Physics with Neutrons I

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Exercise sheet 6 https://wiki.mlz-garching.de/n-lecture06:index

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1. Instrument Resolution I

For neutron scattering experiments, atoms can be well described by delta functions. So far, we have seen that Bragg scattering from a perfect (and sufficiently large) single crystal gives rise to very sharp intensity peaks which are again well described by delta functions. However, in experiment these sharp peaks always have a finite width and have the shape of peak functions (such as Gaussian or Lorentzian distribution). This is no effect of real samples, but of the experimental setup. The two main contributions to this broadening are:

- The incident beam direction. A neutron beam is never perfectly parallel, but has a finite divergence.
- The neutron wavelength. Both, monochromators and velocity selectors let through a certain spectrum of neutrons.

Consider a 1D experiment where we look at scattering under small angles. The incident neutron beam is collimated in a 20m long flight tube with 10mm diameter apertures and the wavelength is determined by a velocity selector. The selector has a triangular shaped spectrum with $\delta\lambda/\lambda = 5\%$ (i.e. the FWHM of the distribution has the value of $\delta\lambda$).

- For incoming neutrons, what is the shape and width of the angular distribution?
- How do angular and wavelength spread affect the scattering process?
- What is the shape and width of Bragg peaks finally observed?
- What changes if we consider scattering under large angles?
- Think about other possible factors influencing the shape of Bragg peaks in an experiment!

Solution

- For a simple 1D setup, the maximum angle under which neutrons can pass the collimation tube (from one edge of entrance to the opposite edge of the exit) is given by

$$\theta_{max} = \arctan\left(\frac{d}{l}\right) = \arctan\left(\frac{10\text{mm}}{20\text{m}}\right) = \pm 0.5\text{mrad} = \pm 0.029^\circ$$

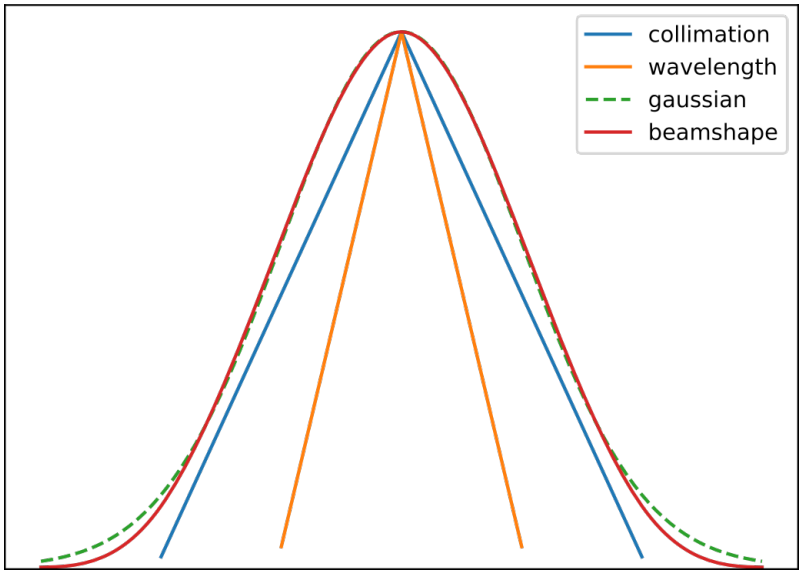
  For a simple flight tube, the relative intensity of all neutrons passing under a certain angle is restricted by geometric constraints (at which points of entrance can neutrons under a certain angle still pass through) and grows linearly from $I(\pm\theta_{max} = 0$ to $I(0) = 1$,i.e. a triangular distribution with FWHM= θ_{max}.

- For scattering under small angles the Bragg equation is:

$$\vartheta = \arcsin\left(\frac{\lambda}{2d}\right) \approx \frac{\lambda}{2d}$$

  meaning, that uncertainties in ϑ and λ both affect the equation linearly. However, while the angular spread is the same for all wavelengths and samples investigated, the wavelength uncertainty has a larger effect on scattering under larger angles (meaning smaller structures). In any case, the sharp delta peaks of Bragg scattering are broadened which is expressed by a convolution of the scattering pattern with the angular and wavelength distribution.

- The convolution of two triangular function with different widths is in mathematically easy, however, a set of case distinctions have to be made which turns out to be quite tedious. However, this step can be easily done by computation.
  Firstl, let us consider the angular spread due to wavelength uncertainty in numbers. For small angle scattering, a typical set of parameters would be $\lambda = 10$Åand $d = 0.1\mu$ m. This results in a scattering angle of $\vartheta = 5\text{mrad} = 0.29°$ and a uncertainty from the wavelengths $\Delta\vartheta = 0.25\text{mrad} = 0.0145°$. This is similar (factor 1/2) to the uncertainty due to collimation. The resulting Peak shape from convolving both distributions is shown below, with a measured HWFM$\approx 0.6\text{mrad} = 0.0348°$. The additional dashed green line is a Gaussian function illustrating the similar shape and the justification, why neutron beam shapes are often assumed to be Gaussian.



- We already saw, that for larger angles, the absolute uncertainty of the scattering angle due to a wavelength spread increases. Additionally, the small angle approximation in no longer valid. Both factors make small angle scattering more precise. (Which is why small angle instruments can use velocity selectors in the first place, instead of single crystal monochromators).

2. Instrument Resolution II

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 $2.2 \cdot 1^{-2}$% 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). The instrument resolution is given by $\Delta\theta/\theta = 9.5 \cdot 10^{-4}$. Since the unit cell is tetragonal, the according reciprocal lattice vector is:

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

We look at the specific Bragg reflex at $(hkl) = (002)$ 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 $d = 2\pi/Q$

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

which means that the peak shifts by

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

$$\leq \left(\frac{\Delta\theta}{\theta}\right)_{\text{instrument}} = 9.5^{-4}.$$

Here we used $\arcsin x \approx x$ for small x. (For typical neutron wavelengths $1\text{Å} \leq \lambda \leq 10\text{Å}$ the scattering angle is not necessarily small, however as we only look at a small relative change, the approximation is still good enough) Therefore, the effect cannot measured with this instrument.

3. Towards disordered systems

As the lecture will now go from Well ordered crystals to liquids and other disordered systems, take some time to play around with disorder in 1D atomic chains. Similar to the computations in exercise 3 on sheet 3, try out some different kinds of disorder:

- different atoms (with different scattering length) in perfect order, complete disorder.
- substitutional defects can be modeled by adding a small chance to have the wrong atom at each lattice point
- imperfect ordering can be introduced by adding a small random shift to each lattice position. this can be either individual for each atomic position, or cumulative.
- some binary alloys tend to cluster. this can be modeled by selecting the Atom for each lattice position randomly, but make it dependent on the previous position with a bias towards the same kind of atom.

This is no exercise with a fixed solution. It is rather meant to illustrate the effects of different kinds of disorder in crystals and how strong they are. Remember to compare them to scattering from liquids, once they are covered in the lecture!
If you have trouble implementing an idea in python, feel free to write me an email.

Solution

The following cases illustrate some examples of disorder and their impact on the scattering image. All computations have been done with a 1D chain of point-like atoms, with a set of $N = 500$ Atoms, a lattice distance $d = 6$Åand a neutron wavelength $\lambda = 1.9$Åwhere not stated otherwise.

- Case 1: Perfect crystal without any disorder for reference.
- Case 2: All atomic positions have a random individual displacement from their original position. The error dt is randomly chosen from a gaussian distribution with $\sigma = 0.3$Å.

```
for n in range(N):
    dt = np.random.normal(0 , 0.3)
    phi = 2 * np.pi*2 * (n * d + dt) * np.sin(np.pi * Theta / 180.) / Wl
    x += np.cos(phi)
    y += np.sin(phi)
I += (x**2 + y**2) / float(N**2)
```

- Case 3: This times, the error of each atom will be added up. The distance from one Atom to the next is still d (plus the error dt) but it is now a local and not a global order. In this case $\sigma = 0.12$Å.

```
dt = 0 for n in range(N):
    dt += np.random.normal(0 , 0.3)
    phi = 2 * np.pi*2 * (n * d + dt) * np.sin(np.pi * Theta / 180.) / Wl
    x += np.cos(phi)
    y += np.sin(phi)
I += (x**2 + y**2) / float(N**2)
```

- Case 4 & 5: For both cases the crystal structure is perfect, but the scattering length is randomly selected from tweo values for each Atom (0.5 or 1.0 for Case 4 and 0.0 or 1.0 for Case 5). These examples were already shown in previous exercises.
- Case 6: the following cases are a sketch of what happens in alloys of two atoms, which tend to cluster and which have a lattice distance depending on the kind of Atom.
  for Case 6, the first variety of Atoms has $d = 6$Åand a chance of 1/20 that the next Atom will be of the second variety, which has $d = 5$Åand a cance 1/7 to switch back to the first kind.

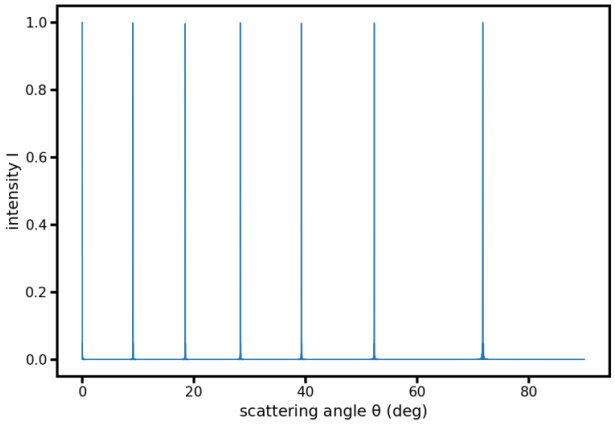
```
D = [6.,5.]
d = 0
m = 0
ch = [[0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,0,1] , [1,1,1,1,1,1,0]]

for n in range(N):
    m = np.random.choice(ch[m])
    d += D[m]
    phi = 2 * np.pi*2 * (d) * np.sin(np.pi * Theta / 180.) / Wl
    x += np.cos(phi)
    y += np.sin(phi)
```

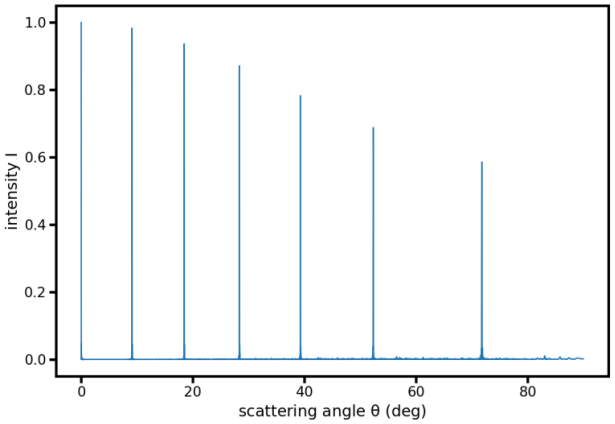
I += (x**2 + y**2) / float(N**2)

- Case 7: Both varieties of atoms have a switching chance of 1/20.
- Case 8: Both varieties of atoms have a switching chance of 1/7.
- Case 9: Both varieties of atoms have a switching chance of 1/20. the distance parameters are now $d = 6.0$Åand $d = 5.1$Å
- Case 10: Both varieties of atoms have a switching chance of 1/20. the distance parameters are now $d = 6.0$Åand $d = 5.95$Å

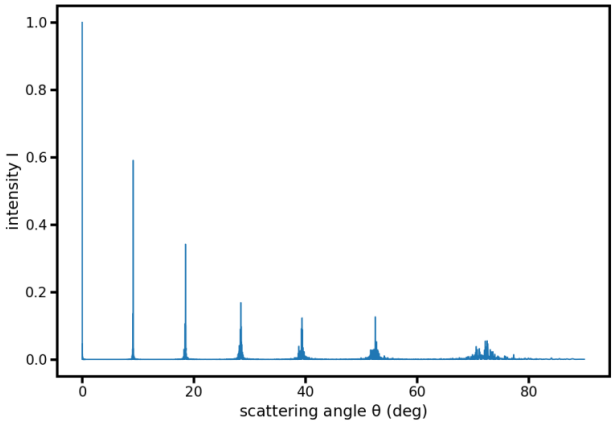
Case 1.



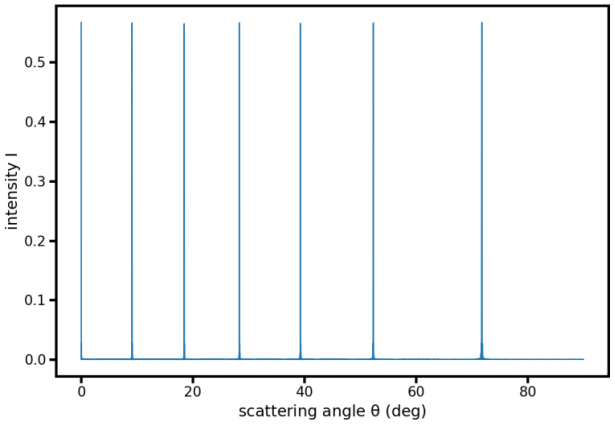
Case 2.



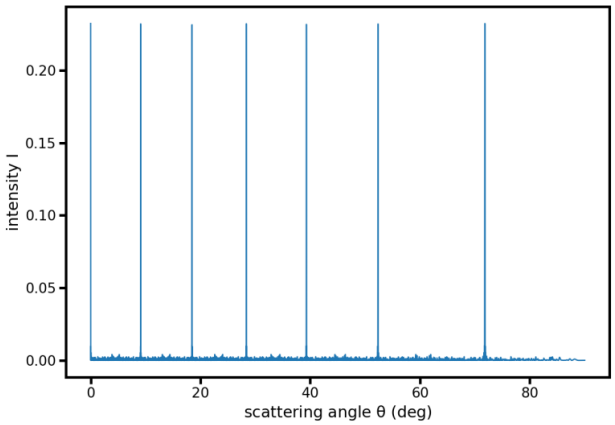
Case 3.



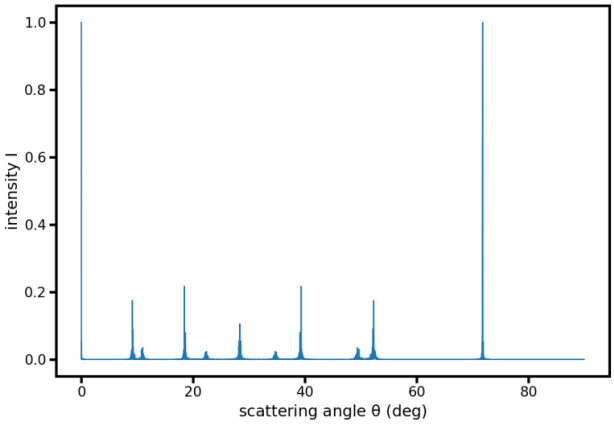
Case 4.



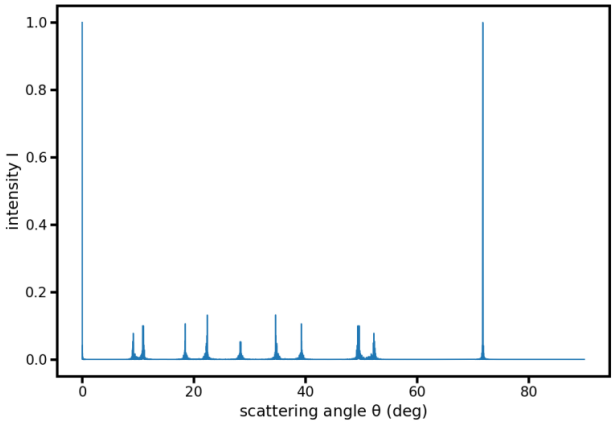
Case 5.



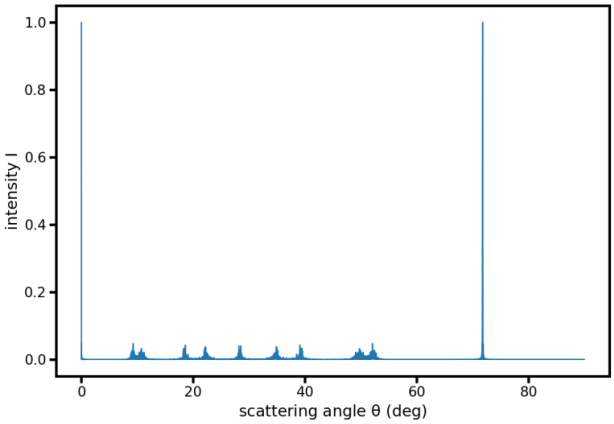
Case 6.



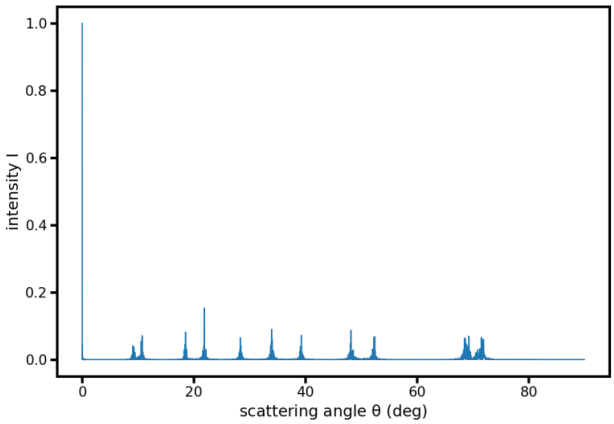
Case 7.



Case 8.



Case 9.



Case 10.