Slow Neutron Velocity Spectrometer Studies of O_2, N_2, A, H_2, H_2O, and Seven Hydrocarbons

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The slow neutron cross sections of free atoms of O, N, and A have been determined to be; O: 3.73, N: 9.96, A: 0.68 barn. Between 1 and 15 ev the N_2 curve shows an $E^{-\frac{1}{2}}$ slope of 0.34 barn$\times$(ev)$^{\frac{1}{2}}$ per atom. Measurements on these elements have been extended to over 10,000 ev with no resonances observed. Dependence of cross section upon neutron energy from 1 ev to 0.003 ev is presented for O_2, N_2, H_2, H_2O, methane, ethane, propane, *n*-butane, cetane, ethylene, 1,3-butadiene, and liquid *n*-butane. At the lowest energies, the large observed H_2 cross section is attributed largely to thermal translational motion of the molecule. The measurements on the hydrocarbons show the effect of the binding of H in various molecules. Comparison with the theories of Bethe and Arley is made. No difference has been detected in cross-section measurements on gaseous and liquid *n*-butane.

INTRODUCTION

THE Columbia University slow neutron velocity spectrometer has been used in numerous studies of the interaction of neutrons with matter.[1,2] The results obtained are in the form of cross section (or transmission) as a function of neutron energy. The previous reports have dealt with the study of materials in the solid or liquid state. The results of application of neutron spectroscopy to samples in the gas phase are presented here. Measurements on two liquid samples have been included for comparison.

For the elements studied the only purely nuclear phenomenon observed at low energies (<100 ev), aside from nuclear scattering, is the $1/v$ slope which is observed in the case of N and A and is attributed to neutron capture. This is indicative of resonances outside of the energy interval investigated. Capture of neutrons by O is known to be unobservably small for transmission measurements in this energy region. The capture cross section of H is small (being about 0.3 barn at 0.025 ev) and the expected $1/v$ slope is easily masked by other effects.

The non-nuclear effects observed below 1 ev are of molecular origin. One of these is chemical binding, the most important case of which is the fourfold increase of the H cross section when bound to a very heavy molecule. In addition, interference effects, inelastic scattering, and effects arising from the molecular thermal motion are observed. Cross-section measurements at low neutron energies (to 0.003 ev) for numerous materials are presented and demonstrate these phenomena, although it is rarely possible to make an experimental separation of the contributions of the various effects. In addition to these the O_2 cross section should have a contribution arising from the paramagnetic scattering of neutrons.

EXPERIMENTAL PROCEDURE

The slow neutron velocity spectrometer, the standard filter technique for making absolute transmission measurements, and the sample holders for liquid and gaseous scatterers have been described in detail in the preceding paper.[3] Where the standard filter has been used, it will be specifically stated. Otherwise, the effect of using the standard filter has been estimated and corrections made accordingly. In no case has the cross section been increased by more than 3 percent for this correction.

Transmission Measurements in the Low Energy Region

Some difficulty has been experienced in making measurements at neutron energies below 0.005 ev. These difficulties usually lead to too high a measured value for the transmission. Since all of the materials under investigation here show decreasing transmission with decreasing neutron energy, and since the intensity at low energies is very small compared with that at the thermal peak ($\sim$0.025 ev) the faster thermal neutrons might somehow influence the low energy results. Specifically, one or more of the following effects may give rise to erroneous results:

(1) Thermal neutrons inside the cyclotron enclosure may be scattered into the beam by air molecules and arrive at the detector at the same time as the slower neutrons under consideration. The possibility of this occurring has been minimized by replacing the 3.5-meter air path of the neutron beam inside the enclosure by a cadmium-shielded argon path, accomplished by the use of argon-filled, cadmium-lined, aluminum ducts. Argon is used because of its very low scattering cross section compared with air and other gases. This gives the additional benefit of diminishing the attenuation of

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[1] L. J. Rainwater and W. W. Havens, Phys. Rev. **70**, 136 (1946). W. W. Havens and L. J. Rainwater, Phys. Rev. **70**, 154 (1946). Rainwater, Havens, Wu, and Dunning, Phys. Rev. **71**, 65 (1947). Havens, Wu, Rainwater, and Meaker, Phys. Rev. **71**, 165 (1947). Wu, Rainwater, and Havens, Phys. Rev. **71**, 174 (1947). Havens, Rainwater, Wu, and Dunning, Phys. Rev. **73**, 963 (1948).

[2] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948).

[3] E. Melkonian, Phys. Rev. **76**, 1744 (1949).

the neutron beam in the air path, the gain in intensity being about a factor of two at 0.003 ev.

(2) For condensed samples like H_2O it is possible for thermal neutrons to be slowed down by inelastic collisions in the scatterer and reach the detector at times corresponding to the slower neutrons. Some simple tests to establish the presence of such an effect have indicated that it is not important, but it could not be ruled out conclusively. However, Jones has given some evidence for the presence of such an effect in his measurements on water.[4]

(3) Sutton *et al*,[5] state that there is a continuous production of neutrons even during those portions of the cycle when the arc is not on. This effect was detected in connection with the present work by measuring the intensity distribution at very long timing by using a repeat time of 32,768 microseconds. The normal intensity should decrease very rapidly with increasing time of flight approximately as (time of flight)$^{-4}$. The observed curve showed this expected decrease to about 16,000 microseconds. Beyond this, the intensity was independent of timing and was attributed to the background production of neutrons. Measurement of the transmission of boron for these background neutrons showed that the mean energy is approximately kT. The intensity of this background varies somewhat with operating conditions but is usually sufficiently intense to distort measurements for neutron energies less than 0.005 ev.

All of these effects are aggravated by the adverse intensity ratio of the main part of the thermal distribution to the lower energy neutrons. All of them can be minimized by the use of Be and BeO filters which discriminate strongly against neutrons of energy greater than 0.01 ev but which become almost completely transparent at lower energies. The use of these filters should reduce the effects discussed above to a negligible amount. All of the results presented here for times of flight greater than 665 microseconds/meter were taken with 13 g/cm^2 of sintered Be in the beam. In a few cases, which will be specifically pointed out, 5.7 g/cm^2 of sintered BeO were used in addition to the Be. This addition of BeO may or may not give better results depending upon the relative amount of background. In any case, the improvement arising from the use of BeO is generally relatively small for measurements to 0.003 ev.

Preparation and Purity of Samples

The samples were obtained from various sources, and the purification required depended upon the amount and nature of the impurities.

O_2: This was obtained from the Matheson Company who stated that there was present as impurity 0.5 percent A and a small amount of N_2. These will not affect the measurements appreciably. To get rid of possible traces of H_2 and H_2O which would have a large effect, the O_2 was dried roughly with $CaCl_2$, passed over platinized asbestos at 300°C, dried with P_2O_5 and passed through a trap immersed in dry ice slush for final drying.

N_2: This was prepurified N_2 from Airco who claimed a purity of 99.999 percent. Mass-spectrographic analyses by the Columbia University Analytical Laboratory and by the Bureau of Standards indicated the presence of 0.2 percent A and 0.1 percent CO_2. In either case the gas was sufficiently pure and was used after passing through a dry ice trap to minimize the water vapor content.

H_2: This was electrolytic H_2 from Airco, who claimed an O_2 impurity less than $\frac{1}{2}$ percent (on a molar basis). Since the "sample thickness" determination is made by weighing (as will be discussed later) this amounts to a large error because of the large ratio of molecular weights. The purification process (to get rid of O_2 here) was essentially the same as that for the purification of O_2. A determination of the molecular weight of the purified sample was made as a check on the presence of heavier gases. The results showed a negligible amount of such impurities. In addition, mass-spectrographic analysis indicated no impurities. The sensitivity for detection of O_2 was such as to show 0.005 percent.

A: This was from Airco who listed the presence of 0.2 percent N_2. However, mass-spectrographic analysis indicated the presence of less than 0.05 percent of impurity (probably N_2). Because of the very large cross section of N_2 compared with that of A, the presence of even a small amount of N_2 is exaggerated. However, removal of the N_2 is very difficult for the amount of sample required (~500 liters at S.T.P.) and therefore the gas was used as furnished.

Methane, ethane, propane, *n*-butane, ethylene, and 1,3-butadiene: These were research grade gases obtained from Phillips Petroleum Company. The stated purities are (see Table I):

Water: Distilled water was used.

Cetane: ($C_{16}H_{34}$) This was obtained from du Pont with a claim of 95 percent purity, the impurities presumably being saturated hydrocarbons having a slightly different number of C atoms (e.g., $C_{15}H_{32}$, $C_{17}H_{36}$, etc.). Attempts at distillation using a packed column appeared to give no improvement in purity as determined by measuring the freezing point. Analysis of the hydrogen atom to carbon atom ratio gave 2.131 compared with 2.125 calculated from the formula.

TABLE I.

Gas	Mol. percent	Probable impurities
Methane (CH_4)	99.59	N_2, ethane, CO_2
Ethane (C_2H_6)	99.69±0.00	Propylene, propane
Propane (C_3H_8)	99.99+	None detected
n-butane (C_4H_{10})	99.78±0.08	Iso-butane
Ethylene (C_2H_4)	99.77±0.01	Propane, CO_2
1, 3-butadiene (C_4H_6)	99.72±0.06	Butene-1, butene-2

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[4] W. B. Jones, Jr., Phys. Rev. 74, 364 (1948).

[5] Sutton, Hall, Anderson, Bridge, DeWire, Lavatelli, Long, Snyder, and Williams, Phys. Rev. 72, 1147 (1947).

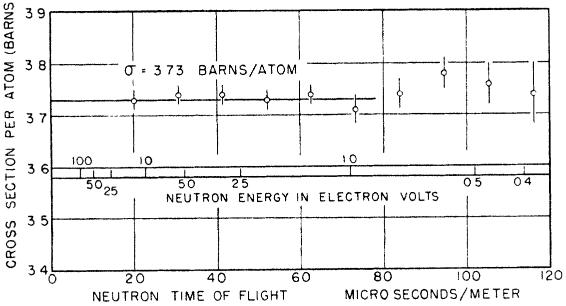

FIG. 1. The slow neutron cross section of O_2 gas in the energy interval 0.4 to 15 ev. The sample contained 6.853 g/cm². A standard filter was used.

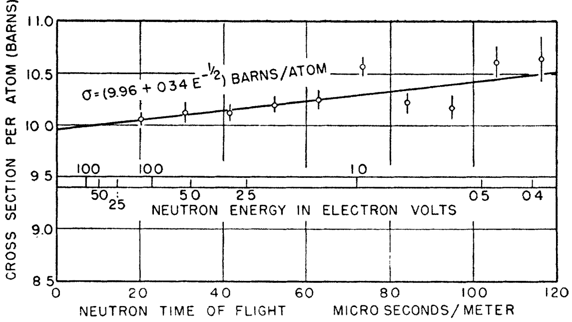

FIG. 2. The slow neutron cross section of N_2 gas in the energy interval 0.4 to 15 ev. The sample contained 2.312 g/cm². A standard filter was used.

Determination of "Sample Thickness"

The important quantity for cross-section calculation characteristic of the samples used is the number of molecules per cm² of area normal to the beam. Since this is most directly associated with the number of g/cm², weighing techniques were preferred over measurement of pressure, thus doing away with the necessity of knowing the exact gas laws. Also, pressure measurements are difficult to make accurately.

For the gas samples containing total weight of sample greater than 100 g (as for O_2, N_2, A), the weight of gas was determined directly by weighing the holder, before and after filling, on a large balance capable of handling 10 kg. Since the cross-sectional area of the holder varied somewhat along the length, the effective area was determined from the volume, measured by weighing water, and the known inside length.

When the total amount of gas sample was appreciably less than 100 g, as was the case for all of the gases containing hydrogen, the above determination was not sufficiently sensitive, and an alternative scheme was used. A small aluminum tank, of known volume and having dimensions and weight suitable for weighing on an ordinary analytical balance, was connected with the gas sample holder and filled with the sample gas at the same time. This allows determination of the density of the gas sample from which g/cm² can be computed using the known inside length of the sample holder.

SLOW NEUTRON CROSS SECTION OF FREE O AND N ATOMS

For the measurements above 0.4 ev neutron energy the cyclotron and detector on times were 64 microseconds. The standard filter technique was used since absolute cross-section values were sought. The results are shown in Figs. 1 and 2.

The molecular effects, which gave rise to the observed $1/E$ slope in the case of H_2, vary inversely as the atomic weight and are undetectable, unless great precision is attained, in the case of O_2 and N_2 as the change in cross section is only a few tenths of a percent. Hence, the usual time of flight abscissa is used on the graphs.

O_2 shows no other effect in this energy region so that the cross section is independent of energy. Using the data above 1 ev, the free oxygen cross section is found to be 3.73±0.04 barns per atom. This supercedes the previously reported value of 3.68.[6,7]

N_2 exhibits neutron capture in the form of a $1/v$ slope. The reactions possible are $N_7^{14}(n, p)C_6^{14}$ and $N_7^{14}(n, \gamma)N_7^{15}$. Nitrogen is one of the very few elements which decay by heavy particle emission following slow neutron capture. For neutron energy above 1 ev the nitrogen cross section is best fitted by:

$$\sigma_N = (9.96 \pm 0.10 + 0.34E^{-\frac{1}{2}}) \text{ barns/atom.}$$

The free nitrogen cross section is thus 9.96 barns/atom (replacing the previously reported value of 9.74). The energy-dependent term should be a good measure of the capture cross section as no other significant effects are present in this energy region.

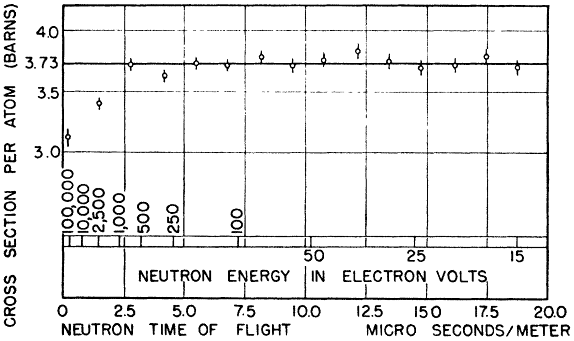

FIG. 3. The slow neutron cross section of O_2 gas above 15 ev using an 8.595-g/cm² sample.

CROSS SECTION OF O, N, AND A FOR NEUTRON ENERGIES GREATER THAN 15 EV

Above 15-ev molecular effects are negligible, and only the nuclear cross sections of the elements present are important. Figures 3–5 show measurements on the elements O, N, and A, respectively, using cyclotron and detector on times of 4 and 8 microseconds. Standard

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[6] E. Melkonian, Phys. Rev. 73, 1265A (1948).

[7] Melkonian, Rainwater, Havens, and Dunning, Phys. Rev. 73, 1399 (1948).

filters are not available for this energy region, and the measured cross section values have been increased by 1 to 2 percent to account for the dependence of detector efficiency upon counting rate. In the case of O and N, the constant portions of the curves were adjusted to the free atom values determined previously. Since the free A cross section has not been determined with the aid of standard filters, we may obtain a fairly good value for it from the constant portion of Fig. 5 and get 0.68 barn/atom.

The measured values for the free atom cross sections of O, N, and A are listed in Table II. Expected potential scattering cross-section values calculated from $\sigma=4\pi R^2$ with $R=1.5\times10^{-13}$ $A^{\frac{1}{3}}$ are included for comparison.

Comparing with expected values, the nitrogen value is abnormally high, the argon value abnormally low, while the oxygen value is relatively in better agreement. Interpreting according to the theory of Feshbach, Peaslee, and Weisskopf[8] one would expect that nearby (relative to a very wide level spacing) energy levels are present. For nitrogen the energy level would be at a lower energy and hence would be virtual (i.e., at negative energy). Figure 4 shows that at higher neutron energies, the nitrogen cross section is dropping rapidly to a more normal value. For oxygen, the energy level is again virtual, and Fig. 3 shows a decrease in the cross section at higher energies. The continuation of these trends at still higher energies is shown in the graphs in the review article by Goldsmith, Ibser, and Feld.[9] The nitrogen cross section decreases continuously with increasing neutron energy, and reaches the expected potential scattering value of the cross section at about 1 Mev. The oxygen cross section decreases with increasing neutron energy to about 250 kev, but the potential scattering value of the cross section appears not to be reached. Instead, there are resonances at approximately 450, 900, and 4000 kev. (See, however, the more recent work of Adair *et al.*[10] where the oxygen cross section is found to be approximately constant at 3.8 barns between 20 and 350 kev.) For argon, the energy level must be at higher neutron energies, and Fig. 5 shows a rapid increase in cross section with increasing neutron energy.

TABLE II. Comparison of observed potential scattering values of the free atom cross sections.

Element	σ_{free}	$4\pi R^2$
N	9.96	1.64
O	3.73	1.80
A	0.68	3.31

CROSS SECTION RESULTS TO 0.003 EV NEUTRON ENERGY

Below 1 ev neutron energy, molecular effects are prominent. The following phenomena influence the cross section when a gaseous sample is used:

(1) *Thermal motion:* The thermal motion of the gas molecules gives rise to an increase in cross section with decreasing neutron energy. In the limiting case of low neutron velocity, this effect may be thought of as arising from a preponderance of cases in which neutrons are knocked out of the beam by molecules striking them from behind and from the sides over those cases where a neutron collides with a molecule directly in its path. In this limit of low neutron velocity, the cross section in the laboratory system varies inversely as the neutron velocity.

The above comments apply if the cross section is approximately constant in the relative coordinate system. They do not apply to a $1/v$ type of dependence which is unaffected by translational motion of the gas molecules.

(2) *Inelastic collisions:* When energetically possible, the neutron can be scattered while giving energy up to the molecule in the form of increased vibrational or rotational energy. Molecules in excited vibrational and

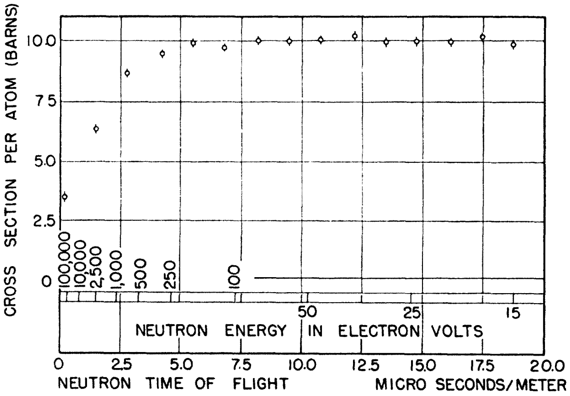

FIG. 4. The slow neutron cross section of N_2 gas above 15 ev using a 3.168-g/cm² sample.

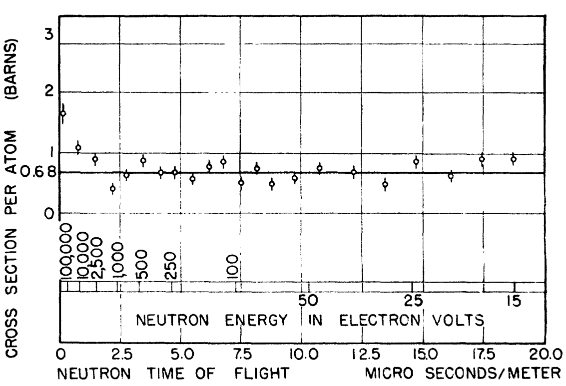

FIG. 5. The slow neutron cross section of A gas above 15 ev using an 11.71-g/cm² sample.

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[8] Feshbach, Peaslee, and Weisskopf, Phys. Rev. **71**, 145 (1947).

[9] Goldsmith, Ibser, and Feld, Rev. Mod. Phys. **19**, 259 (1947).

[10] Adair, Barschall, Bockelman, and Sala, Phys. Rev. **75**, 1124 (1949).

rotational states can also scatter neutrons by giving up energy at the expense of excitation energy (so-called hyperelastic collisions). Both of these give contributions to the cross section in addition to the elastic cross section.

(3) *Interference effects:* For neutron wave-length small compared with atomic spacing in the molecule, the total cross section of the molecule is simply the sum of the individual atomic cross sections. However, when the neutron wave-length becomes comparable with the interatomic distances, interference effects between the waves scattered from the various atoms become important. For a diatomic molecule consisting of identical atoms of zero spin, this interference is constructive and at very long wave-lengths leads to a cross section which is just twice the cross section of the molecule at very short wave-lengths. When nuclear spins are not zero and the scattering amplitudes for the various spin orientations are different, or when different atoms are present, the interference effects may increase or reduce the cross section, depending upon the degree of difference (and in particular a difference in sign) of the scattering amplitudes. In the limit of very slow neutrons (and molecules at rest and in the ground state) the cross section of a polyatomic molecule is given by:

$$\sigma=4\pi\left(\sum_n \frac{m}{m_n}a_n\right)^2,$$

where a_n = scattering amplitude of nth atom (free), m = reduced mass of neutron and entire molecule, m_n = reduced mass of neutron and nth atom. This expression takes into account interference effects and the effect of chemical binding of atoms in the molecule (reduced mass effect). Elastic collisions only are possible in this limiting case as long as the temperature is low enough so that a negligible number of molecules are in excited states.

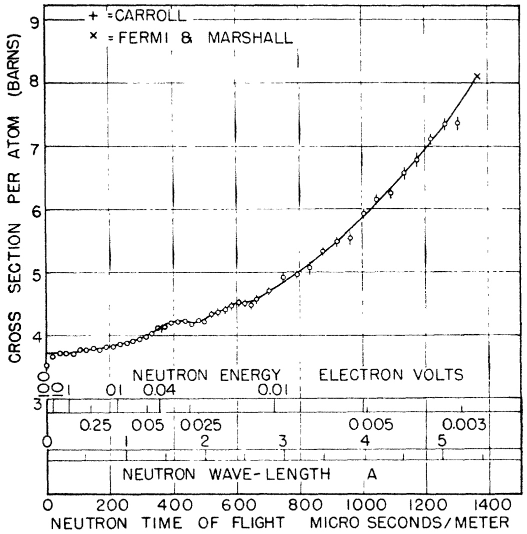

FIG. 6. The slow neutron cross section of O_2 gas to 0.003 ev. The average of several runs is shown, the samples varying from 6.7 to 8.6 g/cm². For timings greater than 665-microseconds/meter BeO filtering was used in addition to the usual Be filtering.

We now consider the results for the various materials investigated.

O_2: The results are shown in Fig. 6.

Oxygen has negligible capture in this energy region so that almost all of the increase in cross section below 1 ev is due to molecular effects. The oxygen nucleus has no spin and oxygen is predominantly of one isotope so that the interference effect tends to double the cross section at very low energies. The binding effect increases the cross section by six percent.

An additional contribution to the O_2 cross section is believed to be due to the paramagnetism of O_2 which gives rise to an interaction between the magnetic moment of the O_2 molecule and that of the neutron. At sufficiently low energies when the form factor for this effect approaches unity, calculations based on a formula given by Halpern and Johnson show that this interaction should contribute about five barns to the molecular cross section.[11, 12]

N_2: The results are presented in Fig. 7.

The cross section for neutron capture contributes an appreciable amount to the total nitrogen cross section in this energy interval. Using the $E^{-\frac{1}{2}}$ slope based on measurements above 1 ev, the capture cross sections per atom are 2.15 and 6.2 barns at 0.025 and 0.003 ev, respectively. The slope below 1 ev is not a good measure of the capture cross section because of the incidence of molecular effects.

The interference effects are influenced by the nuclear spin of one. The binding effect tends to increase the low energy cross section by seven percent.

A: Results for neutron energy less than 1 ev have been published previously.[7] No new measurements have been made. The previous cross section results should be increased uniformly by about 1.5 percent since the standard filter was not used at that time.

H_2: Figure 8 shows the results of measurements.

The rapid increase in cross section at low energies is due primarily to the effect of thermal motion which exhibits its maximum influence in the case of H_2 since it is the lightest molecule. If the H_2 cross section were independent of energy in the absence of thermal translational motion, the cross section would be increased by a factor of 2.4 at 0.003 ev because of it. Actually this assumption is not quite true, and an approximate elimination of the effect of thermal translational motion was made on the assumption that the resulting dependence would be of the form of a constant plus a $1/v$ term. This "corrected" curve is included in Fig. 8 and is seen to be approximately of the form assumed. This curve is expected to be a good approximation to the

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[11] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

[12] O. Halpern, Phys. Rev. **72**, 746 (1947).

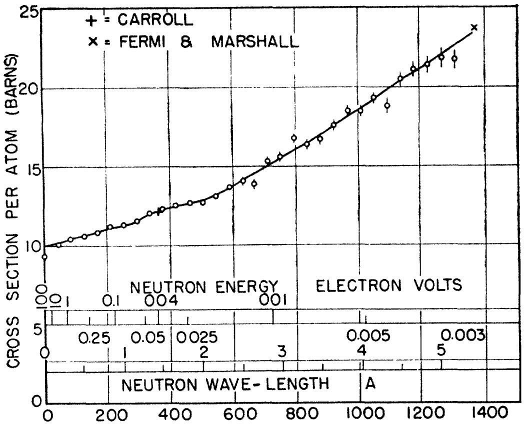

FIG. 7. The slow neutron cross section of N_2 gas to 0.003 ev. For timings less than 665 microseconds/meter a 3.168-g/cm^2 sample was used; while for greater timings the sample contained 1.840 g/cm^2.

cross section as a function of relative velocity for neutron velocity equal to or greater than the mean molecular velocity. For lower neutron velocities, however, it should become progressively poorer because it can be shown that at these energies the observed cross section is more a measure of the cross section at neutron velocity equal to mean molecular velocity than of the cross section at the actual neutron velocity.

The reduced mass effect increases the cross section by $(\frac{2}{3}/\frac{1}{2})^2=1.78$. The scattering amplitudes are of different sign for triplet and singlet interaction so that the interference is sometimes constructive and sometimes destructive.

Methane, ethane, propane, n-butane, cetane, ethylene, and 1,3-*butadiene:* These results, shown in Figs. 9–15, give information concerning the effect of binding of protons in hydrocarbon molecules of varying size and, in some cases, having double bonds. The carbon cross section has been taken as 4.70 barns. For convenient comparison, these curves together with that for H_2 gas are shown together in Fig. 16. Here, approximate elimination of the effect of thermal translational motion has been performed in the way described above in the case of H_2 gas.

In the energy region above 0.1 ev all of the hydrocarbon curves are about the same, indicating that it is the influence of the C—H bond which dominates. At about the energy spacing of vibrational energy levels (0.5 ev) there is a sudden, rapid increase in cross section with decreasing neutron energy. This is probably to be interpreted as the setting-in of the effect of the binding. In the region 0.01 to 0.1 ev the cross sections for a given neutron energy increase in the order of increasing molecular weight. The differences between successively heavier molecules appear to decrease, as expected, although there is some irregularity probably due, in part, to the inaccuracy of the corrections which had to be made since the standard filter was not used for most of these measurements. In particular, the difference between *n*-butane and cetane is small considering the difference in molecular size.

Below 0.01 ev the hydrocarbon curves continue to increase and appear to converge somewhat. Values of cross section approximately four times the free cross section are readily reached, confirming Fermi's prediction of the effect of binding of H in heavy molecules.[13] Bethe has made quantitative calculations concerning the manner in which this binding takes place for neutron energies well below the energies of vibration of the C—H bond.[14] As a model, he considered the proton bound in an anisotropic potential well. His results are shown on the graph for cetane (Fig. 13) and indicate good agreement with the cross section of H in cetaine between 0.01 and 0.1 ev. (Here Bethe's own choice of the values of the vibration wave numbers, 3200 cm^{-1} parallel to the C—H bond and 800 cm^{-1} perpendicular to it, has been used.) Below 0.01 ev the agreement becomes increasingly poorer since Bethe's curve approaches $4\times 20.36=81.4$ asymptotically while the experimental curve shows no such tendency.

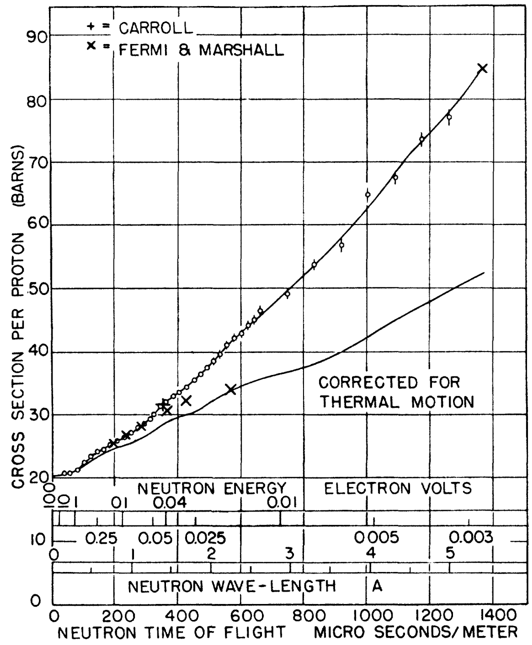

FIG. 8. The slow neutron cross section of H_2 gas to 0.003 ev. For timings less than 324 microseconds/meter the average of measurements on 0.1417- and 0.0825-g/cm^2 samples is shown. Between 324 and 665 microseconds/meter a 0.0825-g/cm^2 sample was used. Beyond 665 microseconds/meter the sample contained 0.0371 g/cm^2. For timings less than 665 microseconds/meter standard filters were used. For timings greater than 665 microseconds/meter BeO filtering was used in addition to Be filtering.

The major cause of this discrepancy is that the binding of the proton is not to an infinite mass but to a carbon atom which can vibrate with respect to neigh-

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[13] E. Fermi, Ricerca Scient. VII-II, 13 (1936).

[14] H. A. Bethe, Rev. Mod. Phys. 9, 69, 117 (1937).

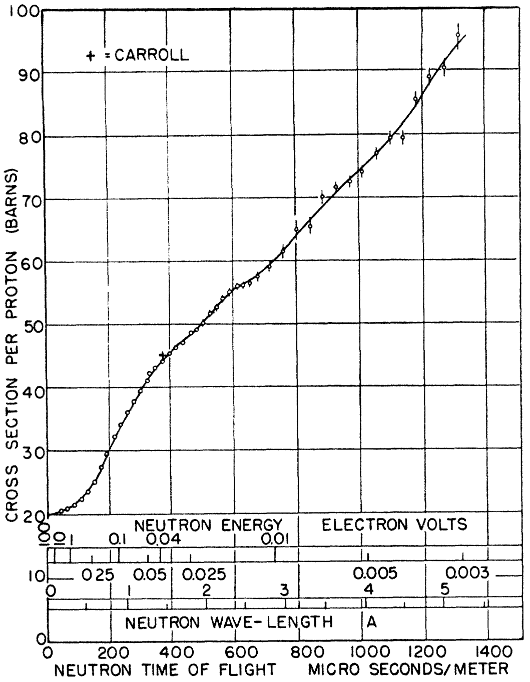

Fig. 9. The slow neutron cross section of H in methane gas to 0.003 ev. For timings shorter than 665 microseconds/meter measurements were made on a 0.2462-g/cm² sample using the standard filter technique. The rest of the curve was taken with a 0.2258 g/cm² sample.

boring carbon atoms. In itself, the small weight of the carbon atom tends to make the cross section level off at 3.41 instead of at 4 times the free value. However, for the heavier molecules, many low energy C−C vibrations are possible which give rise to elastic and inelastic scattering and account for the insufficiency of Bethe's

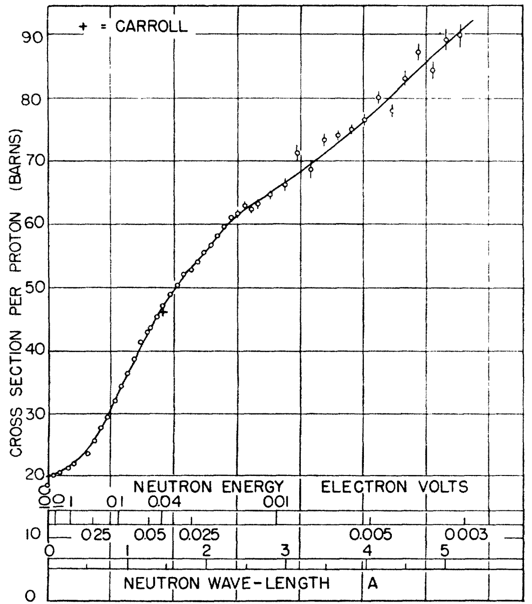

Fig. 10. The slow neutron cross section of H in ethane gas to 0.003 ev. A 0.3055-g/cm² sample was used throughout.

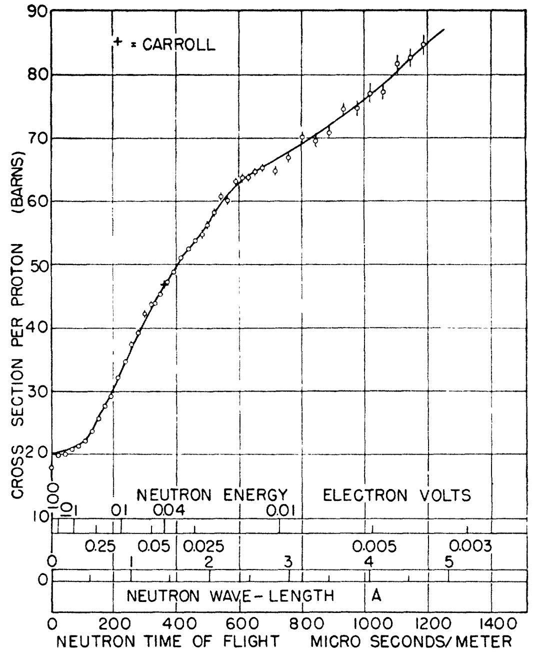

Fig. 11. The slow neutron cross section of H in propane gas to 0.003 ev. For timings less than 324 microseconds/meter a 0.7694-g/cm² sample was used, while the remaining data were taken on a 0.2103-g/cm² sample.

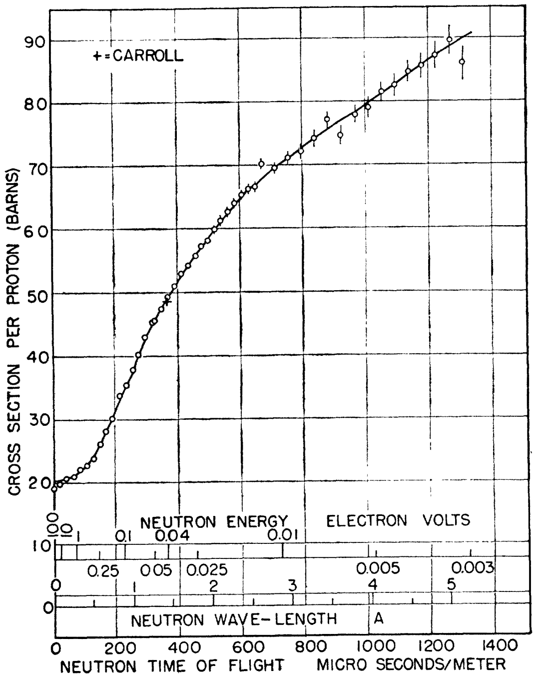

Fig. 12. The slow neutron cross section of H in *n*-butane gas to 0.003 ev. A 0.2789-g/cm² sample was used for timings less than 665 microseconds/meter; while beyond this a 0.0808-g/cm² sample was used.

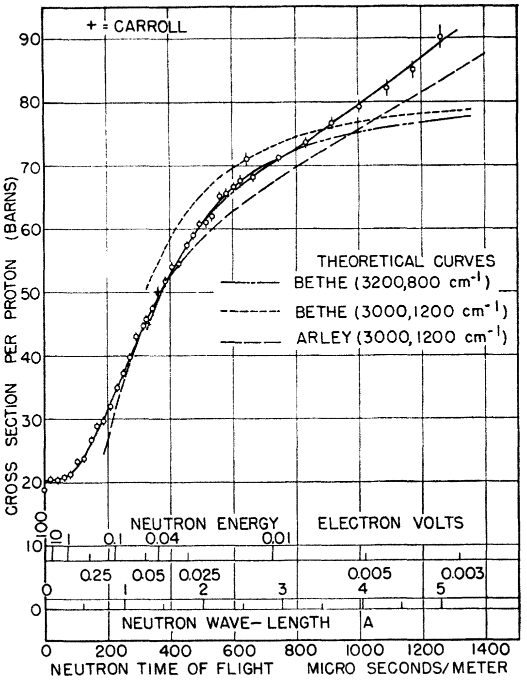

FIG. 13. The slow neutron cross section of H in liquid cetane to 0.003 ev. A 0.2439-g/cm² sample was used. Theoretical curves by Bethe (two choices of vibration frequencies) and by Arley (his own choice of vibration frequencies) are included.

treatment. N. Arley[15] has attempted to take these effects into consideration by using for calculation a model which makes the following assumptions:

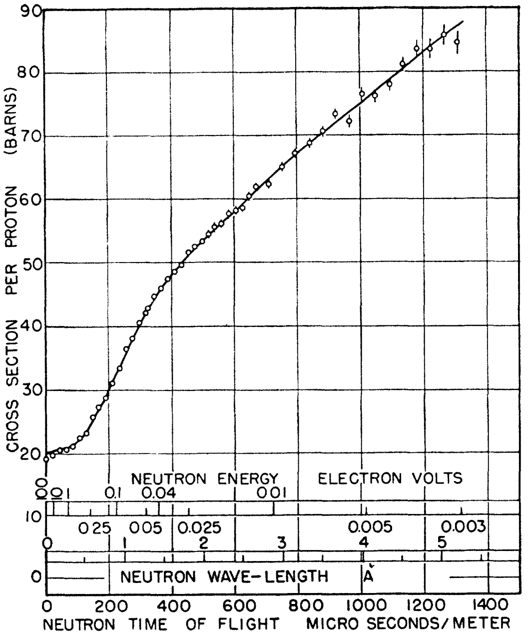

FIG. 14. The slow neutron cross section of H in ethylene gas to 0.003 ev. A 0.3879-g/cm² sample was used.

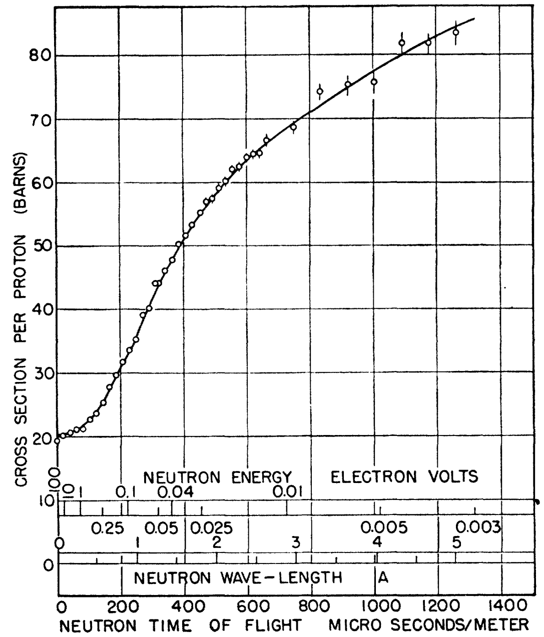

FIG. 15. The slow neutron cross section of H in 1, 3-butadiene gas to 0.003 ev. A 0.4571 g/cm² sample was used for timings less than 665 microseconds/meter, and for larger timings a 0.3347-g/cm² sample was used.

(1) Each proton is assumed to oscillate independently in an anisotropic harmonic potential. The vibration frequency in the direction along the C—H bond is taken as 3000 cm⁻¹ (0.37 ev)

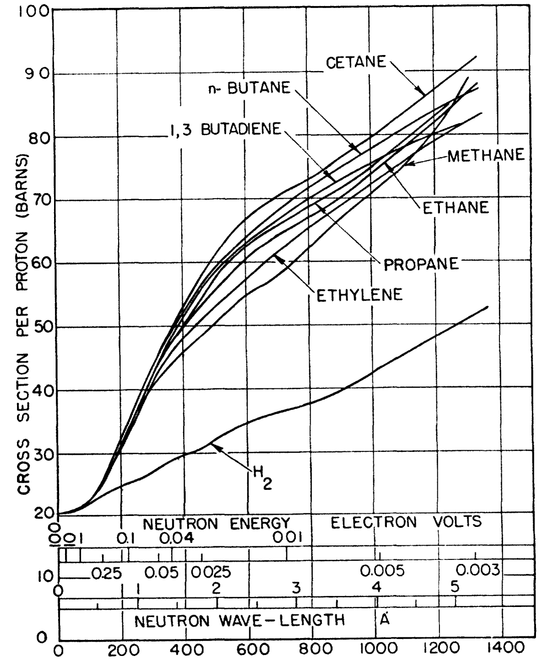

FIG. 16. A composite graph showing the slow neutron cross section of H in the hydrocarbons studied and in H_2 gas. Approximate corrections for the effect of thermal the translational motion have been made for the gas scatterers as described in the text.

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[15] N. Arley, Kgl. Danske Vid. Sels. Math.-fys. Medd. 16, 1 (1938).

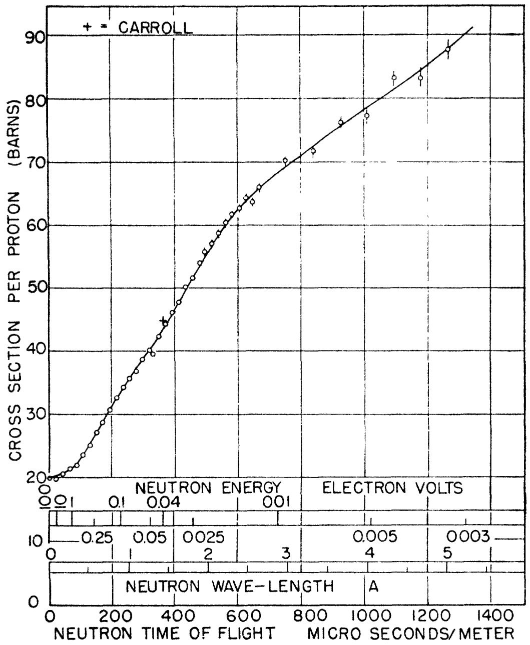

FIG. 17. The slow neutron cross section of H in water to 0.003 ev. A 0.3142-g/cm² sample was used throughout. BeO filtering was used in addition to the usual Be filtering for timings longer than 665 microseconds/meter.

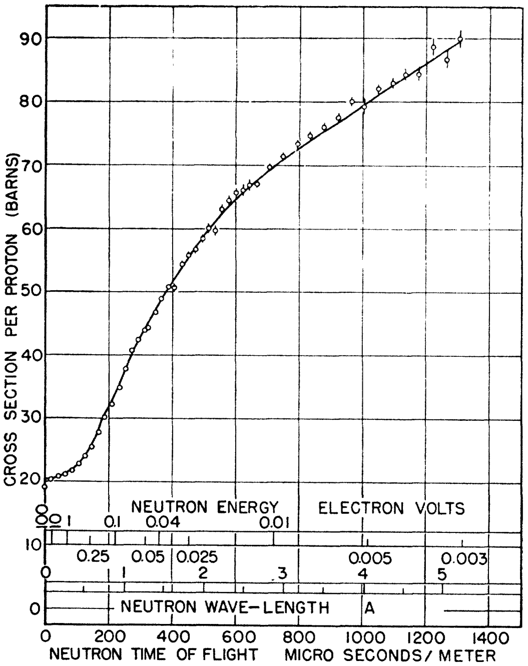

FIG. 18. The slow neutron cross section of H in liquid *n*-butane to 0.003 ev. A 0.272 g/cm² sample was used. The solid curve is that for gaseous *n*-butane.

while 1200 cm^{-1} (0.148 ev) is used for vibrations perpendicular to the C—H bond.

(2) The lower frequencies are taken into account by ascribing an effective mass, chosen as 14 times the neutron mass, to the system consisting of proton plus potential, and assuming that these "molecules" move freely like gas molecules with a Maxwellian distribution.

The results of calculation on the basis of this model are included in Fig. 13. It is seen that the cross section is no longer limited to four times the free cross section as before, but can now follow roughly the experimental curves. Arley's choice of vibration frequencies, somewhat different from that of Bethe, is in better agreement with recently measured values of these frequencies. Figure 13 includes a curve based on Bethe's theory but using Arley's figures. This does not agree as well with the cetane curve, but quantitative agreement is not to be expected.

There are other small effects which have not yet been taken into account. The binding of the carbon atom should contribute about 0.4 barn. Coherent scattering from the spinless carbon atoms should become increasingly important with increasing neutron wavelength and with increase in the number of carbon atoms in the molecule. Interference of the scattered neutrons from protons should add only a little to the cross section since singlet and triplet scattering amplitudes have opposite sign.

H_2O: The results, given in Fig. 17, look similar to those for the heavier hydrocarbons. The oxygen cross section has been taken as 3.73 barns.

Liquid n-butane: The direct determination of g/cm² was difficult and proved to be unreliable. The measured value was increased by 3.5 percent to make the cross-section curve give the correct value of the free proton cross section above 1 ev. The results are shown in Fig. 18. The solid curve of this graph has been taken from Fig. 12 and indicates that there is no significant difference between liquid and gaseous *n*-butane so far as neutron cross-section measurements are concerned.

Comparison with Previous Measurements

Numerous transmission measurements have been made using the neutrons absorbed by cadmium (*C* neutrons). Previous experience[2] indicates that the effective energy is not $kT=0.025$ ev as was supposed, but that when 0.04 ev is assumed agreement with spectrometer data is usually good. Carroll[16] made measurements of this kind on numerous gas, liquid, and solid scatterers. His observed cross-section values are plotted at 0.04 ev neutron energy on the corresponding graphs. Agreement is very good in general. Numerous measurements with *C* neutrons have been made on paraffin.[16,17] Carroll's[16] value of 49.8±0.2 is typical of the best measurements. The only comparable measurement of the present investigation is that for cetane which gives 48.8 at 0.04 ev.

Two comparable sets of measurements have been made using neutron velocity spectrometers of the cyclo-

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[16] Henry Carroll, Phys. Rev. **60**, 702 (1941).

[17] Dunning, Pegram, Fink, and Mitchell, Phys. Rev. **48**, 265 (1935). Goldhaber and Briggs, Proc. Roy. Soc. **A162**, 127 (1937). G. A. Fink, Phys. Rev. **50**, 738 (1936). Powers, Goldsmith, Beyer and Dunning, Phys. Rev. **53**, 947 (1938).

tron arc modulated type. Jones has reported measurements on water over the same energy range covered here.[4] His cross-section results are consistently lower than those of Fig. 17. For neutron energies greater than 0.005 ev this discrepancy amounts to only a few percent and may be attributed to difficulties of making absolute measurements. At lower energies, however, this difference becomes worse and reaches 10 percent at 0.003 ev. The larger cross-section results are probably more reliable because all known difficulties of measurement tend to give too low a value for the observed cross section. Rainwater, Havens, Dunning, and Wu have published spectrometer measurements on paraffin.[2] Between 0.05 and 0.005 ev their results are in agreement with Bethe's curve (with his own choice of vibration frequencies) which is shown in Fig. 13.

Using a crystal spectrometer Fermi and Marshall[18] have investigated H_2 gas at room temperature and at 83°K from 0.75 to 2.5A neutron wave-length. Their results for the gas at room temperature are included in Fig. 8 and indicate good agreement with the present measurements for wave-lengths shorter than 1.2A. At longer wave-lengths, however, there is a marked discrepancy. Again the larger values for the cross section are likely to be more reliable.

Fermi and Marshall[18] have also measured the cross section of O_2, N_2, and H_2 gases for neutrons filtered through BeO. They initially reported the effective wave-length of these neutrons as 5.1A, and their measured cross-section values were about 10 percent larger than the present results as the stated wave-length.[7] In a subsequent report,[19] however, they gave 5.43A as the effective wave-length. This value has been used in plotting their results on Figs. 6–8. The agreement is now much better.

ACKNOWLEDGMENTS

The author wishes to express appreciation to Professor J. R. Dunning who suggested this problem, and to Professors L. J. Rainwater and W. W. Havens, Jr., who took an active interest in the experiments and made numerous invaluable suggestions. Thanks are also also due to Professor T. I. Taylor for the mass-spectrographic analyses of some of the gas samples; Drs. G. Placzek, H. A. Bethe, and O. Halpern for interesting discussions on several points; and Miss Miriam Levin and members of the cyclotron staff for assistance in making some of the measurements. The author also wishes to thank the AEC whose support made these researches possible.

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[18] E. Fermi and L. Marshall, Phys. Rev. 71, 666 (1947).

[19] E. Fermi and L. Marshall, Phys. Rev. 75, 578 (1949).

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The Stopping Power for Protons in Several Metals*

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The rate of energy loss of low velocity (i.e., from 50 to 400 kev) protons has been measured in five different stopping materials: Be, Al, Cu, Ag, and Au. Agreement with previous measurements at higher velocities (greater than 400 kev) is good where such measurements have been published. The stopping measurements were made on foils prepared by an evaporation technique and are believed to be more accurate than measurements made on the usual (hammered or rolled) foils; the results give a value for stopping about 15 percent higher (in Au) and about 7 percent lower (in Al) than those published by Wilcox. A comparison to existing theory at very low and at high velocities (the critical velocity being defined as $e^2/\hbar$) was made with good agreement shown at the low end. The high end theory is of doubtful validity and gives a stopping power about 10 percent higher than observed. It is argued that this discrepancy cannot be accounted for by a proton charge less than unity due to the electron capture-loss process.

I. INTRODUCTION

SEVERAL recent discussions[1] of the stopping power for charged particles in the low velocity region make an experimental determination interesting from a theoretical point of view. An adequate theory of stopping power, in the region where ionic velocity becomes comparable with the orbital electronic velocity, does not really exist. The measurements that will be reported on in this paper are, therefore, compared to qualitative extensions of a familiar theory that is valid at very high proton energies, but rather less accurate for energy about 500 kev—and quite invalid below this point. A recent argument that should be valid in the region of "adiabatic" collisions, i.e., below about 15 kev, will also be discussed. The particular metals on which

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[1] E. Fermi and E. Teller, Phys. Rev. 72, 399 (1947); N. Bohr, Kgl. Danske Vid. Sels. Math.-fys. Medd. 18, 8 (1948) (this work will be referred to as (N.B.) in the following); see also J. Knipp and E. Teller, Phys. Rev. 59, 659 (1941).