THE FUTURE OF INELASTIC SCATTERING STUDIES AT HIGH FLUX REACTORS*

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INTRODUCTION

Neutron diffraction, as we know it today, had its origin at the Oak Ridge National Laboratory in November, 1945, when E. O. Wollan installed a two-axis diffractometer at the Graphite Reactor. A photograph of this first neutron diffractometer is shown in Fig. 1. The second axis was a Geneva type x-ray instrument, which Wollan had used in earlier research in x-ray physics at the University of Chicago. The detector arm was both strengthened and lengthened to accommodate a long BF_3 detector and its associated shielding. However, the shield was much too heavy to be supported by the arm, and it was supported by four guy wires attached to a track above the instrument. The monochromator axis, which held a NaCl single crystal monochromator, was surrounded by shielding of stacked lead and borated paraffin. This shield provided only one exit port for a monochromatic beam, and the angle of the port could not be changed. This instrument remained in almost continuous operation for about ten years, and it is now on exhibit at the Smithsonian Institution in Washington, D. C.

It would be impossible to list the numerous important investigations that were performed on this instrument, but it should be sufficient merely to state that it was used for most of the pioneering work in neutron diffraction by E. O. Wollan and C. G. Shull [1-6] as well as for later experiments by them and their colleagues. At no time during the early experiments could these scientists possibly have visualized the growth of this field and the impact that it would make on solid state physics. It is equally inconceivable that B. N. Brockhouse could have predicted the important contributions from inelastic scattering investigations and the sophisticated techniques now being used in this field of research, when he performed the first experiments on a three-axis crystal spectrometer [7-8] in 1955.

Today both neutron diffraction and inelastic neutron scattering are well understood techniques that are utilized at reactor centers throughout the world. They are among the most powerful tools currently available for the studies of the structural and dynamical properties of solids and liquids. The value of these techniques exists because several characteristics of the neutron make it a unique probe in studying solid-state properties. a) First, and perhaps most important, the energy of thermal neutrons gives them a corresponding wavelength that is comparable to the separation of atoms in condensed systems; hence, thermal neutrons will undergo pronounced interference effects when they are scattered by the atoms in these systems. Furthermore, the neutron energy is less than that usually required to displace atoms in a material, so that the samples are not damaged during an experiment. b) Second, the change in energy of thermal neutrons due to inelastic scattering processes involving the creation and annihilation of excitations in solids and liquids is frequently of the same order of magnitude as the initial energy. Consequently, these energy changes are relatively easy to measure. c) Third, the strength of the nuclear scattering interaction does not vary regularly with atomic number, so that information on light atoms can usually be obtained as easily as information on heavy atoms. Furthermore, since the nuclear scattering interaction varies with isotopes of the same element, it is frequently possible to improve the scattering properties of a

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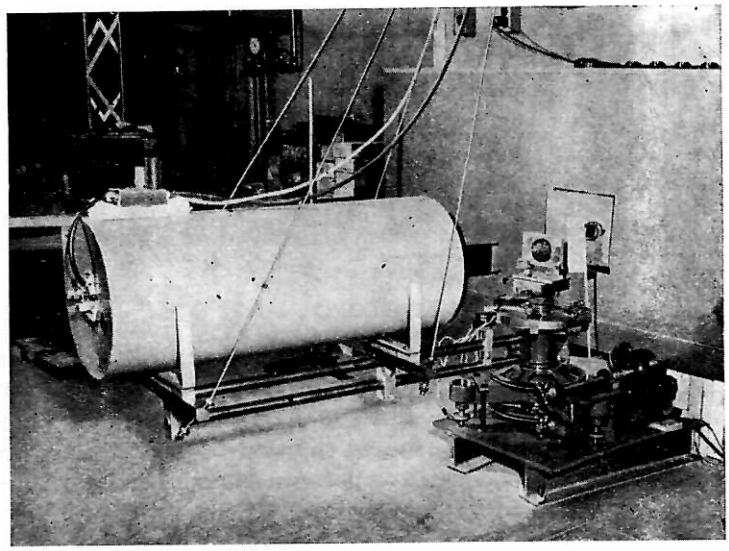

Fig. 1. Photograph of modified x-ray diffractometer used at ORNL in 1945 for the first neutron diffraction research.

sample by substituting isotopes for the elements. d) Fourth, the neutron has a magnetic moment and therefore experiences a magnetic scattering interaction with atoms possessing atomic magnetic moments. This scattering, provides information on both the structural properties and the dynamical properties of magnetic systems. e) Finally, the neutron does not possess an electric charge and can penetrate deeply into solids; hence, information is obtained on the bulk properties of the specimen and not merely on the surface properties, which is usually true for both x-ray and electron scattering.

Just as it was impossible for the pioneers in this field of research to predict the development that has taken place over the past twenty-five years, it is highly doubtful that anyone can accurately predict the future. Although neutron scattering techniques require facilities that are relatively expensive to build and operate, these facilities are not as costly as those required for many experimental investigations in physics. Moreover, these investigations contribute significantly to our understanding of the physical properties of solids and liquids, and they frequently lead to new phenomena or to new materials that have a direct practical application. Consequently, there is no reason to doubt that research of this type will continue to receive support throughout the world and that many significant developments will result from it. All of us involved in the use of neutron beams to study solid-state properties can see many years of important and very productive research ahead of us, and this is particularly true of investigations by inelastic scattering. Such investigations require very high neutron intensities, and there are only a few high-flux reactors currently in operation. This field of research is only half the age of neutron diffraction, and our utilization of it as a tool to study solid-state properties is still in its infancy.

As an attempt to give some insight into the future of inelastic neutron scattering studies, we shall discuss some of the recent developments in this field which we consider to be important. Interesting new techniques together with improvements in older techniques have been developing steadily, and these techniques definitely will be widely used in future research. We shall also discuss some of the problems currently being investigated at research centers with high-flux reactors. Many of these problems form a basis for long-range investigations that will continue for a number of years. Our particular selection of topics is obviously influenced by our personal interests. It is also guided by our current neutron scattering program at the Oak Ridge National Laboratory, where we are fortunate in having the most intense thermal neutron beams presently available for research. This paper is not intended to be a review of the field, and our omission of certain specific experiments or even entire classes of problems should not be misinterpreted as an indication that we do not consider such problems important.

II. EXPERIMENTAL TECHNIQUES

A. General Comments

For experiments in inelastic scattering, any method [9] may be applied, which allows a selection by solid angle and energy for both the primary neutron beam and the scattered neutron beam. There are a few special cases where useful results can be obtained simply by measuring total cross sections or angular distributions, but these cases are exceptional. Energy selection can be performed [10] by filters, by crystal monochromators, by mechanical velocity selectors, and by time-of-flight methods. In most experiments the scattered neutron intensity will be the limiting factor for the resolution that can be obtained, and this intensity requirement frequently dictates the angular resolution of the experiment and the type of energy selection that must be used.

The two techniques that have been the most useful for a wide variety of problems are the triple-axis crystal spectrometer method [11] and the pulsed-monoenergetic-beam time-of-flight method [12] and these techniques will certainly be found at high-flux reactors where inelastic scattering experiments are conducted. Their comparative merits have been discussed many times [13] and there are a number of strong supporters of each technique. The triple-axis crystal spectrometer permits the measurement of the scattered neutron distribution for a particular preselected momentum transfer and energy transfer, and in most machines presently in use only one detector is used. In contrast, the time-of-flight machines measure the scattered neutrons arriving at several detectors at different angles of scattering. Hence, the counting rate is higher than that for crystal spectrometers, but frequently data are collected that have no particular value. Recent evaluations of these techniques by Dorner and Stiller [14] and by Stedman [15] indicate that the time-of-flight techniques are probably preferable for neutron spectroscopic investigations of liquids, but that the crystal spectrometer is preferable for small-angle scattering measurements, for the determination of fine structure in phonon and magnon dispersion curves, and for molecular spectroscopy.

Of course, the best type of instrument also depends strongly on the type of neutron source that is available; e.g., the time-of-flight technique obviously has advantages over the crystal spectrometer if the neutron source is pulsed. Furthermore, the development of new techniques to improve the efficiency of an instrument can frequently alter a particular evaluation, and some of these new developments will be discussed in this section. In Section III a description will be given of the instruments that have been installed at the High-Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. It is our opinion at ORNL that crystal spectrometers and time-of-flight spectrometers supplement each other and that a complete inelastic neutron scattering program must utilize both types. Our instruments have been designed with strong emphasis on maximum flexibility so that any type of experiment can be undertaken.

B. Changes in Neutron Spectra

1. Hot and Cold Neutron Sources

Research reactors usually have moderator temperatures in the range of about 300° to 400°K, depending on the type of moderator used. Consequently, the maximum flux of thermal neutrons is in the vicinity of 0.03 eV, and there are relatively few neutrons with energies ten times higher or lower than this value. Since there are specific types of experiments that require neutrons with energies near the ends of the thermal neutron spectrum, an increase in the intensities of these neutrons can be quite valuable. This intensity increase can be obtained by installing an additional moderator at the reactor end of the beam tube. Cold moderators can provide sizeable gains in the intensity (about an order of magnitude) of neutrons with energies below about 0.1 eV and 1 eV. The most efficient cold sources are liquid H_2 and D_2, and these are about the only substances that can be used in high-flux steady-state reactors, because other substances like methane will be decomposed by the fast neutrons and γ rays. The usual hot sources are BeO and graphite, either heated electrically or by radiation from the reactor. Figure 2 shows the variation of the neutron spectra caused by a graphite moderator [16] placed in the end of a beam tube at the FR2 Reactor at Karlsruhe, when it was operated at temperatures of 1030°K and 1540°K. This graphite moderator was heated by nuclear heating and therefore required very careful thermal insulation. Although the gain in intensity for the higher energy neutrons is appreciable, the graphite block was too small for the neutrons to come into thermal equilibrium at the temperature of the graphite, and the gain was only about 70% of the theoretical gain if complete rethermalization had been accomplished.

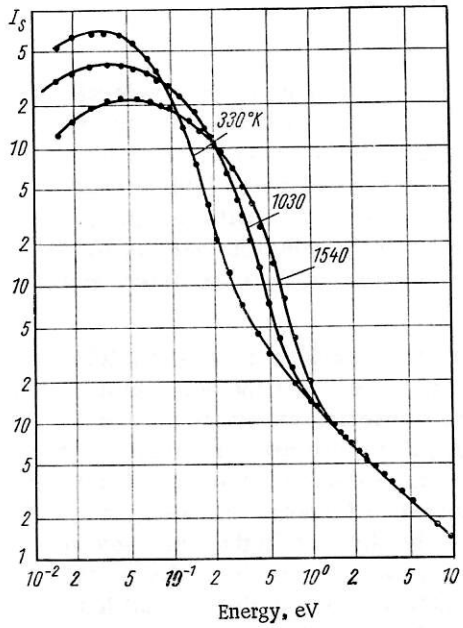

Fig. 2. Neutron spectra obtained from the hot source at the FR2 Reactor at Karlsruhe [16].

At the present time, there are approximately a dozen cold sources either in operation or being planned. There are about half as many hot sources. It is quite possible that this number will increase considerably in the future as more reactors, such as the HFBR at the Brookhaven National Laboratory and the very-high-flux reactor of the Institut Max von Laue-Paul Langevin, are designed with an emphasis on beam hole research. It is highly preferable to design the reactor to accommodate such sources than to try to install them at a later date. Furthermore, in many reactors built primarily for another purpose, such as the HFIR at the Oak Ridge National Laboratory, the beam tubes are too small to permit installation of such sources.

2. Neutron Guide Tubes

One of the most promising techniques of the past several years for neutron scattering experiments is the development of neutron guide tubes [17-19] for obtaining pure beams of thermal neutrons with good collimation. The tubes have totally reflecting surfaces with a very low loss per reflection, and they can be curved tobringthe reflected neutrons away from the high-energy neutrons and γ rays coming from the reactor core. Furthermore, the bent tube behaves as a filter to eliminate those neutrons with a wavelength smaller than some characteristic value. Several tubes can be installed at the same beam port, and since they transmit intense collimated beams of slow neutrons to uncrowded experimental areas well away from the reactor, several different experiments can be performed with the utilization of only one beam port.

A description [20] has been given recently of the first guide tube in operation at CEN Saclay, which is a bent rectangular tube constructed from nickel mirrors. The rectangular cross section is 20 × 50 mm, the radius of curvature is 835 m, and the total length of the tube is 30 m. The average neutron flux in the beam at the end of the guide tube is $2.3 \cdot 10^7$ neutrons/cm^2 · sec, which corresponds to a 95% transmission of neutrons with an energy below 0.005 eV.

Although recent developments indicate that neutron guide tubes can be used for the whole range of thermal neutrons, they appear to be particularly valuable for transmitting low-energy neutrons. Consequently, they should be seriously considered for use with a cold moderator, and a cold source is now planned for the guide tubes in operation at CEN Saclay. At the very-high-flux reactor of the Institut Max von Laue-Paul Langevin, there will be five neutron guide tubes originating from the cold source in addition to others originating from the D_2O moderator. The guide tubes at this reactor have been planned in the reactor design, and they provide neutron beams with different cut-off wavelengths. The long-range planning for such experiments simultaneously with the design of the reactor is necessary in obtaining the most effective installation of these facilities.

C. Improvements in Experimental Efficiency

1. "Small k" Method

In attempts to observe intramolecular vibrational transitions by neutron inelastic scattering experiments, Brugger and his associates [21] have shown that discrete peaks can be observed in the neutron inelastic scattering spectra by observing the spectra at very small momentum changes. Similar peaks are not observed in data at higher momentum changes because the widths of the elastic and inelastic peaks are sufficiently large that they overlap. It was therefore necessary to use warm incident neutrons (0.05 to 0.1 eV) and to observe the inelastic scattering at scattering angles as small as possible (down to 4.8°). By this method it was possible to resolve intramolecular transitions in liquid neopentane, which are unobservable by infrared methods and have appeared only as broad bands in previous neutron scattering experi-

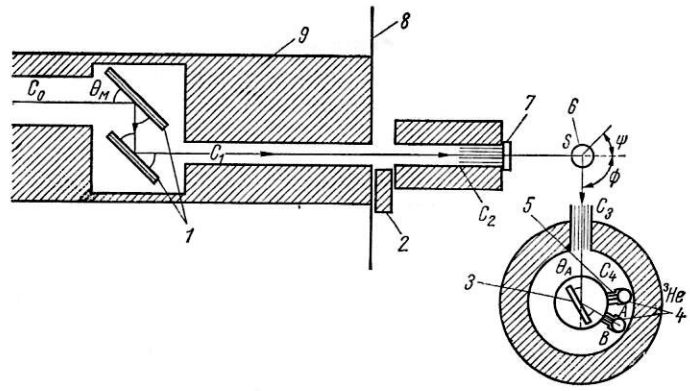

Fig. 3. Schematic diagram of the McMaster University spectrometer at the NRU Reactor at Karlsruhe [16]. 1) Monochromating crystals; 2) gate; 3) analyzing crystal; 4) detectors; 5) collimators; 6) specimen; 7) monitor; 8) reactor face; 9) beam plug.

ments. Although the data in these experiments were obtained with a phased-chopper velocity selector, this technique can also be used with other time-of-flight instruments and with triple-axis crystal spectrometers.

2. New Monochromating Crystals

The need for monochromating crystals with a high reflectivity is obvious in all fields of neutron crystal spectrometry. In triple-axis spectrometry the need is strengthened by the fact that crystals are used both as monochromators and analyzers. T. Riste and K. Otnes [22-23] have recently investigated the reflectivity of pyrolytic graphite specimens, which have an ordered hexagonal c axis but no ordering of the axes within the basal planes. These specimens are presently available with a mosaic spread down to 0.3° and with a cross-sectional area of about 100 × 50 mm. A comparison with other crystals normally used as monochromators at the Norwegian Institute for Atomic Energy, Kjeller, showed that the pyrolytic graphite gave a considerable gain in intensity. It was particularly good at long wavelengths, and reflectivities of about 0.9 could be obtained for 4 Å neutrons. These pyrolytic graphite crystals also can be used to reduce the contamination of unwanted neutrons in the reflected beam. Since they are not three-dimensional single crystals, multiple Bragg scattering is absent. Furthermore, because of the high reflectivity, thin crystals can be used, which reduces the background due to incoherent scattering. Riste [24] also investigated the performance of cylindrically curved pyrolytic graphite crystals, and he found that these crystals could provide a worthwhile gain in intensity for experiments that do not require extreme restrictions on the angular divergence of the neutron beam.

Monochromating crystals with very high reflectivities would be particularly important for spectrometers that use a double monochromator such as the one described by Brockhouse, deWit, Hallman, and Rowe [25] which is shown schematically in Fig. 3. These spectrometers are very simple in construction and are consequently less expensive to build than triple-axis units that utilize a single monochromator. Because only the monochromatic beam emerges from the reactor hole, the required shielding is comparatively modest. Furthermore, the specimen position is fixed in space independent of the incident energy so that the moving parts can be similar to a simple diffractometer. In addition, because of the double reflection, the beam should have a high purity and the background should be correspondingly low.

Improvements in other types of monochromating crystals are also being investigated. H. Maier-Leibnitz [26] has proposed a monochromator crystal of lamellas, each of which is inclined relative to the adjoining ones by the mosaic angle. Such crystals would have particular importance in certain applications of neutron diffraction. Scientists at the Oak Ridge National Laboratory have been searching for better polarizing crystals, because the FeCo crystals now used have a reflectivity that is only about one fifth as high as the reflectivity of good nonpolarizing monochromators. This low reflectivity is of considerable concern in all types of polarized neutron experiments, but it is a particular problem in the newly developed

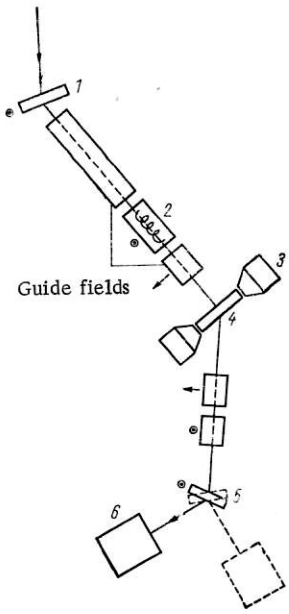

Fig. 4. Experimental arrangement for neutron polarization analysis. Arrows adjacent to the guide fields show the direction of the magnetic fields sensed by the neutrons [36]. 1) Polarizer; 2) flipper; 3) electromagnet; 4) sample; 5) analyzer; 6) counter.

technique of neutron polarization analysis that will be discussed in Section IID. Crystals grown from isotopes of iron and from gallium manganese ferrite look particularly promising for future use in polarized neutron spectrometers.

3. Correlation Techniques

One of the most promising developments for inelastic neutron scattering investigations by time-of-flight techniques is the recent adaptation of correlation methods [27-29] that were originally used in linear network analysis. In the conventional time-of-flight experiments the available primary intensity is used during a very small fraction of time, typically 1%; but these techniques allow the duty cycle to be increased to approximately 50%. In addition to this substantial gain in neutron economy, there can also be a considerable improvement in the energy resolution of the experiment.

The use of correlation techniques is particularly impressive in the measurements of sharp peaks in an intensity distribution, particularly when the ratio of the signal to the uncorrelated background is small and when the time-of-flight spectrum consists of a small number of peaks. In measurements of this type the increase in accuracy is proportional to the ratio of the duty cycles. These techniques give a smaller improvement in measurements of a relatively smooth distribution, and in some cases when the uncorrelated background is quite small, correlation methods may be inferior to the conventional time-of-flight method. Recent investigations [30-31] have shown that the best conditions for performing an experiment frequently do not require the maximum duty cycle available with correlation techniques. Consequently, the ability to change easily the duty cycle of these pseudo-random pulses would be highly desirable in optimizing a particular experiment. These investigations have also included preliminary considerations for using pseudo-random choppers in both the primary and scattered beams.

Another technique for improving the utilization of neutrons in time-of-flight spectroscopy has been proposed [32] which makes use of Fourier synthesis instead of cross correlation. The overall accuracy of this method appears to be comparable with that of the pseudo-random pulsing method, and there are special types of problems in which each method may have specific advantages.

There is little doubt that these new methods will receive widespread use in the future, because there are so many cases connected with inelastic neutron scattering where they offer a substantial improvement over the conventional time-of-flight systems. In view of the possible desirability of using both techniques and also of optimizing the correlation method by selecting the proper pseudo-random sequences, it is apparent that mechanical choppers [29, 33] cannot offer the maximum flexibility needed for time-of-flight facilities. Magnetic choppers previously used [27-28, 34-35] in polarized neutron experiments can offer this flexibility, but they now suffer considerably because of the low reflectivity of the FeCo polarizing crystals. Furthermore, it is not always desirable to use a polarized neutron beam in such experiments. At the present time the most versatile method for time-of-flight experiments appears to be the magnetically pulsed spectrometer that is being developed at the Oak Ridge National Laboratory. A brief description of this instrument will be given in Section IIIB.

D. Neutron Polarization Analysis

A new technique, called neutron-polarization analysis, has been developed recently [36] at ORNL, and it appears that this technique may be extremely important for many types of magnetic scattering investigations. The usual polarized-beam instrument is a two-axis diffractometer in which the measured cross sections involve integration over the final energy and spin distributions. A new dimension is added to these measurements by using a triple-axis spectrometer with polarization-sensitive crystals on both the first and third axes. With such an instrument the distribution of scattered neutrons from an initially polarized

monochromatic beam can be measured as a function of angle, energy, and spin. The reason that investigations of this type must be restricted to very-high-flux reactors is the low reflectivity of the FeCo polarizing crystals. Because it is necessary to use these crystals both as polarizer and analyzer, a factor of about 30 in intensity is lost due to the poor reflectivity of the crystals.

The experimental arrangement for polarization analysis is shown schematically in Fig. 4. FeCo polarizing crystals are mounted in the gaps of permanent magnets on the first and third axes, and the sample is mounted in the gap of an electromagnet located on the second axis. This electromagnet can be rotated so that neutrons can be brought into the field of the electromagnet in either a vertical or horizontal orientation. Radiofrequency coils with a vertical guide field are used as flippers for reversing the direction of the neutron spins. The analyzer is sensitive to the same polarization state as that of the polarizer.

The technique of neutron polarization analysis is most useful in identifying scattering processes that are dependent on the orientation of the neutron spin. In particular, it can be very important in studying magnetic phenomena, because it furnishes a method for separating magnetic scattering from nuclear scattering without changing the sample temperature. Areas in which this technique appears to be particularly useful include (a) separation of nuclear and magnetic Bragg scattering from antiferromagnets, (b) separation of magnon and phonon scattering from ferromagnets and antiferromagnets, (c) separation of paramagnetic scattering from other types of incoherent scattering, and (d) separation of spin-incoherent nuclear scattering in solids and liquids from other types of scattering.

The latter separation could be very important for the application of incoherent inelastic scattering techniques to a wider variety of materials. This technique, which allows a direct measurement of the frequency distribution of the normal modes of vibration of a crystal lattice, has received relatively little attention, because it is difficult to separate the desired incoherent scattering. Of course, the extent to which the technique of neutron polarization analysis can be applied to such problems will depend considerably on the availability of polarizing crystals with higher reflectivity.

III. EXPERIMENTAL FACILITIES AT THE HIGH-FLUX ISOTOPE REACTOR AT ORNL

A. On-Line Computer-Controlled Triple-Axis Crystal Spectrometers

Completely automatic triple-axis crystal spectrometers have been installed at two of the four horizontal beam ports of the High-Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. In order to reduce costs in design and construction, these units are almost identical. Furthermore, the two-axis diffractometer, which has been installed at a third beam port for problems in crystal structure analysis, has identical components for the monochromator axis and its associated shielding. Since these spectrometers are probably representative of crystal spectrometers either in existence or planned for high-flux reactors, a brief description will be given.

These spectrometers have been designed with an emphasis on flexibility so that they can be used for many different types of investigations [37]. The neutron energy from the monochromating crystal can be varied continuously over an unusually large range in scattering angle, which is made possible by a system of wedges that form part of the rotating section of the monochromator shield. The wedges near the direction of the main beam lift above it to allow passage of the beam, while adjacent wedges remain in place to provide shielding. The cavity for the monochromating crystal is relatively large and permits the installation of both a polarizing crystal and a nonpolarizing crystal. Either crystal can be selected and positioned accurately by external controls, so that the spectrometers can be modified relatively quickly for experiments requiring either polarized or unpolarized neutrons. Each spectrometer is controlled on-line by its own computer, a Digital Equipment Corporation PDP-8, through pulsed motors that drive up to six spectrometer axes. The advantages of this system lie in the flexibility of experiment control by stored programs on magnetic tape, which are brought into the fast memory when the conditions of the experiment dictate it. In addition to controlling the spectrometer, each computer has interfacing that allows control of auxiliary equipment such as shutters, velocity selectors, electromagnets, strip-chart recorders, and temperature controllers.

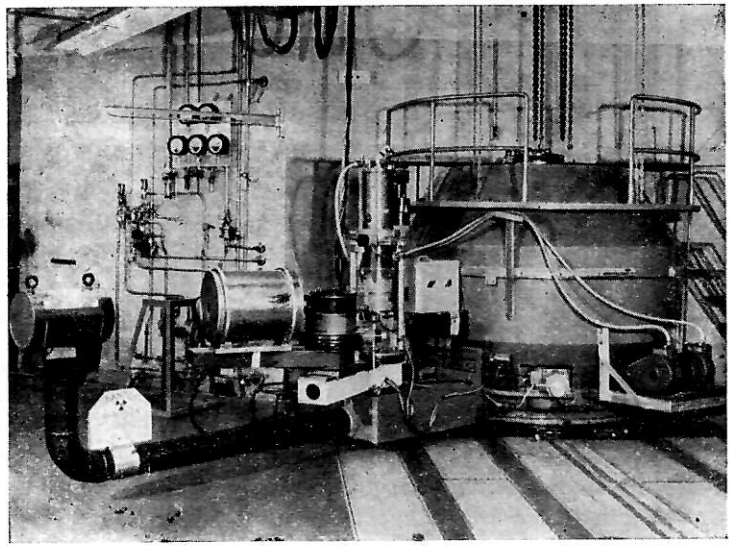

Fig. 5. Photograph of triple-axis spectrometer at beam port HB-3 of the HFIR at ORNL [37].

Although the two spectrometers are identical, one has been used primarily for inelastic scattering investigations with unpolarized neutrons, whereas the other has been used primarily as a polarized beam unit. The spectrometer that has been used for inelastic scattering is shown in Fig. 5. To reduce background scattering three special shields have been installed. The shield at the exit collimator, which is borated paraffin enclosed in Boral, reduces background from air scattering in that region and from scattering by the exit collimator and Soller slits before the sample. The shield around the sample specimen is made from Boral and it has been installed to protect personnel from the thermal neutrons scattered in random directions from large single crystal samples. The shield around the analyzing crystal reduces the number of stray neutrons that might enter the detector. With this shielding arrangement the background not associated with scattering from the sample is approximately one neutron per minute. A low-temperature cryostat is shown on the sample table, and this cryostat allows precise automatic positioning of a single crystal about a vertical axis at any temperature down to the liquid helium region. The second spectrometer, which is used mostly in its polarized-beam mode of operation, is ideally suited for the experiments in neutron polarization analysis described in Section IId.

B. Magnetically Pulsed Time-of-Flight Spectrometer

A very versatile time-of-flight spectrometer, which utilizes a magnetically pulsed neutron beam [38], has been developed at ORNL for inelastic neutron scattering investigations and it is now being installed at the HFIR. This type of spectrometer appears to offer a combination of simplicity, flexibility, and precision, which cannot be obtained with time-of-flight spectrometers now in operation. Furthermore, it is ideally suited for autocorrelation techniques, so that the counting efficiency can be improved considerably for many types of investigations.

The neutron beam is pulsed by diffracting it from a magnetic crystal in which the moment direction can be switched rapidly. Since the intensity of a magnetic reflection is dependent on the direction of the atomic magnetic moments within the crystal relative to the scattering vector, the beam is pulsed by changing the direction of the atomic moments with an applied field. The (111) reflection of $^{7}Li_{0.5}Fe_{2.5}O_4$ is due almost entirely to magnetic scattering; thus the intensity of the beam can be varied from essentially zero with the field applied along the scattering vector to a maximum when the field is applied perpendicular to the scattering vector. Good neutron reflectivity can be obtained with these ferrite crystals, and since the magnetic anisotropy is small, the magnetic spin system can be changed rapidly by small external magnetic fields. The single crystal thus serves both as monochromator and pulser, and any type of pulse shape or length can be readily obtained because the pulsing is accomplished electronically. The pulse repetition rate can also be varied continuously so that the magnetic pulser can operate in any type of autocorrelation mode.

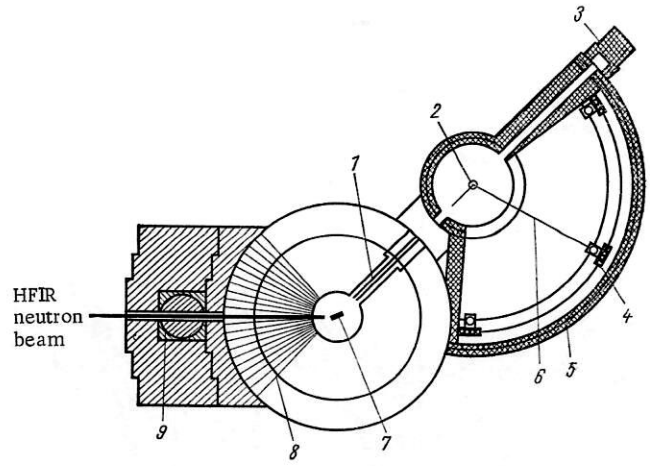

Fig. 6. Schematic diagram of magnetically pulsed time-of-flight spectrometer at the HFIR at ORNL [39]. 1) Exit collimator; 2) sample position; 3) beam stop; 4) neutron detectors; 5) detector shielding; 6) flight path; 7) pulsing crystal; 8) monochromator shield and wedges; 9) shutter.

Since this instrument will use the remaining beam tube at the HFIR, it has been designed for optimum flexibility [39], and a schematic drawing of the spectrometer is shown in Fig. 6. The monochromator-pulser crystal will be mounted in a shield of the same type as those employed on the triple-axis spectrometers described in Section IIIA, and there will be continuous variation of the neutron wavelength over a relatively wide range. The sample assembly and a bank of detectors giving a flight path of 1.5 m will rotate with the monochromator shield. The spectrometer will be on-line to its own digital computer, the Digital Equipment Corporation PDP 15-30, which will operate the magnetic pulser in the optimum manner for a given experiment. If an autocorrelation mode is desired, the computer will pulse the crystal with the desired code of pulses, collect and cross-correlate the time-of-flight data, and display the results of the measurement. The computer will also be used to accurately position the monochromator, sample, and detectors by means of pulsed motors, making it possible to scan along symmetry directions of a single crystal sample.

It is also planned to use the spectrometer with polarized neutron beams in which the polarization is pulsed as a function of time. The neutron beams will have their polarization modulated in a pseudo-random manner so that autocorrelation techniques can be used with samples where the scattered intensity is sensitive to the neutron polarization. The modulation in polarization can be accomplished with a flipper coil similar to the method that has been used previously [24-25, 31-32] or it can be accomplished by reversing the direction of magnetization of a ferrite crystal that will produce a polarized beam. The (111) reflection of GaMn ferrite with the appropriate composition gives a polarized beam with a much higher intensity than that obtained from FeCo polarizers.

The requirement of a very flexible neutron scattering facility has resulted in the construction of a rather elaborate spectrometer with continuously variable incident neutron wavelength and on-line computer control. However, since the magnetic pulsing device itself is extremely easy to construct, a relatively simple time-of-flight spectrometer could be constructed with modest expense, if the on-line computer and continuous wavelength adjustment were not included. This type of device, with the effective intensity increase obtained by autocorrelation techniques, should be seriously considered in the future by anyone planning to start an inelastic scattering program.

IV. PRESENT STATE OF EXPERIMENTAL INVESTIGATIONS

A. General Comments

The importance of inelastic neutron scattering techniques in studying the dynamical properties of different classes of materials has been well demonstrated during the past fifteen years. To obtain an in-

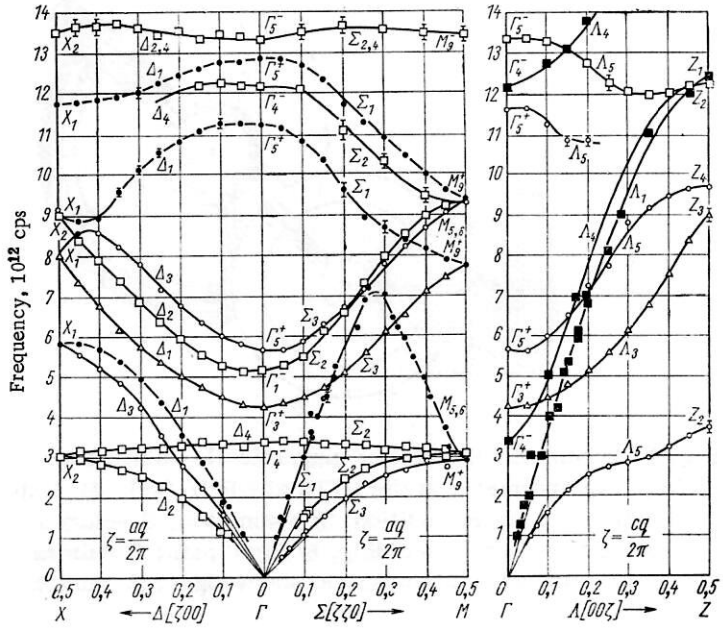

Fig. 7. Low-frequency phonon dispersion curves in the major symmetry directions for TiO_2 [45].

dication of the wide application of these methods in studying condensed systems, it is only necessary to glance through the proceedings of the most recent I.A.E.A. Symposium on Neutron Inelastic Scattering, which was held in Copenhagen in May, 1968. In addition to the papers on Experimental Methods, there are over eighty papers grouped under major categories listed as Dynamics of Solids, Dynamics of Liquids, Molecular Dynamics, and Magnetic Systems.

It is perhaps justifiable to state that in most of these fields inelastic neutron scattering is really just beginning to make a significant impact. As is true of any new technique, the first phase of experiments consisted primarily of exploratory measurements to show the types of information that the technique is capable of providing. The next phase, which involves the use of the technique to investigate particular solid-state phenomena, has been in progress a relatively short time. Since these measurements give the most accurate pictures on a microscopic scale of the atomic forces in solids, liquids, and molecules, there is no visible limit to important investigations that need to be done. The choice of problems is determined primarily by the particular solid-state interests of the scientists.

B. Dynamics of Solids

Coherent inelastic neutron scattering experiments provide the only way at present to get a complete and accurate measurement of the dispersion relations in crystals. Many simple metals and compounds have been investigated, and analyses of these dispersion curves in terms of theoretical models of the crystal dynamics have yielded considerable information about the nature and magnitude of the interatomic forces in solids. However, these investigations have been restricted to simple crystal structures with only three or four atoms per unit cell. The measurements have also been restricted almost entirely to high symmetry directions, where it is easier to separate the longitudinal and transverse modes of vibration, and where the analysis of the normal-mode frequencies in terms of interatomic force constants can be performed with relatively simple calculations. Moreover, most of the models have been based on the Born-von Karman theory of lattice vibrations, which is a phenomenological theory that contains both the adiabatic and harmonic approximations. The former approximation assumes that the electrons in the crystal adjust themselves to the positions of atoms so quickly that they do not have to be considered explicitly and can be included in the force system linking the atoms. The harmonic approximation assumes that the atomic oscillations are small and that any motion of the atoms can be considered as a superposition of the normal modes of vibration of the crystal.

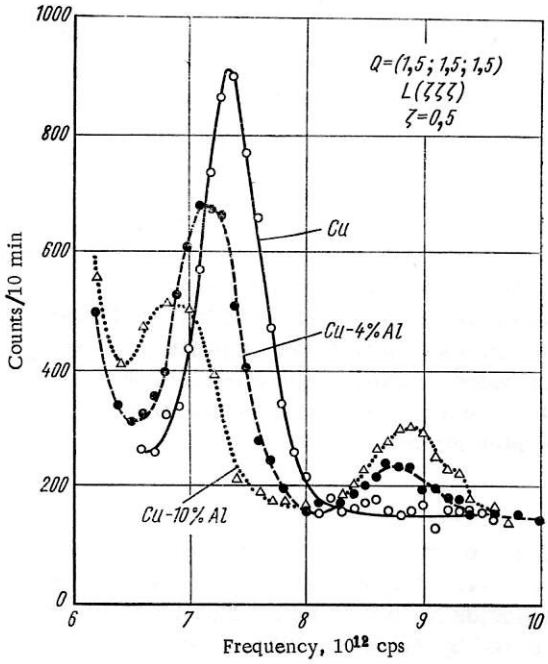

Fig. 8. Constant Q scants obtained for Cu and two CuAl alloys showing peaks from scattering by both in-band and local modes [54].

As the neutron fluxes have become greater and the experimental techniques have been improved, the investigations have been extended to more complicated crystal structures, and many attempts have been made to improve the existing theories. However, except for the alkali metals, these theories [40-44] have not been very satisfactory. The Born-von Karman model with various extensions still provides the best framework for the analysis of experimental results. One of the most complex structures that has been studied [45] is rutile, TiO_2, which has six atoms per unit cell. The lattice dynamics of the rutile structure is of interest, because over twenty compounds with varied dielectric, magnetic, and elastic properties crystallize with this structure. The phonon spectra have been measured for the principal symmetry directions with a triple-axis crystal spectrometer, and some of the measurements are shown in Fig. 7. Calculated curves based on a second-neighbor shell model with tensor forces give a good qualitative fit to the data. A rigid-ion tensor-force model also gives a fair fit to the data, but neither model can adequately describe the qualitative features of the data when they are restricted to axially symmetric forces.

There have been relatively few measurements made of the dispersion curves in the off-symmetry directions, but it is anticipated that many more measurements of this type will be made in the future. Sharp [46-47] has recently performed a relatively thorough investigation of niobium, in which he measured several hundred phonons with wave vectors at general positions in the $[1\bar{1}0]$ reciprocal lattice plane. These measurements show that a number of unusual features of the dispersion relations observed by Nakagawa and Woods [48] along the symmetry directions of niobium can also be traced to the off-symmetry directions. With the assumption that certain of these features are associated with Kohn anomalies, he was able to deduce portions of the Fermi surface of niobium. Kohn anomalies, which are due to singularities in the dielectric function at wave vectors determined by the true Fermi surface, have also been observed in alloy systems [49-50]. Since neutron observations of Kohn anomalies provide one of the few ways to obtain explicit information about the Fermi surface of disordered alloys, measurements of the type made by Sharp could be of particular value in these systems. It has also been shown [51] that sufficiently accurate intensity measurements of the neutron groups produced in one-phonon scattering can permit experimental determination of the polarization vectors of the normal modes and hence the interatomic forces in the crystal. Determination of force constants from an analysis of the polarization vectors is a straightforward method that would eliminate certain problems that can occur in determining them from a nonlinear least-squares fitting of the observed frequencies. Furthermore, according to Brockhouse [52], by proper selections of points in the Brillouin zone for these measurements, the number of measurements need not be very much greater than the number of force constants required.

Coherent inelastic neutron scattering techniques have also been used recently to study the effects of impurities in solids. Since the presence of impurities can seriously alter certain physical properties of solids, it is of considerable interest to determine the influence of these impurities on the dynamical properties of the atoms. An investigation of copper lattices with small amounts (4.1 and 10.0 atomic percent) of aluminum has revealed the existence of localized vibrational modes in these alloys [53-54]. Typical measurements are shown in Fig. 8, which gives a comparison of constant-Q scans at the $\zeta\,\zeta\,\zeta$ zone boundary for pure copper and for the two alloys. The scan for pure copper shows only the expected peak from the coherent neutron scattering by the longitudinal $(\zeta\,\zeta\,\zeta)$ vibrational mode, and this zone-boundary mode has the highest frequency that was observed in copper [55]. The results for the alloys show a decrease in the frequency of the $L(\zeta\,\zeta\,\zeta)$ zone-boundary mode as a function of aluminum concentration, and they also

show an additional peak at a higher frequency. This peak corresponds to the high-frequency vibrations of the light aluminum atoms in the heavier copper lattice and represents the first definite observations of localized modes by coherent inelastic neutron scattering. This technique has the unique advantage of providing information on the degree of spatial localization of the mode in the crystal, and in these CuAl alloys the mode appears to be quite localized; the amplitude of the vibrations is almost completely damped within two unit cells. Additional experiments have been planned with both higher and lower concentrations of aluminum to try to obtain a better understanding of the effects of the impurities on the in-band modes. Evidence for localized modes in an alloy of 88% Ta–12% Nb has also been observed [56], but in these experiments it was not possible to resolve the band phonons and the localized phonon into two separate neutron energy groups. The short range of magnetic forces in insulators makes them quite suitable for the study of localized modes, and such modes have recently been observed in MnF_2 doped with Co [57] and in $KMnF_3$ doped with Co [58]. The resonant perturbation of spin waves caused by Zn impurities in MnF_2 has also been observed recently [59]. However, for a complete understanding of the effects of these various types of impurities, it will be necessary to have the results of many more experiments, in which both the concentration and mass of the impurity atoms are varied systematically.

C. Dynamics of Liquids

Up to the present time thermal neutron inelastic scattering experiments on liquids have been somewhat less rewarding than similar experiments in solids, and our current understanding of liquids is not very satisfactory. The primary difficulty is that the atoms in liquids have less order than those in solids and the simplicity of symmetry cannot be introduced. As discussed by Larsson [59], the recent investigations of importance have been incoherent scattering experiments on molecular liquids and coherent scattering experiments on simple liquids, notably on liquid lead and liquid argon. The work on molecular liquids has given valuable information on the rotation-vibration spectra of the molecules, while the experiments on simple liquids have given further insight to the diffusive and vibratory nature of the atomic motions. The latter experiments definitely show the existence of cooperative vibrational modes, but the vibrational motions appear to be short-time transients which die out into exponential decays. Measurements by Cocking [60] on lead at temperatures slightly above and slightly below the melting point indicate that there is a striking similarity between phonons in liquid lead and solid lead. His results for small $\varkappa$ values show that the phonons in liquid lead fit very closely to the longitudinal phonon dispersion curve for the solid. Hubbard and Beeby [61] have recently proposed a theory of phonons in liquids, based on a treatment of highly correlated many-body systems, and this theory satisfactorily predicts some of the experimental results obtained for liquid lead.

It should be possible to experimentally determine a scattering law $S(\varkappa, \omega)$ that contains all information about the positions and motions of the atoms in a liquid. However, for this determination it is necessary to measure the intensity of scattering in a phonon peak as well as the energy. Furthermore, in order that the Fourier transformation of $S(\varkappa, \omega)$ give accurate values of the space and time dependent pair-correlation function, it is necessary to make the measurements over a wide range of energy and momentum changes. Almost all measurements that have been made of the scattering law up to this time have been of limited statistical accuracy with poor resolution over a relatively narrow range in energy and momentum. With higher flux reactors and more sophisticated instruments, these measurements can undoubtedly be improved. The time-of-flight method with cross-correlation techniques should be particularly appropriate for such measurements.

D. Molecular Dynamics

Although coherent inelastic neutron scattering techniques have been used extensively to study the dynamical behavior of many crystalline solids, investigations of this type are almost nonexistent in the case of molecular solids. While numerous investigations have been made on such solids, almost all of them have utilized incoherent scattering methods with polycrystalline specimens. As previously emphasized [62-63], in order to determine the phonon dispersion curves, from which detailed information on molecular interactions can be obtained, it is necessary to perform coherent scattering experiments on single crystals.

Scattering investigations on single crystal specimens have recently been reported on NH_4Cl [64], ND_4Cl [65], $NaNO_2$ [66], and hexamethylenetetramine [67-68], and it is obvious that this field will grow

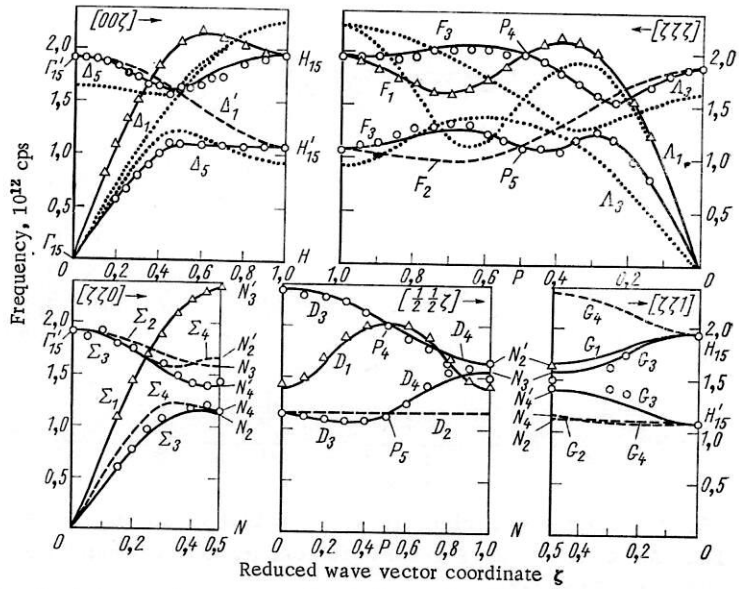

Fig. 9. Dispersion curves for intermolecular modes propagating along directions of high symmetry in hexamethylenetetramine at 100°K. The solid curves are a best least-squares fit to the results for a force model involving first and second nearest neighbor interactions. The dashed curves were computed from this model but represent branches not observed experimentally. The dotted curves are the best fit for a force model involving only nearest neighbor interactions [68].

considerably with higher neutron fluxes and better experimental facilities. Perhaps the most extensive investigation has been made on hexamethylenetetramine [68] which has the chemical formula $(CH_2)_6N_4$; it is certainly the most complicated compound examined by this technique. The measurements were made on a deuterated specimen at 100° and 298°K, and some of the dispersion curves are shown in Fig. 9. The results were analyzed in terms of phenomenological intermolecular force models, which assumed that the molecules were rigid. Models were found that gave a very good fit to the data, but it was necessary to include interactions with both first and second nearest-neighbor molecules. Elastic constants computed from one of the force models show discrepancies with results obtained experimentally by ultrasonic measurements. It is believed that these differences are real and that they can be attributed to anharmonic effects.

E. Magnetic Systems

The investigations of magnetic structures by neutron diffraction has proved to be one of the most important uses of this technique, and magnetic inelastic scattering experiments can be equally rewarding. Such experiments can provide details concerning the behavior of spin waves in various types of magnetic systems, and they can also give direct information on the magnetic exchange forces and anisotropy forces that exist in these systems. Both types of information are extremely important in establishing a better understanding of the magnetic properties of solids. Most of the early investigations of spin waves in magnetic materials were performed using the diffraction technique [69-71], and the techniques involving energy analysis have been employed for only a few years. Not many magnetic materials have been studied, and systematic investigations of the magnetic interactions in these materials are just beginning. Since existing theories appear to give a satisfactory explanation for magnetic exchange interactions between localized magnetic moments in compounds, much of the current work involves investigations of magnetic interactions in metals.

Measurements of magnon dispersion curves in the 3d metals are very difficult because of the high magnon energies that are involved, and some of the first measurements [72-74] were limited by this problem. These measurements indicated long-range magnetic interactions in both Ni and Fe, and they showed

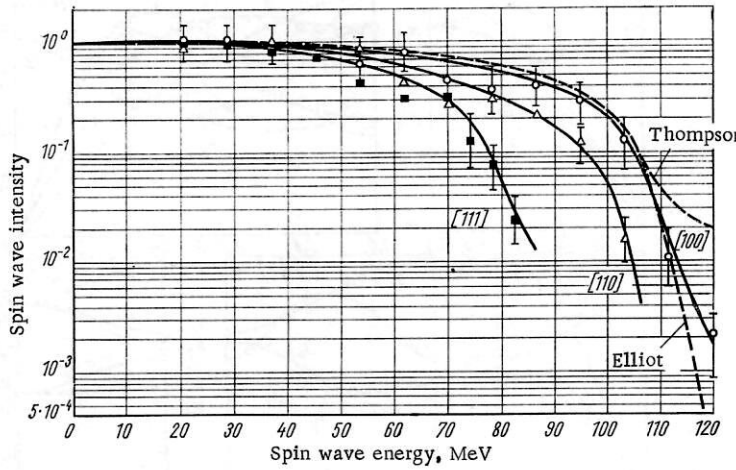

Fig. 10. Spin-wave intensity as a function of energy for ^{60}Ni. The dashed line shows the theoretical fit to the intensity for the [100] direction [75].

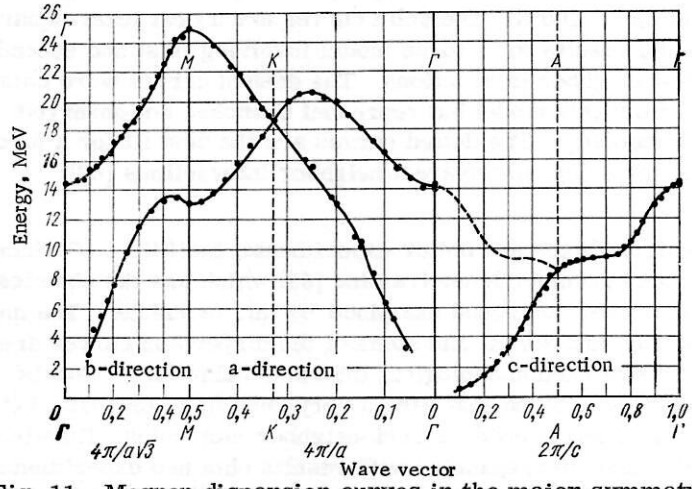

Fig. 11. Magnon dispersion curves in the major symmetry directions for ^{160}Gd at 78°K [85].

well-defined magnons in Fe up to a reduced temperature $T/T_C = 0.995$ but not above T_C. They also showed a strong temperature dependence of the magnon linewidth. Measurements [75-77] of the magnon dispersion curves along the three principal symmetry directions have recently been made to considerably higher magnon energies with large single crystals of ^{60}Ni, Fe, and ^{54}Fe (4% Si). These measurements, which were made on a triple-axis crystal spectrometer, required isotopic specimens to reduce incoherent scattering and contributions from phonon scattering. The dispersion curves were found to rise quadratically in each of the three symmetry directions according to the relation $E = Dq^2$, where D is about 400 MeV · Å^2 for nickel and 280 MeV · Å^2 for iron. At spin wave energies of roughly 100 MeV there was a very sudden decrease in the intensity of the spin waves by more than an order of magnitude. This sudden decrease occurs at somewhat different energies for nickel and iron and at different energies for the three symmetry directions of each metal. The data for ^{60}Ni are shown in Fig. 10. In accordance with calculations the sudden decrease is interpreted as the interaction of the spin-wave dispersion curve with a continuum band of excitations called Stoner excitations. These dispersion curves furnish [78-79] direct experimental information on the energy bands of these metals, and it is highly desirable that they be extended to higher temperatures and to off-symmetry directions.

Measurements of spin wave dispersion curves for the rare-earth metals are of particular value in helping to clarify ideas concerning the very unusual magnetic structures that exist in these metals and

their alloys [80]. The first measurements [81] were made on Tb in the ferromagnetic region at the Risö Research Establishment, and these measurements were extended [82-83] to an alloy of Tb – 10% Ho, which has a wider region of temperature where the spiral phase is stable. Analyses of the results on ferromagnetic Tb did not show the proper conditions for stability of the spiral structure, whereas similar analyses of the alloy results in the spiral region satisfied these requirements. A strong magnon-phonon coupling in the Tb–10% Ho alloy was found to cause a mixing of the transverse phonon and magnon modes propagating in the c direction, and there was pronounced splitting of the two branches. The temperature dependence of the magnon energies [84] shows that the exchange forces may change rapidly with temperature in the helical phase and abruptly at the ferromagnetic transition. In order to understand how variations in both the exchange forces and anisotropy forces can produce the unusual magnetic structures of these metals, it is necessary to make precise measurements of the magnon dispersion curves as a function of temperature in the various magnetic phases. Furthermore, since alloying introduces problems associated with impurity atoms, it seems very desirable to make such measurements on the pure metals. Unfortunately, the high neutron absorption cross sections of several of the rare-earth elements make their investigation quite difficult, and it is necessary to use single crystals grown from low-absorbing isotopes. A program of this type has been initiated at ORNL, and certain measurements have already been made on ^{160}Gd [85] and ^{153}Dy [86]. Gadolinium is of particular interest, because it is a simple ferromagnet below 300°K and behaves as if it contains tripositive ions in $^{8}S_{7/2}$ states. Consequently, the influence of crystalline anisotropy can be expected to be small, and the dispersion curves in the c direction can be taken to be a direct measure of the exchange energy. The magnon dispersion curves for ^{160}Gd at 78°K are shown in Fig. 11, and the analysis shows that at least five interplanar exchange interactions are required to give a satisfactory fit to the data. Similar results have been obtained from measurements on ^{153}Dy and on elemental Ho [87-88] and Er [89], but in all of these metals the out-of-plane anisotropy energy is significant and must be included in the analyses.

IV. CONCLUSIONS

It is hoped that this paper will provide an indication of the types of research programs in inelastic neutron scattering that will exist at high-flux research reactors in future years. We have tried to accomplish this goal by discussing briefly some recent developments in techniques and equipment that will be utilized and by outlining some current investigations that will continue at least into the foreseable future. Our selection of topics was not intended to be complete, and other scientists actively engaged in this field of research might have chosen different experiments to use as illustrations. Several important classes of problems, such as phase transitions, critical scattering, and the dynamics of solidified inert gases, have been arbitrarily excluded. Effects due to changes in temperature, pressure, and external magnetic fields have not been discussed in detail. Relatively little has been mentioned about the interesting information that can be obtained from measurements of phonon and magnon line shapes and intensities. The most important points to emphasize are that thermal neutrons provide a powerful tool to study solid-state phenomena, that the application of this technique is relatively new, and that significant information can still be obtained on all classes of materials.

The long-range future of the field, of course, will depend primarily on the reactor facilities that become available and on the ingenuity of the scientists who use them. It is possible that some of the newest reactors are nearing the limit that can be expected for high-intensity neutron sources. There is still no indication that we are approaching a limit in the ingenuity of the scientists. New and more sophisticated techniques are being exploited continuously, and more difficult problems are being undertaken and solved. Inelastic neutron scattering research will undoubtedly continue to further our understanding of condensed atomic systems for more years than most of us would dare to predict.

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