Performance of the Amorphous Materials Diffractometer "GLAD" at IPNS

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ABSTRACT

The Glass, Liquids, and Amorphous Materials Diffractometer (GLAD) at IPNS has now been commissioned and the first scientific measurements have been carried out. Details of the performance of this instrument, including background, calibration, resolution, data rate, data quality, and data reduction and analysis will be discussed.

I. Introduction

Until recently, neutron diffraction measurements of the structure of disordered materials at pulsed sources were carried out on diffractometers designed essentially for powder diffraction. While much work of high quality was carried out, these instruments fall far short of meeting the requirements for state-of-the-art measurements on disordered systems. Because of this, a few years ago a group of Argonne and university scientists decided to build a new instrument optimized for structural measurements in glasses and liquids at the Intense Pulsed Neutron Source (IPNS). The requirements for this instrument included a large Q range, a nearly continuous range of scattering angles extending to angles as low as possible, and a range of wavelengths extending to ones as short as possible; coupled with the need to maximize intensity while still preserving reasonable Q resolution. These demanding requirements have been met by the design of the Glass, Liquids, and Amorphous Materials Diffractometer (GLAD). This design and various novel features incorporated in the instrument have been described in detail elsewhere.[1-5] Therefore, only a brief description of the instrument is presented below, and the remainder of this article is devoted to discussion of the performance of this instrument.

II. The GLAD Instrument

Figure 1 presents a top view of GLAD. As shown in the figure, GLAD has two sample positions, an upstream position 9.0 m from the moderator for the highest resolution measurements and a downstream position 10.5 m from the moderator, which provides higher data rates at some sacrifice in resolution. Crossed converging soller collimators focused on the forward detector bank collimate the thermal and epithermal neutrons used by GLAD. With this collimation scheme the sample size does not contribute significantly to the resolution,[6] so samples up to ~2.5 x 2.5 cm^2 (1.25 x 1.25 cm^2) can be used at the upstream (downstream) position. A series of massive apertures upstream from the soller collimators defines the beam, and provides the primary collimation for those higher-energy neutrons (hundreds of eV and higher) for which the soller collimators become transparent. The last of these massive apertures

* Work supported by U.S. Department of Energy, BES, contract No. W-31-109-ENG-38.

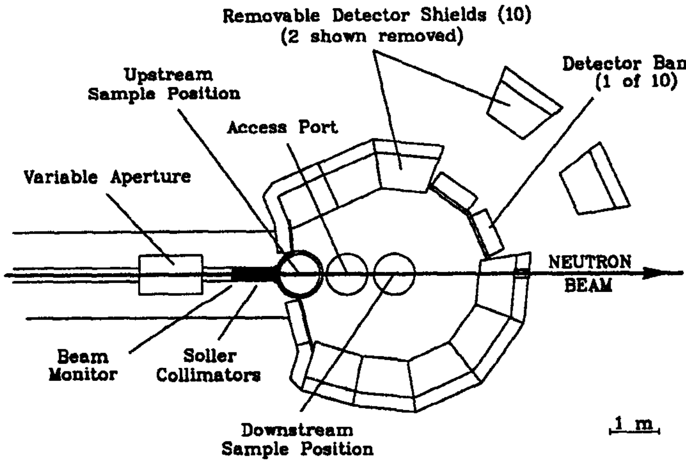

Figure 1. Top view of the GLAD instrument. Two of the ten detector shields are shown removed to reveal the associated detector banks in their positions just outside the evacuated flight path.

is adjustable and can be varied to control the beam size to minimize the amount of background when smaller sample sizes are used. The flight path is evacuated and the detectors are located outside this vacuum (Fig. 1), viewing the sample positions through thin aluminum windows.

GLAD uses linear position-sensitive detectors (LPSDs), which permit it to handle high data rates and to cover low scattering angles. GLAD contains 235 cylindrical aluminum-walled ^{3}He-proportional-counter LPSDs at the present time; eventually to be expanded to a total of 408. These detectors are mounted in 10 banks (Fig. 1); each bank contains from 38 to 53 LPSDs, depending on the location of the bank in the instrument. Each LPSD has a diameter of 1.27 cm and an active length of 60 cm, and contains 10 atm of ^{3}He plus appropriate stopping gases (mostly Ar). Each detector is encoded into 64 segments using charge-division encoding; the resulting full width at half maximum (FWHM) position resolution is currently measured to be ~14 mm. (These detectors and the mounting arrangements are similar to those used earlier on a steady-state small-angle diffractometer at the University of Missouri reactor[7] and on the SAN small-angle diffractometer at the KENS pulsed neutron source,[8] but the encoding and data acquisition electronics for GLAD are quite different from those used in either of these other two instruments.[3]) Segments of the LPSDs located in line with the direct beam have been successfully used to monitor sample transmissions[4] concurrently with the scattered neutron measurements which utilize the remainder of the segments of these same LPSDs.

The currently-installed detectors cover scattering angles from -9° to +18° and +37° to +95° for the upstream sample position and from -17° to +35° and +73° to +150° for the downstream sample position, with the negative sign implying scattering angles measured clockwise from the direct beam (at 0°) when viewed from above as in Fig. 1. Angular coverage is continuous over these ranges except for very small angles and for the small gaps between the detector banks. A beam attenuator, which protects the LPSDs near 0° from overload by the direct beam, obscures the scattering angles $2\theta < 1.7°$ when the upstream sample position is used and $2\theta < 3.4°$ when the downstream sample position is used. The locations of the detector banks on the two sides of the flight path are not symmetric (Fig. 1), so that the scattering angles not covered on one side of the flight path can be covered on the other.

An experiment on GLAD is started by first setting up a run file which contains information about how LPSD segments are grouped together and how the time-of-flight (TOF) histograms are organized. There are virtually no restrictions on how this is achieved; for example, detector segments may be grouped into Debye-Scherrer cones (preferable for isotropic scattering) and the time binning may be chosen with a channel width which is proportional to the TOF. In addition the run file contains calibration information concerning scattering angles, distances, etc. which are used in the subsequent data analysis. The run files are large (up to ~12 Mbyte), so managing the volume of data generated by GLAD presents some problems. A typical experiment results in several run files generated in a short period of time, so efficient programs have been developed for analyzing the data and compressing it to more manageable sizes. The raw run files are archived on secondary mass storage devices (currently 8-mm tapes holding about 2 Gbytes each) after the first-step analysis.

III. Resolution

The best experiments performed on GLAD use detector segments binned in common Debye-Scherrer cones with a user-defined width $\Delta\theta$. Most experiments performed to date have been in the downstream sample position using $\Delta\theta$ of 0.6°, corresponding to roughly 2 detector segment widths over a nominal scattered flight path of 1.5 m. In this operational mode instrument resolution is almost entirely geometric. This is illustrated using the FWHM of the strong (111) peak in the powder diffraction pattern of diamond to determine the resolution $\Delta Q/Q$, where the momentum transfer Q is defined as

$$Q = \frac{4\pi\sin\theta}{\lambda} \tag{1}$$

and

$$\frac{\Delta Q}{Q} = \frac{\Delta d}{d} = \Delta\theta\cot\theta \quad , \tag{2}$$

where d is the crystalline d-spacing. The results are plotted in Fig. 2 as a function of the scattering angle 2θ from 2-95°. The line through the data is 0.006 $\cot\theta$, where 0.006 is the average of the ratio $(\Delta Q/Q)/\cot\theta$ for each angle. Essentially identical curves are obtained using FWHM of other Bragg peaks - thus the resolution function is almost entirely geometric. Resolution improves by roughly a factor of two for samples run in the upstream sample position, but is still dominated by geometry.

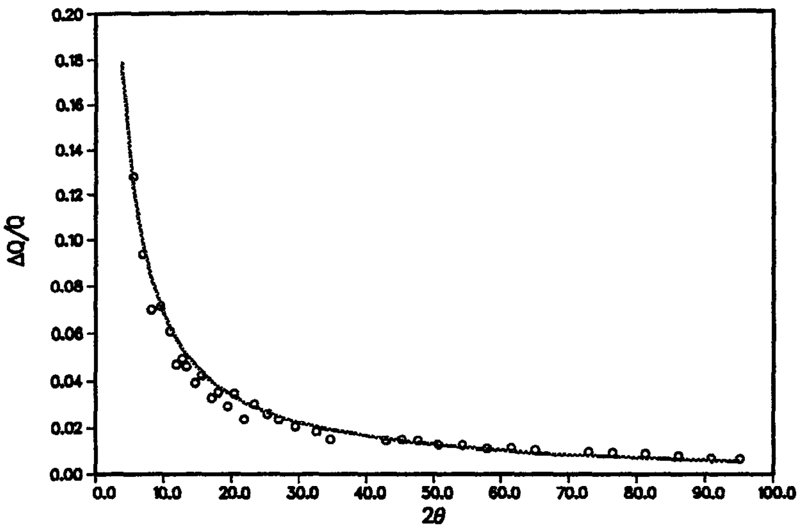

Figure 2. Resolution of GLAD, measured as the FWHM of the (111) peak in the powder diffraction from diamond.

The comparatively low resolution at low scattering angles places limits on the angles at which data are used in routine analyses. While some materials have relatively broad features at low Q, in general the best results for vitreous and liquid samples run in the downstream position are obtained when analysis is confined to data collected at scattering angles > 7°. At the same time, for liquids formed from light atoms and for nearly all oxide glasses, Placek corrections become quite large for values of Q corresponding to wavelengths of 1.5-2.5 Å. This places a practical limit upon the lowest reliable Q obtained from a sample in the downstream sample position of 0.2-0.3 $Å^{-1}$, depending upon the wavelength range appropriate for a particular sample. In the upstream sample position, the minimum reliable scattering angle decreases to 4°, and thus the minimum reliable Q decreases to roughly 0.1 $Å^{-1}$. Work underway is expected to improve the spatial resolution of the low-angle detectors to better than the size of a detector segment, in which case the minimum Q in the downstream sample position will improve by at least a factor of two. Even with the present set-up, however, GLAD provides relatively good overlap with the highest Q values obtained on conventional small-angle neutron diffractometers.

IV. Calibration

At present time-of-flight data is corrected to wavelength using a simple linear relation between t and λ. This is justified empirically in that addition of quadratic or higher-order terms to the equation had a negligible impact on χ^2 for wavelength-calibration data collected on the GLAD prototype.[5] Software is in place for performing wavelength calibrations, and efforts are underway to automate it. Detector segment positions are currently assumed to be equally spaced along LPSDs, with exactly the same active lengths. Flood patterns collected using a moderated PuBe source show that differences in counting rates from one segment to another - a potential rationale for determining detector segment boundaries - are within the counting error for each segment, as might be expected given the fact that the resolution at any point is almost twice the size of a detector segment. Nevertheless, anticipated changes in the resolution of the low-angle detector banks will necessitate a calibration of detector segment positions. Hardware for measuring detector segment positions is in place and has been tested, and software has been developed to calculate detector segment boundaries using data collected with this hardware. A trial position calibration will be performed this Spring, and will hopefully be fully automated and routinely performed by the end of this year.

V. Backgrounds

Amorphous materials display an unfortunate tendency to be weak scatterers, compounded in some instances by a tendency to be very efficient absorbers of thermal neutrons as well. In these cases a successful analysis depends strongly on the reproducibility and magnitude of background scattering in the instrument. We have devoted considerable attention to sources of background in GLAD, and over the wavelength range for which GLAD is optimized its background is currently the lowest among diffractometers at IPNS. Moreover we know the sources of the background that remains, and are taking steps this Spring to greatly reduce the contributions of the most prominent of these. We demonstrate the current state of affairs in Fig. 3, a plot of normalized intensity vs. Q for $Li_2Si_2O_5$ glass for data collected at roughly 30°. Natural Li has a 70 barn absorption cross section (1.8 Å), a negative scattering

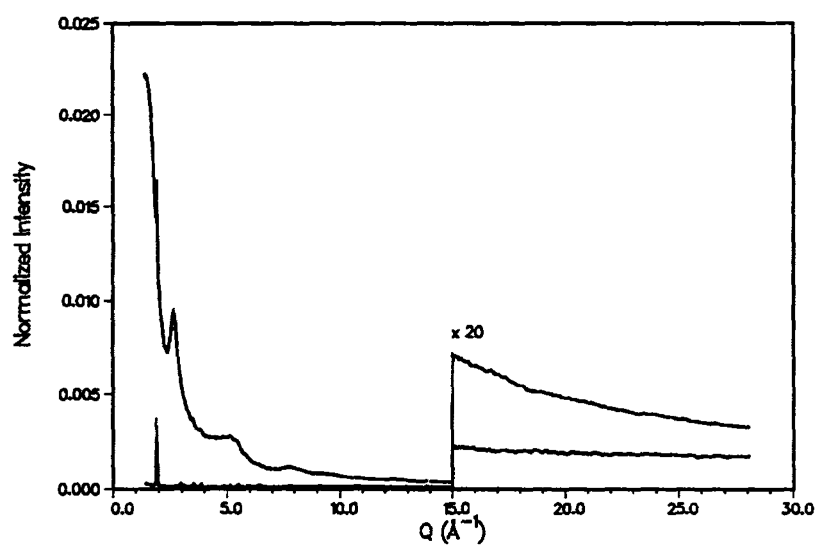

Figure 3. Normalized scattered intensity for $Li_2Si_2O_5$ glass. The lower curve is the measured background with the sample removed.

length, and a relatively large incoherent scattering cross-section, and its presence at the level of 22 atom% not only results in significantly attenuated scattering at long wavelengths but also greatly reduces the amplitude in the interference function. As is clear in the figure, even for this "difficult" sample the sample scattering is far greater than the background at all points.

VI. Data Normalization

Even with low backgrounds, analysis of scattering data in real-space is extremely sensitive to the normalization of the data. Unfortunately, there are no universal criteria for a determining whether or not one has obtained a "good" normalization, except perhaps the success or failure of a real-space representation of structure to replicate the coordination numbers of the various atoms in a known sample. We have adopted an alternative criterion: when a well-characterized sample of known geometry is placed in a beam of known dimensions and flux, and if absorption and multiple scattering corrections are relatively small, then the differential cross-section $d\sigma/d\Omega$ obtained for relatively high-energy neutrons (> 1 eV) should approach the calculated self-scattering (SLF) obtained using the methods described by Placek[9] - in other words, $(d\sigma/d\Omega)/(SLF) \rightarrow 1$ as $Q \rightarrow \infty$. When true, we have correctly accounted for the most important instrument-dependent factors contributing to raw scattering intensity. This criterion is indeed satisfied on GLAD. Figure 4 shows the ratio $(d\sigma/d\Omega)/(SLF)$ for a rod of v-SiO_2 collected at scattering angles of roughly 30, 45, 60, and 90°. Data from the various angles all fall on the same curve, and the ratios clearly converge to 1.0 at high Q.

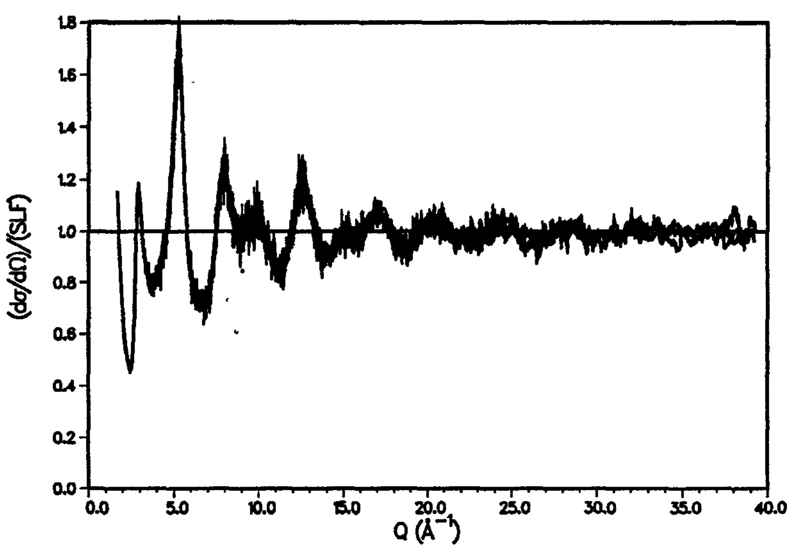

Figure 4. Ratio of the differential scattering cross-sections measured for v-SiO_2 at different scattering angles to the corresponding calculated self-scattering.

VII. Example

These results confirm that the data from GLAD meets or exceeds its design specifications, and thus "all" that remains is to develop optimized procedures for instrument operation while leaving enough flexibility to adapt to the needs of a particular experiment. We confine our demonstration of the capabilities of GLAD to a single example, the structure factor of D_2O. This is germane because the study of amorphous materials containing light atoms figured prominently in the design criteria for GLAD. Briefly stated, analysis of neutron scattering from materials containing light atoms is plagued by the importance of inelasticity corrections. While the Placek correction is adequate for relatively high energy neutrons scattering from atoms with "fixed" equilibrium positions, it is a comparatively poor approximation when the mass of the atoms in question approaches the mass of the neutron, particularly at high scattering angles. Therefore a premium is placed upon collecting data over as broad a Q-range as possible using the highest energy neutrons collected over the smallest range of scattering angles. Moreover, it must be possible to perform these experiments in the extremely limited amount of time typically allotted to experimenters!

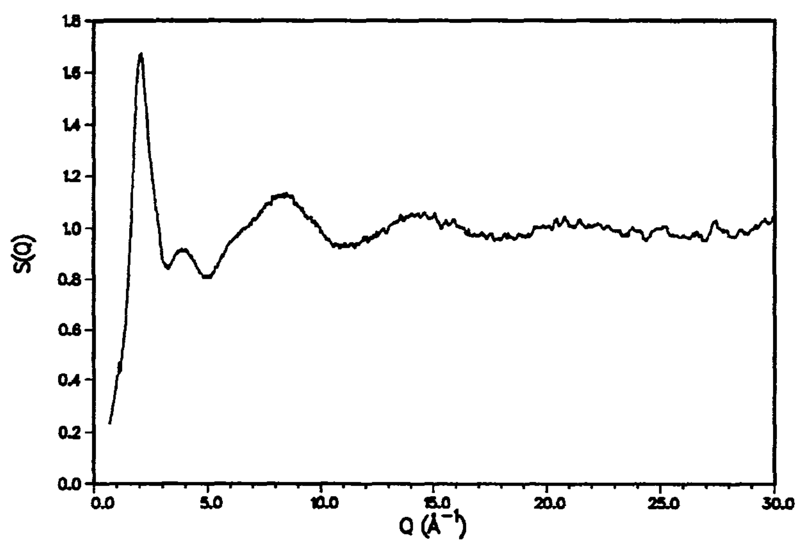

Figure 5. Structure factor for D_2O, based on a measurement range of 8-30° and 0.1-1.0 Å.

Figure 5 shows the structure factor of D_2O obtained using detector segments from 8-30° over a wavelength range of 0.1 - 1.0 Å. The sample was in a 4 mm x 8 cm vanadium container and was run at ambient temperature for 12 hours in the downstream sample position. Although the wavelength and angle range employed here is very restrictive, the Q range of the resulting structure factor encompasses all major diffraction features in D_2O, including the prominent low-Q pre-peak and its much weaker satellite at slightly higher Q, and extends to nearly 30 $Å^{-1}$. When a broader range of scattering angles and/or a larger wavelength range can be employed, the statistics at all points are improved significantly, and the structure factor data can be extended to far higher Q (analyses are routinely performed using data through Q = 40 $Å^{-1}$, and occasionally to Q = 50 $Å^{-1}$).

Acknowledgments

The success of GLAD is due in large part to the innovative mechanical design by Bob Kleb and the state-of-the-art data acquisition system designed and developed by Joe Haumann. The instrument was assembled and installed by the IPNS operations team headed by Don Bohringer and Dave Leach, and Merlyn Faber managed the fabrication and installation of the position encoding electronics. Sherman Susman has played a lead role in the development of the ancillary equipment for GLAD. Funding for the construction of GLAD was provided primarily by the Department of Energy through a proposal submitted by a Participating Research Team (PRT) headed by Simon Moss at the University of Houston.

References

1. R. K. Crawford, D. L. Price, J. R. Haumann, R. Kleb, D. G. Montague, J. M. Carpenter, S. Susman, and R. J. Dejus. *Advanced Neutron Sources 1988*, Proceedings of the 10th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS X), held at the Los Alamos National Laboratory, Oct. 3-7, 1988. Institute of Physics Conference Series Number 97, IOP Publishing Ltd, New York. pp 419-426 (1989).
2. R. K. Crawford, J. M. Carpenter, R. Dejus, J. R. Haumann, R. Kleb, D. G. Montague, D. L. Price, and S. Susman. Proceedings of the 11th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS XI), held at the National Laboratory for High Energy Physics, Tsukuba, Japan, October 22-26, 1990. KEK Report 90-25. pp. 820-829 (1991).
3. R. K. Crawford and J. R. Haumann. *IEEE Trans. Nucl. Sci.* **NS-37**, 72-81 (1990).
4. D. G. Montague, J. M. Carpenter, R. K. Crawford, R. Dejus, D. L. Price, and S. Susman. Proceedings of the 11th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS XI), held at the National Laboratory for High Energy Physics, Tsukuba, Japan, October 22-26, 1990. KEK Report 90-25. pp. 830-839 (1991).
5. R. K. Crawford, J. R. Haumann, and G. E. Ostrowski. Proceedings of the 11th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS XI), held at the National Laboratory for High Energy Physics, Tsukuba, Japan, October 22-26, 1990. KEK Report 90-25. pp. 943-956 (1991).
6. A. C. Nunes. *Nucl. Instrum. Methods* **119**, 291-293 (1974).
7. R. Berliner, D. F. R. Mildner, O. A. Pringle, and J. S. King. *Nucl. Instrum. Methods* **185**, 481-495 (1981).
8. Y. Ishikawa, M. Furusaka, N. Niimura, M. Arai, and K. Hasegawa. *J. Appl. Cryst.* **19**, 229-242 (1986).
9. G. Placek, The Scattering of Neutrons by Systems of Heavy Nuclei, *Phys. Rev.* **86** 377-388 (1952).