

ranged to collaborate with Thomas Koetzle of the Chemical Structure group at Brookhaven to work with one of the x-ray beam lines when it becomes available. They plan to emphasize efforts to determine phases of crystals without a center of symmetry, particularly organic materials. —TVF

References

1. W. N. Lipscomb, *Acta Cryst.* **2**, 193 (1949).
2. S. Miyake, K. Kambe, *Acta Cryst.* **7**, 218 (1954); K. Kambe, *J. Jap. Phys. Soc.* **12**, 13 (1957).
3. R. S. Williamson, *Thesis*, Polytechnic Institute of Brooklyn (1957).
4. M. Hart, A. R. Lang, *Phys. Rev. Lett.* **7**, 120 (1961).
5. B. Post, *Phys. Rev. Lett.* **39**, 12 (1977); *Acta Cryst.* **A35**, 17 (1979).
6. H. Juretschke, *Phys. Rev. Lett.* **48**, 1487 (1982).
7. R. Colella, *Acta Cryst.* **A30**, 413 (1974); L. D. Chapman, D. R. Yoder, R. Colella, *Phys. Rev. Lett.* **46**, 1578 (1981); R. Colella, *Z. Naturforsch.* **37a**, 437 (1982).
8. S.-L. Chang, *Phys. Rev. Lett.* **48**, 163 (1982).

Spallation sources for neutron scattering

Low-energy neutrons are particularly well suited to probe crystalline, glassy and biological materials. The de Broglie wavelength of a thermal neutron is of the same order as interatomic spacings, and its anomalously high cross section for scattering off hydrogen nuclei makes it an especially useful probe for hydrogenous substances. Furthermore, because its electromagnetic interactions are limited to its magnetic dipole moment, the slow neutron has a very large penetration depth with little disruption of the sample under study, and it serves as an excellent probe of complex magnetic structures.

Given the unique capabilities of neutron scattering for the study of condensed matter, the Materials Sciences Division of DOE has recently been at pains to ensure that existing facilities and future plans make the best possible use of its very limited budget. Two years ago a review panel of neutron scattering research, commissioned by DOE and chaired by William Brinkman (Bell Labs), came to the harsh conclusion that the Intense Pulsed Neutron Source (IPNS I), then under construction at Argonne, should be terminated even before its completion, if additional funding for neutron scattering research was not forthcoming.

The panel had been asked to consider the prospect that funding in the near future would be more-or-less constrained to its 1980 level (\$16 million), with little more than modest adjustments for inflation. "If constrained to a constant level of funding (which the panel feels is inadequate to maintain the long-term vitality of this field), we recommend [that the Argonne neutron-scattering] research staff use the facilities at the other laboratories," Brinkman and his colleagues told DOE in their October 1980 report.

But now, after a year of successful operation at IPNS I and a second look by another review panel (again chaired by Brinkman), the prospects for Argonne's pulsed neutron facility are somewhat less gloomy—at least for the

short run. Because IPNS I was to be the world's first intense pulsed spallation neutron source, DOE had decided to see it through a year of operation before phasing it out. IPNS I has now been serving experimenters since November 1981. In the light of this experience, the second Brinkman report (submitted to DOE in September) strongly urges that additional funds be made available to keep IPNS I going through fiscal 1984. The Argonne facility is running "extremely well, and ... the laboratory has made outstanding progress in establishing a strong users program," the report concludes. "The community is gaining invaluable experience in the use of pulsed neutron sources."

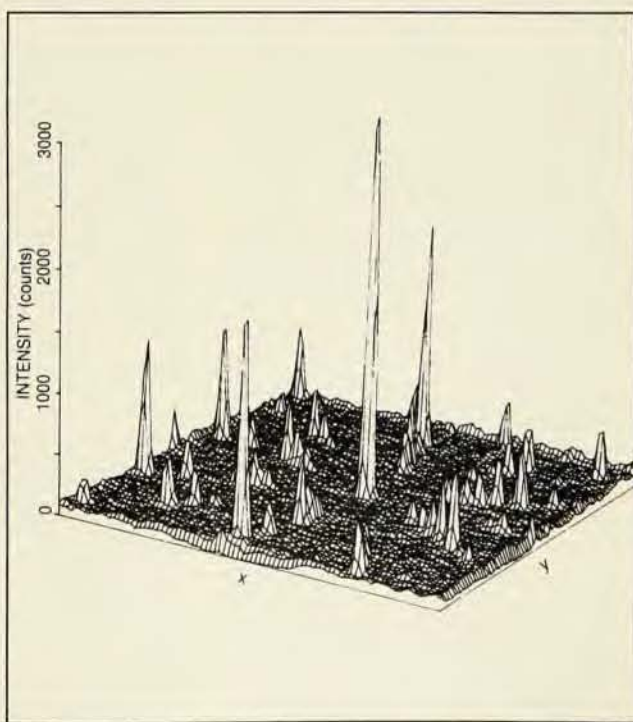
But in the longer run, neutron-scattering research in this country will be centered at Los Alamos, Brookhaven and Oak Ridge. The high-flux reactors at Brookhaven and Oak Ridge have

served as the primary sources for thermal neutron beams during the past decade. In the mid 1970s Argonne and Los Alamos proposed to build second-generation, accelerator-based pulsed spallation sources. Argonne's IPNS II proposal has not been regarded as a live option in recent years, primarily because it would cost about \$100 million. The significantly less expensive Los Alamos proposal to upgrade its Weapons Neutron Research spallation facility with a proton storage ring has been particularly attractive to the cost-conscious neutron-scattering community because it is based on the already existing LAMPF linac, and the cost of constructing the storage ring would be borne by DOE's military applications program. Both Brinkman reports urge that pulsed neutron-scattering research in the latter half of this decade be concentrated at the upgraded Los Alamos spallation facility. Construction of the proton storage ring has already begun.

"Spallation" is a term borrowed from stoneworking and geology. A single high-energy proton hitting a heavy-metal target can dislodge dozens of neutrons. The analogy is to the numerous chips sent flying from a rock by the blow of a "spalling" hammer.

Pulsed spallation sources promise the neutron-scattering researchers two potential advantages (PHYSICS TODAY, December 1979, page 42) over reactor-based, steady-state neutron beams. In an accelerator-based spallation facility, short bursts of high-energy protons (up to a GeV) colliding with a spallation target produce a broad energy spectrum of neutrons with a well-defined

Time-of-flight Laue pattern for a crystal of $K_{0.26}WO_3$ as measured with the single-crystal diffractometer at Argonne's IPNS I pulsed spallation neutron source. The display of Bragg reflection peaks is a sum over 117 time-of-flight channels, corresponding to a wavelength range 1–3 Å. In a three-dimensional representation (x, y and wavelength) the Bragg peaks are fully resolved. The data were taken by an Argonne, Florida State University, Northern Illinois University collaboration.

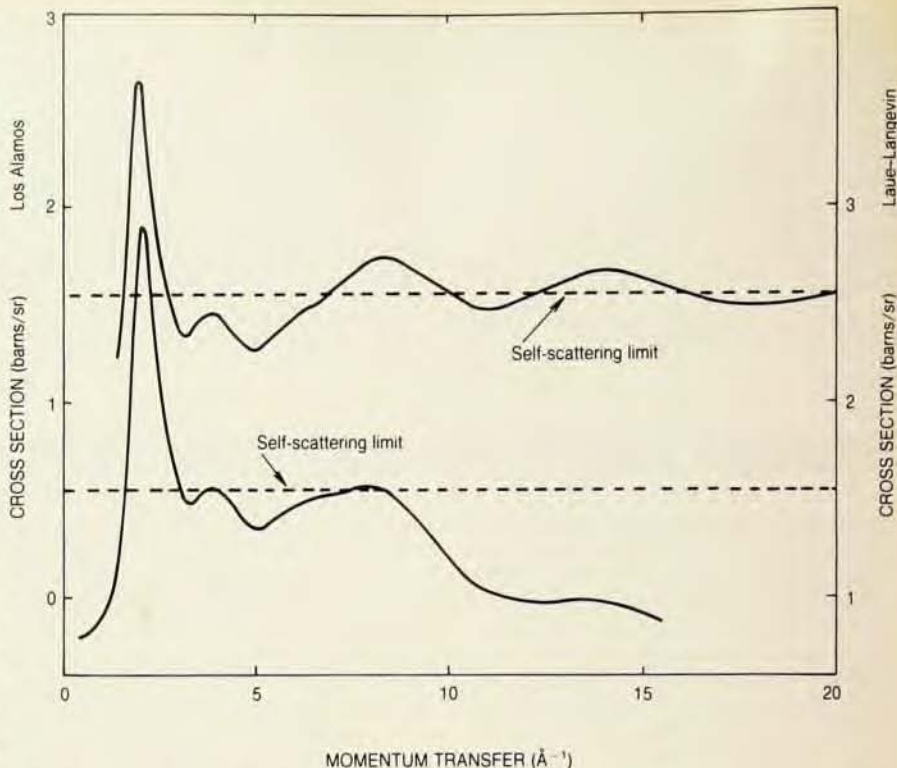


time of origin. Thus one can easily do time-of-flight spectroscopy, and one has neutron energies ranging from a hundredth of an electron volt to hundreds of MeV. The continuous reactor beams, on the other hand, are limited to thermal energies (up to about 0.1 eV), and they do not provide a temporal reference for time-of-flight measurements. On the other hand, the high-flux reactors produce enormously greater time-averaged neutron fluxes than one can hope for with a spallation source. With so many neutrons to spare, one can in fact use beam-chopper techniques to do time-of-flight spectroscopy with thermal neutrons at the reactors. The reactors' principal limitation is the absence of epithermal (0.1 to 1 eV) neutrons.

The Brookhaven and Oak Ridge reactors have served the neutron-scattering community well since the mid 1960s with reliable, high-flux beams. High-intensity spallation sources, by contrast, represent a new and relatively untried experimental mode. Therefore both Brinkman reports conclude that these two "steady-state reactors... will be the mainstays of the DOE neutron-scattering research program for at least the next decade, and that increased funding for these facilities is a first priority." But because these reactors are getting on in years, and the problems of building new reactors are well known, "the pulsed spallation sources may offer the optimal opportunity for future high-intensity neutron sources. Research and development efforts with these sources in necessary for the future of the field."

The second Brinkman panel was "encouraged by the increased collaboration between Argonne and Los Alamos; indeed such cooperation is essential for an orderly transfer of instrumentation to occur in the 1985-86 time frame." Louis Ianiello, director of the DOE materials sciences program, told us that DOE is stressing the development of a cooperative national effort in pulsed neutron research. "We're anxious to pool the expertise of the two laboratories in a joint effort to develop new instrumentation and spallation target designs."

The Argonne group has been developing spallation sources since 1974, when it built ZING P, a small prototype pulsed neutron source using a few tens of nanoamps of 200-MeV protons from a small experimental synchrotron. The proton storage ring now under construction at Los Alamos is scheduled for completion in 1985. It will accumulate 800-MeV protons from the Los Alamos Meson Physics Facility linear accelerator and direct them to a spallation target in intense pulses of 270 nanoseconds duration, with a time-averaged current of 100 microamps.



Comparison of neutron-scattering experiments on D₂O with a spallation neutron source (top curve, Los Alamos data) and a reactor source (bottom curve, Laue-Langevin Institute). One sees an advantage of having higher energies available from the spallation source. Ideally, the cross section fluctuates about the self-scatter limit. At reactors, however, interpretation of the data is made difficult because one needs to go to large angles to reach requisite momentum transfer, bringing in inelastic-scattering terms that drop the cross section below this limit.

Argonne's IPNS I begins with H⁻ ions accelerated in the small 15-MeV linac that originally served as the injector for the Zero Gradient Synchrotron, a proton accelerator for high-energy physics that was shut down in 1979. The electrons are then stripped off and the resultant 50-MeV protons are injected into a rapid-cycling 500-MeV synchrotron that was originally intended as an energy booster for the ZGS, but was not finished before the 1979 shutdown. One starts with H⁻ ions instead of protons because the change of charge sign when the electrons are stripped facilitates injection into the synchrotron. By this trick (which is also exploited at Los Alamos) one is essentially getting around Liouville's theorem, which would otherwise limit the concentration of the beam in phase space.

The synchrotron generates thirty 100-nsec bursts per second of 500-MeV protons with an average current of 8 microamps. Striking one of the facility's two uranium spallation targets, each proton produces an average of 20 neutrons, with energies up to several hundred MeV. The peak neutron flux, about 5×10^{14} neutrons/cm² sec, is substantially higher than that now available at any other pulsed spallation source in the world.

The target used for neutron scatter-

ing experiments is surrounded by hydrogen-rich moderators that slow the neutrons down to thermal and epithermal energies. While thermal neutrons, with wavelengths on the order of an angstrom, are used at IPNS I primarily for neutron-diffraction studies of crystal and magnetic structure, the shorter-wavelength epithermal neutrons, which are not available in reactor beams, have energies well suited to excite higher-energy phonons and other collective excitations.

The second IPNS I spallation target, surrounded by non-moderating reflectors, yields neutron beams with energies up to several hundred MeV for radiation-damage studies. This target area has cryogenic facilities that let one maintain samples at liquid-helium temperatures during intense irradiation. The investigation of fundamental neutron-damage mechanisms is best done at very low temperature because these mechanisms are to a large extent obscured by annealing even at 30 K. The understanding of such mechanisms is important for the prediction of damage to structural materials and superconducting magnets in fusion and fission reactor environments. Because the Los Alamos upgrade plans do not at present include a radiation-damage target area, DOE is looking into the construction of a new cryogenic radi-

ation-damage facility at a steady-state reactor.

About a hundred experiments have been run in the past year at the thirteen neutron beam lines of IPNS I. For elastic neutron-diffraction studies, the facility has two powder diffractometers (for the study of glasses and polycrystalline materials) and a single-crystal diffractometer employing a novel scintillation area detector recently developed at Argonne—an instrument rather like the Anger gamma-ray cameras used for medical imaging.

For such elastic-scattering experiments, measuring the direction and time of flight of the scattered neutron is sufficient to specify the kinematics of the scattering event, despite the fact that the incident beam has a broad energy spectrum. For inelastic scattering phenomena, on the other hand, one needs an additional trick to determine both the incident neutron energy and the energy lost to collective modes. To this end, the IPNS I facility has two chopper spectrometers—rotating-shutter systems in the neutron beam that permit one to select a narrow incident-energy band by time-of-flight restriction on the neutrons before they reach the scatterer. These chopper spectrometers are used primarily for epithermal-neutron experiments, providing energy resolution down to 2%.

The crystal-analyzer spectrometer at IPNS I is particularly useful for the study of proton modes in metal hydrides. The slow-neutron scattering cross section is an irregular and feature-rich function of the atomic number of the scattering nucleus. The cross section turns out to be higher for scattering off a hydrogen nucleus than for almost any other nuclear species. Neutron scattering experiments exploit this anomalously high cross section to probe the arrangement of hydrogen in biological materials, metal hydrides, battery electrodes and other systems of interest. Because the cross section for neutron scattering off deuterons is not anomalously high, one can use the replacement of hydrogen by deuterium as a differential labeling technique for neutron scattering studies of biological materials and polymers.

At Los Alamos, pulsed-neutron-beam experiments began in 1979 with the completion of the Weapons Neutron Research facility—a spallation source built primarily for military research, but shared by condensed-matter and nuclear-physics experimenters. Although the LAMPF linac, which generates the 800-MeV protons for the WNR spallation target, is the world's highest-intensity proton accelerator, WNR can at present produce a peak pulsed neutron flux of only 5×10^{13} neutron/cm² sec—primarily because one can use only a small fraction of the relative-

ly long LAMPF pulses to produce spallation pulses short enough for time-of-flight spectroscopy.

The purpose of the 90-meter circumference proton storage ring now under construction at the WNR facility is to permit the spallation source to make better use of the intense LAMPF proton beam. The ring will serve as an accumulator, converting long proton pulses (750 microseconds) from LAMPF to 270-nanosecond bursts without loss of protons. Each 800-MeV proton from LAMPF will produce an average of about 20 neutrons from the spallation target. When the storage ring is completed in 1985, the 100-microamp average proton current should yield a neutron intensity more than twenty times that of IPNS I.

Because the Los Alamos facility will have only a single spallation target, it will not be used for radiation-damage studies. But in addition to thermal and epithermal neutron-scattering studies, it will serve for high-energy nuclear physics and weapons research. Although the weapons program will require only about 20% of the facility's beam time, the entire construction cost of the storage ring will be borne by the DOE Office of Military Applications. The present design calls for six beam lines for neutron scattering experiments.

"A number of valuable experiments are being performed [currently at the WNR] that explore the unique capabilities of pulsed sources, particularly at epithermal energies," the second Brinkman panel reports. The stress of the current WNR program is on the exploration and development of new techniques and instruments. Two new spectrometers have been built for inelastic neutron scattering. For diffraction studies, a general-purpose diffractometer is being used to study liquids, amorphous materials and powders, and a single-crystal spectrometer with a unique two-axis design is under development. A formal visitors program will begin next year.

The Brinkman panel had made its 1980 report after visiting the neutron-scattering facilities at Argonne, Los Alamos, Brookhaven and Oak Ridge. The 1982 panel was asked by DOE to take a second look at the two spallation facilities last summer before any irreversible step was taken to begin the phase-out of IPNS I. The resulting second Brinkman report, in addition to recommending strongly that funds be sought to keep IPNS I running through fiscal 1984, expressed concern "that the experimental area at the WNR is small, and user accessibility is limited. We consider it essential that an expansion of the experimental area at Los Alamos be undertaken, making it suitable for a national facility of this magnitude."



Position-sensitive neutron detector developed at Argonne for the single-crystal diffractometer at the IPNS I spallation source. A 7×7 array of photomultipliers is behind a panel of Li⁶-loaded glass (not shown). The position of a neutron interacting in the glass is determined with a spatial resolution of 3 mm.

The expansion recommended by the second Brinkman report will require the approval and funding of additional construction.

Charles Bowman, associate physics-division leader at Los Alamos, told us that a number of improvements are under consideration for the proton storage-ring facility, including the addition of six new neutron drift tubes—making a total of eleven for neutron-scattering research. Plans are under study for increasing the size and shielding of the experimental area, he told us. "We believe that the new storage ring can provide the basis for a world-class pulsed-neutron facility."

"We're generally pleased with the recommendations of the second Brinkman panel," says John Carpenter, technical director of the IPNS program. "We would have preferred that the next generation of neutron-scattering research remain at Argonne, which has over the years developed a strong, broad-based materials-science effort," he told us. "What we'd most like to see now is a continuation of the IPNS I until the Los Alamos facility becomes available to the community of experimenters in 1986."

"First and foremost" among the conclusions of the Brinkman report is "that neutron-scattering research is an indispensable component of condensed-matter physics, polymer science, materials research and . . . molecular biology." Outside the United States the premier steady-state neutron scattering source is the Laue-Langevin Institute's high-flux reactor at Grenoble (PHYSICS TODAY, June 1980, page 21). The Rutherford Laboratory's SNS pulsed spallation facility, scheduled for completion in 1984, will provide a neutron intensity comparable to that of the Los Alamos storage-ring facility. —BMS