SLOW NEUTRON SCATTERING EXPERIMENTS WITH THE HARWELL LINAC PULSED SOURCE

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The 45 MeV Harwell electron linac has been used as a source of neutrons for solid state physics experiments. Alternative pulses from the linac are directed to different targets. Short pulses go to the neutron booster which is used for nuclear cross-section measurements and longer pulses to a separate target for slow neutron scattering experiments. The neutron fluxes produced are compared with those available from research reactors and the count rates in equivalent experiments are estimated. Structure factors for Mn_5Ge_3 have been measured using a single crystal specimen. Results are also presented for diffraction from powder samples including a measurement of preferred orientation in Bi_2Te_3 powder compacts and a diffraction pattern for a sample of carbon fibers. Preliminary results are given for scattering from a liquid (heavy water), showing that results over a greater range of the wave vector transfer (Q) can be obtained using the time-of-flight technique with the "hard" spectrum from a small slab of moderator. Inelastic scattering using pulsed sources is shown to be particularly useful in the epithermal region and for experiments at low momentum transfer. It is concluded that electron linear accelerators can provide pulsed sources of slow neutrons which are competitive with thermal reactors and advantageous for particular experiments. Some problems of particular importance which require more study are indicated.

INTRODUCTION

For many years studies of the condensed state using pulsed neutron methods have been carried out on continuous beam reactors using mechanical choppers to pulse the beams. More recently, interest has turned to pulsed neutron sources based on linacs or pulsed fast reactors. In particular, interest has been stimulated by the series of pioneering experiments performed at Dubna with the unique IBR reactors. This paper reviews a small program of research at Harwell that has been aimed at assessing the potentialities of pulsed sources in this field and identifying the experiments for which pulsed sources give a positive advantage.

The Harwell linac [1] was principally designed to inject relatively short (0.23 μsec) electron pulses into a multiplying target or "booster." The booster multiplication was limited to ten in order to keep the neutron pulse length short, but it has the additional advantage over a nonmultiplying target of a greatly reduced "gamma-flash." This booster target has been used chiefly for neutron cross-section measurements over the entire range from 1 eV to 10 MeV [2]. From the start, the linac has been "multiplexed." Although 192 pulses per second have been injected into the booster, the klystrons have been run at 384 pps with a radiofrequency envelope ~2.7 μsec. The pulse length at the electron gun can then be programmed to provide 0.23 μsec pulses at 192 pps with up to 192 pulses of some other pulse length from 10 nsec to 1.7 μsec. A pulsed magnet in phase with the gun can then deflect the intervening pulses into the appropriate cell. Figure 1 shows the linac with the straight ahead beam going to the booster (cell 1) and the

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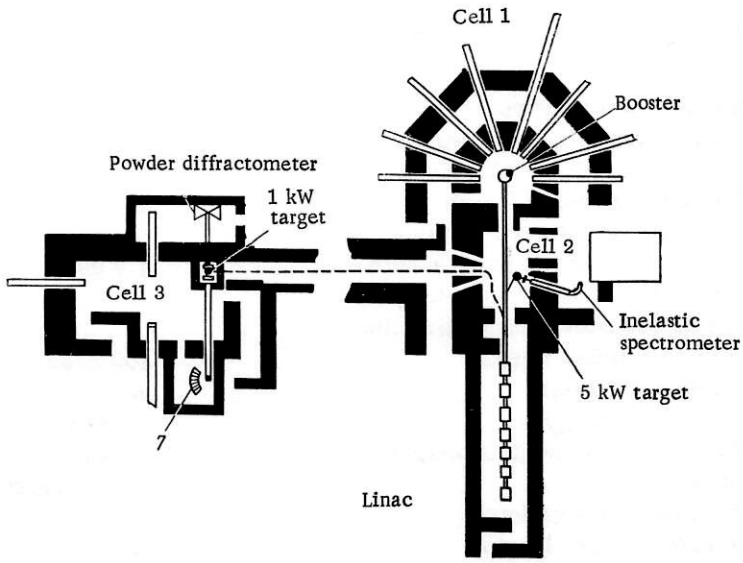

Fig. 1. Harwell linear accelerator laboratory.

deflected beams going to cells 2 or 3 where slow neutron scattering experiments can be performed. This ability to multiplex has led to a flexible facility well able to cope with the changes of research program which have taken place over the years. The short pulses have been used for photonuclear research [3] and fast neutron spectrum measurements [4]; the longer pulses, which were used in the study of thermal neutron spectra in moderators [5], are now used for solid state physics investigations by time-of-flight techniques.

Some parameters of the machine are given in Table 1. If a machine were being designed for multiplexing now, it would be desirable (and seems possible) to multiplex the modulators in order to minimize the mean klystron power. Otherwise if, for example, 8-μsec pulses were required at 100 pps but the machine was run at 400 pps to provide additional short pulses, the mean power would be almost four times greater than necessary.

THE NEUTRON SOURCE

Electron Targets

The electrons are stopped in a material of high atomic number (Z) to produce bremsstrahlung; for a low-powered target uranium can be used, but for higher powers thermal conductivity considerations dictate tungsten, gold, or flowing mercury.

Neutrons are produced by (γ, n) reactions in heavy materials but if fissile elements are used (γ, f) reactions can double the neutron output [6]. Figures 2 and 3 summarize the important factors in the choise of target materials. Some obvious conclusions are that an electron linac for neutron production should run at 40 MeV or more and that neutron producing targets should be made of fissile materials.

The first experiments at Harwell were carried out at 96 pps using a 25 μA (mean current) beam of 35 MeV electrons in 1-μsec pulses. Thus, the target power was 900 W and a target of air-cooled natural uranium could be used with a gold bremsstrahlung converter.

Later experiments at 192 pps have used a 140-μA beam in 1.7-μsec pulses giving a power of almost 5 kW. The target design has been improved by the use of water cooling as is shown in Fig. 4. The electron beam passes through a thin stainless steel window and then falls on two gold disks (thickness 3 mm) followed by nine disks of natural uranium (thickness 6 mm). Roughly half the heat produced is deposited in the two gold disks.

The design of a target to dissipate 50 to 100 kW should be possible using present-day fuel element technology.

TABLE 1

Number of klystrons	7
Peak power of each klystron	7 to 8 MW
Mean power of each klystron	20 kW
Maximum rf pulse length	2.7 μsec
Beam energy at zero current	55 MeV
Maximum electron pulse length	1.7 μsec
Minimum electron pulse length	10 nsec
Normal booster target operation:	
electron pulse length	100 nsec
electron current	500 mA
electron energy	42 MeV
Maximum peak neutron output from booster	$5 \cdot 10^{17}$ neutrons/sec
Short pulse operation:	
electron pulse length	10 nsec
electron current	1000 mA
electron energy	45 MeV
Total number of targets	7
Number of targets run simultaneously	3
Repetition frequency is variable up to	500 pps

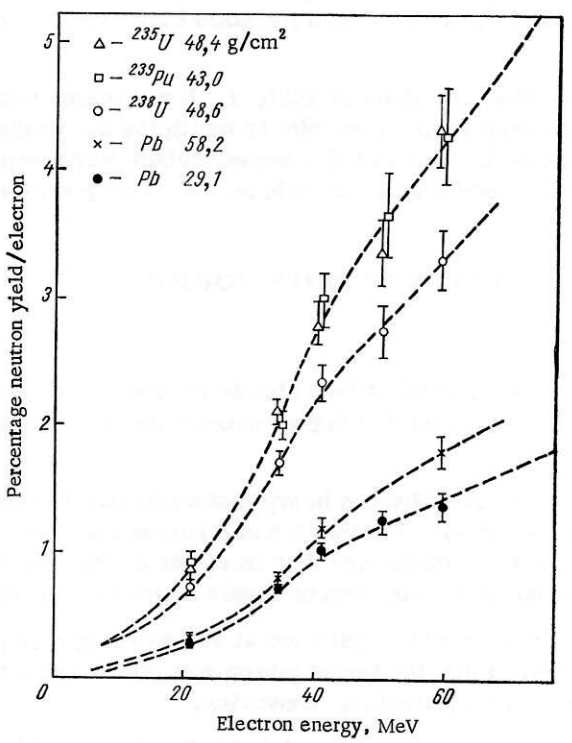

Fig. 2. Dependence of neutron yield on electron energy.

Moderators

The fast neutrons from the target must be slowed down to energies less than one electron volt for solid state physics experiments. Slowing down times in moderators are such that only a hydrogenous moderator is suitable for pulse lengths of order 10 μsec. Simple considerations show that the figure of merit for an unpoisoned moderator is proportional to $N^{1/2}$, where N is the hydrogen atom density. Thus, ignoring more exotic materials, polyethylene, which is 10% better than water, has been used.

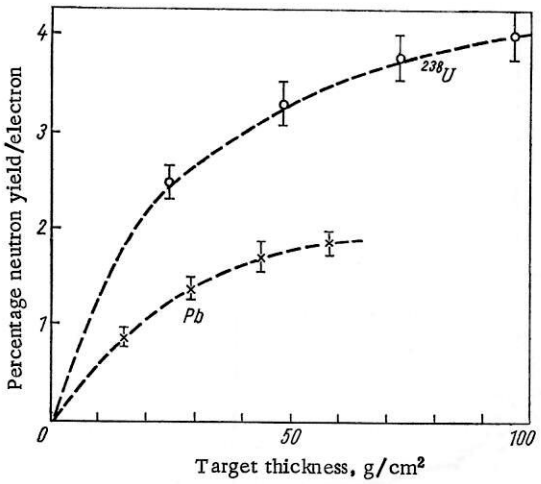

Fig. 3. Dependence of neutron yield on target thickness.

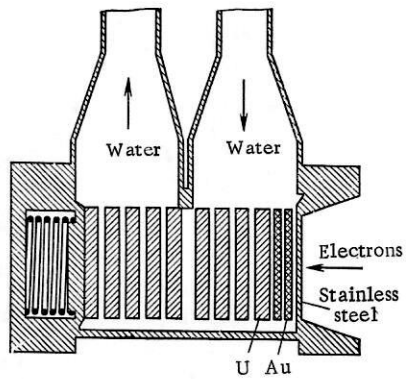

Fig. 4. Five-kilowatt target.

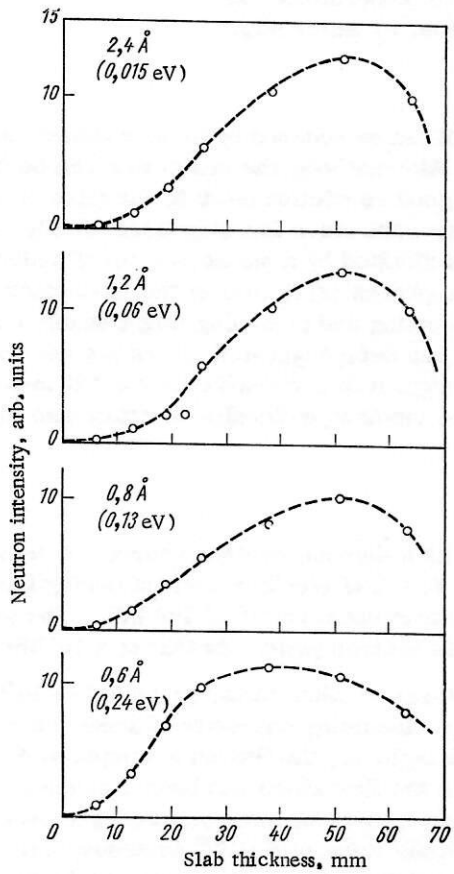

Fig. 5. Variation of intensity with polyethylene slab thickness at various wavelengths.

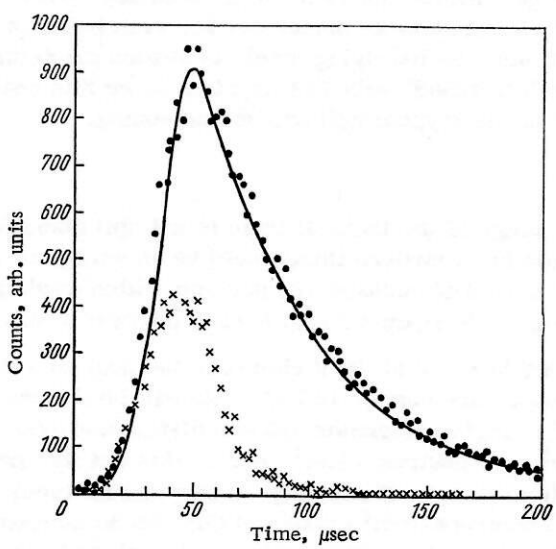

Fig. 6. Shapes of peaks reflected 1.4 Å. ●) Polyethylene slab, 38 mm; ×) polyethylene slab, 13 mm; ——) calculated peak shape.

For plane slabs of polyethylene, the slow neutron flux in the beam varies with thickness [7]; Fig. 5 shows the dependence for various neutron wavelengths. For pure room-temperature polyethylene, the duration of the slow neutron pulse at 10 kT (0.25 eV) is short, since it is dependent on the fast neutron pulse length (~2 μsec) and the slowing down time (also ~2 μsec). However, at lower energies up-scattering of the thermal neutrons becomes possible, and the pulse shape is determined by the decay of the ther-

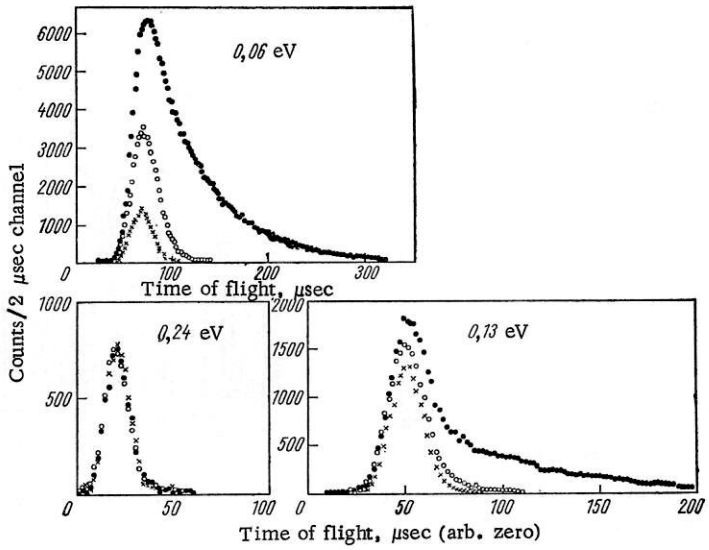

Fig. 7. Measured peak shapes for poisoned moderators. ●) Pure water; ○) water with 7.5 g/liter boron; ×) water with 8.5 g/liter gadolinium.

mal neutron burst in the moderator block. This exponential tail can be reduced by using a thinner slab of moderator (see Fig. 6) but with a reduction in peak intensity. Alternatively the moderator can be poisoned with absorber (see Fig. 7); this probably enables one to obtain good resolution down to energies of order 2 kT (~0.05 eV). However, this field would benefit from further work since investigations of only two poisons at one concentration each and only at a set of energies dictated by a particular set of orders of a single Bragg reflection have been made at Harwell. Another approach [8] is to cool the moderator with liquid nitrogen; hence, the 10 kT limit becomes ~0.06 eV and cooling and poisoning might enable one to reach 0.01 eV. A suitable moderator for even lower energies has been suggested [9] but not yet tried out. Since there must be low-lying levels to absorb the neutron energy, it is proposed to use a 4°K methane moderator "stretched" with 16% krypton, since this enables the methane molecules to rotate and the natural absorption of the krypton will provide poisoning.

Flux

The design of the Harwell linac is not optimized for use as a thermal neutron source. A reasonable specification for a modern linac would be an energy of 100 MeV, 1 A of electron current during the pulse, and 8 μsec pulses at perhaps 125 per sec, which would give a mean beam power of 100 kW. The target shown in Fig. 4 dissipates 5 kW; it is of interest to compare its neutron yield with that of a 10-MW reactor.

A 5-kW beam of 35 MeV electrons incident on a thick natural uranium target gives $1.65 \cdot 10^{13}$ fast neutrons/sec. Since about 10% of these will be thermalized and assuming one-sixth of these leave the surface of a single moderator and are distributed over a solid angle 2π, the flux on a sample at 4 m will be $2.7 \cdot 10^5$ slow neutrons/(cm^2 · sec). This result agrees with the flux which has been measured, $3 \cdot 10^5$ neutrons/(cm^2 · sec). The leakage of thermal neutrons from the moderator surface, averaged over time, is $1.4 \cdot 10^9$ neutrons/(cm^2 · sec) and this can be compared with the value of $6 \cdot 10^{10}$ neutrons/(cm^2 · sec) for the IBR-30 reactor. This ratio of 40 is close to that expected from the mean powers of the two facilities.

To derive a comparable figure for a reactor, consider a pulsed white beam produced with a chopper. Consider a source flux of $5 \cdot 10^{13}$ thermal neutrons/(cm^2 · sec) and close to this a chopper which transmits 100 ten μsec bursts per sec with a transmission of 0.5 (to allow for the restriction of the spectral range transmitted) and a slit area of 10 cm^2. The flux on a sample at 4 m would be $2.5 \cdot 10^5$ neutrons/(cm^2 · sec), which is the same as the flux produced by the linac target.

This is, however, an oversimplified comparison: in fact it is more probable that a reactor experiment will use a monochromated neutron beam. It is important in these comparisons to remember that

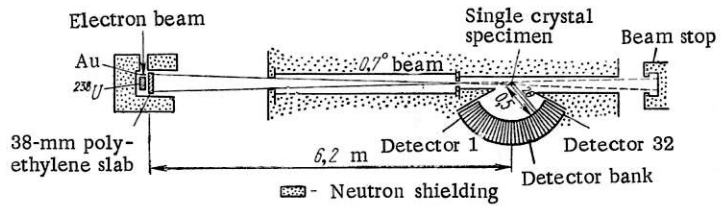

Fig. 8. Time-of-flight diffractometer with 32 detectors.

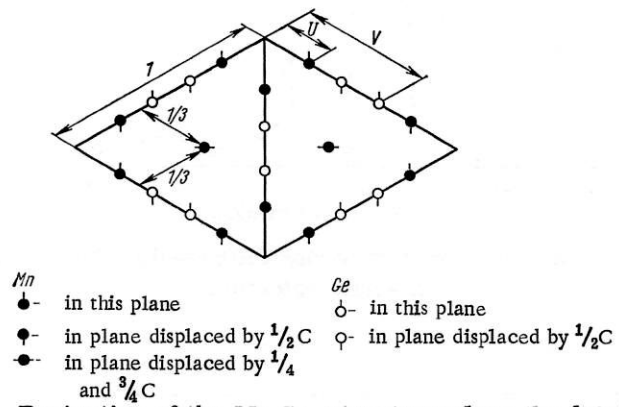

Fig. 9. Projection of the Mn_5Ge_3 structure along the [001] axis.

in any pulsed source experiment the result is obtained in a single run as a time-of-flight spectrum, whereas in the monochromated beam experiment the result is a series of points measured sequentially, an angular distribution, or (for an inelastic experiment) a scan with various settings of the monochromator energy.

For several types of experiment the useful mean flux incident on the specimen is an important factor. For the comparison considered here the reactor will give $\sim 10^6$ neutrons/(cm^2 · sec) in a monochromatic beam of 1 Å neutrons with a resolution of ~2.5%. This is a factor three higher than the linac flux. However, the time-of-flight technique with stationary detectors can, in general, use larger counter arrays than the monochromatic beam method, and including this factor makes the two facilities at least comparable. This conclusion assumes that the white beam experimental methods can produce data of the same quality as that from the highly developed monochromatic beam techniques. The remainder of this paper describes an attempt to test this assumption for a few important types of experiment.

EXPERIMENTAL RESULTS

Diffraction by Single Crystals

The structure of Mn_5Ge_3 has been investigated [10] to identify the problems and to test if the time-of-flight technique could yield accurate structure factor values. The experiment (Fig. 8) was carried out using the 1-W air-cooled target with a 44-mm slab of polyethylene as moderator, since the exponential tail on the pulse was considered tolerable. Measured peak widths at half the maximum intensity varied from 12 μsec at 0.5 Å and 45 μsec at 1 Å to 85 μsec at 2 Å; the corresponding times-of-flight over a 6.7-m flight path were 850, 1700, and 3400 μsec giving a resolution of around 2%.

The specimen was a pillar of Mn_5Ge_3 0.83 cm high and 0.38 × 0.27 cm in section. Mn_5Ge_3 is a hexagonal crystal with lattice parameters a = 7.170 Å, c = 5.043 Å. The projection along the [001] axis is shown in Fig. 9, and the unknown parameters U and V are given [11] as 0.25 and 0.61.

The detector bank consisted of 32 3.5-cm^2 lithium glass scintillators arranged on an arc of 0.5 m radius. Each scintillator subtended 4° at the specimen; and, since the dead spaces between the scintillators were small, the whole arc subtended 128°. The sample was positioned so that a zone of reflections lay fully on the detectors; this procedure would be unnecessary with a large position-sensitive detector.

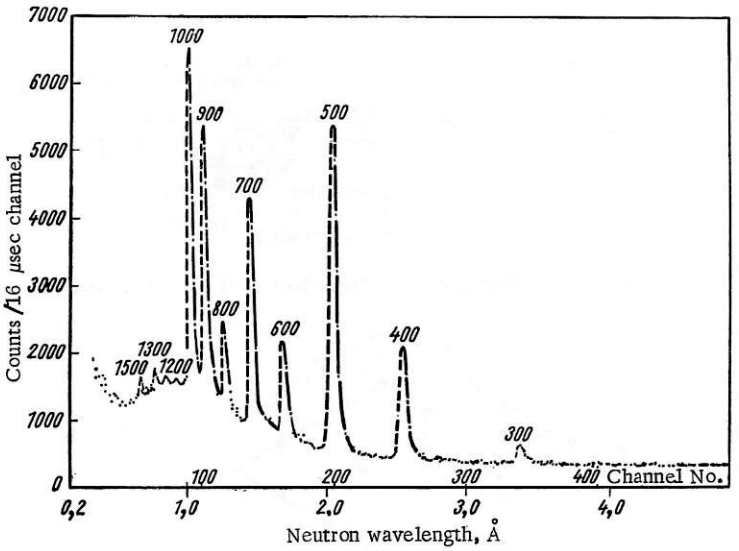

Fig. 10. The spectrum of neutrons diffracted by Mn_5Ge_3 onto a single detector.

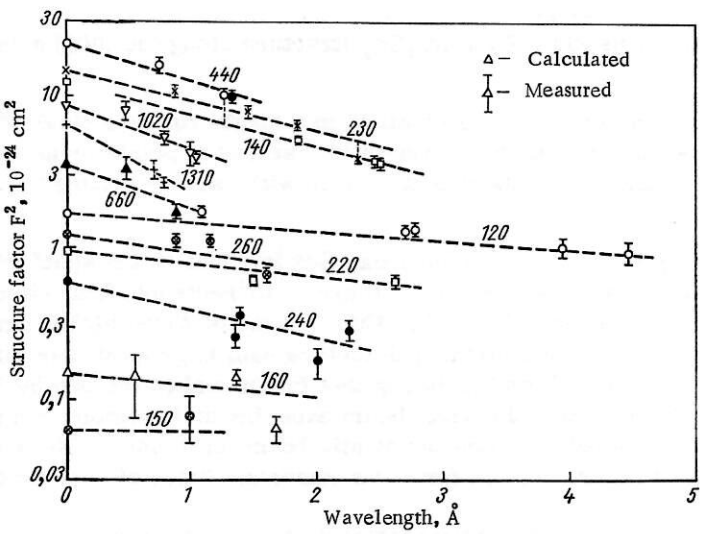

Fig. 11. Comparison of measured and calculated structure factors for Mn_5Ge_3.

The intensities of up to 112 Bragg reflections from 0.3 Å to 4 Å were measured simultaneously. Figure 10 shows the spectrum from the detector. The background under each peak was subtracted and the structure factors calculated, making corrections for counter efficiency and spectrum shape (both of which were derived from an experiment with a vanadium scatterer [12]) wavelength and scattering angle.

One set of results was repeated with good consistency, and a further set was then measured with the crystal reoriented. Since some of the measured reflections were equivalent due to the symmetry of the crystal, some structure factors had been measured at five different wavelengths. The results indicated (Fig. 11) that after correction for the absorption of the manganese there was still a wavelength dependent effect, strongest in the intense peaks which were ascribed to extinction. The results extrapolated to zero wavelength agreed with structure factors calculated for U = 0.23, V = 0.60. But lacking a procedure to correct for such strong extinction, a least-squares refinement was not attempted.

The results show that the structure factors for a single crystal can be measured using the time-of-flight method and a 1-kW electron beam. This method gives additional information on the variation of the

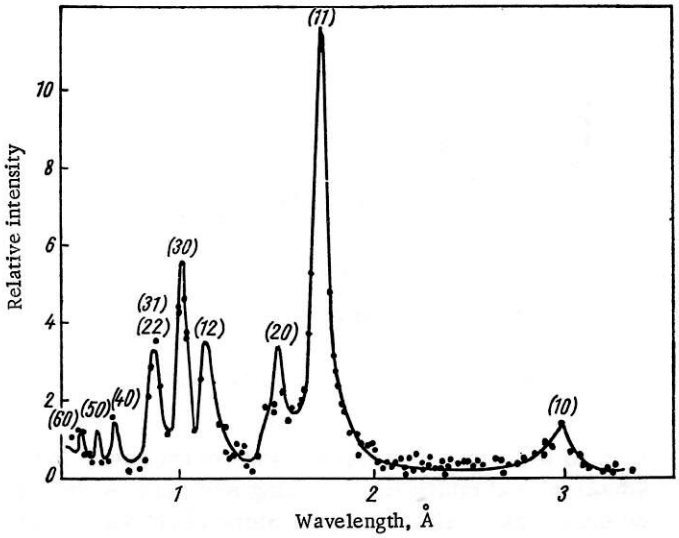

Fig. 12. Time-of-flight neutron diffraction pattern for carbon fibers lying in the plane of scattering. In this orientation the (00l) reflections are absent.

measured structure factors with wavelength. This additional information indicates whether the right absorption corrections have been made and enables one to estimate the size of the extinction effects.

Further work is needed in two directions. The accuracy of the technique should be studied using a simple ionic crystal of low absorption and known extinction. A complex crystal should be studied, measuring ~1000 reflections simultaneously using a large area detector.

Diffraction by Powdered Crystals [13]

Some powder diffraction experiments have been performed using the 800 W target and a 25-mm polyethylene moderator. The resolution of the equipment $\Delta\lambda/\lambda$ was ~1.5% but it was not optimized to give maximum count rate. Kimura [14] has shown that good results can be obtained in a very short time with a linac source if the detectors are arranged to cover the whole Debye-Scherrer cone.

An experiment to measure the orientation in a pressed sample of bismuth seleno-telluride [15] showed the potentialities of the time-of-flight method in obtaining simultaneous results for many reflections.

Figure 12 shows a diffraction pattern measured with a sample of carbon fibers; the widths of the peaks give an indication of the range of crystalline order [16].

A recent experiment [17] to measure phase changes in nickel sulfide at high pressure and low temperature showed that the time-of-flight technique has the advantage of recording all the results at a single angle. As Brugger has mentioned [18], this is a great advantage in high-pressure cells where considerations of strength dictate that only small "windows" can be provided.

Diffraction by Liquids

The diffracted neutron intensity, due to coherent scattering by a liquid, measured as a function of the wave vector transfer ($Q = 4\pi \sin\theta/\lambda$, 2θ is the scattering angle, λ the neutron wavelength), can be analyzed to give a radial distribution function. One of the parameters needed in this analysis is $I(\infty)$, the asymptotic value of the diffracted intensity at large Q. The conventional liquids diffractometer using a monochromatic beam can reach Q values of perhaps 13 $Å^{-1}$. Values greater than this are difficult to achieve because the reactor spectrum and monochromator reflectivity both drop markedly for $\lambda < 0.8$ Å, and $\sin\theta$ cannot be much greater than 0.9. This value of Q may be adequate for atomic liquids, but it is not high enough for molecular liquids.

The time-of-flight technique on a linac, using a position sensitive detector, can give a set of diffraction patterns either at various values of the wavelength or at different scattering angles, depending on the

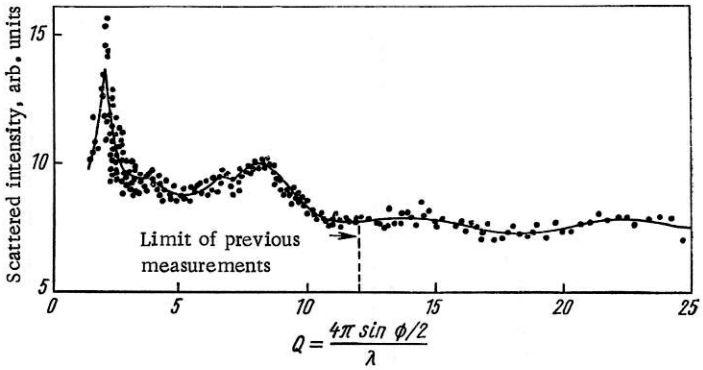

Fig. 13. Scattering of heavy water.

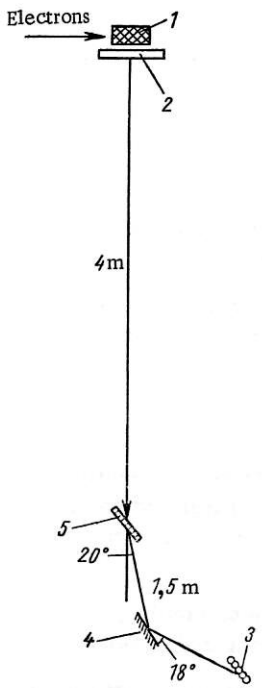

Fig. 14. Inelastic down-scattering experiment. Analyzed neutron energy 0.018 eV: 1) water-cooled target; 2) poisoned water moderator; 3) BF_3 filled proportional detectors; 4) germanium (111); 5) sample.

method of analysis. If the various corrections for inelastic scattering, absorption, and multiple scattering are made correctly, these patterns should collapse onto a single curve. Since short wavelengths are available (because a slowing down spectrum is "harder" than that in the reflector of a reactor and because no monochromator is used), Q values up to 25 $Å^{-1}$ are available. We have carried out a pilot experiment with heavy water (Fig. 13) which showed that there was significant structure out to high Q.

Thus liquids provide a promising field for investigation using pulsed sources. An incidental point is that the use of large position sensitive detector arrays gives a gain in count rates over present point by point methods which may make possible experiments at the triple point. This is a region of considerable theoretical interest [19].

Inelastic Scattering

The study of molecular excitations by observing the energy gain resulting from the incoherent scattering of cold neutrons is limited to the region below 0.08 eV by the Boltzmann factor. This range can be increased by using the energy loss (or down-scattering) technique. Using a crystal monochromator to select neutrons of various initial energies from a reactor beam and a beryllium filter detector to select neutrons scattered to energies less than 0.005 eV, energy transfers up to 0.18 eV (or, in the favorable case of zirconium hydride, 0.4 eV) can be studied with high counting rates [20]. This technique is limited because the epithermal neutron flux and the monochromator crystal reflectivity decline as the energy is increased but also because there is a penalty for using the highly efficient beryllium filter detector. The observation of high energy excitations is hindered by the presence of multiphonon terms. The intensity of the multiphonon effect is proportional to Q^4; thus, to observe a level above 0.2 eV when there is also a high scattering intensity from the lattice vibrations and molecular rotations at lower energies, Q must be minimized. The results of Harling [21] for water and ice illustrate this point well; the 427 MeV level in ice could only be observed at 15° scattering angle. If neutrons are scattered from energy E_1 to E_2 through a scattering angle φ and $E_1 - E_2 = \varepsilon$ for minimum Q, $E_2 = \varepsilon/2$ (cosec $\varphi - 1$) and the minimum value of Q is 21.95 $(\varepsilon \sin \varphi)^{1/2}$. Thus, the final energy should be of the same order as the energy transfer, and the scattering angle should be as small as possible. (Both conditions conflict with the use of a beryllium filter detector.)

Purohit has used the Rensselaer linac with a phased chopper to study down scattering from a fixed (high) initial energy to a range of final energies [22]. The possibilities have been investigated of using a linac for this type of experiment with a fixed final energy selected with a crystal analyzer. The time-of-flight spectrum then gives the whole inelastic scattering distribution. Figure 14 shows the experimental

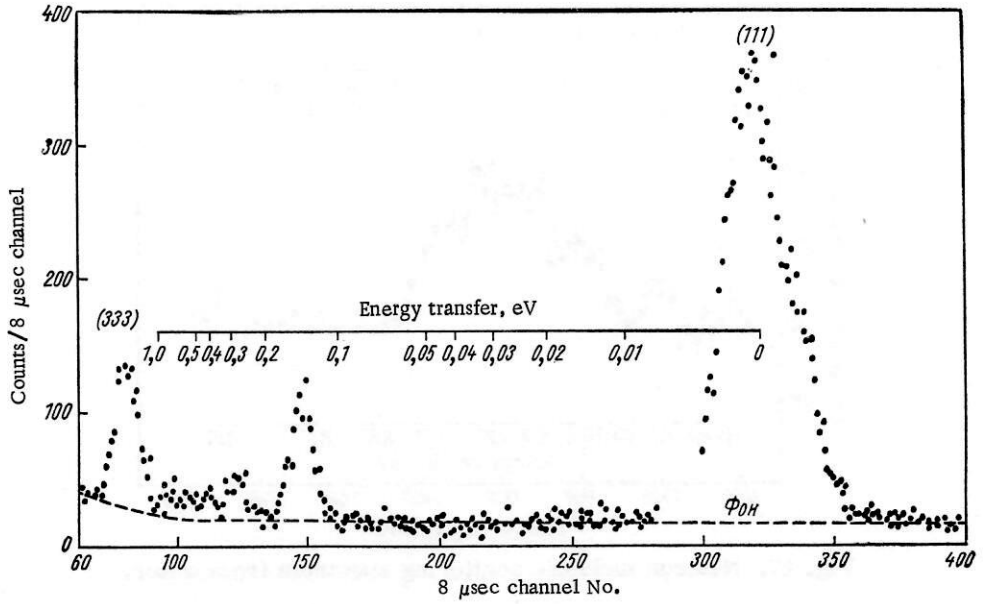

Fig. 15. Time-of-flight spectrum measured for zirconium hydride.

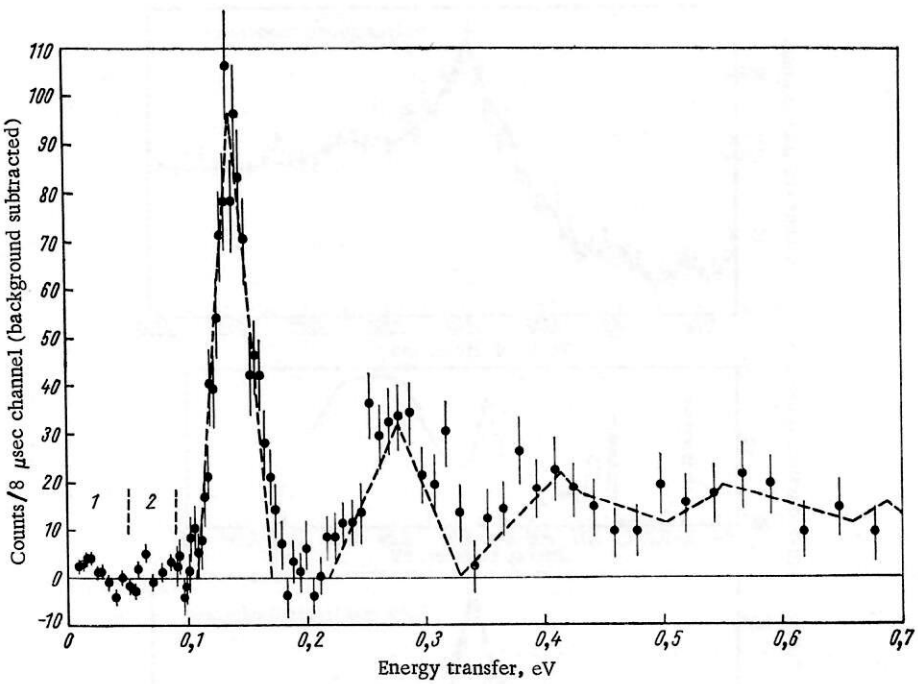

Fig. 16. Inelastic neutron scattering spectrum from zirconium hydride. 1) Means of 8 channels; 2) means of 4 channels.

arrangement; the scattering angle is 20° and the analyzer section is designed to give a "time focusing" effect. Overlap between the higher order elastic scattering and the inelastic spectrum is avoided by choosing the germanium (111) reflection for the analyzer since the (222) is absent; then with a suitable choice of flight path lengths the (333) does not intrude (Fig. 15). The final energy was 0.018 eV; with a more powerful linac it should be possible to raise this energy to 0.27 eV and eliminate the second order with a hafnium filter in a really low momentum transfer experiment. Figures 16-18 show some preliminary results. The zirconium hydride results show that the observation of the higher order modes is limited not by experimental resolution but by the natural widths of the levels. Instrumental resolution varies from

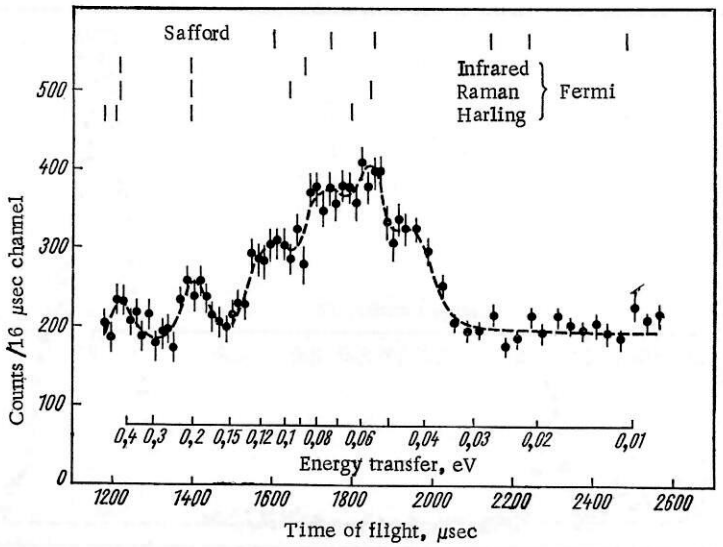

Fig. 17. Neutron inelastic scattering spectrum from water.

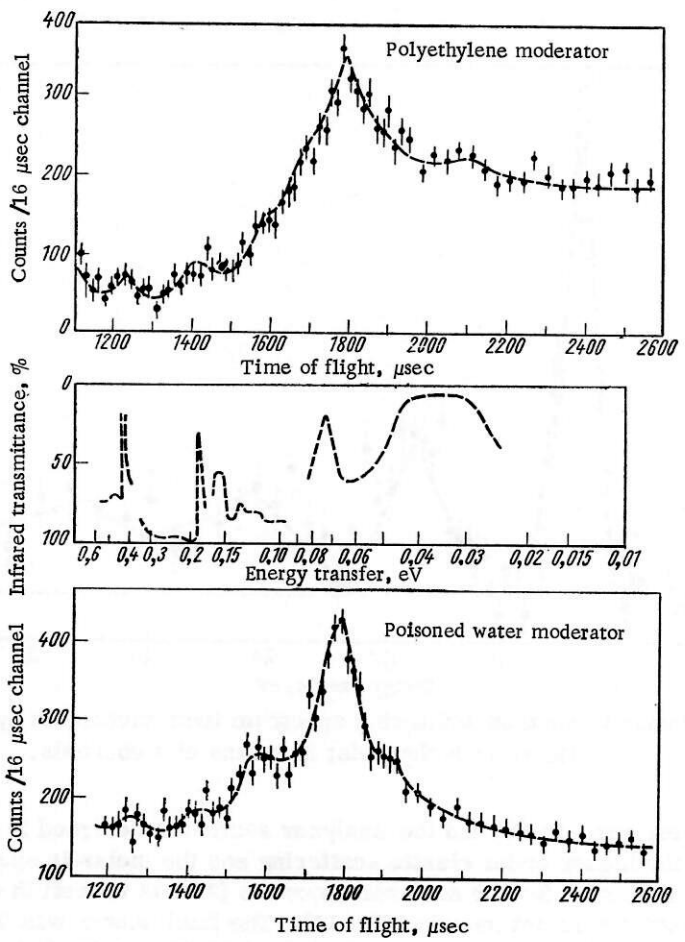

Fig. 18. Neutron inelastic scattering spectrum from sodamide.

6% for an energy transfer of 0.060 eV to 7% for 0.40 eV. The result for water shows that not only can the 0.21 and 0.40 eV vibration levels [21, 23] be observed but at lower energies in the torsional band the three levels observed by Safford [24] are resolved. Thus, one measurement by this technique covers the large and interesting region of energy transfers from 0.04 to 0.4 eV. The lower energy lattice vibrations around 0.025 eV are not observed because the poisoned moderator used to improve the resolution in this experiment absorbs too many neutrons at these energies. There is some indication of a level at about 0.045 eV, but higher statistical accuracy is needed for confirmation.

The scattering from sodamide ($NaNH_2$) has been studied using both polyethylene and borated water moderators. The results are compared with an infrared absorption spectrum [25]. The resolution of the torsional band at ~0.07 eV is considerably improved by the use of the poisoned moderator. It seems likely that the pronounced structure at 0.12 eV can be assigned to the rocking motion of the NH_2 group. With better statistics more details in this region could probably be observed. The structure at around 0.18 eV corresponds to NH_2 deformation and that at 0.4 eV to N−H stretching modes.

CONCLUSIONS

Brugger convincingly argued [26] that the next generation of intense neutron sources will consist of pulsed reactors or linacs with boosters. Recent experience would suggest that a modern linac with a neutron-producing target can produce important results in the study of condensed matter with expenditures rather less than are needed for a comparable reactor. In particular, a linac with "multiplexing" can be used simultaneously for nuclear physics and solid state physics studies.

More generally, reactor techniques have been perfected over the last 15 years, and several fields have been outlined in which effort is now needed in the parallel pulsed neutron techniques.

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