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J. Faber, Jr.

Materials Science & Technology Division  
Argonne National Laboratory  
Argonne, IL 60439

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# SAMPLE ENVIRONMENTS AT IPNS: PRESENT AND FUTURE CAPABILITIES

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Materials Science & Technology Division  
Argonne National Laboratory  
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## ABSTRACT

Argonne's Intense Pulsed Neutron Source, IPNS, was dedicated as a major user-oriented neutron scattering facility two years ago. Most instruments are now equipped to provide for sample environments in the temperature range  $1.5 \leq T \leq 1300\text{K}$ . A special facility provides  $T \leq 1\text{mK}$ , and another provides pressures to 30kbar. Several environmental equipment designs are described that emphasize time-of-flight technique. Methods for achieving time-resolved experiments which take advantage of the IPNS pulsed source characteristics are discussed.

## Resumé:

La Source Intense de Neutrons Pulsés (IPNS) d'Argonne a été inaugurée, il y a deux ans, comme un équipement majeur de diffusion neutronique ouvert à un grand nombre d'utilisateurs. La plupart des instruments sont maintenant équipés de cellules d'environnement permettant de travailler dans l'intervalle de température  $1.5 \leq T \leq 1300\text{K}$ . Un dispositif spécial permet de travailler à  $T \leq 1\text{mK}$ , et un autre fonctionne à des pressions pouvant atteindre 30 Kbar. Plusieurs plans de cellules d'environnement qui utilisent des techniques du temps de vol sont présentés. Les méthodes pour effectuer des expériences de temps résolu qui tirent avantage des caractéristiques particulières de la source pulsée de l'IPNS sont discutées.

## INTRODUCTION

The Intense Pulsed Neutron Source (IPNS) is a major dedicated user-oriented facility for the study of condensed matter physics with thermal neutrons. A linac accelerates  $H^-$  ions to 50 MeV. These are stripped to protons and accelerated to 450-500 MeV in the rapid cycling synchrotron (RCS). Protons extracted from the RCS are transported to a uranium target located inside a shielding monolith in the experimental hall; neutrons are produced by spallation. Moderator configurations in the target assembly act as effective sources for the respective beam lines surrounding the monolith as shown in Figure 1. The source moderators produce sharp time bursts of polychromatic beams with a repetition rate of 30 Hz. The peak flux is  $4 \times 10^{14} \text{ ncm}^{-2} \text{ s}^{-1}$ . As illustrated in Figure 1, ten instruments are positioned around the monolith: six diffractometers and four spectrometers. A comprehensive report of the facilities is available(1). Most instruments have dedicated closed-cycle refrigerators for cryogenic sample environments. Exchange gas cryostats are operational that supplement the limited low temperature capability of the closed-cycle refrigerator systems, and provide  $T \sim 1.5\text{K}$ . In collaboration with University of Illinois, we have capability down to 50 mK with a dilution refrigerator, successfully operated for example on beam line F5. A special ultra low temperature refrigerator provides  $T < 1\text{mK}$ . Several vacuum furnaces are operational and available for either diffractometer or spectrometer sample environment needs and provide  $T < 1300\text{K}$ . A high pressure cell provides hydrostatic pressure to 30 Kbar at room temperature. In the following section, several examples of ancillary equipment design are illustrated.

SAMPLE ENVIRONMENT DESIGN EXAMPLES

The basic design elements for low temperature environments are to allow when possible an unimpeded path for the incident and down-stream beams through the closed-cycle refrigerator or exchange gas cryostat. If positioned well outside the diffraction geometry, thin wall aluminum heat shields are chosen. Nearer to the sample position, thin-wall vanadium cans are used to help insure a homogeneous temperature profile. Provision for loading specimens in vanadium cans containing helium exchange gas provide the method for minimizing  $\Delta T$  between thermometry and sample. The typical sample dimensions are 1.1 cm dia x 5 cm high on the two powder diffractometers located at F5 and F2 illustrated in Figure 1. The motivation for these designs lies in the problems attendant with undermoderated fluxes (epithermal and high energy neutrons) that emerge from the source. The high energy neutrons in the incident beam can "moderate" or multiple scatter from the environmental equipment thus producing sample and environmental equipment dependent apparent backgrounds as much as 50% higher than anticipated on the basis of thermal neutron cross section estimates alone. Boron carbide and boron nitride are typically used in the scattered flight path to provide detector bank or area-detector collimation. Cadmium is ineffective in the epithermal ( $1 < E < 10$  ev) range. Where possible, evacuated sample chambers (that house the environmental equipment) are kept under vacuum to reduce air scattering background.

In the high temperature regime, the powder diffractometers employ a Ta ribbon wound vacuum furnace for  $T < 1300K$ . Figure 2 is a photograph of the high temperature stage. Note the open path for the incident beam. Vanadium foil heat shields (not shown) which surround this assembly, are used to provide temperature homogenization. The gradient in temperature  $\Delta T$  over the sample

area is typically  $\leq \pm 5K$ . Figure 3 shows the use of this furnace to study the monoclinic to tetragonal phase transition in  $\text{LaNbO}_4$  ( $T_c \sim 778K$ ). These are data collected on the high resolution ( $\Delta d/d$  fwhm = 0.25% at  $2\theta = 150^\circ$ ) General Purpose Powder Diffractometer (GPPD) on port F2 of Figure 1. Each diffraction pattern, (a), (b), or (c) in Figure 3 required 3 hours of data collect time. A high temperature furnace is under construction to provide artificial gaseous environments and  $T < 1500K$  for in-situ experiments. For the Laue Single Crystal Diffractometer (SCD) on port H1 in Figure 1 a high temperature single crystal furnace(2) is illustrated in Figure 4. This design clearly shows that large volumes of reciprocal space can be measured. A wide wavelength range,  $\phi$  and  $\chi$  angle adjustments, and a large 2-dimensional area detector provide access to these large volumes. This furnace operates in air, with  $T < 1300K$ . Again, an unimpeded path for the incident beam is provided.

The design of high pressure environmental cells for room temperature studies(3) is illustrated in Figure 5. This design uses massive support and provides uniform compressive stresses on the  $\text{Al}_2\text{O}_3$  sample cell which is then loaded by hydraulic rams to provide pressures up to 30 kbar. The samples are typically loaded in teflon tubes and may contain pressure transmitting fluids and pressure calibrants. The pressure calibrant most often used is CsCl. Plans are in progress to provide pressure capabilities up to 20 kbar at helium temperature.

#### TIME-RESOLVED EXPERIMENTS

The time structure of the neutron bursts generated at IPNS and the use of white beam techniques that utilize time-of-flight methods lead us to consider the potential for time-resolved neutron scattering studies. The prospects for time-resolved experiments were pointed out recently by Windsor.(4) In fact,

pioneering experiments carried out by Niimura and Muto(5) showed that such experiments are feasible, even at much lower neutron fluxes. One particular unique advantage of the IPNS data acquisition system(6) is the use of flexible microprocessor based data histogramming. This is illustrated in Figure 6, where the Bragg peaks (a) and (c) correspond to single detector response at the extremes of a 20 detector bank centered at  $2\theta_0=152^\circ$  ( $\Delta 2\theta/\text{detector} = 0.5^\circ$ ). The Bragg peak in each case is Si(220), with  $a_0=5.4309\text{\AA}$ . The middle peak (b) in the figure shows the results of electronic time-focussing(6). Note however that the time range spanned by these detectors is  $\sim 450 \mu\text{sec}$ . A significant time range of observation follows from the relaxation of time-focussing constraints and we can amplify this idea in the following discussion. The neutron wavelength,  $\lambda$  in angstrom units, is related to tof and path length by

$$\lambda = \frac{h}{mv} = \frac{h}{m} \frac{t}{L} = 3956 \frac{t(\text{sec})}{L(\text{m})} . \quad (1)$$

Time is measured in seconds and path lengths in meters. Suppose that we choose the GPPD at IPNS, with a continuous bank of detectors spanning  $60 < 2\theta < 150^\circ$  as the instrumental resolution function,  $R < 0.7\%$ . The source to sample distance  $L_1=20\text{m}$ , the sample to detector distance  $L_2=1.5\text{m}$ , each detector has  $\delta 2\theta_1 = 0.5^\circ$ , and the total flight path  $L_T=L_1+L_2 = 21.5\text{m}$ . The quantity of interest is the time span for constant d-spacing at the sample position, so with  $\lambda=2d\sin\theta$  and the parameters given above:

$$\Delta t_s = \frac{\Delta \lambda L_1}{3956} = \frac{2L_1 d}{3956} (\sin\theta_{\text{max}} - \sin\theta_{\text{min}}) = 4.7 \times 10^{-3} d . \quad (2)$$

This result is illustrated in Figure 7, where  $\tau$  is the pulsed source period. To estimate the resolution within  $\Delta t_s$ , assume that the instrumental resolution  $\Delta t/t|_{\text{fwhm}} = 4 \times 10^{-3}$  (the case for  $2\theta = 90^\circ$ ), then

$$\Delta t|t_s = 4 \times 10^{-3} \frac{L_1}{L_T} = 3.7 \times 10^{-3}, \quad (3)$$

and since

$$t_s = \frac{2dL_1 \sin\theta}{3956} = 7.15 \times 10^{-3} d, \quad (4)$$

the resolution element at the sample,  $\delta t_s$  is

$$\delta t_s = (\Delta t/t_s) = (26.4 \text{ } \mu\text{sec})d. \quad (5)$$

The number of resolution elements,  $N$ , contained in  $\Delta t_s$  (Figure 7) is approximately equal to the number of detectors spanning the  $2\theta$  range, i.e.,  $N=180$ . If  $d=1\text{\AA}$ , this gives 30  $\mu\text{sec}$  resolution over  $\Delta t_s = 4.7 \text{ msec}$ . As pointed out by Windsor(5), a phased excitation relative to neutrons produced at the source allows us to examine different time ranges in the sample response function. The data acquisition system at IPNS (memory capacity to 16 Mbytes) can easily store the required separate histograms. The method described above is a constant  $d$ -space technique. While we have pointed to one particular  $d$ -spacing, extension of these ideas to many Bragg peaks is straight-forward. Suppose we collect the data from all detectors given in the example above for  $1 < \lambda < 5\text{\AA}$ , then  $5.06 < t_s < 25.3 \text{ msec}$  and  $\Delta t_s = 20.2 \text{ msec}$ . The data reduction scheme is to choose  $\lambda$  and a narrow  $\Delta\lambda$  to sort through all detector histograms, thus producing  $I(2\theta)$  for constant  $\lambda$ . This is the tof

analogue of constant  $\lambda$  experiments, except in our polychromatic tof case we can choose many different  $\lambda$  and  $\Delta\lambda/\lambda$  by which to construct our data representation. The choice of  $\lambda$  correspond to examining the sample function at a particular time. One disadvantage of the scheme outlined here is that counting times are much longer since time-focussing advantages have been relaxed. The main advantage is that sample response times can be easily sampled with 10's of microseconds resolution. Sufficient flexibility exists to couple a desired sample response time program through CAMAC interfaces to ancillary equipment control modules.

#### SUMMARY

After only two years of operating time as a major user-oriented facility, the IPNS sample environment program has produced a number of successful environmental cells for use with time-of-flight techniques. The program will continue development beyond the operational devices discussed here (1). A unique opportunity exists for pulsed spallation neutron source time resolved studies. The advent of higher source fluxes should allow us to more readily realize these possibilities.

#### ACKNOWLEDGEMENT

It is a pleasure to thank G. H. Lander and R. K. Crawford for useful discussion on elements of this paper. A special thanks to A. J. Schultz who provided unpublished details on the single crystal high temperature equipment design, and M. H. Mueller and the IPNS technical staff who helped to make the high temperature powder diffractometer furnace designs a reality. This work is supported under the U.S. Department of Energy.

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FIGURE CAPTIONS

Figure 1. Layout of the experimental hall at Argonne's Intense Pulsed Neutron Source (IPNS). The central structure houses the neutron scattering target and moderator assemblies. The beam lines connect to the following instruments: H3, High Resolution Medium Energy Chopper Spectrometer; H1, Single Crystal Diffractometer; F5, Special Environments Powder Diffractometer; F4, Low Resolution Medium Energy Chopper Spectrometer; F1, Crystal Analyzer Spectrometer; F2, General Purpose Powder Diffractometer; C1, Small Angle Diffractometer.

Figure 2. High temperature heater stage that produces  $T < 1300\text{K}$ . The heater element is tantalum ribbon, 0.05 mm thick. Alumina support posts and boron nitride disks form the support structure for the furnace element. Sample diameters as large as 1.1 cm are located in the center of the furnace.

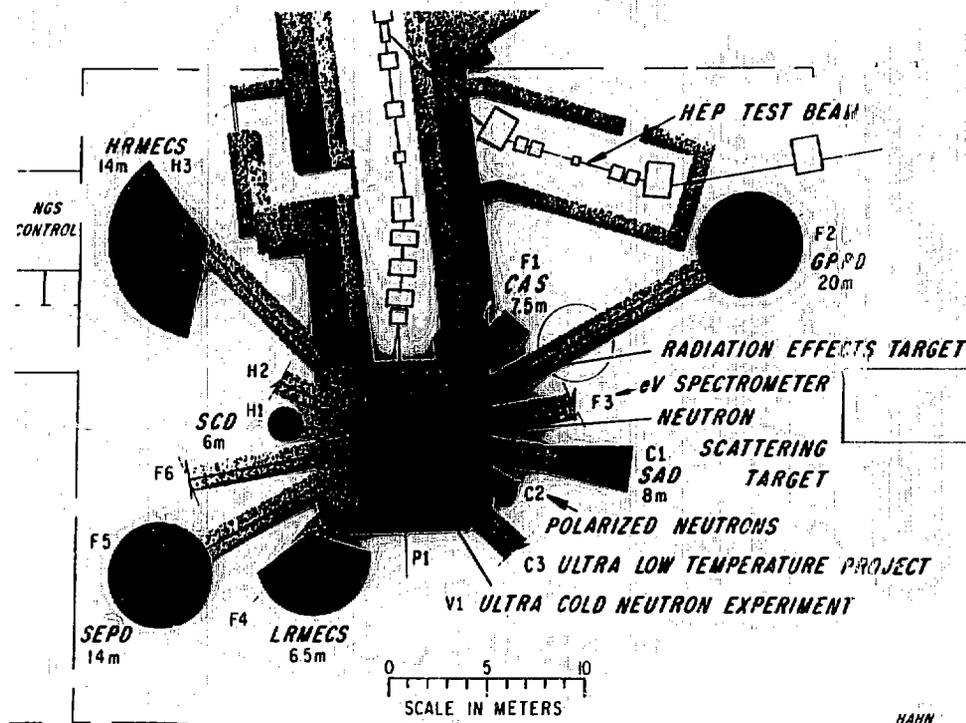
Figure 3. Raw neutron tof data over a limited range showing the evolution of structure of  $\text{LaNbO}_4$  as a function of temperature.  $T_c = 778\text{K}$ . Note that (b) and (c) are displaced 400 neutron counts from (a) for illustrative purposes. This tof range corresponds to  $1.813 < d < 2.576 \text{ \AA}$ .

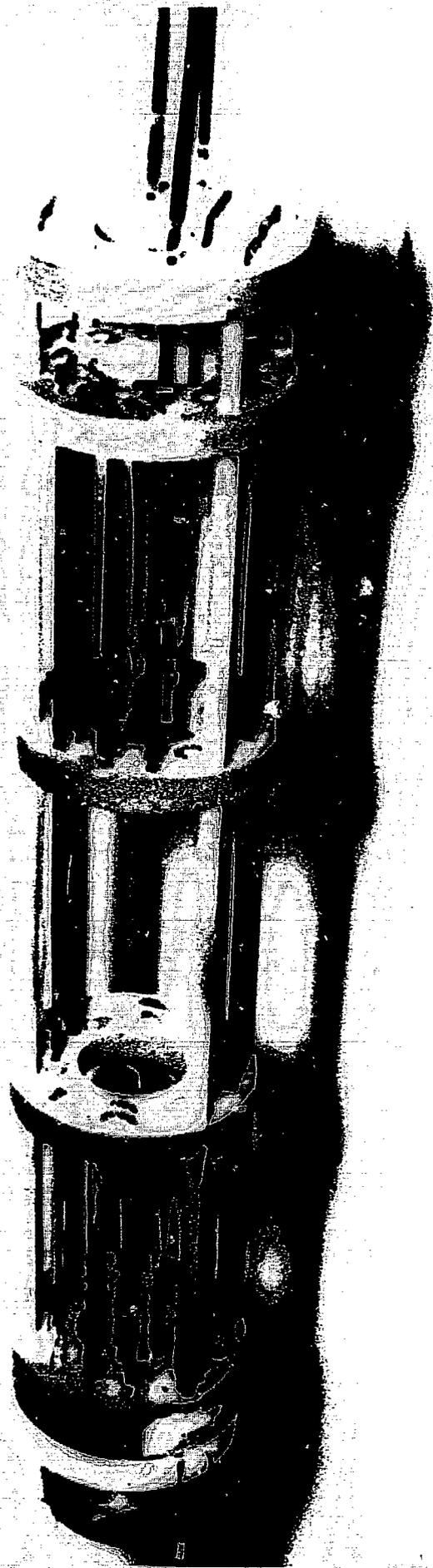
Figure 4. Furnace attachment for the Laue tof single crystal diffractometer at IPNS. The furnace has been operated at  $T=1040\text{K}$ , at atmospheric pressure.

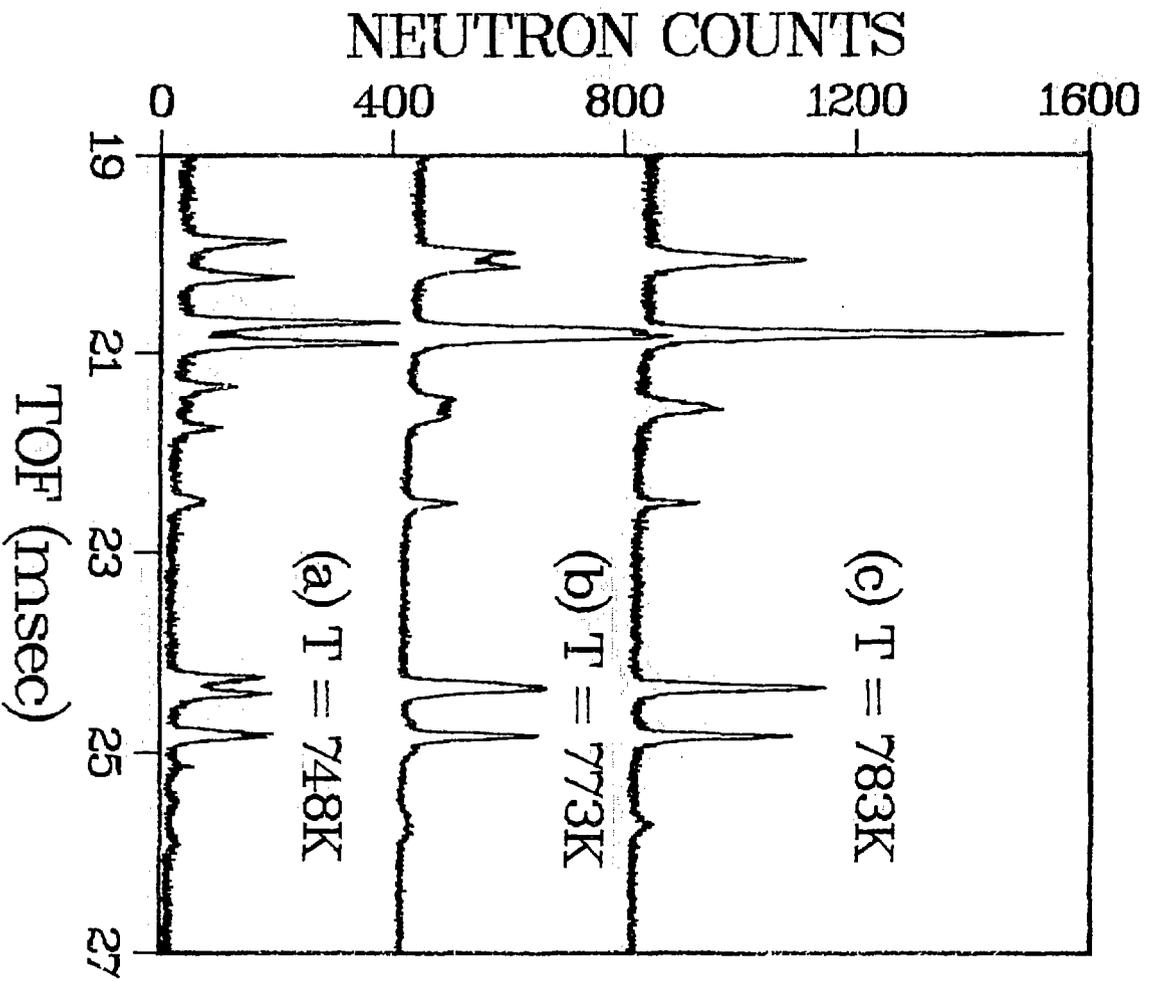
Figure 5. High pressure cell for diffraction studies under pressure to 30Kbar. A  $2\theta=90^\circ$  scattering angle provides minimal interference peaks from cell materials.

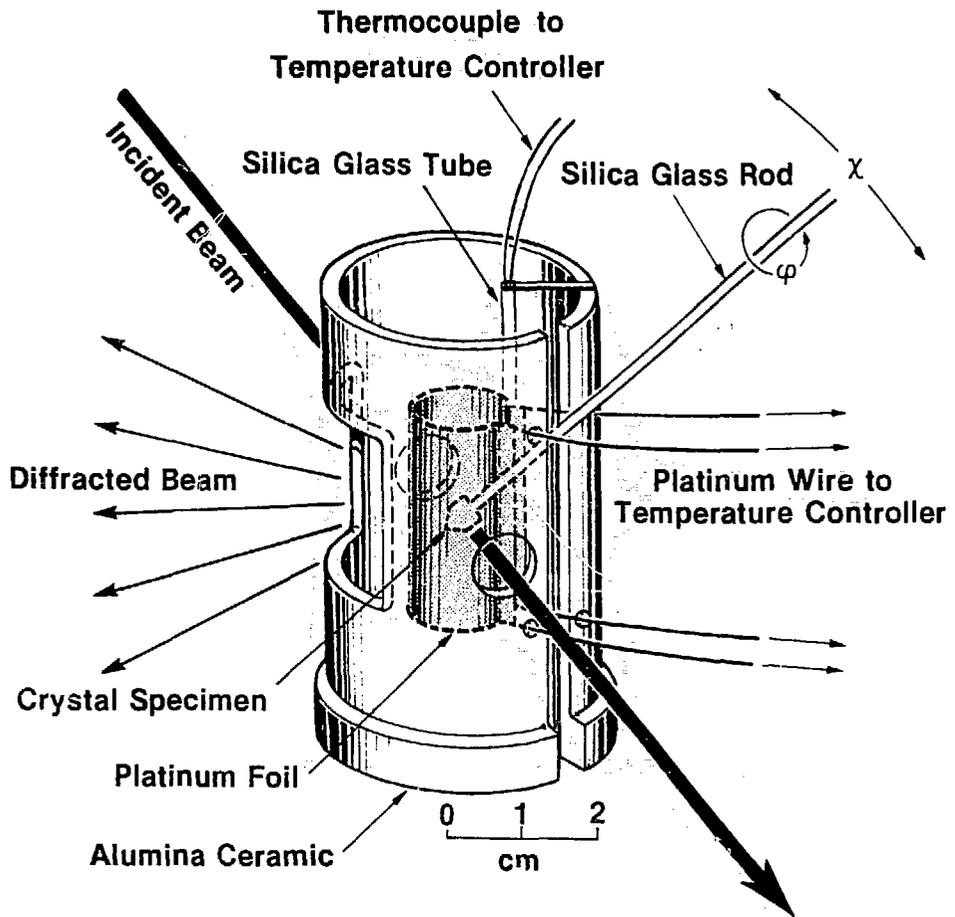
Figure 6. Neutron counts vs tof for the (220) Bragg peak from Si (a) and (c) show single detector response from extreme angles in a 20 detector band centered at  $2\theta_0=152^\circ$ . (b) Show the results of time focussing over this extended solid angle of detectors.

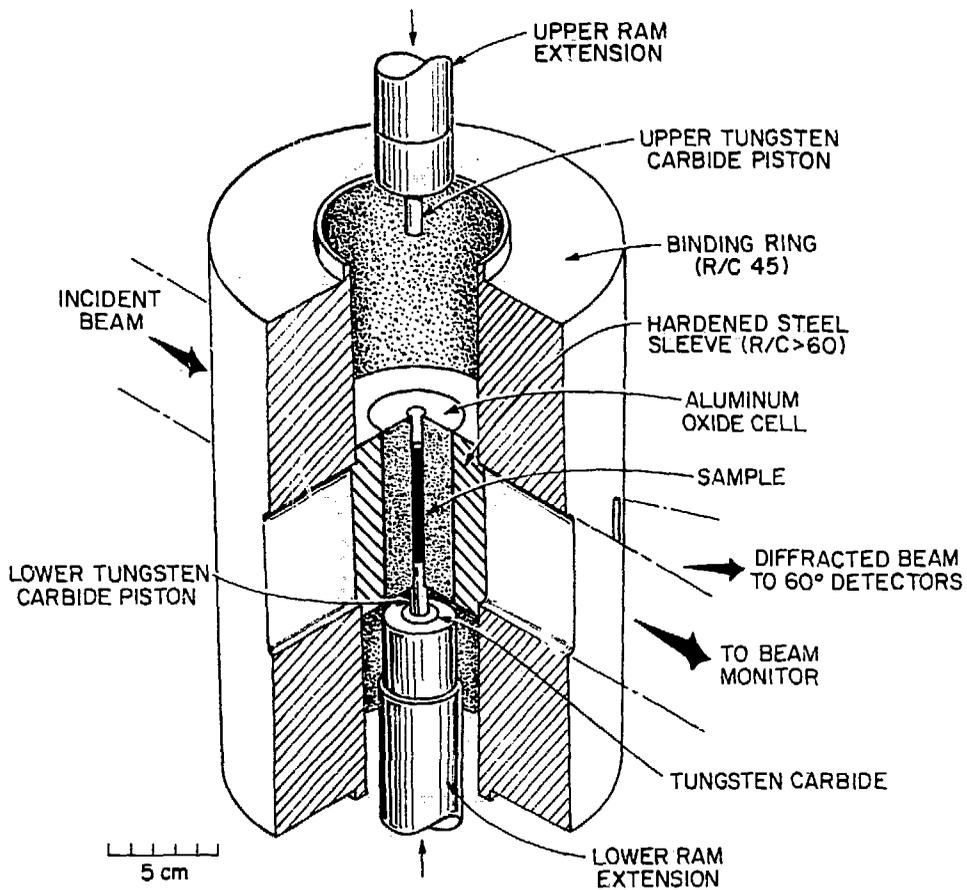
Figure 7. Sample response function as a function of time. Source neutron pulses occur at time interval,  $\tau$ . The time range,  $\Delta t_s$ , is based on detectors over the scattering angle range  $150 < 2\theta < 60^\circ$ , and represents a constant d-spacing in the sample.











**PRESSURE CELL FOR NEUTRON DIFFRACTION STUDIES OF SAMPLES UNDER PRESSURES OF UP TO 600,000 lb/in.<sup>2</sup>**

