\eta) = \pm\frac{k_B}{e}\left(\frac{(r+5/2)F_{r+3/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)} - \eta\right)$. After this all the transport parameters can be calculated.

Another possible way to perform the SPB calculations is to use approximated sigmoid functions proposed by Snyder's group.[7–9]





Calculations

Assuming acoustic phonon scattering (which is reasonable for many of thermoelectrics):[7]

$$\alpha(\eta) = \pm\frac{k_B}{e}\left(\frac{2F_1(\eta)}{F_0(\eta)} - \eta\right)$$

thus

$$\alpha_r = \frac{|\alpha|}{k_B/e} = \frac{2F_1(\eta)}{F_0(\eta)} - \eta$$

at the same time:

$$\sigma = \sigma_{E_0}F_0(\eta) = B_E\left(\frac{e}{k_B}\right)^2 F_0(\eta)$$

while $B_E = \left(\frac{k_B}{e}\right)^2\sigma_{E_0} = \left(\frac{k_B}{e}\right)^2\frac{\alpha^2\sigma\sigma_{E_0}}{\alpha^2\sigma} = \left(\frac{k_B}{e}\right)^2\frac{\alpha^2\sigma\sigma_{E_0}}{\alpha^2\sigma_{E_0}F_0(\eta)} = \frac{\alpha^2\sigma}{\frac{\alpha^2}{(k_B/e)^2}F_0(\eta)} = \frac{\alpha^2\sigma}{\left(\frac{2F_1(\eta)}{F_0(\eta)}-\eta\right)^2 F_0(\eta)} = \frac{\alpha^2\sigma}{\alpha_r^2F_0(\eta)}$

For non-degenerate limit $F_0(\eta)$ reduces to $F_0(\eta) = e^{2-\alpha_r}$

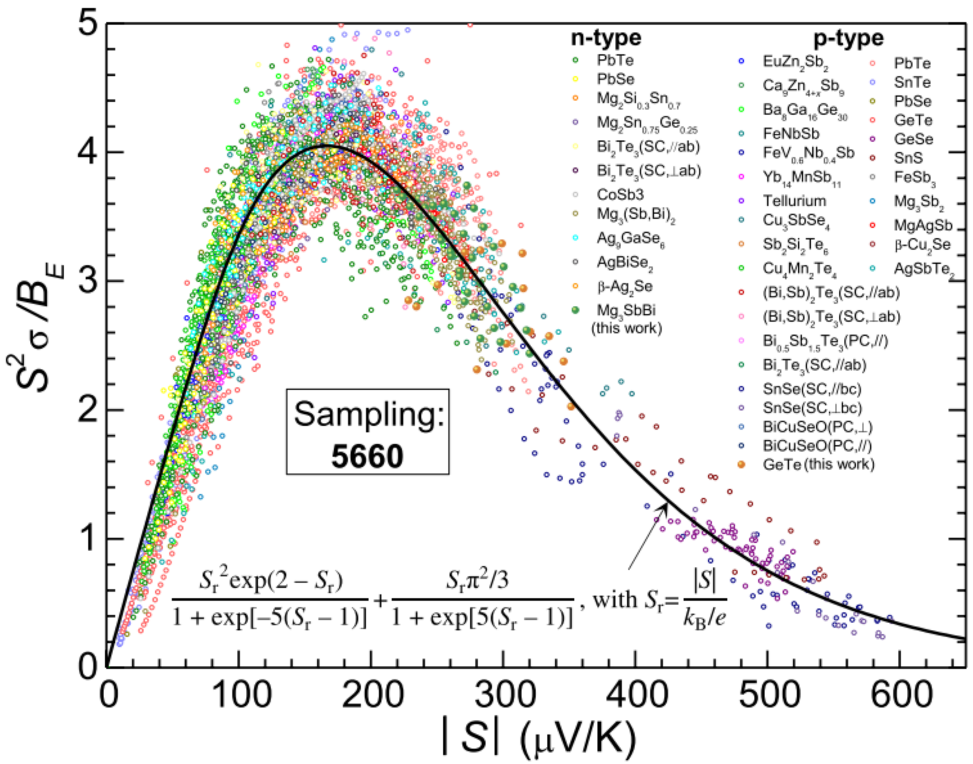
In fully degenerate case $F_0(\eta)$ reduces to $F_0(\eta) = \frac{\pi^2}{3\alpha_r}$

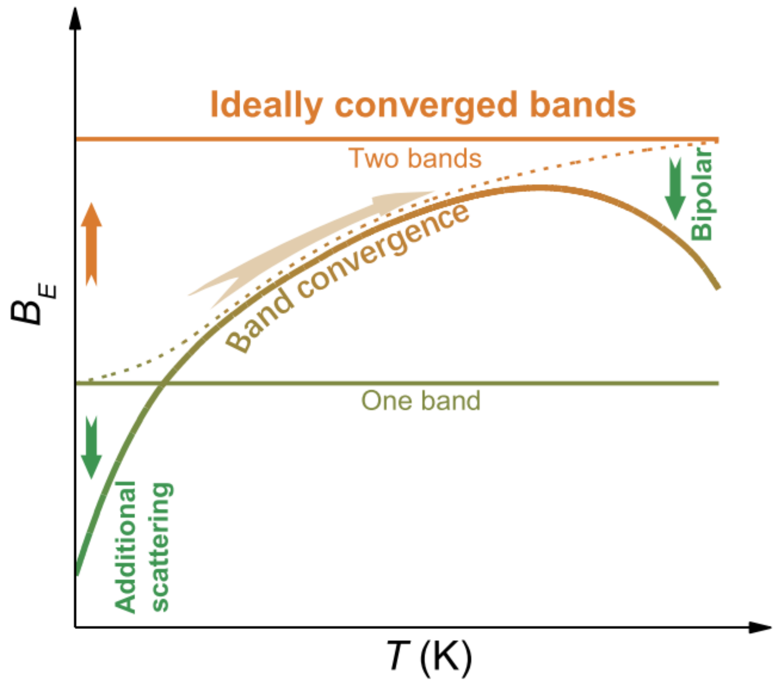
Calculations

Thus, by using sigmoid selection function that smoothly goes between the degenerate and non-degenerate limits the following relation can be written for B_E:[7,8]

$$B_E = \alpha^2\sigma\left[\frac{\alpha_r^2 e^{2-\alpha_r}}{1+e^{-5(\alpha_r-1)}} + \frac{\alpha_r\,\pi^2/3}{1+e^{5(\alpha_r-1)}}\right]^{-1}$$

here $\alpha_r = \frac{|\alpha|}{k_B/e}$, where α is the experimental value of the Seebeck coefficient.





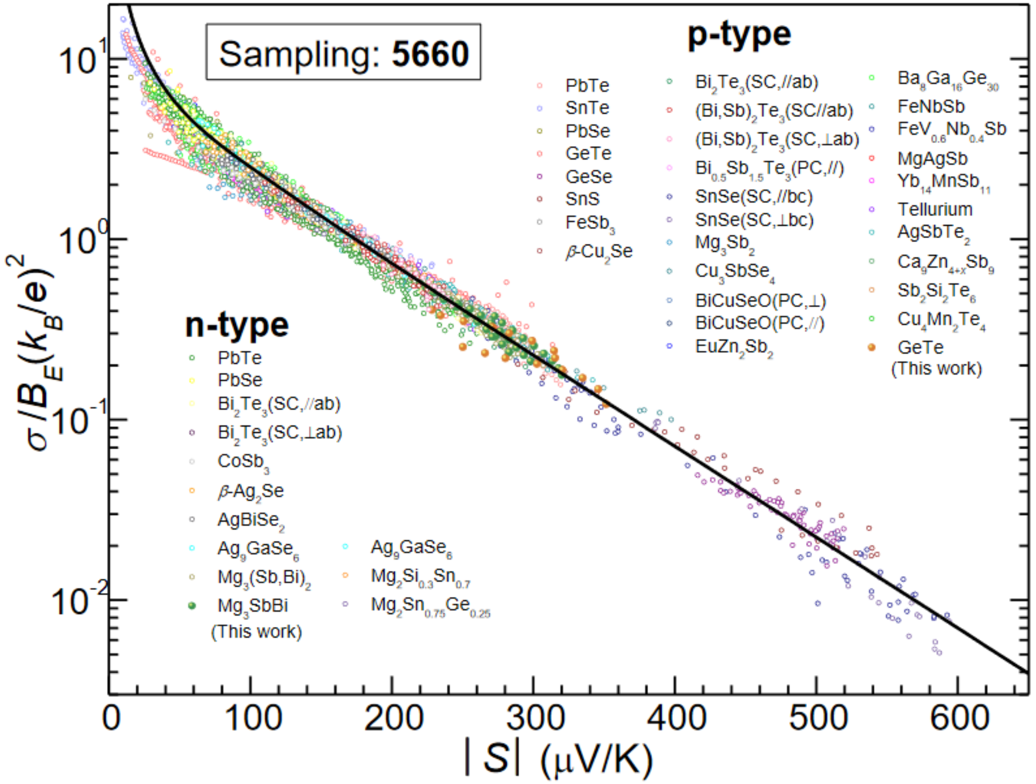
Calculations

Similarly, the electrical conductivity:[7,8]

$$\sigma = \sigma_{E_0}\left[\frac{e^{2-\alpha_r}}{1+e^{-5(\alpha_r-1)}} + \frac{\pi^2/3\alpha_r}{1+e^{5(\alpha_r-1)}}\right]$$

here $\alpha_r = \frac{|\alpha|}{k_B/e}$, where α is the experimental value of the Seebeck coefficient.

In this case the reduced Fermi energy can be calculated from $\sigma = \sigma_{E_0}\ln(1+e^{\eta})$



Calculations

In the same manner (see details in Refs. [7–9]) such relation can be written for the Seebeck effective mass:

$$m_S^* = \frac{h^2}{2k_BT}\left(\frac{3n_H}{16\sqrt{\pi}}\right)^{2/3}\left[\frac{(e^{\alpha_r-2}-0.17)^{2/3}}{1+e^{-5(\alpha_r-1)}}+\frac{\frac{3}{\pi^2}\left(\frac{2}{\sqrt{\pi}}\right)^{2/3}\alpha_r}{1+e^{5(\alpha_r-1)}}\right]$$

and for the weighted mobility:

$$\mu_w = \frac{3h^3\sigma}{8\pi e(2m_ek_BT)^{3/2}}\left[\frac{e^{\alpha_r-2}}{1+e^{-5(\alpha_r-1)}}+\frac{\frac{3}{\pi^2}\alpha_r}{1+e^{5(\alpha_r-1)}}\right]$$

here $\alpha_r = \frac{|\alpha|}{k_B/e}$, where α is the experimental value of the Seebeck coefficient.

Hall and weighted mobilities

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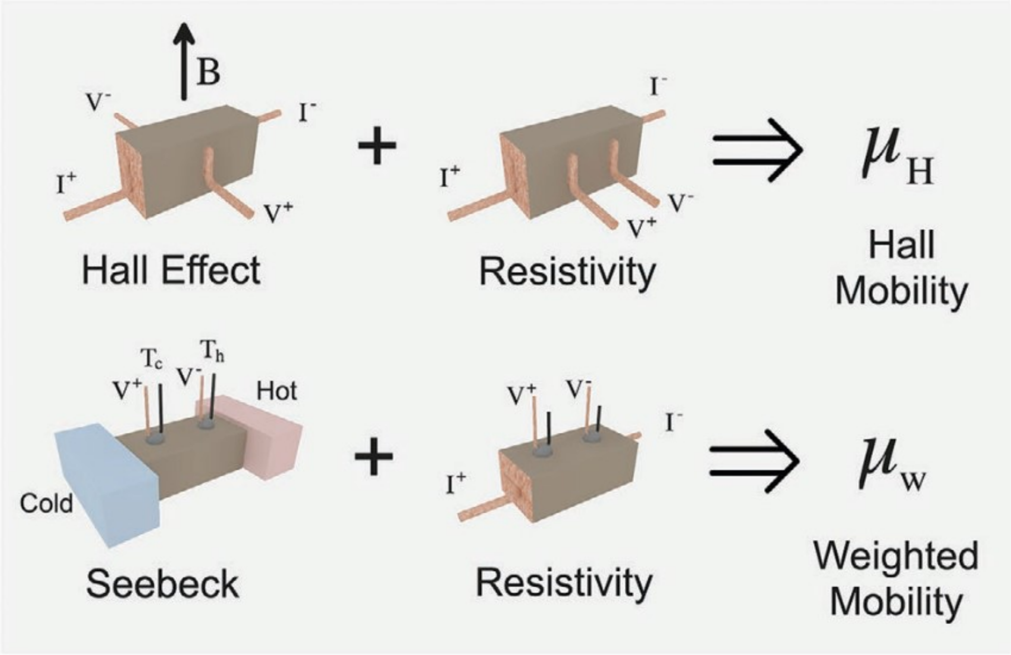
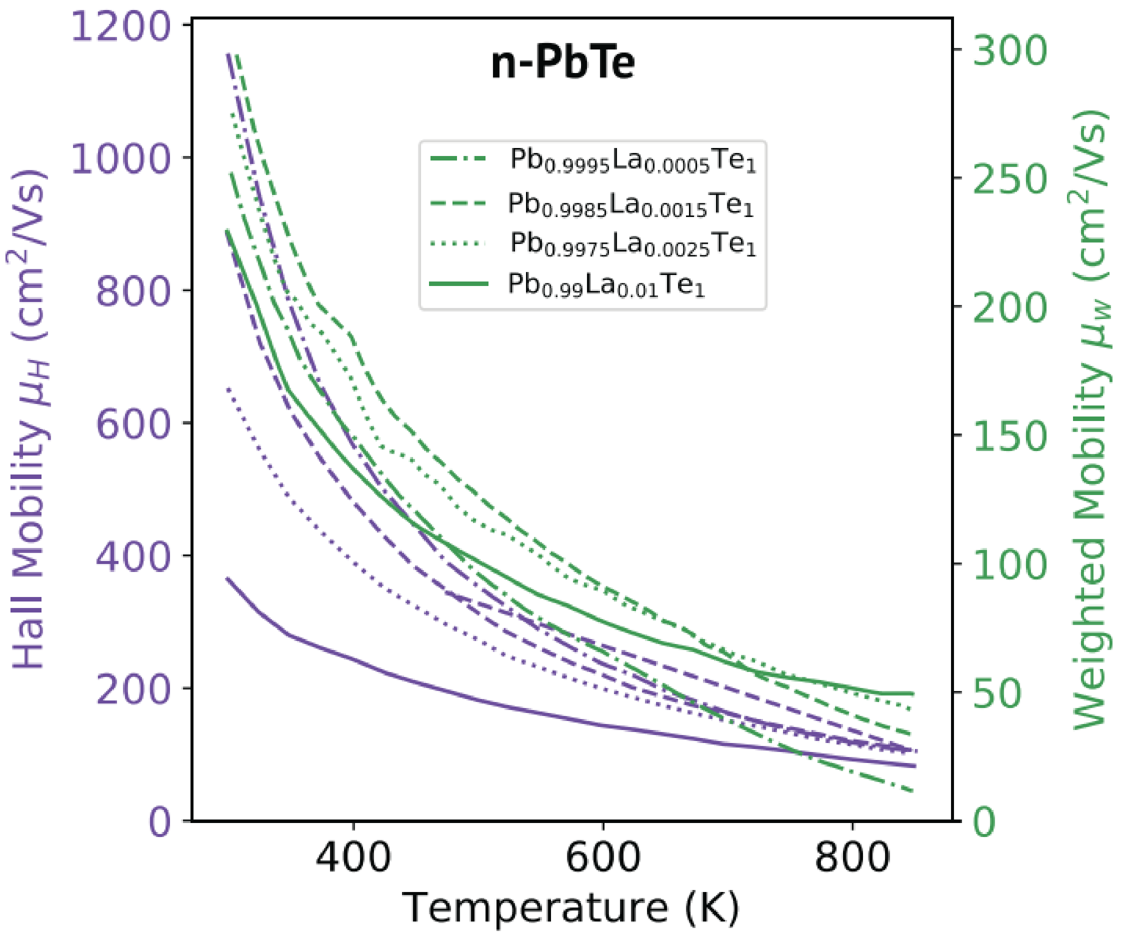
μ_H and μ_w are highly correlated except in samples with very high carrier concentration. This is because the Hall mobility depends on Fermi level through the Fermi integrals. This reduces the μ_H at high carrier concentrations. The weighted mobility should only depend on the intrinsic mobility parameter and the density of states m^*, which shouldn't change much in a parabolic band.[8]



$$\mu_H = \sigma R_H = \mu_0 \frac{F_{-1/2}}{2F_0}$$

$$\mu_w = f(\alpha, \sigma) = \mu_0 \left(\frac{m_d^*}{m_e}\right)^{\frac{3}{2}}$$



Hall and weighted mobilities

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Often the Hall mobility measurements are not accurate enough to notice the trend. The weighted mobility, in contrast, only depends on the mobility parameter and not Fermi level, so it is reasonable to expect it to be a constant with carrier concentration.[8]

$$\left.\begin{aligned} \mu_H &= \mu_0 \frac{F_{-1/2}}{2F_0} \\ \mu_w &= \mu_0 \left(\frac{m_d^*}{m_e}\right)^{\frac{3}{2}} \end{aligned}\right\} \frac{m_d^*}{m_e} = \left(\frac{\mu_w}{\mu_0}\right)^{\frac{2}{3}} = \left(\frac{\mu_w}{\mu_H}\frac{F_{-1/2}}{2F_0}\right)^{\frac{2}{3}}$$

Thus:

$$\frac{\mu_w}{\mu_H} = \left(\frac{m_d^*}{m_e}\right)^{\frac{3}{2}} \frac{2F_0}{F_{-1/2}}$$

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