IEEE Transactions on Nuclear Science, Vol.NS-24, No.3, June 1977

APPLICATIONS OF PULSED NEUTRONS FROM A SPALLATION SOURCE

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Summary. This paper brings to a focus the major current limitation of the application of thermal neutron scattering in condensed matter research - source intensity. The pulsed spallation source provides a means of gaining an order of magnitude increase over the best steady-state reactors, while at the same time it opens up a much broader, and potentially very rich region of excitations to neutron scattering.

I. The Domain of Neutron Scattering - What Limits It?

Thermal neutron scattering began about 30 years ago. It is now an extremely important field of scientific research. The first apparatus which could legitimately be called a neutron spectrometer was built at Argonne National Laboratory by Zinn[1] in 1945. Although the first experimental measurement of the intensities of neutrons diffracted by single crystals, which determined the scattering amplitudes of various nuclei, was made by Fermi and Marshall[2] in 1947 at Argonne at their original heavy water reactor, an even earlier diffraction experiment was carried out by Mitchell and Powers[3] in 1936 using a radium-beryllium neutron source.

About a year and a half ago a Workshop devoted to Uses of Advanced Pulsed Neutron Sources[4] was held at Argonne National Laboratory. A group of about 100 people, representing most of the neutron scattering laboratories in the world, set down its statement of where the science of the application of neutrons for condensed matter research stands now. It is an exciting science and I would like to tell you something about it today.

The Argonne Workshop took place at a time when a number of examples of operating pulsed neutron sources and a number of studies of advanced pulsed source systems were emerging. These include the present and scheduled new Harwell electron linac sources in the U.K., the series of Soviet pulsed fast reactors IBR-I, IBR-30, IBR-II, the Soviet electron linac source at Kurchatov, the Japanese electron linac source operating at Tohoku, the proposed Japan Linac Booster, the KENS spallation source at Tsukuba, Japan, the operating electron linac at Toronto, the studied application of the Oak Ridge Electron Linear Accelerator for neutron scattering, the nearly completed Weapons Neutron Research Facility at LAMF, and the Argonne pulsed neutron sources ZING-P, ZING-P', PPNS, IPNS-I, and IPNS-II.

These systems are the source designers response to the fact that the conventional neutron sources, steady state reactors, have reached limitations imposed by heat transfer and cost, and thus the available neutron fluxes have increased by only about a factor of 3 in the past twenty years. This is illustrated in fig. 1.

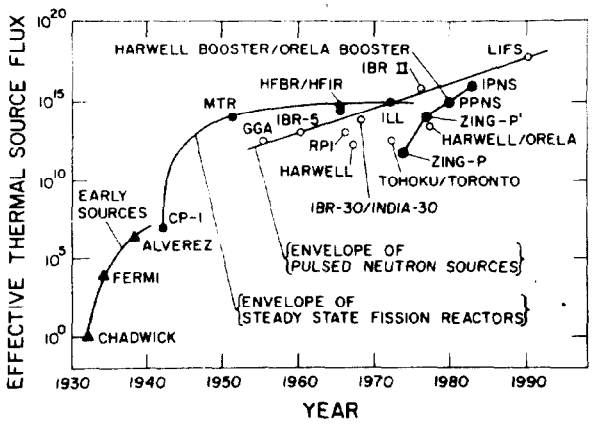

Fig. 1
This figure was prepared by R. Brugger, University of Missouri, and shows the envelope of the highest flux reactors as a function of time. The conclusion must be that within any reasonable constraints on cost, higher flux reactors will not and perhaps cannot be built. The High Flux Reactor at the Institut Laue-Langevin, Grenoble, represents the current highest evolution of the steady state research reactor and provides a thermal neutron flux of about 1.2×10^{15} n/cm^2/sec. Also included on this figure are the projected fluxes for various pulsed neutron sources.

In almost all areas of neutron scattering research the source flux limits what can be done. The limitation is so severe that a great many important measurements cannot even by attempted. Pulsed neutron sources, with which time-of-flight measurement techniques must be used, offer the means for obtaining higher useful neutron fluxes. This is because the source is "on" for only a very small fraction of the time, and the heat which must be dissipated to produce neutrons is averaged out over time. Therefore, very high instantaneous neutron fluxes can be achieved.

In order that we are all tuned in on the same frequency, and I realize that your frequency is usually much higher than mine, I would first like to show you how thermal neutron scattering is carried out at a steady state source and contrast this with how they are carried out at a pulsed source. I will then show you what a first rate spallation source might look like, using Argonne's proposed IPNS as the example. The last half of my talk will then be devoted to a discussion of areas of condensed matter research in which neutron scattering and specifically a pulsed source is particularly useful. I will conclude with a short discussion of an experiment for which a pulsed neutron source is absolutely required.

II. Experimental Techniques at a Steady State and at a Pulsed Source.

Thermal neutron scattering experiments can be divided into two general catagories: a) Elastic scattering (sometimes called diffraction experiments) in which there is little or no exchange of neutron energy with the target during scattering, and b) Inelastic scattering in which the neutron creats or destroys a quantum of energy in the target, thus emerging with an energy different from its incident energy. In the first class of experiments we are probing the static structure of the target, and in the second we are looking at the target's dynamic structure.

There are two interactions of a thermal neutron with a target (solid, liquid or gas) which dominate all others:

a. The nuclear interaction which can be taken to be a series of Fermi pseudo potentials centered at each nucleus in the target, that is

$$V(\vec{r}) = \frac{2\pi\hbar^2}{m} \sum_j b_j \, \delta(\vec{r}-\vec{r}_j) \quad . \tag{1}$$

b. The magnetic interaction, for which

$$V(\vec{r}) = - \vec{\mu}_n \cdot \vec{B}(\vec{r}) \quad , \tag{2}$$

where $\vec{\mu}_n$ is the neutron's magnetic moment, and $\vec{B}(\vec{r})$ is the magnetic induction field inside the target resulting from atomic magnetic moments.

By measuring the scattered neutron intensity as a function of angle and energy transfer, we obtain the Fourier transform of the space and time dependence of these interaction potentials (within a phase factor). The Fourier variables are, of course,

the momentum transfer

$$\hbar\vec{Q} = \hbar(\vec{k}_2 - \vec{k}_1) \quad . \tag{3}$$

and the energy transfer

$$\hbar\omega = \frac{\hbar^2}{2m} (k_2^2 - k_1^2) \quad . \tag{4}$$

In these expressions $\vec{k}_1$ and $\vec{k}_2$ are the incident and final neutron wave vectors.

Let's first talk about an elastic scattering experiment which is schematically shown in the top half of fig. 2. On a steady state source there are two ways to carry out such an experiment. We can use a single crystal and obtain a monochromatic beam by Bragg reflection. This beam is scattered by the sample and the intensity is monitored as a function of scattering angle 2θ. If the sample is crystalline we will observe a series of sharp Bragg peaks. If it is a liquid we will see a series of diffuse peaks which contain information regarding short range atomic correlation of the liquid. Alternatively we could place a chopper between the source and the sample and observe the intensity as a function of time-of-flight at a fixed angle. At least conceptually, these techniques can yield the same information with equal efficiency. In both cases we throw away about 99% of the thermal neutrons coming from the reactor which are potentially useful. With a crystal monochromator we take about a 1% energy slice out of the incident Maxwell-Boltzmann spectrum, and with the chopper we use the whole spectrum, but only 1% of the time.

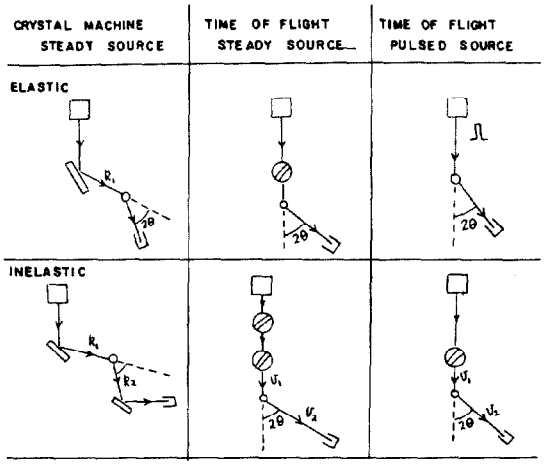

Fig. 2

If we have a pulsed neutron source, we do not need the chopper. We can use neutrons of all energies in the pulse since they are already put on a time base by pulsing the source. Thus, we see immediately the potential advantage that a pulsed source has over a steady state source. In this type of experiment it is clear that it is appropriate to compare peak flux with steady state flux when assessing the relative source luminosities. There is a further advantage which is presently difficult to assess. Because a pulsed source is "off" when the neutrons are being counted, the background can potentially be very low.

There are again two ways to carry out an inelastic scattering experiment at a steady state source as shown in the lower half of fig. 2. We must define both the incident and final neutron wave vectors $\vec{k}_1$ and $\vec{k}_2$. The incident wave vector $\vec{k}_1$ is defined by Bragg reflection from a monochromator at a particular angle, and the final wave vector $\vec{k}_2$ by reflection from an analyzing crystal at some other angle. The difference in the squares of $\vec{k}_1$ and $\vec{k}_2$ gives the energy transfer and the vector difference gives the momentum transfer. This is called a triple-axis spectrometer. Alternatively we can define the incident neutron wave vector $\vec{k}_1$ by selecting a given neutron velocity $\vec{v}_1$ using a pair of phased choppers. The beam is then scattered off the target and the time of arrival at a detector after the choppers opened gives the final velocity $\vec{v}_2$ (or wavevector $\vec{k}_2$). Again, using a single detector, these techniques are roughly equivalent in terms of data collection times. However using a multidetector on the time of flight machine we can potentially increase its efficiency (and cost) enormously. The dis-

advantage in the time of flight technique is that it is difficult to isolate a particular part of the scattering cross section in a local region of $\omega,\vec{Q}$ space and look at it separately and exclusively. Thus, a great deal of data, which we are not interested in will be accumulated in order to get the data we want. Although the triple axis technique is inherently very inefficient, it is very useful in experiments where we either know something about the sample beforehand, or have a limited interest in the general overall features of the scattering cross-section.

If we have a pulsed source, we can eliminate the first chopper. It is clear that we must phase the chopper downstream with the source. In fact, in accelerator based systems we must run the choppers for all the experiments and the accelerator off of a master clock. Thus, the incident velocity $\vec{v}_1$ is determined by the pulsed source and the downstream chopper, while the velocity of the scattered neutron $\vec{v}_2$ is determined by its time of arrival at the detector.

III. A Pulsed Spallation Source

A schematic diagram of the intense pulsed neutron source proposed at Argonne National Laboratory is shown in fig. 3.

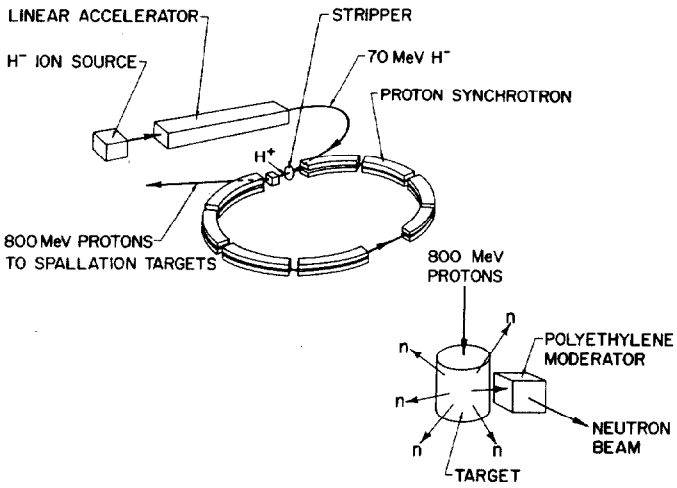

Fig. 3

H^- ions are accelerated to 70 MeV in a linear accelerator and then stripped and injected into a high intensity synchrotron, in which the proton energy is increased to 800 MeV. The machine is to operate at 60 Hz, giving bursts of 5×10^{13} protons/burst every 16.7 ms upon ejection. This beam is to be alternately steered to a uranium target for neutron scattering studies and to a tungsten target for radiation damage studies (which I will not discuss here). At this energy each incident proton yields about 30 fast neutrons by spallation. These neutrons are then slowed down in a hydrogenous moderator leading to an effective peak thermal neutron flux of about 10^{16} n/cm^2/sec. The neutron pulse width is determined primarily by the slowing-down time and is of order 10μs, in the thermal range.

Fig. 4 shows the energy spectrum for a particular moderator design.

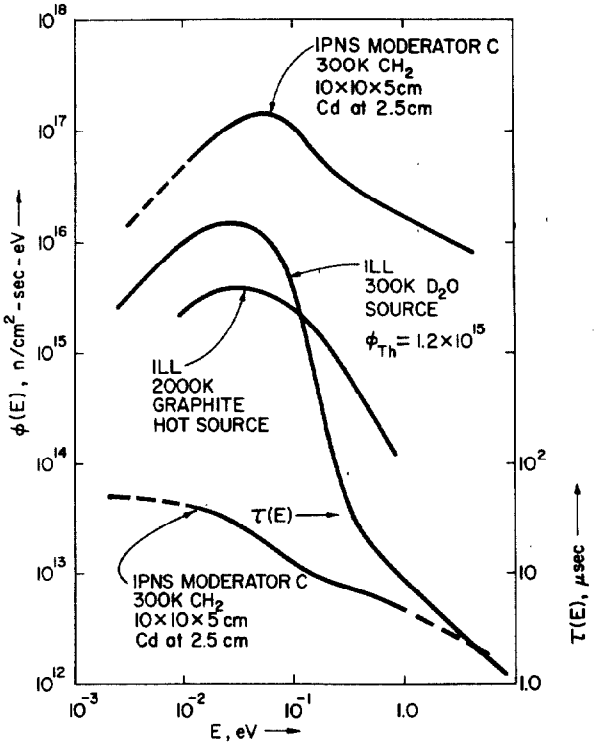

Fig. 4

The neutron energy spectrum for the ILL reactor is also shown for comparison. A very important feature of such a pulsed source is the fact that there will also be a prolific supply of epithermal (>.1 eV) neutrons in addition to the very high thermal flux. These higher energy neutrons will be very useful for a wide variety of experiments, one of which we will discuss shortly. It is for exactly this reason that the ILL reactor has installed a "hot" source in order to compensate somewhat for this general lack of epithermal neutrons in the leakage spectrum of a thermal reactor.

There are four general characteristics of an intense pulsed neutron source, such as IPNS, which has recently led to the considerable enthusiasm among scientists involved in condensed matter research:

- Very high intensity throughout the thermal energy range
- High epithermal flux (>0.1 eV)
- Pulsed nature of the source
- The moderator can be tailored to produce very intense beams of either "cold" or "hot" neutrons.

IV. Problems in Condensed Matter Research

Thermal neutron scattering has grown to become one of the most general experimental techniques in condensed matter research. In many cases it yeilds direct microscopic information which is inaccessible by any other technique, and in many other cases produces results which are completely complementary to the information obtained in other physical measurement methods. Applications spanning biology, chemistry, physics and materials science are well established and are growing constantly.

The region of energy and wave vector transfers in current neutron experiments, using thermal reactors

is, roughly speaking below 0.150 eV and 10 $\mathring{A}^{-1}$.

Experiments of fundamental interest have been proposed which require energy transfers up to 10 eV, and in other cases wave vector transfers out beyond 50 $\mathring{A}^{-1}$. Some of these experiments would be feasible with IPNS. Due to the pulsed nature of the source, very high pulsed magnetic fields, pulsed electric fields, or pulsed pressures can be applied to a sample. Certain time-dependent phenomena having relaxation times in the millisecond to minute range can be investigated utilizing the pulsed nature of the incident flux. A very high incident intensity allows experiments to be done on much smaller samples. Thus, experiments could be done on materials which are very rare, very expensive, highly absorbing, or are small fragments of other materials. A summary of areas of experimental research where an intense pulsed neutron source would be particularly useful and in some cases essential is given in table I. I would like to discuss one of these experiments, which is totally impossible at a steady state source in a little more detail.

V. An Experiment Which Is Impossible at a Steady State Reactor

It appears possible, using the prolific flux of epithermal neutrons from a source such as IPNS, to directly observe the continuum band of Stoner excitations in the ferromagnetic transition metals iron, nickel and cobalt. This would represent a major breakthrough in our understanding of the origin of ferromagnetism in metals.

Current theories of magnetism in metals divide the excitations into two classes. The first class is the collective excitations, in which almost all of the "magnetic electrons" (unpaired 3d electrons in the transition metals) contribute. These are called spin waves, or magnons, and there are thought to be two dispersion branches, an "acoustic" branch for which $\omega \to 0$ as the wave vector $q \to 0$, and an optic branch. A schematic illustration[5] of these excitations for nickel metal is shown in fig. 5. The second class is the Stoner excitations naively viewed as those arising from magnetic excitations of a single electron. This single particle continuum of excitations covers a broad range of energies as shown in fig. 5. There is a clear distinction between the single particle continuum and the collective states. Only the low energy magnons have been observed. The decrease in intensity expected as this branch enters the continuum and interacts with the single particle states has been observed[6].

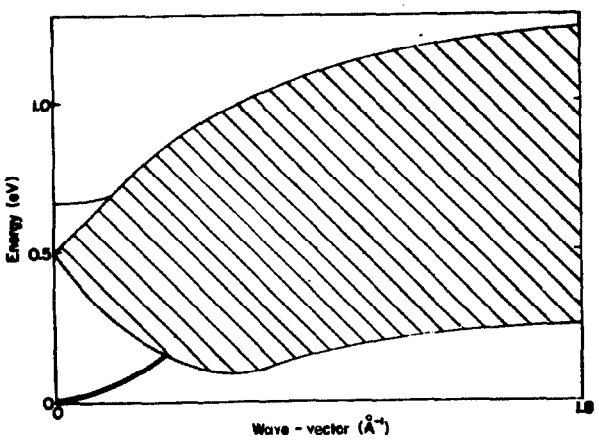

Fig. 5

The difficulty in observing the single particle excitations with neutrons arises from the fact that they occur at relatively high energies and also that they form a continuum where the scattering cross-section in a local region of $\omega, \vec{Q}$ space is very low.

The problem arising from a general lack of epithermal neutrons from a reactor is even more severe in magnetic problems than is at first apparent. Wave vector transfers $\vec{Q}$ must be restricted to less than, say 4 $\mathring{A}^{-1}$, because the magnetic form factor drops off rapidly with increasing $\vec{Q}$, leading to a rapid decrease in the neutron scattering cross-section. In order to close the scattering triangle, thus satisfying both energy and momentum conservation, the incident neutron energy E_o must be considerably greater than the excitation energy $\hbar\omega$. This is illustrated in fig. 6.

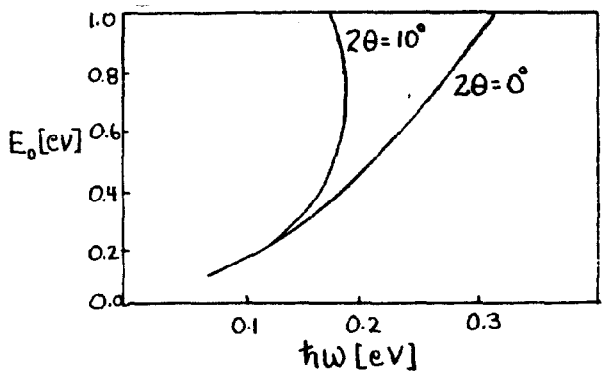

Fig. 6. Minimum incident neutron energy, E_o to observe an excitation of energy transfer $\hbar\omega$ with $Q = 4\ \mathring{A}^{-1}$.

For example, to see an excitation $\hbar\omega = 0.3$ eV we must have a reasonable flux of incident neutrons of energy $E_o = 0.86$ eV. It is clear from this that there is no hope of carrying out an experiment of this type at a steady state reactor. The only hope on the horizon is the intense pulsed spallation source.

VI. Conclusion

I have attempted to give you a birds-eye view of the exciting applications of a first-rate intense pulsed neutron source in condensed matter research. I personally believe that the future scientific horizons in neutron scattering will remain horizons until a well-instrumental pulsed spallation source is constructed.

Table I

Uses of Advanced Pulsed Neutron Sources

CHEMISTRY

-- Molecular vibration in solids, especially modes with large H amplitudes (high E)

-- Molecular vibration in liquids and dense gases (high E, low Q)

-- Spectroscopy of optically forbidden transitions, e.g., triplet-singlet transitions in molecules (high E, low Q)

-- Small particles, surfaces, adsorbed species (high ϕ_{th}, high E)

-- Intercalation, absorbed species and non-stoichiometric compounds (high ϕ_{th})

Uses of Advanced Pulsed Neutron Sources

METALLURGY AND CERAMICS

-- Dynamics of disordered solids, e.g., non-stoichiometric hydrides and oxides (high ϕ_{th})

-- Phonon densities-of-state of superconductors as function of (P,T) (high ϕ_{th})

-- Studies of small distortions of materials due to defects, radiation damage, etc. (high Q)

-- Interatomic potentials in alloys and ceramics from diffuse scattering (high ϕ_{th})

-- Relaxation after pulsed sample condition--shock wave, laser pulse, etc. (high ϕ_{th}, pulsed)

-- Studies of spinodal decomposition near a critical point (high ϕ_{th}, pulsed)

Uses of Advanced Pulsed Neutron Sources

PHYSICS OF LIQUIDS AND GLASSES

-- Neutron Compton scattering--ground state momentum distribution (high E)

-- Determination of interatomic forces from $\int S(Q,\omega)\omega^4 \, d\omega$ (high E)

-- Structures of glasses and molecular liquids at high spatial resolution (high Q)

-- Dynamics of amorphous chalcogenide semiconductors (high ϕ_{th})

-- Detailed studies of transition from hydrodynamic to zero sound regime (high ϕ_{th})

Uses of Advanced Pulsed Neutron Sources

PHYSICS OF SOLIDS

-- Electronic level spectroscopy--semiconductors, metals (high E)

-- Stoner excitations and high energy magnons in itinerant magnets (high E, low Q)

-- Optic modes and higher harmonics in hydrides (high E)

-- Dynamics of very small crystals, e.g., organic charge transfer salts (high ϕ_{th})

-- Measurements at very high magnetic fields, very high pressures (high ϕ_{th}, pulsed)

Uses of Advanced Pulsed Neutron Sources

BIOLOGY

-- Protein crystallography--differential labeling, anomalous dispersion, etc. (high ϕ_{th}, high E)

-- Structure of macromolecular complexes, e.g., viruses, enzyme complexes (high ϕ_{th})

-- Structure of periodic cellular assemblies, e.g., muscle, tendon, nerve myelin sheath (high ϕ_{th})

-- Study of regulation mechanisms (high ϕ_{th})

-- Kinetics of proteins in solution (high ϕ_{th}, pulsed)

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Acknowledgement. I would like to thank Dr. J. M. Carpenter for introducing me to the wonderful idea of pulsed spallation neutron sources and for his encouraging me to think about their application in condensed matter research.