Pulsed spallation neutron sources

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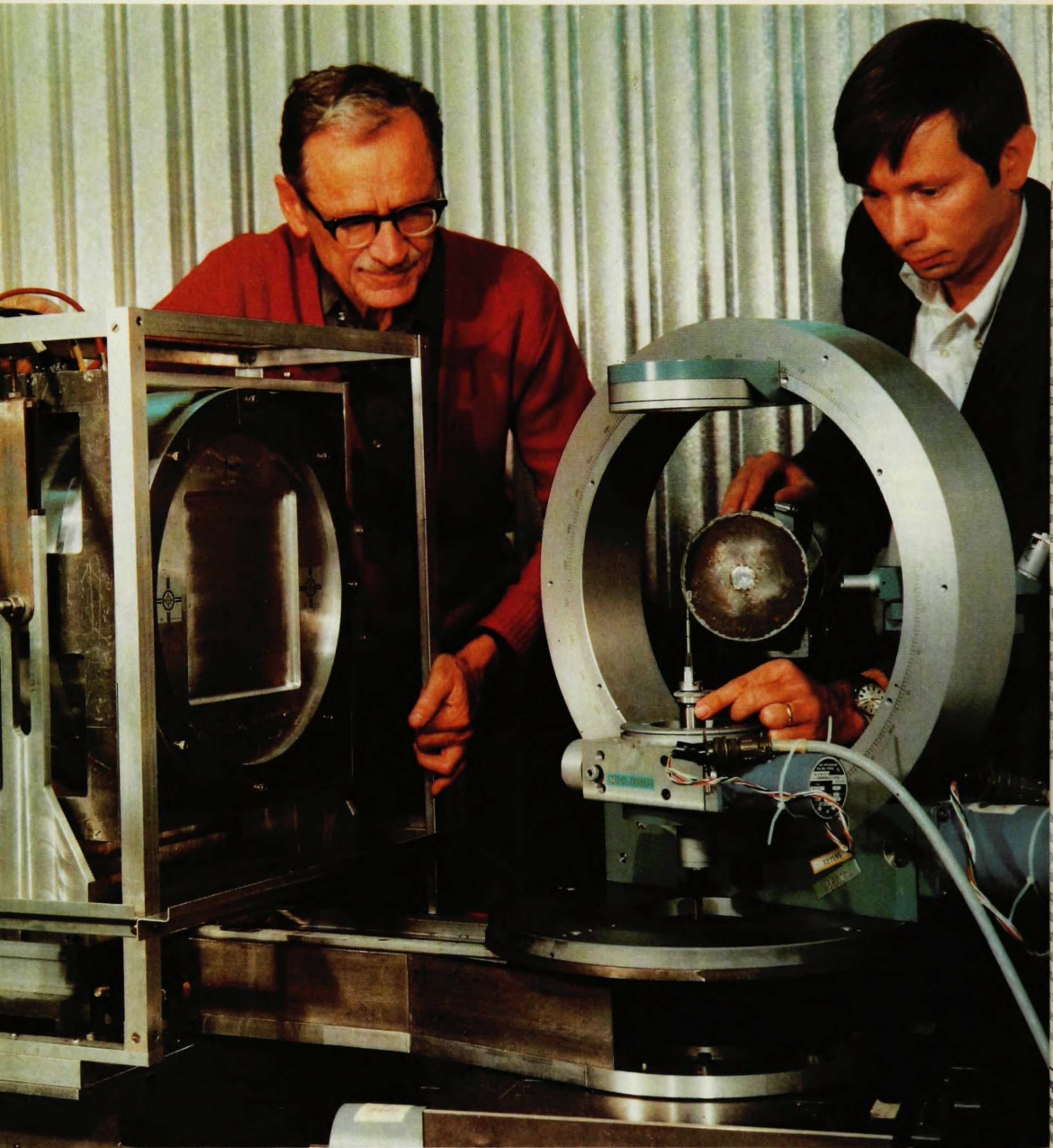

These new, intense neutron sources take up where high-flux reactors leave off, making possible a host of new structural and dynamic studies as well as neutron-irradiation investigations.

The discovery of the neutron in 1932, which indirectly led to the potentially catastrophic development of nuclear weapons on the one hand and to the beneficial development of nuclear-generated electricity on the other, has had another, less well-known, but scientifically important consequence: the provision of a uniquely sensitive and pervasive tool for probing condensed matter. Because they are neutral and because they interact mainly with nuclei, neutrons can penetrate into bulk material and provide information that is difficult to obtain with x rays or charged particles. Until recently, nuclear reactors provided the highest-intensity neutron sources. But lately another kind of source is promising to overcome the limitations of reactors. In these sources high-energy protons are made to collide with heavy nuclei, splitting off a large number of neutrons. The process is rather like making chips fly by hitting a rock with a hammer—what the geologists call "spallation." These neutron spallation sources together with equipment such as that shown in figure 1 have already begun to provide us with useful information accessible in no other way.

Neutron scattering

Within four years after James Chadwick's discovery, it was demonstrated that neutrons from a radium–beryllium source could be diffracted by the periodic lattice of atoms in a crystal because, like x rays, their de Broglie wave vector **k** was similar in magnitude to the reciprocal lattice vectors of the crystal:

$$k = mv/h \approx 2\pi/a \qquad (1)$$

where m and v are the neutron mass and velocity and a is a lattice parameter, typically a few Angstrom units in magnitude. The possibility of diffraction has important practical consequences, because the interaction of neutrons with matter is so different from that of x rays that one can use neutron diffraction to obtain structural information inaccessible to x-ray techniques. Already by 1946 these unique properties were being exploited in diffraction studies at the Argonne and Oak Ridge nuclear reactors.[1]

It was also realized very early that neutrons had another unique property that expanded the scope of the information derived from scattering experiments: Not only is the wavelength of thermal neutrons well matched to the spacings between atoms, but also their kinetic energy is near as the energy of the collective excitations in condensed matter:

$$E = \tfrac{1}{2}mv^2 \sim h\omega \qquad (2)$$

where ω is a lattice vibration frequency. Typically $h\omega$ has a magnitude of a few hundredths of an electron volt, corresponding to a frequency of a few times 10^{13} radians/sec. Thus, one can obtain dynamical information from inelastic scattering by simultaneously measuring momentum (or wavevector) and energy transfer in the scattering process. The extra variable analyzed in the scattering process made higher-powered neutron sources necessary, but even so, this technique was the basis of several experimental programs[2] by the late 1950's.

Because neutrons have no difficulty in penetrating the Coulomb barrier of the nucleus, neutron scattering varies strongly and irregularly with both the nuclear charge and mass. One can thus use neutrons to distinguish light elements and different isotopes of the same element. Furthermore, neutrons, because of their magnetic moment, scatter from unpaired electrons and offer a unique probe of magnetism in materials.

In summary, then, neutron scattering provides a number of advantages for investigating matter:

- They penetrate into the bulk of the sample
- They have a wavelength and energy well matched to atomic spacings and excitation energies in matter
- Their scattering length varies irregularly with A and Z, so they can distinguish elements and isotopes
- Their scattering from nuclei provides direct structural and dynamical information without uncertainties about their interactions
- Their scattering from unpaired electrons provides magnetic information

The technique now has come of age; it is contributing to many areas of solid- and liquid-state physics, metallurgy, chemistry and biology.[3] This development probably reached a plateau at the trinational Institut Laue–Langevin in Grenoble, France, in 1972, with the commissioning of a $60-million high-flux research reactor with an unprecedented array of special beam facilities and neutron instrumentation. We use the word "plateau" deliberately, for it appears prohibitively expensive to build reactors with fluxes much higher than those produced at the Grenoble reactor. At the same time the particle currents it produces are still orders of magnitude weaker than those available with other types of radiation (x rays, light, electrons etc.) and far below what is required for important new classes of experiment (see, "We need more intense neutron sources," by R. M. Brugger, PHYSICS TODAY, December 1968).

As a result of this impasse, the development of a new type of experimental facility called the "pulsed spallation neutron source"[4] is generating much interest around the world. Not only does this source offer a way of breaking through the flux barrier of the high-flux reactors, it also has other totally new features that promise to open up areas hardly explored with the steady-state neutron sources.

The pulsed-spallation concept

Figure 2 illustrates a typical pulsed spallation neutron source. Negative hydrogen ions are accelerated to an energy of about 10^8 eV and then stripped of their electrons; the remaining protons are injected into a rapid-cycling synchrotron. The negative-ion injection enables one to tailor the injected beam precisely to fit the acceptance of the accelerator and to overcome limitations on the brightness of the primary ion source. The protons are then accelerated to about 10^9 eV and ejected in a short burst towards a target where they excite heavy nuclei, with consequent emission of fast neutrons (spallation). Typically each proton produces 10–50 neutrons, depending on its energy and on the target material. These ejected neutrons can then be used directly

Single-crystal neutron diffractometer. Selmer Peterson and Arthur Reis are working on this time-of-flight diffractometer for use at the ZING-P' pulsed neutron source. Figure 1

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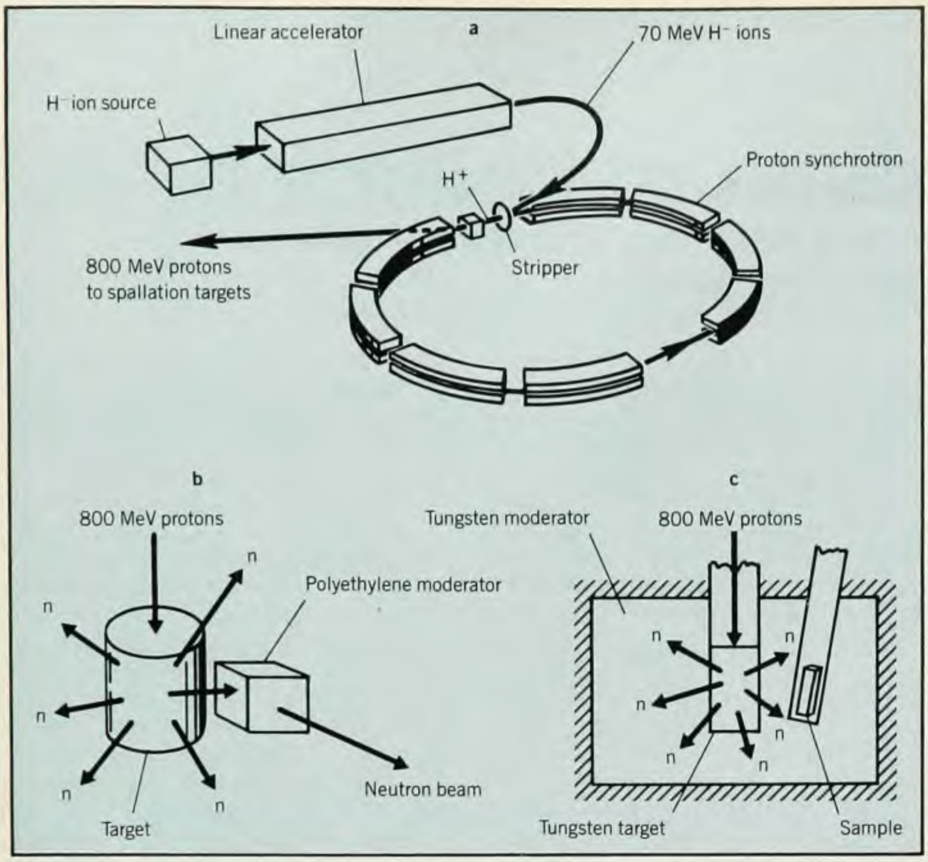


Pulsed spallation neutron source. The diagrams show the proton source (a) and the targets used to produce neutron pulses for scattering (b) and radiation-damage studies (c). Figure 2

or slowed down to the energy desired for a particular experiment by an appropriately designed moderator–reflector assembly.

The advantage of the pulsed source is that very intense bursts of neutrons can be produced in a very short time interval, so that the time-average rate of neutron production and the associated rate of heat production (which is the factor limiting the fluxes available in steady-state reactors) is very low. At the same time, one can use time-of-flight spectroscopic techniques at the pulsed sources, so that the high peak fluxes are as effective for a large class of experiments as are conventional techniques at a reactor with the same flux maintained continuously. To understand the reason, consider figure 3, which illustrates schematically different ways of carrying out a diffraction experiment. At a steady-state reactor one may use either the more conventional steady-state technique (left diagram) or the time-of-flight technique (right diagram). The two methods are equally efficient: the first uses a fraction of the wavelength spectrum all the time, the second uses all the spectrum a fraction of the time. In fact it can be shown that, for the same source intensity, resolution and detector solid angle, the two methods give identical count rates. Obviously, a time-of-flight experiment at a pulsed source is equivalent to one at a steady-state source with a chopper that produces the same pulse characteristics, thus the peak intensity of the pulsed source has the same effectiveness as the continuous intensity of the steady-state source as long as the duty cycle of one source matches the resolution of the other.

This method of producing neutrons is highly favorable in terms of minimizing the associated heat production. In this regard it is second only to fusion, which, however, cannot yet approach the neutron intensities required. Furthermore, accelerators of the type shown in figure 2 can produce the required neutron intensities with existing technology.

As well as providing very high effective neutron intensities, pulsed spallation sources have some characteristics quite different from reactor sources:

- They are especially powerful sources of *epithermal* neutrons (neutrons whose kinetic energy is in the range 0.1–10 eV, just above thermal energies) and thus open up a new range of energies and wavevectors for scattering experiments
- They are pulsed and thus provide capabilities for real-time studies and for measurements under extreme environmental conditions
- Their gamma-ray production rate is very low; they thus provide high fluxes of fast neutrons ideal for studies of radiation effects with low sample heating

Motivated by these powerful new features, several laboratories around the world have embarked on programs to design the build sources of this type. A selective listing is given in the table.

In the US the activities are centered at Argonne National Laboratory and Los Alamos Scientific Laboratory. At Argonne, the IPNS-I facility is under construction and scheduled for start-up in 1981; this will be an intermediate-intensity source, based on an existing accelerator, and will be dedicated for basic materials research. At Los Alamos, the activities are carried out (along with other research on nuclear physics and materials) at the WNR facility, which takes part of the LAMPF proton beam into a neutron-target area; a proton storage ring now being designed and constructed there will provide higher intensities by 1984. The most intense source now under construction, the SNS at the Rutherford Laboratory, is scheduled for completion in 1982. Proposals formulated by Argonne and Los Alamos, if approved, would provide the US with the world's foremost facilities in the late 1980's. Construction of a smaller-scale source in Tsukuba, Japan, is nearing completion.

As an example of the performance that can be achieved with pulsed spallation sources, figure 4 shows the peak neutron spectrum of the IPNS-II facility[5] proposed by Argonne compared with the performance of two sources at the Grenoble reactor. In addition to higher overall flux levels, the intensity of the pulsed spallation source in the eV region is orders of magnitude higher than that of either of the reactor sources. Also, as long as the pulse from the accelerator is sufficiently short, the neutron-pulse width decreases with increasing energy (figure 4) in such a way that the fractional energy-resolution is essentially constant. Together with the high intensity in the high-energy region, this provides the impressive performance in the epithermal region we mentioned earlier.

Structural investigations

Let us now survey the experiments that will be possible with the new sources.[6] We first consider structural measurements, made with the diffraction method illustrated in figure 3. This field naturally splits into studies of

- liquids and amorphous solids
- polycrystalline solids
- single crystals

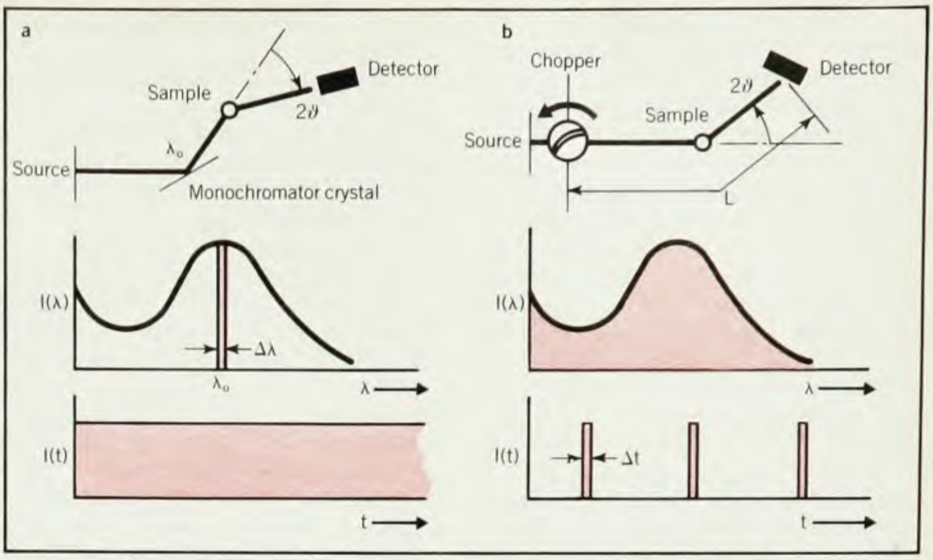
in increasing degree of structural order. The first of these structural types, *liquids and amorphous solids*, is especially attractive for study at pulsed sources: The abundant supply of epithermal neutrons makes it possible to measure accurately the radial distribution function, $g(r)$, which is essentially a Fourier transform of the observed diffraction pattern. For example, to measure $g(r)$ with a spatial resolution of 0.1 Å requires a neutron wavevector around 60 Å^{-1}, which corresponds to an energy of 8 eV. As figure 4 shows, only the pulsed sources have appreciable intensities in this range. The results shown in figure 5 illustrate how increasing wave vectors lead to enhanced fine structure in the measurement of $g(r)$ in the case of the amorphous alloy, $Pd_{0.8}Si_{0.2}$. Kenji Suzuki and his collaborators at the relatively low-level pulsed source at the electron linac of Tohoku University, Japan, have neatly exploited this feature. We can expect answers to many important questions about the structures of liquid metals and metal alloys, glassy solids, molten salts, liquid hydrocarbons and aqueous solutions as more powerful pulsed sources come into operation. In the case of aqueous solutions, for example, we could get information on the arrangement of water molecules around ions from a series of neutron studies involving different concentrations and H:D isotopic ratios.

The field of structural studies undertaken with *polycrystalline samples* is becoming increasingly important. Many new technologically interesting materials—such as many complex hydrides, tenary superconductors and one-dimensional conductors—are unavailable in single-crystal form, so that we cannot use more conventional single-crystal diffraction to investigate them. At the same time, Thomas Worlton, James Jorgensen, Robert Beyerlein and Daniel Decker are using profile analysis to increase greatly the scope and complexity of structural measurements possible with polycrystalline samples. Two factors make these measurements ideally suited to time-of-flight methods at a pulsed source: It is very simple to make use of large arrays of time-focused detectors, and, as we said, the resolution function is essentially constant across the entire spectrum of wavelengths, that is, it is constant for all plane spacings. A good example of the power of these techniques is a diffraction pattern of $LaNi_{4.5}Al_{0.5}D_6$ recently obtained at the low-level prototype ZING-P at Argonne by Jorgensen, Melvin Muller and Selmer Peterson, shown in figure 6.

A particular area where pulsed-source neutron diffraction will probably have a major impact is high-pressure neutron diffraction, where restricted access to the sample makes the time-of-flight, constant-angle technique especially favorable. Jorgensen, Louis Cartz and their colleagues have carried out several successful studies of oxide and nitride ceramics at pressures up to 40 kbar, using a time-of-flight diffraction method (and profile-refinement analysis) with a chopper at the CP-5 reactor at Argonne. With the fluxes available at a pulsed source such as IPNS-II, experimenters will be able to use multiple-anvil presses to carry out diffraction measurements at pressures of 100 kbar.

Many metallurgical systems exhibit static disorder, which gives rise to elastic diffuse scattering in addition to the Bragg diffraction peaks arising from the crystal lattice. Pulsed sources have two advantages for studying diffuse scattering: low background because the detector is simply not "on" when the fast neutrons—which are usually the major problem—arrive, and the ability to distinguish easily the elastic and inelastic components (by simply measuring the time it takes them to reach the detector). Especially important areas for study are: atomic arrangements in transition-metal alloys (in which x rays cannot distinguish between the constituents), measurements of the location of interstitials (such as carbon, nitrogen, oxygen and self-interstitials), and investigation of precipitate nucleation and growth at elevated temperatures.

One can perform structural measurements on *single crystals* at pulsed neutron sources by a simple extension of the x-ray diffraction techniques used by Max von Laue 70 years ago. Again, pulsed sources provide a great advantage over the earlier methods. Whereas in Laue's method it is impossible to separate the intensities of the reflections of different order from a given set of planes (that is, different values of n in the Bragg equation $n\lambda = 2d \sin\theta$), these are easily separated in time-of-flight neutron diffraction, because they arrive at different times at the detector. Although this technique is still under development, it appears that large time- and position-sensitive detectors will make it a powerful tool for studying single-crystal diffraction. The continuous spectrum of wavelengths available from a pulsed source—a spectrum that is, moreover, rich in neutrons with eV energies, makes anomalous dispersion of neutrons an attractive technique. This process relies on the change of coherent scattering amplitude associated with the absorption of the wave packet when the neutron energy approaches a resonance



Steady-state and time-of-flight techniques for neutron-diffraction studies. The diagram on the left shows a typical arrangement for steady-state measurements; the distribution of crystal-plane spacings, d, in the sample is determined by measuring the intensity as a function of the detector angle, 2θ, at a fixed wavelength λ_0. On the right is a typical arrangement for time-of-flight studies. The distribution of spacings d is determined by measuring the intensity as a function of the delay time, and hence wavelength λ (because $\lambda = ht/mL$), at a fixed angle 2θ. In both cases the plane spacing is given from λ by the Bragg condition $2d \sin\theta = \lambda$. Figure 3

Some pulsed spallation neutron sources world-wide

Source	Location	Frequency (Hz)	Proton pulse width (microsec)	Peak thermal neutron flux* (n/cm² sec)	Starting date
ZING-P	Argonne	10	0.15	5×10^{11}	1974
ZING-P′	Argonne	30	0.1	10^{14}	1977
IPNS-I	Argonne	45	0.1	7×10^{14}	1981
IPNS-II	Argonne	60	0.2	1.5×10^{16}	To be determined
WNR	Los Alamos	120	3.3	2×10^{13}	1977
WNR + PSR-I	Los Alamos	12–120	0.27	5×10^{15}†	1984
WNR + PSR-II	Los Alamos	48–120	0.27	1.6×10^{16}‡	To be determined
SNS	Rutherford Lab., UK	53	0.2	7.5×10^{15}	1982
KENS	Tsukuba, Japan	15	0.05	5×10^{14}	1980

* The fluxes in this table are standardized estimates (to permit a uniform comparison of the sources) and may differ slightly from values given in design reports

† At a frequency of 40 Hz.

‡ At a frequency of 48 Hz

level: In particular, it yields information about the phase of the waves scattered by each atom. Benno Schoenborn has suggested that this technique in conjunction with the use of isotopes such as Sm^{149} and Cd^{113} should give useful information for solving structures of large organic and biological molecules. Calculations performed by Peterson and others at Argonne indicate that complex protein structures with cells up to 100 Å in size could generally be determined within a few months at pulsed sources with the highest fluxes.

Dynamic investigations

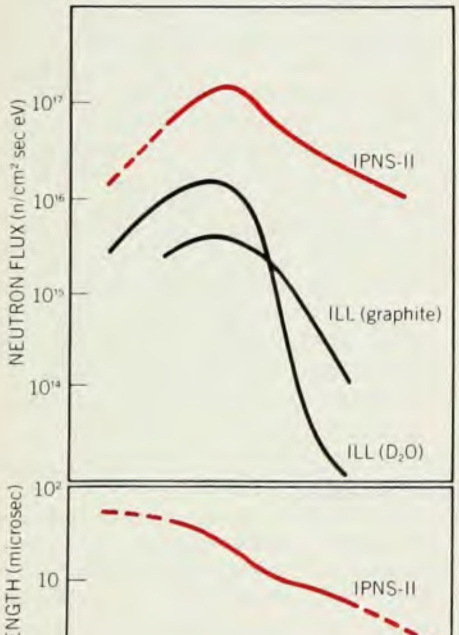
Inelastic neutron scattering—the simultaneous determination of the energy and momentum transfer in the scattering process—provides a unique method for studying the dynamics of condensed matter on a microscopic scale of space and time. This has been effectively exploited over the last two decades in experiments at the steady-state sources for several types of study:

- phonon and magnon spectra in crystals (see, for example, "Exploring phase transformations with neutron scattering" by John D. Axe and Gen Shirane, PHYSICS TODAY, September 1973)
- critical fluctuations in the neighborhood of magnetic and structural phase transitions
- diffusion of light interstitial atoms in solids
- molecular rotation and reorientations in plastic crystals
- crystal-field splittings and excitations associated with magnetic ions in solids
- collective excitations and diffusion in liquids and dense gases
- the dynamics of molecular monolayers adsorbed on certain substrates.

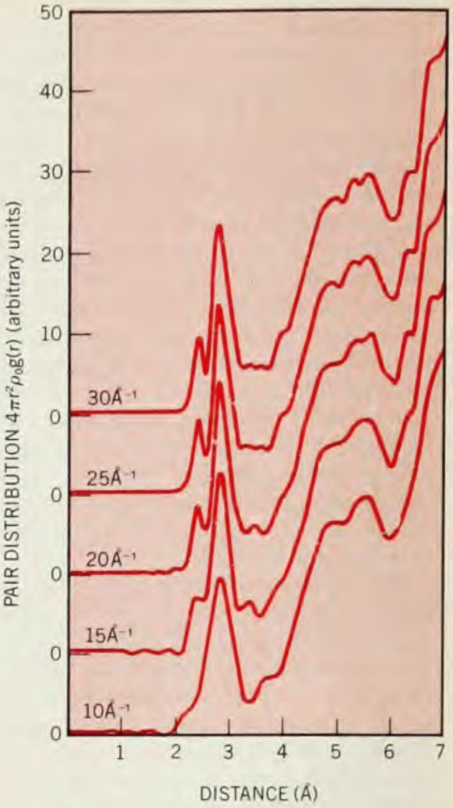
The pulsed spallation sources will be extremely powerful for many such studies, especially on systems with a lesser degree of order—disordered alloys, amorphous and polycrystalline solids, liquids, and polymers. They will make possible experiments on these systems that are not possible at current levels of intensity and resolution. This is because the time-of-flight technique for inelastic scattering (essentially, the method shown on the right in figure 3, with either the incident or final energy fixed) is well suited to measurements that require information over a large range of energy and wavevector transfers. The equivalent steady-state technique, on the other hand, is more suitable for deriving information for particular values of these variables. In addition, the copious supplies of epithermal neutrons at the pulsed sources will open up whole classes of experiments involving energy transfers from 0.1 to 10 electron volts, which are now not possible at all.

The dynamics of liquids and glasses is particularly suitable for study with time-of-flight techniques because the scattering function $S(Q,E)$ is continuous in the variables Q (wave-vector transfer) and E (energy transfer). For example, it will be possible to measure the vibrational densities of states of the tetrahedrally coordinated amorphous semiconductors (Ge, Si) and their alloys with hydrogen and other elements. Until now, it has not been possible to make definitive measurements of this kind because of limited intensities and sample quantities available.

In the case of fluid dynamics, it will be possible to make systematic high-resolution measurements of many different classes of liquids and dense gases. As an example, Kurt Sköld and Charles Pelizzari used the time-of-flight spectrometer at Argonne's CP-5 reactor to measure both the dispersion of zero-sound modes and the amplitude of the spin-fluctuation peak in liquid helium-3 at 1.2 K and 0.04 K. Because of the very high absorption of thermal neutrons by He^3, these measurements are now being made very close to the limits of resolution and intensity at which excitations in the spectrum can be observed. With the new pulsed sources, it will be possible to define the detailed structure of both the spin-fluctuation and zero-sound modes and to extend the measurements to long wavelengths, where connection with the Landau theory can be made. It should even be possible to extend these measurements into the superfluid regimes.



Neutron output of pulsed and steady-state sources. The upper graph shows the neutron flux available from Argonne's proposed IPNS-II source (with a CH_2 moderator at 300 K) and from the ILL reactor with two different moderators (graphite at 2000 K and D_2O at 300 K). The lower graph shows the duration of pulses (Δt in figure 3) available from IPNS-II as a function of the neutron energy. Figure 4



Radial distribution function for atom pairs in the amorphous alloy $Pd_{0.8}Si_{0.2}$. We show distribution functions derived from diffraction patterns extending to different values of maximum wavevector transfer, Q. The curves are labelled with the maximum value of Q used in the Fourier transform to obtain $g(r)$ from the scattering function $S(Q)$. Figure 5

The study of the dynamics of adsorbed molecules on surfaces is another field ideally suited to the application of a high-flux pulsed source to time-of-flight spectrometry, because the large-specific-area samples involved will generally be polycrystalline. By subtracting from the measured scattering from such a surface the corresponding scattering from a "clean" substrate, one can obtain the frequency spectrum of the adsorbed monolayer. Observation of the higher-frequency internal molecular modes of the adsorbed molecules and comparison with the corresponding values of the free molecules leads to information regarding the conformation and geometry of such molecules on the surface. Haskell Taub and his colleagues at the University of Missouri at Columbia have made some measurements of this type at reactors. However, the higher intensity of the pulsed sources, especially in the epithermal region (these modes often have energies of the order of 0.1 eV), will make them much more powerful for this type of measurement.

To obtain the ultimate resolution possible with the new pulsed sources, a group at Argonne has conceived a microvolt-resolution spectrometer.[5] Such an instrument combines the very high resolution of, for example, the back-scattering spectrometer at the Institut Laue–Langevin with the extended range in energy and momentum transfer available at time-of-flight spectrometers. With microvolt resolution one may be able to identify the "tunneling" or "reorientation" modes of various molecular subgroups in glasses that have been postulated to explain the anomalous linear variation with temperature of the specific heat at low temperatures.

The relative abundance of higher-energy neutrons in the pulsed sources has important implications in studying the dynamics of solids. The optical-phonon modes in many crystals have energies greater than 60 meV. These are difficult to study with conventional triple-axis spectrometers, owing to the rapid drop in the source spectrum for the necessary incident-neutron energies beyond 100 meV. An added difficulty stems from the poor reflectivity of crystal monochromators at these energies. An interesting example of an optical-phonon spectrum was actually studied at the first source of this kind ever set up, the ZING-P prototype at Argonne: Aneesur Rahman and his colleagues obtained the optical-phonon spectrum of palladium hydride, $PdH_{0.63}$. Comparing this with the phonon spectrum calculated from the dispersion curves measured by Mike Rowe and collaborators on a single crystal of $PdD_{0.63}$ at Oak Ridge, they found that the palladium–hydrogen force constant is some 20% stronger in the hydride than in the deuteride. This result, in turn, has been used in calculations by Dimitrios Papaconstantopoulos and Barry Klein to explain quantitatively the system's "inverse" isotope effect—that is, the heavier isotope has the higher superconducting transition temperature T_c. Their conclusion is that the high-frequency optical modes play an important role in the superconductivity of these and many other high-T_c systems.

As pulsed sources with more intensity become available, measurements of this kind can be extended to higher energies. At Argonne's newer prototype source, ZING-P′, Kent Crawford has seen at least three higher harmonics of the hydrogen vibrational frequency in ZrH_2 (see figure 7); the third harmonic corresponds to an energy transfer of 0.56 eV. While data of this kind have been obtained at the earlier electron-linac sources,[7] the fact that these measurements are possible at a prototype source and with a relatively short running time suggests that spallation sources will provide a powerful technique for studying these high-energy vibrational modes. In particular, careful comparison of the energy differences between successive levels will enable us to deduce the shape of the potential wells in which the hydrogen atoms reside.

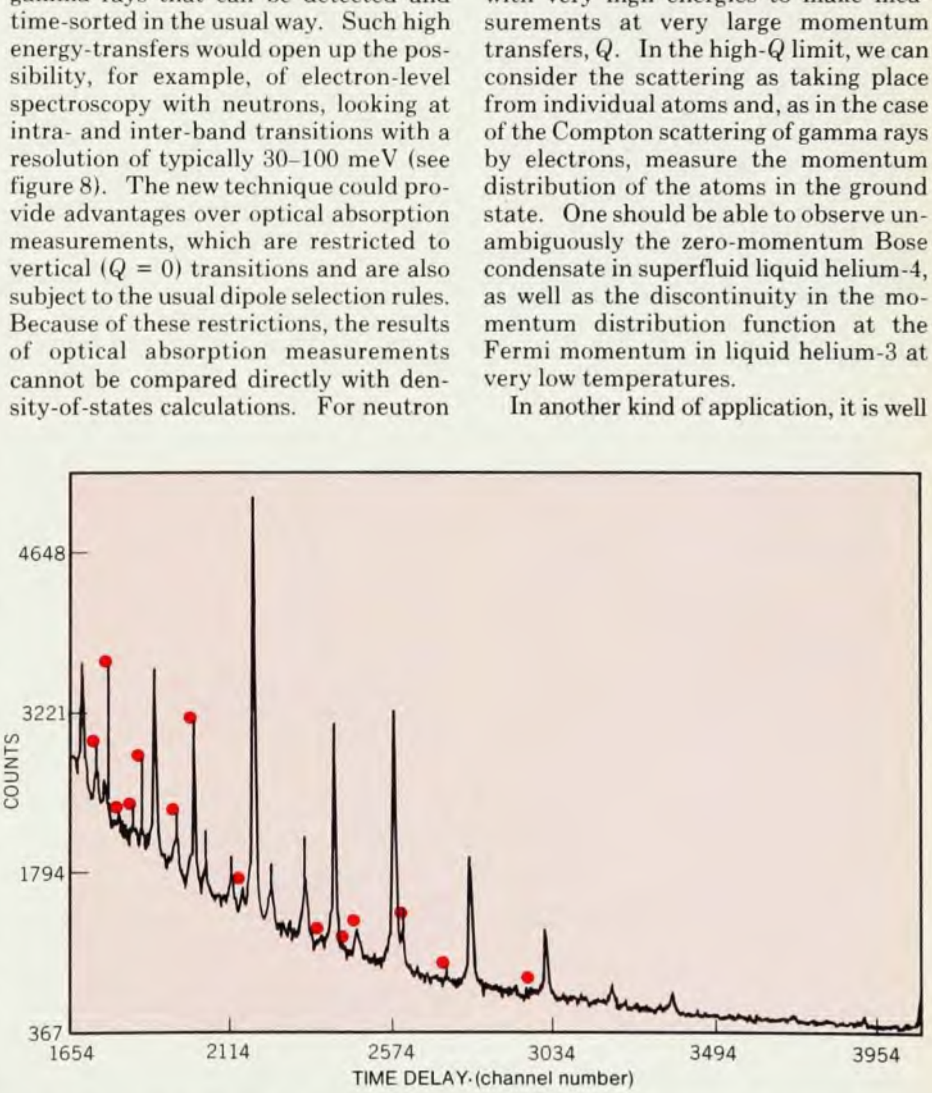
Even more severe limitations exist on experiments performed with steady-state reactors for another family of high-frequency excitations, spin excitations in solids with energies 60 meV and above. The limitations arise because magnetic scattering restricts wavevector transfers to be typically less than 5 $Å^{-1}$; beyond this value the form factor of the magnetic electrons becomes very small. To satisfy the kinematic restrictions imposed by the simultaneous requirement of small wavevector transfer and large energy transfer, one has to make measurements with small scattering angles and high incident energies, often in the range of 1–2 eV, for which pulsed sources become essential. One example is the study of the disappearance of the spin waves into the "Stoner continuum" of single-particle excitations in itinerant-electron antiferromagnets.

With pulsed sources one can consider experiments involving really high energy transfers (in neutron-scattering terms) up to 10 eV or so, perhaps using a resonance detector consisting of a foil of material such as U^{238}. This isotope has a sharp neutron-capture resonance at 6.67 eV and, after absorbing a neutron, emits gamma rays that can be detected and time-sorted in the usual way. Such high energy-transfers would open up the possibility, for example, of electron-level spectroscopy with neutrons, looking at intra- and inter-band transitions with a resolution of typically 30–100 meV (see figure 8). The new technique could provide advantages over optical absorption measurements, which are restricted to vertical ($Q = 0$) transitions and are also subject to the usual dipole selection rules. Because of these restrictions, the results of optical absorption measurements cannot be compared directly with density-of-states calculations. For neutron scattering, on the other hand, one can make measurements at large momentum transfers in such a way that the results give an average over the entire Brillouin zone. The scattering cross section is then proportional to the joint density of states of the electrons. This kind of neutron experiment would be particularly useful and relevant for the mixed-valence and actinide compounds with f-bands lying close to the Fermi energy. The cross sections would be especially favorable in such cases.

An additional advantage of the technique is that neutrons are not sensitive to the surface of the material and thus, as we have said, provide information about the state of the bulk material. Moreover, because neutron scattering probes the spin-response function while x-ray and electron scattering probe the density-response function, a comparison of the two should yield information of great interest to many-body theorists—specifically, the Q-dependence of the exchange enhancement of the spin-response function and of the screening of the density-response function.

Finally, we will be able to use neutrons with very high energies to make measurements at very large momentum transfers, Q. In the high-Q limit, we can consider the scattering as taking place from individual atoms and, as in the case of the Compton scattering of gamma rays by electrons, measure the momentum distribution of the atoms in the ground state. One should be able to observe unambiguously the zero-momentum Bose condensate in superfluid liquid helium-4, as well as the discontinuity in the momentum distribution function at the Fermi momentum in liquid helium-3 at very low temperatures.

In another kind of application, it is well

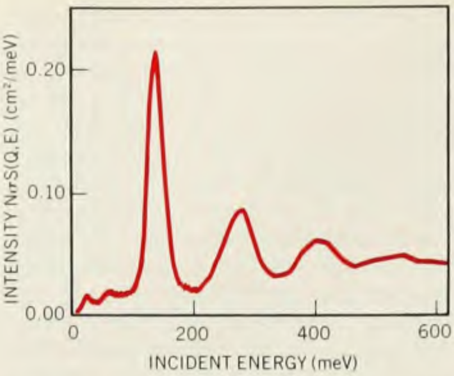


Diffraction pattern of $LaNi_{4.5}Al_{0.5}D_6$ taken at the ZING-P′ pulsed neutron source. The marks indicate peaks not observed in an equivalent measurement at a reactor. Figure 6 known that the interaction of energetic nucleons with solids produces substantial changes in the physical properties of these materials. In simplified terms, the bombarding particle collides with the host material, displacing the atoms from their equilibrium sites, and thereby creating vacant lattice sites and interstitial atoms. These simple defects can interact not only with their opposite kind to annihilate, but also with their own kind to form vacancy and interstitial clusters.

Radiation effects

It is important to realize that the energy, electronic charge, and mass of the bombarding particle are of great importance in determining the number and spatial distribution of defects. One extreme case is that of energetic-electron bombardment, where the energy transferred to the lattice atoms in most instances is small (less than twice the energy required to displace the atom). A single high-energy electron thus produces a single interstitial–vacancy pair; the overall result is a random distribution of isolated pairs of vacancies and interstitials. Bombardment with fast neutrons ($E \sim 10^6$ eV) on the other hand can, in a single collision, transfer an energy several orders of magnitude greater than is required to displace an atom. A fast neutron therefore produces a region of localized damage that contains a high density of vacancies and interstitials.

The understanding of neutron-irradiation effects is, of course, of great practical concern selecting materials for both fusion and fission reactors. One should not overlook, however, that these studies are also of great value in understanding basic defect interactions. The



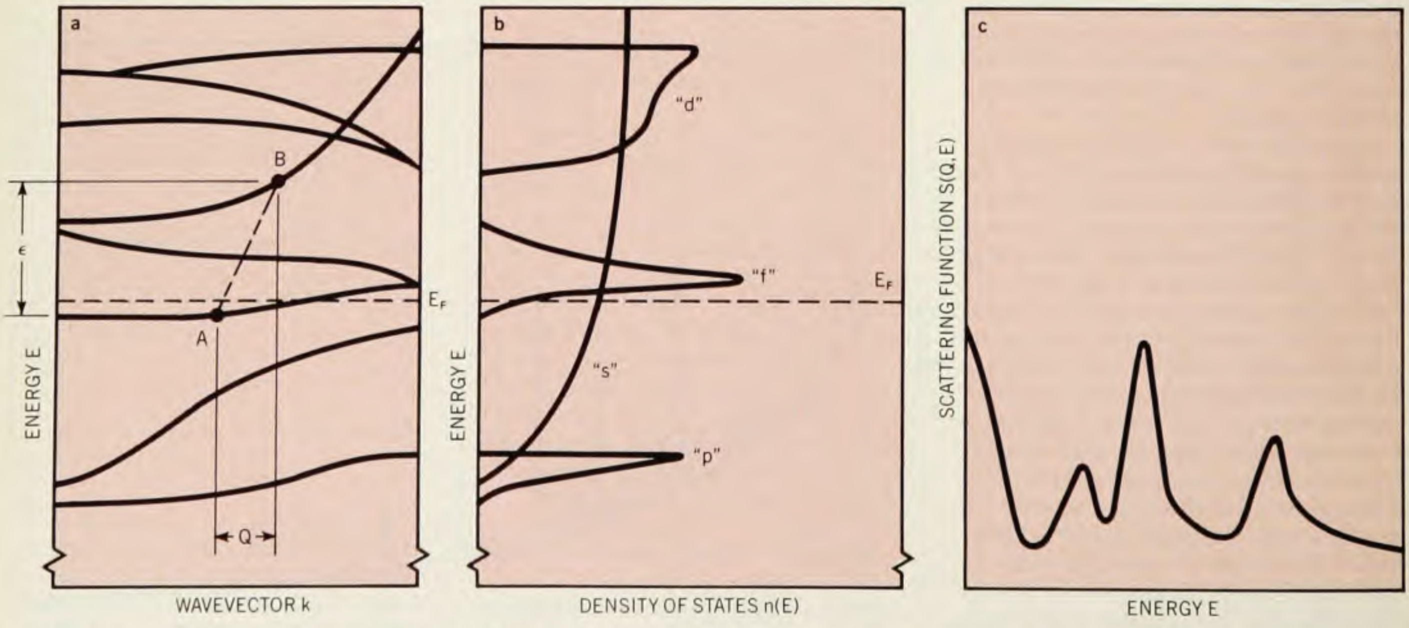
Inelastic neutron scattering from ZrH_2 at 300 K, measured at ZING-P′. The detection system fixes the final energy at 3 meV and the plot shows the scattered intensity (at 90° to the incident beam) as a function of the initial energy. The large peak at left is due to a hydrogen vibrational mode; the other peaks are from its harmonics. Figure 7

major areas that need greater understanding are:

- defect production as a function of neutron energy
- defect agglomeration and annihilation
- defect distributions
- thermal defect recovery
- defect–dislocation and defect–impurity interactions

In the past, fundamental neutron-irradiation effects have largely been studied in low- to medium-flux reactors, cheifly because the high nuclear heating rate encountered in high-flux reactors (as much as several hundred watts per gram, primarily from gamma-ray absorption) makes it difficult to control the experimental environment. *In-situ* basic studies have for the most part been restricted to fluxes of the order of 10^{13} neutrons/cm^2 sec with energies above 10^5 eV. Research reactors are large multipurpose machines serving a variety of functions besides studies of radiation effects. This lack of dedication makes it difficult to provide some of the features required specifically for research on radiation effects. One of the chief requirements is a well-characterized neutron spectrum of high energy and with a minimum of thermal neutrons. To meet this need, it has been necessary to sacrifice flux. Despite these drawbacks, research reactors have proven to be a valuable tool in radiation-effects research, and have provided many irradiation facilities with uniform and well characterized fluxes.

Spallation sources produce neutrons both by evaporation and by spallation. The former are emitted with a spectrum characteristic of fission neutrons, the latter with energies up to that of the bombarding particle. The number of evaporation neutrons increases with atomic weight of the target while the production of spallation neutrons is more or less independent of it; the total spectrum of emitted neutrons can therefore be varied by changing the target material. One of the features of the spallation sources is the very low emission of gamma rays during neutron emission, so that nuclear heating is very much smaller than for fission sources. This makes it possible to obtain good experimental control at high neutron fluxes. At the most intense pulsed sources, such as the proposed IPNS-II facility mentioned above, it should be possible to obtain a flux of about 10^{14}/cm^{2}sec fission-energy neutrons at sample positions, although the sources



Electron-level spectroscopy with neutrons. Graph (a) shows a typical band structure of a conductor; graph (b) shows the resulting density of states. A neutron that transfers energy ϵ wavevector Q to the crystal can induce transitions from A to B. When the conditions are such that the scattering averages over the entire Brillouin zone (powdered sample, large Q) one should see chiefly transitions between the peaks in the densities of states, so the scattering function should have the form shown in (c), due to the effective joint density of states. Figure 8 now being built will have somewhat lower fast-neutron fluxes—between 10^{12} and $10^{13}/\mathrm{cm}^2\mathrm{sec}$. The radiation-effects laboratories can be dedicated to just these studies and completely independent of the scattering target, and they can therefore be optimized for studies of radiation effects.

These unique features of spallation sources offer an excellent opportunity for increasing our knowledge in those areas of radiation effects listed above.[6] In particular, the potential for high fluxes combined with low nuclear heating and the ability to change the neutron spectrum, make low-temperature studies particularly exciting: They will allow new experiments on defect production as a function of neutron energy as well as experiments on radiation effects in superconductors. The large experimental spaces available greatly simplify measurements of mechanical properties, making it feasible to study radiation-defect–dislocation interactions. It is not yet clear to what extent the pulsed nature of the source is useful. With some improvements in high-speed voltage measurements, however, we can look forward to exciting experiments directly measuring lifetimes of defects. Such improvements appear to be entirely feasible over the next few years.

Other applications

The pulsed nature of spallation sources suggests other new experiments. For example, it is natural to consider neutron-scattering studies of samples under pulsed external conditions such as applied stress, magnetic and electric fields, laser excitation pulses, and so on. Two kinds of experiment are possible: In the first, one applies the pulsed condition at the same time as the neutron pulse arrives at the sample; the scattering thus gives information on the state of the sample under this condition. (An interesting example would be the structure of a molecular crystal in its excited-state configurations during irradiation with a laser pulse.) In the other case one applies the pulsed condition at various times prior to the arrival of the neutron pulse and studies the relaxation processes involved. Of course, for these experiments the relaxation times must have the right order of magnitude, say 10^{-5} to 10^{-2} sec.

These sources also appear to offer exciting possibilities for "neutron bottle experiments," in which ultra-cold neutrons (with velocities of 7 m/sec or less) are trapped in a container, due to total internal reflection at all angles from the walls. (See "Ultracold neutrons" by V. I. Luschikov, PHYSICS TODAY, June 1977, page 42.) The conventional way of producing such neutrons at a reactor is to trap the neutrons that emerge from it. At a pulsed source, one can take somewhat faster neutrons ($v \sim 400$ m/sec), which do not suffer such strong absorption during transport, and then slow them down by diffraction from a suitably chosen moving crystal synchronized to the source. A device of this kind is now being tested at the ZING-P′ source at Argonne. The parameter that determines the density of cold neutrons is the instantaneous phase-space density at the source, so pulsed spallation sources appear capable of providing much higher stored densities of ultracold neutrons than are now available. Such sources will permit us, for example, to search for the electric dipole moment of the neutron with an accuracy sufficient to measure the very small, but non-zero, value predicted by some recent theories of the weak interaction.

The examples outlined above are just a few of the many applications we foresee for pulsed spallation neutron sources. Many of the ideas, of course, are still in their infancy, especially those involving epithermal neutrons, which are essentially not available at the steady-state sources. As in other fields, it is likely that many surprises await us in regions of energy and momentum space that are still uncharted territory, and it may well be that some of the most important applications of these sources will become apparent only after the research has reached a certain level of maturity. In any case, there appears to be no doubt that spallation sources together with the existing high-flux steady-state reactors will ensure the importance and vitality of neutron research on condensed matter for some time to come.

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