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ARGONNE NATIONAL LABORATORY

FROM THE EDITOR

The Intense Pulsed Neutron Source (IPNS) facility began operation in 1981. In 1983, the progress made at IPNS in instrumentation, computer hardware and software, scientific results, and user issues was reported in a publication called the *IPNS Progress Report 1981-1983*. Since that time, there have been four more of these reports published, covering the years 1983-1985, 1985-1988, 1988-1990, and 1990-1992. Susan Biggin served as technical editor for the first two IPNS Progress Reports, and I have performed this task for the last three. The now-familiar format of the IPNS Progress Report was established with the initial report and has remained basically unchanged, that is, until this report.

The *IPNS Progress Report 10th Anniversary Edition* is being published in recognition of the first ten years of successful IPNS operation. To emphasize the significance of this milestone, we wanted this report to stand apart from the previous IPNS Progress Reports, and the best way to do this, we thought, was to make the design and organization of the report significantly different. In their articles, authors were asked to emphasize not only advances made since IPNS began operating but also the groundwork that was laid at its predecessor facilities - Argonne's ZING-P and ZING-P' prototype pulsed neutron sources and CP-5 reactor. Each article stands as a separate chapter in the report, since each represents a

particular instrument or class of instruments, system, technique, or area of research. In some cases, contributions were similar to review articles in scientific journals, complete with extensive lists of references. Ten-year cumulative lists of members of IPNS committees and of scientists who have visited or done experiments at IPNS were assembled. A list of published and "in press" articles in journals, books, and conference proceedings, resulting from work done at IPNS during the past ten years, was compiled. And, archival photographs of people and activities during the ten-year history of IPNS were located and were used liberally throughout the report.

I would like to take this opportunity to express my gratitude to those whose efforts made the production of the *IPNS Progress Report 10th Anniversary Edition* possible. First, I would like to thank Diane Hoffmann and Beverly Marzec for their painstaking efforts in compiling and producing the lists of users and visitors, committees, and publications used in the report. I am deeply indebted to Mary Ann Forys of the Media Services Department of Argonne's Information and Publishing Division; she is responsible for "the look and layout" of the report, as she has been for the previous three reports that I have edited. And last, but certainly not least, many thanks to the authors of the articles in the report; the excellent material provided by them made my job a pleasure.

Frank J. Rotella
IPNS Division

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INTENSE PULSED NEUTRON SOURCE
ARGONNE NATIONAL LABORATORY

Operated by The University of Chicago for the U.S. Department of Energy

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A DECADE OF EXCELLENCE

In the course of its 10-year history, Argonne's Intense Pulsed Neutron Source has exemplified how a national scientific user facility should be developed and operated.

The design and construction of IPNS advanced neutron source technology and the techniques of neutron scattering. The upgrade of the uranium target and the addition of each specialized instrument contributed to the progress of neutron scattering as a science.

Even more important, this facility established new, higher standards of "user friendliness". It attracted researchers from disciplines not traditionally associated with neutron scattering, such as biologists and chemists. It was organized and staffed to provide maximum assistance to visiting researchers. Its reliability - more than 95 per cent availability of beam - has enabled users to plan confidently and gain maximum results from their time at the facility.

And it has provided an exceptional return on the taxpayers' investment. Operating under tight fiscal strictures throughout its history, IPNS was built with some components of a decommissioned accelerator at a fraction the cost of a "greenfield" facility. The array of specialized instruments was built up over 10 years of careful budgeting. IPNS has served more experiments for the time it was funded to operate than any other neutron facility in the nation.

The staff and the users of the Intense Pulsed Neutron Source can be proud of their decade of excellence. Argonne National Laboratory shares their pride

Alan Schriesheim
Director and Chief Executive Officer
Argonne National Laboratory

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MASTER

INTENSE PULSED NEUTRON SOURCE FACILITIES

LEGEND

IPNS I

- 1 CONTROL ROOM (IPNS I&II)
- 2 INAC
- 3 BOHNER SYNCHROTRON
- 4 EXPERIMENTAL AREA

IPNS II

- 5 ONLY INAC
- 6 INTENSITY SYNCHROTRON
- 7 R-T-B-B-B
- 8 TO 6 TATTERING TAPET
- 9 HENAI AITA
- 10 AT II EFFECT TATTER

CHAPTER

1

PULSED NEUTRON SOURCE DEVELOPMENT AT ARGONNE

J. M. Carpenter, IPNS Division, Argonne National Laboratory

The Early Years

In 1968, the Argonne Committee on Intense Neutron Sources, headed by Lowell Bollinger and on which I served as a left-over from the Argonne Advanced Research Reactor (A²R²) instrumentation group, found proton-driven pulsed spallation neutron sources to be the best identifiable, practical basis for a new class of slow neutron facilities. That committee was formed in response to the closing down of the A²R² project in the early stages of its construction and commissioned to identify the best means to advance slow neutron research at Argonne. Ingredients of this finding were the Canadian Intense Neutron Generator (ING) study of a steady spallation source concluded in 1966 and the development by Dimov in the Soviet Union of an efficient negative hydrogen ion source, news of which Ron Martin brought back from a visit there and which stimulated his conception of an intensity booster for the Zero Gradient Synchrotron (ZGS) based on a 500-MeV rapid-cycling synchrotron with H⁻ stripping injection. The conception by Ralph Fullwood of a pulsed spallation source for weapons research driven by protons from the 800-MeV LAMPF accelerator at the Los Alamos National Laboratory (LANL) and the demonstration of a uranium spallation target for nuclear physics research at the Nevada synchrocyclotron also reinforced the finding.

The ING study provided essential information on neutron yields from targets irradiated by medium energy protons. A significant advantage over all other practical methods of neutron production is that spallation neutrons are produced in large numbers (about 20 neutrons per proton) and very efficiently, that is, with relatively little accompanying heat, approaching 50 MeV per neutron in nonfissionable materials for proton energies of about 1 GeV.

This is to be compared to about 200 MeV per neutron for fission reactors and to much lower yield and much higher heating figures for electron and low energy charged particle reactions.

Moderators close by the primary neutron source are necessary to slow down the neutrons from the primary source, where they emerge with average energies of about 1 MeV, to energies of a few eV and less that are needed for the intended applications. The most significant problem at that time was that there was almost no information on the intensities of slow neutron beams that could be extracted from the moderators. Considerable information was available about the spectra and shapes of pulses from representative moderators, for example from the work of Rex Fluharty and principally from the work of Kingsley Graham, my student at the University of Michigan, but only Graham had measured absolute intensities, and his measurements applied to 14 MeV neutrons. In 1969, I worked out a preliminary estimate of the intensities available from a proton spallation source based on the planned ZGS Injector Booster, but the resulting figures were rather low and quite uncertain. I called it ZING, for ZGS Intense Neutron Generator, intending a how toward the Canadian effort which had foundered for want of appropriate accelerator technology.

During the academic year 1971-1972, when I spent a sabbatical leave in the Solid State Science Division at Argonne, Oliver Simpson, then division director, encouraged me to work more on the neutron source ideas. We reviewed all the crazy schemes that we could find or think of, still the spallation source came out on top. Dan Conner, then head of the neutron group, encouraged me to build a mockup of a spallation source and moderator to measure

the resulting intensities. I had in mind to resolve also the question that had appeared during Graham's work, that the boron carbide shielding we placed around the moderator to exclude room-return neutrons, increased the intensity of the beam from the moderator by a considerable factor; we had tentatively attributed this to its effect as a reflector, similar to a reactor reflector, but a poor one. Conner pointed me to the stock of beryllium blocks that were left over from the A²R² criticality experiments, and I borrowed a calibrated californium spontaneous fission source from Alex DeVolpi; arranged space in the A²R² mockup bay with help from Roland Arinhardt; put together cadmium-shielded, cadmium-filtered ²³⁵U-fall track-etch detectors; learned to count the tracks under a microscope with the help of Fran Lyon; and went to work. I built a mockup (Fig. 1) with a beam port and cadmium decouplers inside a beryllium reflector, which seemed to be the best material. Measurements for a moderator with and without the reflector showed an increase in the beam intensity of

Fig. 1 Sketch of the ZING mockup, 1972.

about a factor of ten due to the reflector! With Gary Manner (to keep me honest in accelerator matters), I wrote up the results in a little red-covered informal report, ANL/SSS-72-1. The patent people got wind of the result and I wrote up an application. The patent was granted for the reflector idea; I got \$1 for it and \$25 for the effort, as was the arrangement at the time.

The Development of ZING-P

With the enhancement of the reflector, the ZING idea looked good, even realizing that the more complex arrangements in a real source would cost some of the gain, and Bob Kleb and I worked out the concept of a research facility based on the ZGS Booster. So it stood until David Price and I convened a workshop in the Spring of 1973 to evaluate the scientific applications of ZING. Among others, we invited Motoharu Kimura, a nuclear physicist from Japan, who had already gained important experience with an electron linac-based source and neutron scattering instruments at Tohoku University. Moto urged that we build a prototype to test the ideas and returned after the conference with Noboru Watanabe. Kimura laid out (Fig. 2) and, with Bob Kleb, designed a tiny shielded house (Fig. 3) for the instruments; Watanabe helped with the design of a time-focussed powder diffractometer (Fig. 4), an idea I had worked out in the 1960's at Michigan with my student John Sutton for powders and with Kingsley Graham for single crystals; and Torben Bruun and Kurt Skold (Sturdevik, Sweden) developed ideas for a crystal analyzer (pulsed scattering instrument). Bob Kleb assisted with the target and moderator-reflector design, and Kimura and I worked out the principal shielding requirements. The target was one-half of a lead brick (Fig. 5), to which Bob Kleb and Bob Stuliuk soldered a copper tube to carry off the heat. The Laboratory assigned \$30,000 for the project and appointed Tom Benfield, then director of the CP-5 reactor, to oversee the work. Jamie Tulbov assisted with the planning, Chuck Bally oversaw

Fig. 2 Motoharu Kimura's sketch of the shielded house for ZING-P. The date is 18 May 1973.

Fig. 3 ZING-P experiment house, March 1974

the installation of the house. Gus Schulke helped coordinate activities with those at CP-5. We completed the prototype by January 1974 - It was built on top of the earth-mound shield between the Booster and ZGS, in the area called "Skunk Hollow". We called it "ZING-P", "P" for prototype. We had to scurry up and down the muddy bank that winter to get to the instruments. The prototype accelerator, Booster I, was ready at the same time, put together by Jim Simpson and the accelerator technical staff from parts moved for the purpose from the Cornell 2-GeV electron synchrotron. The first moderators were slabs of room temperature polyethylene, with cadmium sheets (Fluharty's idea, explored at length by Graham, so we know how to manipulate spectra and pulse shapes in polyethylene moderators) inserted to control the pulse width. These were our first models of target and moderators. ZING-P had only two, nearly-vertical beam holes (Fig. 6) and operated for three periods during 1974 and 1975, when its operation ended with the completion of tests on the prototype Booster. The data acquisition system (DAS) was a small Nuclear Data multichannel analyzer (Fig. 7), of which we were very proud. Our first work was to determine the intensities, which worked out according to

expectations. And the work with the instruments proved the basis for optimism about the larger facility. Moto Kimura spent extensive periods after his "retirement", working with us at Argonne.

The ZING-P Phase

We prepared a request for construction funds for ZING, based on the plan worked out by Bob Kleb. I worked out the fundamentals of the design of a uranium target. Argonne neutron scientists prepared ideas for instruments for the new facility. "Schedule 44" (requests for construction funds)

flow to Washington in numerous forms, and in 1974, the powers encouraged us to think up a larger-scale facility - ZING was not powerful enough. Jim Simpson, Bob Kustom, and others took up the challenge and conceived the 60-Hz High Intensity Synchrotron to deliver 500 μ A of 800-MeV protons. During these days, we received much encouragement and support from Don Zaffarano, president of the Argonne Universities Association, then Argonne's operating contractor. When Zaffarano left, his replacement, Paul McDaniel, took me aside to recommend that we

Fig. 4 Early powder diffractometer at ZING-P.

Fig. 5 Bob Kleb's drawing of the ZING-P had target. Note that the dimensions are exactly one-half the size of a standard had brick.

Fig. 6 Vertical section through the shielding of ZING-P.

Fig. 7 (Left to right) George Ostrowski, Mel Mueller, Bob Beyerlein, Selmer Peterson, and David Price with the Data Acquisition System (a Nuclear Data multichannel time-of-flight analyzer) inside the ZING-P experiment house in March 1974.

call the new higher-powered facility by a name that could not be pronounced - "ZING" was too cute. I decided to call it Intense Pulsed Neutron Source, IPNS. We prepared and submitted more Schedules 44. All this time, I was commuting between Ann Arbor and Chicago. John King, my colleague at Michigan, encouraged me to work out an effective way to do small angle scattering at a pulsed source; "It's very important!", he said. I still have notes of my thinking recorded on placemats from Howard Johnson's at Benton Harbor. Finally, I decided to resign my position at Michigan and moved to Argonne in January 1975, to devote full time to the work on the neutron source. Just then, we were given \$175,000 to prepare a conceptual design, and the work began in earnest. Sam Warner became our strong supporter, and he and I convened another workshop, held in June 1975 to evaluate the potential of the IPNS. Sam made up the little blue cards of neutron data that we all still use, which he distributed at the workshop; an updated version of this card is being prepared for distribution as part of the IPNS tenth anniversary commemoration. Marcel Barbier joined us for the summer to work out shielding requirements.

One day in the early 1970's, then-Laboratory Director Bob Sachs sent Eugene Wigner, at Argonne for a visit, to see me, no doubt to see whether Wigner could be convinced that pulsed sources were a viable idea for neutron scattering applications. Soft-spoken and kind, he asked his questions, and I launched a lengthy explanation of how they worked, peak vs time-average fluxes, phase space densities, time-of-flight methods... He interrupted part-way through - "Never mind, I understand," he said. Perhaps this saved the project.

Meanwhile, we worked out modifications of ZING-P to provide for more instruments, adding another moderator and, at first, two horizontal beam holes, then later, a third (Fig. 8). The modified prototype was to be ready when the new Booster II accelerator was completed. Bob Klob built a

Fig. 8 ZING-P moderator-reflector arrangement, before filling with beryllium blocks.

water-cooled tungsten target and designed a pneumatically driven actuator which allowed us to share the proton beam with the ZGS. Drawers in the reflector-shield assembly allowed us to change moderators at will and to insert items close to the source for tests. We improved the shielding with help from Marcel Barbier. We built several new instruments. Kurt Sköld, Kent Crawford, and Sow-Hsin Chen (Massachusetts Institute of Technology) designed a crystal analyzer spectrometer. Selmer Peterson conceived, and with Art Reis, built a single crystal diffractometer, and Mel Mueller, assisted by John Faber and Chuck Berse, devised a small angle scattering instrument to share the same beam and the same detector; Frank Linkszus and Paul Day arranged hardware and software to enable data to be recorded on the Chemistry Division Sigma-5 computer. Jim Jorgensen developed a high resolution time-focussed powder diffractometer, Chuck Polizzari, George Ostrowski, and the time-of-flight experts from the Solid State Science Division produced a chopper spectrometer, getting into trouble because someone else wanted the privilege of stacking the thousand solid concrete blocks of the shield. I recall a crucial discussion with the accelerator people, concerning whether

the chopper should be accelerated and decelerated to keep in phase with the synchrotron, or whether the synchrotron should be phased to the chopper. Just this once, I was ready with an effective argument: the "Q" for the accelerator is about 30, the equivalent factor for a chopper is about 10^7 , furthermore, both store about a megajoule of energy. This convinced the electrical engineers that it was better for the accelerator to track the chopper and began the long, cooperative development toward our present system, in which eight choppers run in phase simultaneously and the chopper drive system and the accelerator share the task of adjusting to fluctuations in the power line phase. Other reports in this volume describe how the aforementioned instruments evolved into the current instruments at IPNS.

The modified prototype with most of its instruments was completed in November 1977, ready for startup with Booster II. We called it "ZING-P". Bob Kleb and Henry Thresh designed, and Henry produced, a Zircaloy-clad uranium target in the form of a solid cylinder. Arduous safety reviews were required before it could be tested. Les Colomani, a member of the review panel, was hard to convince - "I don't want to be there when the blue flash comes!", he said.

The uranium target operated successfully during the last year of ZING-P, but not without worries. Silver-110 (possibly a fission product), was found in the water cooling stream, but no other fission products were detected; we feared that the cladding on the target had corroded or split open. Finally, we found that the surge tank in the cooling system, made from a vessel formerly used in a photographic film developing system, though it had been thoroughly cleaned, was contaminated with silver which dissolved in the coolant water and became activated. Sometimes we found xenon-135 gas (another fission product) in the coolant water - it would come and go. The explanation was that the threads on the feedthrough to the thermocouple in the center of the target leaked some gas when the target became hot. Next time, we would clad the thermocouple well. As ZING-P got underway, we began a user program, inviting scientists from outside Argonne to share the use of the instruments.

Bob Kleb and Bob Staffuk designed and built a naturally-circulating liquid hydrogen moderator for tests, using a second-hand helium compressor. This provided us our first experience with cold neutrons. Ted Postel and David Mildner (National Institute of Standards and Technology) helped measure the spectrum and pulse widths. We needed to develop technical data to support IPNS design. In 1978 and 1979, Al Knox, Bob Kleb, Moto Kimura, and I measured the energy deposition in the tungsten and uranium targets, using Larry Greenwood's spallation cross sections for proton current. (The aluminum foil used was a very pure form used by airlines to bake potatoes; the foils were free and potato-sized.) Less heat appears in the tungsten than the energy brought in by the proton beam - the difference we measured is the energy required to liberate neutrons from the tungsten! Jim Bailey began to help around this time also. He, Al Knox, and I worked on the interpretation of the thermocouple time response to

derive from it the local power density. Jim later based his Master's thesis on this subject. Kimura, Mildner, and I determined the heating rates in moderator, decoupler, and reflector materials. Tom Worlton and I made absolute measurements of the neutron beam intensities using resonance gold foil activation techniques. Dick Praef and Shashikala Das modified the VIM computer code to calculate the neutron intensities, which were in satisfactory agreement with measurements.

Of course, the time-average flux at the moderator surface in pulsed sources is rather low and the peak flux during the pulse is rather high (why they are effective!). Realizing that this implied that the peak phase space density is correspondingly high and that the then-popular attempts to measure the electric dipole moment of the neutron depended on the peak phase space density, Tom Dombek, Roy Ringo, Vic Kruhn, Sam Werner, and others conceived a mica crystal "Doppler shifter" to convert the pulsed, long-wavelength neutrons into a steady source of ultra-cold neutrons that could be trapped in a bottle at (in principle) the same phase space density as the peak at the source. Bob Kleb designed the required apparatus, and it was operated on the vertical beam from the hydrogen moderator in ZING-P¹. The principle, an application of Liouville's theorem, was established, but it required one tour de force effort to keep the instrument running and another to provide an effective bottle. We never detected bottled neutrons.

In 1977, it happened that Rex Fluharty, then at LANL, Loe Hobbis from the Rutherford-Appleton Laboratory (RAL), U.K., and Moto Kimura were simultaneously at Argonne. By then RAL faced the shutdown of NIMROD, a situation similar to that of Argonne and ZGS, and thinking I had begun about converting the facilities to a spallation source; the WNR facility was being built at LANL; and the Japanese were constructing a pulsed spallation source at the KEK High Energy Physics Laboratory in Tsukuba. We decided to form a

collaboration among our laboratories to foster the development of pulsed spallation sources. Torben Brun and I worked out the name, ICANS, International Collaboration on Advanced Neutron Sources. ICANS remains a wonderfully useful forum and has met eleven-and-one-half times since its first meeting held later that year at Argonne.

The IPNS Concept

The proposed IPNS project had two phases, IPNS-I and IPNS-II; IPNS-I was to be built early and to operate for several years to provide further experience needed for IPNS-II. Estimates of the cost of IPNS rose as our design evolved. There was unrest in the materials science community. And the synchrotron light people began to agitate for construction of more powerful photon factories. Commitments met. The Laboratory assigned Norm Swanson, an experienced nuclear facilities construction engineer to oversee the construction project. The authorities asked for ever more paper, and more Schedules 44 flew to Washington. We were asked to tell the cost of the IPNS-I prototype separately from the cost of IPNS-II. I recall a critical discussion of this issue with David Price, then director of the Solid State Science Division, and Bob Kleb. We recognized the risk that if we once separated the two components of the project, only IPNS-I might be funded. But if we did not respond, we might lose all. We complied, but not wisely. We honestly determined the cost of IPNS-I alone, on the basis that we would be continuing with IPNS-II; without the IPNS-II activity, IPNS-I would cost more. In the midst of all the fuss, the feared result came about. IPNS-I was funded in 1978 for \$6.4M, our estimated cost, but no allowance was made for the fact that there was no IPNS-II! This required another agonizing decision, whether to accept the project or not, realizing the difficulty of the funding shortfall. The people in Washington understood after all, and accepted our idea of interpreting the IPNS-I project to represent only facilities construction, without instruments, and accepted a

separate proposal for building the initial complement of instruments, another \$3M. Now, as do all engineering estimates, this included a rational contingency allowance, about 25% of recognized costs. Unbelievably, the contingency was not allowed in the amount granted, but we accepted the \$2.4M and, with grim determination, continued the work. Our schedule called for completion of IPNS-I in the Spring of 1981. Although the project was now far beneath his abilities, Norm stuck with the job. Bob Krueger assisted in project management. Bob Hagaman acted as engineering liaison.

IPNS-I was to have two targets, one for neutron scattering, with three beryllium-reflected moderators and twelve neutron beams, the other for radiation damage research, surrounded with a lead reflector, and accepting two vertical cryogenic irradiation thimbles. The Radiation Effects Facility (REF) was provided to support the work of the small but vigorous fast neutron radiation damage group and their colleagues, led by experienced and feisty Tom Blowitt. IPNS-I would have water-cooled, Zircaloy-clad, depleted uranium targets in the form of stacked disks; IPNS-II was to have had a steel-clad, depleted uranium or uranium-molybdenum alloy target, cooled by liquid NaK. Hans Fauske and Mike Grohms checked out my calculations. The moderators of IPNS would be liquid methanol, reflected by a liquid nitrogen-cooled beryllium reflector, decoupled with gadolinium. Both facilities were to benefit from the closing down of the ZGS accelerator and of the CP-5 reactor, which took place in 1979. Ultimately, we estimated that the "junk" we inherited would have cost at least \$35M (in then-current dollars) to replace new.

Building IPNS

As the construction project got underway, Bruce Brown joined the group as operations manager; Gus Schulke joined after the closing of CP-5; and David Price signed on as director of the project. I became the technical director. Don Bohringer and Dave Leach

moved from the High Energy Physics effort to IPNS and became the keystone of our technical crew. The neutron scattering scientists marshalled to support the instrument design and construction effort, and Tom Blewitt and Terry Scott of the Radiation Effects group took part not only in the design of their facility, but also provided invaluable support for the cryogenic moderator systems design. Al Knox took responsibility for coordinating the design and fabrication work on the scattering instruments, the DASs, radiation monitors, and the building to house the DASs (Building 399). John Ball became Norm Swanson's very busy assistant. I recall a long period of difficulty with the contractor that was to supply the moderator cooling systems. The reason finally came out - somehow instructions had been passed to them that the system was to keep the moderator containers cold using liquid methane, but somehow no mention had been made of the requirement that they be kept *full* of liquid methane! Such are communication problems that arise in the hot, busy times of a construction project.

Habib Ahmad, Jim Bailey, Frank DeSousa (Illinois Institute of Technology), and Don Henley, were responsible for the thermal/hydraulic design of the targets and cooling systems. Ben Loomis, Henry Thresh, and Gary Fogel developed the target fabrication process, a unique hot isostatic pressure (HIP) bonding of uranium metal cores in Zircaloy cans. Al Hines led the target disk production program. Bob Kleb worked out the clever chain-link system for moving the target. (His 1/4-scale model is still on display in the IPNS Experiment Hall [Building 375], as is his Styrofoam model of the target-moderator-reflector system.) Tom Worton assisted with non-trivial calculations. Kent Crawford moved from the Solid State Science Division to take responsibility for the instruments and DASs. Tom was responsible for the Digital Equipment Corp. (DEC) VAX central computing systems. Kent devised and, with Joe Haumann and others, built the DEC-computer-based

DAS, linked to the VAXes.

Henry Hubony, a Purdue graduate student, adapted the VIM code for the VAX computer and provided useful systematic studies of the energy- and time-dependent neutron fluxes in the moderators.

ZINC-P' continued operating until 4 August 1980, when it was necessary to shut it down to install the beam transport system connecting the Booster accelerator to IPNS-I. Its operation provided not only a wealth of needed data for IPNS-I and its instruments, but also produced a significant body of scientific results. During the construction years, we lived in the cozy Building 372.

IPNS Begins Operation

IPNS-I was completed on schedule, and the first proton beam was delivered to the REF target on 5 May 1981. First beam on the neutron scattering target was delivered 4 August 1981. Users were invited in after the first scattering instrument commissioning activities. In November 1981, in 1982, Gerry Lauder returned from Institut Laue-Langevin (ILL), France to assume the project leadership, and David Price returned to doing science. Charlie Potts, as accelerator manager, Frank Brunwell, Yanglai Cho, Jim Norom, and others of the accelerator team worked continuously to identify the limitations of the accelerator and improve it. We renamed the Booster the Rapid Cycling Synchrotron (RCS) because, with ZCS gone, there was nothing to boost anymore. When IPNS started up, the accelerator was delivering 3 μ A of current. As this is written, the RCS is routinely delivering 15 μ A! And its reliability is near 100%!

TNTQFS, the hybrid chopper spectrometer built by David Price, Mike Rowe, Bob Kleb, and George Ostrowski, was moved from CP-5 to IPNS and adapted to the pulsed source. Chuck Pelizzari, then I, took over its commissioning. Chuan Loong and David Price later assumed responsibility for the chopper instruments. The Small Angle Diffractometer, using the 20×20 -cm² area detector built by Manfred Köpp at Oak Ridge, was

moved from ZINC-P' to IPNS.

Ernest Epperson and Chuck Borse took over its development, and Frank Williamson assisted with software development. Mike Struss, Frank Lynch, and Raul Branner developed a neutron adaptation of an Anger camera for use on the Single Crystal Diffractometer; Ari Schultz and Jack Williams assumed responsibility for the new machine. Jim Jorgensen and John Fisher designed new powder diffractometers, and Jim introduced the idea of "electronic time focussing", possible now with the powerful computer-based DASs. Kurt Sköld, Mary Vrtis, and others built a special purpose diffractometer to measure the nuclear antiferromagnetic ordering in solid ³He - we were beaten by the French effort at the Melusine reactor. Kent Crawford and I tried and tried to develop a small-Q resonance detector spectrometer - we never could get the background down to acceptable levels. Chopper spectrometers have taken over where we had expected they could not go. Torben Brun, Bob Kleb, Tom Erickson, and Bob Steffuk designed and built a new quasielastic spectrometer, QENS.

Moderators

The first moderators were uncooled, graphite-reflected polyethylene because the cold moderator assembly was not quite ready. In the Fall of 1982, the cold moderators were installed and the results revealed a serious error on my part. I had called for them to be assembled without poisoning, a compromise in favor of intensity at the sacrifice of narrow pulses. I recall the circumstances of that decision, made late during the construction activities. The Booster accelerator had delivered only 3 μ A of proton current during ZINC-P' operation. Committees of mighty accelerator experts had been convened to evaluate the prospects for higher current - they concluded that the most that could ever be hoped for was 8 μ A; the advertised current was 25 μ A. Under a thinly-veiled threat from higher Laboratory management, with whom I was by then not very popular (partly

because of some admitted ineptitudes on my part, but certainly also partly because I insisted on doing things well, not just doing them), I was forced to decide on the poisoning question without access to data or calculations to evaluate the impact of choices. I chose wrongly, weighing the intensity question too heavily and having too little faith in my colleagues on the accelerator.

Rob Robinson (LANL) and Andrew Taylor (then at LANL) came to help me in measuring the pulse shapes of the liquid methane moderators. We found that functions of exactly the same form as invented by Jim Jorgensen to fit the profiles of Bragg peaks in time-of-flight powder diffractometers, with negative α and a rationally wavelength-dependent ratio of the α and β terms, fitted the data very well. It was called the Taylor-Robinson-Carpenter function. Dave Pickett (Birmingham University, U.K.) performed calculations using his liquid methane scattering cross sections and found good agreement with the measurements.

In January 1983, we removed the liquid methane moderators and the cooled central part of the beryllium reflector, replacing them with poisoned, ambient temperature polyethylene moderators and a cadmium-decoupled graphite reflector. The beryllium had become too radioactive to rework. Interest in ultra-cold neutron measurements had not vanished. Roy Ringo, Vic Krohn, and Mel Friedman set up the Ultra-Cold Neutron Generator on top of the main shield, on a vertical beam that viewed a fourth moderator of liquid hydrogen, which we installed with the new graphite reflector assembly.

Realizing that we needed a cold moderator for the small angle scattering instrument, we designed a liquid nitrogen-cooled, grooved polyethylene moderator for the "C" position, which we installed in the Fall of 1983. Later, in February 1984, we redesigned the system for cooling by liquid helium. The spectral temperature was about 60 K, even with the lowest coolant temperature. It was qualitatively easy to understand, since polyethylene has few

internal vibrational modes with low enough energy to efficiently absorb the last few millivolts of energy from the neutrons. We measured the decay time of the Maxwellian and found it to be consistent with diffusion theory calculations for a grooved system.

Very gradually, we came to the realization that radiation damage in polyethylene causes changes in the neutronic performance of polyethylene moderators as well as the changes in their physical properties that we had observed in the ZING-P moderators. Casimir DeGusatis, a Pennsylvania State University student who joined us for the summer, characterized these changes - they are all for the worse: spectra harden, pulse widths broaden, and overall intensities decrease. We would have to replace the polyethylene periodically; this was the first observation of this phenomenon. Although it was no surprise, no one knew before what radiation doses were required to produce these effects.

Susumu Ikeda, from KFKI, visited for a year during 1984-85. We measured the pulse shapes as a function of wavelength for all the polyethylene moderators, over a range up to 1 eV, using Kingsley Graham's detector in a time-focussed arrangement with a pressed germanium crystal made by Mel Mueller years before, cooled to 10 K. Ikeda found a marvelously simple, intuitively satisfying function that fitted the data beautifully. It has been called the Ikeda-Carpenter function.

We designed a solid methane "C" moderator, filled with aluminum foam to provide good internal heat transfer, which we installed in January 1985, at the same time renewing the polyethylene moderators. In an IPNS lunch talk just after its first operation, I reported that the spectral temperature seemed to be about 40 K, lower than we had seen with helium-cooled polyethylene. But Gian Felcher sensed my disappointment that the temperature was not much lower. It was a mistake - the aluminum Bragg edges visible in the spectrum were in the wrong places. We had used the wrong flight path length in

interpolating the data; the temperature was actually closer to 20 K, consistent with my expectation. We observed with delight the coldest neutron spectrum that had been reported to that time, 19.6 K.

The system behaved oddly. Every once in a while, and for no reason apparent at the time, the temperature increased. The pressure of hydrogen inside the container went up and, after some time, a tiny leak formed which allowed the hydrogen into the insulating vacuum and caused the system to warm up. We called the events "burps". We understood the accumulation of hydrogen as due to radiolysis of the methane, and its release as the temperature increased. We learned to plug the leak by admitting methane gas into the cold box before filling it. Theories abounded as to the cause of the temperature fluctuations. Blame was laid on the helium refrigerator, on accumulation of condensed oxygen and nitrogen in the moderator or in the insulating space, on a microscopic low-temperature leak. Yoshikazu Ishikawa, at Tohoku University and KEK, theorized that it was some kind of chemical effect. We built and installed a new container in January 1986. At the same time, we installed *poisoned* liquid methane moderators, this time designed to provide narrow pulses.

Convinced that we had solved the problem, I called for Terry Scott to run the "C" moderator at as low a temperature as he could - 7 K. The system behaved beautifully for two weeks, and the spectrum was colder than ever, but the temperature had gradually crept up to 9 K. Two hours after shutdown at the end of the two-week cycle, the moderator blew up! On St. Valentine's Day! All by itself! This event had a way of concentrating my attention on new possibilities for the cause of the temperature excursions. It was not a leak, it was not the condensation of air, it was not the refrigerator. Three weeks later, to the neglect of my family, my friends, my cat, and my own health, I had it! The moderator had exploded! This hardly sounds like anything that we didn't already

know, but what I mean is that the moderator had suffered a classic thermal explosion, caused by the accumulation of energy-storing, reactive species (methyl radicals, it turns out). The gas chromatographic results, calculations of the rate of production by radiolysis, the energy of the recombination reaction, the thermal properties of the materials, and (how that attention was concentrated properly) the periodicity of the earlier burps and the creeping up of the temperature all added up. I invented a crude theory of the explosions, accounting for the continuous buildup of reactants, and Ulrich Walter helped me code up a calculation from the theory that correlated the results reasonably well. And the theory provided us an estimate of the activation temperature for the recombination reaction, about 150 K. The explanation is that, at modestly low temperatures, recombination goes on rapidly and reactive species do not build up, and, at moderately lower temperatures, reactants build up to a certain moderate critical density until their spontaneous recombination overwhelms the cooling mechanism. At very low temperatures, the recombination of the reactive species is so severely inhibited that their density builds up far more - after two weeks, there was sufficient energy to melt the methane and vaporize a fair fraction of it; there was no holding it back once the release had been triggered. The phenomenon had not been observed before, so maybe we can be forgiven for not having anticipated it. Ishikawa was right (!) although I think he had in mind some reaction involving the high surface area of aluminum in our moderator. Later, I wrote up a report for *Nature*. Gerry Lauder congratulated Ulrich for his contribution.

Proposed Facilities and New Instruments

In 1984, Bob Kustorn, Tai Khoo, Ed Croshaw, and Gada Wustefeld developed the concept of a very high current Fixed-Field Alternating-Gradient (FFAG) synchrotron delivering 4 nA of 1.5-GeV protons in short pulses at 40 Hz, which was the basis for a

proposed new facility called the Argonne Super Pulsed Neutron Source (ASPUN). The idea is an adaptation to protons of the FFAG accelerator which was successfully demonstrated with electrons in the 1950's at the Midwest Universities Research Association (MURA), in Wisconsin. Several scaled-down versions of the ASPUN concept have been formulated and put forward, but no substantial work has been funded. We are prepared to launch a design effort on a smaller but meaningful intermediate-scale facility, the Pulsed Neutron Research Facility (PNRF).

About this time, Gian Felcher conceived and Bob Kleb designed the first-of-its-kind neutron reflectometer using a polarized neutron beam. David Price won the contest to name it - POSEIDON. A shortened form of this name, POSY, has been used to refer to this instrument and its unpolarized sibling, POSY II, to this day.

Moderators Revisited

In March 1986, we put back the leaky previously operated container and similar polyethylene moderators, to limp along until we could build proper replacements. We now understood that the stored energy could be released by periodically warming the moderator and undertook this as standard procedure. In April, we installed a new system, which became leaky after a few cycles of use. Torben Brun recalled his experience with leaky cryostats - we took up his suggestion of filling the cracks by admitting gaseous methanol into the system which froze in the cracks at moderately low temperature. Then, cooling to low temperature allowed us to fill the system with methanol. Patching the system with this cryogenic BARZ-LEAKS worked. It was somewhat fussy, but less of a balancing act than was the patching with methanol itself.

Examination of the design of the methanol container revealed that the final closure weld was faulty - the weld was not a fully penetrating weld, purposely to avoid blowing through into the interior during welding. I found that the weld was placed in a location where the

interior surface of the can was under tensile stress so that the unwelded edges constituted a stress riser - the weld had failed under fatigue at the elevated stress level. We redesigned the container with the weld placed so that its internal surface was under compression and with a fully penetrating weld, which was installed in November 1986.

For the next two years, we continued to run the same liquid methane moderators but had to replace the solid methane container again in January 1988. Meanwhile, Scott Cudmak, a clever young engineer who worked with us part time for several years, worked out, among other things, the response of the "C" moderator container to internal pressure, relating it to the release of accumulated radiolytically produced hydrogen, and accounting for thermal expansion of methane, solubility of hydrogen in methane, and the elasticity of the container.

Charlie Potts retired in 1988, having brought the RCS up to its highly productive, reliable levels. Tony Raudhas took the reins of the accelerator.

The Booster Target

Meanwhile, since 1984, we had been working on the design of a new "Booster" target, identical in shape and size to the existing depleted uranium target, but enriched in ^{235}U . I dared not mention this idea in the earlier years, realizing that we had to establish first that the depleted uranium target operated satisfactorily. But it was not a particularly novel idea, since the lower-powered Booster target at the Harwell linac had been operated successfully since 1958, and the JRR-30 pulsed reactor at Dubna had been used successfully in the Booster mode since the mid-1960's. Also, Kimura had conceived a high-power Booster target for use at an electron bremsstrahlung source. However, the IPNS enriched uranium Booster target would be the first to operate at such high power and with such short pulses. We aimed for a modest gain, a factor of three. This allowed us to use the two existing target cooling systems, ganged together,

to remove the additional heat. Al Knox headed the team, Jim Bailey headed the mechanical design effort, Henry Thresh and Al Hins led the metallurgy effort, and Tom Weinack developed the needed copper-boron decoupling material. Roger Blomquist and Roland Armani performed neutronic calculations, and Tom Worlton assisted with HET code calculations of the spallation source. This time, we could not look forward to building the new target at Argonne. Regulations had changed, and we could not handle the required quantity of fissile material. We contracted with the Oak Ridge Y-12 facility for the alloying, casting, machining, welding, and HIP bonding of the target disks.

Bill Steinkamp, Alan Page, and Tom Fisher led the Y-12 target disk development and production effort. Al Hins provided the technical liaison. The Y-12 people had to relearn and adapt the procedures for machining, cleaning, and welding the cladding cans that we had developed earlier at Argonne. Aside from the different venue and personnel, the casting procedure had to be different in view of criticality considerations. And new problems had to be assessed concerning the rate of thermal cycling and radiation damage growth. The Y-12 casting procedure left a texture in the uranium core material that required quantification and remedial measures to reduce. Mel Mueller and Jim Richardson undertook the texture characterization using the IPNS single crystal diffractometer. Bill McDonnell at Savannah River Laboratory and Argonne's Ben Loomis evaluated the growth rates.

The method for reducing the texture was a "beta quench" of the HIP-bonded target disks, which caused severe stresses on the core-cladding bond and led to frequent bond failures. Clarence Stevens and Larry Walker at Argonne identified the offending surface contaminants, and Ron Simandl of Y-12 worked out a new, rigorous cleaning methodology to promote bonding; "Cleanliness is next to Bondedness", we found. Disk production results were very reliable using the refined

methods. Finally, we estimated that the target would live for a very satisfactory three years or so, based on our conservative estimate of uranium growth (or shrinkage).

Having guided IPNS through five difficult years of growth and increasing productivity, Gerry Lander left in 1987. Bruce Brown took over the leadership, and Gus Schulke became operations manager.

The Booster target developments required about four years and about \$2M for completion, but the new target was ready in the Fall of 1988. The Booster had a $k_{eff} = 0.80$ (far subcritical) in its operating position and consisted of 13 disks of 77.5% enriched uranium. Al Knox prepared the three criticality hazards control documents needed. Roland Armani, Joe Casidlo, Fred Kim, Jim Morman, Stuart Carpenter, and others from Argonne's Criticality Hazards Control Committee took responsibility for independent safety and criticality review. Al Knox, Tony Raczhas, Gus Schulke, and Don Bohringer doggedly worked out the required safety and operational documentation. Al spent his honeymoon with Gus and Tony, revising the final draft of the Safety Analysis Report.

We were required to have a SWAT team present during the final stages of assembly and the initial operation. Don Bohringer oversaw their efforts, which were sometimes spooky - I turned a corner once during their exercises to face a camouflage-clad figure prone at the base of an outbuilding, his laser-aimed M-16 pointed in my direction. We heard swooping helicopters and practice gunfire and became accustomed to finding spent (blank) shell casings inside and outside our facilities. One visiting Department of Energy official inadvertently(?) tripped an approach alarm at the top of the biological shield (also known as "the monolith"), bringing armed SWAT people out of the shadows. I suffered some very sweet times working out the criticality monitoring methods to be employed during insertion of the target into its operating position - the target is so far subcritical that standard reactor

loading procedures are difficult to apply and conventional data difficult to interpret.

The Booster target went into place without a hitch on 5 October 1988, and we immediately irradiated it with a few protons. The beam caused the target temperatures to go down; the thermocouples had been wired backward! We celebrated anyway and later reversed the thermocouples. On 11 October, we did our first high power tests, and Frans Trow collected the first data on QENS, which indicated a gain factor of about 2.5 over the intensity of the depleted uranium target. Subsequent measurements indicated that the gain is not the same for all moderators and averages 2.5 over all the beams where before-and-after comparisons could be made. Our calculations had indicated a factor of about 3, but for cost reasons, they had been carried out for only one moderator which, according to later calculations, turned out to be the one that gave the largest factor. We expected the delayed neutron fraction to rise from what it had been with the depleted target. Measurements indicated that it is now 2.8%, roughly equal to the figure that would arise from a calculation for a ^{235}U subcritical assembly with $k_{eff} = 0.80$ and perfect coupling. It is a coincidence that represents compensating effects of imperfect coupling and the higher delayed fraction due to fast fissions in ^{235}U . We had already prepared for this in software measures in some of the instruments and in "delayed neutron choppers", designed by Kent Crawford and Bob Kleb, which were installed in the powder diffractometers. The thermal and hydraulic performance of the Booster target was almost exactly as predicted.

More on Moderators and More New Instruments

Realizing that we already had problems with the solid methane "C" moderator with the depleted uranium target, we decided to fill that moderator with liquid hydrogen, which could not suffer the burping problems. This cost a factor

of 3.5 in the long-wavelength intensity and produced much broader pulses than solid methane, but the system gives no trouble.

Instrument development continued. PHOENIX, a chopper spectrometer especially well suited to the study of quantum fluids and solids, rose from the "ashes" of the solid ^4He diffraction experiment; it was brought into existence by Paul Sokol (Pennsylvania State University), David Price, and Ralph Simmons (University of Illinois - Urbana). An instrument dedicated to diffraction experiments from amorphous systems (GLAD) was designed and built by a large group of scientists, engineers, technicians, and students, headed by Si Moss (University of Houston), David Price, Dan Montague (Williamette University), Sherman Susman, and Kent Crawford.

Problems and Solutions

Jim Jorgensen noted that the Bragg profiles appeared different than they had before the Booster installation: some peaks appeared to have "porches", most had "tails" at the 1% intensity level, but ten times wider than the main peaks. Another set of problems arose. The intensity of the beams from the "I" moderator diminished over the course of a two-week run cycle, sometimes by as much as 50%. And the shape of the spectrum changed. With these shifts, one could expect the profile functions to change in the powder diffractometers. I convened two working groups to address these and related problems: the Cryogenic Moderator Problems Working Group (CMPWG) and the Diffractometer Problems Working Group (DPWG). (Since the latter represents my prying into some matters that some others might consider their "territory", I report our deliberations in a series of memoranda called *The QUIDNUNC*, which is Latin for "busybody".) We had to improve our beam monitor detectors in order to quantify our intensity and spectrum observations - the higher beam intensity caused overloading problems. Kent Crawford introduced feedback preamplifiers and checked out the counting

electronics. We needed reliable proton intensity monitoring methods. Lawrence Densky developed appropriate signals from RCS which were fed to the data acquisition computers. The profile functions do not fit the observed peaks outside a limited range of wavelengths. We introduced a diamond powder calibrant to extend the range of clear observations to short wavelengths. Bill Dinnin, a student from Lehigh University who was working with me, invented a set of functions of the form of Padé approximants which have the correct behavior at limiting large and small wavelengths and interpolate the data more accurately than existing functions; Bill worked with Frank Rotella to install these functions into the IPNS Rietveld analysis software and test them. Jim Richardson, David Mildner, and I worked out methods for correcting diffractometer data for the presence of chopped delayed neutrons. The tails were very elusive, difficult to quantify because they were just about at the background level. Theories abounded as to their origin, which came down to two after some had been eliminated by other tests - thermal diffuse scattering (a sample effect) or inadequate decoupling of the moderator from the reflector.

The CMPWG identified the cause of the intensity loss in the "F" moderator. Hydrogen accumulates in the circulating methane system at the level of a few mole percent; a flash of insight during one of our discussions clarified the problem - hydrogen is a vapor at the temperature of liquid methane (who didn't know this?), occupies a large fraction of the volume, and pushes methane out into the bolus tanks. Scott Chetnak, and later Bill Dinnin, worked up a numerical model of the effect, which confirmed our understanding. Why didn't the "H" moderator behave this way? It turns out that the internal piping in "H" was slightly different than in "F", and "H" acted like a physical phase separator in which liquid collected in the moderator, while voids collected in "H". We devised a hydrogen separator

based on a heated palladium tube, an adaptation of a commercial hydrogen purifier device. Terry Scott and Al Paugys installed it - it worked perfectly.

Now, Terry Scott had been observing ever-more-frequent blockages of the flow in the "F" moderator. We were prepared to understand this since we already expected heavy hydrocarbons to build up somehow due to radiolysis. Those caught up with us in December 1990, when the system blocked up completely. Terry and Al disassembled the piping and flushed it with every imaginable solvent compatible with the materials of the system. The expected oils came out, but a few flakes of amber-like material appeared also, which proved absolutely refractory to our attempts to dissolve them. The best we have been able to say is that the material has a hydrogen-to-carbon ratio of 1.6. We decided to replace the innards of the system. When the moderator-reflector assembly was raised from the monolith, we found that calcium decoupling material had been omitted from the upper surfaces of the beam voids on both sides of the "F" moderator. The mystery of the tails was solved! The "F" container was replaced with another having internal plumbing like "H", to act as a phase separator. The missing calcium was added and additional tightening of the decoupler incorporated. Subsequent tests indicated that the system, even without the palladium-tube hydrogen separator, now operates stably. And the tails are gone! There never were any porches.

Jim Bailey, Al Knox, and Dave Kupperman had devised an ultrasonic method for monitoring the growth or shrinkage of the target disks. (This idea arose during our design discussions, when one of us mentioned that he would like to be able to "thump" on the vessel to hear whether the disks had come into contact with the vessel walls.) Periodic ultrasonic measurements have shown that the disks are growing radially and axially at about one-fifth of the rate allowed for in the design lifetime calculations. Our metallurgical procedures

seem to be successful in controlling the growth, and from this point of view, it appeared that the Booster would live a long life. Then on Flag Day (14 June) 1991, we detected an increase of the counting rate in the detector monitoring ^{136}Xe in the cooling system. Numerous tests have convinced us that a small leak has developed in the cladding of a target disk, allowing a small amount of fission products to enter the coolant stream. The leaks are far below any that would represent a hazard, but exceed the extremely conservative limits we had set on the concentration. Analysis has shown that we can safely operate at much higher concentrations, and we have applied for permission to modify the limit. Meanwhile, we operate at somewhat reduced proton current, pending approval of our request, and have initiated the production of a replacement Booster target.

The Future

Where might we go from here? The advantages of the solid methane " C " moderator are so attractive that this is our top priority moderator improvement. Measurements show that the intensity of long

wavelength neutrons would be increased by a factor of 3.5. It will be necessary to learn how to manage the buildup of energy-storing species (methyl radicals) and to manage (separately) to remove the accumulated hydrogen. If we can learn how to do that in " C ", then it will also be advantageous to install a solid methane " H " moderator. That should increase the long-wavelength intensity by about a factor of 20, useful for the High Resolution Medium Energy Chopper Spectrometer in the low energy, high resolution applications that Chun Long has been exploring, and for QENS. We have identified some possible smaller improvements in the reflector assembly, which could lead to an increase in beam intensity of 20-40%, but they would be expensive. We intend to perform further calculations to evaluate this prospect. Another dream is to improve upon solid methane as a moderator - it seems that there might be some advantage to using a mixture of protonated and deuterated methane, rather than pure CH_4 , to capitalize on the lower rotational level spacings in the deuterated species. We'll need some calculations, which are

impossible for the time being because the scattering kernels are not available, and some measurements, which would be relatively easy if a stock of CH_3D were available. We look forward to the time when we can begin work on a new facility, PNRF.

We have traveled a long journey from the stack of forlorn beryllium, a chunk of plastic, a borrowed isotope source, and microscopic fission tracks; from a block of lead and a second-hand accelerator to a subcritical, enriched uranium Booster target; from blocks of warm polyethylene to intricate arrangements of liquid and solid methane; from two vertical beams with instruments that could be lifted in one hand to a baker's dozen of world-class scattering instruments. We have sometimes been at sea, and not always happy, like Ulysses. But our ship and most of our fellow sailors have weathered the storms and shipwrecks, resisted the Sirens, passed between the Scyllas and Charybdises, and escaped the Polyphemoses. Unlike Ulysses and his crew, we have never really left home. We carry on our travels. What new adventures will we enjoy?

CHAPTER 2

THE IPNS ACCELERATOR SYSTEM

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Introduction

As we celebrate the 10th anniversary of the IPNS facility, it seems appropriate to dig back a little further in time and review the development of the somewhat older IPNS foundation stones - the proton accelerators. But first, let's review what the IPNS accelerator system consists of today.

The Accelerator System

The basic IPNS accelerator system was designed and built by personnel associated with the Zero Gradient Synchrotron (ZGS) high energy physics accelerator. This machine produced a pulse of 12.5-GeV protons every 3.8 seconds. Operation of this facility for high energy physics usage ceased in late 1979 and the synchrotron portion of the facility has been dismantled. The remaining lower energy components of the ZGS system - a 750-keV Cockcroft-Wallton preaccelerator, a 50-MeV Alvarez linac and a 450-MeV rapid cycling booster synchrotron - have been modified to deliver protons 30 times per second to the IPNS spallation neutron source.

Other Anniversaries

The IPNS 50-MeV linac was built to feed 50-MeV protons to the 12.5-GeV ZGS high energy physics accelerator. The first beam was accelerated through the linac in 1962; we can celebrate 30 years of linac operation next year. The IPNS 450-MeV synchrotron, now called the Rapid Cycling Synchrotron (RCS), was originally built as an injection energy booster to increase the beam intensity in the ZGS. An accelerator log book entry on 4 April 1977, indicates first beam injected and successfully coasted, so the RCS will be 15 years old next year.

Booster I

Actually, the collaboration between neutrons and accelerated protons at Argonne National

Laboratory (ANL) began much earlier than 1977. In 1968, Ron Martin, then-ZGS Division Director, decided to make the first U.S. attempt at H⁺ stripping injection. This technique, pioneered in the U.S.S.R., injects H⁺ ions into closed orbits. These ions are doubly stripped to H²⁺ ions by very thin plastic foils. Since the H²⁺ can recirculate through the foils with very little orbital perturbation, it is possible to inject for a much longer time and, thus, get a much more intense circulating beam.

To make these tests, Martin acquired the surplus Cornell 2-GeV electron accelerator, which was rebuilt as a proton synchrotron at Argonne under the leadership of Jim Simpson. An H⁺ ion source development program was also begun at ANL. The reincarnated Cornell machine was renamed Booster I since the ultimate intent of this program was to increase the ZGS beam intensity by injecting higher (booster) energy particles into the ZGS and, thus, to take advantage of the increased space charge limit resulting. Booster I did successfully demonstrate that H⁺ ion injection produced brighter beams with the orbiting beam being 100 times brighter than the injected beam. It was also the world's first demonstration of non-Liouvilian injection on a practical scale. Injection was possible for up to 180 synchrotron beam revolutions compared to a typical 4 or 5 revolutions with H⁺ injection. Booster I's operating time, however, was limited by schedule conflicts with the ZGS high energy physics program. Since virtually all the world's proton synchrotrons now run on H⁺ injection, others must have thought the Booster I pioneering tests meaningful. Booster I was operated often enough to allow Jack Carpenter to do the first prototype experiments using high energy proton-induced pulsed neutrons. Ron Martin also used the beam to explore medical

applications of protons. This work was done in 1973 and 1974. Looks like an opportunity for a 20 year celebration soon!

Booster II

In 1973, funding was sought to build a Booster II which would be able to deliver 3×10^{14} protons to the ZGS at an energy of 500 MeV and do it at a rate of 30 Hz. Since the ZGS could only use 8 pulses every 4 seconds, it seemed that 90% of the beam would be available for other uses, such as pulsed neutron production.

Unfortunately, Washington was not very enthusiastic about improving the ZGS since it was scheduled for shutdown in the not too distant future. The ZGS high energy user community reaction was mixed with many feeling that the remaining years and available funds should be spent exploiting the ZGS uniqueness - namely, the world's only high energy polarized proton beam and the world's best bubble chamber. A realistic analysis did imply that it would be difficult to design, build, and tune-up a new booster synchrotron in time for productive use before October 1979. The ZGS team hoped that a dramatic increase in beam intensity would delay the shutdown date.

Booster II was started in 1975 as a research and development project with very limited funds. Surplus equipment was used whenever possible. Major components and systems came from the Brookhaven National Laboratory, the Cornell Accelerator Laboratory, the Princeton-Penn Accelerator Laboratory, the University of Chicago, and, of course, from the ZGS. A negative view of this situation might say that Booster II was built on a shoestring, but a more positive view says it's an All-American machine.

Project construction leadership was shared between Jim Simpson, Everett Parker, and Bob Kustom. Many ZGS and High Energy Physics

(HEP) Division staff and technicians were involved. Staff members with extensive participation in the design and engineering were: Ed Crosby, Tai Khoo, Martin Foss, Yanglai Cho, Walter Proeg, Ken Monahan, Don McGhee, Len Bulka, John Monahan, Tom Hardak, Jorgan Madson, Art Crane, Maco Lieberg, Gary Gunderson, Marty Knott, Ron Timm, Andy Gorka, Bob Zisman, Reed Moffatt, Dale Suckloth, and Jerry Volk. Our apologies to others inadvertently left out of this list.

To add to the work load, a west coast contractor defaulted on the ring magnet construction contract requiring ZGS personnel to build these magnets at ANL. The magnet construction job was directed by Frank Brumwell and ably assisted by Chuck Kellogg, Jim Bywater, Jim Biggs, and Ken Thompson.

Booster II (RCS) Tune-up

The first beam was injected into the Booster II (RCS) in April 1977. Beam was being extracted toward the ZGS, but was stopped at the ZINC target. (This name was an acronym for ZGS Intense Neutron Generator, after the ING project proposed by Lewis in Canada in the early 1960's.) The RCS was being operated in a time-sharing mode with a switching magnet at the 50-MeV end of the linac steering either to the ZGS or RCS; beam was directed to the RCS about 2/3 of the time. The linac repetition rate was limited by ZGS management to 10 Hz at this time. The tune-up was slow with many equipment problems encountered. The inadequate funding was also reflected in the limited number of beam diagnostics available. Among others, Yanglai

Cho and Tony Rauchas spent many long hours exploring problems and defining solutions. The ZGS technical staff redoubled their efforts to make the equipment run reliably and to understand the physics of the RCS. Much of the operation was at 300 MeV and low repetition rates to minimize the radiation while new techniques were being explored. Gradually improvements in ring magnet power supply stability, stripping foil life, etc. began to become apparent. In late March 1978, a scheduled program of beam delivery to the ZINC-P⁺ target was begun; ZINC-P⁺ was a prototype pulsed spallation neutron source and forerunner to IPNS. The results were far from impressive as Table I shows. Current analogous data for IPNS (Table II) make a pleasant contrast.

Table I. ZING-P⁺ Operations Summary (1978)

Average beam current (μA)	0.7
Operating availability (%)	87
Scheduled operating time (h)	2681
Available operating time (h)	1706
Total pulses on target ($\times 10^9$)	0.43

Table II. Ten Years of IPNS Accelerator Operation

	First 5 Years Nov. 1981- July 1986	Second 5 Years Aug. 1986- May 1991
Average beam current (μA)	11.06	13.99
Operating availability (%)	90.1	93.8
Scheduled operating time (h)	18,021	15,753
Available operating time (h)	16,358	14,781
Total pulses on target ($\times 10^9$)	1.72	1.55
Total μA hours	178,613	192,822

Operation in this mode continued through October 1979, with gradual but constant improvement of availability and beam current. The ZGS accelerator was permanently shutdown in October 1979, so operation at 30 Hz could begin. ZING-P' continued to operate through August 1980. While ZING-P' neutrons were being used for target yield studies and instrument development, the accelerator people were improving equipment and increasing beam intensity while keeping radiation losses under control. The month of July 1980 showed availability at 87% and beam current at 5.4 μA over 5.7×10^7 pulses. Major accelerator improvements were a rugged, transformer-style septum magnet, developed by Martin Foss and Walter Praeg, and high power transistor amplifiers, designed by Charlie Potts, for dynamic programmable control of the tune correction magnets.

RCS Planned Improvement

Sometime in 1979, some members of the materials science community felt progress toward the promised 15 μA goal (or was it 25 μA ?) was far too slow. We tried to tell them it wasn't as simple as adjusting the control rods on a reactor, but they remained unsympathetic. The later experiences at the ISIS facility of the Rutherford-Appleton Laboratory, U.K., and at the Los Alamos LANSCE facility should have by now convinced all but the most critical that the accelerator team and the accelerator itself were pretty good after all.

In response, Bob Kustom developed a program to increase beam current by raising the repetition rate to 45 Hz along with other improvements to get the projected current to 25 μA . The program was started prior to receiving funding, which was shortly denied, so nothing much came of the program except a few power supply modifications.

It was during this era, too, that the collective spirit of the accelerator division rode a roller coaster as the possibilities of a major large facility, IPNS II, waxed and waned.

Conversion to IPNS Target

Relocating the accelerator extraction point for beam delivery to the present IPNS target location was begun in August 1980. During this extensive shutdown, many changes were made as a result of the accelerator physics studies conducted by Yunglai Cho in 1978 and 1979. The injection and extraction apertures were widened, a higher powered klystron extraction magnet was developed by Jerry Volk, shielding was increased, Hvac cooling was increased, added beam diagnostics were installed, rf system impedance control was improved by Fred Brandeberry, and ring magnet power supply preparation for neutron chopper synchronization was completed.

Others from the ZGS, under Reed Moffatt, were busy installing surplus magnets and power supplies in the new 450-MeV proton transport line to the new IPNS target stations. This line included extensive beam profile monitors and loss monitors. A high energy physics test beam target and magnet shalter were also constructed by this group.

Accelerator restart was scheduled for April 1981. This target date was met, and 10 μA were delivered to the beam dump at an energy of 300 MeV. In May, 4.5 μA of 400-MeV protons was delivered to the new neutron Radiation Effects Facility (REF) target. Evidently, most of the newly installed apparatus worked quite successfully. In August, scheduled running to the Neutron Scattering Facility target was begun. The IPNS Division was formed with many of the old ZGS operating group transferring to the new division. IPNS operation as we now know it had begun under the leadership of Charlie Potts.

IPNS Accelerator Operation: The First Five Years

Within 9 weeks of turn-on for neutron scattering operation, we achieved our short-term goal of 8 μA . Unfortunately, we stayed at that level for almost a year since we were efficiently accelerating all of the beam that our H⁺ ion source could deliver. Vern Stipp started in early 1982 to adapt a PermLab

version of a magnetron-type H⁺ ion source to 30 Hz operation. This involved changing the cathode geometry to improve its coolability. The new H⁺ source was installed in early 1983. This unit, along with better understanding of the injection orbits, high energy synchrotron phase space dilution, "early extraction", and decreasing the pulse sensitivity of the ring magnet power regulator, led to beam currents of almost 12 μA in just 6 weeks. Unfortunately, we were too eager to stuff more and more beam in the synchrotron and experienced damage of the synchrotron vacuum chamber due to excessive lost beam. The vertical aperture was decreased substantially as a result of this damage, but we continued to run a few weeks at reduced current to complete the neutron science schedule. A protective collimator was installed, the chamber damage repaired, and operation resumed better than before with currents regularly from 12 to 13 μA . Two more experiences with beam damage to the vacuum chamber led to the installation of a second protective collimator, establishment of operating practices to monitor and control beam losses, as well as routine tests to determine that the protective collimators were absorbing the beam with extreme vertical betatron motion. Recovery from these vacuum chamber damage incidents was prompt, neutron science interruption minimal, and repair personnel radiation exposure was kept to a minimum.

Experience with running the beam to the REF target, which allowed power line synchronization rather than neutron chopper synchronization, convinced us that improvement in the neutron chopper synchronization control would help us control beam losses while increasing beam current to the scattering target. One of our very bright synchrotron chief operators, Lawrence Donley, took on this task. The result was a patented chopper phase lock system (called the Donley clock) that allowed the once again improved ring magnet power supply to reduce injection orbit deviation from ± 8 G

to ± 4 G out of 2800 G. The result was a jump in average beam current to around 15 μA in early 1986. During this period, former ZGS staff members, not in the IPNS Division, provided assistance when unusual problems developed. Also in 1986, the involvement of the IPNS accelerator group with the ANL Strategic Defense Initiative (SDI) program began.

From the first 5 years of IPNS operation, a plot of beam current versus time revealed a positive slope of about 1.5 μA per year. This may be noted in Fig. 1. One of the big events of this 5 year period occurred on 10 June 1984, when the one billionth pulse of protons struck the target. We had a great party. Now that the IPNS accelerator system has produced over 3 billion pulses, this first milestone seems less important, but given the shoestring budget and construction history of the RCS, it was a very significant milestone.

IPNS Accelerator Operation: The Second Five Years

The IPNS accelerator group's participation in the Argonne SDI program began in 1986, peaked in 1987 and 1988, and continues to the present at reduced level. The SDI scientists wanted to study 50-MeV neutral beams. The IPNS 50-MeV H^- beam was easily neutralized, and the old ZGS building was available as an experiment hall. IPNS personnel, under Bramwell's direction, helped design, build, and operate two beam lines to study methods of neutralization, radiation damage to neutralizing materials, neutral beam properties, and methods of producing large diameter low divergence beams. During IPNS downtime, the IPNS linac beam was often delivered to the SDI beam lines. A total of over 5000 hours of SDI running time was accumulated. At the present time, this operation has ceased, and future plans for these beam lines are uncertain.

SDI payment for IPNS personnel and linac usage was substantial, peaking in fiscal 1987 at over one million dollars. Since the accelerator personnel were too busy to use much of this money on the accelera-

tor, it substantially supplemented funding for other aspects of the IPNS program, as well as preventing layoffs that flat budgets for materials science research would have made likely.

Currently, IPNS personnel have a modest involvement in the construction of a very high current, 7.5-MeV negative ion deuteron linac for SDI. This machine is scheduled for completion in 1992.

During their participation in the SDI program, the IPNS accelerator group quite effectively operated and maintained the accelerators for IPNS neutron operation. Beam current continued to increase, which can be seen in Fig. 1 and Table II. No major improvement programs were undertaken, but the new chopper synchronization electronics definitely decreased problems with power line phase stability.

Extensive studies of beam position and size at injection as a function of different tune correction quadrupole settings were undertaken, but no improvement resulted. New power supplies for the bending magnets in the beam line transporting H^- ions from the linac to the synchrotron added to

the injection stability. This allowed precise repeatable tuning of the injection parameters. Occasionally, pulses as high as 16 μA were observed, and monthly averages exceeding 15.5 μA have been reached. A review of Table II indicates that the second five-year beam current average is up about 4.0 μA over the first five years, so the rate of progress has slowed down when compared to the first five years.

Making the accelerator available for neutron users has always had high priority with the IPNS accelerator crew. Comparing the data in Table II indicates success as availability increased about 4% to 93.8%. Upgrades in computer hardware and extraction kicker power supplies, plus a new transformer-style septum magnet have been among the major contributors to the availability improvement. But perhaps the most important factor has been intelligent restraint in pursuing higher beam currents. Operating the machine for the last few years with a fixed loss limit of about 1.5 μA has effectively controlled the residual radiation build-up, allowing any required

Fig. 1. Average proton current delivered to IPNS targets during the first ten years of operation.

repairs to be completed promptly. Maps of residual radiation levels in the synchrotron tunnel are taken after each run. No residual radiation build-up has been noted over 5 years despite the increases in beam current to the neutron target. This philosophy is the key to repairability and will be used with continued emphasis. Figure 2 shows just how good availability has been over the first 10 years of accelerator operation.

IPNS Accelerator Operation: April 1980 - May 1991

A major milestone, the three-billionth pulse of protons to the neutron target, was accomplished on 19 July 1990, and a 327 million start on 4 billion has occurred already this year. Beam current reached the highest yearly average ever at 14.6 μA , and availability, while not the best ever, was close at 94.7%.

The Next Ten Years

Given the poor funding granted to the construction of the RCS, the extensive inclusion of surplus equipment, and the rather poor beam turn-on in 1979-80, few would have predicted the success achieved by the accelerator and its personnel. Continued hard work, a little luck, and adequate funding will assure that the *next ten years* are even better.

Fig. 2. Average availability of the IPNS accelerator system since start up in 1981.

CHAPTER 3

IPNS COMPUTER SYSTEMS: 1981-1991

R. K. Crawford and T. G. Worlton, IPNS Division, Argonne National Laboratory

Introduction

The IPNS data acquisition systems (DASs) have been an important contributor to the success of IPNS, with their unique features enabling some of the unique designs and methods of the neutron scattering instruments. Many features of the DAS were a result of the direct involvement of neutron scientists in all phases of the DAS design and close collaboration between scientists designing the DAS and those designing the neutron scattering instruments. The most significant new feature was the concept of complete software control of the binning process which allowed events to be "electronically time-focused" and histogrammed in several different ways simultaneously. This allowed the detectors for some of the neutron scattering instruments to be arranged in a circle to simplify shielding and geometry instead of arranging the detectors in a "time-focused geometry". Time focusing is necessary because neutrons of different wavelengths are combined in the data, and when data from detectors at different scattering angles are combined, the product of the neutron path length and the sine of the scattering angle must be kept constant.

Another important feature of the DASs is the ability to flexibly schedule the data acquisition process and to interact with the neutron source and monitor and control the sample environment. Users are able to schedule changing of samples or changes of sample temperature and to determine the length of data collection based on the actual number of neutrons detected.

Since IPNS began operation in 1981, there have been evolutionary changes in the computer systems driven by increases in the number and kinds of instruments and increases in the data rate, as well as advances in the computer industry. The data analysis system began as a single Digital Equipment Corp.

(DEC) VAX computer running the VMS operating system. This has been replaced by a VAXcluster of three MicroVAX computers. The DASs initially consisted of DEC PDP-11 computers coupled to a dedicated data collection microprocessor in a Multibus crate. The basic design of the DAS has not changed, but the PDP-11 computers are being replaced by MicroVAX computers, and the microprocessors are being replaced by faster, better designed microprocessors.

The IPNS Computer Systems

The IPNS data acquisition and control computer systems had their origin in the earlier experience at the CP-5 reactor and the ZING-P² prototype pulsed source. Experience at these facilities left IPNS scientists with some fairly clear ideas about what their DAS should and should not do for them. Based on these experiences, and on a review of the data acquisition and computer systems at the other U.S. neutron scattering facilities, a document setting forth the requirements for the IPNS data acquisition and computer systems was prepared. This document, completed in late 1978, served as the basis for the subsequent design of these systems.

Working from this document, groups from the Applied Mathematics and Electronics Divisions at Argonne prepared alternative proposals for system designs which would meet these requirements. In 1979, the proposal of the Electronics Division group was selected, and a system design/development team was formed. Members of this team were Bob Daly, Joe Haumann, and Clare Morgan from the Electronics Division, Dick Hittormann from the Argonne Materials Science Division, and Kent Crawford, George Ostrowski, and Tom Worlton from IPNS. This team carried out essentially all of the design, development, and implementation of the hardware and software for the IPNS DASs.

The first task of the design team was to formulate the detailed specifications for the computer systems to be used at IPNS. These specifications were completed and put out for bid late in 1979. Detailed specifications for the DAS hardware and software to be developed in-house were then hammered out in a number of meetings of the full design team. Early in 1980, the first five PDP-11/34s and the VAX-11/780 were ordered, and by late Spring 1980, the first PDP had arrived, and we were able to begin the actual writing and testing some of the DAS software.

Development of the data acquisition electronics, microprocessor software, and PDP-to-microprocessor communications hardware and software proceeded concurrently. Most of the computers and electronics were housed in a large, specially refurbished room in the old Electronics Division "barracks" building in the 800 area, and most of the development activity took place there. However, we did have the luxury of one 300 baud acoustic modem connected to a DEC LA-120 printing terminal, so some of the PDP software development could be done from other Argonne locations (e.g., Building 223).

An early "victory" of this data acquisition team was the assignment of a major portion of the space in the then-being-designed Building 530 for use by the DASs and the central VAX-11/780 computer system. This occurred early enough to allow us significant input into some aspects of the building design. Our life in the 800 area prepared us to really appreciate this new building.

The IPNS DAS design called for each detector to have its own signal processing chain (preamplifier-amplifier-discriminator-time digitizer) so that data from individual detectors could be combined on-the-fly in a variety of different patterns. This design also provided for simple algebraic manipulations

to be made on the data before such on-the-fly combination of the results, making possible electronic time-focusing which has subsequently been used so effectively, especially on the powder diffractometers. To provide such flexibility, a dedicated microprocessor housed in a Multibus crate was used to carry out the focusing calculations and to bin the data into histograms in dedicated Multibus memory. The binning was controlled by extensive tables, which were set up by the PDP under user control, and then downloaded to the histogramming microprocessor system. A separate, completely independent PDP-microprocessor-electronics system was to be provided for each instrument.

The use of the dedicated microprocessor with a flexible histogramming algorithm controlled by such binning tables made it possible for all the instruments to use essentially identical hardware and software. This, in fact, was one of the original requirements for the system, since it was assumed from the outset that there would never be adequate staff available to maintain the data acquisition systems and computer hardware and software properly, thus making the use of different, specialized DASs for each instrument out of the question. History has subsequently shown this assumption to have been fully justified.

Having essentially the same hardware and software on each instrument provided an additional advantage for the users (or at least we liked to think so). It meant that the same commands were used on each instrument, making it very easy to move from instrument to instrument for different experiments.

After much effort, the VAX-11/780 and the functional DASs for three instruments were installed in the newly completed Building 399 in 1981. Those three instruments were the Low Resolution Medium Energy Chopper Spectrometer (LRMECS), the Single Crystal Diffractometer (SCD), and the Special Environment Powder Diffractometer (SEPD). A fourth functioning DAS for the General Purpose Powder Diffractometer

(GPPD) was housed in the "green trailer" in the DNS Experiment Hall (Building 376). Not long after start up in 1981, our fifth PDP and associated DAS was installed in Building 399 for use with the Crystal Analyzer Spectrometer (CAS).

A different data acquisition system was developed by Frank Williamson and Gordon Holmblad of Argonne's Biology Division for use by the single crystal and small angle diffractometer prototypes at ZING-1¹. Shortly after IPNS start up, this system was refurbished and moved to the "green trailer" to operate the Small Angle Diffractometer (SAD) in its initial stages.

Despite our best intentions to provide a completely independent DAS for each instrument, construction of new instruments at IPNS quickly outpaced the ability to fund new DASs, and within the first year or two of operation, the LRMECS PDP was also serving for the High Resolution Medium Energy Chopper Spectrometer (HRMECS) and the CAS PDP, for the "Ho Experiment". By 1983, we had added two more PDPs, but one of these was used for the Electron-Volt Spectrometer (eVS), and the other replaced the earlier SAD data acquisition system with a copy of the "standard" IPNS DAS.

From the outset, it was intended to network the PDPs to the control VAX-11/780, but this was given a lower priority than the other system components. Thus, for the early years at IPNS, networking was purely by "sneakernet" - RL02 disk packs containing data files were hand carried from the PDPs and loaded onto a disk drive on the VAX, and the data files were copied to mass storage disks on the VAX. Since data rates were not so high in those years, this did not necessitate too many such trips for most instruments, and anyway, the exercise was probably good for the instrument scientists.

The VAX-11/780 originally had 1 Mbyte of memory, a 9-track tape drive, and a 67-Mbyte disk which was used for both the system software and user and data files. In 1982, a 516-Mbyte disk was added, which proved adequate for the next five years. Initially, three

text terminals and a line printer were connected to the VAX, as was a Versatec plotter for graphics output. A control VS11 graphics display was added to allow color plotting and some plot interaction. By 1984, the VAX-11/780 was heavily loaded, and a VAX-11/750 was added to share part of the load. The VAX-11/780 and VAX-11/750 were connected with a high-speed Ethernet connection, using DECnet software.

In 1985, we were finally able to add DECnet to our PDP systems, providing the luxury of electronic data transfer to the VAX, and allowing the instrument scientists to sit back and relax. By this time, with the additions of the polarized neutron reflectometer POLY and the PHOENIX spectrometer, we were operating 10 instruments with 8 PDPs.

The original IPNS DASs used a Zilog Z8000 microprocessor for histogramming data. However, as data rates increased it became necessary to provide a different microprocessor system to handle the higher rates. This led to the development of the multiple parallel-microprocessor systems using the National N32016 microprocessor boards, which have been in use on all instruments except the Glass, Liquid, and Amorphous Material Diffractometer (GLAD) since 1987.

GLAD was designed to produce data rates an order of magnitude higher than those of any of our other instruments. This necessitated a departure from the use of a microprocessor for histogramming the data and led to the development of the FASTDAS table-driven histogramming board. This system has essentially all the table-driven histogramming algorithms used on the other IPNS instruments, but in this case, they are implemented in hardware rather than in software. FASTDAS can histogram data at rates up to 300,000 events/s⁻¹. Also, GLAD utilizes a large number of linear position-sensitive detectors, and so, FASTDAS must histogram data from over 15,000 individual detector elements at those data rates. This indeed represents a major leap in complexity for data acquisition at IPNS.

IPNS Computer Systems: Then and Now

	Fall 1981	Fall 1991
Data Acquisition Systems		
Instruments:	GPPD, LRMECS, SCD, SEPD	GPPD, LRMECS, SCD, SEPD, HRMECS, SAD, QENS, POSY, PHOENIX, POSY II, GLAD, SAND, HIPD
Total Detectors:	~430 standard detectors 1 area PSD	~800 standard detectors 4 area PSDs ~240 linear PSDs
Instrument Computers:	4 PDP-11/34s	5 PDP-11s, 8 VAXstations
Microprocessors:	Zilog Z8000 - none per instrument	National 332016 - up to four per instrument, FASTDAS board on GLAD
Total Disk Capacity (Mbyte):	80	5900
Data Transfer:	"Sneakernet" manual transfer of RL02 disk cartridges	DECnet/Ethernet
Data Archiving:	9-track tape on central computer	9-track tape or 8-mm cartridge tape jukebox on central VAXcluster, 8-mm cartridge tape on instrument VAXstations
Central Computer System		
Computers: (VUP = VAX Unit of Processing)	VAX-11/780 (1.0 VUP total)	Two MicroVAX 3500s and one MicroVAX 3400 in a VAXcluster + VAX-11/750 (6.0 VUP total)
Total Disk Capacity (Mbyte):	67	8580
Data Archiving:	9-track tape	9-track tape, 8-mm cartridge tape jukebox, VHS cartridge tape, TK70 cartridge tape
Networking:	none	Lab-wide Ethernet, BITNET, HEPNET, TELNET
Print/Plot Hardcopy:	Line printer, Versatec plotter	2 line printers, 7 laser printer/plotters, color ink-jet plotter, color pen plotter

Other significant improvements in the DAS over the years include the interfacing of Displex refrigerator and furnace temperature controllers to the PDPs to provide automated temperature control (1985) and the subsequent automation of sample changers on several instruments.

Recently, the most significant improvement for the DAS has been the gradual replacement of the PDP computers with MicroVAX workstations. The first such workstation was provided for GLAD in 1987, and conversion of the IPNS data acquisition software from the PDP version to a MicroVAX version was begun. The computer and software were installed on the GLAD prototype instrument in 1988, and a similar system was installed on the POSY II neutron reflectometer shortly thereafter. Installation of additional MicroVAXes has proceeded by fits and starts, to the point where 8 of the 13 IPNS instruments are now running on MicroVAX systems. Since the initial PDP-to-MicroVAX software conversion, subsequent changes in the IPNS instrument operating software, although significant, have been evolutionary. However, one recent change, which has had a major effect on the way the users interact with the instrument computer system, has been the conversion of some of the MicroVAX systems to the use of DECwindows software. This has added a new dimension of versatility to the systems, and we are only beginning to explore the ramifications.

For most of the ten years of IPNS operation, data files from the instruments had been archived on

9-track reel tapes using the central VAX computer. As MicroVAX computers replaced the PDPs, large-capacity 8-mm cartridge tape drives were installed on the individual MicroVAXes for data file archiving. To further augment archive and backup capabilities, a robotic tape-changer for 8-mm cartridge tapes with a capacity of 250 Gbytes was installed on the central VAX computer. With the robotic tape-changer, on-line access to virtually every data file ever collected on the IPNS instruments is possible.

The most significant development for the data analysis systems has been in the area of graphics. We have gradually converted from text to graphics terminals and have added sophisticated graphics software so that both interactive and hardcopy graphics are now available to all users. Users can quickly determine the progress of an experiment through graphic displays and the success of their data analysis procedures through interactive graphics. A general-purpose plotting program called EASYPLOT was written by Dick Hiltmann for interactively reading and plotting data. This was based on the DISPLA graphics library from Computer Associates, Inc., as were most of the graphics programs written at IPNS. Unfortunately, DISPLA was only available on one computer at IPNS and was not widely available among our users, so another graphics library called GPLOT¹ was written for general use at IPNS and for distribution to our users. GPLOT provides many of the functions of DISPLA, but it is based on a low-cost GKS package which is widely available.

As disk capacity was added to the VAX computers, backing up the disks became a major problem. We acquired large-capacity helical-scan (VHS) cartridge tape drives to simplify the backup procedure.

The VAX-11/780 and the VAX-11/750 were becoming overloaded in 1988 due to the large increase in data to be processed as a result of the installation of the Booster target and the addition of new neutron scattering instruments. It was decided to replace the VAX-11/780 with a cluster of two MicroVAX 3500 computers and one VAXServer 3400 computer. This provided an increase of about a factor of five in computing capacity. The VAX-11/750 was kept mainly for word-processing applications.

Acknowledgements

We wish to take this opportunity to express our gratitude to the members of the original data acquisition design/development team for getting our data acquisition and computer systems off to a good start. In addition, throughout most of the 10 years of IPNS operation, we have had the extreme good fortune to have Joe Haumann working on the continuing development of electronics for the IPNS DASs. He has been responsible for the successful development of the multiple parallel-microprocessor upgrade of the system, for the development of the position-encoding and the FASTDAS histogramming electronics for GLAD, and for many of the other developments which have helped so much to make the IPNS instruments as effective as possible. His help has made possible much of what we are now able to do.

CHAPTER 4

NEUTRON RADIATION EFFECTS STUDIES AT IPNS

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Introduction

The study of radiation effects produced by neutrons has a long and prominent history at Argonne National Laboratory (ANL), reflecting the Laboratory's central role in nuclear reactor development. The major effort began in 1980 with Tom Blowitt of the Materials Science Division and construction of the CP-5 reactor irradiation facilities. These facilities allowed irradiations using either pure, thermal or fast neutrons. Since irradiation-induced defects are mobile in solids at very low temperatures, in some cases below 4 K, it was necessary that irradiations for fundamental studies be made at temperatures as low as possible. Thus, both neutron irradiation facilities were designed to allow irradiation at liquid helium temperature. When a spallation neutron source was proposed at Argonne, Tom Blowitt had the foresight to be among the early backers and planners for two separate facilities, one dedicated to neutron scattering and the other, to radiation effects. Individuals from his research group were involved from the very beginning in the determination of the basic techniques required for utilization of neutrons produced by spallation.

Designing a neutron irradiation facility based on a spallation source posed quite different problems from that based on a reactor. Preliminary questions centered on neutron production rate, neutron energy distribution, spatial variation of the neutron flux, and techniques for maximizing the neutron flux with reflector materials. An extensive effort was made to measure neutron fluxes in a full-scale mock-up experiment. Neutronic calculations were made by Bob Birtcher, and experimental execution and load brick manipulation was orchestrated by Mark Kirk. Determination of the neutron energy distribution by multiple foil activation techniques was performed by Larry

Greenwood of Argonne's Analytical Chemistry Division. Early work included the first experiments utilizing spallation neutrons which involved measurement of disordering rates in Cu₃Al performed by Bob Birtcher and Terry Scott at the beam stop located at the Los Alamos LAMPF facility. These irradiation experiments helped to verify neutronic and damage expectations for the IPNS radiation facility.

The Radiation Effects Facility

Based on these measurements and additional neutronic calculations, the Radiation Effects Facility (REF) was constructed at IPNS. The REF was designed to be as flexible as possible. The facility used a spallation target area that was dedicated to radiation effects measurements and not parasitic to neutron scattering needs. This allowed control over the irradiation time or dose and dose rate. It was possible to vary the neutron flux by varying the proton intensity in each pulse, the proton pulse rate, and the number of pulses. Unlike a reactor, the neutron flux could be turned off or on within the time of a single proton pulse. This allowed irradiations at very low and controlled doses not achievable at a reactor. The volume surrounding the spallation target was filled with removable neutron reflector sections to allow modification of the neutron and gamma-ray spectra. The original reflector sections were filled with lead to maximize fast neutron intensity while minimizing cost. The tradition of low-temperature neutron irradiations was continued. Two liquid helium cryostats were located on either side of the spallation target at positions of highest neutron flux. Cooling for both cryostats was provided by a commercial 400-W refrigerator, modified by Tom Blowitt and Terry Scott. The cryostats could operate between 4 and 1000 K. The first IPNS operation commenced with irradiations

in the REF. Measurement of the neutron flux and energy spectrum confirmed the design expectations.

After a brief period of calibration, IPNS opened as a national user facility on 28 May 1981 with the first irradiations in the REF. Proposals for beam time at the REF were evaluated by the IPNS Program Advisory Committee. Experiments involved the study of basic defect properties in semiconductors, superconductors, metals, ordered compounds, and composite materials. Applied studies were made of insulator and magnet materials for the Department of Energy (DOE) fusion program. In three years of operation, a total of 15 weeks of neutrons from the REF target, over 150 irradiations were performed at the facility. The precise dose control and ability to completely turn off the neutron flux allowed in situ study of neutron irradiation effects in semiconductors. These studies also utilized the liquid helium irradiation cryostats for in situ measurement of the defect charge states through transient capacitance techniques. Investigation of defect interactions and kinetics in ordered metal alloys was made possible by the irradiation cryostats, which allowed control over a wide temperature range, and the ability to vary the neutron flux through control of the proton current and pulse rate. The large cold volume in each irradiation cryostat provided the opportunity for neutron irradiation of superconducting magnets that were carrying current. Additional low-temperature neutron irradiation studies of superconducting materials included ex situ measurement of changes in critical currents as a function of applied magnetic field and transition temperature. A wide range of polymer matrix composites was irradiated at 5 K and room temperature for ex situ measurement of changes in their mechanical properties such as Young's modulus and ultimate strength.

In 1984, the REF was closed for financial reasons. The liquid helium refrigerator system was transferred by the DOE to the low-temperature neutron irradiation facility at the Bulk Shielding Reactor (BSR) located at Oak Ridge National Laboratory (ORNL). After less than one year of partial operation, ORNL closed the facility.

Beyond the Radiation Effects Facility

Since 1984, irradiation damage experiments have continued at IPNS in two ambient temperature facilities positioned in the Neutron

Scattering Facility (NSF) target area. Irradiations using these facilities have studied the mechanical strength of irradiated composite materials and the burn-up behavior of uranium silicide reactor fuels. Determination of changes in lattice parameters and amorphization of neutron irradiated uranium silicides utilized neutron scattering measurements with the General Purpose Powder Diffractometer (GPPD) at IPNS.

The future of irradiation studies at IPNS improved in January 1991 when the DOE decided to reinvigorate and expand

the neutron irradiation program at ANL. A new irradiation thimble, the Radiation Effects Module (REM), has been designed for use in the Vertical Thimble 3 (VT3) located in the NSF target area. Installation of the REM is expected in 1992. The REM will be operated by the Materials Science Division at ANL; it will provide a large irradiation volume and allow neutron irradiation at temperatures from ambient to 800°C. Experiments are planned on reactor fuels, composite materials, insulators, and basic defect cascade effects.

CHAPTER 5

POWDER DIFFRACTION AT ARGONNE'S PULSED NEUTRON SOURCES

J. D. Jorgensen, Materials Science Division, and J. W. Richardson, Jr. and F. J. Rotella, IPNS Division, Argonne National Laboratory

Introduction

Many of the instrumentation concepts and data analysis techniques for powder diffraction at pulsed neutron sources were either developed and first implemented or enjoyed their first widespread use on Argonne's Intense Pulsed Neutron Source or the various ZINC prototype pulsed sources that preceded IPNS. This pioneering work began in the mid-1970's^{1,2} and has continued for over 15 years. As a result, the IPNS powder diffraction program plays a central role in materials research at Argonne and is recognized as one of the most productive neutron powder diffraction programs in the world. This success has demonstrated that time-of-flight (TOF) powder diffraction methods, when properly implemented on a pulsed neutron source of modest flux, are competitive with fixed wavelength techniques on present-day reactors and, in fact, offer advantages for some experiments.

Early Developments at Argonne's Prototype Pulsed Sources

The first pulsed source powder diffractometer at Argonne was operated on the ZINC-P pulsed neutron source in 1974 and 1975.³ ZINC-P was a prototype source that used beam from an existing 200-MeV proton synchrotron - the Booster I for Argonne's Zero Gradient Synchrotron (ZGS) - to produce neutrons by spallation in a lead target. ZINC-P operated for three one-month periods in 1974 and 1975. Several different instruments were tested on the two available neutron beam lines. Thus, the time allocated to each instrument was limited. The powder diffractometer⁴ achieved a resolution ($\Delta d/d$) of about 0.01 in backscattering ($2\theta = 150^\circ$) with an incident flight path of 4.34 m and a scattered flight path of 0.46 m. Adequate count rates were obtained

by using a relatively large (0.17 m) detector array consisting of eleven 1.59-cm-diameter $\times$ 25-cm-long ^3He detectors in a mechanically time-focused configuration.

Although this instrument operated only briefly, it provided the data and experience that allowed the solution of some of the most important problems concerning the design of pulsed source powder diffractometers and the analysis of data from them. Most notably, the peak shape functions that are used in many of today's pulsed source TOF Rietveld codes, based on the convolution of a Gaussian instrumental peak shape with rising and falling exponentials that model the time dependence of

the source pulse, were developed at this time.^{5,6} The peak shape function was tested on data from iron powder at various d spacings. Figure 1 illustrates the precision of fit that was obtained. This early success illustrated that the asymmetric peak shapes inherent to the pulsed source diffractometers could certainly be modeled well enough to allow Rietveld crystal structure refinement. In fact, those working on pulsed sources at Argonne and elsewhere were pleased to recognize that, with a little careful work, the fit to their asymmetric peak shapes was actually better than the standard Gaussian fits to peaks from most of the constant wavelength diffractometers operating at reactors.

Fig. 1 Data (crosses) for the iron 2 1 1 peak as measured on the TOF powder diffractometer at Argonne's ZINC-P pulsed neutron source and least-squares fit (solid line) with a peak shape function derived by convoluting a Gaussian (describing the instrumental resolution) with rising and decaying exponentials (describing the time dependence of the source pulse). This peak shape function, developed at Argonne in 1975, has been used in most of the subsequent Rietveld codes for pulsed source TOF data. (Figure reproduced from Ref. 4.)

Fig. 3 Schematic of the HRPD at Argonne's ZING-P pulsed neutron source showing the arrangement of time-focused detector arrays at scattering angles of $\pm 90^\circ$ and $\pm 160^\circ$.

Data from the HRPD produced some of the most impressive Rietveld refinement profiles ever published. As an example, Fig. 4 shows a small portion of the refinement profile for Mg_2SiO_4 .¹¹ Although the HRPD operated for less than three years (from December 1977 to August 1980), it was one of the most productive powder diffractometers in the world at that time and firmly established the pulsed source TOF diffraction technique as an attractive alternative to reactor-based methods. Working with a number of collaborators inside and outside of Argonne, important papers were published concerning mineral chemistry,^{11,12} organometallic compounds,¹³ fast ion conductors and electrode materials,^{14,15} and metal-oxide catalysts.¹⁶

The HRPD also serves as a unique reminder of the humble beginnings of Argonne's pulsed source powder diffraction program. The instrument was actually built with a zero budget. Detectors were salvaged from previous instruments that had operated at the CP-5 reactor and ZING-P. The instrument itself was constructed on a 1/2-in-thick aluminum plate supported by a Unistrut frame with the detectors being housed in boxes made from 0.062-in-thick aluminum sheet. The shielding included sheets of

cadmium and polyethylene, 5-gal cans of paraffin, and a large pto-shaped tank of water through which passed a rectangular-cross-section tube lined with cadmium that was used to collimate the incident beam. The instrument was located against the North wall of the current IPNS experiment hall (Building 375) with its helium-filled incident flight path (a piece of aluminum downspout enclosed in a 6-in-diameter aluminum tube with shielding in the concentric space) passing across the yard, and through the corner of a large ventilation fan plenum and a 3-ft-thick concrete shielding wall, to the target area in the Booster building. At that time, Building 375 contained the remains of various shielding, large detectors, flight paths, power supplies, etc., that had been part of the ZGS. It was necessary to construct a crude corrugated sheet metal hut around the HRPD to protect it from scavengers and debris and to allow some degree of control over the temperature and humidity. The TOF data were encoded in a multichannel analyzer and output on punched paper tape.

The Powder Diffractometers at the IPNS: SEPD and GPPD

Two powder diffractometers were included in the original IPNS construction project. The decision to build two instruments was based

on the belief that they could be brought into operation quickly, that a large number of users would propose powder diffraction experiments, and that the instruments would feature prominently in Argonne's own materials science research programs. Those expectations proved to be true. The Special Environment Powder Diffractometer (SEPD) was ready to collect data at the time IPNS was turned on in August 1981 and produced an impressive diffraction pattern from silicon during the first few hours of operation. Both instruments have been oversubscribed by a factor of two to three during the entire ten years of IPNS operation.

The SEPD and the General Purpose Powder Diffractometer (GPPD) at IPNS¹⁷ were designed to achieve resolution comparable to the best instruments in the world at the time of their construction while maintaining high data rates. The two instruments are physically identical but exhibit different resolutions because of their different incident flight path lengths. The common design was employed to reduce the costs of engineering and fabrication and resulted in only minor compromises in instrument performance. Figure 5 shows the physical layout of the SEPD and GPPD. The diffractometers are constructed as self-contained units with the final collimator, sample chamber, detectors, and shielding being incorporated into the mechanical support structure. Each instrument weighs about 22 tons (20,000 kg).

The most important new design feature of the SEPD and GPPD was electronic time-focusing.¹⁸ This focusing method allows the use of arbitrarily large detector arrays at any scattering angle. Initially, each instrument contained 120 detectors distributed in groups at various scattering angles. Two-thirds or more of the detectors are in backscattering ($2\theta = 145\text{--}155^\circ$) and at 90° ($2\theta = 85\text{--}95^\circ$) because these are the scattering angles needed for most experiments. The backscattering detectors offer the highest resolution ($\Delta d/d = 0.0035$ on the SEPD and 0.0025 on the GPPD), while the 90° detectors are critical for many experiments in which a restrictive

Fig. 4 A portion of the Rietveld refinement profile for MgSiO_3 , from data taken on the HRPD at ZING-P. Crosses (+) are the raw data. The solid line is the calculated profile. The difference (observed minus calculated) appears at the bottom. Tick marks below the profile indicate the positions of all allowed Bragg reflections. [Figure reproduced from Ref. 11.]

special sample environment is used. The focusing calculations are done in real time by dedicated microprocessors as the data are collected, allowing a live display of the data.

The SBPD and GPPD both met their design goals and have operated reliably during all of the ten years of IPNS operation. One small change made to the basic instrument design involved detector shielding. After the initial operation, it was determined that a contribution to background was coming from neutrons scattered from various internal surfaces of the instrument and from the thick regions of the wall of the sample chamber not in the direct scattered flight path. In the initial design, the detectors were mounted in large wedge-shaped spaces on the right and left hand sides of the sample chamber. To lower the background levels, tapered shield boxes, made from B₄C cast in epoxy, were installed between each detector array and the surface of the sample chamber so that each detector array could view only the region of the sample. After this was done, instrumental background was negligible.

A second modification to the instruments occurred with the installation of the enriched uranium Booster target at IPNS in 1988. With the anticipated factor of ~3 gain in prompt neutron flux with the Booster target, we expected a concomitant increase of a factor of ~6 in the delayed neutrons produced. Delayed neutrons have no time structure, and thus, they contribute to a diffraction pattern as background, but their effect is most pronounced at large d spacings where the prompt flux is the weakest. In both the SBPD and GPPD, choppers were installed to eliminate the delayed-neutron background at large d spacings. How well do they work? Data from a standard $\alpha\text{-Al}_2\text{O}_3$ sample, collected on the SBPD with the Booster target and with its delayed-neutron chopper operating, showed a signal-to-noise ratio (S/N) for the 113 hexagonal Bragg reflection in the backscattering detectors of 360. This can be compared with an S/N of 246

Fig. 5 Schematic diagram showing the overall layout of the SEPD and GPPD at IPNS. The detectors, 10-atm He proportional counters, 1.27-cm diameter and 38.1-cm active length, mount on a track at a constant radius of 1.5 m from the sample position. [Figure reproduced from Ref. 19.]

for the same sample and instrument configuration with the depleted uranium target and without the chopper. Very well, indeed!

Ancillary Equipment on the SEPD and GPPD

Although the instruments themselves have experienced few changes during the ten years of IPNS operation, considerable effort has been devoted to developing equipment for experiments in special sample environments.²⁰ Such developments have allowed the IPNS powder diffractometers to be used for many experiments where the inherent advantages of the TOF technique (full diffraction patterns at a single scattering angle) are evidenced. Emphasis has been placed on equipment that could be automated so that high overall data rates could be maintained. Most low-temperature experiments have been done in Displex closed-cycle helium refrigerators with the temperature being controlled from

the data acquisition system computer. However, cryostats and helium dilution refrigerators have been used for special experiments that required lower temperatures. The minimum temperature achieved on the SEPD is 0.2 K.²¹

A number of furnaces have been developed to meet various experimental needs. The most heavily used of these is known as the "Miller" furnace (named after the engineer from the Argonne West site in Idaho who designed it). The unique feature of the Miller furnace is the ability to study samples at high temperature in controlled atmospheres. John Eubank, the instrument scientist for the GPPD until 1989, was responsible for the original concept for such a furnace and the development of these techniques on the IPNS powder diffractometers.²² The Miller furnace can operate at temperatures as high as 1400°C.

The most widely known experiments done using the Miller

furnace are those involving studies of the oxide superconductors.^{23,24} The range of temperatures and oxygen partial pressures available with this furnace is ideal for studying the high-temperature phase diagrams of these compounds. The first such experiment, and perhaps the best known, was a measurement of the oxygen ordering associated with the orthorhombic-to-tetragonal phase transition in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.²⁵ This compound accommodates a remarkable range of oxygen stoichiometry - exhibiting a variation of one oxygen atom per formula unit (i.e., $0 < x < 1$). As the oxygen stoichiometry is varied the compound changes systematically from a 90-K superconductor (for $x < 0.1$) to an insulator (for $x > 0.4$). The classic in situ neutron powder diffraction experiments performed in the Miller furnace on the SEPD²¹ provided the first detailed information on the structural changes associated with the varying oxygen stoichiometry and have been of central importance in understanding the relationship between the structural and superconducting properties. Subsequent experiments have included in situ studies of the complex high-temperature formation chemistry of the oxide superconductors.²⁶ For experiments of this type, the IPNS powder diffractometers and their associated sample environment equipment are unique in the world as evidenced by the fact that the experiments on $\text{YBa}_2\text{Cu}_3\text{O}_x$ have not yet been fully duplicated at any other neutron scattering facility.

The study of samples at high pressure is another area where the IPNS diffractometers have produced unique results. The use of TOF techniques for neutron diffraction studies at high pressure began at Argonne in the early 1970's at the CP-5 reactor. Some of the same pressure cells have been used at IPNS, and new equipment has been developed.^{27,28} High-pressure experiments take full advantage of the TOF fixed-angle technique. Cells are designed for data collection at $2\theta = 90^\circ$ where appropriate collimation can completely eliminate scattering from the pressure cell walls. In one recent design, the

collimation is actually incorporated inside the pressure cell wall, less than 1 cm from the sample.²⁰ To date, about twenty high-pressure experiments have been performed on the IPNS powder diffractometers. Systems studied include simple metals (e.g., lithium and sodium),^{21,22} ice,²³ various compounds that undergo pressure-induced structural phase transitions,^{24,25} and several oxide superconductors.²⁶⁻³² The structure of phase II of Nd_2Fe , studied by A. C. Lawson et al on the SEPD,³³ is believed to be the most complex structure ever solved and refined entirely from powder diffraction data from a sample in a high-pressure cell.

Although experiments in special environments have contributed greatly to the success of the SEPD and GPPD, it is important to realize that roughly half of the experiments done on these instruments are at ambient conditions. As the data rates have increased over the years, the number of runs has gone up dramatically. Figure 6 shows the number of data sets collected on the SEPD and GPPD each year for the first 10 years of operation. The present rate of nearly 1000 runs per instrument per year equates to an average of six to eight runs per day. This estimate includes the time spent changing samples, changing and equilibrating the temperature, pressure, or sample atmosphere, etc. With these data rates, it is not surprising that the most heavily used pieces of ancillary equipment are the automatic sample changers designed by Bob Kleb and installed on both the SEPD and GPPD in 1987. Each changer accommodates ten samples and can be programmed to change samples based on elapsed time, neutron pulses, integrated neutron flux, or integrated counts in a given detector. With the sample changers, sample loading can be done at convenient times once or twice each day rather than every 1-3 hours.

Powder Diffraction Software Development at IPNS

Using Von Dreele's Rietveld code³⁴ as a centerpiece, Frank Rotella designed a "package" of computer software that would not only perform Rietveld refinements

from TOF neutron powder data, but also would allow the results of Rietveld analyses to be interpreted numerically and graphically. Programs were adapted for use in this package to determine allowed Bragg reflections from, to utilize Fourier synthesis techniques to locate otherwise unknown atoms in, to calculate distances and angles with standard deviations among atoms in, and to illustrate atomic arrangements within crystal structures. Additionally, codes were written to display raw data versus TOF and d spacing, to determine instrument parameters for standard detector arrangements of the SEPD and GPPD, to update least-squares variables with their refined values for use in subsequent refinements, to produce profile plots (graphical comparisons of observed data and calculated profile versus d spacing), and to simulate TOF neutron powder spectra from known structural models and instrument configurations. The package was assembled between 1979 and 1982, when the first *Users Manual for Rietveld Analysis of Time-of-Flight Neutron Powder Diffraction Data at IPNS* was compiled and distributed to users of the IPNS powder instruments, a tradition which

continues today with the most recent version of the manual.³⁵

Since 1982, software in the IPNS Rietveld analysis package has been augmented, primarily in response to specific user needs. The capability to fit individual or multiple diffraction peaks without regard to crystal structure constraints was developed by Faber and Dick Hitterman in 1983.³⁶ This software is used primarily in residual stress determinations, but it is also useful for extracting integrated intensities from TOF neutron powder profiles. Beginning in 1984, the ability to analyze data from multiple phase samples, that exhibit diffraction peaks that are broadened due to anisotropic sample strain, and that can be modeled by anharmonic thermal vibration for individual atoms in crystal structures was incorporated into the package.³⁷ In response to the inability to model broad oscillations occurring in data, primarily from zeolite samples, due to amorphous or other noncrystalline scattering components in the sample, Jim Richardson and Faber developed a Fourier-filtering/background subtraction technique in 1986 to fit the oscillations and remove their effect from

Fig. 6 Data sets collected per year on the SEPD and GPPD for the years 1981 to 1990.

the crystalline data.¹⁷ The user-friendliness of the package was enhanced when software to control user interaction with programs and computer job submission (RIETVELD) was introduced beginning in 1986; further enhancements occurred in 1989 with software to summarize Rietveld analyses from samples under varying experimental conditions and to extract easily in tabular form or for graphical display the response of any or all structural parameters to the variations. Recently, Hitterman and Rotella have developed software to "automate" the Rietveld analysis procedure; it is expected that this software will provide great utility in the analysis of powder data collected as experimental conditions are varied in real time.¹⁸

Since IPNS commenced operation, software from the IPNS Rietveld analysis package has been exported to some 79 users at 60 institutions internationally. The institutions encompass university, government, and industrial laboratories in Australia, Canada, Denmark, France, Germany, Italy, Japan, Sweden, Switzerland, the U.K., and the U.S.

Summary

Looking back over the last ten years of IPNS operation, it is satisfying to realize that the two powder diffractometers, SEPD and GPPD, have operated continuously from the first days to the present. Over 12,000 data sets have been collected. Most of these have provided results that were eventually published, making the powder diffractometers the most productive instruments at IPNS in terms of the number of publications. It was originally expected that the powder diffractometers would be workhorse instruments, and this expectation has been fulfilled. What was perhaps not fully anticipated was the extent to which neutron powder diffraction would become a dominant force in the materials research programs at Argonne and more generally in the U.S. physics and chemistry communities. The SEPD and GPPD at IPNS have played a major role in that success.

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CHAPTER 6

RESIDUAL AND APPLIED STRESS MEASUREMENTS AT IPNS

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Introduction

The accurate, absolute, and nondestructive measurement of residual and applied stresses in metallic, ceramic, and composite components is one of the major problems facing engineers. As an alternative to the destructive techniques of sectioning and hole drilling, x-ray diffraction has long been recognized as the primary nondestructive technique for the measurement of near-surface stresses. Although ultrasonic, thermoplastic, and magnetic techniques have also been developed for nondestructive analysis, neutron diffraction offers some distinct advantages over all of these techniques. Over the past decade, developments in the application of neutron diffraction to the measurement of bulk stresses in engineering materials have dramatically extended the range of materials accessible to analysis. These developments include important experiments at IPNS which have nicely demonstrated the importance of fixed scattering angle diffraction.

Potential applications of the technique cover a considerable range from measurement of macrostresses in massive engineering materials (e.g., high strength metallic alloys) to detailed analysis of microstresses in more subtle components (e.g., ceramic composites). The General Purpose Powder Diffractometer (GPPD) at IPNS has an excellent combination of instrumental resolution and neutron flux which has been put to good use for routine measurement of residual and applied stresses.

Importance of Residual and Applied Stress Measurements

The presence of residual stress in engineering components significantly affects load carrying capacity and resistance to crack growth. Residual stresses can be introduced unintentionally or deliberately

during fabrication and as a result of differential plastic creep deformation during use. Furthermore, the mechanical properties of advanced ceramic- and metal-matrix composites, such as strength and toughness, can be greatly influenced by the presence of fabrication-induced residual stresses. Stresses are created when two different materials used to form the composite are fired and cooled. Differential contraction can lead to one constituent (e.g., the matrix) compressing the other (e.g., fiber). Very large residual stresses (up to 2 MPa) can result.

It is important in the optimization of mechanical properties to know the stress levels and how to modify them. Control to the problem is the ability to measure changes in lattice spacings when the material is elastically strained. If the strain is in the elastic region and the elastic moduli of the material are known, we can determine the stress inside the material or the average stress of a constituent of a composite.

Time-of-flight Neutron Diffraction as a Stress Measurement Technique

With any diffraction technique, residual or applied stresses are measured in the form of lattice strains, i.e., as proportional changes in crystallographic lattice spacings. Neutron diffraction offers the unique capability of providing bulk measurements, with neutron penetrations of up to many millimeters. If both macrostresses (e.g., from welding or surface finish) and microstresses (those on the scale of the microstructure) are present, the macrostresses are averaged out within the sampling volume, thus making the microstresses directly accessible. This is a significant advantage over x-ray diffraction, which measures only near-surface stresses which may include microstresses. Time-of-flight (TOF) diffraction, in which complete

diffraction spectra are collected at a single diffraction angle, allows simultaneous measurement of data representing many different crystallographic directions, and thus potentially different stress states, while maintaining the same gauge volume (identically the same collection of sample grains are scattering for each point in the spectrum). Instrumental resolution is fundamentally important, and the GPPD offers good resolution over a wide range of scattering angles; the resolution is roughly constant at each angle.

Neutron diffraction was first applied to residual stress measurements at Harwell in 1981.^{1,2} At about the same time, the residual stress measurement capabilities at IPNS were being developed by John Faber in collaboration with Dick Hitterman and Stu MacLewon. In 1983, MacLewon and Faber³ reported the first TOF residual stress measurements from the GPPD. Since that time, many more important developments have followed. For instance, the GPPD has proven to be particularly well suited to the study of composites. The first applications to the study of stresses in composites were in 1988 by Allen et al.,⁴ who found tensile stresses in the matrix of an aluminum-matrix/silicon carbide-fiber composite, and by Krawitz et al.,⁵ who studied the relaxation of residual stresses in a cemented-carbide composite under cyclic loading. More recently,^{6,7} the GPPD has been used to measure the residual strains in a wide variety of other ceramic- and metal-matrix composites. To facilitate the intricate sample manipulations required of many residual stress measurements, Dick Hitterman designed and constructed an apparatus, which can be mounted in the GPPD and which has adjustable incident and scattered beam slits and will center the sample in the beam and translate it under

computer control. The development of the GPPD as a stress monitor was recognized in 1988 with an R&D 100 (formerly IR 100) Award as one of the top 100 technological developments for that year.

Strain measurements are used to calculate stresses for comparison with predictions of analytical models to enhance further the confidence in the measured elastic strain and calculated residual stresses. Although results from elastic analysis of a simplified geometry agree reasonably well with measurements in some situations, relaxation of stresses due to creep and microcracking must be taken into account. The analysis can also be complicated by lack of a clearly defined crystal structure.

The neutron diffraction method is not adaptable as a portable system or on-line nondestructive evaluation (NDE) tool in a manufacturing environment. However, it is valuable for validation or calibration of other techniques, such as ultrasonic and Barkhausen-noise techniques for measuring stress, neither of which is an absolute measurement. It is also valuable for the validation of analytical models that are used to predict residual stress and subsequent mechanical properties.

Calculation of Residual and Applied Stress from Diffraction Data

In applying neutron diffraction, the lattice spacings in various crystallographic directions of stress-free powders and/or fibers are first determined. Shifts in the Bragg peaks of stressed constituents are then determined. For any diffraction peak, the lattice strain is given by

$$\epsilon_{hkl} = (d_{hkl} - d_0)/d_0$$

where d_{hkl} and d_0 are the average lattice spacings in the stressed and stress-free material, respectively, and hkl represents the Miller indices of the diffracting planes. With a pulsed source, changes in lattice spacing are related to shifts in the TOF of Bragg peaks. The relationship of TOF to neutron wavelength is described by the equation

$$\lambda = h/mv$$

where λ is the neutron wavelength, h is Planck's constant, v is the TOF for a neutron to reach a detector after leaving the neutron source, L is the source-to-detector path length, and m is the neutron mass. Bragg's law, $\lambda = 2d \sin \theta$, can then be rewritten as

$$1 = (2d \sin \theta / h) \sin \theta$$

where d is the lattice spacing and θ is the diffraction angle (fixed on the GPPD). The measured strain, therefore, is simply

$$\epsilon_{hkl} = \Delta l / l$$

In general, six independent (nonorthogonal) strain measurements are required to define the stress tensor. However, for special high-symmetry cases, fewer measurements will suffice. For example, for composites with fibers, one measurement with the scattering vector parallel to the fiber axis and two perpendicular to the fiber axis are required. If the fibers are single crystals, randomly oriented in an isotropic matrix, only one measurement is needed to determine the stress tensor. On the GPPD, with detectors at scattering angles $2\theta = \pm 148^\circ, \pm 90^\circ, \pm 60^\circ, 30^\circ$, and 20° , each representing a different orientation of the scattering vector with respect to the beam, some of these measurements can be made simultaneously.

Applications at IPNS

As the capabilities of the GPPD have developed over the past ten years, the scope and objectives of residual stress experiments have been extended to take advantage of the developments. Described below in roughly chronological order are illustrative examples.

Grain interaction stresses

In their early work, Macklem and Faber¹¹ established experimentally that shifts in the positions of diffraction peaks from a uniaxially deformed sample of Zircaloy-2 were the result of grain interaction stresses. This was confirmed by the observation that after a given deformation, prism and basal diffraction peaks, representing crystallographically distinct directions, shifted in opposite directions and that reversing the sign of the deformation reversed the direction of the shifts. This was

one of the first illustrations of how different stress states (e.g., the prism and basal directions in Zircaloy-2) could be probed simultaneously on the GPPD. For each deformation, the positions and breadths of nine peaks were refined from a single diffraction pattern.

Depth profiles: The ability of neutron diffraction to produce depth profiles of residual stress in massive industrial components is very important. Such experiments typically involve collecting diffraction data along stress gradients through the material while maintaining the orientation of the scattering vector. This means that successive measurements must be performed by passing the neutron beam through greater and greater amounts of material. We had our first depth profiling experience in 1988 with experiments by Hans Priessmeyer (KFA, Jülich, Germany) and Yale Harker and Dirk Schumak (B. G. & C. Idaho). Priessmeyer was looking at residual stresses near a friction weld formed between WASPALOY and INCONEL, while Harker and Schumak studied variations in internal stresses in an externally deformed U-bend of stainless steel. The use of cadmium and boron nitride masks close to the samples provided beam collimation down to roughly $5 \times 5 \text{ mm}^2$, and absolute intensity measurements confirmed calculated attenuation factors.

Metal-ceramic composites: Starting in 1987, Krawitz and co-workers^{12,13} initiated a program of investigations into differential thermal stresses and stress distributions in coated WC-Ni composites. Of interest were the effects of nickel composition, tungsten carbide grain size, and fabrication cooling rate. Some of the trends in measured residual stresses went contrary to expectation, forcing them to reconsider theoretical models. The observed broad stress distributions about the mean stress values, exemplified by unusually broad diffraction peaks, were found to diminish with increasing temperature as did the mean stresses. By analyzing the d spacing dependence of peak breadth using a number of diffraction lines, they were able to determine that the

broadening arises exclusively from a stress distribution with no appreciable contribution from particle size effects.

Ceramic-ceramic composites: Recent work by Kupperman et al.^{8,10} on a composite composed of silicon carbide whiskers randomly embedded in an alumina matrix illustrates many of the inherent advantages of

a TOP diffractometer. The whiskers are about 1 μm in diameter and 10-30 μm in length and occupy a fractional volume of 15-30%. In cooling from the fabrication temperature, the alumina squeezes down on the whiskers, inducing residual stresses in the whiskers. As shown in the schematic presented in Fig. 1, the whiskers' axes

are randomly oriented in a plane normal to the passing direction. By orienting the sample 45° to the neutron beam and using two opposing 90° detector banks, we select out the whiskers with a particular orientation and simultaneously measure the strains parallel - using the 0006 diffraction line - and perpendicular - using 1120 - to the long axis of the whiskers. Figure 2 shows measured and predicted residual average hydrostatic strains as a function of temperature for whiskers and matrix. Strains are relieved as the composite is heated toward the fabrication temperature, but the extrapolated point of zero strain is 1400°C, about 300°C below the fabrication temperature. This observation indicates that, during the temperature drop from 1700 to 1400°C, the material is relaxing by creep, and thus, it is not building up residual stresses. The plot in Fig. 2 shows that a satisfactory model to predict the hydrostatic strain as a function of temperature can be developed. Measurement of any anisotropy in the residual stresses associated with the silicon carbide fibers is complicated by the fact that the crystallographic symmetry of the fibers is higher than the morphological symmetry, and scattering along the fiber axis is indistinguishable from scattering normal to it.

In contrast to the poorly defined crystal structure of silicon carbide, silicon nitride has a well-defined hexagonal structure. Therefore, various diffraction peaks can be used to determine strain as a function of angle to the long axis of the whiskers (i.e., 0006 uniquely defines the strain along the whisker axis, 1120 uniquely defines the strain perpendicular to the whisker axis, etc.). Calculated strains as a function of angle relative to the c axis of the whiskers are compared with measured strains in Fig. 3. Clearly there is significant anisotropy. The excellent agreement increases our confidence in the measured data and shows that the data are internally consistent and independent of model.

High-temperature superconductors: A final example of the

Fig. 1 Schematic representation of the pulsed neutron measurement of residual strains in a composite material consisting of randomly embedded whiskers.

Fig. 2 Mutual relaxation of strains in alumina matrix and silicon carbide whiskers as the composite is heated. Straight lines are predictions based on elastic analysis.

application of neutron diffraction analysis to an advanced material involves the new high-temperature ceramic superconductors.¹¹ In the few years since their discovery, these materials have already had a dramatic impact on many industries including magnet technology and the development of electronic devices. Single-phase monolithic material is brittle and difficult to work with. It is expected that the toughness can be improved by adding silver to the ceramic to form a superconducting composite. The silver may also enhance the electrical properties. However, the combination of a metal and a ceramic is likely to result in a buildup of residual stresses during fabrication. The silver will contract more than the ceramic matrix and could pull away from the ceramic material, thus breaking electrical continuity. The ceramic should be bonded well enough to pull the silver into tension when it cools. If the silver is in tension, compensating compressive stresses in the matrix will minimize the propagation of cracks, since they tend to concentrate at the tip of a crack. Figure 4 shows the increasing compressive strain in the superconducting oxide $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, which forms the matrix (and also decreases tensile strain in the silver), as the volume concentration of the silver is increased to 20%. This is a clear indication that good bonding of the oxide to the silver has been achieved. The compressive strain relaxes at higher silver concentration, probably because the oxide is undergoing plastic deformation due to creep. Neutron diffraction may be the only technique that could be used to demonstrate this effect.

Summary

Neutron diffraction techniques provide a unique new tool for NDE and characterization of materials, particularly in the area of internal strains in complex structures. Whereas these techniques are not adaptable to a manufacturing environment, they are very valuable for obtaining absolute measurements for validating and calibrating other methods that are easier to use.

Fig. 3 Comparison of calculated strains (solid line) in silicon nitride whiskers of silicon nitride-alumina composite at various angles to the c-axis at room temperature with strain determined by neutron diffraction.

Fig. 4 Bulk strains in ceramic (YBCO) and silver (Ag) components in a composite form of the high-temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

Experiments on the GPD have been instrumental in illustrating the dramatic advantages of a TOF diffractometer for residual stress measurements. The benefits of fixed 90° scattering angle diffraction are now well recognized and documented. Excellent resolution over a wide range of d spacings offers many opportunities for detailed mapping of stress tensors in even more complex materials. With the growing interest on the part of industry in the use of neutrons, there is every reason to believe the coming ten years will bring many more important developments in this field.

Acknowledgments

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CHAPTER 7

THE SINGLE CRYSTAL DIFFRACTOMETER AT IPNS

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Introduction

The first experimental demonstration of the time-of-flight (TOF) method for determining single crystal structures, using chopped beams at steady state reactor sources, was reported by Buras et al. in 1965.¹ It had been only 13 years earlier, in 1952, when Selmer Peterson and Henri Levy reported the first structure determined by single crystal neutron diffraction using a monochromatic steady-state incident beam.² In the late 1960's, Peterson joined his former student, Jack Williams, at Argonne National Laboratory (ANL) to design and build a single crystal diffractometer at the CP-5 reactor. Then, in the 1970's Peterson led the effort to develop a single crystal TOF Laue instrument for the proposed pulsed spallation neutron source.

The prototype instrument for the ZING-P³ pulsed neutron source was designed by Peterson and Ari Reis, Jr., of the ANL Chemistry Division.⁴ The detector was a 'He-filled, multiwire area position-sensitive detector which was shared with the small angle scattering group and is still in use on the Small Angle Diffractometer (SAD) at IPNS. I believe it was sometime in the spring of 1979 that a NaCl crystal was used to obtain the first TOF Laue diffraction pattern on a pulsed spallation neutron source. In September of that year, Reis left Argonne for Brandeis University, and with Peterson planning to retire in 1980, I acquired the role of instrument scientist for the Single Crystal Diffractometer (SCD). Ray Teller worked with me for about the first 2 years of IPNS operation, followed by Peter Loung for 3 years. A large part of the work during those years involved the development of a program library for data analysis. In June 1989, Rick Goyette became the scientific assistant for both the SCD and the polarized neutron reflectometer, POSY.

In 1981, the SCD was setup on beam line H1 with an initial flight

path of 6.5 m. An entry in my notebook dated 1 April 1981 (not an April Fools joke) reads: "Called Al Knox - we will have to fill 500 downsports with 18,000 pounds of anhydrous borax." With help from the entire Chemical and Electronic Structure Group (Ray Teller, Mark Bonn, Jack Williams, and others), we filled the downsports and constructed a shielded hatch. The SCD was ready for testing by July of that year. In 1986, the SCD was moved to its present location on beam line K6 with an initial flight path of 9.5 m to provide higher resolution and avoid dead time losses in anticipation of the installation of the Booster target.

The most important component of the IPNS SCD is the area position-sensitive 'Li-glass scintillation detector designed and constructed by Raul Branner and Mike Strauss of Argonne's Electronics Division. A scintillation detector based on the principles of the Anger gamma-ray camera seemed most promising in terms of overcoming some of the limitations of the 'He-gas-filled multiwire detector for single crystal diffraction. In particular, without the need to contain a large volume of gas under high pressure, a scintillator detector could have high efficiency, thin windows, and minimum parallax due to the thin scintillation glass. A 22-cm-diameter circular prototype detector was successfully tested on the ZING-P³ SCD.⁴ The detector, in use currently and for the past 10 years at IPNS, is square with an active area of 30 × 30 cm²; it was built and ready for testing on the SCD by August 1981.^{5,6} In 1983, Branner and Strauss received an R&D 100 (formerly IR 100) Award for their work in developing the detector.

A number of outside collaborators have made important contributions to the development of the single crystal TOF Laue technique. Bob Jacobson from Iowa State University assisted us in 1981 by adapting his autoindexing program

BLIND for TOF Laue data obtained with the SCD.⁷ Wolfgang Juch of the Hahn-Meitner Institut, Germany collaborated on a series of studies designed to compare the results from the SCD with those from monochromatic neutron and gamma-ray techniques using the same crystal.^{8,9} The analyses included testing various extinction models, spectral corrections, and integration procedures. In those studies, the accuracy of the TOF Laue technique was shown to be equivalent to conventional monochromatic techniques. Clive Wilkinson (Kings College, London, U.K.) examined procedures for integrating Bragg peaks in the three-dimensional histograms.¹⁰

Ancillary equipment for varying temperature and pressure is required for most experiments. The Displex closed-cycle helium refrigerator (10-300 K) mounts through the ϕ -circle of the diffractometer. In order to overcome the problems of shielding the large area detector from the aluminum scattering of the vacuum container, Bob Kleb of Argonne's Materials Sciences Division designed and fabricated a unique vacuum container with two rotating vacuum seals. Recently, low temperatures approaching 4 K have been achieved with a liquid helium cryostat. The first SCD furnace was designed and constructed by Hiroyuki Horikuchi (University of Tokyo, Japan) in order to study yttria-stabilized zirconia, Zr(YO)_{1.96}, at 1040 K with a voltage across the crystal to generate an O²⁻ ionic current.¹² A furnace of a different design was used to characterize the crystal structure of the nonlinear optical material thallium bismuth phosphate, TlBiOPO₄, at 650°C, well above its ferroelectric-to-paraelectric phase transition ($T_c = 583^\circ\text{C}$).¹¹ The helium pressure cells (0-5 kbar) which mount on the Displex cold stage were designed by Kleb for the study of

the organic superconductor β -(ET)₂I, as described below.

During the past decade a related single crystal TOF Laue instrument was under development at the Los Alamos National Laboratory (LANL) by Phil Vergamini and Allen Larson. The exchange of ideas, software, and hardware designs has been extremely beneficial for both groups.

The following sections describe a few SCD experiments selected for their historical and scientific significance.

The First Experiment: C-H Metal Interactions

The activation of a C-H bond by a metal is a critical step in many catalytic reactions. The nature of the C-H metal intermediate had been investigated by various spectroscopic methods, but the arrangement of the three atoms in the bond and the degree of electron delocalization could only be ascertained from high precision neutron structures. By Fall 1981, only four stable molecules containing 3-center C-H metal bonds had been unambiguously characterized by single crystal neutron diffraction.¹¹⁻¹⁴ For the first scientific experiment using the SCD, Jack Williams proposed the structure determination of $\text{Mn}(\text{CO})_5(\text{C}_4\text{H}_9)$ with crystals obtained from Maurice Brookhart of the University of North Carolina. Data were collected with a sample at 25 K in November and December of 1981, but all attempts to phase the data with the previously determined room temperature x-ray structure met with failure. Since this was the first SCD experiment, Ray Teller and I examined every step of the data analysis procedure in detail for possible errors. We also provided Mark Beno with Bragg data within a limited wavelength range to reduce the effects of extinction, and he was able to obtain a structure solution using the direct methods program MULTAN. It then became clear that we had actually chosen a different monoclinic unit cell, with $a_0 = -a_x$, $b_0 = -b_x$, and $c_0 = 2a_x + c_x$, where the subscripts x and 0 refer to the original neutron and x-ray unit cell

vectors, respectively. Initially, we thought the unit cells were equivalent because of the fortuitous near equivalencies of the magnitudes of $c_0 = 17.132 \text{ \AA}$ and $b_0 = 113.21^\circ$ with $c_x = 17.250 \text{ \AA}$ and $b_x = 112.00^\circ$. Thus, the first single crystal TOF structure determination was solved by direct methods independent of the x-ray structure.

The most important feature of the structure is the C(1)-H(1A)-Mn interaction with H(1A) located almost exactly in an octahedral site about the manganese atom and with a very long C(1)-H(1A) distance of $1.19(1) \text{ \AA}$ (Fig. 1). The average value for the other C-H distances in the structure is $1.09(1) \text{ \AA}$, as expected. This same type of interaction was structurally characterized in a related isoelectronic cationic iron complex with C-H = $1.164(3) \text{ \AA}$.¹⁴ In both of these molecules, an 18-electron configuration is achieved only by delocalization of the two electrons in the C-H bond to form a 3-center, 2-electron C-H metal bond.

Organic Superconductors: The Development of the High Pressure Apparatus

Organic superconductors (or organic metals) have been the subject of a number of investigations on the SCD. The first of these involved the characterizations of the low-temperature commensurate superlattices in the quasi-one-dimensional charge transfer salts (TMTSF)₂X, where X = ClO₄ and BF₄, both of which have T_c's of 1-2 K.¹⁵ The superlattices are due to ordering of the tetrahedral anions which are located on centers of inversion in the high-temperature structural phases. This was the first demonstration using the SCD of the importance of crystallographic

Fig. 1. The structure of $\text{Mn}(\text{CO})_5(\text{C}_4\text{H}_9)$ at 25 K.

order to superconductivity in the organic metals.

The largest number of organic superconductors, and those with the highest T_c values known to date, are based on the donor molecule BEDT-TTF or simply, ET.¹⁶ Whereas the TMTSF salts are all nearly isostructural, ET salts crystallize with many different structural phases, even with the same anion and stoichiometry. Most of the salts are layered materials with layers of conducting donor molecules sandwiched between layers of anions.

One of the most interesting features of the ET molecule is the occurrence of different conformations, as depicted in Fig. 2, and the relation of these conformations to lattice softness, disorder, and T_c. In the case of β -(ET)₂I, we have characterized three distinct structural phases. At ambient temperature and pressure, each ET molecule assumes a random orientation with either an eclipsed or staggered conformation of their ethylene groups. In 1984, the observation of incommensurate superlattice satellite reflections with x-rays and with neutrons on the SCD from

Fig. 2. Schematic view of the two possible ethylene group arrangements of ET where the C-S and C-C bonds of the ethylenedithio units ($-S-CH_2-CH_2-S-$) are represented by wedges to distinguish the C atoms lying above and below the molecular π -framework. The diagram on the right of each molecule depicts the eclipsed and staggered arrangements of the ethylene groups when viewed parallel to the central C=C double bond.

crystals of β -(ET)₂I₄ below ~ 200 K was reported.²⁰ These satellite reflections were shown to be due to (i) displacive modulations of the I₄⁻ anions and the ET molecules as rigid groups and (ii) modulation of the aforementioned conformational disorder.²¹ Upon further cooling, β -(ET)₂I₄ becomes superconducting at $T_c = 1.4$ K. This compares to salts in which the ET molecules are in ordered, eclipsed conformations with $T_c = 2.8$ K for β -(ET)₂I₃Br₂ and $T_c = 4.98$ K for β -(ET)₂AsI₄. The low T_c for β -(ET)₂I₄ was attributed to its partial disorder (incommensurate modulation) although band calculations of the density of states at the Fermi energy indicated it should have the highest T_c .

Then in 1985, Soviet and Japanese scientists reported that the superconducting transition temperature T_c for β -(ET)₂I₄ rises from ~ 1 K at ambient pressure to ~ 8 K with applied pressures above 1.2–1.3 kbar.^{22–24} Simultaneously, I sent off a proposal to the Institut Laue-Langevin (ILL) in France and discussed with Bob Kleb the possibility of obtaining pressure capabilities on the SCD. Kleb came through first with the design and construction of the high pressure

sample cell which mounts on the Displex cold stage of the SCD. To complete the instrumentation, Jim Jorgensen of the Materials Science Division helped us locate a helium pumping station which had been stored in the basement of ANL Building 223 since the shutdown of CP-5 in 1979. Using this equipment, we observed that the satellite reflections, which are present at ambient pressure below 200 K, are absent above a critical pressure of 0.5 kbar applied at room temperature and followed by cooling (Fig. 3).²⁴ The satellite reflections remained absent if the pressure was released while the temperature was maintained at 20 K. It was proposed that the high-pressure structural phase was ordered and the transition from the modulated structure to the completely ordered phase accounted for the rise in T_c . The ordered structure was later confirmed with data obtained at the ILL.²⁵

High- T_c and Related Materials: Studies of Magnetic Properties

Materials related to the high- T_c superconductor La₂CuO₄ were the subject of the first quantitative investigation of magnetic properties using SCD data. La₂CuO₄ is

antiferromagnetic with a Néel temperature that depends on δ and a magnetic form factor in the ordered state which is not in very good agreement with that of the Cu²⁺ free ion due to a plateau at low Q . The two reasons suggested for this discrepancy are either anisotropic magnetization density of the copper site or some small magnetic moment on the out-of-plane oxygen sites. In order to test these theories, measurements on a material in which the out-of-plane oxygen atoms are replaced by chlorine atoms, Sr₂CuO₂Cl₂, were carried out by Xunli Wang from Iowa State University in a collaboration with scientists from Argonne and other laboratories.²⁶ Using the SCD, it was found that the form factor of Sr₂CuO₂Cl₂ is quite similar to that of La₂CuO₄, and it is, therefore, unlikely that any significant moments exist on the out-of-plane oxygen (or Cl) ions. A reasonable fit to the experimental form factor data is obtained if a dipolar spin polarization on the in-plane oxygen ions is assumed. In this model, the in-plane oxygen 2p orbital which overlaps with the two nearest neighbor Cu $d_{x^2-y^2}$ orbitals is spin up on one side and spin down on the other side.

In another series of experiments, the magnetic form factor of La₂NiO₄ was examined.²⁶ Since Ni²⁺ has a much larger moment than Cu²⁺, measurements on La₂NiO₄ should be more precise than those on La₂CuO₄ and Sr₂CuO₂Cl₂. Attempts to measure the magnetic reflections of La₂NiO₄ at a steady state reactor source were not successful due to contamination by multiple Bragg scattering. Using the IPNS SCD, each reflection was measured many times (~ 10) over a wide range of neutron wavelengths so that anomalous intensities could be easily identified and discarded. Again, the experimentally determined data for La₂NiO₄ exhibit a plateau at low Q similar to La₂CuO₄ and Sr₂CuO₂Cl₂. With La₂NiO₄, the larger number of measured magnetic reflections permits the evaluation of the spin density Fourier map corresponding to the difference between the observed density and that obtained by fitting

Fig. 3. (a) Plot of the neutron diffraction intensity distribution in the $h = 4.92$ reciprocal lattice plane of $\beta\text{-(ET)}_x\text{I}$, at 20 K and zero pressure. Satellite peaks at $(5\frac{1}{2}\frac{1}{2}) - q$, $(5\frac{1}{2}0) - q$, and $(5\frac{1}{2}1) - q$, where $q = 0.08a^* + 0.27b^* + 0.21c^*$, are clearly observable. (b) The same $h = 4.92$ plane after applying a pressure of 1.4 kbar, warming to room temperature, and cooling back down to 20 K.

the data to an isotropic Ni^{2+} form factor. No significant density is observed away from the Ni-O plane. As shown in Fig. 4, in the Ni-O plane, there is spin density extending from each copper in the direction of the four in-plane oxygen atoms.

Texture Studies of Polycrystalline Samples

Although the SCD was designed for measurements of

single crystal samples, over the years, we have found that it is also ideally suited for determining the texture (preferred orientation) of polycrystalline materials. A major advantage of using the SCD, with its large area detector and range of wavelengths, is the ability to measure large portions of the pole figures of many d spacings with a single sample orientation in the case of a fine-grained polycrystalline material. For a coarse-grained

material, single crystal orientations for several grains can be derived from one sample setting.

An example of determining preferred orientation of a coarse-grained material is that of mineral grain orientations in earth mantle samples (J. R. Smyth (University of Colorado), P. J. Vorganini (LANL), and A. J. Schultz (ANL)).⁴⁷ In this experiment, slabs approximately $80 \times 25 \times 3$ mm³ were attached to an x-y linear drive stepping motor assembly which was mounted in the sample position of the SCD. The x-y translator also includes one rotation axis and had been constructed in 1981 by Bob Klab, Torben Brun, and Sherman Susman of the ANL Materials Science Division to search for single crystals in boules of LiAl and LiGa obtained from a melt. For the earth mantle experiment, a single histogram of data was recorded for about 35 minutes at one spot, the sample was then translated a few millimeters, and data from another spot was recorded. In this manner, data from as many as 48 spots were collected from each sample. Bragg peaks were indexed for garnet and clinopyroxene using the multiple phase and crystallite features of the GSAS computer program by Allen Larson and Bob Von Dreele of LANL's LANSCE facility. Significant preferred orientation was revealed with respect to the observed foliation of the rocks which may account for seismic velocity anisotropy.

The development of pole figure analysis software was initiated in 1987 in order to study the texture of high-T_c superconductor samples. The pole figure analysis software developed here at Argonne also produces the input required to calculate the orientational distribution function (ODF) using software provided by John Kallend of the Illinois Institute of Technology (IIT). In a round-robin experiment using a highly textured calcite sample, the results from both the IPNS and LANSCE SCDs compared well with the results from instruments at steady state reactor sources.⁴⁸

During the fabrication of wires, rods, or films from high-T_c copper oxide superconductors, the high

aspect ratio of the crystallites leads to preferred orientation. In the case of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, this texturing can give rise to significant increases in the critical current density [J_c] if the a - b planes of the crystallites are aligned in the direction of the electrical conductivity. In an ongoing program with Ken Giannelis (Material and Components Technology Division) and Tony Biondo and Kullend (ITP), various processing techniques, ranging from slip casting to sinter forging, have been evaluated.^{12,13} We are currently extending these investigations to samples of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ superconductor.

Looking Ahead

In the past ten years, the single crystal TOP lane technique has progressed from the proof-of-principle stage to being a valuable scientific tool of proven capabilities. Of course, the entire IPNS staff need to be acknowledged for the design and construction of

everything from the beam line to the data acquisition system. In the near future, plans for installing a chopper to reduce delayed neutron background, obtaining a closed-cycle helium refrigerator capable of cooling to 4 K, and the addition of a second detector, will be evaluated. These improvements could lead to better measurements of low Q data (e.g., first-order magnetic reflections) at lower angles and shorter wavelengths where there is a higher incident neutron flux and less secondary extinction. A second area detector will also reduce data collection times and allow the use of smaller samples and the study of larger crystal structures.

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Fig. 4. The difference magnetization density in the Ni-O plane of LaNiO_3 . Ni atoms are represented by circles and O, by triangles. The solid and dashed contours denote positive and negative densities, respectively. Each contour is $8 \times 10^{-3} \mu_B/\text{\AA}^3$. The Ni atoms at the corner positions have been defined to have positive densities.

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CHAPTER 8

SUPERCONDUCTIVITY

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Introduction

Although present activities on superconductivity at IPNS, as elsewhere, are overwhelmed by research on oxide-based materials (e.g., over half the experiments on the Special Environment Powder Diffractometer (SEPD) during 1988-1990 involved oxide superconductors), the history at IPNS for studies of superconductivity predates the oxides. These activities have included Chevrel-phase materials, transition metals, and organic superconductors. The latter area is covered in Chapter 7 of this report, while the former two are discussed in this section. The bulk of the studies on Cu-O-related compounds are driven by the crucial value of neutrons for studying oxygen-containing materials, since x-rays do not have the necessary scattering strength for oxygen.

Structural Studies

Chevrel Phases: During 1983-1984, there was much activity investigating the large observed variation of superconducting properties of the Chevrel phase superconductors, SnMo_6S_8 and PbMo_6S_8 , have large critical fields and T_c 's of 12.2 and 13.3 K, respectively. In one of the first studies that showed it is possible to locate and identify point defects by powder neutron diffraction,¹ it was revealed that impurity oxygen substitutes at a particular sulfur site in the structure (Fig. 1). This substitution causes a static displacement of the neighboring Sn or Pb cation, and T_c is lowered substantially for even small defect concentrations. These results pointed the way for the synthesis of materials with reproducible properties and led to studies that put the materials on a proper scientific basis.

Later, it was learned that in Chevrel-phase systems having divalent cations (i.e., Sn^{2+} , Pb^{2+} , Ba^{2+} , Eu^{2+} , or Yb^{2+}), it was possible to view these compounds as

members of a generalized temperature-pressure-composition phase diagram containing a rhombohedral, superconducting region and a triclinic, nonsuperconducting region.² T_c was found to increase systematically as the phase boundary is approached, reaching a maximum at the boundary, and then dropping sharply to zero upon entering the triclinic phase. These experiments, carried out primarily at IPNS on the General Purpose Powder Diffractometer (GPPD) and the SEPD, are among the clearest

experimental proof for a direct relationship between T_c 's and structural instability. These studies, as well as the recent ones on oxide superconductors, rely heavily on the extensive use of the Rietveld technique.

Structures of Oxides: The studies of Chevrel phase materials at IPNS were also important because they showed the unique advantages offered by the time-of-flight technique and the direct way that GPPD and SEPD allow for fast, automatic changing of samples. Thus, it was possible to study oxide superconductors quickly under conditions that simulate those during synthesis and to use the results to develop improved synthesis routes. The work on oxide superconductors proceeded at such a feverish pace that the normal IPNS proposal system was overwhelmed by the number of proposed experiments and too slow to allow for competitive research. A "hot proposal" mechanism was created to allow users to obtain rapid access to the IPNS instruments for these important experiments. Beginning in January 1989, several weeks of additional neutron beam time were funded by the National Science Foundation, Science and Technology Center for Superconductivity (grant number DMR 88-09854) allowing scientists from the member institutions of the center (University of Illinois - Urbana, Northwestern University, University of Chicago, and Argonne National Laboratory) to obtain fast access to the IPNS instruments without unduly impacting the ongoing IPNS user program.

The importance of neutron powder work grew as it became apparent that the role of oxygen in the Cu-O superconductors is quite complex and could be revealed by this technique. For the first superconductor having a T_c greater than liquid nitrogen temperature, $\text{YBa}_2\text{Cu}_3\text{O}_x$, (YBCO) with $T_c = 93$ K, a single-crystal x-ray diffraction

Fig. 1 Local structure in the region of an oxygen defect in a Chevrel-phase superconductor.

study was dense but yielded an incomplete structure because of twinning and the inability to locate the oxygen atoms. The first report of the correct structure⁴ was based on work at IPNS, and the second report of the correct stoichiometry of this compound (late by 1-2 days) came from data obtained at IPNS.⁵ As shown in Fig. 2, this unique structure has the by-now notorious Cu-O planes and chains.

In many initial structural refinements for Bi-O- or Tl-O-based superconductors, it was clear that the locations of the oxygen atoms in the respective layers were poorly defined, and it was deduced that the oxygen atoms (and the Bi or Tl to a lesser extent) relax from their ideal positions to achieve more desirable bond lengths. This local distortion was seen in Tl-based-2212 via an analysis of the atomic pair distribution function from IPNS data.⁶ This latter result indicated that the local structure for the Tl-O sublattice does not agree with the average structure based on refinements. Two possible local Tl-O arrangements with either Tl₂O₂ clusters or zig-zag Tl-O chains fit the results. This work was later extended to show that the local structure

changes abruptly at the onset of superconductivity, suggesting that the mechanism involves strong electron-lattice interactions.⁷

Oxygen Occupancies: One of the delightful complexities of YBCO is that the oxygen content can be varied from 6.0 (tetragonal, insulating, and nonsuperconducting) to 7.0 (orthorhombic with $T_c = 93$ K). At $x = 7.0$, there is perfect ordering of the oxygen vacancies, while removal of oxygen leads to random ordering of the oxygen defects at about $x = 6.35$ and the nonsuperconducting tetragonal structure.

Studies at IPNS on the variation of oxygen site occupancies in YBCO as a function of temperature and oxygen partial pressure⁸ are among the most heavily quoted papers in the field, owing to the fact that this *in situ* work (for which the IPNS instrumentation offers unique advantages) had not at that time been duplicated in any other laboratory. The orthorhombic-tetragonal transformation temperature varies from 700°C in 1 atm of oxygen to 450°C in 10^{-4} atm of oxygen (Fig. 3). In the tetragonal phase, the O1 (chain) and O5 (interchain) oxygen sites are equivalent with random partial occupancy.

Work on YBaCuO_{7-x} has also included extensive studies of the relationships between oxygen content, structure, and superconductivity.⁹ It has been shown that oxygen atoms are mobile at room temperature in YBCO.¹⁰ When YBCO is quenched from high temperature to room temperature, the structural parameters and T_c vary with time. The superconducting properties are controlled by the local ordering of oxygen atoms at room temperature to form chain fragments on the available sites, which in turn affects the carrier doping of the Cu-O planes and the T_c 's. Changes in the Cu-O bond lengths, corresponding to an expected electron transfer necessary to change T_c by 20 K, are observed, in agreement with the observed changes in T_c with time.

Ba, K, BiO₂: In the Ba₂K₂BiO₄ system where T_c reaches 30 K for $x = 0.4$, extensive studies of the various phases and high-temperature structural chemistry have been done. At and below room temperature, five structures have been identified, only one of which, a cubic perovskite phase, is superconducting.¹¹ Additional *in situ* diffraction studies at high temperature in controlled atmospheres have elucidated the complex chemistry that controls the solubility of potassium in this compound and provide unique insight into an important method by which new superconducting oxides are synthesized.¹² At the synthesis temperature (near 700°C) in specified reducing atmospheres, a sufficient number of oxygen vacancies are present to reduce the material which allows the incorporation of K⁺ ions. Thus, potassium can be incorporated only if charge-compensation oxygen vacancies are present such that a low bismuth oxidation state is maintained. The desired oxygen stoichiometry compound is formed by further low-temperature oxidation, where K⁺ is immobile. In these studies, the rapid sample throughput was most useful in ascertaining the best methods for preparing phase-pure materials. In some ways, the synthesis of YBCO also requires reducing conditions, followed by oxidation to $x = 7.0$ as the material

Fig. 2 Structure of (a) the superconducting orthorhombic phase and (b) the nonsuperconducting tetragonal phase of YBaCuO_{7-x}.

Fig. 3 Occupancies of the indicated oxygen sites in $\text{YBa}_2\text{Cu}_3\text{O}_7$, versus oxygen partial pressure in situ at 490°C . Results are shown for two different samples.

is cooled to room temperature in an oxygen-containing environment.

Role of Defects: The critical importance of the relationship between the electronic properties of the oxide superconductors and the concentrations and ordering of various defects is becoming clear.¹¹ Many of the current ideas are based on neutron powder diffraction results. Data from IPNS has provided some of the first information on these defects in various systems. For example, in $\text{La}_2\text{CuO}_{4-x}$, superconductivity results from the incorporation of interstitial oxygen (x) in the corrugated La_2O_2 double layer.¹² The presence of this interstitial oxygen and its high mobility lead to an unusual phase separation into superconducting and nonsuperconducting phases that surprisingly occurs near room temperature. Two nearly identical antiferromagnetic phases are observed

(with $x = 0.0$ and 0.08) between 10 – 320 K, with only the latter composition showing superconductivity. The structure of the interstitial oxygen defect was learned from a parallel study of the isostructural compound $\text{La}_2\text{NiO}_{4-x}$, where higher defect concentrations can be reached.¹³

Interstitial oxygen defects also occur in other oxide superconductors. An unusual phase separation was also seen in a study at IPNS of the "electron-doped" high- T_c material $\text{Nd}_{1-x}\text{Co}_x\text{CuO}_4$ (so-called T^* structure).¹⁴ Although the defect structure has not been determined, it is expected that defects also play a key role. The structure of the interstitial defect that is thought to be one of the doping mechanisms in the T^* phases (essentially a combination of the T^* and $\text{La}_2\text{CuO}_{4-x}$ phases) has recently been determined from IPNS powder diffraction

data, by doping some of the oxygen with fluorine.¹⁵

Defects can also destroy superconductivity. In the $(\text{La},\text{Sr},\text{Ca})\text{Cu}_2\text{O}_{7-x}$ system, where Cava has recently found 40 K superconductivity,¹⁷ IPNS powder diffraction data have shown that the nonsuperconducting phases contain excess oxygen in the interlayer site (equivalent to oxygen in the Y-layer site in $\text{YBa}_2\text{Cu}_3\text{O}_7$). It is hypothesized that excess oxygen in this site destroys superconductivity, but the incorporation of calcium into the structure produces a superconducting compound by blocking the formation of these oxygen defects.¹⁸

Phonons

$\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$: A systematic study of the phonon density of states (DOS) of $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ has been very informative. The study combined the inelastic neutron scattering results with DOS calculations via molecular dynamics (MD) simulations.¹⁹ The measured DOS yields broad bands centered at 15 , 30 , and 61 meV, with substantial differences from undoped BaBiO_3 , and the nonsuperconducting $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$. For the superconductor, the bands are broadened and softened at the highest energy (61 meV). This is consistent with strong electron-phonon coupling expected by the BCS theory. Further studies were carried out with substitution of ^{18}O for ^{16}O .²⁰ The observed DOSs are similar, except for a shift of 3 – 4 meV to lower energies in the higher-mass isotope spectrum above 20 meV. The calculated DOS agrees with the results, and the calculated oxygen isotope effect coefficient α , which relates T_c to isotope mass, agrees well with the measured value. Thus, this highly coordinated study shows that $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ is a BCS superconductor, and given the T_c of 30 K, it represents a good base material for considerations of the higher T_c , two-dimensional Cu-O-based materials.

$\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$: Whereas most of the rare earths can substitute for Y in YBCO with little effect on superconducting properties, $\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$ (PBCO) is a nonsuperconducting insulator. $\text{Y}_x(\text{Pr},\text{Ba})_{2-x}\text{Cu}_3\text{O}_7$ compounds are

superconducting when $y \leq 0.5$, with T_c decreasing as a function of Pr content. Measurements of the DOS of YBCO and PBCO on the Low Resolution Modulated Energy Chopper Spectrometer (LRMECS) at IPNS [Fig. 4] show that the phonon modes for unsubstituted YBCO are softer than those for the Pr-substituted PBCO between 75-85 meV, unlike the zone center modes obtained in IR and Raman spectra.⁴¹ The results support a model where the modes related to vibrations of the Cu and O atoms parallel to the basal plane may be of importance to the high T_c 's.

Polarized Neutron Scattering

The use of polarized neutron scattering to determine the depth profile of the magnetization of materials was pioneered at IPNS with the polarized neutron reflectometer POSY, and this technique has also been applied to superconductors. In the first such study, the superconducting penetration depth of a film of Nb metal was measured directly.⁴² Later studies on Pb and Pb-Bi films showed incipient type-II superconductivity in the latter material, supported by the apparent observation of the surface sheath between H_{c1} and H_{c2} (all of these

studies are modified by surface roughness, which complicates the analysis but also gives data on the roughness).⁴³ Studies of YBCO films show most of them to be nonhomogeneous.⁴⁴ Such samples, deposited epitaxially on SrTiO_3 and requiring heat treatments to yield sharp transitions, exhibit poor neutron reflectivity until the outer 0.5 μm has been polished off. Penetration depths of 1400 Å (with a roughness of about 40 Å) at 14 K and an applied field of 400 Oe are comparable to those found from muon resonance measurements.

Magnetic Studies

$\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$: Although detailed studies of antiferromagnetic structures of La_2CuO_4 have been extensively studied at reactor sources, similar work has also been carried out at IPNS. The magnetic response of PBCO has been compared to that of $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$, where sharp peaks due to crystal-field effects are seen. This is interpreted to mean that no interference occurs between the Nd^{3+} ions and the superconductivity because of the low density of charge carriers around the rare earth sites. However, for PBCO, no sharp crystal-field⁴⁵ effects were found, but a

broadened quasielastic component is seen up to 10 meV.⁴⁶ The results indicate that a picture of a single-ion configuration for Pr in pure PBCO is not applicable. It may be that exchange coupling between the Pr f-electrons and the conduction electrons may be responsible for the breaking of Cooper pairs in PBCO.

$\text{Sr}_2\text{CuO}_2\text{Cl}_2$: With the installation of the Booster target, the Single Crystal Diffractometer (SCD) at IPNS has become increasingly useful for studies of magnetic scattering. In one such experiment, the magnetic form factor of $\text{Sr}_2\text{CuO}_2\text{Cl}_2$, a compound related to $(\text{La,Sr})_2\text{CuO}_4$, was derived.⁴⁷ Neutron powder diffraction measurements confirm that this insulating compound, with an ordered moment of 0.34 μ_B at 10 K, is structurally related to the doped La_2CuO_4 superconductor, with the Cl atoms in the out-of-plane axial sites. The derived magnetic form factor of the Sr compound is quite similar to that of La_2CuO_4 . This suggests that deviations from the magnetic behavior of the free Cu^{2+} ion is not due to some small magnetic moment on the out-of-plane site, as had been suggested by band theory. A dipolar spin polarization on the in-plane oxygen ions gives improved fits to the data.

Fig. 4 Measured phonon density of states of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (circles) and $\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$ (dotted line) at 15 K with $E_0 = 120$ meV. The solid and dashed vertical lines represent the measured frequencies of Raman and IR bands, respectively.

Practical Superconductors

The difficulties of preparing flexible wires of oxide superconductors limits the commercial use of these materials. IPNS has been used to follow internal stresses and texture in wires prepared by various routes.

Internal Stresses: Dense specimens of YBCO were prepared by controlling oxygen partial pressure, and the effects of grain size were evaluated by measuring the internal strains using the GPPD.⁴⁸ The strain distributions and particle sizes were determined from the diffraction peak widths. The results indicate that the internal stresses are due to grain anisotropy, which is relieved in large grains by microcracking. Strength was found to increase with decreasing grain size and with a decrease in residual tensile stresses from grain anisotropy.

Texture: Preferred orientation measurements of slip-cast YBCO using the SCD at IPNS revealed clear indications of the expected 001 texture parallel to the settling direction (Fig. 5).¹⁷ This work was preceded by the development of generalized software to yield quantitative stereographic pole figures. This effort also forms the basis for an extensive study of bulk materials processed by sinter forging; the study is now underway.

Fig. 5 Contour plots of single-line slices for a slip-cast sample of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ oriented with the settling direction (a) perpendicular and (b) parallel to the diffraction vector. Note enhancement of $\{00l\}$ directions parallel to the settling direction.

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CHAPTER 9

ORDER IN DISORDERED SYSTEMS

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Introduction

Structural studies of materials are revealing an incredible richness in the varieties of order and disorder which can be exhibited by a condensed system. Ten years ago, materials were basically divided into two types, crystalline and disordered, depending on whether or not they possessed the property of long-range order. From an experimental point of view, methods for tackling the two kinds of structure were well defined: for crystalline materials, unless a suitable single crystal was available, one analyzed powder diffraction data with the Rietveld profile refinement technique whose power had recently been established by work at Argonne National Laboratory (ANL); the Institut Laue-Langevin (ILL), France; and other neutron centers, while for those less fortunate materials like liquids and glasses, not blessed with long-range order, one had to make recourse to an average, neutron-weighted *structure factor* (*sf*) and its image in reciprocal space, the total *radial distribution function* (*rdf*). This dichotomy was partially reflected in the initial complement of diffraction instrumentation at IPNS: for powder measurements, two dedicated diffractometers were established, nicely optimized thanks to the pioneering work of Jim Jorgensen and co-workers at the ZING prototype pulsed sources, while for disordered materials, no such instrument was provided. However, measurements on these systems were generously permitted on the Special Environment and General Purpose Powder Diffractometers (SEPD and GPPD, respectively), and a somewhat makeshift family of analysis routines was generated, by the foolhardy pioneers unwise enough to venture into this territory, to produce the required data in *sf* and *rdf* form.

Ten years later, the situation has radically changed, and we

recognize a great variety of types and degrees of ordering between the two extremes described above. This has come about, in a sense, as a result of advances into the middle ground from both directions. Work on disordered materials has shown that, while long-range order in the form of perfect translational symmetry is lacking, substantial intermediate-range, as well as short-range, order may exist, at the same time as investigators of crystalline materials have found that correlations between displacements of nearby atoms, similar to those described by the *rdf* in amorphous materials, are involved in the complex structures such as oxide superconductors. In the middle of all this is the discovery of

quasicrystals as a fundamentally new state of matter. A crucial aspect of many of these developments is the distinction between translational and orientational order, and one reason for the complex situation that we now perceive is that the two types of order may be present to differing degrees in a given class of materials. This is illustrated in Fig. 1, which shows some of the varieties of types of order that are found in condensed matter. We see that long-range order, which can be both translational and orientational, is a well-defined boundary, but that intermediate and short-range order can vary continuously over a substantial range, and that the combination of these different types of order

Fig. 1 Types of order and disorder.

leads to a rich diversity in atomic structure and the physical phenomena associated with structure.

From an experimental point of view, the importance of disordered materials has been recognized by the funding and construction of a dedicated instrument, the Glass, Liquid, and Amorphous Material Diffractometer (GLAD), for studying the structures of these materials. From a dynamical point of view, the Low Resolution and High Resolution Medium Energy Chopper Spectrometers (LRMECS and HRMECS, respectively) are ideally suited to studying the excitations of disordered materials, especially at higher energies, and measurements of this kind are becoming an increasingly active component of the scientific programs of these instruments.

Work at IPNS has contributed significantly to all these developments, and the aim of this article is to give a flavor of a few of the most outstanding developments. In so doing, it should be remembered that the work at ANL has been carried out in a era of dramatic developments in this field, some of which have been made at other neutron, and some x-ray, centers. In particular, we should mention the pioneering experiments on slow relaxation processes at the liquid-glass transition performed with spin-echo techniques at the ILL, and at the same institute, the high-quality structural measurements on liquids with isotope substitution at the D4 diffractometer. Also, many of the concepts of atomic order and disorder discussed in this article are parallel in the magnetic behavior of materials, but that, as Sherlock Holmes says, is another story.

Intermediate-Range Order in Glasses

The first measurements of disordered structures at IPNS were made on chalcogenide glasses by two different groups, by Faber et al. on GeS₂ using the GPPD¹ and by Price et al. on P₂Se₃ using the SEPID.² The latter study, stimulated by the visit of Masakatsu Misawa (National Laboratory for High Energy Physics, KEK, Japan) to Argonne inaugurated an extensive, systematic study of chalcogenide

and oxide glasses by David Price, Sherman Sushun, and several postdoctoral appointments and students.³ Among other aspects of glass structure revealed by these experiments is the importance of intermediate-range order, representing the organization of short-range structural elements in the glass into an extended network.⁴ Subsequent work has identified the universal nature of intermediate-range ordering, as manifested by the "first sharp diffraction peak" (FSDP) in the *s*f and by "companion" or so-called "defect" lines in the vibrational spectrum over a broad range of amorphous materials. While the intermediate-range order is a unifying feature in the structure of these glasses, a distinguishing feature appears to be the "network dimensionality" of the glasses. From this point of view, P₂Se₃ turned out to be a rare example of zero network dimensionality, being composed of molecularlike P₂Se₃ entities held together by a "glue" of superfluous atoms. The *s*f for P₂Se₃ glass, with

its dramatic FSDP,⁵ is shown in Fig. 2; the inset shows the P₂Se₃ unit which represents the building block of the structure.

Ordered Liquid Alloys

Liquids, normally considered a highly disordered state of condensed matter, may have a degree of short- and even intermediate-range order.⁶ A dramatic example of the latter is provided by eutectic alloys of alkali metals and polyvalent metals, studied extensively by Marie-Louise Saboungi and co-workers. In these alloys, charge transfer, followed by covalent bonding effects, leads to remarkable electronic, thermodynamic and structural properties, including appearance of a pronounced FSDP.⁷ These properties can be explained in terms of complex structural units of the form A₂M₂, consisting of a tetrahedron of atoms of the alkali metal A surrounded by a larger tetrahedron of polyvalent atoms M pointing in the opposite direction. In the crystalline solid, these

Fig. 2 Composite average structure factor $\langle S(Q) \rangle$ for Se (Curve A), $P_{0.01}Se_{0.99}$ (B), $P_{0.1}Se_{0.9}$ (C), $P_{0.2}Se_{0.8}$ (D), $P_{0.3}Se_{0.7}$ (E), and $P_{0.4}Se_{0.6}$ (F). Successive curves are moved up by 0.2. Inset: Structure of the P₂Se₃ molecule.

structural units orient in a particular pattern, pointing in an ordered sequence of four allowed directions. A high degree of both translational and orientational order persists to the liquid phase. Melting can take place in a number of possible ways and, in at least one case (CsPb), orientational melting precedes translational melting, leading to an intermediate, plastic-crystal phase.¹¹ The sf for the three phases of CsPb—crystal, plastic crystal, and liquid—is shown in Fig. 3.

Molten salts can also exhibit a substantial degree of intermediate-

range order. A systematic study of trivalent salts, MX_3 , has shown that some melts are made up of structural units in the form of two edge-linked tetrahedra, M_2X_6 , while others form more or less densely packed arrays of individual M^{3+} and X^- ions. Here again, melting can take a number of different forms. An unusual case is that of $AlCl_3$, which forms a densely packed array in the solid, with Al^{3+} ions in octahedral environments, but on melting becomes a molecular liquid composed of Al_2Cl_6 units;¹² a very large volume change (88%) is

observed on melting. If an alkali halide AX is added to MX_3 , even more complex structures, with three or more edge-linked tetrahedra, can be formed.¹³ This can be understood in terms of the screening effects of the A^+ and X^- ions which create a local environment in which more complex species can be stabilized.

Quasicrystalline and Polycrystalline Materials

Systems with long-range order, and consequently well-defined Bragg peaks, have traditionally been studied with crystal diffraction techniques which address the intensities in the Bragg peaks as a function of wave vector. In this case, one is looking at elastic scattering ($k = 0$) which is related via a Fourier transform to the time-average structure and thus gives the arrangement of the *equilibrium* positions of the atoms, which is normally the subject of interest in a crystalline material. However, the rdf is also a well-defined quantity for such systems, and this can be measured with the same techniques as those used for the amorphous solids and liquids described above.¹⁴ The rdf is related to the sf which, as discussed in the next section, is an integral over all k and therefore, related to the *instantaneous* structure of the material. In many cases, this reveals short-range correlations between nearby atoms which the conventional diffraction techniques cannot observe. This method has been applied to a variety of materials by Takashi Egami and his co-workers at the University of Pennsylvania.

This technique has been applied with great effectiveness to *quasicrystals*, a new state of condensed matter, discovered a few years ago, which is generally agreed to be made up of building blocks or tiles which have perfect orientational order extending over a macroscopic dimension of the sample but without the translational long-range order of normal crystals (see Fig. 1). Such a material will have Bragg peaks, some of which are much larger than others, but in fact these are infinitely dense in reciprocal space. Clearly, the rdf in a system of this kind contains a

Fig. 3 Measured structure factors of CsPb at (a) 293 K, (b) 660 K, and (c) 930 K.

wealth of information. One system studied at IPNS by the University of Pennsylvania group is tetrahedral Al-Mn-Cr-Si.¹⁷ Fig. 4 shows the differential distribution function (ddf) around the Mn atoms obtained from the difference of the scattering from $\text{Al}_{70}\text{Mn}_{30}\text{Si}_{10}$ and $\text{Al}_{70}\text{Mn}_{10}\text{Cr}_{20}\text{Si}_{10}$, assuming a random substitution of Mn by Cr. The Mn ddf is in good agreement with the vortex-vortex distances of a quasicrystalline lattice, indicated by arrows in Fig. 4, confirming the presence of a quasicrystalline substructure, decorated by Al and Mn atoms, in this material.

Even materials which are crystalline in the normal sense may have important information lurking in the rdf. An important class of such materials is the high-temperature oxide superconductors which have been extensively studied at Argonne and elsewhere over the last few years. An example of the power of the rdf technique is given by the study of the superconductor $\text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$, where rdf measurements made by Dmowski et al.¹⁸ at IPNS (Fig. 5) demonstrated Ti-O displacements having only short-range order, while conventional crystal structure analysis was forced to model the Ti-O plane with a disordered structure. Subsequent experiments by Toby et al. revealed short-range correlated displacements in the Cu-O(1) plane which correlate remarkably with the onset of superconductivity.¹⁹

Dynamics of Disordered Systems

Substantial work on the dynamics of disordered materials has been carried out on the chopper spectrometers at IPNS. Here the quantity measured is the scattering function or dynamic structure factor (dsf), which shows the energy dependence $S(\mathbf{Q}, E)$ of the sf $S(\mathbf{Q})$, where $\mathbf{Q}$ is the scattering vector and E , the energy transfer in the scattering event. By measuring the average of the dsf over a wide range of $\mathbf{Q}$, we can obtain the generalized vibrational density of states (vdos). This usually exhibits a number of features characterizing the dominant vibrational normal modes of the system, somewhat similar to those measured in an optical Raman scattering experiment. In the case of

Fig. 4 Differential distribution function around Mn obtained by comparing the scattering intensities from $\text{Al}_{70}\text{Mn}_{30}\text{Si}_{10}$ and $\text{Al}_{70}\text{Mn}_{10}\text{Cr}_{20}\text{Si}_{10}$, assuming a random distribution of Mn by Cr. The arrows indicate the quasicrystalline vortex-vertex distances with the quasicrystalline lattice parameter of 4.60 Å.

Fig. 5 Atomic pair distribution function (pdf) of $\text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$. In the upper plot, the solid line denotes the experimental data and the dotted line, the pdf calculated from a model of local ordering of O and Ti atoms. The lower plot shows the pdf's calculated from other models which do not fit the data as well.

inelastic neutron scattering, however, we can obtain information about these modes from the Q dependence of the $d\sigma/d\Omega$, which follows from the correlated vibrational motions of neighboring atoms in a particular normal mode. This technique has been applied to considerable effect by a group at the Pennsylvania State University on amorphous semiconductors, such as amorphous Ge,¹⁴ and amorphous metals,¹⁵ and by the Argonne group on the chalcogenides and oxide glasses discussed above. A beautiful example is GeSe_2 , where the features observed in the v_{los} , shown in Fig. 6, can be associated with the normal modes of an isolated GeSe_4 tetrahedron which, for this glass in particular, represent the normal modes extremely well.¹² The Q dependence of the $d\sigma/d\Omega$ for one of these modes is shown in Fig. 7 and is seen to be very well matched by that calculated for an isolated tetrahedron.

GLAD

The new Glass, Liquid, and Amorphous Material Diffractometer is one of two instruments in the world expressly designed for structural measurements on disordered materials with very short-wavelength neutrons.¹⁶⁻¹⁸ These have the advantage that the effects of inelasticity on the measured $d\sigma/d\Omega$ of materials with light atoms, and liquids in particular, are minimized, as are those due to absorbing nuclei, which are often encountered in the increasingly important isotope substitution experiments. Funded by the Department of Energy through a proposal submitted by a Participating Research Team (PRT) headed by Simon Moss at the University of Houston and through direct funding at IPNS, GLAD incorporates banks of linear position-sensitive detectors recording time-of-flight spectra from neutrons in the wavelength range of 0.1-2 Å. To measure small wave vectors with the shortest-wavelength neutrons, the smallest scattering angles must be accessible. This is achieved at GLAD by the novel arrangement of having the incident beam pass through one of the detector banks. The enormous

data rates inherent in the GLAD detector scheme are handled with a state-of-the-art data acquisition scheme (FASDAS) designed by Kent Crawford. The large, 3-m-long evacuated flight path, with thin windows in front of the detector banks, is another Bob Klob design special, and an impressive array of sample environment apparatus, involving the capability for han-

dling multiple samples at low, ambient, and high temperatures, and eventually at high pressures, has been assembled by Sherman Susman and Ken Vollrath. The whole diffractometer has been put together by the IPNS operations team headed by Don Bohringer and Dave Louch and carefully got under sail by Dan Montague (Willamette University) and Adam Ellison.

Fig. 6 One-phonon vibrational density of states $G(E)$ from GeSe_2 glass at 13 K.

Fig. 7 Dynamic structure factor $S(Q, E)$ from GeSe_2 glass measured at $E = 25.91$ meV (circles and solid line) compared with that calculated for the v_1 tetrahedral breathing mode (dashed line).

GLAD is operating with six detector banks out of a total complement of ten; others will be added as funds become available. The scientific program with experiments from the PRT is just now getting underway, and general user experiments will start in Spring 1992. An example of the first data from the present phase of GLAD is shown in Fig. 8.

Summary

In this article, we have tried to give a flavor of the complexity of the types and degrees of order exhibited by systems normally considered disordered, and new kinds of order found in systems normally considered ordered. We may expect that still more manifestations of symmetry in materials are waiting to be discovered, and that IPNS, with its powerful complement of instruments that are well suited for investigating the structural and dynamical aspects of order in condensed matter, will remain at the forefront of this rapidly expanding field.

Acknowledgment

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CHAPTER 10

SMALL ANGLE NEUTRON SCATTERING WITH THE SAD AT IPNS

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Introduction

Somewhat contrary to early expectations, pulsed neutron sources equipped with cold moderators prove to be especially well adapted to small angle scattering applications. The IPNS Small Angle Diffractometer (SAD) is a general purpose, short-flight-path, time-of-flight (TOF) instrument, shown schematically in Fig. 1. Neutrons are produced in short bursts by spallation when energetic protons impinge on a uranium target 30 times per second. A cold moderator is required to produce a neutron spectrum suitable for small angle scattering (i.e., a spectrum enhanced in long wavelength neutrons). A cold, single-crystal MgO filter positioned in the incident beam removes most of the fast neutrons. The instrument has a short flight path to provide access to a large wavelength range in the face of "frame overlap" constraints. In view of the modest resolution requirements for small angle scattering, even short flight paths provide adequate wavelength resolution for many purposes.

Collimation of the neutron beam is achieved by use of a crossed pair of Soller elements. Scattered neutrons are detected by an area-sensitive, gas-filled proportional counter, and monitors for the incident and transmission measurements are of the BF₃-type. The SAD uses neutrons in the wavelength range from 0.5 to 14 Å and covers the Q range from 0.005 to 0.35 Å⁻¹.

Pulsed spallation or pulsed reactor neutron sources are currently in use at Argonne National Laboratory (ANL) and at four other laboratories throughout the world. Small angle neutron scattering (SANS) capabilities are being developed at each Institute at the Laboratory for High Energy Physics (KEK, Japan); at the Rutherford-Appleton Laboratory, U.K.; at the Joint Institute for Nuclear Research in Dubna, U.S.S.R.; and in the U.S., at the Los Alamos National Laboratory¹ and at the ANL with IPNS².

At the time development of the IPNS-SAD began, there was no generally agreed upon design for such an instrument at a pulsed source. Rather, its development

has been, and continues to be, an evolutionary process. The tenth anniversary of the IPNS appears to be an appropriate time to recount briefly its evolution, to discuss its present configuration and some of the science now being produced, and finally, to indicate modifications being incorporated in a second small angle neutron diffractometer (SAND) currently being constructed at IPNS.

Neutron Production and Moderation

Neutrons are produced at the IPNS by a pulsed spallation process. The process begins by production of H⁺ ions in the source, acceleration to 750 keV in the preaccelerator, and acceleration to 50 MeV in the linear accelerator. At injection into the Rapid Cycling Synchrotron (RCS), two electrons are stripped away from each H⁺, and the resulting protons are accelerated to 450 MeV. The protons are extracted in a single revolution in the RCS and are directed to the target; this process is repeated 30 times per

Fig. 1 Schematic representation of the components comprising the IPNS-SAD. Drawing is not to scale.

second. Nominally, very short pulses of protons (100 ns) at 30 Hz impinge on the target.

The original target at IPNS was made of depleted uranium, but, in order to gain a factor of about 3 in flux-on-sample, it was replaced by the Booster target. The present Booster target at the IPNS is a water-cooled stack of 77.5% ^{235}U disks, decoupled from its surroundings by a layer of ^{10}B which maintains primary neutron pulses less than 0.3 μs in width. With 450-MeV protons and a time-averaged proton current of 15 μA , about 4×10^{16} neutrons are produced.¹⁰

Currently, the moderator on beam line C1, on which SAD is located, consists of liquid hydrogen at 20 K in a 0.7-L aluminum can which is grooved on the viewed side to increase the neutron beam intensity,¹¹ since resolution is not a prime concern in SANS. The moderator is surrounded by graphite and beryllium as reflector and is also covered by a 0.5-mm layer of cadmium.

Collimation of the Neutron Beam

When short flight paths are required, as with TOP SANS instruments, multiple-aperture converging collimation provides the maximum intensity on the sample for a given resolution.¹⁰ In the SAD, such multiple-aperture collimation of the neutron beam is accomplished by use of a crossed pair of Soller elements, as indicated in Fig. 1. The Soller elements in use on the SAD were fabricated by CHIC Ltd., Cheltenham, U.K. Each element was designed to focus the neutron beam on the detector plane, some two meters beyond the last collimator exit as shown in Fig. 1. Each element contains 22 vanes consisting of aluminized mylar stretched over tapered stainless steel frames, and the vanes are coated with a ^{10}B -containing paint. The collimators were designed to yield a full width beam divergence of 0.43°, and the penumbra measured with the area detector is in excellent agreement with the design calculations.

Unfortunately, the original set of Soller elements produced an anisotropic background as illus-

Fig. 2 Anisotropic background produced by the original set of Soller collimators. A beam stop of $^{6}\text{Li}_2\text{CO}_3$ has removed the direct beam. The underlying grid represents the virtual elements into which area detector data are stored.

trated in Fig. 2. The integral contribution from the "wings" was about half that of the total empty-camera background and amounted to 3×10^{-3} times that of the direct beam. This anisotropic background was significant for weak scatterers and was especially troublesome for anisotropic scatterers. It was found necessary to use software masks to exclude the affected regions of the collected data, a procedure which sacrificed an unacceptable fraction of the solid angle sampled by the area detector. A second set of Soller elements, in use since May 1988 and again manufactured by CHIC with somewhat rougher surfaces (matte finish), has effectively eliminated this anisotropic background component.¹²

Historically, the first functional version of the SAD used a series of "pinhole" apertures fabricated from

B_2C in waterglass and sized to focus a single beam of neutrons at the detector plane. An earlier effort to use a bundle of seven tapered stainless steel tubes as a focusing collimator in a prototype instrument was a disaster.

Experiences with the Neutron Spectrum

In the SAD, the detectors are in a rather hostile environment in that they view the moderator directly. To be sure, unless the beam is being attenuated, a massive beam stop of $^{6}\text{Li}_2\text{CO}_3$ inside the secondary flight path shields the area detector from the direct beam. The ^6Li isotope is used in order to avoid gamma-ray production in the beam stop. Fusing the $^{6}\text{Li}_2\text{CO}_3$ was found to lower the background contribution by a factor of 3.5 relative to that obtained with a beam stop made from powdered

$^6\text{Li}_2\text{CO}_3$ compacted into a thin-walled, high-purity aluminum can.

Early on, a slow recovery of the area detector was noted after bombardment by the fast neutrons and/or gamma-rays present in the prompt pulse. A single-crystal Be filter, refrigerated with liquid nitrogen, was designed initially for use in the beam to eliminate the fast neutrons, but due to the fact that such a filter removes those neutrons with wavelength less than 4 Å, it was not much used in experiments. However, after recognition of the recovery problem, a single-crystal MgO filter, again refrigerated with liquid nitrogen, was installed in the incident beam. This filter has a gradual, short-wavelength cutoff between 1-2 Å and a transmission of approximately 1% for neutrons with wavelength below about 0.1 Å. About 75% of the neutrons with wavelength greater than 2 Å are transmitted. The MgO filter was installed in Fall 1988 and, in the absence of a "prompt pulse removal" chopper, is deemed an essential part of the instrument for removing the unwanted, fast part of the neutron spectrum.

It is perhaps worth recounting as an aside how a special characteristic of the MgO filter is used to advantage in instrument calibration. Short wavelength neutrons are removed from the beam by means of Bragg scattering from the MgO, the result being a series of sharp negative spikes in the transmitted neutron beam in the range below about 2 Å. This pattern has been indexed by Carpenter et al.,¹² so we know the wavelength corresponding to each transmission dip. We use these negative lines as tags on the spectrum and thus are able conveniently to determine the distances from the moderator to the area detector, to the transmission and incident beam monitors, and to the sample position (with a monitor detector temporarily placed in this location).

As a consequence of our using the Bonister target containing the fissionable material ^{235}U , in addition to the prompt neutrons, a contribution from delayed neutrons results. These delayed neutrons are of all wavelengths and are emitted

Fig. 3 The relative distribution of neutron wavelengths viewed by the SAD with the MgO filter in place. The filled circles are raw data, and the open circles, the same data corrected for delayed neutrons.

uniformly over the full duty cycle. The fraction of delayed neutrons has been measured by Epperson et al.¹¹ and found to be 0.0283. (In an earlier measurement when the depleted uranium target was being used at IPNS, a delayed-neutron fraction of 0.0044 was measured.) The number density of neutrons in the prompt spectrum is strongly wavelength dependent, decreasing with increasing wavelength beyond the Maxwellian peak. Not only do the delayed neutrons produce a sample-dependent background component, the fractional contribution from delayed neutrons depends with which part of the spectrum one is dealing; it is especially significant in the long wavelength part of the prompt spectrum which yields the low-Q information. Figure 3 gives an example of a normalized spectrum, as measured and after correction for delayed neutrons. The procedure used to correct for delayed neutrons is formally independent of the nature of the sample, so a general routine is incorporated in the data reduction software (REDUCE) for removing the delayed neutron contribution.

Absolute Intensity Calibration

Since neutrons with a wide range of wavelengths (0.5-14 Å) are being used in the SAD, the scattering data must be normalized for the spectral distribution when the data are being reduced. Typically, the incoherent scattering from a 1-cm water sample is used for determining the detector sensitivity and as an absolute intensity standard in the case of reactor-based instruments. In other cases, the incoherent scattering from vanadium is used for this purpose.

During the initial stages of development of the SAD, we attempted to use a single crystal of vanadium for this purpose; however, this had to be abandoned because of the enormous counting times required to obtain acceptable statistics. Later, a water sample was tried. Since water is a stronger scatterer than vanadium, a few hours of beam time were sufficient to obtain a relative spectrum with adequate statistics. The total number of neutrons seen by the area detector in each time slice has been taken as the relative spectrum. In order to assess the quality of the

spectrum, an irradiated aluminum sample (Al-4) containing micro-scope voids was provided by Kottler¹⁴ for which the Guinier radius ($R_g = 200 \text{ \AA}$) and the absolute scattering cross section ($I(0) = 100 \text{ cm}^{-1}$) were known. Only a limited range of long-wavelength neutrons were used in the SAD determination of R_g because of its magnitude, so this sample was not well suited as a standard. The SAD data initially compared poorly with that obtained from the 30-m machine at the High Flux Isotope Reactor at the Oak Ridge National Laboratory (ORNL). However, when the SAD spectrum, determined from a water measurement, was modified by applying a correction which is based on the fact that the incoherent scattering from water is proportional to the factor $(1-T)/4\pi$, where T is the transmission of the water sample, agreement was greatly improved. Because of the fact that the water measurement required some 6 to 8 hours and that some important questions remain about the water cross section, its use was also discontinued.

In current practice, the spectral distribution of neutrons is determined directly by using the area detector; a cadmium mask filled with tiny holes is used to attenuate the beam without altering the spectral character, and the beam stop is removed. An example of a spectrum measured in this manner, before and after correction for delayed neutrons, is shown in Fig. 3.

The absolute calibration of the SANS data is essential if maximum use is to be made of the data (e.g., to determine the molecular weight or volume fraction of the scattering entities). For this purpose, we use, as a secondary intensity standard, a polymer melt sample ("Bates poly") which is a mixture of equal volumes of deuterated and hydrogenated high molecular weight polystyrene whose absolute cross section was measured at ORNL.¹⁵ The standard was kindly provided to us by Frank Bates (University of Minnesota). By comparison of the reduced SAD data, in relative units, with the absolute Bates-ORNL data, a scale factor is determined which allows our data to be converted to

units of cm^{-1} per unit steradian. An example of this comparison is shown in Fig. 4. This conversion to absolute cross sections is routine practice with IPNS-SAD data.

Detectors and Data Collection System

The detection system for the SAD consists of an incident beam monitor, a transmitted beam monitor, and an area detector for counting the scattered neutrons. In addition, the capability exists to measure the total number of protons striking the target during a run.

The incident beam monitor is a low-efficiency transmission-type detector which samples the onfibre cross section of the incident beam and provides a continuous monitor of the intensity and the wavelength distribution, essential information inasmuch as the proton current varies and there may also be slight excursions in the moderator temperature. Initially, a silicon surface-barrier detector was used for this purpose. Detectors of this

type were found to be unsatisfactory for use as incident beam monitors due to their susceptibility to fast neutron damage. It is not known if a surface-barrier detector would be stable when an MgO filter is used to remove the fast neutrons, as is currently done. A commercial, planar, pulsed-ion chamber containing BF₃ as the neutron absorber and a mixture of 90% Ar and 10% CF₄ as the background gas (Model RS-P1-3462-101 from Ruter-Stokes, Twinsburg, OH) has been in service for approximately six years without noticeable degradation of efficiency or other undesirable behavior.

Initially, transmission measurements were made with the area detector. However, this required repositioning the beam stop, and it was soon recognized that installation of a separate detector for this purpose would result in a much more convenient mode of operation. A second consideration was the difference in size of the resulting data files, inasmuch as disk space was extremely limited at the time.

Fig. 4 Comparison of the scattering from the secondary intensity standard, "Bates poly", measured at ORNL (open circles) and at the IPNS-SAD (filled circles). The scale factor for conversion of SAD data to absolute cross sections is obtained by fitting the profiles in the range $0.035 < Q < 0.055 \text{ \AA}^{-1}$.

For roughly five years, a 2.5-cm-diameter $\times$ 30-cm-long cylindrical gas proportional counter containing BF_3 has been in use. This detector has an efficiency of 2×10^{-4} at 1 Å. It is shielded with a cadmium mask which restricts the area of neutron detection to a 2.25-cm-diameter circle centered on the transmitted beam and ensures that most of the SANS from the sample is excluded from the transmission measurements. This aperture is made slightly oversize to facilitate alignment of the monitor and mask. Positioning of the transmission monitor is under computer control.

The area detector used for the SAD is a gas proportional counter with an active volume of $20 \times 20 \times 3.2 \text{ cm}^3$, and it is filled with a mixture of 65% He and 35% CF_4 to a total pressure of 2 atm. (Originally, the detector contained xenon as the quench gas, but this was replaced with CF_4 to reduce the gamma-ray sensitivity.) This detector is of the Borkowski-Kopp type¹⁶ and contains an anode plane sandwiched between front and rear single-wire cathode planes which yield the x- and y-position information. Pulse-height discrimination is used to eliminate events due to gamma-rays and fast neutrons. Nominal resolution should be about 3 mm, but the lowest achieved is about 5 mm full width at half-maximum (fwhm) when short amplifier time constants (0.5 μs) are used. Longer time constants produce much better linearity of the position encoding, and a time constant of about 5 μs is used, which results in a measured resolution of 9 mm fwhm. Even with the longer time constant, significant regions near the detector edges must be discarded due to nonlinearities, resulting in an effective active area of about $19 \times 19 \text{ cm}^2$. It is of some historical interest to recall that in the days of prototype instrument development on the ZING-1¹⁷ pulsed source, this small area detector was actually shared by the SAD and the single crystal diffractometer.

Digitized signals from the direct-time digitizer are initially stored, along with an encoded

neutron flight time, in a buffer in a CAMAC module. A CAMAC "clock module" provides an 8-MHz timing signal utilized by all the TOF encoding modules in the system. The clock module uses the CAMAC dataway to zero a scalar at the time the proton pulse strikes the target, so the scalar keeps a running count of time (in 0.5 or 1.0 μs clock ticks) since the last pulse of prompt neutrons was produced. When a timing pulse is received from the area detector encoding electronics, the current value of the scalar is the neutron TOF (16 bits), and this, along with 16 bits of x-y information from the area detector position encoding electronics, is loaded into the "first-in-first-out" (FIFO) buffer.

The microprocessor system¹⁷ utilizes multiple microprocessors which are dedicated to histogramming the events from the area detector and the two monitor detectors. When the microprocessor system is not involved in transferring data from the FIFO buffers to a buffer region in the microprocessor memory, it processes data from these buffer regions to form TOF histograms for the two monitors and an x-y histogram for the area detector data. The histogramming is tube-driven and is under complete software control. Normal operation for the SAD involves 67 time slices and 64×64 spatial bins in each time slice.

A Digital Equipment Corp. DEC/PDP-11 computer originally served as the general user interface for the instrument, but it has been replaced by a DEC MicroVAX VAXstation-II workstation. This computer is used to set up the histogramming tables and download them to the microprocessor system, and it also maintains a disk file system for completed histogram data. A color graphics system on this computer allows the user to display live histograms as it is being acquired or file data of previously acquired histograms. Both color density displays of area detector data and standard plots of beam monitor spectra or of particular cuts through the area detector data are available. The memory and

disk file storage limitations of the PDP computer prevented its use for any significant amount of on-line data reduction or analysis. These limitations have been partially removed by the installation of the MicroVAX, which may now be used for data analysis as well as for the instrument interface. Equally important is the fact that the 600-Mbyte disk on the workstation allows storage of a far larger number of data histograms and hence allows a longer time window for data reduction without additional archival and retrieval operations. An Ethernet network connection allows easy transfer of completed histogram files or reduced data to other data analysis computers for further processing or for transfer to transportable media.

Ancillary Equipment

The IPNS-SAD operates on the basis of proposals which are reviewed for scientific merit by the IPNS/LANSCE Program Advisory Committee. The instrument is utilized largely by metallurgists, chemists, biologists, polymer scientists, and solid-state physicists. Consequently, capabilities for a range of sample environments are required. Furnaces, closed-cycle refrigerators, cryostats, pagnols, and pressure cells (sometimes used in combination) are available, as is a seven-position sample changer. The equipment available for use with the SAD and the relevant ranges of conditions are summarized in the table.

Examples of Science Being Done with the SAD

The principal advantage of a small angle diffractometer operating on a pulsed neutron source is the fact that scattering over an extended range in Q space is sampled with a single setting of the sample-detector distance; the extended Q range is a consequence of using a wide band of neutron wavelengths and TOF techniques. For the SAD, wavelengths from 0.5 to 14 Å are used which allows one to scan the range $0.005 < Q < 0.35 \text{ Å}^{-1}$ in a single experiment. This feature, along with

Ancillary Equipment Available at the SAD Instrument

Equipment	Environment	Temperature Range
Sample changer (7-position)	Ambient	0 to 75°C
Biological well (circulating bath)	Ambient	-20 to 100°C
Biological well (resistance heater)	Ambient	Ambient to 200°C
Metallurgical configuration	Vacuum	Ambient
Various furnaces with ceramic sample tubes	Flowing gas atmosphere	100 to 1700°C
Electromagnet ¹	Ambient	Ambient
Electromagnet with Klippert furnace ¹	Flowing helium or Vacuum	Ambient to 750°C
Electromagnet with Displex refrigerator	Vacuum	20 to 300 K
Displex refrigerator	Vacuum	20 to 300 K
Sample rotator (slurry)	Ambient	Ambient
Pressure cell liquids	1 to 2.5 kbar 1 to 2.5 kbar	Ambient 0 to -15°C
Sample stretcher (polymers)	Ambient	Ambient

¹ The existing magnet can achieve about 10 kG with a 1-in pole gap and about 5 kG with a 2-in pole gap.

¹ The use of the furnace requires a 2-in pole gap.

the array of ancillary equipment listed in the table, is now being exploited. A few examples of some of the science being done at the SAD is in order to illustrate the capabilities of the present instrument.

SANS techniques have been used in the field of separation science^{19,21} to provide a better understanding of the interaction of a number of organic solvents which are used to extract selectively such metal ions as Co(III), Pr(III), and

Pu(IV). These studies have yielded information on the size and shape of the polymers formed during the extraction, as well as the nature of bonding in such polymers. It was shown that the polymers formed in the aqueous media assumed various sizes and shapes depending on the conditions of formation. For example, the Pu-polymers prepared in low acid conditions are cylindrical in the aqueous media and are broken in the length direction upon

extraction into the organic phase, as illustrated in Fig. 5.

Studies have been made on lecithin and bile salt mixed colloids in aqueous media at various concentrations, temperatures, and pressures in order to understand the thermodynamics of particle formation.²² Recently, phosphatidyl choline systems have been investigated which exhibit a temperature- and pressure-dependent, reversible gel-to-liquid-

An extensive study of phase separation in supersaturated Ni-Si alloys has been carried out^{20,21} in which it was possible to study, separately, the nucleation/growth and the coarsening stages. It was shown that dynamical scaling behavior takes place in the coarsening regime. SANS measurements shed no light on the possibility that short-range atomic ordering may precede nucleation/growth, and this points to the need to complement such SANS measurements with high-angle diffuse scattering studies.

Aside from a series of more or less minor, incremental changes which would enhance the performance of the JPNS-SAD, several areas have been identified which would result in very significant improvement relative to the present instrument. These improvements are being incorporated into the

Fig. 7 Scattering from polycrystalline Fe-40at%Cr alloy samples. These samples had been annealed 48 h at the respective temperatures, and SANS measurements were made at room temperature.

design for SAND, which is now under construction.

Replacing the present liquid hydrogen moderator with one of solid methane would significantly increase the available neutron flux on the sample; an enhancement of neutron flux in excess of a factor of three could be realized over most of the wavelength range of interest. Such a solid methane moderator was, in fact, in operation for some years at IPNS; however, it was subject to occasional temperature excursions ("burps") in which the radiolytically produced, stored chemical energy was released spontaneously and rapidly, and the aluminum moderator can was sometimes ruptured.¹²⁻¹⁴ Security considerations relative to the current Booster target preclude use of the solid methane moderator until a mode of operation is found in which one can control the release of stored energy and the consequent temperature excursions in the moderator.

An incident beam chopper (or choppers) could be designed to yield several benefits. (1) Elimination of the "initial burst" of fast neutrons would allow the removal

of the MgO filter, which would provide 25% more neutrons at the sample position. (2) Extension of the usable wavelength range downward to a few tenths of an Angstrom would permit nuclear resonance measurements to be carried out at the IPNS-SAD. This would effectively open up a new field for SANS investigation, as has been reviewed by Epperson and Thiyagarajan.¹⁵ (3) At present, some 2.83% of the neutrons appear as delayed neutrons, and many of the most troublesome of these could be removed by a prompt pulse removal chopper. (4) Every second prompt pulse could be chopped out, when desired. This latter optional procedure would allow use of longer wavelength neutrons, one result of which would be to extend the minimum Q range downward and hence, to make the instrument more competitive with steady state source SANS instruments. A chopper for these purposes could be installed and used without the necessity of altering the collimator or the sample-to-detector distance.

One of the principal advantages of a small angle scattering diffractometer of a pulsed neutron

source over its counterpart at a steady state source is the potential for covering a wide-Q range with a single setting of the sample-to-detector distance. This extended Q range is a direct result of having available, and being able to utilize, a wide band of neutron wavelengths. For the important class of crystalline materials, it may not be possible to use the full, available range of neutron wavelengths. These crystalline materials are, of course, capable of Bragg scattering neutrons whose wavelengths are below the Bragg cutoff, defined as twice the maximum interplanar spacing in the material. Such Bragg scattered neutrons are sometimes scattered a second time (or more) into the region sampled by the area detector. When only a few time channels are affected, they can usually be identified and the corrupted data eliminated from further use. However, when the grain sizes are small, this procedure is impractical, and often, all wavelength channels below the Bragg cutoff must be omitted, the net result being a severely restricted Q range for the experiment. The obvious way to cover a wider range of Q without the necessity of using any wavelengths below the Bragg cutoff is to provide a high-angle bank of detectors to supplement the area detector. Figure 8 shows calculated instrumental weighting functions for the present SAD and for SAND with a supplementary array of linear position-sensitive detectors (LPSDs) extending from a scattering angle of 3.5 to 35°. This figure dramatizes the enormous Q range enhancement which would be provided by such a set of LPSDs.

Prompt neutrons are present in the spectrum beyond the 14 Å currently used. To be sure, the flux decreases with increasing wavelength; however, the effect of the longer wavelength neutrons on the measurements is not negligible because the scattering cross section typically increases almost as rapidly as the flux decreases. Mirrors consisting of silicon coated with ¹⁰⁰Ni could be installed in the incident beam and oriented to remove neutrons beyond a specified wavelength.¹⁶ These would, of

Fig. 8. Calculated instrument weighting functions showing the improvement with a bank of high-angle linear position-sensitive detectors (LSPDs).

course, need to be removed if a pulse eliminating chopper were installed to allow use of the longer wavelength neutrons.

Conclusions

Experience has shown that pulsed neutron sources equipped with cold moderators can be exploited for small angle scattering research; a number of the problems encountered in developing SANS capabilities are recounted. A TOP instrument at a pulsed neutron source has one decided advantage relative to its counterpart at a steady state source: it permits measurement over an extended range in momentum transfer space with a single setting of the sample-to-detector distance. Given this inherent advantage and the array of ancillary equipment which has been assembled, good science is emerging from the SAD, as is illustrated with a few examples. Potential improvements in instrumentation have been identified, and these are being incorporated in a second small angle scattering diffractometer, SAND, being built at the IPNS facility.

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CHAPTER **11**

A POSY STORY

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What's in a name?

Did you ever ask yourself what the name "POSY" stands for? The general wisdom is that the name of an instrument should be first, catchy (yeah, like LRMECS, of course!), and second, an acronym of some sort; but if you try to unravel it (and you would not be the first), you would stop hopelessly at the "Y". What is the key? POSY is indeed an acronym - sort of - and a story goes with it.

In November 1984, one of the very ramblentous parties for which IPNS is famous took place. The occasion? Actually there were two: to say goodbye to Roberto Felici, a young Italian scientist who had helped to set up the first reflectivity experiments at beam hole G2, and to celebrate the success of those experiments. Success was such that it had been decided to "permanize" the rickety contraption hastily thrown together and call it an instrument. Any respectable instrument needs a name, and it was decided to choose it in the most democratic way. In the invitation to the party, the revelers were asked to bring their suggestions, from which the name would be chosen by popular vote at the end of the party. Not few were the entries, some of which so colorful that they would not have made their way into a family magazine. When the vote was taken, however, it was the name shown in Fig. 1 that won by standing ovation. Culture had benton budwiness; the godfather, David Prieon, was bunn-ing. He did not know that he had beaten even the entry of the guy who had set up that instrument....

Now seriously, would you use a name like POSEIDON for your instrument? You cannot ask for money from Washington for something that sounds like a sinking ocean liner. You cannot ask with a straight face for a technician to prepare something for POSEIDON. What a silly foreign

Fig. 1 The winning entry for the name of the first reflectometer.

name. Long, too. And if somebody asked to explain the acronym? Embarrassing. On the other hand, other choices were limited indeed. To scuttle the name would have been unsportsmanlike after a full-strength, historic party. The alternative chosen was to snip and shorten the name in the same way in which the kid Poseidon would have been nicknamed by the other kids on the block. True, the nature of that fierce god was somewhat altered, as if by putting in his hand instead of a trident, a bunch of water lilies. But the new name, POSY, was easy on the mouth, and the business with the acronym had been submerged very deep - the only one who knows now is you.

A Prototype

The sketch in Fig. 2 shows that POSY is a very simple instrument indeed: it pinches down a beam of neutrons to a very narrow collimation. The beam strikes the sample surface at grazing incidence and is reflected like light from a mirror.¹ Material mirrors are partially transparent to neutrons, and the amount of neutron "light" reflected

Fig. 2 Logo used when POSY won an R&D 100 (formerly R 100) Award in 1987.

tells a good deal about what the mirror is made of. The reflection takes place because neutrons interact with the material at the surface and the layers beneath it. Actually, there are two kinds of interactions. First, the neutrons are affected by the nuclei with a scattering amplitude, b , which varies not only from element to element, but even for the distinct isotopes of a single element. Also, the neutrons, having a magnetic moment, μ_n , interact with the local magnetic fields, B , in the material; the energy term is $\mu_n \cdot B$. Thus, the reflectivity is linked to the depth profile of the magnetization. The skipping of neutrons off the surface has been compared with playing ducks and drakes with subatomic particles. The analogy is a bit unfair: the slower neutrons have more time to feel the potential of the material and are reflected more readily from the surface rather than sinking into it.

Figure 2 illustrates how the neutrons, with their tiny magnetic dipoles, are reflected from a magnetic surface selectively: the reflectivity depends upon the relative orientation of the neutrons' moments and those of the atoms on the surface and beneath. The prototype reflectometer was set up specifically for magnetic depth profiles near the surface, and early in the game, an unexpected effect was found in magnetic tapes.

Sputtered films of iron oxides form a widely used class of materials for high-density recording media. Traditionally, films of Fe_3O_4 are prepared by reactive sputtering of an iron target in an argon-oxygen plasma. However, as prepared, these films do not make the best magnetic memories (which are those characterized by a high coercive field and a square hysteresis loop). The films improve their magnetic performance if roasted in air at elevated temperatures. It was known that the treatment changes the crystal structure to the γ -phase of Fe_3O_4 ; but how, in detail, does the magnetization change? Figures 3a and 3b show the reflectivity curves for the as-prepared and roasted material. In both cases, the reflectivities are plotted separately

for neutrons polarized parallel and opposite to the magnetic field in which the sample is placed. For Fe_3O_4 , $R^+(\lambda)$ and $R^-(\lambda)$ are roughly parallel, while for $\gamma\text{-Fe}_3\text{O}_4$, they rapidly converge. Figure 3c shows that the heat treatment causes the surface to become magnetically passive for a thickness of 150 Å; later work showed that this was due to the presence of a surface layer of (antiferromagnetic) $\alpha\text{-Fe}_2\text{O}_3$.

Perhaps it was this experiment that convinced the scientific community back in 1984 of the validity of the new technique. Here was a probe capable of measuring at the same time the magnetization at the surface and in the bulk, and it could do so quantitatively and without destroying the sample. Much work has been done since then to elucidate different aspects of magnetism near the surface. Perhaps the most remarkable project was the study of the penetration depth of magnetic fields in superconductors carried out by Ken Gray and Bob Kampwirth of Argonne's Materials Science Division.¹ The instrument is still going strong, and Fig. 4 shows reflectivity data obtained recently from a film of iron (only four atomic planes thick) deposited on magnesium oxide. The data were obtained by Yi-Yun (Carrie) Huang, who wanted to see the minimum thickness at which iron remains magnetic. Carrie is the latest of a string of young scientists that came to POSY from every corner of the world: Roberto Follat, Paolo Bisanti, Russ Hilleko, and Abdelkader Mounour.

The Sequels

Being the first permanent facility of its kind, POSY has inspired other neutron grazing-angle instruments, for which the time was ripe. Those in Europe are graced by splendid names: in the U.K., CRISP (Rutherford-Appleton Laboratory); in Germany, TORREMA (KFA Institut, Jülich); and in France, D2SIR (Saclay) and EVA (Institut Louis-Langevin, Grenoble). With the exception of SPBAR at the Los Alamos LANSCE facility, that trend is not followed in United States; the beautiful data of

Majkrzak and Soltz at the National Institute of Standards and Technology (and Pnasill at Brookhaven National Laboratory) come out from "Beam Hole #8" or something of that sort.

Magnetism was not the driving force behind the blossoming of reflectometry, but rather the possibility of studying thin films in extreme detail by "coloring" subsequent layers with different isotopes. A close analogy is the growth of the small angle scattering

Fig. 3 Spin-dependent reflectivities of (a) Fe_3O_4 and (b) $\gamma\text{-Fe}_3\text{O}_4$. The lines are reflectivities calculated from (c) best-fit magnetic depth profiles for the two oxides.

Fig. 4 Spin-dependent reflectivities for an 8 Å layer of iron on MgO. The continuous lines are calculated for a magnetic moment of $2.2 \mu_B$ per Fe atom.

technique which took place one decade earlier. In both cases, the scientific community that formed the backbone segment of users of the new technique was that dedicated to polymer physics.

A first wind of these interests was felt at Argonne in the Fall 1987, when a scientist from IBM Research, Tom Russell, came to do a few innocent tests on POSY. At least one of the tests was remarkable. The sample was a silicon plate covered with two layers of polystyrene (PS). The upper layer had the hydrogen of its molecules substituted with its heavy isotope, deuterium, forming d-PS. Also, its linear molecules were much shorter (by a factor of twenty) than the molecules forming the bottom layer. Figure 5a shows the reflectivity for the freshly prepared sample and after heating it to weld the two layers. The results were surprising. After heating, the top layer had become thinner (Fig. 5b), the "short" molecules penetrating the mesh of the long molecules of the bottom layer. However, in spite of its motion, the interface remained rather well defined. These features strongly pointed to a special

Fig. 5 (a) The reflectivity of a bilayer of d-PS/PS on silicon before (dots) and after (squares) heat treatment. (b) The concentration profile obtained by fitting the data in (a). Note that despite the migration of PS into d-PS, the interface remains relatively sharp.

character in the diffusion of polymer chains, as described in the reptation model. Simply put, two layers formed of short chains and long chains interdiffuse, but with different mobilities. The body formed of long, entangled chains behaves as a sponge, soaking in the lighter molecules and expanding; as in the sponge, however, its surface remains well defined.

The social consequences of that experiment were quite serious. Tom declared that he had enough projects to saturate the running time of POSY many times over. A "war council" called to cope with the crisis - Bruce Brown, Kent Crawford, and Bob Kleb - proposed to add a new instrument specifically designed for polymer physics. IBM sent, in record time, a check to cover a substantial fraction of its cost. And thus, at the beginning of 1989, POSY begat POSY II. But the enthusiasm of Tom Russell reached even farther. A young Northwestern University student, Alamgir Karim, fell in love with polymers and decided to watch them interdiffuse for his thesis project. A large number of universities (Cornell, Massachusetts, Northwestern, Illinois, Connecticut, Pennsylvania, Minnesota, Queens College) first made inquiries, then tests (Fig. 6), then more and more ambitious experiments. The academic community - spearheaded by Ed Kramer of Cornell University - became very supportive of the new instrument, pressing the National Science Foundation to support more scientific staff for POSY II. That bid was successful (enter Rick Coyette and Bill Dozler), and the instrument too, to a point that nobody - not even Tom Russell - had expected in the beginning. Even POSY II is oversubscribed. Several times over.

POSY II is now a full-fledged user instrument. Its operation is as

Fig. 6 Reflectivity from a thick, single layer of triblock polymer (dPS-PS-dPS) on silicon. The problem is to observe if, in thermal equilibrium, the deuterated tips are uniformly distributed in the melt. (Jon Sokolov and Miriam Hajilovich, Queens College).

nearly "turnkey" as possible, as is evidenced by its frequent successful use by novices in the technique. The initial design has been improved with automatically varying collimation, and the data reduction software is constantly being improved to increase both precision and ease of use. New enhancements for 1991 include a MicroVAX computer for data acquisition, an automatic sample changer, in situ heating, and a polymer sample preparation lab in Building 300. The user base is also continuing to expand even outside of the polymer physics community, as experiments to study the diffusion and configuration of hydrogen in materials have begun. There is now even talk (or at least whispers) of a POSY III!

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CHAPTER **12**

PULSED SOURCE QUASIELASTIC NEUTRON SCATTERING AT ARGONNE

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The Instrument

The Quasielastic Neutron Scattering Spectrometer (QENS) at IPNS can be traced back through three prototypes to the 1970's when the ZING-P pulsed source was being operated at Argonne. The basic principle used in QENS, that of final energy analysis by Bragg scattering from the 002 reflection of graphite analyzer crystals, has remained unchanged during the development of these instruments.

The first inverted geometry time-of-flight spectrometer at ZING-P was developed by Kurt Sköld (Studsvik, Sweden), Kent Crawford, and Sow-Hsin Chen (Massachusetts Institute of Technology).¹ It had one final energy analyzer crystal array with an associated detector bank which was oriented to measure the scattering at 90° to the incident beam. A beryllium filter between the sample and the graphite analyzer crystals was necessary to remove the scattered neutrons with energies greater than 5 meV, which would give higher-order reflections, thus creating an ambiguity regarding the final energy of the detected neutrons (the 002 reflection of the graphite crystals is used for final energy analysis). The focusing of the analyzer crystals onto the detector bank was optimized using Monte Carlo simulation of the neutron paths.

This first prototype was also operated on ZING-P with the addition of two new copper analyzers placed at $\pm 10^\circ$ (Fig. 1), resulting in a larger final energy (to 40 meV compared with 3.1 meV for graphite). During this period, Torben Brun, currently of Los Alamos National Laboratory (LANL), became instrument scientist of this second prototype. A third prototype was developed and built at IPNS in the early 1980's, and it became operational in 1982 as the Crystal Analyzer Spectrometer (CAS). There was a fundamental difference between the first two prototypes and CAS which lay in

Fig. 1 Configuration of the first inverted geometry spectrometer at ZING-P.

the focusing method for the analyzer crystals. The earlier instruments used Monte Carlo simulation to determine the geometry of the final energy analyzer system, while Torben determined that a cylindrical geometry would allow for energy focusing using an analytical form for the sample-to-analyzer-and-detector geometry. In addition, the detectors are separated during data collection which allows them to be time-focused by the analysis software. This results in a spectrum which is both time- and energy-focused with a resolution function which is narrow in energy and broad in momentum transfer. CAS was used for a detailed study of the low-energy (< 100 meV) modes of the solid hydrocarbons from ethane to hexane (with scientists from Schlumberger-Doll Research).²

It was at this point that a new instrument, QENS, was built based on the three prototypes. QENS was built as a Participating Research Team (PRT) instrument, the PRT members being Argonne, Exxon Research and Development, LANL, and Schlumberger-Doll Research, and it became operational in 1988. It differs from the first prototype in three important ways: the analyzer geometry is based on a cylindrical energy-focusing technique, the beryllium filter is located between the analyzer and detector, and there are three analyzer banks located on a rotating base. The elastic resolution is approximately 70 μ eV, and the accessible momentum transfer range is 0.5-2.5 $\text{\AA}^{-1}$. This makes QENS particularly useful for quasielastic scattering measurements in molecular systems, while

it also functions as a low-energy inelastic spectrometer (up to about 50 meV) with a resolution of approximately 3.5% ΔE .¹

In 1988, the enriched uranium Booster target was installed at IPNS which required some modifications of the shielding and incident collimation of QENS, but left the basic design unchanged. During this period, considerable effort was put into making the operation of QENS and its associated ancillary equipment as routine as possible. The spectrometer has been in a stable configuration since approximately the middle of December 1988, and 25% of the available beam time has been used for non-PRT experiments.

The Science

The early science on QENS made use of the good inelastic energy resolution to continue a study started on CAS to determine the torsional frequencies of the tetramethylammonium (TMA) cation occluded in a series of zeolites. The TMA cation functions as a template during the synthesis of the zeolites, such that it favors the formation of a particular zeolite structure. This trapped template molecule has four methyl groups which interlock around the nitrogen atom, and the rotation of the groups is hindered by this strong interaction between neighboring groups. In addition to the internal barriers to rotation, the methyl groups are also affected by the interaction with the zeolite. The series of experiments on CAS and QENS yielded information on these effects by measuring the frequencies of the rotational oscillations of the groups. The results are shown in Fig. 2 for three different zeolite structures and the TMA bromide salt.² Also indicated are the results of an *ab initio* calculation for an isolated TMA cation. The measured frequencies are always higher than the calculated values, as expected on the basis that the additional interaction with a lattice increases the barrier to rotation.

The first quasielastic scattering experiment was carried out by Ken Bradley on the plastic phase of butane.³ Solid butane undergoes a

structural phase transition at 109 K during which it remains in the $P2_1/c$ space group, but there is a change in shape and volume. Quasielastic scattering was observed, on the timescale of the QENS measurements, but only in the higher temperature phase. The

quasielastic scattering was found to be faster than expected from simulation studies (full width at half-maximum (fwhm) ~ 0.5 meV), and an additional very broad component (15 meV fwhm) was also observed. This latter component is interpreted as rotation about

Fig. 2 Inelastic neutron scattering spectra for tetramethylammonium (TMA) cations in (a) TMA-Br, (b) zeolite ZK-4 (LTA), and (c) zeolite omega (MAZ). The assignments of the torsion and bending modes are shown. The bottom vertical lines indicate the scaled torsion and bending mode frequencies from an *ab initio* calculation for the free ion.

the central C-C bond, and these results have been compared with molecular dynamics (MD) calculations to test the quality of the model system.

The inelastic scattering measurements on template ions in zeolites were extended to higher temperatures where quasielastic scattering experiments were done on the TMA template in the zeolites sodalite and onaga (gmelinite cage).⁶ These differ in their shape and volume, the gmelinite cage being somewhat larger and more elliptical in cross section. The activation energies were determined from the measured values of the rotational diffusion constant (proportional to fwhm) at a series of temperatures. These were found to be essentially the same for both types of cages. In spite of this similarity, the geometry of the TMA motion (as determined from the variation in the residual elastic intensity as a function of momentum transfer) is quite different. The measured elastic incoherent structure factors suggest that there is an about a 30% chance of finding the TMA protons in their original position at infinite time for the sodalite cage, while the probability is approximately 60% for the gmelinite cage. This is consistent with rotation about a C_2 axis for the TMA in sodalite and a C_3 axis for TMA in a gmelinite cage.

The zeolite program is continuing and is now concentrating on the diffusion of adsorbates in molecular sieves for a variety of pore topologies and adsorbates. The preparation of the sorbates, as well as the associated equipment and expertise necessary for such measurements, is now available at IPNS on a routine basis. This has allowed for measurements of the diffusion of methane and cyclopropane adsorbed in Silicalite (the aluminum-poor analogue of zeolite ZSM-5) and the dynamics of water adsorbed in the aluminophosphate molecular sieve APO-5. There is an inherent difficulty in making such measurements on molecular sieves as they are only available as powders. This implies an orientational averaging of the measured motion of the adsorbate, resulting in a compli-

cated line shape for the broad component. In addition, the topology in Silicalite implies a one-dimensional diffusion process over segment lengths of approximately 10 Å.⁷ This has been ignored in previous work as there are no analytical models available for such cases. In an attempt to deal with this problem and to come to a more fundamental description of the interactions between sorbate and the molecular sieve, MD simulations have been carried out for these systems. The results for methane in Silicalite demonstrate the importance of pore topology and orientational averaging on the measured neutron spectra from such systems.⁷ This work is continuing, and it is currently dealing with the detailed comparison of MD simulations and quasielastic neutron scattering.

The scientific program on QENS has broadened to include a variety of different problems, which have come from users who are PRT members and non-PRT members alike. The diffusion of hydrocarbons adsorbed on the basal planes of graphite, with particular emphasis on butane and hexane, has been the subject of work done by Huskell Tubb (University of Missouri - Columbia) and collaborators. Previous diffraction measurements and MD simulations for these two molecules adsorbed on graphite have indicated that these molecules behave quite differently when adsorbed on graphite. In particular, the experiments have focussed on the melting of the adsorbed monolayers which is readily probed using quasielastic scattering measurements on QENS. The results demonstrate that, in the case of butane, rotational order about the surface normal and translational order are lost abruptly at the melting point. Hexane behaves differently, the results being consistent with the coexistence of solidlike patches having a herringbone structure and a fluid phase of molecules in a gauche conformation above the melting point of the monolayer. These results are consistent with the diffraction experiments and MD simulations.

In a closely related area, considerable effort has been

devoted to the general area of hydrogen bonded systems, extending the original work done at IPNS on formic acid.⁸ The system chosen was ethanol on graphite which is a logical extension of the large amount of work done on adsorbed hydrocarbons over the last twenty years. QENS provided the inelastic spectrum at low temperatures, in addition to the quasielastic scattering observed above 150 K. The inelastic spectrum is shown in Fig. 3. The first two peaks are ascribed to the librations of the molecule about its long axis and the short axis perpendicular to the graphite surface, while the highest energy excitation shown is the libration of the CH₃ group. The quasielastic scattering at 200 K is interpreted as rotational motion of the molecule, while the scattering at 300 K is clearly due to translational motion. Figure 4 shows the variation of the quasielastic scattering intensity as a function of momentum transfer for these two temperatures; they are clearly different for the two temperatures and characteristic of rotational and translational motion at 200 and 300 K, respectively. This is in complete agreement with diffraction measurements carried out on the High Intensity Powder Diffractometer (HIPD) at IPNS. The influence of hydrogen bonding between the molecules results in a melting point (> 210 K) which is approximately 50 K above that for bulk ethanol.

Some More Thoughts

QENS has evolved from the early inverse geometry inelastic scattering spectrometers into an instrument capable of rotational and translational diffusion measurements for a wide variety of molecular systems. In addition, it has proven to be very useful as a low-energy (< 40 meV) inelastic spectrometer for a wide variety of systems ranging from magnetic excitations⁹ to the torsional motion of trapped molecular ions in zeolites.

The combination of good elastic resolution and useful momentum transfer range with simultaneous measurement of the inelastic spectrum out to 40 meV

Fig. 3 Inelastic spectrum taken at 4 K on QENS for ethanol adsorbed on the basal planes of the partially oriented graphite, Grafoil.

Fig. 4 Variation of the intensity of the broad component of the quasielastic scattering from ethanol on graphite at 200 and 300 K.

makes QENS very useful for the investigation of diffusion and low-frequency excitations. Since the incident beam is not monochromated and an additional detector bank without an analyzer functions as a time-of-flight diffractometer, it is also possible to measure the diffraction pattern from the sample simultaneously. This makes QENS a unique instrument for quasielastic neutron scattering measurements.

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CHAPTER 13

THE CHOPPER SPECTROMETERS AT IPNS: AN UNAUTHORIZED STORY

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A Frigid Start

About ten winters ago, Chicago was buried under snow. (Even the mayor of Chicago claimed to be victimized by the ponderous snow.) A team of scientists, engineers, and technicians were warming up for experiments at the first spallation neutron source ever in the United States. On the deck of a spectrometer, which one might recognize as the Thermal Neutron Time-of-Flight Spectrometer (TNTOFS) at the CP-5 reactor but now was rechristened as the Low Resolution Medium Energy Chopper Spectrometer (LRMECS - what an unattractive name!), was Jack Carpenter. Yes, the IPNS technical director also wore the hat as the LRMECS instrument scientist. Imagine the wonders this would bring to the spectrometer? The next year, LRMECS got a new evacuated collimator housing. A new thin-wall aluminum "HAT" for maintaining a sample cryogenic vacuum was designed and fabricated. Its two chemically milled-down 0.005-in-thick windows would help minimize background scattering by the entrance and exit beam. To test if it would sustain an internal ambient pressure, Jack and Chun Loong (then a newcomer to IPNS) filled the HAT with water and pressurized it with a bicycle pump. As expected, it passed the test. (Ira Bresof, the IPNS Quality Assurance Representative, will verify the faultless performance of the LRMECS HAT for the last ten years.) Although LRMECS resembles the old TNTOFS, it was given a new heart, that is, a new chopper. The new chopper accommodates an aperture size up to 3.5 X 4.5 in² and weighs close to 50 pounds. Different energy ranges could be accessed by changing chopper rotors. Running and synchronizing choppers at a speed of 270 revolution per second were no simple matters. If you have doubt, just ask George Ostrowski about replacing the failed bearings in those sleepless nights during the

first two years of operation. Fortunately, a magic lubricant, capable of lengthening the bearings lifetime by more than a factor of ten, was soon found.

Down at the other end of the IPNS experiment hall, a brand new chopper spectrometer, the High Resolution Medium Energy Chopper Spectrometer (HRMECS), was being commissioned. Although it uses the same chopper rotors, this big brother is more than twice the size of LRMECS (also, a resolution two times better).

Noise, Shielding, and More Shielding

Neutron scattering scientists are obsessed with suppression of unwelcome stray neutrons (noise) by various kinds of neutron absorbers called shielding. Soon after HRMECS started to collect data, David Price noticed that detectors closer to the back wall of the scattering chamber had a higher noise level. Jack Carpenter and David quickly discovered that the noise came from high-energy neutrons, which pulsed sources take pride in producing, bouncing within the scattering chamber (the so-called albedo effect). After testing about a dozen different materials, Jack pronounced boron carbide (B₄C) to be the best of low-albedo neutron absorbers. He showed the IPNS crew a recipe (which he perfected in Japan) of bonding granulated B₄C with the least amount of epoxy and molding the mixture into large plates. Mass production of this gooella, which was later called "crispy mix", soon began. Hundreds of square meters of crispy mix were installed on the inside walls, ceilings, and floors of LRMECS and HRMECS scattering chambers. The LRMECS 120°, 7-ft-radius, pin-shaped chamber has a clearance of less than four feet; Dave Loebe's acrobatic gifts and courageous enthusiasm made this shielding job possible. The improvement in the signal-to-noise

ratio was stunning. Over the years, tons of crispy mix, "flexi mix", "caramel corn" (a neutron concrete), and black lead-shot bricks were produced at IPNS. But the demands of shielding show no end of it. Let's face it, these would not exactly be considered health foods of the 20th century! Rumor has it that the technology of shielding fabrication has now been transferred to an off-site specialty company (or the shielding builders themselves all transferred to other divisions at Argonne).

The Puzzling Resolution Function

The neutron time-of-flight (TOF) technique is, in simple terms, a matter of measuring neutron flight times and distances. In the beginning, the chopper team was alerted by a slight discrepancy in the positions between the observed and calculated elastic peaks. They checked their clocks again and remeasured the distances, but they found nothing wrong in their timers and rulers. Interestingly, Roger Sinclair of A.E.R.E. - Harwell, U.K. reported a similar problem with the chopper spectrometer at Harwell's electron-accelerator-based pulsed neutron source. Things were pointing to the need for an in-depth understanding of the response of a chopper spectrometer to the neutron bursts emitted from the moderator. The opportune moment arrived in 1983 when Susumu Ikeda of the KENS pulsed neutron source at the KEK High Energy Physics Laboratory, Japan visited IPNS for a year. He and Jack Carpenter built a time-focused crystal spectrometer on the beam line of the Single Crystal Diffractometer (SCDD) and succeeded in measuring the neutron pulse shapes over a wide range of energies. The analytical expressions of these measured pulse shapes are now called the Ikeda-Carpenter function. Over the Christmas holidays of that year, Susumu formulated the resolution function for a pulsed source chopper

spectrometer. Results of subsequent studies revealed the solution for the energy calibration problem. The neutron burst of most energy has an asymmetric distribution (pulse shape): It contains more higher speed neutrons, which arrive at shorter times for a given distance from the source than do the slower neutrons in the pulse. A synchronized chopper, acting as a pinhole camera, admits the neutron burst, but reverses its asymmetric 'TOP' profile. As the neutrons travel longer distances, the profile becomes more skewed. A simple analysis of the monitor profile centroids would lead to a slight error in energy calibration.

More Neutrons, More Detectors, More Computers...

Neutron scattering scientists always hunger for intensity. To catch more neutrons, they want more detectors. To cope with the large data sets, they want faster computers and better software. In 1985, a high-angle (85-140°) flight path chamber was added to HRMECS to relieve the demands for high-Q and high-resolution experiments. Efforts are underway to install an intermediate-angle (20-85°) chamber in 1992. The use of pseudo-time in data acquisition was implemented by Kent Crawford in 1985, which enabled the setup of an energy scale for detectors of different sizes and flight distances. Kent also installed new software and hardware for the use of a MicroVAX workstation as the front-end computer on HRMECS. Data reduction for chopper spectrometers usually involves multiple step processes using many analysis programs. The pioneering analysis codes were written mainly by David Peisen. Figure 1 illustrates a portion of the analysis schemes undertaken by Masah Arai of the KENS facility and David for data from glass samples in 1983. As it can be seen,

only very friendly and sympathetic users (or persistent students of forceful professors) managed to uncover the information from the data. In 1984, a process of streamlining the analysis codes took place. An improved version of the data analysis package was presented by Jack Carpenter at the "Workshop on Research Opportunities in Amorphous Solids with Pulsed Neutron Sources" at Argonne in 1985. Currently, more user-friendly software in the directions of on-line help, menu-driven input, and dynamic visualization is being developed by George Ostrowski and John Hammonds.

The Chopper System Command Center

The dynamic range and the resolving power of a pulsed source chopper spectrometer depend on the parameters of the heart of the instrument, namely, the chopper (and controller). Six high-resolution (E_c) choppers are fully operational. Switching neutron energies anywhere from 4 to 2000 meV takes only 1/2 to 4 hours. Choppers have also been developed and built for the suppression of delayed neutrons (these had actors became about six times more populous with the installation of the IPNS Booster target) and the elimination of the prompt neutron pulse (to reduce fast neutron and gamma-ray contributions to background). Two prompt-pulse removal choppers ("stoppers") for the Spacial Environment and General Purpose Powder Diffractometers (SEPD and GPPD, respectively) and three delayed-neutron choppers (" E_c choppers"), designed by Bob Klob, Paul Sokol, and Jack Carpenter, for the three chopper instruments HRMECS, LRMECS, and PHOENIX are now running in addition to the E_c choppers; all of these choppers are synchronized to the accelerator. The good old

"simple" chopper controller system has evolved to a sophisticated chopper system command center, integrated with columns of microcomputers and electronic hardware. What is more, rumor has it all other instruments want a dedicated chopper as well. This 10-year evolution has been highlighted by several major improvements:

- The development of a phase-locked variable-frequency mode in 1984 by Lawrence Donley and George Ostrowski made possible a more stable and efficient operation of the accelerator and chopper systems.
- The implementation of the High-Resolution Chopper Controllers in 1985 by Edwin Jung of Argonne's Electronics Division and subsequent conversion to personal-computer-based units, resulted in cost reduction and flexibility enhancement.
- The expansion to multiple-harmonic rotational frequencies for both the E_c and E_d choppers in 1989 expanded the operation over the entire range of cold-to-epithermal neutron energies with optimal efficiency.

Actually, the real highlight was the 1986 party celebrating the receiving of the first Argonne Laboratory Director's award by Lawrence and George.

And, Finally!

During ten years of operation, the chopper team enjoyed the interactions with users and the support from scientific, engineering, and administrative branches of Argonne. They look forward to another productive decade to come. Meanwhile, the story teller is at peace, at least for now, free from hassle by the Editor for writing another chopper story. [Editor's Note: Until the next IPNS Progress Report!]

Fig. 1 An historic lab notebook page of chopper spectrometer data analysis by David Price and Masu Arai in 1983

7 MAY AND NEUTRON CAPTURE FROM MATERIAL
IN THE LABORATORY, 1964

CHAPTER 14

INELASTIC MAGNETIC SCATTERING USING COLD-TO-EPI THERMAL NEUTRONS

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Introduction

Magnetic neutron spectroscopy, which is based on the magnetic interaction between a neutron and unpaired d- or f-electrons, provides unique information on the electronic configuration and dynamics in solids. The energy range of interest varies from 10^{-1} to 10^4 meV. Traditionally, neutron scattering of magnetic materials is limited to the low-energy region (less than 50 meV) at reactor sources. The advent of pulsed neutron sources has extended the study of magnetic excitations to hundreds of meV, thereby expanding the domain of scientific problems and materials to be studied.

Crystal-Field Excitations in f-Electron Dioxides

High-energy magnetic scattering was recognized as an important area of research ever since the High Resolution and Low Resolution Medium Energy Chopper Spectrometers (HRMECS and LRMECS, respectively) began operation at IPNS. When HRMECS was commissioned in August 1982, Sandy Korn (Colorado State University) spent his first summer visit (and eight more summer visits and a nine-month sabbatical during the next nine years) at Argonne National Laboratory (ANL). He proposed to investigate the crystal-field splitting in PrO_2 , one of his favorite magnetic oxides. Almost twenty years ago, Korn measured the magnetic susceptibility of PrO_2 from which he deduced a Γ_6 ground state. However, the position of the next excited state, Γ_7 , was not determined since the energy is too high for thermal neutron scattering and the opaqueness of this compound precludes any optical study. As the first experiment on HRMECS - with effectively six times lower neutron flux and having half the number of detectors as compared with the current configuration - a magnetic excitation between the Γ_6 and Γ_7

states was clearly observed at 130 meV (Fig. 1a). A subsequent experiment⁴ on a similar compound, BaPrO_3 , brought a new record in energy transfer, a 280 meV crystal-field excitation (Fig. 1b).

Having tasted the first fruits of success, Korn, Gerry Lander, and co-workers quickly contemplated the next harvest. They measured crystal-field transitions in several actinide oxides¹⁻³ (BaUO_3 , UO_2 , NpO_2 , and PuO_2), chloride and fluoride⁵ (UCl_3 and UF_3), and related materials. The electronic spectra of these 5f-systems turned out to be more complex. The results showed a variety of structures: sharp, but densely spaced excitations (UO_2); weak and broad features (NpO_2 and PuO_2); and null outcome (BaUO_3). But they enjoyed the fun of the experiments, such as the Olympian task of fastening the mounting screws of a radioactive PuO_2 sample. (N.B. The sample was enclosed in a doubly-sealed container, and the experimenters were surrounded by health physicists and their equipment.) Some of these IPNS measurements have consequently been redone at the ISIS facility at the Rutherford-Appleton Laboratory, U.K. under improved conditions.

Spin-Waves in Single-Crystal Itinerant Magnets

Quantitative measurement of the dynamic susceptibility, $\chi(Q, E)$, of a many-electron system over a wide range of momentum (Q) and energy transfer (E) has been one of the most important objectives in magnetic neutron scattering at pulsed sources. Magnetic excitations in itinerant metals (e.g., Fe, Ni, and Cr) at energy transfers above 100 meV, in which important issues such as the existence of optic branches of spin-waves dispersion and the transition into the Stoner excitation regime, had not been explored by neutron spectroscopy. In 1983, Jeff Lynn (University of

Maryland) and co-workers carried out the first single-crystal experiment on a pulsed source chopper spectrometer. This experiment⁶ extended Lynn's 1975 measurement of the spin-wave spectrum of ferromagnetic iron to a higher energy (150 meV) for the first time. Similar experiments on Cr and Cr-8at% Fe alloy (Sami Werner of the University of Missouri - Columbia, and K. Mikkola of the Institute of Nuclear Research, Swierk, Poland and co-workers) met with limited success. Even though IPNS has demonstrated the feasibility of performing high-energy single-crystal magnetic scattering on chopper spectrometers, the limitation in intensity and Q -resolution was soon recognized. These measurements are continuing at ISIS on both the MARI and NET chopper spectrometers and on special instruments, such as PRISMA, designed for single-crystal work.

Heavy-Electron Systems

Phenomena due to hybridization between the f-electrons and the conduction electrons in certain rare-earth and actinide intermetallic compounds, called heavy-electron systems, have attracted a great deal of experimental investigations, including magnetic neutron scattering. In contrast to the sharp peaks expected for crystal-field transitions, the excitation spectra of heavy-electron materials are, in general, broad and extend to high energies. The $\gamma \leftrightarrow \alpha$ phase transformation in cerium metal is an interesting example. In a previous neutron scattering study⁷ of a similar transformation in $\text{Co}_{0.7}\text{Th}_{0.3}$ that occurs at 150 K, Steve Shapiro (Brookhaven National Laboratory) and co-workers found that the magnetic response function in the low-temperature α -phase broadens to energies beyond the reach of reactor experiments. Extraction of such magnetic components from the

Fig. 1 (a) Neutron intensity as a function of energy transfer for PrO_2 as observed by detectors with scattering angles less than 7° . Upper left inset: The cubic fluorite structure of PrO_2 . The solid and open circles represent, respectively, the Pr and O atoms. Upper right inset: Schematic of the Γ_8 - Γ_7 splitting of the $J = 5/2$ multiplet. The degeneracy of the Γ_8 ground state is lifted by a dynamic Jahn-Teller effect, and, in the ordered state, by an exchange field. The neutron excitation corresponds to the total cross section between all Γ_8 states to both Γ_7 doublet states. (b) The measured scattering function, $S(E)$, of BaPrO_3 , using HRMECS with an incident energy of 800 meV. Inset: The perovskite structure of BaPrO_3 . The open circles represent O atoms, the solid circles, Pr atoms, and the shaded circles, Ba atoms.

total observed spectrum is difficult because detailed knowledge of the nuclear scattering background is required. Nevertheless, under an improved experimental configuration, the team was able to characterize the magnetic excitations of the $\text{Ce}_{0.74}\text{Th}_{0.26}$ alloy up to an energy transfer of 250 meV over the temperature range 100-200 K and about 900 meV at 10 K.¹⁴ The paramagnetic scattering functions for $\text{Ce}_{0.74}\text{Th}_{0.26}$ are shown in Fig. 2.

Another group of researchers, led by Mike Loewenhaupt of KFA, Jülich, Germany and Ulrich Walter of the University of Cologne, Germany, have studied many heavy-electron actinide compounds (UB_4 , UCu_4 , UGe_4 , USn_4 , UPb_4 , U_2Zn_{17} , and URu_2Si_2) at IPNS and the Institut Laue-Langevin (ILL), France. For example, evidence of a magnetic gap-like excitation in URu_2Si_2 was first seen¹⁵ at IPNS in 1985. They also found that in order to characterize the magnetic response in these materials fully, both high- and low-energy measurements are equally important. In the case of USn_4 , measurements¹⁶ were made on HRMECS and LRMECS at IPNS as well as on the IN4 and IN6 spectrometers at ILL. It took more than two years to schedule and complete the measurements. A question then arises: is a pulsed source chopper spectrometer capable of high-resolution measurements in the 0 to 50 meV energy region in addition to epithermal energies? This indeed has been the main task concerning the IPNS chopper team during 1990 and 1991, and fortunately, the answer to the question is yes. In addition to scattering at epithermal energies, measurements at energy transfers down to about 0.5 meV with a resolution of the order of 150 μeV have been realized recently.

High-Resolution Cold-to-Epithermal Magnetic Scattering

High resolution is essential for measuring the detailed dynamic response of a system, such as the line features in a crystal-field or phonon spectrum. In general, the full width at half-maximum energy resolution, ΔE , of HRMECS varies

between 2-4% of the incident neutron energy (E_0) over the neutron energy-loss region, $E = E_0 - E_1 > 0$ (where E_1 is the scattered neutron energy). Consequently, good resolution at an energy transfer E can be achieved by measurements chosen to maintain a small E_1 . As an example, ΔE is approximately equal to 120 μeV at $E = 0.5$ meV and to 2.5 meV at $E = 80$ meV, if E_0 are 4 and 110 meV, respectively. These experimental conditions are made possible by using the cold-to-epithermal incident neutron spectrum provided by a liquid methane moderator at IPNS in conjunction with a two-rotor energy selector system. Figure 3 shows the excitation spectra¹¹ of a magnetic superconductor $\text{ErBa}_2\text{Cu}_3\text{O}_7$, measured by Lynda Soderholm (ANL), Chun Loong, and Sandy Kern. By using various incident energies between 4 and 110 meV, a total of 13 transitions were observed, corresponding to excitation or de-excitation processes between the crystal-field levels shown in Fig. 4. This experiment demonstrates the versatility of the chopper spectrometers at IPNS, namely, dynamic processes of energies in the 10^{-4} - 10^1 meV range can be investigated in a single experimental setting without disturbing the sample environment.

Conclusion

During the first ten years of the IPNS chopper program, magnetic scattering has been one of the major research areas in inelastic scattering. With steady improvements in the source and the spectrometers, the chopper team is looking forward to another decade of magnetic scattering entering the 21st century.

Fig. 2 Temperature dependence of the observed imaginary part of the magnetic susceptibility for $\text{Ce}_{0.71}\text{Th}_{0.29}$. The solid curves show the results of fitting the data with a spin relaxation spectral function.

Fig. 3 The observed excitation spectra of the magnetic superconductor $\text{ErBa}_2\text{Cu}_3\text{O}_7$. The labeled peaks correspond to excitations or de-excitations between various crystal-field levels, as shown in Fig. 4.

Fig. 4 The crystal-field levels for the ground state term $4I_{15/2}$ of Er^{3+} in $\text{ErBa}_2\text{Cu}_3\text{O}_7$. The labeled transitions correspond to the observed peaks shown in Fig. 3. The crystal-field parameters are obtained by modeling the data with an intermediate coupling scheme calculated by spherical tensor techniques.

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CHAPTER **15**

MOMENTUM DISTRIBUTION MEASUREMENTS AT IPNS

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Introduction

The advent of spallation neutron sources, in particular IPNS, provided a window into a region of energy-momentum space that was not easily accessible using reactor-based neutron sources. One area that benefited immensely when this window opened was the measurement of atomic momentum distributions, $n(\mathbf{p})$, using Deep Inelastic Neutron Scattering (DINS).¹ These measurements require extremely energetic neutrons which IPNS produces copiously. Therefore, while momentum distribution measurements were attempted at reactors in the past, it was not until the advent of spallation sources that DINS was fully exploited.

Early DINS Measurements at IPNS

One of the most exciting areas, and perhaps the main driving force behind the development of this technique, is the study of quantum liquids and solids, such as the heliums. These systems are characterized by light mass and weak interactions leading to a large quantum zero-point motion and a correspondingly large overlap of the wave functions describing the atoms. Quantum mechanical effects are the dominant features determining the behavior of these systems, and many new and unusual properties are observed. For example, neither ^4He , nor its less common isotope ^3He , solidify, even at a temperature of 0 K, unless a substantial external pressure is applied. Even more surprisingly, both of these systems exhibit superfluid phases, albeit with very different transition temperatures. The superfluid phase appears in ^4He at ~ 2 K, while it does not appear in ^3He until ~ 1 mK, nearly two thousand times colder. These effects are intimately linked to the extreme quantum natures of these liquids and the different statistics (Bose-Einstein for ^4He ,

Fermi-Dirac for ^3He) that govern their collective behavior.

The atomic momentum distribution, as measured using DINS, provides a unique perspective on the microscopic dynamics in these quantum systems. For example, in 1938 Fritz London proposed² that the superfluid phase of liquid ^4He was the analogue, for a real system, of the phenomenon of Bose-Einstein condensation, where a macroscopic occupation of a single quantum state develops. This phenomenon manifests itself through the appearance of a delta function singularity in the momentum distribution and is responsible for the "super" properties of liquid helium. However, this very simple and fundamental idea has taken over 50 years to verify since the condensate only directly shows itself in the atomic momentum distribution.

DINS measurements of the momentum distribution, therefore, allow a direct verification of London's intriguing idea. The experimental quest to observe the condensate in the superfluid phase directly began with reactor measurements³ starting in the mid-1960's. However, due to the limited momentum transfers available at reactors, the Bose condensate remained elusive. In fact, the quest has only recently been brought to a successful conclusion with measurements carried out at IPNS.

Thus, it was not surprising that the appearance of IPNS in 1981, with its plentiful supply of energetic neutrons needed for DINS, attracted the attention of experimentalists interested in quantum liquids and solids. In particular, Ralph Simmons and his group at the University of Illinois - Urbana (UI), which have had a long standing interest in quantum solids, were intrigued by the potential of this new source and carried out the first measurements of $n(\mathbf{p})$ in solid helium at IPNS in 1982.

Simmons, with his student Russ Hilleke and postdoctoral fellow Praveen Chaddah, and David Price and Sunny Sinha (currently at Exxon Research and Development) from Argonne measured the momentum distribution of solid helium using the Low Resolution Medium Energy Chopper Spectrometer (LRMECS) at IPNS. Figure 1 shows the observed scattering as reported in the first paper on momentum distributions from IPNS.⁴ The scattering clearly satisfies the minimum requirements for DINS: a symmetric peak centered at the recoil energy. At the same time, the relative resolution, as a fraction of the peak width, was comparable to reactor-based measurements that were carried out at much lower values of Q where deviations from the prediction for DINS were quite apparent. Thus, Simmons and collaborators were able to obtain accurate values for the average kinetic energy per atom, $\langle KE \rangle$, which provides information on the importance of quantum effects.

To put the capabilities of IPNS into context, it is useful to examine the range of momentum transfers covered in this first experiment. While reactor measurements have been carried out at Q values comparable to those used at spallation sources, this has only been at the expense of degrading the resolution so severely that little detailed information can be obtained. In contrast, the chopper spectrometers at IPNS - LRMECS and the High Resolution Medium Energy Chopper Spectrometer (HRMECS) - have relatively good resolution over the entire range of energy and momentum transfers used. Figure 2 shows the location of the peak as a function of momentum transfer. The small shaded area near the origin is the region where measurements of comparable resolution have been carried out

Fig. 1 The dynamic structure factor for solid helium ($19.45 \text{ cm}^3\text{-mol}^{-1}$) at a scattering angle of 105.9° .

Fig. 2 The recoil energy as a function of momentum transfer in solid helium. The small shaded area near the origin is the region covered in reactor-based measurements.

using reactors. The advantages of IPNS are clear.

This measurement marked the beginning of a highly successful series of momentum distribution measurements at IPNS that is still going strong. Paul Sokol, then a postdoctoral fellow at UI, Simmons, and Price began a series of measurements using LRMECS that now cover a large portion of the liquid and solid phases of ^4He . These measurements include studies of the density dependence of the condensate fraction⁵ (it goes down as expected, but does not go to zero at the liquid-solid transition), the difference between the hcp and bcc solid phases and the liquid at the same density⁶ (there is none!), and the Fermi surface⁷ of ^3He (with Kurt Sköld from Studsvik, Sweden). These experiments were very successful in elucidating the properties of quantum systems in the various solid and liquid phases and confirmed the unique ability of IPNS for DINS measurements. By late 1985, the intensity of the effort to study momentum distributions by Sokol, by then at Harvard University, and Simmons was threatening to dominate the scientific program on LRMECS and was beginning to spill over onto HRMECS as well.

A Dedicated IPNS Instrument for DINS

The initial series of measurements in the first half of the 1980's provided a great deal of new information on the quantum behavior of the heliumons. However, the measurements were beginning to press the capabilities of the existing instruments to the edge in terms of the resolution and statistical accuracy attainable. LRMECS did not have sufficient resolution for the more precise studies of the line shape that were now needed. HRMECS offered much better resolution but had only a small complement of the high-angle detectors required for DINS measurements. More importantly, the measurements were beginning to reach the point where detailed comparisons to theory were possible. However, a major limitation in carrying out these

comparisons was the statistical accuracy of the results. There was simply not enough time available, on an instrument in the user program, to carry out the measurements of high statistical accuracy required for a detailed comparison with the increasingly sophisticated many-body calculations of liquid and solid helium.

These studies also faced another major problem, this one related to the user facility status of IPNS. Cryostats, gas handling systems, and associated equipment were continuously being moved onto the instrument to prepare for a run or off the instrument to make way for the next user. This never-ending game of "musical cryostats" only intensified the standard low temperature problems such as leaky cryostats, plugged fill lines, and the occasional exploding sample cell.

In light of these problems, the decommissioning of the Ultra-Low Temperature Diffractometer was viewed with great interest. (A good experimentalist has a great deal in common with one of the less popular members of the animal world, the vulture!) While no one actually circled the dying instrument, it did not take long for Price and Sokol to lay out the plans for a dedicated instrument for DINS measurements on the back of a pickup at Slone's Ice Cream in Harvard Square. Thus, like the mythical Greek bird rising from the ashes, PHOENIX was born.

A Participating Research Team was quickly formed to construct and operate PHOENIX. This team was headed by Sokol and included Simmons, Price, and graduate and undergraduate students from Harvard and UI. Detailed design of the instrument was begun in January 1986, and the first measurements were being carried out that summer. This swift construction was due to the extraordinary efforts of the students involved in putting the instrument together, particularly Tobin Sosnick, Mike Snow, and Bob Blasdel. These students gladly (well, maybe not so gladly) took on the task of casting thousands of pounds of shielding blocks, endlessly stacking and unstacking lead

blocks, and many other intellectually stimulating tasks.

The design and philosophy of PHOENIX was aimed towards satisfying two criteria: to obtain the best resolution possible at high Q and to maximize the statistical accuracy of the results. The goal of high resolution was relatively easy to obtain given the vast experience in instrument design at IPNS. The resulting design, which was constrained by the location of the instrument, had a single high-angle scattering bank, to attain the highest momentum transfer possible, and the longest flight paths obtainable, to attain the best resolution. Maximizing the statistical accuracy proved a much more difficult goal, requiring both high count rates and low background levels. A great deal of effort was expended in this area, including a reevaluation of the chopper shielding, the development of a " 1_ϕ chopper" to remove background contributions from the prompt gamma-ray pulse, the development of special focusing schemes to increase count rates, and the development of sophisticated Monte Carlo codes to describe the instrumental resolution.

DINS Measurements Using PHOENIX

Figure 3 shows the fruits of this labor.⁸ The upper figure is a measurement of the normal liquid at 3.5 K, where no condensate is present. The lower figure is a measurement of the superfluid at 0.32 K, where a condensate should exist. The difference between the scattering in the two phases is quite dramatic. Below the superfluid transition the scattering is much more peaked near $Y = 0$, where the condensate contributes to the scattering. Alas, no distinct condensate peak (the "Holy Grail" in this quest) was observed. In fact, if one attempted to compare the predictions for $n(p)$ from the sophisticated many-body theories⁹ available (shown as the dashed line in the figure), the agreement was quite poor. A skeptical observer might even question the existence of a condensate.

The failure to observe a distinct condensate peak was not a surprise.

It was well known that deviations from the impulse approximation would distort the scattering and smear out the condensate peak. However, the available theories for these deviations lacked sufficient predictive power to allow a direct comparison with the theoretical calculation of $n(p)$. Fortunately, at about the same time that PHOENIX began operation, Richard Silver of Los Alamos National Laboratory developed a theory describing the deviations from the impulse approximation in detail.¹⁰ Silver's treatment was special for two reasons. First, it gave definitive predictions on how final state effects modified the observed scattering. Second, it contained no adjustable parameters. The only inputs to the theory were the pair distribution function and the interatomic potential, both experimentally measured quantities.

The solid lines in Fig. 3 show the many-body predictions corrected for both final state effects, using Silver's theory, and instrumental resolution. The agreement, in both the normal and superfluid phases, is excellent! For the first time, good agreement between theory and experiment for the momentum distribution in superfluid helium was obtained. This agreement provides convincing evidence for the existence of a condensate containing 10% of the atoms and brings the long quest to confirm London's idea to a close.

Since these first measurements showed such beautiful agreement between experiment and theory, the phase diagram of ^4He has been extensively explored by Sokol, Simmons, and their students. While many open questions remain, the picture of the microscopic dynamics that has emerged is very pretty indeed. We can expect to see similar progress on such problems as the momentum distribution in ^3He , ^4He - ^3He mixtures, and many other systems in the future.

While the first studies at IPNS concentrated on helium, attention quickly expanded to include other lines of investigation. These included systems which have internal degrees of freedom (such as condensed molecular hydrogen), heavier

Fig. 3 The Compton profile for (a) normal liquid helium at 3.5 K and (b) superfluid helium at 0.35 K as a function of the scaling variable $Y [= (M/Q)(\omega - \omega_r)]$, where $\omega_r [= Q^2/2M]$ is the recoil energy. The dashed lines are the theoretical predictions with instrumental resolution taken into account. The solid lines include final state effects in the comparison.

systems (the noble gas solids and fluids), and combinations (such as dilute hydrogen embedded in bulk hosts or intercalates and hydrogen adsorbed on surfaces). All of these proved fruitful.

Condensed hydrogen is a fascinating prototype system for neutron study. Calculations of its neutron scattering properties date from the earliest days of neutron research in the 1930's, when the focus of attention was understanding the interaction of the newly discovered neutron with nuclei, particularly, the proton. But only with the advent of IPNS and its chopper spectrometer LRMECS was sufficient resolution available to look at hydrogen in detail.¹¹ It was demonstrated that the DNS technique indeed probed the initial state of the target system, although the final states included a variety of excitations, including rotation and vibration (the latter, for incident neutron energies chosen above the 517 meV threshold). Figure 4, the first measurements of hydrogen at IPNS, clearly shows the different final states excited in the measurement.

Predicted details fall into place, such as the relative strengths of the internal hydrogen transitions, which depend upon energy and momentum transfer, and such as the freedom shown by these molecules to rotate in the liquid and solid states, so that they act as independent scatterers, to satisfy the requirements of the impulse approximation. The first quantitative measurements of the solid and liquid ground-state single-particle kinetic energies were made.¹² Subsequent work on para-hydrogen extended the results over a wide range of solid densities.¹³ Such volume-dependent studies can be carried out over very wide density ranges because solid hydrogen is a "quantum solid" which, like the heliums, owes its unusually large interparticle spacing at low density to quantum zero-point pressure. Thus, hydrogen has an extraordinarily large compressibility. The results obtained in these measurements show gratifying agreement with Monte Carlo simulations.¹⁴

The technique has also been applied to study guest-host interactions and is still developing. Early results have been obtained on the system para-hydrogen in solid argon.¹⁴ They disagree with standard spectroscopic results in the literature which were obtained by IR absorption and Raman methods. The optical spectroscopies were primarily aimed at internal molecular effects, but it seems that the center-of-mass inferences from them are off by up to 50%, so reexamination of the optical spectroscopies, and the associated theoretical simulations, is in order. The technique has also been employed to obtain information on the local environment of hydrogen in intercalates¹⁵ and on surfaces.¹⁶

Another line of investigation pursued heavier noble gases. The elements in this column of the periodic table form a graduated family, which ranges from xenon (thoroughly classical) to neon (a bridge to the quantum solids). This family also provides, in the other direction, a gradually increasing amount of triplet and higher-order interactions and of crystal anharmonicity, which indeed are not yet known quantitatively in the condensed state. First results of these studies have recently been published.¹⁷ They can be summarized succinctly: Boltzmann was right! That is, systems under classical conditions, no matter how condensed they are or the nature of their interactions, show single-particle kinetic energies which follow the equipartition law, $(3/2)kT$. Figure 5 shows this, using reduced coordinates, for the whole family; points for solid neon, argon, krypton, and xenon are shown. The results demonstrate the applicability of this neutron recoil method to determine $\langle KE \rangle$ even of heavy atoms in suitable cases where a value may not be known in advance (e.g., inclusions in other hosts).

Liquid neon has been studied at reactor neutron scattering facilities and was the subject of considerable attention concerning final state effects.¹⁸ Results from IPNS show this prototype liquid can still yield surprises, because the earlier work at reactor sources had

Fig. 4 The scattering from liquid hydrogen showing the various excitations in the final state.

Fig. 5 Single particle kinetic energy as a function of temperature, both in reduced coordinates, for several rare gas solids.

required considerable correction of the observed recoil line shapes toward supposed impulse approximation conditions. Recent results¹⁰ show disagreement in detail with the earlier work, apparently owing to the presence of even-order corrections to the recoil line shape. No doubt, ongoing DINS measurements will provoke additional theoretical consideration of final state and related effects, as the value of this neutron method becomes more widely recognized.

Conclusion

The last ten years have seen great advances in the "state-of-the-art" for DINS. These technological advances have been accompanied by scientific advances both in the solution of long standing questions, such as the superfluid phase of helium, and in new areas, such as the studies of hydrogen in various environments. IPNS, both as a neutron source and as a scientific institution, deserves most of the credit for these advances.

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FIRST IPNS USERS MEETING November 15 16 1983

SECOND IPNS USERS MEETING April 15 16 1985

THIRD IPNS USERS MEETING December 8 9 1986

FOURTH IPNS USERS MEETING November 14 15 1988

CHAPTER 16

THE IPNS USER PROGRAM: THE FIRST TEN YEARS

B. S. Brown and F. J. Rotella, IPNS Division, Argonne National Laboratory

Introduction

From its inception, IPNS has operated as a user facility, and the table shows the growth with time of the number of experiments, users at IPNS, and neutron scattering instruments available for users. These numbers do not, however, tell the whole story. Many of the users not only did one or more experiments at IPNS, but also based a research program at the facility. This took the form of a series of experiments (sometimes 10 or more), a graduate student working full time at IPNS, or an extended assignment or sabbatical at Argonne National Laboratory (ANL). Certainly the collaborations with the Australian National University, Colorado State University, the University of Illinois - Urbana, the Massachusetts Institute of Technology, the Pennsylvania State University, the Transuranium Institute (Germany), and Willamette University stand out as some of our most successful long term collaborations.

The Argonne Fellows program for distinguished visitors to the Laboratory was established in 1982. Of the eleven Fellows who have participated in the program, IPNS has been extremely fortunate to have hosted two. Professor John White from the Australian National University was the first, and he spent a very fruitful 12 months (spread over 4 years) involved in many experimental programs at IPNS and with the catalytic chemistry group. The second Fellow to be hosted by IPNS was Eric Lynn (Director, Nuclear Physics, A.E.R.E. - Harwell); his appointment spanned one year from April 1988 through March 1989. Recently, another Argonne Fellow, Professor John Endersby from the University of Bristol, U.K., spent one year in the Chemical Technology Division involved in research on the structure of liquids using neutron diffraction techniques at IPNS.

Financial Support

IPNS has a policy of paying some or all of the travel and living expenses for university scientists or students working on accepted experimental proposals. This and the availability of a user car is made possible through funds from the University of Chicago. IPNS also receives support from ANL's Division of Educational Programs (DEP) for students and visiting professors.

For the past 3 years, operation of the IPNS has been supported by the National Science Foundation (NSF) Science and Technology Center for Superconductivity. Time is available on IPNS instruments to members of the center and to scientists supported by the NSF for research in superconductivity. This has been particularly useful for performing experiments quickly in this rapidly developing field since a streamlined proposal system is used for this beam time. The NSF has also provided support for a scientist

at IPNS whose major effort will be to develop a program of polymer science research centered currently on the unpolarized neutron reflectometer POSY II.

During the ten years of IPNS operation, the Department of Energy (DOE) has provided support for many of the scientific initiatives promoted at the facility. Conceptual plans for the enriched uranium Booster target, the Glass, Liquid, and Amorphous Material Diffractometer (GLAD) and the polarized neutron reflectometer POSY were made with partial support from ANL discretionary funds, while the development of GLAD and the second small angle diffractometer SAND were and are being funded through grants from the DOE.

Beam Time

Another useful barometer of user interest in IPNS is the number of days requested under our proposal system. Figure 1 shows these

Fig. 1 IPNS statistics since 1982.

TEN-YEAR IPNS USER PROGRAM STATISTICS

	FY82	FY83	FY84	FY85	FY86	FY87	FY88	FY89	FY90	FY91
Number of Experiments Performed	94	110	210	180	212	223	257	323	330	273
Visitors to IPNS for at Least One Experiment:										
Argonne	37	41	49	44	52	55	57	60	61	60
Other Government Laboratories	8	9	8	7	11	15	18	16	19	15
Universities	27	33	45	51	79	70	80	94	120	92
Industry	5	5	9	7	13	24	20	24	36	18
Foreign	12	18	39	35	27	24	17	26	18	27
	89	106	150	144	182	196	201	220	254	212
Number of User Instruments	4	5	6	6	6	6	6	7	7	7
Number of PRT Instruments	1	1	1	2	3	3	4	4	5	5

FY indicates Fiscal Year (e.g., FY91 = October 1, 1990 to September 30, 1991, inclusive).

PRT indicates Participating Research Team.

data, as well as the number of days available on the user instruments. The oversubscription on the individual instruments was generally between 2 and 3 in the early years. This ratio decreased somewhat after installation of the Booster target in 1988, when beam line flux increased by a factor of 2.5.

Although a large fraction of the beam time is allocated based on the proposal system, 25% of the time on the user instruments is retained for research and instrument development by the Argonne scientist responsible for that instrument. In addition, proprietary time is available at a full cost recovery rate of \$4550 per 24 hour day, including technical assistance and use of data analysis capabilities. Approximately 20 days per year are purchased by scientists from industry and other DOE laboratories. As part of an industry-national laboratory program, the Argonne Superconductivity Pilot Center has purchased beam time for collaborative research during the past 3 years.

IPNS Advisory Committee

The IPNS Advisory Committee was established to assist users with any problems or issues arising at IPNS where consultation could be useful. The purview of the committee has expanded to include advising on short- and long-term plans for the facility. The members were chosen to represent the various scientific research areas that utilize IPNS. Listed below are scientists who have served on the committee during the first 10 years of IPNS operation; the institution to which the member was affiliated while serving on the committee is also listed. We acknowledge their efforts on behalf of IPNS.

Current Members

J. White (Chair)
Australian National University

J. Ivers
Northwestern University

E. Kramer
Cornell University

H. Fritzsche
University of Chicago

R. Moen
Oak Ridge National Laboratory

D. Price
Argonne National Laboratory

S. Sinha
Exxon Research

P. Sokol
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S. Cooper
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S. MacEwen
Chalk River Nuclear Laboratory,
Canada

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National Institute of Standards and
Technology

R. Simmons
University of Illinois - Urbana

R. Teller
BP-America

C. Wagner
University of California -
Los Angeles

S. Werner
University of Missouri - Columbia

D. Worcester
University of Missouri - Columbia

IPNS/LANSCE Program Advisory Committee

The Program Advisory Committee for IPNS and the Los Alamos Neutron Scattering Center (LANSCE) meets twice annually to review proposals for beam time at the two neutron facilities. Scientists who have served on this committee during the past 10 years, as well as their institutional affiliations at the time of service, are listed below; their time and effort in this capacity is greatly appreciated.

Current Members

T. Egami (Chair)
University of Pennsylvania

R. Briber
National Institute of Standards
and Technology

B. Brown (ex officio)
Argonne National Laboratory

T. Brun
Los Alamos National Laboratory

D. Cox
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University of Minnesota

T. Riewitt
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E. Bradbury
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W. Brown
AT&T Bell Laboratories

H. Chan
University of Illinois - Urbana

S. Chan
Massachusetts Institute
of Technology

J. Cohen
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S. Cropper
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P. Egelstaff
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D. Engelman
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J. Faber
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E. I. du Pont

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G. Jeffrey
University of Pittsburgh

J. Jorgensen
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M. Kirk
Argonne National Laboratory

S. Krimm
University of Michigan

G. Lander
Argonne National Laboratory

B. Larson
Oak Ridge National Laboratory

C. Loong
Argonne National Laboratory

J. McTague
University of California -
Los Angeles

D. McWhan
AT&T Bell Laboratories

H. Mook
Oak Ridge National Laboratory

F. Morse
Los Alamos National Laboratory

S. Moss
University of Houston

L. Passell
Brookhaven National Laboratory

J. Rhyne
National Institute of Standards
and Technology

R. Robinson
Los Alamos National Laboratory

J. Rowe
National Institute of Standards
and Technology

M. Salamon
University of Illinois - Urbana

S. Shapiro
Brookhaven National Laboratory

R. Silver
Los Alamos National Laboratory

S. Sinha
Argonne National Laboratory

C. Stassis
Iowa State University

P. Vergamini
Los Alamos National Laboratory

J. Weertman
Northwestern University

S. Werner
University of Missouri - Columbia

J. Williams
Argonne National Laboratory

W. Yelon
University of Missouri - Columbia

P. Young
Oak Ridge National Laboratory

Conferences and Workshops

Below is a list of conferences and workshops which were sponsored by IPNS during the first 10 years of its existence. Of the 31 conferences and workshops listed, 27 were held on-site at Argonne. Technical and financial assistance from the University of Chicago and the DEP at ANL for these activities is gratefully acknowledged.

July 20-24, 1981

Faculty Institute: The Intense Pulsed Neutron Source

August 12-14, 1981

International Symposium on Neutron Scattering

November 9-12, 1981

International Conference on Neutron Irradiation Effects in Solids

May 20-21, 1982

Workshop on Resonance Neutron Radiography

June 9-11, 1982

Faculty Institute: Introduction to X-ray and Neutron Diffraction in Biology, Chemistry, and Materials Science

June 27-July 2, 1982

Sixth Meeting of the International Collaboration on Advanced Neutron Sources (ICANS-VI)

July 13-16, 1982

Short Course in Powder Diffraction and Rietveld Analysis

June 27-29, 1983

Workshop on Application of Neutron Scattering to Chemistry (joint with the Los Alamos National Laboratory)

November 15-16, 1983

First IPNS User Meeting

November 17-18, 1983

Workshop on Application of Neutron Scattering to Engineering Materials

February 13-15, 1984

Workshop on High-Energy Excitations in Condensed Matter (joint with and held at the Los Alamos National Laboratory)

April 2-4, 1984

Workshop on Structure Refinement from Powder Data (joint with and held at the National Institute of Standards and Technology)

October 23-26, 1985

Workshop on Scientific Opportunities with Advanced Facilities for Neutron Scattering (held at Shelter Island, New York, in collaboration with Brookhaven National Laboratory, Oak Ridge National Laboratory, National Institute of Standards and Technology, and Los Alamos National Laboratory)

April 15-16, 1985

Second IPNS User Meeting

April 17-19, 1985

Workshop on Research Opportunities in Amorphous Solids with Pulsed Neutron Sources

November 15, 1985

Meeting on Neutron Scattering Studies of Biological Macromolecules

May 12-14, 1986

Short Course in Powder Diffraction and Rietveld Analysis

May 15-16, 1986

Conference on Neutron Scattering in Materials Science

December 8-9, 1986

Third IPNS User Meeting

May 12-13, 1987

Design Workshop for an Advanced Chopper Spectrometer (joint with and held at the Los Alamos National Laboratory)

October 26-29, 1987

International Conference on Techniques and Applications of Small Angle Scattering

November 6-7, 1987

Conference on X-ray and Neutron Scattering from Magnetic Materials

October 3-7, 1988

Tenth Meeting of the International Collaboration on Advanced Neutron Sources (ICANS-X; joint with and held at the Los Alamos National Laboratory)

October 24-26, 1988

Workshop on Momentum Distributions

November 14-15, 1988

Fourth IPNS User Meeting

November 16-18, 1988

Short Course in Powder Diffraction and Rietveld Analysis

June 14-15, 1989

Neutron Scattering in Molecular Sieve Research

April 16-17, 1990

Workshop on Glass and Liquid Diffraction

May 22-23, 1990

Workshop on Application of Neutron Diffraction to the Determination of Residual Stress in Engineering Materials

August 23-25, 1990

Workshop on Neutron Reflection Data Analysis

August 13-15, 1991

International Symposium on Frontiers of Liquid and Amorphous States

Users and Visitors:

1981 through 1991

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CHAPTER 17

NEW INITIATIVES, NEW INSTRUMENTS

B. S. Brown and R. K. Crawford, IPNS Division, Argonne National Laboratory

New Initiatives

Pulsed Neutron Research Facility: Accelerator based pulsed neutron sources have been performing neutron scattering research for just over ten years. During this decade, beam intensities have increased by a factor of 100, and there are now more than 50 spectrometers operating worldwide. The pulsed neutron sources have proven to be highly effective and are complementary to reactor-based sources in that there are important scientific areas for which each type of source has unique capabilities. We have proposed to the Department of Energy (DOE) that the capabilities of pulsed neutron sources be further pursued by means of a Fixed-Field Alternating-Gradient (FFAG) synchrotron concept. The program in the proposal will do the conceptual research and development (R&D) for the accelerator and target systems that will produce 100-200 μA at 500-1000 MeV. Much of the present IPNS system will be incorporated in the design. The neutron source, named the Pulsed Neutron Research Facility (PNRF), will be a source comparable to the most powerful pulsed sources now operating in the world and also will act as a test bed for the basic synchrotron concept for more powerful sources in the future. A preliminary conceptual design was developed in 1984 for a 100 μA , 500 MeV machine and will be the starting point for this R&D proposal.

The need for more intense neutron sources has been the subject of many meetings and reports. The most thorough workshop took place at Shelter Island, New York in October 1984. The major findings of the workshop were that:

- The case for a new higher flux neutron source is extremely strong and such a facility will lead to qualitatively new advances in condensed matter science.

- To a large extent, the future needs of the scientific community could be met with either a 5×10^{17} n-cm⁻²-s⁻¹ steady state source or a 1×10^{17} n-cm⁻²-s⁻¹ peak flux spallation source.

The Advanced Neutron Source (ANS) at Oak Ridge National Laboratory will be the most powerful steady state neutron source in the world. The complementary nature of pulsed and steady state neutron sources argues strongly for also developing advanced pulsed source capabilities.

The rationale for more powerful neutron sources must be made on the basis of the scientific impact. It is, in fact, because of the great strength and diversity of the contributions of neutron scattering research in recent times that the need for more powerful sources has become so important.

Frequently, arguments for more intense sources are based on an individual experiment or type of measurement, and it has been shown that a very strong case results when the sum of these arguments is considered. Certain features are common to many of the specific examples:

- The higher neutron flux means that instruments of higher resolution can be constructed, allowing measurements which are now impossible, plus the development of totally new experimental techniques.
- The polarization analysis technique, which has not been fully exploited because of intensity limitations, will become widely used in many disciplines.
- The ability to use smaller samples will result in a great increase in the number and type of materials that can be studied with neutrons.
- The study of nonequilibrium phenomena through real time diffraction will become important.

It is well known that for almost all applications, neutron scattering is an intensity-limited technique. This means that even experiments performed at present sources would benefit directly from more flux. More importantly, past experience has shown that increases in intensity open new scientific areas, and this will undoubtedly occur with the next generation source. The neutron cross section is small so that rather large samples have to be used. Many materials of interest are available in only very small quantities, or the scattering centers of interest are dispersed dilutely.

The original goal of accelerator-based neutron sources was to prove the viability of this option for the next generation neutron source. The scientific successes of spallation neutron sources over the past few years have verified that this goal has been met. It is, however, important to note that all of the pulsed spallation sources in the world today are based on accelerators that were built for other purposes. The rapid progress in accelerator technology and the proven successes of spallation sources indicate that the field has matured and that it is time for a pulsed neutron source to be built that is dedicated to and maximized for neutron scattering science.

Argonne National Laboratory (ANL) pioneered the use of proton accelerators to produce instantaneously intense bursts of neutrons and time-structured beams that make effective use of time-of-flight measurement methods. The first spallation source of this type was tested at ANL in 1974 using the 200-MeV proton accelerator, Booster I, with a prototype target station, ZING-P. A second prototype ZING-P² was used in the development of targets, moderators, and instruments, and also for some early scientific experiments. Information gained from the ZING-P² prototype led to the IPNS, which was the

world's most scientifically productive pulsed neutron source for the first 7 years of its operation. It has recently been surpassed by the ISIS facility at the Rutherford-Appleton Laboratory in the U.K.

ANL has a long range goal - the construction of a pulsed spallation neutron source which approaches currently envisioned target and accelerator design limits. The design goal is to produce a peak pulsed neutron flux in excess of 1×10^{17} n-cm⁻²-s⁻¹. Neutron intensity would be 100 times the most powerful pulsed source operating today. The source is based on the FFAG synchrotron concept. This concept uses a de-operated magnet system and a pulsed, frequency modulated rf system to accelerate intense proton beams from about 200 to 1500 MeV. The time-averaged proton current is expected to be 3 to 4 mA.

The FFAG concept has many features that make it attractive for generating the proton bursts for an intense neutron source. These include de-excitation of the main ring magnets so that the capture and rf acceleration process can be optimized. Metal vacuum chambers can be used, which allows an extremely low impedance to reduce beam instabilities by permitting higher beam thresholds before disruptive beam instabilities set in. This phenomenon is further helped by the fact that injection and extraction elements are located only on the very inside and outside edges of the vacuum chamber, reducing the perturbation of the smooth wall to a small fraction of the acceleration cycle. The acceleration cycle is so rapid that the beam moves through instability regions before damaging beam growth can occur.

This type of accelerator is a new and exciting entrant in the field of high-current drivers for spallation neutron sources. However, some past experience does exist with this and generically similar machines such as ring cyclotrons and sector focussing cyclotrons. The concept was extensively studied in the early 1960's at the Midwest Universities Research Association (MURA) laboratory in Stoughton, Wisconsin.

Small electron machines were successfully built and operated, and much of the theory was developed at that time. Beam stacking was demonstrated. Two large high-current ring cyclotrons, similar in many respects to FFAG synchrotrons, are currently in operation in Canada (TRIUMF) and in Switzerland at the Schweizerisches Institut für Nuklearforschung (SIN). The latter routinely operates at 1 mA and 500 MeV with a loss rate that is so low that hands-on maintenance of the machine is possible.

Although this previous experience does exist and is suggestive of the capability of a 3-4 mA accelerator, the fact remains that this does represent innovation in the area of high-current accelerators. After looking critically at this reality, it has been concluded that an accelerator that would be an intermediate step would serve as a powerful facility for neutron scattering research while developing the accelerator technology necessary for future larger sources. A design was proposed in 1984 in which the existing Rapid Cycling Synchrotron (RCS) of the IPNS is replaced by an FFAG synchrotron. The details of this design are given in *The Argonne Neutron Plan*, February 1984. The intent of the plan is to provide a configuration affording critical tests of the principal large machine ideas, for instance, efficient capture and acceleration, the design of the magnets, removal of uncaptured beam, and the success or limitations of beam stacking or beam storage. The proposal submitted to the DOE updates the 1984 plan. PNRF will clearly serve as an important scientific facility in its own right, as well as testing concepts for a next generation source.

Location of the new accelerator on the IPNS site is shown in Fig. 1. The use of present accelerator and target components in the new source will be carefully analyzed in order to minimize costs while optimizing research capabilities. The parameters from the 1984 design are shown in Table I and will be used as a framework, but not as a goal or limitation for the new design.

New Instruments

Since the new Glass, Liquid, and Amorphous Material Diffractometer (GLAD) is now being commissioned, design and development efforts have been focused on the new Small Angle Neutron Diffractometer (SAND) which is the next instrument planned for construction at IPNS. Details of this instrument are presented below. Beyond SAND, the priorities for new instrument development are not fixed. However two possible candidates, a μ eV-resolution inelastic spectrometer and a high-resolution powder diffractometer, are briefly discussed below. In addition to the development of such completely new instruments, continuing attention will be given to upgrades of the current instruments and to increasing the capabilities of the complement of auxiliary equipment. Of course, if PNRF is constructed the full complement of instruments will be reviewed with an eye toward modifying or replacing them in order to optimize the match of the instrumental capabilities to this much more intense source.

SAND: The development of the Small Angle Diffractometer (SAD) and the phenomenal growth of its user program has been one of the major success stories at IPNS. In fact, this instrument has been so successful that it has repeatedly been the most oversubscribed of all the IPNS instruments. For this reason, we began considering the development of a second small angle diffractometer at IPNS. A workshop on small angle diffractometers at pulsed neutron sources was held at ANL in Fall 1986, and the proceedings summarized the experience at ISIS, LANSCE at Los Alamos National Laboratory, and IPNS concerning what had worked and what had not for small angle scattering instrumentation. This workshop highlighted a number of areas which required study before a new small angle diffractometer could be designed for IPNS.

For the next few years, GLAD absorbed most of the development efforts of the IPNS technical staff, so work on SAND proceeded at a

Fig. 1 Schematic representation of the proposed Pulsed Neutron Research Facility (PNRF) at Argonne.

Table I. Parameters for the PNRF Accelerator

Injection energy (MeV)	50
Extraction energy (MeV)	500
Stacking energy (MeV)	350
Injection closed orbit length/ 2π (m)	6.407
Extraction closed orbit length/ 2π (m)	9.666
Number of sectors	16
Angular width of magnet ($^\circ$)	5.625
Field index, k	8
Spiral angle, ϵ ($^\circ$)	57
Magnetic field at injection (T)	0.489
Magnetic field at extraction (T)	1.5
Radial betatron frequency, ν_r	3.25
Vertical betatron frequency, ν_z	2.3
Radial beam emittance at injection (mm-mrad)	200π
Vertical beam emittance at injection (mm-mrad)	150π
Maximum rf voltage per turn (kV)	60
Space charge limit	5.25×10^{11}
Extracted beam repetition rate (Hz)	29.9
Number of beam stacks	4
Average beam current (μ A)	100

fairly slow pace. However, a new 40×40 -cm²-active area position-sensitive detector was procured in 1987, and the CS beam line was earmarked for use by the new instrument.

A major problem with the SAD at this time was the "wings" present in the instrument background near the beam stop, and this problem had to be understood and eliminated before it would be practical to use similar collimation for any new instrument. Detailed computer simulations in 1987 indicated that the wings were due to reflections from the absorbing blades of the converging Soller collimators and suggested that the problem should be significantly reduced if similar collimators with rougher surfaces on the absorbing blades could be produced. On the basis of these simulations, a new set of converging Soller collimators was procured for the SAD. These collimators with the rough surfaces on the absorbing blades were installed in the SAD in Summer 1988 and showed a significant improvement over the previous collimators. The way was thus cleared to proceed with a SAND design based on similar collimation.

Experience with the SAD had shown that it was necessary to block the intense prompt pulse of fast neutrons from reaching the detector to prevent detector and electronic overload problems. On the new instrument, this could be done either by placing a filter in the direct beam (as on the SAD), by using a "chopper" in the direct beam, or by moving the instrument out of the direct beam by means of a curved neutron guide or a multiguide "beam bender". Since a short flight path was desired from intensity and frame-overlap considerations, only the beam bender could provide sufficient displacement of the instrument from the direct beam. Computer simulations analyzed the performance characteristics of a beam bender with parameters chosen to provide the desired instrument displacement. However, the simulations showed that such a beam bender did not offer any transmission advantages over an MgO filter similar to that used by

the SAD, so it was decided to go with the direct beam geometry for SAND with the option of using either a filter or a t_p chopper.

One major goal for the new instrument was to provide detectors at higher angles than could be reached with the area detector in order to extend the accessible Q range to higher Q values. The success of the linear position-sensitive detector (LPSD) development on GLAD led naturally to a decision to use a bank of similar LPSDs to increase the angular range covered on SAND.

A second major goal for the new instrument was to reduce the minimum accessible Q value to $0.002 \text{ \AA}^{-1}$. This could be done either by tightening the collimation or by extending the wavelength range. The latter could be done by using a chopper to eliminate the neutrons from every other pulse, thus making the source effectively 15 Hz and doubling the wavelength range which would be available before the next useful pulse arrived. A drum chopper intended for such frame-elimination use was extensively studied in computer simulations. The simulations showed that the chopper might be practical for this purpose, and that in any case, a chopper of this design should be very effective in eliminating the delayed neutron background from the wavelength regions where it is most troublesome.

Production of converging Soller collimators to provide tighter collimation than that currently available on the SAD would not be easy, nor is it certain that reflection from the collimator blades would not again become a problem at this level. Thus, some development on collimators was required. To test various forms of multiple-converging aperture collimators for use on the new instrument, a prototype small angle scattering instrument was installed on the C3 beam line in 1991. The prototype utilized the SAND area detector and the new data acquisition system and Digital Equipment Corp. (DEC) MicroVAX computer which had already been procured for SAND. A drum chopper of the type considered in the simulation

was fabricated so the prototype instrument could also be used to test the proposed uses of this chopper.

Also in 1991, parameters for SAND were finalized, and design of the new instrument was begun. Figure 2 shows schematically the design of the scattering chamber and final flight path for SAND, and the position of SAND in the IPNS experiment hall is shown in Fig. 3. As can be seen in Fig. 2, the sample chamber is large, making it possible to accommodate a wide variety of auxiliary equipment. Table II lists some of the expected capabilities of this instrument. The final version of the instrument will have two automatically interchangeable sets of collimators to allow optimal compromise between Q range and intensity for a given experiment. It will also have a cooled filter (probably MgO) in the incident beam, which can be removed if a t_p chopper is to be used. A $40 \times 40\text{-cm}^2$ area detector and a bank of 57 LPSDs (total active area $\sim 60 \times 63 \text{ cm}^2$) will be used. It is

anticipated that the sample chamber and final flight path will be installed in Summer 1992. At that time, a temporary version of the incident beam line with a single set of converging collimators will permit limited operation of the instrument, while construction of the final components of the incident beam line proceeds. It is expected that the instrument will be fully completed in 1993.

μeV Spectrometer: The recent success of the LAM-80ET instrument at the KENS facility of the High Energy Physics Laboratory, KEK, Japan has led us to reevaluate our position with respect to high-resolution inelastic and quasielastic spectroscopy. We expect to begin a design study next year to explore the use of a back-reflection crystal analyzer spectrometer to provide resolutions in the 1-10 μeV range at IPNS. After completion of such a study, a decision will be made whether or not such an instrument will be the next major IPNS instrument development/construction project after SAND. A design study

Fig. 2 Design drawing of the scattering chamber and final flight path for SAND.

Fig. 3 Floor plan showing the position of SAND relative to neighboring instruments in the IPNS experiment hall.

by Frans Trouw has shown that the use of guides in the incident beam on the Quasielastic Neutron Spectrometer (QENS), with a resolution of $\sim 70 \mu\text{eV}$, would significantly increase the intensity on that instrument, so completion of the design study on the 1-10 μeV resolution instrument will allow the relative priorities of these two high-resolution inelastic/quasielastic spectrometer projects to be assessed. If the 1-10 μeV instrument were to be built, it would have to go on a solid methane moderator, and since all the beam ports on the "C" moderator are already occupied, it would have either to replace or to share a beam line with one of the existing instruments.

High-Resolution Powder Diffractometer: A design study

for a high-resolution powder diffractometer was carried out by Jim Jorgensen several years ago. This was a multiangle instrument with resolution $\Delta d/d$ varying from 0.00085 at $2\theta = 160^\circ$ to 0.0136 at $2\theta = 30^\circ$. It would require a 50-m incident flight path and an approximately 2-m scattered flight path (scattered flight path length may vary with angle) and would make use of at least partial guides in the incident flight path. No decision has yet been made whether or when an instrument along these lines should be built at IPNS. Also in this case, if such an instrument were to be built, it would have to either replace or share a beam line with an existing instrument.

Upgrade of Existing Instruments: Major upgrades of the

chopper spectrometers have been underway for some time. These include construction of the intermediate-angle flight path for the High Resolution Medium Energy Chopper Spectrometer (HRMECS) - design package nearly complete; provision of t_0 choppers for the Low Resolution Medium Energy Chopper Spectrometer (LRMECS), HRMECS, and PHOENIX - HRMECS and PHOENIX in place, LRMECS being fabricated; provision of choppers and collimators for a high-resolution option on HRMECS and LRMECS - collimators in place, chopper being fabricated; and improved shielding - improvements on LRMECS underway. As a long-range project, we are still investigating the use of magnetic-bearing choppers to achieve still better intensity/resolution optimization.

An important upgrade of all instruments is the conversion from DEC-PDP to MicroVAX computers. This is, in turn, leading to considerable expansion of the data visualization and on-line analysis software capabilities for several of the instruments. Software development leading to the collection of powder diffraction data in real time (i.e., in ~ 1 minute intervals) is planned for the upcoming year.

An ever-continuing upgrade for most instruments is the addition of more detectors. Needs are particularly strong for HRMECS and for the powder diffractometers. The Single Crystal Diffractometer (SCD) could also benefit from the addition of another area detector.

An increasingly important area is the development of ancillary equipment. Requirements for new furnaces, cryostats, sample changers, magnets, pressure cells, etc. arise frequently and are expected to occupy an increasing fraction of our mechanical design and computer interfacing efforts.

Table II. Parameters for SAND

Distances (m)	
source-to-sample	9.0
sample-to-area detector	2.0
sample-to-LPSD	1.524
Beam size at sample (cm dia)	
	2.0
Area detector	
active area (cm ²)	40 × 40
active thickness (cm)	2.5
resolution (mm)	4-6
³ He fill pressure (atm)	2.0
CF ₄ fill pressure (atm)	1.4
encoding type	rise time
encoded information	8 bits X and 8 bits Y
LPSD	
diameter (cm)	1.1
active length (cm)	60
resolution along detector (cm)	1-2
number of detectors	57
³ He fill pressure (atm)	10
encoding type	charge division
encoded information	64 segments per detector
Collimators (rad beam divergence)*	
coarse	0.0034
fine	0.0014
Wavelength range (Å)	
noncrystalline samples	1-14
crystalline samples	5-14
Q_{max} (Å⁻¹)	
$\lambda_{\text{min}} = 1.0 \text{ Å}$ (noncrystalline)	> 2.0
$\lambda_{\text{min}} = 5.0 \text{ Å}$ (crystalline)	~ 0.6
Q_{min} (Å⁻¹)	
coarse collimation	~ 0.004
fine collimation	~ 0.002
Estimated intensity (× SAD)†	
coarse collimation	~ 1.7
fine collimation	~ 0.3

* Both coarse and fine are converging collimators focused on the area detector.

† Relative intensity estimates are for the same Q range using the same moderator on the SAD and SAND and are based only on data near the area detector center. Because of the larger area detector and the bank of LPSDs, the total data rate on SAND will be several times that on the SAD even when fine collimation is used on SAND.

CHAPTER 18

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