GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

JOHN JAY HOPKINS LABORATORY FOR PURE AND APPLIED SCIENCE

P.O. BOX 608, SAN DIEGO, CALIFORNIA 92112

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DIFFERENTIAL NEUTRON THERMALIZATION

ANNUAL SUMMARY REPORT

OCTOBER 1, 1965, THROUGH SEPTEMBER 30, 1966

Work done by:

S. Boehm
A. K. Hom
D. Parks
G. Rothbart
W. L. Whittemore

Report written by:

W. L. Whittemore

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Lance E. Williams
Authorizing Official
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ABSTRACT

The General Atomic Neutron Velocity Selector has been used to study the details of neutron scattering in liquid parahydrogen, liquid D₂O, and reactor-grade graphite. The scattering into various angular directions between 30 deg and 150 deg has been studied for incident neutrons with energies ranging up to ~0.7 eV. The energy-transfer cross sections have been evaluated for the larger values of incident energy to provide data in a region not readily accessible to experimenters, using a reactor as a source of neutrons. For the cases of D₂O and graphite, the data are also presented in terms of the Scattering Law. These provide information for much larger values of energy exchange (β up to 10 and above) than previous work, from which the largest values of β were in the order of 2. The cross-section data are compared in detail with theoretical results, where these are available.

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I

INTRODUCTION

The experimental and theoretical work conducted during this contract year has continued the fundamental study of neutron interactions in liquid parahydrogen and reactor-grade graphite and has extended the study to include liquid D_2O . The program has been primarily concerned this year with obtaining information on cross sections in ranges of momentum and energy exchange beyond that already available from previous work in this and other laboratories. As in previous years, the program this year includes:

- a. The experimental determination of the interaction cross sections and the angular and energy distribution for neutrons after single scattering events in various media, and the determination of the related Scattering Law;
- b. Theoretical interpretations of the experimental data for the purpose of making the results available in a form useful to persons interested in fundamental interactions between neutrons and moderating materials.

In this Annual Summary Report, the results of this year's efforts are presented. A few experimental apparatus details are presented, although most of the experimental techniques were discussed in detail in earlier reports.¹⁻⁵ Since the prime interest this year has been in obtaining data for larger energy exchanges (larger β) which require larger values of the incident neutron energy E_0 , an effort was made to improve the experimental conditions for these experiments in particular. These efforts are described in Section II. The subsequent three sections give detailed experimental results on specific moderating media. As this report will demonstrate, the work this year has been devoted in large measure to nonhydrogeneous moderating materials.

II

EXPERIMENTAL TECHNIQUES AND CONSIDERATIONS

As explained in Section I, the aim this year has been to obtain experimental information on double differential cross sections involving larger energy transfers. Since this requires the use of incident neutrons with high energies, an effort was devoted to the related questions of energy resolution and background. Descriptions of some of the developments in experimental technique are given below.

2.1 Improved Shielding Arrangement and Signal-to-Background Considerations

During this contract year, a significant change in the neutron source shield has occurred. The fansteel target was previously shielded by a cube about 2 m on a side, made of borated-paraffin shielding blocks. The interior of this shield contained the fansteel target and massive lead shielding used for attenuating the gamma burst and for reflecting the fast neutrons. An improved version of this target and shield has been built and is illustrated in Figure 1. As illustrated in the drawing, the neutron source is now buried in a dense concrete wall.

Additional shielding to that discussed above has been found very useful in improving the signal-to-noise figure of merit. A section of the 6-m drift tube nearest the neutron source has received additional shielding because this large drift tube has in the past constituted a source of room background neutrons. This is also illustrated in Figure 1. Additional use of B₄C around the detectors and as liners for the 2-m drift tubes between the scattering sample and the detectors has also been beneficial.

The results of the changes in shielding are best demonstrated by displaying a time-of-flight plot for a typical scattering specimen. In Figure 2, we show the scattering of 0.325-eV neutrons by a thin layer of D₂O at 30 deg. This shows that the ratio of counts in the "elastic" peak to the background just before the "elastic" peak is about 20 for this specimen, for which the neutron transmission was about 90%. It is anticipated that additional improvement is still possible through the use of additional dense concrete blocks, since not all of the concrete shielding wall shown in Figure 1 is at present made from dense concrete.

Fig. 1--Experimental arrangement for General Atomic Neutron Velocity Selector and scattering apparatus used in conjunction with the Electron Linear Accelerator.

Fig. 2--A time-of-flight plot illustrating improved signal-to-noise conditions

2.2 Resolution

Since the emphasis this year has been on the study of coherent scatterers such as D_2O and graphite, attention has been directed to improving the angular resolution of the detectors. The He^3 detectors ($1 \times 18 \text{ in}^2$) are mounted in banks of five each. The full angular resolution represented by the 5-in. width is about 3.6 deg. This represents a factor of two improvement over the previous coverage of 7.2 deg with the 10-in.-wide counter banks.

An even more important improvement in resolution has been the increase in the rotational speed of the chopper. In most of the previous work (especially during the past two or three years), the chopper has been used with a rotating speed of 180 rps. By increasing the ac potential applied to the motor, it has been possible to spin the chopper at 240 rps, without other major mechanical modifications. The reader will note that the motor has a nominal 1 hp rating, but can be operated routinely to supply 2 hp, provided only that its rotational speed is in excess of 8000 rpm, which insures adequate air cooling of the motor. The lower thrust bearing for the chopper has previously been a Fafnir Model No. 2MM 304 WI CR (Fafnir Bearing Co., New Britain, Connecticut) with a fibre retaining ring. This bearing has recently been replaced by Barden Model No. 204 HJB (Barden Corporation, Danbury, Connecticut) in the interest of longevity, since the Barden bearing has metal retaining rings and is designed for speeds up to 20,000 rpm and, for the model used, for thrusts in excess of 100 pounds.

The resolution in incident energy has been improved as expected by the $33\frac{1}{3}\%$ increase in rotational speed. For example, the resolution $\Delta E_0/E_0$ is now 6% to 7% at 0.5 eV for 240 rps. Consistent with higher Linac performance, still greater resolution is possible with the same chopper and speed (240 rps) through the use of thinner chopper slits and/or tighter collimation before the chopper. A resolution in the range 2% to 4% at $E_0 = 0.5 \text{ eV}$ is easily obtainable with acceptable beam intensity.

2.3 Transmission of Chopper as Function of Speed

A rotating, neutron chopper with a fixed set of slits (either curved or straight) has a relative transmission which depends on rotational speed. For a set of straight slits and for a fixed speed of rotation, the relative transmission has a low neutron energy cut-off (because of the finite transit time through the chopper-slit system) and rises monotonically with increasing neutron energy (because of a decreasing effect of transit time). By using a curved-slit system, an optimum transmission can be achieved for a given radius of curvature ρ , neutron velocity v , and rotational speed ω .

This relation is given as

$$\rho = v/2\omega \quad (1)$$

For the present slit system, having a radius of curvature of 120.5 in., some typical operating parameters are represented graphically in Figure 3 for speeds of 180 and 240 rps. The present curved-slit system is adequately represented by these data and indicates that a speed of 240 rps cannot be efficiently used to select neutrons with incident energy below ~ 0.2 eV. Figure 3 indicates that lowering the speed to 180 rps will easily accommodate neutrons with energy down to 0.1 eV. Other considerations of Linac usage usually preclude the use of neutrons with energies below about 0.15 eV (because the long "die-away" time in the neutron source for lower energy neutrons seriously reduces the energy resolution).

2.4 Scattering-Sample Holders

A feature of the General Atomic Neutron Velocity Selector is that it accommodates large scattering samples. When the scattering specimens are liquid, care must be exercised to provide uniform specimen thickness. This is sometimes difficult if the additional requirement is made that the specimen holder must have thin walls.

A new approach to the solution of this problem has recently been made. For the D_2O and liquid parahydrogen scattering samples, a holder has been constructed in the form of a manifold of thin-walled aluminum tubes. Recently, it has become possible to obtain inexpensive, thin-walled aluminum tubes* with any reasonable inner diameter and wall thickness. Inner diameters down to 0.005 in can be obtained. The wall thickness can be as thin as 0.001 in. The present tubes for the D_2O runs had 0.082-in. inner diameter and 0.006-in. walls. The liquid parahydrogen specimen was held in tubes with inner diameter of 0.051 in. and wall thickness of 0.003 in.

A point of some interest to other experimenters may be the fact that the aluminum-tube manifold can be constructed for use with liquid hydrogen, using an epoxy cement to seal the joints. Welding is not necessary. The epoxy remains flexible down to very low temperatures and thereby accommodates the rather large changes in size and differential expansions without cracking. The epoxy used for the low-temperature liquid-sample holder was made from equal parts of EPON Resin 826 and hardener Versamid 140. Up to 24 hrs are required for the cement to set. In order to provide for the rather thin, liquid character of the cement when applied, the clearance

*Uniform Tubes Company, Collegeville, Pennsylvania

Fig. 3--Relative transmission of a rotating slit system as a function of neutron energy and chopper speed. Typical behavior is represented for curved and straight slits

between members to be cemented should be kept to a minimum. As shown in Figure 4, one can see the details of the manifold and the piping which prevents gas entrapment in the thin tubes. It may be of interest to note that the holder illustrated in Figure 4 was leak-free the first time it was pressed into service and, at low temperatures, has given no indication of trouble with the seals.

Fig. 4--A photograph of the thin-walled aluminum tubing manifold used for the study of scattering by liquid parahydrogen. The thin-walled tubes are 13 inches high

III

NEUTRON SCATTERING BY LIQUID PARAHYDROGEN

The interest in inelastic scattering of neutrons by liquid parahydrogen stems in large measure from an interest in its use as a neutron shield and as a moderating material for the production of "cold" neutrons. From the very beginning of this program, experimental information has been accumulated on the scattering by liquid parahydrogen.^{1-4, 6, 7, 8} Through the years, theoretical predictions have been made by various authors, including Sarma⁹ and Brimberg¹⁰. More recently, a detailed, complete, theoretical scattering kernel has been generated by Young and Koppel¹¹ for neutron interactions with liquid parahydrogen. Our earlier data has generally agreed with the predictions of theory, though a few annoying differences have existed. The purpose of the present experimental runs was to obtain better data, particularly for the higher-energy incident neutrons and for the cases where the above differences exist.

3.1 General Information

The general considerations of the hydrogen molecule and of neutron interactions in liquid and gaseous hydrogen have been given elsewhere, including our previous publications^{1-4, 12} and will not be repeated here. For ease in discussing the results, however, we will list a few useful relations.

The rotational energy levels in the molecule are given as

$$E_j = 0.015 (J/2) (J + 1) \text{ eV} \quad (2)$$

The kinetics of the collision between a neutron and a hydrogen molecule cannot be represented simply, except for the special case of scattering at 90 deg. For this case, the final energy E of the neutron is related to the incident energy E_o and to the excitation of a molecular level ΔE by the following.

$$E = \frac{E_o - 2\Delta E}{3} \quad (3)$$

This is a special example of the general classical case evaluated by Rothbart.¹³ Rothbart has evaluated the results for the excitation of vibrational levels (0.543 eV) for a variety of incident energies and scattering angles. These are shown in Figure 5. The lowest neutron energy capable of exciting this level is 0.816 eV, which is about $3/2 \times 0.543$ eV, as expected from center-of-mass considerations. An interesting result is that the neutron recoiling in a given direction may have either of two, sometimes widely different, energies.

In Rothbart's considerations of scattering and excitation, the quantum theoretical aspects as well as Doppler effects have been omitted. As Young and Koppel¹¹ point out, vibrational transitions are important only if the incident neutron energy is great enough so that $\kappa^2/8M\omega \gtrsim 1$, where κ is the momentum transfer, M is the proton mass, and $\omega = 0.543$ eV. As an example for our case, a neutron with $E_0 = 0.65$ eV scattered at 90 deg gives $\kappa^2/8M\omega \approx 0.4$. Some effects of the vibrational level may be evidenced in the scattering cross section for such a neutron energy, but these are expected to be generally small.

For completeness and ease in discussing the results, we will include the cross section derived by Young and Koppel¹¹ for hydrogen at 20.4°K. For the "self" scattering cross section per molecule (applicable strictly to gaseous hydrogen, but reasonably accurate for liquid hydrogen if the incident neutron energy is large), they have given the following results:

$$\left(\frac{d^2\sigma}{d\epsilon d\Omega} \right)_{\text{para}} = \frac{\kappa}{\kappa_0} \left(\frac{M}{\pi \kappa^2 T} \right)^{\frac{1}{2}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \times$$

$$\sum_{J=0,2,4} P_J \left[a_c^2 \sum_{J'=0,2,4} + a_i^2 \sum_{J'=1,3,5} \right] \times$$

$$(2J' + 1) \exp \left\{ \frac{- \left| \epsilon + \Delta E + \kappa^2/4M \right|^2}{(\kappa^2 T)/M} \right\} \times$$

$$\sum_{\ell=|J'-J|}^{J'+J} C^2(JJ'\ell, 00) |A_{n\ell}|^2 \quad (4)$$

Fig. 5--A graph plot representation of scattering of neutrons by liquid para-hydrogen in which the 0.543-eV vibrational level is excited

In this expression,

- ϵ = neutron energy transfer;
- κ_0 and κ = initial and final neutron momenta;
- M = proton mass;
- T = temperature,
- $|\vec{\kappa}|$ = neutron momentum transfer;
- P_J = statistical weight of the angular momentum state J ,
- ω = level spacing of the vibrations (0.543 eV);
- a_c and a_i = coherent and incoherent scattering lengths;
- $C(JJ'\ell; 00)$ = Clebsch-Gordon coefficient, and

$$A_{n\ell} = \int_{-1}^1 d\mu \mu^n \exp\left(\frac{-\kappa^2 \mu^2}{8M\omega} + \frac{\ell\kappa\alpha\mu}{2}\right) P_\ell(\mu)$$

3.2 Experimental Conditions

A summary of the more pertinent experimental conditions may be useful to the reader.

The former scattering results were obtained with a sample holder which contained the thin layer of liquid hydrogen between two metal sheets. The first of these holders used a layer of cadmium as one wall and thus could be used only in the reflection position. A later refinement replaced this holder with one having two thin aluminum walls, each 1/16 in-thick. This holder contained a substantial amount of aluminum, but could be used in the transmission position so that 30-deg data were obtained. The large scattering by the aluminum holder was not particularly troublesome because it is mostly elastic, whereas none of the scattering by the liquid hydrogen molecules is elastic (i. e., with $E_0 = E$). The major problem with this holder was the fact that the thickness of the specimen produced by the large unsupported surfaces was nonuniform. This nonuniformity was aggravated by possible buckling at the low temperatures. The latest improvement in liquid-sample holders is the use of the manifold of thin-walled aluminum tubing already discussed in Section 2.4. This latest holder is useful for all angles of scattering, provides uniform scattering thickness, and causes only minimal background scatter.

The transmission of all samples used was about 0.9 for a sample setting at 45 deg to the incident beam. The transmission was essentially the same for all incident energies used (0.224-0.65 eV) because of the relatively slow variation of the hydrogen scattering cross section in this energy range. The contribution of orthohydrogen to the scattering cross section was minimal, since a stabilized mixture of ortho- and para-hydrogen was used for which the ortho content was about 0.21%. Consequently, the parahydrogen sample was about 99.79% pure. The cryostat used for holding the liquid-hydrogen specimen has been described adequately elsewhere.¹

The energies of neutrons selected during several years of study of liquid parahydrogen have ranged from 0.01 eV⁸ in the earliest work to 0.65 eV. During this past year, experimental runs have been made for $E_0 = 0.396$ eV and 0.638 eV. The analysis presented below will be primarily for three values of E_0 : namely, 0.217 eV, 0.396 eV, and 0.638 eV.

3.3 Experimental Results and Discussion

Although the results for neutrons with incident energy of 0.217 eV (Figures 6, 7, and 8) have been discussed before,¹² they will be treated again, for completeness sake, together with the recent results obtained for $E_0 = 0.396$ eV (Figures 9, 10, 11, and 12) and for $E_0 = 0.638$ eV (Figures 13, 14, 15, and 16). These later data obtained for scattering at 30, 60, 90, and 120 deg, and the earlier data, were analyzed to give absolute cross sections, but have not been corrected for the effects of multiple scattering or instrumental resolution. Figures 6 to 16 give the experimental results as a function of energy transfer, together with comparable theoretical calculations of Young and Koppel.¹¹ Typical experimental resolution triangles have been included for convenience.

The reader may note that earlier data obtained for $E_0 = 0.366$ eV and 0.643 eV are missing from the present discussion. These were of poorer quality and suffered in varying degrees from background problems. In view of the significant improvement of the recent data, the older data at about the same energy will be ignored. Although the data obtained at $E_0 = 0.217$ eV and included in the present discussion were also obtained earlier with poorer experimental conditions, its inclusion is justified because the data obtained using lower neutron energy suffered less from the experimental problems.

Taking into account the effects of instrumental resolution, one observes a quite generally acceptable agreement between the theory and experiment. Until a multiple scattering correction is calculated, it will not be possible to conclude whether (a) the absence of deep minima in some of the experimental distributions (particularly at 30 deg scattering as, for

Fig. 6--Scattering of 0.217-eV neutrons at 30° by liquid parahydrogen

Fig. 7--Scattering of 0.217-eV neutrons at 60° by liquid parahydrogen

Fig. 8--Scattering of 0.217-eV neutrons at 90° by liquid parahydrogen

Fig. 9--Scattering of 0.396-eV neutrons at 30° by liquid parahydrogen

Fig. 10--Scattering of 0.396-eV neutrons at 60° by liquid parahydrogen

Fig. 11--Scattering of 0.396-eV neutrons at 90° by liquid parahydrogen

Fig. 12--Scattering of 0.396-eV neutrons at 120° by liquid parahydrogen

Fig. 13--Scattering of 0.638-eV neutrons at 30° by liquid parahydrogen

Fig. 14--Scattering of 0.638-eV neutrons at 60° by liquid parahydrogen

Fig. 15--Scattering of 0.638-eV neutrons at 90° by liquid parahydrogen

Fig. 16--Scattering of 0.638-eV neutrons at 120° by liquid parahydrogen

example, in Figure 6) and (b) the difference between theory and experiments, in some instances, in the relative magnitudes of excitation levels within a single pattern (as, for example, at 60 deg with 0.638 eV scattering, shown in Figure 14) are real. We find generally that the location of levels is in good agreement. In most cases, the relative intensities of peaks agree reasonably well in experiment and theory, as for example at 60 deg for 0.396 eV (Fig. 10), and 30 and 60 deg for 0.217 eV (Figs 6 and 7), and at 90 deg for 0.638 eV (Fig. 15). In some cases, the absolute cross section of the very slowest outgoing neutrons is larger in the experiments, due in part to the effects of multiple scattering in giving a real, large low-energy contribution and in part, perhaps, to the difficulty of accurately determining the background for the lowest energy scattered neutrons.

Although we reported earlier¹² that (a) the widths of some experimental peaks were wider than the theoretical ones after correcting for the effects of instrumental resolution and (b) the experiments found scattering in some instances not predicted by theory, the recent, improved experimental results give agreement, for the most part, between theory and experiment. Some of the observed peaks are still wider than predicted and modified for experimental conditions (at 60° for $E_0 = 0.217$, Figure 7), but it is felt that the effects of multiple scattering may be sufficiently important here to explain the observed results. It may not be necessary to postulate that large liquid-state effects considered earlier¹² are involved, although some effects are possible, since the Debye temperature¹⁴ is about 0.007 eV, and the temperature of the boiling parahydrogen is about 0.0017 eV.

The better data have resolved most of problems found earlier with elastic recoil scattering. As noted above, the theory of Young and Koppel¹² predicts that no scattering will occur for some cases involving pure recoil scattering, even though classical considerations [e.g., Eq. (2)] would lead one to expect such scattering. The theoretical results in Eq. (3) show that such absence of scattering is the effect of the terms A_{nl} , which are approximated by Bessel functions in many instances. The earlier measurements did not verify the lack of such scattering. The situation for scattering of 0.217-eV neutrons remains unchanged from earlier discussions¹² because no new experimental data has been obtained. However, the recent, improved data at other neutron energies verify the predicted results on this point. For example, for 90-deg scattering of 0.396-eV neutrons, the theory predicts a certain amount of recoil scattering (at $E = 0.132$ eV), about as observed in Figure 11. For 90-deg scattering of 0.638-eV neutrons, theory predicts very little recoil scattering at $E = 0.213$ eV, in agreement with the experimental results shown in Figure 15.

One concludes that the agreement is generally good between theory and experiment. Such discrepancies as exist may be explained by the effects of instrumental resolutions and multiple scattering.

IV

NEUTRON SCATTERING BY LIQUID D₂O

4.1 Introduction and General Information

Experimental measurements on the Scattering Law in heavy water, D₂O, have been reported earlier by Egelstaff, Cocking, and Royston,¹⁵ and Haywood and Thorson¹⁶. Their measurements were restricted of necessity to relatively small values of energy transfers and momentum transfers. Additional work on D₂O using "cold" neutrons has been performed by Brockhouse,¹⁷ Hughes¹⁸, and Larssen¹⁹. The theoretical work on D₂O started with the approach of Honeck,²⁰ who treated the scatterer in the "incoherent" approximation. Honeck applied Nelkin's model for H₂O, using appropriate adjustments. This treatment was quite successful in predicting measured neutron spectra and total cross sections. A somewhat similar treatment, with additional refinements, has been given by McMurry²¹ and is called a Scaled Nelkin Model. McMurry²¹ has also used a modification of his earlier treatment of H₂O and includes both free D₂O molecules (17% of total) and two aggregates (each with 41.5% of total) with masses of 167 and 83. A more refined treatment of the elemental neutron interaction with the D₂O molecule has been presented by Butler²² and, later, by Koppel and Young,²³ in which the coherent nature of the scatterer has been treated rather rigorously.

The present experimental program on liquid D₂O has been undertaken to extend the check on theory to higher energy transfers than were previously available. The work in References 15 and 16 used energy transfers ≤ 0.075 eV. The use of the General Atomic Neutron Velocity Selector and the Linac permits measurements to be made for much larger energy transfers, in which all rotational and vibrational energy levels can be excited.

Some general information on liquid D₂O is given for the convenience of the reader. In the following, it will be useful to note that the characteristic frequencies of the D₂O molecule are directly related to similar frequencies in the H₂O molecule, except that the heavier mass of the deuterium atom causes a reduction by a factor of $\sqrt{2}$ in the corresponding frequencies, as shown in Table I. Several experimental determinations of the hindered rotational levels in H₂O and D₂O have been made using "cold" neutrons. Raman spectroscopy has also verified these rotational levels.

More recently, the higher lying vibrational levels have been excited with neutrons in H_2O^{24} and D_2O^5 . Here, we are reporting additional extensive work involving the excitation of these vibrational levels in D_2O . Measurements have been made on both pure D_2O and a mixture of H_2O , HDO , and D_2O . The "pure" D_2O was a biomedical specimen with guaranteed assay $\text{H}/\text{D} < 0.0012$. Recent independent analysis has given a value $\text{H}/\text{D} < 0.025$. A similar analysis of the mixture gave $\text{H}/\text{D} \approx 0.36$.

Table I
CHARACTERISTIC FREQUENCIES IN H_2O AND D_2O

Characteristic Motion	Energy- H_2O (eV)	Energy- D_2O (eV)
Hindered rotation	~ 0.06	~ 0.048
Symmetrical deformation	0.205	0.146
Symmetrical stretching	0.474	0.331
Unsymmetrical stretching	0.488	0.346

4.2 Experimental Procedure and Scattering Results for Pure D_2O

The General Atomic Neutron Velocity Selector described in Section 2.1 was used for the measurements of double differential scattering of neutrons by liquid D_2O . The scattering specimens were contained in thin aluminum tubes having an inside diameter of 0.082 in. and a wall thickness of 0.006 in. and generally discussed in Section 2.4. The transmission of the specimen set at 45 deg to the incident beam was close to 90%. The thickness of the specimens was thereby known with accuracy. Measurements were made at four angles, 30, 60, 90, and 120 deg, except in some cases where the data were taken at 150 deg rather than 120 deg. Table II gives, for the present experiments, the incident energies E_0 used, the spread in incident energy ΔE_0 due to finite emission time ($\sim 7 \mu\text{sec}$) and chopper open time (18-20 μsec), and some typical resolutions ΔE in final energies E .

Using the apparatus described above, double differential scattering results have been obtained for pure D_2O ($\text{H}/\text{D} < 0.025$). Figures 17, 18, 19, and 20 show typical results at four scattering angles for incident neutron energies of 0.157, 0.206, 0.304, and 0.325 eV for the scattering by pure D_2O . Some typical energy resolutions are also indicated on these distributions.

Table II
INCIDENT ENERGIES E_0 USED IN THE D_2O RUNS, WITH
INCIDENT ENERGY RANGE ΔE_0 TYPICAL NEUTRON
ENERGY TRANSFERS E_0-E AND THEIR
RESOLUTION ΔE

E_0 (eV)	ΔE_0 (eV)	E_0-E	ΔE (eV)
0.157	0.008	0.047	0.018
		0.087	0.012
0.206	0.012	0.066	0.026
		0.146	0.014
0.258	0.016	0.058	0.032
		0.158	0.018
0.304	0.022	0.144	0.038
		0.204	0.027
0.325	0.023	0.050	0.069
		0.150	0.043
0.396	0.031	0.146	0.041
		0.316	0.030
0.520	0.047	0.150	0.110
		0.300	0.071
0.646	0.065	0.150	0.125
		0.30	0.090
		0.546	0.046

Fig. 17--The double differential scattering of 0.157-eV neutrons by liquid D_2O

Fig. 18--The double differential scattering of 0.206-eV neutrons by liquid D₂O

Fig. 19--The double differential scattering of 0.304-eV neutrons by liquid D_2O .

Fig. 20--The double differential scattering of 0.325-eV neutrons by liquid D_2O

4.3 Scattering by a Liquid Mixture of D₂O and H₂O

The mixture of light and heavy water will give rise to some mixing of H and D atoms within a single molecule. One expects that if $H/D \approx 0.36$, there will be about 7% H₂O, 39% HDO, and 54% D₂O, assuming that the pairing of the H and D atoms is random. The scattering of 0.404-eV neutrons by this mixed system is shown in Figure 21. In Figures 22 and 23, some experimental energy-transfer cross sections are compared with calculations made by McMurtry²⁵ for this particular case. In general, these data show frequencies for D₂O and H₂O (Table I), as well as other levels, some of which may be due to HDO frequencies (0.174, 0.337, and 0.417 eV), and some of which appear to be combinations of more than one level. Such an example is that for 90-deg scattering of 0.404-eV neutrons (Figure 23), giving an energy transfer of ~ 0.240 eV. No such level exists as a characteristic energy in H₂O, HDO, or D₂O. We suggest that such an energy transfer may be due to multiple scattering in which the 0.205-eV level in H₂O and the 0.048-eV level in D₂O are excited in successive inelastic collisions. Such an explanation is likely, since the sample thickness was chosen to give a transmission of 0.9 for pure D₂O and was thus too thick for the mixture.

The neutron scattering by a mixture of D₂O and H₂O is illustrated by some results shown in Figure 21. These data were obtained before the presence of H₂O was suspected and, as a consequence, the sample thickness (tube diameter, 0.082 in.) was therefore too large. If an inside diameter of 0.082 in. is proper for pure D₂O, then this thickness of scatterer containing $H/D \approx 0.36$ will give a considerable amount of scattering from hydrogen and also increase the effects of multiple scattering.

4.4 Scattering Law for Liquid D₂O

The data for scattering by pure liquid D₂O have been converted to the Scattering Law representation, employing the usual definitions, which are repeated here for clarity.

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b}{4\pi kT} \frac{E}{E_0} e^{-\beta/2} S(\alpha\beta) \quad , \quad (5)$$

where

$$\alpha = [E_0 + E - 2(E_0 E)^{\frac{1}{2}} \cos\theta] / Ak_b T \quad ,$$

Fig. 21--The double differential scattering of 0.404-eV neutrons by a mixture of light and heavy water ($H/D \approx 0.36$). Resolution bars indicate typical widths at half maximum for final energies

Fig. 22--Energy transfer cross section for scattering at 30° by a mixture of light and heavy water. Results computed by McMurry are shown

Fig. 23--Energy transfer cross section for scattering at 90° by a mixture of light and heavy water. Results computed by McMurry are shown

$$\beta = (E - E_0)/k_B T ,$$

$$\sigma_b = \text{bound cross section, } 7.6 \text{ b,}$$

$$A = \text{ratio of mass of scattering nucleus to the mass of the neutron (= 2 for deuterium),}$$

$$k_B = \text{Boltzman constant.}$$

There is some inconsistency in previous treatments by other authors because, in some cases, the value of A is taken to be 2; in others, 20. This makes a factor of 10 difference in the values of α . In the present work we use $A = 2$.

The Scattering Law representation for the pure D_2O data appears in Figures 24 and 25. The accuracy with which the data interleave for all the incident energies of Table II is encouraging. Also shown in Figures 24 and 25 are the Scattering Law calculations of H. L. McMurry²⁵ in the incoherent approximation. It can be seen that the agreement between the theory of McMurry and the present results is satisfactory over the entire range $1 < \beta < 7$. No comparison is made for $\beta = 0$, because instrumental resolution will surely cause the experimental cross sections to fall below predicted cross sections.

The reader's attention should be called to the slightly anomalous behavior of the theoretical Scattering Law curve predicted by McMurry for $\beta = 6$. McMurry's calculations predict that this curve deviates from the general pattern for smaller and larger values of β . This comes about because of a strong resonance at 0.146 eV ($\beta = 5.8$) in all scattering aggregates and free molecules, while there are no resonances near $\beta = 5$ and $\beta = 7$. On the other hand, one does not expect that the curve for $\beta = 2$ will similarly cross the $\beta = 1$ curve because, although one aggregate has a state at 0.053 eV ($\beta = 2.1$), there are also states at 0.025 eV ($\beta = 0.99$) and 0.0265 eV ($\beta = 1.04$). Excitation of these states will keep the $S(\alpha, \beta)$ versus α curves for $\beta = 1$ above the curve for $\beta = 2$. The interested reader is referred to Reference 21 for details of specific frequencies in the various aggregates.

The theory of McMurry has been adjusted as described in detail in Reference 21 to fit the experimental data of Haywood.²⁶ Of course, the experimental data were available for energy transfers only up to $\beta = 3$. It is interesting to note that fit with the present experimental data is good over the whole range tested; namely $1 < \beta < 7$.

Fig. 24--The Experimental Scattering Law data for liquid D_2O compared with calculations by McMurry, where $\beta = 1, 3, 5,$ and 7

Fig. 25--The Experimental Scattering Law data for liquid D_2O compared with calculations by McMurry, where $\beta = 2, 4,$ and 6

4.5 Elastic Scattering by Pure D₂O

Information can be gleaned by examination of the scattering near the "elastic" peak. If the liquid D₂O molecules exist as aggregates only, then recoil scattering by the large aggregate will result in essentially no energy exchange with the scattered neutron. On the other hand, if the scattering is predominantly by single D₂O molecules, the scattered neutron will lose energy in this recoil scattering, with more energy lost for scattering at the larger scattering angles. Figure 26 shows the scattering results calculated by McMurry for a Scaled Nelkin Model (scattering by single molecules only) and for his aggregate model (scattering by single molecules and by larger aggregates). Here one can see that at 30 deg, both models agree on the predicted neutron energy for "elastic" recoil, whereas for scattering at 90 deg, the two models disagree significantly on this energy.

It appears that one should be able to gain information about the size of D₂O aggregates by examining this quasi-elastic scattering. The data in Figure 20 were obtained with resolution adequate for this purpose, and the background scattering, including that from the aluminum holder, has been suitably deducted. At 30 deg, the "elastic" scattering peaks at $E_0 = 0.325$ eV. At 60 deg, the "elastic" peak is shifted slightly (about 0.025 eV) in fair agreement with McMurry's aggregate model. At 90 deg, the "elastic" peak is either greatly reduced compared with some of the inelastic scattering or it is shifted significantly (about 0.05 eV). At 120 deg, the situation is different and indicates the presence again of "elastic" scattering for $E_0 = E$. This appears to be evidence for coherent scattering, which will have to be verified by appropriate calculations. Further work on this interesting point is anticipated in the coming contract year.

Fig. 26--A comparison of calculation based on a scaled Nelkin model and McMurry's aggregate model. Results are shown for scattering at 30° and 90°

NEUTRON SCATTERING BY REACTOR-GRADE GRAPHITE

5.1 Introduction

In previous reports^{5, 27} preliminary data have been presented from a study of neutron scattering by reactor-grade graphite. In this summary report, the reader will find results obtained under better conditions of signal-to-noise and with better resolution. These data have been reduced to a Scattering Law representation and some comparisons are made with earlier data of Egelstaff²⁸ and Young et al.²⁹. An earlier quite complete theoretical treatment of graphite was given by Wikner et al.,³⁰ and a newer treatment has been generated by Young and Koppel³¹.

As pointed out in earlier sections of this Summary Report, a purpose of using the General Atomic Neutron Velocity Selector is to provide Scattering Law data for larger energy and momentum transfers than previously available from experiments using a reactor source of neutrons. In the earlier work in other laboratories, such as that reported by Egelstaff,²⁸ no effect of the higher-lying energy levels could be investigated, since the incident-neutron energy was generally less than 0.1 eV. An exception was the work by Egelstaff and Harris,³² using "cold" neutrons and "hot" graphite which reveal energy levels in graphite at about 0.08, 0.12, 0.18, and 0.215 eV. Young and Koppel³¹ predict energy levels in the phonon spectrum at about 0.02, 0.056, 0.080, 0.108, 0.136, 0.155, and 0.175 up to about 0.208 eV as shown in Figure 27. A discrepancy exists between the predicted level at 0.108 eV and the level observed by Egelstaff at 0.12 eV. This discrepancy was verified in the present program by observations of direct energy-loss scattering which have been reported earlier.⁵

5.2 Experimental Results

Typical experimental results are shown in Figures 28 and 29 for scattering by reactor-grade graphite with $E_0 = 0.227$ and 0.325 eV. These and similar data were converted to a Scattering Law representation, which are presented as $S(\alpha, \beta)$ -versus- α plots in Figures 30 and 31. No correction has been made of these data for the effects of multiple scattering. In addition, one may expect some effects of coherence in the plots shown. At present, neither of these effects has been investigated in detail. However, it is interesting to note that the two points with extra large values on the

Fig. 27--Theoretical frequency spectrum $f(\omega)$ of reactor-type graphite.
(From Young and Koppel, Ref. 31)

Fig. 28--Scattering of 0.227-eV neutrons by graphite at $\sim 24^\circ\text{C}$

Fig. 29--Scattering of 0.325-eV neutron by graphite at $\sim 24^\circ\text{C}$

Fig. 30--Scattering Law representation for graphite for $\beta = 0, 2, 4, 6,$ and 8

Fig. 31--Scattering Law representation for graphite for $\beta = 0, 1, 3, 5,$ and 7

$\beta = 0$ curve occurs for $|K| = 5.38$ and $10.8 \text{ \AA}^{-1}$. The calculation of reciprocal lattice vectors reported in the previous Annual Summary Report⁵ shows several contributions to these two values and makes it likely that these two points are manifestations of the coherent nature of graphite. F. Carvalho³³ at Karlsruhe has observed similar diffraction peaks in the Scattering Law for $\beta = 0$, for graphite, particularly clearly in the range $0.001 < \alpha < 0.1$. It is possible that similar effects of coherence are present in the other Scattering Law curves ($\beta \neq 0$), but detailed calculations must be made to check this point.

5.3 Scattering Law for Graphite; Comparison with Earlier Results

In order to gain some idea as to the relation of the present data to the earlier data of Egelstaff obtained for generally smaller values of β , we have shown, in Figure 32, separate plots of the present data for $\beta = -1$ and -2 for which Egelstaff²⁸ has published data. On the graph for $\beta = -1$, some published calculations of Young, Wikner, and Parks²⁹ are also included. For the case of $\beta = -1$, the present data agree well with the data for Young et al. for all large values α . For the values of α smaller than 1 to 2, both Egelstaff and Young get lower values of $S(\alpha, \beta)/\alpha$, as expected, since the instrumental resolution for the present experiment is expected to result in larger apparent cross sections for small energy transfers ($\beta \approx -1$). Egelstaff's data do not extend to very large values of α , but at his largest values, the values of $S(\alpha, \beta)/\alpha$ tend towards the present experimental values. Egelstaff's data for $\beta = -2$ indicate some problems with accuracy in his data. From the cross comparisons already made (interleaving of Scattering Law data and results of other groups in the lower β range), one concludes that the present results are meaningful except for very small values of β (≤ 1) and α (< 1.5). Additional comparisons will be made in detail with the results of Young and Koppel when these calculations using FLANGE and GASKET become available.

Fig. 32--A comparison of the Scattering Law for graphite at $\sim 24^\circ\text{C}$ for $\beta = -1$ and $\beta = -2$, using the present data, Egelstaff's data, and some theoretical results of Young et al.

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