Progress on The Time-of-Flight Ultra Small Angle Neutron Scattering Instrument at SNS

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Abstract. We describe the Time-of-Flight (TOF) Ultra Small Angle Neutron Scattering (USANS) instrument at the Spallation Neutron Source, Oak Ridge National Laboratory (SNS, ORNL), which is now operating. The instrument, a double-crystal diffractometer (DCD), consists of a pair of triple-bounce symmetrically Bragg-reflecting Si(220) channel-cut crystals with a nominal Bragg angle of 70°, based on the Bonse-Hart multi-bounce concept. We briefly describe the instrument, present the results of measurements of double-crystal rocking curves, and exhibit a few early scientific results.

1. The TOF Ultra-Small-Angle Neutron Scattering Instrument at SNS

The Figure 1 schematically illustrates the SNS instrument [1] based on the Bonse-Hart principle [2]. The \(T_o\) chopper, guide, and premonochromator are not fundamental components but serve to reduce the background and crowding against neighbors. The scattering angle is greatly exaggerated in the figure. Figure 2 is a photograph of the instrument.

![Figure 1. The TOF USANS instrument](image1)

![Figure 2. A view of the SNS USANS instrument.](image2)

The beam at SNS beamline 1A from the 30-K hydrogen moderator, reflected by the 2-D focused premonochromator, enters from the lower left and strikes the triple-bounce crystal monochromator (small table at lower right). The reflected beam passes through the sample...
position (round table with boron nitride apertures). The scattered neutrons pass to the triple-bounce analyzer crystal (upper left) and are reflected from there to the detector. The detector (SNS 8-pack, staggered 0.5-inch diameter $^3$He proportional counters) rests in a massive shield (green, upper right). Stepping motors drive the analyzer angles with precision ~0.03 arc-sec. (This is about equal to the angular resolution of the best terrestrial optical telescopes.) The monochromator-analyzer assemblies rest on a vibration-isolated granite slab (dark platform). All are within a room with temperature held ±.2 C.

2. Single-crystal reflectivity

The single-crystal single-bounce reflectivity function, $R(\theta)$, is similar to the Darwin-Ewald function, with perfect reflection for small deviations of the reflection angle from the nominal Bragg angle and very sharp fall-off for angles greater than the half-width of the Darwin plateau, derived in terms of the (originally, x-ray) theories of diffraction [3,4],

$$\delta \theta_D = \frac{\lambda^2}{\pi \nu \text{cell} \sin(2\theta_B)} \exp(-W) \left| \frac{F_{hkl}}{k} \right|. \tag{1}$$

Atomic motions are accounted for in (1) by including a “temperature factor,” the Debye-Waller factor, $DWF(q) = \exp(-W(q))$, which is of Gaussian form in $q$.

It is convenient to express the reflectivity as a function of the dimensionless ratio $y$:

$$y = (\theta - \theta_D)/\delta \theta_D(\lambda), \text{ where } \theta_D(\lambda) = \sin^{-1}(\lambda/(2d)). \tag{2}$$

For $|y| \leq 1$, the Darwin and Ewald reflectivities are

$$R(y) = 1.0. \tag{3}$$

and for $|y| > 1$,

$$R(y) = \begin{cases} R_D(y) = \left[|y| - \sqrt{|y|^2 - 1}\right]^2, & \text{Darwin, thick crystals, and} \\ R_E(y) = 1 - \sqrt{1 - |y|^2}, & \text{Ewald, finite crystals} \end{cases}. \tag{4}$$

The reflectivity function for the triple-bounce channel-cut crystals is

$$R_3(\theta) = R^3(\theta). \tag{5}$$

The rocking curve in the SNS USANS instrument is the convolution of the two triple-bounce reflectivity functions,

$$R_{\text{rock}}(\Delta) = R^3 \ast R^3 = \int R^3(\theta + \Delta) R^3(\theta) d\theta \tag{6}$$

which is approximately a triangular function with perfect reflectivity at the peak and a full width at half-maximum (FWHM) equal to the full width of the Darwin plateau. The difference between the Darwin and Ewald functions is not significant here. The more accurate functions (4,5,6) are used in data analysis.

With no sample, when the second multibounce crystal is offset by angle $\theta_0$, the instrument samples the scattering at small angles near $\theta_0$. Ignoring the wings on the reflectivity functions, one has a triangular weighting function, the resolution function, which is the convolution of the two rectangular functions (6). The FWHM $2 \delta \theta_0$ of the triangular distribution, the rocking curve, as a function of $(\theta - \theta_0)$, is measured by scanning $\theta$ through
angle around $\theta = 0$. With a sample in place, the minimum value of $q$ that is accessible without the analyzer resolution function overlapping the response to the unscattered beam is $q_{\text{min}} = 4\pi \delta \theta / \lambda$, which is quoted as the instrument resolution of USANS instruments.

In operation for diffraction measurements, the measured scattered-neutron counting rate for each wavelength (orders separated by time-of-flight) as a function of the offset angle $\theta_s$ is a slit-smeared version averaged in the plane perpendicular to the scattering plane, a Kratky camera image. The nominal wave-vector change is $q_n = 4\pi \sin(\theta/2) / \lambda_n$ for each wavelength $\lambda_n$. The convolution-like form is not amenable to direct deconvolution methods.

Feigin and Svergun [5] discuss the data treatment in detail, based on known slit-smearing and wavelength distribution functions, which are specific to each instrument. For USANS this process is carried out by slit-smeared optimized cut-and-try modeling methods. A qualitative feature of the relationship between the point-image and slit-smeared measurements is that d-dimensional power-law $q^d$ pinhole-scattering functions, exhibit $q^{-(d-1)}$ dependence. That is, the slit-smeared function is one dimension lower than the pinhole function.

3. The Debye-Waller factor

For the $n^{th}$-order wavelength, the DWF is

$$DWF_n = \exp(-q_n^2 \langle u^2 \rangle / 3) = \exp(-B \sin^2 \Theta / \lambda_n^2),$$

(7)
in which $\langle u^2 \rangle$ is the mean-squared atomic displacement due to thermal motions,

$$B = \left(4\pi\right)^2 \langle u^2 \rangle / 3$$

(8)
and the factor 3 applies for single-crystal cubic materials. Butt, et al. [6], gives $B = 0.45 \AA^2$.

4. Measured rocking curves

In rocking curve measurements, for each wavelength the monochromator is fixed in orientation, and the analyzer crystal is rotated in small steps through small angles around the straight-through position. summarizes the most recent measurements. Figure 3 shows the rocking curves, representative of recent measurements for the lowest-order reflections, $1 \leq n \leq 6$: Table 1 and figure 4 show the fitted FWHMs.
Figure 3. Rocking curves for wavelengths (orders $n=1,2,3,4,5,6$) and fitted functions. Angle scales differ for the rocking curves for different orders of reflection.

Table 1. Measured Rocking Curve Widths

$FWHM_n = 2 \delta \theta_n, \lambda_n = 3.6 \AA/n$

| Order, n | Wavelength, \AA | FWHM, arc-sec |
|----------|-----------------|----------------|
| 1        | 3.60            | 5.306±0.008    |
| 2        | 1.80            | 1.259±0.003    |
| 3        | 1.20            | 0.516±0.0009   |
| 4        | 0.90            | 0.244±0.0008   |
| 5        | 0.72            | 0.128±0.0006   |
| 6        | 0.60            | 0.097±0.001    |

Figure 4. Rocking curve widths.

5. Conclusions

Resolution distorts measurements of the rocking curves, now under study. The effects on scientific measurements are small, but significant in the rocking curve measurements. We avoid detector dead time effects, but are developing improvements. Meanwhile, we introduce attenuating filters to reduce the counting rates when they are excessive. We are addressing the (thermal) stability of the instrument and the effects of vibrations on scattering measurements. When the instrument thermal environment is disturbed, angle calibration is lost, requiring time to recover. Vibrations driven by nearby rotating equipment (a neighbor’s T0 chopper) sometimes disturb the instrument; this problem has been corrected. None of these preclude scientific use of the Time-of-Flight USANS instrument, which is ready for business.
6. Results of early measurements

Figure 5 displays SANS and TOF USANS measurements on tetragonal- and hexagonal-phase FeS powders, showing also the slopes of the Porod plots.

Figure 5 SANS and USANS diffraction of tetrahedral and hexagonal FeS.

USANS provides the opportunity to investigate particles across a wide range of 0.1-1000 nm length scales. USANS and SANS data (a) show that in both phases, there are two general length scales. Figures (b) and (c) are SEM images. (S. J. Kuhn, et al., 2016)

Figure 6 shows Shear cell measurements on complex fluids under shear.

A new type of electrolytes for Li-ion batteries that exhibit a shear thickening response promises to prevent shorting between electrodes.

(unpublished, G.M Veith, B. Armstrong, and K. Browning, M. Doucet, J. Browning, R. Sacci, and B. Shen, 2017).

Figure 6. Shear cell measurements.
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