Mineral Physics I
Chapter 5. Heat transfer properties
Section 2. Lattice thermal conductivity

TOMOO KATSURA

BAYERISCHES GEOINSTITUT, UNIVERSITY OF BAYREUTH, GERMANY

Inverse proportionality to distance

q Fourier's law: $q = -k(\mathrm{d}T/\mathrm{d}x)$

- Ø heat flux q proportional to the temperature difference ΔT and inversely proportional to distance x
- Ø $q \propto \Delta T / x$
- Ø The origin of the inverse proportionality to the distance is not obvious

q An example for no relation of q to x:

- Ø Heat transfer between surfaces A and B in space:
  - ü $q = \sigma \Delta T \cdot T_{\mathrm{AB}}^{3}$ (5.1.1)
    - § q is a function of $\Delta\, T$ but not x,
    - § We cannot define the thermal conductivity of space.

q The inverse proportionality to the distance is the central issue of the lattice thermal conductivity

Thermal resistance

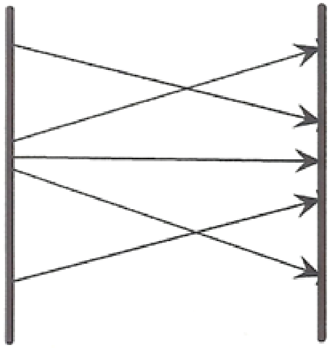
tional to the distance

ieat flow in matters

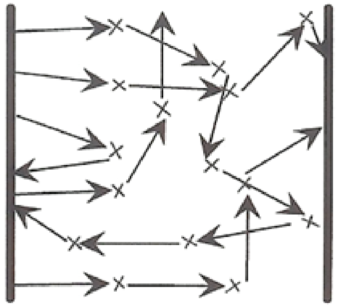
particles turn around

erred by phonons”

e vibration



No thermal resistance



Presence of
thermal resistance

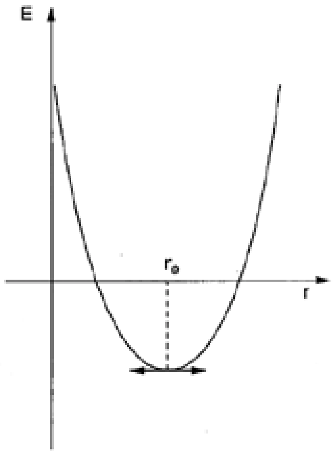
Mean free path & relaxation time

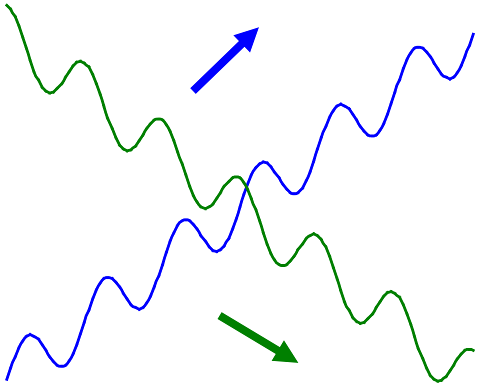
- **Mean free path** l: the distance where a particle can run without colliding any obstacle
  - q is proportional to l

- Lattice thermal conductivity in the quantum mechanics
  - $k \propto ncvl = Cvl$ (5.2.2)
    - n : the number of phonons per volume
    - c : the heat capacity per one phonon
    - v: the average velocity of phonons
  - $C = nc$: the heat capacity per volume

No thermal resistance in lattice thermal conductivity?

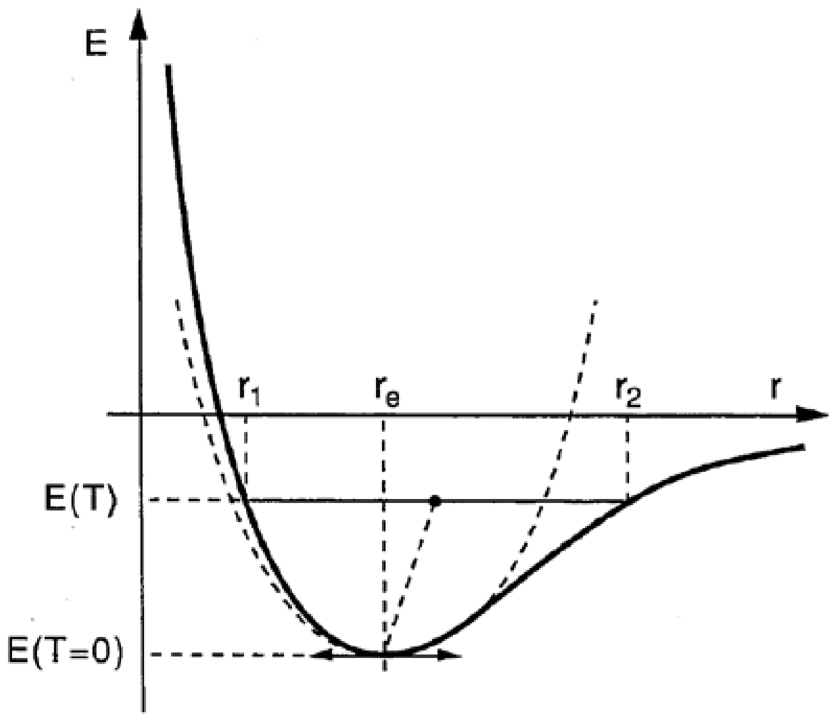
- What works as the thermal resistance?
  - If the atomic force is perfectly proportional to the displacement, the propagating lattice vibration goes straightly and is not attenuated.
    - Harmonic oscillation
  - Even if two waves intersect, two waves go straightly without interaction.
  - No thermal resistance?





Thermal resistance in lattice thermal conductivity

- The causes for thermal resistance of the lattice thermal conductivity
  - Defects in the lattice
    - vacancy, impurity, dislocation, stacking fault…
    - relatively significant at low temperatures
    - not essential for geophysics
  - Anharmonicity of the atomic potential
    - The force between atoms is not proportional to the displacement
    - The same cause for thermal expansion
    - significant at high temperatures
    - geophysically essential



Atomic vibration in the anharmonic potential -1

q An atomic potential $U(x)$ as a function of the displacement from the equilibrium position x:

Ø $U(x) = a_1 x^2 + a_2 x^3$ (5.2.3)

q If it were harmonic, the potential would be

Ø $U(x) = a_1 x^2$ (5.2.4)

q In this case, the equation of motion for the atom is

Ø $m \frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = F = -\frac{\mathrm{d}U}{\mathrm{d}x} = -2a_1 x$ (5.2.5)

ü m: mass of the atom

q The solution is:

Ø $x(t) = A\cos(\omega t)$ (5.2.6)

ü $\omega = \sqrt{2a_1/m}$

Atomic vibration in the anharmonic potential -2

q Now we argue the anharmonic oscillation by taking into account of the $a_2 x^3$ term:

Ø $U(x) = a_1 x^2 + a_2 x^3$ (5.2.7)

q The equation of motion becomes

Ø $m\frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = -2a_1 x - 3a_2 x^2$ (5.2.8)

Ø $\frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = -\omega^2 x + \phi x^2$ (5.2.9)

ü $\phi = -3a_2/m$

q Here, we assume a much smaller contribution of the higher-order term

Ø $|\omega^2 x| \gg |\phi x^2|$

Atomic vibration in the anharmonic potential -3

- For the first approximation, the solution of (5.2.9) $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} = -\omega^2 x + \phi x^2$ is assumed to have a formula with a slightly different angular frequency ω'
  - $x(t) = B\cos(\omega' t)$ (5.2.10)
    - $\omega^2 \gg |\omega^2 - \omega'^2|$ (5.2.11)
  - the accuracy of this solution is to be improved

- Eq. (5.2.9) $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} = -\omega^2 x + \phi x^2$ is modified by adding $\omega'^2 x$ to the both sides:
  - $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} + \omega'^2 x = -(\omega^2 - \omega'^2)x + \phi x^2$ (5.2.12)

- By substituting the first approximate solution Eq. (5.2.10) for the right side of Eq. (5.2.12)
  - $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} + \omega'^2 x = -(\omega^2 - \omega'^2)B\cos(\omega' t) + \phi[B\cos(\omega' t)]^2$

$$= -(\omega^2 - \omega'^2)B\cos(\omega' t) + \phi B^2(\cos(2\omega' t) + 1)/2 \quad (5.2.13)$$

Atomic vibration in the anharmonic potential -4

q Eq. (5.2.13) $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} + \omega'^2 x = -(\omega^2 - \omega'^2)B\cos(\omega' t) + \phi B^2(\cos(2\omega' t) + 1)/2$ is assumed to have a solution of:

Ø $x_a(t) = C_1\cos(\omega' t) + C_2\cos(2\omega' t) + C_0$ (5.2.14)

q By substituting Eq. (5.2.16) for Eq. (5.2.15), we have:

Ø $-4\omega'^2 C_2\cos(2\omega' t) - \omega'^2 C_1\cos(\omega' t) + \omega'^2 C_2\cos(2\omega' t) + \omega'^2 C_1\cos(\omega' t) + \omega'^2 C_0 = -(\omega^2 - \omega'^2)B\cos(\omega' t) + \phi B^2(\cos(2\omega' t) + 1)/2$

Ø $\cos(2\omega' t)$ term: $-3\omega'^2 C_2 = +\phi B^2/2$

Ø $\cos(\omega' t)$ term: $0 = -(\omega^2 - \omega'^2)B$

ü Since $\omega^2 - \omega'^2 \approx 0$, $B \neq 0$

Ø Constant term: $\omega'^2 C_0 = +\phi B^2/2$

Atomic vibration in the anharmonic potential -5

- A particular solution of (5.2.9) $\frac{\mathrm{d}^2x}{\mathrm{d}t^2} = -\omega^2 x + \phi x^2$ can be:
  - $x_p(t) = C_1 \cos(\omega' t) + \frac{\phi B^2}{6\omega'^2}\cos(2\omega' t) + \frac{\phi B^2}{2\omega'^2}$ (5.2.15)
- The assumed solution of (5.2.9) is
  - $x_a(t) = B \cos(\omega' t)$ (5.2.10)
  - $C_1 \approx B$ (5.2.16)
- By combining Eq.(5.2.17) and (5.2.12'), we have an approximate solution of Eq. (5.2.10) as
  - $x_h(t) \approx B \cos(\omega' t) + \frac{\phi B^2}{6\omega'^2}\cos(2\omega' t) + \frac{\phi B^2}{2\omega'^2}$ (5.2.17)

Physical meaning of the solution of the anharmonic oscillation

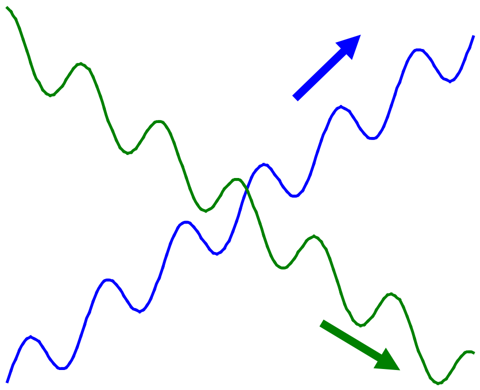
q $x(t) = B\cos(\omega' t) + \frac{\phi B^2}{6\omega'^2}\cos(2\omega' t) + \frac{\phi B^2}{2\omega'^2}$ (5.2.17)

Ø The higher term of the potential produces a higher harmonic oscillation

Ø The amplitude of the higher harmonic oscillation increases with the amplitude **squared** of the base wave

ü When two waves with the same amplitude intersect, the amplitude at the intersection becomes twice larger, and the amplitude of the higher harmonics become four times larger

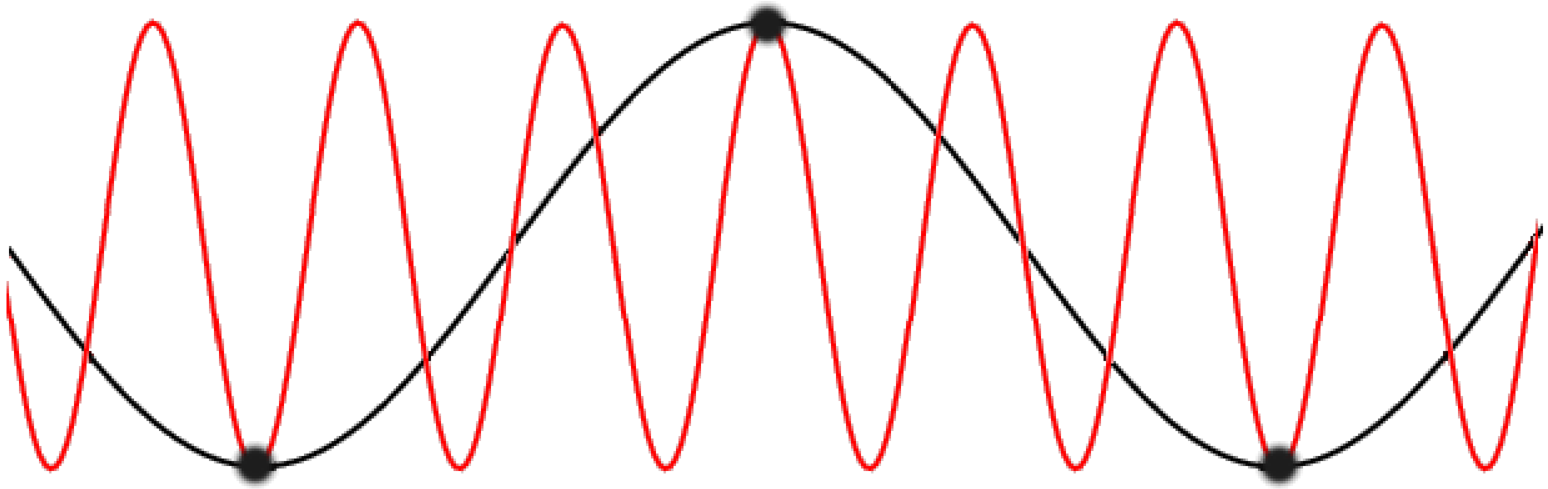
§ Intersection of waves produces a larger magnitude of higher harmonic oscillation



Turning-over of lattice wave

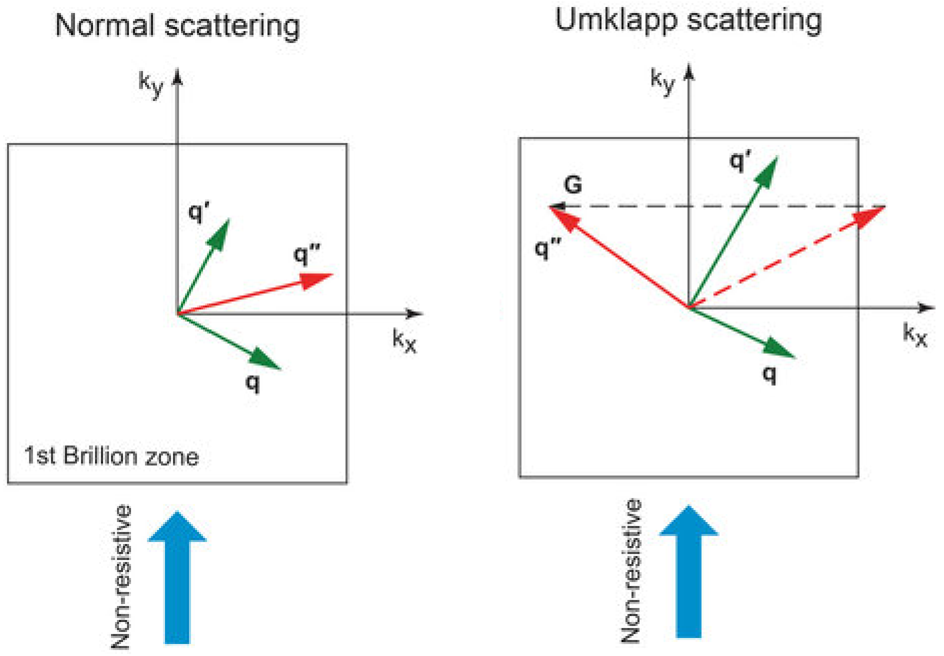
- Wave lengths of lattice waves cannot be shorter than 2✕atomic distance.
  - Chapter 3

- Waves shorter than 2✕atomic distance are identical to long waves propagating opposite to the original wave.



Umklapp scattering

- Phonon-phonon scattering
  - Intersection of two lattice waves → formation of a new lattice wave
- When two propagating waves intersect, the amplitude increases, then the new wave is generated, which propagates in the opposite direction.
  - Umklapp scattering
  - Origin of the thermal resistance of lattice thermal conductivity



Temperature dependence

q $x(t) = B\cos(\omega' t) + \frac{\phi B^2}{6\omega'^2}\cos(2\omega' t) + \frac{\phi B^2}{2\omega'^2}$ (5.2.17)

q The amplitude of the harmonics : proportional to the amplitude of the original wave squared

Ø More intense lattice vibration à higher production of the harmonics

Ø Rate of the Umklapp process: $\frac{\phi B^2}{6\omega'^2}/B = \frac{\phi B}{6\omega'^2}$ à $k \propto \left(\frac{\phi B}{6\omega'^2}\right)^{-1}$

q Amplitude of the lattice wave ∝ Temperature: $\frac{\phi B}{6\omega'^2} \propto T$ à $k \propto \frac{1}{T}$

q Higher temperature à larger amplitude à higher production of higher harmonics à more intense Umklapp scattering à larger thermal resistance à lower lattice thermal conductivity

q Lattice thermal conductivity should decrease with temperature

Pressure, temperature, structural dependence of lattice thermal conductivity

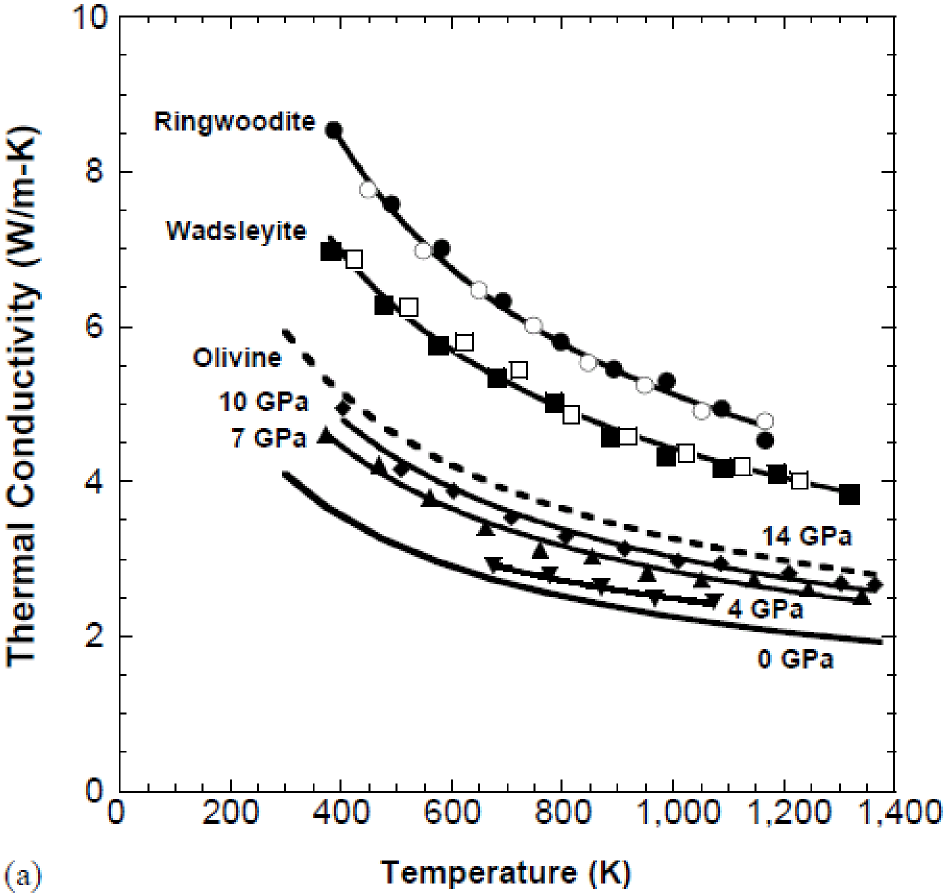
- Thermal resistance (Umklapp scattering) and thermal expansion
  - Caused by the anharmonicity of lattice vibration

- Opposite dependence between thermal conductivity and thermal expansion
  - Atomic distance increase/decrease ⇒ anharmonicity increase/decrease
  - Pressure, temperature and structure

	Thermal conductivity	Thermal expansion
Temperature ↑	↓	↑
Compression ↑	↑	↓
Coordination number ↑	↓	↑

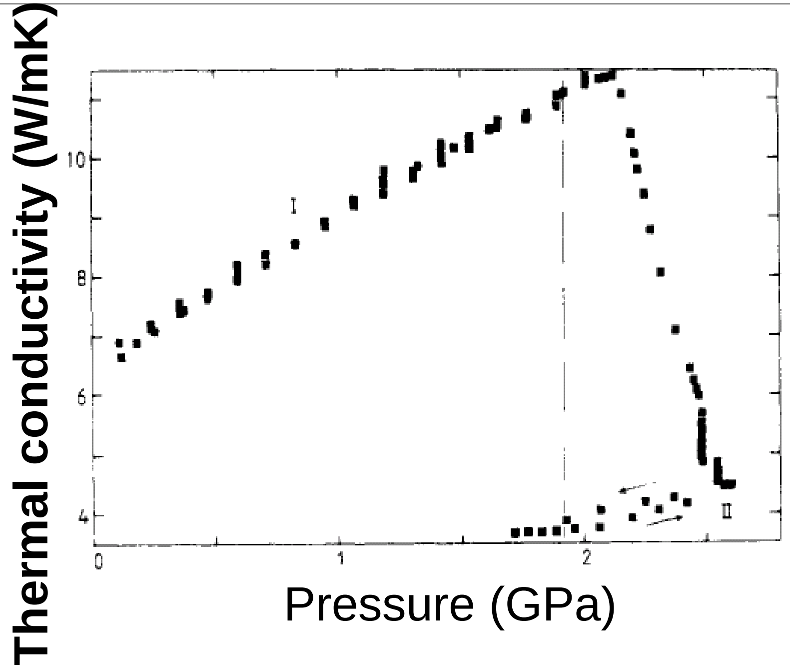
Example of lattice thermal conduction

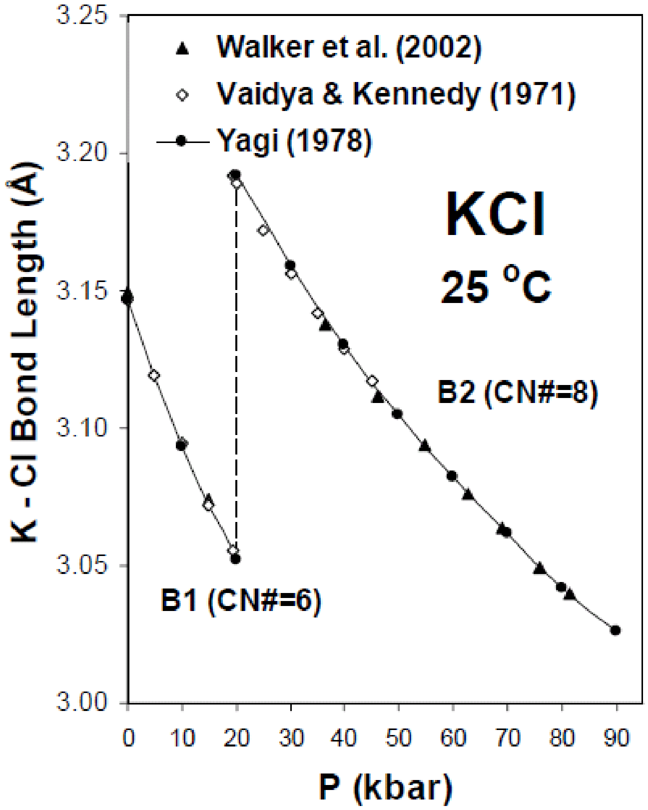
- Thermal conductivity of olivine, wadsleyite and ringwoodite [Xu et al., 2004]
  - Decrease with T
  - Increase with P
  - HP phases have higher conductivity
    - $k = Cvl$
      - HP phase: high sound velocity



(a)

Decrease in thermal conductivity with increase in coordination





- B1-B2 transition in KCl
  - Coordination number increase from 6 to 8
    - Atomic distance increase
    - Lattice thermal conductivity decrease

Mineral Physics I

Chapter 5. Heat transfer properties

Section 2. Lattice thermal conductivity

End