PULSED NEUTRONS: ONE YEAR OF EXPERIENCE WITH THE
NEW SOURCE AT ARGONNE NATIONAL LABORATORY

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NOTICE

ABSTRACT

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The Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory is a spallation neutron source based on a 500-MeV proton accelerator operating at 30 Hz and with an average proton current of ~ 10 μ A. Neutron-scattering instruments for elastic scattering include two powder diffractometers, a single-crystal diffractometer based on the Laue method and employing a large (30 x 30 cm) position-sensitive scintillation detector, a small-angle diffractometer using a position-sensitive detector, and a polarized-neutron diffractometer which will utilize the spin-refrigerator device to obtain a beam of white polarized neutrons. For inelastic scattering we presently have the crystal-analyzer spectrometer and two chopper spectrometers capable of providing monoenergetic incident neutron beams of between 100 and 600 meV.

From its inception IPNS has been operating in a user mode and the selection of experiments is made by a Program Committee twice a year on the basis of the scientific merit of submitted proposals.

MASTER

Invited Review Paper for Yamada VI Conference on Neutron Scattering, Hakone, Japan, 1-4 September 1982.

1. Accelerator System at IPNS

The proton accelerator at Argonne National Laboratory that is part of IPNS consists of an H^- source, a 750-keV preaccelerator, a 50-MeV Linac, and a rapid cycling synchrotron capable of accelerating the protons up to 600 MeV. The accelerator operates at 30 Hz. The protons are then extracted and impinge on uranium targets. Full details of the accelerator systems may be found in References [1-3]. What is important for neutron science is the performance of this machine and since the start up in November 1981 this has been exceptional. From November 1, 1981 to August, 1982 it has delivered protons for a total of 2,985 hours at an average current of 8.02 μA and an operating efficiency of 89 %. Total operating time this fiscal year (up to October 1, 1982) is limited by the budget to $\sim 3,670$ hrs. This year the accelerator has operated at 400 MeV with the average current yields shown in Figure 1. After the initial test periods reliability was found to be too low at 500 MeV so a decision was made to drop the energy to 400 MeV. Tests in May 1982 have shown that the problems present earlier have now been corrected, and 450 MeV operation is planned for October 1982. Improvements to the accelerator system are continually taking place. Next spring we will be installing a new ion source, which has passed all its tests here at ANL, and this, together with the increased energy, should increase our neutron flux by $\sim 2\frac{1}{2}$ times within the next year.

2. Target and Moderators

The targets in both the neutron-scattering and radiation-effects facilities are both Zircaloy-2-clad depleted uranium. All tests and calculations have shown that uranium is the preferred target material in

spallation neutron sources and ANL has developed a special process for fabricating these. At this time IPNS is the only running facility with uranium targets in the world.

For the radiation-effects facility the neutrons are not moderated since the object is to obtain fast neutrons for damage studies. Since the focus of this Conference is neutron scattering nothing further will be said about the radiation-effects facility. The interested reader is referred to Brown et al [4] and Reference [1].

For neutron scattering targets, calculations (and experiments; see for example, the report by Professor Ishikawa in this Proceedings on the KENS pulsed-neutron facility) have shown that a cooled moderator system gives more intensity over most of the thermal spectrum and also a narrower pulse width. Our initial design at 25K and fabrication of a cooled (liquid hydrogen) moderator had a number of unforeseen problems, but a modified design with methane at ~100K has now been tested and will be installed in September 1982. This will improve the capability of a number of instruments, most notably the small-angle diffractometer and special experiments on polarized neutrons, ultra-cold neutrons and the ^3He project. (See below for descriptions). At the present time a polyethylene moderator at room temperature is being used. The pulse width at 100 meV is ~5 μs , and the peak thermal flux is $\sim 3 \times 10^{14}$ n/cm²/sec.

3. Neutron Science - Instruments Available

The parameters of the neutron scattering instruments are shown in Table I. A schematic of the experimental facilities is shown in Figure 2.

(a) Powder Diffractometers

From the first prototype spallation source up to the present day it has been clear that these machines would open up new areas of research. There are two powder diffractometers at IPNS, as the table indicates, and both are fully operational. First, the abundance of epithermal neutrons has allowed measurements out to much higher Q values, possibly up to $\sim 100 \text{ \AA}^{-1}$, and second, the short pulse width, together with long flight times, has allowed new standards of resolution to be attained. For example, both instruments have a resolution of $\Delta Q/Q \simeq 0.003$, which is independent of Q . At the present time three main areas of study have been pursued: (i) Structural work. At present the heaviest demand is for this area, and since a data set can be collected from a reasonable ($\sim 5\text{g}$) size sample in ~ 24 hrs the machines service a good number of users. In fact about half our present users fit into this category. What is of great importance is that the software package for handling this data is "on-line" at ANL. At the heart are the routines TOFPRP and TOFLS (written by R. B. von Dreele of Arizona State University and used extensively at Argonne over the past $2\frac{1}{2}$ years) which perform full-matrix least-squares refinement of crystal structure and peak shape parameters (Rietveld analysis) based on powder data. Programs to determine Bragg reflections for a given structure, to calculate Fourier syntheses, to calculate distances, angles and associated standard deviations from refined structures and to illustrate the atomic arrangement of a given structure are available. Users who have stayed an extra day or two after data collection have been able to leave with nearly complete Rietveld refinements. In addition, we have run a short course on Powder Diffraction and Rietveld Analysis at ANL from July 13-16, 1982.

(ii) Glasses, liquids and amorphous systems. For these studies the high Q capability is particularly important and this has already been exploited in a study of P_xSe_{1-x} glasses by Misawa, Price, and Susman. Another interesting application of the powder diffractometers was in determination of the magnetic scattering from an amorphous ferromagnet $Fe_{0.82}YB_{0.18}$ by Guttman, et al. Here the experimenters used banks of detectors placed symmetrically left and right of the incident beam, and applied a magnetic field $\vec{H}$ so that for one set of detectors $\vec{Q} \parallel \vec{H}$, and for another $\vec{Q} \perp \vec{H}$. Under these conditions the magnetic scattering appears in the $\vec{Q} \parallel \vec{H}$ detectors only and can be separated out. Note that with the time-of-flight method this condition is true for all $\vec{Q}$.

(iii) Measurements of residual grain interaction stresses in deformed alloys. MacEwan et al have exploited the high resolution at all Q values to observe the shifts of individual peaks after materials have been permanently strained. They estimate that residual bulk strains of order 10^{-5} can be detected using the high resolution configuration. Some experimental results for this work are shown in Figure 3, see MacEwan et al [5].

(b) Single-Crystal Diffractometer

This instrument, based on the wavelength-resolved Laue method, uses a 30x30 cm position sensitive 6Li -glass scintillation detector developed by M. G. Strauss and others in the Electronics Division at ANL [6]. The smaller-scale prototype built up at ZING-P' was the first of its kind. As this technique is capable of viewing large portions of reciprocal space it has a wide variety of potential

applications. The first experiments have concentrated on crystallography and the crystal structure of $\text{Mn}(\text{CO})_3 (\text{C}_6\text{H}_5\text{CH}_3)$ at 25K was solved by a joint group from ANL and the University of North Carolina at Chapel Hill [7]. The low-temperature structure was solved independently by direct methods - to our knowledge, the first such case with time-of-flight data.

Other types of experiments which are being performed with this instrument include searches for diffuse scattering, satellite peaks, and superlattice reflections, see Figure 4. The versatility of the instrument is certain to make it particularly attractive for these latter studies.

(c) Small-Angle Diffractometer

The SAD is another recently-developed machine. As such, one expects to encounter new challenges, and the most difficult one is to diminish the background scattering from fast neutrons. The instrument has a 2-dimensional gas-filled proportional counter that sits directly in the incident beam, but the background is now a factor 10^6 lower than the direct beam flux. At present the minimum usable Q is limited to $\sim 0.02 \text{ \AA}^{-1}$; however, once the cold moderator is installed, the Q range will be 7×10^{-3} to $0.35 \text{ \AA}^{-1}$. Experiments are being conducted on both metallurgical and biological samples. One early experiment after the test period has been to try to study the critical scattering from Fe near T_c . The object of this experiment, which has been proposed by F. Mezei of the Institute Laue Langevin, is to use the large wavelength band to make the direct integration

$$\int S(Q, \omega) d\omega = S(Q)$$

and thus obtain the true critical exponents in Fe. Such an experiment is difficult at a reactor because of the limited bandwidths available. Early results are encouraging.

(d) Chopper Spectrometers

The inelastic scattering instruments at IPNS are of special interest because they attempt to exploit in a direct way the high epithermal flux, which is a unique capability of spallation sources. Both these machines run in the so-called 'direct' geometry, i.e. the incident energy E_0 is defined. So far runs have been made with $E_0 = 160$ and 500 meV and momentum transfers from 0.1 to $20\text{\AA}^{-1}$. Measurements of the vibrational densities of states of amorphous SiO_2 and amorphous P have been completed. The electronics for the phasing of chopper and accelerator have been improved and the time in which the accelerator-chopper phase relationship is acceptable ($t_0 - t_c < \Delta T_c$, typically $2 \mu\text{sec}$) is now essentially 100%.

The second chopper machine is now installed and initial tests have been run. Of particular importance is that we are able to run two choppers simultaneously, which presents a complex phasing problem since only one chopper can be used to trigger accelerator extraction. Tests have now shown that two (or more) choppers controlled by a fixed-frequency oscillator can be maintained in acceptable phase relationship with the accelerator.

Experiments on the chopper machines include two important classes of experiments. The first, some form of electronic spectroscopy, needs high-energy at small momentum transfer to minimize the electronic form factor [4]. Experiments of this type have been done on Ce-Th alloys, UAl_2 , and Cr. The second may be characterized

roughly as the neutron analog of Compton scattering in which we wish to have both high energy and momentum transfer. Then the impulse approximation is valid and one has scattering from individual nuclei. The quantity that can be obtained is $n(p)$, the momentum distribution. Such a quantity is of fundamental importance in the calculation of many properties of the system, but is very difficult to derive from first principles. Particular attention is centered on $n(p)$ for the quantum systems ^3He and ^4He . Hilleke and coworkers [8] measured the momentum distribution in hcp ^4He at two solid densities (see Figure 5) and also at one density in normal liquid ^4He .

As expected, the chopper spectrometers have been under great demand for experiments. At the last Program Committee meeting only 43% of the proposals on LRMECS could be accommodated. Although this situation may get better when HRMECS comes on line, the low percentage reflects both the long time required for these experiments and the high interest.

(e) Crystal Analyzer Spectrometer

This machine uses the 'inverse' geometry technique in which the final energy is defined by a cooled Be filter and focussed graphite crystals to be 3.6 meV. The time-of-flight technique is then used to determine the initial energy and thus the energy transfer is known. The CAS is being constructed primarily for studying vibrational modes of hydrogen in metals. Even though the CAS can be used effectively for other studies, we anticipate that most of the experiments will be done on metal-hydrogen systems.

4. Special experiments at IPNS

In addition to the experimental facilities described above that are open to the entire user community on an experiment by experiment basis, three proposals were accepted by the Program Committee in June 1981 for long-term assignment of beams. These are described briefly below. In each case they represent a considerable effort, often collaborative with other institutions.

(a) Nuclear magnetic ordering in ^3He at very low temperature

This experiment is designed to observe antiferromagnetic Bragg reflections from single crystals of solid ^3He below 0.001K. The facility is now in the final stages of assembly. A vibration-free support structure to hold the cryostat has been completed and the dilution refrigerator has been installed at IPNS. In previous testing before this installation it cooled below 0.006K. The nuclear cooling stage is now being installed. The sample cell with a single-crystal silicon window is being leak tested. Other necessary components such as filters, the chopper assembly, position-sensitive detectors, and shielding are now almost completed. Studies of solid ^3He crystal growth will begin soon, and the actual experiments later this summer.

(b) Polarized Neutrons at IPNS

An optical instrument is being installed for neutron reflection studies. The object is to determine the magnetic induction $\vec{B}(z)$ close to the surface of materials. In many instances $\vec{B}$ varies as a function of the distance z from the surface until it reaches a value

$\vec{B}_0$ for the bulk. The goal is attained by measuring the spin dependent reflectivity of the neutron beam by the surface, since this quantity is related by optical laws to $\vec{B}(z)$. The perturbation of the magnetic induction at the surface is detected if significantly different from the bulk over a region not smaller than $5 \text{ \AA}$, nor larger than $1000 \text{ \AA}$. Early experiments will be on Nb, ErRh_4B_4 and on Nickel.

An effort is also underway to test a neutron spin refrigerator, which will produce a white beam of polarized neutrons. The basic principle is that protons are polarized not by "brute force" as in the conventional system, but by coupling to precessing electronic moments. This system is described in greater detail by its originator, G. P. Felcher, at this Conference, so nothing further will be said here.

(c) Ultracold Neutron Experiments

The ultimate aim of this experiment is to measure the electric dipole moment (EDM) of the neutron as a test of time reversal invariance. A finite EDM would show failure of time reversal. A practical system for producing ultracold neutrons (UCN) at high density from a pulsed neutron source was demonstrated at ZING-P'. We now need to show that we can hold these neutrons in a bottle for 100 seconds or so. To be competitive with other measurements of the EDM we need a density of about 10 UCN/cc stored in our bottle. Work on this project has been delayed by the difficulties with the cold moderator, but is expected to continue after its installation in September 1982.

5. User Program

IPNS is a national user facility. What this means is that we encourage and actively seek use of the various instruments by outside users. To achieve this effectively we have developed the following policy:

- o Program Committee (chaired by a non-Argonne scientist -- majority of members from outside Argonne) will review experiment proposals and allocate time to optimize the production of good science.
- o Instrument Scientists will be allotted 25% of time on each instrument for checking, upgrading, calibration and their own experiments, remaining 75% will be allocated by Program Committee.
- o Some beams will be left free for special experiments in which all experimental equipment will be provided by the users.
- o Users will generally provide any non-standard equipment required (beyond conventional sample environment equipment).
- o Users will provide their own travel support (Argonne Universities Association may be able to help university users in special cases).
- o Neutrons will be provided free of charge for scientific experiments meeting criteria established by the Department of Energy.

- o Proprietary experiments may be scheduled with appropriate cost recovery according to the Department of Energy guidelines.

The question is how well has this worked? On the whole extremely well. So far (Nov. 1981 - June 1982) we have run 80 experiments. About 60 outside users have been involved with these experiments, and of these about 30 have actually been at ANL to do their experiments. This is a promising start.

Conclusion

Most people believe that heat transfer limitations prevent more intense reactors from being built for neutron science; the hope, therefore, is that further generations of spallation sources can have higher peak fluxes than the present high-flux research reactors and open up further fields for neutron science. Our objective at ANL with the IPNS source is to start probing those areas unique to pulsed sources - particularly the epithermal region of the spectrum - so that future instrumentation may be based on our experience. We have made a promising start along this line. At least 75% of our experiments could either not be attempted or be very difficult at reactor sources, e.g. the strain experiments, Laue technique, and ^4He experiment. It is, of course, too early to judge the value of these experiments in absolute terms, but the interest expressed in them by various sections of the scientific community is gratifying.

Another objective of the ANL program has been to build a major user community from the university and industrial research sectors. This too, has got off to a good start. The overall user community for neutrons in the U.S. is at present not a large one, certainly much smaller than that in Europe, so that much remains to be done. We are involving outside users from the very

beginning in many of our decisions and, at least so far, they have shown much understanding of the difficult task ahead. In the long term of course, the U.S. must build a sufficiently vocal user community, and prove the excitement of the science, before it can expect to obtain funding for very high intensity spallation source of the sort proposed in Germany and Japan.

Acknowledgements

In a program of this size it goes without saying that a great number of people are involved in a diversity of activities. Special acknowledgements are due to my predecessor, David Price, for so successfully laying the groundwork, to Jack Carpenter for his perseverance and dedication from the first ideas a decade ago, to Bruce Brown and Charlie Potts for running such smooth and efficient operations, and to Ken Kliewer and Walter Massey for their encouragement and support.

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Pulsed Neutrons: One Year at Argonne

Figure Captions

1. Accelerator Current at IPNS since November 1981.
2. Experimental facilities at IPNS.
3. Results of residual stress measurements on Zircaloy-2 specimens using the GPPD at the IPNS. Complete details of these results have been reported by S. R. MacEwen, J. Faber, Jr., and A. P. L. Turner, Scripta Met., in press. Notice in the figure that an anisotropic yield surface in Zircaloy-2 yields Bragg profiles that shift in response to a uniaxial macroscopic deformation. The sign of Bragg peak shift depends upon the sense of deformation and the crystallographic orientation of the particles. These results give an accurate measure of grain interaction stresses, in part because of the high resolution ($\Delta d/d$ fwhm=0.25%) using the GPPD.
4. This is a plot of the Laue pattern for a crystal of $K_{0.26}WO_6$ from an experiment with Professor Bridgett Krause from Northern Illinois University. The intensities are the sums over the wavelength range of 1.0 to $3.0 \text{ \AA}$ for each X, Y channel of the 30 x 30 cm scintillation detector. In the three-dimensional histogram (X, Y, time) there are 117 channels. All Bragg peaks, including a number of satellite peaks, are completely resolved.
5. Dynamic structure factor of hcp ^4He as measured by R. O. Hilleke and coworkers (Ref. 8) is shown at a scattering angle of 105.9° . Altogether, data were taken simultaneously at eight scattering angles (corresponding to Q in the range $12.2 \text{ \AA}^{-1}$ to $22.5 \text{ \AA}^{-1}$) and at two sample densities, $18.20 \text{ cm}^3/\text{mole}$ and $19.45 \text{ cm}^3/\text{mole}$ at 1.70 K. The Gaussian fit to the experimental data corresponds to a Gaussian momentum distribution. The raw data peak intensity is 1300 counts/channel.

FIGURE 1

IPNS-I

EXPERIMENT FACILITIES

FIGURE 2

FIGURE 3

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EXP. NO. 5 HIST. NO. 5

FIGURE 4

FIGURE 5

TABLE 1

IPNS-I EXPERIMENTAL FACILITIES

Facility (Instrument Scientist)	Assignment	NEUTRON SCATTERING			
		Range		Resolution	
		†Wave-vector	Energy	Wave-vector	Energy
Special Environment Powder Diffractometer (J. D. Jorgensen)	F5	0.5-40 Å ⁻¹	*	0.35%	*
General Purpose Powder Diffractometer (J. Faber, Jr.)	F2	0.5-100 Å ⁻¹	*	0.25%	*
Single Crystal Diffractometer (A. J. Schultz)	H1	2-20 Å ⁻¹	*	2%	*
Low-Resolution Medium-Energy Chopper Spectrometer (J. M. Carpenter)	F4	0.1-30 Å ⁻¹	0-0.5 eV	0.02 K ₀	0.05 E ₀
High-Resolution Medium-Energy Chopper Spectrometer (D. L. Price)	H3	0.3-9 Å ⁻¹	0-0.4 eV	0.01 K ₀	0.02 E ₀
Small-Angle Scattering Diffractometer (J. E. Epperson (a), C. Borso (b))	C1	0.001- 0.3 Å ⁻¹	*	0.004 Å ⁻¹	*
Crystal Analyzer Spectrometer (T. O. Brun)	F1	3-16 Å ⁻¹	0.02- 0.5 eV	3%	2%

* No energy analysis
† Wave-vector, $K = 4\pi \sin \theta / \lambda$
(a) Materials Science -- 3 Meter Flight Path
(b) Biology -- 8 Meter Flight Path

NEUTRON BEAMS FOR SPECIAL EXPERIMENTS

Beam Tube	Current Use	Flight Path Length (m)
F3	Vacant	6-70
C2	Polarized Neutron Exp.	6-40
C3	Solid He ³ Project	7.5-25
F6	Irradiations	6-20
H2	Irradiations	6-20
V1	Ultra-Cold Neutron Exp.	2.7-6.7

RADIATION EFFECTS

Facility (Instrument Scientist)	Description
Radiation Effects Facility (R. C. Birtcher)	Two vertical (5 cm ID) tubes with flux 1×10^{12} n/cm ² sec and one horizontal (3.8 cm ID) tube with flux 3×10^{11} for energy greater than 0.1 MeV at 8pA; capabilities for maintaining two samples at liquid helium temperature (4°K) and above

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