Ultra small angle scattering versus diffraction

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Abstract. In the case of ultra small angle (neutron or x-ray) scattering (USANS, USAXS) it may happen that structures under investigations are not fully coherently illuminated by the incident wave. Despite this fact interference effects are observed similar to SAS data. In this case the measured scattering patterns must be different interpreted. We propose a procedure to calculate and adapt such scattering patterns to experimental data.

1. Introduction

The investigation of large particles by means of small angle scattering is the domain of ultra small angle neutron and x-ray scattering, USANS and USAXS. Small angle scattering assumes a coherent illumination of the particles (structures) under investigation and it covers a q–range from $10^{-2}$nm$^{-1}$ down to $10^{-5}$nm$^{-1}$ corresponding to app. 50nm and 50µm particle sizes. USANS is performed with double crystal diffractometers (DCD), but also new “classical” (no DCD) instruments are planned or under construction to cover a range down to $10^{-4}$nm$^{-1}$. Therefore DCDs still keep the record of low momentum transfer measurements for USANS or USAXS and are used worldwide. The advantages of these instruments are the so-called decoupling of momentum transfer resolution from beam divergence and then their compact layout. Due to the high angular correlation of the beam between both crystals the transversal coherence length $\Delta_{c,\text{transv}}$ is much larger than of conventional instruments. For a DCD it is given by $\Delta_{c,\text{transv}} \sim (2 \Delta_{\text{Darwin}} \cdot k_{\text{transv}})^{-1}$ [1], [2], i.e. it depends directly on the Darwin width of the reflecting crystals, which is of the order of sec of arc or in q-space $10^{-5}$nm$^{-1}$. For classical small angle instruments one can estimate $\Delta_{c,\text{transv}} \sim 0.5$µm up to 5µm depending on the beam collimation and geometry.

2. Theoretical considerations

If the size of structures to be investigated becomes of the order of µm small angle scattering investigations must be performed with so-called double crystal diffractometer (DCD) [3]-[5]. The characteristic entity of a DCD is its Darwin width or Darwin – plateau (~µrad), and therefore its extreme small FWHM of a rocking curve, usually of the order of some µrad or sec of arc. This strong
angular-wave length correlation exists only between monochromator and analyzer crystal, and it is
used to detect tiny deviations of a beam scattered by the structure between these crystals. The beam
itself can have a much larger divergence (e.g. 0.5°) which does not (nearly does not) change the width
of the Darwin plateau and so the wavelength sharpness between both crystals. In the case of dynamical
neutron diffraction the so-called Darwin width or Darwin plateau $\Delta \theta_B$ is given by [1]

$$\Delta \theta_B = \frac{N \cdot b_c \cdot \lambda \cdot |F_{hkl}| \lambda}{2 \cdot \sin(2 \theta_B)} \cdot \frac{1}{2\pi \sqrt{|b|}} \cdot DWF$$

(1)

$N = \text{number of atoms/unit volume}$, $b_c = \text{coherent scattering length}$, $\lambda = \text{wave length}$, $F_{hkl} = \text{structure factor}$, $b = \text{asymmetry factor of the reflection}$ and $DWF = \text{Debye–Waller factor}$. If $\Delta k_y / k$ means the transversal $k$—spread of the beam one can write

$$\Delta \theta_B = \frac{\Delta k_y}{k} \cdot \frac{DWF}{\sqrt{|b|}}$$

(2)

and from this [2]

$$\Delta k_y = \frac{N \cdot b_c \cdot \lambda \cdot |F_{hkl}|}{4 \cdot \sin(2 \theta_B)}$$

(3)

The inverse of $\Delta k_y = 1/\Delta k_y$ yields some $\mu m$, depending on the reflection and Darwin width, and it is a
measure for the transversal coherence length $\Delta c,\text{transv}$. Using classical considerations based on the
Huygens-Fresnel principle one can calculate the coherent area of a beam given by the area of the first
Fresnel zone $A_{FZ}$ as

$$A_{FZ} = \frac{R \cdot r \cdot \pi \cdot \lambda}{R + r \cdot \sin(\theta)}$$

(4)

In the case of a DCD, the parameters $R = \text{distance source – object}$, $r = \text{distance object – detector}$, and $\theta$
must be defined. $\theta$ can be assumed to be $\sim 90^\circ$, $R$ and $r$ are of the order of meter or less. For x-rays
these parameters are well defined [6], [7] in the case of neutrons a non perfect graphite crystal as pre-
monochromator serves as virtual source and the calculation are a bit more complicated [8], however,
again one yields a transversal coherence length of the order of $10\mu m$. Contrary to these values we
measured with the V12b DCD with two different independent methods a transversal coherence length
of $\sim 90(20)\mu m$ [9], [10]. To overcome this discrepancy between theoretical transversal coherence
length $\Delta c,\text{transv}$ and experimental measured values we started a new series of experiments using now the
diffraction of neutrons by different thick Cu wires. The thicknesses (together with a coating) were
larger than 90$\mu m$ and depending on the interaction with the wire (fully coherent or partially coherent
interaction) different diffraction patterns were expected. In the case of partially coherent interaction
one can consider the resulting diffraction pattern to be built up by an incoherent sum of diffraction
patterns stemming from different parts of the diffracting object. The particular diffraction patterns can
be calculated on the basis of the Huygens-Fresnel principle taking into account - different to small
angle scattering treatment - phase relationships between partial waves in the sample (details are given
in [11]).

3. Experiments
The experiments were performed with the high resolution double crystal diffractometer V12b at the
BER II reactor of the Helmholtz Centre for Materials and Energy in Berlin (HZB). The instrument was
situated at the Nl3b neutron guide and had in the guide a graphite pre-monochromator with a mosaic spread of 0.5°. The mean wavelength $\lambda$ of the reflected neutrons was 0.533(1)nm, the intensity in front of the Cu samples (between the crystals) $5 \times 10^3$/cm$^2$.sec. The resolution function, i.e. the rocking curve, had a FWHM of 6.0(0.05)sec of arc (2.9x10$^{-5}$rad), in the case of the “Darwin reduction” [9] 1.6(0.05)sec of arc (7.76 x 10$^{-6}$ rad). These values correspond to a momentum transfer of $\Delta q = 3.4 \times 10^{-4}$ nm$^{-1}$ and $9.1 \times 10^{-5}$ nm$^{-1}$, respectively. The Cu wires (coated with an electric isolation) were mounted between the crystals as shown in figure 1.

The distances of the 130µm thick wires from each other were 340µm – 410µm, in the case of the 180µm wires it was app 300µm, however for each individual Cu wire the distances to the adjacent ones were much larger than the expected transversal coherence length $\Delta c_{\text{transv}}$. Because USANS scattering curves did not fit to the measured data the difference between different diffraction scattering curves basing on plane waves and Gaussian shaped waves having a finite lateral coherence length (FWHM) of 90µm (as was measured before [10]) were calculated assuming in both cases fully coherent illumination of the object. Figure 2 (a) and (b) shows the results, the agreement with experimental data was unsatisfactorily, it was worse assuming plane waves.

Figure 1 Layout of the V12b DCD instrument and set of Cu wires (thickness = 130µm + 9µm coating and 180 µm + 16µm coating). Si- monochromator and analyzer were seven-bounce channel cut crystal as in [9] (not shown here).

Figure 2 Scattering curves by (a) 130µm and (b) 180µm thick Cu wires. Incident wave was assumed to have Gaussian shape (FWHM = 90µm), red = experimental data, blue = theory
The next step was to consider that neutron waves traverse the object at different parts. Based on this assumption one can calculate the incoherent sum of scattering curves due to such interactions. In Fig.3 the incident waves traverses the object at three different paths, for the 130µm thick Cu wire 65µm apart from each other and 90µm for the 180µm Cu wire. The different distances were scaled to the object width, i.e. the wider the object the larger the distance to the adjacent wire. These results are shown in figure 3. The now achieved agreement is much improved and it underlines that the proposed theoretical treatment describes the nature of interaction much better than standard theory of small angle scattering. If the distance of the wires from each becomes close to the FWHM of the Gaussian shaped wave the resulting scattering patterns converges to the plane wave treatment [11].

![Figure 3](image-url)

Figure 3 Diffraction pattern calculated by means of incoherent superposition of diffracted waves. The agreement of experimental data with theoretical curves (blue) is much better than in figure 2.

4. Summary
We presented a procedure to explain USANS scattering curves by means of incoherent superposition of diffracted waves emerging from different parts of the object (structure). In the case that structures are larger than the lateral coherence width of the incident (neutron) wave one has to apply a different procedure than classical (U)SANS theory to calculated the correct diffraction pattern. The incoherent superposition of diffracted partial waves apparently fits much better to scattering data than standard theory can yield.

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