The Interaction of Slow Neutrons with Nuclei*

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The influence of the molecular binding of the proton has been investigated by measuring the total cross section per proton, for neutrons absorbed by cadmium (C neutrons), of a series of gaseous hydrocarbons of the form C_nH_{2n+2}. The measurements were also extended to include gaseous hydrogen (H_2), "ordinary" water (H_2O), and solid paraffin. The results show that the cross section per proton increases rapidly from the value for the free proton, varying roughly as the square of the reduced mass of the system. The bound proton cross section as measured for the highest hydrocarbon (paraffin) approaches a limiting value of 2.4 times that for the free proton. The dependence of neutron scattering on structure was investigated from measurements on one of the hydrocarbons, cetane ($C_{16}H_{34}$) in its liquid and solid states. The marked increase in the cross section of the liquid state over the solid state indicates that a large amount of coherent scattering must be present. The total cross sections of the deuteron in "heavy" water (D_2O), and of helium, neon, argon, krypton, oxygen, and nitrogen in the gaseous state, were also measured.

INTRODUCTION

FERMI[1] has pointed out that the proton cross section for slow neutrons should depend upon the binding of the proton within the molecular structure. For sufficiently energetic neutrons, this binding effect should disappear. The expression for neutron-proton scattering as developed by Wigner, Bethe and Peierls[2] has significance only when the proton may be considered "free," but for the slow neutrons absorbed in cadmium used in these experiments, the proton cross section is considerably larger than the "free" proton cross section and the use of this expression without modification is not valid.

The theoretical development by Fermi[1] and its extensions by Bethe[3] and Arley[4] show that the proton cross section for zero energy neutrons, calculated with the Born approximation should vary approximately as the square of the reduced mass of the system composed of the neutron and scatterer. In general, however, the above theory predicts the upper limit of the theoretically calculated cross section rather than the experimentally observed cross section, since binding effects will place some restrictions on the transfer of momentum.

Cadmium absorption neutrons (C neutrons) emerging from paraffin at room temperature ($\sim$300°K) have been shown to have energies corresponding to thermal velocities with a most probable energy of 0.025 ev.[5] As this energy is small compared to the binding energy of the protons for C neutrons, the effect of the proton binding should be large.

To investigate the effect of binding on the proton cross section,[6] a gaseous hydrocarbon series of the form C_nH_{2n+2} was selected and a systematic study of the change in cross section per proton was made as the number of protons per molecule increased. The choice of this series offered a twofold advantage in that:

(a) The series consists of substances whose chain-like molecular structure is similar.

(b) The choice of gases avoids solid state difficulties which result in interference effects from crystalline structure[7,8] and intramolecular binding. In fact, subsequent measurements on a liquid and solid hydrocarbon (cetane) show that a large change in the slow neutron cross section

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[1] E. Fermi, Ricerca Scient. **7**, 13 (1936).
[2] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 117 (1936).
[3] H. A. Bethe, Rev. Mod. Phys. **9**, 124 (1937).
[4] N. Arley, Kgl. Danske Vid. Sels. Math.-fys. Medd. **16**, 1 (1938).
[5] Dunning, Pegram, Fink, Mitchell and Segrè, Phys. Rev. **48**, 704 (1935).
[6] A preliminary report on this work was given in Phys. Rev. **54**, 541 (1938).
[7] H. G. Beyer and M. D. Whitaker, Phys. Rev. **57**, 976 (1940).
[8] F. Rasetti, Phys. Rev. **58**, 321 (1940).

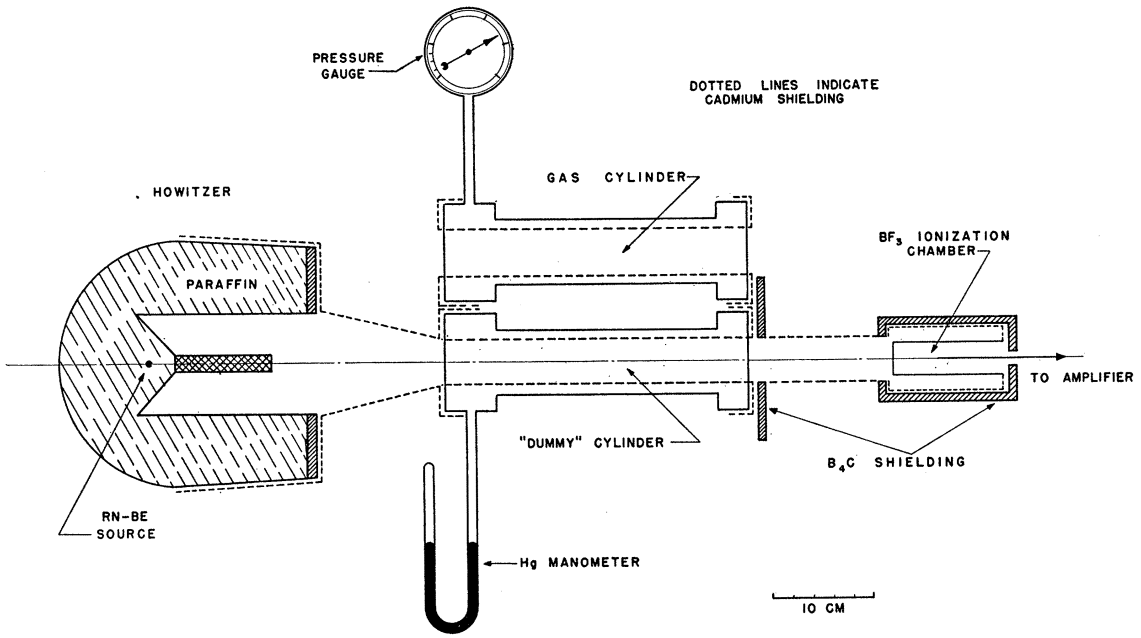

FIG. 1. General schematic arrangement of apparatus for gaseous measurements.

may accompany changes of structure. The use of gaseous substances of low molecular weight, however, offers a further complication due to the higher thermal velocities of the scattering nuclei. This, in the case of light molecules, will necessitate a small correction downward of the experimentally measured cross sections. (See Discussion of results, Section C.) The series investigated was gaseous hydrogen (H_2), methane (CH_4), ethane (C_2H_6), propane (C_3H_8), and butane (C_4H_{10}).

In addition, the cross section per proton for water and solid paraffin in two forms, one of "ordinary" paraffin, which is a mixture of hydrocarbon molecules, whose average molecule was assumed to be $C_{22}H_{46}$ and the other, a pure, single hydrocarbon, normal dotriacontane, ($C_{32}H_{66}$),[9] were measured.

Following the study of the behavior of the proton cross section, measurements were made of the cross section of the neighboring light nuclei, the deuteron, and the alpha-particle. Measurements on deuterium and helium are particularly useful, since the results are related to single isotopes. From the theoretical standpoint, the neutron-proton interaction requires the solution of a two-body problem which is capable of exact mathematical treatment. Beyond this, however, the neutron interaction with more complex particles necessitates the solution of a three-body problem. The mathematical difficulties arising in the general three-body problem are insuperable, though attempts have been made in the special case of the neutron-deuteron problem,[10] which represents a modified three-body problem. Furthermore, according to the generally accepted ideas of nuclear forces and application of the Pauli exclusion principle, the α-particle is to be considered a saturated system, and consequently, its neutron nuclear cross section is of particular interest. The deuteron cross section was measured with deuterium oxide (D_2O) of high purity, and the helium cross section was measured with helium gas as a scatterer.

Previously, the neutron nuclear cross sections of a large number of the elements of the periodic table have been measured,[11,12] but none of these

[9] The sample of normal dotriacontane was kindly supplied by Dr. F. D. Rossini of the National Bureau of Standards.

[10] L. Motz and J. Schwinger, Phys. Rev. **58**, 26 (1940).

[11] Dunning, Pegram, Fink and Mitchell, Phys. Rev. **48**, 265 (1935).

[12] M. Goldhaber and G. H. Briggs, Proc. Roy. Soc. **A162**, 127 (1937).

surveys has included any of the inert gas cross sections, as the substances used in previous experiments were in either the solid or liquid state. In these experiments, neutron measurements have been extended to include the cross sections of neon, argon and krypton in the gaseous form.

The cross sections of oxygen and nitrogen have been previously measured with compounds of these substances in solid form, or the elements

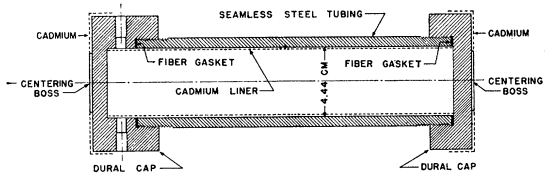

FIG. 2. High pressure gas cylinder details.

in liquid form. Interference effects[7,8] mentioned above made it desirable to redetermine these cross sections for the pure elements, and measurements were made on molecular oxygen and nitrogen in the gaseous form.

APPARATUS

The general experimental arrangement is shown in Fig. 1.

A. Neutron source and detector

The neutron sources used throughout all experiments were of the radon-beryllium type commonly used in this laboratory, whose maximum strength was about 650 millicuries.[13] To obtain slow neutrons, these bulbs were placed in a paraffin "howitzer."[14] Collimation was greatly improved and room scattering into the detector was greatly reduced by the use of hollow metal plaques and channels one cm thick, filled with powdered B_4C.[15] This supplemented the use of sheet cadmium shielding.

In order to approximate as nearly as possible the ideal arrangement which calls for a parallel beam of neutrons, a special ionization chamber was designed and constructed which offered a shielded opening to the neutron beam of only 3 cm diameter. The length of the chamber was 10 cm and its operating pressure of 2 atmospheres of boron trifluoride insured absorption of an appreciable fraction of the transmitted neutrons. Since the distance from source to scatterer was 40 cm and from scatterer to ionization chamber 35 cm, the solid angle subtended at the scatterer by the chamber was only 0.006 steradian.

The ionization chamber was operated from a stabilized power supply at 2000 volts to insure rapid ion collection. The ionization pulses were recorded by a linear amplifier and scale-of-two thyratron system operating a Cenco impulse counter.[16]

B. Gas measurement system

The gas cylinder finally adopted (see Fig. 2) consisted of a seamless steel tube 29 cm long threaded at both ends and closed with flat Duralumin caps 7.00 mm thick. Fiber gaskets and Apiezon Q in the cap threads insured a gas tight seal at pressures running up to 3000 lb. per sq. in. Good collimation was obtained by the use of cadmium diaphragms and B_4C plaques whose axes coincided with the cylinder axis.

In order to correct for neutron attenuation by the cylinder end caps, the experimental arrangement consisted of duplicate cylinders, one of which was evacuated and served as a "dummy." The other gas cylinder was provided either with a two-meter open tube mercury manometer, or a series of calibrated pressure gauges.[17]

During a cycle of measurements, it was

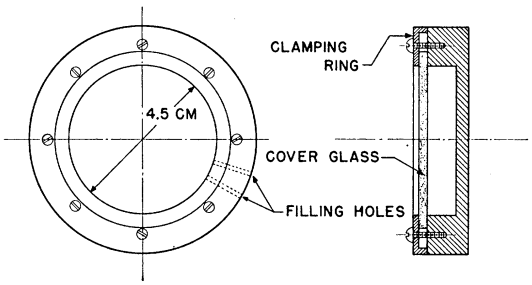

FIG. 3. Liquid cell details.

[13] The radon was obtained through the cooperation of Dr. G. Failla and Dr. J. J. Duffy of the Memorial Hospital, New York.
[14] G. A. Fink, Phys. Rev. **50**, 738 (1936).
[15] The boron carbide was supplied by the Norton Company of Worcester, Massachusetts.
[16] J. R. Dunning, Rev. Sci. Inst. **5**, 387 (1934).
[17] Kindly loaned by Professor C. F. Kayan of the Department of Mechanical Engineering.

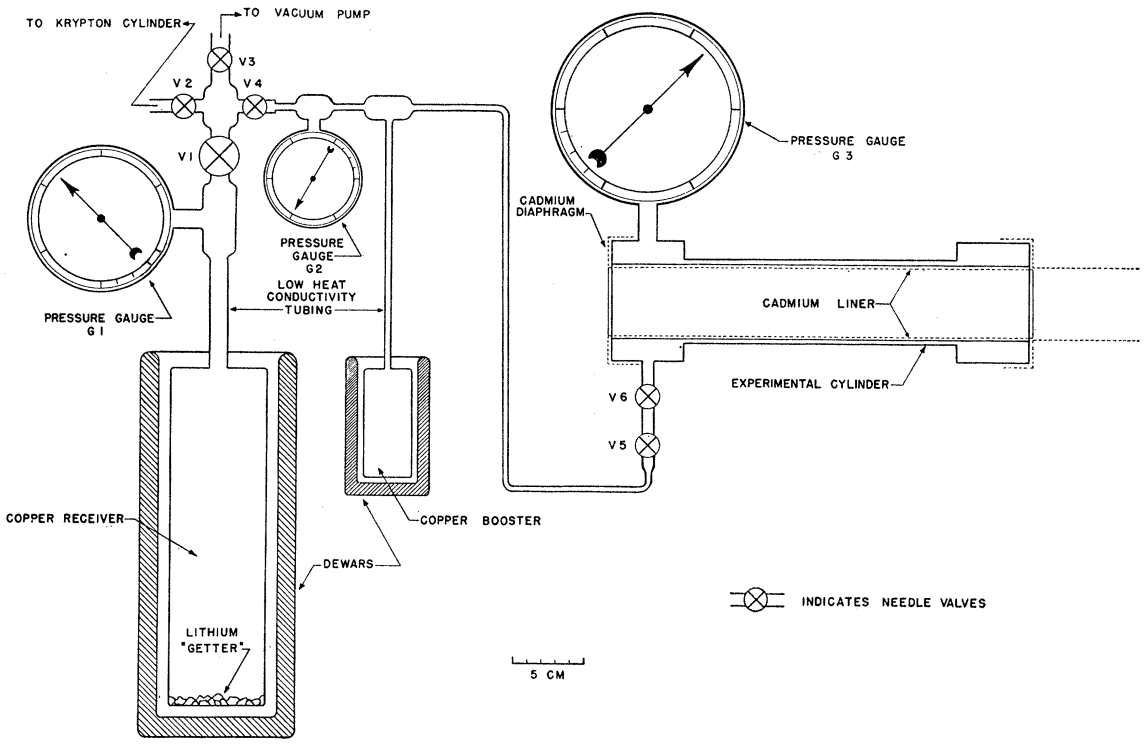

FIG. 4. Schematic arrangement of receiver and booster for high pressure gas transfer.

necessary to interpose the two cylinders alternately in the beam. To insure accurate register of each cylinder axis with the axis of the system, a double cradle and slide was designed consisting of two brass plates with double V notches in which the cylinders were supported. The slide plates moved in lubricated tracks provided with stops on either side. All gas connections were made with copper tubing to provide necessary flexibility, to withstand high pressure and to eliminate excessive absorption of hydrocarbons observed in rubber tubing and waxes.

In order to minimize scattering of neutrons from the room and adjacent apparatus, the entire assembly of Fig. 1, with the exception of the ionization chamber, was mounted in the center of the room on an open pipe-work frame, rigidly braced.

C. Cell design

The cross section measurements of the proton in water and paraffin necessitated the use of accurately constructed cells of uniform bottom thickness. The cell design is shown in Fig. 3. The bottoms were hollow dural disks and the caps were selected Eastman thin slide cover glasses of low boron content. The tolerance on the thickness of both cell bottoms, cover glasses, and a "dummy" plate was held to less than 0.001 cm. Several cells were made to hold suitable thicknesses of absorber, which gave neutron transmissions ranging from 35 percent to 65 percent.

The paraffin results were obtained from hollow dural cells similar to the above, without the cover glasses. The paraffin plaques were cast in these cells by melting the paraffin in a water bath just above the melting point. To investigate the possibility of air trapped during cooling in any way vitiating the results, some of the plaques were cast and allowed to cool under a bell jar at a pressure of about 5 or 6 mm of mercury. No transmission difference was discernible in these casts over paraffin plaques subjected to normal air cooling.

D. High pressure transfer systems

Since many of the gases had small cross sections, it was necessary to use filling pressures which in some cases exceeded 2500 lb./in.^{2}. None

of the gases was supplied at this pressure, which made it necessary to design a system for raising the original pressure to the appropriate value. Actually, two "booster" systems were used, one exclusively for krypton, in order to minimize loss of this valuable gas, and the other for oxygen, helium, etc. The krypton transfer system is shown schematically in Fig. 4. The method of transfer was to condense the krypton, by means of liquid nitrogen, into an intermediate

TABLE I. *Neutron-proton measurements for gaseous hydrogen*

P	$\log_{10} P^{-1}$	Pressure lb./sq. in. gage	g/cm²	Cross Section per Proton $\times 10^{24}$ cm^{-2}
0.726	0.86094	77.0	0.01512	31.9
0.668	0.82478	114.0	0.0211	31.5
0.656	0.81690	119.5	0.0240	31.1
0.502	0.70070	205.0	0.0363	31.4
0.413	0.61595	277.0	0.0484	31.1
0.319	0.50379	375.0	0.0645	30.8

evacuated cylinder of small volume and allow the system to warm up to room temperature. Transfer back to the main reservoir was accomplished in the same manner.

In order to maintain high purity of the krypton, a glass capsule, packed with metallic lithium particles, was shattered inside the receiver and used as a "getter" to clean up any residual contaminating gases. When the krypton was transferred to the "booster," the receiver could be heated and pumped to remove any gases absorbed by the lithium.

A similar procedure was employed with other gases.

Measurements

The cross sections were calculated from the relation

$$P = e^{-n\sigma x},$$

where n = number of nuclei per cubic centimeter of the scatterer, σ = the total cross section of the scatterer, x = thickness of scatterer, P = percent of neutrons transmitted by the scatterer.

In order to calculate the percent transmission, P, four measurements were made in equal time intervals.

1. N_1, the number of neutrons transmitted through the scatterer.

2. N_2, the number of neutrons transmitted through both scatterer and a sheet of cadmium of 0.8 g/cm² placed near the source.

3. N_3, the total number of neutrons traversing the open beam, i.e., with the dummy cell in place.

4. N_4, the number of neutrons transmitted through the sheet of cadmium and the dummy cell.

Then, the percent transmission is given by

$$P = (N_1 - N_2)/(N_3 - N_4).$$

Cyclic runs were taken in all cases. The statistical precision given for the results is based on $1/\sqrt{N}$, where N is the number of neutrons counted. During any experiment, not less than 40,000 neutrons were counted for N_1 and N_3. All absolute cross sections may be considered reliable to 3 percent after allowing for other sources of variation, but the relative values are more precise.

Table I shows the measurements made for a number of hydrogen gas pressures. These served as a check on the cross section value and also as a test of the exponential transmission of the system.

The cross section measurements of the proton in the hydrocarbon series are given in Table II. Included in the table for comparison is the cross section for liquid hydrogen[18] (normal).

TABLE II. *Interaction of slow neutrons with bound protons.*

Substance	State	g/cm²	Percent Transmission	Cross Section per Proton $\times 10^{24}$ cm^{-2}
Hydrogen (H_2) Normal	Liquid*			24.0
Water (H_2O)	Liquid	0.1778	57.2	44.6±0.5
Hydrogen (H_2)	Gas	0.0363	50.2	31.8±0.5
Methane (CH_4)	Gas	0.0683	61.8	45.4±0.3
Ethane (C_2H_6)	Gas	0.1059	54.6	46.4±0.5
Propane (C_3H_8)	Gas	0.1177	53.3	46.9±0.6
Butane (C_4H_{10})	Gas	0.1242	51.2	48.7±0.6
Paraffin ($\sim C_{22}H_{46}$)	Solid	0.1304	54.8	49.8±0.2
Dotriacontane ($nC_{32}H_{66}$)	Solid	0.1368	52.8	50.2±0.2

* Reference 18 in text.

TABLE III. *Interaction of slow neutrons with the nuclei of gases.*

Substance	State	Pressure lb./sq. in.	g/cm²	Percent Transmission	Cross Section per Nucleus $\times 10^{24}$ cm^{-2}
D_2O	Liquid		0.737	70.3	5.73±0.14
Helium	Gas	1980	0.758	83.8	1.51±0.09
Neon	Gas	1600	2.620	80.4	2.79±0.14
Argon	Gas	1600	5.220	92.1	2.53±0.10
Krypton	Gas	465	3.145	53.6	27.3 ±0.5
Nitrogen	Gas	495	1.173	54.4	12.2 ±0.2
Nitrogen*	Liquid				12.8
Oxygen	Gas	1500	3.94	54.0	4.12±0.10
Oxygen*	Liquid				4.05

* Reference 18 in text.

[18] Brickwedde, Dunning, Hoge and Manley, Phys. Rev. **54**, 266 (1938).

The value listed for each substance represents the average of several sets of measurements.

Listed in Table III are the cross sections for deuterium, the rare gases, and nitrogen and oxygen in gaseous form.

The cross sections of nitrogen and oxygen in the liquid state[18] are also included. A comparison of the cross sections of the gaseous and liquid forms shows that the cross section did not change appreciably in going from the gaseous to the liquid state.

Discussion of Results

A. Hydrogen and the hydrocarbon series

The experimental value (31.8×10^{-24} cm^2), Fig. 5, for the proton cross section as obtained from hydrogen gas will be decreased when correction is made for the thermal motion of the molecules. (See below, Section C.) The influence of chemical binding on the cross section may be noted from the increase in cross section per proton for increasing mass of the molecule of the scattering system as shown in Table II. In this series, the cross section of carbon was taken[7,12] to be 4.80×10^{-24} cm^2. The proton cross section was calculated after subtracting the carbon cross section from the cross section of the whole molecule.

The increase of the proton cross section for C neutrons with increase in the square of the reduced mass is more clearly evident in the lower curve of Fig. 6. Arley's treatment of this scattering problem predicts that for zero energy neutrons the proton cross section should vary linearly as the square of the reduced mass. This is shown by the upper curve of Fig. 6, which includes the value for the free proton cross section obtained experimentally by Cohen, Goldsmith and Schwinger[19] using rhodium filtered neutrons and a rhodium detector with a G-M counter and also as determined more precisely by Hanstein[20] for indium and iodine resonance neutrons. Hanstein's value is 21 ± 1 $\times10^{-24}$ cm^2.

From the upper curve, the bound proton cross section would be 84×10^{-24} cm^2 since the squares of the reduced masses of the free and bound proton are in the ratio of one to four. The experimental value for the bound proton cross section (for paraffin) is actually 50×10^{-24} cm^2 for thermal neutrons at 300°K. It can be seen from the graph that while the proton cross section for hydrogen gas shows only a small departure from linearity, which would be slightly larger if corrected (see below, Section C), the departure becomes progressively larger for the heavier molecules. Previous experiments (unpublished) with neutrons emerging from paraffin at ~115°K show that from 300°K the proton cross section increases by a factor of 1.3 to an absolute value of 65×10^{-24} cm^2 which approaches the limiting value for zero energy neutrons.

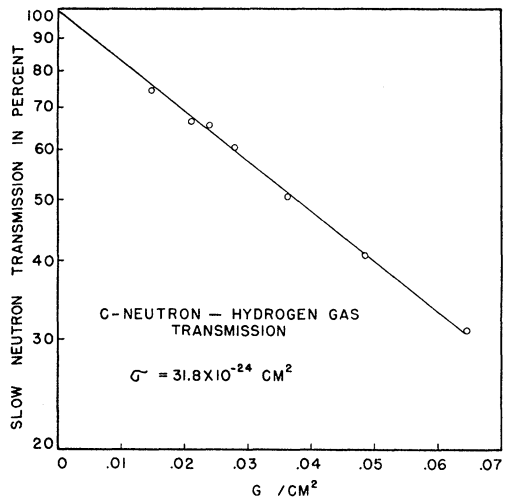

FIG. 5. Above is shown a semi-logarithmic graph of the percent neutron transmission plotted against the grams per square centimeter for a series of hydrogen pressures. As can be seen from the linearity of the plot, the transmission follows quite well an exponential law. The average cross section is $31.8\pm0.5\times10^{-24}$ cm^2.

Results of subsequent measurements on liquid and solid cetane show that the variation of the cross section may in part be due to interference effects. The existence of these effects cannot be disregarded in a critical survey of the relative change in the measured cross sections.

Owing to its complexity, the general problem of neutron-proton scattering has not been solved theoretically. Attempts have been made based on certain simplifying assumptions[4] and their modifications.

1. The original assumption of interaction

[19] V. W. Cohen, H. H. Goldsmith and J. Schwinger, Phys. Rev. **55**, 106 (1939).
[20] H. B. Hanstein, Phys. Rev. **59**, 489 (1941).

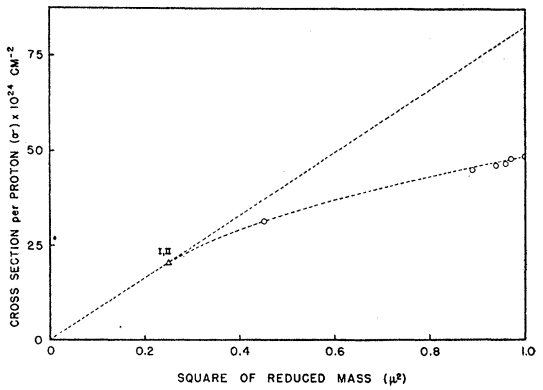

FIG. 6. C neutron cross section per proton for hydrocarbon series. *I*—Cohen, Goldsmith and Schwinger, Phys. Rev. **55**, 106 (1939); *II*—Hanstein, Phys. Rev. **59**, 489 (1941).

between a neutron and bound proton was modified[4] to that of interaction between a neutron and a system of proton "molecules," this system being chosen to have a mass of fourteen times the neutron mass. The system was admittedly artificial in that interference effects were neglected and interactions were assumed to take place by energy transfer to six vibrations of the group composing the molecules with neglect of the rotations of the group.

The actual interaction which takes place between neutron and the paraffin will depend upon the binding between the molecules and it is difficult to determine the aggregate.

2. The original assumption made was that a beam of zero energy neutrons was used.[1] This was modified by Arley to a beam of neutrons whose energy distribution was strictly Maxwellian. While it has been shown by the method of a mechanical velocity selector[5] that the maximum of the energy distribution of C neutrons at room temperature corresponds to an energy of the order of kT, the actual distribution is probably only approximately Maxwellian.

For the various molecules studied in the gaseous state, the theoretical neutron-proton cross section would have to be studied in more detailed fashion as a function of the energy distribution of the neutrons and energy levels effective in the interaction before more accurate agreement between theory and experiment could reasonably be expected.

From the graph, the value of the "chemical factor" for paraffin can be calculated from the ratio of the free and bound proton cross sections. The ratio for C neutrons is 2.4. Arley has calculated this ratio for his paraffin model and found the value to be 2.7.

B. Deuteron cross section

A theoretical calculation of the deuteron cross section has been made by Motz and Schwinger.[10] Their results arrived at by numerical integration of the wave equation for two choices of potential give cross sections of 4.57×10^{-24} cm² and 6.91×10^{-24} cm² as against the value given in Table III of 5.7×10^{-24} cm². In their calculations polarization effects were neglected because of the great difficulties introduced into the calculation.

C. Rare gas series

The small experimental cross section (1.51×10^{-24} cm²) for helium (see Table III) is consistent with the assumption that the α-particle is a saturated system. In calculating this value, the thermal motion of the gas molecules was neglected. Due to their motion, the cross section is increased and a correction should be applied which will give the "effective" cross section for stationary molecules. Schwinger[21] has calculated this effective cross section by using the relative velocity of neutron and scattering molecule, and the molecular velocity distribution function which represents the Maxwellian distribution appropriate to the temperature of the gas, and arrives at the value of 1.25×10^{-24} cm². This correction should become less important as the mass of the molecule increases as was the case for the hydrocarbon series.

Argon[2] and neon[2] have three isotopes each and krypton[2] has six. While no definite inference can be drawn from the experimental cross section of these gases, it is quite possible that in the case of krypton the contribution to the cross section may well be due to one of those which has a large capture cross section for neutrons of the energy used.

D. Nitrogen and oxygen

The cross sections for these gases agree well with the measurements made in the liquid[18]

[21] J. Schwinger, Phys. Rev. **58**, 1004 (1940).

state. However, the measured value of 4.1×10^{-24} cm^2 for gaseous oxygen is significantly higher than the value of 3.3×10^{-24} cm^2 as obtained[7,11] from SiO_2, assuming additivity of cross sections. Effects similar to this have been previously observed[7] and interpreted in terms of interference phenomena.

Dependence of Neutron Scattering on Structure

The results of the measurements on highly purified cetane are given in Table IV.

The melting point of pure cetane is 16.1°C. As this is very close to room temperature, a slight lowering of the temperature of a cell of liquid cetane caused the cetane to solidify. An ice water bath enabled alternate measurements to be made of the slow neutron transmission in both liquid and solid states.

The 10 percent difference in the measured values of the proton cross section can be explained in terms of interference effects since the solid was more transparent than the liquid. A corresponding effect was not observed in the case of lead and bismuth.[22] In certain respects the conditions were more favorable in the case of cetane since (a) the spacing of the interference centers is about 1.5A whereas for bismuth and lead, it is about 5.5A. For the neutron distribution, the most probable wave-length is about 1.7A and only very small interference effects could be expected from bismuth and lead; (b) on solidification, large well-defined crystals of cetane were formed.

The amount of coherent scattering which must be present to account for the observed difference in cross section between solid and liquid cetane in terms of interference effects is fairly large. As pointed out by Schwinger, some discrepancies still exist between the experimental results[18,23] and the theory[21,24] of the scattering of neutrons by ortho- and parahydrogen. Furthermore the theory[25] must be considerably extended if it is to take into account all of the possible contributions to interference effects such as the net effect of the ortho- and para-states in the case of an aggregate of paraffin molecules in a crystalline structure.

The cetane results clearly suggest, however, that the cancellation of the scattering by the singlet and triplet states is not complete so that some coherent scattering results. A further contribution to the coherent scattering is also possible from the carbon atoms. It appears, therefore, that although the inelastic scattering is comparatively large in paraffin, the experiments show that a considerable amount of coherent scattering still exists and that structure does play an important role in neutron scattering even in organic molecules.

Table IV. *Slow neutron-cetane cross section.*

Substance	State	G/cm^2	Percent Transmission	Cross Section per Proton $\times10^{24}$ cm^{-2}
Cetane ($nC_{16}H_{34}$)	Solid	0.1721	47.7	44.9 ± 0.4
Cetane ($nC_{16}H_{34}$)	Liquid	0.1573	47.1	50.1 ± 0.4

It is a pleasure to acknowledge my indebtedness to Professor J. R. Dunning both for suggesting this problem and also for many helpful suggestions and encouragement during the course of this research. Thanks are also due to Dr. F. J. Metzger and Dr. F. R. Balcar for their cooperation in supplying some of the rare gases.

[22] F. Rasetti, Phys. Rev. **58**, 321 (1940).

[23] L. W. Alvarez and K. S. Pitzer, Phys. Rev. **58**, 1003 (1940).

[24] J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937). M. Hammermesh and J. Schwinger, Phys. Rev. **55**, 697 (1939). R. G. Sachs and E. Teller, Phys. Rev. **57**, 1076 (1940). W. Rarita and J. Schwinger, Phys. Rev. **59**, 556 (1941).

[25] G. C. Wick, Physik. Zeits. **38**, 403 (1937); **38**, 689 (1937). O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939). J. H. Van Vleck, Phys. Rev. **55**, 924 (1939).