PULSED NEUTRON SOURCES FOR CONDENSED-MATTER RESEARCH

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Introduction

Slow neutrons provide an important and unique probe of condensed matter. Since their wavelength is comparable to interatomic spacings, diffraction measurements give information about both atomic and magnetic structure, and since their kinetic energy is comparable to the energies of collective excitations, inelastic scattering measurements give information about the dynamics of these structures. The luminosity of neutron sources is much weaker than those of photons or electrons and thus the power of neutron techniques is heavily dependent on the strength of the available sources. The high-flux reactors at Brookhaven, Oak Ridge and Grenoble in France have led to important advances in solid- and liquid-state physics, metallurgy, chemistry and biology. Unfortunately, it appears prohibitively expensive as well as politically difficult to build a reactor with a neutron flux significantly higher than these, and thus the efforts around the world to provide higher neutron intensities are now focussed on accelerator-driven spallation sources in which neutrons are produced by bombardment of heavy metal targets with high energy protons. Spallation sources can be pulsed or quasi-continuous depending on the nature of the proton accelerator. Pulsed spallation sources, based on rapid cycling synchrotrons or storage rings, are especially attractive since a moderate average proton current (10^{-5} - 10^{-3} A) can produce high instantaneous neutron intensities which can be used with time-of-flight methods to provide powerful neutron spectrometers.

The Pulsed Spallation Concept

Fig. 1 illustrates a typical pulsed spallation neutron source. Negative hydrogen ions are accelerated to an energy of about 10^7 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 10^8 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 or slowed down to the energy desired for a particular experiment by an appropriately designed moderator-reflector assembly.

Fig. 1. Schematic diagram of a pulsed spallation neutron source showing the proton source (a) and the targets used to produce neutron pulses for scattering (b) and radiation-damage studies (c).

Table 1: Some Pulsed Spallation Neutron Sources World-Wide

Source	Location	Frequency (Hz)	Proton pulse width (microsec)	Peak thermal neutron flux (n/cm^2 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	5×10^{14}	1981
IPNS-II	Argonne	60	0.2	1.5×10^{16}	To be determined
WNR	Los Alamos	120	8	10^{14}	1977
WNR + PSR-I	Los Alamos	12	0.27	10^{16}	1986
WNR + PSR-II	Los Alamos	48	0.27	1.6×10^{16}	To be determined
SNS	Rutherford Lab., UK	53	0.2	5×10^{15}	1984
KENS	Tsukuba, Japan	15	0.05	10^{14}	1980

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 Table 1.

Structural Investigations

We now survey the experiments that will be possible with the new sources. We first consider structural measurements which split 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 $0.1 \text{ \AA}$ requires a neutron wavevector around $60 \text{ \AA}^{-1}$, which corresponds to an energy of 8 eV. Only pulsed sources have appreciable intensities in this range. K. Suzuki and his collaborators at the relatively low-level pulsed source at Tohoku University, Japan, have neatly exploited this feature.

The field of structural studies undertaken with polycrystalline samples is becoming increasingly important. Many new technologically interesting materials--such as complex metal hydrides, ternary 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 J. D. Jorgensen and collaborators are using profile analysis to increase greatly the scope and complexity of structural measurements possible with polycrystalline samples at pulsed neutron sources. 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 the resolution function is essentially constant across the entire spectrum of wavelengths, that is, constant for all plane spacings. A good example of the power of these techniques is a diffraction pattern of lithium-stabilized beta" alumina (Fig. 2) recently obtained at the ZING-P' prototype source at Argonne by W. L. Roth and collaborators. The material studied was an actual ceramic tubular structure to be used as a separator in a prototype sodium sulfur battery. Analysis of the diffraction data showed that the distribution of sodium atoms in the conduction planes is different from that in beta" aluminas stabilized with other atoms, presumably correlated with the superior conductivity and stability of the lithium-stabilized compound.

Fig. 2. Time-of-flight diffraction spectrum (four successive frames) from ceramic $\text{Na}^+\beta''\text{-alumina:Li}$. The solid curves represent profile refinement fits to the data. (W. L. Roth et al.)

One can perform structural measurements on single crystals at pulsed neutron sources by a simple extension of Laue x-ray diffraction techniques. Whereas in Laue's method it is impossible to separate the intensities of the reflections of different order from a given set of planes, these are easily separated in time-of-flight neutron diffraction because they arrive at different times in 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.

Dynamical 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. The pulsed

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 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.

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 interesting example of an optical-phonon spectrum is a measurement on $\text{VH}_{0.02}$ carried out by T. O. Brun at the ZING-P' prototype (Fig. 3). Despite the low H concentration, two vibrational modes are clearly identified which disappear on warming to room temperature.

Fig. 3. Double-differential scattering cross section (arbitrary units) from $\text{VH}_{0.02}$ at 30°K , with the scattering from pure vanadium subtracted. (T.O. Brun)

Other types of dynamical investigation for which the pulsed sources are especially advantage are spin excitations in solids with energies of 60 meV and above (where the restriction on momentum transfer caused by the magnetic form factor requires the scattering of high-energy neutrons at low angle), measurements of vibrational modes of molecules adsorbed on surfaces, and measurements of ground-state momentum distributions in quantum systems at large neutron momentum transfer.

In another kind of application, it is well 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.

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.

Spallation sources offer an excellent opportunity for increasing our knowledge of radiation effects. In particular, the potential for high fluxes combined with low nuclear heating 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. With improvements in high-speed voltage measurements, we can look forward to exciting experiments measuring lifetimes of defects following the neutron pulse.

Real-Time Studies

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 experiments 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. 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.

Resonance Neutron Radiography

Radiography has been for many years one of the important applications of neutron beams. A pulsed source offers a unique advantage for certain applications of radiography because the various energies can be readily separated by time-of-flight techniques. To demonstrate this method, G. H. Lander and collaborators have carried out a test experiment at the ZING-P' prototype. The target, consisting of foils of gold and indium, is shown in Fig. 4.

-Fig. 4. Target for resonance radiography test.

The detector was a new type of neutron-position detector developed by M. G. Strauss and R. Brenner consisting of a lithium glass scintillator and a photomultiplier assembly using the Anger coding method. Fig. 5 shows the spatial dependence of the neutrons detected in two different time channels corresponding to the In and Au positions, respectively. The two-dimensional histograms give clear indications of the extent of the two foils.

Fig. 5. Two-dimensional neutron radiograms measured in transmission through the target of Fig. 4 (G. H. Lander *et al.*)

Conclusion

The examples outlined above represent just a few of the applications we foresee for pulsed spallation neutron sources. As in other fields, it is likely that surprises await us in previously inaccessible regions of energy and momentum space that will be opened up by these new devices.

References

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