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Measurement of the neutron-proton total cross section at 132 eV

W. Dilg
Physik-Department, E 14, Technische Universität München, München, Germany

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The neutron-proton scattering cross section at "zero energy" (σ_0), which is an important experimental parameter in the analysis of low energy np data, has been accurately redetermined by transmission measurements at 132 eV neutron energy on H_2O, C_6H_6, CH_3OH, and C_7H_8. We obtain the total cross sections per molecule (132 eV) 44.731 ± 0.027, 151.40 ± 0.10, 90.37 ± 0.08, and 197.20 ±0.34 b, respectively. To evaluate σ_0 we subtract the well-known cross sections of carbon and oxygen, and correct for the small contributions ($<0.1\%$) of capture, molecular binding, effective range, and deuterium. Combining all known errors from the experimental and evaluation procedure, we find $\sigma_0 = 20.491 \pm 0.014$ b. This is 0.27%, or 2.4 standard errors, above the best previous measurement by Houk, and ~0.6% above the mean of former data. Our result of σ_0 implies changes in the np effective-range parameters towards higher values, by about twice the present standard errors in a_s, a_t, and r_{0t} and 1 standard error in r_{0s}.

[NUCLEAR REACTIONS $^1H(n,n)$, $E=132$ eV; measured σ_T. Cobalt-resonance neutron beam, targets H_2O, C_6H_6, CH_3OH, C_7H_8.]

I. INTRODUCTION

The accurate determination of the np cross section as a function of energy has been the subject of much experimental effort[1] for several reasons. Such data form the main basis for adjusting theoretical models of the np interaction[2] and provide accurate quantitative information about the charge dependence of the nucleon-nucleon interaction by comparing the singlet np and pp effective ranges.[3,4] Besides, the neutron cross section for hydrogen is of considerable experimental interest as a standard in fast neutron flux measurements.[5-9]

Most analyses[2-9] of the low energy np cross section have used effective-range theory[10] to parametrize the large body of available data. In the shape-independent approximation, which is an excellent approach to about 5 MeV neutron energy,[11] the cross section is written in terms of only four parameters, e.g. the scattering lengths and effective ranges for the singlet and triplet state, respectively. The scattering lengths are found combining the cross section at "zero energy"[4] and the coherent scattering length.[12] The triplet effective range, in turn is determined by the deuteron binding energy, while the singlet range is derived by fitting the total cross section in the MeV range. A list of 1973 best values of these parameters was recently tabulated by Lomon and Wilson.[2]

This paper reports on a remeasurement of the np scattering cross section at zero energy. The presently accepted value, $\sigma_0 = 20.436 \pm 0.023$ b, is due to Houk.[4] A redetermination appeared to be useful for two reasons.

First, the experimental uncertainty in σ_0 is still by far the dominating source of error[13] in each of the parameters a_t, a_s, r_{0t}, and r_{0s}, respectively. One of Houk's conclusions was, therefore, that further improvements in the MeV cross section were not worthwhile unless the zero energy value was correspondingly improved. Since then, other MeV data have indeed become available, both from precision measurements using monochromatic neutrons[14,15] and from advanced time-of-flight experiments.[16]

Secondly, none of the determinations of σ_0 prior to Houk's had been of comparable accuracy,[17] which means that the precision presently ascribed[2] to the effective-range parameters rests entirely upon one measurement.

In this experiment we have performed transmission measurements at 132 eV neutron energy on water, benzene, methanol, and toluene. Liquids were chosen for convenience in sample handling. The carbon and oxygen cross sections are well known (Sec. III), and their subtraction introduces little error (~0.02%) in the hydrogen value. The transmission measurements were performed at the Munich research reactor FRM using a previously described experimental arrangement[18-20] in which the energy selection is made by double-resonance scattering in cobalt. The suitability of this method for precision measurements of cross sections at 132 eV has been demonstrated in preceding experiments[18,19,21] on D, C, O, Si, Pb, and Bi. Though at that time part of our data (Si, C, D) was seriously in conflict with other measurements, some recent revisions[12,22] and new experiments[23] have meanwhile confirmed our results even in these cases.

The limiting factors in the previous determinations of the free-proton cross section[4, 17] were counting statistics and the correction for molecular binding. Our method offers some advantage regarding both. It provides good statistical accuracy in rather short sampling times (better than 0.1% in cross section within 10 h), which allows one to vary some relevant experimental parameters such as transmission coefficient, sample length, and sample compound within reasonable time. Besides, the energy of the cobalt resonance (132 eV) is just optimum to accurately determine σ_0, for the following reason.

The total cross section per proton bound in any hydrogenous molecule is still energy-dependent in the eV range due to three effects. At low energy there is the small contribution of capture, and the more significant effect of molecular vibrations. The latter varies asymptotically as $1/E$ and is of the order of 5% at 1 eV. In addition, the cross section decreases with energy according to effective-range theory, the effect being about -0.7% per keV and linear at low energy. Thus, there is just a rather small region from some tens of eV to a few hundreds of eV where the three terms simultaneously disappear (see, e.g. Fig. 5 in Ref. 24 or Fig. 2 in Ref. 4), and the free-proton cross section can be directly measured. At 132 eV neutron energy none of the required corrections exceeds 0.1% in cross section, and their calculated combined effect is only 0.03%.

II. EXPERIMENT

A. Experimental method and setup

Here we give only a brief outline of the method and experimental conditions which have been previously described in detail.[18-20] The main point is the following: If a "white" spectrum of epithermal reactor neutrons is impinging on a thin foil of cobalt, a small energy band around the prominent resonance at 132 eV will shine up strongly enhanced in the scattered spectrum. The width of that band is of the order of the resonance width $\Gamma \simeq 5.6$ eV, while the enhancement factor obtained with a *thin* foil, i.e. the ratio of the scattering cross section in resonance ($\sim 4\pi g \bar{\lambda}_0^2$) and off resonance ($\sim 4\pi R'^2$), is of the order 2000.

In our transmission experiment (Fig. 1 in Refs. 18-20) we actually use *three* subsequent resonance selections in cobalt foils (double scattering plus "self-indication") to produce a monoenergetic neutron spectrum. First, a thin scatterer is placed near the reactor core in the center position of the evacuated through tube FD of the FRM reactor. Scattered neutrons arising from that "source" are collimated in a system of Soller slits (Fig. 2 in Ref. 20), giving less than 0.8° full beam divergency. The intensity of the beam, after passing a Cd filter and the sample position, is measured in a resonance detector which involves a second scattering[25] from a cobalt foil. In addition, we apply the usual "self-indication" technique[26] to further reduce the remaining effect of off-resonance neutrons, i.e. for background subtraction we use sample-in and sample-out data taken with a cobalt resonance filter[27] inserted into the beam. Neglecting some small corrections (Sec. II C), the sample transmission and cross section are given by

$$T = \exp(-n\sigma) = (C_S - C_{SF})/(C_0 - C_F), \qquad (1)$$

where C_0 is equal to the counts with no sample in the beam $\simeq 800$ counts/sec; C_S is counts with the sample in beam; C_F is counts with the filter in beam; C_{SF} is counts with both sample and filter in beam; n is sample thickness in molecules per barn; σ is total cross section (132 eV) in barns per molecule.

In the present experiment on liquids the sample-out counting of C_0 and C_F is actually made with an empty container in the beam, replacing the sample container (Sec. II B).

The difference procedure used in Eq. (1) provides in itself correction for background due to room-scattered neutrons and γ radiation in the reactor hall ($\simeq 3\%$ of C_0), which contributes equally to the four count rates and is cancelled in both differences. Also, small variations in its magnitude have no effect since filter-in and filter-out runs are made in cycles of 5 min.

It was previously shown[18-20] that the experimental procedure outlined above provides a nearly monoenergetic neutron spectrum, with negligibly small contributions of off resonance and fast neutrons. The off-resonance background is mainly due to low energy neutrons from ~1 eV (Cd filter) to ~100 eV. Its step-by-step reduction in the three-stage resonance selection has been demonstrated by measuring the "self-transmission" (Fig. 3 in Ref. 20) through samples of cobalt. The off-resonant effect remaining with the filter-difference procedure (curve c in that figure) is less than 0.1%, probably about 0.04% (calculated). In addition, we have a small background of keV neutrons due to double scattering from resonances in cobalt at higher energies,[28] predominatly due to the levels at 4.32 and 5.02 keV. According to simple estimates[18, 20] this component should be about 0.3% for the set of cobalt foils used. In the present measurement, most of that background was removed *experimentally* using a permanent filter of 5 mm vanadium in the beam (in addition to the Cd filter), which is nearly "black" for energies from 4 to 7 keV.[29] A direct check of our spectrum in view of the keV neutron background was provided by measuring the exponential attenuation through boron over more than three orders of magnitude in transmission, similar to Fig. 4 in Ref. 20. As no significant "hardening" was observed we estimate the residual contribution of keV neutrons in our spectrum as $\lesssim 0.1\%$. Both neutronic backgrounds discussed above have negligible effect on the determination of the free-hydrogen cross section.

Some slight improvements were made in geometry, compared with the previous experiments.[18-21] The sample-detector distance was increased to 2.40 m. The filter and sample holder were mounted on a bank, together with three circular diaphragms of polyethylene, each 50 mm thick. Two of them, 23 mm in diameter, were placed in front and back of the sample position, while the third, 25 mm in diameter, was set about midway between the sample and the detector foil. Since the beam divergency in our arrangement is determined by the Soller collimators, these diaphragms merely served to define the beam cross section and the detector solid angle ($\sim 3\times 10^{-5}$ of 4π) with respect to the sample position.

The alignment of the whole setup, about 7 m in length from the "source" to the detector foil, was done with a laser beam. The sample holder was adjusted to within 0.3° with respect to the beam axis, using the laser beam reflection from the quartz windows of the sample containers (Sec. II B). Thus, errors due to oblique transmission were also negligible.

B. Samples

The liquids used were reactor grade water (resistivity $\sim 2\ M\Omega\,\mathrm{cm}$), GC-analyzed benzene from Baker,[30] and methanol and toluene of highest available purity from Merck.[31,32] The densities of the organic samples were determined relative to water by weighing in a 25 ml pycnometer which was equipped with a thermometer. Measurements were made at several temperatures near 20°C, to check also the volume expansion coefficients. The maximum error in the densities is estimated as 0.03%, including the uncertainty in temperature (0.2°C). Within this estimate the data were consistent with standard values.[33]

Three sample containers were prepared for the transmission experiments, and were alternately used both as dummy and for the sample. The length of each container was defined by a stainless-steel cylinder, (outside diameter 55 mm, inside diameter 30 mm), the front and back faces of which were machined plane to better than 10 μm. Polished disks of quartz glass (SiO_2, 2 mm thick, 40 mm in diameter,) were used as windows and were kept directly on the respective faces of the cylinders by means of a stainless-steel front ring and an intermediate rubber ring. The g/cm^2 of each disk were determined from its mass and diameter, and three pairs were selected for which the total mass per area agreed to within 0.3%. Since neutron attenuation in 4 mm of SiO_2 is about 8%, the errors in the measured transmission coefficients due to this slight difference were less than 0.03%. They were further reduced, on average, by the alternate use of each can for the sample and for the dummy, and were thus neglected.

The sample thicknesses determined by the lengths of the stainless-steel cylinders, were 12.141, 17.035, and 24.030 mm, respectively. They were measured with a micrometer calibrated with gauge blocks, the certified tolerance of which was 1 μm. The observed variations in length over the area of the cylinders were within $\pm 5\ \mu$m, which were also taken as the error of the average lengths. These were additionally checked by measuring the total lengths of the ready containers and subtracting the thickness of the quartz disks. These determinations, slightly less accurate, were in agreement with the former procedure.

The cans were filled with hypodermic needles through radial holes, 6 mm in diameter, machined in each of the cylinders. Sample temperatures during the transmission experiment were controlled with the same thermometer as used in the pycnometer, inserted into the filling holes. Variations in temperature were checked in periods of 1 to 2 h, and were found to be both small and of long term. (The extreme values observed within three weeks were 20.1 °C and 21.3 °C.) Average sample temperatures were used to evaluate the transmission data for the particular fillings, i.e. for each about 10 h, and their errors were taken as 0.2°C.

The effect of impurities on the cross-section measurement should be negligible both for water and the used sample of benzene.[30] In case of methanol[31] and toluene[32] possible errors were estimated on the basis of the certified assay and the quoted bounds of the water content. The rest was assumed to consist of hydrocarbon groups, the effect of which was estimated by varying the H/C ratio from 1 to 2. This leads to a maximum uncertainty in cross section of 0.06% and 0.15% for CH_3OH and C_7H_8, respectively.

C. Experimental results

Optimum counting statistical accuracy in cross section within a given time is achieved with sample lengths of about twice the free neutron path, i.e. transmissions of 10% to 20%. The transmission coefficients resulting with the three sample lengths and four compounds used were around the optimum

TABLE I. Experimental error in the cross sections per molecule (in percent).

	H_2O	C_6H_6	CH_3OH	C_7H_8
Statistical	0.043	0.048	0.045	0.048
Systematic				
Sample purity	...	...	0.06[a]	0.15[a]
Sample density	b	0.03	0.03	0.03
Sample temperature	0.00	0.02	0.02	0.02
Sample length[c]	~0.03	~0.03	~0.03	~0.03
Dead time	0.02	0.02	0.02	0.02
Inscattering	0.01	0.01	0.01	0.01
Systematic total	0.04	0.05	0.08	0.16
Over all	0.06	0.07	0.09	0.17

[a] Estimated *maximum* uncertainty.

[b] Used for reference.

[c] Uncertainty ±5 μm. Sample lengths were about 12, 17, and 24 mm, respectively.

region (about 7% to 33%), except for the low values obtained with 24 mm of CH_3OH and H_2O (about 4.0% and 2.8%, respectively). Yet, data were taken with each of the 12 combinations to about the same statistical accuracy in cross section (~0.08%), which provided a sensitive test of our experimental and evaluation procedure in view of rate-dependent effects and backgrounds (Sec. II A).

The neutron transmission coefficients were determined by counting in cycles with the four possible sample and filter positions. Filter-in and filter-out runs were interchanged in periods of 5 min, while, in order to obtain better use of reactor time, we actually spent more time with sample-in than with sample-out counting depending on the particular transmission coefficient. Evaluation of sample transmissions from Eq. (1) could be made without reference to a monitor, as variations in the primary intensity due to electronic drifts and variations in reactor power were typically only some tenths of a percent per day. The random error of each transmission coefficient was calculated both from counting statistics and from the standard deviation of the transmissions for the individual pairs of sample-out and sample-in data. The errors determined by these alternative procedures were in no case significantly different.

Data from each combination sample length/compound were individually corrected for counting losses due to electronic dead time[18,19] (+0.24% to 0.50% in cross section), for the additional air scattering during sample-out counting (~ +0.03%), and for inscattering (~ +0.01%). The corrected cross sections per molecule obtained with the different sample lengths showed in no case statistically significant trends. The weighted means are given in the first line of Table II. A listing of the known experimental uncertainties is given in Table I.

III. EVALUATION OF THE FREE-PROTON CROSS SECTION

To evaluate the total cross section per hydrogen atom at 132 eV (Table II) we subtract 4.746 ± 0.005 and 3.761 ± 0.007 b, respectively, per atom of carbon and oxygen. The value of the carbon free-atom cross section rests on three independent experiments. Our measurement[18,19] in 1970 on reactor graphite (132 eV) was 4.746 ± 0.007 b. This was 1.7 standard errors below Houk's and Wilson's formerly reported[34] value for pyrolytic graphite, but agrees exactly with Houk's revised[4] result (4.7461 ± 0.0045 b) including his Doppler correction. Also, the revised[12] mirror-reflection value is now in excellent agreement with the transmission data.

TABLE II. Experimental results and evaluation of the free-proton cross section σ_0.

	H_2O	C_6H_6	CH_3OH	C_7H_8
Total cross section (132 eV)				
per molecule (b)	44.731	151.40	90.37	197.20
per bound H atom[a] (b)	20.485	20.487	20.466	20.497
Evaluated σ_0[b] (b)	20.491	20.495	20.474	20.505
Statistical error	0.010	0.012	0.010	0.012
Systematic error[c]	0.010	0.014	0.019	0.040
Mean of σ_0 (b)		20.491		
Standard error		0.007		
Systematic error		~0.012		

[a] Subtracting 4.746 ± 0.005 b and 3.761 ± 0.007 b per atom of carbon and oxygen, respectively.

[b] Corrected for the contributions of capture, effective range, molecular binding, and deuterium, as discussed in the text.

[c] Including the systematic experimental errors (Table I), the error in the oxygen and carbon cross sections, and the uncertainty due to the molecular-binding correction.

The oxygen cross section quoted above was derived[21] from measurements at 132 eV on quartz glass[21] (SiO_2) and silicone.[18,19] Our result for Si has meanwhile been confirmed by Shull and Oberteuffer.[22] Another measurement on SiO_2 at 5.2 eV was recently performed by Waschkowski,[35] who finds a cross section (uncorrected for the Doppler effect) very slightly above our value.[21] An independent determination of the oxygen scattering length using mirror reflection was reported by Nistler,[36] whose result leads to a free-nucleus scattering cross section of 3.758 ± 0.007 b. Thus, both cross sections used in the present analysis seem now to be well established by different experiments.

To evaluate the free-proton scattering cross section σ_0 at "zero energy," we apply further four corrections (see the detailed discussion by Houk[4]):
(1) We add 2.5 mb for the effect of deuterium (0.015 at% abundance, free-atom cross section 3.39 b [21]).
(2) We subtract 4.6 mb for neutron capture at 132 eV by extrapolating the thermal absorption (332 mb [28]) according to the $1/v$ law.
(3) We add 18 mb for the decrease in the *np* cross section between zero energy and 132 eV, calculated from the effective-range formula ($r_s \simeq 2.76$ fm^2).
(4) We subtract estimates of the residual molecular binding effects at 132 eV using the first-order form of the Placzek-Wick expansion.[37] Asymptotically, one has $\sigma/\sigma_0 = 1 + (\langle K\rangle/3E) + \ldots$, where $\langle K\rangle = \frac{3}{2}\langle T\rangle$ is the mean kinetic energy per bound proton and $\langle T\rangle$ is a corresponding *effective* temperature in eV. For H_2O there are two experimental estimates[38,39] of $\langle T\rangle$ and three theoretical predictions from different models listed in Ref. 39. We adopt $\langle T\rangle = 0.125$ eV ± 10%, which covers all these data and yields a correction of -10 ± 1 mb in cross section at 132 eV. For C_6H_6 we correct -8 ± 1 mb using a theoretical value $\langle T\rangle = 0.1005$ eV from a model due to Sprevak *et al.*,[40] which provides a good fit to the experimental cross section[40] from thermal to eV energies. We are not aware of any specific data concerning the binding effect in CH_3OH and C_7H_8, but we may use the fact that many hydrocarbons[41] and hydrogenous organic liquids[42] show essentially the same asymptotic trends. The correction was, therefore, assumed as -8 ± 3 mb for both.

The data of σ_0 obtained with the four compounds, given in Table II, are reasonably consistent within statistics. The unweighted mean is 20.491 b, the standard error of the mean being 7 mb. Weighing the four data by the inverse or squared inverse of their systematic uncertainties would change the mean by less than 2 mb. For the total systematic error of the mean we take ~12 mb, as estimated for H_2O and C_6H_6. Our final result of the free-proton scattering cross section at "zero energy" is thus

$$\sigma_0 = 20.491 \pm 0.014 \text{ b},$$

the error being the quadratic sum of the systematic and standard error of the mean.

IV. DISCUSSION

A comparison with the best previous measurements of σ_0 by Houk,[4] Neill, Russell, and Brown,[39] and Melkonian[38] is given in Table III. Some other data compiled by Story[8] and still earlier determinations[43] were mostly even lower. Our measurement confirms the trend to a higher value of σ_0 as indicated by Houk's experiment, but it is still larger and outside the error bars of the former results.

For H_2O, a direct comparison can also be made with the data given in Fig. 2 of Neill *et al.*[39] and Melkonian's[38] Fig. 6. Our measurement, when plotted into these figures[44] at $1/E \simeq 0$, does not appear to be really inconsistent with these data regarding their statistical scattering and uncertainties due to the molecular binding effect. It is also worth noting that the molecular parameters $\langle T\rangle$ derived in Refs. 38 and 39 are systematically high compared with theoretical predictions (Table I in Ref. 38 and Table II in Ref. 39) which might be another reason for the low fitted values[38,39] of σ_0.

The discrepancy with Houk's[4] σ_0 might originate to some extent from his data selection. It is evident from Houk's Fig. 2 that most of his data from 1 eV to ~50 eV lie above his fit curve labeled "0°C," while the few points from ~50 eV to 330 eV are systematically low. Houk's final analysis included the data from 6 eV to 330 eV which give his

TABLE III. Comparison with previous measurements of σ_0.

Reference	Sample	Energy (eV)	σ_0 (b)
Melkonian (Ref. 38) (1949)	H_2, C_4H_{10}, H_2O	0.8–15	20.36 ± 0.10
Neill *et al.* (Ref. 39) (1968)	H_2O	0.8–16	20.366 ± 0.076
Houk (Ref. 4) (1971)	H_2	6–329	20.436 ± 0.023
Present	H_2O, C_6H_6, CH_3OH, C_7H_8	132	20.491 ± 0.014

TABLE IV. Low energy *np* parameters. The parameters of Lomon and Wilson[2] are based on Houk's σ_0, $a_H=-3.739(3)$ fm of Koester and Nistler,[12] and $\epsilon=2224.644(46)$ keV. The second column gives the data evaluated from the present σ_0 using the same a_H and ϵ.

Parameters (fm)	Lomon and Wilson 1973 values	Evaluation using present σ_0
a_t	5.414(5)	5.423(4)
a_s	−23.719(13)	−23.749(9)
$\rho_t(-\epsilon, 0)$ [a]	1.750(5)	1.760(5)
r_{0s}		2.77(5) shape independent
	2.76(5) BC model	2.81(5) BC model shape correction[b]
		2.78(5) OPE shape correction[c]

[a] Mixed triplet effective range, model independent. In the shape-independent approximation, $r_{0t}=\rho_t(0,0)\equiv\rho_t(-\epsilon,0)$. For the evaluation of r_{0t} using BC model or OPE shape corrections see Ref. 4 and Refs. 3 and 48, respectively.

[b] Using shape correction for the boundary-condition model (Ref. 49) from Houk (Ref. 4).

[c] Using OPE shape correction from Noyes (Refs. 3 and 48).

first few timing channels a rather heavy weight. He mentions[4] that the fitted σ_0 tends to slightly higher values as the lower energy limit is reduced, but makes no quantitative statement on this point. In fact, an increase in σ_0 by 0.27% as suggested by the present measurement seems to be well compatible with Houk's data up to ~50 eV, but not with the few points beyond. Perhaps these might be systematically low by uncertainties in the first few timing channels due to fast neutron background or resolution. (The latter could affect the effective-range correction.)

Our estimate of σ_0 implies changes in the low energy *np* parameters towards higher values as indicated in Table IV. The first column summarizes the 1973 best values from Table II of Lomon and Wilson,[2] which are based on Houk's σ_0, the measurement[12] of Koester and Nistler of $a_H=-3.739\pm0.003$ fm, and $\epsilon_D=2224.644$ keV. The second column gives the parameters evaluated from the present σ_0, adopting the same values of a_H and ϵ_D. In fitting r_{0s} we use the five most accurate single-energy data below 5 MeV, at 0.4926,[45] 0.5260,[15] 2.535,[14] 3.186[45] (revised energy[46]), and 4.749 MeV.[47] Small corrections for $l>0$ partial waves are taken from Ref. 48. Values of r_{0s} are given for the shape-independent approximation, and for fits including boundary-condition model[49,4] and one-pion-exchange[48,3] (OPE) shape corrections. The error in r_{0s} due to the five data at MeV energies is ~0.04 fm (standard error of the mean), while the uncertainty due to the zero energy data, mainly from σ_0, is 0.02 fm. The changes in the *np* effective-range parameters suggested by the present measurement are about 2 standard errors in a_t, a_s, and r_{0t}, and about 1 standard error in r_{0s}. The fitted r_{0s} remains within the span of theoretical predictions by Noyes[50] (2.73 fm) and Breit *et al.*[51] (2.83 fm), derived from *pp* scattering under the assumption of charge independence.

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