PULSED SPALLATION NEUTRON SOURCES

John M. Carpenter
Intense Pulsed Neutron Source Division
Argonne National Laboratory

This paper reviews the early history of pulsed spallation neutron source development at Argonne and provides an overview of existing sources world wide. A number of proposals for machines more powerful than currently exist are under development, which are briefly described. I review the status of the Intense Pulsed Neutron Source, its instrumentation, and its user program, and provide a few examples of applications in fundamental condensed matter physics, materials science and technology.

HISTORY OF PULSED SOURCE DEVELOPMENTS AT ARGONNE

This is a day for remembering, reflecting and projecting into the future. Think back to the year 1968, 26 years ago. Please don't fix upon the gathering war half a world away, the burnings in our cities, or the riots in our own Grant Park; these were dark rumblings of political change that even now have not played out. Think instead of the scientific scene. ZGS had already been operating for five years and improvements were afoot. New, bigger machines were being designed and built around the world for high energy physics research. Several high flux research reactors were under design, construction or commissioning: HFBR (Brookhaven), HFIR (Oak Ridge), HFR (ILL Grenoble), A^2R^2 (Argonne), British HFR. Argonne had an already long-established tradition in neutron scattering based on its smaller research reactors, beginning with Enrico Fermi and Walter Zinn at CP-3, and continuing at the 5-MW CP-5.

In January of 1968, as a young and dewy professor of Nuclear Engineering at the University of Michigan, where I had built some instruments for neutron scattering research at the 2-MW Ford Reactor, I was invited to serve in an instrument design group, led by Don Connor of the SSS division, that was supposed eventually to provide instruments for A^2R^2. That group

just once, early in 1968. Sam Werner, my colleague and classmate at Michigan, then at Ford Scientific Laboratory, was also on that committee. Almost immediately, the A^2R^2 project was canceled.

When it was canceled, A^2R^2 had progressed to the stage of a hole in the ground, a concrete foundation and a reactor mockup facility with a big supply of Beryllium blocks. In May that year, the Laboratory and the AUA established a Committee on Intense Neutron Sources, which was eventually led by Lowell Bollinger and was assigned to look into possible alternatives to the A^2R^2. Bollinger, in Argonne's Physics Division, had a strong program of nuclear cross section measurements at CP-5 and ultimately went on to establish the ATLAS superconducting heavy-ion linac. I have reviewed the meeting minutes, and found the committee membership to have been a somewhat revolving thing. Lee Teng and Tat Khoe took part, but most of the members were reactor types. I learned about spallation sources and the Canadian ING studies. After about one year, examining a wide range of sometimes wild alternatives, we reported the recommendation to pursue a pulsed slow neutron source driven by a proton accelerator. We noted that the main shortcomings of the idea were that we did not know on the one hand the intensity of slow neutrons that such a source would produce, and on the other hand we did not know how effective such a pulsed source would be for neutron diffraction and inelastic scattering studies.

At about this time, Ron Martin returned from a trip to Russia having learned about Dimov's development of a high-current H^- ion source. Ron conceived that this source, feeding a 30 Hz rapid-cycling synchrotron through the existing 50 MeV linac, could provide a chain of 500 MeV pulses to the ZGS, increasing its current by a large factor according to $\beta^2\gamma^3$. He arranged to bring the decommissioned 2. GeV electron synchrotron from Cornell, to rig it for a test of the H^- injection principle with 300 MeV protons--so-called Booster I, and initiated the design of the 500 MeV Booster II. Jim Simpson took the job of bringing the system to reality. I learned of this in 1969 and dreamed up a proposal to use it to drive a pulsed neutron source during the intervals between ZGS injection pulses. I called it ZIING- the ZGS Injector Intense Neutron Generator. My preliminary estimates of the neutron fluxes indicated that they would be interesting but a little low.

So it stood until 1971, when I came to Argonne on my first sabbatical leave. With Oliver Simpson, SSS Division Director, I spent half my time re-evaluating neutron source options, and half my time making use of the new TNTOFS hybrid chopper spectrometer that David Price, Bob Kleb and Mike Rowe had built at CP-5. I had guessed from experience in moderator measurements with Kingsley Graham at Michigan, that a Beryllium reflector could substantially increase the flux in pulsed source moderators; with the moderators decoupled at low energy, the arrangement would preserve the needed short pulses. With Bob Kleb, I worked up the idea for a

pulsed source with a Uranium target, using the Booster-II accelerator. We called it ZING. Don Connor encouraged me to build up a test for this using A^2R^2 Beryllium blocks, the ZING Mockup. I did this, and found that for the simple geometry that I used, the reflector provided a gain of a factor of 10! The factor would be somewhat less for a multi-moderator system. I wrote up the results with Gary Marmer, whom I brought in to keep me honest in matters of the accelerator. I received a patent on the decoupled reflector idea.

I continued to consult at Argonne after my return to Michigan. We drew up a proposal to build ZING and convened a workshop to evaluate its scientific applications, which took place in 1973. About 50 people came; among them was Motoharu Kimura from Tohoku University in Japan. Kimura already had experience using the 300 MeV Tohoku electron linac as a very effective source for neutron diffraction. Others had used powerful electron machines for inelastic scattering, but they were very limited in this application. The ZING spallation source was much more powerful. Kimura made a crucial suggestion--"You must build a prototype!" He stayed on for several months after the workshop and called in his colleague Noboru Watanabe to assist; I took a six-month leave from Michigan in the Fall of 1973 to work on the project.

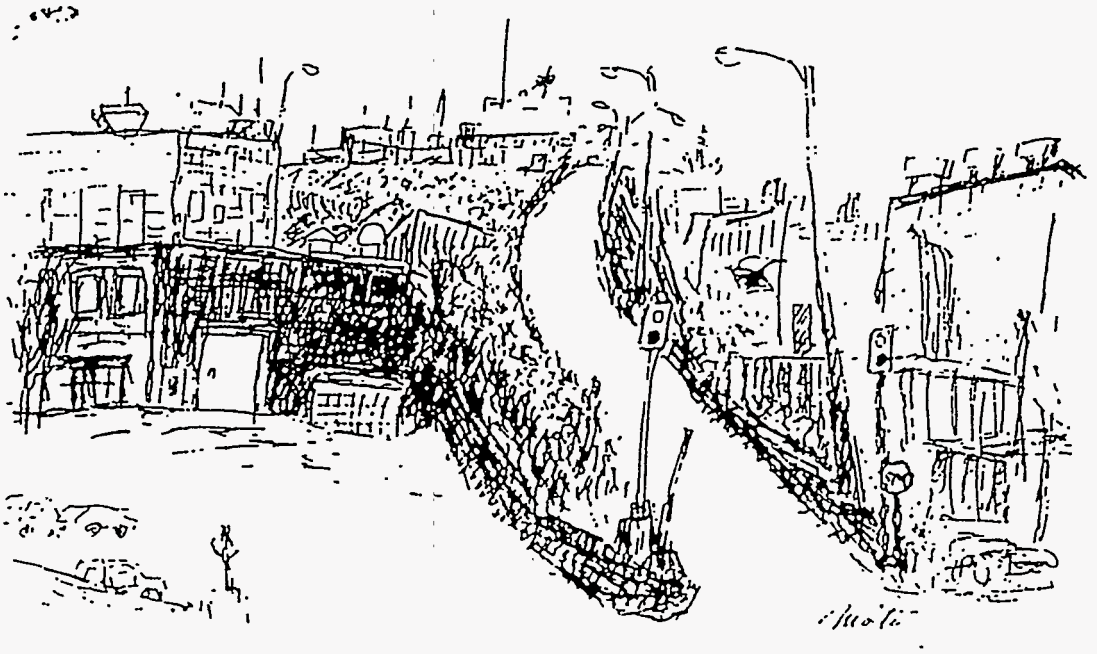

Figure 1. Skunk Hollow, the location of ZING-P, and the entrance ramp to ZGS. Sketch by Motoharu Kimura.

With Bob Kleb's help, we designed a test setup. We called it ZING-P, a little shielded house on top of the beam line between the Booster and ZGS--"Skunk Hollow." Figure 1 shows Kimura's sketch of Skunk Hollow and the ZGS access ramp. The Laboratory eventually

provided $30,000 for the job, and put Tom Banfield, CP-5 director, in charge. We used a stack of armor plate from the dismantled battleship Indiana (Mike Nevitt's ship when he was in the Navy) in the shielding. Kimura had to return to Japan before the project was authorized. He left in disappointment, feeling that his contribution was being ignored. That was not true, it was only administrative delay. We completed the installation in about three months and ran it first in January, 1974. The target was half of a lead brick, with a copper tube cooling pipe. There were two polyethylene moderators, a decoupled Beryllium reflector and two vertical neutron beams. Watanabe helped to design a neutron diffractometer that was built in the SSS shops. The test proved out the basic principles and the intensity estimates. ZING-P ran at intervals until the end of 1975, when it was time to install Booster-II. Figure 2 is Kimura's drawing of the ZING-P experiment area.

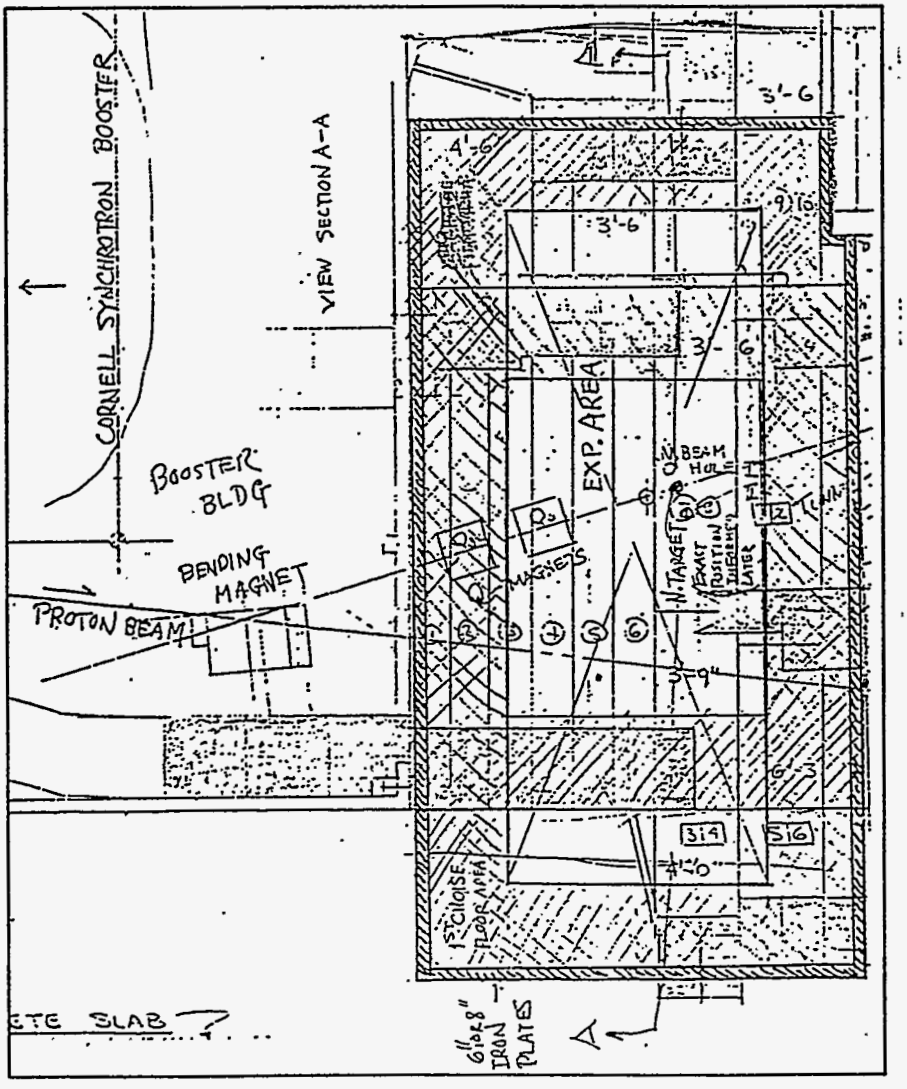

Figure 2. Kimura's drawing of the ZING-P experiment area. The date is May 18, 1973.

Figure 3 shows the lead-brick target and the moderator and reflector arrangement of ZING-P.

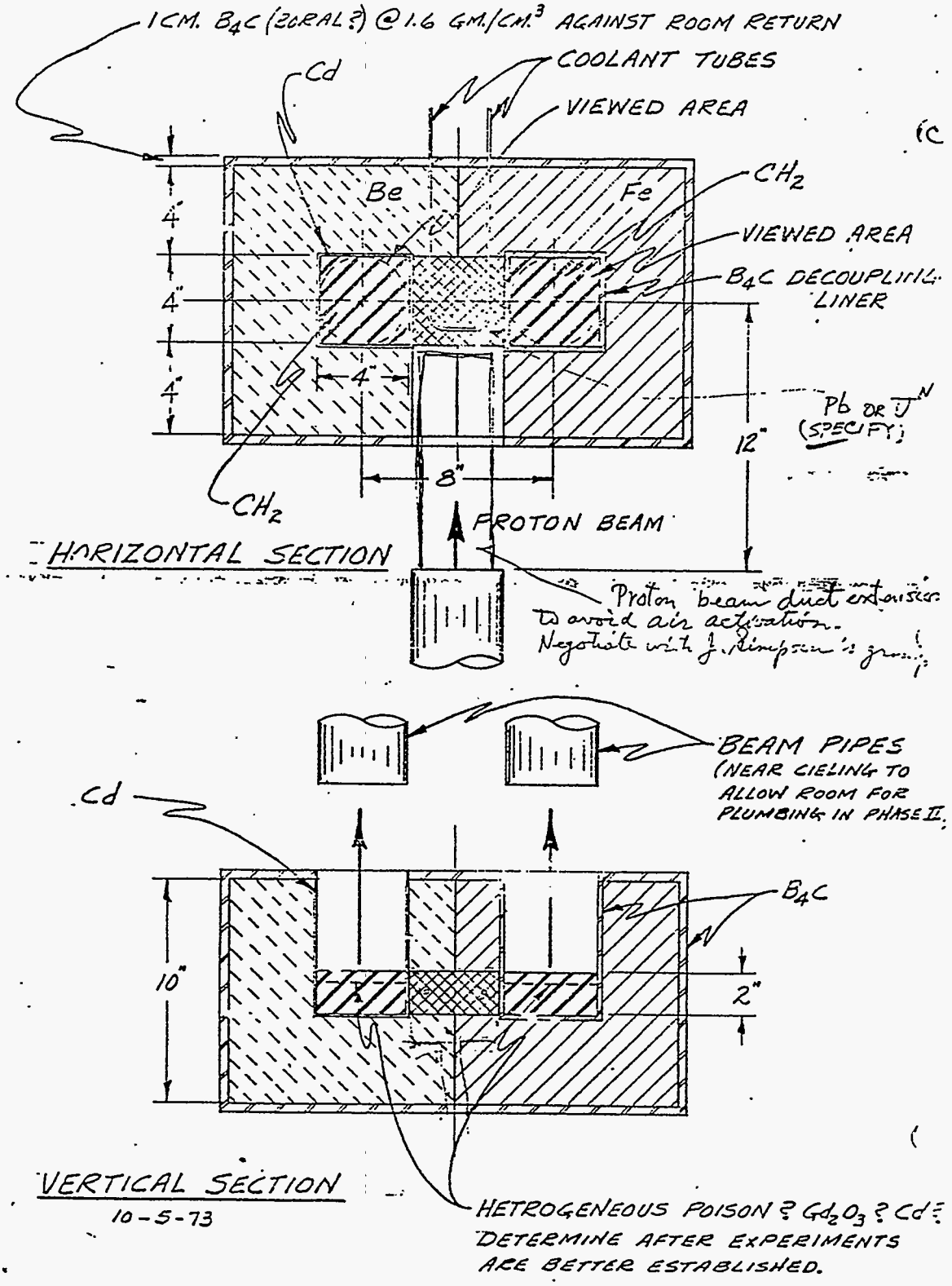

Figure 3. Drawing of the ZING-P target-moderator-reflector arrangement by Bob Kleb.

Figure 4 shows the moderators arranged for time-focusing measurement of the emission-time distribution as a function of neutron wavelength.

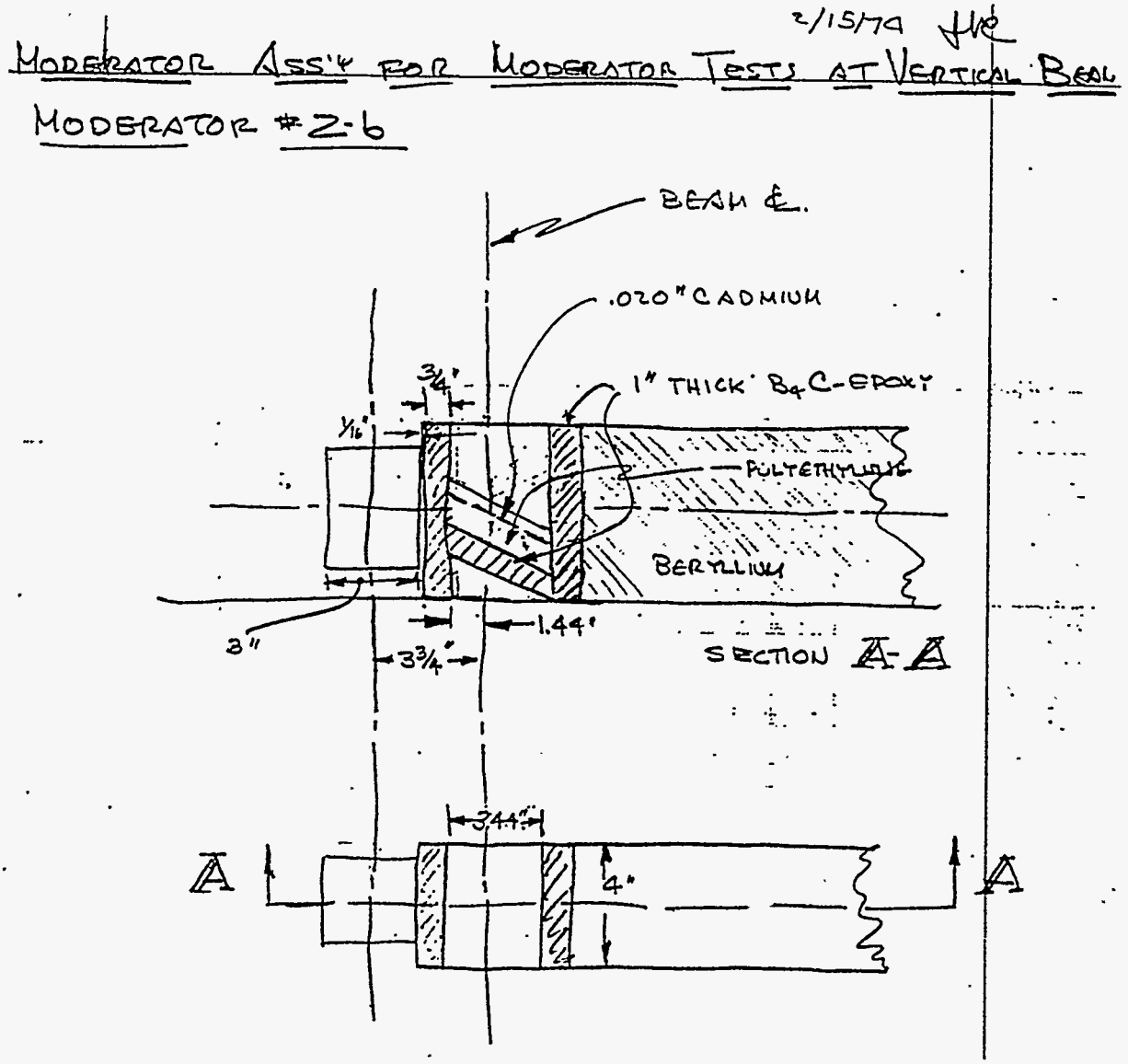

Figure 4. Tilted moderators for time-focused measurements of the shapes of emitted neutron pulses as functions of neutron wavelength.

By then, I had become so involved in the neutron source project that I resigned my full professor position at Michigan and joined Argonne in January, 1975. We designed some modifications to the prototype, including three more horizontal beams and more scattering instruments, to be operated when Booster-II came on-line. That was ZING-P'. Its first target was of W, soon replaced by a target of U, and eventually ZING-P' had a liquid Hydrogen moderator. Prominent in the work were Torben Brun, Bob Beyerlein, David Price, Kurt Sköld, Jim Jorgensen, Bob Kleb, Tom Erickson, Mel Mueller, Selmer Peterson, Art Ries, and Chuck

Pelizzari. Bob Sachs, then Laboratory Director and Mike Nevitt, Deputy Director, encouraged us mightily throughout all this time. ZGS and CP-5 were soon to be shut down and the pulsed source became a matter of higher priority. Kimura returned frequently and assisted with a large number of source performance measurements that were necessary to underpin the IPNS design. In ZING-P and ZING-P' all the essential pieces of the modern pulsed spallation neutron source came together for the first time.

Our ZING proposal ran into trouble in Washington, on the basis that it was too modest--"Go away and come back with a more powerful version," we heard. Jim Simpson, Martin Foss and others worked up the design of a 800 MeV, 0.5 mA High Intensity Synchrotron, HIS, and we laid plans for a new neutron hall. In 1975 we convened another Workshop to evaluate the new proposal, which Sam Werner chaired with me. Paul McDaniel, AUA head, advised me not to name the newly proposed installation frivolously; "Choose an unpronounceable acronym," he counseled, "Make them say it out." We gave up "ZING" and called it the Intense Pulsed Neutron Source, IPNS. We began firing Schedule-44s to Washington.

Booster-II came up with a low current. It ran until 1980, when the current had risen to about 3 μA. Everybody worked on the machine, Frank Brumwell, Yang Cho, Ed Crosbie, Marty Knott, Bob Kustom, Jim Norem, Charlie Potts, Walter Praeg, Tony Rauchas, Jim Simpson, Vern Stipp, Bob Wehrli, all had important hands in the synchrotron development over the years. On the neutron side, ZING-P' was extremely successful. It turned out significant research and we established a user program based on the pattern of ZGS, which was very important.

Our proposals ran into more trouble; now the project was said to be too big, too expensive. We included in later versions of our proposals a quickly-accomplishable intermediate step, IPNS-I, based on Booster-II, (now we called the accelerator the Rapid Cycling Synchrotron, RCS) which was to provide experimental capacity and further experience needed before HIS and IPNS-II could be completed. It finally turned out that only the first phase, IPNS-I, received the funding nod. ZGS shut down on October 1, 1979. We began the IPNS construction project in 1978, based on the use of soon-to-be vacated ZGS areas and soon-to-be-liberated components. IPNS was completed in early 1981, on schedule, and (roughly speaking) within budget. First beam was delivered to the target on May 5, 1981, so today, May 6, 1994, is IPNS's 13th anniversary, plus one day! The reasoning by which IPNS was funded was that it was to be an experiment to test the effectiveness of the new way of doing neutron scattering. By now, we can claim resounding success, but there has never been a more ambitious pulsed source project funded in the US.

Figure 5 shows a plan view of IPNS; in the dim reaches beyond the synchrotron is the ZING-P target.

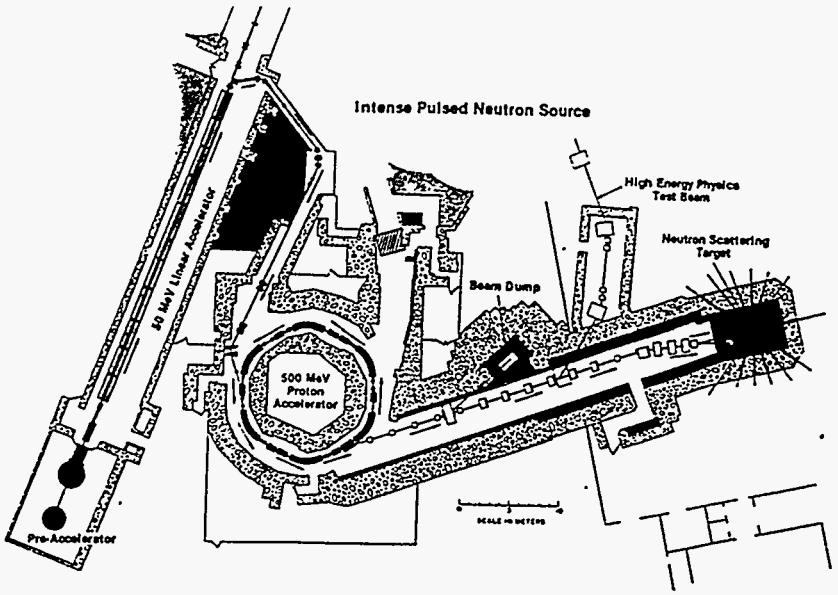

Figure 5. Plan View of IPNS.

Figure 6 shows the experiment hall of IPNS, the former EPB-2 of ZGS. Twelve neutron beams support thirteen neutron scattering instruments. IPNS has three moderators, two of solid methane and one of liquid methane--all cryogenic systems producing cold neutrons as well as copious epithermal neutron beams.

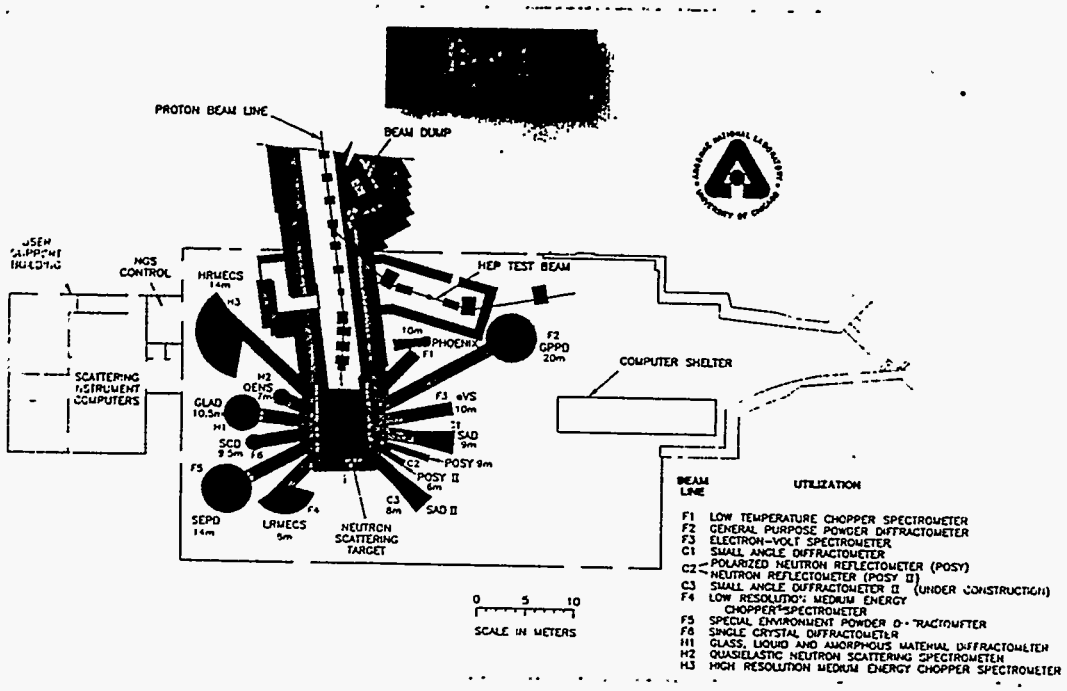

Figure 6. Plan View of the IPNS Experiment Hall.

Figure 7 is a photograph of the IPNS experiment hall.

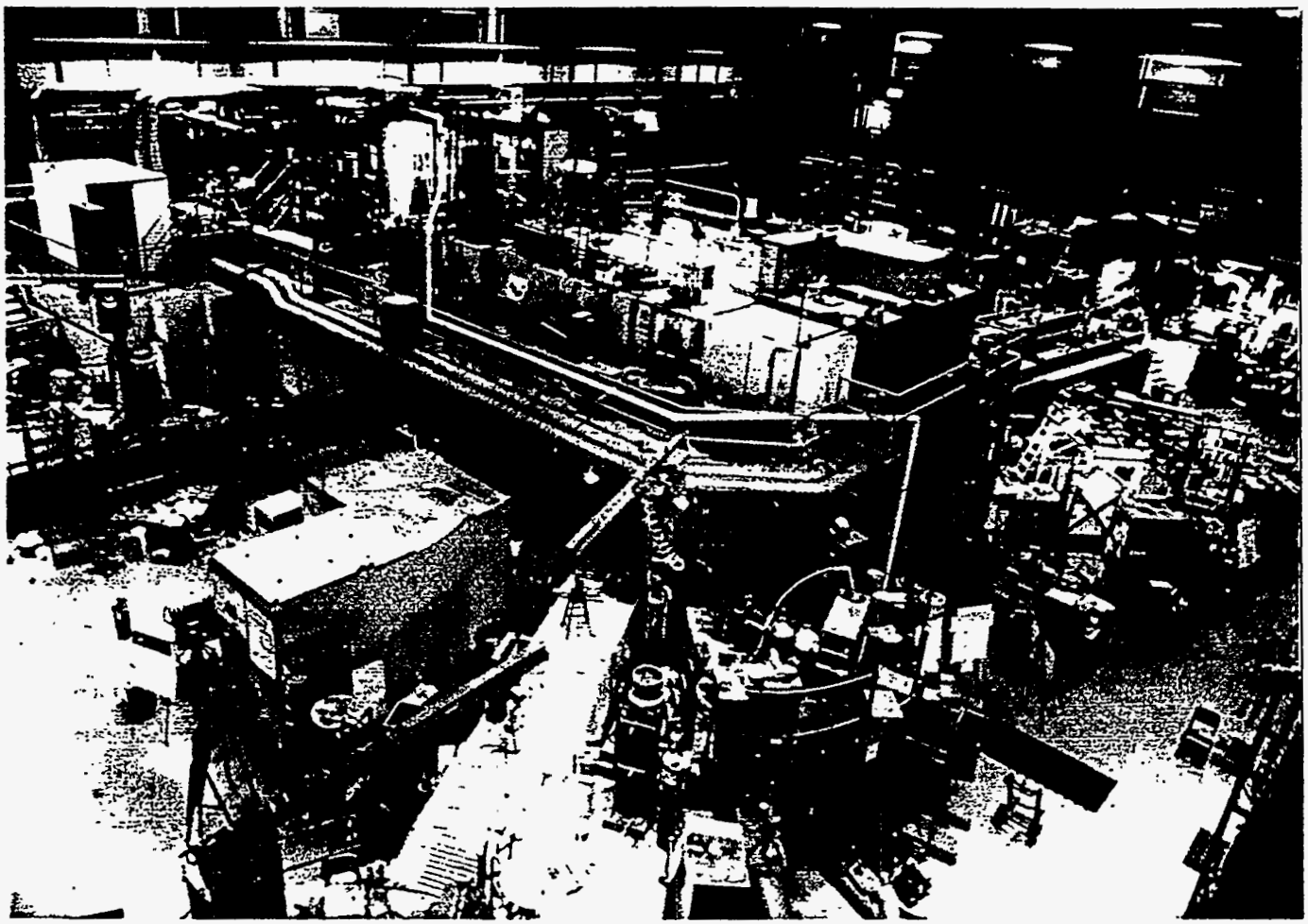

Figure 7. Photo of the IPNS Experiment Hall.

WORLD PULSED SOURCE DEVELOPMENTS

Events transpired elsewhere. In Japan, a pulsed spallation neutron source, KENS, which began operation in 1980 and is an extremely successful, although small research installation, was put up based on the 500 MeV injector to the 12 GeV PS at KEK. The NIMROD accelerator at Rutherford Laboratory in UK was to be shut down. The British adapted the IPNS-II ideas and built what is now ISIS, which started up in 1985 and is now the world leader, an extremely effective research facility. Los Alamos put up a target on LAMPF, originally called WNR, which started up in 1977. It was adapted using a new PSR which started in 1985, now called LANSCE. A giant study was undertaken in Germany, the SNQ, which, unfortunately, never received funding. Construction of a steady spallation source, SINQ, began in Switzerland, which

will be completed in 1995. At the Moscow Meson Factory in Troitsk, work is temporarily stalled but well along toward installing a two-target pulsed/steady spallation source. Table 1 lists the operating pulsed spallation neutron sources and those under construction.

Table 1. Spallation Neutron Sources Operating or Under Construction

Source	Pulsing Frequency	Proton energy, time-average current	Time-Average Beam Power	Startup
KENS (Japan)	20 Hz	500 MeV x 7μA	3.5 kW	1980
IPNS (US)	30 Hz	450 MeV x 15 μA	6.75 kW	1981
ISIS (UK)	50 Hz	800 MeV x 200 μA	160 kW	1985
LANSCE (Los Alamos)	20 Hz	800 MeV x 60 μA	48 kW	1985
SINQ (Switzerland)	Steady source	570 MeV x 1500 μA	860 kW	1995
MMF, INR (Troitsk, Russia)	Pulsed and steady sources	600 MeV x 200 μA	120 kW	?

We really never quit thinking about newer, larger installations. Bob Kustom conceived an FFAG design called ASPUN, and a smaller, prototypical Mini-ASPUN, put forward in 1984. A few years ago, Jim Norem and I put together a series of modifications to IPNS called PNRF. Now we are working with Yang Cho on the design of a new installation, the IPNS Upgrade, to have a 1-MW time-average proton beam. Los Alamos is pursuing the design of its own version of a 1-MW pulsed source, while Brookhaven National Laboratory is studying a "green field" design that starts at 1 MW and is upgradable to 5 MW. Elsewhere, the Japanese have conceived KENS-II as a part of a new facility, the Japan Hadron Project. Austria and an Eastern European consortium have launched the study of a new facility called AUSTRON. The European Union is sponsoring the study of a 5-MW pulsed spallation source, ESS. Table 2 lists the pulsed spallation source studies now under way.

Table 2. Pulsed Spallation Source Studies Underway

Study	Location	Beam Power
IPNS Upgrade	Argonne	1.0 MW
NGSNS	Los Alamos	1.0 MW
AUSTRON	Austria	0.3 MW
KENS-2	Japan	0.2 MW
ESS	Europe	5.0 MW
PSNS	Brookhaven	1.0 —> 5.0 MW

STATUS OF IPNS

The developments that started here at Argonne in 1968 and have proceeded successfully since have spawned this entire new generation of neutron scattering installations, which complement the high flux research reactors and provide capacity for an ever-broadening range of applications of neutrons to the study of materials. Table 3 shows the current status of IPNS. Since startup, the accelerator current has risen while the reliability has stayed at an exemplary 95 %. The original depleted Uranium target was for a time (1988 to 1991) replaced with an enriched Uranium Booster target, which increased the neutron beam intensities by a factor 2.5.

Table 3. What's Happened at IPNS in 10 Years.

Item	Change
Accelerator current	protons i x 4
Booster (U^{235}, k_{eff} = 0.8) Target	neutrons ϕ x 2.5
Cryogenic moderators	cold neutrons ϕ x 150
Number of instruments	4 —> 13
Number of experiments	94 —> 250
Number of visitors each year	89 —> 170
Number of proposals	up 100 %
DOE supported operating time	down 30 %
DOE operating budget (after inflation)	down 9 %
non-DOE operating funds	$1.5 M

The Booster target suffered a cladding failure after three years and is being replaced. The original moderators have been replaced with three cryogenic moderators, two solid methane moderators producing 150 times greater cold neutron flux and other advantages over ambient temperature systems. The number of experiments completed per year has risen to over 300 and the number of users has increased to about 250 different faces each year. The number of instruments has increased from four to thirteen and the number of requested experiment days has doubled. We have completed about 3000 experiments. However, the budget is down in terms that reflect inflation, and consequently operating time is down 30 %. Occasionally, we do work for others, for which we received $1.5 M in FY 1993.

Table 4 shows the details of IPNS utilization for the years 1982 through 1993.

Table 4. Summary of IPNS Utilization.

Fiscal Year	82	83	84	85	86	87	88	89	90	91	92	93	Total
Number of experiments performed	94	110	210	180	212	223	257	323	330	273	210	248	2670
Visitors for at least one experiment													
Argonne	37	41	49	44	52	55	57	60	61	60	53	48	
Other government labs	8	9	8	7	11	15	18	16	19	15	14	18	
Universities	27	33	45	51	79	78	89	94	120	92	62	64	
Industry	5	5	9	7	13	24	20	24	36	18	20	16	
Foreign	12	18	39	35	27	24	17	26	18	27	14	25	
Totals	89	106	150	143	182	196	201	220	254	212	163	171	2087
Number of "user" instruments	4	5	6	6	6	6	6	7	7	7	7	6	
Number of "PRT" instruments	1	1	1	2	3	3	4	4	5	5	5	6	

Industrial research groups make frequent use of IPNS. Table 5 summarizes the names of industrial firms that recently used IPNS.

Table 5 Recent Industrial Use of IPNS

Industry Recent Users of IPNS		
3M Corp	Du Pont	Miles, Inc.
Allied Signal*	Eastman Kodak	Mobay Corp.
Amoco	Exxon	Mobil Oil*
ATT Bell Labs	GE*	SDR
BP America*	Goodyear Tire	Shell Research*
Corning Glass	IBM Almaden	Texaco
Dow	Kraft	

* Purchased beam time for proprietary use

Contributors to IPNS Instruments
BP America
Exxon
IBM
SDR
Texaco

SCIENCE AT IPNS

Following are three examples of experiments completed at IPNS, which illustrate the broad range and high significance of science carried out at IPNS.

DEEP INELASTIC SCATTERING AND THE BOSE CONDENSATE FRACTION IN SUPERFLUID HELIUM

The number of atoms per unit momentum $\vec{p}$ in superfluid materials consists of a degenerate zero-momentum component, the Bose condensate, containing a fraction n_0 atoms and a continuous distribution representing the normal atoms

$$n(\vec{p}) = n_0\delta(\vec{p}) + (1 - n_0) \times \text{continuous function}(\vec{p})$$

If the momentum transfer in scattering is large enough, it is appropriate to represent the scattering function in the "impulse approximation", which describes so-called "deep inelastic scattering,"

$$S(Q,\omega) = \int n(\vec{p}) d^3\vec{p}\,\delta(\omega - \frac{\bar{h}^2Q^2}{2M} - \frac{\bar{h}\vec{p}\cdot\vec{Q}}{M})$$

Since the momentum distribution is isotropic, the result can be expressed in terms of a function of a single variable,

$$= \frac{M}{Q} J(y);$$

where

$$y = \frac{M}{Q}(\omega - \bar{h}\frac{Q^2}{2M})$$

and

$$J(y) = 2\pi\int_{|y|}^{\infty} pn(p)dp\,.$$

Here,

$$\bar{h}\vec{Q} = \bar{h}(\vec{k}_i - \vec{k}_f)\,,$$

is the momentum transfer and the (non-relativistic) energy transfer is

$$\bar{h}\omega = (E_i - E_f)\ \text{, where } E = \frac{\bar{h}^2}{2m_n}k^2\ .$$

Figure 8 shows calculated momentum distribution functions for normal and superfluid liquid Helium.

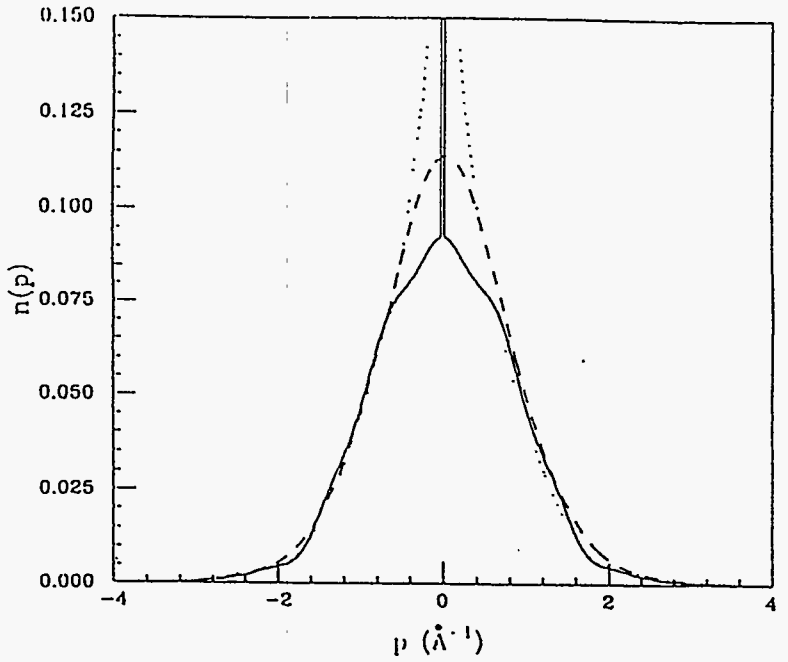

Figure 8. The momentum distribution of normal and superfluid liquid Helium. Solid line, Green's function Monte Carlo; dotted line, variational calculation, dashed line, path integral Monte Carlo calculation for normal liquid.

Measurements are best done at a pulsed source, capitalizing on the high flux of epithermal (higher than thermal) neutrons and the accompanying high pulse resolution, using a chopper spectrometer. Figure 9 shows the results of measurements at IPNS, expressed in terms of J(y), for normal liquid at 3.5 K and for superfluid at 0.35 K.

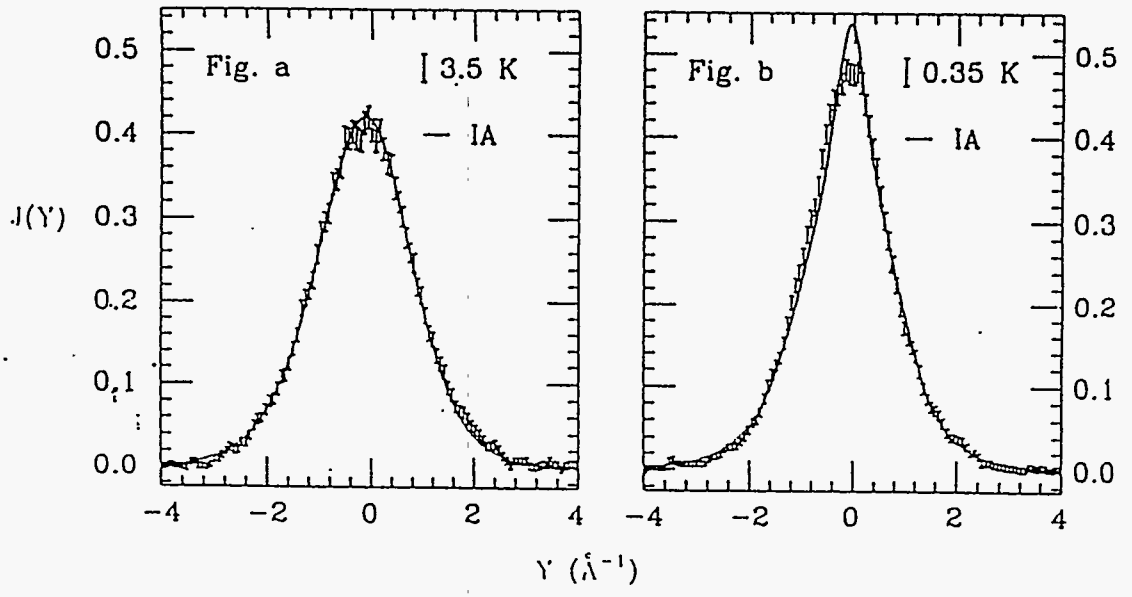

Figure 9. Measured reduced momentum distribution functions J(y), for normal liquid at 3.5 K and for superfluid at 0.35 K.

The impulse approximation (IA) matches the measurements for the normal liquid, but fails in the case of the superfluid. The reason is that the IA fails to account for "final state effects," which broaden both the delta-function and the continuous distribution. Fortunately, theory developed at the same time as the measuring techniques were refined which enabled accounting for these effects, providing the means for extracting the condensate fraction n_0 and producing an essentially perfect fit to the data.

Figure 10 shows the variation of the condensate fraction as a function of temperature for liquid Helium at saturation pressure. The data have been fitted to a function

$$n_0(T) = n_0(0)[1-(T/T_\lambda)^6] \quad ,$$

with $n_0 = 8\%$ and where T_λ is the superfluid transition temperature, 2.17 K.

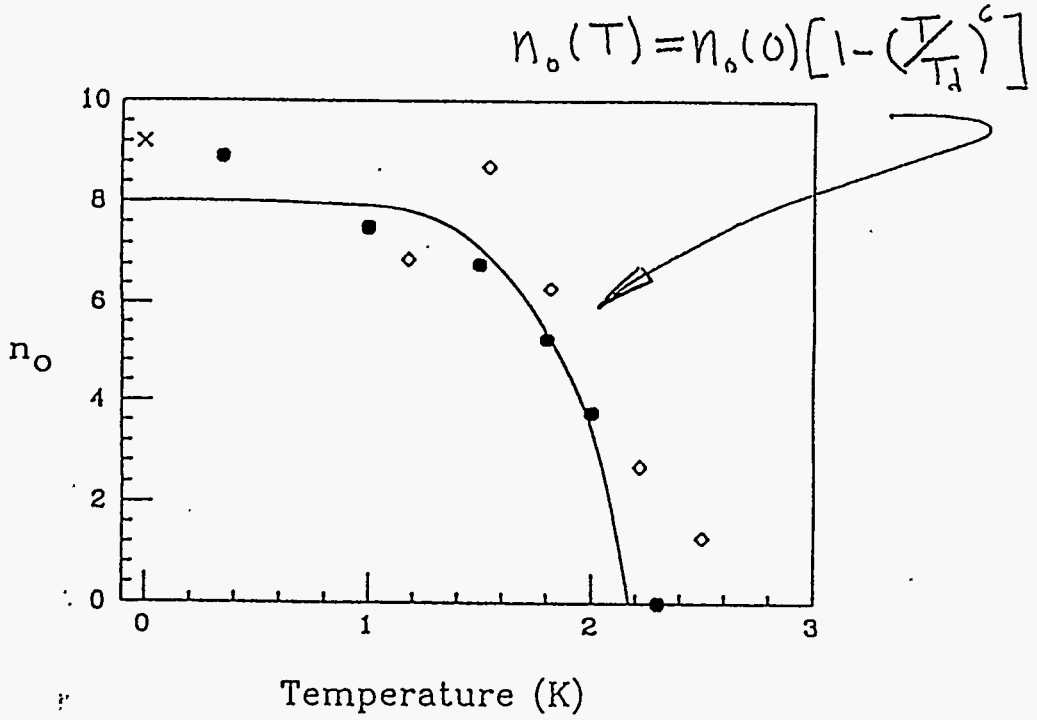

Figure 10. The Bose condensate fraction as a function of temperature. Heavy dots represent the data; the cross is the result of a Green's function Monte Carlo calculation and the diamonds are path integral Monte Carlo results. The solid line is a fit to the data.

The extensive work on this subject has largely been pursued at IPNS, and even began at the prototype ZING-P'. Leading in these accomplishments have been Ralph Simmons (University of Illinois) and Paul Sokol (Penn State University). The work has cleared up many long standing questions of the theory of superfluid Helium, and now has progressed to the study of more complicated systems. He-3/He-4 mixtures, joint pressure and temperature dependence of n_0, n_0 in restricted geometries (He

in porous and layered materials), and Hydrogenous systems; all of these types of measurements have already been undertaken.

STRUCTURE STUDIES OF HIGH-T_C SUPERCONDUCTORS, $YBa_2Cu_3O_{7-\delta}$, &c.

The discovery in 1986 of the new class of copper oxide superconductors launched world wide studies of their properties. The first determination of the structure of YBCO, as it has come to be called, was done at IPNS in the Special Environment Powder Diffractometer, by Mark Beno and his colleagues. Hi-T_C materials come in indefinite number of varieties: $A_aB_bC_cD_dCu_3O_x$, all polycrystalline ceramics. Neutron diffraction, especially pulsed-source neutron diffraction, it turns out, is THE WAY to explore the structure/function relation relationships in these materials. IPNS and all other pulsed spallation sources are still very busy on these materials. Questions addressed span the range from fundamental to practical, from new superconductors to non superconducting prototypical materials; composition, defects and vacancies, crystallographic phase transitions, multiple phases, *in situ* preparation and treatment variables, texture, composite materials. Jim Jorgensen and his colleagues have led the world in their ongoing program of studying these materials.

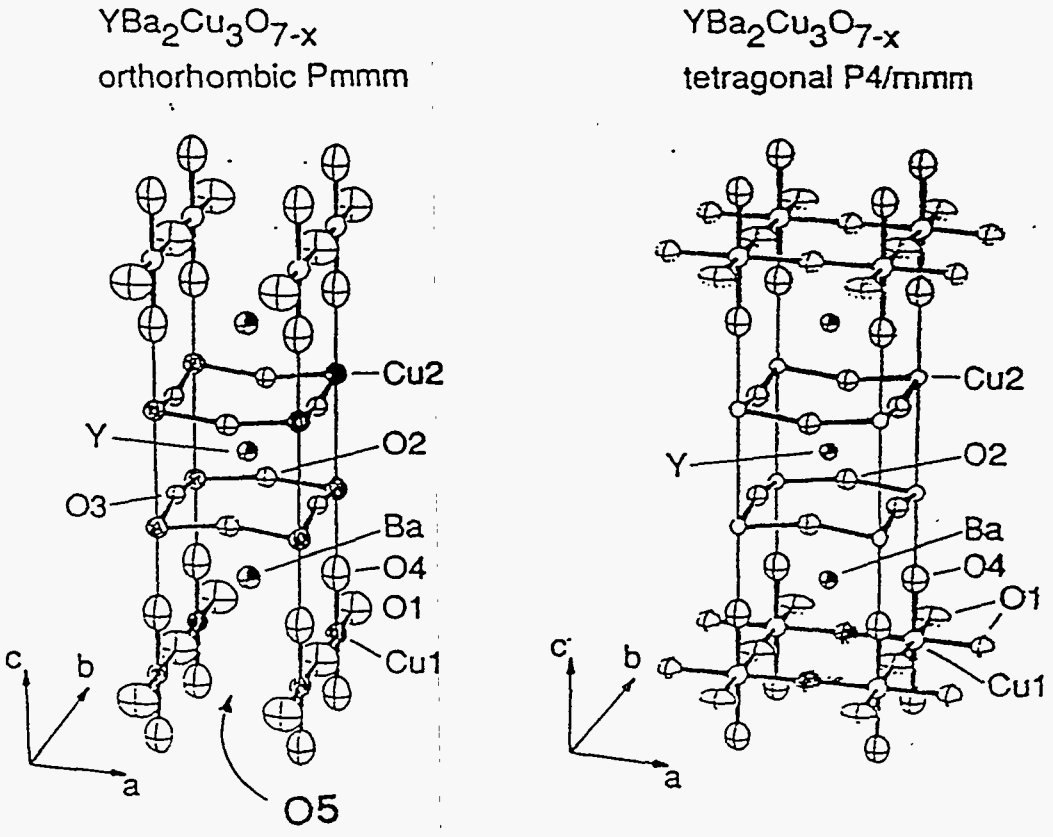

Figure 11. Structures of YBa2Cu3O7-δ. Left, superconducting, orthorhombic phase; right, insulating, tetragonal phase.

Figure 11 shows the unit cell structures of two phases of YBCO, the high-temperature tetragonal, normal (insulating) phase and the low temperature orthorhombic, superconducting phase, determined at IPNS. These measurements and an extensive program of further investigations are possible not only because of the power of the pulsed source diffractometers and the well developed analysis techniques, but also because of the presence at Argonne of excellent capabilities for materials preparation and characterization by other methods.

A TEST OF THE REPTATION MODEL OF POLYMER DIFFUSION

The first instrument for neutron reflectometry, POSY, was constructed at IPNS by Gian Felcher and Bob Kleb and won an IR-100 award in 1987. Since its installation, instruments for this purpose have been built in almost all the neutron facilities in the world. The technique is most powerful and flexible in its time-of-flight form, appropriate for pulsed sources. Neutron reflection is similar in its fundamentals to the critical reflection of light, as, for example, from an air-glass interface. There is perfect reflection for angles less than the critical angle, and no information is to be gained. For angles above the critical angle, the reflection probability is less than unity and depends on the neutron wavelength, the incident angle, and the details of the variation of the refractive index below the surface. Since the refractive index for neutrons depends on nuclear scattering lengths which vary irregularly with (A,Z), the technique is uniquely sensitive to variations of chemical (nuclear, isotopic) concentration beneath the surface. The contrast between H and D, the common and heavy forms of Hydrogen, is especially noteworthy and useful. Because the refractive index varies linearly with wavelength, time-of-flight measurements as a function of wavelength at a fixed angle of reflection reveal the entire variation of the reflectivity. Using polarized beams of neutrons, the reflectivity can be measured as a function of the variation of magnetization density beneath the surface, which is a further unique feature of the method. The measured reflectivities can be analyzed to provide the variation of the index of refraction as a function of depth below the surface.

Bill Dozier (IPNS), Tom Russell (IBM), G. Agrawal (UIC) and others used this method in a series of measurements to investigate the motions of polymer molecules across the interface between two layers. The strength of a polymer interface depends on inter diffusion of molecules across the surface. How this inter diffusion takes place is not only a practical question but also one with deep theoretical implications and is the subject of a theory of DeGennes. The long polymer molecules in the bulk of material diffuse along their length like snakes in a basket of snakes--a motion he dubbed "reptation." The clever IPNS measurements proved out this theory for the first time.

A sample was prepared of two kinds of "block" copolymers. One variant, called HDH, consisted of a block of completely deuterated material in between two blocks of normally hydrogenated polymer. The other variant, DHD, however, was built in the opposite way, a hydrogenated block between two

deuterated blocks. In an ideal case, the materials do not contrast if they can be made to have the same average scattering length densities. A layer of the one was placed on a layer of the other--of course, the two layers are at first distinct. After annealing at elevated temperatures for various lengths of time, the two layers grow together, as shown in Figure 12, illustrated for the case of perfect contrast matching.

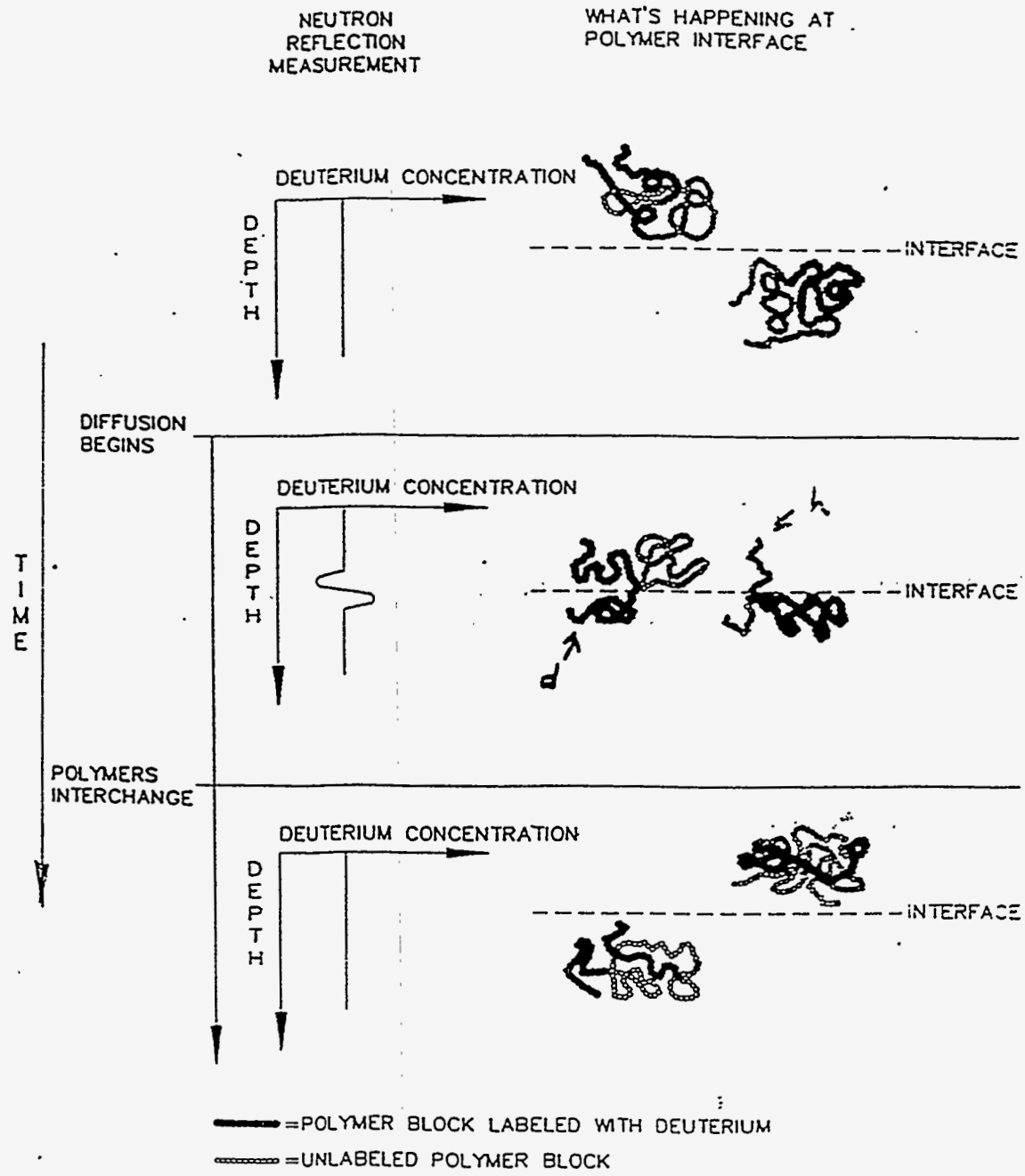

Figure 12. The reptation model of polymer diffusion across an interface. Black spheres represent deuterated polymer blocks, D; white spheres represent normally-hydrogenated blocks, H.

If the reptation model is correct, in early times Ds from the DHD side diffuse into the HDH side and Hs from the HDH side diffuse into the DHD side because these are at the ends of the respective

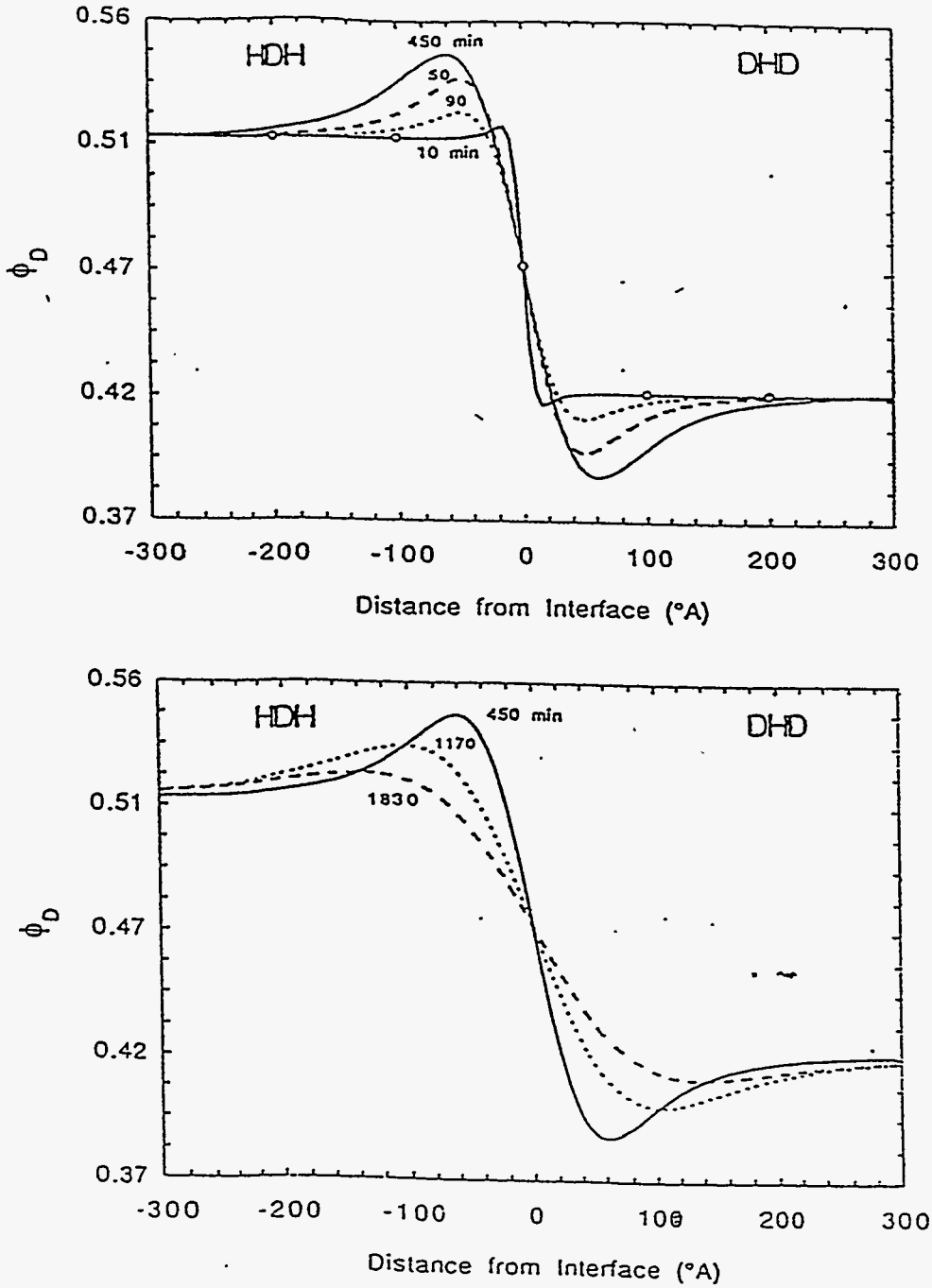

Figure 13. Results of reflectivity measurements of the Deuterium concentration across a labeled polymer interface as a function of distance, for different annealing times.

molecules. This produces an enrichment of Ds and corresponding depletion of Hs on one side of the interface and vice versa, so that a region of contrast develops which influences the neutron reflectivity. After long annealing times, when complete molecules have had the chance to pass across, the interface becomes smeared out and the contrast disappears. Figure 13 shows the deuterium density variation as a function of distance, as determined from reflectivity measurements after various annealing times. In the actual measurement, the two materials are not exactly matched in average Deuterium density, so at first there is a sharp step in contrast, then this step spreads out as annealing progresses.

This is the first definitive evidence that polymer diffusion follows DeGennes's reptation model. The demonstration could only have been accomplished with neutrons. It has clear implications for the understanding of polymer bonding and interlayer adhesion.

CONCLUSION

What began as a modest test using Argonne accelerators has grown to represent a highly significant new category of sources for neutron scattering research and other applications, a powerful complement at least and an alternative perhaps, to research reactors for these purposes. Development proceeds on a world wide scale and Argonne's accelerator group, neutron physicists, engineers and materials scientists can justly reflect that they started something good.