

Performance of the Advanced Cold Neutron Source and Optics Upgrades at the NIST Research Reactor

*Robert E. Williams, Paul Kopetka, Jeremy C. Cook,
and J. Michael Rowe*

NIST Center for Neutron Research

On March 6, 2002, the NIST Research Reactor resumed routine operation following a six-month shutdown for facility upgrades and maintenance. During the shutdown, the original liquid hydrogen cold neutron source was removed, and the advanced cold source was installed. An optical filter was installed on one of the neutron guides, NG-3, replacing a crystal filter for the 30-m SANS instrument and the guide used between the chopper disks of the Disk Chopper time-of-flight Spectrometer (DCS) installed on NG-4 has been recently reconfigured. Additional improvements in the neutron optics of various instruments are being made.

The advanced liquid hydrogen cold neutron source performs as expected, nearly doubling the flux available to most instruments. The measured gains range from about 1.4 at 2 Å, to over a factor of two at 15 Å. Also as expected, the heat load in the new source increased to 1200 watts, but the previously existing refrigerator has easily accommodated the increase. With intensity gains of a factor of two in the important long wavelength region of the spectrum, the advanced cold source significantly enhances the measurement capability of the cold neutron scattering instrumentation at NIST.

The optical filter on NG-3 is also very successful; the 30-m SANS has an additional gain of two at 17 Å. A system of refracting lenses and prisms near the SANS sample position has made possible measurements at low Q (0.0005 \AA^{-1}) that were previously not feasible. The DCS has also seen additional intensity gain factors in excess of two for the majority of experiments and at short neutron wavelengths the gains exceed three. In addition, two new triple axis spectrometers will feature double-focusing monochromators in order to exploit the full size of the available thermal and cold neutron beam tubes.

The success of the advanced cold source and enhanced neutron optics contributed to the recognition of the NIST Center for Neutron Research as "the premiere neutron scattering facility in the United States" by the President's Office of Science and Technology.

Presented by:

Dr. Robert E. Williams
NIST Center for Neutron Research
100 Bureau Drive, Stop 8563
Gaithersburg, MD 20899
USA

robert.williams@nist.gov

INTRODUCTION

The NBSR is a 20-MW, heavy water cooled and moderated research reactor, located at the National Institute of Standards and Technology in Gaithersburg, Maryland. The facility is operated by the NIST Center for Neutron Research for the U. S. Department of Commerce. Its first criticality was achieved in December 1967. The reactor was operated at 10 MW from 1969 to 1985, when the power was increased to its design power of 20 MW. Its thermal neutron beams and irradiation facilities have been used for neutron scattering experiments in support of the condensed matter research program at NCNR, nuclear analytical chemistry applications, and radiation physics research. [1]

The designers of the NBSR also envisioned a cold neutron source. They included a very large, 55-cm diameter, cryogenic beam tube to house a heavy ice (D_2O) cold source, with two dedicated beam tubes, CTE and CTW, for cold neutron beams. The first cold source, however, was not installed until 1987, twenty years after the reactor startup. Since that time, a steady program of cold source development and innovative use of neutron optics for guides, filters, etc., has dramatically increased the research capabilities of the NCNR. The cold neutron guide hall was completed in 1989, and its first instruments were operational in 1990. A liquid hydrogen cold source replaced the D_2O source in 1995, providing a six-fold increase in intensity. That source was itself replaced by the advanced LH_2 cold source last year. This paper presents recent advances in cold source development and neutron optics at the NCNR.

THE FIRST LIQUID HYDROGEN COLD SOURCE

Although the D_2O cold source was a success, increasing the flux of cold neutrons in CTE and CTW by factors of 3 to 5 with respect to the thermal beams, it was quickly realized that a liquid hydrogen source had the potential for a gain of 20-25 with respect to a thermal neutron beam, for neutrons with wavelengths greater than 4 Å. In addition, a LH_2 moderator avoids the need to warm up the solid D_2O every two days to encourage the recombination of D^+ and OD^- radicals in a controlled manner [2]. Hydrogen was chosen as the moderator, rather than deuterium, because the required gas inventory is much smaller, and because the tritium production is negligible. Furthermore, Monte Carlo simulations, using MCNP [3], indicated that a 5-liter LH_2 source would be nearly as bright (about 70%) as a 35-liter LD_2 source. The simulations also indicated that the nuclear heat load would be less than 1000 watts. The Unit 1 LH_2 cold source was first operated in 1995 [4].

A description of the Unit 1 cold source has been presented at previous IGORR meetings, but a brief review is presented here. The 3.5-kW refrigerator, its instrumentation and PLC controls, the hydrogen condenser and expansion tank, and the insulating vacuum system were all unaffected by the installation of the advanced source. Only the cryostat assembly, located in the cryogenic beam port, was replaced. Since the axes of CTE and CTW intersect at a point 20 cm from the center of the source, the LH_2 annulus had to be very large to fully illuminate the guides. The moderator chamber (see Fig. 1) was a 20-mm thick spherical annulus of LH_2 , 320 mm in diameter. The volume between two concentric spheres of Al-6061 defined the annulus. A 200-mm diameter 'bubble' on one side of the inner sphere provided an exit hole through the LH_2 for the cold neutrons streaming toward the eight neutron guides. The inner sphere was entirely filled with hydrogen vapor because it was open to the annulus only through a small tube at the bottom.

The thermosiphon delivering LH_2 to Unit 1, which was very stable and reliable, was retained. Thermal hydraulic tests at NIST-Boulder demonstrated that at least 2200 watts could be removed in this manner. Liquid hydrogen from the condenser flowed by gravity into the moderator chamber, and the mixture of liquid and vapor produced by the 800-850 W heat load returned to the condenser via a concentric tube. This two-phase return flow resulted in very stable operation, driven by natural circulation at a saturation temperature of 20.4 K at the chosen operating pressure of 105 kPa.

Unit 1 was operated successfully for 35 reactor cycles between 1995 and 2001. MCNP calculations were used to predict its performance. The calculated energy spectrum, the cold

neutron gain, and the brightness all agreed well with the measurements. The calculations confirmed another observation made in the early weeks of operation: a reduction in operating pressure from 150 kPa to 105 kPa actually increased the flux of cold neutrons by about 5% at the longest wavelengths, even though the void fraction in the boiling liquid hydrogen had increased. This is because cold neutrons were scattered out of the beam while traveling through the 300-mm of hydrogen vapor in the inner sphere. The presence of the vapor degrades the source performance, which is evidence that the ortho fraction in the LH₂ remains above 50%, and completely dominates the scattering (para-hydrogen is nearly transparent to cold neutrons).

Safety has always been assured by requiring at least two barriers preventing H₂ from mixing with air. Each component was surrounded with a monitored helium containment. The LH₂ system was always open to a 2000-liter ballast tank, providing a passively safe shutdown in the event of a refrigerator failure. (A reactor power rundown protects the cold source from overheating if the thermosiphon is interrupted.) This system has also minimized hydrogen gas handling, and as a result, a negligible amount of hydrogen was lost in 6 years. The advanced source was built and installed with the same rigorous commitment to safety.

THE ADVANCED LIQUID HYDROGEN SOURCE

The installation of Unit 1 was nearly complete before a sufficiently sensitive MCNP model of the core was available to study the effect of variations in the cold source geometry on the reactor. It then became clear that additional D₂O surrounding the cryostat assembly would improve the coupling between the cold source and the reactor fuel. It was also obvious that a lengthy series of calculations was needed to study and optimize the coupling [5,6]. These simulations led to a complicated cryostat assembly, subject to many engineering constraints. A vacuum vessel must surround the moderator chamber, and a helium containment vessel, strong enough to withstand the design basis accidental detonation of liquid hydrogen and solid oxygen, surrounds them. The helium vessel determines the extent of the D₂O volume.

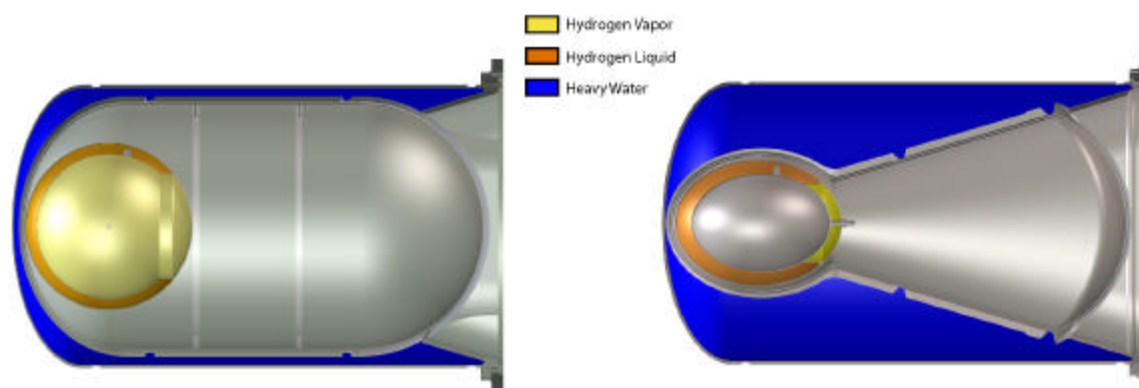


Figure 1. Comparison of the Unit 1 cryostat assembly (left) and that of the Advanced Cold Neutron Source.

The geometry of the advanced source, Unit 2, differs from Unit 1 in many key respects, as shown in Figure 1. The most important change is the addition of 60 liters of D₂O to the cooling jacket, partially surrounding the moderator chamber. This substantially reduced the size of the void through the reactor reflector, increasing the thermal neutron flux in the cryostat region about 40%. Unit 2 is an ellipsoidal, rather than spherical, annulus, with major axes of 320 mm, and a 240-mm minor axis in the transverse horizontal direction. This 5-liter annulus is between two nearly concentric Al-6061 ellipsoids; its average thickness is 25 mm. The center of the inner ellipsoid is offset 5 mm, however, so that the annulus is 30 mm thick near the core, and 20 mm thick at the exit hole. The inner ellipsoid is evacuated through a small vacuum port. Hydrogen vapor fills the elliptical exit hole (200 mm by 150 mm), but in Unit 2, the cold neutron beam passes through only 20 mm of vapor, rather than 300 mm. These changes in the moderator chamber added another 15-20% gain. An ellipsoidal

annulus provides three advantages. Because it has a smaller volume, more D_2O can be introduced in the cryostat assembly. It is also possible to increase the LH_2 thickness while maintaining the same volume as Unit 1. An elliptical shape is also desirable from a neutron optics standpoint; the neutron guides at NIST are all rectangular, most 60-mm wide and 150-mm tall, so they are still fully illuminated with the smaller cold source volume.

This March is the anniversary of the initial startup of the advanced source. Its nuclear heat load increased from 800 W to 1200 W as a result of the added mass of aluminum and the higher neutron flux. Since the refrigerator has a capacity of 3500 W, it was easy to increase its cooling capacity to accommodate the larger heat load. The cold source has operated very reliably during its first seven reactor cycles. Only one aspect of its behavior was unexpected. It was thought that with most of the H_2 vapor removed, there would be an increase in brightness as the system pressure increased, owing to the reduced void fraction in the LH_2 . Instead, the flux decreased as the pressure was raised, just like Unit 1. Even though there is only 20 mm of vapor in the exit hole, the loss from the vapor scattering still outweighs the gain due to the lower void fraction. Therefore, the source is operated at nearly the same pressure as Unit 1 (90-100 kPa).

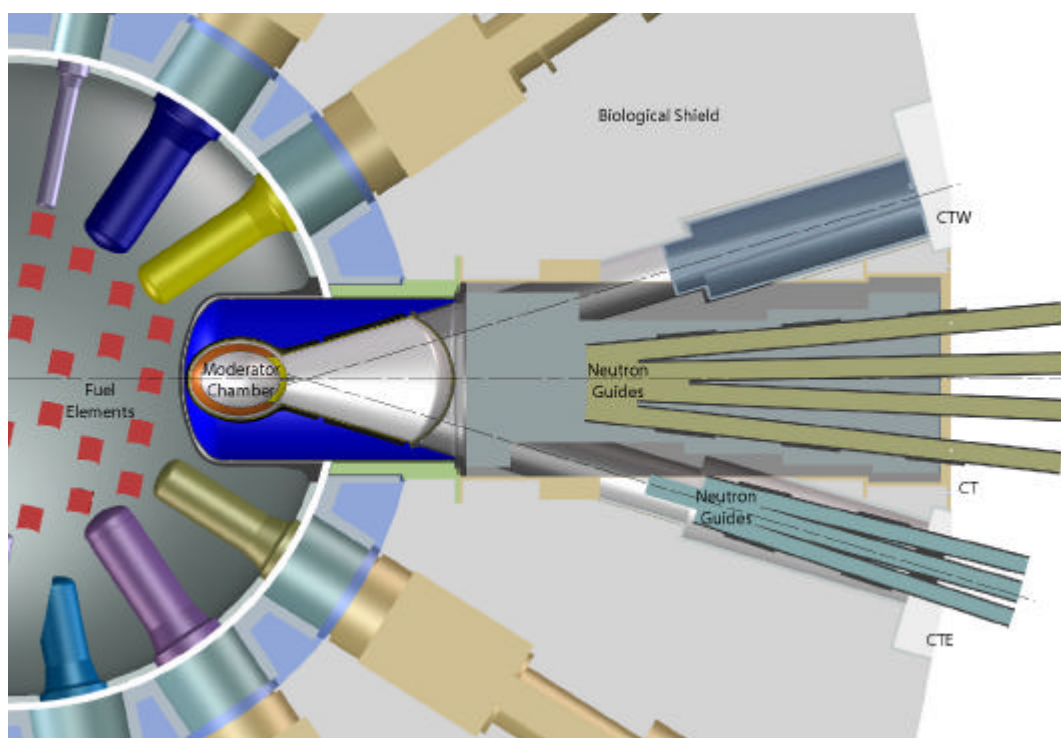


Figure 2. Plan view of the Advanced LH_2 cold Neutron Source. The cryostat assembly was inserted horizontally into the cryogenic beam port. The cold neutron beam ports intersect at a point near the exit of the thimble, about 20 cm from the center of the LH_2 vessel.

The neutronic performance of the new source was benchmarked by duplicating flux measurements made with the Unit 1 cold source at several Guide Hall instruments. Figure 3 is a plot of the cold source gain, defined as the ratio of intensities between Unit 2 and Unit 1, as a function of neutron wavelength. The figure includes the gain factors measured at three spectrometers and the gains calculated using MCNP. While the agreement is excellent, the measured gains were actually somewhat greater than predicted, especially at long wavelengths. This additional gain is likely due to the new guide sections replaced during the cold source installation, an effect not included in the MCNP models. The new cryostat design also substantially reduces the number of fast neutrons from the cold source resulting in improvements in instrument signal-to-noise levels.

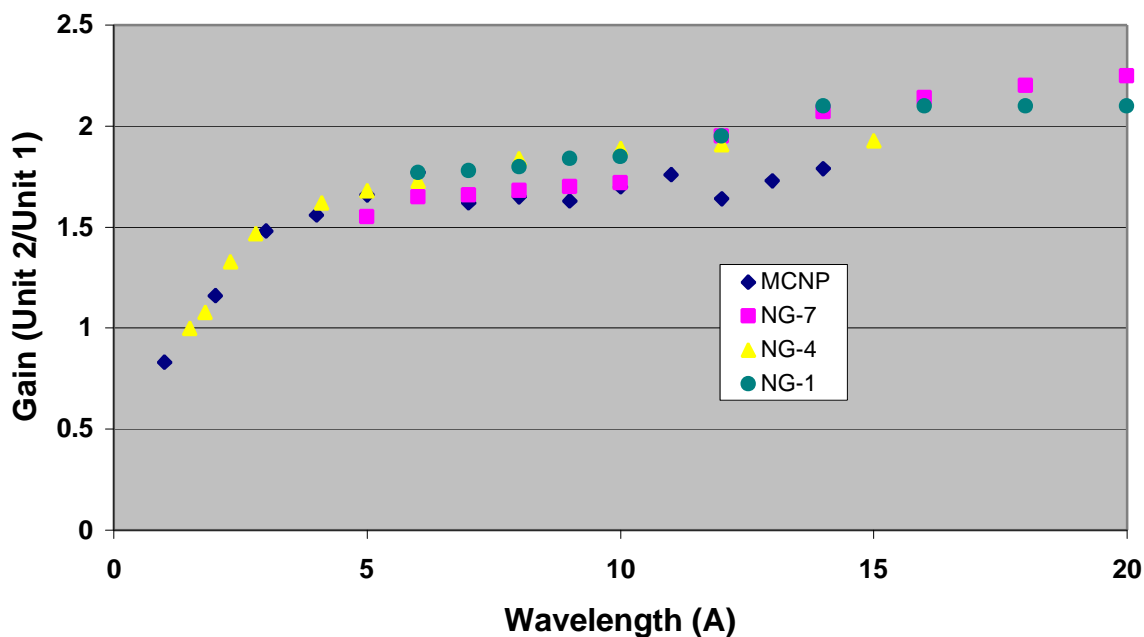


Figure 3. Measured and Calculated Gains of the Advanced Cold Neutron Source. Some gains exceeded predicted values because improvements were made to the neutron guide network.

ADVANCES IN NEUTRON OPTICS

In the last decade, the growth of the NIST Cold Neutron Research Facility has led to many advances in neutron optics, with applications to a wide variety of instruments. During the recent shutdown, the 30-m Small Angle Neutron Scattering (SANS) instrument on NG-3 was upgraded with the installation of an optical filter [7] and a system of refracting lenses and prisms. Later in the year, a section of the NG-4 neutron guide running between the chopper disks of the Disk Chopper time-of-flight Spectrometer (DCS) was reconfigured providing intensity gain factors between two and three for most experiments. These gains were obtained with no measurable increase in the instrumental resolution or background and will be reported elsewhere [8]. In the near future, two new Triple Axis Spectrometers will be built with doubly focusing neutron monochromators and massively parallel detection systems.

Small-Angle Scattering: An optical filter replaced the bismuth/beryllium crystal filter, used to reduce the background at the CHRNS (Center for High Resolution Neutron Scattering) SANS detector on NG-3. The crystal filter was replaced because it scattered some of the long wavelength neutrons from the beam and it eliminated all neutrons with wavelengths less than 4 Å. By excluding a line-of-sight between the entry to the SANS instrument and the reactor core, the optical filter eliminates the fast neutron background and heavily attenuates core gamma ray background. The cold neutrons delivered to the SANS instrument undergo an even number of reflections in the optical filter to emerge in the horizontal direction, but displaced 14.3 cm vertically, as shown in Figure 4. The vertical beam displacement meant that the entire 30-m SANS instrument had to be raised.

The optical filter design was refined with the aid of detailed Monte Carlo calculations of the angular distributions and spectral intensity of the transmitted neutrons [9]. Figure 5 shows the calculated and measured neutron flux gains at the sample due to the optical filter. The two

calculations are for the two supermirror reflectivity models shown in the inset. These results demonstrate that the optical filter transmission is about the same as the previous crystal filter for wavelengths around 5 Å, but becomes substantially better at longer wavelengths where absorption in the crystal filter becomes significant. In fact, the flux gain at long wavelengths approaches the estimated gain for the original guide design with the crystal filter removed. Because the optical filter produces a negligible increase in the divergence of the beam at the sample position, a requirement for the SANS instrument, the long wavelength flux increase is therefore close to optimum.

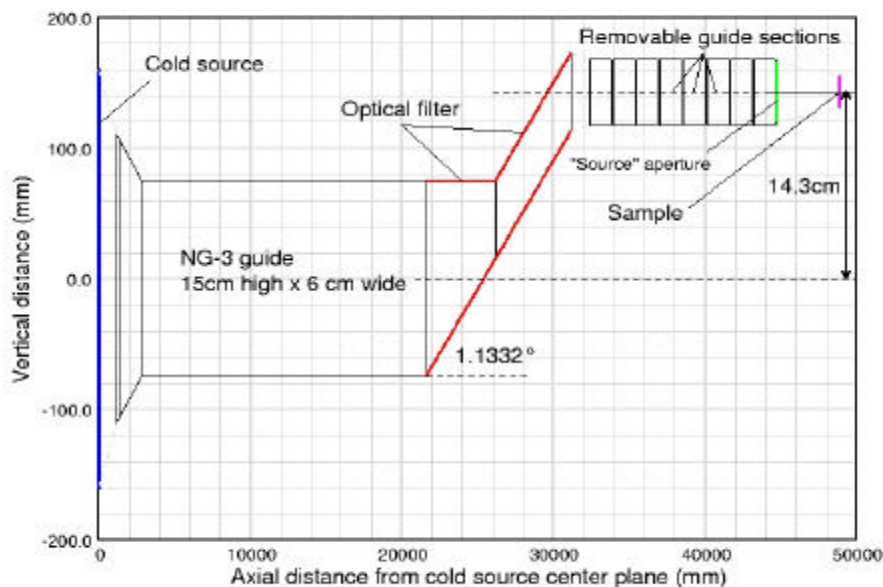


Figure 4. Schematic elevation of the NG-3 optical filter (note vertical scale is exaggerated for clarity). The inclined section shown in red is coated with supermirror with critical angle ~ 3.2 times that of natural nickel. The removable guide sections are in the pre-sample flight path of the CHRNS SANS instrument.

The optics for the CHRNS SANS instrument were further improved by installing a system of refracting lenses and prisms near the sample position to focus 17 Å neutrons onto the detector at its maximum distance, 13 m, from the sample. A similar lens system, consisting of 28 biconcave single crystals of MgF₂ for focusing 8 Å neutrons, had been in use on the NCNR's other 30-m SANS instrument on guide NG-7 for nearly two years [10]. The refracting power of the lenses increases with the wavelength squared, but so does the distance the neutron falls between the sample and detector due to gravity. The vertical spreading of the focus for a beam with a wavelength spread typical of a SANS instrument (about 10-15% FWHM) restricts the utility of the lenses alone to wavelengths of less than 10 Å. A new development, implemented on the CHRNS SANS instrument, was to follow the lenses with prisms that refract in the vertical direction to counteract the effect of gravity.

Figure 6 depicts the arrangement of lenses and prisms currently installed on the CHRNS SANS instrument. Seven MgF₂ biconcave lenses focus 17 Å neutrons at the detector, 13 m from the sample. For this distance a single prism with an apex angle of 161° cancels the beam spreading due to gravity *for all wavelengths* [11]. Such a prism, however, would have to be 250 mm long at its base to intercept the full beam height transmitted by the lenses (~ 2 cm). A more practical scheme, shown in Figure 6, uses a stack of two sets of prisms (each 30 mm long and 5 mm high with apex angle 143° to give the same anti-gravity effect. In this scheme, the bottoms of each prism are coated with Gd₂O₃ to eliminate surface reflections. The lens/prism system extends the minimum scattering vector to $Q = 0.00045 \text{ \AA}^{-1}$, nearly a

factor of two lower than pinhole collimation, and increases the scattered intensity 3 times because a larger area of the sample can be illuminated. There is actually an intensity gain of 48 compared to simply reducing the pinhole aperture to achieve the same minimum Q.

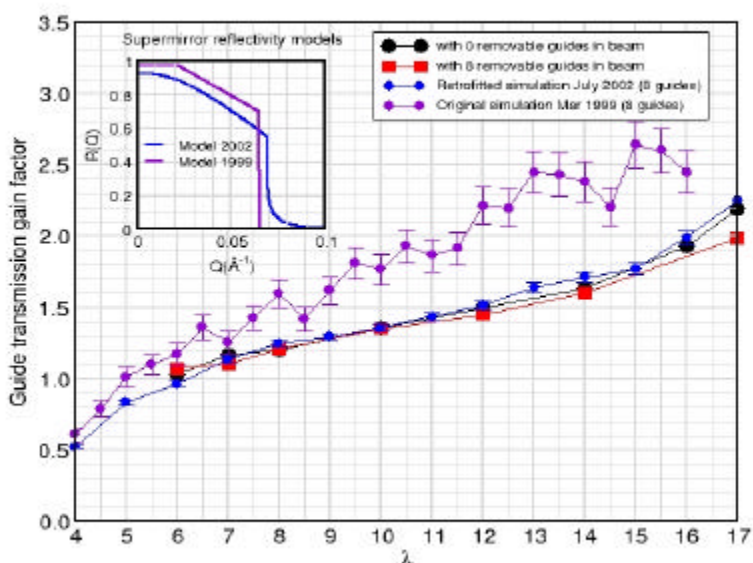


Figure 5. Transmission gains of the NG-3 optical filter. The black circles are measured with no removable guides in the beam. The red squares are measured with 8 removable guides in the beam. The violet circles are the original Monte Carlo simulated gain predictions (March 1999) that used the supermirror reflectivity model indicated by "Model 1999" in the inset. The blue circles represent simulated gains (July 2002) retro-fitted to the measured gain factors by refining the supermirror reflectivity model. The corresponding model, labeled "Model 2002" is also shown in the inset. The fitted model corresponds to "M=3.2" supermirror with $R(Q=0)=0.930$ and RMS surface roughness= 10\AA with $R(Q=0.069\text{\AA}^{-1})=0.55$.

Doubly Focusing Monochromator: For the past few years, NCNR, Johns Hopkins University, and the University of Maryland have been collaborating on a new concept for a low background, doubly focusing neutron monochromator (DFM), with a minimum of structural elements in beam. The devices will be at the heart of three triple axis spectrometers, located at CTW (cold neutrons), BT-7, and BT-9. The DFM envisioned for CTW, shown in Figure 7, consists of 21, variable-thickness Al blades, with 17, 2×2 cm pyrolytic graphite (PG) crystals attached to each [12]. For the cold neutron Multi-Analyzer Crystal Spectrometer (MACS), the entire assembly can be moved along the beam line and rotated through its full range of scattering angles, $35^\circ = 2\theta = 140^\circ$. The 1428 cm^2 beam can be focused onto a $2 \times 4 \text{ cm}^2$ sample, with intensities in excess of $10^8 \text{ n/cm}^2/\text{s}$ at 10 meV. Horizontal focusing is accomplished by independently rotating each of the 21 blades. Vertical focusing is achieved through application of an electronically controlled compressive force. Under this force, the carefully fabricated, variable thickness blades buckle into circular arcs with $0.9 \text{ m} = R = 10 \text{ m}$. The DFM is expected to have broad application in future spectrometers. The focusing performance was verified optically and confirmed a crystal placement accuracy of 0.15° and a repeatability of 0.03° (see also: <http://idg.pha.jhu.edu/DFM>).

The BT-7 Double Focusing Triple-Axis Spectrometer will be the first instrument at the NCNR that uses a DFM. In fact, the instrument will offer at least two such monochromators, one with copper crystals, and one with PG crystals. A DFM, coupled with the large area beam

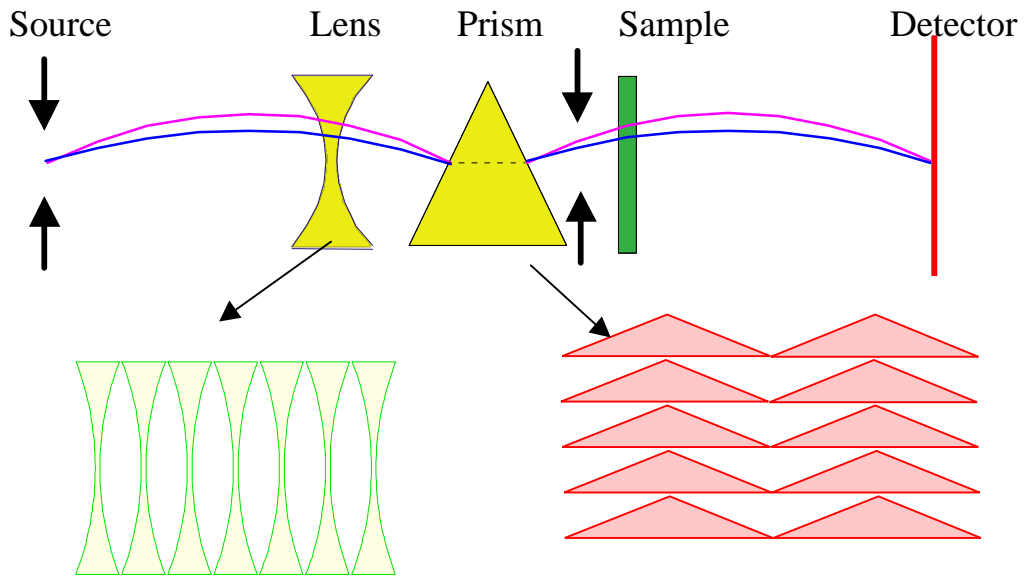


Figure 6. Schematic diagram of the configuration of lenses and prisms used on the 30-m CHRNS SANS instrument. The seven biconcave lenses focus 17 Å neutrons at the detector, 13 m from the sample, and the double stack of prisms refracts the beam vertically to cancel the effect of gravity.

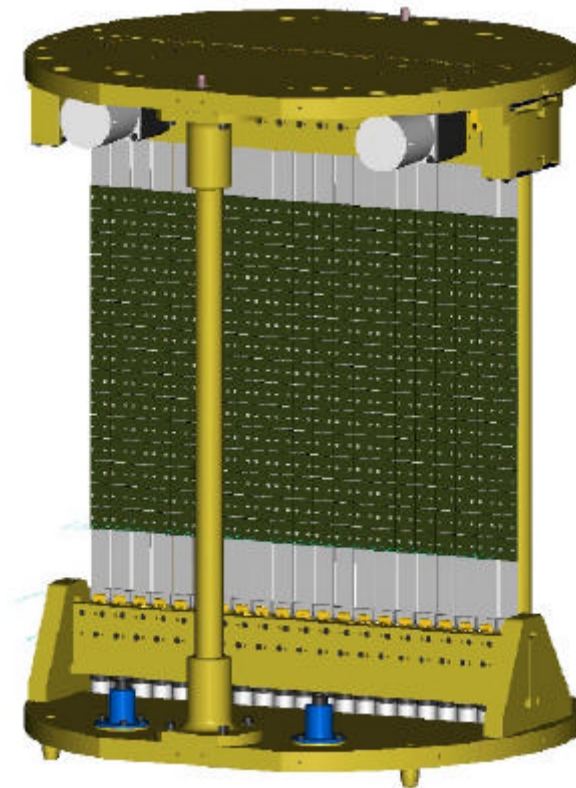


Figure 7. The low background, doubly focusing neutron monochromator for MACS (cold neutrons). Similar monochromators are planned for BT-7, and later, BT-9.

possible with a new in-pile shutter, will create neutron fluxes at the sample position one order of magnitude greater than previously available. In addition, the detector system of the new BT-7 will feature a horizontally focusing analyzer and for many detector configurations, the data collection rates will be increased by two orders of magnitude. More information on the upgrade of the thermal neutron instruments can be found at the NCNR web site: www.ncnr.nist.gov.

CONCLUSION

In the last decade, improved cold neutron sources have resulted in gain factors of 10 to 12 in the beam intensities available to all of the cold neutron instruments in the Guide Hall. In addition, many instruments have enhanced beam quality and intensity through the use of state-of-the-art neutron optics. The development of the DFM will revolutionize the venerable triple-axis spectrometer starting next year. Although the Spallation Neutron Source is expected to start operation in 2006, the NCNR is committed to continued improvements of its capabilities to best serve the neutron scattering community.

REFERENCES

1. R. L. Cappelletti et al, "Materials Research With Neutrons at NIST," J. Res. Natl. Inst. Stand. Technol. 106 (2001) 187-230.
2. R. E. Williams, J. M. Rowe, and P. Kopetka, "The Liquid Hydrogen Moderator at the NIST Research Reactor," Proceedings of the International Workshop on Cold Moderators for Pulsed Neutron Sources, Argonne National Laboratory, Sept. 29 – Oct. 2, 1997, J. M. Carpenter and E. B. Iverson, editors, pp. 79-86.
3. J. F. Briesmeister, ed., "MCNP – A General Monte Carlo N-Particle Transport Code," Version 4B, Los Alamos National Laboratory, LA-12625-M, Los Alamos, New Mexico (March 1997).
4. J. M. Rowe, P. Kopetka, and R. E. Williams, "Performance of the Liquid Hydrogen Cold Source," Proceedings of the Fifth Meeting of the International Group on Research Reactors, IGORR-5, November 4-6, 1996, Aix-en-Provence, France.
5. R. E. Williams and J. M. Rowe, Physica B **311**, 117 (2002).
6. R. E. Williams, P. Kopetka, and J. M. Rowe, "An Advanced Liquid Hydrogen Cold Source for the NIST Research Reactor," Proceedings of the Seventh Meeting of the International Group on Research Reactors, IGORR-7, San Carlos de Bariloche, Patagonia, Argentina, October 26-29, 1999.
7. J. R. D. Copley and J. C. Cook, Physica B **283** (2000) 386.
8. J. C. Cook and J. R. D. Copley, "A significant intensity increase for the Disk Chopper Spectrometer at the NIST Center for Neutron Research", to be submitted to Rev. Sci. Instrum.
9. "NCNR 2002: NIST Center for Neutron Research Accomplishments and Opportunities," NIST SP 993, p. 14, R. Cappelletti, ed. (2001).
10. S-M. Choi et al., J. Appl. Cryst. **33** (2000) 793-796.
11. E. M. Forgan and R Cubitt, Nuclear News **9(4)**, 25 (1998).
12. Smee, S. A. et al, "MACS Low Background Doubly Focusing Neutron Monochromator," Appl. Phys. A **75**, 3 (2002).