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PROPOSAL
FOR A
SPALLATION NEUTRON FACILITY

SCIENCE BOARD

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OCTOBER 1976

Reader's Guide

Readers of this document on the Spallation Neutron Source (SNS) will come to it with varying degrees of familiarity with the tools and techniques of neutron scattering and of their widely ranging applications in scientific research. The presentation of information in the Proposal seeks to take account of this situation and inevitably this leads to some repetition and overlap of material between chapters.

Chapter 1 gives a broad synopsis of the whole proposal.

Chapter 2 is essentially a layman's guide to neutron scattering concluding with brief surveys of the main application areas and their relationship to the SNS.

Chapter 3 summarises the conclusions of the scientific working groups. These are rather specialised but a summary of the important scientific gains which would accrue from the SNS is given in the opening paragraphs.

Chapter 4 is an account of the thinking on neutron sources which has led to the conclusion that the provision of the SNS is the best possible step forward to realise high effective neutron intensities.

Chapter 5 contains an outline description of the SNS and its neutron performance parameters, and information on costs and time scales.

More details on utilization and the project implementation are given in the two appendices.

PROPOSAL FOR A SPALLATION NEUTRON FACILITY

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P R E F A C E

This proposal for a new neutron source for thermal neutron scattering studies has been prepared by a Coordinating Group under the auspices of the Neutron Beam Research Committee, and represents the culmination of several years' study by the NBRC of the long term needs of the field and of the options for their realization. The scientific case was formulated by a Science Panel on the basis of the conclusions of four Working Groups appointed to examine specific areas of science to be served by the proposed facility. The proposals for implementation have been prepared by the Rutherford Laboratory.

The various working parties have demonstrated their enthusiasm for the proposal in a practical way by meeting demanding deadlines and I thank all who participated for their co-operation. We are all grateful to the many scientists in the UK and abroad whose work we have drawn upon without reference in reviewing the progress and prospects of neutron beam science.

The day to day burden of preparing the proposal has fallen on members of the Rutherford Laboratory staff and I should like to record how much their excellent work has been appreciated by everyone else involved.

B E F Fender

Chairman,
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SPALLATION NEUTRON SOURCE SCIENCE PANEL AND WORKING GROUPS

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CHAPTER 1. SYNOPSIS OF THE PROPOSAL

1.1 The proposal presents the case for a high intensity neutron facility designed for thermal neutron scattering. The facility is based on the use of a proton synchrotron to generate very intense neutron pulses from a spallation target. By making use of existing plant and buildings it could be built at the Rutherford Laboratory for a capital cost of £10M compared with an estimated £30M if built on a green field site.

1.2 Why is this new source needed? The question is best answered by considering the scientific successes which have been achieved in thermal neutron scattering to date, the restricted availability of high flux instruments currently accessible to UK scientists, and especially the exciting new science that would follow construction of a high intensity pulsed source with its emphasis on the higher energy region of the neutron spectrum. All the major areas of science at present making use of neutron scattering, involving physicists, chemists and biologists, would benefit from the spallation neutron source (SNS) and we would expect immediate advances in, for example, the liquid and amorphous state, chemical applications of neutron spectroscopy, high energy excitations in crystalline solids, relaxation phenomena in polymers, and kinetic aspects of biological processes. Moreover the enormously enhanced flux of hot neutrons is bound to lead to new and unexpected experiments in a manner analogous to the advances stimulated by the availability of cold neutrons at ILL.

1.3 In Chapter 2 of this proposal, the history of the SRC-supported programme is traced through to today's partnership in the Institut Laue-Langevin (ILL) in Grenoble. Although it has long been appreciated that neutrons play a crucial role in furthering our understanding of fields as diverse as magnetism and the dynamical properties of crystals and liquids, the advent of the ILL reactor has provided striking proof that the special properties of the neutron can be employed to provide unique contributions to our knowledge in almost all fields of condensed matter research. Thus it is possible to point to significant advances over the last year or two in areas as varied as the structure of aqueous solutions, alloy precipitation phenomena, the properties of quantum liquids, the structure and dynamics of polymers, and molecular biology.

1.4 This widening of the scope of neutron beam studies has inevitably led to constraints in the availability of ILL facilities. The new research which has developed round the use of the ILL can in no sense be described as a single 'programme', bearing in mind that it supports and extends a scientifically diverse set of projects for scientists from France, the UK and West Germany. The number and range of users increases yearly, and it is already difficult to accommodate the pressure of good experiments. As far as the UK is concerned this pressure is not surprising, given that the facilities represent only one third of those judged to be necessary when in 1971 the Council accepted the case for a UK high flux reactor.

1.5 The exploitation of the higher neutron intensities available at ILL, despite successes achieved, has already exposed limitations arising from currently available intensities. At every stage in the exploitation of neutrons, higher intensities have revealed new science, for example through the possibilities of improving experimental resolution, of examining systems which are relatively weak scatterers, or of investigating phenomena over wider ranges of experimental conditions. Experiments at the ILL have also demonstrated the value of marked increases in intensity outside the neutron wavelength range of 1-2 Å most frequently employed. It is possible now to see areas of science which will benefit from the increase in intensity associated with the proposed new source, and these are indicated in Chapter 2.

1.6 Useful neutron beam intensities can only be obtained through the provision of expensive central facilities; even at ILL the intensity of an average beam is only 10^8 neutron per second compared with around 10^{11} photons

per second from a laboratory x-ray generator. Yet it is apparent from the comments above, and especially from the range of science discussed in Chapters 2 and 3 that the use of neutron beams - unlike high energy physics where a big machine serves a relatively small number of 'big' experiments - affects many scientists in many disciplines and contributes to the solutions of a large number of 'small science' problems. The concept is more like the use of a large computer processing many different jobs.

1.7 Because of the restraint of the potential scientific programme imposed by the limitation in flux, the Neutron Beam Research Committee (NBRC), through a study programme carried out by the Neutron Beam Research Unit (NBRU), has maintained a continuing review of possible new neutron sources. Recently it has concentrated on the types of source which might give gains in intensity approaching 100 times today's highest fluxes and which would represent an improvement of neutron beam facilities in the 1980's which could serve at least to the year 2000. Chapter 4 summarises the general consideration of new sources which has led to the present proposal.

1.8 It has become apparent that conventional reactors are nearing their limits in terms of flux although it is possible to envisage, after a considerable development programme, gains of $\sim$ 10 using a fluid fuelled reactor. The cost however could hardly be less than £100M and a pulsed reactor of comparable performance - even if judged safe to operate in the UK - would also be prohibitively expensive.

1.9 The advantages of accelerator-based pulsed neutron sources for many experimental regimes have recently been clarified through assessments in this country and abroad. Experience on continuous reactors has shown that instruments based on continuous beams with energy selection by monochromating devices are roughly equivalent in performance with those based on chopped beams in which energy selection is carried out by time-of-flight methods. Instrument design studies confirm this rough equivalence in many circumstances, and so it may be considered that the intrinsic instantaneous brightness of a source is the vital factor, and that a pulsed source with favourable pulse characteristics may for many experiments be equivalent to a continuous reactor of mean brightness equal to the peak brightness of the pulsed source. There will also be a range of experiments where the time-averaged flux is the important variable, and the pulsed source then suffers a disadvantage.

1.10 Consideration of pulsed sources in the UK goes back to 1965 when a detailed technical and scientific examination by the UKAEA favoured the known technology of a high flux reactor over the uncertain technology of the then most promising alternative - an electron linac producing neutrons which are multiplied in a variable reactivity plutonium booster assembly. Since those early considerations of proton and electron accelerators, and in the light of subsequent developments, thinking on such sources has crystallised to a point where we can state that the SNS is the optimum facility which can be built at present irrespective of the favourable siting at the Rutherford Laboratory. The site and availability of plant and buildings at the Rutherford Laboratory must be considered a bonus, and do not of themselves determine the decision.

1.11 In pulsed sources, the primary source neutrons are only partially moderated in order to maintain suitably short pulse lengths. This gives, as well as a roughly Maxwellian distribution of energies corresponding approximately to the moderator temperature (cf a steady-state reactor), neutrons whose intensity at higher energies is inversely proportional to energy. This difference in approach precludes a straightforward comparison with reactor-based facilities. In broad terms however the assessments which have been made in Appendix I show that although the time-averaged intensity of the SNS falls below that of the highest flux reactors, except at relatively high energies, there are, for the reasons alluded to in paragraph 1.9, some remarkably large gains in *effective* flux. This arises from the necessity with continuous sources to discard most of the neutron flux to achieve appropriate energy selection; in pulsed sources the incident beam is inherently 'tailored' for time-of-flight measurement. For those instruments - the majority at present - which can make use of a broad band of incident wavelengths around 1 Å, there are order of magnitude gains over comparable instruments now operating at the ILL. At shorter wavelengths (higher energies) the advantage over ILL instruments is even more marked allowing for the first time the use of a high flux of neutrons with wavelengths less than 0.3 Å. As the wavelength increases beyond 1 Å the gains diminish steadily but we still envisage instruments with distinct advantages in performance even at 6-10 Å.

1.12 The scientific programme which could be carried out at the SNS has been examined by about 60 UK scientists, including representatives from over 20 different university departments, contributing to four main working groups.

In addition two general discussion meetings with a wider audience have been held at the Rutherford Laboratory. The reports of the working groups are summarised in Chapter 3 of this Proposal; the full reports are available in Appendix I.

1.13 The NBRU in collaboration with the Working Groups has compiled a list of 26 instruments (Appendix I) with specifications precise enough to proceed to the design stage. Fourteen basic types of instrument (close to the maximum number which can be accommodated at a single target station) are listed in Table 3.1 and these could cover almost all the scientific developments so far proposed. The precise determination of the instruments to be built and their priority will be determined after an up to date reassessment of the scientific programme and in the light of experience gained on the new Harwell linac. During the planning stage instruments of advanced conception to match novel applications are also likely to emerge.

1.14 Part of our confidence in the new source stems from the experience of UK scientists at the Harwell linac, the only pulsed accelerator in Europe used for neutron scattering studies. Even though the flux is more than 1000 times less than that of the SNS and only a minor share of the facility is available, useful scientific results have been obtained and several different types of instrument have been tested. The new Harwell linac will be an important intermediate step in the development of the Rutherford Laboratory facility, but its intensity (100 times less than the SNS) precludes most of the scientific programme proposed for the SNS.

1.15 The proposal demonstrates that the SNS achieves the features demanded of a next generation source, with gains in performance over a large part of the scientific programme which are larger - often considerably larger - than the differences between medium flux reactors built in the 1950's and 60's and the present generation of high flux reactors. It is recognised however that not all types of experiment benefit. Indeed, some which require high resolution simultaneously in both energy and direction, as in certain phonon and magnon studies, or experiments which make ingenious use of a continuous beam, as in the ultra high resolution spin echo technique, will continue to be best done at the ILL. It is the different time and energy distribution of the neutrons from the two sources which ensures a complementarity between the ILL reactor and the SNS. This is also illustrated by the new fields, such

as ultra cold neutron ($\lambda > 200$ Å) studies and neutron interferometry which are being developed at the ILL but which are not part of the programme proposed for the SNS.

1.16 The SNS and ILL together would provide unrivalled neutron beam facilities and the NBRC's Spallation Neutron Source Coordinating Group recommends that the SNS project begin at the Rutherford Laboratory early in 1977 with a view to starting the first experiments in 1982.

CHAPTER 2. NEUTRON SCATTERING IN THE UK

INTRODUCTION

2.1 Like many other particles and radiations the neutron, although discovered in the search for the ultimate nature of matter, has been utilized as a probe in a wide range of scientific activity. In this sense the properties of the neutron are being exploited as in their turn have been x-rays in crystallography, electrons in diffraction for all disciplines, microwaves in ESR, radio waves in NMR etc. The major difference between the exploitation of neutrons in this way and the other cases, is that to obtain a useful flux of neutrons a large investment is required in the source.

2.2 The characteristics of the neutron which make it so useful for studying condensed matter are:

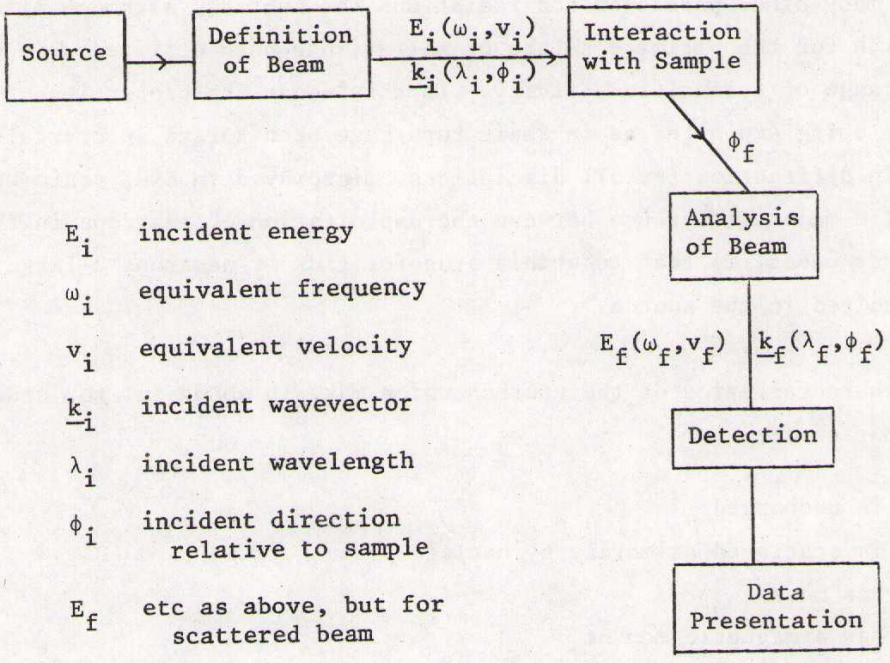
- it is uncharged
- it is scattered primarily by nuclei
- it has mass
- it has a magnetic moment
- it may be polarized
- it usually suffers low absorption (capture) and has a relatively large penetration of condensed matter such that large samples and a wide range of wavelengths may be utilized

2.3 It is this unique combination of characteristics which makes neutron beams such crucial probes for the study of condensed matter. Neutrons should

only be used when their special characteristics permit measurements that cannot be made more cheaply, and they are frequently used in conjunction with other experimental techniques. In order to give some general feel for the continuing and developing applicability of neutrons it is no exaggeration to say that their versatility in the study of matter is much greater than that of the generally more familiar x-rays. The scientific case that we are presenting, however, is no longer a justification in comparison with x-rays; that has been amply demonstrated in experiments using UKAEA medium flux reactors and the high flux reactor at the ILL.

THE PRINCIPLES OF THE NEUTRON SCATTERING METHOD

2.4 The basic nature of a typical neutron scattering experiment is simple, although the hardware may be complicated. The basic experiment may be illustrated in a block diagram as follows:



E_i incident energy

ω_i equivalent frequency

v_i equivalent velocity

$\underline{k}_i$ incident wavevector

λ_i incident wavelength

ϕ_i incident direction relative to sample

E_f etc as above, but for scattered beam

$$\varepsilon = E_f - E_i$$

$$\underline{Q} = \underline{k}_f - \underline{k}_i$$

In such an experiment the intensity scattered per incident neutron per unit solid angle per unit of energy change may be written as

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \frac{k_f}{k_i} \sum_{\substack{mn \\ if}} P_i \, b_m \, b_n \left| M_{if}(mn) \right|^2 \delta(\varepsilon - \hbar\omega) \qquad (1)$$

where $M_{if}(mn)$ is the matrix element for scattering involving atoms m (scattering length b_m) and n (b_n) between initial and final states i and f, P_i is the thermodynamic probability of finding the system in the initial state and the sum is over all pairs of atoms and all initial and final states. The δ-function is zero except when $\varepsilon = \hbar\omega$ when $\delta = 1$. For crystalline systems we describe the crystal by reciprocal lattice vectors $\underline{\tau}$ and the excitations by wavevector $\underline{q}$ and energy $\hbar\omega_q$ and (1) becomes of the form

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \frac{k_f}{k_i} \left| M(\underline{Q},\underline{\tau},\underline{q},\hbar\omega_{\underline{q}}) \right|^2 \delta(\underline{Q} - \underline{q} + \tau) \; \delta(\varepsilon - \hbar\omega_{\underline{q}}) \qquad (2)$$

In non-crystalline systems it is more convenient to express (1) by expressions of the type

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = N \frac{b^2}{\hbar} \frac{k_f}{k_i} S(\underline{Q},\omega) \qquad (3)$$

where $S(\underline{Q},\omega)$ transforms to $G(\underline{r},t)$, the space-time correlation function, which is a composite function including the motion of the same particle and the motion of pairs of particles.

2.5 Experiments may be *elastic* for which:

$$E_i = E_f, \quad \omega_i = \omega_f, \quad v_i = v_f;$$

$$\underline{k}_f - \underline{k}_i = Q, \quad |k_i| = |k_f|$$

and for $\phi_i = 0$ the scattering angle $\phi = \phi_f$ and $Q = \frac{4\pi \sin\phi/2}{\lambda}$.

Elastic experiments represent the structural mode and include all conventional crystallography, general structural diffuse scattering (including small angle scattering - better called small Q scattering) and the determination of atomic positions in disordered systems such as liquids, glasses, amorphous materials, polymers, biological membranes etc. A notable feature in the development of

elastic neutron scattering has been the progress from relatively simple structures to systems of considerable structural complexity. The structural mode also includes all the corresponding magnetic structural information ranging from the magnetic structure of ferro-, antiferro- and ferri-magnetic structures to the diffuse magnetic scattering associated with the distribution of magnetic moments surrounding magnetic ions in non-magnetic hosts and the associated ability to study covalency effects of general chemical - not specifically magnetic - relevance. The intensity scattered elastically into unit solid angle per incident neutron is measured and for the purposes of this discussion will be written as

$$\frac{d\sigma}{d\Omega} = \sum_{\substack{mn \\ if}} P_i b_m b_n \left| M_{if}(mn) \right|^2 .$$

Whatever the system, the information about the structure on atomic positions is contained in $|M_{if}(mn)|^2$ - ie, $|M(\underline{Q},\underline{\tau})|^2$ or $G(\underline{r},\infty)$ - and various procedures have been developed for unravelling this information. The analysis makes substantial use of modern computers and is usually performed at the scientist's home base rather than at the reactor centre.

2.6 Experiments may be *inelastic* for which:

$$E_i \neq E_f \text{ etc and } \varepsilon = E_f - E_i = E_{exc} = \hbar\omega_{\underline{q}}$$

where E_{exc} is an excitational energy of the system;

$$\underline{k}_i \neq \underline{k}_f \text{ and } |k_i| \neq |k_f|$$

and

$$\underline{Q} = \underline{k}_f - \underline{k}_i = \underline{q}$$

where $\underline{q}$ is the corresponding wavevector of the excitation. Furthermore selection rules exist such that from the intensities the nature of the mode of n collective excitations (phonons, magnons) may be deduced. The scattered intensity is measured per unit solid angle per unit of transferred energy per incident neutron and is given by equation (1) - or its alternatives (2) and (3). The excitation may be dynamical or magnetic - ie

motion of atoms or fluctuations in the directions of atomic magnetic moments - or, indeed, mixed. The information about the excitation is contained in $|M_{if}(mn)|^2$ - ie, $|M(\underline{k},\underline{\tau},\underline{q},\hbar\omega_{\underline{q}}|^2$ or $G(\underline{r},t)$ - and again various procedures for unravelling the information have been developed. The underlying physics of this unravelling process is relatively straightforward although in practice computer support is usually required.

2.7 In addition to the elastic/inelastic division, it is convenient to consider the *coherent* and *incoherent* parts of the cross-section. When the scattering is coherent, many-particle information is contained in the scattered beam allowing collective phenomena to be studied. When the scattering is incoherent, then information is obtained on the self correlation function, which describes the dynamical behaviour of individual molecules. The latter is particularly important in the molecular sciences, since the cross-section of hydrogen is both large and almost completely incoherent.

2.8 In inelastic experiments the information obtained is essentially spectroscopic and the spectroscopic mode includes:

- lattice vibrations in crystals (coherent scattering)
- instabilities of vibrational modes associated with the onset of phase changes (coherent)
- magnetic excitations (coherent)
- instabilities of magnetic modes associated with the onset of magnetic phase changes (coherent)
- motions of atoms in liquids and solutions (coherent and incoherent)
- general diffusion in matter (incoherent)
- molecular excitations (coherent and incoherent)
- polymer chain motion (coherent and incoherent)

2.9 In many neutron experiments where there is a magnetic interaction between the neutron moment and either an atomic or nuclear moment density, an increase in the sensitivity of the measurement of the interaction cross-section can be achieved by selecting the neutron spin state in the incident beam. When the polarized beam technique was first introduced it played an

important role in the determination of magnetic structures, but more recently it has been increasingly applied to the measurement of electron spin density distributions and is revealing more and more about the microscopic nature of magnetism. An extension of the technique to a spin analysis of the scattered beam (polarization analysis) enables spin-dependent cross-sections to be determined often in the presence of several other scattering processes; this type of experiment is, however, still in its infancy, and will benefit greatly from an increase in neutron intensity.

GENERAL CONSIDERATION OF INTENSITIES

2.10 The maximum thermal neutron flux Φ in nuclear reactors varies from $\sim 10^{12}$ n cm^{-2} sec^{-1} in small university based reactors to $1 - 2 \times 10^{15}$ in installations such as the ILL. This flux maximum in the reactor moderator is not available for neutron scattering experiments, since definition of the beam in terms of energy and direction reduces the intensity by factors of 10^{-6} to 10^{-11} depending on resolution requirements. The feasibility of an individual experiment further depends on factors such as the magnitude of the scattering from the sample (conceivably a factor of 10^{-1} but generally much less), and the need to match the energy/spatial resolution of the scattered and incident beams, which involves further losses in the range $10^{-4} - 10^{-6}$. For acceptable accuracy one usually needs to collect $\sim 10^{4}$ counts per counting element (more if the background is quite large) so that even with multidetectors (spatial) and time channelling (energy) to allow simultaneous counting in many resolution elements data collection rates are slow.

2.11 In practice reactors of $\Phi = 10^{12}$ are now quite unsuited for condensed matter research. With $\Phi = 10^{13}$ in the early days, some reasonably accurate structural work on simple structures could be pursued as well as simple phonon curves and diffuse scattering from samples with high impurity or defect concentrations. Most of the experiments currently carried out at the ILL would be impossible with $\Phi = 10^{13}$ and not realistically possible for $\Phi = 10^{14}$. At the ILL typical individual experiments range in time from $\sim$ 10 secs (small angle scattering with a 4×10^{3} multielement counter) enabling simple time dependences to be followed in favourable cases (eg alloys and polymers) to $\sim$ 10 weeks for high resolution, inelastic studies on relatively weakly scattering samples or complex crystal structures. In the latter studies the demand for the high flux facilities is such that to complete one phase of a major experimental programme may take several years even for proposals given the highest priority. In addition there is now also increasing interest in

examining *in situ* time dependent processes (in alloys, polymers, glasses, defect crystals etc) and the greatly enlarged scientific scope afforded by high resolution studies from weak scatterers (surfaces, diffusion studies, excitations in phase changes, studies involving polarization analysis etc). The implications are that the next generation source should have an effective $\Phi > 10^{16}$. The SNS is just such a source.

THE PROVISION OF NEUTRON SCATTERING FACILITIES FOR UK UNIVERSITY SCIENTISTS

2.12 The way in which neutron beams have been made available to a growing number of UK University scientists (see the table below) and the impact that neutron scattering studies have made first in physics, more recently in chemistry and biology, can be seen as one of the most successful developments in post-war science.

YEAR	UNIVERSITY STAFF	RES. FELLOWS AND ASSOCIATES	RESEARCH STUDENTS	TOTALS
1967	38	16	62	116
1968	44	24	63	131
1969	39	20	68	127
1970	47	25	76	148
1971	52	41	75	168
1972	59	46	68	173
1973	61	51	72	184
1974	94	52	70	216
1975	103	61	83	247

The important developments were:

1951 - 1965 Limited but growing *ad hoc* use by university scientists of UKAEA reactors, principally DIDO and PLUTO at Harwell with $\Phi = 7 \times 10^{13}$ (now $\sim 10^{14}$) and Herald at Aldermaston with $\Phi = 2 \times 10^{13}$.

1966 SRC sets up NBRC. 25% of the Harwell instruments available to university users, and three instruments at Aldermaston.

1967 France and Germany begin construction of the ILL reactor.

1968 Rental of Harwell instruments increased to 50%.

1971 SRC recommends construction of a high flux beam reactor ($\Phi = 2 \times 10^{15}$) in UK.

1971 Neutron Beam Research Unit created at the Rutherford Laboratory. The unit works in close collaboration with users both at Harwell and the ILL and is a centre for research and development covering a variety of aspects of neutron scattering. It has been the focus of the continuing discussion about the next generation source.

1971 Significant use by university scientists of Harwell linac commences.

1972 ILL reactor operational and the first instruments commissioned.

1973 SRC becomes a *de facto* associate of the ILL giving UK scientists access to about one third of the facilities which earlier, and conservatively, had been predicted as necessary for a wide exploitation of the use of neutron beams.

1975 Most of the ILL instruments working routinely, with a total of over 500 experiments performed ($\sim$ 200 involving UK scientists). Demand on several instruments substantially exceeding availability.

SCIENTIFIC DEVELOPMENTS

2.13 We conclude this general survey of the scope of the neutron scattering technique and its development in the UK by summarising the status in some of the important areas of application and indicating how these would benefit from the provision of the SNS. Such a summary must inevitably be subjective to some extent; the reader can find a full account of the programmes supported since 1967 by consulting the annual reports of the NBRC. The more specific conclusions of the Working Groups on the scientific gains which would accrue from using the SNS follow in Chapter 3.

Magnetism

2.14 Neutron scattering studies have revolutionized the subject of magnetism. After early success with the determination of the simpler magnetic structures quite complicated arrangements of atomic magnetic moments are now determined. The dynamics of these structures can be studied and the motions described in terms of the magnetic excitations or magnons of the structure (cf phonons in lattice dynamics). Measurement of these excitations by inelastic magnetic scattering has been one of the major successes of neutron scattering. Diffuse

magnetic scattering has been able to demonstrate the spread of magnetic influence around a magnetic impurity and this has important applications in probing electronic wave functions in non-magnetic hosts (eg, covalency). The spin glass systems are also being examined by diffuse scattering.

2.15 The future of this field is extremely promising both in relation to the present types of experiment and to the extensions, difficult at present, to higher energies and, by combining polarization and energy analysis, to the separation of magnetic and vibrational effects, especially above critical points. Whereas few phonon excitations occur above 0.1 eV, there exists a large number of magnetic materials exhibiting magnetism due to itinerant rather than localised electrons, whose collective excitations in the ordered state range into the 0.1 - 0.3 eV region. This region extends beyond the upper limit of energy-transfer currently observable using a hot-moderated steady-state source, but should be readily attainable with the SNS. Experiments of immediate interest include investigation of itinerant ferromagnets (eg Fe, Ni), where interaction with Stoner modes has been suggested by conventional experiments, and itinerant antiferromagnets (eg γ-Mn, Cr, NiS) whose steep energy dispersion has limited observations using conventional sources to excitations up to 0.14 eV and 2/3 q_{max}. Investigation of the generalised susceptibility $\chi(\underline{q},\omega)$ in transition metals is another area limited by current sources, the experiments requiring wide ranges of energy transfers without large momentum transfers. The exciting possibility of observing electronic band excitations in magnetic materials should also be mentioned. Cross-sections of only a few millibarns have been predicted, and experiments would need to be carried out with high energy neutrons, of a few eV, at relatively low Q, conditions only attainable on a source like the SNS.

2.16 Another type of experiment which presently suffers from a lack of neutron flux is that of polarization analysis in the measurement of diffuse and inelastic scattering cross-sections. The technique has potentially wide applications in the separation of nuclear spin-incoherent from coherent scattering. In magnetism, in addition to separating pure phonons and magnons, it can be used to identify magneto-vibrational scattering, where the phonons are excited through the magnetic interaction.

Liquids

2.17 The structure of liquids has to be represented by a probability distribution. At any instant the overall distribution will be the same but individual atoms will have moved in a given interval of time. They will diffuse and execute molecular motions (vibrations etc...). All these aspects may be determined by neutron scattering experiments. There is a profitable interaction between theory and experiment in this field and the ability comprehensively to determine probability distributions and the atomic dynamics from the 'microscopic' approach provided by neutron scattering has transformed the experimental study of liquids. Isotopes can be used to study structural and dynamic effects in binary (or higher) systems so that for example in a system AB the distributions r_{AB}, r_{AA} and r_{BB} may be separately determined. Such measurements require high intensities and many natural extensions of the present work, for example, AB, AA and BB motional correlations are barely possible. The work completed has shown the existence of molecular orientation in molecular liquids, structure in aqueous solutions and has allowed atomic distributions in metals and molten salts to be compared directly with the fundamental atom-atom interaction potentials. The structural aspect of this discussion applies also to glasses and the solid amorphous state generally.

2.18 The SNS will clearly serve wide areas of research in the field of fluids and amorphous materials. The case rests on the long-term, continuing programmes of investigation of the structure and dynamics of various classes of materials and their relevance to poorly understood properties. The scientific advantages arise through the ability to study wider ranges of composition of materials, and of materials over a wider range of the thermodynamic phase diagram, than is possible with existing sources. The extended Q range available with the SNS is particularly important for structural studies, providing new information on intramolecular properties. A wide selection of experiments can be immediately identified, for example, the study of aqueous solutions, and liquid alloys, aimed at understanding the correlation of the microscopic behaviour with macroscopic properties. The structure and dynamics of ^{3}He, ^{4}He and their mixtures should be studied to test the theoretical basis of our understanding of quantum fluids; this is particularly topical in the light of the recent discoveries of superfluid ^{3}He phases. The provision of the new source raises the possibility of carrying out kinetic experiments during phase separation and crystallisation processes, giving a new dimension to neutron beam studies.

Phase Changes

2.19 In the last decade there has been considerable interest in both the general formulation and in specific applications of the statistical mechanical and atomic theory of phase changes. The subject is all embracing and manifests itself for example in the helium λ-point, in ferroelectricity, in magnetism and in the classical first order change of melting. Neutron scattering has contributed substantially on the structural side. Many ferroelectric structures, for example, depend on hydrogen (or deuterium) displacements or displacements between ions of similar x-ray scattering power; in both these cases structural information from neutron scattering has been essential. However, in all cases one is interested in the way the instability occurs as the transition temperature is reached and the dynamics, whether motional or magnetic, are also studied by neutron scattering. A related problem is the persistence of order characteristic of temperatures below the transition temperature in local regions at temperatures above the transition temperature. This field is so large that experimentally it is only beginning to be explored. The addition of special perturbations and the separation of magnetic and atomic local ordering effects using polarized neutrons will need higher fluxes; the pulsed nature of the SNS is particularly important for high pressure investigations.

Polymers

2.20 Both structural and dynamic aspects of polymers have now been extensively studied by neutron scattering techniques. Structural work has been concerned primarily with determinations of the chain conformation in concentrated solution and in bulk samples (glassy, crystalline and rubber). Neutron scattering provides a quite unique tool for these studies because of the different coherent scattering cross-sections of H and D. Use of a small concentration of deuterated polymer in a matrix of protonated polymer (or vice versa) gives a scattering contrast which permits the coherent scattering from the individual molecules to be isolated (as for a true dilute solution) and hence enables molecular conformations to be determined. Such experiments established the validity of Flory's hypothesis that polymer chains in their own matrix have unperturbed dimensions. Experiments on crystalline polyethylene have shown that the dimensions of the chains in the crystal are not very different from those in the melt, implying that an individual molecule may form part of several folded crystalline regions in different layers.

2.21 The study of the dynamics of polymers is concerned with side-group motions and with the low frequency long range conformational motions of chains in solution and in the melt. Another important aspect of the study of polymer dynamics concerns the study of phonons. Where single crystals can be grown the information obtained in such work is like that for similar studies of any single crystal, the $\omega(\underline{q})$ relations across the Brillouin zone for modes of various symmetries propagating in specified crystal directions. This gives directly the elastic stiffnesses, for example along the perpendicular to the chains, and provides a very good probe for both intramolecular and intermolecular forces.

2.22 The enormous backlog of experiments and pressure of time available at ILL mean that a small angle scattering instrument of only comparable performance to those at ILL on the SNS would be important, especially in extending polymer work from scientific problems to those of a technological, and relatively more time-consuming, nature. With count rates improved by an order of magnitude however, entirely new experiments on the time dependence of chain conformation (relaxation phenomena) will be possible, opening up a whole new area of polymer science and technology. For inelastic scattering, the pulsed nature of the source will make possible the study of rotational isomerison in relaxing systems, which is especially important because the rotational isomeric model of the polymer chain underpins all accepted models concerning the influence of chemical structure.

Alloys and Materials

2.23 There are many systems showing structural disorder, although a lattice basis exists. For example, precipitation in alloys, defect arrangements in non-stoichiometric compounds and materials disordered by extensive radiation damage. These invariably exhibit diffuse scattering either at large or small angles, or indeed at both. Neutrons can penetrate reasonably sized samples compared with x-rays or electrons and moreover the much longer wavelengths which may be used have enabled much better information to be obtained. In many cases the ability to discriminate between elements of similar x-ray scattering power (eg, Al, Mg, Si) has also been exploited. A number of cases has now been observed in which there are significant anisotropies in the small angle scattering obtained from single crystal samples. These

effects are being related to the shape of the structural fluctuation producing the scattering and experiments are in progress on systems of technological importance. Another important aspect of the present work is the fact that in a few favourable cases the time dependence of ageing or dissolution can be followed so that the growth and decay of the precipitates (say) can be followed atomically.

2.24 An important consequence of the SNS will be the possibility of extending the range of materials, and conditions, under which experiments can be carried out. This is particularly important because of the technological implications of much of materials science. An important area of application is the study of voids and point defects caused by irradiation, where it should be possible with sufficiently high flux to follow annealing processes *in situ* as a function of temperature. It should also be possible to follow the effects of stress on the damage, and the pulsed nature of the beam may be used to investigate the microscopic nature of fatigue by synchronous loading of a sample.

Biophysics

2.25 The impact of neutrons in this field is in its infancy having effectively become possible with the fluxes available at the ILL and even there requiring in many cases long counting times. Information on structures characterized by large repeat distances is being obtained by small angle neutron diffraction from membranes, muscle and collagen. The use of different H_2O/D_2O mixtures to vary the contrast between a particle (or a particular component of a particle) and its surrounding medium allows the size, shape and internal structure of various biological units to be determined by small angle neutron scattering. Applications have been made to the study of viruses, ribosomal subunits, chromatin and such proteins as catalase and tubulin. The interest is such that a wide-ranging international meeting 'Neutron Scattering for the Analysis of Biological Structures' has recently been held at Brookhaven.

2.26 The immediate effect of a new source 'tomorrow' would be to provide facilities additional to those presently available, thus relieving the chronic bottlenecks in beam time and enabling a greater variety of experiments to be covered. Of particular importance will be the development of

studies involving selective deuteration, to give the distribution of proteins in mixed biological systems (eg ribosomes). Among the more ambitious experiments to be attempted with increased fluxes, one can foresee detailed inelastic measurements on biological systems, whereby rapid dynamical processes such as diffusional motions can be studied, while the pulsed nature of the source will be exploited to follow kinetic processes in 'real-time' experiments such as activation and relaxation of muscle, and activation by light of retinal membranes.

General Spectroscopy

2.27 There are large numbers of inelastic experiments carried out to determine energy levels of localized systems not obtainable by other types of spectroscopy. This may be because the selection rules for neutron induced transitions are allowed - where those for transitions induced by electromagnetic radiation are forbidden - as in chemical neutron spectroscopy; or because with rare earth metals or alloys the neutrons can penetrate the sample where electromagnetic radiation cannot. These fields are vast and neutron inelastic spectroscopy is part of the general spectroscopic armoury available to the chemist and physicist, although at present the energy range available is restricted to relatively low energy excitations in comparison with the range of infra-red and Raman spectroscopy. The SNS should enable neutron scattering to fully complement the range covered by the optical methods.

2.28 Closely related to many of these studies are the observations of the very small energy changes associated with diffusive motions, both translational and rotational, manifested by quasi-elastic scattering. Experiments of this type have led to major advances in the understanding of liquid and plastic crystalline phases and the behaviour of hydrogen in metals. A combination of quasi-elastic scattering and neutron spectroscopic techniques has been employed in studies of the vibrations and diffusional motions of adsorbed and intercalated molecules. Such investigations hold the promise of important contributions to surface chemistry but present-day fluxes have so far limited experiments to systems of relatively large surface area. A major function of the SNS will be to provide the possibility of high flux experiments with good resolution for such experiments.

CHAPTER 3. SCIENTIFIC APPLICATIONS OF THE SPALLATION NEUTRON SOURCE

INTRODUCTION

3.1 The general features of the SNS in relation to a high flux reactor have been described in Chapter 1, paragraph 1.11, and the estimated performance of the source is given in Chapter 5, paragraphs 5.12-5.19. The four Working Groups (Chapter 1, paragraph 1.12) have used the latter data in assessing the impact of the new source in the scientific areas of interest to the Science Board, and this chapter presents their conclusions. A group of instruments which might be used for the suggested science programme is summarised (paragraph 3.87) and the chapter also includes some possible other uses of the facility (paragraph 3.88). Appendix I contains the full reports of the Working Groups and preliminary specifications of possible instruments.

3.2 The practical advantages of the SNS can be summarised as follows:

- Wide regions of momentum and energy transfer become accessible to the experimenter. The available domain of $(Q,\hbar\omega)$ is expected to be between $(0.3\ \text{Å}^{-1}, 0\ \text{eV})$ and $(100\ \text{Å}^{-1}, \sim 0.5\ \text{eV})$
- Measurements can be made at higher rates of data collection
- Improved resolution will be available, particularly at higher energy transfers

- The use of smaller crystals or samples than hitherto becomes feasible

- Experimentation will also be possible on samples which are 'small' in the geometrical sense (eg surfaces or thin films) or in the chemical sense (eg dilute solution where the behaviour of the solute is of particular interest)

- The use of fixed scattering angle enables studies of samples to be undertaken under extreme conditions of temperature and pressure which are otherwise difficult or impossible

- The use of higher energies up to and beyond 1 eV enables studies to be made of highly absorbing samples and promotes the employment of anomalous scattering techniques

- Kinematic corrections at high incident energies, particularly for fluid systems, are smaller and better controlled.

3.3 In each of the scientific areas solid state physics, liquids and amorphous solids, structure determination, and chemistry, it is possible to identify experiments which depend on the SNS for their successful execution, and those which though feasible today would benefit markedly from the increased intensity. The latter are not necessarily inferior to the former, and particularly in chemistry or biology the ability to be able to complete a planned series of experiments may lead to scientific advances of greater significance than a single 'difficult' experiment. Examples of each type of experiment are headlined in the following paragraphs.

3.4 New experiments, or experiments that are barely possible at present, which can be performed with the SNS include:

- magnon studies in itinerant ferromagnets and antiferromagnets

- residual spin wave structure in ferromagnets above T_c

- the study of Kondo or spin fluctuating systems

- dispersion curves for electronic excitations in semiconductors

- magnon creation and annihilation studies

- the influence of fluid flow on liquid structure factors

- collision mechanisms from dynamical studies of dense gases
- the transition between binary collisions and collective mode coupling effects in simple fluids
- separation of coherent and self motions in liquid metals
- anomalous scattering techniques in the study of liquid alloys
- the structure of dilute electrolyte solutions
- molecular configurational changes in collisions between flexible molecules
- the structure of amorphous thin films
- phonon lifetimes in glasses
- structural, vibrational and rotational studies of molecules adsorbed on surfaces of area $\sim$ 1 m^2 or less
- high energy transfer vibrational spectroscopy - the energy range and the ease of data collection becomes comparable with infra-red techniques
- relaxation of chain conformations in polymers
- mechanical deformation - structure relationships in polymers
- very high pressure studies of crystal structure

3.5 Experiments currently feasible but given new scope by the higher intensity of the SNS:

- crystal and spin orbit splitting in metals and metallic compounds
- pair potentials and triplet distribution functions in simple fluids and their mixtures
- magnetism in liquid transition and rare-earth metals
- isotopic substitution techniques in aqueous solutions, molten salts and liquid alloys
- structural studies of water
- the structure of molecular liquids

- structural and dynamical studies of ^3He and ^3He-^4He mixtures

- kinetics of glass transitions

- collective motions in liquid crystals and intercalates

- mechanisms involving diffusion of non-hydrogenous atoms

- the spectroscopy of dilute and small samples, lipid bilayers, biological membranes, matrix isolated molecules etc

- kinetics of H-D exchange in biological systems

- the determination of the position of labelled sites in biological macromolecules in solution

- complex structure refinements by very high resolution powder diffraction

- simultaneous studies of the structural change and diffusion kinetics of host-molecule systems

- single crystal structure determination of polymers, and other small crystals

SOLID STATE PHYSICS

3.6 In discussing the impact of the SNS on solid state physics experiments it is convenient to classify these into (a) those which benefit from increased intensity, (b) experiments at the limits of feasibility at the ILL but which could be best carried out on SNS instruments on grounds of the spectral characterisation of the beam as well as flux, and (c) experiments which at present cannot be carried out at any neutron source in the world.

3.7 We have identified experiments in various areas of physics according to this classification, as follows:

'class (a)'

- phonon and magnon dispersion

- crystal field and spin-orbit splittings in paramagnets

- inhomogeneities in solids

'class (b)'

- generalised susceptibility $\chi(\underline{q},\omega)$
- magnon excitations in itinerant ferromagnets and antiferromagnetics
- separation of coherent/spin-incoherent and nuclear/magnetic scattering

'class (c)'

- high energy transitions
- inelastic polarization analysis

3.8 For 'class (b)' experiments, two instruments are suggested for the SNS which would extend the scope of experiments at present attempted only with difficulty. These are (a) a high energy (0.1 to 0.3 eV) inelastic instrument and (b) a total scattering polarization analysis instrument. The former instrument would contribute very significantly to magnon and crystal field studies and it would in addition open up the study of two other important problems in magnetism, viz measurements of the generalised susceptibility $\chi(\underline{Q},\omega)$ and the study of itinerant magnetism. Some experiments in this class are described in paragraphs 3.13 - 3.15 below.

3.9 With the high effective intensities available from the SNS, the use of inelastic polarization analysis techniques becomes possible, essentially for the first time. In addition, a favourable feature of the energy spectrum of the SNS is the intensity gains it provides at the higher neutron energies (1 to 10 eV), and these can be employed to carry out totally new high-energy low-Q inelastic experiments with energy transfers $\sim$ 0.3 to 1 eV. These experiments fall in 'class (c)' and are described in paragraphs 3.16 - 3.19.

Phonon and Magnon Dispersion

3.10 The unique property of thermal neutrons, that both their wavevector and energy match closely those of elementary excitations in solids, has been exploited extensively during the past 15 years to determine the energy dispersion throughout the Brillouin zone of magnons and phonons in many elements and compounds. These measurements have generally involved neutron

energy transfers (excitation energies) of 0-0.1 eV, and have mainly been performed on continuous or chopped beams from steady state reactors. They have yielded important and unique information on the interactions between the magnetic electrons, or between atoms or ions, in materials, and they have been extended to investigate details of the dynamical behaviour of spins and lattices near phase transitions. Information on excitation lifetimes, and the interactions between different kinds of excitations (such as external and internal modes, or magnon-phonon coupling) has also been obtained.

Crystal Field and Spin-Orbit Splittings in Paramagnets

3.11 Crystalline electric field levels in rare earth ions can be measured directly by neutron inelastic scattering. The method has played a key role in the study of metals, since photon spectroscopy is not applicable here, due to the interaction with conducting electrons. Although the lanthanide metals have received considerable attention, there has so far been little success in observing transitions in actinide materials, since here the 5f shell is on the outside of the atom, and the crystal field splittings are somewhat larger ($\sim$ 0.1 eV). Spin orbit splittings in the rare earths fall in the range 0.04 to 0.3 eV. These may be studied in the insulating compounds by optical spectroscopy, but again in the metals and in metallic compounds optical techniques are no longer appropriate. In one particular field, that of intermediate valence compounds (which are usually metallic), the measurement of transitions between spin orbit levels would be an extremely useful tool. Though the splittings are relatively small in SmS, in other rare earths showing intermediate valence behaviour eg, Ce, Tm and Yb, the splittings are much larger.

Inhomogeneities in Solids

3.12 Any deviation from order in a crystal lattice gives rise to scattering which is additional to Bragg scattering and is termed as diffuse. In many instances where this deviation is due to dislocations, point clusters, alloy precipitates, magnetic moment fluctuations, density changes etc, the inhomogeneity gives rise to a $\underline{Q}$-dependent scattering, which is generally best-observed at neutron wavelengths greater than that of the material being studied, so as to avoid multiple Bragg scattering, and at small

scattering angles. Some examples of areas now under study are radiation induced defects, alloy decomposition, non-stoichiometric defects, covalency, flux-line lattices in superconductors, polymers in solution and magnetic impurities. The study of magnetic defects is considerably enhanced by using spin polarized beams. Two major advantages of diffuse scattering experiments with a pulsed neutron beam are the possibility of separating elastic and inelastic scattering if the wavelength spread is limited, and the possibility of synchronising a perturbation of a sample with the neutron pulse and then studying the response of the sample by collecting the scattered neutrons from many such cycles. Possible areas of application are the study of voids, point defects and point defect clusters caused by irradiation, where it becomes possible to follow the annealing of the damage as a function of temperature. It should also be possible to follow the effects of stress on the damage and to investigate the microscopic nature of fatigue by synchronous loading of a sample.

Generalised Susceptibility $\chi(\underline{q},\omega)$

3.13 The investigations of the generalised susceptibility $\chi(\underline{q},\omega)$ in transition metals are of two broad types, (a) nearly magnetic materials like Pd, and (b) ferromagnets like Ni and Fe and antiferromagnets like Cr and Mn, above their ordering temperature. In category (a) calculations have been made of the neutron cross-sections for the case of Pd. These showed that the total response is spread over a wide energy range, of the order of the bandwidth of the d bands. However, many fascinating many-body effects (eg "the paramagnon peak") appear at energies below 0.1 eV at low temperatures. Attempts to study the magnetic scattering have not succeeded with present techniques since the 4d form factor of Pd drops rather rapidly, and with present sources wide ranges of energy transfers cannot be studied without large momentum transfers, though scattering has been observed from the narrow 4f band of αCe. Similar considerations apply to category (b) above. Recent measurements on Ni and Fe above T_c have shown a surprising amount of structure in the paramagnetic response. This has been interpreted as a residual spin wave branch which extends up to 0.1 eV, where the spin waves disappear below T_c. The origin of this scattering is not understood at present.

Magnon Excitations in Itinerant Ferromagnets and Antiferromagnets

3.14 Whereas few phonon excitations occur above 0.1 eV, there exists a large number of magnetic materials, mainly metallic or semi-metallic and exhibiting magnetism due to itinerant rather than localised electrons, whose collective excitations (magnons) in the ordered magnetic state range into the 0.1 - 0.3 eV region. This region extends beyond the upper limit of energy-transfer observable using a conventional triple-axis spectrometer on a beam from a hot-moderated steady-state source, and a number of recent experiments performed on such an instrument has indicated that it contains, tantalizingly, important and fundamental physical effects. Examples of possible experiments include the investigation of the itinerant ferromagnets nickel and iron, where sudden decreases in scattered intensity have suggested the possibility of the magnon interacting with the continuum band of Stoner modes near 0.1 eV, and the study of itinerant antiferromagnets such as γ-manganese, chromium and nickel sulphide, whose steep energy-dispersion (200, 450 and $\sim$ 450 meV Å respectively) has limited observation using conventional sources.

Separation of Coherent/Spin-Incoherent and Nuclear/Magnetic Scattering

3.15 Probably the most important application of a total scattering polarization analysis instrument would be in the study of paramagnetic or magnetic defect scattering. One of the main problems in the usual determination of paramagnetic cross-sections is to separate this from the other scattering processes viz, nuclear disorder, multiple Bragg, thermal diffuse, Bragg, and nuclear spin incoherent scattering. A measurement of the spin-flip scattering cross-section provides a powerful method for separating the paramagnetic scattering. One of the fields of study where an accurate evaluation of the paramagnetic cross-section would make a large impact is dilute magnetic alloys, where it can reveal the existence of the onset of long range magnetic order, interacting impurities and Kondo or spin fluctuating systems.

Inelastic Polarization Analysis

3.16 Many useful polarization analysis experiments such as those mentioned above can be adequately performed without energy analysis. When some

energy-analyser is included in a polarization analysis instrument, the accompanying decrease in flux is sufficient at present to make inelastic experiments effectively impossible. The provision of an inelastic polarization analysis instrument on the SNS will, however, enable a significant number of spin dynamics experiments, both in magnetic as well as non-magnetic materials to be carried out. Inelastic polarization analysis provides the most effective way of separating magnon and phonon scattering, since the latter is nuclear and coherent, hence always non-spin-flip. By selecting both the incident and scattering neutron spin states the observation of magnon creation and annihilation has been clearly demonstrated in the saturated ferromagnet $Fe_{2.5}Li_{0.5}O_4$, though it has not been possible to carry out experiments routinely. In addition to separating pure phonons and magnons, it is also possible to use the technique to identify magnetovibrational scattering, where the phonons are excited through the magnetic interaction, and which exhibits the same polarization-dependent effects as magnetic Bragg scattering.

High Energy Transitions

3.17 The role of neutrons in determining the frequency/wavevector dispersion curves for phonons (see previous section on phonon and magnon dispersion) derives from the ability in neutron experiments to measure, with sufficient accuracy, excitations in the range $0.02 < \hbar\omega < 0.1$ eV over a wavevector range $0 < \underline{q} < 5$ Å^{-1}; this enables energy dispersion information to be obtained within the first Brillouin zone. No other probe eg, x-rays, infra-red, visible light Raman scattering, allows such comprehensive data to be obtained; these other techniques yield information only for restricted regions of the Brillouin zone, especially near $\underline{q} = 0$ and the zone boundaries, and then not with unique assignment.

3.18 The same general problem occurs in the experimental determination of the dispersion curves for electronic excitations. An important class of such materials is the large group of semiconductors having band excitation energies $0.1 < \hbar\omega < 5$ eV throughout the Brillouin zone of $0 < \underline{q} < 5$ Å^{-1}. Methods currently available for studying these excitations include optical absorption, optical reflectivity, photo-emission, electron scattering, and cyclotron resonance, and these provide accurate information for specific parts of the Brillouin zone. However, no complete experimental determination of frequency/wavevector dispersion curves has been possible.

3.19 As occurred with the studies of vibrational excitations, major computing projects have been developed to interpolate between points, and a further recent initiative has been taken by a number of theoretical physicists in the UK using the Rutherford Laboratory IBM 360/195 computer. It is clear that the ability to measure experimentally more complete dispersion curves would be a major advance both in the study of the electronic states of semiconductors and in the growing possibilities of 'electronic design' of materials.

GASES, LIQUIDS AND AMORPHOUS SOLIDS

3.20 The study of fluids and amorphous solids by neutron scattering provides information on the structure and dynamics of the materials. The SNS will have a major impact on all aspects of this work; some of the main features are described below.

Structural and Dynamical Studies of Simple Fluids

3.21 The study of the structure of pure noble-gas fluids is at present heavily influenced by computer simulation which gives an unambiguous description of kinetics and structures of fluids interacting with realistic and hypothetical pair potentials. There is, however, a lack of very accurate measurements of the structure factor, S(Q), to compare with the computed models. Three regions of the p,V,T diagram of particular interest are:

- the dense fluid region, far from the triple point
- the liquid region, near to the triple point, and
- the region of the critical point.

Such information allows pair potentials to be calculated, as well as data related to the triplet distribution function and so provides an opportunity to test various theories of the fluid state. By the use of isotopic substitution the studies can be extended to include mixtures of the noble gases. Attempts to investigate distortion of the structure factor resulting from fluid flow, which would have an impact on the theory of transport properties, should also be worthwhile.

3.22 The SNS will make possible the accurate study of the dynamical structure factor of dense gases of atoms or molecules as a function of density. The scattering from non-interacting rigid molecules can be calculated exactly. Therefore by studying departures from ideal behaviour as a function of collision rate (by increasing pressure or temperature) it becomes possible to learn about the collision mechanisms themselves.

3.23 A fundamental problem in the theory of fluids is the treatment of the competition between simple binary collisions, which dominate at low densities, and collective mode-coupling effects which become increasingly important at high densities. These effects most strongly influence the transition region between hydrodynamic and kinetic behaviour. This transition region has been investigated at liquid densities using both neutrons and computer simulation, but the neutron data are at present limited. Light scattering has been used to investigate this transition in the low density region, but it is not useful for more dense systems. The intermediate density region has not yet been investigated thoroughly by any technique and there is a pressing need therefore to follow a system like ^{36}Ar through the whole density range.

3.24 The gains described above by using the SNS to study gases are equally applicable to studies of dynamics of liquids which are at present poorly understood. From constant low Q measurements with high energy resolution, information on translational (and rotational for molecular systems) diffusion is obtained while at higher Q the nature of the single particle motion is obtained. The validity of measurements possible with the SNS will enable studies on both natural and induced relaxation processes. Such studies are of particular value also for liquid metals, molten salts, aqueous solutions and molecular liquids.

Liquid Metals and Alloys

3.25 The outstanding problem in the physics of liquid metals is to determine the extent to which the distribution of the ions reflects and is influenced by the electron gas. In the simplest case, an effective pair potential, p(r), can be thought of as representing the interactions between the ions, appropriately screened by the electron gas. Self-consistent methods which relate p(r) to S(Q) require an accurate knowledge of S(Q), particularly in the region for $Q < 2\ \text{Å}^{-1}$, over ranges of temperature and pressure. The need to compare x-rays and neutron scattering to a high degree of accuracy has also recently been emphasised. This enables infor-

mation about electron-ion correlations to be obtained, knowledge of which is basic to our understanding of electronic properties, including under extreme conditions, those liquid metals which are known to undergo a metal-insulator transition. In alloys three broad types of behaviour have been observed:

- metallic behaviour with a statistical distribution of ions
- metallic behaviour with clustering of ions
- semi-conducting behaviour implying the existence of ionic, covalent or mixed bonds.

The occurrence of such diverse behaviour is not understood and arises from that area of science common to physicists, metallurgists and chemists. Again research programmes depend heavily on accurate data using isotopic substitution techniques although with the SNS it also becomes feasible to consider anomalous scattering close to resonance absorption edges as a method of extracting partial structure factors. This is a relatively new technique, but it opens up new and exciting possibilities for liquid alloy work, particularly for systems involving heavy elements and those for which no suitable stable isotopes exist.

3.26 A further application of either isotopic substitution or anomalous scattering is in the domain of alloy critical scattering. With more highly developed polarization techniques and an intense neutron beam, the nature of magnetism in liquid transition and rare-earth metals will be investigated. This subject is still in its infancy but is already beginning to reveal interesting new concepts.

3.27 The enhanced flux also enables one to consider, for a single element, the separate measurements of $S(Q,\omega)$ and $S_s(Q,\omega)$ (related to the coherent and self motions respectively) by inelastic polarization analysis. This technique could be applied in particular to a variety of liquid metals including sodium about which considerable theoretical and experimental knowledge has been built up over the last decade or so.

Molten Salts

3.28 The structure of molten salts (ie. disordered systems in which there is a strong interaction through Coulomb forces) is a topic of long standing interest. Fundamental problems in these types of liquid are:

- the short range repulsive part of the ion-ion interaction
- the role of polarization
- the mechanism of ion transport.

A programme to investigate the partial structure factors of molten salts is already under way in the UK and experiments yielding new information have already been carried out on the salts CuCl, NaCl and RbCl. Extension of this programme to other elements, and later, tertiary and more complicated systems, lies to a large extent outside the capabilities of present sources and requires the SNS.

Aqueous Solutions

3.29 Our knowledge of the structure of aqueous solutions has advanced following a series of experiments using the high flux reactor at Grenoble. From these experiments it has proved possible to determine the ion-solvent and ion-ion arrangement respectively but at present the investigations are limited to a small number of favourable isotopes where the degree of statistical uncertainty is minimised, eg saturated sodium chloride and nickel chloride solutions. The new source will enable a study to be undertaken on a wider variety of solutions and a greater range of dilutions, thus leading to a general picture of electrolyte structure including tests of the validity of a variety of computer simulated models. Because of the wider range of solute-solvent correlations, it will be possible to assess the degree to which various ions enhance or reduce the bulk water structure, a fundamental problem in electro-chemistry.

3.30 The new source is also ideally suited to the examination of the effects of pressure while the boost it should give polarisation analysis techniques - which in principle can give new insights in to the proton distribution in hydrogenous liquids - will have immediate importance for the structural study of water.

Molecular Fluids

3.31 Even the simplest molecular fluids, homonuclear diatomic molecules, are structually an order of magnitude more complicated than the noble-gas fluids, because of the additional degrees of freedom associated with the relative orientations of the particles. Neutron structure factor measurements are one means of studying orientational correlations in molecular

fluids. It has been possible with the present facilities to determine favourable configurations for pairs of molecules in liquid nitrogen, oxygen and bromine. However a thorough study of even such simple molecular systems over the thermodynamic phase diagram, combined with an extensive programme of computer simulation would increase our understanding of the liquid state. For more complex small molecules, studies of the structure of the dense gas phase, not possible with present intensities are desirable; modifications in molecular configuration due to collisions should be detectable, and information obtainable on the rigidity of the molecules.

3.32 Dynamical studies on simple molecular gases will prove rewarding both for the study of collision processes themselves and their effect on the internal motion of the molecules. It is also possible to envisage the study of the dynamical behaviour of metal dimers or other more highly associated groups of atoms or ions in the vapour phase.

Quantum Fluids

3.33 The study of liquid ^{3}He and liquid ^{4}He by neutron scattering shares with other liquids the general advantages to be gained from the use of the new source. Some of the relevant aspects are discussed in the following paragraphs.

3.34 As far as the static structure factor S(Q) is concerned, ^{4}He may, to a good approximation, be regarded as a dense classical fluid, but for a stringent test of theory accurate results over a wide Q range are required. As these theoretical approaches also give the population of the Bose Condensate - a quantity of central importance to the understanding of superfluidity - their critical appraisal is essential. The recently discovered superfluid phases of ^{3}He also creates an interest in the ^{3}He-^{4}He interaction which will stimulate structural studies on ^{3}He-^{4}He mixtures for several years.

3.35 Existing studies of the collective excitations in superfluid ^{4}He using neutrons, though extensive, have been significantly limited with present sources. For example, the behaviour of the one-phonon branch at scattering vectors beyond the roton minimum has long been of theoretical interest and recently of experimental interest, but it is highly desirable to improve both the statistics and resolution of existing measurements as well as extending them to higher wave vectors. The inelastic scattering behaviour is dominated by roton-roton interactions and accurate knowledge of lineshapes would allow a critical test of existing theories.

3.36 Although the interaction potential for ^{3}He is similar to that of ^{4}He, its collective excitation spectrum is fundamentally different. Unfortunately the capture cross-section of ^{3}He for thermal neutrons is extremely large and it is only recently that inelastic scattering has been observed and these results are limited. On the basis of theory the most prominent feature of the scattering from ^{3}He is the broad distribution of intensity that results from the excitation of particle-hole pairs. This is analogous to the Stoner excitations in itinerant ferromagnets. For large scattering vectors the continuum of excitations becomes independent of the particle scattering. As for ^{4}He, this region would be accessible with the new source.

Amorphous Solids

3.37 The structures of amorphous solids are again complicated and generally require accurate analysis of data in real space. Since the real space resolution obtained in any experiment is inversely proportional to Q_{max} the new source has a decisive advantage over steady state reactors.

3.38 Most inorganic glass structures are based on covalently bonded networks ranging from the simple one-dimensional chains found in selenium and many phosphates to the fully three-dimensional silicates. The glasses in common use are multicomponent systems (eg. Na_2O-CaO-SiO_2) and the elements present are not amenable to the technique of isotopic substitution. By far the most effective method of studying the structure of these glasses is by systematically varying the composition and a broad programme of structural analysis can be envisaged with the new source which will also allow extremely interesting kinetic studies of the glass transition region.

3.39 A variety of thin films can be made by vapour deposition (eg. Se, As-S, As-Se) and these have structures which are very dependent on preparation conditions. There is considerable interest in the extent to which the network structure of bulk glasses gives way in thin films to a more molecular character. So far the only neutron experiments on thin films have been on As-S and amorphous Ge, the major difficulty being the small amount of sample available. Similar considerations apply to other modes of preparation such as splat cooling and chemical deposition.

3.40 The SNS will encourage studies on the density of vibrational states and it will also be possible to study phonon lifetimes in glasses, which are related to low temperature anomalies.

STRUCTURE DETERMINATION

3.41 Most crystallographic studies form part of a wide investigation of physical, chemical or biological properties and the importance of the neutron part in this acquisition of basic structural data arises from

- the relatively small range of neutron scattering amplitudes which allows the ready detection of light atoms in the presence of atoms of high atomic number
- the irregular variation of scattering amplitudes which often generate significant fluctuations in scattering length between neighbouring atoms in the Periodic Table
- the fact that in many cases absorption is small (compared with x-rays) so that measurements down to liquid helium temperatures or up to 1500°C can be as routine as at room temperature. Experiments at even higher temperatures and at high pressure may also be performed
- the magnetic moment for magnetic structure determinations and magnetic moment distributions.

3.42 The SNS has three distinct advantages which can be employed in opening new areas of structure determination. These are

- the time-of-flight techniques used in conjunction with a white beam which facilitate the study of samples in special environments
- the very high rates of data collection in conjunction with the simultaneous recording of all or a large part of the diffraction pattern, which adds a further dimension to the study of time-dependent processes such as reaction mechanisms and transient phenomena
- the opportunity to examine relaxation processes in the solid state by the synchronisation of an external perturbation with the pulse frequency of the beam. We note that some of these features are also offered by x-ray radiation from a synchrotron source.

However, apart from the general advantages of neutron scattering referred to above, the troublesome preferred orientation effects in powders are much more pronounced for x-rays and the energy dispersive analysis of the synchrotron x-radiation has poor resolution compared with neutron time-of-flight methods.

3.43 Some of the new experiments based on the features in the preceding paragraph which will follow the introduction of the SNS are:

- high pressure experiments up to 100 kbars for studies of phase transitions, geophysical problems, and new phases which exist only under high pressure conditions
- accurate structure determinations at very high temperatures up to 3000°C
- structural changes associated with the application of a small periodic electric field to a piezoelectric crystal (or ceramic) or to a liquid crystal
- the mechanism of dipole switching with ferroelectrics (one such experiment on $NaNO_2$ has already been demonstrated)
- studies of metamagnetic phase transition using small rapid oscillations of a magnetic field
- speculative investigations of muscle contraction effects and structural changes associated with shock waves
- structural studies (complemented by neutron topography and particle size analysis) of reaction mechanisms.

3.44 In conventional crystallography - particularly involving complex chemical or biochemical crystals, the desire to include neutron diffraction (when appropriate) in the attack on the structure is frequently frustrated by the lack of crystals of a suitable size. Moreover, the delicate preparative conditions or the crystal morphology often prevent larger crystals being grown. In some cases involving relatively simple structures the problem can be overcome by using powders and employing profile methods to analyse the diffraction pattern. But either the resolution or the data collection rate is restricted with present fluxes. The improvements possible with the SNS in both single crystal and powder techniques are given below.

Single Crystal Studies

3.45 It is clear that the high intensity of the SNS, coupled with a large area position sensitive detector, would allow single crystal structure

factors to be measured as a function of wavelength and at rates some two orders of magnitude higher than at the most intense reactor source currently available. A feature of the SNS is the extension of data to high values of sin θ/λ. This is vital to many structural problems involving the study of positional parameter changes at phase transitions, electron distributions from x-ray/neutron data, and anisotropic and anharmonic thermal vibrations. The measurement of the wavelength dependence of strong structure factors is the surest way of obtaining the best estimate of their extinction-free values. The same basic instrument can be used for:

- high resolution structural studies of materials with simple structures having cell dimensions of 5 - 10 Å (ambient moderator)
- normal structural studies cell dimensions 10 - 20 Å, wavelengths 0.5 - 1.5 Å (ambient moderator)
- structural studies of complex molecules with large unit cells having dimensions up to 40 Å using wavelengths in the range 1 - 6 Å (cooled moderator).

Studies of magnetisation density distributions in materials with cell dimensions in the range 5-20 Å can also be carried out with the insertion of a polarization filter and Drabkin flipper into the incoming neutron beam with similar improvements in the data collection rate. In both nuclear and magnetic crystallography many experimentalists will need to exchange the possibility of a greatly enhanced data collection rate for a reduction in specimen size.

3.46 One aspect of single crystal structure determinations by time-of-flight methods is that each structure factor is readily obtained at a number of different wavelengths, and when the crystal contains strongly absorbing nuclei and anomalous dispersion effects are present it is possible to solve the well known phase problem. The data collection rate of the SNS will increase the precision with which weak anomalous scattering effects can be measured and encourage the use of the technique as part of the general armoury employed in single crystal structure determination. New techniques like the diffraction of polarised neutrons from dynamically polarised targets will also benefit from the enhanced flux.

Powder Studies

3.47 The recent remarkable profile analysis methods applied to powder

samples now enable refinements to be carried out routinely on structures with 20-30 structural parameters and unit cell volumes up to 1000 $Å^3$. Apart from large gains in effective flux the SNS provides the possibility of very high resolution at reasonable counting rates by the use of the back scattering mode. Two types of diffractometer are envisaged to exploit the SNS. A high resolution diffractometer would very considerably extend the power of powder methods in structure determination. The resolution could be 2-3 times better than conventional diffractometers and only slowly dependent on $\sin \theta/\lambda$. With such an instrument the refinement of structures with at least 100 structural parameters will be possible, over a very wide range of experimental conditions. Other applications, for single crystals as well as powders, are the observation of phase transitions involving very small changes of symmetry, critical scattering associated with a phase transition or the diffuse scattering associated with point defects in the sample. Particle line broadening effects will be observable and the instrument could be used for the study of small amounts of precipitated phase occluded in the bulk material. A moderate resolution instrument with very rapid data collection would be used to exploit the kinetic processes.

3.48 The applications of conventional crystal structure determination on the SNS will reflect the scientific topics of importance in the 1980's. To a greater extent than some of the other sections the neutron measurements are supplementary though that is not to underestimate their importance. To illustrate the general scope of neutron diffraction studies, some current examples are given in fields which will benefit from the SNS.

Phase Transitions

3.49 The structure of several important ferroelectric (and piezoelectric) materials are being intensively studied. Often the structural behaviour is complex; in the $Na_xK_{1-x}NbO_3$ system for example there are twenty different phases, half of them being ferroelectric with a high spontaneous polarization - and the crystallographic study is a necessary first step before an examination of the lattice dynamics. Marked changes in electrical conductivity may be associated with a phase change, and the structural changes occurring with metallic-semiconductor transitions feature in recent reports. Sometimes the transition of interest is one which confers enhanced ionic conductivity and the structural properties of a number of these superionic conductors are being investigated.

3.50 Most of our accurate information about hydrogen bonded structures has been derived from single crystal neutron diffraction studies. Current examples of this work include the clustering of water molecules about H_3O^+, the study of very short hydrogen bonds and the exploration of less common hydrogen bond interactions for example with acceptor phenyl rings. Attempts are also being made to elucidate the detailed hydrogen bonding scheme for molecular crystals which can serve as a model for the packing of side chains in globular proteins. More investigations of this kind should prove rewarding.

Molecular Crystals

3.51 The relative ease with which accurate neutron diffraction experiments can be carried out at low temperatures, coupled with the rapid development of profile analysis techniques applicable to polycrystalline samples, is stimulating considerable interest in the structures of molecular crystals. Generally there are subtle structural changes with variations in temperature and pressure. We can expect this type of study to extend our understanding of intermolecular forces not only in simple plastic crystals or hydrogen bonded systems but also to the large class of ionic compounds (eg hexafluorides) whose behaviour is intermediate between molecular crystals and simple salts. In selected cases the structural determination will be a necessary prelude to a complete dynamical investigation. The extension to more complex moelcular systems, eg organometallic compounds, pharmacologically important organic molecules and simple polymeric compounds is readily envisaged but it is frequently impossible to obtain crystals of sufficient size (for present fluxes) or sufficient instrumental resolution (also limited by flux) for powder experiments.

Defect and Disordered Solids

3.52 The increasing emphasis in recent years on the science of materials has created considerable interest in the structure of inorganic solids including ceramics. Many of the important compounds contain light atoms and frequently the high temperature phases demonstrate broad ranges of non-stoichiometry. Some aspects of the ultra microstructure - short range order

and clustering of defects - is revealed by diffuse or small angle neutron scattering and by electron microscopy, but the contents of the average unit cell can often only be accurately determined by neutron Bragg diffraction. The main leads to our understanding of non-stoichiometry in simple compounds are derived from such investigations and have stimulated considerable theoretical effort. Even so only a small number of structural types has been studied over limited concentration and temperature ranges. Non-stoichiometric compounds represent just one example of the very widespread occurrence of order-disorder phenomena in solids, many of which are profitably studied by neutrons.

Intercalation and Surface Studies

3.53 A feature of recent crystallographic studies has been the exploration of host-guest structures with host lattices such as the transition metal dichalcogenides and the clays. The molecular orientation of pyridine in NbS_2 is one recent determination which gives an unexpected result, but the range of inclusion compounds in this class alone is enormous and we still understand little about the host-guest interaction. In addition these layer hosts allow the examination of the properties of two dimensional monolayers, and could serve as templates for 'tailored' chemical reactions.

3.54 As well as the examination of occluded layers, present fluxes just enable - in favourable cases - direct structural investigation of simple gases on high surface area solids (eg graphite). Microcrystalline clusters are observed and in krypton for instance two solid phases are observed on a layer related to the graphite lattice and a compressed phase with Kr-Kr distances close to those in the pure solid. With the SNS the structure of surface layers of relatively low area will be possible.

Electron Distributions

3.55 It is possible from accurate x-ray and neutron data to derive electron distributions.

Magnetic Structure, Moment Distributions and Covalency

3.56 The interaction of the magnetic moment of the neutron with the magnetization density resulting from unpaired electrons in solids has led

to the determination of hundreds of magnetic structures over the last twenty years. Such investigations will continue to form part of future programmes in magnetic crystallography particularly in connection with novel materials. Much greater emphasis however will be placed in future on the more detailed information which is available by the use of polarised neutrons. In the past complete magnetization density distributions have been restricted to a limited number of relatively simple systems where large crystals were available. Now we can envisage charting the moment distribution for a much wider variety of compounds which can include free radicals. In alloys the information obtained can relate the observed macroscopic behaviour to atomic properties; thus in YCo_5 which exhibits abnormally high magnetic anisotropy it has been shown that this is associated with the extended spin density of Co (in one of the two Co sites) in the basal plane containing Y atoms. For inorganic salts the often marked aspherical moment distribution is related to the extent and nature of the covalent interaction and the measurements, apart from being able to demonstrate effects such as spin polarisation, provide illuminating guidelines for theoretical studies. These moment density maps are much more revealing than the simple determination of atomic moments which have been common hitherto. The evidence from the ILL suggests that with additional and more intense polarised beam facilities there would be very little restriction on the type of ligand (including complexes) or on the d and f block elements which could be studied.

3.57 Parallel with the greatly extended exploration of unpaired electron distributions there is bound to be a more sophisticated probing of magnetic interactions though a study of magnetic phase diagrams, the influence of pressure and the temperature dependence of correlation lengths in magnetic chains.

MOLECULAR SCIENCES - CHEMISTRY

3.58 The chemistry programme is already very diverse. Neutron scattering investigations provide information about the diffusive and rotational motions of molcular systems (quasi-elastic scattering), molecular energy levels (inelastic scattering), the conformation of polymer molecules and the shape and internal structure of biological particles (small angle scattering) and in addition a number of the chemical applications of neutron scattering has already been considered under other headings (Liquids, Structure Determination, etc). Some examples of the current research and likely future developments are outlined in the following sections.

Atomic Diffusion

3.59 The principal area of study here is the important one of hydrogen in metals and in this case the SNS by extending high resolution measurements beyond the first reciprocal lattice points will provide unique information on the diffusion mechanism. There are obvious gains for the study of very low hydrogen concentrations and measurements at low temperature. An important development will be the extension to nuclei with cross-sections small relative to that of the proton. Preliminary experiments have been carried out at the ILL on diffusion in sodium metal and some superionics, but under difficult conditions. The study of carbon in metals or alkali ions in glasses have been proposed as future experiments.

Molecular Crystals

3.60 The study of phonons by coherent scattering from single crystals can give the most definitive information on intermolecular forces. Detailed studies are likely to continue with triple-axis instruments on a reactor, but the SNS has, with the constant-Q spectrometer, a distinct advantage in searching for phonons. This is especially important for investigations of complex molecular crystals and polymers which may have to be carried out without prior calculation of the expected dispersion surfaces and inelastic structure factors. The biggest problem in this area is the growth of suitable single crystals. Higher fluxes will enable smaller crystals to be used, thus allowing a much wider class of materials to be studied.

3.61 The angular part of the intermolecular potential function is of particular importance in many molecular crystals which undergo transitions to orientationally disordered phases. Intramolecular potentials for the rotation of groups within molecules are also of interest. Both these fields can be very effectively studied using both quasi-elastic and inelastic incoherent scattering. Such investigations have already been made for a wide variety of systems, ranging from simple molecules like NH_3, to polymers. In developing this work further, there is a need for

- resolution good enough to separate elastic and quasi-elastic components of the scattering
- a large Q range to provide a sufficiently detailed description of the motion

- instruments of variable resolution and energy scans to cover the wide range of correlation times expected.

All these requirements can be met by the SNS.

Liquid Crystals

3.62 Considerable progress has already been made in the characterisation of the many liquid crystal phases in terms of the self-correlation functions for translational and rotational motion of the molecules. However, little work has been done on the direct study of phase transitions - a problem of general interest. The investigation of collective molecular motions in such systems has hardly begun and is hampered by low inelastic structure factors and the need for fully deuterated single crystal specimens.

Molecular Spectroscopy

3.63 This area is concerned with incoherent inelastic scattering, which, because of the proton's cross-section is essentially the molecular spectroscopy of vibrations involving proton motions. Its development has been hampered by low neutron intensities resulting in data collection rates which are very low in comparison with infra-red spectroscopy, and also by the necessary use of low resolution spectrometers. Increases in flux by a factor of 100 or more thus imply short run times or much improved resolution. This opens up a whole new range of possible applications, for example, in repeated scanning for kinetic experiments where the ease of data collection should be comparable with current infra-red techniques.

3.64 Some systems (eg NH_4ClO_4) show sharp excitations due to tunnelling modes at very low energies, of the order of a few μeV. The availability of a very high resolution spectrometer with a wide energy window will enable the study of a much greater variety of systems. For the first time also, it will be possible to obtain information on rotational and translational dynamics by analysing inelastic vibrational band shapes (cf infra-red and Raman spectroscopy).

High Energy Transfer Vibrational and Electronic Spectroscopy

3.65 With reactor sources there is an upper limit to measurements in energy transfer (generally < 250 meV). The SNS will open up the possibility

of measurements (eg of electron transitions or vibrational harmonics) involving energies greater than 600 meV, but the principal gain will come from the very large increases in flux up to 600 meV.

3.66 The most important application here is the study of hydrogen stretching frequencies which are largely inaccessible on current instruments. In general this implies a study of X-H bonds, frequently in absorbed or occluded phases. Specific instances include:

- The study of the vibrations of hydrogen chemisorbed on a surface, since for many metals the M-H stretching vibration may lie above 200 meV
- The study of metal-hydrogen stretching vibrations in transition metal hydride compounds. It is frequently difficult to identify the stretching frequency in these compounds by conventional optical techniques
- The study of the hydrogen stretching frequency in hydrogen-bonded systems where detailed measurements of the intensity of inelastic neutron scattering will provide information about the potential well in which the hydrogen is moving
- At higher energies the measurements of the relative intensities of overtone modes derived from hydrogen stretching frequencies should enable a much more precise definition of the potential energy curve in X-H bonds. The study of overtone modes in hydrogen in transition metal lattices is also of considerable importance.

3.67 The measurement of crystal field transitions, which up to now has been confined to the f-block compounds, will be possible on the SNS for d-block systems (see also paragraph 3.11).

Surface Chemistry and Other Dilute Systems

3.68 In the study of dilute systems the high counting rates and low background of the SNS will open up exciting areas. The most important field is surface chemistry. Studies already performed on existing sources include:

- the structure and dynamics of physisorbed species
- excitations of hydrogenous molecules chemisorbed on various substrates
- the dynamics of intercalated molecules.

The high effective intensity will make possible experiments with the same accuracy on samples which are up to a thousand times poorer scatterers than

can be currently examined. In favourable cases such as the study of hydrogenous species absorbed on the surface of a crystal with small incoherent scattering, it will be possible to study monolayers down to substrate areas as small as 1 m^2 or less, improving our understanding of catalysis. It will also be possible to study other systems such as multiple water layers on silver iodide crystals, and non hydrogenous species on graphite or concentrated colloids.

3.69 Examples of other dilute systems which will be open to study are:

- lipid bilayers and biological membranes
- matrix isolated unstable species currently studied almost exclusively by infra-red techniques
- species, eg NH_4^+, isolated at low concentration in simple ionic lattices
- hydrogen in metals at low concentration, including metals such as steel where there is considerable technological interest.

Polarisation Analysis

3.70 The major chemical application of polarisation analysis will be the separation of coherent from spin-incoherent scattering. This may be used in two ways:

- the determination of structures of systems which have a large, incoherent background, eg materials which are very difficult to deuterate completely, and biological materials in *in vivo* conditions, ie in light water
- the reverse application - the study of self-correlation function in the presence of a significant coherent signal.

MOLECULAR SCIENCES - POLYMERS

3.71 The application of neutron scattering techniques to polymerised systems has involved the study of rubbers, glasses, partially crystalline materials and polymer solutions. Apart from the intrinsic interest in relationships between chemical structure and physical property for materials composed of long chain molecules there are very important technological applications.

3.72 Neutron scattering has made a unique contribution to the understanding of the structural conformations of polymer molecules and to their molecular dynamics. This is due to the particular wavelength - energy characteristics of neutron beams, and also to the enormous difference in scattering cross-section of hydrogen and deuterium combined with the fact that these isotopes can be substituted for each other with little effect on the general physical properties of the materials. The phenomena of principle interest are outlined below.

Small Angle Neutron Scattering (SANS)

3.73 Most of the work in this field has so far been carried out using D11A at ILL. The work has extended our knowledge of polymer chain conformation into polymers in bulk. Light scattering and small angle x-ray scattering techniques, which hitherto provided all available information, are only applicable to polymer solutions. Neutron scattering results have in the past three years raised Flory's hypotheses on chain conformation in rubbers and glasses (the basis of all theories of amorphous polymers) to the status of fact. Equally important information on the effects of bulk deformations and also of crystallisation on chain conformations is now beginning to emerge.

3.74 The enormous backlog of experiments and pressure on time available at ILL means that an additional SANS instrument even if only of comparable efficiency would be important especially in extending the polymer work to technological problems. However, with count rates improved by an order of magnitude, entirely new experiments on the time dependence of chain conformations (relaxation phenomena) will be possible. This would enable the asymmetry of scattering to be measured as a function of time in a sample undergoing bulk relaxation on a technological time scale. Thus a whole new area of polymer science and technology could be opened up.

Diffraction at Larger Scattering Angles

3.75 The determination of radial distribution functions in the high Q region will give information on local molecular packing in both crystalline and amorphous states. Even more important might be kinetic studies made in this range using a pulsed source while the sample is subjected to relatively low frequency mechanical deformation. The very high epithermal flux should make such kinetic experiments easily possible.

Quasi-Elastic Scattering

3.76 Some of the conformational reorientational motions of polymer chains associated with the onset of main chain motion at the glass-rubber transition, or occurring in polymers in solution, have been investigated by quasi-elastic scattering. In solution, for example, the results support the Zimm model (with hydrodynamic interactions) rather than the Rouse model. The range of interest is $0.01 < Q < 1.0$ Å^{-1} and $\Delta\hbar\omega \sim 1$ μeV. Extension of this work is seriously hindered by pressure of time on ILL instruments and this entirely prevents extension to technological problems. The quasi-elastic instruments proposed would be invaluable in extending the work on rubbers and gels, which in the current situation, will probably be restricted to model systems only.

Inelastic Scattering

3.77 In amorphous systems inelastic neutron scattering has been used mainly to study side-group motions. Measurements on crystalline polymers have involved essentially powders, stretch oriented specimens and in two or three instances specimens approximating to single crystals having macroscopic dimensions. Phonon frequencies have been measured and using perdeuterated samples dispersion curves have been determined. Generally, cold neutron time-of-flight instruments have been used together with triple axis spectrometers for the single crystal work.

3.78 In the single crystal work advance is limited by crystal growth technology rather than by neutron fluxes, but the much greater effective fluxes available with the SNS would make possible the use of significantly smaller crystals. The new source could provide additional capacity for the remaining inelastic work with the possibility of much improved resolution being especially valuable. The pulsed nature of the source would make possible the study of rotational isomerism in relaxing systems, which is important because the rotational isomeric model of the polymer chain is the one which underpins all accepted models concerning the influence of chemical structure.

MOLECULAR SCIENCES - BIOLOGY

3.79 Broadly, the experiments of interest can be classified as follows:

- Studies of single crystals of macromolecules (eg proteins such as myoglobin, lysozyme etc and nucleotides). Here the wavelength range is 1 to 2 Å
- Studies of semi-crystalline macromolecules where Bragg diffraction peaks are observed even though there is inherent disorder (eg fibre diffraction from proteins and DNA, diffraction from collagen, muscle and membranes). Typically 4 to 12 Å neutrons would be involved with these experiments and small angle scattering equipment (eg D11A or D17 at ILL) used
- Neutron quasi-elastic and inelastic scattering studies of the dynamical properties of biological molecules and macromolecular structure
- Small angle scattering studies of separated macromolecules in solution (λ = 4 to 12 Å)
- Kinetic experiments using the pulsed nature of the source (λ = 1 to 12 Å say).

Small Angle Neutron Scattering from Biological Particles in Solution

3.80 These experiments enable the determination of the spherically averaged Patterson function of the macromolecules in contrast against scattering from the solvent. In the absence of crystals of the macromolecules this information is vital to the determination of the low resolution structure and the technique requires that the solvent scattering-length density (usually water) is changed by choosing different D_2O/H_2O mixtures for the solvent and by choosing different small molecule concentrations. Because of the coherent elastic scattering-length-density difference between D_2O and H_2O a variety of contrast conditions are achieved and one can obtain 'fundamental scattering functions' which are related to:

- the outer profile of the particle
- the internal structure
- a 'cross' term of the inner and outer structures
- the distribution of D/H sites within the particle.

Measurements at low angles, in the so-called Guinier region, yield information on the spherically symmetrical component of the scattering-length

density distribution within the macromolecules, but there is interest developing in higher angle measurements, where the deviations from spherical symmetry can be determined and where the details of the internal structure of the particles can be seen.

3.81 Deuteration of the macromolecules at non-labile sites permits the determination of the position of the deuterated sites within the structure. One technique, the so-called triangulation method, involves accurate measurements of differences of scattered intensity as a function of Q for two solutions. The first solution is a mixture of ribosome subunits with both proteins deuterated and both proteins protonated. The second solution is a mixture of ribosome subunits containing the first protein protonated and the second deuterated and subunits with the first protein deuterated and the second protonated. An interference function in the measured difference of intensities as a function of Q gives information about the distance between the scattering length centres of the proteins and a distribution of length between extreme surfaces of the two proteins. By obtaining information on a large number of proteins in the ribosome subunits it should be possible to determine the shape of the proteins. Very high statistics for the scattered neutrons are necessary and the ribosome experiments will take several years to complete so that the value of the increase in flux is obvious.

3.82 In studying dispersions of biological membranes and membrane components in solution the neutron scattered intensity falls off more rapidly than for globular particles in solution. In this case again therefore, higher neutron fluxes provide an important advantage.

Small Angle Studies of Bragg Diffraction in Semi-Crystalline Biological Specimens

3.83 Neutron studies of small-angle Bragg peaks in specimens such as muscle, collagen, biological membranes, are revealing much information on the intact systems and on changes on activating a given system. For example, differences between contracted and relaxed muscle, and between retinal membranes in the dark and activated by light, may be studied. It is also possible to derive information on how the various parts of a structure contribute to the Fourier harmonic corresponding to the amplitude of a given Bragg peak. Many of these specimens are quite highly oriented and therefore information is available at particular Q values. Clearly

information would be lost on spherically averaging the neutron data from an area detector (in an attempt to improve statistics) so that better count rates are a great help in studying the details of a particular Bragg peak.

Studies of Single Crystals of Macromolecules

3.84 Studies of crystals of biological macromolecules using neutrons are in progress both at the ILL and at Brookhaven, and have already provided useful information about hydrogen bonding and the detailed structures of the macromolecular building blocks of amino acids and nucleotides. If it were possible (by virtue of a very high flux of neutrons of $\sim$ 1.5 Å wavelength) to carry out a complete structure determination in one or two weeks, one could look in fine detail at D/H exchange at highly localised sites in the structure. Work at high pressures with cooled samples will reduce the thermal motion in the crystals and enable the full potential of the high resolution capabilities of neutrons to be obtained. These experiments will clearly gain from the high flux of neutrons at 1.5 Å wavelength which would be available from the SNS. The disadvantage of these experiments is that the measurements are not carried out *in vivo*, but there is the potential for a wealth of information of biological importance.

Neutron Quasi-Elastic and Inelastic Scattering

3.85 In biological studies this work has investigated the normal modes of vibration and force constants of poly-α-amino acids. At present the studies have concentrated on polyglycine (the simplest amino acid). More complex systems are now beginning to be investigated. These neutron studies supplement infra-red and Raman studies because they are most sensitive to displacements of hydrogen atoms in vibrational modes and are not restricted by optical selection rules. In this area the possibility of performing an energy analysis in conjunction with small-angle scattering is very attractive.

Kinetic Experiments Using the Pulsed Nature of the New Source

3.86 There is a developing interest in the dynamical aspects of biological processes, eg the kinetics of enzyme action, the rates of H $\rightleftarrows$ D exchange within a structure, changes of structure with electrical signals, magnetic fields, exposure to light, temperature, pressure etc.. These are all studies which will be stimulated by the availability of the SNS.

3.87 A wide diversity of instruments has been considered for carrying out research at pulsed sources. In Appendix I a comprehensive list of 26 instruments has been compiled, from which it will be possible to select the most suitable instruments for the SNS to satisfy the scientific programme envisaged. Present indications are that 12-15 instruments could provide a balanced inventory within the scope of the project. Fourteen basic types of instrument are illustrated in Table 3.1, and these could cover almost all the scientific developments proposed. The precise definition of the instruments to be built and their priority will be determined after an up to date reassessment of the scientific programme and in the light of experience gained on the new Harwell linac. During the planning stage instruments of advanced conception to match novel applications are also likely to emerge.

TABLE 3.1

INSTRUMENTS FOR THE SNS : AMBIENT TEMPERATURE MODERATOR

Instrument	Specification	Field
I. Single Crystal Diffractometer	$0.3 < \lambda < 1.5$ Å $\frac{\Delta Q}{Q} \sim 0.03$ Polarized incident beam option	Single crystal structure determination. Magnetisation density distributions. Structural investigations as a function of temperature, pressure, etc..
II. (a) High Intensity, Medium Resolution Powder Diffractometer	$Q > 2.5$ Å^{-1} ($\phi = 170^\circ$) $\frac{\Delta Q}{Q} \sim 3\ 10^{-3}$ $Q > 0.6$ Å^{-1} ($\phi = 30^\circ$)	Structural determination using powder samples. Kinetic processes, eg diffusion of gases into solids.
(b) High Pressure Spectrometer	$0.3 < Q < 100$ Å^{-1} Pressures up to 50 kbar	Pressure dependence of structure factors of fluids and amorphous solids. Triplet correlation functions.
III.(a) Elastic Discrimination Spectrometer	$1 < Q < 25$ Å^{-1} $\frac{\Delta Q}{Q} \sim 0.02$	Simultaneous measurement of elastic and total diffraction patterns to allow separation of elastic and inelastic scattering to study the eccentricity of atomic thermal vibration tensors. Structure factors of fluids and amorphous solids.
(b) Total Scattering Spectrometer	$0.3 < Q < 100$ Å^{-1}	
IV. Very High Energy Transfer Spectrometer	$\hbar\omega \sim 1$ eV $\frac{\Delta\hbar\omega}{\hbar\omega} \sim 0.1$ $Q < 4$ Å^{-1}	Electronic excitations: Band energies in semiconductors. Valence fluctuations. Crystal field levels in optically opaque transition metal compounds.
V. (a) High Energy Transfer Chopper Spectrometer	$100 < \hbar\omega < 600$ meV $\Delta\hbar\omega \sim 5$ meV $\Delta Q \sim 0.1$ Å^{-1} low Q bank	Vibrational spectroscopy, in particular the study of hydrogen modes on surfaces, in H-bonded systems and in metal hydride complexes. Phonons, magnons, crystal fields, vibrational modes and liquid dynamics. Measurements on the dynamics of the helium liquids. High Q dependence of $S(Q,\omega)$ for amorphous materials.
(b) High Energy, High Momentum Transfer Spectrometer	$Q_{max} \sim 30$ Å^{-1} ($\phi = 150^\circ$)	

VI. (a) Total Scattering Polarization Analysis Spectrometer	Q up to 20 $\AA^{-1}$, with spin analysis of scattered beam	Determination of elastic spin-dependent cross-sections. Structure factor measurements in presence of spin-incoherent scattering.
(b) Inelastic Polarization Analysis Spectrometer	$0 < \hbar\omega < 200$ meV $\frac{\Delta\hbar\omega}{\hbar\omega} \sim 0.01$ Spin selection of both incident and scattered beams	Spin dynamics. Separation of magnon and phonon scattering. Measurements of the dynamical structure factors $S(\underline{Q},\omega)$ and $S_s(\underline{Q},\omega)$ for spin-incoherent scatterers.
VII. Moderate Energy Transfer Chopper Spectrometer	$\hbar\omega < 100$ meV $\frac{\Delta\hbar\omega}{\hbar\omega} \sim 0.02$ low Q bank required $\Delta Q \sim 0.1$ $\AA^{-1}$	Measurements on the dynamical structure factors of liquids. Conventional inelastic spectroscopy. Very low angle bank required for observing inelastic modes in liquids on the energy gain side - the 'small Q method'. Phonon, magnon and crystal field studies.

INSTRUMENTS FOR THE SNS : COLD MODERATOR

Instrument	Specification	Field
VIII. High Resolution Powder Diffractometer	$Q > 6$ $\AA^{-1}$ ($\phi = 170^\circ$) $Q > 0.8$ $\AA^{-1}$ ($\phi = 15^\circ$) $\frac{\Delta Q}{Q} = 0.001$	Resolution of closely spaced peaks in powder diffraction profiles of complex crystals with large unit cells.
IX. (a) Very Low Q Spectrometer (SANS)	$0.005 < Q < 1$ $\AA^{-1}$ $\frac{\Delta Q}{Q} \sim 0.1$ $4 < \lambda < 12$ $\AA$	'Small Angle Neutron Scattering': studies of polymers and biological systems. Defects and structural periodicities on the scale ~ 100 $\AA$.
(b) Elastic Diffuse Spectrometer	Polarised incident beam option	Effects of doping, alloying, heat treating and irradiating condensed systems. Magnetic defects.
X. Total Scattering Spectrometer	$0.3 < Q < 100$ $\AA^{-1}$	Structure factors of fluids and amorphous solids. High resolution at low Q.
XI. Constant Q Spectrometer	$10 < \hbar\omega < 200$ meV $\frac{\Delta\hbar\omega}{\hbar\omega} \sim 0.05$ $3 < Q < 6$ $\AA^{-1}$	Triple axis analogue: scans in energy through $(\underline{Q},\omega)$ space at constant $\underline{Q}$. Coherent excitations.
XII. Time-of-flight MARX Spectrometer	$0 < \hbar\omega < 50$ meV $\frac{\Delta\hbar\omega}{\hbar\omega} \sim 0.02$ $\frac{\Delta Q}{Q} \sim 0.1$	Conventional inelastic scattering. Quasi-elastic scattering at good resolution and high Q, diffusive modes of plastic and liquid crystals, hydrogen in metals, etc.. Intermolecular modes of crystals, magnetic crystal field levels.
XIII. Long Wavelength Chopper Spectrometer	$4 < \lambda < 10$ $\AA$ $\Delta\hbar\omega/E$ comparable to that of IN5	High resolution quasi-elastic studies. Low energy inelastic modes (< 20 meV) observable in neutron energy gain.
XIV. White Beam Backscattering Spectrometer (L = 25 m)	$\Delta\lambda = 3$ $\AA$ Quasi-elastic mode: $\Delta\hbar\omega \sim 15$ μeV $-1000 < \hbar\omega < 1000$ μeV Inelastic mode: scan up to 100 meV with $\frac{\Delta\hbar\omega}{E} < 8\ 10^{-3}$	Very high resolution quasi-elastic studies. Slow diffusive processes. Tunnelling transitions. Inelastic band shapes.

OTHER USES OF THE FACILITY

3.88 The SNS has been designed as a high intensity pulsed neutron source for thermal neutron scattering experiments in condensed matter research. However the 800 MeV proton synchrotron, a machine comparable with the best pion factories of the world, would also be a source for the production of intense fluxes of other elementary particles. The provision of beams of these particles, primarily pions and muons, could find use over a wide range of multi-disciplinary studies which could include:

- studies in solid state physics and chemistry using muon spin rotation techniques
- chemical analysis and structure using μ-mesic atomic x-rays
- elementary particle physics
- nuclear structure studies
- biomedical studies and pion therapy
- isotope production
- radiation damage studies

Further information on these possible applications is given in Appendix I, where consideration is also given to the provision of a storage ring facility which, by creating smooth spill conditions, would considerably enhance the scope of many of these investigations. These possibilities are not part of the present Proposal.

CHAPTER 4. NEUTRON SOURCES

INTRODUCTION

4.1 We now return in this chapter to the subject of neutron sources and review the considerations which have led us to conclude that provision of the spallation neutron source at the Rutherford Laboratory is the best way to realise a significant flux increase. An outline of the proposed facility itself is given in Chapter 5.

4.2 Most neutron scattering applications use neutron beams from steady state nuclear reactors. Starting with the first experiments at Oak Ridge in 1946 research instruments have been installed on many reactors built for other applications such as materials testing, including the UKAEA reactors, DIDO and PLUTO at AERE, Harwell. The most powerful reactors available for condensed matter research today are listed in Table 4.1.

Table 4.1 High Flux Reactors

Reactor	Location	Power MW	Thermal Flux $n\ cm^{-2}\ s^{-1}$	First Operation
HFR	ILL-Grenoble	57	1.5×10^{15}	1972
HFIR	Oak Ridge	100	1.5×10^{15}	1967
HFBR	Brookhaven	40	0.7×10^{15}	1965

4.3 The HFR and HFBR were designed primarily as beam reactors for condensed matter studies; HFIR was designed mainly for isotope production. The standard arrangement allows neutrons from the high flux region of the moderator volume to pass to the experiment by beam tubes piercing the reactor vessel and radiation shielding. The neutron spectrum (white beam) corresponds to the Maxwellian velocity distribution at the moderator temperature and can be enhanced in the low and high velocity regions by the provision of special cold and hot sources.

4.4 The instruments on steady state sources commonly require monochromatic neutrons at the sample under investigation. They are selected from the white beam by monochromating crystals or by mechanical choppers; in either case the selection process must discard nearly all of the incident neutron flux.

4.5 Many neutron scattering experiments can be done in principle using an intrinsically pulsed white beam and the time-of-flight technique, instead of a monochromated continuous beam. Roughly speaking it is then the maximum neutron flux in the pulse which is the relevant criterion rather than the time-averaged flux. An important advantage of the pulsed source when it can be used in this way is the greatly reduced mean power to achieve the same effective flux as a steady state reactor.

4.6 Practical pulsed sources can be either pulsed reactors or accelerator based systems in which the burst of fast (MeV) primary neutrons has first to be moderated in, for example, a block of polyethylene whose surface(s) form the effective source for the scattering experiment. Ideally the pulse lengths of the moderated neutrons should not exceed a few tens of microseconds so that the required energy resolution can readily be obtained.

4.7 Relevant experience with pulsed reactors is limited to the Soviet Union where a small (30 kW) system (IBR-30) has been working since 1969; a much larger system (4 MW, IBR-2) is now under construction there. In the UK considerable experience has been obtained using the condensed matter cell on the AERE Harwell linear electron accelerator (linac) and there is similar experience in North America and Japan; the basic fast neutron production process involved is the $(e\gamma)$, (γn) reaction in a heavy target of, for example, tungsten or uranium. Proton accelerators can produce fast

neutrons by nuclear reactions in heavy elements (spallation reactions) and although not yet in use as 'production' sources, proton synchrotrons of energy around 1 GeV are now generally regarded as the best candidates for the next stage of source development.

POSSIBLE FUTURE SOURCES

4.8 Since 1973 the NBRC has kept the 'next generation source' under review with the NBRU actively considering ways to achieve an effective improvement in flux of at least 10 in comparison with the best available today. In terms of a steady state source this means at least 10^{16} n cm^{-2} s^{-1}; for a pulsed source the criterion is not so easily described and its merits must be examined by careful evaluation of 'typical' experiments, taking account of the neutron output characteristics of the source in question.

4.9 Three candidates have been carefully considered, namely:

- steady state reactors
- pulsed reactors
- accelerator systems, with and without neutron boosters

Fusion neutron sources, employing either magnetically or inertially (laser driven) confined plasmas, are much too far from practical realisation to merit serious consideration today.

Reactors

4.10 Reactor studies at Oak Ridge have suggested that pressing solid fuel technology to the limit might yield a gain of 5 in steady state performance. A flowing liquid fuel system, studied at Los Alamos, is in principle capable of attaining at least 10^{16} n cm^{-2} s^{-1}. However, such systems would require considerable development programmes to establish their design parameters and to estimate costs but the capital cost would be at least £100M and operating costs would be proportionately higher than present day levels, probably greater than £10M per annum. Problems in materials technology, radiation damage and safety would be severe and would inhibit the attainment of high operational efficiency.

4.11 The IBR-2 pulsed reactor under construction at Dubna (completion ∿ 1977) is designed to produce a peak thermal flux of at least 10^{16} n $cm^{-2}s^{-1}$ at peak power of 7700 MW when operating at 5 pulses per second with a power pulse halfwidth of 90 μs. It is unlikely that a pulsed reactor like IBR-2 would be much cheaper to build than an advanced steady state reactor of comparable performance, and although there should soon be useful operating experience in the USSR when IBR-2 is working, it is doubtful if the new technology could be readily transferred to Western Europe, implying the need for an independent development programme before such a project could begin. Also, the safety aspects would constitute a formidable obstacle to obtaining a nuclear site licence for such a research installation in an SRC laboratory. Finally, the inherently long power pulse is a distinct disadvantage.

4.12 Thus reactors, either steady state or pulsed, do not provide an acceptable solution. They would be far too costly, the technology is not adequately established, and in any case, they could barely offer the flux gains we are seeking.

Accelerator based systems

4.13 Particle accelerators are well established as neutron sources for many applications and continue to be considered for new requirements. Processes and parameters relevant to our interest are given in Table 4.2.

Table 4.2 Neutron production processes

Process	neutron/incident particle	target energy/neutron
$(e^-\gamma)$, (γn) in heavy element	∿ 10^{-2}/electron (30 MeV)	∿ 1500 MeV
(dn) in tritium	∿ 10^{-4}/deuteron (400 keV)	∿ 3000 MeV
(dn) in lithium	∿ 3×10^{-2}/deuteron (30 MeV)	∿ 1000 MeV
proton spallation in heavy element	∿ 30/proton (800 MeV, ^{238}U target)	∿ 55 MeV
(fission)	(∿ 1/fission)	(∿ 200 MeV/fission)

4.14 Amongst the accelerator options, proton spallation is clearly the most prolific and most efficient in terms of target power although to draw overall conclusions it is necessary to examine the details of the various systems

and to compare their costs. For instance, on the basis of a Canadian proposal (the unadopted ING project) a continuously rated proton spallation source producing a thermal neutron flux of 10^{16} n cm^{-2} s^{-1} would cost more than £100M.

4.15 For pulsed neutron sources however accelerators command special attention, for the following reasons. High intensity fast neutron bursts can be generated without excessive target heating; pulsing is straightforward (sometimes inherent); the pulse length and repetition rate can be varied within limits; the pulse length of the fast neutron burst is not significant in determining the moderated neutron pulse lengths; high performance can be realised using state of the art technology; operating costs are much less than those of reactors and there are no special problems of nuclear safety.

4.16 Since 1974 therefore, the Rutherford Laboratory has concentrated study on the use of electron linacs and proton synchrotrons for future neutron sources including the possibility of using a fission fuelled booster to increase the fast neutron production from the accelerator target.

PULSED SOURCES USING ELECTRON AND PROTON ACCELERATORS

4.17 Table 4.3 compares various pulsed sources, present or future, that can be realistically considered at this time. Neutron performances are quoted as the time average of fast neutrons produced, a valid criterion for comparison since in all cases the primary neutron burst length is much less than that of the moderated pulse.

4.18 The present Harwell electron linac (Table 2.3, line 1) has a well established condensed matter facility which is exploited in the SRC/AERE joint programme and has demonstrated the advantages conferred by the special features of such pulsed sources. Even at this very modest level of performance this facility is competitive with a high flux reactor for some limited applications. The new Harwell linac with 10 times the beam power will extend this range of usefulness.

4.19 The performance of an electron linac system must ultimately be determined by target dissipation capability. This is likely to be limited to

Table 4.3 Neutron sources based on electron and proton accelerators

Facility	Beam intensity or average target power; proton/pulse or kW	Pulse rate s^{-1}	Particle, energy	Target	Time averaged intensity at target n s^{-1}	Comments
1.Present Harwell electron linac	5 kW	200	e^-, 30 MeV	^{238}U	2×10^{13}	Closing December 1976
2.New Harwell electron linac	45 kW	150	e^-, 60 MeV	^{238}U	2×10^{14}	Under construction. Operational in 1978
3.Oak Ridge electron linac	65 kW	500	e^-, 140 MeV	Ta	1×10^{14}	In operation. Booster study 1975
4.Purpose built e^- linac	450 kW	150	e^-, 450 MeV	^{238}U	2×10^{15}	Estimated capital cost ~ £12M including instruments
5.Los Alamos weapons neutron research facility	1×10^{12}	120	p, 800 MeV	W	1×10^{15}	Uses protons from Los Alamos Meson Facility, LAMPF
6.KENS	6×10^{11}	15	p, 500 MeV	^{238}U	2×10^{14}	Japanese project to use injector synchrotron of KEK proton HEP facility
7.Optimised proton synchrotron + spallation target	2.5×10^{13}	50	p, 800 MeV	^{238}U	4×10^{16}	Estimated capital cost of accelerator + instruments on green field site - approx. £30M (Argonne IPNS estimated at $70M) Estimated cost if built at Rutherford Laboratory (SNS) using existing Nimrod plant and buildings and NINA power supply, £10M

the region of 450 kW indicating a maximum time average production rate of $\sim 2 \times 10^{15}$ n s^{-1} from a uranium target. Such an installation with buildings and instrumentation would cost about £12M (Table 4.3, line 4).

4.20 A static booster (operating below criticality at constant reactivity) using uranium fuel can readily provide a gain of 10; such a booster has been in use at AERE for many years, although not on the condensed matter cell. Plutonium boosters could have some advantages including that of a higher gain factor. It may be noted that a plutonium static booster added to the new Harwell linac would raise its output to a level of about 4×10^{15} n s^{-1} for an additional cost including instruments of about £6M.

4.21 Gains of several hundred would be possible with dynamic boosters, in which the reactivity is varied so as to exceed criticality for delayed neutrons, but not for prompt neutrons, with the reactivity peak occurring in phase with the primary neutron burst; the device is very similar to a repetitively pulsed reactor. A superbooster of this type was considered by the UKAEA (in consultation with University representatives) in 1965 but rejected because insufficient was known of the technology and on broad scientific grounds. Outside the Soviet Union dynamic booster technology is still not established and the safety implications remain to be examined fully.

4.22 The much smaller target energy generated by protons means that a target heat production rate similar to that considered as a limit for the electron linac ($\sim$ 400 kW) will allow fast neutron yields more than 10 times larger. This advantage has stimulated a number of proposals. In addition to the present UK proposal (Table 4.3, line 7), there is one from the Argonne National Laboratory for a closely similar design but on a green field site (IPNS, costed at $70M), and a more modest proposal in Japan (Table 4.3, line 6).

4.23 Design data for IPNS have been based partly on studies using an existing injector synchrotron at Argonne and the project has been the subject of two study workshops (1973 and 1975) involving potential users in the United States and invited visitors from Europe and Japan. The Argonne proposals have been widely supported and approval to proceed is being sought.

4.24 The choice of 800 MeV primary proton energy in both the IPNS and the SNS designs arises from the optimisation of neutron intensity at the moderator surfaces and from cost considerations. Neutron yield increases rather linearly with incident proton energy, above about 200 MeV, and increases with increasing target mass number. In target nuclei having a low fission threshold energy such as ^{238}U, a significant contribution comes from fissions. Thus protons of several hundred MeV and heavy-element targets are desirable. Proton energies greater than about 1 GeV are not advantageous because of their longer range and consequent extended neutron source distribution. About 90% of the neutrons are from an evaporation spectrum (similar to that of fission neutrons) of average energy 3 MeV, while the remaining 10% result from the nuclear cascade in a distribution extending upward to the incident proton energy. These high energy neutrons make necessary a heavy shield, as much as 5 m of steel.

4.25 The SNS will be about 100 times more powerful than the new Harwell linac and over 10 times that of the best purpose built electron linac. In cost per neutron it is very considerably cheaper than any alternative and at an estimated cost of £10M for the new components, including instruments, is strikingly economic.

4.26 Although provision of the SNS is clearly the best single step to take now to achieve higher neutron fluxes, the eventual addition of a neutron booster should be recognised as offering important development potential for the longer term.

CHAPTER 5. OUTLINE OF SPALLATION NEUTRON SOURCE PROJECT

DESCRIPTION OF THE FACILITY

5.1 The main features of the proposed facility will be a high repetition rate, high intensity, 800 MeV proton synchrotron, using as injector a 70 MeV H^- RF linear accelerator, and delivering the high energy protons to a uranium spallation target located within a substantial shielding blockhouse round which the neutron scattering experiments are arranged. Table 4.1 lists the main parameters; Fig. 5.1 shows a schematic layout. The installations are described in detail in Appendix II.

Table 5.1 SNS main parameters

Mean radius of synchrotron	26.0 m
Long straight length	9.19 m
Injection energy	70 MeV
Maximum energy	800 MeV
Injection scheme	H^-, charge exchange
Repetition rate	53 Hz
Frequency range of RF system	0.67-1.54 MHz
Harmonic number of RF system	1
Design intensity	2.5×10^{13} protons per pulse
Duration of extracted proton pulse	0.22 μs
Neutron production, time average, uranium target	4×10^{16} n s^{-1}
Target power	350 kW

5.2 The machine parameters have been chosen so as to optimise the neutron performance while pressing accelerator technology close to reasonable practical limits. An output energy of 800 MeV is about optimum for neutron

production intensity from the moderators, and a magnet ring for this energy with suitable spaces in which to fit the necessary injection and ejection devices and the RF accelerating cavities, fits conveniently inside the existing Nimrod magnet building. The synchrotron ring will completely replace the existing Nimrod magnet ring.

5.3 A repetition rate near 50 Hz is about the maximum for an economic design of the laminated synchrotron magnets, their power supply and the RF accelerating system; it also enables the existing NINA power supply to be used. Combined with the space charge limited intensity of 2.5×10^{13} protons per pulse, such a repetition rate yields a high time averaged neutron produciton rate and it is compatible with the pulse interval needed to avoid frame overlap in the experiments.

5.4 The injector will be the recently commissioned 70 MeV proton linear accelerator; its pre-injector will need modifying to supply the required current of H^- ions and the modulators supplying the RF circuits will have to be uprated from 1 Hz to 53 Hz operation. H^- injection into the synchrotron using charge exchange stripping will ensure that the accumulated beam before acceleration is limited only by space charge effects.

5.5 To obtain an even higher proton intensity per burst would require a higher injection energy and/or a larger magnet aperture with further consequences for many other parts of the system such as RF cavities and extraction magents. There are strong technical and financial deterrents in this direction.

5.6 Single turn extraction is used to give a proton burst of 0.22 μs duration, so short that the corresponding primary neutron burst is not significant in determining the moderated neutron pulse lengths. Standard beam transport techniques are used to direct the proton beam on to the target in the neutron cell. Fig. 5.1 shows the beam horizontal at the target. A vertical proton beam may facilitate geometrical optimisation of the neutron beams and in principle it could be provided but with some fairly strong cost penalties which need further examination.

5.7 It is proposed to use a target of ^{238}U, which, through the contribution of fission neutrons, has about double the neutron yield of non-

fissionable heavy metals. Target dissipation will be 350 kW. The target itself will be 30cm long and 8x8cm^2 in cross-section and can be conveniently water cooled by arranging it in the form of plates, clad in Zircaloy-2 to provide a corrosion and contamination barrier. Dimensional changes in the uranium plates caused by irradiation effects will limit the lifetime of individual target assemblies. The minimum lifetime is expected to be about 10 weeks and by careful attention to the design, target life might be extended to 25 weeks. In any case the target must be regarded as a consumable item.

5.8 Adjacent to the target will be moderator/reflector assemblies operating at ambient and below ambient temperatures, and arranged geometrically with respect to the target and beam tubes in the shield so that fast neutrons from the target cannot reach a sample directly. A possible arrangement provides for 2 moderators serving 12 beam tubes. This will accommodate the optimised selection of instruments given in Chapter 3, Table 3.1. If required guide tubes up to 75 m long can be arranged conveniently on ground at the level of Hall 3; longer ones could be provided by excavating some of the nearby ground. Optimisation of target, moderator, reflector, shield and experiment geometries is an important activity which will be carried out during the design phase and initial period of machine construction.

5.9 It is proposed to construct only one target station initially, in Hall 3, but a second could be added later in Hall 1, or Hall 1 could be used for the applications other than neutron scattering mentioned in Chapter 3 and Appendix I.

5.10 The SNS beam current is significantly higher than that so far achieved in any cyclic accelerator. While this performance is realisable essentially within state of the art machine technology, attention will be needed to confine beam loss to chosen locations in the synchrotron ring and the target station so as to localise the major incidence of radiation effects. Active handling arrangements will be provided where necessary.

5.11 A sophisticated control system will be provided drawing extensively on experience in the development and operation of the CERN SPS. This will be an important feature for the safe and efficient operation of the facility.

NEUTRON PERFORMANCE PARAMETERS OF SNS

5.12 In order to make meaningful assessments of potential scientific applications of the spallation source, it is necessary to know the spectral and temporal characteristics of the neutron burst as well as the gross features such as mean intensity, repetition rate, moderator dimensions and experiment geometry. The information summarised below in paragraphs 5.13-5.19 is treated in more detail in Appendix I.

5.13 Fast neutrons may be moderated to useful energies (ie to energies around and below 1 eV) by interaction with a few centimetres of a light material of high scattering cross-section, in practice hydrogenous. The moderated spectrum has an approximately 1/E dependence in the neutron slowing-down region, corresponding to E greater than $\sim 5\ k_B T_{eff}$ (where k_B is Boltzmann's constant and T_{eff} is the effective temperature of the moderator); at lower energies the spectrum assumes the form of a Maxwellian distribution characteristic of the effective moderator temperature.

5.14 The neutron yield from the moderator surface for an optimized target/reflector/moderator configuration has been estimated from published experimental data to be

$$\phi(E) = \frac{10^{13}}{E} \text{ n eV}^{-1} \text{ ster}^{-1} \text{ s}^{-1}$$

in the slowing-down region of the spectrum. The time-averaged intensity at a specimen distance L from the moderator within an energy window ΔE at E is then given by

$$I(E)\ \Delta E = \frac{10^{13}}{L^2} \quad \frac{1}{E} \ \Delta E \quad \text{n cm}^{-2} \text{ s}^{-1}$$

5.15 In the slowing-down region the pulse shape is relatively narrow and the full width at half maximum may be approximated by the empirical expression

$$\Delta t(\mu s) = 2/\sqrt{E(eV)}$$

5.16 The slowing-down region of the spectrum may be extended by reducing the moderator temperature, which moves the Maxwellian portion of the spectrum to lower energies and maintains the short slowing-down pulse width. For example:

Moderator	Temperature	kT_{eff}	1/E Range
$(CH_2)_n$, H_2O	300K	∿ 35 meV	> 200 meV
CH_4, NH_3	77K	∿ 11 meV	> 50 meV
CH_4, H_2	20K	∿ 3 meV	> 15 meV

Thus Δt is 2 μs for 1 eV neutrons from an ambient moderator, and only 13 μs at 25 meV if a 20K moderator is used.

5.17 In the Maxwellian region (E less than ∿ 2 $k_B T_{eff}$) the pulse width broadens considerably, and the pulse shape is asymmetric and cannot be expressed in a simple analytic form. The width depends on the neutron energy, the moderator material, temperature and configuration, and may be broadened by some moderation within reflectors.

5.18 Estimates of beam intensities from slab moderators (polyethylene and methane) at different temperatures are illustrated in Figure 5.2. The spectra have been normalised to unity at 1 eV, an energy which is in the slowing-down region; F(E) is the ratio of the flux per unit energy at an energy E to that at 1 eV. The time averaged neutron intensity at a specimen at a distance L from the moderator within an energy window ΔE at an energy E is thus

$$I(E)\ \Delta E = \frac{10^{13}}{L^2}\ F(E)\ \Delta E \quad n\ cm^{-2}\ s^{-1}$$

5.19 The above estimates have been based on a diversity of published experimental data, and more experimental work and computations will be needed to define closely the characteristics of SNS, particularly for energies in the Maxwellian region for any moderator.

COSTS

5.20 Striking economies are possible in the overall cost of the proposed facility because of the extent to which use can be made of existing plant and buildings; notably, the new Nimrod injector, Nimrod beam handling magnets, quadrupoles, vacuum and control equipment, the extensive service network, steel and concrete shielding, and also the NINA magnet power supply. No new buildings will be required. The present day total value of all the plant and buildings which will be used in whole or part is estimated at £20M. The use of existing buildings confers great flexibility on the construction programme in addition to substantial cost savings.

5.21 The new money capital cost of the accelerator system and target station including moderators and active handling services is estimated at £7.6M. A further £2.6M should cover the cost of 15 instruments at $\sim$ £150K each plus ancillaries such as cryostats. Estimated capital costs are itemised in Table 5.2.

5.22 The exact cost of instruments and their ancillaries will depend on the inventory chosen. Instruments are likely to be selected for this over a period of several years with the last ones possibly being decided even after the facility is in use. Also, as is normal in planning the exploitation of a facility like SNS, a somewhat arbitrary line has to be drawn between the initial and ongoing provision of instruments.

5.23 Assuming a 5 year programme covering detailed design and machine construction the total recurrent expenditure (excluding staff costs and overheads) over the same period is estimated at £1.5M and the total effort at 570 man years.

5.24 For the equilibrium operating phase corresponding to use of a full complement of instruments on one target station, annual recurrent costs (excluding staff costs and overheads) are estimated at £1.1M, capital expenditure at £0.3M per annum and the direct effort at 135 man years per annum. These resources would allow 24 hour operation for 80% of the calendar year (7000 hours). The total corresponding annual cost including staff costs and overheads, would be approximately £2.75M per annum. Details of these estimates are given in Appendix II, Table 5.5.

Table 5.2 SNS estimated capital costs. January 1976 prices

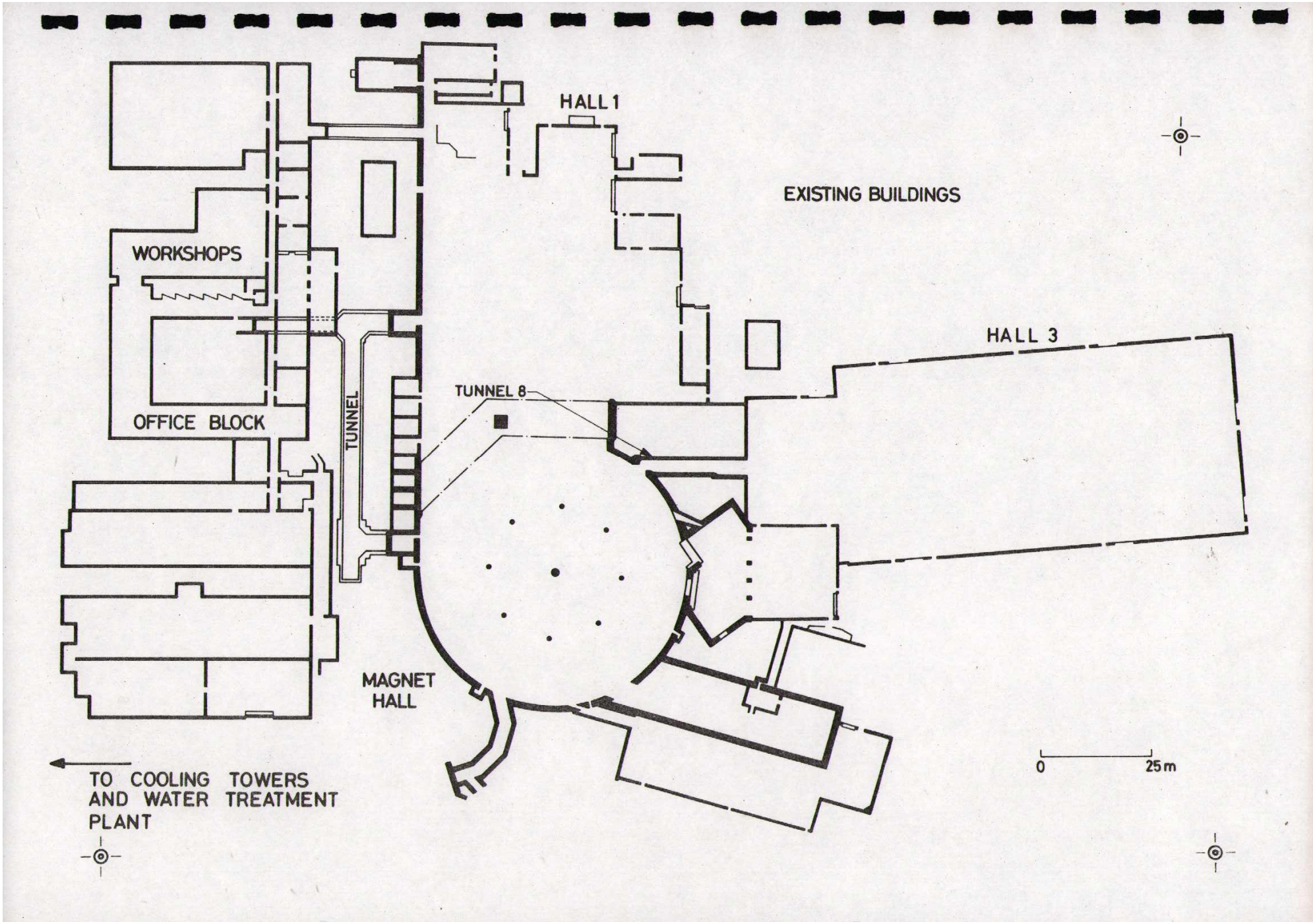
	£K	£K
Machine		
Modifications to 70 MeV injector	415	
Injection system	125	
Magnet system	2600	
Vacuum system	570	
Radiofrequency system	895	
Extraction system	500	
Controls	445	
Services	80	
Dismantling of Nimrod	100	
	5730	
VAT	458	6,188
Target station		
Target	160	
Moderators	250	
Shield	600	
Special services	300	
	1310	
VAT	105	1,415
Experimental facilities		
Neutron scattering instruments (15)	2250	
Instrument ancillaries	200	
	2450	
VAT	196	2,646
TOTAL		10,249

OVERALL PROGRAMME

5.25 Table 5.3 shows an optimised programme model which, starting from 1977, would allow the first neutron scattering experiments to begin towards the end of 1982. Instrument construction starts later than that of the machine and continues into the first years of operation with some ongoing provision of instruments beyond. The start of construction is phased to match an assumed Nimrod closure date of December 1978. On this programme the peak in expenditure, approximately £5M, would fall in the fifth year. The programme might be extended somewhat if that were necessary to match the available funding rate, but this would be less economic overall apart from the delay in starting the new science. A programme for the first experiments to begin early in 1984 would have its peak expenditure at approximately £4M in the sixth year.

Table 5.3 SNS programme model. Costs in £M, January 1976 prices

Year	1 77/78	2 78/79	3 79/80	4 80/81	5 81/82	6 82/83	7 83/84	8 84/85	9 85/86	10 86/87	11 87/88
Machine		↑ Nimrod closure									
Design		├	─	─	─	─					
Manufacture and installation		├	─	─	─	─					
Commissioning				├	─	─	─				
Target station											
Design		├	─	─	─	↕	800 MeV				
Manufacture and installation		├	─	─	─	─	beam				
Commissioning				├	─	─					
Experimental facilities											
Design		├	─	─	─	─	─	─	─	─	─
Manufacture and installation				├	─	─	─	─	─	─	─
Commissioning						├	─	─	─	─	─
						↑ neutron scattering experiments begin					
COSTS											
Design and construction											
Recurrent	.25	.53	.23	.17	.16	.05					
Capital			.62	2.32	3.47	2.70	.9	.24			
Operation											
Recurrent						.46	.87	.93	1.05	1.1	1.1
Capital									.15	.3	.3
Staff costs and overheads	.44	.95	1.25	1.30	1.40	1.35	1.35	1.35	1.35	1.35	1.35
TOTAL	.69	1.48	2.10	3.79	5.03	4.56	3.12	2.52	2.55	2.75	2.75
Direct effort, MY	44	95	125	130	140	135	135	135	135	135	135


HALL 1
EXISTING BUILDINGS
WORKSHOPS
HALL 3
TUNNEL 8
TUNNEL
OFFICE BLOCK
MAGNET HALL
0
25m
TO COOLING TOWERS AND WATER TREATMENT PLANT

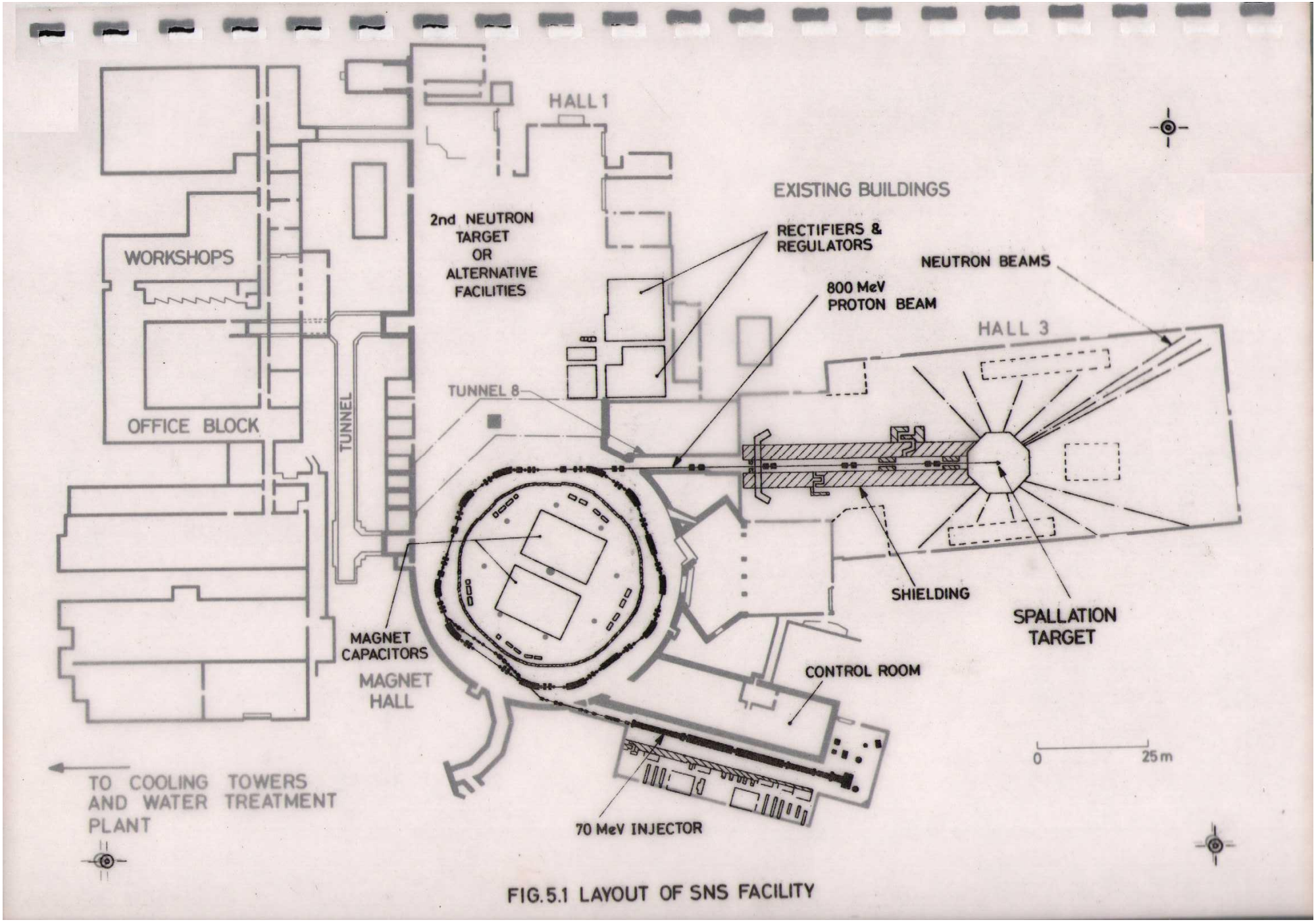


FIG. 5.1 LAYOUT OF SNS FACILITY

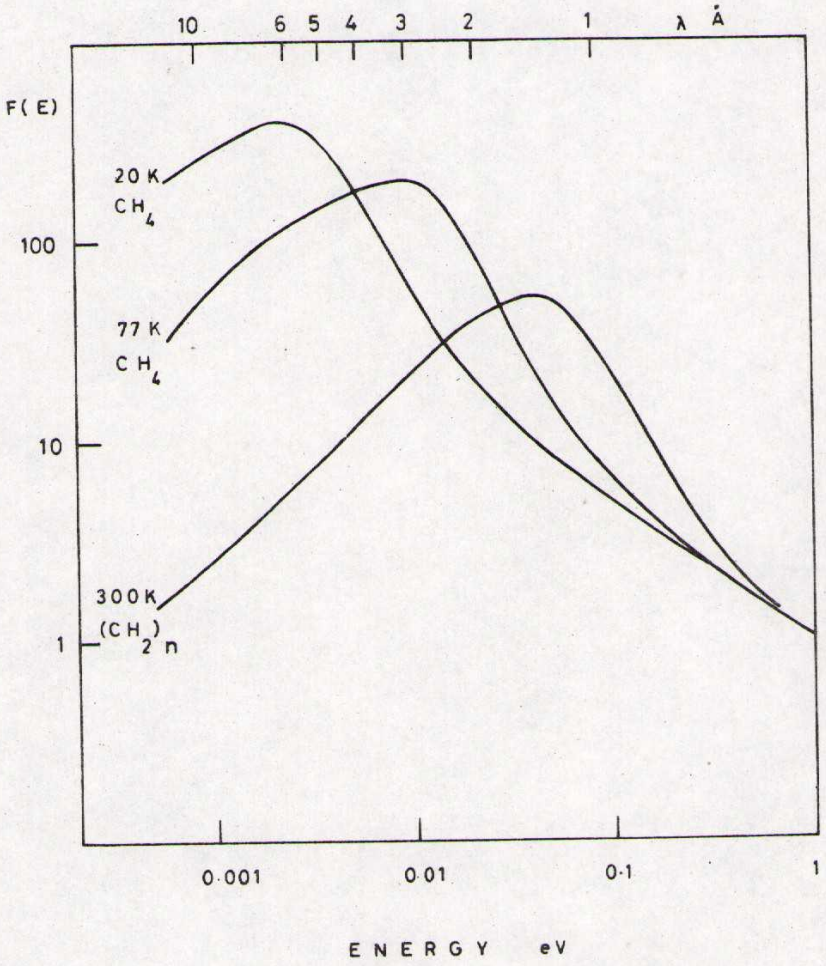


FIGURE 5.2 The spectral distribution of neutrons from various moderators.