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NEW SMALL ANGLE NEUTRON SCATTERING (SANS) INSTRUMENT AT ORNL
USING A POSITION SENSITIVE AREA DETECTOR

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NEW SMALL ANGLE NEUTRON SCATTERING (SANS) INSTRUMENT AT ORNL
USING A POSITION SENSITIVE AREA DETECTOR*

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ABSTRACT

This paper describes the construction and operation of the small-angle neutron scattering instrument which has been set up at the Oak Ridge Research Reactor at ORNL. Motivation for work on this system to date has been the demonstration of the feasibility of the Borkowski-Kopp position-sensitive area detector and the use of pyrolytic graphite monochromating crystal pairs for small angle neutron scattering research. For a relatively modest cost, a serviceable experimental capability has been achieved, results from which are given in several papers at this conference. It is planned that some of the experience developed at this facility will be incorporated in a proposed large-scale apparatus at the High Flux Isotope Reactor (HFIR) and that this facility will continue in use for preliminary scattering tests.

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INTRODUCTION

The performance of small-angle neutron scattering experiments with useful K-space resolution and acceptable data collection rates at available neutron sources requires the use of a position sensitive neutron detector (Abend, Schmatz, Schelten, and Müller, 1970; Schmatz and Schelten, 1971; Schelten, 1972; Ibel, 1976). When an area detector is used with a long beam path geometry, pinhole collimation can be employed. The advantage obtained is that slit geometry collimation corrections are eliminated and one is in a position to measure scattering on an area detector which has non-cylindrical symmetry distribution such as precipitates or voids in a single crystal or fibrous polymer and biological samples. Resolution in K-space is improved by the use of long wavelength neutrons and multiple Bragg scattering is avoided when the wavelength is greater than the Bragg cut off limit. Beyond 4 or 5 Å the number of available neutrons from the reactor spectrum is greatly reduced and unless an operating cold source is available use of 4 to 5 Å wavelengths is both practical and recommended, since Bragg cut off and K-space resolution advantages are retained with adequate neutron flux. In addition, one may make measurements over a wider range of scattering space which may be required to achieve the desired real space structure resolution.

This paper describes the construction and operation of a small-angle neutron scattering instrument set up at the Oak Ridge Research Reactor (ORR) which was designed with the above described considerations. Motivation for work on this instrument has been the demonstration of two experimental features: (a) the use of the Borkowski-Kopp position

sensitive area detector (Borkowski and Kopp, 1968; 1975) and (b) the use of multiple pyrolytic graphite crystals as a monochromator for small-angle neutron scattering research. For a relatively modest cost a serviceable experimental capability has been developed which has demonstrated the success of both features. During the period of operation of this experiment the ORR was on a reduced intermittent operating schedule; nevertheless, the apparatus supported some of the Oak Ridge research work presented at this conference. It is planned to incorporate the experience developed at this facility into a proposed large-scale apparatus at the High Flux Isotope Reactor (HFIR) and to continue to use the facility for preliminary small-angle scattering investigations.

DESCRIPTION

A 20 cm by 20 cm area position sensitive detector which was built by C. J. Borkowski and M. K. Kopp as a prototype in the Instrumentation and Controls Division of ORNL was made available for this experiment. The apparatus was set up at beam port HB-6 of the ORR which has been used previously for a long beam path time-of-flight spectrometer. A data collection system with several communication peripherals is based on the use of a Hewlett-Packard 21MX computer. At present, the principal mode of data output is ASCII-coded punched paper tape which is then used for input into the IBM and PDP-10 systems for reduction and analysis. These data can also be processed on the x-ray small angle scattering system in the Metals and Ceramics Division (Hendricks, 1978) which is based on a MODCOMP computer. The compatibility between the neutron and x-ray system is assured by the fact that the two area detectors are identical in construction

except for gas filling. A schematic drawing of the instrument is shown in Figure 1.

Monochromator and Filter

At HB-6 a movable steel and concrete shield which formerly housed a rotating chopper was used to enclose the liquid nitrogen cooled beryllium filter and the pyrolytic graphite crystal monochromator. A rotating steel shutter was reworked and fitted with a steel collimator channel which tapered from a 2" x 2" opening on the core side to a 1" x 2" opening on the outside over 24". A 6" beryllium filter consisting of an assembly of 12-1/2" x 1/2" x 6" bars cadmium plated on the sides and contained in a 2" x 1-1/2" channel through an aluminum block was placed just outside the shutter. Liquid nitrogen filled a vacuum insulated stainless steel chamber containing the aluminum block. A thermocouple on the aluminum block monitors the temperature and a Brown chart controller activates a liquid nitrogen fill system in order to maintain the filter temperature. The filter delivers a transmitted flux of 7.3×10^6 neutron/cm²-sec to the monochromator position. The ratio of flux delivered by the cooled filter to that from the uncooled filter is 2.2. The beryllium filter is enclosed in a steel and benelex shield within the movable steel and concrete shield room.

The monochromator assembly (shown in Figure 2) consists of 3 pairs of pyrolytic graphite crystals. Each pair is set to diffract a beam from the beryllium filter through two reflections at approximately 90°. This arrangement was originally employed at the ORR by J. W. Cable in a low angle diffractometer in which 4.4 Å neutrons are scattered into a linear position

sensitive detector over a range of 3° to 30° . This double crystal scheme provides a beam which is parallel to but displaced from the reactor beam which has a strong fast neutron component. The wavelength in the second beam can be changed by adjusting the crystal pairs. Furthermore, the width of the wavelength slice from the filtered spectrum can be broadened by using several crystal pairs which are tilted with respect to one another. In the current experiment an inexpensive construction has been used in which the graphite crystals are permanently fixed into position on an aluminum mounting frame. The pyrolytic graphite crystals have a nominal mosaic spread of 1° which gives a high peak reflectivity at $4.74 \text{ \AA}$ ($2\theta_M$ equal 90°). In order to use the available long beam path, a rather small displacement of $2\text{--}3/4''$ between the monochromatic and reactor beams was selected. The monochromated beam emerges through a $1''$ diameter hole through $24''$ of a steel/masonite laminated reactor beam shield. The three pairs were fixed to monochromate neutron beams through 88° , 90° and 92° ($2\theta_M$) respectively. The angular range of high reflectivity in each of the crystals is on the order of $1/4^\circ$. Therefore, small misalignments in the crystal are accommodated. An improvement of more than 20% could be achieved if adjustments could be made on each crystal.

Monochromated intensity for the arrangement described here will be proportional to the square of monochromator crystal peak reflectivities and to the monochromatic wavelength spread. The through-put efficiency of the double monochromator approaches 95% as determined by measured and calculated peak reflectivities (Riste and Otnes, 1969). In this respect there is an advantage over velocity selectors in which a fraction of the neutrons are removed by the rotating vanes and the collimator effect

(Brugger, 1965). However, the wavelength spread which is accepted in a high collimation small angle scattering system in the double monochromator is very small (0.2%) compared to that in a velocity selector ($\sim 10\%$). This necessitates the use of multiple crystal pairs. For an equivalent K-space resolution, one can afford to relax the geometrical resolution in the monochromator system in order to gain some intensity (Schmatz, Springer, and Schelten, 1974).

Detector System

The principles governing the RC position encoding of the Borkowski-Kopp detector are described in detail in several publications (Borkowski and Kopp, 1968; 1969; 1970; 1972; 1975; 1978). The area detector used in this experiment consists of a 20 cm x 20 cm anode grid plane, an x-coordinate cathode grid plane 3 mm above the anode and a y-coordinate cathode grid plane below the anode and a 5 cm drift field made up of 4 wire grid planes above the anode. The anode is held at 1.8 kv and the cathode grid planes are at -500 v. The drift field maximum voltage is -5 kv. The cathode grid planes are made of a continuous 75 micron stainless steel wire strung in a serpentine manner with a 2 mm spacing between wires on a frame having beryllium copper leaf springs for each wire which keep the wire segments in tension. A special low noise preamplifier (Kopp, 1971) is connected to each end of both the x-coordinate and y-coordinate cathode wires. The wire planes are mounted in an aluminum chamber which is filled with 400 mm of ^3He which is the neutron detecting element, 850 mm of Xe to cut down H and ^3T ion track lengths, and 50 mm CO_2 for electron quenching for a total fill pressure of 2 atm. The

detector window was 0.100" thick. With this gas filling the detection efficiency is 60% and position determining resolution is approximately 5 mm.

The counting electronics used with this detector are shown diagrammatically in Fig. 3. The signals coming from the preamplifiers are processed by the main amplifiers and passed on to cross-over timing units which set up appropriate delay signals. Each x and y set of electronics is essentially a time analyzer which converts the detected neutron pulses to time signals proportional to x and y. The timing information originating from the ends of each cathode wire are converted into time differences which upon conversion in a TAC becomes a voltage amplitude. At the same time, the signals from all four cathodes are summed and processed through a conventional voltage discriminator which rejects the gamma background. The discriminator gates in the voltage from the TAC's to the ADC through the interface system, which will be described later.

Once the counting electronics are set up which mainly consists of setting the main amplifiers so that the neutron signals are amplified without position dependence, adjustments are needed in the Timing SCA's and the Time-to-Amplitude Converters. Adjustment of the amplitude range in the TAC's varies the position-amplitude proportionality and it must be set so that position conversion in the Analogue to Digital converter (ADC) has adequate sensitivity. As this multiplicative factor is changed it is necessary to shift delay settings so that the events on the useful area of the detector fall in the range of the x and y ADC's. On-line scope display greatly facilitates instrument adjustment procedures. Finally,

the discriminator voltage window is generally opened to 10 V and the base line is set to the edge of the low voltage (gamma ray) noise.

A problem encountered after a preliminary gas filling was a loss of energy discrimination due to water vapor and oxygen contamination. Because of ion cloud attachment, these contaminants spread the ion charge spectrum and the gamma ray and neutron pulses badly overlapped. Periodic checks for degraded energy discrimination may be necessary in a system that has not been carefully cleaned or where there are virtual or actual vacuum leaks. A detector gas cleaning system has been set up (Kopp, 1977) so that periodic detector rejuvenation can be done conveniently. Improved detector chamber design may eliminate this problem.

System Geometry and Resolution

The system geometry was adopted primarily on the basis of accessible floor space and available flight tube equipment. A seven meter evacuated tube of 8" diameter was placed between a fixed specimen position and the shield housing for the beryllium filter and the graphite monochromator. Two flight tubes for the scattered beam were made; one is 4.7 meters in length and 38 cm in diameter. Both are evacuated by a mechanical pump. The initial approach of using relatively inexpensive separate experimental components dictated that the detector be enclosed in a parafin/boron carbide shielding structure with limited frontal access for scattered neutrons. At present, the incident beam stop is positioned on the external face of the detector shield (approximately 20 cm from the detector window). Additional shielding is placed at the specimen position to help cut down on the general background coming from the reactor. Air scattering

originates over a 50 cm to 80 cm length at the specimen position and a 25 cm length immediately in front of the detector.

Geometrical collimation and resultant resolution are fixed by the incident and scattering path lengths, collimation apertures at the monochromator and specimen positions, and the detector position resolution. The detector resolution was estimated from the half intensity width of a beam projected on the detector from a 1 mm diameter hole in a boron carbide mask at the specimen position. The half intensity width was 10 mm and was taken to be the square root of the sum of the squares of the geometrical cross fire at the detector and the detector resolution. The incident path length was 730 cm, the first collimation aperture diameter was 2 cm, and the path length from the specimen position to the detector was 120 cm. A detector resolution of 11 mm is calculated.

The resolution and collimation characteristics of the present apparatus are illustrated in Table I. The K-space resolution is calculated from the relationship (equation 10) in Schmatz, Springer, and Schelten (1974) and the minimum and maximum K-values, K_{MN} and the K_{MX} from the beam diameter D_p projected onto the detector which is calculated from simple ray optics. The wavelength is $4.74 \text{ \AA}$ and the relative wavelength spread $\Delta\lambda/\lambda = 0.03$.

The calibration of the channel position with respect to detector position was done with a cadmium mask placed in front of the detector which was perforated with 2 mm holes spaced 4 cm apart on a 16 cm x 16 cm grid. The shielding was removed from the specimen position so that the

monochromated beam could flood the detector face. Figure 4 shows the pattern of the mask displayed on the Tektronic oscilloscope. The peak positions were determined from x and y centroids. In a 64 by 64 channel array deviations from position linearity did not exceed 0.3 channel numbers which in this instance represents 0.8 mm in real position, and is much less than the spacial resolution of the detector.

Data Collection System

The timing information from a specific pulse provided by the x and y channel electronics is delivered to the ADC if the energy discriminator determines the event is an acceptable neutron pulse. The ADC's convert the times into digital data and delivers them to the interface. The interface reduces the event into a single 10 or 12 bit word with the x and y addresses in the two halves, disables the gate to the TAC's so that no other events can be accepted, and sets an interrupt flag in the computer. The interface also contains a prescaler, overflows from which are recorded in channel zero to be used by the computer to time the data accumulation period. This prescaler can be fed by either an internal clock oscillator or by an external monitor counter mounted just off center of the incident beam. The interface disables the TAC's whenever the prescaler overflows or an ADC input has been accepted so that no other events are allowed for the approximately 30 μ s needed for processing of the data by the computer.

The computer adds the word from the interface to the correct starting address in the memory for the data region desired and increments this location to record the count. The software checks for overflow and can

accommodate channels containing 2^{16} , 2^{20} , or 2^{24} counts if sufficient memory is available. The software also increments and checks channel zero for each prescaler overflow to accumulate data for the desired time (or total monitor count). At the end of processing, the computer generates a reset pulse which enables all the gates so that the next event can be accepted. The memory of the computer is currently 16 K 16-bit words but this will be doubled in the near future. Data accumulation is accomplished under software control but direct memory access capability is present if required for higher counting rates.

Only a limited capability is available for on-line data reduction at present but the increase in memory will facilitate development of data analysis programs. The data can be recorded on IBM compatible magnetic tape for later use but, more usually, it is punched out on a high-speed paper tape punch and transmitted to the Laboratory's PDP-10 time sharing computer system where it is available immediately on the PDP-10 or for subsequent transmittal to the IBM 360 computer for detailed analyses. Since a run consists of 1024 (32×32) or 4096 (64×64) numbers, some form of computer analysis is required and background corrections, radial averages, and contour plots are routinely made of virtually every data set.

The on-line scope provides a method of displaying the data as it is being accumulated either in the form of intensity vs position of any desired x or y slice through the array or as a contour shaded map of the array with intensity shown as alternating bright and dim areas (See Figure 4). The on-line high speed teletype is used to input three letter commands with their parameters to the computer for controlling various

operations of data accumulation, data computation, input, output, etc., and also functions as an output device. A high-speed paper tape reader is available for input of programs and previously collected data sets.

SYSTEM PERFORMANCE

In its present form the SANS apparatus is useful in the study of high cross section scattering problems. In most studies to date background corrected scattering can be measured in a range including about three decades of intensity. The incident beam current measured by the detector through a 1 cm aperture at the specimen position is approximately 80,000 neutron counts per minute. Background count rates for a 64 x 64 channel arrangement are 0.001 counts per minute per channel with reactor down, 0.08 counts per minute per channel with cadmium over the detector (reactor up) with the detector set at approximately 1 meter from the specimen and an empty cell (no specimen) background of 0.4 counts per minute per channel. In many problems one can take advantage of radial averaging of the area detector data and thereby increase the statistical precision of the scattering function determination. Massive shielding will be required to reduce the background to levels where scattering intensity can be reliably measured in a range of more than 4 decades.

Accurate determination of the detector efficiency over its area has been difficult. In Figure 5 the intensity measured in a fixed 1.5 cm beam is plotted as a function of detector displacement. The apparent efficiency does not vary by more than 10%. However, in some scattering experiments there is an indication of efficiency variation on a small scale which may be attributed to nonuniformity in cathode wire spacing which causes

variation in gas multiplication. So far, we have not found a scattering standard of sufficient uniformity and scattering strength to make convenient detector calibrations.

Examples of the kinds of data that can be collected on the SANS system are illustrated in Figures 6 through 8. In Figure 6 is shown the Tektronik scope display of the scattering from a graphite block 8 mm thick. The pattern was collected in less than 1 hour with the 1.2 meter scattering path and the scattering is attributed to microvoids in the sample. The scattering is spherically symmetric and it is estimated that with radial averaging of the pattern, a 5 to 10 minute collection time is all that is required to achieve an analyzable data set. The advantage of the area detector in the study of non-symmetric scattering problems is illustrated in Figure 7 which shows a contour plot produced after processing of the collected data from a Cu-8%Ti alloy quenched and aged to produce an over-aged spinoidal structure. The sample was large grained so that the scattering from the composition fluctuations is measured in only a few grains which were properly oriented for diffraction. Figure 8 shows a contour plot of smoothed intensity data from a single crystal of aluminum in which faceted voids were produced by radiation damage. In this instance the tails of the intensity function which are perpendicular to the octahedral faces of the void are clearly seen. The detector was set at 120 cm from the specimen so that a sufficient range in K-space was covered by the detector to pick up the faceting effects. Such effects are at an intensity level of two to three orders of magnitude lower than the small-angle scattering limit for the void scattering.

SUMMARY

A SANS apparatus has been assembled for a relatively small cost which has demonstrated the usefulness of the Borkowski-Kopp area detector and the multiple pyrolytic crystal monochromator. Although the apparatus was constructed with little attention given to geometric optimization and background scattering suppression, a useful capability has been demonstrated and used to carry out research investigations. Several improvements are under consideration and include modification of the monochromator to increase the diffracted beam displacement from the reactor beam and to add more graphite crystal pairs with adjustments for intensity peaking. A shielding scheme which incorporates flexible detector positioning is under consideration. Resumption of full reactor operations will obviously lead to steady progress in developing the facility. Improvements in data reduction can be affected by expanding the core memory of the data processor in order to do more rough data reduction. Finally, utilization of the small-angle x-ray scattering system developed by Hendricks (1978) should prove beneficial from two points of view. First, many of the analytic tools have been implemented for data reduction of data which are virtually identical, and second, joint x-ray and neutron scattering studies evolve naturally from such interaction.

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Table 1. Resolution and Collimation Characteristics
of SANS Apparatus at ORR

	ΔK ($\text{\AA}^{-1}$)	K_{MN}	K_{MX}	D_p
	($\text{\AA}^{-1}$)	($\text{\AA}^{-1}$)	($\text{\AA}^{-1}$)	(cm)
High Resolution*	0.0019	0.0039	0.0350	2.32
Low Resolution**	0.0100	0.0300	0.1530	2.60

* Incident beam path $\ell = 730$ cm, scattered beam path $L = 480$ cm,
first aperture $d_E = 10$ mm, sample aperture $d_S = 10$ mm, detector
resolution element $d_0 = 5$ mm.

** Geometry as above except $L = 120$ cm and $d_E = d_S = 20$ mm.

FIGURE CAPTIONS

- Figure 1. A schematic drawing of the new SANS apparatus at Oak Ridge.
- Figure 2. A photograph of the double pyrolytic graphite monochromator assembly. The six shiny black rectangles (three at each position) are the graphite crystals.
- Figure 3. Block diagram of the electronic components of the SANS instrument.
- Figure 4. A photograph of the pattern displayed on the on-line scope for a mask of 2 mm holes in cadmium spaced 4 cm apart on a 16 cm x 16 cm grid used to calibrate channel positions with respect to detector position.
- Figure 5. Intensity for a fixed 1.5 cm beam vs position along y as the detector is translated through the beam. A variation in sensitivity of less than 10% is observed.
- Figure 6. A photograph of the on-line scope display of the scattering intensity from a graphite block containing microvoids. The data were collected at 1.2 meter scattering path for 1 hour.
- Figure 7. A contour plot of the non-symmetric intensity distribution from a Cu-8%Ti alloy specimen consisting of large grains. The sample was quenched and aged to produce an overaged spinoidal structure.
- Figure 8. A contour plot of smoothed intensity data from a single crystal of aluminum in which faceted voids were produced by radiation damage. The incident beam is parallel to a $\langle 110 \rangle$ crystal direction and the tails of the intensity function perpendicular to the octahedral faces of the voids are clearly seen.

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Irradiated Al Xstal $\vec{k}_i \parallel (110)$
Corrected for Background