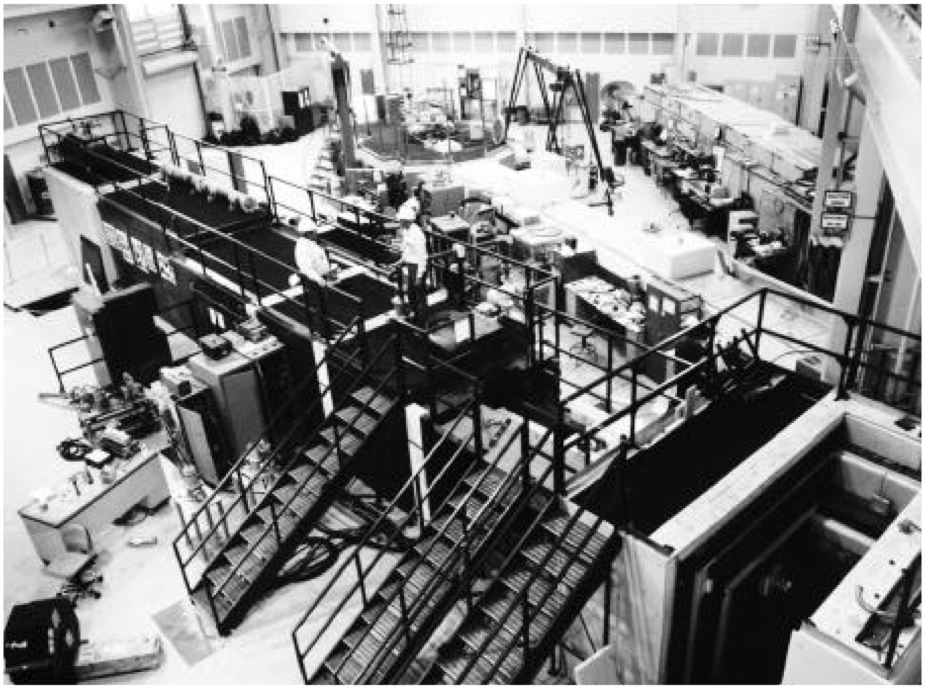


Experimental hall at LANSCE

NEUTRONS in our future

a proposed high-flux spallation neutron source

Roger Pynn

There is a paradigm in scientific research that repeats itself continually—the discovery that earned yesterday's Nobel Prize becomes the tool for today's research. Take x rays, lasers, and transistors, for example. Each was worth a Nobel in its day, and each is now found not only in almost every research laboratory but also in hospitals, supermarkets, and homes. The same paradigm applies to neutrons. Discovered by James Chadwick in 1932, these neutral particles were the stuff of esoteric research until fast fission and politics combined to make them central players in the Los Alamos story. Nuclear reactions in which neutrons participate are at the heart of all of the nuclear weapons designed here and elsewhere. Other neutron reactions—those in which neutrons are scattered rather than absorbed by nuclei—are the basis for the use of neutrons as probes of the structures of materials. That area of research, referred to simply as neutron scattering, is an important part of today's scientific agenda, which stresses industrial competitiveness and quality of life. To design new and improved materials for industrial applications, scientists build on their understanding of existing materials, a large part of which comes from information about their structures. Neutron scattering provides that information, often in situations where other techniques fail.

Successful neutron-scattering experiments require large number of neutrons to be directed at a sample because only a small fraction of the neutrons are scattered. The first neutron sources that were sufficiently intense for such experiments were nuclear reactors, and neutron scattering began as a parasitic activity at research reactors that were built in the 1950s to obtain data for nuclear-power programs. To this day the most productive neutron-scattering program is to be found at a reactor—the Institut Laue Langevin (ILL) in Grenoble, France. However, the situation is changing. A newer technique for producing neutrons at proton accelerators rather than nuclear reactors is fast becoming competitive. The technique, proton-induced spallation of heavy-metal nuclei, is currently the basis of the neutron source at the Laboratory's Manuel Lujan, Jr. Neutron Scattering Center and will remain the basis of a more intense neutron source that the Laboratory hopes to build. An upgrade of the LAMPF proton accelerator will make the more intense neutron source possible—which brings us back once more to our paradigm. LAMPF was built more than twenty years ago to study nuclear reactions that involve energetic protons or pions. Now, one of those reactions, proton-induced spallation, may be the basis for a new neutron-scattering facility.

The success of neutron scattering and its continuing importance are a result of several properties of the neutron. Because of its neutrality and the weakness of its interactions with matter, the neutron—unlike x rays or light— can penetrate deeply into solids and liquids and provide information about bulk, as opposed to surface, structure. In addition, because neutrons are scattered by both the nuclei and the unpaired electrons in matter, they provide information about both atomic and magnetic structure. The thermal neutrons generated by nuclear reactors or spallation sources have energies that are comparable to those of vibrating or diffusing atoms in solids. Therefore neutrons can probe not only the equilibrium positions of atoms in solids but also temporal structural changes. Because the neutron-scattering power of atomic nuclei varies erratically and often considerably with atomic number, neutrons can often distinguish between neighboring elements and can easily distinguish the lightest element, hydrogen, even in the presence of much heavier elements. The latter property makes neutrons a particularly powerful probe of biological molecules and man-made polymers, both of which contain substantial amounts of hydrogen.

For more than forty years neutron scattering has played an indispensable role in studies of condensed matter, providing essential information about materials as different as antiferromagnets, ribosomes, and shape-memory alloys. Often the information has been unobtainable by other means. Even a partial list of contributions from the past decade is impressive. During that period neutron scattering revealed the structure of the first high-temperature superconductors; the structure and excitations of buckminsterfullerenes, or bucky balls; the conformation of molecules in a polymer melt; the interfacial structure of artificially produced polymeric and magnetic layers; the structure and dynamics of new catalysts; the spin dynamics of highly correlated electron systems; and the condensate fraction in superfluid helium. It is safe to say that a large part of the conceptual and theoretical underpinning of the modern theory of solids would be unverified and incomplete without neutron scattering. And without that knowledge our current technology could not exist.

LANSCE has made its share of contributions during the five years it has been operating. The discovery by Gregory J. Kubas of the Laboratory's Inorganic and Structural Chemistry Group that certain metal complexes can coordinate molecular hydrogen is widely regarded as one of the most significant developments of the 1980s in inorganic chemistry. Studies at LANSCE of the vibrational and rotational dynamics of those dihydrogen ligands have provided insight into the nature of this unique chemical bond—the first known example of stable intermolecular coordination of a sigma bond to a metal. The system mimics a catalytic reaction "frozen" in an intermediate state of a type that is usually too ephemeral to study and understand. The dihydrogen ligand is important in catalysis because it can easily exchange hydrogen with other ligands in a complex. It is conceivable, for example, that hydrogen could be added to other ligands such as ethylene (catalytic hydrogenation) at a much lower cost in energy than the 104 kilocalories per mole required to break the hydrogen-hydrogen bond of uncoordinated molecular hydrogen.

Polymers and other macromolecules absorbed at solid or fluid surfaces have many applications to a wide variety of technologies. They are a means for achieving colloidal stabilization in water-treatment schemes, ceramic processing, inks, and fuels; they are used for mechanical protection of solids against friction and wear in motors and computer disks; and surface-active molecules at liquid-liquid interfaces are used to clean up oil spills and to enhance emulsification and blending. The variation of polymer density close to an absorbing surface had been studied theoretically but was difficult to study experimentally until neutron reflection provided the answer. Work at LANSCE verified theoretical predictions for the profile of the "polymer brush" formed by the stretching of polymer molecules away from a solid surface into a surrounding fluid and provided a characterization of the "polymer mushrooms" that occur as the grafting density of the absorbed polymers (the number of attached polymers per unit area) is decreased.

As the transportation industry struggles to improve fuel efficiency, it is turning increasingly to new composite materials—such as aluminum reinforced with silicon-carbide particles—that provide the dual advantages of strength and lightness. To understand the mechanisms of failure of such materials and to assess their lifetimes in real components, it is important to understand the residual stresses induced in the materials during fabrication. Depending on their distribution, such stresses can be devastating—aircraft fuselages have disintegrated in flight and railroad tracks have cracked and caused train crashes—or beneficial—wine barrels have been held together by metal hoops for centuries. Unfortunately no conventional technique for measuring residual stress, such as strain-gauge sectioning or hole drilling, is truly nondestructive. Over the last five years, neutron diffraction has proved to be a unique, nondestructive alternative and has been systematically exploited at LANSCE. Our work on composite materials has allowed sophisticated computer models for stresses—residual stresses as well as stress induced by applied load—to be verified and, in some cases, improved.

In spite of the successes and acknowledged importance of neutron scattering, the technique is on the verge of extinction in this country, leadership having passed to our European colleagues over a decade ago. With one exception (the neutron source at the National Institute of Standards and Technology), all the neutron sources in the U.S. that support neutron-scattering programs are run by the Department of Energy. Those sources are old and, by modern standards, poorly instrumented. The high-flux reactors at Brookhaven and Oak Ridge national laboratories may reach the end of their useful lives before the end of this decade. A pulsed spallation source at Argonne National Laboratory provides only one-tenth of the intensity of the ISIS facility at the University of Oxford. The LANSCE source has a peak neutron flux that is slightly higher than the ISIS source but has suffered from poor reliability and short annual operating periods. The problems of LANSCE have been exacerbated by constant erosion of the operating budget of LAMPF over the past five years. LANSCE is now threatened with closure because LAMPF is no longer the highest priority of the nuclear-physics community. Without a new neutron source, U.S. researchers will not remain competitive. Fundamental research as well as technology will suffer.

Nor are our competitors standing still. A consortium of European laboratories has proposed a design study for an advanced spallation source (the European Spallation Source, or ESS) that would provide capabilities well beyond those available at the ILL. The proposed ESS—consisting of a high-energy linac and an accumulator ring—looks very much like an upgraded version of LAMPF and the Proton Storage Ring. It is ironic that just as the Laboratory's spallation source faces shutdown, the Europeans are realizing that spallation sources are the way of the future.

Recognizing the national need for new neutron-scattering capabilities, as well as the value of existing infrastructure at LAMPF and the strength of its expertise in neutron scattering, Los Alamos National Laboratory has proposed the construction of a new pulsed spallation source with an initial power of 1 megawatt (the power of the present LANSCE source is 60 kilowatts) and a possible future power of 5 megawatts. On August 19, 1992, Laboratory Director Sig Hecker announced the proposal to a visiting review committee, noting that the Laboratory wants to change the emphasis of research at its 800-MeV linac from nuclear physics to neutron scattering. The committee Hecker addressed was chaired by Walter Kohn of the University of California, Santa Barbara, and had been charged by Will Happer, head of the DOE's Office of Energy Research, to examine the relative merits of reactor and spallation sources for the country's future neutron-scattering program. After several contentious weeks, the committee finally concluded that the country would be best served if two new sources—one of each type—could be built.

Since its beginning in 1987, LANSCE has been operated as a National User Facility, open to scientists from industry, academia, and other national laboratories. Experimental proposals submitted by potential users are peer-reviewed to ensure that the best use is made of the facility. Most of the national laboratories host a user facility of some sort in fulfillment of one of the DOE's most important missions in the area of basic research. The new pulsed source proposed by the Laboratory will remain a user facility, and the community of users will define the facility specifications that best suit its needs. Of course, we have an idea of the facility we would like to build—it is described below—but it is important to recognize that the final design parameters will come from the users rather than from the Laboratory.

A new spallation source at the Laboratory will make use of a number of existing LAMPF assets that would be expensive to reproduce elsewhere and are very appropriate as part of a modern accelerator complex. The 700-meter-long shielded tunnel that contains the present linac will remain, as will buildings, cooling towers, 30 megawatts of site electrical power, and a 600-meter part of the existing accelerator called a coupled-cavity linac. The latter is basically a copper pipe—albeit of somewhat exotic design—that is not expected to wear out.

All of the high-tech parts of the proposed accelerator complex will be new and will take full advantage of accelerator technology developed here and elsewhere as part of the Strategic Defense Initiative. Our present reference design calls for injection of 800-MeV protons from the upgraded linac into an accumulator ring that is similar in concept to the the existing Proton Storage Ring. However, we are studying an option that would increase the proton energy and, perhaps, permit an easier upgrade to 5 megawatts of beam power in the future.

The new accelerator complex will produce 60 proton pulses per second, each of about 0.5 microsecond in duration, and distribute them between two neutron-production targets. One target will receive 40 pulses per second and the other 20 pulses per second. We expect the 40-hertz target to provide about five times the average neutron flux generated by the ISIS source. Coupled cold moderators at the 20-hertz target will give twenty-five times the peak flux of either the ISIS or the present LANSCE source. There will be room for between twelve and sixteen beam lines around each target.

What does all this buy us? How does it compare with the ILL, for example? This question was answered by a group of European and American neutron-instrumentation experts who advised the Kohn panel. That eminent group concluded that a 1-megawatt pulsed spallation source could duplicate the capabilities of the ILL *and* provide facilities that exceed those at the ILL for experiments requiring the intense high-energy neutron beams produced by spallation. In other words, a 1-megawatt spallation source would give the U.S. the same capability as the ILL *plus* the obvious advantages over ISIS. Such a source would also be complementary to the reactor—the so-called Advanced Neutron Source—that the DOE proposes to build at Oak Ridge National Laboratory. Although there is a large area of overlap in the capabilities of these two sources, each has unique strengths.

Any prediction of the scientific impact of a 1-megawatt spallation source is bound to be incomplete, at best. Nevertheless, the impact is fairly obvious in those areas that are extrapolations of current research. For example, many experiments are beyond our current capabilities because samples of sufficient size are not available. Neutron scattering is inherently a signal-limited technique because, as mentioned above, only a small fraction of the neutrons incident on a sample are scattered. If the sample is not large enough, the informative scattered neutrons—the signal—cannot be discriminated from background neutrons that have suffered spurious scattering processes. One way of overcoming this limitation is to increase the flux of incident neutrons as our new source is designed to do. Such a source would make many important experiments possible. For example, we would be able to probe the collective excitations of high-temperature superconductors and fullerenes and perhaps understand the bases for their bizarre properties. Experiments of this sort now require single crystals of a size that cannot be grown. The problem of sample size is even more critical in structural biology, where the structures of only about one in every two hundred interesting proteins are now accessible to neutron scattering—sufficiently large crystals of the others just cannot be produced. Although some improvement in sample size is likely in some instances, there are other areas—the study of interfacial structure by neutron reflection, for example—where the scattering volume is inherently small and will always remain so. For such systems the only way forward is through the use of neutron sources with higher flux.

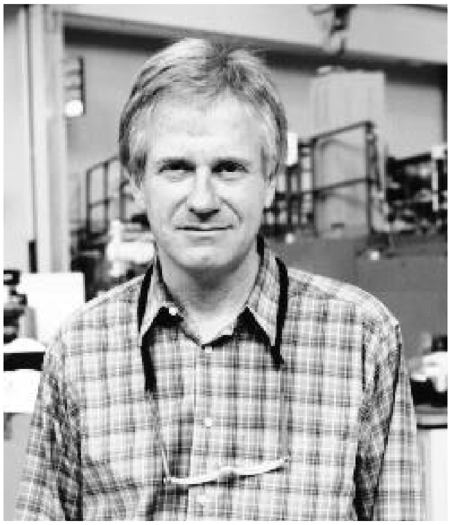
Higher-flux sources will also offer scientists the possibility to study structures as they evolve over time. Examples include changes in the structure of interfaces during corrosion and of electrolytes during battery discharge; phase transformations induced by propagating shock waves; and conformational changes of polymers during extrusion molding. Presently such experiments are restricted to model systems that change relatively slowly with time or to systems that can be arrested or cycled repeatedly. Examining the change in structure of a catalyst during its active phase, for example, is beyond current capabilities because the entire reaction is completed in a time that is much shorter than that needed for a neutron measurement.

The techniques used for neutron scattering at high-flux pulsed sources are well adapted to neutron-scattering experiments in which samples are subjected to high pressures or high magnetic fields. The equipment required to achieve high pressures would fail if it had to have large windows to let neutrons in and out, and high magnetic fields can be maintained only for short periods. Powder-diffraction measurements at

LANSCE have already been made at pressures above 100 kilobars, a pressure that is three or four times higher than has been achieved by similar experiments elsewhere in the country. Achieving still higher pressures, such as those needed to mimic some geological conditions, will require the use of smaller samples and, concomitantly, more powerful neutron sources.

One of the most exciting new capabilities offered by a 1-megawatt pulsed source couples the characteristics of the source with the great progress that has been made in computer science over the past two decades. Twenty-five years ago, neutron spectrometers were deliberately designed to avoid collecting too much data—it would have been just too confusing to the poor scientists! Spectrometers were designed to focus on phenomena that occurred over a small range of length and time scales and to ignore the rest. Although that approach delayed some discoveries a decade or two, it worked reasonably well for simple samples, especially those that could be grown in the form of single crystals. Unfortunately, many of the complex materials of interest today—both in materials science and structural biology—are interesting precisely because they have structure on a wide variety of length scales. Examples range from DNA molecules packaged as chromatin to the fractal structures found in porous media. To study such materials with neutrons requires spectrometers with access to a large range of length scales, a feature provided quite naturally by pulsed sources. However, to find some meaning in the vast quantities of information obtained by such spectrometers requires the speed of modern computers and the wizardry of modern techniques for data manipulation and display. The payoff could be immense, however. One can easily imagine a neutron spectrometer at a new pulsed source with 1 million or 10 million parallel information channels instead of just one.

Although this article has focused on neutron scattering, we expect that a new spallation source at the Laboratory would support other types of research as well. Indeed, it would be indefensible from the taxpayers' point of view *not* to exploit synergistic uses of the facility, some of which might help to resolve important issues in areas such as the management of radioactive waste. And there are exciting experiments in basic physics and nuclear-physics research to be done with neutrons, as well as complementary investigations of materials by muon spin resonance. Experiments with ultracold neutrons—those with velocities of only a few meters per second—can accurately measure the lifetime of the neutron and determine whether it has an electric dipole moment. Both of those properties are important inputs to the standard model used to understand our universe. High-power spallation sources also have practical applications. They can produce neutron-poor radioisotopes, many of which have become indispensable to modern nuclear medicine; they can be used to study radiation damage of materials in regimes that are relevant to fusion-energy systems; and they are the basis for many transmutation schemes that have been proposed to solve problems ranging from the production of tritium to the destruction of long-lived fission products and plutonium from the weapons stockpile. We envisage that those other types of research could be carried out without jeopardizing the primary mission of neutron scattering. The prospects of such a multidisciplinary research facility are indeed exciting and a fitting continuation of Los Alamos expertise in the science of neutrons. ■



Roger Pynn was born and educated in England. He received his M.A. from the University of Cambridge in 1966 and his Ph.D. in neutron scattering, also from the University of Cambridge, in 1969. He was a Royal Society European Fellow to Sweden in 1970; he did two years of postdoctoral research in Norway; and then he was an associate physicist for two years at Brookhaven National Laboratory. After spending eleven years at the world's leading center for neutron scattering, the Institut Laue Langevin in Grenoble, France, Pynn was appointed director of the Laboratory's Manuel Lujan, Jr. Neutron Scattering Center. He served as a member of the Kohn panel whose deliberations are described in this article.