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12.1 HIPPO, The High-Pressure Preferred Orientation Diffractometer at LANSCE for Characterization of Bulk Materials

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Abstract

United States National Laboratory researchers and University of California faculty, representing a broad range of scientific disciplines, is building a novel time-of-flight (TOF) neutron diffractometer and associated *in situ* equipment at the Manuel Lujan Jr. Neutron Scattering Center (Lujan Center), under the auspices of the United States Department of Energy. The goal with the High-Pressure Preferred Orientation Instrument (HIPPO) is to investigate dynamic processes in heterogeneous bulk materials in a variety of environments. The instrument, which will become available in summer 2001, has the extremely high count-rates necessary to study time-dependent processes in small (1-mm diameter) and large (2-cm diameter) samples, and in a large variety of environmental conditions (10-2000 K cryostats and furnaces, 0-20 GPa pressure vessels, straining cells, goniometers, magnets, etc.). The 3-D arrangement of detectors allows direct measurements of crystal orientation distributions in polycrystalline materials. The analysis of TOF diffraction patterns with versatile Rietveld codes provides simultaneous information on crystal structure, texture, microstructure and phase proportions. While this instrument has many applications in materials science, it is also of great interest for geology and geophysics. Some applications include: kinetics of reactions, structure of silicate glasses and melts, high-pressure investigations of complex systems, evolution of texture and anisotropy during deformation and recrystallization. The Lujan Center aims at creating an instrument with high data through-put and easy access to researchers and students. While the HIPPO instrument will be part of the national user facility operated by the Lujan Center, the scientific program will be guided by the University of California consortium with the goal of satisfying national priorities and establishing an environment of scientific excellence. [For more information search for "UCMRD" or http://lansce.lanl.gov/research/index_res_99.html on the web].

1. Introduction

Advanced neutron source technology and future generation spallation facilities will enable major advances in neutron instrumentation and research capabilities for condensed matter research. Important new applications of neutron diffraction have emerged already in both applied and fundamental research on condensed matter. The new High Pressure Preferred Orientation (HIPPO) diffractometer is designed to exploit the power of time-of flight (TOF) neutron diffraction for materials science research by innovative engineering design, flexible sample environments, and fast data acquisition routines. A salient characteristic of neutrons *vis-à-vis* X-rays for scattering research is the low absorption by most elements. This makes it possible to study bulk properties, to utilize sophisticated sample environments for studies at high or low temperatures and pressures, and to investigate orientation distributions and internal stresses in polycrystals. Due to the high spectral resolution that is available, refinements of crystal structures, phase proportions, textures and strains can all be performed simultaneously, e.g. with the Rietveld method [1,2]. The objective of HIPPO is to be a high count rate diffractometer with moderate resolution and flexible sample environments.

The HIPPO diffractometer is part of a synergistic effort by both the University of California campuses and US National laboratories to (1) attain scientific excellence (2) advance our present knowledge of condensed matter and (3) make neutron diffraction an available tool for younger generation scientists. Costing less than $4M and chosen by the US Department of Energy (DOE) for accelerated construction, the HIPPO diffractometer is presently under construction. It is anticipated to become available for testing in fall 2000 with a user program initiated in summer 2001. The instrument is designed to study properties of polycrystalline materials and liquids. Of particular interest is the investigation of small ($1mm^3$) and large ($2cm^3$) sample volumes at high (<2000K) and low (>10K) temperatures, at high pressure (<20GPa), and in different atmospheres. The diffractometer which we are building will be highly visible in the fields of phase transformations, high pressure research, polycrystal anisotropy (texture-strain-stress) and complex materials crystallography. It will be possible to study the dynamics of reactions, recrystallization and deformation of bulk anisotropic samples at a wide range of temperature and pressure conditions. No existing instrument, world-wide, has this range of environmental capabilities.

A major limitation of neutrons has been the weak intensity. With the new diffractometer we can overcome this problem, taking advantage of the improved neutron source at LANSCE, a short flight path (flux at the sample: 10^8 n $cm^{-2}s^{-1}$) and a novel three-dimensional arrangement of detector banks with 1400 ^{3}He-tubes, on conical rings. The usual data collection time will be reduced from hours to minutes, enabling us to study time-dependent processes in bulk samples with anisotropic properties. A flexible sample environment chamber (large 75-cm diameter sample well) accommodates ancillary equipment such as goniometers, furnaces, cryostats, straining stages, high-pressure cells, magnets etc. A new state of the art DAQ system enables remote and fast data acquisition and experiment monitoring via the world wide web from a user's laboratory.

The HIPPO Diffractometer will be housed at the Manuel Lujan Jr. Neutron Scattering Center as part of an over $42 M, facility-wide upgrade project for the Los Alamos Neutron Science Center (LANSCE) sponsored by the US DOE. LANSCE is a unique spallation (100 μA proton beam at 20 Hz with upgrade to 200μA at 30 Hz) source that produces intense bursts of polychromatic neutrons that can be captured in time-of flight (TOF) for fundamental condensed matter research. To learn more about LANSCE or the Short Pulse Spallation Upgrade project at LANL please see the web site http://lansce.lanl.gov/overview/index_over.html.

2. HIPPO Technical Description

For a more detailed description of the project, technical design and installation progress visit the HIPPO website http://www.seismo.berkeley.edu/~wenk/hippo.html or search for UCMRD.

Overview

The goal for this instrument as stated above is to have the highest intensity available with moderate resolution. This is to be achieved by an instrument design consisting of detectors positioned over as much of the azimuthal angle about the incident beam as possible with the instrument located on a quite short flight path. To keep the resolution of the instrument independent of the detector azimuthal angle, we use a neutron flight path at the Lujan Center that is normal to a moderator surface. A Monte Carlo simulation routine (program NISP, Neutron Instrumentation Simulation Package) [3] was prepared to simulate a conventional time-of-flight powder diffractometer and it has been used to verify the choice of design features of HIPPO.

The HIPPO diffractometer features a short initial flight path of 9m at Flight Path 4 of the Lujan Center and an array of more than 1400 10-atm ^{3}He detector tubes covering nearly 4.6m^2 with five detector banks at scattering angles ranging from backscattering (nominally 150°) to low forward scattering (nominally 10°). The characteristics of each detector array are listed in Table 1 and their placement is shown in Figure 1. Figure 1 shows an exploded view of the diffractometer along the incident beam line. The detector panels are tilted relative to the scattered neutron paths to give a more constant resolution across their surfaces. The tilt compensates the change in resolution due to the angular range for each panel with a corresponding change in the sample-to-detector flight path (L, Table 1). The collimation views a 12-cm diameter round portion of a high intensity ambient water moderator, and converges to a maximum round beam size of 2-cm diameter at the sample position. A T_0 chopper removes the fast neutron prompt pulse. For most applications samples ought to be fully immersed in the neutron beam and should not exceed this size. However, smaller beam sizes at the sample position can be produced with adjustable collimation. Motorized neutron collimating elements (see Figure 1) are positioned along the beam path to provide addition tailoring of the beam profile at the sample (down to 5mm diam.). The cost of such an adjustment is, of course, a loss of intensity. In addition to reduce the effects of air scattering in the secondary flight paths between the sample chamber and forward-scattering detectors, we use 1.0 mil aluminized Mylar balloon filled with Helium gas that reduce the aborption from air by nominally 7%.

It is anticipated that the count rate for some experiments on HIPPO will be approximately 20-60 times what is currently obtained on the present High Intensity Powder Diffractometer (HIPD) on Flight Path 3 at the Lujan Center, and will enable measurements in as little as 5-10s. The time-average flux on a sample positioned at 9m, as measured on FP-3 (HIPD) with the neutron source operating at 70μA, is ~$9x10^7 ns^{-1}cm^{-2}$ for all neutron energies of which~$1x10^7$ $ns^{-1}cm^{-2}$ is in the "thermal" (<1eV) range suitable for diffraction work. It is expected that the flux on FP-4 will thus be very similar to that observed for HIPD; scaled to 200μA beam current, we expect to have a time average flux of ~$2x10^8$ $ns^{-1}cm^{-2}$ for all neutron energies and ~$3x10^7$ $ns^{-1}cm^{-2}$ for thermal neutrons. The data acquisition will be based on current VME technology and make use of web-based visualization and control software. Experiments can be controlled remotely, e.g. from the laboratory of the user.

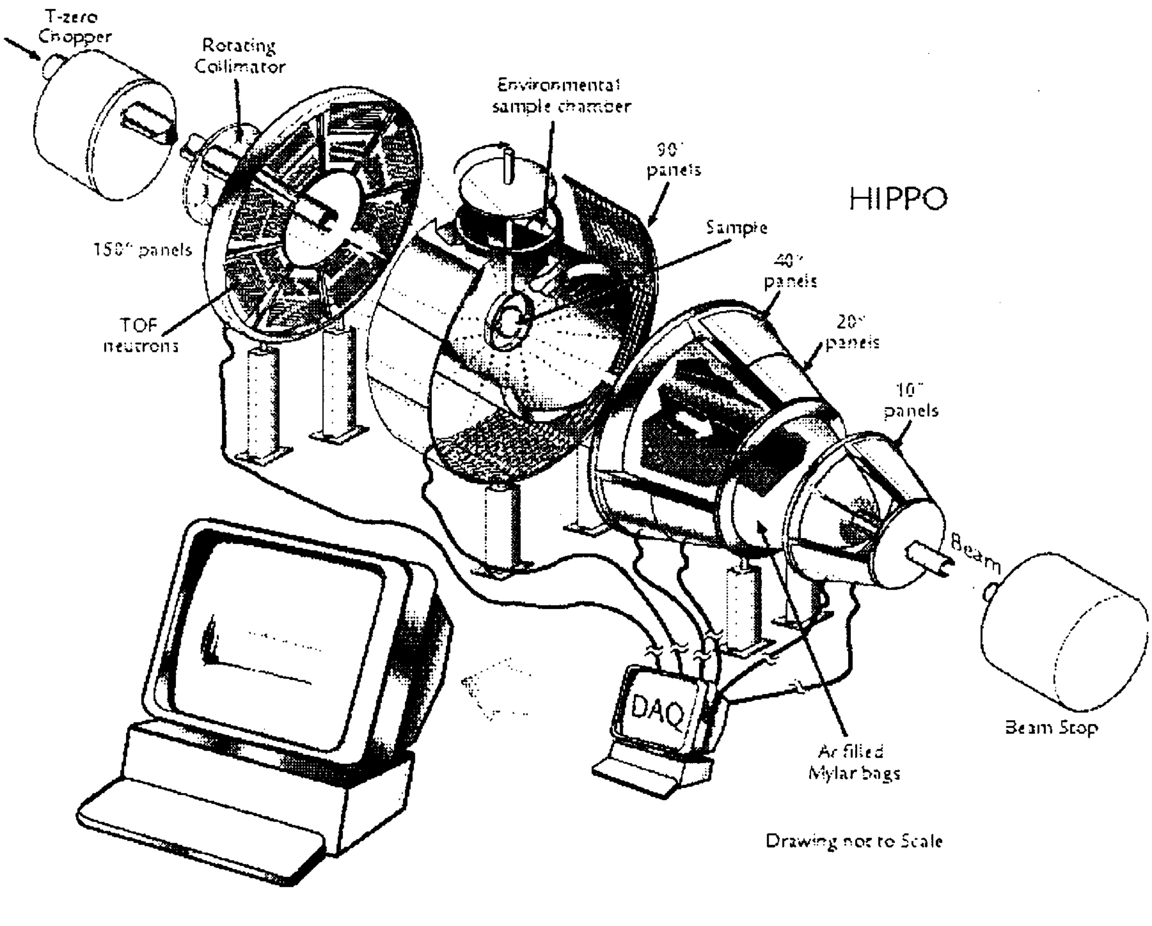

Figure 1. Exploded view of HIPPO diffractometer showing sample chamber surrounded by 5 conical rings of over 1400 ^{3}He detector tubes. The TOF incident beam travels from left to right of figure, beginning at the 150 ° panels. A 60 Hz T_0 chopper removes the fast epithermal neutrons. A rotating collimator allows adjustable circular collimation from (2-cm diam. to 5-mm diam. spot size). The large sample chamber accomodates special environment equipment such as displexes, goniometers, furnaces and high-pressure cells.

An Helium-filled Mylar bag is used to reduce incoherent scattering in the forward scattering secondary flight paths.

Table 1. Detector layout for HIPPO. The d and Q ranges are for 0.5-9 Å wavelength range. The resolution is full width at half maximum over position (100% ΔT/T). The resolution was determined by comparison with the corresponding detectors currently in use on HIPD and Monte Carlo simulations with the program MONTE

Nominal angle	angular range	L_2,m	d-range, Å	Q range, Å^{-1}	ΔT/T
150	143°54'-155°48'	1.25	0.12-4.80	1.31-52.4	0.37%
90	76°46'-103°15'	0.75	0.17-6.90	0.91-37.0	0.75%
40	33°29'-47°35'	1.00	0.35-13.9	0.45-18.0	1.5%
20	18°17'-25°47'	1.50	0.65-26.1	0.24-9.67	2.6%
10	7°17'-12°45'	2.00	1.19-47.5	0.13-5.28	5.0%

Sample Chamber and Ancillary Equipment

The HIPPO diffractometer uses a large sample with a 75cm (29.52") opening at 81.28cm (32") from the neutron beam centerline. It is designed for easy interchangeability of sample environment equipment (Figure 2.). The aluminum chamber is fabricated as a vacuum vessel and can be evacuated to approximately 10^{-6} torr so to provide adequate insulation for low temperature equipment (Displexes or cryostats) and adequate protection for the high temperature furnace. However at present most ancillary stages for HIPPO are closed-looped, self-sufficient systems that do not require the chamber vacuum. The window areas of the chamber are thinned (0.20") aluminum to minimize attenuation of secondary beams to the detector panels.

Because of the low absorption of neutrons, we can use many environmental stages that consist of materials mostly transparent to neutrons. Environmental stages (i.e. high temperature, low temperature, and stress/strain) can be used on HIPPO for *in situ* observation of texture changes [6]. To accommodate the breadth of research applications for HIPPO, we have purchased or fabricated several different pieces of ancillary in a large variety of environmental conditions (10-2000 K cryostats and furnaces, 0-20 GPa pressure vessels, straining cells, goniometers) to insert into the sample chamber. Some stages are standard. Some environments are specially designed.

An APD Displex system is standard equipment. It is capable of ca. 10K to 300K operation with 0.1K precision. Lower temperature capability (1-2K) will be achieved by the use of a liquid He cryostat (ILL "Orange" cryostat or equivalent) in house at the Lujan Center. We also have a two-stage displex for reaching temperatures as low as 4 K.

In addition we have a conventional 3-axis goniometer using Kappa geometry for standard texture measurements. The design of this goniometer is based on one by A. Schultz (IPNS). Its operation will be optimized to produce complete coverage of the orientation space taking into account the fact that the goniometer would inevitably obscure some detector panels. Because modern data analysis techniques utilizing Rietveld techniques

[4,5] can accommodate any set of sample orientation angles, there is no requirement that the pole figures be sampled on a uniform grid as used in classical pole figure measurements. The azimuthal array of detector panels on HIPPO allows texture measurements to be done with very few sample orientations. In Figure 3 we show how a single sample setting gives considerable coverage of the orientation space; resulting in 20-32 points on a pole figure simultaneously from the 150, 90 and 40 degree detector banks.

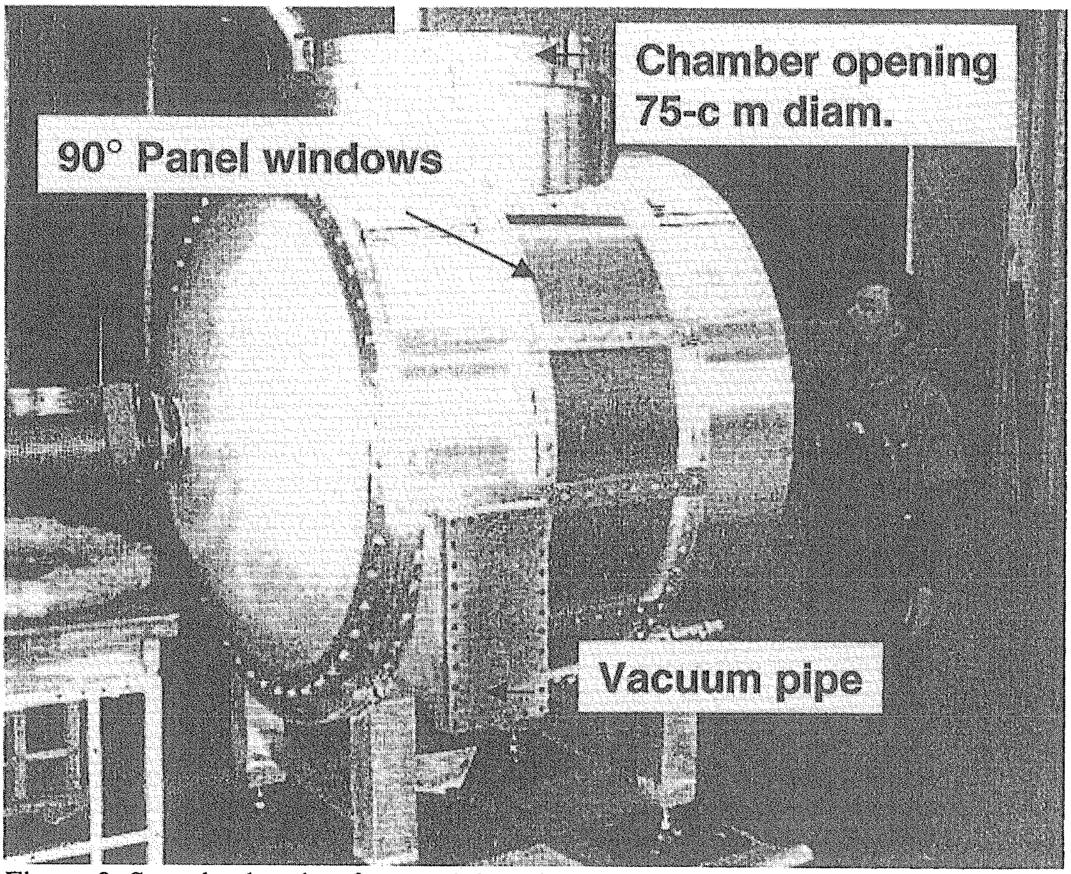

Figure 2. Sample chamber for special environment equipment after its fabrication. The chamber is made out of 6061 aluminum and has been vacuum tested to nominally 10^{-6} torr. A number of sample environments (cryostat, furnace, texture goniometer, high-pressure cells, sample changer) are accommodated within the sample chamber for various research applications.

Other specially designed equipment include an innovative "sample changer" fabricated by Zygo Engineering, Inc. that holds 100 samples at a time for rapid powder-diffraction measurements and 32 samples at a time for rapid texture measurements. It will facilitate composition driven materials studies as well as enable handling a suite of samples from several experimenters. Because of the high throughput of HIPPO, this sample changer is essential for efficient utilization of the neutron diffractometer. In addition we purchased a specialized combined furnace and gas flow apparatus for *in situ* and kinetics experiments constructed by AS Scientific Products, Ltd and provided by Sandia National Laboratory. This furnace is based on a standard ILL 1800°C furnace and is equipped with a single rotation axis to permit examination of texture evolution under high temperature.

Two existing 240Ton Paris-Edinburgh high pressure cells, which are capable of 10GPa for a 100mm^3 sample and 7.5GPa for a 350mm^3 sample, will be used on HIPPO. They also are capable of simultaneous high temperature (1600K) and high pressure (7GPa) with an internally heated furnace assembly. In addition we fabricated a Toroidal Anvil Press (TAP-98), with a 500-Ton capacity with target simultaneous P-T conditions of 20 GPa and 2000 K. TAP-98 is a compact format with multiple neutron access ports, large tooling space and a self-alignment stage.

A future development for texture studies on HIPPO will include a texture goniometer equipped with a computer controlled X,Y,Z stage and 90° radial collimators; this will permit the determination of position dependent texture within the dimensions of an extended sample. The collimation should be sufficient to determine a 2x2x2-mm^3 gauge volume with a sample positioning precision of 0.1mm or less. Each goniometer requires only modest angular precision for the setting angles and 0.1deg is more than adequate. In addition, design and fabrication of a rotatable stress rig are in process at the Technical University of Hamburg. It will be used for the simultaneous determination of texture and strain evolution with applied unidirectional stress.

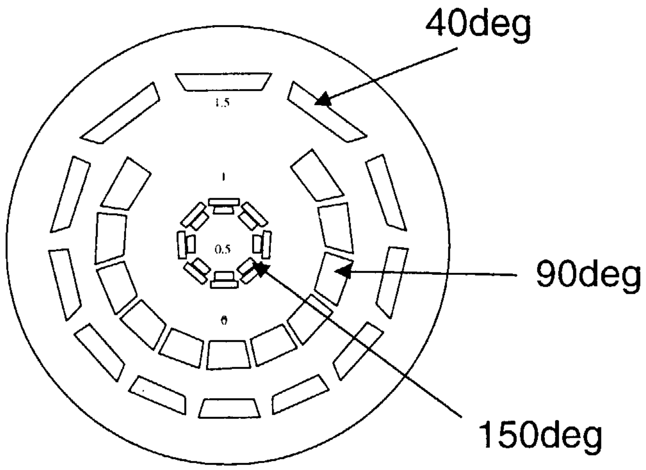

Figure 3. A stereographic projection of the 150, 90 and 40 degree sets of detector panels on a pole figure for one sample orientation. Incident beam direction is along the projection axis. The gap in one set of detector panel images arises from the gap in the 90 degree bank where the sample chamber access port is located. The detector panel images for the 150 and 40 degree banks have no gaps.

3. Data Acquisition System

The HIPPO (DAQ) system conforms to the new standards being developed at the Lujan Center. There are three primary components of the DAQ: the Graphical User Interface (GUI), the real time system and the server. The user is connected to the GUI which provides access to the system to allow initiation of data collection, display of spectra, control of special environment equipment, etc. The detector systems are attached to the real time portion that captures the data in digital form and generates the TOF histograms.

with phase transitions in the deep mantle can also be studied by high P-T neutrondiffraction experiments.

Established research focus groups for HIPPO include (1) Liquids and amorphous materials, contact Brian Annis, ORNL, annisbk@ornl.gov (2) High pressure, contact Raymond Jeanloz, UCB, jeanloz@uclink.berkeley.edu (3) High temperature contact Mark Rodriguez, Sandia, marodri@sandia.gov (4) Texture and strain, contact Rudy Wenk, UCB, wenk@seismo.berkeley.edu.

5. User Program and Conclusion

The HIPPO diffractometer will be used for investigations of scientific relevance, and be accessible for a wide, multidisciplinary range of experiments in physics, chemistry, materials science, engineering and earth sciences. It is planned to perform 150-250 experiments each year during an 8-month run cycle, making neutron diffraction not only a method for a few dedicated neutron specialists, but also a viable resource for the 'mainstream' materials and earth science communities.

A formal user program with Los Alamos University of California Directors Research and Development (UCRDR) support is now in place to assist University of California and New Mexico University users. It includes the following provisions: (1) 20 travel subsidies (from $500 to $1000) per experiment) per year for students and faculty from Campuses and New Mexico Universities to visit Los Alamos for experiments (2) one graduate or undergraduate student stipend to spend extended time (e.g. 3 months in summer) at LANSCE to conduct research in neutron diffraction (3) a one-year fellowship for a graduate student to engage in a Ph.D. thesis project in materials or earth sciences that emphasizes the HIPPO diffractometer and (4) Postdoctoral researcher to help users with data processing and data interpretation. Proposals for HIPPO will be accepted in Spring 2001 and may be submitted as (1) *standard*, a single defined experiment (2) *extended*, a project over 1 or 2 years or (3) *exploratory*, short term for sample identification. For more information on the program contact wenk@seismo.berkeley.edu.

References

[1] R.B. Von Dreele, Quantitative texture analysis by Rietveld refinement, *J. Appl. Cryst.*, (1997) 577.

[2] L. Lutterotti L., S. Matthies, H.-R. Wenk, A.J. Schultz and J.W. Richardson, Combined texture and structure analysis of deformed limestone from time-of-flight neutron diffraction spectra, *J. Appl. Phys.* (1997) 594.

[3] P. A. Seeger, L. L. Daemen, T. G. Thelliez, and R. P. Hjelm, Jr., Neutron instrumentation simulations in the next millennium, Physica B, (2000) 433.

[4] H.-R. Wenk, Texture analysis with TOF neutrons, *Trans. Am. Cryst. Ass.*, (1993) 95.

The data acquisition server section then provides the link between the other two portions as well as routing instrument control information (temperature settings, goniometer angles, etc.) to the diffractometer and creating the Hierarchical Data File (HDF) formatted data files for archival at the end of each run.

Our real time system uses a commercial computer interface from VME Microsystems, Inc. based on time-slicing direct memory access processors with network capability. The HIPPO system consists of thirteen VME crates that perform time-stamping, buffering, vetoing and readout of the detector banks. Each VME crate consists of a VME processor module that provides histogram building and data readout and handles 100K-1M TOF channels from these detector banks. A 128Mb-memory module in each crate is used for program storage and provides sufficient storage for multiple histograms generated, for example, in a time series kinetics experiment. The detector interface is equipped with double buffers so that one buffer can be processed while the other is being filled. The buffers collect data in alternate neutron pulses; this scheme is needed to handle the high event rate expected from this instrument. The 100Mbit/s dedicated network will ensure that the data in each VME module (3.2-16Mbits each) can be read in less than 1s. We are using VxWorks (WindRiver) to control the real time system on a 600 MHz commercial PC running MicroSoft Windows NT. And the GUI interface will be implemented to operate within a browser (e.g. Netscape) so that the data display facilities can be accessed remotely.

4. Research Applications

Though we cannot mention all the various research possibilities on HIPPO, we list a few applications here. We designed HIPPO to accommodate a myriad of research efforts. HIPPO can be applied to research in kinetics of reactions; high-pressure investigations of complex systems with large sample volumes; the evolution of texture in polycrystals during deformation processes; and recrystallization and phase transformation studies. Areas of research might include texture and anisotropy studies of rocks, for example, granite-mylonite and mantle peridotites; crystal-structure studies of zeolites; and structure studies of liquids and melts, including aluminum-silicon melts and glasses. It will be possible to perform real-time experiments on bulk anisotropic samples at a wide range of temperature and pressure conditions. In addition texture information that can only be obtained with this instrument is a prerequisite to interpreting, for example, high-resolution residual stress data.

Using HIPPO we may perform in-situ high pressure and temperature (P-T) neutron-diffraction experiments, providing unique capabilities to study texture, hydrogen bonding, magnetic moments, and structural and thermal parameters of light elements (for example, hydrogen, lithium, and carbon) and heavy elements (for example, tantalum, uranium, and plutonium), which are virtually impossible to determine by x-ray diffraction techniques. For example, we can derive thermoelasticities and Debye-Waller factors as functions of pressure and temperature using in-situ high P-T neutron diffraction techniques [7]. These applications can also be extended to a much broader spectrum of scientific problems. For instance, puzzles in Earth science, such as the carbon cycle and the role of hydrous minerals for water exchange between lithosphere and biosphere, can be directly addressed. Moreover, by introducing in-situ shear strain, texture accompanied

[5] H.-R. Wenk, S. Matthies and L. Lutterotti, Texture analysis from diffraction spectra, *Proc. ICOTOM 10, Mater. Sci. Forum*, (1994) 473.

[6] K. Bennett, H.-R. Wenk, W.B. Durham, L.A. Stern and S.H. Kirby, Preferred crystallographic orientation in the ice I-II transformation and the flow of ice II, *Phil. Mag. A.*, (1997) 413.

[7] Y. Zhao, R. B. Von Dreele, J. Zhang, and D. J. Weidner, Thermal Equation of State of Monoclinic Pyroxene: $CaMgSi_2O_6$ Diopside, *Review of High Pressure Science & Technology*, (1998) 25.

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