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The Scientific Case for the High Flux Beam Reactor

February 1970

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THE SCIENTIFIC CASE FOR THE HIGH FLUX BEAM REACTOR

Foreword

This report has been drawn up as part of the submission of the H.F.B.R. Main Working Group to Council. It contains the scientific case for building the H.F.B.R. for use by university chemists and physicists and, to a smaller extent, by structure biochemists.

It has been contributed to by members of the Neutron Beam Research Committee and by members of the Working Party on Neutron Beam Instruments.

E.W.J. Mitchell.

February 1970.

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THE SCIENTIFIC CASE FOR A HIGH FLUX BEAM REACTOR

SUMMARY

a. Neutron scattering is playing an increasingly important part in the study of the solid and liquid states of matter. Although the early work was directed at problems of interest to physicists the use of the technique has spread rapidly during the last few years into chemistry and, in a preliminary way, into biology.

b. The importance of neutron experiments is their uniqueness of interpretation. This arises from the basic properties of the neutron: the way it is scattered by nuclei; its magnetic moment; its mass; and in a large number of cases its low absorption.

c. Chemists and physicists would not choose to use neutron scattering to solve structural problems if they could be solved using x-rays. However, using x-rays it is notoriously difficult to determine the positions of light atoms in structures also containing heavy ones. This is because the heavy atoms have more electrons and therefore give rise to more of the observed scattering. With neutrons the scattering is determined by nuclear forces and the scattering factors, while different for different nuclei, do not show the very large differences which occur in x-ray atomic scattering factors. This characteristic enables neutrons to be readily used for determining the positions of light atoms – especially hydrogen or deuterium – in crystal structures.

d. The existence of the neutron magnetic moment means that neutrons can interact with the magnetic states of matter. This enables magnetic structures to be determined – as in ferromagnetic, ferrimagnetic and antiferromagnetic crystals – and also through inelastic scattering, the dynamics of magnetization. Such studies are impossible with x-rays and neutron work has already given a new boost to our attempts properly to understand magnetic phenomena.

e. The magnitude of the mass of the neutron enables it easily to interact with the individual and collective motions of atoms in all forms of condensed matter. This is allowing the properties of lattice waves to be determined completely for the first time. Many of the exciting possibilities of extensions of this work to individual phonon lifetimes, to the dynamics of phase transitions through mode instability (particularly in the case of ferroelectric transitions) are held back though lack of flux.

In chemistry the detailed information which can be obtained about atomic motions from neutron studies is more complete than can be obtained with any other techniques or combinations of techniques. This has already been demonstrated by the work – still in its infancy – on polymers and organic liquids. The further extensions into chemistry seem boundless.

Neutron techniques are playing a crucial role in the development of our understanding of liquids. Although at present tantalizingly limited by the available fluxes, it is possible with neutron studies to determine both the velocity and position co-relations of atoms, where previously with x-rays only position co-relations were possible. We expect that higher fluxes would make this another major growth point.

f. Finally the relatively low absorption in a number of materials means that experiments may be carried out using long wavelengths, much longer than possible with x-rays. This opens up a new technique for studying disordered systems – crystal defects, non-stoichiometric compounds, alloys and amorphous solids. Experiments in the 5–15Å region are yielding encouraging results and the possibilities of the extension of this work and of the wavelength range for the study of composite materials is being examined. Apart from this possibility studies of defect clusters and of the ordering of defective oxides are actively being pursued.

SUMMARY (Cont.)

g. At present the limitation of available fluxes is preventing the realization of the full impact of neutron studies to the new areas of study. Individual experiments are hindered or are not attempted because of lack of flux and this is true across the whole range of studies of condensed matter. In this connection the impetus given to the neutron study of ferroelectric transitions by the Brookhaven reactor is an indication of the general threshold state of the application of neutrons. The lack of flux also puts severe limitations on the throughput of experiments, a limitation which prevents a fuller exploitation of neutron techniques in chemistry where the ability to study many systems is important. Experiments using polarized neutrons are also hindered and the wide application of polarized neutron studies awaits higher fluxes.

h. The preceding summary shows that the case for a high flux thermal neutron source rests on:

(i) the crucial nature of the experiments which can be carried out using neutrons;

(ii) the number of experiments which are hindered or which cannot be attempted because of lack of flux;

(iii) the increasing scope of the areas of useful applications of neutrons into chemistry and biology which higher fluxes would make possible, both for individual experiments and the throughput of experiments.

i. A substantial improvement over present UK fluxes ($2 \times 10^{13} - 10^{14}$ n cm^{-2} sec^{-1}) is, therefore, required. Bearing in mind the Brookhaven reactor (7×10^{14}) and the Franco-German reactor of even higher flux under construction, we recommend that the U.K. source should be designed for at least 1.5×10^{15} n cm^{-2} sec^{-1}.

j. As to the nature of the source we confirm the previous conclusions and are convinced that the High Flux Beam Reactor is the only practicable solution. It gives an adequate number of beam holes for the full range of experiments for the 150 estimated number of users. It also provides for the simultaneous operation of all types of experiment at near optimum conditions.

k. Without the reactor, a field of science in which we have always been in the forefront, is bound to suffer. Conversely the provision of the reactor will give an incalculable boost to this field of the physics and chemistry of condensed matter. The number of users and the areas of science covered fully justifies the S.R.C. commissioning the H.F.B.R. for university users.

Chapter 1. RADIATION AND MATTER – A BRIEF SURVEY

1.1. The history of solid and liquid state physics shows a series of examples of how our understanding of condensed matter has been increased by the study of its interaction with each new kind of radiation. The optical work of the nineteenth century was followed by the use of x-rays, of electrons in electron diffraction and in electron microscopes, of radiofrequency sources in the study of dielectrics and in n.m.r. and of μ-waves in e.s.r. The application of each new radiation has opened up whole new areas of investigation. After a specialist period these new techniques become the essential tools for solid state work, and more generally the study of condensed matter.

1.2. Further progress has followed in two stages. First, there is the spread of the techniques to other sciences, particularly chemistry and biology. When this occurs it soon happens in a big way and for example infra-red and n.m.r. are being used more in chemistry than physics, and electron microscopy is being used more in biology than in physics.

1.3. The second further stage – and there is no sharp boundary in time between the stages – is the use of the techniques in technology. Primarily in the past this has occurred in the applications of chemistry and in metallurgy. We have in mind the use of all forms of spectroscopy, of x-ray and electron diffraction and of electron microscopy. Equally e.s.r. and n.m.r. have become as important to the development of the applications of chemistry as to chemistry itself. Since the war we should add to these technologies, the technologies based on solid state physics itself, although industrially they have not reached the unified scale of chemical technology and metallurgy.

1.4. Neutrons were discovered in 1932 and became available in intense fluxes after the war with the rapid development of nuclear reactors. Physicists were quick to realize that the interaction of this new type of radiation with matter could again lead to major advances in our understanding of solids. The first type of work to be done was neutron diffraction, precisely equivalent to x-ray diffraction, but taking advantage of some favourable nuclear parameters to determine atomic positions not capable of being studied with any accuracy by x-rays. The neutron also interacts through its magnetic moment with the magnetic moments which arise from electron spins in atoms. This gives rise to magnetic scattering and enables magnetic structures to be investigated for the first time. Indeed, it can be said that the scope of the problem of magnetic structure was only realized as a result of research work with neutron beams. Perhaps of greater interest now and in the future, however, is the use of neutrons for studying the dynamic and static imperfections in crystals. In the former case it turns out that thermal neutrons have just the appropriate energy/momentum relationship for studying the atomic vibrations in molecules and solids. In the study of static imperfections one utilizes the fact that the neutrons are scattered by the nuclei and are relatively unaffected by the distorted electronic structure near the imperfections. The favourable properties of neutrons also make it possible to study the atomic motions as well as positions in liquids. This is the step which is likely to be as significant to the study of liquids as was the Braggs' work to the study of solids.

1.5. It has been argued strongly in the past, in the N.I.R.N.S. report of 1963, and especially by Professor J.S. Anderson in the 1965 report, that neutron techniques would be of crucial significance not only in physics but to a wide variety of chemical problems. The work done during the last few years has amply borne out these predictions.

Thus neutron diffraction is already playing a substantial role in the elucidation of molecular structures of importance in chemistry and molecular biology because of its power in locating light atoms (principally ^{1}H) in the presence of heavy atoms. The place of neutron spectroscopy has also been established. In contrast to widely used methods of optical spectroscopy, not only can optically inactive molecular motions be observed but also the amplitudes of nuclear motions can be determined from intensity measurements and perhaps most important of all, the time scales of local motions over distances of ~10Å can be measured.

1.5. (cont.)

Thermodynamic and optical spectroscopic measurements have given the chemist a working knowledge of equilibrium properties of molecules and assemblies of molecules. Diffraction studies continue to provide information on the structures of molecules and ordered arrays of molecules. The understanding of transport phenomena, of the dynamics and energetics of flexible molecules, and of local order and molecular diffusion in amorphous states is at a more primitive level. It is here that neutron spectroscopy has an important role to play. Technologically these studies bear directly on areas which include electrochemistry, polymer technology, fluid mechanics and catalysis.

The current position of neutron techniques in chemistry is analogous to that pertaining in n.m.r. spectroscopy in 1955. Then the need was for higher magnetic fields of greater homogeneity and for more sensitivity, the promise of the technique having already been demonstrated. Achievement of these instrumental improvements within the next half decade brought n.m.r. spectroscopy to the fore as a major tool in chemistry. Higher neutron flux and better spectrometers and diffractometers similarly will expand the role of neutrons in chemistry to provide answers to problems of molecular structure and properties of materials where at present progress is denied. The importance of the field has been acknowledged in 1969 by the award of the Marlow Medal of the Faraday Society to John White for his pioneering efforts in chemistry.

1.6. The pattern of use of optical spectroscopy, x-ray diffraction, electron diffraction, radiofrequency waves and microwaves may be seen to be repeating itself with neutrons. The difference on this occasion is set by the source which produces the neutrons. We are not, this time, dealing with a compact piece of apparatus which can be set up and operated in any industrial or university laboratory. A large and expensive central facility is required.

Chapter 2 THE SCIENTIFIC CASE FOR A HIGH FLUX THERMAL NEUTRON SOURCE

2.1. The scientific case which we describe here is, in its essentials, the same as that fully described in 1965. However, the range of the applications which have grown since 1965, particularly in chemistry and to a lesser extent in biology, provides a background of experience which confirms what in 1965 were sound, but unproven, predictions. This experience should give extra confidence that the general views put in the past are correct.

In this chapter, therefore, we restate the scientific case and discuss the areas in physics and chemistry where significant advances in understanding can be expected if higher fluxes become available. In doing so we outline the current limits of application. In the following chaper (3) we pick out, by way of illustration, some of the advances which have been made over the last few years.

2.2. The characteristics which make neutrons so important for the study of condensed matter may be summarized as follows:

(a) The nuclear scattering cross-sections for neutrons, while varying, do not show the very large differences which occur for different atoms in x-ray scattering. This characteristic enables the position of light atoms – hydrogen is a particular case – to be located precisely in the presence of heavy atoms.

(b) The neutron has no electric charge but does possess a magnetic moment. These features make the neutron unique amongst available radiations in being able to react to the magnetic state of materials.

(c) The mass of the neutron is comparable with atomic masses so that it is possible to measure changes in both energy and momentum in the range significant for the dynamics of atom movement. Thermal neutron energies are about 1/40 eV so that changes of energy corresponding to the quanta of processes with a resonant frequency in the region of 10^{13} Hz are easily measured. (Electromagnetic radiation of this frequency has a wavelength of one three-hundredth of a centimetre, and so cannot be used for investigation of spatial detail on an atomic scale. Conversely electromagnetic radiation with wavelengths of atomic dimensions – x-rays – has such high energy ($\sim 10^4$ eV) that the energy levels of thermal interest cannot be studied).

These three characteristics (a.b.c.) mean that the neutron is able both to respond to atomic positions and magnetization on the atomic scale, and to exchange energy of thermal and epithermal magnitude with a scattering sample. Thus it is possible to make unambiguous deductions about many basic properties of those materials which we can investigate. Results of experiments using other techniques can be interpreted in terms of these basic properties, but it is exceptional for any other techniques to be able to isolate the individual effects so convincingly as can the appropriate neutron experiment. Perhaps it is worth noting that it is this certainty of interpretation, rather than any major extension into new fields of physics, which gives neutron beam experiments their paramount value: this is in sharp distinction to the situation of high energy physics.

2.3. The scientific programme may conveniently be divided under three main headings

(a) Crystallography
  (i) Non-magnetic
  (ii) Magnetic

2.3. (cont.)

(b) Inelastic scattering

Inelastic experiments may be divided into two energy ranges which, although somewhat arbitrary, turn out to be convenient for the later discussion in the report. These are low energies (less than, say 20 meV, or wavelengths longer than, say 2Å and extending to at least 15Å), and high energies (taken here for convenience as 20 – 500 meV). Each range includes experiments on:

(i) Phonons in crystals.
(ii) The dynamics of the fluctuations of magnetization in crystals.
(iii) The dynamics of atom movements in liquids.
(iv) Molecular rotations and vibrations.

(c) Defect scattering

(i) Structural defects and magnetic and non-magnetic substitutional atom defects; order-disorder studies.

2.4. In this section we consider the programme under the headings given above

(a) Neutron crystallography

(i) Non-magnetic

The positions of light atoms in structures and the differentiation of atoms of similar atomic number, may be explored with neutrons, thus removing an irritating set of restrictions on x-ray work which has held up studies of hydrogen bonding, hindered rotations in ions, hydride structures, heavy element oxides, carbides and nitrides, and studies of ordering in many interesting metallic systems. Single crystal work on comparatively simple non-hydrogenic substances with about thirty atoms per unit cell requires, at present fluxes, samples of linear dimensions of about 1 mm. Complex hydrogenic substances, such as the simplest crystals of biological interest, may require samples with dimensions of 1 cm. All polycrystal work requires bigger samples, which although easier to produce, yield results which can usually only be interpreted to a very limited extent.

The provision of a high flux source would widen the field of application enormously, would allow use of separated isotopes and would bring us at least to the borderline of a study of the role played by hydrogen in simple substances of biological interest. It is confidently predicted that higher fluxes, together with streamlined experimental procedures, will lead to a considerably wider application of neutron techniques to chemical problems.

(ii) Magnetization studies in crystals

Despite the fact that the close connection between strong magnetic properties and the anti-symmetry condition for electron wave functions was recognised by Heisenberg in 1926, we are still unable to predict the magnetic structures of matter. Even in technologically important materials, such as ferrites and garnets, we have only a skeleton empirical knowledge of the facts and this has been provided mostly by neutron techniques. The magnetic electrons are the outer electrons and are of interest for chemical as well as physical reasons.

At present, single crystals for this work must be comparatively large (approaching 1 mm^3) and may need to be single domains. This is a very serious limitation since large crystals are generally difficult to make. Because of this limitation work has concentrated mainly on metallic systems where large single crystals are readily available and has partially neglected oxides. An increase of neutron flux would enable smaller crystals to be used and make possible a much wider range of investigations.

2.4. (cont.)

(b) Inelastic scattering

(i) Phonons

Inelastic scattering of neutrons from crystals in which the neutrons experience a change in energy gives us a direct method of studying the vibrations of atoms in crystal lattices. The very first vibrational studies to be completed in detail (at Chalk River in Canada) showed that whilst some of the forces between atoms in a metal were best thought of as comparatively local, some are of very long range, and probably represent forces derived from the electron gas in some way. In ionic and covalent crystals the phonon frequencies showed some unpredicted effects, now regarded as evidence for polarization by deformation. A study of lead has shown anomalies which are due to the interaction between the electron Fermi surface and the phonons – the interaction which is the basis of the current explanation of superconductivity.

(ii) Spin waves

Inelastic scattering of neutrons also provides a means of studying directly fluctuations in the magnetization of crystals (spin waves). The same apparatus is used for spin wave studies in ferromagnets, antiferromagnets and ferrimagnets, and once again this is the only available method for work on bulk substances (there is a resonance technique available for thin films of ferromagnets). The explanation of the results is still uncertain, especially in metallic systems where inadequacies in the original formulation of the problem are now clearly exhibited for the first time.

Most of the studies in these two categories demand samples of about 20 cm^3 in volume and are, therefore, limited to compounds of elements with low neutron absorption and of which very large single crystals may be grown. With a high flux source and with large crystals such as these, some experiments on the life time of phonons would be possible, and a detailed picture of the basic processes determining thermal conductivity should result. These experiments are quite beyond our reach with currently available fluxes. We should like to emphasize the importance of studying low energy excitations, i.e. low energy phonons and magnons. Such studies require the provision of a copious supply of cold neutrons.

(iii) The dynamics of atom movements in liquids

Inelastic scattering from liquids allows us to study the dynamics of the atoms and molecules. For many years, the correlations of atomic positions in liquids have been known from x-ray studies. With inelastic scattering techniques it is possible to study correlations of the velocities of the atoms and already it has been shown that the effective mass for diffusion is about 100 atomic masses. It has also been shown that co-operative modes, analogous to phonons in crystals, occur at high wave numbers.

In the study of diffusion it is possible to examine the details of a single diffusion step. The diffusion step occurs on a time scale of about 10^{-12} sec., and to study that period requires scattering wave vectors of 0.5$Å^{-1}$ and energy transfers of ~0.5 meV. Transfers of this order have to be measured precisely and the scattered intensity determined at each point so that the shape of the curves may be compared with the predictions of diffusion theories. In this way one can decide, for example, whether the diffusion process consists of atoms jumping from site to site, as in a solid, or of combined movements of groups of atoms, analogous to Brownian motion, or, as actually occurs, the process is a combination of several mechanisms. The importance of this knowledge in the study of chemical reactions

has been realized in the last few years but the accuracy of individual experiments, and what is equally important to chemistry, the throughput of different compounds, is severely limited by present fluxes and facilities.

Advance in understanding liquid diffusion is directly related to the precision of the neutron measurements and very high resolution measurements on liquids will be one of the first experimental programmes with a higher available flux.

The discussion of the collective motion in liquids is a complicated matter since these modes are heavily damped due to their strong interactions with one another. Since it is just at this point that the theory of liquids is in difficulty the neutron measurements of this effect are eagerly awaited, but again they require accurate and time-consuming measurements. For example, at present the measurement to an accuracy of 0.5Å of the wavelength change occurring in the scattering of 8Å neutrons may take one week, whereas with a sufficiently higher flux source ($\sim 10^{15}$ n cm^{-2} sec^{-1}) the same measurement to an accuracy of 0.1Å would take about one day. These figures are for reasonably favourable processes. However, some of the most interesting processes give low scattered neutron intensities and have longer counting times – those occurring at the minima of the diffraction pattern for example. Most important amongst these are the study of collective modes having a wavelength of several atomic spacings, because modes of this kind determine the degree of long range order (or disorder) in the liquid.

Present experiments are yielding tantalizing results which will be far more useful when further factors of improvement in energy and momentum resolution are available. We forecast that the use of neutron techniques will be as important for furthering a knowledge of the structure and dynamics of liquids as the use of x-rays was for solids.

(iv) Molecular rotations and vibrations

We have discussed in (i) above the use of neutrons for the study of atomic vibrations in crystals. A major growth point has also been the application of neutron techniques to the study of molecular spectroscopy in gases, liquids and solids of interest to chemists. The studies include work on small molecules, on synthetic and natural macromolecules and on molecules absorbed on surfaces, and they complement infra-red and Raman spectroscopic techniques which are already widely used by chemists. The special features of the neutron technique include the removal of the restrictions imposed by optical selection rules and very different, and often simpler, intensity relationships.

The insatiable demand which has already occurred from chemists is mainly for the study of low frequency modes of vibration and other lower energy excitations using cold neutrons. Poor spectral resolution at present limits the quantitative information which can be deduced but nevertheless important results have been obtained and it is clear that much more information will be forthcoming when the spectral resolution is improved. In particular, experiments on the dynamics of molecular crystals and on polymer chains will be more fruitful and a much better understanding of intermolecular and intramolecular forces between non-bonded atoms will result. An equally exciting prospect is the extension of work begun by White on surface species present in heterogeneous catalysis. The new inelastic technique of polarization analysis promises to be very important for molecular neutron scattering.

To date relatively little work on higher energy excitations using thermal neutron scattering in Stokes rather than the anti-Stokes mode, has been carried out. The demand for this form of vibrational spectroscopy is growing and preliminary studies indicate a particularly bright future, provided present facilities can be replaced by a vastly improved installation.

Neither presently available fluxes nor the existing facilities for molecular spectroscopic studies by inelastic neutron scattering are adequate to provide the precision required or the throughput of samples.

(c) Defect and domain scattering

Long wavelength neutrons may be scattered from crystals only if deviations from the crystal perfection are present, so that the scattering of neutrons which originate in a cold moderator, or have been otherwise selected to have low energy, gives a powerful method of studying defects on a size scale of 2–10Å. This is just below the resolution of the electron microscope and there is considerable promise in this method of study of radiation damage.

Foreign atoms in magnetic substances may disturb the magnetic structure, so that we can study local modifications of the magnetic lattice. This has already thrown new light on the behaviour of nickel and iron based alloys and provides a searching test of the validity of electron band theory for alloys. Experiments are now under way at Harwell in collaboration with I.C.I., and Manchester University to measure domain sizes in composite polymeric materials by low angle neutron scattering. X-ray scattering has proved to be of very limited value and so success at Harwell would provide a major technique for investigating the morphology of these commercially important materials. Their structure/property relations are not understood.

Two rather different types of apparatus exist with long programmes of exciting work ahead of them. Very large samples of highly controlled perfection are demanded by these programmes and samples five to seven cm square and about one cm thick are required. Where such samples can be obtained, a higher flux would mean that essential information could be obtained on the wavelength dependence of the scattering. Where only smaller samples are available results of the present useful quality would be obtained for many more samples.

As in the case of neutron crystallography, the availability of high fluxes and the streamlining of techniques will lead to a much wider application of neutrons by chemists to the study of the defect solid state. The chemists are particularly concerned with defect arrangements in non-stoichiometric compounds where the short and long range ordering of both defects and ions are thought to be important. Encouraging progress has been made in this field.

2.5. Summary and conclusion

We have highlighted some of the scientific programme which could be carried out with a high flux source and have discussed some of the difficulties which limit the scope and achievement of work at the present time

We conclude that the scientific case for a high flux thermal source rests on four basic points:

(a) the crucial nature of the experiments which can be carried out with neutrons. It is not possible – even in principle – to obtain the desired information with other techniques or combinations of techniques;

2.5. (cont.)

(b) the number of such experiments which are hindered or which cannot be attempted because of lack of flux;

(c) the increasing scope of the areas of useful application of neutrons (e.g. advancing into chemistry and biology) which higher fluxes would make possible;

(d) the increased rate of throughput of samples which is critical to the use of spectroscopic and diffraction techniques in chemistry and biology, when a larger number of relative measurements rather than a small number of absolute measurements is required to maintain progress.

Chapter 3 PRESENT FACILITIES AND EXAMPLES OF THEIR USE

3.1 Following the report R.R.3 1965 from the former A.E.A./S.R.C. Research Reactor Committee the S.R.C. agreed to rent neutron beam time at A.E.R.E. and at A.W.R.E. for use by university scientists. At Harwell the instruments have primarily been developed by A.E.R.E. staff, while at Aldermaston instruments have primarily been developed by the university users. Two booklets have been prepared and circulated giving the details and characteristics of the various instruments. The latest of these should be consulted for full information but the following section provides a summary of the 20 instruments currently available at Harwell and the 7 at Aldermaston. For 1970/71 the A.E.A./S.R.C. Agreement allows university scientists to use the Harwell instruments for 40% of the time. At Aldermaston there are four instruments which can be used full-time; the three others are available for 50% of the time. The Agreement is operated by the Neutron Beam Research Committee (N.B.R.C.) of the Science Board. The N.B.R.C. allocates time to university scientists, makes small financial grants for items special to particular experiments, arranges for new instruments to be developed and provides funds which enable the users to travel and stay at Harwell and Aldermaston.

3.2 The experimental equipment at present available at DIDO, HERALD and PLUTO to research workers in the neutron beam field has been built up over a number of years by a programme of continuous expansion. This has increased both the total numbers of instruments and their capability of collecting data. The installation of multiple detector assemblies and the use of small computers for on-line control and data handling has significantly increased the effectiveness of the equipments. In addition to the major facilities, there is a considerable investment in ancillary equipment such as specimen furnaces, cryostats, magnets, sample changers and pressure cells.

(a) Crystallographic studies

Apparatus available for single crystal studies comprises four on-line computer controlled four-circle single-crystal diffractometers together with one prototype instrument which operates under punched tape control: all these are at Harwell.

For powder diffraction three instruments are in use at HERALD, one is a conventional instrument with on-line control by PDP-8 computer and has a continuously variable wavelength from 0.2 to 2.4 Å. The second is a pulsed diffractometer using time-of-flight techniques – an area where the accumulation of experience will be of great future benefit. The third instrument has a fixed wavelength, on-line control and moves in a vertical plane. This has been built primarily to provide an additional facility but also to gain experience in operating in the vertical plane as this mode will be used on the angled holes in H.F.B.R. On DIDO and PLUTO there are three large-sample instruments, one having a variable input wavelength, and one a multi-detector assembly. A pair of small general purpose instruments also exist and are used partly for specimen orientation prior to use of the single crystal instruments as well as for conventional powder diffraction.

Also in use is a new equipment employing the Laue method in conjunction with a proportional counter as the neutron detector.

Two polarized neutron diffractometers are installed in PLUTO both using Co-Fe polarizing monochromators with r.f. spin inverters, one instrument having a variable input wavelength.

(b) Dynamical studies

The equipment for experimentation in this field comprises four time-of-flight spectrometers, two of which view a moderator of liquid hydrogen. The instruments have wavelength resolutions between 3% and 15% with 8 detector positions at the HERALD apparatus and 21, 30 and 64 positions respectively on the three Harwell instruments. In all cases data handling is by PDP-8 computer.

3.2 (Cont.)

There are two triple-axis spectrometers at Harwell, one being on a hot source in DIDO, but this is being moved to an ambient hole in 1970 and replaced by a beryllium filter detector instrument.

(c) Defect scattering

Four long wavelength (> 4 Å) instruments exist for studying defect scattering. Two are on HERALD, and are PDP-8 controlled. One is on the cold source and employs a filtered beam which is monochromated by a mechanical velocity selector possessing a subsidiary disc for pulsing the beam. Elastically scattered neutrons are selected by time-of-flight in 16 angle positions. The other instrument at HERALD uses a velocity selector and is employed for transmission measurement experiments. These instruments operate out to wavelengths of 15 Å.

The two instruments at PLUTO output their data by punched tape for off-line processing and both use a rotating disc to pulse a filtered neutron beam: the scattering is measured as a function of angle using time-of-flight techniques.

The equipment summarized under (a), (b) and (c) above represents a basis for a national programme of research and there is no item which can be abandoned without detriment to some branch of the study of condensed matter. Further facilities will be required to take advantage of the new areas of work being developed especially those of interest to chemists. An essential feature of the present S.R.C. arrangements, as with any future higher flux source, is that users have access to a complete range of instruments. In relation to higher fluxes this aspect is discussed in a later chapter.

3.3 The research work which has been carried out by university scientists under the S.R.C. scheme has been described in the annual reports of the N.B.R.C. for 1967, 1968 and 1969. The following statements highlight some of the progress which has been made, but the reports, and the publications referred to in them should be consulted if further details are required.

(a) Progress in crystallography of chemical and biochemical interest

The progress achieved in chemical crystallography in the major international centres of neutron beam research over the last few years has been due very largely to the use of the automatic diffractometers, which enable the pattern of scattered neutrons to be explored in 3-dimensions in a reasonable amount of time. This has resulted not only in a greatly increased accuracy of parameter determination (for example a standard deviation of ± 0.008 Å is claimed for the location of the hydrogen atoms in hexamethylene tetramine) but in providing general conclusions on groups and series of compounds, simply because of the greatly increased output of information. For example, the high accuracy of hydrogen location which neutron beams permit has enabled the problem of random and rigid-body motion to be examined accurately in some of the relatively simple molecules which are capable of theoretical study by quantum chemists. Thus naphthalene, resorcinol, salicylic acid and hexamethylene tetramine have been studied in detail by university groups at Harwell. In the case of the latter substance it has also been demonstrated that a concurrent refinement of x-ray and neutron data produces a more satisfactory result than the use of either data separately.

A study which opens up great future possibilities has been the determination of the structure of samarium bromate which has utilised the Ramaseshan method of solving the phase problem by making measurements at two different neutron wavelengths, one on either side of the resonant wavelength of 0.92 Å for Sm^{149}. Samarium has now been substituted into zinc insulin, giving the prospect of completing the analysis of insulin by examination of this single derivative – in contrast to the five derivatives which had to be prepared and examined in the

3.3 (Cont.)

preliminary x-ray study by the isomorphous replacement method. The only real difficulty in the way of success is the limited neutron flux which is at present available.

It is significant that a large programme to study organic crystals is planned for the high-flux reactor at Grenoble and the protein myoglobin is already under investigation at Brookhaven. In fact, bearing in mind the close relationship with Oxford University an H.F.B.R. at the Chilton/ Harwell complex would give us a much stronger position than either of the above foreign centres to contribute in the biological field. It should also be noted that neutrons possess two other key advantages over x-rays in studies of these large molecules. First, because of the vastly lower energy of the neutrons, there is no deterioration of the crystal through radiation damage caused by the primary beam. Secondly, the fact that hydrogen and deuterium atoms scatter neutrons with opposite phases means that deuterium can be used as a marker to follow changes in, say, enzyme molecules. There are thus major prospects that neutrons can be used to study the function of biological molecules, and not merely to determine their structures.

(b) Progress in magnetic crystallography

Most of the recent investigations in this field have sought to increase our fundamental knowledge of the magnetic interactions between atoms and the factors which determine magnetic arrangements. They have concentrated on binary compounds, such as Fe_2B, Fe_2U and Fe_3P, and on alloy systems in which the magnetism associated with a particular atom, such as manganese, can be 'diluted' by the introduction of other atoms, e.g. Au, Cu, Zn which can be inserted in a variety of ways. In a majority of cases powdered materials have had to be used – with a consequent loss of intensity, which has been the more detrimental because of the high angular resolution which is also required. The latter requirement arises on account of the abundance of phase changes which occur in these systems as a result of the interplay of magnetic forces, thermal motion and interatomic separation: these phase changes are associated with small changes of crystal symmetry which are often very difficult to discern. Real progress will only come when sufficient neutron flux is available to provide beams of magnetically-discerning neutrons which give an angular resolution comparable to that achieved with x-rays. It is however very indicative of the interest in this field (mainly from physicists predominantly interested in magnetism) that the two powder diffractometers at Harwell – which are at present our best instruments for this purpose – have been heavily over-subscribed since the S.R.C. support scheme was introduced.

From a chemical viewpoint the outer electrons which determine the magnetic properties also determine the nature of the valency bonds and, in particular, their ionic and covalent content.

(c) Progress in the study of covalency in inorganic complexes

The magnetic properties of transition metal ions are modified by interaction of the metal with ligand groups. Determination of magnetic moment density can therefore provide important quantitative information about the delocalisation of d orbitals. Recently, groups at Cambridge, Harwell and Oxford have used highly accurate magnetic moment determinations (from several neutron diffraction techniques) to examine the extent of covalency in a number of transition metal compounds with simple ligands, such as O^{2-}, F^-, S^{2-}, Se^{2-}, CO_3^{2-}, as well as unpaired electron distributions in a number of alloy systems. In some compounds neutron diffraction studies provide a unique route to the measurement of orbital overlap, in others the information obtained complements that provided by resonance techniques (nuclear magnetic, electron spin and nuclear quadrupole resonance). When comparisons of the results of different techniques are possible important information about the relative contribution of σ and π bonding is obtained.

So far nearly all investigations have been on magnetically ordered compounds; higher fluxes would enable these studies to be pursued in magnetically dilute systems and in complexes of special interest to inorganic chemists.

Apart from providing greater insight into physical properties affected by covalency such measurements provide essential guide lines for the development of more sophisticated theories of chemical bonding in transition metal compounds and complexes.

(d) Progress in inelastic methods applied to chemistry

Spectroscopy in which the neutrons observed have been scattered inelastically and incoherently is rapidly increasing in importance as a technique in Molecular Spectroscopy. This is because the selection rules are totally different from those governing optical spectra and the intensity of a vibrational band is simply related to the amplitudes of nuclear displacement in the normal mode. Particular use is made of the abnormally high scattering cross-section of the proton; substitution of ^2H for ^1H greatly reduces the scattering cross-section and provides an important diagnostic tool.

Recent applications include:

(i) observation of optically inactive or very weak vibrational modes, which are of importance in determining the thermodynamic and dynamic properties of simple molecules, e.g. torsional modes in $CH_3.CH_3$, $CH_3.CF_3$ and $CH_3.CC\ell_3$.

(ii) studies of the dynamics of polymer main-chains and side-chains and crystal lattices to obtain information relevant to structure-property relations in polymers.

(iii) observation of hydrogen bending vibrations to determine the molecular symmetry of metal hydrides (important as homogeneous catalysts) e.g. $HCr(CO)_5$, $H_3Re_3(CO)_{12}$.

(iv) direct observation of hydrogen bonds.

(v) study of conformations and vibrations of molecules absorbed on catalytic surfaces.

Energy-gain spectra using cold neutrons provide spectral information at frequencies <500 cm^{-1}. Higher frequencies, up to 1200 cm^{-1}, can be obtained in energy-loss experiments using thermal neutrons. There is no doubt that higher fluxes and better spectrometers would produce immediately a vast and timely increase in the data forthcoming from such experiments. Already the time available to chemists in the U.K. is oversubscribed by worthy projects by a factor of 2.

Diffusional modes of molecular motion are now observed as a Doppler broadening of the quasi-elastic peak. Measured as a function of momentum transfer, the time required for a molecule to diffuse through a distance of $\sim$ 10Å can be determined from the broadening. Measurements have been made on liquid H_2, CH_4 and n-paraffins and one can now compare models of viscous flow with experiments at the molecular level. The same technique is being used to obtain the time scale of the localized mobility of molecules trapped in clathrates and of the molecular processes which occur at phase transitions in solids and on melting. There is now the prospect of detecting density fluctuations associated with the movement of reactant and product groups in reactions carried out in condensed phases and this provides a novel probe for the

chemical kinetics of fast reactions. c.f. 2.4 (b) (iii). These experiments have ultimate technological importance since problems of flow, diffusion and chemical reaction in fluids and melts are of great importance in the oil and polymer industries.

A further important application lies in the field of catalysis. It has been shown that the inelastic techniques can study molecules absorbed on catalytic surfaces. Inferences have been made about the nature of binding and mobility of the absorbed molecules. The method has great advantages over infra-red techniques not only because of the scattering cross-section of protons but also because diffusional as well as structural information is potentially available.

All these applications would benefit greatly in precision and scope if higher fluxes were available.

(e) Progress in crystal dynamics

It is convenient to consider progress since 1965 and to examine briefly what has happened since the review by Dolling and Woods (1965). Up to 1965 experimental studies of many of the simpler monatomic metals (Na, Al. . . .), covalent elements and compounds (Ge, GaAs. . .) and ionic compounds (NaI, KBr. . .) were made by neutron spectroscopy.

In the past few years the same, or related, materials have been investigated with greater accuracy, particularly for metals in order to investigate phenomena which involve electron-phonon interaction, and sometimes over a wide temperature range in order to study anharmonic effects, i.e. phonon-phonon interaction. A notable example is the investigation of magnesium by Squires and his students using the DIDO cold neutron apparatus. A relatively fundamental interpretation of the phonon dispersion curves proved to be possible (Pynn and Squires 1968, Pindor and Pynn 1969). Metals with more complicated crystal structures have also been studied; a good example is provided by the investigation of gallium by Waeber (1969) using the PLUTO triple-axis spectrometer. There are 12 phonon branches for this material and group theory was used successfully to predict their various neutron scattering cross-sections and thereby distinguish them. This experiment, which required about ten weeks of machine time, probably represents the limit of what an exceptional scientist can accomplish using existing fluxes and instruments at Harwell.

Experimental difficulties prevented the investigation of crystals of the "inert gas" elements in the initial phase of neutron spectroscopy. More recently solid helium has been investigated over a range of temperature and pressure with results particularly interesting to theoretical physicists (Choquard 1967). The phonon dispersion relations for argon at 4°K have been determined by Batchelder et al. (1970) of Q.M.C.

The study of solid state phase transitions by neutron spectroscopy, which had been proceeding steadily at Harwell and elsewhere on magnetic materials and at Chalk River on ferroelectrics, has noticeably accelerated with the coming into use of the high flux reactor at Brookhaven. A review of neutron spectroscopy experiments by Shirane (1969) was the highlight of the second International Conference on Ferroelectricity.

(f) Progress in magnetic excitations

Neutron inelastic scattering experiments provide direct measurements of the energies of magnetic excitations in solids, leading to detailed information about the interactions underlying magnetic ordering and allowing powerful tests of general theory relating to elementary excitations and critical phenomena. For example, measurements at Harwell of spin wave

dispersion relations in the ionic antiferromagnets MnF_2 and $RbMnF_3$, and of spin correlation in the 3d transition metal nickel, and measurements at Risø on the rare earth metal terbium show that the magnetic exchange interaction is completely different in character in these three types of magnetic system. Studies of this sort are already yielding details of the outer electron structures in magnetic substances. Inelastic neutron scattering has been widely used to study critical effects in magnetic systems, and in particular recent work on nickel and $RbMnF_3$ at Harwell and on iron, nickel and $RbMnF_3$ at Brookhaven has greatly advanced the understanding of fluctuations in spin systems and critical phenomena generally. The technique may also be used to study the excited levels of paramagnetic ions in crystalline electric fields; experiments at Harwell on cerium-group V compounds have provided direct information about the structure of the cerium ion. Some experiments on the interaction of spin waves with other elementary excitations such as phonons have already been carried out, for example at Chalk River on UO_2. Increasing interest in this topic may be expected in future, especially if higher neutron fluxes are available to facilitate the difficult measurements required, and this will lead to a proper understanding of the interactions of elementary excitations in solids.

(g) Progress in liquid dynamics

As has been pointed out the great merit of neutron studies of liquids is that they allow velocity and positional correlation functions to be simultaneously determined. In thinking about the dynamics it is convenient to distinguish between diffusive motions of molecules in small groups and the collective motions in the liquid analogous to phonons in solids. The difficulties of the latter measurements have already been referred to.

Studies of the breadth of the quasi-elastic peak have been pursued, however, both on Aldermaston and Harwell instruments to characterize the diffusing species in benzene and toluene by Walker and colleagues, and by White in alcohols and acetic acids. Studies of scattering from solute ions have also been used to study the dynamics of solvents.

In some cases structural information about the solute and its environment have been required. Advantage has been taken by Mason of the differences between neutron scattering from different isotopes to study solutions in D_2O of copper perchlorate (Cu^{63}, Cu^{65}) and potassium chloride (K^{35}, K^{37}). Nickel perchlorate (Ni^{58}, Ni^{60}) is also being studied.

Substantial progress has been made in the studies of liquid metals and semiconductors. The work of Enderby on liquid rubidium, zinc, tellurium and zinc at varying temperatures and pressures is being related by March to current developments in the theory of the liquid state.

(h) Progress in the study of non-stoichiometric compounds and solid solutions

Bragg diffraction experiments give information on the average unit cell and early work by Roth, and by Willis demonstrated the value of neutron diffraction studies in locating light atoms in solids with very high (5% or more) lattice imperfections. As a result of improvements in diffractometer design most grossly defective solids can now be studied more profitably with neutrons than with x-rays. Work currently in progress suggests a rapid growth in this type of structure determination, particularly as it becomes possible to study high temperature equilibrium phases directly.

An important recent development is the use of longwave length (greater than 5Å) neutrons to investigate diffuse scattering by non-stoichiometric compounds and solid solutions. These studies have only been in progress for a short time, but is already clear that the technique has great potential for providing information about clustering and short-range ordering of atoms in

a wide variety of solids. Not only may light atoms be readily detected by neutrons, but the use of long wavelength neutrons allows the diffuse scattering to be observed with greatly enhanced sensitivity, (x-rays cannot be used because at long wavelengths absorption is prohibitively high).

Very recent experiments include the determination of non-metal atom distribution in metallic hydrides, carbides (e.g. NbC_{1-x}) and nitrides (e.g. TiN_{1-x}). The previously unsuspected short-range ordering revealed in these systems points to extensive structural studies in future (and their possible correlative with other physical properties) on these often technologically important alloy systems.

Defect clustering of a complex nature has also been demonstrated in a number of "ionic" systems (e.g. $Fe_{1-x}0$ and defect fluorite structures). The extension to other defect structures is feasible and the detailed picture eventually presented by these studies is likely to influence considerably our understanding of grossly defect solids.

Emphasis in the near future will be on systems at or near equilibrium but there is obvious scope for the application of these techniques to the study of the ultra-microcystalline changes associated with the pre-precipitation of new phases.

(i) Progress in lattice disorder studies

Using the S.R.C. liquid hydrogen/deuterium source it has been possible to study defects by elastic diffuse scattering of neutrons of wavelengths up to 15Å. This is much greater than can be achieved with x-rays and all complications of double Bragg reflections can be avoided. In this way the Reading group has revealed the importance of pore closure on an atomic scale in reactor irradiated graphite. When disorder is introduced into germanium by irradiation in a reactor both regions of amorphous material and groups of vacancies with associated strain fields can be seen, from the long wavelength scattering, to be present.

Because of the wider wavelength range over which data may be obtained neutrons are proving invaluable for the study of amorphous solids and as well as work on the conventional glassy materials interesting studies are planned on some of the glassy semiconductors. The full range of S.R.C. instruments at Aldermaston is being used for this work by Lorch, Leadbetter and Wright.

Chapter 4. THE NEED FOR HIGHER FLUXES.

4.1. Some examples of cases when higher fluxes are needed have already been referred to. The importance of attaining the highest possible neutron flux is illustrated by the following simple calculation. Neutron beams from the highest flux reactors at present available in the United Kingdom are produced by collimation from a total thermal flux of about 10^{14} n cm^{-2} sec^{-1}, or a few times higher. A well collimated beam, say ½° x 1° divergence, selects only 1.6×10^{-5} of the original flux, so that a 10 cm^2 source in a flux of 10^{14} n cm^{-2} sec^{-1}, yields a collimated beam of this area with a total flux of 1.6×10^{10} n sec^{-1}. This includes neutrons of all thermal energies; if a wavelength spread of a few per cent about the mean is called for, the best flux will be about 5×10^{8} n sec^{-1}, or 5×10^{7} n cm^{-2} sec^{-1}. After partial scattering by the sample, a further selection of directions and energies may be required, so that even with samples of many square centimetres in cross-section, counting rates in some experiments are reduced to a few counts per second. Some weak scattering processes have been studied at significant counting rates of only 5-10 events an hour. At such levels random spurious counts can easily defeat the aim of an experiment.

4.2. For some types of experiment at present fluxes, notably inelastic scattering and defect scattering, the solids which may be studied are necessarily selected far more by consideration of whether a sufficiently large crystal of adequate perfection can be grown, than by their intrinsic scientific interest. In the case of crystallographic studies extinction effects put a severe limit on the maximum size of crystal which can be used and there is then no alternative to a higher neutron flux if success is to be achieved.

4.3. In the crystallographic work with present day fluxes studies of crystals with say thirty atoms per unit cell, including several hydrogen atoms, demand single crystals of a cubic mm or more in volume. To increase the flux by a factor of 20 would allow crystals of this size with perhaps a thousand atoms per unit cell to be studied so that we could begin a wider study of simple biological materials or, alternatively, we would be able to use crystals thirty times smaller and thus only a few micrograms in weight. Such changes would tremendously widen the types of crystals in which we could positively place the hydrogen atoms. To be able to use small single crystals would also permit the wider use of isotopic substitution, with its rewarding possibilities for structure determination.

4.4. Even with inelastic and defect scattering, however, the limitations are set not only by crystal growing technique (though that is, of course, highly important), but also by the fact that the need for big samples rules out the possibility of experiments on compounds containing strong neutron absorbers, and on samples made from costly separated isotopes. Each increase in flux by a factor of 10 enables experiments of present quality to be carried out on a wider selection of substances; probably it is no exaggeration to say that a hundred times as wide a choice is available for each reduction by a factor of 10 in size of crystal demanded.

4.5. Phonon lifetime. The vibrational spectrum of a solid reflects the interatomic forces produced when atoms undergo very small displacements. It is of interest to study larger displacements and the shape and energy width of phonon states could be examined in detail in favourable cases if higher fluxes were available. Such studies are beyond the capabilities of the present U.K. reactors.

4.6. Present fluxes put limitations on defect studies because the concentration of defects required has to be greater than 0.1%, at the very least. Higher fluxes would give a big impetus in this field and make it far less restricted. Thus with a cold source on an H.F.B.R. it would be possible to carry out the most sophisticated experiment in the radiation damage field – namely the study of the angular variation of scattered neutrons from defects produced by electron irradiation. The direct experimental

4.6. (cont.)

study of the relaxation of atoms surrounding impurities or defects can be studied only in very special cases. Higher fluxes are needed to extend the technique to a wide variety of solid solutions.

A higher flux reactor with cold source and guide tubes will allow the wavelength range available for experiments to be extended to longer wavelengths. This will allow additional information about structural disorder to be obtained, information lying outside the scope of normal diffraction or electron microscopy.

4.7. The ability to study many more types of substance is an essential requirement for the deployment of neutron techniques in chemistry. In the chemical application the pattern of results from a range of substances is at least as important as the more detailed measurements on individual substances in physical applications. The availability of higher fluxes will both provide the freedom to investigate more types of substances and enable more significant experiments to be carried out on those few large crystals that are available.

4.8. General comments and conclusion.

It has been argued since 1963 that the neutron fluxes available at the DIDO, PLUTO and HERALD reactors would not be adequate to sustain a U.K. programme comparable with what would be achieved with planned H.F.B.R.'s. The outcome of the record of indecision since 1963 is now beginning to hit us as we see the demonstrably better and wider range of work being done on the Brookhaven reactor at a flux of 7×10^{14} n cm^{-2} sec^{-1} The Franco/German reactor will provide even higher fluxes by 1973.

Without fluxes at least as good as these the U.K. effort which in the past has been as good as any, and which by 1960 had largely overcome the disadvantages of a late start relative to the U.S.A., will have to be limited to secondary problems. The exciting possibilities of biomolecular studies, the studies of phase transitions, of liquids, of defects and of the wide ramifications to chemistry – to all of which U.K. scientists have been playing leading roles – will eventually diminish on the scale of international significance.

Because of delays in the past it is important now to provide some beams with a flux substantially greater than 7×10^{14} n cm^{-2} sec^{-1} – say twice this amount. The importance of further facilities on the same reactor to back up these high flux holes, cannot be over emphasised, since more routine work requiring a high throughput will be expected to be developed by 1976.

The reactor design specification should be 12 high flux (not less than 1.5×10^{15} n cm^{-2} sec^{-1}) beams in the horizontal plane, and the ability to incorporate the special cold and hot sources as required in at least three of them (with multiple viewing on two of these special sources). Such a reactor will allow some number of additional beams at around 7×10^{14} n cm^{-2} sec^{-1} and as many of these should be provided as possible without prejudicing the high fluxes.

Chapter 5. THE NUMBER OF USERS.

5.1. In the 1965 report we estimated that there would be 150 scientists using the reactor of whom 25 would be A.E.A. staff and 25 scientists pursuing "centre programmes". Since that time it has become clear that, although the numbers are not too far out, there are significant differences between the pattern of use which has occurred and that predicted.

5.2. In 1965 and in earlier reports we were having to convince the chemists that the neutron beam technique would become a major one for chemistry. Professor J.S. Anderson was very clear about this and it was emphasized in the 1965 report. What we did not judge was the rate at which this would take-off and the fact that inelastic experiments using cold neutrons would become the largest single category of chemical interest.

5.3. In 1965 we also misjudged the way in which crystallographers would use neutrons – not the total neutron time needed, but the manner in which it is used. Our concept in 1963 and 1965 was that, although there would be some crystallographers with continuing programmes, a major part of the use would be as occasion demanded to solve a particular problem. In practice the use has been primarily of the former type and very little of the casual user. Thus some crystallographers are finding that neutron studies are providing crucial information on the structures of interest on a continuing basis. In this context the severe limitations on available neutron time may have inhibited potential casual users so we should not exclude this element from future plans.

5.4. Another factor emerging since 1965 has been the limitations on the A.E.A. programmes, such that the number of scientists currently engaged on neutron beam work is 15.

5.5. During 1967, 1968 and 1969 the S.R.C./A.E.A. Agreement for use of neutron beams has been in operation as already outlined.

	A.E.R.E. Time on instruments	A.W.R.E. Effective use of total reactor
1967	25%	33%
1968	25%	33%
1969	30%	33%
1970	40%	33%

These figures are a simplification of a more complicated situation which has already been described. The figure for 1970 is the one recently approved by The Science Board.

5.6. With the limitations on available time, investigators have faced serious difficulties in planning their research programmes. Several investigators have commented at times of particular difficulty that if the technique were not so important they would abandon it since the lack of facilities made the running of their research groups so difficult. It is a mark of the importance of the technique that, with one exception, they have felt it necessary to continue. At present all neutron programmes need more time, without the further provision coming into the category of adequate for the science which investigators think should be done. The limitations on available time has undoubtedly held back the full impact of neutron techniques on other areas. Even so the chemical uses have become clearer and may be expected to spread. The work on bio-molecules has also shown promise. Both these areas of use can be expected to expand if the supply of neutrons were greater – both in flux and in reactor time. Another effect of the present limitations is that two Departments which have often expressed interest in starting major groups – which they feel their subject (magnetism) demands – have not been

5.6. (cont.)

able to. In one case a staff member has not been appointed, although the work of the Department needs the neutron information, because adequate assurances about neutron time could not be given.

5.7. The use of neutron beams for the study of condensed matter is a growing point of science. It would not be unreasonable to envisage a doubling by 1980 on the merits of the science, and it is quite clear that with the removal of current limitations on time and flux, the doubling will occur. Some of this will come from people using neutron techniques for their problem who previously have not, i.e. a change in the balance of techniques by existing scientists. There will also be new people taking up the technique.

5.8. We believe that in the longer term several parts of Industry are going to find neutron beam techniques important. We should encourage C.A.P.S. in this field as a means of examining with Industrial Laboratories the potentialities of neutron beam work for their field of interest. The I.C.I. has already shown an awareness of this situation and has had a member of its staff seconded for a period to work with one of the university groups.

5.9. General comments and conclusion

(a) Present and projected university users.

	1967	1968	1969	1975	1980
University Staff	34	36	47	60	60
Res. Assts. and Fellows	15	10	19	40	45
Research Students	28	31	29	50	55
	77	77	95	150	160

The 1967, 68 and 69 figures have been obtained from the Annual Report of the N.B.R.C. The other figures follow from the long term interests expressed by the major groups and our knowledge of the expressed intentions of other scientists to use the reactor if it was built.

(b) The Centre scientific staff.

The need for the Centre to have scientific staff with a continuing scientific interest in the use of neutron beams has been stressed in all previous reports and forms part of the present thinking of the N.B.R.C. The number might be ca. 10% of university users possible growing in the light of experience to, but not exceeding, 20% by 1980.

(c) Total

The numbers of scientists who will make use of the reactor may be conservatively put at 150 + 15 = 165 in 1975 and 160 + 30 = 190 by 1980. In this document we base our case on 150 users.

(d) Some estimate of the number of beam instruments needed to maintain the programme of 150 scientists may be estimated. Each advanced project will need about 3 months reactor time per annum. Assuming that 11 months reactor time is available p.a. and that 150 users are engaged on the equivalent of 100 advanced projects from universities and 10 from the Centre, then 330 instrument months would be required, on 30 beam instruments. Allowing for more than one instrument on some beam tubes, this number of instruments will require close to 20 beam tubes. While this is clearly an approximate approach, equally clearly it shows the scale of beam provision that 150 users will need. We have made no allowance here for the growth of use in technology. It is not going to be possible to provide for this scale of use with the part time use

5.9. (cont.)

of foreign reactors. Thus a one third availability of a reactor having 10 holes operating for 11 months p.a. would give only 37 beam-tube months, compared with the 220 beam-tube months estimated.

It is interesting to note that at present approximately (20 x 0.4 x 11) + (4 x 1.0 x 11) + (3 x 0.5 x 11) = 148 beam-instrument months are available for 80 users and the N.B.R.C. is continuously under pressure about the inadequacy of time made available for crucial experiments in all fields.

(e) A conservative estimate gives 150 users who will need a minimum of 30 beam instruments.

Chapter 6 POSSIBLE SOURCES

6.1. The present reactors

Excluding the universities' teaching reactors (London, Liverpool/Manchester, Scottish) where fluxes are too low to be relevant to the present discussion, there are three reactors in use in the U.K. for neutron beam experiments. These are HERALD at Aldermaston operating at 5MW and DIDO and PLUTO at Harwell operating at 15 and 22MW. The thermal flux accessible at HERALD is 2×10^{13} n cm^{-2} sec^{-1}. Later this year DIDO will be uprated to 22MW and the flux accessible in both DIDO and PLUTO will then be approximately 10^{14} n cm^{-2} sec^{-1}.

The Harwell reactors were designed as materials testing reactors, i.e. for irradiation rather than beam experiments, and the conditions at the reactor face are less than ideal for beam work because of high radiation backgrounds and limited working space. Additionally the thermal flux at one end of a beam tube is less than that which can be achieved in principle for the reactor power quoted. This is a consequence of the core arrangement which has been designed to meet the materials testing requirements. While these disadvantages are accepted at the beginning of a programme they become less acceptable as beam experiments move towards high levels of sophistication.

To achieve the full potential of the use of neutrons in the study of condensed matter which we have outlined, a high flux source designed principally for beam experiments is required. If the source is to provide for the improvements in energy and angular resolution which are considered necessary and for the extension to the range of samples and scope of experiments previously referred to, the unperturbed flux to be aimed at has to be ca 10^{15} n cm^{-2} sec^{-1}. We have already stated that we believe that a reactor becoming available for beam experiments in 1976 should have flux ca x 2 than what the Brookhaven H.F.B.R. provides now (7×10^{14}) and we believe it to be of prime importance that the proposed source should give fluxes of 1.5×10^{15} n cm^{-2} sec^{-1}. This we believe represents an acceptable limit based on known reactor technology. Such figures (ca 10^{15}) are beyond the practical limits of any proposals for the uprating of DIDO and PLUTO.

6.2. The proposed U.K. H.F.B.R.

This reactor is described in more detail in the accompanying papers. Its design has evolved over the last 10 years to the present system. The underlying principle is of a compact core somewhat undermoderated so that the ends of the beam holes can be near the peak thermal flux occuring just outside the core. The design includes 12 beam holes viewing 1.5×10^{15} n cm^{-2} sec^{-1} and a number of high angle (9) and low angle (7) holes are included viewing $6\text{–}7 \times 10^{14}$ n cm^{-2} sec^{-1}. Special sources – cold and hot – are included in the design to enhance the flux at low energies ($< 5 \times 10^{-3}$ eV) and high energies (0.1 – 0.5 eV) respectively. Each of the special sources would be viewed by 3 of the high flux beam holes. The actual discussion about the special sources is not yet final and it is certainly arguable that for the trends of S.R.C. interest two cold sources would be preferable. We shall examine this further.

Since 1963 when the case for a U.K. higher flux source was first established, it has been this reactor (in its various stages of evolution) that the beam users have argued is necessary to meet the needs of individual experiments (flux) and the scope of experiments (flux and number of beam holes). This was reaffirmed in detail in 1965 and holds good today, although we feel that we should indicate what other sources could be considered as developable or usable.

6.3. Lineac sources

The simple (i.e. unboosted) lineac source comprises a ca 100MeV linear electron accelerator delivering a mean electron power of ca 300kW and peak of 150MW to a heavy target. By a sequence of

6.3. (Cont.)

γ production and (γn) reactions, pulses of fast neutrons are produced. The number of fast neutrons produced can be calculated with reasonable accuracy.

The utilization of the fast neutrons depends on the detailed moderation arrangements used. In all the lineac based sources it is unlikely that one moderator system could satisfy the differing requirements of different types of experiments. A number of moderator systems have been considered including that of a cylinder of moderator of up to 6 cm thickness surrounding a uranium target of 8 cm diameter and 18 cm long. While there are many possible variations and while development of ideas about moderator composition may lead to improvements the critical factor is the external "moderator core" area over which thermal neutrons emerge. This is not likely to differ significantly from the above, whatever the details, so that we will use it in estimating thermal fluxes.

The other critical number is the number of fast neutrons produced by the electron pulse. This while increasing with the peak power cannot be increased indefinitely by decreasing the electron pulse time, because at short times there is inadequate filling of the wave guides.

The best figures we have available have been supplied by Mr. M.C. Crowley-Milling of the Daresbury Nuclear Physics Laboratory. They are:

148 MeV
1 A peak. 8μs. 250 p.p.s.
Peak n production 3.6 x 10^{12} n/pulse. (4.5 x 10^{17} n sec^{-1})

These figures are thought to be the reasonable limit on extrapolated klystron technology and are approximately the same as estimated independently in the super-booster proposal of 1965. The above figures of 3.6 x 10^{12} fast neutron per pulse (250 p.p.s.) are substantially better than those estimated for the Glasgow lineac (4 x 10^{11} at 240) and the Harwell lineac (ca 5 x 10^{10} at 200).

In the process of moderation the pulse becomes longer and exhibits tailing and would undoubted have to be shaped by external choppers. However, for a typical 30μs pulse we could have emerging from the moderator surface (total area ca 10^3 cm^2) about 2 x 10^{13} thermal neutrons cm^{-2} sec^{-1} (energy up to 0.1 eV). In this system the epithermal flux above 0.1 eV (say to 0.5) will be greater than that found in a reactor reflector for the same thermal flux.

These are the essential characteristics of the best lineac system. It is in fact not clear that all the implicit favourable circumstances can be achieved. We shall use these ideal figures later.

6.4. The super-booster

In the super-booster the lineac pulse is injected into a plutonium fuelled fast reactor assembly. The reactivity of the assembly is cycled mechanically and the pulse is injected at maximum reactivity to achieve high multiplication. The need for mechanical cycling limits the pulse repetition frequency (p.r.f.) to 200 p.p.s.

This system has been fully described in AERE-R-5205 and assessed in TNSG/14. As with the lineac the details of utilization depend on the moderator arrangements. Using a 7MW reactor in conjunction with the lineac assumed above, peak fluxes of 1.5 x 10^{16} n cm^{-2} sec^{-1} in 50μs pulses at 200 p.p.s. could be achieved. As will be discussed later a detailed examination of the super-booster performance over the range of experiments envisaged led in 1965 to the conclusion that the H.F.B.R. was preferable to the super-booster. In addition to the comparison of neutron fluxes, it is also the

6.4 (Cont.)

case that a considerable technological development would be necessary to produce the super-booster and although in 1965 its cost was estimated as being comparable with the H.F.B.R. substantially larger contingencies would be required because of the much greater design uncertainties.

6.5 Proton sources

High energy protons represent a more efficient way of producing neutrons per particle than electrons. An accelerator producing 60mA of protons at 1 GeV is fired into a heavy target such as bismuth. The target is surrounded by D_2O and an estimated continuous flux of 2 x 10^{16} thermal n cm^{-2} sec^{-1} could be produced. This was essentially the Canadian I.N.G. (Intense Neutron Generator) project. The technological uncertainties in this project are enormous and several years development work is needed even to produce a good cost estimate. The chances of the cost being less than £30M are negligible and the likelihood is that it would be much greater. The Canadian Government recently stopped work on this proposal.

6.6 The SORA project

This is an Italian plan, which has consistently failed to get EURATOM support, for a mechanically pulsed reactor. The pulsing mechanism would be a beryllium block on the end of a 1m rod rotating at 3000 r.p.m. Thus by varying the reflector in this way the thermal neutron flux would be pulsed at 50 p.p.s. For an average power of 0.5MW the peak neutron power corresponds to 150–200 MW in 50μs pulses at 50 p.p.s. (ref. "pulsed high intensity fission sources" Washington, Feb. 1965. p.35).

Much depends on the detailed design and a number of technological problems have to be overcome. We estimate that the above system would provide ca. 3 x 10^{15} n cm^{-2} sec^{-1} in a pulse and in principle the figures could be scaled up to that for a mean power of 5MW rather than 0.5MW. The latter possibly giving 3 x 10^{16}, is conjecture.

In connection with the SORA project it is interesting to note that a mechanically pulsed reactor has been built in the U.S.S.R. In this case the reactivity has been cycled and an average 3kW system has been cycled at 80 p.p.s. with a pulse width of 36μs to give a peak power of 1.08MW. This has demonstrated the feasibility although the particular design was limited by the air cooling employed. We understand that the reactor has now been closed down, but time-of-flight crystallography experiments were carried out with a data collection rate corresponding to a 1MW reactor using conventional crystallography. This was on a limited class of problems and we are not aware of any new result in the neutron study of condensed matter coming from the use of this reactor.

6.7 The Franco-German Reactor

The Franco-German reactor, which is under construction at Grenoble, is designed specifically for beam experiments and uses the principle of the under-moderated core to bring the peak in the thermal flux to an accessible position outside of the core. The reactor has a uranium core with heavy water moderation and cooling, surrounded by a light water pool. It will operate at 57MW giving an unperturbed thermal flux of 1.5 x 10^{15} n cm^{-2} sec^{-1}. There are to be 13 horizontal beam tubes, 3 of which converge on a hot source. The beam tube viewing the cold source position is designed to accommodate a number of neutron guide tubes. The other beam tubes will have cross sectional areas of about 100 cm^2, some of them being tapered along their length. Planning for the construction of the reactor envisages criticality just after the middle of 1971.

6.8 The Brookhaven Reactor

This reactor has been operating since 1968 as a high flux beam facility. It provides nine beam tubes with thermal fluxes of 7 x 10^{14} n cm^{-2} sec^{-1}. The benefit to the study of condensed matter can already be seen and has been particularly evident in crystallographic problems and the dynamics of

6.8. (cont.)

ferroelectric phase transitions, two subjects of special interest at Brookhaven. The U.K. users would see little point in having a copy of the Brookhaven reactor available in 1975/6 with its limited number of beam tubes and fluxes which would have been available for 7 or 8 years, which appears incapable of development beyond 60 MW without extensive modification.

6.9. Other developments

The preceding sections cover the major possibilities. Liquid fuel reactors have been conjectured but are not serious possibilities in the present context. There are also obvious speculations which can be made of permutations of some of the above systems. These would be even more nebulous in design than some of the systems considered.

Chapter 7. COMPARISON BETWEEN CONTINUOUS AND PULSED SOURCES

7.1 This comparison is not a straightforward one – it depends on the type of experiment being carried out, the energy range, the resolution and the mode of operation of the sources. Thus a reactor may be used as a continuous source or, with external choppers, for time of flight work, while a pulsed source may be used with its intrinsic (neutron) pulse or with pulses shaped by rotors.

There are, however, two broad comparisons which can be made initially:

(a) Experiments in which the performance is determined by the time average flux;

(b) Experiments requiring pulsed beams.

7.2 In the first category we have all conventional crystallographic methods, triple axis-spectrometer for inelastic work and velocity selector methods for long wavelength diffuse scattering experiments. For a thermal flux F_c n cm^{-2}sec^{-1} from a reactor, F_p n cm^{-2}sec^{-1} thermal neutrons within a pulse from a pulsed source, a pulse repetition frequency p (p.p.s.) and a pulse width $\Delta\tau$, we use a symbol P_p^c as a performance ratio of continuous to pulsed source and find that:

$$P_p^c = \frac{C_c}{C_p} = \frac{F_c}{F_p \times \Delta\tau \times p} \qquad \text{(a)}$$

where C_c and C_p are the time averaged fluxes in the continuous and pulsed cases respectively. The following are of interest:

(a) $F_c = F_p$, $\Delta\tau = 50\mu$s. p = 200.

$$P_p^c = 100$$

(b) DIDO/PLUTO to best lineac

$$P_p^c = \frac{10^{14}}{2 \times 10^{13} \times 30 \times 10^{-6} \times 250} = 700$$

(c) H.F.B.R. to best lineac

$$P_p^c = \frac{1.5 \times 10^{15}}{1.5 \times 10^{11}} = 10^4$$

(d) H.F.B.R. to superbooster

$$P_p^c = 10$$

(e) H.F.B.R. to 5 MW pulsed reactor

$$P_p^c = 20$$

Values of C_c and C_p are given in the table below.

7.3 In the second category of experiment, those requiring pulsed beams, P_p^c depends very critically on the condition of the experiment. Here we include time of flight crystallography in which a chopped beam is diffracted by the sample and the arrival of different wavelengths at a few angles recorded as the

7.3. (cont.)

time of flight spectrum. This has not been used apart from demonstration experiments although clearly it is possible in principle. This category also includes all time of flight dynamical studies. In the experiments in this category demands of resolution put different requirements on the pulse widths and other characteristics. The broad comparisons are:

(a) $F_c = F_p$, equal $\Delta\tau$, $p_c = 2000$, $p_p = 250$.

$P_p^c = 8$

(b) Upgraded DIDO to best lineac ($\Delta\tau = 30\mu s$)

$P_p^c = 40$

(c) H.F.B.R. to best lineac ($\Delta\tau = 30\mu s$)

$P_p^c = 600$

(d) H.F.B.R. to super-booster ($\Delta\tau = 50\mu s$)

$P_p^c = 1$

(e) H.F.B.R. to 5 MW pulsed reactor. ($\Delta\tau = 50\mu s$)

$P_p^c = 2$

Values of $N_c = F_c \times p_c$ and $N_p = F_p \times p_p$ are given also in the table.

Summary of performances

Source	P.R.F. (p.p.s.)		Pulse width μs	Th. flux in pulse $n.cm^{-2}s^{-1}$	C*	N*
	EXT.	INT.				
5×10^{13} reactor	2000	–	–*	5×10^{13}	5×10^{13}	10^{17}
10^{14} reactor	2000	–	–*	10^{14}	10^{14}	2×10^{17}
1.5×10^{15} reactor	2000	–	–*	1.5×10^{15}	1.5×10^{15}	3×10^{18}
Lineac	–	250	30	2×10^{13}	1.5×10^{11}	5×10^{15}
Super-booster	–	200	50	1.5×10^{16}	1.5×10^{14}	3×10^{18}
SORA	–	50	50	3×10^{15}	7.5×10^{12}	1.5×10^{17}
Higher power pulsed reactor	–	50	50	3×10^{16}	7.5×10^{13}	1.5×10^{18}

* C means C_c or C_p depending on source. For reactor $\Delta\tau$ of 30 μsec is assumed when comparing with 30 μsec pulsed source and 50 μsec when comparing with 50 μsec pulsed source. Similar considerations apply to N.

7.4. The point of this broad comparison is to bring out the general conclusions, some of which have previously been drawn. Thus the best lineac (unboosted) is in general inferior to DIDO/PLUTO (10^{14} n cm^{-2} sec^{-1}) in both categories of experiment. (For a restricted range of experiments there is the possibility of improving the performance of lineac work if external shaping choppers are dispensed with. This would allow larger samples to be used compared with a time of flight experiment on a reactor).

The only sources whose performances approach the H.F.B.R. are the super-booster, which is strictly comparable in pulsed experiments but inferior in the continuous experiments, and the 5MW pulsed reactor which is inferior by a factor of 2 only in pulsed experiments but substantially inferior for continuous experiments.

If time of flight crystallography could be made comparable with conventional crystallography, the comparison between the super-booster and the H.F.B.R. depends on the details of resolution etc. as discussed in the 1965 report.

Unboosted lineac sources are not comparable with the H.F.B.R. They are inferior to existing sources except in the special case of high energies (0.1–0.5eV) referred to below.

7.5 From the general comparison given above it is clear that the H.F.B.R. and the super-booster offer comparable performance. It was this fact which led to the detailed examination of their relative performances, optimized for each group of experiments, which was contained in the 1965 report of "The Study Group on high intensity sources of thermal neutrons" TNSG/16, A.E.R.E.

We do not propose to give again all the detailed arguments in that report and its supporting papers. The following table is from TNSG/16 and summarizes the overall comparison (Page 28).

The H.F.B.R. is much preferable for elastic and inelastic experiments using incident neutrons of energy $\leqslant$ 20 meV. The inelastic experiments in this energy region represent one of the most important growing points as described in Chapters 2 and 3. The H.F.B.R. is also preferable for special inelastic methods (e.g. constant $\underline{q}$) in the energy range 20–100 meV..

In the energy range 100–500 meV the super-booster with external choppers is about the same as the H.F.B.R. with a suitably chosen hot source. It is here that the super-booster begins to come into its own. To maintain parity of performance in this region the temperature of the H.F.B.R. hot source would have to be adjusted to suit the neutron energy. For energies greater than 500 meV the super-booster would be superior but there is practically no scientific demand for work in this energy range.

For inelastic experiments in the 20–100 meV range involving general surveys the super-booster with external choppers is preferable at a resolution of 0.5% ($\Delta\lambda/\lambda$) but the two are equivalent at 2%. If the super-booster were run without external choppers it would be preferable at 2% resolution, but this would not allow it to run at optimum conditions for other experiments.

In the crystallography field the H.F.B.R. is preferable for the continuous method. The time of flight method can be shown to be preferable in principle to the continuous method, but there is little to choose in performance between the H.F.B.R. and the super-booster with external choppers.

7.6. We have no reason to differ from the conclusion given on page 3 in TNSG/16 that:

"Over the whole scientific area considered, the versatility of the H.F.B.R. allows something approaching the optimum performance to be achieved in all the individual areas simultaneously".

and that:

SUBJECT	SPECIAL FEATURES		PREFERABLE SOURCE	ADDITIONAL REMARKS	CHOICE FOR EACH SUBJECT
CRYSTALLOGRAPHY	Continuous		H.F.B.R.	Well tried; considerable experience of data processing in relation to crystal structure determination.	H.F.B.R.
	Time of flight		H.F.B.R. = S.B.(c)*	Either potentially better than continuous: optimum data handling in relation to structure determination to be developed.	
INELASTIC SCATTERING	< 20 meV		H.F.B.R.	Requires cold source.	Depends on importance of energy range. H. F. B. R. or S. B.
	20–100 meV	Special (e.g. const. Q)	H.F.B.R.	Well tried triple axis spectrometer, both apparatus and method.	
		General: 2% resolution	S.B.(c) = H.F.B.R.	S.B. without choppers better than either – but would not allow S.B. to run at optimum in other experiments.	
		General: ½%	S.B.(c)	Requires development of large counter arrays.	
	100–500 meV		S.B.(c) = H.F.B.R.	H.F.B.R. with a series of hot moderators comparable with S.B.(c) performance, but moderator temperatures would have to be adjusted to suit required neutron energy.	
	> 500 meV		S.B.	S.B. completely superior – new field.	
ELASTIC DIFFUSE SCATTERING	< 20 meV		H.F.B.R.	Requires cold source	H.F.B.R.

TABLE SUMMARIZING NEUTRON USERS' COMPARISON OF THE TWO SOURCES

*S.B.(c) = Super-booster with choppers

7.6 (cont.)

"On the grounds of versatility, certainty of success and satisfactory accommodation of the large number of neutron users, a High Flux Beam Reactor would best meet the users' needs".

Since 1965 the large growth of interest among chemists in long wavelength inelastic scattering – a region when the H.F.B.R. is undoubtedly superior – reinforces the earlier decision.

7.7 The lineac based sources, whether boosted or not, have the characteristic that their epithermal flux (>0.1 eV) is greater in comparison with their thermal flux than is the case for a reactor. In this region not only does the super-booster begin to be substantially better than the H.F.B.R. but also the best unboosted lineac sources becomes comparable with DIDO. There is however little demand for experiments in this area although a number of possible points of interest were listed in Appendix E of TNSG/16.

Chapter 8 THE NEUTRON BEAM EQUIPMENT

8.1 Previous discussions of neutron beam equipment for the H.F.B.R. have been up-dated in a report (HFBR/MWG/P3) by a working group of university and A.E.A. users. In listing the instruments required it is convenient to use the headings of Chapter 2, viz. (a) crystallography, (b) inelastic scattering and (c) defect scattering.

8.2 The lists of instruments given below are not intended as a final specification, but as an indication of the range of equipment required on the basis of present-day ideas about experiments. Accordingly the lists will be subject to modification as ideas and techniques evolve.

8.3 As well as allowing greatly improved resolution, shorter counting times and the use of smaller samples, the high fluxes available at the H.F.B.R. will bring into the realm of feasibility new neutron beam techniques. An example is polarization analysis of the scattered neutrons using a 3-axis crystal spectrometer fitted with polarizing monochromator and analysing crystals. The additional information obtained from measurement of the change of neutron spin associated with scattering is of very considerable importance in the interpretation of experiments on materials with complex magnetic structures. In other situations where both coherent and incoherent scattering are present the use of polarization analysis allows the two types of scattering to be distinguished thus providing definitive information from experiments which in many cases would otherwise be largely uninterpretable: this is likely to be of great value for measurements on liquids, molecular crystals, polymers and catalysts and whenever hydrogen atoms are present. A second area which will be broached by the combination of high fluxes and the introduction of new beam methods is that of very long wavelength experiments. It is likely that new techniques for use here will be developed on the basis of focussing and energy analysis of neutron beams through the action of magnetic fields on the dipole moment of the neutron. Long wavelength neutron investigations will lead to important extensions in the work on diffraction from materials with very long structural repeat distances, e.g. biological materials, on defects, on diffusion in liquids, on order-disorder phenomena and on the study of fast chemical reactions.

8.4 To carry out the full range of experiments indicated in chapter 2, it is necessary to have beams of neutrons whose energy distributions correspond to Maxwell temperatures both above and below room temperature. This can be achieved by inserting into the reactor small blocks of hot and cold moderator – i.e. hot and cold sources. The reactor design incorporates provision for mounting two such sources, each viewed by four separate beam tubes (these beam tubes are in addition to the tube used for insertion of the source). These two source positions will allow the use of one hot (say BeO) and one cold (liquid H_2/D_2) source. Among other things the hot source will be used in experiments involving relatively high energy transfers (e.g. in molecular spectroscopy when light atoms are present) or high crystal momentum transfers (e.g. in the measurement of a radial distribution function at large scattering vectors). The cold source will be used for the evaluation of slower diffusion processes in liquids, for measurements relating to very fast chemical kinetics, for observation of scattering from defects and for many other things.

8.5 In this section we list the proposed set of instruments. It should be noted that, as several instruments can in some cases be mounted in parallel or series on a single hole, the planning envisages a total number of instruments which exceeds the total number of holes.

(a) Crystallography

3 powder diffractometers
8 automatic single crystal diffractometers[i, ii, iii]
4 general purpose diffractometer
1 simple time-of-flight diffractometer with single flight path
2 Laue facilities
1 3-axis crystal spectrometer for operation with polarized neutrons[iv]

8.5 (Cont.)

Key to superscripts used for comment: i with optional polarized beam operation, ii one on hot source for operation at short wavelengths, iii two on cold source for operation at long wavelengths, iv included also in list of instruments for inelastic scattering.

The powder and general purpose diffractometers and the Laue facilities will be used for the location and initial examination of Bragg reflections arising from magnetic and light atom cross sections. The single crystal diffractometers will be used for highly accurate measurement of such reflections leading to the determination of precise atom and spin positions. The machine on the hot source will be available for extinction checks and for measurements at high values of momentum transfer; the diffractometers on the cold source operating at longer wavelengths will be used for materials with a large crystallographic repeat distance. The 3-axis spectrometer is an instrument with provision for polarization (and energy) analysis of the scattered neutrons (by means of a second diffracting crystal). The ability to measure the polarization change associated with a scattering process is of great importance as a tool in the interpretation of magnetic scattering from complex structures. Polarized beam operation of the single crystal diffractometers has similar utility – it should be noted that the introduction of samples containing nuclear polarizations will greatly extend the range of possible experiments with corresponding increases in the information obtainable. One use of the time-of-flight diffractometer will be with a pressure cell. This technique allows a "white" beam of neutrons to fall on the sample in bursts and distinguishes among the various Bragg reflections in the scattered beam by means of time-of-flight sorting. A single fixed flight path for the scattered neutrons may be used, thus allowing a simple design of pressure cell to be employed.

(b) Inelastic scattering

5 time-of-flight spectrometers [i, ii]
5 crystal spectrometers for measurement of inelastic scattering (3-axis or Be-filter detector instruments) [iii, iv]
1 spectrometer using a magnetic field to analyse the energy distribution of the scattered neutrons [v]

Key to superscripts used for comment: i two on the hot source utilizing higher energy incident neutrons, ii two on the cold source for use with long wavelength incident neutrons, iii one on the hot source, iv one capable of operation with polarized beams, v for use on the cold source at very long wavelengths.

The basis of neutron time-of-flight spectrometry is to allow bursts of monochromatic neutrons to fall on a sample and then to time the arrivals of the scattered neutrons at a counter. The change in energy and momentum involved in the scattering can thus be inferred. The bursts of monochromatic neutrons incident on the sample are provided either by a mechanical chopper system whose action is analogous to "green-wave" traffic lights; by a Bragg reflection which is switched by rotation; or by a non-mechanical method based on magnetic field control of a magnetic Bragg reflection in a crystal used as a monochromator. In the crystal spectrometers for inelastic scattering measurement, a beam of neutrons from a first monochromating crystal is brought to a sample and the energy distribution of the scattered neutrons is analysed either by means of a second Bragg diffraction or by "Be-filter" placed over a counter. The Be-filter assembly passes only very long wavelength neutrons not capable of being deflected by Bragg scattering in the Be crystal. The advantage of the Be-filter detector method is the large solid angle that can be employed to collect the scattered neutrons. In general the instruments on the hot source will be used in experiments in which higher energy levels are excited – modes involving light atoms. The

8.5 (Cont.)

long wavelength neutrons from the cold source will be utilized in making accurate measurements of low energy levels in solids and liquids, in examining diffusion processes in liquids, in investigating the details of chemical changes both in solution and at surfaces. The use of neutrons from the cold source will be extended to very long wavelengths using probably a spectrometer in which the monochromation of the incident neutrons and the energy analysis of the scattered neutrons is done with magnetic fields which act on the magnetic dipole moment of the neutron.

(c) Defect scattering

2 spectrometers based on helical velocity selectors [i]
2 small angle scattering equipments [ii]
1 Be-filter and chopper diffuse scattering apparatus

Key to superscripts used for comment: i both on cold source, ii one on cold source.

The basis of defect scattering experiments is the study of the pattern of diffuse intensity scattered from a sample placed in an incident beam of monochromatic neutrons. Under appropriate conditions of defect concentration etc., the scattering can be interpreted in terms of a fourier transform of the defect. Thus, the larger the defect the smaller the quotient of angular spread in the scattering pattern over radiation wavelength; this immediately makes clear the utility of long wavelength neutrons. In the case of the first instruments listed above, the incident neutrons are monochromated by helical velocity selectors. The principle involved here is that of a screw rotating about its axis at a fixed speed. Neutrons aimed at the end of the screw and in a direction parallel to its axis will only avoid contact with the screw, i.e. absorption and scattering, if their velocity bears the appropriate relation to the speed of rotation and pitch of the screw. The small angle scattering instruments listed will probably use crystal monochromators. The Be-filter and chopper apparatus obtains a crudely defined long wavelength beam by using a Be-filter which rejects all neutrons with wavelengths short enough to suffer Bragg scattering in the Be. The chopper breaks the beam up into pulses in time and some further selection is carried out by time-of-flight analysis.

8.6 In drawing up the list of 36 instruments given in the last section, the working party noted a number of areas where more or less major development on instrumentation is required. Neutron guide tubes and position sensitive area counters are obvious examples; another is more efficient polarizing monochromator crystals; a fourth is suitable techniques for moving in and out of position the large and heavy instruments. A full list is given in report HFBR/MWG/P3. The benefit of such work to the effectiveness of the neutron beam equipment will be considerable and as early a start as possible should be made so that the results of the development can be incorporated into the instrument designs without delaying them.

8.7 It is clear that computer techniques will constitute a very important aspect of the instrumentation associated with the H.F.B.R. The beam instrument proposals incorporate the use of a number of small computers, each computer being associated with one, two or three beam experiments, for on-line control and data handling. These small computers can thus be regarded in a very real sense as part of the beam instrumentation.

It is estimated that off-line analysis of data and theoretical model testing etc. will require a computing capacity equivalent to the full-time use of a fairly large machine. However, this capacity need not all be at the H.F.B.R. site and a substantial proportion could be associated with university laboratories etc.

Chapter 9 CONCLUSIONS

9.1 The scientific case rests on the crucial nature of the experiments which can be carried out with neutrons in the study of the physics and chemistry of condensed matter. The experiments cover the chemical structure and atomic movements in crystals, polymers, amorphous solids and liquids, and also the magnetic structure and magnetization dynamics of magnetic materials.

9.2 The work carried out during the last few years has shown the importance of neutron techniques for chemical, as well as physical problems and has indicated the possibilities for the study of molecules of biological interest.

9.3 The present fluxes limit the full realization of the neutron method both in individual experiments in all fields and also in the throughput of experiments. A high throughout is of great importance in the applications to chemistry.

9.4 The design figures should be for a source supplying at least 1.5×10^{15} n cm^{-2} sec^{-1}. Over the whole range of experiments something like optimum performance can be achieved if this flux is provided by a High Flux Beam Reactor. We have discovered no technological development which leads us to modify the conclusion of 1965 that the proposed H.F.B.R. is preferable to the "Super-booster" concept. We conclude unequivocally that the reactor of thermal flux of 1.5×10^{15} n cm^{-2} sec^{-1} and with special cold and hot sources is the best solution from the neutron users point of view.

9.5 The number of users expected to use the facility is 150. This is a realistic and, if anything, conservative estimate. One of the consequences of this number is that the reactor should have at least 20 beam holes, so that an adequate amount of neutron beam time is available.

9.6 The scale of use as envisaged on a balanced programme means that instruments should be available in all the main categories – single crystal diffractometers, powder diffractometers, time-of-flight experiments with cold, thermal and hot neutrons, triple axis spectrometers, polarized neutron spectrometers, long wavelength instruments.