A low-temperature sample orienting device for single crystal spectroscopy at the SNS

T E Sherline, L Solomon, C K Roberts II, D Bruce, B Gaulin and G E Granroth

1 Neutron Sciences Directorate, Oak Ridge National Laboratories, Oak Ridge, TN, USA
2 Department of Physics and Astronomy, McMaster University, Hamilton, ON, Canada

E-mail: sherlinete@ornl.gov

Abstract. A low temperature sample orientation device providing three axes of rotation has been successfully built and is in testing for use on several spectrometers at the spallation neutron source (SNS). Sample rotation about the vertical (ω) axis of nearly 360° and out of plane tilts (φ and ν) of from -3.4° to 4.4° and from -2.8° to 3.5°, respectively, are possible. An off-the-shelf closed cycle refrigerator (CCR) is mounted on a room temperature sealed rotary flange providing ω rotations of the sample. Out-of-plane tilts are made possible by piezoelectric actuated angular positioning devices mounted on the low temperature head of the CCR. Novel encoding devices based on magnetoresistive sensors have been developed to measure the tilt stage angles. This combination facilitates single crystal investigations from room temperature to 3.1 K. Commissioning experiments of the rotating CCR for both powder and single crystal samples have been performed on the ARCS spectrometer at the SNS. For the powder sample this device was used to continuously rotate the sample and thus average out any partial orientation of the powder. The powder rings observed in S(Q) are presented. For the single crystal sample, the rotation was used to probe different regions of momentum transfer (Q-space). Laue patterns obtained from a single crystal sample at two rotation angles are presented.

1. Introduction

The need for a device capable of orienting and cooling a single crystal sample to temperatures significantly below 10 K on a time-of-flight instrument is well known in the neutron scattering community. A device capable of both of these tasks, which has been built and is in testing, will be described.

When performing single crystal experiments, the ability to align the crystallographic axes with respect to both the incident neutron beam and the detectors is necessary in order to fully probe Q-space and to take advantage of the symmetry of the detector bank. Though it is highly desirable to maximize the Q-space available for measurement, technical difficulties limit the ability to tilt the crystal out of the horizontal plane of the instrument. Typically, the orientation of a single crystal used in an inelastic neutron scattering experiment is known a priori, so only small out-of-plane tilts are needed to bring the crystal into proper alignment.

When considering measurements on a powder sample, concern arises that the individual grains may preferentially align along one crystallographic axis, creating a texture in the resultant scattering pattern. Rotating or oscillating the powder sample during the neutron measurement tends to average out any texture which may be present. Since this device has the ability to...
rotate from $0^\circ$ to $360^\circ$ and back, texture effects may be reduced or eliminated by oscillating between these two values.

2. Apparatus
The low temperature sample orienting device consists of a CCR [1] mounted vertically on a room temperature sealed rotary flange [2] providing $\omega$ rotations. Two piezoelectric actuated tilt stages [3] for $\phi$ and $\nu$ rotations with in-house developed encoders are mounted within an exchange gas can which is thermally anchored to the bottom of the CCR. The resolution of the encoders is sufficient to determine the angle of each tilt stage to better than $0.1^\circ$ over much of the angular range. Further developments will improve the performance of the encoders. An aluminum cold shield mounted to the nominally 50 K intermediate stage of the CCR surrounds the low-temperature cold head and exchange gas can. The entire assembly, as shown in Fig. 1, rests in a stainless steel thimble whose flange forms the upper seal of the sample vacuum vessel of the spectrometer, e.g. that described in Ref. [4]. The neutron beam, shown as a dashed horizontal blue line in Fig. 1, is 749.3 mm below the thimble flange. For operation on the tilt stage, mount to sample center distance is 52.3 mm while CCR mount to sample distance is 151.9 mm. The tilt stage is capable of carrying a total of 60 g.

![Figure 1. Schematic representation of the low temperature sample orientation device showing a) the thimble (arrow indicates the vacuum sealing surface), b) the CCR, c) the sealed rotary flange, d) the Al cold shield, e) the Cu thermal link and upper flange of the exchange gas can, f) the bottom portion of the exchange gas can, and g) the tilt stages. Note that everything below the sealed rotary flange is within the sample vacuum space. Dimensions are represented in mm. The dashed horizontal blue line represents the neutron beam.](image)

The thermal performance of the CCR has been tested both with and without a load and with two different heat shields. Results shown in Fig. 2 indicate that the addition of a thermal load increases the total cooling time from less than 40 minutes to well over two hours. Additionally, the base temperature increases from 3.1 K to 6.5 K and creeps up over time. Based on experience with other heat shield configurations on identical CCRs, this likely originates from a degrading thermal link between the upper and lower portions of the heat shield. Nevertheless, this device has been effective for measuring several samples [5].

3. Neutron Scattering Results
The rotating cryostat has been used on the ARCS [6] and SEQUOIA spectrometers at the SNS. The intensity vs. detector bank position measured using the rotating cryostat on two example systems are shown.
Figure 2. Two cooldown curves showing the thermal performance of the CCR. a) The black curve shows the cooldown of the CCR without any thermal load and using a solid thick walled cold shield. The red curve shows the cooldown of the CCR in the configuration shown in Fig. 1. b) and c) Expanded views showing the behavior of the final temperatures achieved for the two configurations.

Diffraction rings from a power sample of NaNiO$_2$ contained within a cylindrical Al can mounted via a Cu thermal link to the bottom of the CCR can clearly be seen in Fig. 3. The measurement was made using an incident energy of 60 meV while the sample, maintained at 5 K, was rotated from 0° to 360° and back in order to negate possible texture effects.

Figure 3. Intensity vs. detector bank position taken an the ARCS spectrometer for the powder sample NaNiO$_2$.

In a single crystal measurement on La$_{2-x}$Ba$_x$CuO$_4$ \( [X \approx 0.09] \), the pre-aligned crystal was mounted to the bottom of the CCR via a Cu thermal cold finger extension similar to that used in the powder experiment. While being maintained at 6 K, the rotating cryostat was used to scan through one plane of reciprocal space in order to obtain Laue patterns at two distinct values of \( \omega \) as shown in Fig. 4 and Fig. 5.

4. Conclusion
The low-temperature rotating cryostat has been successfully used on an operating beamline at the SNS. Observations of both diffraction rings from a powder sample and Laue patterns from multiple Q values in a single crystal sample have been made possible using this device. Further development of the tilt stages will enable more extensive single crystal investigations in the near future.
Figure 4. Laue image for La$_{2-x}$Ba$_x$CuO$_4$ [X≈0.09] in one rotational orientation.

Figure 5. Laue image for La$_{2-x}$Ba$_x$CuO$_4$ [X≈0.09] in a second orientation rotated 40° from the pattern shown in Fig. 4.

Acknowledgments
The authors would like to acknowledge Doug Abernathy, Matt Stone, and Mark Loguillo of the ARCS instrument team for providing the data and plots for both the powder and intensity plots. This submission was sponsored by a contractor of the United States Government under contract DE-AC05-00OR22725 with the United States Department of Energy.

References
[1] ARS Model DE-210S. Advanced Research Systems, Inc., 7476 Industrial Park Way, Macungie, PA 18062 USA. (610)-967-2120, http://www.arscryo.com.
[2] Thermionics Model RNN-600MS. Thermionics Vacuum Products, 231-B Otto Street, Port Townsend, WA 98368 USA. (800)-962-2310, http://www.thermionics.com.
[3] Attocube Models ANGp100 and ANGt100. attocube systems AG, Koenigstrasse 11aRGB, 80539 Muenchen, Germany. 49(0)89-2877800-0, www.attocube.com.
[4] G. E. Granroth, D. H. Vandergriff, and S. E. Nagler, Physica B 385-386, 1104 (2006).
[5] M. D. Lumsden et al., Phys. Rev. Lett. 102, 107005 (2009).
[6] D. L. Abernathy, Notiziario Neutroni e Luce di Sincrotrone 13, 4 (2008).