Physics with neutrons 2

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Exercise sheet 5
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EXERCISE 5.1

The Debye–Waller factor is used in to describe the attenuation of coherent neutron scattering caused by thermal motion:

$$f_{\mathrm{DWF}} = e^{-Q^2/3\langle u^2\rangle} = e^{-2W(\mathbf{Q})}.$$

For a cubic Bravais lattice we can make the following approximation:

$$2W = \frac{Q^2\hbar}{6MN}\int \frac{Z(\omega)}{\omega}\coth\left(\frac{\hbar\omega}{2k_BT}\right)d\omega, \tag{1}$$

where $Z(\omega)$ is the phonon density of states, M is the mass of the atom and N is the number of atoms in the crystal.
Within the Debye approximation, when the velocity of sound is frequency independent, we can express the phonon density of states for a cubic crystal with side length L by (in analogy with the theory of the black body radiation):

$$Z(\omega) = \frac{1}{2\pi^2}L^3\left(\frac{1}{c_L^3}+\frac{2}{c_T^3}\right)\omega^2. \tag{2}$$

c_L and c_T are the longitudinal and transverse velocity of sound, respectively. The total number of normal modes is $3N$. Therefore, we can put:

$$3N = \int_0^{\omega_D} Z(\omega)d\omega. \tag{3}$$

ω_D is the maximum frequency of the normal mode and $\Theta_D = \frac{\hbar\omega_D}{k_B}$ is the Debye temperature.

1. Calculate ω_D from the equations (2) and (3).
2. Calculate the asymptomatic behaviour of $2W$ for $T \ll \Theta_D$ and $T \gg \Theta_D$.
3. Copper crystallizes in fcc-lattice ($a = 3.615$ Å, $\rho_{\mathrm{Cu}} = 8920 kg/m^3$, $c_L = 4760 m/s$ and $c_T = 2320 m/s$);
   a) Calculate θ_D and show that the obtained value is reasonable.
   b) Figure 1 and 2 show $Z(\omega)$ and the dispersion relation for copper, respectively (Nilsson 1973). What are the reasons of deviations to the Debye model?

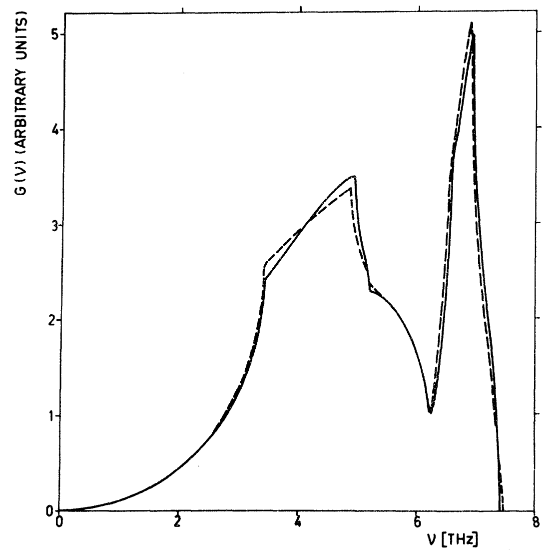

Figure 1: Phonon frequency distributions calculated from the Born-von Kármán models.

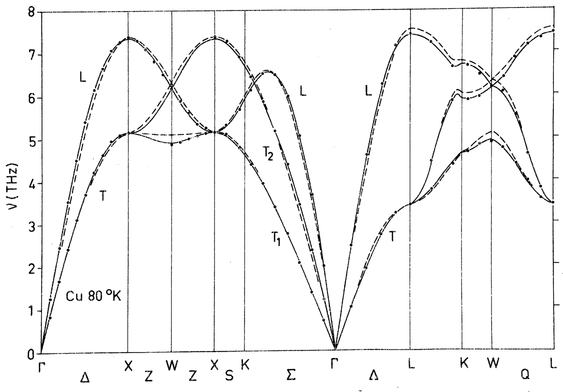

Figure 2: Dispersion curves for Cu at $80K$.

4. Calculate the mean amplitude $\langle u^2\rangle$ for copper at $T = 20K, 100K, 500K, 1000K$.
5. Estimate the attenuation in a neutron powder diffraction measurement with wavelength $\lambda = 1.188$ Å of (100) and (440) reflex due to the Debye-Waller factor ($T = 200K$).
6. a) Why do soft materials have a larger Debye-Waller factor than condensed matter?
   b) What is the influence of mass on the Debye-Waller factor?

Solution. 1. Within the Debye approximation we have

$$3N = \int_0^{\omega_D} Z(\omega)d\omega = \int_0^{\omega_D} \frac{1}{2\pi^2}L^3\frac{3}{c^3}\omega^2 d\omega = L^3\frac{\omega_D^3}{2\pi^2 c^3}.$$

Plugging this into Eq. (2), we get

$$Z(\omega) = \frac{3}{2\pi^2 c^3}L^3\omega^2 = 9N\frac{\omega^2}{\omega_D^3}.$$

2. Plugging the Debye approximation into the Eq. (1), we get

$$2W = \frac{Q^2\hbar}{6MN}\int_0^{\omega_D} 9N\frac{\omega^2}{\omega_D^3\omega}\coth\left(\frac{\hbar\omega}{2k_BT}\right)d\omega = \frac{3Q^2\hbar}{2M\omega_D^3}\int_0^{\omega_D}\omega\coth\left(\frac{\hbar\omega}{2k_BT}\right)d\omega$$

We substitute $x = \hbar\omega/k_BT$, $x_D = \hbar\omega_D/k_BT$:

$$\frac{3Q^2\hbar}{2M\omega_D^3}\left(\frac{k_BT}{\hbar}\right)^2\int_0^{x_D} x\coth(x/2)\,dx = \frac{3Q^2k_B^2T^2}{2M\hbar\omega_D^3}\underbrace{\int_0^{x_D} x\coth(x/2)\,dx}_{I} \tag{4}$$

For high temperatures $\Theta_D \ll T, x_D \ll 1$, the asymptotic behaviour is obtained from the series expansion

$$\coth(x/2) = \frac{1}{x/2} + \frac{x/2}{3} + \frac{(x/2)^3}{45} + \ldots$$

yielding for the integrand ($x_D \ll 1$)

$$I \approx \int_0^{x_D} 2 + \frac{x^2}{6} dx = \left[2x + x^3/12\right]_{x=0}^{x_D} = 2x_D + x_D^3/12 \approx 2\frac{\hbar\omega_D}{k_B T}.$$

Plugging this into Eq. (4), we get

$$2W = \frac{3Q^2 k_B^2 T^2}{2M\hbar\omega_D^3} 2\frac{\hbar\omega_D}{k_B T} = 3\frac{Q^2\hbar^2 T}{M k_B \Theta_D^2}. \tag{5}$$

For low temperatures $\Theta_D \gg T, x_D \gg 1$, we have to write the integrand in such a way, that it is valid for small and large x, since the integration range lies between 0 and x_D. We use the series:

$$\coth(x/2) = \frac{1+e^{-x}}{1-e^{-x}} = 1 + 2\sum_{n=1}^{\infty} e^{-2nx}, \qquad (x > 0).$$

Inserting this we get

$$I = \int_0^{x_D} x\left(1 + 2\sum_{n=1}^{\infty} e^{-2nx}\right) dx = \int_0^{x_D} x dx + 2\sum_{n=1}^{\infty}\int_0^{x_D} x e^{-nx} dx$$
$$= \frac{x_D^2}{2} + 2\sum_{n=1}^{\infty}\left(\frac{1}{n^2} - \frac{n x_D + 1}{n^2} e^{-n x_D}\right) = \frac{x_D^2}{2} + \frac{\pi^2}{3} - 2\sum_{n=1}^{\infty}\left(\frac{n x_D + 1}{n^2} e^{-n x_D}\right).$$

The sum is convergent, since $(n x_D + 1)/n^2 < 1$ for $n \geq x_D + 1$ and $\sum_{n=1}^{\infty} \exp(-n x_D) = 1/(\exp(n) - 1)$. Moreover, for $x_D \gg 1$ the sum is dominated by the exponential, hence

$$2\sum_{n=1}^{\infty}\left(\frac{n x_D + 1}{n^2} e^{-n x_D}\right) \approx 0.$$

Inserting this into (4) finally yields

$$2W = \frac{3Q^2 k_B^2 T^2}{2M\hbar\omega_D^3}\left(\frac{x_D^2}{2} + \frac{\pi^2}{3}\right) = \frac{3Q^2\hbar^2}{4k_b\Theta_D M} + \frac{Q^2\pi^2\hbar^2 T^2}{2k_b\Theta_D^3 M}. \tag{6}$$

The first factor origins from the zero-point motion and the second from low-energy phonons.

3. a) The highest frequency mode ω_D depends on the crystal geometry. Using $\frac{1}{c_{\mathrm{ef}}^3} = \frac{1}{3}\left(\frac{2}{c_{\mathrm{t}}^3} + \frac{1}{c_{\mathrm{l}}^3}\right)$ and $N/L^3 = \rho_{\mathrm{Cu}}/m_{\mathrm{Cu}}$ with the molar mass of Copper $m_{\mathrm{Cu}} = 63.59/6 \cdot 10^{23} g$ we get,

$$\omega_D = c_{\mathrm{eff}}\left(\frac{6N\pi^2}{V}\right)^{1/3} = c_{\mathrm{eff}}\left(\frac{6\pi^2\rho_{\mathrm{Cu}}}{m_{\mathrm{Cu}}}\right)^{1/3} = 45\mathrm{THz} = \frac{7.2}{2\pi}\mathrm{THz},$$

$$\Theta_D = 340K.$$

The results agree with Fig. 2.

b) In reality, $\omega = \omega(k)$ is dispersive. Therefore $Z(\omega)$ is not proportional to ω^2 (Debye-Model), but

$$Z(\omega) = \frac{V}{8\pi^3} \int \frac{d\sigma}{\nabla_q \omega(q)}.$$

If the dispersion becomes flat, deviations from the Debye-Model arise.

4. For a cubic Bravais lattice, the mean displacement due to lattice vibrations is given by

$$\langle \mathbf{u}^2 \rangle = 6W(\mathbf{Q})/\mathbf{Q}^2.$$

At low temperatures we get with Eq. (6)

$$\langle \mathbf{u}^2 \rangle_\mathrm{l} = \frac{9\hbar^2}{M k_B \Theta_D} \left(\frac{1}{4} + \frac{T^2 \pi^2}{6\Theta_D^2} \right)$$

and for high temperatures with Eq. (5)

$$\langle \mathbf{u}^2 \rangle_\mathrm{h} = \frac{9\hbar^2}{M k_B \Theta_D} \left(\frac{T}{\Theta_D} \right).$$

The results are shown in Table 1. The nearest-neighbour distance for Copper is $d = a/\sqrt{2} = 2.5$ Å. The mean thermal amplitude at $T = 1000K$ is approximately $1/10d$.

Table 1: Mean displacement of atoms in Co.

		low T		high T	
$T(K)$	T/Θ_D	$\langle u^2 \rangle_\mathrm{l}$ (Å^2)	$\sqrt{\langle u^2 \rangle_\mathrm{l}}$ (Å)	$\langle u^2 \rangle_\mathrm{h}$ (Å^2)	$\sqrt{\langle u^2 \rangle_\mathrm{h}}$ (Å)
0	0	0.00506	0.0711		
20	0.059	0.00517	0.0719		
100	0.29	0.0065	0.0876	0.0059	0.0768
200	0.58	0.0107	0.1034	0.0117	0.1084
500	1.44			0.0291	0.171
1000	2.94			0.0594	0.243

5. Copper: fcc, $a = 3.615$ Å.

$$Q_{(100)} = \frac{2\pi}{a}\sqrt{1^2 + 0^2 + 0^2} = 1.7381\text{Å}^{-1}$$

$$Q_{(440)} = \frac{2\pi}{a}\sqrt{4^2 + 4^2 + 0^2} = 9.83\text{Å}^{-1}$$

We get the Debye-Waller factor with $f_\mathrm{DWF} = \exp(-2W) = \exp(-\mathbf{Q}^2 \langle u^2 \rangle/3)$. The results are given in Table 2.

6. a) Soft materials have a "small" spring-constant, e.g. the harmonic potential is flat and $\langle u^2 \rangle$ large

$$\omega = \sqrt{\frac{K}{M}}.$$

K small ⇒ many low-energy oscillations ⇒ ω_D small ⇒ Θ_D small.

Table 2:

$\exp(-2W)$	$T = 20K$	$T = 200K$	$T = 1000K$
(100)	0.994	0.98	0.94
(440)	0.85	0.83	0.15

b) We use the linear approximation of the exponential. For low temperatures $\Theta_D \gg T$ we get

$$f_{\text{DWF}} \propto \frac{1}{M}.$$

For high temperatures $\Theta_D \ll T$ we have

$$f_{\text{DWF}} \propto \frac{T}{M}.$$

□