Physics with neutrons 1

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Exercise sheet 7
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Exercise 7.1

Insulating organo-metallic compound $NiCl_2 - 4SC(NH_2)_2$ (know as DTN) demonstrates magnetoelastic properties (Phys. Rev. B 77, 020404(R) (2008)). In an applied magnetic field its c-axis first shrinks by $6 \cdot 10^{-3}\%$ and then expands up to $2.2 \cdot 10^{-2}\%$ in comparison to the zero field value. Calculate whether it is possible to detect such a change in length of the c-axis using powder neutron diffractometer HRPT located in PSI (the instrumental resolution is equal to $\Delta\theta/\theta = 9.5 \cdot 10^{-4}$ for $Q = (002)$). The unit cell is tetragonal (space group I4 number 79) and the lattice parameters (zero magnetic field) are: $a = b = 9.558$ Å, $c = 8.981$ Å.

Solution. In the magnetic field the c-axis shrinks by $6 \cdot 10^{-3}\%$ and then expands by $2.2 \cdot 10^{-2}\%$ (Figure 1, 2). The instrument resolution is given by $\Delta\theta/\theta = 9.5 \cdot 10^{-4} \approx 10^{-3}$. Since the unit cell is tetragonal, we have

$$c^* = \frac{2\pi}{V_0} a \times b = 2\pi \frac{a \times b}{a \times b \cdot c} = \frac{2\pi}{||c||}.$$

We look at $Q = (002)$ and get

$$Q = (0, 0, 2) = \left(0, 0, \frac{4\pi}{||c||}\right) = (0, 0, 1.3992\text{Å}^{-1}).$$

The largest relative change between the shrinked and expanded c-axis is

$$\frac{\Delta d}{d} = 6 \cdot 10^{-3}\% + 2.2 \cdot 10^{-2}\% = 2.8 \cdot 10^{-4}$$

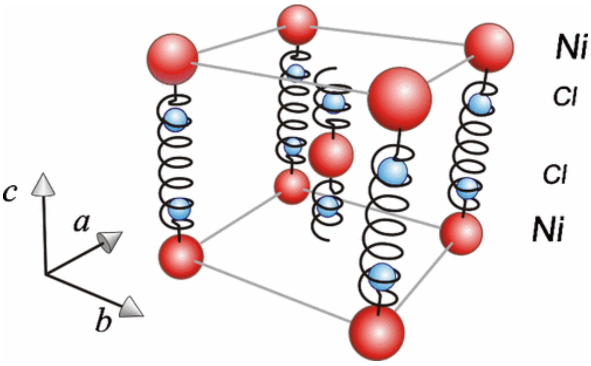
Now use Bragg's law and $d = 2\pi/Q$

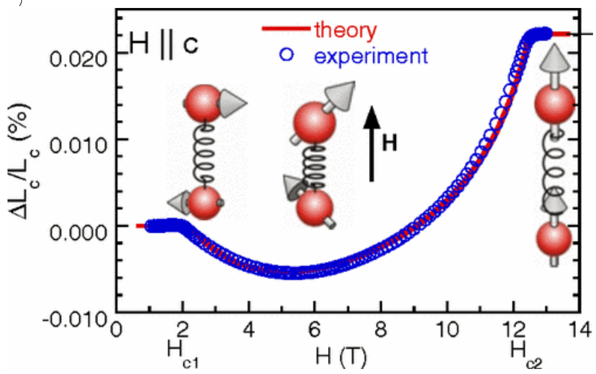
$$\sin\theta_{(002)} = \frac{\lambda}{2\pi}\frac{Q}{2} = \frac{\lambda}{2\pi}\frac{4\pi}{2||c||} = \frac{\lambda}{||c||},$$

which means that the peak shifts by

$$\frac{\Delta\theta}{\theta} = \left| \frac{\arcsin\left(\frac{\lambda}{c + c\Delta d/d}\right) - \arcsin\left(\frac{\lambda}{c}\right)}{\arcsin\left(\frac{\lambda}{c}\right)} \right| \approx \left| \frac{\left(\frac{\lambda}{c + c\Delta d/d}\right) - \left(\frac{\lambda}{c}\right)}{\left(\frac{\lambda}{c}\right)} \right| = \left| \frac{1}{1 + \Delta d/d} - 1 \right| = 2.8 \cdot 10^{-4}$$
$$\leq \left(\frac{\Delta\theta}{\theta}\right)_{\text{instrument}} = 10^{-3}.$$

. Here we used $\arcsin x \approx x$ for small x. Therefore, the effect cannot measured with this instrument.





□

Figure 1: Unit cell of tetragonal $NiCl_2 - 4SC(NH_2)_2$.

Figure 2: Comparison of experimental c-axis magnetostriction data as a function of H for $H||c$ with a model.

Exercise 7.2

Highly oriented pyrolytic graphite (HOPG) is used as one of the most efficient monochromators for thermal and cold neutrons. In addition, HOPG is used as a filter for neutrons. Graphite has a hexagonal crystal structure. Along the $[00l]$ direction, the crystal planes are regularly stacked thus exhibiting the properties of a single crystal. Within the hexagonal planes, the atomic sheets are oriented randomly, i.e. like a powder. Calculate the energies for the cut-offs of the first few reflections (002), (004), (006), (101), (102), (103), (104), (105) and (106). The lattice constants are $a = 2.4612$ Å and $c = 6.7079$ Å. The stacking along the c-direction is such that the peaks with $(00l)$, l odd, are extinguished.

Solution. Neutron wavelength filters are usually polycrystals with high coherent scattering crosssection and low absorption cross-section. Such a polycrystal of sufficient thickness will scatter all neutrons below a certain wavelength cutoff $\lambda_c = 2d$ out of the beam (where d is the largest distance of lattice planes for which Bragg scattering is allowed). One of the materials used as a filter is Beryllium, whose cutoff wavelength is $\lambda_c = 3.9$Å. If shorter neutron wavelengths should be transmitted, it is hard to find crystals with such small lattice constants. Here HOPG can be used, which is not a polycrystal but is well-ordered along the $[00l]$ direction. In the perpendicular directions, the orientation of the hexagonal planes is random. Therefore, using a HOPG with c oriented along the beam as a filter, not all wavelengths are scattered out of the beam, but only those that match a (hkl) reflex with h or k nonzero. (Note: for a three-dimensional single crystal, that would only be possible for one specific (hkl) and its multiples; due to the rotational disorder, the Bragg condition can be fulfilled for all reflexes.) To calculate the energies of scattered-out neutrons, we look at basic properties of the hexagonal system: the base vectors are

$$\mathbf{a}_1 = (a, 0, 0), \mathbf{a}_2 = (a/2, a\sqrt{3}/2, 0), \mathbf{a}_3 = (0, 0, c);$$

the reciprocal vectors are

$$\mathbf{b}_1 = 2\pi\frac{2}{a\sqrt{3}}(\sqrt{3}/2, -1/2, 0), \mathbf{b}_2 = 2\pi\frac{2}{a\sqrt{3}}(0, 1, 0), \mathbf{b}_3 = 2\pi\frac{1}{c}(0, 0, 1).$$

The modulus of a reciprocal lattice vector $\mathbf{G}_{hkl} = h\mathbf{b}_1 + k\mathbf{b}_2 + l\mathbf{b}_3$ is

$$|\mathbf{G}_{hkl}| = 2\pi\sqrt{(h^2 + k^2 + hk)\frac{4}{a^2 3} + \frac{l^2}{c^2}}.$$

From this, we can find the $d_{hkl} = 2\pi/|\mathbf{G}_{hkl}|$ for a given reflection in the hexagonal crystal system:

$$\frac{1}{d_{hkl}} = \sqrt{\frac{4(h^2 + k^2 + hk)}{3a^2} + \frac{l^2}{c^2}}$$

and the Bragg angle ϑ_{hkl} betwenn the neutron path - along [001] - and the lattice planes for $[hkl]$:

$$\vartheta_{hkl} = \frac{\pi}{2} - \cos^{-1}\frac{\mathbf{G}_{hkl}\cdot\mathbf{G}_{00l}}{|\mathbf{G}_{hkl}||\mathbf{G}_{00l}|} = \frac{\pi}{2} - \cos^{-1}\frac{l/c}{\sqrt{4(h^2+k^2+hk)/(3a^2)+l^2/c^2}}.$$

The wavelength that is scattered out is then

$$\lambda_{hkl} = 2d_{hkl}\sin\vartheta_{hkl}.$$

Results:

h	k	l	λ (Å)	$E(meV)$
0	0	2	6.71	1.82
0	0	4	3.35	7.29
0	0	6	2.24	16.31
1	0	1	1.23	54.82
1	0	2	1.93	22.03

h	k	l	λ (Å)	$E(meV)$
1	0	3	2.13	18.10
1	0	4	2.07	19.12
1	0	5	1.92	22.21
1	0	6	1.75	26.68

In particular the (101) reflection is notable: for a wavelength of 2.54 Å HOPG has a good transmission, while 1.23 Å is filtered out very well by this reflex. □

Exercise 7.3

Derive the Lorentz factor

$$L(\theta) = \frac{1}{\sin\theta\sin 2\theta}$$

The origin of the Lorentz factor is twofold:

1. The statistical distribution of the crystallites in a polycrystalline sample has to be considered.
2. The detector covers only part of the Debye-Scherrer cone, which describes the Bragg scattering from polycrystalline materials. As sketched in Figure 3, the wavevector $\mathbf{k}'$ of the scattered neutrons lies on a cone, known as Debye-Scherrer cone, where the axis of the cone is along the wavevector $\mathbf{k}$ of the incoming neutrons and θ is the Bragg angle.

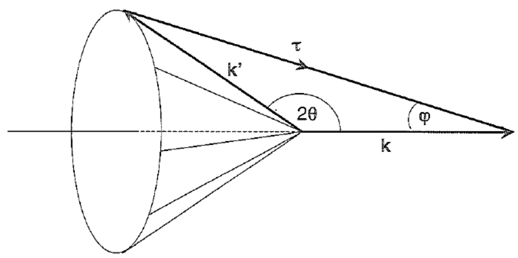


Figure 3: Debye-Scherrer cone for Bragg scattering from polycrystalline materials.

Solution. First we consider the statistical distribution of the crystallites in a polycrystalline sample. The fraction of microcrystals oriented to fulfill Bragg's law $\lambda = 2d\sin\theta$ can be obtained by considering Figure 4. All crystallites with reciprocal lattice vectors lying in the dotted surface area of a sphere with radius τ contribute to the scattering. The active surface area amounts to $2\pi\tau^2\cos\theta d\theta/4\pi\tau^2$. Hence, the total scattering for the Debye-Scherrer cone is given by

$$\sigma_{\text{cone}} \propto \int_0^{\pi/2} \delta(k'-k)cos\theta d\theta, \quad (1)$$

where $\delta(k'-k)$ confines the integration to elastic scattering. Using the geometry sketched in Figure 3 we find

$$k'^2 - k^2 = \tau^2 - 2\tau k\cos\phi = \tau^2 - 2\tau k\sin\theta = (k'+k)(k'-k),$$

where we used the relation $\theta = \pi/2 - \phi$. Setting $k' \approx k$ yields

$$k' - k = \frac{1}{2k}(\tau^2 - 2\tau k \sin\theta). \tag{2}$$

Combining Eqs. (1) and (2) yields

$$\sigma_{\mathbf{cone}} \propto \int_0^{\pi/2} \delta(\tau^2 - 2\tau k \sin\theta) \cos\theta d\theta. \tag{3}$$

We solve the integral in Eq. (3) by the substituion $x = 2\tau k \sin\theta$:

$$\sigma_{\mathbf{cone}} \propto \int_0^{\pi/2} \delta(\tau^2 - x)\frac{1}{2\tau k} dx = \frac{1}{2\tau k}.$$

Setting $\tau = 2k \sin\theta$ from Bragg's law we find

$$\sigma_{\text{cone}} \propto \frac{1}{\sin\theta}. \tag{4}$$

If the neutron detector with diameter d is at a distance r from the sample, it intercepts a fraction $q = d/2\pi r \sin(2\theta)$ of the neutrons in the cone. Multiplying σ_{cone} of Eq. (3) with the θ-dependent term of q yields the final result for the Lorentz factor:

$$L(\theta) = \frac{1}{\sin(\theta)\sin(2\theta)}$$

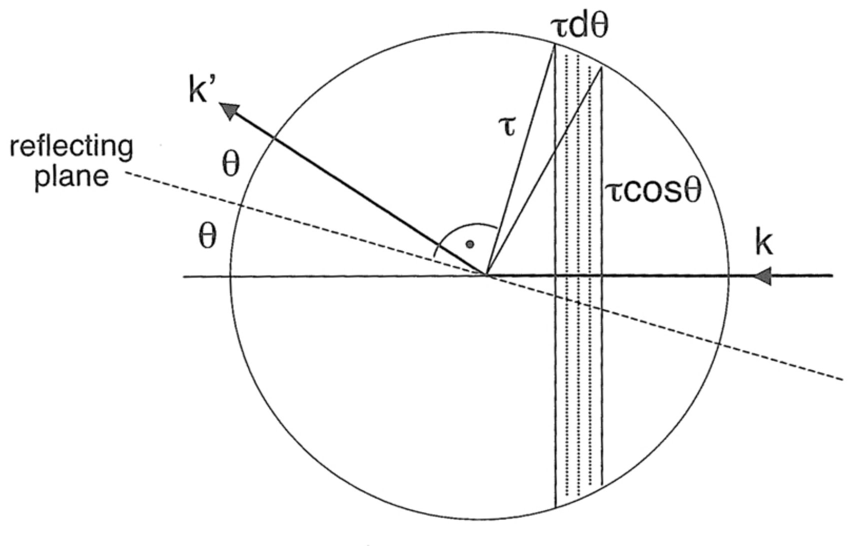


Figure 4: Sketch showing the fraction of crystallites satisfying the Bragg condition.

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