A Precise Determination of the Slow Neutron Cross Section of the Free Proton

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(Received August 19, 1949)

The total neutron cross sections of H_2, *n*-butane, and water have been measured with the Columbia slow neutron velocity spectrometer in the energy range 0.8 to 15 ev. Analysis of these data gives a value of 20.36 ± 0.10 barns for the free proton cross section. In this energy interval the measurements confirm Placzek's theoretical prediction of the variation of the H_2 cross section. The technique of making absolute transmission measurements and the corrections for multiple scattering are discussed. Measurements on H_2 for neutron energies above 15 ev show a constant cross section to several thousand ev. Measurements on fused quartz in the energy interval 0.003 ev to 10 ev show interference effects.

INTRODUCTION

THE slow neutron cross section of the free proton is one of the important constants of nuclear physics inasmuch as the neutron-proton interaction involves elementary particles and hence lends itself to theoretical investigation. In particular, the free proton cross section used in conjunction with the results of the experiments on the scattering of neutrons by parahydrogen enables one to calculate the range of nuclear forces for the triplet interaction between neutron and proton.[1]

Because of the importance of the value of the free proton cross section, it has been measured by many investigators. An examination of these experiments indicated that a better determination was desirable. It is the purpose of this report to describe a precise determination of this quantity using measurements on H_2, *n*-butane, and H_2O. Comparison with previous results will be presented.

EXPERIMENTAL PROCEDURE

The Slow Neutron Velocity Spectrometer

This apparatus has been previously described in detail.[2] There has been one major change. The fundamental timing is now provided by a one-megacycle oscillator instead of a 100-kilocycle oscillator.[3] Much better resolution is thus made available. The following brief description emphasizes other improvements and adaptations and has been kept sufficiently general to serve for the following paper.[4]

The neutron spectrometer system uses the method of cyclotron arc modulation which confines neutron production to intervals of four to 1024 microseconds duration out of a 1024- to 32,768-microsecond cycle. Neutrons slowed down in a paraffin "source" slab are detected by BF_3 proportional counters at a distance of 6.0 meters (unless otherwise stated) from the paraffin source. A special selector system counts selectively those neutrons detected during adjustable time intervals after the neutron production. Knowledge of time of flight and distance enables calculation of neutron energy within the resolution limits.

Two paraffin source slabs have been in use. The "thin source" is effectively one inch thick and is used for timings less than 2048 microseconds to keep source delay time corrections to a minimum. For longer timings a "thick source" effectively three inches thick has been used to give greater intensities of low energy thermal neutrons. Appropriate delay time corrections are made.

The collimating system is similar to that used previously, but provision is made for handling the one-meter long gas sample holders. The mean position of the samples (gas or liquid) is 1.1 meters from the BF_3 counter. The slow neutron flux is collimated to 6.3-cm diameter. The detector subtends less than 3×10^{-4} of the 4π-solid angle viewed from the sample position; therefore no "geometry" corrections have been made in general except in the case of the determination of the free proton cross section where a high degree of refinement was sought. Even here the correction was less than 0.5 percent.

Absolute Measurement of Cross Section—The Standard Filter Technique

In the past, attempts at various times to repeat measurements on a given sample had frequently yielded sets of results each internally consistent, but differing among themselves by a few percent, which was considerably greater than the statistical uncertainty. This situation was climaxed by the occurrence of discrepancies in the measurements made with the two separate channels. Both counters were replaced with new ones with the hope of correcting this situation. Measurements using the new counters gave cross-section values systematically higher than any of the previous results. Exploratory tests seemed to indicate that these discrepancies arose because of a slight dependence of detection efficiency upon the counting rate. (The background rate was negligibly small in all cases.) The re-

[1] Wu, Rainwater, Havens, and Dunning, Phys. Rev. **69**, 236 (1946); M. Hamermesh and J. Schwinger, Phys. Rev. **71**, 678 (1947).
[2] Rainwater, Havens, Wu, and Dunning, Phys. Rev. **71**, 65 (1947).
[3] Havens, Rainwater, Wu, and Dunning, Phys. Rev. **74**, 1216A (1948).
[4] E. Melkonian, Phys. Rev. **76**, 1750 (1949).

sults could be explained if the counting efficiency were smaller for higher counting rates ("sample out" count) than for lower counting rates ("sample in" count). It was not known whether this dependence upon counting rate was caused by the counters or by the associated circuits, although subsequent tests seemed to indicate that the latter were at fault. This difficulty could be eliminated by making the counting rates approximately the same in the "in" and the "out" positions. This can be accomplished, for example, by using in the "out" position a sample of appropriate transmission whose cross section is known from other considerations to the required accuracy. Since the transmission of no such substance was known, a B_4C aperture system was constructed whose transmission is known absolutely from geometrical considerations. This device is denoted by the term "standard filter." The use of the standard filter has given results which are consistent within statistical uncertainty. The new results are systematically higher in cross section by a few percent (3 percent or less) in agreement with the assumptions given above.

A standard filter (for one channel) consists of two parts: (1) a "stator" which always remains in the beam in a fixed position and (2) a "rotor" which is used only when the sample is not in the beam.

The standard filter to give transmission equal to $\frac{1}{3}$ will be described. Standard filters to give other transmission values have been made in a similar way.

The stator consists of a disk of B_4C paraffin mixture one inch thick and five inches in diameter in which three equal wedge-shaped openings are cut out leaving three spokes each $\frac{1}{4}$ inch wide (Fig. 1). The cut-out portions extend radically out far enough (1.5 inches) so that the opening in the collimating system is more than fully exposed in the open sections of the stator.

The rotor consists of a similar disk but with only one wedge-shaped opening going to the exact center of the disk, the central angle of the opening being the same as in the case of the stator. When placed properly over the stator, the rotor covers two openings completely, leaving the third opening completely exposed. The stator spokes are wide enough so that the rotor edge is sufficiently far back that it cannot be "seen" by the neutron beam, and so that neutrons coming through the covered openings at the steepest angle possible cannot get through. *Thus each opening acts independently of the others, so that the number of neutrons going through a given opening is the same whether the other openings are covered or not.* When this device is accurately made, and the neutron beam is completely uniform across the three openings, then the number of neutrons reaching the detector when the rotor is used would be exactly $\frac{1}{3}$ the number without the rotor. Thus, for a sample having transmission approximately $\frac{1}{3}$, one alternates sample and rotor in the beam and gets a direct comparison of the sample transmission with that of the standard, at the same time getting approximately the same counting rates in both "in" and "out" positions and so avoiding errors resulting from the rate dependence of the detection efficiency.

For the neutron beam actually used the intensity is not the same for the three openings (as much as 30 percent variation from the average having been observed). Although the openings do not separately give exactly $\frac{1}{3}$ of the total intensity, the desired effect is obtained by exposing the three openings in sequence, each for $\frac{1}{3}$ of the time allotted to the "out" count. A residual effect still remains, as the counting rate (and hence detection efficiency) for the various openings is still not exactly the same as that for the "in" count. However, the variation of counting rate is now much less than without the standard filter. Also, there is a strong tendency to cancel out the error arising from this variation as long as the average counting rate with the standard filter matches that with the sample. A simple calculation shows that the error arising from this non-uniformity of the beam is negligible.

A solid one-inch thick piece of B_4C paraffin mixture is periodically placed in the rotor position to determine what counts would get through the solid portions of the standard filter system. Only data are used for which this background count is negligible. In general, the standard filter described here cannot be used for energies above 15 ev since the filter is not sufficiently opaque at these higher energies.

In the discussion so far it has been tacitly assumed that the efficiency of detection for this spectrometer system depends solely upon the instantaneous counting rate. This appears to be at least partially true. There is, however, some evidence to indicate that the detection efficiency at any instant depends somewhat upon the intensity in previous portions of the timing cycle. In the extreme limit, the detection efficiency might depend upon the intensity averaged over the entire cycle. For measurements between 0.5 and 15 ev, the repeat time used is 1024 microseconds, and cadmium is used to prevent overlapping with subsequent cycles. For the samples studied, as in the case of the determination of the free proton cross section, the transmission of the sample varies very little (~10 percent) over this interval. The ratio of "sample in" to "sample out"

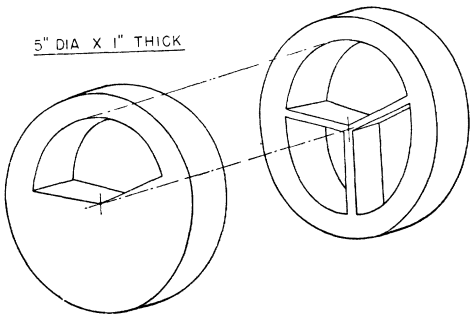

FIG. 1. Diagram showing standard filter to give a transmission of $\frac{1}{3}$. It is made of a B_4C paraffin mixture.

intensity is thus approximately the same whether instantaneous or averaged transmission values are considered, and there is no difficulty in choosing the appropriate standard filter. When longer repeat times are to be used where the sample transmission varies considerably, the choice of the proper standard filter transmission value is somewhat more difficult since it is not known whether the sample transmission of the portion of the cycle being studied should be matched, or whether the sample transmission averaged over the entire cycle should be matched. This problem arose in the case of H_2

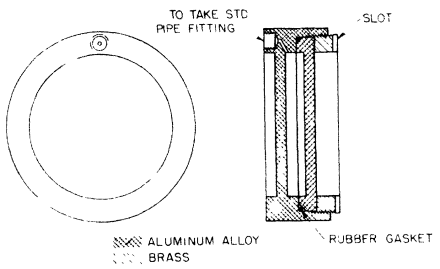

FIG. 2. Holder for liquid scatterer.

and methane when measurements with standard filter were made to timings of 4096 microseconds out of a repeat time of 8192 microseconds. In such cases compromise values were adopted between the possible extremes.

It should be stressed that the discrepancies corrected by the standard filter technique are at most 3 percent (under otherwise good conditions) and are usually less with new counters. Hence, uncertainties in the absolute values of previous measurements made with the spectrometer are only of this order. Most of the previous work was concerned primarily with the shape of the transmission curve (e.g., in the neighborhood of a resonance) and this is only slightly affected by the variation of detection efficiency with counting rate. Only when a precise determination of an absolute value for the cross section is desired is the use of the standard filter technique essential (with the present equipment).

An unsuccessful attempt to use fused quartz as a secondary standard filter in place of the less convenient primary standard filter is described in the Appendix.

Sample Holders

The holders used for liquid and gas samples were constructed of aluminum alloy to minimize attenuation of the neutron beam in the ends.

Several liquid sample holders of different design have been used. A particularly versatile design is shown in Fig. 2. A variety of sample thicknesses may be obtained by using interchangeable plates hollowed to different depths. Use of the valve allows filling with liquids having vapor pressure above atmospheric pressure (e.g., liquid *n*-butane). A "blank" having the same thickness of the same aluminum alloy was used in the sample "out" position.

The gas sample holder, shown in Fig. 3, can be used safely with gas pressures to 1000 p.s.i. These pressures are necessary when the gas being investigated has a low cross section. The brass to aluminum seals of the inlet, used in both gas and liquid sample holders, are gastight at the highest pressure used in testing (about 3000 p.s.i.). The seal itself is made in the seat below the threads. An identical evacuated sample holder was used in the "sample out" position.

Preparation and Purity of Samples, and Determination of Sample Thickness

The scatterers used in this investigation are included in those reported on in the following paper[4] to which reference is made for details on the preparation and purity of the samples and the determination of the number of grams per cm^2 in the path of the beam.

RESULTS

Neutron Energies between 0.5 and 15 ev

For the final measurements under the best experimental conditions, several different materials were in-

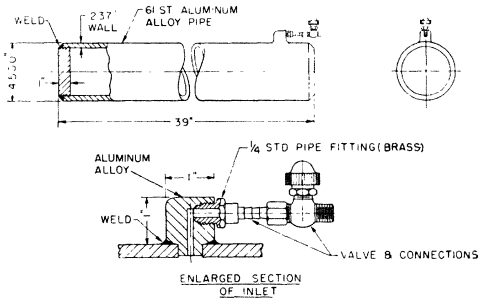

FIG. 3. Holder for gas scatterer.

vestigated to avoid possible systematic errors depending upon the material or sample used. Water was selected because it is typical of condensed substances, because there can be no doubt of the sample composition, and because several other investigators have used it. H_2 was used because it is the simplest hydrogen-containing substance and because it is a gas. (This latter point is important in connection with the multiple scattering correction which will be discussed later.) In addition, theoretical calculations of the expected σ *vs.* E dependence are most readily made for H_2. Gaseous *n*-butane was selected as a third scatterer somewhat different from the other two.

Measurements at sufficiently high energies to make the molecular binding effects completely negligible are not feasible as the poorer intensity accompanying the good resolution required makes it difficult to get the desired statistical accuracy. Also, the standard filter becomes partially transparent above 15 ev and hence

cannot be used. If measurements are made at energies at which binding effects are not negligible, suitable corrections must be made. Placzek has developed a theory[5] which shows that, at neutron energies large compared with the energy of spacing of the molecular vibrational levels, the cross section is linear in the reciprocal of the neutron energy, i.e.:

$$\sigma = \sigma_0 + \beta/E.$$

Thus, a plot of the cross-section data against the reciprocal of the neutron energy should give a straight line whose intercept is the free proton cross section. For the case of H_2, Placzek has given a calculated value for β which depends upon well-known constants of the H_2 molecule not related to neutron measurements. Only approximate calculations have been made in the case of water and *n*-butane.

The energy range chosen for measurement was approximately 0.5 to 15 ev. A repeat cycle of 1024 microseconds with cadmium filtering was used. Cyclotron on time and detection intervals of 32 and 64 microseconds were used, although most of the measurements were done with the latter for the sake of higher counting rates.

The experimental results are shown in Figs. 4–6, cross section being plotted against $1/E$. These data represent the average of several runs in each case. Correction has been made for capture of neutrons by protons assuming a $1/v$ variation of cross section, with the capture cross section taken as 0.3 barn at 0.025 ev. This correction is quite small, being only 0.05 barn at 1 ev. The statistical accuracy is indicated by vertical bars. Points above 15 ev have not been included because the standard filter becomes partially transparent.

The straight lines shown in these graphs have been obtained by means of least-square solutions using only the data above 0.8 ev. In this energy region, the straight line fit is seen to be good in accordance with Placzek's theory. Points below 0.8 ev have been included for H_2 and *n*-butane to show how deviation from the $1/E$ behavior begins.

The values of σ_0 and β (with their statistical standard deviations) for the best straight lines are listed in Table I. Placzek's theoretical values are also included for comparison. The agreement between observed and calculated values of β in the case of H_2 is considered satisfactory. The calculations in the case of H_2O are much more difficult and have been done roughly, so that only approximate agreement is expected.

The main contribution to the value of β arises from the existence of the "zero point energy" of an oscillator. In most phenomena, only energy differences contribute, so that the zero point energy is generally undetectable. It is interesting to note that we have here an experimentally observed phenomenon which depends almost entirely upon the presence of this quantum-mechanical effect.

The measured values of the cross section must be corrected for the effect of neutrons reaching the detector after one or more collisions in the scatterer. In spite of the very good geometry used, this effect of "in scattering" is not negligible although it is quite small. This effect has sometimes been corrected for by other investigators on the assumption that only the effect of single scattering is important so that the true transmission (i.e., for perfect geometry) is increased by an amount proportional to $(1-T)$, where T is the sample transmission. This is approximately true for T almost equal to unity. However, this treatment is sometimes applied when T is quite different from unity. Actually, this estimate would probably serve as a good upper limit to the correction for any magnitude of the transmission where it not for the fact that the detector used has a $1/v$ detection efficiency. Neutrons reaching the

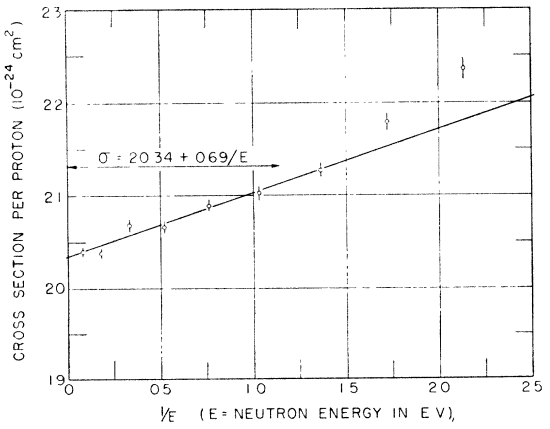

FIG. 4. The slow neutron cross section of H_2 gas in the energy region 0.5 to 15 ev. The average of measurements on samples of 0.0825 and 0.1417 g/cm^2 is shown. Correction for capture has been made.

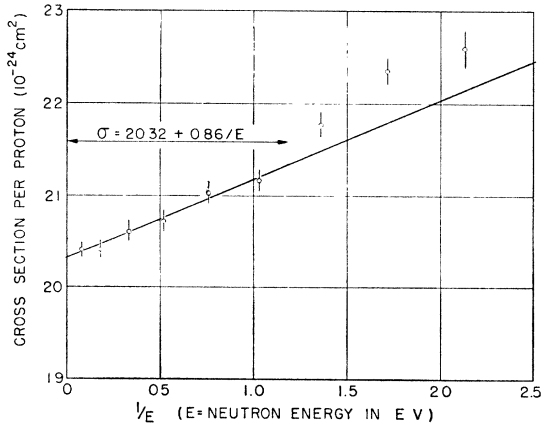

FIG. 5. The slow neutron cross section of H in gaseous *n*-butane in the energy region 0.5 to 15 e.v. The sample used contained 0.4351 g/cm^2. The carbon cross section was taken as 4.70 barns. Correction for capture has been made.

[5] The author is indebted to Dr. G. Placzek for communication of his results prior to publication.

TABLE I. Results of measurements and calculations.

Scatterer	σ_0 in barns: As measured	σ_0 in barns: Corrected for "In" scattering	β: Observed	β: Calculated (Placzek)
H_2	20.34±0.05	20.37	0.69±0.09	0.65
n-butane	20.32±0.08	20.35	0.86±0.14	
H_2O	20.27±0.06	20.37	1.36±0.11	1.0

detector after only one collision in the scatterer have essentially unchanged energy. However, neutrons reaching the detector after two or more collisions may have

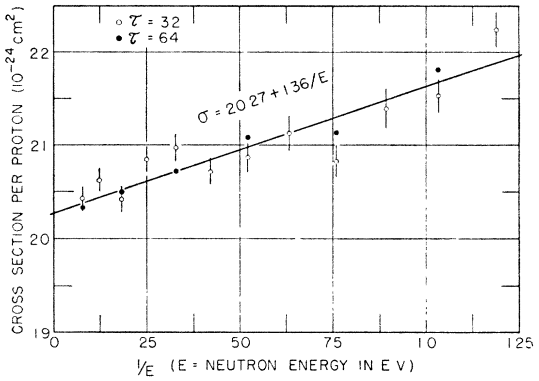

FIG. 6. The slow neutron cross section of H in H_2O in the energy region 1 to 15 ev. A 0.9407-g/cm² sample was used. The oxygen cross section was taken as 3.73 barns. Correction for capture has been made.

considerably reduced energy and, being detected more efficiently, contribute a large amount to the inscattering effect. Because of this, single and double scattering have been considered separately in the present investigation in order to arrive at the corrections to be applied to the measured values of the cross section.

The correction for single scattering considers only those neutrons which reach the detector after one collision in the scatterer. This amounts to an increase in the cross section of 0.025 for the water sample and 0.03 for the gas samples. (The latter correction is slightly larger because the "geometry" of the 94-cm long gas scatterer is effectively poorer than that of the water sample even though the mean positions are the same.)

The correction for double scattering considers only those neutrons which reach the detector after two collisions in the scatterer. In carrying out the calculations, the following facts have been taken into account:

(a) The detection efficiency for neutrons is proportional to $1/v$.

(b) Since the total time of flight for a slowed-down neutron must be the same as that for a neutron reaching the detector with no collision in the scatterer, the former neutron must have had greater energy initially than the latter. On a time of flight basis, a given spread is reflected in a larger spread after slowing down.

(c) The detected intensity has been taken as uniform on a time of flight basis. Actually there is a small decrease in detected intensity with increasing time of flight in the energy region used. This would tend to make the corrections slightly larger than calculated.

For the case of the gas scatterer the double-scattering correction is less than 1/20 of the single-scattering correction and hence may be neglected. However, for the water scatterer, this double-scattering correction turns out to be about as large as that for single scattering. Calculation of corrections for higher orders of scattering becomes increasingly complicated and has not been carried out. However, there is no reason to expect that these will be negligible. As an estimate of the total correction for water, the measured cross section has been increased by 0.10 which is twice the combined correction for single and double scattering. This correction is still only ½ percent of the measured cross section.

The reason for the difference in magnitude of the double-scattering correction for gas and liquid scatterers may be seen on the basis of simple considerations. Both liquid and gas samples have been so chosen that the sample thickness is approximately equal to the neutron mean free path in each case. For the gases, this is 94 cm; whereas for water (as well as for other condensed substances such as cetane, paraffin, etc., which have been used in the past) it is about 0.7 cm. The diameter of both water and gas samples is about 6.5 cm. Thus, for a gas scatterer, a neutron being deflected by a large angle after the first collision has a good chance of leaving the sample before making a second collision. For water, on the other hand, such a neutron has many mean free paths equivalent of material to pass through before leaving the sample, and thus has a good chance to make a second collision.

The corrected results are included in Table I. The agreement of the three determinations is excellent, but not as much confidence should be placed on the results on H_2O because of the uncertainty in the inscattering correction. It does serve to indicate, though, that no serious systematic errors are present. The results on H_2 should be weighted most heavily because the most data were taken on it and because the measured and theoretical values of the $1/E$ slope agree.

The free proton cross section is taken as 20.36 from these measurements. Statistical errors arising from

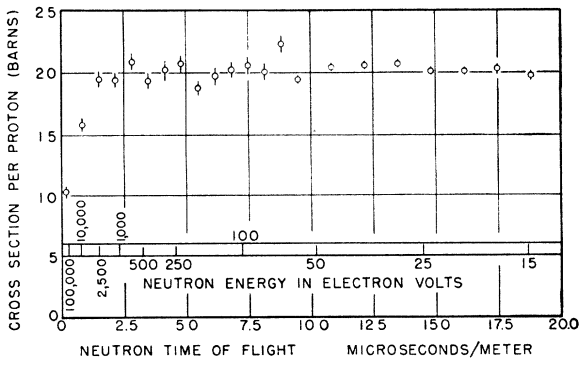

FIG. 7. The slow neutron cross section of H_2 for neutron energy greater than 15 ev. The sample used contained 0.1417 g/cm² of H_2.

counting and errors from determination of sample thickness indicate that a probable error of 0.10 should be placed on the stated figure.

An abstract of this work has been published.[6] Results reported still earlier[7] were made without the use of the standard filter and did not take into account the variation of the proton cross section above 1 ev.

The best previous determination of the free proton cross section has been made by Jones[8] who, using a similar set-up, obtained good statistical accuracy and corrected for molecular binding effects using Placzek's theory. His measurements on water and cyclohexane gave 20.0 ± 0.3 and 20.1 ± 0.3, respectively. His geometry was somewhat poorer than that used for the present measurements, and his final results include an additive correction of 0.25 for the effect of inscattering which he has evaluated on the assumption that the transmission is increased by an amount proportional to $(1-T)$. It is interesting to note that Jones' result is increased to $\sim$20.3 (in good agreement with the results of the present measurements) if the inscattering correction method of this paper is used. The small difference between the results of this paper and those of Jones is thus probably due mainly to his use of too small a correction factor.

A previous determination with the Columbia slow neutron velocity spectrometer has been made by Rainwater, Havens, Dunning, and Wu[9] who reported a value of 20.6 ± 1 from measurements on a paraffin scatterer.

Still earlier determinations have been summarized in reference 9. These measurements are characterized by: (1) use of liquid or solid scatterer, (2) relatively poor statistical accuracy in most cases, (3) poorer geometry than that for the determinations already discussed, and (4) no corrections for the effect of molecular binding where present.

Neutron Energies above 15 ev

In this energy region molecular binding effects are negligible. The neutron-proton cross section should decrease with increasing neutron energy as shown theoretically and checked by measurements to 6 Mev.[10]

Figure 7 shows measurements on H_2 using 4- and 8-microsecond cyclotron and detection on-times (without the standard filter technique). The cross section is seen to be constant until 2500 ev. Above this the results behave approximately as expected, but a detailed comparison with expectation is meaningless because of the relatively poor resolution used at the higher energies.

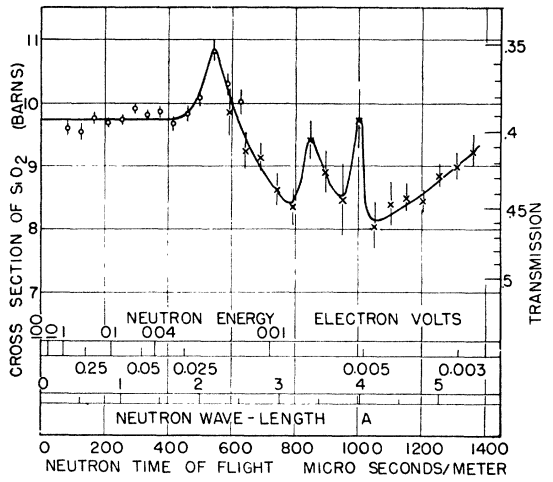

FIG. 8. The slow neutron cross section of fused quartz. The sample contained 9.583 g/cm². ○—Source detector distance = 6.0 meters. ×—Source detector distance = 5.0 meters.

APPENDIX

Measurements on Fused Quartz

The standard filter technique is somewhat inconvenient to use and leads to almost a factor of two loss of detected intensity so that a calibrated secondary standard would be useful. Fused quartz (SiO_2) has elements with negligible capture in the slow neutron region and was expected not to show microcrystalline interference effects because of its supposed lack of long range crystalline structure. Thus it was expected to show a transmission nearly independent of neutron energy and hence to be suitable for use as a secondary standard. Figure 8 shows measurements on fused quartz made against a standard filter. Fairly large interference effects are evident which make this particular sample useless as a secondary standard. The shape of the curve is somewhat similar to that which would be expected for a liquid and is included here because of its interest in connection with the structure of vitreous materials.

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 due 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.

[6] Melkonian, Rainwater, and Havens, Phys. Rev. **75**, 1295A (1949).
[7] E. Melkonian, Phys. Rev. **73**, 1265A (1948).
[8] W. B. Jones, Jr., Phys. Rev. **74**, 364 (1948).
[9] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948).
[10] D. Bohm and C. Richman, Phys. Rev. **71**, 567 (1947).